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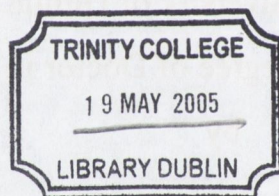
**Structural characterisation and Kerr
magnetometry of Fe/Mo(110) epitaxial
system**

A thesis submitted to the University of Dublin, Trinity College,
in application for the degree of Doctor in Philosophy
by

Victor Usov

Physics Department
Trinity College Dublin
October 2004

Structural characterization and Rietveld
magnetometry of Fe₃(Mo₂(PO₄)₄)₂ spinel
system



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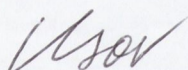
Declaration

This thesis is submitted by the undersigned for the degree of Doctor in Philosophy at the University of Dublin.

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Victor Usov

Acknowledgements

I would first like to thank my supervisor Prof. Igor Shvets, for giving me the opportunity to work in his excellent group. He gave me critical advice during the course of this study, while allowing me some degree of independence. The group has expanded dramatically over his determination and hard work and I wish him best of luck for the future.

I thank Shane Murphy, for his interest in my work, correcting my corrupt English and help with the experiments. Thanks to Dima and Sergey, for sharing a house, helping me with the setup and most of all, being good friends. All the people in the group have made it better by their presence, so thanks to; Nikolai and Roman, for Euro-2004 nights out; Sunil, Jin and Yung, for XRD; Bala, for Indian sweets; Maina, for putting on the angles inside a MOKE system via a 4.5" flange; Guido, for computers; Marie, for administrative support; Gops, for STM tips; Anselm, for cakes and road trips; William, for pizza and wine parties; Luca, for starting the 'mokie' and Kevin, for taking over it; Giuseppe, for discussions on thermal conductivity; Ken, for explaining me the meaning of the words I could never find in any dictionary; Sumesh, Floreano, Rafael and Ciaran, for SFI vs. Fitzgerald match; Alex, for not destroying the lab, Sergio, for his advise on printing this thesis and a summer student David for being a generous sponsor of Russian militia.

Thanks to the help provided by Prof. I.T. McGovern, Prof. J.M.D. Coey, Prof. J. McGilp, Dr. S. Sanvito, Prof. E. Finch, Dr. V. Weldon and the members of their groups, especially Lee Carroll and Dr. M. Venkatesen.

The group could not operate without the efforts of all the departmental staff; T. Burke, J. Kelly, D. Grouse, M. Reilly, N. Carroll, J. McCauley, K. Concannon and all the secretarial staff, especially Susan and Michelle.

Thanks to my friends Misha, Tanya and Daniel for warm welcomes at their place. Thanks to all my friends from Trinity basketball club for all the great games, trainings and fun we

had together and making me to give up a notion that people go to basketball varsities to play basketball.

Thanks to my parents Alexander Viktorovich and Galina Viktorovna and my brother Sergei, for providing me with all possible help and always being there for me, even when it all seemed to go wrong. Finally, I would like to thank my family, my son Sasha for being a good boy and keeping his record of yellow cards at school below fifteen per annum and to my wife Natasha, who gave up her carrier of a medical doctor and came with me to Ireland to share all difficulties and delights of living in a new county. They have all my love.

Abstract

This thesis describes the investigation of the growth and magnetic properties of ultrathin Fe films on the Mo(110) surface. The surface magneto optical Kerr effect (SMOKE) setup was designed and built by author and used in conjunction with the surface characterisation techniques to study magnetic anisotropy and magnetic phase transition in Fe/Mo(110) films. The emphasis is made on correlation between magnetic properties and film morphology.

The surface characterisation was conducted using scanning tunnelling microscopy (STM), low energy electron diffraction (LEED), Auger electron spectroscopy (AES). The SMOKE setup designed to study magnetic properties of ultra-thin films in ultra-high vacuum (UHV) is described in detail. A phenomenological approach used in the description of polarised light, which is based on the Jones vectors and complex plane representation is described. A general overview of the magneto-optical experimental techniques is given and it is shown how the elements of the sample's Jones matrix are related to the magneto-optical parameters measured in the experiment.

The SMOKE system was applied to study the magnetic anisotropy of ultrathin Fe films grown on a vicinal Mo(110) surface. For this purpose Fe films in a thickness range $2ML \leq \theta \leq 7.1ML$ ($3.4 - 12 \text{ \AA}$) were deposited at room temperature on a vicinal Mo(110) substrate. The STM data shows that the films grow in the Stranski-Krastanov mode. The typical staircase pattern of the substrate is maintained with the loss of the straightness of the surface steps with increasing thickness. A uniaxial magnetic anisotropy with the easy magnetisation axis along [001] direction is present in these films down to 2 ML thickness. This is exactly opposite to the seemingly similar Fe/W(110) system and contradicts the results of recent theoretical calculations. The easy axis of two-fold step-induced anisotropy lies along the surface step edges. The influence of the steps on magnetic behavior decreases with increasing film thickness due to the loss of the step's straightness. When the room temperature grown films are annealed in UHV at 750 K, Fe

atoms not contained in the first pseudomorphic layer agglomerate into islands with a characteristic wedge shape which are elongated along the step edges. The magnetic behaviour of these films is governed by the islands' shape anisotropy. Thicker films produce islands with larger vertical aspect ratios leading to larger saturation fields and coercivity. The elongation of the islands at an angle to the [001] direction shifts the easy axis of the effective magnetic anisotropy from [001] crystallographic axis towards the steps due to the shape anisotropy contribution.

The magnetic critical behaviour in Fe films grown on a low-vicinal Mo(110) surface was investigated. The thickness dependence of the Curie temperature was studied in the thickness range $1.9\text{ML} \leq \theta \leq 2.8\text{ML}$. The critical exponent for these films is close to the value $\beta = 0.125$ given by the Ising model. Two Fe films of the same thickness 1.95 ML were grown at 300 K and 510 K. It was found that the growth conditions have a dramatic influence of the on magnetic critical behaviour. A Curie temperature is higher by ~ 30 K and the ferromagnetic-paramagnetic phase transition is broader for the room-temperature grown film compared to the film grown at elevated temperature.

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V. Usov, S. Murphy, I.V. Shvets, *Study of ferromagnetic-paramagnetic phase transition in two-dimensional Fe/Mo(110) epitaxial films*, J. Magn. Magn. Mat. **290-291**, 764 (2005).

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List of abbreviations

2D - two dimensional

3D - three dimensional

AES - Auger electron spectroscopy

bcc - body centered cubic

CMA - cylindrical mirror analyser

DOS - density of states

e-beam - electron beam

FVM - Frank-Van-der-Merwe

LCP –left circular polarisation

LDOS - local density of states

LEED - low energy electron diffraction

MBE - molecular beam epitaxy

ML – monolayer

MOKE - magneto-optical Kerr effect

OD - outer diameter

OFHC - oxygen free high conductivity

RCP - right circular polarisation

RTSTM - room-temperature scanning tunneling microscopy

SK - Stranski-Krastanov

SMOKE – surface magneto-optical Kerr effect

STM - scanning tunneling microscopy

TPD - temperature programmed desorption

TSP - titanium sublimation pump

UHV - ultra high vacuum

XRD - X-ray diffraction

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Introduction

In the past decade research efforts in nanoscience and nanotechnology have grown explosively worldwide. This surge of interest to learn more about the atomic scale structure of matter stems from the remarkable effects that may arise from the size reduction. Interesting unexpected properties (catalytic, magnetic, ferroelectric, mechanical, optical and electronic) appear when the dimensions are reduced to a nanometer scale. Magnetic ultrathin metallic systems have attracted much scientific interest due to their novel magnetic properties such as the reorientation of the preferential direction of magnetisation, oscillatory exchange coupling phenomena and related giant magnetoresistance [1]. Intensive studies of thin magnetic films started in the 1950s when these films began to be considered in terms of their potential for information storage. The fundamental experimental research was inspired by the prospects of discovering new magnetic phenomena expected from theoretical considerations [2]. However the techniques for the ultrathin films growth and their structural, compositional and magnetic analysis were not available at that time. The situation changed with significant progress in vacuum science and molecular beam epitaxy (MBE) ultrathin structures and the concurrent development of magnetic and surface characterisation techniques. Application of these techniques to growth and studies of magnetic materials triggered an extensive experimental research of magnetic phenomena in low-dimensional systems.

The first magneto-optical effect (Faraday effect) was discovered in 1845 by Michael Faraday. He found that the light's polarisation plane rotated after passing through a glass specimen placed in a magnetic field. The similar effect in reflection was discovered thirty two years later by John Kerr when he was examining the light reflected from a polished electromagnet pole. The phenomenon was called magneto-optical Kerr effect (MOKE). It describes the changes in the light's state of polarisation when it is reflected from a magnetised medium. When the incident light is linear polarised, the changes are referred to as Kerr rotation and Kerr ellipticity. Depending on the magnetisation orientation with respect to light's scattering plane there are three distinct MOKE configurations which are

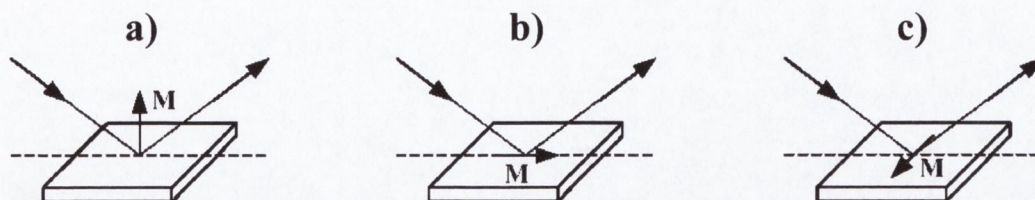


Figure 1 Three configurations used in magneto-optic Kerr effect experiments: a) polar, b) longitudinal c) transverse.

shown in figure 1. In the polar Kerr effect, the direction of magnetisation is perpendicular to the film plane. In the longitudinal geometry, the magnetisation is in the film plane and in the plane of incidence of the probing light. In the transverse Kerr effect, the magnetisation is also in the film plane but it is perpendicular to the plane of incidence. Using *p*- or *s*- linear polarised incident light (where the electric field vector oscillates in the plane of incidence or perpendicular to it) simplifies the algebra describing the effect and makes the interpretation of the experimental data easier.

Today MOKE has attracted much interest due to its application as a read out technique in perpendicular high-density magneto-optical recording [3, 4]. Without any mechanical contact the detection system is capable of resolving between two orientations of magnetisation corresponding to two logical states. Moog and Bader were the first to place a MOKE system into ultra-high vacuum (UHV) to study ultrathin Fe/Au(100) films [5, 6]. Since then the local character and submonolayer sensitivity have made MOKE an effective tool for probing magnetic properties of ultrathin magnetic structures. The acronym SMOKE is used to denote *surface* magneto-optic Kerr effect when this technique is applied to explore the magnetism of ultra thin films with a thickness of several atomic layers. A significant advantage of the SMOKE technique is the relative ease with which it can be incorporated into UHV. For magnetic measurements under ambient conditions a passivating layer of a non-reactive material such as Au is typically required. In the case of ultrathin film measurements, this layer can affect the magnetic properties of the system being measured [7]. *In-situ* SMOKE measurements performed in UHV have the advantage that a capping layer is not required.

Among the diverse bimetallic systems, Fe films epitaxially grown on the refractory metal substrates such as Mo or W have received particular attention regarding the effects of film-substrate d-band hybridisation and misfit strain on magnetic properties of Fe films [8, 9]. In a practical sense, there is the additional advantage that the tungsten and molybdenum are relatively easy to clean for surface studies, while surface alloying can be neglected in both systems [10]. In this study, the magnetic properties of ultrathin Fe films grown epitaxially on the Mo(110) surface are investigated using the SMOKE technique. This UHV SMOKE setup was designed and developed by the author as an annex to a larger UHV system, which includes other surface-sensitive techniques such as scanning tunneling microscopy STM [11], low energy electron diffraction LEED [12] and Auger electron spectroscopy AES [13]. The morphology of Fe on Mo(110) surface has been extensively studied in the last decade though there is no experimental data available on the magnetism of this system. The previous theoretical work indicates that Fe(110)/Mo(110) exhibits magnetic properties similar to that of the more widely studied Fe/W(110) system. This thesis presents the first detailed study of magnetism of ultrathin Fe films grown on Mo(110) surface.

Chapter 1

Polarised light

In this chapter the description of light polarisation is summarised. A magneto-optical media is considered as a device changing the light polarisation so the transformations of a polarisation state by polarising optical elements are also described. Although polarisation optics is well known and well described in references [14-19], a brief overview of this topic is given in order to define the sign convention used in the further sections.

1.1 Elliptical polarisation

A light wave is considered as a time-harmonic electromagnetic wave. The electric field vector $\mathbf{E}(\mathbf{r}, t)$ is chosen to define the state of polarisation. The electric field of a monochromatic plane wave, propagating in the z -direction can be represented as a function:

$$\mathbf{E}(z, t) = \text{Re}(\mathbf{E}_0 \exp(i(\omega t - kz + \delta)))$$

where E_0 is a complex amplitude of the electric field, ω is a circular frequency, k is a wave number and Re stands for the real part. In general, totally polarised light is elliptically polarised, which means that the electric field vector of the plane wave, propagating in the z -direction, traces an ellipse in the xy -plane, perpendicular to the wave vector at a given point. The polarisation ellipse is described by its *azimuth (rotation)* θ , the angle the major axis of the ellipse makes with the x -axis of the chosen coordinate system, and the *ellipticity*, $\tan(\varepsilon)$, which is the ratio of length the semi-minor axis to the length of the semi-major axis, where ε is the ellipticity angle, defined as in figure 1.1. Any state of elliptical polarisation is either ‘right-handed’ or ‘left-handed’. In case of the right-handed state the tip of the electric field vector

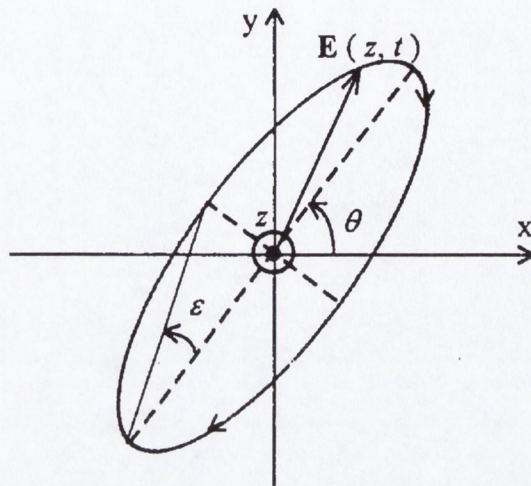


Figure 1.1 Right-handed elliptically polarised light. The tip of the electric field vector travels clockwise as seen by an observer looking from the positive z -direction towards the light source. The state of polarisation is described by rotation θ and ellipticity $\tan \epsilon$.

travels around its elliptical locus in a clockwise direction, as seen by an observer looking from the positive z -direction towards the light source and the counter-clockwise rotation corresponds to the left-hand state of polarisation. The positive and negative sign of ellipticity correspond to right and left-handed polarisations respectively.

1.2 Jones vector formalism

There are several formalisms employed for describing polarised light and its transformations by polarisation optical elements. The classical method of dealing with a totally polarised light is using the Poincare sphere. For analysis of partially polarised light one can employ four-component Stokes vectors and corresponding Mueller matrices.

The focus of this chapter is on the approach, based on Jones vectors and the complex plane representation. While this method can only be used to describe the fully polarised light, it is advantageous in understanding magneto-optical phenomena and explaining experimental techniques.

In the Jones calculations, the polarisation of light is characterised by a Jones vector $\mathbf{J}$ in two-dimensional complex vector space. The coordinates of the Jones vector are specified with respect to a specific basis. In general, the basis can be constructed of any two linearly independent vectors corresponding to different polarisations, but it is usually preferred to choose the basis spanned by two orthogonal polarisation states, satisfying the condition [14]

$$\mathbf{J}_1^\dagger \mathbf{J}_2 = \mathbf{J}_2^\dagger \mathbf{J}_1 = 0 \quad (1.1)$$

where the “†” superscript indicates a Hermitian adjoint. Any polarisation can be expressed as a linear superposition of any two orthogonal components “1” and “2” having the amplitudes a_1 and a_2 and their absolute phases are shifted by $\delta = \delta_1 - \delta_2$. Taking these two components as a basis and after introducing $\tan \alpha = a_1 / a_2$, one can write the coordinates of the normalised Jones vector with respect to the given basis as

$$\mathbf{J}_{12} = \begin{pmatrix} \cos(\alpha) \\ \sin(\alpha) e^{i\delta} \end{pmatrix} \quad (1.2)$$

where the subscripts 1 and 2 indicate the basis. Two of the most important bases are:

the Cartesian basis, made by two linear polarisations along the x and y directions:

$$\begin{pmatrix} 1 \\ 0 \end{pmatrix}, \quad \begin{pmatrix} 0 \\ 1 \end{pmatrix} \quad (1.3)$$

and *the circular basis* made by right and left circular polarisations (RCP and LCP respectively):

$$\frac{1}{\sqrt{2}} \begin{pmatrix} 1 \\ i \end{pmatrix}, \quad \frac{1}{\sqrt{2}} \begin{pmatrix} 1 \\ -i \end{pmatrix} \quad (1.4)$$

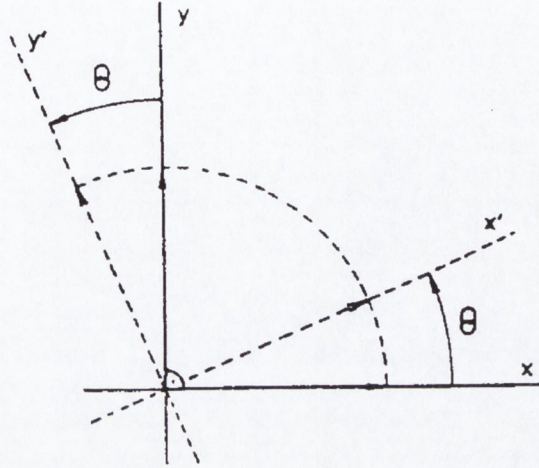


Figure 1.2 Rotation of a Cartesian coordinate system. xy is the original coordinate system; $x'y'$ is obtained by rotating xy about the origin by angle θ .

The switching between the Cartesian $\mathbf{J}_{xy} = \begin{pmatrix} E_x \\ E_y \end{pmatrix}$ and circular $\mathbf{J}_{LR} = \begin{pmatrix} E_L \\ E_R \end{pmatrix}$ Jones vectors is equivalent to a linear transform, which can be expressed in the matrix form:

$$\mathbf{J}_{xy} = \frac{1}{\sqrt{2}} \begin{pmatrix} 1 & 1 \\ -i & i \end{pmatrix} \mathbf{J}_{LR} = \mathbf{F} \mathbf{J}_{LR} \quad (1.5)$$

$$\mathbf{J}_{LR} = \frac{1}{\sqrt{2}} \begin{pmatrix} 1 & i \\ 1 & -i \end{pmatrix} \mathbf{J}_{xy} = \mathbf{F}^{-1} \mathbf{J}_{xy} \quad (1.6)$$

where $\mathbf{F}$ is called the transition matrix. The principal Cartesian and circular Jones vectors are summarised in Table 1.

Another important case is a transformation of the Jones vectors under rotation of a Cartesian coordinate system (Fig. 1.2). The corresponding transition matrix (or rotation matrix) represents the orthogonal transformation, which rotates all vectors in a clockwise direction:

$$\mathbf{R}(\theta) = \begin{pmatrix} \cos \theta & \sin \theta \\ -\sin \theta & \cos \theta \end{pmatrix} \quad (1.7)$$

Its inverse is easily obtained, because $\mathbf{R}^{-1}(\theta) = \mathbf{R}(-\theta)$.

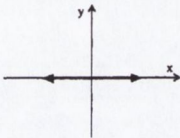
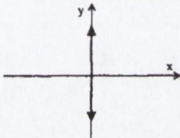
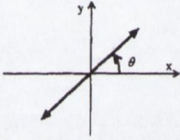
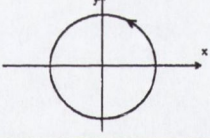
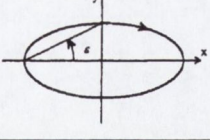
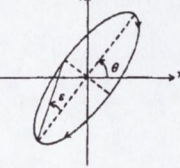
<u>Light polarization</u>	<u>Cartesian</u> $\mathbf{J}_{xy} = \begin{pmatrix} E_x \\ E_y \end{pmatrix}$		<u>Circular</u> $\mathbf{J}_{LR} = \begin{pmatrix} E_L \\ E_R \end{pmatrix}$
Linear along x direction	$\begin{pmatrix} 1 \\ 0 \end{pmatrix}$		$\frac{1}{\sqrt{2}} \begin{pmatrix} 1 \\ 1 \end{pmatrix}$
Linear along y direction	$\begin{pmatrix} 0 \\ 1 \end{pmatrix}$		$\frac{1}{\sqrt{2}} \begin{pmatrix} 1 \\ -1 \end{pmatrix}$
Linear with azimuth θ	$\begin{pmatrix} \cos \theta \\ \sin \theta \end{pmatrix}$		$\frac{1}{\sqrt{2}} \begin{pmatrix} \cos \theta + i \sin \theta \\ \cos \theta - i \sin \theta \end{pmatrix}$
Circular right-handed	$\frac{1}{\sqrt{2}} \begin{pmatrix} 1 \\ i \end{pmatrix}$		$\begin{pmatrix} 0 \\ 1 \end{pmatrix}$
Circular left-handed	$\frac{1}{\sqrt{2}} \begin{pmatrix} 1 \\ -i \end{pmatrix}$		$\begin{pmatrix} 1 \\ 0 \end{pmatrix}$
Elliptical azimuth $\theta = 0$	$\begin{pmatrix} \cos \varepsilon \\ i \sin \varepsilon \end{pmatrix}$		$\frac{1}{\sqrt{2}} \begin{pmatrix} \cos \varepsilon - \sin \varepsilon \\ \cos \varepsilon + \sin \varepsilon \end{pmatrix}$
Elliptical with azimuth θ , ellipticity ε	$\begin{pmatrix} \cos \theta \cos \varepsilon - i \sin \theta \sin \varepsilon \\ \sin \theta \cos \varepsilon + i \cos \theta \sin \varepsilon \end{pmatrix}$		$\frac{1}{\sqrt{2}} \begin{pmatrix} (\cos \varepsilon - \sin \varepsilon) e^{i\theta} \\ (\cos \varepsilon + \sin \varepsilon) e^{-i\theta} \end{pmatrix}$

Table 1.1 Jones vectors of various polarisations.

The Cartesian representation is widely employed because it provides a clear geometric interpretation of the polarisation state. On the other hand the circular representation is useful for the systems where the circular polarisations can propagate independently.

In the case of a Cartesian basis, the following procedure can be applied to obtain the general elliptical polarisation (with specific θ and ε). For an ellipse with the major axis parallel with x ($\theta = 0$) and the angle ε , the azimuth can be introduced by a counter-clockwise rotation of the ellipse (*i.e.* by the clockwise rotation of the coordinate system) using the rotation matrix $\mathbf{R}(-\theta)$ (1.7)

$$\mathbf{J}_{xy} = \begin{pmatrix} \cos \theta & -\sin \theta \\ \sin \theta & \cos \theta \end{pmatrix} \begin{pmatrix} \cos \varepsilon \\ i \sin \varepsilon \end{pmatrix} = \begin{pmatrix} \cos \theta \cos \varepsilon - i \sin \theta \sin \varepsilon \\ \sin \theta \cos \varepsilon + i \cos \theta \sin \varepsilon \end{pmatrix} \quad (1.8)$$

It is also possible to express the general elliptical polarisation directly using the definition of the normalised Jones vector (1.2) with respect to the Cartesian basis, when we specify the quantities α and δ . These two expressions of the Jones vector are equivalent: in the expression (1.8) the polarisation ellipse is determined by the geometrical parameters θ and ε specifying the shape of the ellipse, while in the second case the ellipse is described using the parameters α and δ , which explain how the orthogonal polarisations of the basis "mix" together. The two sets of parameters are related by the following equations [19]:

$$\tan 2\theta = \tan 2\alpha \cos \delta \quad (1.9)$$

$$\tan 2\varepsilon = \sin 2\alpha \sin \delta \quad (1.10)$$

The Jones vector in the circular basis can be obtained from the Eq. (1.8) using the Eq. (1.6) which describes the transformation from the Cartesian basis to the basis spanned by RCP and LCP polarisations:

$$\mathbf{J}_{LR} = \frac{1}{\sqrt{2}} \begin{pmatrix} (\cos \varepsilon - \sin \varepsilon) \exp(i\theta) \\ (\cos \varepsilon + \sin \varepsilon) \exp(-i\theta) \end{pmatrix} = \begin{pmatrix} \cos(\pi/4 + \varepsilon) \exp(i\theta) \\ \sin(\pi/4 + \varepsilon) \exp(-i\theta) \end{pmatrix} \quad (1.11)$$

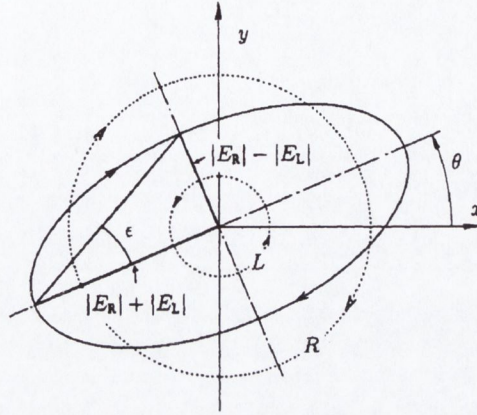


Figure 1.3 Arbitrary polarisation with azimuth θ and ellipticity ε can be expressed as a sum of left and right circular polarised waves. Azimuth θ is determined by the phase difference of these waves, while the ellipticity $\tan \varepsilon$ is attributed to the difference in their amplitudes.

In this case one can also use the definition (1.2) and express the Jones vector as a linear combination of the two vectors corresponding to RCP and LCP waves:

$$\mathbf{J}_{LR} = \begin{pmatrix} E_L \\ E_R \end{pmatrix} = \begin{pmatrix} \cos \alpha \\ \sin \alpha e^{i\delta} \end{pmatrix} \quad (1.12)$$

Comparing Eqs. (1.11) and (1.12) we obtain

$$\tan \alpha = \frac{a_L}{a_R} = \tan \left(\frac{\pi}{4} + \varepsilon \right) = \frac{1 + \tan \varepsilon}{1 - \tan \varepsilon} \quad (1.13)$$

$$\delta = \delta_R - \delta_L = -2\theta$$

The result, given by the Eqs. 1.13 is very important, because it shows that if any elliptically polarised wave (having the azimuth θ and the ellipticity ε) is expressed as a sum of the LCP and the RCP waves, then the azimuth θ is determined by the phase difference $\delta = \delta_R - \delta_L$ of these RCP and LCP waves, while the ellipticity $\tan \varepsilon$ is attributed to the difference in their amplitudes (Fig. 1.3):

$$\theta = -\frac{\delta}{2} = -\frac{1}{2}(\delta_R - \delta_L) \quad (1.14)$$

$$\tan \varepsilon = \frac{\tan \alpha - 1}{\tan \alpha + 1} \quad (1.15)$$

It should be pointed out that this is an exceptional case, because normally θ and ε are related to both parameters α and δ , according to Eqs. (1.9) and (1.10).

A Jones vector, in general, has two complex components and contains information not only on the polarisation state itself, but also on the amplitude and the absolute phase. The Jones vector can be normalised if the absolute value of the amplitude is not required. After that only three parameters remain and one of them is the absolute phase. The Jones vector in this form is still more general than that defined by Eq. (1.2) and can be used for studies of the interference phenomena. If the total phase is not needed, one can obtain the Jones vector in the form given by the Eq. (1.2) by multiplying the original Jones vector by a complex number and/or dividing it by its first component. This procedure produces the Jones vector with the unitary first component. The second component is in general a complex number containing the total information about the polarisation state. This number is sometimes called the *complex polarization parameter* χ , which is defined as a ratio of the second to the first component of the Jones vector (1.2)

$$\chi_{12} = \tan \alpha \cdot \exp(i\delta) \quad (1.16)$$

so that the absolute value and the argument of χ_{12} are given by:

$$|\chi_{12}| = \tan \alpha = \frac{a_2}{a_1} \quad (1.17)$$

$$\arg(\chi_{12}) = \delta_1 - \delta_2 = \delta \quad (1.18)$$

In this approach each polarisation state is represented by a point on the complex plane so that the method is called the complex plane representation. Since the Jones vector is defined with respect to a certain basis, it is evident that the complex polarisation parameter is related to the same basis.

The complex parameter χ_{xy} of an arbitrary elliptical polarisation with respect to the Cartesian basis can be obtained from (1.8)

$$\chi_{xy} = \frac{E_y}{E_x} = \frac{\sin \theta \cos \varepsilon + i \cos \theta \sin \varepsilon}{\cos \theta \cos \varepsilon - i \sin \theta \sin \varepsilon} = \frac{\tan \theta + i \tan \varepsilon}{1 - i \tan \theta \tan \varepsilon} \quad (1.19)$$

When the angles θ and ε are small, it is possible to replace the tangents by the first terms of Taylor expansions: $\tan \theta \approx \theta$, $\tan \varepsilon \approx \varepsilon$. Neglecting the second order term in the denominator of (1.19) gives:

$$\chi_{xy} \approx \theta + i\varepsilon \quad (1.20)$$

For the circular representation the parameter χ_{12} be calculated from Eq. (1.11)

$$\chi_{LR} = \frac{E_R}{E_L} = \tan\left(\frac{\pi}{4} + \varepsilon\right)e^{-2i\theta} \quad (1.21)$$

If the circular complex parameter is known, the azimuth θ and the ellipticity ε quantities can be calculated from Eqs. (1.14) and (1.15), using (1.17) and (1.18):

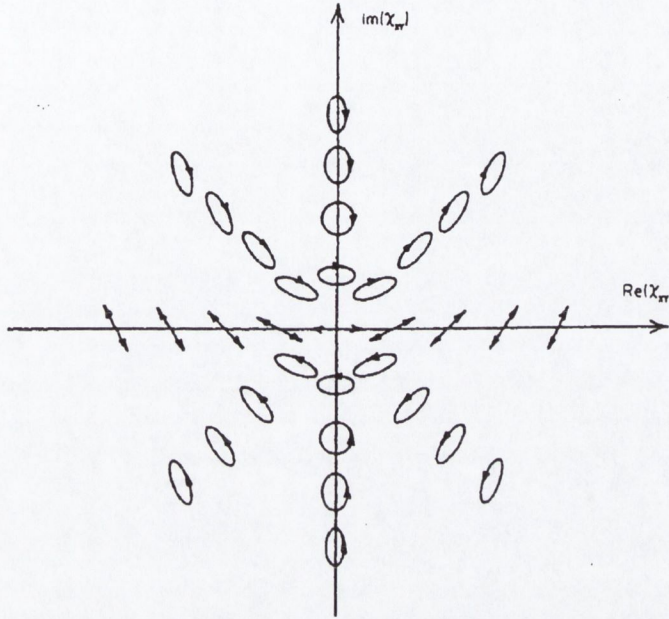


Figure 1.4 Complex plane representation of polarisation states in a Cartesian basis. Any state of polarisation is represented by a single complex number χ_{xy} defined by Eq. (1.16). Each χ_{xy} is associated with a representative point on the complex plane.

$$\tan \varepsilon = \frac{|\chi_{LR}| - 1}{|\chi_{LR}| + 1} \quad (1.22)$$

$$\theta = -\frac{1}{2} \arg(\chi_{LR}) \quad (1.23)$$

The assignment of the polarisation states to the different points of the complex plane in the Cartesian representation is shown in Fig. 1.4.

The distributions of the loci representing the different polarisation states in the complex plane can be obtained by the stereographic projection from a spherical surface [14]. The representation is determined by the orientation of the sphere, when the projection is performed. This representation of the polarization states was first introduced by H. Poincare in 1892. If the diameter of the Poincare sphere equals S_0 , then any elliptical polarisation corresponds to one locus on the sphere, having the Cartesian coordinates:

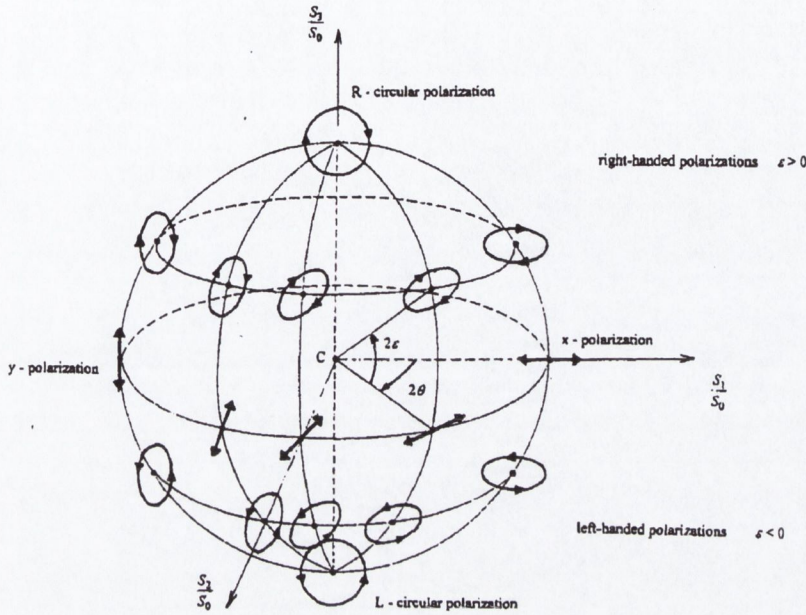


Figure.1.5 Poincare sphere. The centre C represents unpolarised light. The surface of the sphere represents the fully polarised light. The angles θ and ε are the azimuth and the ellipticity of the polarised light respectively.

$$S_1 = S_0 \cos 2\varepsilon \cos 2\theta \quad (1.24)$$

$$S_2 = S_0 \cos 2\varepsilon \sin 2\theta \quad (1.25)$$

$$S_3 = S_0 \sin 2\varepsilon \quad (1.26)$$

From these equations it is evident, that the longitude of the given point is equal to double of the azimuth, while the latitude of this point equals to double of the ellipticity. The set of numbers (S_0, S_1, S_2, S_3) is the Stokes vector $\mathbf{S}$, which defines the state of polarisation. Eqs. (1.24-1.26) define the Stokes vector of fully polarised light. From the Stokes parameters we can easily obtain the information on the azimuth and the ellipticity angle:

$$\theta = \frac{1}{2} \arctan \left(\frac{S_2}{S_1} \right) \quad (1.27)$$

$$\varepsilon = \frac{1}{2} \arcsin \left(\frac{S_3}{\sqrt{S_1^2 + S_2^2 + S_3^2}} \right) \quad (1.28)$$

The advantage of the Stokes formalism is that it is suitable for description of not only fully- but also partially- and un-polarised light. The *degree of polarization* P , which is the ratio of intensity of the fully polarised component to the total intensity, can be found from:

$$P = \frac{\sqrt{S_1^2 + S_2^2 + S_3^2}}{S_0} \quad (1.29)$$

The transformation of the Stokes vectors by an optical system is defined by the 4x4 Mueller matrix $\mathbf{M}$:

$$\mathbf{S}_0 = \mathbf{M} \mathbf{S}_i \quad (1.30)$$

where $\mathbf{S}_0$ and $\mathbf{S}_i$ are the output and input Stokes vector respectively. The approach using the Poincare sphere is quite elegant (Fig 1.5), because it provides a clear geometrical idea and the components of the Stokes vector are real numbers. However, the Jones formalism and the complex plane representation are used in the following mostly because of the authors' preferences.

1.3 Jones matrices

The *Jones matrixes* describe the transformation of the state of polarisation by an optical element. If the Jones vectors $\mathbf{J}_i$ and $\mathbf{J}_o$ with the components E_{ix} , E_{iy} and E_{ox} , E_{oy} describe the wave at the input and the output of the optical system (Fig 1.6), then the relation between them is given by

$$\begin{pmatrix} E_{ox} \\ E_{oy} \end{pmatrix} = \begin{pmatrix} T_{11} & T_{12} \\ T_{21} & T_{22} \end{pmatrix} \begin{pmatrix} E_{ix} \\ E_{iy} \end{pmatrix} \quad \text{or} \quad \mathbf{J}_o = \mathbf{T} \mathbf{J}_i; \quad (1.31)$$

where the 2x2 matrix $\mathbf{T}$ is the Jones matrix of the polarising optical element.

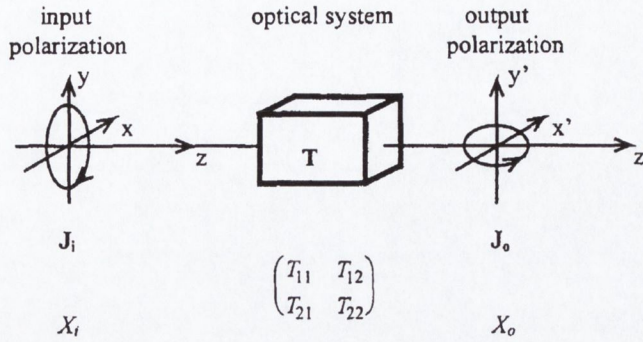


Figure 1.6 Transformation of polarisation state by optical elements. If the Jones vectors J_i and J_o describe the wave at the input and the output of a polarising element, then the relation between them is given by $J_o = T J_i$, where T is the Jones matrix of the polarising element.

If the polarisation of the incident light J_i , is transformed by a sequence of several polarising elements with Jones matrices $T_1, T_2, \dots, T_n$, then the Jones matrix of this set of components is a product of the Jones matrices of the individual optical elements, multiplied in the reversed order and the output polarisation is characterised by the Jones vector, given by

$$J_o = T_n \dots T_2 T_1 J_i = T J_i \quad (1.32)$$

where $T = T_n \dots T_2 T_1$ is the Jones matrix of the given set of optical components. Under the rotation of the coordinate system the Cartesian Jones matrix of an optical system is transformed as

$$T_{x'y'} = R(\theta) T_{xy} R(-\theta) \quad (1.33)$$

where $R(\theta)$ is the rotation matrix, defined by equation (1.7). The transition of the Jones matrix from the Cartesian to the circular basis is given by the formula

$$T_{LR} = F^{-1} T_{xy} F; \quad (1.34)$$

where F is the transition matrix, defined by (1.5). The examples of the Cartesian and circular Jones matrices, which are frequently employed in calculations of the magneto-optical experimental configurations, are presented in Table 2.

Polarization component	Description	Cartesian Jones matrix T_{xy}	circular Jones matrix T_{LR}
Linear polarizer x direction	output light is linearly polarized at x direction	$\begin{pmatrix} 1 & 0 \\ 0 & 0 \end{pmatrix}$	$\frac{1}{2} \begin{pmatrix} 1 & 1 \\ 1 & 1 \end{pmatrix}$
Linear polarizer α rotated from x axis	output light is linearly polarized at angle α from x direction	$\begin{pmatrix} \cos^2 \alpha & \sin \alpha \cos \alpha \\ \sin \alpha \cos \alpha & \sin^2 \alpha \end{pmatrix}$	$\frac{1}{2} \begin{pmatrix} 1 & e^{i2\alpha} \\ e^{-i2\alpha} & 1 \end{pmatrix}$
Wave retarder optical axis parallel with x	$\delta = 2\pi d(n_o - n_e) / \lambda$ δ - retardation angle $(n_o - n_e)$ - birefringence of the medium d - thickness of the plate	$\begin{pmatrix} 1 & 0 \\ 0 & e^{-i\delta} \end{pmatrix}$	$e^{-i\frac{\delta}{2}} \begin{pmatrix} \cos \delta / 2 & i \sin \delta / 2 \\ i \sin \delta / 2 & \cos \delta / 2 \end{pmatrix}$
Half-wave plate $\delta = \pi$	- reverses the direction of the polarization with respect to optical axis - changes the sense (handedness) of the polarization	$\begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}$	$\begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix}$
Quarter-wave plate $\delta = \pi/2$	transforms the linear polarization at 45° from its optical axis to circular one and vice versa	$\begin{pmatrix} 1 & 0 \\ 0 & -i \end{pmatrix}$	$\frac{1}{2} \begin{pmatrix} 1-i & 1+i \\ 1+i & 1-i \end{pmatrix}$
Rotator	output light is rotated at the angle α $\alpha = \pi d(n_R - n_L) / \lambda$ $(n_R - n_L)$ - optical activity, circular birefringence	$\begin{pmatrix} \cos \alpha & -\sin \alpha \\ \sin \alpha & \cos \alpha \end{pmatrix}$	$\begin{pmatrix} e^{i\alpha} & 0 \\ 0 & e^{-i\alpha} \end{pmatrix}$

Table 1.2 Jones matrices of basic optical components.

In the complex plane representation the polarising optical elements are described by a polarisation transfer function, which is determined by the elements of the Jones matrix $\mathbf{T}$ from Equation (1.32). Using the definition of the complex polarisation parameter χ we obtain:

$$\chi_o = \frac{T_{22}\chi_i + T_{21}}{T_{12}\chi_i + T_{11}} \quad (1.35)$$

where χ_i and χ_o are complex polarisation parameters of the input and output polarisations respectively.

The states of polarisation, which are not changed when the light is transmitted through an optical system, are called the eigenmode polarisations of this system. The eigenpolarisations are described by the Jones vector $\mathbf{J}_e$ and defined by an eigenmode equation:

$$\mathbf{T} \mathbf{J}_e = V_e \mathbf{J}_e \quad (1.36)$$

where V_e is a complex number called the eigenvalue.

1.4 Reflection and transmission matrices. Magneto-optical observables

The principal process in magneto-optics is the transformation of the incident light polarisation by a magnetic system. The influence of the magnetised media on the polarisation of the reflected or transmitted light can be described within the Jones formalism using the Cartesian representation. Ellipsometry defines the principal *s*- and *p*-linear polarisations for an isotropic media along the *x*- and *y*-axes respectively (Fig. 1.7). *s*-polarisation (transverse electric TE) is perpendicular to the plane of incidence and *p*-polarisation (transverse magnetic TM) lies in the plane of incidence. In this case the effect of the sample is expressed by:

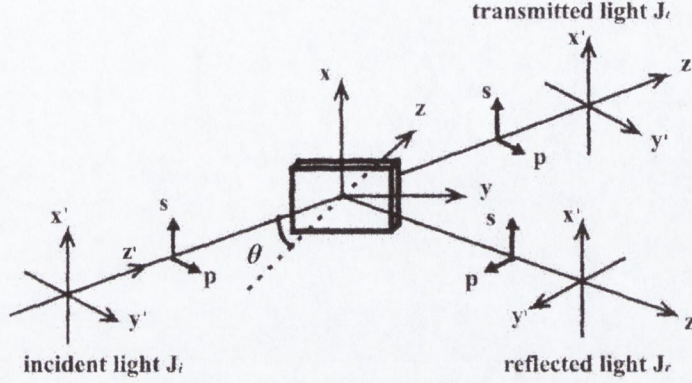


Figure 1.7 Definition of coordinate systems for transmission and reflection. x', y', z' is a local coordinate system with the z' -axis along the wave normal. For p - and s - linear polarisations the electric field vector oscillates in the plane of incidence or perpendicular to it respectively. x' - and y' -axes are chosen along s - and p - principal polarisations respectively

$$\mathbf{J}_r = \begin{pmatrix} r_{ss} & r_{sp} \\ r_{ps} & r_{pp} \end{pmatrix} \mathbf{J}_i = \mathbf{R} \mathbf{J}_i \quad (1.37)$$

$$\mathbf{J}_t = \begin{pmatrix} t_{ss} & t_{sp} \\ t_{ps} & t_{pp} \end{pmatrix} \mathbf{J}_i = \mathbf{T} \mathbf{J}_i \quad (1.38)$$

where $\mathbf{R}$ is the *reflection matrix*, $\mathbf{T}$ is the *transmission matrix*. $\mathbf{J}_i$, $\mathbf{J}_r$ and $\mathbf{J}_t$ are the Jones vectors of the incident, reflected and transmitted light respectively. The matrix elements r_{ij} and t_{ij} $i, j = s, p$ are the *amplitude reflection and transmission coefficients*, also called the *magneto-optical coefficients* when applied to describe magneto-optical phenomena [20]. They are defined as the ratio of the i component of the reflected $E_i^{(r)}$ (or transmitted $E_i^{(t)}$) light to the j component of the incident light $E_j^{(i)}$.

$$r_{ij} = \frac{E_i^{(r)}}{E_j^{(i)}}; \quad t_{ij} = \frac{E_i^{(t)}}{E_j^{(i)}}; \quad (1.39)$$

The Jones matrixes $\mathbf{R}$ and $\mathbf{T}$ provide the full description of the magneto-optical properties of the sample. In the case of a linear dielectric the matrixes $\mathbf{R}$ and $\mathbf{T}$ are diagonal, which indicates the absence of interaction between the s and p waves. The diagonal elements are related to the sample structure and to the optical properties of

individual layers. In-general, when the sample is magnetised, the off-diagonal elements become non-zero.

