MASTER

Ultracold electron diffraction using an ultracold source

van Mourik, M.W.

Award date:
2013

Link to publication

Disclaimer
This document contains a student thesis (bachelor's or master's), as authored by a student at Eindhoven University of Technology. Student theses are made available in the TU/e repository upon obtaining the required degree. The grade received is not published on the document as presented in the repository. The required complexity or quality of research of student theses may vary by program, and the required minimum study period may vary in duration.

General rights
Copyright and moral rights for the publications made accessible in the public portal are retained by the authors and/or other copyright owners and it is a condition of accessing publications that users recognise and abide by the legal requirements associated with these rights.

- Users may download and print one copy of any publication from the public portal for the purpose of private study or research.
- You may not further distribute the material or use it for any profit-making activity or commercial gain.
Ultracold electron diffraction using an ultracold source

M.W. van Mourik

CQT - 2013 - 12

Eindhoven University of Technology
Department of Applied Physics
Coherence and Quantum Technology

Supervisors:
dr. ir. W.J. Engelen
dr. ir. E.J.D. Vredenbregt
prof. dr. ir. O.J. Luiten

January 2013 - November 2013
Ultrafast electron diffraction (UED) enables the study of molecular dynamics on nanometer length scales and sub-picosecond time scales. In UED, structural changes of a sample are induced by a femtosecond pump laser pulse, and are probed by an ultrashort (picosecond) electron bunch. The quality of the resulting diffraction pattern is limited by an intrinsic beam parameter that represents beam quality, the coherence length. The ultracold charged particle (UCP) set-up described in this work has been developed to improve beam quality in UED, with the future prospects of studying time-resolved dynamics of macromolecules. In the UCP set-up, magneto-optically trapped and cooled rubidium atoms are photo-ionized at near-threshold. The released electrons are accelerated, producing an ultracold electron beam. Magnetic lenses control the beam, allowing it to transmit through a sample before reaching a detector, on which diffraction patterns can be captured. Previous work has quantified the quality of the UCP beam by measuring the source parameters, the source size and the effective transverse source temperature.

In this work, the first diffraction experiments using ultracold, picosecond-long electron bunches are conceived and executed, using polycrystalline and monocrystalline graphite samples. The goal is to directly measure the beam quality through diffraction patterns. The set-up has undergone several improvements to accommodate diffraction experiments. For example, to monitor the electron beam’s path and keep it aligned, alignment plates have been placed at two points along the beam path. A double-lens configuration has been introduced to improve control over the beam’s waist position and angular spread. An analytical study of the beam path has provided the optimal lens configuration for diffraction experiments. Using polycrystalline graphite samples, diffraction ring patterns have been produced. Diffraction rings are difficult to analyze accurately, causing the measured ring width to be limited by analysis constraints; therefore, no absolute statements about beam quality can be made from ring patterns. A monocrystalline graphite sample has been produced by manual exfoliation of natural graphite blocks. When focusing the electron beam through this sample onto the detector, sharp diffraction spots are acquired, of which the size is limited by the resolution of the detector. A lower limit of the lattice distance that can be resolved with 100 µm samples is 3 nm. When focusing the beam onto the sample, broad spots are detected. By varying the effective transverse source temperature between 10 – 300 K, beam quality is shown to decrease accordingly: at 10 K, a coherence length up to 0.56 nm is found for a beam size at the sample of 3.3 µm. Increasing the source temperature to 300 K puts the coherence length to 0.39 nm with a beam size at the sample of 8.9 µm. These results are in good agreement with values expected from analytical beam theory, and simulated results.
## Contents

1 Introduction 4  
   1.1 Prospects of ultrafast electron diffraction 4  
   1.2 Beam quality 6  
      1.2.1 Phase-space 6  
      1.2.2 Emittance 6  
      1.2.3 Coherence length 8  
   1.3 Beam quality in ultrafast electron diffraction 8  
      1.3.1 General 9  
      1.3.2 Ultracold UED 9  
   1.4 This thesis 10  
      1.4.1 Chapter overview 10  
2 Experimental Set-up 11  
   2.1 Magneto-optical trap 11  
      2.1.1 Laser cooling and trapping 11  
      2.1.2 Rubidium 14  
   2.2 The UCP set-up 15  
      2.2.1 Lasers 17  
   2.3 Photo-ionization - source temperature 18  
   2.4 Ultracold charged particle beam 20  
      2.4.1 Beam source 20  
      2.4.2 Beam path 20  
3 Methods 23  
   3.1 Beam alignment 23  
   3.2 Beam parameters 24  
      3.2.1 Analytical beam model 24  
      3.2.2 Source size - Space charge scan 25  
      3.2.3 Effective transverse temperature - Waist scans 26  
      3.2.4 Beam energy - Time-of-flight scan 28  
4 Setting up for diffraction 29  
   4.1 Beam line parameters 29  
      4.1.1 Astigmatism 30  
      4.1.2 Double lens configuration 32  
      4.1.3 Temperature dependence 34  
   4.2 Diffraction theory 34  
      4.2.1 Required sample thickness 35  
      4.2.2 Reciprocal lattice space 37  
      4.2.3 Bragg condition 37  
      4.2.4 Diffraction from graphite 38
5 Results

5.1 Graphite samples .................................................. 42
  5.1.1 Sample preparation ........................................... 43

5.2 Diffraction rings ................................................... 44
  5.2.1 Ring patterns .................................................. 44
  5.2.2 Ring analysis .................................................. 45
  5.2.3 Waist scans ..................................................... 48
  5.2.4 The need for a monocrystalline sample ....................... 50

5.3 Diffraction spots ................................................... 50
  5.3.1 Sharp diffraction spots ........................................ 50
  5.3.2 Diffraction experiments: temperature dependence ........... 52
  5.3.3 Coherence length .............................................. 55
  5.3.4 Implications ................................................... 57

6 Discussion and Conclusions ......................................... 58

6.1 Research goals ..................................................... 58
  6.1.1 Set-up improvements .......................................... 58
  6.1.2 Experimental results .......................................... 59

6.2 Outlook ............................................................. 60
  6.2.1 Recommendations ............................................... 60
  6.2.2 Ultracold and ultrafast electron diffraction: Outlook ....... 61

A Submitted paper ..................................................... 67

B Space charge .......................................................... 73

C Diffraction angle ..................................................... 75
Chapter 1

Introduction

This report presents the work done during the course of the authors graduation project at the group Coherence and Quantum Technology (CQT) at the Eindhoven University of Technology (TU/e). The ultracold charged particle (UCP) set-up has been used for experimental work in the field of ultrafast electron diffraction (UED). In this chapter, the project is introduced, and the layout of the rest of this thesis is presented.

1.1 Prospects of ultrafast electron diffraction

Fundamental processes in physics, chemistry, and biology are often well understood in macroscopic terms. For example, the process of a polymer melting is macroscopically well known. However, when trying to understand such processes at atomic or molecular levels, solving the physics of it all becomes cumbersome, to say the least. Modern technology enables us to take films and photography to staggering scales: high-speed cameras record films at up to a million frames per second and certain microscopes such as STMs allow us to take pictures of single atoms. However, one of the big dreams in physics, chemistry, and biology remains to be able to combine the two, and making the so-called ‘molecular movie’ [1], recording ultrafast structural dynamics.

The study of ultrafast structural dynamics [2] is applicable in many fields of research. An example with a clear real world relevance is being able to monitor the structural changes in the folding and unfolding processes of proteins. Diseases such as Alzheimer’s, Parkinson’s, Diabetes Type II and BSE [3] are all attributed to incorrect folding of proteins. Understanding such processes is key in finding methods to prevent or correct such processes. However, protein folding processes typically occur at time scales between $10^{-9}$ s and $10^{-6}$ s [2] and length scales of a few nanometers [4]. ‘Filming’ a protein folding process at such time and length scales, remains problematic.

In 2009 the first X-ray Free Electron Laser (XFEL) has been developed which shows promise as a tool for the study of ultrafast structural dynamics [5]. The XFEL source produces $10 - 100$ fs X-ray pulses with $10^{13}$ photons. Such pulses are highly coherent and have been used in proof-of-principle demonstrations in which the molecular structure of protein crystals have been determined using diffraction [6]. However, using ultrafast X-ray diffraction for structural dynamics experiments has some setbacks: firstly, GeV electrons are needed to produce the X-ray beam, requiring $\sim 3$ km linear accelerators. Secondly, the energy transfer during a diffraction collision is high enough that damaging the sample under investigation is practically inevitable.

One prominent up-and-coming technique to overcome the obstacles present in the field of ultrafast structural dynamics, and a viable alternative to X-ray diffraction, is ultrafast electron diffraction (UED) [7, 8]. Electron diffraction provides insight to crystal lattice structures and sizes with sub-Angstrom resolution, and fs-laser induced pump-probe techniques allow monitoring of these lattice properties and changes at picosecond time scales. UED experiments are plentiful and indeed show the potential of using ultrashort electron pulses for diffraction in time-resolved
analysis of structural dynamics of relatively simple molecules [9, 10]. For example Siwick et al. [11] have studied processes in Al melting at atomic levels and picosecond time scales, in which the transition from a structured solid to an unstructured liquid is made visible.

A conceptual sketch of ultrafast electron diffraction is shown in Figure 1.1. An extremely short (ps) electron bunch is created by femtosecond photo-emission of a target. Electrons are extracted by means of an electric field, and accelerated to tens or hundreds of keV. The resulting electron beam can be modified both in the transverse direction (e.g. by magnetic lenses) or in the longitudinal direction (e.g. by an RF cavity). The beam passes through a thin crystal sample and undergoes diffraction. The diffracted beam is then imaged onto a detector. The ionization laser pulse passes a beam splitter before arriving at the target. The split-off beam arrives, with a tunable delay, at the crystal sample under investigation. Such a pulse, the so-called probe pulse, can alter the properties of the sample by, for example, inducing heat. These changes in sample properties are reflected by changes in diffraction images. By repeating shots for different delay times of the probe pulse and analyzing the diffraction results for each separate delay time, time-resolved measurements of crystal transitions are made, at time scales limited by the length of the electron pulse, typically picoseconds.

One of the foreseeable difficulties in reaching the next level, UED of macromolecules such as proteins, is providing a sufficiently high beam quality. Generally speaking, the larger and more complicated the crystal lattice becomes, the higher the quality of the beam used for diffraction needs to be. In this case, beam quality is quantified in terms of beam size and transverse angular momentum. Also, protein samples are delicate, meaning that a dose of high energy electrons can easily damage the sample. Ideally, diffraction patterns of such structures are captured with a single electron bunch, before the sample is damaged, so-called single-shot diffraction.

A proposed solution to reaching this new stage in UED is to replace the ‘standard’ photocathode [13] with a cold source, effectively decreasing the beam’s initial transverse angular momentum spread. Two experimental set-ups for investigating ultracold UED exist today, one at the group CQT in Eindhoven, and the other in Melbourne, Australia [14]. Using cold electrons provides exciting new possibilities in electron diffraction experiments. When compared to photocathodes, the beam quality is typically an order of magnitude higher, as will be quantified later in this chapter. When (naively) putting the two sources side by side, we see that under the same beam conditions (i.e. beam size at source, beam size at sample) we could either study samples that are ten times as small, or samples that have ten times the lattice distance, with the same resolution. While this comparison is admittedly not entirely justified, the advantage of using an ultracold source is apparent. Furthermore, other experimental parameters, such as bunch charge, can theoretically reach similar orders of magnitude in both cases.

In this work, an ultracold source for ultrafast electron diffraction is investigated with the UCP set-up at CQT. In the following sections, some background information in beam physics and the necessity for an ultracold source is introduced. With that information in mind, the goal of this research is described. Lastly, the layout of the rest of this thesis is described.
1.2 Beam quality

Depending on the application of the particle beam, it is important to have a suitable way to quantify the beam quality. In this section, the relevant parameters in beam physics are discussed, with a focus on applications in pulsed electron diffraction experiments.

1.2.1 Phase-space

In beam physics we refer to a particle beam when a group of (usually charged) particles share a common direction. A particle beam can be continuous, pulsed, or even a single shot. In the latter two cases, the group of electrons in each individual shot is referred to as a bunch. Each individual particle within a particle bunch can be described in terms of its position $x$, $y$, $z$ and its momentum, $p_x$, $p_y$, $p_z$. The position and momentum information about the entire set of electrons in a bunch are described in a 6-dimensional phase space [15, 16]. It is convenient to break down this 6-dimensional system to 3 2-dimensional representations of phase space, each describing the position and momentum in their respective Cartesian direction. This split is allowed since the motions in each direction is in most cases decoupled (an exception being the action of magnetic solenoid lenses, covered in 2.4.2).

The collection of the information of particles in phase space, more specifically the volume they take up, is used to define beam quality in a parameter called emittance.

1.2.2 Emittance

Let us look closer at the 2 dimensional phase space in one direction, $x$. In Figure 1.2 we see a typical distribution of particles in phase space, position $x$ on the x-axis and momentum $p_x$ on the y-axis. The particles occupy an effective area in phase space, outlined in red. The choice of where to place the border of this area is somewhat arbitrary; for example, it could be drawn to include 99% or 95% of the particles. However, in many cases the distribution in both position and momentum is Gaussian, making it a mathematically useful choice to define the border in terms of standard deviation, thus including 46% of the particles.

As a rule-of-thumb, the smaller the area that confines the particles in phase space in each separate dimension, the better the quality of the beam. The area of this ellipse (times a factor
1/$\pi$) is called the beam’s emittance $\epsilon$. The emittance is usually normalized to $\epsilon_n = \epsilon/(m_e c)$ ($m_e =$ electron mass, $c =$ speed of light), called the normalized emittance.

Normalized emittance in the x-direction is given by (1.1):

$\epsilon_{x,n} = \frac{1}{m_e c} \sqrt{\langle x^2 \rangle \langle p_x^2 \rangle - \langle xp_x \rangle^2}$,

(1.1)

Here the brackets $\langle \cdots \rangle$ indicate an average over the ensemble of electrons in the bunch. For convenience, all the emittances referred to in this work are normalized, and the subscript n will be dropped. In practice, a lower emittance means better beam quality, though the actual emittance value that constitutes as a good quality beam is dependent on the beam type and applications. What is not included in this measure of ‘beam quality’ is the beam current $I$, or the bunch charge. This is typically included in a parameter called beam brightness, $B = I/(8\pi^2 \epsilon_x \epsilon_y)$. In this work, the beam brightness is not a significant parameter, so will not be further developed in this thesis. However, in the future, when realizing single-shot diffraction, this parameter becomes more important.

In beam physics it is conventional to define the coordinate system so that the common direction of travel of the particles is in the z-direction, centered around the origin of the $x$ and $y$ plane ($\langle x \rangle = \langle y \rangle = 0$ and $\langle p_x \rangle = \langle p_y \rangle = 0$).

In a beam waist, the particles’ positions and momenta are uncorrelated, $\langle xp_x \rangle = 0$, so that Eq. 1.1 reduces to

$\epsilon_x = \frac{1}{m_e c} \sigma_x \sigma_{p_x}$,

(1.2)

where $\sigma_x = \sqrt{\langle x^2 \rangle}$ and $\sigma_{p_x} = \sqrt{\langle p_x^2 \rangle}$ are the root-mean-square (rms) spot size and momentum, respectively.

Emittance is typically a conserved quantity, though many phenomena can contribute to increasing it, such as particle scattering and space charge forces. Decreasing emittance of a beam is also possible, but often either very complicated (e.g. laser-cooling) or comes at the cost of reduced current (e.g. diaphragms). Phenomenologically, conservation of emittance tells us that it is possible to reach small beam spot sizes $\sigma_x$ by increasing angular spread $\sigma_{p_x}$. Conversely, a non-spreading beam can be achieved, but only for large beam sizes. Intuitively, emittance thus represents the ‘focusability’ of the beam.

Because the requirements of a bunch are different for the transverse and longitudinal directions, a distinction is made between transverse $\epsilon_{x,y}$, and longitudinal emittance $\epsilon_z$. In this research the longitudinal emittance is not a limiting factor, so we will focus primarily on the transverse emittance.

A consequence of conservation of emittance is that the quality of the beam is already determined at the beam’s source, which conveniently can be taken as a beam waist. Emittance at any point along a beam path can therefore be taken in terms of source parameters, as

$\epsilon_x = \frac{1}{m_e c} \sigma_{x,i} \sigma_{p_x,i}$,

(1.3)

where $\sigma_{x,i}$ and $\sigma_{p_x,i}$ are the source size and momentum spread, respectively. The transverse momentum spread of the electrons at the source can also be written in terms of transverse source temperature $T$ which is a more intuitive and interpretable parameter. Strictly speaking, the term temperature might be misleading, as the electrons’ momentum may not be described by a Maxwell-Boltzmann distribution. Still, we can relate the electron momentum spread to an effective temperature as

$\frac{1}{2} \frac{\sigma_{p_x,i}^2}{m_e} = \langle U_x \rangle = \frac{1}{2} k_B T$,

(1.4)

with $U_x$ an electron energy associated with the motion in the x-direction, and $k_B$ Boltzmann’s constant. Combining Eq. 1.3 and Eq. 1.4,

$\epsilon_x = \sigma_{x,i} \sqrt{\frac{k_B T}{m_e c^2}}$.

(1.5)
we see that source size and source temperature are the crucial parameters when defining beam quality.

1.2.3 Coherence length

When applying a beam for diffraction experiments, it is more useful to discuss beam quality in terms of a parameter called (transverse) coherence length, defined in an electron bunch as [17]

\[ L_{\perp} \equiv \frac{\hbar}{m_e c \epsilon_x}, \]  

(1.6)

where \( \hbar \) is Dirac’s constant. The transverse coherence length is a parameter with quantum mechanical explanations and implications. In quantum mechanics, it is related to the transverse spatial extent of a particle’s wave function. In electron diffraction it basically describes to what extent an electron ‘sees’ the potential landscape of the crystal from which it undergoes diffraction. Without delving too deep into quantum mechanical jargon, the wave function of an electron with a high transverse coherence length passes through several lattice distances of a crystal simultaneously, and interferes with itself to form a diffraction pattern. This quantum mechanical description of coherence length is not necessary for understanding and analyzing electron diffraction in this work, but is a useful analogy to understand the following crucial point: in order for diffraction to take place, the coherence length of a particle beam \( L_{\perp} \) should be larger than the lattice spacing \( a \) of the examined crystal.

The coherence length \( L_{\perp} \) is not a conserved quantity, but depends on the size of the beam \( \sigma_x \). Because of this, the coherence length could be made arbitrarily large by increasing the size of the beam. However, in order to avoid wasting electrons in diffraction experiments, a beam should pass in its entirety through the sample under investigation. This requires that the beam size at the sample \( \sigma_x = \sigma_{\text{smpl}} \) is no larger than the size of the sample \( \sigma_{\text{smpl}} \leq s \). In diffraction experiments, the coherence length should be made no larger than

\[ L_{\perp, \text{max}} = \frac{\hbar}{m_e c \epsilon_x} \frac{s}{\sigma_{\text{src}}} = C_{\perp} s. \]  

(1.7)

Here we have introduced another quantity, \( C_{\perp} = L_{\perp}/\sigma_x \), the relative transverse coherence length. The value of this parameter is conserved in a beam if emittance is conserved, and can thus be calculated using source parameters. It is a useful parameter because it describes the required quality of the beam in terms of the sample properties, namely \( C_{\perp} \geq a/s \).

The coherence length in diffraction experiments depends on the source quality (beam emittance, \( \epsilon_x \)) and the size of the sample. Rewriting Eq. 1.7 in terms of source properties (effective transverse source temperature \( T \) and initial spot size \( \sigma_{\text{src}} \)) results in

\[ L_{\perp, \text{max}} = \frac{\hbar}{m_e k_B T} \frac{s}{\sigma_{\text{src}}}. \]  

(1.8)

This implies that the quality of the beam for diffraction is improved by decreasing the source size and temperature. An obvious limitation is that a small source size results in a low charge per pulse. Specific charge requirements are not a major focus of this work, but will be touched on in the discussion, Chapter 6.

1.3 Beam quality in ultrafast electron diffraction

Now that we have defined a method to quantify the beam quality, we focus on the more specific quality requirements of UED.
1.3.1 General
UED typically consists of 4 primary steps: 1) electrons are extracted from a source, 2) are accelerated, steered and focused, 3) pass through a sample and diffract, and finally 4) reach a detector where the diffraction pattern is registered. Beam quality in the extraction phase (1) must be high enough so that the coherence length at the point of the sample, where diffraction occurs (3), is larger than the lattice distance of the crystal under investigation. Electron extraction typically occurs by laser-induced photo-ionization or emission. For UED experiments, it is crucial that the ionizing laser pulse is short, $10^{-13} - 10^{-9}$ s, depending on the required temporal resolution. In molecular dynamics, motions occur at a wide range of time scales, ranging from $10^{-15} - 10^{-12}$ s for molecular vibrations, to $10^{-9} - 10^{-6}$ s for folding processes in proteins [2]. To achieve such short pulses, a femtosecond laser is used.

An example of a set-up used in UED is the 100 kV DC photogun in use by the group CQT [18]. This state-of-the-art device, in combination with a femtosecond laser system, creates 100 kV electron bunches that can be brought below 100 fs bunch length. These bunches have already successfully been used in diffraction experiments on gold and graphite, and are being set up for pump-probe experiments where time resolved structural dynamics of graphite can be investigated.

The 100 kV DC photogun, and in fact any conventional photocathode electron source, is characterized by high source temperatures, typically $T \geq 1000$ K, and up to $T \approx 5000$ K for the photogun at CQT. In Eq. 1.5 we saw that the transverse source temperature plays a part in defining the beam quality, emittance. For many applications, photocathodes have proven to have sufficient coherence to produce (time-resolved) diffraction patterns. For example, the Group of Ultrafast Science in Toronto have ‘filmed’ phonon vibrations in graphite using UED with pump-probe techniques [19].

While this field is still developing, we already look towards the next step: what is necessary for studying motions in crystals with larger lattice distances? For example, as mentioned in the general introduction, can we observe the folding processes of protein crystals, with lattice distances of $a \approx 1 - 5$ nm, and time scales of $10^{-9} - 10^{-6}$ s, using electron diffraction? A larger lattice distance means a larger required coherence length at the sample. Additionally, the synthesis of such samples is often complicated, limiting the size of samples to (sub-)micrometer sizes.

From Eq. 1.8, it can be seen that in order to increase coherence length, the source size and/or the source temperature must be decreased. Since decreasing source size has its own disadvantages and limits, such as low bunch charge or a laser beam’s diffraction limit, we aim to increase beam quality for diffraction experiments by implementing an ultracold source.

1.3.2 Ultracold UED
At the CQT-group a unique new type of electron source has been developed that combines the ultrafast with the ultracold: the ultracold charged particle (UCP) set-up. As a source for ultrafast electron diffraction, a laser-cooled and trapped cloud of rubidium atoms is ionized by nanosecond or femtosecond laser pulses at near-threshold. An accelerator structure then extracts electrons and sends them towards a detector. In previous works it has been shown that the UCP source is capable of producing electron beams with effective transverse temperatures $T$ as low as 14 K. Additionally, a relative coherence length of $C_\perp = 3.6 \pm 0.4 \times 10^{-4}$ has been measured [20], which is sufficient for studying diffraction patterns on $s = 10 \mu m$ crystals with $a \approx 3$ nm.

