

ULTRATHIN MAGNETIC FILMS

STRUCTURAL AND MAGNETIC PROPERTIES OF A CHEMICALLY ORDERED MANGANESE ALLOY FILM

By

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Abstract

FeMn has often been successfully used as a pinning layer in exchange bias systems. Ultrathin FeMn films have been studied little, even though the importance of the volume of a data storage system has been increasingly stressed recently. A 4ML Ni/W(110) substrate is used to establish a (slightly strained) (111) f.c.c. template upon which 3ML Fe is deposited and annealed to 580K. LEED, Auger electron spectroscopy (AES) and crystallographic channeling of Auger electrons (Directional AES) confirm that the resulting substrate has very good short and long range f.c.c. (111) order, and is Fe rich at the surface. Mn is grown sequentially to 1.6ML through multiple depositions, with annealing at 580K for 20 seconds between consecutive depositions to maintain the epitaxial structure. LEED and AES reveal that the annealing caused the Fe and Mn to form a chemically ordered FeMn alloy, while Ni is relatively inert to this thermal effect. The magnetism of each alloy film is studied *in situ* by the Kerr effect. The alloy films formed from 0.3 or 0.5ML of Mn have magnetic properties similar to the FeNi substrate. Alloy films formed from 0.8 to 1.6ML of Mn show a marked increase in the width of the susceptibility peak, and develop a slanted hysteresis loop with a reduced coercive field that is consistent with a Mn gradient within the film creating a wide distribution in T_c . This suggests a paramagnetic or antiferromagnetic Mn alloy formation.

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Chapter 1

Introduction

Magnetism has many manifestations in materials studies. The origin of the phenomenon is related to the spin of electrons. People's great interest in it largely arises because the alignment of electron spins exhibit some macroscopic effects that are useful in science and industry. According to this criterion, a magnetic material can be categorized as a disordered state or an ordered state. Disordered magnetic states seem to be more common and have more degrees of freedom. The spins are freed from interactions and don't contribute to detectable magnetization when no field is applied. There are two categories of disordered materials, depending upon how they react to an external field. In a paramagnetic state, the spins tend to align with the external field so that the effective field inside the material will be intensified as a result of the reorganization of the spins. In a diamagnetic state, the spins rotate antiparallel to the external field and the effective field inside the material will be weakened as a result of the reorganization of the spins. In either circumstance, the occurrence of internal magnetization is triggered by an external field.

In contrast, in an ordered magnetic state, spins align spontaneously and thus have fewer degrees of freedom. The alignment of spins provides a very useful representation of information. A very obvious one, already applied in technology, is in binary computation where we regard one direction of magnetization as 1 and the opposite direction as 0. Because of these applications, ordered states have been more widely studied than disordered states. As we look into the microscopic picture of magnetic

ordering, it is a quantum effect. A simple analogy is the singlet and triplet states for a Helium atom. If the singlet, in which two spins are antiparallel, has lower energy, then the total "magnetization" is zero; if the triplet, in which two spins are parallel, has lower energy, then the total "magnetization" is nonzero. The comparison of these two energy states is determined by a complicated wavefunction calculation. Even in such a simple system, the work is nontrivial; a practical magnetic system usually consists of 10^{23} atoms. In this case, people think of some phenomenological models to simplify the many-body interactions. Some models consider spins as classical vectors and the spin interaction as constant. These models sometimes fit the experimental data, such as hysteresis or susceptibility, rather well. Though this strategy is making progress with aid from experimental data and supercomputers, magnetism remains one of the least developed fields in solid state physics.

The two ordered states that correspond to the triplet and singlet atomic states are ferromagnetic and antiferromagnetic. The ferromagnetic (FM) state has its magnetization as order parameter. Most of the spins align, or at least rotate toward an identical direction and the symmetry breaks. Scientists defined ferromagnetism early in the 20th century, and its practical use such as in a compass dates back to before Christ. The counterpart, the antiferromagnetic (AFM) state, has its sublattice magnetization as order parameter. Neighboring spins point to opposite direction. So the symmetry also breaks though the net magnetization is zero. The lack of magnetization greatly disguises the ordering in macroscopic measurements, which accounts for the much later discovery of AFM than FM. Apart from these two states, there are ferrimagnetic, antiferromagnetic and superparamagnetic states.

As an isolated atom, nearly all elements are magnetic. Nonetheless, magnetic solids all contain elements of rare earth or transition metals, with unfilled d or f levels because for other elements they lose their magnetic moments when incorporated into a crystal. The main reason is the delocalization of the electrons due to the overlap of the wavefunctions of neighboring atoms. The delocalization leads to a reduction of the kinetic energy and is an important contribution to the binding energy of the crystals. This affects in particular the outer (s, p)-valence-electrons. As a consequence orbital moments are reduced in a solid. A second reason for the reduction of the orbital

moments is the reduction of symmetry of the potential induced by the presence of neighboring atoms, the so-called crystal-field potential. The orbital moments are strongly reduced or even totally "quenched". While of the main group elements only five are magnetic (Cr, Mn, Fe, Co and Ni), when deposited as thin films on surfaces other elements like V, Ru or Rh can also become magnetic.

The core topic of this thesis is ultrathin magnetic films, with a thickness of only several atomic layers, or monolayers (ML). To address low energy excitations, spin waves with long wavelength are of major concern. In the ultrathin limit, the wavelength exceeds the film thickness and along this direction the magnetic state is uniform. As a result only in-plane degrees of freedom remain and the system closely approaches the physical realization of a two dimensional system. The reduced dimensionality causes many interesting properties. Some ultrathin films are able to follow the behavior of the two-dimensional Ising model; some others have two distinctly ordered states below the Curie temperature, the magnetization reorienting from parallel to perpendicular to the film surface as temperature is lowered. A number of theoretical [5, 6, 7], experimental [8, 9, 10, 11], and numerical studies [12] have been done. Moreover, imagine an ultrathin film with two layers at the surfaces and very few layers in the body. The atoms in the interfaces account for a considerable portion of the total number of atoms. This is distinctive from bulk materials, in which the proportion of interfacial atoms is negligible. It is straightforward to see that interfacial effects are more dominant in ultrathin film than elsewhere. For instance, we have to take the interface energy into account if the film is thinner than $100\text{\AA}$, as proposed by Néel [13]. This peculiar property enables us to study and gain insight into the interfaces of materials. People have studied spin reorientation [14] and exchange bias [15] using ultrathin systems.

Apart from their intriguing magnetic properties, the structure of ultrathin films is also worth exploring. Recently, a novel growing technique has been developed: molecular beam epitaxy (MBE) [16]. The importance of this technique is that certain thin films have the ability to maintain the same structure as the substrate (below a certain thickness). In this pseudomorphic growth mode, the energy increase due to the deviation from the equilibrium bulk structure is compensated by a decrease of

energy due to binding with the substrate. Therefore, this only occurs below a certain thickness. A common example is the deposition of face-centered cubic (FCC) Fe on Ni(111) [17]. The Fe maintains a FCC (111) surface up to a thickness of three ML. Beyond this limit, extra Fe reverts to the bulk structure, BCC (111). Using this technique, exotic materials structure may be realized and the role of structure in determining the intrinsic properties of materials can be studied. Of course, there are numerous ways to manipulate film structures. Other techniques, such as the use of a glass substrate to start amorphous growth [18, 19] or the use of a seed layer to buffer the mismatch between the substrate and films [18, 20], are important and widely-used.

While a majority of the work in magnetic films has concentrated on ferromagnetism, little has been done in antiferromagnetism, especially in the ultrathin limit. Antiferromagnetic thin films are more difficult to fabricate. Unlike ferromagnetism which exists in many elements, antiferromagnetism usually exists in the form of alloys. A common strategy uses co-deposition of several sources to form alloys as-deposited. As the global stoichiometric ratio of each element is achieved by pre-calibration of the evaporator, the local ratio can't be guaranteed. Therefore, in order to ensure an antiferromagnetic state, the antiferromagnetism has to span a wide range of the elemental ratio, like that of FeMn[21]. As in bulk AFM materials, thermal annealing is a convenient method to get homogeneous films, for both sequential deposition and co-deposition. However, many potential antiferromagnetic films use copper as a substrate template to form a face centered cubic (FCC) film structure. Unfortunately copper is easily intermixed with the film at elevated temperatures. Another difficulty of studying antiferromagnetic films is due to the lack of a macroscopic magnetic signal. Techniques typically used to detect AFM, such as neutron scattering, have a small magnetic scattering cross section. The signal to noise (S/N) ratio for a 1nm thin film, as opposed to a 1mm plate, has to be improved by 6 orders of magnitude. One innovative scientist stacked thousands of identical thin antiferromagnetic films to solve the problem.

Previous work in our lab has investigated the structure of an ultrathin Mn alloy film. Some appealing aspects of this film have been discovered. As we know, mag-

netism is usually influenced by the structure, so a question naturally arises: What are the magnetic properties of this alloy film? The basic composition of the film as given by the sequence of deposition of the layers reads $x\text{Mn}/3\text{Fe}/4\text{Ni}/\text{W}(110)$. Due to annealing during fabrication (described in chapter 4) we believe alloys form and the film is better described as $\text{FeMn}/\text{FeNi}/\text{W}(110)$. Certainly, this is not an absolutely accurate formulation since the local ratio of elements is unknown. In bulk and thick films, FeNi is a ferromagnetic material and FeMn is an antiferromagnetic material over a wide range of $\text{Fe}:\text{Mn}$ ratio. If this also applies in the ultrathin limit, we can consider the alloy film above as a potential AFM/FM system in which exchange bias could be realized. Exchange bias studied in sputtered films shows it is an interface effect, but the interface is not well characterized. We are looking for a well characterized interface. Our ultrathin film, approximately 7ML, is thick enough to contain the interface region. A problem might be that it is not as easy to see the exchange bias effect as in sputtered films since a very thin AFM layer can't firmly pin the FM layer (See Sec.2.7). However, searching for exchange bias in this film is still meaningful. We hope to see an exchange bias occurring in a chemically ordered interface. Also, its simple composition meets the requirement for technological use since very thin layers are increasingly important in magnetic devices.

The remainder of this thesis will be organized as follows: in chapter 2, some important theory will be summarized. It guides the experimental work and is indispensable for explaining phenomena; chapter 3 concentrates on a description of the lab facilities and techniques. As film engineering is a very sensitive work, we need special equipment to obtain good data. Then in chapter 4 I will present the progress I have made in the lab, mostly an initial study of a Mn alloy film. Part of the results have been published as a paper. Chapter 5 gives the conclusion and some suggestions for further study.

Chapter 2

Theory

2.1 Introduction

This chapter covers theoretical background that is necessary to understand the thesis. As we analyze a magnetic system, many factors should be taken into account. Some simple models are not quantitatively consistent with experimental data, but can give some qualitative insight. In order to achieve a better understanding, we need to add more contributions to the Hamiltonian, or use quantum mechanics, or use relativistic models instead of classical models. In this thesis, phenomenological models are good options in most cases.

2.2 Heisenberg model

The Heisenberg model is a phenomenological approach to describe the exchange interaction between spin pairs. In principle, this interaction is short range, and consideration of nearest neighbor exchange coupling of spin pairs is enough:

$$H = -J \sum_{i,j}^{NN} \mathbf{S}_i \cdot \mathbf{S}_j \quad (2.1)$$

J is the exchange interaction constant and S is a classical unit vector representing the spin. To show that Eq. 2.1 originates from a quantum effect, or more ex-

actly, the Pauli principle, we consider a two-electron system with a spin-independent Hamiltonian[22]. In the independent electron approximation, the Schrödinger equation takes the form:

$$(h_1 + h_2)\psi(\mathbf{r}_1, \mathbf{r}_2) = E\psi(\mathbf{r}_1, \mathbf{r}_2) \quad (2.2)$$

where

$$h_i = -\frac{\hbar^2}{2m}\nabla_i^2 - \frac{e^2}{|\mathbf{r}_i - \mathbf{R}_1|} - \frac{e^2}{|\mathbf{r}_i - \mathbf{R}_2|}, i = 1, 2. \quad (2.3)$$

$\mathbf{r}_1, \mathbf{r}_2$ are the positions of two electrons and $\mathbf{R}_1, \mathbf{R}_2$ are the positions of two ions which we take to be fixed. Because of the Pauli principle, the wavefunction with the spin part falls into two categories, the singlet

$$\psi_s(\mathbf{r}_1, \mathbf{r}_2) = [\phi_1(\mathbf{r}_1)\phi_2(\mathbf{r}_2) + \phi_1(\mathbf{r}_2)\phi_2(\mathbf{r}_1)]|S = 0, S_z = 0 > \quad (2.4)$$

and the triplet

$$\psi_t(\mathbf{r}_1, \mathbf{r}_2) = [\phi_1(\mathbf{r}_1)\phi_2(\mathbf{r}_2) - \phi_1(\mathbf{r}_2)\phi_2(\mathbf{r}_1)]|S = 1, S_z = -1, 0, 1 > \quad (2.5)$$

where ϕ_i is simply an eigenstate for h_i , S and S_z being the total spin and its z component. The spin wavefunctions are shown Tab.2.1. It is then straightforward to calculate the singlet-triplet splitting

$$E_s - E_t = \frac{(\psi_s, H\psi_s)}{(\psi_s, \psi_s)} - \frac{(\psi_t, H\psi_t)}{(\psi_t, \psi_t)} \quad (2.6)$$

where $H = h_1 + h_2$. In the limit of large spatial separations this splitting can reduce to

$$\begin{aligned} \frac{1}{2}(E_s - E_t) = \int d\mathbf{r}_1 d\mathbf{r}_2 [\phi_1(\mathbf{r}_1)\phi_2(\mathbf{r}_2)] & \left(\frac{e^2}{|\mathbf{r}_1 - \mathbf{r}_2|} + \frac{e^2}{|\mathbf{R}_1 - \mathbf{R}_2|} \right. \\ & \left. - \frac{e^2}{|\mathbf{r}_1 - \mathbf{R}_1|} - \frac{e^2}{|\mathbf{r}_2 - \mathbf{R}_2|} \right) [\phi_2(\mathbf{r}_1)\phi_1(\mathbf{r}_2)] \end{aligned} \quad (2.7)$$

Because this is a matrix element between two states that differ solely through the exchange of the coordinates of the two electrons, the singlet-triplet energy difference is referred to as an exchange-splitting or, when viewed as a source of magnetic interaction, as the exchange interaction.

Since the atomic orbital $\psi_i(\mathbf{r})$ is strongly localized in the neighborhood of $\mathbf{r} = \mathbf{R}_i$, the factors $\phi_1(\mathbf{r}_1)\phi_2(\mathbf{r}_2)$ and $\phi_2(\mathbf{r}_1)\phi_1(\mathbf{r}_2)$ in the integrand of Eq. 2.7 ensure that the

State	S	S_z
$\frac{1}{\sqrt{2}}(\uparrow\downarrow\rangle - \downarrow\uparrow\rangle)$	0	0
$ \uparrow\uparrow\rangle$	1	1
$\frac{1}{\sqrt{2}}(\uparrow\downarrow\rangle + \downarrow\uparrow\rangle)$	1	0
$ \downarrow\downarrow\rangle$	1	-1

Table 2.1: Singlet and triplet wavefunctions for a two-electron system.

singlet-triplet energy splitting will fall off rapidly with the distance $|\mathbf{R}_1 - \mathbf{R}_2|$ between protons.

