Structural and Magnetic Properties of Fe/Co (001) and Fe/V (001) Superlattices

BY

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ABSTRACT

The structural and magnetic properties of Fe/Co (001) and Fe/V (001) superlattices have been investigated. The thin film structures were deposited on MgO (001) substrates using a four source ultra-high-vacuum based sputtering equipment.

Reflection high energy electron diffraction showed that the body-centered cubic phase of Co could be stabilized up to a thickness of about eight atomic monolayers. X-ray diffraction measurements confirmed that the Fe/Co superlattices were single-crystalline with a pure body-centered cubic structure. The crystalline quality and the interface profile were found to be strongly dependent on the growth temperature. Furthermore, the Fe/Co superlattices were also found to exhibit novel magnetic properties. The first order magnetic anisotropy constant for the body-centered cubic Co phase was determined to be negative with a magnitude of 110 kJ/m³. Magnetization and Mössbauer measurements showed that the Fe and Co magnetic moments in the superlattices were enhanced. Spin polarized neutron reflectometry proved that all the magnetic moments were collinear.

The interlayer exchange coupling and the giant magnetoresistance effect in Fe/V superlattices were also studied. The strength of the coupling was found to be dependent on the thickness of the Fe and the V layers. This was also true for the giant magnetoresistance effect. Mössbauer measurements showed that the V-on-Fe interfaces (V deposited on Fe) were more abrupt and less diffuse than the Fe-on-V interfaces. Finally, the lattice parameters of a series of different Fe/V superlattices were determined by x-ray diffraction. The experimental lattice expansion was found to be in good agreement with a simple theoretical model.

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XIV  Phase transitions of hydrogen in quasi-two-dimensional vanadium lattices
My contributions to the included papers:

I  I have grown all the samples, performed the XRD investigation and written most of the paper. I have taken an active part in the planning of the project and also been involved in the neutron measurements.

II I have done everything myself.

III I have done everything myself.

IV I have grown all the samples and been responsible for the XRD measurements.

V I have grown all the samples, performed the XRD investigation and written part of the paper. I have also taken an active part in the planning of the project.

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Paper VIII-XIV:

I have grown all the samples and performed all the XRD measurements.
Chapter 1

Introduction

Magnetism has been known to mankind for at least three thousand years. It has been used in different ways to help people in their everyday life, in everything from a simple device like a compass to today’s advanced computer memory. In the last decade there has been a dramatic increase in interest in the magnetic properties of materials. The development of ultra-high-vacuum (UHV) systems has made it possible to grow thin epitaxial films, only a few atomic monolayers thick, with negligible contamination, low defect densities and well defined interfaces. The crystalline quality as well as the epitaxial orientation of the films can be readily controlled by varying the growth conditions. Magnetic multilayers can be made by depositing alternating thin films of magnetic and non-magnetic materials on top of each other. If the crystalline quality of the multilayer is high it is called a superlattice. The two most commonly used growth techniques today are sputtering and molecular beam epitaxy (MBE). These new methods to deposit thin films, multilayers and superlattices have led to the observation of a wide range of new magnetic phenomena which are not found in bulk materials. Two examples of novel magnetic properties are the oscillatory interlayer exchange coupling of magnetic moments across a non-magnetic spacer film and the giant magnetoresistance (GMR) effect [1-4]. Furthermore, as the film thickness is reduced, the role of interfaces becomes more important and may even dominate that of the bulk. In films only a few atomic monolayers thick, an interface contribution to the magnetic anisotropy is capable of rotating the magnetic moments from in the film plane to perpendicular to the film plane [5-7]. Another novel property is the enhancement of the magnetic moments in thin magnetic films due to the reduced dimensionality [8-10]. Especially the giant magnetoresistance effect has attracted a lot of attention because it can be exploited to read information from a magnetic disc but also the
perpendicular magnetization in thin films can be utilized. On a magnetic disc the information is stored in form of small magnetized regions. The perpendicular alignment of the magnetic moments makes it possible to decrease the size of those regions and thereby increase the storage areal density.

Fe/Co and Fe/V are two material systems which exhibit many of the above mentioned novel magnetic properties. In this thesis, sputter deposited Fe/Co (001) and Fe/V (001) superlattices on MgO (001) substrates have been studied. The structural as well as the magnetic properties of these two systems have been investigated using different characterization methods like x-ray diffraction (XRD), reflection high energy electron diffraction (RHEED), spin polarized neutron reflectometry, SQUID magnetometry and Mössbauer spectroscopy. The thesis is organized as follows; the growth of superlattices and their crystalline structure are discussed in chapter 2, an introduction to thin film magnetism is given in chapter 3, the characterization techniques are described in chapter 4 and the last chapter is a short summary of the results reported in the papers included in this thesis.
Chapter 2

Crystalline structure and growth of thin films

2.1 Crystalline structure of thin films and multilayers

Multilayers are made by depositing in an orderly fashion alternating thin films of two or more materials. In metallic magnetic multilayers the materials are metals and at least one is magnetic. The properties of these multilayer are uniquely determined by the thickness of the individual layers, the crystalline structure and the properties of the constituents. In a multilayer, the layers can be amorphous, poly-crystalline, textured or single-crystalline [11]. If the multilayer has a large crystallographic coherence in all directions it is referred to as a superlattice. In a superlattice the layers are grown epitaxially which means that the constituents adjust their lattice constants to match and grow as an oriented single crystal multilayer. In addition to the interatomic distance, superlattices also exhibit a periodicity in the growth direction called the superlattice period as shown schematically in Fig. 1.

2.1.1 Substrate

The choice of substrate is one of the most important issues in thin film growth. Epitaxial growth of superlattices requires matching the film and substrate with respect to lattice parameters, atomic positions and crystallographic orientation. Superlattices with high crystallographic coherence in all directions are possible only if the lattice mismatch between the constituents is less than a few percent. If the mismatch is too large, a textured or a poly-crystalline structure are generally obtained. Furthermore, the substrate should not react chemically with the film, which makes oxides like MgO and Al₂O₃ suitable when films are grown at high temperatures.
The substrate surface must also be clean, smooth and uniform to ensure a uniform, homogenous film with high crystalline quality. In general, the crystalline quality of a superlattice will not be better than the quality of the substrate \[12\]. By varying the crystalline orientation of the substrate, it is also possible to control the epitaxial orientation of the film. A buffer layer between the substrate and the film can be used to reduce substrate related defects or to gradually decrease lattice misfit. Thin films are also often capped with a protective layer to avoid oxidation.

2.1.2 Strain

Lattice mismatch between the constituents in a superlattice will give rise to strain in the crystal structure. Strain can also be induced by the deposition process or by differences in thermal expansion coefficients. If the lattice mismatch is not too large the strain can be accomodated by introducing a tensile strain in one constituent and a compressive strain in the other. The constituent with the larger lattice parameter will be compressed in-plane whereas the constituent with the smaller lattice parameter will be expanded. Due to the Poisson response, opposite changes will occur for the out-of-plane
2.1 Crystalline structure of thin films and multilayers

lattice parameters. The regime in which the constituents have the same in-plane lattice parameter is called the coherent regime. The elastic energy of the crystal associated with the coherent regime is proportional to the strained volume. At a certain critical thickness it becomes more energetically favorable to introduce misfit dislocations which reduce the strain. In this regime, which is called incoherent, the in-plane lattice parameters of the constituents are no longer the same. Besides misfit dislocations also other crystalline defects like stacking faults, twin boundaries, interstitials and vacancies are often present. All these defects will influence the properties of thin films, multilayers and superlattices.

