Shaped electron bunches from ultracold plasma

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Cover illustration: “The phase of The Force”. One step in the phase retrieval technique described in Ch. 5, for a cold atom cloud in the shape of Master Yoda.
“Ephemeral answers – eternal questions!”

– Dr. R. V. Sellwood
Abstract

This thesis presents the development of a new apparatus, and imaging techniques, used to produce shaped cold electron bunches from ultracold plasma (UCP). Due to their low temperature, cold electron bunches from UCP have the potential to provide a compact electron source with sufficient brightness and coherence to enable single-shot ultrafast diffractive imaging of nano-scale samples, such as bio-molecules.

To create the electron bunch, a cold (70 µK) cloud of $10^8$ rubidium-85 atoms was prepared in a magneto optical trap (MOT). Some of the atoms were excited to an intermediate state using a laser with a spatially varying intensity profile, then ionised by a second laser pulse. The shaped excitation laser allowed two-dimensional control of the electron density. The ionisation laser could, in principle, also be spatially varying, to allow three dimensional manipulation of the electron bunch shape.

Due to Coulomb repulsion between electrons within a bunch, the brightness of the source is critically dependent on its initial shape, and therefore the shape of the atom cloud. Uniform density bunches are particularly desirable due to their linear space-charge expansion. To optimise the source through production of uniform bunches, knowledge of the spatial density distribution of atoms in the cloud is required. Conventional imaging techniques for cold atoms are either technically demanding or destructive to the atom cloud, and are unsuitable for this application, providing direct motivation for the development of new methods.

I have developed two new imaging techniques for use in shaped electron bunch production. The new techniques are particularly suited to our application,
but are applicable to the cold atom research community in general, offering several advantages over conventional methods. The first method adapts a phase-contrast imaging technique to measure the spatial distribution of atoms in a specific excited state. The second approach allows single-shot imaging of inhomogeneous atom clouds; that is, where both the density and refractive index may be spatially varying. The method uses a perturbative approach in conjunction with phase retrieval based on the transport of intensity equation. This technique is also potentially valuable for studies of atomic coherence effects in cold atoms and was demonstrated using spatially modulated electromagnetically induced transparency.

In collaboration with other members of our research group and in parallel with the imaging research, I also designed and constructed a new apparatus to produce UCP. The experiment first produced electron bunches in late 2009, and using the imaging techniques I developed, the results of the first shaped electron bunches from UCP are presented here. Due to the low temperature of the electrons, such shaped bunches can only be produced and observed using a cold electron source. Conventional thermal sources, including photoemission and field emission sources, produce hot electrons whose high temperature immediately diffuses any initial structure. The effects of increased electron temperature on the quality of the bunch shape are investigated here, resulting in the conclusion that cold electrons are essential for observing, and thus optimising, shaped bunches. Arbitrarily shaped bunches are demonstrated for the first time, using an intensity shaped excitation laser beam followed by uniform ionisation of the excited atom distribution. The laser intensity profile is adjusted using a spatial light modulator (SLM). Variations in atomic density which would degrade the bunch shape are mea-
sured using the imaging techniques developed. Adjustment of the excitation laser intensity to compensate for atomic density is shown to produce uniform bunches in two dimensions. Quantitative analysis of the acuity of the bunch edge provides an upper limit to the electron temperature of $T = 35\,\text{K}$. Unlike photoemission sources, the electron bunch shaping mechanism demonstrated here can easily be generalised to three dimensions. In addition, field ionisation of Rydberg atoms is observed to play an important role in the ionisation process, and suggested as a future avenue of research.
Declaration

This is to certify that

1. the thesis comprises only my original work towards the PhD except where indicated in the Preface,

2. due acknowledgement has been made in the text to all other material used,

3. the thesis is less than 100,000 words in length, exclusive of tables, maps, bibliographies and appendices.

Signed,

David V Sheludko
I have been extraordinarily fortunate to work with A/Prof. Rob Scholten over the last several years. He has proven to be an endless source of inspiration and motivation, advice and guidance, without which this project would not have been even remotely possible. He is selfless and enthusiastic in his supervision, scientifically creative and yet invaluably realistic! I value his friendship greatly and wish him every continued success in the future. My supervisory panel members Dr. Jeff McCallum and Dr. Andy Martin have also both encouraged and supported my research and career development over the last four years, for which I am very grateful.

Andy McCulloch, Simon Bell, Mark Junker, Seb Saliba, Liam McGuinness, Alastair Stacey, Martijn Jasperse and Christoph Hofmann. These are the guys in the lab that you rely on to lend a hand aligning, fibre-coupling, greasing vacuum bolts, soldering PCBs and occasionally popping balloons with high power lasers and freezing mandarins in liquid nitrogen. We are lucky to work with such a great group of friends, and I appreciate all of your help and hard work over the years.

I am very grateful to Prof. Keith Nugent and the ARC Centre of Excellence for Coherent X-ray Science (CXS) for supporting this project, both financially and scientifically. Without their support, the Melbourne ultracold plasma would still be a sketch on a page.

My family have continually supported and encouraged me throughout my education. In particular, my parents have been incredibly patient with my perpetual student status over the last fourteen years at university. I promise
I’ll get a haircut and a “real” job and will now fix your computers!

My wife Alicia has put up with my constant grumbling about lasers, atoms and electronics, almost every night for the last four and a half years. When the vacuum viewports arrived without glass in them, she bought me a bottle of red and reminded me that it wasn’t the end of the world. When we first saw electrons, she drove me home from the “celebrations” that followed. I can’t begin to imagine a way to thank you for everything, but perhaps some slow-roasted pork belly would be a step in the right direction?
Contributions

The following work was contributed by the author:

- Review and analysis of current UCP literature and methods presented in Chapter 2.

- Design and CAD drawings of the UCP apparatus (Chapter 3).

- Design and assembly of the tapered amplifier laser, including engineering drawings and printed circuit board design.

- Significant contributions to the construction and bakeout of the vacuum apparatus.

- Creation of all experimental control software (Labview) and hardware interface programming (Perl, C) required for the project.

- Switching and timing electronics for magnetic and electric fields and laser control.

- Experimental measurements and images of atomic density and electron density presented in Chapter 3.

- Theoretical, computational and experimental adaptation of the imaging and phase retrieval technique presented in Chapter 5 to image cold atom clouds.

- Use of a three-level semiclassical (quantum master equation) model to enable imaging of electromagnetically induced transparency in cold atoms and extraction of spatially varying refractive index.
• Theoretical, computational and experimental work on excited state diffraction contrast imaging (Chapter 4).
The author thanks the following people for their significant contributions to the project, and published work included here:

- Simon Bell and Mark Junker designed and implemented the Zeeman slower which enhanced the loading rate and efficiency of the UCP significantly. They were also responsible for simulations and design of the charged particle accelerator structure, and assisted during assembly and bakeout of the UCP vacuum apparatus. Simon Bell also developed the 776 nm laser and frequency locking scheme used to demonstrate the new imaging technique in Ch. 4.

- Andy McCulloch provided assistance with image acquisition and experimental work during testing of the SCPI method using electromagnetically induced transparency (Ch. 5). His work on characterising the behaviour of the spatial light modulator used in Ch. 3 enabled and enhanced the bunch shaping results presented there. He was also instrumental in the construction of the apparatus.

- Discussions with Martijn Jasperse during theoretical work on modelling electromagnetically induced transparency in a magneto optical trap led to the theoretical predictions of refractive index in Fig. 5.5.

- Russell Anderson provided the analytical result for the refractive index of a three-level atom used in Ch. 4.

- Christoph Hofmann assisted with early work to set up the magneto-optical trap used in Ch. 4 and the spatial light modulator used to produce shaped laser beams.
• Harry Quiney provided valuable advice during the work to adapt his SCPI method to cold atom imaging (Ch. 5).

• Edgar Vredenburg and his research group at Technische Universiteit Eindhoven, The Netherlands for providing guidance, insight and feedback on the design of our new apparatus. Discussions with A/Prof. Vredenburg also led to the explanation of spatially dependent Zeeman shifts observed in excited state atom clouds in Ch. 4.
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Chapter 1

Introduction

Research into ultracold plasma (UCP) offers unique insights into both fundamental and applied physics. Plasma is only produced when there is sufficient energy (for example, thermal energy) to separate atoms and molecules into positive ions and negative electrons. Most often, ionisation occurs when thermal energy exceeds the binding energy, creating a weakly coupled plasma. In weakly coupled plasma, the energy of the Coulomb attraction between the charged particles is weaker than their thermal energy. This type of plasma is commonly found in fluorescent light tubes.

In contrast, ultracold plasmas offer a method to produce strongly coupled plasma, through photoionisation of an ultracold atom cloud [1]. The recent advent of cheap, reliable sources of laser light has enabled rapid development of methods to cool and trap atoms. One such technique is a magneto-optical trap (MOT) [2], in which a combination of laser beams and magnetic fields are used to trap and cool a cloud of atoms to sub-millikelvin temperatures. Photoionisation of the ultracold atoms using a narrow-linewidth laser pulse, such that the atoms are just barely ionised, results in cold electrons and ions. With careful adjustment of the ionisation process, the resulting cloud of cold ions and electrons can have temperatures as low as a few Kelvin [3]. With
such a low thermal energy, ultracold plasma approaches the strongly coupled regime.

The physics of strongly coupled plasmas is a dynamic new frontier of physics research. Dynamical processes such as the formation of Wigner crystals [4,5], propagation of plasma waves [6,7] and plasma heating mechanisms provide a clear motivation for research of ultracold plasma. Electrons in ultracold plasmas have been suggested as an analogous laboratory-based system in which to study star cluster dynamics [8]. Additionally, the potential applications of cold electrons and ions extracted from the plasma have resulted in a burgeoning tide of interest in the field over the last decade. For example, ultracold plasma may provide sufficiently bright electron bunches to enable single shot diffractive imaging of biomolecules [9], while cold ions can be used to improve resolution in industrial applications such as focused ion beam milling and semiconductor fabrication [10].

While recent UCP simulations [11,12] and experiments [13] indicate that the electron bunch brightness may be sufficient for single-shot diffractive imaging, the brightness is critically dependent on the initial electron distribution, or bunch shape. Since we are producing the electrons via photoionisation of cold atoms, the distribution of cold atoms is critical. This project develops two methods of imaging the distribution of cold atoms for direct use in shaping electron bunches produced from UCP. The techniques are particularly suited to our application, but are applicable to imaging of cold atoms in general.

A new ultracold plasma experiment was designed and constructed at the University of Melbourne. The new imaging techniques presented in this thesis were used to determine the spatial atomic density distribution prior to
ionisation, enabling the first demonstration of shaped electron bunches from an ultracold plasma.

1.1 Motivation

Our primary interest in development of an ultracold plasma is the production of cold electrons for use in diffractive imaging of, for example, biological molecules and nanometre scale defects in solid state devices. With sufficient electron bunch brightness, ultrafast time resolved imaging could even reveal important information about the structural dynamics of membrane proteins. Determination of the structure and function of membrane proteins is critical for biological applications, including rational drug design and disease analysis [14]. Nutrients, viruses and toxins enter the cell through structures controlled by membrane proteins. Understanding the structure of the proteins allows the design of targeted drugs to block or allow passage through the cell membrane on a selective basis.

Numerous techniques exist to assist in determining the structure of these molecules, such as x-ray crystallography and nuclear magnetic resonance [15, 16]. Since the late 1950s, x-ray crystallography [15] has provided a means of extracting structural information on an atomic level from ordered protein crystals. Unfortunately, most membrane crystals are not easily crystallised. Since crystallographic techniques rely on the periodic lattice structure of the crystal to enhance the signal to noise ratio of a diffracted intensity pattern, they are not easily adapted to membrane proteins.

Coherent diffractive imaging (CDI) presents an alternative technique, in
which the object is placed in an intense x-ray beam and the diffracted intensity is measured on a detector. To determine the transmission function of the object, the phase of the wave at the detector is found via iterative techniques [17] and back-propagation yields the exit surface wave at the object. Extensive work on improving the resolution [18,19], computation time [20,21] and relaxation of experimental conditions [22,23,24] continues to develop the technique.

While great progress has been made in adapting CDI to imaging of biological structures [25,26,27], the inherently weak strength of the interaction between x-rays and matter leads inevitably to the requirement of extremely high brightness sources of x-rays. Accordingly, x-ray based imaging currently requires access to synchrotrons or X-ray Free Electron Lasers (XFELs). Recent developments in high-harmonic generation may potentially offer a more compact source of high brightness x-rays in future, but the limitations imposed by the weak interaction strength remain, resulting in long experimental runs and challenging statistical post-processing of data.

In contrast, the interaction strength for electrons is approximately ten thousand times stronger than x-rays, offering massive gains for diffractive imaging applications. In addition, the field of electron optics is very advanced. However, the charge of the electron requires that Coulomb repulsion be carefully managed in order to maintain the brightness and coherence of the electrons. In conventional electron imaging, this is normally achieved by using very low electron fluxes, such that space-charge effects can be neglected. Conventional electron imaging is therefore a slow process, preventing its use for “snapshot” imaging of dynamic processes.
More recently, 4D ultrafast electron microscopy (UEM) [28] has offered the possibility of femtosecond time resolution through generation of ultrafast (< 100 fs) electron bunches. In UEM, a femtosecond laser pulse is incident on a photocathode to generate the electron bunch. To avoid space-charge expansion of the bunch, pulses typically contain an average of one electron per pulse [29]. The same laser is used to “pump” the sample into a well-defined state for imaging, with the delay time between pump pulse and incident probe electrons very well defined. Using such pump-probe techniques, the sample can be reliably prepared into the same state prior to imaging on each shot. Coupled with the short temporal bunch length, this enables exposure times of several seconds to still provide “snapshot” images of the sample in a particular dynamic state. The technique has primarily been demonstrated on solid state samples [30] which exhibit both high damage threshold and long term stability. Although the technique represents a valuable tool for investigation of dynamic processes in such systems, its application to biological molecules is challenging due to motion in the sample during the long exposure time, and the effects of damage to the sample [31].

Single-shot diffractive imaging using electrons (Fig. 1.1) requires an electron bunch to propagate through the sample, meaning that space-charge effects must be taken into account. Space-charge expansion of an electron bunch is, in general, non-linear. Consequently, such expansion cannot be reversed with conventional linear electron optics. The unique production mechanism of ultracold plasma offers the opportunity to produce electron bunches that are shaped in three dimensions, such that their space-charge expansion can remain linear [9], and can therefore be reversed. With sufficient brightness and coherence, shaped electron bunches can then be used for the “holy grail”
Figure 1.1: Single-shot diffractive electron imaging of biomolecules. A uniform ellipsoidal electron bunch is incident on the sample, resulting in a diffraction pattern on the detector which can be inverted computationally to determine the structure of the sample.


To create shaped electron bunches from UCP, we require information about the spatial distribution of cold atoms, from which the electrons are produced. Conventional cold atom imaging techniques are limited in their application to complex multi-level systems involving many laser fields, necessitating the development of new imaging methods for our application.

While the imaging methods developed here were motivated primarily by their suitability to cold electron production, they are widely applicable to cold atom experiments in general. For example, studies in atomic coherence phenomena such as electromagnetically induced transparency (EIT) [32], coherent frequency upconversion [33], generation of entangled states in the Rydberg blockade regime [34, 35], and “slow light” [36] in a magneto-optical trap (MOT) have typically been studied using techniques without spatial resolution. Phase imaging offers the potential for obtaining additional information on these physical effects. Capture and storage of three dimensional
light fields using EIT [37] could benefit from imaging to provide spatial information about the atomic coherence of the atoms involved. Combining diffraction-based phase imaging with control of the internal state of the imaged atoms using a probe laser, we could potentially explore techniques for enhancing the imaging (e.g. by modifying the refractive index), or for directly measuring the control process itself. In addition to their use in the UCP project, the new phase imaging techniques presented here enabled single-shot measurements of the spatial variation in refractive index of a cold atom cloud. The refractive index was modified using a spatially modulated EIT scheme (Ch. 5).

1.2 Electron production

The project described in this thesis provides the basis for a pulsed ultrabright electron beam for diffractive imaging. The brightness of electron beams can be expressed in terms of the transverse brightness, \( B_\perp \) [9]:

\[
B_\perp = \frac{mc^2 J}{2\pi k_B T_e}
\]  

(1.1)

where \( m \) is the electron mass, \( J \) is the peak current density, \( k_B \) is the Boltzmann constant, \( c \) is the speed of light and \( T \) is the electron temperature. The brightness of non-relativistic sources, such as those used in electron microscopy, is often expressed in terms of the reduced brightness, \( B_r \). For comparison, it is related to the transverse brightness by [38]

\[
B_r = \frac{2eB_\perp}{mc^2} = \frac{eJ}{\pi k_B T_e}
\]  

(1.2)
where \( e \) is the electron charge. It is clear from equations 1.1 and 1.2 that increased brightness can result from either increased current density, or reduced electron temperature.

Intrinsically linked with the brightness of the beam is the normalised r.m.s. emittance, \( \varepsilon \), which indicates the focusability of the beam. The emittance in the transverse direction \((x)\) is given by

\[
\varepsilon_x = \frac{1}{mc} \sqrt{\langle x^2 \rangle \langle p_x^2 \rangle - \langle xp_x \rangle^2}
\]  

where \( \langle \rangle \) indicates averaging over the distribution, \( m \) is the electron mass, \( c \) is the speed of light and \( p_x \) is the momentum in the \( x \)-direction. For a given beam divergence, a lower emittance value indicates a smaller achievable spot size. From Eq. 1.3 we can see that a lower emittance corresponds to a smaller phase-space distribution, and higher phase space density. Contributions to the emittance at the target arise from two sources: the intrinsic emittance which is a characteristic of the source, and emittance growth due to nonlinear expansion of the bunch as it propagates to the target.

At the source, there is no correlation between momentum and position, such that \( \langle xp_x \rangle^2 = 0 \) and the emittance can be re-written as

\[
\varepsilon_x = \frac{1}{mc} \sigma_x \sigma_{p_x}
\]  

where \( \sigma_x \) and \( \sigma_{p_x} \) are the transverse r.m.s. beam size and momentum spread, respectively. For a thermal distribution, \( \sigma_{p_x} = \sqrt{mkT} \) so that the emittance becomes

\[
\varepsilon_x = \sigma_x \sqrt{\frac{k_B T}{mc^2}}.
\]
Recalling that the current density is related to the peak current $I$ by

$$J = \frac{I}{2\pi \sigma_x \sigma_y},$$

(1.6)

the transverse brightness of the beam (Eq. 1.1) can now be equivalently expressed in terms of the emittance:

$$B_{\perp} = \frac{I}{4\pi^2 \epsilon_x \epsilon_y}.$$  \hspace{1cm} (1.7)

We seek to minimise the emittance, thus maximising the transverse brightness of the source.

Current state-of-the-art sources [39, 40] increase brightness by reducing the emission area. For example, photofield emission from needle cathodes have recently demonstrated high currents (150 pC, 2.9 A peak current [40]). A source temperature of the order $10^4$K [41, 42], coupled with an emission area of only 50 $\mu$m results in a transverse brightness of order $10^{13}$ A/m$^2$sr. Carbon nanotube (CNT) field emission sources reduce the emission area even further: transverse brightnesses of up to $8 \times 10^{14}$ A/m$^2$sr have been reported [43, 38], however these sources are limited to operation in the regime of very small (nA) currents due to very small emission area, typically only a few nanometers wide.

1.2.1 Emittance growth due to space-charge repulsion

In contrast to the sources mentioned above, electrons produced from ultracold plasma can have temperatures of just a few degrees Kelvin, offering the potential for massive gains in brightness if comparable current densities can
be achieved. While simulations have indicated that these gains are achievable [9, 44], they rely on control of the initial bunch shape to prevent emittance growth due to space-charge (Coulomb) repulsion as the bunch propagates.

To illustrate this point, consider the evolution of an electron bunch with Gaussian initial density distribution (Fig. 1.2, top). The electric field at the center of the bunch is zero due to symmetry. However, the electrons near the center of the bunch will experience greater Coulomb repulsion than the outer areas, due to the higher electron (and hence charge) density. These electrons will therefore acquire more momentum and diverge more rapidly than electrons at the outer regions of the bunch. As the bunch propagates, the non-uniform electron density results in non-linear expansion of the bunch, and therefore unwanted emittance growth. In contrast, the evolution of a “waterbag” bunch (an ellipsoid with uniform initial density, Fig. 1.2, bottom) avoids the problem. For this bunch, the Coulomb repulsion is linear with position across the cloud so that the expansion remains linear, and emittance growth (but not expansion) is avoided. While the concept is most easily visualised for the waterbag distribution, other initial charge distributions also exhibit this desirable behaviour, since they spontaneously evolve into a uniform ellipsoid. In particular, a flat sheet of charge with half-circle radial charge density distribution (a “pancake” bunch) and an elongated “cigar” shaped bunch have both been shown to evolve to a uniform ellipsoidal charge distribution, and therefore exhibit only linear space-charge expansion [44].

In 2008, Musumeci et al. demonstrated production of pancake bunches using an intensity-shaped ultrafast (35 fs) laser pulse to illuminate the cathode on an RF photogun [45]. The bunch was shown to evolved under its own space-charge forces to a uniform ellipsoid, resulting in suppression of emittance.
growth and reversible Coulomb expansion. In addition, the use of ultrafast laser pulses creates ultrashort bunches, such as that which would be required for time resolved imaging. Although demonstrating impressive brightness ($10^{13}$ A/m$^2$sr) and low emittance, this technique remains limited by the temperature of the electrons produced by the laser pulse, typically thousands of degrees. Production of shaped bunches from a *cold* source has the potential to improve on these results even further.

**Figure 1.2:** Qualitative representation of the evolution of a Gaussian initial density distribution (top) and uniform ellipsoid (bottom). Momentum is represented by a colour scale. Although the central region of the Gaussian bunch remains roughly linear, it is clear that unlike the uniform ellipse, whose momentum remains linear with position all the way to the edge of the bunch, the Gaussian evolves to a non-linear momentum distribution.

### 1.2.2 Bunch shaping

To control the initial electron density produced from the UCP and thereby avoid emittance growth, the scheme outlined in Fig 1.3 has been proposed [9].
Beginning with a cold atom cloud with Gaussian density distribution, an excitation beam with controllable intensity profile excites a fraction of the atoms to an intermediate excited state, shaping the excited atom cloud in two dimensions. The ionisation laser, also with shaped intensity profile, photoionises only those atoms in the excited state, along an orthogonal axis. The overlap of the two intensity-shaped laser beams enables shaping of the electron bunch in three dimensions, and hence production of an ellipsoidal bunch with uniform initial density and minimal space-charge emittance growth.

![Diagram](image)

**Figure 1.3:** Proposed scheme for production of uniform density electron bunches. Two-step ionisation allows shaping of the electron bunch in three dimensions by shaping the overlap of the excitation and ionisation laser beams. The initially Gaussian distribution is made uniform by reducing the laser intensity in the center of both beams, illustrated in the plot of excitation intensity beam as a function of position.

### 1.3 Imaging cold atoms

The example shown in Fig. 1.3 assumes that the initial atom density is Gaussian. If the atomic distribution is *not* perfectly Gaussian (as is typically the case for experiments) then the intensity profile of the lasers has to be altered.
1.3 Imaging cold atoms

to take these density perturbations into account. To determine the correct intensity profile for the lasers, we require information about the specific spatial distribution of atoms in the MOT. To obtain this information, two new imaging techniques were developed, detailed in chapters 4 and 5.

The first imaging technique (Ch. 4) uses diffraction contrast imaging (DCI) to provide information about the distribution of excited atoms in the trap [46]. This information can be used to feedback to the profile of the ionisation laser. Although advantageous over conventional imaging techniques, DCI is limited in its application in that it cannot image inhomogeneous objects, i.e. those with inherent spatial variations in refractive index. DCI assumes that the refractive index varies only with the density of atoms, relying on this assumption to extract a map of atomic density. In many cold atom experiments, the atom cloud is illuminated by many lasers, each of which cause a spatial variation in the refractive index of the atoms.

The second imaging method (single-plane curved-beam phase imaging, SCPI, Ch. 5) demonstrates the application of a technique which is able to image such inhomogeneous objects via phase retrieval using the transport of intensity equation (TIE) [47]. Retrieval of either the atomic density or the spatial variation in refractive index is possible, provided that the other is known, making this technique particularly valuable for atomic coherence experiments in cold atoms.

The imaging techniques were demonstrated using an existing magneto optical trap, while the ultracold plasma apparatus was designed and constructed in parallel. The imaging techniques were applied to the new apparatus after construction.
1.4 UCP experiment

Ultracold plasma is a new venture at the University of Melbourne. At the commencement of this project, no apparatus or laboratory space existed for the experiment. Chapter 3 details the design and construction of the new University of Melbourne UCP laboratory and apparatus together with initial experimental results from the ultracold plasma.

![Conceptual diagram of the UCP experiment.](image)

**Figure 1.4:** Conceptual diagram of the UCP experiment. Hot atoms effuse from the rubidium oven (right), and are slowed before being cooled and trapped in a magneto optical trap. The atoms are ionised via a two-step process before electrons (or ions) are extracted with a parallel plate accelerator.

