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MASTER

Ultracold electron source for ultrafast electron diffraction

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Ultracold Electron Source
for Ultrafast Electron Diffraction

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Single-shot ultrafast electron diffraction (UED) was introduced as a viable way to measure ultrafast processes, such as structural transitions in materials. For UED, the dynamics in a sample are induced by a femtosecond laser pulse and ultrashort electron bunches are used to probe the ultrafast processes in the sample. The ultracold charged particle cloud (UCPC) source researched in this work is an extended electron source with the potential to produce electron bunches with the extreme quality required for single shot UED. The UCPC source uses rubidium atoms that are trapped and cooled in a magneto-optical trap. The rubidium atoms are near-threshold photoionized in a two step ionization process to form the ultracold charged particle cloud. The electrons in the ionization volume are accelerated by a DC electric field.

In previous work, nanosecond long, narrow bandwidth ionization pulses were used to create ultracold electron bunches. In this work, measurements of the transverse source temperature are presented for bunches that are ionized with femtosecond long, broad bandwidth ionization pulses. For near-threshold ionization laser wavelengths from 489 nm to 477 nm, temperatures were measured from 25 to 240 K. Very low bunch temperatures were measured despite the broad bandwidth of the ionization laser. Transverse coherence lengths up to 20 nm are reported, which is sufficiently long to perform diffraction experiments on typical protein crystal samples. Individual electron bunches contain typically $10^3$ electrons, less than what is ideally required for single shot UED. The temperature measurements are compared to simulations performed with a semi-analytical model based on electron trajectories in the combined potential energy function of the rubidium atoms and the electric field of the accelerator. The overall shape of the simulated temperature as a function of the ionization laser wavelength mirrors the measured data. From additional measurements, we saw that the transverse source temperature varies sinusoidally as a function of the polarization angle of the ionization laser. The mean value of the oscillations of the transverse source temperature increases monotonously for decreasing ionization laser wavelengths, as does the amplitude of the oscillations. This behavior is also confirmed by the model.

The first diffraction patterns that have been measured with the UCPC source setup are also reported in this thesis, measured with a graphene sample. With crystal domains that are several times smaller than the electron beam at the sample, the diffraction pattern consists of rings. The diffraction angles of the first two diffraction rings were measured as a function of the beam energy and match with values calculated using the grating equation. The angular spread determined from the measured diffraction patterns is however many times larger than what is expected based on the transverse source temperature. To improve experiments, a quadrupole lens is added to the setup to counter astigmatism and a second solenoid lens is added. It is left up to future researchers to use the improved setup.
Preface
# Contents

1 Introduction 3  
   1.1 Ultrafast electron diffraction 3  
   1.2 Quantification of beam quality 4  
   1.3 Ultracold charged particle beams 7  
   1.4 Scope of this thesis 8  

2 UCPC Concepts and modelling 9  
   2.1 Ultracold charged particle cloud 9  
      2.1.1 Rubidium 9  
      2.1.2 Cooling and trapping 11  
      2.1.3 Photo-ionization 12  
      2.1.4 Acceleration 14  
   2.2 Photo-ionization model 14  
      2.2.1 Excess energy 14  
      2.2.2 Narrow bandwidth model 16  
      2.2.3 Broadband model 19  
      2.2.4 Laser polarization 21  

3 Setup and measurement techniques 23  
   3.1 UCPC setup 23  
   3.2 Source emittance measurements 28  
      3.2.1 Waist scans 28  
      3.2.2 Ion spotsize scan 32  
      3.2.3 Independent emittance measurement 34  

4 Temperature and charge measurements 36  
   4.1 Wavelength dependence 36  
   4.2 Polarization effects 38  

5 Pragmatic diffraction theory 43  
   5.1 Scattered fractions 44  
   5.2 Ring pattern 46  

6 Diffraction experiments 51  
   6.1 Diffraction patterns 51  
   6.2 Experimental improvements 55  
      6.2.1 Point to point focussing 56  

7 Conclusions and outlook 60  
   7.1 Beam quality measurements 60  
   7.2 Diffraction patterns 61  
   7.3 Outlook 61
Chapter 1

Introduction

This thesis concludes the graduation project that was performed by the author in the group Coherence and Quantum Technology at the Eindhoven University of Technology. The work entails the characterization and the application of a novel electron beam source concept, the ultracold charged particle cloud (UCPC) source. The source is developed towards the goal of researching ultrafast dynamics by performing single shot ultrafast electron diffraction (UED). Successful implementation of this experimental method will unlock a new regime of sample characterization, important for e.g. biological and chemical research. The main challenge for single shot UED is the extremely high electron beam quality that is required. In this chapter, a short impression will be given of the important applications of ultrafast research, the beam quality is discussed quantitatively and the positioning of this work is discussed with respect to other work that has been performed using the UCPC electron source.

1.1 Ultrafast electron diffraction

The holy grail in chemistry would be to be able to make a movie of atomic and molecular motion [1]. Using such a movie, reaction mechanisms of e.g. chemical bond formation could be precisely determined, which would greatly increase the understanding and control of chemical experiments. The typical length scales that a camera should be able to resolve for such a movie is in the order of the size of individual atoms, which is 0.1 nm. Present day transmission electron microscopes (TEM) setups are able to resolve the required features, individual atoms in a lattice [2, 3]. The typical time scale that the atomic motion takes place is the period of the vibrations of the atoms in a solid, which is in the order of 0.1 ps [4]. The temporal resolution that is required to resolve the motion of atoms has never been combined with extreme high spatial resolution.

Another area that would greatly benefit from the use of a molecular video camera is physical biology [1]. The formation and the operation of proteins in organisms is not well understood [5]. Proteins perform their tasks by folding back and forth between predefined shapes that have the required functionality. In the muscle tissue of the body for example, proteins contract and relax collectively for the organism to be able to flex or relax the muscle [6]. Sometimes, the folding of proteins is influence on a larger scale, this can have serious repercussions. For example Alzheimer’s disease and CreutzfeldtJakob disease are both caused by proteins misfolding in the human brain. With a molecular camera, such processes could be studied in detail, providing vital information towards finding a cure.

Since the development of the femtosecond laser system in the 1980s, the field of research of ultrafast dynamics has been opened up [7]. Ultrafast electrons diffraction (UED) is one of the techniques that is applied to measure effects that take place on the ultrashort timescales [8]. Ultrafast processes are pumped using femtosecond laser pulses. Part of the femtosecond laser pulse is split off and is used to create an ultrashort electrons bunch by photo-ionization. These electrons are accelerated, focussed on the pumped sample and scattered in the sample. The scattered
electrons then form a diffraction pattern, which is recorded using a detector. The diffraction pattern changes as a function of the delay between the pump laser pulse and the probe electron bunch. By varying the delay between the pump and the probe beams, the ultrafast dynamics that are driven in the sample can be measured. A schematic representation of the pump-probe experiment is displayed in figure 1.1. Using non-single shot UED, Siwick et al. studied the process of melting in thin gold and aluminum samples [9].

In other work on UED, single electron bunches are utilized [3]. Single electron bunches do not degrade due to Coulomb repulsion, so that both the longitudinal and the transverse beam quality are maintained. To build up a complete diffraction image, the pump-probe experiment is repeated many times. This method is only applicable if the process that is pumped in the sample is reversible and highly reproducible and if the probe beam or pump beam do not damage the sample. Protein dynamics cannot be studied using this many shot approach, since the process of misfolding is irreversible. To study irreversible processes, or samples that have a very low damage threshold, the diffraction pattern must be recorded in a single shot. When many electrons are in the small volume of the ultrashort electron bunch, the bunch will very quickly expand due to the Coulomb repulsion, becoming too long to measure the desired ultrafast dynamics. Using a lens as indicated in figure 1.1 the transverse size of the bunches can be decreased. For elliptical bunches with linear internal fields, the longitudinal expansion can be reversed using a radiofrequency cavity [11].

Many effects were mentioned here that play a role in performing single shot UED measurements. To perform UED measurements with sufficient charge to be able to measure a diffraction pattern with only one bunch, we saw that the quality of the bunches should be very high. In the following section, the quality of the bunches is quantified, to provide a framework in which to discuss the requirements of UED.

### 1.2 Quantification of beam quality

When describing electrons as individual particles, then every electron has its own position $\vec{r} = (x, y, z)$ and momentum $\vec{p} = (p_x, p_y, p_z)$. The whole ensemble of electrons, all with a unique position and momentum, can be described by a distribution function in six dimensions [12]: $f(x, y, z, p_x, p_y, p_z)$. A collection of particles is called a beam or bunch, if they share a common direction of travel. In other words, for a beam one momentum component is much larger than the other two. By convention, the $z$-axis is chosen to align to this average direction of travel of the electrons. In case the electrons travel at constant speed and noting that the motions of the electrons in the three directions are usually decoupled, the distribution function can be written as

$$f(x, y, z, p_x, p_y, p_z) = f_x(x, p_x)f_y(y, p_y)f_z(z, p_z). \quad (1.1)$$
The coordinates span a 6-dimensional space known as phase space. An electron bunch will occupy a certain volume inside phase space, the smaller this volume, the higher the quality of the bunch. In accelerator physics, the phase space volume is usually quantified by use of normalised root-mean-square (rms) emittances. In the \(x\)-direction it is defined as

\[
\varepsilon_{n,x} = \frac{1}{m_e c} \sqrt{\langle x^2 \rangle \langle p_x^2 \rangle - \langle xp_x \rangle^2},
\]

(1.2)

where \(m_e\) is the electron mass, \(c\) the speed of light and the brackets \(\langle .. \rangle\) indicate the average over all electrons in the bunch. The origin of the phase space axes are set to correspond to the average values of the electron coordinates, namely the center of the bunch.

The normalised rms emittances \(\varepsilon_{n,y}\) and \(\varepsilon_{n,z}\) for the \(y\)- and \(z\)-direction are defined similar to equation (1.2). The product \(\varepsilon_{n,x}\varepsilon_{n,y}\varepsilon_{n,z}\) is a measure of the volume the bunch occupies in phase space and the focusability of the bunch as will be explained next.

Consider for example an expanding bunch of electrons traveling in the \(z\)-direction as pictured in figure 1.2a. Plotting the components of their position and momentum along the \(x\)-axis results in the phase space plot shown in figure 1.2b. The area that the bunch of electrons occupies in phase space is now quantified as the root-mean-square (rms) ellipse, indicated by the red ellipse. This area is a conserved quantity during propagation of the bunch when all forces acting on the electrons are linear [12]. It can be shown that the normalised rms emittance \(\varepsilon_{n,x}\) is proportional to the area enclosed by this rms ellipse and thus is also a constant of motion.

At positions where the particles do not have a correlation between position and momentum, for instance at the source of the bunch or in a beam waist, the normalised rms emittance reduces to

\[
\varepsilon_{n,x} = \frac{1}{mc} \sigma_x \sigma_{p_x},
\]

(1.3)

where \(\sigma_x = \sqrt{\langle (x - \langle x \rangle)^2 \rangle}\) is the rms bunch size and \(\sigma_{p_x} = \sqrt{\langle (p_x - \langle p_x \rangle)^2 \rangle}\) is the rms momentum, with the axes chosen so that \(\langle x \rangle = 0\) and \(\langle p_x \rangle = 0\). An aspect of the rms momentum spread that is of importance to this work, is that it can be described at the source by an effective electron temperature \(T\). Although the velocity distribution is not a Maxwell-Boltzmann distribution, the temperature is used rather than the angular spread due to the familiarity with the concept temperature. In the transverse directions the average kinetic energy of the electrons equals
\[ \frac{\sigma_{px}^2}{2m} = \frac{\sigma_{py}^2}{2m} = \frac{1}{2} k_B T, \]  

(1.4)

where \( k_B \) is Boltzmann’s constant. The effective source temperature that is described here is used throughout this work as a figure of merit for the quality of the electron source.

In practice not the momentum spread of electrons is measured, but their angular spread. The transverse momentum \( p_x \) can be related to the angle \( \theta_x \) between an electron’s transverse velocity \( v_x \) and longitudinal velocity \( v_z \) as

\[ p_x = \gamma m_e v_x \approx \gamma \beta m_e c \frac{v_x}{v_z} = \gamma \beta m_e c \theta_x. \]  

(1.5)

In this equation \( \gamma \) is the Lorentz factor \( \left(1 - \beta^2\right)^{-\frac{1}{2}} \) and \( \beta = \frac{v}{c} \) is the relative speed of the electron. The approximation is based on the condition that \( v_z \gg v_x \) and is called the paraxial approximation. Substituting this relation in the definition of the normalised rms emittance leads to

\[ \varepsilon_{n,x} \equiv \frac{1}{m_e c} \sqrt{\langle x^2 \rangle \langle p_x^2 \rangle - \langle xp_x \rangle^2} \approx \gamma \beta \sqrt{\langle x^2 \rangle \langle \theta_x^2 \rangle - \langle x \theta_x \rangle^2} = \gamma \beta \varepsilon_x. \]  

(1.6)

In this expression the (non-normalised) rms emittance \( \varepsilon_x \) is introduced, also known as the geometrical emittance. It is proportional to the area of the rms ellipse in trace space, the space spanned by the coordinates \( x \) and \( \theta_x \). Note the approximation in equation (1.6) assumes negligible spread in longitudinal velocity, so the factor \( \gamma \beta \) is in that case constant. The distribution function \( g_x(x, \theta_x) \) of the electrons in trace space is then related to the phase space distribution function of equation (1.1) by \( g_x(x, \theta_x) = f_x \left( x, \frac{p_x}{\gamma \beta m_e c} \right) \). The example bunch of figure 1.2 has thus the exact same distribution in trace space as plotted in figure 1.2b, but with \( \theta_x \) on the vertical axis.

If there is no correlation between the position and angle of the electrons, which is true in a beam waist, equation (1.6) simplifies to

\[ \varepsilon_{n,x} \approx \gamma \beta \sigma_x \sigma_{\theta_x}, \]  

(1.7)

where \( \sigma_{\theta_x} \) is the rms angle, or angular spread of the electrons. Of course the same is true for the rms emittance \( \varepsilon_y \) in the transverse trace space as a function of position \( y \) and angle \( \theta_y \). From equation (1.7) it is clear that the minimal spot size of the beam at the waist is determined by both the focussing angle and the emittance of the beam. Since practical considerations often limit the focussing angle, it is important Using these results, the angular spread at the source can be expressed in terms of the effective source temperature as

\[ \sigma_{\theta_x} = \frac{1}{\beta \gamma} \sqrt{\frac{k_B T}{m_e c^2}}. \]  

(1.8)

This expression for the angular spread can only be used at the source and at a waist in the beam line.

When electron bunches are accelerated or decelerated in the longitudinal direction, the angles \( \theta_x \) and \( \theta_y \) change. Consequently, the geometrical emittance changes and is not a constant of motion anymore. However, the normalised rms emittance remains invariant under linear forces [12]. This means the normalised rms emittance is a useful quantity to reliably compare the quality of electron bunches from sources with different acceleration field strengths.
The aimed use of the electron bunches produced with the UCPC source is to perform diffraction experiments. The visibility of an electron diffraction pattern is limited by the momentum spread of the electrons in the bunch that is incident on the sample of interest. The more coherent a bunch of electrons is, the larger the structures are that can be measured with it. In this thesis, the transverse coherence length will be used to express the degree of coherence in the electron bunches. The transverse coherence length of a focussed electron bunch can be written as

\[ L_{c,x} = \frac{\hbar}{\sigma_{p_x}} = \frac{\hbar}{m_e c \varepsilon_{n,x}}, \tag{1.9} \]

where \( \hbar \) is the reduced planck’s constant. To be able to perform diffraction experiments on samples with lattice parameter \( a \), the transverse coherence length should be at least as large as the lattice parameter, so that for UED experiments, we require \( L_{c,x} > a \). The demands for the transverse beam quality will become stricter when samples are used with large lattice parameters, such as protein crystals.

### 1.3 Ultracold charged particle beams

At the start of this chapter, several of the important aspects and requirements of single shot UED experiments were discussed qualitatively. Now that a framework is provided in which to define beam quality, a more quantitative set of requirements can be listed for single single shot UED. The preferred kinetic energy \( U \) of the electron bunches is in the range from 100 to 300 keV [14]. If we aspire to measure protein samples using UED, we need to be able to image the large structures of the protein. A protein crystal will typically have a lattice parameters of 1 to 10 nm. For the bunches that are used to perform UED on protein samples we thus require \( L_{c,x} > 1 \) nm [15]. Moreover, to research irreversible processes that occur for example in proteins, single shot measurements should be performed, for which the requirement ideally is \( 10^4 \) to \( 10^6 \) electrons per bunch, depending on the sample [8].

At the CQT group of the Eindhoven University of Technology, a source concept is developed that has the potential to meet all requirements of single shot UED: The Ultracold Charged Particle Cloud (UCPC) source [16, 17]. The UCPC source uses rubidium atoms that are trapped and cooled in a magneto-optical trap [18]. With a two step photo-ionization process, a part of the rubidium atoms is excited and subsequently ionized. The bunches are subsequently accelerated from the magneto-optical trap using a DC operated accelerating field. In the current UCP setup, the electrons bunches can be accelerated up to energies of 15 keV, falling short of the preferred energy range.

In previous work on the UCPC source, electron source temperatures as low as 11 K were measured, with typical source sizes of 25 \( \mu \)m [19, 20]. During those experiments, photo-ionization was performed with a nanosecond pulse length ionization laser, resulting in nanosecond duration electron bunches. The emittance of the beam at the source, calculated using equations (1.7) and (1.8), was 1 mm-rad. The transverse coherence length, calculated with equation (1.9), was 20 nm for the reported emittance. The number of electrons in single bunches was reported to be typically \( 10^3 \) to \( 10^4 \) [10]. We thus see that the current UCPC setup is capable of producing electron bunches of extremely high quality that meet the coherence length requirement for UED, but that do not have enough charge to perform single shot UED, that do not have the preferred kinetic energy and that are not short enough to measure the ultrafast dynamics of atoms.