The laser cooled atoms typically have densities around $\sim 10^{16} \text{ m}^{-3}$, but can theoretically reach $\sim 10^{18} \text{ m}^{-3}$ [21]. If we take an ionization volume of $\sim 10^3 \text{ m}^3$, we could extract $\sim 10^6$ electrons, enough for single-shot diffraction.

Ultrafast electron diffraction therefore shows promise in the study of single-shot diffraction at ps time scales. Upgrading UED by implementing an ultracold source is a strong potential candidate for single-shot ultrafast electron diffraction of more complicated (larger) crystal structures.
1.4 This thesis

In previous research [22, 23] the main properties of ionized beams from ultracold gases and the influence of the accelerator structure was studied. A method to determine source properties, such as temperature and size has been developed and studied in detail [24]. Additionally, the influence of external parameters such as electric field and ionization laser polarization on the source temperature has been studied, of which the results have been published in [17, 25, 26]. The obtained results have been explained by comparison to analytical models [27].

At the start of this research, the very first diffraction images had already been obtained on commercial graphite samples [24, 28].

Previous results have predicted the advanced capabilities of an ultracold source for diffraction experiments, be it for studying complicated structures, or very small samples. The goal of this project is to provide quantitative evidence that the source and beam properties indeed match predictions and meet the requirements for new advances in crystallography. The focus of the research during this project, in its broadest terms, has been to implement, improve and investigate ultrafast electron diffraction from the ultracold source. More specifically, this can be divided into the following two points:

- Improve the ultracold charged particle (UCP) set-up to enable more elaborate diffraction experiments. The main requirements here are to be able to create a stable and controllable beam line. For example, all components must be well enough aligned so that under change of parameters such as magnetic lens strength and electron energy, the beam still passes through the sample.

- Set up experiments to acquire and analyze diffraction images from an ultracold source. With this we intend to investigate the properties of diffraction patterns showing that the ultracold electron beam is not only capable of diffraction, but that its properties reflect its ultracold nature. With this we aim to provide more direct evidence that electron beams from ultracold sources are usable in ultrafast diffraction experiments of macromolecules.

The first of these two points is more technical and has resulted in higher quality results in the second point. The improvements resulting from the first point will be covered in the experimental set-up section, but are not a focus of this thesis.

1.4.1 Chapter overview

The rest of this thesis covers how these goals have been achieved. In Chapter 2 an overview of the experimental set-up of the UCP beam is given. It covers the main components necessary for creating ultracold beams and diffraction images. Chapter 3 introduces the methods used for parametrization of electron diffraction experiments, such as determining source parameters. Chapter 4 covers important theory, specific to the set-up. Among others, the necessary concepts of diffraction are covered. In Chapter 5 the results are presented and analyzed. These results have also been presented in a scientific paper submitted to Nature Communications, which can be found in Appendix A. Finally, the discussions, conclusions and future prospects are given in Chapter 6.
Chapter 2

Experimental Set-up

The ultracold charged particle (UCP) set-up is a state-of-the-art beam source designed for diffraction applications [29]. It is built as a conceptual demonstration of ultracold and ultrafast electron diffraction experiments. With aid from experimental results obtained with this source, a design for a second generation set-up is being conceived, which intends to fully demonstrate pump-probed ultrafast electron diffraction of macromolecules. While this is a future prospect, we aim to use the UCP set-up to provide evidence and further investigate the capabilities and advantages of using an ultracold source in UED.

Figure 2.1 shows a schematic overview of the general stages in ultracold electron diffraction experiments. (a) The source of ultracold electrons is a magneto-optically cooled and trapped cloud of rubidium atoms. (b) Two pulsed laser beams excite and ionize a specific volume within this cloud at near-threshold, creating a local plasma. (c) An electric field extracts electrons and sends them down a beam line, in a bunch. (d) The bunch passes several lens elements, some of which controllable. (e) The bunch is transmitted through a graphite sample, where it undergoes diffraction. (f) Electrons arrive at an MCP detector, which is coupled to a phosphor screen, and are imaged by a CCD camera. In the following sections, these steps are individually covered in more detail.

2.1 Magneto-optical trap

The magneto-optical trap (MOT) is a set-up designed for trapping and cooling atoms [30], in our case, rubidium. MOTs have revolutionized scientific research by allowing scientists to study the behavior of atoms in their lowest quantum state, giving rise to many interesting quantum effects, such as quantum mechanical interference in Bose-Einstein condensates (BECs). Laser-cooling and trapping atoms [31] has won its inventors the 1997 Nobel Prize in physics, while applying MOTs for quantum interference studies of BECs [32] has led its investigators to winning the 2001 Nobel Prize in physics. All in all, MOTs have opened exciting new worlds in the study of quantum properties of atoms, molecules, etc.

Below a brief description of laser cooling and trapping will be given, the principle of which is schematically shown in Figure 2.2. A more in-depth theoretical overview of atomic trapping and cooling can be found in [33].

2.1.1 Laser cooling and trapping

In a MOT, atoms are cooled down below 1 mK by 3 pairs of counter-propagating laser beams, one pair per axis. Cooling, however, is not enough; we want to localize the cooled atoms in a dense volume. This is achieved by trapping. In order to understand these mechanisms, the atomic energy levels, up to the hyperfine levels, must be known.
Atomic energy levels are energy levels coupled to specific quantum states that an electron can occur in, when coupled to an atom. These levels are energy solutions of the Schrödinger equation for an electron in the coulomb potential of a nucleus [34]. In solving this equation, magnetic coupling terms are included. A typical notation for describing an electron’s energy level is $nL_JF$. Here, $n$ is the principal quantum number that represents the orbit level which the electron occupies. It represents the $n$th eigenvalue of the Hamiltonian of the time-independent Schrödinger equation for an electron in a coulomb potential of a charged point source. The number $L$ is the azimuthal quantum number, related to the angular momentum of the electron. This value is limited to $L = 0, 1, \ldots, n - 1$. The azimuthal quantum number is commonly denoted by a letter, where $L = 0, 1, 2, 3, \ldots$ are denoted by $S, P, D, F, \ldots$. Depending on $L$, the electron orbital (or more accurately, its wavefunction) takes on different shapes. Within these states, the projected angular momentum of the particle (typically defined in an arbitrary $z$-direction) can take on values denoted by the quantum numbers $m_z = -L, -L+1, \ldots, L-1, L$. In a first order approximation, the energies of these momenta are degenerate. In reality, effects such as spin-orbit coupling cause these energy levels to be split into a so-called fine structure. The influence of an externally applied magnetic field can further split these states. This property, known as Zeeman splitting, contributes to trapping. The number $J$ represents the sum of the angular momentum and electron spin quantum numbers, $s = \pm 1/2$. Finally, the electron-nucleus spin coupling quantum number $I$ is included in $F$, given by $F = J + I$. This coupling results in hyperfine splitting of energy levels.

The quantification of electron energy levels is taken advantage of in atomic trapping and cooling. The process of cooling and trapping is described below. We limit ourselves to a phenomenological description.
Cooling

Consider a hypothetical atom with only two energy states, denoted by \( |g\rangle \) and \( |e\rangle \), the electron’s ground state and excited state, respectively. The energy difference between these states is given by \( E_{ge} \). This atom is placed in a laser beam with wave vector \( \vec{k} \). Photons in this beam have a momentum of \( \vec{p} = h\vec{k} \), with \( h \) Dirac’s constant. If the atom absorbs a photon from this beam, the momentum is transferred to the atom, giving it a ‘kick’ of magnitude \( h|\vec{k}| \) in the direction of the laser beam’s propagation. Absorption in our atom is only possible if the photon’s energy, \( E_p = h\omega_p \) (\( \omega_p \) is a photon’s angular frequency), matches the transition energy \( E_{ge} \). If the energy is off, the probability of absorption decreases. Absorption results in transferring an electron from \( |g\rangle \) to \( |e\rangle \).

After a typical time \( \tau = 1/\Gamma \), with \( \Gamma \) the natural linewidth of the transition, the electron falls back to \( |g\rangle \) spontaneously, emitting a photon in a random direction. As long as the atom remains in the beam, this process can repeat itself. Since the emission process occurs in a random direction, the net force due to emission is nil. Therefore, there is a total net force in the propagation direction of the beam.

Now suppose the atom has a velocity component along the propagation axis of the beam. The atom ‘sees’ the incoming photons as Doppler shifted: red shifted if the direction of the atom’s motion is the same as that of the beam, and blue shifted if it is moving in an opposite direction.

We now have the ingredients to cool electrons, schematically shown on the left side of Figure 2.2. We take a two-level atom with transition frequency \( \omega_{ge} \), and place it in the path of two counter-propagating laser beams, with an angular frequency \( \omega_p = \omega_{ge} - \delta \) slightly lower than the transition frequency, detuned by \( \delta \). Suppose the atom has a velocity towards the right. It will see the beam coming from the right as blue shifted, and the one coming from the left as red shifted. The right beam’s detuning is thus compensated, while the left beam is further detuned.

An atom moving to the right therefore ‘feels’ a net force towards the left, and vice versa. The atom therefore always undergoes a deceleration, or cooling. For a small detuning and low atom speeds, the net force \( \vec{F} \) on a moving atom is linear with speed \( \vec{v} \), \( \vec{F} = -\alpha\vec{v} \). Here \( \alpha \) is a damping factor, dependent on the natural linewidth, \( \Gamma \), the detuning, \( \delta \), and the laser intensity. Of course, the atom is now only cooled in one dimension; to cool atoms in all three dimensions, 3 orthogonal pairs of laser beams are typically used.

The cyclic process of cooling always includes spontaneous emission, randomly emitting photons. Atoms therefore are never fully stop moving, resulting in a lower theoretical limit in temperature when Doppler cooling, the Doppler limit, \( T_D = \frac{\hbar k}{2\pi c} \).

Trapping

The atoms are now cooled, but not yet spatially confined. To achieve this, magneto-optical trapping is required. Magnetic trapping introduces two components to the set-up: a magnetic field is applied by two coils in the anti-Helmholtz configuration, and each beam of all pairs of laser beams are oppositely circularly polarized. To understand why this results in a trapping force, we need to take a closer look at the atomic energy levels of our hypothetical atom.

Let us assume that the atomic transition that has been taking place during cooling is between an \( S \) (\( L = 0 \)) and a \( P \) (\( L = 1 \)) state, being the ground and excited state respectively. The \( P \) state can be further divided into three degenerate energy levels, due to different angular momentum quantum numbers \( M = -1, 0, 1 \). Under the influence of a magnetic field, these levels split and their energies become distinguishable.

In the anti-Helmholtz configuration, the central point between the coils has 0 T magnetic field. So, at the exact center, atoms do not behave differently than described in the cooling section. By approximation, the field at and around the center has a constant gradient in each direction, \( \frac{\partial B_x}{\partial x} = \frac{\partial B_y}{\partial y} = \frac{\partial B_z}{\partial z} = \text{constant} \). Therefore, if an atom is off-center the energy levels split, linearly with position.

We now look again at the photon-atom interaction. Polarized light intrinsically carries angular momentum of which the projected quantum number is +1 for positively polarized light, \( \sigma^+ \), and −1 for negatively polarized light, \( \sigma^- \). When a photon is absorbed, this must also be transferred
to the excited electron. In the ground state (the $S$ orbital), an electron has an angular momentum quantum number $M = 0$. Therefore, $\sigma^+$ polarized light can only excite an electron to the $L = 1$, $M = 1$ state, while $\sigma^-$ polarized light only excites to the $L = 1$, $M = -1$ state. The MOT is designed so that if an atom is off-center, closer to where the $\sigma^+$ polarized beam is coming from, the $M = 1$ state is shifted down in energy, bringing it closer to resonance with the detuned laser. The $\sigma^-$ beam is consequently further out of resonance with the transition to $M = -1$. The atom is therefore more likely to absorb $\sigma^+$ polarized light, which ‘pushes’ it back to the center. This effect is similar for an offset in any direction from the center; the atom will always be pushed back towards its central position. For small deviations, the net trapping force is proportional to the atom’s displacement from the center, $\vec{F} = -\beta \vec{x}$, where $\beta$ is dependent on the laser detuning, intensity and magnetic field gradient. The process described here is schematically shown on the right side of Figure 2.2.

Conveniently, the detuned lasers used for trapping are also by default cooling lasers. So, combining a non-uniform magnetic field and 3 orthogonal counterpropagating pairs of oppositely circularly polarized laser beams will provide a net force $\vec{F} = -\alpha \vec{v} - \beta \vec{x}$. The atoms are now cooled and trapped.

The above description has been kept rather general to focus on the physical aspects of a MOT. We will now take a closer look at the atom which we trap and cool, rubidium, and focus more on the specific requirements for creating a rubidium MOT.

### 2.1.2 Rubidium

A variety of atoms could be used for optical trapping and cooling. Based on the theory of trapping and cooling given above, it is useful (but not necessarily a requirement) to choose an atom whose outer electron is alone in its ground state in the $S$ orbital, i.e., the alkali metals. Rubidium is a common choice for MOTs. One of the beneficial properties is that it is an alkali metal with a relatively low melting point (312.5 K) making it simple to create a suitable rubidium vapour, without excessive heating.

Rubidium most commonly occurs in two isotopes, $^{87}$Rb and $^{85}$Rb. The latter is cooled and
trapped in our UCP setup (the incompatibility stemming from the hyperfine splitting due to electron-nucleus magnetic moment coupling).

Figure 2.3 shows the relevant part of the electron energy level diagram for $^{85}$Rb (not to scale). The $5S_{1/2}$ and $5P_{3/2}$ states are split into the hyperfine $F = 2, 3$ and $F = 3, 4$ levels (among others), respectively. The trapping laser operates in the optical cycle between the $5S_{1/2}$ and $5P_{3/2}$ states at 780.24 nm. The excitation beam is detuned, by $\delta = -14.5$ MHz. It is possible that an electron leaves this cycle by being excited to the wrong level, $5P_{3/2}$ instead of $5P_{3/4}$. From there the electron can relax to the $5S_{1/2}$ state, from where it cannot normally re-enter the cooling cycle. Therefore the cooling lasers are not sufficient for keeping the cooling cycle running. ‘Lost’ electrons are re-pumped back into the cycle by means of a repump laser, which brings electrons from the $5S_{1/2}$ state back to $5P_{3/2}$, from where it can relax to $5S_{1/2}$. Lost electrons are thus reapplied to the cooling cycle, keeping the MOT from dissipating.

From its ground state, a photon energy of at least 4.18 eV is required to ionize Rb. In practice, this is done in two stages: first an excitation stage, which operates the same transition as the cooling laser, and an ionization stage, requiring at least another 2.58 eV to extract an electron from the $5P_{3/4}$ level, assuming no external electric field is applied. The ionization process is crucial in determining the temperature of the released electrons. More on this is covered in Chapter 4.

2.2 The UCP set-up

The above sections have provided the fundamentals of magneto-optical trapping of $^{85}$Rb. In this section the experimental set-up used at CQT for ultracold charged particle production is discussed.

The MOT is housed within a vacuum vessel, shown in Figure 2.4. The cross section of the vessel is shown in Figure 2.5. The coordinate system in the bottom right of the schematic is used consistently throughout this report. The vessel is kept at $10^{-8}$ mbar by an adjacently placed ion pump. An ion pump locally ionizes gas, and extracts ions using a high electric field. Ions are accelerated away from the vessel and captured, thus removed from the vacuum. The advantage of using an ion pump over a conventional turbo pump is that the former does not have any moving parts, ideal for experiments where mechanical vibrations need to be kept at a minimum. In our set-up, simply clapping nearby one of the lasers can bring it out of its lock on resonance, showing the high need for a vibration-free environment. The ion pump only operates at high-vacuum; several pre-pumps can initially be used to pump the vessel (or parts of the beam line) down from atmospheric pressures.

The 6 red arrows in Figure 2.4 represent the 6 entry points for the cooling and trapping lasers. Four of the beams enter the vessel from the 4 diagonals. The other two enter from the bottom (in the image shown as two arrows; note that these are not in the same plane) and are further reflected internally, as seen in Figure 2.5.
Figure 2.4: Schematic of the vacuum vessel which houses the MOT viewed on the x-y plane (see coordinate system bottom-left). An ion getter pump keeps the vessel at vacuum. Several ports allow visual access and access to MOT lasers. Four laser beams for trapping enter the vessel at the four diagonals, while the other two enter from the bottom, and are reflected to the z-axis internally (see figure 2.5). The ion getter pump and turbo pump are used to bring and keep the vessel at about $10^{-8}$ mbar. The MOT is viewed from above and from the side using CCD cameras.

Figure 2.5: Cross section of the vacuum vessel, in the y-z plane (see coordinate axes at bottom system). The MOT is in the center. Components for creating the MOT and accelerating electrons are visible. Not shown are the 4 laser paths in the x-y plane, which contribute to cooling and trapping. In the top right diagonal, a heated rubidium block provides the vessel with a rubidium background gas.

In practice, there are in fact only 4 laser entry points; two from the bottom, and two on each bottom diagonal. The diagonal beams are allowed to pass through the vessel, and exit through the upper diagonals. The beams then pass through a quarter-wave plate, hit a mirror, and are thus returned back into the vessel, once again passing the quarter-wave plate on the way back. This effectively allows us to use the same laser beam twice. The effect of passing a quarter-wave plate twice results in changing the polarization of the beam from $\sigma^+$ to $\sigma^-$, resulting in two oppositely polarized, counter-propagating beams.

While this set-up simplifies the laser beam path, it has one disadvantage: a returning beam inside the vessel has passed a viewing window of the vessel twice, a quarter-wave plate twice, and is reflected by a mirror, all of which results in losses in laser intensity. The returning beams have marginally less power than the incoming ones. Realizing the importance of beam intensity in the theory of cooling and trapping, it is apparent that this imbalance will cause the MOT to be off-center. In the case of our set-up, the incoming beams come through the bottom two diagonal
entry points, and are reflected back through the top two diagonals, causing the MOT to be pushed upwards. A well-centered MOT is vital to performing reliable and reproducible electron diffraction experiments, so this offset needs to be corrected. This is done by increasing the intensity of the returning beam by decreasing its size. A lens is placed in the beam path before reaching the quarter-wave plate and the mirror. The lens focuses the beam slightly ‘beyond’ the mirror. The returning beam, after passing through the lens again, is slightly converging, having undergone a telescopic effect. The lens is adjustable in position, allowing us to hand-tune the position of the MOT.

Returning to Figure 2.5, the last two laser beams can be seen, cooling and trapping in the z-direction. The two beams enter from the bottom, and are reflected inside the vessel onto the z-axis. All 6 beams cross each other nearly perpendicularly in the center of the vessel, where the MOT is created. The beams have intensities around 10 mW/cm². The two MOT coils, responsible for trapping the Rb, are visible. Each of the two coils are made of hollow copper tubes, with four 100 mm diameter windings. The coils are spaced 100 mm apart from each other. In typical MOT experiments, the water cooled coils are operated in anti-Helmholtz configuration at a steady current of 175 A, producing a quadrupolar magnetic field, with gradients of 10−20 G/cm [23].

A heated rubidium vapor cell (top right in Figure 2.5) creates a rubidium background gas in the vessel. The trap captures rubidium atoms that come close enough and have a sufficiently low velocity. Two CCD cameras view the MOT from two directions (above and along the x axis, as seen in Figure 2.4). From these images, the central MOT position in 3 dimensions can be determined. Additionally, based on the intensity of fluorescence (spontaneous emission), an estimate of MOT particle density can be made.

Also shown in Figure 2.5 is the accelerator structure. The accelerator is axially symmetric. The inner part of the accelerator is attached to a HV feedthrough, and can be set to voltages of up to ± 30 kV. The outer part of the accelerator is grounded. The MOT is placed in the center of the accelerator, where the electric field strength is $F = 37 \text{ kV/m per kV input voltage}, V_{\text{acc}}$. The accelerator can be set to both a positive and negative polarity, allowing us to choose whether ions or electrons are sent down the beam line. Electrons are, of course, used for electron diffraction experiments, while ions can be used for beam alignment (section 2.4) and source size determination (section 3.2.2).

The excitation and ionization laser pulses are injected via the bottom and top of the vessel, respectively. The excitation laser is reflected into the z-axis via an internal mirror (2.5). The excitation and ionization laser beams are thus orthogonal to each other, propagating along the z and y-axis, respectively.

### 2.2.1 Lasers

In the previous sections the following lasers have been introduced:

- Trapping
- Repump
- Excitation
- Ionization

Recalling that the trapping and repump lasers enter the vessel sixfold and fourfold, respectively, we in essence have $6 + 4 + 2 = 12$ optical paths over which we require independent control. The optical paths are thus quite elaborate, but will not be discussed here.

An overview of the lasers used in the UCP set-up is given in table 2.1 [23]. Cooling and trapping is done with a 780 nm diode laser. The frequency is stabilized to the required atomic line transition of rubidium (see section 2.1.2) by using frequency modulation spectroscopy. This technique is based on measuring Doppler-free absorption of rubidium and locking the laser beam to that frequency. The beams are further fine tuned by acousto-optical modulators (AOMs), to introduce detuning. Additionally, the AOMs can be used to switch the beams on and off in
Table 2.1: List of the lasers used in the UCP experiments [35]

<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 FWHM</td>
</tr>
</tbody>
</table>

-Legend with OPerA Solo

microseconds. The same laser also produces the beam for excitation. This signal is typically not detuned with respect to the original rubidium line transition. Another AOM controls the frequency and output (switching) of this beam.

Another 780 nm diode laser is used as repump laser. The frequency of this laser is locked to the trapping laser, with a frequency offset to accommodate the repump transition, $S_{\frac{1}{2},2} \rightarrow 5P_{\frac{3}{2},3}$.

There are two lasers available for ionization, one with a pulse length of several nanoseconds (Quanta-Ray) and another with femtosecond pulses, with an output pulse length of 58 fs. The output wavelengths of both laser systems are tunable, and can be accurately measured with wavemeters. The femtosecond laser is produced by a Ti:Saph system with an output of 800 nm [36]. To be able to tune this beam to the required wavelength, the beam is passed through an optical parametric amplifier (OPA) [37, 38]. The OPA is designed to output a tunable wavelength between 472 nm up to 2.6 μm. This broad range is achievable by altering the optical paths in the OPA. In its current set-up, however, the output is tunable between 472 and 533 nm by changing the angle of a BBO crystal in the OPA. The optical path of the femtosecond laser pulse covers more than 10 meters before reaching the MOT. Some beam lengthening is expected during its journey, resulting in a pulse length of over 100 fs when it reaches the MOT.

Before beginning the ionization process, the trapping and repump lasers are first temporarily shut off to allow the rubidium atoms to fall back to their ground state. Due to the extremely low temperature of the MOT, the position and velocity distribution changes negligibly during this short μs off-time. Meanwhile, the excitation and ionization lasers enter the MOT. The excitation and ionization lasers overlap orthogonally (propagating along the z and y-axis respectively), creating a well defined volume of ionized rubidium.