In building the new experiment, the design of a new ultrahigh vacuum (UHV) system was of primary concern due to the long manufacturing time required to produce customised UHV components. Plans for the new UCP system progressed from the conceptual outline of the experiment, explained via Fig. 5.1. Hot rubidium atoms effuse from an oven and are slowed as they travel to the main experimental chamber. The atoms are cooled and trapped using a magneto optical trap [2]. The atoms are imaged to determine their spatial distribution, before a shaped excitation beam excites a fraction of the atoms
to the 5P state (see Fig. 1.5). The excited atoms are imaged on the 776 nm transition, and ionised using a 480 nm pulsed laser. An electric field between parallel electrodes extracts the electrons and ions in opposite directions for diagnostic analysis, and ultimately for the applications described above.

![Energy level diagram for rubidium](image)

**Figure 1.5:** Simplified energy level diagram for rubidium, indicating transitions relevant to production of cold electrons from UCP as shown in Fig 1.4. The 420 nm transition can also provide a measurement of the excited state fraction via fluorescence imaging, as demonstrated in Ch. 4.

The project to produce bright electron bunches depends on UCP. UCP is a new field, with tremendous developments since its inception by Rolston and Killian in 1999. An appreciation of the physics of UCP is clearly important to the project. Chapter 2 reviews the relevant aspects of the field and our understanding of UCP.
Chapter 2

Ultracold Plasma

2.1 Introduction

The first ultracold neutral plasma was produced using laser-cooled xenon atoms in 1999 [1], triggering rapid development and interest in this new and exciting field. Funding of the Melbourne UCP system commenced in 2005, design of the experimental apparatus in 2007, and the first electrons and ions were extracted in late 2009. With such rapid development of a new and complex experimental apparatus, a plethora of new physical concepts were encountered in a relatively short period. During the design phase in particular, these concepts were critical to ensuring successful operation of the new UCP system, so a brief overview of the relevant ideas is presented in Secs. 2.2 and 2.3.

We begin by examining the defining characteristics of a plasma, and investigate how cold plasma differs from more familiar hot plasma through a brief overview of strong coupling. During the first experiments to produce cold plasma, the dominant cause of plasma heating was assumed to be excess energy from the ionisation laser, which could easily be reduced to the point where strong coupling should be observed. However, the signatures of
strongly coupled plasmas proved more elusive than expected [1, 48], which provoked thought that further investigation of the heating processes was re-
quired. Indeed, as the plasma evolves, several heating mechanisms alter
the behaviour of the plasma, and limit the achievable temperature of the
extracted electrons [49]. Since our application also depends on this temper-
ature, an understanding of the limiting mechanisms is required in order to
minimise their effects.
2.2 Plasma Fundamentals

2.2.1 Debye Shielding

Chen [7] described plasma as “a quasineutral gas of charged and neutral particles which exhibits collective behaviour”. Mathematically, “quasineutrality” is defined in terms of the Debye length, $\lambda_D$, and the dimensions of the system, $L$:

$$\lambda_D << L \quad (2.1)$$

where

$$\lambda_D = \left( \frac{\epsilon_0 k_B T_e}{n e^2} \right)^{1/2} \quad (2.2)$$

$n$ is the plasma density and $T_e$ is the electron temperature, $\epsilon_0$ is the permittivity of the vacuum, $k_B$ is the Boltzmann constant and $e$ is the electron charge. The Debye length is a measure of the Debye shielding distance. For UCPs, the “system size” is normally taken to be the rms radius of the Gaussian plasma distribution, denoted $\sigma$. If the shielding distance is much smaller than the size of the plasma, then fields are effectively screened at the edges of the plasma, and cannot penetrate the bulk.

Debye shielding is a term used to describe the ability of a plasma to exclude applied electric potentials, due to the motion of free charges in the plasma. Just as free charges in a conductor will move such that the electric field in a conductor is zero, the charges in a plasma will move to shield the bulk of the plasma from an applied electric potential. However, in a plasma the charged particles are completely free and have some thermal energy, so that the screening is less successful than that of a conductor. There will be some
distance from the edge of the plasma at which the energy density of the electric field falls to the same order as the energy of the thermal motions of the electrons, $k_B T$. At this distance, (the Debye length) charged particles in the plasma have sufficient energy to escape the electric potential. As a result, fields with potential of the order $k_B T/e$ or less will be able to exist in the plasma.

The \textit{plasma parameter}, $N_D$, is the number of of particles in a “Debye sphere” (sphere of radius $\lambda_D$). For the concept of Debye shielding to be statistically valid, we require the number of particles in this volume to be significant, i.e.

$$N_D = \frac{4}{3} \pi \lambda_D^3$$

$$= 1.38 \times 10^6 \frac{T^{3/2}}{\sqrt{n}}$$

and we require that for a plasma,

$$N_D >>> 1$$

\subsection{2.2.2 Plasma Waves}

The \textit{plasma frequency} is defined as the frequency at which displaced electrons oscillate around their mean positions due to the electrostatic restoring forces of the (massive) ions. To understand the fundamental concept, consider an electron and its parent ion. If the electron is pushed away from the ion, it will be pulled back by the Coulomb attraction, but will overshoot due to inertia, such that it is pulled back toward the ion, overshoot again, and so on, resulting in an oscillation around the parent ion. The frequency at which this
oscillation occurs is called the *plasma frequency*. Mathematically, it depends only on the equilibrium electron density $n_0$:

$$\omega_p = \left(\frac{n_0 e^2}{\epsilon_0 m}\right)^{1/2}$$

(2.6)

where $m$ is the electron mass. Following of the method of Chen [7], this surprising relation can be obtained by solving Maxwell’s equations for the dispersion relation of a purely electric (no magnetic field) *longitudinal* wave.

With no magnetic field, we have only Gauss’s law:

$$\nabla \cdot \mathbf{E} = \frac{\rho}{\epsilon_0}$$

(2.7)

where $\rho$ is the total charge density. For a longitudinal wave, $\mathbf{E}(x,t) = E e^{i(kx-\omega t)} \hat{x}$ as usual so that

$$i k E e^{i(kx-\omega t)} = \frac{\rho(x,t)}{\epsilon_0}.$$  

(2.8)

Now, the continuity equation can provide the charge density $\rho$:

$$\nabla \cdot \mathbf{J} = -\frac{\partial \rho}{\partial t}$$

(2.9)

where $\mathbf{J}$ is the total current density. For a longitudinal wave, the current will also be longitudinal, so that

$$J_x(x,t) = J e^{i(kx-\omega t)}$$

(2.10)

$$\rho(x,t) = \rho e^{i(kx-\omega t)}.$$  

(2.11)
Substituting these into Eq. 2.9, we have

\[ i k J e^{i(kx-\omega t)} = i \omega \rho e^{i(kx-\omega t)} \]

\[ \Rightarrow \rho = \frac{k}{\omega} J. \]  \hspace{1cm} (2.12)  \hspace{1cm} (2.13)

For electrons, the current density is simply the charge density multiplied by the velocity. For a neutral plasma with density \( n_0 \) then, the electron current density is simply \( J = -en_0 v \). For a cold plasma \( (T = 0) \) we can find \( v \) from Newton’s second law \( (F = ma) \), setting

\[ v_x(x,t) = v e^{i(kx-\omega t)} \]  \hspace{1cm} (2.14)

so that

\[ F = -eE_x = m \frac{dv_x}{dt} \]  \hspace{1cm} (2.15)

\[ -eE e^{i(kx-\omega t)} = -i \omega m v e^{i(kx-\omega t)} \]  \hspace{1cm} (2.16)

\[ \Rightarrow v = \frac{e}{i \omega m} E. \]  \hspace{1cm} (2.17)

The current density is now

\[ J = -\frac{e^2 n_0}{i \omega m} E \]  \hspace{1cm} (2.18)

and using Eq. 2.13 we have

\[ \rho = \frac{k}{\omega} \left( -\frac{e^2 n_0}{i \omega m} \right) E. \]  \hspace{1cm} (2.19)
Substituting back into Eq. 2.8 gives

\[ ikE e^{i(kx - \omega t)} = \frac{\rho e^{i(kx - \omega t)}}{\epsilon_0} \]

\[ \Rightarrow ikE = -\frac{k e^2 n_0}{i \omega^2 \epsilon_0 m} E \]  \hspace{1cm} (2.21)

\[ \Rightarrow \omega^2 = \frac{n_0 e^2}{\epsilon_0 m} = \omega_p^2 \]  \hspace{1cm} (2.22)

as required, where we have used Eq. 2.11. Although this result might at first seem odd for a dispersion relation, the physical interpretation is quite simple: if the electrons are perturbed, they will oscillate at this frequency regardless of the wavelength. It should be noted that this derivation assumes an infinite plasma at zero temperature. For non-zero temperatures, the dispersion relation is modified to the Bohm-Gross relation [50, 7]:

\[ \omega^2 = \omega_p^2 (1 + 3 k^2 \lambda_D^2). \]  \hspace{1cm} (2.23)

We can see that in the cold plasma limit \((k \rightarrow 0)\) this describes a wave with frequency \(\omega_p\), and a linearly dispersive electron sound wave ("Langmuir wave" [51]) for high \(k\). An appropriate \(k\) value estimation can be approximated using the size of the plasma \(\sigma\):

\[ k = \frac{2 \pi}{\sigma}. \]  \hspace{1cm} (2.24)

For UCP sizes and temperatures, the second term in equation 2.23 can be significant, such that temperature cannot be disregarded when considering wave propagation in UCPs [6, 7].

For an ionised gas to be a plasma, its motion must be controlled by electro-
magnetic rather than hydrodynamic forces. If \( \tau \) is the average time between collisions with neutral atoms, then for electromagnetic forces to dominate, the plasma oscillation frequency must be faster than \( 1/\tau \), i.e.

\[
\omega_p \tau > 1.
\]  

\[(2.25)\]

For a plasma with density \( 10^{10} \text{ cm}^{-3} \), the corresponding plasma frequency, \( f_p \), is about 897 MHz (\( \omega_p \approx 5.6 \text{ GHz} \)). A plot of plasma frequency versus density is shown in figure 2.1.

![Figure 2.1: Plot of plasma frequency versus electron density, for electron densities comparable to our expected experimental range.](image)

Together, equations 2.1, 2.5 and 2.25 form a mathematical definition of a plasma.

A dispersion relation similar to Eq. 2.23 can also be derived using a similar
technique for transverse waves, by setting

\begin{align*}
E_y &= E e^{i(kw - \omega t)} \quad (2.26) \\
J_y(x, t) &= J e^{i(kw - \omega t)} \quad (2.27) \\
v_y(x, t) &= v e^{i(kx - \omega t)} \quad (2.28)
\end{align*}

Use of Newton’s law results in the same expression for the current density, Eq. 2.18. Faraday’s and Ampere/Maxwell’s laws are:

\begin{align*}
\nabla \times \vec{E} &= -\frac{\partial \vec{B}}{\partial t} \quad (2.29) \\
\nabla \times \vec{B} &= \mu_0 \vec{J} + \mu_0 \epsilon_0 \frac{\partial \vec{E}}{\partial t}, \quad (2.30)
\end{align*}

so that substituting the \( e^{i(kr - \omega t)} \) waveform gives

\begin{align*}
\imath k \times \vec{E} &= \imath \omega \vec{B} \quad (2.31) \\
\imath k \times \vec{B} &= \mu_0 \vec{J} - \frac{\imath \omega}{c^2} \vec{E}. \quad (2.32)
\end{align*}

Eliminating \( \vec{B} \) we have

\begin{align*}
\imath k \times \frac{1}{\imath \omega} (k \times \vec{E}) &= \mu_0 \vec{J} - \frac{\imath \omega}{c^2} \vec{E} \quad (2.33) \\
\Rightarrow -\imath \omega \mu_0 \vec{J} &= \left( -k^2 + \frac{\omega^2}{c^2} \right) \vec{E}. \quad (2.34)
\end{align*}

Substituting Eq. 2.18 for the current density the field cancels and we find

\begin{align*}
-\imath \omega \mu_0 \left( -\frac{n_0 e^2}{\imath \omega m} \right) &= \frac{\omega}{c^2} - k^2 \quad (2.35) \\
\Rightarrow \frac{1}{c^2} \omega_p^2 &= \omega^2 - k^2 c^2 \quad (2.36) \\
\Rightarrow \omega^2 &= \omega_p^2 + k^2 c^2. \quad (2.37)
\end{align*}
Solving for $k$ gives

$$k = \frac{\sqrt{\omega^2 - \omega_p^2}}{c}.$$  \hspace{1cm} (2.38)

This result gives rise to an interesting property of plasmas, which we will rely upon later. Frequencies below the plasma frequency propagate with an imaginary $k$, and decay exponentially. As a result, they cannot propagate in the plasma. In the context of Debye shielding, this result is unsurprising: free charges can easily move to cancel out the applied field, but they can only do so up to a point. If the oscillation is faster than the charges can respond (which will depend on the density of the plasma), the field will not be cancelled, and the wave will propagate. When the density varies across the plasma, the cutoff frequency varies with position, and causes an interesting phenomenon known as “Tonks-Dattner resonances”. Such resonances can provide diagnostic information about the density and temperature of the plasma, and are described in Sec. 2.4.2.

Up to this point we have only considered electron plasma waves, since ion acoustic waves have a much lower velocity, the same as the plasma free expansion velocity $[6]$:

$$v_{iaw} = \sqrt{k_B T_e / m_i}.$$ \hspace{1cm} (2.39)

While Coulomb repulsion prevents direct ion-ion collisions, an ion sound wave (longitudinal pressure wave) can still propagate through the intermediary of an electric field. The dispersion relation for an ion acoustic wave is $[7]^2$

$$\frac{\omega}{k} = \left( k_B T_e + \gamma_i k_B T_i / m_i \right)^{1/2}$$ \hspace{1cm} (2.40)

---

\(^2\text{Page 96}\)
where $\gamma_i$ is 3 for one-dimensional plane wave compressions of the ions, $T_i$ and $T_e$ are the ion and electron temperatures, respectively, and $m_i$ is the ion mass.

### 2.2.3 Strongly Coupled Plasma

A plasma is defined as “strongly coupled” when the Coulomb interaction energy between charged particles exceeds the average kinetic energy [52]. Such plasmas are also termed “collisionless”, since particles of like charge lack the necessary kinetic energy to overcome the Coulomb repulsion between them and collide [7]. The characteristic measurement of strong coupling is defined as

$$\Gamma = \frac{e^2}{4\pi \epsilon_0 ak_B T}$$

(2.41)

where $\Gamma$ is the “Coulomb coupling parameter” and $a$ is the Wigner-Seitz radius characterising the separation between particles in a plasma of density $\rho$:

$$a = \left[ \frac{3}{4\pi \rho} \right]^{1/3}.$$  

(2.42)

A $\Gamma$ value greater than one indicates strong coupling.

After the production of the first UCP in 1999 [1], the initial temperature of electrons in UCP was believed to be as low as 100 mK, limited only by the energy spread of the photoionisation laser. For an electron density of $10^{11}$ cm$^{-3}$, we would then expect a $\Gamma$ value of over 100 immediately after ionisation. Such a high value of $\Gamma$ may result in formation of Wigner crystals, the crystalline phase of a plasma. Wigner crystals are formed when thermal motions are much smaller than electron-electron repulsion forces, resulting
2.2 Plasma Fundamentals

in a regular lattice. Crystallisation to a cubic lattice is predicted at or above \( \Gamma \approx 170 \) [53,54,55].

As predicted by Kuzmin and O’Neil in 2002 [3], intrinsic heating of the plasma (see Sec. 2.3) reduces the \( \Gamma \) value to below threshold on short timescales, such that their (accessible) temperatures are typically between 1-1000K, with ion temperatures around 1K [52]. In a rubidium-85 magneto optical trap, atomic densities of order \( 10^{12} \text{cm}^{-3} \) are typically achieved. Assuming that approximately one third of the atoms are ionised, an electron temperature of 10K results in a \( \Gamma \) parameter of \( \sim 1.8 \). While this indicates strong coupling, Wigner crystals will not form.

In contrast to neutral UCP, one-component plasmas, such as laser-cooled \(^9\text{Be}^+\) can have temperatures in the \( \mu \text{K} \) range, resulting in correspondingly high \( \Gamma \) values. This is possible due to the direct laser cooling of the ions inside a Penning trap. Such systems have exhibited crystallisation [56], providing an attractive candidate for study of the phase transition. They are of particular value in investigations of quantum control and noise reduction for quantum information processing [57,58].
2.3 Heating processes

As outlined above and shown explicitly in Eq. 1.1, the achievable brightness of the electron source depends critically on the initial temperature of the electrons. It is therefore important to properly understand the various heating mechanisms which influence the electrons during the initial stages of evolution. This in turn will indicate the timescales on which the electrons must be extracted as well as suggest possible avenues of future work in order to minimise the effects of any heating processes.

2.3.1 Disorder-induced Heating

When a strongly coupled plasma is formed via laser photoionisation, the resulting plasma is not in true thermal equilibrium. While a large Γ parameter would typically indicate expected ion-ion and electron-electron spatial correlations, the uncorrelated initial atomic states results in minimal correlations in the resulting plasma. As these correlations develop in the plasma, the energy difference between the uncorrelated plasma and the thermal equilibrium state causes heating of the plasma. This limits the strength of the correlations which can ultimately be reached in the plasma [3]. This effect is also sometimes referred to as correlation induced heating [59].

An alternate approach is to think of the electrons as being placed (ionised) onto a potential slope, and immediately moving down the potential gradient. If the average interparticle distance is $a$ (Wigner-Seitz radius) then an electron gains an energy of $e^2/4\pi\varepsilon_0a$ when travelling this distance, and the
2.3 Heating processes

The timescale for this heating, $\tau_{\text{disorder}}$, is

$$
\tau_{\text{disorder}} = \frac{a}{\sqrt{e^2/4\pi \varepsilon_0 a m_e}} 
\approx \frac{1}{\omega_p}.
$$

By reducing the density, thereby increasing the interparticle separation, we can increase the time taken for this heating to develop. As shown earlier, in our experiments we expect a plasma frequency of the order $\omega_p \approx 5 \text{ GHz}$, corresponding to heating on picosecond timescales. To extract the electrons before they are heated by this process would require extraordinarily fast switching of large electric fields. Alternatively, the problem could be addressed prior to ionisation, by preparing the atoms in a correlated spatial arrangement using an optical lattice \[60,61\]. The ability to use such a trap to minimise heating in future experiments was an important consideration during the design of the experimental apparatus (Sec. 3.1).

### 2.3.2 Three-body Recombination

Three-body recombination is a process in which an ion and two electrons interact to form an energetic electron and an excited neutral atom \[52\]. The TBR rate varies with temperature as $T^{-9/2}$ \[62\], so that at low temperatures TBR is the dominant recombination process. Measurements of the TBR rate can thus provide an indication of electron temperature during the first stages of plasma evolution \[63\].

The TBR rate varies with the square of the electron density, but linearly\footnote{Equation IV.18, page 12}
with the ion density, and is normally given by [63]:

\[ R_{TBR}(s^{-1}) = K_{TBR} T_e^{-9/2} \int n_e^2(r) n_i(r) 4\pi r^2 dr, \]  

(2.45)

where \( K_{TBR} \) is the three body rate constant, with an accepted value of \( K_{TBR} = 3.9 \times 10^{-21} \text{m}^6\text{K}^{9/2}\text{s}^{-1} \) [62]. Strictly speaking, this TBR rate expression is not valid in the strongly coupled regime, but has been used to determine the electron temperature in UCPs with densities lower than that required for strong coupling. The TBR rates are calculated by integrating over all possible final atomic states, \( n_f \). At very low temperatures (< 1 K) corrections to the rate can be implemented using a density dependent cut-off for high Rydberg states, \( n_{f_{\text{max}}} \) [64], resulting in a temperature and density dependence of \( T^{-1/2} n_e^{5/6} \). In our experiments, we expect intrinsic heating of the plasma to increase the electron temperature above 1 K, where \( T^{-9/2} \) scaling is valid.

Other contributing processes to recombination include radiative recombination (electron capture and photon emission) and dielectronic recombination (electron capture, creating a doubly unstable Auger state and subsequent photon emission), however due to the \( T^{-9/2} \) scaling of TBR, these processes are not expected to contribute significantly to recombination at UCP temperatures [62].

### 2.3.3 Continuum Lowering

The initial kinetic energy of the electrons in a UCP is predominantly due to the excess energy gained from the photoionisation laser. One might expect then, that if the effects of disorder-induced heating can be reduced through
ordering of the atomic spacing, the lower limit to the achievable electron temperature is ultimately the linewidth of the ionisation laser. However, the long-range interaction potential of the ions actually lowers the appropriate zero energy from that of the atom to the collective plasma, meaning that the electrons will always have this initial energy (and hence temperature limit) regardless of the excess energy from the ionisation laser. This effect is known as “continuum lowering” or the “threshold lowering effect (TLE)”.

Consider a single atom to be photoionised. Immediately after ionisation, the atom sees the potential of the parent ion as usual (Fig. 2.2a). If we now place additional ions next to the first (Fig. 2.2b), the potentials can overlap, such that a free electron now sees an effectively lower potential than for a single ion. If the ions are placed closer together (i.e. the density is higher) then the overlap is greater and the zero level shifts even lower. At low excess ionisation energies, the minimum attainable electron temperature is thus determined by the density (via the Wigner-Seitz radius, $a$)

$$T_{e,min} = \frac{2a_B C_P E_h}{3a k_B}$$  

where $C_P$ is a constant ($\approx 11 \pm 5$) [59, 65, 66], $a_B$ is the Bohr radius and $E_h$ is the Hartree energy. At $n \approx 10^9$ cm$^{-3}$, this corresponds to a $T_{e,min} \approx 20$ K.

**Figure 2.2:** Continuum lowering due to long range Coulomb interaction between ions. (a) Potential from a single ion; (b) Potential due to several ions, with overlap of potential wells, resulting in (c) lowering of the effective zero energy.
2.4 Plasma diagnostics

We have seen that the initial temperature of the plasma is critical for production of bright electron bunches, and that it is easily increased by several physical mechanisms during the first stages of plasma formation and evolution. To optimise the electron output, it will be important to be able to probe not only the temperature, but also the density distribution of the plasma after production.

Direct observation of the ions in the plasma (for example, using absorption imaging) would provide an ideal means of extracting information, however unlike strontium [67], singly ionised rubidium does not have an electron transition in the optical wavelength range. Observation of the expansion of the electron bunch can provide a means of temperature measurement, but requires a variable propagation distance or electron optics, both of which are planned upgrades to the experimental system. Several less direct methods of extracting electron temperatures and densities exist. For our experimental apparatus, two of these methods are of particular interest, since they permit indirect extraction of electron temperature.

2.4.1 Three body recombination

The accepted scaling of TBR rate with electron temperature of $T^{-9/2}$ means that for low electron temperatures where TBR is the dominant recombination process, the TBR rate can provide a measurement of electron temperatures. [63].

When the plasma is created, Rydberg states near the ionisation threshold
are populated. Electrons in Rydberg states are then ionised using a microwave RF pulse, typically about 50 \( \mu s \) after the initial ionisation pulse. The electrons are accelerated to a time-resolving detector. The Rydberg states are then refilled via TBR and a second RF pulse is applied to ionise the (now filled) Rydberg states after some delay time (typically several hundred nanoseconds, see figure 2.3). By measuring the number of electrons ionised by the second pulse, the refilling rate is measured. Since TBR is the dominant process at these temperatures, the refilling rate gives a direct measurement of the TBR rate, which can then be used to extract the electron temperature (figure 2.4).

**Figure 2.3:** Image from [63]. Electron emission signal from an expanding UCP, averaged over 40 runs. The double peaks at 55 \( \mu s \) are the response due to a pair of short (100 ns) microwave pulses. The first of the two pulses is held fixed and ionizes Rydbergs that have formed in the plasma. Following this pulse, the Rydberg population is refilled by three body recombination. The second pulse is applied at varying times, and the number of Rydbergs ionized is counted as a function of the time between the two pulses (inset).
Figure 2.4: Image from [63]. (a) Electron temperature as determined by measurement of TBR rate. $T_e$ is calculated using TBR theory with assumed self-similar Gaussian expansion of the plasma. The initial energy is $\Delta E = 3$ K. For comparison, also plotted are earlier $T_e$ measurements for $\Delta E = 10$ K (squares) [59], simulation results for $\Delta E = 66$ K (dashed line) and simulation results for $\Delta E = 3$ K (solid line). (b) The corresponding plasma coupling parameter, calculated using simulation results.

2.4.2 Tonks-Dattner Resonances

Tonks-Dattner resonances are a closely spaced series of resonances which occur when a plasma is subjected to a transverse RF electric field. If the electron density in the plasma varies spatially, such that it is higher in the center of the plasma than the outer regions, then the plasma frequency is also higher in the center than the outer regions. This means that a plasma wave which can propagate in the outer regions (real $k$) may be reflected
by the inner core of the plasma, where $\omega < \omega_p$. If the phase shift due to
propagation of the field between the edge of the plasma and the inner core
is $2n\pi$ (where $n$ is an integer) then a standing wave will occur, resulting in a
resonance. This effect was originally observed and explained in hot plasmas
using cylindrical geometry [68, 69, 70], but recent work [6] has observed the
effect in an expanding (Gaussian) UCP. This was detected via peaks in the
electrons emitted from the plasma after some delay (typically of the order
tens of microseconds, see figure 2.5). The delayed electron emission arises due
to the excitation of the collective plasma oscillation mode. Energy from the
RF field is transferred into the plasma to excite the oscillation, which in turn
heats the plasma when the energy is redistributed among all the electrons
(typically within 10-1000 ns). The evaporation rate from the Coulomb well
is then temporarily increased, resulting in a pulse of electrons being ejected
from the plasma [48].