We see the splitting depends only on the value of the total spin S . Through a simple transformation, we can convert the spin-independent Hamiltonian into an equivalent spin-dependent Hamiltonian

$$H^{spin} = \frac{1}{4}(E_s + 3E_t) - (E_s - E_t)\mathbf{S}_1 \cdot \mathbf{S}_2 \quad (2.8)$$

where $\mathbf{S}_1$ and $\mathbf{S}_2$ are the spins of the two electrons. By redefining the zero energy and letting $J = E_s - E_t$, the Hamiltonian simplifies to

$$H^{spin} = -J\mathbf{S}_1 \cdot \mathbf{S}_2 \quad (2.9)$$

which now looks like Eq. 2.1. When the number of ions N and spin S are large, the system contains $(2S + 1)^N$ different configuration and the spin Hamiltonian becomes exceedingly complicated. What is remarkable, however, is that for many cases of interest the form of the spin Hamiltonian is simply that for the two-spin case, summed over all spin pairs of ions. Similarly, the summation only considers pairs of short distance, which in practice are pairs of nearest neighbors. Sometimes demanding work requires inclusion of the next nearest neighboring pairing or higher.

The Heisenberg Hamiltonian has some intuitive solutions. If J is a positive number, all the spins point to one direction in order to reach the minimum total energy. The spin alignment can exhibit macroscopic magnetization because of the magnetic moment $\boldsymbol{\mu} = -g\mu_B\mathbf{S}$ associated with the spin. Then the ground state is a ferromagnetic state. If J is a negative number, neighboring spins point to opposite directions

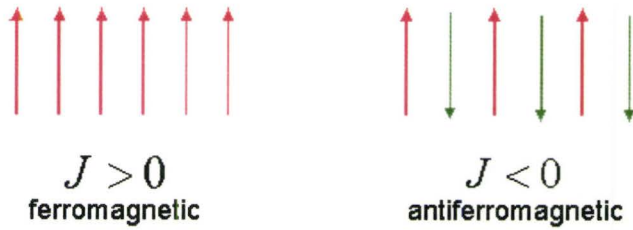


Figure 2.1: The ground states for a one dimension magnet in Heisenberg model when J is a positive or negative constant.

in order to reach the minimum total energy. Then the ground state is an antiferromagnetic state and there is no macroscopic magnetization. The Heisenberg model is very useful in studying the above two states in statistical physics.

However, one difficulty arises if we go through the Hamiltonian carefully. It is an isotropic model which means all directions are equivalent. The spins can align in any direction as long as they meet the minimum energy requirement. Qualitatively speaking, the system tends to fluctuate between alignment directions. So some systems that are described by a bare Heisenberg model can't realize ordering in spite of satisfying a minimum energy condition. The Mermin-Wagner theorem [5] pointed out that there is no spontaneous long-range ferromagnetic order for isotropic Heisenberg models with a short-range interaction ($\sum_j J_{ij} r_{ij}^2 < \infty$). Specifically, one dimensional and two dimensional Heisenberg systems can only achieve ordered states at 0K.

2.3 Anisotropy

Actual magnetic systems are rarely isotropic and we have seen many examples of ordering in one dimension and two dimensions. This is because of the presence of magnetic anisotropy; that is, all directions are not equivalent. Without the effect of magnetic anisotropy, we wouldn't even be able to notice magnetism. A magnetic field would only switch the direction of magnetization instead of imposing a torque on the materials. Almost all applications of magnetic materials depend on the fact that it is more favorable to magnetize a magnetic material along one direction than other. This difference makes the material *magnetically anisotropic*. We usually call the favorable

direction the *easy axis* and the most unfavorable direction the *hard axis*. There are many sources of anisotropy. It can be extrinsic such as an applied field and intrinsic such as dipolar field. Sometimes the anisotropy from different sources can counteract each other and create an effective isotropy. In the ultrathin films studied here, two types of anisotropy are dominant: crystalline anisotropy and shape anisotropy. Their magnitudes are a function of film thickness, shape, structure etc.

2.3.1 Shape anisotropy

The fundamental magnetic phenomenon is the dipolar interaction between magnetic dipoles. Consider two magnetic dipoles $\mathbf{M}_1$ and $\mathbf{M}_2$ separated by the vector $\mathbf{r}$. In classical electrodynamics the dipole energy of the two magnetic moments is:

$$U = \frac{1}{4\pi\mu_0 r^3} [\mathbf{M}_1 \cdot \mathbf{M}_2 - \frac{3}{r^2} (\mathbf{M}_1 \cdot \mathbf{r})(\mathbf{M}_2 \cdot \mathbf{r})] \quad (2.10)$$

where μ_0 is the permeability of vacuum. In a ferromagnet, this expression depends only on the angle between $\mathbf{r}$ and the direction of magnetization. The simplest case is a monoatomic chain, in which we immediately see that spins prefer an orientation of the chain direction. It agrees with our common sense as a compass or a magnetized needle always has its poles at tails. In a thin film with magnetization perpendicular to the film, $\mathbf{M} \cdot \mathbf{r} = 0$ and antiparallel moments are favored. The total effect of the dipolar field then creates striped domains that align antiferromagnetically.

In a ferromagnetic material, all the magnetic moments build up a dipolar field. As this field usually acts against ferromagnetic ordering, it is also called the demagnetization field. If the material has a uniform magnetization, the field in the material is proportional to the magnetization, however not necessarily uniform. It is only in the case of an ellipsoid that the demagnetization field becomes uniform for a uniform distribution of magnetization[23, 24]. Then the proportionality is expressed by N_x, N_y, N_z for the magnetization components along the three principal directions:

$$\mathbf{H}_D = -\frac{1}{\mu_0} (N_x \mathbf{M}_x + N_y \mathbf{M}_y + N_z \mathbf{M}_z) \quad (2.11)$$

N_x, N_y, N_z are called demagnetization factors. They are determined by the shape of the materials. Some examples for special shapes are listed in table 2.3.1

Geometry	N_x	N_y	N_z
sphere ($x = y = z$)	$\frac{1}{3}$	$\frac{1}{3}$	$\frac{1}{3}$
flat plate ($x = y = \infty, z = 0$)	0	0	1
long cylinder ($x = y, z = \infty$)	$\frac{1}{2}$	$\frac{1}{2}$	0

Table 2.2: Table of demagnetization factors for different geometries

We can see that the dipolar field is strikingly influenced by the shape of the ferromagnetic material. In an irregular bulk ferromagnetic specimen, the dipolar field is determined by how the finite size body is bounded by its surface. The effect of the dipolar field is therefore termed shape anisotropy. Only considering one pair of spins, the dipole effect is far smaller than the exchange interaction. But the dipole interaction is a long range effect which decays much more slowly than the exchange interaction. Any spin interacts with many more spins through the dipole interaction than through the exchange interaction. In the film or the bulk regime, it has comparable impact on the magnetic configuration [4].

2.3.2 Crystalline anisotropy

Spherical samples, which bear no anisotropy from the dipole interaction, still show easy and hard axes. This effect comes from the relative orientation between the magnetization and the crystal lattice. For the cubic magnetic material iron, the easy axis is the cubic axis. And for a tetragonal or hexagonal magnetic material, *e.g.* cobalt, the easy axis is the *c* direction. Crystalline anisotropy (or crystal magnetic anisotropy or magnetocrystalline anisotropy) originates from strong coupling between electron spin and orbital momenta. It arises in the relativistic Dirac Hamiltonian and contributes a term to the scalar relativistic Hamiltonian of the form[4]

$$[E + eV(r) + \frac{\hbar}{2m} \nabla^2 - \frac{\mu_B}{2mc} \xi(r) \mathbf{L} \cdot \mathbf{S}] \psi = 0 \quad (2.12)$$

where:

$$\xi(r) = \frac{1}{2m^2} \frac{1}{r} \frac{\partial V}{\partial r} \quad (2.13)$$

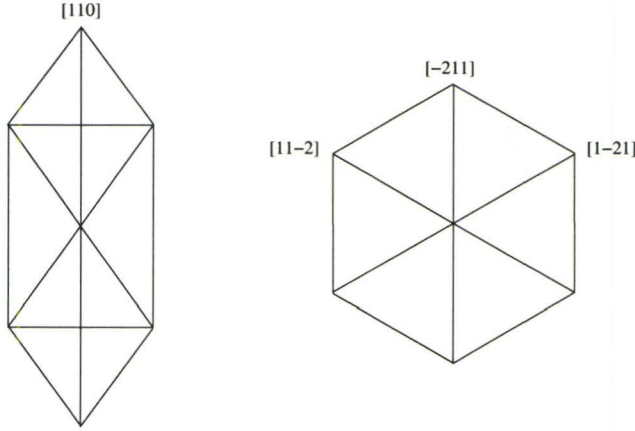


Figure 2.2: Relative orientation for the substrate W (left) and Fe (right) in our films. Unstrained FCC (111) Fe has three equivalent easy axis: $[-211]$, $[1-21]$, $[11-2]$. An expansion for Ni along the $[110]$ of W ($\text{Fe}[-211]_{\text{FCC}}$) makes it the sole easy axis for our films.

V is the atomic potential between an electron and nucleus. From Eq. 2.13 we expect ξ will be large for heavy atoms but small for lighter ones. The relations above indicate the direction of spin and orbital momenta are interconnected. Electrons in a crystal feel the crystal potential rather than a single atomic potential, so their orbital momentum are related to the crystal symmetry. In a crystal, orbital momentum $\mathbf{L}$ is mostly quenched as explained in chapter 1. Therefore the spin moment, and therefore the total magnetic moment, possesses the symmetry of the lattice structure. On a macroscopic scale the effect is that magnetization has a preferred direction for alignment and deviation from this direction increases the magnetic energy.

For example, cubic materials usually have three equivalent easy axes along cubic directions. In my study, FCC Fe grows on a 4Ni/W(110) substrate. The 4ML Ni is strained due to a mismatch of lattice parameters with that of W. This is especially true of the interface layer, which shows a (7×1) reconstructed LEED pattern [25]. This crystalline strain selects the FCC $[-211]$ direction as the sole easy axis. In this circumstance, crystal anisotropy is a kind of uniaxial anisotropy. Some materials have uniaxial symmetry, such as hexagonal, which exhibits uniaxial anisotropy parallel to the c axis of the crystal at room temperature. As the internal magnetization deviates

from the easy axis at an angle of Φ , the anisotropy energy increases. We can express this energy by expanding it in a series of powers of $\sin^2 \Phi$:

$$E_a = K_{u1} \sin^2 \Phi + K_{u2} \sin^4 \Phi + \dots \quad (2.14)$$

Usually it is sufficient to use the first term only to express the anisotropy energy (Table 2.3.2). The anisotropy energy is also dependent on the azimuthal angle about the c axis, but this term is as small as the third term of Eq. (2.14). We can see from

Class	Material	K_1	K_2	Symmetry
3d Metals	Fe	$5.2 \cdot 10^4$	$-1.8 \cdot 10^4$	cubic
	Ni	$-1.2 \cdot 10^5$	$3.0 \cdot 10^4$	cubic
	Co	$7.0 \cdot 10^5$	$1.8 \cdot 10^5$	uniaxial
4f Metals	Tb	$-5.7 \cdot 10^7$	$-4.6 \cdot 10^6$	uniaxial
	Er	$1.2 \cdot 10^7$	$-3.9 \cdot 10^6$	uniaxial
Hard Magnets	SmCo ₅	$7 \cdot 10^6$	–	uniaxial
	NdCo ₅	$-4 \cdot 10^7$	–	uniaxial

Table 2.3: Magnetic anisotropy constants K_1 and K_2 for some cubic and uniaxial magnetic materials in J/m^3 at T=4.2K [1].

table 2.3.2 that cubic materials generally have smaller K 's than uniaxial systems. 4f metals or its compounds can have rather high anisotropy constants. Inserting the constants of Co into Eq. 2.14 we can calculate the magnetic field that moves the bulk magnetization from easy axis to hard axis, approximately 10^{-1} Tesla. So the system is quite stable under crystalline anisotropy.

2.3.3 Magnetic domains

Because the dipole energy depends upon the shape of the sample, a uniform ferromagnetic or antiferromagnetic state does not give the minimum energy solution. Instead, the film breaks up into many regions, each of which is ferromagnetic or antiferromagnetic, but their alignment is along different directions. These are called magnetic domains. While the domains are not necessarily intercorrelated, there is a region of continuous spin rotation connecting two neighboring domains. The zone

of transition layers between the adjacent domains is called a magnetic domain wall, which was first investigated by F. Bloch [26] in 1932. In a Bloch wall, spin rotation is parallel to the wall surface and in a Néel wall spin rotation is parallel to the film surface. The domain wall width in the absence of external field is given by

$$w \sim \sqrt{J/K} \quad (2.15)$$

where J is the exchange constant and K is the magnetocrystalline constant. We see that exchange interaction prefers thick domain walls while crystalline anisotropy prefers thin ones, both for the sake of energy minimization.

Spin rotation induces extra exchange and magnetocrystalline anisotropy energy. We usually refer to this energy as the energy of a domain wall. There is another extra energy which is mostly seen in dynamics, the pinning energy. It is an energy barrier caused by pinning sites such as defects. In an inhomogeneous film, pinning sites dissipate additional energy while the wall is moving. This effect may change the shape of a hysteresis or a susceptibility curve.

2.4 Irreversible magnetization processes

When a small external field is applied to a ferromagnetic material, the system undergoes a reversible process: that is, after the field is removed, all the magnetic moments return to their original states. However, if the external field grows large and exceeds a certain critical value, the system won't recover to its original state after removal of the field and an irreversible magnetization process has occurred. Irreversible magnetization processes are one of the most striking features of ferromagnetic materials. In a microscopic view, they are a combined effect of irreversible magnetization rotation and irreversible displacement of the domain wall.

2.4.1 Irreversible rotation magnetization

Here we consider materials with a single domain[27]. In Fig. 2.3 an ellipsoid particle has uniform magnetization M_s . The easy axis of the particle, presumably the

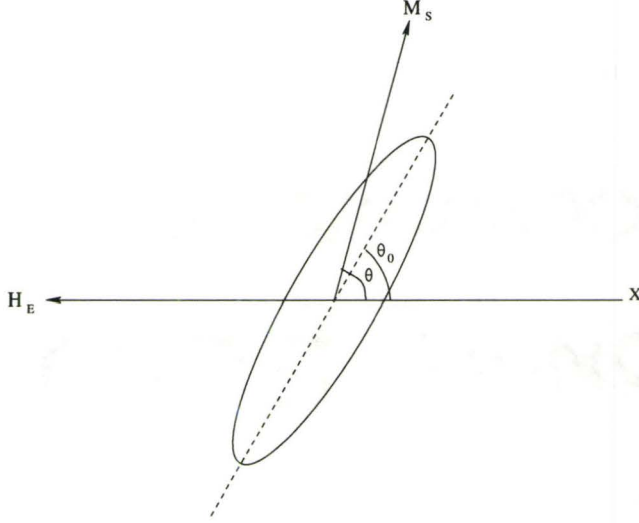


Figure 2.3: An external field drives the magnetization rotation.

major axis of the ellipsoid, makes an angle of θ_0 with the x axis. An external field H_E points along the negative sense of the x axis. Then M_s makes an angle of θ with the x axis in a compromise of the external field and the anisotropy. Considering Eq. 2.14 and energy of the field, the energy of the system is

$$E = -K_u \cos^2(\theta - \theta_0) + M_s H_E \cos \theta \quad (2.16)$$

where K_u is the uniaxial anisotropy constant. The equilibrium direction of M_s is obtained by the extremum energy condition

$$\frac{\partial E}{\partial \theta} = 0 \quad (2.17)$$

and the sign of $\frac{\partial^2 E}{\partial \theta^2}$ determines if the equilibrium is stable. As H_E increases, M_s gradually rotates until the equilibrium becomes unstable at a critical field H_0 when M_s jumps to another equilibrium direction that is stable. At this instant, it must be that

$$\frac{\partial^2 E}{\partial \theta^2} = 0 \quad (2.18)$$

By solving Eq. 2.16, 2.17, 2.18, we get

$$\sin \theta = \sqrt{\frac{4 - p^2}{3p^2}}, \cos \theta = \pm 2\sqrt{\frac{p^2 - 1}{3p^2}} \quad (2.19)$$

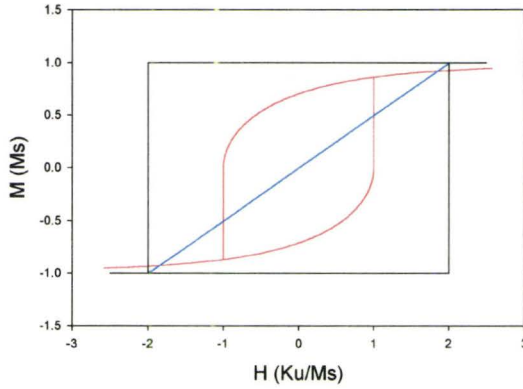


Figure 2.4: Hysteresis loops for a single domain particle calculated from Eq. 2.17 and 2.18. $\theta_0 = 0, 45^\circ, 90^\circ$ for black, red and blue loops.

where $p = \frac{M_s H_0}{K_u}$. A second result coming out of the group of equations is

$$\sin 2\theta_0 = \frac{1}{p^2} \left(\frac{4 - p^2}{3} \right)^{3/2} \quad (2.20)$$

Eq. 2.20, if differentiated, shows the reversal of magnetization takes place most easily when $\theta_0 = 45^\circ$ and the critical field $H_0 = \frac{K_u}{M_s}$. Before H_0 , the magnetization curve is reversible. At the field H_0 , the system undergoes an irreversible magnetization reversal and such a hysteretic effect is illustrated in Fig. 2.4.