2.3 Photo-ionization - source temperature

An important parameter of the ultracold charged particle beam is its initial effective transverse temperature, defined in Chapter 1 as $T = \frac{\sigma_{p,i}^2}{m_e k_B}$, with $\sigma_{p,i}$ the transverse rms momentum, $m_e$ the electron mass, and $k_B$ Boltzmann’s constant. Engelen [27, 24] has developed a model which predicts the source temperature as function of ionization laser wavelength and acceleration field. This model is partially based on the work done by Bordas $et$ $al.$ [39, 40], which describes the equations of motion of electrons in Stark-shifted Coulomb potentials. The validity of Engelen’s model has been investigated by Bakker [28], and is not a focus of this research. However, a brief overview of the important aspects of the model are described in this section.

In the two-stage ionization process, the ionization laser frees an electron from rubidium’s $5P_{\frac{3}{2},4}$ state. In the absence of an external electric field, this requires at least $E_i = 2.58$ eV of energy. If photons in the ionization beam have a higher energy $E_i$ than required for ionization, ionized electrons will inherit part of the excess energy, $E_{exc} = E_i - E_i$ in the form of kinetic energy. Applying a field, as is the case in our set-up, changes the potential landscape of the rubidium atom, causing the required ionization energy to decrease, as is schematically shown in Figure 2.6 for several field strengths, $F$. The dashed line, the field-free Coulomb potential, is shifted linearly by the field, causing the minimal required electron escape energy to decrease. This shift, the Stark
Figure 2.6: Plot of the energy of an electron in a rubidium ion potential, under the influence of various external electric field strengths in the $z$ direction. The black dashed line, the field-free potential, is shifted linearly by an external field (colored dashed lines), resulting in the colored potential lines. An electron trapped in a Stark-shifted Coulomb potential requires less energy to escape to a positive $z$, compared to a regular Coulomb potential. [37]

shift, is given by:

$$E_{\text{Stark}} = 4Ry \sqrt{\frac{F}{F_0}},$$

with $Ry = 13.6$ eV the Rydberg constant, and $F_0 = 5.14 \times 10^{11}$ V/m the atomic unit of electric field strength. The excess energy thus becomes:

$$E_{\text{exc}} = E_l - E_i + E_{\text{Stark}} = 2\pi\hbar \left( \frac{1}{\lambda_l} - \frac{1}{\lambda_i} \right) + 4Ry \sqrt{\frac{F}{F_0}},$$

with $\lambda_l$ the ionization laser wavelength, and $\lambda_i = 479.06$ nm rubidium’s field free threshold ionization wavelength (see Figure 2.3). As mentioned, most of the excess energy ends up as kinetic energy of the electrons, though this does not provide us with any information of how the energy is spatially distributed. In fact, it has been found experimentally that the measured temperature was much lower than the expected temperature based on the electrons’ excess energy. In order to understand this, a closer look at the electron’s trajectory after ionization is made, using the Bordas model.

Without delving into the mathematics of it, the fundamentals of the Bordas model are simple to understand. The model makes the following assumptions: electrons, upon ionization, radially expand in a random direction from a central point with a velocity determined by $E_{\text{exc}}$. The potential landscape an electron finds itself in is determined by the Coulomb force and the electric field (applied in the $z$-direction). A saddle point thus is formed at a certain distance from the center in the $z$-direction, in Figure 2.6 at the local maximum of the Stark-shifted potential in the positive $z$ direction. Regardless of the starting direction, if an electron is to escape, it must pass through this saddle point, a sort of gully from the electrons point of view. That is not to say that all starting angles result in an escaped electron; in fact the model also predicts which maximum angle results in an escaped electron, dependent on the ionization energy $E_l - E_i$ and the applied field $F$. The saddle point acts as a collimator or a funnel for escaped electrons, restricting the allowed transverse spread. The potential landscape thus suppresses the transverse temperature of released electrons.

Up until now, the assumption that all electrons gain one discrete value of excess energy has been made. For a narrow-bandwidth laser such as the ns laser, this is an accurate approximation. However, the broadband nature of the fs laser results in a broader range in excess energies. To find the resulting transverse temperature, one must integrate over all these energies, weighted by the photons’ spectral distribution function. Even if the mean energy of photons in a fs pulse is lower than the required ionization energy, the tail of the energy spectrum can still cause ionization to occur. The model thus also distinguishes between a narrow or broad band spectrum.
2.4 Ultracold charged particle beam

The previous section has described the necessary ingredients for creating an ultracold charged particle beam. In this section, the electron beam will be described in more detail, focusing on the experimental set-up for controlling and analyzing the beam.

2.4.1 Beam source

As mentioned in Chapter 1, the two main source parameters that influence the beam’s quality are the initial source temperature $T$ and the source size $\sigma_{i,(x,y)}$. The electron temperature after ionization is a function of the ionization laser wavelength $\lambda_i$ and the applied electric field $F$. This relation was covered in 2.3. The source size is governed by the rms sizes of the excitation and ionization lasers, $\sigma_{\text{ion}}$ and $\sigma_{\text{exc}}$, which propagate along the z and x-axis respectively. The source volume is thus given by [24]

$$\{\sigma_x, \sigma_y, \sigma_z\} = \{1/\sqrt{\sigma_{\text{ion}}^2 + \sigma_{\text{exc}}^2}, \sigma_{\text{exc}}, \sigma_{\text{ion}}\}. \quad (2.3)$$

The longitudinal beam quality is generally of importance in UED experiments because it defines the possible temporal resolution. Since this is not a focus in this research, we will only concern ourselves with the transverse source sizes, $\sigma_{x,y}$.

Combining all the aspects of the previous section, a charged particle beam can be created. The accelerator structure, with potential $V_{\text{acc}}$, accelerates electrons to an energy $U_e = 0.47efV_{\text{acc}}$, with $e$ the elementary charge and $f \approx 1$ a correction factor in the case that the MOT is not entirely centered within the accelerator.

2.4.2 Beam path

The electrons in the beam line cover a total of 1.53 meters from the center of the MOT until the detector, passing a series of elements in the beam line. An overview of these elements and positions is given in table 2.2, and shown schematically in Figure 2.1. Some of these elements in the beam line exert forces on electrons within the beam, typically linear with radial position of the electron. They can be modelled as lens elements, similar to those working on light beams. Each element therefore has a corresponding focal length, possibly depending on the electrons’ longitudinal energy. The focal lengths of the lenses in the beam line are also shown in Table 2.2. These focal lengths have been determined using particle tracking simulations, GPT [41]. GPT is based on full 3D particle tracking techniques that can follow individual charged particles through a variety of electrical and magnetic elements. In theory, the lenses do not affect the total emittance of the beam.

Electrons are accelerated in an axially symmetric field. However, the field is radially diverging at the exit of the acceleration structure. Because of this, the exit acts as a negative lens for the electrons, with focal length $-0.033$ m [24]. This gives accelerated electrons a so called exit kick. The focal length of the exit kick is independent of the acceleration potential, or of the type of particle.

After having left the accelerator structure, electrons pass one of the MOT coils used for trapping rubidium. This coil carries a current of 175 A and, needless to say, produces a non-negligible field. This field is conveniently axially symmetric so that the forces in the x and y-axes are identical. However, the field, being that of a solenoid lens, causes a rotation of the beam around its central axis. The MOT coils act as a positive lens, proportional to the beam energy $U_e$.

A rather inconvenient focusing element in the set-up is that of an unwarranted magnetic field, which is by speculation attributed to a small mirror used for directing the z-axis trapping and repump laser beams, seen on the right side of Figure 2.5. This mirror might be magnetic. The dipolar term of this field can alter the direction of the beam, but otherwise leaves the beam properties unchanged. The quadrupole term of this field, however, has a clear effect on measurements:
Table 2.2: List of the focusing elements that influence the size of the electron beam. The position and the focal length are listed for every element. The values of $U$ are in eV, and the values of $I$ are in A. (partially taken from [28])

<table>
<thead>
<tr>
<th>Element</th>
<th>Position (m)</th>
<th>Focal length (m)</th>
</tr>
</thead>
<tbody>
<tr>
<td>MOT center</td>
<td>0</td>
<td></td>
</tr>
<tr>
<td>exit kick</td>
<td>0.010</td>
<td>$-0.033$</td>
</tr>
<tr>
<td>MOT coils</td>
<td>0.057</td>
<td>$0.019 + 4.8 \times 10^{-5} \cdot U$</td>
</tr>
<tr>
<td>quadrupole $(x)$</td>
<td>0.228</td>
<td>$+0.009 \cdot U$</td>
</tr>
<tr>
<td>quadrupole $(y)$</td>
<td>0.228</td>
<td>$-0.009 \cdot U$</td>
</tr>
<tr>
<td>alignment plate 1</td>
<td>0.69</td>
<td></td>
</tr>
<tr>
<td>solenoid lens 1</td>
<td>0.56</td>
<td>$(1.6 + 5.8 \cdot 10^{-6} \cdot U) \cdot I^{-2}$</td>
</tr>
<tr>
<td>solenoid lens 2</td>
<td>1.12</td>
<td>$(2.74 \cdot 10^{-11} \cdot U^2 + 1.221 \cdot 10^{-5} \cdot U) \cdot I^{-2}$</td>
</tr>
<tr>
<td>graphite sample (plate 2)</td>
<td>1.245</td>
<td></td>
</tr>
<tr>
<td>beam block</td>
<td>1.415</td>
<td></td>
</tr>
<tr>
<td>MCP detector</td>
<td>1.53</td>
<td></td>
</tr>
</tbody>
</table>

A quadrupole lens focuses in one transverse direction, while defocusing in the other. The focusing strength and effective position of this lens seem to be quite dependent on experimental parameters. The lens parameters of this quadrupole, have been characterized on several occasions [28, 42], of which the most recent is presented in Table 2.2. When including this lens in fitting models (see Chapter 3.2.3) the lens strength and position are included as fitting parameters.

Two perpendicular sets of steering coils are placed right outside the MOT vessel, about 32 cm into the beam line. These coils create a dipolar field which simply alters the path of the electron bunch, without changing internal properties. These coils can be used for fine tuning the beam path, making sure it travels straight down the beam pipe.

Two magnetic solenoid lenses are placed at 0.56 and 1.12 m into the beam line. The first lens has inner radius $\rho_i = 5.8$ cm, outer radius $\rho_o = 6.2$ cm, length $L = 5$ cm, and 200 windings. The second lens has $\rho_i = 2.9$ cm, $\rho_o = 4.6$ cm, $L = 4.2$ cm, and 600 windings. Solenoid lenses have variable focal lengths, depending on the current they carry [43]. The two-lens system provides high control over the beam path, allowing the user to choose the position of the beam waist. In fact, the second solenoid lens is a more recent inclusion to create a higher degree of control over the beam’s transverse momentum spread and size. How these lenses are used to control the beam for diffraction experiments is covered in section 4.1.2. The magnetic lenses are mounted on translation stages, allowing them to be moved independently in the x and y-direction. This conveniently allows us to center the lenses around the electron beam, instead of having to adjust the beam itself. A side effect of solenoid lenses is that in addition to focusing a beam, they introduce a rotation of the beam around its central axis.

Introduced in this research are two metallic plates, placed at 0.69 and 1.245 m into the beam line. These plates are used in alignment of the beam. This has become a necessity in our experiments, because little was known about the path of the electrons. In fact, even the exact source position was poorly defined; the only reliable information about the beam was where it landed on the detector. The plates, schematically shown in Figure 2.7, consist of several holes, in a crosshair pattern. The first plate is attached to a rotatable arm, with the plate off-center from the rotational axis. The plate can thus be rotated in and out of the beam line. When in the beam line, the crosshair is centered within the beam pipe. The plate is used to ensure that particles are extracted from the radial center of the accelerator structure. During alignment, ions are used instead of electrons, since ions are negligibly affected by magnetic elements. By changing the positions of the excitation and ionization laser beams, the ionization volume can be moved until the ions pass through the plate centrally. The second plate is mounted on a translator, which can vertically move the plate in and out of the beam line, in steps of 0.25 $\mu$m. The second plate ensures that the beam pipe itself, which has a rotation pivot point at 0.76 m, is in a straight line with the beam. Again, ions are used for alignment. When these aspects are aligned, the plates
Figure 2.7: Schematic of the alignment plates in the beam line, and alignment images using ions. The first plate has hole-pattern crosshair. The second plate a larger pattern, as the beam is typically larger at this point in the beam line. The second plate also holds the diffraction sample and contains spot size determination edges. The images are taken during the alignment process, using rubidium ions. All values are in mm.

can be used again to control the beam path of electrons. The second plate also contains a sample holder, and sharp slanted edges for spot size determination.

A graphite sample (on a TEM grid) is placed in the sample holder of the second plate. Using the magnetic lenses to transmit the beam through the sample, electrons in the beam are diffracted (see section 4.2). The undiffracted beam is typically still much more intense than the diffracted beamlets, so for good contrast the central beam is blocked by an 8 mm diameter, circular beam block on a thin stem, which can be translated horizontally into the beam line.

After travelling a distance of 1.53 m, the beam reaches its final destination, the microchannel plate (MCP) detector [44]. The MCP detector amplifies the signal of incoming electrons or ions by means of an internal cascade of secondary electrons, which are accelerated towards a phosphor screen. A single incident particle can induce up to $10^4 - 10^6$ secondary electron at the output. The phosphor screen lights up when electrons are incident upon it, which is captured by a CCD camera. A spatially resolved image of where the beam has arrived can thus be acquired.

An electron cascade broadens out as it passes through the MCP. A single electron is detected on the phosphor screen with a size of 95 $\mu$m [23]. This is a limiting factor in detector resolution. The CCD camera images the phosphor screen with a maximum resolution of $2.07 \times 10^3$ pixels/mm$^2$, meaning each pixel represents an area of $22^2 \mu$m$^2$ on the phosphor screen. For speed and image size considerations, however, images can be binned so that an average over a set amount of pixels is taken. A binning of 5, meaning $5 \times 5$ pixels are combined into 1, is used throughout this work. Images are thus captured with a resolution of 110 $\mu$m.

The output current of the MCP is sent to an oscilloscope, where the relative charge of the bunch is detected. The detected current is dependent on the MCP voltage settings, and has not been absolutely calibrated, but is useful for measuring relative charge.
Chapter 3

Methods

In Chapter 2, an overview of the set-up that we have to our disposal for ultracold electron diffraction is given. In this chapter, the methods for measurements using our set-up are described.

3.1 Beam alignment

In most of our experiments a well-aligned beam is extremely important. This is especially true for diffraction experiments, where it is crucial that the central position of the beam at the sample and at the detector are not dependent on changes in parameters such as source temperature and magnetic lens currents.

It has been discussed in the previous section how the first alignment plate in the beam line is used to ensure particles are extracted from the radial center of the accelerator structure. Additionally, the second alignment plate is used to adjust the beam pipe itself from a pivot point midway the beam line. Both of these alignment procedures make use of an ion beam, as it is negligibly affected by magnetic fields.

Now we switch to an electron beam. First off, it is noticeable that electrons will not appear on the same point on the detector as the ions. Two main contributors to this offset are the Earth’s magnetic field and the unwanted magnetic quadrupole element in the beam line. The Earth’s magnetic field is mostly corrected for by current-carrying wires placed parallel to the beam line, though inaccuracies in this correction might still have some influence on the electron’s path. Secondly, the dipole term of the unwanted magnetic element alters the electrons’ path, but its exact influence is not yet entirely understood. As a result, the path of an electron beam is different than that of ions. Extra steering coils are installed near the source of the beam to adjust the path. The current through these coils is adjusted until the beam once again passes centrally through the second alignment plate. The electron beam spot will still not necessarily end up on the same location on the detector as the ions, but this is not a crucial difference. The second alignment plate also holds samples for diffraction, thus it is most important that electrons are allowed to pass centrally through it. The plate is attached to a vertical translator; the sample can thus be shifted into the beam line at exactly the place of the center of the crosshair, seen in Figure 2.7.

The magnetic lenses, which are placed on translation stages (mobile in the $x$ and $y$ direction), need to be adjusted so that these are centered around the electron beam. Simply put, the position of the center of the electron spot on the detector should remain unchanged under the influence of magnetic focusing (though obviously the size is expected to change). In other words, the center of the beam should follow the radial center of the solenoid. This then, warrants a method of alignment: ensuring that the position of the electron spot remains unchanged for varying magnetic field strengths. The physical position of the magnetic lens is determined by four degrees of freedom: two translation, and two tilt. This means that for any given coil current, two-dimensional alignment of the electron beam is overdetermined. It is therefore necessary to scan
through various coil currents, methodologically adjusting the tilt and translation of the coil. It has proven useful to choose one current, adjust the beam, and then repeat for the same negative current. After a few iterations of switching back and forth between the same positive and negative current, a higher current is chosen, and the process is repeated. This alignment is done for both lenses separately, where in both cases the lens not being aligned is inactive.

3.2 Beam parameters

To understand and to be able to analyze our measurements, we need a good understanding of the beam’s dynamics. Having an appropriate model of the beam is, for example, important for determining the source parameters, the effective transverse source temperature and source size. There are two main models used here for analyzing the beam: an analytic ray tracing model and particle tracking simulations using the General Particle Tracker (GPT) software [41].

We will introduce this section by describing how the beam line is modelled. These models are then used in analyses of source measurements, described further in this chapter.

3.2.1 Analytical beam model

As described in the Introduction, the beam size and angular spread are essential components for describing a beam. The value of both for a specific set-up can be modelled analytically for any point along a beam line, given that the source parameters, the size and angular spread (i.e. temperature) are known. It is assumed that the electrons in the beam have a Gaussian distribution in phase space (position and angle with respect to the z-axis in the two transverse dimensions), so that we can define the size and angular spread of the beam as the root-mean-squared of the ensemble, $\sigma_{x,y}$ and $\sigma_{\theta}$.

Let us first look at a single electron in the beam. It is convenient to describe the phase space parameters of the electron in a vector for each dimension:

$$\vec{x} = \begin{pmatrix} x \\ \theta \end{pmatrix},$$

with $x$ the position, and $\theta \approx \frac{p_x}{p_z}$ the particle’s directional angle with respect to the z-axis. Given a specific phase space vector $\vec{p}$ at a certain position in the beam line, further development of the electron in the beam can be modelled using a matrix formalism [45], where a set of $2 \times 2$ matrices describe the evolution of the electrons’ phase space as it travels down the beam line. These matrices represent transformation operations of the phase space. We can model the position and angle of an electron at any point down the beam by using a set of these matrices acting on the known initial beam parameters, $\vec{x}$.

Two operations occur in the beam path that require a transformation matrix: the beam propagates freely down the beam line, and the path of the beam is changed by a lens element (see table 2.2). Beam drift is described by the matrix element

$$M_d = \begin{pmatrix} 1 & d \\ 0 & 1 \end{pmatrix} \tag{3.1}$$

where $d$ is the length of the drift space. When applying this matrix operation to a phase space vector $(M_d \vec{x})$ we see that the angular direction remains unchanged, but that the electron approaches or moves away from the central axis according to its angular direction. At a lens, which we will assume is thin, the matrix operation is given by

$$M_f = \begin{pmatrix} 1 & 0 \\ -\frac{1}{f} & 1 \end{pmatrix} \tag{3.2}$$
where \( f \) is the focus distance of the lens, as given in table 2.2. Understandably, the matrix operation does not influence the position of the electron, but alters its angular direction, depending on its radial position at the lens.

Along the length of the beam path, electrons pass several drift spaces and lenses, one after another, seen in Figure 2.1(d-f). The action of each separate operation \( M_n \) accumulates, and provides final positions and angles at the detector as a function of initial parameters, with a single \( 2 \times 2 \) matrix, \( M_{\text{tot}} \).

\[
\bar{x} = (M_N(M_{N-1}(\ldots(M_2(M_1(x_i))))) = M_{\text{tot}}x_i. \quad (3.3)
\]

In our set-up, drift lengths \( d_n \) and lens focus lengths \( f_n \) as function of lens current \( I \) and/or electron energy \( U_e \) are known, and given in Table 2.2. We thus have a model that predicts the final conditions at the detector \( \sigma_{\text{det}} \) as a function of source parameters and lens parameters. In terms of distributions of the entire electron ensemble, the final (rms) beam spot size is then given by:

\[
\sigma_{\text{det}}^2 = M_{11}^2 \sigma_{x,i}^2 + M_{12}^2 \sigma_{\theta,x,i}^2 = f(\sigma_{x,i},\sigma_{\theta,x,i},U_e,I_1,I_2) \quad (3.4)
\]

where \( I_1 \) and \( I_2 \) are the currents through the magnetic lenses. \( M_{ij} \) are elements of \( M_{\text{tot}} \), where \( i \) is the column number and \( j \) the row number.

The final spot size can also be written in terms of source temperature, using Eq. 1.4: \( \sigma_{\theta,e,i} = \sigma_{p,e}/p_z = \sqrt{k_BT/2U_e} \).

\[
\sigma_{\text{det}}^2 = M_{11}^2 \sigma_{x,i}^2 + M_{12}^2 \frac{k_BT}{2U_e} \quad (3.5)
\]

and analogously for the y direction. We see here, again, the importance of the source size and temperature in beam dynamics.

These models, together with simulation data, are used to determine source parameters, the effective transverse source temperature and source size, described in the following sections. Once the source size and temperature are known, analytic and simulated models of the beam path, using these source parameters, can be used to predict experimental results.

Note that this model has made some simplifications and assumptions:

- Non-conservative effects such as expansion due to space charge are not included in this model. Unless mentioned otherwise, space charge has negligible effect on beam properties in our experiments, so can be ignored in the analyses.

- We assume the lenses can be modelled as thin lenses, meaning that the (de-)focusing effect of the lens is immediate at a fixed plane. While this may be a simplification for thicker lenses such as the solenoids, the focal lengths are based on realistic GPT simulations which incorporate the width of the lens. We can thus confidently say that the presented focal lengths for the model thin lens are an accurate representation of the actual lens action.

- Solenoid lenses introduce a rotation in the beam. Formally, a \( 4 \times 4 \) matrix acting on \( \bar{x}_{4D} = (\sigma_z^2, \sigma_{\theta,z}^2, \sigma_y^2, \sigma_{\theta,y}^2)^T \) should be used, where the magnetic lens matrices include off-diagonal terms to ‘mix’ \( x \) and \( y \) properties. It is therefore more convenient to keep track of the long and short axes of the ellipsoidal beam. The properties of these axes remain unchanged under rotation, thus can be separately analyzed with \( 2 \times 2 \) matrices.

### 3.2.2 Source size - Space charge scan

One of the two primary source parameters is the size of the ionization volume \( \sigma_{i,x} \) and \( \sigma_{i,y} \). This is determined by a space charge scan, which will be described below.

In space charge scans, ions are used instead electrons (simply by reversing the polarity of the acceleration structure) for two reasons:
• Nearly all of the excess ionization energy ends up in the electrons, leaving the ions very cold. The angular spread due to temperature is thus negligible in ions.

• Ions are practically unaffected by the magnetic fields present along the beam line. This considerably simplifies analysis of this scan, as we are effectively only dealing with two defocusing actions: the exit kick and space charge expansion.