In the same work, the dependence of the transverse source temperature on the wavelength of the ionization laser and on the applied accelerating voltage were studied. In work performed by Engelen, the dependence of the transverse electron beam quality on the polarization of the ionization laser was measured using the nanosecond pulse length ionization laser [21]. Engelen moreover introduced a semi-analytical model based on electron trajectories in the combined potential energy function of the rubidium atoms and the electric field of the accelerator [22]. Simulations performed using the model match the beam quality measurements of the electron beams.
1.4 Scope of this thesis

In this work, we research the beam quality of electrons beams produced with femtosecond ionization laser pulses. The photo-ionization model developed for ionization using the femtosecond laser is included in this thesis in chapter 2. The specifics of the UCPC setup and the measurement techniques that were used to determine the beam quality are discussed in chapter 3. The measurements of the transverse beam quality and the simulations performed with the broadband photo-ionization model are discussed and compared in the first half of chapter 4. A paper on the measurements of the beam quality and on the broadband photo-ionization model has recently been submitted to Nature Communications [23]. Measurements of the dependence of the transverse beam temperature on the polarization angle of the femtosecond ionization laser are performed and reported for the first time in this work. The results of the measurements are presented and compared to simulated results in the second part of chapter 4.

The first diffraction patterns that are produced using the UCPC setup are also measured as a part of this work, and presented and discussed in this thesis. A pragmatic model on electron diffraction is presented in chapter 5, aimed at understanding the diffraction patterns that are measured. The results of the diffraction experiments are discussed in the first part of chapter 6 in the framework of the diffraction model. These experiments comprise the first attempt at diffraction using the setup, so that based on the experiments, several suggestions can be formulated to improve the quality of the images. Some of these suggestions have been worked out in the second part of chapter 6.

The last chapter of this thesis concludes the work that was performed on the transverse beam quality of ultrashort electron bunches produced with the UCPC source setup and on the first diffraction patterns that were measured using this setup. An outlook is given on the possible future experiments that can be performed to further the understanding of the UCPC source to be able to perform meaningful experiments on ultrafast dynamic systems.
In the introduction, single shot ultrafast electron diffraction was introduced as a viable way to measure ultrafast processes. An electron beam that is used as probe beam during UED experiments is required to have extreme characteristics: high charge, a long transverse coherence length and an ultrashort pulse length. The ultracold charged particle (UCP) source was introduced as a likely candidate to produce such extreme electron bunches. In this chapter, the concepts of the source and the photo-ionization model that is used to predict the performance of the source are discussed.

### 2.1 Ultracold charged particle cloud

The UCP source uses rubidium atoms that are trapped and cooled in a magneto-optical trap (MOT), using a quadrupolar magnetic field and six counter-propagating laser beams. The ultracold atoms are ionized in a two-stage process to form an UCP cloud. The electrons or ions are then accelerated from the UCP cloud towards the beam line to form a charged particle beam with small transverse velocities. The characteristic of rubidium will be discussed first in this section. Then, the different processes required for creating UCP bunches: cooling and trapping, photo-ionization and acceleration will be discussed in this section. All processes are displayed schematically in Figure 2.1.

#### 2.1.1 Rubidium

Rubidium is a common element to be used for experiments that involve trapping and cooling in a MOT [24]. To be able to trap and cool rubidium, the vacuum chamber around the MOT should be filled with rubidium vapor. Rubidium has the beneficial property that is has a low melting point at 312.5 K, only slightly above room temperature. Considerable flux can thus be achieved from a rubidium cell without having to heat it to extreme temperatures. Two isotopes of rubidium are abundant in nature, $^{85}$Rb and $^{87}$Rb. The specific isotope that is used for the UCP source described in this work is $^{85}$Rb [25]. Laser cooling takes place by using photons to excite an electron of the rubidium atoms to an excited state, after which the electron falls back to the ground state emitting a photon in a random direction. A notation will be introduced here to describe the atomic energy levels in $^{85}$Rb that are used to perform laser cooling.

An energy level of an atom can be labeled using several quantum numbers that all have meaning regarding the specific state of the electron. An energy level can be written as $n L J F$. $n$ is the principle quantum number, that represents the semi-classical orbit of an electron around the nucleus. The parameters $L$ is the angular momentum state of the electron, which can range from 0 to $n – 1$ in integer values. A common spectroscopic notation of $L$ is to express its value with a letter, for example $L = 0$ corresponds to the spherically symmetric $S$ orbital. The quantum number $J$ is the angular momentum quantum number including electron spin. The states linked to quantum number $J$ are called the fine structure of the atom. $F$ is the total angular momentum.
(a) Six counter-propagating laser beams, depicted as transparent red lines, are used to cool a cloud of rubidium atoms at their point of intersection. Two magnetic field coils, with current $I$, trap the cloud of rubidium atoms at the center of the MOT.

(b) A small portion of the rubidium atoms is excited using a laser beam travelling in the $z$-direction. After the excitation step, a laser beam travelling in the negative $y$-direction ionizes a portion of the excited atoms. The volume of intersection of the two lasers is the ionization volume.

(c) Using a parallel plate accelerator with an applied accelerating voltage $V_{\text{acc}}$, the electrons are accelerated in the $z$-direction. The neutral rubidium atoms remain at the center of the MOT.

Figure 2.1: Schematic representations of the three processes that are required to create an ultracold electron beam: cooling and trapping, photo-ionization and acceleration.

quantum number including nuclear spin. The states indicated with $F$ constitute the hyper fine structure of the atom.

In figure 2.2 a part of the energy level diagram of $^{85}\text{Rb}$ is displayed (not to scale). The energy levels and transitions that are depicted are the ones that are relevant for both the laser cooling process and for photo-ionization. We see four labeled energy levels, $5S_{\frac{1}{2}, 2}$, $5S_{\frac{1}{2}, 3}$, $5P_{\frac{3}{2}, 3}$ and $5P_{\frac{3}{2}, 4}$, and above those energy levels we see the ionization continuum, labeled as $\text{Rb}^+$. Transitions that are indicated by arrows labeled laser are pumped with laser light in the MOT. The two laser pulses that are used to photo-ionize the electrons are the excitation laser pulse, tuned to the transition from the $5S_{\frac{1}{2}, 3}$ to the $5P_{\frac{3}{2}, 4}$ level and the ionization laser pulse, that brings the electron past the ionization threshold. The process of photo-ionization is more elaborately discussed in subsection 2.1.3. In the top part of the diagram, the Stark shift $E_{\text{Stark}}$ of the ionization threshold is depicted. The Stark shift is caused by the electric field of the accelerator and will be explained in detail in subsection 2.1.4. The energy indicated as $E_{\text{exc}}$ is the excess energy, the energy that an ionized electron retains after traversing the Stark shifted ionization threshold. The excess energy is discussed in section 2.2. The cooling transition is
pumped by the trapping laser. In the diagram, we see that the trapping laser is detuned 14.5 GHz from resonance. The reason for detuning the laser and further information on the process of laser cooling can be found in subsection 2.1.2. Finally, we see that a fourth laser beam is listed in the energy level diagram, the repump laser. The necessity for a repump laser is discussed here.

Laser cooling requires the atoms to switch between the excited state and the ground state repeatedly. The laser cooling transition should be a closed system to be able to continue cooling for timescales in the order of 1 s. The typical time $\tau$ that an atom needs to relax to the ground state by spontaneous emission is $\tau = 1/\Gamma$, where $\Gamma$ is the natural linewidth of the cooling transition. For $^{85}$Rb, $\tau = 1.7 \cdot 10^{-7}$ s, so that millions of cycles can be completed within a second. In the diagram we see that photons from the trapping laser have a small chance of exciting an electron not to the $5P_{3/2, 4}$ level but to the $5P_{3/2, 3}$ level, indicated by the dashed arrow. About one in every thousand electrons is excited to the $5P_{3/2, 3}$ level. From the $5P_{3/2, 3}$ energy level, those electrons have a chance of relaxing to the $5S_{1/2, 2}$ energy level. These electrons in the $5S_{1/2, 2}$ state are removed from the cooling cycle. To re-enter electrons from the $5S_{1/2, 2}$ energy level into the cooling cycle, the repump laser beam excites electrons from the $5S_{1/2, 2}$ to the $5P_{3/2, 3}$ level. Without the repump laser beam, the population of the cooling transition disappears within several milliseconds, causing the trapped cloud to dissipate.

2.1.2 Cooling and trapping

The atoms in a MOT are laser cooled using an effect called Doppler cooling. The atoms absorb a photon from the trapping laser, getting a momentum kick $p = h/\lambda$, where $h$ is Planck’s constant and $\lambda$ is the wavelength of the photon. The absorbed photon excites the outer electron of the rubidium atom from the ground state to an excited state. The subsequent spontaneous emission caused by the electron relaxing to the ground state occurs in a random direction, so that the effect of a large amount of momentum kicks caused by the spontaneous emissions average out to zero. The net result of the absorption and emission process is thus a kick in the propagation direction of the laser. Six laser beams are directed on the atoms, from each perpendicular direction, as can be seen in Figure 2.1a.

Looking at one atom only in the $x$-direction of the problem, we see that the atom is positioned between two counterpropagating laser beams. The photon energy of the laser light is set lower than the atomic transition energy, which means that the laser is red detuned. If we now assume the atom is moving in the positive $x$-direction, then the light of the laser beam travelling in the
negative \(x\)-direction will become Doppler blue-shifted. The Doppler shift brings the photon energy closer to resonance with the cooling transition, making absorption of the photons by the atom more likely. The other laser beam propagates in the positive \(x\)-direction, same as the atom, which means that the light of that laser beam is Doppler red-shifted for the atom. The photon energy of that laser beam is thus brought further from resonance, so that the photons are less likely to be absorbed. Concluding, we see that an atom absorbs more photons from a laser beam that travels in the contrary direction of the atom. In the previous paragraph it was explained that a laser beam exerts a force on an atom by the atom absorbing photons from that laser beam. Combining these two finds, the atom experiences a force pointed contrary to its motion, and as a result the atoms are cooled. Note that this process does slow the atoms down, but does not bind them to a specific location. The lasers do not trap the atoms.

The dressed state model, a two state model of atoms [18] can be used to calculate the influence of the detuning and of the Doppler shift on the absorption rate of the atom. With these absorption rates, the resulting force of the laser beams on the atom can be calculated. The force \(F\) is proportional to the particle velocity \(v\) for small velocities, \(F = -\alpha v\), where the damping constant \(\alpha\) depends on the laser frequency detuning and the beam intensity. Another interesting result of the model for laser cooling is the minimum achievable temperature using Doppler cooling, the so-called Doppler temperature limit \(T_D\). The Doppler temperature occurs when an equilibrium is reached between the cooling and heating effects in the atom cloud. Heating in the atomic cloud is caused by the spontaneous emissions of the atoms. The heating can be modelled as a diffusive, random force that is proportional to the spontaneous emission rate \(\Gamma\) introduced in subsection 2.1.1. The Doppler temperature limit for \(^{85}\text{Rb}\) is \(T_D = 142.41\ \mu\text{K}\).

The Doppler cooling mechanism does not favor a specific position. To spatially confine, or to trap the atoms, a position dependent force has to be applied. A quadrupole magnetic field applies the restorative, position dependent force, if it is combined with circularly polarized trapping beams. The field coils that generate the magnetic field are positioned in the anti-Helmholtz configuration, as can be seen in figure 2.1a. The center of the trapping region is located where the magnetic field crosses the zero point, so that particles at that position do not experience any field. If an atom moves away from the zero field location, the atom will experience an increasing magnetic field causing a splitting of the quantum state described by quantum number \(J\) called the Zeeman splitting. To understand the influence of the splitting of the levels, we look at the simple example of a cooling transition from a \(J = 0\) to a \(J = 1\) energy level. For the excited \(J = 1\) level, three degenerate sublevels that are labeled with magnetic quantum numbers \(M = -1, 0, 1\) become distinguishable due to the magnetic field, which means that the levels can be separately excited. Quantum mechanical selection rules now dictate that \(\sigma^+\) light can only excite an electron to the \(M = 1\) state and \(\sigma^-\) polarized light can only excite to the \(M = -1\) state. The MOT design is so that the \(M = 1\) state of the atom is shifted closer to resonance when the atom is positioned outside the zero point of the magnetic field, at the side where the \(\sigma^+\) polarized laser comes from, so that the \(\sigma^+\) radiation is more likely to be absorbed and the atom is pushed back to the trapping location. Vice versa, the particle is pushed back by the \(\sigma^-\) laser to the trapping position due to the \(M = -1\) energy level moving closer to resonance. For small deviations from the trapping location, the trapping force \(F\) is proportional to the atomic displacement \(x\) as \(F = -\beta x\), where the spring constant \(\beta\) depends on the laser detuning, intensity and the magnetic field gradient. Changing the polarization of the trapping laser beams does not influence the laser cooling effect.

The combined effect of the circularly polarized trapping laser beams and the quadrupole magnetic field is a force that both damps the velocity of the atoms and restores the position of the atoms to the trapping position. For small atomic velocities and displacements, the force is linear in both the velocity of the atom and the atomic displacement with respect to the trapping location as \(F = -\alpha v - \beta x\). The atoms are trapped and cooled in the MOT.

### 2.1.3 Photo-ionization

During stable operation of the MOT, all atoms switch continuously between the ground state and the excited state. The ionization step to form a UCPC takes place by removing the outer electron.
from the excited atoms using an separate laser that is designated as the ionization laser. In figure 2.1b, we see the geometry of the situation, along with an axis system indicating the definition of the directions of the problem. The positive-z direction is the direction of propagation of the electron beam in the electric field of the accelerator. The x- and y-direction are the horizontal and the vertical direction respectively, both perpendicular to the z-axis. In the schematic representation of the photo-ionization process, the ionization laser pulses travel in the negative y-direction and the excitation laser pulses travel in the positive z-direction. The specific function of the excitation laser will be explained here.

The typical size of the ionization laser is tens of microns, while the typical size of the rubidium cloud in the MOT is 1 mm. If the MOT laser would be turned on permanently, a large fraction of all atoms in the cloud will be in the excited state. When the ionization laser is utilized to simply illuminate a part of the MOT, many electrons will be ionized on its path, resulting in electron bunches that are cylindrical in shape, with the same size as the MOT in the y-dimension. To be able to produce Gaussian shaped bunches of controlled charge and initial volume, an extra step is added to the ionization process: the excitation step. During the photo-ionization process, the trapping lasers are turned off for a small period of time, during which the excited electrons can relax to the ground state. During the time the trapping laser are turned off, the speed of the atoms is sufficiently low for the atoms to remain in the MOT region. After all atoms have relaxed to the ground state, a well-defined portion of the atoms is excited by a separate excitation laser beam. The excitation laser beam is visible in figure 2.1b as the red beam. In the schematic line diagram of $^5\text{Rb}$, we see that the excitation laser frequency is tuned to the resonant frequency of the transition from the $5S_{1/2,3}$ to the $5P_{3/2,4}$ energy level.

Following the excitation step, a part of the excited atoms is ionized using the ionization laser. The ionization energy for atoms in the excited $5P_{3/2,4}$ state corresponds to a wavelength $\lambda_0 = 479.06$ nm. In figure 2.2 the transitions excited by the excitation laser and ionization laser are depicted, where the ionization laser can be seen to excite the electrons past the ionization threshold. The wavelength of the ionization laser is tunable, so that the energy the electrons retain after traversing the ionization threshold can be tuned using the ionization laser. The shorter the wavelength of the ionization laser, the more energy the electrons finally have.

Using the excitation- and ionization lasers a UCPC is formed in the MOT. The volume of the UCPC is defined by the region of overlap between the excitation- and ionization laser beams, and is referred to as the ionization volume. The charge and the initial bunch volume are thus tunable by changing both the energy per pulse and the transverse size of the excitation- and ionization laser. Both the ionization laser and the excitation laser have a typical rms cross section of 30 $\mu$m.

The typical initial size of the electron bunches is the same as the size of the ionization volume.

The process of photo-ionization does not allow for charged particle bunches to be created with source temperatures arbitrarily close to the Doppler limit. Assuming that all electrons are ionized simultaneously, then the limiting effect for the minimal source temperature is disorder induced heating [20]. Disorder induced heating is caused by the spatial distribution of the ionized atoms being random. The Coulomb potential energy is minimal when all particles are distributes over the charged particle cloud evenly, which means that the random distribution of charged particles contains an excess of Coulomb potential energy. This excess potential energy is released by the spatial electron distribution shifting until the kinetic energy is in equilibrium with the Coulomb energy throughout the charged particle cloud, effectively heating the cloud. The minimum achievable temperature associated with disorder induced heating $T_{\text{dih}}$ depends on the density of the electrons in the UCP cloud $n_e$ as $T_{\text{dih}} \sim n_e^{1/3}$. For typical numbers of the electron density, $n_e = 10^{18}$ m$^{-3}$, the minimum temperature is $T_{\text{dih}} = 9$ K. The collective charged particle motion in the UCP cloud takes place at timescales expressed as the inverse plasma frequency in the UCP cloud,

$$\frac{1}{\omega_{\text{plasma}}} = \sqrt{\frac{m_e e_0}{n_e q_e^2}},$$  \hspace{1cm} (2.1)$$

where $e_0$ is the permittivity of free space. For an electron density $n_e = 10^{18}$, the equilibrium will be achieved typically in 25 ps.
2.1.4 Acceleration

Following the formation of the UCP cloud either the electron bunches (or ion bunches) are accelerated in the positive $z$-direction using a DC operated acceleration structure. The acceleration step is schematically shown in Figure 2.1c, where the accelerator is visualized by two parallel plates. The atoms are accelerated to a longitudinal mean energy $U$, which is determined by the applied electric field $F$ and the distance from the center of the MOT to the end of the accelerator $d_{\text{acc}}$ as $U = q_e F d_{\text{acc}}$. The rms longitudinal energy spread $\sigma_U$ is dominantly determined by the longitudinal size of the ionization volume $\sigma_z$, since particles ionized in the front of the accelerator have a lower final energy than particles ionized further back, which results in a longitudinal energy spread of $\sigma_U = q_e F \sigma_z$. With a typical rms size of the ionization volume of 20 $\mu$m and an accelerating distance of 1.35 cm, the energy spread relative to the mean energy is typically 0.2 %.

In the rest of this work, the magnitude of the electric field in the accelerating structure will be given in terms of the accelerating voltage $V_{\text{acc}}$. The electric field inside the accelerator is proportional to the accelerating voltage as $F = V_{\text{acc}}/d_{\text{acc}}$. For a typical accelerating voltage of 6 kV, the magnitude of the electric field is 0.44 MV/m.

