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Development of a Modulation-Based Magneto-Optic Kerr Effect (MOKE) Microscope

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Abstract

The magneto-optic Kerr effect (MOKE) is the rotation and ellipticity change of polarised light upon reflection from a magnetic material. This thesis outlines the development of a MOKE setup using a modulation technique to fully characterise the polarisation state of visible light. Whereas alternative polarimetry techniques require separate measurements to quantify the Kerr rotation, $\theta_K$, and the ellipticity angle, $\epsilon_K$, the dual photoelastic modulator (PEM) technique used here simultaneously measures both with a single optical path.

A full description and analysis of the optical and electronic setup is given. Emphasis is placed on thermal regulation of the entire setup to reduce signal drift, as well as the identification and minimisation of errors in the dual PEM detection system. The typical angular resolution of both MOKE angles is 12$\mu$rad, and a long timescale drift of up to 1.2$\mu$rad/hour is present. The spatial resolution of MOKE measurements is 4$\mu$m, as limited by the reflective objective lens used. This sets the resolution of magnetic domain images which are obtained by scanning the position of focus across the sample surface using piezoelectric transducers.

Three magnetic materials are investigated with the developed MOKE setup. Permalloy ($\text{Ni}_{80}\text{Fe}_{20}$) deposited on a nickel seed layer has a large permeability and is of interest as a core material in micro-inductors. Persistent domain structures within separated elements of 5$\mu$m thick permalloy are imaged using the scanning MOKE setup. This simple domain structure is only observed in high aspect ratio elements ($20\times300\mu\text{m}^2$) with large shape-induced uniaxial anisotropy which is useful for future micro-inductor design.

Magnetostrictive galfenol ($\text{Fe}_{1-x}\text{Ga}_x$) deposited on a piezoelectric 128° X-cut LiNbO$_3$ substrate is investigated for potential incorporation into surface acoustic wave (SAW) devices. A uniaxial magnetic anisotropy in the Kerr and ellipticity angles align with a strain potentially induced by the lattice mismatch between
galfenol and LiNbO$_3$. This anisotropy is present in 250nm thick galfenol films. Despite successful imaging of stripe domains in 250nm thick galfenol following AC demagnetisation along the magnetic hard axis, there is no evidence in the MOKE signal of SAWs controlling magnetostriction in the galfenol surface.

Finally, single crystals of Mn$_3$Sn, a hexagonal antiferromagnet with a large anomalous Hall effect signal, are aligned and polished for MOKE measurements. Evidence of a longitudinal ellipticity angle up to 0.12mrad, rather than the previously observed Kerr rotation, is presented. Square magnetic hysteresis is measured within 10° of the easy axis in (0110), and (2110) surface cuts. In both cases, the (0001) direction serves as the hard axis for which there is no measurable field dependence of $\theta_K$ or $\epsilon_K$. 
Lay Summary

The magneto-optic Kerr effect (MOKE) is the rotation of the plane of linearly polarised light upon reflection from a magnetic material. This rotation is coined the Kerr rotation and is referred to as the angle $\theta_K$. Practical applications of MOKE include magneto-optic drives which were popular data storage devices in the 1980’s, exploiting MOKE to read bits of data. Nowadays, MOKE is mainly used for research purposes - for magnetometry (measuring magnetism) and for imaging magnetic domains (regions with uniform magnetism in magnetic materials) - however, it may be exploited within future ultra-fast all-optical data storage devices.

In addition to a rotation upon reflection, the polarised light also becomes elliptically polarised, with a respective ellipticity angle $\epsilon_K$ subtended between the major and minor axes of the polarisation ellipse. Both $\theta_K$ and $\epsilon_K$ fully characterise the polarisation state of reflected light, and both may reveal information about the magnetic structure of the reflective material. Many techniques exist to measure $\theta_K$ and $\epsilon_K$, but the technique used in this thesis is the only one able to measure both simultaneously with a single optical path. Two photoelastic modulators (PEMs) are used to vary the polarisation state of light at 10’s of kHz, allowing synchronous measurement of $\theta_K$ and $\epsilon_K$.

This thesis outlines the development of an optical setup that can measure $\theta_K$ and $\epsilon_K$ rotations as small as $12 \mu$rad (less than one thousandth of a degree) using a dual PEM detection setup. A scanning mode that can be used to image magnetic domains with a resolution of $4 \mu$m is also described.

Three magnetic materials are then investigated with the developed MOKE setup. The first of which is small raised elements (mesas) of $5\mu$m thick permalloy (Ni$_{80}$Fe$_{20}$). Magnetic domain images within high aspect ratio ($20 \times 300 \mu$m$^2$) mesas are presented, showing a simple domain structure. This is relevant for the design
of micro-inductors - small electronic components to be used within micron-sized power supplies on chips that can reduce the footprint of electronic devices.

Secondly, the magnetic properties of thin films of galfenol (Fe$_{\sim85}$Ga$_{\sim15}$), a material that changes shape when magnetised (magnetostriction), as deposited on a LiNbO$_3$, a material that changes shape when a voltage is applied (piezoelectricity), substrate are investigated. The motivation of this study is to use galfenol and LiNbO$_3$ to develop surface acoustic wave (SAW) devices in order to control the magnetostriction in galfenol with a voltage applied to LiNbO$_3$. SAW devices are particularly useful in electronic elements, such as filters and transformers, and can also be used as sensors to measure pressure, temperature, strain, or torque. An anisotropy in measured $\theta_K$ and $\epsilon_K$ angles suggests that a crystal lattice mismatch between LiNbO$_3$ and galfenol induces additional magnetostriction in galfenol. This anisotropy is observed in 250nm thick galfenol films and not 600nm thick galfenol films, indicating a relaxation of the magnetostriction in galfenol after a few hundred nanometres. Despite successful imaging of stripe magnetic domains in 250nm thick galfenol following demagnetisation, there is no evidence in the MOKE signal of SAWs controlling magnetostriction in the galfenol surface.

Finally, MOKE measurements are presented on single crystals of Mn$_3$Sn, an antiferromagnet (a type of magnetic order that looks non-magnetic on a macroscopic scale) with a hexagonal crystal structure that has a large anomalous Hall effect signal which is typically linked to MOKE. Investigations into materials like Mn$_3$Sn may lead the way to future efficient high-density data storage mechanisms. The alignment, polishing, and magnetic characterisation of Mn$_3$Sn single crystals are described. Evidence of an in-plane ellipticity angle up to 0.12mrad, rather than the previously observed Kerr rotation, is presented. Square magnetic field sweeps (hysteresis) of $\epsilon_K$ within 10° of two directions (the ‘easy’ magnetic axes) in Mn$_3$Sn are observed. A ‘hard’ magnetic axis is also identified, in agreement to previous reports, for which there is no measurable field dependence of $\theta_K$ or $\epsilon_K$. 

Declaration

I declare that this thesis was composed by myself, that the work contained herein is my own except where explicitly stated otherwise in the text, and that this work has not been submitted for any other degree or professional qualification except as specified.

(Christopher Clark, February 2020)
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6.13 (a-l) Change in measured longitudinal Kerr rotation with field when reflected from the centre of the crystal in Figure (6.12a). The angle between the incidence plane of p-polarised light from the (01\(\bar{1}\)0) axis is indicated in the top right corner of each plot. Error bars represent the standard deviation from averaging over ten loops for (a-h) and seven loops for (i-l). . . . . . . . . . . . . . . . . . . . . . . . . . . . . . . . 145
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2.1 Key: MFM (magnetic force microscope), NV Centre (nitrogen vacancy), SEMPA (scanning electron microscope with polarisation analysis), TEM (transmission electron microscope), X-PEEM (X-ray photoemission electron microscope), XMCD (X-ray magnetic circular dichroism). Data from [81, 85–89].

3.1 Effective electronic noise due to each device. \(^{†}\)Responsivity=0.44A/W
*Gain=5×10\(^7\) (V/A).

5.1 Growth conditions and the relative compositions of the galfenol films using LiNbO\(_3\) as a substrate. Films were analysed and RF sputtered using a power of 60W by David Czerski.
Chapter 1

Introduction

This thesis outlines the development of an optical setup that is capable of quantifying the polarisation state of light reflected from magnetic materials. The unique aspect of the detection technique used here is the synchronous measurement of both the rotation of the plane of polarisation and the ellipticity angle. These measurements, coupled with the ability to scan the position of focus, allow the building of magnetic domain images that can be expressed in terms of both the rotation and elliptical angles.

This chapter provides an introduction to the relevant topics of this PhD thesis. A brief historical overview of magneto-optics, the interaction between light and magnetism, is provided and the linear magneto-optic effects are described. The different polarimetry techniques commonly used to measure magneto-optical effects are outlined and compared. Finally, an overview of recent magneto-optic studies is included to give context for the utility of magneto-optics, and motivation for the development of highly sensitive magneto-optic polarimeters.

1.1 The Magneto-Optic Kerr Effect (MOKE)

The interaction between light and magnetism was first experimentally observed in the mid 19th century by Michael Faraday [1]. Faraday observed the rotation of the plane of polarised light when transmitted through lead borosilicate glass while in the presence of a horse-shoe magnet. This rotation has been coined the Faraday effect, and in large part was a definitive illustration of the electromagnetic
theory of light that allowed James Clerk Maxwell to compile the works of other 19th century scientific powerhouses [2] which were further reduced to the four eponymous Maxwell equations by Oliver Heaviside [3].

Émile Verdet repeated Faraday’s experiment with more experimental rigour, and determined the equation denoting the exact Faraday rotation of the plane of polarisation ($\theta_F$) linking the distance polarised monochromatic light traverses through the material ($l$) and the magnetic field component in the direction of propagation ($H$), to a material-specific Verdet constant ($V_{\text{Verdet}}$) as the linear equation [4]

$$\theta_F = V_{\text{Verdet}} l H.$$  

(1.1)

Inspired by Faraday’s work, Rev. John Kerr investigated the effect of reflecting polarised light from a polished pole of an electromagnet. Indeed, Kerr measured a linear rotation of the plane of polarisation in response to the strength of the magnetic field from the pole piece [5], analogous to the Faraday effect. Thus, there are two linear effects describing the interaction between polarised light and magnetism: the Faraday effect in transmission, and the magneto-optic Kerr effect (MOKE) in reflection. The same physical origin is responsible for both effects.

Whilst MOKE is often referred to as the Kerr effect, this can be confused with the electro-optic Kerr effect, the change in refractive index of a material in the direction an electric field is applied. Therefore, the magneto-optic Kerr effect is referred to here as MOKE to remove ambiguity that is often found in literature.

Along with the two linear effects, there are higher order magneto-optic effects. The Voigt effect rotates and adds ellipticity to polarised light when at normal incidence but is proportional to the square of the field, making the polarisation rotations even in field [6]. The source of the Voigt effect is a linear birefringence [7, 8]. Similarly, the Cotton-Mouton effect is a linear birefringence that manifests in liquids with particles that are anisotropic (electrically and magnetically) [8, 9]. These higher order effects must be appreciated when analysing polar MOKE data to avoid mischaracterising the magneto-optic response of a material.

MOKE is illustrated in Figure (1.1a). Linearly polarised light incident on a magnetic material has its angle of polarisation rotated and ellipticity angle changed upon reflection. The reflected polarisation ellipse is defined by two angles, the rotation of the major ellipse relative to a reference axis $\theta_K$, and the ellipticity angle $\epsilon_K$ subtended between the polarisation ellipse major and minor
Three different orientations relating the magnetisation direction and the incident plane are illustrated in Figure (1.1b). Polar MOKE is observed at near normal incidence, and to first order is sensitive to out-of-plane magnetisation components. Longitudinal and transverse MOKE are sensitive to in-plane magnetisation components, with the longitudinal configuration sensitive to magnetisation parallel to the incidence plane, and transverse perpendicular to this. The type of incident polarisation also needs to be considered: both s- and p-polarisations may be used to measure polar and longitudinal MOKE, whereas the transverse MOKE is only observed for p-polarised light, where it manifests as an intensity change in reflected light [10] (this is true for magnetically isotropic materials; if there is any magnetic anisotropy in the material, transverse MOKE may actually lead to a nonzero $\theta_K$ and $\epsilon_K$ upon reflection [11]). Both longitudinal and transverse MOKE are most sensitive to large incidence angles, though both may be observed at any incidence angle that is not normal to the surface plane. In most practical experiments, the measured MOKE signal will in fact be a combination of all three effects due to a mixing between the polarisation state of light used, the incidence plane, and the magnetic field direction.
1.1.1 MOKE Measurement Techniques

Measuring any magneto-optic effect comes down to characterising a change in the polarisation of light before and after reflection from the material in question. The basic principles of quantifying the change in polarisation state requires a linear polariser to generate a known linear polarisation state which is reflected from the magnetic material. How this reflected light is then analysed can be one of several broad methods.

Crossed Polarisers

The simplest technique to measure the rotation of polarisation uses crossed polarisers. Light is sent through a linear polariser, reflected from the sample and then passed through another polariser at 90° to the first, acting as an analyser, before being detected. Figure (1.2) illustrates this polariser-sample-polariser setup. Light passing through this system should be extinguished unless there’s a change in the polarisation state due to the sample. Kerr’s original experiment was indeed carried out using this same method, with crossed Nicol prisms [5]. Nowadays, polarisers such as Glan-Thompson prisms, with typical extinction coefficients of $10^6$, are used to construct cheap and easily adaptable polarimeters.

There are then two methods for converting the electronic signal on the detector to a rotation of the plane of polarisation. The ‘null’ scheme has the analysing

![Figure 1.2](image_url)  

**Figure 1.2** A basic crossed polarisers setup to measure MOKE from a magnetic sample aligned using electromagnets. From [12].
polariser physically rotated to extinguish light incident on the detector. The typical resolution of a ‘null’ method is the same as that of the goniometer controlling the polariser rotation, typically 0.005°-0.01° (≈ 0.1mrad) [13]. The second method relies on a slight offset angle between the two polarisers for which changes in the intensity of light measured by the detector may be calibrated to a change in angle [12].

Additionally, a quarter waveplate may be placed between the sample and analysing polariser. This waveplate induces a $\frac{\pi}{2}$ phase difference between s- and p-polarisations, making the analyser-detector sensitive to the ellipticity angle rather than Kerr angle. Hence the polarisation ellipse may be fully quantified through two separate measurements.

**Balanced Photodiodes**

A balanced photodiode setup is illustrated in Figure (1.3). The Wollaston prism splits light into two beams of orthogonal polarisations, s- and p-polarisations, which are focused onto separate photodetectors. The difference in intensity on each detector is taken to give an output current that is proportional to the difference between s and p components of the polarised light. A waveplate (retarder) is required between the sample and the Wollaston prism. This half waveplate is typically rotated to ensure light is evenly split between s- and p-polarisations. A calibration procedure by varying the input linear polarisation and relating this to a change in differential current is used to quantify the measured polarisation rotation.

![Figure 1.3](image)  
*Figure 1.3* A balanced photodiode setup using a Wollaston prism to separate s- and p-polarisations to measure MOKE. Adapted from [14].
Modulation-Based Detection

Both the above methods may incorporate an additional optical device to modulate the polarisation state or intensity of light. The advantage that modulation provides is to allow AC detection which generally gives a larger signal-to-noise ratio when compared to the DC measurement. The simplest modulator is a rotating shutter (chopper) which may vary the light intensity at effective frequencies up to 10kHz. In this case, the photodetector is linked to a lock-in amplifier which may then extract the signal at the reference frequency of the chopper.

An alternative modulation technique changes the polarisation state of light rather than the total intensity. This modulation is most commonly carried out with either an electro-optic modulator (EOM), or a photoelastic modulator (PEM). PEMs are resonant devices used to periodically change the polarisation state of light passing through them. PEMs were originally invented in the 1960’s by Billardon & Badoz as a device to modulate light [15]. Much of the experimental development for the use of PEMs as a polarimetry tool was then carried out by Kemp [16] and Jasperson & Schnatterly [17].

PEMs function by exploiting the photoelastic effect - linear birefringence brought about by mechanical stress. A piezoelectric transducer is attached to a transparent optical element, as in Figure (1.4). The transducer periodically stresses the optical element at its resonance frequency, which in turn changes

![Figure 1.4](image_url)  
**Figure 1.4** Photoelastic modulator (PEM) operation. Strain applied to the optical element by the vibrating transducer modulates the linear birefringence $n_x$ relative to $n_y$. Retardance between $x$ and $y$ varies sinusoidally with the transducers vibration, periodically altering the polarisation state of incident light.
the refractive index in the direction of the applied stress, $n_x$. This change in $n_x$ periodically slows down and speeds up components of light in the $x$ direction (relative to $y$) inducing a phase shift between the components. The amplitude of the AC voltage applied to the PEM controls the maximum phase change between $x$ and $y$ directions and can be tuned to transform complete linear polarisation to complete circular, and every elliptical state in-between for a given wavelength of light. As the sinusoidal AC voltage may drive PEMs at frequencies up to 50kHz [18], there is a smaller contribution from flicker noise (a type of electronic noise which has many sources but has a power spectral density of the form $1/f$ [19]) when compared to lower frequencies, increasing the signal-to-noise ratio of any detected signal.

A PEM setup is similar to the crossed polariser setup and is illustrated in Figure (1.5). The PEM is oriented at 45° relative to the input polariser and analyser. The AC intensity incident on the detector at the frequency of the modulator is separated into the first and second harmonics (1f & 2f) which can be used in tandem with the DC signal to calculate the approximate Kerr angle and Kerr ellipticity in a single measurement [16, 17, 21]. This setup is useful for determining small rotations of the plane of polarisation (which needs to be approximately known before the measurement and aligned at 45° relative to the PEM) and the ellipticity angle synchronously.

A dual PEM setup has the advantage of simultaneously determining the polarisation angle and ellipticity angle, fully quantifying the polarisation state of light in a single measurement without approximations [22, 23]. This is the

![Figure 1.5](image.png)  
**Figure 1.5** A typical single photoelastic modulator (PEM) setup. From [20].
stand-out feature of a dual modulation-based setup and is the setup used in this thesis, which is described in detail in Section (2.4.1).

Operating on a similar principle to PEMs, an EOM relies on the Pockels effect - in certain materials linear birefringence is linearly proportional to the applied electric field. Hence, EOMs may alternatively be used to modulate the light polarisation. The Pockels effect is exploited in EOMs with an AC voltage which can be applied at frequencies up to 100MHz. EOMs can therefore act as extremely fast voltage-controlled waveplates, much like PEMs but with the advantage of much higher frequencies. The usable area of EOMs is usually far smaller than PEMs however, making them difficult to implement in some setups.

Interferometry

Interferometry relates to a class of techniques that can measure small displacements to high precision. The basic function relies on the constructive or destructive interference of light that experiences different paths. A type of interferometer known as the Sagnac interferometer was first used as a tool to measure the rotation of the plane of polarised light when sent through a magnetic material in the early 1990’s [24].

The Sagnac interferometer operates by splitting circularly polarised light of opposite handedness into two counter-propagating paths around a circular loop, as illustrated in Figure (1.6). Light passes through an EOM that periodically induces a phase shift between the two paths to allow phase-sensitive detection. After completing the loop, light is recombined and interferes at a detector. If the loop is reciprocal (i.e. if the path length is independent of direction travelled for each polarised state) the light will interfere constructively. Any dependence on the direction travelled however, will manifest itself as a phase difference at the detector and there will be destructive interference and an intensity change. For example, inserting a material into the loop will reveal any internal magnetic effects by inducing a phase shift between the two paths light travels. Such an apparatus has been shown to be able to discern Kerr rotations of order 10 nrad [25] at low temperatures, though only under ambient external fields, or for field-cooling measurements. This nrad sensitivity means the Sagnac interferometer is currently the most sensitive polarimetry tool.
Figure 1.6  Sagnac interferometer for measuring Faraday rotation $\theta_F$ of a sample using an electro-optic modulator (EOM). From [26].

1.2 Overview of Previous MOKE Experiments

MOKE is a commonly used characterisation technique for magnetic materials. As a tool for magnetometry, a MOKE setup may provide complementary information on magnetic materials to that provided by a SQUID (superconducting quantum interference device), VSM (vibrating-sample magnetometer), or torque magnetometer. The advantages of MOKE include:

- **Non-perturbative/non-destructive** - typically the probing light at optical wavelengths does not influence the magnetisation of the material which itself is unaffected by light.

- **Experimental setup** - for many ferromagnetic materials, the MOKE setup may be cheap and the optical components may be physically removed from the sample, allowing the sample to be placed in a vacuum and then manipulated in terms of temperature, stress, or magnetic fields applied.

- **Direction-sensitivity** - the incidence plane of light determines the sensitivity to magnetisation so that it can provide information on any magnetic directionality in the material.

- **Surface sensitivity** - for metals, optical light is reflected from the surface which allows the detection of magnetic surface states, separated from bulk effects.

- **Position sensitivity** - optical light may be focused to a sub-micron area so that nano-scale patterned features or magnetic position dependencies may be investigated.
• **Speed** - MOKE remains one of the best suited tools that can be designed to measure dynamic magnetic processes.

Disadvantages of MOKE include:

• **Small signal** - ferromagnetism in many exotic materials results only in a small MOKE signal, making the detection difficult.

• **Surface quality** - due to being a reflection-based technique, the surface quality has to be smooth enough over the scale of the wavelength of probing light. Scratches and surface defects can lead to spurious offsets.

• **Length Scale** - the resolution is determined by the wavelength of optical light and so typically may not be used as a tool to study nanometre-scale magnetism without clever preparation.

While Section (2.1.2) will discuss MOKE as a magnetic domain imaging tool, the following provides an overview of significant contributions MOKE has provided to the understanding of magnetic materials, and the various detection techniques employed are described.

In the mid 20\textsuperscript{th} century the discovery of microwave magneto-absorption in semiconductors [27] spurred interest in experimental magneto-optic techniques and led to them being used to investigate semiconductor band structures. Using magneto-optics as a spectroscopy tool by controlling the wavelength of light became common in the 1960's, with all manner of materials being investigated. Polar MOKE detection of both Kerr rotation and ellipticity was mainly carried out with crossed polariser setups, sometimes with Faraday cells acting as modulators, over the broad wavelength ranges 0.5-6eV (0.2-2.5\textmu m) with a general sensitivity of 0.1° (1.7mrad), though 0.001° (17\textmu rad) was claimed over visible wavelengths [28]. Several overviews of these spectroscopy measurements are recommended [28–31].

As technological progress dictates much of scientific research into functional materials, this steered magneto-optic development towards commercial applications. The first of which for MOKE was in the 1980's when magneto-optical drives became a viable data storage technique [32]. In the writing phase, a laser is used to locally heat a spot on a magnetic disk which can be written using an out-of-plane magnetic field to orient the magnetisation up or down. The reading
phase measures a polar MOKE signal, registering a positive or negative Kerr rotation from an up or down oriented grain in the magneto-optic disk. These magneto-optic disks therefore had to be made of magnetic materials that prefer the magnetisation to point out-of-plane. Whilst magneto-optic drives have been surpassed by higher density, and faster writing techniques for memory storage, the usefulness of MOKE remains both as an investigative tool as well as potential for future ultra-fast all-optical data storage [33, 34].

**Thin Film Measurements**

Magnetic thin films are important for technological applications. In the ultra-thin limit, magnetic films may be controlled as a single entity by applying magnetic or electric fields. How these individual films are understood further allows the building of heterostructures to perform complicated tasks [35].

Within magnetically layered structures, interfaces can produce unique electric and magnetic behaviours. Further to this, the reduced dimensionality of thin films can result in completely different physics on the surface compared to bulk effects of the same material. The first experimental study of surface magnetism in atomically thin Fe(100) films grown onto Au(100) was carried out with a MOKE setup, with the authors able to obtain in-plane longitudinal hysteresis with sensitivity that varied on changing atomic thickness and temperature [36]. This study used crossed polarisers, recording the intensity change as a function of applied magnetic field and only gave an estimated calculation for the Kerr rotation. Since then, MOKE has become a staple for monitoring growth of magnetic thin films, providing information on the magnetism during the deposition [12, 37]. The separation of the optics and the sample allow the sample to be placed in extreme environments, whereas alternative magnetometry tools such as a SQUID cannot be used during the growth process.

For measuring the magnetic dynamics of thin films within heterostructures (in-situ), magnetic second harmonic generation (SHG) is commonly used as it is highly sensitive to surfaces and interfaces [38]. SHG is a non-linear effect resulting from inversion symmetry breaking. While the MOKE signal is detected at the frequency of incident light, SHG represents the small fraction of light that is reflected with doubled frequency [39]. With clever application of linear MOKE, it has been demonstrated that MOKE may be additionally used as a depth-sensitive tool to characterise the local magnetic state of nested thin films provided enough
is known about each layer beforehand [40]. This technique however, requires several measurements with careful control and variance of the incidence plane, frequency of light used, and detection sensitivity. Hence, MOKE is rarely used in place of SHG for measuring buried interfaces.

The Spin Hall Effect

The classical Hall effect is the accumulation of charge on the lateral surfaces of a current-carrying sample. Analogous to this, the spin Hall effect (SHE) is a spin accumulation on the lateral surfaces of a current-carrying sample [41, 42]. There are two mechanisms leading to the SHE: the extrinsic spin-dependent Mott scattering of charge carriers with unpolarised impurities [43], and the intrinsic mechanism arising from spin-orbit coupling within the material with large Rashba spin-splitting without the need for impurities to scatter from [44].

The first experimental observation of the SHE was in the semiconductor GaAs at 30K using magneto-optical detection [45]. Data from this first success is

![Figure 1.7](image)

**Figure 1.7** The first experimental observation of the spin Hall effect in GaAs. (a) the schematics of the GaAs sample, (b) the spin accumulation $n_s$ (left) and the corresponding Reflectivity (right) of the GaAs bar, (c) the Kerr rotation laterally across the GaAs bar for different external fields applied, and (d) the peak Kerr rotation $A_0$ across the sample. Adapted from [41, 45].
displayed in Figure (1.7). The spin accumulation on either edge of the GaAs bar (transverse to the current direction) is shown in the 2D scan in Figure (1.7b). Figure (1.7c) shows the effect of applying an external field parallel to the spin current direction, with the relative Kerr rotation being ±2µrad on either edge in zero field. The measurement system used in this experiment used a pump-probe technique. A normally incident circularly polarised pump pulse was used to excite spin-polarised electrons, a brief time delay then separated the linearly polarised probe pulse (from a Ti:sapphire laser), which experienced the Kerr rotation [46]. A balanced photodiode detection system was used to measure the polar Kerr rotation, with a Glan-Taylor prism to separate orthogonal polarisation states [45–47].

Detecting the SHE in metals is more difficult, as spin accumulation as in Figure (1.7b) cannot be resolved due to the spin diffusion length $l_s$ being several nm compared to several µm in semiconductors [48]. Despite several attempts having no success in detecting a pure intrinsic SHE [49, 50], recently however, the Kerr rotation was successfully measured on the non-magnetic metals Pt & W [48]. The experimental setup measured the longitudinal Kerr rotation using a balanced photodiode and sinusoidally modulated current passed through the metal samples. Results of SHE-induced Kerr rotation for applied current density and incidence direction for Pt and W are shown in Figure (1.8). The opposite sign of the slope for Pt and W shows the effect of the different sign of the SHE.

![Figure 1.8](image.png)

**Figure 1.8** Spin Hall effect-induced Kerr rotation on non-magnetic metals Pt (Left, 15nm thick) & W (Right, 10nm thick) as a function of current density and incident direction (the sign of $s$ represents the sign of incidence angle used) [48].
Time resolved Measurements

The optical study and control of ultra-fast magnetic dynamics is currently of great interest for data storage, spintronics, and quantum computing [51]. Nowadays, light pulses can be controlled on the femtosecond time scale [52] however, the limitation still comes from the spatial resolution which is determined by the wavelength of light used.

Time resolved measurements usually follow a pump-probe technique with different intensities, polarisation states and/or frequencies used for the pump and probe beams [51]. The pump beam generally affects the material magnetically through local heating (heat assisted magnetic recording) [53], or via spin transfer torque where the momentum of circularly polarised light is transferred to the magnetisation [54]. A time delay then separates the probe beam which measures the response of the material to the pump, commonly as a time resolved MOKE measurement, and different wavelength ranges provide complementary information on the magnetisation dynamics [51].

Magnetisation dynamics on the sub-picosecond timescale were first observed in the mid 1990’s using longitudinal MOKE with a crossed-polariser technique and demonstrating hysteresis loops in the intensity of light passing through the analyser [55]. A 22nm thick film of polycrystalline Ni was used and the magnetic hysteresis before and after a pump beam was applied were compared, with the effect of the pump beam being a demagnetisation of the Ni. The interpretation of the dynamics in a relatively simple metal such as Ni still proved difficult due to a separation in the magnetic response of the material, and the optical response observed through MOKE, which depends heavily on the wavelength of light used for probing [56].

A recent example of a time resolved MOKE measurement is illustrated in Figure (1.9). A pump pulse excites up and down spin electrons in an Fe layer (emitter), physically spaced by a Au layer from another Fe layer (collector) the probe pulse is reflected from. An unequal transmittance of up and down spins through the Fe-Au interface results in a spin-polarised current making its way to the Fe collector layer. This spin current transfers momentum, resulting in spin transfer torque to the magnetisation of the Fe collector layer [57].

In Figure (1.9), the temporal sensitivity of the probe beam is clear, with oscillations in both Kerr rotation and ellipticity. The $\frac{\pi}{2}$ phase shift between
Figure 1.9 Extracted spin transfer torque-induced magnetisation dynamics as measured using a pump-probe technique and time resolved MOKE. From [57].

longitudinal (green) and polar (red, blue) MOKE signals demonstrates the precession of the magnetisation in the Fe collector layer following the pump pulse in the Fe emitter layer. The MOKE setup used a modulated balanced photodetector method, with a 1MHz pulsed Ti:sapphire laser in addition to separate choppers for the pump and probe beams. Both Kerr rotation and ellipticity were measured synchronously by splitting the reflected probe beam into two paths with a beamsplitter. Each path was individually detected using a balanced photodetector, with the ellipticity-sensitive path having a quarter waveplate placed before the Glan-Taylor prism. This technique requires careful calibration to ensure the effect of the beamsplitter is negated (a different s- to p-polarisation transmittance/reflectance can lead to spurious offsets in the polarisation state emitted/reflected). The sensitivity of this setup is around 1µrad.

Low Temperature Measurements

Measuring at low temperatures (less than 300K, and down to near absolute zero) comes with experimental difficulties. Low temperature samples must be isolated in a vacuum to adequately cool them and prevent condensation forming on the sample and any electronics. Accessing the sample with light is the main obstacle for measuring variable temperature MOKE. There are two main methods for overcoming this challenge: having a glass window built into the cryogen, or using fibre optics to guide light into the cryogen.
Using a view-port to access the sample requires the glass to transmit light at the measurement wavelengths (usually in the visible range) whilst being opaque for infra-red wavelengths to reduce heating effects. Placement of the objective lens used to focus light is also important. The simplest systems use a large working distance lens, which may or may not take into account the thickness of the glass, generally adding aberrations and reducing the focus quality (for examples, see [58, 59]). For the highest quality focusing, the objective lens is placed after the window, and is usually cooled with the sample (for example see [60, 61], though some setups may keep the lens at room temperature [62]). The relative positions of the objective lens, window and sample are illustrated in Figure (1.10). Both techniques require the glass view-port to be made stress-free to reduce birefringence that can lead to spurious rotations in the polarised light. A common choice for this glass is fused silica which is strong, has relatively small stress-induced birefringence, and has a transmission range of 0.2-2.1µm [63–65]. Additional stress induced on the glass from the holder, or the pressure difference of the vacuum compared to air must also be minimised. The simplest MOKE setups using crossed polarisers or balanced photodiode techniques typically add in a retarder before the analyser which is rotated to counter any window birefringence when calibrating the signal. Additionally, the reflection of light from the window may be an issue, leading to the use of anti-reflection coatings, although these may also have an effect on the polarisation state of light used for the MOKE measurement [66].

