Transient behavior of EUV emitting discharge plasmas

a study by optical methods
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Chapter 1

General introduction

Abstract

Extreme ultraviolet lithography (EUVL) is generally considered to be the most promising candidate to become the successor of deep UV lithography techniques for further reduction of feature sizes in computer chips. EUVL requires the application of a high-intensity light source at a wavelength of 13.5 nm. Discharge plasmas can be applied for extreme ultraviolet (EUV) light generation in a cost-effective way. This thesis deals with the characterization of such EUV light producing discharge plasmas. The main focus is on experimental investigations of the plasma with the use of optical diagnostics. At the end of this chapter, an outline of the remainder of the thesis is given.
1.1 Introduction

Pulsed discharge plasmas are nowadays considered by many people in the field as the most serious candidate sources of extreme ultraviolet (EUV) light in future generations of lithography tools for the computer industry.

As plasmas, they are characterized by the fact that charged particles (electrons and ions) play a prominent role in determining their properties. The plasma state is the predominant state of matter, if all visible matter in the universe is considered. Plasmas are mainly found in the form of stars and interstellar nebulae. Although less obvious to the general observer, plasmas also play an important role on earth. Besides purely academic ones, there are “technological” plasmas that have a very wide range of applications in industry, reflecting the enormous ranges over which plasma parameters can be varied. Reactive plasmas are used for deposition and etching of materials, for example in the production of solar cells or computer chips. Large, particularly hot plasmas are proposed for future controlled nuclear fusion power plants. Further, plasma torches are used for large-scale cutting and welding. Another important field of applications is as light sources, for example in gas discharge lamps, plasma display panels, and certain types of lasers. Using plasmas as sources of EUV radiation is a relatively new type of application as a light source. The production of EUV radiation for semiconductor lithography puts a set of specific, very demanding requirements on the properties of (discharge) plasmas. Among others, the required high temperatures make pulsed operation necessary.

As the world’s leading developer and producer of lithography systems, the Veldhoven-based company ASML has a large interest in the development of EUV light sources. For this reason, not only an EUV research laboratory was started in 2001, but also in September of the same year, a joint research project was started by ASML and the Eindhoven University of Technology (TU/e) covering both experimental and theoretical investigations of pulsed discharge plasmas. For the group Elementary Processes in Gas discharges (EPG) in the Department of Applied Physics at TU/e, the project meant the addition of a new subfield of research to the existing range of plasma types under study.

This thesis describes a part of the work done within this research project, and focuses mainly on the experimental aspects. This chapter serves as a general introduction to the subjects covered in more detail in the subsequent chapters. First, the following section gives a brief introduction into semiconductor lithography in general, and in particular the motivation for the development of EUV lithography. In Sec. 1.3, general properties of EUV producing discharge plasmas are described. Finally, in Secs. 1.4 and 1.5, the scope of this work is described in more detail, and an outline is given of the remaining parts of the thesis.
1.2 Next-generation lithography

The lithography step in the production of ICs

A fundamental production step in the fabrication of silicon-based integrated circuits is that of optical lithography. In a lithography tool, a pattern, as present on the reticle (or mask), is projected by an optical system onto a silicon wafer that was previously coated with a photosensitive material (the photoresist). After the illumination, in a different apparatus either the unexposed or exposed parts of the photoresist—depending on whether a negative or a positive resist was applied—are etched away (chemically, or by use of a plasma), and the pattern is etched into the top layer of the wafer. The physical patterns on the wafer that are thus created are further processed to form electronic pathways. This procedure is depicted in a simplified way by Fig. 1.1. It is repeated several times to form the interconnected multilevel semiconductor electronic structure called the integrated circuit (IC), more commonly known as the microchip.

Resolution and depth of focus

In the computer industry, there is a continuing drive to reduction of the sizes of the basic features in computer microchips. Smaller features in chips will enable chip producers to increase memory sizes and the IC clock speed, thereby increasing the computational power of the chip, while at the same time keeping the heat production and energy consumption in general within acceptable levels. According to Moore’s law [2], first formulated in the 1960s, feature sizes in integrated circuits are reduced by a factor of $\sqrt{2}$ roughly every eighteen months to two years. This law has proven valid for forty years already.

The sizes of structures that can be created in computer chips are for a large part determined by the properties of the optical lithography system. Hence, in the past two decades it have been mainly the enormous advances in optical lithography that have enabled the
Chapter 1: General introduction

Fig. 1.2. A demonstration of focusing principles. Parts (a) and (b): when the half-acceptance angle \( \theta \), and thereby also the numerical aperture \( NA \), is increased (from \( \theta_1 \) to \( \theta_2 \) in this case), the resolution \( L_w \) shrinks; however, the depth of focus \( DOF \) shrinks faster. Part (c): a shorter wavelength of light can be applied to further reduce the resolution.

shrinking of feature sizes. The minimum size of a feature etched into the resist layer is determined by the resolution and the depth of focus. See Fig. 1.2 for a schematic representation of these quantities. The resolution is here defined as the minimum width \( L_w \) on the wafer of a line that was projected onto the wafer from the reticle. Its value is limited by diffraction; the resolution is related to the numerical aperture \( NA \) and light wavelength \( \lambda \) as

\[
L_w = k_1 \frac{\lambda}{NA}, \tag{1.1}
\]

where the numerical aperture is defined as \( NA = \sin \theta \), with \( \theta \) the half-acceptance angle of the lens, i.e., the largest angle between a light ray and the optical axis, measured in the space between the last optical element and the wafer. The proportionality constant \( k_1 \) depends on specifics of the optical system and the photoresist.

The second parameter, the depth of focus (DOF), is an indication for the vertical distance away from the focal plane of the optical system for which the image of the reticle is still “good”, i.e., the image size of the spot is within a certain percentage of the resolution. The depth of focus is given by

\[
DOF = k_2 \frac{\lambda}{NA^2}. \tag{1.2}
\]

Here, \( k_2 \) is another proportionality constant. Its value is dependent on the precise definition of the DOF, but typically it is on the order of 0.5.

Now, the resolution of a given system can be improved, in principle, in three different ways: by reduction of \( k_1 \), by increase of the numerical aperture, and finally by reduction of the wavelength. All three methods have been pursued in the past. For instance, over the past decade or so, lithography tools have been developed using as light sources mercury arc lamps, emitting the so-called I-line at 365 nm, and krypton fluoride and argon fluoride excimer lasers operating in the deep ultraviolet (DUV) range, at 248 nm and 193 nm, respectively. The 193 nm systems are the current state-of-the-art in commercially available systems.
1.2 Next-generation lithography

In these systems, the value for $k_1$ is about 0.3; this is already close to the fundamental limit (for dense line patterns) that is found at $k_1 = 0.25$. Hence, only a marginal further resolution improvement could come from further reduction of $k_1$. Alternatively, the resolution could be improved further by increasing the numerical aperture of the optical system with respect to the wafer, but this goes at the cost of loss of depth of focus, which makes vertical positioning of the wafer more critical. Also, a lower limit for the depth of focus is given by the resist layer thickness. These are practical limits for increasing the numerical aperture. A more fundamental limit is imposed by the fact that $\sin \theta$ cannot be larger than unity. In practice, $NA \approx 0.9$ is the largest achievable value, and current systems are already close to this limit.

Therefore, currently another way of improving the resolution is being pursued: this is the so-called **immersion lithography**. This concept was first introduced in commercial systems recently. In immersion lithography, a liquid is introduced between the bottom surface of the last lens and the top surface of the wafer. Now, Eqs. (1.1) and (1.2) are modified in the following ways,

$$L_{w,imm} = k_1 \frac{\lambda}{n_{liq} \sin \theta_{liq}},$$

(1.3)

$$DOF_{imm} = k_2 \frac{\lambda}{n_{liq} \sin^2 \theta_{liq}},$$

(1.4)

where $n_{liq}$ is the refractive index of the liquid at the wavelength of interest, and $\theta_{liq}$ is the half-acceptance angle as measured inside the liquid. If an appropriate liquid (with $n_{liq} > 1$) is selected, the resolution can be improved compared to “dry” lithography. The depth of focus is also reduced, but the loss is only linearly proportional to the refractive index, which is better than the quadratic dependency on $NA$ as expressed by Eq. (1.2).

Another common way of looking at the application of a liquid is to redefine the numerical aperture as $NA = n_{liq} \sin \theta_{liq}$—note, however, that this definition cannot simply be substituted into Eq. (1.2) to reproduce Eq. (1.4). With this modified definition, numerical apertures larger than 1 can be obtained, which has led to the introduction of the term **hyper NA lithography**.

Nevertheless, also with immersion lithography there is a limit to the achievable $NA$. The application of 193 nm systems is likely to be extended to the so-called 45 nm **node**. This is the technology in which the smallest combined line and space width within a dense group of lines is 90 nm. On the other hand, it is expected that only a further decrease of illumination wavelength can help to achieve the next node at 32 nm.

Application of a fluorine laser at 157 nm was long regarded as the next step on the roadmap. However, it is as yet unclear if immersion lithography will be feasible at this wavelength. If not, the change to 157 nm will not bring any benefits over immersion lithography at 193 nm, and hence will probably be skipped by important players in the semiconductor industry. In this case, a larger jump in wavelength will have to be made.
Chapter 1: General introduction

The next step
When the source wavelength is reduced further, a problem that is encountered is that the conventional optical systems can no longer be used. These systems are based on focusing of light by lenses. Important properties of lenses are sufficiently high optical transparency and refractive index. However, no materials can be found that have the required properties at wavelengths below about 150 nm.

In the past, several alternative techniques have been proposed or developed to achieve better imaging resolution. These include proximity X-ray and electron beam lithography. In the former case, no imaging system is used at all between the reticle and the wafer. The reticle, which is partly covered by a non-transparent layer, is positioned close to the wafer, and the wafer is illuminated directly through the pattern that is formed by the openings in the absorbing reticle layer. In general, radiation with a wavelength between 1 and 2 nm is applied. One drawback of this method is that the magnification factor between reticle and wafer is, by definition, equal to one, and therefore this technique requires printing very small features on the reticle. Also, the reticle needs to be at least partly transparent to the X-rays, so that it has to be very thin and it will hence be very fragile. Because of the extreme fragility, this technique is not usable for mass production of computer microchips (which is commonly referred to as “high volume manufacturing” or HVM).

Electron beam (e-beam) writing suffers from the same problem of limited throughput. It is used for the production of masks for optical lithography, but the technique is far too slow (and therefore expensive) for HVM. Since a few years, the MAPPER Lithography company [3] has been working on a scheme that applies a large number of parallel electron beams, which could partially overcome the throughput limitation. Although this may be a viable method, at this point the technology does not seem to be mature enough to be introduced commercially any time in the near future.

The EUV wavelength range
Yet another option is to use mirrors instead of lenses as optics between the reticle and the wafer. For achieving good imaging resolution and small aberrations with a limited number of optical elements, the use of near-normal incidence mirrors is indispensable. Unfortunately, near-normal incidence mirrors made from a single material have an extremely low reflectivity in the deep ultraviolet range. This problem can be solved by combining materials of high and low refractive indices into multilayer (ML) stacks: when each bilayer is given a thickness of roughly half the wavelength, constructive interference of the light reflected off each interface in the material can lead to acceptable reflectivity values. That is, the mirrors are designed to work as Bragg reflectors. The best results have been achieved for mirrors consisting of alternating layers of silicon (Si) and molybdenum (Mo); their highest reflectivity is around 70%, for a mirror designed for a wavelength near 13.5 nm.\(^1\)

\(^1\) Alternatively, molybdenum/beryllium (Mo/Be) mirrors have a good reflectivity curve around 11 nm; however, due to related manufacturing and safety problems, Si/Mo mirrors are preferred. Furthermore,
1.2 Next-generation lithography

![Diagram of EUV lithography tool with labels: Source, Collector mirror, Spectral purity filter, Intermediate focus, Condenser optics, Projection optics, Reticle, Wafer position.]

Fig. 1.3. Design example for the mirror optics in an EUV lithography tool. After a “collector mirror” and possibly a spectral purity filter of some kind, the light gets focused to a so-called intermediate focus. The condenser optics illuminate the reticle (mask); the projection optics produce an image of the mask on the wafer.

Therefore, the development of a lithographic technique using mirror optics has focused on the application of just this wavelength.

Originally, this technique was called “soft X-ray lithography”, but this name was dropped by the industry in favor of “extreme ultraviolet lithography” (EUVL) in the nineties of the last century to avoid confusion with the near-contact printing technique mentioned above. As a result, the term “extreme ultraviolet” (EUV) has also become a general name for light in the wavelength range of roughly 10–20 nm, although the term is still mainly used in conjunction with application in lithography. Note that the limits of both terms “EUV” and “soft X-ray” are not very well defined.

As indicated above, the reflectivity of a ML mirror is still far from unity, even at the optimum wavelength. Also, its full width at half maximum (FWHM) as a function of wavelength is only about 4%. Since in an optical system, ten or more optical elements have to be combined to produce a good image on the wafer, the FWHM of the transmission spectrum of the entire system is reduced to about 2% of 13.5 nm; and the peak transmission of a system with ten ML mirrors is not more than about 2.8%.

It is for this reason that very demanding requirements are put on the properties of light sources for EUV lithography. The general design of EUVL allows for the definition of an intermediate focus (IF); see Fig. 1.3 for a design example. The required light power emitted by the source is usually defined as the power required in a 2% wavelength band.

the exact choice of wavelength has been influenced by the shape of the spectrum of lithium; see for details Ref. 4.
around 13.5 nm at this IF point. The current status—as of February 2005—is a required power of 115 W. A more detailed discussion of the industry demands on light sources is given in Sec. 2.1.1.

### 1.3 Discharge plasmas as EUV light sources

A wavelength of 13.5 nm corresponds to a photon energy of about 92 eV. Although there are several alternative ways to create such photons (some of which will be discussed in the next chapter), the easiest and most cost-effective way of making a high intensity source is to use atomic line radiation. Transitions between excited levels of an atom or ion can, in principle, only create photons with energies below the ionization potential of that specific atom or ion. Line radiation with photon energies as high as 92 eV can therefore only be generated by multiply ionized atoms. Examples of ions that exhibit emission spectra with considerable peaks at or near 13.5 nm, are O$^{4+}$, Xe$^{10+}$, Li$^{2+}$ and Sn$^{8–11+}$.

Such ions can only be generated and electronically excited in sufficient quantities in a hot plasma, with electron temperatures in the range of 20–50 eV. High electron densities (on the order $10^{25}$ m$^{-3}$) are required for collisional excitation of the ions to the proper radiating levels.

There are basically two ways in which a sufficiently hot and dense plasma can be generated. The first is to feed the required energy to a target by firing a laser pulse onto it; the plasmas generated in this way are aptly named *laser produced plasmas* (LPPs). The other way is to expose the target material to a strong electric current; in this case energy is fed to the plasma through Ohmic heating. Plasmas generated in this way are called *(gas)* *discharge produced plasmas* (GDPPs) or simply “discharge plasmas”.

In both cases, in view of the required energy input and the resulting heat load, it is possible to sustain a sufficiently hot plasma only for a very short time. Additionally, for both LPPs and GDPPs it turns out to be impossible to fully confine the plasma to the location where it has been generated. Instead, the thermal energy of the particles is quickly converted into a directed expansion velocity. Not only is the thermal energy lost for excitation of ions; also the ions (and electrons) themselves are lost. Hence, all known efficient EUV producing plasmas have a pulsed character. Due to the short lifetimes of EUV generating plasmas, their properties change very rapidly during their evolution. Typically, the effective time for EUV generation varies from a few ns for LPPs to—at most—a few tens of ns for GDPPs.

*Discharge* plasmas for EUV generation are, contrary to LPPs, often started in a medium that is *less* dense than the final plasma. Before the onset of expansion, an increase in density is achieved by the so-called “pinch” effect, a magnetic compression caused by the electric current. Typically, up to a factor ten reduction of plasma radius can be achieved in this way. Usually, the shape of the plasma is not stable under this process; instead,
1.3 Discharge plasmas as EUV light sources

Fig. 1.4. Part (a): the EUV spectrum of a capillary discharge in xenon, reproduced from Ref. [8]. The oxygen lines shown in the spectrum originate from contaminations in the xenon gas. Part (b): a typical tin discharge spectrum shown for comparison; see for further details Chapter 5. The 2% wavelength band around 13.5 nm is highlighted.

various kinds of pinching instabilities can occur [5–7]. These, too, can limit the lifetime of the pinch plasma.

All types of discharge plasmas have certain characteristics in common. The plasma is usually generated in a small space between two electrodes. These are connected to one or more rings of capacitors through a low-inductance circuit, to ensure that a large amount of electrical energy can be fed to the plasma in a very short time. The capacitors, in turn, are connected to an external power supply, which can recharge them on a longer timescale after they have been discharged by the current pulse. Each discharge pulse is started by closing a fast switch in the capacitors-plasma circuit, or by application of some kind of external trigger. In the latter case, the gap between the electrodes itself is changed from electrically insulating to conductive, and hence serves as a switch. More details on the schematics and operation of discharge plasmas, as well as on the differences between the various types, are given in the following chapter.

Of the chemical elements mentioned above, xenon has the advantage over all the other ones that it is a noble gas. Hence, it is not only in a gaseous state at ambient conditions, but it is also chemically inert. Therefore, it has been commonly applied as the working element in the development of various types of discharge plasmas. Because of the similarities in plasma temperatures and lifetimes of these plasmas (compared to the typical time constants needed for population of the various ionization stages of xenon), the pulse-integrated EUV spectra of these xenon plasmas also tend to be rather similar. An example of such an EUV spectrum, as reported in the literature [8], is given in part (a) of Fig. 1.4.

More recently, tin has attracted increasing attention as an alternative working element, despite the drawback that it is solid at ambient conditions so that more effort needs to be put into introducing it into the discharge, and into keeping it from polluting the mirrors. The reason is that the shape of the EUV spectrum of tin is, in general, much more favorable.
than that of xenon for high intensity emission around 13.5 nm; an example is shown in part (b) of Fig. 1.4.

### 1.4 Scope of the thesis

The demanding requirements from the industry on the properties of EUV light sources, make it a far from trivial task to design and produce such a source. The further development and improvement of EUV emitting plasmas require a good understanding of the fundamental processes that dominate the plasma evolution. Such understanding can only follow from measurements of the plasma parameters, in combination with efforts in the fields of plasma theory and computer modeling. Therefore, the goal of this work has been to further the understanding and characterization of EUV producing discharge plasmas, and, more generally, to develop experimental techniques that can be applied with the same goal in the future.

The scope of the research project that the present thesis is a part of, is limited to the study of discharge plasmas. However, many of the elementary processes in plasmas are common to both laser produced and gas discharge produced plasmas. Hence, previous efforts to describe LPPs can be applied to discharge plasmas, and similarly, certain results and conclusions of this work will also be applicable to the case of LPPs.

As stated above, the main focus of this work has been on experimental methods and the results of their application to discharge plasmas. However, plasma theory has been applied and expanded where necessary to justify the applicability of certain experimental techniques, or to use it as an aid in the interpretation of specific experimental results. Computer modeling has been applied for the simulation of EUV spectra, used for a comparison with their experimental counterparts. No serious experimental work is possible without interaction with theory and modeling, and vice versa.

The focus on optical diagnostics is not one of arbitrary choice, but follows from the properties of the plasmas under study. Plasma sources of EUV radiation are generally characterized by their small spatial dimensions, short durations and high densities and temperatures. These properties make the application of electrical probes extremely difficult if not impossible. A Langmuir probe would probably not survive the first plasma pulse, but even if it did, the discharge would be strongly disturbed by its presence. Furthermore, a study of the electric parameters of the discharge (current, voltage etcetera) can give only a limited, qualitative picture of the processes that are going on inside the plasma. Therefore, a different group of experimental methods, optical diagnostics, are indispensable for obtaining detailed knowledge on the evolution of any EUV producing plasma. Additionally, optical diagnostics are simply a logical choice considering that light generation is the purpose of these plasmas.

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2 Nevertheless, this method has been routinely applied—in combination with other diagnostics—to both the hollow cathode and laser-triggered tin discharge sources; see, e.g., Sec. 5.3.
A basic subdivision of optical diagnostics can be made into *passive* and *active* ones. Active diagnostics are those in which the plasma is irradiated by an external source of some kind; an example of this type is Thomson scattering. Passive optical diagnostics are those in which such an external source is not present, such as with plasma imaging, emission spectroscopy, and line broadening measurements. Each specific diagnostic gives a different type of information about the plasma, and has its own limitations and ranges of applicability. For this reason, the most successful approach to studying any type of plasma is a suitable combination of different types of diagnostics; this is exactly what has been done in the present work.

Finally, a good understanding of these pulsed plasmas cannot be obtained without closely studying the *transient effects* that occur during their evolution. This means that time-integrated measurements over complete discharge pulses typically provide insufficient information. For this reason, in all experiments described in this thesis, measurements have been done *as a function of time* during the discharge pulse. Hence, in all experiments, a time resolution in the ns range had to be achieved.

### 1.5 Outline

The following two chapters are devoted to a more detailed introduction into two different aspects related to this work, namely the various types of EUV sources, and optical diagnostics. First, Chapter 2 deals with EUV sources. The demands on such sources for application in lithography are discussed in more detail. Also, an overview is given of a wide range of sources, but, given the scope of this thesis, the main focus is on discharge produced plasmas. Two specific types of discharge plasmas have been the subjects of experimental investigation in this work, and these two types are described in greater detail.

Chapter 3 describes a number of optical diagnostics that have been, or could in principle be applied to EUV producing discharge plasmas. Their general characteristics, as well as the extents of their applicability to this type of plasma, are discussed. However, specific experimental details or issues related to the interpretation of experimental results, are left to the individual chapters in which those experiments are described.

The remainder of this thesis is devoted to the results of different diagnostic techniques, as applied to two specific types of discharge plasmas: a hollow-cathode discharge in xenon, and a laser-triggered discharge in tin vapor. In Chapters 4 and 5 time-resolved imaging and emission spectrometry in the EUV range are described for the hollow-cathode and tin vapor plasmas, respectively. Time-resolved plasma imaging gives important qualitative insight in the dynamics of the plasma. For the case of the tin plasma, EUV imaging has been complemented with imaging in the visible wavelength range, to extend the range of applicability to cooler phases of the plasma evolution. In Chapter 6 the interpretation of the time-resolved EUV spectra is dealt with in more detail. An existing computer model, designed for LPPs, has been adapted and applied to the case of an ionizing...
discharge plasma. The modeling efforts encompass first of all calculations of the atomic
data of the relevant ions, using the COWAN computer package [10]. Further, ionic excited
state distribution functions have been determined using an analytical model. Finally, from
these, the emitted spectra have been derived. The adjustments that had to be made to
the model and its input parameters, needed to obtain a good match between simulated
and experimental spectra, provide insight in which processes play important roles in the
production of EUV radiation.

Chapter 7 deals with the study of Stark broadening of certain atomic lines in the tin
plasma in order to obtain electron densities. Chapters 8–10 are devoted to Thomson
scattering, the study of spectra of laser light scattered off free electrons in the plasma.
A first set of experiments, carried out using an existing setup for Thomson scattering,
is described in Chapter 8 and the results are presented. It was found that for certain
phases of the plasma evolution, the background radiation as emitted by the plasma was
too strong to clearly distinguish the Thomson scattered signal. Therefore, a new system
for sub-nanosecond Thomson scattering has been designed and built. Chapter 9 gives a
description of that new setup and discusses test experiments that have been performed to
characterize it. After construction and testing, the new setup has been applied to the tin
vapor discharge. These experiments and their results are presented in Chapter 10.

Chapters 4–10 have each been, or are intended to be published as separate articles in
scientific journals. Therefore they can in principle be read separately from one another.
However, it is reminded to the reader that more background information on plasma sources
and optical diagnostics can be found in Chapters 2 and 3. Further, cross references between
the various chapters have been included or updated where appropriate.

Finally, in Chapter 11 the findings of the various experiments as presented in the
preceding chapters are discussed in relation with one another, general conclusions based
on this work are presented, and certain recommendations for future work are given.

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Chapter 2

Plasma sources of EUV radiation for application in lithography

Abstract

Joint requirements for EUV lithography light sources have been established by the major lithography companies. EUV radiation can be generated in the laboratory in several ways; however, only plasma sources of EUV radiation can be considered to be serious candidates to meet the requirements at a reasonable cost. Plasma sources can be subdivided into two groups, laser produced plasmas and discharge produced plasmas. Two types of the latter group that are particularly relevant to the work described in this thesis are a hollow cathode triggered discharge operated in xenon, and a laser-triggered discharge in tin vapor.
2.1 Introduction

2.1.1 Demands from industry on future EUV lithography light sources

To guide the development efforts of various groups in the world, the major players in the field of semiconductor lithography tools have defined, and regularly update, joint requirements for the properties of extreme ultraviolet lithography (EUVL) light sources for high volume manufacturing (HVM) application [1]. These requirements are summarized in Table 2.1. Their meanings, and some of their consequences for the designs of EUV sources, are discussed below. Here we will pay the most attention to the consequences for (discharge) plasma sources, since these are the subject of research in this thesis. See, for a detailed discussion, also Ref. [2].

The first two requirements listed in Table 2.1 on wavelength and in-band power, have already been touched upon in the previous chapter. “In-band” is defined as within the 2% wavelength range around the central wavelength of 13.5 nm, as this is the range that corresponds to the full width at half maximum (FWHM) of a typical lithographic tool’s transmission curve. This number is valid for the intermediate focus (IF), as explained in Chapter 1. The IF itself is indicated in Fig. 1.3 of that chapter. This position was chosen since it makes the required power independent of the precise source and collector mirror designs. Of course, these designs, and the presence or absence of a spectral purity filter (see below), will have a large influence on whether or not the requirement of sufficient output power can be met.

Specifically, for the case of discharge plasmas, the collector optic will consist of a number of concentrically placed grazing-incidence mirrors. The larger their acceptance angle, the larger the part of the emitted EUV radiation that can be collected.

The collection efficiency is also related to characteristics of the source. Firstly, radiation may be blocked by the electrodes or other source parts. Secondly, the total collectable power is the result of the power flux density integrated over the so-called etendue. The source etendue is the integral of the collectable solid angle over the (effective) surface of the source. For the required EUV power, only the radiation emitted within the specified etendue may be taken into account. In the case of a plasma, the solid angle of emitted radiation cannot be reduced; it is always $4\pi$ sr. Therefore, to keep overall collection efficiency at an acceptable level, the best approach is to make the collection solid angle as large as possible, and at the same time try to reduce the plasma size to match the etendue. With a reasonably large collection solid angle of $\pi$ sr, the apparent plasma size needs to be limited to about 1 mm$^2$ or less for efficient collection of the radiation.

A related issue is the solid angle input from the IF to the illuminator optics. This will put certain constraints on the design of the collector optics, and hence on its efficiency and on the collectable solid angle from the source.

Even with a $\pi$ sr collector and matching plasma size, typically some 5–10 times more
Table 2.1. The joint requirements as specified by the three largest lithography companies—Canon, Nikon and ASML. In some cases, final agreement on upper or lower limits has not been reached, as they may, for example, depend on the details of the optical design of the lithography system. In those cases, the limit is indicated as a range rather than one number. Where relevant, the numbers are defined for the so-called “intermediate focus” of the optical system, as indicated in Fig. 1.3 in Chapter 1. Data are taken from Ref. [1] and were still valid in February 2005.

<table>
<thead>
<tr>
<th>Source characteristic</th>
<th>Requirement</th>
</tr>
</thead>
<tbody>
<tr>
<td>Wavelength</td>
<td>13.5 nm</td>
</tr>
<tr>
<td>In-band EUV power</td>
<td>115 W</td>
</tr>
<tr>
<td>Etendue of source output</td>
<td>$\leq 3.3 \text{ mm}^2\text{sr}$</td>
</tr>
<tr>
<td>Solid angle input to illuminator</td>
<td>$\leq 0.03–0.2 \text{ sr}$</td>
</tr>
<tr>
<td>Spectral purity, 130–400 nm</td>
<td>$\leq 3–7%$ of in-band</td>
</tr>
<tr>
<td>Id., above 400 nm</td>
<td>To be determined</td>
</tr>
<tr>
<td>Repetition frequency</td>
<td>$&gt; 7–10 \text{ kHz}$</td>
</tr>
<tr>
<td>Integrated energy stability</td>
<td>$\pm 0.3%$, $3\sigma$ over 50 pulses</td>
</tr>
<tr>
<td>Source cleanliness at IF</td>
<td>$\geq 30000 \text{ hours}$</td>
</tr>
</tbody>
</table>

...in-band power will need to be emitted by the source into $2\pi$ solid angle than can actually be collected in the IF. With conversion efficiencies of “wall-plug energy” to EUV into $2\pi$ sr currently being on the order of 1–2% for the most efficient EUV plasmas, an in-band power at IF of 115 W implies an input power on the order of 100 kW. Most of this input power will be converted into heat deposited on the surfaces of solid parts near the plasma. Therefore, a major issue in the design of future EUV sources is how to transport this heat load away effectively. A large collection efficiency means that the necessary input power into the source can be reduced, which will in general be beneficial to the lifetime of the source.

Apart from in-band radiation, also various types of out-of-band radiation will be (partly) transmitted by the optical system. Out-of-band radiation can influence the performance of a lithographic tool in different ways. First of all, the photoresists that are currently proposed for use in EUVL, are not only sensitive to EUV radiation, but also to light at longer wavelengths in the deep ultraviolet (DUV). Because of the longer wavelength, an image of the mask on the wafer in the DUV range will have a much worse resolution than the intended image. Since the multilayer (ML) mirrors show a broad peak in their reflectivity near 200 nm, especially the out-of-band radiation near this wavelength will need to be limited. Further, out-of-band radiation, when it gets absorbed, can also contribute to heating of optical elements. This will especially be the case for the first elements of the illuminator optics, where the radiation power is the highest. How high the maximum

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1 Actually, it may perhaps make more sense to define a requirement for spectral purity in a smaller wavelength band around 200 nm, instead of just in the broad range of 130 to 400 nm as given in Table 2.1.
allowable absorbed power will be is still uncertain, as it is not fully clear yet how effective
the cooling systems of the optical elements will be in pumping away the absorbed power.

Due to the uncertainties in the requirements, and even more so due to those in the
measurement of out-of-band power of existing source concepts, it still remains uncertain
whether these sources will be ready for application without a spectral purity filter. Such
an additional wavelength filter would of course have the major drawback that it would
further reduce the transmission efficiency of the complete optical system, so that even
more in-band power from the source would be required.

As EUV plasmas will always be pulsed sources, a further requirement is a high repetition
frequency. This requirement is connected to the fact that commercial EUVL tools will be
scanning systems. A high repetition frequency is needed to ensure sufficiently homogeneous
illumination of all parts of the wafer as it is being scanned. Certainly, a high repetition
frequency will also be necessary to achieve the required output powers. Therefore, this
explicit requirement does not in practice put any additional constraints on the source
designs. Also connected to the homogeneity of illumination, however, is the integrated
energy stability, measured over 50 pulses. Most source suppliers use some kind of pulse-
to-pulse feedback mechanism to compensate for individual pulses that are much stronger
or weaker than average, hence ensuring stable long-term operation.

Finally, a highly important requirement is the cleanliness of the source. Apart from
radiation, plasma sources also tend to emit both thermal and non-thermal atoms and ions,
as well as (especially in the case of discharge plasmas) larger droplets or clusters of the
working element or nearby electrodes. All this so-called “debris” material can degrade
the quality of the illuminator optics, and hence, its creation needs to be avoided or it
needs to be kept from reaching or passing the IF. For the latter goal, various types of
debris mitigation are under development, including the application of buffer gases or gas
curtains, secondary plasmas, magnetic fields, and “foil trap” debris filters. Apart from
this, methods are being investigated to clean optical surfaces during or after the deposition
of debris on them, and to make the optics themselves more resistant against degradation.
Several contributions on these topics can be found a.o. in Refs. [3–5]. To encompass the
various effects that the debris can have on the optics, the requirement is specified in general
terms of lifetime until a 10% loss of EUV throughput in the optics after the IF is reached.

Even when one or more sources will prove to meet all the requirements listed above, there
will be other issues that determine whether they will indeed be applicable in a commercial
EUVL tool. First of all, the lifetime of the collector optic is not explicitly specified.
However, unless the lifetime of this optic is sufficiently long, and/or the replacement of
the optic after it has reached its lifetime is a simple and quick procedure, the downtime of
the machine for servicing the source may become too long and hence too expensive. More
generally, the cost of ownership will become an important factor. This includes not only the
purchase price of the source, but also the price of consumables (e.g., xenon gas, electrical
input power, replacement electrodes), replacement collector optics, servicing, covered floor
space, etcetera.

### 2.1.2 Sources of EUV radiation

In Chapter 1 and in the discussion above, we have already focused on plasmas as the most likely EUV light sources for application in lithography. However, a number of other concepts of EUV generation exist as well. In the following, we will describe some of these concepts, and also explain the reasons why they are currently not considered as candidate sources for EUVL. More or less complete overviews of technological EUV sources were given by De Boer in 1998, by Attwood in 1999, and by Samson and Ederer in 2000. A more recent overview of coherent vacuum ultraviolet (VUV), EUV, and X-ray sources is given in a special issue, published in 2004, of the IEEE Journal of Selected Topics in Quantum Electronics. Concepts for generation of EUV radiation include synchrotrons, free electron lasers (FELs), high-harmonic generation (HHG) in gases, Cherenkov radiation from thin foils, X-ray lasers (XRLs), and electron-impact X-ray tubes. These will be briefly discussed below.

A **synchrotron** consists of a source for electrons, an accelerator to get the electrons to a specific required energy, and a storage ring. The electrons emit photons when they are accelerated in a magnetic field. In the case of the bending magnets that are used to keep the electrons in the storage ring, the radiation is emitted into a broad spectrum. **Wigglers** and **undulators** are insertion devices that are placed in straight sections of the storage ring with the specific goal of generating a certain type of radiation. Both consist of an array of magnets, such that the electrons travel over an oscillating path. In the case of a wiggler, the emitted radiation is simply the sum of that of the individual magnets: much stronger (compared to a single magnet), but still broadband radiation is produced. In the case of an undulator, the shape of the magnetic field sensed by an electron is such that coherent interference causes the emission of narrowband radiation. Although synchrotrons are not regarded as actual candidate sources for lithography, they are routinely applied in EUV research and development; for example for ML mirror reflectivity measurements and for the calibration of light sensors.

A further increase of radiation brightness from undulators can be achieved when the electron bunch is short compared to the emitted wavelength. In this case, the electric fields from different electrons add up coherently, and self-amplified spontaneous emission (SASE) can take place. This is the principle of the **free electron laser** (FEL). First operation of a SASE FEL in the VUV range has been reported in Ref. A more recent overview of coherent vacuum ultraviolet (VUV), EUV, and X-ray sources is given in a special issue, published in 2004, of the IEEE Journal of Selected Topics in Quantum Electronics. Several facilities operating in the VUV to soft X-ray ranges have been proposed. These will be based on linear electron accelerators, and they will be seeded by either another undulator or a high harmonic generated beam.

**High-order harmonic generation** is another method of generating short-wavelength light. Atoms ionized by strong laser fields can emit coherent radiation of photon energy much higher than their ionization potential, if the “quasi-free” electron recombines into the...
Chapter 2: Plasma sources of EUV radiation

ground state of the atom. Wavelengths into the EUV range have been produced by application of ultraviolet laser irradiation [14]. Further developments have gone into two directions. On the one hand, HHG down into the so-called “water window” of 2.3 to 4.4 nm has been achieved by applying ultrashort, high intensity laser pulses [15, 16]. Due to the short pulse durations and mixing of various harmonics, a continuum spectrum is produced. On the other hand, a high spectral purity, fully tunable source in the 40–100 nm range has been created by applying a wavelength-tunable laser pulse of ~300 ps duration, in combination with a spectrometer to separate the different harmonics [17].

Cherenkov radiation [18] is created when a high-energy (MeV range) electron beam is targeted at a material in which the electron velocity exceeds the phase velocity of light. It is emitted into a narrow cone in the forward direction; hence, to avoid reabsorption, thin foils are applied. Cherenkov radiation can only be created at wavelengths for which the refractive index of the material is larger than unity. In the EUV and soft x-ray ranges, this only occurs in narrow bands near the inner-shell absorption edges of the material. Therefore, the radiation is narrowband, and the wavelengths can be selected by an appropriate choice of the foil material. The etendue of a Cherenkov radiation source can be very small, since the emitting area is determined by the electron beam spot size, and radiation is emitted only into a small solid angle. Therefore, despite the modest total output, the source brightness can potentially be very high.

X-ray lasers (XRLs) are based on population inversion of ionic excited states in plasmas. The two main pumping schemes are recombination pumping in a relatively cool, but still dense plasma, and collisional pumping, followed by stimulated emission to a strongly radiative lower level. Both methods have already been demonstrated in the 1980s in laser-produced plasmas [19, 20], but gain-length products remained too low for practical applications. More recently, new schemes using transient excitation of ions have improved the lasing efficiency. This has become possible by application of femtosecond laser pulses, which generate optical field ionization (OFI). These developments have opened the path to table-top experiments. Soft X-ray lasing is also possible in discharge produced plasmas. Rocca et al. [21] have developed a relatively compact setup for lasing in a capillary discharge at the 46.86 nm line in neonlike argon.

Finally, in electron-impact X-ray tubes keV electrons produce X-rays in the anode material by photo-ionization and bremsstrahlung processes.

All methods except synchrotrons and FELs exhibit very low output powers and/or conversion efficiencies. Although these output powers may be scalable to a certain extent, such scaling would probably require using a large number of sources in parallel, which would complicate the design of the optical system. Even in that case, the achievement of high enough powers for use in commercial EUVL would remain questionable at best. Of course, this does not rule out the possibility that these sources may be used for certain low-power EUVL related applications, e.g., where ultrashort pulse duration, narrow bandwidth, low divergence, and/or strongly coherent radiation is required.
Synchrotrons and FELs, on the other hand, are in principle capable of producing a sufficiently high amount of in-band radiation for HVM, while simultaneously meeting the other requirements. However, both types of sources would require (by comparison) huge capital investments and amounts of floor space. Even though one source could be shared by multiple lithography machines, the investments and the inflexibility of such a source would make the source economically unacceptable.

The only remaining concept that is potentially commercially viable, is that of pulsed plasmas. Therefore, the remainder of this chapter is devoted to the discussion of plasmas as sources of EUV radiation. As indicated in Chapter 1 they can be subdivided into two groups: laser produced plasmas (LPPs) and discharge plasmas. The two are generated in different ways, and also the physical processes by which they are governed are partly different. However, as stated in Sec. 1.4 they also have many characteristics in common. The physics of both LPPs and discharge plasmas, as well as overviews of their subtypes, are given in Secs. 2.2 and 2.3 respectively. More attention is paid to the discharge plasmas, since they form the main research subject of this thesis. The two specific types of discharge plasmas on which experiments, described in this thesis, have been performed, are a hollow cathode triggered discharge in xenon and a laser-triggered discharge in tin vapor. These two concepts are treated in more detail in Secs. 2.4 and 2.5 respectively.

2.2 Laser produced plasmas

2.2.1 Physics of LPPs
The basic principle of laser produced plasmas is extremely simple. A high-intensity laser pulse is fired onto a target material of some kind. Through absorption of the laser energy, a hot plasma is created that emits (EUV) radiation. Some details will be given below; a more elaborate description, which was the basis for a computer model of the evolution of an LPP, can be found in Ref. [22].

For an LPP to start, first some initial electrons need to be generated. Since usually the photon energy of the laser is not high enough to directly ionize atoms in the target, the plasma initiation can only happen through the nonlinear process of multiphoton ionization, in which an atom absorbs several photons in a very short time, after which it ionizes.

After the electron density has risen sufficiently high, the process of inverse bremsstrahlung (IB) [23, 24] takes over as the main absorption process of laser energy—see also Sec. 8.3.2 of this thesis for a description of the IB absorption coefficient. As IB increases strongly with electron density, for sufficiently high laser power the electron density will quickly reach the critical density, which is the density at which the laser frequency equals the plasma frequency. Above this density, the laser light can no longer propagate through the plasma, and most of the light is reflected. Only at the edges of the plasma, where the density is still below the critical density, can further absorption still take place. Hence, a laser produced plasma is often characterized by an electron density that is close
Chapter 2: Plasma sources of EUV radiation

to the critical density for the applied laser wavelength, irrespective of the exact properties of the laser or the target. Shorter laser wavelengths lead to denser plasmas.

Critical electron densities are $9 \times 10^{27}$, $4 \times 10^{27}$, $1 \times 10^{27}$, and $1 \times 10^{25}$ m$^{-3}$, for laser wavelengths of $\sim 355$ nm, 532 nm, 1064 nm, and 10.6 $\mu$m, respectively. These are the wavelengths of—in the same order—xenon fluoride (XeF) excimer or third harmonic neodymium-doped yttrium aluminum garnet (Nd:YAG), second harmonic and fundamental Nd:YAG, and carbon dioxide (CO$_2$) lasers. With the exception of the latter, these electron densities are so high that—given the typical electron temperatures of 20–50 eV in EUV producing LPPs—the plasma will be in or close to local thermodynamic equilibrium (LTE). When we consider population densities of ionization stages and excited states, this means that radiative processes can be ignored in comparison with collisional (de)excitation and ionization, and three-particle recombination, of the relevant ions. Under these circumstances, the population densities of excited states within one ionization stage of an element can be described using the Boltzmann expression,

$$\eta(p) = \eta(p_1) \exp\left(-\frac{E_{\text{exc}}}{k_B T_e}\right),$$

(2.1)

where $\eta(p)$ and $\eta(p_1)$ are the population densities per statistical weight of a certain excited state $p$ and of the ground state $p_1$ of the same ion, respectively, and $E_{\text{exc}}$ is the energy of the excited state relative to the ground state. Further, the different ionization stages of an element are populated following the Saha balance,

$$\eta_z = \frac{1}{2} \eta_{z+1} n_e \frac{\hbar^3}{(2\pi m_e k_B T_e)^{3/2}} \exp\left(\frac{E_{\text{ion}}}{k_B T_e}\right),$$

(2.2)

in which $\eta_z$ and $\eta_{z+1}$ are the ground-state population densities per statistical weight of consecutive ionization stages of a certain element, and $E_{\text{ion}}$ is the ionization energy of the lower stage.

Due to the high electron densities, the excited states of the ions are relatively strongly populated, and therefore a large amount of radiation is generated. A negative effect of the high densities is, however, a strong reabsorption of the radiation. As a consequence, LPPs often radiate close to the black body limit over a large part of the spectrum.

Shapes and sizes of LPPs of course depend on the experimental characteristics; however, either through choice of a narrow laser focus, or of a small size liquid or solid target (e.g., a micro-droplet), the etendue of emitted EUV radiation can be easily tuned to meet the industry requirements as quoted in Table 2.1. In case of such a mass-limited target, the zone of EUV emission will often have a more or less spherical shape, as the plasma generated by the laser starts to expand homogeneously in all directions.

It is also the plasma expansion that limits the effective lifetime of an LPP. Since the LPP is not spatially confined in any way, thermal energy is easily converted to the kinetic energy of expansion. Due to the associated drop of electron density, the laser energy can no
2.2 Laser produced plasmas

longer be absorbed efficiently. Because of the plasma cooling and the decreasing densities caused by the expansion, the emission of intense EUV radiation stops.

2.2.2 LPP types

Laser produced plasma types can be distinguished by the properties of the laser pulse(s), and by the selected target material. Parameters of the laser pulse include not only the laser wavelength mentioned above, but also the pulse duration, the diameter of the focus, and the peak intensity. Additionally, multiple beams may be fired on a single target simultaneously from different directions to achieve sufficient power. Also, one or more short prepulses may deliver very high peak intensities that enhance the nonlinear processes that are required to start the plasma.

Targets may be, e.g., solid material surfaces, thin metal tapes, liquid jets or droplets or gas jets. Also, targets based on solid clusters or jets of xenon have been developed; see below. Large solid targets tend to produce a lot of debris in the form of solid material clusters ejected into the vacuum; therefore, mass-limited targets, such as trains of clusters or droplets from a spray jet, are favored. On the other hand, higher density plasmas are more efficient radiators, and therefore solid surfaces typically result in somewhat higher conversion efficiencies than mass-limited targets.

In the nineties of the last century, research and development of laser produced plasmas for generation of EUV radiation was carried out, among others, at Sandia National Laboratories in Livermore, California [25], in the group of Richardson at the Center for Research and Education in Optics and Lasers (CREOL) of the University of Central Florida, at University College Dublin by O’Sullivan and co-workers [26], at TRW [27], and at Lawrence Livermore National Laboratory [28, 29]. In Ref. [29], a series of experiments is described in which the EUV emission around 13.4 nm was measured for a large number of target elements, and it was already found in that work that tin is one of the most efficient radiators. At the CCLRC Rutherford Appleton Laboratory (RAL), a facility was built and operated for the creation of (soft) X-ray emitting LPPs by Turcu and co-workers [30]. At Philips Research in Eindhoven, a water droplet LPP has been in operation for some time [31, 32]. The parameters of this source were the basis of the model described in Ref. [22]. In the Swedish company Innolite, a spin-off of the Royal Institute of Technology in Stockholm, Sweden, a method was developed for creating a stable, collimated (either liquid or frozen) xenon jet, which was used as a target for creating an LPP [33–35].

