Production and Characterisation of Nanoscale Structures using Atom Lithographic Techniques

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This work has not previously been submitted for a degree or diploma in any university. To the best of my knowledge and belief, the thesis contains no material previously published or written by another person except where due reference is made in the thesis itself.

Joshua P. Beardmore
Acknowledgements

During my PhD candidature I have been privileged enough to see some truly unique places and meet amazing people. The opportunities afforded to me have changed my life in many more ways than I could have thought.

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A metastable neon (Ne$^*$) beam generated by a liquid nitrogen cooled, DC discharge source was purified to an atomic beam consisting of a single metastable state. The atomic beam was cooled in the transverse direction by a two dimensional optical collimator, slowed in the longitudinal direction by a novel dual beam Zeeman slower, and then guided through a 30° arc by a hexapole magnetic guide. This resulted in a pure, UV free metastable atomic beam with a flux of $(4.4\pm1.1)\times10^9$ atoms s$^{-1}$.

The metastable neon atomic beam was used to investigate the patterns formed in resist based atom lithography experiments utilising alkanethiol self-assembled monolayer resists. It was observed that very short chain alkanethiols, such as ethanethiol, do not form viable resist layers. They are likely desorbed from the surface during exposure to the metastable beam and replaced by background mechanical pump oil molecules. Above the critical dosage ($\sim7\times10^{14}$ atoms cm$^{-2}$) these samples react in a manner similar to bare gold samples and form a carbonaceous resist layer. This dosage was found to be significantly affected by the vacuum infrastructure, highlighting the role contamination plays in the formation of negative contrast patterns in resist based atom lithography.

Using ellipsometry the growth of a carbonaceous film during exposure to a metastable atomic beam was characterised. The desorption cross-section of carbonaceous material from a silicon surface via Ne$^*$ impact was determined to be many times larger than the polymerisation cross-section. The values determined, along with simple estimates for the mean residence time,
volume, and cross-section of the contaminants involved provide insight for the application of the theory to other metastable atom experimental apparatus.

Direct deposition lithography without laser cooling of the atomic beam was achieved and patterning observed for iron atoms with a local average transverse velocity of up to 4 m s$^{-1}$. A broadening of the experimentally deposited samples, from a full width half maximum of 35 nm predicted by simulations to >80 nm on SiOx substrates, was observed. The broadening is attributed to a substrate dependent diffusion mechanism and the scattering and interference of the standing wave light mask near the substrate.

An initial characterisation of the magnetic properties of co-deposited iron-nickel (Fe-Ni) structures has been conducted using the longitudinal magneto-optic Kerr effect. Variations in the Fe-Ni concentrations influence the coercivity of the deposited structures. A reduction in the coercive field in regions with line structures was observed when applying a magnetic field parallel to the co-deposited lines. This has been attributed to the nucleation of magnetic domains in regions were the Fe-Ni alloy possesses a lower magnetic moment per atom. A magnetic anisotropy induced by the incident angle of the Ni atomic beam was also observed in regions without nanostructuring.
List of Publications

Below is a list of peer reviewed publications with the contributions made towards each. The relevant sections in this thesis are referenced in each case. Copies of these publications are provided in Appendix A at the end of this thesis.


- The design and implementation of the dual laser beam input to the Zeeman-slower (Section 2.5.3).
- Designed, constructed, and characterised the hexapole magnetic guide (Section 2.5.4).
- Optimisation of the experimental apparatus and taking of results.
- Three-dimensional atom trajectory simulation through the magnetic guide (Section 2.5.4).


- Aiding the setup and optimisation of the laser system including the installation of a single mode, polarisation maintaining optic fibre.
• Implementation of the standing wave light mask (Section 4.2.2).
• Generating samples and assisting in their analysis using atomic force microscopy and magneto-optical Kerr effect microscopy (Section 4.4).


• Review of the theory of thin film growth (Section 3.2.2).
• Assembly and operation of the experimental apparatus.
• Writing the data acquisition and analysis programs.
• Analysis of the data (Section 3.4.5).


• Design of the sample carousel (Section 3.4.2).
• Installation of the residual gas analyser.
• Set up of the contact angle goniometer (Section 3.3.1).
• Assisting the general operation of the experiment and data analysis.
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Chapter 1

Introduction

The interaction between light and matter has interested scientists for centuries. One of the earliest observations on the ability of light to exert a force on a physical system was made by Kepler [1]. He observed that the tail of a comet always points away from the sun and concluded that it must be created by the sun’s rays. In the centuries following Kepler’s observation the nature of light was investigated extensively with fundamental work performed by Descartes, Hooke, Huygens, Newton, Young, and Fresnel [2]. In the 1860’s Maxwell developed a series of equations that showed that light consisted of oscillating electric and magnetic fields and would go on to show how it could apply pressure to a material [3]. This concept was also investigated by Bartoli who discussed its implications to the second law of thermodynamics [4]. Radiation pressure was not realised experimentally until 1900 when Lebedev measured the deflection of metal discs in vacuum [5]. The results of this experiment were confirmed by Nichols and Hull the following year when they measured the radiation pressure exerted on a silvered glass plate [6].

One of the problems still facing the scientific community of the time was that Maxwell’s theory of light did not explain all the phenomena observed. The most notable of which was that the energy imparted by light on particles is dependent on its frequency rather than its intensity. In 1900 Planck hypothesised that the energy involved in the absorption or emission of electromagnetic radiation is quantised into multiples of $E = h\nu$, where $h$ is Planck’s constant and $\nu$ is the frequency of the radiation [7]. Einstein,
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After his explanation of the photoelectric effect, would use Plank’s idea and show that the energy quanta would also carry momentum \( p = \frac{h}{\lambda} \), where \( \lambda \) is the wavelength of light \[8\]. The momentum of a photon was verified experimentally in 1923 by Compton, who observed the scattering of X-rays from atoms \[9\]. This description of the photon provides a more intuitive method of realising the effect of photons on matter. When a photon is absorbed, the momentum is imparted to the material in order to conserve momentum. If the photon is reflected from a surface, the material acquires twice the momentum due to recoil. Thus, with a sufficiently intense source of photons matter is able to be manipulated in a significant way.

1.1 Neutral atom manipulation/cooling

Following the development of the laser \[10\], manipulation of the motion of atoms underwent significant advances \[11–15\]. By using the momentum imparted to the atoms via the absorption of photons, their motion can be damped, or cooled. Laser cooling has enabled the reduction of the broad velocity distributions, characteristic of thermal atomic beams. This led to the spatial confinement of an atomic beam in three dimensions with the development of the magneto-optical trap \[16\]. Laser cooling was also the first step towards a new state of matter with the experimental realisation of Bose-Einstein condensation (BEC) \[17, 18\]. The enhancement of atomic beams has allowed much weaker forces to be utilised in atom manipulation. For example, the optical dipole force and the magnetic force arising from inhomogeneous magnetic fields. This has given rise to the field of atom optics \[19, 20\].

Atom optics uses these forces to alter the trajectories of atoms in ways analogous to photons in conventional optics. As with conventional optics, atom optical elements may be divided into a number of classes \[21\]. Zeroth-order elements, such as lenses and mirrors, only manipulate the atom as a particle and do not make use of the wave nature of the atom. Higher-order elements utilise the quantum mechanical properties to coherently divide the atomic wave-packet into two or more states, such as in beam splitters and interferometers. A number of atom optical elements have been experimentally achieved including a variety of lenses \[22–28\], mirrors \[29–31\], beamsplitters \[32\], and diffraction gratings \[28, 33–36\]. These elements may be used in
conjunction with laser cooling to increase the brightness and phase-space density of an atomic beam. Of particular importance to many atom optical experiments is the compression of the transverse and longitudinal velocity components of the atomic beam.

The transverse velocity compression, or collimation, of the atomic beam is readily achieved using laser cooling in two-dimensions. An arrangement of particular interest is the multiple reflection, two-dimensional optical collimator [37] which is covered in more detail in Section 2.5.2. Compression of the spatial distribution, i.e. increasing the phase-space density, of the atomic beam has also been accomplished [37–39]. One arrangement utilises magneto-optical forces and forms a two-dimensional magneto-optic trap where the atoms are simultaneously compressed and cooled in the radial direction [38, 39]. An alternative method involves use of an atom optical lens and subsequent re-collimation stage [37]. For these methods to be efficient the longitudinal velocity must first be reduced and compressed. Reducing the longitudinal velocity greatly increases the interaction time of the atoms with the compression and collimation elements, while narrowing of the velocity distribution will remove any effects analogous to chromatic aberrations in conventional optics. Methods for the longitudinal slowing of atomic beams utilising counter-propagating light must account for the Doppler shifts induced as the atoms are slowed. One technique, Zeeman slowing, compensates by using a tapered magnetic field [14], while another ‘chirps’ the laser frequency [40].

### 1.2 Atom lithography

Enhanced atomic beams have found practical application in the creation of nanostructures, a technique termed atom lithography. This process involves the manipulation of atomic beams to create nanoscale structures on surfaces [41]. Atom lithography is analogous to optical lithography where the conventional roles of light and matter are reversed. In atom lithography an atomic beam incident on a sample may be spatially modulated by using either a physical transmission mask, the optical dipole force of a near-resonant laser standing wave [28], or manipulation of the atomic wavefront by holographic means [42]. For light based masks, the quantum mechanical nature of the interaction between the neutral atoms and the light field has enabled the
There are two distinct branches of atom lithography; direct deposition \[45, 46\] and resist based atom lithography \[47\]. In the first scheme, the spatial profile of the atomic beam on the sample is controlled precisely by one of the previously mentioned modulation methods and atoms accumulate on the surface of a substrate. The second technique relies on the exposure of a surface covered with a specific resist to energetic or chemically reactive atoms. Patterns are then transferred to the substrate by the application of an etching technique. Resist based atom lithography has primarily utilised metastable atomic beams (Ne*, Ar* or He*) formed in gas discharge sources. However, other elements, such as indium, gallium, barium, and the chemically reactive species cesium, have also been used. The two schemes for atom lithography are depicted in Figure 1.1.

Recent reviews of the field of atom lithography are given in \[41, 48–50\].
1.2. ATOM LITHOGRAPHY

1.2.1 Direct deposition

Atomic nanofabrication was first demonstrated by the direct deposition of a thermal sodium beam in 1992 [45]. In this experiment, a beam of sodium atoms was focused directly onto a silicon wafer using a one-dimensional standing wave of laser light tuned near the atomic transition at 589 nm and aligned above the sample surface as shown in Figure 1.1 b). The deposited line structures were observed to have the expected periodicity of λ/2. Refinement of this technique resulted in structures with widths as small as 20 nm [26]. Following the success with sodium atoms, the development of laser control for chromium atoms allowed for more permanent nanostructures to be created [46]. By using chromium the sample may be exposed to air after the deposition process. This is due to the formation of a very thin (∼1 nm) protective oxide layer and allows the deposited material to be investigated using a variety of techniques, including an atomic force microscope (AFM).

Further development allowed the formation of structures with even smaller periodicity. A technique where the detuning of the standing wave is switched from positive to negative enabled the double exposure of an individual sample [51]. In this way, atoms were deposited into the previously unexposed regions and produced λ/4 periodic structures. Another technique utilising the strong polarisation gradient formed by two counter-propagating, orthogonally polarised beams resulted in structures with λ/8 periodicity [44]. The polarisation grating formed by the light field induces Raman coherences between the magnetic sublevels of the chromium ground state and results in structures with four times greater periodicity. Experiments using light fields other than a one-dimensional standing wave have created structures such as square [52, 53] and hexagonal arrays [54].

Nanofabricated features have also been observed with other elements such as aluminium [55], ytterbium [56], and iron [57, 58]. Iron nanostructures are of particular interest as they can allow the study of low-dimensional and single domain ferromagnetic phenomena [59].

1.2.2 Resist based atom lithography

The alternative method of atom lithography is much more akin to conventional lithography. In resist based atom lithography a resist coated sample is exposed to the atomic beam and subsequently etched. The etchant dif-
ferentially removes material from the regions exposed to the atomic beam and transfers the pattern into the substrate. Damage of the resist layer can be achieved either by using a reactive species of neutral atoms, such as the alkali metals, or by transferring internal energy from the atom to the molecules in the resist. The transfer of internal energy is readily achieved by using metastable rare gas atoms. Noble gas atoms in a metastable state exhibit internal energies in the range of 8-20 eV which is sufficient to break chemical bonds within a resist layer upon collision.

The selection of an appropriate resist layer is crucial to achieving high resolution pattern transfer. The damage mechanisms involved in resist based atom lithography are highly localised and as such will not penetrate thick resist layers. Self-assembled monolayers (SAMs) [60] are ideal candidates for resist layers in atom lithography. Two commonly used SAMs are alkanethiols, which form on gold substrates, and siloxanes which form on silicon. They form a tightly bound, dense, monomolecular layer which is resistant to wet chemical etching until damaged by the atomic beam. The small footprint of these molecules on the sample surface is essential to achieving maximum resolution from the lithographic process.

In addition to SAMs, the formation of a contamination resist has been observed in atom lithography experiments using beams of metastable argon and neon [61, 62]. The creation of these resists is highly dependent on the partial pressure of background oil vapour in the vacuum system. During exposure to the atomic beam, oil vapour is adsorbed onto the surface and is polymerised by the impact of the metastable atoms. This results in the exposed regions becoming more resistant to the etching process. A representation of the two resist layers and the patterns formed are shown in Figure 1.2.

Resist based atom lithography was first realised with a beam of metastable argon (Ar\(^*\)) [47]. It was used to expose a dodecanethiol resist through a copper grid physical mask with 5 \(\mu\)m wide lines. Upon impact, the internal energy of the Ar\(^*\) atom is transferred to the resist and results in local damage of the SAM. Chemical etching produces a pattern in the underlying substrate, and in this experiment, generated structures with an edge resolution of \(\sim\)80 nm. This technique was extended to metastable states in both helium (He\(^*\)) [63, 64] and neon (Ne\(^*\)) [67, 68] and resulted in resolutions \(\leq\)50 nm. Structures have also been observed in experiments using cesium beams.
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Figure 1.2: The formation of a) positive contrast patterning or b) negative contrast patterning in the underlying substrate is dependent on the length of exposure to the metastable atomic beam. Below a critical dose of metastable atoms, the SAM is damaged and is less resistant to the etching process. Once exposed above the critical dosage, sufficient contamination resist has formed to make the exposed regions more resistant to the etchant. Note: the contamination resist will also form in the absence of the SAM.
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[42] 69 72, indium and gallium beams 73, and atomic beams consisting of
the ground and metastable states of barium 74 75.

The first resist based atom lithography experiment utilising an optical
mask was conducted with a cesium beam 71. The resulting pattern was a
series of parallel lines 120 nm wide and separated with periodicity 426nm,
half the wavelength of the cesium D2 line. One-dimensional standing wave
optical masks have since been used in Ar* 76 and Ne* 68 experiments and
produced structures with widths ≤65 nm. Experiments with He* have im-
plemented both one- 77 and two-dimensional 78 optical masks but with
reduced resolution. The first holographic optical mask for atom lithogra-
phy was demonstrated in 2002 42. A two-dimensional intensity modulated
pattern was stored in a LiNbO3:Fe crystal and transferred to a laser beam
detuned from the cesium D2 line, forming the holographic mask. Cesium
atoms were focused by the light pattern and resulted in a series of horizontal
lines with λ/2 periodicity and a superimposed vertical line pattern with 23.4
µm spacing.

1.2.3 Metastable atoms

Metastable states of atoms occur when the excited state is unable to decay
back to the ground state via a single photon radiative transition. Decay from
these states is only possible via higher order processes and as such they are
long-lived, typically >1 s. The metastable states of noble gas atoms possess
internal energies far above the ground state (8-20 eV). Of particular interest
to this work is the 33P2 metastable state in neon. Selected energy levels of
two excited states of neon are shown in Figure 1.3.

Neon has two metastable states, the 33P0 and 33P2, highlighted in Fig-
ure 1.3. Electric dipole transitions from these states to the 21S0 ground
state are forbidden by the quantum mechanical selection rules 79. The
33P0 state can decay to the 33P1 state through a magnetic-dipole radiative
transition, or to the 33P2 state through an electric-quadrupole radiative
transition. For the 33P2 state, the only decay mechanism is via a spin de-
pendent magnetic quadrupole transition. The 33P0 and 33P2 are therefore
long lived metastable states, and for neon are indicated by the designation
Ne*.

The second excited state of neon consists of the triplet states 33D3,
33D2, 33D1 and the singlet state 31D1. A transition from the 33P2 to the
1.2. ATOM LITHOGRAPHY

Figure 1.3: Partial energy level diagram of neon showing the closed optical transition from the \(^3P_2\) metastable state to the \(^3D_3\) state. The two metastable states are highlighted grey.
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$^{3}\text{D}_3$ state exists where the only decay channel is back to the metastable $^{3}\text{P}_2$ state. This forms a two-level transition which is ideal for laser cooling. Multiple photon excitations may occur between these states without loss through decay to one the short lived $^{3}\text{P}_1$ or $^{1}\text{P}_1$ states. The transition is therefore termed a closed optical transition. The wavelength required for this transition is easily obtained using commercially available dye lasers. The relevant parameters of the $^{3}\text{P}_2$ state, and the $^{3}\text{P}_2 \rightarrow ^{3}\text{D}_3$ transition are provided in Table 1.1.

Table 1.1: Parameters of the $^{3}\text{P}_2$ metastable state of neon and the $^{3}\text{P}_2 \rightarrow ^{3}\text{D}_3$ transition.

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<thead>
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<th>Symbol</th>
<th>Value</th>
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<tr>
<td>Atomic mass</td>
<td>$m$</td>
<td>$3.32 \times 10^{-26}$ kg</td>
</tr>
<tr>
<td>Internal energy</td>
<td>$E_i$</td>
<td>$\sim 16.6$ eV</td>
</tr>
<tr>
<td>$^{3}\text{P}_2$ state lifetime</td>
<td>$\tau_{^{3}\text{P}_2}$</td>
<td>14.73 s</td>
</tr>
<tr>
<td>Transition wavelength</td>
<td>$\lambda$</td>
<td>640.40 nm</td>
</tr>
<tr>
<td>Transition linewidth</td>
<td>$\Gamma$</td>
<td>8.20(2$\pi$) MHz</td>
</tr>
<tr>
<td>Transition lifetime</td>
<td>$\tau = 1/\Gamma$</td>
<td>19.42 ns</td>
</tr>
<tr>
<td>Saturation intensity</td>
<td>$I_s$</td>
<td>4.22 mW cm$^{-2}$</td>
</tr>
<tr>
<td>Capture limit</td>
<td>$v_c$</td>
<td>5.43 m s$^{-1}$</td>
</tr>
<tr>
<td>Doppler limit</td>
<td>$v_D$</td>
<td>0.29 m s$^{-1}$</td>
</tr>
<tr>
<td>Recoil velocity</td>
<td>$v_r$</td>
<td>0.031 m s$^{-1}$</td>
</tr>
</tbody>
</table>

The conventional method of producing metastable atomic beams is via gas discharge systems. However, it is an inefficient process with only $\sim 0.01\%$ of the gas atoms emerging from the discharge in the metastable state [80]. The resulting atomic beam is a combination of metastable and ground state atoms and also contains electrons, ions, and UV photons, all of which may lead to undesirable lithographic patterning.

1.3 Magnetic nanostructures

The ability to fabricate ever smaller electronic devices, particularly those relying on magnetic memory, requires an accurate knowledge of the behaviour of structures with dimensions approaching the fundamental lengths, such as domain wall width. Miniaturisation of magnetic structures has led to the discovery of giant magnetoresistance [81, 82] and the development of
1.3. MAGNETIC NANOSTRUCTURES

the field of spintronics [83]. The production of nano-sized structures using magnetic materials is enabling research into oscillatory exchange coupling, spin-polarised tunnelling, and the effects of proximity and confinement in magnetic memory applications [84, 85]. Large arrays of well defined nanostructures are also providing an opportunity to study important magnetic phenomena such as surface induced anisotropy, domain wall motion, and pinning [59].

A number of techniques have been applied to the formation of magnetic nanostructures including epitaxial film deposition, optical and electron beam lithography, electrodeposition, and scanning probe lithography. Reviews of the various fabrication methods are provided in [84–86].

Atom lithography is particularly well suited to the formation of nanostructured magnetic materials. Direct deposition experiments have already achieved nanostructuring of iron [57] [58] using a standing wave optical mask. However, direct deposition results in the nanostructures being formed on a ‘pedestal’ of the deposited material. If this layer is relatively thick the induced anisotropy in the sample will be limited. An advantage of this technique lies in the selectivity of the optical mask and allows multiple species to be deposited resulting in a doped layer with the concentrations modulated with at least \( \lambda/2 \) periodicity [87].

Previous experiments utilising resist based atom lithography have shown the ability to form micrometer sized structures in iron [88]. These structures were created by exposing an iron coated silicon substrate, capped with a thin gold film and subsequently grown alkanethiol resist layer, to a metastable neon beam through a physical mask. A three step chemical etching process then transferred the patterning to the underlying iron layer. By implementing an optical mask, and refinement of the etching process for higher resolutions, structures with nanometer dimensions should be achievable. The key benefit of resist based atom lithography is that it is not limited to the production of structures using an atomic beam of the desired material. It can therefore be used to pattern semiconductor and dielectric substrates once the required resist layers and etching procedures are established.
1.4 Motivation

To continue the miniaturisation of circuitry and data storage beyond the current 45 nm resolution, alternative lithographic solutions need to be established [89]. Atom lithography is an alternative of interest as the de Broglie wavelength of atoms is significantly shorter than that of light used in optical lithography. The diffraction limit for atoms therefore does not limit resolution for features larger than $\sim 1 \text{ Å}$. However, as with optical lithographic techniques, the continuing advancement relies on further research into resist, source, and mask technologies.

The majority of this work utilises the $^3\text{P}_2$ metastable state of neon and the $^3\text{P}_2 \rightarrow ^3\text{D}_3$ transition for atom lithography. The development and enhancement of a high flux, UV free atomic beam is crucial for atom lithography to be viable as a next generation technique. The low percentage of metastable atoms present in the atomic beam emerging from the source limits the flux to $\sim 10^{14}$ atoms sr$^{-1}$ s$^{-1}$. The presence of UV photons and other energetic constituents in the atomic beam may also limit the achievable resolution. The apparatus described in Chapter 2 was designed in order to increase the flux of the metastable atomic beam and remove all other constituents. Enhancement of the atomic beam is achieved by reducing the velocity and spacial distributions and selectively deflecting the metastable atoms.

Resist layers play an integral role in the formation and resolution of patterning in atom lithography. Without sufficient contrast of the surface properties between exposed and unexposed regions, etching will result in poorly defined features. Investigations into alternative resists and an accurate knowledge of the processes involved during exposure to the metastable atom beam are vital for further development of this technique. Previous experiments using a short chain alkanethiol molecule as a resist [90] were non-quantitative and initial investigations showed the possibility of producing negative contrast lithographic patterns more rapidly than a longer chain alkanethiol. It was the aim of the experiments outlined in Chapter 3 to extend this investigation and clearly define the processes involved in the formation of negative contrast lithographic patterns.