General issues further arise from vibrations of the compressor, vacuum pump, or scroll pumps commonly used for cooling to cryogenic temperatures. These vibrations from physical movement may be somewhat counteracted by damping vibrations from leads connecting to the sample stage, or by mechanically

![Figure 1.10](image) The possible relative positions of the window, objective lens, and sample when imaging in a cryogen. Adapted from [67].
separating the sample from the moving parts. Using fibre-optics, this problem is reduced, as the objective lens and sample are mounted together and are detached from the outside optics whereas the relative movement between the outside optics and the lens-sample mount when using a view-port is more of a problem (when using a low magnification lens placed outside the vacuum, this problem is reduced). Additionally, if a magnet is used to change the magnetic state of the sample, the effect this external field has on either the glass view-port, the fibre-optics, or the objective lens has to be compensated for. As Faraday rotation arises when the magnetic field is aligned with the polarised light momentum vector, mirrors may be used to ensure light passes through the window when the surface is aligned with the external field [68].

In terms of cryogenic measurements using polarisation maintaining fibre-optics to guide light, the Sagnac interferometer has seen great success. This technique is naturally insensitive to sources of linear birefringence that are found even in polarisation-maintaining optical fibres which any other MOKE measurement technique would have to account for. The downside to the Sagnac interferometer however, is the inability to apply large magnetic fields due to complicated Faraday rotations in the optical fibres. The advantage of techniques not using optical fibres, such as the dual PEM technique used in this thesis, is the application of large magnetic fields.

Despite the limitation of static magnetic fields, useful information may be obtained by comparing field cooling (where the magnetic field is fixed, and the temperature is decreased, usually across a magnetic transition in the sample being investigated). A few examples of significant contributions the Sagnac interferometer as a tool to measure MOKE at ultra-low temperatures include:

- Detecting a deviation of 65nrad in $\theta_K$ at $T\approx1K$ in Sr$_2$RuO$_4$ [69] supported evidence for a time reversal symmetry breaking (TRS B) state within the superconducting state, and helped narrow down the most probable type of order parameter in Sr$_2$RuO$_4$. Additionally, the thickness dependence of the conducting and ferromagnetic states in SrRuO$_3$ deposited on SrTiO$_3$ show a transition to insulating and antiferromagnetic states below 4 monolayers [70].

- Kerr rotations of $1\mu$rad at $T\approx1K$ with an onset at $T\approx150K$ in copper oxide systems indicate the transition into a charge-ordered chiral state [71–73].

- Investigating the multiple superconducting phases in UPt$_3$ revealed a
deviation of 400nrad in $\theta_K$ at 350mK compared to 500mK [74]. This was obtained through separate field-cooling sweeps for oppositely aligned magnetic fields, as shown in Figure (1.11). The top red plot represents positive field cooling; the bottom blue curve represents negative field cooling. The $\Delta \theta_K$ occurs separately to the superconducting transition, indicating that TRSB does not occur in all phases which, when combined with other studies, narrows down the possible form of the order parameter in UPt$_3$.

![Figure 1.11](image)

**Figure 1.11** The change in Kerr signal, $\Delta \theta_K$, from UPt$_3$ upon field cooling to train the magnetisation measured using the Sagnac interferometer. From [74].

### 1.2.1 Comparison With Alternative MOKE Detection Systems

The detection scheme that forms the basis of the developed MOKE system described in this thesis is a dual PEM setup. An overview of this method of polarisation detection is included in Section (2.4.1) and an analysis of the errors associated with it is included in Section (3.4). The key advantage of this system is the synchronous quantification of both $\theta_K$ and $\epsilon_K$ that fully describe the polarisation state of light. This is in addition to the possibility of applying large magnetic fields that techniques using optical fibres may not (due to parasitic Faraday rotations). Like alternative techniques, the MOKE setup here may be
used to image magnetic domains by raster scanning the position of focus. The domain images may, however, be imaged in both the Kerr rotation and ellipticity angle, for which some materials may be more sensitive to depending on their bandstructure.

Whilst alternative polarimetry techniques can attain greater angular sensitivity than the dual PEM setup used here (a balanced photodiode setup or the Sagnac interferometer can attain near nrad sensitivity in comparison to the $\mu$rad sensitivity of a dual PEM setup), these selectively measure $\theta_K$ or $\epsilon_K$. To measure both $\theta_K$ and $\epsilon_K$ using alternative techniques requires subsequent measurements or a splitting of the measurement light beam which may be influenced by optical interfaces differently. The cost of the system described in this thesis is comparable to alternative techniques that also require modulation of the polarisation state/intensity of light. The simplest and cheapest (and least sensitive) magnetic imaging or polarisation detection method uses crossed polarisers which may be incorporated into conventional optical microscopes.

### 1.3 Thesis Outline

The outline of the remainder of this thesis is as follows:

- **Chapter 2** provides the theoretical background to work pertinent to this thesis. A classical and quantum explanation of both magnetism in ferromagnetic materials, and the magneto-optic Kerr effect will be provided. How polarisation is characterised will then be described, and the theory behind the dual photoelastic modulator detection setup, as used in the remaining chapters, will be outlined and its limitations analysed.

- **Chapter 3** highlights the experimental development that forms the bulk of this thesis, with consideration going to analysing the optical layout, the parameters defining the setup in terms of spatial resolution and sources of drift and noise. Work to adapt the MOKE setup for cryogenic measurements and the image processing software to produce magnetic domain images by scanning the MOKE microscope will also be discussed.

- **Chapter 4** presents measurements of persistent magnetic domains within raised permalloy ($\text{Ni}_80\text{Fe}_{20}$) structures deposited on a nickel seed layer using the scanning MOKE setup.
Chapter 5 is focused on MOKE measurements recorded on thin films of galfenol (Fe$_{1-x}$Ga$_x$), a magnetostrictive alloy, as deposited on a LiNbO$_3$ piezoelectric substrate. Longitudinal and transverse MOKE hysteresis loops, along with domain images following demagnetisation of the galfenol film, will be presented and discussed.

Chapter 6 will discuss longitudinal MOKE measurements recorded on the hexagonal antiferromagnet Mn$_3$Sn. Preparation of Mn$_3$Sn single crystals, in terms of alignment and polishing, for optical measurements is also outlined.

Chapter 7 will finally conclude the work presented in this thesis, and outline the direction future work may take.
Chapter 2

Theoretical Background

This chapter outlines the theoretical background to magnetism in ferromagnetic materials, with classical and quantum mechanical descriptions compared. Similarly, a description of the magneto-optic Kerr effect will be given, as well as an overview of the theory describing the different measurement orientations possible. This is followed by the fundamentals to characterising polarisation states using Stokes parameters and Mueller matrices. This will lead onto the calculation describing the dual photoelastic modulator detection system used to measure the polarisation state of light in the remainder of this thesis.

2.1 Ferromagnetism

The magnetisation \( M \) in magnetic materials is defined by the magnetic moment \( m \) per unit volume \( V \):

\[
M = \frac{m}{V}.
\]  

Magnetic materials are categorised according to their response to an external magnetic field. Ferromagnets are notable in that they retain a spontaneous magnetisation in zero field when cooled below their Curie temperature. The interactions between magnetic moments cause a real-space alignment direction to be favoured. Whilst paramagnets and diamagnets both have zero net magnetisation, the presence of an external magnetic field distinguishes them. Moments in diamagnets arrange themselves to cancel out any external field within the material, and moments within paramagnets arrange themselves with the
external field to enhance it within the material. Antiferromagnets also have zero net magnetisation however, moments are ordered such that their macroscopic magnetisation is zero. An imbalance of moments within antiferromagnets can lead to ferrimagnetism, a net moment in one direction, or weak ferromagnetism which is the result of a canted antiferromagnetic structure.

2.1.1 Magnetic Domains

The competing interactions in a ferromagnet are described by the Landau-Lifshitz equation, which itself leads to a key feature of ferromagnetic materials: the formation of domains. Magnetic domains are regions with uniform magnetisation; neighbouring domains have magnetisation vectors pointing in different directions. Magnetic domains naturally arise from minimising the total free energy, given by [75]

\[
E_{\text{tot}} = \int V \left( E_{\text{ex}}(m) + E_{\text{an}}(m) + E_{\text{de}}(m) + E_{\text{st}}(m) + E_{\text{ms}}(m) \right) dV. \tag{2.2}
\]

Each term represents an energy density, which may be integrated over the volume to obtain the total energy. The origin and relevance of each term in Equation (2.2) is as follows.

The exchange interaction is most readily understood with a Heisenberg lattice, named after Heisenberg who first considered it [76]. The exchange term arises from the Coulomb interaction, causing a repulsion for aligned neighbouring spins, and the Pauli exclusion principle, preventing two electrons from occupying the same state. This behaviour is encapsulated in an exchange constant, \( J_{ij} \), which may be positive or negative. For localised spins \( S_i \) and \( S_j \), the interaction between them has an energy

\[
E_{\text{ex}}(m) = -\sum_{i \neq j} J_{ij} \langle \vec{S}_i \cdot \vec{S}_j \rangle. \tag{2.3}
\]

Thus it is favourable to have parallel spins if \( J_{ij} > 0 \), leading to ferromagnetism, or anti-parallel spins if \( J_{ij} < 0 \), leading to antiferromagnetism.

An often-quoted characteristic of magnetic materials is the exchange stiffness, \( A \),
which is related to \( J_{ij} \) by

\[
A \approx \sum_{i \neq j} J_{ij} \frac{\langle \vec{S}_i \cdot \vec{S}_j \rangle}{a_0} = \frac{k_B T_{\text{Curie}}}{2a_0},
\]

(2.4)

where \( Z_c \) is a constant depending on the symmetry of the crystal structure, and \( a_0 \) is the lattice constant. The exchange length \( l \) characterises the difference between the exchange energy and the demagnetising energy with a saturation magnetisation \( M_s \) and is defined as

\[
l = \sqrt{\frac{A}{\mu_0 M_s^2}}.
\]

(2.5)

The anisotropy term accounts for directional dependence of the magnetisation. Some crystal structures, for example, have a magnetic easy axis or an axis along which the spontaneous magnetisation produces a minimum in energy. This magnetic anisotropy generally shares symmetry with the crystal structure so is commonly referred to as magnetocrystalline anisotropy. The magnetocrystalline anisotropy energy depends on the structure of the material, as well as the angle of the applied external field relative to each axis. For a uniaxial material (the simplest example), the anisotropy energy is given by expanding in a series of \( \sin^2(\phi) \) terms with \( \phi \) being the angle between the applied external field and the magnetic easy axis [77]:

\[
E_{an}(m) = K_{u1} \sin^2(\phi) + K_{u2} \sin^4(\phi) + ... \tag{2.6}
\]

with anisotropy constants \( K_{un} \).

An additional type of anisotropy may arise due to the physical dimensions of a magnetic material. For example a sub-micron sized elongated ellipsoid will preferentially have magnetisation pointing along its long axis, and the semi minor axis would correspond to its hard axis. On the long edges of the ellipsoid there is a component of magnetisation normal to a surface which acts to increase the self energy of the particle. This shape anisotropy is one route of generating a demagnetising field which represents an additional term in Equation (2.2).

The demagnetising energy arises due to the difference between the magnetic field inside and outside the ferromagnetic material. The demagnetising field for a
sample magnetisation $M_0$ is given by

$$H_{de} = \frac{N_i M_0}{\mu_0}, \quad i = x, y, z \quad (2.7)$$

with a demagnetising factor $N_i$ along each principle axis. And so the demagnetising energy may be expressed in terms of the overlap between the demagnetising vector and the magnetisation:

$$E_{de}(m) = -\frac{1}{2}\mu_0 \vec{M}_0 \cdot \vec{H}_{de}. \quad (2.8)$$

Applying an external stress $\sigma_{ij}$ can induce a strain in the material. How this applied stress couples to the material depends on the elastic strain tensor $\epsilon_{ij}$ which in turn contributes an energy

$$E_{st}(m) = -\sum_{i,j} \sigma_{ij} \epsilon_{ij}. \quad (2.9)$$

In an isotropic material, this physical stress may be modelled as a uniaxial anisotropy with an effective anisotropy constant of $K_u = \frac{3}{2} \lambda_S \sigma$, with $\lambda_S$ being a dimensionless parameter indicating the strength of the magneto-elastic interaction.

In a similar vein, magnetostriction is a physical change in dimensions of a ferromagnet due to stresses caused by an internal magnetic field. This local stress acts to elongate the physical dimension that the stress is applied along. For a uniaxial ferromagnet the spontaneous magnetostriction elongation for length $l$ may be expressed as

$$\Delta \left( \frac{\delta l}{l} \right) = \frac{3}{2} \lambda_s \left( 1 - \cos(\phi)^2 \right), \quad (2.10)$$

where $\phi$ is the angle of the applied field relative to the magnetic easy axis.

The formation of magnetic domains naturally occurs following energy arguments. Figure (2.1a) illustrates a single domain particle and its corresponding magnetic field lines. The saturated state has magnetic poles at either edge, contributing demagnetisation energy. Dividing the single domain particle into multiple domains as in Figure (2.1b-c) reduces the stray field energy. If the energy of the wall is less than the demagnetising energy, the total free energy will be lower for a multi-domain particle and this domain splitting process will continue until it
Figure 2.1  The stray fields from a single domain particle (a) as they develop towards a multi-domains particle ((b)-(c)) to reduce demagnetising energy. Either state (d) or (e) may be energetically preferred depending on the material and exact dimensions. From [77].

is not energetically favourable. Two possible outcomes for the simple particle are illustrated in Figure (2.1d-e). The resulting size of domains, and pattern depends on the ferromagnet itself in terms of Equation (2.2).

**Magnetic Domain Walls**

Separating domains is an area of either slow or abrupt rotation of the magnetisation direction, known as a domain wall. Two types of walls are defined corresponding to the plane of the rotation of magnetic moments:

- **Bloch walls** have magnetic moments rotate in the plane of the wall, and normal to the surface (Figure 2.2a).

- **Néel walls** have moments rotate in the plane of the domains, parallel to the surface (Figure 2.2b).

The type of domain wall in a material is that which minimises Equation (2.2), with the key parameters being the exchange and anisotropy terms. In many materials, the type of domain wall is largely determined by physical dimensions of the material. Very thin materials (with thicknesses smaller than the exchange length) for example strongly favour in-plane rotations (a Néel wall). The key...
Figure 2.2  Comparison between the moment rotation across a) Bloch walls and b) Néel walls. From [78].

length scale determining the switch over between wall types for a given material however, is the exchange length.

The Hysteresis Loop and Magnetisation Processes

Magnetic hysteresis, or memory, is a quintessential feature of ferromagnets. After being magnetised, a ferromagnet will retain its magnetisation when the external field is reduced to zero. An oppositely directed external field is then needed to change, or switch, the magnetisation. There are three main processes for the switching of magnetisation:

- Coherent rotation of moments
- Domain wall movement
- Nucleation and growth

With similarity to the type of domain wall present in a magnetic material, the switching process depends on the dynamics of Equation (2.2). A source of anisotropy such as the physical dimensions, or an applied directional stress can influence the most energetically favourable process. It is common for isolated micron-sized ferromagnets to behave as a single domain particle. This type of particle is too small to have domains form and so the switching process occurs with all spins rotating together in a coherent fashion.
If there is strong shape anisotropy, as in an ellipsoidal particle, then the hysteresis behaviour will vary depending on the direction of the applied field. Figure (2.3) demonstrates this magnetic anisotropy for such an ellipsoid that is known as a Stoner-Wohlfarth particle. When the applied field is directed along the easy axis (0°), it is energetically favourable for the magnetisation to point along this direction so that there is little change in magnetisation when the field is reduced from saturation until a critical point, the coercivity, flips the magnetisation direction. As the direction of the applied field is rotated towards the hard axis, the measured magnetisation component decreases more readily until the response of the single domain particle is linear at 90°.

Figure (2.4) displays the distribution of moments for each stage of the hysteresis loop. Starting in the demagnetised state (position O), moments may point in any direction. As the field is increased (O-B-C), moments preferentially point towards the direction of the applied field, with saturation occurring with a small range of possible angles moments may point. Decreasing the field from saturation (C-D) allows moments to ‘relax’ and point in a larger range of angles when the field is reduced to zero. This magnetisation at position D is noted as the remnant magnetisation $M_R$. Reversing the magnetic field (D-E) leads to an overall zero net magnetisation at E, as moments may point in any direction. This field value at E is the coercivity of the material $H_C$. 

**Figure 2.3** Hysteresis curves under the Stoner-Wohlfarth model. Numbers indicate the angle between the external field direction and the magnetic easy axis for each curve. From [75].
2.1.2 Domain Imaging Techniques

Magnetic domain imaging techniques have progressed since the early twentieth century, when stray fields were used to align a ferrofluid placed on the surface of a magnetic material. The ferrofluid aligns along domain walls which are regions of high magnetic flux. This technique is known as the Bitter technique, after the original inventor, Francis Bitter [80].

MOKE is a common technique used to image magnetic domains, owing to its relatively simple implementation nowadays. A conventional microscope can be adapted to obtain MOKE images. Using crossed polarisers, digital difference techniques can be used to subtract a non-magnetic background [81]. Further to this, quantitative Kerr microscopy can separate different components of magnetisation orthogonal to each other. Despite the name, quantitative MOKE microscopy does not ascribe a physical rotation to the measurement but rather, can separate domains with different magnetisation vectors. This is achieved by separating signals from different incidence planes.

The method for imaging magnetic domains used in this thesis is by scanning the focus position of the MOKE microscope. This technique is known as scanning MOKE microscopy [82]. The advantages over a fixed MOKE microscope are that it is quantitative, providing a physical rotation and ellipticity for each measurement point, and it allows for detection of smaller signals. The main disadvantage however, is the time taken to complete an image with a scanning method which must be balanced with the resolution required.

Similar to the Bitter method, magnetic domains can be visualised using the stray
field through magneto-optical methods. Magneto-optic indicator films (MOIFs) are thin single crystalline layers of a magneto-optically active material that can be placed on a ferromagnetic sample to increase its magneto-optic contrast [83]. MOIFs are useful for imaging the magnetic structure of a ferromagnetic material with typically weak Faraday or Kerr rotations. How MOIFs improve magneto-optic contrast is illustrated by Figure (2.5). The local stray field from the ferromagnetic material (FM) penetrates the MOIF and aligns the ferrite garnet layer (IG). This IG layer has a Faraday rotation that is usually orders of magnitude larger than Faraday or Kerr rotation of the FM being probed. Hence it is easier to distinguish between different magnetic domains using a MOIF than without one. Alternatively, domain images that are sensitive to the stray field can be made using a magnetic force microscope (MFM), or the relatively recent technique using nitrogen-vacancy (NV) centres [84, 85].

The comparison of various other techniques for direct domain imaging are summarised in Table (2.1). A more in depth comparison of each of these techniques may be found in [81, 86].
<table>
<thead>
<tr>
<th>Method</th>
<th>Spatial Resolution</th>
<th>Depth Sensitivity</th>
<th>Speed</th>
<th>Quantitative?</th>
<th>Surface Quality Requirement</th>
<th>Allowed Field Range</th>
<th>Infrastructure Cost</th>
</tr>
</thead>
<tbody>
<tr>
<td>Scanning MOKE</td>
<td>0.3µm</td>
<td>20nm</td>
<td>10 mins-hours</td>
<td>Yes</td>
<td>High</td>
<td>Any</td>
<td>Medium</td>
</tr>
<tr>
<td>Widefield MOKE</td>
<td>0.3µm</td>
<td>20nm</td>
<td>10ms-1 second</td>
<td>No</td>
<td>High</td>
<td>Any</td>
<td>Low</td>
</tr>
<tr>
<td>MFM</td>
<td>&lt;10nm</td>
<td>1µm</td>
<td>5 mins-hours</td>
<td>No</td>
<td>None</td>
<td>1T</td>
<td>Medium</td>
</tr>
<tr>
<td>NV Centre</td>
<td>&gt;4nm</td>
<td>1µm</td>
<td>seconds-hours</td>
<td>Yes</td>
<td>None</td>
<td>Any</td>
<td>Low</td>
</tr>
<tr>
<td>SEMPA</td>
<td>10nm</td>
<td>1nm</td>
<td>1min-2hours</td>
<td>Yes</td>
<td>High</td>
<td>0.02T</td>
<td>High</td>
</tr>
<tr>
<td>Lorentz TEM</td>
<td>&lt;5nm</td>
<td>Whole sample</td>
<td>~0.05-30 seconds</td>
<td>Yes</td>
<td>None</td>
<td>0.6T</td>
<td>High</td>
</tr>
<tr>
<td>X-PEEM</td>
<td>50nm</td>
<td>50nm</td>
<td>0.1-10 seconds</td>
<td>Yes</td>
<td>High</td>
<td>Any</td>
<td>High</td>
</tr>
<tr>
<td>XMCD</td>
<td>0.3µm</td>
<td>10nm</td>
<td>0.1 seconds-10 mins</td>
<td>Yes</td>
<td>None</td>
<td>Any</td>
<td>High</td>
</tr>
<tr>
<td>Bitter</td>
<td>0.1µm</td>
<td>1µm</td>
<td>1 second</td>
<td>No</td>
<td>Moderate</td>
<td>0.02T</td>
<td>Low</td>
</tr>
</tbody>
</table>

Table 2.1 Key: MFM (magnetic force microscope), NV Centre (nitrogen vacancy), SEMPA (scanning electron microscope with polarisation analysis), TEM (transmission electron microscope), X-PEEM (X-ray photoemission electron microscope), XMCD (X-ray magnetic circular dichroism). Data from [81, 85–89].
2.2 The Magneto-Optic Kerr Effect (MOKE)

The magneto-optic Kerr effect was briefly described in Section (1.1), and illustrated in Figure (1.1a). Magnetic materials follow the general trend that $\theta_K \propto M$; a larger magnetisation leads to a larger polar Kerr rotation, as displayed in Figure (2.6) for a number of ferro-, ferri-, and antiferro-magnets.

![Figure 2.6](image)

**Figure 2.6** The relation between magnetisation ($M$) and polar MOKE signal ($\theta_K$) for various magnetic materials. Adapted from [90].

This section first presents a superficial semi-classical explanation for MOKE in terms of the normal states of light propagation through materials, and then goes on to present how MOKE is fundamentally understood as due to the spin-orbit interaction.

### 2.2.1 Semi-Classical Description of MOKE

Both linear magneto-optic effects (MOKE and the Faraday effect) arise from the same physical process. Hence, the classical description of MOKE is the same as that of the Faraday effect, only the former concerns the rotation of reflected polarised light, and the latter the rotation of transmitted polarised light.

The classical explanation of the MOKE is as follows. Linear polarisation can be decomposed into the superposition of phase- and amplitude-coherent left and right circular polarisations (LCP and RCP respectively). These are the normal modes of light propagation within a material. The electric field of LCP (RCP) that propagates through a medium will excite electrons, causing them to orbit in
left (right) circular orbits. With no magnetic field in the medium, the orbits will have the same radius; linear polarisation (that is, the superposition of LCP and RCP) will be unchanged. Once a magnetic field is present however, the electrons will feel different Lorentz forces, either away from, or towards, the centre of the orbit and the radius of orbit will either increase or decrease. Once light has exited the material the superposition of LCP and RCP will be different in comparison to the incident light; the linear polarisation state will have rotated, and different absorption rates between LCP and RCP will have changed the ellipticity.

This classical explanation is qualitatively correct however, to quantify magneto-optic effects in ferromagnets would require an effective field of the same order as the Weiss field [91]. The Weiss field is due to the exchange interaction between electrons, representing an effective magnetic field that aligns electron spins rather than an actual magnetic field that can affect electron motion. It is the physical motion of electrons that produces a current that in turn determines optical properties. Hence, the Weiss field cannot be used to explain optical properties, and the classical understanding is quantifiably incorrect.

To calculate the magnetic response of a simple material to light, it is natural to consider the dielectric tensor $\tilde{\varepsilon}$ of a homogeneous medium. As electric dipole moments are proportional to the radii of electron orbits, this indicates a difference in $\tilde{\varepsilon}$ for LCP and RCP. The electric field ($E$) inside the medium can be related to the displacement field ($D$) through the relation

$$D = \tilde{\varepsilon} E. \quad (2.11)$$

For polar MOKE, with applied field (and magnetisation vector) parallel to the propagation direction, the dielectric tensor of a material with cubic symmetry will be of the form [92]

$$\tilde{\varepsilon} = \begin{pmatrix} \tilde{\varepsilon}_{xx} & \tilde{\varepsilon}_{xy} & 0 \\ -\tilde{\varepsilon}_{xy} & \tilde{\varepsilon}_{yy} & 0 \\ 0 & 0 & \tilde{\varepsilon}_{zz} \end{pmatrix} \quad (2.12)$$

where $\tilde{\varepsilon}_{ij}$ (with $i, j = x, y, z$) is a complex number. This holds for a field parallel to any principal axis. Off-diagonal terms in Equation (2.12) have linear field dependence; these are the terms responsible for linear magneto-optic effects (MOKE and the Faraday effect). Diagonal terms are responsible for the optical activity of materials ($\tilde{\varepsilon}_{xx}$ and $\tilde{\varepsilon}_{yy}$ are independent of external field, $\tilde{\varepsilon}_{zz}$ goes as field
squared, but the difference between them is negligible [92]). Off-diagonal terms are far smaller than diagonal terms, a result that means magneto-optic effects are generally very small.

It is conventional also to set the magnetic permeability tensor $\mu(\omega)=1$ for optical frequencies (i.e. there is no induced magnetisation in the material due to the light) [92, 93]. For magneto-optic effects, physical properties can be explained by considering the conductivity tensor $\tilde{\sigma}$ that relates the electric field to induced current

$$ \mathbf{J} = \tilde{\sigma} \mathbf{E}, \quad (2.13) $$

where $\mathbf{J}$ is the current density. The conductivity tensor is easily relatable to the dielectric tensor by

$$ \tilde{\varepsilon} = \varepsilon_0 + \frac{4\pi i}{\omega} \tilde{\sigma}, \quad (2.14) $$

where $\varepsilon_0$ represents the linear response (and the $\frac{4\pi}{\omega}$ is due to the Gaussian units used to define Maxwell’s equations in order to be consistent with the given references). Equation (2.14) therefore demonstrates that either the dielectric tensor or the conductivity tensor may be used interchangeably to qualitatively describe magneto-optic effects.

### 2.2.2 Quantum Mechanical Origin of MOKE

Quantum theory can quantify magneto-optic effects. The interaction of photons with electron spins via the spin-orbit interaction is responsible for linear magneto-optic effects [91, 92, 94]. This statement is justified as follows.

The isolated one-electron Hamiltonian is

$$ \mathcal{H}_0 = \frac{\mathbf{p}^2}{2m} + V(\mathbf{r}) \quad (2.15) $$

where $\mathbf{p} = (\hbar/i)\nabla$ is the electron momentum operator, $m$ is the electron mass, and $V(\mathbf{r})$ is the effective potential of an electron in a crystal.

Magneto-optic effects can be built into the Hamiltonian as [91]

$$ \mathcal{H} = \mathcal{H}_0 + \frac{1}{2m^2c^2} [\nabla V(\mathbf{r}) \times \mathbf{p}] \cdot \mathbf{s} + \frac{e}{mc} \mathbf{A}(\mathbf{r}, t) \cdot \mathbf{p} \quad (2.16) $$

where $\mathbf{s} = (\hbar/2)\mathbf{\sigma}$ is the electron spin operator, $e$ is the electronic charge, and
\( A(\mathbf{r}, t) \) is the vector potential of the electromagnetic field within the crystal. \( \nabla V(\mathbf{r}) \times \mathbf{p} \cdot \mathbf{s} \) is the spin-orbit coupling; the orbital motion of an electron in an electrostatic field causes the spin moment to precess. Immediately from Maxwell’s fourth \((\nabla \times \mathbf{H} = \mathbf{J}_{\text{total}} + \epsilon \partial \mathbf{E} / \partial t \) [2]), one would expect this to lead to an effective current since it links electron motion \((\mathbf{p})\) to its spin \((\mathbf{s})\).

The third term in Equation (2.16) is the interaction between the electron and the electromagnetic field in the crystal (leading to optical activity). Treating this interaction term as a perturbation to the Hamiltonian, an expression for the current density of an electron in the presence of \( A(\mathbf{r}, t) \) may be constructed (see Argyes for the full treatment [91]) as

\[
\mathbf{J}_{\text{total}} = \mathbf{\sigma} \cdot \mathbf{E} + \mathbf{\alpha} \cdot \partial \mathbf{E} / \partial t
\]

(2.17)

which in turn allows one to construct both conductivity \((\mathbf{\sigma})\) and polarisability \((\mathbf{\alpha})\) tensors of a medium.

**Polar MOKE**

With a light beam propagating along the \( z \) axis through a medium with cubic symmetry, the conductivity tensor is obtained from combining Equations (2.14) \& (2.12) and is of the form

\[
\mathbf{\sigma} = \begin{pmatrix}
\sigma_{xx} & \sigma_{xy} & 0 \\
-\sigma_{xy} & \sigma_{xx} & 0 \\
0 & 0 & \sigma_{xx}
\end{pmatrix}
\]

(2.18)

in a Cartesian basis. In a circular basis, the conductivity tensor will be diagonalised and of the form

\[
\mathbf{\sigma} = \begin{pmatrix}
\sigma_{xx} + i\sigma_{xy} & 0 & 0 \\
0 & \sigma_{xx} - i\sigma_{xy} & 0 \\
0 & 0 & \sigma_{xx}
\end{pmatrix},
\]

(2.19)

where the diagonal elements \( \sigma_{xx} \pm i\sigma_{xy} \) are components in the left and right circular basis respectively. When thinking of magneto-optical effects, it is natural therefore to use this circular basis which represent the normal modes of propagation.
Maxwell’s equations can be combined with Equation (2.17) to calculate all magneto-optic effects [91]. For plane polarised light at normal incidence to a magnetised sample, the reflected beam can be described in terms of the differences in birefringence of left ($n_l$) and right ($n_r$) circular polarisations [91]:

\[
\theta_K \approx -3 \left[ \frac{(n_r - n_l)}{(n_r n_l - 1)} \right], \quad \text{and} \quad \epsilon_K \approx -\Re \left[ \frac{(n_r - n_l)}{(n_r n_l - 1)} \right] \tag{2.20}
\]

where the Kerr angle ($\theta_K$) represents the imaginary component, and ellipticity ($\epsilon_K$) corresponds to the real component. It should be noted that these equations are approximations and only valid for small $n_r$ or $n_l$ (i.e. $|n_r - n_l| << n_r, n_l$), where they may be defined in terms of the permittivity as

\[
n^2_{r,l} = \epsilon_{xx} \pm i \epsilon_{xy} \tag{2.21}
\]

Thus, it is a difference in refractive index for RCP and LCP that leads to a phase/amplitude shift and, consequently, a change in the linear and elliptical polarisation state.