2.2 Photo-ionization model

As explained in the introduction, the important characteristic of the beams that are produced using the UCPC setup is the low angular spread of the source, or differently said, the low transverse source temperature. Precisely how low the transverse temperature can become can be determined semi-analytically using a classical model for the electron trajectories in the combined potential of the accelerating field and the potential of a rubidium atom. In the first part of this section, the excess energy will be introduced and explained. In the second part of this section, a narrow bandwidth trajectory model will be introduced to determine the angular spread of the source. In the last part of this section, the narrow bandwidth model will be generalized to be able to calculated the angular spread of the source for large ionization laser bandwidths.

2.2.1 Excess energy

The excess energy is defined as the kinetic energy that an electron retains after leaving the MOT and the accelerator. The excess energy depends on the zero-field energy of the photon that was used to ionize the atom and on the voltage applied to the accelerator. The dependence of the excess energy on the ionizing photon energy is trivial, since the energy of the photon that is in excess of the zero-field ionization threshold $E_{\lambda}$ is transferred almost completely to the electron of the ionized atom, with a negligible part of the energy going to the atom. The energy of a photon can be expressed in terms of the wavelength of the photon as $E_{\lambda} = h c / \lambda$. The dependence of the excess energy on the accelerating voltage requires more explanation. The electric field applied in the accelerator lowers the ionization threshold of the atoms by an amount referred to as the Stark shift. The Stark shift $E_{\text{Stark}}$ can be understood by imagining potential energy of an electron in the combined electric field of a rubidium atom and an externally applied homogeneous electric field, that can be described as a slanted infinite well shape. This shape is depicted in figure 2.3a in the $x$-$z$ plane, with distances expressed in atomic units (the atomic unit for length is the Bohr radius $a_0$). The Stark shift is visible as the height difference between the saddle point in the potential landscape that the electrons traverse and the zero level that corresponds to the zero-field ionization threshold. The starting angle $\beta$ can be defined using figure 2.3a as the initial angle of the electron trajectory with respect to the $z$-axis. The azimuthal angle of the trajectory with respect to the $x$-axis in the $x$-$y$ plane has no impact on the trajectories, because the situation is cylindrically symmetric.

The excess energy can be expressed in experimental parameters and physical constants as
(a) Surface plot of the potential energy \( U \) as a function of the \( x \)- and \( y \)-coordinates, with distances expressed in atomic units (the atomic unit of length is the Bohr radius \( a_0 \)). Visible are both the Stark shift \( E_{\text{Stark}} \) and two examples of electrons escaping with a starting angle \( \beta \).

(b) Contour plot of the potential energy of the electron offset with the photon excess energy \( E_\lambda \). Several possible escape trajectories for different starting angles \( \beta \) up to the classical maximum \( \beta_{\text{max}} \) are shown for particles with zero-field ionization energy \( E_\lambda \).

Figure 2.3: Graphic representations of the potential energy of an electron in the combined electric field of the accelerator and the Rb\(^+\) atom.

\[
E_{\text{exc}} = E_\lambda + E_{\text{Stark}} = h c \left( \frac{1}{\lambda_{\text{ion}}} - \frac{1}{\lambda_0} \right) + 4 R_y \sqrt{\frac{V_{\text{acc}}}{d_{\text{acc}} F_0}},
\]

(2.2)

where \( \lambda_{\text{ion}} \) is the ionization laser wavelength, \( R_y \) is the Rydberg constant and \( F_0 \) is the atomic unit of electric field strength.

Although the accelerating voltage and the ionization laser wavelength can both be used to tune the excess energy of the charged particle bunches, they are not interchangeable. To understand this, we consider the potential energy plot in figure 2.3a. If the ionization laser wavelength is changed, the energy of the electrons in the potential landscape will change, which causes an offset in the potential energy, but the shape of the surface will remain unchanged. A change in the accelerating voltage however causes the potential landscape to change all together. Although the
(a) The transverse velocity of an electron versus the starting angle $\beta$ of the electron with respect to the $z$-axis, plotted for $E_{\text{exc}} = 5, 15$ and $25$ meV, for $V_{\text{acc}} = 2$ kV, for the case of a hydrogen ion potential. The electrons escape the hydrogen potential for starting angles $\beta$ up to $\beta_{\text{max}}$.

(b) The transverse velocity of an electron versus the starting angle $\beta$ of the electron with respect to the $z$-axis, plotted for $E_{\text{exc}} = 5$ meV and for $V_{\text{acc}} = 2$ kV, for the case of a rubidium ion potential. The electron can escape the potential well of the ion for all starting angles.

Figure 2.4: Simulated transverse velocities of an electron as a function of the starting angle $\beta$ [22].

excess energy is a convenient parameter for intuitive understanding of the ionization process, it is not a unifying parameter for the ionization laser wavelength and the accelerating voltage. The excess energy will therefore not be used without mention of either the accelerating voltage or the wavelength of the ionization laser.

2.2.2 narrow bandwidth model

In this subsection a model is presented for the quality of electron bunches that are produced with photo-ionization of atoms trapped in a MOT. The parameter that is used to represent the beam quality is the transverse source temperature of the electron bunches, calculated from the angular spread with equation (1.8). The model presented here assumes that the electrons all have the same excess energy. Assuming that the Stark shift is equal for all atoms, then the assumption of equal excess energies implies that all atoms are ionized by photons with the same wavelength, which means that the ionization laser pulses are assumed to be monochromatic. The electrons source is modelled as a single point emitter, which means that all electrons start from the same point in space. This model will be referred to as the narrow bandwidth photo-ionization model. A very narrow bandwidth laser with pulses of nanosecond duration is present in the laboratory setup.

The motion of electrons in the combined potential of a homogeneous electric field and a positively charged ion has been researched by Bordas et al. [26, 27]. Bordas found that the equations of motion of the electrons can be expressed analytically as a function of the parameters that describe the potential energy of the electron, so that a precise trajectory can be calculated for every electron. Bordas assumed the $1/r$ hydrogen potential for the positively charged ion, which can be seen as a simple model for a singly ionized rubidium ion. The radial velocity $v_r$ of the electron trajectories for $z \to \infty$ follows directly from the Bordas model. The radial velocity depends on the excess photon energy of the electron $E_\lambda$, on the Stark energy $E_{\text{Stark}}$ and on the starting angle $\beta$: $v_r = v_r (E_\lambda, E_{\text{Stark}}, \beta)$. The radial velocities calculated using the Bordas model form the basis for all simulations in this thesis. The Bordas model was adapted to the conditions of the UCP source setup by Engelen [22].

Several of the trajectories calculated for electrons using the Bordas model have been displayed in figure 2.3b. Under the trajectories, a contour plot of the potential energy of the electrons is shown. The potential energy of the electrons is offset with the zero-field excess photon energy $E_\lambda$, which means that classically, the electrons are not allowed to cross the zero equipotential line,
displayed as the thicker line in the image. We see that the electron trajectories vary strongly with the starting angle $\beta$, and that trajectories are shown for starting angles up to a maximum starting angle $\beta_{\text{max}}$. A simple expression for the maximum escape angle follows from the Bordas model, namely

$$\beta_{\text{max}} = 2 \arccos \left( \frac{|E_\lambda|}{E_{\text{Stark}}} \right).$$

(2.3)

From equation (2.3), we see that $\beta_{\text{max}} \to 0$ for $E_\lambda \to 0$ and that $\beta_{\text{max}} \to 180^\circ$ for $E_\lambda \to \infty$. Electrons that are emitted from the ion with a starting angle $\beta > \beta_{\text{max}}$ cannot escape the potential, which is caused by the perfect $1/r$ potential of hydrogen, where $r$ is the radius in a spherical coordinate system. The $1/r$ dependence of the hydrogen potential allows for closed electron orbits within the potential well of the ion. Electrons that start with $\beta > \beta_{\text{max}}$ will thus remain trapped in the potential well of the ion.

The transverse velocity calculated with the hydrogen model is plotted versus the starting angle $\beta$ for three different excess energies, $E_{\text{exc}} = 5$, 15 and 25 meV, in figure 2.4a for $V_{\text{acc}} = 2$ kV. What we see from the electron trajectories is that the transverse velocity increases with the escape angle, until the starting angle is so large that the electrons bounce off the potential wall and come back towards the $z$-axis. In figure 2.3b, we see that the trajectory with the largest starting angle crosses the $z$-axis twice. The transverse velocity goes to zero right when the trajectory becomes parallel to the $z$-axis and increases again for further increasing $\beta$, which is caused by the trajectories going back towards the $z$-axis. For high excess energies, we see in figure 2.4a that the electrons can pass the $z$-axis multiple times, giving rise to multiple zeros in the transverse velocity curve.

The model that was described here uses the $\text{H}^+$ ion as a model for the $\text{^{85}Rb}^+$ ion. The rubidium ion is however different from the hydrogen ion. This difference is caused by the 36 electrons that the singly ionized rubidium atom has left, shielding the charge of the nucleus. In the far field potential of the ion, this difference is negligible. In the near field potential however, the influence of the bound electrons of the rubidium ion is very noticeable. Simulations of the case of a realistic rubidium potential have been conducted by Engelen using the General Particle Tracer code [28] to research the differences between the analytical hydrogen model and the more realistic rubidium model. For the case of an electron escaping the rubidium potential with $E_{\text{exc}} = 5$ meV and $V_{\text{acc}} = 2$ kV, the transverse velocity is plotted in figure 2.4b. This image can be compared with the $E_{\text{exc}} = 5$ meV line from figure 2.4a. What we see, is that for $\beta < \beta_{\text{max}}$, the curves are close to identical. For the rubidium atom however, the electrons are able to escape the potential well for all starting angles, which means that the rubidium potential does not allow for closed orbits of the electrons, because of the bound electrons of the rubidium atom. The electrons with $\beta > \beta_{\text{max}}$ can escape after they make several recursions around the ionic core. After those recursions, the electrons come close enough to the rubidium ion to be scattered by the ionic core are able to leave the potential well.

The realistic rubidium model shows us that the charge of individual electron bunches should not depend on the maximum escape angle that was calculated using the hydrogen model, because all electrons can escape, independent of their starting angle. Since all electrons have the same energy after photo-ionization in the narrow-bandwidth model, the total charge of the electron bunches should not depend on the ionization laser wavelength all together, except for the case of wavelengths that are so long that the laser is not capable of ionizing the rubidium atoms.

Calculations of the transverse source temperature with the realistic rubidium potential using GPT and with the hydrogen model using the Bordas model were compared to see the influence of the bound electrons of the rubidium ion on the quality of the electron bunches. It was found that the temperature calculated with the Bordas model matches the result of the GPT simulations. The difference is so small, that it was decided that the less laborious hydrogen model would be used to perform all temperature simulations that are presented in this work.

The transverse source temperature increases when the transverse velocity of the electrons increases. Two effects that increase the transverse velocity for increased excess energies are visible in figure 2.4a. In the low excess energy regime, an increase of the excess energy causes an increase
of the maximal escape angle $\beta_{\text{max}}$. The electrons that escape for those increased escape angles have larger transverse velocities relative to the smaller escape angles. This effect stops to play a role when $\beta_{\text{max}} \rightarrow 180^\circ$. For all excess energies, the transverse velocities for starting angles below $\beta_{\text{max}}$ increase for higher excess energies. These two effects both contribute to an increased transverse source temperature for higher excess energies.

To determine the chance $P(\beta)$ for an atom to emit an electron with starting angle $\beta$, we need the polarization direction of the ionization laser. Assuming the polarization of the ionization laser is in the $z$-direction, then the chance that an electron is emitted with starting angle $\beta$ is

$$P(\beta) = \frac{1}{A_{\beta}} \cos^2(\beta),$$

where $A_{\beta}$ is a normalization constant. With the polarization in the $z$-direction, there is a maximum chance of an electron being emitted in the $\pm z$-direction and there is no chance of an electron being emitted in the $x$-direction. The model assumes that the distribution of the electrons along the starting angle matches the $P(\beta)$ distribution, so that the electrons with starting angles of $0^\circ$ and $180^\circ$ are preferred, and no electrons are emitted with a starting angle of $90^\circ$. This assumption is valid in the case that the number of electrons is very large, so that deviations from the expected behavior are averaged out. In the rest of this subsection, the polarization of the ionization laser is assumed to be in the $z$-direction, so that indeed $P(\beta) = \frac{1}{A_{\beta}} \cos^2(\beta)$.

The model implementation written by Engelen assumes a finite number of electrons. The starting angles of the electrons are distributed as $P(\beta)$ and $P(\beta)$ is determined by the polarization angle of the ionization laser. For now, we assume that the polarization of the laser is in the $z$-direction. With the radial velocity $v_r\, (E, E_{\text{Stark}}, \beta)$, the spread in radial velocities for $N$ electrons is

$$\sigma_{vr} = \sum_{i=1}^{N} \frac{v_r(E, E_{\text{Stark}}, \beta_i)^2 P(\beta_i) \sin(\beta_i)}{2P(\beta_i) \sin(\beta_i)}.$$  

(2.4)

When the model is used for a situation with a single excess energy, the angular spread is calculated for electrons ionized by a very narrow bandwidth laser. The transverse source temperatures calculated from the model, assuming a narrow bandwidth ionization laser is displayed in figure 2.5 plotted versus the ionization laser wavelength that determines the excess energy. Note that on the $x$-axis, the wavelength is displayed in reverse. This choice is made so that the highest energy particles are in the right most part of the graphs. We see that for each accelerating voltage, the curve for the transverse source temperature starts at a specific wavelength, the critical wavelength $\lambda_c$, $\lambda_c$ is the wavelength for which the excess energy is exactly zero, and depends only on the accelerating voltage. Wavelengths lower than $\lambda_c$ are not able to excite electrons past the ionization threshold in the classical model. The source temperature of the electrons is zero for the case of a wavelength exactly at the $E_{\text{exc}} = 0$ benchmark. The temperature then quickly increases after which it forms a small plateau. This plateau becomes increasingly long for higher accelerating voltages. Beyond the plateau, the behavior for all curves becomes the same. The unified behavior of the curves follows the classical relation $T = \frac{2}{3\hbar} E_\lambda$ for $E_{\text{exc}} \rightarrow \infty$.

In the previous section, a minimal source temperature was introduced for the UCPC electron source based on disorder induced heating. The minimal temperature associated with disorder induced heating is 9 K. The source temperatures that the model predicts become lower than this minimal temperatures for wavelengths close to $\lambda_c$. The model predicts these temperatures since the disorder induced heating effect is not included in the model. Temperatures lower than the limit imposed by disorder induced heating that are predicted by the model should be disregarded.

The behavior of the temperature curve is understandable from the combined potential of the accelerating field and the rubidium ion. For low excess energies, the electrons are surrounded by an insurmountable potential well, with only a small saddle point where they can escape. The width of that saddle point is determined mainly by the accelerating voltage, which determines the shape of the potential. The width of the saddle point then determines the maximum escape angle and that is the dominant contribution to the electron source temperature. For low excess energies, the accelerating voltage is thus the dominant factor in determining the source temperature.
For high excess energies, the landscape that the electrons see is quite different. They see an easily escapable well, with behind that a straight insurmountable slanted wall. The shape of that wall hardly varies for different accelerating voltages and all electrons are easily capable of escaping, so there is no maximal escape angle to influence the temperature. The temperature is thus dominantly determined by the wavelength of the ionization laser. This explains the unified behavior of the curves for high wavelengths, where the temperature is only determined by the energy provided by the ionization laser.

### 2.2.3 broadband model

As stated in section 1.4, one of the major aims of this thesis is to measure and understand the effect of a large spectral bandwidth of the ionization laser on the behavior of the electrons. With a broadband ionization laser with a mean wavelength $\mu_\lambda$ and a width of $\sigma_\lambda$, electrons are released into the potential landscape with a whole range of excess energies. Within the whole ensemble of ionized electrons, fractions of electrons can be identified that have approximately the same excess energies. The source temperature of each of these fraction can be calculated using the narrow bandwidth model. The number of electrons in each fraction depends on the relative laser intensity for that specific wavelength within the spectrum, the excess energy distribution of electrons will thus be Gaussian. This generalization can mathematically be described by the convolution integral of the narrow bandwidth temperature curve with the spectral envelope of the broadband laser with boundaries $0 < E_{exc} < \infty$. Strictly speaking, this is not a convolution since that is defined as an integral from $-\infty$ to $\infty$. For mean wavelengths $\mu_\lambda < \lambda_c + \sigma_\lambda$ however, a significant part of the wavelengths in the ionization laser spectrum are below $\lambda_c$. That part of the spectrum is not capable of ionizing electrons and should therefore not be included in the model, hence the $E_{exc} = 0$ lower boundary for the integration.

The spectral envelope of the ionization laser, the narrow band temperature curve, and the broadband temperature curve are displayed in figure 2.6a. The width of the ionization laser is assumed to be $\sigma_\lambda = 4$ nm, a typical value for femtosecond laser systems. We see that the broadband laser is capable of producing electron bunches in wavelength regimes where the narrow band laser is not. This can be explained by keeping in mind that the wavelengths displayed on the $x$-axis in figure 2.6a are the mean wavelengths of the ionization laser spectrum. The long flanks of the spectrum still contain many wavelengths capable of ionizing electrons. Moreover, we see that
the broad bandwidth laser is capable of producing electron bunches with a low angular spread, despite containing many lower wavelengths that create high excess energy electrons. The key to this low temperature is the narrow saddle point that the electrons pass.

When comparing two laser pulses that contain equal energy, but where one has \( \mu_\lambda < \lambda_c + \sigma_\lambda \), and where the other has \( \mu_\lambda > \lambda_c + \sigma_\lambda \), then part of the second laser’s spectrum are wavelengths incapable of ionizing electrons. The second laser pulse will therefore result in electron bunches with lower charge than the first laser pulse. The charge of the electron bunches will therefore depend on the wavelength, assuming equal laser pulse energies. The dependence of the charge on the wavelength can be described as a finite boundary integration of the Gaussian spectral envelope of the ionization laser. This integration is defined as the error function \( \text{erf}(x) \), a function that saturates to \(-1\) for \( x \to -\infty \) and to 1 for \( x \to \infty \). The charge can be expressed as a function of the wavelength as

\[
Q(\lambda) = Q_0 + Q_1 \text{erf} \left( \frac{\lambda - \lambda_c}{\sqrt{2} \sigma_\lambda} \right).
\]

Both the position and the width of the charge curve should follow from the characteristics of the ionization process, namely from the critical wavelength \( \lambda_c \) and the width of the spectral envelope \( \sigma_\lambda \). The charge signal will saturate to zero for \( \lambda \to \infty \), and the saturation value for very high excess energies will depend on many parameters such as the MOT density and the ionization laser pulse energy. The saturation effect for \( \lambda \to \infty \) places a constraint on the parameters \( Q_0 \) and \( Q_1 \), namely \( Q_0 - Q_1 = 0 \). With the constraint that the initial charge of the bunches is zero, this model has one free parameter, the charge for \( \lambda \to 0 \).