To extract information about the temperature and density of the plasma, the
experimental results are fitted to a theoretical model. Theoretical predictions
of the resonances make use of the Vlasov equation. The Vlasov equation is
a special case of the Boltzmann equation, and can be used in kinetic theory
descriptions of a plasma, when collisions can be neglected. [7] It has the form:

$$\frac{\partial f}{\partial t} + \mathbf{v} \cdot \nabla f + \frac{q}{m} (\mathbf{E} + \mathbf{v} \times \mathbf{B})) \cdot \frac{\partial f}{\partial \mathbf{v}} = 0 \quad (2.47)$$

where $f(r, \mathbf{v}, t)$ is the velocity distribution function for each species, describ-
ing the density of particles at position $\mathbf{r}$ and time $t$ with velocity components
$v_x, v_y, v_z$. Numerical solutions have been computed, but unlike cylindrical
plasmas where the plasma wall is well defined, a freely expanding UCP has
Figure 2.5: Image from [6]. Typical electron emissions from an expanding ultracold plasma. RF electric fields with the noted frequencies are applied to the expanding plasma, yielding distinct peaks that depend on frequency. The applied RF power is adjusted to make the peaks visible. Note the distortion of the curves at times immediately after the prompt peak for higher RF frequencies, due to the higher RF power required to observe resonance peaks at those frequencies.

an ambiguous boundary. For simplicity, Fletcher et al. used a value of $3\sigma$ for the plasma “wall” of a freely expanding Gaussian UCP. By fitting the electron density and temperature to experimentally measured data, the resonances provide a measurement of these parameters. Figures 2.5 and 2.6 show typical electron emission signals, and the fitted theoretical predictions of the resonance frequencies. The fits indicate a temperature of $T = 19$ K and peak initial electron density of $n_0 = 1.9 \times 10^{-9}$ cm$^{-3}$ which is consistent with previous results [59]. The method is experimentally straightforward, and although the theoretical model requires further refinement for a freely expanding geometry in order to provide a thoroughly rigorous measurement, Tonks-Dattner resonances can at least provide confirmation of temperature
2.5 Ultracold Plasma Production

2.5.1 Photoionisation

Photoionisation provides several advantages when producing ultracold plasma. We have previously described how a two-step ionisation process can allow shaping of the resulting electron cloud in three dimensions. In addition, pre-
cise control over the laser wavelength allows the initial electron temperature to be minimised, which in turn maximises electron brightness. However, any wavelength spread in the ionisation laser will result in a range of initial electron temperatures, which will increase emittance and decrease brightness. For a typical laser linewidth\(^4\) of 0.7 cm\(^{-1}\), this implies an energy spread of \(\Delta E \approx 0.01\) meV, corresponding to a temperature spread of about 100 mK. The heating processes described above show that this additional temperature range is unlikely to be a limiting factor in the production of high brightness electrons.

Photoionisation of cold rubidium atoms in a MOT has also been achieved using a single photon at 296 nm (4.177 eV) produced by frequency doubling yellow light in a beta barium borate (BBO) crystal [71]. Ciampini \textit{et al.} [71] measured the single photon ionisation cross sections from the ground (296 nm) and excited 5P states (420 nm). The cross sections were extracted from a comparison of trapped atom loss rates. The cross section for ionisation from the excited state was found to be two hundred times greater than from the ground state:

\[
\begin{align*}
\sigma_{5S_{1/2}} &= (0.76 \pm 0.15) \times 10^{-19}\text{cm}^2 \quad (2.48) \\
\sigma_{5P_{3/2}} &= (1.34 \pm 0.16) \times 10^{-17}\text{cm}^2. \quad (2.49)
\end{align*}
\]

The larger cross section for the 5P state means that even when we include incomplete excitation of the 5P state (typically \(\sim 30\%\)), our two-step ionisation method is more efficient than direct ionisation of the ground state and also avoids technical complexities typically associated with short wavelength

\(^4\text{Quantel Brilliant B-10, http://www.quantel-laser.com}\)
2.5 Ultracold Plasma Production

laser sources. Higher excitation fractions are also possible via coherent excitation (using $\pi$-pulses) or stimulated Raman adiabatic passage (STIRAP) techniques [72, 73].

2.5.2 Penning Ionisation

Dense Rydberg gases have been observed to evolve into an ultra cold plasma, due to inter-atomic collisions [74, 75, 76, 77]. The cross section for atom-atom collisions is normally very small in cold atom clouds, due to the relatively large inter-atomic distance, compared to the size of the atom. In Rydberg gases this cross section is dramatically increased, as the atoms are highly excited and hence have an increased size and dipole moment. The dipole–dipole interaction for two Rydberg atoms scales with interparticle distance $R$ as $1/R^3$ [78, 79], while van der Waals interactions scale as $1/R^6$ [80]. The van der Waals interaction coefficient in turn scales with principle quantum number $n$ as $n^{11}$ so that at typical trapped atom densities, long-range interactions are significantly enhanced for Rydberg states. Ionisation can occur when an inter-atomic collision ionises one atom while returning the other to a less excited state.

While laser-cooled Rydberg atoms are mostly cold, some hot (300 K) Rydberg atoms are formed when the cloud is excited, usually by a laser pulse. It is these atoms which begin the ionising collisions. Studies of these effects are typically performed by pulsed Rydberg formation and subsequent electron or ion extraction to a time resolved MCP detector [74, 81]. As electrons leave the plasma, the cloud becomes net positively charged, thus trapping electrons subsequently formed in the MOT. These trapped electrons rapidly
collide with the remaining Rydberg atoms, causing an avalanche ionisation. Even in the event that electrons are not actively extracted, the avalanche will still occur due to the early electrons escaping the plasma [82, 77].
Chapter 3

The Melbourne UCP

The design of the Melbourne UCP apparatus is detailed in Sec. 3.1, with particular attention given to the vacuum system, cooling laser system and switching electronics. While largely technical in nature, an accurate description of these components is required in order to understand the experiments presented in Sec. 3.4. Section 3.1 also highlights the physical concepts which support the technical design decisions taken over the course of the project. The construction of the vacuum apparatus, control electronics, laser systems and computer control system are described in Sec. 3.3, and initial results characterising the plasma are presented in Sec. 3.5.

3.1 Design

At the commencement of this project in 2007, no apparatus existed for creation of the ultracold plasma. The initial design focused on the key requirements of the system, while simultaneously seeking to improve upon design limitations in existing UCP systems such as that of our collaborators in Eindhoven, The Netherlands.

During a four week visit to the UCP laboratory at Technische Universiteit
The Melbourne UCP

Eindhoven (TUE), their existing UCP was analysed to determine the optimum approach to building the new Melbourne experiment. Although many of the vacuum components used are available “off-the-shelf”, consideration of the possible electronic, optical, computational, imaging, laser and magnetic field requirements was also necessary, resulting in the decision to create a new and radically different type of UCP system.

Several design priorities resulted directly from investigation of the Eindhoven UCP system, specifically:

- Minimisation of in-vacuum optical and magnetic components which caused difficulty during alignment procedures
- Maximisation of optical access to the cold atoms, e.g. for imaging
- Provision of multiple points of entry for electrical (often high-voltage) vacuum feedthroughs for accelerator and electro-optical components
- Large-volume vacuum pumps with high-conductance connections to allow rapid pump-down of the vacuum chamber to ultra-high-vacuum (UHV) conditions
- Maximisation of safe bakeout temperature for all components to aid pump-down and minimise final chamber background pressure
- Ability to load the MOT from either a rubidium dispenser or a high-flux Zeeman slower source [83].
- Accommodation of a large-diameter accelerator structure [84] to ensure uniform homogeneous acceleration fields.
The UHV apparatus was designed using a 3D CAD program (*Solidworks 2006*) with regular consultation with other members of the Melbourne and Eindhoven groups. The scale and complexity of the experiment required the design and construction work to be shared between members of the Melbourne group. The atom source (rubidium oven and Zeeman slower [83]) and internal accelerator structure specifics were designed primarily by other members of the Melbourne group, in consultation with the Eindhoven group [84]. I produced the design, modelling and mechanical drawings of the main experimental chamber, and ensured that all parts of the final system would successfully facilitate our experimental goals. To begin the design process of such a large-scale experiment, a clear list of these goals was required, which could then be subdivided into distributable projects.

### 3.1.1 System requirements

The requirements of our UCP system are dictated primarily by Eq. 1.1 for the transverse brightness of an electron beam.

\[
B_\perp = \frac{mc^2J}{\pi k_B T_e}
\]  

(3.1)

We want to maximise the brightness, by generating as many electrons as possible, as cold as possible, in the smallest possible volume. Hence, we aim to maximise the density of cold atoms in our trap, while minimising their temperature. This problem has been examined previously [85,86] resulting in the finding that the density in the trap is essentially limited by two processes. Inter-atomic collisions between excited- and ground-state atoms can result in partial transfer of the excitation energy into sufficient kinetic energy for
an atom to escape the trap potential. In addition, absorption of re-scattered light can result in sufficient radiation pressure to balance that of the confining laser beams. Background gas (thermal, untrapped atoms) reduces the trap density and trapped atom numbers, primarily through collisions with cold trapped atoms, and also through scattering of the cooling beams which can lead to subsequent absorption and heating of the trapped atoms. Accordingly, high pumping speed is important to minimise the background pressure, and our source of atoms should ideally load atoms specifically into the trap, thereby minimising background pressure and maximising trapped atom number and density.

While minimising background pressure is important, we also require a high flux of atoms so that the trap can be loaded quickly. We aim to produce electron bunches at an initial repetition rate of 10 Hz (set by the repetition rate of our pulsed ionisation laser) so the atom source requires sufficient flux to maintain a “full” trap, such that the trapped atom number does not decrease with subsequent ionisation pulses. While many possible atom sources were considered [87, 88, 89, 90, 91], a single-layer Zeeman slower [83] was chosen to provide the necessary balance between low background pressure and high atomic flux. In addition, a standard rubidium dispenser source was mounted inside the main chamber as a secondary source in case of unforeseen equipment failure.

The magneto optical trap has become the workhorse of modern atomic physics research, resulting in many variations in design geometry. For our application, we require the cold atoms to be located inside a parallel plate accelerator structure to extract the electrons. This structure will restrict optical access to the cold atoms. Unlike a conventional MOT which requires
optical access along all six cooling beam axes \cite{2}, we chose to utilise an adaptation of the “mirror” MOT design, shown in figure 3.1. Our design was developed separately but almost simultaneously with the very similar quasi-mirror-MOT design at the National Institute of Standards Technology (NIST) in Gaithersburg, MD \cite{92}. This provides ease of optical alignment, while removing the requirement for optical access from all sides of the atom cloud. This adaptation retains a full six-beam MOT (as opposed to the four beams in a standard mirror-MOT \cite{93}) which provides greater control over the spatial extent of the trap potential. It also allows removal of other optics and the magnetic coils from within the vacuum chamber. This is a significant simplification of the Eindhoven design, which had several mirrors and high-current coils inside the chamber. Internal optics made alignment of the MOT beams difficult, and the relatively few windings of their internal coils required fast switching of very high currents (approximately 180 A). Our parallel plate design allows removal of the coils to the exterior of the chamber, allowing for more turns and thus reducing the current required for operation and improving optical access. The close spacing of the plates near the plasma also reduces the potential needed to extract the electrons, in turn allowing easier fast switching of that potential.

To facilitate the new Melbourne design, two of the cooling beams must pass through one of the accelerator plates. That plate is custom-made anti-reflection coated glass, also coated with indium-tin-oxide (ITO), which allows electrical conduction of the accelerating potential. The mirror plate is gold-coated copper, while the final plate is solid copper. All edges of the plates were rounded to prevent high-voltage breakdown due to the high electric field strengths that can result from sharp edges. The plates were
Figure 3.1: Rendered CAD image of the quasi-mirror MOT arrangement inside the main chamber. The trapping and cooling beams (shown in red) in the horizontal plane are reflected from the polished surface of the central accelerator plate, and pass through the anti-reflection coated surfaces of the right hand plate. Atoms are cooled and trapped in the central region of overlapping beams. After ionisation, a potential difference between the plates accelerates the electrons towards the detector (not shown). The third accelerator plate forms a second stage of accelerator. This design minimises emittance growth of the electron beam and the accelerating potential required, simplifying the fast switching requirements of the accelerator. Each plate is approximately 11 cm in diameter, and 4 mm thick. From left to right, the plate separations are 1 cm and 5 cm with central hole diameters of 2 cm, 2 cm and 0.6 cm. The indicated voltages are the nominal maximum design voltages.

mounted in a cylindrical structure made from UHV-compatible, insulating PEEK (polyether ether ketone) which was secured to the inner wall of the main chamber. The plates are secured on one side by a stainless steel ring, into which electrical connections are secured. Commercially available UHV-safe polyimide-coated wires, in-vacuum connectors and high-voltage electrical feedthroughs connect the external power supplies to the accelerator.
3.1 Design

3.1.2 Vacuum system design

The experimental chamber is shown in figure 3.2. The main features of the system are:

- Rubidium oven atom source
- Zeeman slower to provide high flux of slow atoms
- Titanium sublimation and ion getter pumps, backed by turbo pumps during UHV bakeout
- Internal surface area of 0.73 m² resulting in a minimum theoretically achievable pressure of 1.8 \times 10^{-11} \text{ torr} limited by the internal surface area gas load of \( Q = 6.8 \times 10^{-9} \text{ torr litres per second} \) [94]
- Central plasma chamber combining:
  - six cooling and trapping beams in quasi-mirror MOT arrangement
  - seven electrical feedthroughs for accelerator plates, refocusing lens, and alternate atom sources (rubidium dispensers)
  - two direct line-of-sight axes for diagnostic imaging
  - three possible axes for ionisation and/or excitation beam
  - microchannel plate electron detector and phosphor screen for electron beam measurements
  - access for slow atoms from a Zeeman slower, for high flux atom source
  - large diameter connection to high speed ion pump for high conductance and effective pump rate
– direct optical access through the chamber for the Zeeman slower laser beam.
Figure 3.2: (a) Rendered CAD drawing of the UCP experimental chamber design, (b) top-down section view through line in (a), and (c) exploded view of the trapping region with accelerator plates. The rubidium oven and Zeeman slower are on the right, main ion pump on the left and the main chamber in the center, showing cooling and trapping beams in red. The Zeeman slower laser enters through a port on to the left of the ion pump, and travels through the ion pump and main chamber to the oven aperture on the right.
3.1.3 Laser systems

Ionisation laser

The ionisation laser consists of a 480 nm pulsed dye laser\(^1\) using Coumarin 102 dye pumped by a frequency upconverted Q-switched Nd:YAG laser\(^2\) at 355 nm. The pulse length of the pump is 5 ns full-width at half-maximum (FWHM) and the repetition rate is fixed at 10 Hz. The usable wavelength range is from 465–490 nm and the pulse power of the pump laser (and hence that of the dye laser) is adjustable by varying the time delay between the flashlamp and Q-switch. The maximum pulse energy of the blue laser is approximately 10 mJ.

The ionisation laser pulse can be synchronised with the experiment via the TTL I/O connections to and from the flashlamp and Q-switch on the Nd:YAG laser. Synchronisation can be achieved in two ways. In the first mode, both the flashlamp and Q-switch are externally triggered from a digital timing card (Sec. 3.3). This method has the advantage of controlling all aspects of the experimental sequence from a single clock source, however it quickly becomes impractical due to technical restrictions. The pump laser requires that the flashlamp is fired at least 80 times before the first trigger of the Q-switch, corresponding to an eight second delay before light is emitted. To then alter the Q-switch delay (and hence the output power), the timing card must be stopped, a new program uploaded, and the new program started, which takes much longer than the 100 ms between flashlamp pulses (typically 500 ms). Since no flashlamp input pulses are received by the laser

during this reprogramming, the laser registers the flashlamp as “off”, such that it then requires another eight seconds before emitting light with the new Q-switch delay.

The second method, which has been used for experiments presented here, uses the internal clock of the Nd:YAG laser to trigger the flashlamp independently. A TTL signal from the flashlamp circuit is sent to the digital timing card to act as a trigger to begin the rest of the experimental sequence. Although somewhat more cumbersome, this method allows variation of the Q-switch delay in real time, increasing the efficiency of data collection significantly.

**MOT lasers**

A high power, narrow linewidth laser source was required for cooling and trapping the atom cloud. While several commercially available systems exist, they have proven unreliable in past experiments and required frequent and costly returns to European manufacturers for repairs. To avoid the cost and long experimental delays resulting from returns, I designed a compact tapered amplifier (TA) laser system, capable of amplifying a seed laser up to a maximum output of 2W, with currently available diodes (see figure 3.3). As laser diode technology develops, this output power may well be extended even higher, and is limited only by the diode used, rather than the design. Combined with the seed laser (“master oscillator”) this amplifier forms a “master oscillator power amplifier” (MOPA) and, using appropriate diodes, can amplify light at several wavelengths used in the project [95].

The seed laser is normally one of our existing external cavity diode lasers

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For example, the Toptica DLX110: [http://www.toptica.com](http://www.toptica.com)
which provides approximately 20mW of injection seed with linewidth typically below 300kHz. Multiple beams from several ECDLs can be combined, thereby amplifying several wavelengths simultaneously. The seed laser is focused onto the input facet of the TA diode using a high numerical aperture (NA) lens\(^5\) mounted in a fixed \(x - y\) position with adjustable \(z\)-focus via a fine (M12×0.5) thread. The TA is injection locked to the seed, and output beam astigmatism (caused by the diode shape) is corrected using a double lens system. The first high NA aspheric lens\(^6\) corrects the very fast vertical divergence of the output. In the horizontal plane, this results in a beam waist approximately 1” from the output facet. A second cylindrical lens placed at its focal distance from this waist corrects the horizontal divergence. While the unit has been designed to use fixed \(x - y\) mounts, if \(x - y\) adjustment is desired the high NA lens mounts can easily be replaced with a modified \(x - y\) fibre positioner\(^7\). The second output lens can be adjust for optimum beam size and quality, but is nominally an \(f = 80\text{ mm}\) cylindrical lens. The output passes through an optical isolator with fixed \(x - y\) position before being used in the experiment, to prevent damage to the diode caused by optical feedback. Although the use of fixed position elements may seem restrictive at first glance, they offer the advantage of very stable long-term operation and reliability.

The TA diode is mounted on a commercially supplied “SDL” mount, on which the case and an insulated pin provide electrical connections to the c-mount diode. Temperature stabilisation is achieved using a 50W thermoelectric cooler (TEC) and a thermistor temperature sensor for active feedback. The

\(^4\)MOGlabs rev. 3: http://www.moglabs.com/
\(^5\)Thorlabs C230TM-B f=4.5 mm 0.55NA
\(^6\)Thorlabs C330TM-B f=3.1 mm 0.68NA
\(^7\)Newport 9051(M) Fiber Launcher
hot side of the TEC is in contact with a plate which can be water cooled in warm ambient environments, although water cooling is not required in normal laboratory conditions. Electrical connections to the unit are via 9- and 25-pin D-Sub connectors on a single-layer PCB, incorporating a diode protection circuit, safety interlock connector, and connections for the TEC, thermistor and laser diode.

Our design offers several advantages over commercial systems. By locking to a seed laser, we can effect small changes in the laser frequency by altering the seed laser beam, for example using an acousto-optic modulator (AOM). This is far more efficient than altering the amplified beam directly, as it avoids loss of beam power due to the non-optimal efficiency of the AOM. Such a technique has been utilised to change the detuning of our cooling lasers, as described in Ch. 4.

In addition, we can use our existing frequency locking techniques and equipment to stabilise the seed laser to an atomic transition, avoiding the additional cost of purchasing often inferior frequency locking electronics. The design has proved useful for colleagues at the National Institute of Standards Technology (NIST) in Gaithersburg, Maryland USA, who have used it to successfully produce high power lasers for experiments on quantum squeezing and laser cooling.

A diagram of the MOT cooling, repump and excitation lasers is shown in Fig. 3.5. All continuous wave lasers for the experiment are located on a separate optical bench and transported to the vacuum chamber via single mode optical fiber. Two external cavity diode lasers (ECDLs) are locked relative to the $5S_{1/2}(F = 3) \rightarrow 5P_{3/2}(F' = 4)$ transition (“cooling” and ”excita-
Figure 3.3: Rendered CAD image (top) and photograph (bottom) of the tapered amplifier, with case removed. The beam direction is from bottom right to top left.
3.1 Design

Figure 3.4: Schematic of shifted saturated absorption spectroscopy locking technique used to frequency stabilise ECDLs. Lenses and waveplates have been omitted for clarity. The locking beam is split between a double-pass AOM and the rubidium vapour cell. The AOM-shifted beam then counter-propagates through the vapour cell, resulting in a Doppler-free saturated absorption spectrum that is shifted by the frequency at which the AOM is driven. The AOM driver is frequency modulated at 250 kHz to create an AC locking signal. Demodulation of the detected photodiode (PD) signal provides feedback to the ECDL. The laser frequency can be easily shifted by changing the AOM driving frequency, for example via a voltage controlled oscillator.

Fig. 3.6 shows the arrangement of the horizontal cooling beams at the vacuum

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8The tapered amplifier presented here was used successfully for approximately eight months, after which the commercial laser diode failed. To avoid delays to the experiments, a Toptica DLX110 was modified for use as an injection locked tapered amplifier, and used while the replacement diode was awaiting delivery.
Figure 3.5: Schematic of the cooling and excitation ($5S_{1/2}(F = 3) \rightarrow 5P_{3/2}(F' = 4)$), and repump ($5S_{1/2}(F = 2) \rightarrow 5P_{3/2}(F' = 3)$) lasers. The seed cooling laser is used to injection lock a tapered amplifier (TA), the output of which is sent to the MOT through single mode optical fibre with the repump beam. AOMs allows fast switching of all beams for timed experiments.

chamber. After exiting the fiber, the beam immediately passes through a polarising beam splitter, which ensures that any polarisation drift due to misalignment of the fiber polarisation axis causes an equal intensity drifts on both arms of the horizontal beams. During early experiments, such drifts caused slow variations ($\sim 20$ s) in the MOT position and density, resulting in large shot-to-shot variation in results. The addition of this PBS ensures that any polarisation drifts do not change the relative power balance between the two horizontal arms of the MOT beams, thereby improving stability considerably. The beam is then split and collimated to a $1/e^2$ width of approximated 4cm using $f = 75$ mm lenses. The power of each beam is approximately 20 mW at the vacuum viewport.

The vertical MOT beam is also collimated using a $f = 75$ mm lens before entering the chamber through the bottom port. The beam is retro-reflected at the top of the chamber.
**Figure 3.6:** Schematic of the horizontal MOT cooling beams at the vacuum chamber. BB: Beam block. The first polarising beam splitter immediately after the fiber is to avoid *relative* intensity drifts between the two main arms, which can be caused by polarisation drift due to misalignment of the polarisation maintaining fiber axis.

### 3.2 Construction

The design was finalised in early 2008 and sent to manufacturers (MDC, USA) for production. The estimated date of delivery for the completed vacuum chamber was in early May 2008, however due to numerous mistakes by the manufacturers the parts were not delivered until late November 2008.

During the wait for delivery of UHV components, a new laboratory space was commissioned as part of the project, which had to be set up for experimental physics work. This involved installation of optical benches, overhead equipment racks, power supplies, IT and networking infrastructure, furniture, water cooling, compressed air supplies, blackout curtains and safety equipment. Although a daunting task, over the second half of 2008 our group successfully prepared this new laboratory for the ultracold plasma project with minimal workshop assistance.

Construction of the UCP apparatus proceeded in three stages: UHV assem-
bly, chamber bakeout, and assembly of the optical and electronic components around the chamber. Figs. 3.7 and 3.8 show the laboratory in mid-2008 (before the chamber was assembled), assembly of the (internal) accelerator plate structure and vacuum system, the system during bakeout, and the operational UCP system.

UHV bakeout is an essential but time consuming process. In any UHV system, gas molecules (mostly water) desorb from the internal walls of the chamber at some outgassing rate. If the chamber remains at room temperature, the system will reach a quasi-equilibrium state when the effective pumping rate is limited to the outgassing rate. If we heat the chamber, thereby temporarily increasing the available thermal energy, the molecules will desorb from the walls faster, and the outgassing rate will increase. This results in the pressure temporarily increasing, and with it, the effective pumping speed. When the system is returned to room temperature after some time, the outgassing rate will be much lower, since most of the contaminants have been pumped away. A new quasi-equilibrium is established at lower pressure, since much of the water has been removed.