When the incident light is linearly polarised the Magneto-Optical Kerr Effect is characterised by complex reflection coefficients or by the rotation and ellipticity of the reflected light, measured in the experiment. The consideration of only s or p -polarised light simplifies the algebra and the analysis of the experimental results. For the case of the s -polarised incident beam the reflected light is described by the Jones vector:

$$\begin{pmatrix} r_{ss} & r_{sp} \\ r_{ps} & r_{pp} \end{pmatrix} \begin{pmatrix} E_s^{(i)} \\ E_p^{(i)} \end{pmatrix} = \begin{pmatrix} r_{ss} & r_{sp} \\ r_{ps} & r_{pp} \end{pmatrix} \begin{pmatrix} 1 \\ 0 \end{pmatrix} = \begin{pmatrix} r_{ss} \\ r_{ps} \end{pmatrix} = r_{ss} \begin{pmatrix} 1 \\ r_{ps}/r_{ss} \end{pmatrix} = r_{ss} \begin{pmatrix} 1 \\ \chi \end{pmatrix} \quad (1.40)$$

The elliptically polarised reflected light is described by the complex number χ , given by (1.19). The Kerr effect is a small perturbation [21, 22] and consequently the approximation (1.20) can be used.

$$\chi \approx \phi_s = \theta_s + i\varepsilon_s = \frac{r_{ps}}{r_{ss}} \quad (1.41)$$

In this case χ is denoted as ϕ and called the *complex Kerr rotation*. θ_s and ε_s are the *Kerr rotation* and *Kerr ellipticity* respectively. The index s is attributed to the state of polarisation of the incident light.

In case of the p -polarised incident beam the reflected light is described by the Jones vector given by:

$$\begin{pmatrix} r_{ss} & r_{sp} \\ r_{ps} & r_{pp} \end{pmatrix} \begin{pmatrix} E_s^{(i)} \\ E_p^{(i)} \end{pmatrix} = \begin{pmatrix} r_{ss} & r_{sp} \\ r_{ps} & r_{pp} \end{pmatrix} \begin{pmatrix} 0 \\ 1 \end{pmatrix} = \begin{pmatrix} r_{sp} \\ r_{pp} \end{pmatrix} = r_{pp} \begin{pmatrix} r_{sp}/r_{pp} \\ 1 \end{pmatrix} = r_{pp} \begin{pmatrix} \chi' \\ 1 \end{pmatrix} \quad (1.42)$$

In the same manner the inversely defined complex number χ' is related to the rotation and ellipticity as:

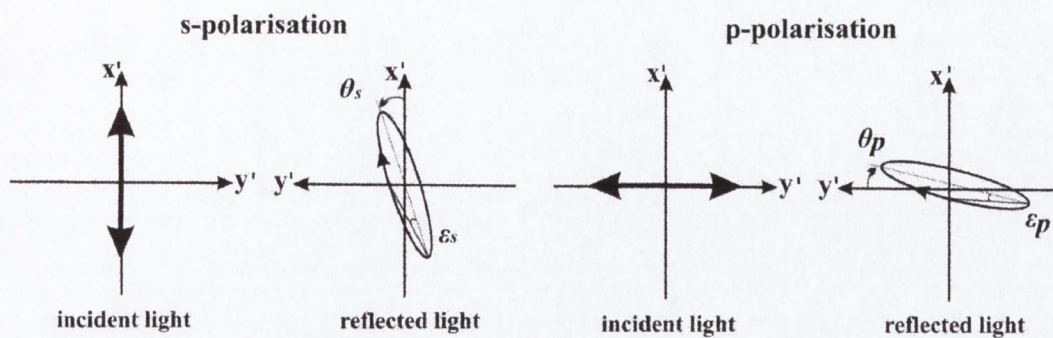


Figure 1.8 Kerr rotation and ellipticity defined for s - and p - polarised incident light.

$$\chi' = \frac{E_x}{E_y} = \frac{-\tan \theta_p + i \tan \epsilon_p}{1 + i \tan \theta_p \tan \epsilon_p} \approx \phi_p = -\theta_p + i \epsilon_p = \frac{r_{sp}}{r_{pp}} \quad (1.43)$$

The sign convention used to define the magneto-optical parameters is the same to that used by G. Metzger *at al.* in [23]. It becomes clear from Fig. 1.8.

Chapter 2

Macroscopic formalism of magneto-optic Kerr effect

2.1 Permittivity tensor

Magneto-optics is presently described in the context of either microscopic or macroscopic theories. Microscopically, the coupling between the electric field of light and the electron spin within the magnetised medium occurs through the spin-orbit interaction. The spin-orbit interaction provides the relation between the motion of electrons and their magnetic moments and is consequently responsible for magneto-optic properties. Kittel showed that it is the change of the electron wavefunctions due to the spin-orbit interaction that gives rise to a correct order of magnitude of the magneto-optic effects. [24]. Argyres later gave a full derivation of the magneto-optic effect in a ferromagnet using perturbation theory [25].

The macroscopic theory, described in this chapter, relates the experimentally measurable parameters such as reflectance, polarisation changes and the optical phase shifts to the general parameters that describe the media response, *i.e.* the conductivity tensor, dielectric tensor or index of refraction. It is based on the Maxwell theory, where propagation of light through the media is described in terms of the vectors of the electric field $\mathbf{E}$, magnetic field $\mathbf{H}$, electric displacement $\mathbf{D}$ and magnetic induction $\mathbf{B}$, mutually coupled by the material relations:

$$\begin{aligned}\mathbf{D} &= \epsilon' \mathbf{E} \\ \mathbf{B} &= \mu' \mathbf{H} \\ \mathbf{j} &= \sigma' \mathbf{E}\end{aligned}\tag{2.1}$$

where vector $\mathbf{j}$ is the conductivity current density, ε' is the permittivity tensor, μ' is the magnetic permeability tensor and σ' is the electrical conductivity tensor of the medium. The interaction between the electromagnetic wave and electron spins is important in the radiofrequency, microwave and far-infrared regions, but becomes negligible at optical frequencies [26, 27]. Thus only the interaction of light with the orbital motion of electrons needs to be considered. In this case the relative permeability tensor in (2.1) is represented by a unit matrix and the optical properties of a medium are established by a dielectric tensor that is determined by the motion of the electrons in the medium.

The relation between the complex permittivity and conductivity tensors can be obtained from the wave equation

$$\Delta \mathbf{E} - \frac{\varepsilon'}{c^2} \frac{\partial^2 \mathbf{E}}{\partial t^2} = \frac{4\pi\sigma'}{c^2} \frac{\partial \mathbf{E}}{\partial t} \quad (2.2)$$

Here the dielectric properties of the substance are expressed by the real permittivity tensor ε' , whereas the losses are described by the real conductivity σ' . Considering that $\mathbf{E}$ is harmonic function of time ($\mathbf{E} = \mathbf{E}_0 \exp(i\omega t)$ where ω is the circular frequency) at any given point of space, the first and the second time derivatives are related as

$$\frac{\partial^2 \mathbf{E}}{\partial t^2} = i\omega \frac{\partial \mathbf{E}}{\partial t}. \quad \text{The term containing the conductivity } \sigma' \text{ can be added to the second term}$$

on the left of the equation (2.2) and σ' can be formally included into ε' as its imaginary part. Now the imaginary part of the permittivity tensor corresponds to the losses due to the light adsorption. In the same way ε' can be included in the conductivity tensor [28]. The complex permittivity tensor is related to the permittivity and permeability tensors by

$$\varepsilon_{ij} = \varepsilon'_{ij} - i \frac{4\pi}{\omega} \sigma'_{ij} \quad (2.3)$$

Here the same notation ε for both ellipticity and the permittivity tensor is used, but the two will be easily distinguished from the context. Under the assumption, that the field

acts as a small perturbation of the system we can expand the components of the permittivity tensor [29]. Using the Einstein summation convention ε_{ij} can be written:

$$\varepsilon_{ij} = \varepsilon_{ij}^0 + K_{ijk}M_k + G_{ijkl}M_kM_l + \dots = \varepsilon_{ij}^0 + \varepsilon_{ij}^1 + \varepsilon_{ij}^2 + \dots \quad (2.4)$$

Here the indices i, j, k represent the Cartesian x, y, z axes respectively, M_k are the components of the magnetisation vector, ε_{ij}^0 denotes the components of the permittivity tensor at zero magnetisation ($\mathbf{M} = 0$), K_{ijk} and G_{ijkl} are the components of the linear and quadratic magnetic optical tensors.

Important features of permittivity follow from the symmetry arguments. Under the time-reversal operation, the displacement $\mathbf{D}$ and electric field $\mathbf{E}$ vectors remain unchanged, but the magnetisation field $\mathbf{M}$ changes sign. In this case Onsager's relation gives [30]:

$$\varepsilon_{ij}(\mathbf{M}) = \varepsilon_{ji}(-\mathbf{M}) \quad (2.5)$$

The 3×3 dielectric tensor can be decomposed into symmetric and asymmetric parts [31] and it follows from Eq. (2.5) that the symmetric part of the permittivity tensor is even with $\mathbf{M}$, while the antisymmetric part is odd. The permittivity tensor for different crystal systems has been obtained by Visnovsky [32]. Here we use only the permittivity tensor elements, obtained to the first order of $\mathbf{M}$ (linear magneto-optical effect) in order to describe a crystal with the cubic symmetry.

A general form of the permittivity tensor can be deduced with help of the Neumann's principle, which can be stated as follows: the symmetry elements of any physical property of a crystal must include the symmetry elements of the point group of the crystal [33, 34]. We suppose that a cubic crystal is placed into a magnetic field oriented along the z -direction. The homogeneous magnetisation can be thought of as generated by a current loop perpendicular to $\mathbf{M}$. The cubic crystal holds the 4 and mmm symmetry elements and therefore the permittivity tensor must remain unchanged with respect to the

rotation of the Cartesian coordinate system about z -axis by 90° and its reflection of the z -axis. These conditions can be written as [35]:

$$\boldsymbol{\varepsilon}(\mathbf{M}) = \begin{pmatrix} 0 & 1 & 0 \\ -1 & 0 & 0 \\ 0 & 0 & \pm 1 \end{pmatrix} \boldsymbol{\varepsilon}(\mathbf{M}) \begin{pmatrix} 0 & -1 & 0 \\ 1 & 0 & 0 \\ 0 & 0 & \pm 1 \end{pmatrix} \quad (2.6)$$

The 2×2 blocks are the transformation matrixes for $i, j = x, y$ which give the rotation by 90° about z -axis (see Eq. 1.7) and the diagonal elements equal to -1 give the reflection by the xy plane. The general form of the permittivity tensor, satisfying Eq.(2.6) is

$$\boldsymbol{\varepsilon}(\mathbf{M}) = \begin{pmatrix} \varepsilon_{xx} & \varepsilon_{xy} & 0 \\ -\varepsilon_{xy} & \varepsilon_{xx} & 0 \\ 0 & 0 & \varepsilon_{zz} \end{pmatrix} \quad (2.7)$$

with $\varepsilon_{xx} = \varepsilon_{zz}$ for a cubic crystal. Reflection by a plane containing the z -axis changes the sense of direction of current in the loop, which is equivalent to a magnetisation reversal. Due to the cubic symmetry, $\boldsymbol{\varepsilon}(\mathbf{M})$ must be invariant to the reflection by a plane perpendicular to the x -axis. This condition can be written as:

$$\boldsymbol{\varepsilon}(-M_z) = \begin{pmatrix} -1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 1 \end{pmatrix} \boldsymbol{\varepsilon}(M_z) \begin{pmatrix} -1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 1 \end{pmatrix} \quad (2.8)$$

Substituting $\boldsymbol{\varepsilon}(\mathbf{M})$ given by Eq. (2.7) into Eq. (2.8) leads to

$$\begin{aligned} \varepsilon_{xx}(-M_z) &= \varepsilon_{xx}(M_z) \\ \varepsilon_{xy}(-M_z) &= -\varepsilon_{xy}(M_z) \end{aligned} \quad (2.9)$$

Thus the diagonal elements of the permittivity tensor are even functions of magnetisation while the off-diagonal are odd and so they cancel in a non-magnetised medium. As a

result the first term in (2.4) corresponds to the symmetric part of the permittivity tensor and for a cubic crystal can be written as:

$$\boldsymbol{\varepsilon}^0 = N^2 \begin{pmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 1 \end{pmatrix} \quad (2.10)$$

where $N = \sqrt{\varepsilon_{xx}}$ is the principal refractive index of the medium. The form of the asymmetric part $\boldsymbol{\varepsilon}^1$, that gives rise to the Kerr effect, can be obtained using (2.5) that gives for the permittivity tensor elements linear in M_k

$$\varepsilon_{ij}^1 = K_{ijk} M_k = -K_{jik} M_k = -\varepsilon_{ji}^1 \quad (2.11)$$

which leads to

$$K_{iik} = 0 ; K_{ijk} = -K_{jik} \quad (2.12)$$

The crystal symmetry when it is present simplifies the second term $\boldsymbol{\varepsilon}^1$ in (2.4) even further and for a cubic system its components are given by [32]

$$\begin{pmatrix} \varepsilon_{yz}^1 \\ \varepsilon_{zx}^1 \\ \varepsilon_{xy}^1 \\ \varepsilon_{zy}^1 \\ \varepsilon_{xz}^1 \\ \varepsilon_{yx}^1 \end{pmatrix} = \begin{pmatrix} K_{123} & 0 & 0 \\ 0 & K_{123} & 0 \\ 0 & 0 & K_{123} \\ -K_{123} & 0 & 0 \\ 0 & -K_{123} & 0 \\ 0 & 0 & -K_{123} \end{pmatrix} \begin{pmatrix} M_x \\ M_y \\ M_z \end{pmatrix} \quad (2.13)$$

If we choose the coordinate system as shown in the Fig. 1.7 (z-axis is perpendicular to the sample surface and x-axis is perpendicular to the scattering plane), the permittivity tensor for the *polar magnetisation* ($\mathbf{M}$ is along z-axis) takes the form

$$\boldsymbol{\varepsilon} = N^2 \begin{pmatrix} 1 & K_{123}M/N^2 & 0 \\ -K_{123}M/N^2 & 1 & 0 \\ 0 & 0 & 1 \end{pmatrix} = N^2 \begin{pmatrix} 1 & iQ & 0 \\ -iQ & 1 & 0 \\ 0 & 0 & 1 \end{pmatrix} \quad (2.14)$$

$Q = -iK_{123}M/N^2$ is the magneto-optic Voigt constant, which is proportional to the magnetisation [23, 36]. For the *longitudinal* ($\mathbf{M}$ is along y -axis) and *transverse* ($\mathbf{M}$ is along x -axis) configurations (2.13) gives:

$$\boldsymbol{\varepsilon} = N^2 \begin{pmatrix} 1 & 0 & iQ \\ 0 & 1 & 0 \\ -iQ & 0 & 1 \end{pmatrix} \quad (2.15)$$

$$\boldsymbol{\varepsilon} = N^2 \begin{pmatrix} 1 & 0 & 0 \\ 0 & 1 & iQ \\ 0 & -iQ & 1 \end{pmatrix} \quad (2.16)$$

2.2 Zak formalism for magneto-optics; the medium boundary matrix

Zak and co-workers provided guidelines, useful for the description of magneto-optic phenomena in ultrathin films and multilayer film structures [37,38]. They created a multiple reflection formalism, which can describe the magneto-optic effects to the first order, in a magnetic field, in layered magnetic materials with the magnetisation vector in each layer pointing in an arbitrary direction. In this formalism both magnetic and non-magnetic films are treated on the same footing and each layer in a multilayer stack can be either magnetic or non-magnetic. The relation between magneto-optical coefficients, defined by (1.39), is obtained by using the *medium boundary* and *medium propagation* matrices. This part presents a brief guide to the formalism and the final results for the most commonly used longitudinal and polar configurations.

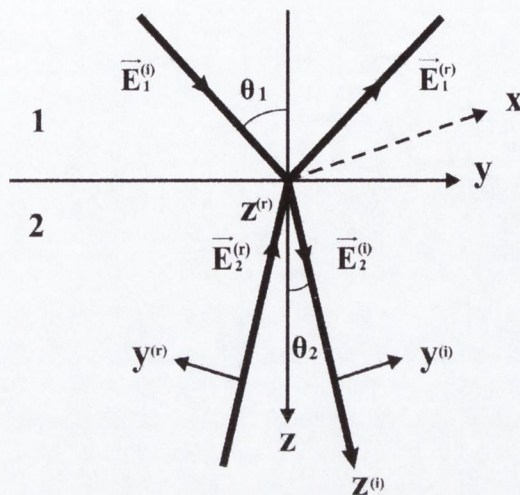


Figure 2.1. Two media, separated by an xy – plane boundary. The waves going in z -direction are denoted as $E^{(i)}$ and the ones in the opposite direction $E^{(r)}$. $(y^{(i)}, z^{(i)})$ and $(y^{(r)}, z^{(r)})$ are the local coordinate systems.

Zak *et al.* started by considering that a beam of light, travelling from a medium 1 into medium 2 conserves the tangential components of its electric E_x , E_y and magnetic H_x , H_y fields, where the xy – plane is the boundary separating the two media like in figure 2.1. The set of fields $E_s^{(i)}, E_p^{(i)}, E_s^{(r)}, E_p^{(r)}$, which defines the magneto-optic coefficients in Eq. (1.39) is connected to these tangential components via the medium boundary matrix A :

$$F = AP \quad (2.17)$$

where F and P are column vectors defined as:

$$F = \begin{pmatrix} E_x \\ E_y \\ H_x \\ H_y \end{pmatrix}; \quad P = \begin{pmatrix} E_s^{(i)} \\ E_p^{(i)} \\ E_s^{(r)} \\ E_p^{(r)} \end{pmatrix} \quad (2.18)$$

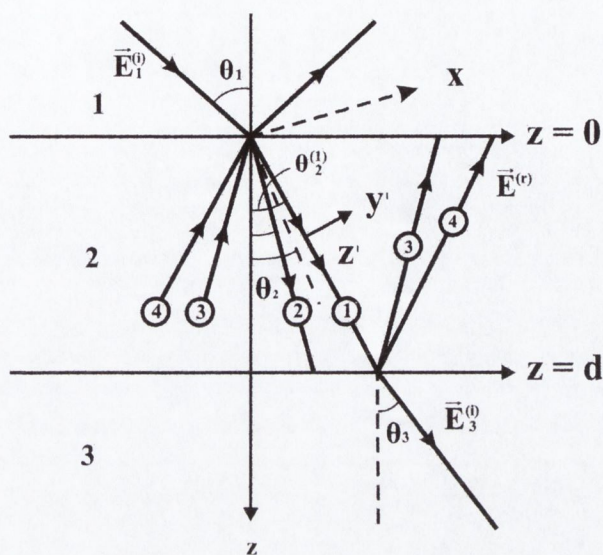


Figure 2.2 Three media system; (y', z') is a local coordinate system for the wave 1. $z = 0$ and $z = d$ are the film boundaries. 1, 2 denote two circular polarised waves in the magnetic medium 2 that add up to form $E^{(i)}$; 3 and 4 are the same for $E^{(r)}$; $\theta_2^{(i)}$ is the direction angle for the wave 1. $\theta_2^{(j)}$, $j = 2, 3, 4$ are not shown in the figure.

If neither of the two media is magnetic then the angles θ_1 and θ_2 are the angles that appear in Snell's law:

$$N_1 \alpha_{1y} = N_2 \alpha_{2y} \quad (2.19)$$

where N_1 and N_2 are the refractive indices of the media 1 and 2 respectively and $\alpha_{1y} = \sin \theta_1$ and $\alpha_{2y} = \sin \theta_2$. If one of the media is magnetic (e.g. medium 2) there are two circularly polarised waves, propagating in each direction in the magnetic medium [30] (Fig. 2.2). The two rays $E^{(1)}$ and $E^{(2)}$ correspond to $E^{(i)}$, and $E^{(3)}$ and $E^{(4)}$ to $E^{(r)}$. The dependence between the components of $E^{(j)}$, ($j = 1, 2, 3, 4$) is used to construct the A matrix elements. To find this dependence the electrical displacement vector $\mathbf{D}$ is used, which is connected to the electric field components via the dialectic tensor ϵ_{ij} :

$$D_i = \varepsilon_{ij} E_j \quad (2.20)$$

where ε_{ij} is determined by equations (2.14) and (2.15) for the polar and longitudinal MOKE respectively. As a result the A matrix elements are constructed from the Q and N values of the media as well as the geometric angles of the problem. A detailed discussion on construction of the boundary and propagation matrices and derivation of the magneto-optic coefficients can be found in Refs. [37, 38].

In the simple case of the two-media, one-boundary system (Fig. 2.1), the boundary matching condition can be written as:

$$A_1 P_1 = A_2 P_2 \quad (2.21)$$

If there are more than two media in the stack, Eq. (2.21) still holds for any boundary, however one needs to describe the wave propagation inside the film from the boundary at $z = 0$ to the boundary at $z = d$, where d is the film thickness. This is achieved by means of the *medium propagation matrix* $\bar{D}$, defined as:

$$P_2(z = 0) = \bar{D} P_2(z = d) \quad (2.22)$$

Using the boundary matching conditions (2.21) and the definition of the propagation matrix (2.22) one can write for the most frequent three media – two boundaries case:

$$A_1 P_1 = A_2 P_2(0) = A_2 \bar{D} P_2(d) = A_2 \bar{D} A_2^{-1} A_2 P_2(d) = A_2 \bar{D} A_2^{-1} A_3 P_3(d) \quad (2.23)$$

where A_1 and A_3 are the medium boundary matrixes for the media 1 and 3, while $A_2 \bar{D} A_2^{-1}$ is the matrix for the medium 2. The equation (2.23) can be generalised for any number of films by replacing $A_2 \bar{D} A_2^{-1}$ by the product of the film matrixes:

$$A_i P_i = \left(\prod_m A_m \bar{D}_m A_m^{-1} \right) A_{sub} P_{sub} \quad (2.24)$$

where the subscripts i and sub correspond to the initial media (1 in the previous notation) and the substrate respectively. All three media can be either magnetic or non-magnetic.

The expression (2.24) can be put in the form $P_i = T P_s$, where

$$T = A_i^{-1} \prod_m A_m \bar{D}_m A_m^{-1} A_{sub} = \begin{pmatrix} G & H \\ I & J \end{pmatrix} \quad (2.25)$$

The magneto-optical coefficients can be obtained using the 2×2 matrixes G and I composed of the magneto-optic parameters:

$$G^{-1} = \begin{pmatrix} t_{ss} & t_{sp} \\ t_{ps} & t_{pp} \end{pmatrix} \text{ and } I G^{-1} = \begin{pmatrix} r_{ss} & r_{sp} \\ r_{ps} & r_{pp} \end{pmatrix} \quad (2.26)$$

The equation (2.24) significantly simplifies in the ultrathin limit defined by

$$\frac{2\pi}{\lambda} \sum |N_m| d_m \ll 1 \quad (2.27)$$

where λ is a wave length, N_m is the refractive index of the m -th film in the stack, d_m is its thickness and the sum is over all films in the stack. Equation (2.27) defines the condition to distinguish between the surface magneto optic Kerr-effect (SMOKE) and the magneto optic Kerr effect (MOKE), observed at the reflection of light from the bulk sample. In the ultrathin limit, the equation for the magneto-optical coefficients (2.24) can be solved analytically for small angles of incidence:

$$r_{ps}^{(POL)} = r_{sp}^{(POL)} = -\frac{4\pi}{\lambda} \frac{N_i}{(N_i + N_{sub})^2} \sum N_m^2 Q_m d_m \quad (2.28)$$

$$r_{ps}^{(LON)} = -r_{sp}^{(LON)} = \frac{4\pi}{\lambda} \frac{\alpha_{iy} N_i^2 N_{sub}}{(N_i + N_{sub})^2} \sum Q_m d_m \quad (2.29)$$

$$r_{ss} = \frac{N_i - N_{sub}}{N_i + N_{sub}} ; \quad r_{pp} = \frac{N_{sub} - N_i}{N_{sub} + N_i} \quad (2.30)$$

where the summation is over the magnetic films only. The equations (2.28) and (2.29) establish the additive law for the Kerr effect in the ultra-thin limit, experimentally confirmed in multiple studies [39, 40, 41]. By substituting the magneto-optical coefficients into the equations (1.41) and (1.43) one can obtain the expressions for the complex Kerr rotation for *s* or *p*-polarised light reflected from a multilayer system. For a system, like that used in our experiments, consisting of a single magnetic film, characterised by *N*, *Q* and *d* on a non-magnetic substrate the polar and longitudinal Kerr effects become:

$$\phi^{(POL)} = \mp \left(\frac{4\pi}{\lambda} \right) \left(\frac{N^2}{1 - N_{sub}^2} \right) Qd ; \quad \phi^{(LON)} = \left(\frac{4\pi}{\lambda} \right) \left(\frac{N_{sub}}{1 - N_{sub}^2} \right) \theta Qd \quad (2.31)$$

where $\mp$ stand for the *s* and *p* – polarised incident light respectively.

Though Eqs. (2.31) were obtained for a particular case they are also relevant to the experimental Kerr effect study. It follows from these equations that $\phi^{(POL)} \gg \phi^{(LON)}$ due to an extra *N* factor. Due to this fact a close attention needs to be paid to the alignment of the applied magnetic field with respect to the film plane in the longitudinal MOKE measurements. The sample misalignment will produce an out-of-plane component of magnetisation and a polar contribution to the Kerr effect will produce a distorted hysteresis loop, similar to that shown in Fig. 2.3. The longitudinal Kerr effect is independent of the refractive index of the magnetic layer; however it is dependent on the

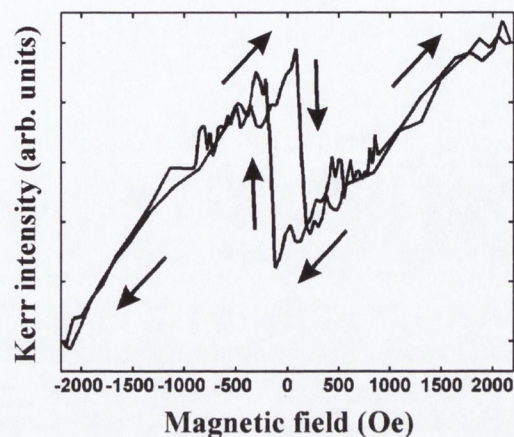


Figure 2.3 Longitudinal hysteresis loop measured along magnetically hard [1-10] direction of a 4 monolayer Fe film on Mo(110) surface. The loop is distorted by polar Kerr effect contribution, which appears due to misalignment of the applied magnetic field and the film plane.

angle of incidence so that a better sensitivity to the longitudinal magnetisation can be achieved by using a larger angle of incidence. Both $\phi^{(POL)}$ and $\phi^{(LON)}$ have $(N_i^2 - N_{sub}^2)$ in the denominator and the Kerr intensity can be enhanced by a proper choice of a substrate.

Chapter 3

Experimental methods of magneto-optics

When the Kerr effect is used to probe the magnetic properties of the bulk magnetic samples the magneto-optic angles of about $0.2\text{--}0.5^\circ$ are observed [21, 22] and the changes of the state of polarisation can be easily detected. However, the effect is reduced by one or two orders of magnitude for thin and ultrathin magnetic films or multilayers [41, 42] and the requirements for the sensitivity of the experiment significantly increase. This chapter describes the four experimental techniques, most frequently used in magneto-optical studies.

3.1 Nearly crossed polarizers intensity detection

The nearly crossed polarizers technique is the earliest method of magneto-optics. With some modifications it is still favoured for many applications due to its simplicity. It is used for integral and local probing of the magnetic samples, high resolution magnetic domain imaging [43] and quality control of recording media. Figure 10 outlines a straightforward DC detection scheme. It includes a monochromatic light source, polarizer, sample, analysing polarizer and photodiode. The axis of polarisation of the analyser is set at an angle α from the extinction position.

In case of a p -polarised incident beam, as used in our experiments, the Jones vector describing the polarisation of light that reaches the photodiode is given by a product:

$$\mathbf{J} = \begin{pmatrix} 1 & 0 \\ 0 & 0 \end{pmatrix} \begin{pmatrix} \cos \alpha & \sin \alpha \\ -\sin \alpha & \cos \alpha \end{pmatrix} \begin{pmatrix} r_{ss} & r_{sp} \\ r_{ps} & r_{pp} \end{pmatrix} \begin{pmatrix} 0 \\ 1 \end{pmatrix} = \begin{pmatrix} r_{sp} \cos \alpha + r_{pp} \sin \alpha \\ 0 \end{pmatrix} \quad (3.1)$$

The intensity of the output beam is:

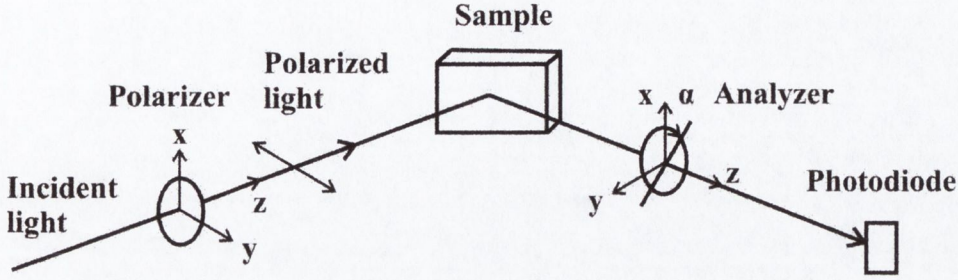


Figure 3.1 Schematic of a nearly crossed polarizers intensity detection MOKE experiment. A polarizer is used to produce *s*- or *p*- polarised incident light. The analyser is set at an angle α from the extinction position. The intensity of light reaching the photodiode changes when the sample is magnetised and the photodiode output signal is proportional to Kerr rotation.

$$I = \mathbf{J}^+ \mathbf{J} = I_0 |r_{pp}|^2 \left(\sin^2 \alpha + \theta_p \sin 2\alpha + \frac{|r_{sp}|^2}{|r_{pp}|^2} \cos^2 \alpha \right) \approx I_M + \Delta I \quad (3.2)$$

where I_0 is the intensity of the light transmitted through the first polarizer, $\theta_p = \text{Re}(r_{sp}/r_{pp})$ is the Kerr rotation and $|r_{pp}|^2$ is the intensity reflection coefficient of the sample. The first term I_M represents the classical Malus law, while the second ΔI , which has its maximum at $\alpha = \pi/4$, appears due to the magneto-optical contribution from the sample and referred to as the Kerr intensity. The third quadratic term can be neglected because of the small magnitude of the Kerr effect. From the experimental point of view a high contrast ($\Delta I/I_M$) is important. It can be maximised by the appropriate choice of the offset angle α . Depending on the magnitude of the Kerr intensity and the quality of polarizers α is usually set between 2° and 10° .

The setup can be modified for ellipticity measurements. To do so a quarter wave plate with the fast axis perpendicular to the polarising axis of the analyser can be placed after the sample. In this case, the Jones vector of the output light is given by a matrix product:

$$\mathbf{J} = \begin{pmatrix} 1 & 0 \\ 0 & 0 \end{pmatrix} \begin{pmatrix} -i & 0 \\ 0 & 1 \end{pmatrix} \begin{pmatrix} \cos \alpha & \sin \alpha \\ -\sin \alpha & \cos \alpha \end{pmatrix} \begin{pmatrix} r_{ss} & r_{sp} \\ r_{ps} & r_{pp} \end{pmatrix} \begin{pmatrix} 0 \\ 1 \end{pmatrix} = -i \begin{pmatrix} r_{sp} \cos \alpha + r_{pp} \sin \alpha \\ 0 \end{pmatrix} \quad (3.3)$$

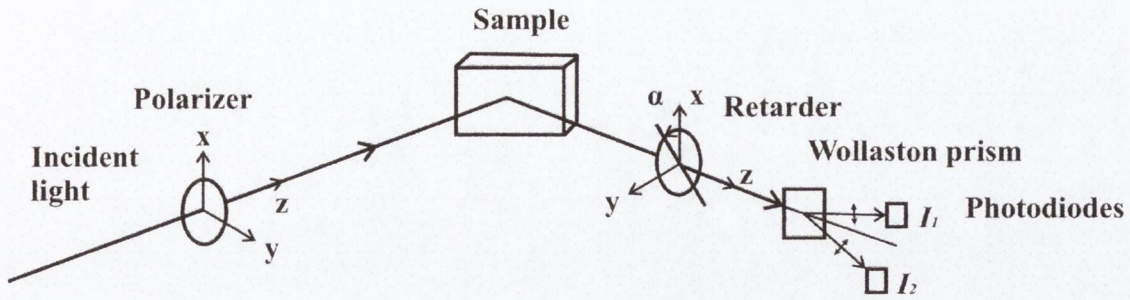


Figure 3.2 Schematic drawing of a differential intensity detection MOKE experiment using a Wollaston prism. The Wollaston prism splits the reflected light into two orthogonally linear polarised beams. Initially the intensity difference between the beams is set to zero. When the sample is magnetised, the change in the intensity difference is proportional to the Kerr effect.

and the Kerr intensity becomes sensitive to the ellipticity $\varepsilon_p = \text{Im}(r_{sp}/r_{pp})$ of the transmitted light:

$$I = \mathbf{J}^+ \mathbf{J} = I_0 |r_{pp}|^2 \left(\sin^2 \alpha + \varepsilon_p \sin 2\alpha + \frac{|r_{sp}|^2}{|r_{pp}|^2} \cos^2 \alpha \right) \approx I_M + \Delta I \quad (3.4)$$

The expressions for intensity of a *s*-polarised wave are similar to equations (3.2) and (3.4) and can be obtained in the same manner.

3.2 Differential intensity detection

The differential intensity detection setup shown in figure 3.2 is also quite simple but due to the differential detection is less sensitive to the intensity instability of the light source and has larger signal-to-noise ratio than the crossed polarizers intensity detection method. This makes it the basic technique for the magneto-optical reading in erasable data storage systems [44]. The optical setup consists of a light source, polarizer, with the polarisation axis oriented along *s* (*x*) or *p* (*y*) – direction, sample, retarder, Wollaston prism or polarising beam splitter and two photodiodes. The Wollaston prism separates the light

into two orthogonal linear polarised waves and the difference in the intensity between these two waves is measured. By utilising a half-wave plate (retardation π radians) the setup can be used to measure the Kerr rotation of the reflected light. In this case the Jones vector for the initially s -polarised wave at the input of the Wollaston prism is given by the matrix product:

$$\mathbf{J} = \frac{1}{\sqrt{2}} \begin{pmatrix} \cos \alpha & -\sin \alpha \\ \sin \alpha & \cos \alpha \end{pmatrix} \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix} \begin{pmatrix} \cos \alpha & \sin \alpha \\ -\sin \alpha & \cos \alpha \end{pmatrix} \begin{pmatrix} r_{ss} & r_{sp} \\ r_{ps} & r_{pp} \end{pmatrix} \begin{pmatrix} 1 \\ 0 \end{pmatrix} \quad (3.5)$$

where α is the angle between the fast-axis of the half-wave plate and the x -axis. The Jones vector takes the form:

$$\mathbf{J} = \frac{1}{\sqrt{2}} r_{ss} \begin{pmatrix} \cos 2\alpha + \frac{r_{ps}}{r_{ss}} \sin 2\alpha \\ \sin 2\alpha + \frac{r_{ps}}{r_{ss}} \sin 2\alpha \end{pmatrix} \quad (3.6)$$

The difference between the intensities of the two orthogonal components measured by the photodiodes is

$$\Delta I = I_1 - I_2 = \mathbf{J}_x^\dagger \mathbf{J}_x - \mathbf{J}_y^\dagger \mathbf{J}_y = \frac{I_0}{2} |r_{ss}|^2 \left[\cos 4\alpha \left(1 - \frac{|r_{ps}|^2}{|r_{ss}|^2} \right) + 2 \sin 4\alpha \operatorname{Re} \left(\frac{r_{ps}}{r_{ss}} \right) \right] \quad (3.7)$$

where I_0 is the intensity of light after the polarizer. First, the intensity difference between the two photodiodes is set to zero by setting the angle $\alpha = \frac{\pi}{8}(2n+1)$ (n is any integer number) in the absence of the magnetic field ($r_{ps} = 0$). When the magnetic field is applied the differential signal is proportional to the Kerr rotation and is given by:

$$\Delta I \approx I_0 |r_{ss}|^2 \operatorname{Re} \left(\frac{r_{ps}}{r_{ss}} \right) = I_0 |r_{ss}|^2 \theta_s \quad (3.8)$$

The calibrating constant, proportional to the reflectivity of the sample $|r_{ss}|^2$ can be found from the change of the signal, corresponding to a small rotation $\Delta\alpha$ of the polariser. Substituting a half-wave plate for a quarter-wave plate makes the setup sensitive to the ellipticity. In this case the Jones vector for initially s -polarised light can be found from:

$$\begin{aligned} \mathbf{J} &= \frac{1}{\sqrt{2}} \begin{pmatrix} \cos \alpha & -\sin \alpha \\ \sin \alpha & \cos \alpha \end{pmatrix} \begin{pmatrix} 1 & 0 \\ 0 & -i \end{pmatrix} \begin{pmatrix} \cos \alpha & \sin \alpha \\ -\sin \alpha & \cos \alpha \end{pmatrix} \begin{pmatrix} r_{ss} & r_{sp} \\ r_{ps} & r_{pp} \end{pmatrix} \begin{pmatrix} 1 \\ 0 \end{pmatrix} = \\ &= \frac{1}{\sqrt{2}} r_{ss} \begin{pmatrix} \cos^2 \alpha - i \sin^2 \alpha + \frac{r_{ps}}{r_{ss}} (1+i) \sin \alpha \cos \alpha \\ (1+i) \sin \alpha \cos \alpha + \frac{r_{ps}}{r_{ss}} (\sin^2 \alpha - i \cos \alpha) \end{pmatrix} \end{aligned} \quad (3.9)$$

The difference between the s and p – components of light incident on the Wollaston prism is then:

$$\Delta I = \frac{I_0}{2} |r_{ss}|^2 \left[\cos^2 2\alpha \left(1 - \frac{|r_{ps}|^2}{|r_{ss}|^2} \right) + 2 \sin 2\alpha \cos 2\alpha \operatorname{Re} \left(\frac{r_{ps}}{r_{ss}} \right) - 2 \sin 2\alpha \operatorname{Im} \left(\frac{r_{ps}}{r_{ss}} \right) \right] \quad (3.10)$$

For $\alpha = \frac{\pi}{4} (2n+1)$ the intensity difference becomes proportional to the ellipticity:

$$\Delta I = I_0 |r_{ss}|^2 \operatorname{Im} \left(\frac{r_{ps}}{r_{ss}} \right) = I_0 |r_{ss}|^2 \epsilon_s \quad (3.11)$$

These calculations can be modified for the p -polarisation of the incident light.

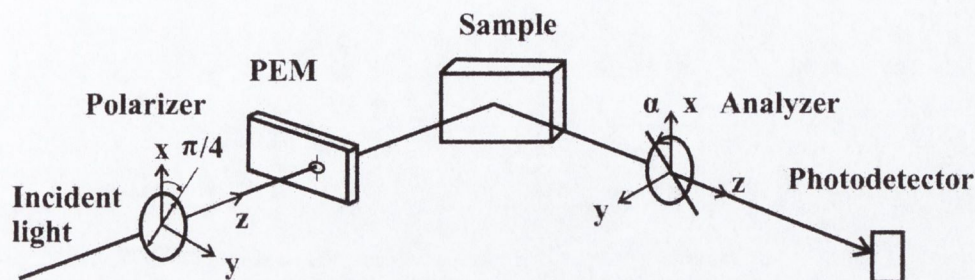


Figure 3.3 Schematic of the ellipticity modulation MOKE setup using a photoelastic modulator, which introduces harmonic oscillations into the incident light polarisation. The photodiode signal, which is proportional to Kerr effect, is detected using the lock-in detection technique.

3.3 Modulation techniques

Modulation techniques use special optical devices to introduce time-harmonic changes in the polarisation state. As a result the detected signal contains an oscillating component, which can be measured using lock-in detection. There are two principal modulation methods used in magneto-optics; the *ellipticity modulation technique* and the *azimuth modulation technique*. The first one makes use of the photoelastic effect to introduce an oscillating phase shift between two orthogonal directions and the second one employs the Faraday effect to oscillate the angle of the plane of polarisation.

Figure 3.3 shows a typical optical layout for the *ellipticity modulation technique* [45]. The main feature of this setup is the use of a photoelastic modulator (PEM). Typically it is an isotropic optical material (silica or calcium fluoride) stressed by compression and stretching at the frequency of ω_m along the modulator's principal optical axis. This periodic deformation is produced by a piezoelectric transducer at $\omega_m \sim 50$ kHz (the resonant frequency of the piezoelectric ceramic) making the modulator act as a phase plate with the time-harmonic phase shift $\delta = \delta_0 \sin \omega_m t$. The amplitude of the shift δ_0 is adjusted by changing the voltage applied to the transducer. To illustrate the operation of the ellipsometer with a PEM the calculation is performed for the following sequence of optical elements: a polarizer, making the angle $\pi/4$ to the s -direction, a PEM with the principal axis oriented along the x (s)-axis, sample and analyzer. A detailed analysis of

this technique has been done by Z. Yang in [46]. The result presented here is obtained for the polar geometry with the normal incidence of light. In this case the difference between the s and p -polarised waves disappears and the calculations simplify [47]. Using (2.28) and (2.30) the Jones vector of the output light is obtained from the matrix product:

$$\begin{aligned} \mathbf{J} &= \frac{r}{\sqrt{2}} \begin{pmatrix} \cos^2 \alpha & \sin \alpha \cos \alpha \\ \sin \alpha \cos \alpha & \sin^2 \alpha \end{pmatrix} \begin{pmatrix} 1 & -\phi \\ -\phi & -1 \end{pmatrix} \begin{pmatrix} e^{i\delta/2} & 0 \\ 0 & e^{-i\delta/2} \end{pmatrix} \begin{pmatrix} 1 \\ 1 \end{pmatrix} = \\ &= \frac{r}{\sqrt{2}} \left(\cos \alpha e^{i\delta/2} - \sin \alpha e^{-i\delta/2} - \phi [\cos \alpha e^{-i\delta/2} + \sin \alpha e^{i\delta/2}] \right) \begin{pmatrix} \cos \alpha \\ \sin \alpha \end{pmatrix} \end{aligned} \quad (3.12)$$

where r is the complex reflection coefficient of the sample and ϕ is the complex Kerr rotation. Using the small magneto-optic angles approximation we obtain the expression for the output intensity:

$$I = \mathbf{J}^+ \mathbf{J} = \frac{|r|^2}{4} (1 - \cos \delta \sin(2\alpha + 2\theta) + 2\epsilon \sin \delta) \quad (3.13)$$

Since δ is a harmonic function of time the $\cos \delta$ and $\sin \delta$ can be expanded in series of δ using:

$$\begin{aligned} \sin(\delta_0 \sin \omega_m t) &= 2 \sum_{k=1}^{\infty} J_{2k-1}(\delta_0) \sin((2k-1)\omega_m t); \\ \cos(\delta_0 \sin \omega_m t) &= J_0(\delta_0) + 2 \sum_{k=1}^{\infty} J_{2k}(\delta_0) \cos(2k\omega_m t) \end{aligned} \quad (3.14)$$

where $J_k(\delta_0)$ gives the i -th order of the Bessel function. Substituting (3.14) into (3.13) and negating the harmonics higher than $2\omega_m$ gives:

$$I \approx \frac{|r|^2}{4} (1 - J_0(\delta_0) \sin(2\alpha + 2\theta) + 4\epsilon J_1(\delta_0) \sin \omega_m t - 2 \sin(2\alpha + 2\theta) J_2(\delta_0) \cos 2\omega_m t) \quad (3.15)$$

Thus the ratio between the intensities of the first and second harmonics to the DC component:

$$\frac{I_\omega}{I_{dc}} = \frac{4\epsilon J_1(\delta_0)}{1 - J_0(\delta_0) \sin(2\alpha + 2\theta)}; \quad \frac{I_{2\omega}}{I_{dc}} = \frac{-2 \sin(2\alpha + 2\theta) J_2(\delta_0)}{1 - J_0(\delta_0) \sin(2\alpha + 2\theta)} \quad (3.16)$$

$J_0(\delta_0)$ can be set to zero by the proper choice of the amplitude of the modulation signal δ_0 . For a Kerr rotation $\theta \ll \pi/4$ the intensity of the $2\omega_m$ component has two maximums when the analyzer is oriented at $\alpha \approx \pm \pi/4$. For $J_0(\delta_0) = 0$ these maximums become symmetric with respect to the zero intensity and the magnitude of δ_0 can be accurately adjusted. When it is done the output intensity becomes:

$$\frac{I_\omega}{I_{dc}} = 4\epsilon J_1(\delta_0); \quad \frac{I_{2\omega}}{I_{dc}} = -2 \sin(2\alpha + 2\theta) J_2(\delta_0) \quad (3.17)$$

The first harmonic ω_m is now proportional to the Kerr ellipticity, while the second harmonic $2\omega_m$ to the Kerr rotation for $\alpha = n\pi$ (n is any integer).