The charge of the ions extracted from the ionization volume is given by
\[ Q = eN f_{ex} P \] [24]. Here, \( N = \sqrt{\frac{2\pi^3}{\sigma_x,i\sigma_y,i\sigma_z,i\eta_{MOT}}} \) is the number of atoms in the ionization volume, with MOT atom density \( \eta_{MOT} \). The excited state fraction \( f_{ex} \) and the ionization probability per excited atom \( P \), are determined partially by the intensity of the excitation and ionization lasers, respectively. The intensity of the ionization laser beam is easily varied by implementing a neutral-density (ND) filter. ND filters decrease the intensity of the beam by a factor \( 10^{\text{ND}} \), evenly in the frequency spectrum.

A freshly ionized and extracted volume of charged particles will undergo a Coulomb explosion; space charge causes the particles to expand away from each other. If the charge density of the initial volume is low enough, this effect is negligible. However, if the ionization laser is intentionally intensified by lowering the value of the ND-filter, a noticeable expansion occurs.

In a space charge scan, we scan through beam charge by varying the ND-filter value of the ionization laser beam. The charge in the bunch is linearly related to the intensity of the laser beam. So, for example, reducing the ND filter from ND = 2 to ND = 1 results in an increase in charge by a factor of 10. The final spot size at the detector, a function of initial spot size and bunch charge, is monitored for the different bunch charges. The results of these scans are compared to similar scans performed with GPT simulations, with varying initial source sizes (in two dimensions). The experimental source size is then determined by finding the set of simulation parameters that best matches the experimental data, in terms of the lowest rms residual.

An example of an ion source scan is given in Figure 3.1. Here, the ion spot size is given in two dimensions at the detector as function of laser ND filter. A higher ND filter results in a weaker laser pulse, thus a lower charge, and finally less space charge expansion. We compare these data to simulated data for various initial sizes. The rms residuals between simulation and experimental data is shown in the inset. The spot size is determined as the simulated initial size for which the residual is minimal.

Note that this method is different from that presented in earlier work [24, 37, 28]. Previously, the data from the experimental space charge scan was fitted to an analytical beam model that models the expansion due to space charge as a defocusing lens. We have found that this model, while sufficiently accurate for many scans, is incomplete, and requires further investigation. More on this note can be found in Appendix B.

3.2.3 Effective transverse temperature - Waist scans

In this section the method for determining the effective source temperature, \( T \), is introduced. In Eqs. 3.4 and 3.5 we have shown that the spot size at the detector is a known function of several parameters:
\[ \sigma_{\text{det},(1,2)} = f(\sigma_{x,i}, \sigma_{y,i}, T, U_e, I_1, I_2) \]. In the previous section, the method of finding the source size \( \sigma_{(x,y),i} \) has been described. So, for known values of \( \sigma_{x,i}, U_e, I_1 \) and \( I_2 \) and a measurable value of \( \sigma_{\text{det}} \), it is possible to derive \( T \).

In practice a single measurement is not enough to accurately determine the source temperature \( T \), considering measurement uncertainties, and for example the unpredictable effects of the magnetic quadruple, discussed in section 2.4.2. For this reason, a much more reliable technique is to measure the spot sizes for varying beam parameters, under the same source conditions. Looking back at Eq. 3.5, an evidently obvious choice is to vary the currents of the magnetic focusing lenses. In practice, only the first lens is used in such experiments. By scanning through increasing lens currents, the waist of the beam is moved from an effective position behind the detector to somewhere in front of the detector. As we are placing the waist of the beam at different positions this type of measurement is referred to as a waist scan.
Figure 3.1: Example of a source size scan. The ion spot size at the detector is shown for two dimensions. The ND filter of the laser is related to space charge expansion: a higher ND value results in a lower bunch charge. A high ND filter value therefore results in minimal expansion. The experimental data is compared to space charge scans from GPT simulations. The residual between the experimental and simulation data is shown in the inset for varying simulation source sizes. The source size is determined by finding the set with the lowest rms residual, in this case at $54 \times 32 \mu m^2$.

The measured spot size $\sigma_{\text{det}}$ is limited by the resolution of the detector $\sigma_{\text{res}}$. In most cases this does not present a problem, since spot sizes are typically in the order of millimeters ($\sigma_{\text{det}} > 110 \mu m$). However, when the waist of the beam is placed at the sample, this limit is reached, and the final measured size is different from the actual beam size, $\sigma_{\text{real}}$. The measured size (and so also the beam line model) is therefore adjusted by:

$$\sigma_{\text{det}} = \sqrt{\sigma_{\text{res}}^2 + \sigma_{\text{real}}^2}$$  \hspace{1cm} (3.6)

The final spot size at the detector is measured using a two-dimensional Gaussian fit of the spot’s intensity profile, resulting in the Gaussian standard deviation for the long and short axes $\sigma_1$ and $\sigma_2$ of the elliptical spot. Again, we do not look at the spot size along the x and y-axes because of rotation of the beam caused by the magnetic lenses. The resulting data, the spot size on the detector as function of magnetic lens current, is fitted against the analytical beam model (Eq. 3.5), with temperature as fitting parameter. Also included as fitting parameters are the position $d_{\text{quad}}$ and strength $k_{\text{quad}}$ of the quadrupole. Finally, a (subtle) magnetic lens correction factor $C_{\text{sol}}$ is included, which ensures that the waist of the theoretical curve is at the same position as the data, correcting for inaccuracies in lens strength or beam energy. Included in the fit model is the source size, which is determined using an ion space charge scan (section 3.2.2).

Figure 3.2 shows examples of waist scans. In Fig. 3.2(a) some analytical examples are given for various source temperatures. Here it is clear that there is a unique fit for each temperature. A higher temperature results in a consistently larger spot size. In Fig. 3.2(b) an example of a measured data set is plotted, with a best-fit curve. The two data sets represent the rms size of the short and long axis of the detected spot. Note that (a) and (b) are taken data sets with different initial conditions, so do not overlap. In (a), for convenience, the size of only the long axis is shown. The minimum in the curve, around 1.15 A, is the current when the beam waist is at the detector. In (b) it is clear that the minima of the short and long axes curves do not occur at the same current. Astigmatism here is caused by the magnetic quadrupole in the set-up (see Table 2.2). This is included in the fit model.
Figure 3.2: Examples of waist scans, showing the beam size at the detector as function of magnetic lens current, \( I_1 \) and \( I_2 \). (a) According to the analytical beam line model, higher source temperatures result in consistently higher beam sizes. For convenience, the size of only one of the axes of the spot is shown. (b) An example of the waist scan model fitted to experimental data is shown. The beam size at the detector is determined in two dimensions, for the long and short axis of the elliptical spot. The waist of the short and long axes do not coincide due to the magnetic quadrupole in the beam.

3.2.4 Beam energy - Time-of-flight scan

The electron beam has an average longitudinal energy of \( U_e = 0.47 f e V_{\text{acc}} \), with \( V_{\text{acc}} \) the potential over the accelerator, \( e \) the elementary charge, and \( f \approx 1 \) a correction factor for the case that the ions are not extracted from the exact center of the accelerator structure. While the latter deviates minimally from unity, it is not a negligible term. Therefore it is useful to have a method of measuring the beam energy. A conventional method of doing this is a time-of-flight (TOF) scan. In such a scan, the arrival time of electrons relative to ionization time is measured for varying acceleration potentials, \( V_{\text{acc}} \). The scope signal of the charge on the MCP is measured as the arrival time (section 2.4.2). The resulting data is fitted to the model,

\[
\begin{align*}
    t_{\text{TOF}} &= \frac{d_{\text{det}}}{c \sqrt{1 - \left(\frac{m_e c^2}{m_e c^2 + m_i c^2}\right)^2}} + t_0, \\
    \text{with } d_{\text{det}} &= 1.53 \text{ m the distance from the center of the MOT to the detector. The factor } t_0 \text{ is a constant factor stemming from a delay in the electronic signal from the detection of the ionization pulse and electron pulse.}
\end{align*}
\]
Chapter 4

Setting up for diffraction

This chapter covers the theoretical groundwork of the diffraction experiments presented in this work. The following topics will be covered:

- In section 4.1, a quantitative description of the double lens configuration for diffraction is given; we focus on the controllability of the beam waist and angular spread, and expected output at the detector. Theoretical dependence of beam parameters due to temperature in this set-up is then evaluated.

- In section 4.2 an overview of diffraction theory is presented, focusing on expected diffraction patterns from our beam through graphite.

4.1 Beam line parameters

We have several methods of analyzing a beam at our disposal: particle tracking simulations (GPT), an analytical beam model (with lens parameters determined by GPT, given in 2.4.2), and physical measurements. Using the former two, a feasibility study of diffraction experiments can be made before starting the hands-on laboratory work. Recapping the goal of this research, we wish to demonstrate the beam quality directly from diffraction patterns, providing evidence that the low temperature properties of the source are a vital new step for advances in UED. A proposition for diffraction experiments is made based on simulations and analytical calculations. There are a few practical considerations to keep in mind when conceiving experiments:

- The beam pipe is 40 mm in diameter; a 20 mm radius is an obvious upper limit to beam size at any point along the beam line, so the rms size should be $\sigma < 7$ mm. Compared to other restrictions listed below, this particular restriction is not a limiting factor.

- The graphite samples are typically only a few hundred microns wide. At best, in the case of the Graphene Supermarket polycrystalline samples, they are about 2 mm wide. The beam should pass in its entirety through the sample. There is thus an upper limit to the beam size at the position of the sample, depending on the type of sample being analyzed.

- The undiffracted beam should be blocked in its entirety by the 8 mm diameter beam block (Figure 2.1).

- When doing diffraction experiments to analyze beam quality, the beam should be large enough at the detector so that detector resolution is not a limiting factor.

- We wish to provide evidence for improved quality of the low temperature source; it is therefore useful to be able to assess changes in diffraction patterns as function of source temperature changes. It is then also important that the above points hold true for higher temperatures, ($\sim 100$ K).
Possible astigmatic effects should not become a hindrance.

In the following sections, it is discussed how these considerations have been utilized in setting up the beam.

4.1.1 Astigmatism

Astigmatism is an optical aberration in which the focal length of a lens (system) is dependent on the transverse direction. The presence of astigmatism in our set-up has already been accounted for [24, 28, 37]. It is attributed to the unintentional quadrupole in the beam line (see section 2.4.2 and table 2.2), which by definition has an equal but opposite lens effect in two lateral dimensions. The effect of astigmatism can be seen in, for example, waist scans such as the one in Figure 3.2b, where the waists of the two axes of the bunch clearly are in two different places. Astigmatism in waist scans is accounted for in the fitting models used to calculate temperature, so it is not much of a hindrance in these scans. However, in diffraction experiments, the physical location of the waist of the beam is important. In practice there are two logical places along the beam line where one would want a waist in such experiments: 1) The waist is placed at the detector, minimizing the diffraction spot sizes, for maximal spot visibility. 2) The beam waist is placed at the sample, minimizing the spot size at the sample. As will be apparent in the results, Chapter 5, the first scenario is useful for sample analysis, while the second is useful for beam analysis. In the first case, if a as-sharp-as-possible diffraction pattern is desired, ideally the two axes have the same waist position. In the second case, the analysis of beam quality in terms of coherence length (which will be done in section 5.3.3), makes the assumption that the waist is at the sample. Astigmatism can therefore be a hindrance for the analysis of diffraction patterns. This has been noted by Bakker [28], and it has been suggested to improve this by including a tunable quadrupole. This has been shown to work, but is sensitive to specific beam parameters such as beam energy $U_e$, initial temperature $T$, etc., making it hard to control.

Under the beam and lens parameters that are of interest in our research, astigmatism is much less prominent than first expected. Suppose the beam waist $1$ is placed at the sample, at 1.25 m along the beam line. Previously, a single magnetic lens was used to achieve this. We now have a second lens at our disposal.

Figure 4.1 shows an overview the GPT simulated beams at and around the sample when using one or two lenses. For this simulation, the beam energy has been set to $U_e = 10.8$ keV. The source size is $\sigma_x, i \times \sigma_y, i \times \sigma_z, i = 20^3 \mu m^3$, and the source temperature $T = 0$. On the left hand side are the resulting transverse spatial electron distributions for a single lens ($I_1 = 2.5$ A), with (a) and (c) the two distributions where $\sigma_1$ and $\sigma_2$ are separately minimal. The distribution at $\sim 1.25$ m is also shown, which is where the beam area ($\sigma_1 \sigma_2$) is minimal. The right side shows the same, but using two lenses, $I_1 = 3.0$ A and $I_2 = 1.23$ A $^2$. The gray bar is used to indicate the relative positions in the beam line of the distribution images. Here the large difference in waist positions for a single lens is clear. In (d) the bunch size in two dimensions $\sigma_1$ and $\sigma_2$ are shown as they develop along the beam line, and each go through their respective minima. Astigmatism, in terms of distance between waist positions, is reduced from 28 cm to 1.3 cm by including a second lens.

Both configurations confirm the presence of astigmatism, as we see that the beam reaches a minimal size in two directions, though at a different position along the beam line. Consequently, there are two thin ‘pancake’ shapes, and one fairly uniform circular shape. The beam thus never reaches its theoretical minimum size (in terms of area). However, it is apparent that including a second lens conveniently brings the positions of minima much closer to each other. The effective area at the waist is thereby more than halved, as can be seen by comparing b.i. and b.ii. The reason for this is as follows: in the two lens configuration, the beam’s angular spread around the

---

1 Up until now we’ve said that due to astigmatism there are two physical waist positions in the beam. In this case we define the waist as the place where the product of the beam sizes in both dimensions is minimal, $(\sigma_1 \sigma_2)_{\text{min}}$.

2 Note that in this configuration the beam undergoes a double-crossover, i.e. there are two waists, one between lens 1 and lens 2, and one after lens 2. The one after lens 2 is placed at the position of the sample, and is the one we are concerning ourselves with.
$I_1 = 2.5 \text{ A, } I_2 = 0.0 \text{ A}$

$I_1 = 3.0 \text{ A, } I_2 = 1.23 \text{ A}$

Figure 4.1: Overview of (simulated) astigmatic effects when placing the beam waist at the sample, for a beam with energy $U_e = 10.8 \text{ keV}$. On the left (i), only the first lens is used in focusing the beam with $I_1 = 2.5 \text{ A}$. On the right (ii), two lenses are used, with $I_1 = 3.0 \text{ A}$ and $I_2 = 1.23 \text{ A}$. The lateral particle distributions of the separate minima of $\sigma_1$ (a) and $\sigma_2$ (c) and the beam 'waist' (b) are shown. The gray bar schematically shows the relative positions where the distribution images have been taken, showing the small positional difference in the waists when using a double lens set-up, when compared to that of a single lens. Also, the development of the bunch sizes along the beam line around the waist is shown in (d).
sample is dominated by the second lens, instead of the quadrupole. The added positive or negative angular spread due to the quadrupole is more visible if only a single, weak lens is used.

In the double lens configuration, the distance between the two size minima $\delta d$ is about a centimeter, as opposed to $\delta d = 28\,\text{cm}$ when using a single lens. When analyzing diffraction patterns, we require $\delta d \ll h$, where $h (= 285\,\text{mm})$ is the distance between the sample and the detector. For diffraction experiments in our set-up, therefore, the double lens set-up is a necessary improvement.

### 4.1.2 Double lens configuration

Having established that the ‘separation’ of the transverse beam minima is reduced by increasing the angular spread, we now take a closer look at the double lens configuration, focusing on the restrictions and considerations presented at the beginning of this section. Simulations have been run to estimate the regimes, in terms of lens currents, in which the conditions are met. For these simulations, typical diffraction experiment properties have been used ($U_e = 10.8\,\text{keV}$, $T = 100\,\text{K}$, $\sigma_{x,i} = \sigma_{y,i} = \sigma_{z,i} = 50\,\mu\text{m}$).

Figure 4.2 shows the spot sizes of the beam at the detector $\sigma_{\text{det}}$ (for simplicity taken in only one direction) as a function of magnetic lens currents, $I_1$ and $I_2$. The three plots are based on (a) actual measurements, (b) the ray tracing model, and (c) GPT simulations. The similarity in the three techniques is apparent. In fact the similarity in GPT simulations and the analytical model is so good, that we have enough confidence to further analyze the beam properties in this section using only the analytical models of section 3.2.1.

When looking at the curve along the surface plot where $I_2 = 0$, the curve of a standard waist scan can be seen, as shown in Figure 3.2. The trench along the surface plots represent the area where the waist of the beam is placed on the detector. From this, a relation between $I_1$ and $I_2$ for focusing the beam on the detector (for this specific beam energy) can be derived. The second trench, seen in the high current area of the plots (top right), is in a regime where there are two beam waists, one after the first lens, and one after the second lens, on the detector. This is a so-called double crossover.

The plots in Figure 4.2 are useful for determining which lens configuration to use: for example, if we want the beam waist to be on the sample, the lens configuration should be such that it corresponds to a trench in the surface plot. Such surface plots can be made along various points along the beam line. Looking back at the beam requirements mentioned at the beginning of this section, it is possible to use the analytical model to determine the desired lens configuration. The requirements were: the beam should pass fully through the sample, should be blocked by the beam block, and should be maximally responsive to source temperature changes. Based on these conditions, the proper beam settings have been determined, as is summarized in Figure 4.3.

Figure 4.3(a) shows a contour plot of the beam size as function of lens currents $I_1$ and $I_2$, at
Figure 4.3: Determination of lens currents. Based on several conditions, a choice can be made for the beam path. Plots (a)-(d) all have the same scale. In (a) and (b) the areas where the beam size meets the requirements is outlined. (c) is a contour plot showing the relative response to source temperature change, where the red area is minimally responsive. From these plots, the optimal beam settings are extracted, shown in (d). The beam path of this set-up is schematically shown in (e).
4.1.3 Temperature dependence

A final check that needs to be made is that the waist of the beam (placed at the sample, as described in the previous section) remains unchanged as a result of different source temperatures. For this, a GPT simulation is run with temperature as variable. The results are shown in Figure 4.4. The beam size as function of position is shown for temperatures ranging from 10 to 300 K. For clarity, only one of the beam waists is shown. The angular spread of the beam is clearly temperature dependent (as can be seen by the slope $\frac{d\sigma}{dz}$), but the position of the waist remains more or less constant, as is shown in the inset. A change of a few mm is considered negligible for analyzing diffraction results as it meets the requirement $\delta d \ll b$, from the previous section. Also noticeable in the figure is that the spot size in the waist increases at higher temperatures. This is a direct consequence of conservation of emittance, $\sigma_x \frac{d\sigma_y}{dz} \propto \sqrt{T}$, valid in a beam waist.

4.2 Diffraction theory

The quantum mechanical wave properties of an electron allow it to interfere when passing through a lattice structure, much like light passing through a grating. In order to be able to understand, interpret, and analyze diffraction patterns, some basic knowledge of diffraction is necessary. This section covers the essential aspects of electron diffraction theory. A more thorough description can be found in [46].
4.2.1 Required sample thickness

When doing transmission diffraction experiments, the sample under investigation should be thin enough that electrons are allowed to transmit through the sample. On the other hand, the sample should be thick enough so that there is an appreciable interaction between the electrons and sample. It is therefore instructive to take a closer look at the fraction of scattered electrons in a beam as function of sample thickness.

The quantum mechanical description of electron diffraction dictates that an electron in fact interferes with itself. We will therefore follow a single electron as it passes through a crystal structure, and see what happens. For simplicity, we assume the crystal is neatly layered, like graphite, with a constant interlayer distance $c_l$, and that the electron travels through the sample perpendicular to the layer planes.

One of three events can happen each time the electron passes a new layer of atoms [47]:

1. The electron does not interact with the layer, passing through undeflected.
2. The electron is scattered inelastically due to atom-electron interactions. For example, the electron might be used in ionizing an atom, releasing a secondary electron. Some of the incident electron’s energy is lost to the ionization process. Inelastically scattered electrons do not produce interference patterns.
3. The electron is scattered elastically through atom-electron interactions. Classically stated, the electrons simply bounce off of an atom site, losing a negligible amount of energy. An elastically scattered electrons wave function is such that only certain outgoing electron momenta interfere, resulting in a diffraction pattern.

The electrons that contribute to the diffraction pattern are those that have scattered elastically only once, with no further interactions. Since we are interested in the amount of electrons undergoing diffraction, it is useful to discuss the outcome of a bunch through a sample of thickness $\tau$, in terms of probabilities. The collisional path of an electron through a medium is often described in terms of the mean free path $\Lambda$, which describes the average distance an electron travels before it encounters a collision event, elastically, $\Lambda_e$ or inelastically, $\Lambda_i$. This means that, on average, after traversing a sample of thickness $\tau$, electrons have undergone $\bar{n}_i = \tau/\Lambda_i$ inelastic and $\bar{n}_e = \tau/\Lambda_e$ elastic collisions during their path$^3$.

Noting that the sample thickness can be written in terms of number of layers, $N$, as $\tau = Nc_l$, we see that it is convenient to discuss scattering probabilities in terms of Poisson statistics:

$$P(k) = \frac{\bar{n}^k e^{-\bar{n}}}{k!} \quad (4.1)$$

where $P(k)$ is the probability that a certain event with average occurrence $\bar{n}$ occurs $k$ times. The probability that no scattering event occurs $P_u$ after $N$ layers is then:

$$P_u = P_e(0) \cap P_i(0) = e^{-Nc_l/\Lambda_i} e^{-Nc_l/\Lambda_i} = e^{-Nc_l(1/\Lambda_i + 1/\Lambda_i)} \quad (4.2)$$

with $P_e$ and $P_i$ the elastic and inelastic scattering probabilities, respectively. The probability that an electron has undergone inelastic scattering at least once is

$$P_i(\ge 1) = \sum_1^\infty \frac{Nc_l e^{-Nc_l/\Lambda_i}}{\Lambda_i k!} = 1 - P_i(0) = 1 - e^{-Nc_l/\Lambda_i} \quad (4.3)$$

The probability that an electron undergoes elastic scattering only once is

$$P_e(1) = \frac{Nc_l}{\Lambda_e} e^{-Nc_l/\Lambda_e} \quad (4.4)$$

$^3$This assertion is not entirely true, as it does not exclude the possibility of scattering multiple times at the same site, which physically does not occur. In the statistics presented here, this event has such a low probability, that it is acceptable to overlook this.
Figure 4.5: Probabilities of various scattering events of $U_e = 10$ keV electrons, depending on the amount of graphite layers they pass through. At more than 20 layers, electrons are mostly lost due to inelastic scattering (and a few more due to multiple elastic scattering events). Around 10 layers is optimal for diffraction experiments.