It is important to ensure than the temperature during bakeout remains as uniform as possible, since any “cold spots” will act as a trap for contaminants. To maintain a uniform temperature during bakeout, a multi-channel temperature and pressure logging system was created. The system automatically monitored the temperature at ten locations on the chamber, together with the pressure and current readings from the two ion pumps, during the entire four-week bakeout. In addition, it allowed temperature monitoring and automated alerts for sensitive areas, such as near ion-pump magnets and anti-reflection coated windows, which can be damaged if heated excessively.
Figure 3.7: Construction of the UCP laboratory and apparatus. Top: UCP lab after installation of optical benches and mains power. Middle: UCP vacuum components (foreground) and ionisation laser (background) after installation of drop-down equipment racks, water cooling and networking. Bottom: accelerator structure (left) mounted inside the main experimental chamber (right).
Figure 3.8: Insulated UCP vacuum system during bakeout as seen from oven end (top left), after bakeout from opposite end (top right) and operating UCP system with optics (bottom).
The system consisted of an SRS SR630 Thermocouple controller with RS232 interface connected to a PC running Ubuntu Linux. Device communications were programmed using Perl, which was in turn called from a series of shell scripts. Raw data output was plotted in real time using gnuplot and shared on-line using Apache web server such that it could be viewed from any worldwide internet connection in real time. Automated monitors sent alerts to lab users via email and SMS text message if either the temperature or chamber pressure was outside a designated range. Chamber pressures and ion pump currents were monitored via a second RS232 interface to a Terranova Model 752 Ion pump controller.

The chamber was heated using a combination of flange-mounted heater bands and heater tapes, powered by variable-AC power supplies. The entire chamber was insulated using standard foil-backed fiberglass wool insulation. A plot of the temperature for all ten channels over the four week bakeout is shown in Fig. 3.9. The pressure and corresponding ion pump current, is shown in Fig. 3.10. The chamber heaters were turned off on July 20th, when the temperature and pressure are observed to drop rapidly to a their final equilibrium levels.

The first ions from the system were observed on November 9th 2009, with electrons following soon after, on November 11. The entire UCP system was constructed from an empty laboratory in under two years.
Figure 3.9: Plot of temperature at points on the chamber indicated on the CAD schematic, during UCP system bakeout. The sharp drop on the “All metal turbo valve” on July 6th was due to a nearby shutter approaching a dangerous temperature. To reduce the temperature at the shutter, some insulation had to be loosened, resulting in the temperature drop.
Figure 3.10: Plot of ion pump current, and corresponding pressure, for the two ion pumps used during UCP system bakeout. The “oven” pump is a 60 L/s pump while the “main” pump is 500 L/s, connected to the main chamber via an 8” flange. The smaller ion pump was turned off and the gate valve between the oven and Zeeman slower was closed on July 9th, when a sharp rise in current was detected. This was later found to be due to a faulty transformer in the ion pump controller.
3.3 Electronics and timing

The UCP experimental sequence is shown in Fig. 3.11. Low voltage TTL signals (3.3V) are generated by a 24-channel digital timing card\(^9\). A TTL input from the flashlamp on the ionisation laser triggers the start of the sequence, which begins with a pulse of 780 nm light to excite atoms to the 5P state. A few microseconds later the ionisation laser Q-switch is triggered, the plasma is created, and electrons (or ions) are extracted by a DC electric field applied to the first stage accelerator. Pulsed, two-stage acceleration is possible using this apparatus, but is yet to be implemented. The MOT and Zeeman slower cooling lasers and their magnetic fields are then switched on to load the trap for the next cycle. A few milliseconds before the cycle restarts, the lasers and magnetic fields are shut off, and the cloud can be imaged using one of the techniques described in Chaps. 4 and 5. The timing card waits for the next trigger from the flashlamp to begin the cycle again. The MOT loading time is adjusted to ensure that the total time for an experimental sequence matches the repetition rate of the laser.

The UCP experiment requires fast switching of the magnetic fields of the MOT anti-Helmholtz coils and the Zeeman slower coil. Fast switching of magnetic fields remains a challenge for modern experimental physics. As the magnetic field collapses, it induces a *counter* electromotive force, or “back-EMF” voltage across the coil which, instantaneously, can be orders of magnitude larger than the original driving voltage. The back EMF, \( \epsilon \), is related

\(^9\)Spincore Pulseblaster PCI mounted in external USB enclosure.
3.3 Electronics and timing

Excitation pulse (5 $\mu$s)
Ionisation pulse (5 ns)
Q-switch delay (~ 280 $\mu$s)
MOT + Zeeman slower, (~ 97 ms)
t_{\text{off}} (~ 2 ms)
Imaging strobe (~ 20 $\mu$s)
CCD shutter (~ 1.5 ms)

**Figure 3.11:** Experimental timing sequence for electron extraction from UCP. The TTL output from the flashlamp was used as a trigger input to start the sequence, which was repeated at 10 Hz. The total time of a single sequence was maintained at 100 ms by adjusting the MOT loading time when other parameters were changed.
to the inductance of the coil, $L$, and the change in current, $I$, by [98]

$$\epsilon = -L \frac{dI}{dt} \quad (3.2)$$

To collapse the field quickly, (i.e. maximising $dI/dt$) the switching circuit must allow the coil to generate a large back-EMF.

In addition to the back-EMF, nearby conducting objects, such as the vacuum chamber itself, can act as secondary windings (much like in a transformer), with additional eddy currents induced in the conducting material as the primary field collapses. The total magnetic field can be much slower to dissipate than the driving current, simply due to the presence of the vacuum chamber [98]. To minimise eddy currents, slits were cut in the stainless steel former around which the Zeeman coil is wound. Further reduction of eddy currents requires active damping of the magnetic field via feedback to the driving current. Future electron diffraction experiments may require such active damping, or alternatively loading of a non-magnetic atomic trap (e.g. dipole trap [99]) prior to ionisation, thereby relaxing the requirement of fast magnetic field switching.

Experimentally, the coils are switched using high-current field-effect transistors\(^\text{10}\) (FETs). Fig. 3.12(a) shows the circuits used to connect the TTL signals from the timing card to the coil circuit. During normal operation, the standard diode ensures that current flows only through the coils. When the FET is opened (by a TTL high - the circuit is inverting) the back EMF results in reverse current flow through the Zener diode, and fast dissipation of the energy stored in the coil.

\(^{10}\)IXYS IXFN200N10P Polar high-power MOSFET 200A 100V
Figure 3.12: Circuit for computer control of MOT and Zeeman slower coils during experimental sequence.

A similar circuit is used for switching the magnetic field of the Zeeman slower, Fig. 3.12(b) [83]. The Zeeman slower uses two coils connected in series to generate positive and negative sections of magnetic field. To balance the current between the coils, an additional FET is used as a “shunt” to allow current to flow through the FET instead of the second coil. By adjusting the gate voltage on the shunt, either manually or using a computer controlled analog output, the magnetic field can be adjusted to maximise loading rate at the MOT.

The measured decay of the magnetic fields is shown in Fig. 3.13. The Zeeman
slower field decays less rapidly, most likely due to a combination of eddy currents in the stainless steel tube around which the coil is wound, and the higher current. The fields decay to below background levels in approximately 1.5 ms.

![Magnetic field decay](image)

**Figure 3.13:** Decay of the magnetic field of the Zeeman and MOT coils when switched off by the TTL signal. The magnetic field was measured approximately 1 cm from the top anti-Helmholtz coil, using a linear hall effect sensor (634SS2). The MOT coils generate a field gradient of 10 G/cm at the atom cloud.

A control program to run the experiment was created using the National Instruments Labview environment to interface with the various hardware components. The primary task of the software is to allow user-friendly adjustable programming of the digital timing card. In addition, the UCP control program interfaces with CCD cameras for measurement of electron signals on the MCP and imaging of the atom cloud. It also includes image analysis tools to allow image processing and optimisation, an implementation of the inversion algorithm for diffraction contrast imaging (Ch. 4), control of the ionisation laser wavelength and power, and software control of the various shutters and AOMs used in the experiment.
3.4 Experiment

Our approach to the production of shaped electron bunches depends on spatial variation of the excitation and ionisation lasers. The procedure for achieving shaped laser intensities using a spatial light modulator (SLM) is described in Sec. 3.4.1. In addition, we require knowledge of the initial atomic density distribution, acquired via imaging as described in Sec. 3.4.2. Such imaging could in future be used to continuously optimise the electron cloud, by adjusting the excitation laser intensity profile to compensate for any measured atomic density fluctuations. In Sec. 3.5, this feed-forward technique is demonstrated to enhance the uniformity of the bunch considerably. In addition, the detected electron signal can also be used to feed-back to the laser intensity profiles. Continuous measurement and algorithmic adjustment of the ionisation process has potential to allow unprecedented control over the electron bunch shape, which would in turn result in significant improvements in brightness and coherence. The experiments presented here use a simplified geometry to demonstrate the techniques required to achieve these gains, and include the first demonstration of shaped cold electron bunches. Future work is anticipated to implement the algorithmic optimisation techniques and produce a compact electron source capable of diffractive imaging.

Section 1.2.2 described how our apparatus can produce electron bunches that are shaped in three dimensions using two shaped laser beams. As a proof-of-principle experiment which demonstrates the advantages of cold electron bunches, the experiments presented here shape only the excitation laser beam, resulting in an electron bunch that is shaped in two dimensions. The ionisation laser illuminates the entire cloud, so that the electron density
is defined by the combination of the excitation laser intensity profile and the atomic density. The advantages of cold electrons using two-dimensional bunch shaping will only be enhanced by control of the third dimension in the future.

### 3.4.1 Intensity shaping

Figure 3.14 shows the arrangement for producing an intensity shaped excitation beam. A 780 nm laser locked to the $5S_{1/2}(F = 3) \rightarrow 5P_{3/2}(F' = 4)$ transition is transported to the experimental optical bench via single mode fibre. The beam is expanded to a $1/e$ radius of approximately 5 mm and approximately 5 mW is incident on the SLM\(^{11}\) at near-normal incidence.

The SLM is computer-controlled via a standard DVI interface. Eight-bit grey-scale values on each pixel are mapped to $0 - 2\pi$ effective phase shifts internally. The Holoeye SLM was supplied with proprietary software which uses an optimised Gerchberg-Saxton algorithm to calculate the phase pattern required to produce an arbitrary far-field intensity distribution. Phase patterns were also initially calculated using a standard two-plane Gerchberg-

\(^{11}\)Holoeye Pluto-NIR2 1920x1080 8\(\mu\)m pixels $0 - 2\pi$ 8-bit phase-only SLM, http://www.holoeye.com
3.4 Experiment

Figure 3.15: Production of diffracted intensity pattern from phase-only SLM. (a) Desired intensity distribution (logo of the ARC Center of Excellence for Coherent X-ray Science, "CXS"), (b) measured intensity distribution and (c) phase pattern (cropped)

Saxton algorithm [100], but the Holoeye software was found to be adequate. The Holoeye software also allowed adjustment of additional superimposed phase patterns, such as Fresnel lens phase, prism (grating) phase and lateral offsets in real time, after the initial calculation of the phase mask. This provided real time adjustment of diffraction efficiency, beam direction, position and focal plane during experiments, and was used to produce the results presented here.

After diffraction from the SLM, the excitation beam was divided using a polarising beam splitter, between the experimental chamber and an optically-identical diagnostic path which enabled imaging of the beam shape as it would be at the atom cloud inside the vacuum chamber. Diffraction efficiency was typically 40%, resulting in a shaped beam power of 2 mW in the shaped beam. An example of the desired intensity distribution, calculated required phase pattern, and measured intensity pattern is shown in Fig. 3.15. The
excitation beam entered the vacuum chamber through a view-port opposite the MCP, propagating along the acceleration direction to the atom cloud (Fig. 3.16. A slight downward angle on the beam prevented it from being incident on the MCP at the far end of the chamber.

The MCP\textsuperscript{12} is mounted on a standard 4.5” conflat vacuum flange which is attached to the main chamber. One side of the MCP is biased with 1850 V and the polarity set appropriately for electrons or ions, while the other side is electrically grounded. To measure temporal charged particle signals, the ground-side of the MCP can be connected to a 50Ω-coupled oscilloscope. An electrically grounded, gold-coated wire mesh mounted in front of the MCP shields incoming charged particles from the MCP bias voltage. At the rear of the MCP is a phosphor screen which allows the electron distribution to be imaged. For analysis, images of the phosphor are acquired using an $f = 50 \text{ mm}$\textsuperscript{13} lens and CCD\textsuperscript{14} camera, controlled using the Labview software detailed in Sec. 3.3. A ring-shaped permanent magnet was inserted between the MCP and the 50 mm lens to reduce the divergence of the bunch, caused by the defocussing effect of the aperture in the accelerator structure. At the position of the MCP, the field strength was approximately 50 G. In addition, transverse movement of the magnet relative to the beam axes allowed control of the bunch position on the MCP. The magnet was used in Figs. 3.20 and 3.22.

\textsuperscript{12}Photonis Chevron, 10 µm pore size, $10^7$ electron gain at 2400 V (maximum) Model number: APD 3025FM 12/10/12 I 60:1 P20 4.5”FM

\textsuperscript{13}Sigma 50 mm $f/1.8$ AF

\textsuperscript{14}Apogee Alta U2000 1600x1200 pixels
3.4.2 Imaging

Imaging of the cloud provided the required knowledge of the spatial distribution of atoms in the MOT, for creation of shaped electron bunches corrected for the actual cloud shape, described in Sec. 3.5.1. Two imaging beams were used, at 780 nm and 776 nm for ground- and excited-state imaging respectively. The lasers and their frequency-stabilisation mechanisms, along with the imaging techniques themselves are described in Ch. 4 and 5.

For all images presented here, the imaging beam (either 780 nm or 776 nm) expanded from a single-mode optical fibre and was collimated to a $1/e^2$ radius of 5 mm. An additional $f = 100$ mm achromatic doublet lens was inserted to focus the beam for SCPI imaging, as explained in Ch. 5. After passing through the vacuum chamber, images were acquired with an interline transfer CCD$^{15}$ and a 200 mm lens.$^{16}$ Imaging was performed along an orthogonal axis to both the excitation and ionisation laser beams. While imaging would ideally be performed on the same axis as excitation (to allow direct compensation for specific atomic density variations) the MCP detector obstructed the view in this geometry. However, since the atomic density distribution will remain roughly Gaussian, imaging along the orthogonal axis can still provide enough information to determine the $1/e$ radius of the cloud, which (as shown later) allows first order correction for creation of uniform density electron bunches. Later experiments will allow imaging along the excitation axis and thus higher order corrections to atomic density fluctuations.

Figure 3.17 shows initial images of the atom cloud, using several different imaging techniques. The observed variation in cloud size and shape, result-

$^{15}$Apogee Alta U2000 ML 1600x1200 pixels
$^{16}$Nikon micro-Nikkor AF 200mm f/4
Figure 3.16: Experimental arrangement for demonstration of shaped electron bunches. (a) Side-view showing 480 nm ionisation and 780 nm imaging beams, orthogonal to acceleration direction. In future experiments, the excitation beam will counter-propagate along the same axis as the imaging beam, for feed-forward of atomic density information. (b) Top-down section view through (a), showing shaped excitation beam (see also Fig. 3.14), cold atom cloud and electron extraction to the MCP and phosphor. A permanent magnet refocused the bunch on the MCP. The phosphor was imaged using a 50 mm lens and CCD.
Figure 3.17: Diagnostic images of the cold atom cloud. (a) SCPI image, \( z = 20 \text{ mm}, \) 1.5\( \Gamma \) probe detuning, 30\( \mu \text{s} \) imaging pulse. (b) Fluorescence image, 5 ms exposure. (c) Ground state diffraction contrast image, \( z = 140 \text{ mm}, \) 2.5\( \Gamma \) probe detuning, 30\( \mu \text{s} \) imaging pulse. (d) Excited state diffraction contrast image, \( z = 150 \text{ mm}, \) \( \Delta_{776} = 2.0\Gamma, \Delta_{780} = 1.5\Gamma. \) (e) Ground state absorption image, 30\( \mu \text{s} \) imaging pulse.
ing primarily from optimisation of cooling laser frequency and alignment, clearly demonstrates the need for imaging of the atomic distribution prior to creation of shaped electron bunches. The SCPI image (Fig. 3.17a) exhibits more evidence of etalon fringes than the other techniques, resulting from the additional lens in the imaging beam. As explained in Ch. 5, this technique is best suited to imaging inhomogeneous clouds, which we anticipate in future experiments. For homogeneous clouds, diffraction contrast imaging remains the most robust and reliable imaging technique. The fluorescence image is clearly saturated - a common problem with this technique, even at the minimum exposure time of our CCD camera. Nevertheless, it provides a useful indication of cloud size. The resonant absorption image shows good contrast, but results in heating of the cloud and, as demonstrated in Ch. 5, the quantitative output is very sensitive to probe laser detuning and defocus. By far the most successful imaging technique is diffraction contrast imaging, shown in Figs. 3.17(c) and 3.17(d) for ground and excited state distributions respectively. DCI avoids the problem of saturation encountered in fluorescence imaging and, due to the detuning of the probe laser, results in less heating of the atom cloud. In addition, the large defocus distance enables easy subtraction of unwanted fluorescence, which appears as a defocussed background, so that unlike in-focus absorption imaging, the trapping lasers can remain on during imaging. From the ground state image, the cloud size is $1.7 \pm 0.3 \text{mm}$ full width at half maximum (FWHM) on the narrow axis and $3.0 \pm 0.4 \text{mm}$ on the long axis with a peak atomic column density of $3.4 \times 10^{13} \text{m}^{-2}$. Assuming a similar Gaussian density distribution along the imaging axis, the peak atomic density is then approximately $1.2 \times 10^{10} \text{cm}^{-3}$, with a total of $3.0 \times 10^8$ atoms, consistent with similar rubidium traps [2].
The excited state image shows an excitation fraction of approximately 40%. This image was taken using the MOT cooling beams to excite the atoms, resulting in a slightly higher excitation fraction than we expect when using the dedicated (and lower intensity) excitation beam. As explained in Ch. 4, the signal to noise ratio remains an issue for excited state imaging in low density traps. Future work to implement an optical dipole trap, to increase the electron bunch charge significantly, will simultaneously improve the imaging signal.

### 3.4.3 Temperature Measurement

When the cooling beams are switched off, the atom cloud will expand due to the thermal velocity of the atoms in the trap. The r.m.s. thermal velocity is related to the temperature of the atom cloud, $T$:

$$v_0 = \sqrt{\frac{2k_B T}{m_{Rb}}} \quad (3.3)$$

where $m_{Rb}$ is the mass of a rubidium atom. The radius of the atom cloud $R$ after a free expansion time $t$ is related to this velocity via

$$R(t) = \sqrt{R_0^2 + (v_0 t)^2} \quad (3.4)$$

where $R_0$ is the initial 1/e radius of the cloud, with spatial distribution modelled as a Maxwell-Boltzmann function $n(r) = n_0 \exp\{-r^2/R_0^2\}$ [101].

Using the Labview control system described in Sec. 3.3, 780 nm absorption images were taken with free expansion times ranging from 1 ms to 10 ms. Five images were taken at each delay time and the cloud radii determined
The Melbourne UCP

Expansion time (ms)

Cloud radius (mm)

\( v_0 = 116 +/- 9 \text{ mm/s} \)

\( R_0 = 0.84 +/- 0.01 \text{ mm} \)

\( T = 69 +/- 11 \mu\text{K} \)

Figure 3.18: Measured cloud radius versus free expansion time and fit to Eq. 3.4. Experimental parameters for all images were: 3 ms exposure, 40 \( \mu\text{s} \) imaging pulse duration, \( \sim 20 \text{ mW} \) per beam cooling beam power.

from a Gaussian fit. The average radius versus free-expansion time is plotted in Fig. 3.18 with a fit to Eq. 3.4. From the fit, the temperature of the cloud was \( T = 69 \pm 11 \mu\text{K} \), which is well below the Doppler cooling limit for \(^{85}\text{Rb} \) (\( T_D = 143 \mu\text{K} \) \(^{[102]}\)), indicating some polarisation gradient cooling \(^{[103]}\). Temperatures as low as 10 \( \mu\text{K} \) have been reported in the literature for similar \(^{85}\text{Rb} \) traps \(^{[104]}\), though usually with careful nulling of extraneous magnetic fields, optimisation of detuning and laser power and very low background pressure. Several mechanisms to reduce the temperature are well known and can be implemented at a later date should further cooling be required.

3.5 Results

Figure 3.19 shows images of the initial electron and ion signals on the MCP. In Fig. 3.19(a), the electron signal is the strong vertical line. The linear shape is due to the presence of the magnetic fields generated by the MOT coils. A
strong background signal is also present, believed to be due to ionisation laser light scattering off the vacuum view-ports and ionising background rubidium vapour which is not trapped in the MOT. Although anti-reflection coatings were specified to manufacturers, for both 480 nm and 780 nm wavelengths, the windows were delivered with only the 780 nm coating, leading to substantial 480 nm background. Figure 3.19(b) shows the improved electron signal after implementation of magnetic field switching described in Sec. 3.3. Although the shape of the bunch is much improved, the electron density is reduced due to the expansion of the MOT (and resulting decrease in atomic density) during the time between switching the magnetic fields off and ionising the atoms. The reduction in density can be avoid through implementation of a non-magnetic trap (e.g. dipole trap [99]) or faster damping of the magnetic fields using active feedback [98].

Due to their larger mass, ion bunches were far less susceptible to stray magnetic fields. Some distortion is still present (Fig. 3.19c), although the apparent symmetry suggests the distortion may be due to the combination of thermal expansion and space-charge effects. Time-of-flight measurements were conducted by monitoring the voltage on the ground-plane of the MCP, using a 50 Ω coupled oscilloscope. Example traces are shown in Fig. 3.19(d). For the acceleration voltages and flight distances listed in Fig. 3.19, the time-of-flight of both electron and ion bunches were seen to agree well with simple theoretical predictions of $90 \pm 14 \text{ ns}$ and $16 \pm 1.5 \mu \text{s}$ respectively, indicating that the apparatus operates within expected design parameters.
Figure 3.19: Preliminary electron (blue) and ion (red) observations (false colour). All images are 10-shot averages (1 s exposure duration). (a) Initial electron distribution prior to installation of magnetic field switching. The acceleration voltage was 30 V. The signal due to electrons from the cold atoms is indicated. Significant background noise was present due to scattered light from the ionisation laser. (b) Electron distribution at an acceleration voltage of 300 V, after installation of magnetic field switching. The time between field turn-off and ionisation pulse was 3.5 ms. (c) Preliminary ion distribution, 300 V acceleration voltage. (d) Ion (red, 300 V accelerating potential) and electron (blue, 60 V accelerating potential) time-of-flight measurements detected at the ground-plane of the MCP. The acceleration region distance was 2.5 cm and the free-flight distance from the accelerator to the MCP was 25 cm. The ion signal has been multiplied by a factor of 5 for clarity and the time axes are relative to the beginning of the 5 ns ionisation pulse.
3.5 Results

3.5.1 Shaped bunches

Using the spatial light modulator and the arrangement described above, the excitation beam was shaped into the intensity pattern shown in Fig. 3.15 and used to excite the atom cloud along the acceleration \((z)\) axis, immediately prior to the ionisation pulse. The experimental timing sequence used was the same as that shown earlier (Fig. 3.11).

Using a semi-classical model for a two level atom \([102]\), the transverse excited state density distribution \(\rho_{\text{ex}}(x,y)\) is proportional to the on-resonant excitation laser intensity, \(I(x,y)\).

\[
\rho_{\text{ex}}(x,y) = \frac{|\Omega(x,y)|^2}{\Gamma^2 + 2|\Omega(x,y)|^2} \rho(x,y)
\]  

(3.5)

where \(|\Omega(x,y)|^2 = \Gamma^2 I(x,y)/2I_{\text{sat}}\) and \(\Omega(x,y)\) is the Rabi frequency of the excitation transition with spatial dependence in the transverse plane. \(I_{\text{sat}}\) is the saturation intensity of the transition, \(\rho(x,y)\) is the column density of the ground state atom cloud and \(\Gamma\) is the transition linewidth. The ionisation laser illuminates the entire cloud with Rabi frequency \(\Omega_i\) and natural linewidth \(\Gamma_i\), but is resonant only with atoms in the excited state. As the ionisation laser couples the electron to a continuum state, the linewidth \(\Gamma_i\) is determined by the recombination rate \([105]\), which is the equivalent loss mechanism for the transition, analogous to spontaneous emission in a two-level atom. Furthermore, as shown in Sec. 3.5.2, the coldest electrons were produced through field ionisation of excited Rydberg states, for which the transition linewidth \(\Gamma_i\) is well defined \([106]\). For incoherent two-step ionisation, the initial transverse density distribution of the electron bunch \(\rho_{\text{e}^{-}}(x,y)\) is then proportional to the product of the excitation laser intensity and the
ground state atomic density.

\[
\rho_e(x, y) = \frac{|\Omega_i|^2}{\Gamma_i^2 + 2|\Omega_i|^2} \rho_{ex}(x, y) \tag{3.6}
\]

\[
\propto \frac{|\Omega(x, y)|^2}{\Gamma^2 + 2|\Omega(x, y)|^2} \rho(x, y). \tag{3.7}
\]

As described above, the electron bunch shape depends on the initial atomic density. Fig. 3.20 show the ground and excited state atom distributions and the resulting electron signal on the phosphor, for two different initial atomic density distributions. If we first consider the result from the highly asymmetric cloud (top row, a – c), it would appear that a surprisingly large portion of the desired electron bunch shape is apparent, perhaps more than would be expected given the extreme asymmetry of the atom cloud. It is important to remember however, that imaging was performed on an orthogonal axis, so that although the cloud appears too narrow to include the whole excitation beam illumination, it may be much wider in the longitudinal direction. It is for this reason that future experiments will image and excite along the same axis, so that atomic density variations can be directly accommodated. Nevertheless, the current geometry provides sufficient information about the size and shape of the atom cloud to enable a first-order correction for the atomic density, as demonstrated in Sec. 3.5.2. Without this information, compensation for atomic density variations and production of uniform density electron bunches would not be possible. The excited state distribution also displays some asymmetry. The difficulties associated with obtaining high signal-to-noise ratio (SNR) images of the excited state distribution in low density atomic traps would be mitigated by future implementation of
Figure 3.20: Shaped electron bunches produced using the excitation laser intensity pattern shown in Fig. 3.15. The top row (a – c) shows the electron bunch from an elongated asymmetric atom cloud and the bottom row (d – f) the result after adjusting the MOT alignment to minimise the cloud asymmetry and improve the bunch shape. Left column (a & d): Ground state atomic distribution imaged using DCI. Parameters were $z = 130\, \text{mm}$, $\Delta = 2.5\, \Gamma$. Centre column (b & e): Excited state atomic distribution imaged using DCI (see Ch. 4). Parameters were $z = 150\, \text{mm}$ defocus, $\Delta_{776} = 1.5\, \Gamma$, $\Delta_{780} = 1.5\, \Gamma$. Right column (c & f): Images of the resulting electron distribution on the MCP and phosphor. Both images are one second exposures (10-shot average) and have been normalised using the same factor, to illustrate the intensity variation in the electron signal. After optimisation of the excitation laser alignment, (g) and (h) show a 3 s exposure (30 shot average) and density plot of the shaped bunch. The Gaussian envelope of the atomic density distribution is clearly visible in (h).
a dipole trap for increased atomic density. As explained above, the resulting increased atomic density will simultaneously improve imaging SNR and electron bunch brightness.