2.4.2 Irreversible displacement of the domain wall

Ordinary ferromagnetic materials have domain structures. In such cases irreversible displacement of the domain walls is exclusively responsible for any hysteretic effect. Phenomenologically, the irreversible displacement is attributed to pinning sites that form energy barriers between equilibrium states. These pinning sites can be voids, non-magnetic defects, deformed lattice sites, etc. In the presence of an external field, the wall can overcome these energy barriers. Once the field is removed, the wall is trapped in a new position.

To gain some quantitative insights, let's consider a plane domain wall which travels in the medium where the wall energy fluctuates as shown in Fig. 2.5. If the wall is

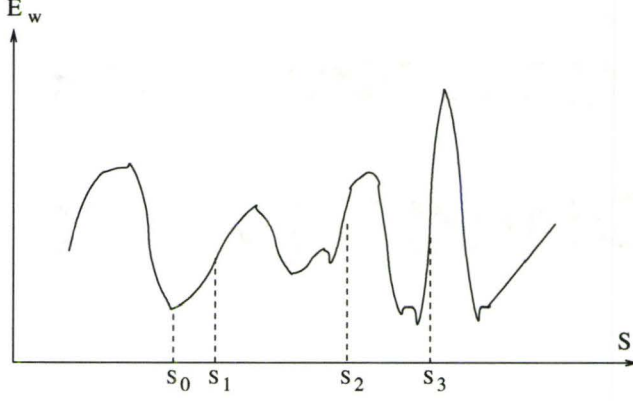


Figure 2.5: Wall energy as a function of wall position for a plane domain wall.

balanced by the external field and inner interaction at position S_i , we must have [27]

$$\left(\frac{\partial E_w}{\partial S}\right)_{S_i} = 2H_E M_s \cos \theta \quad (2.21)$$

where M_s is the magnetization per unit length and θ and $\pi - \theta$ are the angles between the external field and the magnetic moments in the two domains. The free domain stays at S_0 . If an external field is applied such that Eq. 2.21 is satisfied at the point S_1 , then the wall will move to S_1 . Let's further assume that S_1 has a maximum gradient in the neighborhood. Then further increase of the field intensity will result in an irreversible displacement of the wall from S_1 to S_2 where the gradient is larger and thus can counterbalance the field. If the field is reduced, the wall will come back, not to S_0 but to the nearest minimum point to S_2 . If the largest maximum gradient exists at S_3 , the wall will finally reach S_3 and then will discontinuously move to the final goal upon an additional increase of H_E . Thus the critical field for one domain wall is

$$H_0 = \frac{1}{2M_s \cos \theta} \left(\frac{\partial E_w}{\partial S}\right)_{max} \quad (2.22)$$

It turns out that the shape of E_w depends mostly on pinning sites and has little to do with field direction. So H_0 is inversely proportional to $\cos \theta$. Unlike the irreversible rotation magnetization, whose minimum H_0 is at $\theta = 45^\circ$, here H_0 is at $\theta = 0$.

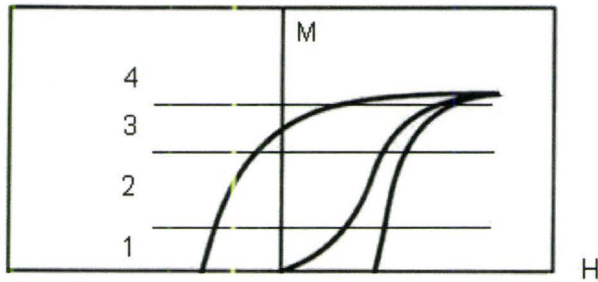


Figure 2.6: Magnetization curve and the classification of magnetization mechanism: 1. Initial permeability range; 2. Range of irreversible magnetization; 3. Range of rotation magnetization; 4. Range of approach to saturation.

2.4.3 Magnetization curves

When a ferromagnetic substance is put into an increasing magnetic field, its magnetization is increased and finally reaches the saturation magnetization. Such a process is called technical magnetization, because it is essentially achieved by a change in the direction of domain magnetization, as opposed to a change in the intensity of spontaneous magnetization. Starting from the demagnetized state ($M = 0$, $H = 0$), the substance undergoes a narrow reversible stage, which is called the initial permeability range (Fig. 2.6). In this range, a major domain wall displacement as well as some domain magnetization, both reversibly, is attributable for the change of the magnetization. As the magnetic field further increases, the system enters the range of irreversible magnetization where the gradient of the magnetization curve increases to greater values. This range is mainly attained by irreversible domain wall displacement from one stable position to another. The irreversible displacement completes with the field increasing and is replaced by the range of rotation magnetization. In this range, the less steep magnetization curve is mainly caused by reversible rotation magnetization. Beyond this range the magnetization gradually approaches the saturation magnetization. The magnetic field gradually perfects the spin alignment. Normally the effect is small and proportional to the intensity of the magnetic field.

The release and reversion of the magnetic field is just an issue of reversing the sequence of above ranges. The total heat generated during one cycle of hysteresis,

which is measured by the area enclosed by a hysteresis loop, is equal to the total energy loss dissipated by discontinuous magnetization processes.

2.5 Critical behavior

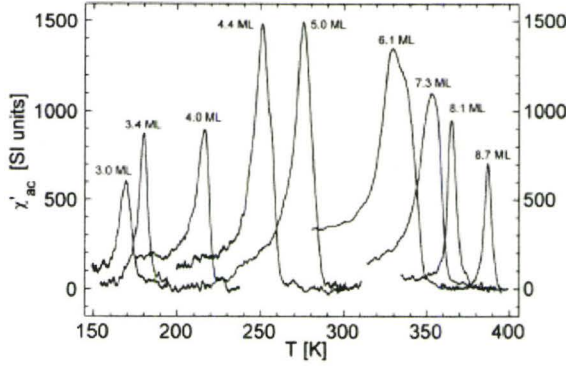
Phase transitions are immensely influential in every corner of the universe—indeed it is widely argued that the very existence of the observable universe is attributable to a phase change in the state of some pre-existing vacuum. In the field of solid state physics, one of the most famous phase transitions is that of a paramagnetic phase to a ferromagnetic or antiferromagnetic phase. The critical temperature that divides two phases is denoted as T_c .

Statistical mechanics is interested in several thermodynamic potentials (such as Gibbs free energy G and Helmholtz free energy F) in the sense that other important quantities can be expressed as derivatives of them[28]. In fact, these potentials all have an analytic part and a singular part¹. In the vicinity of a phase transition, the behavior of the singular part causes some quantities to obey power laws: specific heat c , magnetization M , susceptibility $\chi = (\frac{\partial m}{\partial h})_{h=0}$, correlation length ξ , and the connected two-point correlation function $\Gamma^{(2)}$. The power laws and related critical exponents are given in Tab.2.4. In particular let's consider the case of β and γ . These two values are positive for magnetic materials. As T approaches T_c from below, we expect the magnetization decreases to zero. Far from T_c , a small oscillating field can only wiggle the magnetization at the beginning of the initial permeability range because of a large coercivity of the material. Then the susceptibility is the minute gradient of the magnetization curve at the origin. As the T_c is approached, the coercivity decreases quickly to a point when the magnetization is easily switched by a negligible field. Then the susceptibility becomes much larger. In a real system, χ goes to a sharply peaked value instead of diverging at T_c due to the finite size of the sample (described in the following section), which gives us an accurate way of measuring T_c .

¹By 'singular' we mean non-analytic dependence on the independent variable T .

Exponent	Definition
α	$c \propto T - T_c ^{-\alpha}, T \rightarrow T_c, H_E = 0$
β	$M \propto (T - T_c)^\beta, T \rightarrow T_c \text{ from below}, H_E = 0$
γ	$\chi \propto T - T_c ^{-\gamma}, T \rightarrow T_c, H_E = 0$
δ	$M \propto H_E^{1/\delta}, T = T_c, H_E \rightarrow 0$
η	$\Gamma^{(2)}(r) \propto 1/r^{d-2+\eta}, T = T_c, H_E = 0$
ξ	$\xi \propto T - T_c ^{-\nu}, T \rightarrow T_c, H_E = 0$

Table 2.4: Definition of critical exponents

Figure 2.7: Calibrated in-plane a.c. susceptibility χ'_{ac} versus temperature for several Ni/Cu(001) films of different thickness [2].

2.6 Finite size effect

Conceptually, we know that a finite system cannot have a true singularity at a non-zero temperature. This is very easy to understand in the case of discrete models such as the Ising model in any dimension. Experimental systems, of course, are finite, albeit very large. For all practical purposes, true phase transitions do occur in these finite systems and we have to reconcile this fact with the rigorous proof that no phase transitions can occur in a finite system. This is one of the roles of finite size scaling theory.

Take, for example, a finite discrete magnetic system characterized by the Ising

model. In statistical mechanics the average spin per site takes the form

$$\langle S(\mathbf{r}) \rangle = \frac{\text{Tr}(S(\mathbf{r})e^{-\beta H})}{\text{Tr}(e^{-\beta H})} \quad (2.23)$$

where the Hamiltonian includes an external field term

$$H = -J \sum_{\mathbf{r}, \mathbf{r}'}^{NN} S(\mathbf{r})S(\mathbf{r}') + h \sum_{\mathbf{r}} S(\mathbf{r}) \quad (2.24)$$

A derivative of Eq. 2.23 with respect to the effective field h yields the susceptibility per site in terms of the spin-spin correlation function Γ

$$k_B T \chi(T) = \sum_{\mathbf{r}'} \Gamma(|\mathbf{r} - \mathbf{r}'|) \quad (2.25)$$

The asymptotic form of the correlation function in an infinite system is directly given here without proof[29]

$$\Gamma(|\mathbf{r} - \mathbf{r}'|) \sim \frac{\exp(-|\mathbf{r} - \mathbf{r}'|/\xi(T))}{|\mathbf{r} - \mathbf{r}'|^{d-2+\eta}} \quad (2.26)$$

As a result, the susceptibility will saturate instead of diverging when the correlation length $\xi(T)$ is comparable to the system size.

Bulk ferromagnetic materials or thick films have well defined T_c values depending exclusively on their composition [30]. However, as the film approaches the ultrathin limit the correlation length will saturate before that of an infinite system. T_c is reduced. For a qualitative understanding, a large fraction of the atoms have a smaller coordination number in the ultrathin limit than in the bulk region. The exchange interaction per atom becomes smaller and the ordering is more vulnerable to thermal fluctuation. So the variation of T_c with thickness is a finite size effect. Such an effect was first observed for Ni(111)/Re(0001) [31]. More recently, it has been found from susceptibility curves that increasing the thickness of the ultrathin Ni films increased the T_c [2]. If the thickness is further increased there will be an intermediate crossover region leading to a region where three-dimension T_c is observed.

2.7 Antiferromagnetism in films

The difficulties of detecting antiferromagnetism in films arises mainly from the lack of a macroscopic magnetism. Neutron scattering is useful in thicker films or

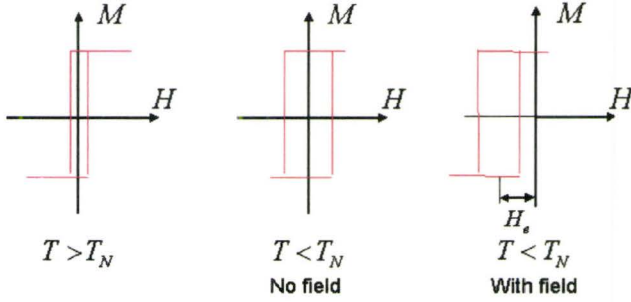


Figure 2.8: Exchange bias may appear when an AFM/FM coupled system is cooled to lower than T_N .

in specimens with a large number of multilayers, but otherwise the signal is very weak. Polarized neutron reflectometry (PNR) supplies a very powerful technique in solving this issue[32]. But the specimen has to be taken out of the ultrahigh vacuum environment which increases technical requirements.

The interaction created at the interface between an AFM and a FM material offers a promising technique for studying antiferromagnetism. When AFM and FM interfaces are cooled through the Néel temperature (normally with an external field), a unidirectional anisotropy for the magnetization of the system will be induced [33]. It causes an increased coercive field and a hysteresis loop shifted from the origin along the applied field axis (Fig. 2.8) which has been used as a signal of appearance of AFM. The size of the shift, that is, the distance between the origin and the center of the hysteresis loop, is usually called exchange bias field (H_e).

Although there have been a number of applications [18, 34], a clear understanding of exchange bias at the microscopic level is still lacking. Some models [35, 36] are able to interpret some experimental data in a quantitative manner. Here, however, I just give an intuitive picture of how exchange bias is formed[3].

When an FM-AFM bilayer is cooled down from $T_N < T < T_C$ with an external "cooling field", the FM spins line up with the field. Due to the exchange coupling at the interface, the AFM spins next to the FM will align ferromagnetically (Fig. 2.9) or antiferromagnetically to those of the FM since $T < T_N$. If the external "cooling field" is removed, the system is trapped in a metastable state, with the interfacial exchange

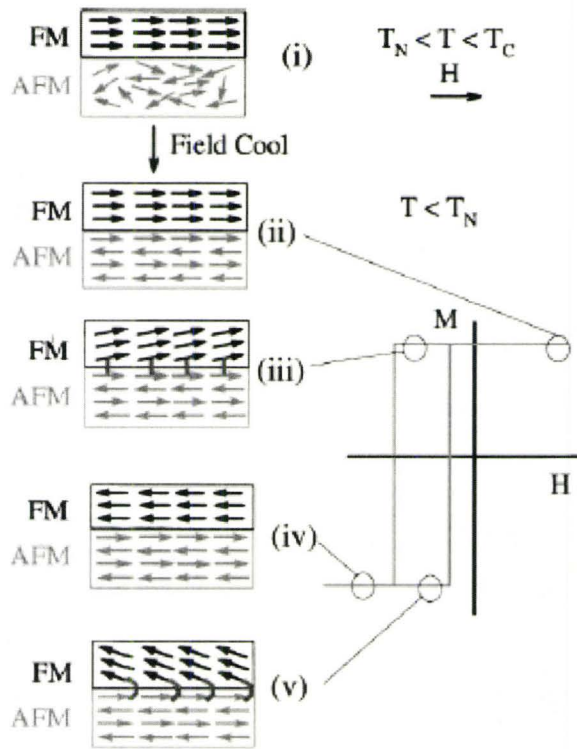


Figure 2.9: Schematic diagram of the spin configuration of an FM-AFM bilayer at different stages [3].

interaction with the AFM creating an effective anisotropy for the ferromagnet. If an external field is used to rotate the FM spin, the AFM spins in the interface will exert a microscopic torque to keep them in the original direction. Therefore, the FM spins have only one single stable configuration, the anisotropy being unidirectional. If we consider the force of AFM on FM as an effective field, this field is equal to H_e .