![Figure 2](image)

*Figure 2. Two-dimensional layer by layer growth (a), three-dimensional island growth (b) and initial two-dimensional growth followed by three-dimensional growth (c).*

It has been shown that strain also plays an important role in the initial growth morphology of thin films [13-14]. Experimental results point to the existence of three distinct growth modes (Fig. 2). The most preferred growth mode is the two-dimensional growth in which the film grows layer by layer. Two-dimensional growth leads to high crystalline quality. Films can also grow as three-dimensional islands which coalesce. Three-dimensional growth generally leads to textured or poly-crystalline films. The third growth mode can be described as initial layer by layer growth up to a few monolayers film thickness, then the growth mode changes to three-dimensional growth. The growth mode is also strongly affected by the growth temperature, the sputter gas pressure and the surface energies of the deposited materials.
Materials with high surface energy minimize the energy by growing as three-dimensional islands, whereas materials with low surface energy can grow layer by layer.

### 2.1.3 Interfaces

![Figure 3](image.png)

Figure 3. Three different types of interfaces: a perfect interface (a), an interface with roughness (b) and an interface with interdiffusion (c).

The interfaces between the layers can be smooth or rough, sharp or diffuse. Roughness is in general the result of limited surface diffusion during growth. High growth temperatures promote surface diffusion but can also lead to unwanted interdiffusion between the layers. Three different types of interfaces are shown in Fig. 3. The interfaces in a real multilayer or superlattice are never perfect, i.e. some roughness and interdiffusion are always present. Two different types of interfacial roughness can be distinguished, one which is correlated between different layers and one which is random or uncorrelated. The correlated roughness is often induced by the substrate which is not exactly flat but contains steps which are replicated from layer to layer. In most cases, the roughness can be described as lying between these two extremes, the roughness is partially correlated. The roughness can also increase from layer to layer, this is referred to as cumulative roughness. If the growth is not phase locked the incomplete formation of the last monolayer of each individual layer will also give rise to an interfacial roughness. The different types of roughness are schematically depicted in Fig. 4.
2.2 Growth of thin films and multilayers

2.2.1 Physical vapour deposition (PVD)

The two most commonly used methods for preparing thin films and multilayer structures are sputtering and molecular beam epitaxy (MBE). In both techniques, multilayers are prepared by sequential deposition onto a substrate by exposure to different materials in the vapour phase. In deposition by MBE, the different materials are evaporated whereas sputtering is a simple momentum transfer process.

Since sputtering has been used in the present work, a more detailed description of this technique is given. In a vacuum chamber an inert gas is ionized through an electric discharge and accelerated towards a negatively biased solid target and on impact, atoms are ejected. The ejected atoms migrate through the plasma and deposit on the substrate. The applied electric field can be DC (Direct Current), which is suitable for conducting targets (metals) or RF (Radio Frequency) for insulating targets. The most commonly used sputter gas is Ar (argon) because it is inert and also relatively heavy, but also other noble gases like Kr, Ne and Xe can be used. In the case of reactive sputtering, O₂ or N₂ are often used. To make the sputtering process more effective, a strong magnet is placed behind the target to confine the ionized gas. In this way the ionization is enhanced and the plasma can be sustained at sputter gas pressures as low as 1 mtorr. A schematic description of the sputtering process is found in Fig. 5.
The kinetic energy of the sputtered atoms depends on the strength of the applied electric field and on the atomic mass ratio between the target material and the sputter gas. A small mass ratio will result in sputtered atoms with high kinetic energy. The sputtering rate is a function of both the strength of the applied electric field and the sputter gas pressure. A higher pressure results in a higher sputtering rate. The sputter gas pressure is usually 1-50 mtorr and typical film deposition rates are 0.5-10 Å/s. The deposition rate at the substrate is also strongly dependent on the target material, size of target and distance between target and substrate. By varying the pressure of the sputter gas it is possible control the energy distribution of the sputtered atoms. At low sputter gas pressures, the mean free path of the sputtered atoms is large and they can move from the target to the substrate without colliding with other atoms. The kinetic energy per atom can be as high as several electron volts. This energy will increase the surface mobility of the deposited atoms. The mean free path of the sputtered atoms decreases as the sputter gas pressure is increased. A reduced mean free path will result in an increased collision frequency and the sputtered atoms will lose kinetic energy by gas scattering. If the sputter gas pressure is high enough, the sputtered atoms can not travel from the target to the film without colliding with other atoms, they are thermalized.
2.2 Growth of thin films and multilayers

Experiments have shown that there is a clear transition in the growth morphology of thin films at the thermalization pressure [15].

Ions from the plasma can also be accelerated towards the growing film by applying a negative potential to the substrate. These energetic ions will affect the growth conditions of the film. Both increased as well as decreased surface/interface quality has been observed in thin films and multilayers which have been ion irradiated during growth [16]. Another important film growth parameter is the substrate temperature as discussed above.

2.2.2 Freyja

Growth of thin films, multilayers and superlattices requires ultra-high-vacuum (UHV) conditions in order to obtain high quality films free of contaminants. In UHV the pressure is of the order of $10^{-8}$-$10^{-10}$ torr. The particle density at $10^{-10}$ torr is about $10^{12}$ times lower than at atmospheric pressure. The time it takes to form a monolayer of N$_2$ at $10^{-10}$ torr is of the order of 6-7 hours which should be compared to nanoseconds at atmospheric pressure. As described above, in deposition by sputtering a gas is necessary in order to create a plasma. This gas, usually argon, should be of very high purity. If the background pressure is too high or if the sputter gas is not pure enough contaminants will be incorporated into the growing film and affect the quality as well as the properties of the film. Furthermore, the materials to be deposited should also be of as high purity as possible.

The UHV based sputtering system Freyja which has been used to deposit the Fe/Co and Fe/V superlattices in this thesis is pumped by a 120 l/s turbomolecular pump, and two cryopumps. After bake out at 150 °C a base pressure of $10^{-10}$ torr is reached. The purity of the argon gas used is 99.9999% and the purity of the target materials is 99.95% (Fe, Co and Pd) and 99.7% (V). Freyja has four magnetrons (1” or 1.3”) which can be operated with either DC of RF power.
Chapter 2  Crystalline structure and growth of thin films

The sample can be heated to about 800 °C and rotated during deposition to prevent film thickness gradients. The pneumatic shutters in front of each magnetron are computer controlled. The system is also equipped with an in situ RHEED normally operated at 10 kV.

2.3 Growth of Fe, V and Co

The magnetic properties of a material and its structure are strongly linked. In each group of the transition metals the elements tend to have the same crystalline structure. For example, V which is a non-magnetic 3d transition metal has the body-centered cubic (bcc) crystal structure just like the other elements in its group. However, this regularity is broken by the ferromagnetic 3d transition metals Fe and Co. Fe has the bcc crystal structure instead of the hexagonal close packed (hcp) like the other elements in its group. Co on the other hand has in the bulk a hcp structure instead of the expected face-centered cubic (fcc) structure. These deviations can be attributed to a magnetic contribution to the energy of the crystal which alters the ground state structure.