The schematic optical setup for the *azimuth modulation technique* is displayed in Figure 3.4. It makes use of a Faraday cell to modulate the orientation of the polarisation plane of the incident linear polarised light with an amplitude generally smaller than 3° . The Faraday cell consists of a quartz or glass rod placed inside a water cooled solenoid, which generates an oscillating magnetic field along its axis. The amplitude β_0 and frequency ω_m of modulation are determined by the amplitude and frequency of the solenoid current. The full sequence of the optical elements includes a monochromatic light source, a

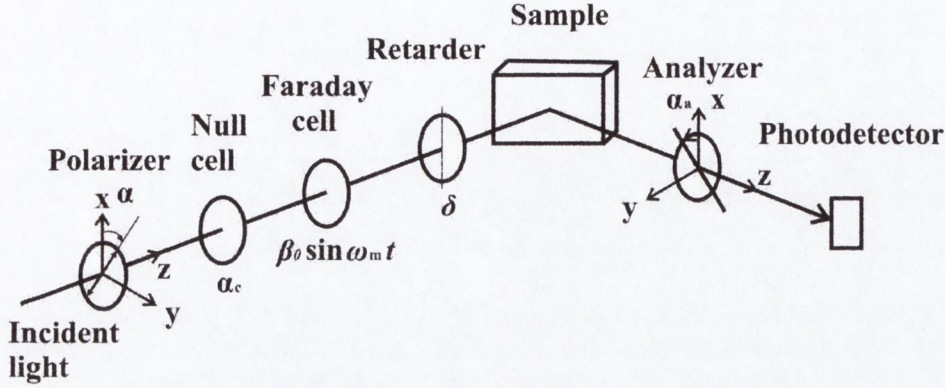


Figure 3.4 Schematic drawing of the azimuth modulation MOKE setup using a Faraday cell to modulate the orientation of the polarisation plane. The output voltage of the photodetector is applied to the input of the negative feedback circuit, which controls the current in the null cell coil.

polarizer (oriented at the small angle α to the s -axis), Faraday compensation (or null) cell (producing the rotation of the polarisation plane α_c), Faraday cell, phase plate (with the fast axis along the s -axis and retardation δ), sample, analyzer (oriented at α_a), and photodetector. The output voltage of the photodetector is applied to the input of the negative feedback circuit, which controls the current in the compensation cell coil [48, 49]. The Jones vector for this sequence can be found from the matrix product:

$$\mathbf{J} = \frac{r}{\sqrt{2}} \begin{pmatrix} \cos^2 \alpha_a & \sin \alpha_a \cos \alpha_a \\ \sin \alpha_a \cos \alpha_a & \sin^2 \alpha_a \end{pmatrix} \begin{pmatrix} r_{ss} & r_{sp} \\ r_{ps} & r_{pp} \end{pmatrix} \begin{pmatrix} e^{i\delta/2} & 0 \\ 0 & e^{-i\delta/2} \end{pmatrix} \times \quad (3.18)$$

$$\begin{pmatrix} \cos(\beta_0 \sin \omega_m t) & -\sin(\beta_0 \sin \omega_m t) \\ \sin(\beta_0 \sin \omega_m t) & \cos(\beta_0 \sin \omega_m t) \end{pmatrix} \begin{pmatrix} \cos \alpha_c & -\sin \alpha_c \\ \sin \alpha_c & \cos \alpha_c \end{pmatrix} \begin{pmatrix} \sin \alpha \\ \cos \alpha \end{pmatrix}$$

Denoting $\alpha + \alpha_c + \beta_0 \sin \omega_m t$ as Ω the Jones vector can be written:

$$\Omega \equiv \alpha + \alpha_c + \beta_0 \sin \omega_m t \quad (3.19)$$

$$\mathbf{J} = \begin{pmatrix} \cos^2 \alpha_a (r_{ss} e^{i\delta/2} \cos \Omega + r_{sp} e^{-i\delta/2} \sin \Omega) \\ \sin^2 \alpha_a (r_{ps} e^{i\delta/2} \cos \Omega + r_{pp} e^{-i\delta/2} \sin \Omega) \end{pmatrix} \quad (3.20)$$

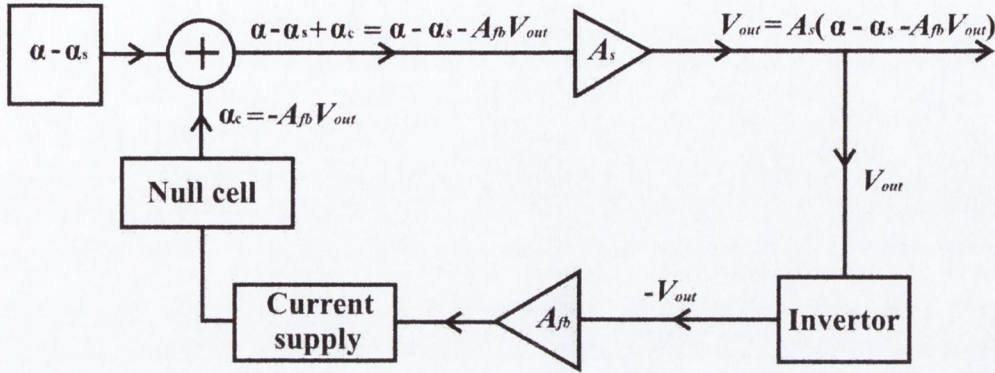


Figure 3.5 Negative feedback loop used in the azimuth modulation technique. The role of the null cell is to adjust the output signal to zero using negative feedback, to compensate for any changes of the intensity due to the magneto optic effect.

For the analyser oriented along the p -direction ($\alpha_a = \pi/2$) and small angles α , α_c and β_0 the intensity of the output signal is:

$$I = \mathbf{J}^+ \mathbf{J} \approx \frac{1}{2} \left[|r_{ps}|^2 + |r_{pp}|^2 \Omega^2 + (r_{ps} r_{pp}^* e^{i\delta} + r_{ps}^* r_{pp} e^{-i\delta}) \Omega \right] \quad (3.21)$$

Thus the contribution from the component oscillating at ω_m frequency is given by:

$$I_\omega \approx |r_{pp}|^2 (\alpha + \alpha_c - \alpha_s) \beta_0 \sin \omega_m t \quad (3.22)$$

where $\alpha_s = -\text{Re} \left(\frac{r_{ps}}{r_{pp}} e^{i\delta} \right)$ corresponds to the impact from the magneto-optical effect when the light is reflected from a magnetised sample.

The role of the null cell is to adjust I_ω to zero through negative feedback, to compensate for any changes of the intensity due to the magneto optical effect. This can be realised by using the circuit shown in Figure 3.5, where the amplification coefficients of the setup and the negative feedback circuit are marked as A_s and A_{fb} respectively:

$$A_s = \frac{\partial V_{out}}{\partial (\alpha + \alpha_c - \alpha_s)} \quad (V / \text{deg}); \quad A_{fb} = \frac{\partial \alpha_c}{\partial V_{out}} \quad (\text{deg}/V) \quad (3.23)$$

If the inverted output signal is applied to the input of the compensation cell coil then

$$\alpha_c = -A_{fb} V_{out} \quad \text{and} \quad V_{out} = A_s (\alpha - \alpha_s - A_{fb} V_{out}) \quad (3.24)$$

which yields

$$V_{out} = \frac{A_s}{1 + A_s A_{fb}} (\alpha - \alpha_s) \quad \text{or} \quad \alpha_c = -\frac{A_s A_{fb}}{1 + A_s A_{fb}} (\alpha - \alpha_s) \quad (3.25)$$

The product $A = A_s A_{fb}$ is the total amplification of the feedback loop and in case $A \gg 1$:

$$\alpha_c + \alpha - \alpha_s \approx 0 \quad (3.26)$$

Substituting the first equation of (3.24) into (3.26) yields:

$$V_{out} \approx \frac{\alpha - \alpha_s}{A_{fb}} \quad (3.27)$$

Thus the output signal of the photodetector is proportional to the magneto-optical effect α_s , produced by the magnetised sample. One can make this technique sensitive to the Kerr rotation or ellipticity defined by Eq. (1.43) if the incident beam makes a small angle with the sample normal. Using the relation $r_{ps} = r_{sp}$ for the polar configuration, given by Eq. (2.28) yields:

$$\alpha_s = -\text{Re} \left(\frac{r_{sp}}{r_{pp}} e^{i\delta} \right) = -\text{Re}(\phi_p e^{i\delta}) = \theta_p \cos \delta + \varepsilon_p \sin \delta \quad (3.28)$$

For the longitudinal MOKE $r_{ps} = -r_{sp}$ (Eq. 2.29) leads to:

$$\alpha_s = \operatorname{Re} \left(\frac{r_{sp}}{r_{pp}} e^{i\delta} \right) = \operatorname{Re} (\phi_p e^{i\delta}) = -\theta_p \cos \delta - \varepsilon_p \sin \delta \quad (3.29)$$

Thus for an arbitrary retardation δ of the phase plate the output signal is a superposition of the Kerr rotation and ellipticity. The pure rotation is measured in the absence of a phase plate ($\delta = 0$) and a quarter-wave plate ($\delta = \pi/2$) is used to measure the pure ellipticity.

Chapter 4

Experimental details

The presence of adsorbates on a surface can have a variety of effects such as altering surface stress, causing surface reconstruction, quenching magnetic moments *etc.* Therefore, the presence of contaminants can significantly affect results obtained with surface-sensitive techniques such as Scanning Tunnelling Microscopy (STM), Low Energy Electron Diffraction (LEED), Auger Electron Spectroscopy (AES), SMOKE *etc.* Consequently, a high degree of surface cleanliness ($< 1 \text{ at } \%$) is required during the observation time of an experiment in order to obtain a meaningful result. For studies of transition metals such as Fe and Mo, which are highly reactive, with a particularly strong affinity for oxygen, a high degree of surface cleanliness can only be achieved for long periods under ultra-high vacuum (UHV) conditions.

4.1 UHV system

The UHV SMOKE system has been designed and developed as an annex to a larger UHV system. The whole UHV system consists of four chambers: the room-temperature STM (RTSTM) chamber, LEED and AES chamber, SMOKE chamber and a preparation chamber also used for tip and sample preparation. The layout of the system is shown in figure 4.1. Each chamber can be valved off from the rest of the system via a series of UHV gate valves (VAT [61]) and brought to atmospheric pressure for maintenance without breaking vacuum in the rest of the system. Samples and STM tips are introduced to the system via a fast-entry loadlock connected to the preparation chamber by a gate valve. When not in use, this loadlock is usually maintained in the low 10^{-8} Torr range by a 20 l s^{-1} differential ion-pump (Physical Electronics [50]). Tip and sample transfer throughout the chamber is facilitated by a series of wobblesticks and magnetically-coupled linear drives (Vacuum Generators Ltd.), which can access each sample stage. The pressure in each chamber is monitored using nude Bayard-Alpert type ionisation gauges (Anelva).

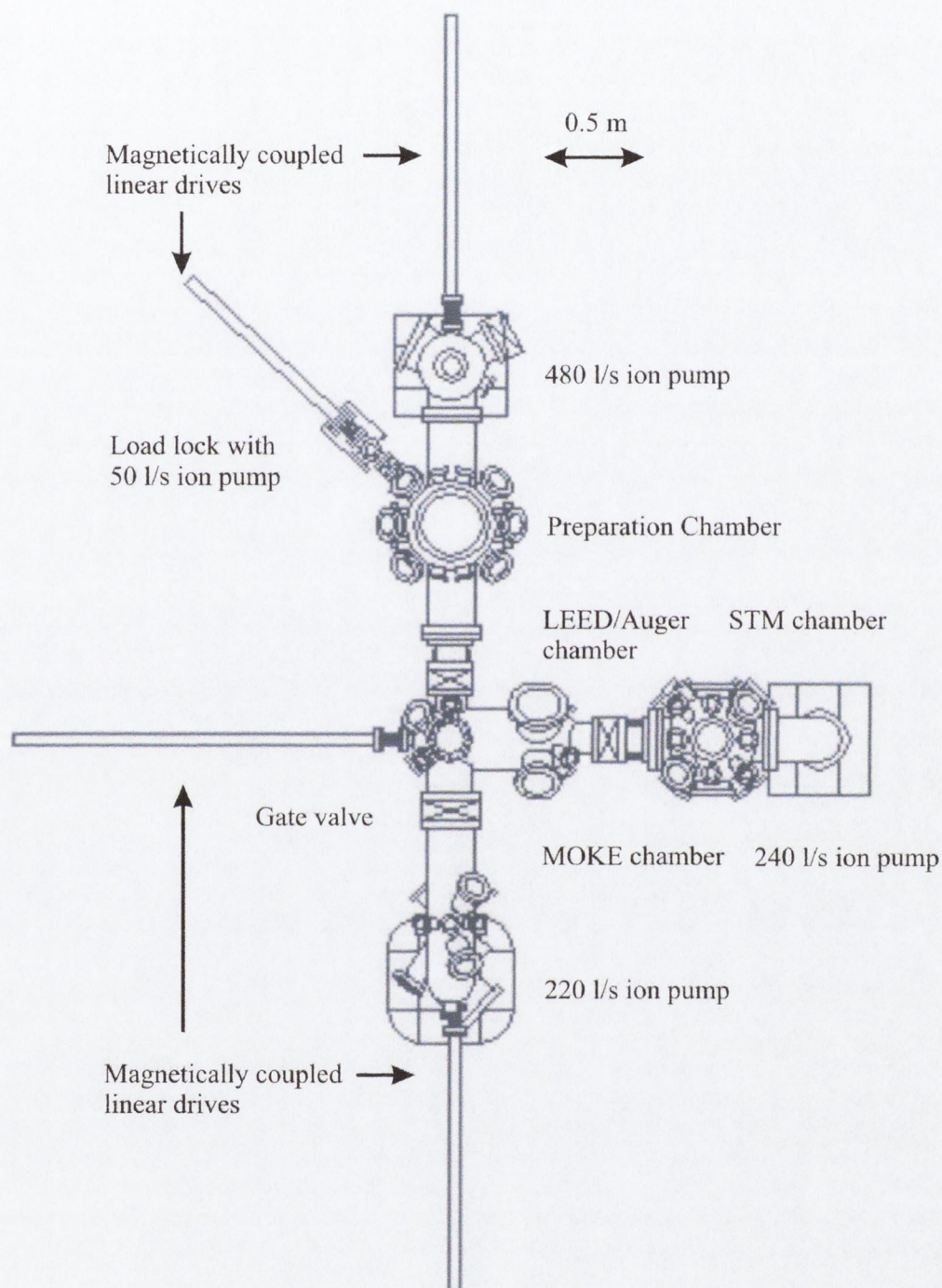


Figure 4.1 Top view schematic of the UHV system.

The entire system is supported on a stainless steel box-section frame which can be floated on pneumatic dampers to isolate the system from building vibrations during STM operation. The box-section is filled with gravel to minimise hollow pipe vibrations.

The *preparation chamber* contains all the facilities for *in-situ* tip and sample preparation: electron-beam and resistive heaters, quartz crystal deposition monitor (Inficon [51]), water cooled electron-beam evaporator (Omicron [52]), high-temperature fusion cell (MBE-Komponenten [53]), ion-gun (VG Microtech [54]) and precision leak valves for controlled introduction of high purity oxygen and argon gases.

A number of pumps are used to achieve UHV conditions in the chamber. A TMU 260 l/s turbomolecular pump (Pfeiffer Vacuum [55]) backed by a 0.7 l/s two-stage rotary vane pump is used to pump the chamber down from atmosphere and handling heavy gas loads during the bake-out, ion-etching and oxidation procedures. A double-ended 480 l/s differential ion-pump (Physical Electronics) is used to maintain UHV conditions. A liquid nitrogen cryoshroud is inserted in the base flange of the ion-pump which houses a titanium sublimation pump (TSP). The maximum pumping speed obtainable when titanium is deposited on a surface at 300 K is 3-9 l/s for hydrogen and oxygen, while it is increased by a factor of 2-5 upon cooling to 70 K [56].

To attain UHV, the preparation chamber, along with the whole system, is normally baked out for 2 days at 120-150 °C by heating tapes. The system is wrapped in aluminium foil and a heavy bake-out blanket to provide thermal insulation. A base pressure in the low 10^{-10} Torr is usually obtained in the system after bake-out.

The *Room Temperature STM (RTSTM)* chamber houses the RTSTM which is mounted inside this chamber via the O.D. 254 mm base flange. This chamber also contains a tip and sample storage carousel. The *LEED/AES chamber* to the side of the RTSTM chamber (Caburn-MDC [57]) houses the four-grid LEED/Auger optics. Both chambers are pumped from atmosphere by a 260 l/s turbo pump which is connected via a right-angle UHV valve (VG) and a 1 metre long formed bellows. The UHV is maintained by a 240

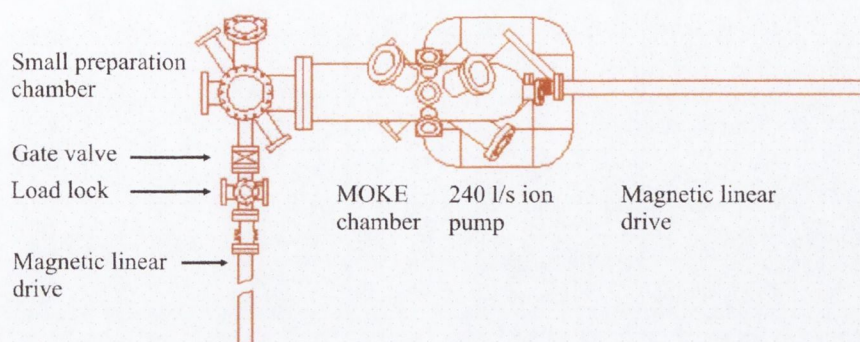


Figure 4.2 Top view schematic of the MOKE/SMOKE setup with a small preparation chamber attached to it.

l/s differential ion-pump with TSP and cryoshroud, which is connected to a mitred elbow on the opposite side of the STM chamber. This eliminates stray magnetic fields from the vicinity where LEED and AES experiments are carried out.

The *MOKE/SMOKE* chamber [58, 59] is an O.D. 156 mm $\times$ 3.0 mm stainless steel (304 grade) mitred elbow with twenty-seven access ports. The drawings of the chamber and the port listing can be found in appendix A. The chamber is pumped from atmospheric pressure by a TMU 60 l/s turbo-molecular pump (Pfeiffer Vacuum) valved directly onto the chamber via a 114 mm gate valve and backed by a 0.7 l/s two-stage rotary vane pump. The UHV is maintained by a double-ended 240 l/s differential ion-pump (Physical Electronics). A TSP SB -1000 (TLI Thermoelectronics Laboratory Inc. [60]) is mounted on a metered elbow to the bottom flange of the ion-pump.

The functioning of the MOKE/SMOKE setup had to be tested before it was joined together with the main system, shown in figure 4.1. For that purpose a *small preparation chamber* was initially connected to the MOKE chamber (figure 4.2). It contained facilities for epitaxial thin film growth: fast-entry lock, quartz-crystal deposition monitor, precision leak valve connected to a high purity oxygen source and a home-built e-beam evaporator, e-beam and resistive heaters. The fast-entry lock was a six-way cross, which was valved onto the preparation chamber via a 70 mm gate valve(VAT [61]) and connected to the turbo pump via a formed bellows.

4.2 Sample preparation

Sample cleaning, electron beam heater

As-grown 5N purity crystals of refractory metals such as W and Mo, usually contain bulk impurities (C, K, S, *etc.*) in concentrations of a few parts per million. The typical procedure for cleaning refractory metal surfaces involves annealing the crystal in an O₂ atmosphere of partial pressure $1 \times 10^{-7} \leq P_{O_2} \leq 1 \times 10^{-6}$ Torr at elevated temperatures ~ 1200 -1300 K. At these temperatures the bulk impurities diffuse to the surface where they form chemically stable oxides. These oxides, along with the surface oxide, are then removed by *flash-annealing* the surface to very high temperatures, in the range $2200 \leq T_{flash} \leq 2600$ K for short periods (5-20 seconds) under UHV conditions. These flashes are accompanied by sharp pressure bursts in the vacuum, so that some time must be allowed between flashes to allow the sample to cool and the vacuum to recover.

Conventional resistive heaters are not suitable for rapidly heating samples to such high temperatures. Instead, an electron beam heater (e-beam heater), designed by Osing [62] was used. A schematic illustration of the e-beam set-up is shown in figure 4.3. The sample is clamped in a sample holder by a Ta cap that screws onto a Mo body; the thread is deliberately machined for a loose fit. These measures are taken to prevent the two pieces of the sample holder from welding together. This holder is mounted into a Ta stage which is held at a potential of +1 kV. A current ($3 \leq I \leq 3.5$ A) is passed through a grounded thoriated tungsten filament ($\varnothing$ 0.15 mm, 0.6 % Th) which generates thermionic emission of electrons. These electrons are accelerated towards the cap of the sample holder, where upon collision, their kinetic energy is transferred to the sample as heat. Because the filament is situated to one side of the sample there will be a temperature gradient across the sample, however, this set-up produces a contamination free surface. Surface temperatures are measured from outside the chamber using an infra-red pyrometer (Iacon UX -20P [63]) and an emissivity value of $\varepsilon = 0.35$ for Mo. The absolute accuracy of these measurements is hindered by the position of the filament,

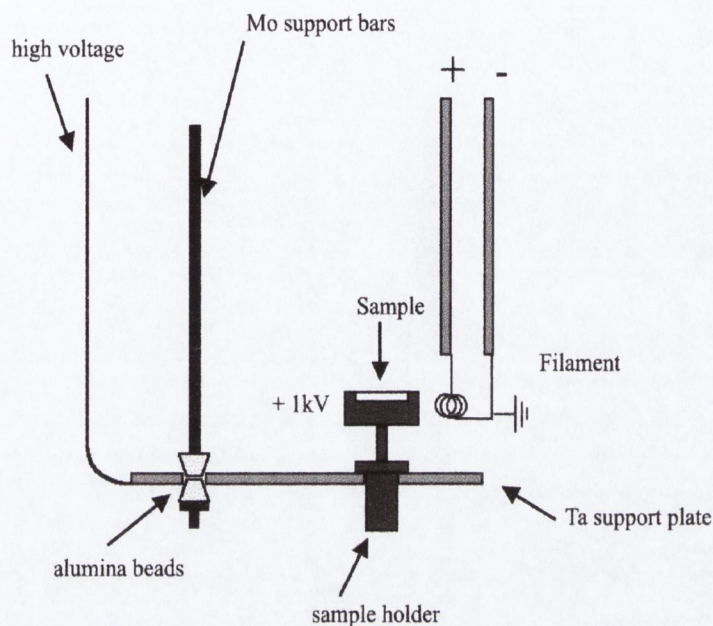


Figure 4.3 A schematic illustration of the e-beam heater used to clean refractory metal samples. Reproduced from Ref. [62].

which contributes to the level of radiation measured. However, the temperatures measured are within a similar range to the values used by other groups to obtain clean surfaces [64, 65].

Fe film deposition

The ultrathin Fe films were deposited using an electron beam evaporator shown in figure 4.4 originally designed by C. Kempf. The principle of operation is the same as that for the e-beam heater. Electrons originating from a hot thoriated tungsten filament are accelerated towards a 3N purity $\varnothing$ 1.6 mm Fe rod which is at a potential of + 1.9 kV with respect to ground. A heat-shield is formed around the Fe rod and filament by a tube of Ta foil which sits into grooves cut in the Mo end-caps of the cell. Alumina beads isolate the filament from these end-caps. Apertures in each Mo cap allow the Fe rod into the cell at one end and the evaporated Fe out through the other. The entire cell fits inside a double-

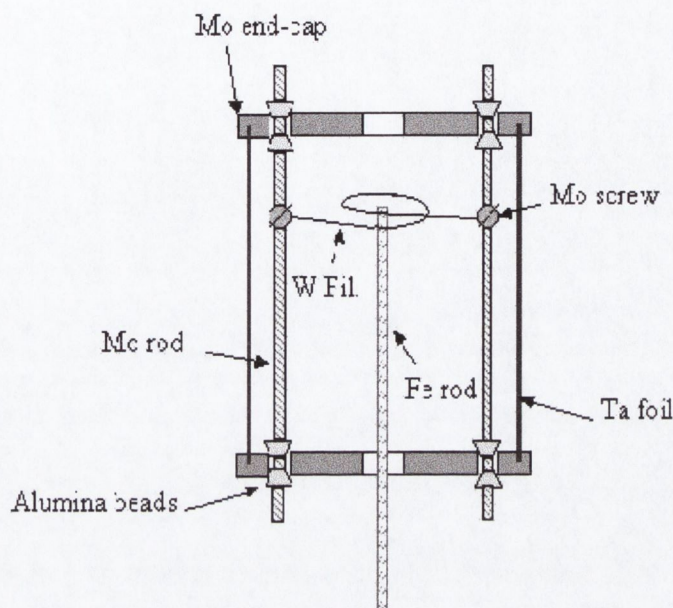


Figure 4.4 A schematic illustration showing a cross-sectional view of the e-beam evaporator used to deposit ultrathin Fe films. Electrons originating from a hot thoriated tungsten filament are accelerated towards a 3N purity Fe rod which is at a potential of + 1.9 kV with respect to the ground.

walled liquid N_2 cooled cryoshroud. During operation, cooling this cryoshroud minimizes outgassing from the environs of the evaporator. A new Fe rod is outgassed for several hours before it is suitable for use, it is also outgassed for a further 3-4 hours before each subsequent deposition. Typical values for the filament and emission currents during deposition are 2.8 A and 2 mA respectively. The chamber pressure usually rises to no more than the mid- 10^{-10} Torr.

The sample sits upside-down inside a resistive heater (described later), which has an aperture machined in its base. This set-up allows the sample surface to be heated for depositions at elevated temperatures. This heater stage is also equipped with a water-cooled quartz crystal balance and deposition monitor (Inficon). The quartz crystal balance measures the mass of iron arriving at its surface by the dampening effect of this mass on the resonant oscillations of the quartz crystal sensor. The measured mass may be converted into a film thickness value by the deposition monitor, which is programmed with values for the density of bcc Fe (7.86 g.cm^{-3}) and the exposed sensor area (diameter

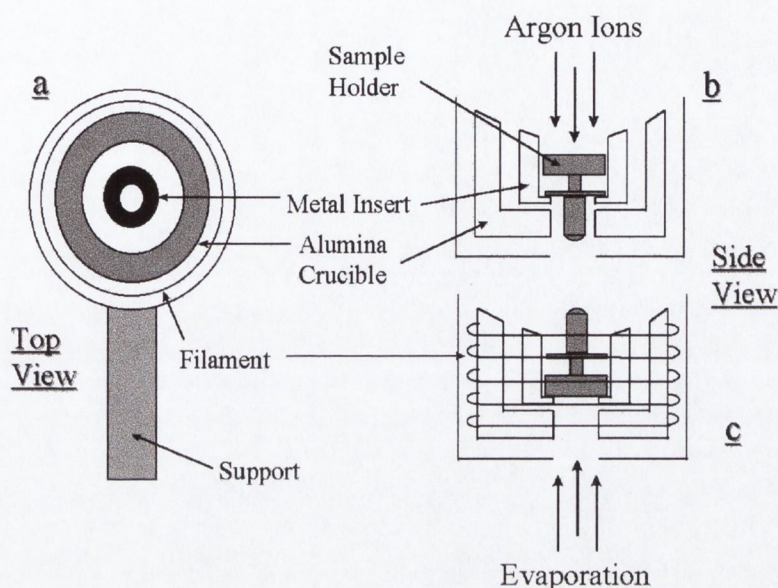


Figure 4.5 Schematic illustration of the resistive heater. The sample sits face-down in the heater so that Fe can be deposited on it through the aperture in the base.

= 8 mm). However, the thickness value determined by this method need to be taken with caution as it assumes that Fe deposited on the sensor surface has the density of bulk bcc Fe, which may not be the case.

The heating stage and crystal sensor are mounted on a linear drive so that the sample or quartz monitor can be moved into position over the evaporator aperture. During a deposition the quartz crystal is first positioned over the evaporator aperture. The filament current is slowly increased until a stable deposition rate is achieved, this is usually about 0.5 ML/min (0.015 Å/s). Then the sample, which is cleaned 20-45 minutes prior to deposition, is rapidly moved into the deposition beam - this takes roughly 10 seconds. As a result the amount of material deposited upon the surface is not exactly controlled, however, the deposition rate is typically so low that only a very small fraction of the total coverage can be deposited in this time. Once the required deposition time has elapsed the filament current and accelerating voltage are quickly ramped down.

Resistive heater

The heater, originally designed by C. Kempf and modified by author is shown in figure 4.5. It consists of a crucible which is an alumina tube with a screw thread on the outside around which a 0.2 mm W wire is wrapped. The sample sits inside a Ta insert which fits inside the crucible. The Ta insert has a concentric aperture machined in its base. The sample is inserted face-down for film depositions. A provision for placing the sample face-up for Ar⁺ ion bombardment has been also made. The crucible fits inside a stainless steel can which is connected to the feedthrough for the quartz crystal monitor. This feedthrough comprises two water cooling pipes and a BNC feedthrough to carry the quartz crystal signal to the monitor. A separate feedthrough carries the connections to the heater filament and a K-type (Omega) thermocouple connected to the Ta insert. The heater was calibrated for temperature (°C) *versus* filament power (W) with the thermocouple attached to the face of a sample-holder mounted face-down in the heater and the calibrating curve is given in appendix B.

4.3 Sample analysis

Room-temperature STM

Comprehensive reviews of the theory and operating principles of scanning tunnelling microscopy (STM) and spectroscopy (STS) are given in dedicated texts by Chen and Wiesendanger [11, 67, 68]. The fundamental theory of STM is modelled on the quantum-mechanical description of an electron with energy E travelling in a 1D potential $U(z)$ of the form shown in figure 4.6 (a). This electron is described by a wavefunction $\psi(z)$, which satisfies the Schrödinger equation:

$$-\frac{\hbar^2}{2m} \frac{d^2}{dz^2} \psi(z) + U(z)\psi(z) = E\psi(z) \quad (4.1)$$

where m is the electron mass and $\hbar = h/2\pi$ (where h is Planck's constant). In the classically allowed region where $E > U(z)$, this equation has solutions of the form:

$$\psi(z) = \psi(0)e^{\pm ikz}, \quad k = \frac{\sqrt{2m(E - U)}}{\hbar} \quad (4.2)$$

where the electron can move in either the positive or negative sense of direction. In the classically forbidden barrier region where $E < U(z)$, the Schrödinger equation has the solution:

$$\psi(z) = \psi(0)e^{-kz}, \quad k = \frac{\sqrt{2m(U - E)}}{\hbar} \quad (4.3)$$

The k term describes the decay of the electron wavefunction within the barrier region. For a finite potential $U(z)$, there is a non-zero probability P of finding the electron at a position z inside the barrier region, which is given by:

$$P(z) \propto |\psi(0)|^2 e^{-2kz} \quad (4.4)$$

Consequently, if the width of the tunnel barrier is sufficiently narrow, there is a finite probability that the electron can tunnel through the barrier region.

In the tip-vacuum-sample configuration of an STM junction (figure 4.6 (b)), the height of the tunnel barrier is determined by the work function ϕ of the tip and sample (assumed to be identical for convenience), which is the minimum energy required to remove an electron from the metal to vacuum. An electron at the tip or sample surface, with Fermi energy $E_F = -\phi$, will have the greatest probability to tunnel through the barrier, since by definition the Fermi level denotes the upper limit of electron occupancy in the metal. In the absence of an externally applied bias, the electron can tunnel through the barrier in either direction so that there is no net tunnel current. By applying an external voltage V ,

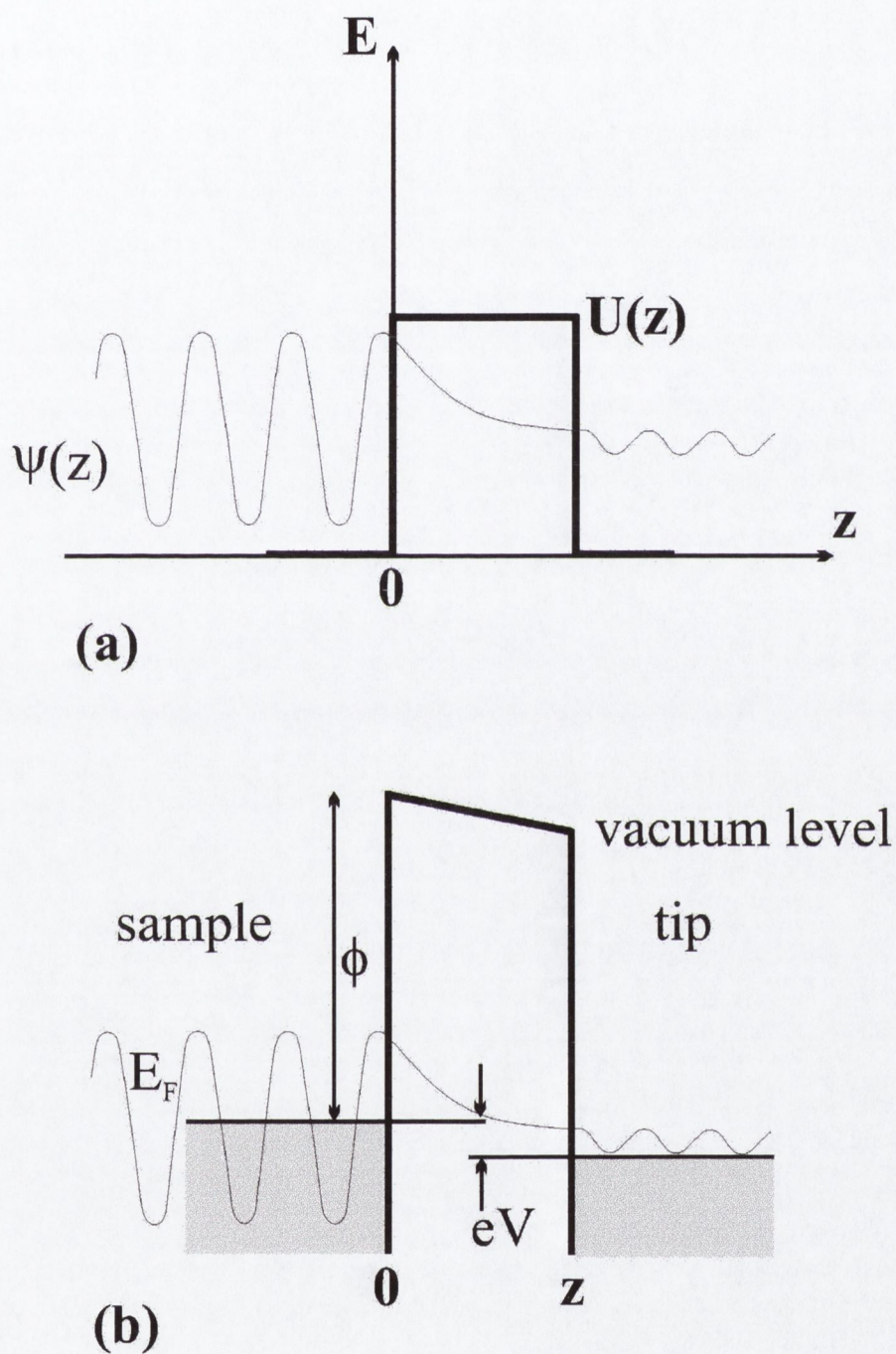


Figure 4.6 Schematic representation of a STM tunnelling junction (a) an electron described by the wavefunction $\psi(z)$ travelling in a one-dimensional potential $U(z)$. In the absence of an external bias, electrons can tunnel through the barrier in either direction so that there is no net tunnel current. (b) a STM junction under an applied bias V . By applying an external voltage V , the net electron tunnel current from the sample to the tip is established.

electrons in the sample within the energy range $E_F - eV \leq E \leq E_F$ may tunnel through the barrier. If $eV \leq \phi$, then only electron states very near to the Fermi level are probed. The probability for an electron in the n th of these states to tunnel through a barrier of width W is given by:

$$P \propto |\psi_n(0)|^2 e^{-2kW}, \quad k = \frac{\sqrt{2m\phi}}{\hbar} \quad (4.5)$$

Taking all the possible states in the energy range $E_F - eV \leq E \leq E_F$ into account, the tunnel current is:

$$I_t \propto \sum_{E=E_F-eV}^{E_F} |\psi(0)|^2 e^{-2kW} \quad (4.6)$$

This equation may be re-written in terms of the local density of states (LDOS) at the Fermi level of the sample surface $\rho(0, E_F)$:

$$I_t \propto V \rho_s(0, E_F) e^{-2kW} \quad (4.7)$$

It is clear from this equation that: (1) the tunnel current is directly proportional to the bias applied across the junction, (2) it decays exponentially as the distance between the tip apex and the sample surface is increased and (3) the local density of states of the surface is sampled.

The significance of the probe-tip LDOS contribution is realised through a time-dependent perturbation model of metal-insulator-metal tunneling proposed by Bardeen [69]. Here, a Transfer Hamiltonian H_T is used to describe the transfer of a tunneling electron from a sample state ψ to a tip state χ . The tunnel current can be taken as a convolution of the sample LDOS ρ_s and the tip LDOS ρ_t .

$$I_t = \frac{4\pi e}{\hbar} \int_0^{eV} \rho_s(E_F - eV + E) \rho_t(E_F - E) |M|^2 dE \quad (4.8)$$

I_t also includes a tunneling matrix element M , describing the amplitude of electron transfer across the tunnel barrier (through the overlap of the ψ and χ states). The integral describing M is evaluated over any surface lying within the barrier region.

$$M = \frac{\hbar}{2m} \int (\chi^* \frac{\partial \psi}{\partial z} - \psi^* \frac{\partial \chi}{\partial z}) dS \quad (4.9)$$

All STM images described in this thesis were obtained by scanning the tip across the surface in constant current mode. In this configuration the tunneling current between the tip and the sample surface is maintained at a constant value by means of a feedback-loop which controls a piezoelectric tube scanner holding the tip. The room-temperature scanning tunneling microscope (RTSTM) used in this study is similar to one designed by Quinn [66]. The STM head (Fig. 4.7) is constructed from Macor and comprises a piezo tube scanner and a fine approach walker based upon the system developed by Pan at the University of Basel, Switzerland [70]. The sample is mounted on top of a polished quasi-cylindrical sapphire rod which is clamped between a set of six piezo stacks. These piezo stacks are arranged pair-wise in a triangular fashion about the sapphire rod with one pair of stacks spring-loaded against it. The rod is moved towards or away from the tip in two steps: first the stacks shear sequentially so that the rod is clamped while each individual stack shears. Then they are allowed to relax simultaneously, dragging the rod in the direction opposite to the shear direction. A full description of this mechanism is discussed in [71, 72]. This fine approach system is controlled using a Scala control unit from Omicron.

The tube scanner is sectioned into four outer electrodes which carry the Z-X, Z+X, Z-Y and Z+Y voltages, while the inner electrode is grounded. The scanner has a dynamic range of $\pm 13000 \text{ \AA}$ in the z-direction and $\pm 20600 \text{ \AA}$ in the x- and y- directions. These ranges were calibrated on Cu(100) monatomic steps and HOPG atomic resolution images

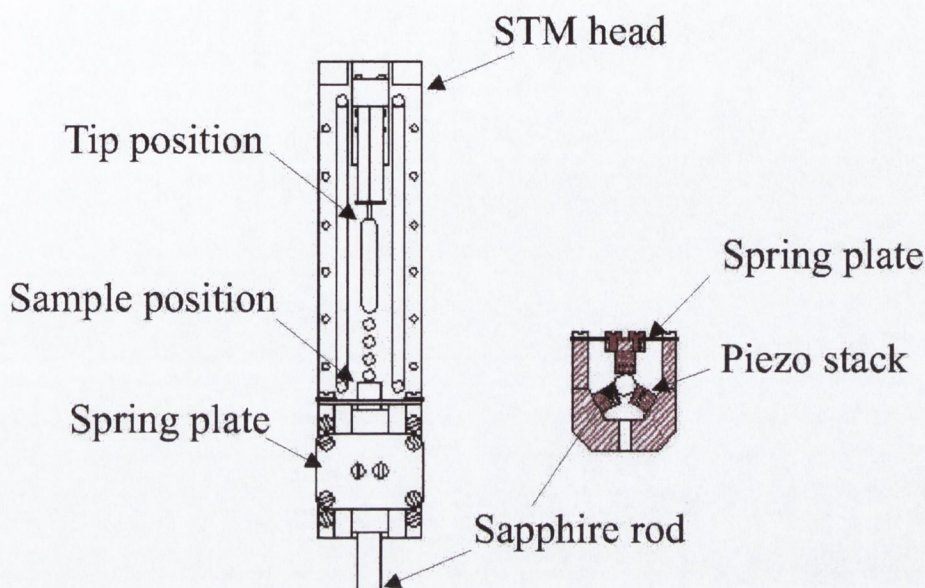


Figure 4.7 Schematic representation of a STM head and a cross-section of the fine approach walker.

respectively. The STM head is isolated from vibrations by a two-stage spring system [66] which works in conjunction with the pneumatic dampers on the system frame.

The STM controller used for these experiments is a SCALA system by Omicron GmbH [52]. The accompanying software provides some functions for data analysis, however data and commercial software from NTMDT, Russia [73] and Nanotec Electronica S.L [74] was used in the analysis.

Four-grid LEED/AES optics

The four-grid reverse view optics was manufactured by Omicron and primarily used for low-energy electron diffraction (LEED). The optics are mounted on a O.D. 200 mm CF custom elbow which is tilted at an angle of 45° to the horizontal. The optics are surrounded by a mu-metal cylinder to provide shielding from stray magnetic fields. The sample sits in the transfer fork of a grounded magnetic drive. Scattered electrons are collected by the grids and screen located behind the gun.

A schematic illustration of the four-grid LEED is shown in figure 4.8. The grid nearest the sample M1 is earthed so that electrons scattered by the sample, initially travel in field-free space. A negative potential is applied to the two centre grids M2a and M2b to suppress inelastically scattered electrons, while elastically scattered electrons are accelerated towards the phosphorescent screen by its +5 keV potential. The fourth mesh M3 is also grounded to reduce the field penetration of the suppressing grids by the screen.

LEED patterns of the refractory metal surfaces were taken at energies between 40 and 200 eV, using an emission current of 0.5 mA, the target current could not be measured. LEED patterns were recorded by photographing the screen with a digital camera. The spot-separation in these photographs could be used to calculate the surface net parameters of the surfaces analysed [66].