Figure 3.20(d – f) show the results for a more symmetric atom cloud obtained by improving the alignment of the MOT cooling and trapping laser beams. The electron bunch density distributions are brighter and more evenly distributed. The excited state peak density is slightly higher than for the asymmetric cloud, which is reflected in the electron density. Although this example is slightly exaggerated, the comparison shown in Fig. 3.20 clearly illustrates the importance of the atomic density distribution when producing shaped electron bunches. Fig. 3.20(g) and (h) show a 30-shot average, with reduced background noise and a three dimensional plot of electron density. A Gaussian envelope is clearly visible superimposed on the binary excitation intensity distribution, caused by the Gaussian atomic density distribution. Compensation for atomic density variations is demonstrated in the following section.

Fig. 3.20 represents the first proof-of-principle demonstration of electron bunch shaping using an ultracold plasma source. It is important to note that although a shaped laser pulse could, in principle, produce 2D shaped bunches from a conventional photoemission source [45], the high electron temperature would result in significant diffusion of the bunch, so that arbitrary bunch shapes like those shown above would not be observable. Temperature effects are investigated explicitly in Sec. 3.5.2. Unlike UCP, shaping a photoemission source would not be generalised to three dimensions. By using an ultracold plasma to create the bunch, we begin with cold electrons with low emittance, which maintain the bunch shape as it propagates to the detector.
3.5 Results

High emittance sources have a larger phase-space distribution, resulting in increased diffusion of the bunch as it propagates and destruction of the initial pattern. As a result, observation of macroscopic shaped electron bunches and their subsequent optimisation is currently a unique capability of UCP sources.

3.5.2 Uniform bunches and temperature effects

The results above clearly demonstrate the effect of atomic density on the electron bunch shape. The bunch shaping approach inherently allows compensation for such atomic density variation, by adaptive variation of the excitation laser profile. To that end, an inverted Gaussian intensity pattern with a hard edge was created using the SLM with the aim of creating a 2D uniform density electron bunch. The image of the atomic density in Fig. 3.20(d) was used to estimate the size of the atom cloud, and hence the magnitude of the Gaussian intensity fluctuation required. The desired laser intensity pattern, required SLM phase mask and measured excitation laser intensity are shown in Fig. 3.21. A slight linear gradient is present across the measured laser intensity, which could be eliminated by future implementation of iterative refinement schemes [107]. The shaped beam illuminates the atom cloud as in Fig. 3.16.

Fig. 3.22 shows a series of images of electron bunches shaped using the excitation laser intensity in Fig. 3.21(b). Each image was taken at the ionisation laser wavelength indicated on the image. Lower wavelengths correspond to higher excess ionisation energies and therefore hotter electrons. The difference between the energy required for ionisation from the $5P_{3/2}$ state, $E_i$ [108],
Figure 3.21: (a) Desired intensity to produce 2D uniform electron bunches. (b) Excitation laser intensity distribution measured using CCD. (c) Required SLM phase pattern.
and the actual energy of the ionisation laser at the indicated wavelength, $E_\lambda$ is indicated as the excess ionisation energy $\Delta E = (E_\lambda - E_i)/k_B$. Units of Kelvin are used to indicate the relative scale of heating occurring due to excess ionisation energy, but do not reflect the absolute electron temperature. Line profiles from the $\Delta E = 171$ K and $\Delta E = -96$ K are shown in Fig. 3.23. The bunch at 480.675 nm (bottom left) appears to be the clearest, most defined bunch. This result is unsurprising since the bunch has the lowest excess energy, and should therefore be the coldest electrons. Bunches at lower wavelengths display blurred edges and smearing, characteristic of hotter electron temperatures as expected.

It is interesting to note that the optimum bunch shape and intensity occurs well below threshold, at $-96$ K. The wavelength required to ionise $^{85}$Rb from the $5P_{3/2}$ state is 479.06 nm [106], yet electron signals are observed using an ionisation wavelength over a nanometre longer than this. The shift in ionisation threshold is attributed to DC-field ionisation of high-lying Rydberg states [78].

The atoms were photoionised in a DC electric field of 160 V cm$^{-1}$. A classical saddle-point model using a Coulomb-Stark potential of $V(r) = -1/r - Fz$ [78,109,110,111] where $F$ is the electric field, predicts an ionisation threshold shift due to Rydberg field-ionisation of $\Delta E = -4R_y\sqrt{F/F_0}$ where $R_y$ is the Rydberg constant and $F_0$ is the atomic electric field unit ($e/4\pi\varepsilon_0a_0^2$). If $\Delta E$ is in units of cm$^{-1}$ and $F$ is in V/cm, then $\Delta E \approx -6.12\sqrt{F}$ [110]. The predicted energy shift corresponds to a shifted ionisation threshold wavelength of 480.9 ± 0.2 nm or an excess energy of $\Delta E = -110 \pm 15$ K = 9.6 ± 0.7 meV. Differences with respect to the theoretical shift are thought to arise primarily from stray fields which have not been measured or nulled, and systematic
error in measurement of the distance between the accelerator plates. The images in Fig. 3.22 are in good agreement with the predicted shift, in that an electron signal remains visible until the final image. The penultimate image at 481.001 nm is particularly dim, indicating that the threshold shift is 1 meV smaller than anticipated by the classical model. Slightly smaller shifts have also been reported in the literature for Ar and Cs Rydberg field ionisation measurements [110, 112].

Fig. 3.24 presents a quantitative examination of the effects of electron temperature on the spatial derivative of the bunch edge. The data points represent the acuity of the bunch edges in each image of Fig. 3.22. Gradients across the edge of the clouds were calculated, over a range of distances between 5 and 50 pixels. The gradients were calculated using a least-squares linear fit to the relevant section of a line profile from the image, similar to those in Fig. 3.23. This process was repeated for 100 line profiles from each bunch and the results averaged, so that each data point represents an average of 1000 gradients across the bunch edge. For each image, the mean intensity of the electron signal was used to normalise the entire image to unity before the gradients were calculated, to eliminate intensity variation effects. The x-axis represents excess ionisation energy, $\Delta E$, relative to direct ionisation $E_i$, which is proportional to the electron temperature.

The ionisation threshold is clearly visible near $\Delta E = -100\,\text{K}$ as predicted by the classical model described above. Field-ionised Rydberg atoms provide the clearest signal immediately above threshold, however the blurring at $-75\,\text{K}$ suggests that some heating is occurring. This can be understood via the energy level diagrams in Fig. 3.25. The electric field was constant for all measurements, so that its energy contribution to ionisation remained con-
3.5 Results

![Figure 3.22: 2D uniform electron bunches at increasing ionisation laser wavelength (decreasing energy). Excess field-free ionisation energies are shown in units of degrees Kelvin, but do not indicate electron temperature. No normalisation has been applied to the images, to preserve relative intensity information. The brightness of the entire $\Delta E = -116\,\text{K}$ image has been increased by 10\% for clarity. Parameters for all images were: 5.7 ms delay between switching magnetic field off and excitation pulse, 800 V acceleration voltage, 1 s exposure, 10 Hz repetition rate, 40 $\mu$s excitation pulse, 5 mJ ionisation laser pulse energy.](image-url)
Figure 3.23: Lineprofiles from uniform electron density distributions for warm ($\Delta E = 171$ K) and cold ($\Delta E = -96$ K) electrons. The profile of the bunch is drastically improved for cold electrons, even over this small temperature difference.

Figure 3.24: Electron bunch edge sharpness versus excess ionisation energy, relative to field-free ionisation energy. Error bars represent one standard deviation.
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stant. When the ionisation laser energy plus the field energy add exactly to the field-free ionisation energy (Fig. 3.25a), there is no excess energy to cause heating so we expect sharp bunch edges, as observed at $\Delta E = -96 \text{ K}$. When the ionisation laser energy was increased, but still below field-free threshold (Fig. 3.25b), the excess ionisation energy is imparted to the electron as kinetic energy (heat), blurring the bunch edges as observed at $\Delta E = -41 \text{ K}$. Increasing the energy of the ionisation laser further still results in a combination of field-free (direct) ionisation to the continuum and Rydberg field-ionisation via discrete Rydberg levels. The directly ionised electrons are cold, improving the sharpness again, but the field-ionised electrons are warm, resulting in only a slightly sharper bunch. This interesting result indicates that due to the contribution from field ionisation, the path to production of the coldest electron bunches may in fact be through field-ionisation of Rydberg atoms rather than direct ionisation. Of course, if the electric fields are pulsed on after ionisation, then the contribution from field-ionised Rydberg atoms would be minimised. Such speculation warrants further investigation in future work. The interactions and behaviour of Rydberg atoms is a large and dynamic field, beyond the scope of this thesis, but the reader is directed to reference [78] for further insight. Clearly, investigations into Rydberg atom production and field ionisation mechanisms will be critical in the long-term production of cold electron bunches.

An upper limit to the electron temperature can be extracted from a fit to the data in Fig. 3.24 which can then be used to estimate the emittance of the source $\epsilon_x$. To derive a fitting formula, we first assume that any transverse expansion of the bunch is due only to electron temperature, and that the accelerator and lens system acts only in the acceleration direction. This
Field ionisation of Rydberg atoms producing cold and warm electrons. (a) Ideal field ionisation producing cold electrons. (b) Excess ionisation laser energy, resulting in warm electrons.

Figure 3.26: Schematic of the electron path to the detector. The electrons are assumed to experience constant acceleration while inside the accelerator (distance $d_i$), then travel at constant velocity to the detector (distance $d_2$) by which time a point source has expanded to a size $\Delta x$ due to its initial temperature.
3.5 Results

assumption ensures that we extract an upper limit to the temperature: transverse expansion may in fact be caused by the electron transport system (and is in fact expected to be caused by the accelerator apertures), but its effect would be to lower the estimated temperature of the electrons. We now consider Fig. 3.26 which shows the expansion of a point source within the bunch as it propagates to the detector. Since the transport system is assumed to act only in the $z$-direction, the mean transverse velocity of the electrons will be given by $v_\perp = \Delta x / t$ where $t$ is the propagation time from the plasma to the detector. The total propagation time is given by $t = t_1 + t_2$ where $t_1$ is the propagation time inside the accelerator at constant acceleration and $t_2$ is the time of flight from the accelerator to the detector at constant velocity $v_2$. These times are easily derived using classical mechanics as follows, where $F$ is the electric field, $a$ is acceleration and $m_e$ is the electron mass. During acceleration, we have

$$m_e a = eF \quad (3.8)$$

$$d_1 = \frac{1}{2} a t_1^2 \quad (3.9)$$

$$\Rightarrow t_1 = \sqrt{\frac{2m_e d_1}{eF}}. \quad (3.10)$$

The time of flight from the accelerator to the detector $t_2$ is given by

$$t_2 = \frac{d_2}{v_2} \quad (3.11)$$

$$v_2 = \frac{eF}{m_e} \times t_1 \quad (3.12)$$

$$\Rightarrow t_2 = \frac{d_2m_e}{eF} \sqrt{\frac{eF}{2d_1m_e}} \quad (3.13)$$
so that the transverse velocity $v_\perp$ is

$$v_\perp = \frac{\sqrt{2d_1\Delta x}}{(2d_1 + d_2)\sqrt{\frac{eF}{d_1 m_e}}}. \quad (3.14)$$

The acceleration field is static, meaning that by the time the last electrons are ionised by the 5 ns laser pulse, the first electrons have already left the acceleration region. Coupled with the low initial density of the atom cloud, this means that Coulomb interactions within the electron cloud are negligible for DC acceleration fields. Any expansion is assumed to be purely due to thermal expansion of the cloud, with a transverse r.m.s. expansion velocity of

$$v_0 = \sqrt{\frac{2k_B T}{m_e}}. \quad (3.15)$$

Equating $v_0 = v_\perp$ and solving for $\Delta x$ as a function of $T$ obtains the relation

$$\Delta x = \frac{(2d_1 + d_2)}{d_1}\sqrt{\frac{d_1 k_B T}{eF}}. \quad (3.16)$$

The linear derivative measured in Fig. 3.24 is inversely proportional to $\Delta x$ so that $dI/dx = C_1/\Delta x$ where $C_1$ is a constant. Equation 3.16 then becomes

$$\frac{dI}{dx} = C_1 \frac{d_1}{(2d_1 + d_2)\sqrt{\frac{eF}{d_1 k_B T}}} \quad (3.17)$$

The clearest bunch shape achieved experimentally was at an excess ionisation energy of $\Delta E = -96$ K corresponding to our coldest bunch. Now assume that after adjustment for the ionisation shift, this bunch has an excess energy of $\Delta E' = \Delta E + 96K = 0$ K so that any remaining expansion of the bunch must be due to intrinsic heating of the electrons, for example due to the processes described in Ch. 2. Once again, if this assumption is invalid, the
3.5 Results

estimated temperature would be greater than the actual temperature, so that we are calculating an upper limit to the initial source temperature. The total electron temperature will then be \( T = T_0 + \Delta E' \) where \( T_0 \) is the minimum experimentally achievable electron temperature limited by intrinsic heating processes. Substituting this into Eq. 3.17 we obtain the following relation between the derivative of the bunch edge and the excess ionisation energy.

\[
\frac{dI}{dx} = C_1 \frac{d_1}{(2d_1 + d_2)} \sqrt{\frac{eF}{d_1(k_B(T_0 + \Delta E'))}} \quad (3.18)
\]

A fit of Eq. 3.18 to the data using \( C_1 \) and \( T_0 \) as free parameters is shown with the data in Fig. 3.27. The scaling parameter \( C_1 \) is effectively set by the higher \( \Delta E' \) data points and takes into account the unknown distortions of the electron transport system, while the initial temperature \( T_0 \) is set by the \( \Delta E' = 0 \) K data point. This is particularly elegant since any magnification caused by the accelerator would change only the absolute scale of the edge derivative as a multiplicative factor, rather than the relative change in edge acuity with temperature. As the scaling parameter \( C_1 \) accommodates this factor, the magnification of the accelerator is expected to have minimal effect on the calculated temperature limit \( T_0 \). When systematic errors are included with the fitting error, the resulting upper limit on electron temperature is \( T_0 \leq 35 \) K. Equation 1.5 for the one-dimensional transverse emittance,

\[
\epsilon_x = \sigma_x \sqrt{\frac{k_B T}{m c^2}}, \quad (3.19)
\]

gives an upper limit to the transverse emittance. Using the measured cloud size of \( \sigma_x = 2 \) mm the upper limit to source emittance is \( \epsilon_x = 0.15 \) mm mrad. This result compares very favourably with recent UCP results in the liter-
Figure 3.27: Electron bunch edge sharpness versus excess ionisation energy, with fit to Eq. 3.18. Error bars represent one standard deviation.

ature of 0.3 mm mrad [13] but it is purely a source emittance and does not take into account any emittance growth which may result from propagation through the electron transport system. It also assumes that the initial edge is infinitely sharp. Since the spatial derivative is typically of order several hundred $\mu$m and the optical diffraction limit much smaller ($10 \mu$m at this working distance) the finite initial bunch edge resolution is expected to have minimal effect on the result.

The source size $\sigma_x$ used to calculate the emittance limit is based on the diameter of the excitation laser beam. This size can be significantly reduced and is ultimately limited by optical access to the vacuum chamber and the size of the SLM. For diffraction limited optics at a working distance of 250 mm and using the entire surface of the SLM, we expect a minimum source size of approximately 50 $\mu$m, which would result in a much lower emittance of
3.5 Results

$3 \times 10^{-3}$ nm mrad.

While reducing the source size improves the emittance, it simultaneously reduces the number of electrons in the bunch. Since reduced electron number is undesirable, the brightness is a more useful measure of source quality for diffractive imaging applications. At this early stage of the experiment, a Faraday cup has yet to be incorporated into the apparatus, so that direct measurements of the total bunch charge are not possible. Nevertheless, we can estimate the achievable brightness based on reasonable excitation and ionisation fractions observed in the literature [13,71]. Beginning with $5 \times 10^8$ trapped atoms and using excitation and ionisation fractions of 30%, the total extracted charge should be 7 pC. For a pulse length of $\tau = 5$ ns this equates to a peak current of $I_p = 1.4$ mA. Eq. 1.7 for the transverse brightness in terms of the emittance,

$$B_{\perp} = \frac{I_p}{4\pi^2 \epsilon_\epsilon_y},$$

then gives a lower limit to the transverse brightness of $10^9$ A/m$^2$sr.

For diffractive imaging applications, we also require the coherence length to be of the same order as the sample size, $L_c > 10$ nm. The intrinsic coherence length of the electron source due to its initial temperature can be written as [12]

$$L_c = \frac{\hbar}{\sqrt{m_e k_B T}}.$$  (3.21)

For $T < 35$ K the coherence length at the source is $> 5$ nm.
Table 3.1: Summary of beam parameters. The predicted best UCP performance, and values marked with an asterisk, are based on reasonable extrapolation from experimental parameters, as described below.

### 3.5.3 Electron source comparison

Table 3.1 shows a comparison of source parameters for this work and existing electron sources, together with the requirements for ultrafast electron diffraction (UED). Carbon nanotube fieldemission sources operate in a low current continuous regime and are therefore unsuitable for use in diffractive imaging, but are included for completeness as they represent the current state-of-the-art in achievable brightness. The values shown are the transverse thermal emittance from Eq. 3.19, transverse brightness $B_\perp$, source coherence length (Eq. 3.21), source temperature $T_0$, peak current $I_p$ and temporal bunch length, $\tau$. It is important to realise that the parameters given in Tab. 3.1 represent only the intrinsic properties of the source, and do not include any degradation of beam quality which may occur during transport of the bunch to the sample, for example through emittance growth during acceleration. These effects are particularly problematic for photofield emission sources [40], as the extremely small emission area leads to exaggerated space-charge forces and associated emittance growth [11]. In contrast, due to the low temperature of the electrons, the UCP source can operate with a larger source size while maintaining brightness and coherence. Com-
combined with further suppression of emittance growth due to three dimensional shaping of the electron bunch, UCP is unique in its ability to produce high brightness, high coherence electron bunches for diffractive imaging. While standard photoemission sources can achieve remarkably high currents and very short bunch lengths, the large thermal emittance remains well beyond that required for UED.

Table 3.1 includes the estimated achievable brightness and coherence of the Melbourne UCP source, following implementation of the improvements suggested earlier. These improvements include a dipole trap for increased density through reduced source size (100 µm), together with improved (possibly coherent) excitation and ionisation processes to increase the total bunch charge. After optimisation, we expect $5 \times 10^9$ atoms in the initial magneto-optical trap. When the atoms are transferred into an optical dipole trap [99] we expect some loss, so that for a worst-case scenario, approximately $10^7$ atoms are anticipated to remain. The estimated best parameters listed in Tab. 3.1 are based on 50% excitation and ionisation fractions of these atoms, together with reduction of the electron bunch length below 100 ps using compression during acceleration [9]. These parameters indicate that although some additional work is required to achieve the ultimate potential of the apparatus, the Melbourne UCP can meet or exceed the requirements of diffractive imaging, through implementation of well established experimental techniques.
Chapter 4

Excited State Diffraction Contrast Imaging

4.1 Preface

The material presented in this chapter is published work [46]. The paper explains the new diffractive imaging technique developed to obtain images of excited state atom distributions which are used to optimise electron bunch shaping as described in Sec. 3.5.

State-selective imaging of cold atoms
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Abstract: Atomic coherence phenomena are usually investigated using single beam techniques without spatial resolution. Here we demonstrate state-selective imaging of cold $^{85}$Rb atoms in a three-level ladder system, where the atomic refractive index is sensitive to the quantum coherence state of the atoms. We use a phase-sensitive diffraction contrast imaging (DCI) technique which depends on the complex refractive index of the atom cloud. A semiclassical model allows us to analytically calculate the detuning-dependent refractive index of the system. The predicted Autler-Townes splitting and our experimental measurements are in excellent agreement. DCI provided a quantitative image of the distribution of the excited-state fraction, and compared with on-resonance absorption and blue cascade fluorescence techniques, was found to be experimentally simple and robust.
4.2 Introduction

We have developed an off-resonant imaging technique to investigate atomic coherence phenomena such as electromagnetically induced transparency (EIT) [32], coherent frequency upconversion [33], and “slow light” [36] in a magneto-optical trap (MOT). These processes have typically been studied using techniques without spatial resolution. Imaging offers the potential for obtaining additional information. For example, capture and storage of three-dimensional light fields using EIT [37] could benefit from imaging to provide spatial information about the atomic coherence of the atoms involved. Combining diffraction-based phase imaging with control of the internal state of the imaged atoms using a probe laser, we could potentially explore techniques for enhancing the imaging (e.g. by modifying the refractive index), or for directly measuring the control process itself.

Imaging of the distribution of excited-state atoms has become of interest recently, for example to control the formation of samples of cold Rydberg atoms [78,114,115] with defined spatial distribution. Cold Rydberg gas is created by laser cooling and trapping atoms and then exciting them to a Rydberg state, usually in a two-step process which relies on an excitation laser to provide the first excitation step from the ground state (see Fig. 1). The shape of the Rydberg sample can be controlled by spatially profiling the laser beams for either excitation process. Excited-state imaging can provide the feedback to control this process, for example in dipole blockade [116,117] and coherent excitation [118] experiments.

Imaging feedback is also needed for creating ultra-cold plasma (UCP) with controlled spatial distribution [1]. UCP is created by photoionization of an ultra-cold atom cloud, leaving a plasma with very low initial electron (and ion) temperature, which could allow generation of a very high brightness electron beam [52]. It has been calculated [9] that the emittance of such a source could be optimized by controlling the initial spatial distribution of the electrons, which is determined by the distribution of excited-state cold atoms and the spatial profile of the photoionization laser. Thus emittance optimization will require imaging of the excited-state atom distribution, with feedback to the spatial profiles of the excitation and photoionization lasers.
Figure 4.1: Simplified Rb level scheme showing Rydberg states, ionization threshold, $5S - 5P - 5D$ ladder system used for state-selective imaging, and $6D$ state which leads to blue fluorescence at 420 nm. Atoms are cooled and maintained in the excited $5P$ state by the 780 nm laser. A 480 nm beam will excite to Rydberg states or photoionize the atoms, producing cold electrons. The excited-state atom distribution was determined by imaging the $5P - 5D$ transition at 776 nm.

Conventional imaging techniques such as on-resonant absorption imaging are simple and effective. However, absorption imaging is inherently destructive to the cold atom cloud, and critically sensitive to experimental parameters such as defocus, detuning of the imaging laser, and optical alignment. For excited-state imaging, these difficulties are compounded by the need for filters to remove unwanted fluorescence from the image, which introduce noise and interference fringes. Phase imaging techniques, including Zernike phase-contrast, are experimentally demanding and quantitative only for a limited range of phase shifts. We have previously demonstrated diffraction contrast imaging (DCI) \[119\], an off-resonant imaging technique based on retrieving the object phase from Fresnel propagation of a diffracted imaging beam. The technique is quantitative, minimally destructive, and less sensitive to experimental parameters than either conventional absorption imaging or classical phase imaging.

Excited-state imaging is experimentally challenging regardless of the imaging method. The expected incoherent excitation fraction of the $5P$ state is limited to 50% and the natural linewidth of the 776 nm excited-excited
transition is only 600 kHz, approximately ten times smaller than the ground-excited 780 nm transition. The narrow linewidth, and hence slow decay rate from the 5D state, limits the absorption of the 776 nm beam. Coupled with the smaller population in the 5P state, it results in low relative absorption (5%) of the 776 nm imaging beam. In combination, the signal-to-noise ratio for simple 776 nm absorption imaging is expected to be 20 times lower than for 780 nm. Fortunately, high atomic density and small cloud size, which are inherently desirable for many applications of interest including UCP production and EIT imaging, enhance the imaging contrast, particularly for diffractive imaging (DCI).