2.3.1 Fe and V

As previously discussed, epitaxial growth of single-crystalline films requires matching the film and substrate with respect to lattice parameters, atomic positions and crystallographic orientation. MgO substrates with the (001) orientation are ideal for growing epitaxial single-crystalline films of Fe and V. If Fe and V are combined and deposited on top of each other an Fe/V (001) superlattice is obtained. The epitaxial relationship is [17]:

\[
\text{Fe(V)} [001] \parallel \text{MgO} [001],
\]

\[
\text{Fe(V)} [110] \parallel \text{MgO} [100].
\]
Fe and V have lattice parameters 2.8664 Å and 3.0274 Å, respectively. This arrangement gives a nominal lattice mismatch of about 3.5% between Fe and MgO and 2% between V and MgO whereas the mismatch between Fe and V is about 5.5%.

2.3.2 Body-centered cubic Co

Crystal structures that are not thermodynamically stable in the bulk can be stabilized as thin epitaxial films. The body-centered cubic (bcc) Co phase which is not found in nature is an example [18-21]. Bulk Co is at ambient pressure hexagonal close packed (hcp) up to a temperature of about 425 °C where it transforms to the face-centered cubic (fcc) structure. The FeCo alloy system on the other hand has a bcc crystal structure up to a Co concentration of 75% [22]. Higher concentrations yield mixed phases of bcc, fcc and hcp. Linear extrapolation of the FeCo alloy lattice parameter gives an estimated lattice constant of 2.82 Å for pure bcc Co. GaAs as well as MgO substrates can be used to stabilize the bcc Co phase. The advantage of using GaAs is the very small lattice mismatch. X-ray diffraction measurements on a (011) oriented bcc Co film grown on GaAs (011) have indicated a lattice parameter of 2.827 Å for the bcc phase, which gives a lattice mismatch of about 0.2%. For MgO (001) substrates, on the other hand, the lattice mismatch is larger, about 5%, but an Fe buffer layer can be used to reduce it to about 1.7%. Fe and bcc Co grow on top of each other just like Fe and V, i.e. an Fe/Co (001) superlattice on MgO (001) has the same epitaxial relationship as an Fe/V (001) superlattice. The advantage of using MgO is that it is more thermally stable than GaAs and it is also easier to prepare. It is also possible to stabilize the fcc Co phase as a thin film. Thus, thin films of Co can have the hcp, the bcc and the fcc crystal structure at ambient pressure and temperature [23].
Chapter 3

Thin film magnetism

"Few subjects in science are more difficult to understand than magnetism."


### 3.1 The magnetic moment

Magnetism has its origin in the intrinsic magnetic moment of the electron (spin magnetic moment) and the motion of the electrons around the atomic nucleus (orbital magnetic moment). Quantum mechanics restricts the spin of the electron to have two possible projections along a certain direction. These quantum mechanical states are referred to as 'up' or 'down'. All free atoms with unfilled shells are magnetic and each unpaired electron contributes with 1 Bohr magneton ($\mu_B$) to the total spin magnetic moment. For isolated atoms, there is also a contribution from the orbital angular momentum of the electrons to the total magnetic moment. However, in the 3d transition metals it has been found experimentally that the orbital magnetic moment is approximately zero, it is quenched. The 3d transition metals V, Fe and Co have the electronic configurations; [Ar]3d$^3$4s$^2$, [Ar]3d$^6$4s$^2$ and [Ar]3d$^7$4s$^2$, respectively. This gives, if Hund’s rules are applied, the magnetic moments 3 $\mu_B$, 4 $\mu_B$ and 3 $\mu_B$, respectively for the free atoms.

In bulk form on the other hand, V loses its magnetism whereas Fe and Co still are magnetic but with a much lower average magnetic moment per atom, 2.2 $\mu_B$ and 1.7 $\mu_B$, respectively. The reason is that the outer electrons now are free electrons and are no longer bound to a particular atom. Whether a bulk metal is magnetically ordered or not depends on the balance between the kinetic energy and the exchange Coulomb energy resulting from the Pauli exclusion principle.
A parallel alignment of the spin magnetic moments reduces the exchange energy but increases the kinetic energy of the electrons and vice versa. The famous Stoner criterion for ferromagnetism is a statement of this energy balance:

\[ g(E_F)I > 1, \]

where \( I \), the exchange integral, is a measure of the Coulomb interaction between the spins. The increase of the total kinetic energy of the electrons is described by \( g(E_F) \), which is the density of states of the electrons at the Fermi energy. Thus, for a bulk material to be ferromagnetic both the exchange energy and the density of states at the Fermi energy should be large. In the 3d transition metal series only Fe, Co and Ni satisfy the Stoner criterion and are ferromagnetic, V does not.

In thin films the number of nearest neighbours is reduced compared to bulk. A monolayer thick film is bounded by two interfaces and can be regarded as a two-dimensional crystal whereas the topmost atomic layer of a thicker film has a dimensionality lying between the dimensionality of a single monolayer and bulk. This reduced dimensionality leads to an enhancement of the average magnetic moment per atom. This has been verified theoretically as well as experimentally. For example, theoretical calculations have predicted a magnetic moment of 2.87 \( \mu_B \) for a single atomic monolayer of V which is very close to the value for a free atom [24]. Furthermore, measurements have shown that the average magnetic moment for a rough Fe surface is significantly enhanced [10].

### 3.2 Magnetic anisotropy

#### 3.2.1 Magnetocrystalline anisotropy

In the ferromagnetic 3d transition metals, Fe, Co and Ni, the spin-orbit interaction couples the total magnetic moment to the crystal axes.
3.2 Magnetic anisotropy

This results in an alignment of the magnetic moments along certain crystallographic directions. In zero applied magnetic field the moments are aligned along so-called magnetic easy directions which correspond to the lowest energy. To align the magnetic moments in other directions, an applied magnetic field is necessary. The magnetocrystalline anisotropy energy for a material with cubic symmetry is given by the expression

$$E_{mc} = K_1(\alpha_1^2\alpha_2^2 + \alpha_2^2\alpha_3^2 + \alpha_3^2\alpha_1^2), \quad (2)$$

where the $\alpha$ are the direction cosines of the magnetization relative to the crystal axes and $K_1$ is the first order anisotropy term (the higher order terms have been omitted) [25]. In Fe ($K_1 > 0$) the [100] direction is the easy direction whereas the [110] is the intermediate and the [111] is the hard. In Ni ($K_1 < 0$) on the other hand, the [111] direction is the easy axis whereas the [100] is the hard.

3.2.2 Magnetoelastic anisotropy

Strain changes the magnetocrystalline anisotropy and can thereby alter the direction of the magnetization. The energy associated with this effect can for a crystal with a cubic symmetry be written as

$$E_{me} = -\frac{3}{2}\lambda_{100}\sigma(\alpha_1^2\gamma_1^2 + \alpha_2^2\gamma_2^2 + \alpha_3^2\gamma_3^2) - 3\lambda_{111}\sigma(\alpha_1\alpha_2\gamma_1\gamma_2 + \alpha_2\alpha_3\gamma_2\gamma_3 + \alpha_3\alpha_1\gamma_3\gamma_1), \quad (3)$$

where the $\lambda$ are the magnetostriction constants which can be either positive or negative and $\sigma$ is the stress [26]. The $\alpha$ and the $\gamma$ are the direction cosines relative to the crystal axes and the stress respectively. The magnetoelastic contribution to the magnetic anisotropy is especially important in multilayers and superlattices because they are often strained due to the lattice mismatch between the constituents.