For surface diffraction in a 2D system, the electron beam must conserve both its energy and the component of its momentum parallel to the surface.

$$k_{\parallel}^2 + k_{\perp}^2 = k_{\parallel}'^2 + k_{\perp}'^2, \quad k_{\parallel}' = k_{\parallel} + g_{hk} \quad (4.10)$$

where $k_{\parallel}$ and $k_{\perp}$ are the parallel and perpendicular momentum components of the incident beam, while $k_{\parallel}'$ and $k_{\perp}'$ are those of the diffracted beam. The reciprocal lattice vector g_{hk} is related to the beam energy E_{eV} , electron mass m_e and diffraction angle α by:

$$|g_{hk}| = |ha^* + kb^*| = |k| \sin \alpha = \frac{\sqrt{2m_e E_{eV}}}{\hbar} \sin \alpha \quad (4.12)$$

where a^* and b^* are the reciprocal lattice primitive net vectors. These are related to the

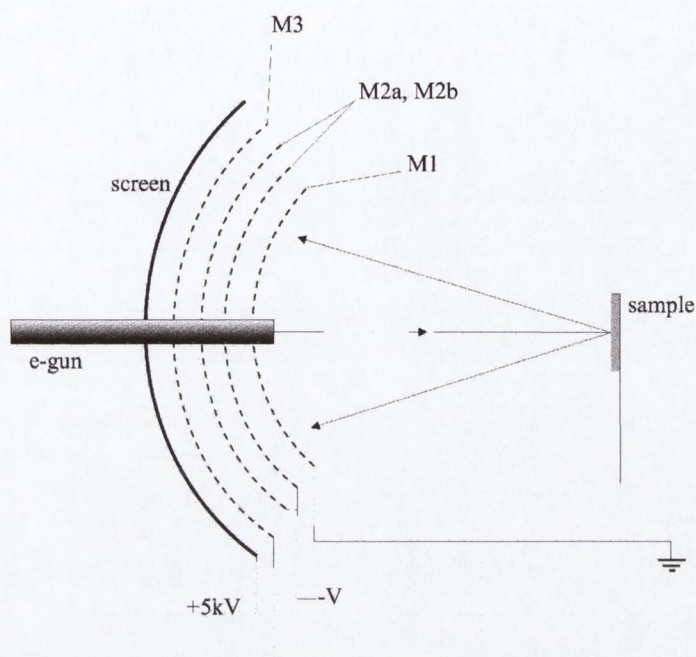


Figure 4.8 Schematic of four-grid optics operating in LEED mode.

real space vectors $\mathbf{a}$ and $\mathbf{b}$ by [12]:

$$\mathbf{a} \cdot \mathbf{a}^* = \mathbf{b} \cdot \mathbf{b}^* = 2\pi, \quad \mathbf{a} \cdot \mathbf{b}^* = \mathbf{b} \cdot \mathbf{a}^* = 0 \quad (4.13)$$

The reciprocal lattice vector $\mathbf{g}_{hk}$ lies in a direction that is orthogonal to the plane of the real space lattice that is denoted by the Miller indices h and k . The Miller indices of the diffracting planes are used to index the diffraction spots of the LEED pattern. The spacing between atomic planes with these Miller indices is:

$$d_{hk} = \frac{2\pi}{|\mathbf{g}_{hk}|} = \frac{2\pi\hbar}{\sin\alpha\sqrt{2m_e E_{eV}}} \text{ [\AA]} \quad (4.14)$$

For a centred (or primitive) rectangular lattice the spacing of atomic rows is given by [75]:

$$\frac{1}{d_{hk}^2} = \frac{h^2}{|\mathbf{a}|^2} + \frac{k^2}{|\mathbf{b}|^2} \quad (4.15)$$

The angle α may be determined from the spot-separation d_r between indexed spots in the LEED pattern by using the geometry of the LEED screen, shown in figure 4.9. The actual spot separation can be calculated from the distance d_{photo} measured in a photograph of the pattern using:

$$d_r = \frac{d_{photo} D_r}{D_{photo}} \quad (4.16)$$

where D_r is the actual screen diameter and D_{photo} is the screen diameter measured in the photograph.

For the bcc (110) surface the primitive net cell described by the primitive net vectors does not represent the full symmetry of the surface. A conventional unit cell is required, with lattice parameters a_0 and b_0 , that incorporates the full centered-rectangular structure of the surface. The distinction between the primitive and conventional cells is highlighted in figure 2.10 and is discussed by Ashcroft and Mermin [76]. By convention the diffraction patterns of bcc (110) surfaces are indexed with respect to the conventional unit cell, leading to missing (10) and (01) beams [67, 77]. Using this convention, the parameters a_0 and b_0 of the conventional surface net can be found.

Cylindrical mirror analyser for Auger electron spectroscopy (AES)

A model 10-155A Physical Electronics cylindrical mirror analyser was used for AES analysis of Mo(110) samples. The cylindrical mirror analyser (CMA) operates as an electrostatic deflection analyser. It consists of concentric inner and outer cylinders, with a coaxially mounted electron gun (figure 4.11). A primary electron beam is produced by

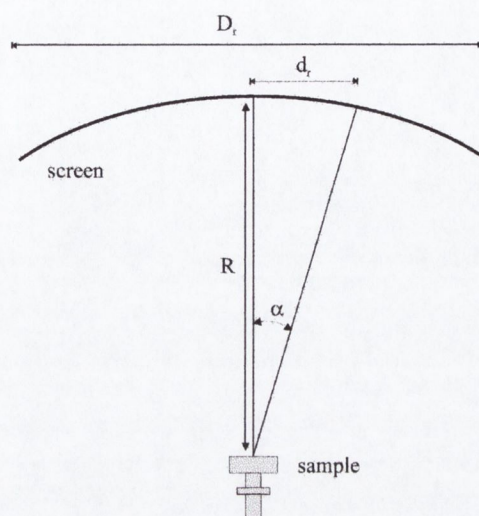


Figure 4.9 Schematic of the LEED screen geometry. The sample is placed at the focal point of the screen. The angle α can be determined from $\tan \alpha = d_r / R$. The distance d_r is the separation between the $h k$ spot and the specular spot.

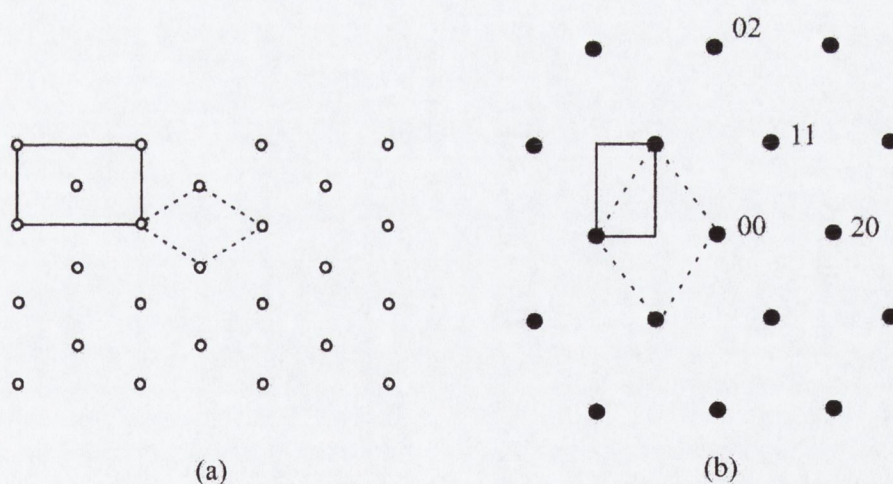


Figure 4.10 Illustration of the conventional (solid line) and primitive (dashed line) cells in (a) real space and (b) reciprocal space. The conventional unit cell highlights the centred-rectangular structure of the surface but forms a too small reciprocal lattice cell. The diffraction spots are indexed according to a convention, where the 10 and 01 spots are absent.

thermionic emission from a hot filament (F1 and F2) and acceleration through a potential V2. The beam current can be varied by changing the extraction voltage V1 to vary the level of emission. The primary beam could be focussed on the sample to produce a beam diameter of 100 μm .

Backscattered electrons enter the analyser through an annular aperture in the inner cylinder. This cylinder is grounded, while a deflecting potential is applied to the outer cylinder. The trajectory of electrons travelling in the resulting electrostatic field will be deflected by an amount which depends inversely upon their kinetic energy. Electrons with lower energies will be deflected to a greater extent than those with higher energies. The deflecting potential is chosen so that only electrons of a particular energy E_0 will have the correct trajectory to pass through the exit aperture at the opposite end of the inner cylinder. The analyser current will be given by the number of electrons $N(E_0)$ passing through the exit aperture. An electron multiplier is used to amplify the analyser current to a level which can be detected by a lock-in amplifier. Sinusoidal modulation of the deflecting potential shifts the energy window for electron transmission above and below the value specified by the dc component. The first harmonic component of this signal, which can be measured using lock-in detection, gives the differential signal $dN(E)/dE$.

Because the primary beam is generated from a hot filament source, the incident electrons will have a finite angular (and energy) spread. As a result, electrons with the same energy may be backscattered so that they enter the analyser at different angles. To avoid the resulting deterioration in resolution, the entrance aperture of the analyser is designed so that it accepts a conical annulus of electrons ($\pm 6^\circ$ [67]) about a mean angle of $\alpha = 42.3^\circ$. The geometry of the analyser is such that at this angle, electrons with the same energy but different entrance angles are focussed onto the exit aperture. This arises because electrons entering the analyser at a steeper angle must travel a larger distance in the electrostatic field and are therefore deflected through a larger extent.

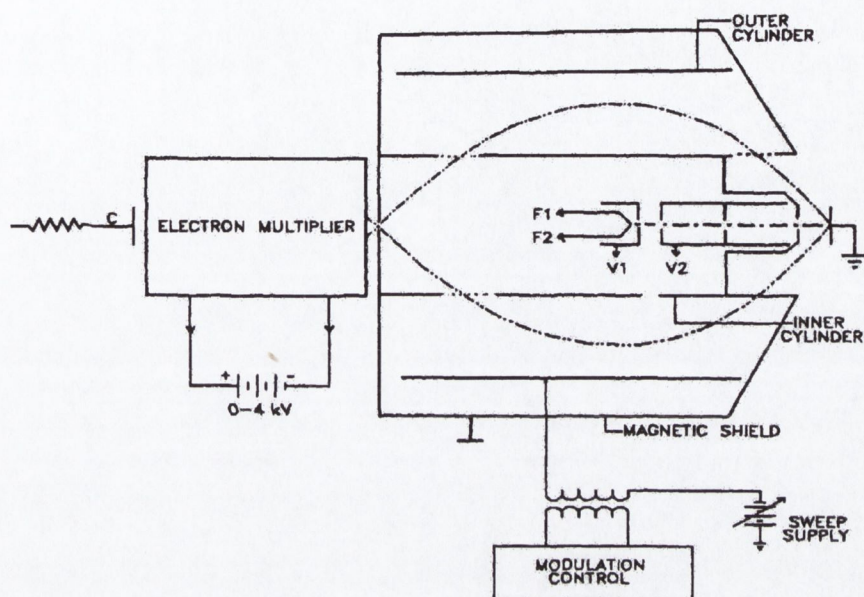


Figure 4.11 A Schematic illustration of a cylindrical mirror analyser. Reproduced from Ref. [78].

Because of this arrangement, the resolution of the CMA is fixed by the dimensions of the apertures and the mean electron path length inside the analyser, which is related to the inner cylinder radius. Since $\Delta E/E$ is constant, the transmission width ΔE widens as E increases, which means that more electrons can pass through. Moreover, the response of the analyser depends strongly on the working distance between the optics and the sample. As the sample moves closer to the entrance aperture, there is a shift in peak energies to higher values. This is simply because electrons of a given energy enter the analyser at a steeper angle and require greater deflection to reach the exit aperture. The correct working distance is obtained by measuring an elastic peak of known energy, then moving the sample and focussing the primary beam until the analyser response is optimised.

In the experimental set-up, the sample sits in a stainless steel insert, which is isolated from ground by a macor block (figure 4.13). The latter is fixed to a double-sided O.D. 70 mm CF flange by three threaded stainless steel rods (~ 300 mm long). A coaxial cable connects the steel insert with a BNC feedthrough mounted on one side of the double-

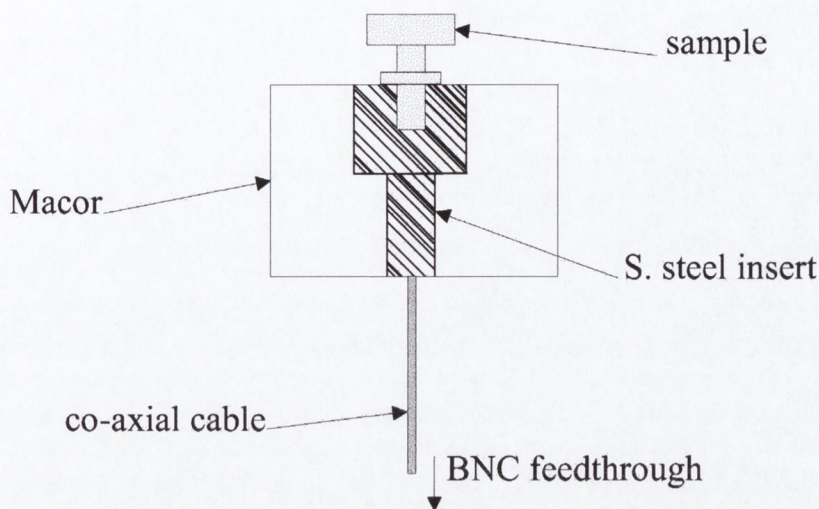


Figure 4.12 Schematic illustration of the sample stage used for AES analysis. The macor block isolates the stainless steel insert and sample from ground. A co-axial cable connects the insert to a BNC feedthrough, which can be grounded directly or through an ammeter to measure the target current.

sided flange. The whole assembly is mounted on an x-y-z translator so that the sample may be positioned directly beneath the CMA optics.

A beam energy of 3 keV was used for all measurements, the filament and emission currents were 3.2 A and 0.6 mA respectively, giving a target current of $8\text{ }\mu\text{A}$. The deflecting potential was ramped by a Perkin-Elmer 11-500A AES control unit. A SR 850 DSP lock-in amplifier from Stanford Research Systems [79] was used to output a 0.5 V_{rms} sinusoidal signal of frequency 12 kHz . This signal was fed into the AES control and acted as a reference for the modulation of the deflecting potential. A peak-to-peak modulation of 2 eV was used in all experiments. The current passing through the exit aperture of the analyser was amplified using a Channeltron model 4839 electron multiplier [80], held at an accelerating potential of $\sim 2\text{--}3\text{ kV}$ using a Perkin-Elmer 32-100 multiplier power supply. The first harmonic of the collector current was separated and converted to a voltage using the lock-in amplifier. A lock-in sensitivity of $100\text{ }\mu\text{V}$ was used to detect the Auger signal. This signal (y-axis) and a signal proportional to the dc component of the deflecting potential (x-axis) were fed through a digital multimeter to a GPIB PC interface card, where the data was acquired in *.dat format, using a programme written by D.

Kashanin in the Lab Windows CVI environment. The scan speed was always set at 1 eV/s, with scan ranges of 0-750 eV, so that a single scan takes approximately 12.5 minutes.

Atomic concentrations are calculated in the manner described by Davis *et al.* [81] and McGuire [82]. The atomic concentration C_X of element X is given by:

$$C_X = \frac{I_X / S_X d_X}{\sum_{\alpha} (I_{\alpha} / S_{\alpha} d_{\alpha})} \quad (4.17)$$

where I_X is the peak-to-peak amplitude of the largest principal Auger transition measured for the element, S_X is a relative Auger sensitivity factor, taken from reference [81]. The main Auger transition peaks for pure elemental standards are marked in spectra also given in reference [81]. The summation is over one peak per element present on the surface. The scaling factor d_X is included so that concentrations may be determined from peaks measured under different conditions, this is given by:

$$d_X = L_X E_{M,X} I_{P,X} \quad (4.18)$$

where L_X is the lock-in sensitivity, $E_{M,X}$ is the modulation energy and $I_{P,X}$ is the primary beam current. When calculating concentrations using peak-to-peak values, taken from the same spectrum, this scaling factor can be disregarded.

Chapter 5

Surface magneto-optic Kerr effect system

5.1 Electromagnet

To magnetise a sample in SMOKE experiments an electromagnet capable to produce magnetic fields of a few hundred millitesla for polar, longitudinal and transverse geometries must be integrated with a vacuum chamber. A pair of Helmholtz coils placed in UHV provides a simple and inexpensive solution [83]. A more commonly used configuration of the in-vacuum magnet is a C-shaped core manufactured from the UHV compatible soft magnetic material of low coercivity around which is wrapped a solenoid, made of polyimide-coated wire [84]. The outgassing of the wire coating is the major drawback of this design especially when several amperes current passes through the solenoid. This reduces the time for measurements to just several minutes. An in-vacuum split-coil superconducting magnets, providing maximum field of 2 Tesla are used in some systems [85].

In other experimental designs, a sample is placed between the poles of a high field water cooled external electromagnet [86]. To achieve an appreciable field in the gap the electromagnet needs to be mounted at a pinched-down section of the chamber and therefore the access to the sample is limited. An external electromagnet with the pole pieces inside the vacuum allows one to obtain a field of several hundred millitesla and avoid outgassing problems at the same time [87, 88]. In all configurations switching between polar and longitudinal geometries is done by using two pairs of crossed magnet poles [89] or by rotating the magnet or sample by 90° [90, 91].

A design with the pole tips located inside the UHV chamber and the rest of the magnet assembly outside was chosen for our setup. The cross-section of the chamber with the magnet in place is schematically shown in Fig. 5.1. This arrangement allows one to change the direction of the applied magnetic field from in-plane to out-of-plane to

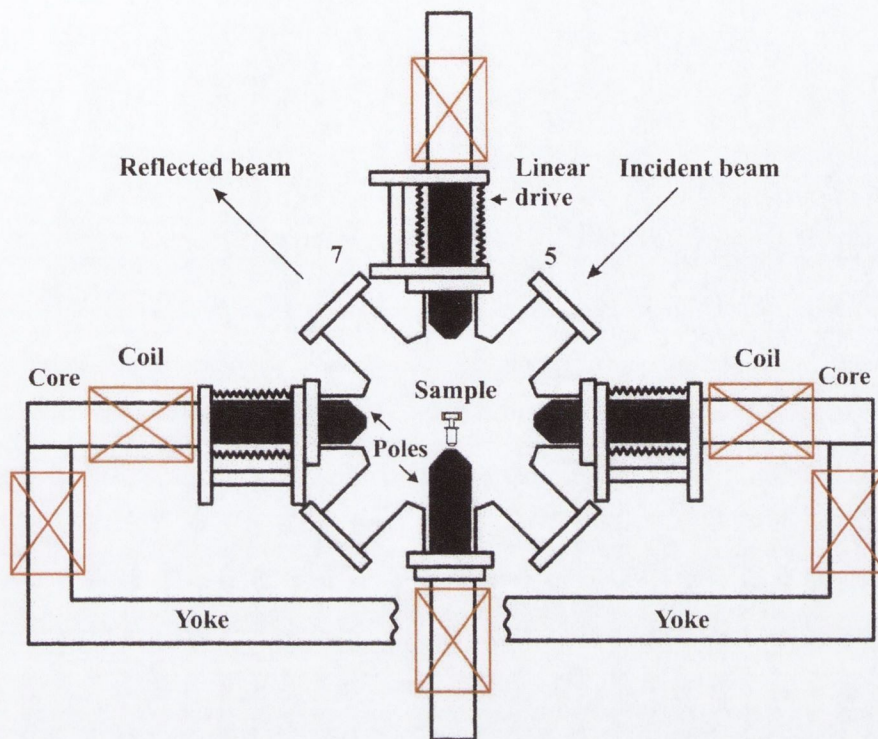


Figure 5.1 Schematic view of the SMOKE setup. Two pairs of magnetic poles are arranged in a cross-like geometry, one for polar and one for longitudinal Kerr effect measurements. The poles are located inside the UHV chamber while the rest of the magnet is outside

the longitudinal and polar Kerr measurements without displacing the sample or changing the optical path. Two pairs of magnetic poles were machined from Permenorm 5000 H2 alloy (supplied by Vacuumschmelze GmbH [92]) and arranged in a cross-like geometry. The poles were tapered to achieve higher fields and prevent blocking of the laser from the sample. They were coupled to the external core (made from low carbon steel, grade En1A) via the customized vacuum feedthroughs machined from the same alloy (figure 5.2). The feedthroughs were connected the linear translators (Vacuum Generators [93]) so that the poles could be withdrawn for the sample transfer. Thus the size of the air gap was determined only by dimensions of the sample holder and was chosen to be 14 and 18 mm for the longitudinal and the polar configurations respectively. A yoke (machined of En1A grade steel) connects the core pieces and closes the magnetic field path. Slots were milled at the ends of the core pieces so that they could slide along the yoke when the

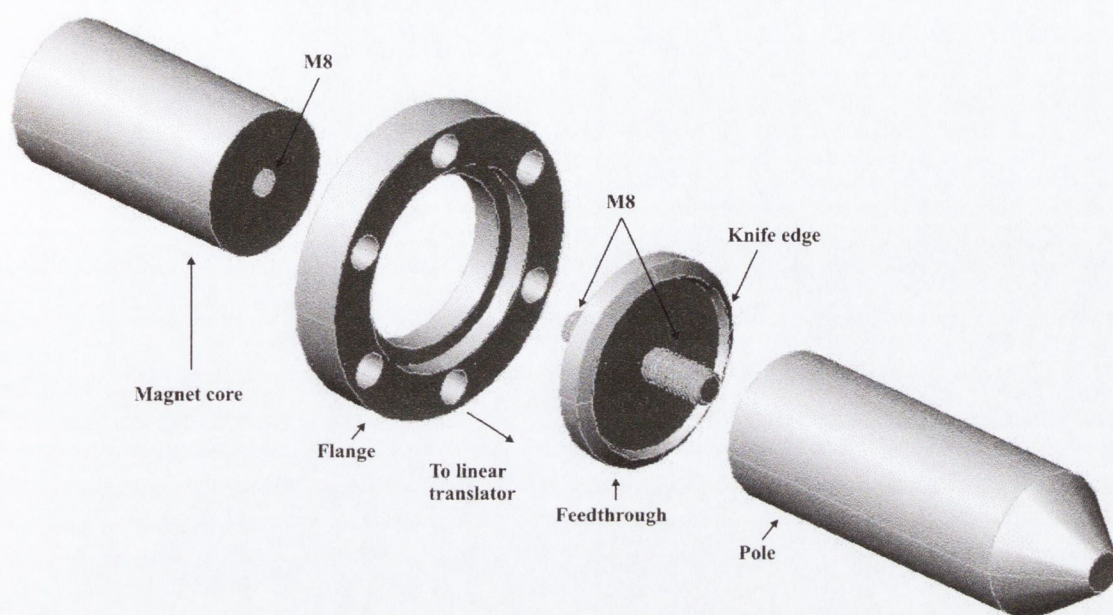


Figure 5.2 Magnetic pole assembly. The poles are coupled to the external core via the customised vacuum feedthroughs. The feedthroughs were connected to linear translators so that the poles could be withdrawn for the sample transfer.

poles were moved. The magnetic field is produced by four external coils made of 2 mm insulated copper wire. There are 400 turns on the coils seated on this core and 300 turns on the coils on the yoke sides. They can be air-cooled; however this is not necessary during routine operation.

The magnetic flux density B in the gap was estimated using a formula for a ring of magnetic material wound with a solenoid which carries current i [94]:

$$B = \frac{Ni}{\left(\frac{L}{\mu_r \mu_0} + \frac{L_a}{\mu_0} \right)} \quad (5.1)$$

where N is the number of turns in the solenoid, L is the path length in the magnetic material, L_a is the path length in the gap, μ_r is relative magnetic permeability, and $\mu_0 = 4\pi \cdot 10^{-7}$ is magnetic permeability of vacuum. The induction $B = 27 \text{ mT}$ was

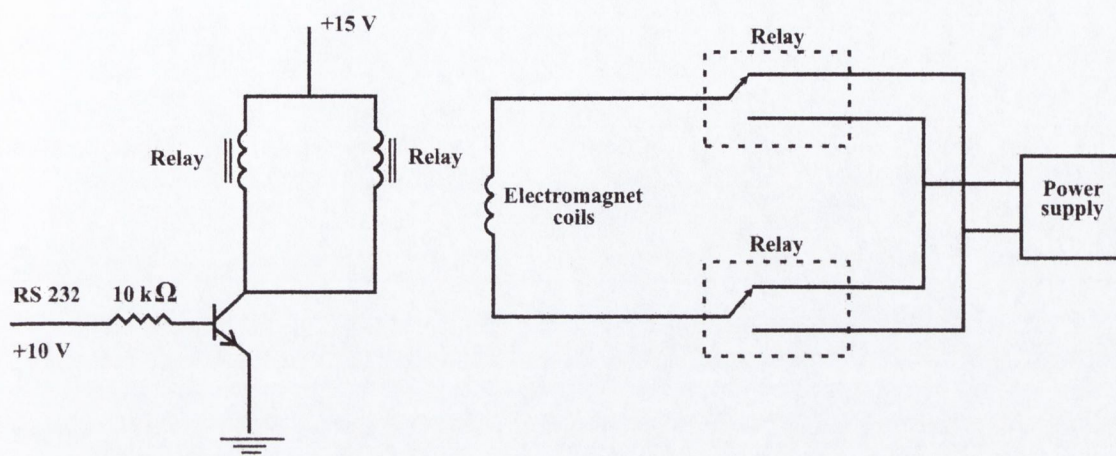


Figure 5.3 Schematic and wiring diagram of the magnetic field reversal switch.

obtained for $N = 1400$, $i = 0.2 A$, $L = 2m$, $L_a = 10 mm$, and $\mu_r = 7 \cdot 10^3$ (relative magnetic permeability of Permenorm) [92]. This simple approach gives a reasonable estimate of magnetic flux in the gap and shows that magnetic field sufficient to saturate the thin films of most magnetic materials can be obtained with relatively small current in the solenoid. However the magnetic field measured for the same current was only $\sim 20 mT$ as the true shape of the magnet, losses of magnetic energy through the stray field, smaller permeability of mild steel [96] and inhomogeneous distribution of the solenoid windings were not taken into account.

The magnetic field in the gap was calibrated *versus* current using a Hirst GM 05 Gaussmeter [95]. The calibration curves for different values of maximum current are shown in appendix B. The permeability decreases as the magnetisation approaches saturation. As a result, after $6 A$ is reached a further increase of $1 A$ adds to the magnetic field in the gap by only $10 mT$ ($100 Oe$). Hence any further increase of current is not of benefit. Therefore $6 A$ was the largest current used in our measurements, providing a magnetic field of about 220 millitesla ($2200 Oe$). The resistance of the coils connected in-series is about 3Ω and a 120 W Hewlett Packard bipolar power supply is sufficient to carry out the experiments. A switch, containing a pair of magneto-mechanical relays is inserted between the power supply and the coils. The wiring diagram and the schematic

of the switch are shown in figure 5.3. The relays are actuated by a signal from the computer RS 232 port and send the current in the opposite direction, reversing the magnetic field in the gap of the electromagnet.

5.2 Sample stage

The sample stage of the experimental setup comprises the thermal block, adapter and sample holder, machined from oxygen free high conductivity (OFHC) copper. They are connected to the cooling stage as shown in figure 5.4. The *cooling stage* is a stainless steel tube which is fixed to an O.D. 70 mm CN flange and operates as a continuous flow heat exchanger. The liquid nitrogen (LN) comes from a Dewar vessel, evaporates inside the tube and thus cools down the tube surface.

The *thermal block* is a copper cylinder, attached to the cooling stage by a thread. There is a resistive heater, wired to the block, which is made of multiple bore alumina tubes inserted into the cylinder body with a high resistivity Nichrome wire ($34 \Omega/m$ [97]) passed through them. The block can be cooled down to 90 K and heated up to 450 K. The temperature is measured with a K-type thermocouple, jammed into a copper piece which is screwed to the cylinder. The accuracy of the measurements below 300 K is $\pm 1.5 - 2$ K [97]. In order to maintain the desired temperature above the cryogenic limit but below room temperature the thermocouple and resistive heater are wired to LakeShore-331 PID temperature controller [97] via feedthroughs placed on the cooling stage flange. During the experiment the cooling power remains constant while the heater input is changed depending on the difference between the setpoint and measured temperatures.

The *adapter* is a circular plate with a threaded rod at its rim manufactured as one piece. The plate is attached to the thermal block by three stainless steel screws through the slits in the plate. The slits are machined in a pattern to allow the adjustment of the sample position in the gap of the electromagnet. The cylindrical *sample holder* is screwed on the adapter and spring loaded from inside to provide higher rigidity and better thermal contact. A molybdenum sample holder (see figure 4.3) sits in a hole in the cylinder and is

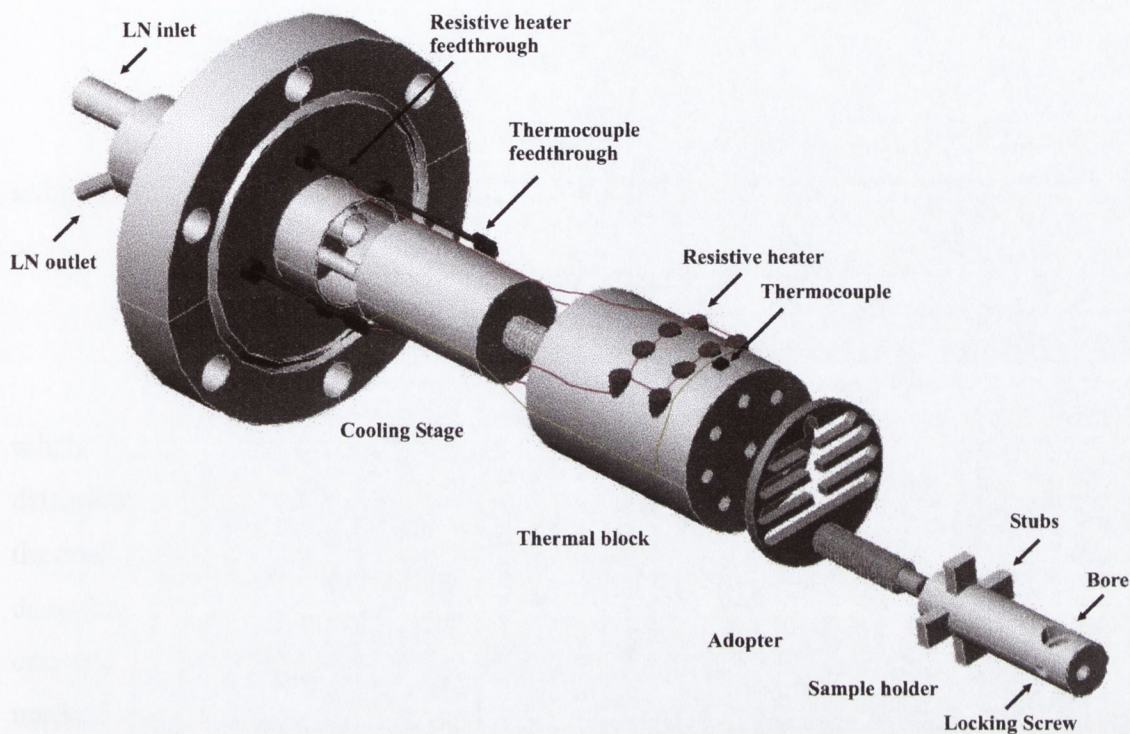


Figure 5.4 Schematic illustration of the sample stage. The sample sits in the bore of the sample holder. It is cooled down by liquid nitrogen and heated by a resistive heater. Temperatures below 290 K are maintained by a PID controller which keeps a balance between the cooling and heating power.

held in place by a locking screw. To tighten the screw the wobble stick used to transfer the sample was modified to use as screw driver. There are four stubs left on the surface of the holder so that it could be rotated about its axis by a wobble stick and a reflected beam could be positioned to the centre of the output viewport.

Below a certain thicknesses ultrathin films of ferromagnetic materials become paramagnetic at room temperature. To perform SMOKE experiments on such samples the films were cooled down below Curie temperature (T_c) and then warmed by stepping the temperature of the thermal block until T_c was reached. The hysteresis loops were measured at each step after the temperature stabilised at the desired setpoint. The time required for a sample to attain the temperature of the block was estimated by solving the heat diffusion equation:

$$\frac{\partial T(x,t)}{\partial t} = D \frac{\partial^2 T(x,t)}{\partial x^2} ; D = \frac{k}{\rho C} \quad (5.2)$$

subjected to the following initial and boundary conditions:

$$T(x,0) = T_1 ; T(L,t) = T_2 ; \frac{\partial T(x,t)}{\partial x} = 0 \text{ at } x = 0 \quad (5.3)$$

where $T(x,t)$ is the temperature which is a function of position and time, D is the thermal diffusivity ($\sim 1.1 \text{ cm}^2/\text{s}$ for Cu and $\sim 0.5 \text{ cm}^2/\text{s}$ for Mo at room temperature [98]), k is the thermal conductivity, ρ is the mass density and C is the specific heat. Equation (5.2 - 5.3) describes the heat transport in an initially thermally isolated rod at temperature T_1 , where one end $x = L$ was brought into contact with a heat bath at T_2 . The Fourier transforms method was used to solve the equation [99]. It was found that when the temperature of the thermal block is increased by 5 K the sample will reach nearly the same temperature in about 20 seconds. Therefore, 30 seconds after the temperature of the thermal block is stabilised it can be taken with confidence as the temperature of the sample.

Changes of the heat load by radiation and gas conduction due to the changes of the sample temperature were not taken into account. The convection was excluded because at room temperature a pressure of 10^{-4} Torr is already sufficient that the mean free path is $\sim 1 \text{ m}$ and the thermal conductivity becomes a function of molecule concentration. At these conditions the flow of heat transferred by gas can be estimated using the equation for two parallel surfaces at temperatures T_1 and T_2 [100]:

$$\dot{Q} = B A a_0 P (T_2 - T_1) \quad W \quad (5.4)$$

where P is pressure in the chamber in Pa, A is surface area, $0 \leq a_0 \leq 1$ is related to the individual accommodation coefficients and areas of the surfaces and for the gases usually encountered in the cryogenic design can be taken as 0.3. B has the approximate value of 4.4 for hydrogen which is the main component of the UHV residual atmosphere.

For the surface of the sample holder $A = 13 \text{ cm}^2$, $P = 10^{-8} \text{ Pa}$, $T_2 = 300 \text{ K}$ and $T_1 = 100 \text{ K}$ the heat transferred by gas in 1 minute is $2 \cdot 10^{-8} \text{ J}$, which is more than 9 orders of magnitude smaller than $Q = 50 \text{ J}$, required to increase the temperature of the sample holder by 5 K .

The radiative thermal load from the inner surface of the chamber at $T_{\text{chamber}} \sim 300 \text{ K}$ to the sample holder at $T_{\text{sample}} \sim 250 \text{ K}$ can be estimated using Stefan-Boltzmann law:

$$\dot{Q} = A F e_{\text{eff}} \sigma (T_{\text{sample}}^4 - T_{\text{chamber}}^4) ; e_{\text{eff}} = \frac{1}{(1/e_1) + (1/e_2) - 1} \quad (5.3)$$

A is the surface area of the sample holder, $\sigma = 5.67 \cdot 10^{-8} \text{ W/m}^2\text{K}$ is the Stefan-Boltzmann constant. $e_1 = e_2 \approx 0.5$ are emissivities of the chamber and sample holder, so the effective emissivity is $e_{\text{eff}} = 0.33$. This assumption allows some degree of polish on metal surfaces. The view factor F which corrects for the solid angle of the transmitting surface as seen from the receiving surface can be taken as 1 for simplicity [101]. As seen from equation (5.3) the reduction of the radiative heat flow due to the increase of the sample temperature by 5 K is about 10^{-2} W or 0.6 J/min , which can be neglected.

When the measurements are performed above the room temperature the radiative heat transfer from the sample to the chamber walls increases according to the Stefan-Boltzmann law given by Eq. (5.3). Therefore the temperature of the thermal block can not be considered as the temperature of the sample anymore. To compensate for the thermal offset a thermocouple was attached to the sample and the sample temperature was calibrated against the thermal block temperature. The offset appears at about 340 K and grows as temperature increases.

5.3 Optical setup and signal processing

The nearly crossed polarizers technique described in section 3.1 is employed in our SMOKE measurements. The optical layout of the experimental setup is shown in figure 5.5. The light source is a $685 \pm 10 \text{ nm}$ diode laser LDA-4040 [102] with a collimated

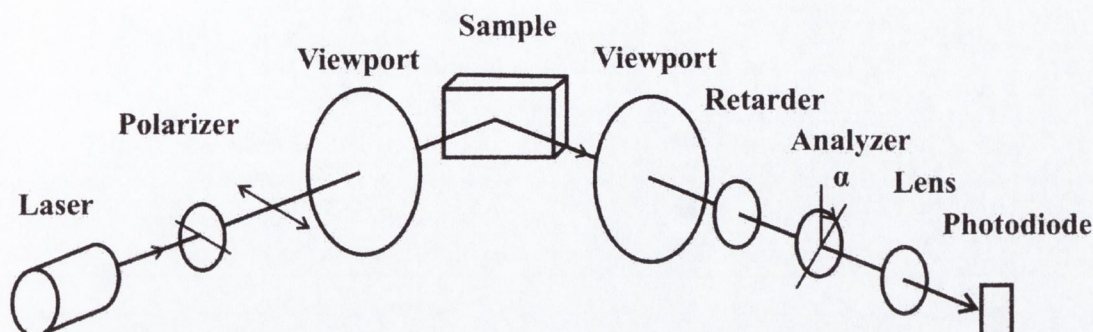


Figure 5.5 Schematic of the optical setup for the SMOKE measurements. The polarizer is used to produce a *p*-polarised light. Analyser is set at an angle $\alpha = 2 - 3^\circ$ from the extinction position. A retarder is used to compensate for the birefringence produced by the UHV windows.

beam. Dichroic sheet polarizers with the extinction ratio 10^{-4} [103] are used to define the polarisation of the incident light and as an analyser in front of the light detector. These polarizers are mounted on the rotation stages [104] to allow for the control and reproducibility of positioning. The polarizer in front of the laser is set to polarise the beam in the plane of incidence (*p*-polarised). The light intensity is detected by an IPL photodiode with an integrated amplifier [105]. It has an active area of 1.75 mm^2 making focusing of the beam difficult and increasing the noise arising from the mechanical instabilities of the setup. However when it was substituted for a photodiode of larger area the increased junction capacitance and smaller shunt resistance of the latter reduced the output signal-to-noise ratio. Therefore, a photodiode with a small active area is used and a convex lens is placed in front to focus the beam providing a noise free “optical gain”. The UHV windows introduce unwanted birefringence mostly due to the strain which is induced when they are tightened on to the chamber. The birefringence changes the polarisation of the incident light from linear to elliptical therefore the extinction cannot be realised. To remove the ellipticity a retarder is placed in front of the analysing polarizer [106]. The optical system is assembled on two mounting base plates, connected to the vertical optical rails. The adjustable height posts, translation and rotation stages from Melles Griot [107] were used individually or combined to provide positioning and alignment of optical components. All hardware elements are made from aluminium or brass to avoid their interaction with the stray magnetic field.

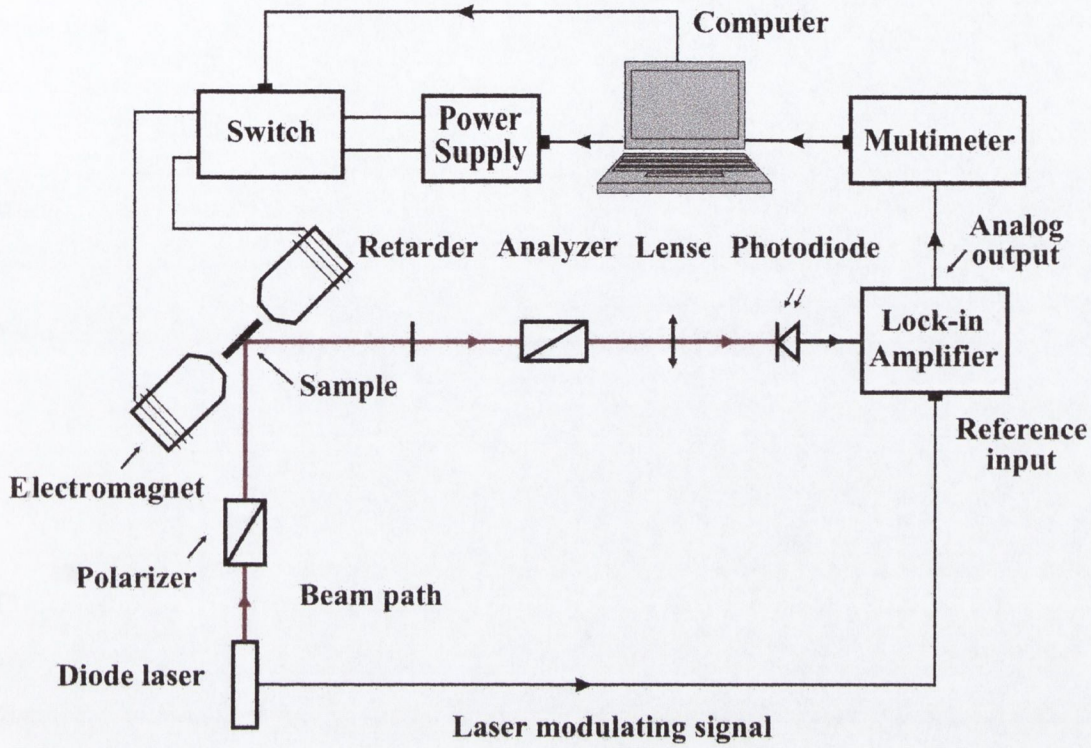


Figure 5.6 Block diagram of the SMOKE experiment. The beam path is shown by red arrows. Applied magnetic field H is swept in both positive and negative directions. The intensity of the output light changes with magnetisation and the photodiode signal plotted *versus* H traces a hysteresis loop.

An analysing polarizer is set at an angle α_m from the p -axis, where the maximum contrast is achieved. It cannot be set to the full extinction since in this case the rotation of the polarisation plane in the opposite directions will result in identical transmitted intensities. Besides in this case the detector signal will be proportional to the square of magnetisation. Under the assumption that the Kerr effect is real (no ellipticity) the contrast can be found from [108]:

$$C \equiv \frac{\Delta I}{I} = \frac{\sin^2(\alpha + \theta) - \sin^2 \alpha}{\sin^2 \alpha + \delta} \approx \frac{2\alpha\theta + \theta^2}{\alpha^2 + \delta} \quad (5.5)$$

where ΔI is the change in the light intensity produced by the Kerr rotation θ and I is

the transmitted intensity resulting from the depolarising effects and the analyser offset from the position of minimum transition. $\delta = I_{\min} / I_{\max}$ is the extinction ratio of the system where $I_{\min}$ and $I_{\max}$ are the intensities detected by the photodiode when the analysing polarizer is set to the positions of minimum and maximum transitions respectively. In the case of practical interest $4\delta \gg \theta^2$ the maximum contrast is obtained when the polarizer is set at $\alpha_m \approx \sqrt{\delta}$ and the equation (5.5) becomes [109]:

$$C = \frac{2(\alpha / \alpha_m)}{1 + (\alpha / \alpha_m)} \frac{\theta}{\sqrt{\delta}} \quad (5.6)$$

The equation (5.6) shows that the extinction ratio determines the performance of the optical system. It was found for our setup that the maximum contrast can be achieved at $\alpha = 2 - 3^\circ$. It still can be significantly improved by replacing the sheet polarizers mentioned before with the Glan-Thompson polarizers, with extinction ratio equals to 10^{-5} .

The block diagram of the SMOKE experiment is shown in figure 5.6. To eliminate the stray light from other sources and enhance the signal-to-noise ratio the laser intensity is internally modulated at the frequency of about 1.5 kHz. This modulating signal is provided to the reference input of the lock-in amplifier (model 5210 by EG&G [110]). The photodetector output signal is applied to the input of the lock-in amplifier, while the output is connected to a Hewlett Packard digital multimeter. A Hewlett Packard power supply is used to energise the electromagnet coils. A computer reads the data from the multimeter and controls the power supply via a GPIB interface. The field reversing switch is actuated via a RS-232 serial port. A Lab Windows CVI programme is used to step the current and record the photodiode signal. To speed up the measurements the current step can be set smaller in the region of magnetic reversal and larger at the ends of the hysteresis loop. It takes from 10 to 60 seconds to record a single loop. Prior to the measurements the magnetic field was repeatedly reversed to cycle the film and the

electromagnet around the hysteresis loop. After several reversals the tips of the loop become reproducible.

The photodiode signal *versus* current is acquired in *.dat format and the data is processed using programs written in Matlab. So as to improve the signal-to-noise ratio the hysteresis loop is swept multiple times and then averaged. The results of calibration shown in figure B2 of appendix B are used to convert current I into magnetic field H . The branches 1-2, 2-3 of the calibrating curves (figure B2(c)) were fit to an exponential associate function:

$$H = H_0 + H_1(1 - e^{-I/t_1}) + H_2(1 - e^{-I/t_2}) \quad (5.7)$$

and 3-4, 4-1 to an exponential growth function:

$$H = H_0 + H_1 e^{I/t_1} \quad (5.8)$$

where H_0 , H_1 , H_2 , t_1 and t_2 are the fitting parameters. Then the obtained Kerr intensity *versus* H loops were scaled to bring the positive and negative states of saturation magnetisation M_s to arbitrary intensities of 1 and 0 respectively.

Sometimes variations in the ambient temperature occur which change the laser light intensity and causing a distortion of the hysteresis loops. Figure 5.7a shows six successive easy axis hysteresis loops, obtained with the longitudinal SMOKE from a 3.5 monolayer iron film, deposited on a molybdenum (110) surface. The traces are shifted one to another, due to the thermal drift and their averaging results in a distorted loop displayed in figure 5.7b. This thermal drift can not be eliminated by the lock-in detection technique; however when the scan time is less than the drift fluctuation time the drift approximates reasonably well to a constant ramp over the scan period and can be subtracted by a computer from the recorded curves. The overlay for the same loops after the drift correction and the result of their averaging are displayed in figures 5.7c and 5.7d, respectively.