Although initially it was found that tin was a particularly efficient radiator near the wavelengths of interest for EUVL, attention quickly shifted to xenon since it causes fewer problems with the generation of debris [25]. However, more recently, attention has shifted back to alternative elements (mainly lithium and tin), since they make a better chance of meeting the industry requirements on the topic of in-band power.

Current development on LPPs, or parts thereof, is continuing in a number of places [3]. Richardson and co-workers at UCF—in partnership with JMAR, a laser systems
company—are working with tin-droplet targets using laser apparatus inherited from TRW. The group of O’Sullivan at UCD is performing research on LPPs produced from both solid tin and composite targets containing tin. CYMER, a San Diego-based company that is also a large supplier of excimer lasers for DUV lithography tools, has committed itself to LPPs from lithium droplets. Contrary to tin, lithium has a spectrum that consists of a limited number of isolated lines, one of which is at 13.5 nm, and therefore emits relatively little out-of-band radiation. CYMER intend to use xenon fluoride (XeF) excimer lasers for the further development of a commercial EUV source.

As of February 2005, XTREME Technologies in Germany, which is a joint-venture of Lambda Physik and JENOPTIK Laser, had not yet made the choice between LPP and discharge plasma as their main candidate EUV source (see also below); their LPP work is focusing on using solid-state and CO\textsubscript{2} lasers on xenon droplet targets. In Japan, the EUVA research consortium are considering the application of a xenon droplet supply system in combination with CO\textsubscript{2} lasers as the way to reach the required levels of output power. In France, EXULITE is working on Nd:YAG laser pulses on xenon targets; however, due to the acousto-optical Q-switching that they apply and the resulting long laser pulses, their conversion efficiencies are staying behind considerably with the state-of-the-art in this field. Finally, Powerlase in the UK are focusing on the development of laser modules that can be applied for LPPs, rather than on development of complete LPP systems.

LPP sources have in common that in principle, it is not difficult to scale them to a source that produces the required 115 W output power; but to do so at an acceptable cost is a big challenge. The capital investment for the required laser modules will be enormous. An advantage of LPP systems compared to discharge plasmas is the fact that due to the relatively high conversion efficiency of laser power to EUV, the heat load near the target is limited. On the other hand, an additional conversion step, from electrical power to laser light, is required, so that the overall power consumption will be much larger for LPPs.

## 2.3 Discharge plasmas

### 2.3.1 General discharge plasma evolution

Another method to create a hot, dense plasma is to make a discharge in gaseous material. Two metallic electrodes are typically arranged in a cylinder-symmetric configuration, such that an electric current can flow in the axial direction. The electrodes are connected to a high voltage power supply, which usually consists of a battery of capacitors, which is in turn connected to another, external power supply. The way the discharge is ignited depends on the specific type of discharge. After ignition, Ohmic heating leads to further ionization of the plasma. In all cases, the magnetic field that is generated by the multiple-kA current causes a Lorentz force acting on the charged particles in the plasma. On average, for both the ions and the electrons, this force is directed towards the axis of symmetry of the discharge, due to their non-zero average velocities caused by the current. In a stable
2.3 Discharge plasmas

situation, the magnetic pressure is balanced by the thermal pressure in the plasma. This pressure balance is known as the Bennett equilibrium. However, if the magnetic pressure gets larger than the thermal pressure, the balance gets disturbed and the plasma will start to compress in the radial direction. This is called the pinch effect. Compression can end if the pressure imbalance is reversed, e.g., through further (compressional and Ohmic) heating of the plasma, or a decrease of the electric current. The latter can happen either due to the pinch effect itself, which will cause an increase of plasma inductance and resistance, or as a result of the inherently oscillating nature of the current in an LC circuit.

The pinch effect leads to an increase of the electron density in the plasma. Still, typical densities in discharge plasma EUV sources are much lower than those in LPPs; it is estimated that the peak electron density is typically on the order of $10^{25} \text{m}^{-3}$. Under these circumstances, and taking into account the high temperatures and multiple ionization of the plasma, the assumption of LTE is not valid. Instead, radiative deexcitation and recombination of ions play important roles in determining the excited level and ion stage populations. Under such circumstances, an increase of the electron density leads to a decrease of the relative importance of radiative deexcitation for the excited state populations, and hence to an increase of the absolute amount of emitted radiation. Also, the transient nature of the discharges can play a role in the ion stage and excited level populations. A detailed discussion of these populations in discharge plasmas can be found in Sec. 6.2.

During the pinching of a discharge plasma, several types of instabilities can occur. One of these is the Rayleigh-Taylor instability, analogous to the instability that occurs when a light fluid supports a heavier one in a gravitational field. An overview of instabilities in (pinch) discharge plasmas is given in Ref. [36]. Pinch instabilities can form the onset of radiative collapse, which occurs when energy losses from the plasma due to (line) radiation are larger than the energy gained from Ohmic heating. This can lead to a deep compression, the result of which is called a “micropinch”. This effect has been investigated extensively by Koshelev and others [37, 38]; see also Sec. 2.3.4 below. Under the moderate conditions of devices designed for EUV generation, however, micropinchig is less likely to occur.

In the remainder of this section, a number of discharge plasma types that have been or are still being investigated, will be discussed. The last two sections of this chapter describe in some more detail the two types that have been the subject of the research work described in this thesis: a hollow cathode triggered discharge in xenon, and a laser-triggered discharge in tin vapor.

2.3.2 “Classical” Z-pinch

Gas-puffed Z-pinch discharge plasmas have already been considered since the early 1980s as potential radiation sources of (proximity) X-ray lithography [39, 40], with input energies per pulse in the several kJ range. In more recent years, Z-pinch designs have been changed to make them suitable for EUV generation. A schematic drawing of the most basic form of Z-pinch is shown in Fig. 2.1. A discharge takes place between two parallel electrodes,
Fig. 2.1. Schematic picture of a basic Z-pinch discharge, adapted from Ref. [41]. The dashed arrows show the direction from which the emitted radiation can be collected and/or observed.

that both have large holes in them for introduction of the working gas, and viewing of the output radiation. The discharge starts near the insulating wall, and moves to the discharge axis due to the pinch effect described above. Some form of active preionization is required to stabilize the discharge. A spark-gap switch is required in the electrical circuit to build up the electrical potential between the electrodes fast enough.

Z-pinches as EUV sources have been developed at PLEX LLC in the USA by McGeoch [42] and at XTREME Technologies in Germany [43].

The Z-pinch source that was developed by McGeoch, made use of strong rf preionization, which reportedly resulted in a diffuse and arc-free discharge. It was operated in xenon. The minimum pulse energy was reported to be 150 J, which would make high-repetition rate operation difficult. Also, given its length and diameter of about 4 cm and 2 mm, respectively, it did not fit the maximum etendue requirements as quoted at the beginning of this chapter. Further development of the Z-pinch source has been abandoned at this company in favor of the Star pinch device (see below).

Unlike the Z-pinch source of PLEX, the version of XTREME Technologies is still under development as a candidate source for high-volume manufacturing. In this source, preionization is achieved through a “sliding” surface discharge between the cathode and an additional third electrode that doubles as a gas inlet. Input energies per pulse are up to 10 J. With pinch sizes of between 1 and 4 mm, also the plasma sizes are much smaller than in the case of the PLEX Z-pinch. However, matching the lithography etendue still appears to be somewhat a problem for this type of plasma. Xenon and, more recently, also tin have been applied as working elements. Porous metal cooling of the electrodes is applied to make high repetition rate operation possible.

2.3.3 Capillary discharge

Capillary discharge EUV sources are similar to the “classical” Z-pinches described above, apart from the fact that the diameter of the insulating ring between the electrodes is much smaller (typically several mm instead of a few cm), and is now almost on the same order as the pinch plasma diameter. As a result, the channel wall plays an important
role in stabilizing the plasma column, which makes active preionization of the plasma unnecessary. However, a high-voltage switch is still required in the electrical circuit between the capacitors and the discharge region.

The plasma material may consist of a gas that was injected into the capillary, or of wall material in the case of an ablative capillary discharge. Even if ceramics are used as the wall material, the close vicinity of the insulator leads to relatively high levels of debris. Another drawback is that, in principle, the available solid angle for collecting the emitted radiation is very small; see Fig. 2.2.

The EUV emission of xenon from a capillary discharge was studied among others by Klosner and Silfvast at CREOL [44, 45] in cooperation with Sandia. In their source, the effective viewing angle was enhanced by electrode design and pumping “tricks”, which caused the EUV radiation to be emitted mainly from a position near one electrode, rather than from the entire length of the plasma [41]. The inductance of the electrical circuit as reported in Refs. [44, 45] was about 150 nH, leading to a current risetime of about 200 ns. Input energies per pulse were in the 6–9 J range.

A capillary discharge source in xenon is currently under development in the Gotenba branch of EUVA in Japan [46]; first operation in tin has also been reported.

2.3.4 Plasma focus

The arrangement of the electrodes in a so-called “plasma focus” device is different from that in the Z-pinch and capillary discharge devices described above. Plasma focus devices, just like Z-pinches, have an axisymmetric geometry. However, the electrodes in a plasma focus are cylinder-shaped and arranged concentrically, with the anode usually in the center. Both electrodes are connected by insulating material near the end of the central electrode. Again, Lorentz forces play an important role, and shortly after the discharge is started near the insulator surface, the current starts flowing in radial direction, creating an azimuthally directed magnetic field. This magnetic field then pushes the plasma outward until it reaches the open end of the cylinder. There, the geometry of the device causes the discharge to curve over the edge of the anode and “focus” into its hollow interior—hence the name of this plasma type—where subsequently pinching takes place like in the Z-pinch configuration. See Fig. 2.3 for a schematic drawing. Due to the “run-down” phase before pinching, the
current pulse can be an order of magnitude longer than in the source designs discussed above, which makes the design of the electrical circuit easier \cite{41}. The large opening angle for collection of the emitted EUV radiation is another advantage of the plasma focus source.

On the down side, this plasma type often suffers from its inherently bad stability, in the sense of pulse-to-pulse reproducibility of EUV output and emission zone position. This problem is caused by radiative collapse. Dependent on initial conditions, radiation during the pinching phase may serve as an efficient energy loss channel, leading to greatly enhanced compression of the plasma and a very small final plasma diameter. Typically, the diameter after pinching is only a few microns after radiative collapse, while it is a few hundred microns without it.

A plasma focus device, the SPEED 2, was used to study instabilities in pinch plasmas \cite{37,48}. In this device, argon was injected into the deuterium working gas. Apart from the micropinch mode (MPM), also an initially unexpected stable column mode (SCM) was found, and in Ref. \cite{48}, the discharge parameters in which both modes occurred, were investigated. In the SCM case, fast, runaway deuterium ions were found to provide a mechanism for stabilization of the pinch.

Soft x-ray emission from a plasma focus device was also studied by Bergmann, Lebert, and others at the RWTH and the Fraunhofer Institut für Lasertechnik in Aachen, Germany \cite{47,49,50}.

The so-called “Dense Plasma Focus” device, operated in xenon, has been investigated for some time as a possible high-intensity EUVL light source at CYMER \cite{51}, before it was finally abandoned as a candidate source, as stability and electrode erosion remained difficult issues to solve. Typical input energies were in the 10–20 J range. Apart from the configuration described here, also experiments with the reverse electrode configuration have been performed. These offered the advantage of reduced heating and erosion of the central electrode.

### 2.3.5 Star pinch

The “star pinch” \cite{52,54} can best be described as a single pinch plasma, which is connected not to a single cathode and anode, but instead to a large number (typically 24) of cathodes and anodes, which are arranged in a star-like configuration. It was introduced by McGeoch
2.4 Hollow cathode triggered discharge

of PLEX LLC in 2002. In this concept, before the main discharge all electrodes (both cathodes and anodes) act as separate pseudospark switches (see the next section), for which the main discharge volume acts as a common hollow cathode. Small beamlets of xenon ions from the various pseudospark discharges coincide in the main discharge volume. Via charge-exchange collisions with the background gas, a sufficiently high density of xenon atoms is reached without a too high electric potential in the center. Subsequently, due to the action of a switch, the main voltage is applied over the electrodes. During the first half-cycle of the current pulse, of about 800 ns duration, the plasma pinches and of the total electrical input energy of about 25 J, some 10 J is dissipated in the plasma. The remainder is recovered by re-opening the switch after the completion of the current half-cycle.

The main advantage of the star pinch over the previously mentioned types (the capillary discharge in particular), is that all plasma-facing surfaces are relatively far from the pinch volume, so that the heat load from the plasma is spread over a large surface area. Specifically, in the axial direction towards the cathode, where most of the ion flux is directed, the distance to the closest surface can be several centimeters.

More recently, operation of the star pinch in tin has been proposed. This is reached by introducing a tin droplet into the main discharge, which is started in a background of helium with a small amount of xenon added. Notably, the main advantage of this step is not quoted to be the improved conversion efficiency associated with the shape of the spectrum of tin, but rather the reduced etendue of an evaporated droplet (such as in an LPP) as compared to that of a normal pinch plasma.

2.4 Hollow cathode triggered discharge

Another variant of the Z-pinch device is the hollow cathode triggered (HCT) discharge. As the name implies, the main aspect that makes it different from a “classical” Z-pinch is the way the discharge is ignited. Unlike the other discharges, this device is operated in the left-hand branch of Paschen’s curve, meaning that the mean free path of the electrons for ionization is larger than the inter-electrode distance. This has a number of consequences. For example, the operating pressure can be much lower (which also leads to lower reabsorption of EUV light); and the source can be operated without an electrical switch in the main discharge circuit, so that the inductance of the circuit can be very low. Copies of the HCT discharge from Philips EUV are used in the ASML EUV Laboratory. They have been subject of the research described in Chapters 4 and 6 of this thesis. In the following, a brief history of the research and development of this type of source, and some more details on the physical principles are given.

The concept of the hollow cathode triggered discharge has been studied extensively in the past for its application as a so-called “pseudospark switch”, due to its property that it can be used in electric circuits to switch high voltages on very short timescales. Considerable effort was put into experimental investigations and numerical modeling of these switches.
Chapter 2: Plasma sources of EUV radiation

Fig. 2.4. Schematic electrode geometry and plasma shape of the hollow cathode discharge. Note that, in contrast to the devices in Figs. 2.1–2.3, there is no switch in the electric circuit, so that the inductance of the circuit can be very low.

The discharge was investigated for the first time as a possible source of EUV light, through operation with xenon gas, at the Lehrstuhl für Lasertechnik of the RWTH, and the Fraunhofer Institut für Lasertechnik (ILT), both in Aachen, by Bergmann and others. Later, the company AIXUV started developing the source further for laboratory applications, while at Philips EUV, a joint-venture between ILT and Philips Electronics, it was further developed as a potential source for high volume manufacturing.

The device consists of two parallel plate electrodes; see Fig. 2.4. Gas (usually xenon) is introduced into the space between the electrodes from the sides. As said, the electrodes are connected directly to a ring of high-voltage capacitors, without any switch in between. Operating gas pressure and plate distance are such that no discharge takes place directly between the plates. Rather, the discharge is initiated at the position of a hole in the cathode near the center of the plate assembly.

The breakdown phase of the discharge was already extensively described by Boeuf and Pitchford in 1991. When a voltage is applied over the electrodes, electron multiplication starts to take place inside the hollow cathode (HC), where the electric field is relatively weak. The ionization cross section in the space between the electrodes is assumed to be very low at this time, as free electrons easily obtain kinetic energies that are too high for efficient ionization. The free electrons that are generated inside the HC are focussed to a relatively small, intense beam into the inter-electrode space.

Now, collisions of the electrons with neutral atoms in this space lead to further ionization. As the ions are much heavier than the free electrons, they are extracted from this position by the potential differences far more slowly. Hence, a positive space charge is created between the electrodes near the HC position, and it grows towards the opening of the HC (the borehole space).

After sufficient time, the presence of the positive space charge causes a significant deformation of the electric field in the device. Specifically, a region of high electric potential will penetrate more and more into the HC. This in turn causes enhanced electron multiplication inside the HC, as the newly developed electric field causes high energy electrons to...
be trapped inside the HC and “bounce” back and forth between its walls; this is called the *pendulum effect*. As they travel long distances, the fast electrons are able to ionize a large number of atoms. Secondary ionization is especially efficient if it takes place in regions with large electric field, so that newly created electrons can themselves be accelerated to high energies. A strong high-energy electron beam out of the HC into the inter-electrode space, and through a hole in the anode to the surrounding vacuum, is generated. This beam has been observed experimentally in the Philips EUV devices at ASML, and it was used for triggering the optical diagnostics.

With increasing ionization degree, the positive space charge region is transformed into a low-impedance plasma that penetrates further into the HC. Further increase of the electric current is followed by the final breakdown of the discharge. By this time, not only ion collisions, but also other processes such as field-enhanced thermionic emission, contribute to the emission of electrons from the cathode [67].

From this point onwards, the evolution of the discharge is similar to that of the other Z-pinch-like discharge concepts described above. The electric current causes the plasma to collapse onto the symmetry axis of the device through the magnetic field that it creates. The EUV light that is emitted in this phase can be detected through the anode hole.

After the end of the pulse, a residual degree of ionization of the gas inside the HC can contribute to the ignition of the subsequent pulse. This makes it possible to operate the source in a “self-breakdown” mode, i.e., without any external triggering or switching. The capacitors that are part of the main discharge circuit, can simply be charged slowly until the discharge voltage is reached. Next, the HC effect will take care of the ignition. Note that the gas pressure and electrode geometry can be tuned to let the device discharge at the desired voltage. For example, higher gas flow (and hence higher pressure) will lead to discharge at a lower built-up potential.

Apart from this, the device can also be made to operate at a lower or higher voltage than the intrinsic one, by the introduction of a third electrode inside the HC. Depending on the polarity of the potential between the cathode and this third electrode, ignition can be either stimulated or inhibited. However, the experiments presented in Chapter 4 have been carried out on a device without such an additional electrode.

The fact that the source is operated in the left-hand branch of Paschen’s curve leads to a number of advantages over other sources. First, due to the low inductance of the circuit, the electrical energy can be coupled into the plasma relatively efficiently. Hence a low energy (on the order of 1 J) is sufficient to operate the discharge (although typically a somewhat higher energy of around 3 J is applied), which makes it relatively easy to achieve high repetition rates. Further, the operating pressure is typically in the range of 10–30 Pa, which is much lower than for other discharge types. The low background pressure of xenon gas reduces the reabsorption of emitted EUV radiation, as mentioned above. And finally, no insulating material is needed near the discharge volume, as the plasma “automatically” chooses its position where the longest field lines are (i.e., between the electrode holes).
Therefore, debris from insulators is not a problem. More generally, the heat generated by the pinch (and transferred in the form of radiation and fast particles) can be spread over a relatively large surface area.

Still, it is projected by Philips EUV that the finally achievable in-band output powers will not be sufficient for high-volume manufacturing in lithography. Therefore, operation of the device in alternative working elements (specifically, tin halides) is being pursued.

2.5 Laser-triggered discharge in tin vapor

Around the year 2002, the progress in the development of the existing EUV plasma concepts towards commercially viable EUV sources had been quite fast. Still, it was becoming increasingly clear that plasma sources working on xenon gas might in the end not be capable of producing sufficient output power to meet the industry demands. For this reason, several groups started to investigate both laser produced and discharge produced plasmas in alternative working elements, which have a more favorable shape of their spectrum in the EUV range (see also Sec. 2.2.2). Around the same time, ASML became involved in the development of an alternative EUV plasma concept that had been invented at ISAN in Troitsk, Russia: a laser-triggered vacuum arc discharge. Although it has been operated with vapors of different elements (including lithium and iridium), for reliable operation as a lithography EUV source, tin vapor seems to be the most promising.

Apart from the application of a more efficient radiator, the concept offers a few other potential advantages. These will be explained below; first, the principle of operation and the evolution of a basic version of the discharge will be described in more detail.

Like in the hollow cathode discharge concept, the discharge electrodes are directly connected to one or more rings of capacitors, without any additional switch in the electric circuit. The capacitors can be charged at a relatively slow rate; a high voltage on the electrodes can be built up without problem due to the fact that they are separated by a vacuum or a gas at very low pressure. Typically, the cathode is put at a negative voltage while the anode is kept grounded.

A schematic picture of the typical setup is shown in Fig. 2.5. The bottom electrode, the cathode, is covered with a layer of liquid tin, which can be heated from below using a heating spiral. Like in other discharge concepts, there is a hole in the anode. However, in this case it does not serve for observing the plasma or collecting the EUV radiation, but to admit the passage of a laser pulse that is focused onto the cathode surface.

Due to absorption of laser energy in the tin pool, some of the liquid tin gets vaporized and (partly) ionized. After the end of the laser pulse, the tin vapor plume expands, and after some time (of typically a few hundred ns), the front of the plume reaches the edge of the anode. Once the plasma density near the anode has become sufficiently high and a conducting path between both electrodes has been created, an electric discharge can start.

Again, from here on, the discharge evolves, in principle, in a similar way as in the other
Z-pinchlike discharge concepts: the discharge itself causes further ionization and heating of the plasma, and the pinch effect occurs due to the Lorentz force created by the electric current. The pinch plasma finally dies due to the lack of confinement of the plasma in the axial direction, and the finite duration of the current half cycle. The plasma expands into the vacuum or low pressure background gas, and conditions are restored for the start of the subsequent discharge pulse.

Summarizing, the evolution of the plasma can be roughly split up into four main phases, similar to those of other discharge plasma types:

i. an *ignition* phase, in which only a laser-induced plasma exists, and the electric current has not started yet;

ii. a *prepinch* phase, in which a strong electric current has taken over as the energy supply for further heating and ionization of the plasma;

iii. a *pinch* phase, with (relatively) high density plasma, caused by radial compression; and

iv. a *decay* phase, that starts as soon as the pinch plasma begins to expand, and ends with the restoration of vacuum between the discharge electrodes.

As mentioned above, this type of source offers some specific advantages that are related to its geometry and way of operation. Of the two main ones, the first is related to the fact that it has a relatively open geometry. As illustrated in Fig. 2.5, emitted EUV radiation cannot only be observed and collected through the anode hole, but also from directions in a large solid angle perpendicular to the axis of symmetry of the pinch plasma. It has been observed that by far the most debris created by the source, both in the forms of atomic tin and tin droplets, is emitted in a direction close to the symmetry axis. Perpendicular to the pinch, relatively little contamination by metallic atoms is found. A possible explanation of this distribution is offered in Sec. 5.3. The debris distribution partly compensates for the fact that source debris is in itself more of a problem when tin is used than in the case of xenon.
The fact that the collector optics in a lithography tool can be placed perpendicular to the axis of the pinch plasma, is an advantage over other pinch plasma concepts. However, the drawback of this choice is that the plasma exhibits a larger etendue when viewed perpendicularly to the pinch. This means that the pinch length should be very limited to match the etendue required by the optical design.

The second main advantage of this discharge type is the way the heat load problem is dealt with. Laser ignition of the discharge makes so-called multiplexing of the source relatively straightforward. In a modified geometry of the setup, the electrodes can be replaced by large rings, which rotate around a common axis at a certain angular frequency—note, however, that in this case, the “local” cylindrical symmetry of the electrodes with respect to the discharge position may be broken. For each discharge pulse, the ignition laser can be focused to the same position in space, but due to the rotation of the electrodes, in each pulse, the heat gets dissipated at different parts of the electrode surfaces. In this way, all EUV pulses originate from a fixed position relative to the collection optics, but the associated heat load is spread evenly over a large area of both the cathode and anode rings. Hence, the electrodes can be cooled more easily. Given a sufficient number of discharge locations on the rings, high repetition frequency operation (on the order of 5 kHz) can be achieved with relatively moderate rotation frequencies of the electrode rings, of only around 5 Hz.

Many of the experiments described in this thesis have been performed on a basic version of this discharge type (without rotating electrodes or multiplexing). A relatively small high voltage power supply and ignition laser were used, and operation up to about 10 Hz was possible. The main reason to select this discharge type for various experiments, is the fact mentioned above that the plasma can be easily accessed and observed from various directions, not just “head on” from the rotational axis of symmetry. Experiments on this plasma type and their results are described in Chapters 5–8 and 10.

References


Chapter 2: Plasma sources of EUV radiation


Chapter 2: Plasma sources of EUV radiation


Abstract

Optical diagnostics are indispensable for obtaining detailed knowledge on EUV producing plasmas. These diagnostics can be subdivided in multiple ways; the most important distinctions are those between active and passive diagnostics, and those performed “at-wavelength” versus those at other wavelengths. To capture the inherently transient behavior of pulsed EUV discharges, it is important to perform any kind of measurement in a time-resolved way. Optical diagnostics include plasma imaging (in the visible and EUV wavelength ranges), EUV spectrometry, absolute intensity measurements, line broadening measurements, Schlieren photography and laser interferometry, and Thomson scattering.
3.1 Introduction

As discussed in Sec. 1.4, optical diagnostics in general are indispensable for obtaining detailed knowledge on EUV producing discharge plasmas. When selected and applied properly, they may give space and time resolved information on electron temperatures and densities, atom or ion excited energy level populations, plasma optical constants, etcetera, without excessively disturbing the evolution of the plasma. Several of these methods have been applied in the present work; they are the subjects of different chapters of this thesis.

In this chapter, a general (but not necessarily complete) overview will be given of optical diagnostics that have been or could be successfully applied to EUV producing plasmas. Given the wide range of plasma parameters that occur during the evolution of a plasma pulse, the range of different diagnostics is also very wide. Only those that require very stable plasma properties (due to, e.g., long integration times) are less suitable for application to this type of plasma. Just some basic principles and characteristics of the various diagnostics are discussed here; if used in this work, their specific experimental details or other considerations are discussed in the respective chapters. References to those chapters are included in the text below.

A general distinction can be made between passive and active optical diagnostics; here “passive” means that only light emitted by the plasma itself is considered, whereas in active diagnostics, the plasma is irradiated by an external light source, and the observer records light that is subsequently scattered, refracted, reflected, absorbed or emitted by the plasma. The advantage of passive over active diagnostics is that with passive diagnostics the plasma cannot be influenced in any way, whereas with active diagnostics, the possible influence of the external light source on the plasma properties always needs to be considered carefully. On the other hand, active techniques can make it easier to derive certain properties, especially when spatially resolved information is desired.

A further subdivision of passive techniques, in the context of EUV producing plasmas, can be made into at-wavelength and other wavelength diagnostics. Here, we define “at wavelength” as within the EUV range of roughly 8 to 21 nm. At-wavelength diagnostics give the most direct information of the plasma properties in terms of the future application. Also, in general, for those parts of the plasma evolution in which the most EUV radiation is emitted, also the most information is contained in the EUV range of the spectrum, since this is where the more highly ionized species in the plasma tend to radiate. However, in other, cooler phases, low ionization stages of the same element tend to dominate, and they will in general radiate at larger wavelengths. Therefore, it is also useful to apply diagnostics at larger wavelengths (usually in the visible light range) to learn more about those phases of the discharge, and hence obtain a more complete picture. Also, it is much easier to obtain sufficient spectral resolution at visible light wavelengths than it is in the EUV range.
As a final general remark, the fact that the plasmas under study are transient by nature, makes the application of time-resolved measurements necessary. The typical required time resolution is about 5–10 ns. Time-resolved optical measurements can usually be performed by switching the detector on and off; often this is the intensifier of an intensified charge coupled device (ICCD) camera. However, usually the integration time is too short to collect a sufficiently strong and noise-free signal in a single pulse. In such cases, the detector is gated at the appropriate time during each of a certain sequence of pulses, and the signals from the individual pulses are integrated—either on the detector itself, or digitally on a computer.

In the following sections, a number of techniques will be discussed. We start with certain passive diagnostics: plasma imaging (Sec. 3.2), EUV spectrometry (Sec. 3.3), absolute intensity measurements (Sec. 3.4), and Stark broadening (Sec. 3.5). The chapter is concluded with two groups of active techniques: Schlieren photography and laser interferometry (Sec. 3.6) and Thomson scattering (Sec. 3.7).

### 3.2 Plasma imaging

#### 3.2.1 General

A very important and often underestimated imaging device is the human eye. It can provide a good and fast indication of discharge position and stability; however, it is, of course, limited in terms of, a.o., time resolution and wavelength sensitivity. Also, external tools are required to properly record and reproduce an observed image.

In the context of EUV producing discharges, time-resolved imaging is of particular importance, as the discharge never reaches anything close to stable conditions. As plasma imaging belongs to the passive methods, the only way to obtain time resolution is to use some method of gating the camera. Therefore, any experiment must consist of optics, a sensor (the camera) which can be gated, and synchronization of the gating to the evolution of the discharge.

The wavelength sensitivity of the method is determined by the optics and the sensor together; for light of a certain wavelength to be recorded, it should be transmitted and properly imaged by the optics, and the sensor should also be sensitive to the particular wavelength. Below, two (broad) wavelength ranges of particular interest will be discussed: the extreme ultraviolet (EUV) and the visible.

#### 3.2.2 Visible

The visible wavelength range is not so much of interest because it can be directly observed by the eye, but rather because high-quality lenses and sensors are readily available. Achromatic lenses provide the best resolution, even if only a narrow wavelength range is recorded. Commercially available ICCD cameras can be used as sensors. These typically are sensitive to a broad wavelength range within the visible part of the spectrum, and
they can be gated with a time resolution (typically on the order of 1 ns) that is sufficient for the application. In this work, visible light plasma imaging has been carried out for the tin vapor discharge; the experiment and its results are described in Secs. 5.2 and 5.3 respectively. We have not performed such experiments for the hollow cathode discharge.

3.2.3 EUV

Imaging of the plasma in the EUV range—especially around the wavelength of 13.5 nm—is of interest not only for improved qualitative understanding of the plasma, but also directly for the future application, in which the plasma will be used as a light source in a larger optical system. Hence, some form of plasma imaging in the EUV is quite generally applied to characterize proposed EUV sources.

As was stated already in Sec. 1.2 of this thesis, at these wavelength ranges, lenses cannot be used as optical elements; an alternative is the application of mirrors. The simplest useful imaging system consisting of mirrors is that of the Schwarzschild microscope, which is a combination of one concave and one convex mirror. Since both are near-normal incidence mirrors, they must have multilayer coatings for good efficiency, and are hence automatically optimized for a certain small wavelength range. This can be an advantage for the above-mentioned application, in which the apparent shape and size of the plasma at a specific wavelength needs to be known.

A more simple alternative is the application of a pinhole, which transmits only the light that passes through a small aperture in a foil. The resolution of the image that the pinhole produces, is directly related to the wavelength of the light. Simple expressions exist in two limiting cases of large and small pinhole radii, in which the resolution is limited by geometrical imaging and by diffraction, respectively. These expressions are given by

\[
R_{\text{geo}} = r \left(1 + \frac{L_{\text{s-p}}}{L_{\text{p-c}}}ight), \quad \text{and} \\
R_{\text{diff}} = 0.61L_{\text{s-p}} \frac{\lambda}{r},
\]

respectively. Here, \(r\) is the pinhole radius, \(\lambda\) the wavelength used for imaging, \(L_{\text{s-p}}\) the distance between the light source and the pinhole, and \(L_{\text{p-c}}\) the distance between the pinhole and the sensor (camera or otherwise). \(R_{\text{geo}}\) and \(R_{\text{diff}}\) are the radii of smallest resolvable features inside the plasma. The resolution for intermediate cases cannot be given by such a simple expression [1]; however, remarkably, it tends to be better than the largest of the two limiting values given above. For a given pinhole radius, the optimum value for the source to pinhole distance is roughly \(L_{\text{s-p}} = r^2/\lambda\), assuming that the distance from the pinhole to the sensor is much larger than that from the source to the pinhole; i.e., the magnification factor \(M = L_{\text{p-c}}/L_{\text{s-p}}\) is much larger than unity. In this case, the smallest resolvable features in the plasma have roughly the same size as the pinhole itself.
3.3 EUV spectrometry

As the imaging resolution is related to wavelength, it is adversely affected if light at wavelengths larger than the intended range is also recorded. To ensure good spatial resolution, and to limit the image to the wavelength range of interest, thin foil transmission filters can be used. Also, the sensor can be selected to be sensitive only in a certain wavelength range.

EUV sensitive photographic films are frequently used sensors for plasma imaging. Their main drawback is, of course, that they cannot be gated (at least not on a ns timescale) so that only pulse-integrated images can be obtained.

Time-resolved pinhole images in the EUV wavelength range have been recorded for both the hollow cathode triggered discharge and the tin vapor discharge; the results are presented in Chapters 4 and 5 respectively. For these measurements, a *multichannel plate* (MCP) was used to gate the detector, to convert the EUV radiation to visible light, and as a vacuum-to-air interface. Since the MCP is only sensitive to wavelengths below roughly 100 nm, it helped to filter out light at longer wavelengths. More details, including schematic images of the pinhole camera, can be found in the respective chapters.

3.3 EUV spectrometry

For EUV spectrometry, even more than for EUV plasma imaging, a setup with a minimum of optical elements is desired. Multilayer mirrors can not be used in this case as these are themselves strongly wavelength-selective, so that a spectrum over a reasonable wavelength range cannot be produced. This leaves only two main types of gratings that are commonly used in EUV spectrometers: transmission gratings and curved grazing-incidence reflection gratings.

In *transmission grating* spectrometers, no focusing is done at all. This means that the spectral resolution is directly limited by both the size of the source and the size of the grating. On the other hand, the grating should also not be too small, since for too small gratings the resolution is diffraction limited. Hence, it is inversely proportional to the number of periods, or, equivalently, to the total grating width. Further, with decreasing grating size, the efficiency of the spectrometer becomes very low.

Transmission grating spectrometers have the advantage that they are easy to align and can be applied over a relatively large wavelength range with reasonable resolution and efficiency.

*Curved reflection gratings* provide not only wavelength dispersion but also focusing of the light. Reflection grating spectrometers for the EUV can be subdivided into two frequently used types: those employing “classical” ruled gratings with equally spaced lines, and so-called flat field spectrometers.

Curved gratings usually have a spherical shape. The *Rowland circle* is the circle that has a radius that is just half the radius of curvature of the grating, and touches the grating surface in the center. If, in the case of *equally spaced* grooves, the entrance slit of the
spectrometer is located on the Rowland circle, then so is the tangential focal curve. This means that for all wavelengths, the focus in the wavelength direction is on the Rowland circle.

In the case of an MCP or CCD detector, which is flat by definition, it is impossible to follow the Rowland circle exactly. For this case, there are two commonly applied configurations: one in which the detector surface is (more or less) tangent to the circle, and another in which the surface of the detector is (approximately) normal to the incident light. In the latter case, the light is only accurately focused for a single wavelength, corresponding to the point where the Rowland circle crosses the detector plane. However, in the case of a sufficiently small, distant light source, also off-Rowland-circle resolution is still fairly good and a spectrum can be recorded over a relatively large wavelength range in a single experiment. See Fig. 3.1 for a schematic representation of this configuration. It has been used in our EUV spectrometry experiments as described in Chapters 4 and 5 for the hollow cathode triggered discharge and the tin vapor discharge, respectively.

Flat field spectrometers have gratings that are typically not produced by mechanical ruling, but by illumination of a photoresist with a (laser) interference pattern, followed by etching and further processing. The distances between the grooves, rather than being equal for all grooves, are adjusted such that the focal curve is nearly linear for a certain wavelength range. For flat (electronic) detectors, these spectrometers provide high spectral resolution over a larger wavelength range.

A spherical grating also focuses the dispersed light in the sagittal direction (perpendicular to the wavelength axis); however, the focal distance in this direction is much larger than in the tangential direction. This is due to the near-grazing incidence angles at which EUV reflection gratings are typically used to obtain good efficiency. Therefore, the curvature of the grating is not sufficient to obtain spatially resolved spectra. Instead, an additional slit, perpendicular to the entrance slit of the spectrometer, can be placed between the entrance slit and the light source. This additional slit works in much the same way as the pinhole discussed in the previous section. The curvature of the grating can be taken into account as just a small correction to the geometrical magnification factor provided by the slit. The main difference with pinhole imaging is that the slit provides imaging resolution in only one spatial dimension; if the plasma is observed from along its axis of symmetry,
Abel inversion can be applied to obtain radial emission profiles from the “raw” recorded spectra.

Once the detected spectral features have been identified, the results from EUV spectrometry can be used to derive which species, i.e., which ions of a certain element, are present in the plasma at a given time; this is done in Chapters 4 and 5. However, a more elaborate interpretation of the results requires both information on the—absolute or relative—sensitivity of the spectrometer over the relevant wavelength range, and a theoretical model on the emission of the plasma as a function of its basic parameters; see below.

3.4 Absolute intensity measurements and construction of ASDFs

Absolute line intensity measurements of optical transitions in the visible light range are a commonly used technique to derive atomic state distribution functions (ASDFs) in plasmas. Line intensities are connected to the populations of the upper excited states of the transitions through their optical transition probabilities. In case the plasma is sufficiently close to local thermodynamic equilibrium (LTE), the ASDF will follow a Boltzmann distribution, meaning that the population per statistical weight will follow an exponential function of energy, the slope of which (on a logarithmic scale) is related to the electron temperature. An absolute calibration of the intensities (using, e.g., a ribbon lamp) can relate the excited state densities to that of the ground state, which makes the determination of the electron temperature more accurate. In case of LTE, the electron density can be determined by using the Saha balance, or by measuring intensities of ionic lines as well. If LTE is not sufficiently established, more elaborate collisional-radiative models (CRMs) can be used to relate the measured excited state densities to the basic plasma parameters $T_e$ and $n_e$.

In the EUV wavelength range, similar measurements are not straightforward for a number of reasons. First of all, for the heavy, complex ions that are usually present in EUV producing plasmas, the spectra in the EUV range often do not consist of individually resolvable lines, but rather form so-called unresolved transition arrays (UTAs). These are spectral features that consist of up to hundreds or even thousands of closely spaced spectral lines—usually sharing the same upper and lower level electron configurations. Secondly, absolute intensity calibration of the recorded spectrum is less straightforward, since comparison with a source of well defined intensity over the entire wavelength range is usually impossible.

Finally, as mentioned in Sec. 2.3.1, EUV producing discharges tend to be far from LTE. Not only do radiative deexcitation of excited states and radiative and dielectronic recombination of ions play an important role, also the transient behavior of the discharges
Chapter 3: Optical diagnostics

causes deviations from equilibrium, in the form of “lagging” of the distribution of ion stages compared to the instantaneous electron temperature and density. This means that the deduction of plasma parameters from the measured spectrum is not straightforward. A more detailed discussion is given in Sec. 6.2. The model described there is used in Chapter 6 to derive physical information from the EUV spectra of the xenon hollow-cathode triggered and tin vapor discharges.

Also from the point of view of application in lithography tools, absolute emission intensity measurements—both at the operating wavelength of 13.5 nm and at other wavelengths—are of great interest. Several dedicated tools have been developed for this goal. The best known of these is the so-called “Flying Circus” (FC) \[2\]. It was designed in cooperation between the research organization FOM and the companies Philips and ASML, with the specific goal to set a standard for the measurement of in-band powers of EUV sources from different potential suppliers, so that a reliable comparison between those sources could be made. The name of the tool was derived from the fact that it was actually transported to various locations for measurements. In the FC tool, (EUV) radiation is wavelength filtered by reflection off a multilayer mirror and transmission through a thin foil filter, before it is collected on a photodiode. As the mirror, filter and photodiode are all well-characterized, the photodiode signal can be related to the absolute power emitted by a source at a certain wavelength.

Section 6.3 describes the wavelength-dependent sensitivity calibration of an EUV spectrometer using a tool similar to the Flying Circus mentioned above, in which multilayer mirrors optimized for different wavelengths were placed. A wavelength dependent sensitivity correction has been applied to the experimental EUV spectra used in the same chapter for comparison with modeling results.

3.5 Line broadening

Apart from intensities integrated over a spectral line, also linewidths and shapes can give information about the plasma. Again, such measurements are more straightforward in the visible wavelength range than in the EUV, since for the latter, one needs (by comparison) a very large, high accuracy spectrometer to resolve the shapes of individual lines.

Spectral lines can be broadened or even split up into separate contributions by a number of different mechanisms. These include natural broadening (due to the finite lifetimes of the energy levels), pressure broadening (due to collisions with neutral atoms) and broadening and level splitting due to the presence of macroscopic electric and magnetic fields. However, in plasmas with sufficiently high degree of ionization, Doppler broadening and Stark broadening are usually the only significant contributors to the shape of a spectral line. Doppler broadening is caused by Doppler shifts due to the velocities of the individual atoms or ions that emit the photons, and results in a Gaussian spectral shape if the velocity distribution is Maxwellian.
3.6 Schlieren photography and laser interferometry

Stark broadening on the other hand, which is due to collisions of the radiating atom or ion with charged particles, leads to a Lorentzian spectral profile. The combination of both broadening mechanisms leads to a so-called Voigt profile. In principle, if a sufficiently high resolution spectral profile has been measured, the overall line broadening can be separated into the Lorentzian and Gaussian parts. Next, from the width of the Gaussian (Doppler) part, the gas temperature can be derived.

The interpretation of the Stark broadening part in terms of plasma parameters is somewhat less straightforward. Stark broadening for non-hydrogenic lines is determined for the most part by electron impact broadening, which leads to a linear dependency of the Stark width on electron density [3]. Apart from this, there is also a (generally much weaker) dependency on electron temperature. The electron density in a plasma can be derived from a Stark width by looking up the Stark broadening parameter of the spectral line in the literature for a plasma of similar (but not necessarily exactly the same) temperature. Experimental data are available for large sets of neutral atoms and low ionic stages and they have been compiled into databases and review articles, e.g., Refs. [4] (and several supplements to this work), [5], [6], and several references in Ref. [3]. Still, experimental data cannot be available for all possible optical transitions and temperatures. For a given optical transition, an experimental Stark broadening parameter is often known only for a single temperature.

An alternative is to use theoretical calculations of Stark widths. Full quantum mechanical calculations can be performed—see, e.g., Ref. [3] for an overview—but more simple, approximating formulas are also available [7]; see also the discussions in Secs. 6.2.4 and 7.2.1. To a certain extent, the theoretical temperature dependencies of Stark widths can be used to extrapolate the available experimental data to plasma conditions for which the Stark broadening parameters are not yet known.

Measurements of Stark broadening of certain spectral lines of Sn$^{+}$ and Sn$^{2+}$ ions are discussed in Chapter 7. For these lines in the tin vapor discharge plasma, it was found that all other broadening mechanisms are negligible, except for a possible contribution of the macroscopic magnetic field in the prepinch phase. Also, it was derived from theory that the temperature dependency of the Stark widths was small in the range of interest. Since literature data on the broadening of the lines emitted by Sn$^{2+}$ were not available, yet another method was applied to determine their Stark broadening parameters, namely a cross-calibration with the width of a Sn$^{+}$ line recorded simultaneously. The newly obtained Stark broadening parameters have been validated by comparison with Thomson scattering data.

3.6 Schlieren photography and laser interferometry

In the field of active diagnostic techniques, Schlieren photography and laser interferometry are two types that are quite frequently applied to both laser produced and discharge EUV
Chapter 3: Optical diagnostics

Fig. 3.2. The imaging principle of Schlieren photography. The laser beam is focused by a lens onto the edge of a knife (solid lines), while the same lens produces a sharp image of the plasma on the detector (dashed lines). Local electron density gradients in the plasma cause the laser focus to end up higher or lower relative to the knife, and thereby create laser intensity variations in the image on the screen.

producing plasmas. Both are based on measuring the refractive index of the plasma at a certain (laser) wavelength, and therefore they are treated together in this section.

In the case of Schlieren photography, a laser beam is sent through a plasma setup, and then focused by a lens onto the edge of a “knife”, so that—in the case that the plasma is absent—just half of the laser intensity is blocked by the knife and the other half gets recorded by a detector (film or camera). The position of the detector is chosen such that the lens produces a sharp image of the plasma on the detector; see Fig. 3.2. Now, electron density gradients in the plasma are the cause of gradients in the refractive index in the plasma, which can lead to refraction of the laser light. Those gradients that are perpendicular to the knife edge will cause the light to be blocked by the knife to a smaller or larger extent, and intensity variations on the detector will be the result. This way, the intensity of the recorded light at a certain position on the detector is a measure for the electron density gradient in one direction, integrated over a line of sight through the plasma.

While Schlieren photography is based on refractive bending in the plasma, laser interferometry is based on phase shifts caused by the plasma refractive index. In laser interferometry, the laser beam is split into two parts by a beam splitter, and one part is sent through the plasma; then, this part of the laser beam is made to interfere with the other part, which bypasses the plasma. Now, the integrated refractive index along the laser beam in the plasma causes an optical path length difference, and hence a phase difference that leads to either constructive or destructive interference. Again, the plasma is imaged onto a detector by means of a lens. Line-of-sight-integrated electron densities can be derived from the image by “counting fringes”.