One application of atom lithography is the fabrication of nanostructured magnetic materials. In Chapter 4 the direct deposition atom lithography
1.5. THIS THESIS

process was used to improve the understanding of thin magnetic films with nanometer scale features. In initial experiments, a beam of iron was deposited onto a silicon oxide surface through a standing wave light mask without the implementation of laser cooling. By removing the requirement of laser cooling many technologically interesting elements that do not have closed optical transitions, such as Ga, In, and Si [91], may be used in atom lithography experiments. The geometries of the deposited structures also provide insight into the limitations of the atom lithography process and the effect of substrate dependent diffusion. This work culminated in investigations into the co-deposition of iron and nickel to create nanostructured magnetic films with periodic variations in the spatial concentrations.

1.5 This thesis

In the following chapter the experimental apparatus used to enhance and purify the flux from a DC discharge metastable neon source is detailed. The theory of atom optical centre-of-mass manipulation is also reviewed and applied to simulating the trajectories of metastable neon atoms through a multiple element hexapole magnetic guide. A comparison of the transmission efficiencies resulting from theory and experiment is presented and discussed. Chapter 3 presents the application of the metastable neon beam to atom lithography. Self-assembled monolayers are used as a resist and the damage mechanisms due to metastable atom impact are outlined. The comparison of patterning achieved using long and short chain alkanethiol monolayers is discussed and the importance of background mechanical pump oil in the formation of negative contrast patterns is highlighted. A theory describing the deposition of molecules on a surface is provided and applied to the characterisation of the contamination resist layers.

Work utilising the direct deposition lithographic technique is presented in Chapter 4. This work was performed at the Eindhoven University of Technology. The experimental apparatus is described and its application to the creation of nanoscale iron features is presented. The co-deposition of iron and nickel is also presented. The surface composition and magnetic properties of the deposited material is investigated and discussed.
CHAPTER 1. INTRODUCTION
2.1 Introduction

This chapter details the apparatus used in the experiments described in Chapter 3. The motivation behind the design and construction of the apparatus was to produce a pure, monochromatic, UV free metastable atomic beam for application in atom lithography. A beam of metastable neon atoms is generated in a DC discharge source. This is then cooled in the transverse direction using light from a ring dye laser frequency stabilised near an atomic resonance. Charged constituents of the atomic beam are also removed in this region. In order to produce an atomic beam of purely metastable neon atoms in the $3^3P_2$ state requires subsequent manipulation. The method chosen to extract a single metastable state was to use an arrangement of permanent magnets to deviate the atoms from the original beam path. To facilitate this technique, the atoms are slowed along the beam line using a Zeeman slower with a novel laser beam input arrangement to optimise the atom flux incident on the ‘magnetic guide’.

The design of the magnetic guide is based on similar devices constructed using magnets that produce quadrupole and octupole magnetic fields to deflect alkali metal beams [92, 93]. Using a series of hexapole magnetic lens elements a novel approach to the guiding of metastable neon atoms is presented [94]. Hexapole fields were chosen due to the linearly increasing force exerted upon the atoms as they deviate from the axis of each element. This approach also allows for subsequent focussing of the atomic beam. The de-
vice may also be beneficial for the loading of magneto-optical traps operating in UHV environments, as the magnetic guide could be used simultaneously as a conductance aperture for a differential pumping stage while also removing unslowed or undesirable constituents of the atomic beam.

The laser systems required for the experiments detailed in Chapter 3 are described in Section 2.2. Details of the theory describing how the laser light is used to manipulate the atoms is also covered. Section 2.5 outlines each component of the vacuum system including discussion of the operating parameters and performance, particularly in regard to the hexapole magnetic guide. The magnetic field gradients present in multipole elements and the forces they exert on the neutral atom are also discussed. The systems utilised to detect and analyse the atomic beam are detailed in Section 2.4.

2.2 Laser system

Experimentation was carried out using laser light supplied by two ring dye laser systems, a *Coherent 899* and a *Spectra Physics 380D*. The dye lasers operated with Kiton-red dye and were both pumped by *Coherent Verdi V5* solid state lasers operating at 528 nm that could provide pump powers up to 5.5 W. Power output from the *Spectra Physics 380D* ring dye laser is lower than that of the *Coherent 899* due to differences in the configuration of the internal laser cavity. The output specifications of each laser system are outlined in Table 2.1.

<table>
<thead>
<tr>
<th></th>
<th>Coherent 899</th>
<th>Spectra Physics 380D</th>
</tr>
</thead>
<tbody>
<tr>
<td>Wavelength (vacuum)</td>
<td>640.40 nm</td>
<td>640.40 nm</td>
</tr>
<tr>
<td>Output power</td>
<td>~450 mW</td>
<td>~300 mW</td>
</tr>
<tr>
<td>Linewidth</td>
<td>1 MHz</td>
<td>~1 MHz</td>
</tr>
</tbody>
</table>

An overview of the laser and optics used is shown in Figure 2.1. The linearly polarised output from each laser system passed through a $\lambda/2$ plate and polarising beam splitter (PBS) so that a small amount of light could be reflected and used for laser frequency stabilisation via saturated absorption spectroscopy (see Section 2.2.3). The beam was detuned from atomic resonance using an acousto-optic modulator (AOM).
Figure 2.1: Schematic overview of the laser apparatus.
2.2.1  Coherent 899 ring laser

The laser output from the Coherent 899 single mode ring dye laser was frequency locked using an intracavity etalon assembly and detuned from the $3^3P_2 \rightarrow 3^3D_3$ atomic transition by +40 MHz using an IntraAction AOM40 acousto-optic modulator placed before the saturated absorption cell. In this arrangement, when the laser was locked to the atomic transition frequency using the saturated absorption cell, as described in Section 2.2.3, the laser cavity was actually detuned from atomic resonance to compensate for the shift given by the AOM. This reduced the loss of laser power due to modulation efficiency of the AOM, as a majority of the output power from the Coherent 899 ring dye laser (∼300 mW) was required by the two-dimensional atomic beam collimator, where the optimal frequency detuning is +40 MHz. Therefore only ∼120 mW passed through the AOM and was detuned back to the atomic transition frequency. From this beam only 10 - 15 mW was required for saturated absorption spectroscopy, the remainder was used for a re-collimation stage after the Zeeman slower.

The beam for the two-dimensional collimator passed through an Optics for Research Faraday rotator to prevent back reflections from destabilising the ring dye laser cavity. The beam was then sent to the experiment through a single mode optical fiber using a Newport fiber launcher. 200 mW of light was out-coupled and collimated to a 15 mm ($1/e^2$) diameter and subsequently split into four beams, each of ∼40 mW. These beams constitute the two-dimensional optical molasses used for atomic beam collimation as described in Section 2.5.2.

The beam that passed through the AOM was divided again using another λ/2 plate and PBS. This allowed a small amount of light to be diverted to the saturated absorption cell while the remainder was sent to the experimental apparatus. This beam was again collimated to a $1/e^2$ diameter of 15 mm and directed into the re-collimation chamber through a 2 3/4” viewport. A particular arrangement of mirrors mounted in vacuo, shown in Figure 2.17, allowed a single beam to be retroflected and produce a two-dimensional optical molasses.

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2.2. LASER SYSTEM

2.2.2 Spectra Physics 380D ring laser

The output from the Spectra Physics 380D ring dye laser passed through a Spectra Physics 380D locking unit to lock the laser frequency. This beam was then split using a \( \lambda/2 \) plate and PBS and 10 mW was sent to another saturated absorption cell. The remaining beam was launched into a single mode optical fiber using a Newport fiber launcher and directed to the experiment. Approximately 200 mW was out-coupled and divided into two beams. These beams each passed through a \( \lambda/4 \) plate to create \( \sigma^+ \) polarised light which was then incident on the novel dual mirror arrangement, shown in Figure 2.16 and used to longitudinally slow the atomic beam.

The optimal detuning of these beams was - 140 MHz, achieved via the application of DC offset to the external frequency control input on the laser frequency control electronics. This technique of frequency control was used as the large detuning required was not easily achieved using available AOM’s. This also meant that laser frequency locking to the saturated absorption peak could not be used. However, it was found that during experimentation the long term drift of the laser frequency of the Spectra Physics 380D was less than one natural linewidth of the atomic transition over the period of an hour and could be easily monitored and manually corrected.

The DC offset applied to the laser frequency control was frequency calibrated using the saturated absorption peaks of the two most abundant isotopes of neon, \(^{20}\text{Ne}\) and \(^{22}\text{Ne}\), which are separated by 1.6 GHz as shown in Figure 2.2. A LabView program was written to enable an adjustable, calibrated detuning to be applied once the laser is referenced to the saturated absorption peak at zero detuning.

2.2.3 Saturated absorption spectroscopy

The ring dye laser were both frequency locked, however, fluctuations of external parameters such as ambient temperature and humidity can induce absolute frequency drifts of up to \(~100\) MHz hr\(^{-1}\). In order to lock the laser to the atomic transition over long time periods, saturated absorption spectroscopy was performed using a reference cell containing neon gas. The saturated absorption cell, shown in Figure 2.3 consists of a glass tube filled with 2 Torr of neon and two ring electrodes separated by 70 mm. An electrical discharge created between the two electrodes excites a small portion
of the atoms into the $3^3\text{P}_2$ state. An incident beam of $\sigma^+$ polarised light passes through the cell and optically ‘pumps’ the atoms to the $3^3\text{D}_3$ atomic state if the laser frequency is resonant with the atomic frequency. The beam is then retroreflected and after passing through the cell again is directed onto a photodiode. If the incident laser is accurately tuned to the atomic frequency it will saturate the atomic transition for atoms with no Doppler detuning, i.e. the $\langle v \rangle = 0$ velocity group. The returning ‘probe’ beam cannot excite any further atoms into the $3^3\text{D}_3$ state and the cell is essentially transparent (for the $\langle v \rangle = 0$ velocity group of atoms), resulting in a maximum signal on the photodiode. If, however, the laser frequency is slightly detuned from the resonance condition, the ‘pump’ and ‘probe’ laser beams interact with different velocity groups within the cell. By scanning the laser frequency over a relatively large range (achieved by scanning the laser Brewster plates in this case) a saturated absorption spectrum showing the peak at the resonance condition, as shown in the inset of Figure 2.3 can be obtained. This peak has a FWHM of $\sim 20$ MHz and enables the laser to be locked to the atomic transition frequency to within one atomic linewidth.

In order to scan across the atomic transition while maintaining a laser frequency lock, an AC coil is positioned around the cell to provide an oscillating magnetic field. The magnetic field present induces a Zeeman shift
and detunes the atomic resonance from the laser frequency. By dithering the field, the signal of the ‘probe’ beam on the photodiode produces the saturated absorption peak. By comparing the differentiated signals of the ‘probe’ intensity and the magnetic field, an error signal can be generated and applied to the external frequency control input of the laser electronics \[95, 96\]. Using this technique enables the laser frequency to be altered to compensate for long term drift. Measurements of the long term frequency stability utilising this locking system resulted in a frequency drift of 4.0±0.1 MHz over several hours. There is also a separate coil located around the saturated absorption cell that can detune the atomic transition via the application of a DC magnetic field. However, this technique for detuning the laser frequency was not used in these experiments.

### 2.3 Laser interactions with atoms

This discussion of the interaction of a light field with an atom takes a semi-classical approach based on Ehrenfest’s theorem \[97\]. The interaction is simplified to a two level system, with transitions occurring between the ground and excited states denoted by \( |1\rangle \) and \( |2\rangle \) respectively. The total
CHAPTER 2. EXPERIMENTAL APPARATUS

Hamiltonian of the system can be written as

\[ \hat{H} = \frac{\hat{P}^2}{2M} + \hat{H}_0 - \hat{d} \cdot \hat{E}(\hat{R}, t), \]  

(2.1)

where \( \hat{P}^2/2M \) is the kinetic energy of the atom associated with the centre of mass momentum operator \( \hat{P} \), \( \hat{H}_0 \) is the Hamiltonian describing the internal states of the atom, \( \hat{d} \) is the electric dipole operator, and \( \hat{E}(\hat{R}, t) \) is the electric field operator at the centre of mass position operator \( \hat{R} \). We can describe the time evolution of the momentum and position operators using their commutation relations with the Hamiltonian,

\[ \frac{d}{dt} \hat{R} = \frac{1}{i\hbar} [\hat{R}, \hat{H}] = \frac{\partial \hat{H}}{\partial \hat{R}} = \frac{\hat{P}}{M}, \] 

(2.2)

and

\[ \frac{d}{dt} \hat{P} = \frac{1}{i\hbar} [\hat{P}, \hat{H}] = -\frac{\partial \hat{H}}{\partial \hat{P}} = \nabla (\hat{d} \cdot \hat{E}). \] 

(2.3)

Using Ehrenfest’s theorem \[97\] the force on the atom, \( \vec{F} \), can be defined by

\[ \vec{F} = \frac{d}{dt} \langle \hat{P} \rangle = \langle \nabla (\hat{d} \cdot \hat{E}(\hat{R}, t)) \rangle \] 

(2.4)

and since \( \hat{d} \) is independent of \( \hat{R} \)

\[ \vec{F} = \langle (\hat{d} \cdot \nabla \hat{E}(\hat{R}, t)) \rangle. \] 

(2.5)

The general form of the electric field operator of a light field is given by

\[ \hat{E}(\hat{R}, t) = \hat{e} \vec{E}(\vec{r}, t), \] 

(2.6)

where \( \hat{e} \) is the unit polarisation vector, \( \vec{r} = \langle \hat{R} \rangle \), and \( \vec{E}(\vec{r}, t) \) is the electric field vector. As the spatial variation of the electric field is negligible over the scale of an atom, the electric dipole approximation, the force exerted by the atom can then be defined as

\[ \vec{F} = \langle (\hat{d} \cdot \hat{e}) \nabla \hat{E}(\vec{r}, t) \rangle. \] 

(2.7)

The expectation value can be evaluated using the atomic density matrix, \( \rho \), which describes the internal states of the two level atom. Taking the dipole moment matrix element, \( \langle 1|\hat{d} \cdot \hat{e}|2 \rangle \equiv \mu \), to be real, the expectation value is
then
\[ \langle \hat{d} \cdot \epsilon \rangle = \mu (\rho_{12} + \rho_{21}), \] (2.8)

where \( \rho_{12} \) and \( \rho_{21} \) represent the optical coherences between the two states. This can also be expressed in terms of the two components of the Bloch vector, \( \vec{u}(t) \) and \( \vec{v}(t) \), as
\[ \langle \hat{d} \cdot \epsilon \rangle = \mu (\vec{u}(t) \cos \omega_0 t - \vec{v}(t) \sin \omega_0 t), \] (2.9)

with \( \omega_0 \) being the natural transition frequency between states \( |1\rangle \) and \( |2\rangle \). By taking only the resonant terms in the interaction, using the rotating wave approximation, the components of the Bloch vector are
\[ \vec{u}(t) = (\rho_{12} e^{i\omega_0 t} + \rho_{21} e^{-i\omega_0 t}), \] (2.10)
\[ \vec{v}(t) = -i(\rho_{12} e^{i\omega_0 t} - \rho_{21} e^{-i\omega_0 t}), \] (2.11)

and
\[ \vec{w}(t) = (\rho_{11} - \rho_{22}), \] (2.12)

where \( \rho_{11} \) and \( \rho_{22} \) are the populations of the atomic states \( |1\rangle \) and \( |2\rangle \) respectively. If we now define an arbitrary monochromatic light field,
\[ \vec{E}(\vec{r}, t) = E(\vec{r})(e^{-i(\omega t - \theta(\vec{r}))} + e^{i(\omega t - \theta(\vec{r}))}), \] (2.13)

where \( E(\vec{r}) \) is the spatially varying amplitude of the electric field, \( \omega \) the frequency of the light, and \( \theta(\vec{r}) \) is the phase. The position dependent Rabi frequency is then
\[ \Omega(\vec{r}) = -\frac{\hat{d} \cdot E(\vec{r})}{\hbar}. \] (2.14)

Using the steady state values, \( u_s \) and \( v_s \), of the Bloch vector components in Equations 2.10 and 2.11 allows the force to now be expressed as
\[ \vec{F} = -\hbar (u_s \nabla \Omega(\vec{r}) + v_s \Omega(\vec{r}) \nabla \theta(\vec{r})). \] (2.15)

From Equation 2.15, it is clear there are two components to the force on
CHAPTER 2. EXPERIMENTAL APPARATUS

the atom. The first is proportional to the gradient of the Rabi frequency, and hence the amplitude of the electric field, and the second is proportional to the gradient of the phase of the field. These two components represent the dipole and scattering forces respectively. By making the approximation that the amplitude and phase of the electric field do not vary during the lifetime of the atomic transition, the steady state values of \( u_s \) and \( v_s \) are obtained from the optical Bloch equations and are given by

\[
\begin{align*}
    u_s &= \frac{-4\Omega(\Delta_L + \dot{\theta})}{4(\Delta_L + \dot{\theta})^2 + \Gamma^2 + 2\Omega^2}, \\
    v_s &= \frac{-2\Gamma\Omega}{4(\Delta_L + \dot{\theta})^2 + \Gamma^2 + 2\Omega^2},
\end{align*}
\]

(2.16)

and

\[
\begin{align*}
    \vec{F} &= \frac{\hbar}{4(\Delta_L + \dot{\theta})^2 + \Gamma^2 + 2\Omega^2} \left( -\Delta_L \nabla \Omega^2 + \Gamma \Omega^2 \nabla \theta \right).
\end{align*}
\]

(2.18)

2.3.1 The scattering force

If a travelling wave is now considered which has a phase of \( \theta(\vec{r}) = \vec{k} \cdot \vec{r} \), the electric field can be written as

\[
\vec{E}(\vec{r}, t) = \vec{E}_0(\cos \omega t - \vec{k} \cdot \vec{r}).
\]

(2.19)

The Rabi frequency defined in Equation \( \text{2.14} \) is now constant and hence the force on the atom is now

\[
\vec{F} = \frac{\hbar \Gamma}{4(\Delta_L - \vec{k} \cdot \vec{v})^2 + \Gamma^2 + 2\Omega^2} \Omega^2.
\]

(2.20)

where \( \vec{v} = \partial \vec{r}/\partial t \) is the velocity of the atom. The equation can be further simplified by defining an effective detuning, \( \Delta_{\text{eff}} \), which includes both the laser detuning from the atomic resonance frequency and the Doppler shift of the optical field due to the motion of the atom. The Rabi frequency can
2.3. LASER INTERACTIONS WITH ATOMS

also be expressed in terms of the light intensity, $I$, and the decay rate, such that

$$\Omega = \Gamma \sqrt{\frac{I}{2I_s}} = \Gamma \sqrt{\frac{s}{2}},$$  \hspace{1cm} (2.21)

where $s$ is the saturation parameter at resonance, given by the ratio of the light intensity to the saturation intensity of the transition, $I_s$, which is defined as

$$I_s = \frac{\pi hc}{3\lambda^3 \tau},$$  \hspace{1cm} (2.22)

where $\lambda$ is the wavelength of the light and $\tau$ is the lifetime of the transition, given by $\tau = 1/\Gamma$. Substituting these values into Equation 2.20 results in an expression for the scattering force of

$$\vec{F}_{\text{scat}} = \hbar \vec{k} \frac{\Gamma^2}{4} \left[ 1 + s + \frac{s}{1 + 4\Delta_{\text{eff}}^2/\Gamma^2} \right] = \hbar \vec{k} \mathcal{R},$$  \hspace{1cm} (2.23)

where $\mathcal{R}$ represents the scattering rate. This equation shows that the photon momentum of each photon in the light field, $\hbar \vec{k}$, is imparted to the atom at the rate defined by $\mathcal{R}$. However this is only if the atom decays back to the ground state via spontaneous emission as the photon is then emitted in a random direction. Over many spontaneous emission events the net effect on the atomic momentum due to emission is zero. Using increased light intensity to maximise the scattering force is limited because at very high intensity the rate of stimulated emission also increases. The momentum transfer for a stimulated emission event directly opposes the momentum gained during absorption from laser generated photons. The laser intensities used in the experiments mentioned throughout this thesis do not lead to the stimulated emission process playing a significant role.

Neglecting the effect of stimulated emission, the maximum scattering rate occurs for high intensities where the transition saturates, i.e. the populations of atoms in the ground and excited states are equal. This is the case when each spontaneous emission event is immediately followed by the absorption of another photon. This results in a maximal scattering force of $F_{\text{max}} = \hbar \vec{k} \Gamma / 2$. The Doppler shift present in the effective detuning of Equation 2.23 results in a velocity dependence of the force applied to the atom and enables this force to be utilised for the laser manipulation of atoms.
2.3.2 Optical molasses

An arrangement of light fields which is particularly important to the field of laser cooling of atoms is two near resonant, counter-propagating travelling waves. An atom at rest within the fields will experience an equal force from each of the two waves and the net force on the atom will be zero. If however the atom is moving relative to the propagation axis of the light fields the scattering rate from one wave will be greater than the other. This is due to the Doppler shift induced by the atomic motion. With the correct light detuning, usually $\Gamma/2$ below the atomic resonance frequency, the wave counter-propagating to the atomic motion will be shifted closer to resonance and will scatter more photons than the co-propagating wave. The net force therefore opposes the atomic motion. For low atomic velocities and low intensity, the total force on the atom can simply be expressed as the sum of the two scattering forces

$$\vec{F}_{OM} = \hbar \vec{k} \frac{\Gamma}{2} \left[ \frac{s}{1 + s + 4(\Delta L + \vec{k} \cdot \vec{v})^2/\Gamma^2} - \frac{s}{1 + s + 4(\Delta L - \vec{k} \cdot \vec{v})^2/\Gamma^2} \right].$$  \hspace{1cm} (2.24)

The force can also be expressed in terms of a damping coefficient, $\alpha$, as

$$\vec{F}_{OM} = -\alpha \vec{v},$$  \hspace{1cm} (2.25)

where

$$\alpha = 4\hbar k^2 s \left[ \frac{2\Delta L/\Gamma}{1 + s + (2\Delta L/\Gamma)^2} \right].$$  \hspace{1cm} (2.26)

The fact the force is proportional to the velocity for low velocities ($||\vec{k} \cdot \vec{v}|| \ll \Gamma$), resulting in a viscous damping, lead to the name of this arrangement as an ‘optical molasses’. From Equation $2.24$ it would be expected that the force would reduce the atomic velocity to zero, violating the laws of thermodynamics. However, there is a minimum velocity achievable due to the discrete changes in momentum of $\hbar \vec{k}$, resulting in a diffusion of the atomic momentum. This minimum velocity is called the Doppler cooling limit and for the $3^3P_2$ to $3^3D_3$ transition in metastable neon this velocity is $29$ cm s$^{-1}$.
2.4 Atomic beam detection systems

Three parameters are required to accurately describe the atomic beam, the atom flux, the beam divergence, and the longitudinal velocity distribution. In order to detect and characterise these parameters three separate detection systems were used. Metastable states of noble gas atoms have the advantage that the metastable excited state has significantly more internal energy than the ground state of the atom. The detection systems were based on the liberation of this internal energy, \( \sim 16 \text{ eV for Ne}^* \). The energy of the excited state is typically at least two orders of magnitude greater than the kinetic energy of the centre of mass motion of the atoms constituting the metastable atom beam.

