Soft X-Ray Resonant Scattering from Magnetic Heterostructures

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Johannes Grabis

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Erstgutachter Prof. Dr. Dr. h.c. H. Zabel
Zweitgutachter Prof. Dr. J. Pelzl
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Introduction

Heterogenous magnetic materials such as multilayers and superlattices of ferromagnetic metals with non-ferromagnetic spacer layers are of great interest both because of their relevance for current and future technological applications and since they provide model systems to understand magnetic behavior and interactions.

Magnetic properties of artificial multilayered media are strongly influenced by the preparation conditions. The morphology of the interface and particularly its roughness play a major role for the electronic and magnetic properties. The reliable determination of lateral and perpendicular interface characteristics is crucial for further developments in material science. X-ray scattering is a standard tool for non-destructive studies of the structure of buried layers and interfaces. While not yielding direct imaging information, as obtained by the complementary techniques of electron microscopy, scanning tunneling microscopy or atomic force microscopy, x-ray scattering is capable of examining quantitative global statistical information. Conventional x-ray techniques are sensitive to the electron density, i.e. structural information only. For a long time polarized neutron reflectometry (PNR) was the only tool yielding similar information on the magnetic structure.

The availability of high brilliance synchrotron sources with tuneable photon polarization enabled the application of magneto-optical effects in the hard and soft x-ray ranges. By tuning the photon energy close to a specific absorption edge, not only much larger effects than in laser-based investigations are observed, but it is also possible to study the magnetism of different elements separately. The most prominent among these effects is x-ray magnetic circular dichroism (XMCD) \cite{1}, because it allows the quantitative, element-specific determination of spin and orbital magnetic moments via sum rules with sub-monolayer sensitivity \cite{2, 3}. XMCD is defined as the difference in the absorption coefficients of right and left circularly polarized light in a sample with the magnetization along the photon propagation direction. In case of 3$d$ transition metals the absorption process involves electronic transitions from 2$p$ core states to 3$d$ valence states lying in the soft x-ray range of 500-1000 eV.

X-ray resonant magnetic scattering (XRMS) combines the advantages of XMCD and conventional x-ray scattering \cite{4, 5}. It provides not only an alternative tool to PNR but also yields additional and complementary information. While neutrons probe the magnetic induction in the sample, resonant x-rays are sensitive to the atomic magnetic moment. The high flux of modern synchrotron sources allows a much smaller sample volume and a faster data acquisition as compared to neutron scattering. The surface sensitive of soft x-rays due to the shorter penetration depth can be an advantage in the field of thin film magnetism.

Within the framework of the present thesis the new diffractometer ALICE for soft
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XRMS has been constructed. The instrument comprises a two-circle goniometer and works in horizontal scattering geometry. It opens up the possibility of XRMS measurements at low temperatures down to 30 K in presence of a variable external magnetic field up to ±0.27 T. This large parameter range is unique at least in Germany. The diffractometer was tested successfully for the first time at BESSY II, the synchrotron facility in Berlin, in December 2002. Since then XRMS measurements have been performed on various magnetic multilayer systems and nanostructures. Two of the studied physical systems are presented in this thesis:

Antiferromagnetically exchange-coupled superlattices represent ideal model systems to demonstrate the feasibility of soft x-ray resonant magnetic scattering. The Fe/Cr(001) system has been one of the most frequently and thoroughly studied systems in the field of exchange-coupled thin films over the last two decades and exhibits a large variety of magnetic phenomena [6, 7, 8]. Astonishingly, there is no literature available about soft x-ray resonant magnetic scattering of Fe/Cr superlattices, although the oscillatory interlayer exchange-coupling was observed first in this transition metal system. In this study antiferromagnetically and non-collinearly coupled Fe/Cr superlattices are studied by XRMS with linearly and circularly polarized light at the Fe $L_{2,3}$ absorption edges. It is demonstrated that a layer-resolved vector magnetometry is possible, similar to the case of PNR [9].

The study on Co$_2$MnGe Heusler thin films and superlattices takes full advantage of the element-sensitivity of XMCD and XRMS. The ternary Heusler alloy is predicted to be a ferromagnetic half metal possessing a 100% spin polarization at the Fermi level [10]. This rare property predestinates it to be ideally suited as electrode in spintronic devices e.g. for spin injection [11], spin filtering [12] or in tunnelling magnetoresistance (TMR) applications [13]. The Co$_2$MnGe compound grows in the $L_{21}$ structure – a cubic lattice combining four fcc sublattices occupied by the Co, Mn and Ge atoms, respectively. The focus in the present work is on the effect of site disorder both in the film and at the interfaces, which has been suggested to be the main reason for the moderate performance of GMR and TMR devices based on the fully spin polarized Heusler compounds achieved until now [14, 15]. Perfectly ordered $L_{21}$ symmetry is difficult to achieve even in bulk single crystals of the Heusler phase. In thin film heterostructures, which must be processed at rather low temperatures in order to prevent excessive interdiffusion at the interface, site disorder can hardly be avoided completely. While XMCD is an ideal tool to study the element-specific magnetic moments, only resonant magnetic scattering enables to determine a magnetization depth profile across the interface to the non-ferromagnetic layer.

The present thesis is divided into three parts: The first part provides the theoretical background to the magneto-optical effects at soft x-ray wavelengths. The different techniques used in the present study are discussed: XMCD (Chap. 2), XRMS in the specular reflectivity (Chap. 3) and in the off-specular diffuse scattering (Chap. 4). A matrix formalism is presented which allows the calculation of the specular reflected intensity for radiation of arbitrary polarization and incidence angle onto structures having arbitrary magnetization distributions. The second part deals with the instrumentation necessary to perform XRMS experiments. After a brief discussion of the properties of synchrotron radiation (Chap. 5) the ALICE diffractometer is described in Chap. 6. The experimental results are discussed in the third part starting with the measurements
on interlayer exchange-coupled Fe/Cr superlattices (Chap. 7). Subsequently, x-ray absorption spectroscopy and magnetic dichroism studies on a Co$_2$MnGe thin film at the $L_{2,3}$ absorption edges of Co and Mn are presented in Chap. 8. The magneto-optical constants determined from the absorption measurements are used in Chap. 9 for the quantitative analysis of the XRMS measurements on a [Co$_2$MnGe/Au]$_{50}$ multilayer in order to determine element-specific magnetization depth profiles of Co and Mn.
Introduction
Part I.

X-ray resonant magnetic scattering
1. Magneto-optical effects

1.1. Introduction

1.1.1. Overview of magneto-optical effects

Magneto-optical (MO) effects generally describe the dependence of optical and spectroscopic properties on the magnetic order in a solid. A huge variety of MO effects is observed at visible and x-ray wavelengths, depending on the geometry and measured quantity in an experiment:

- The first magneto-optical effect was already observed in 1845 by Faraday. The Faraday effect describes the rotation of the polarization vector during transmission of linearly polarized light through a magnetic material \[16, 17, 18\].

- The analogous effect in reflection, the magneto-optical Kerr effect (MOKE), was observed much later in 1877. There are typically three different geometries used to measure the Kerr effect: In the polar Kerr (P-MOKE) geometry the magnetization is perpendicular to the sample surface. In the longitudinal Kerr (L-MOKE) geometry, the magnetization is parallel to scattering plane and sample surface. Usually the so-called Kerr rotation of the linear polarization of the incoming light is measured in L-MOKE and P-MOKE \[19, 20, 21\]. Conversely the transverse Kerr effect (T-MOKE) measures a change in intensity. In T-MOKE the magnetization direction is perpendicular to the scattering plane along the sample surface \[22, 23, 24\].

- The Voigt effect (discovered 1898), which is sometimes denoted as linear magnetic birefringence, describes the rotation of the polarization plane of linearly polarized light when passing through a sample that has magnetic moments perpendicular to the propagation direction of the light \[25\]. In contrast to the Faraday and the Kerr effects the Voigt effect is proportional to the square of the atomic magnetic moment. Therefore antiferromagnetic materials can be studied.

- Faraday, Kerr and Voigt effects were first observed at visible wavelengths and were studied using conventional optics. The observation of MO effects at x-ray wavelengths was strongly dependent on the availability of high-brilliance synchrotron light sources with tuneable photon energy and polarization. Moreover, it is technically challenging to perform a polarization analysis of the transmitted or reflected light \[1\].

\[1\] The references cited here and in the following refer to the respective MO effects measured at x-ray wavelengths.
1. Magneto-optical effects

light at (soft) x-ray energies. Therefore x-ray absorption measurements have earlier become available as a standard method: Erskine and Stern [26] predicted a difference in the absorption cross section of circularly polarized light at the $M_{2,3}$ absorption edges\(^2\) of ferromagnetic Ni for the magnetization direction parallel or antiparallel to the light propagation direction. This effect is known as \textit{x-ray magnetic circular dichroism} (XMCD) today. It took more than ten years until XMCD was first observed in 1987 by Schütz et al. [1] at the iron $K$ edge.

- In the meantime, 1985, Thole et al. [28] proposed a different kind of dichroism effect depending on the orientation of the magnetization direction relative to the electric field vector of \textit{linearly} polarized light. The \textit{x-ray magnetic linear dichroism} (XMLD) was observed one year later at the $M_{4,5}$ absorption edge of rare earth materials by van der Laan et al. [29]. Similar to the Voigt effect, the XMLD is quadratic in the magnetization. Therefore it is a suitable method to study antiferromagnets [30].

- The first \textit{non-resonant x-ray magnetic scattering} experiments were performed already in 1972 by de Bergevin and Brunel [31] using a standard x-ray tube. They observed weak magnetic $(1/2, 1/2, 1/2)$ and $(3/2, 3/2, 3/2)$ reflections in antiferromagnetic NiO due to the doubling of the magnetic over the chemical periodicity. However, it was shown by Blume [32] that for photon energies below 10 keV the ratio of magnetic and charge scattering cross sections is of the order of $10^{-6}$ or smaller.

- Gibbs et al. [34] found a strong \textit{resonant} enhancement of the $(0,0,2+\tau)$ magnetic satellite reflex in Ho at the $L_3$ absorption edge, where $\tau$ is the wave vector of the magnetic spiral present in Ho at temperatures below $T_N = 133$ K. Their experimental results were theoretically explained by Hannon et al. [5].

Although magnetic peaks are not accessible due to the long wavelength at soft x-ray energies, \textit{x-ray resonant magnetic scattering} (XRMS) was also measured at the $L_{2,3}$ edges of 3$d$ transition metals in several different geometries, such as specular reflection [22, 33, 34], diffuse scattering [35, 36, 37], small angle scattering (SAS) [38] and diffraction [39, 40, 41].