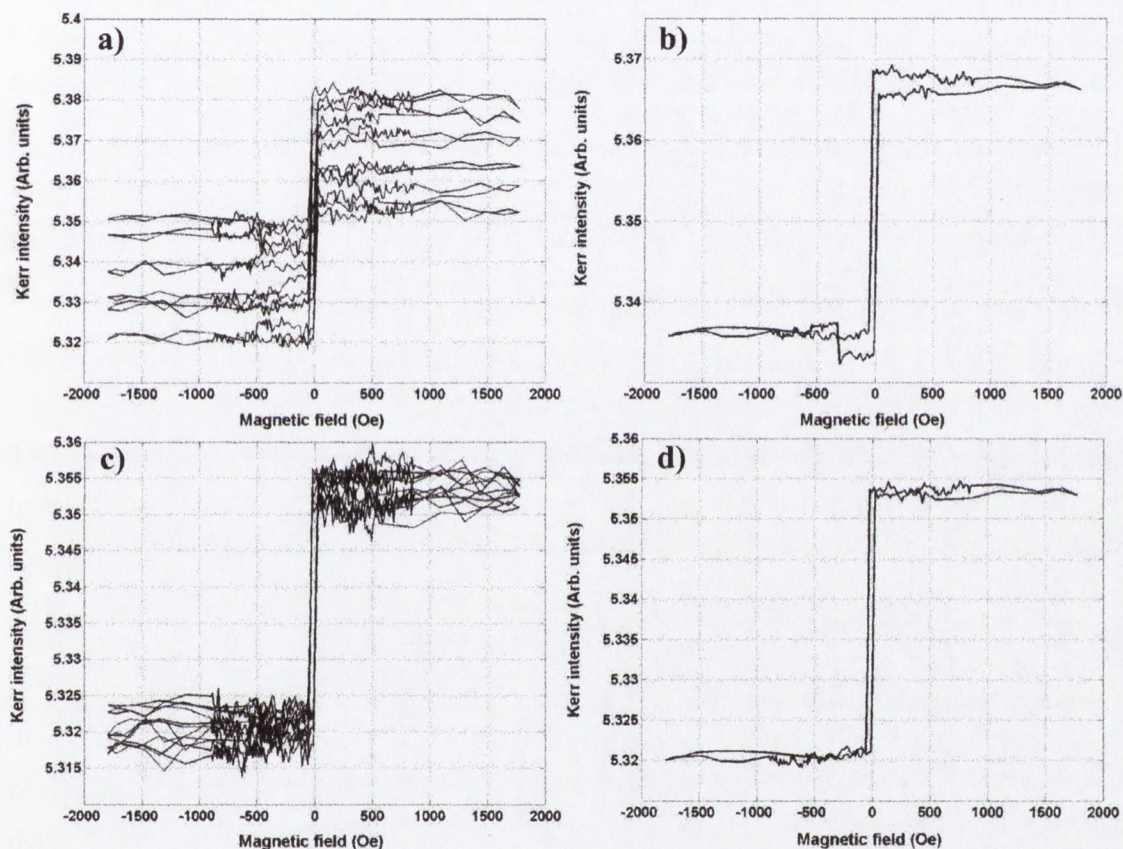


Figure 5.7 Easy axis longitudinal hysteresis loops for a 3.5 ML Fe/Mo(110) film; a) six loops shifted dew to the laser thermal drift; b) distorted hysteresis loop, obtained by multiple averaging of the traces displayed in figure 5.7a; c) the result of applying the slopping baseline drift correction procedure to the curves in figure 5.7a ; d) average of the curves in figure 5.7c.

The Voigt effect on the UHV windows is another unsought phenomenon impeding the SMOKE measurements (figure 5.8). If the effect is described in the context of the macroscopic theory used in chapter 2 then its contribution to the dielectric tensor will be given by the third term of equation (2.4). In simple terms, the Voigt effect consists in changing the light's ellipticity and rotating its plane of polarisation as it propagates through a medium placed in a magnetic field orthogonal to the wave vector [111]. If the medium is isotropic then the eigenmodes are the fields linearly polarised along and perpendicular to the magnetic field. Due to the asymmetry between the two eigenmodes the medium acquires linear birefringence and dichroism. Since changing the sign of the magnetic field does not change this asymmetry between the eigenmodes, there are no linear magneto-optical effects and the lowest order term of Voigt effect is quadratic in the applied magnetic field.

The graph shown in figure 5.8a was recorded using the settings for longitudinal MOKE measurements described above with a beam reflecting from a bulk Mo polycrystalline sample. Mo is a paramagnetic material with magnetic susceptibility $\chi = 1.19 \cdot 10^{-4}$ [112] therefore no changes in the light intensity should be observed in the available range of magnetic field. However it is clear from the figures 5.8a and 5.8c that there are changes in the output signal proportional to H^2 which are of the same magnitude as Kerr intensity obtained from the ultrathin Fe films. Later it was found that magnetisation curves distorted by Voigt effect on the UHV windows had been presented in a number of SMOKE experiments in which the magnetic field of several thousand Oe was applied along the hard magnetisation axis of the samples with the large magnetic anisotropy [113, 114]. Mild steel tubes slotted over the view ports were used to shield the glass from the stray magnetic field produced by the electromagnet. The curve shown in figure 5.8b was obtained from the same polycrystalline Mo sample with the magnetic shields in place which clearly shows that the shielding eliminates this unwanted magneto-optical activity.

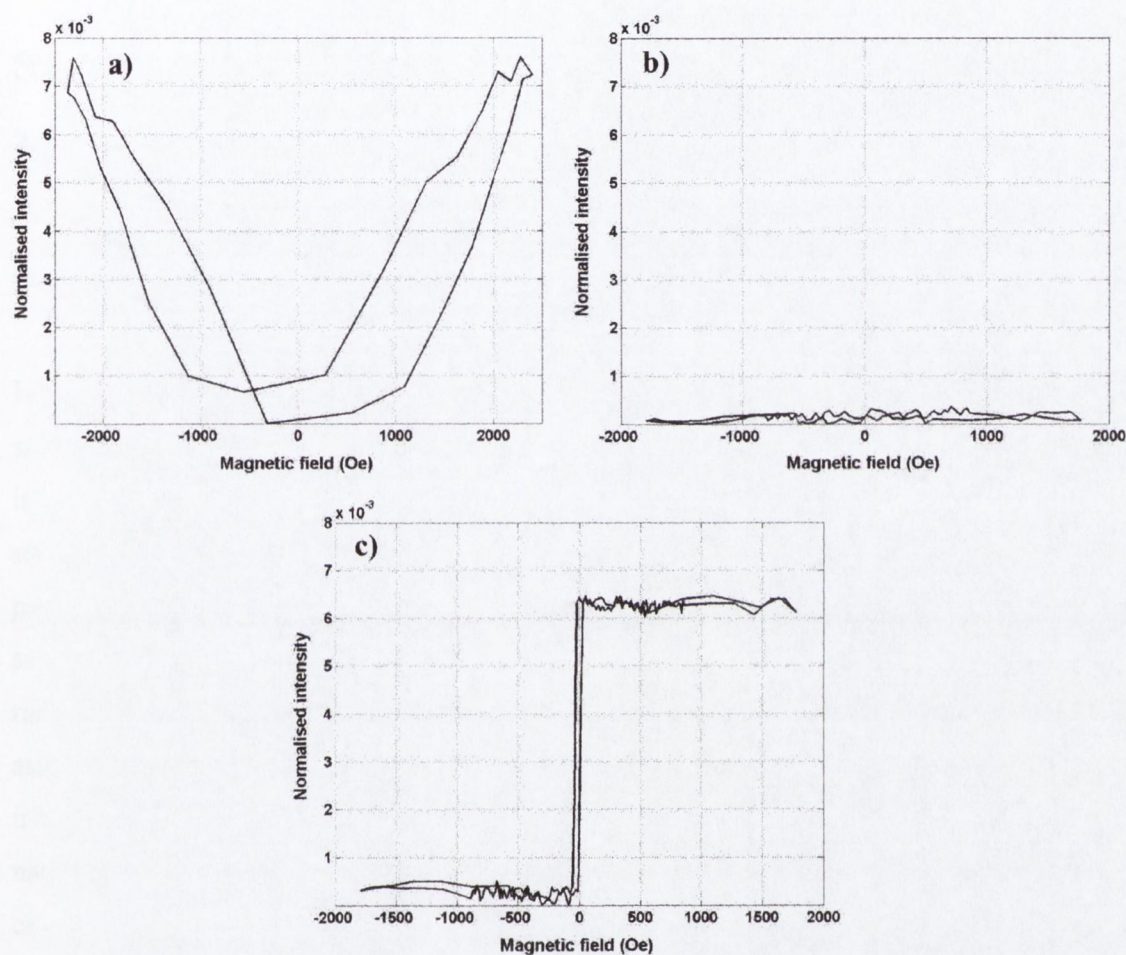


Figure 5.8 Intensity of light transmitted through the analyser (normalised to the intensity at $H = 0$) versus longitudinal magnetic field; a) the sample is a disk made of polycrystalline molybdenum. The changes in the output intensity proportional to H^2 result from the Voigt effect on the UHV windows; b) same as a) but with the magnetically shielded viewports; c) easy axis MOKE loop produced by a 6 ML Fe/Mo(110) film with the magnetically shielded viewports.

Chapter 6

Study of magnetic anisotropy of ultrathin Fe films on Mo(110) surface

6.1. Magnetic anisotropy

Ultrathin hetero-epitaxial films of magnetic 3d transition metals have stimulated much scientific interest as the reduced film thickness and the presence of a heterogeneous interface can substantially modify magnetic properties. A main factor that affects the shape of the hysteresis loop is magnetic anisotropy. The anisotropy defines the preferential (easy), intermediate and hard directions of magnetisation. The two main sources of the magnetic anisotropy are dipolar and spin-orbit interactions. Due to its long range, the dipolar interaction generally results in a *shape – dependent* contribution to the anisotropy (shape or dipolar anisotropy), which is largely responsible for the in plane magnetisation usually observed. With the spin-orbit interaction a small orbital momentum is induced which couples the total (spin plus orbital) magnetic moment to the crystal axes. As result, the total energy is minimised when spontaneous magnetisation is oriented along certain crystallographic directions (*easy axes*) as compared to those less preferred (*hard axes*). This dependence of total energy on the orientation of the magnetisation relative to the crystalline axes reflects the symmetry of the crystal. This is known as the *magnetocrystalline contribution* to the anisotropy. The lowered symmetry at the surface strongly modifies this contribution as compared to the bulk, yielding a so-called *surface anisotropy* as pointed out by Neel. The spin-orbit interaction is also responsible for the *magnetoelastic anisotropy*, induced in the system when it is strained, a situation which is frequently encountered in thin films due to the lattice mismatch between the constituent film and substrate.

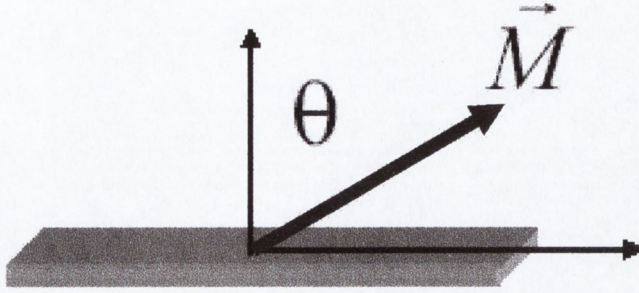


Figure 6.1 Definition of an angle θ between magnetisation and the film normal.

The shape effect of the dipolar interaction in ferromagnets is described via an anisotropic demagnetising field $\mathbf{H}_d = -N_d \mathbf{M}$. Here N_d is a shape-dependant demagnetising tensor which elements depend on the shape of a sample. For a thin film in the shape of a disk as is used in these experiments, all tensor elements are nearly zero except for the direction perpendicular to the layer: $N_d \approx 1$. The magnetostatic energy per unit volume can be expressed as:

$$E_d = -\frac{\mu_0}{2V} \int_V \mathbf{M} \cdot \mathbf{H}_d dV \quad (6.1)$$

where V is the volume of the sample. For a thin film, it results in an anisotropy energy contribution [115]:

$$E_d = -\frac{1}{2} \mu_0 M_s^2 \sin^2 \theta \quad (6.2)$$

where M_s is the saturation magnetisation, μ_0 is the magnetic permeability of vacuum and θ is an angle between the film magnetisation vector $\mathbf{M}_s$ and the surface normal (usually z -direction) as shown in Fig. 6.1. According to this expression, this contribution favours in-plane orientation of magnetisation.

The *magnetocrystalline anisotropy* is closely related to the spin-orbit interaction and results from the action of the crystalline field, produced by surrounding atoms or ions in the solid, on the considered atom or ion. This field has the symmetry of the crystal and the magnetic electrons of the atom involved are coupled to the lattice axes. Thus, the magnetisation process is different along different crystallographic directions. A Russian physicist Akulov showed that the crystal anisotropy energy can be expressed in terms of a series expansion of the direction cosines of the saturation magnetisation vector $\mathbf{M}_s$ relative to the crystal axes. In a *cubic crystal*, this expansion can be written as:

$$E = K_0 + K_1(\alpha_1^2\alpha_2^2 + \alpha_2^2\alpha_3^2 + \alpha_3^2\alpha_1^2) + K_2(\alpha_1^2\alpha_2^2\alpha_3^2) + \dots \quad (6.3)$$

where α_i are the direction cosines of the $\mathbf{M}_s$ vector. The odd power coefficients vanish due to the inversion symmetry of a cubic system. The second order coefficient has a factor $(\alpha_1^2 + \alpha_2^2 + \alpha_3^2) = 1$ and so does not contribute to the anisotropy. The fourth and sixth order coefficients K_1 and K_2 are called the first and the second order anisotropy constants. They determine the strength of anisotropy and have the units $J\cdot m^{-3}$. K_1 gives the magnitude of four-fold (biaxial) symmetry while K_2 is related to the six-fold symmetry.

A uniaxial (two-fold) symmetry is characterised by a unique easy axis. For example in cobalt, which has a hexagonal close-packed structure, a hexagonal c -axis is the direction of easy magnetisation and any direction in the basal plane is equally hard. In this case, the anisotropy energy depends only on a single angle θ between the $\mathbf{M}_s$ vector and the c -axis. Therefore, for a uniaxial anisotropy equation 6.3 can be rewritten as:

$$E = K_0 + K_1 \sin^2 \theta + K_2 \sin^4 \theta \quad (6.4)$$

Induced anisotropy arises when the easy axis of magnetisation is induced by creation of some atomic scale texture during the film growth or annealing, or by applied stress (*magnetoelastic anisotropy*). The strain in a ferromagnet changes the magnetocrystalline anisotropy. It is of particular importance for the cubic magnets where the uniaxial stress

gives rise to uniaxial anisotropy contributions. For a uniaxially strained elastically isotropic medium the energy per unit volume, associated with this effect can be written as:

$$E_{me} = -\frac{3}{2}\lambda_m\sigma\cos^2\theta \quad (6.5)$$

where σ is the stress, which is related to the strain ε via the elastic modulus E_e by $\sigma = \varepsilon E_e$. The magnetostriction constant λ_m , having the physical meaning of the spontaneous magnetostriction in the magnetisation direction, can be either positive or negative. θ is the angle between the magnetisation and the strain axis.

Phenomenologically the anisotropy is described in terms of volume and surface contributions. The constant term K_0 in equation (6.4) has no bearing on the equilibrium conditions and can be omitted. A second order term $K_2 \sin^4(\theta)$ is needed sometimes, but usually is much smaller than the first term. Therefore, a uniaxial description is widely used to account for experimental observations:

$$E = K_1 \sin^2\theta \quad (6.6)$$

where θ denotes the angle between the magnetisation and the film normal (Fig. 6.1). A positive K_1 describes the case of a preferred direction of magnetisation perpendicular to the film plane. The anisotropy energy K_1 includes all contributions from the various sources and sometimes it is referred to as the effective anisotropy K^{eff} .

One can distinguish between the contributions from the *surface* or interface and from the bulk, which are given by the anisotropy constants K_S ($J\cdot m^{-2}$) and K_V ($J\cdot m^{-3}$) respectively.

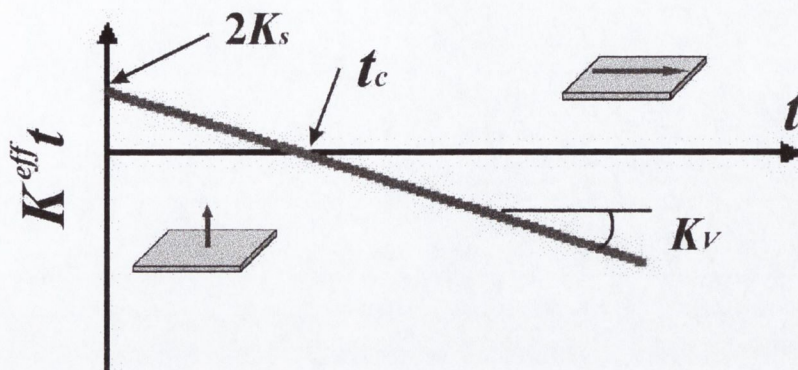


Figure 6.2. Schematic plot of $K^{\text{eff}} t$ versus t . Perpendicular magnetisation is established for a thickness smaller than the critical thickness t_c . The y -axis intercept equals twice the surface energy. The slope gives the volume anisotropy constant.

In this case, the average anisotropy of a magnetic layer of thickness t is given by

$$K^{\text{eff}} = K_v + 2K_s / t \quad (6.7)$$

K_s and K_v can be obtained by plotting the product $K^{\text{eff}} t$ versus film thickness t as shown in figure 6.2. The factor 2 appears because the film has two surfaces: film/vacuum and film/substrate. At some critical thickness $t_c = -K_s / K_v$ ($K^{\text{eff}} = 0$) the surface contribution to anisotropy overcomes the magnetostatic energy and the easy axis of magnetisation changes its direction from in-plane to out-of-plane [116].

Magnetic *surface anisotropy* originates from reduced local symmetries. Neel was first to recognise in his phenomenological pair-bonding model that the loss of the nearest neighbours in magnetic surfaces and interfaces gives rise to anisotropies different from the bulk [117]. According to Neel, in a bulk bcc (cubic body centred) lattice (inherent to Fe) the interactions from 8 nearest neighbours sum up to a constant contribution to the energy. In creating a (110) surface two nearest neighbour bonds of the central atom (grey in Fig. 6.3) to the atoms above the surface are removed. Atomic steps, which are always present at the surface of an oriented single crystal, break the rotational symmetry of the

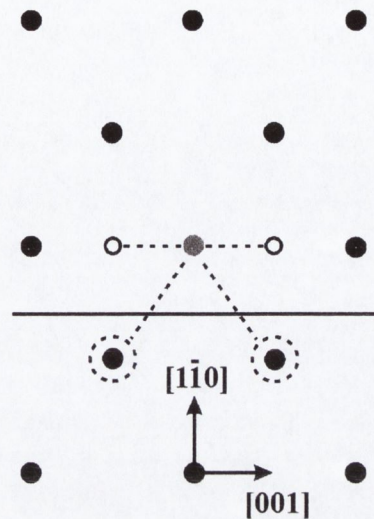


Figure 6.3 Top view of a bcc (110) surface. Full circles represent the atoms lying in (110) plane; open circles correspond to the empty atomic positions above the surface. The solid line shows a step along the [001] crystallographic direction and the dashed lines indicate the missing bonds to the removed atoms.

surface in the directions parallel to the surface plane. For example in figure 6.3 a step running along (001) direction is created by removing the bonds to the circled atoms and reducing the coordination number of the central atom sitting at the step edge from 8 to 4. Magnetic moments at the step-edge and corner sites tend to align either parallel or perpendicular to the step orientation and produce additional uniaxial *step-induced* anisotropy within the film plane. Allowing for the step-induced anisotropy the equation (6.7) for the anisotropy energy can be written as [118]:

$$K^{eff} = K_v + 2 \frac{K_s}{t} + \frac{K_{sl}}{td} \quad (6.8)$$

where K_{sl} is the anisotropy constant of the spin-induced anisotropy in $J \cdot m^{-1}$ and d is the atomic terrace width. The anisotropy effects from the surface and the steps become less important with the increasing film thickness. Similarly, in the systems with wider terraces the step effects are not significant.

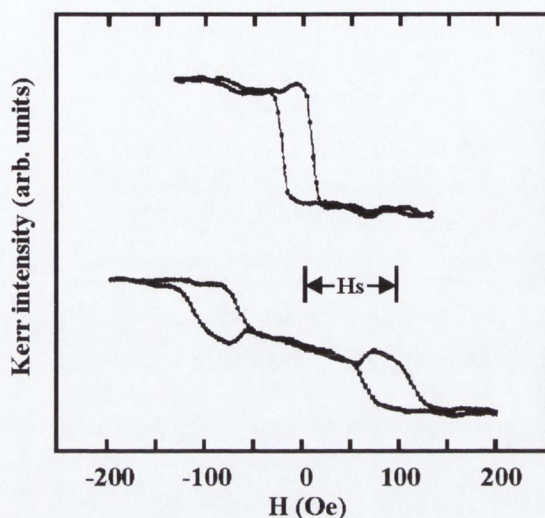


Figure 6.4. Hysteresis loops for a 2 ML Fe film grown on a 4.7° miscut W(001) surface (Ref. [120]). The square loop is for magnetic field (H) perpendicular to the step edges. The split loop obtained for H parallel to the step edges indicates the presence of the step induced anisotropy with the easy axis perpendicular to the steps. The shift field H_s is directly proportional to the step-induced anisotropy constant.

A uniaxial step-induced anisotropy has been observed in several magnetic thin films grown on stepped substrates, such as Co/Cu(100) [119], Fe/W(100) [120] and Fe/Ag (100) [121]. All these systems had a four-fold in plane symmetry of a bcc or fcc (100) surface broken by the steps along the [100] crystallographic direction. The split Kerr loops with zero magnetic remanence (Fig. 6.4) were obtained when the in-plane magnetic field was applied along the steps for Fe/W(100) or perpendicular to the steps for Co/Cu(100) and Fe/Ag (100). The same films produced square loops when applied magnetic field was rotated by 90° clearly pointing to the easy axis of the step-induced anisotropy. The split loops indicated the direction of the hard axis and the shift field (H_s) of the split loops was directly proportional to the step-induced anisotropy [122].

When the step edges break the in-plane two-fold symmetry of a (110) surface of a bcc system, such as in Fe/Mo(110) films, the resultant in-plane anisotropy is still uniaxial and the step-induced contribution can be found from comparison of the anisotropy constants obtained for the stepped and flat surfaces. The anisotropy constants can be found from the anisotropy field H_a , which is defined as that needed to saturate the magnetisation in

the hard direction. The expression for H_a can be calculated by minimising the total energy of the system when the field is applied at a right angle to the easy axis. For a uniaxial system the total energy is, from equation (6.6):

$$E = K_1 \sin^2 \theta - \mu_0 M_s H \cos(90^\circ - \theta) \quad (6.9)$$

where the second term is the Zeeman energy. Minimising E ($\partial E / \partial \theta = 0$) and setting $\theta = 90^\circ$

$$H_a = \frac{2K_1}{\mu_0 M_s} \quad (6.10)$$

The serious disadvantage of MOKE when it is applied to anisotropy measurements is the fact that it does not measure the magnetisation directly. The loops measured along the hard axis only provide the anisotropy fields. To obtain the anisotropy constants, a value for the saturation magnetisation has to be assumed. In addition high accuracies cannot be obtained for large anisotropies because at large fields, the change in the Kerr signal is too low to reliably determine anisotropy [123]. However, when a film is grown on a vicinal surface with steps propagating between the [001] and [1-10] directions, the orientation of the preferred axis of step-induced anisotropy can be determined from the shift of the anisotropy axes away from the main crystallographic directions.

6.2. Fe/Mo(110) and Fe/W(110) epitaxial systems

Bcc Mo(110) and W(110) surfaces are suitable substrates for epitaxial growth of 3d transition metals films. In particular these systems are thermodynamically stable due to the higher surface energy of the substrate ($\gamma_{Mo} = 2.95 \text{ J}\cdot\text{m}^{-2}$ and $\gamma_W = 3.3 \text{ J}\cdot\text{m}^{-2}$) compared to the film ($\gamma_{Fe} = 2.0 \text{ J}\cdot\text{m}^{-2}$) [124]. The absence of surface alloying or inter-diffusion up to about 900 K [10] and high mobility of Fe atoms allow to grow a variety of nanostructures by changing the substrate temperature, deposition rate, orientation and density of atomic steps. In addition, these systems are of interest to study the effects of

the film strain on magnetic properties as a large tensile film strain is induced by the lattice mismatch at the film-substrate interface ($(a_{Mo} - a_{Fe})/a_{Fe} = 9.8\%$) and $(a_W - a_{Fe})/a_{Fe} = 10.4\%$) [119]. The fact that magnetic behaviour is strongly correlated with the film morphology, strain, electronic and defect structures makes Fe/W(110) and Fe/Mo(110) attractive system to explore fundamental and applied issues of magnetism.

The growth of Fe on W (110) and Mo(110) has been well studied in the last decade. At room temperature the growth starts in a layer-by-layer or Frank-Van-der-Merwe (FVM) mode, though the roughness of the film increases at higher coverages as a new layer nucleates before the underlying layer is fully closed. Below a critical thickness of about 100 Å (~60 ML) Fe films grown on W(110) exhibit an in-plane rotation of the easy axis of magnetisation from the bulk [001] to [1-10] direction. This switching behaviour was ascribed to the dominant contribution of the surface anisotropies to the overall energy density for decreasing film thickness [125].

In the films grown at elevated temperatures ($400 \leq T \leq 800$ K), the strain, induced in the film due to the large lattice misfit between the film and the substrate is released through the formation of three-dimensional wedge-shaped Fe islands on the top of a closed pseudomorphic monolayer, (layer-plus-island or Stranki – Krastanov (SK) growth). The thickness of these islands can be significantly larger than the thickness measured during the film deposition. Sander *et al.* demonstrated for a 20 Å (~12 ML) Fe film grown at room temperature on W(110) that the in-plane easy axis changes its orientation from [1-10] to [001] when the film is annealed at 700 K [126]. This reorientation results from the change of the growth mode when deposited Fe atoms assemble into islands and the thickness of these islands becomes higher than the critical thickness for the reorientation of the easy axis. Fe islands grown on Mo(110) also have the easy axis of magnetisation along [001] [127]. The height of these islands strongly depends on the annealing conditions as at higher temperatures Fe agglomerates into larger islands, which propagate across a larger number of atomic steps and as result have larger thickness [64].

The Fe/W(110) system exhibits a variety of interesting magnetic properties when the Fe coverage (θ) is below 2 monolayers (*ML*). At 300 K Fe films begin as an array of unconnected monolayer thick islands, which are superparamagnetic at 115 K [128]. At a coverage greater than 0.6 *ML*, ferromagnetic order is induced by magnetic percolation through island coalescence (Fig. 6.5a). The first closed Fe layer becomes ferromagnetic below 225 K [128, 129], with the in-plane easy axis of magnetization along the [1-10] direction [128, 130]. Dislocation – free second layer islands are formed on top of this layer in the $1 \leq \theta \leq 1.5$ *ML* coverage range (Fig. 6.5b), which have a strain-induced out-of-plane magnetization and are ferromagnetic at 300 K [130]. As the film coverage increases above 1.5 *ML*, the average Fe island area also increases and dislocations begin to form. The resulting strain relief causes a reorientation of the magnetic easy axis back into the film plane. At elevated temperatures $500 \leq T \leq 800$ K Fe atoms arrange themselves into arrays of alternating monolayer/double-layer thick stripes by the step flow mechanism (Fig. 6.5c). These stripes grow only if the atomic steps on W(110) surface propagate in [001] direction or close to it. The double-layer thick stripes have an out-of-plane magnetisation and antiferromagnetically couple to one another by dipolar interactions [131]. Exposure to the UHV residual atmosphere forces the reorientation of the easy-axis of magnetisation back into the film plane [132]. Ferromagnetic order is not established in the elevated temperature grown Fe/W(110) when the substrate steps are aligned along the [1-10] direction below a coverage of 1 *ML*. Fe atoms in this case form chains of two-dimensional triangular islands growing along the steps (Fig. 6.5d) [133].

The growth of Fe on Mo(110) surface has been extensively studied [64, 124, 134]. However very few data is available on the magnetism of Fe/Mo(110) system. It is known that in Mo/Fe/Mo(110) sandwiches the magnetisation easy axis lies along the [001] direction for the Fe layers in the thickness range $7.5 \leq d \leq 100$ Å ($4.4 \leq d \leq 59$ *ML*) with a large in-plane anisotropy field of about 0.5 *T* between the [001] and [1-10] axes. In contrast, the [1-10] direction is the easy axis in W/Fe/W(110) multilayers [135] for Fe layers in the same thickness range. The differences in these two systems were ascribed to the differences in the structure and hybridisation at the interfaces. Theoretical studies of magnetic anisotropy for single and double Fe layers on Mo(110) and W(110) substrates

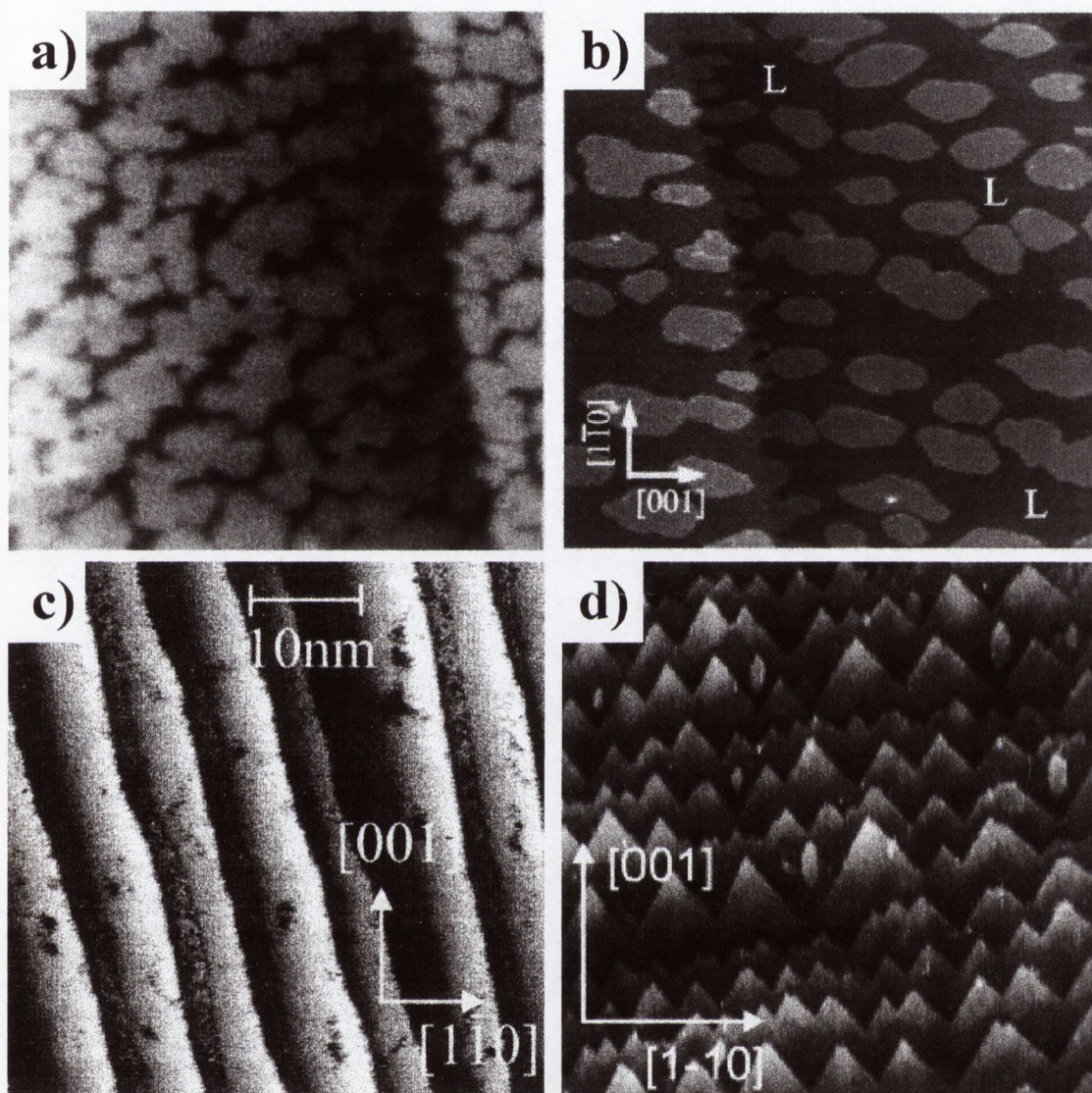


Figure 6.5 a) $700 \times 700 \text{ \AA}^2$ image of 0.66 ML Fe/W(110) film grown at 300 K (Ref. [128]). Above 0.6 ML coverage, ferromagnetic order is induced by magnetic percolation via island coalescence. First closed Fe monolayer is ferromagnetic below 225 K ; b) $900 \times 900 \text{ \AA}^2$ STM image of a pseudomorphic Fe/W(110) 1.45 ML film prepared at 300 K (Ref. [130]). The film consists of double layer islands on top of a pseudomorphic Fe monolayer, indicated by *L*. Dislocation-free 2 ML islands have a strain-induced out-of-plane magnetization and are ferromagnetic at 300 K ; c) $450 \times 450 \text{ \AA}^2$ STM image of 1.8 ML Fe/W(110) film grown at 700 K (Ref. [131]). Fe atoms arrange themselves into arrays of alternating monolayer/double-layer thick stripes. 2 ML stripes have an out-of-plane magnetisation and antiferromagnetically couple to one another by dipolar interactions; d) $650 \times 650 \text{ \AA}^2$ STM image of a W(110) surface with steps along $[1-10]$ covered by 0.5 ML Fe film grown at 300 K and post-annealed at 700 K (Ref. [133]). Fe atoms form chains of 1 ML triangular islands. No ferromagnetic order is established in these films up to 0.9 ML coverage.

were performed using the first-principle full-potential linearised augmented plane-wave calculations (FP-LAPW) [136]. The in-plane [1-10] direction was found to be the easy axis of magnetisation for both systems with a stronger in-plane anisotropy for Fe/W(110). Multiple experiments confirm this result for Fe/W(110) but no in-situ measurements of magnetic properties of ultrathin Fe films on Mo(110) has been done so far. Previous experimental data were obtained ex-situ with a metal overlayer on top of the Fe film. However, the magnetic behaviour of the metal-coated films and uncoated films can be notably different [7, 8, 137], so no reliable conclusions can be drawn without *in-situ* measurements. This chapter presents the details of magnetic anisotropy investigation of uncapped Fe films grown on a vicinal Mo(110) substrate measured under UHV conditions.

6.3 Sample preparation

The sample was prepared from a 4N purity single Mo crystal (Surface Preparation Laboratory, The Netherlands [138]). The surface was aligned to within a few degrees off the (110) crystal plane and finished by mechanical polishing to a surface roughness of $0.03\ \mu\text{m}$. The sample was fixed in a sample-holder with a Ta cap and inserted into the UHV system described previously in section 4.1. The procedure for cleaning the sample in UHV is described in section 4.2. Initially, the surface was oxidised by annealing at $\sim 1300 \div 1400\ \text{K}$ in an oxygen atmosphere of partial pressure $\sim 5 \times 10^{-7}\ \text{Torr}$ for 30 to 60 minutes. The sample was then flash-annealed to temperatures reaching $2300 \pm 100\ \text{K}$ for 15 to 20 seconds to remove the oxide layer. It was then left to cool for 5 to 10 minutes before the next flash, which allowed the chamber pressure to recover to the low $10^{-10}\ \text{Torr}$. This cycle was repeated several times. LEED and AES analysis were used to assess the cleanliness and structural order of the surface. After considerable repetition of this procedure, it was found that a carbon peak did not appear above the detection limit of the auger system, indicating a concentration below 1 at. %. It should be noted that the main *LMM* Auger transition for sulfur at $152\ \text{eV}$ is obscured by a molybdenum peak at $148\ \text{eV}$. While this means that the presence of sulfur contamination cannot be completely

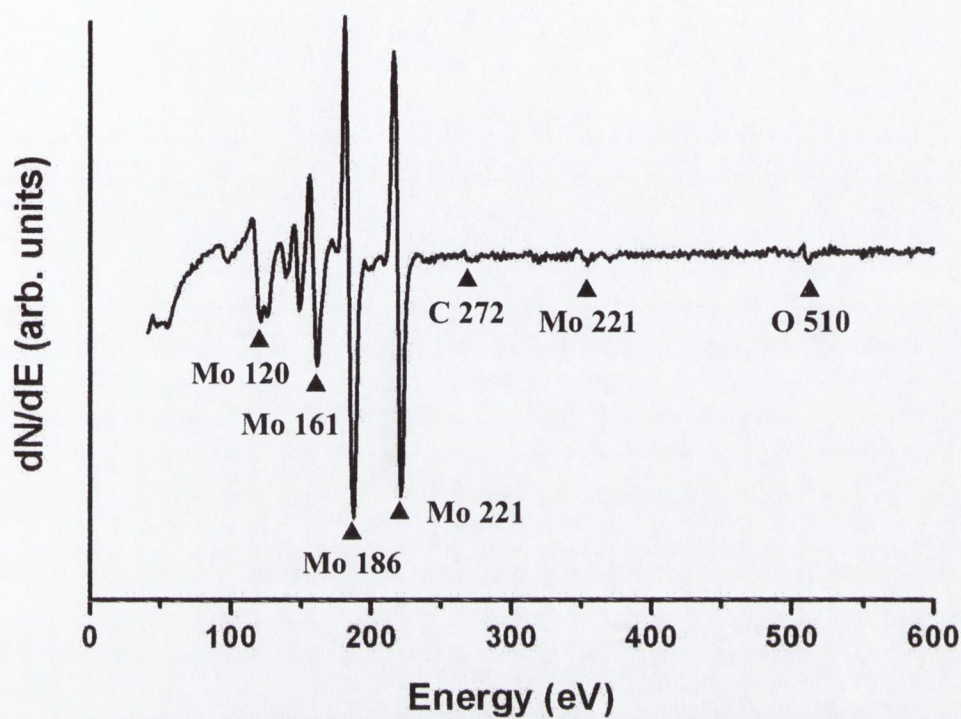


Figure 6.6 Auger spectrum of clean Mo(110) surface after cleaning by flash-annealing.

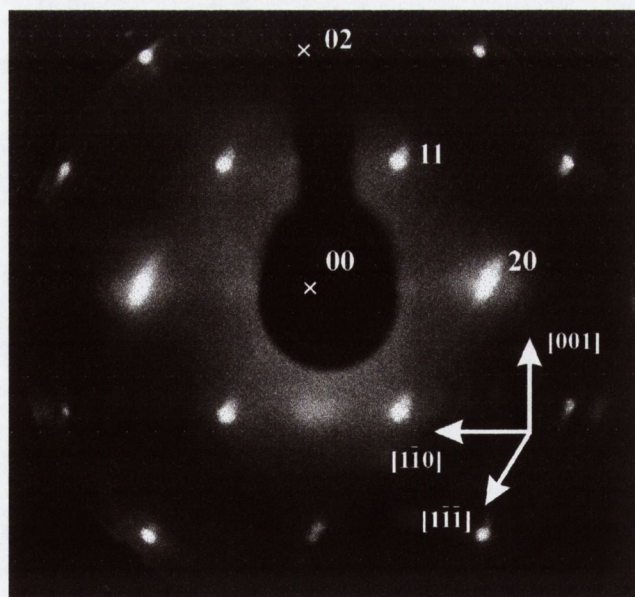


Figure 6.7 LEED pattern of clean high-index Mo(110) surface. The high step density on the vicinal surface produces the spot smearing along $[1-1-1]$ direction. $E_{\text{beam}} = 186 \text{ eV}$.

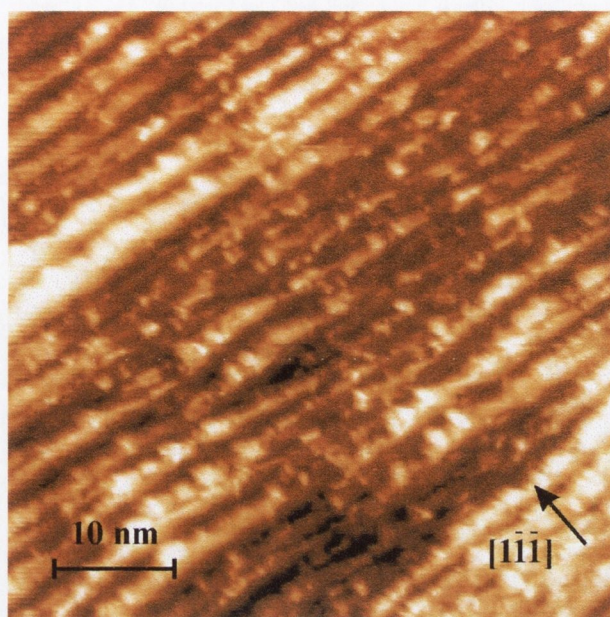


Figure 6.8 500×500 Å² STM image of the clean Mo(110) surface with global plane fitting. . The surface topography is characterised by a high density 'staircase' arrangement of terraces with an average width $d = 27 \pm 6$ Å, separated by mostly monatomic steps.

excluded, the cleaning procedure was assumed to produce concentrations of sulfur of the same order as the carbon level. Oxygen peaks were found to persist in Auger spectra after annealing. As result, the oxidising treatment was abandoned after a carbon-free surface was obtained and further cleaning was carried out by flash-annealing alone.

6.4 Clean Mo(110) vicinal surface

Figure 6.6 shows a typical Auger spectrum taken after cleaning by flash-annealing. A low Auger signal ratio $O/Mo < 0.03$ indicates desorption of oxygen from the surface leaving a clean Mo surface. A typical LEED pattern obtained from the clean surface is shown in figure 6.7 with some of the beams indexed. The presence of sharp diffraction spots and low background intensity suggests that the surface is well ordered. Following the approach outlined in section 4.3, a molybdenum lattice parameter was calculated for different energies, to an average value of $a_{Mo} = 2.98$ which is 5.3 % lower than the known

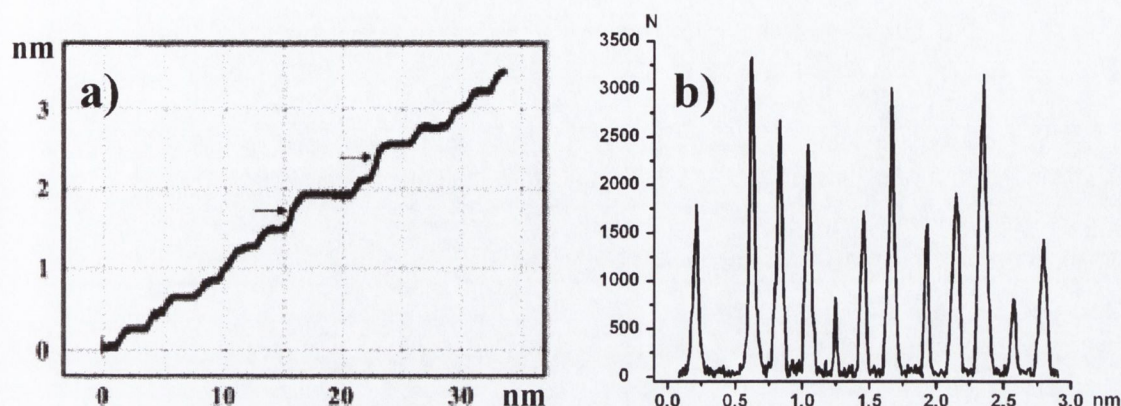


Figure 6.9 Line profile and histogram of Mo(110) terraces after local plane fitting to a terrace: a) a line profile along the direction marked in figure 6.8 shows that the surface follows a 'staircase' topography. The arrows mark double steps. b) a histogram of figure 6.8 which shows that the step heights are mostly monatomic, with an average value of $h = 2.17 \text{ \AA}$.

value of 3.147. This is within the geometrical error associated with the experimental set-up as laid out by Quinn [66].