3.2.3 Shape anisotropy
Chapter 3  Thin film magnetism

Also the shape of the magnetic material will affect the magnetic anisotropy. The shape anisotropy originates from the long range magnetic dipolar interaction between the magnetic moments and favours, for thin films an in-plane orientation of the magnetic moments. The shape anisotropy energy is in this case given by the equation

\[ E_s = \frac{1}{2} \mu_0 M^2 \cos^2 \theta, \]  

where \( M \) is the magnetization and \( \theta \) is the angle relative to the film normal [27].

3.2.4 Volume and surface contributions

The magnetic anisotropy in thin films and multilayers can be markedly different from bulk materials. As the film thickness is reduced, surfaces and interfaces become more important. The magnetic anisotropy can be manipulated by varying the thickness of the layers. A very dramatic example of this is the phenomenon refered to as perpendicular magnetic anisotropy in which the direction of magnetization in a multilayer is changed from an in-plane orientation to an out-of-plane by varying the thickness of the layers [5-7]. This effect is attributed to the surface or interface anisotropy which favours an out-of-plane orientation of the magnetic moments. Contributions from surfaces and interfaces can be taken into account by defining an effective anisotropy constant:

\[ K^{\text{eff}} = K_v + \frac{K_s}{t}, \]  

where \( K_v \) and \( K_s \) are the effective volume and surface anisotropy constants respectively and \( t \) is the film thickness. The anisotropy energy can then be written as

\[ E = -K^{\text{eff}} \cos^2 \theta, \]  

where \( \theta \) is the angle of the magnetic moments relative to the film normal [27]. It should be pointed out that the volume contributions are not necessarily independent of the film thickness. Strain can in some cases induce a \( 1/t \) dependence in \( K_v \). Thus, the above expression should therefore be used with caution.
As discussed above, surfaces and interfaces are never perfect, roughness and interdiffusion are always present and influence the magnetic anisotropy [28]. Furthermore, the magnetic anisotropy is also strongly temperature dependent [29].

3.3 Interlayer exchange coupling and giant magnetoresistance

3.3.1 Interlayer exchange coupling

The magnetic moments in the different layers in a multilayer can also be affected by the magnetic moments in the other layers. Often multilayers are grown so that magnetic layers are separated by non-magnetic spacer layers. It has been found experimentally that the magnetic layers can couple to each other across these spacer layers. While each magnetic layer is ferromagnetic, the magnetic moments of adjacent layers are aligned parallel or antiparallel. The latter type of magnetic coupling is referred to as antiferromagnetic interlayer exchange coupling and has been observed in almost all magnetic/non-magnetic multilayers [30]. Also other types of arrangements are possible, one is a perpendicular orientation of the magnetic moments in the different layer, a so-called biquadratic coupling. The magnetic moments usually have an in-plane orientation due to the shape anisotropy. Furthermore, the exchange coupling has also been found to oscillate from ferromagnetic to antiferromagnetic and so forth as the thickness of the non-magnetic spacer layers is varied (Fig. 6) [2-3]. Not only the type of coupling changes but also the strength, which decreases rapidly with increased spacer layer thickness. The exchange coupling is also strongly dependent on the thickness of the magnetic layers. The strength of the antiferromagnetic exchange coupling can be estimated from

\[ J = \frac{\mu_0 M_s H_{sat} d}{4}, \]  

(7)
Chapter 3  Thin film magnetism

Figure 6. The strength of the interlayer exchange coupling as a function of spacer film thickness. Note that the type of coupling also changes with spacer film thickness.

where $M_s$ and $H_{\text{sat}}$ are the saturation magnetization and saturation field respectively and $d$ is the thickness of the magnetic layers [31]. Different models have been proposed to explain the interlayer exchange coupling. The two most commonly used are the quantum well description and the RKKY model. However, it has been shown that both models are the limiting cases of a more general theory [32-34].

3.3.2 Giant magnetoresistance

The giant magnetoresistance (GMR) effect is closely linked to the interlayer exchange coupling. It has been found that antiferromagnetically coupled multilayers exhibit a very large change in electrical resistance when a current is passed in the direction parallel to the layers and the magnetic configuration is changed from antiferromagnetic to ferromagnetic by an applied magnetic field. The relative change can be as large as 50-100% in multilayers like Fe/Cr and Cu/Co [3-4, 35]. This should be compared to the ordinary magnetoresistance effect in bulk materials which shows a relative
3.3 Interlayer exchange coupling and giant magnetoresistance

change of only 1-2%. It should also be mentioned that the giant magnetoresistance effect is observed as well if the current is passed in the direction perpendicular to the layers. A typical magnetoresistance curve is shown in Fig. 7.

![Magnetoresistance curve](image)

Figure 7. Magnetoresistance of an antiferromagnetically coupled [Fe(7 ML)/V(13 ML)]₃₀ (001) superlattice. The magnetic field and the current are in the plane of the film.

The giant magnetoresistance effect can be explained in terms of spin dependent electron scattering (Fig. 8). In a magnetic material there are two types of electrons; those with their spin aligned with the magnetization and those with their spin aligned opposite to the direction of magnetization. It is known experimentally that the probability of electron scattering in which the orientation of the spin changes is very low. Thus, the current carried by the spin up electrons is independent of the current carried by the spin down electrons. In the ferromagnetic 3d transition metals Fe, Co and Ni the spin up and spin down d bands are split and the densities of states at the Fermi energy are therefore very different (Fig. 9). Since the scattering probability is proportional to the density of states at the Fermi energy, the spin up and spin
Figure 8. Schematic illustration of the principle of giant magnetoresistance. The scattering probability of the spin up and spin down electrons depends on the magnetic configuration. Antiferromagnetic (a) and ferromagnetic configuration (b).

down electrons will experience different resistivities. Furthermore, it also means that the resistivities of the spin up and spin down electrons in a multilayer will depend on the magnetic configuration, i.e. if the magnetic layers are antiferromagnetically or ferromagnetically aligned. The electrical resistance of the multilayer is lower in the ferromagnetic state than in the antiferromagnetic as seen in Fig. 7.

The magnitude of the magnetoresistance effect in a multilayer generally depends on two parameters; the spin asymmetry ratio and the mean free path of the electrons. The spin asymmetry ratio is the ratio of the spin up and spin down resistivities in the magnetic material and the mean free path is the average distance an electron moves before it is scattered. The combination of a large spin asymmetry ratio and a long electron mean free path yields a large magnetoresistance effect. It should be pointed out that the mean free path in each layer must be longer than the thickness of the layer in order to observe a giant magnetoresistance effect. If the spin asymmetry is zero or if the mean free path is very short no magnetoresistance effect will be observed. The magnitude of the magnetoresistance effect is also related the strength of the exchange coupling, a strong coupling gives a large magnetoresistance effect and vice versa.
3.3 Interlayer exchange coupling and giant magnetoresistance

![Schematic representation of the densities of states in the 4s and 3d bands of ferromagnetic Fe (left) and Co (right). The bands are filled up to the common Fermi level (E_F).](image)

It has been assumed in this discussion that the spin dependent scattering takes place in the ferromagnetic layers (bulk scattering) but experimental results have indicated that scattering at the interfaces also plays an important role (interface scattering). Increased as well as decreased magnetoresistance has been observed with increased interface roughness. For example, Fe/Cr multilayers show an enhanced magnetoresistance with increased interface roughness whereas Cu/Co multilayers exhibit the opposite behaviour. Thus, it is possible that the relative importance of the bulk and interface scattering varies strongly from material system to material system [36-39].
Chapter 4

Characterization techniques

4.1 X-ray diffraction (XRD)

X-ray diffraction is a very powerful technique for investigating the structural properties of thin films, multilayers and superlattices. Information on crystal structure as well as on crystalline quality can be obtained. It is a non-destructive method and no additional sample preparation is needed. This chapter gives a review of the basic theory of x-ray diffraction along with a discussion of how the structural properties of superlattices can be determined by investigating the reciprocal space.