A model of magnetization rotation predicts that in order to observe exchange bias, the following condition is required [3]

$$K_{AFM}t_{AFM} > J_{INT}n_{INT} \quad (2.27)$$

where K_{AFM} is the anisotropy constant of the AFM with a unit of $J \cdot m^{-3}$, t_{AFM} being its thickness and J_{INT} is the exchange constant between AFM and FM at the interface with a unit of J . n_{INT} is the density of AFM and FM nearest-neighbor coupling pairs at the interface, with a unit of m^{-2} . If Eq. 2.27 can't be satisfied, it is energetically more favorable for the AFM and FM to rotate together. Thus no loop shift should be observed, only an increase in coercivity. Experiments have indicated that exchange bias is roughly inversely proportional to the thickness of the FM layer

$$H_e \propto (t_{FM})^{-1} \quad (2.28)$$

Qualitatively, this is because a thinner FM layer feels a weaker rotation torque from an external field compared with a thicker FM layer. But the pinning torque from the FM/AFM interface is the same. This relation holds for rather thick FM layers (several hundred nm), as long as the thickness is smaller than the FM domain wall size. However, if the FM layer is too thin, the relation is no longer valid, probably because the FM layer becomes discontinuous.

Chapter 3

Experimental methods

This chapter describes the apparatus and techniques that are central to these experiments. As is the case for all the nanomaterials research, the experimental conditions are very demanding. The essential condition is realization of ultrahigh vacuum (UHV). This is obvious because the films under investigation are vulnerable to even minute contamination. After films are grown in the UHV chamber, all the observations and measurements are made *in situ*, using a number of advanced techniques such as low energy electron diffraction (LEED), Auger electron spectroscopy (AES), and the magneto-optical Kerr effect (MOKE). The operation of these apparatus is relatively easy and straightforward compared to an understanding of the underlying mechanisms.

3.1 Apparatus

3.1.1 Ultrahigh vacuum chamber

The UHV chamber, twelve inches in diameter, has a base pressure of 10^{-10} Torr. In such an environment, one monolayer of gaseous contaminant will be accumulated on the substrate in around three hours. The chamber is evacuated through different pumping systems for each different pressure range. The rotary pump makes a preliminary vacuum of 10^{-5} Torr. The chamber is then baked at 150°C overnight

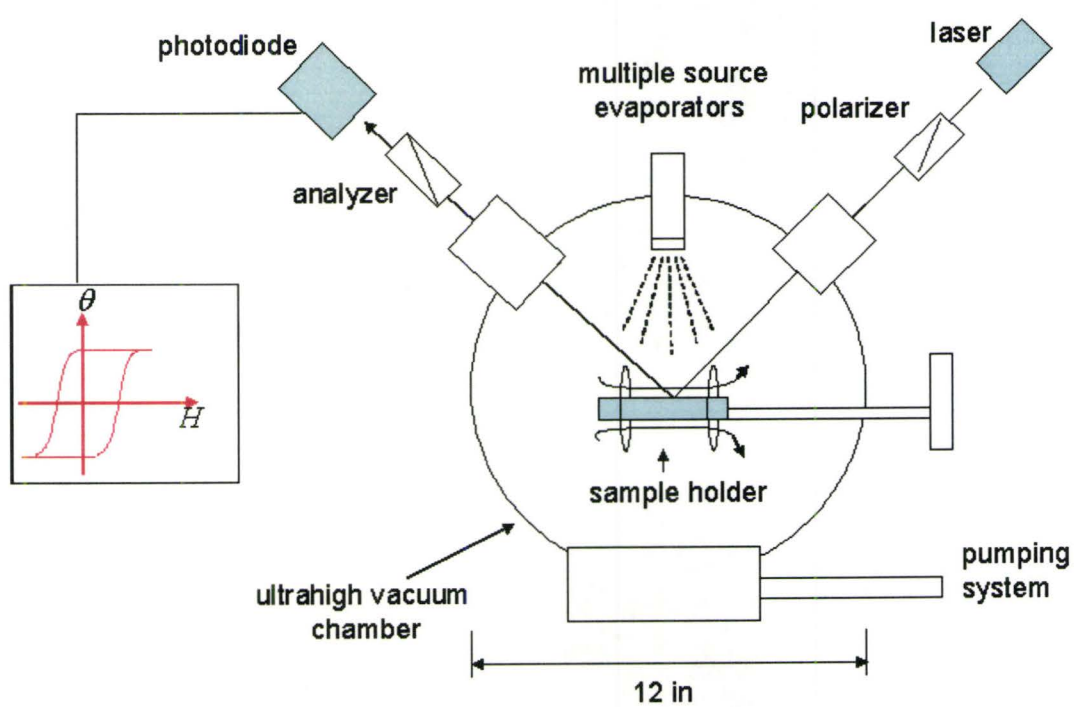


Figure 3.1: A sketch of deposition units.

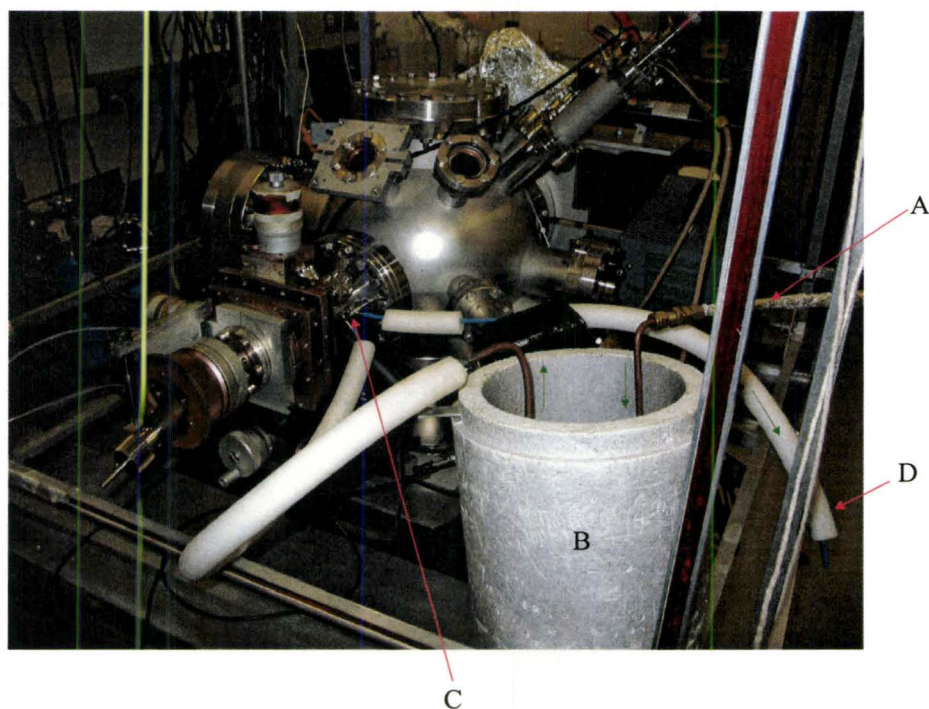


Figure 3.2: Out-of-chamber part of the cooling system. A. Gaseous nitrogen supply pipe; B. Liquid nitrogen dewar; C. Inlet and outlet tube connection; D. Nitrogen exhaust pipe. The green arrows indicate the flowing direction of gaseous nitrogen.

with an ion pump working to 10^{-8} Torr. The post-baking process includes degassing filaments, cleaning the substrate, and flashing a titanium sublimation pump (TSP). In order to obtain the best vacuum, the TSP should be flashed daily.

Some parts inside the chamber are depicted in Fig. 3.1. The substrate upon which films are deposited is a tungsten (110) single crystal. It is mounted on a manipulator which is able to control three translational coordinates and the orientation defined by azimuthal and polar angles. A Helmholtz coil is mounted on the sample holder to supply an in-plane field and a single coil is mounted right under the substrate to supply a field perpendicular to the sample surface. A current supply controls the magnitude of the field. Fig. 3.2 shows a cooling system for the substrate. Gaseous

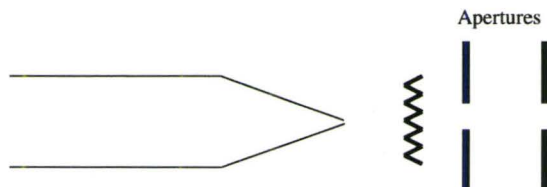


Figure 3.3: Sketch of an electron beam evaporator. From left to right: pure source wire, tungsten filament, first aperture, second aperture.

nitrogen is liquified in a coiled tube immersed in a dewar of liquid nitrogen, and flows into the chamber. The nitrogen flows down to a low-mass, low-volume copper heat exchanger via a small bore coiled capillary tubing. The exchanger, which is mounted adjacent to the sample mounting plate, then cools the sample holder to as low as around 100K.

There are six pyrex windows in the chamber for observation and measurements. Two $2\frac{1}{4}$ in. windows angled at 45° to the horizontal form a optical path for laser light, for the purpose of MOKE. This geometry gives a optimal compromise for using the longitudinal, transverse, or polar MOKE geometry (described in the MOKE section). We also have a mass-spectrometer which enables monitoring contaminant and leak detection with a sensitivity of 10^{-14} Torr.

3.1.2 Evaporators and film growth

Electron beam evaporation

A number of methods for creating atomic beams for film growth have been developed over the evolution of film engineering [37]. For ultrathin film growth, a very low atom flux is required, and during the growing process we need to maintain ultrahigh vacuum. These selective requirements can be well met by the introduction of electron beam evaporators, one of which is shown schematically in Fig. 3.3.

The design is quite simple. A source wire is made of 99.99% pure material. In operation, we apply 1750V to the wire and about 3.5A through a circular tungsten filament in front of it. Electrons emitted by the hot filament are accelerated by the high voltage and hit the tip of the source wire. Due to the bombardment, the

tip melts and source atoms are evaporated. We use two apertures to calibrate the direction and flux of the atomic beam [38]. The evaporated atoms have a chance of being bombarded by electrons while passing the tungsten filament. A certain fraction of the atoms are then ionized. An electrometer measures the ion current intercepted by the second aperture. A bias of -25V applied to the second aperture rejects all the electrons emitted by the filament. The ion current is a linear measure of the atomic flux. By holding the ion current constant, we can achieve a very stable rate of deposition. Also, the rate can be doubled by doubling the ion current. For an ion current of a few nA, the rate of deposition is about 0.1–0.3ML/min, much lower than with sputtering. But the slow process maintains a good vacuum in the chamber and allows a precise control of thickness.

Growth mode

The manner in which atoms are deposited on a substrate to form a film is referred to as the growth mode. Phenomenologically it falls into three categories. The simplest and most favorable mode is Franck-Van der Merwe (FV) under which a film grows layer-by-layer. In other words, growth of the $(N + 1)^{th}$ layer doesn't start until the N^{th} is complete. The films are homogeneous and the thickness is easy to calibrate (described in the Auger spectroscopy section) in this mode. Two other growth modes are less preferred in film engineering, Stranskii-Krastanov (SK) and Volmer-Weber (VW). In SK mode, the second layer doesn't grow until the first is complete. But the growth of second layer and thicker layers are accompanied by formation of tall crystallites, usually with bulk lattice parameters. In VW mode, tall crystallites form at the very beginning of film growth. The latter two modes result in inhomogeneous structure and incur difficulties in thickness calibration and property measurements. Sometimes, by choosing the proper substrate or reducing the substrate temperature during growth, FV growth mode can be realized.

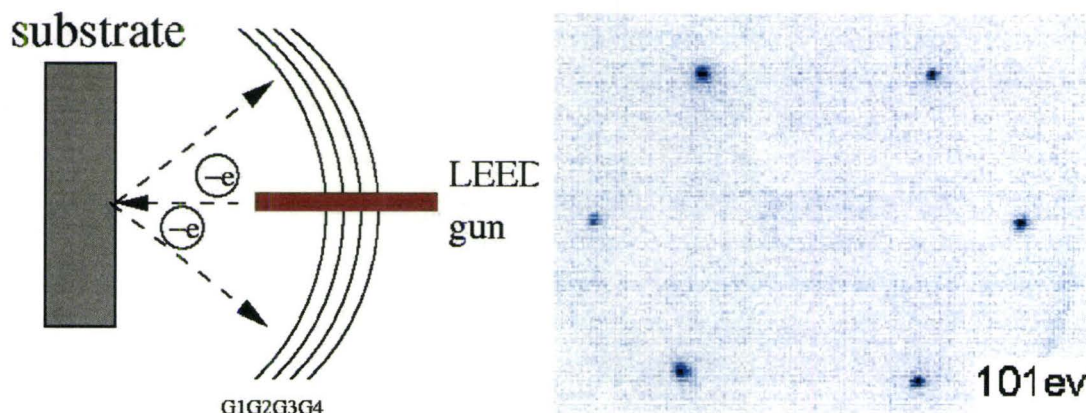


Figure 3.4: LEED working units (left) and a LEED pattern of clean tungsten (110) (right). G1–G4 are four retardation grids.

3.1.3 LEED

Basic knowledge

LEED is a principal technique of materials surface studies. Its setup is shown in Fig. 3.4. A beam of well defined low energy electrons (0–500 eV) are normally incident to a crystal under study. After they are backscattered they go through a set of filtering grids which remove inelastically scattered electrons using a decelerating field. Only elastically scattered electrons survive to reach a fluorescent screen to display a pattern. In order to get an excellent pattern, it is required that the sample be a single crystal with a relatively smooth surface.

The small penetration depth of low energy electrons restricts the sensitivity of LEED to the top few layers of the surface. One can gain qualitative information about the sample, such as its translational symmetry and rotational symmetry and quantitative information such as the surface lattice spacing and the normal interlayer distance [39]. Fig. 3.5 shows an example of a digitalized image. In film engineering, LEED is also used to check the cleanness of a substrate since the growth of contamination seldom follows the structure of a substrate. For instance, an oxygen contaminated tungsten (110) may display a $P(2 \times 2)$ pattern and a carbon contaminated surface may display a complicated pattern with many diffraction spots.

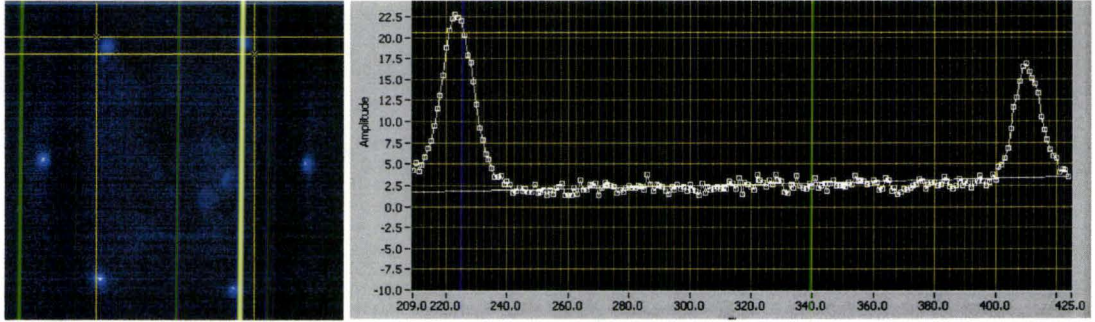


Figure 3.5: Labview program digitalizes the intensity (right graph) of LEED spots (left graph) that are selected by cursors.