So now the probability that an electron passes through $N$ layers and elastically scatters only once, with no further inelastic scattering, thus used effectively in diffraction, is given by

$$P_{\text{diff}} = P_e(1) \cap P_i(0) = \frac{Nc_l}{\Lambda_e} e^{-Nc_l(1/\Lambda_e + 1/\Lambda_i)} \quad (4.5)$$

On the other hand, the probability that an electron is lost due to inelastic scattering and/or multiple elastic scattering is

$$P_{\text{lost}} = P_e(\geq 1) \cup P_i(\geq 1) - P_{\text{diff}}$$

$$= 1 - P_u - P_{\text{diff}}$$

$$= 1 - (1 + \frac{Nc_l}{\Lambda_e}) e^{-Nc_l(1/\Lambda_e + 1/\Lambda_i)} \quad (4.6)$$

The sample thickness that will provide the most singly elastically scattered electrons is found from

$$dP_{\text{diff}} \frac{dN}{dN} = 0 \quad (4.7)$$

from which the number of layers $N$ is found to be

$$N = \frac{\Lambda_e \Lambda_i}{c_l(\Lambda_e + \Lambda_i)} \quad (4.8)$$

In our diffraction experiments, electrons have a mean energy around 10 keV, and traverse through a graphite sample, perpendicular to the graphite layers. According to the NIST database [48], the elastic mean free path length for electrons with this energy is $\Lambda_e = 14.8$ nm, and the inelastic length is estimated to be $\Lambda_i \approx \frac{Z}{20} \Lambda_e = 4.43$ nm, with $Z = 6$ the atomic number of carbon. The interlayer distance is $c_l = 0.335$ nm. Figure 4.5 shows the resulting scattered fractions. Around 10 graphite layers is optimal for diffraction purposes. At more than 30 layers, most electrons are lost to inelastic scattering and/or multiple elastic scattering. At best, we can expect about 1 in 10 electrons to contribute to the diffraction pattern. At 50 layers, this decreases to about 1 in 100, and at 100 layers, 1 in 10,000, becoming exponentially less. For a reasonable diffraction output, the layer thickness should therefore not be larger than 50 layers ($\tau \lesssim 17$ nm).
4.2.2 Reciprocal lattice space

We now turn to predicting the diffraction patterns resulting from sending an electron through a crystal lattice. The change in angle of an incident electron at the interference maxima is given by Bragg’s equation

\[ \sin(2\theta) = \frac{\lambda_e}{d_{hkl}} \]  

(4.9)

with \( \lambda_e = h/\sqrt{2m_e U_e} \) the De Broglie wavelength of the electron, and \( d_{hkl} \) the crystal lattice distance with Miller indices h, k and l. Representing the change in angle as \( 2\theta \) is a convention derived from the geometry for scattering, shown in Figure 4.6. Classically speaking, an electron reflects off of a lattice plane. The angle of incidence \( \theta \) is the same as the reflected angle, so the total change in angle of an electron is \( 2\theta \).

The lattice constant \( d_{hkl} \) is dependent on the geometry of the crystal. It is an effective distance, closely related to the physical distance between lattice planes \( a \). Determining the lattice parameter can conveniently be done by representing the crystal structure in reciprocal space.

To make the transformation to reciprocal space, a crystal should first be defined in ‘regular’ space. A crystal can be drawn out by points in real space, each point representing the site of an atom. The crystal is assumed to be infinitely large, so there is an infinite amount of points spanning real space. We will call this set of points in real space our real image. Due to a crystal’s periodic nature, the origin of our coordinate system can be translated to certain positions where exactly the same image is retained. For any 3D crystal, there are only specific translations possible so that this image remains unchanged. These translations are given by an integer multiple of three basis vectors, \( \vec{a}_1, \vec{a}_2, \) and \( \vec{a}_3 \), all combinations of which create a set of points that span 3D space. The eight points given by all different combinations of

\[ c_1\vec{a}_1 + c_2\vec{a}_2 + c_3\vec{a}_3 \]  

(4.10)

with the constants \( c_i \) = \( \{0, 1\} \), are the corner points of a parallelepiped called the unit cell.

The periodicity of the crystal structure allows us to describe this set of points in reciprocal space, a spatial Fourier transformation. The set of points spanning reciprocal space are now given by multiple integers of three new basis vectors, \( \vec{b}_1, \vec{b}_2, \) and \( \vec{b}_3 \), given by

\[ \vec{b}_1 = 2\pi \vec{a}_2 \times \vec{a}_3 / V_e, \vec{b}_2 = 2\pi \vec{a}_1 \times \vec{a}_3 / V_e, \vec{b}_3 = 2\pi \vec{a}_1 \times \vec{a}_2 / V_e \]  

(4.11)

with \( V_e \) the volume of the unit cell. Note that points in real space are not transformed one-to-one to points in reciprocal space; the collection of points in reciprocal space gives us information about the periodicity of the set of points in real space.

Viewing the crystal lattice in reciprocal space provides insight into the expected diffraction pattern: the lattice points \( \vec{g} = (h\vec{b}_1 + k\vec{b}_2 + l\vec{b}_3) \) are directly related to the lattice distances \( d_{hkl} \) from Eq. 4.9, as \( d_{hkl} = 2\pi / |\vec{g}| \). The positions of the points in reciprocal space are thus correlated to the positions of diffraction spots. In fact, as will become apparent in the following section, the acquired diffraction spot pattern is an exact 2D projection of the reciprocal lattice.

4.2.3 Bragg condition

Using the above, a more precise definition of allowed scattering directions can be made. An electron is incident on a scattering site in the crystal with wave vector \( \vec{k}_i = \vec{p}_0 / \hbar \). An elastically scattered electron leaves the sample with wave vector \( \vec{k}_f \), which must satisfy two conditions:

\[ \vec{k}_f - \vec{k}_i = \vec{g} \]  

(4.12)

\[ |\vec{k}_f| = |\vec{k}_i| \]  

(4.13)
Figure 4.7: Schematic diagram of the molecular structure of a graphite layer. All red points are carbon atoms. The basis vectors \( \vec{a}_i \) are indicated, which can be determined as a function of the carbon-carbon bond length, \( l_{\text{c-c}} \).

Bragg’s law, Eq. 4.9, is actually a simplification of those conditions, with: \( |\vec{k}_f - \vec{k}_i| = |\vec{k}| \sin(2\theta) = 2\pi/\lambda \) and \( |\vec{g}| = 2\pi/d_{\text{hkl}} \). These two conditions, Eqs. 4.12 and 4.13, can be combined into one equation, the diffraction condition:

\[
2\vec{k}_i \cdot \vec{g} = |\vec{g}|^2
\]  

(4.14)

According to Eq. 4.12, the change in the electron’s wave vector \( \Delta \vec{k} \) must be equal to a lattice vector. Eq. 4.13 tells us that the magnitude of the ingoing and outgoing wave vector must remain unchanged (i.e. conservation of energy in elastic scattering), meaning that the possibilities for the final wave vector are bound to the surface of a sphere in reciprocal space with radius \( |\vec{k}| \). This sphere is referred to as an Ewald sphere. Any two lattice vector points on the surface of the Ewald sphere represent an allowed transition \( \Delta \vec{k} \), resulting in a limited amount of allowed outgoing wave vectors. These constitute the final diffraction pattern.

4.2.4 Diffraction from graphite

With the theory of electron diffraction through crystals worked through, we now take a closer look at the sample that has been investigated in this research, graphite. Graphite is a naturally occurring allotrope of carbon, as commonly used in everyday life as in experimental physics. Graphite is highly anisotropic; it contains planes of 1-atom thick sheets, stacked and held together by Van Der Waals forces. Each individual layer, has an intensely high tensile strength parallel to the plane. The stacked layers, however, are weakly bonded and can easily shear or simply be peeled or scratched off.

A diagram of the molecular structure of a single graphite layer is shown in Figure 4.7. The carbon atoms are arranged in a hexagonal lattice, also known as a ‘chicken wire’ structure. The basis vectors \( \vec{a}_i \) are indicated in the image, with \( \vec{a}_3 \) pointing perpendicularly out of the plane of the page. Graphite sheets can be stacked on top of each other in various different geometries, with an interplane distance \( c = 0.335 \). The most common naturally occurring stacking type in graphite is AB stacking, meaning two neighboring planes are shifted by a distance \( l_{\text{c-c}} \) with respect to each other. One neighboring sheet further is again identical to the first sheet. The lattice vector \( |\vec{a}_3| \) is therefore twice the distance between two planes, \( |\vec{a}_3| = 2c \).
As a reminder, the basis vectors (and any integer combination thereof) represent translations of the crystal that result in exactly the same crystal structure. The choice of basis vectors is not unique, and many other documents on graphite crystallography [49] use different basis vectors. The volume of a unit cell, number of atoms per unit cell, and more importantly the resulting calculated lattice distances \(d_{hkl}\) are fixed for graphite, regardless of the choice in basis vectors.

In terms of \(a = |\vec{a}_1| = \sqrt{3}a - c\) and \(c = |\vec{a}_3|/2\), the basis vectors can be written as:

\[
\vec{a}_1 = \begin{pmatrix} 0 \\ a \\ 0 \end{pmatrix}, \quad \vec{a}_2 = \begin{pmatrix} a \sin(60^\circ) \\ a \cos(60^\circ) \\ 0 \end{pmatrix}, \quad \vec{a}_3 = \begin{pmatrix} 0 \\ 0 \\ 2c \end{pmatrix} \quad (4.15)
\]

Using Eq. 4.11, the reciprocal space vectors are calculated to be:

\[
\vec{b}_1 = 2\pi \left( -\frac{a\sqrt{3}}{2} \right), \quad \vec{b}_2 = 2\pi \left( \frac{2a\sqrt{3}}{3} \right), \quad \vec{b}_3 = 2\pi \left( 0 \quad \frac{1}{\pi} \right). \quad (4.16)
\]

The scattering points in reciprocal space are given by:

\[
\vec{g} = h\vec{b}_1 + k\vec{b}_2 + l\vec{b}_3 = 2\pi \left( \frac{-1}{a\sqrt{3}} h + \frac{2}{\sqrt{3}a} k \right), \quad (4.17)
\]

and finally, the lattice constants \(d_{hkl}\) are given by

\[
d_{hkl} = \frac{2\pi}{|\vec{g}|} = \frac{1}{\sqrt{\frac{4}{5a^2}(h^2 + k^2 + hk) + \frac{c^2}{4}}} \quad (4.18)
\]

For graphite, with \(a = 0.2346\) nm [49], the first order lattice distance, defined as the lattice distance that results in the smallest diffraction angle \(2\theta\) is found for Miller indexes

\[
(h, k, l) = \pm \begin{cases} (1, 0, 0) \\ (0, 1, 0) \\ (1, -1, 0) \end{cases} \quad (4.19)
\]

resulting in \(d_{110} = 0.2131\) nm.

The collection of scattering points \(\vec{g}\) in 2 dimensions (for \(l = 0\)) is shown in Figure 4.8. The collection of points represent the various scattering transitions, thus represents the expected diffraction image in real space for electrons incident along the z direction. The circles connect points that are equidistant from the central point, \((h, k, l) = (0, 0, 0)\). These represent the expected diffraction pattern for an electron transmitting through a graphite sample with randomly oriented flakes. The diffraction spots are then smeared out over the entire circumference of the ring.

In our experiments, an electron beam travels through the sample in the z direction, with momentum \(p_z = \sqrt{2m_eU_e}\). Let’s first assume that the transverse angular spread is negligible. The beam is thus characterized by initial wave vector \(\vec{k}_i = (0, 0, k_z') = (0, 0, p_z/h)\). Figure 4.9 shows a 2D representation of the Ewald sphere surface (blue curve) for this situation, and the allowed scattering transitions (orange points) in reciprocal space. The lattice points represent \(\vec{g} = h\vec{b}_1 + k\vec{b}_2 + l\vec{b}_3\) for different combinations of \(h\) and \(l\), with in this example \(k = 0\). Nowhere does the surface of the Ewald sphere overlap exactly with a scattering site, \(\vec{g}\), most certainly not for points where \(l = 0\). This also follows from the diffraction condition, using \(k_i = (0, 0, p_z/h)\) in Eq. 4.14, which becomes

\[
p_z = \frac{4c\hbar}{3a^2l} (h^2 + k^2 + hk) + \frac{l\hbar}{4c} \quad (4.20)
\]

revealing that for any diffraction to occur with \(l = 0\), \(p_z\) is required to approach infinity. Luckily, the diffraction condition is not as strict as it has been made out to be up until now: an excitation
Figure 4.8: The reciprocal lattice of graphite, viewed in the $l = 0$ plane. The points represent scattering sites, thus represent the positions of diffraction spots for a beam incident along the $z$ direction. The rings connect points with equal distances from the center. [50]

Figure 4.9: Cross section of an Ewald sphere in the reciprocal lattice of graphite. The image is scaled to a typical experimental value of $\hat{\mathbf{k}}$. The inset shows a close-up of the mismatch between the sphere and an allowed diffraction transition. For high $|\hat{\mathbf{k}}|$, the mismatch is small. The excitation error $\hat{s}$ bridges this gap.
error can be introduced to cover small deviations between the Ewald sphere and scattering sites. In the figure, this excitation error is given by the vector $\vec{s}$, which connects the lattice point to the Ewald sphere, adjusting Eq. 4.12 to $\vec{k}_f - \vec{k}_0 = \vec{g} + \vec{s}$. The excitation error vector is parallel to the incoming wave vector $\vec{k}_0$. This deviation, however, comes at a cost: the larger the excitation error, the smaller the probability that the scattering event takes place, thus resulting in a lower diffraction intensity. The actual intensity depends on a variety of factors, including $\vec{s}$, well covered in [51]. In this research, the relative intensities of diffraction points is not of direct interest, and will not be further elaborated on.

Up until now in this section, diffraction has been described by a beam consisting of electrons all travelling in only the $z$-direction. We now know what spots to expect for a perfectly coherent beam, though in practice, the beam has transverse angular spread, $\sigma_{\phi_0}$, which will result in an angular spread in the diffracted beam. In Appendix C we derive that for small deviations in incoming angles, the outgoing angular spread of the diffracted beamlets is the same as the incoming spread, only altered in direction by a fixed angle $2\theta$.

Knowing the beam properties at the sample, we thus we know exactly what to expect on the detector. In most cases the spot size on the detector $\sigma_d$ is dominated by the transverse angular spread $\sigma_\phi$ and not the size of the beam on the sample, so the spot size on the detector can be written as $\sigma_d = \sigma_\phi h$, with $h$ the length between the sample and the detector. This holds for the spots as well as the undiffracted beam. In fact the diffraction spot sizes are expected to be the same as the spot size on the detector in the absence of a sample.

As a final extension to expected diffraction patterns, it should be noted that one of the samples investigated in this work is polycrystalline graphite, meaning the sample contains domains of randomly orientated graphite flakes. As the beam size is much larger than the typical domain size, angular information about the diffraction spots is lost. In other words, the spots are smeared out azimuthally, resulting in ring patterns. The width of the ring should be about the same as the long axis of the spots.
Chapter 5

Results

In this section the results of ultracold diffraction experiments with the UCP set-up are presented. Using the ultracold electron beam for diffraction is still a relatively new development. The first ultracold diffraction images have been acquired and presented by Bakker [28] and Engelen [24]. This is not only a first in our lab, but, as far as we know, the first ultracold diffraction pattern in the world. While much work has been done to achieve such images, the experimental set-up has required further improvement in order to do diffraction experiments with varying parameters, for investigating the beam’s coherence properties. The direct improvements to the set-up have been essential for being able to run reproducible diffraction experiments, the results of which are described in this chapter.

The two types of samples used in this research, mono- and polycrystalline graphite, are described in 5.1, including the fabrication process of sufficiently thin monocrystalline graphite.

As mentioned in the Introduction, the goal is to determine the beam quality quantitatively directly from diffraction patterns. Both samples have been investigated in diffraction experiments. Diffraction through the polycrystalline graphite sample has produced sharp ring patterns. The method that these patterns have been analyzed is described in section 5.2.2. Diffraction experiments, such as waist scans and temperature dependence of ring width are then shown. One of the conclusions of these experiments is that diffraction rings are as of yet poorly suited for accurate beam quality measurements. Because of this, we have undertaken the process of developing thin monocrystalline samples. The diffraction spots from the new samples have shown to be much more reliable indicators of beam quality, as is shown in section 5.3. The sample has been used in several diffraction experiments, similar to those attempted for the polycrystalline sample. The quality and reproducibility of results from diffraction spots has proven to be high enough to run accurate temperature dependent diffraction experiments, using the lens set-up described in section 4.1.2. The results of these experiments, shown in section 5.3.3, have been submitted to Nature Communications (see Appendix A).

5.1 Graphite samples

During this research, two graphite samples have been used:

- Chemical vapor deposited graphite, supported by a 2000 mesh TEM grid [52]. The sample is polycrystalline, thus consists of randomly oriented flakes. According to specifications, the sample is \( \sim 6 \) layers thick, though AFM measurements done by van Lieshout [50] have shown that \( \sim 30 \) layers is probably a more accurate estimate.

- A natural graphite [53] film placed on a 200 mesh TEM grid. The graphite sample has been manually exfoliated. Based on light absorption with an optical microscope images, we estimate that the sample is 20 – 30 layers thick.
The polycrystalline sample has been used in this research for measuring and analyzing powder diffraction patterns. However, the analysis of these measurements has proved inaccurate, leaving desire for diffraction spot patterns from which more accurate conclusions can be made. Monocrystalline graphite samples are available from Naturally Graphite [53], but do not come ready-made for electron diffraction experiments. The graphite samples are available as cylindrical blocks of 1 mm diameter, and 1 mm height. When using the samples for diffraction experiments, they are required to be sufficiently thin (< 50 layers, as has been determined in Chapter 4). Additionally, it goes without saying that a free-standing graphite sample of just a few tens of layers thin is far from handleable, so a support grid is required. In the following section we give a methodological overview of the fabrication of few-layer graphite samples on TEM grids, usable for electron diffraction.

5.1.1 Sample preparation

The method of exfoliating graphite to just a few layers is similar to the methods used by Nobel Prize winner A. Geim, now dubbed the ‘Scotch tape technique’, where layers of graphite are peeled off of a bulk sample bit by bit, until a single layer is left [54]. We have found that our sample preparation is not an exact reproducible scientific method, but more of a trial-and-error method, to some extent prone to good or bad fortune. The method is taken from experience-based advice from Klarenhaar [55].

A fresh block of graphite is placed on a piece of Scotch tape. Alternatively, a previously peeled layer of graphite, already on Scotch tape, is used. The graphite layers are parallel to the tape. Providing that the sample is thin enough, a TEM grid (200 mesh) is placed over the sample, onto the tape. One of the edges of the tape is folded over itself, creating a non-adhesive handle. Meanwhile, a small chunk of crystal bond (Quickstick 135 Mounting Wax) is placed on a clean microscope slide. The slide is placed on a heating plate (∼ 150 – 180 °C), allowing the wax to melt. The tape with graphite sample is then placed onto the wax. Some light pressure is applied over the sample so that the TEM grid is firmly attached to the molten wax. The slide is removed from the heating plate and allowed to cool (few minutes). The wax has once again hardened, and, if all is well, firmly supports the TEM grid with graphite. The tape can be removed slowly, by pulling on the handle, making a highly acute angle in the tape. Some graphite layers will most likely be removed in this process.

The graphite layers can now be peeled off. A new piece of tape, with a folded non-adhesive handle, is placed over the graphite sample, and lightly pressed onto it with a non-sharp pair of tweezers. The tape should be pressed on so that it is visibly attached to the graphite, but gently, so that the graphite does not become cracked or otherwise damaged. The tape can now once again slowly be removed, peeling at an acute angle. This process is repeated, preferably placing the tape on the sample from a different angle each time, until the sample is thin enough. Judging thickness is partly based on experience, but as a rule of thumb, if the sample starts becoming slightly transparent, it is approaching the desired thickness. Especially for thinner samples, the risk becomes high that the full sample is removed from the TEM grid, so care must be taken not to overdo the process; it requires, in practice some effort and luck to end up with a sample with the correct thickness.

Once a proper thickness is found, the TEM grid, together with the sample, needs to be removed from the crystal bond and microscope slide. As the sample will, in the end, solely be connected to the TEM grid via (weak) van-der-Waals forces, it is understandable that removing the TEM grid along with the sample is a delicate procedure. The bond material is dissolved by applying a few drops of acetone around (not on) the sample. The acetone will slowly dissolve the bond from the outside inwards. This may require a few repetitions, as acetone dries up rather quickly. It is important not to use too much acetone, since the graphite can simply float away if there is a drop of acetone on top of the TEM grid. In fact, the most successful TEM grid removals have been achieved not with pure acetone, but a 1:3 acetone-to-water mixture. The cohesive properties of water allow it to stay more localized as a drop on the glass slide, instead of spreading out. Sharp tweezers are used to gently remove the TEM grid.
Once the grid is removed, there will most likely be bond material stuck to the bottom. Some extra cleaning is required. Again, the graphite flake is very prone to floating off; therefore, the TEM grid is placed on top of a 1:3 acetone-to-water droplet. The grid floats on this droplet, and the graphite flake in turn remains firmly on the grid. The grid is allowed to stay on the droplet for a while. This process is repeated until the TEM grid is clean and allowed to dry. We now have a sample that can be placed directly in the UCP beam line, for electron diffraction experiments.

Figure 5.1 shows optical microscope images of typical thin graphite samples, lit from (a) below and (b) above. From the transmission image, (a), it has been determined that \( 40 - 50\% \) of light is absorbed in the marked area. Graphite absorbs \( p = 2.3\% \) of light per layer [56]. This can be used to estimate the amount of graphite layers, using \( N = \frac{-\ln(I/I_0)}{1-p} \), where \( I \) is the measured intensity, and \( I_0 \) is the background intensity. From this we estimate that the graphite sample contains \( 20 - 30 \) layers.

5.2 Diffraction rings

As mentioned in the introduction of this chapter, the ring patterns from the polycrystalline graphite sample were studied because more complicated crystals, which we plan to investigate in the future, will in many cases also be polycrystalline. It is therefore instructive to be able to analyze ring patterns of ultracold diffraction experiments, for example to know where expected difficulties and inaccuracies lie. Also, it is useful to know what types of experiments can be done, and what the limits and advantages are of using an ultracold source in such experiments. For example, to what extent is the quality of the beam observable in diffraction rings? Only the first solenoid (see Figure 2.1 and Table 2.2) lens was used in acquiring results in this section. Using this lens, the beam is focused behind the detector.

5.2.1 Ring patterns

Figure 5.2 shows a typical example of a diffraction image in our set-up, using polycrystalline graphite as a sample. In the image, two rings are visible, labelled (1) and (2). The beam block (3), used to block the 0th order beam is outlined. The outer outlined circle is the edge of the detection area (4), 40 mm in diameter. To capture this pattern, electrons have been accelerated to \( U_e = 11.3 \) keV, by an acceleration potential of \( V_{acc} = 24 \) kV. The ionization volume is roughly \( 40 \times 40 \mu m^2 \) and has been produced by the nanosecond laser (Table 2.2). Typical images are a result of about 100 shots at a 10 Hz repetition rate. The temperature of the source is not yet relevant at this stage, so has not been measured for this particular set-up, but based the model presented in section 2.3, with \( \lambda_l = 480 \) nm and \( V_{acc} = 24 \) keV, the temperature is around \( T = 100 \) K.
Figure 5.2: Diffraction ring pattern acquired with a $U_e = 11.3$ keV electron beam, from an ultracold source. In the image a first (1) and second order ring (2) can be seen, though most of the second ring falls out of the detection range. The beam block (3) and the edge of the 40 mm diameter detector (4) are outlined.