4.1.1 X-ray diffraction and the reciprocal space

An ideal crystalline solid material consists of an infinite array of identical structural units. These units may be single atoms, groups of atoms or molecules. The structure of the solid can be defined in terms of a lattice, with a group of atoms called a basis attached to every lattice point. An infinite three-dimensional lattice can be described by

\[ L(\vec{r}) = \sum_{x,y,z} \delta(\vec{r} - \vec{r}_{x,y,z}), \]

where \( \delta \) is the Dirac delta function and \( \vec{r}_{x,y,z} = a\vec{x} + b\vec{y} + c\vec{z} \) is the generic lattice vector.

The electron distribution of the basis of a crystal can be represented by a function \( \rho(\vec{r}) \) and the distribution for the whole crystal is then the convolution

\[ \rho_w(\vec{r}) = \rho(\vec{r}) * L(\vec{r}). \]

X-rays are scattered by the electrons in the crystal and the Fourier transform (T) of the electron distribution gives the amplitude and the phase of the scattered wave:
Chapter 4  Characterization techniques

\[ T[r_{\infty} \rho] = T[\rho \cdot L(r)], \quad (10) \]

where

\[ T[L(r)] = \frac{1}{V} \sum_{h,k,l=\infty}^{\infty} \delta(z - \bar{s}_{h,k,l}) \quad (11) \]

and

\[ T[\rho(r)] = \int \rho(r) \exp(2\pi i \bar{s} \cdot r) dV. \quad (12) \]

\( \bar{s}_{h,k,l} \) is the generic lattice vector of the reciprocal lattice and \( V \) is the volume of the unit cell. The electron density in Eq. 12 can be replaced by the atomic scattering factor, \( f_i \), for each atom in the unit cell:

\[ \rho(r) \rightarrow \sum_i f_i \delta(r - r_i), \quad (13) \]

where \( f_i \) is the effective number of electrons concentrated at the atom center and the summation runs over all the atoms in the unit cell. Thus, the well known expression for the structure factor, \( F_{hkl} \), is obtained:

\[ T[\rho(r)] = F_{hkl} = \sum_i f_i \exp(2\pi i \bar{s} \cdot r_i). \quad (14) \]

The structure factor is the effective number of electrons concentrated at the cell origin. The intensity of the scattered x-rays is proportional to the square of the modulus of the wave scattered from the crystal:

\[ I_{hkl} \propto |T[r_{\infty} \rho]|^2 = N^2 |F_{hkl}|^2, \quad (15) \]

where \( N \) is the number of unit cells in the crystal. From Eq. 11 it can be seen that the amplitude of the scattered wave is non-zero only if the scattering vector is a vector of the reciprocal lattice (the Laue diffraction condition). Thus, the electron distribution in the unit cell determines the intensity as well as the direction of x-rays scattered from a crystal. The scattering vector, \( \vec{K} \), is defined as

\[ \vec{K} = \frac{1}{2\pi} (\vec{k}_d - \vec{k}_i), \quad (16) \]

where \( \vec{k}_d \) and \( \vec{k}_i \) are the diffracted and incident wave vectors respectively. The magnitude of \( \vec{k}_d \) and \( \vec{k}_i \) is \( 2\pi / \lambda \).
4.1 X-ray diffraction (XRD)

It should be pointed out that the measured intensity is less than the one given by Eq. 15 owing to various instrumental correction factors such as the Lorentz factor, the polarization factor, the absorption and the Debye-Waller factor. Furthermore, the atomic scattering factor, $f_i$, is not a constant but depends strongly on the scattering vector. The theory outlined above is the kinematical x-ray scattering theory in which it is assumed that the x-rays are only scattered once and that the energy of the radiation is conserved [40-44].

4.1.2 Crystalline quality and coherence

The coherence of a crystal is often described by the structural coherence length. Crystalline defects limit the coherence length. For a perfect single-crystalline material the coherence length is infinite in all directions whereas it is zero for a material with no structural order. In reciprocal space, limited long range order in a three-dimensional crystal will change the position, shape and weight of the reciprocal lattice points. Thus, by examining the these points, information on the disorder can be obtained. There are different ways to define a relationship between the structural coherence length ($\zeta$) and the width of a reciprocal lattice point. One relation that can be used is

$$\zeta = \frac{1}{\Delta K},$$

(17)

where $\Delta K$ is the linewidth (FWHM in Å⁻¹) of the diffracted x-ray beam corresponding to a certain reciprocal lattice point. It should be noted that $\zeta$ is the structural coherence length in the direction defined by the scattering vector. By investigating different reciprocal lattice points the coherence length in different crystallographic directions can be obtained.

As discussed earlier, strain is often present in superlattices due to lattice mismatch between the constituents. If the in-plane lattice parameters of the two constituents are equal in the superlattice the layers are said to be perfectly epitaxial.
In the other extreme the elastic strain is totally relaxed and the constituents have their bulk lattice parameters. In most superlattices however, the strain is partially relaxed, often through the formation of misfit dislocations which introduce a slight orientational spread in the crystal. All these cases can be fully characterized by using x-ray diffraction.

For diffraction to occur in a crystal the wave length of the radiation must be of the same order as the interatomic distance. Therefore is x-ray radiation with a wave length of the order of 1 Å used. The most commonly used radiation is Cu K_α (λ=1.54 Å) but also K_α radiation from other metals like Mo (λ=0.71 Å) can be used. Diffraction also requires the radiation to be coherent. Two different coherence lengths can be distinguished, longitudinal and lateral. The longitudinal coherence length depends on the spectral purity (Δλ/λ) of the radiation whereas the lateral coherence length is determined by the divergence (Δα) of the radiation. The two types of coherence lengths define a volume in real space. An x-ray diffraction measurement furnishes average structural information over this volume. In reciprocal space on the other hand, the coherence lengths will define a volume called the resolution element which is inversely proportional to the volume in real space. Large coherence lengths give a small resolution element which means that the reciprocal space of a crystal can be probed with high accuracy. Monochromators and collimators can be used to increase the coherence of the radiation. A drawback is that they will also reduce the intensity.

In general, an x-ray diffraction measurement probes the smaller of the two types of coherence lengths. Hence, in order to measure a property of a the crystal the coherence length of the x-ray radiation should be much larger than the structural coherence length.