Main theory

By the principles of wave-particle duality, the beam of electrons may be equally regarded as a succession of electron waves incident normally on the sample. These waves will be scattered by regions of high localised electron density, i.e. the surface atoms, which can therefore be considered to act as point scatterers.

The wavelength of the electrons is given by the de Broglie relation

$$\lambda = \frac{h}{p} \quad (3.1)$$

Consider, for example, a one dimensional crystal as shown in Fig. 3.6. An electron wave is incident on the chain. The wave scattered by two adjacent atoms and collected at infinity has a path difference of

$$\Delta \ell = a \sin \theta \quad (3.2)$$

The condition of constructive interference requires the path difference be an integer multiple of wavelength λ , which gives

$$\sin \theta = \frac{n\lambda}{a} \quad (3.3)$$

where n is the order of constructive interference. If we generalize the condition of constructive interference in arbitrary dimensions, it is required that[22]

$$\mathbf{d} \cdot (\mathbf{k}_f - \mathbf{k}_i) = 2n\pi \quad (3.4)$$

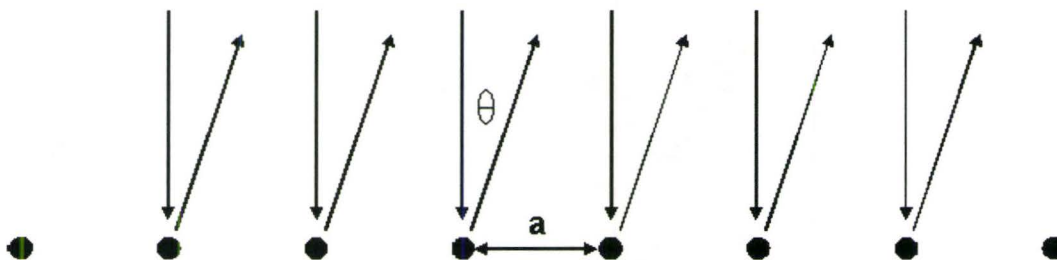


Figure 3.6: A beam of electrons are normally incident on the sample and scattered with an angle of θ .

where $\mathbf{d}$ is a lattice vector of the crystal and $\mathbf{k}_i$, $\mathbf{k}_f$ are wavevectors of incident and scattered electron beams. In our experiments, $\mathbf{k}_i$ is normal to $\mathbf{d}$. So the only requirement is the projection of $\mathbf{k}_f$ in the film plane forms the reciprocal lattice of the film. Then the constructive pattern we see in the LEED screen shows the reciprocal space of the surface layer. In Fig.3.4 the LEED pattern has a b. c. c. (110) plane and the horizontal direction is [001]. This tells us the real surface also has a b. c. c. (110) surface but the horizontal direction is [110] (Fig.2.2), *i.e* the LEED pattern is rotated from the real pattern by an angle of $\frac{\pi}{2}$.

3.1.4 Auger electron spectroscopy

Auger electron spectroscopy was named after a French physicist Pierre Auger who discovered this effect in cloud chambers [40]. After decades of development, it has evolved to be one of the most important techniques in surface science research. The spectroscopy of Auger electrons consists of three stages, described in the following.

Ionization of specimen by high energy electrons

The specimen is exposed to high energy electrons with a primary beam energy of 1-10 keV. These electrons make either elastic or inelastic collisions with target atoms. As their kinetic energy is high, there is a favorable chance that the incident electrons will make an inelastic collision with atoms in which an electron transition occurs. The Auger effect requires that one core electron of the target atom obtains energy from an

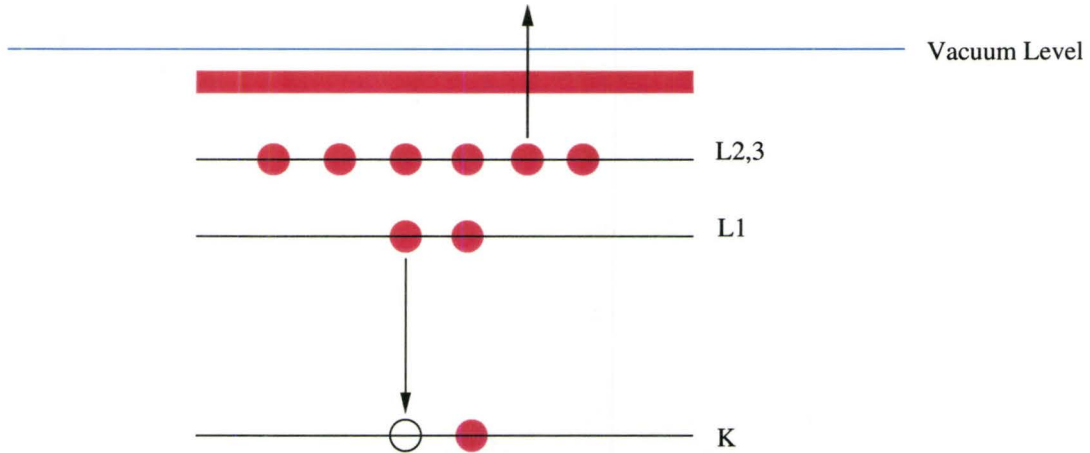


Figure 3.7: Illustration for Auger emission. After transition of two L level electrons there will be two holes in the atom.

incident electron and escapes to the vacuum level, a process called ionization. After this process, the atom displays positive charge for the moment. Usually, the kinetic energy of the primary beam is high enough to ionize all levels of light elements and higher core levels of heavy elements.

In some studies, the initial ionization process is instead carried out using soft x-rays ($h\nu = 1000 - 2000$ eV). In this case, the acronym XAES is sometimes used. As we shall see, however, this change in the method of ionisation has no significant effect on the final Auger spectrum.

Emission of Auger electrons from specimen due to core electron transitions

The ionized atoms are in unstable, excited states and therefore will relax to the ground state by X-ray fluorescence or Auger emission. Here, I will only describe the latter. This process is schematically illustrated in Fig. 3.7.

An electron in the L_1 level jumps to the K level to fill the core hole and gives out energy $\Delta E_{LK} = E_{L1} - E_K$. If another core electron in the L_{23} level receives this energy, it escapes to infinity with kinetic energy of

$$E_{LL} = E_{L23} + E_{L1} - E_K \quad (3.5)$$

It should be clear from this expression that the latter two energy terms could

be interchanged without any effect—i.e. it is actually impossible to say which electron fills the initial core hole and which is ejected as an Auger electron; they are indistinguishable.

An Auger transition is therefore characterized primarily by :

1. The location of the initial hole.
2. The locations of the final two holes.

The illustration shown above is denoted as KL_1L_{23} transition. Since the initial ionization is non-selective the initial hole might be in various shells. It is conceivable that the Auger electrons bear many specific energies across a wide range of energy. Auger spectroscopy measures such a spectrum that shows the intensity of electrons at different energies. Each element has its characteristic spectrum that has several signature peaks.

Signal filtering and analysis

The same apparatus may be used for LEED or retardation analyzer Auger electron spectroscopy. The electron detector is the LEED screen, and the four grids (Fig.3.4) are used to select electrons with energies E . The first grid is grounded to create a field-free region. The 2nd and 3rd grids are placed at energy E , with a small harmonic amplitude of frequency ν superimposed on this potential. The total number of electrons reaching the detector is

$$N(E - \epsilon \sin \nu t) = N(E) - \epsilon \frac{\partial N(E)}{\partial E} \sin \nu t + \frac{1}{2} \epsilon^2 \frac{\partial^2 N(E)}{\partial E^2} \sin^2 \nu t \quad (3.6)$$

As N mainly consists of secondary electrons that have undergone inelastic scattering, the huge and "flat" background easily overwhelms the much weaker, but sharp Auger signals. But the higher order derivatives are dominated by the Auger signals. We use a lock-in amplifier [41] to create the harmonic potential of frequency ν , and measure the collected current at the harmonic of 2ν ($\sin^2 \nu \propto \cos 2\nu$). The second derivative of the spectrum still contains signals from secondary electrons, but it is small and smooth and mainly in the lower energy range (compared with the beam

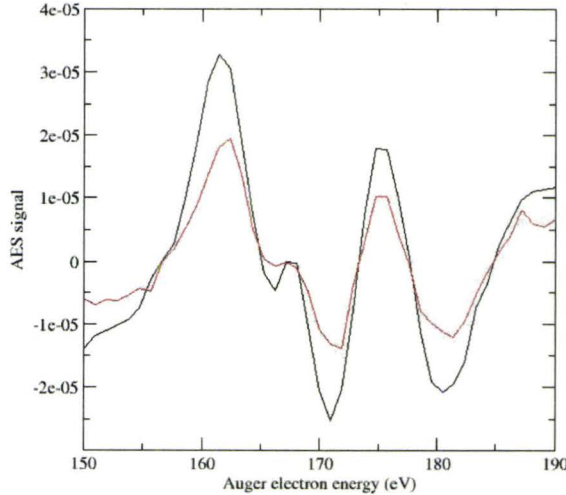


Figure 3.8: Auger spectrum without polynomial background for bare tungsten substrate (black) and that covered by 1ML Fe film (red).

energy). For convenience, a polynomial in energy can be fit to a spectrum over a restricted range to remove the background. In thickness calibrations (later paragraph of this section), the tungsten (bare or covered) spectra have a polynomial background subtracted, so only the Auger signals survive. Then the processed spectra are used for thickness calibration. Fig. 3.8 shows such sample spectra of tungsten in the energy range specified. Three negative peaks at 168eV, 173eV and 182eV are characteristic of tungsten.

Auger spectroscopy has manifold applications in surface science:

1. All elements in a film make a contribution to the Auger signal. These are superimposed to make an Auger spectrum. The composition of each film is qualitatively understood by making a least square fit to a combination of the Auger spectra of bulk materials or rather thick films. To perform this analysis, we need the standard spectrum I_n of each element and the spectrum I of the film. These spectra should be measured at the same energy points. For a film,

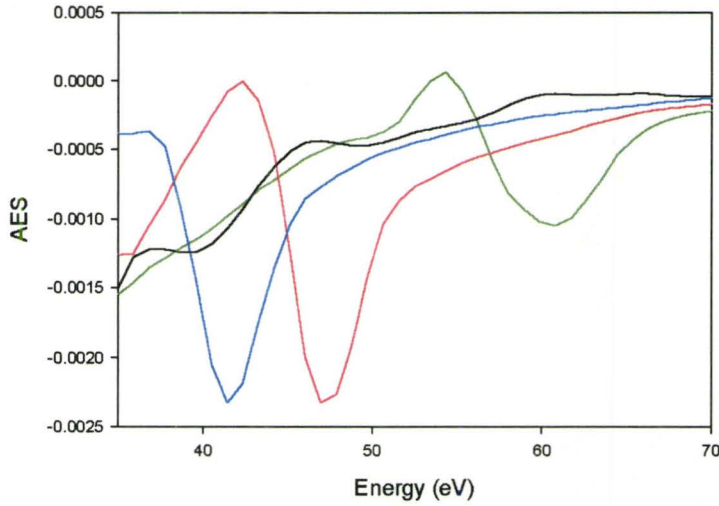


Figure 3.9: AES for very thick pure films: Fe (red), Ni (green), Mn (blue), tungsten (black).

which, for instance, has three elements

$$I = \alpha I_1 + \beta I_2 + \gamma I_3 \quad (3.7)$$

where α , β , γ are calculated using the least square criterion. They reflect the amount of elements 1,2,3 in a non-rigorous manner as any change in those fitting coefficients implies a change in the number of atoms or in their vertical location. Fig. 3.9 shows such a collection of spectra for 4ML Fe, 4ML Ni, 6ML Mn and W substrate respectively, all considered as standard spectra. These are taken using a main electron beam of 1keV in the energy range 35 to 70eV. Fe, Ni and Mn all have characteristic peaks with wide displacement and similar magnitude. The tungsten substrate doesn't have a remarkable peak in this region. The tungsten stays stable underneath the film and is not a major factor in determining the film profile. Fig. 3.10 shows an AES least square fitting for a film with Fe, Ni and Mn on W¹. The coefficients with standard errors for Ni, Fe, Mn and W are 0.21 ± 0.02 , 0.43 ± 0.01 , 0.11 ± 0.02 , 0.20 ± 0.01 .

2. In ultrathin film research, thickness calibration by Auger spectroscopy has an

¹See Sec. 4.1 for detail of the film.

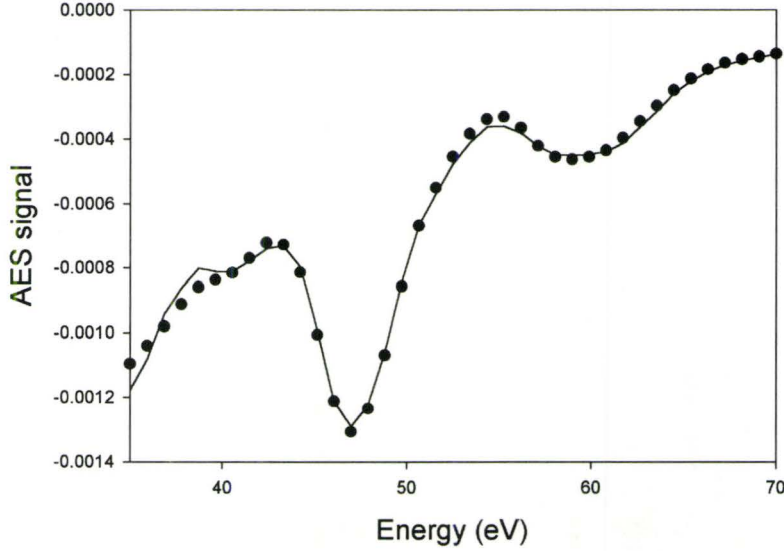


Figure 3.10: Original (dotted line) and fitted (solid line) AES for a Mn alloy film with 0.5ML equivalent Mn.

advantage over many other techniques [38]. The growth of a film increases the Auger signal of the film element and decreases that of the substrate element. Both Auger signals can be used for calibration. As a thicker film is deposited, more of the Auger electrons escaping from the substrate will be scattered inelastically by the film layers. The Auger electrons received by the analyzer decrease exponentially with respect to thickness. We can define an attenuation factor α to quantify the amount the signal decreases,

$$I = \alpha I_0 \quad (3.8)$$

where I_0 is the Auger signal of a bare substrate and I is the Auger signal of the substrate with film on top of it, both subtracting the polynomial background. This attenuation factor is easy to get as we do a least square fitting for I and αI_0 . As shown in Fig. 3.8 the attenuation factor is 0.60 for 1ML Fe on top of the tungsten substrate. In certain cases the attenuation factor and film thickness have a simple relation which makes the thickness calibration easy and consistent. This is true for film that grows in the FV mode. The layer-by-layer

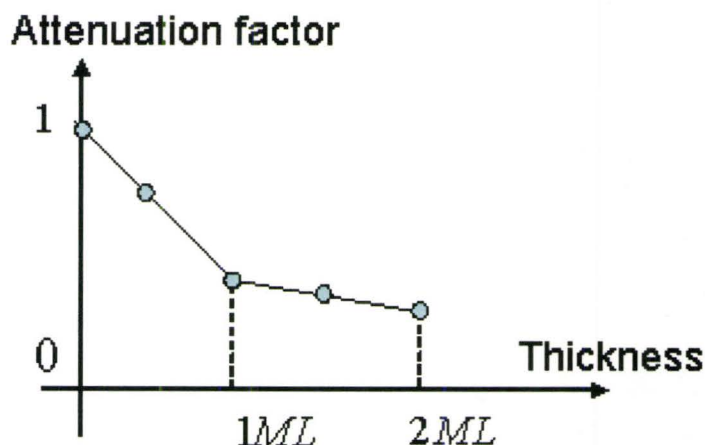


Figure 3.11: The attenuation curve for FV growth mode. Completion of certain single monolayer is called break point.

growth mode doesn't initiate the $(N + 1)^{th}$ layer until completion of the N^{th} layer. As the N^{th} layer grows, the Auger signal is a linear combination of that from a N layer film and a $N - 1$ layer film. Therefore the attenuation factor changes linearly during the growing of any single layer. After the completion of certain layer, the attenuation curve starts another linear segment which has a different slope (Fig. 3.11). The connecting point (break point) for adjacent segments then represents completion of a monolayer. So the calibration of thickness for FV mode is straightforward. The SK growth mode has one break point in its attenuation curve which still allows precise calibration. The VW mode has no break point but its exponentially decreased attenuation can be used for a less precise calibration of thicker films.