Both methods suffer from certain rather severe limitations. First of all, only line-of-sight measurements are possible, so that fully spatially resolved determination of electron densities is not possible; rather, assumptions on the length and homogeneity of the plasma are needed to interpret the results. Further, both methods can only be used if the electron density gradients are not too large; in the case of Schlieren photography the measured intensity can only go down to zero or up to 100% of the laser beam intensity. In the case of
laser interferometry, fringes cannot be counted if their mutual distances are smaller than the imaging resolution of the system, or if they are smeared too much due to evolution of the plasma within the duration of the laser pulse. As the difference of the plasma refractive index from unity is, in general, proportional to the square of the probing wavelength, a reduction of the laser wavelength can help to limit the number of fringes in the image. High density plasmas can be probed using (extreme) ultraviolet laser beams. For example, a 15.5 nm laser was applied to measure electron densities above $10^{27} \text{ m}^{-3}$ \cite{8}; and a 46.9 nm laser was used to probe a pinched discharge plasma \cite{9}. On the other hand, interferometry at small wavelengths is not very sensitive to low electron densities, as it is not straightforward to determine a small fraction of a fringe.

With Schlieren photography, there is the additional difficulty that absorption of the laser light in the plasma can influence the result. Often, only qualitative conclusions are drawn from Schlieren photography results.

Advantages of both methods compared to Thomson scattering (discussed in the next section) are that they are experimentally less complex, and they are more easily applicable to higher plasma densities—within the limitations caused by the gradients and laser absorption discussed above. The main disadvantage of both methods is the smaller amount of obtained information: they do not provide completely spatially resolved measurements, or any information on electron temperature.

In the field of EUV generating plasmas, a laser system (at 337.1 nm) for simultaneous Schlieren photography and laser interferometry of Z-pinch plasmas was built by Kalantar et al. \cite{10}. Fornaciari and co-workers \cite{11} used laser interferometry at 532 nm for probing a capillary discharge intended for EUV generation. They used a prescribed shape of the electron density distribution to fit their experimental results. Both laser interferometry and Schlieren photography have further been applied to an EUV producing Z-pinch plasma by Katsuki et al. at Kumamoto University, Japan \cite{12, 13}. Neither of both methods has been applied in the experiments described in the present work.

### 3.7 Thomson scattering

An important tool for the characterization of a wide variety of laboratory and industrial plasmas is Thomson scattering (TS) spectroscopy. This technique, in most standard applications, is based on the scattering of laser photons off individual electrons in a plasma. In general, the parameters that can be derived from the scattered spectra are local electron temperatures and densities, $T_e$ and $n_e$. The light scattered from a focused laser beam can be imaged onto the entrance slit of a spectrograph; by translation of the plasma relative to the laser focus, fully three-dimensional spatially resolved data can in principle be obtained.

In this section, it is investigated to what extent Thomson scattering can be applied to gain knowledge on these parameters in the pulsed plasmas that are used for the generation of EUV radiation. To this end, the theoretical shape of the spectrum is studied.
tive behavior of the electrons in EUV plasmas is shown to play an important role; the consequences of the collective effects are discussed in detail.

Within the framework of this thesis, collective Thomson scattering experiments have been performed on the tin vapor discharge. These experiments and their results are discussed in Chapters 8 and 10. The design of a new setup for Thomson scattering, based on some of the considerations in this section, is presented in Chapter 9.

### 3.7.1 Theoretical spectrum; the Salpeter approximation

Noncollective (or incoherent) Thomson scattering is based on the scattering of photons on individual electrons in a plasma. When a photon is scattered off an electron, the induced photon frequency shift is proportional to the velocity of the electron in the direction of the scattering vector. When the electron energies have a Maxwellian distribution, the velocity distribution will be Gaussian. As long as the scattered photons from different electrons are not phase correlated, the spectrum of scattered photons will also have a Gaussian shape, and the temperature of the electrons can be derived by fitting the experimentally observed spectrum to a Gaussian profile. By calibration of the absolute value of the radiated power, the electron density in the plasma can be derived.

The assumption of noncollective scattering fails when electrons that can sense each other’s presence in the plasma, undergo phase-correlated motion in the laser field. A measure for the distance over which screening of electron charges takes place, is the Debye length,

$$\lambda_D = \sqrt{\frac{\varepsilon_0 k_B T_e}{e^2 n_e}}.$$  \hspace{1cm} (3.3)

Scattering is noncollective if electrons within this distance experience random phases of the laser light as observed from a certain scattering direction. This is the case when $k\lambda_D \gg 1$. Here, $k = 4\pi \sin(\phi/2)/\lambda$ is the scattering wavevector, in which $\phi$ is the angle between the incident laser beam and the scattering direction ($\phi = 0, \pi$ correspond to the limits of fully forward and backward scattering, respectively). For $k\lambda_D \lesssim 1$, collective effects play an important role. Hence, the scattering parameter $\alpha = 1/k\lambda_D$ determines the nature of the scattering profile.

Under certain approximations (collisionless plasma, and a not too large ratio $T_e/T_i$), the frequency dependence of (partially) collective TS light is given by the Salpeter approximation [14, 15]. The scattered power per solid angle, as a function of frequency, is given by

$$\frac{dP_s(\omega)}{d\Omega} = \frac{d\sigma}{d\Omega} P_L L_e n_e S(k, \omega),$$  \hspace{1cm} (3.4)

with $P_s$ the scattered power, $P_L$ the incident laser power, $L_e$ the length over which scattered
3.7 Thomson scattering

Light is collected, and $\sigma$ the single-electron scattering cross section. In this equation, the frequency dependence is enclosed in the so-called form factor, $S(k, \omega)$, which is given by

$$S(k, \omega) = \left| \frac{1}{1 + \alpha^2 W(x_e)} \right|^2 \frac{\exp(-x_e^2)}{\pi^{1/2}} dx_e + Z \left( \frac{\alpha^2}{1 + \alpha^2} \right)^2 \left| \frac{1}{1 + \beta^2 W(x_i)} \right|^2 \frac{\exp(-x_i^2)}{\pi^{1/2}} dx_i$$

$$= \pi^{-1/2} \Gamma_\alpha(x_e) dx_e + Z \left( \frac{\alpha^2}{1 + \alpha^2} \right)^2 \pi^{-1/2} \Gamma_\beta(x_i) dx_i,$$  \hspace{1cm} (3.5a)

where

$$\beta^2 = Z \left( \frac{\alpha^2}{1 + \alpha^2} \right) \frac{T_e}{T_i},$$  \hspace{1cm} (3.5b)

$$\Gamma_\zeta(x) = \frac{\exp(-x^2)}{|1 + \zeta^2 W(x)|^2} \left( \zeta = \alpha, \beta \right),$$  \hspace{1cm} (3.5c)

and

$$W(x) = 1 - 2x \exp(-x^2) \int_0^x \exp(p^2)dp - i\pi^{1/2}x \exp(-x^2).$$  \hspace{1cm} (3.5d)

Here, $x_e = \omega/(kv_e)$ and $x_i = \omega/(kv_i)$. $\Gamma$ is called the Salpeter function, $W$ the plasma dispersion function. Further, $v_e$ and $v_i$ are the electron and ion mean thermal speeds, respectively, defined as

$$v_e = \left( \frac{2k_B T_e}{m_e} \right)^{1/2}, \quad v_i = \left( \frac{2k_B T_i}{m_i} \right)^{1/2}.$$  \hspace{1cm} (3.6)

The right-hand side of Eq. (3.5a) consists of two terms; these are named $S_e(k, \omega)$ and $S_i(k, \omega)$. In the limit of strongly collective scattering, they correspond to scattering from longitudinal plasma waves of electron- and ion-acoustic nature, respectively. In principle, both terms can be used to derive information about the plasma. Since typically $v_e \gg v_i$, the electron contribution will always be much wider than the ion contribution, so that the two are almost completely separated in the frequency or wavelength domain. The two terms are treated separately in the remainder of this section.

3.7.2 The electron term

In principle, the electron term can be used to derive the electron temperature and density, $T_e$ and $n_e$, simultaneously. However, a distinction should be made between the cases of moderately (or partially) and strongly collective scattering.

For $\alpha$ being not too large, the shape of the contribution can be fitted to the first term of Eq. (3.5a) to find simultaneously $\alpha$ and $T_e$ from the actual shape and the width of the spectrum, respectively. Eq. (3.6), in combination with the definition of $x_e$, shows that for given $\alpha$ the width of the spectrum is proportional to the square root of $T_e$. In a practical
Fig. 3.3. The electron contribution to the form factor in the collective Thomson scattering spectrum for values of $n_e$ between $10^{21}$ and $10^{25}$ m$^{-3}$, with $T_e = 35$ eV, $\lambda = 532$ nm and scattering angle $\phi = \pi/2$. On the horizontal scale, the shift of the scattered light from the incident laser light is given both in frequency and in wavelength units.

implementation, the fitting procedure can be programmed to accept $n_e$ and $T_e$ as input parameters, and to calculate $\alpha$ from these two.

Examples of the electron contribution are plotted in Fig. 3.3 for an electron temperature $T_e = 35$ eV, incident laser wavelength 532 nm and scattering angle $\pi/2$ (which corresponds to a perpendicular scattering geometry); the electron density is varied from $10^{21}$ to $10^{25}$ m$^{-3}$ in steps of a factor 10. At $n_e = 10^{21}$ m$^{-3}$ and $10^{22}$ m$^{-3}$, the distribution is still (nearly) Gaussian; however, at $n_e = 10^{23}$ m$^{-3}$ the distribution starts to deviate from this shape: the intensity in the center drops and increases slightly in the wings. At $10^{24}$ m$^{-3}$, already some rudimentary satellite peaks can be observed; and at $n_e = 10^{25}$ m$^{-3}$, finally, the central contribution has practically vanished, and only a sharp satellite peak remains at roughly the plasma frequency. This phenomenon is discussed in more detail below. The horizontal scale in Fig. 3.3 is plotted not only in frequency units but also in terms of wavelength shift, since this is the quantity that is more commonly used in the interpretation of experimental spectra.

From the discussion above, it follows that for moderately collective spectra, it is in principle possible to derive both electron density and temperature independently without an absolute calibration of the scattered intensity. However, such an absolute calibration, for example by recording Rayleigh or rotational Raman scattering in air or nitrogen gas at atmospheric pressure, will give additional information on the stability of the plasma and the reliability of the fitting results. The implementation of a fitting procedure, including absolute calibration of the signal, is discussed in Secs. 8.2 and 10.2 for the experiments of Chapters 8 and 10, respectively.

The intensity of the electron contribution to the form factor, integrated over $\omega$, is
3.7 Thomson scattering

The real and imaginary parts of $W(x)$ plotted against $x$. For large $x$, $\text{Re}(W(x))$ behaves as $-1/(2x^2)$.

$$S_e(k) = \frac{1}{1 + \alpha^2}.$$ \hspace{1cm} (3.7)

The total scattered power in the electron part is now given by

$$\frac{dP_{s,e}}{d\Omega} = \frac{d\sigma}{d\Omega} P_L L_a n_e S_e(k) = \frac{d\sigma}{d\Omega} P_L L_a n_e \frac{1}{1 + \alpha^2}$$ \hspace{1cm} (3.8)

In this equation, the dependency on the plasma parameters is fully enclosed in the factor $n_e/(1 + \alpha^2)$.

### 3.7.3 Strongly collective limit

As mentioned above, for large $\alpha$, a strong, sharp peak appears in the spectrum. This happens at values of $x_e$ (or $\omega$) for which the denominator of $\Gamma_\alpha$ approaches zero. For $\alpha$ sufficiently large, i.e., larger than about 3, the real part of $W(x)$ behaves roughly like $1/\sqrt{2}x^{-2}$ (see Fig. 3.4) while $\text{Im}(W(x))$ is very small. Hence, a maximum in $\Gamma_\alpha$ is found near the value of $x$ where $1 + \alpha^2 \text{Re}(W(x_e)) = 0$,

$$1 - \frac{1}{2} \alpha^2 x_e^{-2} \approx 0,$$ \hspace{1cm} (3.9)

or, in terms of frequency shift,

$$\omega = kv_e \alpha/\sqrt{2} = \frac{v_e}{\sqrt{2\lambda_D}} = \left(\frac{k_B T_e}{m_e}\right)^{1/2} \left(\frac{\epsilon_0 k T_e}{\epsilon^2 n_e}\right)^{-1/2} = \sqrt{\frac{\epsilon^2 n_e}{\epsilon_0 m_e}} = \omega_p.$$ \hspace{1cm} (3.10)

In other words, two peaks appear at frequencies which are shifted from the laser frequency by the plasma frequency—which is just the frequency of the plasma waves from which the laser light is scattered. Hence, in this case, the total width of the spectrum is directly related to the electron density.
In this strongly collective limit, not much information is contained in the shape of the spectrum. In the case of a collisionless plasma, the full width at half maximum (FWHM) of the peak is approximately given by

$$\Delta x_e = \frac{1}{2} \sqrt{\pi \alpha^2 \exp\left(-\frac{\alpha^2}{2}\right)}$$  \hspace{1cm} (3.11)

for the range where \(\text{Re}(W(x_e))\) can be approximated as \(\frac{1}{2} x_e^{-2}\), or \(\alpha \gtrsim 3\). In terms of scattering from a plasma wave, this finite peak width is caused by the Landau damping of the wave. However, for the conditions that govern a pinched EUV discharge, at large \(\alpha\), the width of the peak will be determined by collisional damping of the wave rather than by Landau damping \cite{16}. In the pinch plasma, the electron-ion collision frequency for momentum transfer is larger than the electron-electron one (due to the fact that the ions are multiply charged), and hence it dominates the total damping. In the frequency domain, the peak width will be on the same order as this collision frequency, which is given by \cite{17}

\begin{align*}
\nu_{ei} &= n_e \frac{4\sqrt{2\pi}}{3} \left(\frac{e^2}{4\pi \epsilon_0 m_e}\right)^2 \left(\frac{m_e}{k_B T_e}\right)^{3/2} Z \ln \Lambda_i, \quad (3.12a) \\
\Lambda_i &= \frac{3k_B T_e 4\pi \epsilon_0}{Ze^2} \sqrt{\frac{\epsilon_0 k_B T_e}{e^2 n_e}}. \quad (3.12b)
\end{align*}

Here, \(\Lambda_i\) is the corresponding Coulomb logarithm. If we assume, for a pinched EUV discharge, a density of \(n_e = 2 \times 10^{25} \text{ m}^{-3}\), \(T_e = 35 \text{ eV}\), and \(\lambda = 532 \text{ nm}\), then we get \(\nu_{ei} = 1.2 \times 10^{13} \text{ s}^{-1}\). The corresponding linewidth is on the order of \(\Delta \lambda = 1.8 \text{ nm}\), which is much larger than the width \(\Delta \lambda \approx 7 \times 10^{-5} \text{ nm}\) that would result from Landau damping alone (with \(\alpha = 6.09\)).

In theory, from the collision frequency, as calculated from the peak width, the electron temperature could be derived, provided that the theoretical shape of the spectrum is carefully derived (such as, e.g., in Ref. \cite{18}). However, in practice, a finite volume is probed in finite time, which leads to corresponding variations in plasma parameters. Also, there will always be pulse to pulse variations in the plasma parameters. Therefore, the measured profile will be smeared beyond the width of the theoretical spectrum; the broadening due to electron density variations can be much larger than the theoretical width of the satellite.

Hence, determination of \(T_e\) can only be possible in practice by using the total scattered intensity. For \(\alpha \gg 1\), Eq.\((3.8)\) can be replaced by

\begin{align*}
\frac{dP_{se}}{d\Omega} &\approx \frac{d\sigma}{d\Omega} P_{L_e} L_n \frac{1}{\alpha^2} \\
&= \frac{d\sigma}{d\Omega} P_{L_e} L_n \left(\frac{4\pi \sin(\phi/2)}{\lambda}\right)^2 \frac{\epsilon_0 k_B T_e}{e^2}. \quad (3.13)
\end{align*}
so that as far as the plasma parameters are concerned, the total scattered power is just proportional to $T_e$.

In Fig. 3.5 for certain arbitrary experimental parameters, the dependency of total scattered power on $n_e$ is given for different temperatures $T_e$. It clearly shows the transition from a proportionality to $n_e$ only, for low $n_e/T_e$ ratios, to a proportionality to $T_e$ only for the strongly collective regime. Note that in the latter case, the roles of $n_e$ and $T_e$ are exactly reversed compared to the case of noncollective scattering, where $n_e$ determines the integrated intensity of the scattered light, and $T_e$ determines the width of the spectrum.

Note, finally, that the problems mentioned above—the gradients in the pinch plasma and pulse-to-pulse variations—can also make a reliable measurement of the absolute intensity of the spectrum difficult.

### 3.7.4 The ion-acoustic term

Apart from the electron term, also the ion-acoustic term in the form factor could be used to study EUV producing plasmas. Integration of the second term in Eq. (3.5a) over $\omega$ yields

$$S_i(k) = \frac{Z\alpha^4}{(1 + \alpha^2)[1 + \alpha^2(1 + ZT_e/T_i)]}. \tag{3.14}$$

Unlike the electron contribution, Eq. (3.14) shows that for $\alpha \to \infty$, the ion contribution does not go to zero, but tends to a constant value, $Z/(1 + ZT_e/T_i)$. In other words, even for very large $\alpha$, the power in the ion feature is a finite fraction of the scattered power for noncollective Thomson scattering. For $\alpha^2$, $Z \gg 1$ and $T_e \approx T_i$, such as expected in a fully pinched discharge plasma, $S_i(k) \approx 1$.

This property makes the ion-acoustic term suitable for measurements in the regime of
strongly collective Thomson scattering. To start with, the integrated intensity of the ion term provides an additional way to determine the electron density $n_e$ in the detection volume. Note, however, that the width of the ion contribution, given by (3.15), is very low. For $Z = 10$, $T_e = 35$ eV, $\alpha \gg 1$, and a 532 nm laser scattered over angle $\pi/2$, the wavelength difference between the incident light and the ion peaks is less than 0.05 nm. This makes the usual stray light filtering, as described in Sec. 9.3, impossible. Therefore, any experiment for determining the total power in the ion feature should be set up such that the recorded stray light level is very low.

Secondly, similar to Eq. (3.10), for sufficiently high $\beta$ [as defined in Eq. (3.5b)] a peak is found in the ion contribution at

$$\Delta \omega/k = v_i \beta / \sqrt{2} = \sqrt{\frac{2 k_B T_i}{m_i}} \sqrt{\frac{1}{2} Z^{\left(\frac{\alpha^2}{1+\alpha^2}\right)} \frac{T_e}{T_i}} = \sqrt{\frac{Z k_B T_e}{m_i}} \sqrt{\frac{\alpha^2}{1+\alpha^2}} = c_s,$$ (3.15)

where $c_s$ is the ion-acoustic sound speed. With a high resolution spectrometer, the width and shape of the ion contribution could be determined, and the product $ZT_e$ could be derived. Assuming that $Z$ is already known with sufficient accuracy from EUV spectrometry, $T_e$ can be determined in this way. Simulated examples of the ion-acoustic wave term are shown in Fig. 3.6. Note that for these curves, neither collisions nor strong coupling effects were taken into account, which can both be significant for the relevant plasma parameters—however, these effects are beyond the scope of the present discussion. See, e.g., Ref. [19] for a detailed discussion of the effect of strong coupling on the shape of the ion-acoustic wave term.

Ion-acoustic Thomson scattering has been used for the probing of laser produced plasmas [20–22] and a gas-liner pinch [23, 24]. Neither of these were designed as EUV sources.
most of these works, the ion temperature was higher than our values, and the atomic mass of the working element was (much) lower than that of xenon or tin, which helps in obtaining a broader, more easily resolvable ion-acoustic term than in the case given above.

### 3.7.5 Experimental limitations

This section is concluded with a discussion of two types of difficulties that can limit the reliable, independent determination of \( n_e \) and \( T_e \) from Thomson scattering, especially in the case of relatively high density plasmas.

Firstly, in active spectroscopic techniques, a primary concern is always the disturbance of the plasma by absorption of laser light. In the context of EUV producing plasmas, two main absorption processes can be recognized; these are inverse bremsstrahlung and absorption in line transitions. The effects of both processes on the (measured) plasma parameters, for the conditions relevant to the tin vapor discharge, are discussed in detail in Chapters 8 and 10. Nonlinear processes (such as multiphoton ionization) have been found to be negligible for the moderate laser powers as used in this work.

An additional theoretical problem that arises from laser absorption is a systematic error in the absolute intensity calibration, as the actual laser intensity in the plasma is decreased compared to the intensity in the calibration experiment. However, this error will only start to play a role when the plasma is already strongly disturbed, and hence does not play any role in practice.

As the second main difficulty, as explained in the previous section, the total power in the electron contribution tends to a certain limit when the plasma density is increased with constant temperature. At the same time, the background radiation emitted by the plasma will increase further with increasing density. Hence, the electron contribution will have to compete increasingly with fluctuations and statistical noise in the background. In fact, in the experiments of Chapter 8 this phenomenon was found to make measurements possible for only one specific part of the discharge evolution. This problem can be solved by measures that increase the signal to background ratio; these include a narrowing of the laser focus (in combination with a smaller plasma volume being recorded by the spectrograph), and—more importantly—application of shorter laser pulses. These considerations led to the design and construction of a new setup for Thomson scattering. This setup is described in detail in Chapter 9 and has been used for Thomson scattering experiments on the remaining phases of the tin vapor discharge as described in Chapter 10.

The problem of background radiation could in principle also be solved by application of a laser at lower wavelength, e.g., 248 nm (krypton fluoride excimer) or 266 nm (fourth harmonic Nd:YAG). In the example of Sec. 3.7.3 the total intensity in the electron part of the spectrum would be about 4.2 times larger at 248 nm than at 532 nm (for perpendicular scattering). Further, the total width of the spectrum would be only about 22 nm instead of 78 nm. On the other hand, the plasma emission near 248 nm is not accurately known, but it is reasonable to assume a \( 1/\lambda^2 \) dependence of the background radiation intensity per
Chapter 3: Optical diagnostics

wavelength (see, e.g., Sec. 6.6). Overall, an improvement of the signal to background ratio of roughly a factor 3 could be expected. An additional benefit of the lower wavelength would be a less collective spectrum—for the same example, we get $\alpha = 2.84$ at $\lambda = 248$ nm instead of $\alpha = 6.09$ at $\lambda = 532$ nm—so that more information could be derived from the shape of the spectrum.

However, operation of TS experiments at wavelengths in the ultraviolet (UV) range is made unpractical by experimental difficulties. Firstly, the laser wavelength is invisible to the human eye, which would make the system much more difficult to align. Further, vacuum windows, lenses, gratings, and ICCD cameras are less readily available for the UV range than they are for the visible wavelength range. And finally, nonlinear absorption processes are more likely to occur at shorter laser wavelengths.

References


Chapter 4

Time resolved pinhole camera imaging and extreme ultraviolet spectrometry on a hollow cathode discharge in xenon

Abstract

A pinhole camera, an extreme ultraviolet (EUV) spectrometer, a fast gatable multichannel plate EUV detector, and a digital camera have been installed on the ASML EUV laboratory setup to perform time-resolved pinhole imaging and EUV spectroscopy on a copy of the Philips EUV hollow cathode discharge plasma source. The main properties of the setup have been characterized. Time-resolved measurements within the plasma pulse in the EUV have been performed on this source. Specific features of the plasma, such as a ring shape in the initiation phase and a propagating sphere during the pinch phase, have either been discovered or confirmed experimentally. Relative populations of various ionization stages in the pinch plasma have been estimated on the basis of line intensities and calculated transition probabilities. The changes in relative line intensities of a single ionization stage can be explained by a combination of temperature and excitation/deexcitation balance effects. Experiments with argon dilution on a newer version of the source show considerable effect on the shape of the xenon EUV spectrum.

Chapter 4: Time-resolved imaging and EUV spectrometry on a HC discharge

4.1 Introduction

Background

Various concepts of discharge plasmas are currently under development by a number of groups worldwide as candidate sources of extreme ultraviolet (EUV) radiation for application in future lithography tools (for a recent overview, see, e.g., Refs. [1, 2]). Common properties of these plasmas are that they have small spatial dimensions (typically less than 1 mm) and occur in short pulses (on the order of 100 ns–1 µs duration) rather than as steady-state discharges.

To gain insight in the characteristics of such discharges, it is necessary to develop and apply diagnostics that not only show the required spatial resolving power but also operate on timescales shorter than the typical pulse duration.

Basic description of the Philips source

In the ASML EUV laboratory in Veldhoven, The Netherlands, an early state-of-development version copy of the Philips EUV hollow cathode discharge plasma source was installed in an experimental setup in the end of 2001. The concept of this EUV source was described first in Refs. [3–5]. See Fig. 4.1 for a schematic representation of the source. Initially, the total capacitance of the electrical circuit for this specific source was 48 nF. The source was operated with voltages up to 15 kV. This version was used as a source of photons for testing purposes (especially of mirror lifetime during exposure to the plasma) and as a study object for the initial characterization of the pinch plasma.

The source received intermediate updates from the supplier, including an adjustment of the electrode geometry and a change in the electrical parameters towards a higher capacitance of 674 nF and lower working voltage of 3 kV at maximum. All experiments described here, except for those concerning dilution of the working gas with argon, have been performed on the “basic” version of the source.
4.2 Experimental

This work
In the framework of a joint research project between ASML and TU/e, involving both theoretical and modeling efforts and experimental investigations, fast EUV imaging and fast spectroscopy on this source have been combined to further the knowledge on the dynamics of the EUV plasma pulse. The combination of time-resolved EUV imaging and spectroscopy has led to better understanding of the dynamics of the pinch plasma in the Philips EUV source. Owing to good reproducibility in timing of both the plasma pulse and the trigger signal, a time resolution of 10 ns or better could be achieved while integrating light from several pulses, making it possible to resolve the different phases of the plasma during the discharge pulse. Abel inversion was applied to space-resolved EUV spectrometry to plot the EUV line intensities during a ring-shaped phase of the discharge. Calculations based on the COWAN computer code [6] were used to derive rough estimates of the relative populations of different ionization stages of xenon in the plasma.

4.2 Experimental

For typical experiments as described here, the source was operated in a repetitive mode at 100 Hz repetition frequency. The dead time of the high-voltage supply for the multichannel plate (MCP) camera limited the recordings of pinhole images or EUV spectra to just one pulse per second. Several pulses (typically 10 or 30) were integrated for each image to gather sufficient intensity and reduce the effect of short time scale variations in the plasma characteristics.

4.2.1 Pinhole camera

A pinhole camera, consisting of a pinhole, a filter, an MCP image intensifier from the ISAN institute in Troitsk, Russia [7], and a commercial-type digital camera, was applied to record time-resolved images of the discharge plasma in the EUV wavelength range. For low power applications, the system provides a simple and cost-efficient imaging alternative to mirror projection systems. Images were recorded with the camera in two different positions: one on the axis of symmetry of the source and one at an angle of 22.5° with respect to that axis. A schematic representation of the setup is shown in Fig. 4.2.

Just behind a (electrical spark or mechanically drilled) 50 or 100 µm diameter pinhole, a 150-nm-thick niobium/silicon filter was positioned. It has a transparency of about 50% to EUV light with a wavelength between roughly 10 and 17 nm, but it blocks over 99% of any vacuum ultraviolet (VUV) light emitted by the source at larger wavelengths. The supporting mesh of the filter has 0.5 mm spacing and is visible in some of the pinhole images. The pinhole image is projected onto the surface of an MCP with phosphorus screen, which intensifies the signal and converts the EUV radiation into visible light. The MCP was gated by a fast 6.25 kV high voltage pulse over the plate and the adjacent gap to the phosphorus screen. The typical duration of the pulse was 10 ns. However, variation
of the pulse duration showed that the effective exposure time was only 5 ns, a fact that is probably caused by capacitive properties of the current circuit. The phosphorus screen also served as a vacuum-to-air interface, and the intensified image was finally recorded by a high-sensitivity commercial digital camera.

Depending on the distance between the plasma and the pinhole, and pinhole diameter, the spatial resolution in the plasma that could be achieved was on the order of 70 µm. The two main contributors to this resolution are a geometrical contribution and diffraction from the pinhole at the EUV wavelengths. Serious blurring of the image due to diffraction at larger wavelengths was prevented by the niobium/silicon filter and the drop of sensitivity of the MCP above 100 nm [8].

The distance from the pinhole to the MCP surface was fixed at 770 mm; the distance from the source to the pinhole was 140 mm for the off-axis position and 130 mm for the on-axis position, resulting in magnification factors from source to MCP of 5.5 and 5.9, respectively.

4.2.2 EUV/VUV spectrometer

For the EUV spectrometer, partly the same arrangement was used as for the pinhole camera. A 100 µm slit was mounted at the position of the MCP camera. A 1200 l/mm grazing-incidence reflective grating was used as the dispersing element to record the EUV spectrum from roughly 9 to 18 nm. A different grating with 300 l/mm could be fitted to cover the VUV range from roughly 30 to 90 nm. However, since the higher ionization stages of xenon (7+ and above) all radiate in the EUV, and interpretation of the EUV part of the spectrum turned out to be relatively simple, only spectra recorded with the 1200 l/mm grating are discussed in the present work.

A valve with a bypass tube was used to prevent degradation of the optical elements during operation of the source without using the spectrometer, while keeping the spectrometer at vacuum pressure. Again, the MCP and digital camera were used for intensification, con-
version, and recording of the spectrum. The MCP was mounted in an off-Rowland circle configuration for simplification of alignment and mounting.

Both gratings of the spectrometer have 1000 mm radius of curvature. The grazing angle of incidence is $4^\circ$ and the center of the MCP is located at a grazing angle of $10^\circ$ from the grating. The slit is on the Rowland circle of the grating. The MCP is mounted onto a holder with adjustable distance to the grating, so that the wavelength for which it is “focused” onto the Rowland circle can be adjusted. A spectral resolution of $\Delta \lambda = 0.1$ nm or better can be achieved with this setup.

The background pressure of xenon gas along the path between the pinhole optics and the MCP was kept below 0.1 Pa to minimize reabsorption of the EUV radiation. For this pressure, the absorption in Xe in the EUV range over 1 m path length is 6% at most. A 3000 l/s turbo pump was used to pump off the xenon flow of about 8 sccm (cubic centimeter per minute at standard temperature and pressure) that was required to run the discharge. In the experiments with argon dilution, an argon gas buffer was applied in combination with a downstream flow restriction (not obstructing the view to the pinhole camera or EUV spectrometer). In this case, the total gas flow was much larger; however, due to the much lower absorption cross section for argon, the contribution of argon to the absorption of EUV radiation is negligible.

To obtain spatially resolved spectra from the on-axis position, an additional slit was inserted at the pinhole position and orientated perpendicular to the spectrometer entrance slit. The spatial resolution of the spectra is comparable to that of the pinhole images. Since the source electrodes have cylindrical symmetry, Abel inversion has been applied to obtain line intensities in the plasma as a function of the distance to the symmetry axis.

An example of the EUV spectrum of xenon in this plasma, with classification of its main features, is shown in Fig. 4.3. The identifications are supported by the literature and/or calculations based on the COWAN computer code. Some difficulty arises with the lines around and below 11 nm, since many lines from stage XII and higher tend to overlap here. The contribution from stage XIII was derived from a distinctly different behavior of the intensity at 10.8 nm compared to that of larger wavelengths, during variation of the plasma parameters. Since the contribution had to come from one or more stages higher than XII, and stages XIV and higher are unlikely to be populated under the given plasma settings, it was ascribed to Xe XIII.

For evidence of the presence of different ionization stages at a certain place or time in the plasma, the $4d$-$5p$ transition features have been used wherever possible, because of the fact that they are well separated for the different stages. However, in the initiation phase of the discharge, the intensities of these features are very low, and the stronger $4d$-$4f$ transitions had to be applied for identification of the stages. For an overview of the EUV lines used for identification of the ionization stages, see Table 4.1.
Fig. 4.3. Sample EUV spectrum recorded using a 1200 l/mm grazing-incidence grating spectrograph with MCP.

Table 4.1. An overview of the EUV lines in the spectrum of the discharge used for ion identification. The listed intensities refer to typical maximum measured intensities of the line during the discharge, and serve only for relative comparison within one part of the spectrum. For Xe X, the weighted transition probability (gA) values are those of the strongest transition at the given wavelength.

<table>
<thead>
<tr>
<th>$\lambda_{\text{obs}}$ (nm)</th>
<th>Stage</th>
<th>Lower level</th>
<th>Upper level</th>
<th>$gA$ ($10^9$ s$^{-1}$)</th>
<th>$I$ (counts)</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>17.77</td>
<td>VIII</td>
<td>$4d^{10}5s$</td>
<td>$4d^85s5p$</td>
<td>2000</td>
<td></td>
<td>[10]</td>
</tr>
<tr>
<td>12.01</td>
<td>IX</td>
<td>$4d^{10}1^1S_0$</td>
<td>$4d^94f \ ^1P_1$</td>
<td>4554</td>
<td>6500</td>
<td>[9]</td>
</tr>
<tr>
<td>16.53</td>
<td>IX</td>
<td>$4d^{10}1^1S_0$</td>
<td>$4d^95p \ ^1P_1$</td>
<td>241</td>
<td>6000</td>
<td>[9]</td>
</tr>
<tr>
<td>11.49</td>
<td>X</td>
<td>$4d^9$</td>
<td>$4d^84f$</td>
<td>14400</td>
<td>5500</td>
<td>[9]</td>
</tr>
<tr>
<td>14.80</td>
<td>X</td>
<td>$4d^9$</td>
<td>$4d^85p$</td>
<td>321</td>
<td>5500</td>
<td>[9]</td>
</tr>
<tr>
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<td>XI</td>
<td>$4d^8$</td>
<td>$4d^74f$</td>
<td>5000</td>
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<td>[11]</td>
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<tr>
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<td>3500</td>
<td></td>
<td>[6, 11]</td>
</tr>
<tr>
<td>10.96</td>
<td>XII</td>
<td>$4d^7$</td>
<td>$4d^64p$</td>
<td>2500</td>
<td></td>
<td>[11]</td>
</tr>
<tr>
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<td>$4d^7$</td>
<td>$4d^65p$</td>
<td>3500</td>
<td></td>
<td>[6]</td>
</tr>
<tr>
<td>10.80</td>
<td>XIII</td>
<td>$4d^6$</td>
<td>$4d^54f$</td>
<td></td>
<td></td>
<td>[6]</td>
</tr>
</tbody>
</table>
4.2 Experimental

4.2.3 Triggering and other metrology

As stated above, fast and reproducible gating of the imaging system was achieved by applying short high-voltage pulses to the MCP camera. Given the unavoidable delays caused by the application of the high-voltage pulse generator, a Stanford DG535 delay generator, and some lengths of coaxial cables, a sufficiently strong and low-jitter trigger signal was needed to be available at least 160 ns before the onset of any EUV radiation from the discharge. Since the source operated in a “self-breakdown” mode, no such signal was available from the control electronics of the source.

However, it is a common characteristic of hollow cathode discharges, known both from experiments and modeling (see, e.g., Refs. [12, 13], and the discussion in Sec. 2.4), that they emit an electron beam before the discharge in the main electrode gap. Such an electron beam produces a negative voltage on a Faraday-cup-type collector that is placed close to the axis behind the anode hole. A sufficiently strong signal with a jitter of no more than \( \sim 2 \) ns could be picked up on-axis at least 300 ns before the main discharge. In the present setup, the cup was placed at a slightly off-axis position to allow space for the pinhole camera. Still, a sufficiently strong signal was picked up in time for recording even the earliest phases of EUV emission in the discharge.

The same Faraday cup signal can also be used to detect the arrival of ions from the discharge plasma expansion. From the time delays between the plasma pinch and the arrival at the Faraday cup, the ion velocity distribution can be derived.

A copy of the so-called “Flying Circus” tool [14], consisting of a set of photodiodes behind near-normal incidence multilayer mirrors, was used to measure the absolute in-band EUV output of the source to have a simple measure of the proper operation of the source, and its stability.

4.2.4 Derivation of relative ionization stage populations

Relative population densities of the different xenon ionization stages have been estimated by comparison of the measured EUV spectra with theoretical calculations of the positions and transition probabilities of xenon lines in the EUV. The theoretical data were provided by calculations using the COWAN computer code [6]. The spectral resolution of the spectrometer was about 0.1 nm; therefore, lines within a range of 0.1 nm in total were taken to contribute to the measured intensity at a certain wavelength.

Furthermore, local thermodynamic equilibrium (LTE) and a fixed electron temperature \( T_e = 35 \) eV have been assumed to relate line intensities to the ground state population densities. In short, the relative population densities were calculated as

\[
n_Z = \frac{n'_Z}{\sum_i n'_i}, \tag{4.1a}
\]

with

\[
n_Z = \frac{n'_Z}{\sum_i n'_i}, \tag{4.1a}
\]
\[ n'_Z = \frac{I_{\text{meas}}}{\sum g_n A_{nm}} \exp \left( -\frac{\Delta E}{kT_e} \right), \] (4.1b)

where the first summation is over all considered ionization stages and is carried out for normalization purposes. The second summation goes over the relevant transition lines from the computer code output as discussed above.

The assumption of LTE is necessary for this derivation. However, since all the applied lines are related to similar transitions and lie relatively close together in terms of wavelength and energy levels, it is likely that any deviations from LTE will affect the intensities of the different lines in similar ways. Therefore, these calculated relative population densities may also serve as rough estimates of the densities in the non-LTE conditions that are encountered in different phases of the discharge.

## 4.3 Results

Inspection of the combined results of pinhole imaging and EUV spectroscopy for the basic version of the source has led to the qualitative identification of three different phases in the main discharge, being an initiation of the pinch with a “ring-shaped” plasma, the pinch itself, and a decay phase with a propagating spherelike plasma “bulb.” Quantitative results for each of the phases could subsequently be obtained. Details of the measurement results for each of the phases are described below.

### 4.3.1 “Ring” shape before the pinch

The first evidence of a ring-shaped plasma before the main pinch was provided by on-axis pinhole images. These are presented in a matrix form in Fig. [4.4]. Each image corresponds to an area of 2.4×1.3 mm² in the source. The exposed area of each image is limited by the edge of the MCP zone (to the bottom) and a grid line of the filter support (to the top right).

The images are shown in chronological order during the plasma pulse, with time increasing from left to right and from top to bottom. The time step between consecutive images was 5 ns for the first 12 pictures and 10 ns for the remaining ones. Since the source was operated in self-breakdown mode, the definition of an absolute “zero” on the time scale is not straightforward. If the time of maximum pinching—that is, minimal effective radius of the EUV emission—is taken as the zero (between the seventh and eighth image in the series), then the time range between −35 and +85 ns is covered.

The source was operated at a discharge voltage of 13.9 kV, resulting in a pulse energy of 4.6 J. A 50 µm pinhole was used and for every image, 30 exposures of (effectively) 5 ns each were integrated, and a dark image from the charge-coupled device was subtracted. The first four pictures and the last one were finally intensified by a factor of 3.2 with respect
Fig. 4.4. Pinhole images of the discharge viewed from an on-axis position. First 12 images were taken with 5-ns time steps; for the remaining images time steps of 10 ns were applied. The first four and the last images were digitally intensified 3.2 times compared to the other ones.
to the other ones to make the weakest features better visible. Darkest regions correspond to the highest EUV intensities.

The ring phase can be recognized in the second to fourth images, the pinch itself in the subsequent four, and the plasma decay in the remaining images. Note that the limits between the phases, as defined here, are somewhat arbitrary.

From these images it can be derived that the initial phase is relatively short (only about 15 ns) compared to the total duration of the discharge (over 100 ns). The radiation intensity from this phase of the discharge is weak compared to the pinch radiation. From the images, the inner diameter of the ring at the time of its first appearance in the EUV can be derived to be 1.2 mm; the half-maximum thickness of the ring is about 0.5 mm. The maximum compression velocity is evaluated to be $8 \times 10^4 \text{ m s}^{-1}$.

To identify the EUV lines, and hence the ionization stages, that are responsible for the emission during the initiation phase, spatially resolved spectroscopy was applied to this part of the discharge. As stated already in Sec. 4.2.2, the most intense spectral features in this phase are those that are ascribed to the 4$d$-4$f$ transitions. The relative intensities of the strongest lines for the ionization stages 8+ up to 11+ have been plotted as a function of distance to the axis of symmetry in 5 ns time steps, covering the ring phase and the subsequent compression to the pinch, from the –30 to 0 ns time marks. The plots are shown in parts (a) to (f) of Fig. 4.5 together with the corresponding pinhole images from Fig. 4.4. Note that, since the spectra and pinhole images were recorded in separate experiments, the correspondence is not perfect, and the pinhole images should only be regarded as a quick reference.

For each time step, the relative populations of the different ionization stages were estimated using Eq. (4.1). Any possible populations of higher or lower stages than those considered here were simply ignored, and would have led to only a small correction of the relative populations. The result is plotted in Fig. 4.6. In Fig. 4.7, the effective average charge derived from these results is plotted versus time, together with the total intensity of the lines under consideration in arbitrary units. Note that the integrated intensity of the considered 4$d$-4$f$ lines was already decreasing while the pinch had not yet reached its minimum radius. This point will be discussed in Sec. 4.4.3 of the Discussion.

### 4.3.2 Propagating plasma after the pinch

The third phase of the discharge, the decay after the main pinch, is characterized by a considerable drop of the radiation intensity below the maximum of the discharge, and by expansion of the plasma. The expansion appears to have a preferential direction; this can be derived from the off-axis pinhole images as shown in Fig. 4.8. The source settings were similar to those of the on-axis pictures in Fig. 4.4. A number of 20-pulse integrated images with 5 ns effective exposure time were recorded using the 100 µm pinhole while varying the delay time in steps of 10 ns. The width and height of each picture correspond to an area of $1.85 \times 1.85 \text{ mm}^2$ in the source.
Fig. 4.5. Measured space-resolved, Abel-inverted relative line intensities of $4f-4d$ transitions belonging to different ionization stages of xenon. $d$ is the distance to the axis of symmetry of the discharge. Different spectra were produced in steps of 5 ns during the ring phase of the discharge, integrating ten shots for each image. For comparison, on-axis pinhole images are shown in the top right corner of each graph. These are from a different set of experiments, so the correspondence is not perfect.
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Fig. 4.6. Estimated relative populations of different ionization stages of xenon as a function of time, in the ring phase of the plasma, and during compression to the moment of minimum pinching radius.

Fig. 4.7. Total intensity of the considered 4d-4f EUV lines and effective average charge number as a function of time in the ring phase of the discharge.
4.3 Results

Fig. 4.8. Shape of the plasma pinch in the EUV as a function of time during the discharge. The MCP gating time was 10 ns; 20 pulse exposures were integrated per image. The time step between consecutive images is 10 ns. The actual size of each image is $1.85 \times 1.85$ mm$^2$.

In this figure, the first five or six images correspond to the first two phases of the discharge, the ring, and the compression to the pinch. In these two phases, not much EUV radiation is visible from the off-axis position because of shielding of the plasma by the edge of the anode. However, in the subsequent pictures, an apparently nearly spherical “ball” of plasma appears that travels in the vertical direction away from the cathode. Because of the axial symmetry of the source, the plasma must in reality be traveling along the symmetry axis. From this assumption, a propagation velocity can be derived of $3.6 \times 10^4$ m s$^{-1}$. Given that the sonic velocity will be on the order of $1 \times 10^4$ m s$^{-1}$ at most (for temperatures in the 50 eV range) this means that the compression must be supersonic. The propagation velocity compares well in order of magnitude to the average velocity found for the fast ions emitted by the source, as measured by the Faraday cup device.

4.3.3 Comparison between phases

Relative ionization stage populations

After the above discussion on qualitative characterization of the different phases during the main discharge, line intensity information from time-resolved spectroscopy can now be studied, giving information on the relative populations of the different ionization stages.

In Fig. 4.9 line intensities (in arbitrary units) for different stages are plotted against time during the discharge. On the time scale, $t = 0$ ns corresponds to the maximum EUV intensity from the plasma. For the EUV spectra, for all ionization stages $4d$-$5p$ lines have been selected for best comparability. However, in the case of Xe XII, next to the (presumed)
5p line at 12.39 nm, also the 4f line at 10.96 nm is shown for comparison. The line intensity for Xe xiii was calculated by correcting the intensity at 10.80 nm for the presence of Xe xii radiation: 0.4 times the signal intensity at 10.96 nm was subtracted from the gross value. This ratio of intensities was found by a parameter study of the spectrum from the discharge.

Note that the Xe xii line array at 12.39 nm coincides with radiation from Xe viii. The intensity was not corrected for this contribution, although it is probably dominant at $t \geq 80$ ns, where the other intensities from the highest ionization stages have nearly dropped to zero.

From the on-axis intensities in Fig. 4.9 it can be derived that the various ionization stages, ranging from Xe$^{7+}$ to Xe$^{11+}$, get populated during a relatively short time interval of about 20 ns. Despite the short time, it can be seen that the ionization stages get populated in order of increasing charge number. The time interval appears to coincide with the ring and compression phases observed in the pinhole camera images. Depopulation of all but the highest ionization stages happens on a much longer time scale. Radiation in the EUV can be observed for about 100 ns after the moment of maximum intensities, but in the VUV part of the spectrum between 30 and 90 nm (not shown), line radiation can be observed for as long as 300 ns after the maximum intensity was reached.