- Finally, any MO effect can be utilized as a contrast mechanism for magnetic imaging. Common laser-based techniques are Kerr microscopy [42] and Lorentz microscopy [43]. X-ray microscopy techniques imaging the photoelectrons (photoemission electron microscopy, PEEM) [44] or the transmitted x-rays using Fresnel zone plates [45] can yield much finer resolution due to the shorter x-ray wavelength. For reviews on x-ray magnetic microscopy techniques see e.g. Refs. [46, 47].

The different MO effects and geometries are summarized in Fig. 1.1.

Except non-resonant magnetic scattering all MO phenomena listed above are closely connected to each other. The effects originate from electric dipole transitions\(^3\) driven by

\(^2\)For a definition of the nomenclature of the different absorption edges see e.g. Ref. [27].

\(^3\)More generally resonant scattering is due to electric \textit{multipole} transitions [5]. The magnetic satellite peaks observed in Ho [34] are caused by excitations of the $2p_{3/2}$ core electrons to the $5d$ (dipole) and $4f$ (quadrupole) levels, respectively.
the electric field of the electromagnetic radiation. In contrast, non-resonant scattering arises from the interaction of the magnetic radiation field with both the spin and orbital moments.

There is an intimate relation between scattering and absorption phenomena following from the optical theorem and the Kramers-Kronig relations: The absorption cross section is proportional to the imaginary part of the scattering amplitude. Furthermore, imaginary and real part of the scattering amplitude depend on each other due to the Kramers-Kronig relations. If one component is known for a sufficiently large energy range, e.g. the imaginary part from absorption measurements, the other component can be calculated. Therefore magnetic scattering may be thought of as being caused by XMCD or vice versa [48].

The microscopic origin of all MO effects is based on electronic transitions from spin-orbit split initial states to exchange-split final states \[49\]. At photon energies of visible light intraband transitions are probed. The spin-orbit interaction within the \(d\) shell of \(3d\) transition metals is of the order of 0.1 eV and accordingly the MO effects are small. The situation changes significantly for \(2p \rightarrow 3d\) interband transitions, since the spin-orbit splitting of \(L_3\) and \(L_2\) edges ranges between 11.2 eV and 17.3 eV for the elements Mn to Ni, respectively \[50\]. Therefore much larger MO effects are observed. The required photon energies lie in the soft x-ray range between 512 eV and 870 eV for \(3d\) transition metals V to Ni, respectively.

The use of x-rays for studies of magnetic properties is more than a simple extension of laser-based investigations because it offers two more unique capabilities: By tuning the photon energy to a specific absorption edge, the magnetism of different elements can be investigated separately, which is a great advantage in the field of magnetic heterostructures. Furthermore, it is possible to study the magnetic contributions of different kinds of valence electrons by probing different absorption edges of the same element according to the dipole selection rules. For example the magnetism of \(5d\) and \(4f\) levels in rare earth metals can be separately investigated at the \(M_{4,5} (3d \rightarrow 4f) \[51\] and \(L_{2,3}\) edges.
1. Magneto-optical effects

\[ (2p \rightarrow 5d) \text{[52]}, \text{respectively.} \]

1.1.2. Outline

The purpose of this chapter is to elucidate the basic theory of magneto-optical effects. At least two formalisms are available: In Sec. 1.2 a phenomenological approach is discussed utilizing classical electrodynamics and the dielectric tensor in magnetic materials. The solution of Fresnel’s equation yields the normal modes of the electromagnetic waves in presence of a sample magnetization and the phenomenological magneto-optical constants for different geometries, i.e. for the relative orientation of the photon propagation direction and the magnetization. The origin of the energy-dependent elements of the dielectric tensor cannot be explained within the frame of classical electrodynamics. Therefore the quantum mechanical treatment and its application to absorption and scattering are presented in Sec. 1.3. Both phenomena are due to the same term in the interaction Hamiltonian in first- and second-order perturbation theory, respectively. In contrast, non-resonant magnetic scattering is caused by other contributions of the interaction Hamiltonian directly involving the electron spin, as will be briefly discussed at the end of Sec. 1.3. An analysis of the polarization dependence of the scattering factor in Sec. 1.4 exhibits again the same MO effects as shown before for the classical theory and, indeed, both approaches are equivalent within dipole approximation. The chapter is concluded by a discussion of the optical theorem and the Kramers-Kronig relations in Sec. 1.5.

In the subsequent chapters the three methods, which are the topic of the present work, are described in more detail: X-ray magnetic circular dichroism in Chap. 2 magnetic reflectivity in Chap. 3 and magnetic diffuse scattering in Chap. 4.

1.2. Classical theory

In this section the properties of electromagnetic waves in a magnetic medium are discussed on the basis of Maxwell’s classical theory. In an isotropic, amorphous medium the dielectric tensor \( \epsilon(\omega) \) is diagonal with equal diagonal elements, i.e. it reduces to a scalar. The presence of an external magnetic field or a magnetic moment breaks the symmetry and the degeneracy is lifted. The material properties and therewith the dielectric tensor must be invariant under the symmetry operations of the magnetization [53]. In presence of a non-vanishing magnetization vector \( \vec{M} \) with orientation parallel to the \( z \) axis the axial symmetry leads to the following form of the dielectric tensor:

\[
\epsilon(\omega) = \begin{pmatrix}
\epsilon_{xx} & \epsilon_{xy} & 0 \\
-\epsilon_{xy} & \epsilon_{xx} & 0 \\
0 & 0 & \epsilon_{zz}
\end{pmatrix}.
\]

(1.1)

Additionally to the geometrical symmetry considerations, the Onsager relation \( \epsilon_{ij}(\vec{M}) = \epsilon_{ji}(-\vec{M}) \) can be derived from general statistical arguments [54]. The Onsager relation

\[\text{The dielectric tensor should not be mistaken with the polarization vector of the electromagnetic wave, denoted by } \vec{\epsilon}.\]
1.2. Classical theory

causes the diagonal elements to be even in $\vec{M}$ and the off-diagonal elements to be odd in $\vec{M}$. A further reduction of the dielectric tensor due to symmetry arguments is not possible. The same form of the dielectric tensor is valid for cubic systems with the magnetization along (001) directions as well.

It has to be pointed out that this description is only strictly correct for bulk materials while in thin films and interfaces the broken symmetry may modify the magnetic properties [55]. Nonetheless this approach is a starting point and is widely used to describe magneto-optical effects in thin-film systems.

Now the consequences of the dielectric tensor on the normal modes of electromagnetic waves are discussed. For a monochromatic plane wave $\vec{E} = \vec{E}_0 \exp(i\vec{k} \cdot \vec{r} - i\omega t)$ with wave vector $\vec{k}$ and frequency $\omega$, Maxwell’s equations can be written as

\[
\begin{align*}
\vec{k} \cdot \vec{D} &= -4\pi i \rho, \\
\vec{k} \cdot \vec{B} &= 0, \\
\vec{k} \times \vec{H} &= -\frac{4\pi i}{c} \vec{j} - \frac{\omega}{c} \vec{D}, \\
\vec{k} \times \vec{E} &= \frac{\omega}{c} \vec{B},
\end{align*}
\]

where $\vec{E}$ and $\vec{D}$ are the electric field and electric displacement, $\vec{H}$ and $\vec{B}$ are the magnetic field and magnetic induction, $\rho$ is the density of free charges, $\vec{j}$ is the current density and $c$ is the velocity of light in vacuum. Even for ferromagnetic metals with a static sample magnetization $\vec{M}$ or a non-zero external magnetic field $\mu(\omega) = 1$ at optical frequencies, so that $\vec{B}(\omega) = \vec{H}(\omega)$ [56, 57]. The material equations and Ohm’s law are then represented by:

\[
\begin{align*}
\vec{D} &= \epsilon \vec{E} \quad (1.6) \\
\vec{B} &= \vec{H} \quad \text{and} \\
\vec{j} &= \sigma \vec{E}. \quad (1.8)
\end{align*}
\]

Taking the cross product of $\vec{k}$ with Eq. (1.5) and using Eqs. (1.6)–(1.8) yields the Fresnel equation

\[
\epsilon \vec{E} = n^2 \left( \vec{E} - \hat{k}(\vec{E} \cdot \hat{k}) \right),
\]

where $\hat{k}$ is a unit vector pointing in direction of the wave vector $\vec{k} = \frac{\omega}{c} \vec{n} \hat{k}$. Equation (1.9) is a system of three coupled equations. From its solution the normal modes of the electromagnetic wave in the material and the corresponding refractive indices $n$ are obtained. Since the electric field is generally no longer a transverse wave in a magnetic material, another way of obtaining the normal modes is described here, following the procedure in Ref. [56]. In transparent media the conductivity tensor $\sigma$ is zero. It follows directly that $\vec{j}$ and $\rho$ vanish as well, if no external charges are present. In this case Eqs. (1.2)–(1.5) have a simple geometrical interpretation. The vectors $\vec{k}$, $\vec{D}$, $\vec{E}$ and the Poynting vector $\vec{S} = \vec{E} \times \vec{H}$ lie in a plane perpendicular to $\vec{H}$ and are pairwise orthogonal. In contrast to the electrical field $\vec{E}$, the electric displacement $\vec{D}$ is a transverse wave. The relative orientation of all vectors is shown in Fig. 1.2.
1. Magneto-optical effects

Figure 1.2.: Relative orientation of electric and magnetic fields, the wave vector and the Poynting vector in a magnetic medium.