2.4.1 Faraday plate

A metastable atom incident on a surface with a work function of \( \Phi \) can eject an electron from the surface if the internal energy of the excited atom exceeds the work function. The resulting electron current provides a readily measurable quantity for the determination of the atomic flux. The mechanism of the electron ejection from the surface is dependent on the metastable internal energy and the surface composition.

For a metastable atom incident on a conducting surface, Resonance ionisation (shown in Figure 2.4(a)) of the metastable atom may occur. The electron in the metastable state tunnels through the barrier into a vacant energy level in the surface above the Fermi energy. If the energy for recombination of the ion is at least twice the work function of the surface (that is, \( E^+ \geq 2\Phi \)), the resonance ionisation event may be followed by Auger neutralisation (Figure 2.4(b)) of the ion to the neutral ground state. This process involves the tunnelling of an electron from the surface into the vacant ground state hole left in the metastable ion and the simultaneous ejection of a secondary electron from the conduction band of the surface.

If however there is no vacant energy levels in the surface or the surface is contaminated by an adsorbed layer, Resonance ionisation is suppressed and a subsequent Auger de-excitation process may occur (Figure 2.4(c)). In this process an electron from the surface/contaminant tunnels to the ground state of the atom and the electron in the excited state is ejected.

The efficiencies of these processes is dependent on the metastable atomic
species and the surface composition. The efficiency, defined by the secondary electron coefficient $\gamma$, from a chemically cleaned, stainless steel surface has been measured using various methods by [99], [100], and [101]. The secondary electron coefficient for Ne$^*$ ranges from 0.3 to 0.9 from these experiments, however, the most reliable and widely accepted value of 0.30 ± 0.08 was determined via photoionisation by a CW laser [101]. The measured electron current, $I_-$, can now be related to the metastable atom flux, $F$, incident on a surface by

$$F = \frac{I_-}{e\gamma \Delta \Omega},$$

where $\Delta \Omega$ is the solid angle of the detection surface in steradians.

The detector used in the experiments mentioned in the following section and Chapter 3 consisted of a 15 mm diameter 304 stainless steel plate and a stainless steel ring positioned 5 mm in front of the plate, as seen in Figure 2.5. The ring was connected to a 200 V supply to prevent the recombination of the secondary ejected electrons which would result in a reduced electron current. The plate was electrically isolated from the system and connected to a Keithley 485 picoammeter via an electrical feedthrough.

### 2.4.2 Thin wire detectors

In order to measure the spatial distribution of the atomic beam and subsequently the divergence, a detector was constructed using the same principles as the Faraday plate but consisted of a length of wire. The stainless steel
Figure 2.5: Schematic of the Faraday plate detector and thin wire detector mounted to a linear drive feedthrough.
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wire had a diameter of 0.13 mm and a length of 40 mm and was electrically isolated from the square ring mount which was connected to a 200 V supply. The detector was mounted to a stepper motor driven linear feedthrough to enable the wire to be scanned across the atomic beam (see Figure 2.5). The signal from the wire was acquired by a National Instruments PCI-6229 DAQ card and was correlated to the stepper motor position using LabView software.

The atomic beam profile is determined from the measurement by integrating the distribution of the atomic beam along the length of wire

\[ I(x) \approx \gamma e^{d_w} \int_{-l_w/2}^{l_w/2} I_{\text{meas}}(x, y) dy, \]  

(2.28)

where \( d_w \) and \( l_w \) are the diameter and length of the wire respectively, and \( I_{\text{meas}}(x, y) \) is the measured distribution of the atomic beam.

The total atom flux, \( \dot{N} \), can be determined by integrating this equation along the scan direction and can be approximated by the central beam profile, \( I(0) \), and the FWHM beam diameter, \( d_b \),

\[ \dot{N} = \int I(x) dx \approx I(0) d_b. \]  

(2.29)

2.4.3 Time-of-flight analysis

The final parameter required for the characterisation of the atomic beam is the longitudinal velocity distribution, determined through the time-of-flight technique. In this technique the atomic beam is ‘pulsed’ by using a mechanical chopper and allowed to temporally spread, due to the distribution of atomic velocities, before being measured by a position sensitive detector (PSD). A schematic of the system is shown in Figure 2.6.

The mechanical chopper consisted of a 100 mm diameter aluminium disc with a 0.5 mm wide radial slit. The disc, also called the ‘chopper wheel’, was driven by a DC motor and rotated at 90 Hz. Another aluminium plate behind the chopper wheel acted as a physical atomic beam block and atoms were only transmitted through to the detector when the slit in the wheel aligned with a corresponding slit in the plate. An electro-optical switch mounted to this plate allowed the rotation speed of the chopper wheel to be monitored and also provided a trigger signal for the data acquisition electronics.
Figure 2.6: Schematic of the time-of-flight detection system.
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The PSD is a series of three 25 mm diameter multi-channel plates (Quan-
tar Technology 3300) followed by a resistive anode. Upon collision with the
first plate a metastable atom, or other energetic particle (i.e. electron, ion,
or UV photon), ejects a secondary electron from the surface. A 3 kV poten-
tial applied across the plates amplified the signal which was then detected
by the resistive anode. Each impact on the PSD was accumulated by a
multi-channel scaler (MCS) (Norland 5700). The MCS resolved the events
into 1024 10 µs channels. The MCS sweep was gated by the electro-optical
switch and the resulting spectrum was a sum over many individual atomic
pulses.

2.5 Vacuum system

The following section outlines the major components of the experimental
apparatus. The experimental optimisation of the atomic flux through the
Zeeman slower and hexapole magnetic guide are also presented in the fol-
loving sections.

2.5.1 Supersonic DC discharge Ne* source

Atomic beam sources typically consist of a reservoir of gas atoms that ex-
and through an aperture into a region of lower pressure. The expansion is
dependent on the geometry of the aperture, the pressure differential across
the aperture, and the temperature of the gas reservoir. The atomic source
used in the experiments mentioned in this Chapter and Chapter 3 produces
a beam of metastable neon atoms that supersonically expand through the
nozzle aperture.

A schematic of the metastable neon source is provided in Figure 2.7 and
a detailed view of the nozzle in Figure 2.8. The source was mounted to a 6”
flat plate flange that was bolted to the source chamber via a flexible bellows.
This allowed the position of the source nozzle to be translated between 2-18
mm from the skimmer aperture via adjustment of the four micrometers. The
source body consisted of two concentric stainless steel tubes and the sealed
cavity between the inner and outer tubes formed a liquid nitrogen jacket
which allowed the source gas and nozzle assembly to be actively cooled.
The liquid nitrogen jacket was gravity fed from a dewar located ex vacuo.
The inner stainless steel tube provides the conduit for the source gas, which

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Figure 2.7: Schematic of the source assembly with associated connections.

enters the region via a KF25 flange. The gas pressure in this region was monitored with a MKS 122A Baratron pressure gauge mounted on the gas inlet line and controlled via a Granville-Phillips 203019 variable leak valve.

A pyrex tube was mounted coaxially within the conduit and tapers down to a 1.5 mm diameter outlet behind the nozzle. This tube was attached with TorrSeal™ to a stainless steel T-junction that enables the region behind the nozzle to be evacuated by a Pfeiffer-Balzers DUO 20 M 20 m³ h⁻¹ rotary vane pump connected via a KF25 flange. This pump also removes excess gas from the region behind the nozzle during source operation. A KF16 flange provides the mounting point and electrical feedthrough for the high voltage cathode. The cathode was constructed of a stainless steel rod sharpened to a conical tip and tapered to fit within the pyrex tube. The cathode was electrically insulated apart from the sharpened tip to inhibit the cathode from discharging to the stainless steel T-junction.

The source gas enters the region behind the nozzle through a spiral groove around the outer edge of a Teflon spacer. This increases the collisions between the source gas and the liquid nitrogen jacket and reduces the thermal energy of the atoms in the atomic beam. The Teflon spacer also promotes the initiation of the discharge from the cathode to the skimmer rather than to the liquid nitrogen jacket. The nozzle itself was constructed
Figure 2.8: Schematic of the metastable neon source nozzle assembly.
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from boron nitride, an electrical insulator and good thermal conductor, and was sealed to the liquid nitrogen jacket by a Viton o-ring while maintaining thermal contact via an aluminium ring. The aperture in the nozzle had a diameter of 350 µm and was 250 µm long.

The cone shaped brass skimmer had a 1.5 mm aperture and serves multiple purposes; the aperture provides geometric collimation of the atomic beam and also provides differential pumping between the source chamber and the remainder of the experimental apparatus. The skimmer also forms the anode for the electrical discharge and was able to have a positive potential provided to it, to assist the initiation of the electrical discharge, via an electrical feedthrough on the source chamber.

The source chamber providing the low pressure environment, required to achieve a supersonic expansion, was pumped by an Edwards 700 l s\(^{-1}\) oil diffusion pump using Santovac 5 diffusion pump oil. This was backed by a Leybold D253/WS 25 m\(^3\) hr\(^{-1}\) rotary vane pump through an Edwards foreline trap to limit the backstreaming pump oil into the source chamber. The pressure in this chamber was monitored by a Pfeiffer PBR260 full range Bayard-Alpert pressure gauge.

Source operation

During operation the neon gas built to a pressure of between 1-10 Torr behind the nozzle. This gas then supersonically expanded into the source chamber which rose from a background pressure of \(\sim 2 \times 10^{-7}\) Torr, to \(\sim 1 \times 10^{-4}\) Torr during operation. Once gas pressure reached equilibrium the discharge was stuck between the cathode and the skimmer via a 0-2 kV constant current (0-25 mA) supply connected to the cathode electrical feedthrough. Neon atoms were excited to both the \(3^3P_2\) and \(3^3P_0\) metastable states by impact with the electrons in the discharge. The relative proportions of the metastable states are 5/6 and 1/6 respectively, however the excitation process is typically very inefficient with only \(\sim 0.01\%\) of the emerging gas atoms being in an excited state \(\text{SU}\). A majority of the beam was therefore ground state neon, which can lead to an increased background pressure in atom trapping experiments, with a smaller proportion of the beam consisting of electrons, neon ions, and UV photons, results of Penning ionisation and de-excitation of the metastable state.

A more thorough discussion of the characterisation and optimisation of the
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metastable atomic beam source used is provided in [103]. The optimal operating conditions are given in Table 2.2.

Table 2.2: Optimal source operating conditions.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Nozzle to skimmer distance</td>
<td>8 mm</td>
</tr>
<tr>
<td>Nozzle temperature</td>
<td>130±3 K</td>
</tr>
<tr>
<td>Nozzle pressure</td>
<td>3-4 Torr</td>
</tr>
<tr>
<td>Source chamber pressure</td>
<td>1.6 × 10^{-4} Torr</td>
</tr>
<tr>
<td>Cathode current</td>
<td>15 mA (-450 V)</td>
</tr>
<tr>
<td>Max. metastable neon flux</td>
<td>(6.9±1.7)×10^{14} atoms sr^{-1} s^{-1}</td>
</tr>
<tr>
<td>Most probable longitudinal velocity</td>
<td>515±7 m s^{-1}</td>
</tr>
<tr>
<td>Half angle divergence</td>
<td>~100 mrad</td>
</tr>
</tbody>
</table>

2.5.2 Atomic beam collimation

In order to maximise the atomic flux for metastable atom lithography experiments, a scheme to collimate the diverging atom beam from the source was implemented. The method utilised in this experiment was developed by Hoogerland et al. [37], and consisted of multiple laser reflections from two orthogonal pairs of nearly parallel mirrors as shown in Figure 2.9. This arrangement allows the force from an optical molasses, of Equation 2.24, to be applied to atoms with a velocity in the transverse direction of ±50 m s^{-1} at the beginning of the collimation region and reduce the transverse velocity to as small as feasibly possible by the end of the collimator, ~3.6 m s^{-1} in these experiments. This is achieved by variation of the angle of the laser beam with respect to the transverse axis of the atomic beam and hence the effective detuning of the laser.

An atom, with a velocity \( \vec{v} \), interacting with the dominant laser beam of an optical molasses, with a wavevector \( \vec{k} \) as shown in Figure 2.10, experiences a shift in the laser frequency given by the Doppler detuning, \( \Delta_D = -\vec{k} \cdot \vec{v} \). This can be divided into the two orthogonal components,

\[
\Delta_D = -kv_{||}sin(\beta) + kv_{\perp}cos(\beta) \approx -kv_{||}sin(\beta) + kv_{\perp}, \tag{2.30}
\]

where \( \beta \) is the angle of the laser relative to the transverse direction and is assumed to be small such that \( cos(\beta) \approx 1 \). The angle of the mirrors in
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Figure 2.9: One dimension of the optical collimator showing the parameters that affect the performance.

Figure 2.10: Vector diagram for the determination of the Doppler components.
Figure 2.11: Part diagrams for the mirror mount of the two-dimensional optical collimator. The mirrors were fixed to individual holders and the angle of the mirror was adjusted using a precision spring-loaded screw assembly.

Figure 2.9 can therefore be chosen such that the laser detuning matches the Doppler shift for a range of atomic velocities along the collimator.

The mirrors of the collimator were mounted to a slotted aluminium frame via a cantilevered spring and screw assembly, shown in Figure 2.11 which allows precise adjustment of the mirror angle. The mirrors were custom made by Thorlabs and are 98% reflective protected aluminium on an optical glass substrate 150 mm long and 20 mm wide. The mirrors were aligned \textit{ex vacuo} using a helium-neon laser. The collimator mirror mount was located within the vacuum chamber 55 mm after the skimmer.

The vacuum chambers of the collimator apparatus, shown in Figure 2.12, consisted of a stainless steel extension with 6” conflat flanges and four 2 3/4” flanges for the coupling of the laser into the vacuum system, mounted to a glass chamber with metal flanges. The glass allowed for observation of the laser reflections on the mirrors and enabled alignment of the four laser beams. This chamber was then mounted to a 6-way cross which housed the
first thin wire detector. This chamber also contained a pair of electro-static
deflector plates 40 mm wide and 70 mm long. These plates were connected
to a high voltage, DC power supply through an electrical feedthrough and
applied an electric field in the transverse direction of $\sim 60 \text{ kV m}^{-1}$. The ap-
plied field at which all charged constituents of the atomic beam are removed
was determined by ramping the DC voltage applied to the deflector plates
whilst monitoring the electron current from the Faraday plate. The collim-
ometer chambers were evacuated by a Varian Turbo V550 MacroTorr 550
ls$^{-1}$ turbo-molecular pump, which maintained a pressure of $\leq 1 \times 10^{-6}$ Torr
while the source was operating. A second thin wire detector located 680 mm
downstream (not depicted in Figure 2.12) from the first allowed the atomic
beam divergence to be determined. The optimal operating parameters for
the collimator are outlined in Table 2.3.

Table 2.3: Optimal operating parameters for the two-dimensional collimator.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Symbol</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Mirror length</td>
<td>$l$</td>
<td>150 mm</td>
</tr>
<tr>
<td>Mirror separation</td>
<td>$d$</td>
<td>60 mm</td>
</tr>
<tr>
<td>Mirror angle</td>
<td>$\alpha$</td>
<td>2.4 mrad</td>
</tr>
<tr>
<td>Laser input angle</td>
<td>$\beta_0$</td>
<td>120 mrad</td>
</tr>
<tr>
<td>Laser detuning</td>
<td>$\Delta_L$</td>
<td>+40 MHz</td>
</tr>
<tr>
<td>Number of reflections</td>
<td>$n$</td>
<td>28</td>
</tr>
</tbody>
</table>

Four 40 mW laser beams from the Coherent 899 ring dye laser were cou-
pled into the collimator chamber and aligned to achieve 28 reflections from
the internal collimator mirrors. The metastable neon source was operated
at the conditions outlined in Table 2.2 and the thin wire detectors were
used to measure the divergence reduction of the atomic beam. The data is
shown in Figure 2.13 where the black line indicates the atomic beam cross-
section while the collimator was disabled and the red while the collimator
was operational.

The FWHM of the profiles obtained from the first and second thin wire
detectors, while the collimator was operational, were 14.7 mm and 19.4 mm
respectively. This results in an atomic beam divergence of 7±3 mrad. At
a most probable longitudinal velocity of 515±7 m s$^{-1}$ the mean transverse
velocity of atoms emerging from the collimator was therefore 3.6±1.6 m s$^{-1}$. 
Figure 2.12: Experimental apparatus showing the source and collimator chambers. A second thin wire scanner was located in another vacuum chamber 680 mm further along the beam line when performing divergence diagnostics.
Figure 2.13: Measured thin wire current with the optical collimator on (red) and off (black) for (a) the first thin wire detector and (b) the thin wire detector located 680 mm further along the beam line.
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2.5.3 Atomic beam slowing

The atomic beam emerging from the collimator was significantly less divergent than that directly from the source, however, the velocity spread in the longitudinal direction, defined by the axis of the source, was still relatively large, \( \sim 122 \text{ m s}^{-1} \). To reduce this spread and also enable the implementation of the hexapole magnetic guide, the atomic beam was slowed in the longitudinal direction by a Zeeman slower [106].

In order to continually apply the scattering force to atoms over a large range of velocities a method of spatially varying the effective detuning is required. The scheme used for longitudinal velocity slowing utilises the anomalous Zeeman effect to alter the atomic transition frequency and maintain resonance with a counter-propagating laser beam. The anomalous Zeeman effect describes the perturbation of the internal atomic states due to an external magnetic field. The field acts to lift the degeneracy of the magnetic substates, \( m_j \), due to the possible orientations of the magnetic dipole of the atom relative to a magnetic flux density, \( \vec{B} \). To the first order the energy shift of each state in an LS coupling scheme is given by

\[
\Delta E = g_j m_j \mu_B B, \tag{2.31}
\]

where \( \mu_B \) is the Bohr magneton and \( g_j \) is the Landé \( g \) factor. This shift in energy relates to detuning of the atomic transition frequency relative to an unperturbed atom. In a transition between two atomic states both states are shifted according to Equation 2.31 and the overall energy shift of the transition due to the magnetic field is

\[
\Delta E_{\text{trans}} = (g_j' m_j' - g_j m_j) \mu_B B = \mu_{eff} B, \tag{2.32}
\]

where the prime denotes the upper state and \( \mu_{eff} \) is the effective magnetic moment of the transition. The Landé \( g \) factors for the \( 3^3\text{P}_2 \) and \( 3^3\text{D}_3 \) states in metastable neon are \( 3/2 \) and \( 4/3 \) respectively. By using \( \sigma^+ \) circularly polarised light the absorption of a photon is limited to states where \( \Delta m_j = +1 \). After several absorption-emission events the metastable ground state is optically pumped into the highest magnetic sub-states and therefore only transitions between the \( 3^3\text{P}_2(m_j = +2) \) and \( 3^3\text{D}_3(m_j = +3) \) states are allowed.

By using a magnetic field that varies in the direction of the atomic beam
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Figure 2.14: The magnetic flux density of the theoretical field required for the Zeeman slowing of atoms from $v_\parallel = 720 \text{ m s}^{-1}$ and that measured along the axis of the constructed Zeeman slower with all coils carrying 15A. propagation, the field strength can be chosen so that the detuning of the atomic transition compensates for the Doppler shift due to the atoms longitudinal velocity. The effective detuning in the scattering force of Equation 2.23 is now

$$\Delta_{\text{eff}} = \Delta_L + \frac{\mu_{\text{eff}}}{\hbar} B(z) - \vec{k} \cdot \vec{v},$$  \hspace{1cm} (2.33)

where $B(z)$ is the spatially varying magnetic field with $z$ in the longitudinal direction.

The field profile of the Zeeman slower was measured using a translatable Hall probe and is shown in Figure 2.14. The Zeeman slower was constructed from two coaxial 1030 mm long stainless steel tubes welded to 6” conflat flanges. The region between the tubes acts as a water cooling jacket, to limit the heat build up during operation, while the inner tube forms the vacuum chamber. Coils of enamel coated copper wire are wound around the outer tube and consist of 26 layers of varying lengths. The coils were wound such that the inner coils, all 930 mm long providing a bias field, and the outer coils, of varying length which provide the taper of the field in Figure 2.14, could be connected to independent current supplies.

The magnetic field produced by the Zeeman slower was designed to com-
pensate for the Doppler shift of an atom moving at $v_\parallel = 720 \text{ m s}^{-1}$, the capture velocity, at the beginning of the coils with 15 A supplied to all coils. With the laser tuned to the natural transition frequency ($\Delta_L = 0$) it is possible to slow the atoms to zero velocity, an undesirable consequence as we require the beam for subsequent experiments. The laser detuning must therefore be non-zero for operation and the final velocity of the atoms will depend on this detuning and the magnitude of the field at the end of the Zeeman slower. The atoms entering the Zeeman slower below the capture velocity will all be slowed into a narrow velocity group about this final velocity.

**Dual beam Zeeman slowing**

The Zeeman slower was previously arranged to have a single laser beam aligned co-axially and counter-propagating with respect to the atomic beam \[107\]. The laser beam was reflected from a mirror in vacuum and on axis. The mirror had a 1 mm hole bored through it to allow the on axis slowed atoms to pass into subsequent experimental chambers. This small aperture significantly reduced the flux of the subsequent atomic beam. The residual transverse velocity from the collimator also contributes more significantly once the atomic beam is slowed longitudinally, resulting in a larger beam divergence.

To increase the atomic flux available after Zeeman slowing a novel dual mirror input was designed and implemented. This arrangement provides a larger aperture for the atomic beam and also imparts a slight transverse force, in one dimension, to the atomic beam by overlapping the two slowing laser beams throughout the Zeeman slower. To reduce the transverse velocity after longitudinal slowing a re-collimation stage was also used. The Zeeman slowing apparatus is shown in Figure 2.15.

Two 20 mm diameter linearly polarised laser beams from the *Spectra Physics* 380D ring laser passed through $\lambda/4$ plates to create $\sigma^+$ polarised light. These were then coupled through anti-reflection coated viewports into the vacuum chamber and onto the slowing mirrors. The mirrors were fused silica prisms with a protected aluminium reflective coating. They were mounted 8 mm apart by the holder depicted in Figure 2.16. The angle of the laser beams on the mirrors were adjusted so that they enter the Zeeman slower at an angle of 15 mrad with respect to the longitudinal axis, and
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Figure 2.15: Schematic of the Zeeman slower apparatus connected to the final chamber of the optical collimator.

completely overlap at the mid-point of the Zeeman slower. This ensures that the beams overlap over the entire length of the coils and interact with the atomic beam.

Re-collimation stage

During longitudinal slowing the divergence of the atomic beam increases due to the growing significance of the residual transverse velocity from the collimation stage. In this apparatus the divergence of the atomic beam increases from $7 \pm 3$ mrad before the Zeeman slower to $\sim 45 \pm 19$ mrad after longitudinal slowing. To maximise the atomic beam flux for subsequent experiments the divergence was reduced by implementing a re-collimation stage, shown in Figure 2.17. It consists of two fused silica prisms with a protected aluminium reflective coating rigidly mounted in an arrangement which allows a single 20 mm laser beam to be retroflected to create a two-dimensional optical molasses. The mirror holder located the re-collimation stage 40 mm from the end of the Zeeman slower. A 40 mW laser beam from the *Coherent* 899 ring laser was used for re-collimation.