The complex Kerr rotation for polar incidence may be written in terms of the conductivity by substituting Equation (2.21) into Equation (2.20), and using Equation (2.14) to rearrange, as [28]

\[
\theta_K(\omega) + i \epsilon_K(\omega) \approx \frac{-\epsilon_{xy}(\omega)}{\sqrt{\epsilon_{xx}(\omega)(\epsilon_{xx}(\omega) - 1)}} = \frac{-\sigma_{xy}(\omega)}{\sigma_{xx}(\omega) \sqrt{1 + \frac{4 \pi i}{\omega} \sigma_{xx}(\omega)}} \tag{2.22}
\]

Calculations of the polar MOKE response of a material therefore boils down to calculating $\sigma_{xy}$, the off-diagonal conductivity term.

Equation (2.22) holds for polarised light at normal incidence. To generalise the calculation for the polar contribution to the complex Kerr signal at an arbitrary incidence angle ($\theta_i$) to the sample surface needs only a cosine term multiplied to the off-diagonal component of the permittivity:

\[
n^2_{r,l} = \epsilon_{xx} \pm i \epsilon_{xy} \cos(\theta_i) \tag{2.23}
\]

This is correct to first-order in $\epsilon_{xy}$, and leads Equation (2.22) to be expressed as

\[
\theta_K(\omega) + i \epsilon_K(\omega) \approx \frac{-\sigma_{xy}(\omega) \cos(\theta_i)}{\sigma_{xx}(\omega) \sqrt{1 + \frac{4 \pi i}{\omega} \sigma_{xx}(\omega)}} \tag{2.24}
\]
which similarly is correct to first order.

**Longitudinal MOKE**

The general theory for calculating magneto-optic terms for an arbitrary angle of incidence was originally carried out by Hunt [95]. This theory only considers terms that vary linearly with magnetisation (linear in $\epsilon_{xy}$) however, it is usually accurate for cubic systems.

The relations were further simplified by You & Shin [96] using Snell’s law and symmetry arguments. The analytical expressions derived are a product of two main factors: the normal polar Kerr rotation, and a function of optical parameters describing the system.

The basic derivation considers light in a non-magnetic medium with refractive index $n_1$ incident at an angle $\theta_i$ to a magnetic medium with refractive index $n_2$ and a refraction angle $\theta_t$. Solutions to Maxwell’s equations using Equation (2.12) allows the magneto-optic Fresnel reflection matrix to be defined as

$$
\begin{pmatrix}
E_s^r \\
E_p^r
\end{pmatrix}
= 
\begin{pmatrix}
r_{ss} & r_{sp} \\
r_{ps} & r_{pp}
\end{pmatrix}
\cdot 
\begin{pmatrix}
E_s^i \\
E_p^i
\end{pmatrix}
$$

(2.25)

where $r_{ij}$ are the magneto-optic Fresnel reflection coefficients and represent the ratio of incident ($E_i$) to reflected ($E_r$) polarised electric field components. A general form for each $r_{ij}$ component is defined elsewhere [96]. The separate Kerr rotations for s- and p-polarisation are defined as

$$
\theta^s_K = \frac{r_{ps}}{r_{ss}}, \quad \& \quad \theta^p_K = \frac{r_{sp}}{r_{pp}}.
$$

(2.26)

The total Kerr rotation for an arbitrary mix of s- and p-polarisations therefore, will be a weighted sum of each component. Subbing the magneto-optic Fresnel coefficients into Equation (2.26), the polar Kerr rotations are [96]

$$
\theta^s_K = -\frac{\cos(\theta_i) \tan(\theta_i)}{\cos(\theta_i - \theta_t)} \frac{i n_1 n_2 Q}{(n_2^2 - n_1^2)}
$$

(2.27a)

$$
\theta^p_K = \frac{\cos(\theta_i) \tan(\theta_i)}{\cos(\theta_i + \theta_t)} \frac{i n_1 n_2 Q}{(n_2^2 - n_1^2)}
$$

(2.27b)

where $Q = i \epsilon_{xy} / \epsilon_{xx}$ is the Voigt parameter. It should be noted that Equations
(2.27a) & (2.27b) are the product of two terms: the first half is reliant on the incidence and refraction angles, and the second half is the normal polar Kerr signal. Hence, when calculating the magneto-optic response for an arbitrary angle of incidence, it is convenient to first calculate the polar response, and then generalise this with a correction term.

2.2.3 Plasma Frequency

The dependence of frequency in Equation (2.22) means care must be taken when interpreting polarised light experiments. Ellipsometry is the field of study that is focused on the frequency dependence of the polarisation state of reflected light. The measured spectra can give information on optical parameters and is commonly used for determining material thickness by inserting obtained energy transitions into dispersion models [13]. This is partly separated from magneto-optic measurements which want to reveal the magnetic effect on the reflected polarisation state. Hence, for magneto-optic studies, the effect of an external magnetic field must be isolated from non-magnetic contributions to any change in polarisation state for a specific wavelength of light.

As the reflection of light is an absorption-emission process (this mechanism is the electric dipole transition), both interband as well as an intraband transitions must be accounted for. An external magnetic field can alter the electronic structure leading to changes in the allowed transitions and manifesting as a magneto-optic effect.

For incident $\omega < \omega_{\text{plasma}}$, the electromagnetic wave is damped and so heavily absorbed, and for $\omega > \omega_{\text{plasma}}$ the wave propagates through the material and so the reflectivity goes to zero. The intraband conductivity follows a Drude form

$$\sigma_{\text{intra}}(\omega) = \frac{\sigma_0}{1 - i\omega\tau}$$

with a characteristic material-dependent decay constant $\tau$. This $\tau$ represents the finite lifetime effects of the absorption-emission process. In ferromagnets, the intraband contribution to off-diagonal terms of $\sigma(\omega)$ is negligible compared to interband transitions [28]. Only when calculating the spectra for non-magnetic metals are both intraband and interband contributions taken [97]. Particularly for calculations of magneto-optic spectra, the relevant allowed transitions are vital.
Tuning the probing frequency can lead to an enhancement of the magneto-optic signal if the plasma edge in optical reflectivity is steep. This even occurs in paramagnetic materials [28]. Figure (2.7) shows a classic resonance curve for the polar Kerr rotation $\theta_K(\omega)$ and ellipticity $\epsilon_K(\omega)$ from Ag. $\theta_K(\omega)$ follows the standard real component curve shape, rising as frequency increases towards resonance before passing through zero and mirroring the behaviour for higher frequencies. $\epsilon_K(\omega)$ then follows the standard imaginary component shape, displaying a peak at the resonance frequency. For Ag, the plasma edge falls in the UV region at 3.8eV (325nm).

Figure 2.7  The polar Kerr rotation $\theta_K$ and ellipticity $\epsilon_K$ for Ag in a field of 1T, showing the plasma edge at 3.8eV (325nm). From [28, 98].

2.2.4 Penetration Depth

An important parameter to be aware of when investigating materials through optical methods is the penetration depth ($\delta_p$). Defined as [99]

$$\delta_p = \frac{1}{\alpha} = \frac{\lambda}{4\pi \kappa},$$

(2.29)

where $\alpha$ is the extinction coefficient, $\lambda$ the wavelength of light, and $\kappa$ the complex index of refraction, the penetration depth is the distance light travels through a material before the intensity has fallen to $1/e$ of its original value.

Any magneto-optic measurements taken are therefore only sensitive to the surface
layer of the material, where the surface layer thickness may be taken as $\delta_p$. As an example calculation, light of wavelength 633nm reflected from a silver surface with extinction coefficient $\kappa=4.2776$ [100] will penetrate to a depth of $\approx 12$nm. Typically for metals therefore, magneto-optic measurements are sensitive to a thickness of hundreds of atoms.

### 2.3 Polarisation Fundamentals

#### 2.3.1 Stokes Parameters

In order to investigate the magneto-optic properties of a material, the polarisation state of reflected light must be fully characterised. Introduced by Sir George Gabriel Stokes in 1852, the Stokes parameters [101] fully describe the polarisation state of any light beam, whether totally or partially polarised. Experimentally, characterising polarised light therefore comes down to measuring the Stokes parameters.

Often represented by a four-component Stokes vector ($S$), the Stokes parameters consist entirely of observables - intensities - and can be compiled as [102]

$$ S = \begin{pmatrix} I \\ Q \\ U \\ V \end{pmatrix} = \begin{pmatrix} I_x + I_y \\ I_x - I_y \\ I_{+\pi/4} - I_{-\pi/4} \\ I_{RCP} - I_{LCP} \end{pmatrix} = \begin{pmatrix} E_{0x}^2 + E_{0y}^2 \\ E_{0x}^2 - E_{0y}^2 \\ 2E_{0x}E_{0y}\cos(\delta) \\ 2E_{0x}E_{0y}\sin(\delta) \end{pmatrix}, \quad (2.30) $$

where $I$, $Q$, $U$, and $V$ are the four Stokes parameters, $E_{0x}$ and $E_{0y}$ are the amplitudes of the electric field in $x$ and $y$ directions, and the difference in phase between $x$ and $y$ components of electric field is $\delta=\delta_x-\delta_y$. $I_{x,y,\pm\pi/4,RCP,LCP}$ are the intensities of light in the $x$, $y$, $\pm\pi/4$ to the $x$ axis, right, and left circular directions respectively.

The physical significance behind each parameter is as follows: $I$ is the total intensity of a light beam, $Q$ is the difference between linear polarisation in the horizontal and vertical directions (the $x$ and $y$ directions), $U$ is the difference between linear polarisation in $\pm\pi/4$ directions to the $x$ axis, and $V$ is the difference between right and left circular polarisation.
Due to the Cauchy-Schwarz inequality [103], the expression

\[ I^2 \geq Q^2 + U^2 + V^2 \]  

holds, with a light beam totally polarised for the case where the equality stands, and partially polarised if \( I^2 > Q^2 + U^2 + V^2 \).

The orientation of linear polarisation, the Kerr rotation \( \theta_K \), is defined as [102]

\[ \theta_K = \frac{1}{2} \tan^{-1}\left(\frac{U}{Q}\right). \]  

(2.32)

If totally polarised, the ellipticity, or fraction of circular polarisation, is calculated from

\[ \epsilon_K = \frac{1}{2} \sin^{-1}\left(\frac{V}{I}\right). \]  

(2.33)

Both \( \theta_K \) and \( \epsilon_K \) can be visualised with an elliptical polarisation state according to Figure (1.1a). \( \theta_K \) is the angle between the \( x \) axis and the major axis of the polarisation ellipse; \( \epsilon_K \) is the angle subtended between the major axis and the point that the minor axis meets the ellipse. \( \theta_K \) and \( \epsilon_K \) may take any value between \( \pm \pi/4 \).

As \( S \) is composed only of intensities, they are directly measurable, and so convenient quantities to use. For any number of light waves, the Stokes parameters may be used to find the resultant superposition, so long as there is no fixed amplitude or phase relations between them. When there are amplitude variations, Jones calculus is usually more convenient [102]. Jones calculus can deal with interference effects when there is an amplitude or phase relation between beams; the Stokes method cannot account for interference. Jones vectors involve complex quantities however, and can not be used to simply describe light that is not fully polarised. So, for a single monochromatic light source traversing a single path (i.e. not being separated and recombined) as is used in this thesis, Stokes vectors are preferred.

### 2.3.2 Mueller Matrices

When a light beam passes through or reflects from an optical element, the polarisation state may be affected. Changes to the Stokes vector from \( S \) to \( S' \)
may be described by a 4-by-4 matrix, the Mueller matrix, defined by the equation $S' = M \cdot S$, where

$$M = \begin{pmatrix}
m_{00} & m_{01} & m_{02} & m_{03} \\
m_{10} & m_{11} & m_{12} & m_{13} \\
m_{20} & m_{21} & m_{22} & m_{23} \\
m_{30} & m_{31} & m_{32} & m_{33}
\end{pmatrix}. \quad (2.34)$$

The effect of passing through/reflecting from multiple optical elements on the polarisation state of incident light $S$, may then be calculated from

$$S' = M_B \cdot M_A \cdot S \quad (2.35)$$

where the first element $A$ has corresponding Mueller matrix $M_A$, and second element $B$ has Mueller matrix $M_B$. Polarisation detection methods are conceived from such calculations.

### 2.4 Polarisation Detection

#### 2.4.1 Dual Photoelastic Modulator (PEM) Detection

The following outlines the calculation that provides the theoretical basis for measuring the Stokes parameters using a dual PEM and analyser system. The calculation uses Mueller calculus and has been carried out previously [22] as well as having error analysis applied [23, 104]. Arbitrarily polarised light (initially $S = (I, Q, U, V)^T$) passes through two PEMs (the first is orientated at an angle $\alpha$, the second at angle $\beta$, to the optic axis) followed by an analyser (at angle $\gamma$) before reaching a detector as illustrated in Figure (2.8).

The Mueller matrix calculation follows according to

$$S' = M_{ANALYSER} \cdot M_{PEM2} \cdot M_{PEM1} \cdot S. \quad (2.36)$$

The detector is sensitive only to the $I'$ component, the final total intensity
The detection setup consisting of two PEMs and a linear polariser orientated at angles $\alpha$, $\beta$, and $\gamma$ relative to incoming light.

Reaching the detector (in a simplified form):

$$I' = \frac{1}{2} \left( \sqrt{Q^2 + U^2 + V^2} ight. \right.$$

$$- \sin(2(\beta - \gamma)) \cos(\Delta_2) \left[ \cos(2(\alpha - \beta)) \cos(\Delta_1) (U \cos(2\alpha) - Q \sin(2\alpha)) + (Q \cos(2\alpha) + U \sin(2\alpha)) \sin(2(\alpha - \beta)) + V \cos(2(\alpha - \beta)) \sin(\Delta_1) \right]$$

$$+ \cos(2(\beta - \gamma)) \left[ \cos(2(\alpha - \beta)) (Q \cos(2\alpha) + U \sin(2\alpha)) + \cos(\Delta_1) (-U \cos(2\alpha) + Q \sin(2\alpha)) \sin(2(\alpha - \beta)) - V \cos(2(\alpha - \beta)) \sin(\Delta_1) \right]$$

$$- \sin(2(\beta - \gamma)) \sin(\Delta_2) \left[ V \cos(\Delta_1) + (Q \sin(2\alpha)) \sin(\Delta_1) - U \cos(2\alpha) \right] \right).$$

$$I' = \frac{1}{2} \left( \sqrt{Q^2 + U^2 + V^2} \right.$$

$$- \sin(2(\beta - \gamma)) \cos(\Delta_2) \left[ \cos(2(\alpha - \beta)) \cos(\Delta_1) (U \cos(2\alpha) - Q \sin(2\alpha)) + (Q \cos(2\alpha) + U \sin(2\alpha)) \sin(2(\alpha - \beta)) + V \cos(2(\alpha - \beta)) \sin(\Delta_1) \right]$$

$$+ \cos(2(\beta - \gamma)) \left[ \cos(2(\alpha - \beta)) (Q \cos(2\alpha) + U \sin(2\alpha)) + \cos(\Delta_1) (-U \cos(2\alpha) + Q \sin(2\alpha)) \sin(2(\alpha - \beta)) - V \cos(2(\alpha - \beta)) \sin(\Delta_1) \right]$$

$$- \sin(2(\beta - \gamma)) \sin(\Delta_2) \left[ V \cos(\Delta_1) + (Q \sin(2\alpha)) \sin(\Delta_1) - U \cos(2\alpha) \right] \right).$$

(2.37)

Here, $\Delta_1$ and $\Delta_2$ are the PEM modulations, of the form $\Delta = \Delta_0 \sin(\Omega t)$, at frequency $\Omega$ and with amplitude $\Delta_0$. $I'$ is a combination of DC and AC terms, the latter of which varies at select frequencies according to the retardation frequencies $\Omega_1$, $\Omega_2$. As such, a phase-sensitive measurement may be used to discern these rapidly varying AC signals.

Using the Jacobi-Anger expansion [103]

$$\sum_{n=-\infty}^{\infty} i^n e^{i\theta n} J_n(z) = e^{iz \cos(\theta)}$$

(2.38)
sine and cosine terms in equation (2.37) may be expanded out as Bessel functions of the first kind \((J_n, \text{ with integer } n)\) to give

\[
\begin{align*}
\cos(\Delta) &= \cos(\Delta_0 \sin(\Omega t)) = J_0(\Delta_0) + 2J_2(\Delta_0 \cos(2\Omega t) + \ldots \quad (2.39a) \\
\sin(\Delta) &= \sin(\Delta_0 \sin(\Omega t)) = 2J_1(\Delta_0) \sin(\Omega t) + 2J_3(\Delta_0) \sin(3\Omega t) + \ldots \quad (2.39b)
\end{align*}
\]

The expansions, Equations (2.39a) & (2.39b), can be substituted in for sine and cosine terms in Equation (2.37) to give

\[
\begin{align*}
\Gamma' &= \frac{1}{2} \left( \sqrt{Q^2 + U^2 + V^2} \right. \\
&- \sin(2(\beta - \gamma))(J_0(\Delta_{20}) + 2J_2(\Delta_{20}) \cos(2\omega t))(cos(2(\alpha - \beta))(J_0(\Delta_{10}) + 2J_2(\Delta_{10}) \cos(2\omega t)) \\
&\quad \times (U \cos(2\alpha) - Q \sin(2\alpha)) + (Q \cos(2\alpha) + U \sin(2\alpha)) \sin(2(\alpha - \beta)) \\
&\quad + V \cos(2(\alpha - \beta))(2J_1(\Delta_{10}) \sin(\omega t) + 2J_3(\Delta_{10}) \sin(3\omega t)) \right] \\
&+ \cos(2(\beta - \gamma)) \left[ \cos(2(\alpha - \beta))(Q \cos(2\alpha) + U \sin(2\alpha)) + (J_0(\Delta_{10}) + 2J_2(\Delta_{10}) \cos(2\omega t)) \\
&\quad \times (-U \cos(2\alpha) + Q \sin(2\alpha)) \sin(2(\alpha - \beta)) \\
&\quad - V \sin(2(\alpha - \beta))(2J_1(\Delta_{10}) \sin(\omega t) + 2J_3(\Delta_{10}) \sin(3\omega t)) \right] \\
&- \sin(2(\beta - \gamma)) \sin(\Delta_2) \left[ V \left( J_0(\Delta_{10}) + 2J_2(\Delta_{10}) \cos(2\omega t) \right) \\
&\quad + (Q \sin(2\alpha)) \sin(\Delta_1 - U \cos(2\alpha)) \right]. \\
\end{align*}
\]

(2.40)

The detector is sensitive to RMS voltages hence, the \(\sin(\Omega t)\) terms in Equation (2.40) can be evaluated knowing that

\[
\sqrt{\frac{1}{2\pi} \int_{-\pi}^{\pi} \sin^2(\Omega t) \, dt} = \frac{1}{\sqrt{2}}. 
\]

(2.41)
The detected intensity can thus be expressed as

\[
\Gamma = \frac{1}{2} \left( \sqrt{Q^2 + U^2 + V^2} + \cos(2(\beta - \gamma)) \left( \cos(2(\alpha - \beta))(Q \cos(2\alpha) + U \sin(2\alpha)) \right) \\
- \sqrt{2} V J_1 (\Delta_{10}) \sin(2(\alpha - \beta)) \\
+ \sin(2(\alpha - \beta)) \left( J_0 (\Delta_{10}) + \sqrt{2} J_2 (\Delta_{10}) \right) (-U \cos(2\alpha) + Q \sin(2\alpha)) \\
- \sqrt{2} J_1 (\Delta_{20}) \sin(2(\beta - \gamma)) \\
\times \left( V \left( J_0 (\Delta_{10}) + \sqrt{2} J_2 (\Delta_{10}) \right) + \sqrt{2} J_1 (\Delta_{10}) (-U \cos(2\alpha) + Q \sin(2\alpha)) \right) \\
- \sqrt{2} V J_1 (\Delta_{10}) \sin(2(\beta - \gamma)) \cos(2(\alpha - \beta)) \left( J_0 (\Delta_{20}) + \sqrt{2} J_2 (\Delta_{20}) \right) \\
+ \cos(2(\alpha - \beta)) \left( J_0 (\Delta_{10}) + \sqrt{2} J_2 (\Delta_{10}) \right) (U \cos(2\alpha) - Q \sin(2\alpha)) \\
+ (Q \cos(2\alpha) + U \sin(2\alpha)) \sin(2(\alpha - \beta))) \right).
\]

(2.42)

Sorting into terms proportional to \( J_n \)'s and discarding terms that are combinations of multiple \( J_n \)'s provides

\[
\Gamma = \frac{1}{2} \left[ \sqrt{Q^2 + U^2 + V^2} + \cos(2(\beta - \gamma)) \cos(2(\beta - \gamma))(Q \cos(2\alpha) + U \sin(2\alpha)) \\
+ J_0 (\Delta_{10}) \cos(2(\beta - \gamma))(-U \cos(2\alpha) + Q \sin(2\alpha)) \sin(2(\alpha - \beta)) \\
- \sqrt{2} J_1 (\Delta_{10}) V \cos(2(\beta - \gamma)) \sin(2(\alpha - \beta)) \\
+ \sqrt{2} J_2 (\Delta_{10}) \cos(2(\beta - \gamma))(-U \cos(2\alpha) + Q \sin(2\alpha)) \sin(2(\alpha - \beta)) \\
- J_0 (\Delta_{20}) \cos(2(\beta - \gamma)) \sin(2(\alpha - \beta)) \sin(2(\beta - \gamma)) \\
+ J_0 (\Delta_{20}) (Q \cos(2\alpha) + U \sin(2\alpha)) \sin(2(\alpha - \beta)) \sin(2(\beta - \gamma)) \right].
\]

(2.43)

We are free to choose the PEM retardation amplitude \( \Delta_0 \) and hence, the value \( J_n(\Delta_{n0}) \) takes. It is common to choose a value that simplifies Equation (2.43) to reduce the number of components needed to be detected. Figure (2.9) is a plot of the first three Bessel harmonics. A value of \( \Delta_0 = 2.406 \) rad (corresponding to 0.383 \( \lambda \)) zeroes \( J_0(\Delta_0) \), reducing the \( J_0(\Delta_{10}) \) and \( J_0(\Delta_{20}) \) terms in equation (2.43) to zero. This is the value carried forward for the following calculations. Another common choice is \( \Delta_0 = 3.054 \) rad (corresponding to 0.486 \( \lambda \)) which is the point that \( J_2(\Delta_0) \) is most stable.

Invoking \( \Delta_0 = 2.406 \) rad into Equation (2.43) reduces the detected intensity to
the final reduced form:

\[
\Gamma' = \frac{1}{2} \left[ \sqrt{Q^2 + U^2 + V^2 + \cos(2(\alpha - \beta)) \cos(2(\beta - \gamma))} (Q \cos(2\alpha) + U \sin(2\alpha)) 
- \sqrt{2} J_1(\Delta_{10}) V \cos(2(\beta - \gamma)) \sin(2(\alpha - \beta)) 
+ \sqrt{2} J_2(\Delta_{10}) \cos(2(\beta - \gamma)) \left( -U \cos(2\alpha) + Q \sin(2\alpha) \right) \sin(2(\alpha - \beta)) 
- \sqrt{2} J_2(\Delta_{20}) \left( Q \cos(2\alpha) + U \sin(2\alpha) \right) \sin(2(\alpha - \beta)) \sin(2(\beta - \gamma)) \right].
\]

The voltage signals to be measured are a combination of DC and AC terms detected with lock-in amplifiers:

\[
V_{\text{signal}} = V_{\text{DC}} + V_{\omega 1} + V_{2\omega 1} + V_{2\omega 2}
\]

where the subscripts relate to the DC, first, and second harmonics of the AC signal, and the order relates each term to the relevant line in Equation (2.40). So, each of the AC voltage signals are expressed in terms of the initial PEM and polariser parameters, as well as the Stokes parameters:

\[
V_{\omega 1} = -\frac{V}{\sqrt{2}} J_1(\Delta_{10}) \sin \left( 2(\alpha - \beta) \right) \cos \left( 2(\beta - \gamma) \right),
\]

\[
V_{2\omega 1} = +\frac{1}{\sqrt{2}} J_2(\Delta_{10}) \sin \left( 2(\alpha - \beta) \right) \cos \left( 2(\beta - \gamma) \right) \left( Q \sin(2\alpha) - U \cos(2\alpha) \right),
\]

\[
V_{2\omega 2} = -\frac{1}{\sqrt{2}} J_2(\Delta_{20}) \sin \left( 2(\alpha - \beta) \right) \sin \left( 2(\beta - \gamma) \right) \left( Q \cos(2\alpha) + U \sin(2\alpha) \right).
\]

It is desirable to maximise all three measured voltage signals so that the signal to electronic noise ratio is as large as possible. The common feature between
the three AC voltages is the sin \((2(\alpha - \beta))\) term. To maximise this requires \(\alpha - \beta = \pi/4\); the PEMs should be set so that their modulated axes are \(\pi/4\) relative to each other. Both Equations (2.46a) & (2.46b) share the cos \((2(\beta - \gamma))\) term however, Equation (2.46c) is orthogonal to this. In order to maximise all three equations equally, the sine term should equal the cosine term and so \(\beta - \gamma = \pi/8\).

Combining these choices, if we choose our reference axis aligned to the retardation axis of the first PEM, then we impose \(\alpha = 0\) which implies \(\beta = \pi/4\), and \(\gamma = \pi/8\).

With these three conditions satisfied we can rewrite Equations (2.46a)-(2.46c) to express each AC voltage signal in terms of each of the Stokes parameters as

\[
V_{1\omega 1} = -\frac{V}{2} J_1(\Delta_{10}) \quad \text{or} \quad V = -\frac{2V_{1\omega 1}}{J_1(\Delta_{10})} \quad \text{(2.47a)}
\]

\[
V_{2\omega 1} = -\frac{U}{2} J_2(\Delta_{10}) \quad \text{or} \quad U = -\frac{2V_{2\omega 1}}{J_2(\Delta_{10})} \quad \text{(2.47b)}
\]

\[
V_{2\omega 2} = -\frac{Q}{2} J_2(\Delta_{20}) \quad \text{or} \quad Q = -\frac{2V_{2\omega 2}}{J_2(\Delta_{20})} \quad \text{(2.47c)}
\]

Additionally, the DC signal will be given as

\[
V_{DC} = \frac{1}{2} \sqrt{Q^2 + U^2 + V^2}. \quad \text{(2.48)}
\]

From the Stokes parameters, the rotation of the major axis of the polarisation ellipse, the Kerr rotation, is defined as Equation (2.32) and the ellipticity angle is given by Equation (2.33). Or, in terms of the measured voltages, and linking Equations (2.47a)-(2.47c) to Equation (2.32), the Kerr rotation is

\[
\theta_K = \frac{1}{2} \tan^{-1} \left( \frac{V_{2\omega 2} J_2(\Delta_{10})}{V_{2\omega 1} J_2(\Delta_{20})} \right), \quad \text{(2.49)}
\]

and the ellipticity angle is

\[
\epsilon_K = \frac{1}{2} \sin^{-1} \left( \frac{V_{\omega 1}}{J_1(\Delta_{10}) \sqrt{\left(\frac{V_{\omega 1}}{J_1(\Delta_{10})}\right)^2 + \left(\frac{V_{2\omega 1}}{J_2(\Delta_{10})}\right)^2 + \left(\frac{V_{2\omega 2}}{J_2(\Delta_{20})}\right)^2}} \right). \quad \text{(2.50)}
\]

Hence, the voltages required to calculate both \(\theta_K\) and \(\epsilon_K\) are the three AC terms outlined in Equation (2.46a-c).
Stokes Parameters of a Rotated Waveplate

The unique ability of the dual PEM setup to simultaneously determine the polarisation ellipse major axis angle and ellipticity angle can be demonstrated using a quarter waveplate. Figure (2.10) shows the measured and calculated Stokes parameters when linearly polarised light passes through the waveplate which is rotated from 0 to \( \pi \) rad, as detected by the dual PEM setup. Three AC voltages are measured and converted into the relevant Stokes parameter using Equations (2.47a-c), which were normalised to give a total intensity of unity. The initial polarisation state \((S = (1, 1, 0, 0)^T)\) is realised at angles of 0 & \( \pi \) rad where the waveplate has no effect on the polarisation state. At \( \frac{\pi}{4} \) & \( \frac{3\pi}{4} \), the linearly polarised light is converted to purely circular, resulting in the maximum \( V \) signal, whilst the linear components \( U \) & \( Q \) go to zero. Any other angle of the waveplate results in a mix of the Stokes parameters. Hence, any arbitrary polarisation state may be quantified.

For most materials, it is usually a small rotation upon reflection that is of interest. Figure (2.11) demonstrates the quarter waveplate being rotated about 0 rad in increments corresponding to the smallest gradation on the goniometer controlling the waveplate angle: \( \sim 1.5 \) mrad. The linear \( Q \) component here is close to constant, and the \( U \) & \( V \) components cross zero at the same angle (within the best achievable waveplate angle accuracy), illustrating the correct zero position of the waveplate. This demonstrates how the voltage signal corresponding to the \( U \) component is more significant than the \( Q \) component when calculating the Kerr angle (Equation (2.49)), and similarly the \( V \) component is most important when calculating the ellipticity angle (Equation (2.50)).
Figure 2.10  Measured Stokes parameters of linearly polarised light transmitted through a quarter waveplate that is coarsely rotated $0 - \pi$ rad.

Figure 2.11  Measured Stokes parameters of linearly polarised light transmitted through a quarter waveplate that is finely rotated about 0 rad. The $Q$ signal is reduced to 5% of its magnitude for clarity.
Chapter 3

Experimental Development

This chapter details the development of the MOKE measurement setup. The full optical layout is explained, and the selection and placement of each component is detailed. Following this the electronic circuit is described with reference to measured signal noise and stability. Finally, the imaging techniques used to maintain image focus and accurately build domain images are outlined.

3.1 Optical Layout

The optical layout is illustrated in Figure (3.1). There are two light paths: the 633nm wavelength laser diode produces red light which is used for the MOKE measurement, and the 565nm LED produces green light which is used for imaging. All optics are mounted on the breadboard (Thorlabs MB3060/M) and are mechanically coupled using a Thorlabs 30mm cage system (apart from the PEMs which are independently bound). The optics are thermally regulated as detailed in Section (3.2).

3.1.1 MOKE Measurement

Following the red light path in Figure (3.1), light is produced by a 5mW laser diode (Thorlabs HL6312G) held in an isolated thermally regulated enclosure (TCLDM9). The diverging red light is collected by a close lens (f=2.5mm), focused through a 25µm pinhole, and collimated by a second lens (f=25mm).
This lens-pinhole-lens system acts as a spatial filter to ‘clean up’ light from the laser diode by removing unwanted high-intensity fringes and transforming the intensity distribution into a Gaussian. The output beam diameter is controlled by the choice of focal lengths of the two lenses (typically 120µm for this setup). A remote-controlled shutter (SH1/M) stops red light if required for imaging purposes.

The now collimated beam is reflected from two silver mirrors (PF10-03-P01) and passed through the initial Glan-Thomson polariser (GTH10M-A) to generate linearly polarised light. After passing through the first beamsplitter (BSW10R), light exits the thermally regulated box and is incident on the sample mount that houses the 40× magnification objective lens (LMM-40X-P01) and sample. The angle of incidence at which light is focused onto the sample is controlled by the position light is incident on the objective lens.