In figure 2.6, the transverse source temperature is displayed as a function of the mean ionization laser wavelength for several different accelerating voltages for the case of a broadband ionization laser. In the high excess energy regime, the behavior for the temperature is similar to the behavior of the narrow bandwidth model curve, following the \( T = \frac{2}{3E} E_\lambda \) line. For low excess energies, the behavior of the temperature curve flattens out forming a plateau, but unlike the narrow bandwidth model, the plateau goes on for negative mean excess energies. The value for the temperature was calculated for mean excess energies down to \(-20\) meV.

The fact that the electrons only escape with limited angles raises a question of how correct it is
to use the term temperature to describe the angular spread of the source. The term temperature suggests that the process is isotropic. The strong forward directionality of the electron trajectories however is quite clearly not isotropic. Temperature must thus be seen in a different light when used in this thesis. Temperature gives a handhold for comparing the UCPC source with different, isotropic electron sources. Moreover, the very fact that a feeling is already associated with the word temperature enables the user to quickly develop a feeling for the beam dynamics that are associated with a certain beam temperature. For these reasons, we will continue using the term temperature throughout this thesis, purely as a measure for the angular spread of the beam.

2.2.4 Laser polarization

Earlier, the laser polarization was briefly mentioned in the description for the source temperature determination. The polarization is used to determine how many electrons are ionized at each starting angle. The results that were presented were calculated for the ionization laser polarization in the z-direction. Using a quarter wave plate, the polarization of the ionization laser can be rotated. The angle of the linear polarization can be varied in the x–z-plane. We will describe the effect of changing the laser polarization on the source temperature in the upcoming subsection.

The polarization angle \( \phi \) is defined as the angle between the

With the polarization in the z-direction, the distribution function for the electrons as a function of the starting angle is \( \cos^2(\beta) \), favoring starting angles 0° and 180°, and with no emitted electrons for a starting angle of 90°. The angle of the polarization is defined with respect to the positive z-axis, the direction of propagation for the electrons. A rotation of the polarization results in a phase shift in the electron distribution function. If the polarization of the ionization laser is for example in the x-direction, then the electron distribution function turns into \( -\cos^2(\beta) \), so that no electrons are emitted for starting angles 0° and 180° and that the preferred starting angle is 90°.

Looking at figure 2.4a, we see that the change in the distribution function influences the transverse velocities of the particles. The result of this effect is that the transverse source temperature will vary sinusoidally with the laser polarization direction.

Aside from the model, a source temperature difference for different polarization directions of the ionization laser can also be explained conceptually. As discussed in the previous section on photo-ionization, the ionization laser induces disorder in the electron beam during the ionization step. This disorder will of course be more evident in the polarization direction of the laser. Assuming the laser is polarized in the z-direction, the disorder will also be induced mainly in the z-direction, with is the longitudinal dimension for the electron beam. The laser induced disorder will therefore show up less in the transverse source temperature. Assuming the laser polarization is in the x-direction, the laser induced disorder will predominantly be in the transverse dimensions, causing a higher transverse source temperature.

The results of the polarization model are displayed in figure 2.7 for three different wavelengths, calculated for \( V_{\text{maxacc}} = 6 \) kV. The predicted sinusoidal behavior of the transverse source temperature as a function of the ionization laser polarization is clearly visible. Both the amplitude of the oscillations and the mean of the oscillations increase with the ionization laser wavelength. We see that the temperature is maximal for the polarization in the x-direction and is minimal for the polarization in the z-direction. The polarization direction of the ionization laser is indicated with arrows in the figure.

In figure 2.8a, the mean temperature of the oscillation is displayed as a function of the ionization laser wavelength. The behavior of the mean temperature resembles the behavior of the transverse source temperature curves displayed in figure 2.6, as is expected. The main difference is that the mean temperature curve is always higher than the source temperature curve of figure 2.6. Since figure 2.6 is plotted for a z-polarized ionization laser, it should indeed be lower.

The amplitude of the source temperature oscillations also increases monotonicly, as seen in figure 2.8b. The shape of the line also strongly resembles the earlier displayed temperature curves. The absolute value of the oscillations is about a factor 4 smaller than the mean of the oscillations. Finally, the charge of the electron bunches is not expected to depend on the polarization of the
Figure 2.7: Modelled transverse source temperature as a function of the polarization angle of the ionization laser with the positive z-axis, for three different wavelengths of the ionization laser, calculated using the broadband hydrogen model for $V_{\text{milleracc}} = 6$ kV. The arrows labeled $\hat{x}$ and $\hat{z}$ indicate a laser polarization in the $x$-direction and in the $z$-direction respectively.

Figure 2.8: Model curves of the characteristics of the transverse source temperature oscillations, calculated using the broadband hydrogen model for $V_{\text{milleracc}} = 6$ kV.

ionization laser. For a given distribution of excess energies, all ionized electrons escape from the rubidium potential, independent of the laser polarization.

The analytical broadband point source model can be used to make predictions of the source temperature of the UCPC electron source. The predictions were described and displayed in this chapter. In the upcoming chapter, the specifics of the UCPC setup and the measurement techniques that are employed to conduct the actual source temperature measurements are discussed. The results of the measurements are discussed and compared to the model results in chapter 4.
Chapter 3

Setup and measurement techniques

In the previous chapters, the process of magneto-optical trapping, photo-ionization and acceleration was introduced as a viable source of high coherence electron bunches. In this chapter, the experimental necessities and techniques are introduced that are required to create charged particle bunches in the laboratory. A model is introduced that enables the calculation of the size of the electron beams throughout the setup. The model, together with the experiments give the experimenter the tools needed to measure the beam quality of the UCPC charged particle source.

3.1 UCPC setup

For both the trapping and the repump laser applications, commercially available 780 nm diode laser systems are used, as described in table 3.1. The fine tuning of the wavelengths of both laser beams is performed using computer controlled acousto-optical modulators. The polarization of the lasers is controlled by first ensuring linear polarization with a polarizing beam splitter cube and subsequently making the polarization circular using a quarter wave plate.

The MOT coils that are used to create the magnetic trapping field in the experiment are fabricated of hollow copper tube, with windings of a diameter of 100 mm. The two coils are spaced 100 mm from each other. The coils are water cooled so that operation with a steady current of 175 A is possible [29]. The typical gradient produced by the coils is in the order of 10 - 20 G/cm. A second set of coils, the MOT correction coils, are present in the setup to slightly tweak the position of the zero field crossing, so that the position of the atom cloud in the MOT can be changed.

The light of the excitation laser beam is tuned exactly to the resonant frequency of the transition from the $5S_{1/2,3}$ level to the $5P_{3/2,4}$ level. The excitation laser light originates from the same laser as the trapping laser light and is tuned to the correct transition using a separate AOM. The characteristics of the laser that is used to both cool and to excite the atoms can be seen in table 3.1.

<table>
<thead>
<tr>
<th>Laser</th>
<th>use</th>
<th>Wavelength</th>
<th>Power/Energy</th>
<th>Pulse length</th>
</tr>
</thead>
<tbody>
<tr>
<td>Toptica DLX 110</td>
<td>trap, excite</td>
<td>780 nm</td>
<td>900 mW</td>
<td>CW</td>
</tr>
<tr>
<td>Toptica DL 100</td>
<td>repump</td>
<td>780 nm</td>
<td>100 mW</td>
<td>CW</td>
</tr>
<tr>
<td>Quanta-Ray PDL3</td>
<td>ionize</td>
<td>470 – 490 nm</td>
<td>100 µJ</td>
<td>2.5 ns rms</td>
</tr>
<tr>
<td>Coherent Mantis, -Evolution,</td>
<td>ionize</td>
<td>472 – 533 nm</td>
<td>50 µJ</td>
<td>110 fs</td>
</tr>
</tbody>
</table>

Table 3.1: List of the lasers used in the UCPC experiments [29]
Two different lasers were used to ionize the atoms in the experiments presented in this work, both listen in Table 3.1, one with a pulse length of several nanoseconds by Quanta-Ray, one with a pulse length of several hundred femtoseconds by Coherent. A half wave plate was placed into the setup to be able to rotate the polarization of the ionization laser.

The wavelength of the nanosecond laser can be tuned to great precision, using a dye system together with a high precision spectrometer as feedback. The femtosecond laser however is a Ti:Saph system, which has an output of strictly 800 nm laser light. To be able to modulate the wavelength, a separate device, an Optical Parametric Amplifier (OPA) is used, marketed by Coherent to work with the Legend laser system. The OPA is capable of modulating the mean wavelength from 472 nm, up to several µm [30].

The calibration of the OPA is dependent on the settings of several stepper motors in the device. The output of the OPA however also depends on the input beam characteristics, e.g. compression and alignment. To be able to correctly interpret experimental results, the OPA spectral output must therefore be measured each time experiments are conducted, both the mean and the spread. To this end, an Ocean Optics spectrometer is integrated into the setup. An example of a calibration of the OPA output can be seen in figure 3.1.

After the ionization step an electron bunch or an ion bunch is accelerated towards the further beam line using a DC operated acceleration structure. The exit from the accelerating structure defocusses the beam, an effect known as the exit kick. Negative voltages to accelerate electrons can be applied up to $V_{\text{acc}} = -30$ kV, positive voltages can be applied up to $V_{\text{acc}} = 30$ kV. A more detailed schematic of the cross section of the accelerating structure is displayed in Figure 3.2. The outer vacuum chamber is visible with the MOT coils, excitation laser beam path and the rectangular accelerating structure.

The vacuum inside the vacuum chamber and the further beam pipe is achieved using a turbo pump and two ion getter pumps. The turbo pump is used to pump down from atmospheric conditions, to pressures that are low enough for the ion getter pumps to be effective. The ion getter pumps are not suitable to pump down the setup from atmospheric conditions. The main advantage of ion getter pumps is operation without the use of rotating parts, ensuring vibration free operation. One ion getter pump is connected to the vacuum chamber, one ion getter pump is installed at the end of the beam line, the turbo pump is also connected to the beam pipe.

Upon leaving the magneto optical trap, the electrons experience several passive effects. First, at 1 cm after the MOT, the electron beam is defocussed by the accelerator kick that has a focal...
point $f_{ak} = -0.033$ m [19]. Secondly, at $z = 2.4$ cm, the electron beam is focussed by the magnetic field of the MOT coils. The focal point of the MOT coils lies at $f = 0.78 \cdot U \ [m/keV]$, where $U$ is the mean energy of the electrons. A quadrupole inducing component is present in the setup, that influences the beam dynamics. The quadrupole effect is attributed to a mirror that is mounted inside the MOT chamber, that might be magnetic. The beam is focussed in one of the transverse dimensions, and defocussed in the other. Waist scans therefor have unequal beam sizes for the two transverse dimensions. The focal distance of the quadrupole component can be determined using [20]

$$f_\pm = \pm \frac{\sqrt{2m_eU}}{q_e} \frac{1}{kl}$$

(3.1)

where $m_e$ is the electron mass, $q_e$ is the elementary charge and $kl$ is a measure for the strength of the quadrupole.

The earth magnetic field causes the electrons to gyrate, so that they follow a helical trajectory. The radius of the helix trajectory is in the order of meters for typical accelerating voltages. To be able to transport the electron beam along the beam line that has an inner diameter of 40 mm, the effect of the earth magnetic field must be cancelled. An additional magnetic field is applied along the entire beam line to enable the electrons to follow a straight trajectory. The magnetic field is created using two wires that are positions along the length of the beam line and that carry a current of 5 A.

After the passive effects, the electrons bunches can be manipulated using magnetic electron optics while traversing the beam line. The electrons first pass through a steering coil that can be used to change the beam pointing in both transverse directions. Following the steering coil, the beam arrives at the magnetic lens that is used to perform the waist scans that were described in the previous chapter. The magnetic solenoid lens that is installed in the setup has $N_w = 200$ windings, an inner radius $\rho_{in} = 5.8$ cm, outer radius $\rho_{out} = 6.2$ cm and a length of $L = 5.0$ cm. The magnetic field of a solenoid lens on the optical axis of the lens can be approximated as [31]
\[ B_z(0, z) = \frac{\mu_0 N_w I_s}{(\rho_{\text{in}} - \rho_{\text{out}})} L \left( \frac{L}{2} + z \right) \ln \left[ \frac{\rho_{\text{out}} + \sqrt{\rho_{\text{out}}^2 + \left( \frac{L}{2} + z \right)^2}}{\rho_{\text{in}} + \sqrt{\rho_{\text{in}}^2 + \left( \frac{L}{2} + z \right)^2}} \right] + \ldots \]

\[ \ldots \frac{\mu_0 N_w I_s}{(\rho_{\text{in}} - \rho_{\text{out}})} L \left( \frac{L}{2} - z \right) \ln \left[ \frac{\rho_{\text{out}} + \sqrt{\rho_{\text{out}}^2 + \left( \frac{L}{2} - z \right)^2}}{\rho_{\text{in}} + \sqrt{\rho_{\text{in}}^2 + \left( \frac{L}{2} - z \right)^2}} \right], \quad (3.2) \]

where \( \mu_0 \) is the permeability of free space and \( I_s \) is the current. The maximum current that the solenoid can withstand is 5 A, which is enough to be able to over-focus beams with the maximal energy of 15 keV, so that all beam geometries can be formed using the lens. The focal point of the magnetic lens is calculated using the equation for the focal point of a solenoid lens

\[ f_{\text{sol}} = \frac{4 \gamma^2 m_e^2 v_z^2}{q_e^2} \int_{-\infty}^{\infty} \frac{1}{B_z^2} \, dz, \quad (3.3) \]

where \( \gamma \) is the Lorentz factor, and \( v_z \) is the mean velocity of the electrons. Since the magnetic field of the solenoid lens increases with the current, we see that the focal point of the lens decreases by increasing the current. When the velocity of the electrons is increased, the focal point of the solenoid lens will increase. The current that is needed to focus high energy electrons is thus higher than the current required to focus lower energy electrons. To be able to align the solenoid lens, it is mounted on a tip-tilt stage, which in turn is mounted on an \( x,y \) translation stage.

Following the solenoid lens, a second steering coils is installed in the setup to be able to direct the electron bunches towards the detector. Right before the second steering coil, a rotation point has been designed to the beam line, so that the alignment of the beam line itself can be changed slightly. This rotation point can be used to fine tune the alignment of the detector with respect to the particles traversing the beam line. Since the steering coils are capable of steering the electron bunches, the position of the detector is kept constant during experiments.

A charge detector, a so called Faraday cup has been installed in the setup after the second steering coil. The Faraday cup consists of a conducting plate connected to a charge amplifier whose signal is measured using an oscilloscope. The conducting plate that is placed in the beam line is several tens of millimeters across, which is large enough for all charge to arrive on the Faraday cup. The Faraday cup is calibrated using a well known electric signal, the calibration constant is \( 5.92 \, \text{fC/V}_{\text{scope}} \). The Faraday cup is mounted on a translation stage, to be able to remove the Faraday cup from the beam line, to enable the electrons to pass by and reach the alternative detector setup.

The size of the beam can be measured using two stacked multi channel plates (MCPs), a phosphor screen, a lens and a CCD camera. A schematic representation of the detector setup is visible in figure 3.3, along with an event cascade that is caused by a single electron. The MCPs amplify the signal of the single electron by emitting a cascade of secondary electrons. The cascaded electrons are accelerated towards the phosphor screen by increasing voltages applied over the two MCPs and the phosphor screen. The potential of the second MCP is set to 1 kV with respect to ground, the potential of the phosphor screen is \( 3.2 \, \text{kV} \). The potential of the first MCP plate with respect to ground is varied between 0.0 and 0.7 kV to fine tune the amplification of the signal to match the requirements of the performed experiments. The broadening of the MCPs is in fact the dominant term in the detector resolution, which is measured to be 95 \( \mu \text{m} \). For experiments that do not require the resolution to be minimal, the camera offers a binning feature to speed up experimentation. Setting the binning of the camera to \( n \) means the camera will force...
Figure 3.3: Schematic representation of the detector setup, with the event cascade caused by a single electron shown. The detector consists of two MCPs, a phosphor screen, an imaging lens and a CCD camera. Bias voltages are applied to both MCP plates and to the phosphor screen.

Figure 3.4: Logarithmic plot of the current signal through the MCPs as a function of the MCP voltage $V_{\text{MCP, in}}$.

the CCD chip to read out $n \times n$ pixels as one. Individual pixels on the camera are 22 $\mu$m across, so that e.g. binning 10 would result in a detector resolution of 220 $\mu$m.

An advantage of using the MCPs as amplifier for the electronic signal, is that the current that passes through the two MCPs can be measured. The MCP current is proportional to the number of electrons that reach the MCPs and can thus be used as a relative charge measurement. The charge amplification of the two MCPs varies exponentially with the voltage that is applied to the first MCP. The exponential behavior of the current signal as a function of the MCP voltage is visible in figure 3.4, measured for different wavelengths of the ionization laser. Less charge is released when using higher wavelengths. An absolute calibration of the MCP current signal using the Faraday cup has not been performed as of yet.

In figure 3.5, the complete setup as just described is displayed schematically. All components that were mentioned, the vacuum chamber with pumps and valves, the beam line with the rotation point and the charged particle optics and the two detector setups are labeled.
3.2 Source emittance measurements

With the setup that was just described, we can trap a cloud of ultracold gas, ionize a part of the gas using photo-ionization and accelerating either ions or electrons towards the further beam line. Inside the beam line, the ions fly straight towards the detector unaffected by the magnetic charged particle optics or the earth magnetic field. The electrons pass through the beam line reacting strongly to all magnetic fields, making the trajectories complex, yet also granting more control over the beam dynamics. How these aspects of the setup can be used to determine the quality of the UCPC charged particle source is described in this section.