Results

The efficiency of the Zeeman slower was determined by using the time-of-flight method mentioned in Section 2.4. The Zeeman slower apparatus of Figure 2.15 was connected via a bellows to the 6-way cross chamber of the collimator. The chamber containing the re-collimation stage and slowing
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Figure 2.16: The dual beam Zeeman slowing mirror arrangement.

Figure 2.17: Mirror mount for the re-collimation optical molasses.
mirrors was evacuated by a *Pfeiffer-Balzers* TMH 071-P 60 l s\(^{-1}\) turbo-molecular pump. This was then connected to the time-of-flight apparatus which consists of the mechanical chopper wheel mounted within a 4-way cross chamber followed by a 520 mm flight path to the PSD, mounted within the PSD chamber. The PSD chamber was pumped by a *Pfeiffer-Balzers* TPH520 500 l s\(^{-1}\) turbo-molecular pump.

The metastable neon source was operated at liquid nitrogen temperatures with a discharge current of 15 mA. The optical collimator was setup to the parameters outlined in Table 2.3 and the slowing laser beam power per beam was 70 mW. The re-collimation stage had not yet been implemented at the time of performing these characterisations.

The Zeeman slower was operated at a number of different laser detunings and Zeeman slower coil currents and the time-of-flight data analysed. It was found that optimal slowing efficiency was obtained when the inner and outer coil currents of the Zeeman slower were 13 A and 18 A respectively and the laser was detuned -140 MHz from the atomic transition. At this detuning the capture velocity of the Zeeman slower is lowered to \(\sim 680\) m s\(^{-1}\). The velocity distribution obtained for these parameters is given in Figure 2.18.

A significant proportion of the unslowed atomic beam was still present with only a very small slowed atom peak present at 80 m s\(^{-1}\). The slowed atom peak was significantly reduced due to the residual transverse velocity from the collimator and as such many do not traverse the 0.5 mm slit in the mechanical chopper wheel. Previous experiments with the single mirror with a 1 mm hole were unable to resolve slowed atoms below \(\sim 300\) m s\(^{-1}\). Unfortunately, due to the increased atomic flux through the 8 mm aperture in the dual slowing beam apparatus, a velocity profile for an unslowed atomic beam, i.e. when the Zeeman slower is turned off, was not able to be obtained due to saturation of the PSD. Therefore, in order to determine the slowing efficiency the remainder of the unslowed atomic beam in Figure 2.18 was fitted with a Maxwell-Boltzmann distribution and integrated. Using this method an estimate for the total unslowed atomic beam flux was obtained. Upon comparison to the integrated flux of the profile in Figure 2.18 the observed atomic beam flux was reduced by 45%. A significant proportion of the atomic beam is therefore undetected, with a large number being slowed to 80 m s\(^{-1}\) or lower.
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Figure 2.18: The velocity distribution obtained by the time-of-flight method for a laser detuning of -140 MHz and coil currents of 13A and 18A for the inner and outer coils respectively. A slowed atom peak at 80 m s$^{-1}$ is observed. The peaks above 200 m s$^{-1}$ are the remainder of the unslowed atomic beam.

2.5.4 A hexapole magnetic guide

The atomic beam from the source consisted not only of divergent, supersonic metastable neon atoms in the $3^3P_2$ state but also neon atoms in the $3^3P_0$ metastable state, ground state neon atoms, neon ions, electrons, and UV photons. The atoms in the $3^3P_2$ state were collimated and the charged constituents removed in the two-dimensional collimator chambers. The collimated atoms were subsequently slowed by the Zeeman slower and re-collimated. The resulting atomic beam now consists of both supersonic and slowed metastable neon atoms in the $3^3P_2$ and a significantly lower proportion of the $3^3P_0$ metastable neon atoms, ground state neon atoms, and UV photons. To selectively deflect the slowed metastable neon atoms from the atomic beam path, a magnetic guide was constructed using hexapole magnetic fields.

The hexapole magnetic guide relies on the paramagnetic behaviour of Ne$^*$ atoms in the $3^3P_2(m_j = +2)$ state. A paramagnetic atom in a magnetic field will align its magnetic moment, $\vec{\mu}$, with the external magnetic field, $\vec{B}$, such that it minimises the potential energy. The potential is given by
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\[ V = -\vec{\mu} \cdot \vec{B}. \]  

For an inhomogeneous field where \( \vec{\mu} \) is constant and aligned with \( \vec{B} \) the force on the atom is

\[ \vec{F} = \vec{\mu} \cdot \nabla \vec{B} = \mu_B g_j m_j \nabla \vec{B}. \]  

Ne\(^*\) atoms are ‘low-field seekers’ as they minimise the potential energy by aligning the dipole parallel to the applied magnetic field. To apply a significant force on the atom, relatively large magnetic field gradients must be realised. Such gradients can be obtained by the arrangement of rare-earth magnets into multipole magnetic elements. Friedberg and Paul [108] showed that a thermal beam of low-field seeking atoms can be focused by using a hexapole magnet to provide a spatially varying magnetic field.

Figure 2.19 (a) shows a schematic representation of a hexapole magnetic element constructed from \( M \) trapezoidal rare-earth magnets. Due to a relative susceptibility nearing unity, the magnetisation of the rare-earth magnets remains constant for a large range of applied magnetic field strengths. The total field from an arrangement of rare-earth magnets can be calculated using the superposition principle. Halbach [109] showed that the magnetic field within a multipole magnetic element of order \( 2N \), where \( 2N = 6 \) for a hexapole arrangement, with \( M \) trapezoidal magnets can be approximated by

\[ B^*(\rho) = B_r \sum_{v=1}^{\infty} \left( \frac{\rho}{r_1} \right)^2 \frac{n}{n-1} \left[ 1 - \left( \frac{r_1}{r_2} \right)^{n-1} \right] K_n, \]  

where

\[ n = N - vM \]  

and

\[ K_n = \cos^n \left( \epsilon \pi / M \right) \frac{\sin(n\pi M)}{n\pi M}. \]  

In these equations \( B_r \) is the complex magnetisation vector, \( \rho \) is the radial position inside the multipole element, and \( r_1 \) and \( r_2 \) are the radii of inner and outer boundaries of the magnets, as shown in Figure 2.19 (b). \( \epsilon \) represents
a physical form factor for each trapezoidal magnet defining the actual size relative to the maximum possible size, given by $2\pi/M$.

The spatial variation for a perfect multipole magnetic element without segmentation is given by

$$|B(\vec{\rho})| = B_r \left( \frac{\rho}{r_1} \right)^{N-1}.$$  \hspace{1cm} (2.39)

For a hexapole ($2N = 6$) magnetic element this results in a quadratic dependence with the radial position, $\rho$, from the centre of the element. The magnetic field gradient is therefore linear, resulting in a force that increases linearly as a low-field seeking atom deviates from the axis of the element. This has led to the elements being termed a hexapole lens as, in the ‘thin lens’ approximation [110], a focal length for atoms of mass, $M$, traversing the lens of length, $L$, at a velocity, $v_\parallel$, can be defined by

$$f = \frac{Mv_\parallel^2/2}{\mu_B(\partial^2 B/\partial \rho^2)L}.$$  \hspace{1cm} (2.40)

**Design and setup**

The magnetic guide was constructed of nine individual magnetic hexapole lens elements [94]. Each hexapole lens element consists of twelve permanent rare earth magnets held in formation by an aluminium housing, shown in Figure 2.20, with a central aperture of 23.5 mm and an inner radius to the magnets, $r_1$, of 12.7 mm. The individual magnets used had the dimensions
Figure 2.20: Schematic of a hexapole element assembled into an aluminium housing. The arrows indicate the direction of the magnetic field.

6.4×6.4 mm, were 12.7 mm long, and had a surface field strength of ∼0.44 T [111]. As this magnet arrangement is significantly different from the trapezoidal wedges used by Halbach [109], Equation 2.36 may not accurately describe the field within these lens elements.

The magnetic field measured using a Hall probe for the central region of the hexapole lens is shown in Figure 2.21 (a). Line profiles were taken over multiple angles between 0° and 60°, where the angle is defined in Figure 2.21 (a). The field at all other angles could then be determined by using the symmetry of the magnetic field. The individual line profiles of 2.21 (b) show that the uniformity of the magnetic field gradient is lost once the atoms are more than ∼10 mm from the centre of the lens due to the geometry of the magnets used. While the theory of Halbach [109] could not be used directly to analyse the fields within the hexapole lens, by including the second term in the Taylor expansion of Equation 2.36 it predicts the magnetic field to within 1% until approximately 11 mm from the centre of the lens.

A two-dimensional simulation of the atomic trajectories through nine hexapole lens elements with a magnetic field profile given by Figure 2.21 (a) was used to establish the optimum orientation of the elements. The simulation was performed using MATLAB and assumed a constant field
Figure 2.21: (a) The measured magnetic profile at the centre of the element shown in [2.20]. (b) A plot displaying the angular dependence of the magnetic field and showing how the theory diverges from the measured flux at large distances (>11 mm) from the axis of the element.
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Figure 2.22: Simulation of the atomic trajectories through a 9 hexapole element magnetic guide. Metastable neon atoms in the $m_j = +2$ state were incident on the central 8mm of the guide with a longitudinal velocity of 80 m s$^{-1}$ and a maximum transverse velocity of ±4 m s$^{-1}$.

Profile over the length of each lens element and zero magnetic field between successive elements. The atoms entered the first element within the central 8 mm defined by the separation of the Zeeman slower mirrors. The atoms were simulated to have a most probable longitudinal velocity of 80 m s$^{-1}$ and a maximum transverse velocity of ±4 m s$^{-1}$. The typical result of these simulations for atoms in the $m_j = +2$ magnetic substate is shown in Figure 2.22.

From the two-dimensional simulations the hexapole lens elements were arranged such that the initial and final two elements are axially aligned, while the inner five elements are aligned on a 30 degree arc with a radius of curvature of 30 cm, as shown in Figure 2.23. The entire lens system was mounted within a stainless steel chamber with 6” conflat flanges and all interior surfaces were coated to prevent the reflection of UV photons through the chamber. The final experimental setup is shown in Figure 2.24. Faraday detectors were mounted in the 6-way cross collimator chamber, 80 mm before, and immediately after the magnetic guide chamber to enable analysis of the transmitted atomic beam flux. The first two Faraday detectors were on translatable mounts to facilitate insertion and removal from the
CHAPTER 2. EXPERIMENTAL APPARATUS

Figure 2.23: The arrangement of hexapole magnetic elements mounted within the chamber.

atomic beam line. A Leybold TurboVac 151 150 l s$^{-1}$ turbo-molecular pump was connected to the final Faraday detector chamber to aid evacuation the magnetic guide.

Numerical simulation of transmission efficiency

In addition to the two-dimensional magnetic field profile, the magnetic field profile in the $xz$-plane, relative to Figure 2.21 (a), of an individual hexapole lens was also measured. The profile is shown in Figure 2.25. The profile shows that there is a significant variation of the magnitude of the magnetic field along the 12.7 mm length of the hexapole lens. By convoluting this variation with the two-dimensional profile obtained by the theory of Halbach [109], a full three-dimensional profile of the magnetic field was obtained. From this, a simulation of the atomic trajectories in all dimensions was performed. Similarly to the two-dimensional simulation, the magnetic field strength outside of the hexapole lens was neglected.

From Equation 2.35 we witness that the force applied is dependent on the magnetic substate of the atom, and is only positive when $m_j > 0$. Atoms in the $m_j = 0$ experience no force, while atoms in the negative magnetic substates will be accelerated outwards from the centre of the hexapole lens. Therefore simulations were only carried out for atoms in the $m_j = +1$ and
2.5. VACUUM SYSTEM

Figure 2.24: Schematic of the final experimental apparatus used for the characterisation of the hexapole magnetic guide.
Figure 2.25: Measured radial magnetic field profile over the length of a hexapole lens.
To determine the transmission efficiency of the hexapole magnetic guide, an atomic beam with the velocities determined from the output of the Zeeman slower ($v_\parallel = 80 \text{ m s}^{-1}$, and a $v_\perp$ spread of $\pm 4 \text{ m s}^{-1}$) was simulated over the full aperture of the first hexapole lens element. Monte Carlo simulations of the atomic trajectory were performed for approximately 130 million atoms incident at numerous positions within the 23.5 mm aperture. Simulations were performed by using a standard differential equation solver in MATLAB on the Griffith University HPC Cluster V20z.

The results for the transmission efficiency relative to the incident position of atoms in the $m_j$ +1 and +2 substates are presented in Figure 2.26. The region exposed to the atomic beam by the aperture in the Zeeman slower mirrors is denoted by the area inside the rectangle. In Figure 2.26, the radius of curvature is defined by the point $x = -300 \text{ mm}$. It was observed that the maximum transmission probability for the atoms in the $m_j = +2$ substate corresponds to the centre of the initial hexapole lens, with transmission probabilities $>80\%$ over the central $\sim10 \text{ mm}$ of the aperture. Optimal efficiency for the $m_j = +1$ substate, however, occurs closer to the outer radius of the magnetic guide. This occurs due to the reduced force experienced by the atoms in the lower magnetic substate, which therefore require the larger initial force by being displaced from the centre to successfully traverse the magnetic guide.

By integrating over the area exposed through the Zeeman slower mirrors, a value for the experimentally achievable transmission efficiency can be obtained. It was found that 74% of the $m_j = +2$ atoms and 43% of the $m_j = +1$ atoms should be transmitted by the guide when exposed through the Zeeman slower mirrors. Assuming that all magnetic substates are equally populated, that is the atom is not optically pumped into the highest substate, this would result in total atomic flux transmission efficiency of approximately 23%. It was also found from these simulations that by increasing the incident atomic beam divergence by a factor of two, the transmitted atom flux is nearly halved.

It should be noted that to reduce the complexity of the simulation the hexapole lens elements were assumed to be evenly spaced around the $30^\circ$ arc. This is not a completely accurate representation of the experiment arrangement shown in Figure 2.23, however, investigation of the transmis-
Figure 2.26: Transmission efficiency as a function of the initial position for atoms in the (a) $m_j = +1$ substate and (b) $m_j = +2$ substate. The aperture of the Zeeman slowing mirrors is outlined by the rectangle.
sion through the central seven lens elements showed that the transmission efficiency should be within ±5% of the experimental value.

Results

The metastable neon source was operated at the conditions shown in Table 2.2 and the collimator was setup as described in Section 2.5.2. Using a nitrogen buffer gas, the metastable atoms were collisionally de-excited, or ‘quenched’, and UV photons were found to contribute 6% of the total measured current on the first Faraday detector. The resulting atomic beam flux measured by the Faraday detector was \((1.6\pm0.4)\times10^{12}\) atoms s\(^{-1}\). After being longitudinally slowed, the total atom flux measured by the second Faraday detector was \((5.2\pm1.3)\times10^{10}\) atoms s\(^{-1}\) which increased to \((8.5\pm2.1)\times10^{10}\) atoms s\(^{-1}\) when the re-collimation stage was implemented. This increase is comparable to the missing flux observed in the time-of-flight analysis of Section 2.5.3. By considering the divergence of the atomic beam and comparing the measured atomic flux to that measured for the unslowed atomic beam, the Zeeman slower efficiency was determined to be approximately 58%.

The atomic beam incident on the magnetic guide still consists largely of ground state neon atoms but the relative proportion of the other constituents changes significantly after the Zeeman slower. Metastable atoms in the \(^3\text{P}_2\) state now contribute to nearly 100% of the measured current on the second Faraday detector, with less than 2% of the total flux in the \(^3\text{P}_0\) metastable state and only \(~0.7\%\) attributed to UV photons. Once the atomic beam traversed the magnetic guide the flux measured by the final Faraday detector was \((4.4\pm1.1)\times10^{9}\) atoms s\(^{-1}\), giving a transmission efficiency of the magnetic guide of \(~9\%\). However, the emerging beam is purely metastable neon atoms in the \(^3\text{P}_2\) state, as all other constituents are not guided through the 30 degree angle, confirmed by quenching the metastable atoms. The measured transmission efficiency was lower than that predicted by simulations of the magnetic guide. There are a few possible contributions to this discrepancy. Firstly, it was assumed that all the magnetic substates were equally populated as there is no magnetic field present between the Zeeman slower and the magnetic guide to lift the degeneracy of the magnetic substates. As seen from Figure 2.26 if the \(m_j = +2\) magnetic substate was less populated, the measured efficiency will significantly decrease. This
would be the case if there was some residual field and the atoms were optically pumped out of the $m_j = +2$ magnetic substate. In this experiment the re-collimation beams were linearly polarised which has the effect of optically pumping the atoms into magnetic substates with low magnetic projection quantum numbers. As mentioned previously, variations between the geometric arrangement of the magnetic guide will also have some small but finite (expected to be $\approx 5\%$) contribution to the discrepancy observed. Finally, by not being able to accurately measure the proportion of atoms that have been slowed, the calculated transmission efficiency may be underestimated.

The transmission efficiency of the hexapole magnetic guide is relatively low when compared to another Ne$^*$ deflection scheme realised by Scholz et al. [112]. They have measured a transmission efficiency of 25% using a two-dimensional magneto-optic molasses tilted at 68°. With a transmitted atom flux of $\sim 2 \times 10^7$ atoms s$^{-1}$ it is significantly lower than the transmitted flux of the hexapole magnetic guide but by implementing an optical pumping scheme between the Zeeman slower and first hexapole lens, comparable transmission efficiencies should be achieved. It should be noted that the 30° arc of the hexapole magnetic guide was arbitrarily chosen and that it may be extended to any angle. Though for larger angles it likely has an associated drop in transmission efficiency.

### 2.6 Conclusions

A metastable neon beam generated by a liquid nitrogen cooled, DC discharge source was purified to an atomic beam consisting of only one metastable state. The atomic beam was first cooled in the transverse direction by a two-dimensional optical collimator which decreased the atomic beam divergence to $7 \pm 3$ mrad.

The atomic beam was then slowed in the longitudinal direction by a novel dual beam Zeeman slower. This arrangement significantly increases the transmitted slowed atom flux compared to previous experiments. Atoms with longitudinal velocities below $\sim 680$ m s$^{-1}$ were slowed to a velocity of 80 m s$^{-1}$ with an efficiency of approximately 58%. The slowed atoms were re-collimated by a two-dimensional optical molasses at the end of the Zeeman slower.

This beam was then incident on a hexapole magnetic guide. Metastable
neon atoms in the positive magnetic substates were guided through a 30° arc and are subsequently deflected from the other constituents of the atomic beam. This results in a pure, UV free metastable atomic beam with a flux of $(4.4 \pm 1.1) \times 10^9$ atoms s$^{-1}$. The design is cost effective and easily adaptable to any atomic beam line. It is straightforward to adapt the device so that it could also act as a conductance aperture for differentially pumped UHV vacuum systems. However, the current flux is insufficient for use in atom lithography experiments without further enhancement of the phase space density.
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Chapter 3

Metastable atom lithography

3.1 Introduction

To create lithographic patterns on a sample using metastable noble gas atoms requires the selection of an appropriate resist layer. The areas of the resist exposed to the metastable atom beam must undergo a change which either aids or inhibits a subsequent etching procedure. Of particular interest is the change which will inhibit the etching of the exposed regions. If the metastable atom beam incident on the sample is spatially modulated by a physical ‘grid’ mask, the resulting pattern after etching will yield ‘islands’ of the underlying substrate. This pattern is a negative contrast image of the mask. Such an array created in a ferromagnetic surface is potentially interesting for research into nano-magnetics [85].

Previous atom lithography experiments using metastable neon successfully utilised dodecanthiol (DDT) as a resist and observed the formation of negative contrast imaging at dosages of $1.2 \times 10^{15}$ atoms cm$^{-2}$ [113]. An initial investigation into an alternative resist, ethanethiol (ET), showed negative pattern formation after exposure to only $(1.3 \pm 0.3) \times 10^{14}$ atoms cm$^{-2}$. This prompted further investigation into the possibility of using the shorter chain alkanethiol to more rapidly produce negative contrast lithographic patterns. However, the vacuum system had been significantly altered since the results with DDT were produced. A replacement turbo-molecular pump had been installed on the sample chamber and fore-line traps placed after all mechanical rotary pumps. As a result, the partial pressure of mechani-
The dosage required to obtain positive patterns using DDT as a resist remained similar to those reported previously at \((1.7 \pm 0.4) \times 10^{14}\) atoms cm\(^{-2}\), but the production of negative patterning became difficult to replicate. A dosage of \((8.4 \pm 2.1) \times 10^{15}\) atoms cm\(^{-2}\) was required for reliable negative contrast images in the current vacuum system. This corresponds to a dosage of \(\sim 17\) Ne\(^*\) atoms for every alkanethiol molecule, an order of magnitude higher than in previous experiments \([113]\).

The availability of MPO contaminant is clearly dependent on the individual vacuum apparatus and is critical to the metastable dosages required to produce negative lithographic patterns. This reinforces the findings of Close et al. \([114]\) and indicates that the deposition of background MPO and cross-linking of the MPO with the resist molecules is the most likely mechanism for the production of durable negative resists.

This chapter presents the investigation of three resists used to create lithographic patterns on a gold coated, silicon substrate using the atomic beam described in Chapter 2. The two alkanethiol resist layers are described in Section 3.2.1. A theory describing the growth of the third resist, a contamination resist, is presented in Section 3.2.2. Section 3.4.4 examines the lithographic patterns formed using the short-chain ET molecule and compares them to those produced using DDT as a resist layer. The growth of a carbonaceous resist due to the presence of background mechanical pump oil vapour is characterised in Section 3.4.5. The techniques used to characterise the surfaces are outlined in Section 3.3.

### 3.2 Resists in atom lithography

#### 3.2.1 Alkanethiols on gold

To be a viable resist and accurately reproduce the desired pattern, the resist layer must form a dense uniform layer, have highly localised damage upon metastable atom impact, and provide selectivity to the etching process after exposure to the atomic beam. A self-assembled monolayer (SAM) of alkanethiol molecules \((\text{CH}_3(\text{CH}_2)_n\text{SH})\) on gold, shown in Figure 3.1, has been found to be particularly useful \([115]\). The formation of the SAM is achieved by submersing a chemically cleaned, gold coated silicon wafer in a dilute
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Figure 3.1: Illustration of a self-assembled monolayer of alkanethiol molecules on a gold surface. The molecules shown are dodecanethiol where the film thickness is \( \sim 15 \text{ Å} \) and the molecular spacing is \( \sim 5 \text{ Å} \). The tilt angle of 30° is induced by van der Waals forces.

solution of the alkanethiol in ethanol.

The sulphur head of the alkanethiol molecule binds to the gold surface via chemisorption and allows a high surface mobility of the adsorbed molecules. The exothermic nature of this process results in the molecules forming a closely packed layer, as all surface binding sites are filled. This causes the van der Waals forces between the alkyl chains \( ((\text{CH}_2)_n) \) to become significant and over the period of several hours, re-orients the molecules into the highly ordered monolayer shown in Figure [3.1]. The alkyl chain is typically orientated 20° - 30° relative to the surface normal. The resulting surface consists of the terminal methyl group \( (\text{CH}_3) \) and provides a hydrophobic nature which benefits the selectivity of a wet chemical etchant.