After reflecting from the sample, light is re-collimated by the reflective objective lens and re-enters the thermally regulated box. Light is reflected from the first beamsplitter, passes through a long focal length lens (f=200mm) and reaches the second beam splitter (BSS10R), splitting light intensity into 70:30 transmitted:reflected ratio. Reflected light travels to another lens and the CMOS...
sensor (DCC1645C), forming the image. Transmitted light now passes through the detection system detailed in Section (2.4.1), consisting of two PEMs (Hinds PEM-100) and a linear polariser (GTH10M-A). Finally light is passed through a dichroic filter (FL05635-10) and lens (f=25mm) to be focused onto the photodiode (SM05PD2A).

The sample is mounted onto a cold finger on top of three Attocube transducers (two ANPx101 and one ANPz101) to control the $x$, $y$, $z$ position of focus on the sample. An additional Attocube rotator (ANR101) to control the sample orientation may also be included. The objective lens is separately mounted to the cold finger to keep its position fixed relative to the optics board and the sample. Figure (3.2) is a photograph of this mount, showing the brass arms which are attached to the cold finger by threading into the bottom brass block. The sample-objective lens mount may be raised or lowered to place it in line with the lower vertical electromagnet (for polar MOKE measurements) and the higher horizontal cored electromagnets (for longitudinal or transverse MOKE measurements) depicted in Figure (3.1). Additionally, the optics board is held on a separated manual $x$, $y$ translator to align the light beam with the objective lens and control the incidence plane of light focused onto the sample.

### 3.1.2 Imaging

The green optical path in Figure (3.1) represents diffuse light used for forming a live image. Green light originates from the 565nm LED (Thorlabs M565L3) and
is focused by collector and field lenses. Light is then passed through an aperture acting as the field diaphragm and reflected from a dichroic mirror (DMLP605). The green imaging light then follows the same path as the red measurement light, being reflected from the sample and the second beam splitter to form an image on the CMOS sensor. The transmitted light is prevented from reaching the photodiode with a dichroic filter, although in practice the LED is also switched off during the measurement phase to prevent the small fraction of light leakage.

**Spatial Resolution**

The spatial resolution of the red probing light is determined by the objective lens and the width of the laser beam incident upon the objective. The numerical aperture (NA) of an objective lens defines the range of incidence angles the lens can focus light to, and is given by

\[
NA = n \sin (\theta) = n \sin \left( \arctan \left( \frac{D}{2f} \right) \right)
\]

(3.1)

with \(n\) being the refractive index of surrounding medium (air), \(\theta\) the half angle of the cone of focused light, \(D\) is the width of the incoming beam, and \(f\) is the focal length of the lens used. These parameters are illustrated in Figure (3.3), showing the green light using the full entrance pupil of the lens \(D\).

If the incident light only fills part of the objective lens diameter, as for the red

![Figure 3.3](image-url)  
**Figure 3.3** Definition of numerical aperture (NA) from the half angle cone of focused light \(\theta\), the width of the used entrance pupil \((D\) for green light, \(d\) for red light), and the focal length of the lens \(f\).
Figure 3.4  Measured 635nm red laser beam profile directly before the objective lens.

Figure 3.5  Measured focused 635nm red laser beam profile that sets the resolution limit for the MOKE measurement system.

light path in Figure (3.3), the effective NA is less than that defined by the lens. From equation (3.1), $D$ is replaced by the smaller beam width $d$, and as $\frac{d}{f} < \frac{D}{f}$, the numerical aperture is decreased. This reduced diameter limits the obtainable resolution.

The resolution may be quantified using the Rayleigh criterion, defined as the distance between the overlap of two Airy disks, each centred on the first minimum of the other [99]. The contrast between the peaks of the two Airy disks is such that both may be resolved. For this to occur there must be a dip of at least 26% of each peak’s intensity separating them. The Rayleigh criterion is expressed as the distance

$$r = \frac{0.61\lambda}{\text{NA}}. \quad (3.2)$$

For the reflective objective used with a NA of 0.5, the best obtainable resolution with 635nm light is $r = 0.77\mu$m.
The collimated red laser beam has a beam width of \( \approx 120 \mu m \). This is shown in Figure (3.4) - the beam profile measured with a CMOS sensor placed before the objective lens. The Airy disk can be observed (the first minimum is non-zero possibly due to blooming of the pixels from the high intensity of light although it may also be an accurate representation).

This laser beam is focused by the objective lens to a spot which is similarly imaged by a CMOS sensor in Figure (3.5). This focused spot has a clear minimum, and the FWHM is \( 4.10 \mu m \), far larger than the best obtainable resolution of the objective lens (0.77\( \mu m \)). This is the resolution of the MOKE measurements detailed in this thesis and may be reduced with the choice of a higher NA objective lens and/or expanding the laser beam diameter before focusing.

### 3.1.3 Reflective Objective Lens

The objective lens is perhaps the most important component in an optical setup, as it is this that determines the field of view and resolution of the projected image. Using polarised light further complicates the choice, as the objective lens must have as small an effect on the polarisation as possible. There are two types of lenses one can choose: refractive (glass) or reflective (mirror) lenses.

For refractive lenses, it is usual to choose a specifically manufactured strain-free polarised light objective. These objectives limit the number of micro-lens components within the objective in addition to using “strain-free” glass and spacers to hold the components in fixed positions. Such lenses, however, may still be made of glass that can have a measurable Faraday rotation, requiring a careful calibration procedure to separate the effect of the lens from that of the sample.

Reflective objectives function like a standard Cassegrain reflector telescope, with light reflecting from two polished surfaces between the object and the observer. Figure (3.6) is a diagram of a reflective objective, showing the primary and secondary mirrors used to focus light, and the light path (red dashed line). Reflective objectives have the advantage of easier manufacture, since polishing a glass surface (which acts as a substrate for the mirror) is easier than forming a complete lens. This mirror material is typically a reflective metal such as aluminium, silver, or gold. These lenses have the benefit of no chromatic aberrations (unlike refracting glass lenses) and are commonly used for high laser
power applications. With reflective objectives however, there is a fractional obscuration from the spider mounts used to hold the primary mirror in place, which typically reduces the overall image intensity by $\approx 20\%$. A reflective objective is used for all MOKE measurements in this thesis unless otherwise stated.

Offsets in Measured Kerr and Ellipticity Angles

For a MOKE measurement system, changes in the polarisation state of light must be separated into components of non-magnetic origin, e.g. linear birefringence, and components of magnetic origin. When passing through or reflecting from any optical component, it is usual for the state of polarisation of light to change unless the polarisation state is carefully maintained.

For the current system, the incidence angle of light upon a sample is determined by the objective lens used. From Equation (3.1), the cone of incidence half angle is controlled by the NA of the objective lens. For an incident beam with width less than the diameter of the objective lens, the incidence angle $\theta_i$ is controlled by the position upon which the beam enters the objective.

Due to different reflection coefficients for s- and p-polarised light, reflective objectives induce both a slight rotation of the plane of polarisation as well as a slight ellipticity for any beam that is not perfectly s- or p-polarised. This arises due to a difference in reflection amplitude and reflection phase between Fresnel’s
$r_s$ and $r_p$ coefficients, defined as [13]

$$
\begin{align*}
    r_s &= \frac{n_1 \cos(\theta_i) - (n_2 + ik_2) \sqrt{1 - \frac{n_1^2 \sin^2(\theta_i)}{(n_2 + ik_2)^2}}}{n_1 \cos(\theta_i) + (n_2 + ik_2) \sqrt{1 - \frac{n_1^2 \sin^2(\theta_i)}{(n_2 + ik_2)^2}}} \\
    r_p &= \frac{n_1 \sqrt{1 - \frac{n_1^2 \sin^2(\theta_i)}{(n_2 + ik_2)^2}} - (n_2 + ik_2) \cos(\theta_i)}{n_1 \sqrt{1 - \frac{n_1^2 \sin^2(\theta_i)}{(n_2 + ik_2)^2}} + (n_2 + ik_2) \cos(\theta_i)},
\end{align*}
$$

(3.3a)

(3.3b)

where both have been expressed in terms of the real ($n_{1,2}$) and imaginary ($k_{1,2}$) parts of the complex refractive index of the reflecting material as well as the angle of incidence ($\theta_i$). It is assumed that air (medium 1) has $k_1 = 0$.

In terms of the above Fresnel reflection coefficients, the polarisation state of light may be described by the ellipsometric parameters [106]

$$
\frac{r_p}{r_s} = \tan(\psi) \exp^i \Delta,
$$

(3.4)

where

$$
\psi = \tan^{-1} \left[ \frac{|r_p|}{|r_s|} \right]
$$

(3.5)

represents the amplitude ratio and

$$
\Delta = \arg[r_p] - \arg[r_s]
$$

(3.6)

represents the phase change of p- and s-polarised waves upon reflection. These ellipsometric parameters may be re-expressed to give the MOKE parameters [106] defining the polarisation ellipse relative to the optic axis, as illustrated in Figure (1.1a): the Kerr angle

$$
\theta_K = \tan^{-1} \left[ \frac{\sin(2\psi) \cos(\Delta)}{\sqrt{1 - \sin^2(2\psi) \sin^2(\Delta) - \cos(2\psi)}} \right],
$$

(3.7)

and the ellipticity angle

$$
\epsilon_K = \frac{1}{2} \tan^{-1} \left[ \frac{\sin(2\psi) \sin(\Delta)}{\sqrt{1 - \sin^2(2\psi) \sin^2(\Delta)}} \right].
$$

(3.8)

If polarised light that is not perfectly s- or p-polarised is reflected from an optical component at an angle other than normal incidence, there will be an induced
rotation and ellipticity, with parameters described by Equations (3.7) & (3.8). Efforts must be dedicated to minimise any error in the initial polarisation state generated. For a measurement setup, this effect on the polarisation state is a constant offset to the measured angles $\theta_K$ and $\epsilon_K$ and is independent of any magnetic behaviour.

When reflected from a silver mirror ($n_2=0.157$, $k_2=3.82$) in a perfect vacuum ($n_1=1$, $k_1=0$), light ($\lambda=635$nm) that is initially 99% s-polarised and 1% p-polarised results in a rotation of the plane of polarisation and a larger induced ellipticity angle, as illustrated in Figure (3.7). This is the case for any incidence angle $\neq 90^\circ$. Reflecting from a silver mirror at 45° for example leads to $\Delta \theta_K \approx 0.14$ mrad and $\Delta \epsilon_K \approx 3.5$ mrad, which are both significant rotations. These rotations would then propagate through the system, causing larger effective rotations the more interfaces light passes through. This is one reason for minimising the number of optical components placed after the position that the polarisation state of light is generated (the linear polariser).

For the current system, the use of a reflective objective adds a larger offset to the incident $\theta_K$ & $\epsilon_K$. This is due to the reflection of light from two metal surfaces within the objective both before and after reflecting from the sample under question. Using Equations (3.3a-b), the calculated change in $\theta_K$ & $\epsilon_K$ for light incident upon an objective lens is given in Figure (3.8). In these calculations, the initial incident light is linearly polarised along the $y$-axis, and for each $x,y$
position on the plots, the colour scale indicates the effective rotation for $\theta_K$ or $\epsilon_K$ that would occur if light was incident upon that coordinate of the objective lens, with (0,0) being the centre of the objective lens. The input parameters are such that match the entrance diameter and NA of the reflective objective lens used in the MOKE setup. This calculation only considers the rotations due to the incidence angle on the ‘sample’ which is taken as silver. Complex rotations due to reflections within a reflective objective lens are not included in this calculation.

To calculate $\theta_K(x, y)$ in Figure (3.8a), the incidence plane and polarisation state relative to this incidence plane is controlled using polar coordinates such that

$$x = r \cos(\phi), \quad (3.9a)$$
$$y = r \sin(\phi). \quad (3.9b)$$

Here, $r$ is the distance from the centre of the objective. This position determines the incidence angle by $\theta_i = \tan^{-1}(\frac{r}{2f})$ which follows from Equation (3.1) and the geometry of Figure (3.3). This uses a thin lens approximation for the reflective objective. The Kerr rotation is then calculated by weighting Equation (3.7) and the effective $r_s$ and $r_p$ given by Equations (3.3a-b) according to the position of incidence i.e. setting

$$\psi(x, y) = \tan^{-1}\left[\frac{\cos(\phi) |r_p|}{\sin(\phi) |r_s|}\right] = \tan^{-1}\left[\frac{x |r_p|}{y |r_s|}\right]. \quad (3.10)$$

Similarly, the phase shift given by Equation (3.6) is altered to accommodate the position-dependence of the incident polarisation state as

$$\Delta(x, y) = \arg[r \cos(\phi) r_p] - \arg[r \sin(\phi) r_s] = \arg[x r_p] - \arg[y r_s]. \quad (3.11)$$

When plotting, $x$ & $y$ are taken to cover the entrance aperture of the corresponding objective lens resulting in Figure (3.8a). Both Equations (3.10) & (3.11) are also incorporated into Equation (3.8) to produce the ellipticity angle $\epsilon_K(x, y)$ in Figure (3.8b).

Figures (3.8a) & (3.8b) display largely similar behaviour, with the input linearly polarised light changing behaviour along the $x$- and $y$- axis of the lens. As expected, with increasing position from the centre, both $|\theta_K|$ & $|\epsilon_K|$ increase (as the incidence angle is increasing). The induced rotation and ellipticity however, show opposite behaviours i.e. at positions that $\theta_K$ has a large positive rotation,
Calculated polarisation rotation $\theta_K$ (a) and ellipticity $\epsilon_K$ (b) due to incidence position on an NA=0.5 reflective objective lens. Light is originally polarised along the $y$ axis and the incident position is linked to the effective incidence angle and polarisation state upon a silver mirror.

$\epsilon_K$ has a corresponding negative rotation. It should be noted that the calculated offset in ellipticity is far larger than that of the Kerr rotation for identical positions of incidence. In experiments using linearly polarised light along $y$, the incidence position on the objective should be along either the $x$- or $y$-axis for s- or p-polarised measurements. These planes reduce measured offsets as a result of the incidence plane which is separate to any magnetic effects.

In comparison to the above calculated offsets in measured $\theta_K$ & $\epsilon_K$, Figure (3.9) shows the corresponding experimentally measured offsets for the reflective objective. The obscuration due to the three spider legs holding the primary mirror in the objective are visible in the DC voltage as shown in Figure (3.9c). Qualitatively the change in $\theta_K$ as a function of incidence position on the objective lens in Figure (3.9a) is the same for experiment as it was for the calculated result in Figure (3.8a). The magnitude of the experiment is about five times that of the calculation, owing to the additional reflections within the objective lens that have not been calculated.

The measured $\epsilon_K$ offset in Figure (3.9b) similarly shows the same qualitative behaviour as the calculation in Figure (3.8b) though with some discrepancies. The large negative ellipticity at (1.5,-2.0) for example is possibly a result of the
interference with the spider legs of the objective. Overall the general behaviour broadly agrees with the calculation. The magnitude is also similar to that measured in the Kerr rotation, suggesting the reflections within the objective lens can both increase and reduce the ellipticity angle of polarised light.

These measurements show how important alignment of incident light with the planes of the objective lens is. A small offset in position that polarised light is incident upon the objective lens can drastically change the effective incidence polarisation state. Whilst magnetic contributions from a sample during a MOKE measurement should be easily separable from these spurious offsets (e.g. by measuring a change in Kerr or ellipticity angle as a function of field), having a larger ellipticity angle incident upon the sample naturally reduces the linear component of incident polarisation, as the total polarisation state must be conserved. This reduced linear polarisation component will result in a smaller measured Kerr rotation.
Figure 3.9 Measured polarisation rotation $\theta_K$ (a) and ellipticity $\epsilon_K$ (b) due to incidence position on an NA=0.5 reflective objective lens. Incidence light is polarised along the $y$ axis and the incident position is linked to the effective incidence angle and polarisation state upon a silver mirror. The corresponding DC signal (c) shows the three spider legs that obscure light.
3.2 Thermal Stability

The stability of the measurement system is vital. Temperature variations have an affect on various aspects of the detection system, with the most significant being:

- Responsivity of the photodiode
- Output power of the laser diode
- Peak retardation applied by the PEMs
- Physical position of all optical components

Measurements can take several hours when scanning across a sample to produce domain images, or varying the temperature or magnetic field applied to the sample being measured. The measurement system therefore, has to be stable over timescales of at least this duration.

A simple solution is to reduce the effects of drift due to thermal changes in the lab environment. This may be achieved by separately regulating the temperature of the optics relative to the ambient temperature. Whilst the laser diode is separately regulated (as small variations can change the wavelength and power of light produced), the remaining components are thermally regulated together. The optical design detailed above has all components, except the objective lens and sample, mounted onto a single breadboard (300mm×600mm), as illustrated in Figure (3.10). The aluminium breadboard is sandwiched between two 1mm thick copper sheets to promote even heat distribution. On the outer side of the breadboard are mounted four 16.8W thermoelectric Peltier devices (54mm×54mm). The hot side of the Peltiers are thermally connected to the copper plate with an adhesive thermal interface sheet, and the cold sides are connected to heat sinks. The temperature of the breadboard is regulated by heating the four Peltiers with the input power regulated by a 225W controller (TED4015), using PID settings from the temperature measured by the thermometer placed in the centre of the four Peltiers within the aluminium breadboard. This setup can heat the breadboard by over 10°C however, the setpoint is chosen to be 1-2°C above room temperature to reduce the temperature differential between the outside lab environment and the inside.
Figure 3.10  Side view of the temperature regulated optics box, built around an aluminium breadboard and regulated above room temperature with Peltier devices.

An aluminium frame is built around the optics enclosed within the thermally regulated box, and the whole box is enveloped by two layers of 9mm thick (nitrile rubber) insulation sheets. This frame screws directly into the breadboard and the top lid is attached with Velcro to allow easy access. Two cut-outs are made in the frame, one for light to exit the box towards the sample mount and re-enter at the same position after reflecting from the sample, and another for the cables required to power the optical equipment (the LED, laser diode, two PEMs, photodiode, monitoring thermometers). Two monitoring thermometers are placed within the box, and an additional one is placed outside the box to record room temperature variations. The entire frame is mounted to a bracket via four plastic legs to thermally isolate the box. This allows the MOKE measurement system to be adapted to different setups for applying different sample conditions.

The power required to warm the enclosure may be calculated from the total energy needed to heat the Al breadboard, most of the optical equipment (which is Al and glass but assumed mostly Al), and the two copper sheets:

\[
E_{\text{warming}} = \left( \left( c_{Al} \times m_{Al} \right) + \left( c_{Cu} \times m_{Cu} \right) \right) \times \Delta T \\
= \left( \left( 0.9 \frac{kJ}{kgK} \times 13kg \right) + \left( 0.4 \frac{kJ}{kgK} \times 3.5kg \right) \right) \times 2K \\
= 26.2kJ
\]

(3.12)

where the masses and heat capacities of Al and Cu are \( c_{Al,Cu} \) & \( m_{Al,Cu} \).
respectively, and the desired temperature increase is $2K$. Assuming the enclosure is thermally separated from the air surroundings via the nitrile rubber insulation layer with thermal conductivity $k_{nitrile}$, the rate of energy loss will be

$$E_{loss} = k_{nitrile} \times \Delta T \times \frac{A}{l}$$

$$= 0.034 \frac{W}{m \times K} \times 2K \times \frac{0.75m^2}{0.018m}$$

$$= 2.8W$$

(3.13)

where the surface area ($A$) of the insulation is taken as that of the box, of dimension $0.3 \times 0.6 \times 0.25m^3$, and the thickness ($l$) is for two layers of the nitrile rubber. This is neglecting any heat loss through the hole that light exits the enclosure, and the cables which are attached to the various optical devices inside the enclosure. Thus, to heat up the enclosure within an hour requires a power of

$$P_{required} = \frac{E_{warming} + E_{loss} \times t}{t}$$

$$= \frac{26.2kJ + 2.8 \frac{W}{s} \times 3600s}{3600s}$$

$$= 10.1W$$

(3.14)

where $t$ is the time taken to complete the warming. Additionally, a power greater than $E_{loss} = 2.8W$ is required to maintain its temperature. The four Peltiers used have a combined maximum power output of 64W which is more than adequate to warm and maintain the temperature of the enclosure within a reasonable timescale, even if heat loss through the various cables is significant.

The measured thermal performance of the enclosure is shown in Figure (3.11). The regulating thermometer is denoted “Optics Board T” (blue), and is maintained at 22.5°C, controlling the power supplied to the four Peltiers. The room temperature thermometer (green) shows large variations of 0.5°C over the course of 24 hours, as well as a short timescale variation due to air currents from the air conditioning system (indicated by the spread of points). The sample mount (red) is maintained at 21.85°C in air with only a heater coil resulting in blips at times of large temperature change. The “Optics Frame” (magenta) thermometer is mounted inside the box to the Al optics cage system, and shows small variations of 0.025°C in time with the external room temperature changes (the absolute temperature has been shifted in Figure (3.11) to separate it from the regulating temperature). Short timescale variations in temperature due to
Figure 3.11 Thermal stability of the enclosure - see text. The three optics temperatures have been offset for clarity.

Air currents are also eliminated by the enclosure. Similarly, the “Optics Cable” (cyan) is mounted on the entry to the enclosure in thermal contact with required cables. This thermometer shows a larger variation of 0.15°C, highlighting the weak point in the thermal performance of the enclosure. To improve the thermal performance of the enclosure, having the cables thermally insulated would help (however, the PEM cables are matched to the controllers so extra-long versions would be required). Additionally, the room temperature affects the mounting bracket of the optics board (as the optics and sample mount themselves are detached) so overcoming this by maintaining the bracket temperature to reduce thermal expansion effects would help.

The corresponding signal stability to the above thermal stability is illustrated in Figure (3.12). Overall, small jumps in the Kerr signal of 0.03 mrad are observed coincident with room temperature changes. Measurements are usually taken avoiding these two time periods in each day. A small drift is also present which, at its worst, is roughly 1.2 µrad/hour. This drift in $\Delta \theta_K$ is smaller than the average standard deviation from 200 measurements of 12 µrad.
3.3 Signal Noise

This section contains the analysis of the electronic side of the MOKE measurement. The electronic noise calculated is compared with that measured, and the stability of the voltage signals is also discussed.

3.3.1 Electronic Noise

Section (2.4.1) outlined how two PEMs and a linear polariser can accurately quantify the full polarisation state of light. The noise on any calculation of $\theta_K$ and $\epsilon_K$ will have an electronic component, as well as additional components that represent artefacts of the measurement procedure - vibrations or thermal fluctuations of the devices themselves for example.

From Equation (2.49), $\theta_K$ relies upon the second harmonics of high frequency voltage signals from both PEMs and from Equation (2.50) $\epsilon_K$ relies upon these as well as the first harmonic of one PEM. The electronic circuit used for detection is illustrated in Figure (3.13). BNC coaxial cables are used for the entire circuit to shield against external noise sources. The photodiode is negatively biased (with -20V) by a DC voltage source connected through a shielded RC filter to dampen
Figure 3.13 Simplified circuit diagram for the electronics. The RC filter to negatively bias the photodiode, and the pre-amplifier are both separately shielded.

High frequency noise ripples. Current induced on the photodiode by incident light is converted to a voltage signal by the pre-amplifier which is detected by the DC voltmeter and three lock-in amplifiers (routed using T-connectors). The reference frequencies from both PEM controllers are connected to the lock-ins to act as the reference signals. The electronic noise on the high frequency signals will therefore be determined by each component the signal passes through: the photodiode, the pre-amplifier, and the lock-in amplifier.

Light detectors generally have their performance rated by the noise equivalent power (NEP), which is a measure of the detectors sensitivity to light. Expressed in units of either $A/\sqrt{Hz}$ or $V/\sqrt{Hz}$, the NEP per unit area is defined as the signal power that would give rise to a signal-to-noise ratio of one in a one hertz bandwidth [107]. In other words, the NEP is the power that would have to fall on the detector to produce a signal equal to detectors noise. It is therefore desirable to choose a detector with an NEP that is at least an order of magnitude smaller than the signal to be detected. Table 3.1 compares the noise from each device in the setup used (as provided by the supplier’s manuals). From face value, the pre-amplifier has the largest noise contribution to the electronic signal.

<table>
<thead>
<tr>
<th>Model</th>
<th>Photodiode</th>
<th>Pre-amplifier</th>
<th>lock-in amplifier</th>
<th>DC voltmeter</th>
</tr>
</thead>
<tbody>
<tr>
<td>Noise</td>
<td>Hamamatsu S1223†</td>
<td>Femto LCA-100K-50M*</td>
<td>Signal Recovery 7265</td>
<td>Keithley 2010</td>
</tr>
<tr>
<td>9.4x10^-15 WHz^-1/2</td>
<td>3x10^-14 AHz^-1/2</td>
<td>5x10^-9 VHz^-1/2</td>
<td>110 nV</td>
<td></td>
</tr>
<tr>
<td>Effective NEP (nVHz^-1/2)</td>
<td>210</td>
<td>1500</td>
<td>5</td>
<td>110</td>
</tr>
</tbody>
</table>

Table 3.1 Effective electronic noise due to each device. †Responsivity=0.44A/W *Gain=5x10^7 (V/A).

The source of electronic noise in each component will be a sum of the different
noise sources, with thermal (Johnson–Nyquist) and shot noise being most significant at these kHz frequencies. Shot noise arises due to the discrete nature of electrons and is apparent on any signal crossing an energy barrier. A signal current $I_{\text{signal}}$ shows shot noise as a statistical variation in the measured current and can be described by the Schottky formula [19]

$$I_{\text{shot}} = \sqrt{2f_{BW}qI_{\text{signal}}},$$  \hspace{1cm} (3.15)

where $f_{BW}$ is the measurement bandwidth, and $q$ is the charge of an electron. Unlike shot noise, thermal noise varies according to ambient temperature $T$, as well as the resistance ($R_{\text{load}}$) across which the current is measured, and is given by

$$I_{\text{thermal}} = \sqrt{\frac{4f_{BW}k_BT}{R_{\text{load}}}},$$  \hspace{1cm} (3.16)

where $k_B$ is Boltzmann’s constant. Electronic noise is usually limited by the thermal noise [19].

Figure (3.14) shows the calculated voltage noise from Equations (3.15) & (3.16) on an effective incident voltage passing through the pre-amplifier. The pre-amplifier

**Figure 3.14** Comparison between the calculated noise sources and standard deviation of each of the three voltages used in determining the Kerr rotation and ellipticity. Data was taken for an arbitrary elliptical polarisation as the laser diode power was swept.
(Femto LCA-100K-50M) load resistance is given as $> 10k\Omega$ in the datasheet. An effective bandwidth of 0.3335Hz is used corresponding to the measurement bandwidth of the lock-in amplifiers (SR7265) with a time constant of 500ms, and a slope of 12dB/Octave. For the pre-amplifier, the thermal noise is most significant for the measurement intensity used (between $10\mu V$ and 1V). The measured standard deviation on a highly elliptical polarised light state is also plotted in Figure (3.14) for all three measurement voltages used to determine $\theta_K$ and $\epsilon_K$. The laser diode power was swept to collect the data, and the voltage noise was determined from the standard deviation of 100 points with the above lock-in amplifier settings.

The experimental data in Figure (3.14) roughly corresponds to the calculated thermal noise, though differences arise likely due to values in Equations (3.15) & (3.16) deviating from the manufacturers quotations (which will also be different for the measurement frequencies used). At low power, the noise:signal falls as $1$/signal due to the pre-amplifier Johnson noise. For higher power, above 0.1V, the noise:signal improves at a lesser rate and follows the same slope as the calculated shot noise, although the data is nearly two orders of magnitude larger than the calculation.

### 3.3.2 Measured Noise

The experimentally observed noise varies with the power incident upon the photodiode. Figure (3.15) is a plot of the standard deviation of 100 measurements of $\theta_K$ against the effective incident power, presented as the proxy DC signal on the photodiode. The measurement was taken with light reflected from a silver mirror placed after the first beam splitter in Figure (3.1) within the thermally regulated box. This optical path emulates that of an actual experiment minus the extra path length travelled to the objective and sample mount. As the incident power is increased, the measured noise in $\theta_K$ decreases as expected, reaching $\approx 12\mu rad$ for an incident DC signal of 1.15V.

Also plotted in Figure (3.15) is the red curve representing $1$/DC scaled by a factor of 13 to overlap with the data. The broad agreement with the measured $\sigma_{\theta_K}$ shows how the Johnson noise is largely dominant in the calculated $\theta_K$ signal, at least up to 1V. Whether this $1$/DC relation extends for higher power in $\sigma_{\theta_K}$ or whether the noise follows the shot limit as shown in Figure (3.14) has not been determined. A simple solution to reducing the noise in measured $\theta_K$ however,
could be realised by increasing the power of the laser diode. Increasing the laser power from 5mW to 50mW, with no other changes in the optical setup, could reduce $\sigma_{\theta_K}$ to 1.2$\mu$rad, assuming the noise continues to follow an inverse power law.

### 3.4 Dual PEM Uncertainties

This section is focused on uncertainties in the dual PEM detection system that may produce errors in any measured Kerr rotation or ellipticity angle. These consist mainly of errors in the angle settings of the optical components, and variations in their function which themselves may arise due to physical movements or temperature variations. These uncertainties are considered separately from those arising in the electronic signal between the photodiode and lock-in amplifiers which itself was treated in Section (3.3).

#### 3.4.1 Error Analysis of Dual PEM Detection

There are two things to consider with any measurement system: signal offset and signal noise, where the ‘signals’ in this case are the measured voltages, and the angles $\theta_K$ & $\epsilon_K$ which are calculated from these. A signal offset here is considered as any measured angle $\theta_K$ or $\epsilon_K$ that does not originate from the sample under question, or the initial polarisation state generated. Similarly, a signal noise is a
variation in $\theta_K$ or $\epsilon_K$ that occurs during the acquisition of a single measurement.

The dual PEM detection system consists of three components (the two PEMs and a polariser as shown in Figure (2.8)). These three components have an error in the angle they are set ($\delta\alpha$, $\delta\beta$, $\delta\gamma$), and the two PEMs have an additional error in the peak retardation applied to polarised light transmitted through them ($\delta\Delta_{10}$, $\delta\Delta_{20}$). This retardation error may be due to the electronic signal sent to the PEM head being different to the input setting, or from the PEM being misaligned from the optical plane that can increase the path length light passes through the optical element of the PEM. Thus, from these five errors, there will be an offset in any measured $\theta_K$ or $\epsilon_K$, and if any of these errors are causing a variation in measured voltage within one measurement cycle, there will be a noise added.

**Uncertainty in PEM Retardation Amplitude** - $\delta\theta_K(\Delta_{10,20})$ & $\delta\epsilon_K(\Delta_{10,20})$

Taking the full expression for measured AC signals before inputting angles or retardation amplitudes (Equation (2.43)), the difference in the actual Kerr rotation to that measured due to the uncertainty in PEM retardation amplitude ($\delta\theta_K = \theta_K(\Delta_0) - \theta_K(\Delta_0 + \delta\Delta_0)$) is plotted in Figure (3.16). Here, the input polarisation is purely linear ($V = 0$) and so a mix of $Q$ and $U$ relative to the optic axis (which determines the PEM angle settings). As the actual polarisation state

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**Figure 3.16** How the calculated linear polarisation angle differs from the actual polarisation angle with different uncertainties in the PEM retardation amplitudes $\delta\Delta_{10,20}$. 