3.2.1 waist scans

One of the most instinctive ways to measure the angular spread of the beam would be to simply move a camera along the beam line and measure the size of the beam at various \( z \)-positions. The increase in size of the beam along the beam line can be used to calculate the angular spread. With the current beam line however, the angular spread that is measured in that case would not be the source angular spread. The beam passes by several focussing or defocussing elements in the beam line so that the angular spread measured further down the beam line has changed with respect to the angular spread directly after the source.

Using the method of shifting the camera down the beam line does however allow the researcher to determine the emittance of the beam at the position of the camera in the beam line. The emittance was introduced in section 1.2 as a measure for the beam quality. The emittance is most easily determined by measuring the beam size around a waist. For a waist in the beam, the normalized emittance can be calculated using equation (1.7). The waist in the beam can be created using the solenoid lens described in the previous section.

The emittance of the electron beam is a conserved quantity along the beam line. The source emittance is thus equal to the emittance measured in the waist further down the beam line. If the size of the source can be measured, the angular spread of the source can be calculated, again using equation (1.7). A technique to measure the transverse size of the ionization volume, the ion spot size scan, is discussed later in this section.

The UCPC setup does not allow for the camera to be moved along the beam line to measure the beam size around a waist. An alternative way of measuring the characteristics of the beam waist is by moving the waist along the camera, in stead of moving the camera along the waist. By increasing the current through the solenoid lens, the waist can be placed either behind the camera,
on the camera or before the camera. The measurement technique just described to measure the emittance of the beam is called the waist scan.

The waist scan can be modelled by calculating the size of the beam throughout the setup, taking into account all components present in the beam line. The beam is assumed to be Gaussian in both transverse dimensions, and is also assumed to have a Gaussian angular spread distribution. The electrons bunches can thus be described by a two dimensional ellipse in phase space. In the so-called ellipse formalism, the size of a Gaussian beam in the two transverse dimensions $\sigma_x$ and $\sigma_y$ and the transverse velocity spread of the beam in both transverse parameters $\sigma_{\theta_x}$ and $\sigma_{\theta_y}$ is kept track of. As the shape of the position and velocity distributions is assumed not to change, keeping track of the transverse sizes and angular spreads is sufficient to know the entire phase space distribution. Two vectors are defined with the phase space parameters, for each transverse dimension one,

$$\vec{x} = [\sigma_x, \sigma_{\theta_x}]$$
$$\vec{y} = [\sigma_y, \sigma_{\theta_y}].$$

The effect of a drift or a focussing element in the beam line is modelled using a matrix formalism. Two-by-two matrices are defined that represent either a drift or a focussing element. To define these matrices, we assume abberation-free operation of the elements present in the setup, all optics are perfect. The vectors that represent the phase space distribution are multiplied by the matrices to calculate the change of the phase space distribution throughout the beam line. For the matrices, the on-axis elements are unity since the size of the beam and the angular spread of the beam are not influenced by their own size for a drift or a perfect focussing element. During a drift, the angular spread of the beam influences the size of the beam. This is represented by a contribution of the upper right matrix element, making the matrix describing drift

$$M_d = \begin{pmatrix} 1 & d \\ 0 & 1 \end{pmatrix}.$$

For a focussing element, the angular spread of the beam is changed dependent of the size of the beam. This relation can be described using the bottom left matrix element, so that

$$M_f = \begin{pmatrix} 1 & 0 \\ -\frac{1}{f} & 1 \end{pmatrix}.$$

To calculate the final phase space distribution vectors $\vec{x}_f$ and $\vec{y}_f$, the initial phase space distribution vectors $\vec{x}_i$ and $\vec{y}_i$ are multiplied by either $M_d$ of $M_f$ for every drift or focussing element in the beam line. The multiplication needs to be in reverse order of the elements occurring in the setup, so that the multiplication for the first optical element is performed first

$$\vec{x}_f = (M_N (M_{N-1} (\ldots (M_2 (M_1 \vec{x}_i)))).$$

In essence, the waist scan can be modelled as a drift-lens-drift geometry, with the initial conditions of the beam being a waist, as shown in figure 3.6. The matrix representation of the drift lens drift configuration is $M_{\text{waistscan}} = M_{\text{d,2}} \cdot M_f \cdot M_{\text{d,2}}$. The final beam size on the detector $\sigma_{\text{f,x}}$ is calculated using $M_{\text{waistscan}}$ to be

$$\sigma_{\text{f,x}} = \frac{\sigma_{\text{i,x}}}{f_x} \sqrt{\frac{\sigma_{\text{n,x}}^2}{\sigma_{\text{i,x}}^2} \left( (f_x - l_{\text{det}}) l_0 + f_x l_{\text{det}} \right)^2 + [f_x - l_{\text{det}}]^2},$$

(3.4)

where, as is also indicated in figure 3.6, $f_x$ is the focal point of the magnetic solenoid lens, $l_0$ is the distance from the first waist to the lens and $l_{\text{det}}$ is the distance from the lens to the detector.
Using equation (3.4), combined with equations (3.2) and (3.3) for the focal point of the solenoid lens, the size of the beam on the detector can be calculated as a function of the current through the magnetic solenoid lens. The shape of a waist scan depends strongly on the beam emittance. A fit to a measured waist scan with the emittance as a fit parameter and the current through the solenoid lens thus gives a value for the emittance of the beam.

The waist scans that are measured in the UCPC setup are not so perfect as the schematic situation in figure 3.6. Several components of the beam line influence the shape of the beam before it reaches the lens. As mentioned in the previous section, after leaving the waist at the source, the beam is defocussed by the accelerator kick, it is focussed by the field of the MOT coils and it is focussed in one dimension and defocussed in the other dimension by the quadrupole in the vacuum vessel. All these effects are included in the model for the beam line in the form of drift and focussing matrices with the appropriate focal distances and drift lengths.

To be able to perform an accurate fit to measured waist scan data in the realistic beam line model, several more fit parameters need to be introduced besides the emittance of the beam. Since the exact origin of the quadrupole component in the vacuum vessel is still unknown, both the position and the strength of the quadrupole are fit parameters. The strength and the position can depend on the alignment of the beam and the mean beam energy, and indeed the characteristics of the quadrupole vary between experiments. Imperfections in the solenoid lens are accounted for by using a multiplication factor for the focal distance of the lens: the magnetic lens factor $C_{\text{sol}}$.

The magnetic lens factor is also a fit parameter, capable of changing the position of the minimum in the waist scan. If the value of the magnetic lens factor deviates far from unity, the measurement was performed under poor conditions and will not be trusted.

In figure 3.7, an example of a waist scan measurement is displayed, along with modelled waist scan curves fit to the data. The waist scan was measured using the femtosecond ionization laser with a mean wavelength of 489 nm and with an accelerating voltage of 6 kV. We see that the beam size at the CCD camera detector is in the order of millimeters. The beam size in the two transverse dimensions differs due to the quadrupole contribution. The beam size initially decreases for low currents, suggesting the focus of the solenoid lens lies behind the detector. For increasing currents, the lens focusses stronger, decreasing the size of the beam on the detector. Due to the quadrupole component, the $\sigma_2$ dimension reaches its minimal spot size first at approximately 1.1 $\mu$m. At 1.3 A, the $\sigma_1$ dimension is focussed. For even higher currents, the beam is overfocussed, so that the spot size increases with the current.

In figure 3.7, we see that the sizes of the beam are not indicated as $\sigma_x$ and $\sigma_y$, but as $\sigma_1$ and $\sigma_2$. The labelling of the transverse beam dimensions with more arbitrary labels is due to a side effect of focussing using a solenoid lens. The focussing capabilities of a solenoid lens are derived...
from the bundle rotating due to the magnetic fields while traversing the solenoid. The rotation depends on the applied magnetic field, and thus changes during waist scan measurements. To deal with this effect, the long axis and the short axis of the bunches on the CCD detector are used as the coordinate system. Linking $\sigma_1$ and $\sigma_2$ to the source’s $x - y$ coordinate system requires complex modelling and does not benefit the experimental results.

The fit parameters determined from the measurement are $d_{quad} = 0.40$ m, $kl_{quad} = 0.15$ T, $C_{sol} = 1.14$ and an emittance of $\varepsilon_{n,x} = 1.4 \pm 0.2$ nm-rad. The beam size, determined using the ion spot size technique discussed in the next section, is $23 \times 30$ $\mu$m. The angular spread calculated using equation (1.7) is $0.10 \pm 0.01$ mrad. The transverse source temperature associated with this angular spread is $18 \pm 2$ K. Measurements of the transverse beam quality as a function of the wavelength and the ionization laser polarization are presented in the next chapter.

The waist scan is an indirect measurement of the emittance of the beam, which raises questions on the accuracy of the technique. Using the beam line model, the influence of the change of the source emittance on the shape of the waist scan was researched. In figure 3.8, we see the data points of same waist scan measurement as is displayed in figure 3.7, with the fit produced from the model. Using the same beam line parameters, initial spot size, magnetic lens factor and quadrupole strength and position, simulations were performed for source emittances a factor $\sqrt{2}$ higher and lower than the measurement, so that the transverse source temperature is a factor two lower and higher.

The simulations for $\sigma_1$ displayed in figure 3.8a show a clear difference between the waist scan curves for the different values of the emittance. The simulated curves have the waist at the exact same current of the solenoid lens, with the same beam size. The beam size in the waist should be smaller for a smaller emittance. The limiting factor for the beam size in the waist is however not the emittance but the detector resolution, which is equal for all three curves. The difference is primarily visible for the lower currents in the solenoid lens, where the measured data points lie neatly in between the simulated curves with different emittance. The waist scan is thus a technique capable of measuring subtle differences in emittance, with sub-nm-rad resolution. The theoretical minimum for the angular spread predicted by disorder induced heating is approximately $7 \cdot 10^{-5}$ rad, which corresponds to an emittance of 0.7 nm-rad assuming an ionization volume size of 10 $\mu$m, so that the accuracy of the waist scan measurement is sufficient.
Ion spotsize scan

It was mentioned several times in the previous discussion of the waist scan that the measured quantity of the waist scan is the emittance of the beam. To be able to determine the angular spread and with that the temperature of the source, the size of the ionization volume must be determined. To measure the size of the ionization volume, we use the ion spot size scan. As the name suggests, not electrons but ions are used for this application.

Ions have several beneficial characteristics for the determination of the initial spot size. Ions are ionized with a very low excess energy. All the energy from the laser is absorbed by the electrons, so that the initial angular spread of the ions is negligible. Moreover, due to the large mass of the ions, the influence of magnetic fields on the trajectories of the ions is negligible. These two effects make for an easy determination of the initial spot size from the ion spot sizes measured on the CCD camera detector.

Several effects do however influence the trajectory of the ions. Firstly, since the accelerator utilizes electric fields, the ions are affected by the accelerator kick. The ions also drift across the entire beam line, which gives rise to expansion of the ion bunches. These two effects can be combined into a single magnification term for the ion beam. Assuming these two effects were the only effects present, a single shot of the ion spot size would be sufficient to trace back the transverse ionization volume size.

The ions are however also affected by charge expansion within the time of flight from the MOT to the CCD camera. Charge expansion in a charged particle bunch typically takes place on the time scales that can be expressed as a modified inverse plasma frequency, \( \tau = \sqrt{\frac{2m_e e_0 \epsilon_0}{2n_e q^2 e}} \), where \( \epsilon_0 \) is the permittivity of free space and \( n_e \) is the particle density [8]. For ions, the inverse plasma frequency is significantly lower than the time of flight. The ions expand during their time of flight, with an expansion rate that depends on the charge density in the pulse. The effect of the charge expansion can be included in the beam line model for the ions as a defocussing element with a focal distance defined by [10]

\[
\frac{1}{f_{ce}} = -\eta \sqrt{\frac{n_e q^2}{3\epsilon_0 U}}
\]

where \( \eta \) is a constant determined by the Gaussian distribution of the bunches and \( n_e \) is the density of electrons in the bunch. With this focal distance, the final size of the ion spot can be calculated as
Figure 3.9: Example of an ion spot size scan performed to measure the initial ionization volume size in the $(x,y)$-plane.

\[\sigma_{x,f} = \left( M_{11} + M_{12} \eta \sqrt{\frac{nuq}{3\epsilon_0 U}} \right) \sigma_{x,i}, \quad (3.6)\]

where constants $M_{11}$ and $M_{12}$ depend on the accelerator structure and the beam line geometry. We see now that the final size depends on the root of the density of the pulses, with a constant offset. For pulses with less and less charge, the root of the density will go to zero, so that only the constant $M_{11}$ determines the final spot size from the size of the ionization volume. If the size of the ion spot size is measured for decreasing charge in the bunches, the measurement points will tend towards this asymptote. This asymptotic value divided by the measured beam size will give the ionization volume size.

The charge of the ion bunches can be decreased by decreasing the intensity of the ionization laser. This is performed using neutral density (ND) filters, that decrease the laser intensity with a factor of $10^{ND}$, with a approximately flat frequency response. The charge of the ion bunches is in good approximation linear dependent on the intensity of the laser. If the ND filtering is increased linearly, the charge in the bunches will therefor decrease exponentially. The final spot size depends on the root of the density in equation (3.6), so that it will depend on the ND filtering of the ionization laser as $10^{-ND/2}$. A fit can be performed to the asymptotic curve measured by increasing the ND filtering, so that the ion spot size for zero charge can be determined. In figure 3.9 an example of an ion spot size scan is visible, along with a fit described by \( y = y_0 + A \cdot 10^{-ND/2} \). The measured values for the ion spot size in figure 3.9 are scaled with $M_{11} = 46.7$, so that the value for $y_0$ is equal to the ionization volume size.

Just as for the waist scans, the sizes of the beam are listed in the long-axis, short-axis coordinate system, that depends on the orientation of the ellipsoidal shape of the bunches. For the case of ions however, the coordinate system formed by the long axis and the short axis of the ion bunch is easily related to the source’s $x$–$y$ coordinate system. The ion bunches are not rotated by the magnetic fields, so that the source’s $y$-dimension is also the vertical dimension on the camera.

In the example of the ion spot size scan, we see that the predicted shape of the curve is mirrored by the measured data. The final spot size that is measured in this example is $24 \times 14 \mu m$, a typical size for the ionization volume. This measurement suggests an elliptical transverse profile of the ionization volume. An elliptical ionization volume was measured consistently during experiments, which is explained by the ionization laser profile being narrower than the excitation laser profile.
This is confirmed by the long axis coinciding with the \( y \)-direction of the source.

### 3.2.3 Independent emittance measurement

The results presented in this thesis depend heavily on the waist scan and ion spot size scan measurement techniques. The waist scan and ion spot size scan measurement techniques both rely on the beam line model, so that changes in the setup that might occur during normal operation could influence the accuracy. The measured values for the angular spread and the emittance of the beam measured with the waist scan and ion spot size scan techniques are believable, but an independent conformation of the results is important to be able to fully rely in the techniques. An independent measurement of the angular spread and the emittance of the beam is presented by the work of Rammeloo [10].

The pepper-pot measurement technique relies on placing a TEM-grid in the beam line, so that the electron beam is partly clipped by the grid, and small beamlets are transmitted. A schematic representation of the situation is presented in figure 3.10. We see the incoming beam being separated into several smaller beamlets. From the emerging beam profile, the size of the beam at the TEM grid can be determined from the imprint of the TEM grid in the beam, since the size of the TEM grid wires is known. The angular spread of the beam at the TEM grid can be determined using the profile of individual beamlets.

The angular spread of the beam causes all electrons that pass by close to the edge of the TEM grid line to spread out, so that the cut-off caused by the TEM grid becomes less and less sharp further down the beam line. Since the angular spread distribution of the beam is Gaussian, the shape of the cut-off caused by the TEM grid lines is an integrated Gaussian, an error function. The width of the edge of the cut-off thus is a measure for the angular spread in the beam. By fitting error functions to the edges in the profile of individual beamlets, the angular spread at the grid can be determined. Using the local angular spread and the beam size at the grid, the emittance of the beam can be determined.

A raw image of a pepper-pot measurement is visible in figure 3.11a. The signal dips caused by the grid lines of the TEM grid are clearly visible in the profile of the beam. A one dimensional
line integration of the raw image is presented in figure 3.11b. The position of the grid lines of the TEM grid are indicated in the integration as circles. These circles enable the rescaling of the x-axis of the figure to size on the grid in stead of size on the CCD camera to determine the size of the beam at the position of the grid. The flanks of the dips in the integration provide the necessary information for the determination of the angular spread. The signal at the dips does not go all the way down to the constant background level at approximately 1700 counts. This effect is attributed to a signal dependent Gaussian background signal. This can for example be cause by bleeding of the pixels to adjacent positions. The signal dependent background is fitted and removed from the images prior to determining the slope of the dips.

The emittance determined from the pepper-pot measurements is comparable to the emittances found using the waist scan technique. The results of the pepper-pot method match well with the waist scan data for the low excess energy regime. The low emittance listed for the UCPC setup of 1 mm·rad is confirmed by the pepper-pot measurements, so that the high beam quality is independently measured using two different techniques. This makes the claim of high beam quality from this novel source for ultra short electron bunches that much stronger. Practical problems with using the pepper-pot measurement technique, such as signal dependent background, make the results of pepper-pot measurements less dependable than the results from waist scan measurements. For that reason, the emittance will be measured using the waist scan technique in this work.
Chapter 4

Temperature and charge measurements

A model for the angular spread or the transverse source temperature of the electron beams produced in the UCPC setup using a femtosecond ionization laser is presented in chapter 2. The dependence of the beam temperature on the ionization laser wavelength, on the accelerating voltage and on the ionization laser polarization is simulated. Several predictions were also made for the dependence of the charge of the bunches on these same parameters. In this chapter, measurements of the transverse source temperature and the bunch charge are presented, gathered using the measurements techniques presented in chapter 3. A paper on the source temperature measurements presented in this chapter and on the model presented in chapter 2 has been submitted to Nature Communications [23].