The image in Figure 5.2 is an average of several images, to reduce noise. The CCD camera often detects ‘dead counts’, resulting in hot pixels, i.e. bright detected peaks unrelated to the beam. These pixels are removed in image post-processing, either by subtracting a background image in which the hot pixels are also visible, or by manually ‘cleaning up’ the image.

Also visible in the figure, is a semicircular sharp bright spot near the bottom of the detection area. This is a commonly recurring pattern in our diffraction images, especially when the diffraction signal is relatively weak, so that a long camera exposure time is required. This bright area is caused by a second electron source: during the acceleration process, ions accelerated in the opposite direction of electrons towards the rear of the accelerator structure, seen in Figure 2.5 to be a mirror for the MOT beams. Upon bombardment with this mirror, secondary electrons are generated, which are accelerated down the beam line, with $U_e \approx eV_{ac}$. The path of these electrons is poorly defined, but what can be seen in Figure 5.2 is a part of this electron cloud passing under the sample plate. The upper part of the semicircle is the bottom edge of the sample plate, schematically shown in Figure 2.7, and the rounded lower part comes from the inner edge of the beam pipe. The secondary electron cloud can be focused onto the detector by increasing the current of the first magnetic lens. The current at which the cloud is focused on the detector is consistent with a beam energy of $U_e \approx 24$ keV.

5.2.2 Ring analysis

To draw conclusions about the beam from diffraction images, it is necessary to quantify the properties of the ring pattern, the most important of which being the ring width. To start of with, the center of the rings must be accurately determined. Ring width can easily be only several pixels wide, typically no more than 10; therefore it is crucial to find the ring center with an accuracy down to a pixel. If this is not done, azimuthally integrating over the image from the center will result in a wider ring profile than actually present in the image. The method of finding the center of the ring is described below, and is graphically shown in Figure 5.3.

To find the center, first a guess of the central position and radius of the first order ring is made (Fig 5.3a). Then, radial line profiles are taken for a large set of angles between 0° and 360°, with the guessed point as a center. The collection of these line profiles as function of angle is shown in Fig. 5.3b. The x-axis is the angle from 0° to 360°, and the radial profile is shown along the y axis. At the bottom of the image the beam block can be seen. The beam block’s stem is visible in the middle of the image. If the center had been chosen appropriately, the diffraction ring would appear on the image as a straight line. This is clearly not the case in the upper profile. To correct for this, the position of the maximum around the guessed radius (red line) is determined. The difference
between the guessed radius and measured position of the maximum is plotted as function of angle in Fig. 5.3c. If the center and radius had been chosen correctly, the difference should be about zero for all angles, which is, again, not the case. The offset is given by a sinusoidal profile. A sinusoidal fit results in a correction for the center and radius. The process is repeated with the new guess, until the amplitude of the offset is sufficiently reduced, typically when the amplitude of the fitted sine is less than 1 pixel.

With the center accurately determined, a radial profile can be made. A simple method to do this is to simply average the radial profiles of the various angles, such as in Fig. 5.3. Only the relevant angles, where the ring is present (so not where it is outside the detection area or behind the beam block) should be taken into account in averaging. Figure 5.4 shows the resulting profiles.

In 5.4a the diffraction peaks of the two rings can be seen, with their radial position indicated by the dashed lines. The general declining curve is caused by a cloud of inelastically scattered electrons. The angular distribution function of inelastic electron scattering is such that fewer electrons are scattered at higher angles, also visible in the diffraction image of Fig. 5.2. The relevant angular scattering cross section is a rather complicated function, given by Eq. (7.16) in [51]. This function, however, poorly matches our data. Instead, an empirically determined even rational polynomial function, 

\[
\frac{a_1}{(r^2+a_2)^2} \frac{a_3}{(r^2+a_3)^2}
\]

is used as a fitting function, where \( r \) is the radial distance and \( a_{1,2,3} \) are fit parameters. This more closely matches the background signal. This fit is shown by the red dashed line. This inelastic background is removed from the profile. The intensity of the rings is smeared out over a larger area at higher radii. A correction is therefore applied to the profiles by multiplying the intensity by the radius radius. The resulting curve is shown in Fig. 5.4b. A double Gaussian fit is used to characterize the separate peaks, shown by the red dashed line. A double Gaussian is used because of detector artefact that causes a background signal around spots, or in our case, rings. The origin of this artefact is documented in [57].
Figure 5.4: Average radial profile of the diffraction ring in 5.2. Two peaks are present in the radial profile, at 15.4 mm and 26.5 mm away from the center. In (a), the overall declination of the curve is attributed to inelastically scattered electrons. This background is removed, resulting in (b). The two peaks are fitted with a Gaussian fit.

The data found from the Gaussian fits of the two peaks is presented in the following table:

Table 5.1: Properties of the diffraction pattern from Figure 5.2

<table>
<thead>
<tr>
<th></th>
<th>Ring 1</th>
<th>Ring 2</th>
</tr>
</thead>
<tbody>
<tr>
<td>Radius (mm)</td>
<td>15.5 ± 0.1</td>
<td>26.5 ± 0.1</td>
</tr>
<tr>
<td>$2\theta$ (measured, mrad)</td>
<td>55 ± 1</td>
<td>93 ± 2</td>
</tr>
<tr>
<td>$2\theta$ (Bragg, mrad)</td>
<td>53.8 ± 0.2</td>
<td>93.3 ± 0.3</td>
</tr>
<tr>
<td>Width (mm)</td>
<td>1.23 ± 0.03</td>
<td>0.51 ± 0.02</td>
</tr>
</tbody>
</table>

In Table 5.1, the width of the ring is taken as the square root of the variance of the Gaussian fit that describes the peak (and not the second Gaussian that corrects for the detector artefact). The measured diffraction angles are determined using $\theta_{1,2} = \tan^{-1}(r_{1,2}/h)$, where $h = 285$ is the distance between the sample and the detector. These values are in good agreement with the theoretical angles calculated using Bragg’s law (Eq. 4.9) with $\lambda_e = 2\pi\hbar/\sqrt{2m_eV_e}$.

The large difference in the width of the first and second order ring is unpredicted. Since the small-angle approximation is valid, we expect the widths of each ring to be the same (see section 4.2.4). Part of the reason for the difference is that a double Gaussian fit was necessary to more accurately fit the first peak. The peak is thus not perfectly described by a Gaussian, unlike the second peak, which hardly required modifying. Since the exact mechanism of the detector background is not well understood, it is difficult to determine the accuracy of the double Gaussian fit in determining the actual spot size. Another issue is apparent from Fig. 5.2: the first ring runs into the brighter secondary electron area, which can distort the intensities around the ring. It also appears from the figure as though the width of the first ring varies along its perimeter: at about 8 o’clock the ring seems particularly thin, while in the lower and upper part of the ring, it seems thicker. Analyses of separate parts of the ring have confirmed that the determined width is highly dependent on which portion of the ring is taken into account. This is another possible reason for the difference in the 1st and 2nd ring widths.

It should be noted, however, that the analyzed image is not a typical example of diffraction images, but of a lesser quality. Firstly, the image is an average of several images. Spreading due to beam jitter cannot be ignored in this case (though we would expect to see equally much influence of beam jitter in the first and second order rings). Secondly, an image with more noticeable secondary electron noise (the semicircular bright area in Figure 5.2) has been chosen for analysis. In most images, the intensity of the diffraction patterns was sufficiently much higher than the noise, that we have confidence in the accuracy of the resulting measured ring widths. This particular image has been shown to focus on some of the difficulties involved in acquiring data from polycrystalline samples.
5.2.3 Waist scans

A waist scan (see 3.2.3) is an accurate method from which we can deduce the coherence length. If a waist scan is done using the ring widths instead of spot size $\sigma_d$, the coherence length, being a quantum property, is measured directly through a diffraction measurement, which is by definition a quantum effect.

Waist scans have been done with a $U_e = 10.3$ keV electron beam. The ns laser has been used for ionization. Only the first magnetic lens is used, as is conventional in waist scans [24]. The source size, $\sigma_{x,i} \times \sigma_{y,i} = 21 \times 44 \mu m^2$ has been determined using space charge scans (section 3.2.2).

The results of the waist scans are shown in Figure 5.5. Firstly, a ‘standard’ waist scan of the 0th order spot is shown by the red circles and blue squares. The two different data sets are the rms sizes of the short and long axes of the elliptical beam spot at the detector. This waist scan is done without a sample and is shown as reference, to have something to compare 1st order results to. The minima of the two data sets do not occur at the same magnetic lens strength, caused by astigmatism (section 4.1.1). The two lines are waist scan fits, from which the source temperature ($T = 130 \pm 6$ K) has been determined, following the method of section 3.2.3.

These scans have been repeated, now with the graphite sample in the beam, but still looking at the 0th order (undiffracted) spot. We expect the same results as the normal waist scan. However, such a scan is good as a second reference, since any deviations will most likely be carried over into the 1st order waist scans. The red and blue crosses in Figure 5.5 show this data set. Between currents of 2.0 and 2.6 A, the 0th order results through the sample exactly match those without a sample. However, outside these ranges, deviations occur. At currents below 2.0 A, both axes of the spot are smaller than expected. At these currents, the beam is clipped by the edges of the sample holder (Figure 2.7), about 2 mm in diameter. With the sample holder acting as an aperture, the resulting spot size is smaller than for an unrestricted beam. Curiously, at high magnetic lens currents, the size of the long axis is larger than that of the sample-free beam, while the same clipping behavior as for low currents is expected. The fact that the spot size is larger in this
regime suggests that there is some other scattering or reflection occurring, most likely from the edges of the sample holder or the sample itself, though the exact nature is unknown. Comparing 0th order data, we see that 1st order data will not be representative of the beam if the lens current is outside of the range $2.0 \ A < I_1 < 2.6 \ A$.

Finally, we look at ring patterns in these same scans. The method of acquiring ring widths from diffraction images is described in the previous section. The results are shown by the green triangles in Figure 5.5. The ring widths behave accordingly with 0th order data: the shape of the curves of the 0th and 1st order data are very similar; the current at which the minimum ring width occurs matches that of the 0th order beam.

The ring widths are consistently higher than the average of the 0th order spot sizes, about 0.3 mm more. A few explanations for this are listed below:

- The ring size is defined in one dimension, its width, while spot sizes are defined in two dimensions, along the long and the short axes. The final ring width, which is integrated over the full visible part of the ring, is a blend of the long and short axes of the 0th order spot. While this does not explain the full 0.3 mm difference, it is clear that the ring cannot be as low as the minimal spot size.

- Individual diffraction images require more time to be captured than images used in waist scans. In our case, diffraction images have been taken with an exposure time of 10 seconds, while 0th order data has used exposure times of typically $1 - 2$ seconds. It has previously been shown [28] that measured sizes of spots and/or rings increase with increasing exposure time due to instabilities in the beam.

- As seen in the previous section, the images have certain imperfections which can influence the measured width. The effect of these is not always well known, but imperfections will never lead to a smaller width.

- A small broadening could also occur if the choice of the center of the ring is off by a pixel or two. The difference between the 0th and 1st order data is about 400 $\mu$m, which is equivalent to about 4 pixels. The analysis of ring patterns might therefore not be accurate enough to properly determine the actual width.

- Graphite flakes in the polycrystalline sample are distributed in random orientations in a fixed plane. However, it is possible that some flakes are angled out of plane, which can result in broader diffraction angles.

- If the sample itself is tilted with respect to the incoming beam, some ring broadening might occur. Also, as a result the circle might not be azimuthally symmetric.

Despite the offset, the results of this waist scan give hope that relevant information can be extracted from diffraction patterns. As can be seen in the examples of various waist scans in Figure 3.2, much of the information about source temperature is extracted from the slopes of waist scan data. In Fig. 5.5 the relevant slopes are measured in the current ranges $1.8 - 2.2 \ A$ and $2.4 - 2.7 \ A$. The slopes are expected to increase roughly by a factor of 1.7 by an increase of temperature from 10 K to 100 K, which is much larger than the error between the slopes of the 1st and 0th order data, so a ball-park figure of temperature can still be deduced from ring waist scans. Furthermore, a lower limit of the beam quality can be deduced from the diffraction image. Based on the width of the 1st ring, the radii of the rings can be scaled down by a factor of 9 while keeping the 1st and 2nd rings distinguishable. This means under the same beam conditions, a diffraction pattern can still be resolved if the graphite sample is replaced by a sample with a 9-times larger lattice spacing, $d = 1.9 \ nm$.

In conclusion, the widths of the rings in waist scans follow the expected trend, but have a consistent offset. The measured ring widths are therefore not representative of the quality of the beam.
5.2.4 The need for a monocrystalline sample

Given the inaccuracies and problems encountered in this particular investigation, ring patterns and analyses thereof are not sufficient for accurate beam quality measurements. We have therefore fabricated few-layered graphite by manual exfoliation of a monocrystalline graphite block (section 5.1). Instead of rings, these samples produce diffraction spots. Spots can provide more accurate results from diffraction experiments for a few reasons:

- Since diffracted electrons are now concentrated in select spots instead of being spread out over a ring, the diffraction intensity, in relation to the background is expected to be larger.

- Due to the higher intensity per spot, a shorter integration time is possible for acquiring diffraction images. Beam jitter will thus have less influence on the measured spot sizes.

- Also due to the increased intensity per spot, we can allow for larger spot sizes without them being ‘overshadowed’ by background noise.

- Instead of a single ring, there are several spots from which the size can be determined. Furthermore, the size of the spots is measured in two dimensions, further detailing the spots.

- Analysis of spots is much easier than of rings. Two-dimensional Gaussians are already used to analyze 0th order spots from our set-up; the same method can be used for 1st order spots.

- Defects caused by random flake orientations do not occur in the monocrystalline sample.

5.3 Diffraction spots

The diffraction spots described in this section are based on two beam line configurations, shown schematically by the upper and lower beam diagrams in Figure 5.6. The upper diagram shows the two magnetic lenses working together to focus the beam onto the graphite sample. The lower set-up uses only the first lens to place the waist of the beam (had it not been blocked) on the detector. Each of these two set-ups can be described by a point in the lens current diagram presented in Figure 4.3, shown again in Figure 5.6. The upper set-up is in the region of interest for beam analysis in diffraction experiments as described in section 4.1.2. The set-up produces large diffraction spots on the detector which are expected to change noticeably due to source temperature changes. Also, the beam is focused onto the sample, giving a minimal spot size at the sample, $\sigma_{\text{smpl}}$. The lower set-up provides sharp diffraction spots. These are useful in sample analysis, and also tests the limits in producing high-visibility diffraction patterns. The position in the current diagram of Fig. 5.6 is also in the acceptable region, although the influence of temperature changes is minimal. We therefore do not expect to be able to do temperature dependent experiments with this set-up. Furthermore, the spot sizes on the detector are expected to be smaller than the detector resolution, so information about beam quality is lost. The results of diffraction experiments that have been run with the two beam set-ups are presented in the following sections.

5.3.1 Sharp diffraction spots

Figure 5.7 shows a diffraction pattern produced with a $U_e = 13.2$ keV electron beam created by fs photo-ionization. As the source temperature minimally influences the spot size in this measurement (see 4.1.2), it is not important to know it exactly, but it is estimated to be 70 – 80 K, using the model presented in section 2.3, for a femtosecond ionization beam. Shots are fired at a repetition rate of 100 Hz with a total exposure time per image of 10 s. Diffraction images are thus the result of $10^3$ shots, each shot containing a few hundred electrons. Since the spots are very bright, decent images can also be acquired at shorter exposure times. However, as will be elaborated later, the spot size is limited by the resolution of the detector; beam jitter thus
Figure 5.6: Two magnetic lens configurations, resulting in different beam lines. The upper schematic shows how two lenses are used to place the beam waist onto the sample, resulting in broad spots. The lower schematic shows the beam focused onto the detector, resulting in sharp diffraction spots. The points in the current diagram from section 4.1.2 represented by these set-ups are circled.

only has a relatively minor contribution to spot broadening. It is therefore acceptable to increase exposure time for a less noisy image. The image shown in Figure 5.7a is an average over 10 separate images, for illustrative purposes. The presented analyses are, however, based on individual 103-shot diffraction images. In Figure 5.7a, five of the six 1st order spots (1) and one 2nd order spot (2) are visible. The beam block (3) and the detector edge (4) are outlined. In (b) a profile along one axis of the spot is shown. A Gaussian fit is used to determine the spot size, $\sigma_d = 180 \, \mu m$ and $210 \, \mu m$ along the short and long axes of the spot, respectively.

Analyzing spots from a diffraction image is considerably easier than rings: a 2-dimensional Gaussian fit already results in an accurate measurement of the spot’s size and position. The only extra step taken is to take a look at the background level surrounding the spot, and subtracting the plane that fits through that background. A profile along one of the axes of the spot is shown in Figure 5.7b. The figure also shows a Gaussian fit through the data, from which the spot size is determined. The spot size, based on fits of multiple spots in the same image, has been calculated to be $\sigma_{d,1} = 180 \, \mu m$ and $\sigma_{d,2} = 210 \, \mu m$ for the short and long axes of the spot, respectively. The diffraction angle $2\theta$ can be determined from the distance between the points.
and center, where the 0th order beam would arrive were it not for the beam block. However, the distance can be determined much more accurately by measuring all intra-spot distances and averaging these (possibly correcting by a factor \( \sqrt{3} \) or 2 due to the geometry of the hexagonal structure). For 6 spots, as in Figure 5.7, the diffraction angle can be based on an average of 15 different intra-spot distances. In this particular case, this distance is measured to be \( s = 14.3 \) nm. With a distance from the sample to the detector of \( h = 285 \) mm, a diffraction angle of \( 2\theta = \tan^{-1}(s/h) = 50 \pm 1 \) mrad is calculated, in excellent agreement with the value calculated using Bragg’s law, \( d_1 \sin(2\theta) = 2\pi h/\sqrt{2m_eU_e} \), resulting in \( 2\theta = 49.9 \) mrad. Here \( d_1 = 0.2131 \) nm is the first order lattice constant of graphite. An overview of the properties of the spot pattern are listed in Table 5.1. The measured spot size \( \sigma_d \) is an average of the sizes of the separate spots, which are all within 10% of each other.

<table>
<thead>
<tr>
<th>Distance between neighboring spots</th>
<th>14.3 ± 0.5 mm</th>
</tr>
</thead>
<tbody>
<tr>
<td>( 2\theta ) (measured)</td>
<td>50 ± 1 mrad</td>
</tr>
<tr>
<td>( 2\theta ) (Bragg)</td>
<td>49.9 ± 0.5 mrad</td>
</tr>
<tr>
<td>( \sigma_d )</td>
<td>(180 ± 10) ( \times ) (210 ± 10) ( \mu m )</td>
</tr>
</tbody>
</table>

It is useful to analyze the overall quality of the diffraction image and relate that to beam quality. The measured spot size is \( 180 \times 210 \) \( \mu m^2 \), and the distance between spots is 14.3 nm. This means that we could bring the spots 12.9 times closer to each other before the visibility decreases to 88%, the optical criterion for visibility of interference fringes [58]. Here, visibility is defined as \( \nu = \frac{I_{\text{max}}-I_{\text{min}}}{I_{\text{max}}+I_{\text{min}}} \), with \( I_{\text{max}} \) and \( I_{\text{min}} \) the maximum and minimum intensities of the diffraction pattern, in this case estimated by overlapping Gaussians. Being able to shift the spots 12.9 times closer, while retaining sufficient visibility has a very exciting implication: the diffraction pattern will still have resolvable spots even if the graphite sample is replaced with another sample with a lattice constant of \( 0.2131 \) nm \( \times \) 12.9 = 2.7 nm. Macromolecular crystals of interest, such as protein crystals, typically have lattice constants in that range. The fact that diffraction patterns of crystals with such large lattice constants can be resolved is directly derived from the visibility of the spots in Figure 5.7, and not any information about source or beam parameters. The measured sizes of the spots are limited by the resolution of the detector; GPT simulations predict that the spot sizes should be as small as \( \sigma_d = 30 \) \( \mu m \). This implies that if the resolution of the detector is not a limiting factor, crystals with even larger lattice spacings could be studied, up to \( d = 19 \) nm. Furthermore, GPT simulations show that the beam size at the sample \( \sigma_{\text{smpl},1} \times \sigma_{\text{smpl},2} = 180 \times 260 \) \( \mu m^2 \). The \( d = 19 \) nm crystals could thus be less than 1 mm across and still provide resolvable diffraction patterns.

### 5.3.2 Diffraction experiments: temperature dependence

The image in 5.7a does not provide conclusive information about the actual beam quality, mainly due to the fact that the size of the spots is limited by the resolution of the MCP detector. To demonstrate that the high quality of diffraction patterns is a direct result of low temperature, the beam line is set up as shown in the upper schematic of Figure 5.6. Consequently, we set the currents of the magnetic lenses to those described by the ‘area of interest’ in Figure 4.3. To be exact, the beam currents are set to \( I_1 = 2.2 \) A and \( I_2 = 0.935 \) A, with a beam energy of \( U_e = 10.8 \) keV. As the schematic suggests, the spot sizes expand considerably. However, the spots remain visible, a direct consequence of the ultracold properties of the beam source.

To analyze diffraction experiments and compare to expected values from simulations and analytical models, the source parameters (effective temperature \( T \) and source size \( \sigma_{(x,y),i} \)) must be determined first. Using waist scans on a non-diffracting beam (see section 3.2.3), the source temperatures have been determined for varying ionization excess energies, for an acceleration potential \( V_{\text{acc}} = 23 \) keV. The resulting temperatures are shown in Figure 5.8, both as function of ionization laser wavelength \( \lambda_l \) and ionization excess energy \( E_{\text{exc}} \). At negative (mean) excess
energies, the source temperature reaches $\sim 10$ K. The uncertainty in the data points is partly due to a systematic error in fitting a waist scan. Added in the figure is the temperature calculated using the model presented in section 2.3, shown by the dashed red line. It is apparent that the model and measured temperature do not overlap very well. Some insight on possible reasons for this distinction is given in [28], though this has only been done for lower acceleration potentials ($V_{\text{acc}} = 6$ kV). It is possible that the model works less well for high acceleration voltages. Still, at low temperatures, the model is within the uncertainty range of the measured temperatures and at high temperatures, the slopes of the model curve and measured data are similar. The difference between the model and this measured data set has not further been investigated, and we will further take the measured data as the correct source temperature.

The source size, $\sigma_{x,i} \times \sigma_{y,i} = (32 \pm 2) \times (54 \pm 2)$ µm$^2$ has been determined by a space charge scan (see section 3.2.2). Diffraction images have been captured for various ionization laser wavelengths, $\lambda_l = 500 - 475$ nm, with the acceleration plates at a potential of $V_{\text{acc}} = 23$ kV ($U_e = 10.8$ keV). As with the sharp diffraction images, the femtosecond ionization laser has been used, resulting in picosecond electron bunches.