In case of an absorbing material the situation is more complicated. Formally the dielectric tensor $\epsilon$ and the conductivity tensor $\sigma$ can be subsumed to the complex dielectric tensor

$$\epsilon^c = \epsilon + \frac{4\pi i}{\omega} \sigma.$$  \hspace{1cm} (1.10)

Analogously $\vec{D}$ and $\vec{j}$ are combined to a complex electric displacement vector

$$\vec{D}^c = \vec{D} + \frac{4\pi i}{\omega} \vec{j}.$$  \hspace{1cm} (1.11)

With these definitions the Maxwell equations are rewritten. They are formally identical to the equations for transparent media, but now all quantities are complex-valued. Especially the condition

$$\vec{k} \cdot \vec{D}^c = 0$$  \hspace{1cm} (1.12)

is still valid, i.e. the $\vec{D}$-vector is again a transverse wave. The superscript $c$ is omitted again in the following. Inserting $\vec{E} = \epsilon^{-1} \vec{D}$ into Eq. (1.9) leads to an expression for $\vec{D}$ analogue to the Fresnel equation. This treatment has the advantage that all quantities can be transformed to a coordinate system, where one axis ($\vec{e}_z$) is parallel to the wave vector $\vec{k}$, and only two coupled equations are remaining as follows:

$$D_a = n^2(\epsilon^{-1})_{aa}D_a + n^2(\epsilon^{-1})_{ab}D_b,$$

$$D_b = n^2(\epsilon^{-1})_{ba}D_a + n^2(\epsilon^{-1})_{bb}D_b.$$  \hspace{1cm} (1.13, 1.14)

A detailed derivation of the refractive indices and normal modes is given in Ref. [58]. Here only the results are stated. If the electromagnetic wave is propagating along the magnetization direction, $\vec{k} \parallel M \parallel \vec{e}_z$ ($a = x$, $b = y$), the solution for the refractive index is

$$n_{\pm}^2 = \epsilon_{xx} \pm i\epsilon_{xy}.$$  \hspace{1cm} (1.15)

Negative values of the refractive index correspond to waves propagating in the negative direction. The normal modes are circularly polarized waves, $D_x = \pm iD_y$. In this special case the electrical field is a circularly polarized transverse wave, too. This geometry
corresponds to the Faraday effect, XMCD, polar MOKE and the longitudinal MOKE at grazing incidence.

If the wave is propagating in a direction perpendicular to the magnetization, $\hat{k} \perp \vec{M} \parallel \vec{e}_z$, the normal modes are linearly polarized. Only the refractive index $n_\perp$ for the field component perpendicular to $\vec{M}$ is sensitive to the magnetization:

$$n_\perp^2 = \epsilon_{xx} + \frac{\epsilon_{xy}^2}{\epsilon_{xx}}, \quad n_{||}^2 = \epsilon_{zz}. \quad (1.16)$$

This situation corresponds to XMLD or the Voigt effect. It is second order in the magnetic quantity $\epsilon_{xy}$.

The general solution of Eqs. (1.13) and (1.14) is rather complicated:

$$n_{1,2}^2 = \frac{2\epsilon^2 + \epsilon_{xy}^2 \sin^2 \gamma \pm \sqrt{\epsilon_{xy}^4 \sin^4 \gamma - 4\epsilon^2 \epsilon_{xy}^2 \cos^2 \gamma}}{2\epsilon}, \quad (1.17)$$

where $\gamma$ is the angle between the propagation direction and $\vec{M}$. For simplicity $\epsilon_{xx} = \epsilon_{zz}$ is assumed here. The $D$-wave is now elliptically polarized. In case of $\gamma = 0$ the general result of Eq. (1.17) reduces to Eq. (1.15), for $\gamma = \pi/2$ to Eq. (1.16).

The magnetic contribution to the refractive index is of the order of $10^{-3}$ at the $L_{2,3}$ edges of the $3d$ transition metals. Therefore an expansion to first order in the magnetic parameter still leads to results, which are in very good agreement with experimental data on ferromagnetic samples. The dielectric tensor is commonly rewritten in the form

$$\epsilon(\omega) = N^2 \begin{pmatrix} 1 & iQ & 0 \\ -iQ & 1 & 0 \\ 0 & 0 & 1 \end{pmatrix}, \quad (1.18)$$

$$\epsilon_{xx} = \epsilon_{zz} = N^2, \quad \epsilon_{xy} = iN^2 Q, \quad (1.19)$$

where again the difference between the diagonal elements $\epsilon_{xx}$ and $\epsilon_{zz}$ has been neglected. $Q$ is the Voigt parameter. Different sign conventions for $Q$ are used in literature [59, 60, 61]. Here the scheme of Ref. [59] is followed. Expanding all quantities in Eqs. (1.13) and (1.14) to first order in $Q$,

$$n_{1,2}^2 = N^2(1 \pm Q \cos \gamma) \quad (1.20)$$

is obtained. In this approximation the $D$-waves are again circularly polarized independently of the propagation direction:

$$D_a = \pm iD_b. \quad (1.21)$$

Equations (1.20) and (1.21) are the starting point for the simulation of the soft x-ray resonant magnetic specular reflectivity, which is described in Chap. 3.

So far the dielectric tensor has been only used as a phenomenological parameter. It has to be pointed out that $\epsilon$ is strongly frequency-dependent. Within the frame of Maxwell’s theory this cannot be explained. For the description of $\epsilon$ a microscopic theory is necessary, which is outlined in the next section.
1. Magneto-optical effects

1.3. Quantum mechanics

The starting point for the quantum mechanical description of the interaction of light with matter is the non-relativistic approximation of the Dirac equation \[62\]. The Hamiltonian for an atomic electron in a quantized electromagnetic field is given by \[32\]

\[
\mathcal{H} = \frac{1}{2m} \left( \vec{p} - \frac{e}{c} \vec{A} \right)^2 + V_0(\vec{r}) - \frac{e\hbar}{2mc} \vec{s} \cdot \nabla \times \vec{A} - \frac{\hbar}{2(mc)^2} \vec{s} \cdot \vec{E} \times \left( \vec{p} - \frac{e}{c} \vec{A} \right) + \sum_{k\sigma} \hbar \omega_k c \left[ c^\dagger(k,\sigma) c(k,\sigma) e^{i(k \cdot \vec{r} - \omega t)} + e^{*}(k,\sigma) c^\dagger(k,\sigma) e^{-i(k \cdot \vec{r} - \omega t)} \right].
\] (1.22)

All quantities like \(\vec{p}\) and \(\vec{r}\) have to be considered as operators in the following. The Hamiltonian comprises of kinetic and potential energy of the electrons, Zeeman energy, spin-orbit coupling and the free radiation field. According to Maxwell’s theory, by choosing the Coulomb gauge \(\nabla \cdot \vec{A} = 0\), the electromagnetic radiation can be described by the vector potential \(\vec{A}\) alone. It is linear in photon creation and annihilation operators \(c^\dagger\) and \(c\),

\[
\vec{A} = \sum_{k,\sigma} \left( \frac{2\pi \hbar c^2}{V \omega_k} \right)^{1/2} \left[ \epsilon(k,\sigma) c(k,\sigma) e^{i(k \cdot \vec{r} - \omega t)} + \epsilon^{*}(k,\sigma) c^\dagger(k,\sigma) e^{-i(k \cdot \vec{r} - \omega t)} \right],
\] (1.23)

where the sum is over all wave vectors \(\vec{k}\). The two different polarization states are labelled by the index \(\sigma = 1, 2\). The quantization volume \(V\) drops out of any physical expression.

In order to apply time-dependent perturbation theory, the Hamiltonian in Eq. (1.22) is split into contributions of the undisturbed electron system, free radiation field and interaction Hamiltonian, \(\mathcal{H} = \mathcal{H}_0 + \mathcal{H}_{\text{rad}} + \mathcal{H}_{\text{int}}\), where

\[
\mathcal{H}_{\text{int}} = -\frac{e}{mc} \vec{A} \cdot \vec{p} + \frac{e^2}{2mc^2} \vec{A}^2 - \frac{e\hbar}{mc} \vec{s} \cdot \left( \nabla \times \vec{A} \right) - \frac{e\hbar}{2(mc)^2} \frac{e^2}{c^2} \vec{s} \cdot \left( \dot{\vec{A}} \times \vec{A} \right)
\]

\[
\equiv \mathcal{H}_1 + \mathcal{H}_2 + \mathcal{H}_3 + \mathcal{H}_4.
\] (1.24)

The interaction Hamiltonian is subdivided into four terms with different \(\vec{A}\)-dependence. The physical processes caused by \(\mathcal{H}_1 - \mathcal{H}_4\) are outlined in the following paragraphs: \(\mathcal{H}_1\) describes absorption processes in first order and resonant scattering processes in second order perturbation theory, non-resonant Thomson scattering is caused by \(\mathcal{H}_2\). The spin-dependent terms \(\mathcal{H}_3\) and \(\mathcal{H}_4\) are responsible for non-resonant magnetic scattering.

1.3.1. Absorption

The absorption cross section is defined as the number of excited electrons per unit time divided by the incident photon flux \(I_0 = c/V\),

\[
\sigma_a = \frac{w_{i-f}}{I_0}.
\] (1.25)

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The leading contribution of the interaction Hamiltonian to absorption stems from $H_1$. The transition amplitude per unit time to first order perturbation theory is given by Fermi’s golden rule,

$$w_{i\rightarrow f} = \frac{2\pi}{\hbar} \int dE_f |\langle f|H_1|i\rangle|^2 \rho(E_f)\delta(E_f - E_i - \hbar\omega), \quad (1.26)$$

where $\rho(E_f)$ is the density of final states. In case of absorption the initial and final states are given by

$$|i\rangle = |i\rangle_{el}|\vec{k}_i, \sigma\rangle_{ph}, \quad |f\rangle = |f\rangle_{el}|0\rangle_{ph}. \quad (1.27)$$

Now the matrix element in Eq. (1.26) has to be evaluated by inserting Eqs. (1.24) and (1.23). Since the final state does not contain a photon, only the term proportional to the annihilation operator $c(\vec{k} = \vec{k}_i)$ gives a non-zero contribution to the matrix element,

$$\langle f|H_1|i\rangle = -\frac{e}{mc} \left(\frac{2\pi\hbar c^2}{V\omega}\right)^{\frac{1}{2}} \left\langle f \left| \vec{e} \cdot \vec{r} \right| i \right\rangle_{el} e^{-i\omega t}. \quad (1.28)$$

The exponential function $e^{i\vec{k} \cdot \vec{r}}$ is expanded as

$$e^{i\vec{k} \cdot \vec{r}} = 1 + i\vec{k} \cdot \vec{r} - \frac{1}{2}(\vec{k} \cdot \vec{r})^2 + \ldots. \quad (1.29)$$

The terms on the right hand side describe electric dipole ($E1$), magnetic dipole ($M1$), electric quadrupole ($E2$) etc. transitions, respectively. Since the present work solely deals with $2p \rightarrow 3d$ excitation processes of 3d transition metals, the photon energy is in the soft x-ray range with a wavelength $\lambda = 1.5 - 2$ nm so that it is sufficient to consider dipole transitions only. Replacing the exponential function by one reflects the fact that the excited core electron is localized around the nucleus and therefore does not see the spatial variation of the electromagnetic wave.