The two alkanethiol molecules utilised in these experiments are dodecanethiol (DDT - \( \text{CH}_3(\text{CH}_2)_{11}\text{SH} \)) and ethanethiol (ET - \( \text{CH}_3(\text{CH}_2)_1\text{SH} \)). DDT on gold forms a uniformly 15 Å thick layer with the alkyl chains tilted at \( \sim 30° \) and having an intermolecular spacing of 5 Å. Ethanethiol does not achieve the same level of order due to the significantly shorter alkyl chain.

Upon impact with the resist, a metastable neon atom de-excites and emits a secondary electron with sufficient energy to break chemical bonds and modify the surface. These secondary electrons only have a mean free path within the surface of \(< 1 \text{ nm} \), resulting in very localised damage of the SAM. Figure [3.2] outlines the possible bond breaking mechanisms for alkanethiols exposed to a metastable atom [114].
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Figure 3.2: Damage mechanisms of SAM molecules exposed to a metastable atom beam. (1) C-H bond breaking results in a surface less resistant to the chemical etchant. (2) C-C bond breaking yields surfaces more resistant to the chemical etch process. Figure adapted from [114].

The damage mechanisms shown in Figure 3.2 are divided into two branches. The first results from the scission of a C-H bond. In this case, a radical CH group is formed and through radical transfer may react with a background species and form a polar bond. Alternatively the radical transfer may lead to the dissociation of the sulphur head and alkyl chain and the subsequent desorption of the molecule from the surface. These mechanisms lead to an increase in the surface energy and make the surface more susceptible to a wet chemical etchant. If, however, a C-C bond is broken, the alkanethiol chain can be fragmented and the radical transfer leads to a crosslinking either between neighbouring SAM molecules or the SAM molecules and a background species. The resulting surface becomes more resistant to the wet chemical etching process, yielding a negative contrast pattern of the mask.

In previous experiments [61, 62, 116] lithographic patterns were observed to change from positive contrast to negative for metastable atom dosages above a critical dosage of metastable atoms. We have also found that the critical dosage was highly dependent on the background partial pressure of MPO. Close et al. [114] investigated the fragmentation of DDT molecules.
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exposed to low energy electrons and found that there was no evidence of C-C bond breaking and only C-H and C-S bond scission occurred. The likely mechanism for the creation of negative contrast patterns is therefore from the interaction of the metastable atoms with the background MPO. The resist layer would then be formed by either the cross-linking of the alkyl chains and the MPO or, if the SAM has already been desorbed by breaking the C-S bond, a purely cross-linked hydrocarbon layer.

3.2.2 Formation of contamination resists

To investigate the growth of a purely hydrocarbon resist we will consider an uncoated sample in vacuum. For any surface in vacuum where there is some background partial pressure of hydrocarbon vapour, MPO in this case, a small amount of the vapour will be adsorbed and could possibly form a thin film. The theory that describes the interaction of the background vapour and surface was developed by Irving Langmuir in the early 20th century. Much of the theory here is described in his work [117–119], but is also expressed in the formalism used by Hirsch [120].

There is a flux of molecules incident on the surface per unit area and time, defined by \( F \). If some proportion of the molecules are adsorbed by the surface, given by the sticking coefficient \( s \), and reside for some mean time, \( \tau \), before re-evaporating from the surface, the rate equation defining the surface density of adsorbed hydrocarbons, \( N \), is given by

\[
\frac{dN}{dt} = F \cdot s - \frac{N}{\tau}.
\]

(3.1)

It was expressed by Langmuir [118] that “when gas molecules impinge against any solid or liquid surface they do not in general rebound elastically, but condense on the surface, being held by the field of force of the surface atoms”. As such the sticking coefficient \( s \) can assumed to be unity. The solution is then

\[
N = \tau \left[ F - c_0 e^{\left(\frac{-t}{\tau}\right)} \right],
\]

(3.2)

where \( c_0 \) is a constant dependent on the initial state of the surface.

If the surface is subsequently exposed to a flux, \( f \) (atoms cm\(^{-2}\) s\(^{-1}\)), of metastable atoms, there are two additional loss mechanisms of the adsorbed hydrocarbon. The first is by desorption due to metastable atom impact,
where the desorption cross-section is given by $Q_0$. The adsorbed hydrocarbons can also be polymerised or cross-linked. The polymerised material is more strongly bound to the surface and the cross-section for polymerisation is denoted by $\sigma$. The surface density of adsorbed hydrocarbons is now given by

$$\frac{dN}{dt} = F - N \left[ \frac{1}{\tau} + (Q_0 + \sigma) f \right],$$

where time is now zero from the moment the atomic source was initiated, and the solution is given by

$$N = \frac{1}{\frac{1}{\tau} + (Q_0 + \sigma) f} \times \left[ F - c_1 e^{-\left(\frac{1}{\tau} + (Q_0 + \sigma) f\right)t} \right],$$

where $c_1$ can be calculated by using the value of $N$ from Equation 3.2 at the time when the atomic source was started.

As we wish to investigate the growth of the hydrocarbon resist it is more important to have an expression for the increasing surface density of polymerised material, $P$. The rate at which polymerisation occurs is given by

$$\frac{dP}{dt} = \sigma f N - Q f P.$$  \hspace{1cm} (3.5)

The second term represents a loss of polymerised material due to sputtering, with $Q$ representing the cross-section of polymerised hydrocarbon. Sputtering losses using a metastable atomic beam are negligible and this equation simplifies to

$$P = \frac{\sigma f}{\frac{1}{\tau} + (Q_0 + \sigma) f} \times \left[ Ft + \frac{c_1}{\frac{1}{\tau} + (Q_0 + \sigma) f} e^{-\left(\frac{1}{\tau} + (Q_0 + \sigma) f\right)t} \right] + c_2.$$ \hspace{1cm} (3.6)

At $t = 0$, $P = 0$,
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\[ c_2 = \frac{-\sigma f c_1}{(1 + (Q_0 + \sigma)f)^2}. \]  

(3.7)

3.3 Characterisation of the surface

In order to examine the processes involved during resist exposure to the metastable beam, two techniques were employed. The first, contact angle goniometry, probes the surface energy of the sample. This provides insight into the integrity of the SAM and the possible damage mechanisms involved. The second is an ellipsometric technique that investigates the formation of thin films on surfaces. This allows the refractive index and thickness of the film to be measured and can also indicate the possible constitution of the film. Imaging and analysis of structures on the surface of the samples was undertaken using atomic force microscopy (AFM).

3.3.1 Contact angle goniometry

Contact angle goniometry is a technique that utilises a small droplet of liquid to measure the wetting properties of a surface. Wetting occurs when the fluid and surface are brought into contact and is quantified by the contact angle defined in Figure 3.3. A droplet will spread over, or wet, a surface that has a high surface energy, such as a surface bound by strong chemical bonds (i.e. the metallic bonds in gold). On surfaces with low surface energy, like that formed by the SAM, the droplet will form a contact angle greater than 90°.

The wettability is determined by three interfacial boundaries; the solid-liquid (SL), solid-gas (SG), and liquid-gas (LG) interfaces. For a static droplet in thermodynamic equilibrium with the surface, the contact angle, \( \theta \), is related to the interfacial tensions by Young’s equation [121]

\[ \gamma_{SG} = \gamma_{SL} + \gamma_{LG}\cos \theta, \]  

(3.8)

where \( \gamma_{SG} \) is the surface tension between the solid and gas, \( \gamma_{SL} \) between the solid and liquid, and \( \gamma_{LG} \) between the liquid and gas. The contact angle is specific to each solid-liquid-gas system and provides information on the interaction between the surface and the liquid, and the properties of the surface. A contact angle of less than 90° means that the surface is wettable or hydrophilic while an angle \( \geq 90° \) generally characterises a
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Figure 3.3: The contact angle (θ) of a droplet on a surface is defined by the surface tensions between the solid and liquid, $\gamma_{SL}$, liquid and gas, $\gamma_{LG}$, and solid and gas, $\gamma_{SG}$.

The contact angle (θ) of a droplet on a surface is defined by the surface tensions between the solid and liquid, $\gamma_{SL}$, liquid and gas, $\gamma_{LG}$, and solid and gas, $\gamma_{SG}$.

surface as not-wettable or hydrophobic [121]. Superhydrophobic surfaces have contact angles greater than 150°.

To investigate the wetting properties of the resist layers, a contact angle goniometer, based upon the design of Aurenty et al. [122], was constructed. A schematic of the contact angle measurement system is shown in Figure 3.4. The apparatus consisted of a Hamilton 701N 10 µL syringe, a Panasonic WV-BP334 CCD camera, and a translatable sample mount. Small droplets, $\leq 2 \mu$L, of liquid (in this work, 18 MΩ grade H$_2$O) were suspended from the tip of the syringe, and the sample mount raised until the droplet made contact with the substrate. The sample mount was then lowered slowly to transfer the droplet from the syringe tip to the sample. The CCD camera was used to image the resulting drop. By determining the width of the contact area and the height of the droplet, depicted in Figure 3.5, the contact angle can be determined using geometry and is given by

$$\theta = 2\arctan\left(\frac{h}{w/2}\right).$$

(3.9)

It must be noted that this method for determining the contact angle is only valid for small droplets, $\leq 8 \mu$L, as larger droplets are deformed due to gravity. The results of contact angle measurements conducted on surfaces with DDT, ET, and contamination resists can be found in Section 3.4.4.
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Figure 3.4: Depiction of the contact angle goniometer apparatus used to determine the wetting properties of surfaces.

Figure 3.5: Depiction of an imaged droplet indicating the height, $h$, width, $w$, and contact angle, $\theta$. 
3.3.2 Ellipsometry

Ellipsometry is a well-established technique for determining the optical properties and thickness of a thin film [123]. It is particularly useful for studying the formation of thin films on surfaces. Ellipsometry is a non-destructive technique that utilises the property that the polarisation of light changes upon reflection from a surface. When the surface is also covered by a thin film, the entire system influences the change in polarisation. This technique is extremely sensitive to changes in the film thickness and can be used to determine film thicknesses on the order of angstroms.

Since its origin in 1889 [124] ellipsometry has undergone much refinement and can be divided into several experimental configurations including null [125], rotating analyser [126], spectroscopic [127], and imaging [128, 129]. The rotating analyser configuration has been found to have an advantage in the speed of measurement while maintaining an accuracy on the order of angstroms [120]. The ellipsometer utilised in Section 3.4.5 to study the kinetics of the contamination resist formation is based on the computer-assisted ellipsometric thickness analyser developed by Hauge and Dill [120] and is shown schematically in Figure 3.6.

The intensity of light incident on the photodetector, regardless of it’s polarisation, can be described by [126]

\[ I(\theta) = I_0(1 + a_2\cos 2\theta + b_2\sin 2\theta), \]  

(3.10)

where \( I_0 \) is the average intensity and \( \theta \) is the angle of the analyser’s polarisation axis with respect to the incident plane. The coefficients \( a_2 \) and \( b_2 \) were determined through Fourier analysis in MATLAB and contain all the polarisation information. These coefficients can then be used to determine the ellipsometric parameters \( \psi_a \) and \( \Delta_a \), where \( \tan \psi_a \) is the p-wave (the polarisation state normal to the surface) to s-wave (polarisation in the plane of the surface) amplitude ratio and \( \Delta_a \) is the relative phase between the p-wave and s-wave at the analyser,

\[ a_2 = -\cos 2\psi_a, \]  

(3.11)

\[ b_2 = \sin 2\psi_a \cos \Delta_a. \]  

(3.12)
Figure 3.6: Schematic of the rotating analyser ellipsometer. The optics were secured to rails which may be independently rotated about a pivot point located directly below the sample.
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The polarisation at the analyser can now be represented by the complex parameter $\rho_a$,

$$\rho_a = \tan\psi_a e^{i\Delta_a}. \hspace{1cm} (3.13)$$

The polarisation of the light incident and reflected from the surface can now be expressed by the parameters $\rho_i$ and $\rho_r$ respectively. The incident polarisation is determined by aligning the optical rails in Figure 3.6 such that the light is not reflected by the surface and directly enters the analyser (therefore $\rho_a = \rho_i$). The apparatus is then returned to the configuration shown in Figure 3.6 and the polarisation measured is that reflected from the sample ($\rho_a = \rho_r$). From these two measurements the complex ratio $\rho$ can be calculated,

$$\rho = \frac{\rho_r}{\rho_i} = \tan\psi e^{i\Delta}. \hspace{1cm} (3.14)$$

The reflecting surface can be regarded as a three layer system consisting of the ambient medium (vacuum in this case), film, and substrate shown in Figure 3.7. The p-wave and s-wave components of light have reflection coefficients from the vacuum-film interface of

$$r^p_{12} = \frac{n_2 \cos\phi_1 - n_1 \cos\phi_2}{n_2 \cos\phi_1 + n_1 \cos\phi_2} \hspace{1cm} (3.15)$$

and

$$r^s_{12} = \frac{n_1 \cos\phi_1 - n_2 \cos\phi_2}{n_1 \cos\phi_1 + n_2 \cos\phi_2}, \hspace{1cm} (3.16)$$

where $n_1$ and $n_2$ are the refractive indices of the ambient medium and the film respectively, and $\phi_1$ and $\phi_2$ are the angles of incidence and refraction respectively.

Similar expressions can be defined for the film-substrate interface, giving $r^p_{23}$ and $r^s_{23}$. The total reflection coefficients for the three layer system are then given by

$$R^p = \frac{r^p_{12} + r^p_{23} e^{iD}}{1 + r^p_{12} r^p_{23} e^{iD}} \hspace{1cm} (3.17)$$

and

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Figure 3.7: Reflection of light from a surface covered by a thin film.

\[ R^s = \frac{r_{12}^s + r_{23}^s e^D}{1 + r_{12}^s r_{23}^s e^D}. \]  
(3.18)

The value of \( D \) is defined by

\[ D = -\frac{4\pi n_2 \cos \phi_2 d}{\lambda}, \]  
(3.19)

where \( d \) is the thickness of the film and \( \lambda \) is the wavelength of light used.

The ratio of the p-wave to s-wave total reflection coefficients is equal to the ratio of ellipsometric polarisation states of the reflected and incident light, defined using the coefficients \( a_2 \) and \( b_2 \), and is given by

\[ \rho = \frac{R^p}{R^s} = \tan \psi e^{iD}. \]  
(3.20)

Substitution of the total reflection coefficients into Equation 3.20 results in a quadratic of the form

\[ C_1(e^D)^2 + C_2(e^D) + C_3 = 0, \]  
(3.21)

where \( C_1, C_2, \) and \( C_3 \) are complex functions of the form \( f(n, \phi, \Delta, \psi) \). For a given measurement, Equation 3.21 results in two solutions for \( D \), and hence film thickness \( d \). The complex coefficients result in complex values of \( d \), but as the film thickness is a real quantity, the solution to Equation 3.21
which yields a real value is the true solution. Experimental errors result in imaginary components for both solutions and the true thickness is chosen as the one with the smallest imaginary component. The imaginary part of the solution for the thickness is then used as a relative measure of error.

3.4 Exposure to metastable atoms and etching

3.4.1 Sample preparation

In order to obtain reproducible, high resolution lithographic images, the samples must be cleaned and prepared in a strict and meticulous manner. The substrates used in this work were (100) oriented test grade silicon wafers (Addison Engineering) of 100 mm diameter and 474 - 575 µm thickness. The wafers were then cleaved into 25 mm × 25 mm samples and cleaned with analytical reagent (AR) grade acetone, then isopropanol, and subsequently dried with nitrogen gas.

The samples were immediately transferred to an Emitech K575D dual head argon ion sputter coater for vapour deposition. The sputter coater is able to coat the sample with either of two target materials, an oxidising target (chromium in this case) or a Noble target (gold). The silicon sample was first coated with a thin layer (∼10 nm) of chromium as it readily adheres to the native silicon oxide layer present on the sample and enables a subsequent gold layer of ∼40 nm to be deposited. The rms surface roughness of the gold layer was determined to be 2.5±0.1 nm by an AFM. The average grain size was on the order of 20 nm.

After coating, the samples were immersed in a 2 mMol solution of either DDT or ET in AR grade ethanol for 16 hours. This produced complete SAM coverage while avoiding issues that arise from excess immersion [130]. The samples were removed from the solution and dried with nitrogen before being transferred directly to the experimental apparatus.

3.4.2 Exposure to the atomic beam

The prepared samples were loaded into a four sided sample carousel depicted in Figure 3.8. One side of the carousel housed a 15 mm diameter Faraday plate detector to measure the atomic beam flux. The remaining sides contain recesses for the samples which were then secured behind a transmission
3.4. EXPOSURE TO METASTABLE ATOMS AND ETCHING

Figure 3.8: The sample mount used for lithographic pattern formation. Three samples may be mounted behind the transmission masks and a Faraday plate detector was mounted on the remaining face.

mask. The transmission masks consist of copper transmission electron microscope (TEM) grid masks attached over a $3 \times 3$ array of holes in a thin stainless steel sheet. The TEM mask dimensions are 60 $\mu$m pitch, 20 $\mu$m bar, and 40 $\mu$m $\times$ 40 $\mu$m square holes. They were rigidly adhered to the stainless steel sheet and located $\sim 100$ $\mu$m from the sample surface.

The sample carousel was positioned 395 mm downstream from the skimmer and mounted to a rotary vacuum feed through in place of the first thin wire detector shown in Figure 2.12. The chambers were then evacuated and the metastable neon source initiated. The samples were exposed to various metastable atom dosages to determine the critical dosage for the transition from positive to negative contrast lithographic patterns.

The metastable flux was measured before and after sample exposure and was determined to remain constant over all exposures. As the atomic beam collimator of Section 2.5.2 was unnecessary for these experiments, the atom flux was also relatively uniform over the area of the samples. The measured Ne$^*$ atom flux was $(3.0 \pm 0.7) \times 10^{11}$ atoms cm$^{-2}$ s$^{-1}$. The partial pressure of MPO during sample exposure was measured using a Stanford Research Systems RGA-100 residual gas analyser (RGA) and was typically $8 \times 10^{-9}$ Torr. The exposed samples were then removed from vacuum and chemically etched as described in the next section.

Contact angle goniometry was also performed on SAM coated and bare
gold samples exposed to various metastable atom dosages. This allowed the wetting properties of the surfaces to be defined as a function dosage. To enable this, the transmission masks previously used were replaced by masks with a relatively large 8 mm × 8 mm open aperture. A DDT, ET, and bare gold sample was placed in each of the three recesses on the sample carousel and exposed to identical atomic beam dosages. The samples were removed from vacuum and placed immediately on the contact angle goniometer to limit contamination. Five droplet images were recorded from different positions within the exposed portion of each sample. This was repeated for a number of metastable atom dosages and the results are given in Section 3.4.4.

3.4.3 Chemical Etching

Etching of the exposed samples was performed using an aqueous ferro/ferricyanide etchant. The etching of gold is an electrochemical process where the oxidation of gold determines the rate of the reaction. Ferrocyanide (K₄Fe(CN)₆) is included in the solution as it is beneficial in reducing the number of defects in the gold pattern during etching. The etching solution was made using 250 mL of distilled water (18 MΩ quality). The composition of the aqueous ferro/ferricyanide etchant is outlined in Table 3.1.

<table>
<thead>
<tr>
<th>Material</th>
<th>Concentration (M)</th>
</tr>
</thead>
<tbody>
<tr>
<td>KOH</td>
<td>1.0</td>
</tr>
<tr>
<td>K₂S₂O₃</td>
<td>0.1</td>
</tr>
<tr>
<td>K₃Fe(CN)₆</td>
<td>0.01</td>
</tr>
<tr>
<td>K₄Fe(CN)₆</td>
<td>0.001</td>
</tr>
</tbody>
</table>

Etching times were typically 8-10 minutes for positive pattern transfer with DDT resists. Samples were periodically removed from solution during the process and inspected with an optical microscope to determine the pattern contrast. Etch times were significantly longer to transfer negative patterns with DDT resists, typically 15-20 minutes. Bare gold samples and samples prepared with an ET resist generally only required ~5 minutes to transfer negative contrast patterns.

Samples were rinsed in de-ionised water once sufficient pattern contrast
3.4. EXPOSURE TO METASTABLE ATOMS AND ETCHING

was observed. They were subsequently dried with nitrogen and placed in a
desiccator before further analysis.

3.4.4 Results with alkanethiol resists

The Ne\(^*\) source outlined in Section 2.5.1 was not operated under liquid ni-
trogen conditions during the following experiments as a cooled atomic beam
was not necessary. Collimation of the atomic beam was also unnecessary
due to the proximity of the samples being exposed. The resulting atomic
beam had a most probable velocity of \((580\pm20)\) m s\(^{-1}\). The source chamber
maintained a pressure of \((8.5\pm0.1)\times10^{-5}\) Torr while the source was oper-
ating. Charged particles were removed before the sample carousel by the
electrostatic deflector plates. The sample chamber (collimator chambers in
Figure 2.12) was pumped by a Leybold Turbovac 151 turbo-molecular pump
backed by a Varian SD-451 dual stage rotary vane pump. This chamber
achieved an ultimate pressure of \(<1\times10^{-7}\) Torr, rising to \((2.0\pm0.1)\times10^{-6}\)
Torr while the source was operating.

Previous results showed the onset of positive and negative contrast pat-
terning on DDT to occur at dosages of \(1.9\times10^{14}\) atoms cm\(^{-2}\) and \(1.2\times10^{15}\)
atoms cm\(^{-2}\) respectively and negative pattering on bare gold samples was ob-
served at dosages above \(6.2\times10^{14}\) atoms cm\(^{-2}\) \[113\]. The dosage for the transi-
tion from positive to negative contrast patterns corresponds to a dosage of
\(~2.4\) Ne\(^*\) atoms for every SAM molecule. Similar experiments yield dosages
of 4-10 Ne\(^*\) atoms per SAM molecule \[67, 68\]. This indicates that the critical
dosage is very dependant on the experimental apparatus. The MPO partial
pressure during the previous exposures was \(~4\times10^{-7}\) Torr.

The vacuum system had been significantly altered since these initial
results were produced. The turbo-molecular pump connected to the sample
chamber replaced an older model and fore-line traps were placed after all
mechanical rotary pumps. As a result, the partial pressure of MPO in the
system was reduced by more than an order of magnitude to \(~8\times10^{-9}\) Torr.
The dosage required to obtain positive patterns on DDT samples in the
new system remained similar at \((1.7\pm0.4)\times10^{14}\) atoms cm\(^{-2}\), however, the
production of negative patterning became difficult to reproduce. A dosage
of \((8.4\pm2.1)\times10^{15}\) atoms cm\(^{-2}\) was required for reliable negative patterning
of DDT, ET and bare gold samples. This corresponds to a dosage of \(~17\)
Ne\(^*\) atoms for every SAM molecule, an order of magnitude higher than in

79
The reduction in the levels of MPO contaminant is critical to the meta-
stable dosages required to produce negative lithographic patterns. This
reinforces the findings of Close et al. \cite{114} and indicates that the deposition
and cross-linking of the MPO with SAM molecules is the most likely mech-
anism for the production of durable negative resists. In the case of the bare
gold coated samples it is clear that the negative resists are the result of a
MPO contamination resist.