Two examples of diffraction images from this experimental set are shown in Figure 5.9a and c, for source temperatures of $T = 10$ and $T = 250$. Again, for illustration purposes, the presented images are an average of 10 separate images. As a guide to the eye, graphite’s expected hexagonal pattern is indicated by the thin lines. The spots are clearly much broader than in Figure 5.7. Looking at the spots more closely, in the inset of (b) and (d) and their respective line profiles, it is apparent that the size of the spot is smaller for a lower source temperature. Consequently, the low source temperature provides higher quality diffraction images, in terms of visibility. Two-dimensional Gaussian fits of the diffraction spots provide the sizes of the long and short axes. Only two of the six spots are fully within the detection area, being the top and bottom middle spots, so the presented spot sizes are an average of the sizes of those two. They are $\sigma_{d,1} \times \sigma_{d,2} = 1.8 \times 1.3$ mm$^2$ at $T = 250$ K, and $1.1 \times 0.9$ mm$^2$ at $T = 10$ K. The intra-spot distance, $s = 15.4 \pm 0.5$ mm, agrees with the value calculated using Bragg’s law, Eq. 4.9.

The diffraction spot sizes are plotted as a function of source temperature in Figure 5.10, where the two different sets represent the rms sizes of the short (green triangles) and the long (blue squares) axes of the elliptical diffraction spots. Each individual data point is the average of the spot sizes obtained from 10 separate images. The dotted line and surrounding gray band represents the spot size (and uncertainty) obtained from GPT simulations, using the measured source size
Figure 5.9: Examples of diffraction images for different source temperatures, obtained by placing the waist of a 10.8 keV electron beam on the sample. (a) and (c) show captured images for temperatures of $T = 250$ K and $T = 10$ K, respectively. In (b) and (d) a line profile along one axis of one of the spots (inset) is shown.

Figure 5.10: Measured diffraction spot sizes (green triangles and blue squares) as a function of effective source temperature. The gray bands are simulated spot sizes, using GPT.
and temperatures as parameters. The experimental data is in good agreement with simulated data. This shows that the spot sizes of the diffraction patterns behave as expected on the basis of source properties. This proves that accurate information about the beam properties can be extracted directly from diffraction images, unlike the diffraction rings, where a clear offset is present that has to be dealt with.

Despite a much better overlap between data and expected values, the match is not perfect. When comparing the individual measured spot sizes \( \sigma_{\text{det}} \) to those predicted by GPT, \( \sigma_{\text{GPT}} \), the average difference between the two is calculated as \( \langle \sigma_{\text{det}} - \sigma_{\text{GPT}} \rangle \) to be 100 \( \mu \)m for the long axis (blue data) and 30 \( \mu \)m for the short axis (green data). Looking at the spread in data, defined as \( \sqrt{\langle (\sigma_{\text{det}} - \sigma_{\text{GPT}})^2 \rangle} - \langle \sigma_{\text{det}} - \sigma_{\text{GPT}} \rangle^2 \), we find 110 \( \mu \)m for the long axis and 160 \( \mu \)m for the short axis. The data thus has systematic error compared to GPT data, most notably for the long axis. The short axis demonstrates more scatter. This scatter in the data set is attributed to beam jitter. This is most likely a result of pointing instabilities of the femtosecond ionization laser. The ionization laser propagates in the y direction (Figure 2.4), entering the MOT from above. Instabilities caused by this laser will thus result in unstable source positions, varying in the x and z direction. The x component of the source size is the short axis. It is therefore not surprising that the short axis data scatters more than that of the long axis.

### 5.3.3 Coherence length

Having confidence that the resulting spot sizes are a good representation of the beam quality, it is now instructive to quantify this. In Chapter 1 the term coherence length was introduced as a beam parameter that describes whether a diffraction pattern can be resolved or not. The coherence length is given by

\[
L_\perp \equiv \frac{\hbar}{m_e c} \frac{\sigma_x}{\epsilon_x^x},
\]  

(5.1)

with \( \sigma_x \) the rms beam size, and \( \epsilon_x \) the beam emittance, in this case taken in the x direction. At a beam waist, this can be rewritten as

\[
L_\perp = \frac{\hbar}{\sigma_{p_x}},
\]  

(5.2)

with \( \sigma_{p_x} \) the momentum spread. The momentum spread can be written in terms of angular momentum spread, \( \sigma_{\theta_x} = \sigma_{p_x} / p_z \). The longitudinal momentum \( p_z \) can then be expressed in terms of a diffraction angle, using Bragg’s law: \( p_z = \frac{2\pi \hbar}{d(\sin \theta)} \). Finally, the geometric relations under the small-angle approximation and infinite crystal approximation\(^1\) give \( 2\theta \approx s/h \) and \( \sigma_{\theta_x} \approx \sigma_d/h \) with \( s \) the intra-spot distance and \( h = 285 \) mm the distance between the sample and the detector. Combining all of this, the coherence length can be written as

\[
L_\perp = \frac{d_1 s}{2\pi \sigma_d}.
\]  

(5.3)

This equation tells us that the coherence length can be extracted directly from the information of a diffraction pattern, assuming that the waist of the beam is on the sample. No information about the source or the beam size at the sample, or, in fact, any prior beam information is required for this. The reasoning behind this seems obvious when considering the implication of the definition of coherence length: it is a term that describes the possible visibility of diffraction patterns. The coherence length should therefore be derivable from diffraction images.

Using Eq. 5.3, the coherence lengths as function of source temperature can be calculated using the data from Figure 5.10 and \( s = 15.4 \) mm, and is shown in Figure 5.11 (blue squares and green triangles). Also shown are the data from GPT simulations (dotted line ± gray area). The coherence length for a beam with \( T = 10 \) K source temperature is \( L_{\perp,1} = 0.65 \) nm and

---

\(^1\)The premise of this approximation is that the spot size at the detector is dominated by the angular spread, not the size of the beam at the sample, \( \sigma_{\text{sampl}} \). In other words, \( h\sigma_\theta \gg \sigma_{\text{sampl}} \). Although we haven’t said anything about \( \sigma_{\text{sampl}} \) up until now, we assure that this condition is met.
$L_{\perp,1} = 0.48 \text{ nm}$. Increasing the source temperature from 10 K to 300 K results in a reduction in coherence length by a factor of 1.5. We can thus visibly see in terms of spot size, and physically understand in terms of coherence length, that an increase in temperature deteriorates the beam quality, needed for diffraction experiments.

The set-up does not yet have a method for measuring the beam size at the sample $\sigma_{\text{smpl}}$ with $\mu$m precision. To determine values of $\sigma_{\text{smpl}}$ we use GPT simulations. Alternatively, from the measured coherence length it is possible to estimate the beam size at the sample as function of temperature, using Eq. 5.1 and measured source parameters, assuming that conservation of emittance applies. The resulting $\sigma_{\text{smpl}}$ using both of these methods are plotted in Figure 5.12. The calculated spot size at the sample is shown as function of source temperature. Here we have taken as source size the geometrical average of the calculated sizes, $\sqrt{\sigma_{\text{smpl,1}}\sigma_{\text{smpl,2}}}$. The curve shows the simulated size. The good overlap between the two data sets suggests that indeed, beam emittance is conserved. As is the case with the previous two graphs, there is some scattering around the expected value. Again, this is attributed to beam jitter caused mainly by instabilities in the femtosecond ionization laser.
5.3.4 Implications

At 10 K and 300 K the beam size at the sample is 3.3 µm and 9.1 µm, respectively. This is consistent with the decrease in coherence length, which follows from $\sqrt{T} \propto \sigma_{\text{smpl}}/L_{\perp}$. This too is evidence that our simulation models are accurate, emittance is conserved, and the detected diffraction patterns are representative of the beam parameters.

The coherence lengths presented here are only a few times larger than graphite’s lattice constant. Of course, as mentioned, we have intentionally increased the angular momentum at the sample in order to emphasise the importance of the low source temperature. This allows us to make extrapolations to theoretical limits in our set-up. For example, bringing down the source size by an order of magnitude to about 3 µm will allow us to bring the beam size at the sample down to $\sigma_{\text{smpl}} \approx 300$ nm, while maintaining the same coherence length. This implies that the UCP set-up can be used for time-resolved nanodiffraction, used for samples that are severely size-limited. This is only possible with a 10 K source due to its high coherence. The conventional $T \geq 1000$ K photocathode electron sources will not even be able to resolve the simplest of crystals under these conditions. Additionally, using a 3 µm source size makes time-resolved diffraction of micron-sized protein crystals possible.

Whatever the beam configuration, through our diffraction experiments we have effectively shown that the UCP set-up allows for an improvement of an order of magnitude, compared to conventional planar photocathodes, in either low beam size at the sample, bringing us closer to the realm of nanodiffraction, or in coherence length, allowing for the study of (macro)molecular crystals with lattice distances of $d > 1$ nm.
Chapter 6

Discussion and Conclusions

In this thesis we have presented and analyzed diffraction patterns using an ultrafast electron source. In diffraction experiments, the beam is quantified in terms of the parameter coherence length, $L_\perp$, with the requirement that it is larger than the lattice spacing of the sample under investigation. In order to improve the coherence length for ultrafast electron diffraction (UED) experiments, an ultracold electron source is used to decrease the initial angular spread of the electron beam. At CQT, such an ultracold charged particle set-up has been designed. Here we provide evidence that indeed, a cold source improves beam quality and makes ultrafast diffraction experiments of crystals with large lattice spacings possible. Previously, the properties of the beam have been quantified with methods presented in Chapter 3. Using these parameters, the coherence length of the ultracold beam has been inferred. However, this large coherence length, due to a low source temperature, has not been directly demonstrated in diffraction experiments. The goals of this research have been to improve the set-up and methods in order to enable diffraction experiments, and then employ such experiments to investigate the beam quality. In this chapter, first a synopsis of to what extent these goals have been reached will be given. Secondly, we discuss the implications of the results presented in Chapter 5. Suggestions for further measurements and experimental improvements are given. Finally, we look at the future prospects of ultracold UED.

6.1 Research goals

6.1.1 Set-up improvements

In the span of the project, the experimental set-up and methods have been improved in order to extend experimental possibilities and improve analysis of electron diffraction on graphite. One of the most crucial experimental requirements for diffraction is that the position of the beam at the sample, which is typically very small, should not move due to changes in source temperature, or lens parameters. The additions and changes made during this research is summarized below:

- Two alignment plates have been introduced in the beam line to create a more centralized beam path. The inclusion of the metallic alignment plates have played an important role in better understanding and modifying the beam path. Firstly, it is now possible to ionize rubidium atoms accurately from the radial center of the accelerator. Secondly, ensuring that electrons are centrally steered through the beam pipe is a lot less complicated. Also conveniently, the sample holder is directly below the second alignment crosshair. This gives the advantage of being able to align the electron beam onto the crosshair, then translating the plate on which it is, upwards, after which the beam is automatically aligned on the sample.

- A method of lowering the position of the MOT without using Helmholtz coils was implemented, using extra lenses with variable position to be able to tune the returning trapping laser intensity. Positioning the MOT can now be done with a higher degree of control.
• A highly effective addition to doing diffraction experiments was including the second magnetic lens. The advantage of this has thoroughly been discussed in Chapters 4 and 5. Using these lenses, the position of the waist and the angular spread of the beam can be controlled.

• The ionization laser beam’s drift is corrected for by placing an electronically adjustable mirror near the beginning of the laser beam, and a camera near the end. A simple control loop adjusts the mirror position based on the beam position detected at the camera, ensuring that the beam spot is kept at the same position. While we still see the clear effects of jitter due to quicker instabilities, the overall beam drift is now successfully corrected, keeping the position of the ionization volume constant at longer time scales. This is a necessary improvement, since diffraction experiments can typically last hours.

• A modest new development was replacing the old beam block with a more sensible design. The thin stem blocks a minimal part of the beam, while the central part is still large enough to block the 0th order beam. This is especially an improvement for ring patterns and broad spots, for which the previous beam block obstructed large portions of the interference pattern.

• An addition that has not been utilized in this work, but can be of assistance in later experiments is the addition of two slanted sharp edges in the plate that holds the sample. The change in intensity of a beam at the detector is monitored while scanning the edges (with minimum steps of 0.25 µm) vertically across the beam. The beam size at the sample can be determined from the resulting intensity as function of plate position. Due to beam instabilities it is not yet accurate enough to measure spot sizes under 100 µm.

• The method of analyzing a space charge scan to determine the source size has been improved. The measured data is compared to simulation data, instead of being fitted with an analytical model, which has shown to be inaccurate in certain regimes.

With these modifications, stability, controllability, quality, and reproducibility have been improved in our set-up, enabling good quality diffraction experiments.

6.1.2 Experimental results

The electron beam has been used to investigate thin, poly- and monocrystalline, graphite samples, providing ring and spot patterns. While the radii of the diffraction rings match the theory, the measured widths contain inaccuracies and inconsistencies as a result of beam jitter and limitations in analysis. This has made it difficult to make quantitative statements about the quality of the incident beam. The ring widths are thus not a good representation of the actual high quality of the beam.

A method has been developed to fabricate thin (20–30 layer) monocrystalline graphite samples. Using such a sample, spot patterns were produced. It is possible to produce high visibility, sharp, spot patterns by focusing the electron beam onto the detector. With these patterns we have shown that the set-up as it is now is capable of producing diffraction patterns of samples with lattice constants up to 2.7 nm, provided that the sample is large enough (hundreds of microns) and that it is sufficiently thin to be able to transmit ~ 10 keV electrons. However, this result is a poor representation of the actual beam quality, as the measured spot sizes are limited by the resolution of the MCP detector.

In order to investigate the beam’s coherence properties and provide evidence for the improved beam quality due to a low source temperature, a two-lens set-up was used to focus the beam onto the sample. The diffraction spot patterns measured with the two-lens set-up do a much better job in matching the expected spot sizes, allowing us to extract beam quality information, most notably the coherence length at the sample. The coherence length in this case was obviously larger than the sample’s lattice spacing, but decreases with increasing temperature, coinciding with theoretical expectations. Combining the measured coherence length of 0.65 nm and with source properties, an estimate was made for the beam size at the sample. The fact that the values
were comparable to those found using GPT simulations gives further confidence that the beam line of the ultracold source behaves as theoretically predicted.

We have intentionally done measurements where the coherence length is not spectacular, as low as 0.5 nm at 10 K and 0.3 nm at 300 K. However, the consistency of the results with theoretical and simulated values allows us to extrapolate our results to predict diffraction visibility in other regimes. For example, if the source temperature had been increased even further, all the way up to 1000 K (comparable to the temperature of electrons produced by planar cathode photo-emission), the coherence length would be reduced to maximally \( L_\perp = 0.2 \) nm, and the beam size at the sample would be increased to more than \( \sigma_{\text{smpl}} = 10 \) µm. In this example, this is no longer sufficient to resolve graphite diffraction, because \( L_\perp < d = 0.2131 \).

To take a relevant example, we envision what is possible if we utilized the same experimental conditions used by Kirchner et al. [13]. In this work, electrons are emitted one-by-one from a gold-plated photo-cathode. The collection of electrons have an effective temperature of 1350 K. The source size used is \( \sigma_{\text{src}} = 3 \) µm and the size of the beam at the sample is \( \sigma_{\text{smpl}} = 77 \) µm. A coherence length of 20 nm has been obtained with this set-up. If we replace this source with an ultracold source, with \( T = 10 \) K, the coherence length would increase by \( 11.6 \times \), to > 200 nm. Alternatively, if the beam size at the sample was decreased to 0.7 µm, we could perform nanodiffraction experiments on size-limited samples, while still retaining a coherence length of 2 nm.

To conclude, we have provided direct experimental evidence that the ultracold source is a viable technique for ultrafast electron diffraction of micron-sized (macro)molecular crystals with large lattice spacings, \( d > 1 \) nm.

### 6.2 Outlook

The first ultracold diffraction images have been captured and analyzed, using picosecond electron bunches. However, there is still much to be done before such a source can be applied to time-resolved crystallography. In this section, a few recommendations for short-term research with the UCP set-up are covered, followed by some long term goals that should eventually result in reaching the goal of making biomolecular movies.

#### 6.2.1 Recommendations

Two major limitations in our set-up have been discussed in Chapter 5. These are beam jitter and detector resolution.

**Beam stability:** Beam stability is, for one, a very important factor to have under good control. The more diffraction is pushed to the limits in terms of waist size and coherence length, the more crucial beam stability becomes. In regular waist scans, beam jitter plays a minor role: each image is captured relatively quickly, and the spot size is typically much larger than the beam’s jitter-influenced displacement. However, in diffraction images, which tend to take longer to capture, jitter becomes more significant, not only in the diffraction pattern itself, but also the beam position on the sample. Especially when moving to smaller samples, the beam should not be allowed to drift away from its position on the sample. However, measurements where the position of an unfocused electron beam is monitored in time, have shown that the spot position can vary by hundreds of microns in the course of a measurement, typically tens of minutes to over an hour.

A strong recommendation is to try to further understand and control the mechanisms involved in beam instability. A major influence is the pointing instability of the ionization laser. It should be investigated to what extent a higher degree of control of the beam can be attained. For example, a more sensitive camera, coupled to the adjustable mirror, can be used to more accurately steer the beam at a shorter intervals. Decreasing the optical path of the laser beam should also result in higher positional stability. Furthermore, the output intensity of the OPA, which converts the 800 nm femtosecond laser pulse to a 472 – 533 nm pulse, also shows instabilities that are highly
(and non-linearly) dependent on the 800 nm input. A more systematic approach to aligning both the fs laser and the OPA can result in a more stable beam, both in position and intensity.

**Electron detector:** Another recommended improvement is to replace the electron detection system. Much sharper diffraction spots could be produced than had been done in Figure 5.7, but were limited by the resolution of the detector. Ideally, the detector should register electrons with micrometer accuracy, and with high sensitivity.

**Sample beam size:** In this thesis, the spot size has been determined through simulations and calculations based on conservation of emittance. However, it is desirable to be able to measure $\sigma_{\text{smpl}}$ directly. It has been mentioned that the experimental set-up does not include a diagnostic for accurately determining the spot size at the sample, for sizes of $\sigma_{\text{smpl}} < 100 \, \mu\text{m}$. The translation stage with the sample holder has a plate with sharp edges, which can be scanned across the beam to determine the spot size by monitoring the current at the detector. This method, however, requires edges that are sharp and straight, up to micrometer accuracy. The beam jitter must be negligible during measurement time, which is the current limit in attempting such a scan. Furthermore, it is difficult to derive the size in both dimensions and the orientation of the elliptical axes from such a scan, since the plate can only translate vertically. An alternative method is to place a high mesh (~2000) TEM grid at the position of the sample and image it using electrons. Based on the image, the size of the beam at the sample can be determined. This will, however, not work as a direct measurement of beam size when the beam waist is at the sample: by definition, position and angular spread of electrons in a beam are uncorrelated in a waist.

**Bunch length:** Further experimentation can be done on the bunch length, which provides a theoretical limit to the possible time resolution possible in ultrafast electron diffraction. An effective method to measure the bunch length is to use a streak cavity [59], which translates the length of a chirped bunch to a lateral momentum spread. Bunch length is thus effectively imaged onto the MCP detector. Streak cavities have proven to work successfully as bunch length detectors, and can be implemented in the UCP beam pipe. Additionally, from the bunch length the energy spread can be determined, which provides a theoretical basis for determining the longitudinal coherence length, invariably linked to spatial resolution of diffraction patterns.

**Source size scan:** the space charge model, used for determining the beam source size, should be improved. This model has not been a focus of this research, but should be investigated further for correctness. As an alternative to the space charge model for source size measurements, in this research the source size has been determined by comparing a space charge scan to a simulated scan with various source sizes. While such a method provides an accurate source size, it is time consuming. It is therefore useful to look into a deriving a more complete analytical model to use as a fit instead. Some information about the shortcomings of the old model can be found in Appendix B.

### 6.2.2 Ultracold and ultrafast electron diffraction: Outlook

The UCP set-up has been built as a proof of principle for ultracold UED. Some long term changes are required to actually use the ultracold source for ultrafast electron diffraction:

- An important aspect of UED is the use of pump-probe techniques. The ionization laser beam doubles as a pump beam, which modifies the sample. The ultrafast electron pulse is used to analyze these modifications at ultrashort time scales. The pump-probe technique is currently being studied in another UED set-up at CQT. One major goal of the group is to combine ultracold diffraction with pump-probe techniques.

- In some samples, a pump laser induces a non-reversible change. Also, a high-charge energy electron beam can cause damage to, for example, biological samples. For these reasons, achieving a diffraction pattern using only a single electron bunch is required in some cases. From experience we have seen that reasonable diffraction images of graphite require $10^5 - 10^6$ electrons. Achieving such amounts of electrons in a single bunch is thus an important criterion for capturing single-shot diffraction patterns. One possibility to reach such a bunch charge is to expand the ionization volume, but this comes at the cost of beam quality.
Another option is to increase the source density. The MOT particle density is currently $\sim 10^{16} \text{ m}^{-3}$, but can theoretically be increased to $\sim 10^{18} \text{ m}^{-3}$ [21]. In this case, a source size of $20^3 \mu \text{m}^3$ would be enough to achieve $10^5$ electrons per bunch. Of course, in this case space charge is non-negligible and will have to be dealt with. Also, a higher amount of electrons is required for the more complicated diffraction patterns of biomolecules, compared to graphite samples.

- Electrons in the UCP set-up can be accelerated to a modest maximum energy of 15 keV. We have already seen how this reflects on the stringent conditions for thickness of graphite samples. Many samples cannot be produced at comparable thicknesses. Therefore, electron energies should be $> 100$ keV. Simply placing such a DC field around a MOT is problematic (the MOT already undergoes visible changes when applying $V_{\text{acc}} \approx 20$ kV). Possibilities are being sought in fast-switching electric fields [60] or multi-stage accelerator systems.

Ultrafast electron diffraction still has some way to go before being able to reach the goal of time-resolved microscopy of protein membrane crystals. However, using an ultracold source effectively brings us one order of magnitude closer to this goal. This research has provided evidence that structure determination using diffraction is possible with such a source, and that the expected quality of diffraction patterns due to the low source temperature can be reached.
Bibliography


Appendix A

Submitted paper

The diffraction spot results presented in section 5.3 have been incorporated into a scientific paper, which has been submitted to Nature Communications. This manuscript can be found in the following pages.
Ultrafast electron diffraction using an ultracold source

M.W. van Mourik, W.J. Engelen, E.J.D. Vredenbregt, and O.J. Luiten
Department of Applied Physics, Eindhoven University of Technology,
P.O. Box 513, 5600 MB Eindhoven, The Netherlands

We present diffraction patterns from micron-sized areas of mono-crystalline graphite obtained with an ultracold and ultrafast electron source. We show that high spatial coherence is manifest in the visibility of the patterns even for picosecond bunches of appreciable charge, enabled by the extremely low source temperature (~10 K). For a larger, ~100 μm spot size on the sample, spatial coherence lengths >10 nm result, sufficient to resolve diffraction patterns of complex protein crystals. This makes the source ideal for ultrafast electron diffraction of complex macromolecular structures such as membrane proteins, in a regime unattainable by conventional photocathode sources. By further reducing the source size, sub-μm spot sizes on the sample become possible with spatial coherence lengths exceeding 1 nm, enabling ultrafast nano-diffraction for material science.