When the momentum operator is replaced by the commutation relation $\vec{p} = -im/\hbar[\vec{r}, H_0]$, the absorption cross section finally results in

$$\sigma_a = \frac{4\pi^2 e^2}{\hbar c} \hbar\omega |\langle f|\vec{e} \cdot \vec{r}|i\rangle|^2 \rho(E_f = E_i + \hbar\omega). \quad (1.30)$$

As will be discussed in Chap. 2 in more detail, the dipole selection rules for linearly and circularly polarized light follow from the matrix elements of $\vec{r}$:

$$\Delta j = 0, \pm 1, \quad \Delta l = \pm 1, \quad \Delta s = 0, \quad \Delta m = \begin{cases} 0 \text{ linear}, \\ +1 \text{ right circular}, \\ -1 \text{ left circular}. \end{cases} \quad (1.31)$$

The selection rules simply reflect the principle of angular momentum conservation. Although the photon helicity does not couple directly to the electron spin but to the orbital angular momentum, a spin-dependent excitation can occur due to spin-orbit coupling of the core state in the case of circularly polarized light. Since the density of final states is different for both spin directions in a magnetic material, a difference in the absorption of left and right circularly polarized light can be observed, which is the origin of XMCD. This mechanism will be explained in more detail within a simple two-step-model. But the further discussion of absorption and XMCD is postponed until Chap. 2. First the discussion of the interaction Hamiltonian is continued.
1. Magneto-optical effects

1.3.2. Resonant scattering

The microscopic interaction of x-rays with matter is commonly described in terms of the scattering amplitude

\[ f(\vec{q}, \omega) = f^0(\vec{q}) + f'(\omega) + if''(\omega). \]  

(1.32)

The dispersion corrections \( f' \) and \( f'' \) have to be included, if the photon energy is close to a resonance. The scattering amplitude is related to the differential cross section by the fundamental definition for single scatterers

\[ \frac{d\sigma}{d\Omega} = |f(\vec{q}, \omega)|^2. \]  

(1.33)

The leading contributions of the interaction Hamiltonian involving two-photon processes are \( \mathcal{H}_2 \) in first order and \( \mathcal{H}_1 \) in second order perturbation theory, resulting in

\[ f(\vec{q}, \omega) = \frac{V\omega}{2\pi\hbar c^2} \left( \langle f|\mathcal{H}_2|i \rangle + \sum_n \frac{\langle f|\mathcal{H}_1|n \rangle \langle n|\mathcal{H}_1|i \rangle}{E_i - E_n + \hbar\omega + i\Gamma/2} \right) \]  

(1.34)

for the scattering amplitude. The initial and final states are now given by

\[ |i\rangle = |i\rangle_{el}|\vec{k}_i, \sigma_i\rangle_{ph}, \quad |f\rangle = |i\rangle_{el}|\vec{k}_f, \sigma_f\rangle_{ph}. \]  

(1.35)

The first term in Eq. (1.34) gives rise to the usual Thomson scattering, which is the non-resonant contribution to scattering from all electrons in the atom. The non-resonant charge scattering \( f^0(\vec{q}) \) is generally given by the Fourier transform of the atomic charge distribution. However, at the 2\( p \) edges of 3\( d \) metals the scattering vector is small compared to the atomic radii, and the scattering amplitude can be approximated by \( f^0(\vec{q} = 0) = -r_e Z \), where \( r_e \) is the classical electron radius and \( Z \) the atomic charge number.

The second term is the origin of resonant scattering. It might be the dominant contribution to scattering, if the photon energy is tuned close to an absorption edge, \( \hbar\omega = E_n - E_i \), where \( n \) is an intermediate atomic state. The process can be viewed as an absorption of the initial photon exciting an electron to an intermediate state. The decay of the intermediate state again leads to the emission of a photon. The matrix elements are the same as in the case of absorption, Eq. (1.28). The scattering amplitude for an electric 2\( L \)-pole (\( EL \)) resonance has been derived by Hannon et al. \[5\] for magnetic ions, yielding

\[ f_{EL}^{\text{res}}(E) = 2\lambda \sum_{M=-L}^L \left[ \vec{e}_f^* \cdot \vec{Y}_{LM}(\hat{k}_f)\vec{Y}_{LM}^*(\hat{k}_i) \cdot \vec{e}_i \right] F_M^L(\hbar\omega), \]  

(1.36)

where \( \vec{Y}_{LM} \) are vector spherical harmonics and

\[ F_M^L(\hbar\omega) = \sum_{i,n} p_ip_i(n)\frac{\Gamma_x(i, M, n, EL)}{E_i - E_n + \hbar\omega + i\Gamma/2}, \]  

(1.37)

are dimensionless transition matrix elements. The probability that the initial state \( |i\rangle \) is occupied, and the transition probability to the state \( |n\rangle \) are given by \( p_i \) and \( p_i(n) \),

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respectively. \( \Gamma \) denotes the total line width, including all radiative and non-radiative decays. The partial width for the \( EL \) radiative decay \( |n\rangle \rightarrow |i\rangle \) is

\[
\Gamma_x(i, M, n, EL) = \frac{8\pi^2 e^2}{\lambda} \frac{L + 1}{L} |\langle n|j_L(qr)Y^*_M(\hat{r})|i\rangle|^2 ,
\]

where \( j_L \) is the spherical Bessel function of order \( L \). If again only dipole transitions are considered, the complete scattering amplitude for non-resonant and resonant scattering is

\[
f = (\hat{\epsilon}_f^* \cdot \hat{\epsilon}_i)(-r_e Z + F^{(0)}) + i(\hat{\epsilon}_f^* \times \hat{\epsilon}_i) \cdot \vec{m} F^{(1)} + (\hat{\epsilon}_f^* \cdot \vec{m})(\hat{\epsilon}_i \cdot \vec{m}) F^{(2)},
\]

with

\[
F^{(0)} = \frac{3\lambda}{8\pi} \left[ F^1_{-1} + F^1_1 \right],
\]

\[
F^{(1)} = \frac{3\lambda}{8\pi} \left[ F^1_{-1} - F^1_1 \right],
\]

\[
F^{(2)} = \frac{3\lambda}{8\pi} \left[ 2F^1_0 - F^1_{-1} - F^1_1 \right].
\]

The unit vector \( \vec{m} \) is pointing along the magnetization direction, which defines the quantization axis of the system. Terms proportional to \( \hat{\epsilon}_f^* \cdot \hat{\epsilon}_i \) describe non-resonant and resonant charge scattering. The term involving \( (\hat{\epsilon}_f^* \times \hat{\epsilon}_i) \cdot \vec{m} \) is first order in the magnetization and yields circular dichroism and Kerr effects. The term proportional to \( (\hat{\epsilon}_f^* \cdot \vec{m})(\hat{\epsilon}_i \cdot \vec{m}) \) is second order in the magnetization and causes linear dichroism or the Voigt effect. As mentioned before, Eq. (1.39) was originally derived for a magnetic ion. The same basic terms are also found in theoretical descriptions of atomic scattering factors of itinerant metallic systems [63].

The energy-dependent material properties are contained in the functions \( F^{(0,1,2)} \). These can be derived from band structure calculations in principle. As it is shown below the functions are directly connected to the diagonal and off-diagonal elements of the dielectric tensor. A theoretical description of the functions \( F^{(0,1,2)} \) is beyond the scope of this thesis. The geometrical dependence of the resonant magnetic scattering amplitude is contained in the prefactors. These are further discussed in Sec. 1.4.

It should be emphasized again that resonant magnetic scattering results from electric dipole transitions and not from the interaction of the magnetic field of the electromagnetic wave with the electron spin and orbital momentum. The magnetic sensitivity is achieved, because only transitions to unoccupied states in the conduction band are allowed. The direct interaction of the magnetic field \( \vec{H} \) of the electromagnetic radiation with the electron spin results in non-resonant scattering and is briefly discussed in the following section.

1.3.3. Non-resonant magnetic scattering

In order to conclude the discussion of the Hamiltonian the last two terms in Eq. (1.24) are considered. These give rise to non-resonant magnetic scattering, which is mentioned for the sake of completeness although it is not a topic of the present work. There are
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Figure 1.3.: Classical picture of the dominant mechanisms of photon scattering from a free electron (taken from Ref. [64]). The top diagram shows charge scattering, while the others illustrate interactions between the photon field and electron spin. \( \vec{\mu} = -2\mu_B\vec{S} \) is the magnetic dipole moment of the electron.

Two processes leading to a direct interaction of the photon field and electron spin, which are schematically depicted in Fig. 1.3. The Zeeman interaction of the magnetic field \( \vec{H} = \vec{\nabla} \times \vec{A} \) with the electron spin results in a torque on the spin magnetic dipole moment. Additionally the spin-orbit term describes the interaction of the electron spin with the magnetic field generated by the orbital movement of the electron in the electric photon field. The corresponding scattering amplitude is given by [48]

\[
 f_{mag} = i r_0 \frac{\hbar \omega}{mc^2} \left[ \frac{1}{2} \vec{L}(\vec{q}) \cdot \vec{A} + \vec{S}(\vec{q}) \cdot \vec{B} \right], \quad (1.43)
\]

where \( \vec{L}(\vec{q}) \) and \( \vec{S}(\vec{q}) \) are the Fourier transforms of the atomic orbital and spin magnetization densities. The complicated polarization dependence is given by the vectors \( \vec{A} \) and \( \vec{B} \):

\[
\vec{A} = 2(1 - \hat{k}_i \cdot \hat{k}_f)(\vec{e}_f \times \vec{e}_i) - (\hat{k}_i \times \vec{e}_i)(\hat{k}_i \cdot \vec{e}_f) + (\hat{k}_f \times \vec{e}_i)(\hat{k}_f \cdot \vec{e}_f) \quad (1.44)
\]

\[
\vec{B} = \vec{e}_f \times \vec{e}_i - (\hat{k}_f \times \vec{e}_f) \times (\hat{k}_i \times \vec{e}_i) + (\hat{k}_f \cdot \vec{e}_i)(\hat{k}_f \cdot \vec{e}_f) - (\hat{k}_i \cdot \vec{e}_f)(\hat{k}_i \cdot \vec{e}_i). \quad (1.45)
\]

An advantage of non-resonant magnetic scattering is that the spin and orbit contributions to the magnetic density can be distinguished by means of the different polarization
dependence. Because the spin scattering amplitude is reduced by a factor \( \tau = \hbar \omega / mc^2 \) compared with charge scattering, hard x-rays are necessary. Additionally the spin scattering amplitude scales with the net spin polarization, which is small even in magnetic systems. Therefore pure magnetic non-resonant scattering intensities are typically of order \( 10^{-6} \) compared to charge scattering. Although it is extremely difficult to observe non-resonant magnetic scattering, the first experiments on NiO were performed using a standard x-ray tube [31]. Nowadays the problem of small intensities is overcome with high-brilliance synchrotron sources. However, the contribution of non-resonant magnetic scattering to the total scattered intensity is negligible for all experiments shown in the present work.