In previous work \cite{113} X-ray photoelectron spectroscopy was performed
on DDT and bare gold samples that had been exposed to the Ne* beam at
dosages above and below the critical dosage required for negative contrast
patterning. Above the critical dosage there was a significant increase in
the carbon (1s) and oxygen (1s) peaks in the recorded spectrum. This is
indicative of the formation of a carbonaceous layer from the background
MPO, a result also observed by Johnson et al. \cite{61}. Further investigation
into the formation of the contamination resist is discussed in Section 3.4.5.

Figures 3.9 and 3.10 show AFM scans of the lithographic patterns ob-
tained. The footprint of the AFM tip used was <15 nm. It should also be
noted that all measurements have been corrected for the angle of the line
profile relative to the step edge. The step profile for positive (Figure 3.9(a))
and negative (Figure 3.9(b)) patterns on DDT samples reveal a FWHM step
width of \(\sim 120\) nm and a step height of \(\sim 60\) nm. The negative contrast pat-
ttern on the bare gold sample (Figure 3.10(a)) displays similar dimensions
of \(\sim 100\) nm and \(\sim 45\) nm respectively. ET samples (Figure 3.10(b)) how-
ever, exhibit a much larger step width of \(\sim 640\) nm and the step height is
significantly reduced, indicative of a resist which is not as resilient to the
chemical etching process. This is consistent with the results observed by Xin
et al. \cite{90} confirming that ET is not a viable resist under these conditions.
The surface features observed on the unetched regions in Figure 3.9(b) are
likely the result of adsorbed water or poor SAM coverage, resulting in a
non-uniform negative resist formation.

To investigate the reduced resolution of the patterning in ET compared
to those in DDT and bare gold samples, the surface energy of the exposed
surfaces was quantified using contact angle goniometry. Figure 3.11 shows
contact angle as a function of metastable atom dosage for DDT, ET and
bare gold samples.
3.4. EXPOSURE TO METASTABLE ATOMS AND ETCHING

Figure 3.9: AFM images and line profiles of gold patterns produced using DDT as a resist in metastable atom lithography. (a) Positive contrast patterns were created with a metastable atom dosage of $(1.7 \pm 0.4) \times 10^{14}$ atoms cm$^{-2}$ and (b) negative contrast patterns were created with a dosage of $(8.4 \pm 2.1) \times 10^{15}$ atoms cm$^{-2}$. The white line on the images indicate the region of the line profile.
Figure 3.10: AFM images and line profiles of negative contrast patterns produced in gold using (a) a bare gold substrate and (b) a substrate using ET as a resist in metastable atom lithography. Both patterns were created with a metastable atom dosage of \((8.4 \pm 2.1) \times 10^{15}\) atoms cm\(^{-2}\). The white line on the images indicate the region of the line profile. Bright spots on the images indicate dust contamination.
Figure 3.11: Contact angle measurements for DDT, ET, and bare gold surfaces after exposure to a metastable neon beam.
CHAPTER 3. METASTABLE ATOM LITHOGRAPHY

Unexposed DDT was observed to be hydrophobic and had a contact angle of 108°, indicating a highly ordered and defect free monolayer. Exposure to the metastable beam initially results in a reduction of hydrophobicity, with the best wettability contrast between exposed and unexposed DDT occurring at a dosage of \( \sim 7 \times 10^{14} \text{ atoms cm}^{-2} \). Further exposure to the metastable atoms increases the contact angle. This is an indication that the surface has begun to cross-link between the SAM molecules and the background MPO. Patterns formed at dosages in this transition region were observed to contain a mixture of both positive and negative contrast features.

The ET and bare gold unexposed samples were hydrophilic and it can be seen from Figure 3.11 that there was no significant initial reduction in the contact angle with increasing metastable atom dosage. For ET the contact angle only increased and no positive patterns were observed for these samples. The bare gold data shows a gradual increase in the contact angle, consistent with the growth of a carbonaceous resist layer from the MPO contaminant. No positive patterns were produced using bare gold substrates.

After a dosage of \( \sim 7 \times 10^{14} \text{ atoms cm}^{-2} \) it was observed that the ET and bare gold samples exhibit similar increasing trends. This indicates that ET is most likely desorbed from the surface upon exposure to the metastable beam. The short alkyl chain in ET leads to weak interchain van der Waals interactions, resulting in a poorly ordered and defect prone monolayer. This is evidenced by the hydrophilic nature of the unexposed sample. The disordered monolayer allows metastable atom penetration to the C-S bond and subsequent desorption of the entire ET molecule, as inferred by Xin et al. \[90\]. As the ET is desorbed, the gold surface is then open to the deposition of background MPO. This will lead to a negative resist formation similar to that developed on the bare gold sample. Negative patterns in ET resist samples are therefore likely formed by desorption of the SAM followed by deposition of MPO and subsequent cross-linking and carbide formation.

The long alkyl chains of DDT exhibit strong interchain van der Waals interactions and produce an ordered, close packed monolayer with few defects. The long, ordered chains provide adequate resistance to scission of the C-S bond. As such, C-H bond breaking and polar molecule formation is the likely mechanism for the formation of positive contrast patterns. The
3.4. EXPOSURE TO METASTABLE ATOMS AND ETCHING

unexposed layer was highly hydrophobic and Ne\textsuperscript{+} exposure resulted in a significant reduction in hydrophobicity and effective wetting and rapid positive patterning with strong contrast was observed. Negative patterns are most likely formed by the cross-linking of SAM molecules with background MPO.

3.4.5 Contamination resist formation

To examine the growth of a purely hydrocarbon resist, the ellipsometer depicted in Figure 3.6 was adapted to attach to a vacuum chamber. The sample was mounted to a ULTIMA kinematic optical mount within this chamber and was located approximately 400 mm downstream of the skimmer aperture. This enabled the \textit{in situ} measurement of the hydrocarbon film growth. A schematic of the experimental arrangement is shown in Figure 3.12. The experimental apparatus was operated under the same conditions as those mentioned in the previous section. The samples used in this analysis were prepared as described in Section 3.4.1 but were left as uncoated, bare silicon samples. All rotary pumps on the experiment used Inland19 mechanical pump oil and the partial pressures of the contaminants was measured using an RGA.

The ellipsometer was constructed around a 6-way cross vacuum chamber with 6 inch conflat flange viewports enabling sufficient optical access. A \( \sim \)3 mW, 0.5 mm diameter, collimated beam from a Spectra Physics 117a intensity stabilised He-Ne laser passed through a linear polariser set at 12° relative to the incident plane (x-z plane as shown in Figure 3.12) before passing through a calibrated quarter-wave retarding plate. The laser light passed through the viewport before reflecting from the silicon surface. Upon re-emerging from the vacuum system the light reaches the analyser. The analyser consisted of a linear polariser mounted in a computer controlled rotation mount, rotating at \( \sim \)0.1 Hz, and a Thorlabs DET110 fast photodetector. The laser, optical elements, and analyser were rigidly mounted to optical rails which pivot about the plane normal to the sample and allowed the angle of incidence to be well defined.

The metastable beam source was initiated after allowing the background MPO to form a thin film on the silicon sample. In this investigation periods greater than \( 5 \times 10^4 \) sec from the initiation of the vacuum pumps were found to be sufficient. The flux of metastable atoms was measured using a translatable, biased, stainless steel Faraday plate mounted directly in front of the sample.
Figure 3.12: Schematic of the experimental apparatus used to characterise the growth of a carbonaceous film.
of the sample mount. The measured flux of atoms remained constant at 
\((3.0 \pm 0.7) \times 10^{11}\) atoms cm\(^{-2}\) s\(^{-1}\) during source operation. Contributions to 
the partial pressure of MPO due to source operation was also examined, 
using an RGA, and found to be negligible.

The data taken from the ellipsometer was analysed in MATLAB using 
the bulk refractive indices of silicon and background pump oil of 3.917 + 
0.0122i \[131\], and 1.4796 \[132\] respectively. The native oxide layer present on 
the silicon sample was assumed to be of uniform thickness over the probed 
area and was accounted for via the introduction of a thickness offset in 
subsequent analysis. It was also assumed that the carbonaceous layer was 
void free as similar experiments show that the layers are nearly 100\% carbon 
\[67\] \[133\]. The refractive index of void free carbon was then taken to be 1.95 
\(- 0.79i \[134\]. Figure 3.13 shows a typical time evolution of film thickness 
on a silicon substrate as measured using the \textit{in situ} ellipsometer.

There were two stages of growth during this experiment: the deposition 
of oil onto the surface from the background vapour, described using Equation 
\[3.2\] and the growth of a carbonaceous layer due to the bombardment of 
metastable atoms, described by Equation \[3.6\] There are, however, two 
regions of interest in the initial stage of the film growth labelled A and 
B in Figure 3.13. During the initial evacuation of the vacuum apparatus, 
the deposition of oil is rapid due to the high partial pressure of oil vapour 
from the rotary vane pumps. The hydrocarbon flux in this region is varying 
rpidly and is not well known due to the limitations of the RGA and the 
switching of vacuum pumps and valves. Analysis of this stage of growth 
is not required as sufficient data is obtained from region B of Figure 3.13 
the point where the hydrocarbon flux stabilises, to accurately determine the 
constants of Equation \[3.2\] From \(~5\times 10^{4}\) sec the hydrocarbon flux remained 
relatively constant at \((5.5 \pm 2.5) \times 10^{10}\) molecules cm\(^{-2}\) s\(^{-1}\).

Region C of Figure 3.13 shows the growth of the carbonaceous film 
due to exposure to metastable neon atoms. The hydrocarbon flux while 
the metastable atom source was operating was constant at \((9.9 \pm 1.0) \times 10^{11}\) 
molecules cm\(^{-2}\) s\(^{-1}\). The growth rate of the carbonaceous film was found 
to be only 0.12\pm0.02 nm hr\(^{-1}\). This is significantly lower, by a factor of 
14, than that reported by another group using Ne\(*\) \[67\]. However there are 
significant differences in metastable atom flux, the background hydrocarbon 
species and flux, and the method of film thickness measurement.
Figure 3.13: Example of a typical film thickness measurement. Regions A, B, and C show the growth during chamber evacuation, while under relatively constant vacuum, and during exposure to the metastable atom beam respectively. Variations in the measured thickness of ±1 nm were observed across the samples and are attributed to variations in the silicon oxide layer thickness. However, the growth rate during exposure to the atomic beam was consistently measured to be $0.12 \pm 0.02$ nm hr$^{-1}$. 
3.5. Conclusions

By introducing a volume of the MPO molecules to the surface density defined in Equation 3.2, we have an expression for the thickness of deposited material with respect to time. This enabled the determination of two important variables via curve fit optimisation to region B of Figure 3.13. A value for the mean residence time of an oil molecule on the surface, $\tau$, was found to be $(99.1 \pm 0.3)$ seconds. This is an order of magnitude greater than the value derived by Hirsch [120] but is closely approximated by the steady state solution to Equation 3.1. The value for the volume of an oil molecule was also obtained and determined to be $(8.04 \pm 0.01) \times 10^{-22}$ cm$^3$. This value was found to be well approximated by assuming a spherical molecule and knowing the molecular mass and bulk density of the contaminant, a result also observed by Hirsch [120] in the calculations of collision cross-sections.

Using the value of $\tau$, analysis of the growth of the carbonaceous layer during metastable atom exposure was undertaken. An estimate for the polymerisation cross-section was obtained by projecting the area of the oil molecule onto the surface. The desorption, or dissociation, cross-section is, however, unknown. Hirsch [120] assumed that polymerisation was a more probable process during ion impact and as such, $Q_0 \ll \sigma$. In this investigation it was found that the desorption cross-section was 100 times greater and was the dominant process during metastable atom exposure. By defining an expression for the thickness of the polymerised layer, as done previously for the hydrocarbon layer, and fitting to the data of region C of Figure 3.13, the cross-sections were determined to be $(4.1 \pm 0.3) \times 10^{-15}$ cm$^2$ and $(395.8 \pm 23.6) \times 10^{-15}$ cm$^2$ for polymerisation and desorption respectively. The volume of a carbon atom was also determined from analysis of the carbonaceous growth. The volume measured was $(3.6 \pm 0.3) \times 10^{-24}$ cm$^3$, yielding an atomic radius of $(0.95 \pm 0.06)$ Å. This is in agreement with the radius for carbon atoms in a lattice, 0.914 Å, reported by Pauling [135], and supports the assumption that carbon films grown over long time scales contain little to no voids.

3.5 Conclusions

Comparison of the patterns formed by metastable atom lithography utilising alkanethiol SAM resist layers has shown that very short chain alkanethiols, such as ET, do not form a viable resist. By examining the surface energies
using contact angle goniometry it was found that samples using ET as a resist only produce negative contrast patterns due to the similar wetting properties for exposed and unexposed regions below the critical dosage of $7 \times 10^{14}$ atoms $\text{cm}^{-2}$. It is likely that the ET molecules are desorbed from the surface during exposure to the metastable beam and replaced by a background MPO molecule. Above the critical dosage these samples react in a manner similar to bare gold samples and form a carbonaceous resist layer. The presence of ET molecules may however lead to defects in the carbonaceous resist and result in the poor patterns observed for ET coated samples.

The experimental vacuum apparatus was found to have a significant effect on the dosages required to form negative contrast patterns on all samples. We confirmed that the partial pressure of background MPO is a critical factor for the transition from positive to negative pattern formation on DDT samples. The formation of negative patterns on DDT samples is more likely to form due to the cross-linking of the DDT molecules with background MPO than with adjacent DDT molecules, as predicted by Close et al. [114].

Using ellipsometry the growth of a carbonaceous film during exposure to a metastable atomic beam was able to be characterised. It was found that the desorption cross-section of carbonaceous material from a silicon surface via Ne* impact was many times larger than the polymerisation cross-section. This contradicts an assumption made by Hirsch [120] for growth during ion bombardment. The values determined, along with simple estimates for the mean residence time, volume, and cross-section of the contaminants involved, provide reasonable initial values for the application of the theory, outlined in Section 3.2.2, to other metastable atom experimental apparatus.
Chapter 4

Direct deposition of iron and nickel

The experimental work described in this chapter was undertaken in the Coherence and Quantum Technology research group at the Eindhoven University of Technology.

4.1 Introduction

This chapter presents the alternative method for atom lithography, direct deposition. In particular, the deposition of ferromagnetic elements and the magnetic properties arising from the nanostructuring of the surface is investigated. There are three ferromagnetic elements at room temperature; iron (Fe), cobalt (Co), and nickel (Ni). The physical and optical properties of these elements are provided in Table 4.1.

Table 4.1 shows that Co has many undesirable properties for atom lithography. Primarily, the presence of a nuclear spin, which leads to a complicated hyperfine structure in the ground state reducing the population which can be addressed using a single laser. The wavelength of the transition is also technically challenging to create. The large natural linewidth of the transition and the high saturation intensity also compound the complexity of using this element.

The remaining elements, while better candidates, also present some problems. The wavelength of the Ni transition is also difficult to produce, requir-
### Table 4.1: Properties of the three room temperature ferromagnetic elements; iron, cobalt, and nickel.

<table>
<thead>
<tr>
<th>Element</th>
<th>Magnetic Properties</th>
<th>Isotopes and Abundances</th>
<th>Atomic Properties</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Fe</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Co</td>
<td></td>
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<tr>
<td>Ni</td>
<td></td>
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#### Magnetic Properties

<table>
<thead>
<tr>
<th>Element</th>
<th>Curie temperature (K)</th>
<th>Wien's mean free path (Å)</th>
<th>Atomic magnetic moment (μB)</th>
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<tr>
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#### Isotopes and Abundances

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<thead>
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<th>Isotope</th>
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</thead>
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<tr>
<td>56Fe</td>
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<tr>
<td>58Ni</td>
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<td>28</td>
<td>0.6</td>
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</table>

#### Atomic Properties

<table>
<thead>
<tr>
<th>Percentage</th>
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<th>1% Co</th>
<th>1% Ni</th>
<th>1% Zn</th>
<th>1% Cu</th>
<th>1% Ag</th>
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</tbody>
</table>

#### Table 4.1: Properties of the three room temperature ferromagnetic elements; iron, cobalt, and nickel.
4.1. INTRODUCTION

ing the frequency doubling of a dye laser. The dye laser would also require
a linewidth and frequency stability of better than 500 kHz to maintain reso-
nance within one natural linewidth of the transition in Ni. The Fe transition
is more accessible using a frequency doubled titanium-sapphire laser. How-
ever, the transition from the $^5\text{D}_4$ state to $^5\text{F}_5$ state is not closed and can
decay to one of three intermediate states with a combined probability of
$1/243$. While this poses a problem for laser cooling it has a reduced effect in
atom lithography due to the large frequency detuning and short interaction
time for a standing wave light mask. Fe also has a larger magnetic moment
than that of Ni which may be advantageous for the investigation into the
creation of magnetic nanostructures and it is for these reasons that Fe was
chosen as the element used for these experiments.

This chapter begins with a brief overview of the vacuum and laser sys-
tems used in the direct deposition experiments of Fe. The methods used to
investigate the surfaces composition and magnetic properties are outlined
in Section 4.3. The results from an investigation into the formation of Fe
structures without using laser cooling is presented in Section 4.4.1. By us-
ing a small aperture atomic beam source placed at a relatively long distance
from the sample, the beam incident on the substrate is locally geometrically
collimated. The section of the atomic beam incident on each lens of the
standing wave light field has a local offset angle relative to the substrate.
This affects the height, width, shape, and position of the created structures.
The effects present in the simulated results are partially obscured in the
experiment by a substrate dependent diffusion of atoms and the scattering
and interference of the standing wave light field just above the substrate
which limits the quality of the lithographic mask.

The chapter concludes with the application of this technique to the co-
deposition of Fe and Ni. By simultaneously depositing Ni, which is un-
affected by the standing wave light mask, the film has a variation in its
composition as well as surface nanostructuring with $\lambda/2$ periodicity. This
produces an alternating hard and soft ferromagnetic material. By suitable
nanostructuring of the soft and hard phases of a composite magnetic mate-
rial, improvements in the permanent-magnetic performance beyond that of
the hard phase alone is achievable [136]. The initial investigations into the
magnetic properties of nanostructured Fe-Ni surfaces created using atom
lithography are given in Section 4.4.2.
CHAPTER 4. DIRECT DEPOSITION OF IRON AND NICKEL

Figure 4.1: Vacuum apparatus for the deposition of iron and nickel. Also shown are the load lock/sample storage chamber and the magnetic translator used to exchange the samples in the deposition chamber.

4.2 Experimental setup

4.2.1 Vacuum system

A schematic of the experimental vacuum system is provided in Figure 4.1. The apparatus can be separated into three atomic beam sources, the deposition chamber, and the load lock/sample storage.

Atomic beam sources

The three atomic beam sources utilised in experiments were the Fe source, a Ni source, and a silver (Ag) source. The Ni source was used for the co-deposition experiments outlined in Section 4.4.2 while the Ag source was used to coat the deposited structures with a non-magnetic, protective film to prevent oxidation of the Fe layer and enable ex vacuo analysis of the samples.

To deposit Fe at a reasonable flux, temperatures up to 2000 K are required. This was achieved using a custom-built effusive Knudsen cell depicted in Figure 4.2. A 1000 W graphite heating element is used to heat an Al₂O₃ crucible. The exterior of the oven is insulated using tantalum
4.2. EXPERIMENTAL SETUP

(Ta) foil and was actively water cooled. This thermal source had a 1 mm diameter opening and was located 740 mm from the sample. The Fe atoms expand through the aperture into the source chamber which was pumped by a \textit{BOC Edwards} EXT250M 250 \text{ls}^{-1} turbo-molecular pump. The turbo-
molecular pump was backed by a \textit{BOC Edwards} oil-free, dry scroll pump (XDS10). During operation of the Fe source this chamber maintained a pressure between \(10^{-5}\) to \(10^{-6}\) Torr. The source chamber was separated from the following chamber by a skimming aperture which enables differential pumping. This chamber was a 6-way cross and allows the implementation of a laser cooling stage which was not utilised in these experiments. The chamber was pumped by a \textit{BOC Edwards} EXT250 250 \text{ls}^{-1} turbo-molecular pump which was backed by the previously mentioned dry scroll pump. The characteristics of the Fe atomic beam are given in Table 4.2.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Symbol</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Atom source aperture diameter</td>
<td>(D) (mm)</td>
<td>1</td>
</tr>
<tr>
<td>Atom source temperature</td>
<td>(T) (K)</td>
<td>2000</td>
</tr>
<tr>
<td>Source to sample distance</td>
<td>(L) (m)</td>
<td>0.74</td>
</tr>
<tr>
<td>Average longitudinal velocity</td>
<td>(&lt;v_{\text{long}}&gt;) (m s(^{-1}))</td>
<td>(1.0 \times 10^{3})</td>
</tr>
<tr>
<td>Longitudinal velocity spread (RMS)</td>
<td>(\sigma_{v_{\text{long}}}) (m s(^{-1}))</td>
<td>357</td>
</tr>
<tr>
<td>Transverse velocity spread (RMS)</td>
<td>(\sigma_{v_{\text{trans}}}) (m s(^{-1}))</td>
<td>0.35</td>
</tr>
<tr>
<td>Deposition rate</td>
<td>(\delta_{\text{thickness}}) (nm hr(^{-1}))</td>
<td>(\sim 10)</td>
</tr>
</tbody>
</table>

The Ni source was attached to the deposition chamber at an angle of 45 degrees relative to the Fe source and sample. The cell was a commercially available Createc high temperature effusion cell (HTC-40-2-180-WK-Hor) with an Al\(_2\)O\(_3\) crucible and 1 mm diameter output aperture. The cell was located 365 mm from the sample and deposits \(\sim 6\) nm hr\(^{-1}\) when operating at 2150 K. The chamber containing the Ni source was evacuated by a \textit{BOC Edwards} EXT70M 65 \text{ls}^{-1} turbo-molecular pump. The 45\(^\circ\) angle of incidence on the sample may introduce further complexities to the magnetic structures due to an induced anisotropy. An effect that has previously been observed in evaporated thin films [138].

An effusive Ag source was present between the deposition and sample storage chambers of Figure 4.1. This allowed a protective capping layer to be applied before removing the samples from the load lock. The Ag source,
Figure 4.2: Schematic of the Fe source mounted to a 6 inch flange. Current for the graphite heating coil (3) is provided via two copper electrodes (1). The molten Fe (4) is contained within an Al₂O₃ crucible (2). The external housing of the oven is insulated by Ta foil (5) and is water cooled(6). A skimmer (7) allows differential pumping between the source and subsequent vacuum chambers.
4.2. EXPERIMENTAL SETUP

Figure 4.3: The Ag source used to cap the deposited Fe structures with a protective layer. The tungsten crucible (1) is heated by a Ta filament (3) wound around an alumina spacer (2). The temperature of the crucible is monitored by a thermocouple (6). The exterior of the oven is insulated by Ta foil (4) and water cooled (7). Exposure to the Ag atomic beam is controlled using a mechanical shutter (5).

depicted in Figure 4.3 was custom-built with a tungsten crucible and 2 mm diameter nozzle aperture. The crucible is heated to 1140 K using a Ta filament wound around an alumina spacer. The exterior of the source is insulated using a Ta foil stack and was water cooled. The source aperture was placed 70 mm from the sample surface and deposits $\sim 12 \text{ Å min}^{-1}$. Samples were typically only exposed to the Ag source for 5 minutes.