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varies from 0 to $\frac{\pi}{2}$, the error due to the PEM amplitude setting is maximum when the polarisation state is an equal mix of $Q$, and $U$, i.e. at $\frac{\pi}{8}, \frac{3\pi}{8}$. For a positive error in PEM retardation amplitude ($\delta \Delta_{10,20}$), the first and second PEMs have equal amplitude dependencies but are odd with respect to each other.

If the actual polarisation rotation is small, as will be the case for most MOKE measurements which will be measuring the rotation from perfectly s- or p-polarised light ($S = (I, Q, \delta U, \delta V)^T$), then the uncertainty for an actual polarisation near $n\frac{\pi}{4}$ (for integer $n$) is more important. Figure (3.17) is a heatmap of uncertainty in measured rotation, as in Figure (3.16), for errors in $\Delta_0$ and over a smaller range of rotation from s-polarised light ($Q = 1, U = 0$). For an actual rotation of 10mrad, the spurious offset in the measured rotation is up to 45$\mu$rad if the uncertainty in PEM retardation amplitude is 0.002 (0.5% of $\Delta_0$ at 0.383$\frac{2\pi}{\lambda}$) for either of the two PEMs. Note that the $\theta_K$ measurement error is of opposite sign for the uncertainty in retardation amplitudes of each PEM (i.e. $\delta \Delta_{10} = -\delta \Delta_{20}$).

![Figure 3.17](image)

**Figure 3.17** How the calculated linear polarisation angle differs from the actual polarisation angle with different errors in the PEM retardation amplitudes $\delta \Delta_{10,20}$.

The error in measured ellipticity angle due to the uncertainty in each of the PEM retardation amplitudes is different for each of the PEMs. Additionally, the state of polarisation changes how sensitive the ellipticity angle is to errors in $\Delta_{10,20}$.

The elliptical angles $\theta_K$ & $\epsilon_K$ can be used to express the Stokes parameters $Q$, $U$, & $V$. This may be done geometrically or algebraically as in the following. Acknowledging the equality from Equation (2.31) in Equation (2.33)
(i.e. assuming light is completely polarised) and rearranging for $V$ gives

$$V = \sqrt{Q^2 + U^2 + V^2 \sin (2\epsilon_K)} = \sin (2\epsilon_K). \quad (3.17)$$

The two additional Stokes parameters may be expressed by rearranging Equations (2.31), (2.32), & (3.17) to give

$$Q = \cos (2\theta_K) \cos (2\epsilon_K), \quad (3.18a)$$

$$U = \sin (2\theta_K) \cos (2\epsilon_K). \quad (3.18b)$$

Equations (3.17) & (3.18) can therefore be used to control the normalised polarisation state in calculations using the two angles $\theta_K$ & $\epsilon_K$.

The error in measured ellipticity angle $\delta\epsilon_K$ may be plotted as a heat map with actual ellipticity angle $\epsilon_K$ vs. the error in peak retardation of the first PEM (Figure (3.18a)), and that of the second PEM (Figure (3.18b)) for different angles

Figure 3.18  Calculated error in measured ellipticity angle ($\delta\epsilon_K$) as a function of the actual ellipticity angle ($\epsilon_K$) and the uncertainty in (a) the first PEM retardation amplitude $\Delta_{10}$, and (b) the second PEM retardation amplitude $\Delta_{20}$. The three plots for each case represent calculated $\delta\epsilon_K$ for different choices of the plane of polarisation ellipse (determined by U:Q).
of the major ellipse.

In both Figures (3.18a) & (3.18b), the maximum error in measured ellipticity angle occurs around the actual ellipticity angle of \( \frac{\pi}{8} \). Additionally, for both PEMs, the maximum error occurs when the orientation of the polarisation ellipse is aligned with the PEM modulation axis. For the first PEM oriented at \( \frac{\pi}{4} \) to the optic axis, the maximum \( \delta \epsilon_K \) occurs when \( U = 0 \) i.e. when the elliptical polarisation has its major axis aligned at \( \frac{\pi}{4} \) to the optic axis. For the second PEM oriented along the optic axis, the maximum \( \delta \epsilon_K \) occurs when \( Q = 0 \) i.e. when the elliptical polarisation has its major axis aligned along the optic axis.

Differences in the maximum \( \delta \epsilon_K \) between the two PEM retardation amplitudes should be noted. The first PEM described in Figure (3.18a) has twice as large a \( \delta \epsilon_K \) over the plotted region, showing an error of 2.3mrad when the uncertainty in \( \Delta_{10} \) is 0.002 (or 0.5% of the set amplitude). Another thing to note is the complete insensitivity to error for the ellipticity angle for the second PEM amplitude when the polarisation ellipse is aligned along the PEM modulation direction, as shown by the \( U = 0 \) case in Figure (3.18b).

For MOKE measurements, the circular component is usually very small as input polarisation is most typically highly linearly polarised. For the typical 0.5% error in each of the PEM amplitudes, the error in measured ellipticity angle is

![Figure 3.19](image)

**Figure 3.19** Calculated error in measured ellipticity angle (\( \delta \epsilon_K \)) as a function of the actual ellipticity angle (\( \epsilon_K \)) for a fixed uncertainty in the two PEMs retardation amplitude \( \delta \Delta_{10,20} \). Curves for the three planes of elliptical polarisation described in the text are included.
summarised in Figure (3.19), for the three combinations of $U:Q$ as previously plotted in Figure (3.18). The largest errors occur for the uncertainty in the first PEM amplitude, rising to 8 µrad for when the actual ellipticity angle is 1°≈ 17 mrad. This corresponds to 0.05% of the angle.

**Uncertainty in PEM & Analyser Angles - $\delta \theta_K(\alpha, \beta, \gamma)$ & $\delta \epsilon_K(\alpha, \beta, \gamma)$**

Similarly to the consideration of errors in setting the PEM retardation amplitudes, the possible misalignment of the two PEMs and the analyser are considered starting from the full expressions for the AC signals in Equation (2.43). Here, the PEM angles are denoted $\alpha$ & $\beta$, and the analyser angle is $\gamma$.

Rotating the first PEM ($\alpha$) essentially changes the coordinate system from which the MOKE angles are measured. This means an unintentional misalignment of $\alpha$ is exactly the “error” in $\theta_K$ relative to the optic axis i.e. rotating the first PEM by 1° causes the “error” in the measured Kerr rotation to be 1°.

Figure (3.20) is the calculated error in the measured Kerr rotation ($\delta \theta_K$) as a function of actual polarisation angle ($\epsilon_K$) for offsets in the second PEM angle ($\beta$) and the analyser angle ($\gamma$). These are calculated from the differences ($\delta \theta_K = \theta_K(\beta) - \theta_K(\beta + \delta \beta)$) & ($\delta \epsilon_K = \theta_K(\gamma) - \theta_K(\gamma + \delta \gamma)$). Both devices show

![Figure 3.20](image-url)  
Figure 3.20 Calculated error in measured Kerr angle ($\delta \theta_K$) as a function of the actual linear polarisation angle ($\epsilon_K$) for a few misalignments of the second PEM angle ($\beta$) and the analyser angle ($\gamma$).
similar errors in $\theta_K$ as a function of actual polarisation angle but out of phase by $90^\circ$. Additionally, the maximum $\delta\theta_K$ occurs with polarisation angles of $\frac{\pi}{4}$ and $\frac{3\pi}{4}$. The maximum $\delta\theta_K$ is the offset angle of the second PEM or analyser. This provides a simple method to identify misalignments in either the analyser or second polariser relative to the first PEM (which defines the optic axis): if the linear polariser used to generate the linear polarisation is mounted onto a goniometer, the peak measured $\theta_K$ relative to the theoretical $\theta_K$ from the linear polariser angle can be taken as a misalignment of the PEM or analyser.

The error in measured ellipticity angle $\delta\epsilon_K$ due to misalignments of the PEMs and analyser are summarised in Figure (3.21). The measured ellipticity angle is insensitive to a misalignment of the first PEM ($\delta\alpha$). A misalignment of the second PEM or analyser however, has the most marked effect on elliptically polarised light when the major axis is aligned along the second PEMs modulation direction, and when the light is moderately elliptical ($\epsilon_K = \frac{\pi}{8}$). This manifests as a maximum $\delta\epsilon_K$ equal to the angular missetting of the PEM/analyser.

For typical large signals from MOKE measurements of $\theta_K = \epsilon_K = 15\text{mrad}$, estimated spurious offsets due to the misalignment of the PEMs or analyser are summarised in Figure (3.22). Similarly to Figure (3.21), the ellipticity measured is insensitive to the first PEM angle, and has opposite sign to a positive misalignment of the second PEM or analyser. Over this misalignment range (just over $1^\circ$) $\delta\epsilon_K$ is slightly more sensitive to $\delta\gamma$, the analyser angle.
Figure 3.22  Calculated error in measured ellipticity angle ($\delta \epsilon_K$) as a function of misalignments of the first PEM ($\alpha$), second PEM ($\beta$), and the analyser ($\gamma$). The polarisation state used has typical $\theta_K = \epsilon_K = 15\text{mrad}$.

Conclusion

To conclude, spurious offsets in both $\theta_K$ & $\epsilon_K$ appear when there is a misalignment of either the two PEMs or the analyser, as well as if there is an error in either of the PEMs retardation amplitudes. The typical error for the setting of PEM amplitudes of 0.5% can lead to an offset in the measured $\theta_k$ of 40$\mu$rad, and $\epsilon_K$ of 20$\mu$rad on a large 17mrad signal. If the PEM amplitude drifts over time according to this 0.5%, a drift in the measured $\theta_k$ and $\epsilon_K$ of order 10$\mu$rad should also be expected.

For a large MOKE signal of $\theta_K = 1\degree \approx 17\text{mrad}$, a misalignment of 1.5mrad of either the second PEM or analyser may contribute to an offset in the measured $\theta_k$ of 53$\mu$rad. Similarly, a misalignment of 1.5mrad corresponds to an offset in the measured $\epsilon_k$ of roughly 2$\mu$rad for either the second PEM or analyser.

It should be noted that other sources of actual $\theta_K$ & $\epsilon_K$ that are not caused by the detection system or the sample exist. Each optical component reflected from or transmitted through will produce a non-zero $\theta_K$ & $\epsilon_K$. This is due to simple Fresnel reflection coefficients upon reflection, and linear birefringence which may be brought about by residual stress from the manufacture or holding each optical component.
3.5 Cryogenic Adaptation

The enclosed optical layout described in Figure (3.1) allows the MOKE measurement setup to be adapted to different experimental layouts. This for example allows the MOKE microscope to measure samples placed within cryostats to investigate the low temperature MOKE response of materials. Figure (3.23) is an image of the continuous He-flow cryostat (Cryogenics Ltd.) available and the adapted brass sample mount. The He-flow cryostat can generate magnetic fields up to 9T in the vertical orientation and operates in the temperature range 300-1.8K. The sample mount is screwed onto the end of a metre-long variable temperature insert that is loaded into the top of the cryostat. Electrical connections for the various thermometers and piezoelectric actuators are fed through to the top of the insert.

A viewing port is built into the underside of the cryostat, allowing light to be directed into the sample chamber. Two separate windows are required to separate the inner He-flow chamber from the insulating vacuum layer, and the vacuum layer from the outside air. Both glass windows are made from 2mm thick Spectrosil B, a type of quartz that reflects infra-red wavelengths that would heat

![Image](https://example.com/image.png)

**Figure 3.23** Images of the continuous He-flow cryostat and the sample mount as described in the text. The brass sample mount is 11cm long and has a 5cm outer diameter.
the inner chamber, transmits optical wavelengths, and has reduced birefringence. The thermally regulated optics board is mounted onto a mechanical \(xy\) stage which is mechanically coupled to the cryostat by securing it onto the same aluminium frame. Light for the MOKE measurement and widefield imaging can be aligned and transmitted through the viewing port glass to be focused by the objective lens onto the sample.

The sample mount is constructed of brass and is oriented as in Figure (3.23), with the sample stuck onto a copper sheet that is screwed onto three piezoelectric transducers allowing \(x, y, z\) translation. A PTFE (Teflon) dampener is attached to the mount in order to reduce vibrations by wedging it against the side of the inner cryostat chamber. Whilst the vibrations can be greatly reduced with this dampener, they are not totally eliminated as the chamber walls are not decoupled from the compressor; small vibrations of amplitude \(\approx 1\mu\text{m}\) can be observed in the widefield imaging feed.

In this setup, only polar MOKE measurements are possible due to the fixed position of the superconducting magnet. Various problems must be overcome in order to accurately measure MOKE within such a top loading He-flow cryostat.

One of the outstanding problems of the adaptation of the MOKE setup to this cryostat is due to thermal expansion of the aluminium arms holding the optics board to the cryostat bracket. Within the room, temperature variations over the day cause a thermal expansion within the cryostat frame which extends each of the four aluminium arms slightly differently. This can be observed over a timescale of hours as the focused red laser beam shifts the position incident upon the objective lens within the cryostat. A shift of several 10’s of microns over several hours translates into a large drift in the measured MOKE angles which are not simply linear with the external room temperature (which is an open lab with sporadic temperature fluctuations). To overcome this issue, either the aluminium arms can be replaced with a material with reduced thermal expansion, or an active system to control the temperature of the bracket may be designed.

### 3.5.1 Window Faraday Rotation

An additional problem that requires overcoming is the parasitic Faraday rotation that can be measured from the quartz windows of the cryostat. Any glass will have some residual birefringence which may be exacerbated by any stress applied
when mounted. In vacuum systems, low stress mounting techniques are used however, a small portion of stress will remain in the glass from the clamps and from the pressure difference between the vacuum and atmosphere. With a residual birefringence, any external field applied to the window will cause Faraday rotation of polarised light propagating parallel to the field direction. This may commonly be subtracted from polar MOKE measurements, as Faraday rotation is linear according to Equation (1.1). For highly sensitive measurements however, the slope of the Faraday rotation with field may vary with the temperature of the glass, or the exact position light is transmitted due to residual stress or a mounting stress.

There are a couple of common techniques used to overcome this parasitic Faraday rotation. Taking a reference measurement from an in-situ “null” sample (for example a metal like silver with a small magneto-optic signal) and comparing that to the measured rotation of the magnetic sample in question is one such method. Otherwise, for specific MOKE orientations and experimental setups, light may be transmitted through the window ensuring it is normal to the applied magnetic field (as in [68]). Here, the feasibility of a novel setup utilising two quarter waveplates (QWP) to convert linearly polarised light to circular polarised light to negate Faraday rotation in the windows of the cryostat when conducting polar MOKE measurements is presented.

Faraday rotation can be simulated in Mueller matrix calculations by using a rotation matrix that is a function of magnetic field. In a reflection measurement, polarised light passes through a glass window twice with the outgoing polarisation state calculated from

$$S' = M_{\text{Window}} \cdot M_{\text{Mirror}} \cdot M_{\text{Window}} \cdot S.$$  \hspace{1cm} (3.19)

The Mueller matrices for the mirror and window (treated as a rotation matrix) are defined in Appendix (A). For a 0.5cm thick SiO$_2$ window, the Faraday rotation of polarised light transmitted through it is 0.22 rad. This is assuming a Verdet constant of 0.44 rad/cm/T at 633nm [108]. The resultant Stokes vector for an arbitrary input polarisation state when passing through the window is then

$$S' = \begin{pmatrix} \sqrt{Q^2 + U^2 + V^2} \\ Q (\cos^2(0.22B) - \sin^2(0.22B)) - 2U \cos(0.22B) \sin(0.22B) \\ U (\sin^2(0.22B) - \cos^2(0.22B)) - 2Q \cos(0.22B) \sin(0.22B) \\ -V \end{pmatrix}, \hspace{1cm} (3.20)$$
where the window does not change the total intensity or circular component of polarisation ($V$). The $Q$ & $U$ components are effectively mixed, rotating the plane of polarisation. When plotted versus field for a linearly s-polarised input light beam ($S = (1, 1, 0, 0)^T$), the Kerr rotation and ellipticity (calculated from Equation (3.20) and definitions in Equations (2.32) & (2.33)) are as in Figure (3.24).

The proposed method to reduce Faraday rotation from the window is modelled according to Figure (3.25). The two QWPs are functions of their orientation angles ($\theta_1$ & $\theta_2$) as well as their retardances ($\phi_1$ & $\phi_2$). The outgoing polarisation state ($S'$) is calculated by multiplying out the following Mueller matrix series

$$S' = M_{QWP1}(-\theta_1, \phi_1) \cdot M_{Window}(V_{\text{Verdet}}, -B) \cdot M_{QWP2}(-\theta_2, \phi_2) \cdot M_{Mirror} \cdot M_{QWP2}(\theta_2, \phi_2) \cdot M_{Window}(V_{\text{Verdet}}, B) \cdot M_{QWP1}(\theta_1, \phi_1) \cdot S,$$

(3.21)

where the relevant Mueller matrices are defined in Appendix (A). Upon reflection, it should be noted that the relative orientations of the QWPs are reversed ($\theta_{1,2} \rightarrow -\theta_{1,2}$) and the magnetic field is also reversed ($B \rightarrow -B$).

The first QWP should be aligned at $\frac{\pi}{4}$ relative to the input linearly polarised light, $S$, in order to convert the polarisation into perfectly circular light when transmitted through the window. The second QWP then may theoretically be aligned at any orientation however, this controls the plane of linear polarisation incident upon the mirror. These parameters ($\theta_1 = \frac{\pi}{4}$, $\phi_1 = \phi_2 = \frac{\pi}{2}$) may be input
into Equation (3.21) to give the final polarisation state as

\[
S' = \begin{pmatrix}
\sqrt{Q^2 + U^2 + V^2} \\
Q \\
U \cos(4\theta_2) - V \sin(4\theta_2) \\
V \cos(4\theta_2) + U \sin(4\theta_2)
\end{pmatrix}
\] (3.22)

Note that this is independent of the applied magnetic field. If the input polarisation is chosen to be \(S = (1, 1, 0, 0)^T\) then \(S' = S\) and the measurement is insensitive to the Faraday rotation of the window.

**Quarter Waveplate Errors**

The above describes the perfect scenario. In practice however, both misalignment of the QWPs and values of their retardances will manifest as a change in both measured \(\theta_K\) & \(\epsilon_K\). In the best case, the orientation error of the QWPs will be limited by the goniometer used to rotate them. A typical precision rotation mount will have a resolution of \(\approx 1.5\text{mrad}\) [109]. The actual retardance of zero-order QWPs also typically have a manufacturer-quoted accuracy of \(\frac{\lambda}{100}\) for 633nm light [110]. Figure (3.26) is the corresponding calculated field dependence of \(\theta_K\) & \(\epsilon_K\) for Equation (3.21) assuming inaccuracies in both \(\theta_{1,2}\) & \(\phi_{1,2}\) in this best case scenario. The Kerr rotation is drastically reduced from \(220\text{mrad}\) to \(14\text{mrad}\) with the two QWPs however, the ellipticity angle has an even response in field reaching a value of \(\approx 1.6\text{mrad}\) at \(\pm 1\text{T}\) compared to the flat response without the QWPs.
Whilst the great improvement in the polarisation rotation may be achieved using the dual QWP trick, it adds a field-dependent ellipticity if the measurement is sensitive to this component. This is not to mention the effect that a magnetic field has on the QWP inside the vacuum chamber (which must be placed between the window and the mirror/sample). As QWPs are naturally birefringent devices, a magnetic field will cause a large change in the effective birefringence that transmitted polarised light will experience. Additionally, in an experiment, the perfect mirror is replaced with a sample, and likely an objective lens too, which both produce a change in the polarisation state of light via a MOKE signal or linear birefringence. This change in $\theta_K$ & $\epsilon_K$ will act the same as an alignment error in the return path through the QWPs and window. Hence, the measured $\theta_K$ & $\epsilon_K$ will be different to the actual MOKE signal from the sample.

3.5.2 Autofocus Routine

Temperature variations of a sample mount inevitably lead to defocusing of the image formed. Thermal expansion/contraction of the apparatus holding the sample and objective lens change the distance between the sample and objective. This can be problematic even for room temperature measurements if a high NA objective lens with shallow depth of field is used and not adequately thermally regulated.

As an example for the 11cm long brass sample mount described in Figure (3.23),
the linear thermal expansion coefficient $\alpha_L$ defined as
\[ \alpha_L = \frac{\Delta L}{L \Delta T}, \]  
(3.23)

where $L$ is the length of material, and $T$ is the temperature. For yellow brass (65% Cu, 35% Zn), the thermal contraction below 300K is summarised in Figure (3.27). As a crude model, one would expect a contraction of about 400µm when cooling to near zero kelvin relative to the length at 300K. Considering the depth of field of an NA=0.5 objective lens is roughly 1µm, a system to keep focus maintained is vital.

To compensate for this change in distance, the sample is placed onto a vertical piezoelectric stack, providing control of the sample-objective distance.

Piezoelectric actuators work using the stick-slip mechanism which is described in Figure (3.28). During the sticking phase, the rising slope of the saw-tooth voltage pattern is applied to the piezoelectric stack, causing it to expand and pushing the clamped table a distance $\Delta x$. The voltage is then removed quickly in the slipping phase, returning the piezoelectric stack to its original size, but leaving the clamped table in its new position due to the retracting force of the guiding rod overcoming the friction between the guiding rod and the clamped table.

This stick-slip method is fairly reliable for moving small distances and has a quoted direction repeatability $\leq 5\%$ [112]. For maintaining a fixed distance (the sample-objective lens distance in the current case) some type of feedback is required. Built-in “closed loop” piezoelectric controllers exist for this purpose and use either capacitive sensors or an interferometer. The method chosen to
Figure 3.28 Working principle of a piezoelectric actuator (described in text). From [112].

maintain the focus distance uses image analysis.

The image of the sample surface formed on the CMOS sensor is used to quantify degree-of-focus. While there are various techniques to determine image focus (for example reading the total image intensity with a higher intensity indicating better focus, or contrast detection along a straight line over a specified feature), the most repeatable and consistent method was found to be circular edge detection. The field diaphragm in front of the LED is partially closed to project a near-circular decagon onto the sample, resulting in the image formed as in Figure (3.29c) when in focus. If the image is out-of-focus, the blades of the diaphragm are blurry, as in Figure (3.29a).

Circular edge detection is performed with the “IMAQ Find Circular Edge VI” in Labview. Figure (3.30a) illustrates the method used. An image searches in an annular region, between the user-selected green circles, along the blue paths, using edge detection to find points of largest contrast which is assigned the position of

Figure 3.29 Image of the field diaphragm projected onto the sample surface showing the progress in focusing from under-focused (a) & (b) to in focus (c).
Figure 3.30  (a) Image collected during the autofocus routine overlaid with the search region (between green circles), (blue) line scans, and (red) circle detected. (b) The score given by circular edge detection for different distances between sample and objective lens.

the edge. Each of these individual edges are then combined to produce a single best-fit circle. The confidence in the circle is quantified with a ‘score’ that has a higher value for higher confidence. An example of the score given as the focal distance is swept from under-focused to over-focused is given in Figure (3.30b). In this example, a score of 58 would be a focused image.

This autofocusing method may be used when imaging flat surfaces however, the edge of the image is cut off due to the diaphragm, as visible in Figure (3.29). The discrete steps taken by the piezoelectric transducer generally is a drawback and can cause small jumps in the measured MOKE signal. Particularly with the transducers that were used here, a threshold voltage is needed to move them, corresponding to a distance of ~100nm which is a fairly large jump relative to the depth of focus. As such, the piezoelectric transducers may also be operated in a DC mode. The transducer remains in the sticking phase of Figure (3.28) and a constant DC voltage is supplied to the piezoelectric actuator, expanding smoothly with additional supplied voltage and finely controlling focus. Whilst control units that can do this DC mode in addition to the slip-stick mode may be purchased, this was not available here. Instead a switch to change inputs to the transducer between a DC voltage source and the regular saw-tooth voltage supply was used.

Figure (3.31) represents the logic used in the written LabView program to control the focus. Initially a frame is captured and the circular edge detection as described
above is run. If the score is large enough then the MOKE measurement may be taken at this time. If the score is lower than the threshold then focus is changed using the transducer and another frame is captured to re-run the circular edge detection. If this new score is greater than the previous score, the piezo moves in the same direction. If the score is less than the previous score, then the direction is reversed. This sequence is repeated until the program ends and good focus is achieved.

3.6 Domain Imaging Routine

Magnetic domain images may be formed by scanning the focused spot across a sample, and making note of the $x, y$ coordinates for each position. The resolution here is dictated by the size of the focused spot as well as the step size used when scanning. Here, the sample is mounted on top of two orthogonal horizontal piezoelectric positioners (Attocube ANP-x101), which can be moved relative to the objective lens.

Due to the asymmetry in steps taken with the slip-stick piezoelectric positioners, object detection is used to accurately place each measurement position on the sample.
3.6.1 Program & Setup

The object tracking program is entirely within the Labview environment, using the NI-IMAQ API. For a video with each frame representing the position of the focused spot, the first frame is processed by the user to alter the brightness, contrast, and gamma before convoluting with a Gaussian blur kernel. Convolution is carried out by multiplying each pixels value with that of its neighbours which are weighted depending on the relative position. In practice, a simple 3-by-3 blurring kernel is of the form

\[
\frac{1}{16} \begin{pmatrix} 1 & 2 & 1 \\ 2 & 4 & 2 \\ 1 & 2 & 1 \end{pmatrix},
\]

(3.24)

where the central element is the pixel in question and the total intensity is normalised by the prefactor. This convolution with a blurring kernel smooths out the intensity of any object to be tracked and reduces noise, making the tracked object appear more uniform in-between frames. Following convolution, a threshold is applied to the frame, converting it from greyscale to binary, and making it easier for any object to be identified. For uniform illumination a global threshold is used, however if the illuminating light is uneven then a local threshold may instead be used.

The last step of the setup is for the user to highlight at least one object to be tracked. A traditional mean shift algorithm is then used to ‘learn’ this object and follow it through subsequent frames. The mean shift technique ‘learns’ the object through its intensity vs. position histogram, and the bounding rectangle is used to give the position of the object. Each frame is subsequently read, processed, and the position of the tracked object is calculated relative to its initial position \((x_{\text{initial}})\). The \(x, y\) co-ordinate of the centre of the bounding rectangle for the current frame \((x_{\text{current}})\) is related to the previous frame \((x_{\text{previous}})\) and the absolute position of the object relative to the start position with the first frame at \((0, 0)\) by

\[
x_{\text{coordinate}} = x_{\text{current}} - x_{\text{previous}} - x_{\text{initial}},
\]

(3.25)

where the subscripts indicate the frame for the position of the tracked object, and the same relation for the \(y\) position holds.

If the object is ‘lost’ then the current frame can be reprocessed, or a new object to
be tracked may be selected on the previous frame (and related to the old tracked object position) to continue the tracking. The new object position overwrites the initial coordinate so that the absolute coordinates are carried forward.

The accuracy of the coordinate position can be naively estimated by repeating the tracking routine for the same scan and comparing the output positions. Figure (3.32) displays the final end positions of ten repeats of the tracking routine on a scan consisting of $25 \times 10$ steps. The standard deviation for the error in final positioning of these ten scans is $0.61 \mu m$ for the x-coordinate and $0.89 \mu m$ for the y-coordinate. This equates to an error of roughly 1%. Considering the spot size of the measurement is $\approx 4 \mu m$, this method for tracking is adequate and generally reliable. If the measuring spot size was reduced, and the number of steps taken in the scan increased however, then the error of 1% is still significant enough to distort obtained images. In such situations a closed loop system using an in-built interferometer (that may be purchased in some positioner models) is the best choice.

The limitations of object tracking are mainly that it requires an object to always be in the field of view, and that checking from a user is necessary to ensure sensible positioning. For patterned materials this isn’t an issue however, for a flat sample either surface defects or non-magnetic tracker particles are required to generate accurate coordinates.

![Figure 3.32](image)

**Figure 3.32** The difference in finishing position given by ten repeats of the tracking routine for a scan over a region consisting of $25 \times 10$ steps equating to $\approx 150 \times 50 \mu m^2$. 89
3.6.2 Example Scan

As a demonstration of the necessity of using some sort of tracking for the raster scanning system, and to highlight the efficacy of the image-based object tracking method developed, a patterned Ni$_{80}$Fe$_{20}$ sheet is measured. As imaged in Figure (3.33a), the rectangular structures have dimensions 5µm × 25µm and a spacing of 5µm. Here, the regular structures are easy to recognise and track; on a typical sample, surface defects/dust are generally used to track position. The sample was under a constant 20mT field along the +x direction and focus was scanned across using two piezotransducers, one of which (for the y displacement) has a large asymmetry in movement position. Whilst the asymmetry can be partially corrected for by controlling the voltage supplied to the piezoelectric transducers, or the number of steps in each direction, this correction needed can vary with temperature or position of the transducer along its movable region.

The raw untracked raster results in the heatmap for ∆θ$_K$ displayed in Figure (3.33b). This shares the same ∆θ$_K$ scale as Figure (3.33c) and first order interpolation has been used to blend each measurement point. The result in Figure (3.33b) does not represent the physical structure as in Figure (3.33a).

Running the tracking program results in Figure (3.33c) where the x, y scale has been converted from pixels of the image to microns by calibration with a 1951 USAF resolution test chart (Thorlabs R1DS1N). Here, the structure is clear and the raster path is highly distorted along the y axis. Additionally, the resolution limitation from the finite spot size can be seen through the rounding of the mesa edges. The corresponding raster path that links Figures (3.33b) to (3.33c) is displayed in Figure (3.33d).
Figure 3.33 (a) Patterned Ni$_{80}$Fe$_{20}$ structures, (b) the Kerr rotation of an untracked scan over the patterned mesa structures in (a), (c) the corresponding tracked scan Kerr signal, and (d) the raster path obtained from the tracking.
3.7 Conclusion

This chapter outlined the experimental development of the MOKE detection system. The optical layout was described, and emphasis was placed on the choice of a reflective rather than a refractive objective lens. In comparison to refractive lenses, polarised light focused with reflective lenses does not incur a Faraday rotation and these lenses are typically more reliable for cryogenic applications. Reflective lenses can affect the polarisation state of transmitted light due to a change in the effective incidence plane relative to the input polarisation state, as explained in Section (3.1.3).

The resolution of focused light was measured to be $4\mu m$, and the typical angular resolution of measured $\theta_K$ and $\epsilon_K$ was measured to be $12\mu rad$, from the standard deviation of 100 separate measurements. This noise followed a $1/power$ relation up to the maximum $5mW$ laser power used, suggesting that Johnson (thermal) noise is limiting the sensitivity of the developed setup. In comparison to other measurement systems, this dual PEM setup is less sensitive than both the Sagnac interferometer and balanced photodiode setups, which both can reach near nrad sensitivity. It does retain the benefit of synchronously measuring $\theta_K$ and $\epsilon_K$, however. Additionally, a signal drift of up to $1.2\frac{\mu rad}{hour}$ was measurable for both MOKE angles, even after applying temperature regulation of the entire optical setup.