3. Of course, there are many other applications of Auger spectroscopy. It is an excellent means of monitoring surface cleanliness of samples, with a much higher sensitivity than LEED. Scanning Auger microscopy (SAM) has been widely used to provide spatially-resolved compositional information on heterogeneous samples.

3.2 Techniques

3.2.1 Kerr effect

Introduction

Classically, if a light wave is incident on a ferromagnetic solid, the solid generates a current according to Maxwell equations. The flowing electrons feel the Lorentz force $e\mathbf{v} \times \mathbf{B}$ due to the ferromagnetic moment. This causes an additional current which in turn causes an additional electromagnetic wave. The new electromagnetic wave makes a contribution to the reflected and transmitted waves. In 1845 Michael Faraday discovered that the plane of polarization of the transmitted wave is rotated from that of the incident wave. In 1877 an English clergyman J. Kerr discovered a similar rotation of the plane of polarization of the reflected wave. These are the notable Faraday and Kerr effects. Today, the Kerr effect is more important [42] since in most applications one deals with highly absorbing media, which do not allow optical techniques in transmission.

We categorize the Kerr effect into longitudinal, transverse and polar depending upon the orientation of the magnetization and the wavevector of the light. In the polar Kerr effect, the magnetization lies perpendicular to the reflecting surface. The largest effect occurs for normal incidence. In the longitudinal Kerr effect, the magnetization lies in the plane parallel to both the reflecting surface and the plane of light incidence. The largest effect occurs for grazing incidence. In the transverse Kerr effect, the magnetization lies in the plane parallel to reflecting surface and normal to the plane of light incidence. If one uses p-polarized light at non-normal incidence, the transverse Kerr effect manifests itself as a dependence of the reflectivity on the direction of the magnetization, and not in the change of the light polarization. For s-polarized light and transverse magnetization there is no usable effect because of symmetry. In most cases, the signal in the polar geometry is larger by roughly one order of magnitude than in the longitudinal geometry. This is one of the reasons that the polar geometry is the preferred one for magnetic data storage. Also, in general the geometries are not mutually exclusive. If one can't realize the optimum geometry for measuring a certain

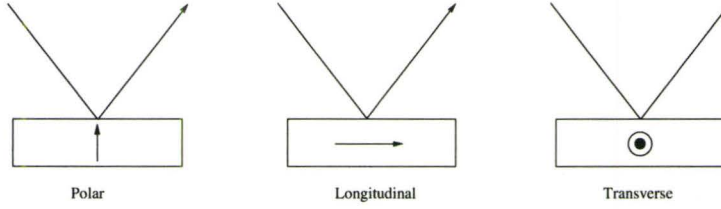


Figure 3.12: Three types of Kerr effect. The directions of magnetization are denoted by arrows.

magnetization direction, the experiment will also be sensitive to other magnetization components.

The general property that distinguishes magneto-optical Kerr effects from other magneto-optical effects in solids is that all manifestations of the Kerr effect are proportional to the magnetization M and vanish at temperatures above the Curie temperature T_c . It is therefore a powerful and convenient technique for probing magnetism in thin films. Many remarkable experiments have been achieved using the Kerr effect [14, 43]. However, Kerr effect measurements do not yield directly quantitative information on magnetic properties, *e.g.* magnetization. The direct measurements usually use arbitrary units or rotational angles. To calibrate measurements the Kerr data need to be analyzed with great attention.

Classical description of the Kerr effect

The current which occurs as a result of the Lorentz force can be incorporated in the phenomenological description by off-diagonal elements in the dielectric tensor. For a cubic magnetic material the tensor has the form

$$\epsilon = \begin{pmatrix} \epsilon_{xx} & \epsilon_{xy} & 0 \\ \epsilon_{yx} & \epsilon_{yy} & 0 \\ 0 & 0 & \epsilon_{zz} \end{pmatrix} \quad (3.9)$$

The coordinate system is chosen such that the magnetization is along the z direction. Due to crystal symmetry, $\epsilon_{xx} = \epsilon_{yy} \neq \epsilon_{zz}$ and $\epsilon_{xy} = -\epsilon_{yx}$. In a non-absorbing medium the diagonal elements of the tensor are real and off-diagonal elements are imaginary. In an absorbing medium both are complex. Suppose now that linearly polarized

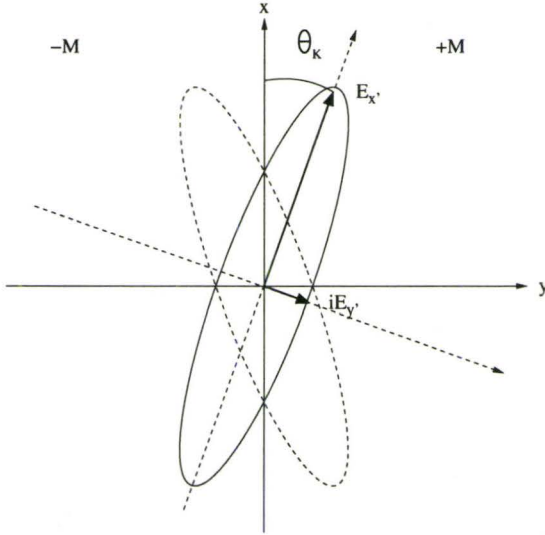


Figure 3.13: The incident polarization is along x direction. The reflection light has a Kerr rotation θ_K and ellipticity $\epsilon_K = E_{y'}/E_{x'}$ [4]. The dashed ellipse shows the polarization state for reversed magnetization. The Faraday effect is similar.

incident light interacts with a solid. In combination with proper boundary conditions the electrodynamic equations yield elliptically polarized waves in both reflection and transmission. As the transmission corresponds to the Faraday effect and reflection corresponds to the Kerr effect, we illustrate both situations in Fig. 3.13. Given the empirical dielectric tensor the electrodynamic theory can be used to derive analytical expressions for the Kerr rotation and ellipticity.

The surface magneto-optical Kerr effect (SMOKE) is associated with reflection from ultrathin magnetic films compared to the traditional magneto-optical Kerr effect (MOKE) associated with reflection from bulk magnetic materials. In the ultrathin condition, which is defined by

$$\frac{2\pi}{\lambda}|N|d \ll 1 \quad (3.10)$$

with λ the wavelength of the light and N the refractive index of the material, the magnitude of the longitudinal and polar SMOKE is proportional to the thickness of the film d [44].

Practical measurements

Our SMOKE measurements utilize a simple setup (Fig. 3.1). Light is provided by a 5mW He-Ne laser, with an intensity stability of 0.2%. Before the light strikes the sample, it passes through a Glan Taylor polarizer and a pyrex window. (The Glan Taylor polarizer consists of two calcite prisms which are separated by an air space. To produce linearly polarized light, it divides an entering unpolarized beam into two rays: one is the extraordinary ray that is transmitted through to the other side and the other is the ordinary beam that is totally internally reflected and absorbed.) The reflected beam passes through another pyrex window and enters the analyzer. The analyzer is just another polarizer but set nearly crossed with the first one. Light transmitted through the analyzer is detected by a photodiode. The reflected light is altered by both the Kerr effect and the Faraday effect, as the reflection is a mixture of reflection from the film and reflection from the substrate. For DC SMOKE the output from the photodiode is preamplified by about ten times before entering the data acquisition (DAQ) system. Each data point is averaged from ten readings. For AC SMOKE the output is fed into a lock-in amplifier which supplies a reference signal for the magnetic coils. The amplifier filters the Kerr response in phase with the reference signal. The coils for magnetic field supply are mounted *in situ* so that relatively small currents are required.

In an ideal situation, the interaction between the light and windows wouldn't change the light polarization and the effects of the windows could be neglected. The first polarizer could be adjusted to make the incident beam a pure S or P state with respect to the sample, depending on which SMOKE signal (polar, longitudinal, transverse) is favorable for the measurement geometry. Based on the Fresnel reflection coefficients no ellipticity is expected for the reflection of S or P polarized light from the sample, since they are eigenpolarizations for reflection. The polarization rotation would then be of purely magnetic origin through the Kerr effect.

However, imperfections exist in practical measurements:

1. The two polarizers can't achieve extinction. We define the extinction ratio e as the ratio of the intensity transmitted by crossed polarizers to that transmitted

by parallel polarizers. Then the measured intensity takes the form

$$I_m = I_0(\sin^2 \theta_m + e) \quad (3.11)$$

where I_0 is the intensity of the reflected light and θ_m is the angle between the polarization of the reflected light and the extinction angle of the analyzer. A change of magnetization in the sample will cause a slight rotation in the polarization of the reflected light, $\delta\theta$. This gives an alteration in the measured light intensity

$$\delta I = 2I_0 \sin \theta_m \cos \theta_m \delta\theta \quad (3.12)$$

Combining Eq. 3.11 and 3.12 gives the calibration scheme

$$\delta\theta = \frac{(\sin^2 \theta_m + e)\delta I}{2I_m \sin \theta \cos \theta} \quad (3.13)$$

In practice, θ_m is a very small angle and we can use the approximations $\sin \theta_m = \theta_m$, $\cos \theta_m = 1$. θ_m is chosen to minimize the signal to noise ratio, $\frac{\delta I}{I_m}$. This gives a measurement angle $\theta_m = \sqrt{e}$. Using Eq. 3.11, this angle is easily found by setting the polarizer angle so that the detected intensity I_m is twice that at extinction. This allows evaluation of I_0 at θ_m as $I_0 = \frac{I_m}{2e}$. Substituting into Eq. 3.12 gives

$$\delta\theta = \frac{\sqrt{e}\delta I}{I_m} \quad (3.14)$$

Eq. 3.14 can be used to calibrate the signal and plot hysteresis in units of rotational angle. Typically, uniformly magnetized ultrathin FCC iron rotates the polarized He-Ne laser beam by tens of microradians. This can be converted into a magnetic unit given enough information. It is easy to get a pair of commercial polarizers with extinction ratio $e < 10^{-6}$. But the extinction ratio of the system will be degraded if the experiment takes place in a noisy environment, which is often the case.

2. The AC susceptibility measurement is different from the hysteresis measurement as a lock-in amplifier supplies a reference signal for the magnetic coils and meanwhile extracts SMOKE signals in phase with the reference signal. These

dynamic measurements also have different sources of noise than that of hysteresis loops. In an earlier study [45], it was found that a larger measurement angle $\theta_m = 3.4 \text{ mrad}$ maximized the signal to noise ratio. The calibration to rotational angles still uses Eq. 3.13. The calibration also needs to consider the impedance of the coils which causes a phase lag between the magnetic field and the reference signal. For experiments at 150Hz, the phase lag is 305° . Typically, ultrathin FCC iron has a peak susceptibility of a few microradians/Oe at the critical temperature.

3. The two windows in the optical path are not ideal. Local strain in the materials creates birefringence and the transmitted light is elliptically polarized. The introduction of a polarization component in the minor axis of the ellipse of polarization degrades the extinction ratio of the system to 10^{-4} . While using normal incidence reduces the ellipticity [46], that which remains must be compensated. A simple iterative process is used to find the optimal path where the ellipticity generated by the two windows is compensated by the ellipticity generated by reflection from the sample when it is tipped slightly away from the S or P polarization condition. Then the light entering the analyzer is linearly polarized. Initially, the incident light is set to the S or P state and an intensity minimum is found by adjusting the analyzer angle. Then the incident polarization is rotated by a few arcminutes and the transmitted intensity is minimized again. If the extinction ratio improves, the incident polarization should be adjusted again in the same direction. Otherwise, the incident polarization should be rotated in the opposite direction and the operation is repeated. Iteration continues until a global minimum is found in the extinction ratio. It is found that the minimum extinction ratio occurs around five degrees from the pure S or P state [47].
4. A Glan Taylor polarizer is used as the analyzer. As the analyzer divides the laser beam into an extraordinary ray and an ordinary ray, the ordinary ray is not perfectly internally absorbed. After multiple reflections it can escape from the analyzer and cause a diffuse glow. The extinction ratio is limited at this stage

by the diffuse glow from the scattered light. Even though the diffuse glow is much less intense than the laser beam, it falls on the full area of the photodiode and creates a significant signal. A corrective scheme is to introduce an aperture in front of the analyzer. However this can cause difficulties if the main beam is deflected due to sample vibration and misses the aperture opening. The beam often deviates from a 0.3mm diameter aperture due to tiny sample vibrations. A larger aperture of 1mm diameter is a good compromise for both concerns [47].

Chapter 4

Results

4.1 Published Paper

The following section contains a paper published in the Journal of Applied Physics[48]. Qiangyong Li's previous work on this system (summarized in Ref. [49]), was to finalize the film growth and annealing procedures. He used LEED and directional AES (DAES) to study the the film structure. He has been included as an author in the present paper because of his efforts in training and passing the expertise on. All the experiments reported here are performed by me, including the discovery of a new pattern in LEED, the exploration of magnetic properties, and the data analysis. References to supplementary work are added as footnotes to the original paper.

Structural and Magnetic Properties of a Chemically Ordered FCC (111) Mn Alloy Film

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Abstract

A 4ML Ni/W(110) substrate is used to establish a (111) f.c.c. template upon which 3ML Fe is deposited and annealed to 580K to form a substrate with very good short and long range f.c.c. (111) order, that is Fe rich at the surface. Mn alloy films are formed by annealing a subsequent Mn deposit of 0.3 to 1.6 ML. LEED and AES show that an ordered alloy is not formed until an annealing temperature of 580K, upon which a multilayer alloy with a $P(2\sqrt{3} \times 2\sqrt{3})R30^\circ$ LEED pattern is created. The alloy films formed from 0.3 to 0.5 ML of Mn have magnetic properties similar to the FeNi substrate. Hysteresis loops and ac-susceptibility curves measured using the Kerr effect give square loops with a ferromagnetic moment along the in-plane f.c.c. $[-211]$ direction and a Curie temperature T_C of about 460K. There is an increase in coercive field likely due to the inhomogeneities introduced by the Mn. Alloy films formed from 0.8 to 1.6 ML of Mn show a marked increase in the width of the susceptibility peak, and a decrease in the peak temperature. The hysteresis loop becomes slanted with a reduced coercive field. The measurements are consistent with a paramagnetic or antiferromagnetic Mn alloy forming an uneven interface within the FeNi film, so that the remaining FeNi film has a wide distribution in T_C .