4.3 Theory

Figure 4.2 shows the arrangement for diffraction contrast imaging. An off-resonance probe laser beam incident on a cloud of cold atoms experiences a spatially dependent absorption and phase shift, and then propagates to a spatially resolving detector, such as a CCD camera, which records the object diffraction pattern. Algebraic linear inversion of the Fresnel diffraction relation in Fourier space returns a quantitative measurement of the column density \( p(x) \) of the sample for transverse spatial coordinates \( x \equiv (x, y) \). The column density of the object is defined as the integral of the atomic number density, \( N(r) \), along the optical path, \( z \):

\[
p(x) = \int_{-\infty}^{0} N(r) \, dz,
\]

where \( r \equiv (x, y, z) \) and \( z = 0 \) is to the right of the cloud, as in Fig. 4.2. The relation between the Fourier transform of the normalized contrast of the

![Figure 4.2](image-url)

**Figure 4.2:** Diffraction contrast imaging records the diffraction pattern of an off-resonant plane wave incident on a cold atom sample.
diffraction pattern, $\mathcal{F}\{(I - I_0)/I_0\}$, and the Fourier transform of the column density at the object, $\mathcal{F}\{p(x)\}$, is given by [119]:

$$\mathcal{F}\left\{\frac{I - I_0}{I_0}\right\} = 2k \left[ \delta \sin(\pi \lambda z u^2) - \beta \cos(\pi \lambda z u^2) \right] \mathcal{F}\{p(x)\} \quad (4.2)$$

where $k = 2\pi/\lambda$, $\lambda$ is the wavelength of the probe laser, $z$ is the propagation distance and $u$ is the spatial frequency conjugate to $x$. $\delta$ and $\beta$ are the phase and absorption coefficients, where $\phi = k\delta p$ and $\mu = k\beta p$ are the phase shift and absorption of the atomic cloud. The refractive index of the atom cloud is then:

$$n(x) = 1 + p(x) \frac{\sigma_0 \lambda}{4\pi} (\delta + i\beta). \quad (4.3)$$

The cross section for the imaging transition, $\sigma_0$, is defined in terms of the total electron angular momentum quantum numbers $J$ and $J'$ of the ground and excited states of the transition [120]:

$$\sigma_0 = \frac{(2J' + 1) \lambda^2}{(2J + 1) 2\pi} \quad (4.4)$$

where we sum all allowed hyperfine levels and magnetic substates for any given probe polarization. A general explanation of the calculation of the refractive index of an atomic gas is given in App. 6. Quantitative retrieval of the column density requires knowledge of the absorption/phase ratio, $\beta/\delta$, for all transverse position coordinates. Previous work [119] has assumed a two-level atom approximation to derive an analytic absorption/phase ratio depending only on the probe detuning. For excited-state imaging, a two-level approximation is not appropriate due to the perturbing effect of the excitation laser field. Rubidium atoms in the 5P state decay to the ground state via spontaneous emission with a lifetime of approximately 26 ns, much shorter than typical imaging exposure durations (10 – 100 $\mu$s). To maintain atoms in the 5P excited state, the 780 nm cooling/excitation light must remain on, perturbing the atomic eigenstates.
4.3 Theory

4.3.1 Complex refractive index

Treating the atom-field interaction semiclassically, the refractive index can be determined using a density matrix formalism [121,122]. The density matrix elements were calculated using optical Bloch equations [123] (OBEs) for three-level atoms (5S, 5P, 5D; see Fig. 4.1) with two coupling laser fields. The system is described by six coupled differential equations, which can be solved by numerical integration for steady-state conditions, but an analytical expression for the density matrix elements was achieved by recasting the optical Bloch equations into vector form [124]:

$$\frac{\partial \tilde{\rho}}{\partial t} = \mathbf{M} \cdot \tilde{\rho} \tag{4.5}$$

where $\mathbf{M}$ is a $9 \times 9$ matrix for a three level system. Since we want the steady-state solution, the left hand side of Eq. (4.5) is $\tilde{0}$. Finding the steady-state solution amounts to finding the nullspace of the matrix $\mathbf{M}$, simplifying the problem considerably. The density matrix elements, and hence the refractive index, are given by the eigenvectors of the system.

The exact solutions for both the real and imaginary parts of the susceptibility were found using this method, each of which contained approximately 100 terms. In the weak coupling limit on the 776 beam, a first-order Taylor expansion gives a more manageable result for the absorption ratio, $\beta/\delta$:

$$\frac{\beta}{\delta} = \frac{\Gamma_{776} (\Gamma^2 + 4\Delta_{776}^2) + \Gamma \Omega_{780}^2}{2\Delta (\Gamma^2 + 4\Delta_{776}^2) - 2\Delta_{776} \Omega_{780}^2} \tag{4.6}$$

where $\Gamma_{\lambda}$ and $\Delta_{\lambda}$ are the linewidth and detuning of the transitions at wavelength $\lambda$, $\Gamma = \Gamma_{780} + \Gamma_{776}$, $\Delta = \Delta_{780} + \Delta_{776}$, and $\Omega_{780}$ is the Rabi frequency for the 780 nm laser [108]. The absorption ratio depends entirely on known parameters: beam detunings, natural linewidths, and the cooling Rabi frequency, which can be calculated from the measured excitation beam intensity. The calculated absorption and phase components of the 776 nm susceptibility are shown in Fig. 4.3, for $\Delta_{780} = -3\Gamma$. Autler-Townes energy level splitting [125,126] is evident in the predicted spectrum of the 776 nm laser, caused by the strong perturbing influence of the applied 780 nm field. The energy level splitting ($\Delta E$) increases with 780 nm detuning and with 780 nm
power \cite{127}:

\[ \Delta E = \hbar \sqrt{\Omega_{780}^2 + \Delta_{780}^2}. \]  

(4.7)

To maximise the phase shift and thus imaging contrast, the 776 nm probe must be detuned, e.g. to \(+40\Gamma_{776}\). Although not investigated here, imaging as a function of \(\Delta_{776}\) provides a detailed measurement of the Autler-Townes effect, with spatial resolution. Note that near resonance, the 6 MHz linewidth of the 780 nm transition broadens the effective linewidth of the 776 nm transition. The broadening is reduced by any detuning of the 780 nm excitation beams, so that the expected SNR for excited-state imaging remains low.

\begin{figure}[h]
\centering
\includegraphics[width=0.5\textwidth]{figure4.3.png}
\caption{Analytical result for the phase and absorption on the 776 nm transition for a three-level model of Rb, in the weak coupling limit on the 776 nm beam. The two-level results (dashed) are clearly not valid for excited-state imaging.}
\end{figure}

A comparison of the first-order approximation (Eq. 4.6) with the exact result is shown in Fig. 4.4, for typical experimental parameters. The energy level splitting shows good agreement. Note that in our image retrieval results we use the exact result for the ratio.

\subsection{4.3.2 Simulations}

The term in square brackets in equation (4.2) is the contrast transfer function (CTF), \(h(u, z)\):

\[ h(u, z) \equiv \delta \sin(\pi \lambda z u^2) - \beta \cos(\pi \lambda z u^2). \]  

(4.8)
Figure 4.4: Calculated absorption and phase for the 776 nm transition, showing exact analytic (solid) and first-order approximation (Eq. 4.6; dashed), for typical experimental parameters ($\Delta_E = -3\Gamma$, $\Omega = 30$ MHz).

Spatial frequencies corresponding to the zero values in the CTF must be regularized during reconstruction of the column density, to avoid division by zero. This is done by means of the Tikhonov method [128] which smoothly interpolates across the poles. The Tikhonov filtered CTF is given by

$$\frac{1}{h'(u, z)} = \frac{h(u, z)}{h(u, z)^2 + \alpha^2},$$

(4.9)

where a large value of the Tikhonov parameter, $\alpha$, increases SNR, but reduces fine detail and increases error in the retrieved column density. A value of $\alpha = 0.1$ to 0.3 is generally acceptable [129].

To determine the accuracy of the retrieval process, a simulation was performed using diffraction patterns calculated from known input column densities. Immediately after the atom cloud, the output wave (at $z = 0$) is described by multiplying the incident wave, $f_0$, by the complex transmission function of the cloud:

$$f(x, z = 0) = f_0 \exp \left[-\mu(x) + i\phi(x)\right].$$

(4.10)

The wave field at an image plane ($z > 0$) is found by Fourier transforming, multiplying by the Kirchoff propagator $\bar{h}(u, z)$, and inverse Fourier transforming:

$$f(x, z) = \mathcal{F}^{-1} \left\{ \mathcal{F} \left\{ f(x, z = 0) \right\} \bar{h}(u, z) \right\}.$$

(4.11)
The plane wave free-space propagator is given by

\[ \tilde{h}(u, z) = \exp \left[ 2\pi iz \sqrt{\frac{1}{\lambda^2} - u^2} \right]. \]  

(4.12)

The column density was then retrieved from the calculated diffraction pattern using equation (4.2), and compared to the known input column density. A typical diffraction pattern and the retrieved column density is shown in Fig. 4.5. The incident beam and the atom cloud were constructed and placed asymmetrically within the image array to ensure that any boundary artefacts of the fast Fourier transform were immediately apparent. The input column density was defined as two overlapping Gaussian column density distributions with different dimensions and peak column densities. The retrieved column density shows good agreement with the initial column density, with noticeable differences introduced only at the peaks of the atom cloud. This error can be reduced significantly by reducing the value of the Tikhonov parameter, \( \alpha \), as shown in Fig. 4.6, but at the cost of increased high spatial frequency noise. The FWHM of the retrieved column density profiles remains consistent with the input column density over a much larger range of \( \alpha \).

4.4 Apparatus

Experiments were performed with a magneto-optical trap (MOT) in a \( \sigma^\pm \) (circularly polarized) arrangement [102, 2]. Cooling and trapping were performed on the \( 5S_{1/2} F = 3 \rightarrow 5P_{3/2} F' = 4 \) hyperfine transition of \( ^{85}\text{Rb} \), with external-cavity diode lasers (ECDLs) frequency stabilized using saturated absorption spectroscopy [130,131,132]. A semiconductor tapered amplifier [133] provided a total cooling beam power of 150 mW at the MOT, after fiber coupling. The cooling beam diameter at the MOT was approximately 20 mm (1/e²). Anti-Helmholtz coils produced a quadrupole magnetic field with a gradient of 10 G/A cm, with a maximum coil current of 6 A. An additional 780 nm repump laser, locked to the \( F = 2 \rightarrow F' = 3 \) hyperfine transition, was co-propagated along one axis of the cooling beams. Rubidium vapor was produced by a dispenser source.
4.4 Apparatus

Figure 4.5: Simulation of diffraction contrast imaging, showing input excited-state column density (left), calculated excited-state diffraction pattern at $z = 130 \text{mm}$ (center), and retrieved column density (right). Vertical column density line profiles through the peak are shown below. The 776 nm probe beam was simulated with a Gaussian intensity profile of 5 mm FWHM, detuned $2 \Gamma_{776}$ from absorption resonance. The retrieval was regularized with $\alpha = 0.2$.

Imaging probe beams at 780 nm and 776 nm were produced with two additional ECDLs. The 780 nm imaging laser was frequency stabilized to the $F = 3 \rightarrow F' = 4$ transition using saturated absorption spectroscopy. The 776 nm imaging laser was frequency-stabilized using Doppler-free two-colour spectroscopy (Fig. 4.7). The 776 nm locking reference beam was counter-propagated through a heated rubidium vapor cell ($50^\circ \text{C}$), coaxial with a frequency-shifted 780 nm reference produced from the stabilized 780 nm imaging laser via a double-pass acousto-optic modulator (AOM). The AOM frequency was modulated at 250 kHz, and the lock signal generated by demodulation of the dispersive absorption signal of the 776 nm beam. The 776 nm beam could be detuned easily and accurately by varying the 80 MHz AOM carrier frequency. The laser frequencies were offset from their respective resonances to allow for the frequency shift of single-pass AOMs used to provide high-speed switching of the imaging beam. The 776 nm and 780 nm beams were coupled into the same fiber, so that comparable ground and excited state images could be acquired consecutively, using either the 776 nm or 780 nm
Figure 4.6: Normalised error (difference divided by sum) in the peak column density retrieved from simulated excited-state diffraction patterns at a propagation distance of 130 mm, for the object shown in Fig. 4.5. The shaded region indicates the typical range of $\alpha$ values used. The dashed line indicates the value used in experiments.

Excitation to the 5P state was provided by the cooling and trapping lasers, which remained on during imaging.

At the vacuum chamber, the imaging beam from the fiber was collimated to produce a Gaussian beam with $1/e^2$ diameter 10 mm (Fig. 4.8). A 200 mm lens (micro-Nikkor AF 200 mm f/4) allowed the effective defocus distance to be varied, enabling both DCI and in-focus imaging to be performed with minimal changes to the apparatus. Images were taken with a CCD (Apogee Alta U2000 ML 1600 × 1200 pixels).

Atoms excited to the 5D$_{5/2}$ state can decay to the 6P$_{3/2}$ state, emitting 5 $\mu$m infrared and 420 nm blue light (Fig. 4.1). The 420 nm fluorescence is proportional to the population in the 5D$_{5/2}$ state and thus proportional to the 776 nm absorption. Blue fluorescence images were obtained by directing a 776 nm excitation beam off the imaging axis, to avoid saturating the CCD, via a removable mirror.

780 nm fluorescence created a strong background, substantially reducing the image contrast for 776 nm in-focus absorption imaging, and completely saturating blue fluorescence imaging. To separate the imaging probe beam at 776 nm from the fluorescence at 780 nm, i.e. only 4 nm separation, a very narrow bandpass filter was required. An effective filter was created from a heated rubidium vapor cell. Absorption of the 780 nm fluorescence was much
stronger than on the 776 nm probe beam, and a 10 cm vapor cell at 85°C reduced the 780 nm fluorescence to negligible intensity in a one second exposure. The temperature dependence of the 776 nm and 780 nm attenuation is shown in Fig. 4.9. In addition, a hot/cold filter (Thorlabs FM03, < 10% transmission below 630 nm) was used during 776 nm absorption imaging to remove any blue fluorescence. Both filters were removed for 780 nm imaging.

Initial 776 nm imaging results exhibited poor signal-to-noise ratio (SNR). To increase the atomic density, a compressed MOT (CMOT) technique was employed [134]. Initially, the coils (and hence the trap) were off, to record a background image. The rubidium dispenser was then driven at high current (5 A), the cooling beam detuning set at $-6 \text{ MHz} (-\Gamma_{780})$ and the coils set at 1 A (10 G/cm) for 30 s to load the trap. The coil current was ramped to a variable maximum (up to 6 A) over times ranging from 5 ms to several seconds. An image was acquired, and the dispenser then returned to a low current (2.5 A) to minimize background gas load, and the coils turned off. The values given are those which optimized the density of the cold atom cloud for our apparatus, limited by a background pressure of $10^{-9} \text{ Pa}$.

It is common to use an additional cooling step with far detuned light, prior to
Figure 4.8: Schematic of the imaging beam path and MOT vacuum chamber. Only two of the six cooling beams are shown.

Figure 4.9: Attenuation of 780 nm and 776 nm light by a heated 10 cm long vapor cell narrow-bandwidth atomic filter.
4.5 Results

Figure 4.10: 780 nm absorption images showing compression due to increase in magnetic field gradient. Uncompressed (left): field gradient 10 G/cm. Compressed (right): 100 ms at 50 G/cm. Cloud full width at half maximum from Gaussian fit, 735 ± 20 µm and 322 ± 12 µm respectively. Both images: 10 ms exposure, 20 µs pulse duration, 1.5 mW probe beam power.

A comparison of 780 nm images taken before and after compression is shown in Fig. 4.10. Spatial compression of the MOT is clearly apparent, with a peak magnetic field gradient of 50 G/cm compressing the MOT to approximately half the initial diameter, and a peak density up to four times higher. The compression was negligible for field gradients below 30 G/cm and also showed only marginal improvement for higher field gradients of 60 G/cm, consistent with findings reported elsewhere [134].

4.5 Results

4.5.1 Autler-Townes

Our theoretical prediction of Autler-Townes splitting was first tested by measuring the total MOT blue fluorescence, using an integrating detector (not shown in Fig. 4.8), consisting of a photomultiplier and camera lens (Nikkor AF 50 mm f/4) filtered using a 420 ± 10 nm interference filter. The blue fluorescence was recorded as the 776 nm laser was scanned through a frequency range of approximately 180 MHz. The measurement was performed for several 780 nm cooling beam detunings, from −25 MHz to −10 MHz. Example spectra and theoretical predictions are shown in Fig. 4.11. The dependence on cooling beam power was also verified separately. From Eq. (4.7) we ex-
Figure 4.11: Theoretical (dashed) and experimental (solid) 420 nm fluorescence in the MOT for 780 nm cooling beam detunings of $-10.0 \pm 0.5$ and $-20 \pm 3$ MHz.

Expect the energy splitting to be less sensitive to cooling beam intensity than detuning, since $\Delta E \propto \Omega_{780} \propto \sqrt{I_{780}}$. This relative insensitivity to beam power explains the close fit of the data, despite the apparent complexity of the system, and also reduces the uncertainty in our imaging results which might arise from fluctuations in cooling beam power. The small shoulder on the right edge of the large peak in the $\Delta_{780} = -20$ MHz spectrum is believed to be due to hyperfine splitting of the 5D state. The observed separation of approximately 10 MHz is consistent with published values of the 5D hyperfine splitting [135]. The broadening of the $-10$ MHz line is due to the larger size of the MOT at this detuning. A larger cloud size results in increased Zeeman detuning across the spatial extent of the atom cloud, broadening the observed fluorescence peaks. The effect is reduced for the much smaller cloud produced by the $-20$ MHz cooling beam detuning.

The cooling beam diameter (20 mm) was much greater than the size of the atom cloud (1 mm). Combined with the relative insensitivity to beam intensity, this minimized any spatial dependence of the refractive index caused by the Gaussian intensity profile of the cooling beams. The excellent agreement for the Autler-Townes splitting provides confidence that our Bloch equation model can reliably calculate the detuning dependence of the refractive index for the 776 nm transition.
4.5 Results

Figure 4.12: Blue fluorescence image (false color) of the uncompressed MOT for 780 nm excitation and 776 nm probe. 1 second exposure with 21 G/cm magnetic field gradient, 85°C vapor cell filter temperature. The fluorescence intensity is shown for a profile along the dashed line, with a Gaussian fit.

4.5.2 Blue fluorescence

Blue fluorescence images of the uncompressed MOT were taken using the heated rubidium vapor cell to remove 780 nm fluorescence. The hot/cold filter was removed for these images, and the trapping beams remained on at all times. An example image and line profile are shown in Fig. 4.12. Due to the long exposure time required, fluorescence images showed considerable blurring, due to the movement of the cold atom cloud on the time scale of the imaging exposure. This lowered the spatial resolution and prevented quantitative imaging.

4.5.3 Excited state absorption

On-resonant in-focus 776 nm absorption images of the compressed cloud were also obtained (Fig. 4.13). Considerable noise and interference fringes were caused mainly by the many uncoated optical surfaces of the vapor cell and hot/cold filter used in the beam line. The best images were obtained for exposures of 150 µs with probe power approximately 150 µW. From the line profile in Fig. 4.13, the SNR ≈ 3, considerably lower than an equivalent ground-state image. This was expected given the relatively low absorption (5%) on the 776 nm beam. Using a Gaussian best fit to the column density line profile, the peak column density calculated from the excited state ab-
Excited State Diffraction Contrast Imaging

Figure 4.13: In-focus absorption image of atoms in the 5P state using 776 nm probe beam. Experimental parameters: 60 G/cm peak magnetic field gradient, 4 ms compression, 300 μW probe beam power, 300 μs pulse duration, 1 ms exposure, 85°C filter cell temperature. Line profile (right) and Gaussian best-fit through the dashed line in image.

Absorption image is $(3.1 \pm 0.3) \times 10^{13}$ atoms m$^{-2}$. The peak column density of the ground-state atom cloud is $(7.0 \pm 0.4) \times 10^{13}$ m$^{-2}$; thus the excited state fraction is $44 \pm 15\%$.

4.5.4 Excited state diffractive contrast

An excited-state diffraction pattern and its column density reconstruction are shown in Fig. 4.14, together with a ground-state image for comparison. The excited state population is $45 \pm 6\%$, which is in agreement with the absorption image measurement. Using a two level approximation for our experimental beam powers and detunings, we expect a theoretical excited state fraction of $46\%$, in good agreement with our results.

The FWHM of the excited-state cloud, as determined by both DCI and absorption imaging, is much smaller than that of the ground state cloud (0.42 mm versus 1.18 mm). This difference can be attributed to the combination of the narrow linewidth of the 776 nm transition and the strong magnetic field gradient used to compress the cloud. From Fig. 4.11, the effective linewidth of the 776 nm transition is approximately 3 MHz for our experimental parameters. The FWHM of the ground state cloud is 1.18 mm, corresponding to a Zeeman shift of about $\pm 3$ MHz at the half-peak-density edges of the cloud, for a field gradient of 60 G/cm. The Zeeman shift changes the effective detuning of the probe beam at the outer edges of the cloud, decreasing the absorption and phase shift of the probe light. We anticipate
Figure 4.14: Ground and excited state diffraction patterns, column density reconstructions and column density line profiles. Experimental parameters for ground state image: 60 G/cm peak magnetic field gradient, 4 ms field ramp, $-6$ MHz maximum cooling beam detuning, 850 $\mu$W probe beam power, 30 $\mu$s pulse duration, 1 ms exposure, $+2.5 \Gamma_{780}$ probe detuning, 120 mm effective defocus. For excited state image: 60 G/cm peak magnetic field gradient, 4 ms field ramp, $-6$ MHz maximum cooling beam detuning, 150 $\mu$W probe beam power, 100 $\mu$s pulse duration, 1 ms exposure, $+12.5$ MHz probe detuning ($+2 \Gamma_{776}$ from absorption peak), 130 mm effective defocus.
that future work to implement fast switching of the magnetic field coils will enable a direct measurement of this effect.

The DCI reconstructions were less sensitive to noise and interference fringes than in-focus imaging, particularly in the ground-state image. For DCI, the imaging system is inherently defocused from the atom cloud, and so fluorescence (780 nm and 420 nm) will not be in focus. The vapor cell and hot/cold filters were therefore not required, and their interference effects and reflection losses were removed. Results were consistent using defocus distances between 40 and 150 mm and detunings up to $5\Gamma$ from the main absorption peak. Due to the narrow linewidth of the 776 nm transition, the reduction in sensitivity to imaging beam detuning compared to absorption imaging makes DCI a robust measurement technique for excited-state imaging.

Excited state DCI also offers advantages over other techniques for measuring the excited-state fraction, including recoil-ion momentum spectroscopy (RIMS) [136], as it does not require additional equipment such as ion beams or RIMS detectors. Our method shows promise for our intended direct application (production of shaped ultra-cold plasmas) due to its experimental simplicity and minimally destructive interaction. Additionally, by measuring the time evolution of the excited state fraction and distribution, DCI has the potential to provide valuable information on processes such as stimulated Raman adiabatic passage (STIRAP) [137], and to allow measurement of the evolution of atomic coherence effects with spatial resolution.

4.6 Conclusion

Diffraction-contrast imaging offers a simple yet powerful method for imaging cold atom samples, providing spatial information on atomic coherence processes and effects. Quantitative imaging requires knowledge of the ratio of the real to imaginary components of the refractive index of the sample. We have derived an analytic expression for the three-level two-laser system using a semi-classical approach. The model predicts Autler-Townes splitting of the rubidium 5P excited state, which was found to be in good agreement with experimental measurements.
4.6 Conclusion

We have demonstrated and compared several techniques for state-selective imaging of cold atoms in a MOT. Blue fluorescence imaging provided good SNR, but poor resolution. Excited-state DCI achieved a good SNR without the need for filters, and showed reduced sensitivity to imaging beam imperfections and experimental parameters such as detuning and defocus. The excited state fraction was found to be $45 \pm 6\%$, in agreement with theoretical predictions. Conventional (destructive) absorption imaging provided a comparable image, but required a more complicated experimental setup. The need for insertion of filters and a vapor cell in the imaging beamline increases noise and interference fringes in absorption images.

The DCI technique can be adapted to provide spatially resolved measurements of atomic coherence phenomena, such as electromagnetically induced transparency and “slow light”, or expanded to investigate more complex multi-level atomic systems. More directly, the technique has been applied in feedback control of the spatial distribution of an ultra-cold plasma, for production of shaped electron bunches outlined in Ch. 2.
Chapter 5

Single-plane Curved-beam Phase Imaging

5.1 Preface

The following chapter is published work [47]. Additional unpublished supporting material is presented at the end of the chapter (Sec. 5.8). The additional material includes spectroscopic measurements of electromagnetically induced transparency in cold atoms (Fig. 5.7), a brief overview of the method used to solve the transport of intensity equation, details of the calculation and elaboration on application of the error correction factor $\eta$ (Fig. 5.6), and additional technical information about the imaging process.

Non-iterative imaging of cold atoms using phase retrieval from a single diffraction measurement

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5.2 Introduction

Measurement of the spatial distribution of cold atoms has become increasingly important with the development of ultracold atom trapping and experiments in a diverse range of applications. Atomic coherence processes, such as generation of entangled states in the Rydberg blockade regime [34, 35], Electromagnetically Induced Transparency (EIT) [138] and slow light [139] are all typically investigated using simple probe beam techniques which do
not provide spatially resolved information. More recently, transfer of optical information between spatially separated Bose-Einstein condensates [140] has provided further motivation for investigation of more complicated atomic coherence phenomena in cold gases. Imaging of the atomic distribution can potentially provide increased information on the underlying processes.