Relative line strengths within one ionization stage

Multiple line intensities from one and the same ionization stage do not always show similar time behavior. The clearest case of such dissimilarities in behavior is found for the Xe ix system, and is shown in Fig. 4.10. When compared to the $4d$-$5p$ lines, the $4d$-$4f$ line shows a relatively larger intensity in the initiation and compression phases of the discharge. The same dissimilarity can be observed in the Xe x spectrum, and, albeit less clearly, in the viii and xi spectra.
4.3 Results

-20 0 20 40 60 80 100 120

0.0 0.5 1.0 1.5 2.0 2.5 3.0 3.5 4.0 4.5

Intensity (arb. units)

t (ns)

$4d-5p$ 16.53 nm
$4d-5p$ 16.17 nm
$4d-4f$ 12.01 nm

4.3.4 Variation of the discharge voltage

Due to the self-breakdown operating mode of the source, the only way to change the discharge voltage and the pulse energy was to change the gas flow to the electrodes. As the hollow cathode discharge is operated on the left-hand side of Paschen’s curve, an increase of the gas flow (and thereby the background pressure) leads to a decrease of the discharge voltage.

EUV spectra have been recorded at different pulse energies. For each ionization stage that has lines in the EUV, the strongest intensity of the $5p$ transitions was selected for analysis—except for Xe $^{13+}$ ($12^+$), of which the $5p$ transition is overshadowed by other lines, and the top of the $4f$ array was selected instead. The same procedure as described in Eq. (4.1) and used in Sec. 4.3.1 was applied to estimate relative ion stage population densities.

The results are presented in Fig. 4.11. The figure shows that the relative populations of the different stages vary relatively slowly with input energy. On the contrary, the total EUV emission shows a sharp, faster than linear increase with pulse energy. The combined effect of a slightly increasing relative population of the $10^+$ ionization stage and the behavior of the absolute EUV emission causes a strong increase of the conversion efficiency with pulse energy within the probed regime, up to 4.7 J input energy.

In time-resolved measurements (not shown here) it was found that the dynamics of the pinch are also influenced by the pulse energy. With increasing energy, the time between the trigger signal and the beginning of the main discharge as well as the time between the appearance of the ring and the moment of minimal pinch radius decreased.

4.3.5 Dilution with argon gas

As mentioned in Sec. 4.1 during an upgrade of the source, the capacitance of the capacitor bank was increased from 48 nF to 674 nF, and an adjustment of the electrodes
geometry was made including a larger anode hole, enabling a larger collection angle of the EUV radiation from the pinch. In the adjusted setup, a small flow of xenon between the electrodes was combined with a much larger flow of argon, introduced between the anode and a downstream flow restriction. The application of argon introduced an additional free parameter for adjustment of the plasma properties.

It was found that the conversion efficiency decreased sharply when the xenon flow was reduced from the “standard” value of 8.6 sccm to a low value of 1.4 sccm, while at the same time the argon flow was increased from 193 sccm to 226 sccm to maintain a constant discharge voltage. The spectra, integrated in time over the discharge pulse, are shown in parts (a) and (b) of Fig. 4.12, for high and low xenon flows, respectively.

When both spectra are compared qualitatively, the following appear to be the main differences:

i. At the higher xenon flow, no Ar lines are visible. At low xenon flow, Ar VIII lines appear in emission.

ii. At low xenon flow, the xenon lines belonging to 4d-4f transitions are relatively intense compared to the 4d-5p lines.

iii. From high to low xenon flow, there is a relative increase of the intensity of the Xe XII 5p unresolved transition array (UTA) relative to the intensity of the Xe XI 5p UTA. A similar increase is visible in the Xe XII 4f intensity relative to that of Xe XI. Also, the intensity at 10.80 nm (tentatively attributed to Xe XIII, since higher stages are unlikely to be present in large quantities) grows relative to that of Xe XII at 10.96 nm.
4.4 Discussion

Fig. 4.12. Time-integrated EUV spectra for pulses with high (a) and low (b) xenon flow into the discharge region. In spectrum (b), EUV lines from argon appear in emission (identification according to Ref. [15]).

In all cases, radiation from higher ionization stages is favored when the xenon flow is decreased.

4.4 Discussion

4.4.1 Origins of the ring shape

The start of the discharge plasma in the main gap in a ring shape was an unexpected effect observed by pinhole imaging. In case the pulse would start as a diffuse discharge with a flat current distribution, the Lorentz force on a moving charged particle would scale linearly with the distance from the axis of symmetry. From this, it can be derived that the current distribution within the compressing plasma would remain flat (within a decreasing radius). However, any initial inhomogeneity in the distribution could grow due to a corresponding deviation in the Lorentz force.
Now, the question is what could be the cause of the initial inhomogeneity in the current distribution. One factor is the cathode geometry: The generation of secondary electrons by ions on the cathode surface during the high-current phase will take place mainly on the cathode edges, which have a ring shape, causing also a ring-shaped distribution of the current in the plasma phase.

Second, the skin effect, i.e., the exclusion of the fast-varying current from the center of the plasma, might also cause the current to run mainly along the edges of the plasma at the start of the high-current phase.

4.4.2 Preferential direction of plasma expansion

In the pinhole images of Fig. 4.8 it was seen that the pinch plasma does not decay simply isotropically, but it has a preferential direction along the axis. In the present results, only expansion in the forward direction is visible. However, the most likely explanation of the anisotropy is the anisotropy in the Lorentz force. While the pinch plasma is confined in the radial direction by the Lorentz force, it is free to expand in the axial direction. Therefore, a similar expansion in the inward direction (i.e., towards the cathode hole) is expected. A second contribution may come from the “zippering effect,” in which the plasma is accelerated in an axial direction due to an axial component in the Lorentz force.

4.4.3 Causes of spectral changes over time

In this subsection the main differences between the xenon EUV spectra from different phases of the discharge are discussed. In the discussion, the focus is on the relative populations of the different stages, and for radiation from a single stage, mainly on the lines emitted by Xe IX, since these lines (and their transition probabilities) are well established in the literature, and also resolvable individually (instead of only in line arrays). The intensities of the main Xe IX EUV lines versus time during the discharge were plotted in Fig. 4.10.

All of the identified transitions in the EUV are resonant, i.e., the lower level is (very close to) the ground state of the ion. This means that the wavelength of the line is a direct measure of the energy of the upper level. Therefore, the intensity of the lower wavelength lines (the 4d-4f transitions) relative to that of the larger wavelength (4d-5p) lines, is an indication of the electron temperature of the plasma, assuming a given excitation/deexcitation balance.

From rough estimates of the ion density (up to $10^{24} \text{ m}^{-3}$), average ionization stage ($\sim 10$), and electron temperature (up to $\sim 35$ eV) in the pinching plasma, it can be deduced that the excitation/deexcitation balance in most phases of the plasma will be neither pure corona nor pure LTE, but rather in between the two. In case of LTE, and an optically thin plasma, the intensities of the individual lines will be proportional to their transition probabilities. On the other hand, the intensities have an extra $1/({\Delta E})^3$ dependency in pure corona balance compared to LTE, since in this case the intensity of the radiation is
proportional not to the optical transition probability but to the electronic excitation rate from the ground state. This means that with similar electron temperature, a less dense plasma will have relatively stronger radiation at larger wavelengths compared to a more dense one.

During the ring phase of the plasma, the $4f$ intensities are relatively high, in spite of the necessarily low density of the plasma in this phase. This is an indication of a high electron temperature in this phase.

During compression of the ring to the actual pinch, the $4f$ line intensities remain high and that of the $5p$ lines increase. The increase of the intensity of the $5p$ lines can easily be explained by an increase of the density of the plasma (and resulting shift to a balance that is closer to LTE). The fact that the total intensity of the $4f$ lines does not increase correspondingly shows that the electron temperature must already be dropping at this time. This assumption is supported by the slight decrease of average ionization stage shown by detailed EUV spectroscopy in the ring phase (Fig. 4.7).

Admittedly, similar trends would be observed if, in this phase, the plasma would become optically thick to some of the $4f$ lines. However, neither the details of the experimentally determined EUV spectra nor comparison with the Planck limit derived from calculated transition probabilities and the above plasma parameters point in this direction.

During the decay of the pinch, the $4f$ intensity decreases quickly, while in the case of Xe IX the $5p$ line intensity stays almost constant for some time (about 40 ns). The relatively fast decrease of the $4f$ line intensity can be explained by cooling of the plasma and a shift of the population balance back towards corona. For the $5p$ lines, the cooling is compensated by repopulation of the Xe$^{8+}$ ionization stage by recombination from higher stages.

Even with a low electron temperature, the relatively low intensity of the $4f$ line is remarkable. Also, it may be noted that after $t = 60$ ns, both $5p$ intensities become nearly equal, while there was a clear difference in the intensities earlier in the discharge. As the upper energy levels for both lines are very close together, such a change cannot be explained by a change in the plasma parameters alone. A possible cause for the change is a “capture radiative cascade” effect, where recombination into an excited level of Xe$^{8+}$ and subsequent radiative decay to lower levels leads to a spectrum that is different from either normal LTE or corona balance.

Summarizing, it is stated that in the initial (ring) phase, the excitation/deexcitation balance is between pure LTE and corona balances, and the electron temperature is high; during the pinching, the density increases and the excitation balance moves closer to LTE, but electron temperature drops; and during the plasma decay, temperature drops further and the balance moves back towards corona. In the latter phase, specific recombination effects might have an influence on the shape of the spectrum.

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1 After publication of this chapter, EUV spectrum simulations have been performed that seem to indicate that opacity does play a certain role in determining the shape of the spectrum; see Chapter [8] for a detailed discussion.
4.4.4 Absence of Ar IX lines and changes in the Xe spectrum at low Xe flow

When the source was operated with the argon buffer gas at low xenon flow, some argon lines became visible. All these can be identified as belonging to the Ar VIII spectrum \cite{15}. However, comparison of ionization energies of the most abundant xenon ions (180 eV, 205 eV, 231 eV, and 258 eV for 8+ up to 11+ \cite{6}) with those of Ar\textsuperscript{7+} and Ar\textsuperscript{8+} (143 eV and 422 eV \cite{16}) shows that in the pinch, almost exclusively, eight times ionized argon should be present: Ar\textsuperscript{7+} has such low ionization energy that it should be completely ionized through to the next stage, whereas that of Ar\textsuperscript{8+} is so high that Ar\textsuperscript{9+} will not be populated considerably. Also, it is known that Ar\textsuperscript{8+} has some lines in the EUV range of 8–20 nm \cite{15}. However, the upper states responsible for these lines all have energies in excess of 300 eV above the ground state of this ion, and therefore will hardly be populated in the pinch plasma with a temperature of “only” about 35 eV. This explains their absence in the relatively cool pinch plasma.

The difficulty to either further ionize or excite the Ar\textsuperscript{8+} ions present in the plasma may be responsible for an increase of the electron temperature compared to the discharge with larger xenon flow. The electrons simply cannot lose their energy easily in either excitation or ionization. The increase in electron temperature is the most likely cause of the shift towards higher ionization stages observed in the xenon contribution to the spectrum. It may also be a large contributor to the shift towards relatively more intense radiation from the short-wavelength 4\textit{f} radiation compared to the 5\textit{p} transitions in xenon.

It should be noted that there might be a second contributor to this shift, which is the optical density of the plasma. Even though there are no indications in the present experimental results that the radiation from the plasma is optically thick at any time during the discharge, it could play a role: it is possible due to the dilution with argon that at low xenon flow, the 4\textit{f} transitions are less optically dense than at high xenon flow. Hence they might be enhanced compared to the 5\textit{p} transitions of the same stages.

4.5 Conclusions

The experimental results presented in this chapter may serve as proof of the good applicability of the cost-efficient combination of fast-triggerable MCP and photocamera diagnostics for characterization of EUV sources by pinhole imaging and EUV spectroscopy.

The described experiments have led to improved understanding of the pinch dynamics of the Philips hollow cathode discharge. A plasma was observed inside the cathode, it was shown that high ionization stages are present already before the main pinch occurs, and it was observed that the initiation of the main discharge occurs as a ring-shaped plasma.

In the EUV band from 9 to 18 nm, contributions from many ionization stages of xenon (ranging from 7+ to 12+) can be distinguished, and their relative populations during the plasma pulse can, in principle, be monitored.

The behavior of individual line intensities, especially for the Xe IX spectrum, can give
further insight into the characteristics of the plasma. Already, they provide indications of shifts in temperature and the excitation/deexcitation balance during the discharge. It was argued that they indicate a relatively high electron temperature and a balance between LTE and corona during the ring phase of the discharge, and lower electron temperature and the possible presence of radiative cascade processes during the decay of the plasma.

References

Chapter 4: Time-resolved imaging and EUV spectrometry on a HC discharge


Chapter 5

Characterization of a vacuum-arc discharge in tin vapor using time-resolved plasma imaging and extreme ultraviolet spectrometry

Abstract

Discharge sources in tin vapor have recently been receiving increased attention as candidate extreme ultraviolet (EUV) light sources for application in semiconductor lithography, because of their favorable spectrum near 13.5 nm. In the ASML EUV laboratory, time-resolved pinhole imaging in the EUV and two-dimensional imaging in visible light have been applied for qualitative characterization of the evolution of a vacuum-arc tin vapor discharge. An EUV spectrometer has been used to find the dominant ionization stages of tin as a function of time during the plasma evolution of the discharge.

5.1 Introduction

Future tools for semiconductor lithography, as currently under development, will apply light in the extreme ultraviolet (EUV) wavelength range for the projection of small-scale patterns onto wafers. Due to the limited reflectivity of the proposed multilayer optics for these wavelengths, and high throughput demands for commercial tools, very powerful EUV light sources are required for this application. Promising candidate sources include various types of discharge plasma sources.

Since the end of 2001, an EUV Laboratory has been in operation at ASML in Veldhoven. One of its aims is the characterization of candidate EUV sources, and their suitability for application in lithography. In the framework of a research cooperation between ASML and TU/e, a hollow cathode discharge in xenon from Philips EUV has been investigated using time-resolved pinhole imaging and EUV spectrometry; see Chapter 4 of this thesis [1].

Recently, various research groups worldwide have put an increased effort in investigating the replacement of xenon by tin as the working element in EUV discharges (several contributions in Ref. [2]). Although it creates some new challenges in the field of debris mitigation, tin has the major advantage over xenon that it has an EUV spectrum that is strongly peaked at the desired wavelength (around 13.5 nm).

A triggered vacuum arc in Sn vapor from the Russian Institute of Spectroscopy (ISAN) has been in operation in the EUV laboratory since March 2003. The discharge region consists of a flat, grounded cathode that is covered with a thin layer of liquid tin, and an anode located about 3 to 3.5 mm above the cathode. The anode has either a round or a semicircular hole in its center; the version with a semicircular hole was used in the experiments presented here. A schematic picture of the electrode cross section is given in Fig. 5.1. Before the start of the discharge, a positive electrical potential of 4 kV is applied to the anode. The discharge is started by the creation of a cloud of partly ionized tin vapor above the cathode, which expands towards the edges of the anode. Once the plasma near the anode has reached a sufficiently high density, a discharge is started. The current through the discharge increases to almost 20 kA in only a few tens of ns time, and a multiply ionized, EUV emitting plasma is formed. The strong current causes a pinch effect, meaning that due to Lorentz forces acting on the charged particles in the plasma, the plasma is compressed in radial direction to a needlelike shape on the axis of the discharge, with a diameter on the order of 100 \( \mu m \). In view of the dynamics of the plasma, the time development of the discharge can be roughly split up into four main phases: the trigger plasma, before the discharge current has started (1); the prepinch phase, in which the plasma is heated and ionized by the strong electrical current, and starts to compress (2); the pinch itself (3); and a decay phase (4). The actual pinch phase has a duration on the order of 10 ns only, and it is therefore very short-lived compared to the other phases of the discharge. This makes the center of the pinch phase a suitable zero on the time scale when
5.2 Experimental

Fig. 5.1. A schematic cross section of the electrodes of the tin vapor discharge.

one is describing the properties of the plasma in the other phases. The pinch occurs about 300 ns after the ignition of the discharge. The repetition rate of the source was limited to 5 Hz in the current work.

This chapter describes a series of experiments, similar in nature to those presented in Chapter 4, for the characterization of the tin discharge. Time-resolved pinhole images in the EUV range have been recorded. Time-resolved imaging has also been performed in the visible light, to achieve better understanding for those phases of the discharge in which the plasma is not hot enough to emit EUV radiation. Combined, they can be used to describe the plasma evolution during the entire discharge.

Time-resolved EUV spectrometry, supported by COWAN code calculations [3], serve to determine the dominant ionization stages in the hottest phases of the plasma, and help to compare the characteristics of different spatial parts of the discharge.

5.2 Experimental

Basic descriptions of the pinhole camera and EUV spectrometer setups have already been given in Chapter 4. In the present case, it was more convenient to use smaller distances between the various parts of the imaging system. Figure 5.2 shows a schematic view of the pinhole imaging setup. For pinhole imaging, the distance between source and pinhole was reduced to 39 mm, whereas a distance of 215 mm was used for the distance from the pinhole to a multichannel plate (MCP) detector, resulting in a geometrical magnification factor of $M = 5.5$. A 150 nm thick silicon/niobium filter was mounted just behind the pinhole to avoid blurring of the image by radiation from wavelengths above about 20 nm. The output light from the MCP was recorded by a commercial-type digital camera. The gain provided by the MCP could be varied by changing the input voltage, to reveal weak features of EUV emission early and late in the pulse, while preventing saturation of the MCP and the camera during the compression and pinch stages.

A schematic picture of the EUV spectrometry setup is shown in Fig. 5.3. A 1200 l/mm grating with 1 m radius of curvature was used at a 4° grazing angle of incidence for the
Fig. 5.2. Schematic view of the pinhole setup for plasma imaging at EUV wavelengths. Pumping holes in the pinhole holder were used for pumping down the space between the pinhole and the MCP. The numbers give the distances between the source, the pinhole and the MCP front surface in mm. The camera that was mounted behind the MCP assembly is not shown here.

Fig. 5.3. A schematic top-view image of the setup for EUV spectrometry on the tin discharge. On the left the plasma source is shown in its vacuum chamber. The space between both slits is pumped down through apertures in the first slit holder. The inside of the EUV spectrometer is pumped down through a separate connection to the turbo pump. The arrow marked by (1) indicates the direction where the EUV power meter was mounted. Not all sizes are drawn to scale.

recording of EUV spectra in the 5–30 nm wavelength range. Spatial resolution, in the vertical dimension of the discharge, was achieved by inserting a horizontally orientated slit into the spectrometry setup in front of the vertical EUV spectrometer entrance slit. Distances between source, first slit, second slit, grating and MCP were 118 mm, 217 mm, 70 mm and 210 mm, respectively. This resulted in a spatial magnification factor of 4.14 on the front surface of the MCP. Both for pinhole imaging and for spectrometry, signals were recorded from a direction perpendicular to the discharge’s axis of symmetry.

A wavelength calibration was achieved by comparing the Sn spectrum with the spectrum from an experiment where the tin cathode was replaced by a cathode that was filled with a mixture containing lithium and magnesium, for which the line wavelengths in the EUV are accurately known (as compiled in Ref. [4]). The lines that were used for the calibration were the Mg \( \text{v} 2p^3 3s (2D) 1D_2 - 2p^4 (1D) 1D_2 \) line and the Mg \( \text{vi} 2s2p^4 (3P) ^2 P_{3/2} - 2s^22p^3 (2P) ^2 D_{5/2} \) line at 14.29 nm \[5, 6\] and 27.04 nm \[6\], respectively. Both lines were chosen
because they are far apart in the spectrum, they were clearly visible above the background and they were well isolated, so that their position could be determined with good accuracy.

For two-dimensional imaging in the visible light range, an intensified charge-coupled device (ICCD) camera from Andor, type DH510-18, was mounted behind a set of two achromatic lenses with 600 mm focal length, arranged to achieve a magnification factor of 1.14. The discharge was imaged into a horizontal direction, perpendicular to the axis of symmetry of the discharge, and through a glass window. The wavelength sensitivity of this imaging system was limited by the transparency of the glass window and lenses, and the sensitivity curve of the camera’s photocathode, to about 300–850 nm. To avoid overexposure of the ICCD camera (even at the lowest gain setting of its MCP), an aperture of 3 mm diameter was placed between the two lenses. As in the case of the EUV diagnostics, gating of the imaging system was achieved by switching of a high voltage over the MCP.

The triggering of the diagnostics was in both cases synchronized to the ignition of the discharge. Typically, the time resolution that could be achieved relative to the evolution of the plasma, was on the order of 10 ns, and was limited by both the pulse-to-pulse jitter in the discharge of about 5 ns, and the effective optical gate time of 5 ns.

A magnetic field probe in the electric circuit provided a current signal, displayed on a fast oscilloscope. This signal served as a monitor for the stability of the source. During the EUV experiments, an additional monitor was provided by a voltage signal from a photodiode, that was mounted behind an arrangement of two multilayer mirrors and a silicon/niobium filter. The arrangement was positioned diametrically across the EUV spectrometer, in the horizontal plane of the discharge. This tool provided information about the absolute “in-band” EUV emission per pulse in a 2% wavelength band around 13.5 nm.

### 5.3 Plasma evolution

In this section, a detailed, although mainly qualitative, description of the evolution of the discharge as a function of time during the discharge pulse will be given on the basis of a series of visible light and EUV images. The visible light images, which cover the entire time range relevant for the discharge, are shown in Fig. 5.4. In the plasma imaging experiments, the position where the discharge current attached to the anode was about 3 mm away from the cathode surface. However, the region closest to the anode is obstructed from view by some small structures that are on the anode between the discharge and the camera.

The timing indicated in the top left corner of each image is relative to the pinch, as explained in Sec. 5.1. For the first seven images, the radiation was very weak, so that their intensity had to be digitally increased by a factor 50 relative to the other images. Figure 5.5 shows typical plots as a function of time of the discharge current and the integrated EUV emission, as derived from the MCP images.

The first six images of Fig. 5.4 (up to 135 ns before the pinch) only show a weak, decaying plasma that was created by the discharge ignition. What follows is a fast transition to the
Fig. 5.4. Time-resolved visible light images recorded during the tin discharge. Each image corresponds to an area in the source of 5.2 mm wide times 3.9 mm high. The horizontal dashed line in the first image indicates the position of the cathode surface and corresponds to a length of 1 mm. The anode is near the top of each image; however, the plasma very close to the anode is obstructed from view by some small structures on the anode between the discharge and the camera. The time indication in the top left corner of each image is relative to the center of the pinch phase. The ignition of the discharge happens at about $-300 \text{ ns}$. 

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5.3 Plasma evolution

Fig. 5.5. Discharge current (solid curve) and EUV emission derived from MCP images (dotted curve) during the discharge. The absolute value of the current is derived from the electrical circuit parameters, and is only approximate.

Fig. 5.6. A four-times intensified image recorded at 65 ns before the pinch. Visible light emission shows up not far from the anode (near the top of the image).

The high-current phase of the discharge, as can be derived from Fig. 5.5, is the appearance of a bright spot on the cathode, followed by increased emission from a region near the cathode. Also, a spot appears near the anode about 65 ns before the pinch. A four-times intensified image of the visible emission at this time, showing the emission near the anode, is shown in Fig. 5.6. The emission becomes visible on the intensity scale of Fig. 5.4 only in the images at $-35$ and $-25$ ns.

The anode spot must be generated by a thin, but very hot plasma. This is proven by the fact that it also shows up in the EUV images, as shown in Fig. 5.7. The image in this figure was recorded 40 ns before the pinch at an MCP voltage of 6.24 kV, and the signal was increased by a factor 4 during digital processing.

Compression of the discharge plasma starts about 30 ns before the actual pinch. During compression, the EUV emission rapidly increases, as is shown in the first four images of Fig. 5.8. This series of images was recorded between 20 ns before and 15 ns after the pinch at an MCP voltage of 5.25 kV. Effectively, the sensitivity of these recordings was about 30
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Fig. 5.7. EUV pinhole image of the discharge at 40 ns before the pinch, recorded at an increased MCP voltage of 6.24 kV. The area of the image corresponds to an area of 1.6 mm wide and 4.2 mm high in the source. The dashed line gives an indication of the position of the surface of the cathode. Like in the visible light images, the real anode position corresponds to the top of the frame, but the region closest to the anode is obstructed from the view. In this image, some EUV emission is visible from the region just outside this obstruction.

times lower than in Fig. 5.7

The images for 0–15 ns after the pinch show that a ball of plasma moves in the direction away from the cathode, at a velocity of about $4 \times 10^4$ m s$^{-1}$. A similar phenomenon can be observed in the visible light images of Fig. 5.4. A possible explanation comes from the fact that the current-induced Lorentz force does confine the plasma in the radial, but not in the axial direction. Since the surface of the cathode is close to the bottom of the pinch, the plasma can only escape in the upward axial direction.

Just after the pinch, visible light plasma radiation appears along the cathode surface. Since a similar emission feature does not appear in the EUV, it must be generated by a much cooler plasma. Most likely, the current through the pinch plasma and absorption of EUV radiation lead to heating of the liquid tin on the cathode surface. Evaporation and partial ionization of tin from the cathode can result in a relatively cool plasma.

In the next 150 ns, the “cool” plasma expands and travels towards the anode. About 100 ns after the pinch, a filamentlike structure appears near the anode. This structure might be created by evaporation of tin from the anode that was deposited there during previous discharge pulses. The intensity of the filament emission increases after 150 ns, perhaps due to a collision with the expanding plasma from the cathode. The anode plasma expands back towards the cathode, but after this time the emission gradually decays. Since these dynamics are not relevant for the EUV emission, we have not studied them in detail.

5.4 Time-resolved EUV spectra

A typical example of the EUV spectrum of the tin source is shown in Fig. 5.9. The spectrum in this figure corresponds to the emission from near the cathode in the pinch phase, during
5.4 Time-resolved EUV spectra

![Fig. 5.8. Series of time-resolved EUV pinhole images during the tin discharge. The images were recorded from 20 ns before until 15 ns after the pinch, in 5 ns time steps. The dashed line in the first image corresponds to a length of 1 mm in the source, and indicates the position of the cathode surface. Each frame corresponds to a size of 1.6x4.2 mm². The feature below this line, near the bottom of each image is due to a reflection of the plasma EUV emission from the surface of the cathode. The anode is just outside the upper edge of each image.]

Table 5.1. The wavelength ranges that were used for integration of the radiation from the 5p-4d unresolved transition arrays (UTAs) of tin ionization stages from 7+ to 10+.

<table>
<thead>
<tr>
<th>Ioniz. stage</th>
<th>(\lambda_{\text{min}}) (nm)</th>
<th>(\lambda_{\text{max}}) (nm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>7+</td>
<td>22.2</td>
<td>22.8</td>
</tr>
<tr>
<td>8+</td>
<td>19.5</td>
<td>20.1</td>
</tr>
<tr>
<td>9+</td>
<td>17.3</td>
<td>18.1</td>
</tr>
<tr>
<td>10+</td>
<td>15.5</td>
<td>16.3</td>
</tr>
</tbody>
</table>

which the EUV emission is strongest. Calculations using the COWAN computer code\[3\] reveal that the large peak at 13.5 nm is caused by 4f-4d transitions from a number of ionization stages of tin, ranging from 8+ to 11+. The 5p-4d transitions of these ions and the 7+ ion form unresolved transition arrays (UTAs) in the wavelength range from 14 up to 23 nm. These UTAs are identified in Fig. 5.9. The wavelength ranges for each of the 5p-4d UTAs from 7+ to 10+ are listed in Table 5.1. The features at lower wavelengths (below 12 nm) are believed to be caused by transitions to the 4d ground state configuration from higher energy levels (5f, 6p) of the same ions.

The fact that the 5p-4d emission features are separated in wavelength for the different ionization stages of tin, makes them suitable for deriving information on the time dependency of the populations of the various ion stages. To this end, time-resolved EUV spectra have been recorded from 75 ns before until 125 ns after the pinch. They have been integrated over three separate spatial regions in the plasma: close to the cathode (from 0.1 up to 0.45 mm from the cathode surface), further away from the cathode (from 1.15 to 1.5 mm
Fig. 5.9. An example of a recorded EUV spectrum from the tin source, including tentative identifications of the UTAs. This particular spectrum corresponds to the region near the cathode during the pinch phase. The dotted curve is the same spectrum for the lower wavelength part, but on a ten times magnified scale.

away) and near the anode (3.0 to 3.35 mm from the cathode surface). For each ionization stage, the emission in a spectrum has been integrated over the corresponding wavelength range as given in Table 5.1. For the near-cathode region, the integrated intensities have been plotted versus time in Fig. 5.10. Since the figure shows that the absolute emission from the 7+ and 8+ stages is much lower than the emission from the 9+ and 10+ ions, the curves have been replotted in Fig. 5.11 in which each curve has been normalized to its peak value.

Figure 5.11 shows that from 65 to 15 ns before the discharge, the spectrum tends towards increasing ionization stages. The emission from the 7+ ion even decreases after 35 ns before the pinch as it probably gets ionized further to higher stages. During this time, the plasma is heated by the strong electric current. However, during the pinch (around 0 ns on the time scale) the average ionization stage does not seem to increase further. The emission from the 10+ ion still increases somewhat relative to the Sn\(^{9+}\) emission; but on the other side, the relative importance of the 7+ and 8+ ion emission also increases. The plasma seems to be cooler than before the pinch. Perhaps cooler material near the discharge axis is mixed with hotter plasma from further away from the axis during the compression process. Additionally, the 9+ and 10+ radiation might be relatively more limited by opacity of the plasma, although it is still much weaker than the emission in the 4f-4d peaks at the same time.

Under the conditions that govern EUV discharges, with electron temperatures of a few tens of eV and electron densities up to about \(10^{25} \text{ m}^{-3}\), the main recombination process is radiative recombination. For example, in the model of Colombant and Tonon [7],
5.4 Time-resolved EUV spectra

Fig. 5.10. Relative intensities of the 5p-4d UTAs of various tin ionization stages as a function of time during the pulse, as measured near the surface of the cathode.

Fig. 5.11. 5p-4d UTA intensities measured near the cathode surface. Each curve is normalized to its own peak intensity during the discharge.
the contribution of three-particle recombination is less than 2% relative to the radiative recombination for the 9+ to 10+ ionization step, and taking plasma parameters $T_e = 35$ eV and $n_e = 1 \times 10^{25}$ m$^{-3}$ that are typical for pinch plasmas. This means that the average ion charge will hardly decrease with increasing electron density, so that the increasing density due to the pinch effect can be excluded as the direct cause of the shift to lower ionization stages.

The EUV emission from the region further away from the cathode is much weaker than the emission from near the cathode, as is indicated by the intensity scale in Fig. 5.12. The same measurements were used as for Fig. 5.10 and the same procedure was followed. The figure shows that during the pinch, the emission from lower ionization stages in this region is relatively less important than in the near-cathode region, which is an indication of a hotter plasma. The EUV emission from this region has a longer duration than near the cathode. In this figure, a strong peak in the emission from the lower ionization stages appears about 50 ns after the pinch near the cathode. From the EUV pinhole images in Fig. 5.8, we know that this peak is probably caused by a volume of plasma that is moving in the upward axial direction away from the pinch. From the time delay between the appearance of the peak in the two regions, we can derive a velocity of the plasma of around $2 \times 10^4$ m s$^{-1}$. This velocity is about a factor two lower than the one derived in the previous section. However, such a difference can easily be explained by inaccuracies in the experiments and changes over time in the detailed behavior of the discharge.

Near the anode, the emission was even weaker and therefore too noisy to derive reliable time–dependent data in the manner described above. The EUV emission from this region is strongly peaked in time around 35 ns before the pinch, and consists almost exclusively of a peak in the spectrum near 13.5 nm, as is shown in Fig. 5.13. This behavior confirms that the emission near the anode is generated by a thin, but very hot plasma, as was already

Fig. 5.12. Relative 5p-4d UTA intensities measured further away from the cathode surface.
5.5 Conclusions

The time-resolved visible and EUV images of the tin discharge plasma show a plasma that is created by the discharge current, and compresses to form a short–lived pinch on the axis of the source. After the pinch, a certain volume of plasma moves along the axial direction away from the cathode. The EUV spectra suggest that the ionization of the plasma takes place mainly before and during the compression of the plasma, and that the highest temperature is reached before the actual plasma pinch. These features all show strong qualitative similarities of the tin discharge with other EUV producing discharge plasmas, in particular the hollow cathode discharge.

However, a remarkable difference with the characteristics of the hollow cathode discharge is the sudden increase of the emission from relatively low ionization stages (7+, 8+ in the case of the tin discharge) during the pinch phase. Also for the hollow cathode discharge it was noticed that the average ionization degree seemed to be already slightly decreasing before the end of the compression phase, as discussed in Chapter 4, but the effect appears to be much stronger in the case of the tin discharge.

The spectra in the EUV range of the near-cathode region of the discharge confirm the suitability of the source for efficient production of EUV for application in lithography. The relative contribution of the region near 13.5 nm to the total spectrum is much larger than in the case of xenon plasmas.

Although some long-term changes in the behavior of the source cannot be excluded, the stability and reproducibility of the source on the short term were found to be good enough to perform more elaborate time-resolved experiments in which integration over

Fig. 5.13. The EUV spectrum as emitted by the plasma near the anode, 35 ns before the pinch.
a large numbers of pulses is necessary. One technique that requires such experiments is Thomson scattering, a laser spectroscopic technique in which the spectrum of light from a laser pulse, scattered by free electrons in the plasma, is recorded. On the basis of our findings, initial measurements using this technique have been performed, and the results of these can be found in Chapter 8.

References


Chapter 6

Comparison of experimental and simulated extreme ultraviolet spectra of xenon and tin discharges

Abstract

Xenon and tin both are working elements applied in discharge plasmas that are being developed for application in extreme ultraviolet (EUV) lithography. Their spectra in the 10–21 nm wavelength range have been analyzed. A fully analytical collisional-radiative model, including departure from equilibrium due to a net ionization rate, was used to simulate the EUV spectra. Detailed Hartree-Fock calculations, using the COWAN package, were applied for determination of the energy levels and optical transition probabilities of the 8+ to 12+ ions of both elements. For the calculation of the radiation, the opacity of the plasma was taken into account. Time-resolved measurements of the spectra from ionizing phases of two different discharge plasmas were corrected for the wavelength-dependent sensitivity of the spectrometer, and compared to the results of the simulations. Fairly good agreement between the experiments and the model calculations has been found.

Secs. 6.1 to 6.5 have been published as E.R. Kieft, K. Garloff, J.J.A.M. van der Mullen, and V. Banine, Phys. Rev. E 71, 036402 (2005).
Chapter 6: Experimental and simulated EUV spectra of xenon and tin

6.1 Introduction

Discharge plasmas are currently regarded as the most promising concept for application as sources of high-power extreme ultraviolet (EUV) radiation in semiconductor lithography. Various types have been developed or are still under development by a number of groups in the world [1–8]. The working element in such a plasma has to be selected for its emission near 13.5 nm, since this is the wavelength for which the silicon/molybdenum (Si/Mo) multilayer mirrors in the optical system of the lithographic apparatus will be optimized. The two most popular elements are xenon and tin. Xenon has the advantage of being a noble gas, whereas tin is a solid at ambient conditions, and therefore might get deposited onto mirrors and other optical surfaces. However, tin is a much more efficient radiator at the desired wavelength. Other efficient radiators, such as oxygen and lithium, are less popular due to their chemical reactivity.

Since xenon and tin are both large, complex atoms, their spectra in the EUV range of roughly 10–21 nm do not consist of sharp, well separated peaks, but rather form a quasi-continuum of a large number of peaks that cannot be resolved individually in a compact EUV spectrometer. The result of such a broad spectrum is that apart from the desired “in-band” radiation in a 2% wavelength band around 13.5 nm, also a large amount of out-of-band radiation is emitted. While this out-of-band radiation does not contribute to the illumination of the wafer in the lithography apparatus, it does contribute to the undesired heating of the discharge electrodes and the first optical elements in the system.

Therefore, from the application point of view, there is a desire to obtain detailed information on the shapes and intensities of the EUV spectra of xenon and tin and insight into the mechanisms that determine the relative intensity of the in-band radiation.

EUV spectrometry can be applied to obtain information about the (time-resolved) spectra of xenon or tin in a discharge plasma. In our EUV laboratory at ASML, such experiments have been performed on two different types of discharge plasmas: a hollow cathode discharge in xenon (see Chapter 4 [9]) and a triggered vacuum arc in tin vapor (Chapter 5 of this thesis [10]). Basic descriptions of the working principles of these two discharge plasmas are given in Ref. [7] and in Sec. 2.4 and Chapter 4 of this thesis; and Sec. 2.5 and Chapter 5 of this thesis, respectively.

So far, the interpretation of the results has been performed without a detailed consideration of the wavelength dependency of the spectrometer sensitivity. Either the behavior of individual contributions to the spectrum was studied as a function of time during the pulse, or line intensities were compared among each other that were relatively close together in wavelength. In the latter case, a flat sensitivity curve for the spectrometer could be assumed without potentially making large errors. This assumption was supported by the fact that from theoretical considerations, no large deviations from a smooth curve (i.e., no sudden “jumps”) were to be expected. However, both for evaluating the effects of the
spectrum emitted by the source on the mirror optics in a lithography tool, and for comparison with simulated EUV spectra, it was desirable that the theoretical considerations would be checked experimentally, and that the spectrometer sensitivity would be evaluated over the larger EUV wavelength range of (at least) 10–21 nm.

Studies of experimental EUV spectra and comparisons with simulations have been made in the past for various elements in different plasmas, including impurities in tokamaks. Examples are works on the spectra of tungsten \[11, 12\], krypton and argon \[13\], and calcium \[14\], where the latter was aimed at benchmarking the analyses of solar spectra. The typical densities, temperatures, and lifetimes of tokamak plasmas are, however, quite strongly different from those of plasmas that are designed for application in EUV lithography.

Elaborate analyses of the latter have been presented previously by Gilleron \textit{et al.} \[15\] for a laser-produced plasma (LPP) in a dense spray of droplets and Böwering \textit{et al.} \[16\] for a dense plasma focus device. Both authors used xenon as the working element and have regarded only time-integrated EUV spectra, which complicates the interpretation of the results. Also, Gilleron uses a local thermal equilibrium (LTE) approach. Böwering claims to use a non-LTE approach, but still applies a simple Boltzmann factor for the emission from the excited states, which essentially results in the same shape of the spectrum. Although an LTE-like approach is appropriate for LPPs, it may give less accurate results for the less dense discharge plasmas. Both works take opacity effects into account to explain the high observed ratio of intensities between the features around 13.5 and 11 nm. However, for a correct evaluation of opacity effects it is essential to use a reasonably accurate description of line broadening, details of which can be found in neither of these works. Recently, Richardson \textit{et al.} \[17\] have reported that they are working on a non-LTE radiation transport code for providing detailed spectra of laser-produced plasmas in xenon and tin.

The aim of this work is twofold. First, we present a theoretical and experimental investigation of our spectrometer sensitivity, so that we can obtain sensitivity-corrected spectra that are suitable for evaluation over larger wavelength ranges, and comparison with simulation results.

Second, we present a model which calculates the emitted EUV radiation from the ion density, electron temperature, size and geometry, and an effective net ionization rate of the plasma at a given time. The simulated spectra are matched, by variation of certain input parameters, to previously obtained experimental results which are now corrected for the spectrometer sensitivity. The aim is to obtain estimates for the main plasma parameters, and, more importantly, to gain insight in what mechanisms govern the population of excited states and the shapes and intensities of the radiated EUV spectra.

The spectral model was based on the radiation module of an existing model describing the evolution of laser-produced plasmas (LPPs) \[18\], but it was adjusted for the properties of discharge plasmas. The adjustments include the application of an analytical collisional-radiative model (CRM) for the calculation of excited state densities and more detailed calculations of line broadening and its influence on the opacity of the plasma. Also, an
effective net ionization rate has been introduced to account for the fact that in an ionizing plasma, the ion stage distribution lags behind the instantaneous electron temperature. The plasma model, and the atomic data calculations which were used as input to the model, are described in the following section.

In Sec. 6.3, we discuss the theoretical dependencies of the sensitivity of our EUV spectrometer on wavelength, and the sensitivity curve derived in this way is compared to the results of an experimental calibration.

In Sec. 6.4, the results of the simulations for both the xenon and the tin source are presented. In Sec. 6.5, the simulated and experimental spectra are compared, the remaining differences are discussed, and certain conclusions on the plasma properties are drawn.

We conclude this chapter with two sections that present and discuss possible extensions to the model. Sec. 6.6 deals with longer wavelength spectra, while in Sec. 6.7, the influence of doubly excited states is discussed.

### 6.2 Spectral model

#### 6.2.1 Radiation module

In Ref. [18], a computer model was described that simulates the evolution and radiation of a laser-produced plasma for generation of EUV radiation. Due to the modular design of the model, it was possible to separate the calculation of the radiation from the remaining parts of the model and to compile it into a separate executable file. The model requires as its input information on the geometry, chemical composition, total atom density, and electron temperature of the plasma. Also, atomic data for the elements present in the plasma are fed to the model in the form of input files.

From the total atom density and the temperature, first the electron density and the distribution of the species over the various ionization stages is calculated self-consistently, based on an LTE assumption with Saha equilibrium for the population of the different ions. Second, the population densities for the excited states are calculated assuming Boltzmann distributions. As a next step, the model calculates the radiation emitted by the plasma. Finally, in the original model, the total radiation at each wavelength is cut off at the Planck level calculated over the total area of the plasma surface, at the given electron temperature. For this, the plasma is assumed to have a spherical shape. The output is written in the form of spectra over a certain wavelength range, giving the total radiation, the contributions from free-free (bremsstrahlung), free-bound (recombination), and bound-bound (line) radiation, and the radiation uncorrected for opacity effects.

For the work presented here, some changes have been made to the original radiation module to make it more appropriate for application to discharge plasmas, which typically have far lower electron densities than LPPs. In this section, these changes will be discussed in detail, first for the calculation of the distribution function of the ions over the ionization stages, then for the excited state densities, and next for the emitted radiation. Finally, a
description will be given of the atomic data calculations leading to information about the energy levels and optical transition probabilities of the relevant ions.

6.2.2 Ion densities

First of all, an analytical collisional-radiative model (CRM) was introduced as an alternative to LTE for the calculation of the population densities of the various ions and excited states. The CRM is based for the largest part on the corona model described in Refs. [19, 20], which is valid for stationary and ionizing plasmas. In this model, the excited state density is given as a function of the effective principle quantum number (pqn) \( p \), which is given by

\[
p = Z \sqrt{\frac{R_y}{E_{\text{ion}} - E_{\text{exc}}(p)}}.
\]

Here, the symbol \( p \) is used instead of the frequently used symbol \( n \) to avoid confusion with densities. \( Z \) represents the charge number of the ion without the outermost electron, so that \( Z = z + 1 \), with \( z \) being the charge number of the ion under consideration. \( R_y \), \( E_{\text{ion}} \) and \( E_{\text{exc}}(p) \) are the ionization energy of hydrogen, the ionization energy of the ion under consideration, and the energy of the excited state relative to the ground state, respectively. Some of the equations that are discussed below were originally derived for hydrogen atoms or hydrogenic ions in which the effective pqn and the actual pqn of an excited state are equal; however, with the appropriate adjustments they can also be applied as approximations for complex ions.

An important role is played in the CRM by the level \( p = p_{cr} \), which is the boundary between radiative and collisional levels: for levels with \( p < p_{cr} \), the decay is mainly radiative and we have a coronalike balance, whereas for \( p > p_{cr} \), it is mainly collisional, and we are in an excitation saturation balance (ESB). The position of this boundary level \( p_{cr} \) is determined by the criterion \( n_e K(p_{cr}) = A(p_{cr}) \), which means that for the critical level, the total collisional destruction equals the total radiative destruction. For the collisional destruction rate, Ref. [20] gives

\[
K(p) = 6 \times 10^{-14} \text{ m}^3 \text{ s}^{-1} Z^{-2} p^4 \frac{\sqrt{T_e}}{T_e + 2Z^2} (1 + \varepsilon_p/4) \ln (2/\varepsilon_p + 1.3),
\]

where \( T_e \) is the electron temperature in eV and \( \varepsilon_p = [E_{\text{ion}} - E_{\text{exc}}(p)] / k_B T_e \). Note that by using a single electron temperature, we implicitly assume a Maxwellian electron energy distribution function (EEDF). In Sec. 6.5 the validity of this approach for the plasmas under study will be verified.

In our calculations we use the above expression, but with two adjustments: first, we omit the \( Z^2 \) scaling in the denominator, since distorted wave calculations [21] that we have performed for certain collisional excitation cross sections of \( \text{Xe}^{8+} \) ions do not seem to confirm the quasi-hard-sphere collision behavior for high values of \( \varepsilon_p \) in ions, as suggested
Chapter 6: Experimental and simulated EUV spectra of xenon and tin

in Ref. [20]. Also, we doubled the prefactor to account for the fact that in complex ions the typical energy distances between near-lying levels are smaller than in hydrogenic ions; this effect enhances the collisional destruction rate. The resulting expression is

\[
K(p) = 1.2 \times 10^{-13} \text{ m}^3 \text{ s}^{-1} \ Z^{-2} p^4 \frac{\sqrt{T_e}}{T_e + 2} (1 + \epsilon_p/4) \ln (2/\epsilon_p + 1.3). \tag{6.3}
\]

Further, for the radiative destruction rate we use

\[
A(p) = \gamma Z^4 p^{-5} (3 \ln p - 2 \ln p_1 - \zeta) \tag{6.4}
\]

in which \( \gamma = 7.87 \times 10^{-9} \text{ s}^{-1} \), \( p_1 \) is the effective pqn of the ground state of the ion, and the opacity-dependent parameter \( \zeta \) equals 0.25 for the optically open case. Here, the expression from Ref. [20] was adjusted to account for the fact that \( p_1 > 1 \) for ions that carry more than two electrons, and excited states cannot radiate to levels which have an effective pqn below that of the ground state of the ion.