4.1.3 Investigating the reciprocal space
An x-ray diffraction measurement can simply be described as an investigation of the reciprocal space of a material. During a measurement, the scattering vector is scanned in a certain direction in the reciprocal space. When the scattering vector is a vector of the reciprocal lattice diffraction will occur and the intensity of the diffracted x-rays will be given by Eq. 15. Four different types of scans are often used to investigate the reciprocal space (Fig. 10):

1. The radial or offset scan is directed nearly parallel to the reciprocal lattice vector of a particular lattice point. \( \Delta \omega / \Delta 2\theta = 1/2 \). \( \omega \) generally differs from \( 2\theta / 2 \).
2. A special case of the radial scan is the specular scan, which is directed along a reciprocal lattice vector. In this case, \( \Delta \omega / \Delta 2\theta = 1/2 \) and \( \omega \) is equal to \( 2\theta / 2 \).
3. The transverse or rocking scan runs perpendicular to the radial scan. \( 2\theta \) is fixed and \( \omega \) is varied.
4. The detector scan, \( \omega \) is fixed while \( 2\theta \) is varied.

In the definition of the reciprocal space that is used here, the length of the scattering vector is related to the angle \( \theta \) through the expression
\[ K = \frac{2}{\lambda} \sin \theta, \quad (18) \]

where \( \lambda \) is the wavelength of the radiation and \( \theta \) is defined in Fig. 10.

The scans described above are one-dimensional scans across the reciprocal space. It is also possible to map areas in the reciprocal space. In reciprocal space mapping (RSM), radial and transverse scans are combined. Another two-dimensional scan is the texture scan in which the length of the scattering vector is fixed while it is scanned across the reciprocal space.

### 4.1.4 The reciprocal space of a superlattice

High- and low-angle specular x-ray diffraction scans can provide valuable information on the crystalline structure of a superlattice. In the high-angle region (\( 2\theta = 20-110^\circ \)) where \( K \) is about 0.25-1.0 Å\(^{-1}\), the probed length scale (\( \approx 1/K \)) is of the order of the interatomic distance. This means that information on lattice constants, strain state and crystalline quality can be obtained. At lower angles (\( 2\theta = 0-20^\circ \)) on the other hand, the probed length scale is of the order of the superlattice period or longer (\( K < 0.25 \) Å\(^{-1}\)). In this region information on the superlattice period, the interface quality, and the total film thickness can be obtained. The x-ray diffraction data must in general be fitted to a theoretical model in order to obtain quantitative information. Computer models like GIXA or SUPREX are therefore often used [45-47]. It should be pointed out that crystalline imperfections will in general both reduce the intensity of the diffracted peaks as well as broaden them. There is no clear distinction between a particular type of disorder and its effect on a particular feature in an x-ray diffraction scan, all diffraction features are affected to some degree [48-53]. High- and low-angle specular scans from an Fe/V (001) superlattice are shown in Fig. 11.
Figure 11. High-angle (top) and low-angle (bottom) x-ray diffraction scans of an \([Fe(9 \text{ ML})/(11 \text{ ML})]_{30} \text{(001)}\) superlattice. Distinct superlattice satellites are seen at both high and low angles.

4.2 **Reflection high energy electron diffraction (RHEED)**

Reflection high energy electron diffraction (RHEED) is a useful technique for investigating the epitaxial growth of thin films in ultra-high-vacuum (UHV). The grazing incidence geometry (1-3°) makes it very surface sensitive, only the 1-2 outermost atomic layers are probed. It is ideally suited for the study of clean,
Chapter 4   Characterization techniques

well ordered single-crystalline surfaces. RHEED gives information on the surface
crystal structure, the surface orientation and the surface roughness. It can also be used to
determine the growth mode of the film, i.e. whether it grows layer by layer or as three-
dimensional islands.

4.2.1 Basic principles

\[ k_i = k_d \]

Figure 12. The Ewald sphere construction. A cross section in the plane of incidence that shows how a reciprocal lattice rod gives rise to a diffraction maximum.

In RHEED, a high energy (10-100 keV) electron beam is focused on a sample surface. Due to the wave character of the electrons, maxima in the intensity of the diffracted beam will occur for directions satisfying the Laue diffraction condition which states that the change in wave vector must be a vector of the reciprocal lattice. For a surface, which can be described as a two-dimensional lattice, the reciprocal lattice is a two-
dimensional array of infinite rods perpendicular to the surface. The rod spacings are
inversely proportional to the atomic row spacings. Since electrons with RHEED energies and a grazing angle of incidence will be scattered elastically, energy is conserved \((k_i = k_d)\).
4.2 Reflection high energy electron diffraction

The Ewald sphere construction, shown schematically in Fig. 12, provides the schematic description of conservation of energy and the Laue diffraction condition.

4.2.2 Interpretation of RHEED patterns

For a perfect single-crystalline surface, the reciprocal lattice rods are effectively delta functions in width. The intersections of the Ewald sphere with those rods are points. If those points are projected onto a screen, they will appear as spots lying on semicircles. The width of a reciprocal lattice rod in a certain direction is inversely related to the long range order in that direction. This means that surface imperfections like steps, roughness and asperities will change the shape of the reciprocal lattice rods and consequently change the appearance of the RHEED pattern. Limited long range order in the surface plane will broaden the reciprocal lattice rods uniformly. A RHEED pattern from such a surface is shown in Fig. 13 (a). The streaking is caused by the grazing geometry of RHEED.

Figure 13. RHEED patterns of four different types of surfaces.
Chapter 4  Characterization techniques

The RHEED pattern from a well ordered crystalline surface with steps will exhibit diffracted beams which are split (Fig. 13 (b)). If the surface is sufficiently rough, a so-called transmission pattern will be obtained. Electrons passing through the asperities will see the additional periodicity of the atomic planes below the surface. As a result, the reciprocal lattice will be an array of points instead of rods. The regular array of spots of a transmission pattern is shown in Fig. 13 (c). Another commonly observed feature in RHEED is superlattice reflections which appear at fractional spacings between the integral-order reflections (Fig. 10 (d)). Superlattice reflections indicate that a surface reconstruction is present, i.e. the size and the symmetry of the topmost layer are different from the bulk structure. RHEED can also be used to monitor changes in the lattice constant of the topmost layer during growth. The distance between the streaks in a RHEED pattern is inversely proportional to the distance between the surface atoms in the direction perpendicular to the direction of the incident electron beam. Because the relative positions of the streaks can be measured accurately, changes in the in-plane lattice constant can be monitored as function of film thickness. The RHEED pattern will also give information on the symmetry of the surface lattice, i.e. a cubic surface lattice will exhibit a RHEED pattern with cubic symmetry. Excellent descriptions of the RHEED technique are found in [54-56].

4.3 Spin polarized neutron reflectometry

The major advantage of spin polarized neutron reflectometry is that the absolute values as well as the direction of the magnetic moments in the individual layers of a magnetic multilayer can be obtained. Techniques like SQUID and MOKE only provide average sample information. Spin polarized neutron reflectometry is therefore a very powerful method for investigating the magnetic structure in magnetic thin films and multilayers.
4.3 Spin polarized neutron reflectometry

In this chapter the basic theory and principles of spin polarized neutron reflectometry will be outlined.