4.1 Wavelength dependence

The transverse source temperature was measured for electrons photo-ionized using the femtosecond ionization laser as a function of the ionization laser wavelength scanned from 489 to 477 nm with 1 nm increments. The electrons were accelerated to a mean energy of 3 keV with an accelerating voltage of 6 kV. The results of the measurement are displayed in figure 4.1. Note that on the x-axis, the wavelength increases to the left of the graph, to ensure that high energy particles are displayed on the right side of the graph. The results of a simulation performed for the same parameters using the model from chapter 2 are displayed alongside the experimental results as a solid line. The measured results mirror the trend predicted using the model.

For low excess energies, or for high ionization laser wavelengths in figure 4.1, the temperatures match the simulation results very well. Up to 483 nm, the data points follow the simulation with a slight offset. The prediction of the model that the UCPC setup should be capable to consistently produce low transverse temperature and ultra short beams is confirmed by these results. For wavelengths lower than 483 nm, the measured temperature departs from the simulation curve. The data shows monotonous increasing behavior, like the simulation. The temperature increase of the measured data for lower wavelengths is however more rapid than the increase of the simulated temperature. This result suggests additional heating effects for higher excess energies.

The imperfections that occur in the various components of the beam line, such as aberrations in the magnetic solenoid lens and stray magnetic fields, are not included in the beam line model. The imperfections degrade the quality of the electron beam and thus broaden the beam. If the beam is broadened by factors that are not included in the beam line model, then the fit model will contribute all those broadening effects to the temperature of the beam. Effects such as aberrations of the magnetic optics or stray magnetic fields thus cause source temperature to be measured that is higher than the actual source temperature.

Abberations caused by an imperfect optic depend both on the field geometry of the optic and
Figure 4.1: Transverse source temperature versus the wavelength of the ionization laser. The measured data and the simulation are shown for electrons ionized with the femtosecond ionization laser.

The absolute value of the charge of the electron bunches was also measured as a function of the wavelength of the ionization laser scanned from 489 to 477 nm with 2 nm increments, for $V_{\text{acc}} = 6$ kV. The results of the measurements are displayed in figure 4.2. The results were measured using the Faraday cup described in chapter 3. The laser energy was measured using a standard light intensity meter from the brand Coherent. The energy of individual laser pulses $E_{\text{ion}}$ was measured directly after the MOT to be $0.46 \pm 0.02 \mu J$.

We see that an electron bunch that is photo-ionized using the femtosecond laser contains in the order of $10^3$ electrons. The data shows saturation for lower wavelengths within the measured wavelength range. From the photo-ionization model described in chapter 2, the charge of the bunches should depend on the ionization laser wavelength as described by equation (2.5). The function $Q(\lambda)$ has only one free parameter which is fitted to the data. The resulting fit function is displayed as the solid curve. The critical wavelength according to the model is $485.8$ nm for $V_{\text{acc}} = 6$ kV, the width of the error function is $4$ nm and the initial charge is fixed to 0 C by applying the constraint $Q_0 - Q_1 = 0$. The final bunch charge, the only fit parameter, is $0.38 \pm 0.05$ fC. The fitted curve, for which only the amplitude was fitted and three parameters came directly from the model, matches the measured data well, validating the model.
Figure 4.2: Charge of the electron bunches versus the wavelength of the ionization laser, for $V_{\text{acc}} = 6$ kV. The solid line is a fit of equation (2.5) to the data.

4.2 Polarization effects

Using the broad bandwidth photo-ionization model, predictions were made for the dependence of the transverse source temperature on the polarization angle of the ionization laser, by assuming the distribution of electrons over the starting angles follows the angular distribution of the intensity of the linearly polarized ionization laser. The temperature curve is expected to be sinusoidal when measured as a function of the polarization angle. A minimum in the temperature is expected to occur when the ionization laser is polarized in the $z$-direction and a maximum is expected when the ionization laser is polarized in the $x$-direction. Defining the polarization angle to be $0^\circ$ for a polarization in the $z$-direction and $90^\circ$ for a polarization in the $x$-direction, then the oscillations in the source temperatures are described by a negative cosine of the polarization angle. Measurements of the transverse source temperature as a function of the ionization laser polarization angle are presented in this section.

The polarization effect on the transverse source temperature was measured twice on different days. Each of these measurement series has strong and weak points. The results of the two datasets are similar, but shed light on the model in a complementary way. Both datasets will therefore be displayed and discussed in this section. To disambiguate the two datasets, they are labeled dataset 1 and dataset 2, labeled in chronological order based on measurement date.

In figure 4.3 the transverse source temperature is displayed as a function of the polarization angle for wavelengths from 485 to 475 nm with 2 nm increments, for both measured datasets. Sine fits to the data points are shown alongside the data as solid curves as a guide to the eye. In figures 4.3d and 4.3c, the temperature measurements from dataset 1 are displayed, with the sinusoidal behavior clearly visible. During experimentation, the half wave plate that was used to change the polarization angle of the ionization laser was rotated from $0^\circ$ to $180^\circ$ with $10^\circ$ increments. The polarization angle was thus scanned from $0^\circ$ to $360^\circ$ with $20^\circ$ increments. Two waist scans were performed for every setting of the polarization angle to improve the accuracy of the source temperature measurements. We see that for lower wavelengths, so for higher excess energies, both the mean temperature and the oscillation amplitude of the temperature oscillations increase. We see that for the high temperature results in figure 4.3d, the zero polarization angle coincides with a minimum in the source temperature, resulting in a $-\cos$ as expected. In dataset 2, visible in figures 4.3b and 4.3a, we also see the sinusoidal behavior as a function of the laser polarization. During experimentation for dataset 2, the increments of the half wave plate were reduced to $5^\circ$.
and one waist scan was performed per setting of the polarization. This resulted in more noise in the measurements, making the \(-\cos\) signal less well defined. Especially for the 477 nm series, we see that noise plays a large role. The first minimum of that fit does not coincide with the zero of the polarization angle. Due to the bad quality of the 477 nm measurements from dataset 2, no conclusions can be drawn from it.

To quantify the temperature oscillations, three parameters can be determined from performing a sine fit. Two of these parameters were already mentioned in chapter 2, the mean and the amplitude of the oscillations. According to the simulations, both parameters increase monotonously as a function of the wavelength and have the same general shape, similar to figure 4.1. The third fitting parameter is the phase shift of every oscillation. The phase should be 0° for all the measurements judging from the model, resulting in negative cosine curve shapes as a function of the polarization angle. Small changes in the experimental conditions can result in deviations from this ideal behavior, so that fitting the phase is justified. All three fitting parameters will be discussed in the rest of this section.

The mean of the transverse source temperature oscillations as a function of the ionization laser wavelength is displayed in figure 4.4 for both datasets. Together with the wavelengths already displayed in figure 4.3, the dependence on the polarization was also measured for \(\lambda_{\text{ion}} = 481\) 483 and 485 nm. For dataset 2, we see that the mean temperature matches the simulated data in the same manner that the data matches the simulation in figure 4.1. For higher wavelengths, the data
matches really well, yet for lower wavelengths, the measured temperature increases faster than the simulated temperature. This behavior was explained in the previous section. For dataset 1, we see that the measured source temperature follow the simulated data even closer, showing the same ever increasing behavior with increasing slope. The measured data is however significantly lower than the simulated data for \( \lambda_{\text{ion}} = 477 \) and 479 nm. Since the model includes no imperfections, it is deemed impossible for the measured temperature to be significantly lower than the simulated temperature. To explain the discrepancy, we assert the measured initial spotsize of dataset 1 is too large, so that the angular spread comes out too small.

In figure 4.5 the amplitude of the measured oscillations is displayed alongside the predictions from the broadband photo-ionization model. The same behavior is visible in the amplitude curve as in the mean temperature curve, a plateau for higher wavelength and a sharp increase for the lower wavelengths. This behavior is mirrored by the measured data. We see however, that the measured amplitude is systematically lower than the amplitude predicted by the model. This effect is worse for dataset 1 than for dataset 2. This can be explained by the temperatures for dataset 1 being lower all together, so that the amplitude will also be lower than measured in dataset 2. Regardless, both for dataset 1 and for dataset 2, the amplitudes are lower than was predicted, which we would like to explain. The polarization state of the ionization laser light was measured to see whether the direction was set properly, and whether the polarization did not become elliptical during experiments. With a polarizing beam splitter cube and a light meter, the components of the light in each direction were measured. The ratio of the intensity in the intended direction \( I_{\text{int}} \) and the perpendicular direction \( I_{\text{perp}} \) is at least 30. Such small imperfections in the polarization of the ionization laser light do not seem a likely cause for the temperature oscillation amplitude being that much smaller. Another likely cause could not be found.

The phase shift, or the starting angle of the oscillations, was a fit parameter in the model. We define that the phase shift is 0 rad in the case that the temperature oscillation behaves as expected, which is that it has a minimum for a zero polarization angle. The phase shift is defined to be \( 1/8\pi \) rad if the oscillation starts in the equilibrium position for a zero polarization angle. We have already seen that for the oscillation at lower wavelengths visible in figure 4.3, the phase shift is approximately zero as expected. This is visible in figure 4.6 in the left half of the graph. For higher wavelengths however, the phase shift changes for both dataset 1 and dataset 2. For both datasets, the phase shift goes towards \( 1/8\pi \) rad, so that the temperature oscillations are a negative sine function, starting with the mean temperature and subsequently decreasing for higher
polarization angles. This behavior occurs for both datasets and is probably related to a persistent effect. The oscillations that are measured for the high wavelengths show clear sinusoidal behavior, so that the phase shift is not due to noise in the data, as it is with the 477 nm datapoint from dataset 2. The cause for the measured phase shift for higher wavelengths is as of yet unknown.

The model does not predict any dependence of the charge of the electron bunches on the laser polarization. Engelen did however notice a dependence of the charge on the polarization during experimentation with the narrow band ionization laser with nanosecond pulse lengths [21]. The behavior seen by Engelen is a sinusoidal oscillation of the charge as a function of the polarization angle with minimal charge for a polarization in the z-direction, similar to the oscillations seen in temperature. The relative amplitude of the oscillations typically measured by Engelen is approximately 64%, varying e.g. from 0.4 fC for a polarization angle of 0° to 1.1 fC for
90°. Although the oscillations of the bunch charge are not well understood, the relative charge was measured for the broad band polarization experiments presented in this work. The relative charge signal was measured using the current through the multi channel plates in the detector setup. The results of the relative charge measurements are presented in figure 4.7a for $\lambda_{\text{ion}} = 475$, 477 and 479 nm for both datasets.

In the measurement series, no clear dependence can be discerned of the charge of the bunches on the polarization angle of the ionization laser. The charge signal cannot be compared between dataset 1 and dataset 2, since the MCP voltage setting was different between the two. The absence of an effect of the laser polarization angle on the charge can be explained by the broad bandwidth of the laser. A paper was published by Freeman et al. on the polarization dependence of the cross section of photo-ionization of rubidium atoms [32]. In this paper, the cross section for photo-ionization is displayed for both the polarization direction parallel and perpendicular to the acceleration direction, for wavelengths in the range from 295 to 301 nm, that lie around the ionization wavelength for ground state rubidium atoms. The cross-section oscillates vary rapidly both in the $x$- and the $z$-direction, in counter-phase, so that maxima in the cross-section in the $x$-direction coincide with minima for the cross section in the $z$-direction. Using a narrow bandwidth laser such as used by Engelen, one single value of the oscillating cross-section applies. With the broadband laser used in this work however, the oscillating cross-section is convoluted with the broad Gaussian spectral envelope of the ionization laser, which results in an averaged value for the cross-section. The effect of the averaging of the oscillations in the cross-section is that the difference between the $x$- and $z$-direction becomes negligible, so that the polarization angle no longer influences the bunch charge.

In this chapter we have seen that the transverse source temperature can be measured reproducibly. Source temperatures as low as 20 K are reported, with emittances under 1 nm·rad. The dependence of the transverse source temperature on the wavelength of the ionization laser was measured for $V_{\text{acc}} = 6$ kV and found to match the predictions from the broad band single source photo ionization model. The charge of the bunches follows the behavior predicted in the model for varying ionization laser wavelengths, saturating for shorter wavelengths. Also, the effect of the variation of the polarization angle of the ionization laser was measured. The measurements were found to mirror the simulations, which predict sinusoidal oscillations in the source temperature. Simulated results for the mean, the amplitude and the phase shift of the oscillations were presented alongside the measured data, with again a good match. Finally, the charge of the bunches as a function of the polarization was researched. No clear dependence was seen. In the next chapters, theory on electron diffraction and diffraction experiments are presented, as performed using the UCPC setup.
Chapter 5

Pragmatic diffraction theory

As we saw in the Introduction, a major goal of the UCPC project is to perform diffraction with macromolecular targets. The beam quality must however first be quantified using a well known, robust sample. The angular spread of the electron beam can be measured directly during diffraction experiments. The determination of the angular spread from a diffraction pattern will be discussed along with other diffraction related subjects in this chapter.

Samples for diffraction experiments using the UCPC setup must be sufficiently thin and have a sufficiently low density for the relatively low energy electrons to be able to pass through the sample. Extremely thin graphite samples that consist of one to six monolayers of graphene will be used to conduct the diffraction experiments, with research and interest in the area of graphite and graphene recently increasing.

An important reason graphene is specifically suitable for experiments using ultrafast electron diffraction is that the coupling of electrons to phonons in the graphene plane is extremely fast [33]. The coupling takes place on two different timescales. An initial relaxation takes place at timescales in the range of $70 - 120$ fs, while a second relaxation takes place in $0.4 - 1.7$ ps. Using fs electron bunches, these couplings can potentially be resolved in the time domain and be studied.

The diffraction sample is mounted at the position previously occupied by the Faraday cup, at $28.5 \pm 1$ cm from the detector. The sample is mounted on a TEM grid with a diameter of 3 mm. With a minimal spot size smaller than $100 \mu m$ as seen in figure 3.7, the sample diameter is sufficient.

The driving principle behind diffraction are electrons passing through a sample material and being scattered from the atoms present in the sample. Two different scattering events are possible, elastic events and inelastic events. Elastically scattered electrons leave the sample with specific angles, giving rise to diffraction patterns as can be seen in figure 5.1. Electrons that are inelastically scattered exit the sample at random angles, providing little extra information and causing a background signal. Lastly, electrons can pass through the sample unscattered, so that the intensity of the central spot of the diffraction pattern increases.

The elastically scattered electrons form a pattern dependent on the sample characteristics. If a sample is monocrystalline, well defined diffraction spots will be formed on the screen. A diffraction pattern measured of a sample of monocrystalline graphite is displayed in figure 5.1a. If a sample is polycrystalline, the diffraction spots that are visible for a monocrystalline sample will occur for a large set of azimuthal angles, so that the pattern of spots is rotated around the central spot. This rotation results in a pattern of circles, as can be seen in figure 5.1b.

The samples that are used during experiments in this research consist of flakes of tens of microns in size, while the electron beam is several times larger in size. The surface area of the electron beam is about an order of magnitude larger than the domain size, so that the sample is effectively polycrystalline for the current beam geometry. With a polycrystalline sample, we expect to see diffraction rings during experiments.
5.1 Scattered fractions

By studying electron matter interactions, several parameters can be modelled analytically to guide the diffraction experiments that are planned for the UCPC setup. The calculations presented in this section are performed for electron bunches that are accelerated to a mean longitudinal energy of 10 keV. The elastic cross section, a measure for the probability of elastic scattering events, for electrons traversing a graphite sample is documented by NIST [35]. The elastic cross section can be used to calculate the amount of elastically scattered electrons. The value of the cross section for graphene is \( \sigma_e = 0.2244 \cdot a_0^2 \), where \( a_0 \) is the Bohr radius.

The mean free path \( \Lambda \) is the expected distance an electron can traverse before it is scattered in the target material, either elastically or inelastically. The mean free path can also be defined specifically for either elastic scattering events \( \Lambda_e \) or for inelastic scattering events \( \Lambda_i \). The elastic mean free path is expressed in material constants and physical constants as

\[
\Lambda_e = \frac{A}{N_A \sigma_e \rho},
\]

where \( A \) is the atomic weight of the target material in gram/mole, \( N_A \) is Avogadro’s constant and \( \rho \) is the mass density [36]. For a graphite sample, the elastic mean free path is \( \Lambda_e = 14.8 \) nm.

The inelastic mean free path and the total mean free path can also be determined using equation (5.1), by substituting the appropriate cross sections. The inelastic cross section can be determined from the elastic cross section using a simple empirical relation \( \sigma_i = \frac{20}{Z} \sigma_e \) where \( Z \) is the atomic number. From this relation we see that for graphite (\( Z = 6 \)) the inelastic cross section is more than three times larger than the elastic cross section. Electrons will therefore be more likely to scatter inelastically, giving rise to a shorter inelastic mean free path: \( \Lambda_i = 4.43 \) nm. The total cross section that represents both scattering processes is simply equal to the sum of both cross sections, so that the total mean free path is \( \Lambda = 3.41 \) nm.

Using the calculated mean free paths, the unscattered, elastically scattered and inelastically scattered fractions of the total number of electrons can be calculated using the Lambert-Beer law.
This approximation only holds for sample thicknesses \( d \) much smaller than the total mean free path \( \Lambda \) so that the chance of multiply scattered electrons is negligible. The unscattered fraction is \( F_u = \exp \left( -\frac{d}{\Lambda} \right) \). The elastically scattered fraction and the inelastically scattered fraction are both calculated as \( F_{e/i} = 1 - \exp \left( -\frac{d}{\Lambda_{e/i}} \right) \). Calculating these fractions for example for a sample with a thickness of 0.335 nm, we see that \( F_u = 0.906 \), \( F_e = 0.022 \) and \( F_i = 0.073 \).

A sample thickness of 0.335 nm is not coincidental, since this is the interlayer spacing in graphite. The interlayer thickness is an order of magnitude smaller than the mean free path, so that multiple scattering events are negligible, as can be seen by the fractions adding up to 1. To be able to calculate the fraction of the electrons that undergo a single elastic scattering event for thicker samples, a multilayer approach is utilized. The thicker sample is divided into multiple layers, each much thinner than the mean free path. Each layer either doesn’t interact with the electron, or scatters the electron either elastically or inelastically. In a two layer system for example, we end up with a fraction of the electrons that is unscattered, a fraction that scattered elastically once, a fraction that scattered inelastically once, a fraction that scattered elastically twice, a fraction that scattered inelastically twice and a fraction that scattered once elastically and once inelastically. By keeping track of all these fractions, a realistic estimation can be made of the scattering behavior of thicker samples.