Figure 6.8 shows a globally plane-fitted $500 \times 500 \text{ \AA}$ STM image of the clean Mo (110) surface. The surface topography is characterised by a 'staircase' arrangement of high-density terraces of average width $d = 27 \pm 6 \text{ \AA}$ separated by mostly monatomic steps (see figure 6.9(a)). The average value of the step height is measured from histograms of locally plane fitted images, like the one shown in figure 6.9(b). This histogram distributes the total pixel count of the image, N , as a function of the colour-scale height value of each pixel. As a result, terraces appear as peaks while the step height is determined from the separation between them. In this case, the measured step height is $h = 2.17 \text{ \AA}$, which is within 2.2 % of the value for the unrelaxed surface ($2.22 \text{ \AA}$). The discrepancy can be attributed to the error in the z-calibration of the piezo tube-scanner in the microscope [66]. Taking these values for terrace width and step height and applying them to the simple geometrical expression:

$$\tan \alpha = \frac{h}{d} \quad (6.11)$$

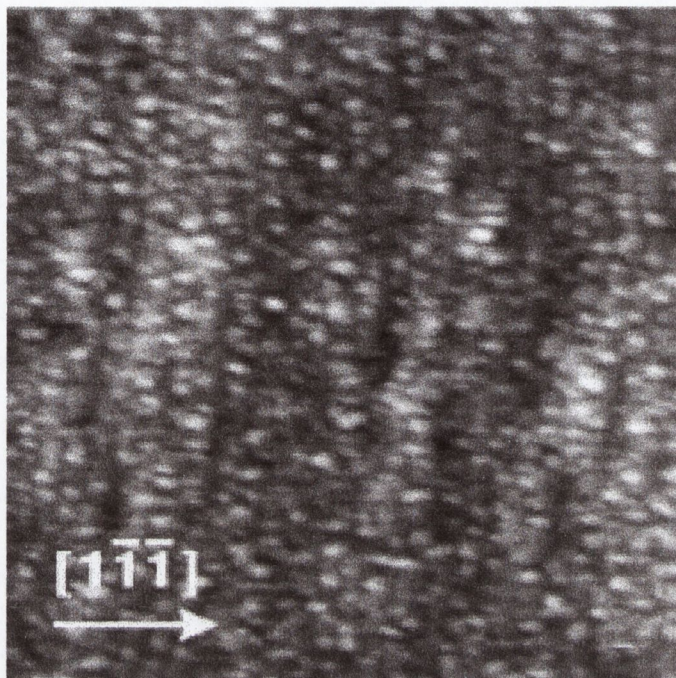


Figure 6.10 $600 \times 600 \text{ \AA}^2$ STM image of a 0.6 ML Fe film grown on a vicinal Mo(110) surface at room temperature. The film growth is initiated by the formation of a large number of pseudomorphic monolayer Fe islands with a mean diameter of 10-20 $\text{\AA}$. As the growth proceeds these islands merge together to form a closed monolayer pseudomorphic to the substrate. Reproduced from Ref. [62].

yields a value for the miscut angle of 4.6° off the (110) plane. Using a mark on the cap of the sample-holder as a reference, the direction of step-edges in the STM images was correlated with LEED data. It was found that terraces were separated by $[1-1-1]$ steps where the step-edges on the surface run parallel to the $[1-1\ 2]$ crystallographic direction which makes 35.26° angle with $[001]$ crystallographic axis. The direction of the miscut along $[1-1-1]$ axis was confirmed by high-resolution X-ray diffraction measurements (Bede Scientific Instruments Ltd [139]).

6.5 Fe growth on vicinal Mo(110) near room temperature

Following the procedure described in section 4.2, the Mo(110) surface was cleaned 30-60 minutes prior to deposition. Films in a thickness range $2ML \leq \theta \leq 7.1ML$ (3.4 - 12 Å) were grown at room temperature with the deposition rate $0.5ML \cdot \text{min}^{-1} \leq D \leq 0.55ML \cdot \text{min}^{-1}$. Film coverages θ for all films (including those where the misfit strain is relieved) are described in pseudomorphic monolayers (ML) where $\theta = 1$ corresponds to the atomic packing density of a Mo(110) plane $n_{Mo} = 1.43 \times 10^{19} m^{-2}$. The coverage was calculated from the deposited Fe thickness, determined using a quartz crystal monitor. For bulk bcc Fe, the atomic density of a single (110) layer is $n_{Fe} = 1.72 \times 10^{19} m^{-2}$ and the layer thickness d_{Fe} is 2.03 Å. Therefore the coverage for a deposited thickness of 2.03 Å corresponds to $\theta = n_{Fe} / n_{Mo} = 1.2ML$.

STM images of Fe films grown on the Mo(110) substrate at room temperature with a nominal thickness of 2.4 ML (4 Å), 3.5 ML (6 Å) and 7.1 ML (12 Å) are shown in Fig. 6.11. At room temperature, the film growth is initiated by the formation of a large number of pseudomorphic monolayer Fe islands with a mean diameter of 10-20 Å (Fig. 6.10). As the growth proceeds these islands merge together to form a closed monolayer pseudomorphic to the substrate. At higher film thicknesses the new Fe layers have an increasing tendency to nucleate before the previous layer is fully completed, leading to an increased film roughness (Fig. 6.11(a)). This represents the transition to Stranski-Krastanov growth driven by the high step density, which provides numerous high coordination nucleation sites and reduces the adatom diffusion length between the steps compared to the low-index surface. The typical staircase pattern of the substrate is maintained, though with a loss in the straightness of the surface steps with increasing thickness (Fig. 6.11 (b, c)). There is no evidence of dislocation formation in the STM data. The LEED pattern (Fig 6.12) corresponds to the Mo(110) surface and does not show any additional features indicating that the film grows in one-to-one registry with the substrate. This may be compared to the case for Fe films deposited on a low-index

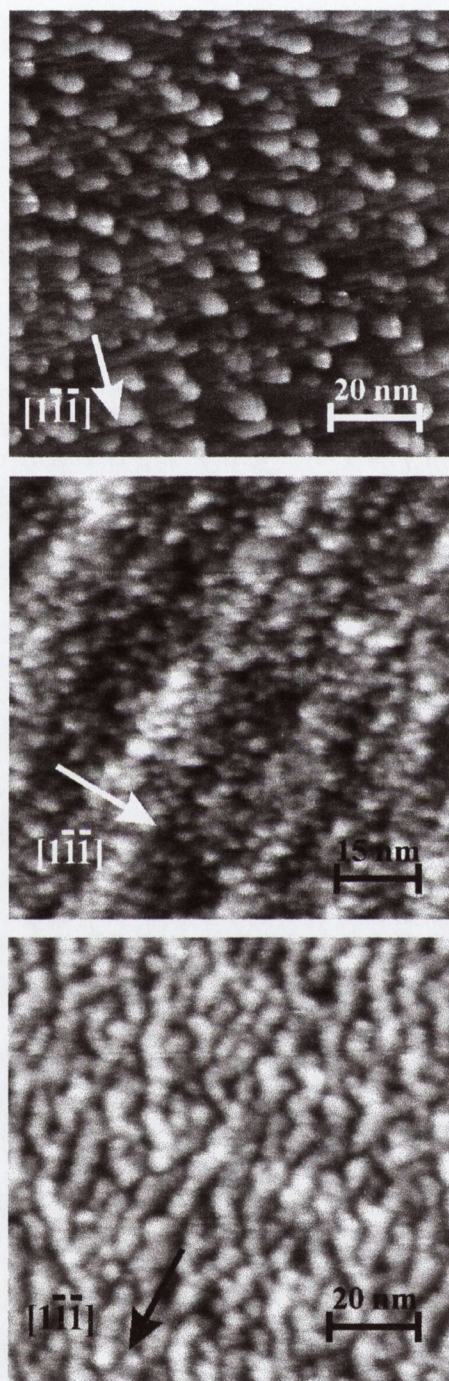


Figure 6.11 STM images of Fe films grown on vicinal Mo(110) surface at room temperature. (a) $1000 \times 1000 \text{ \AA}^2$ image of a 2.4 ML (4 Å) nominal thickness Fe film on Mo(110); (b) $850 \times 850 \text{ \AA}^2$ STM image of a 3.5 ML (6 Å) Fe film on Mo(110); (c) $1000 \times 1000 \text{ \AA}^2$ STM image of a 7.1 ML (12 Å) Fe film on Mo(110). The film surface roughness is increased and the straightness of the atomic steps is lost with the increasing film thickness.

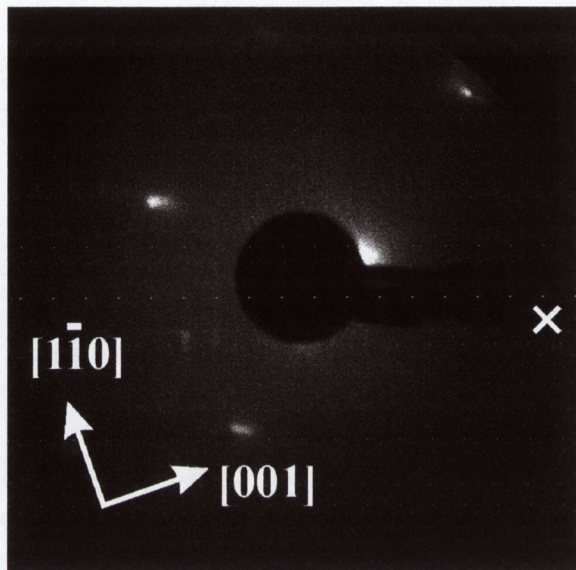


Figure 6.12 LEED image of a 3.5 *ML* (6 Å) Fe film grown at room temperature taken with a primary beam energy of 68 *eV*. The pattern corresponds to a Mo(110) centered-rectangular unit cell. No additional diffraction spots appear indicating that the film grows in one to one registry with the substrate.

Mo(110) surface, where dislocation lines were observed to form at a nominal thickness of 2.7 Å (1.6 *ML*) and a two-dimensional dislocation network was formed at 4 Å (2.4 *ML*) [64]. Typically, the Fe islands formed on the vicinal surface are much smaller than those formed on a low-index surface due to the high step density, which enhances island nucleation. The mismatch-induced strain in these smaller islands is effectively relieved by the relaxation of atom positions at the island edges, thereby precluding the necessity to form strain-relieving dislocation lines or networks.

6.6 Elevated temperature growth

The films were grown at room temperature following the procedure described in the previous section and then annealed in UHV for two hours at 750 K. Figure 6.13 displays STM images of the films with deposited coverages 2.4 and 3.5 *ML* (4 and 6 Å). There is

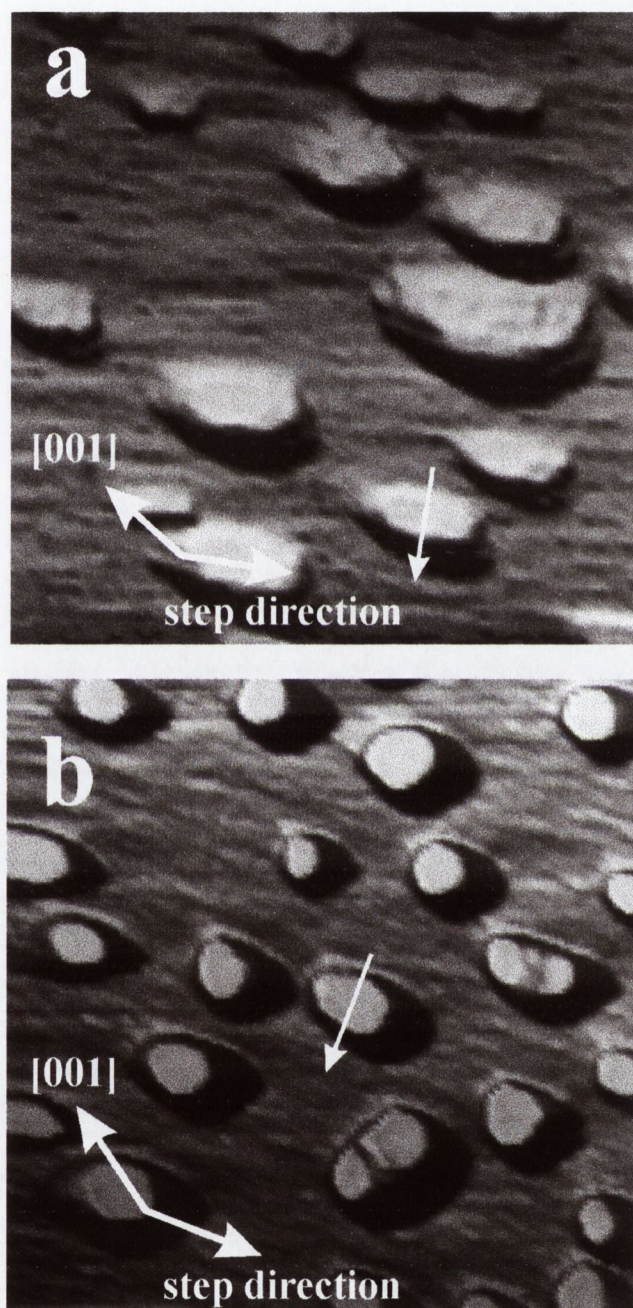


Figure 6.13 STM images of Fe films grown on vicinal Mo(110) surface at room temperature and post-annealed at 750 K for two hours. The films are characterised by large well-separated Fe islands supported on a closed Fe monolayer (Stranski-Krastanov growth). (a) $2000 \times 2000 \text{ \AA}^2$ image of a 2.4 ML (4 Å) nominal Fe thickness (b) $3000 \times 3000 \text{ \AA}^2$ image of a 3.5 ML (6 Å) Fe thickness.

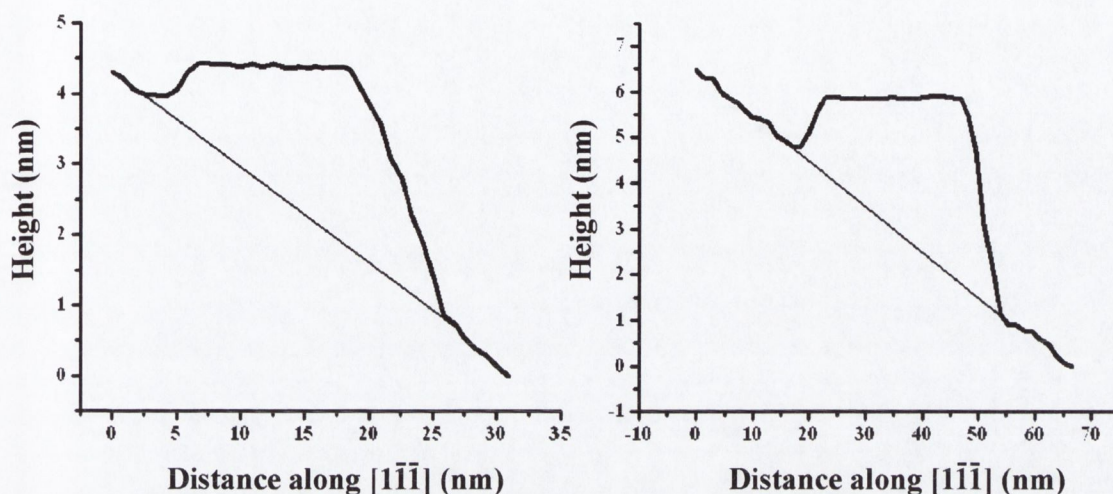


Figure 6.14 Line profile along $[1-1-1]$ direction marked in figure 6.13 for 2.4 and 3.5 ML deposited Fe coverage. The wedge-shape Fe islands form as they propagate over several terraces while maintaining a flat (110) surface.

a significant transformation in the film morphology after annealing. The films are characterised by large well-separated Fe islands supported on a closed Fe monolayer (Stranski-Krastanov growth). These islands have a well-defined shape where their edges are generally straight and lie along the low-index crystallographic directions such as the $[1-1-1]$ and $[001]$ directions. Many of these islands are elongated along a direction making a $0 - 12^\circ$ angle with the atomic step edges, whereas preferential growth along the $[001]$ direction is typical for the low-index Fe/Mo(110) system [64]. The preferential growth direction is changed because the $[001]$ axis lies close to the step direction and the high step density provides a large number of high coordination bonding sites which are readily occupied by the Fe atoms. Each island continues across several terraces in the direction of the miscut. While propagating across these terraces, the islands minimise their surface free energy by maintaining a flat (110) surface that is unbroken by steps. The local island thickness increases by one monolayer for each substrate step-edge crossed. The thickness changes from an initial a few ML to a few nanometers at the thick end. As a result, the islands have a distinctive wedge-shaped cross section along the miscut direction, which is highlighted in figure 6.14. This phenomenon has been

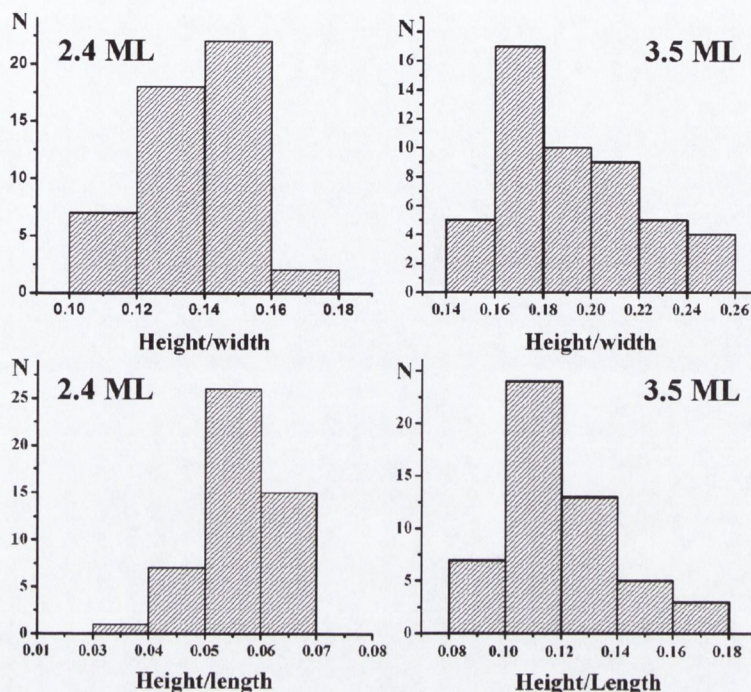


Figure 6.15 Histograms of vertical aspect ratios obtained from the analysis of the STM images for 2.4 and 3.5 ML Fe films grown on Mo(110) at 300 K and post-annealed in UHV at 750 K. The maximum island thickness is referred to as an island height. Islands with larger vertical aspect ratio are formed for thicker films.

observed previously for both the Fe/W(110) and Fe/Mo(110) systems [62, 64, 134, 140]. The islands sides can be much steeper than that shown in Fig. 6.14, but appear sloping due to the convolution of the STM tip and island shape. Figures 6.13 - 6.15 show that islands of higher thickness and vertical aspect ratio (height/length and height/width) are produced when the deposited thickness is increased. The shape has a strong impact on the magnetic behaviour of these islands as will be demonstrated in the following section. The in-plane demagnetising field is increased in the islands of larger vertical aspect ratio resulting in a higher magnetic coercivity.

The presence of the pseudomorphic monolayer cannot be detected by STM because in the areas between the islands the film morphology follows the topography of the underlying substrate. However, the presence of the closed monolayer was detected by Murphy *et al.*

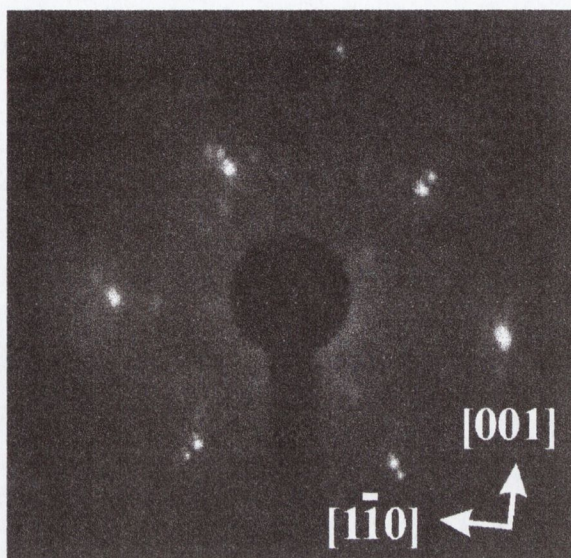


Figure 6.16 LEED pattern produced by a 3.5 ML ($6\text{\AA}$) Fe film grown at room temperature and post annealed in UHV at 750 K for two hours. The first order spots of the unstrained Fe(110) lie outside of those of the pseudomorphic monolayer covering the Mo(110) substrate. $E_{\text{beam}} = 117\text{ eV}$.

in a quantitative analysis of the area and local thickness of islands in STM images and a comparison with the amounts deposited for each film as measured by the quartz crystal balance [134]. In an AES, LEED and thermal desorption (TPD) study of the Fe/Mo(110) system by Tikhov and Bauer [10], it was observed that the Mo Auger signal increased for all coverages except the first Fe layer, upon annealing films grown at 300 K. This was attributed to Fe agglomeration, which did not occur in the first Fe layer. The presence of a pseudomorphic Fe monolayer was also observed in similar films grown on W(110) [114, 126].

Figure 6.16 shows the LEED pattern observed on a 6-ML film grown at 300 K and post-annealed at 750 K. The LEED pattern is consistent with the unstrained 1×1 Fe(110) surface of the islands superimposed on that of the pseudomorphic monolayer covering the substrate between the islands. This indicates that the strain induced in the Fe films due to the large mismatch at the film-substrate interface is effectively relieved in the 3D Fe islands.

6.7 Magnetic anisotropy in Fe/Mo(110) films grown at room temperature

The longitudinal SMOKE setup described in chapter 5 was employed to investigate the magnetic anisotropy in Fe/Mo(110) films. Hysteresis loops were measured at 300 K for different azimuthal orientations of the crystal with respect to the direction of the applied magnetic field. The sample in-plane rotation angle φ was measured from the [00-1] crystallographic axis in an anticlockwise direction as shown in Fig. 6.17. The normalised angle-dependent hysteresis loops for 3.5 ML and 5.9ML (6 and 10 Å) films are displayed in Fig. 6.18. Qualitatively, films of other thicknesses behaved similarly at room temperature down to 2.4 ML (4 Å) thickness, aside from a Kerr intensity change that generally scaled with the film thickness according to Eqs. (2.31). Rectangular loops with nearly unitary M_r/M_s (where M_r and M_s are the remanent and saturation magnetisations respectively) were obtained when the magnetic field was applied close to the [001] direction ($\varphi = 5^\circ$ and $\varphi = 185^\circ$ in Fig 6.18), signifying that the in-plane [001] direction was the easy axis of magnetisation in the films. The magnetisation is completely reversed within about 1 Oe. Applying the magnetic field nearly along the [1-10] direction ($\varphi = 275^\circ$ in Fig. 6.18), results in a typical hard axis loops, indicating that two-fold in-plane anisotropy is present in the films. Similar hard-axis loops were obtained in the polar MOKE configuration when the magnetic field was applied along [1-10] direction. The dependence of M_r/M_s on φ followed a $|\cos(\varphi)|$ law. Figure 6.19 shows that there were deviations from this $|\cos(\varphi)|$ dependency, which were more pronounced for thinner films. These deviations were produced by the uniaxial step-induced anisotropy, which appeared due to the large surface miscut where both hard and easy axes were oriented at an angle to the axes of the magnetocrystalline anisotropy.

Using equation (6.4) the anisotropy energy density for two uniaxial anisotropies with preferred directions are at some angle α to each other can be expressed (neglecting the second order terms) by

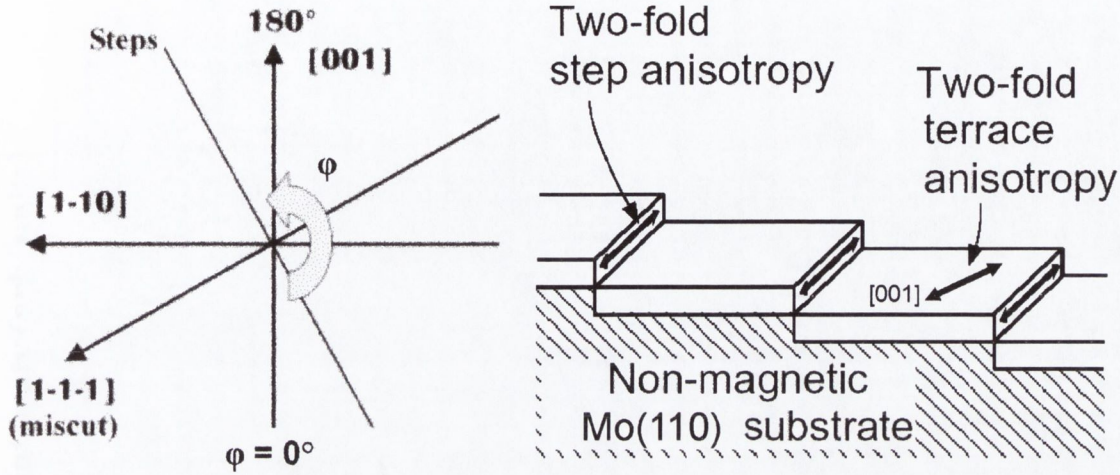


Figure 6.17 Crystallographic axes of the Mo(110) substrate with respect to the direction of the miscut. φ denotes the angle between $[00-1]$ direction and the applied magnetic field.

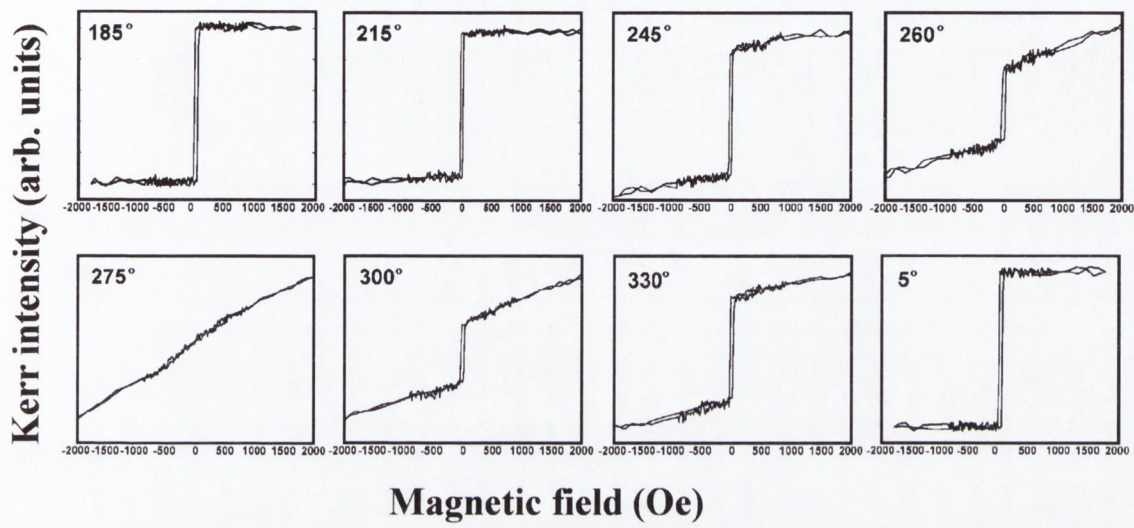
$$E = K_1 \sin^2(\theta) + K_\alpha \sin^2(\alpha - \theta); \quad K_1, K_\alpha > 0 \quad (6.12)$$

In our case K_I is the effective uniaxial anisotropy constant (in J/m^3) for a Fe film on a flat Mo(110) surface where the easy axis of magnetisation lies along the $[001]$ direction. According to equation (6.7) it includes the bulk contribution of Fe, stretched to match the atoms of Mo(110) substrate and surface anisotropy, which is inversely proportional to the film thickness. K_α corresponds to the step-induced anisotropy; θ is the angle between the magnetisation vector and the easy axis of the first anisotropy term. Equation (6.12) is equivalent to a single uniaxial anisotropy, where the easy axis makes an angle β to the easy axis of the first term:

$$E = K_0 + K \sin^2(\beta - \theta) \quad (6.13)$$

By expanding the $\sin^2$ terms in both (6.12) and (6.13) and equating the coefficients of each power term one can find that K_0 , K and β are related to K_I and K_α and α by the following equations:

3.5 ML



5.9 ML

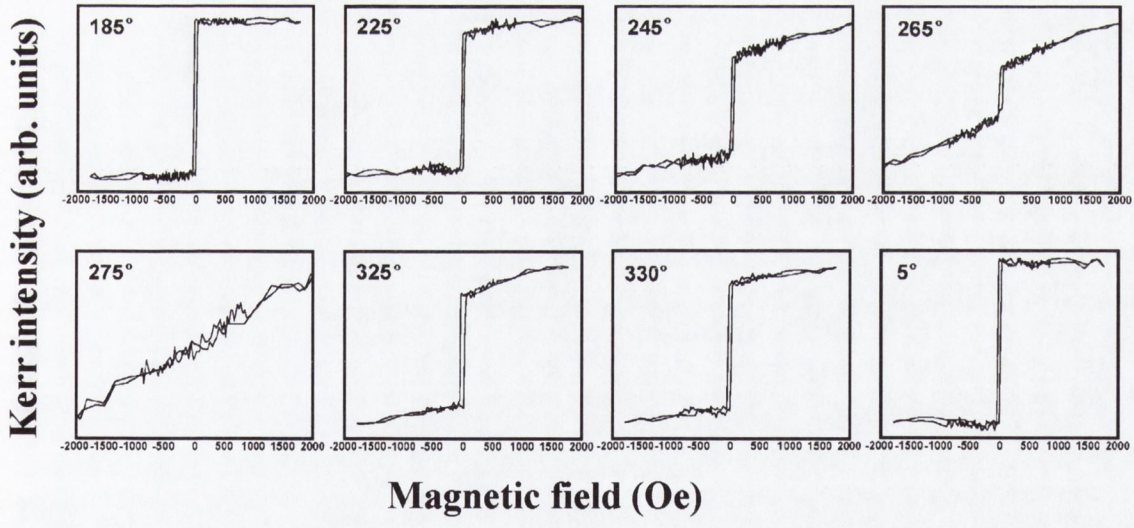


Figure 6.18 Selected normalised in-plane magnetic hysteresis loops for 3.5 (6 Å) and 5.9 ML (10 Å) Fe film on the stepped Mo (110), measured at 300 K. The angle between the [00-1] direction and the direction of the applied magnetic field is marked for each loop.

$$K_1 + K_\alpha \cos 2\alpha = K \cos 2\beta \quad (6.14)$$

$$K_\alpha \sin 2\alpha = K \sin 2\beta \quad (6.15)$$

$$K_\alpha \sin^2 2\alpha = K \sin^2 2\beta + K_0 \quad (6.16)$$

Solving the system of equations (6.14- 6.16) one can find that K is related to K_1 , K_α and α as:

$$K = \sqrt{K_1^2 + K_\alpha^2 + 2K_1K_\alpha \cos 2\alpha} \quad (6.17)$$

and β is given by:

$$\beta = \frac{1}{2} \arccos \left(\frac{K_1 + K_\alpha \cos 2\alpha}{K} \right) \quad (6.18)$$

The equation (6.16) determines the constant term K_0 , which has no bearing on the equilibrium conditions and can be ignored. Simple analysis of the equations (6.17) and (6.18) shows that two uniaxial anisotropies with easy axes orthogonal to each other ($\alpha = \pi/2$) are equal to a uniaxial anisotropy with a constant $|K_1 - K_\alpha|$ and the easy axis along that of the stronger anisotropy. If $\alpha < \pi/2$ then the new easy axis is confined between the preferred directions of two anisotropies given by the first and the second terms in equation (6.12) and lies closer to the easy axis of the larger constant anisotropy.

The constant of the resulting anisotropy K can be found in the experiment as it is related to the anisotropy field H_a by equation (6.10). However, the maximum magnetic field of 2200 Oe, accessible from the electromagnet described in chapter 5.1, was not enough to saturate these films along the hard axis ($\varphi = 275^\circ$ in Fig. 6.18). As result the anisotropy could only be roughly estimated from the equation (6.10) using $M_s = 1.7 \cdot 10^6$ A/m for bulk Fe and found to be stronger than 1.9×10^5 J/m³ in the studied thickness range.

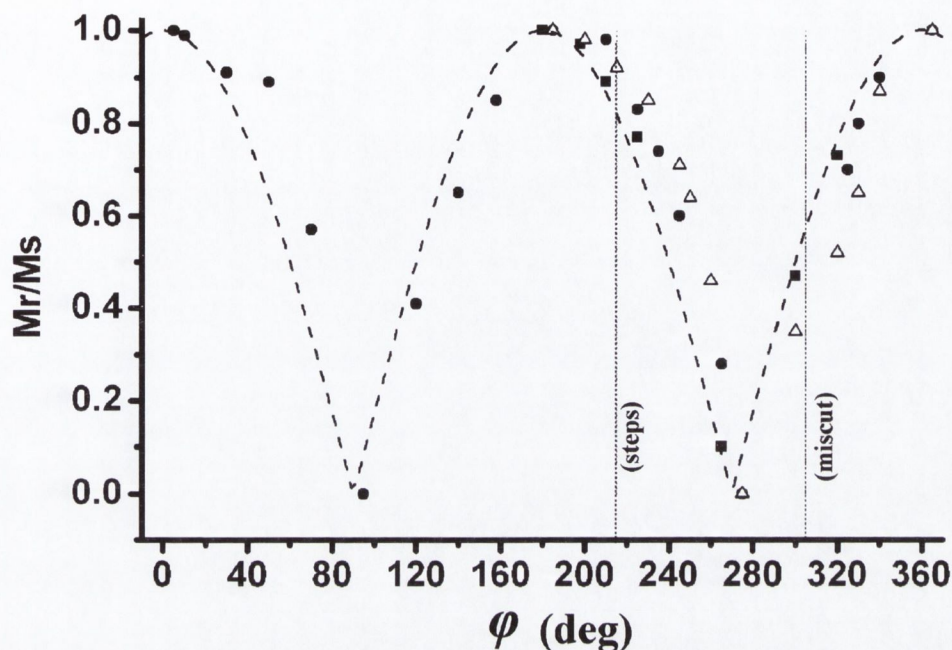


Figure 6.19 Ratio of remanent to saturation magnetisation *versus* azimuthal angle φ for 3.5 ML (6 Å) (Δ), 5.9 ML (10 Å) ($\bullet$) and 7 ML (12 Å) ($\blacksquare$) Fe films epitaxially deposited on the Mo(110) stepped surface. The dashed line depicts a $|\cos(\varphi)|$ function. $\varphi = 0^\circ$ corresponds to the magnetic field applied along the [00-1] direction. Maximum of M_r/M_s *versus* φ is shifted from the [001] direction towards the atomic steps, indicating that the easy axis of the step-induced anisotropy lies along the atomic step-edges.

Nevertheless the shift of the easy axis from the [001] direction towards the step edges was found by plotting M_r / M_s *versus* φ as shown in Fig. 6.19. This indicates that the easy axis of the step-induced anisotropy lies along the atomic step-edges and the hard axis coincides with the miscut direction. It is clear from figure 6.19 that the influence of the step-induced magnetic anisotropy weakens when the Fe coverage is increased. No influence of the miscut on the magnetic behaviour of the Fe films was observed for the thicknesses above 7 ML (12 Å). At this thickness the staircase pattern of the substrate is lost in the film and the M_r / M_s *versus* φ dependence follows the $|\cos(\varphi)|$ law shown by the dashed line in Fig. 6.19.

The presented experimental data shows that in the Fe/Mo(110) ultrathin films M_r / M_s follows the $|\cos(\varphi)|$ law; the coercive field H_c is small compared to the

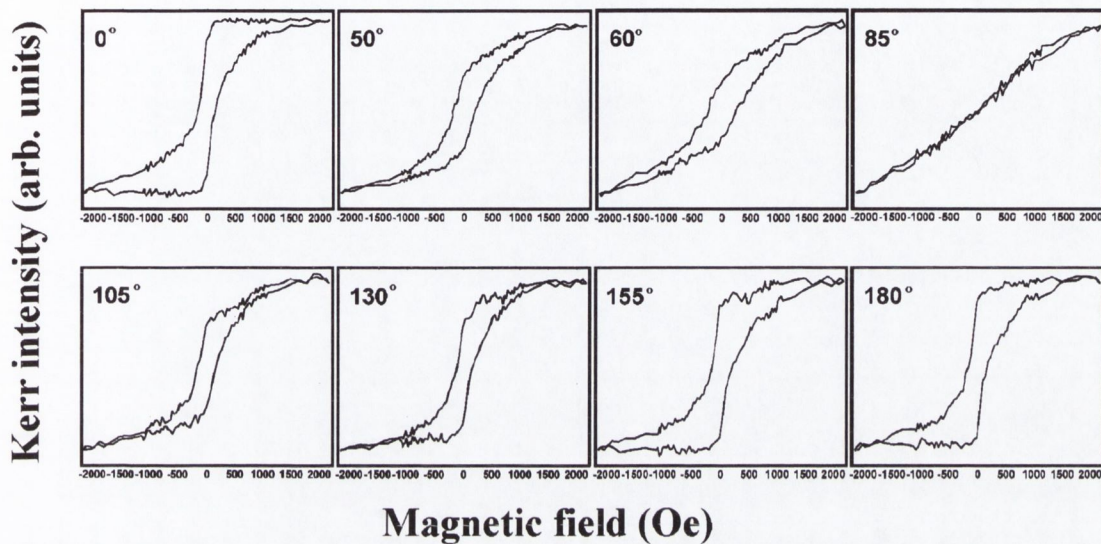
anisotropy field and the magnetisation along the easy [001] axis rapidly flips in the opposite direction as the applied magnetic field exceeds H_c . This indicates that the magnetisation reversal along the easy axis proceeds by a nucleation event followed by a nearly instantaneous propagation of a single domain wall, while it mainly occurs via the homogeneous rotation of magnetic moments when the magnetic field is applied along the hard [1-10] axis. When the magnetic field is applied in any other direction, the magnetisation reversal is a combination of these two processes. The domain wall nucleation is followed by the switch of magnetisation along the easy axis after the projection of the magnetic field on the easy axis exceeds H_c . The magnetisation process is completed via the subsequent coherent rotation of magnetisation into the direction of the applied magnetic field.

In contrast to the Mo/Fe(7.5Å)/Mo(110) system, which was found to be superparamagnetic at 300 K [135], Fe/Mo(110) remained ferromagnetic down to ~ 2.4 ML (4 Å) at room temperature. Thinner Fe films down to 2 ML (3.4 Å) produced typical easy axis loops, like the one in Fig. 6.18 ($\varphi = 5^\circ$), along the [001] axis below 250 K. The hard axis loops were measured close to the [1-10] axis, with no sign of saturation up to 2200 Oe. This contradicts the results of recent theoretical calculations [136], predicting an easy axis of magnetisation to lie along the [1-10] direction. The dramatic rise of H_c from 100 to about 3000 Oe reported for the Mo/Fe(7.5Å)/Mo(110) system [135] was not found in our experiments and is rather the result of the presence of an additional Fe/Mo(110) interface than an intrinsic property of the Fe/Mo(110) system.

6.8 Magnetic measurements on Fe/Mo(110) films grown at elevated temperature

The switch from Frank-Van-der-Merwe to Stranski-Krastanov growth resulted in significant transformation of magnetic properties of Fe/Mo(110) films. Hysteresis loops presented in figure 6.20 were measured for different azimuthal orientations of the crystal with respect to the direction of the applied magnetic field using the longitudinal MOKE.

3.5 ML



5.9 ML

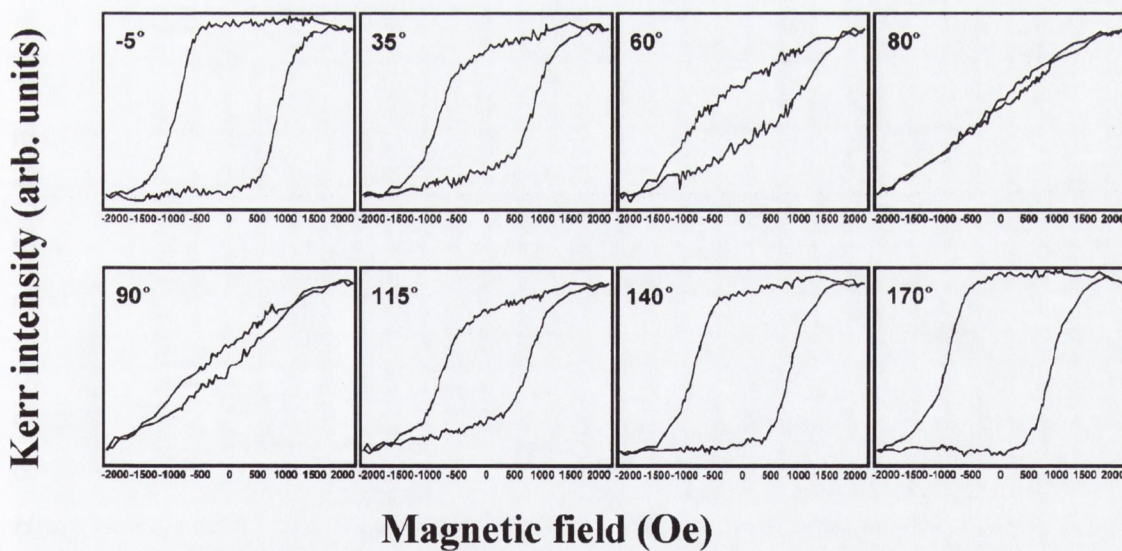


Figure 6.20 Selected normalised longitudinal magnetic hysteresis loops for $\theta = 3.5$ ML (6 Å) and $\theta = 5.9$ ML (10 Å) Fe films grown at room temperature on a stepped Mo(110) and post-annealed at 750 K. The loops were obtained at 300 K. Indicated angle φ gives the direction of the applied magnetic field. φ was measured from [00-1] crystallographic axis in the clockwise direction.

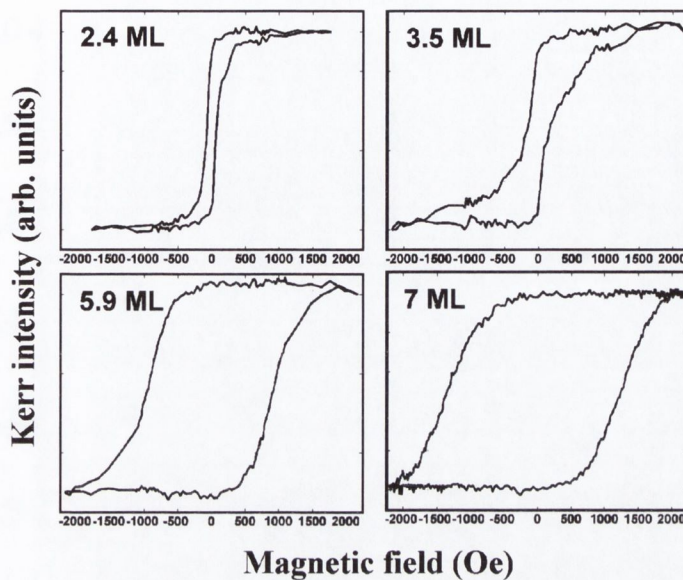


Figure 6.21 Normalised hysteresis loops taken along the magnetic easy axis of Stranski-Krastanov Fe/Mo(110) films for indicated deposited Fe coverages. H_c increases with increasing deposited Fe thickness due to a larger demagnetising field produced in the SK islands, comprising the film.

The shape of the loops shows that the magnetisation reversal proceeds by a succession of nucleation/propagation events so the region of magnetisation reversal has a finite width. The in-plane anisotropy remained uniaxial with an easy axis of magnetisation lying close to the [001] direction. The anisotropy field H_a was measured from the hysteresis loops obtained with the magnetic field applied close to [1-10] direction (Fig. 6.20 $\varphi = 85^\circ$ for 3.5 ML and $\varphi = 80^\circ$ for 5.9 ML). The strength of the magnetic field available in the experiment was not sufficient to saturate the magnetisation along the hard axis and H_a could only be estimated to be larger than 2200 Oe. The coercivity H_c and saturation field H_s increased dramatically compared to the room temperature grown films due to the large demagnetising field H_d induced in the Fe islands. The demagnetising field is always present when magnetic poles are created in the material. It is proportional to the magnetisation $\mathbf{M}$ which creates it and acts in the direction opposite to $\mathbf{M}$:

$$H_d = N_d M \quad (6.19)$$

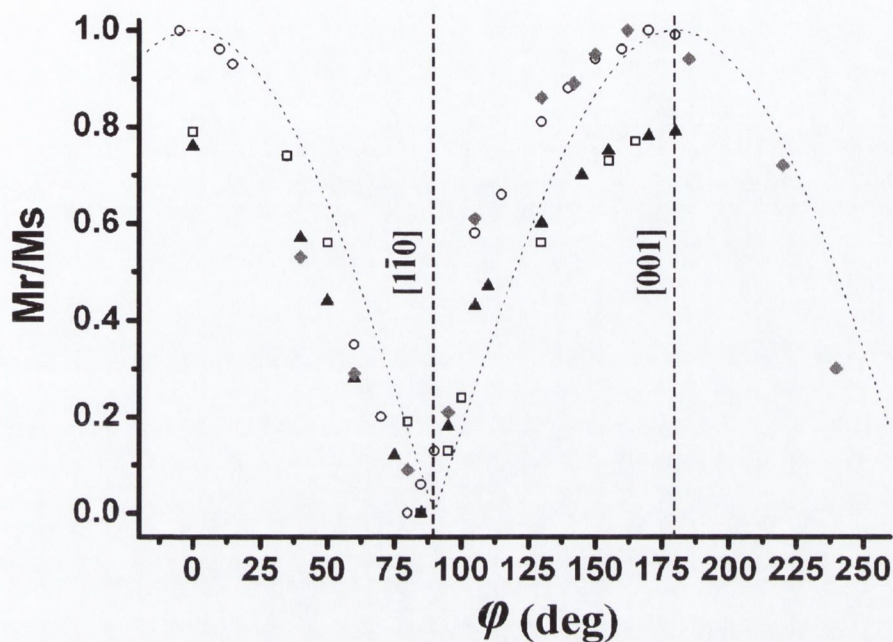


Figure 6.22 Ratio of remanent to saturation magnetisation *versus* azimuthal angle ϕ between [00-1] direction and applied magnetic field. ϕ was measured from [00-1] axis in a clockwise direction. The data was collected at room temperature for 2.4 ML (4 Å) ($\square$), 3.5 ML (6 Å) ($\blacktriangle$), 5.9 ML (10 Å) ($\circ$) and 7 ML (12 Å) ($\blacklozenge$) Fe films epitaxially deposited on the Mo(110) stepped surface at room temperature and post-annealed at 750 K.

where N_d is a demagnetising factor, which depends on the shape of the magnetised body.