Sample holder

The load lock/sample storage chamber of Figure 4.1 allows up to four samples to be exposed to the atomic beams without breaking the vacuum. The sample holder, shown in Figure 4.4 may be moved to and from the deposition chamber by using a magnetic translator. The sample storage and
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Ag source chambers were pumped by a Pfeiffer-Balzers TPH240 240 l s\(^{-1}\) turbo-molecular pump.

The substrate and mirror were both rigidly mounted to the sample holder to suppress fluctuations of their positions relative to one another. This allowed the 8×8×3 mm, 99% reflective mirror to be aligned perpendicular to the substrate \textit{ex vacuo} to within 1 mrad. Adjustment of the mirror orientation was made by shifting the position of a small steel leaf spring, shown in Figure 4.4. The disadvantage to having a rigidly mounted sample and mirror is that the position of the sample holder within the deposition chamber must be highly reproducible. This was achieved experimentally by a seat for the sample holder which was rigidly mounted within the deposition chamber.

The substrates used in these experiments were 8×10 mm silicon \{100\} wafers with a thick oxide (SiO\(_x\)) layer. This oxide layer is amorphous and prevents ordered growth effects due to the crystalline substrates. The sample holder was placed in the deposition chamber, which was pumped by a BOC Edwards EXT250M 250 l s\(^{-1}\) turbo-molecular pump and maintained a pressure of 4×10\(^{-6}\) Torr during sample exposure. The Fe atom beam had a diameter of 7 mm at the substrate surface and its position relative to the mirror is shown in Figure 4.5. The area that line structures may be created

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Figure 4.4: \textit{Left:} The sample holder used for direct deposition experiments. The sample was clamped to a macor isolator which was mounted to the stainless steel sample holder. The use of macor was retained from previous experiments [57] where electrical isolation was required. \textit{Right:} The cross-section shows the leaf spring used to position the mirror.
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Figure 4.5: *Left:* Alignment of the sample and mirror relative to the Fe atomic beam. The position of the standing wave light mask and area of expected line structures is also shown. *Right:* Alignment of the standing wave mask relative to the edge of the sample showing the 50% intensity clipping used in these experiments.

due to the standing wave light mask is also shown.

4.2.2 Laser system

A schematic of the laser system is shown in Figure 4.6. To focus iron atoms using a standing wave mask requires light tuned to the $^5D_4 \rightarrow ^5F_5$ transition at 372 nm. This was achieved by frequency doubling the output of a titanium-sapphire (Ti:S) laser with a lithium triborate (LBO) crystal in a ring cavity. The Ti:S laser was then frequency locked using polarisation spectroscopy \cite{139}. An acousto-optic modulator (AOM) was used to introduce a detuning of $\Delta = 150$ MHz to the standing wave mask.

Frequency doubling

The *Coherent* 899 Ti:S laser was pumped by a *Coherent* Verdi V-18 solid state laser operating at 532 nm. The Ti:S laser outputs approximately 1 W of 744 nm light. This was then coupled into the folded ring cavity shown in Figure 4.6. The LBO crystal was placed between two 99.9% reflective spherical ($R = 7.5$ cm) mirrors which focus the 744 nm light to a beam waist of $\sim 25 \mu m$ within the crystal. The facets of the crystal are cut at Brewster’s angle to limit reflection losses and increase the finesse of the cavity. The mirrors are transparent for UV light and the 372 nm radiation generated in the crystal leaves the cavity through mirror M4.

Mirror M2 was mounted on a piezo transducer and enabled the cavity
Figure 4.6: Depiction of the laser system used to generate the 372 nm light used in Fe deposition experiments. A Verdi V-18 laser pumps a Ti:S laser outputting \(~1\) W at 744 nm. This was coupled to a frequency doubling cavity containing an LBO crystal. The 372 nm output from the doubling cavity was stabilised by analysing the light emitted from mirror M1 using a \(\lambda/4\) wave plate, a polarising beam splitter (PBS), and photodetectors 1 and 2 (PD1 and PD2). Feedback was then provided to the piezo mounted mirror M2. The laser then passed through an acousto-optic modulator (AOM) where a majority of the intensity was unaffected and coupled into an optic fiber to be transported to the vacuum apparatus. The small proportion of the beam deflected into the first order by the AOM was used to lock the laser system to the Fe atomic resonance using polarisation spectroscopy in a hollow cathode discharge cell (HCD).
4.2. EXPERIMENTAL SETUP

to maintain resonance with the incident 744 nm light. The locking scheme makes use of the birefringence of the LBO crystal and the rotation of polarisation within the cavity [140]. A linearly polarised beam travelling through the LBO crystal becomes elliptically polarised due to a small polarisation mismatch with the crystal orientation. By analysing the reflection from mirror M1 with a λ/4 wave plate and a polarising beam splitter (PBS), a dispersive signal which can be used for locking is obtained. Locking using this method resulted in output power fluctuations from the cavity of less than 5%. Typical power output from the doubling cavity was 220 mW of 372 nm light. This light then passed through an AOM where a small proportion, ∼20 mW, was deflected into the first order diffracted beam with a detuning of -150 MHz relative to the unaffected laser frequency. This beam was used to frequency lock the laser system to the \(^5D_4 \rightarrow ^5F_5\) transition in iron using polarisation spectroscopy. The unaffected beam passing through the AOM, which was detuned + 150 MHz from the resonant atomic frequency due to the offset used for the locking system, was then used for the standing wave light mask.

Polarisation spectroscopy

This technique is similar to the saturated absorption scheme described in Section 2.2.3 however, the two counterpropagating laser beams within the reference cell have different polarisations. The iron atoms in a hollow cathode discharge cell, depicted in Figure 4.7, are pumped into either the highest or lowest magnetic sublevel using a circularly polarised pump beam. A linearly polarised probe beam passing through the cell then experiences a slight rotation of its polarisation due to the nonuniform distribution of the magnetic sublevels of the atoms. The emerging probe beam is slightly elliptically polarised which is then analysed with a polarising beam splitter. A dispersive signal, as shown in Figure 4.8, can be obtained from the two components separated by the beam splitter and used to lock the laser system. The basic theory of polarisation spectroscopy can be found in [141].

The hollow cathode discharge cell achieves a partial pressure of iron atoms of \(\sim 4 \times 10^{-5}\) Torr through argon ion sputtering. The discharge is struck between two stainless steel anode rings and a central hollow core iron cathode in the presence of 1 - 2 Torr of argon. The iron cathode is mounted to a water cooled copper heat sink. The cell was configured in the
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Figure 4.7: Depiction of the hollow cathode discharge cell used for locking the laser frequency to the Fe atomic resonance. The discharge is struck between the two stainless steel anode rings and the hollow core Fe anode. The cathode is sealed to two pyrex half tubes using viton o-rings.

Figure 4.8: The dispersive locking signal obtained by polarisation spectroscopy and varying the laser frequency. The signals from two other isotopes of Fe are also visible.
4.2. EXPERIMENTAL SETUP

arrangement shown in Figure 4.6 for polarisation spectroscopy.

To improve the signal to noise ratio and isolate the polarisation spectroscopy signal the pump beam was pulsed at 2 kHz using a mechanical chopper. Using this method the laser could be locked to the atomic resonance to within 0.2 MHz for hours [139].

Standing wave light mask

As the atoms pass through a standing wave they experience a force due to the interaction between the induced atomic dipole moment and the amplitude gradient of the standing wave. Recalling from Section 2.3 Equation 2.15 there are two components to the force on an atom in a light field. The general form of the electric field of a standing wave can be described by

$$\vec{E}(\vec{r}, t) = E(\vec{r}) \cos(\omega t).$$

(4.1)

For the standing wave system there is no phase dependence of the electric field and the force of Equation 2.15 is only dependent on the dipole force component. The dipole force is then given by

$$\vec{F}_{\text{dip}} = \frac{-\hbar \Delta L \nabla \Omega^2}{4 \Delta^2 L + \Gamma^2 + 2\Omega^2}.$$  

(4.2)

The dipole force may also be derived from the potential of the light field,

$$U = \frac{\hbar \Delta L}{2} \ln \left[ 1 + \frac{s_0}{1 + (2\Delta L/\Gamma)^2} \right],$$

(4.3)

where $s_0$ is dependent on the spatially varying intensity of the standing wave $I(\vec{r})$. The dipole force can then be written as

$$\vec{F}_{\text{dip}} = -\nabla U = -\nabla \frac{\hbar \Delta L}{2} \ln \left[ 1 + \frac{I(\vec{r})}{I_s} \frac{1}{1 + (2\Delta L/\Gamma)^2} \right].$$

(4.4)

From this equation it can be seen that the direction of the dipole force is dependent on the sign of the light detuning $\Delta L$. For positive detuning, or blue shifted light frequency, the force on the atoms is towards the intensity minima and for negative detuning, red shifted light frequency, the atoms are forced into the regions of high intensity. The dipole force vanishes for light that is tuned to the resonant frequency of the atoms.

While the dipole force does not saturate for increasing light intensity the
shape of the potentials for red and blue detuned light varies significantly, as can be seen in Figure 4.9. The potential gradient near the nodes of the standing wave increases for both red and blue detuned light, however, due to the opposite signs of the potentials the atoms are more confined in the minima of a blue detuned standing wave, as indicated by the circles in Figure 4.9. This results in narrower features of the deposited structure. For this reason blue detuned standing wave light masks are more appropriate for use in atom lithography.

After passing through the AOM, the blue detuned beam was coupled into a polarisation maintaining fiber. The experimental setup of the standing wave mask after out-coupling from the optic fiber is shown in Figure 4.10. The 372 nm light was focussed onto a mirror in the vacuum to create a standing wave with a waist size between 60 and 150 µm. The beam waist was determined prior to experimentation by reflecting all laser light into the photodetector shown in Figure 4.10 and using a translatable knife edge (not shown in Figure 4.10). The power of the laser beam entering the vacuum chamber was between 20 and 55 mW. The laser beam waist position was adjusted in the x-direction, defined in Figure 4.5 using the translatable lens in Figure 4.10. The retroreflected beam was aligned to the incoming beam to less than 1 mrad deviation using the translatable mirror. The mirror was then positioned such that the power in the retroreflected beam was
4.3. CHARACTERISATION OF MAGNETIC SURFACES

Figure 4.10: A depiction of the optics setup used to create the standing wave light mask. The 372 nm light was out-coupled from the optic fiber and expanded to the required size by the appropriate positioning of a collimating lens ($f = 35$ cm). The number of mirrors was chosen to increase the degrees of freedom and enable the accurate positioning of the laser beam on the translatable mirror. The translatable lens ($f = 30$ cm) enables adjustment of the focal point so that it coincides with the mirror on the sample mount within the deposition chamber. The intensity of the reflected beam was monitored using the photodetector (PD).

50% of the maximum power reflected off the mirror, as shown in Figure 4.5. The power in the reflected beam was measured using the glass plate and photodetector in Figure 4.10.

4.3 Characterisation of magnetic surfaces

In addition to imaging surfaces using AFM, methods for investigating the constitution and magnetic properties of the deposited film were also utilised. The AFM imaging was conducted under ambient conditions on a Digital Instruments Dimension 3100 AFM in tapping mode, using Si cantilevers with a radius of curvature less than 10 nm. As the deposited structures had
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typical height to width ratios below $10^{-1}$, tip artefacts were disregarded. The spatial composition of co-deposited Fe-Ni films were investigated using a technique called energy dispersive X-ray spectroscopy (EDX).

4.3.1 Energy dispersive X-ray spectroscopy

EDX is an analytical technique used for the compositional characterisation of a sample. The technique is usually performed on a scanning electron microscope (SEM) and relies on the emission of characteristic X-rays from the area of the substrate exposed to the focussed electron beam. The energy spectrum emitted is analysed and the peaks can be assigned to specific emission lines of individual elements. In principle all elements from atomic number 4 to 92 can be detected, though some instruments may not detect the lighter elements. Many of the elements also have overlapping spectral lines which cannot be resolved and may limit the accuracy of some measurements. The measurements made in this investigation do not suffer these limitations and were carried out using an FEI Sirion SEM. To determine the concentrations of the elements within the sample, the measured line intensities for each element must be compared to those in calibration standards of known composition.

The electron beam in the SEM bombards the sample and may eject an electron from an inner shell orbital. The vacancy may then be filled by an outer shell electron and eject an X-ray in the process. The spectral lines are denoted by a capital letter (K, L, or M) indicating the orbital containing the electron vacancy (where K is the innermost shell). A greek letter is used to indicate the group that the spectral line belongs to in decreasing order of importance ($\alpha$, $\beta$, etc.). Finally, for multiple lines within a group, numbers are used to indicate their intensity. The most intense K spectral line is then $K\alpha_1$, if the second line is unresolvable from the first the combined spectral line is simply designated $K\alpha$.

Bombardment of the sample by electrons not only produces the characteristic spectral lines but also a continuous X-ray spectrum spanning all energies from zero to the incident electron energy. This is due to interactions between the incident electrons and the atomic nuclei of the sample. These emissions contribute to the background counts observed in the measured spectra.
**4.3. CHARACTERISATION OF MAGNETIC SURFACES**

4.3.2 Magneto-optic Kerr effect

The magnetic properties of the deposited films were analysed using the magneto-optic Kerr effect (MOKE) [142, 143]. When light is reflected from a magnetic surface the polarisation state and reflected intensity can be influenced by the samples magnetisation. The polarisation state of the incident light can be affected in two ways. Firstly, the plane of a linearly polarised beam may be rotated by an angle $\theta_k$, the Kerr rotation angle. The direction of rotation is determined by the orientation of the samples magnetisation. The plane polarised light may also become slightly elliptical which is quantified by the Kerr ellipticity $\varepsilon_k$. By applying an external magnetic field to the sample and detecting the changes in the polarisation of the reflected light a magnetic hysteresis loop may be obtained and the coercivity of the material determined.

The magneto-optic Kerr effect may be divided into three experimental arrangements; the polar, longitudinal, and transversal MOKE. The differences arise from the direction of the samples magnetisation relative to the plane of incidence and the sample surface, as shown in Figure 4.11. In the polar MOKE, the magnetisation is normal to the sample surface and parallel to the plane of incidence. This type of MOKE is frequently studied at near-normal angles of incidence as the sample surface must be accessed through a hole in one pole of the external magnet. The polar arrangement is the only case where the MOKE may be observed at normal incidence.

The longitudinal MOKE is observed when the magnetisation is parallel to both the plane of the surface and the plane of incidence. This is the
most commonly used experimental arrangement and measures the in plane magnetisation of the sample. The arrangement is schematically shown in Figure 4.12.

The intensity of light incident on the detector in both the polar and longitudinal orientations can be written as

\[ I(t) = I_0 [1 + 2 \theta_k \cos(A_0 \omega t) - 2 \varepsilon_k \sin(A_0 \omega t)], \]  

where \( I_0 \) is the average intensity of the light and \( A_0 \omega t \) is the time-dependent retardation of the photoelastic modulator. Continuous measurement of the light intensity enables the components \( \theta_k \) and \( \varepsilon_k \) to be determined which are directly proportional to the magnetisation of the sample.

The final orientation, the transversal MOKE, has the magnetisation perpendicular to the plane of incidence but still parallel to the plane of the sample. In this configuration instead of measuring the polarisation of light after reflection, the reflectivity of the surface is measured. The change in reflectivity is proportional to the magnetisation and is only observed in light polarised parallel to the plane of incidence. Whether the polarisation amplitude increases or decreases is dependent on the direction of magnetisation.

A microscope utilising the MOKE can be constructed and enables the imaging of magnetic domains [144]. An evico magnetics magneto-optical Kerr microscope was utilised to map the spatial variations in the magnetic coercivity of samples, as shown in Figure 4.22. Such a microscope can also
be used to investigate the dynamics of domain wall motion.

4.4 Results and discussion

4.4.1 Direct deposition of iron without laser cooling

The atomic beams used in atom lithography experiments are usually transversely laser cooled to increase the beam brightness and minimise the transverse velocity spread. One method for increasing the beam brightness has been shown previously in Section 2.5.2. However, for atomic species that do not have a closed optical transition from the ground state, more complex laser cooling schemes are required. The minimisation of the transverse velocity spread was previously thought to be an essential requirement for atom lithography [48], though was shown not to be necessary by Smeets et al. [145].

The local transverse velocity spread of an atomic beam incident on a single atom lens in the standing wave light mask needs to be small to enable sharp imaging. This can be achieved in a scheme termed geometric collimation, depicted in Figure 4.13. An atomic beam emerging from a small source opening with a large source to sample distance has a small local transverse velocity spread over the region of a single atom lens at the sample. A laser cooled atomic beam globally collimates the atomic beam, while geometric collimation aligns the atomic beam perfectly orthogonal to the light mask at a single point, but not over the entire standing wave. The beam at each lens element in the standing wave may be considered as collimated but incident at a small angle, \( \phi \), defined by the local average transverse velocity, \( \langle v_{\text{trans}} \rangle \).

Lithographic structure formation with Fe has been experimentally observed over areas of up to 400 \( \mu \text{m} \times 6 \text{ mm} \) on a single substrate. An example of an AFM image of Fe line structures with 186 nm periodicity and a height of \( \sim 6 \text{ nm} \) is shown in Figure 4.14. The experimental parameters of the Fe source are as outlined in Table 4.2. Since line structures have been observed over a region up to 6 mm long in the \( x \) direction (perpendicular to the lines), \( \phi \) varies by \( \sim 8 \text{ mrad} \) over the deposition. Similarly, \( \langle v_{\text{trans}} \rangle \) was calculated to vary by \( \sim 8 \text{ m} \text{s}^{-1} \), more than an order of magnitude larger than the RMS transverse velocity spread, \( \sigma_{v_{\text{trans}}} \). The effect that this has on the geometry of the deposited lines is the topic under investigation.

Simulation of the atomic trajectories of Fe through the focussing light
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Figure 4.13: Overview of atom lithography using geometric collimation, creating a locally collimated atomic beam. D is the diameter of the open aperture of the atom source, L is the atom source to sample distance, and φ is the local angle of incidence of the atom beam with respect to the sample.

Figure 4.14: A $2 \times 2 \, \mu m^2$ AFM image (left) and the average line cross-section (right) of the Fe lines deposited without laser cooling.
field have been carried out using a dressed state, Monte Carlo method. Using typical experimental parameters for the laser detuning, power, and beam width, the results showed that the trajectories do not have a common focal point. This is primarily due to the large spread in the atomic longitudinal velocities, 357 m s\(^{-1}\) RMS. Broadening of the atomic distributions at the extremities of the laser beam width was also observed due to the lower light intensity. The final effect observed through simulation was the asymmetry of the deposited lines and their offset relative to the light intensity minima in the areas away from the centre of the Fe atom beam. This is due to the increase in the local average transverse velocity. Fitting Gaussian functions to the simulated line profiles allow the peak height and full width half maximum (FWHM) of the simulated atomic distributions to be compared to the experimental results.

The profiles of the experimentally deposited samples were measured using an AFM. By measuring along the \(y\) direction, defined in Figure 4.14, the influence of the local light intensity could be observed, where \(y = 0\) is the position of maximum light intensity. Measuring along the \(x\) direction gives insight into the effect of the transverse velocity of the Fe atom beam, where \(x = 0\) corresponds to \(\langle v_{\text{trans}} \rangle = 0\). Measurements were performed on areas 250 nm along the lines and 2 \(\mu\)m perpendicular to the lines, such that 11 lines were visible. The image was averaged over the \(y\) direction and each line in the resulting cross section was fitted with a Gaussian giving an average and spread value for the peak height, \(h\), and FWHM, \(w_h\).

The experimental and simulated results for \(h\) and \(w_h\) as a function of \(y\) position of the deposited Fe lines is shown in Figure 4.15. The profiles were taken at \(x = 0\) (therefore \(\langle v_{\text{trans}} \rangle = 0\)) and should represent the highest and narrowest of the lines deposited. Three settings of incident laser power, \(P\), and beam waist, \(w\), are shown. The variation observed in the incident laser power is due to the maximum power available from the laser on the day of experimentation, however, this has a limited effect on the results as all settings used are well above the saturation intensity of the transition in iron. In Figure 4.16 the experimental and simulated results are shown as a function of \(x\) position, which is proportional to \(\langle v_{\text{trans}} \rangle\). The profiles of the deposited lines shown are for the region of maximum intensity of the light field, \(y = 0\) mm.

From Figure 4.15 it is clear that the formation of line structures is in-
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Figure 4.15: Simulated (red dots) and experimentally measured (black squares) $h$ (left) and $w_h$ (right) of deposited Fe lines at different $y$ positions and laser field settings. From top to bottom: $w = 60 \ \mu\text{m}$ and $P = 50 \ \text{mW}; \ w = 90 \ \mu\text{m}$ and $P = 20 \ \text{mW}; \ w = 150 \ \mu\text{m}$ and $P = 55 \ \text{mW}$. The error bars observed for the simulated results were determined from the fitting routine and indicate difficulties in fitting to $w_h$ at the extremities of the structured deposition. A normalised laser intensity profile (grey area) is shown in the left-hand images to indicate the expected line profiles.
fluenced by the local intensity of the light field. The simulated results show a bell-shaped dependence of the height of the lines on their lateral, $y$, position within the standing wave. The profile is more broad and flat topped than the lasers intensity profile. This indicates that at high light intensities the atomic focussing is less sensitive to changes in the intensity, similar to a saturation of the focussing efficiency. This is reiterated in the simulated values of $w_h$, which remain reasonably constant over the region of $|y| \lesssim w$. For larger values of $y$, corresponding to lower light intensities, $w_h$ increases. The experimental results for $h$ correlate well with the simulations at the extremities of the light field but differ greatly for $|y| \lesssim w$. At $y = 0$, the observed height of the lines is two to three times smaller than the expected simulated values. The experimentally observed values of $w_h$ seem to be independent of the $y$ position. The values fluctuate between 80 and 120 nm, which correspond to the values at the extreme $y$ positions in the simulations. Simulated values of $w_h$ around $y = 0$ are also significantly smaller, down to 35 nm.

The influence of the local $\langle v_{\text{trans}} \rangle$ on structure formation is shown in Figure 4.16. Experimentally, structures were observed up to large values of $x$, $>3$ mm, corresponding to $\langle v_{\text{trans}} \rangle = 4 \text{ m s}^{-1}$. This is more than an order of magnitude larger than $\sigma_{v_{\text{trans}}}$ and nearly two orders of magnitude larger than the transverse velocity associated with Doppler cooled beams, where $\sigma_{v_{\text{trans}}} = v_d = 0.1 \text{ m s}^{-1}$. At the values of $\langle v_{\text{trans}} \rangle$ observed, atoms in the atomic beam may pass through a region of maximum intensity in the optical potential and are therefore no longer restricted to interacting with a single atom lens.