An efficient way of keeping track of all scattering events in this multilayer model is by introducing a polynomial with powers of the fractions \( F_{u/e/i} \) to keep track of scattering events. Looking again at the example of scattering in a two layer system, the total unscattered fraction after both layers is e.g. \( F_{u,\text{tot}} = F_u^2 \). The polynomial that can be used to calculate the scattered fraction for an \( N \) layer sample is

\[
(F_0 + F_{el} + F_{inel})^N
\]

Each term in the expanded polynomial represents a unique combination of scattering events in the modelled layers of the sample. Using this polynomial, direct expressions can be derived for several fractions that are relevant to the problem. The total fraction of unscattered electrons after \( N \) layers is simply

\[
F_{u,\text{tot}} = F_u^N.
\]

Those electrons all end up at the central spot. The total fraction of once elastically scattered electrons, the electrons that form the diffraction pattern, is

\[
F_{e,\text{tot}} = NF_u^{N-1}F_e.
\]

The total fraction of inelastically scattered electrons that form a large Gaussian background is

\[
F_{i,\text{tot}} = 1 - (F_u + F_e)^N.
\]

The results of the calculations are shown in figure 5.2. A maximum can be seen for the once scattered fraction. The position of this maximum can be calculated using differentiation of the expression for \( F_{e,\text{tot}} \), which results in \( N_{e,\text{max}} = -1/\ln(F_u) = 10 \). This maximum can be understood as an interplay of few electrons being scattered for thin samples, and multiply scattered electron fractions becoming dominant for thick samples. The maximum scattered fraction thus lies at higher sample thicknesses for higher order scattering events.

Looking at figure 5.2 for thicknesses that agree with the present samples, so in the range of 1 to 6 monolayers of graphene, we see that the unscattered fraction is quite large. E.g. for a sample of 4 monolayers, the fractions are \( F_u = 0.67 \), \( F_e = 0.067 \) and \( F_u = 0.26 \). These fractions, combined
with the fact that many of the elastically and inelastically scattered electrons also end up in the central spot, suggest that the central spot will be orders of magnitude brighter than the diffraction rings. To be able to conduct diffraction experiments without saturating the central spot on the camera, it is useful to include a beam stop in the beam line to be able to block the central spot.

## 5.2 Ring pattern

The multilayer theory predicted the fraction of electrons that are scattered elastically. To determine how the elastically scattered electrons are distributed over the diffraction pattern, another model is developed in this section. To determine how a radial lineout or azimuthal integration would look, we need to know the width, intensity and radial position of each ring, assuming the ring shape is Gaussian. In figure 5.3 various parameters are indicated surrounding the formation of a diffraction pattern.

Important parameters to quantify the diffraction patterns are the characteristics of the incoming beam at the sample (angular spread $\sigma_{\theta_{x,y}}$, size $\sigma_{x,s}$ and energy $U$), characteristics of the sample (lattice parameter $d$) and the geometry of the setup (distance from sample to detector $D$). Also several parameters are introduced to quantify the diffraction pattern: central spot size $\sigma_{x,d}$, diffraction angle $\theta$, angular spread of the diffracted ring $d\theta$, radius of the ring on the detector $R$ and width of the ring on the detector $dR$.

From the detail in figure 5.3 the angle $\theta$ for which the first order of the diffraction ring occurs can be determined. A peak in the pattern is caused by constructive interference between the scattered electron wave functions. Constructive interference occurs between scattered electron waves when the path length difference is exactly an integer multiple of the wavelength. This criterion is made visible in the red triangle in the detail of figure 5.3, which leads to the grating equation

$$d \sin (\theta) = \lambda. \quad (5.2)$$

The wavelength of the electrons can be calculated using the de Broglie wavelength equation. For electrons travelling at non relativistic speeds ($U \ll 511$ keV), the de Broglie wavelength $\lambda_{\text{deBroglie}}$ is given by
Figure 5.3: Schematic representation of the diffraction pattern being formed on the detector by a diverging beam passing through a sample. Important parameters are indicated for the incoming beam ($\sigma_{x,y}, \sigma_z$), the sample ($d$) and the setup geometry ($D$). Parameters to quantify the diffraction pattern are $\sigma_{x,A}, \theta, d\theta, R$ and $dR$.

\[ \lambda_{\text{de Broglie}} = \frac{h}{\sqrt{2m_eU}}, \]

in terms of Planck’s constant $h$, the electron mass $m_e$ and the mean electron energy $U$. The relativistic correction for electrons moving with $u = 10$ keV is only a few percent, so that the assumption of non-relativistic physics is justified.

The lattice parameter $d$ that partly determines the scattering angle arises from the structure of the sample, in our case graphene. The structure of a graphene sheet is a hexagonal grid of carbon atoms. The structure repeats itself with a certain real space distance $a$, the distance from one carbon atom to his second nearest neighbor. Looking at the hexagonal geometry, $a$ is thus related to the carbon-carbon bond length $l_{C-C}$ as $a = \sqrt{3}l_{C-C} = 0.246$ nm. To be able to parameterize the two dimensional graphene plane, two lattice vectors are necessary. Several equivalent choices exist, one of which is two vectors $\hat{a}_1$ and $\hat{a}_2$ both with length $a$ and with a mutual angle of $60^\circ$.

To make the way $d$ follows from the lattice vector plausible, the Miller indices formalism will be introduced.

To be able to characterize a three dimensional sample of multiple graphene layers, three unit vectors are needed together with a unit cell. The three vectors are the two previously discussed vectors $\hat{a}_1$ and $\hat{a}_2$, and a third vector $\hat{\zeta}$ that is perpendicular to the first two vectors with a length $\zeta$, where $\zeta$ is twice the interplane distance between graphene sheets, 0.67 nm. The length of the vector $\zeta$ is twice the interlayer distance, since the position of carbon atoms in graphene planes in consecutive layers is not equal.

Steps in the lattice can be parameterized by linear combinations of the unit vectors. A step in the lattice quantified by vector $\vec{x} = \eta\hat{a}_1 + k\hat{a}_2 + \zeta\hat{\zeta}$ can be parameterized using the Miller indices $(\eta k l)$. A first simple example of the use of Miller indices is to quantify the beam geometry. The electron beam is assumed to enter perfectly perpendicular to the graphene plane, so that it’s Miller indices are $(Ml)_{\text{beam}} = (001)$.

For diffraction, it is known that structures that are in the direction parallel to the beam do not contribute to the diffraction pattern. With a perfect (001) beam, structures with a nonzero
Miller index therefor will not play a role. Also, the structures with the shortest Miller indices vector provide the smallest rings. For example the structure (100) will contribute to the first ring. Five other structures ((010), (−100), (0 − 10), (−110) and (1 − 10)) also contribute to this first ring. In the monocrystalline diffraction pattern visible in 5.1a these six structures are all separately visible as equidistant diffraction peaks.

Reimer and Kohl [36] present a simple equation for calculating the lattice parameter d for different sets of Miller indices. For a hexagonal lattice that is

\[ d = \frac{a}{\sqrt{\frac{4}{3} (\eta^2 + k^2 + \eta k) + (a/c)^2 l^2}}. \]

which is derived from the transformation of the unit vectors to reciprocal space. Filling out the sets of Miller indices for the first ring results in \( d_{(100)} = \sqrt{3}a/2 \). For the second ring we can now also see the lattice parameter. The second ring is formed for the combination of \( \eta \) and \( k \) for which the term \( \eta^2 + k^2 + \eta k \) is the smallest value larger than one. The next ring occurs for Miller indices (110), (−1 − 10), (2 − 10), (−210), (1 − 20) and (−120), with a lattice parameter of \( d_{(110)} = a/2 \). To calculate the width of the ring on the detector the infinite crystal approximation is used. The assumptions made for the approximation are an infinitely large crystal and an infinitely large distance from the sample to the detector. In this case, finite beam sizes on the sample do not contribute to the size of the ring on the detector. Only the angular spread in the beam at the sample \( \sigma_{x,s} \), or alternatively, the temperature of the beam at the sample \( T_s \) determines the ring width, so that \( d\theta = \sigma_{x,s} \). The angular spread can now be expressed as

\[ d\theta = \frac{\sigma_{x,s}}{p_z} \approx \sqrt{\frac{k_b T_s}{2U}}. \]

To compare the angular predictions with the experiments, the ratio of the angular spread and the diffraction angle \( \theta \) can be compared to the ratio of the width of the ring and the radius of the ring. These ratios are equal in the small angle approximation. If the small angle approximation is then also applied to the expression for the diffraction angle \( \theta \) from equation (5.2) then the ratio of \( \frac{d\theta}{\theta} \) can be expressed purely in simulation- and problem parameters as

\[ \frac{dR}{R} \approx \frac{d\theta}{\theta} \approx \frac{\sqrt{3}}{2} \frac{d}{h} \sqrt{m_e k_b T_s}. \] (5.3)

The infinite crystal approximation is a good first approximation in case the beam on the sample is extremely small. If we however assume a finite beam size on the sample and a short distance from the sample to the detector, the infinite crystal approximation breaks down because the beam size at the sample is no longer negligible compared to the broadening of the ring due to angular spread in the beam. The approximation can be improved greatly by simply adding the beam size on the sample to the broadening effect of the angular spread. The equation for the ring width

\[ dR = \sigma_{x,s} + R \frac{d\theta}{\theta}, \] (5.4)

is derived by converting the expression in equation (5.3) to \( dR = R \frac{d\theta}{\theta} \) and subsequently adding \( \sigma_{x,s} \).

The new model for the ring width requires the size of the beam on the sample. The size of beam on the sample can be determined from independent waist scan measurements combined with modelling of the beam line using the beam line model introduced in chapter 2. By performing a waist scan, the strength and the position of the quadrupole and the magnetic lens factor can be

48
determined. If these parameters are inserted in the beam line model along with a value for the solenoid lens current, the size of the beam can be calculated at every position along the beam line. The width of the diffraction ring can now be determined purely from problem parameters, simulated results and results from other, independent experiments.

The model for the width of the diffraction rings that was just discussed also provides the insight needed to be able to measure the beam quality using a single diffraction pattern, which is one of the major goals of performing diffraction experiments on the UCPC setup. By determining the width of the ring experimentally, and independently measuring the size of the beam on the sample, the angular spread can be determined from the ring pattern. This measurement would present a powerful and direct proof of the high beam quality that can be produced using ultracold charged particle clouds.

The relative intensity of the diffraction peaks can be predicted using kinematical diffraction theory [36]. The full treatment of diffraction in reciprocal space needed to perform these calculations is beyond the scope of this thesis. Some results taken from literature can however give a strong indication of what is to be expected for the relative intensity of the first two peaks in the diffraction pattern with respect to each other.

For bulk graphite, the structure of the sample is intrinsically three dimensional. Performing calculations on the graphite structure, the intensity of the first diffraction ring will be a factor of 3 lower than the second ring [37]. The same calculation can however also be performed for the intrinsically two dimensional graphene sample. In this case the second ring is 0.8 time as intense as the first ring. The two calculations however do not give any insight for the regime in between the two extremes.

During experiments, it is to be expected that the measured ratio between the second and the first ring for a sample of multiple domains with 1 to 6 monolayers of graphene will be in between the two theoretical extremes of 0.8 and 3. Measurements have previously been performed on a graphene like system, so that a likely value can be found from literature. In Fauteux et al. [38], the the second ring is 1.8 times as intense as the first ring, making the ratio between peaks 1.8. Since theory does not seem to give a ready answer to this problem, the relative intensities found by Fauteux et al. will be assumed for simulating purposes.

With knowledge on the position of the rings, the width of the rings and the relative intensity, an azimuthal integration of the diffraction pattern can be simulated. To simplify comparison of the simulated data to the measured data, the radial position with respect to the central spot on the detector is chosen as the running variable. The calculated angles $\theta$ and $d\theta$ can be converted to distances using the distance from the sample to the detector $D = 28.5 \pm 1$ cm. For the simulated diffraction lineout visible in figure 5.4, the electron beam is assumed to have a mean energy 11 keV, a beam size on the sample of 400 $\mu$m and the transverse beam temperature of 110 K. These values are chosen to match actual experimental conditions.

The simulated diffraction patterns has line positions at $15.6 \pm 1$ mm and $27.1 \pm 2$ mm. As can be seen in equation (5.4), the width of both rings is equal in the model and is $417 \mu$m. Measured diffraction patterns will be presented in the next chapter. The measurements will be analyzed to fit the framework that is presented in this chapter and will be compared to the analytical results.
Figure 5.4: First two diffraction rings simulated using calculations for ring position, width and relative intensity. Assumed values are mean electron energy of 11 keV, 400 µm beam size on the sample and 110 K beam temperature.
Chapter 6

Diffraction experiments

As was mentioned in the previous chapter, one of the present goals of the UCPC experiments is to show the beam quality to the community in a convincing manner. When using a diffraction sample and measuring the width of the diffraction rings or spots that are formed on the detector, the angular spread of the beam can be determined. Using the UCPC setup, the first diffraction patterns were recorded using a sample of flakes of graphene deposited on a TEM grid, which will be presented in this chapter.

6.1 Diffraction patterns

The diffraction patterns recorded during experiments required a very large shutter time to be able to clearly see the diffraction rings in the scaled pictures. Twenty separate images were recorded using identical settings, each with an integration time of hundred seconds. Using the nanosecond laser with a repetition rate of 10 Hz, this brings the total amount of electron bunches used for an image to 200. With approximately $10^4$ electrons per pulse, the total amount of charge is estimated to be 0.3 pC. Using the femtosecond ionization laser, the amount of charge per pulse is about a factor of ten lower, only $10^3$ electrons per pulse, as seen in figure 4.2. The repetition rate however is ten times higher, so that the amount of charge used to build up a diffraction pattern is approximately equal between the two used ionization lasers.

The diffraction patterns were all recorded using an ionization laser wavelength $\lambda_{\text{ion}} = 480$ nm. The wavelength was chosen to balance two effects. For high wavelengths, so for low excess energies, not many electrons are ionized. For low wavelengths, so for high excess energies, the transverse source temperature is high, compromising the quality of the diffraction patterns. As a running parameter for the experiments, the bunch energy $U$ was varied from 6 to 11 keV with 1 keV increments, using each of the two lasers. The current in the magnetic solenoid lens was set so that the entire electron beam passes through the sample, which was checked for each measurement series by measuring the central spot without the beam block. This is achieved for magnetic lens currents in between $I_{\text{magn}} = 1.7 - 2.2$ A. Waist scans are performed during each measurement series to extract necessary information about the electron beam, being the transverse source temperature $T$ and the beam size on the sample $\sigma_{\text{xs}}$.

One of the diffraction patterns, recorded using the femtosecond laser and a beam energy of 11 keV, is displayed in figure 6.1. The diffraction pattern consists of two clearly visible diffraction rings, along with a very bright central peak. The central peak seems to be highly saturated, but that is due to scaling of the figure. The inset at the upper right section of figure 6.1 shows the image without scaling. In the inset, the diffraction rings are not visible, due to the overpowering intensity of the central peak. The inset also shows that the beam block that was used during these experiments is not sufficient to block the outer flanks of the Gaussian central peak during the extended shutter times of the experiments.

The background that is visible in the image is caused by artifacts in the camera, and by actual
beam dynamics. As mentioned in the previous chapter, more electrons are scattered inelastically than elastically. The inelastically scattered electrons form a large Gaussian background signal centered at the same position as the pattern. Also, the flanks of the extremely bright central peak pass by the beam block to add a large amount of signal to the central peak.

Several defects in the detector setup cause distortion of the pattern. The MCP detectors have previously been shown to measure a signal dependent background, e.g. during pepper-pot measurements by Rammeloo [10]. The detector also contains randomly distributed hot pixels that fire continuously due to failure of these pixels in the camera. Furthermore, a large structure of pixels (visible in the bottom left corner of figure 6.1) lights up consistently. Background measurements in the absence of charge arriving at the detector confirm that both the hot pixels and the structure in the corner are detector artifacts independent of the arriving charge. The larger bright structure was found to be caused by a fault in the screening for ambient light and will be ignored. The hot pixels are filtered from the raw camera images.

In figure 6.1 the yellow rectangle indicates the area where the signal contains least noise and other artifacts. The bottom right corner of the yellow rectangle rests at the centre of the diffraction pattern. The centre of the pattern is determined manually. Next, every pixel in the indicated area is assigned a radial position. The values for all pixels with identical radial position is summed and divided by the total amount of pixels. The raw azimuthal integration described here is displayed in figure 6.2. We see the very intense central peak, that is partly blocked by the beam block. The
very large Gaussian background can be seen spanning the entire visible range. The diffraction peaks, much less intense than the central peak, are visible as small bumps on the signal.

To analyze the diffraction pattern, the background signal of the peaks in the integration is subtracted, in the form of a third order polynomial that is fit to the background signal. The relevant parameters of the measured rings are determined by fitting a Gaussian function to each of the peaks in the resulting peaks. The width of the ring is defined as the variance of the Gaussian, the surface under the Gaussian curve is taken as the intensity, the position is trivial. The first parameter that we compared to theory is the position of the diffraction peaks. The diffraction angle $\theta$, that can be directly determined from theory, is plotted versus the electron beam energy in figure 6.3. The measured ring positions on the detector were converted to angles using the distance from sample to detector. This distance determines the error bars of the data. The results agree with the calculated angles well within the error bars. The grating equation is thus suitable for predicting the positions of diffraction rings.

The isolated peaks created from the azimuthal integration the diffraction pattern measured using the femtosecond laser with $U = 11$ keV are displayed in figure 6.4. Displayed alongside the measured peaks is a simulation of the diffraction pattern. The simulation is calculated for a beam with $T = 110 \pm 5$ K and $\sigma_{xs} = 0.40 \pm 0.04$ mm, both results from a waist scan. The beam size on the sample was calculated making use of the beam line model described in section 3.2.1 with parameters from the waist scan as input. The relative intensity of the simulated peaks is chosen to match Fauteux et al. [38], the absolute intensity is chosen so that the rightmost diffraction rings are equally high. The simulated diffraction peaks match the measured peaks quite well. The measured intensity ratio is 1.2, within the boundaries set by theory. It does not perfectly match the factor 1.8 measured from literature. This can be explained by a difference in sample thickness between the two experiments, where the lower ratio of in our experiments suggests a higher degree of monolayer graphene presence in our sample. Further analysis of the absolute or relative intensity of the diffraction peaks falls outside the scope of this thesis.