The critical level is thus given by

\[
\rho^0_{cr} = 6.6 \times 10^{22} n_e^{-1} Z^6 \frac{T_e + 2}{\sqrt{T_e}} \left( \frac{3 \ln p_{cr} - 2 \ln p_1 - 0.25}{(1 + \epsilon_p/4) \ln (2/\epsilon_p + 1.3)} \right) \tag{6.5}
\]

with the electron density \( n_e \) in units of \( \text{m}^{-3} \). A further simplification is made by setting the value of the expression between brackets in Eq. (6.5) equal to 1.5, which turns out to be a reasonable value for the simulations in this study.

A further parameter used in this work is \( p_{hc} \), which determines whether collisional excitation processes or deexcitation processes are dominant, and is given by

\[
p_{hc} = Z \sqrt{\frac{Ry}{(3 k_B T_e)}}. \tag{6.6}
\]

For \( p > p_{hc} \), the ESB is called hot, otherwise it is cold.

Now, the distribution over the subsequent ionization stages is derived from a balance between collisional ionization, radiative recombination, three-particle recombination and an effective net ionization rate, in the following manner:

\[
\frac{n_{z+1}}{n_z} = \frac{n_e S_z}{n_e \alpha_{r,z+1} + n_e^2 \alpha_{3p,z+1} + \nu_i}, \tag{6.7}
\]

where \( S_z \) is the collisional ionization coefficient of the ion with charge number \( z \), \( \alpha_{r,z+1} \) is the radiative recombination coefficient of the next ion, and \( \alpha_{3p,z+1} \) is the three-particle recombination coefficient of that ion. \( \nu_i \) represents an effective net ionization rate, as explained below. \( n_z \) and \( n_{z+1} \) are the total densities of the ions with charge numbers \( z \) and \( z + 1 \), respectively.

For the collisional ionization coefficient, we use a sum of the direct ionization coefficient of Vriens and Smeets [22] \( K(p_1, +) \) and the collisional excitation coefficients from the ground state to the different levels that are in hot ESB,
6.2 Spectral model

\[ S_z = K_z(p_1, +) + \sum_{p=p_1, p_1+1, \ldots \leq 24} K_z(p_1, p) \]

\[ = K_z(p_1, +) + 1.6 \times 10^{-11} \text{ m}^3 \text{ s}^{-1} \sum_p f_{p_1 p} g(p_1, p) \frac{\Delta E}{T_e} \exp \left( -\frac{\Delta E}{k_B T_e} \right), \quad (6.8) \]

in which \( p_1 \) is the largest of \( p_{cr}, p_{hc}, \) and \( p_1 + 1; \) \( f_{p_1 p} \) is an analytical approximation for the optical oscillator strength of the transition, as given in Eq. (3.9) of Ref. [20], \( \Delta E \) is the excitation energy in eV of the level with \( pqn \). The gaunt factor \( g(p_1, p) \) is set equal to 0.16, where we use the fact that it approaches a constant value for near-threshold excitations in ions [23], and the numerical value was derived from the distorted wave calculations [21] as mentioned above.

By applying this ionization rate, we take into account that the stepwise excitation processes that finally lead to ionization are very fast compared to the first excitation to a level that is in hot ESB. Since both \( f_{p_1 p} \) and \( \exp \left( -\frac{\Delta E}{k_B T_e} \right) \) are rapidly decreasing functions of \( p \) for the relevant values of \( p \), by far the largest contribution to \( S_z \) is formed by just the first term in the summation over \( p \), and because typically \( p_1 < 5 \), the cutoff for the sum, \( p \leq 24 \), does not have any appreciable effect on the result. Also, the calculations show that direct ionization contributes only a few percent to the total ionization rates, so that the stepwise ionization process is dominant.

The three-particle recombination coefficient \( \alpha_{3p,z+1} \) is derived from the collisional excitation rate by imposing Saha equilibrium in the limit of high electron density; the radiative recombination coefficient \( \alpha_{r,z+1} \) was taken from Ref. [24] as cited in Ref. [25].

In discharge EUV plasmas, the time constants for reaching ionization equilibrium are typically not very small compared to the lifetime of the plasma, as discussed, e.g., in Ref. [26]. Therefore, nonequilibrium effects during the ionization phase of the discharge will have a non-negligible effect on the distribution over the different ionization stages. The radiation module, being based on a quasi-steady-state assumption for the distribution of the ions, is not capable of taking these effects fully into account. However, to mimic the effect of an ionizing plasma, an optional additional parameter, the effective net ionization rate \( \nu_i \) was introduced, which can be interpreted as the time derivative of the average ion charge number \( z_{av} \). The value of \( \nu_i \) compared to the recombination rates of the different ions is an indication of the relative importance of the ionization nonequilibrium.

6.2.3 Excited state densities

After the densities of the different ions have been calculated, the population densities of the excited states are determined. These are expressed as an overpopulation compared to the Saha density, which is given by

\[ n_z^S(p) = \eta_{z+1}(p_{1,z+1}) \frac{1}{2} n_e \frac{h^3}{(2\pi m_e k_B T_e)^{3/2}} \exp \left[ \frac{\left( E_{ion} - E_{exc} \right)}{k_B T_e} \right]. \quad (6.9) \]
Here, \( \eta_z(p) \) stands for the density per statistical weight of the levels with effective \( pqn \) \( p \), and hence \( \eta_{z+1}(p_{1,z+1}) \) represents the density per statistical weight of the ground state of the ion with charge number \( z + 1 \). For \( p < p_{cr} \), we are in the corona domain. Here, the overpopulation of the excited level is expressed as a fraction of the overpopulation of the ground state [based on Eq. (9.6) in Ref. [20]]:

\[
 b(p) - 1 = \frac{\eta_z(p) - \eta_z^S(p)}{\eta_z^S(p)} = (b_1 - 1) \max \left( \frac{n_e K(p, p_1)}{A(p)}, 1 \right), \tag{6.10}
\]

where \( b_1 - 1 \) is the normalized overpopulation of the ground state, which is derived from the actual ratio of \( z \) to \( z+1 \) ground state densities that follows from Eq. (6.7), compared to the Saha ratio (6.9). The expression contains both the collisional deexcitation to the ground state \( K_z(p, p_1) \) and the total radiative deexcitation \( A(p) \). The collisional deexcitation rate is given by

\[
 K(p, p_1) = 1.6 \times 10^{-11} \text{ m}^3 \text{ s}^{-1} \frac{f_{pp1} g(p_1, p)}{\Delta E \sqrt{T_e}} \\
 \approx 3.0 \times 10^{-13} \text{ m}^3 \text{ s}^{-1} \frac{p_1^5}{Z^2 p^2 y^4 \sqrt{T_e}}. \tag{6.11}
\]

Here, \( y = 1 - p_2^2 / p^2 \) is the ratio of the excitation energy to the ionization energy of the ion.

For the radiative decay, we use an approximation to Eq. (6.4):

\[
 A(p) = 1.3 \times 10^{10} \text{ s}^{-1} Z^4 p^{-4.5} \sqrt{1 - 2/p}, \tag{6.12}
\]

valid for \( p > p_1 \approx 2.6 \) for the relevant ions.

In the corona domain, the relative overpopulation of an excited state compared to the Saha density (corresponding to Saha equilibrium with the ground state of the next ion) is now given by

\[
 b(p) - 1 = (b_1 - 1) \max \left( 2.3 \times 10^{-23} n_e \frac{p_1^5}{Z^6 y^4 \sqrt{T_e} p (1 - 2/p)}, 1 \right). \tag{6.13}
\]

This expression is a strongly decreasing function of the excitation energy for the energy levels and plasma conditions of interest in this work, so that radiating levels with lower energies are favored over levels with higher energies, compared to a Boltzmann (or Saha) distribution.

In ESB \( (p > p_{cr}) \), the relative overpopulation to the Saha density is set proportional to \( p^{-6} \). The prefactor is adjusted such that the ESB density matches the corona density for \( p = p_{cr} \). In Fig. 6.1, the positions of the corona and ESB domains are indicated in a schematic plot of the densities per statistical weight as a function of the excitation energy.
6.2 Spectral model

Fig. 6.1. A schematic plot of the densities per statistical weight as a function of the excitation energy. The positions of the Corona and ESB domains are indicated relative to \( p_{cr} \). The arrows at the top indicate the directions of the ionization and recombination processes, as explained in Sec. 6.2.2.

6.2.4 Radiation

The calculation of the broadening of each individual line was adjusted to include the effects of natural and Doppler broadening. The line profile is now calculated as a pseudo-Voigt profile in which the Gaussian contribution corresponds to Doppler broadening and the Lorentzian part accounts for natural and Stark broadening. For the half width at half maximum of the Stark broadening contribution, the approximation of Eqs. (24) and (25) in Ref. [28] has been applied, which is given in frequency units by

\[
\begin{align*}
\omega_{se} &= 8 \left( \frac{\pi}{3} \right)^{3/2} \frac{\hbar}{m_e a_0} \eta_e \left( \frac{R y}{k_B T_e} \right)^{1/2} \\
&\times \left[ \left\langle i \mid r^2 \mid i \right\rangle \bar{\eta}_{se} \left( \frac{3k_B T_e}{2|\Delta E_i|} \right) + \left\langle f \mid r^2 \mid f \right\rangle \bar{\eta}_{se} \left( \frac{3k_B T_e}{2|\Delta E_f|} \right) \right], \\
&\text{with} \\
\left\langle x \mid r^2 \mid x \right\rangle &= \frac{p_x^2}{2z^2} \left[ 5p_x^2 + 1 - 3l_x (l_x + 1) \right] \alpha_0^2. \\
\end{align*}
\] (6.14a)

Here, \( x = i, f \) denotes either the initial or final state of the transition and \( \bar{\eta}_{se}(\xi) \) is an effective Gaunt factor, evaluated as

\[
\bar{\eta}_{se}(\xi) = \max \left( 0.2, \frac{\sqrt{3}}{2\pi} \ln \xi \right). \\
\] (6.15)
As an order-of-magnitude approximation of the energy distance to the nearest perturbing level the expression \( |\Delta E| = \frac{1}{2} Z^2 \text{Ry} \left[ \frac{1}{p^2} - \frac{1}{(p + 1)^2} \right] \) is used, where the prefactor \( \frac{1}{2} \) is included to account for the fact that compared to a purely hydrogenic ion, the distance to the nearest perturbing level will be reduced due to the fine structure in combination with the \( l \)-dependent quantum defect. We believe that such an approximation is sufficiently accurate because \( \overline{\gamma}_n(\xi) \) is only a very weak function of its argument.

To evaluate Eq. (6.14a) for every line, the \( l \) value of each relevant energy level has been included in the atomic data input files to the model.

The cutoff procedure for calculation of opacity effects was replaced by a somewhat more sophisticated method, in which the part of the radiation that escapes the plasma approaches the blackbody limit exponentially at each wavelength as the total emitted radiation increases. Again, the blackbody limit was evaluated over the entire surface area of the plasma, but the geometry of the plasma was changed from spherical to cylindrical, to better match the shape of a discharge plasma. Finally, the resulting emission can be convoluted with a Lorentzian-shaped spectrometer profile for easier comparison with experimental results.

### 6.2.5 Atomic data

A faithful representation of the EUV spectra of both xenon and tin requires detailed information on the fine structure of energy levels of their ions, and transition probabilities for the main optical transitions between the excited and ground states. Such information was obtained from atomic data calculations using the COWAN package [29].

For both xenon and tin, the 8+ to 12+ ions are the main contributors to the EUV spectrum in the 10–21-nm-wavelength range. For these ions, apart from the \( 4d^n \) ground state configuration, also the \( 4d^{n-1}s, 6s, 5d, 5p, 6p, 4f, \) and \( 5f \), and \( 4p^54d^{n+1}(pd) \) configurations were included in the calculations. Exceptions are Xe\(^{8+} \), for which the \( pd \) configuration does not exist due to a full \( 4d \) shell, and Xe\(^{12+} \), for which the \( 6s \) and \( 6p \) configurations were omitted. On the other hand, the \( 7p \) configuration was also included for Sn\(^{8+} \) and Xe\(^{8+} \), the \( 6f \) configuration was included for Xe\(^{8+} \), and the \( 6d \) configuration was included for Xe\(^{8+},9+ \). Similar calculations have also been performed for lower and higher stages of both elements. However, since the simulations show that these play minor roles in the plasmas, these are not discussed in detail here.

Relativistic terms were included in the calculations, and the Coulomb integrals were scaled down to 0.85 times their original values to account for weak interactions with other configurations.

To reduce file sizes and save calculation time, some of the weakest optical transitions were removed. All lines with an optical transition probability \( gA \) larger than \( 10^8 \) s\(^{-1} \) and \( 3\times10^9 \) s\(^{-1} \) for xenon and tin, respectively, were retained, and saved to input files for the radiation module described above. For all ions, the transition probability of the strongest line was at least about four orders of magnitude larger than the cutoff value.
6.3 EUV Spectrometer sensitivity

6.3.1 Theoretical dependencies on wavelength

In the ASML EUV laboratory, a grazing incidence vacuum ultraviolet (VUV) spectrometer from ISAN, Troitsk (Russia), combined with a multichannel plate (MCP) detector and a digital camera, has been used in the experiments for recording of EUV spectra. In this section, first the theoretical considerations regarding the wavelength dependency of the sensitivity of this spectrometer will be briefly discussed, and a theoretical sensitivity curve will be constructed. After that, this curve will be compared to the results of an experimental wavelength-dependent sensitivity calibration.

The grating that was used for the recording of the EUV spectra has a ruling of 1200 l/mm and a blaze angle of 1°. It consists of a substrate covered with a 60 nm thick gold coating and was used at a grazing angle of incidence of 4° and an angle of refraction of about 10° (depending on the wavelength). In the calculation of the theoretical efficiency of the spectrometer, four different factors should be taken into account:

i. The geometrical efficiency of the spectrometer, given the groove density, the angle of incidence (and, connected to that, the angle of refraction), and the blaze angle,

ii. the EUV reflectivity of gold at a grazing incidence angle of 5°,

iii. the inverse wavelength dispersion at the surface plane of the MCP detector (a larger spread of the signal will result in a lower signal per camera pixel), and

iv. the wavelength-dependent sensitivity of the MCP.

The latter depends on the responsivity of the photocathode of the MCP, onto which a thin layer of gold has been deposited. From the literature on this subject [30], it is known that the gold photocathode efficiency strongly depends on the degree of surface contamination. Since usually no special measures are taken in the handling of the MCP (for example, it is regularly exposed to air between experiments, and pumped down to a vacuum of only about $1 \times 10^{-5}$ mbar, without any bake-out procedures), a contaminated surface was assumed. The quantum efficiency was copied from the data of Ref. [31] as quoted in Ref. [30].

The gold reflectivity was taken from the CXRO website on X-ray interactions with matter [32]. The geometrical efficiency combined with the inverse wavelength dispersion, the gold reflectivity, and the MCP efficiency for the EUV range are shown in part (a) of Fig. 6.2. The combined effect of all three factors can be divided by the incident photon energy to get the total (relative) energy conversion efficiency as shown in part (b) of Fig. 6.2.
Fig. 6.2. (a) Geometrical spectrometer efficiency (dashed curve), $5^\circ$ grazing-incidence gold reflectivity (dotted curve), and MCP efficiency (solid curve). (b) The total energy conversion efficiency of the spectrometer as a function of wavelength.

6.3.2 Experimental validation

The calibration of the wavelength dependency of the spectrometer for the EUV range was carried out by comparing pulse-integrated EUV spectra from the hollow cathode source with signals from a copy of the Flying Circus (FC) tool [33]. The original FC tool was designed in cooperation between the research organization FOM and the companies Philips and ASML, with the goal to set a standard for the measurement of in-band powers of EUV sources from different potential suppliers, so that a reliable comparison between those sources could be made. The name of the tool was derived from the fact that it was actually transported to various locations for measurements. In the FC tool, (EUV) radiation is wavelength filtered by reflection off a multilayer mirror and transmission through a thin foil filter, before it is collected on a photodiode. The FC has two channels. In our experiments, one of them was always equipped with a curved multilayer (ML) mirror designed for 13.5 nm wavelength. In the mirror holder for the second channel, for each individual measurement a different flat ML mirror from PhysTex, that was optimized for a different wavelength, was placed. In total, measurements for eight different wavelengths have been done.

For each measurement, the pulse-integrated signals for both photodiodes were averaged over four discharge pulses of the source. Simultaneous to the measurement of the FC signal, a spectrum for the same pulses was recorded using the EUV spectrometer. The MCP was gated on a time scale long enough to record the total EUV emission for each pulse. The recorded spectrum was processed in the same way as in previous experiments.
Then, the spectrum was multiplied at each wavelength with the known photodiode sensitivities, filter transmission, mirror reflectivities, and aperture cross sections for both channels of the FC tool. Next, the results for both channels were integrated over wavelength. The resulting numbers are

\[ I_{c1} = \pi r_1^2 \int_{10}^{21} J_\lambda (\lambda') R_1 (\lambda') T (\lambda') \eta_{\text{diode},1} (\lambda') d\lambda', \]  
\[ I_{c2,\lambda} = \pi r_2^2 \int_{10}^{21} J_\lambda (\lambda') R_2 (\lambda') T (\lambda') \eta_{\text{diode},2} (\lambda') d\lambda'. \]  

(6.16a)

(6.16b)

Here, \( r_1 \) is the radius of the curved mirror aperture, \( r_2 \) is the filter radius of the second channel, \( T \) is the transmission of each filter, and \( R_i \) and \( \eta_{\text{diode},i} \) are the mirror reflectivity and the diode energy conversion efficiency of channel \( i \) (\( i = 1, 2 \)), respectively. \( J_\lambda (\lambda') \) are the raw spectrometer data, where \( \lambda' \) is given in nm. \( I_{c1} \) and \( I_{c2,\lambda} \) represent the calculated signals of the first and second channel, respectively. Now, if the spectrometer would have a flat sensitivity curve, the ratio of these two numbers would be equal to the ratio of measured FC signals. The difference between the two ratios therefore gives information about the relative sensitivity of the EUV spectrometer, in the following way,

\[ \frac{I_{c2,\lambda}}{I_{c1}} = \frac{\eta_\lambda}{\eta_{13.5 \text{ nm}}} \frac{I_{m2,\lambda}}{I_{m1}}, \]  

(6.17)

where \( I_{m1} \) and \( I_{m2,\lambda} \) represent the measured values from the first and second channel, respectively, and finally \( \eta_\lambda/\eta_{13.5 \text{ nm}} \) is the spectrometer efficiency at wavelength \( \lambda \), relative to the sensitivity at 13.5 nm.

To partially eliminate some uncertainties in the filter transmissions and diode sensitivities, the end results were normalized using measurements in which both mirror holders of the FC held 13.5 nm mirrors. The results obtained in this way are summarized in Fig. 6.3. The square blocks represent the measured data; for the open squares, the two photodiodes were exchanged compared to the solid squares. The solid curve in the figure represents the theoretically constructed sensitivity curve as discussed above, normalized to unity at 13.5 nm. As the plot shows, there is a quite large spread between the sensitivity values of the individual data points. However, the experimental data do seem to confirm the general trend of the theoretical prediction; at least no large systematic deviation from the theoretical curve can be detected. The relatively large differences between individual data points and the theoretical curve can probably be ascribed to the large uncertainties in filter transmission, diode sensitivity and especially the mirror reflectivity. The experimental results in the remainder of this chapter have been corrected using the theoretical curve as described above.
6.4 Simulated EUV spectra

For both the tin and xenon discharge plasmas, three different spectra have been selected from existing experimental results to be used as a basis for matching the model simulations. In the case of the xenon discharge, the same set of data as discussed in Chapter 4 has been applied. These are spatially integrated, but time-resolved, spectra recorded from the axial direction of the electrode geometry. The three spectra, to be referred to as (A), (B), and (C), correspond to timings roughly 20, 10, and 0 ns before the pinch. For the tin discharge, three spectra were selected from the data discussed in Chapter 5. These are space- and time-resolved spectra, recorded from a direction perpendicular to the axis of symmetry of the discharge. The signals from an area between 0.1 and 0.45 mm from the cathode have been integrated. The spectra (D), (E), and (F) were recorded for timings of 40, 20, and 0 ns before the pinch. In both cases, signals from multiple pulses (30 and 10 for the xenon and tin discharges, respectively) were added to obtain more accurate results.

An automated fitting procedure of the model results to the experimental data would have become far too involved; also, the accuracy of the experimental results would not always have allowed for such a procedure. Therefore, an alternative approach was followed. For the model calculations, we started with certain initial guesses for the ion density, electron temperature, plasma geometry, and the effective rate of ionization $\nu_i$, derived from pinhole images (Chapters 4 and 5) and Thomson scattering (Chapter 8) results,
where available. Next, these values were varied until a reasonable agreement, to the eye, between simulation and experiment was achieved. In doing so, however, the length of the plasma cylinder was not changed. Also, the values for the total ion density and the plasma radius were chosen such that the total number of ions in each plasma was roughly preserved for the different spectra of each type of discharge; the applied values were about $1.3 \times 10^{14}$ and $6 \times 10^{13}$ for the xenon and tin plasmas, respectively. In other words, the total number of ions in each type of plasma could only be changed for all three spectra simultaneously. This left just three parameters to be varied independently for each spectrum: the electron temperature $T_e$, the effective ionization rate $\nu_i$, and the plasma radius $r$.

The optimized simulated spectra will be referred to by the lower case equivalents (a)–(f) of the experimental spectra names (A)–(F). They are presented in Figs. 6.4 and 6.5 respectively. Even though the intensity units are in principle arbitrary, the same multiplication factors were used for different parts of each graph. Here, the simulated spectra were convoluted with a spectrometer profile with a full width at half maximum of 0.08 nm to make the model results better comparable to the experimental data. To demonstrate the effect of this convolution, the unconvoluted and convoluted versions of spectrum (f) are shown in Fig. 6.6 together with the blackbody radiation curve for the given plasma temperature and geometry. It can be seen that even though the convoluted spectrum stays well below the blackbody limit at all wavelengths, in the actual spectrum many lines do reach the blackbody limit.

For spectra (b) and (e), the contributions of the bound-bound radiation of the individual ions to the simulated spectra are plotted separately in Figs. 6.7 and 6.8 respectively. The individual contributions may not add up exactly to reproduce the spectra of Figs. 6.4 and 6.5, since in Figs. 6.7 and 6.8 first of all free-free and free-bound radiation are not included, and second, opacity effects were only considered for the individual contributions separately, so that overlaps of lines from different ions were not accounted for. As the figures show, for both xenon and tin the features corresponding to $4d-4f$ and $4p-4d$ transitions (around 11 and 14 nm, respectively) tend to overlap for different ionization stages, but on the other hand, the $4d-5p$ arrays of the different ions are well separated, which makes identification of the different ions from an experimental spectrum possible. The contribution of Sn$^7^+$ is not shown in Fig. 6.8, but the COWAN calculations show that the $4d-5p$ feature of this ion has its peak between 22 and 23 nm.

Details on the applied input parameters and the resulting electron densities and total radiation are given in Table 6.1. These data suggest electron temperatures in the range of 23–27 eV for both plasmas, and electron densities up to about $1 \times 10^{25}$ and $3 \times 10^{25}$ m$^{-3}$ in the pinch phase for the xenon and tin plasmas, respectively.

An example of the effect of variation of parameters in general is given in Fig. 6.9. The parameters of the alternative simulated spectra of Fig. 6.9 are included in Table 6.1.

For the simulated spectra corresponding to the time of the pinch, the distributions over the ion stages are as follows: 2.6% of 8+, 28% of 9+, 51% of 10+, 16.9% of 11+, and 1.07%
Fig. 6.4. Comparison of experimental xenon spectra (A), (B), and (C), as defined in the text (dashed curves) with simulation results (a), (b), and (c) (solid curves). The units for the scales of the three different plots are equivalent, and the ratios between the scales of the experimental and simulated results were kept fixed.
Fig. 6.5. Similar plots as in Fig. 6.4 but for the spectra of the tin discharge. Solid curves: simulated spectra (d), (e), and (f); dashed curves: experimental data (D), (E), and (F).
Fig. 6.6. Simulated spectrum (f) for the tin discharge, before (grey curve) and after (solid curve) convolution with a hypothetical spectrometer profile of 0.08 nm full width at half maximum. The dotted curve represents the blackbody limit under the applied input parameters of the model.

Fig. 6.7. The line radiation contributions of the individual ions $\text{Xe}^{7+}$ (grey curve with open circles), $\text{Xe}^{8+}$ (dashed), $\text{Xe}^{9+}$ (solid), $\text{Xe}^{10+}$ (dotted), and $\text{Xe}^{11+}$ (grey curve with crosses) to the simulated xenon spectrum (b). The $\text{Xe}^{7+}$ and $\text{Xe}^{11+}$ curves have been magnified 15 times to make them visible on the same scale as the other ones. The solid horizontal line at the top of the graph shows the approximate position of the different $4d-4f$ contributions, whereas the $4d-5p$ contributions are indicated by the dashed line.
6.4 Simulated EUV spectra

Fig. 6.8. The individual ion contributions of Sn$^8^+$ (dashed curve), Sn$^9^+$ (solid), Sn$^{10^+}$ (dotted), and Sn$^{11^+}$ (grey curve with crosses; magnified two times with respect to the other spectra) to the simulated tin spectrum (e). The solid horizontal line at the top of the graph shows the approximate position of the different 4d-4f contributions, whereas the 4d-5p contributions are indicated by the dashed line.

Table 6.1. The input parameters and certain characteristics of the simulation results corresponding to the spectra shown in Figs. 6.4 and 6.5. Here $n_i$ represents the total ion density, $T_e$ is the electron temperature, $\nu_i$ is the effective ionization rate, and $r$ and $l$ are the radius and length of the cylinder-shaped plasma, respectively. Further, $z_{av}$ is the average ionization degree, $n_e = n_i z_{av}$ is the electron density and $I_{tot}$ represents the total energy emitted by the plasma in 5 ns time, in the wavelength range under consideration (10–21 nm for Sn, 10–19 nm for Xe).

<table>
<thead>
<tr>
<th>Name</th>
<th>$n_i$ ((10^{23} \text{ m}^{-3}))</th>
<th>$T_e$ (\text{(eV)})</th>
<th>$\nu_i$ (\text{(10}^7 \text{ s}^{-1})</th>
<th>$r$ (\text{(mm)})</th>
<th>$l$ (\text{(mm)})</th>
<th>$z_{av}$</th>
<th>$n_e$ (\text{(10}^{25} \text{ m}^{-3})</th>
<th>$I_{tot}$ (\text{(mJ)})</th>
</tr>
</thead>
<tbody>
<tr>
<td>Xe (a)</td>
<td>1.35</td>
<td>24.5</td>
<td>4</td>
<td>0.55</td>
<td>1</td>
<td>8.32</td>
<td>0.112</td>
<td>3.0</td>
</tr>
<tr>
<td>Xe (b)</td>
<td>2.6</td>
<td>27</td>
<td>4.5</td>
<td>0.4</td>
<td>1</td>
<td>9.18</td>
<td>0.24</td>
<td>10.1</td>
</tr>
<tr>
<td>Xe (c)</td>
<td>10</td>
<td>25.5</td>
<td>1</td>
<td>0.2</td>
<td>1</td>
<td>9.86</td>
<td>0.99</td>
<td>24</td>
</tr>
<tr>
<td>Sn (d)</td>
<td>0.78</td>
<td>24</td>
<td>1.5</td>
<td>0.7</td>
<td>0.5</td>
<td>8.54</td>
<td>0.067</td>
<td>2.3</td>
</tr>
<tr>
<td>Sn (e)</td>
<td>3.1</td>
<td>23</td>
<td>3</td>
<td>0.35</td>
<td>0.5</td>
<td>9.21</td>
<td>0.29</td>
<td>9.1</td>
</tr>
<tr>
<td>Sn (f)</td>
<td>31.5</td>
<td>23</td>
<td>5</td>
<td>0.11</td>
<td>0.5</td>
<td>9.73</td>
<td>3.1</td>
<td>36</td>
</tr>
<tr>
<td>Sn (f1)</td>
<td>31.5</td>
<td>21</td>
<td>5</td>
<td>0.11</td>
<td>0.5</td>
<td>9.12</td>
<td>2.9</td>
<td>29</td>
</tr>
<tr>
<td>Sn (f2)</td>
<td>31.5</td>
<td>25</td>
<td>5</td>
<td>0.11</td>
<td>0.5</td>
<td>10.32</td>
<td>3.3</td>
<td>41</td>
</tr>
</tbody>
</table>
Fig. 6.9. Experimental tin spectrum (F) (dotted curve), and the corresponding simulation result (f) (solid curve) for $T_e = 23$ eV. For comparison, alternative simulation results with $T_e = 21$ eV (f1) (solid curve with open circles) and 25 eV (f2) (solid curve with crosses) have been included. Spectra (f1) and (f2) match the absolute intensity and the ratio of 13.5- and 17.5-nm peak intensities less well than the simulated spectrum (f), and they give incorrect ratios of intensities of the spectral features at 16 and 17.5 nm, indicating that the average ion charges for those simulations are wrong.

of 12+ for xenon; and for tin, 0.14% of 7+, 4.6% of 8+, 33% of 9+, 48% of 10+, 13.9% of 11+, and 0.80% of 12+. All the other ions give contributions of less than 0.1%. Since by far the most EUV radiation in each case is emitted by the plasma at the time of pinching, these simulations confirm that the ions mentioned above are the main contributors to the overall emission of both plasmas.

6.5 Discussion and conclusions

6.5.1 Comparison of experimental and simulated spectra

Figures 6.4 and 6.5 show that there is quite good agreement between different aspects of the experimental results and the model calculations. This is especially true for the shapes and positions of the individual features of the spectra, which are basically a direct result of the atomic data calculations. In general, in these atomic data calculations the lines appear at slightly lower wavelengths than in the experiments. The effect seems to be the largest for the $4d-4f$ transitions of different ions. This is not much different from what has been reported earlier [16, 35–37].

Also, the absolute intensities of the simulated spectra compare well to experimental data. There is no direct absolute calibration available for the spectrometer, but the simulated in-band radiation (emitted in a 2% wavelength band around 13.5 nm) can be compared
6.5 Discussion and conclusions

to power measurements that use a photodiode behind a silicon/niobium (Si/Nb) filter and a multilayer mirror that also acts as a wavelength filter, such as in the Flying Circus in the case of the xenon source. Such measurements show that the total emitted in-band radiation per pulse is about 60 mJ for the tin source and 25 mJ for the xenon source, under the settings applied in this work. The simulated spectra for the pinch phase show in-band emission of 5.7 and 2.0 mJ, respectively, in 5 ns time. As experiments show that the typical effective durations for in-band emission are about 30 and 50 ns, respectively (taking experimental timing jitter into account), it can be concluded that the absolute in-band emission of both plasma EUV sources is reproduced rather well. The differences could be explained by the fact that the actual radiating volumes of plasma are somewhat larger than those assumed for the simulations.

Finally, the ratio of emission intensities of the $4d-4f$ features compared to the $4d-5p$ lines in the experiments is reproduced fairly well in the simulations. The COWAN code produces optical transition probabilities that are much larger for the $4d-4f$ lines than for the $4d-5p$ ones. Our calculations show that this difference is balanced for the largest part by stronger opacity effects for the $4d-4f$ lines, and the applied CRM, which relatively favors the population of the $5p$ levels over the $4f$ ones compared to a Boltzmann distribution. Still, the relative $4d-4f$ intensities tend to be overestimated somewhat by the model; see the discussion below for possible explanations.

All this having been said, there are also some differences between the simulated spectra and the experimentally obtained ones. One notable effect that has been reported previously by other workers [16] is also seen here. This is the fact that, compared to the simulations, the experimental spectra seem to exhibit an additional broad, (quasi)continuum emission background that contributes strongly to the total emission in particular between the main emission features. Even though some of the weakest optical transitions were left out of our calculations, these are not strong enough to account for the difference. Another possible cause could be the underestimation of free-bound radiation in our work. However, our calculations show that the much (about 30–100 times) stronger free-bound contribution that would be needed to explain the observed difference, would also lead to a large emission feature at wavelengths below the main $4d-4f$ emission peak for both elements. Such a feature has not been observed experimentally. A third explanation might be radiation emitted from doubly excited states. Such states have not been included in our calculations.

More generally, certain limitations in the model prohibit a better agreement between the simulations and the experiments. First of all, spatial variations in density, temperature, and optical density in the actual plasma can lead to observed spectra that cannot be simulated under the assumption of perfect homogeneity of the plasma. A zone of cooler plasma around the strongest EUV emitting region might, for example, absorb the $4d-4f$ radiation relatively more than the $4d-5p$ part. Also, a jitter in the timing of the experiments can lead to contributions of plasma with different properties to the same observed spectrum. Such an effect might, for example, have caused the apparently relatively strong $4d-5p$ emission,
which could be due to cooler, and hence more optically dense plasma, in spectrum (e) of the tin discharge. And finally, even though opacity of the plasma has been taken into account for the calculation of the radiation, its effect on the densities of excited states (a certain shift from corona towards LTE balance, due to the decreased importance of radiative decay) has not been included. In view of the fact that both plasmas appear to be only partially optically dense, and the reasonably good results obtained with the current model, we believe that the error in the produced spectra made due to this omission is not very large.

A fundamentally different and far more elaborate approach would be required for a further improvement of the results. This approach would include a time- and ideally also space-dependent model of the evolution of the plasma, and a calculation of the population density of each excited state based on detailed information on optical transition probabilities, reabsorption of radiation and electronic (de)excitation cross sections, rather than using analytical expressions that depend on the excited state energy only. However, such an approach was beyond the scope of this work.

### 6.5.2 Validity of the single electron temperature approach

In Sec. 6.2 we mentioned that we assume the electron energy distribution function (EEDF) to be Maxwellian. Here we will verify this assumption by discussing the influence of the main equilibrium disturbing processes for both extreme cases of lowest [spectra (A) and (D)] and highest [spectra (C) and (F)] electron densities. In discharge plasmas such as we are considering, the strongest processes to disturb a Maxwellian EEDF would be expected to be excitation of ions and acceleration due to the external electric field.

First we will evaluate the importance of ion excitation by comparing it to the equilibrium restoring process of energy redistribution due to electron-electron collisions. A good measure for the net effect of excitation of ions is the amount of emitted radiation, since the amount of energy “lost” to radiation is much larger than the amount of energy that is actually stored in the plasma in the form of excitation and/or ionization. From our experiments and simulations, it can be derived that the photon emission rates per free electron are roughly $5 \times 10^7 \text{s}^{-1}$ and $5 \times 10^8 \text{s}^{-1}$ for the lowest and highest density cases, respectively.

For the characteristic electron-electron collision rate for energy transfer, we use the equation for the case of a near-Maxwellian EEDF [38], given in numerical form by

$$
\nu_e = 2.9 \times 10^{-12} \text{ m}^3 \text{ eV}^{3/2} \text{ s}^{-1} n_e \Lambda T_e^{-3/2},
$$

where the value of the Coulomb logarithm $\Lambda$ is about 6 for the plasmas under consideration. Now, $\nu_e$ is between $1 \times 10^{11} \text{s}^{-1}$ and $2 \times 10^{11} \text{s}^{-1}$ and between $1 \times 10^{12} \text{s}^{-1}$ and $5 \times 10^{12} \text{s}^{-1}$ for the low and high density cases, respectively. In both cases, the equilibrium restoring process is at least three orders of magnitude faster than the disturbing one, so that ion excitation is not capable of causing any significant deviation from a Maxwellian EEDF.
The importance of the external electric field can be derived from the magnitude of the electric current that is caused by it. From experiments as well as source design parameters, we know that the maximum current during the evolution of the discharge is about 20 kA. When we ignore the contribution of the ions, and divide this number by the elementary charge and the linear electron density in the direction of the current—which is about $10^{18}$ m$^{-1}$ in all cases—we find an average directed electron velocity on the order of $10^5$ m s$^{-1}$.

The kinetic energy associated with this velocity, about 0.03 eV, is negligible in comparison with the electron temperatures that we have found. Therefore, also the external electric field in the plasma does not lead to a significant deviation from a Maxwellian EEDF, and it can be concluded that, at least for the spectra that are the subject of this work, a description of the EEDF by a single electron temperature $T_e$ is justified.

### 6.5.3 Plasma properties

Considering the uncertainties in the spectrometer calibration, the aforementioned inherent limitations in the model, and the limited accuracy with which the simulated spectra could be matched to the experimental ones, the comparison of experimental and simulated spectra should not be viewed as a way to obtain high accuracy data on plasma parameters such as electron densities and temperatures. However, it is a useful method to derive certain estimates for those parameters. More importantly, it is a way to evaluate which phenomena play an important role in determining the shapes and intensities of the EUV spectra.

It should be noted that it has proven possible to obtain reasonable agreement between experimental and simulated spectra using realistic values for all plasma parameters. The plasma dimensions match the results from pinhole imaging experiments fairly well, and the densities were chosen such that the total number of ions in the plasma remains roughly constant during the pinch evolution. For the xenon discharge, the initial ion density before pinching roughly corresponds to the background gas density if the initial radius of the plasma is taken to be about 4.5 mm, which is a realistic value given the geometry of the electrodes.

For spectra (d) and (e) of the tin discharge, the electron temperatures and densities can be compared to the results of Thomson scattering (TS) experiments discussed in Chapter 8 [34]. The densities given there, about $4 \times 10^{23}$ and $1 \times 10^{24}$ m$^{-3}$, respectively, are somewhat lower than the results presented here for the same time in the discharge evolution. However, the experimental jitter and the fact that the EUV emission increases strongly with time in this phase of the discharge may be responsible for this. Effectively, the EUV spectra will be more representative of the emission somewhat later in the discharge evolution, when the electron density is higher.

The electron temperatures derived from TS increase from about 17 to over 30 eV in the relevant time interval. The temperatures in Table 6.1 are in the same range, although they are much more constant as a function of time. It is worth mentioning that the
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TS measurements give three-dimensional spatially resolved data, whereas the spectrum simulations are based on measurements in which radiation from a certain cross section of the plasma is integrated, which might affect the effective temperatures. For the actual pinch phase of the tin discharge, no experimental data are available.

The values of the input parameter \( \nu_i \) are in agreement with the rate of increase of the average ion charge \( z_{av} \) to within roughly a factor 2. The difference for xenon is an indication that perhaps the ionization coefficient used in this model is still somewhat too low, in spite of the adjustments that have been introduced in Eq. (6.3).

The electron temperatures that were needed to get good agreement between the simulations and the experiments, are not extremely high: \( T_e \) was lower than 30 eV in all cases. Leaving out the ionization effect (i.e., setting \( \nu_i \) equal to zero), the electron temperatures required to obtain the same average ion charges, keeping all other input parameters the same, vary from 18 to 25 eV. The difference to the actual electron temperature decreases from about 6 to less than 1 eV, when going from the first to the last spectrum of each plasma. In other words, ionization effects should be taken into account; however, our results seem to indicate that they are not very strong, and the electron temperature is never more than a few eV higher than in the equilibrium case for the same average ion charge.

The importance of opacity to practical applications seems to depend on the type of plasma. As was mentioned above, the \( 4d-4f \) lines were affected the most by reabsorption in the plasma in our model. For the xenon pinch plasma, also the intensity of the Xe\(^{9+}\) \( 4d-5p \) feature around 15 nm was strongly reduced by opacity. On the other hand, the \( 4d-5p \) emission of Xe\(^{10+}\) around 13.5 nm was relatively unaffected. This is an indication that dilution of the plasma might perhaps be helpful to increase the energy conversion efficiency for the tin discharge, but not so much for the xenon discharge. Such a dilution would reduce the amount of energy needed to run the plasma, while keeping the level of emitted in-band radiation actually escaping from the plasma at a (nearly) constant level. It is only useful when the plasma is optically dense for at least a part of the wavelength range of interest.

Summarizing, we have shown that atomic and plasma physics calculations can result in fairly good reproduction of experimentally observed EUV spectra of discharge plasmas. To obtain a reasonable agreement, however, several factors have proven to be essential. These include first of all a correction for the wavelength dependency of the spectrometer sensitivity; and further an (albeit rough) treatment of ionization effects; a non-LTE approach to the population of the excited states of the ions; and a good description of the broadening mechanisms for the spectral lines, to account for opacity effects (especially during the pinch phase) in a correct manner.

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1 This was still true at the time of writing. However, newer TS experiments have given a confirmation of the electron density in the pinch phase. See Chapter 10 and the discussion in Chapter 11.
6.6 Addendum: Larger wavelength spectra

The validity of the model used in this chapter to simulate the spectra of xenon and tin pinch plasmas, is in principle not limited to the EUV wavelength range. Provided that the input files contain all relevant optical transitions, it can also be used to simulate the spectra at both smaller and larger wavelengths. As an example, we have taken the input parameters as presented in Table 6.1 for both the xenon and tin plasmas, and we have used them to calculate spectra in the extended range of 2 to 1000 nm. The results are presented in Figs. 6.10 and 6.11 respectively.

All relevant ionic configurations needed to simulate these spectra were already taken into account in the original atomic data calculations as discussed in Sec. 6.2.5. However, originally, only the optical transitions in the limited wavelength range of 10–19 nm (xenon) or 10–21 nm (tin) needed to be available, and all other transitions were omitted from the final atomic data files. Now they were needed for a much larger wavelength range. Still, to prevent the input files from becoming impractically large for certain ions, some of the weakest transitions had to be omitted. Optical transitions tend to have larger transition probabilities for smaller wavelengths. Hence, in order to retain the strongest transitions for each part of the spectrum, we chose to base the criterion for keeping or removing a specific transition not just on the $gA$ value, but rather on the value of $\lambda^2 gA$. The limiting values were $10^{11}$, $10^{12}$, $4 \times 10^{12}$, and $5 \times 10^{12}$ nm$^2$s$^{-1}$ for Xe$^{9+}$ to Xe$^{12+}$, and $10^{12}$, $4 \times 10^{12}$, $6 \times 10^{12}$, $5 \times 10^{12}$, $2 \times 10^{12}$, and $10^{11}$ nm$^2$s$^{-1}$ for Sn$^{6+}$ to Sn$^{11+}$, respectively.

Further, the spectra as presented in Figs. 6.10 and 6.11 were produced in four parts each, in order to select an appropriate number of data points per unit of wavelength for each part of the spectrum. They were later combined to be presented in a single figure for each element. The full width at half maximum of the spectrometer profile was, somewhat arbitrarily, kept constant at 0.08 nm over the entire wavelength range.

From the spectra presented in Figs. 6.10 and 6.11 several conclusions can be drawn. First of all, they show that the pinch spectra are dominated by line radiation between about 8 and 200 nm (xenon) and between roughly 10 and 100 nm (tin). At smaller wavelengths, the emission is dominated by free-bound radiation, while at larger wavelengths, free-free radiation (bremsstrahlung) is the main emission process. The equations for free-free and free-bound radiation can be found in the work of Garloff et al. [18]. Although these contributions were also taken into account for the spectra presented in Sec. 6.4, they are negligible compared to line radiation for the EUV wavelength range. These conclusions are supported by closer inspection of the raw data, as produced by the model, which are split up by type of radiation. Additionally, both plots show that the pinch plasmas are optically open in the ultraviolet and visible wavelength ranges, which is of great importance for optical diagnostics at those wavelengths.

Unlike the EUV emission, the pulse-integrated emission of the plasma in the ultraviolet
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Fig. 6.10. Extended-wavelength simulations of the xenon spectra with input parameters as given in Table 6.1. Solid lines (from light to dark): simulated spectra (a), (b) and (c) for 20, 10, and 0 ns before the pinch; dotted lines: Planck limits for the same simulations (with matching greyscales).

Fig. 6.11. Extended-wavelength simulations of the tin spectra with input parameters as given in Table 6.1. Solid lines (from light to dark): simulated spectra (d), (e) and (f) for 40, 20, and 0 ns before the pinch; dotted lines: Planck limits for the same simulations (with matching greyscales).
and visible wavelength ranges is strongly affected by emission from cooler phases of the discharge. Most notably, the relatively large and high density, but cool plasma in the early decay phase of the tin vapor discharge, as discussed in Chapter \[10\] \[39\], can contribute strongly to this pulse-integrated emission. Hence, the relative contribution of non-EUV emission to the total pulse-integrated radiated energy will be larger than suggested by Figs. 6.10 and 6.11. To evaluate this contribution properly, the spectra would need to be simulated as a function of time, in sufficiently small steps, for the entire discharge. For this, two types of supplementary information are needed. First of all, sufficiently accurate input parameters as a function of time are required. Such data could, for example, be derived from the Stark broadening data of Chapter \[7\] \[40\] and the Thomson scattering results as presented in Chapter \[10\]. Second, experimental data over a sufficiently large part of the spectrum are needed to serve as a check of the validity of the model for the cooler phases of the discharge. Unfortunately, such data are not available at present.

Additionally, if detailed spectrally resolved information is needed, the atomic data will need to be reevaluated. The energy level calculations used in this work are accurate enough to quite reliably predict the positions of spectral lines in the EUV, but the energy level errors lead to relatively large wavelength errors for lines in the visible part of the spectrum.