1.4. Polarization dependence in resonant scattering

In this section the polarization dependence of the resonant scattering amplitude and the different magnetic contributions occurring in different geometries are discussed. Explicit expressions of the polarization dependence have been given e.g. by Hill and McMorrow [65].

1.4.1. Linear polarization

It is useful to write the polarization factors in Eq. (1.39) as \( 2 \times 2 \) matrices [64]. The polarization states are chosen either perpendicular (\( \vec{\epsilon}_\sigma \)) or parallel (\( \vec{\epsilon}_\pi \)) to the scattering plane, which is spanned by the wave vectors of the incident and scattered photons (see Fig. 1.4). Note that \( \vec{\epsilon}_\sigma, \vec{\epsilon}_\pi \) and \( \hat{k} \) form a right-handed coordinate system. The first term in Eq. (1.39), describing charge scattering, does not change the polarization state of the photon and the matrix is diagonal,

\[
\vec{\epsilon}_f^* \cdot \vec{\epsilon}_i = \begin{pmatrix} 1 & 0 \\ 0 & \hat{k}_f \cdot \hat{k}_i \end{pmatrix} \text{spec.} = \begin{pmatrix} 1 & 0 \\ 0 & \cos(2\Theta) \end{pmatrix}.
\] (1.46)

The second equality sign holds only for the specular direction, i.e. if the incidence and exit angle are the same [64]. While \( \sigma \rightarrow \sigma \) scattering is independent of the angle of incidence \( \Theta \), \( \pi \rightarrow \pi \) scattering is zero for \( 2\Theta = 90^\circ \). Since the second term of the scattering amplitude in Eq. (1.39) is proportional to the cross product of the polarization states, resonant magnetic scattering allows for a change of the polarization state, i.e. \( \sigma \rightarrow \pi \) and \( \pi \rightarrow \sigma \) scattering,

\[
\left( \vec{\epsilon}_f^* \times \vec{\epsilon}_i \right) \cdot \vec{m} = \begin{pmatrix} 0 & \hat{k}_i \cdot \vec{m} \\ -\hat{k}_f \cdot \vec{m} & \left( \hat{k}_f \times \hat{k}_i \right) \cdot \vec{m} \end{pmatrix} \text{spec.} = \begin{pmatrix} 0 & m_y \cos \Theta + m_z \sin \Theta \\ -m_y \cos \Theta + m_z \sin \Theta & -m_x \sin(2\Theta) \end{pmatrix}.
\] (1.47)

Here a coordinate system has been introduced with the \( x \) and \( y \) axes in the sample plane perpendicular and parallel to the scattering plane, respectively, and \( z \) perpendicular to

\[5\] Throughout the present thesis the angle of incidence \( \Theta \) is defined with respect to the sample surface.
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\[ \varepsilon_{\pi,f} \]

\[ f_k \]

\[ i_{\sigma,i} \]

\[ \varepsilon_{\sigma,i} \]

\[ \varepsilon_{\pi,i} \]

\[ \varepsilon_{\sigma,f} \]

\[ \varepsilon_{\pi,f} \]

\[ k_f \]

\[ k_i \]

scattering plane

Figure 1.4.: Definition of polarization vectors.

the sample surface. According to Eq. (1.33) the differential cross section is proportional to the absolute square of the scattering amplitude. Therefore contributions linear in the magnetization stem from the charge-magnetic interference term. Since charge scattering does not change the polarization state and magnetic scattering is forbidden, only \( \pi \rightarrow \pi \) scattering contains a contribution linear in \( \vec{M} \) leading to the T-MOKE effect. The T-MOKE signal is located at the same position in reciprocal space as the charge scattering. In contrast, the intensity of \( \sigma \rightarrow \pi \) and \( \pi \rightarrow \sigma \) scattering is quadratic in \( \vec{M} \) and gives rise to satellite peaks in magnetically periodic structures. Examples for the T-MOKE and XRMS using linearly polarized light are given in Chaps. 3 and 7.

A derivation of explicit expressions for the polarization dependence of the second-order magnetic term in Eq. (1.39) is more tedious. The final result in the specular case is [65]

\[
(\vec{\varepsilon}_i^* \cdot \vec{m})(\vec{\varepsilon}_i \cdot \vec{m}) = \begin{pmatrix}
    m_x^2 \\
    -m_x(m_y \sin \Theta + m_z \cos \Theta) \\
    m_x(m_y \sin \Theta - m_z \cos \Theta) \\
    \cos^2 \Theta (m_y^2 \tan^3 \Theta + m_z^2)
\end{pmatrix}. \tag{1.48}
\]

The contribution of the term proportional to \( F^{(2)} \) to XRMS is generally assumed to be small at the \( L_{2,3} \) edges of ferromagnetic 3d transition metals [55]. It is neglected in the data analysis throughout the present thesis.

1.4.2. Circular polarization

Several conventions are used in literature for the definition of circular polarization vectors differing in an arbitrary over-all phase factor. Here the definition of Lovesey is used [48]. Right (left) circularly polarized light is defined as rotating counterclockwise (clockwise) when facing into the oncoming wave in a fixed point in space. This corresponds to positive (negative) helicity with an angular momentum of \( +\hbar \) (\( -\hbar \)). The corresponding polarization vectors are written as a linear combination of \( \vec{\varepsilon}_\sigma \) and \( \vec{\varepsilon}_\pi \),

\[
\vec{\varepsilon}_\pm = \pm \frac{1}{\sqrt{2}} (\vec{\varepsilon}_\sigma \pm i\vec{\varepsilon}_\pi), \tag{1.49}
\]

where the positive sign indicates right circular polarization. A calculation of the first and second polarization factor in Eq. (1.39) in the basis of circular polarization states
yields

\[ \vec{e}_{f,\pm}^* \cdot \vec{e}_{i,\pm} = \frac{1}{2} \left( 1 + \hat{k}_f \cdot \hat{k}_i \right), \tag{1.50} \]

\[ \left( \vec{e}_{f,\pm}^* \times \vec{e}_{i,\pm} \right) \cdot \vec{m} = \pm \frac{i}{2} \left( \hat{k}_f + \hat{k}_i \right) \cdot \vec{m} + \frac{1}{2} \left( \hat{k}_f \times \hat{k}_i \right) \cdot \vec{m}, \tag{1.51} \]

and

\[ \vec{e}_{f,\pm}^* \cdot \vec{e}_{i,\mp} = 0, \quad \left( \vec{e}_{f,\pm}^* \times \vec{e}_{i,\mp} \right) \cdot \vec{m} = 0. \tag{1.52} \]

Neither charge nor magnetic scattering change the polarization state of circular polarized light. In ferromagnetic samples the dominant contribution to scattering is again caused by the charge-magnetic interference term. Experiments using circular polarization are usually performed in the L-MOKE geometry, i.e. the magnetization lies along the sample surface within the scattering plane. In this geometry it is equivalent to switch either to helicity or the external magnetic field. The second term in Eq. (1.51) indicates that T-MOKE can be measured with circularly polarized light as well, because any circular polarization state can be thought of to be a superposition of two linear polarization states and therefore contains π polarized light. However, numerical examples in Chap. 3 show that the T-MOKE using circular polarization is small and can be neglected for incidence angles far from the Brewster angle, which is Θ \approx 45^\circ at x-ray wavelengths.

1.4.3. Comparison of both formalisms

The general equivalence of the two formalisms using the dielectric tensor and the atomic scattering factor, respectively, is seen by noting the one-to-one correspondence of terms describing the same polarization dependence for the same normal modes \[55\]. The refractive index is related to the scattering factor through \[66\]

\[ 1 - n(\omega) = \frac{r_e \lambda^2}{2\pi} \sum_{i} \rho_i f_i(\omega, q = 0), \tag{1.53} \]

where \( \rho_i \) is the number density of species \( i \) in the sample and \( f_i(\omega, q = 0) \) is the corresponding scattering amplitude in the forward direction \( \vec{k}_i = \vec{k}_f \). Equation (1.53) is used to compare both descriptions for certain polarization states and geometries. Starting with circularly polarized light propagating along \( \vec{M} \) an insertion of Eqs. (1.15), (1.39), (1.50) and (1.51) in Eq. (1.53) leads to

\[ n_{\pm} = 1 + \rho \frac{r_e \lambda^2}{8\pi^2} \left( 4\pi r_e Z - 3\lambda F^1_{\mp 1} \right) \tag{1.54} \]

for each circular normal mode and

\[ n_+ - n_- = 3\rho \frac{r_e \lambda^2}{16\pi^2} \left( F^1_1 - F^1_{-1} \right) = \rho \frac{r_e \lambda^2}{2\pi} F^{(1)} \tag{1.55} \]

for their difference, respectively. Similarly, the same considerations for linearly polarized light in the Voigt geometry with the electric field vector parallel and perpendicular to the magnetization direction, respectively, yield

\[ n_{||} = 1 - \rho \frac{r_e \lambda^2}{8\pi^2} \left( 4\pi r_e Z + 3\lambda F^1_0 \right), \tag{1.56} \]
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\[ n_{\perp} = 1 - \rho r e \frac{\lambda^2}{8\pi^2} \left( 4\pi r e Z + \frac{3}{2} \lambda[F_1^1 + F_{-1}^1] \right), \]  
(1.57)

and

\[ n_{\perp} - n_{\parallel} = \rho r e \frac{3\lambda^3}{16\pi^2} \left( 2F_0^1 - F_1^1 - F_{-1}^1 \right) = \rho r e \frac{\lambda^2}{2\pi} F^{(2)}. \]  
(1.58)

From the above expressions, Eqs. (1.55) and (1.58), a direct correspondence of first-order and second-order magneto-optical effects of both descriptions is evident. The equivalence is no longer valid for higher order multipole transitions. A classical analogue to the corresponding polarization factors in Eq. (1.36) is missing.

Since the classical theory utilizing the dielectric tensor and the quantum mechanical description are equivalent with respect to dipole transitions, it is possible to choose the appropriate method for each specific application. On the one hand, for the simulation of specular reflection from multilayer systems an algorithm based on classical theory is presented in Chap. 3 which was originally developed for Kerr measurements in the visible spectrum. On the other hand, a theoretical description of diffuse scattering is more convenient in a notation using the atomic scattering amplitude, as will be discussed in Chap. 4.