4.3.1 Basic theory

Like photons and electrons, neutrons have a wave character and will be diffracted if the Laue diffraction condition is satisfied. Neutrons are scattered by the atomic nuclei which means that they can be used to determine the spatial arrangement of atoms in a crystal. Furthermore, neutrons also carry an intrinsic magnetic moment which may interact with the magnetic moments in a magnetic material and give information on the magnetic structure. The effective potential a neutron experiences in a magnetic material can be described by

\[ V = \frac{2\pi\hbar^2}{m_n}Nb - \mu_n \cdot B, \]  

(19)

where \( m_n \) is the neutron mass, \( N \) is the atomic density, \( b \) is the neutron scattering length of the material, \( \mu_n \) is the neutron magnetic moment and \( B \) is the magnetic induction in the material. The first term on the right hand side corresponds to an effective potential associated with the neutron-nucleus interaction. The second term corresponds to the Zeeman interaction and so depends upon the relative orientation of the neutron spin with respect to the magnetic induction within the magnetic material. The neutron plane waves for the spin up (+) and the spin down (-) states in a thin magnetic film are given by the one-dimensional Schrödinger equations:

\[ \left( \nabla^2_z + \frac{K^2}{4} - 4\pi N(b + p\sin\theta) \right)\Psi_+ + (4\pi N p \cos\theta)\Psi_-' = 0, \] 

(20)

\[ \left( \nabla^2_z + \frac{K^2}{4} - 4\pi N(b - p\sin\theta) \right)\Psi_- + (4\pi N p \cos\theta)\Psi_-' = 0. \] 

(21)
Here $K$ is the length of the scattering vector, $p$ is the magnetic scattering length and $\theta$ is angle between the neutron polarization axis and the sample magnetization. As seen, if the magnetization vector of the sample is parallel to the polarization axis ($\theta = 0^\circ$) the spin up (+) and the spin down (-) neutrons will remain in their spin state, no spin-flip will occur. On the other hand, if the magnetization vector makes an angle with respect to the polarization axis the probability for spin-flip is non-zero. The spin-flip probability depends both on the magnitude of the magnetization and its direction. This spin-flip phenomenon is a purely magnetic property and does not occur in non-magnetic materials ($p=0$).

### 4.3.2 Principles of spin polarized neutron reflectometry

In spin polarized neutron reflectometry, a spin polarized neutron beam is directed at low angles ($\theta=0-10^\circ$) towards the sample surface. The beam can be either monochromatic or have a distribution of wavelengths. Thermal neutrons with a wavelength of the order of 1.8 Å are often used.
A schematic outline of the scattering geometry for spin polarized neutron reflectivity studies is shown in Fig. 14. The incident neutrons are polarized with their magnetic moments parallel to a certain direction. The polarization is achieved via a weak guiding magnetic field and a so-called supermirror which separates the spin up (+) and spin down (-) neutrons. A supermirror is also used to analyze the spin direction of the scattered neutrons. Furthermore, spin-flippers are placed between the polarizer and the sample and between the sample and the analyzer. With this experimental arrangement, four different spin dependent reflectivities can be measured; the two non-spin-flip cases (+,+) and (-,-) and the two spin-flip cases (+,-) and (-,+). The two spin-flip cases are usually degenerate, i.e. their reflectivities are equal. By changing the direction of the magnetization with an external magnetic field and analyzing the different spin dependent reflectivities information on the magnetic structure of a sample can be obtained. The experimental data must in general be fitted to a theoretical model in order to obtain quantitative information on magnitude and direction of the magnetic moments. The input parameters are the scattering lengths, atomic density, layer thicknesses and interface quality etc.

It should be pointed out that the scattering geometry described in Fig. 14, can not resolve any z-component of the magnetization vector, only in-plane components can be detected. To resolve any z-component of the magnetization vector, a scattering geometry with the scattering vector parallel to the surface is required. Excellent descriptions of spin polarized neutron reflectometry are found in [57-60].

4.4 Superconducting quantum interference device (SQUID)

SQUID magnetometers are high-resolution magnetometers based on the Josephson effect. They provide the ultimate in resolution for magnetic field measurements, the resolution can be as high as $10^{-14}$ T [61-62].
A SQUID magnetometer measures how the magnetization in a sample responds to an applied magnetic field. Due to the use of superconducting magnets, magnetic fields as high as 5-6 T can be applied. Furthermore, the temperature of the sample can be varied from about 4.2 K (liquid He) to room temperature. It is important to realize that the magnetic properties of the whole sample is measured. In the case of thin films and multilayers, the magnetic properties of the magnetic film as well as the properties of the substrate, buffer layer and capping layer are measured. Thus, only average sample information is obtained from a SQUID measurement as pointed out above.

4.5 Mössbauer spectroscopy

Mössbauer spectroscopy is a technique for probing the interaction between the atomic nuclei and the electric and magnetic fields in solids. The technique is based on the recoilless resonant emission and absorption of $\gamma$-radiation by atomic nuclei bound in solids. This process is referred to as the Mössbauer effect. Recoilless absorption and emission means that no energy is lost due to recoil [63-65].

The by far most important Mössbauer isotope is $^{57}$Fe but also other elements exhibit significant Mössbauer effects. $^{57}$Fe has a 14.4 keV nuclear transition energy between the excited $I=3/2$ state and the $I=1/2$ ground state. It is $\gamma$-rays from this transition that is used for Mössbauer spectroscopy in $^{57}$Fe. Only the $^{57}$Fe isotope, which amounts to 2% of natural Fe, exhibits the Mössbauer effect, $^{56}$Fe does not.

The interactions between an atomic nucleus and the electric and magnetic fields produced by the surrounding electrons are called hyperfine interactions. The hyperfine interactions can be divided into three parts; electric monopole interaction, electric quadrupole interaction and magnetic dipole interaction. These interactions affect the nuclear states in the following way:
• The electric monopole interaction shifts the energy levels up or down. This leads to the isomer shift.
• The electric quadrupole interaction splits the I=3/2 nuclear state into two states; the \( m_I = 3/2 \) and the \( m_I = 1/2 \) state. The I=1/2 ground state is unaffected.
• The magnetic dipole interaction lifts the degeneracy of both the I=3/2 and the I=1/2 nuclear state.

Allowed transitions between different states are given by the selections rules; \( \Delta m_I = 0, \pm 1 \). The isomer shift depends on the electron density at the nucleus. The fact that only the s electrons contribute significantly to that density means, in the case of \( ^{57}\text{Fe} \), that changes in the 4s electron density can be estimated by observing the isomer shift. The electric quadrupole interaction on the other hand depends on the electric field gradient at the nucleus whereas the magnetic dipole interaction depends on the magnetic field. Both fields are determined by the electron distribution in the immediate surroundings of the nucleus. This means that the Mössbauer effect can be used as a local probe to investigate the direction and the strength of the electric and magnetic fields. Hence, information on the structural as well as the magnetic environment in a solid can readily be obtained by measuring the energy levels of the probe atoms. This very special property of Mössbauer spectroscopy can be used to investigate magnetic thin films, multilayers and superlattices. By depositing \( ^{57}\text{Fe} \) atoms at different positions, either at the interfaces or at the interior of a film, information on interface roughness as well as magnetic moments can be obtained.
Chapter 5

Summary of results

This chapter is a short summary of the papers included in this thesis. The reader is referred to the different papers for a more detailed and comprehensive discussion of the results.