### 6.7 Addendum: The contribution of doubly excited states

#### 6.7.1 The UTA formalism

In Sec. 6.5.1, it was mentioned that compared to the simulated spectra, the experimental ones exhibit an additional broad, quasicontinuum emission background. This effect is remarkable since it also occurs at wavelengths where no strong optical transitions appeared to be present at all. Hence, unlike any other mismatches in the spectra, it could not be explained merely by uncertainties of the model, e.g., in the population densities, opacity calculations, or due to plasma inhomogeneity. Instead, an entirely different mechanism appears to be responsible. It was already mentioned in Sec. 6.5.1 that this mechanism could be radiation from doubly excited states. In this section, we look more closely at the possible role of this special class of states.

The main difficulty with modeling doubly excited states is their enormous number. In a doubly excited state, there are two excited electrons instead of just one, and the number of possible combinations of individual electron states is such that they cannot all be treated individually in a spectral model such as ours. Therefore, an alternative approach is required to include doubly excited states in the model.

Sasaki et al. \[41\] used a so-called UTA formalism to include “satellite lines” from multiply excited states in their simulated spectra of laser produced plasmas. A similar approach is followed here to evaluate the contribution of doubly excited states to the spectra of pinch plasmas.

In this formalism, spectral lines from doubly excited states are not treated individually,
but instead, all lines between a certain pair of electron configurations are grouped together and treated as a single line with a fixed width. Since in practice, such a group of lines is difficult to spectrally resolve experimentally, it is referred to as an unresolved transition array (UTA)—hence the name of the formalism.

For this work, we used the expressions as given in Refs. \[42\] and \[43\] to approximate the effective strength of each array, its central wavelength, and its width. The expressions contain several (combinations of) Slater and spin-orbit integrals of the relevant configurations. These were calculated using the COWAN atomic data package \[29\]. The transitions included in the calculations were the 4d-5p, 4d-4f, and 4p-4d transitions, each with a “witness” electron in either an excited 5s, 5p, 4f, 5d, or pd state. To simplify the calculations, we used for the width of each array (in terms of energy) the expression for the case without an excited witness electron. In view of the present goal, which is just a qualitative evaluation of the influence of doubly excited states rather than a detailed calculation of the resulting spectrum, such an approximation seems to be justified.

Another simplification is that in the UTA formalism, the effect of opacity on the emission of doubly excited states is not evaluated line by line but only for the array as a whole. This means that the formalism can only be used if the individual lines do not meet the blackbody limit. In the present case, this requirement is fulfilled given the high energies of the upper levels and the corresponding low population densities.

### 6.7.2 Results

In a first attempt at simulating EUV spectra including the emission of doubly excited states, we simply assumed that these states followed the analytical excited state distribution function as derived in Sec. 6.2.3. In practice, this means that they would be distributed according to the excitation saturation balance (ESB) as explained there. In fact, given their high energies, the overpopulation to the Saha density is so small that they could even be considered to be in partial local Saha equilibrium (pLSE) \[20\], which can be regarded as a limiting case of ESB. However, we found that this “naive” approach does not lead to good results, as Fig. 6.12 demonstrates for spectrum (c) with input parameters as given in Table 6.1.

Within the assumption of pLSE, the simulated spectra show very strong features that are caused by 4d-4f and 4p-4d transitions of doubly excited states. In order to obtain more realistic spectra, we had to artificially reduce the calculated probabilities of these transitions by a factor 40—which is basically just equivalent to assuming that they are strongly underpopulated compared to pLSE. Two examples of the resulting spectra are displayed in Fig. 6.13. The figure shows an improved match between experimental and simulated spectra, compared to the simulations without doubly excited states being taken into account. The effect is especially clear for the xenon spectrum between roughly 11.5 and 15 nm.
6.7 Addendum: The contribution of doubly excited states

![Simulated EUV spectrum of xenon for case (c) of Table 6.1 with doubly excited states populated according to ESB (dashed line). For comparison, the simulation without doubly excited states (solid line), and the experimental spectrum (dotted line) are also shown—clearly, the assumption that doubly excited states are populated according to ESB, does not lead to a good match with the experimental spectrum.]

6.7.3 Discussion and conclusions

The results presented here, although based on a fairly crude model of the energy levels and populations of doubly excited states, show that these states could indeed in part be responsible for the discrepancies between experimental and simulated spectra as observed in Sec. 6.5.

The fact that the doubly excited states seem not to be populated according to the ESB or pLSE, makes good sense from a theoretical point of view. Collisional population and depopulation of these states to singly excited levels with close-lying energies is quantum mechanically forbidden. Hence, an important underlying assumption of ESB is not fulfilled for these specific states.

Further, in the model described earlier in this chapter, the population densities of individual excited states are independent of their main optical transition probabilities. In the case of ESB, this is true because radiative depopulation can be neglected in comparison with collisional (de)population. In the case of corona balance, the collisional excitation rate from the ground state to the excited state and the optical transition probability back to the ground state are in principle both proportional to the oscillator strength of the transition. Hence, in the expression of the population density of the excited state, the oscillator strength cancels out.

However, doubly excited states are populated by collisional excitation from certain singly excited states rather than directly from the ground state. Also, population “from above” (i.e., in a radiative cascade caused by dielectronic recombination) can be an efficient mechanism. In these cases, the population density of a state may no longer be independent of
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Fig. 6.13. Simulations of spectra (c) and (f) with input parameters as given in Table 6.1 shown in parts (a) and (b), respectively. Solid lines: simulations including doubly excited states, with reduced probabilities for 4d-4f and 4p-4d transitions; dashed lines: without doubly excited states; dotted lines: experimental spectra.

its specific optical transition probabilities. Doubly excited states with 4f or pd electrons have much stronger optical transition probabilities to lower lying levels than doubly excited states with just a 5p electron. Hence, the former could be strongly underpopulated compared to the latter—which is just what we found in the present calculations.

The possible influence of (capture) radiative cascade was already discussed for certain lines of Xe8+ in the decay phase of the hollow cathode triggered discharge; see Fig. 4.11 in Chapter 4 [9] and the discussion in Sec. 4.4.3. Now we see that radiative cascade can affect the population of doubly excited states even in a not-yet-recombining plasma.

References

References


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Chapter 7

Stark broadening experiments on a vacuum arc discharge in tin vapor

Abstract

Pinched discharge plasmas in tin vapor are candidates for application in future semiconductor lithography tools. This chapter presents time-resolved measurements of Stark broadened linewidths in a pulsed tin discharge. Stark broadening parameters have been determined for four lines of the Sn III spectrum in the range from 522 to 538 nm, based on a cross-calibration to a Sn II line with a previously known Stark width. The influence of the electron temperature on the Stark widths is discussed. Results for the electron densities in the discharge are presented and compared to Thomson scattering results.

7.1 Introduction

Discharge plasmas as powerful sources of extreme ultraviolet (EUV) radiation have been or are being developed by a number of groups in the world; a selection can be found in [1–7]. An important potential application of such sources lies in the field of the next generation of semiconductor lithography, which requires radiation in a 2% wavelength band around 13.5 nm. Traditionally, in many of these sources, xenon is applied as the working element, since it not only exhibits a transition array near the desired wavelength, but also, being a noble gas, it is not easily deposited on or chemically reacting with the surfaces of (multilayer mirror) optics. However, because of doubts whether the xenon-based sources will in the end be able to meet the power requirements from the industry, the center of attention has recently shifted towards the use of tin as an alternative working element [8]. Despite the obvious drawbacks (tin is a solid under ambient conditions), it is an interesting element because of the highly favorable shape of its spectrum in the EUV range.

Since early 2003, a triggered vacuum arc in Sn vapor from the Russian Institute of Spectroscopy (ISAN) has been in operation in the ASML EUV laboratory in Veldhoven, The Netherlands. Several techniques have been applied for the characterization of this source, and, more generally, to identify the mechanisms and elementary processes that play important roles in this type of discharge. Among these techniques are time-resolved plasma imaging, EUV spectrometry (see Chapter 5 [9]), and Thomson scattering (see Chapter 8 [10]). Since detailed descriptions of the evolution of the plasma are given in Sec. 2.5 and Chapters 5 and 8, only the main points will be repeated here.

Before ignition of the discharge, a ring of capacitors that is connected to the electrodes is charged to a potential of about 4 kV. The cathode, which serves as the bottom electrode, is covered with a thin layer of liquid tin and the discharge is ignited by the vaporization of a small amount of the tin (trigger phase). A current starts to flow once the partially ionized cloud of tin vapor has expanded towards the edge of the ring-shaped anode.

Due to the low inductance of the electrical circuit, the current rises very quickly to about 20 kA (prepinch phase). The large electric current has two effects. First, it causes strong heating of the plasma and multiple ionization of tin up to at least Sn$^{11+}$. The electron temperature can reach values up to around 30 eV in this phase. Second, the electric current creates the “pinch” effect: the Lorentz forces acting on the charged particles in the plasma cause the plasma to collapse to a needlelike shape with radius of about 100 $\mu$m on the axis of symmetry of the discharge (pinch phase). Measurements show that the strongest EUV radiation is emitted in this phase.

Since the magnetic field confines the plasma only in the radial direction, the plasma can escape along the discharge axis and the pinch dies. Simultaneously, the electric current drops and the plasma starts to cool down and expand (decay phase). At the same time, the heat that was deposited into the tin layer on the cathode by the strong current during
7.1 Introduction

Anode

Cathode

Pinch plasma

Liquid tin

Fig. 7.1. A schematic cross section of the electrodes of the tin vapor discharge. The curved shape in the center represents the position of the pinch plasma. The horizontal rectangle shows the orientation and the approximate position of the plasma cross section as it was imaged by the spectrometer onto the ICCD camera.

the prepinch and pinch phases causes additional evaporation of tin and the creation of a new, much cooler plasma.

For the recording of Thomson scattered spectra, a spectrometer was used that was optimized for wavelengths near the second harmonic at 532 nm of a pulsed neodymium-doped yttrium aluminum garnet (Nd:YAG) laser. The arrangement was such that a horizontal cross section of the discharge could be imaged onto an intensified charge-coupled device (ICCD) camera, while the other dimension was reserved for wavelength information. A schematic image of the discharge electrodes and the imaged part of the plasma is shown in Fig. 7.1.

During the Thomson scattering experiments, recordings were also made for the background radiation as emitted by the plasma itself, in wavelength ranges including the one of roughly 527–538 nm. The presence of a few tin emission lines in this range with linewidths that were varying strongly during the evolution of the plasma led to a closer analysis of this part of the tin spectrum and the major line broadening mechanisms.

It was concluded that under the plasma conditions of the tin discharge, Stark broadening—due to collisions of the radiating atom with charged particles—is the dominant broadening mechanism. In principle, it is possible to derive electron densities from the Stark widths of broadened lines; this is a well-established spectroscopic technique \[11, 12\]. Thus, measurement of the plasma emission would provide an additional method for determination of electron densities independent of the Thomson scattering technique. Using the same setup as the one for Thomson scattering provides the benefits of an accurate wavelength calibration and good time resolution due to the presence of an ICCD camera.

However, we found that the calculation of electron densities from the widths of the detected lines was not straightforward. Of the four tin lines in the 527–538 nm range, one was identified as a \(\text{Sn I} \) line, while the other three were found to be \(\text{Sn II} \) lines. Only for the \(\text{Sn II} \) line was the Stark broadening parameter known from the literature \[13, 14\], but in certain spectra only the widths of the \(\text{Sn III} \) lines could be determined with
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sufficient accuracy. Further, we knew that for certain phases of the discharge, the electron temperature varies strongly, and is much higher than the temperature of $1.0 \times 10^4$ K and $1.16 \times 10^4$ K, for which the broadening parameter was given.

In the following section, an approach will be presented for a cross-calibration of the Stark widths of the Sn III lines to the width of the Sn II line. Also, a theoretical justification is given for the interpretation of Stark broadening measurements at higher electron temperatures, and the importance of other possible broadening mechanisms is discussed. In Sec. 7.3, a brief description of the setup and the experimental procedure for the determination of the electron densities is given. In Sec. 7.4, the Stark broadening parameters for four Sn III spectral lines (three in the 527–538 nm range and one at a smaller wavelength) are presented. Further, electron density values have been derived for all phases of the discharge except the pinch phase. These values are presented and compared to the Thomson scattering results for the prepinch phase. In the same section, the validity and accuracy of the results are discussed.

7.2 Stark widths

7.2.1 Stark broadening parameters of the observed lines

The following lines have been detected and identified as belonging to the Sn II and III spectra:

Sn II  
$6d^2D_{3/2} \rightarrow 6p^2P_{1/2}^{o}$, 533.39 nm (a),

Sn III  
$6p^3P_{1}^{o} \rightarrow 5d^3D_{1}$, 529.3 nm (b),
$6p^3P_{1}^{o} \rightarrow 5d^3D_{2}$, 535.1 nm (c),
$6p^3P_{0}^{o} \rightarrow 5d^3D_{1}$, 537.1 nm (d),
$6p^1P_{1}^{o} \rightarrow 6s^1S_{0}$, 522.6 nm (e).

In the following, these lines will also be referred to by their labels, (a)–(e), as given above. Lines (a)–(d) are in the 527–538 nm wavelength range, and could therefore all be recorded together in a single measurement. Line (e) is at a lower wavelength, and could therefore only be recorded together with line (b) in a single measurement over the 511.7–531.5 nm range.

Energy level and wavelength information for line (a) is given in Ref. [13, 14]. References [13] and [14] also give Stark broadening values of $0.27 \pm 0.04$ nm and $0.30 \pm 0.04$ nm, respectively, defined as the Lorentzian half-width at half maximum (HWHM) of the line at an electron density $n_e = 10^{23}$ m$^{-3}$. The four Sn III lines (b)–(e) have been identified in the present study with the help of energy level information from Ref. [15].

Apart from these data, the literature information is, in view of our application, rather limited. For line (a), the broadening parameter has only been measured for electron tem-
7.2 Stark widths

Temperatures of $1.16 \times 10^4$ and $1.0 \times 10^4$ K, by Miller and Martínez, respectively, while the temperatures in the tin discharge under study can get up to tens of eV. Furthermore, for lines (b)–(e), to our knowledge no broadening information is available from the literature at all.

To solve the latter problem for lines (b)–(d), we have made a cross-calibration of their widths to the width of line (a), for those spectra for which both could be determined with sufficient accuracy. Nearly all of the data points are in the decay phase of the discharge. The widths of lines (b)–(d) were assumed to be directly proportional to the width of line (a), and for each line, the Stark broadening parameter was calculated from the proportionality constant as derived from the experimental data, and the Stark broadening parameter of line (a). For the latter, the average of the literature values was applied.

Line (e) has not been recorded simultaneously with line (a) in a single measurement, and therefore only an indirect cross-calibration of the Stark width using line (b) was possible. A Stark broadening parameter for this line has been calculated, but the indirect derivation of this value was not considered to be accurate enough to be useful for the subsequent determination of electron densities.

Although the Stark broadening is mainly determined by the electron density $n_e$ in the plasma, the electron temperature $T_e$ also has a certain influence. Before we can do an interpretation of the results obtained at higher electron temperatures, we need to evaluate the temperature dependency of the line broadening.

The electron impact broadening as presented by Griem [16] is given by the following expression,

$$w_{se} = 8 \left( \frac{\pi}{3} \right)^{3/2} \frac{\hbar}{m a_0} n_e \left( \frac{E_H}{k_B T_e} \right)^{1/2} \left[ \langle i | r^2 | i \rangle \bar{g}_{se} (\xi_i) + \langle f | r^2 | f \rangle \bar{g}_{se} (\xi_f) \right],$$

where $i$ and $f$ denote the initial and final states of the transition, respectively, $\xi_{i,f} = \frac{3}{2} k_B T_e / |\Delta E_{i,f}|$, where $\Delta E_{i,f}$ are the energy differences to the nearest perturbing levels for those states, $\bar{g}_{se} (\xi)$ is an effective Gaunt factor, and $w_{se}$ is the HWHM of the Lorentz profile in frequency units. The other symbols have their usual meanings. $w_{se}$ is proportional to the full width at half maximum (FWHM) line broadening $\Delta \lambda$ in wavelength units, through the expression $\Delta \lambda = w_{se} \lambda^2 / (\pi c)$.

According to Eq. (7.1a), $w_{se}$ depends on the electron temperature both through an explicit $1/\sqrt{T_e}$ dependency and through the effective Gaunt factors, which have $T_e$ in their arguments. It can be rewritten as

$$w_{se} = A \frac{\bar{g}_{se} (\xi_i)}{\sqrt{\xi_i}} + B \frac{\bar{g}_{se} (\xi_f)}{\sqrt{\xi_f}},$$

where the prefactors $A$ and $B$ do not depend on $T_e$. In the approximation of Van Regemorter [17], the effective Gaunt factor $\bar{g}_{se} (\xi)$ for positive ions tends to a constant value of 0.2 for $\xi \to 0$ and behaves roughly as $(\sqrt{3}/2\pi) \ln \xi$ for large values of $\xi$. 

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Fig. 7.2. The function $g_{se}(\xi) / \sqrt{\xi}$, with $g_{se}(\xi)$ as defined in [17]. The plot shows the very weak dependency of the function on $\xi$.

Now, the function $g_{se}(\xi) / \sqrt{\xi}$ exhibits only a very weak dependency on $\xi$ for a large range of values of $\xi$. This point is illustrated in Fig. 7.2. Therefore, as long as the $\xi_{i,f}$ values for both the initial and final states of a measured profile are not very far from those applicable to the calibration of the same line, an estimation of the electron density will be possible without prior detailed information on the electron temperature in the plasma. Also, Fig. 7.2 shows that $w_{se}$ is constant within a factor 2 as long as both $\xi_{i,f}$ stay within a very wide range of roughly $1 < \xi < 200$. In the following, we will use the literature and calibration values of the Stark broadening parameters for the calculation of the electron densities, and check the validity of the method by evaluating the values of $\xi_{i,f}$ both for the calibration and for the electron density measurements.

### 7.2.2 Other broadening mechanisms

Apart from Stark broadening, several other mechanisms can contribute to the broadening of spectral lines that are emitted by plasmas [11]. These include natural line broadening, Doppler broadening, and pressure broadening due to collisions with neutral atoms. However, given realistic plasma properties, natural line broadening is negligible compared to all other mechanisms. With an electron temperature of about 30 eV or lower, Doppler broadening of tin lines near 532 nm will not exceed about 0.025 nm. Further, since the interaction between charged particles is much stronger than the interaction with neutrals, and given the fact that the tin discharge has a considerable degree of ionization for all relevant phases, the pressure broadening due to collisions with neutral atoms will be much smaller than Stark broadening. Therefore, it can be concluded that Stark broadening will dominate over all of the broadening mechanisms mentioned above, provided that it is not very small compared to the width of the apparatus profile, which is about 0.16 nm FWHM, as discussed in Sec. 7.3.

Macroscopic (external) electric and magnetic fields can also contribute to line broadening.
An external electric field will cause Stark splitting of the emission lines. For large fields, the (energy) splitting of a level is on the order of \( S = e a_0 p^2 E \), where \( E \) represents the magnitude of the electric field, and the prefactor is a typical electric dipole matrix element with a perturbing level, where \( a_0 \) is the first Bohr radius and \( p \) represents the principal quantum number of the upper level. However, as long as this value is small compared to the distance \( \Delta \) to the closest perturbing level, only a small quadratic contribution remains (similar to the expression derived in Ref. 18 for the simplest case of only two strongly interacting levels),

\[
\Delta E \approx \frac{S^2}{\Delta} = \frac{(e a_0 p^2)^2}{\Delta} E^2.
\]

(7.2)

The magnitude of the electric field in the plasma will never exceed a value of about \( 1 \times 10^6 \) V/m, which is near the values that we find when we divide the maximum potential of 4 kV by the typical electrode distances. During the discharge, the typical electric fields will be even smaller since most of the potential drop between the electrodes will then be concentrated in the plasma sheaths. Now, for all lines, \( S \) is on the order of 2 meV. The lower level of line (a) has an energy gap of 11 meV to the nearest perturbing level; the other lines all have energy differences \( \Delta \) = 0.7 eV or larger. This results in a line broadening of not more than 0.08 nm full width for line (a) and less than 1.2 \times 10^{-3} \) nm for the other lines. Therefore, only for line (a) could the electric field result in a detectable contribution to the broadening.

A similar argument is valid for the motional Stark effect, which results from the electric field that is sensed by a moving charged particle in the presence of a magnetic field. For the phases of the discharge for which measurements have been done, the magnetic field can reach a value of up to about 3.6 T, as is discussed in detail below. For realistic plasma velocities of about \( 10^4 \) m/s, the magnitude of the “motional electric field”, \( E_{\text{mot}} = |v \times B| \approx 4 \times 10^4 \) V/m, is even much smaller than the value of the electric field used for the estimations above.

Finally, the magnetic field can contribute to the line broadening directly through the Zeeman effect. Roughly, the line splitting in terms of energy will be on the order of \( \mu_B B \), where \( \mu_B = e \hbar/2m_e = 9.3 \times 10^{-24} \) J/T is the Bohr magneton. Of all the phases for which measurements are available, the magnetic field will be the strongest in the prepinch phase. In this phase, the current increases from nearly zero to almost 20 kA between about 90 and 30 ns before the pinch. An example of a current plot is shown in Fig. 7.3. Assuming a flat spatial current density distribution, and a constant plasma radius of \( r \approx 0.7 \) mm before the start of the compression, the cross section averaged magnetic-field strength increases from zero to about 3.6 T. This can lead to a full width line broadening of up to about 0.06 nm at \( t = -30 \) ns, which might give a significant contribution to the total broadening of the lines. The effects of the electric and magnetic fields on the results will be discussed in Sec. 7.4.
Fig. 7.3. Example of the discharge current (solid curve) and EUV emission derived from plasma imaging experiments (dotted curve) during the discharge. The absolute value of the current is derived from the electrical circuit parameters, and is only approximate.

7.3 Experiment

For all experiments, the triple grating spectrograph (TGS) was applied as originally designed and built by Marco van de Sande [19] for application in Thomson scattering experiments, and slightly modified and used for work on the tin discharge, as discussed in Chapter 8 [10]. Depending on the focal length of the last lens in the system, the effective spectral range of the system could be selected to be either about 10 nm or 20 nm. The stray light reduction properties of the TGS were used only for simultaneous Thomson scattering (TS) measurements.

Stark widths have been determined both from the actual spectra that have been recorded as background in the Thomson scattering experiments, and from separately recorded plasma emission spectra, mainly for the trigger and decay phases of the discharge, in which TS was not feasible. The TS spectra are over the wavelength ranges of 511.7–531.5 nm and 532.6–552.9 nm, while the separately recorded spectra cover the 526.9–537.7 nm range. In both cases, only the vertically polarized signal has been considered. In the former case, only one or three of the four lines (a)–(d) could be used, respectively, for determination of the electron density. In the latter case, lines (a)–(d) were all available.

The camera’s optical gating time was always 5 ns. The camera was synchronized to the ignition of the discharge, and a delay generator was used to vary the timing of the measurements relative to the evolution of the discharge. The time resolution of the measurements was about 10 ns, being limited both by the optical gating time of the camera and the pulse-to-pulse jitter in the timing of the discharge evolution, which also contributed about 5 ns. Both single-shot and multiple-shot averaged experiments have been performed.
The width of the apparatus profile of the TGS in the spectral direction has been determined using the widths of the Sn III lines from the outer regions of the plasma during the decay phase, where the electron densities are very low and hence the contribution of the apparatus profile to the total measured widths of the lines could be expected to be dominant. The apparatus profile has been found to have a FWHM of 0.16 nm.

Subsequently, the line profiles have been fitted to a Voigt profile, with a fixed Gaussian contribution to take the apparatus profile into account, and a Lorentzian contribution with a variable width. The Lorentzian contribution was fully ascribed to Stark broadening. The spectra over the 527–538 nm range for which the widths of lines (a)–(d) could all be determined with sufficient accuracy were used for the determination of Stark broadening parameters, as discussed in the previous section.

After this, for every spectrum the electron density was calculated as a simple average of the electron density values derived from the individual lines for which the broadening could be determined with sufficient accuracy.

In principle, radially resolved spectra of the axisymmetric discharge could have been obtained by Abel inversion at each wavelength of the entire spectrum. However, such a procedure would have been extremely laborious and therefore we have chosen to limit ourselves to fitting of only the central spectrum from the lateral profile, thereby effectively obtaining an electron density estimate for the entire cross section of the discharge. Since the highest density regions of the plasma will be emitting the strongest radiation and therefore contribute to the measured spectrum the most, we assume that this effective electron density will be close to the peak value of the density profile, to within about 25%.

7.4 Results and discussion

An example of the plasma emission spectrum in the 527–538 nm range is shown in Fig. 7.4. In Table 7.1, FWHM values are given for the linewidths in the same spectral range for different times in the evolution of the discharge.

The Stark broadening parameters for lines (b)–(d), derived from cross-calibration to the widths of line (a) for those entries in Table 7.1 for which all widths are present, are given in Table 7.2. Also, the Stark parameter for line (e), derived from comparison of its width with that of line (b), is included.

In Sec. 7.2 some limitations were given for the temperature ranges of the Stark results. However, the parameters in Table 7.2 were determined almost exclusively from measurements in the decay phase, which is a recombining plasma and therefore necessarily has a low electron temperature. From the model of Colombant and Tonon [20], it follows that in a stationary plasma the electron temperature must be between 1 and 3 eV when both Sn$^+$ and Sn$^{2+}$ are abundant. For a recombining plasma, the electron temperature will be even lower. As a result, the actual temperature will not be far from the values used in Refs. [13] and [14], and the Stark broadening parameter of line (a) will be close to the literature
Fig. 7.4. Example of a plasma spectrum in the 527–538 nm wavelength range, recorded 120 ns after the pinch. Apart from the experimental data, also the individual line fit curves (a, Sn II 533.39 nm; b, Sn III 529.3 nm; c, Sn III 535.1 nm; d, Sn III 537.1 nm) and their sum have been plotted. In this case, the Gaussian apparatus profile was negligible compared to the Lorentzian Stark contributions. A flat background contribution was subtracted from the experimental curve and has not been included in the graph.

Table 7.1. Full widths at half maximum of lines (a)–(d) for measurements in the 527–538 nm range. The last column gives the electron density values derived using the calibration data as described in the text.

<table>
<thead>
<tr>
<th>$t$ (ns)</th>
<th>$\Delta \lambda_a$ (nm)</th>
<th>$\Delta \lambda_b$ (nm)</th>
<th>$\Delta \lambda_c$ (nm)</th>
<th>$\Delta \lambda_d$ (nm)</th>
<th>$n_e$ ($10^{23}$ m$^{-3}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>270</td>
<td>0.82</td>
<td></td>
<td></td>
<td></td>
<td>1.5</td>
</tr>
<tr>
<td>240</td>
<td>0.85</td>
<td></td>
<td></td>
<td></td>
<td>1.5</td>
</tr>
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<td>210</td>
<td>0.69</td>
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<td></td>
<td></td>
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<td></td>
<td></td>
<td></td>
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<td></td>
<td></td>
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</tr>
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<td></td>
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<td>90</td>
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<td></td>
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<td>1.7</td>
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<td>1.9</td>
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<td>0.32</td>
<td>4.5</td>
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<tr>
<td>60</td>
<td>6.95</td>
<td>0.90</td>
<td>0.70</td>
<td>0.68</td>
<td>11</td>
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<tr>
<td>70</td>
<td>7.88</td>
<td>1.05</td>
<td>0.94</td>
<td>0.83</td>
<td>14</td>
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<tr>
<td>80</td>
<td>7.00</td>
<td>1.09</td>
<td>0.94</td>
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<tr>
<td>90</td>
<td>6.94</td>
<td>1.00</td>
<td>0.92</td>
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Table 7.1. (Continued.)

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<th>$t$ (ns)</th>
<th>$\Delta \lambda_a$ (nm)</th>
<th>$\Delta \lambda_b$ (nm)</th>
<th>$\Delta \lambda_c$ (nm)</th>
<th>$\Delta \lambda_d$ (nm)</th>
<th>$n_e$ ($10^{23}$ m$^{-3}$)</th>
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<tr>
<td>100</td>
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<td>0.98</td>
<td>0.89</td>
<td>0.85</td>
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<td>110</td>
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<td>0.95</td>
<td>14</td>
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<td>120</td>
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<td>0.73</td>
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<td>0.24</td>
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<td></td>
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</tr>
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<tr>
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<td>1.25</td>
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<td></td>
<td>2.2</td>
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<tr>
<td>360</td>
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<td></td>
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<tr>
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<td></td>
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<td></td>
<td></td>
<td>0.82</td>
</tr>
<tr>
<td>540</td>
<td>0.31</td>
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<td>0.56</td>
</tr>
<tr>
<td>570</td>
<td>0.33</td>
<td></td>
<td></td>
<td></td>
<td>0.59</td>
</tr>
<tr>
<td>600</td>
<td>0.29</td>
<td></td>
<td></td>
<td></td>
<td>0.52</td>
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<td>700</td>
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<td></td>
<td></td>
<td>0.34</td>
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<tr>
<td>900</td>
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<tr>
<td>1100</td>
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<td></td>
<td>0.16</td>
</tr>
<tr>
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<td>0.12</td>
<td></td>
<td></td>
<td></td>
<td>0.21</td>
</tr>
<tr>
<td>1500</td>
<td>0.04</td>
<td></td>
<td></td>
<td></td>
<td>0.073</td>
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</tbody>
</table>

Similarly, the Stark broadening parameters in Table 7.2 can be interpreted as being valid for an electron temperature of 1 eV. We estimate the errors in the Stark broadening parameters due to uncertainties in the electron temperature to be not more than about ±20%.

Other possible error sources include the uncertainties in the literature data on the broadening of line (a), the spatial gradients in the electron density, and macroscopic electric and magnetic fields in the plasma. However, since the calculation of Stark parameters is based on a comparison between linewidths, the errors due to the spatial variations in the electron density will partially cancel out. Further, electric and magnetic fields do not play an important role in the decay phase of the discharge, as is discussed below. Since more
Table 7.2. Stark broadening parameters $\Delta \lambda$ for four lines of the Sn III spectrum, given in the form of half-widths at half maximum of the Lorentzian profiles in wavelength units for $n_e = 10^{23} \text{ m}^{-3}$ and $T_e = 1 \text{ eV}$. Also given are the estimated relative errors in these numbers, as well as theoretical broadening parameters $\Delta \lambda_{se}$, derived using the semiempirical formula by Griem [16].

<table>
<thead>
<tr>
<th>Line</th>
<th>$\Delta \lambda$ (nm)</th>
<th>Rel. error</th>
<th>$\Delta \lambda_{se}$ (nm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>(b)</td>
<td>$6p^3P_{1}^o \rightarrow 5d^3D_1$</td>
<td>0.043</td>
<td>0.030</td>
</tr>
<tr>
<td>(c)</td>
<td>$6p^3P_{0}^o \rightarrow 5d^3D_2$</td>
<td>0.030</td>
<td>0.030</td>
</tr>
<tr>
<td>(d)</td>
<td>$6p^3P_{0}^o \rightarrow 5d^3D_1$</td>
<td>0.032</td>
<td>0.030</td>
</tr>
<tr>
<td>(e)</td>
<td>$6p^1P_{0}^o \rightarrow 6s^1S_0$</td>
<td>0.061</td>
<td>0.036</td>
</tr>
</tbody>
</table>

Data are available for lines (b) to (d) than for line (e), and the Stark parameter for line (e) has been derived only from an indirect measurement, the error estimate for line (e) is larger than for the other three.

When we compare the experimentally determined Stark widths in Table 7.2 with the semiempirical formula of Griem (7.1a), using his approximation

$$\langle x \mid r^2 \mid x \rangle = \frac{p_z^2}{2(z+1)^2} \left[ 5p_z^2 + 1 - 3l_x (l_x + 1) \right] a_0^2,$$

(7.3)

where $x$ represents either $i$ or $f$, $p_x$ is the effective principal quantum number, $l_x$ is the orbital quantum number, and $z$ is the charge number of the level under consideration, we see that all results match to within a factor 2. A more detailed, quantum-mechanical calculation of the Stark broadening parameters was beyond the scope of this work.

In Fig. 7.5, the electron densities in the plasma, as derived from the 527–538 nm spectra, have been plotted versus time. The durations of the trigger plasma, prepinch phase, and decay phases of the main discharge are indicated for reference. Electron densities from the Stark broadening measurements in the prepinch phase are shown together with electron densities and temperatures from Thomson scattering experiments in Fig. 7.6. Results from multiple measurement series of Stark broadening, including those from the background spectra of actual Thomson scattering experiments, have been included in this figure to give an impression of the spread in the results. The figure shows that there is excellent agreement in magnitude and trend for both sources of electron density information.

Again, the temperature limitations as discussed in Sec. 7.2 should be checked. From energy level data in Ref. [15], it follows that for line (a), $\Delta E_{i,f}$ are rather small with values of only about 0.1 and 0.01 eV, respectively. This leads to large values of $\xi$ for the temperatures that can typically be found in the tin discharge plasma. However, the width of line (a) was used almost exclusively for measurements in the trigger and decay phases, which are both recombining plasmas. Following the discussion above, we can conclude that the electron temperatures in those phases will not be far from 1 eV, so that the Stark broadening parameters from the literature can be applied.
7.4 Results and discussion

Fig. 7.5. Electron densities as calculated from Stark broadening. The time scale has been subdivided into three parts: A, trigger plasma; B, prepinch phase; C, decay phase. No Stark broadening data are available for the pinch phase (around 0 ns on the time scale).

On the other hand, lines (b), (c), and (d) also play a role in the determination of the electron densities in the ionizing prepinch plasma. For these measurements, the electron temperature has values up to 16 eV, as can be derived from Fig. 7.6 For these lines, the tables in Ref. [15] give values for $\Delta E_{i,f}$ of 0.7 and 2.0 eV, respectively, and hence the energy differences to the nearest perturbing levels are much larger than for line (a). Now, $\xi_{i,f}$ vary from 2 and 0.75, respectively, in the decay phase (assuming $T_e \approx 1$ eV) to up to 34 and 12 in the prepinch phase for the initial and final states, respectively (with $T_e = 16$ eV). For this range of $\xi$ values, Fig. 7.2 shows that $\bar{g}_{se}(\xi)/\sqrt{\xi}$ is only a very weak function of temperature, and hence also for the Sn III lines the condition derived in Sec. 7.2 is met.

Finally, the possible contributions of the electric and magnetic fields to the line broadening were mentioned in Sec. 7.2.2 For the electric field, it was derived there that it could give an additional broadening to line (a) of not more than 0.08 nm. As table 7.1 shows, this contribution would be significant only after $t = 500$ ns. However, this is already far into the decay phase of the discharge and the capacitors will already have been drained of their charge almost completely, so that no large electric field is present anymore. Comparison of the estimated magnetic field contributions to the actually measured linewidths for the measurements in the prepinch phase show that the presence of the magnetic field might lead to an overestimation of the electron density by about 20%. In the other phases, the magnetic field is much smaller and/or the linewidths are much larger, so that the magnetic field does not lead to significant additional broadening.

Taking all sources of error into account, it seems reasonable to assume an error margin of about ±50% for the measured electron densities in the discharge.
Fig. 7.6. (a) Electron densities as derived from multiple series of Stark broadening data, and maximum fitted electron densities per profile from Thomson scattering, plotted as a function of time before the pinch. (b) Profile-averaged electron temperatures from Thomson scattering, weighted by the electron density.

7.5 Conclusions

In this work, measurements of electron densities have been presented using the Stark broadening of emission lines of singly and doubly ionized tin in a vacuum arc pulsed discharge. The Stark broadening parameter was known from the literature for only one line in the part of the spectrum under study. From our measurements, we have been able to derive Stark broadening parameters for four new Sn III lines with up to ±30% accuracy. Since a part of the measurements was done simultaneously with Thomson scattering experiments, the results from both techniques could be compared. Even when taking into account that a certain part of the tin line broadening in the prepinch phase is caused by the presence of a magnetic field, a good agreement both in the absolute values and in the trend of the data is found. The agreement in the absolute values can be considered as a confirmation of the validity of the cross-calibration of the Stark broadening parameters.

The measurements show that the electron density in the prepinch phase, which is on
the order of $10^{23}$ m$^{-3}$ before the start of the discharge current, increases to several times its earlier value in only about 50 ns time, thereby confirming the trend in the Thomson scattering results. Also, a decrease of the density from about $10^{24}$ m$^{-3}$ to below $10^{22}$ m$^{-3}$ in a longer time span of about 1.5 µs during the decay of the plasma has been shown.

In the discharge under study, the electron temperature varied over a large range, and in some cases was far from the values for which the Stark broadening parameter of the Sn II line was known from the literature. Despite these unfavorable circumstances, we have shown both in theory and in experiment that the Stark broadening technique can provide fairly accurate electron density information in the Sn EUV discharge over a range of more than two decades. This is due to the fact that the Sn III lines are about an order of magnitude narrower than that of the Sn II line. Stark broadening can therefore be a useful additional tool for characterization of this type of plasma.

References


Chapter 7: Stark broadening on a vacuum arc in tin vapor


Chapter 8

Collective Thomson scattering experiments on a tin vapor discharge in the prepinch phase

Abstract

Partially collective Thomson scattering measurements have been performed on a triggered vacuum arc in tin vapor, which is a candidate source of extreme ultraviolet light for application in semiconductor lithography. In this chapter, results on the electron densities and temperatures are presented for the prepinch phase of the discharge. Electron densities and temperatures increase from \(1 \times 10^{23} \text{ m}^{-3}\) to \(1 \times 10^{24} \text{ m}^{-3}\) and from 5 eV to over 30 eV, respectively, in about 100 ns. The results are confirmed by Stark broadening data.

8.1 Introduction

Extreme ultraviolet (EUV) lithography tools that are currently under development for application in the semiconductor industry require high-intensity sources of light in a wavelength band of 2% around 13.5 nm. In recent years, discharge plasmas have been regarded as the most promising candidates to meet the requirements set by the industry, and various types have been investigated by different groups worldwide. In particular, discharges in xenon have received much attention. More recently, increased effort has been put into the development of discharges in tin vapor, since tin has the advantage over most other elements under consideration that its EUV spectrum is strongly peaked at the desired wavelength (several contributions in Ref. [1]).

A triggered vacuum arc in Sn vapor from the Russian Institute of Spectroscopy (ISAN) has been in operation in the ASML EUV laboratory in Veldhoven, The Netherlands since March 2003. The discharge region consists of a flat cathode that is covered with a thin layer of liquid tin, and a ring-shaped anode located a few mm above the cathode. A schematic picture of the electrode cross section is given in Fig. 8.1. Before the start of the discharge, a positive electrical potential of 4 kV is applied to the anode. The discharge is started by the creation of a cloud of partly ionized tin vapor above the surface of the cathode, which expands towards the edges of the anode. Once the plasma near the anode has reached a sufficiently high density, a discharge starts. The current through the discharge increases to almost 20 kA in only a few tens of ns, and a multiply ionized, EUV-emitting plasma is formed. The strong current causes a pinching effect, meaning that due to Lorentz forces acting on the charged particles in the plasma, the plasma is compressed in the radial direction to a needlelike shape on the axis of the discharge, with a diameter on the order of 100 \( \mu m \). In view of the dynamics of the plasma, the time development of the discharge can be roughly split up into four main phases: (i) the trigger plasma, before the discharge current has started; (ii) the prepinch phase, in which the plasma is heated and ionized by the strong electrical current, and starts to compress; (iii) the pinch itself; and (iv) a decay phase. Since the pinch phase is very short-lived compared to the other phases of the discharge, its center is suitable to serve as a zero on the time scale when one is describing the properties of the plasma in the other phases.

The various phases of the discharge have been characterized by using time-resolved EUV and visible light imaging and EUV spectrometry, in a similar way to that described in Chapter 4 [2] for a hollow cathode discharge in xenon. First results have been presented in Ref. [3]; a more detailed description of the results can be found in Chapter 5 of this thesis [4].

The above-mentioned passive imaging and spectroscopic techniques have given valuable information on the qualitative behavior of the discharge. Plasma imaging has confirmed the identification of the four phases mentioned above and revealed details of the plasma.
evolution within these phases. EUV spectroscopy has shown the appearance of various ionization stages of tin (from 7+ to 10+) one after another during the prepinch phase, and the apparent cooling of the plasma during the pinch phase. A drawback of these methods, however, is that they do not give direct information on electron densities and temperatures, which are “fundamental” parameters for understanding and modeling plasmas. Instead, extensive approximations and assumptions (e.g., on the deviations from equilibrium in the plasma) are required to derive these parameters from the experimental results.

A method that can give direct information on both electron densities and temperatures is Thomson scattering (TS) spectroscopy. In this technique, a laser pulse is fired through the plasma, and the spectrum of the laser light scattered from free electrons in the plasma is recorded. In the noncollective scattering limit, the electron density in the plasma can be derived from the total intensity of the Thomson scattered light, whereas the width of the spectrum, caused by the Doppler effect, gives information on the velocity distribution, and hence on the temperature, of the electrons in the plasma. For higher electron densities, corrections for collective scattering need to be made.

In the work presented here, the Thomson scattering technique has been tested for the various phases of the discharge. Until now, we have been able to produce good data only for the prepinch phase. In the other phases, the extraction of good Thomson spectra was impossible due to the relatively high levels of plasma background radiation.

In addition, the measurement of Stark broadening of plasma lines is a frequently used method to determine electron densities in the plasma in a passive way. A few Sn lines in the wavelength range of interest for Thomson scattering were found to be broadened well beyond the apparatus spectral resolution, and the linewidths were used to derive electron density information. Here, this information is used only as a check on the validity of the TS results. A more detailed discussion of the Stark broadening results is given in Chapter 7.

In the following section, the collective Thomson scattering theory and experiment will be discussed. After that, the results for the prepinch phase of the discharge are presented. Finally, a possible approach will be discussed for making the TS technique feasible for the other phases of the discharge.
8.2 Collective Thomson scattering

8.2.1 The experimental setup

The setup used for the TS measurements was basically the same as the one described extensively in Ref. [5]. It has been used in the past for diagnostics on a range of laboratory plasmas, as described in Ref. [5] and, e.g., in Refs. [6–8]. A schematic overview of the setup is shown in Fig. 8.2.

The Thomson scattered photons originated from a frequency doubled neodymium-doped yttrium aluminum garnet (Nd:YAG) laser pulse of 7 ns duration, operated at a reduced repetition rate of 5 Hz, with an incident wavelength of $\lambda_i = 532.0$ nm, which was focused into the plasma through a plano-convex lens with a focus length $f = 1$ m. A pulse energy of 170 mJ was applied. Due to certain losses at the optics, effectively about 160 mJ of pulse energy was delivered into the plasma. Brewster-angle entrance and exit windows and a diaphragm just outside the entrance window helped to minimize stray light originating from the vacuum chamber. The laser beam passed through the discharge region between the anode and the cathode; the laser focus was aligned approximately 0.5 mm above the center of the cathode surface. This is the region where the most interesting features of the discharge (such as the trigger plasma and the pinch) occur. As the laser passed through the plasma in a horizontal direction, horizontal profiles of the plasma parameters could be derived from the measurements.
A triple grating spectrograph (TGS) was used to image the light scattered by free electrons inside the plasma onto the entrance plane of an intensified charge-coupled device (ICCD) camera. An image rotator is located inside the TGS so that horizontal spatial information is imaged onto the camera in the vertical direction. In this way, horizontal cross sections of the plasma parameters can be made and wavelength dispersion can take place in the horizontal plane. The first and second spectrographs are arranged in a subtractive configuration. An important role is played by a mask between the first and second spectrographs, which greatly reduces the Rayleigh scattered signal from the plasma and the stray light due to reflections from surfaces near the probed plasma volume.

The optical gate of the ICCD camera was reduced to 5 ns to minimize the level of background radiation emitted by the plasma itself. For each measurement, the signals from 250 pulses, recorded in 50 s, were added, mainly with the aim of averaging out fluctuations in the background radiation emitted by the plasma.

In the standard configuration of the TGS, the central wavelength was imaged onto the center of the CCD camera, so that a wavelength range of 10.9 nm was recorded. In this way, both sides of the TS spectrum could be recorded up to just over 5 nm away from the central wavelength. This configuration was chosen in the past for the recording the spectra of relatively cool plasmas with a sufficiently good spectral resolution. However, it was less suitable for application in plasmas with higher electron temperatures and/or densities. Therefore, compared to this standard configuration, the last lens in the TGS, the second lens of the third spectrograph, was replaced by one with a focal length $f = 300$ mm instead of the 600 mm focal length of the other lenses. Also, the alignment of the gratings was adjusted to project the incident wavelength onto the edge of the covered range instead of onto the center. Accordingly, the mask between the first and second spectrographs was replaced by an edge filter. Depending on which side of the spectrum was selected, a wavelength range of $511.7 - 531.5$ nm or $532.6 - 552.9$ nm could now be covered. A schematic view of the TGS is given in Fig. 8.3.