1.5. Optical theorem and Kramers-Kronig relations

Absorption and scattering can be viewed as two sides of the same coin. The origin of this connection is given by two fundamental theorems, the optical theorem and the Kramers-Kronig relations. The optical theorem states that the absorption cross section is proportional to the imaginary part of the scattering amplitude in the forward direction,

\[ \sigma_a = \frac{4\pi r e}{k} f''(\vec{q} = 0). \]  
(1.59)

The imaginary part of the forward scattering amplitude measures the loss of intensity that the incident beam suffers because of the scattering.

The second step is to relate the real and imaginary part of the scattering amplitude, or alternatively, the refractive index. The refractive index is commonly split into real and imaginary part according to

\[ n_{\pm} = 1 - \delta_{\pm} + i\beta_{\pm} = 1 - (\delta \pm \Delta\delta/2) + i(\beta \pm \Delta\beta/2), \]  
(1.60)

where \( \delta \) and \( \beta \) are the dispersive and absorptive contributions, respectively, and \( \Delta\delta \) and \( \Delta\beta \) are the corresponding magnetic contributions to the refractive index. The modified Kramers-Kronig relations in case of a magnetic medium are

\[ \delta_{\pm}(E) + \delta_{\mp}(E) = -\frac{2}{\pi} \mathcal{P} \int_0^{\infty} E \frac{\beta_{\pm}(E') + \beta_{\mp}(E')}{E'^2 - E^2} dE', \]  
(1.61)

\[ \delta_{\pm}(E) - \delta_{\mp}(E) = -\frac{2E}{\pi} \mathcal{P} \int_0^{\infty} \frac{\beta_{\pm}(E') - \beta_{\mp}(E')}{E'^2 - E^2} dE', \]  
(1.62)

where \( \mathcal{P} \) denotes the principal value. The back transformations are

\[ \beta_{\pm}(E) + \beta_{\mp}(E) = \frac{2E}{\pi} \mathcal{P} \int_0^{\infty} \frac{\delta_{\pm}(E') + \delta_{\mp}(E')}{E'^2 - E^2} dE'. \]  
(1.63)
\[ \beta_+(E) - \beta_-(E) = -\frac{2}{\pi} \mathcal{P} \int_{0}^{\infty} \frac{E' \delta_+(E') - \delta_-(E')}{E'^2 - E'^2} \, dE'. \] (1.64)

Similar relations are valid for several physical quantities like the scattering amplitude, the electric susceptibility and the dielectric tensor. If now magnetic circular dichroism is observed for a certain material, the optical theorem and Kramers-Kronig relations imply that both imaginary and real part of the refractive index have magnetic contributions. Resonant magnetic scattering can therefore be viewed as being caused by magnetization-dependent absorption. The physics behind XRMS and XMCD is essentially the same.

It is possible to determine the complete refractive index from an absorption experiment by applying Eq. (1.59) with \( \beta = 2\pi \rho r f''/k^2 \) and Eq. (1.61). An example of this procedure is shown in Chap. 8.

Both, the optical theorem and the Kramers-Kronig relations, have a profound physical basis: The optical theorem arises from the conservation of probability, which is a consequence of the Hermitian property of the Hamiltonian (see e.g. [68]). The Kramers-Kronig relations are based on the principle of causality [69] stating that the response of a system is at a later instant than the reason that caused the response.
1. Magneto-optical effects
2. X-ray Magnetic Circular Dichroism

2.1. Introduction

X-ray magnetic circular dichroism (XMCD) is the difference in absorption of left and right circularly polarized light exhibited by ferromagnetic materials with a magnetization component along the x-ray propagation direction. In Chap. 1 Eq. (1.30) the absorption cross section for dipole transitions has been derived in first-order perturbation theory. The transition matrix element is further evaluated in the next section leading to a simple two-step model of XMCD. One of the most attractive features of XMCD is the possibility to determine the element-specific spin and orbital magnetic moments via sum rules. Sum rules and their limitations are discussed in Sec. 2.3. Most x-ray absorption experiments are performed by measuring the total photoelectron yield which is discussed in the last section.

2.2. Two-step model of XMCD

The spin-dependent absorption process can be explained in a simple two-step picture schematically depicted in Fig. 2.1: In the first step the excited photoelectron becomes spin-polarized depending on the photon helicity due to the spin-orbit splitting of the core state. In a second step the unoccupied final states serve as a spin detector for

![Figure 2.1.: Two-step picture of the spin-dependent absorption process.](image-url)
the photoelectron. Due to the magnetization of the system the spin-polarized density of states is different for spin-up and spin-down electrons. According to Fermi’s golden rule, Eq. (1.26), the transition probability for photoelectrons is then strongly spin-dependent as well, leading to a difference in absorption of left and right circularly polarized light.

The absorption cross section for a dipole transition has been given in Eq. (1.30). The transition matrix element introduced in Eq. (1.30) is now further evaluated, following the procedure as presented for example in Ref. [70]. The transition matrix element introduced in Eq. (1.30) is now further evaluated, following the procedure as presented for example in Ref. [70].

The absorption process probes the angular momentum quantum numbers $l$ and $m_l$. Since the absorption process is sensitive to the orbital angular momentum quantum number and not to the total angular momentum, the initial and final states $|nlm_j\rangle$ have to be written as a linear combination of $|nlm_{j\pm}\rangle$ using Clebsh-Gordon coefficients. The decomposition of the total angular momentum states in this basis for the $p_{3/2}$ and $p_{1/2}$ levels is given in the first column of Table 2.1.

A coordinate system is chosen with the $z$ axis parallel to the quantization axis, i.e. the magnetization direction. The scalar product of the position and polarization vectors in Eq. (1.30) is written as

$$\bar{\epsilon}_{\pm 1,0} \cdot \vec{r} = R_{\pm 1,0}, \quad (2.1)$$

with

$$R_{+1} = -\frac{1}{\sqrt{2}}(x + iy), \quad R_0 = z, \quad R_{-1} = \frac{1}{\sqrt{2}}(x - iy). \quad (2.2)$$

where $\bar{\epsilon}_{+1}$, $\bar{\epsilon}_{-1}$ and $\bar{\epsilon}_0$ correspond to right and left circularly and linearly polarized light, respectively. For circularly polarized light the propagation direction is chosen to point along the $z$ axis. For linearly polarized light the electric field vector of the electromagnetic wave is parallel to the quantization axis with a propagation direction in the $xy$ plane. Since $R_q$ with $q = \pm 1, 0$ are irreducible spherical tensor operators, the Wigner-Eckart theorem can be applied. In this way geometrical and dynamical aspects of the matrix elements are separated, which appear in the Clebsh-Gordon coefficients and reduced matrix elements, respectively. In the special case of $2p \rightarrow 3d$ dipole transitions and circularly polarized photons the quantum numbers of the final state are $l' = l + 1$ and $m_{l'} = m_l \pm 1$ due to the dipole selection rules, which were already stated in Sec. 1.3.

<table>
<thead>
<tr>
<th>$j, m_j$</th>
<th>$m_l, m_s$</th>
<th>$I^+_{jm}/R^2$</th>
<th>$I^-_{jm}/R^2$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\frac{3}{2}, +\frac{3}{2}$</td>
<td>$</td>
<td>+1\uparrow\rangle$</td>
<td>$\left(\frac{2}{5}\right)_\uparrow$</td>
</tr>
<tr>
<td>$\frac{3}{2}, +\frac{1}{2}$</td>
<td>$\sqrt{\frac{1}{3}}</td>
<td>+1\downarrow\rangle + \sqrt{\frac{2}{3}}</td>
<td>0\uparrow\rangle$</td>
</tr>
<tr>
<td>$\frac{3}{2}, -\frac{1}{2}$</td>
<td>$\sqrt{\frac{2}{3}}</td>
<td>0\downarrow\rangle + \sqrt{\frac{1}{3}}</td>
<td>1\uparrow\rangle$</td>
</tr>
<tr>
<td>$\frac{1}{2}, +\frac{1}{2}$</td>
<td>$\sqrt{\frac{2}{3}}</td>
<td>+1\downarrow\rangle - \sqrt{\frac{1}{3}}</td>
<td>0\uparrow\rangle$</td>
</tr>
<tr>
<td>$\frac{1}{2}, -\frac{1}{2}$</td>
<td>$\sqrt{\frac{1}{3}}</td>
<td>0\downarrow\rangle - \sqrt{\frac{2}{3}}</td>
<td>1\uparrow\rangle$</td>
</tr>
</tbody>
</table>
2.2. Two-step model of XMCD

Eq. (1.31). The electron spin is conserved in the absorption process. The matrix element results in

$$\langle n'l' = l + 1, m_l' = m_l \pm 1 | R_{\pm l} | nlm_l \rangle = -\sqrt{\frac{(l \pm m_l + 3)(l \pm m_l + 1)}{2(2l + 3)(2l + 1)}} R, \quad (2.3)$$

where the radial matrix element

$$R = \langle n'l' || \vec{R} || nl \rangle = \int r^3 R_{nl}^*(r) R_{n'l'}(r) d^3r \quad (2.4)$$

is independent of the magnetic quantum number and therefore independent of the photon helicity. The angular parts of the matrix elements in Eq. (2.3) for right and left circularly polarized light are listed in the second and third columns in Tab. 2.1, respectively. The arrows indicate the relative amount of spin-up and spin-down photoelectrons. As can be seen directly see from Eq. (2.3), it is equivalent to switch either the photon helicity or the magnetization, i.e. the magnetic quantum number $m_l \rightarrow -m_l$. By adding all contributions to one spin direction, one obtains the probability to excite e.g. a spin-up electron with a right circularly polarized photon at the $L_3$ edge,

$$p_{+1,\uparrow}(L_3) = \frac{\frac{2}{5} + \frac{2}{15} + \frac{1}{15}}{\frac{2}{5} + \frac{2}{15} + \frac{2}{15} + \frac{1}{45} + \frac{1}{15}} = 62.5\%, \quad (2.5)$$

and similarly

$$p_{-1,\uparrow}(L_3) = 37.5\%, \quad p_{+1,\uparrow}(L_2) = 25\%, \quad p_{-1,\uparrow}(L_2) = 75\%. \quad (2.6)$$

This is the first step of the model: The previously degenerated core electrons become spin-polarized in the absorption process. Right circularly polarized photons excite mainly spin-up core electrons at the $L_3$ edge and spin-down electrons at the $L_2$ edge. For left circularly polarized light the situation is vice versa. For a given photon polarization the sum of $2p_{3/2}$ and $2p_{1/2}$ contributions shows no spin-polarization, i.e. $p_{+1,\uparrow}(L_3 + L_2) = p_{+1,\downarrow}(L_3 + L_2) = 50\%$. Therefore the spin-polarization of the core electron arises through the spin-orbit splitting of $2p_{3/2}$ and $2p_{1/2}$ levels.