Errors in MOKE signals arising from a misalignment or imperfection in the retardation amplitude provided by the dual PEM setup were theoretically analysed in Section (3.4). Whilst the magnitude of each error source depends on the magnitude of $\theta_K$ and $\epsilon_K$, it was found that a typical MOKE signal of $1^\circ \approx 17\mu rad$ would have an error of $\sim 40\mu rad$ in $\theta_K$ and $\sim 20\mu rad$ in $\epsilon_K$ for a 0.5% error in the PEM retardation amplitude. Similarly, an error in $\theta_K$ of $50\mu rad$, and an error in $\epsilon_K$ of $2\mu rad$, was calculated if the second PEM or analyser angle setting was off by $1.5\mu rad$ - the resolution of the goniometers used.

Section (3.5) highlighted progress to adapt the MOKE setup to measure materials under cryogenic conditions. A PTFE dampener was attached to the constructed brass sample mount to reduce vibrations within the cryostat to $<1\mu m$. A long timescale drift in MOKE signals hampered the development, however. This was measurable as a movement of the position that light was incident on the objective lens, which was likely caused by ambient temperature fluctuations causing a
change in length of the cryostat-optics support frame. An attempt to reduce the Faraday rotation from the cryostat view-port was also theoretically analysed. This involved placing quarter waveplates either side of the cryostat windows to convert linearly polarised light into circularly polarised light that is immune to Faraday rotation. The sensitivity of this technique to angular settings of the waveplates however, indicated that this idea is ineffectual.

Finally, the sample scanning technique utilising image processing to capture magnetic domain images was described. This traditional mean shift processing procedure, as outlined in Section (3.6), was tested to be accurate to within 1% for moderate scanning areas. A similar image processing technique to retain image focus using a circular edge detection algorithm was outlined. This will be beneficial for future temperature dependent MOKE experiments.
Chapter 4

Permalloy - Ni$_{80}$Fe$_{20}$

In this chapter, MOKE measurements on patterned permalloy films are presented. This is included to verify the sensitivity and resolution of the developed scanning MOKE setup however, the results are relevant to micro-inductor design. Despite a large variation in measured Kerr rotation and ellipticity angle across patterned structures, under certain conditions stable domain structures can be imaged using the developed scanning MOKE microscope.

4.1 Introduction

Permalloy is a nickel-iron alloy typically consisting of roughly 80% Ni and 20% Fe. It is well characterised and has been commonly used throughout the last century within devices requiring a material with high magnetic permeability and low coercivity (and therefore also small hysteresis losses). The large spin polarisation in permalloy has led to it featuring prominently in spintronic research, particularly as a spin-detection material [113, 114].

Previously, MOKE studies on permalloy have utilised widefield imaging to record domain evolution, particularly for isolated micron sized elements [115–117], as well as for magnetometry. Much of the recent work with permalloy has used thin films: <100nm. The type of domain wall switches from Néel to Bloch walls when increasing the permalloy thickness past ~35nm [118]. Research using thick films (>1µm) of permalloy are mostly reserved for larger scale applications requiring its large permeability.
Inductors store energy in magnetic fields and can be used to provide power over time to electronic components. Typically being made from a conductive wire, the energy stored by an inductor may be increased by increasing the magnetic field through the use of a ferromagnetic core. A typical choice of core material is permalloy due to its high permeability. This core can either be inserted within a coiled wire, or instead it may envelope the current-carrying wire which is known as a planar core and is more compact in size. Miniaturising and increasing the power that can be stored within micro-inductors is needed to construct power supply on chip packages which are themselves a requirement for making electronic devices more compact [119].

One method of increasing the power that can be stored within inductors is to pattern the planar core. Whilst this patterning reduces the total inductance due to the reduced volume of ferromagnetic material, the Q-factor (ratio of energy stored to energy dissipated) increases as eddy current losses within the core are decreased by physically restricting the current path. Such patterns may be in the form of small rectangular mesas as depicted at the top of Figure (4.1a). Figure (3.33a) also depicts a top down image of these micron-sized permalloy patterns. In these devices, nickel is used as a seed layer for depositing the permalloy in order to increase the performance of the inductor by reducing eddy current losses in the cores [120]. Additionally, Parylene-C, a dielectric polymer, is used within the inductor to act as an insulating barrier between the wire and the core.

The magnetic structure within these patterned permalloy sheets is important to understand for the design of efficient micro-inductors. Understanding how the magnetic field from the inductor is affected by different patterns and sizes

Figure 4.1  (a) A planar inductor showing the current carrying copper wire surrounded by a permalloy core, from [121].  (b) Profile of permalloy test structures.
of permalloy core structures can help in achieving a large Q-factor. As such, test structures of various dimensions using a 300nm-thick nickel seed layer were synthesized by Coinneach Dover (Scottish Microelectronics Centre, University of Edinburgh) following the same deposition technique as described in [122]. The side profile of these structures is illustrated in Figure (4.1b), and they are referred to here as mesas.

### 4.2 MOKE Measurements on Permalloy Mesas

Long rectangular permalloy mesas with dimensions $20 \times 300 \mu m^2$, $5 \mu m$ thick, and with $10 \mu m$ separation are measured here. A scanning routine of the MOKE measurement as detailed in Section (3.6) was undertaken with step sizes of $\sim 2 \mu m$ vertically and $\sim 4 \mu m$ (equal to the spot size) horizontally. The incidence plane of s-polarised light ($\lambda = 633nm$) was aligned along the $x$-axis at an angle of $25^\circ$, and the sample temperature was held fixed at 295K. All heat maps are plotted with second order interpolation between measurement positions. Figure (4.2a) is the measured DC signal over three vertically arranged permalloy mesas after AC demagnetisation along $x$. These three structures are in the centre of a vertical stack of 31 identical mesas. The simultaneous measurement of the Kerr rotation and ellipticity angle are also printed in Figures (4.2b) & (4.2c). The first thing to note about the MOKE measurements is that the Kerr angle varies by more than $3mrad$ from the top to bottom edge of the structure. In contrast, the ellipticity angle is more consistent across the $y$-axis. Both $\theta_K$ and $\epsilon_K$ measurements are affected by the edge of the structures.

The cause of the large change in $\theta_K$ between the top and bottom edges of the permalloy mesas is not certain. The mesas themselves are not perfectly flat and are likely to be thicker in the centre. This would change the reflection angle into the reflective objective lens and, from Section (3.1.3) can lead to spurious offsets in measured MOKE angles. However, there is not as large a change in $\epsilon_K$ in Figure (4.2c) that would also be expected. Alternatively, the cause of $\theta_K$ variation could be from the focused light bleeding over the edges of the mesas and changing the measured MOKE angles due to including various scattering angles and a drop in intensity. This would explain the near uniformity of $\Delta \theta_K \approx 0mrad$ within the centre of the permalloy.

The local magnetic hysteresis of the surface may also be measured by the MOKE
Figure 4.2 (a) DC reflection intensity, (b) change in Kerr rotation, and (c) change in ellipticity angle for a scan over three demagnetised (along the $x$ axis) rectangular permalloy structures.
microscope. Focusing light at various points on the rectangular permalloy mesas can reveal how the local magnetisation changes with field. Figure (4.3) depicts two field sweeps when light is focused near the top and bottom edges of the centre of one 20×300µm² rectangular mesa. When light is focused on the top edge there is clear hysteresis between increasing (magenta) and decreasing (cyan) field directions. The total change between saturation in θ_K is ~0.3mrad and in ε_K is ~0.5mrad. The saturation field is ~4mT. Interestingly, the hysteresis loop is not centred on zero field and so when the field is reduced to zero, the relative Δθ_K and Δε_K are always the same sign. Increasing the field to positive values (up to 200mT) does not further change θ_K or ε_K. This indicates that the top edge of the mesa is preferentially choosing a magnetisation direction at zero field. Similarly for the MOKE measurement at the bottom edge of the permalloy, the hysteresis is not centred on zero field and in fact Δθ_K and Δε_K are of the opposite sign to that measured at the top edge at 0mT.

An additional feature to note is the shape of the curve depending on the field sweeping direction which is discernible in both θ_K and ε_K in Figure (4.3). When the field is moving towards 0mT (increasing for the top of the mesa - magenta; decreasing for the bottom edge of the mesa - blue) there is a sharp jump in

![Figure 4.3](image.png)

**Figure 4.3** Change in longitudinal Kerr and ellipticity angles for field sweeps when light is focused on the top and bottom half of the 20×300µm² rectangular permalloy structure.

98
measured angle. This is clearer in $\Delta \epsilon_K$ as a -0.35mrad jump followed by a smooth decrease until saturation in the magenta curve. Sweeping the field in the opposite direction however, results in a smooth change in the MOKE angles (decreasing for the top of the mesa - cyan; increasing for the bottom edge of the mesa - red). This suggests that moments are rotating away from the preferred direction when the field is large enough. Both the general shape and the field offset of the hysteresis loops indicates the presence of a persisting domain structure within these $20 \times 300 \mu m^2$ permalloy mesas.

4.2.1 Imaging Magnetic Domains

The above position-dependent MOKE response of the permalloy structures may be exploited to reveal the stable magnetic domain structure. Figure (4.4a) is a plot of the difference in measured ellipticity angle as a function of position across the same three mesas as in Figure (4.2) when saturated with a 8mT field along $+x$ and $-x$. This is essentially a plot showing the total $\Delta \epsilon_K$ as measured by the hysteresis plot in Figure (4.3). Looking instead at the difference between $\epsilon_K$ measured under $-8mT$ and $0mT$ as well as $+8mT$ and $0mT$ can reveal more information. This has been done for Figures (4.4b) & (4.4c). The headers $(\epsilon_K^{-8mT} - \epsilon_K^{0mT})$ and $(\epsilon_K^{+8mT} - \epsilon_K^{0mT})$ denote the $\Delta \epsilon_K$ that is plotted for each figure, which share the same colour scale.

Adding the data from Figures (4.4b) & (4.4c), i.e. $(\epsilon_K^{-8mT} - \epsilon_K^{0mT}) + (\epsilon_K^{+8mT} - \epsilon_K^{0mT})$, produces the plot in Figure (4.5b). This was similarly done for the $\Delta \theta_K$ data as presented in Figure (4.5a). From both $\Delta \theta_K$ & $\Delta \epsilon_K$ scans, the permalloy mesa shows a persistent domain structure. Split along the $y$-axis, the top and bottom of the mesas have MOKE signals, proportional to the local magnetisation, of opposite sign. These long mesas show typical shape-induced uniaxial magnetic structure. The persistent structure is illustrated in Figure (4.5c), resembling Figure (2.1). Particularly in the $\Delta \epsilon_K$ scans, Figure (4.5b), the top domain appears to be slightly thicker in all three permalloy mesas - the cause of this is unknown but it is possibly due to a small remanent field from the electromagnet pole pieces.
Figure 4.4  The difference in change of ellipticity angle measured over three permalloy structures between (a) positive and negative saturation fields of ±8mT, (b) fully saturated with a -8mT field and the zero field measurement, and (c) fully saturated with a +8mT field and the zero field measurement. The applied field and plane of incidence, and so MOKE sensitivity direction, is along the horizontal x-axis.
Figure 4.5 Measured change in MOKE signal (according to the equations included in the figures) over three permalloy structures when saturated with $\pm 8 mT$ along the $x$ axis. Both (a) the Kerr rotation, and (b) the ellipticity angle are included. The plane of incidence, and so MOKE sensitivity direction, is aligned with the field along the $x$-axis. (c) Illustration of the measured domain structure.

Figure (4.5) displays MOKE signals sensitive to components of magnetisation along the horizontal $x$-axis, and shows all three permalloy mesas aligned in the same sense: the top half has a positive Kerr/ellipticity rotation and the bottom half has a negative Kerr/ellipticity rotation. It is interesting to know whether these mesas, separated by 10 $\mu m$, interact with each other as this would indicate an adequate stray field from each mesa reaches neighbouring mesas. This could explain the formation of the observed simple domain structure. Figure (4.6a) plots a scan of the domains visualised in the $\Delta \varepsilon_K$ signal over the edge of five
neighbouring mesas. Both this and Figure (4.5) suggests that the mesas are not interacting. The ‘sense’ of the magnetic structure appears to be random, with the simple domain structure in neighbouring mesas sometimes identical, and sometimes reversed.

Edge domains are an important aspect, particularly in clarifying that the observed simple domain structure is correct. Figure (4.6b) represents a scan over the edge of one of the $20 \times 300 \mu m^2$ mesas in the same orientation as previous scans in this section. Horizontal magnetisation components along the top and bottom, separated by a domain wall along $y = 10 \mu m$ are distinguishable. Applying the magnetic field and focusing s-polarised light along the vertical $y$-axis of the permalloy mesas provides sensitivity along the $y$-axis. Figure (4.6c) represents a scan in this experimental orientation with a field amplitude of 20mT applied. The edge domain, completing the previously observed simple domain structure in

\[
\left( e_x^{0 \text{mT} - e_x^{0 \text{mT}}} \right) - \left( e_x^{0 \text{mT} - e_x^{0 \text{mT}}} \right)
\]

Figure 4.6 (a) The difference in change of ellipticity angle measured over four vertically aligned permalloy mesas with horizontal field and plane of incidence. (b) A zoomed in scan over the right edge of the lowest structure in (a). (c) The same permalloy mesa as in (b) but with the field applied along the $y$ axis.
the mesa, is clear. The resolution due to the 4µm focused spot limits the clarity of the edge domain and does not show triangular structure as could be expected from a typical edge domain as illustrated in Figure (2.1d).

4.3 Discussion

In the 5µm thick permalloy deposited onto a 300nm thick magnetic nickel seed layer, the measured change in ellipticity angle is greater than the change in Kerr angle. Within the 150×50µm² mesas, the measured Δθₖ is ~0.3mrad and Δεₖ is ~0.5mrad as shown in Figure (4.3). While it is difficult to find comparable longitudinal θₖ and εₖ measurements for the same permalloy structure as looked at here, the larger ellipticity response has previously been observed. In Ni₈₁Fe₁₉(8nm)/Fe₅₀Mn₅₀(20nm)/Co(5nm) structures treated with field cooling of 10mT from 240°C [123], MOKE signals were measured using a single PEM setup with an incidence angle of 45°, giving Δθₖ ≈ 0.5mrad and εₖ ≈ 1.4mrad. Taking into account the difference in incidence angle used here compared to [123], the MOKE signals should differ by approximately the ratios of cosines of the angles to the longitudinal direction, from Equation (2.27a), i.e. cos(45°)/cos(65°). Weighted by this ratio, the Δθₖ and Δεₖ measured here are ~0.50mrad and ~0.84mrad respectively which is in agreement considering the differences in permalloy structure.

The persistent domains imaged in Figure (4.5) represent the simplest form of closed magnetic domain structure. This has previously been imaged in rectangular micron-sized permalloy structures by transmission electron microscopy (TEM) [124]. 2×1µm² elements, as reprinted in Figure (4.7a), display a simple domain structure with large closure domains. Contrary to this, Figure (4.7b) shows a similar measurement for higher aspect ratio 4×1µm² elements with a complicated domain structure. Considering the aspect ratio of the 300×20µm² mesas studied here is 15, far larger than those imaged in Figures (4.7a) & (4.7b), a more complicated, non-persistent, domain structured should perhaps be expected. Widefield MOKE images captured on 16×16µm² elements in Figure (4.7c) further illustrate the domain evolution within isolated structures as the external field varies. Possible explanations for the simple domain structure observed in the scanning MOKE images on these high aspect ratio mesas here include the nickel seed layer, which affects the magnetisation of the whole structure. Also, the thicknesses of the permalloy elements in [124] are 26nm, within the Néel wall
regime, in contrast to the thick 5µm elements used here.

Efforts to measure the domain structure within permalloy mesas with different aspect ratios were unsuccessful. Using the scanning MOKE microscope, the same issue of a large variation across the length of mesas caused Kerr and ellipticity rotations far larger than the ∼0.4 mrad difference between opposite magnetisation directions in Figure (4.3). When attempting the same method to image magnetic structure by recording several field values at each position during the scan, for a mesa with dimensions 150×50 µm², the hysteresis loop was not consistent upon repetition. In other words, the domain structure didn’t evolve with field repeatably; there was not a single persistent domain structure as found for the highly uniaxial 20×300 µm² mesas. This highlights one of the limitations of the scanning MOKE setup when compared to widefield MOKE microscopes.

The stable domain structure observed in these thick permalloy mesas with a nickel seed layer is useful in regard to micro-inductor design. The uniaxial domain structure can serve to increase the overall magnetisation when the hard axis is oriented parallel to the alternating magnetic field within an inductor. Further investigating the critical thickness and dimensions that still give a large shape-induced uniaxial anisotropy within non-interacting elements may lead the way to increasing the energy stored within micro-inductors. The observed simple domain structure that limits the number of domain walls reduces one form of energy loss during the charging/discharging process in inductors. Further systematic studies are required for optimising the inductor performance with a core permalloy mesa.
structure, however.

4.4 Conclusion

The developed MOKE microscope has been used to measure the local magnetic response of permalloy films. 5µm thick permalloy structures grown on a 300nm thick nickel seed layer show MOKE hysteresis with $\Delta \theta_K = 0.3$ mrad and $\Delta \epsilon_K = 0.5$ mrad. The scanning mode of the MOKE microscope has been used to image the persistent domain structure in permalloy mesas. Despite constant field scans resulting in no magnetic contrast, taking multiple measurements whilst applying different external fields at each position of a scan allows a stable magnetic structure to be revealed. This however, was only the case for very high aspect ratio (20×300µm²) mesas with correspondingly high shape-induced uniaxial anisotropy. Furthermore, many of these rectangular mesas when vertically aligned with 10µm separation showed seemingly random chirality, indicating that at this distance they are non-interacting.

The strength of the developed MOKE microscope is in the clarity and separation of $\theta_K$ and $\epsilon_K$ in local magnetic hysteresis measurements. Scanning the focus point of the MOKE measurement has been observed to be strongly affected by the edges of structures. Moreover, the time required to obtain domain images using a scanning MOKE setup is significant - often taking several hours to collect a single image. As such, a widefield MOKE system would be better for imaging the domain evolution. Alternatively, improvements could be made to the scanning MOKE microscope by reducing the spot size of focused light with a higher NA objective lens. Further work on the mesa structures as part of full inductor devices as illustrated in Figure (4.1a) could be undertaken to measure the magnetic structure within different sized mesas, different pattern shapes, as well as the magnetic evolution whilst the inductor is charging/discharging. This could help to optimise the inductance of (and hence feasibility of incorporating) micro-inductors into power supply on chip packages.
Chapter 5

Galfenol - Fe$_{1-x}$Ga$_x$

This chapter details MOKE measurements on thin films of the magnetostrictive alloy galfenol deposited on LiNbO$_3$. In-plane and out-of-plane components of magnetisation that the developed MOKE microscope is sensitive to are given for the galfenol films. Additionally, captured magnetic domain images are presented.

5.1 Introduction

Iron-gallium alloys, commonly referred to as galfenol, are of commercial interest due to their moderate magnetostriction when exposed to small, easily obtainable, magnetic fields. Magnetostrictive materials themselves are useful for any application requiring some conversion between kinetic and magnetic energy, for instance in sonar devices, vibrational energy sensing/harvesting, and precision actuators. Until the late 1990’s, Terfenol-D (Tb$_x$Dy$_{1-x}$Fe$_2$ with $x \approx 0.3$) was the dominant magnetostriction material of choice and is particularly prevalent in underwater sonar systems. Despite the large magnetostriction of up to 2,000$\mu$m/m under external fields of 200mT at room temperature [125], the usefulness of Terfenol-D is limited by the brittle nature of this alloy. This lack of ductility is usually overcome by alloying with other metals and polymers which naturally reduces the magnetostrictive properties.

In contrast, galfenol is readily machinable into various shapes such as sheets and wires. FeGa, however, does have a reduced magnetostriction (in comparison to Terfenol-D) of $\approx$400$\mu$m/m although this does require a smaller applied field.
How the maximum magnetostriction along (100) of single crystal galfenol changes with varying Fe:Ga is described in Figure (5.1a) [126]. There is a difference in the magnetostriction of FeGa that depends on the growth conditions. For furnace cooled samples, two maxima in the magnetostriction occur for Ga concentrations of \(\approx 17\% \) & \(27\%\). This double peak in the magnetostriction of FeGa is explained as due to the combination of increasing magnetoelastic energy (up to \(19\%\) Ga) and a non-linear softening of the shear elastic constant (up to \(27\%\)) [126]. Between \(20 - 27\%\) Ga, a mixed phase state is generally accepted as the cause of this elastic constant softening.

The origin of the mixed phase state may be understood from the iron-rich portion of the FeGa phase diagram in Figure (5.1b). The trough in magnetostriction coincides with the appearance of the B2 and D0\(_3\) structures upon cooling (at \(\approx 23\%\) Ga). The magnetostriction peak for \(27\%\) Ga occurs in the L1\(_2\) structure, having passed through the hexagonal D0\(_{19}\) structure during the growth.

Upon quenching, magnetostriction continues increasing with Ga concentration past that of equivalent un-quenched samples, peaking at \(\approx 19\%\) Ga rather than \(\approx 17\%\) Ga. Quenching suppresses the mixed phase state observed in FeGa.

![Figure 5.1](image)

**Figure 5.1** (a) Magnetostriction for varying Fe:Ga concentrations of single crystal galfenol (from [126]). (b) The FeGa phase diagram and various crystal structures at elevated temperatures. The \(T_C\) line represents the Curie transition, and shaded regions represent mixed phase states (adapted from [127]).
when slow cooling. There is not a current consensus fully explaining this suppression however, the distribution and subsequent interaction between Ga atoms is the source of an internal strain that stabilises a structure and increases the magnetostriction [128–130].

Thin film galfenol has promising applications in many micro-electromechanical systems, including surface acoustic wave (SAW) devices which will be outlined below in Section (5.6). As the magnetostriction of polycrystalline thin film galfenol will naturally be reduced compared to single crystal FeGa, growth conditions need to be understood to maximise its potential magnetostriction and minimise its coercivity. Si (100) (or its oxidised form quartz) is commonly chosen for the substrate upon which galfenol is deposited, particularly when investigating the effect of growth conditions on the magnetostriction of galfenol (several highlights include [131–133]). This choice is due to Si being a well-studied, cheap and high-quality substrate material that has similar crystal dimensions to FeGa. Different substrates for galfenol have also received interest. A decrease of 20% saturation magnetisation was observed in otherwise identical in-plane magnetisation hysteresis loop for Fe$_{89.5}$Ga$_{10.5}$ compared to Fe$_{75.7}$Ga$_{24.3}$ when deposited onto GaAs (001) [134], consistent with hysteresis loops using a Si substrate. MgO (100) has also been used as a substrate for FeGa when investigating growth of galfenol [135] and in particular, larger saturation magnetisation have been observed for films deposited on MgO (100) in comparison to quartz and silicon [136].

MOKE studies on galfenol include the following. Widefield MOKE domain imaging has been used to observe domain evolution of mechanically strained galfenol bars [137, 138], as well as their stable domain structures that highlight 90° and 180° domain walls [139, 140]. Normalised magnetic hysteresis using MOKE have distinguished similarly shaped loops with varying remanent fields for a few Fe:Ga 50nm thick films [132, 133]. The use of MOKE in looking at thin films of galfenol so far have focused on SAW devices [141, 142].

The basic operation of SAW devices for magnetostrictive applications follows Figure (5.2) [141]. The piezoelectric substrate converts a MHz frequency electronic signal into an acoustic standing wave that is set up between the two interdigitated transducers (IDTs) deposited on the substrate. In-between the IDTs the magnetostrictive film is deposited (in this case galfenol is used). Due to magnetostriction in galfenol, the acoustic wave that stretches the film decreases the coercive field of galfenol at the antinodes and does not affect the coercivity at
the nodes. This allows a periodic magnetic structure to be written by applying a field less than the normal state coercive field but greater than that of the coercive field when galfenol is strained. Such devices have been constructed using quartz as the substrate, and successful magnetic patterns have been written, including micron-sized spots using angled IDTs [141].

For the samples studied here, galfenol was deposited onto a LiNbO$_3$ substrate by David Czerski (Heriot-Watt University). To date, no other studies have been published investigating FeGa thin films deposited onto LiNbO$_3$. Lithium niobate itself is an attractive substrate material due to a few of its piezoelectric parameters. The electromechanical coupling coefficient ($k_s$) is a measure of the efficiency when converting between electric and acoustic energy. It may be defined in terms of the acoustic velocity for a clean surface ($v_f$) and the acoustic velocity for a surface with a conductor deposited on it ($v_m$) as

$$k_s^2 = \frac{2v_f - v_m}{v_f}. \quad (5.1)$$

X-cut quartz has $k_s^2 =0.16\%$, whereas 128° X-cut LiNbO$_3$ has a substantially larger $k_s^2 =5.5\%$ [143]; lithium niobate is far more efficient in converting between acoustic energy and electric energy. LiNbO$_3$ also has a higher sound velocity of $v_s=3960\text{ms}^{-1}$ in comparison to $v_s=3158\text{ms}^{-1}$ for SiO$_2$ [143]. The velocity of sound in turn determines the radio frequency at which conversion between electric and acoustic energy is maximum. For IDTs with periodicity $d$ and corresponding wavelength $1/d$, this frequency ($f_s$) is given by $f_s = v_s d$. For a fixed $d$, therefore, a higher $v_s$ allows the use of higher frequencies and so faster response times.

The obvious main drawback to using LiNbO$_3$ as a substrate for FeGa however, is the mismatch in lattice parameters. Hexagonal LiNbO$_3$ has lattice parameters...
<table>
<thead>
<tr>
<th>Ar Pressure ($\times 10^{-3}$mbar)</th>
<th>Time (mins)</th>
<th>Thickness (nm)</th>
<th>Fe %</th>
<th>Ga %</th>
</tr>
</thead>
<tbody>
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<td>2</td>
<td>20</td>
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<td>12.5</td>
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<td>15.5</td>
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<td>6</td>
<td>40</td>
<td>616</td>
<td>84.2</td>
<td>15.8</td>
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Table 5.1 Growth conditions and the relative compositions of the galfenol films using LiNbO$_3$ as a substrate. Films were analysed and RF sputtered using a power of 60W by David Czerski.

$a = b = 0.515$nm, and $c = 1.386$nm at 293K [144]. Lattice parameters of galfenol (Fe$_{85}$Ga$_{15}$) at 300K are $a = b = c = 0.287$nm [145]. For the D0$_3$ structure, this equates to 0.574nm, differing to the a and b directions of LiNbO$_3$ by $\approx 10\%$. The c direction of LiNbO$_3$ differs to that of galfenol by $\approx 59\%$ however.

In the following, all samples were provided and grown using RF sputtering by David Czerski (Heriot-Watt University), following a similar method to [146] but with a LiNbO$_3$ substrate. In particular, galfenol from a Fe$_{81}$Ga$_{19}$ target was deposited at room temperature on 1mm thick 128° X-cut LiNbO$_3$. Table (5.1) lists the thickness and relative composition of the six galfenol films that were accessible and measured using the developed MOKE system. They are broadly split into two categories depending on the time allowed for deposition: the ‘thin’ samples $\approx 250$nm and the ‘thick’ samples $\approx 600$nm. The orientation ($\phi$) of the plane of incidence/magnetic field, and sensitivity of MOKE measurements, relative to the y axis (1100) of the LiNbO$_3$ substrate is shown in Figure (5.3).

![Figure 5.3](image)

Figure 5.3 Orientation angle ($\phi$) of the plane of incidence (yellow) and magnetic field direction relative to the y axis of the LiNbO$_3$ substrate for the galfenol films.
5.2 Anisotropy in Hysteresis

Figure (5.4) displays longitudinal MOKE signals for s-polarised light ($\lambda=635$nm) reflected from 254nm thick Fe$_{84.5}$Ga$_{15.5}$ (dimensions of roughly 10mm × 10mm) for several orientations of the film plane when the external field is swept. The sample temperature is held at 300K during all measurements. The field is aligned with the film surface and the plane of incidence of light. For each sample orientation, the loops are calculated from the average of seven complete field sweeps, with red points representing the increasing field direction, and blue points the decreasing field direction. Error bars represent the standard deviation on the mean from the seven measurements. The film orientation ($\phi$) relative to the orientation of the $y$ axis of the LiNbO$_3$ substrate is included in the top left corner of each plot. It should be noted that the general shape of the magnetic hysteresis was found to be independent of the position that light was focused on the sample surface (at least more than 10$\mu$m away from edges).

As the film is rotated from 0° to 90°, both $\Delta \theta_K(B)$ and $\Delta \epsilon_K(B)$ show a transition from a sharp transition with large hysteresis, to a smooth change with little enclosed loop area. The observed orientation dependence of the MOKE hysteresis indicates the presence of uniaxial anisotropy with the magnetic easy axis along the $y$ axis of LiNbO$_3$, in Figure (5.4a), and the hard axis 90° to this, in Figure (5.4f). For all film orientations the saturation signal in both $\Delta \theta_K$ and $\Delta \epsilon_K$ remains approximately constant, indicating that the saturation in magnetisation is identical for all orientations. The other two films listed in Table (5.1) with similar thicknesses showed the same orientation-dependent change in the shape of hysteresis. The linkage to the orientation of the LiNbO$_3$ substrate indicates a potential residual strain in the galfenol film that permeates to the surface that the MOKE measurement is sensitive to.
Figure 5.4  MOKE hysteresis loops for 254nm thick Fe$_{84.5}$Ga$_{15.5}$ at different film orientations relative to the $y$ axis of the LiNbO$_3$ substrate (noted in the upper left corner of each plot). Red points are for increasing field and blue points are for decreasing field.
Figure (5.5) shows plots of the loop areas for the Kerr rotation and ellipticity angles obtained from plots such as in Figure (5.4) for the 254nm thick Fe$_{84.5}$Ga$_{15.5}$ film. The loop area was calculated by subtracting the linear interpolation of the increasing field sweep from that of the decreasing field sweep, as fitting a traditional sigmoidal function was not always accurate for the odd-shaped loops as in the $\Delta \epsilon_K$ signal in Figure (5.4e). The red points in Figure (5.5) represent the calculated loop areas from measured hysteresis loops, and the blue points represent their 180° rotations.

The “figure-8” shape of the $\Delta \theta_K$ loop area shows the large increase in energy loss when the external field is aligned along the easy axis compared to the hard axis and is the typical shape a uniaxial material shows. The $\Delta \epsilon_K$ area however, shows a different picture. Whilst the overall loop area of $\Delta \epsilon_K$ is an order of magnitude less than $\Delta \theta_K$, the ellipticity shows a maximum energy loss at 45° to the observed easy axis in the Kerr rotation.

Similarly to Figure (5.4), Figure (5.6) shows MOKE field sweeps on a 664nm thick galfenol film for various film orientations. As this 664nm film is rotated from 0° to 90°, the change in shape of the hysteresis loops is far less marked for the thicker Fe$_{86.4}$Ga$_{13.6}$ film in Figure (5.6). In the limited range of Fe:Ga compositions

![Figure 5.5](image)

*Figure 5.5* Area calculated from longitudinal hysteresis measurements on 254nm thick Fe$_{84.5}$Ga$_{15.5}$ film deposited on LiNbO$_3$ for (a) Kerr rotation and (b) ellipticity angle. The areas were calculated from hysteresis loops as in Figure (5.4). Red points indicate the calculated areas, and the blue points indicate the same points rotated about 180°.
looked at here, the difference in the change in hysteresis for different sample orientations as measured by MOKE is independent of gallium concentration. With the only difference between Figures (5.4) & (5.6) being the thickness of the galfenol, the factor that seemingly controls the anisotropy measured is the galfenol film thickness.