The trigger of the discharge, the Q-switch of the TS laser, and the optical gate of the ICCD camera were all synchronized to a flash-lamps-synchronized output from the TS laser, using two Stanford DG 535 delay generators. The relative jitter between the TS laser and the camera was on the order of 1 ns (mainly due to the laser), whereas the jitter of both relative to the plasma was on the order of 5 ns, due to pulse-to-pulse fluctuations in the timing of the start of the discharge after the trigger. These fluctuations, combined with the optical gate time of the camera, put a limit to the time resolution of the measurements.

A rotatable polarization filter was mounted onto the front of the TGS, and the polarization filter on the camera itself was removed. Both the horizontally and the vertically polarized signals were recorded in the presence of the TS laser pulse. The horizontally polarized signal was used to subtract the plasma background from the vertically polarized signal that contained the TS spectrum. This method was used to eliminate any influence of laser heating on the plasma background signal.
8.2.2 Interpretation of collective TS spectra

For the plasma parameters expected inside an EUV discharge, the TS spectrum would be partially collective. Here, the term collective refers to the fact that the free electrons in the plasma respond collectively to the incident laser light; another frequently used name for this type of scattering is coherent Thomson scattering. For fitting the experimental curves, the expression for the collective Thomson spectrum as given by Salpeter [9] was used. The spectral intensity per solid angle is given by

\[
\frac{dP_s(\omega)}{d\Omega} = \frac{d\sigma}{d\Omega} P_L L_s n_e S(k, \omega)
\]  

(8.1)

with \(P_s\) the scattered intensity, \(P_L\) the incident laser intensity, \(L_s\) the length over which scattered light is collected, and \(\sigma\) the single-electron scattering cross section. \(k\) and \(\omega\) are the scattering wave vector and the angular frequency of the laser light, respectively. The so-called form factor, \(S(k, \omega)\), is given by

\[
S(k, \omega) d\omega = \frac{1}{\sqrt{\pi}} \Gamma_\alpha(x_e) dx_e + Z \left( \frac{\alpha^2}{1 + \alpha^2} \right)^2 \frac{1}{\sqrt{\pi}} \Gamma_\beta(x_i) dx_i,
\]  

(8.2)
where
\[
\Gamma_\alpha(x_e) = \frac{\exp(-x_e^2)}{|1 + \alpha^2 W(x_e)|^2},
\]
\[
\Gamma_\beta(x_i) = \frac{\exp(-x_i^2)}{|1 + \beta^2 W(x_i)|^2},
\]
\[
\beta^2 = Z \left( \frac{\alpha^2}{1 + \alpha^2} \right) \frac{T_e}{T_i},
\]
\[
W(x) = 1 - 2x \exp(-x^2) \int_0^x \exp(p^2) dp - i\pi x \exp(-x^2).
\]

(8.3a)
(8.3b)
(8.3c)
(8.3d)

Here, \(x_e = \omega/(kv_e)\) and \(x_i = \omega/(kv_i)\), with the electron and ion mean thermal speeds, respectively, defined as
\[
v_e = \sqrt{\frac{2k_B T_e}{m_e}}, \quad v_i = \sqrt{\frac{2k_B T_i}{m_i}}
\]

(8.3e)

where \(k_B\) is the Boltzmann constant and \(T_{e,i}\) and \(m_{e,i}\) are the electron and ion temperatures and masses, respectively. Further, \(Z\) represents the average ionization degree and \(\alpha = 1/k\lambda_D \sim \sqrt{n_e/T_e}\) is the scattering parameter \(\alpha\), of which the square appears as a prefactor of the plasma dispersion function \(W(x)\) in the denominator of the shape function \(\Gamma_\alpha(x)\).

In Eq. (8.2), the first term represents the so-called electron contribution, whereas the second is known as the ion contribution. In this work, only the electron contribution has been studied since the ion contribution is so narrow and close to the incident wavelength that it cannot be separately measured and resolved spectrally. The shape of the electron contribution is governed by the scattering parameter \(\alpha\), of which the square appears as a prefactor of the plasma dispersion function \(W(x)\) in the denominator of the shape function \(\Gamma_\alpha(x)\).

The real part of \(W(x)\) contains an integral function that cannot be expressed in closed analytical form, and this is impractical in the actual fitting procedure. Therefore, an approximation \(W'(x)\) of \(W(x)\) was constructed, with \(\text{Im}[W'(x)] = \text{Im}[W(x)]\) and
\[
\text{Re}[W'(x)] = -\frac{1}{2x^2} - \frac{3}{4x^4} + \left( \frac{15}{8} + \frac{5}{4x^2} + \frac{3}{4x^4} \right) \exp(-x^2)
\]
\[
\quad - 0.612 \ x^2 \exp(-0.8913 \ x^2)
\]
\[
\quad - \left[ 0.0038 \ x \sin(3.2 \ x) + 0.0015 \ x^4 \right] \exp(-0.4 \ x^2).
\]

(8.4)

This leads to the definition of
\[
\Gamma_\alpha'(x_e) = \frac{\exp(-x_e^2)}{|1 + \alpha^2 W'(x_e)|^2}
\]

(8.5)
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which was used instead of $\Gamma_\alpha(x_e)$ for the calculation of $S(k, \omega)$. In this approximation, the generated Thomson spectrum was found to be accurate to better than 0.1% of the peak value for $\alpha \leq 1.5$ and 1% or better for $\alpha < 3$. As an example, Fig. 8.4 shows a plot of $\Gamma_\alpha(x_e)$ for $\alpha = 1.5$, together with a 1000 times magnified plot of the difference $\Gamma'_\alpha(x_e) - \Gamma_\alpha(x_e)$.

In the partially collective regime, $\alpha$ and $T_e$ can be determined from just the shape and the width of the spectrum, respectively. Once both $\alpha$ and $T_e$ are known, $n_e$ can be calculated. Therefore, an absolute calibration of the signal is in principle unnecessary. However, in the present work an intensity calibration of the scattered signal was done by measuring the Raman scattered signal from nitrogen at atmospheric pressure. In the fitting procedure, $\alpha$ was treated as a function of $n_e$ and $T_e$. In total, three free parameters were used: $n_e$, $T_e$ (from which two $\alpha$ was calculated), and a third parameter $\xi$, set to be the ratio of the measured TS intensity and the theoretical one. The value of $\xi$, which would be equal to 1 in the ideal case, was used as a check of the validity of the results and for evaluating the stability of the source.

8.3 Results and discussion

8.3.1 Electron temperatures and densities

An example of a measured Thomson scattering spectrum is shown in Fig. 8.5. The plot covers the lower half (in the wavelength domain) of the Thomson spectrum taken 35 ns before the pinch, at a position horizontally 1.0 mm away from the center of the discharge, towards the source of the TS laser pulse. The negative counts at the lower wavelength limit are due to the background subtraction.

For each measurement, five pixels on the CCD camera were grouped in the spatial direction to form “superpixels”; for each of these spatial positions, a fit of the Thomson
Fig. 8.5. Example of a measured TS spectrum. The negative counts at the lower wavelength end are due to the background subtraction. The smooth curve is a fit of the theoretical coherent-scattering spectrum to the experimental data. Fit results were \( n_e = 2.3 \times 10^{23} \text{ m}^{-3} \) and \( T_e = 16 \text{ eV} \), resulting in \( \alpha = 0.96 \).

spectrum was performed. An example of the resulting spatial profile is shown in Fig. 8.6. In some cases, especially at the edges of the plasma, the value of the fit parameter \( \xi \) was well below 1, meaning that the actual intensity of the Thomson signal, compared to the absolute calibration, was substantially lower than the one that would correspond to the fitted plasma parameters \( n_e \) and \( T_e \). This is an indication of spatial and/or temporal gradients in the plasma, or pulse-to-pulse fluctuations of the discharge: apparently a plasma with certain density and temperature was present at the probed position, but only in a certain fraction of the volume, gate time, or of the number of pulses used in the integration. This fraction is reflected in the ratio of measured and expected signal intensity. The effective electron density, as plotted in Fig. 8.6, is the fit result multiplied with this fraction.

The electron temperature in Fig. 8.6 has a hollow spatial profile. A similar shape of the profile was observed in most of the other measurements during the prepinch phase. Near the boundaries between the plasma and the surrounding vacuum, the ion densities will be lower than in the center of the plasma. This is confirmed by the electron densities presented here, combined with the assumption of quasineutrality of the plasma. Hence, in the outer regions the electrons will undergo fewer collisions with other species in which they can lose their energy. Therefore, a certain Ohmic heating of the electrons will lead to higher electron temperatures here than in the central part of the cross section.

For each time step, the spatial maximum of the fitted electron density has been selected and a weighted average over the profile has been calculated for the electron temperature (the weight being the effective local electron density). Both have been plotted against time in Fig. 8.7.
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Fig. 8.6. Profiles of electron densities (symbols and solid curve) and electron temperatures (dashed curve) recorded 35 ns before the pinch.

Fig. 8.7. Maximum fitted electron density for each profile [part (a)] and spatially averaged electron temperature weighted by electron density [part (b)] from Thomson scattering plotted as a function of time before the pinch. In part (a), results from Stark broadening measurements have been included for comparison.
The electron densities in the prepinch phase were found to increase from roughly $n_e = 10^{23} \text{ m}^{-3}$ to $10^{24} \text{ m}^{-3}$ in a time of about 100 ns, while the electron temperature increased from some 5 eV to about 35 eV. The plasma radii in the prepinch and pinch phases can be estimated from the TS electron density profile widths and plasma imaging in the EUV [4], respectively. Given the electron densities in the prepinch phase, and comparing the plasma radius with the final radius in the pinch phase, one can derive that in the pinch, the electron density should reach a value of just over $10^{25} \text{ m}^{-3}$, assuming that the average ionization degree does not change much anymore during the compression.

As mentioned already in Sec. 8.1, some line radiation was also detected in the plasma background during the prepinch phase. Using the Stark broadening effect, electron densities could be calculated from the linewidths. For one line, the $6d^2D_{3/2} \rightarrow 6p^2P_{1/2}^{o}$ Sn II line at 533.39 nm, the Stark broadening parameter was known from the literature [10, 11]. The width of this line was used to cross-calibrate the Stark widths of three Sn III lines in the same spectral range. The method could thus be extended to higher densities and higher temperatures, at which the Sn$^+$ ion was absent. Electron densities derived using this method have been included for reference in Fig. 8.7. The figure shows that there is excellent agreement to within experimental error for both the magnitude and the trend of both sources of electron density information. Only at –90 ns is the TS result considerably larger than the Stark broadening result. A more elaborate discussion of the Stark broadening measurements is given in Chapter 7.

8.3.2 Influence of the laser pulse on the plasma

Special care has been taken to evaluate the heating of the plasma, and more specifically of the electrons in the plasma, by the action of the laser. Nonlinear effects (such as multiphoton or tunneling ionization) do not play a role under the moderate laser conditions as used in this work; under the present plasma parameters, the main absorption processes are inverse bremsstrahlung (IB), as described in Refs. [12–14], and absorption in spectral lines of low ionization stages of tin. The absorption coefficient for IB is given by

$$\kappa_{IB} = \frac{\omega_{pl}^2}{\tilde{n}(\omega_{laser})} \frac{\nu_{ei}}{\omega_{laser}^2 + \nu_{ei}^2}, \quad (8.6)$$

where

$$\nu_{ei} \approx \frac{1}{6\pi} \left( \frac{1}{2\pi m_e} \right)^{1/2} \left( \frac{1}{k_B T_e} \right)^{3/2} \frac{n_e Z e^4}{\varepsilon_0^2} \ln \sqrt{1 + \Lambda^2} \quad (8.7)$$

and

$$\Lambda = \left( \frac{k_B T_e}{m_e} \right)^{1/2} \frac{4\pi \varepsilon_0 k_B T_e}{Z e^2} \frac{1}{\omega_{laser}}, \quad (8.8)$$

in which $\omega_{laser}$ represents the laser angular frequency, $\omega_{pl}$ the plasma frequency, $\tilde{n}$ the refractive index (which is very close to 1 for the laser frequency and range of plasma
parameters under consideration), \( \nu_{ei} \) the electron-ion collision frequency for momentum transfer, and \( Z \) the average ion charge. \( \Lambda \) contains the laser angular frequency \( \omega_{\text{laser}} \) rather than the plasma frequency \( \omega_{\text{pl}} \), as indicated in Ref. [13]. From \( \kappa_{\text{IB}} \), the laser energy \( E_{\text{laser}} \), the laser focal radius \( r_0 \approx 200 \mu\text{m} \), and the electron density, an upper limit for the energy gained per electron can be derived under the assumption of absence of any loss channels. This leads to a maximum increase of the electron temperature given by

\[
\Delta T_{e,\text{max}} = \frac{2}{3} \frac{\kappa_{\text{IB}} E_{\text{laser}}}{k_B \pi n_e r_0^2}.
\] (8.9)

An estimate of the temperature increase averaged over the duration of the laser pulse (which is relevant for the Thomson scattered signal) was obtained by taking half of the total laser pulse energy, or about 80 mJ, for \( E_{\text{laser}} \). An evaluation of Eqs. (8.6) and (8.9) using this energy shows that for the measurements presented in this chapter, \( \kappa_{\text{IB}} \leq 0.08 \text{ m}^{-1} \) (assuming that \( Z \leq 10 \)), so that \( \Delta T_{e,\text{max}} \) never exceeds 0.2 eV. For the first part of the prepinch phase, until 45 ns before the pinch, \( n_e \leq 3 \times 10^{23} \text{ m}^{-3} \) and \( T_e \leq 15 \text{ eV} \) so that \( \Delta T_{e,\text{max}} \) even stays below 0.05 eV. Therefore, inverse bremsstrahlung in all cases only leads to a very small relative error in the measured electron temperature. In the pinch phase, assuming \( n_e \approx 1 \times 10^{25} \text{ m}^{-3} \), \( T_e \approx 30 \text{ eV} \), and \( Z \approx 10 \), \( \Delta T_{e,\text{max}} \) would be about 2 eV.

Absorption of laser energy due to line transitions mainly plays a role in the relatively cool first part of the prepinch phase, when Sn\(^{+}\) and Sn\(^{2+}\) are still abundant. Especially the Sn II line mentioned above will contribute to the absorption since it is sufficiently broadened for the wings of the line profile to reach to the laser wavelength at 532.0 nm. The effect of absorption and stimulated emission at the laser wavelength will be that the upper and lower levels of the transition become strongly coupled, so that their densities per statistical weight become nearly equal. The distribution function of the other excited states, which is assumed to be Boltzmann-like when absorption is absent, will also become disturbed. As a result of this, the ionization potential of Sn\(^{+}\) will effectively get lowered by a certain fraction of the photon energy of 2.33 eV. This effect is illustrated schematically in Fig. 8.8. We estimated that in the most extreme case, the electron density will show an increase of about 30%. Indeed, we believe that the difference in Fig. 8.7 between Stark and Thomson results at 90 ns before the pinch can partially be explained by this effect.

A second result of the disturbed distribution function is that electronic deexcitation from the upper level to the lower level will happen more frequently than electronic excitation from the lower to the upper level. Because the free electron loses energy in the electronic excitation process and gains energy in the reverse process, the imbalance will lead to a net heating of the free electrons in the plasma. We used a simple stationary-state collisional-radiative model similar to the one described in Ref. [15], and the following expression for the approximate collisional (de)excitation rates as used in Refs. [16] and [17], to evaluate
Fig. 8.8. A schematic plot of the excited states distribution function of the Sn\(^{+}\) and Sn\(^{2+}\) systems in the presence of the laser field. Dashed line, undisturbed Saha-Boltzmann distribution; solid dots, densities per statistical weight of the lower and upper levels coupled by the laser field; solid line, approximate disturbed distribution. The densities in the plot are normalized to the density of the ground state of Sn\(^{+}\). The effective lowering of the ionization energy of Sn\(^{+}\) is indicated by the horizontal arrow. See the text for further explanation.

the order of magnitude of this effect,

\[
X(T_e, l, u) = \frac{6 \times 10^{-12} f(l, u)}{\Delta E\sqrt{T_e}} \exp \left( {-\frac{\Delta E}{T_e}} \right) \quad (8.10a)
\]

\[
X(T_e, u, l) = \frac{g_l}{g_u} \frac{6 \times 10^{-12} f(l, u)}{\Delta E\sqrt{T_e}}. \quad (8.10b)
\]

Here, \(l\) and \(u\) denote the lower and upper states of the transition, and \(\Delta E\) is the energy difference between both states in eV. The oscillator strength \(f = 0.73\) was derived from the optical transition probability of the Sn \(\Pi\) line, \(A = 8.6 \times 10^7 \text{ s}^{-1}\), as given by Ref. [18]. We found that the line absorption of laser light might lead to an increase of the electron temperature of up to a few eV in the most extreme case, and only when Sn\(^{+}\) is the dominant ion. Later in the prepinch phase the average charge of the ions will have increased to higher values, so that the Sn\(^{+}\) and Sn\(^{2+}\) ions are no longer abundant. At that time, heating and ionization by line absorption will not be important any longer.

It is worth noting that both effects are virtually independent of the laser power, as long as the power is large enough to disturb the Boltzmann distribution of the excited states of Sn\(^{+}\). However, the increase of electron temperature does depend linearly on the pulse duration.

From extrapolation of the measured Thomson scattered intensities to the expected electron temperatures and densities in the trigger and pinch phases of the discharge, and comparison with the levels of background radiation measured for those phases, it was found that the peak intensity of the Thomson spectrum would amount to only 2\% and 3\%,
respectively, of the intensity of the background. During the trigger phase, the background consists mainly of line radiation, while during the pinch phase it forms a (quasi)continuum. In both cases, pulse-to-pulse fluctuations in the background emission render the accurate determination of the Thomson signal impossible under the present experimental limitations.

8.4 Conclusions and future work

In this work, the feasibility of Thomson scattering on actual EUV producing plasmas has been proven. Electron densities in the prepinch phase have been found to increase from about $10^{23}$ m$^{-3}$ to near $10^{24}$ m$^{-3}$, while simultaneously the electron temperature increased from around 5 eV to more than 30 eV. The increase in the electron density can be explained both by the first onset of the plasma compression, and by a strong increase of the ionization degree of the plasma due to the increasing electron temperature.

It has been shown that the heating of the plasma by means of inverse bremsstrahlung absorption of the TS laser pulse does not significantly affect the results. In the very beginning of the prepinch phase, absorption of laser light in line radiation may be the cause of a certain overestimation of both the electron density and the electron temperature. The line absorption could lead to an increase of electron density of about 30%.

For the largest part of the prepinch phase, measurements of the electron densities using the Stark broadening of tin lines in the plasma confirm both the magnitude and the trends in the TS data.

The present experiments have also shown that the applicability of Thomson scattering to the very early part of the discharge, and the pinch and decay plasmas, is limited due to an excessively high level of background radiation (formed by line radiation, a quasicontinuum, and again line radiation, respectively). Efforts are underway to come to the necessary adjustments of the setup that will make sub-ns experiments possible. These include the application of a stimulated Brillouin scattering (SBS) cell to compress a laser pulse to sub-ns duration, and a sub-ns gated ICCD camera that is properly synchronized to the TS laser light pulse. The better signal-to-background ratio will enable probing the remaining discharge phases. The theory shows that the shorter pulse length will help to reduce the increase of electron temperature due to absorption in spectral lines. Also, measurements as a function of laser pulse energy will become possible, so that the assumptions about the laser-induced plasma heating in the pinch phase can be checked experimentally.

References


1 See Chapters [7] and [10] for the results of the efforts mentioned here.
References


Chapter 9

Sub-ns Thomson scattering setup for space and time resolved measurements with reduced background signal

Abstract

In order to reduce the level of recorded background radiation in Thomson scattering (TS) experiments, we have designed and built a new setup for time and space resolved sub-ns TS. Compared to our old setup, the new one is based on a faster camera, a laser with shorter pulse duration, and an optical delay line in the laser path. It has been characterized both by ray-trace simulations of the spectrograph and test experiments using Rayleigh and rotational Raman scattering on atmospheric pressure nitrogen gas. Although it was designed in the first place for experiments on a vacuum arc discharge in tin vapor, our setup can also be applied to other plasmas that emit high levels of radiation or in which fast phenomena play a role.

Chapter 9: Sub-ns Thomson scattering setup

9.1 Introduction

Thomson scattering (TS) spectroscopy is a reliable technique to measure the density $n_e$ and temperature $T_e$ of the electron gas in a plasma simultaneously. In this technique, a laser pulse is fired through the plasma and the light scattered by free electrons is recorded as a function of wavelength. It is often applied to plasmas that are located far from any vessel walls, such as plasmas for future nuclear fusion reactors. In such cases, scattering from atoms and ions, stray light reflections of the laser light, and light emission of the plasma itself are often not a problem for the measurement of the Thomson scattered spectrum.

However, in plasmas with very low ionization degree, the wings of the Rayleigh scattered light from atoms competes with the TS spectrum. Further, in many types of “technological” plasmas, such as deposition plasmas or lamps, the electrodes and the walls can be very close to the regions of interest in the plasma, and a certain amount of reflected laser light from solid surfaces cannot be avoided. In these cases, a notch filter is needed to block the central wavelength in the scattered light spectrum. In the past, an atomic absorption cell has been applied for this purpose [1]. Another method is the application of a so-called triple grating spectrograph (TGS) [2, 3]. In our group, a TS setup including a TGS has been designed and built by Marco van de Sande [3]. It has been applied to a number of different plasma types; see, e.g., Refs. [3–7].

Another problem that can occur in determining the shape of the TS spectrum from recorded images, is the competition of the TS signal with background radiation that is emitted by the plasma itself. In certain cases, the plasma radiation will consist just of a limited number of spectral lines. In such cases, it can be sufficient to select a wavelength for Thomson scattering at which the background spectrum is relatively clean, for example by application of a wavelength-tunable laser. However, if the plasma radiation forms a (quasi) continuum over a large wavelength range, simply selecting a suitable laser wavelength or blocking specific spectral lines is not enough to solve the problem. Instead, a more general method needs to be applied to improve the signal to background ratio.

This was found to be true for the case of our vacuum arc discharge in tin vapor. Pulsed discharge plasmas in tin vapor are candidate sources of extreme ultraviolet (EUV) light for application in next generation lithography tools. Our vacuum arc discharge is an experimental EUV source that was designed and built at the Institute of Spectroscopy of the Russian Academy of Sciences (ISAN). Due to its geometry, it has proven to be particularly accessible to different diagnostic techniques—see Chapters [3] [7] and [8] [10]. In a previous series of experiments, we used the existing TS setup mentioned above, with some small modifications, to perform Thomson scattering on this plasma (Chapter [8] [10]). Our TS experiments were successful in providing useful data for the so-called “prepinch phase” of the discharge, during which a strong electric current heats and ionizes the plasma at a high rate.
However, in the same set of experiments it was found that it was not possible with the existing setup to do measurements for the other phases of the discharge, which are the ignition, pinch, and decay phases. This is due to a too high level of background radiation emitted by the plasma in those phases. A background subtraction method was used to distinguish the Thomson scattered signal from the background, but random pulse-to-pulse fluctuations in the background prevented this method from working sufficiently well. In principle, longer experiments (i.e., integration over more discharge pulses) could be used to reduce the relative magnitude of the background fluctuations, but this approach is limited by long-term drifts in the behavior of the discharge, for example due to electrode erosion. Therefore, an alternative method to reduce the relative level of background radiation was needed. Such a reduction can, in principle, be achieved in two ways: the first is to acquire a certain TS signal from a smaller detection volume—that is, to use a narrower laser focus in combination with a narrower entrance slit for the spectrograph. The second is to record the signal in a shorter time.

In this chapter, we present the design and construction of a new setup for space and time resolved sub-ns Thomson scattering. It has been built as a flexible tool that can be applied to a range of different plasma types, but with the specific application mentioned above kept in mind. Both methods of background signal reduction have been pursued, with the main emphasis being on the latter.

The design of the new setup consists of two main parts. First, in Sec. 9.2, the need for an optical delay line in the laser path is discussed, and the layout of the laser table is given. In Sec. 9.3, the new triple grating spectrograph (TGS-II) is described, and we present the results of ray-trace simulations of this spectrograph. Finally, in Sec. 9.4 we describe the results of test experiments that we have performed to characterize the setup as a whole. In this chapter, we frequently refer back to specifics of the old system to indicate which changes have been made.

The new setup has already been applied for actual experiments on the tin vapor discharge; the results of these measurements are presented in Chapter 10.

### 9.2 Laser path

#### 9.2.1 Synchronization of laser and camera

A detailed description of the existing, previously used setup for TS can be found in Ref. [3] and the adjustments made for application to the tin vapor discharge are described in Chapter 8. In this old setup, a frequency doubled neodymium-doped yttrium aluminum garnet (Nd:YAG) laser pulse (at 532 nm) of about 170 mJ pulse energy and 7 ns full width at half maximum (FWHM) duration was applied, and the camera gate time was set to 5 ns (with the camera delay being optimized to the maximum of the laser pulse intensity). Compared to this previously existing situation, a reduction of the time needed for a single measurement means, in the first place, that a shorter gate time has to be applied for the
intensified charge-coupled device (ICCD) camera. In the new setup, we use a 4Picos camera from Stanford Computer Optics, which has a minimum specified gate time of 200 ps. To ensure sufficiently homogeneous illumination of the sensor by the intensifier, we applied a slightly longer gate width in practice.

To “fit” the laser pulse into the time window imposed by the camera gating, also a shorter laser pulse length had to be applied. In the new setup, the laser pulse is provided by the SL312 laser from Ekspla, Lithuania. Through stimulated Brillouin scattering, a Nd:YAG laser pulse is compressed from a few ns to 150 ps duration. The maximum output of the laser at the second harmonic is about 120 mJ per pulse.

Finally, to ensure that the TS signal indeed arrives at the camera just when the camera is being gated, a proper synchronization of the laser pulse and camera gate is required. Commercially available laser systems that provide the desired laser wavelength, pulse duration, and pulse energy, can in general not be operated with sufficient accuracy in the time domain; also, electronic synchronization signals from the SL312 and other laser systems exhibit too much time jitter—on the order of 1 ns—to use them for triggering the camera. With such poor synchronization, the effective average Thomson signal recorded during the gating of the camera, would be strongly reduced compared to the signal at the peak of the laser pulse.

We solved this problem by asking the laser manufacturer to place a fast photodiode inside the laser head; a beam splitter, located in the laser path before the amplification stage, sends a small part of the pulse energy to the photodiode, which converts it into a highly stable electric signal that can be used to trigger the camera. This approach has one obvious drawback: the trigger signal for the camera is only generated when the laser pulse has already been formed. It takes the signal some 15 ns to travel from the laser to the camera and the camera takes at least another 65 ns to gate its intensifier after arrival of the trigger signal. Therefore, the TS light should arrive at the camera at least 80 ns after the generation of the trigger signal.

In the old design, the optical path, from the laser head to the plasma and from the plasma to the camera entrance, was only about 10 m long, which corresponds to about 33 ns delay. Therefore, for the new setup, a dedicated delay line in the optical path for the laser pulse was needed to provide the additional delay.

A schematic overview of the design of the sub-ns TS system is shown in Fig. 9.1. The two main parts, apart from the plasma chamber itself, are the laser table and the triple grating spectrograph. The latter is discussed in Sec. 9.3 first we focus on the delay line.

### 9.2.2 Optical delay line

The first requirement for the delay line can be derived from the numbers quoted above: a sufficiently long additional delay of at least 50 ns has to be generated; this corresponds to a path length of about 15 m. However, equally important requirements are good efficiency (so
that sufficient laser energy can be focused into the plasma), ease and stability of alignment, and resistance against high laser powers.

This combination of requirements was met by making use of solid-glass corner cubes (CCs). These have the special property that any incident light ray (within a certain solid angle) gets reflected in the exactly parallel, opposite direction. The principle of using corner cubes for multipass optical cells was discussed by Vitushkin [11]. Klövekorn et al. [12] used a CC in a variable-length optical delay line that is independent of the mechanical quality of the translation stage.

In our setup, we use two CCs of unequal sizes. They are arranged to have parallel optical axes, with the axis of the largest one being directly above that of the smallest. The path of the laser beam is depicted in Fig. 9.2. A 45° mirror first directs the laser beam towards the top half of the large CC. The reflected beam is shifted both horizontally and vertically, and is directed towards the small CC. The beam is reflected and shifted horizontally by the small CC and returns to the large CC. After another reflection by the large CC, the
Fig. 9.2. Schematic drawing of the laser path between the corner cubes in the optical delay line. The parts of the optical path inside the corner cubes have been omitted for clarity. Distances between the optical elements are not to scale; in particular, in the actual setup, the large corner cube is placed much further away from the other elements, to obtain a long path length.

beam passes over the top of the small CC and along the side of the $45^\circ$ mirror, and reaches a normal-incidence (n.i.) mirror. From the n.i. mirror, the laser beam is reflected over exactly the same path; hence, it travels the distance between both CCs for a total of eight times, and exits the delay line in the same position and opposite direction it entered it in.

This delay line provides a total path length of about 18 m, which is limited just by the size of the table. This is more than the requirement derived above; however, any excess delay of the TS signal pulse can be compensated for by setting an appropriate delay in the internal delay generator of the ICCD camera.

The laser pulse is guided from the laser head to the delay line via a combination of a plate polarizer and a quarter wave plate. Initially, the pulse is horizontally polarized and gets transmitted by the polarizer, which is positioned at the Brewster angle with respect to the laser beam. After two passes through the quarter wave plate (one before and one after passing through the delay line) it has become vertically polarized, and it gets completely reflected by the polarizer. This way, both beams are separated, and next the returning beam is sent to the plasma chamber. The complete delay line is shown at the bottom left of Fig. [9.1] an enlargement of part of the delay line, including the entrance and exit, can be found at the top left.

The laser beam is focused into the plasma by a lens with 1 m focal length; the beam crosses the plasma in the horizontal direction. In the case of the tin vapor discharge setup, it was guided into and out of the vacuum chamber using Brewster angle windows, to minimize energy losses and production of stray light.
9.3 Spectrograph

9.3.1 Principle of the triple grating spectrograph
(Thomson) scattered laser light from the plasma is collected by two achromatic lenses and focused onto the horizontal entrance slit of a triple grating spectrograph, the TGS-II. Different positions along the entrance slit correspond to different positions along the plasma-laser beam intersection. The design of the TGS-II is based on that of the original TGS by Van de Sande [3]. In this design, three spectrographs are placed in series, of which the first two are arranged in a subtractive configuration. A mask, placed in the image plane of the first spectrograph, blocks the central wavelength. Next, the second spectrograph folds the spectrum back into a single line, so that the light can pass through an intermediate slit between the second and third spectrographs. Any light at the laser wavelength that was able to pass the mask due to imaging imperfections and stray reflections inside the TGS, is blocked by the intermediate slit. This arrangement turns the first two spectrographs into a strong stray light filter. Finally, the third spectrograph, behind the intermediate slit, provides the spectral dispersion needed to record the spatially resolved spectrum on the ICCD camera. The changes to the original design that have been made in the new one—as schematically depicted in Fig. 9.1—are discussed next.

9.3.2 Design of the TGS-II
In the new design, first of all, we use different gratings, with only 600 instead of 1800 grooves per mm. This is needed to expand the spectral range of the spectrograph from about 20 to 50 nm. Dependent on the orientation of the gratings, we can now record both wings of the TS spectrum symmetrically up to 25 nm away from the central wavelength, or select just one side of the spectrum and record it up to 50 nm away from the laser wavelength. The former method makes fitting of the spectrum to a theoretical curve easier, as information from both sides of the spectrum is available; however, the latter is necessary to do measurements on the highest density parts of the tin vapor discharge, for which the TS spectrum is strongly collective. In this case, the “notch mask” that blocks just the central wavelength, is replaced by an “edge mask”, since only one half of the spectrum is recorded by the camera.

Further, the focal lengths of all lenses but the last were reduced from 600 to 400 mm. This allowed us to apply smaller diameter lenses while maintaining a large collection angle. A larger focal length of 600 mm was used for the last lens to magnify the image 1.5 times and thus obtain better spatial resolution. Also, an extra mirror was placed just before the camera to fold the optical path, and be able to fit the design onto a somewhat smaller table than the original one.

A narrower entrance slit, of only 100 instead of 250 µm width was applied for two reasons: first, in combination with a narrower laser focus, it helps to improve the signal-to-
Table 9.1. Root mean squares $r_{\text{RMS}}$ of the distances of individual ray-trace points from each spot center, produced by zemax for different horizontal positions inside the plasma and different wavelengths. $x$ and $\Delta \lambda$ are defined as the shifts from the central position and the central wavelength, respectively, which is 532 nm in this case. For evenly distributed points in a round image spot, the spot radius $R$ equals $\sqrt{2}r_{\text{RMS}}$.

| $x$ (mm) | −5.0 | −2.5 | 0.0 | 0.0 | 0.0 | 0.0 | 2.5 | 5.0 | 5.0 |
| $\Delta \lambda$ (nm) | 0.0 | 0.0 | −25.0 | −12.5 | 0.0 | 12.5 | 25.0 | 0.0 | 0.0 | −25.0 |
| $r_{\text{RMS}}$ (µm) | 52.9 | 16.7 | 75.3 | 35.3 | 7.3 | 21.6 | 33.4 | 16.8 | 53.3 | 83.2 |

background ratio. Second, it partly compensates for the reduction of spectral resolution caused by the new set of gratings.

Finally, in our design, certain translation stages for optical elements that were present in the old design, were left out to improve the stability of the alignment against positional drift. Such drifts during a single experiment, or between experiments, can seriously reduce the efficiency of the spectrograph, and make frequent realignment of the system necessary. Instead, in certain cases we accepted a small deviation from the “ideal” optical path for certain optical elements. Such deviations can be compensated for by similar, but opposite shifts of other elements (which do still have the required degree of freedom of alignment), without seriously affecting the overall resolution of the system (see below). Specifically, of each set of two lenses, only one had a translation stage in the horizontal direction perpendicular to the optical axis. For the first spectrograph, the horizontal positioning of the focus was done by adjustment of the first folding mirror, which is located after the second lens. Further, we required a precise translation stage in the longitudinal direction for only one of each pair of lenses (with the other just being placed on a carrier on an optical rail).

### 9.3.3 Ray-trace simulations

The zemax optical design program was used to do ray trace simulations of the TGS-II, including the two lenses in front of it. In this way, we have been able to evaluate its spatial and spectral resolution. Calculations have been performed for multiple spatial positions inside the detection volume in the plasma, and for multiple wavelengths. Examples of image spots in the camera plane are depicted in Fig. 9.3. Table 9.1 summarizes the results.

The same program has been used to test the influence of possible misalignments, such as discussed above, on the final resolution. The following cases have been tested: (a) a rotation of the third grating by 0.2 degrees in the horizontal plane; (b) a horizontal shift of 1 mm perpendicular to the optical axis of the first lens of the last spectrograph; (c) a similar shift of the first lens of the second spectrograph, compensated for by an appropriate shift of the second lens of the same spectrograph; and (d) a similar shift of the first lens of the first spectrograph, compensated for by a shift of the first folding mirror, and a
Fig. 9.3. Ray-trace simulated image spots in ZEMAX for four different cases: (a) $x = 0$ mm, $\Delta \lambda = 0$ nm; (b) $x = 0$ mm, $\Delta \lambda = -25$ nm; (c) $x = 5$ mm, $\Delta \lambda = 0$ nm; and (d) $x = 5$ mm, $\Delta \lambda = -25$ nm, for a central wavelength of 532 nm. Here, $x$ and $\Delta \lambda$ represent shifts from the center of the image in the spatial and wavelength directions, respectively. The vertical bars represent sizes of 100 $\mu$m for parts (a) and (c), and 200 $\mu$m for parts (b) and (d).

longitudinal shift of the second lens (to keep the optical path length between that lens and the mask constant). In cases (a) and (b) no compensation by a shift of another element is necessary since these would just lead to a small shift of the image on the camera, which could easily be digitally compensated for during data processing. Therefore, we included compensating shifts of other elements only for cases (c) and (d).

For all tested cases, we found that the alignment deviations have little or no influence on the resolution of the spectrograph.

To evaluate the overall spatial and spectral resolution of the system, the numbers given in Table 9.1 should be compared to the width of the image of the entrance slit on the camera, and the width of the apparatus profile of the camera intensifier itself. These are 164 $\mu$m and on the order of 100 $\mu$m, respectively. For the points near the center of the image, these numbers are limiting the overall resolution; at the lower wavelength limit, and at the outsides of the spatial profile, the smallest resolvable features according to Table 9.1 are somewhat larger, up to just over 200 $\mu$m in diameter. However, in most experiments, only measurements near the spatial center of the image, $x = 0$ mm, have been done. Therefore, it can be concluded that ideally, the spatial and spectral resolution of the system—taking into account the geometrical magnification of the system, and the dispersion caused by the gratings—are about 70 $\mu$m and 0.4 nm, respectively.
9.4 Characterization

Test experiments have been performed to determine the efficiency of the delay line and the spectrograph, and to check that their combination works properly.

First, we tested the delay line efficiency by simply comparing laser power measurements just behind the laser exit and at the end of the delay line, just before the beam is focused into the plasma chamber. We have measured an efficiency of 78% for fully optimized alignment of the laser table. We found that, in practice, only a quick optimization of the orientation of both mirrors inside the delay line itself is needed to have it function properly; therefore, day-to-day optimization is just a matter of seconds. On the other hand, a full optimization of the efficiency of the optical path requires a bit more work. In this case, the angles to the optical axis of the 45° mirrors and the plate polarizer need to be as close as possible to their nominal values to minimize transmission and reflection losses, and the orientation of the quarter wave plate needs to be optimized to ensure that the returning beam from the delay line is perfectly vertically polarized.

Although the maximum output from the laser was about 120 mJ per pulse, in our actual experiments, we limited the laser pulse energy inside the plasma to about 50 mJ, mainly to limit disturbance of the plasma. It should be noted, however, that already at this setting, we found ourselves close to the limit of laser power that the delay line could handle. We found evidence of surface damage on the large corner cube at a n only slightly higher laser energy.

To test the triggering of the camera, we performed Rayleigh scattering measurements on atmospheric pressure nitrogen gas. Naturally, the mask inside the TGS-II was removed for this measurement. We found that the internal delay generator of the camera had to be set to about 13.7 ns additional delay, to optimize the recorded Rayleigh scattered signal. This number corresponds to our expectation. The time jitter in the triggering of the camera could not be measured exactly, but it was found to be less than 50 ps.

From Rayleigh scattered images with both slits removed from the TGS-II, we could also derive that after focusing, the full width at half maximum (FWHM) diameter of the laser beam was roughly 150 µm. As the effective focal width in the old setup was around 400 µm, the new width is a considerable improvement, which helped, in combination with the narrower entrance slit, to achieve a better signal-to-background ratio in TS experiments.

Measurements with the slits in place showed that the apparatus profile in the spectral direction for the complete detection system was 0.6 nm FWHM. This is more than sufficient for Thomson scattering on EUV discharge plasmas. With this resolution, reasonably accurate results can be obtained as long as the electron temperature $T_e$ is larger than about 1.0 eV.

The suppression of stray light and Rayleigh scattered signal was verified by measuring the rotational Raman scattered signal from nitrogen gas. Raman scattering, being an
Fig. 9.4. Example of rotational Raman scattered spectrum from nitrogen gas. In this case, the signal was integrated over all spatial channels.

inelastic scattering process, produces larger wavelength shifts from the laser wavelength than Rayleigh scattering does, but Rayleigh scattering has a much larger cross section. Therefore, observing a (rotational) Raman scattered spectrum without a strong Rayleigh and/or stray light peak in the center is a good test for the notch filter that has to block the central wavelength. Although the spectral resolution of the system—as given above—is not sufficient to resolve the individual Raman lines, the characteristic asymmetric shape of the overall spectrum could be clearly recognized; see Fig. 9.4 for an example.

In our TS experiments, we used a polarization filter to remove the plasma emission background from the TS signal. For this, we made use of the fact that the TS signal is fully linearly polarized in the vertical direction (as it enters the TGS), whereas in principle, the plasma emission is unpolarized. Hence, if a horizontally polarized image is subtracted from the vertically polarized one, only the TS signal is left in the result. This procedure makes polarization and wavelength dependent sensitivity measurements of the TGS-II necessary. Such measurements have been done with a ribbon lamp in the plasma position, and a rotatable polarization filter just in front of the second folding mirror. Later, the same filter was also used in TS experiments.

Our test experiments have shown that the new setup is suitable for Thomson scattering experiments. It can be used to study fast phenomena in plasmas in a spatially resolved way. When appropriate synchronization is available, up to 1 ns time resolution can be achieved if the plasma jitter is low enough. If the plasma setup can accept a trigger signal that comes less than about 60 ns in advance of the laser pulse (corresponding to the length of the delay line), a time resolution as low as 300 ps can even be achieved.

After the initial construction and characterization of the setup was finished, several series of TS experiments have been performed on the tin vapor discharge. To allow for a detailed discussion of the discharge evolution and the related plasma physics, these experiments
and their results are presented separately in the following chapter.

References


Chapter 10

Sub-ns Thomson scattering on a vacuum arc discharge in tin vapor

Abstract

In a previous series of Thomson scattering (TS) experiments on an EUV producing vacuum arc discharge in tin vapor, background radiation emitted by the plasma was found to make measurements impossible for all parts of the discharge except the prepinch phase. To reduce the level of recorded background radiation, we have built a setup for time and space resolved sub-ns TS. Results obtained with this new setup are presented for experiments on previously inaccessible parts of the discharge—the ignition phase, pinch phase and decay phase. For the first two, measurements have been performed at different heights in the plasma. Electron densities for the pinch phase have been derived. For the decay phase, the electron densities confirm previous Stark broadening data. From the overall results, a more complete picture of the plasma evolution can be formed.

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10.1 Introduction

Several types of pulsed discharge plasmas have in recent years attracted attention due to their capability of emitting extreme ultraviolet (EUV) radiation relatively efficiently; see, e.g., Refs. [1–6]. This capability makes them potential candidate light sources for the next generation of semiconductor lithography, in which high-power sources of radiation in a 2% band around 13.5 nm are required. Certain types are being developed further to improve their characteristics in a.o. the fields of output power, lifetime and cleanliness (see, e.g., Refs. [7, 8] and several contributions in Ref. [9]).

In further development and optimization of EUV producing discharges, a better understanding of the discharge evolution can be of great help. Such understanding could come from direct observation, or from the results of computer modeling. However, computer models in general need measurements of plasma parameters—such as electron temperatures and densities—to test their validity.

One specific type of discharge plasma is a laser-triggered vacuum arc discharge in tin vapor. In our EUV Laboratory at ASML, an experimental version of such a type of source, originally from the Institute of Spectroscopy of the Russian Academy of Sciences (ISAN) in Troitsk, Russia, has been in operation since early 2003. Results from time-resolved imaging and spectrometry in the EUV range on the plasma have been presented in Chapter 5 [10]. The setup consists of two electrodes, of which the cathode (the bottom electrode), is covered with a layer of liquid tin. Both electrodes are electrically connected to a ring of capacitors and separated by a vacuum, or a background gas at very low pressure. See Fig. 10.1 for a schematic picture. Before the start of the discharge, the capacitors are charged to a potential of 4 kV. Figure 10.2 shows a typical plot of the electric current during the discharge pulse. The pulse evolution can be described by splitting it up into four main phases, which will be briefly described here.

In the ignition phase (1), caused by the firing of a laser pulse onto the surface of the cathode, a plume of vaporized and partly ionized tin expands into the vacuum until it
reaches the edge of the anode. As soon as a sufficiently conducting path has been created, a discharge starts, and an electric current heats and ionizes the tin plasma further—the prepinch phase (2). Under influence of the azimuthal magnetic field, associated with the strong electric current, the plasma starts to compress in the radial direction, until it finally reaches a narrow, elongated shape: the pinch phase (3). Due to the increase in density that is caused by the compression, the emission of EUV radiation is strongest in this phase. Because of the lack of confinement of the plasma in the axial direction (as the magnetic field confines the plasma only in the radial direction), and the finite amount of energy stored in the capacitors connected to the electrodes, the plasma quickly starts to expand and decay—the decay phase (4)—until finally vacuum between the electrodes is restored for the next discharge pulse.

Apart from the measurements mentioned above, two types of plasma diagnostics have been applied to characterize the evolution of the plasma of this source; these are Stark broadening and Thomson scattering (TS); see Chapters 7 and 8 respectively [11, 12].

Stark broadening of spectral lines of tin ions was used to measure electron densities. Spatially resolved measurements were difficult to perform since, as it is a passive spectroscopic technique, measurements are always over a line of sight. Fortunately, the intensity of the line radiation is strongly dependent on the local density, so that the spectrum measured along a line through the center of the plasma quite closely represents the peak electron density in a cross section of the plasma.

Spatially resolved measurements are more straightforward with TS, an active spectroscopic technique. A laser pulse is fired into the plasma, and the spectrum of light that is scattered by free electrons is recorded, and, after processing, compared to a theoretical spectrum. With this technique, not only electron densities, but also temperatures can be measured. Results for the prepinch phase of the tin vapor discharge have been presented
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in Chapter 8. The applicability of this technique was limited to the prepinch phase, since in the other parts the accurate determination of the TS spectrum was impossible. The cause were random fluctuations in the level of background radiation, emitted by the plasma itself. Yet, measurements for these other phases were desired to obtain a more complete picture of the plasma evolution. The ratio of the TS signal to the background level would have to be improved to make reliable measurements for the other phases possible.