The fast pace at which the new field of ultrafast structural dynamics is currently evolving is largely due to spectacular developments in ultrafast X-ray [1–3] and electron [4–6] beams. A particularly interesting development is the ultracold electron source, which is based on near-threshold photo-ionization of a laser-cooled and trapped atomic gas [7–12]. Recently it was shown that the ultracold electron source can be operated at femtosecond timescales while, surprisingly, retaining its high spatial coherence [11, 12]. Here we present the first diffraction patterns produced by electron bunches generated by such an ultracold and ultrafast source. Even when focusing the electron beam to a micron-sized spot on a graphite sample, we maintain high-visibility diffraction patterns. This is not possible with femtosecond beams generated with conventional planar photocathodes, which lack the required coherence. This opens the door to new possibilities, such as few-shot femtosecond electron diffraction of membrane protein crystals and ultrafast nanodiffraction, with the prospect of real-time monitoring of biomolecular dynamics.

Typical ultrafast electron diffraction (UED) experiments are performed using a planar photocathode source [4], characterized by effective electron temperatures \( T \geq 1000 \text{ K} \). Kirchner et al. [13] have shown that by focusing the femtosecond photoemission laser to a small spot, the root-mean-square (rms) source size can be reduced to \( \sigma_{\text{source}} = 3 \mu \text{m} \). By combining this with an rms beam size at the sample of \( \sigma_{\text{sample}} = 77 \mu \text{m} \), they achieved sizeable coherence lengths,

\[
L_\perp = \frac{h}{\sqrt{k_B T} \sigma_{\text{source}}} \approx 20 \text{ nm},
\]

with \( h \) Dirac’s constant, \( m \) the electron mass and \( k_B \) Boltzmann’s constant. To resolve a diffraction pattern, \( L_\perp \) should be larger than the lattice spacing \( a \) of the sample under investigation. Therefore, \( L_\perp = 20 \text{ nm} \) is more than sufficient for protein crystal diffraction (typically \( a = 1 - 5 \text{ nm} \)), as shown in Ref. [13] on an organic salt with \( a \approx 1 \text{ nm} \). Unfortunately, the crystallites in thin-film samples for transmission experiments are often limited to (sub-)micron sizes due to limitations in sample synthesis. Conventional photocathode sources cannot attain the required coherence length for \( \sigma_{\text{sample}} \leq 1 \mu \text{m} \).

The ultracold and ultrafast electron source, previously shown to have source temperatures as low as 10 K [11, 12], provides a means to realize ultrafast diffraction on (sub-)micron sized samples.

Fig. 1 shows a schematic overview of the set-up, which is described in more detail in Refs. [9, 14]. Electrons are created by near-threshold photoionization of a laser-cooled and trapped cloud of \(^{85}\text{Rb} \) atoms. Rubidium atoms are first excited from the 5s to the 5p state and subsequently ionized by \( a \leq 100 \text{ fs} \) full-width-at-half-maximum (FWHM) long laser pulse with a tunable central wavelength \( \lambda_l \) (Fig 1a). Typically, a few hundred electrons are produced per shot. The laser-cooled gas cloud is trapped inside an accelerator structure (Fig. 1b). Electrons are extracted from the cloud by an electric field with strength \( F \), and are accelerated to a final energy \( U = eF d_{\text{acc}} \), with \( e \) the elementary charge, and \( d_{\text{acc}} = 12.7 \text{ mm} \). The combination of \( \lambda_l \) and \( F \) determines the kinetic energy distribution of the released electrons, and thus the effective source temperature \( T \) [14]. Using the waist scan method (see Methods and [11]) we have established that the source temperature \( T \) can be varied from 300 K to 10 K by tuning \( \lambda_l \) from 477 nm to 500 nm at \( F = 0.85 \text{ MV/m} \). The source size \( \sigma_{\text{source}} = 32 \pm 2 \text{ (54 ± 2) } \mu \text{m} \) has been measured by means of an ion space charge scan (see Methods and [14]). The combination of \( \sigma_{\text{source}} \) and \( T \) fully characterizes the source.

At 1.245 m from the source, the beam is sent through a 13 – 20 nm thick monocrystalline graphite sample [15] on a 200 mesh copper TEM grid. The detector is placed at a distance \( h = 0.285 \text{ m} \) from the sample. Two magnetic lenses (at 0.53 m and 1.12 m) provide control over the spot size and the angular spread of the beam on the sample. To obtain sharp diffraction patterns, we focus the beam on the detector, resulting in a converging beam going through a relatively large area on the sample. Alternatively, focusing the beam on the sample, as shown
in Fig. 1b, allows us to investigate the smallest spot size that can be used in ultrafast diffraction.

Our diffraction images are the result of $10^9$ shots acquired at a 100 Hz repetition rate. Each shot contains a few hundred electrons. For illustrative purposes the recorded diffraction patterns shown here are an average over 10 images. Analyses of data are, however, based on separate $10^9$-shot images.

Fig. 2a shows an electron diffraction pattern produced with the waist of the beam on the detector, using only the first lens. The pattern was recorded with beam parameters $U = 13.2$ keV and $\lambda_1 = 485$ nm.

Five of the six 1st order spots (1) of the expected hexagonal pattern are visible, centered around the beam block (3). The sixth 1st order spot is blocked by the stem of the beam block (4). The 1st order beamlets arrive at the detector at a distance of $s = 14.3$ mm from the central ($0^\text{th}$ order) beam. The 1st order diffraction angle is $\theta = \tan^{-1}(s/h) = 50 \pm 1$ mrad, in excellent agreement with the theoretical value from Bragg’s law, $\theta = \sin^{-1}(\lambda_c/a_1) = 49.9$ mrad. Here $\lambda_c = 2\pi\hbar/\sqrt{2mU}$ is the electron’s De Broglie wavelength, and $a_1 = 0.2131$ nm is the first order lattice constant of graphite.

The rms spot size on the detector (magnified and profiled in Fig. 2b) is measured to be $\sigma_{d,x(y)} = 180 (210)$ $\mu$m, with x (y) the minor (major) axis of the elliptical spot. The diffraction spots can be brought 12.9 times closer to each other before spot visibility decreases to 88%. This implies that we could resolve diffraction patterns with lattice distances 12.9 times larger than that of graphite, such as (macro)molecular crystals, with $a$ up to 2.7 nm. The size $\sigma_a$ of the diffraction spot is actually expected to be as small as 30 $\mu$m, on the basis of measured source temperature and size, but is limited by the detector resolution. Had this not been an issue, crystals with even larger lattice spacings could be studied, up to $a = 19$ nm.

We wish to unambiguously demonstrate the full quality of the beam without being limited by detector resolution. Therefore, we have done measurements with the beam focused to micron-sized spots on the sample (Fig. 1b). In this configuration diffraction spots expand to a much larger size, but remain distinguishable; a direct consequence of the low-temperature properties of the source. For an electron energy $U = 10.8$ keV, diffraction images have been taken for ionization laser wavelengths $\lambda_1 = 500 - 476$ nm ($T = 10 - 300$). From GPT particle tracking simulations [16], we find a spot size on the sample $\sigma_{\text{sample}} = 3.3 \mu$m for 10 K and $\sigma_{\text{sample}} = 8.9 \mu$m for 300 K.

Figs. 2c and e show two examples of diffraction images from this data set, at ionization laser wavelengths of 478 and 498 nm, respectively, corresponding to measured source temperatures of $T = 250$ and 10 K. The thin gray lines are guides to show the hexagonal diffraction pattern. The spots inside the blue squares have been magnified and profiled in (d) ($T = 250$ K) and (f) ($T = 10$ K). Two-dimensional Gaussian fits are used to determine the size of the spots. For (d) and (f) these are $\sigma_{d,x(y)} = 1.8 (1.6)$ mm and $\sigma_{d,x(y)} = 1.1 (0.88)$ mm, respectively.

The diffraction spot sizes $\sigma_d$ are plotted as function of source temperature in Fig. 3, where the two sets represent the rms sizes of the short (green triangles) and long (blue squares) axes of the elliptical spots. Each individual data point is the average over spot sizes obtained from 10 diffraction images. The results are in good agreement with the values from particle tracking simulations (dotted line ± shaded area), which are based on the measured source temperature and spot size shown in Fig. 4 and the known electric and magnetic fields in the beam line [11]. This shows that the spot sizes of the diffraction patterns behave as expected on the basis of source properties.
scatter in the data points is attributed to pointing instabilities in the femtosecond ionization laser, which causes the position and size of the ionization volume, thus the final spot size, to vary.

It is instructive to discuss the results shown in Fig. 3 in terms of coherence length: since $\sigma_{\text{sample}} \ll \sigma_d$, $\sigma_d$ is dominated by the angular spread of the beam. This allows us to write $L_\perp$ as

$$L_\perp = \frac{a_1 s}{2\pi \sigma_d},$$

implying that $L_\perp$ can be determined directly from diffraction data, independent of the source parameters. For the 10 K data we thus find for the short (long) axis of the elliptical spot $L_\perp = 0.65$ (0.48) nm, which drops by a factor 1.5 for the 300 K data. GPT simulations show that $\sigma_{\text{sample}}$ increases by a factor 2.7 for an increase in $T$ from 10 K to 300 K. The values of $L_\perp$ calculated using Eq. (2) are therefore consistent with Eq. (1), i.e. $\sqrt{T} \propto \sigma_{\text{sample}}/L_\perp$.

In conclusion, we have produced sharp diffraction patterns using an ultracold and ultrafast electron source. From the quality of these patterns we infer that our ultracold source is suitable for ultrafast diffraction of macro-molecular crystals with large (> 1 nm) lattice constants. Furthermore, we have shown that we retain high visibility diffraction patterns even if we focus the beam down to a 3 $\mu$m spot size on the sample. By varying the source temperature we have shown that our results are consistent with theoretical and simulated models. We can thus extrapolate that for similar beam source and sample sizes, a high temperature ($T \geq 1000$ K) source would have a coherence length around 50 pm, no longer sufficient for diffraction of even the most basic crystals. We have thus shown the advantage of using an ultracold source over conventional photocathodes.

We have used a source size of a few tens of microns across, but this can, at least in principle, be reduced to the same 3 $\mu$m used by Kirchner et al. [13]. Combined with a source temperature of 10 K, this would enable us to focus the beam down to $\sigma_{\text{sample}} \approx 300$ nm, and to study sub-$\mu$m-sized samples, while still maintaining the visibility of Fig. 2e. In addition, for a 3 micron source size, time-resolved diffraction of micron-sized protein crystals becomes possible.

Methods

Ionization process Rubidium atoms are ionized in a two-step process: Rb atoms are excited from the 5$s$ to the 5$p$ state by an excitation laser pulse, and are subsequently ionized by a laser pulse with tunable ionization wavelength $\lambda_i$. The excitation and ionization laser pulses propagate along perpendicular directions and overlap in a well-defined region within the magneto-optical trap (MOT), resulting in an ionized cloud with
a volume governed by the two laser beam sizes. In the photo-ionization process, a few hundred electrons are released with an excess energy given by

$$E_{exc} = 2\pi hc \left( \frac{1}{\lambda_0} - \frac{1}{\lambda} \right) + 2E_h \sqrt{\frac{F}{F_0}} \quad (3)$$

with $c$ the speed of light, $\lambda_0 = 479.06$ nm, the zero-field ionization threshold wavelength, $E_h = 27.2$ eV the Hartree energy, $F$ the electric field strength inside the accelerator, and $F_0 = 5.14 \times 10^{11}$ V/m the atomic unit of field strength. The excess energy $E_{exc}$ of an absorbed photon is mostly transferred to kinetic energy of the released electron. Due to its broadbanded nature, a femtosecond pulse still has a possibility of ionizing even if the mean photon energy is below the required ionization energy.

**Source temperature** - In the paraxial approximation, electron trajectories at any point along the beam line can be modelled by a $2 \times 2$ transfer matrix $M$, which is a known function of magnetic and electric field strengths and positions of various lens elements present in the beam line. The final spot size $\sigma_d$ can be described in terms of source parameters as:

$$\sigma_d^2 = M_{11}\sigma_{source}^2 + M_{12}\sigma_{source}^2 \quad (4)$$

where $\sigma_{source}$ and $\sigma_{source}$ are the root-mean-square (rms) source size and angular spread, respectively. The angular spread at the source is related to the electron temperature according to $\sigma_{\vartheta,source} = \sqrt{k_B T/2U}$. The values of $M_{11}, M_{12}$ are determined via ray tracing models, and $\sigma_d$ is measured at the detector. In the so-called waist scan method, $\sigma_d$ is measured as function of the focusing strength of a magnetic lens, thus changing the values of $M_{11}, M_{12},$ and $\sigma_d$. The resulting data are fitted to Eq. 4, yielding $\sigma_{\vartheta,source}$ and thus $T$. Fig. 4a shows temperatures determined using waist scans, for excitation laser wavelength $\lambda_i = 500 – 476$ nm and electric field strength $F = 0.85$ MV/m.

**Source size** - The source size is determined by means of an ion space charge scan, in which the spot size of an ion bunch is measured at the detector as function of bunch charge. The spot size is partly governed by the repulsive effects of space charge. Ions are used instead of electrons primarily because the former are negligibly heated during the ionization process, so that angular spread due to temperature can be ignored. We negligibly heated during the ionization process, so that anions primarily because the former are negligibly heated during the ionization process, so that angular spread due to temperature can be ignored. We negligibly heated during the ionization process, so that anions primarily because the former are negligibly heated during the ionization process, so that angular spread due to temperature can be ignored. We negligibly heated during the ionization process, so that anions…

![Diagram](image-url)

**FIG. 4. Source parameters.** (a) Effective transverse source temperature as function of ionization laser wavelength, for an electric field strength of $F = 850$ kV/m. For large wavelengths, temperatures reach $T \approx 10$ K. The uncertainty of the data points is partly due to a systematic error in fitting a waist scan. (b) Results of an ion space charge scan to determine $\sigma_{source}$. The spot size at the detector, in two dimensions (green triangles and blue squares), is shown as function of bunch charge. The inset shows the normalized residual between experimental and simulation data for various simulated source sizes. From this we determine the source size, of which simulated final spot sizes are also plotted (dotted lines).


Acknowledgements
This research is supported by the Dutch Technology Foundation STW, applied science division of NWO and the Technology Programme of the Ministry of Economic Affairs. Author contributions M.W.v.M. and W.J.E. performed the experiments and analysed the data; M.W.v.M. wrote the manuscript, with help from E.J.D.V. and O.J.L.; the project was conceived and supervised by E.J.D.V. and O.J.L.
Appendix B

Space charge

In Chapter 3, a method has been described for determining the beam’s source size. A space charge scan is done, where the resulting data is matched to data taken from GPT simulations. This method differs from methods presented earlier. Previously the expansion due to space charge was modelled as a defocusing element in the analytical ray tracing model. This method, however, is incomplete and fails to work in the spot size scan at low bunch charges. In this chapter, first a brief introduction of the old space charge model is given. Then we discuss the shortcomings of this model. Providing a more complete model is outside the scope of this thesis, but should be considered in future work.

The space charge model attempts to represent the effects of space charge as a transformation matrix working on the 2-dimensional vector \( \vec{x} \), defined by the position and momentum angle of particles in a beam (see 3.2.1). The premise of the model is as follows: suppose we have an ion on the outside edge of an ion cloud with density \( n_i \), placed on the x axis. The ion’s initial potential energy is given by

\[
V_i = \frac{1}{4\pi\varepsilon_0} \frac{q^2 N}{x} \quad \text{(B.1)}
\]

with \( \varepsilon_0 \) the dielectric constant, \( q \) the elementary charge, \( N = \frac{4}{3} \pi x^3 n_i \) the amount of ions in the cloud, and \( x \) the distance of the ion from the center of the cloud. Due to space charge, the cloud explodes, and the ion’s potential energy is converted to kinetic energy, \( K = V_i \). The final velocity of the ion \( v_x \) is then given by

\[
v_x = \sqrt{\frac{2q^2 n_i}{3\varepsilon_0 m_i}} x \quad \text{(B.2)}
\]

The non-relativistic angle \( \theta \) (with respect to the z axis) for an ion accelerated in the z direction with energy \( U \), is given by

\[
\theta = \frac{v_x}{v_z} = v_x \sqrt{\frac{m_i}{2U}} = \sqrt{\frac{q^2 n_i}{3\varepsilon_0 U}} x. \quad \text{(B.3)}
\]

The acquired angle is thus modelled as being proportional to the position \( x \). This transformation is familiar to us in the ray tracing matrix formalism, as a focusing element with focal length \( f_{sc} \) given by

\[
\frac{1}{f_{sc}} = -\sqrt{\frac{q^2 n_i}{3\varepsilon_0 U}}. \quad \text{(B.4)}
\]

This focal length has been used in the beam model. The corresponding focusing matrix is placed at position \( z = 0 \), working on the source, giving a final spot size at the detector of \( \sigma_d = (M_{11} + A \cdot \ldots \right) \).
\( \sqrt{n_i} \sigma_{\text{src}} \), with \( M_{11} \) and \( A \) known constants, based on the beam’s complete transfer matrix \( M_{\text{tot}} \). In a space charge scan, the value of \( n_i \) is changed by varying the intensity of the ionization laser using ND filters, \( n_i \propto 10^{-ND} \). For these varying bunch charges the final spot size, \( \sigma_d \) is measured. The model is fitted to the resulting data in order to find the initial source size \( \sigma_{\text{src}} \).

This model makes the assumption that ions instantly acquire the velocity of Eq. B.2. The requirement for this assumption to be valid is that the characteristic time of space charge expansion \( \tau \) is smaller than the time \( t \) it takes the bunch to reach the first lens element, the accelerator’s exit kick. This allows us to effectively place the model lens at the source. The characteristic space charge expansion time \( \tau \) is given by the (modified) inverse plasma frequency

\[
\tau = \frac{\sqrt{32m_i\epsilon_0}}{27n_iq^2}
\]  

(B.5)

The time it takes a rubidium ion to reach the exit of the accelerator is on the order of \( t \approx 10^{-6} \) s for \( V_{\text{acc}} = 6 \) kV. At about \( n_i \approx 5.8 \times 10^{13} \) m\(^{-3} \) we have \( t \approx \tau \). The initial particle densities \( n_i \) in a space charge scan are typically varied between \( 10^{11} \) and \( 10^{15} \) m\(^{-3} \). It is apparent that at the low charge density regime of a space charge scan, the model is inaccurate. At very low charge densities (\( \sim 10^{12} \) m\(^{-3} \)) this does not matter, since the effect of space charge is negligible anyway. However, around \( \sim 10^{14} \) m\(^{-3} \) space charge is non-negligible. Around this regime the model insufficiently describes the expansion due to space charge in the beam line.
Appendix C

Diffraction angle

An important part of the research presented in this thesis is based on the assumption that if a beam passes through a sample with a certain angular spread $\sigma_\theta$, that the same angular spread is expected from the diffracted beamlets. Based on the diffraction theory presented in Chapter 4, it is not trivial that this is the case. Here we present a derivation to show that as long as the angular spread of the incident beam is small, the spread remains unchanged in the outgoing beamlets.

Consider an electron passing through a graphite sample with initial wavenumber $\vec{k}_i$. The electron is moving in the $z$ direction, with a slight angular offset $\phi$ in the $y$ direction:

$$\vec{k}_i = \begin{pmatrix} k_{i,x} \\ k_{i,y} \\ k_{i,z} \end{pmatrix} = \begin{pmatrix} 0 \\ |k_i| \sin \phi \\ |k_i| \cos \phi \end{pmatrix}.$$  \hfill (C.1)

The electron undergoes diffraction, making the first order transition $\vec{g} = (0, g_2, 0)$ with $g_2 = \frac{4\pi}{\sqrt{3}a}$, and $a = 0.2346$ nm the real space crystal lattice constant. This is taken from Eq. 4.17, using $(h, k, l) = (0, 1, 0)$. An excitation error $\vec{s}$ is introduced in order to maintain conservation of energy. The outgoing wavenumber is then given by $\vec{k}_f = \vec{k}_i + \vec{g} + \vec{s}$. This is schematically shown in Figure C.1. Here the diffraction angle is represented by $\theta$ instead of $2\theta$ for convenience. The excitation error is parallel to the incoming wavenumber, so it can be written as $\vec{s} = \gamma \vec{k}_i$, with $\gamma$ a scalar constant. We thus have $\vec{k}_f = k_i(1 + \gamma) + s$. Or, writing out the separate vector components (ignoring the first):

$$k_{f,y} = (1 + \gamma)|\vec{k}_i| \sin \phi + g_2$$  \hfill (C.2)
$$k_{f,z} = (1 + \gamma)|\vec{k}_i| \cos \phi$$  \hfill (C.3)

A solution for $(1 + \gamma)$ can be found by requiring conservation of energy, $|\vec{k}_f| = |\vec{k}_i| = k$.

$$k_{f,y}^2 + k_{f,z}^2 = k^2 = ((1 + \gamma)k \sin \phi + g_2)^2 + ((1 + \gamma)k \cos \phi)^2$$  \hfill (C.4)

Solving this equation for $(1 + \gamma)$ and for convenience making the substitution $g_2/k = \alpha$ gives

$$1 + \gamma = -\alpha \sin \phi \pm \sqrt{\alpha^2 \sin^2 \phi - \alpha^2 + 1}.$$  \hfill (C.5)

In our case, only the + solution is physically relevant.
Now we can calculate the scattering angle $\theta$ using the vector cosine similarity:

$$\cos \theta = \frac{\vec{k}_i \cdot \vec{k}_f}{|\vec{k}_i||\vec{k}_f|}$$

$$= \frac{k_i^2(1 + \gamma) + k_{i,y}g_2 + k_{i,z}(1 + \gamma)}{k^2}$$

$$= \frac{k^2(1 + \gamma) \sin^2 \phi + k^2(1 + \gamma) \cos^2 \phi + kg_2 \sin \phi}{k^2}$$

$$= (1 + \gamma) + \alpha \sin \phi$$

Then filling in the definition of $(1 + \gamma)$ results in

$$\theta = \cos^{-1} \left( \sqrt{\alpha^2 \sin^2 \phi - \alpha^2 + 1} \right)$$

For small angles $\phi$, this can be expanded as

$$\theta = \theta_0 - \frac{\alpha^2}{2\sqrt{1 - \alpha^2}} \phi^2 + O(\phi^4).$$

Here $\theta_0$ is the diffraction angle for a beam entering at $\phi = 0$. For a $U_e = 10$ keV electron beam passing through graphite, the ratio $\alpha = 0.0096$. Since $\phi$ is typically in the order of milliradians at the sample, or less, the second term on the right hand side is very small, about $10^6$ times less than $\theta_0$ and can be ignored. This tells us that electrons in a beam with angular spread $\sigma_{\phi}$ will all be deflected by the same angle $\theta = \theta_0$. So, the diffracted outgoing beam will have the same angular spread $\sigma_{\phi}$ as the incoming beam.

Note that this derivation is based on an incoming beam’s angle, offset in the y direction. The same derivation can be done for the x direction, but we see by symmetry that in the case for an angle offset in the x direction, the deflection angle $\theta$ is always the same for all electrons, not only by the small angle approximation.