In the second step the $d$ band serves as a spin-resolving detector: According to Fermi’s golden rule the absorption is proportional to the density of final states. In a ferromagnetic metal the final $d$ states are exchange-split. If there are only unfilled spin-down states, for example, the detector is only sensitive to spin-down electrons and the dichroism effect is maximized. If both, spin-up and spin-down unfilled states are present, the dichroism is proportional to the difference of the spin-dependent density of states, i.e. to the magnetic spin moment.

From the discussion above it follows that the $L_3/L_2$ ratio of the integrated XMCD spectrum should be $-1 : 1$. In contrast to that, experimental spectra show a stronger XMCD at the $L_3$ edge. This effect is attributed to the spin-orbit coupling in the final states and therewith a non-vanishing orbital magnetic moment: The degeneracy of the $3d$ states is lifted and different final states are probed at the $L_3$ and $L_2$ edges according to the dipole selection rules \([71]\). Therefore the orbital moment is related to the sum of the $L_3$ and $L_2$ XMCD spectra, which is discussed in the next section.
2.3. Sum rules

A reason for the success of XMCD as a spectroscopic tool is the possibility to determine the spin and orbital contributions to the element-specific magnetic moment by means of sum rules. The sum rules were originally derived by Thole et al. [2] and Carra et al. [3] for single ions in a crystal field with partly filled valence shell. Even though the validity is limited by certain approximations, the applicability of the sum rules has been theoretically [63, 72, 73, 74] and experimentally verified for the 3d metals Fe and Co [75].

For transitions from the initial 2p3/2 and 2p1/2 into the 3d valence band, the orbital and spin magnetic moments \( m_{\text{orb}} \) and \( m_{\text{spin}} \) can be determined from XAS and XMCD spectra by the following equations [75]:

\[
\begin{align*}
m_{\text{orb}} &= -\frac{4}{3} \int_{L_3} (\mu_+ - \mu_-) dE \left( 10 - n_{3d} \right), \\
m_{\text{spin}} &= -\frac{6}{3} \int_{L_3} (\mu_+ - \mu_-) dE - \frac{4}{3} \int_{L_2} (\mu_+ - \mu_-) dE \left( 10 - n_{3d} \right) \left( 1 + \frac{7}{2} \frac{\langle T_z \rangle}{\langle S_z \rangle} \right)^{-1},
\end{align*}
\]

where \( n_{3d} \) is the number of occupied 3d states of the specific transition metal atom. \( \langle T_z \rangle \) is the expectation value of the magnetic dipole operator and \( \langle S_z \rangle \) is equal to half of \( m_{\text{spin}} \). The orbital and spin magnetic moments are given in units of \( \mu_B/\text{atom} \). The \( L_3 \) and \( L_2 \) denote the energy integration range. To apply the sum rules to measured data, the contribution of photoelectron excitations into continuum states must be subtracted from the absorption cross section. This background is usually removed by a step function [75], which can be written e.g. as [76]

\[
s = \frac{h}{3} \left[ 1 + \frac{2}{\pi} \arctan \left( \frac{E - E_{L_3}}{\Delta E} \right) \right] + \frac{h}{6} \left[ 1 + \frac{2}{\pi} \arctan \left( \frac{E - E_{L_2}}{\Delta E} \right) \right].
\]

The function mimics a step of 2/3 and 1/3 of the total step height \( h \) at the \( L_3 \) and \( L_2 \) edge energy, respectively, corresponding to the number of electrons in the core state. The steps are broadened over an energy range \( \Delta E \) due to finite instrumental resolution, temperature and final state lifetime.

Several approximations are made deriving the magneto-optical sum rules. It is therefore important to check under which conditions the application of the XMCD sum rules is justified and to find the accuracy for the magnetic moment deduced from them. First, \( 2p \to 4s \) transitions, which comply the dipole selection rules as well, are not taken into account. This is justified by the small density of states in the 4s band and the small radial matrix elements for transitions into 4s final states as compared to 3d final states. Therefore the \( 2p \to 3d \) transitions dominate the absorption process to 95% [71]. Second, the radial matrix element in Eq. (2.4) is assumed to be independent of the photon energy. Wu et al. [73] have shown that the calculated radial matrix elements for Ni increases by as much as 30% from the bottom to the top of the d band, partly explaining the breakdown of the spin sum rule in case of Ni. Third, the spin sum rule demands that the magnitude of angular momentum of the core hole, i.e. that of the core spin-orbit multiplets \( 2p_{1/2} \) and \( 2p_{3/2} \) in the \( 2p \to 3d \) absorption, is a good quantum number [77].
2.4. Total electron yield and saturation effects

While this condition seems to be satisfied for Fe, Co and Ni, the Coulomb interaction of the core hole with the valence electrons becomes comparable to the spin-orbit interaction for the early 3\textit{d} metals Sc, Ti, V and Cr, resulting in large errors for the spin sum rule \[78\]. Mn with five 3\textit{d} electrons seems to be at the border between early and late 3\textit{d} transition metals and the applicability of the spin sum rule is questionable as is further discussed in Chap. 8 \[79, 80\].

Another source of errors are the values of \(n_{3\textit{d}}\) and \(\langle T_z \rangle\), which have to be taken from theory. The magnetic dipole operator \(\langle T_z \rangle\) is a measure of the anisotropy of the spin density. While for systems with cubic symmetry the magnetic dipole term can be neglected, it becomes sizable if the cubic symmetry is broken e.g. at surfaces and interfaces. Neglecting \(\langle T_z \rangle\) can result in an error of up to 50\% e.g. for the Ni(001) surface \[73\].

2.4. Total electron yield and saturation effects

The most direct and accurate method to measure x-ray absorption spectra and circular dichroism is to measure directly the transmitted light \[75\]. In this case the absorption coefficient \(\mu_{\pm}\) is directly obtained from the transmitted intensity by the equation \(I = I_0 \exp(-\mu_{\pm} d)\), where \(d\) is the sample thickness. But only in very few cases the absorption has been measured in transmission geometry because it is often not practical. The small penetration depth of soft x-rays limits the transmission method to very thin free-standing films, which are difficult to prepare.

An alternative approach to obtain absorption spectra is to monitor the total electron yield (TEY) from a sample, which is defined as the sum of all photoelectrons created in the absorption process that escape from the sample \[81\]. The x-ray absorption cross section of an atom is directly proportional to the number of core holes created in the absorption process. These core holes are subsequently filled by Auger electron (99.2\%) and fluorescence decay (0.8\%) \[82\]. The further away the excited atom is from the surface, the less likely the primary Auger electrons will escape from the sample. The TEY signal is dominated by the secondary electrons, i.e. the inelastically scattered electron cascade originating from the primary Auger electrons \[83\]. In contrast to transmission measurements the TEY is a surface-sensitive method. The probed surface layer thickness is of the order of the electron escape depth \(\lambda_e \approx 2\) nm for 3\textit{d} transition metals \[84\]. The secondary electrons created by x-rays, which are absorbed deeper in the sample, cannot be detected. In this work the TEY has been measured by means of the sample drain current.

It has been shown that the TEY \(Y(E)\) is proportional to the product of absorption coefficient and photon energy, \(Y_\pm(E) \sim E \mu_{\pm}(E)\), if saturation effects can be neglected \[84, 85, 86\]. In transmission measurements, saturation in the signal occurs when either the sample thickness \(d\) or the absorption coefficient \(\mu_{\pm}\) are too large. At these conditions, the transmitted intensity \(I\) „saturates“ at zero, no matter what the variation in the energy-dependent \(\mu_{\pm}\) spectra might be. Correspondingly saturation occurs in TEY measurements, when the electron escape depth \(\lambda_e\) is of the same order as the x-ray penetration depth \(\sin \Theta / \mu_{\pm}(E)\) where \(\Theta\) is the angle of incidence measured with respect to the sample surface. Then again all x-rays are absorbed in the surface region and the
TEY saturates at the maximal signal possible for the incoming intensity $I_0$. Taking into account the fraction of photons absorbed in an increment $dz$
\[
\frac{\mu_{\pm}(E, z)}{\sin \Theta} dz
\]  
and the fraction of electrons escaping from the sample into vacuum
\[
\exp \left( -\int_0^z \frac{dz'}{\lambda_e(z')} \right),
\]
an expression for the TEY can easily be derived, depending on the photon energy and incidence angle \[81, 85, 86\]:
\[
Y_{\pm}(E) = T_0 N(E) \int_0^d \frac{\mu_{\pm}(E, z)}{\sin(\Theta)} \exp \left( -\int_0^z \left( \frac{\mu_{\pm}(E, z')}{\sin(\Theta)} + \frac{1}{\lambda_e(z')} \right) dz' \right) dz.
\]  
Here $T_0$ is the incident intensity transmitted into the sample and $N(E)$ is the number of created electrons per absorbed photon moving in direction of the sample surface. For x-rays $N(E)$ is in good approximation proportional to the photon energy. In the simplest case of a homogenous sample of semi-infinite thickness Eq. (2.12) reduces to
\[
Y_{\pm}(E) \sim E \frac{\mu_{\pm}(E)}{\sin \Theta} \left( 1 + \frac{\lambda_e}{\sin(\Theta)} \right)^{-1}.
\]  
In the limit $\sin \Theta/\mu_{\pm} \to 0$ this expression is independent of the absorption coefficient $\mu_{\pm}$ resulting in the saturation as described above. Since the electron escape depth $\lambda_e$ is a few nm while the x-ray penetration length $1/\mu$ is of the order of 50 - 100 nm, saturation effects lead to systematic errors smaller than 10% for incidence angles larger than 30° \[81\].
3. Specular reflectivity

3.1. Introduction

X-ray specular reflectivity methods have been widely applied over the last decades to characterize the structural properties of thin film systems and multilayers and their surfaces and interfaces. The quantitative analysis of specular reflectivity measurements is usually performed by numerical simulations. The sample properties are obtained by a best fit to the measured data. In this way thickness and interface roughness of buried layers can be determined, i.e. a electron density depth profile. Since at small angles total and multiple reflection effects cannot be neglected, the reflectivity has to be calculated dynamically in general. Kinematical diffraction theory, which does not take into account multiple reflection effects, is only appropriate for incidence angles that are large compared to the angle of total reflection. A standard dynamical approach to model the specular reflected intensity is the Parratt formalism [87] and its generalization to rough or graded interfaces [88].