Figure 5.6 MOKE hysteresis loops for 664nm thick Fe80.4Ga13.6 at different film orientations relative to the y axis of the LiNbO3 substrate (noted in the upper left corner of each plot).
5.3 In-Plane Switching

Looking at the ‘thin’ 254nm thick galfenol, the switching process is useful to
determine whether LiNbO$_3$ be used as a substrate for any application.

From two separate MOKE measurements, the in-plane (longitudinal) and out-
of-plane (polar) responses can be distinguished [147]. Rather than changing
the direction of the external field and making a traditional polar MOKE
measurement, only the longitudinal MOKE setup is needed, with hysteresis loops
being taken for positive (+$\theta_i$) and negative ($-\theta_i$) incidence angles. The separation
of longitudinal and polar responses stems from the first order approximation that
the MOKE signal is proportional to the magnetisation i.e. $k \cdot M$. For MOKE
measurements with equal but opposite incident angles not normal to the surface
($\pm\theta_i$), the polar component is common to both measurements and so may be
expressed for both Kerr rotation and ellipticity angle as

$$\theta_{K_{Polar}} = \frac{\theta_{K}^{+\theta_i} + \theta_{K}^{-\theta_i}}{2},$$  \hspace{1cm} (5.2a)

$$\epsilon_{K_{Polar}} = \frac{\epsilon_{K}^{+\theta_i} + \epsilon_{K}^{-\theta_i}}{2}. \hspace{1cm} (5.2b)$$

The longitudinal component however, is equal but opposite and may therefore
be given by the difference between the two measurements

$$\theta_{K_{Longitudinal}} = \frac{\theta_{K}^{+\theta_i} - \theta_{K}^{-\theta_i}}{2},$$ \hspace{1cm} (5.3a)

$$\epsilon_{K_{Longitudinal}} = \frac{\epsilon_{K}^{+\theta_i} - \epsilon_{K}^{-\theta_i}}{2}. \hspace{1cm} (5.3b)$$

These simple symmetry arguments allow more information to be gathered from
obtained hysteresis loops by the relatively easy reversal of the position that
incident light enters the objective lens. This may be realised by moving the
optical breadboard relative to the objective lens with the $x, y$ translator.

For the 251nm thick Fe$_{87.5}$Ga$_{12.5}$ film with an orientation angle of $\phi=61^\circ$, two
hysteresis loops for the same position on the sample but with reversed incidence
angle are displayed in Figures (5.7a) & (5.7b). Taking the difference and sum
of these hysteresis loops, according to Equations (5.3) & (5.2), provides the
longitudinal component of the Kerr rotation and ellipticity angle as in Figure
(5.7c), and the polar components as in Figure (5.7d). The flat response with
field of the polar contribution highlights how the switching process in this ‘thin’ galfenol film is confined to have moments only in the plane of the film, and no detectable polar component appears.

Figure 5.7  Hysteresis loops of 251nm thick Fe87.5Ga12.5 for (a) negative, and (b) positive incidence angles. The in-plane longitudinal (c) and out-of-plane polar (d) components are then extracted by combining the two hysteresis loops according to the equations included (and described in the text).
5.4 Domain Images Upon AC Demagnetisation

The anisotropy observed in the ‘thin’ galfenol films deposited on LiNbO$_3$ suggests that magnetic domains would form preferentially pointing along the easy axis. Domains themselves usually form naturally in order to reduce the total magnetic energy cost. Once the material has been placed under a field greater than its saturation however, domains may remain aligned to this field direction even when the external field is reduced to zero. In these circumstances, some sort of magnetic asymmetry must be injected into the films in order to introduce magnetic domains.

The LiNbO$_3$ substrate was found to be too brittle to mechanically bend to produce enough additional magnetostriction in the FeGa to promote the appearance of detectable magnetic domains. Instead, an AC demagnetisation routine along the hard axis was applied. This routine had the field oscillating between positive and negative values starting above the maximum switching field from the above hysteresis loops and decreasing in amplitude for each successive loop.

Figure (5.8) corresponds to the same 251nm thick Fe$_{87.5}$Ga$_{12.5}$ film as in Figure (5.4), however this plot shows the transverse MOKE measurements during a field sweep. In these transverse MOKE measurements, the external field is applied in the plane of the film but nearly perpendicular to the incidence plane of light (the incidence plane here is offset from the perpendicular position to highlight the following case). Following AC demagnetisation, the field is increased from zero (following the black curve) before completing a conventional field sweep (red points are for increasing field, blue points are for decreasing field). The difference between Figures (5.8a) & (5.8b) are the position on the film light was focused for the MOKE measurement. In each case, the relative change in $\theta_K$ immediately following AC demagnetisation is different, with Figure (5.8a) showing a constant Kerr rotation with increasing field, and with Figure (5.8b) reversing sign.

The general shape of the transverse $\theta_K$ hysteresis loop is in agreement with that expected from the Stoner-Wohlfarth model for in-plane switching, as described in Figure (2.3) [75]. The position-dependence and Stoner-Wohlfarth-like behaviour suggests the galfenol film has split into domains following AC demagnetisation, larger than the focused spot used for the MOKE measurements (4$\mu$m). The Stoner-Wohlfarth model only considers smooth spin rotation upon switching and
Figure 5.8  Transverse hysteresis loops for two locations on 251nm thick Fe\textsubscript{87.5}Ga\textsubscript{12.5}. The difference in behaviour of the Kerr rotation following AC demagnetisation (black points).

does not consider domain wall movement. It is therefore possible that domains are pinned at the MOKE measurement positions chosen, possibly due to local surface defects or composition variations.

To test this hypothesis, domain images can provide information on what is happening. Figures (5.9), (5.10), & (5.11) refer to three scans (denoted (a), (b), & (c)) across the FeGa surface displaying the change in Kerr rotation ($\Delta \theta_K$), ellipticity angle ($\Delta \epsilon_K$), and DC voltage ($\Delta \text{DC}$) respectively.

Figures (5.9a), (5.10a), & (5.11a) are the data from a single scan with a constant field of 150mT applied along the $+x$ direction, the easy axis. In this state, owing to the hysteresis loops in the above sections, the sample should be saturated along the direction of the field, and any change in signal should only be due to surface defects/changes in reflectivity of the surface. Any large surface defects show up in the DC scan, Figure (5.11a), which are also observable in the MOKE measurement scans Figures (5.9a) & (5.10a). For example, the scratch with negative slope centred at (120,500) is easily observable in all three images. Other locations of the same sample showed similar patterns, with MOKE signals broadly corresponding to the dc signals for both positive and negative DC fields applied during the scan.
Figures (5.9b) & (5.9c) show the change in Kerr rotation across approximately the same area of the FeGa surface following two separate AC demagnetisation sweeps (with the sample being uniformly magnetised with a 200mT field along the -x axis in-between to “reset” the magnetic structure). In both figures, stripe domains can be clearly distinguished when compared to the saturated scan in Figure (5.9a). In these figures, positive/negative changes in Kerr rotation correspond to domains with net magnetisation pointing along the +x and -x directions. The domains average roughly 30µm thick, and >150µm along the easy direction, x. The placement of domains across Figures (5.9b) & (5.9c) is broadly similar in the top half of the scan (above y=400µm) however, differences are evident in the bottom half, with domains having a positive Kerr rotation much preferred in Figure (5.9b). These similarities between the two scans indicate that in this FeGa film, domains may be manipulated to some degree however, some local conditions may dictate the preferred domain direction certain sections of the film may take. Local conditions could either be due to imperfections in the film surface (which may easily be reduced with correct storage/capping of the film), or they may appear from the local growth conditions of the substrate/film.
Figure 5.9  FeGa domain images for the Kerr rotation angle component along the $x$-axis. (a) constant $+150\text{mT}$ along $x$, (b) & (c) different scans after AC demagnetisation along the hard axis (in the $y$-direction).
Figure 5.10 FeGa domain images for the ellipticity rotation angle component along the $x$-axis. (a) constant $+150\text{mT}$ along $x$, (b) & (c) different scans after AC demagnetisation along the hard axis (in the $y$-direction).
Figure 5.11  The DC signal corresponding to the domain images presented in Figures (5.9) & (5.10). (a) constant +150mT along $x$, (b) & (c) different scans after AC demagnetisation along the hard axis (in the $y$-direction).

5.5 Discussion

The above plots illustrate an in-plane anisotropy prevalent in $\sim 250$nm thick samples of FeGa deposited on LiNbO$_3$, and magnetic isotropy in $\sim 600$nm thick samples. There is seemingly a crossover in anisotropy in-between 300-600nm
thickness of FeGa films when lithium niobate is used as a substrate under the growth conditions used as highlighted in Table (5.1).

Evidence of in-plane uniaxial anisotropy in FeGa thin films has previously been observed after applying a dc magnetic field during sputtering [131]. In the above study, DC magnetron sputtering was used with Si (100) as a substrate. In this case, 660nm thick Fe$_{81}$Ga$_{19}$ films were grown using zero applied external field and a DC field of 550Oe for which the corresponding hysteresis loops measured using a VSM are included in Figure (5.12a). With no applied field during sputtering, the Fe$_{81}$Ga$_{19}$ film has an isotropic response in the plane of the field. Applying a field during sputtering however, creates a slight anisotropy in the plane of the film, reducing the saturation magnetisation by about 13% along the direction perpendicular to the growth field, with no change in the coercivity. In comparison, the MOKE hysteresis measurements here showed an average decrease in saturation of $\Delta \theta_K$ by $\sim$28%, from 0.39mrad in Figure (5.4a) to 0.28mrad in Figure (5.4f), whilst the average $\Delta \epsilon_K$ increased by $\sim$50%, from 0.08mrad to 0.17mrad. These differences in MOKE signal are illustrated by combining Figures (5.4a) & (5.4f) into one plot, as in Figure (5.12b).

The difference in the $\theta_K(B)$ and $\epsilon_K(B)$ response of the galfenol films is interesting. The change in Kerr rotation is generally larger than the change in ellipticity.

![Figure 5.12](image-url) (a) VSM measurement of 660nm thick Fe$_{81}$Ga$_{19}$ deposited using DC magnetron sputtering with a constant 5500e field applied determining the easy axis (from [131]). (b) Combined MOKE hysteresis plots extracted from Figures (5.4a) & (5.4f) with the film orientation indicated.
angle, where there is a difference of \( \sim 40-80\% \) between \( \theta_K(B) \) and \( \epsilon_K(B) \) along the easy and hard axes in 254nm thick \( \text{Fe}_{84.5}\text{Ga}_{15.5} \) and \( \sim 28\% \) for 664nm thick \( \text{Fe}_{86.4}\text{Ga}_{13.6} \). From just the MOKE data, interpreting the change of magnetic response in galfenol deposited on \( \text{LiNbO}_3 \) is difficult. For the 664nm thick films, the constant ratio of \( \theta_K(B):\epsilon_K(B) \) is perhaps more indicative for unstrained galfenol, where the small \( \sim 28\% \) difference is due to absorption rates and propagation speeds of right and left circular polarisation states for \( \lambda=635\text{nm} \). In 254nm films however, the large difference in \( \theta_K(B):\epsilon_K(B) \) of \( \sim 40-80\% \) between easy and hard axes indicates an asymmetry in the crystal structure that arises due to magnetostriction. This change in structure may lead to a change in the allowed inter- and intra-band transitions for the measurement frequency, as discussed in Section (2.2.3). Such a structure change should be further investigated with alternative techniques, however.

Alternative origins of in-plane anisotropy in thin film galfenol are found for varying the growth conditions. With cubic \( \text{MgO} \) (100) as a substrate, high sputtering powers (>35W) were shown to induce anisotropy in FeGa, measured using ferromagnetic resonance [135]. In this situation the saturation magnetisation was unchanged along the hard and easy axes. The sputtering power of 60W used here does satisfy the limit set out by [135] however, the diminishing anisotropy with film thickness observed here indicates that the sputtering power is not solely responsible. At the very least, \( \text{LiNbO}_3 \) is significant in producing the anisotropy.

Changes in the argon pressure during sputtering onto \( \text{Si} \) (100) has also been observed to induce an in-plane anisotropy [133]. The authors found a negative correlation between \( M_{\text{remanence}}/M_{\text{saturation}} \), and a positive correlation for the required saturation field for the range \( P_{\text{Ar}} = 3-7\mu\text{bar} \). This amounts to an increase in film strain with increased Ar pressure. Films grown with \( P_{\text{Ar}} < 3\mu\text{bar} \) were additionally magnetically isotropic. In the films studied with MOKE in this thesis, there was no indication of an Ar pressure dependence on magnetic hysteresis from the three available samples of thickness \( \sim 250\text{nm} \), however. \( P_{\text{Ar}} = 2, 4, 6\mu\text{bar} \) were used for growths and all three samples showed similar anisotropy in the response of \( \Delta \theta_K \) & \( \Delta \epsilon_K \) to an external field.
5.6 Surface Acoustic Wave Device

SAW devices similar to Figure (5.2) were constructed by David Czerski using LiNbO$_3$ and galfenol films in the observed ‘thin’ regime. Titanium was used for the IDTs which run at a frequency of 20MHz (which is controlled by the separation distance of arms in the IDTs). The IDTs are connected to the output of a MHz signal generator through an LC filter, acting as a matching network and reducing reflections of the electronic signal at the connection. The circuit is illustrated in Figure (5.13a). The corresponding wavelength of SAWs in this device is 200µm, as determined by the IDT arm separation. The acoustic waves can be visualised by sprinkling on 7µm silica micro-spheres (Whitehouse Scientific), applying a signal of 10V$_{\text{peak-peak}}$ at 20MHz to the IDTs and lightly blowing compressed air over the surface. The micro-spheres coalesce at nodes as in Figure (5.13b).

Despite being able to see the SAW wave using the micro-spheres, efforts to view/manipulate the domain pattern on the LiNbO$_3$ devices were unsuccessful. Figure (5.14) shows the typical hysteresis loop with the SAW turned off (red and blue points). This was collected using a plane of incidence and external field applied along the vertical direction in Figure (5.13b). A large number of slow field sweeps about the positive switching field (+5.3mT) were carried out with light focused at different locations with the reflection plane in the vertical direction in Figure (5.13b) (perpendicular to the direction that the standing SAW is set up.

![Figure 5.13](image)

**Figure 5.13** (a) Circuit diagram for the SAW device showing the matching network consisting of a trimmer capacitor and inductor. (b) The SAW visible with 7µm silica micro-spheres deposited on the devices used here. The vertical axis is the easy magnetic axis of the galfenol film.
in the galfenol). Two of such field sweeps are included in Figure (5.14) as the purple and green points. These two had light focused at points separated by the distance between nodes and anti-nodes (50 µm). For all positions measured, the switching field was identical and irrespective of whether the SAW was applied or not.

Application of a field that was 85% of the un-strained coercivity was adequate to form stripe domains in 57nm thick Fe$_{81.6}$Ga$_{18.4}$ deposited on quartz [141]. Here, for 250nm thick Fe$_{85}$Ga$_{15}$ deposited onto LiNbO$_3$, there was no observable change in the required coercivity within 1%, as measured by MOKE. It should be noted that aligning the field and plane of incidence for the MOKE measurement along the hard axis of the SAW device was also attempted to no avail. Additionally, scans using the MOKE setup as in Figure (5.9) with a constant field applied at various values about the measured coercivity (±5.3mT) proved unsuccessful in imaging any induced magnetic structure from the SAW.

The lack of success in observing SAW-induced magnetic structure could be a result of the anisotropy in the galfenol film from the LiNbO$_3$ substrate, preventing the SAW from inducing enough magnetostriction in the film. Alternatively, the galfenol films here may be too thick to have the magnetostriction permeate to the upper surface that the MOKE measurement is sensitive to. An additional improvement may be found by tuning the Fe:Ga ratio closer to the peak

![Figure 5.14](image_url)

**Figure 5.14** Hysteresis loop of unstrained galfenol (red & blue points) compared to field sweeps around the positive switching field for two locations separated by 50µm along the SAW modulation direction with an applied SAW (purple & green).
magnetostriction value found at 19% Ga.

5.7 Conclusion

In this chapter, the developed MOKE microscope has been used to investigate the magnetic properties of galfenol films deposited onto a 128° X-cut lithium niobate substrate. The presented MOKE measurements on galfenol are the first to distinguish between $\theta_K$ and $\epsilon_K$. From both MOKE angles, a source of uniaxial anisotropy in FeGa is evident and correlates to the alignment of the LiNbO$_3$ substrate. This anisotropy was observed in $\sim$250nm thick films but not in $\sim$600nm thick films. In 254nm thick Fe$_{84.5}$Ga$_{15.5}$, an increase in $\Delta\theta_K(B)$, from 0.56mrad to 0.78mrad, was measured along the easy axis compared to the hard axis. Conversely, a decrease in $\Delta\epsilon_K(B)$, from 0.34mrad to 0.16mrad, in the same film was measured. In 664nm thick Fe$_{86.4}$Ga$_{13.6}$, both $\theta_K(B)$ and $\epsilon_K(B)$ remained similarly shaped. A decrease in $\Delta\theta_K(B)$, from 0.72mrad to 0.60mrad, and a similar decrease in $\Delta\epsilon_K(B)$, from 0.52mrad to 0.44mrad, was observed.

One possible origin of the measured surface anisotropy is a mismatch of lattice parameters between the galfenol and lithium niobate, inducing a strain that permeates through the galfenol but decays between 300-600nm. The strength of this conclusion is weak however, as only two broad thicknesses of galfenol films were available, grown under limited conditions. Previous published investigations into induced anisotropy during sputtering of galfenol films include the variation of sputtering power or argon pressure. However, the MOKE measurements on the available films here do cast doubt on the possibility of these explanations being valid for FeGa deposited on 128° X-cut LiNbO$_3$. X-ray diffraction or a high energy electron diffraction technique performed on various thicknesses of FeGa deposited onto LiNbO$_3$ could reveal the surface structure of FeGa, and how this contributes to the observed anisotropy.

Stripe domains were additionally successfully induced and imaged using the MOKE microscope in the studied galfenol films by AC demagnetisation along the hard axis of the films. AC demagnetisation along the easy axis was not successful in inducing domains, however. This could be due to an energetic favour for stripe domains to be slightly canted relative to each other due to a distortion of the crystal structure from magnetostriction, or that demagnetisation along the easy axis results in very large domains for the film thickness. There is no direct
evidence for either of these suggestions however, so additional measurements to analyse the structure should be made. Despite the demonstration of domains within the films, manipulating such domains with galfenol on LiNbO$_3$ as part of a SAW device has been unsuccessful. Possible changes that could promote the detection of SAW-induced magnetic domains include decreasing the film thickness from 250nm, and changing the stoichiometry of galfenol from Fe$_{85}$Ga$_{15}$ closer to the peak in magnetostriction found with 19% Ga.
Chapter 6

Mn$_3$Sn

This chapter is focused on the hexagonal antiferromagnet Mn$_3$Sn. The sample preparation, bulk-sensitive magnetic characterisation, and surface-sensitive MOKE measurements using the developed MOKE system on single crystals of Mn$_3$Sn are presented.

6.1 Background

MOKE and the Faraday effect are most commonly observed in metals with a net magnetisation - either in ferromagnets, ferrimagnets or canted antiferromagnets. The recent discovery of a large anomalous Hall effect (AHE) (∼20Ωcm at 300K) in the non-collinear antiferromagnet Mn$_3$Sn [148], a metal with a very small net moment, has stirred interest in this and its isostructural companions Mn$_3$Ge and Mn$_3$Ga. This interest led to the measurement of a large zero field polar MOKE signal of 0.35mrad in Mn$_3$Sn [90]. The relatively soft magnetic response, requiring a switching field of ∼30mT [148], coupled with potential fast spin dynamics and inherently small stray fields offers a path for new non-volatile data storage and processing technologies that exceeds those of current ferromagnetic materials in use.

The crystal structure of hexagonal Mn$_3$Sn (space group P6$_3$/mmc) is illustrated in Figure (6.1a); the corresponding lattice constants are a=b=5.671Å and c=4.536Å [149]. Kagome lattices of Mn moments as illustrated in Figure (6.1b) are stacked in planes along the (0001) direction, with an inverse triangular spin
structure. Within each Mn spin triangle, only one moment points along the easy axis - towards the nearest in-plane Sn site - with the other two moments slightly canting towards the easy axis. This slight canting results in geometrical frustration that slightly distorts the kagome lattice, visible by the 0.01Å difference in distance between Mn moments along (2110) in Figure (6.1b), and leads to a net ferromagnetic moment of $\sim 0.002 \mu_B$ per Mn atom.

The spin structure in Mn$_3$Sn varies with temperature. Below the Néel temperature of 420K, there is a further magnetic transition at 50K, where Mn$_3$Sn enters a spin glass phase and spin canting towards the c axis results in a larger ferromagnetic moment [150]. Between these two temperatures, a transition from the inverse triangular state to an incommensurate structure occurs at 270K upon cooling when samples are slightly Mn-deficient. Within the spiral spin structure at around 200K there is an additional magnetic transition observed by magnetic torque measurements which is attributed to a crossover in the easy axis direction between (1120) and (0001) [149]. For samples with excess Mn, the magnetic transitions between 50K-420K may not be present [148].

The magneto-optic response of Mn$_3$Sn was measured in [90] by a commercial NanoMOKE3 (Quantum Design) system at a near normal incidence angle for polar MOKE measurements and 45° for in-plane measurements using a 660nm laser diode. The measured polar Kerr angle along (2110) between positive and negative saturation was $\sim 0.6\text{mrad}$, with a coercive field of $\sim 12\text{mT}$. The wavelength dependence of the zero field polar Kerr angle along (2110) is included in Figure (6.2b), showing a peak in rotation around 600nm.
Within the (01\(\overline{1}\)0), (0001) plane, the measured longitudinal Kerr rotation shows a uniaxial anisotropy [90] which is expected since the Mn moments remain in the hexagonal plane. This anisotropy is shown in Figure (6.2a), with \(\phi\) being the angle from the (01\(\overline{1}\)0) axis. The sharp transition in hysteresis along (01\(\overline{1}\)0) of \(\sim0.38\text{mrad}\) between \(\pm\)fields to a hard-axis-like loop along (0001) with a non-zero slope is as expected. Between these two orientations, the hysteresis shows more structure. For example in the pink \(\phi=60^\circ\) curve in Figure (6.2a), increasing the field from negative saturation shows a sharp drop of \(\theta_K=5\text{mdeg}\) to \(1\text{mdeg}\) at \(40\text{mT}\), followed by a smooth decrease until \(\theta_K=-5\text{mdeg}\) and a further sharp drop to \(-6\text{mdeg}\) at \(130\text{mT}\). The origin of this complicated hysteresis is not explained by the authors however, as the MOKE measurement is locally sensitive due to the focus of the probing light, sharp jumps in the signal may be caused by domain walls passing through the focus position. The movement of AF domain walls in Mn\(_3\)Sn has previously been postulated to be the source of a real space Berry curvature, providing an additional component to the AHE [151]. This was observed from the difference between magnetisation and AHE hysteresis loops and attributed to the inhomogeneous magnetisation within the AF walls, similar to topological Hall effects in other materials.

Both the AHE and MOKE signals are proportional to the off-diagonal components of the conductivity tensor \(\sigma_{xy}(\omega)\). They may also be ascribed to the AF spin structure which, in Mn\(_3\)Sn, can be switched along with the small residual
moment of $\sim 0.002 \mu_B$. As such, the MOKE and AHE in Mn$_3$Sn are expected to be more or less in agreement. Differences between the Kerr rotation and AHE measured however, have recently been observed. In [152], the AHE and longitudinal Kerr rotation in Mn$_3$Sn have been measured. Kerr rotations were measured by balanced photodiodes, as outlined in Section (1.1.1), using a 633nm p-polarised laser diode with a 45° incidence angle.

Figure (6.3a) shows the difference in coercive fields as measured by the AHE and MOKE: the Hall resistivity has a coercivity of $\sim 50$ mT whereas the coercivity as measured by the Kerr rotation is $\sim 120$ mT. As the temperature is decreased, the MOKE signal becomes undetectable at 267K (Figure (6.3b)) whilst the AHE remains until 261K (Figure (6.3c)) where both measurements become independent of the applied field within $\pm 300$ mT. The same authors also measured the temperature dependence of the amplitudes of $\rho_H$ and $\theta_K$. A discrepancy between the temperatures that both amplitudes decreased to zero was observed - at $\sim 270$K for $\rho_H$ and $\sim 260$K for $\theta_K$. Similarly, they observed a difference of $\sim 10$K between the AHE and MOKE sensitivity of the Néel temperature at 420K and 410K respectively. The authors ascribe this temperature difference to a different magnetic behaviour in the surface as compared to bulk Mn$_3$Sn [152].

Whilst Higo et al. observe a consistent coercivity between the Kerr rotation and bulk magnetic measurement [90], Balk et al. note a difference in coercivity of over a factor of two between AHE and MOKE measurements [152].
difference could be attributed to the difference between Mn concentrations in the two studies: Higo et al. use crystals with excess Mn (Mn$_3$Sn$_{0.94}$) whereas Balk et al. use Mn-deficient crystals (as is typical for most grown Mn$_3$Sn crystals). The other difference between the two longitudinal MOKE studies is the orientation of Mn$_3$Sn relative to the incoming p-polarised light which may also factor into this discrepancy. Both studies measure the Kerr rotation along the (0110) direction however, Higo et al. measure the (2110) surface and Balk et al. measure the (0001) surface.

Magneto-optic imaging of the domain evolution of the (2110) surface in Mn$_3$Sn by widefield MOKE microscopy has been published [90]. Growth of the antiferromagnetic domains during the switching process between $\pm 21\text{mT}$ was imaged, with domains $>10\mu\text{m}$ being observable. Local magnetic structure has also been imaged in rectangular 50nm thick films of Mn$_3$Sn using scanning thermal gradient microscopy [153], essentially measuring the thermo-voltage due to the focusing of a 10mW $\lambda =800\text{nm}$ laser at positions along the film. The same authors also demonstrated writing of domains $>10\mu\text{m}$ in the Mn$_3$Sn film when focusing a 50mW laser and applying a 0.5T field, as imaged by the thermo-voltage signal.

The above MOKE studies on Mn$_3$Sn have only reported the Kerr rotation, $\theta_K$, [90, 152] and it is not clear whether these measurements distinguish between the ellipticity angle, $\epsilon_K$, or whether the measurements are sensitive to the combined complex Kerr angle, $\theta_K + i\epsilon_K$. Thus, the MOKE microscope developed in this thesis has been used to separate the ellipticity and Kerr contributions to measured MOKE signals.

### 6.2 Sample Preparation

Two batches of single crystals of Mn$_3$Sn were grown and provided by the Christoph Geibel group (Max Planck Institute for Chemical Physics of Solids, Dresden) and Klára Uhlířová (Charles University in Prague). The latter of the two were grown using a flux growth method with an Sn-rich flux in an Al$_2$O$_3$ crucible, as in [150]. Whilst MOKE measurements are included only for those provided by the Christoph Geibel group, measurements on the second batch of samples were consistent with these. An in-house X-ray Laue diffractometer was used to confirm the orientation of the provided samples, and to align the single
crystals for cutting. The Mn$_3$Sn crystal was mounted onto a three-axis goniometer during the capture of the Laue diffraction image.

Figure (6.4) is one such Laue backwards diffraction image upon exposure from a 3kW Mo X-ray source for 30 minutes operated at 35kV and 30mA. Diffracted spots that satisfy the Bragg condition can be distinguished by eye. The program “Orient Express” was used to fit the calculated diffraction image by inputting the crystallographic parameters of Mn$_3$Sn and selecting several spots on the image in Figure (6.4). For the given image, the c axis (0001) is parallel to the beam (and normal to the page).

The sample, unmoved from the same goniometer stand the Laue image was recorded on, could then be transferred to an in-house spark eroder for cutting desired surfaces. The combination of the fit obtained from the Laue image, and the mounting goniometer allows planes to be selected and cut to an accuracy of 0.1°. In the case of this Mn$_3$Sn crystal, (0110), (2110) and the original (0001) surfaces were cut to be measured by the MOKE microscope.

Before the optical MOKE measurement however, samples need to be polished to a mirror finish, with surface flatness less than the wavelength of probing light.

**Figure 6.4** Laue diffraction image of Mn$_3$Sn oriented with the c axis (0001) out of the page.
(635nm in this case). Polishing was done by hand using the equipment shown in Figure (6.5). The sample is adhered to a sapphire puck using superglue (wax may also be used to hold the sample in place). This sapphire puck is inserted into an epoxy resin case that holds the puck flat. This case is then secured into the brass holder using four socket screws. The brass holder is large enough to be easily gripped by hand and its weight provides a constant downwards force of \(\sim 4\text{N} \) on the sample surface during polishing. Hand polishing is most consistent when applying a constant downwards force onto a flat surface (either a granite or glass plate), and using continuous movements of “figure-8” or circular motions. For the finer polishing steps, a piece of letter paper placed between the flat surface (which, depending on its history, may have fine scratches) and the polishing pad helps to maintain an even pressure on the sample surface.

The underside of the sample is first polished roughly with 10\(\mu\text{m} \) SiC paper. The sample is then released from the sapphire puck by dissolving the superglue in acetone in an ultra-sonic bath, flipped over and re-adhered to the sapphire puck. This ensures that the polished surface is parallel to the back surface so that the sample lays flat relative to incoming light when mounted for the MOKE measurement.

Figure (6.6) details the effect of the different stages of polishing that, through trial and error, were found to work well for the Mn\(_3\)Sn crystals. In all stages, ethanol as a lubricant was adequate however, it does require regular reapplication to prevent complete evaporation. Initially the sample is roughly polished using...
Figure 6.6  Polishing stages for Mn$_3$Sn. Grade of polishing paper used from left to right: pre-polishing, 10µm SiC, 5µm SiC, 1µm C, $\frac{1}{2}$µm C, $\frac{1}{2}$µm C.

10µm SiC film (Struers grinding paper) to remove any surface defects. 5µm SiC film is next used, followed by 1µm diamond film, and finally $\frac{1}{2}$µm diamond film. Ideally, for each stage, polishing should continue until the surface appears homogeneous with scratches of equal thickness to the grade of film used. This is difficult in practice due to mistakes and stray particles getting in the way. Larger scratches may also be buffed out in subsequent stages albeit at a cost of time. Polishing continues until a scratch-free mirror-like surface is achieved as in the far-right of Figure (6.6). The time required for each polishing stage increases with decreasing polishing particle size; each stage generally takes 2-15 minutes but can depend on the material being polished, downward force applied, and experience.

Final polishing with different combinations of pads (Struers MD-Dac, DP-Floc, MD-Chem, OP-Nat) and solvents/pastes (Struers Diapro $\frac{1}{2}$µm, OP-U 0.04µm, OP-S 0.04µm, DP-Paste P $\frac{1}{2}$µm) was carried out. However, these gave no discernible improvement (in measurement signal or visually under a high-powered microscope) over the $\frac{1}{2}$µm diamond film polishing stage. Should a resolution much greater than that used here (4µm) be required, finer polishing may be necessary.