The predicted width of the rings depends on the angular spread of the beam and the size of the beam on the sample. The parameters determined from the waist scan result in an angular spread $d\theta$ that corresponds to a theoretical broadening of the rings of 12 $\mu$m, calculated with equation (5.3). This broadening due to angular spread thus is negligible compared to the contribution of the beam size. This result also holds for the other diffraction pattern in the dataset. The experiments...
Figure 6.3: The diffraction angles of the measured diffraction rings are plotted versus the bunch energy, alongside simulated results using the grating equation. The error bars of the measured data are determined by the error in the distance from the sample to the detector.

Figure 6.4: Azimuthal integration of figure 6.1 within the yellow rectangle with the background subtracted. In red, a simulation of the two diffraction rings is displayed.

performed are thus not suitable for determining the beam quality directly from the diffraction patterns. Later on in this chapter, several suggestions will be discussed for reaching the goal of measuring the beam quality directly using a diffraction measurement.

The measured width of the diffraction rings is not only dominated by the size of the electron beam as suggested by the model. The measured width is consistently several hundreds of microns bigger than the predicted width. This suggests several other broadening effects are present outside the thermal angular spread. Two of such effects are broadening of the ring by all kinds of multiple scattering events and contributions of diffraction peaks with nonzero miller index $l$, due to the beam not traversing the sample with a perfect (001) geometry. The two broadening effects just mentioned are probably small effects. What is considered to be the dominant broadening effect is the time integrated beam jitter. The beam position on the sample and on the detector is not constant, but moves around shot to shot. The beam size on the sample is measured with a shutter
time of 1 s. The long shutter times during the diffraction experiments causes the signal of all the bunches to be summed, so that the result is a larger spot than the size of the individual bunches. This effect of jitter in the beam affects the measured width of the rings of the diffraction pattern.

The precise contribution of the integrated jitter cannot be quantified, since that would involve determining the spotsize for individual electron bunches. A strong indication for jitter can however be found in the dataset. For every diffraction pattern, twenty shots were taken to build up statistics. To research the effect of jitter on the same timescale of the shutter time of individual images (100 s), the width of the diffraction rings can be determined as a function of the amount of images averaged. For two different measurement series, the width of the ring as a function of the integration time is displayed in figure 6.5. We see that the measured width of the diffraction rings increasing with integration time. This increase primarily takes place at the shorter time scales, and saturates for longer times. The saturation suggests that no very long term (∼30 min) jitter effects are present in the setup. The effect of the jitter on the beam size that is visible on the measured timescales is a few percent. It is expected however that the largest jitter effect is present at much smaller timescales. It is probable that most of the extra broadening can be attributed to this effect.

### 6.2 Experimental improvements

In the previous section the first diffraction experiments were described that were performed using the UCPC setup, both with ns electron bunches and with ultrashort, fs electron bunches. In the bigger picture of the UCPC project, it is an important step that diffraction using ultrashort electron bunches was seen for the first time, since the ultimate goal of the ultracold electron source is to be able to perform pump probe experiments using ultrafast diffraction as a diagnostic. Although the experiments are not suitable for direct measurement of the beam quality, several improvements can be made based on the previous experiments.

In the inset in figure 6.1, it is visible the outer flanks of the Gaussian central peak are still much brighter than the diffraction rings. Although it is not imperative for improved diffraction experiments, it will be beneficial for the signal to background ratio to increase the size of the beam block in the setup. We recommend to make a beam block that is about a factor of two wider. A second consideration concerning the beam block is that the position of the center of the diffraction...
pattern was relatively constant during experiments. The beam block design can be optimized with that knowledge, by shortening the beam block so that it only covers half the width of the detector. The electrons that form the diffraction rings are not stopped by the beam block in that case.

To improve the accuracy with which the diffraction pattern can be measured, a decrease in the necessary integration time would decrease the influence of the beam jitter on the measured widths. From figure 6.5 we can conclude that if the integration time of individual images can be reduced, whilst keeping the diffraction pattern visible, the beneficial effect on the beam size can be very significant. In that case, the center of the pattern must be determined for each individual image and those centers must then be overlapped before combining images for one series.

The shutter time of the images can be decreased more and more if the amount of electrons per bunch is optimized. Aside from optimizing the alignment of the various lasers involved in the creation of the electron bunches, the intensity of the excitation and the ionization lasers can be increased by at least an order of magnitude. Increasing the laser intensity increases the amount of charge per bunch linearly as seen in section 3.2.2. When the charge of the bunch increases with a factor the integration time decreases with the same factor.

To be able to better see the effect of the angular spread as a contribution to the total ring size, the angular spread should be increased, while also decreasing the size of the beam on the sample. To make the beam as small as possible on the sample, a second solenoid lens has been added to the setup closer to the sample. The shorter distance from lens to sample enables the use of a larger focussing angle, so that the eventual waist can be smaller. The subsequent angular spread is also larger after the waist. The lens that has been placed was previously used by van Oudheusden [8]. The lens is characterized by are a number of windings of 600, inner radius of 5.84 cm, outer radius of 9.16 cm and lens length of 4.2 cm.

A simple emittance calculation provides an indication for the achievable beam size on the sample. The lens is placed 34 cm before the sample, which is 92 cm from the MOT position at z = 0. Assuming the size of the beam is about 600 µm at the second magnetic lens (this is the same size the beam has at the first solenoid lens), then the focussing angle is 1.8 mrad. Assuming a normalized emittance of 1 nm·rad, a nominal value for the UCPC setup, then the minimal spot size in the waist located at the sample is calculated to be about 1 µm using equation (1.7). The beam line model discussed in section 3.2.1 has been altered to incorporate the second solenoid lens using equations (3.3) and (3.2).

### 6.2.1 Point to point focussing

The setup improved with the second solenoid lens is however plagued by the astigmatism present in the setup. With the large focussing angle, the astigmatism in the beam can cause a large discrepancy between the beam sizes on the sample of both transverse dimensions. To achieve a small beam size on the sample, the focus should be at the same longitudinal position for both transverse dimensions. An extra element should be added to the setup to be able to focus both dimensions to a single point. By adding a quadrupole lens to the setup, point to point focussing can be achieved. Note that the single quadrupole lens is enough to enable point to point focussing, but that full correction of the astigmatism in the beam is not possible.

In figure 6.6a a typical astigmatic waist scan can be seen. The important parameter for enabling point to point focussing is the current difference between the minima for both transverse dimensions δI, indicated in the figure using an arrow. If using the quadrupole lens, the two minima were made to overlap, the goal of point to point focussing is attained. Practically, the quadrupole lens is installed in the setup by rewiring the first of the two steering coils so that it operates as a quadrupole lens. The lens is positioned 47 cm from the MOT.

Both the quadrupole current $I_{quad}$ and the quadrupole angle $\alpha_{quad}$ must be set correctly to enable point to point focussing. The angle of the quadrupole is chosen relative to a zero position with the axes of the lens coinciding with the $x-$ and $y$ direction defined for the setup. Experiments were conducted using the nanosecond ionization laser operated at 480 nm and accelerating voltage at 6 kV, to ensure stable beam operation. Waist scans were recorded for a coarse scan of $\alpha_{quad}$ and $I_{quad}$ to try and see which approximate settings decrease $\delta I$. After the coarse scan, a more
Waist scan of a beam geometry that lacks the capability of point-to-point focusing. The waist occurs for different focusing currents for the two transverse dimensions. The current difference between the two minima has been indicated.

Waist scan of a beam geometry that was altered using the quadrupole lens to be able to perform point-to-point focusing. The beam waist now occurs at the same focusing current for both beam dimensions.

Figure 6.6: Waist scans of the beam with different settings for the quadrupole lens.

detailed scan is performed of the interesting part of parameter space. Since the astigmatism is assumed to be caused by a charged mirror that is positioned directly under that beam, a clear symmetry is assumed to be present in the angle of the quadrupole.

The optimal settings for the quadrupole lens are $\alpha_{\text{quad}} = 90^\circ$ and $I_{\text{quad}} = 0.15$ A. The waist scan that is measured using these settings is displayed in figure 6.6b. We see that the minima for both transverse dimension coincide, so that point to point focusing is achieved. The beam size however still varies between the two dimensions, so that the astigmatism in the beam is not completely solved. The optimal angle of $90^\circ$ satisfies the symmetry argument given earlier.

For the listed beam parameters of $\lambda_{\text{ion}} = 480$ nm and $V_{\text{acc}} = 6$ kV, the quadrupole settings for point to point focusing were found. The exact origin and the precise mechanism of the astigmatism in the beam is however not known. The astigmatism may depend on the beam energy, either longitudinal or transverse, but also on the bunch charge, or the charge density. It is therefore unsure whether the listed settings for the quadrupole lens are universal for all ionization laser wavelengths, for both nanosecond and femtosecond electron bunches and for all acceleration voltages. Several waist scans that were measured following the work on point to point focusing suggest that the listed setting are not sufficient to ensure point to point focusing for significantly different beam characteristics. A more detailed analysis of the astigmatism problem is left for future investigation.

The beamline model was updated to incorporate the quadrupole lens that was introduced here. The quadrupole lens was pragmatically introduced by copying the already present quadrupole term and setting the position to $d_{\text{quad}} = 0.47$ cm. The $kl$ value that is used to indicate the strength of the quadrupole lens in that general description can be related to $I_{\text{quad}}$ using the waist scan results presented in figure 6.6. The relation is however not absolute, since the quadrupole term presumably caused an magnetic element in the vacuum vessel has the tendency to vary in between measurements, which in turn varies the $kl$ value found in the model for the quadrupole lens.

Using the changed beam line model with the additions of the second solenoid lens and the quadrupole lens, a likely scenario can be simulated for the beam geometry during future diffraction experiments. To create a small focus on the sample, a balance must be found for maximizing the beam size on the second solenoid lens, whilst minimizing lens aberrations. The effect of aberrations increases for electrons that are positioned further off axis. Lens aberration also increase for higher lens currents. To balance the desired high focusing angle with the aberration, the beam will be collimated using the first solenoid, and subsequently focussed on the sample using the second
solenoid lens. Simulations conducted by van der Geer confirm the suggested beam geometry as being the most proficient at making a small focus.

A simulation of the geometry is displayed in figure 6.7. We clearly see the disturbance of stigmatic beam geometry at $z = 0.25$ cm, a value often found during experiments. Subsequently, we see that the quadrupole lens defocuses the more convergent beam and focusses the more divergent beam, to enable the first solenoid lens to create a neatly collimated beam. The second solenoid lens focusses the beam on the sample, and in the simulation a minimal spot size is achieved on the sample of $5 \times 7 \, \mu m$. The spot size on the detector finally becomes $390 \times 570 \, \mu m$, making the diffraction pattern size obviously dominated by the angular spread over the spot size.

An added bonus of focussing the beam down onto the sample, is that the size of the beam on the sample is projected to become smaller than the single crystal domain size of the sample. For this we assume the single crystal domains are indeed tens of microns across, which has not been verified. With the beam illuminating only one or several domains, the diffraction pattern will change from a ring pattern to one or several spot patterns, all with the telltale sixfold symmetry of graphene. An advantage of the monocrystalline diffraction pattern is the increased intensity of the individual diffraction peaks, because the charge is distributed over a smaller area. The higher intensity again reduces the integration time needed to properly image the diffraction pattern. Also the added information visible in the images. The size of the diffraction peaks can be determined both the radial as in the azimuthal directions, giving an indication of the astigmatism left in the beam.

Summing up the suggestions and projected improvements for future diffraction experiments:

* Increase the amount of charge per bunch by optimizing MOT density and increasing both excitation laser and ionization laser intensity. With the increased charge, the integration time can be reduced, decreasing the broadening effect of beam jitter.

* Use a quadrupole lens to enable point to point focussing, so that a single focal point position can be achieved for both transverse dimensions of the electron beam. This minimizes the contribution of the electron spot size on the sample to the size of the diffraction patterns.

* Use a second solenoid lens installed closer to the diffraction sample to increase the focussing angle to the sample. The second lens also decreases the electron spot size on the sample, both by increasing the focussing angle and by decreasing the effect of aberration on the focus.
The previous suggested changes might result in a possible extra advantage by the diffraction pattern switching from rings to individual diffraction peaks, due to the beam size becoming smaller than sample’s single crystal domain size. Having separated diffraction peaks both increases the intensity of the diffraction pattern due to the electrons being spread out over a smaller area, and increases the amount of information in the diffraction pattern, since both the radial and the azimuthal peak size can be measured. The increased intensity decreases the necessary integration time and thus decreases the influence of beam jitter on the spot sizes.
Chapter 7

Conclusions and outlook

In this thesis, measurements are presented of the beam quality of electron beams that are extracted from our UCPC source, together with the first diffraction patterns that were measured with these beams. The transverse source temperature was measured as a function of the ionization laser wavelength and as a function of the polarization angle of the ionization laser. A photo-ionization model was developed with which the source temperature was calculated, and compared to experimental results. A pragmatic model was developed to analyze the diffraction patterns. Results from the model were compared to the measured diffraction patterns.

7.1 Beam quality measurements

In chapter 1 we saw that electron bunches used for UED experiments are required to be of an extremely high quality. The length of the bunches must be shorter than the processes under investigation (typically 100 fs), the bunches must ideally contain at least $10^4$ to $10^6$ electrons to be able to image a diffraction pattern in a single shot, the transverse coherence length should be larger than the lattice parameter of the sample and the emittance should be small enough to be able to focus the bunch to a spot of approximately 100 μm. In this thesis, electron bunches were reported with a transverse coherence length of 20 nm, larger than typical lattice parameters of protein crystals. The emittance of the bunches is measured as low as $1.4 \cdot 10^{-6}$ nm·rad, easily low enough to meet the minimal spot size requirement. The typical number of electrons in the measured bunches was $10^3$, one order of magnitude less than would ideally be required for single shot diffraction experiments. The electron bunches photo-ionized with femtosecond long pulses have a typical length of several picoseconds [22], too long to measure dynamics on the shortest time scales. Simulations performed using the photo-ionization model that is presented in this thesis corroborate the experimental finds.

The lowest source temperatures is measured using the femtosecond ionization laser with a wavelength of 489 nm and a rms bandwidth of 4 nm and with $V_{acc} = 6$ kV. The transverse source temperature of the UCPC setup was measured for $V_{acc} = 6$ kV for wavelengths from 489 nm to 477 nm, resulting in temperatures from 25 to 240 K. The low emittance predicted by the model is confirmed by the measurements. For shorter wavelengths, the measured source temperature becomes increasingly higher than the simulated results. Deviations of the measured temperature from the simulated curve are attributed to aberrations in the optics related to larger beam sizes. The overall shape of the simulations is mirrored by the measured data, which validates the proposed model.

The polarization of the ionization laser is an important parameter in the photo-ionization process. According to the model, the highest electron beam quality is attained with the polarization direction parallel to the direction of propagation when using femtosecond long ionization pulses and the behavior for the interlaying polarization angles is sinusoidal. For $\lambda_{ion} = 488$ nm and $V_{acc} = 6$ kV, the mean of the simulated oscillation is 25 K with an amplitude of 5 K. For $\lambda_{ion} = 478$
nm and $V_{\text{acc}} = 6$ kV, the mean is 125 K and the amplitude is 25 K. Measurements of the source temperature as a function of the polarization angle confirm the sinusoidal behavior. The mean value of the oscillations of the transverse source temperature as a function of the ionization laser wavelength follows the increasing behavior predicted by theory, but the measured mean increases quicker for short wavelengths than the simulated temperature. The measured amplitudes of the oscillations of the transverse source temperature are lower than the predicted amplitudes. No clear cause can be indicated for this discrepancy.

7.2 Diffraction patterns

The first diffraction patterns that have been measured with the UCPC source setup are also reported in this thesis. The diffraction sample consists of overlapping graphene flakes deposited on a TEM grid, 1 to 6 monolayers thick. The crystal domains of the sample are several times smaller than the electron beam at the sample, so that the diffraction pattern consists of rings. The diffraction angles of the first two diffraction rings of graphite were measured as a function of the beam energy and match with values calculated using the grating equation.

The angular spread of the beam at the position of the sample can be determined from the diffraction patterns. To find a low angular spread using a diffraction sample would be very strong proof that the beam quality of the UCPC source is high enough to perform UED experiments. Assuming the beam is focussed on the sample, the width of the diffraction rings is determined by the size of the waist and the angular spread in the waist. The angular spread determined from the measured diffraction patterns is many times larger than is expected based on the transverse source temperature. To improve the experiment, the beam dynamics are improved by adding a quadrupole lens to the setup to solve astigmatism issues and by adding a second lens to increase the focussing angle towards the sample. It is left up to future researchers to use the improved setup to deliver the definitive proof of the extreme capabilities of the UCPC source setup.

7.3 Outlook

Currently, the creation of electron bunches of several picoseconds in length, with approximately a thousand electrons in charge and with an emittance in the order of 1 nm·rad is everyday business with the UCPC source setup. By performing additional diffraction experiments, the final proof of the high quality of the electron beams can be delivered. Then, with the various aspects of the transverse beam quality well understood, the characterization of the electron bunches can continue in the longitudinal dimension. With the installation of a streak cavity, the simulations of the bunch length can be verified [39]. The understanding of the characteristics of electron bunches that are produced by the unaided UCPC source is then very far advanced.

At that stage, several beam line components can be added or altered to be able to meet several additional requirements of single shot UED. To meet the ideal bunch energy requirement, the accelerator structure can be updated to be able to produce 100 keV electrons. To compress the electron bunches to sub-picosecond lengths, a radiofrequency cavity can be added to the beam line. To be able to excite ultrafast processes, a pump beam line can be added to the setup including a delay stage. When such important improvements are made to the setup, it would be wise to also update the currently used magneto-optical trap setup to fiber-only operation, and to replace the currently used vacuum vessel with a much smaller one. With this improved setup, single-shot UED experiments are within reach, enabling the study of a whole new range of physical, chemical and biological reactions, both on a very small spatial and temporal scale.
Bibliography