Figure 6.21 displays the hysteresis loops measured along the easy axis for different deposited Fe thicknesses. H_d was found to be larger for thicker deposited Fe films due to the larger vertical aspect ratios of the islands formed for higher coverages (Fig. 6.21). It was shown in section 6.6 that the thickness of wedge-shaped islands changes from a few monolayers to some maximum referred to as the island height. Calculating the demagnetising field for these structures is not trivial. Moreover, the demagnetising factors can be calculated exactly only for ellipsoid shapes [141] because only ellipsoids can be magnetised uniformly. However, tables and charts of approximate calculated demagnetising factors are available for solids of other shapes [142]. An analytical expression for a rectangular uniformly magnetised prism was applied in this work to

estimate the demagnetising field H_d taking the mean island dimensions from STM measurements [143]. The saturation magnetisation value $M_s = 1.7 \cdot 10^{-6} \text{ A/m}$ for bulk iron at room temperature was used for calculations. This approach is justified because the strain induced in the Fe/Mo(110) films is relieved in the formed 3D Fe islands and the lattice constant is effectively reduced to its bulk value. For the islands shown in Fig. 6.13 with an average size $45 \times 17 \times 2.3 \text{ nm}$ (for $\theta = 2.4 \text{ ML}$) and $33 \times 20 \times 3.8 \text{ nm}$ (for $\theta = 3.5 \text{ ML}$) H_d along the direction of island elongation was calculated to be $\sim 1070 \text{ Oe}$ and 2068 Oe respectively, which compares with $H_s \approx 1000 \text{ Oe}$ and $H_s \approx 2000 \text{ Oe}$ measured with MOKE for these films (Fig 6.21). This shows that most of the applied magnetic field energy is spent to overcome the energy of the demagnetising field produced in the Stranski-Krastanov islands.

The plot of M_r / M_s versus azimuthal angle φ is shown in Fig. 6.22. The magnetisation reversal is homogeneous in the SK films and $M_r / M_s(\varphi)$ is not expected to follow the $|\cos(\varphi)|$ dependence like in case of the room temperature grown films. Moreover, 80 % remanent states were observed for 2.4 and 3.5 ML films when the external field was applied along the easy axis (Fig 6.21). However, $M_r / M_s(\varphi)$ graphs are obviously asymmetric about the main crystallographic axes with a discernible shift of the maximum of M_r from the [001] direction towards the step edges. The rotation of the easy axis of the effective magnetic anisotropy is produced by the film shape anisotropy, where easy axis lies parallel to the elongation direction of the SK islands.

Chapter 7

Magnetic phase transition in ultra-thin Fe films on Mo(110)

7.1. Theoretical and experimental studies of two-dimensional magnetic systems

The general theory of phase transitions formulated by Landau [144] unifies many different physical systems with an identical background symmetry and order parameter into universality classes regardless of the length scale of the underlying microscopic interactions. The main concept of this theory is the establishment of *long-range order* as a violation of the background symmetry of the system. Landau distinguished between induced and spontaneous violations of the symmetry. The latter is responsible for *second order phase transitions*, in which, similarly to the ferromagnetic-paramagnetic transition at the Curie temperature (T_c), the order parameter increases continuously from zero below the ordering temperature without volume discontinuity or latent heat.

The magnetic phase transition at T_c in a two-dimensional (2D) system has been a subject of interest for a long time. Reduced dimensionality simplifies the task and the exact solutions for some models have been obtained. Mermin and Wagner proved that an isotropic 2D Heisenberg system with short-range interactions does not have a long-range magnetic order at any finite temperature [145]. Instead, in the limit of 2D Ising model representing a 2D ferromagnet with an infinite uniaxial anisotropy the exact Onsager solution shows [146, 147] that the magnetisation M follows a universal asymptotic power law in the critical region:

$$M = \text{const} \times (1 - T / T_c)^\beta \quad (7.1)$$

with the *critical exponent* $\beta = \frac{1}{8}$.

According to the universality hypothesis formulated by Griffith [148] the critical exponents and amplitudes depend on a small number of parameters such as the dimensionality of the system and the number of the equivalent spin components. A 2D magnetic system with two equivalent in-plane directions of spin orientation is described by the planar or *XY* model. In its isotropic form it does not reveal any conventional phase transition to an ordered state, but the Berezinskii-Kosterlitz-Thouless phase transition to a state of infinite correlation length without spontaneous long-range order [149, 150]. The introduction of the cubic anisotropy in this model leads to a non-universal character of the phase transition with the critical exponents depending on the strength of the anisotropy [151]. The theoretical work on a *finite-size XY* lattice shows that this system can develop a finite magnetisation, which approaches power law behaviour at a finite T_c with the critical exponent $\beta \approx 0.23$ [152]. Real ultra-thin films possess finite magnetic anisotropies and Binder and Hohenberg demonstrated using Monte Carlo simulations that an anisotropic 2D Heisenberg system becomes Ising-like near T_c [153] in the presence of magnetic anisotropy. Later Bander and Mills rigorously proved it using renormalisation group analysis [154].

The situation in the experiment is different from the theoretical studies where an analytical and numerical treatment is easier for a reduced dimensionality. A preparation of a system that can be considered as a model low-dimensional ferromagnet is more difficult than that of a high quality three-dimensional (3D) crystal. In addition, the measurements of magnetic properties of 2D films require instruments of much greater sensitivity. Using modern techniques a single crystal can be grown with a contamination level of a few *ppm* and areas of a perfect crystallographic structure extending over a few micrometers. The magnetic properties of such crystals depend only weakly on the structural defects and the spontaneous magnetisation shows a sharp transition near T_c . Rounding of a phase transition due to the finite-size effects (discussed later in more detail) is insignificant and the magnetisation versus temperature dependence shows a

characteristic “tail” above T_c , which occupies only a limited temperature range $T \approx (10^{-3} - 10^{-4}) \cdot T_c$.

3D effects in early experiments were minimised by using layered antiferromagnetic crystals like K_2NiF_4 , which are equivalent to ferromagnets in their critical behaviour [155]. The measured values of the critical exponent were $\beta \geq 0.15$ which is smaller than $\beta = 0.34$ [156] for the bulk, but clearly larger than $\beta = 0.125$ for the 2D Ising model. The preparation of ultra-thin films with a thickness of a few atomic layers suitable for studying magnetic phenomena on 2D scale became possible due to the advances in vacuum science and the development of molecular beam epitaxial (MBE) deposition techniques. However, these films are still far from ideal. Despite the ultimate growth pressure of 10^{-10} - 10^{-11} Torr now available, the films still contain 1 at % or larger concentration of gas impurities entrapped during the deposition or adsorbed from UHV. Besides a high density of defects such as atomic steps separated by flat terraces reduces the spin-to-spin correlation length ξ in the film plane at T_c according to the scaling dependence [157]:

$$\xi = \xi_0 (1 - T/T_c)^{-\nu} \quad (7.2)$$

where ξ_0 is a characteristic microscopic length and ν is the universal critical exponent which equals 1 for the 2D Ising model. The reduced correlation length in the direction normal to the film translates into a reduction of T_c with a decreasing film thickness according to the power law:

$$\frac{T_c(\infty) - T_c(n)}{T_c(n)} = C_0 n^{-\lambda} \quad (7.3)$$

where $T_c(\infty)$ is the bulk transition temperature, $T_c(n)$ is the transition temperature of the n ML thick film, C_0 is a constant and λ is a shift exponent varying from 1 for the Ising model to ~ 2 for a system with the periodic boundary conditions.

Much of the experimental work is done on 1-5 ML transition metal films deposited on a single crystal non-magnetic metallic substrate. The experiments are usually conducted to study the variations of T_c as a function of film thickness or determine the magnetisation critical exponent from the temperature dependence of spontaneous magnetisation. Magnetic thin films grown on the surfaces of cubic crystals can be divided into two classes [158]. The films that belong to the class 1 have a uniaxial anisotropy, either parallel or perpendicular to the film plane. The films of class 2 have magnetisation in the film plane but their magnetic anisotropy is not uniaxial. Examples of studied systems that belong to the class 1 include Fe/Ag(100) [159], Fe/W(110) [160] (including the phase transition in the submonolayer films [133]), Fe/Pd(100) [161], Ni/W(110) [162], Fe/Cu(100) [163]. The systems of the second type are Fe/Au(100) [164], Ni/Cu(111) [165], Co/Cu(111) [166], Co/Cu(100) [167] *etc.* There is also a growing interest in the critical behaviour of ferromagnetic multilayer structures due to their importance for fundamental research and potential applications in data storage technologies [160].

The surface magnetic anisotropy in ultra-thin films can be extremely large. Magnetic anisotropy energies (calculated per unit volume of magnetic material) can reach a few MJ/m^3 , nearly as strong as anisotropies reported for permanent magnet materials such as YCo_5 [169]. Such anisotropy energy corresponds to an anisotropy field of several Tesla, which is strong in comparison with the bulk magneto-crystalline anisotropy of magnetic transition metals. However, corresponding anisotropy energies per atom of $0.1 \div 0.3 \text{ meV/atom} = k_B \cdot 1.2 \div 3.5 \text{ K}$ are small in comparison with the exchange energies of the order of $k_B T_c$. Therefore, the films of the first class represent an anisotropic Heisenberg model rather than a 2D Ising model. Nevertheless almost all the systems with experimentally determined β (called *effective critical exponent*) close to the Ising value of 0.125 belong to the first class. Films with $\beta \approx 0.25$ belong without exception to the second film class, which can be considered as an experimental realisation of a finite size 2D XY model system.

7.2. Fe/Mo(110) as a system for experimental studies of critical behaviour on 2D scale

There are a number of requirements in the experiments conducted to study magnetic phase transitions on 2D scale. The system must be a well-characterised epitaxial ultra-thin film and its morphology must be preserved in the temperature range where the magnetic phase transformation takes place. Morphology of Fe/Mo(110) has been quite extensively studied and well understood in the last decade. This system is thermodynamically stable and does not show any surface alloying or inter-diffusion up to about 900 K. This is in contrast to Fe/Ag, Fe/Au or Fe/Cu systems for which the inter-diffusion of the substrate and film atoms occurs at the temperatures of about 600 K [137], 400 K [170] and 200 K [171] respectively and alters magnetic properties of these films [113, 172].

Most of the experimentally studied ultra-thin films of magnetic transition metals were grown on noble metal surfaces because of a close match of their lattice constants and the dissimilarity of the electronic structures between early transition metals and noble metals. This insures minimum hybridisation between the *d*-bands of the film and the interface substrate atoms. Full potential linearised augmented plane wave (FP-LAPW) calculations show that the ferromagnetic moment of a magnetic overlayer on a non-magnetic noble metal substrate decreases by only a small amount from a free monolayer value. For example for Fe/Ag(100) [173] or Fe/Cu(100) [174] the magnetic moment of the Fe overlayer is $2.86 \mu_B$ and $2.85 \mu_B$ respectively. It is larger than that of the bulk Fe ($2.2 \mu_B$) and close to the theoretical magnetic moment of a free standing Fe(100) monolayer ($2.96 \mu_B$) [175], which indicates the lack of interaction with the substrate. Fe/Mo(110) system adds a complexity of strong *d*-band hybridisation across the interface and diminishing of the magnetic moment is expected. However, for the monolayer coverage a magnetic moment of $2.59 \mu_B$ was calculated for this system [176]. This value is very close to that of Fe(110) free surface layer ($2.65 \mu_B$) [175] and again larger than that of calculated bulk Fe ($2.2 \mu_B$). For two layers of Fe on Mo(110) the magnetic moment of the top Fe layer is

further enhanced to $2.81 \mu_B$ while the interface Fe layer has its moment reduced to $2.3 \mu_B$. It is believed that the effect of hybridisation between Fe and non-magnetic Mo substrate is compensated by the lattice expansion arising from a 9.8 % misfit in the lattice constant. The retainment and even enhancement of magnetic moment by Fe films on Mo(110) insures a strong magneto-optical signal and allows one to apply SMOKE technique to study magnetic properties of these films down to a monolayer thickness.

Another important aspect in analysis of the experimentally observed magnetic phase transition is the influence of the induced magnetic moments of the interfacial substrate atoms. The polarisation of the substrate has the effect of raising T_c in the monolayer range because the interfacial spins increase the coordination number of nearest neighbor spins around the Fe sites. Calculations show that Fe films on a noble metal surface induce a magnetic moment of $0.04 - 0.05 \mu_B$ onto neighbouring substrate atoms [173, 174]. A magnetic moment induced by a single or double Fe layer onto neighbouring Mo atoms is larger. However, of the three low-index surfaces of Mo, the Mo(110) surface has the smallest magnetic moment of approximately $0.08 \mu_B$ followed by $0.17 \mu_B$ for Mo(111) and $0.33 \mu_B$ for the Mo(100) surface [177]. Therefore, from the point of view of Fe induced magnetic moment the surface of a noble metal crystal does not offer a significant advantage over Mo(110) substrate.

To describe a critical phenomenon, one has to define an appropriate long-range order parameter. For the ferromagnetic to paramagnetic phase transition the spontaneous magnetisation M_{sp} is the order parameter and the temperature at which M_{sp} vanishes defines the T_c of the magnetic system. However, in a static magneto-optical experiment a measured observable is not M_{sp} but the component of the remanent magnetisation M_r parallel to the direction of the measurements. M_r coincides with M_{sp} only if the system is in a single domain state up to T_c and the measurements are taken along an easy axis. This is the case for ultra-thin films with an in-plane easy axis [178]. Most of the ultra-thin films with the uniaxial anisotropy like Ni/Cu(100) or Fe/Ag(100) have an out-of-plane preferred axis of magnetisation, which is set up as result of competition between surface and volume anisotropies. Vanishing of M_r in these films corresponds to the formation of

domains, which yields an abrupt decrease of the remanent signal below real T_c . In this case, choosing M_r as an order parameter leads to incorrect conclusions about T_c [178]. Besides, the out-of-plane easy axis persists only up to a certain critical thickness [179]. Only a limited number of ultra-thin magnetic systems like Fe/Ag(111), Fe/W(110) or (as was shown in the previous chapter) Fe/Mo(110) have a strong in-plane uniaxial anisotropy. In addition, Fe/Mo(110) displays square hysteresis loops of full remanence M_r indicating a well established single domain state [180], which makes it particularly suitable for ferromagnetic to paramagnetic phase transition studies.

7.3. Sample preparation

The sample was prepared from a 4N purity single Mo crystal (Surface Preparation Laboratory, The Netherlands [138]). The surface was oriented to within 1° of the (110) crystal plane and finished by mechanical polishing to a surface roughness of $0.03 \mu\text{m}$. The sample was cleaned following the same procedure as that described in sections 4.2 and 6.3. Initially, the surface was oxidised by annealing at $\sim 1300 \div 1550 \text{ K}$ in an oxygen atmosphere. The sample was then flash-annealed to temperatures reaching $2300 \pm 100 \text{ K}$. LEED and AES were employed to analyse the structural order and cleanliness of the surface. This cycle was repeated until the carbon and oxygen impurity levels were below the detection limit of the AES setup ($< 1 \text{ at. } \%$) and the surface produced a sharp 1×1 LEED pattern, with the bulk Mo(110) surface. The sample was further cleaned prior to each deposition by flash-annealing alone after a clean surface was obtained.

7.4. Clean Mo(110) low-vicinal surface

A typical LEED pattern obtained from the clean surface is shown in figure 7.1 with some of the beams indexed. The diffraction spots are sharp and background intensity is low indicating that the surface is well ordered. No spot smearing similar to that of the vicinal

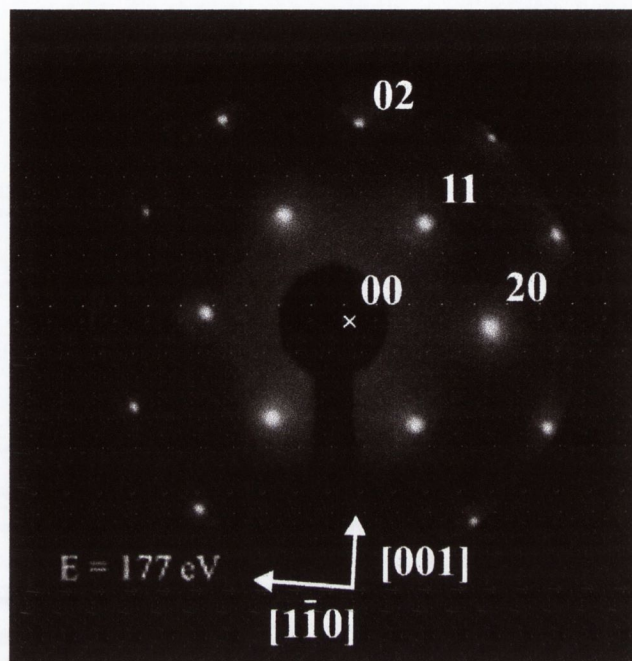


Figure 7.1 LEED pattern of the clean low-index Mo(110) surface. $E_{beam} = 177 \text{ eV}$.

surface (Fig 6.6) has been observed. Following the approach outlined in section 4.3, a molybdenum lattice parameter was calculated for different energies, to an average value of $a_{Mo} = 3.04$ which is 3.7 % lower than the known value of 3.147.

Figure 7.2 shows a globally plane-fitted $1500 \times 1500 \text{ \AA}$ STM image of the clean Mo(110) surface. The surface topography is characterised by a 'staircase' arrangement of terraces of average width $d = 185 \pm 50 \text{ \AA}$ separated by monatomic steps (see figure 7.3). The average value of the step height is measured from the separation between the peaks on histograms of locally plane fitted images, like the one shown in figure 7.4. In this case, the measured step height is $h = 2.13 \text{ \AA}$, which is within 4 % of the value for the unrelaxed surface ($2.22 \text{ \AA}$). The discrepancy can be attributed to the error in the z-calibration of the piezo tube-scanner in the microscope [66]. Taking these values for terrace width and step height and applying them to:

$$\tan \alpha = \frac{h}{d} \quad (7.4)$$

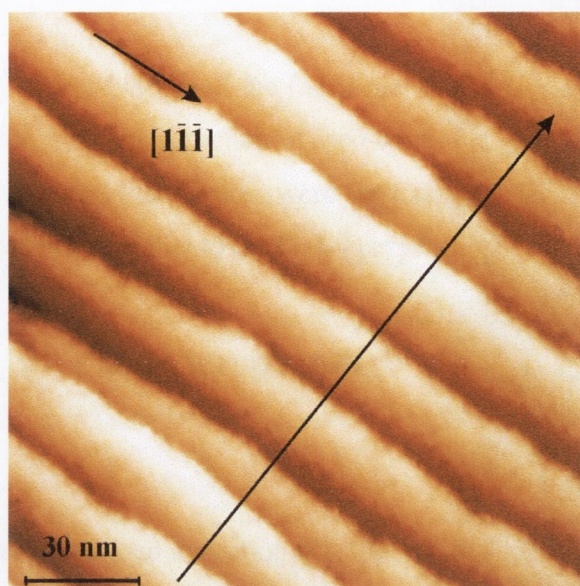


Figure 7.2 $1500 \times 1500 \text{ Å}^2$ STM image of the clean Mo(110) surface with global plane fitting. The arrow marks the location of the line profile shown in figure 7.4 after local plane fitting. The surface topography is characterised by a 'staircase' arrangement of terraces of average width $d = 185 \pm 50 \text{ Å}$ separated by monatomic steps.

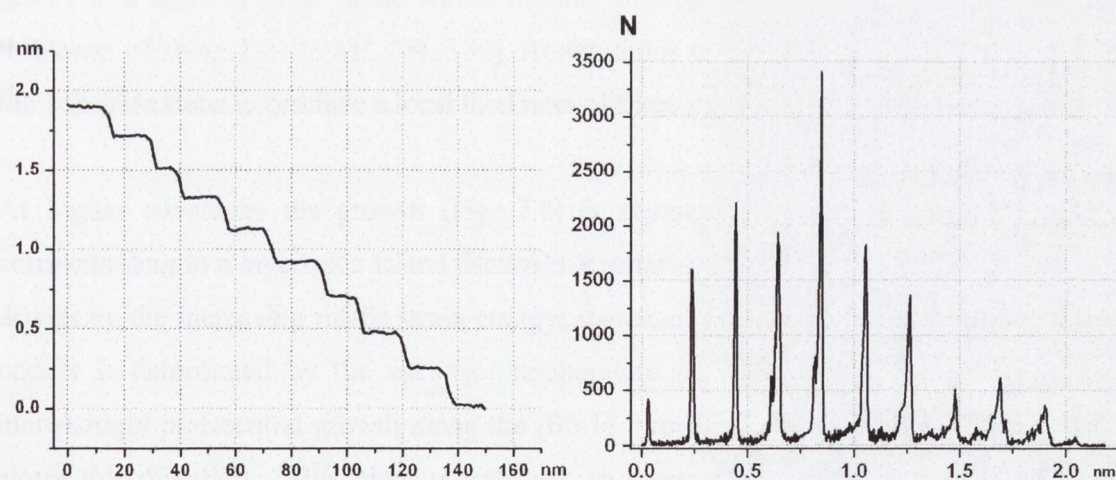


Figure 7.3 (on the left). A line profile of the Mo(110) surface along the direction marked in the Fig. 7.2.

Figure 7.4 (on the right). A histogram of figure 7.2 shows that the step heights are monatomic, with an average value of $h = 2.13 \text{ Å}$.

yields a miscut angle value of 0.66° off the (110) plane. Using a mark on the cap of the sample-holder as a reference, the direction of step-edges in STM images was correlated with LEED data. It was found that terraces were separated by the $[1-1\ 2]$ steps where the step-edges on the surface run parallel to the $[1-1-1]$ crystallographic direction.

7.5. Fe growth at room temperature on low-vicinal Mo(110) surface

Following the procedure described in section 4.2, the Mo(110) surface was cleaned 30-60 minutes prior to deposition. Eight films in a thickness range $1.9\text{ML} \leq \theta \leq 2.8\text{ML}$ ($3.3 - 4.6\ \text{\AA}$) were grown at room temperature with the deposition rate $0.5\text{ML} \cdot \text{min}^{-1} \leq D \leq 0.55\text{ML} \cdot \text{min}^{-1}$. Figure 7.5 shows an $800 \times 800\ \text{\AA}$ STM image of a $3.3\ \text{\AA}$ (1.95 ML) Fe film grown at room temperature with a deposition rate $0.5\ \text{ML} \cdot \text{min}^{-1}$. It is known that the first Fe layer wets the Mo(110) surface, despite the cost in elastic energy, because of the lower surface energy of Fe compared to Mo [124]. The film grows in a layer-by-layer mode with a mixture of step-flow and island growth up to a thickness of about 1.5-1.6 ML [64, 134]. At about this thickness Fe islands start crossing the substrate steps to produce a local thickness of three monolayers.

At higher coverages the growth (Fig. 7.5) is characterised by increasing roughness corresponding to a layer-plus-island (Stranski-Krastanov) growth mode. This transition is driven by the increasing misfit strain energy; the exact coverage at which this transition occurs is determined by the substrate temperature and the deposition rate. There is increasingly preferential growth along the $[00-1]$ direction with an elongation of islands along this direction. This phenomenon was encountered in other bcc (110) epitaxial systems [140, 181, 182] and was attributed to both anisotropic sticking probabilities [181] and the local diffusion behavior at the island edges [182]. Other factors which may contribute to the island shape are anisotropic surface diffusion due to the two-fold symmetry of the bcc (110) surface [183] and stress anisotropy within the film, due to the

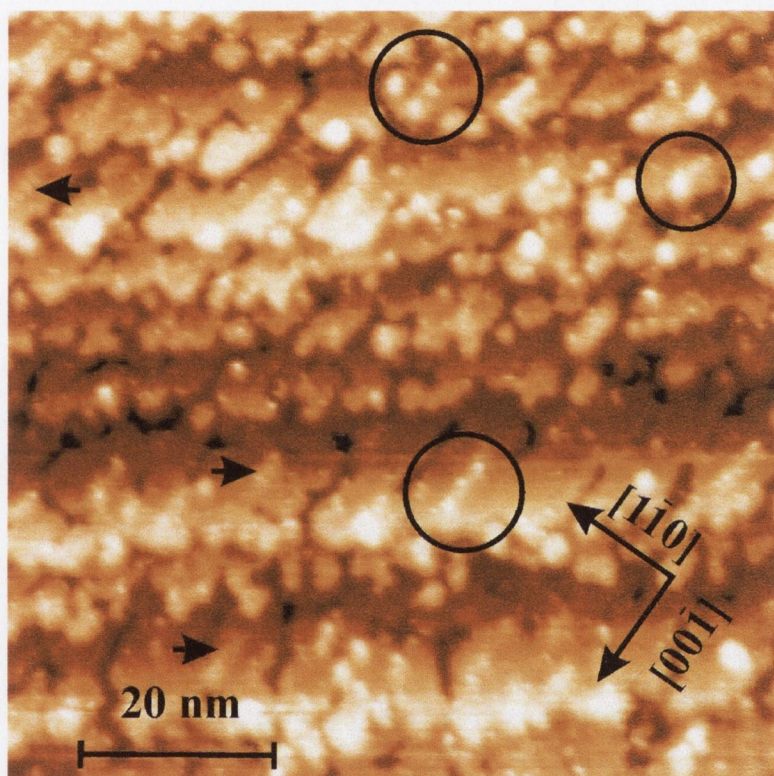


Figure 7.5. $800 \times 800 \text{ \AA}^2$ STM image of a 1.95 ML ($3.3 \text{ \AA}$) Fe film grown at room temperature showing the preferential film growth along $[00\bar{1}]$ crystallographic direction. The dislocations are marked by arrows. Examples of Fe islands growth at the positions of dislocations are circled.

higher cost in elastic energy required to pseudomorphically strain an Fe layer along the $[1-10]$ direction compared to the $[001]$ direction [184].

Because of the comparatively large lattice mismatch between Fe and Mo (9.8 %), only the first layer of the film maintains pseudomorphic registry with the underlying substrate. Linear dislocations (which appear as white lines in the STM image) form along the $[001]$ direction (marked by arrows in Fig. 7.5). Previous LEED and STM studies of the Fe/W(110) and Fe/Mo(110) systems also found evidence of dislocations propagating along the $[001]$ direction [185,186]. These dislocations cause the film to relax along the orthogonal $[1-10]$ direction. The film relaxes along the $[1-10]$ direction first because of the higher cost in energy associated with elastic strain of the film lattice along this direction [187]. These dislocations are formed by the insertion of an extra row of Fe atoms into Fe layer along the $[001]$ direction [64, 134]. These extra Fe atoms cause a relaxation of the interatomic spacing along the $[1-10]$ direction. The lateral offset of the atoms from the pseudomorphic lattice positions along the $[1-10]$ direction establishes a strain field about each dislocation line, which induces an anisotropic diffusion that channels adatoms towards each dislocation. The localised lattice compression at each dislocation facilitates an increased binding energy for the Fe adatoms that enhances the nucleation probability at these defects. As a result, the dislocations form favorable nucleation sites for islands of successive Fe layers (circled in Fig 7.5).

7.6 Elevated temperature growth

To see the effect of the growth conditions on magnetic properties, a 1.95 ML ($3.3 \text{ \AA}$) film was grown at $510 \pm 15 \text{ K}$. The thickness just below 2 ML was chosen because the thicker Fe films deposited at elevated temperatures form large well-separated wedge-shape Fe islands. As was described in section 6.6 the height of these islands can reach a few nanometers and the film cannot be considered as a 2D ferromagnetic system. The first and second Fe layers are formed by the step-flow mechanism, resulting in the formation of uniformly wide Fe stripes along the substrate steps ($[1-1-1]$ direction). An STM image of a nearly completed 2 ML film on Mo(110) surface is shown in Fig. 7.6a. The substrate

terraces are completely covered by a closed pseudomorphic monolayer. The stripes of the second Fe layer cover nearly the entire surface of the substrate forming a good experimental approximation of a two-dimensional ferromagnetic system. The first and second Fe layers can be distinguished in the STM images by the presence of fractional steps that run along the $[1-1-1]$ direction. These steps are marked with arrows in Fig. 7.6 (a, b). A similar effect was observed by STM on clean Fe films deposited on the W(110) surface [128,132,188]. This fractional step is either due to a difference in the local density of states or is a result of pseudomorphically fitting the smaller Fe atoms (atom radius 140 pm) on top of the larger Mo atoms (atom radius 145 pm). The first Fe layer in both images is defect free, but regular stress-relieving dislocations spaced along $[1-1-1]$ direction by ~ 100 Å are formed along the $[001]$ direction in the second layer. An important feature of this film is that the second Fe layer stripes grow along the sides of the fractional steps but do not cross them on iron grown on a lower step to locally form a third Fe layer. This occurs because of the additional cost in elastic energy associated with the maintaining pseudomorphic growth on top of another Fe layer.

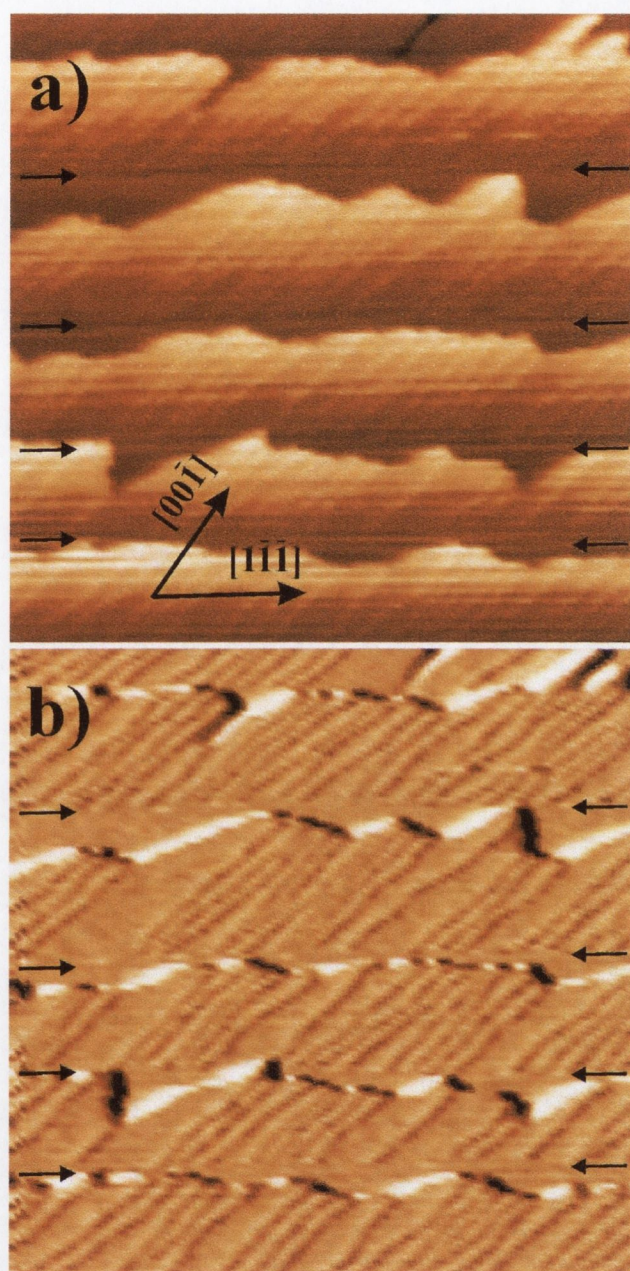


Figure 7.6 Iron stripes formed by step flow on Mo(110) at elevated temperature a) $1000 \times 1000 \text{ \AA}^2$ STM image of a 1.85 ML film grown at $510 \pm 15 \text{ K}$. The first layer is completely closed and free of dislocations. The fractional steps indicating the transition between the first and the second Fe layers are marked by arrows. Periodic dislocations are formed along $[00-1]$ direction in the second layer stripes. b) image (a) differentiated and contrast enhanced to highlight the positions of fractional steps.

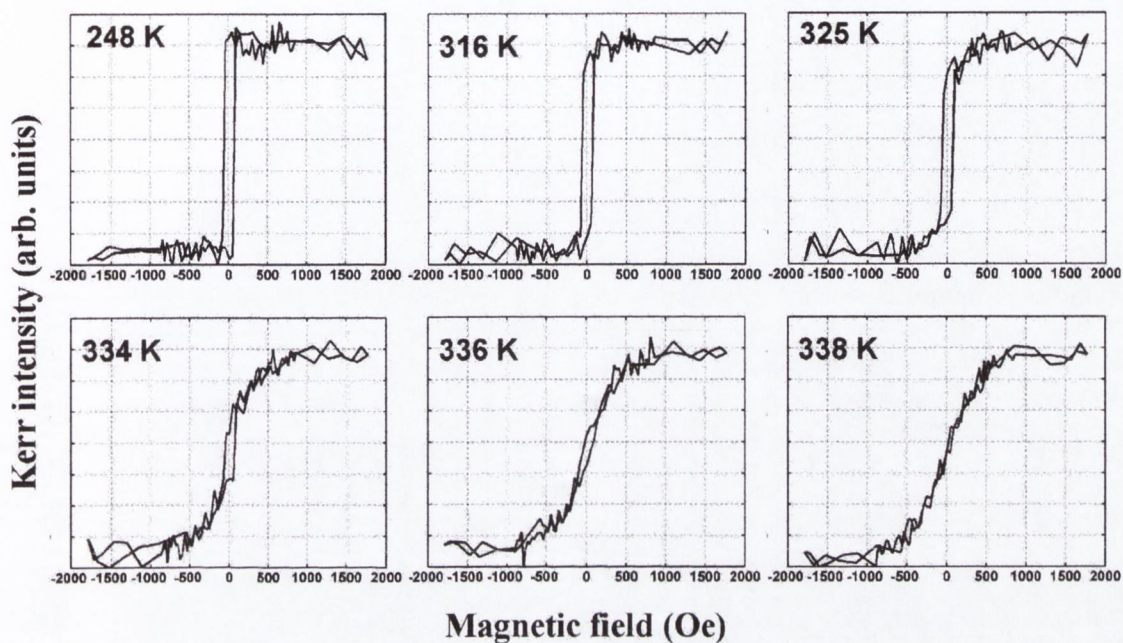
7.7 Magnetic phase transition

The SMOKE technique described in chapter 5 was used to obtain $M(T)$ dependence in order to extract the Curie temperature and effective exponent β_{eff} . First Fe films grown at room temperature were studied. Magnetic hysteresis loops were generated as described in chapter 5.3 by sweeping the magnetic field H and recording the intensity of the reflected light. H was applied along the in-plane [001] crystallographic direction which was found to be the easy magnetic axis for Fe/Mo(110) [180]. Magnetic phase transitions were studied by slowly changing the sample temperature and the recording the detected Kerr intensity in the remanent state.

Longitudinal hysteresis loops recorded at different temperatures for two films of different thickness are shown in Fig. 7.7. The magnetic behaviour of other films is similar aside from a Kerr intensity increase for thicker coverages. Well below the transition temperature, the loop is square with the full remanence indicating a well-established single domain state with the spontaneous magnetisation along the in-plane [001] direction. The remanence decreases with increasing temperature and finally disappears. The temperature at which the remanence vanishes is operationally defined as the Curie temperature and denoted for clarity as T_c' . The thickness dependence of T_c' for the films grown at 300 K is displayed in Fig. 7.8. As expected the Curie temperature is significantly reduced with respect to the bulk value (1043 K for Fe) and decreases monotonically with decreasing Fe coverage from 344 K (2.8 ML) to 249 K (1.95 ML). This reduction is consistent with the loss of the nearest neighbours with decreasing film thickness.

The temperature dependence of the remanent Kerr intensity was recorded in order to study the magnetic phase transition near the Curie temperature. The results for the films grown at room temperature are plotted in Fig. 7.9. All films undergo a second-order magnetic phase transition with the thickness dependent value of T_c . Experimental points in Fig. 7.9 were obtained during the warm-up and cool-down indicating that the transition

2.72 ML



2.36 ML

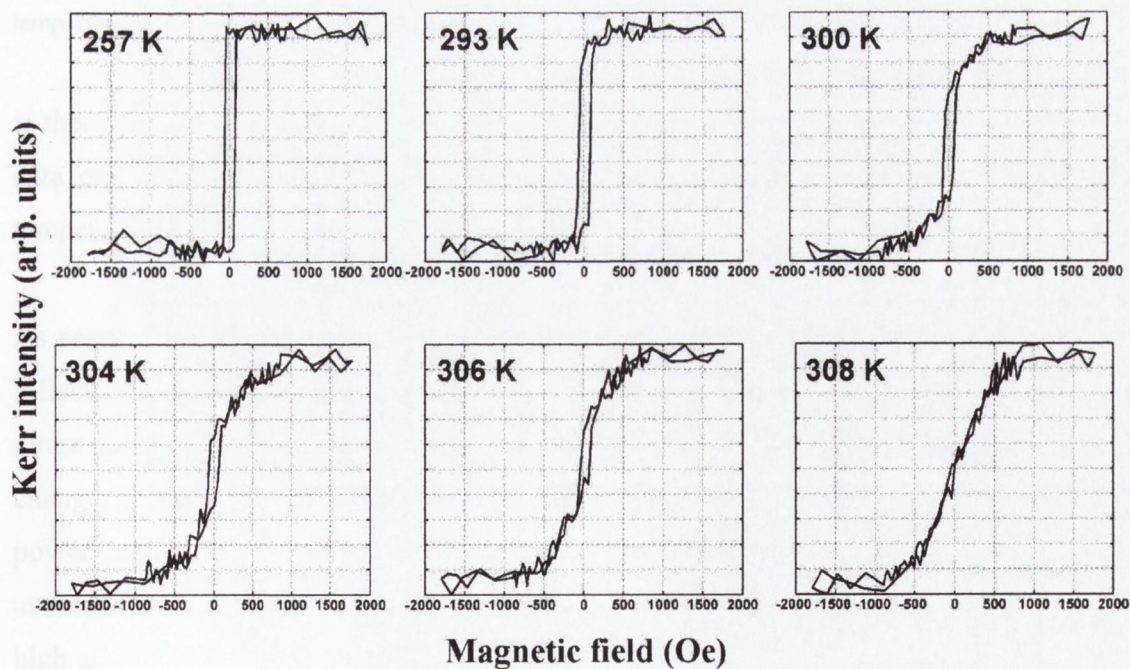


Figure 7.7 Selected longitudinal hysteresis loops measured at indicated temperatures for a 2.72 and 2.36 ML Fe film grown on Mo(110) at room temperature.

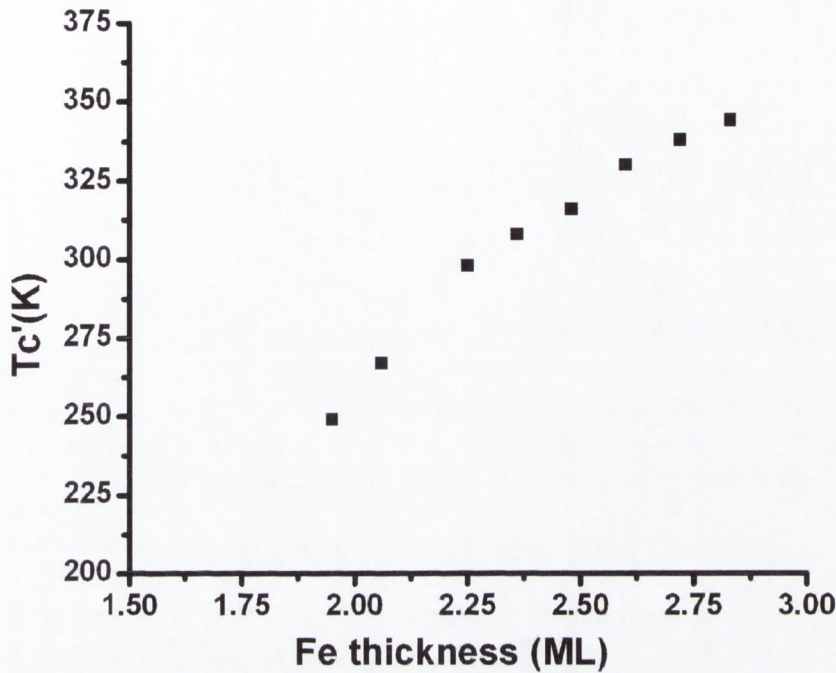


Figure 7.8 Thickness dependence of the Curie temperature T_c' for Fe/Mo(110) films grown at room temperature. T_c' is defined as the temperature at which the remanence vanishes.

is thermally reversible. Since the Kerr intensity is proportional to magnetisation M the data can be fit to a power law given by equation (7.1) in the vicinity of the Curie temperature with T_c and β as fitting parameters.

As seen from Fig. 7.9 the power law fits the data except for a tail observed above the inflection point. This means that M does not follow a power law in the whole temperature range leaving a problem of determining T_c . T_c itself is a fitting parameter and a small change in the assignment of its value affects the value of β . Another problem is that the power law is valid only within a certain temperature region ΔT below T_c . ΔT is not a universal parameter and its choice introduces uncertainty in the data analysis. When a high accuracy is not required, the inflection point is often taken as the Curie temperature of the system; otherwise the T_c value is chosen to maximise the range of a straight line fit in a $\log(M)$ versus $\log(1 - T/T_c)$ plot [164, 189]. The slope of the straight line gives the effective critical exponent of the phase transition. This fitting method is based on a

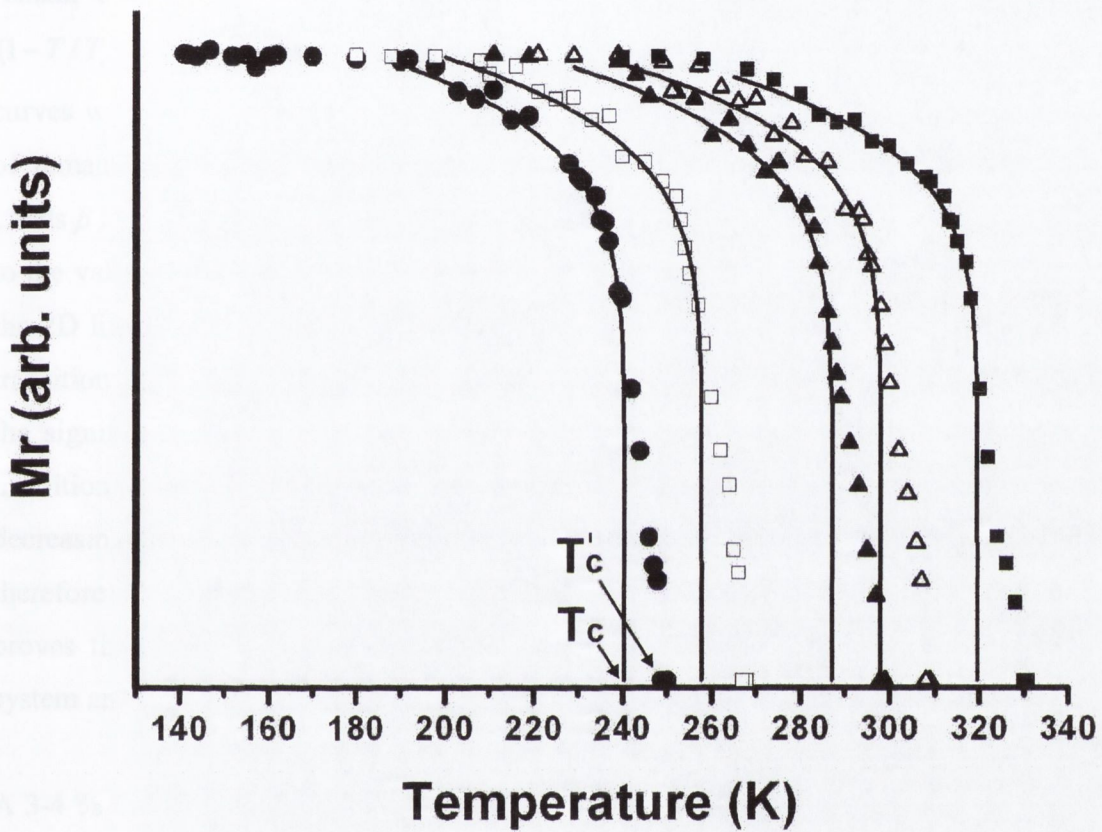


Figure 7.9. Temperature dependence of remanent Kerr intensity for different Fe films deposited on Mo(110) at 300 K. The solid lines show the fit of the data to the theoretical power law $M \propto (1 - T/T_c)^\beta$ with T_c and β used as fitting parameters: 1.95 ML ($\bullet$, $T_c = 240.4 \pm 0.3 K$); 2.06 ML ($\square$, $T_c = 257.4 \pm 0.3 K$); 2.25 ML ($\blacktriangle$, $T_c = 287.4 \pm 0.4 K$); 2.36 ML ($\triangle$, $T_c = 298 \pm 0.4 K$); 2.6 ML ($\blacksquare$, $T_c = 319.5 \pm 0.5 K$).

physical assumption that the system follows the power law up to a certain temperature, above which a deviation occurs due to the finite-size effects. This method is line with the stringent criterion for determining β , which states that the fitting parameters need to remain constant in the asymptotic critical region (the region of *reduced temperature* $(1 - T/T_c)$ where the power law is valid) [190]. The solid lines in Fig. 7.9 are the fitting curves with parameters T_c and β determined as described. Fig. 7.10 shows a log-log plot of remanent magnetisation *versus* reduced temperature. The slope of the straight line yields β in the interval $0.123 - 0.131 \pm 0.004$ for different film thicknesses, which is close to the value of 0.125 given by the Ising model. This agrees with the expectations that in the 2D limit a Heisenberg system with a uniaxial anisotropy reveals an Ising-like phase transition [153, 154]. No trend with the film thickness was recorded for β regardless of the significant difference in T_c , due to the universal character of the magnetic phase transition and its independence of the strength of exchange interaction. T_c decreases with decreasing film thickness due to the reduced average number of nearest neighbours and therefore the total exchange energy per spin. In contrast, the thickness independence of β proves that it is a universal parameter, which depends only on the symmetry of the system and spin dimensionality.