The magnetic properties of the samples were characterised to compare to previously published results. Magnetisation was measured using a superconducting quantum interference device (SQUID) within a commercial magnetic properties measurement system (MPMS-XL from Quantum Design). The sample is held in place between two cling film covered plastic straws within another clear plastic straw. Both cling film layers are coated in a thin layer of vacuum grease to hold
Figure 6.7 (a) Magnetic hysteresis of Mn$_3$Sn measured at 295K with a SQUID for external field aligned along the directions (2110), (0001), and (0110) placed in the inset (same axes) for clarity. (b) How the sample is mounted between two vacuum grease-coated cling film layers in the MPMS.

the sample in place. This setup is illustrated in Figure (6.7b). It should be noted that this measurement setup is not ideal, and the alignment of the sample axis with the applied magnetic field can only be accurate to within a few degrees.

Figure (6.7a) shows the magnetisation as a function of applied field along three directions in Mn$_3$Sn. The (01\overline{1}0) was measured on a 1.80mg cut of single crystal Mn$_3$Sn and the (2\overline{1}10) and (0001) directions were measured on a 6.60mg cut of the same crystal. These results show similar shaped magnetisation curves along (01\overline{1}0) and (2\overline{1}10) with a linear field dependence above 0.2T that is in agreement with \cite{148, 150}. Along the (0001) direction however, the small hysteretic behaviour measured in Figure (6.7a) is different to the linear response in the literature. The cause of this difference is likely to be a misalignment of the Mn$_3$Sn as measured here due to the mounting method, meaning a component along a different axis is being included in the plotted data.

In contrast to MOKE measurements, the SQUID magnetometer is sensitive to the bulk of the sample and not just the surface layer. As such, the comparison between Figure (6.7a) and published results serves to highlight how the bulk of the available Mn$_3$Sn here are broadly in agreement.
6.3 Surface Tarnishing

Following the polishing of Mn$_3$Sn single crystals, the surface was observed to degrade over a timescale of $\sim$ two weeks. Figure (6.8a) is a microscope image showing this tarnishing that occurred when the crystals had been left in air for two weeks after polishing. This surface damage affected the MOKE measurements, decreasing the measured dc signal and hence the surface reflectivity over time.

An energy-dispersive X-ray spectroscopy (EDS) measurement was undertaken (with the help of Thomas Glen) to compare a freshly polished (0001) Mn$_3$Sn

![Microscope images of Mn$_3$Sn surface](image)

**Figure 6.8** (a) Degradation of the freshly polished (LEFT) Mn$_3$Sn surface after two weeks in air (RIGHT). (b) SEM images and the EDS line scan of freshly polished Mn$_3$Sn, and (c) EDS line scan for a single crystal from the same growth as in (b) but having been left in air for two weeks after polishing.
surface with a separate cut from the same crystal that had been polished and then left in air for two weeks. Figure (6.8) highlights the results from the EDS analysis. Figure (6.8b) represents a line scan (as superimposed on the SEM image of the surface) of EDS measurements that distinguish local atomic proportions over 10µm. As determined by the EDS, the average atomic percentages of the freshly polished sample were 74.78% Mn and 25.22% Sn. Conversely, for the cut that had been left in air for two weeks, Figure (6.8c) shows how the surface looks rough over this micron scale. Comparing the image of the line scan with the EDS measurement, Sn inclusions appear to have conglomerated on the surface, at a size of ∼0.6µm. This is in addition to an oxide layer having formed on the surface. These Sn inclusions can faintly be seen on the freshly polished sample in Figure (6.8b) (which itself has been exposed to air for three hours before the EDS measurement). It is probable that either there is an excess of Sn that is diffusing to the surface, or that the oxidation of the surface is removing the top surface layers but leaving these Sn clumps exposed.

This layer of tarnish could not be removed with ethanol or acetone, and re-polishing of the surface was required to return it to a mirror finish. Keeping Mn$_3$Sn in a protected atmosphere to prevent tarnishing is therefore vital to ensuring surface stability over the timescale of weeks. Some sort of electro-polishing or another chemical treatment could alternatively be attempted to remove the tarnished surface layer in single crystal Mn$_3$Sn. This would have to be repeated frequently in-situ for any long-running experiment, however. Recently for STM measurements, cleaving within a vacuum followed by a 10V pulse on the Mn$_3$Sn has been found to smooth the surface to be locally atomically flat [154].

### 6.4 MOKE Measurements

The following longitudinal MOKE measurements have been carried out at 295K in air with p-polarised light (λ =635nm) in the same plane that the field is applied. This is the same orientation used in previous reported MOKE measurements on Mn$_3$Sn [90, 152]. Here, sample orientation is controlled with an Attocube ANR-100 piezoelectric rotator with a resistive feedback mechanism (as imaged in Figure (3.2)). The light is focused to a 4µm spot at an incidence angle of ∼25°, as determined by the NA=0.5 reflective objective used, which is less than the ∼45° incidence used in the above previous MOKE studies. The expected change in longitudinal MOKE signal observed here should therefore be $\Delta \theta_K < 0.11$ mrad as
measured along (2\bar{1}10) for a surface (0001) cut [152], and $\Delta \theta_K < 0.38 \text{mrad}$ along (01\bar{1}0) for a (2\bar{1}10) surface cut [90].

Figure (6.9) displays longitudinal hysteresis loops measured on different cuts from the same single crystal along two directions. Each plot is the average of ten individual loops obtained within four hours of the surface being polished and the error bars represent the standard deviation over these measurements. Figure (6.9a) represents MOKE measured along (2\bar{1}10) of a (01\bar{1}0) surface, and Figure (6.9b) represents MOKE measured along (01\bar{1}0) of a (2\bar{1}10) surface. Photographs of these two orientated cuts are included in Figure (6.10a) and Figure (6.12a) respectively. Both orientations have the (0001) axis, the hard magnetic axis, in the plane and so hysteretic signals are maximised when measuring 90° to this, as observed in previous AHE and MOKE measurements [90, 148, 151, 152].

Along both (2\bar{1}10) & (01\bar{1}0) directions, the $\theta_K$ signal is too noisy to distinguish any hysteretic signal. Figure (6.9b) has the same measurement geometry as [90], who report $\Delta \theta_K = 0.38 \text{mrad}$ between opposite saturation directions. Here, there does not appear to be any hysteretic signal within $\Delta \theta_K = 0.10 \text{mrad}$ along (01\bar{1}0) or (2\bar{1}10). In the $\epsilon_K$ signal however, hysteresis is visible along both (2\bar{1}10) & (01\bar{1}0) which have similar shaped loops. Figure (6.9a) has $\Delta \epsilon_K \approx 0.12 \text{mrad}$ which

![Graphs](image_url)

**Figure 6.9** Measured longitudinal Kerr rotation and ellipticity angle as a function of external field on (a) (01\bar{1}0) surface, and (b) (2\bar{1}10) surface, of Mn$_2$Sn. The plane of incidence and magnetic field is aligned along (2\bar{1}10) and (01\bar{1}0) respectively.

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becomes saturated at ±150mT and Figure (6.9b) has a lower $\Delta \epsilon_K \approx 0.06$ mrad with a saturating field of ±120mT. This larger measured signal along (2110) is in contrast to the slightly larger magnetisation and Hall signal as measured along (0110) compared to (2110) [148].

Following on from Figure (6.9a), Figure (6.10b-m) extends the field dependence of the longitudinal ellipticity signal for a variety of angles from the (2110) direction at 0° to (0001) at 90°. The ellipticity displays hysteresis between 0°~10°, after which the resolution of $\Delta \epsilon_K$ is not fine enough to distinguish details. Figure (6.11a-l) displays the corresponding $\Delta \theta_K$ loops for the same angles in Figure (6.10b-m) for completeness. No hysteretic behaviour can be observed at any angle in $\Delta \theta_K$.

Similarly, longitudinal ellipticity measurements following on from the observed hysteresis in Figure (6.9b) are shown for a number of different angles from the (0110) direction at 0° to (0001) at 90° in Figure (6.12b-m). The distortion that is symmetric in field for some of the loops is discussed below. The hysteretic behaviour is mostly gone by 18° in Figure (6.12f) however, at 28° in Figure (6.12g) there appears to be a finite loop area, with saturating field $\sim$-300mT. This could be an anomaly however, as it was not systematically reproducible. From Figure (6.12j-m), the crystal surface was re-polished and re-mounted onto the MOKE microscope, and the maximum field applied was increased. From here the ellipticity angle was constant with field within the spread of $\sim 50\mu$rad. Figure (6.13a-l) finally displays the corresponding $\Delta \theta_K$ loops obtained concurrently with those in Figure (6.12b-m). No hysteretic structure in $\Delta \theta_K$ was observed for any angle in the (2110) surface.
Figure 6.10 (a) Polished Mn$_3$Sn single crystal with (0110) surface. (b–m) Change in measured longitudinal ellipticity angle with field when reflected from the centre of the crystal in (a). The angle $\phi$ between the incidence plane of $p$-polarised light from the (2110) axis is indicated in the top right corner of each plot. Error bars represent the standard deviation from averaging over ten loops.
Figure 6.11 (a-l) Change in measured longitudinal Kerr rotation with field when reflected from the centre of the crystal in Figure (6.10a). The angle between the incidence plane of p-polarised light from the (2110) axis is indicated in the top right corner of each plot. Error bars represent the standard deviation from averaging over ten loops.
Figure 6.12  (a) Polished Mn$_3$Sn single crystal with (2110) surface.  (b-m) Change in measured longitudinal ellipticity angle with field when reflected from the centre of the crystal in (a). The angle $\phi$ between the incidence plane of $p$-polarised light from the (0110) axis is indicated in the top right corner of each plot. Error bars represent the standard deviation from averaging over ten loops for (a-h) and seven loops for (i-l).
Figure 6.13  (a-l) Change in measured longitudinal Kerr rotation with field when reflected from the centre of the crystal in Figure (6.12a). The angle between the incidence plane of p-polarised light from the (0110) axis is indicated in the top right corner of each plot. Error bars represent the standard deviation from averaging over ten loops for (a-h) and seven loops for (i-l).
6.5 Discussion

The longitudinal MOKE measurements presented here show hysteretic behaviour only in the ellipticity angle, $\epsilon_K$. The large spread in Kerr rotation as a function of external field, for example in Figure (6.9), obscures any actual $\Delta\theta_K$ signal $< 100\mu$rad. The measured $\Delta\epsilon_K$ here is 32% of $\Delta\theta_K$ as measured by [90] along $(01\overline{1}0)$. From the unresolved $\Delta\theta_K$ signal, and considering the steeper incidence angle used here, the magnitude of MOKE signals are in broad agreement. The $\theta_K$ signal in Mn$_3$Sn reported by others [90, 152] does not necessarily distinguish between the ellipticity angle $\epsilon_K$. The measurement can instead be sensitive to the magnitude of the complex Kerr angle, $|\theta_K + i\epsilon_K|$, depending on the exact experimental setup. Here, the larger observed ellipticity signal along $(2\overline{1}10)$ compared to $(01\overline{1}0)$ needs further investigating as there are no other systematic studies to compare the current MOKE data to.

Both $\theta_K$ and $\epsilon_K$ depend on the real and imaginary components of the optical conductivity tensor and there is no analytical method for separating each component. Therefore, the physical significance of a larger ellipticity angle compared to the Kerr rotation as observed in Mn$_3$Sn here is not simply put. The larger measured $\epsilon_K(B)$ response is likely linked to the surface structure and the allowed inter-band transitions of Mn$_3$Sn for the used frequency of light (corresponding to $\lambda=635$nm). Previous measurements of the Kerr rotation have been compared to the AHE [90, 152] - the real part of the off-diagonal element of optical conductivity at zero frequency, $\Re(\sigma_{xy}(\omega = 0))$. Similar to the published density functional theory calculations of MOKE in the cubic antiferromagnets Mn$_3$X (X=Rh, Ir, Pt) [155], calculations of the full optical conductivity tensor in the hexagonal Mn$_3$Y (Y=Sn, Ge) system would aid in the interpretation of the observed ellipticity signal here, and Kerr signal elsewhere [90, 152].

The most obvious shortcoming of the presented longitudinal MOKE data on Mn$_3$Sn is the irregular shape of some hysteresis loops. Many of the presented $\Delta\theta_K(B)$ and $\Delta\epsilon_K(B)$ plots show symmetric (quadratic) behaviour in field. This was not always repeatable at any given sample orientation but was found to be position-dependent on the sample surface. The Voigt effect, which is symmetric in field, is one possible explanation although no other studies have alluded to this. The cause could instead be a slight movement of the objective-sample mount. The objective lens used here has a stainless-steel component which does feel a magnetic force. A very slight shift with field can be observed using the widefield
imaging on the MOKE microscope however, this results in a fraction of a micron movement of the focused spot on the sample surface. Whilst for most samples, a very small movement will not affect the measured MOKE signal, with the Mn$_3$Sn crystals here, Sn inclusions are present as outlined in Section (6.3). Figure (6.8c) shows the size of Sn clumps to be up to 0.6µm and they can be densely placed. Hence, a movement of ~200nm can change the Mn$_3$Sn composition that the 4µm focused spot is measuring.

Position-dependence of the Kerr rotation in Mn$_3$Sn has previously been noted, producing a change by nearly a factor of two in $\Delta \theta_K$ which is likely linked to local composition changes on the surface [152]. Although in this situation the shape of the hysteresis loop did remain constant. An additional factor that could affect collected MOKE data is the sample dimensions. AHE measurements on varying aspect ratios of Mn$_3$Sn single crystals has been shown to alter the enclosed area and symmetry of $\rho_H(B)$ [151]. The two cuts used for the MOKE data presented here, with surface areas 1×2mm$^2$ and 1×3mm$^2$, both have larger dimensions along their respective easy axes (perpendicular to (0001)).

Additional measurements on a (0001) cut surface (in the (2\,\bar{1}\,\bar{1}0),(01\,\bar{1}0) plane as in [152]) were attempted however, the MOKE data showed no signs of hysteresis at any angle. The easiest method to reduce the noise of the obtained MOKE hysteresis plots is to average over many more cycles, which would require a much longer acquisition time. For Mn$_3$Sn, where surface tarnishing occurs relatively quickly, this means it must be kept, and measured, in a protected atmosphere. Balk et al. [152] state that measurements were conducted with Mn$_3$Sn in dry air, so placing the sample in an enclosed dry air space may suffice for this setup too. Alternatively, a higher powered laser source could be used to increase the signal to noise ratio using the same acquisition time.

### 6.6 Conclusion

In this chapter, the preparation of single crystals of the hexagonal antiferromagnet Mn$_3$Sn for MOKE measurements has been outlined. This consisted of aligning large single crystal chunks using a Laue X-ray diffractometer and then cutting to reveal three faces: (01\,\bar{1}0), (2\,\bar{1}\,\bar{1}0), and (0001). A hand polishing method to produce optical quality surfaces of these single crystal cuts was then summarised, and the bulk magnetic properties of the three cut orientations were comparative
with previous studies on Mn$_3$Sn.

Following this, longitudinal MOKE measurements revealed evidence of a large change in ellipticity angle rather than Kerr angle with applied field, contrary to previous publications [90, 152]. Square magnetic hysteresis was observed within 10° of the local easy axis for two orientations of Mn$_3$Sn: for a (01$\bar{1}$0) surface, $\Delta \epsilon_K \approx 0.12$ mrad along (2$\bar{1}$10), with a coercive field of $\pm 125$ mT; for a (2$\bar{1}$10) surface, $\Delta \epsilon_K \approx 0.06$ mrad along (01$\bar{1}$0), with a coercive field of $\pm 115$ mT. The resolution of $\theta_K$ and $\epsilon_K$, as well as the reduced in-plane magnetic field range, limited the quality of obtained MOKE data. These constraints also meant that the MOKE setup was unable to distinguish changes in $\theta_K$ and $\epsilon_K$ along other orientations of Mn$_3$Sn.

Future work requires minor improvements to the experimental setup. To prevent Mn$_3$Sn tarnishing, an enclosed volume around the sample space should be evacuated or filled with an argon atmosphere/dry air. Additionally, the low temperature evolution of both $\theta_K$ and $\epsilon_K$ could be investigated by adapting the MOKE setup to a cryogenic system.
Chapter 7

Conclusions & Outlook

7.1 Summary of Main Findings

This thesis has focused on the development of an experimental MOKE system to fully quantify the polarisation state of light upon reflection from magnetic samples. In comparison to alternative polarimetry techniques such as crossed polarisers, balanced photodiodes, and Sagnac interferometry, the dual PEM setup used in this thesis is unique for being able to synchronously measure the Kerr rotation, $\theta_K$, and the ellipticity angle, $\epsilon_K$.

Chapter 3 outlined the experimental development that formed a large part of the work presented in this thesis. In addition to a description of the optical setup, consisting of a MOKE-measuring light path as well as a separate imaging light path, emphasis was placed on the use of a reflective objective to focus light onto the sample. In comparison to refractive lenses, reflective lenses do not suffer from parasitic Faraday rotations and are typically more reliable for cryogenic applications. As noted in Section (3.1.3) however, reflective lenses can affect the polarisation state of transmitted light due to a change in the effective incidence plane relative to the input polarisation state.

The typical angular resolution of $\theta_K$ and $\epsilon_K$ in the developed setup was measured to be $12\mu$rad, from the standard deviation of 100 separate measurements. This noise level broadly followed a $1/\text{power}$ relation, suggesting that Johnson (thermal) noise within the detection circuit is the limiting factor. For low laser powers, calculated Johnson noise from the pre-amplifier agrees with the measured noise.
For higher powers however, the measured noise appears to follow the same power relation as shot noise from the pre-amplifier, although it is two orders of magnitude higher than is quoted from the amplifier specification. Additionally a small drift of up to $1.2 \mu \text{rad} \text{hour}^{-1}$ was observed, even after this had been reduced by temperature regulation of the entire optical setup.

Errors in measured MOKE signals due to a misalignment or imperfection in the retardation amplitude provided by the dual PEM setup were theoretically analysed in Section (3.4). The severity of each error source depends on the magnitude of $\theta_K$ and $\epsilon_K$. For a typical MOKE signal of $1^\circ \approx 17 \text{mrad}$, a 0.5% error in the PEM retardation amplitude causes an error of $\sim 40 \mu \text{rad}$ in $\theta_K$ and $\sim 20 \mu \text{rad}$ in $\epsilon_K$. From the resolution of the goniometers used, a 1.5mrad misalignment of the second PEM or analyser was calculated to result in an offset of $50 \mu \text{rad}$ in $\theta_K$ and $2 \mu \text{rad}$ in $\epsilon_K$.

Efforts to adapt the MOKE setup to measure materials under cryogenic conditions were outlined in Section (3.5). The constructed brass sample mount with an attached PTFE dampener successfully reduced vibrations within the cryostat to $< 1 \mu \text{m}$. However, a long timescale drift of the position that light was incident on the objective lens caused a drift in MOKE signals and hindered progress. This drift subjected the MOKE measurement to ambient temperature fluctuations and is likely due to thermal expansion of the cryostat-optics support frame. An attempt to reduce the Faraday rotation, $\theta_F$, from the cryostat view-port was also theoretically analysed. This consisted of placing a quarter waveplate either side of the window to convert linearly polarised light into circularly polarised light that is immune to Faraday rotation. Calculations incorporating a slight (but realistic) misalignment or retardance error of either waveplate led to the discovery of large field-dependent variations in the measured $\epsilon_K$, despite successfully reducing $\theta_F$. This additional ellipticity signal negates the advantage of the dual PEM setup and, together with the likely massive field-dependence of the retardance provided by each waveplate, indicates that this idea is ineffective.

Piezoelectric transducers holding the sample allowed a raster scanning procedure to map out magnetic domains. An asymmetric movement from the transducers however, required the development of an image processing routine to accurately place coordinates of measured points on the sample surface. This procedure, as outlined in Section (3.6), tracks the relative position of surface features between measurements using a traditional mean shift algorithm, and was shown to be accurate to within 1%. Additionally, an image processing routine was developed.
to maintain image focus. This autofocus procedure is most useful for temperature-dependent MOKE experiments, where thermal expansion of the sample-objective lens mount can defocus the image.

Chapter 4 presented the longitudinal MOKE response of 5µm thick rectangular permalloy (Ni$_{80}$Fe$_{20}$) mesas grown on a 300nm thick Ni seed layer. The longitudinal $\theta_K$ and $\epsilon_K$ signals varied by over 1mrad across 20×300µm$^2$ mesas. This variation was much larger than the field-dependent $\Delta \theta_K=0.3$mrad and $\Delta \epsilon_K=0.5$mrad for any specific position on the mesas. This discrepancy indicated that, particularly near the edges of the permalloy mesas, the finite size of focused light distorted the MOKE signal. It was still possible to image a persistent domain structure however, by measuring $\theta_K(B)$ and $\epsilon_K(B)$ for multiple field values at each position during a raster scan. This method relies on the same domain structure reappearing each hysteresis cycle, and subsequently was only observed in high aspect ratio mesas, with a large shape-induced uniaxial anisotropy. Further to this, scans over multiple 20×300µm$^2$ mesas separated by 10µm showed seemingly random chirality, indicating that the mesas were non-interacting.

Chapter 5 investigated the magnetic characteristics of magnetostrictive galfenol (Fe$_{1-x}$Ga$_x$) films deposited on a 128° X-cut LiNbO$_3$ substrate. Uniaxial anisotropy in the relative orientation of the reflection plane of the MOKE measurement and the $y$ axis (1¯100) of LiNbO$_3$ indicated the presence of a strain induced by the difference in lattice parameters between galfenol and LiNbO$_3$. This anisotropy was observed in ~250nm thick galfenol films but was not as marked in ~600nm thick films, suggesting that the strain caused by the FeGa-LiNbO$_3$ interface relaxes within galfenol. In 254nm thick Fe$_{84.5}$Ga$_{15.5}$, a decrease of 28% in $\Delta \theta_K$ (between saturation), from 0.78mrad to 0.56mrad, was measured along the easy axis compared to the hard axis. This contrasts with a measured increase of 50% in $\Delta \epsilon_K$, from 0.16mrad to 0.34mrad, in the same film. In 664nm thick Fe$_{86.4}$Ga$_{13.6}$, both $\theta_K(B)$ and $\epsilon_K(B)$ remained similarly shaped, with a increase in $\Delta \theta_K$ of 17% (0.60mrad to 0.72mrad), and a similar increase in $\Delta \epsilon_K$ of 15% (0.44mrad to 0.52mrad). The conversion between $\theta_K(B)$ and $\epsilon_K(B)$ with rotation of the galfenol film could be linked to a structural change due to magnetostriction caused by the LiNbO$_3$-galfenol interface. This asymmetry in crystal structure can lead to a variation in $\theta_K(B)$ and $\epsilon_K(B)$ signals by changing the allowed inter- and intra-band transitions that determine the MOKE signals.

The switching process within galfenol films was also isolated by separating the longitudinal and polar components of MOKE signals. Moments remain within
the galfenol surface plane during the switching process, and no measurable out-of-plane component was observed. Further to this, transverse MOKE measurements at various positions on the 251nm thick Fe\(_{87.5}\)Ga\(_{12.5}\) surface following AC demagnetisation along the hard axis indicated the presence of magnetic domains. These different domains within the thin galfenol film were subsequently imaged using the scanning MOKE setup, appearing as stripes along the hard axis. Finally, a surface acoustic wave device with 250nm thick galfenol was investigated with the MOKE microscope. Standing waves of induced strain in galfenol were set up by exciting the piezoelectric LiNbO\(_3\) substrate. However, no evidence of periodic magnetic structure in galfenol was observed in the MOKE signal for any orientation. Considering the average domain width upon AC demagnetisation was \(\sim 30\ \mu\text{m}\) in the galfenol film, manipulation of the domain positions should have been possible using the SAW device with \(\lambda=200\ \mu\text{m}\), corresponding to a 50\(\mu\text{m}\) distance between nodes and anti-nodes. This comparable distance between domain width and standing wave periodicity should be close to optimum. The domain positions could potentially be controlled by tuning the intrinsic domain width by changing the film thickness or Fe:Ga ratio, or instead by changing the wavelength of SAW/using a higher harmonic of SAW excitation (from 20-40MHz would reduce the node-anti-node distance to 25\(\mu\text{m}\)).

Chapter 6 was dedicated to measuring the longitudinal MOKE response of single crystals of the hexagonal antiferromagnet Mn\(_3\)Sn using p-polarised light (\(\lambda=635\text{nm}\)). The alignment, cutting, and polishing of the single crystals was outlined, resulting in three main surface cuts of Mn\(_3\)Sn: (01\(\bar{1}\)0), (2\(\bar{1}\)\(\bar{1}\)0), and (0001). In terms of the external field dependence of MOKE measurements on Mn\(_3\)Sn, previously only a \(\theta_K(B)\) signal was reported on. Here however, only a \(\epsilon_K(B)\) signal was observed. A longitudinal square-shaped ellipticity signal was measured along the easy axis in (01\(\bar{1}\)0) and (2\(\bar{1}\)\(\bar{1}\)0) surface cuts. For the former cut, \(\Delta\epsilon_K \approx 0.12\text{mrad}\) along (2\(\bar{1}\)\(\bar{1}\)0), with a coercive field of \(\pm 125\text{mT}\); and for the latter, \(\Delta\epsilon_K \approx 0.06\text{mrad}\) along (01\(\bar{1}\)0), with a coercive field of \(\pm 115\text{mT}\). For the above two orientations, the (0001) axis served as the hard axis for which no field dependent MOKE response was measurable, in agreement with previous measurements. A field-dependent Kerr rotation was not discernible along any orientation, although a significantly large spread in \(\theta_K\) prevented resolving a Kerr rotation within 100\(\mu\text{rad}\).

The significance of a larger \(\epsilon_K(B)\) compared to \(\theta_K(B)\) signal lies in the structure of Mn\(_3\)Sn. This is due to how the MOKE angles are dependent on the off-diagonal
optical conductivity, $\sigma_{xy}(\omega)$, which in turn is determined by the allowed inter-band transitions dictated by the electronic structure. Future theoretical work should calculate the complete optical conductivity tensor of Mn$_3$Sn. One of the major limitations of the MOKE experiments here was found to be a long term ($\sim$two weeks) decay of the Mn$_3$Sn surface quality. EDS measurements revealed a tarnishing of the surface, with an oxidation layer forming as well as Sn inclusions up to 0.6$\mu$m in diameter appearing on the surface. As such, future optical measurements on Mn$_3$Sn should be undertaken within a protected atmosphere to prevent surface tarnishing.

7.2 Future Work

Ideas for further work to be done on each of the three materials looked at in this thesis have been included in each of the conclusion chapters respectively. Additionally, further improvements can be made to the experimental setup. Improving the signal-to-noise ratio is a high priority. The easiest change to the current setup is to use a higher power laser source, greater than the current 5mW laser diode used. Seeing as the pre-amplifier was calculated as being the limiting factor in the electronic noise floor, an alternative pre-amplifier with smaller noise equivalent power could be used. Changing the wavelength of the photodiode is an easy alteration to make with the setup. Choosing a visible wavelength that maximises the magneto-optic response of a material would help to increase the signal to noise ratio of MOKE measurements. Additionally, adding a tunable laser source would provide the ability to undertake spectroscopic MOKE measurements which adds another degree of freedom to the measurement that can reveal magneto-optic transitions in the bandstructure.

An alternative objective lens with a larger numerical aperture (>0.5) may increase longitudinal MOKE signals due to the larger incidence angles (>25°). A higher power objective lens would also increase the resolution of magnetic domain images by decreasing the focused spot size. However, the range of reflective objectives with high numerical aperture are currently limited and so a refractive lens may instead be used to increase image resolution.

Additional work is also required to improve the long timescale stability of low temperature measurements using the MOKE setup. Thermal expansion of the frame that supports the optics beneath the available cryostat cause a long-term
drift in the position that light is incident upon the objective lens. This in turn causes a long-term drift in $\theta_K$ and $\epsilon_K$. Actively correcting for this temperature variation, or changing the material of the aluminium supporting frame, could reduce the drift. An optical cryogen, which is designed to minimise mechanical vibrations and drift, could instead be procured. As the MOKE system developed in this thesis is relatively compact (and attached to a single, portable breadboard) it may be transferred to different experimental setups, including an alternative cryogen. Such a system would also provide a vacuum environment for the sample which would reduce tarnishing. This was shown to be vital for a material such as Mn$_3$Sn.
Appendix A

Mueller Matrices

This appendix is lists Mueller matrices that are commonly used in calculations which may be found in most polarisation oriented textbooks e.g. [13, 102, 156]. A concise overview is also provided in [157].

A linear polariser at angle $\gamma$ to the optic axis:

$$\begin{pmatrix}
1 & \cos(2\gamma) & \sin(2\gamma) & 0 \\
\cos(2\gamma) & \cos^2(2\gamma) & \cos(2\gamma)\sin(2\gamma) & 0 \\
\sin(2\gamma) & \cos(2\gamma)\sin(2\gamma) & \sin^2(2\gamma) & 0 \\
0 & 0 & 0 & 0
\end{pmatrix}.$$  (A.1)

A general retarder with fast axis at angle $\theta$ to the optic axis, and with retardance $\phi$:

$$\begin{pmatrix}
1 & 0 & 0 & 0 \\
0 & \cos^2(2\theta) + \cos(\phi)\sin^2(2\theta) & \cos(2\theta)(1 - \cos(\phi))\sin(2\theta) & \sin(2\theta)\sin(\phi) \\
0 & \cos(2\theta)(1 - \cos(\phi))\sin(2\theta) & \cos(\phi)\cos^2(2\theta) + \sin^2(2\theta) & -\cos(2\theta)\sin(\phi) \\
0 & -\sin(2\theta)\sin(\phi) & \cos(2\theta)\sin(\phi) & \cos(\phi)
\end{pmatrix}.  \quad (A.2)
$$

A polarisation modulator (e.g. a Photoelastic Modulator (PEM)) may be modelled by taking the retardance $\phi$ as a time ($t$) variable retardance $\Delta = \Delta_0\sin(\Omega t)$ where $\Delta_0$ is the retardance amplitude and $\Omega$ is the modulation frequency.
A rotator:
\[
\begin{pmatrix}
1 & 0 & 0 & 0 \\
0 & \cos(\beta) & -\sin(\beta) & 0 \\
0 & \sin(\beta) & \cos(\beta) & 0 \\
0 & 0 & 0 & 1
\end{pmatrix}.
\] (A.3)

Such a rotator may either rotate the frame of reference (if rotating an optical device relative to the optic axis), or instead can be used as a Faraday rotator with \(\beta = V_{\text{Verdel}}B\).

A fixed isotropic rotator and reflector:
\[
\begin{pmatrix}
1 & -\cos(2\kappa) & 0 & 0 \\
-\cos(2\kappa) & 1 & 0 & 0 \\
0 & 0 & \cos(\delta)\sin(2\kappa) & \sin(\delta)\sin(2\kappa) \\
0 & 0 & -\sin(\delta)\sin(2\kappa) & \cos(\delta)\sin(2\kappa)
\end{pmatrix},
\] (A.4)

with a phase shift \(\delta\) and rotation \(\kappa\). A perfect mirror is the general case of an isotropic reflector with \(\kappa = \frac{\pi}{4}\) and \(\delta = \pi\):
\[
\begin{pmatrix}
1 & 0 & 0 & 0 \\
0 & 1 & 0 & 0 \\
0 & 0 & -1 & 0 \\
0 & 0 & 0 & -1
\end{pmatrix}.
\] (A.5)
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