FeMn alloy is an example of a metallic antiferromagnet with applications in science and industry. Bulk $\text{Fe}_x\text{Mn}_{100-x}$ alloys are antiferromagnetic with a fcc structure in a concentration range between $x = 45$ and $x = 75$ at room temperature, with the highest Néel temperature (T_N) of about 500K at $x = 50$. However no chemically ordered structure is found in this phase diagram[21]. Similarly, rather thick (8nm) antiferromagnetic FeMn[18, 50] films that are common pinning layers in exchange biased spin-valves, do not form a chemically ordered alloy. Epitaxial ultrathin films(1-26 ML) grown on Cu(100) by co-evaporation are also chemically disordered[15, 51, 52], except for the Cu interface layer. Recently we have been able to grow a chemically ordered Mn ultrathin film alloy by annealing[49] up to 2 ML Mn into a FeNi f.c.c. (111) substrate layer with a Fe rich surface, while retaining the substrate structure. The structure of this system has been reported previously[49]; here we describe an investigation of its magnetic properties.

The films were grown on a W(110) substrate by electron beam evaporation of pure Ni and Fe wires and pure Mn pieces in a UHV chamber with a base pressure of 1×10^{-10} torr. The evaporators have an integral ion flux monitor to ensure a stable deposition rate that can be calibrated by Auger electron spectroscopy(AES)[38]. The first monolayer of Ni was deposited at 550K then annealed at 600K for 1 minute. An additional 3 ML Ni gave a clear f.c.c. (111) LEED pattern. Subsequent deposition of 3 ML Fe at room temperature gave no clear LEED pattern until the film was annealed at 580K for 20s. The resulting FeNi alloy film showed very good f.c.c. (111) structure as revealed by the LEED pattern and Directional AES(DAES)[49]. Measurements of the susceptibility (described below) gave a T_c of 465K within an error of a few degrees. The chemical and magnetic stability of the FeNi alloy substrate was confirmed before Mn deposition by annealing cycles similar to those used for formation of the Mn alloy. No marked changes in the Auger spectra¹ and susceptibility curves² were observed. Mn alloy films were obtained by depositing x equivalent ML of Mn on the FeNi alloy substrate film ($x = 0.3$ to 1.6) in successive increments, with short annealing(20s) at 580K between depositions.

¹Refer to Sec. 4.2.1

²Refer to Sec. 4.2.3

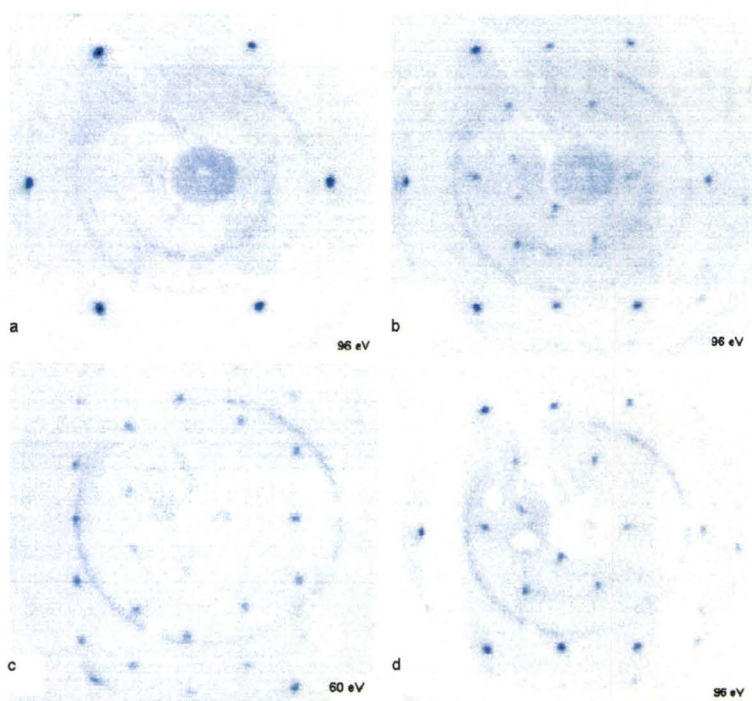


Figure 4.1: LEED pattern for the $x\text{Mn}/3\text{Fe}/4\text{Ni}/\text{W}(110)$, $x = 0$ (a), and $x = \frac{1}{2}$ (b), and $x = 1\frac{1}{2}$ (c, d). The primary energy of the electrons is indicated. The circle in the background is reflection of LEED filament light coming out of a ring-shaped crack.

Selected LEED patterns of the Mn alloy are shown in Fig. 4.1. The as-deposited Mn on top of the substrate showed no clear LEED pattern. Subsequent annealing of $x \leq 0.3$ ML Mn gave a clear $P(1 \times 1)$ LEED pattern similar to that for the $x = 0$ pattern in Fig. 4.1a. Alloys of $0.3 < x < 1.6$ ML Mn resulted in a clear $P(2\sqrt{3} \times 2\sqrt{3})R30^\circ$ LEED pattern, as shown in Fig. 4.1b-1d. To understand the $P(2\sqrt{3} \times 2\sqrt{3})R30^\circ$ LEED pattern, we note the following points. First, this pattern appears for $x > 0.3$ ML Mn only after annealing, where AES shows the Mn signal decreases³. This indicates it is not a simple surface alloy. (If the real structure was formed by Mn staying on the surface, this LEED pattern would be complete at $\frac{1}{12}$ ML Mn.) Second, the intensity ratio of the $(\frac{1}{2}, 0)$ spot to the $(1, 0)$ spot monotonically increases with Mn amount to at least $x = 1$. In contrast the intensity ratio of the $(0, \frac{1}{\sqrt{3}})$ spot to the $(1, 0)$ spot increases with Mn amount up to $x = 0.5$ and then remains nearly a constant⁴. This suggests that independent $P(2 \times 2)$ and $P(\sqrt{3} \times \sqrt{3})R30^\circ$ patterns are coherently superimposed to give the ordered $P(2\sqrt{3} \times 2\sqrt{3})R30^\circ$ pattern. Third, the LEED shows only $P(2 \times 2)$ spots at higher electron energy (96eV). The extra $P(2\sqrt{3} \times 2\sqrt{3})R30^\circ$ spots (which had not been noticed earlier[49]) are only noticeable at lower electron energy (60eV). This indicates the formation of $P(\sqrt{3} \times \sqrt{3})R30^\circ$ [53] near the surface and $P(2 \times 2)$ deeper in the film. Since the Fe and Ni are not ordered in the f.c.c. (111) substrate and the LEED pattern changes from $P(1 \times 1)$ only once Mn is deposited and annealed, it is highly likely that only the Mn orders chemically; that is, some ordered compound $Mn_\alpha X_\beta$ is formed, where at any particular site X is either Fe or Ni determined randomly according to the local concentration. Recall that since the surface of the substrate is Fe rich, X is most often Fe. The $P(\sqrt{3} \times \sqrt{3})R30^\circ$ is consistent with a X_2Mn surface termination seen in some surface alloys[54, 55, 56]. The deeper $P(2 \times 2)$ structure may be XMn_3 , X_3Mn or three symmetry related domains of XMn . It is not yet possible to distinguish between these alternatives.

The magnetic susceptibility measurements used an a.c. Surface Magneto-Optical Kerr Effect(SMOKE) technique described elsewhere[45], with a modulation field of 2.1 Oe applied along the in-plane f.c.c. $[-211]$ direction. The modulation frequency was

³Refer to Sec. 4.2.1

⁴Refer to Sec. 4.2.2

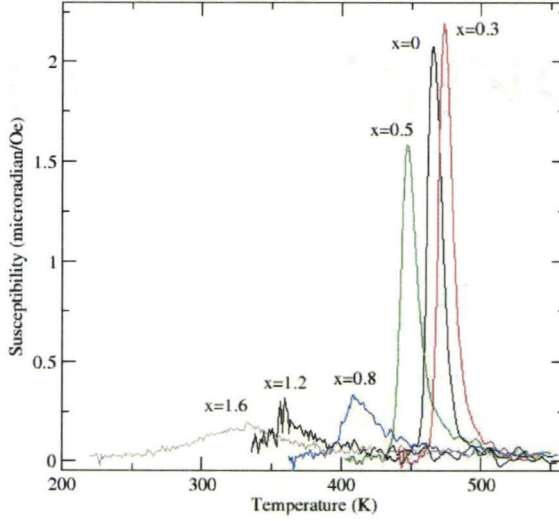


Figure 4.2: Susceptibility for $x\text{Mn}/3\text{Fe}/4\text{Ni}/\text{W}(110)$.

150 Hz. The susceptibility was measured in the cooling process right after annealing and is shown in Fig. 4.2. Near the peak in the susceptibility the cooling rate was smaller than 0.3K/s. After cooling to 340K the hysteresis loops in Fig. 4.3 were measured in the same geometry. For $x = 1.6$ no hysteresis was observed at 340K, but further cooling to 210K produced a loop, as shown in Fig. 4.3.

The bare FeNi substrate had a square hysteresis loop with a coercive field of $H_c = 38\text{Oe}$. Upon adding the first 0.3 ML Mn on FeNi, the hysteresis loop of the as-deposited film⁵ was essentially unchanged. Subsequent annealing induced an increase of H_c as seen in Fig. 4.3. It appears that the diffusion of Mn has created inhomogeneities which enhanced the coercivity. In the vicinity of 0.8 ML Mn the coercive field and saturation magnetization start to decrease, and the hysteresis loop becomes slanted. For the alloy of 0.8 ML Mn it takes about 5 Oe for the magnetization to switch from $+M_s$ to $-M_s$ whereas for the alloy of 1.2 ML Mn it takes about 20

⁵Refer to Sec. 4.2.3

Oe for the switching. The corresponding susceptibility peak values in Fig. 4.2 move to lower temperature, and the full width at half maximum(FWHM) increases, from 10K for the FeNi substrate; to 20K for the alloy of 0.8 ML Mn; to 70K for the alloy of 1.6 ML Mn.

For a qualitative understanding, we attribute these phenomena to a partially antiferromagnetic or paramagnetic film. As the film incorporates more Mn, the top layers become paramagnetic(PM) or antiferromagnetic, a direct result of which is to decrease the thickness of the ferromagnetic FeNi layer underneath. In this model the movement of the susceptibility peak to lower temperature is caused by finite size scaling[57], which reduces the Curie temperature T_c . When T_c decreases, the hysteresis loop measuring temperature(340K) gets closer to the critical point where hysteresis disappears. Approaching the transition point causes the decrease of M_s and H_c for alloys of more than 0.8 ML Mn. As the FeNi layer thickness was reduced by Mn diffusion, it was not uniform. Regions of different thickness possess different T_c s and the superposition of the different susceptibility signals from these regions form the broad susceptibility peak measured experimentally. No MOKE signal was found for field applied along the orthogonal in-plane or perpendicular directions.

It remains to determine whether the Mn alloy is paramagnetic or antiferromagnetic. It may be possible to resolve this question by field cooling the film and searching for the effects of an exchange bias on the hysteresis loop.

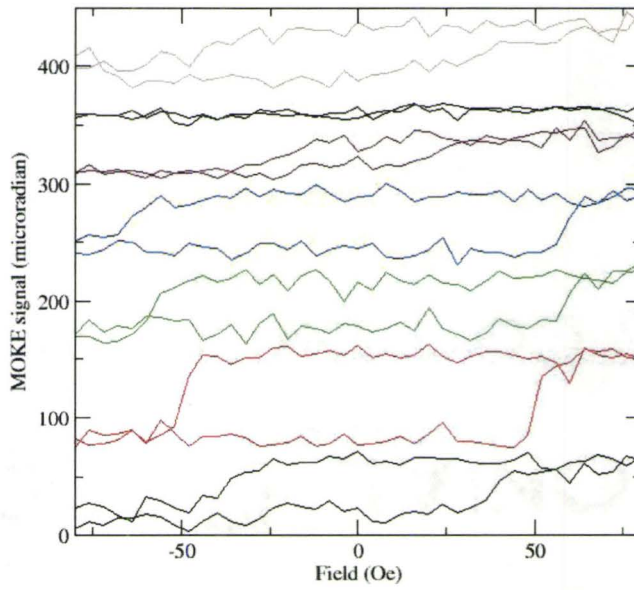


Figure 4.3: Hysteresis loops for $x\text{Mn}/3\text{Fe}/4\text{Ni}/\text{W}(110)$ in 340K except the top one which was measured in 210K for $x = 1.6$. From bottom to top, $x = 0, 0.3, 0.5, 0.8, 1.2, 1.6, 1.6$. The curves have been displaced vertically for clarity.

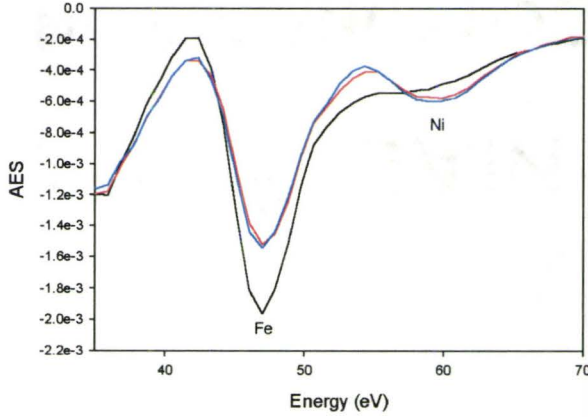


Figure 4.4: AES for FeNi substrate before annealing (black), after the first (red) and the fourth (blue) annealing. The peak for each element is denoted.

4.2 Supplementary work

In this section, I am dealing with the same Mn alloy films as in Sec. 4.1. Some data used but not shown in the paper are supplied here.

4.2.1 From AES

The FeNi substrate is formed by an initial annealing procedure, and then undergoes several more annealing cycles in the sequential Mn deposition. It is important that there is no substantial change in the substrate after the initial annealing because otherwise it would be difficult to extract properties of the FeMn layer. This expectation is verified in a single experiment when a substrate alone is annealed a number of times. In Fig. 4.4, the initial annealing has a significant effect. Large amounts of Fe and Ni mix in the interface. However, subsequent annealing cycles incur only small change.

The annealing cycles change the concentration profiles of films once Mn has been deposited. In Fig. 4.5 the Mn peak exhibits a large decrease after one annealing cycle while the Fe peak and the Ni peak remain almost unchanged. It directly indicates

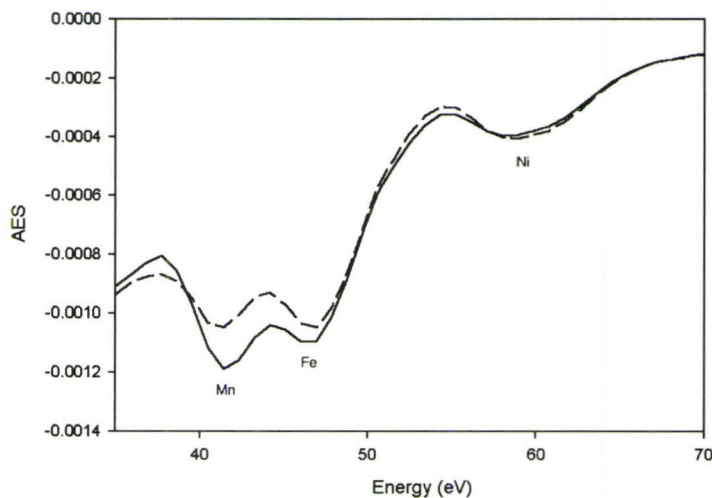


Figure 4.5: AES for a Mn alloy film before (solid line) and after (dashed line) annealing, with 1.2ML equivalent Mn on the FCC (111) surface. The peak for each element is denoted.

the Mn atoms diffuse into the body of the film from the surface. At proximity with the Fe, it is believed the Mn mainly mixes with Fe and forms FeMn alloy layers. At this point $x\text{Mn}/3\text{Fe}/4\text{Ni}/\text{W}(110)$ just denotes the amount of an element deposited. There is no clear interface between elements and their relative concentration varies with depth.