The simulation results of Figure 4.16 indicate that the highest and narrowest lines are deposited at $x = 0$ mm, where $\langle v_{\text{trans}} \rangle = 0 \text{ m s}^{-1}$. They also show that the line height decreases rapidly for non-zero $\langle v_{\text{trans}} \rangle$. Experimental results show a maximum for $h$ at $x = 0$ mm but the peak is significantly flatter, not displaying the strong $\langle v_{\text{trans}} \rangle$ dependence expected by the simulated results. The measured line widths, $w_h$, are again significantly larger than the simulated values around $x = 0$ mm but comparable at the values at the extremities of the $x$ position. The experimental widths are similar to those observed in Figure 4.15 all are between 80 and 120 nm. The results for both the $x$ and $y$ position dependence of the line structure formation indicate that the simulations predict broad, low structures co-
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Figure 4.16: Simulated (red dots) and experimentally measured (black squares) $h$ (left) and $w_h$ (right) of deposited Fe lines at different $x$ positions (which is $\propto \langle v_{\text{trans}} \rangle$) and laser field settings. From top to bottom; $w = 60 \, \mu\text{m}$ and $P = 50 \, \text{mW}$; $w = 90 \, \mu\text{m}$ and $P = 20 \, \text{mW}$; $w = 150 \, \mu\text{m}$ and $P = 55 \, \text{mW}$. The asymmetric bounds of the experimentally measured data in the top and middle graphs is due to a slight positional offset of the sample mounted mirror with respect to the experimental axis.
4.4. Results and Discussion

Figure 4.17: Measured (black squares) and simulated (red circles) product of $h$ and $w_h$ for a light field of $w = 90 \, \mu m$ and $P = 20 \, mW$ as a function of $y$ position (left) and $x$ position (right).

directly, but fail in the case of optimal focusing. This indicates the presence of an additional broadening mechanism that was not accounted for in the simulations.

This can be confirmed by a comparison of the simulated and experimental values for the product of $h$ and $w_h$, a measure of the total amount of focused material. In Figure 4.17 the product for one light field setting is shown. Results are similar for the other settings used in Figures 4.15 and 4.16. The simulated product of $h$ and $w_h$ for the deposited lines is comparable to the experimental results, with a discrepancy on the order of 20%. This may be accounted for in the amount of material which forms the pedestal upon which the Fe lines are deposited.

The broadening of deposited structures in atom lithography experiments has been observed previously [147, 148]. Two explanations have been proposed: the distortion of the light field by the presence of the substrate and the mobility of deposited atoms on the surface.

The effect that the substrate has on the light field has been modelled by Anderson et al. [147]. Assuming that the surface and mirror are fully reflecting and that the incoming Gaussian light field was cut off at the maximum intensity by the substrate, as shown in Figure 4.15, the light field near the centre of the substrate can be calculated. A dark zone extending approximately 10 $\mu m$ immediately above the surface was found and is due to the interaction between the sample and the light field. The maximum
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Intensity of the light field is nearly two times higher at about 20 µm from the surface. At distances farther from the surface the intensity pattern returns to the expected Gaussian pattern. The approximation that the substrate and the mirror are fully reflective is an overly simplified case. During exposure to the atomic beam, Fe coats the SiOx substrate and modifies the reflection coefficients and associated phase terms.

The dark area above the surface allows defocusing of the atomic beam to occur. From the simulations it was found that $\sigma_{v_{trans}}$ in the plane of the sample was 0.53 m s$^{-1}$ for the strongest focused beams. This leads to a maximal broadening of $w_h$ by $\sim$11 nm. Since the maximum intensity is slightly higher, the focusing strength may be altered and lead to stronger defocusing. However, the divergence of the atomic beam over the dark zone above the substrate cannot fully account for the broadening of the structures from 35 nm in simulations to over 80 nm for those observed experimentally.

The mobility of deposited atoms on a surface and the resulting effects on deposited structures have been an area of interest for atom lithography. Ballistic deposition models [149], terrace and step-edge diffusion [150], and polluted adatom limited diffusion [148] have been proposed. The latter is consistent with the experimental data of Anderson et al. [147], but there is no complete understanding of atoms mobility on a substrate. Very clear effects have been observed using this experimental apparatus for structures deposited on SiOx and Si $\langle111\rangle$ substrates. Previously reported Fe structures on Si $\langle111\rangle$ with a native oxide layer had a typical $w_h$ of 50 nm [145].

A reference experiment was performed to determine the extent of substrate dependent diffusion of the deposited atoms [146]. Fe was deposited through 100 nm thick silicon nitride physical masks with line patterns 80±5 nm wide and several hundred µm long. These masks enable the deposition of well defined, free-standing (i.e. no pedestal layer) structures onto Si $\langle111\rangle$ or SiOx substrates. 15 to 20 nm thick layers of Fe were deposited through these masks and their geometry measured with AFM. It was found that the substrate had a significant influence on the shape of the structures deposited. This has been previously observed for other materials, e.g. Tun et al. [151] observed very different structure formation of gold deposited on various substrates through physical masks.

In this experiment the deposited structures were compared to the expected deposition profile based on the geometry of the mask, the divergence
4.4. RESULTS AND DISCUSSION

Figure 4.18: Cross sections of nanolines created by Fe deposition through a physical mask on SiOx (left) and Si ⟨111⟩ (right). The dotted red curve shows the geometrically expected structures from the 80 nm wide mask openings. The measured AFM results are indicated by the solid black curves while the dashed blue curves are the results of a model convoluting the geometric expectation with a Gaussian of σ = 23 nm (left) and σ = 12 nm (right).

of the atom beam, and the mask-substrate distance. The substrate influence was modelled as a convolution of the expected profile with a Gaussian function, of which the standard deviation σ gives a measure of the substrate dependent diffusion. The remarkable agreement between the measured profiles and those predicted by the convolution model can be observed in Figure 4.18. The values of σ were determined by to be 12 nm for Fe on Si ⟨111⟩ and 23 nm for SiOx. However, the value of σ is not easily translatable into an increase in the width of structures in atom lithography. It should be noted that the FWHM of the expected deposition profile, convolution model, and both measurements in Figure 4.18 are approximately equal.

The surface diffusion and broadening effects in experiments utilising a standing wave light mask, which create Fe nanostructures on an Fe pedestal, will be different from those of the freestanding structures shown in Figure 4.18. However, these results indicate that the diffusion effects of Fe on SiOx and Si ⟨111⟩ have a range of tens of nm. It is also observed that the value of σ for SiOx is approximately twice the value for Si ⟨111⟩. This is reflected in the differences observed between the values of \(w_h\) for atom lithographic lines on Si ⟨111⟩ (\(w_h \gtrsim 50\) nm), reported previously [145], and those deposited on SiOx (\(w_h \gtrsim 80\) nm), observed in Figures 4.15 and 4.16.
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It is likely that atom diffusion and broadening affect narrow and high structures more than broad structures, as observed in Figures 4.15 and 4.16. However, these results do not indicate how diffusion and broadening affect structures formed by varying the flux distributions. It’s observed that substrate dependent surface diffusion effects can contribute significantly and may account for the observed differences between the $w_h$ of the Monte Carlo simulations and the experimentally deposited lines.

The simulated asymmetry of lines deposited in the regions displaced from the centre of the Fe atomic beam was also observed in the experimental results. The asymmetry was quantified by fitting a skewed Gaussian distribution to the AFM measured line profiles. The trend of increasing asymmetry at larger $x$ positions, and hence $\langle v_{\text{trans}} \rangle$, was observed for values of $\langle v_{\text{trans}} \rangle$ from -1.25 to 1.25 m s$^{-1}$. However, the broadening of the deposited structures reduced the magnitude of the observed asymmetry compared to the simulated results.

4.4.2 Co-deposition of iron and nickel

The co-deposition of two ferromagnetic elements with a periodic variation in the spatial concentrations is potentially interesting for spintronics applications [152]. The periodic variations in film thickness achieved using atom lithography would induce periodicity in the spin wave dispersion relation due to its dependence on the magnetic film thickness [153]. The spin structure at grain boundaries is also of great importance. The experiments detailed in this section represent the initial investigations into the properties of a co-deposited Fe-Ni thin film.

One property of the film which is of critical importance is the spatial composition profile of the deposited film. This characterisation has been performed using EDX, described in Section 4.3.1. As an initial investigation on the applicability of this technique to the thin films of interest, a series of Fe-Ni films were deposited on an SU-8, structured polymer substrate. SU-8 is an epoxy based photoresist which can be coated onto a silicon wafer and hardened using a UV laser. In this way periodic line patterns can be established in the SU-8 by exposing it to an optical standing wave. An AFM image of the structured SU-8 substrate can be seen in Figure 4.19. Using these substrates enables a large quantity of co-deposited samples to be created more easily than if a standing wave light mask was used to modulate
4.4. RESULTS AND DISCUSSION

Figure 4.19: A 5 × 5 µm² AFM image (left) and the cross-section (right) of the line structures developed in SU-8 photoresist.

In contrast to samples patterned using a standing wave light mask, exposing the structured SU-8 substrate to the Fe and Ni atomic beams results in a layer where the flux of Ni is modulated. The modulation of the Ni beam is provided by ‘shadow’ masking. Since the Ni beam is incident at 45° relative to the sample surface and the line structures are perpendicular to the plane of incidence, the peak of each line will create a ‘shadow’ on its far side and no Ni will be deposited in those regions. The variations in the Fe-Ni concentrations is then simply a function of the line profile. Higher, narrower lines will yield larger variations in the concentration while the average composition over the entire sample is determined by the incident flux of both Fe and Ni.

The sample shown in Figure 4.19 was simultaneously exposed to beams of Fe and Ni with approximately equal flux. This was then capped with a 5 nm Ag layer to limit the oxidation of Fe. The sample was then analysed using EDX in an attempt to determine the spatial variations in the layer composition. However, initial experiments have shown insufficient signal from the Fe and Ni lines to obtain accurate composition information, as shown in Figure 4.20. The carbon and oxygen peaks present are due to the underlying SU-8 substrate. Layer thickness will have a substantial influence on the strength of the signal obtained and may be used to improve the re-
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Figure 4.20: Energy spectrum of the co-deposited Fe-Ni layer on an SU-8 substrate. The signal strength of the Fe and Ni lines was insufficient for compositional analysis. The carbon and oxygen peaks are present due to the SU-8 photoresist which is grown on a silicon wafer.

The magnetic properties of a co-deposited Fe-Ni film have been compared to those of a purely Fe film using the longitudinal MOKE arrangement of Figure 4.12. The magnetisation of the films was measured in the plane of the sample with the applied magnetic field perpendicular and parallel to the deposited lines. Hysteresis loops for a ~10 nm thick Fe layer with 3 nm high lines are shown in Figure 4.21. The measurements shown were taken at the centre of the region with line structures and are averaged over many lines due to the diameter of the laser spot used in the MOKE setup (~200 µm). The coercive field measured with the field applied perpendicular to the lines was 19±1 mT, and when applied parallel to the lines was 16±1 mT. The coercive field was also measured at an unstructured region of the sample and resulted in a value of 13±1 mT regardless of the orientation of the applied magnetic field. This indicates that the magnetic anisotropy observed is due to the deposited lines and not a crystalline growth of the deposited film. These values are slightly lower than previously measured values \[154\] for samples deposited through a physical mask. The discrepancy is most likely due to
the height of the deposited line being substantially smaller.

The measurements were repeated for a co-deposited sample of Fe-Ni with an average concentration of 60(Fe):40 (Ni). The deposited film was \( \sim 30\text{nm} \) thick with 3 nm high lines. The coercive field was measured as a function of position on the sample with the applied field again perpendicular and parallel to the deposited lines. The resulting ‘coercivity maps’ are shown in Figure 4.22. From the figure it is clear that there is once again an anisotropy present due to the deposited line. The coercive field perpendicular to the lines was 30±1 mT, and parallel to the lines 26±1 mT. The magnitude of the change is similar to the Fe film however the absolute value of the coercive field is significantly larger. This will likely be due to the combined effect induced by the difference in magnetic moments of Fe and the Fe-Ni alloy and the significant differences in the average layer thicknesses.

It was also observed that there was a magnetic anisotropy present in the regions outside of the deposited lines. In the regions without structuring the coercive field measured 23±1 mT perpendicular to the line structures and 28±1 mT parallel to them. This may be an effect of the Si substrate, although the oxide layer should be sufficiently thick to allow the polycrystalline growth of the layer, as was observed for the Fe deposition previously. An alternative explanation may be that the incident angle of the Ni atomic beam influences the growth of the layer and induces an anisotropy.

Figure 4.21: Magnetic hysteresis loops for Fe deposited through a standing wave light mask. MOKE measurements were taken with the applied magnetic field a) perpendicular and b) parallel to the deposited lines. The coercive field is indicated by the blue lines.
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Figure 4.22: Coercive field over the region of co-deposited Fe-Ni lines (recall lines are periodic in the x-direction). The coercive field has been mapped for the applied magnetic field a) perpendicular and b) parallel to the co-deposited lines. Note: The distances on the axes are not relative to the maximum line profiles, i.e. $x=0$ is not where $\langle v_{\text{trans}} \rangle = 0$. 

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Figure 4.22: Coercive field over the region of co-deposited Fe-Ni lines (recall lines are periodic in the x-direction). The coercive field has been mapped for the applied magnetic field a) perpendicular and b) parallel to the co-deposited lines. Note: The distances on the axes are not relative to the maximum line profiles, i.e. $x=0$ is not where $\langle v_{\text{trans}} \rangle = 0$. 

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4.5. CONCLUSIONS

The final characteristic observable in Figure 4.22 is that the coercive field was lower within the structured region when the magnetic field was applied parallel to the lines. The nucleation of the magnetic domains within the line structures may be due to the variations in the Fe-Ni concentrations, specifically the Invar effect [155]. Invar is a particular alloy of Fe and Ni (Fe$_{64}$Ni$_{36}$) and has the unique property of an extremely low coefficient of thermal expansion. It also has the anomalous behaviour of lowering the magnetic moment per atom and hence the coercivity of the material. This lowering of the coercivity may account for the effect observed in the co-deposited samples.

4.5. Conclusions

Atom lithography without laser cooling of the atomic beam has been achieved and patterning was observed for atoms with a local average transverse velocity of up to a few m s$^{-1}$. The local $\langle v_{\text{trans}} \rangle$, which corresponds to an offset angle, $\phi$, of the atomic beam with respect to the standing wave light mask, influences the height, width, and symmetry of the deposited structures. These effects have been observed in experimentally deposited Fe lines. However, due to a substrate dependent diffusion mechanism, the observed structures were broader and less pronounced than in simulated results. The scattering and interference of the standing wave light mask near the substrate also limits the resolution of the deposited structures. Simulated results yielded line structures with FWHM’s $\geq 35$ nm, while experimentally deposited lines on SiOx substrates were limited to structures of $w_h > 80$ nm. Previous experiments on Si (111) substrates resulted in smaller structures of $w_h \geq 50$ nm [145]. The appropriate selection of a substrate could therefore produce narrower structures.

Removing the requirement of laser cooling enables technologically interesting elements that do not have closed optical transitions, such as Ga, In, and Si [91], to be used in atom lithography experiments. Atom lithography without laser cooling could be further improved by altering the geometry of the experiment and the atomic beam source to reduce $\sigma_{v_{\text{trans}}}$ and $\phi$.

An initial characterisation of the magnetic properties of co-deposited Fe-Ni structures has indicated there are interesting effects that should be investigated further. Primarily, the effect the variations in Fe-Ni concentrat-
tions have on the coercivity of the deposited structures. A reduction of the coercive field in regions with line structures, when the magnetic field is applied parallel to the lines, has been attributed to the nucleation of magnetic domains by regions where the Fe-Ni alloy possesses a lower magnetic moment per atom. In order to further investigate the effect of these alloys, the spatial composition of the Fe-Ni layer would need to be determined by repeating the EDX experiments or using an alternative technique. Substrates containing the concentrations of Fe-Ni near that of Invar should produce some anomalous effects. The source of the magnetic anisotropy induced by the incident angle of the Ni atomic beam should also be considered. For a complete characterisation of the co-deposition system the effects of line height, width, and height to pedestal height ratio would also need to be examined.

Currently only the in-plane magnetisation has been studied using the magneto-optic Kerr effect. It has been observed in simulations that one likely orientation of the magnetisation follows the profile of the deposited lines [156] and as such the extent of the out-of-plane component of magnetisation should also be determined. MOKE experiments average the magnetic properties over a relatively large area on the sample surface, to determine the magnetic properties of individual deposited lines the application of magnetic force microscopy should be considered. Obtaining such results would provide a clearer indication of the domain structure and switching behaviour of these samples.
Summary

This thesis has detailed the continuing improvement of two experiments utilising the alternative techniques of atom lithography.

Resist based atom lithography

The metastable neon (Ne\(^*\)) atomic beam utilised in the resist based atom lithography experiment has been significant enhanced. Undesirable energetic constituents produced by the atomic beam source may limit the ultimate resolution of the lithographic patterns that can be produced. Therefore a UV free beam of Ne\(^*\) atoms that are purely in the \(3^3\text{P}_2\) state has been developed.

The Ne\(^*\) beam was generated by a liquid nitrogen cooled, DC discharge source. It was then cooled in the transverse direction using a two dimensional optical collimator, decreasing the atomic beam divergence to 7\(\pm\)3 mrad. The beam was then slowed in the longitudinal direction using a novel dual optical beam Zeeman slower that significantly increases the transmitted slowed atom flux compared to previous experiments. Atoms with longitudinal velocities below \(\sim 680\) m s\(^{-1}\) were slowed to a velocity of 80 m s\(^{-1}\) with an efficiency of approximately 58%. The slowed atoms were re-collimated by a two dimensional optical molasses at the end of the Zeeman slower.

The cooled atomic beam was then incident on a magnetic guide constructed with multiple hexapole magnetic elements. Metastable neon atoms in the positive magnetic substates were guided through a 30\(^\circ\) arc and de-
flected from the other constituents of the atomic beam. The resulting pure, UV free metastable atomic beam had a flux of \((4.35 \times 10^9 \pm 2 \times 10^7)\) atoms s\(^{-1}\).

The design of the hexapole magnetic guide is cost effective and easily adaptable to any atomic beam line. The device could be adapted to also act as a conductance aperture for differentially pumped UHV systems. The phase space density of the emerging atomic beam could be further improved by the implementation of a magneto-optic compression element.

The formation of patterns using alternative resist layers was also investigated in an attempt to reduce the exposure times required to form negative contrast patterns. Gold coated silicon substrates with self-assembled monolayers of dodecanethiol (DDT) and ethanethiol (ET) were exposed to the Ne\(^*\) beam for various dosages to determine the properties of the resist layers. A comparison of the patterns formed verified that very short chain alkanethiols, such as ET, do not form a viable resist. Examining the surface energies of the resist layers using contact angle goniometry has shown that samples using ET as a resist only produce negative contrast patterns. This is due to the insufficient contrast in the wetting properties between exposed and unexposed regions below a critical dosage. Above the critical dosage these samples react in a manner similar to bare gold samples. It is postulated that negative pattern formation for ET coated substrates results from the desorption of ET from the surface during exposure and the subsequent deposition of a carbonaceous resist due to the presence of a background contaminant.

The experimental vacuum apparatus was found to have a significant effect on the dosages required to form negative contrast patterns on all samples. This highlighted the importance of the background contaminant, mechanical pump oil (MPO) in this case, in the pattern formation process. We confirmed that the partial pressure of MPO was a critical factor for the transition from positive to negative pattern formation on DDT coated samples. To accurately understand the rate determining factors of the process, the growth of contamination layers in the vacuum system was characterised using ellipsometry.

The growth of the carbonaceous film during exposure to the metastable beam was studied using a custom built rotating analyser ellipsometer. It was found that the desorption cross-section is many times larger than the polymerisation cross-section for a carbonaceous film grown on a silicon surface via Ne\(^*\) impact. The theory of film growth can be applied to other meta-
stable atom apparatus using the values determined and simple estimates for the mean residence time, volume, and cross-section of the contaminants involved.

The growth of carbonaceous films on these samples may be enhanced by increasing the partial pressure of a carbon containing material within the vacuum. However, care must be taken not to introduce levels of contamination that influence the operation of the experimental apparatus, such as mirrors in vacuo.

Direct deposition lithography

In contrast to the increasing complexity of the resist based atom lithography experiment, the direct deposition lithography experiment was simplified by the removal of any laser cooling apparatus. However, co-deposition experiments of iron (Fe) and nickel (Ni), where the Ni beam is incident at an angle of 45° relative to the substrate surface, introduces an increased complexity of the magnetic properties of the deposited structures.

Deposition of Fe atoms without laser cooling of the atomic beam has been achieved and patterning observed for atoms with a local average transverse velocity, $\langle v_{\text{trans}} \rangle$, of up to 4 m s$^{-1}$. The local $\langle v_{\text{trans}} \rangle$, which corresponds to an offset angle $\phi$ of the atomic beam with respect to the standing wave light mask, influences the height, width, and symmetry of the deposited line structures. These effects were observed experimentally, however, due to a substrate dependent diffusion mechanism, the observed structures were broader and less pronounced than those in simulated results. The scattering and interference of the standing wave light mask near the substrate also limits the resolution of the deposited structures. While simulated results yielded line structures with FWHM's $\geq 35$ nm, experimentally deposited lines were limited to structures with widths $> 80$ nm on SiOx substrates.

An initial characterisation of the magnetic properties of co-deposited Fe-Ni structures indicated that there are interesting effects that should be investigated further. The variations in Fe-Ni concentrations within the deposited structures produced a reduction in the coercive field when the magnetic field is applied parallel to the lines. This has been attributed to the nucleation of magnetic domains by regions where the Fe-Ni alloy possesses a lower magnetic moment per atom. A magnetic anisotropy was also ob-
served in the unstructured regions of the sample. This is likely induced by the incident angle of the Ni atomic beam and should also be investigated further.

The effect that the variation in alloy concentrations has on the magnetic properties of the sample should be thoroughly studied. To achieve this the spatial composition of the Fe-Ni layer would need to be determined using further energy dispersive X-ray spectroscopy or an alternative technique. Substrates containing the concentrations of Fe-Ni near that of Invar (Fe$_{64}$Ni$_{36}$) should produce some anomalous effects.

Currently only the in-plane magnetisation of the samples has been studied using the magneto-optic Kerr effect (MOKE). This should be expanded to include an investigation of the out-of-plane component of the magnetisation. MOKE experiments average the magnetic properties over a relatively large area on the sample surface. To determine the magnetic properties of individual deposited lines the application of magnetic force microscopy should be considered. Obtaining such results would provide a clearer indication of the domain structure and switching behaviour. For a complete characterisation of the co-deposition system the effects of line height, width, and height to pedestal height ratio would also need to be examined.
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[111] Magnets were sourced from K&J magnetics inc., Part No. B842.


[132] Value given by Inland Vacuum Industries, Inc.


Appendix A - Publications arising from this work

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