More directly, we are interested in imaging of cold atoms as a means of controlling electron bunch shapes generated from Ultracold Plasma (UCP). UCP is produced by photoionization of a cold atom cloud, resulting in cold ions and electrons [13]. Several laser fields, each with a spatially varying intensity profile, will interact with the cold atoms to produce the UCP. The effect of these fields on the refractive index of the cloud will result in an inhomogeneous atom cloud. Imaging of the cloud can provide feedback to control the final electron density distribution and ultimately the electron brightness, via changes in the ionization laser intensity profiles. Electron bunches extracted from an UCP are potentially much brighter than conventional thermal sources [9] and in the long-term offer the possibility of sufficient brightness for single-shot diffractive imaging of bio-molecules.

Conventional imaging techniques for cold atoms involve absorption of a probe laser tuned to the atomic resonance [141]. An in-focus absorption “shadow” image is recorded, and knowledge of the atomic refractive index is used to retrieve a spatial map of atomic column density. While experimentally simple, such absorption imaging is very sensitive to experimental parameters such as the frequency stability and detuning of the illuminating laser beam and defocus of the imaging optics. For example, an inadvertent half-natural-line-width detuning from resonance results in a factor of three error in the resulting atomic density measurement. Furthermore, for high-density clouds such as Bose-Einstein Condensates (BECs), saturation of the absorption signal can result in an inaccurate quantitative measurement.

Phase imaging techniques, such as Zernike phase-contrast imaging, offer an alternative solution which greatly reduces this sensitivity, but this comes at the cost of increased experimental complexity and such techniques are quantitative only for a limited range of phase shifts. Diffraction Contrast Imaging (DCI), [119,142,143] offers an experimentally simpler method of quantitative phase imaging. DCI computationally inverts off-resonant diffraction data to
provide a map of atomic density. The inversion relies on the assumption of an *homogeneous* object, that is, one having a refractive index which varies only with the density, without any inherent spatial dependence. Processes involving atomic coherence typically rely on many laser fields interacting with the cold atoms, each of which have spatially varying intensity profiles. The effect of these laser fields on the atomic refractive index results in an *inhomogeneous* atom cloud. Accordingly, such objects of interest cannot be imaged using DCI, even with knowledge of the spatial variation of atomic refractive index.

To enable imaging of inhomogeneous objects, we use techniques developed in the context of x-ray phase contrast imaging and coherent diffractive imaging [144], adapted to imaging of cold atoms. Using a non-iterative phase retrieval method, we quantitatively extract the atomic column density of an atom cloud from a single plane of diffraction data, given knowledge of the atomic refractive index. In addition, we use a previously measured column density to extract the complex refractive index of an inhomogeneous cloud. The Single-plane, Curved-beam Phase Imaging method (SCPI) retains the experimental advantages of diffractive imaging techniques like DCI, while avoiding the restrictive homogeneous object requirement.

### 5.3 Method

A conceptual diagram of the imaging technique is shown in Fig. 5.1. The atom cloud is defined in terms of the atomic column density, \( \rho(\mathbf{r}) \), which is given by the integral of the atom number density \( N(\mathbf{r}, z) \) along the optical path \( z \):

\[
\rho(\mathbf{r}) = \int_{-\infty}^{0} N(\mathbf{r}, z) dz \tag{5.1}
\]

where \( z \) is in the direction of propagation and \( \mathbf{r} \) is the transverse coordinate. For a thin object, the exit surface wave \( \psi(\mathbf{r}, z_1) \) is related to the incident field \( \psi_0(\mathbf{r}, z_1) \) via the transmission function, \( T(\mathbf{r}) \):

\[
\psi(\mathbf{r}, z_1) = \psi_0(\mathbf{r}, z_1) T(\mathbf{r}). \tag{5.2}
\]
5.3 Method

Figure 5.1: Conceptual arrangement for Single-plane Curved-beam Phase Imaging. A phase curved Gaussian beam is incident on the atom cloud at plane $z_1$, diffracts and propagates a distance $Z$ to a CCD detector at plane $z_2$.

The transmission function can be written as:

$$T(r) = \exp \left[ ik \{ \alpha(r) + i\beta(r) \} \rho(r) \right]$$  \hspace{1cm} (5.3)

where $\alpha(r)$ and $\beta(r)$ are the spatially varying real and imaginary parts of the complex atomic refractive index, $k = 2\pi/\lambda$ and $\lambda$ is the wavelength of the probe laser beam. Hence the atomic column density is given by:

$$\rho(r) = \frac{1}{k} \left\{ \frac{\| \ln (T(r)) \|^2}{\alpha^2(r) + \beta^2(r)} \right\}^{\frac{1}{2}}.$$  \hspace{1cm} (5.4)

Alternatively, if the density is known and the refractive index variation is desired, rearranging Eq. 5.3 gives an expression for the complex refractive index in terms of the known column density, $\rho(r)$:

$$\alpha(r) + i\beta(r) = \frac{1}{ik \rho(r)} \ln \left( \frac{\psi(r, z_1)}{\psi_0(r, z_1)} \right).$$  \hspace{1cm} (5.5)

We measure the intensity at the detector plane, $|\psi(r_2, z_2)|^2$. The central section of the diffraction data is essentially a point projection hologram, while high-angle scatter provides additional information. To retrieve the exit surface wave, $\Psi(r_1, z_1)$, we also require the phase of the wave at the detector,
which we can obtain using the Transport of Intensity Equation (TIE) [145]:

$$\nabla_\perp \cdot I(r, z) \nabla_\perp \Phi(r, z) = -k \frac{\partial I(r, z)}{\partial z}. \quad (5.6)$$

The TIE is a continuity equation which relates the phase of a wave, $\Phi(r, z)$, to its intensity, $I(r, z)$, and longitudinal intensity derivative, $\partial I/\partial z(r, z)$, using the condition of energy conservation. The measured diffraction pattern is the required intensity input. Conventionally, the derivative term is found by taking two diffraction images separated by some small propagation distance, then approximating the derivative using finite difference methods [146]. Solving the TIE provides the phase of the wave at the detector plane, and back-propagation yields the complex wave-field in the sample plane [147, 148].

The key to the approach described by Quiney et al. [144] is to use a priori information about the unperturbed imaging beam, to calculate an approximate intensity derivative from a single diffraction image. Adaptation of this method to imaging of cold atom clouds provides the advantage that unlike other methods which solve the TIE using a single diffraction image [149], this approach does not assume a homogeneous object.

The calculation of the approximate longitudinal derivative follows the method of Quiney et al. [144]. For an axially aligned Gaussian beam (as in Fig. 5.1), we describe the unperturbed wave at the detector plane as $\psi_0(r_2, z_2) = \exp(-\zeta r^2)$. The real part of $\zeta$ is related to the $1/e$ radius of the beam, $r_0$, by $\text{Re}(\zeta) = 1/r_0^2$. As shown by Quiney et al. [144], the functional relation between the intensity of the unperturbed wave at the detector and its longitudinal derivative is given by $H[I_0]$:

$$H[I_0] \equiv \frac{\partial I_0}{\partial z} = -\frac{\zeta}{Z} \int \int w^2 g_0(w) \exp \left( \frac{2\pi iw \cdot r}{\lambda Z} \right) dw$$

where $Z = z_2 - z_1$ and $g_0(w)$ is the autocorrelation of $\psi_0(r_1, z_1) \exp(i\pi r_1^2/\lambda Z)$. The autocorrelation $g_0(w)$ is the inverse Fourier transform of the unperturbed intensity $I_0(r_2, z_2)$, and $w$ is the transverse coordinate of the autocorrelation. The central approximation of the method is to assume that the functional
form of Eq. (5.7) remains unchanged for the perturbed beam, i.e.

\[
\frac{\partial I}{\partial z} \approx H[I].
\]  

(5.8)

To state this explicitly, we assume that \( H[I] \) is of the same form as in Eq. (5.7) so that we can replace the unperturbed intensity measurement used to calculate \( g_0(w) \) in Eq. (5.7) with the perturbed diffracted intensity measurement. This assumption is essentially perturbative: as long as the absorption and phase shifts caused by the object are small and slowly varying, with no discontinuities in their derivatives, the approximation in Eq. 5.8 demonstrates good agreement with the exact result. In addition to these requirements, the illuminating beam must have significant curvature at the object plane, typically requiring a Fresnel number of five or greater. Such curvature ensures that the far-field diffraction pattern is essentially a magnified version of the short-distance Fresnel diffraction pattern, so that the TIE can be solved using standard techniques for short propagation distance diffraction [150].

These requirements are all readily accommodated in the context of cold atom imaging. By detuning the probe laser from resonance, we have the advantage of being able to effectively tune the absorption and phase shifts imposed by the object. In addition, cold atom clouds are, by design, smooth Gaussian objects: the phase and absorption perturbations will be slowly varying with no discontinuities or phase vortices [151]. The validity of the approximation as a function of probe detuning is investigated using simulations of atom clouds with typical experimental parameters (section 5.4).

It should be noted that while we have demonstrated the use of a Gaussian incident beam for experimental simplicity, the technique is more generally applicable. As long as the form of \( \psi_0(r, z) \) is known and the above assumptions are satisfied, an expression equivalent to Eq. 5.7 can be constructed and used to calculate a derivative for input to the TIE.

Retrieval of the exit wave phase and therefore the column density of the atom cloud (Eq. 5.4) is performed using standard techniques to solve the TIE [150]. The method involves division by the measured intensity distribution, which necessarily falls smoothly to zero at the boundaries of the image. To maintain
a stable solution in these areas well beyond the extent of the object, we divide by a modified form of the intensity:

\[
I(r_2, z_2) = \begin{cases} 
I(r_2, z_2) & I(r_2, z_2) \geq \epsilon \\
I(r_2, z_2) + \epsilon & I(r_2, z_2) < \epsilon
\end{cases}
\]

(5.9)

where \( \epsilon \ll 1 \) is a regularization parameter chosen to minimize quantitative errors while maintaining a stable solution [128,152]. Intensities were normalized to one and \( \epsilon = 10^{-3} \) for simulations and experiments. Smaller values of \( \epsilon \) introduced numerical errors in the outer regions of the image, while larger values decreased accuracy.

Propagation of the retrieved wave field back to the object plane is performed using the paraxial Fresnel diffraction formalism (Eq. 5.10):

\[
\Psi(r_2, z_2) = \frac{-i}{\lambda Z_{rad}} \exp \left( \frac{2\pi i Z_{rad}}{\lambda} \right) \exp \left( \frac{i\pi r_2^2}{\lambda Z_{rad}} \right) \mathcal{F} \left\{ \Psi(r_1, z_1) \exp \left( \frac{i\pi r_1^2}{\lambda Z_{rad}} \right) \right\}.
\]

(5.10)

\( Z_{rad} = ZR/(Z + R) \) is the rescaled propagation distance needed for phase-curved illumination according to the Fresnel scaling theorem [152], \( R \) is the radius of wavefront curvature at the object, and \( \mathcal{F} \) indicates a Fourier transform. Finally, the recovered wave field at the object plane is rescaled by the geometric transverse magnification factor \( M = (Z + R)/R \).

Having now retrieved the exit surface waves for both the perturbed and unperturbed beams, Eq. 5.4 provides the desired map of atomic column density, \( \rho(r) \), or if the column density is known, the refractive index can be extracted using Eqs. 5.2 and 5.3.

### 5.4 Simulations

To test the validity of the assumption made in Eq. 5.8, a range of simulations were performed using realistic experimental parameters (eg. Fig. 5.2).
5.4 Simulations

Figure 5.2: Simulation of diffraction from an atom cloud when illuminated by a near-resonant (detuned +1.5 natural line widths from resonance) curved Gaussian beam, and reconstruction of the column density. (a) input column density ($10^{13}$ atoms/cm$^2$), (b) simulated normalized diffraction intensity, (c) approximate intensity derivative (see Eq. 5.8), (d) reconstructed atomic column density ($10^{13}$ atoms/cm$^2$). (e) Horizontal line profiles of column density through the point of maximum density in (a), for the input, raw output and worst-case corrected output column densities, as outlined in section 5.4. Green lines indicate retrieved imaginary part of refractive index (solid) and simulation input (dashed) for inhomogeneous simulations (Sec. 5.4.3). The refractive index outside the cloud has been removed for clarity.

5.4.1 Homogeneous object simulations

The atomic column density was constructed from two overlapping atom clouds with spherically symmetric Gaussian density profiles, positioned to construct an asymmetric cloud, off-centre from the optical axis. Several different sizes and shapes of cloud were used. The refractive index of the object was calculated as a function of probe beam detuning, $\Delta$, using optical Bloch equations for a two-level $^{85}$Rb atom [119]. The unperturbed incident Gaussian beam was multiplied by the transmission function (Eq. 5.3) to yield the exact exit surface wave. Propagation to the detector was performed using Eq. 5.10. Equations 5.8 and 5.7 were then used to construct the approximate and exact derivatives for the perturbed and unperturbed beams. The phase of each wave was retrieved via solution of the TIE, and the waves were back-propagated to the object plane. Eq. 5.4 provided the retrieved atomic column density for comparison to the input.
As can be seen from Fig. 5.2(d), the reconstructed column density shows good qualitative agreement with the input. A line profile of the input and retrieved column densities is shown in Fig. 5.2e. Although the shape is clear, there is a discrepancy in the magnitude of the reconstructed column density, arising from two sources. Firstly, the approximation made in Eq. (5.8) assumes that the object introduces only a small, smoothly varying perturbation to the measured intensity of the unperturbed wave. For our Gaussian probe beam, the form of Eq. (5.7) can be expressed in terms of the second transverse derivative using standard Fourier techniques [153], i.e., \( H[I] \propto \nabla^2_w I \). Strong perturbations will introduce additional terms which are not accommodated by that approximation. Secondly, regularization (Eq. 5.9) results in a small quantitative change to the recovered phase, even though the shape remains well defined. Propagation back to the object plane magnifies the effects of these two sources of error.

### 5.4.2 Correction factor

The difference between the input and retrieved peak column densities provides a quantitative measurement of the magnitude of this discrepancy, for a given perturbation, as defined by the probe detuning and peak column density. Since the detuning is known and the raw output density is measured, this information can be inverted to calculate a correction factor, \( \eta \), to partially account for the errors introduced by the approximations outlined above, such that:

\[
\rho_{\text{corrected}} = \eta \rho_{\text{raw}}.
\]  

(5.11)

\( \rho_{\text{corrected}} \) and \( \rho_{\text{raw}} \) are the peak column densities of the corrected and raw outputs respectively, and \( \eta = \rho_{\text{input}} / \rho_{\text{raw}} \). The correction factor \( \eta \) is a constant (calculated from the peak values of the atomic densities), but is applied across the entire array. By performing multiple simulations across a range of detunings from \( 0 \Gamma \leq \Delta \leq 3 \Gamma \) and peak column densities between \( 10^{12} \leq \rho_{\text{input}} \leq 3 \times 10^{13} \) atoms/m\(^2\), \( \eta \) was calculated for the range of perturbations typically achieved in our experiments. The calculated correction factor corresponding to the measured detuning and raw column density was applied to the raw density output. Since the correction factor is calculated using the
peak column densities but applied to the entire array, some discrepancy between the input and corrected densities remains at the outer edges of the cloud. Simulations showed that this difference between input and corrected density was less than 10% for probe detunings greater than $\sim 1.5\Gamma$ and peak column densities within the range investigated here.

When experimental uncertainty of the probe beam detuning was considered, the simulations indicated that beyond $1.5\Gamma$ the sensitivity of the technique to such uncertainty is greatly reduced. For example, at $\Delta = 2.0\Gamma$, an experimental uncertainty of $\Gamma/2$ in the detuning of the probe beam results in an error in corrected peak density of $\sim 30\%$. By comparison, the same uncertainty would result in a 200% error in reconstructed density when using conventional on-resonance absorption imaging. The red curve in Fig. 5.2e shows the “worst-case” corrected column density profile corresponding to this scenario, where the value of $\eta$ was calculated using a $\Gamma/2$ error in probe beam detuning. When used to correct experimental data, the correction factor increases the quantitative accuracy of the technique while simultaneously indicating the optimum conditions for reducing sensitivity to experimental parameters.

### 5.4.3 Inhomogeneous object simulations

Simulations of imaging EIT in cold atom clouds were also performed. These simulations required a realistic model of the target atom cloud, with a spatially varying refractive index consistent with practical implementations of EIT schemes as discussed later (section 5.6.2). Optical Bloch equations were used to calculate the refractive index for a 3-level cascade EIT system between the $5S \rightarrow 5P \rightarrow 5D$ transitions of rubidium-85 [96,124].

An analytical expression for the steady-state off-diagonal density matrix element $\rho_{12}$ was obtained, containing over 100 terms. This exact result was used in simulations, but to provide some insight a first-order Taylor expansion was calculated for $\rho_{12}/\Omega_1$, which is proportional to the refractive index
of the probe beam:

\[
\frac{\rho_{12}}{\Omega_1} \approx \frac{2i\pi (\Gamma_2 + 2i (\Delta_1 + \Delta_2))}{4\pi^2 (\Gamma_1 + 2i\Delta_1) (\Gamma_2 + 2i (\Delta_1 + \Delta_2)) + \Omega_2^2}
\]  

(5.12)

The off-diagonal density matrix element is dependent on the detunings \((\Delta_1, \Delta_2)\) and Rabi frequencies \((\Omega_1, \Omega_2)\) of the probe and coupling lasers. The subscripts 1 and 2 indicate the probe \((5S \rightarrow 5P, 780 \text{ nm})\) and coupling \((5P \rightarrow 5D, 776 \text{ nm})\) laser respectively, and \(\Gamma_1\) and \(\Gamma_2\) are the natural line widths of the corresponding transitions.

To simulate imaging of a cloud with EIT-induced spatial structure, the pump Rabi frequency \(\Omega_2\) was spatially modulated across the image array. A vertically elongated Gaussian was used as the intensity of the pump laser, running through the atom cloud slightly off-centre, as in experiments discussed below. The intensity-dependent refractive index across the cloud was calculated using the exact result for the density matrix element (approximated in Eq. 5.12). The resulting array was then used to create the transmission function for the inhomogeneous cloud. Simulated diffraction data were created and used to solve the TIE, from which the object exit wave and unperturbed wave were recovered. The spatially dependent refractive index was extracted using Eq. 5.5. Other forms of single plane diffractive imaging are unable to separately extract the complex refractive index from a known column density and diffraction pattern.

The retrieved refractive index across the cloud was compared to the simulation input, as shown for the imaginary part of the refractive index by the green lines in Fig. 5.2(e). In regions where the column density was large, quantitative recovery of the spatially varying refractive index showed excellent agreement, to within 15% of the input. This is evident in the discrepancy between the solid and dashed green lines in Fig. 5.2(e). In regions of low atomic density, reconstructions used Tikhonov regularization, to avoid the singularity in Eq. 5.5 [128].
5.5 Experiment

Imaging experiments were performed on both homogeneous and inhomogeneous atom clouds, using a rubidium-85 magneto-optical trap (MOT) in $\sigma^{\pm}$ configuration [2, 102]. Cooling and trapping were performed on the $5S_{1/2} (F = 3) \rightarrow 5P_{3/2} (F' = 4)$ hyperfine transition at 780 nm with external cavity diode lasers (ECDLs) [154, 131, 132] and a semiconductor tapered amplifier [95]. Frequency stabilization was achieved using saturated absorption spectroscopy [96]. An additional repump laser resonant with the $F = 2 \rightarrow F' = 3$ hyperfine transition was co-propagated along one axis of the cooling beams. Atoms were provided using a dispenser source. The peak density of the cloud could be controlled using a combination of the detuning of the trapping beams, variation of the trap magnetic fields and the current through the dispenser source. An acousto-optic modulator (AOM) was used to switch the imaging beam on and off (for timed exposures) before the beam was guided to the MOT via single-mode polarization maintaining optical fiber.

At the vacuum chamber, the imaging beam was collimated to a $1/e^2$ diameter of 5 mm. An $f = 50 \text{ mm}$ achromatic doublet lens was used to focus the beam 70 mm behind the atom cloud (Fig. 5.3). The Fresnel number of the beam at the object plane was limited to a maximum of $N_F \sim 5$, due to restricted optical access to the vacuum chamber. A 200 mm lens allowed the effective defocus distance to be varied easily. Images were acquired with an interline transfer CCD.

5.5.1 Homogeneous imaging

For the homogeneous cloud, the imaging laser was detuned a frequency $\Delta$ from the $F = 3 \rightarrow F' = 4$ transition (natural linewidth $\Gamma = 6 \text{ MHz}$). The calculated refractive index of a two-level Rb-85 atom was used to extract a quantitative measurement of the atomic column density.

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1Nikon micro-Nikkor AF 200 mm f/4
2Apogee Alta U2000 ML 1600 x 1200 pixels
Figure 5.3: 2-D and 3-D schematics of the imaging and pump beam paths, and MOT vacuum chamber (not to scale). Waveplates have been omitted for clarity, as have coils in 3D. The pump beam was blocked for homogeneous imaging (section 5.5.1) and unblocked to create the inhomogeneous atom cloud (section 5.6.2). BS: Beam splitter; PD: Photodiode.

5.5.2 Inhomogeneous imaging

For imaging of an inhomogeneous atom cloud, an additional laser was used to modify the refractive index of the object. This “pump beam” was used to couple the upper transition in a cascade electromagnetically induced transparency scheme on the $5S_{1/2}(F = 3) \rightarrow 5P_{3/2}(F' = 3) \rightarrow 5D_{5/2}(F'' = 2, 3, 4)$ transitions [155,156]. Only a section of the cloud was illuminated with this beam, so that only part of the cloud exhibited EIT behavior. Accordingly, the refractive index of the illuminated section was modified by the presence of the pump beam, while the remainder of the cloud was unaffected, resulting
in a spatial modulation of refractive index across the extent of the cloud.

To create the EIT scheme, a second semiconductor tapered amplifier (maximum output power $\sim 300$ mW) seeded by a 776 nm ECDL provided the high power pump beam on the upper transition. The imaging beam also acted as the probe beam on the lower transition. To illuminate only a section of the MOT, the 776 nm beam was focused through a slit (width $\sim 500 \mu$m) using a cylindrical lens ($f = 100$ mm). The slit was re-imaged at the MOT using two $f = 300$ mm lenses, to create a “light sheet” (Fig. 5.3).

The pump beam was frequency locked to the two step $5S_{1/2}(F = 3) \rightarrow 5P_{3/2}(F' = 4) \rightarrow 5D_{5/2}(F'' = 5)$ transition [157] then shifted 130 MHz using an AOM, to be on resonance with the $5P_{3/2}(F' = 3) \rightarrow 5D_{5/2}(F'' = 2, 3, 4)$ transitions at the MOT.

For imaging, the probe laser was locked $+72$ MHz blue of the $5S_{1/2}(F = 3) \rightarrow 5P_{3/2}(F' = 3)$ transition and shifted back to resonance using an AOM, which also allowed switching of the imaging beam for timed exposures. Variation of the lock point allowed detuning of the imaging beam from resonance, while variation of the 776 nm AOM frequency allowed the relative frequency of the EIT resonance to be independently adjusted. The imaging beam was sampled after the MOT, and focused onto a photodiode to provide spectroscopic confirmation of the EIT resonance. During all EIT experiments, the MOT repump laser was tuned to the $F = 2 \rightarrow F' = 2$ hyperfine transition to avoid additional coupling between states which would complicate the atomic coherence.

## 5.6 Results

### 5.6.1 Homogeneous object

A typical experimental image and reconstruction of column density is shown in Fig. 5.4. The raw diffraction pattern exhibits considerable noise and evidence of etalon fringes due to various optical surfaces in the beam line. To reduce the noise, a two dimensional Gaussian fit was performed on the unperturbed intensity $I_0(r_2, z_2)$, recorded when the trap was empty. The noise,
$N(\mathbf{r}_2)$, was defined as the difference between the fitted Gaussian, $G_0(\mathbf{r}_2)$ and the unperturbed intensity: $N(\mathbf{r}_2) = I_0(\mathbf{r}_2, z_2) - G_0(\mathbf{r}_2)$. Before solving the TIE for the phase of the waves, the noise was subtracted from the experimental images, resulting in a significant reduction of noise in the processed diffraction pattern:

$$I'_0(\mathbf{r}_2, z_2) = I_0(\mathbf{r}_2, z_2) - N(\mathbf{r}_2),$$

$$I'(\mathbf{r}_2, z_2) = I(\mathbf{r}_2, z_2) - N(\mathbf{r}_2).$$

**Figure 5.4:** Experimental results for Single-plane Curved-beam Phase Imaging. (a) Raw experimental diffraction pattern; (b) after noise subtraction; (c) reconstructed column density cropped to region inside intensity threshold, retrieved from diffraction pattern in (b), and (d) plot of column density through dashed line in (c). The blue line in (d) indicates the column density retrieved from a conventional in-focus, on-resonance absorption image taken on the same day. The diffraction pattern was recorded at a defocus distance of $Z = 20\,\text{mm}$, a probe beam detuning of $\Delta = 1.5\Gamma$ and imaging probe pulse duration of $13\,\mu\text{s}$. A correction factor of $\eta = 1.3$ was used to obtain the density map (see Sec. 5.4). The circular discontinuity in the column density arises from the threshold function (Eq. 5.9).