Following Eq. 3.7 and other procedures in Sec. 3.1.4, the Auger fitting coefficients for each alloy film are calculated and plotted in Fig. 4.6. There are three quite stable coefficients, coefficients of W, Ni and summation of all elements. Overall, the W coefficient is the smallest because W stays in the bottom. The Ni coefficient has a slight decrease as the amount of Mn increases and screens the Ni Auger electrons. This result confirms that the Ni doesn't move to the surface upon annealing. The Fe coefficient falls significantly because the surface is Fe rich before Mn is deposited. As expected, the Mn coefficient increases continuously. This shows that to a great extent, an alloy of Fe and Mn is being created. Gradually, the Fe and Mn coefficients approach an equilibrium point. This occurs when the Fe and Mn are relatively evenly mixed. This fitting procedure is self consistent because the sum of all the coefficients

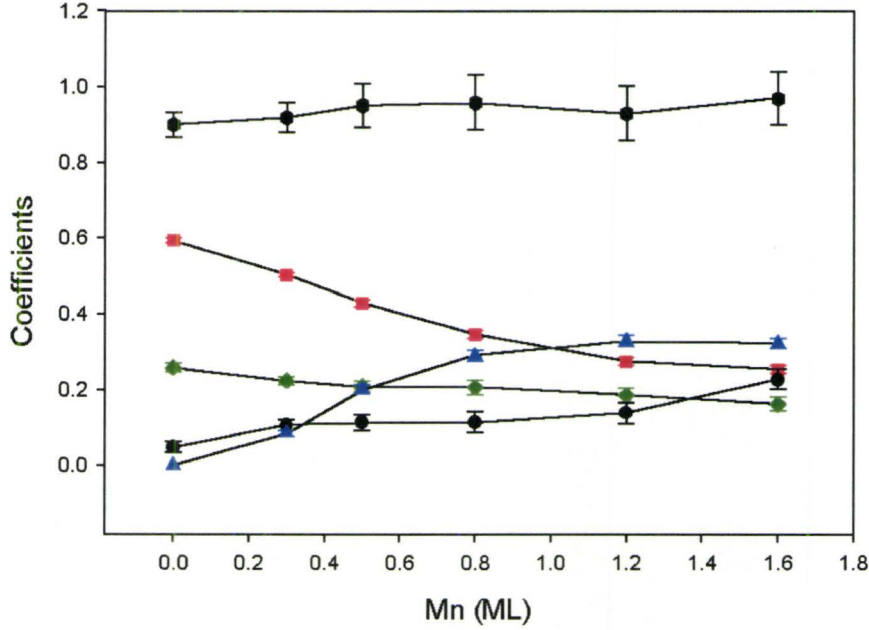


Figure 4.6: Least square Auger fitting coefficients for Fe (red), Ni (green), Mn (blue), tungsten (black) and their summation (grey).

normalizes to one. The error bars for standard deviation are small. If we return to Fig. 3.10 the example shows that position and magnitude of the peaks are fitted well. The small discrepancies are attributed to apparatus quality and theoretical imperfection. The mean free path, and therefore the attenuation of AES, is not a constant in different electron energies [58]. This effect is more evident in 35-70eV than in 150-190eV.

4.2.2 From LEED

The FeNi substrate of the film has a $P(1 \times 1)$ pattern. As Mn is deposited and annealed, extra spots appear in the LEED pattern (Fig. 4.7). We notice the brightness of these extra spots are different, and their variation with thickness not equivalent (Tab. 4.2.2). It is believed that the $(\frac{1}{2}, 0)$ spots and $(0, \frac{1}{\sqrt{3}})$ spots are from different sub-lattices. How these sub-lattices are superimposed upon each other and form the $P(2\sqrt{3} \times 2\sqrt{3})R30^\circ$ structure has been interpreted in Sec.4.1.

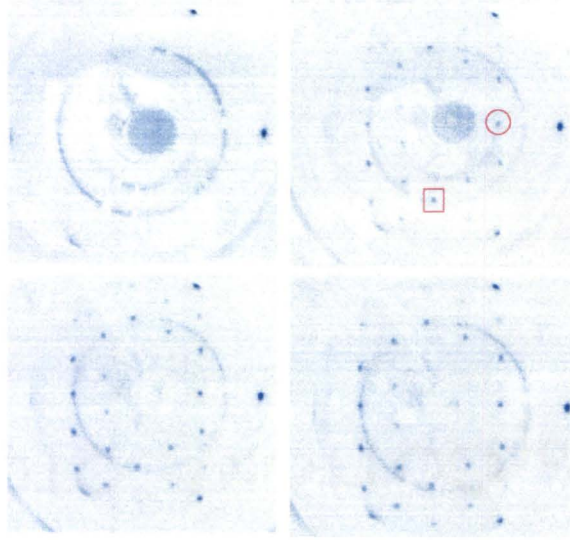


Figure 4.7: LEED patterns with beam energy 60eV for annealed xMn/3Fe/4Ni/W(110). $x=0.3$ (top left); $x=0.5$ (top right); $x=0.8$ (bottom left); $x=1.2$ (bottom right). The $(0, \frac{1}{\sqrt{3}})$ spot is noted by a rectangle; the $(\frac{1}{2}, 0)$ spot is noted by a circle, of which the $(1, 0)$ spot is to the right.

	0.3ML	0.5ML	0.8ML	1.2ML
$(0, \frac{1}{\sqrt{3}})$	0.00	0.29	0.31	0.31
$(\frac{1}{2}, 0)$	0.00	0.29	0.39	0.42

Table 4.1: The intensity ratio of the $(0, \frac{1}{\sqrt{3}})$ spot and the $(\frac{1}{2}, 0)$ spot to the $(1, 0)$ spot for annealed xMn/3Fe/4Ni/W(110).

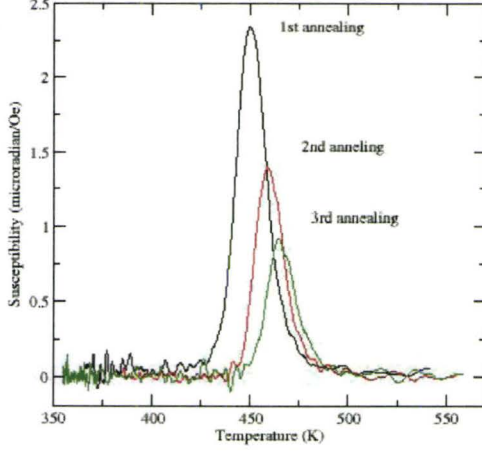


Figure 4.8: Susceptibility for 3Fe/4Ni/W(110) measured after the annealing cycle as indicated.

It is noteworthy that the $P(2\sqrt{3} \times 2\sqrt{3})R30^\circ$ pattern appears right after the annealing, *i.e.* at 580K. This observation excludes the possibility that the new LEED pattern is caused by magnetic ordering, which, if is true, implies the Néel temperature of the FeMn is higher than 580K. However, the highest reported Néel temperature of bulk FeMn is around 500K[21].

4.2.3 From SMOKE

The susceptibility of 3Fe/4Ni/W(110) (Fig. 4.8) has been measured. Like for its Auger spectrum (Fig. 4.5), we hope it reaches equilibrium after the first annealing. The T_c and FWHM stay fairly stable. And it is noticed that the second and third annealing increase the T_c . We think that the T_c of 0.3Mn/3Fe/4Ni/W(110) in Fig. 4.2 should be largely caused by annealing the FeNi rather than formation of dilute Mn alloy. However, when Mn is more than 0.5ML, the trend of T_c and FWHM is largely caused by Mn alloy formation since it bears no resemblance to that in Fig. 4.8.

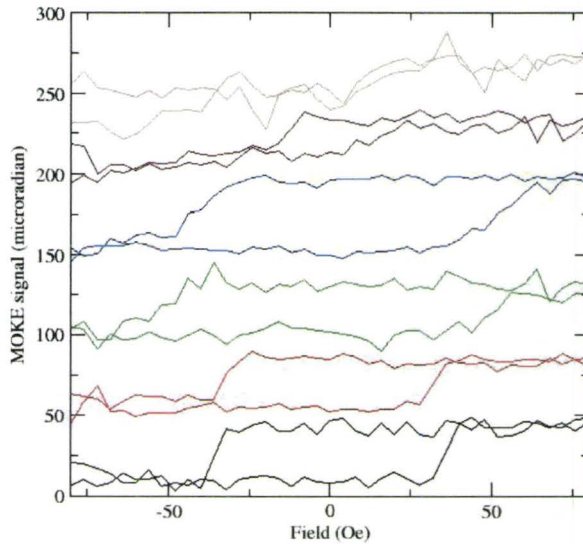


Figure 4.9: Hysteresis loops for $x\text{Mn}/3\text{Fe}/4\text{Ni}/\text{W}(110)$ at 400K, from bottom to top: $x=0$ after anneal; $x=0.5$ before anneal; $x=0.5$ after anneal; $x=1$ before anneal; $x=1$ after anneal; $x=1.5$ before anneal.

The magnetic properties of the films can be significantly changed by annealing. As shown in Fig. 4.9 deposition of Mn without annealing does not profoundly change the hysteresis loop. This is probably because Mn is not ferromagnetic and simply lies onto the ferromagnetic films. After annealing, Mn mixes with the Fe to form an alloy and greatly changes the hysteresis loop. Also, the formation of the alloy affects the homogeneity of the film, changing its coercivity.

The phase diagram of a particular alloy film has two parameters, thickness and temperature. At a critical thickness and a critical temperature, a Curie phase transition occurs. We have studied the hysteresis loops versus Mn concentration in section 4.1. In that experiment, the measuring temperature is constant but the decreasing thickness of the ferromagnetic substrate drives the Curie temperature T_c down. The hysteresis versus thickness diagram shows one axis of the phase diagram. If we measure the same film under increasing temperatures (Fig. 4.10) the coercivity

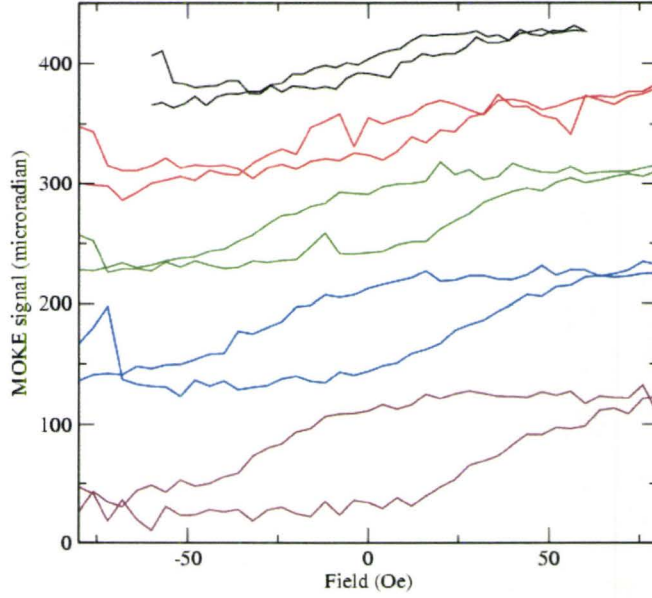


Figure 4.10: Hysteresis loops for a Mn alloy film with 1.5ML equivalent Mn, from bottom to top: 330K, 340K, 350K, 360K, 380K.

and magnetization both decrease. The shape remains slanted. We clearly see the approach to the magnetic transition along the temperature axis.

In order to confirm the trend of the susceptibility curves in Fig. 4.2, a second, identically prepared sample was measured. The FWHM and T_c s of the two samples are plotted in Fig. 4.11 for comparison. The trend of decreased T_c and increased FWHM as the Mn content of the alloy is increased, is confirmed. However, both parameters are not reproduced exactly. In ultrathin film engineering, a small variation in fabrication conditions can have a large influence.

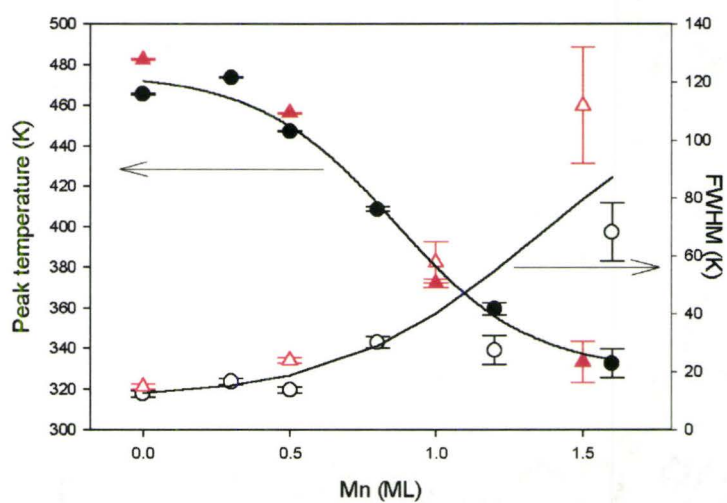


Figure 4.11: Full width at half maximum (FWHM) and T_c s for two similar samples (red and black). The lines are guide to the eye.

Chapter 5

Conclusion

This thesis has presented structural and magnetic properties of a chemically ordered manganese alloy film. The film is grown by electron beam evaporation and the calibration technique allows for a thickness error of under $\pm 10\%$.

The 3Fe/4Ni/W(110) substrate, after annealing, has a Fe rich surface. Further similar annealing cycles don't change its Auger spectrum substantially, showing that it is near equilibrium after the first annealing. Also, the substrate doesn't have a f.c.c. (111) surface until it is annealed. Compared with the Mn as-deposit film, the annealed film has a much smaller Mn peak in the AES spectrum. This is a sign that Mn diffuses into the film and mixes with Fe or Ni, making the film an alloy. The depth profiles of the films are studied by a least square fitting method, which assumes the Auger signal is a linear combination of the component signals, with fitted coefficients. While globally the Mn coefficient increases and the Fe coefficient increases, the Ni coefficient stays approximately constant which means the Ni doesn't come to the surface in a considerable amount. So the alloy should be composed of a majority of FeMn. With more than 0.3ML Mn, the film turns into a chemically ordered alloy. The film has a $P(2\sqrt{3} \times 2\sqrt{3})R30^\circ$ LEED pattern, clearer at lower beam energy. At higher beam energy the LEED pattern looks more like $P(2 \times 2)$. Since these LEED patterns only appear after annealing, the film is not a simple surface alloy. These unique LEED patterns may be a superposition of a surface $P(\sqrt{3} \times \sqrt{3})R30^\circ$ from a MnFe₂ alloy and a $P(2 \times 2)$ from the deeper alloy layers of MnFe, or MnFe₃ or Mn₃Fe.

AC susceptibility and hysteresis loops are measured using MOKE. The magnetic field is applied along the in-plane f.c.c. $[-211]$ direction, easy axis of the film. From susceptibility curves, we see the Curie temperature decreases and the full width at half maximum (FWHM) increases with higher Mn concentration. These changes in the ferromagnetic response of the film are attributed to the formation of the Mn alloy. The hysteresis loops are rather square for alloys with less than 1ML Mn. The initial addition of Mn creates inhomogeneities and enhances the coercivity. With more than 1ML Mn, the Curie temperature decreases, reducing the magnetization and coercivity at a fixed temperature. No change of easy axis is detected in this process. Alloys made with 1ML Mn or more have a slanted hysteresis loop below the Curie temperature. This indicates a greater degree of inhomogeneity, either due to a wide distribution of pinning sites or a distribution of transition temperatures from uncorrelated regions of the sample.

All the data and analysis are consistent with a paramagnetic or antiferromagnetic, chemically ordered FeMn alloy. But the question remains how to discriminate between these two. It is also possible that the Mn alloy is paramagnetic near room temperature but will order as antiferromagnetic at lower temperatures. The next step is to field cool the alloy film and search for an exchange bias.

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