5.1 Fe/Co (001) superlattices

Paper I

Structural and magnetic properties of bcc Fe/Co (001) superlattices

This paper discusses the structural and magnetic properties of Fe/Co (001) superlattices deposited on MgO (001) substrates. X-ray diffraction (XRD) is used to determine the crystalline quality and the interface profiles of the superlattices. Reciprocal space mapping (RSM) and texture scans show that the superlattices are single-crystalline with a pure body-centered cubic (bcc) structure. The out-of-plane crystalline coherence length is estimated to be of the order of 300 Å at the optimum growth temperature of 250-300 °C. The low-angle x-ray diffraction data indicates that there is an asymmetry in the interface roughness. The Fe-on-Co (Fe deposited on Co) interfaces are found to have an interface roughness which is somewhat larger than the Co-on-Fe interfaces, 3.0 Å compared to 1.5 Å. The Fe/Co thickness ratio in the superlattices is varied in order to investigate the magnetic anisotropy and the magnetization. The first order anisotropy constant for the body-centered cubic Co phase is determined to be negative with a magnitude of 110 kJ/m$^3$ at 10 K. Furthermore, the magnetic moments in the Fe and Co layers are parallel and behave as a single-component system.
Chapter 5  Summary of results

Finally, the average magnetic moment per atom in the superlattices is found to be significantly enhanced as compared to the average value calculated from the bulk magnetic moments.

Paper II

Structural properties of ultrathin bcc Co (001) layers

Single-crystalline body-centered cubic (bcc) Fe/Co (001) superlattices with different Fe and Co thicknesses are deposited on MgO (001) substrates at different temperatures in order to investigate the structural properties of the bcc Co phase. The crystalline quality is determined from radial and transverse (rocking) x-ray diffraction scans across the Fe/Co (002) superlattice Bragg peak. It is found that superlattices with a high content of body-centered cubic Co show a much stronger temperature dependence than superlattices with a high Fe concentration. The crystalline quality of the Co rich superlattices exhibit a marked improvement with increased growth temperature in contrast to the Fe rich samples. Furthermore, the Co-on-Fe (Co deposited on Fe) interfaces are found to be abrupt up to a substrate temperature of 500 °C whereas the Fe-on-Co interfaces become more diffuse at 350 °C due to interdiffusion.

Paper III

Growth of ultrathin cobalt films on Fe (001) studied by XRD and RHEED

It is well known that thin films of the body-centered cubic (bcc) Co phase can only be stabilized up to a certain number of atomic monolayers. The exact thickness has not previously been determined. In this investigation, thin layers of bcc Co (001) are deposited on single-crystalline Fe (001) films in order to determine the stability limit and to study the surface crystalline structure.
5.2 Fe/V (001) superlattices

The reflection high energy electron diffraction (RHEED) investigation reveals that there is a structural phase transformation from the body-centered cubic phase to the hexagonal close packed (hcp) at about 8 atomic monolayers and that the bcc Co surface is more atomically rough as compared to the Fe buffer layer but improves with increased growth temperature. An x-ray diffraction (XRD) investigation of a thick Co film (300 Å) show that the hcp phase is textured and contains a small amount of Co with the face-centered cubic structure.

Paper IV

Interface roughness/intermixing and magnetic moments in a Fe/Co (001) superlattice

This paper is a Mössbauer spectroscopy study of the Fe magnetic moment distribution in a symmetric $[^{57}\text{Fe}(5 \text{ ML})/\text{Co}(5 \text{ ML})]$ (001) superlattice deposited on a MgO (001) substrate. The experimentally obtained magnetic moment distribution as a function of the distance from the Fe/Co interface is found to correlate very well with recent theoretical calculations. The results show that the magnetic moment of Fe is enhanced at the interfaces whereas the moment just a few atomic monolayers away from the interfaces is bulk like. Furthermore, the obtained results also indicate that the Fe/Co interfaces are abrupt without any detectable interdiffusion. The sensitivity of Mössbauer spectroscopy as local probe method is demonstrated by a comparison with x-ray diffraction (XRD). The Mössbauer results clearly show that two different types of superlattices, in this case the Fe/Co superlattice and an $[^{57}\text{Fe}(5 \text{ ML})/\text{V}(5 \text{ ML})]$ (001) superlattice, can have very different interface profiles although x-ray diffraction data indicates the opposite.

5.2 Fe/V (001) superlattices
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Paper V
Interlayer exchange coupling and magnetoresistance in Fe/V (001) superlattices

It has been shown that the ferromagnetic Fe layers in Fe/V superlattices can couple to each other across the non-magnetic V spacer layers ferromagnetically or antiferromagnetically. The type of coupling as and the strength depend on the thickness of the Fe and V layers. In this study, the influence of the Fe and V thicknesses on the interlayer exchange coupling and the giant magnetoresistance (GMR) effect in Fe/V (001) superlattices deposited on MgO (001) substrates are examined. It is found that the strongest interlayer exchange coupling is obtained if the Fe layers are 5-6 atomic monolayers and the V layers are 13 atomic monolayers thick. This is also true for the giant magnetoresistance effect. For other combinations of Fe and V, the exchange coupling is weaker. Furthermore, it is also shown that the exchange coupling is antiferromagnetic only for a limited number of thickness combinations. The thickness of the Pd capping layer is found to strongly affect the magnitude of the giant magnetoresistance. The magnetoresistance effect for an [Fe(5 ML)/V(13 ML)] superlattice with a 20 Å thick Pd layer is about 8%. This should be compared to a relative change of only 3% for a superlattice with a 100 Å thick Pd layer. The capping layer reduces the amount of current that goes through the superlattice and thereby changes the magnetoresistance ratio.

Paper VI
A study of the different interfaces in Fe/V superlattices

There are two different types of interfaces in an Fe/V superlattice, the Fe-on-V (Fe deposited on V) interface and the V-on-Fe interface. In this paper these two different interfaces are investigated by Mössbauer spectroscopy.
Two types of single-crystalline Fe/V (001) superlattices were deposited on MgO (001) substrates with \(^{57}\)Fe as a local probe; two different samples with two atomic monolayers of \(^{57}\)Fe at the Fe-on-V interfaces and two different samples with two atomic monolayers of \(^{57}\)Fe at the V-on-Fe interfaces. The Mössbauer investigation shows that the V-on-Fe interfaces are more abrupt and less diffuse than the Fe-on-V interfaces. This is consistent with reflection high energy electron diffraction (RHEED) measurements which indicate a somewhat rougher V surface as compared to Fe. Furthermore, it can also be concluded from the Mössbauer data that the interlayer ferromagnetic exchange interaction is weaker when the non-magnetic spacer layers are thicker and that the intralayer ferromagnetic exchange interaction is weaker when the magnetic layers are thinner.

**Paper VII**

**In-plane magnetic anisotropy of Fe/V (001) superlattices**

Lattice mismatch between the constituents in a superlattice will give rise to strain in the crystal structure. This is seen in a series of Fe/V (001) superlattices deposited on MgO (001) substrates where the Fe layer thickness is constant (15 atomic monolayers) and the thickness of the V layers is varied (1-12 atomic monolayers). The average in-plane and out-of-plane lattice parameters of the Fe/V superlattices, as determined by reciprocal space mapping (RSM), are found to increase with V layer thickness. The results are compared to a simple theoretical model which neglects the influence of the substrate and the interfaces. The experimental lattice expansion is found to be in good agreement with the theoretical results. The observed deviations may be attributed to strain relaxation in the crystal. Furthermore, if the anisotropy energy, given by the area enclosed by the [100] and [110] magnetization curves, is extrapolated to zero strain, a lower value than in bulk Fe is obtained. This may be explained by an interface anisotropy term which favours the [110] direction.
Finally, the measurements also indicated a somewhat lower saturation magnetization in the superlattices as compared to bulk Fe. This reduction is attributed to a negatively coupled magnetic moment in the V layer at the interface and a reduced Fe moment at the interface, a fact that has also been obtained in theoretical calculations.