Given the possibility of plasma disturbance by the laser, a simple further increase of the laser pulse energy (which is, in principle, proportional to the detected TS signal intensity) was undesirable. If the TS signal intensity could not be increased, the level of background radiation would have to be lowered. Our main approach to achieve this was to reduce the time that is needed to record a single measurement. For this, we have designed and built a new setup for sub-ns Thomson scattering. Details on this design and characterization of the final setup are given in Chapter 9 [13].

In this chapter, we present the results of sub-ns Thomson scattering on the tin vapor discharge. First, in Sec. 10.2, properties of the new setup are summarized, and the experimental procedure is explained. In Sec. 10.3, the experimental results are presented and discussed separately for the decay phase, the ignition and prepinch phases, and the pinch phase of the discharge, respectively. Finally, our main conclusions are summarized.

10.2 Experiment

Thomson scattering experiments can give information about electron densities and temperatures in a plasma simultaneously. In this technique, a laser pulse is focused into a plasma, and the light scattered by free electrons is recorded. A double Doppler shift due to the velocities of the electrons causes a broadening of the spectrum of the scattered light. In case of a Maxwellian electron energy distribution and noncollective scattering, the resulting spectrum has a Gaussian shape, the width of which is directly related to the electron temperature. The intensity of the scattered light is a measure of the electron density. On the other hand, at sufficiently high ratios of electron density over electron temperature, \(n_e/T_e\), the scattering process is collective. In the collective scattering limit, the laser light is scattered off plasma waves rather than individual electrons, and sharp peaks are found in the scattered spectrum that are separated from the central laser frequency by the plasma frequency. For the intermediate case of partially collective scattering, the shape of the spectrum is given by the Salpeter approximation [14]. In this case, compared to non-collective scattering, not only the total scattered intensity but also the shape of the spectrum provides information on the electron density. The degree of collectivity is determined by the so-called scattering parameter \(\alpha\), which depends on the ratio \(n_e/T_e\) as indicated above.

In our setup, we combine a laser with 150 ps pulse duration, the SL 312 from Ekspla, with the 4Picos camera from Stanford Computer Optics, which has a minimum gate width
of 200 ps. The laser is a frequency-doubled neodymium-doped yttrium aluminum garnet (Nd:YAG) laser at 532 nm. A photodiode signal is used to synchronize the camera to the laser pulse; an optical delay line on the laser table provides enough time for the operation of the triggering electronics of the camera. In our experiments, the laser pulse energy in the plasma was limited to 50 mJ; the pulse energy directly from the laser had to be somewhat higher due to losses in the optical path of the laser beam. The waist diameter of the laser focus was measured to be about 150 µm.

We used a triple grating spectrograph (the TGS-II) to record the Thomson scattered light. The design of this spectrograph was based on the original design of the setup built by Marco van de Sande [15]. In this design, three separate spectrographs are placed in series. The first two are arranged in a subtractive configuration. A notch or edge mask is located behind the first spectrograph, and an intermediate slit behind the second. In this arrangement, the first two spectrographs serve as a powerful stray light filter. In our case, the maximum achievable spatial and spectral resolution of the spectrograph are about 70 µm and 0.6 nm, respectively. The lowest electron temperature that can be measured reliably with this setup, is about 1 eV.

The TGS-II covers a wavelength range of about 50 nm width in total. The actual range can be changed by an appropriate rotation of the three gratings; either the entire TS spectrum can be recorded symmetrically up to 25 nm from the central wavelength on both sides, or only one side of the spectrum can be covered up to 50 nm from the central wavelength. This is done when particularly broad spectra are expected.

The procedure for recording of the TS spectra and their subsequent processing, was largely similar to the procedure followed for our previous experiments; see Chapter 8. First of all, before each series of experiments, we recorded a spectrum of rotational Raman scattering in nitrogen gas, in order to obtain an absolute intensity calibration of the system.

Further, during TS experiments, a rotatable polarization filter in front of the camera was used to separate the plasma background from the TS signal. Here, we made use of the fact that the Thomson scattered light has only one polarization direction, while in principle, the background is unpolarized. We measured a signal for two orthogonal polarizations both with the laser pulse present, and subtracted one signal from the other to obtain just the Thomson spectrum. This way, we could eliminate any systematic effect that the presence of the laser pulse may have on the emitted plasma spectrum. Before subtraction, both signals were corrected for the polarization and wavelength dependent sensitivity of the TGS-II.

In our experiments, recorded signals from typically 100 laser shots were integrated to form one camera image, to partially average out the background fluctuations. Pixels in the camera images were grouped in both spatial and spectral directions, to form new “superpixels” which have a size that roughly matches the resolution of the system, namely 181 µm (spatially) × 0.34 nm (spectrally). This procedure, called binning, reduces the noise in the signal and the amount of data to be processed. After binning, each image was
corrected for the corresponding polarization-dependent sensitivity, and the background subtraction was performed.

Next, for each horizontal position along the laser beam, the obtained TS spectrum was fitted to the theoretical curve, giving the local electron temperature $T_e$ and density $n_e$. For the theoretical shape of the curve we used the Salpeter approximation [14], with a numerical approximation for the plasma dispersion function. More details on the fit function are given in Chapter 8.

Even though an absolute intensity calibration from rotational Raman scattering was available, the total scattered intensity was typically left as an additional free parameter, the intensity factor $\xi$. In other words, $n_e$ and $T_e$ were derived from just the shape and width of the spectrum, without actually using the integrated intensity. Thus, errors that may be present in the calibration experiment and those that are caused by alignment drifts during TS experiments, could be avoided. This was possible because of the relatively large values of the scattering parameter, $0.6 < \alpha < 2.3$, in nearly all cases.

The intensity factor is further influenced by the spatial and temporal gradients and jitter of the plasma. In the ideal case, $\xi$ would always be unity—that is, the measured intensities would be exactly equal to the theoretical ones. However, if, for example, a plasma of certain density is hit by the laser pulse in only 50% of all shots, an intensity factor $\xi = 0.5$ results. Now, the directly fitted electron density $n_{e,\text{fit}}$ represents the actual density in the plasma. On the other hand, the effective density $n_{e,\text{eff}} = \xi n_{e,\text{fit}}$ represents the density averaged over all shots, including the ones in which the plasma was “missed”.

In the limit of noncollective scattering, the effects of $\xi$ and $n_{e,\text{fit}}$ on the spectrum cannot be separated, and only their product $n_{e,\text{eff}}$ provides a meaningful number. In such cases (typically when $\alpha$ was smaller than about 0.6), $\xi$ was kept fixed during the fitting procedure. The same thing was done in a very limited number of cases in which the shape of the spectrum was particularly disturbed by background radiation. Good estimates for $\xi$ could be derived from the values in neighboring data points.

Finally, for each time step, the maximum fitted electron density $n_{e,\text{max}}$ and a weighted average $T_{e,\text{av}}$ were determined over the entire horizontal spatial profile. For the relative weight of each $T_e$ value, needed to calculate $T_{e,\text{av}}$, the effective density $n_{e,\text{eff}}$ at the same spatial position was used. $n_{e,\text{max}}$ and $T_{e,\text{av}}$ could be used to characterize the plasma as a function of time during the discharge pulse.

In this procedure, only the relative values of $\xi$ within each spatial profile are relevant, and the absolute values are not important. However, it is worth mentioning that within each profile, the maximum value of $\xi$ was typically between 0.4 and 0.8. The difference from unity can be ascribed partly to some instability of the discharge, but mainly to a certain drift of the alignment of the setup during each experiment. In most cases, the value of $\xi$ was lower at the sides of the profile than in the center, which could be the result of some positional instability of the plasma, or of the dynamical processes of expansion and compression that take place in the plasma evolution.
10.3 Results

In the following three subsections, the experimental results will be discussed for the decay phase, the ignition and prepinch phases, and the pinch phase, respectively.

10.3.1 Decay phase

The results of a scan through the decay phase of the discharge have been plotted in Fig. 10.3. For this scan, the wavelength range of 507 to 557 nm was used. In the same plot, results from previous Stark broadening measurements, as presented in Chapter 7, are shown.

It is interesting to see that the TS measurements closely reproduce the Stark densities in the decay phase; even some detailed features in the shape of the curve between $t = 1$ and $2 \mu s$ can be recognized. It is worth mentioning that the decay of the electron density is much slower than would be expected from a simple expansion of the pinch plasma; the main cause is additional evaporation of tin from the cathode surface due to the energy absorbed during the discharge—as observed in Chapter 5. Further, an oscillating electric
current between both electrodes is still present and still delivers energy to the plasma in
this phase.

In our previous work on Thomson scattering, described in Chapter 8, the possible influence
of laser absorption on the plasma has already been mentioned. The possible effect of
laser energy absorption in line transitions (as discussed there for the early prepinch phase)
of Sn ions is also present here, and could lead to a detectable increase of \( n_e \) of about 30% in
the most extreme case. However, given the good match with the Stark broadening results
(for which no laser pulse was present), the effect seems to be very limited here.

On the other hand, it is likely that laser absorption has significantly influenced the
electron temperature in the early decay phase (about 0.3–0.6 \( \mu s \) on the time scale). For
this part, one would expect a lower temperature, for example such as suggested by the
dotted line in Fig. 10.3. The main reasons to expect a lower temperature are the additional
evaporation of tin, as mentioned above, which would lead to a relatively cool plasma; the
presence of strong line radiation of relatively low ionization stages of tin—which can only
exist if the temperature is not too high; and the absence of strong EUV emission in other
experiments at this time in the evolution of the discharge; see Sec. 5.3 [10].

In order to verify the laser-induced increase of \( T_e \), we have performed separate laser
absorption measurements. These revealed absorption of up to 4% of the laser pulse energy
in the early decay phase. This number corresponds to an energy addition of 2 mJ to the
plasma. If we assume an electron density of \( 1.5 \times 10^{24} \text{ m}^{-3} \), a total length for absorption of
about \( l = 5 \text{ mm} \), an average beam diameter of \( r = 100 \mu \text{m} \), and an increase of the electron
temperature by 15 eV, the energy needed for electron heating amounts to

\[
E = n_e l \pi r^2 \frac{3}{2} k_B \Delta T_e = 0.85 \text{ mJ},
\]

a number that is somewhat smaller than the 2 mJ reported above. However, the electron
temperature increase as derived from Fig. 10.3 is an effective number integrated over the
laser pulse. The accumulated temperature increase at the end of the pulse could be larger
than this value. Further, the increase in excitation and ionization of Sn atoms and ions,
as mentioned above, could form another, although probably smaller, destination for the
absorbed laser energy. In other words, the actual heat capacity of the plasma is larger
than the value used in Eq. (10.1) above.

10.3.2 Ignition and prepinch phases

To study the ignition and prepinch phases in more detail, measurements have been done
for different vertical positions in the plasma (or, equivalently, different distances to the
cathode). The cross sections of the plasma for the different experimental series are depicted
schematically in Fig. 10.4. For these measurements and those presented in the following
subsection, the spectrograph alignment was shifted to the wavelength range of 482 to
532 nm. This is because of the high electron densities that were expected for the pinch
phase, which, in the case of collective TS, lead to spectra that extend far away from the laser
10.3 Results

Anode

Cathode

Fig. 10.4. Cross section of the discharge electrodes, with horizontal lines corresponding to the laser beam positions for different TS experiments. The different line patterns match those of the plots in Fig. 10.3. Distances from the cathode are, from bottom to top, 0.65, 0.85, 1.2, and 1.9 mm.

wavelength (see the next subsection). The lower half of the spectrum (in the wavelength domain) was chosen rather than the higher one, since it appeared to have a more “clean” background spectrum.

In Fig. 10.5, the results from these experiments are represented by the various patterned lines. From these plots, it can be clearly observed that a plasma appears first near the cathode, a relatively short time after the ignition laser pulse was fired. Later, while at the lowest positions the electron density and temperature are decreasing somewhat, a less dense and colder part of the plasma appears further away from the cathode. From the density plots, a velocity of the plasma front of about $2 \times 10^4$ m s$^{-1}$ can be derived.

Further, it can be concluded that the electron density near the anode reaches a few times $10^{22}$ m$^{-3}$ before the discharge starts. The moment of the start of the discharge was detected by a current probe, but it can also be derived from the fact that at this time, the electron temperature near the cathode starts to increase again—which is most likely caused by Ohmic heating due to the electric current. The electric current leads to fast ionization and heating of the plasma. In the second half of the prepinch phase, compression of the plasma starts to contribute to a further increase of electron density. To illustrate this, the plasma radius derived from background radiation for one of the experimental series has been plotted in Fig. 10.6.

In Chapter 8 it was mentioned that the electron temperature in the prepinch phase generally has a “hollow” shape. This phenomenon has been confirmed by our new experiments. It is more pronounced in the higher profiles, as these have lower temperatures at the start of the electric current. By contrast, the profiles of the laser-induced plasma in the ignition phase have their highest electron temperatures in the center. Examples of density and temperature profiles are shown in Fig. 10.7.

Like in the decay phase, illumination by the laser may cause a small relative density increase in the ignition phase and first part of the prepinch phase. Theoretically, it is
Fig. 10.5. Electron densities and temperatures derived from Thomson scattering, as a function of time (relative to the ignition laser pulse) and for different heights in the plasma. The various patterned lines correspond to measurements in the ignition and prepinch phases; the vertical bars represent density measurements for the pinch phase.

Fig. 10.6. Plasma radius derived from the background radiation for the experimental series recorded at 0.85 mm from the cathode. Compression already starts around \( t = 200 \) ns, but becomes much stronger only after \( t = 230 \) ns. Note that for the last data point, the measured radius may be limited by the resolution of the spectrometer.
the strongest in a high density, cool (1–2 eV) plasma. In our results, we have either low densities or higher electron temperatures, so that the effect is probably very limited. Also, the influence on $T_e$ is expected to be very small in this case, as on the one hand the particle densities are much lower than in the decay phase, and on the other hand, the pulse duration of the laser is much shorter than in the previous experiments, so that there is much less time available for electron heating.

### 10.3.3 Pinch phase

In the pinch phase, fitting of measured TS spectra to a theoretical spectrum, defined by single values for $n_e$ and $T_e$, was found to be impossible. The duration of the pinch itself is only on the order of 10 ns, and the diameter is around 200 $\mu$m or less. Therefore, both spatial and temporal gradients of the plasma parameters are very large. Even gradients of $n_e$ and $T_e$ across the laser beam can be significant. Also, jitter in the timing ($\sim 3–5$ ns) and spatial position of the pinch plasma is present and plays a significant role. Hence, the recorded signal is always a mixture of signals from regions or periods of time with different densities and temperatures.

Theoretically, for the plasma parameters expected for this part of the discharge, almost the complete electron contribution to the TS intensity is concentrated in sharp peaks separated from the central laser frequency by the plasma frequency, which is a function of the electron density only. Therefore, the electron density could be determined just from recording the position of the peak in the spectrum.

In practice, the sharp peak at the plasma frequency is “smeared out” to a broader feature, reflecting an electron density distribution rather than a single value. An example of such a spectrum is shown in Fig. [10.8](#). Similar distributions have been recorded for
different distances from the cathode, and they have been found for different positions on
the time scale. The upper and lower limits of these distributions are indicated by the
vertical bars in Fig. 10.5.

Two trends can be observed in the results: first, the time at which the pinch effect occurs,
depends on the height in the plasma; pinching is slower at higher positions. This is known
as the “zippering effect”, which is caused by the fact that the magnetic field in the plasma
varies with height. Second, the densities in the pinch seem to decrease with distance from
the cathode. The central density of the distribution varies from $n_e = 3.1 \times 10^{25}$ m$^{-3}$ near
the cathode to $0.8 \times 10^{25}$ m$^{-3}$ close to the anode. This reflects the fact that already at the
start of the discharge, the atomic density of tin is highest near the cathode, so that at this
position, more material is available for the pinch plasma.

Averaged electron temperatures could in principle be determined from the integrated
intensities of the spectra. This follows from the fact that, in the limit of strongly collective
scattering, the TS intensity tends to a constant factor times the electron temperature. How-
ever, plasma jitter and gradients, and alignment imperfections, also affect the measured
intensity. Further, the method would be highly sensitive to any signal offset that remains
after background subtraction, which is a relatively difficult procedure in this phase. Fi-
nally, inverse bremsstrahlung absorption of laser radiation could become significant for the
highest recorded electron densities; theoretically, it could lead to a temperature increase of
up to about 10 eV. Therefore, we did not attempt to perform actual electron temperature
measurements. However, in general, the intensities are roughly consistent with $T_e$ values
of 25–40 eV.
10.4 Conclusions

After our previous findings from Thomson scattering experiments on a tin vapor discharge in the prepinch phase, we have designed and built a new setup for sub-ns Thomson scattering. With the new setup, we have been capable of probing parts of the discharge that were previously inaccessible to Thomson scattering experiments. The evolution of the tin vapor discharge has now been described more quantitatively and in more detail, especially for all parts of the discharge other than the prepinch phase.

Specifically, for the ignition phase we have found electron densities and temperatures near the cathode of up to $2 \times 10^{23} \text{ m}^{-3}$ and around 10 eV, respectively, while those further away from the cathode both were much lower, and in fact, the discharge current did not start until the density near the anode reached about $2 \times 10^{22} \text{ m}^{-3}$. Next, the plasma behavior in the prepinch phase roughly followed the results already found in our previous work. We can now confirm that the fast increase in electron density is both the result of further ionization and the onset of plasma compression. Another effect, the hollow temperature profile, has also been confirmed, and it was found that it is strongest further away from the cathode, and that a similar effect is not present in the ignition phase. We have been able to determine electron densities for the pinch phase of the discharge, and have found that these decrease with distance from the cathode. Further, the time of pinching increases with distance from the cathode. And finally, electron density measurements for the decay phase provided an independent confirmation of the Stark broadening results for a regime for which they were not previously available. At the same time, the good match with Stark broadening data is an indication that additional ionization due to the presence of the laser pulse is limited.

Some limitations were found for the interpretation of the TS results, though. Care should be taken especially in those cases where strong line radiation is present. Further, although it was possible to probe both electron densities and temperatures in the compressing plasma, once the pinch plasma has reached its final, narrow radius, inherent gradients and plasma jitter were found to make independent determination of the electron temperature practically impossible.

However, in general, we have demonstrated that it is possible to measure extremely wide ranges of $n_e$ and $T_e$ values (over more than four, and almost two orders of magnitude, respectively) in what is basically a single experiment.

References


Chapter 11

General conclusions

Abstract
The main goals of the work presented in this thesis can be subdivided into the further development of optical diagnostic methods for application to EUV producing discharges, and improving the understanding of the physical properties of these discharge plasmas. This chapter presents the main conclusions regarding the various experimental methods. Further, the main phenomena that have been observed using these methods are discussed in relationship to each other, and general conclusions regarding the underlying plasma physics are drawn. Finally, some recommendations for future work are given.
Chapter 11: General conclusions

The main goals of the work described in this thesis have been twofold: the first aim was to apply experimental plasma diagnostics and other methods to EUV producing discharge plasmas, in order to improve the understanding of their physics—more specifically, the relationships between elementary plasma processes and the plasma dynamics. Two types of discharge have been the subject of investigations: a hollow-cathode triggered discharge in xenon gas, and a laser-triggered discharge in tin vapor. However, the extent of the obtained understanding is not necessarily limited to these specific devices, but may also comprise other devices with similar properties.

Second, the development of new diagnostic methods, and the adjustment or expansion of existing ones for application to EUV producing discharge plasmas, should be regarded as a goal in itself. The demonstration of their applicability, and usefulness in terms of obtained results, is of benefit for their future application to similar devices. Particularly, the experiments described in this work have shown the great benefit of applying all types of diagnostics in a time-resolved manner.

In this chapter, a summary of the main conclusions of the this work is presented according to the subdivision given above. First, the following can be stated about the diagnostic techniques that have been developed and/or applied to EUV producing discharge plasmas:

- The combination of a pinhole, a multichannel plate image intensifier, and a digital camera, is a cost-efficient way of recoding EUV images of the plasma during a discharge pulse. Sufficient time resolution and repeatability of the discharge can be achieved to obtain a good picture of the discharge evolution.

- The spectral separation of the various $5p-4d$ spectral features of different ions of both xenon and tin makes it possible to follow the presence, and—to a certain extent—the relative populations, of different ionization stages of those elements as a function of time during the discharge pulse, without the requirement of detailed modeling of the radiation. Symmetry considerations can be used to obtain partially spatially resolved spectra, although the results will still be integrated over a certain line of sight through the plasma.

- EUV spectra for the prepinch and pinch phases of discharge plasmas can be successfully simulated on a computer by using a model that applies relatively simple analytic expressions for the excited state densities. Through matching of simulated spectra to experimentally obtained ones, combined with some basic information about the plasma, such as its size, plasma parameters like electron density and temperature can be extracted from the recorded EUV spectra.

- Stark broadening measurements provide very useful information on electron densities in the cooler phases of tin discharges with relatively little effort. Reliable conclusions can be drawn without prior accurate knowledge of the plasma temperature. The ap-
General conclusions

The applicability of this method to EUV producing tin plasmas has been extended by the determination of four new Stark broadening parameters for Sn\(^{2+}\) ions.

- Thomson scattering (TS) can be applied as an experimental method to all phases in the evolution of EUV producing discharge plasmas. To our knowledge, Chapters 8 and 10 represent the first studies in which TS was applied to actual plasma devices designed for EUV generation. The problem of the high plasma emission background that emerges with the application of “conventional” TS systems can be solved or at least greatly reduced by the use of sub-ns pulses for both the laser and the camera gating. With this method, electron densities can be determined for the pinch phase of the discharge. For the other phases, even simultaneous space and time resolved measurements of the electron density and temperature are possible.

For these measurements, inverse bremsstrahlung has been shown to be unimportant as a mechanism of laser energy absorption in the plasma. On the other hand, the possibility of absorption of laser radiation in line transitions of low ionization stages should be carefully considered when the results of TS measurements are interpreted.

Several plasma-related phenomena have been observed by the use of these individual diagnostic techniques. Below, they are summarized roughly in order of discharge evolution. Furthermore, the results obtained using different techniques are discussed in connection with each other. Where possible, general conclusions are drawn regarding the underlying plasma physics.

- No EUV emission was observed from the expanding plasma plume in the ignition phase of the tin vapor discharge; however, electron temperatures up to about 10 eV have been measured for this phase. These are sufficient for strong, even multiple ionization. Hence, the ionization degree of the laser plasma cannot be considered to be the limiting factor for the onset of the discharge current. Rather, the high-current phase will begin when the plume has expanded enough to get sufficiently high overall particle densities near the anode. Thomson scattering experiments have indicated that the electron density should reach a value of about \(2 \times 10^{22} \text{ m}^{-3}\) for the discharge current to start.

- In both types of devices, EUV emission has been observed well before pinching, showing that ionization of the background gas (or vapor) to a relatively high degree already takes place before the start of plasma compression. Only a relatively small further increase of the average ionization degree seems to take place during compression, until just before the final pinch radius is reached. The pinch effect seems to serve mainly to create sufficiently high densities for efficient collisional population of excited states, and hence to produce strong EUV radiation, rather than to raise the electron temperature and thereby to further ionize the plasma.

The apparent absence of a further temperature increase during pinching is supported by the computer simulations of EUV spectra. The compressional heating of the plasma
may be compensated for by other effects. One is the increase of energy loss through radiation. Also, a decrease of Ohmic heating due to a declining electric current may play a role. The latter could be caused by the pinch effect itself, which leads to increased plasma inductance and resistivity, and by a drop in the electric current that is related to the inherently oscillating nature of the discharge circuit—the timing of the pinch effect is usually in the second part of the first current half-cycle. An additional possible explanation of the limited further increase of the ionization degree, as derived from the EUV emission, is given below.

Before pinching, EUV emission has been observed from a “ring-shaped” plasma. The phenomenon was the clearest for the hollow cathode discharge in xenon, for which images could be recorded along the symmetry axis, but it can also be seen from images of the tin vapor discharge, which could be recorded only from the side. Further, a hollow electron temperature profile has been measured in the prepinch phase of the tin vapor discharge, which is further confirmation that the effect is probably not device specific.

Two explanations can be given for both observed phenomena. First, the parameters of the plasma and the electric circuit are such that the “skin effect” could play a role at the onset of the high current phase. In other words, in the early prepinch phase, the current will flow mainly along the edges of the plasma, while later in the prepinch phase, and during compression and pinching, it will be more evenly distributed over the plasma. Hence, especially in the early prepinch phase, heating and ionization will take place mainly at the edges of the plasma. Second, the lower densities at the edges of the plasma lead to fewer collisions and the electrons can therefore acquire more energy. This means that they will be more capable of exciting and ionizing the atoms—so that again, ionization degrees needed for EUV emission are reached faster at the plasma edges.

The stable or even decreasing average ionization degree during pinching, as deduced from EUV spectrometry, could also in part be related to the hollow temperature profile. In the prepinch phase, the EUV emission will be dominated by the hot, relatively highly ionized outside regions. On the other hand, during compression the plasma may become more mixed, and in this case the trend in the overall emission will reflect the increased contribution of the cooler plasma from the center.

As a final note, with the plasma at the edges of the discharge being hotter rather than cooler than the plasma in the center, there is no reason to assume that there is considerable reabsorption in the outside regions, of the EUV radiation generated by the pinch plasma. On the contrary, such reabsorption will mainly take place either in the dense central part of the pinch plasma itself, or in the neutral background gas (in case of the xenon discharge).
In the tin vapor discharge, the distribution of the pinch electron density as a function of distance from the cathode clearly reflects the way the material is introduced in the inter-electrode space, namely by laser evaporation from the cathode surface.

The influence of capture radiative cascade is seen both in the EUV spectra of the decay phase of the xenon discharge, and in the shape of the contribution from doubly excited states as derived from the spectra of both xenon and tin plasmas. Even in non-recombining plasmas, the radiative cascade of doubly excited states can have a significant influence on the shape of the spectrum.

Both types of discharges show a preferential expansion of the plasma along the symmetry axis due to electromagnetic forces—specifically, due to the fact that the magnetic confinement of the plasma works only in the radial direction. As a consequence, the ionic and atomic debris as produced by these sources is not evenly distributed to all directions. This is actually an important advantage of the laser-triggered tin vapor discharge over other discharge concepts, since it allows the EUV radiation to be collected in a direction perpendicular to the pinch axis.

In the tin vapor discharge, considerable evaporation of additional material from the cathode takes place during and after the pinch phase. This effect can be observed in visible light images of the discharge (but not in EUV images), and was also derived from Thomson scattering and Stark broadening measurements. This additionally vaporized material does not contribute to the generation of EUV radiation, but it does add considerably to the production of debris. Therefore, a further optimization of this type of source in terms of debris production, by reduction of the additional evaporation of tin, might be possible.

This chapter is concluded by a few recommendations for future work. This discussion is limited to issues directly related to plasma diagnostics and their application, rather than the further development of EUV producing discharges or EUV lithography in general.

First of all, the experiments described in this thesis clearly demonstrate the usefulness of combining two or more different diagnostic techniques to probe a plasma. In many cases different techniques can confirm the results of each other (thereby increasing their reliability) or provide complementary information about the plasma. As an example from the present work, the Stark broadening and Thomson scattering experiments have provided independent data on the electron density. Further, the electron density $n_e = 3 \times 10^{25} \text{ m}^{-3}$ for the tin vapor discharge in the pinch phase, as derived from simulations of the EUV spectrum, has later been confirmed by a direct TS measurement. As already discussed above, pinhole images and TS profiles both indicate emission of radiation from a ring-shaped volume (directly, and by showing a strongly hollow temperature profile, respectively). Finally, in more general terms, quantitative measurements in these transient discharges always need to be
combined with plasma imaging in order to be able to relate the results of the measurements to the plasma dynamics.

Stark broadening measurements have provided much information about the coolest discharge phases. By appropriate selection of spectral lines, the method could be extended to hotter discharge phases, in which the low ionization stages Sn$^+$ and Sn$^{2+}$ are no longer abundant. The two main challenges related to such measurements are the relative difficulty of constructing a sufficiently high resolution spectrometer for the wavelengths at which higher ionization stages tend to radiate, and the absence of literature data on the Stark broadening of the spectral lines of those stages. The latter problem could be solved by performing more detailed quantum mechanical calculations of the appropriate Stark broadening parameters. Also, impurities of other elements, of which the line profiles can be more easily measured or for which Stark broadening data are more readily available, could be introduced into the plasma. Such an approach is presented in Ref. [1], in which lithium is added as an impurity to a vacuum spark in tin.

The spectral model described in Chapter 6 could be applied to a more complete set of input data, reflecting not just the pinch phase but also cooler parts of the discharge evolution. In this way, a pulse-integrated spectrum over a larger wavelength range could be obtained, which is of interest for determining the relative contributions of out-of-band radiation in different wavelength ranges to the overall plasma emission. It should be noted, though, that in the decay phases of discharge plasmas, radiative cascade could play an important role, which cannot be accurately simulated by the current analytic model—at least not as far as the details of the spectrum are concerned. Therefore, the accuracy of the results at larger wavelengths might be limited. Also, an accurate reproduction of the spectrum would require more precise determination of the wavelengths of optical transitions in the visible light range.

Finally, experimental results such as presented in this thesis can and should be used as benchmarks for existing and future computer models that are intended to describe the discharge evolution. Such models are indispensable for further development and optimization of discharge sources of EUV radiation, but their validity cannot be proven without comparison to good sets of experimental data.

References

Summary

Pulsed discharge plasmas are considered to be important candidate sources of extreme ultraviolet (EUV) radiation for application in future lithography tools for the high-volume manufacturing of computer chips. Two specific types of such plasmas have been the subject of research in this work: (1) a hollow-cathode triggered source, developed by Philips EUV in Aachen, Germany; and (2) a laser-ignited discharge in tin vapor, from the Institute of Spectroscopy (ISAN) in Troitsk, Russia. In the evolution of the discharge pulses of these and similar types of EUV sources, generally four different phases can be distinguished. After a device-dependent ignition phase, a strong current starts to flow (“prepinch phase”). A Lorentz force, associated with the electric current, causes a strong radial compression of the plasma (“pinch phase”). After this, in the decay phase, the plasma cools down, expands, and finally dies out.

For the further development and optimization of discharge plasma EUV sources, a better understanding of the plasma properties and dynamics is needed; to obtain such understanding, time-resolved measurements of the plasma properties are indispensable. Optical diagnostics are strongly preferred because they provide a lot of information about the plasma while in general they cause little or no disturbance of the plasma itself. In this work, a number of different optical diagnostic techniques have been applied to both types of discharges; their results are summarized below.

First of all, time-resolved imaging of the plasma, both in the EUV and in the visible light ranges of the spectrum, serves mainly to obtain basic, qualitative understanding of the evolution of the discharge pulse. The visible light images help to visualize the parts of the plasma that are not hot enough to emit EUV radiation. Plasma imaging has, for instance, helped to identify a preferential direction of expansion of the plasma along the axis of symmetry in both types of discharges, with supersonic velocities of roughly $4 \times 10^4 \text{ m s}^{-1}$.

Also, time and space resolved spectra have been recorded for the EUV wavelength range. These have shown, together with EUV plasma imaging, that high ionization stages already exist in a ring-shaped plasma in the prepinch phase, before the onset of compression. The recorded spectra have further been compared to EUV spectra produced by a computer model. This comparison gives information about which plasma processes play a prominent role in the studied part of the discharge; detailed consideration of, among others, radiative deexcitation and the influence of both Doppler and Stark broadening on the opacity of the radiation, proved to be important to produce good reproductions of the experimentally obtained spectra. Furthermore, the “lagging” of the ionization stage populations compared to the instantaneous electron temperature, had to be taken into account in the form of an effective net ionization rate—hence, the ionization stage population was shown to be essentially non-stationary. Finally, doubly excited states were shown to play a role in determining the shapes of the EUV spectra of both discharges. Best matches between
simulated and experimental spectra were obtained with electron temperatures near 25 eV and pinch electron densities of about $1 \times 10^{25}$ m$^{-3}$ and $3 \times 10^{25}$ m$^{-3}$ for the xenon and tin plasmas, respectively. These and other plasma parameters agree fairly well with the results of other diagnostic techniques.

The Stark broadening of specific spectral lines of tin ions in the visible wavelength range has been measured as a function of time during the evolution of the tin vapor discharge. A cross-calibration has led to the determination of four new Stark broadening parameters for lines of doubly ionized tin. From the spectral line widths, information about electron densities for various parts of the plasma evolution have been derived.

Finally, the Thomson scattering (TS) technique has been applied to the tin vapor discharge to determine space and time resolved electron temperatures and densities simultaneously. First experiments were performed with an existing TS setup. However, to obtain a better ratio between the TS signal and the background radiation, generated by the plasma itself, a new setup for sub-ns Thomson scattering has been designed and built. A laser with a shorter pulse duration, a camera with a shorter gate time, and improved synchronization between the two, together have enabled this signal-to-background ratio to be improved by more than an order of magnitude. This has greatly expanded the applicability of the TS technique to EUV generating discharge plasmas.

In application to the tin vapor discharge, electron temperatures and densities of up to about 10 eV and $2 \times 10^{23}$ m$^{-3}$, respectively, have been found for the laser-induced ignition plasma. Once the electron density near the anode reaches a value of around $2 \times 10^{22}$ m$^{-3}$, an electric current can start to flow. In the subsequent high-current phase, both densities and temperatures increase fast, with temperatures reaching to about 30 eV after 100 ns. Especially in the beginning of this phase, the plasma exhibits a hollow radial temperature profile. Compression leads to the pinch phase, in which electron densities on the order of $3 \times 10^{25}$ m$^{-3}$ are achieved. After the end of the pinch phase, additional evaporation of tin from the cathode results in a cooler plasma with relatively slowly decreasing electron densities, from initially above $10^{24}$ m$^{-3}$ to around $10^{21}$ m$^{-3}$ after 4 $\mu$s. Nearly all densities, except for those in the pinch phase, have been confirmed independently by the Stark broadening results.

The experiments described above have led, first of all, to a more complete understanding of the plasma evolution. Certain phenomena have been confirmed, others have been newly discovered—such as the aforementioned EUV emission from a ring-shaped plasma.

Furthermore, extensive information, both temporally and spatially resolved, has been gathered on the electron temperatures and densities in the tin vapor discharge. Such data could serve as a benchmark for future computer simulations of the evolution of discharge plasmas.
Samenvatting

Gepulste ontladingsplasma’s worden beschouwd als belangrijke kandidaten voor toepassing als bron van extreem ultraviolette (EUV) straling in toekomstige lithografie-apparaten voor de hoogvolumeproduktie van computerchips. Twee specifieke plasmatypes zijn in dit onderzoek nader bestudeerd: (1) een holle-kathodegetriggerde bron, ontwikkeld door Philips EUV in Aken (Duitsland), en (2) een met behulp van een laser geënitieerde ontlading in tindamp, afkomstig van het Instituut voor Spectroscopie (ISAN) in Troitsk (Rusland). Gedurende een ontladingspuls van deze en vergelijkbare EUV-bronnen kunnen in het algemeen vier verschillende fasen worden onderscheiden. Na een apparaatspecifieke ontstekingsfase, begint een sterke elektrische stroom door het plasma te lopen (“prepinch”-fase). De Lorentzkracht die hiermee verbonden is, veroorzaakt een sterke radiële compressie (“pinch”-fase). Hierna koelt het plasma af en expandeert het, tot het uiteindelijk uitsterft.

Voor de verdere ontwikkeling en optimalisatie van ontladingsplasma-EUV-bronnen is een beter begrip van de plasma-eigenschappen en -dynamica vereist. Hiervoor zijn tijdopgeloste metingen onmisbaar. Optische diagnostieken genieten sterke voorkeur omdat ze veel informatie over het plasma verschaffen, terwijl in het algemeen het plasma zelf niet of nauwelijks verstoord wordt. In dit onderzoek is een aantal verschillende optische technieken toegepast op beide ontladingstypes; de resultaten worden hieronder samengevat.

Om te beginnen dienen tijdopgeloste afbeeldingen van het plasma, zowel in het EUV- als in het zichtbare golflengtegebied, voornamelijk ter bevordering van het kwalitatieve begrip van de ontwikkeling van de ontladingspuls. De zichtbaar-lichtopnamen helpen die delen van het plasma te visualiseren die niet heet genoeg zijn om EUV-straling uit te zenden. Met behulp van dergelijke opnamen is onder andere bevestigd dat het plasma in beide ontladingstypes expandeert in een voorkeursrichting langs de symmetrie-as van de ontlading, met een supersonische snelheid van rond \( 4 \times 10^4 \) m s\(^{-1}\).

Daarnaast zijn ook tijd- en plaatsopgeloste spectra opgenomen in het EUV-golflengtegebied. Samen met de EUV-plasma-afbeeldingen hebben deze aangetoond dat al in een ringvorming plasma, vóór de start van de plasmacompressie, hoge ionisatietrappen voorkomen. De opgenomen spectra zijn verder vergeleken met gesimuleerde spectra uit een computermodel, met het doel informatie te verkrijgen over welke plasmaprocessen een belangrijke rol spelen in het onderhavige deel van de ontlading. Zo bleken gedetailleerde beschouwingen van stralingsdeexcitatie en van de invloed van Doppler- en Stark-lijnverbreding op de optische dichtheid van het plasma van belang om de experimentele spectra goed te kunnen reproduceren. Verder moest rekening worden gehouden met het achterlopen van de bezetting van de ionisatietrappen ten opzichte van de instantane elektronentemperatuur, door middel van het invoeren van een effectieve netto-ionisatiesnelheid. De bevolking van de ionisatietrappen bleek, anders gezegd, essentieel niet-stationair te zijn. Tot slot is aangetoond dat dubbel aangeslagen toestanden van belang zijn voor de vorm van de...
EUV-spectra in beide ontladingen. Een goede overeenkomst tussen de gesimuleerde en de experimentele spectra werd bereikt door het aannemen van elektronentemperaturen rond 25 eV en elektronendichtheden in de “pinch”-fase van ongeveer $1 \times 10^{25} \text{m}^{-3}$ en $3 \times 10^{25} \text{m}^{-3}$ voor respectievelijk de xenon- en de tinontlading. Deze en andere plasmaparameters komen overeen met de resultaten van andere diagnostieken.

De Starkverbreding is gemeten voor een aantal specifieke spectrale lijnen van tinioniën als een functie van de tijd gedurende de tindamp-ontlading. Door middel van een ijking aan de breedte van een andere lijn, zijn voor vier lijnen van dubbel geïoniseerd tin nieuwe Starkparameters vastgesteld. Uit de lijnbreedtes is informatie afgeleid over de elektronendichtheden in diverse fases van de ontwikkeling van het plasma.

Tot slot is de Thomsonverstrooiingstechniek toegepast op de tindamp-ontlading om gelijktijdig tijd- en plaatsopgeloste elektronentemperaturen en -dichtheden te kunnen bepalen. In eerste instantie is hiervoor een bestaande meetopstelling gebruikt. Om een betere verhouding te verkrijgen tussen het Thornonsignaal en de achtergrondstraling die wordt uitgezonden door het plasma zelf, is echter een nieuwe opstelling voor sub-ns-Thomsonverstrooiing ontworpen en gebouwd. Een laser met korte pulsduur, een camera met een korte sluitertijd, en een betere synchronisatie tussen beide, zijn gebruikt om de signaal-achtergrondverhouding met meer dan een ordegrootte te verbeteren. Dit heeft geleid tot een sterke uitbreiding van de toepasbaarheid van Thomsonverstrooing op EUV-producerende ontladingsplasma’s.

Bij toepassing op de tindamp-ontlading zijn elektronentemperaturen en -dichtheden tot respectievelijk circa 10 eV en $2 \times 10^{23} \text{m}^{-3}$ gemeten voor het laser-geïnduceerde ontstekingsplasma. Zodra de elektronendichtheid bij de anode een waarde heeft bereikt van ongeveer $2 \times 10^{22} \text{m}^{-3}$, kan een elektrische stroom gaan lopen. Door de sterke stroom stijgen zowel dichtheden als temperaturen vervolgens snel, tot na 100 ns een temperatuur van circa 30 eV wordt bereikt. Met name in het begin van deze fase vertoont het plasma een “hol” radieel temperatuurprofiel. Compressie leidt tot de bovengenoemde pinchfase, waarin de elektronendichtheid oploopt tot rond $3 \times 10^{25} \text{m}^{-3}$. Na het einde van de pinchfase zorgt additionele verdamping van tin voor een koeler plasma, met relatief langzaam afnemende elektronendichtheid, van initieel meer dan $10^{24} \text{m}^{-3}$ tot rond $10^{21} \text{m}^{-3}$ na 4 µs. Vrijwel alle dichtheden, behalve die in de pinchfase, zijn bevestigd door onafhankelijke Starkverbredingsmetingen.

De hier beschreven experimenten hebben in de eerste plaats geleid tot een beter begrip van de ontwikkeling van het plasma. Bepaalde verschijnselen zijn bevestigd; andere zijn nieuw ontdekt—zoals de voornoemde EUV-emissie door een ringvormig plasma.

Verder is uitgebreide informatie, opgelost in zowel plaats als tijd, verkregen met betrekking tot elektronentemperaturen en dichtheden in de tindamp-ontlading. Zulke gegevens zouden gebruikt kunnen worden als referentie voor toekomstige computersimulaties van de ontwikkeling van ontladingsplasma’s.
Hoewel het verrichten van promotieonderzoek en het schrijven van een proefschrift soms tamelijk eenzame bezigheden lijken (of misschien wel juist daarom), had dit werk nooit tot stand kunnen komen zonder de hulp en ondersteuning van een groot aantal personen. Op deze plaats wil ik hen graag de eer bewijzen die hen toekomt.

Allereerst gaat mijn dank uit naar Joost van der Mullen, mijn copromotor en dagelijks begeleider in de groep EPG. Joost, ondanks dat je me soms het tegendeel wilde doen geloven, heb ik ruimschoots geprofiteerd van onze discussies—met name op het theoretische vlak—en je doorwrochte commentaar op het geschreven werk. Daarnaast wil ik Joost, en Vadim Banine en Jos Benschop van ASML, bedanken voor het feit dat ze vier jaar geleden het nu lopende onderzoeksproject gestart zijn, en mij de kans en hun vertrouwen hebben gegeven om me op dit onderwerp te storten.

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At this place, I would also like to thank my second promotor, Marco Haverlag, and the other members of my core committee—Prof.Dr. H.-J. Kunze, Dr. Konstantin Koshelev, and prof.dr. M.J. van der Wiel—for investing their time to read through this thesis and giving their final approval.

Een groot aantal (oud-)leden van de groep EPG heeft in een of andere vorm aan dit werk bijgedragen. Ik zal beginnen met degene die al het langst weg is: Marco van de Sande, de door jou nagelaten Thomsonverstrooingsopstelling en het bijbehorende proefschrift zijn voor mij van grote waarde geweest. Daarnaast zou ik je speciaal willen bedanken voor de nuttige e-mailcontacten bij mijn pogingen om “laser-induced breakdown” in xenon te creëren, en de latere, succesvollere inspanningen om zelf met Thomsonverstrooiing aan de slag te gaan.

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Dankwoord

van “debris” (puin) en de mitigatie daarvan sterk heeft vergroot. Veel succes met je eigen promotie-onderzoek!

Geen enkele experimentele onderzoeksgroep kan resultaten produceren zonder goede technische ondersteuning. Charlotte, Loek, Evert en Huib wil ik graag bedanken voor hun vele kleine en grotere bijdragen aan mijn werk. Charlotte zou ik daarnaast speciaal willen bedanken voor de ray-trace-simulaties in ZEMAX, die een belangrijk onderdeel vormen van het werk gepresenteerd in hoofdstuk 9 van dit proefschrift. Tot slot mag ik natuurlijk Rina niet vergeten (op straffe van een grote bos bloemen!). Een goede secretaresse is haar gewicht in goud waard.

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Erik Kieft
juli 2005
About the author

Erik Kieft was born in Wageningen, the Netherlands, on the 24th of May, 1977. He received his secondary education at the CLV in Veenendaal. From 1995 until 2001, he studied Applied Physics at the Eindhoven University of Technology. As part of his studies, he did a four month traineeship at the group Environmental Fluid Dynamics at Arizona State University in Tempe, Arizona (United States), on “Laboratory modeling of two-dimensional complex-terrain flow processes in a valley cross-section”. His graduation project in the group Equilibrium and Transport in Plasmas was carried out at TNO-TPD in Eindhoven. His Master’s thesis is titled “Mass spectrometry and ex-situ ellipsometry as analysis tools for plasma enhanced deposition of ZnO thin films”. From 1995 until 1998, he also studied Applied Mathematics at the same university. He passed the propedeutic exam and completed the first two years of the study program.

Erik started his PhD research project in the group Elementary Processes in Gas discharges at the Eindhoven University of Technology in October 2001. This thesis is the result of that project. The subject was the experimental study of discharge plasmas for EUV generation. As part of the project, he paid a research visit to the Laboratory of Plasma Spectroscopy at the Institute of Spectroscopy of the Russian Academy of Sciences (ISAN) in Troitsk, Russia, from October until December 2002.

During his PhD, he was also active as a board member of the AiOOE (the association for PhD students in Eindhoven), representing it at the meetings of the LAIOO (national representative body of PhD students), and as the secretary of VENI, the alumni association of the Department of Applied Physics at TU/e.
Publications related to this work

This thesis is for a large part based on the following publications:


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