Images demonstrated a good qualitative signal-to-noise ratio (SNR) using probe detunings between 0 and $3\Gamma$ and defocus distances out to $Z = 40\,\text{mm}$. At large defocus distances and probe detunings, a high peak column density
was required to produce a good SNR, however the recovered images consistently demonstrated good agreement with alternative quantitative imaging techniques, such as absorption [141] and diffraction contrast imaging [143]. For direct comparison of SCPI with conventional techniques, the quantitative density profile retrieved from an on-resonance absorption image taken on the same day is also shown in Fig. 5.4(d), showing excellent agreement with the SCPI result. The range of defocus distances investigated was only limited by optical access to the vacuum system and the size of the CCD. Further iterative refinement of the images using coherent diffractive imaging algorithms (e.g. Gerchberg-Saxton and error-reduction algorithms [17]) has the potential to provide enhancement of fine detail and structure within the cloud.

### 5.6.2 Inhomogeneous Imaging

In Fig. 5.4b, etalon fringes reduce the signal-to-noise ratio (SNR) in the diffraction pattern, even after noise subtraction, due to shot-to-shot variation in probe laser frequency. To further reduce artefacts at higher spacial frequencies, including etalon fringes, the diffraction data for EIT imaging was spatially filtered by convolution with a Gaussian of variable full width at half maximum (FWHM), at the cost of slightly decreased spatial resolution.

Figure 5.5 shows experimentally acquired diffraction data and retrieved atomic refractive index of the atom cloud with spatial modulation due to the EIT pump beam, calculated using Eq. 5.5. The column density in Eq. 5.5 was measured with the pump beam blocked, and retrieved numerically using Eq. 5.4. The refractive index maps and line profiles have been smoothed using a five-pixel box-car average to reduce the effects of noise. Although this reduces the resolution of the result, our interest is in the variation of refractive index on the much larger scales of the atom cloud and laser beam profile.

The EIT pump beam is seen clearly as an area of low absorption and phase shift in a near-vertical region of the atom cloud. The extracted imaginary part of the refractive index shows good agreement with the theory. In regions of high atomic density, the discrepancy between measured and theoretical imaginary refractive index is less than 15%. In the outer regions of the
Figure 5.5: Imaging of an inhomogeneous atom cloud and extraction of the complex refractive index. Orange arrows indicate the propagation direction of the EIT pump laser beam. (a) Inhomogeneous diffraction data. The intensity of the unperturbed Gaussian has been subtracted for clarity. (b) Diffraction data from a homogeneous (pump beam off) cloud, unperturbed beam subtracted. (c) Column density reconstructed from (b). (d) Retrieved absorption coefficient, $\beta$, calculated using Eq. 5.5. (e) Retrieved phase coefficient, $\alpha$. (f) Experimental line profile (black) through dashed line in (d), with theoretical absorption curve shown in red. (g) Phase line profile, with theoretical curve, as in (f). Images were taken at $Z = 20$ mm defocus distance, $\Delta = 2\Gamma$ from the $F = 3 \rightarrow F' = 3$ transition. Theoretical lines were calculated using the exact version of Eq. 5.12, with parameters ($\Omega_1$, $\Omega_2$, $\Delta_1$, $\Delta_2$ and slit width) fitted to the data, within their respective experimental error margins. The data was convolved with a Gaussian of FWHM 186 $\mu$m prior to reconstruction.

cloud, where the density fell below $10^{11}$ atoms/m$^2$, Tikhonov regularization increases the maximum discrepancy to 40%. The mismatch between the retrieved and theoretical real part of the refractive index on the right-hand
side of Fig. 5.5(g) is due to the hard edges of the EIT pump beam, caused by the slit through which it passed. Although the edges provide a more distinct and visible change in refractive index, they result in areas of rapidly varying phase which are not well accommodated by the approximations of this technique. For the Gaussian beams typically used in atomic coherence measurements, this artifact would be significantly reduced, if present at all.

Using the edges of the pump beam in Fig. 5.5d as a reference, we estimate the resolution of the refractive index retrieval as \(\sim 40 \mu m\). However, the diffraction data used to reconstruct that image was spatially filtered, and the reconstruction subsequently averaged over a five pixel radius, resulting in a substantial reduction in final resolution. In general, the technique does not impose any additional constraints on resolution, which is ultimately limited only by the probe wavelength and detector size. Furthermore, although Eq. 5.7 has been shown to work well for Gaussian objects, it can be adapted to suit more complex structures, relaxing the requirement for a smoothly varying object [144].

## 5.7 Conclusion

Single-plane Curved beam Phase Imaging (SCPI) was demonstrated, providing quantitative images of a cold atom cloud over a range of probe laser detunings and defocus distances. Images showed a good signal-to-noise ratio and quantitative agreement with conventional methods, as seen in Fig. 5.4(d). Unlike other non-iterative diffractive methods, SCPI can retrieve a quantitative image of the column density of an inhomogeneous object, given knowledge of the spatial variation in refractive index. By extracting the complex exit surface wave, the complex refractive index can be separated from the atomic column density. Either the refractive index or the column density can be recovered, provided that the other is known. Combined with the reduced sensitivity to experimental parameters outlined in section 5.4, SCPI can be a useful tool for a wide range of cold atom applications, including ultracold plasma diagnostics and studies of atomic coherence phenomena in ultracold gases. Additionally, the method could provide an excellent starting point for further iterative refinement of the images [17].
Simulation of imaging of cold atoms using typical experimental parameters yielded quantitative reconstructions of column density to within 10%. Using the results from the simulations, a detuning-dependent correction factor was calculated, which partially compensated for the errors introduced by the approximations used in the SCPI technique. Simulations also indicated an optimum region of the available experimental parameter space for imaging. The technique was implemented experimentally and the resulting images were of high quality compared to in-focus, on-resonance absorption imaging of homogeneous objects.

Although conventional absorption imaging remains one of the simplest methods of quantitative imaging, its sensitivity to the frequency of the probe laser beam and any defocus result in large uncertainties when used as a quantitative technique. Using SCPI, this sensitivity is significantly reduced by deliberately detuning the probe and measuring a diffraction pattern. Although other imaging techniques have demonstrated such reduced sensitivity [119], they are unable to separate the refractive index from the column density, limiting their use to homogeneous objects. SCPI retains the advantages of these techniques, while enabling imaging of atomic coherence phenomena in inhomogeneous cold atom clouds. Experimental reconstructions of an atom cloud with spatially modulated refractive index created using a cascade EIT scheme showed good agreement with theoretical predictions. Agreement to within 15% of theoretical predictions was achieved in areas of high atomic density where Tikhonov regularization did not affect the results. The resolution of spatially filtered and smoothed experimental images of inhomogeneous clouds was estimated as $\sim 40 \mu m$, although the resolution of the technique is ultimately limited by wavelength and detector size.

This technique is particularly suited to imaging of atomic coherence effects in cold atom clouds, due to their typically Gaussian density profiles. Many experiments in atomic physics also utilize Gaussian laser beams. Accordingly, any induced inhomogeneities in the refractive index of the cloud will be smoothly varying, minimizing artefacts in quantitative reconstructions, such as those induced by the hard edges of the EIT pump beam in Fig. 5.5(g). Accordingly, SCPI offers the potential to yield greater information from atomic coherence experiments in cold atoms than commonly used probe
beam techniques, while maintaining experimental simplicity.

We have shown an adaptation of the reconstruction process and experimental implementation of SCPI for cold atom imaging. The technique was used to image a complex object involving atomic coherence effects. Further work will involve application to electron bunch shaping in an ultracold plasma, and possible iterative refinement to increase fine detail. The simplicity of the technique makes it a useful tool not only for cold atom applications, but also for x-ray and phase imaging applications in general.
5.8 Additional material

The following supporting material was not included in the published work due to length restrictions.

5.8.1 Correction factor

The results of the simulations described in Sec. 5.4.2 are shown in Fig. 5.6. The plot shows the correction factor, $\eta$, as a function of input column density and probe laser detuning and supports the conclusion that SCPI exhibits reduced sensitivity to systematic experimental uncertainties when compared to conventional methods, as explained in Sec. 5.4.2.

![Figure 5.6: Correction factor, $\eta$, as a function of probe laser detuning and peak input atomic column density. At detunings larger than $\Delta = 1.5\Gamma$, the error remains almost constant over the range of input column densities. Within this region, the technique remains relatively insensitive to uncertainties in probe detuning or raw density.](image-url)
5.8 Additional material

5.8.2 Spectroscopic EIT measurement

To determine the magnitude of the refractive index change caused by the pump beam, and to more accurately control the relative frequency of the EIT resonance, spectroscopic measurements of EIT in the atom cloud were obtained. Part of the imaging beam was split off, after the MOT, and focused onto a photodiode. The probe laser frequency was scanned through the relevant hyperfine transitions and spectra recorded such as that shown in Fig. 5.7. Two EIT peaks can be identified, corresponding to the $5D_{5/2}(F'' = 4, 3)$ hyperfine states separated by 9.4 MHz [158]. A third peak was also observed when the frequency of the 776 nm beam was increased, corresponding to the $F'' = 2$ hyperfine state. During all EIT experiments, the MOT repump laser was tuned to the $F = 2 \rightarrow F' = 2$ hyperfine transition to avoid additional coupling between states which would complicate the atomic coherence.

5.8.3 Pump beam locking

Figure 5.8 shows a schematic of the locking scheme for the 776 nm EIT pump laser beam.

5.8.4 Transport of Intensity Equation

The Transport of Intensity Equation (TIE, Eq. 5.6) is used to retrieve the phase of the wave at the detector plane, which is required for back-propagation to the sample plane and extraction of the atomic column density. To solve the TIE, we follow the method of Paganin and Nugent [150, 152]. From Eq. 5.6, the expression for the phase of the wave at the detector plane
Figure 5.7: Transmission of the 780 nm imaging probe laser through the MOT, scanning the laser frequency through the $F = 3 \rightarrow F' = 2, 3$ transitions, with and without the 776 nm pump laser. Two EIT peaks are seen in the top trace, corresponding to the $5D_{5/2}(F'' = 4, 3)$ hyperfine states. The vertical line indicates the frequency lock point used in Fig. 5.5, at $\Delta = 2\Gamma$, where the absorption is dramatically reduced when the cloud is illuminated with the pump beam.

Figure 5.8: Locking scheme for the 776 nm EIT pump laser. Waveplates, lenses and mirrors omitted for clarity. The frequencies given are relative to the $F = 3 \rightarrow F' = 4$ (780 nm) and $F' = 4 \rightarrow F'' = 5$ (776 nm) transition, such that the 776 nm beam is resonant with the $F' = 3 \rightarrow F' = 2, 3, 4$ transitions at the MOT.
\[ \phi(x, y, z) = -k \nabla_\perp^{-2} \left( \nabla_\perp \cdot \left\{ \frac{1}{I(x, y, z)} \nabla_\perp \left[ \nabla_\perp -2 I(x, y, z) \frac{\partial I(x, y, z)}{\partial z} \right] \right\} \right). \] (5.15)

Application of the Fourier derivative theorem allows the gradient and inverse Laplace operators to be represented in terms of Fourier transforms, so that

\[ \nabla_\perp = i \mathcal{F}^{-1}(k_x, k_y) \mathcal{F} \]  
\[ \nabla_\perp^{-2} = -\mathcal{F}^{-1} \frac{1}{k_x^2 + k_y^2} \mathcal{F} \]  

where \( \mathcal{F} \) indicates a Fourier transform. Given the required input intensity and longitudinal intensity derivative, Eq. 5.15 can now be calculated using computationally efficient fast Fourier transforms. The singularity at \( k_x = k_y = 0 \) is avoided by replacing \( 1/(k_x^2 + k_y^2) \) by zero at the origin [152]^4.

^3Pg. 299
^4Pg. 300
Chapter 6

Conclusion

The research described in this thesis has demonstrated the techniques required to produce shaped cold electron bunches from a new cold atom source. Through shaping of the electron bunch, a cold electron source has the potential to overcome the brightness and coherence limitations of conventional thermal sources, enabling their use in ultrafast diffractive imaging. Targets of particular interest include biological samples such as membrane proteins, and defects in solid-state devices, for example studying dynamic propagation of cracks at an atomic scale. The structure of membrane proteins is exceedingly difficult to determine using conventional techniques due to the difficulties associated with crystallisation of the sample, which is required to enhance the scattered x-ray signal. Electrons interact far more strongly with the sample than x-rays, potentially removing the requirement of signal enhancement via crystallisation. UED can also provide dynamic information on picosecond timescales, to investigate biological processes with temporal resolution [159,160]. For example, observation of the dynamics of protein folding may aid development of synthetic proteins used in rational drug design and disease prevention [161].

Ultracold plasma is a dynamic field undergoing rapid development since the
creation of the first UCP by Killian et al. in 1999 [1]. Chapter 2 presented a review of our current understanding of the physics of UCP with a focus on cold electron production for diffractive imaging. Following a review of plasma fundamentals, particular attention was given to intrinsic heating mechanisms which limit the ultimate electron temperature. Threshold lowering [59] and disorder induced heating [3] were observed to contribute significantly to this limit, resulting in expected electron temperatures of no less than 10 K. The effects of disorder induced heating may be mitigated by inducing order prior to ionisation, for example by using an optical or ponderomotive lattice [162] to arrange the atoms in a regular structure [60, 61]. The threshold lowering effect (TLE) was seen to be a density-dependent heating mechanism, minimised at low density. As low densities are undesirable for bright electron bunches, the dynamics of Rydberg field-ionisation may become an important field of interest during the future development of the project, as this may provide a means of minimising TLE without reducing density.

Several UCP diagnostic techniques were investigated, offering indirect means of extracting density and temperature information from the plasma. Indirect probes of electron temperature will be of particular importance in optimising the source, since (unlike strontium or metastable argon [67, 163]) singly ionised rubidium has no electronic transition in the visible or infrared wavelength range. Conventional direct temperature measurements via imaging or spectroscopy of the ions are therefore not possible in a rubidium UCP. The diagnostic techniques presented to measure plasma temperature and density, although not yet demonstrated on the Melbourne UCP, will undoubtedly form the basis for future measurements and optimisation of the source. Two methods of indirect temperature measurement which can be relatively eas-
ily applied to our system have been previously demonstrated [6, 63]. The accepted scaling at low temperatures of the three body recombination rate of $T^{-9/2}$ can be used to extract temperature information via measurement of the refilling rate of highly excited Rydberg states following field ionisation [63]. Alternatively, observations of microwave-frequency Tonks-Dattner resonances can be fitted to theoretical predictions using temperature as a free parameter [6]. Since both of these techniques involve fitting to experimental parameters, the most reliable temperature measurements will utilise both techniques as a means of verification.

In Ch. 3, the design and construction of the Melbourne UCP apparatus were described, along with initial characterisations of the system. Design considerations were based around versatility and flexibility, with an emphasis on achieving excellent ultrahigh vacuum conditions as quickly as possible. To that end, the system was baked out and pumped down to $10^{-10}$ mbar or lower (below the reliable operating range of our pressure readings) in around four weeks. Excellent vacuum conditions remain, after 12 months of system operation. Although optical access is presently sufficient for our current experiments, additional access may be desired in the future, for which a spherical main chamber may be more practical. Planned upgrades to the system also include an additional electron diffraction chamber for sample interchange and replacement of non-AR coated vacuum view ports with the correct parts to reduce unwanted scattered light inside the chamber.

Chapter 3 also presented the extensive electronic, optical, software, laser and timing systems that were developed for the UCP apparatus. Fast switching of magnetic fields was of particular importance during initial charged particle extraction. Sec. 3.3 described the electronics used to achieve millisecond
switching of both MOT and Zeeman slower magnetic fields which was later shown to reduce electron bunch distortion considerably. Use of an optical dipole trap may relax switching speed requirements in future experiments.

Diagnostic measurements of the trapped atom cloud determined the temperature to be $69 \pm 11 \mu K$, peak atomic column density $3.4 \times 10^{13} m^{-2}$ and maximum cloud size $3.0 \pm 0.4 mm$ full width at half maximum, in good agreement with design expectations. The first electrons and ions were extracted from the Melbourne UCP system in November 2009, two and half years after design commenced and 18 months after the laboratory space was commissioned. Temporal electron and ion signals obtained from the MCP showed good agreement with classical time-of-flight predictions.

Two-step ionisation using a spatial light modulator to shape the excitation beam followed by uniform ionisation of the excited state distribution resulted in the first demonstration of arbitrarily shaped electron bunches. Unlike thermal electron sources, in which the shape of the bunch diffuses due to the large initial (random) electron velocity spread, cold electrons were seen to maintain the shape of their initial distribution.

The quality of the bunch shape was shown to depend heavily on the initial atomic density distribution. To determine the atomic distribution, two new imaging techniques were developed. Excited state diffraction contrast imaging (DCI, Ch. 4) provided a method of determining the excited state fraction and distribution via propagation-based phase imaging. The technique exhibits several advantages over conventional imaging, in particular with regards to its insensitivity to experimental parameters. In addition, the off-resonant technique is less destructive to the atom cloud. Images with high
signal-to-noise ratios proved difficult to obtain in low-density atom clouds. Under these conditions, the technique is best utilised as an indication of excitation efficiency. Future experiments anticipate use of an optical dipole trap. The increased density achieved in such traps will result in both better images and brighter bunches. Imaging of such high density clouds along the excitation axis will allow direct feed-forward of ground-state density and feedback of excited-state density information to the excitation laser intensity, to further improve the bunch shape.

The second imaging technique uses a perturbative approximation to estimate from a single image the longitudinal intensity derivative. Using this information, the transport of intensity equation is solved for the phase at the detector plane and the wave is then back-propagated to find the object exit surface wave. The method can provide a map of either the atomic density or complex refractive index, provided the other is known. It is most useful as a complementary technique to DCI, for use in situations involving multiple light fields which cause inhomogeneities in the atomic refractive index across the spatial extent of the cloud. It was demonstrated to retrieve the spatial variation in refractive index in a cold atom cloud prepared using spatially-varying electromagnetically induced transparency. The technique is particularly suited to optical investigations of such atomic coherence systems, although it may be easily generalised to x-ray or electron regimes. It is experimentally simple, and the phase retrieval process robust. More extensive use of the technique is anticipated in future experiments when high intensity lasers (such as those used in optical dipole trapping, or femtosecond excitation pulses) cause more extreme variations in refractive index across the cloud than are currently observed.
The atomic density distribution in the Melbourne UCP was measured using the imaging techniques developed, and shown to influence the electron bunch shape considerably. Clearly, the imaging of and correction for variation in atomic density will be valuable for producing bright, coherent electron bunches. The excitation laser intensity was modulated to compensate for the measured Gaussian atomic density distribution and resulted in production of 2D uniform electron bunches. The result supports the feasibility of 3D shaped bunch production, which is anticipated in future work. The effect of higher initial electron temperature, adjusted via the wavelength of the ionisation laser pulse, was investigated and seen to degrade the bunch shape as expected. Accordingly, UCP remains an advantageous source of electrons for shaped bunch production and diffraactive imaging applications.

For static acceleration fields, optimum bunch shaping results were achieved via field-ionisation of Rydberg atoms, which produced the coldest electrons. The measured ionisation threshold shift was observed to agree well with the theoretical value of $8.3 \pm 0.7 \text{meV}$.

In Sec. 3.5, a quantitative analysis of the bunch edge sharpness provided an upper limit to the electron temperature of $T < 35K$. Using this measurement, the transverse brightness of the source was estimated as $10^9 \text{A m}^{-2} \text{sr}$, three orders of magnitude brighter than previous UCP work [13]. Although impressive, the brightness achieved to date remains well below the estimated potential of the source. Future implementation of experimental upgrades is estimated to result in a further increase in brightness of another three orders of magnitude, primarily due to increased atomic density and decreased temporal bunch length. When compared to alternative electron sources, UCP remains unique in its capability to produce high brightness shaped bunches.
which can overcome emittance growth due to space charge forces. The estimated potential brightness, emittance and coherence of the source were seen to meet or exceed the requirements for ultrafast electron diffraction of biomolecules [11].

This thesis has presented the design, construction and initial results of a new experiment to produce UCP. Even at this early stage of the experiment lifetime, initial results have, in general, exceeded design expectations. Advances in atomic imaging demonstrated here have enabled new avenues of research into electron bunch shaping. Coupled with state-of-the-art charged particle accelerators and electron optics, shaped electron bunches from UCP have been predicted to achieve the coherence and brightness required for single-shot ultrafast diffractive imaging of bio-molecules [11]. The results of two dimensional shaping demonstrated in this thesis have indicated that not only is 3D bunch shaping a realistic objective for the future, but cold electrons prove to be the critical requirement for effective bunch shaping. The new UCP experiment presents a plethora of opportunities for further research, and rapid advancement of the field is anticipated.
Appendix A

Refractive Index, Susceptibility, and Density Matrices

The complex refractive index, $n$, of a material is defined as\(^1\) [122]:

$$n = n' + in'' = \frac{kc}{\omega}$$  \hspace{0.5cm} (A.1)

where $k$ is the wavenumber, $c$ is the speed of light in vacuum, and $\omega$ is the frequency of the applied field. It is in general complex, and is related to the complex electric susceptibility, $\chi_e$, by\(^2\) $n^2 = 1 + \chi_e$. Denoting the susceptibility as $\chi_e = \chi' + i\chi''$, then

$$n = \sqrt{1 + \chi' + i\chi''}. \hspace{0.5cm} (A.2)$$

For small $\chi_e$, a binomial approximation is valid, so that the real and imaginary parts of the refractive index, which correspond to the phase and ab-

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\(^1\)Page 23, equation 2.4.  
\(^2\)Page 23, equation 2.2.
The susceptibility (and hence the refractive index) relates the induced polarisation, $P$ of the atomic gas due to the applied field, $E$:

$$P = \chi_e E.$$  \hspace{1cm} (A.7)

The polarisation of the atomic gas can be determined using a density matrix formalism, where $P$ is given by the off-diagonal elements of the density matrix for the system, $\rho$ [121]. These elements are calculated from the Liouville equation:

$$\dot{\rho} = -\frac{i}{\hbar} [H, \rho] + L$$  \hspace{1cm} (A.8)

where $H$ is the Hamiltonian for the system and $L$ describes the spontaneous decay. For an atom with $N$ levels, this reduces to $N$ coupled differential equations: the optical Bloch equations (OBEs). Thus for small $\chi_e$, the off-diagonal elements of the density matrix are proportional to the refractive index of the material.
To determine the refractive index from the density matrix elements, we denote the real and imaginary parts of the refractive index by $\delta$ and $\beta$ \cite{119,165}:

$$n(r) = 1 + N(r)(\delta + i\beta)$$  \hspace{1cm} (A.9)

$$= 1 + \rho(x)\frac{\sigma_0 \lambda \Gamma}{4\pi \Omega} \rho_{ge}.$$  \hspace{1cm} (A.10)

$\Gamma$ is the natural linewidth, $\Omega$ is the Rabi frequency of the driving laser, $n(r)$ is the spatially varying refractive index of the atom cloud, $N(r)$ is the atom number density, $\rho(x)$ is the atomic column density, $\sigma_0$ is the atomic cross section and $\rho_{ge}$ is the complex off-diagonal density matrix element.

For a two-level atom, the atomic cross section is \cite{102}:

$$\sigma_0 = \frac{3\lambda^2}{2\pi}.$$  \hspace{1cm} (A.11)

This is a gross simplification which assumes a maximum interaction. For a two level atom then, we have:

$$n(r) = +\rho(x)\frac{\sigma_0 \lambda \Gamma}{4\pi \Omega} \rho_{ge}.$$  \hspace{1cm} (A.12)

$$= \rho(x)\frac{\sigma_0 \lambda \Gamma i\Omega + 2\Delta \Omega}{4\pi \Omega \Gamma^2 + 4\Delta^2}.$$  \hspace{1cm} (A.13)

$$= \rho(x)\frac{\sigma_0 \lambda \Gamma i + 2\Delta}{4\pi \Gamma^2 + 4\Delta^2}.$$  \hspace{1cm} (A.14)

$$= \rho(x)\frac{\sigma_0 \lambda i + \bar{\Delta}}{4\pi \frac{1}{1 + \tilde{\Delta}}}.$$  \hspace{1cm} (A.15)

where $\bar{\Delta} = 2\Delta/\Gamma$ (i.e. the detuning in units of half linewidths).

For excited state imaging, the separation of the hyperfine levels is much smaller, so we must use the cross section specific to the transition of interest.

\(^3\)Milonni, pg 219, eqn 7.4.3
\(^4\)Metcalf, pg 27
[129], that is,

$$\sigma_0 = \frac{(2J' + 1) \lambda^2}{(2J + 1) 2\pi}$$  \hspace{1cm} (A.16)

where $J$ and $J'$ are the total electron angular momentum quantum numbers for the ground and excited states respectively. For 776 nm imaging ($5P \rightarrow 5D$) we have:

$$\sigma_0 = \frac{3\lambda^2}{4\pi}$$  \hspace{1cm} (A.17)

So that:

$$n(r) = 1 + \rho(x) \frac{3\lambda^3}{16\pi^2 \omega^2} \rho_{ge}.$$  \hspace{1cm} (A.18)
References


REFERENCES


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