A 3-4 % tail extending above the fitted T_c value appears due to the finite-size effect that limits the correlation length of the system via the film surface defects and atomic steps. Landau demonstrated in his calculations that a finite-size Ising square lattice with both free and periodic boundary conditions reveals a rounded transition with a pronounced tail extending above T_c [191]. The correlation length (i.e. the size of the largest magnetic fluctuation, which does not lose its coherency) is given by equation (7.2). Approximating ξ_0 by a lattice constant a and using $\nu = 1$ of the Ising model we obtain that a 3-4 % tail corresponds to 70 – 100 Å distance between coherence breaking defects. The fact that the thickness of the films is much smaller than the correlation length insures a 2D nature of the system.

The influence of the deposition temperature on the films magnetic properties was studied by investigating the phase transition in two films of the same coverage (1.95 ML) grown

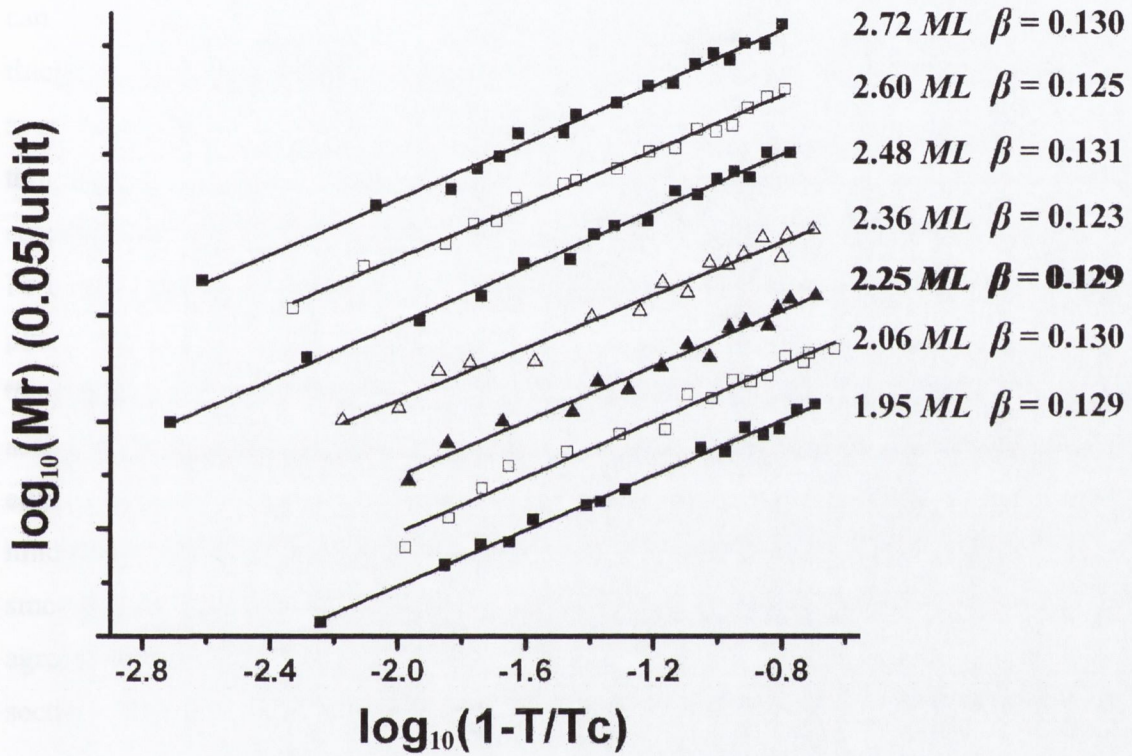


Figure 7.10 Log-log plots of the remanent Kerr intensity *versus* reduced temperature ($1 - T/T_c$) for room temperature grown Fe/Mo(110) films (1.95, 2.06, 2.25, 2.36, 2.48, 2.6 1.95 ML). The data sets are shifted for clarity. The slope of the solid lines gives the effective critical exponent $\beta = 0.123 - 0.131 \pm 0.004$, which is close to the Ising model value of 0.125.

at room temperature and 510 ± 15 K respectively. The temperature dependence of remanent magnetisation and a log-log plot of remanent magnetisation *versus* reduced temperature for these films are displayed in Fig. 7.11. T_c was found to be larger by approximately 30 K for a room temperature grown film compared to the film grown in layer-by-layer fashion at 510 K. Such a dramatic difference in T_c for the same coverage can be explained by the difference in the film morphology. In the film grown at 300 K the thickness locally increases from 2 to 3 ML (as described in chapter 7.5). As a result, the average number of nearest neighbours increases and leads to a larger value of the Curie temperature. In contrast, in the film grown at elevated temperature the second Fe layer stripes grow along the sides of the fractional steps without crossing them to form a third Fe layer. The film morphology was also found to influence the sharpness of the magnetic phase transition. The tail above the fitted T_c is larger for the film deposited at room temperature (3.5 % against 2 % of T_c). The broadening of the phase transition can be assigned to the increased density of the island edges compared to the density of step edges in the film deposited at elevated temperature. The island edges serve as additional hindrances reducing the range of the long-range fluctuations. The tail of 2% found for a smooth film grown at 510 K gives a correlation length of ~ 150 Å, which is in good agreement with the terrace width of 180 Å measured by STM as reported in section 7.4. $\beta = 0.128 \pm 0.003$ was found for the film grown at high temperature providing a further proof of the universal nature of the critical exponent.

The plot of fitted T_c *versus* θ is shown in Fig.7.12. The data below and above the inflection point can be approximated by a linear function. Similar behaviour of $T_c(\theta)$ was previously observed in other ultra-thin film magnetic systems such as Fe/Ag(100) [192] or Co/Cu(100) [193]. A crossover from a power law at higher coverage to linear behaviour at lower coverage was also observed for Ni/Cu(100) [194], Ni/W(110) [195] or Gd/W multilayers [196]. Generally, fitting $T_c(\theta)$ data for linear and power law regions to a single curve given by equation (7.3) leads to an effective exponent λ which is outside of the theoretically predicted range $1 \leq \lambda \leq 2$. Willis and Zhang suggested that this discrepancy can be avoided by fitting $T_c(\theta)$ to different functions above and below some critical thickness N_0 , where N_0 was defined as a “mean range of spin-spin interaction”

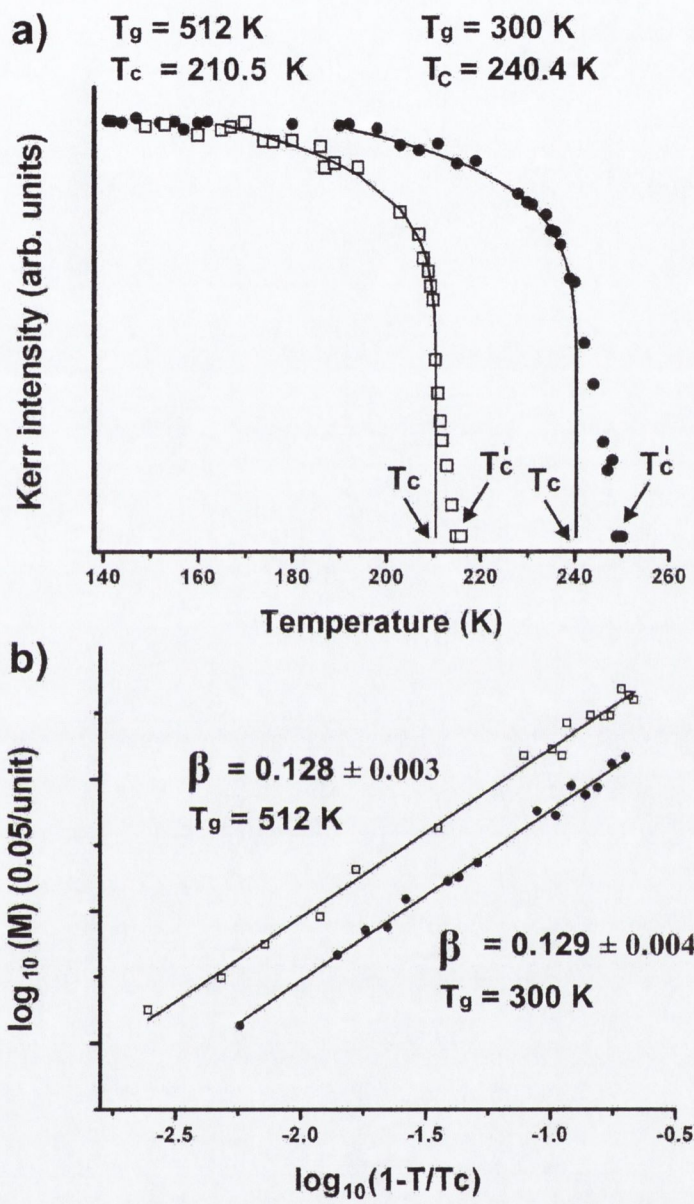


Figure 7.11 a) Remanent Kerr intensity *versus* temperature for 1.95 ML (3.3 Å) Fe films deposited on Mo(110) at room temperature (circles) and at 512 K (squares). The solid lines fit the data to the power law $M \propto (1 - T/T_c)^\beta$ with T_c and β used as fitting parameters. b) Log-log plots of the remanent magnetisation *versus* reduced temperature $(1 - T/T_c)$ for the same films. $\beta = 0.129 \pm 0.004$ and $\beta = 0.128 \pm 0.003$ are determined from the slope of the graph for the room and elevated temperature grown films respectively. T_g denotes the growth temperature and T_c the fitted Curie temperature.

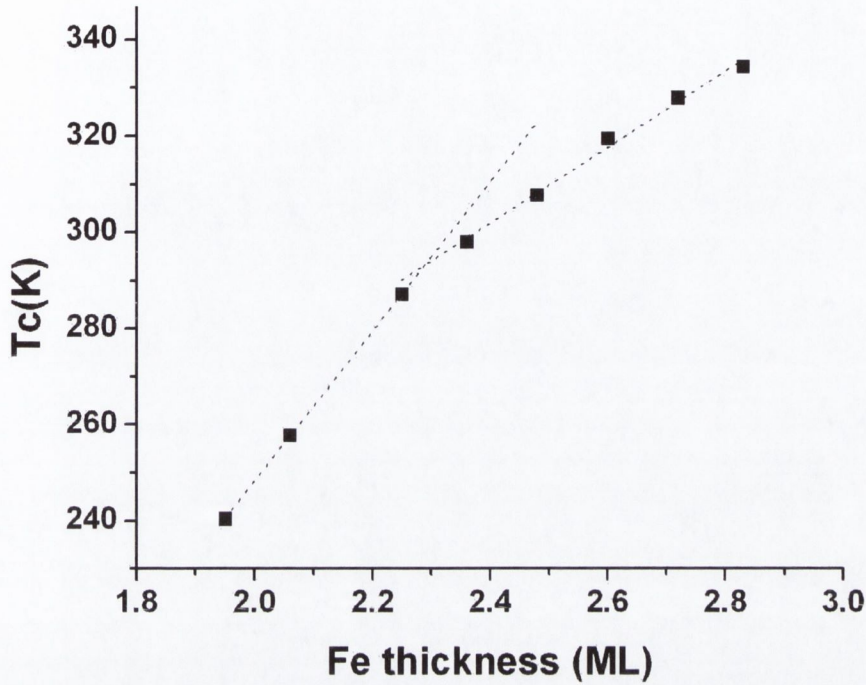


Figure 7.12 Thickness dependence of fitted T_c versus θ for Fe grown at room temperature on Mo(110) surface. The straight lines are the eye guides.

[197]. This model assumes that the spins on lattice sites are coupled to a cluster of identical spins, localised within the radius of interaction of N_0 monolayers (Fig. 7.13). In this approximation, T_c is directly proportional to the number of pair-wise spin-spin interactions N and the energy of these interactions E_0 , which is independent of the distance between the spins within the cluster:

$$k_B T_c = N \cdot E_0 \quad (7.4)$$

where $k_B = 1.38 \cdot 10^{-23} \text{ J/K}$. The Curie temperature is reduced from the bulk value when the number of nearest neighbours and therefore the total exchange energy per spin decreases with the decreasing film thickness. According to this model this reduction is stronger for the coverage below N_0 and $T_c(\theta)$ can be fit to a simple linear function for $\theta < N_0$ from which the range of exchange interaction can be estimated:

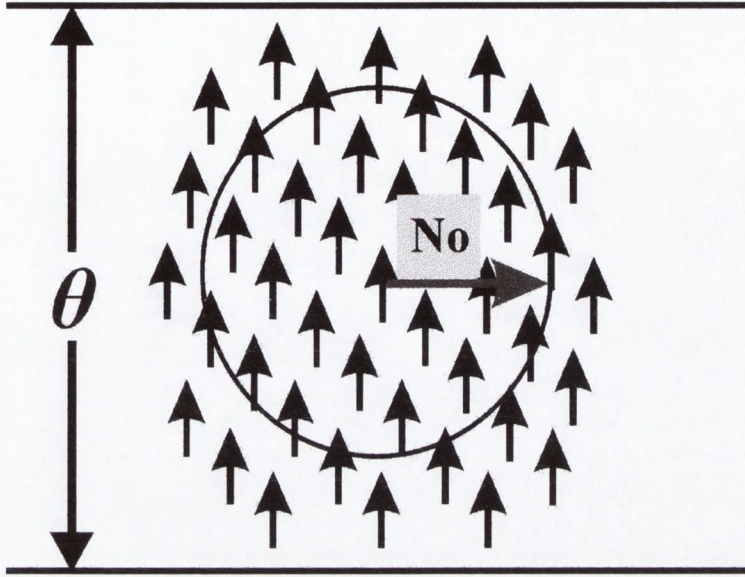


Figure 7.13 Simple model showing the range of spin-spin interactions N_0 (ML) relative to the film coverage θ . Reproduced from reference [197].

$$\frac{T_c(\theta)}{T_c(\infty)} \approx \frac{\theta - 1}{2N_0} \quad (7.5)$$

For $\theta > N_0$, $T_c(\theta)$ needs to be fit to a power law to extract λ , using the value of N_0 found from Eq. (7.5):

$$1 - \frac{T_c(\theta)}{T_c(\infty)} \approx \left[\frac{N_0 + 1}{2\theta} \right]^\lambda \quad (7.6)$$

Apparently, the Fe coverage studied in our experiment is not sufficient to accurately examine the scaling relation given by (7.3) or (7.6). However using the approach described above we obtain $N_0 \approx 2.17$ ML, which is in agreement with other experimental data reporting the average range of exchange interaction for Fe to be close to 2 ML [198]. The inflection of $T_c(\theta)$ curve at $\theta \approx 2$ ML indicating the transition to the power law region confirms the validity of this approach.

Conclusions

A UHV SMOKE setup has been designed and assembled as a part of a larger UHV system for *in-situ* characterisation of structural and magnetic properties of ultrathin magnetic films. The setup is equipped with an electromagnet with four poles arranged in a cross-like geometry capable of producing magnetic fields of up to 2200 Oe for both longitudinal and polar SMOKE configurations. The poles are seated on linear translators so that the size of the gap of the electromagnet is determined only by the size of the sample. A cooling stage, which operates as a continuous flow heat exchanger, allows cooling the samples down to 85 K so that films with the Curie temperature below 290 K can be investigated.

The structural and magnetic properties of Fe films deposited on both high- and low-index Mo(110) surfaces have been investigated using STM, LEED, AES and SMOKE. The LEED and STM analysis of the clean high-index Mo(110) surface showed that the surface was composed of terraces with an average width of 27 Å separated by mostly monatomic steps running parallel to the $[1-1\ 2]$ direction. Fe films in a thickness range $2ML \leq \theta \leq 7.1ML$ were grown at room temperature on the high-index Mo(110) at deposition rates in the range $0.5ML \cdot min^{-1} \leq D \leq 0.55ML \cdot min^{-1}$. The film growth is initiated by the formation of a large number of pseudomorphic monolayer Fe islands with a mean diameter of 10-20 Å merging together to form a pseudomorphic monolayer on the substrate. At higher film thickness a new Fe layer nucleates before the previous layer is fully completed leading to an increased film roughness. The typical staircase pattern of the substrate is maintained, but with a loss in the straightness of the surface steps with increasing thickness. The LEED and STM data shows that the film maintains a one-to-one registry with the Mo substrate. The mismatch strain induced in the small islands is effectively relieved by the inward relaxation of atomic positions at the island edges.

SMOKE hysteresis loops obtained for different orientations of the sample revealed that a two-fold in-plane anisotropy was present in these films with the easy axis along [001] direction. Rectangular loops with unitary M_r/M_s ratio were obtained along the [001]

direction down to 2 ML Fe thickness. Applying a magnetic field nearly along the [1-10] direction produced typical hard axis loops. Similar hard-axis loops were obtained when the magnetic field was applied along the film normal. This result is exactly opposite to the seemingly similar Fe/W(110) system which has its easy axis along the [1-10] direction for the thicknesses below 60 ML. It also contradicts the conclusions of the recent theoretical calculation for single and double Fe layers on Mo(110), predicting that the [1-10] direction is the easy axis for both Fe/W(110) and Fe/Mo(110). The step-induced anisotropy appeared to have the easy axis along the surface step edges. The impact of the step-induced anisotropy on the films' effective anisotropy decreased with increasing film thickness due to the loss of the step straightness during the film growth. The effective anisotropy constant was estimated to be larger than $1.9 \times 10^5 \text{ J/m}^3$. The magnetisation reversal along the easy axis proceeds by a nucleation event followed by a nearly instantaneous propagation of a single domain wall, while it mainly occurs via the homogeneous rotation when the magnetic field is applied along the hard [1-10] direction. When the magnetic field is applied between the [001] and [1-10] directions, the magnetisation reversal is a combination of these two mechanisms.

A significant transformation in the film morphology occurs in Fe films grown at room temperature after annealing in UHV at 750 K. Large well-separated Fe islands are formed, which are supported on a closed pseudomorphic Fe monolayer. Many of these islands are elongated along directions making a $0 - 12^\circ$ angle with the atomic step edges. The preferential growth direction is changed from the [001] direction because the [001] crystallographic axis lies close to the step direction and the high step density provides a large number of high coordination bonding sites which are readily occupied by the Fe atoms. The LEED data shows that the strain induced in the Fe films is effectively relieved in the formed 3D Fe islands. The magnetisation reversal in these Stranski-Krastanov films proceeds by a succession of nucleation/propagation events so the region of magnetisation reversal has a finite width. The in-plane effective anisotropy remains uniaxial with an easy axis of magnetisation lying close to the [001] direction. The coercivity H_c and saturation field H_s increase dramatically compared to the films grown at room temperature due to the large demagnetising field H_d induced in the Fe islands. H_d is larger for the thicker deposited Fe films because of the larger vertical aspect ratios in

the islands produced by the thicker films. Despite the complex island shape, H_d is reasonably well reproduced by the demagnetising field of an approximating rectangular prism. The elongation of the islands at an angle to the [001] direction shifts the easy axis of the effective magnetic anisotropy from [001] crystallographic axis towards the step direction.

Magnetic critical behaviour was investigated in the Fe films grown on a low-index Mo(110) surface. From the LEED and STM analysis it was determined that the surface was composed of terraces with an average width of 185 Å separated by mostly monatomic steps of height 2.13 Å running parallel to the [1-1-1] direction. The miscut angle was estimated to be 0.66° off the (110) plane. Eight films in a thickness range $1.9\text{ML} \leq \theta \leq 2.8\text{ML}$ were grown at room temperature. The growth is characterised by the roughness increasing with the thickness corresponding to a layer-plus-island (Stranski-Krastanov) growth mode. There is increasingly preferential growth along the [00-1] direction with an elongation of islands along this direction. When the Fe islands cross the substrate steps they produce a local thickness of three monolayers. The thickness dependence of the Curie temperature for these films showed that the Curie temperature is significantly reduced with respect to the bulk value (1043 K for Fe) and decreases monotonically with decreasing Fe coverage. The T_c reduction is consistent with the loss of the nearest neighbours with decreasing film thickness. The average range of exchange interaction for Fe was estimated to be $N_0 \approx 2.17\text{ ML}$, from the $T_c(\theta)$ dependence, which is close to other experimental data reporting N_0 close to 2 ML. The temperature dependence of the remanent Kerr intensity was recorded in order to study the magnetic phase transition near the Curie temperature. The T_c value was chosen to maximise the range of a straight line fit in a $\log(M)$ versus $\log(1 - T/T_c)$ plot. The slope of this plot yielded β in the interval $0.123 - 0.131 \pm 0.004$ for different film thicknesses, which is close to the value of 0.125 given by the Ising model. To see the effect of the growth conditions on magnetic properties, two Fe films of the same thickness 1.95 ML were grown at 300 K and 510 K. The film deposited at 510 K was characterised by pure step-flow growth. An important feature is that the second Fe layer stripes in this film grew along the sides of the fractional steps but did not cross them to locally form a third

Fe layer. As a result, an average number of nearest neighbours was reduced and T_c was found to be smaller by ~ 30 K for the film grown in the layer-by-layer fashion at higher temperature. The film morphology was also found to influence the sharpness of the magnetic phase transition. The tail above the fitted T_c , attributed to a finite-size effect, was larger for the film deposited at room temperature (3.5 % against 2 % of T_c). This was explained in terms of increased density of the island edges compared to the density of the atomic step edges. The island edges served as additional hindrances reducing the range of the long-range fluctuations. The tail of 2% found for a smooth film grown at 510 K gives a correlation length of ~ 150 Å, which is in good agreement with the terrace width measured by STM. $\beta = 0.128 \pm 0.003$ was determined for both films providing a further proof of the universal character of the critical exponent β .

Further work

The new SMOKE setup will significantly improve the analytical capabilities of the existing system. By using SMOKE, surface science and molecular beam epitaxy techniques, the correlation between the film structure and magnetic properties can now be investigated for a number of systems such as Fe/SrTiO₃, Fe/Ge, Fe/FeO, Fe/Fe₃O₄/MgO, Ni/Mo *etc.*, which are currently studied in our group. Studies of the Fe/Mo(110) system will also continue. Magnetic anisotropy and critical behaviour of monolayer and submonolayer Fe films on Mo(110) remains unexplored and the effects of gas adsorption on the magnetic properties of Fe/Mo(110) films needs to be investigated. The research will be also extended towards nanostructures with at least one lateral dimension in the nm range. One approach to self-assembling metal nanostructures on the surfaces is the oxidation of metallic alloys. Oxidation of the Mo(110) surface is expected to result in the formation of epitaxial nanowires along the [001] direction [199] due to the anisotropic strain, *i.e.* the lattice parameters of the oxide are well matched to those of the substrate along the wire axis but not along the perpendicular direction. These structures can serve as a template for nucleation of transition metals nanowires. The future studies will be dedicated to the growth, structural and magnetic characterisation of these self-assembled nanostructures.

Appendix A

MOKE / SMOKE chamber

PORT	FLANGE	TUBE OD (mm)	FOCAL POINT	LENTH ± 0.5 (mm)	φ° ± 0.5°	θ° ± 0.5°	USE
1	CF-200-156	156 O.D. × 3.0	A	350	90	0	GATE VALVE
2	CF-114-63	63.5 O.D. × 1.6	A	213	30	33	VIEWPORT
3	CF-70-41	41.2 O.D. × 1.6	A	180	90	45	VIEWPORT or HALL PROBE
4	CF-70-41	41.2 O.D. × 1.6	A	120	270	270	MAGNET POLE
5	CF-70-41	41.2 O.D. × 1.6	A	140	315	270	VIEWPORT
6	CF-70-41	41.2 O.D. × 1.6	A	120	0	90	MAGNET POLE or HALL PROBE
7	CF-70-41	41.2 O.D. × 1.6	A	140	45	90	VIEWPORT
8	CF-70-41	41.2 O.D. × 1.6	A	120	90	90	MAGNET POLE
9	CF-70-41	41.2 O.D. × 1.6	A	140	135	90	ION GAUGE
10	CF-70-41	41.2 O.D. × 1.6	A	120	180	270	MAGNET POLE
11	CF-70-41	41.2 O.D. × 1.6	A	140	225	270	VIEWPORT
12	CF-114-63	63.5 O.D. × 1.6	A	255	280	215	COLD FINGER
13	CF-114-63	63.5 O.D. × 1.6	A	213	30	165	VIEWPORT
14	CF-114-63	63.5 O.D. × 1.6	A	155	90	135	WABBLESTICK
15	CF-70-41	41.2 O.D. × 1.6	A	319	0	169	VIEWPORT
16	CF-70-41	41.2 O.D. × 1.6	B	290	90	180	MAGNETIC DRIVE
17	CF-200-156	156 O.D. × 3.0	C	266	180	0	ION PUMP
18	CF-200-156	156 O.D. × 3.0	C	150	90	135	TURBO PUMP
19	CF-70-41	41.2 O.D. × 1.6	A	160	270	315	SPARE

Table A.1 MOKE/SMOKE chamber port table.

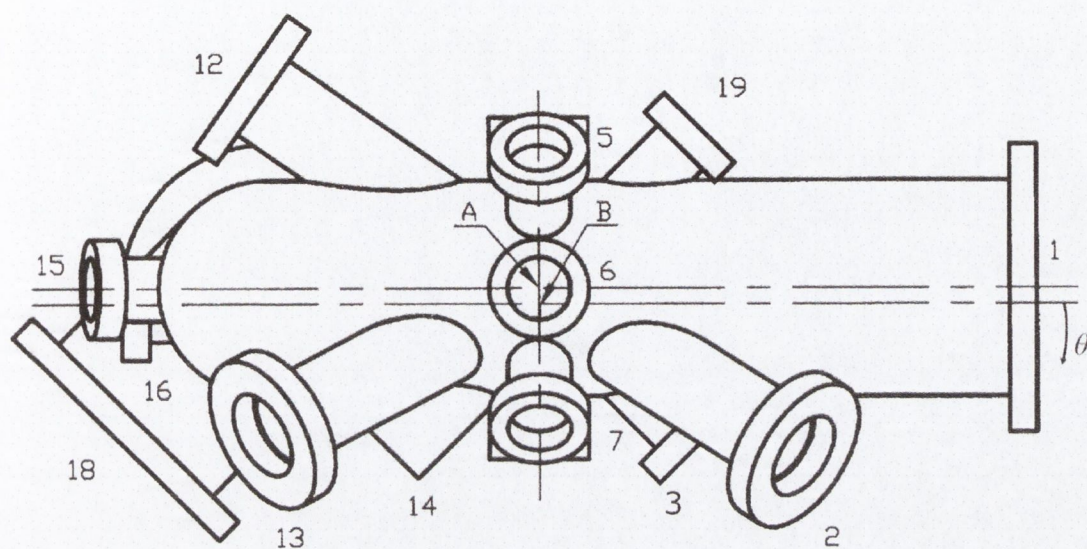


Figure A.1 SMOKE chamber; top view.

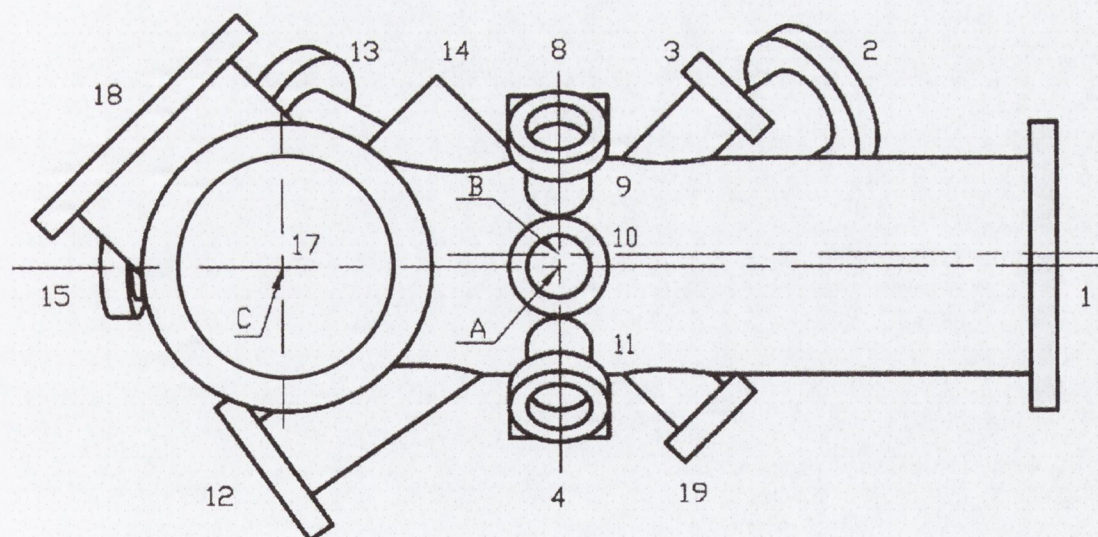


Figure A.2 SMOKE chamber; bottom view.

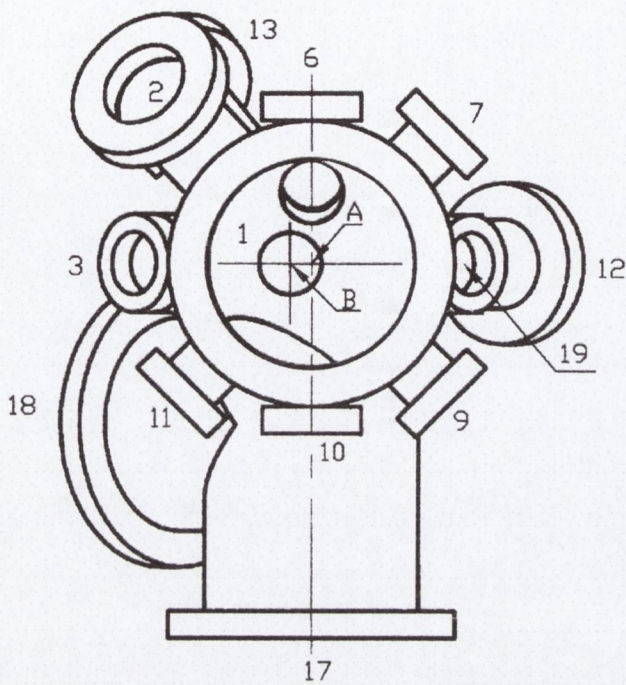


Figure A.3 SMOKE chamber; front view.

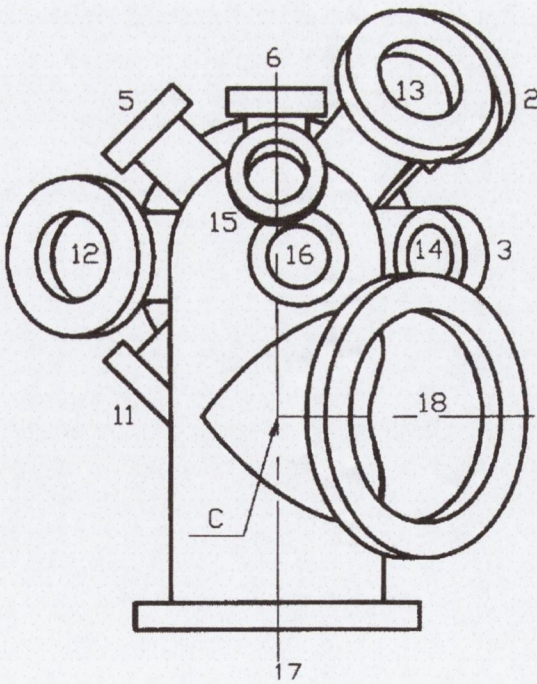


Figure A.4 SMOKE chamber; rear view.

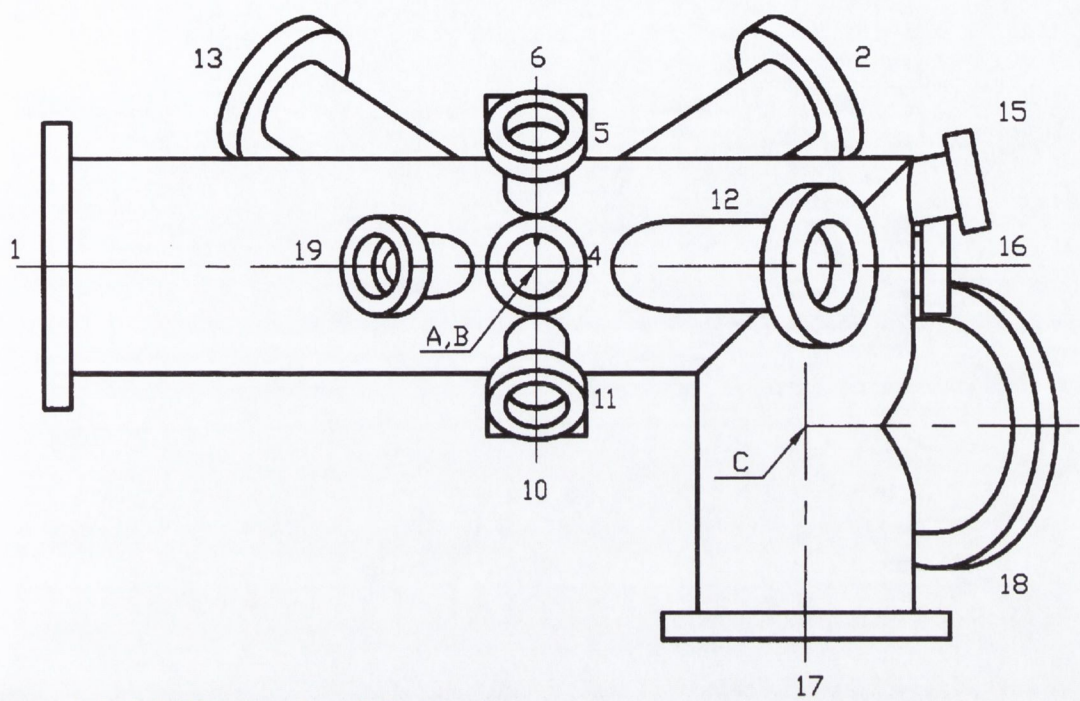


Figure A.5 SMOKE chamber; left side view.

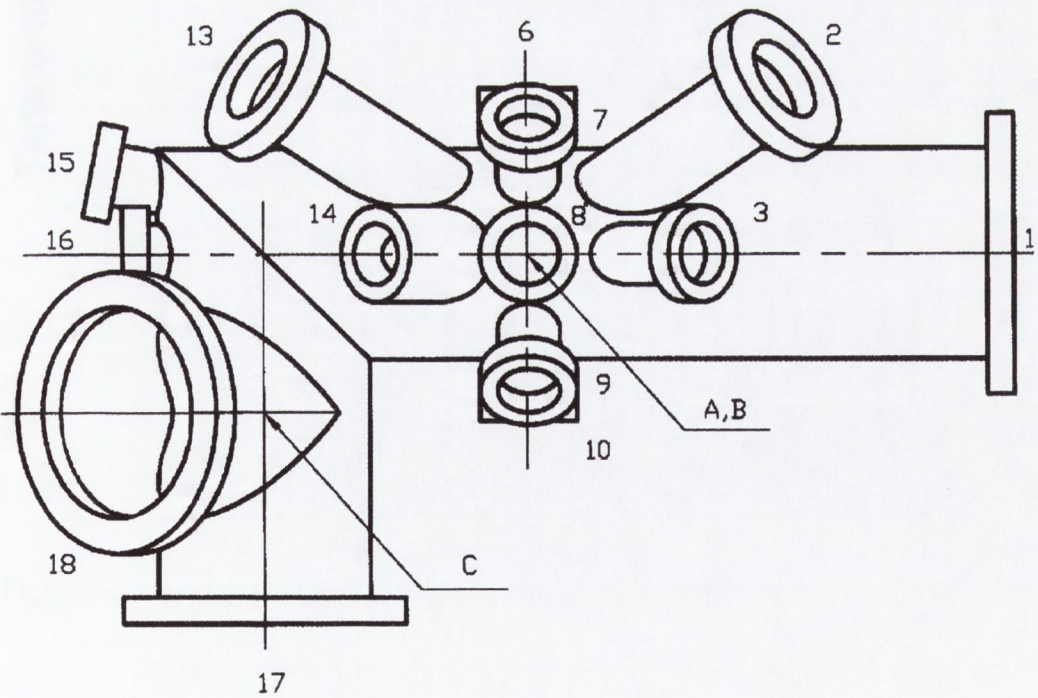


Figure A.6 SMOKE chamber; right side view.

Appendix B

Calibration curves

B.1 Resistive heater calibration curve

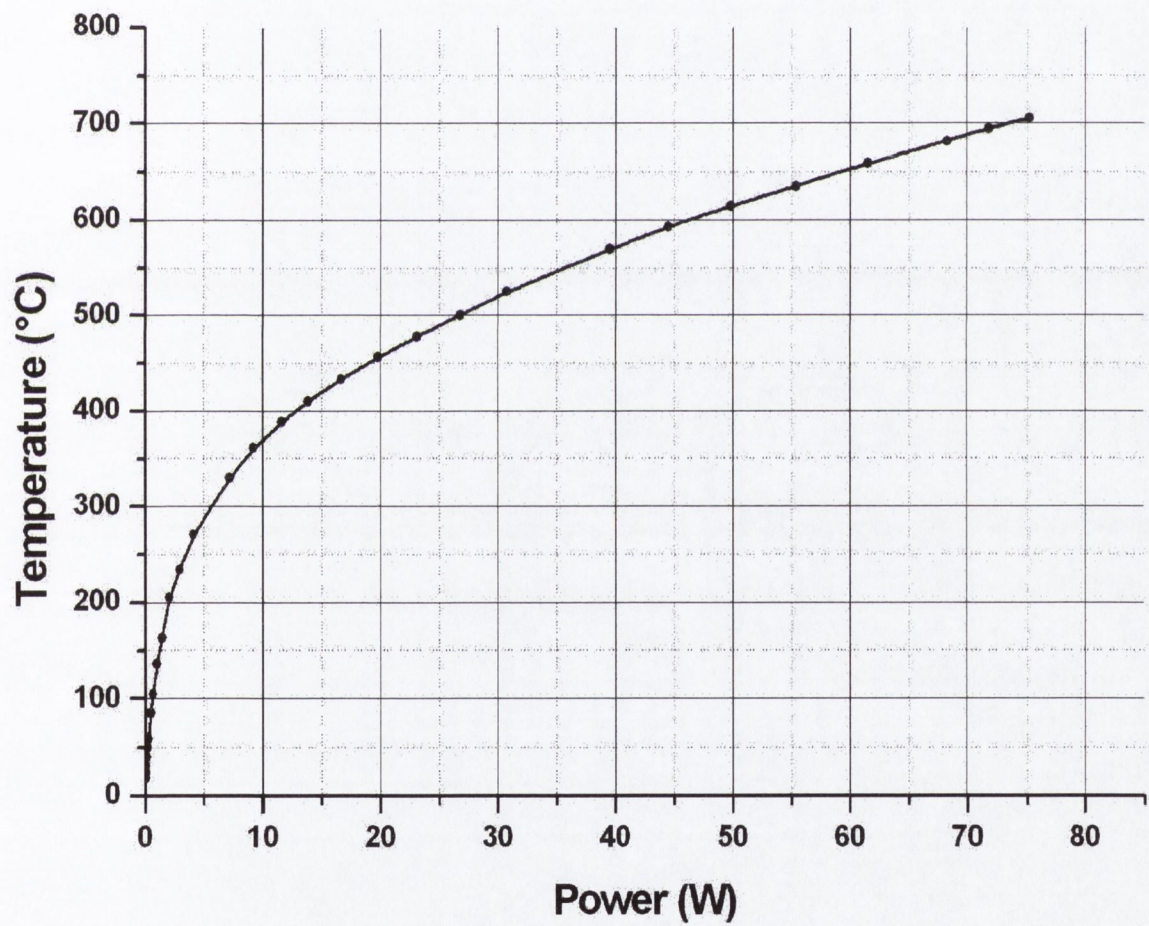


Figure B1 Temperature *versus* power curve for resistive heater.

B.2 Electromagnet calibration curves

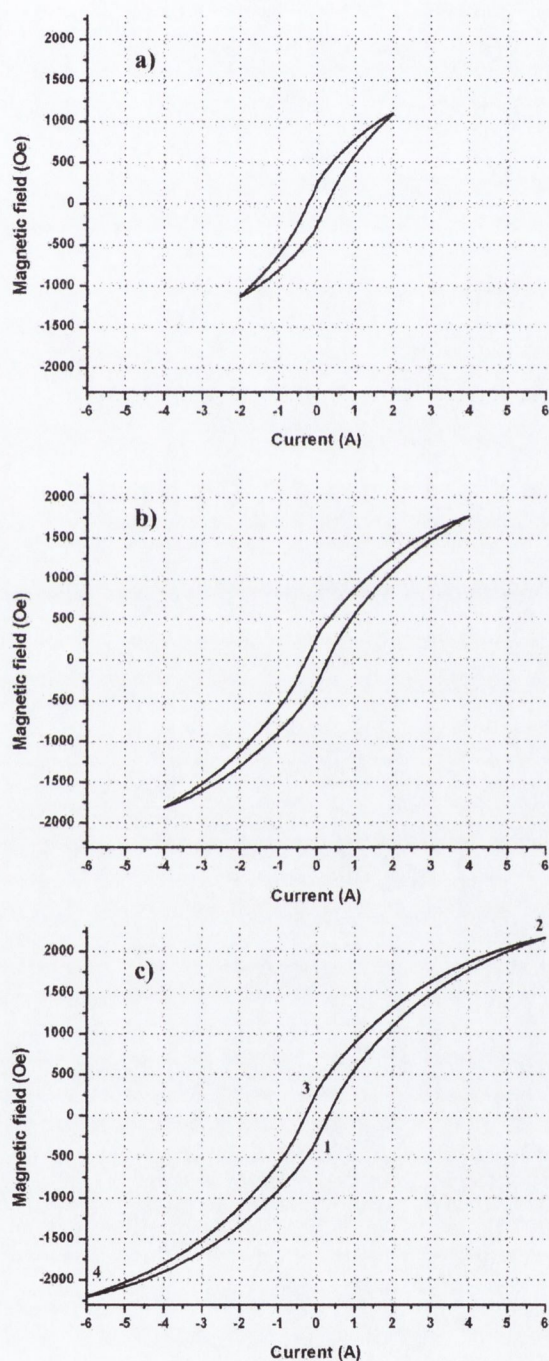


Figure B.2 Electromagnet calibration curves for maximum solenoid current of a) 2 A, b) 4 A and c) 6 A. Magnetic fields obtained for same current values differ due to the coercivity of the magnet core.

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