X-ray resonant magnetic scattering (XRMS) offers the unique possibility to determine element-specific magnetization depth profiles of layered structures [33, 89, 90]. However, an adequate formalism for the simulation of specular XRMS cannot be a simple modification of the Parratt formalism: Even the basic assumption that the normal modes in a magnetic layer are transverse electromagnetic plane waves is not fulfilled in general as has been discussed in Sec. 1.2. In this chapter a formalism for magneto-optics with arbitrary magnetization direction is presented, which was developed by Zak et al. [59, 91, 92]. Since there are no restrictions regarding the geometry, i.e. the magnetization direction, angle of incidence and polarization, the formalism is most suitable for the simulation of all soft x-ray experiments as long as they fulfill the specular condition [55, 93]. The formalism deals with magneto-optical effects up to first order in the Voigt parameter $Q$ which is equivalent to a negligence of the term proportional to $(\vec{m} \cdot \vec{\epsilon}_f)(\vec{m} \cdot \vec{\epsilon}_i)$ in the resonant magnetic scattering amplitude, as has been shown in Sec. 1.4. Therefore second-order effects like x-ray magnetic linear dichroism (XMLD) cannot be described. Within the framework of the present study a computer code has been written based on the Zak formalism. Structural and magnetic roughness have been taken into account in terms of graded electron density and magnetization profiles using a slicing method.

In the meantime Lee et al. have given a theoretical formulation of x-ray resonant magnetic specular reflectivity [94] and diffuse scattering [95] from rough surfaces and interfaces within the distorted-wave Born approximation. However, both approaches should yield equivalent results.

The chapter is built up as follows: In the next two sections the basic ideas and formulas of the magneto-optical approach for arbitrary layered media with flat interfaces...
are summarized following the papers by Zak et al. \[59, 92\]. In Sec. 3.4 it is shown how roughness can be included in the formalism. Subsequently several simple examples for the specular reflectivity of a bulk Fe substrate and a thin Fe layer in different geometries are given to illustrate the dependence of the resonant magnetic specular reflectivity on the magnetization direction and incoming polarization.

3.2. Two media separated by a single boundary

The incoming and outgoing electromagnetic waves in a scattering experiment are usually described in a basis of polarization states perpendicular (\(E_{\sigma}\)) and parallel (\(E_{\pi}\)) to the plane of incidence. These fields can be collected in a four-component vector

\[
\vec{P} = \begin{pmatrix}
E_{\sigma}^{(i)} \\
E_{\pi}^{(i)} \\
E_{\sigma}^{(r)} \\
E_{\pi}^{(r)}
\end{pmatrix},
\]

(3.1)

where \(i\) and \(r\) denote the incident and reflected wave, respectively. Because a beam travelling from medium 1 into medium 2 (see Fig. 3.1(b)) conserves the tangential components of its electric (\(E_x, E_y\)) and magnetic (\(H_x, H_y\)) fields, it is more useful to change to this basis and define

\[
\vec{F} = \begin{pmatrix}
E_x \\
E_y \\
H_x \\
H_y
\end{pmatrix}.
\]

(3.2)

Now a matrix can be defined, that connects \(\vec{F}\) with \(\vec{P}\). This matrix is of central importance in this approach and is called the medium boundary matrix \(A\),

\[
\vec{F} = A\vec{P}.
\]

(3.3)

The boundary matching condition (\(\vec{F}_1 = \vec{F}_2\)) can now be expressed as

\[
A_1\vec{P}_1 = A_2\vec{P}_2.
\]

(3.4)

Once the medium boundary matrix \(A\) is found, only a simple vector equation is left for the calculation of reflected and transmitted waves. The arbitrary direction of the magnetization \(\vec{M}\) is specified by the polar coordinates \(\phi\) and \(\gamma\) in the \(xyz\) system (Fig. 3.1(a)),

\[
M_x = M \sin \phi \cos \gamma
\]

(3.5)
\[
M_y = M \sin \phi \sin \gamma
\]

(3.6)
\[
M_z = M \cos \phi.
\]

(3.7)

The dielectric tensor is written in the notation introduced in Eq. (1.18) for the polar geometry with \(\vec{M}\) along the \(z\) direction. The charge and magnetic contributions to the complex refractive index are given by \(N = 1 - \delta + i\beta\) and \(Q = -\Delta\delta + i\Delta\beta\), respectively,
3.2. Two media separated by a single boundary

Figure 3.1.: (a) Spherical coordinates for the magnetization in the $xyz$ coordinate system. (b) The four normal modes of the electromagnetic wave in a magnetic material. $\theta^{(i)}$ are the angles between the propagation directions and the $z$ direction. Both figures are taken from Ref. [59].

c.f. Eq. (1.20). A coordinate transformation yields an expression for $\epsilon$ for an arbitrary direction of $\vec{M}$,

$$
\epsilon(\omega) = N^2 \begin{pmatrix}
1 & i \cos \phi Q & -i \sin \gamma \sin \phi Q \\
-i \cos \phi Q & 1 & i \cos \gamma \sin \phi Q \\
i \sin \gamma \sin \phi Q & -i \cos \gamma \sin \phi Q & 1
\end{pmatrix}.
$$

(3.8)

As it was explained in detail in section 1.2, the components of $\vec{E}$ in an normal mode in the magnetic material are related by the equation,

$$
\frac{Dy'}{Dx} = \pm i,
$$

(3.9)

where $y'$ is in the direction perpendicular to both the D-wave propagation vector $\vec{k}$ and the $x$ direction (Fig. 3.1(a)). The $\pm$ signs represent the two circularly polarized waves, see Eq. (1.21). As shown in Fig. 3.1(b) there are four waves propagating in the magnetic layer corresponding to incident (1,2) and reflected (3,4) circularly polarized D-waves with positive and negative helicity. Snell’s law holds for each of these waves,

$$
N_1 \sin \theta_1 = N \sin \theta = n^{(1,2)} \sin \theta^{(1,2)} = n^{(3,4)} \sin \theta^{(3,4)},
$$

(3.10)

where $N_1$ is the refractive index of medium 1, $\theta_1$ is the angle of incidence (measured from the sample normal), $N$ is the refractive index of the magnetic medium and $\theta$ is measured with respect to the sample normal, in contrast to $\Theta$, which is measured from the sample surface.

\[1\text{In order to be consistent with the original work by Zak et al. [59], in this section the angle of incidence}\]
3. Specular reflectivity

an auxiliary angle. The refractive index for these normal modes was already given in Eq. (1.20) to first order in the Voigt parameter $Q$,

$$n_{i,r} = N(1 \pm \frac{1}{2}g_{i,r}Q), \quad (3.11)$$

where

$$g_{i} = \alpha_{z} \cos \phi + \alpha_{y} \sin \phi \sin \gamma, \quad g_{r} = -\alpha_{z} \cos \phi + \alpha_{y} \sin \phi \sin \gamma \quad (3.12)$$

with $\alpha_{z} = \cos \theta$ and $\alpha_{y} = \sin \theta$. It is now tedious but straightforward to find the relations between the components of the electric field vector $\vec{E}$ in the magnetic medium using Eqs. (3.8)-(3.11). The calculation is not presented here. Details can be found in Refs. [59, 91, 93]. According to Maxwell’s equations the components of the magnetic field vector are connected to $\vec{E}$ via $\vec{H} = \vec{n} \times \vec{E}$, where $\vec{n}$ is in the propagation direction and its magnitude is the refractive index. Finally one gets the following expression for the medium boundary matrix:

$$A = \begin{pmatrix} 1 & 0 & 0 & 0 \\ \frac{i}{2}g_{i}QN & -N & \frac{i}{2}g_{r}QN & 0 \\ 0 & -N & \frac{i}{2}g_{i}QN & 0 \\ \frac{i}{2}g_{r}QN & 0 & -N & -\frac{i}{2}g_{r}QN \end{pmatrix}. \quad (3.13)$$

In the non-magnetic case $Q$ is set equal to zero and $A$ reduces to

$$A = \begin{pmatrix} 1 & 0 & 1 & 0 \\ 0 & \alpha_{z} & 0 & -\alpha_{z} \\ 0 & -N & 0 & -N \\ \alpha_{z}N & 0 & -\alpha_{z}N & 0 \end{pmatrix}. \quad (3.14)$$

3.3. Multilayers

So far only a single interface between two media has been considered. In a multilayer system a medium propagation matrix $\bar{D}$ is needed additionally to the medium boundary matrix $A$. $\bar{D}$ describes the phase shift and absorption during propagation in the layer:

$$\bar{D} = \begin{pmatrix} U & U\delta_{i} & 0 & 0 \\ -U\delta_{i} & U & 0 & 0 \\ 0 & 0 & U^{-1} & -U^{-1}\delta_{r} \\ 0 & 0 & U^{-1}\delta_{r} & U^{-1} \end{pmatrix}, \quad (3.15)$$

with

$$U = \exp(-i\frac{2\pi}{\lambda}N\alpha_{z}d), \quad (3.16)$$
3.4. Roughness and resolution

\[ \delta_{i,r} = \frac{\pi}{\lambda} Nd \frac{Q}{\alpha_z} g_{i,r}, \]  

where \( d \) is the film thickness. The matrix \( \bar{D} \) is again given to first order in \( Q \). Finally, the boundary matching condition in Eq. (3.4) for an arbitrary multilayer system can be written in the form

\[ \bar{P}_i = M \bar{P}_f, \]  

with

\[ M = A^{-1}_i \prod_{m=1}^{l} (A_m \bar{D}_m A^{-1}_m) A_f \equiv \begin{pmatrix} G & H \\ I & J \end{pmatrix}. \]  

Here \( G, H, I \) and \( J \) are \( 2 \times 2 \) matrices. The matrix \( A_i \) describes the boundary to vacuum, the matrix \( A_f \) represents the last interface to the (infinitely thick) substrate. With the knowledge of the refractive index, Voigt parameter, magnetization direction and angle of incidence, the matrix \( M \) can be calculated. The vector \( \bar{P}_i \) contains only the transmitted beam, a reflected beam in the substrate does not exist. The polarization of the incident beam is given by the first two components of \( \bar{P}_i \). Therefore one is left with a system of four coupled linear equations for the four unknown quantities \( E_{\sigma}^{(t)}, E_{\pi}^{(t)}, E_{\sigma}^{(r)} \) and \( E_{\pi}^{(r)} \). It can be shown that the \( 2 \times 2 \) matrices defined in Eq. (3.19) are related to the transmission (\( t \)) and reflection (\( r \)) magneto-optic coefficients by

\[ \begin{pmatrix} t_{ss} & t_{sp} \\ t_{ps} & t_{pp} \end{pmatrix} = G^{-1}, \quad \begin{pmatrix} r_{ss} & r_{sp} \\ r_{ps} & r_{pp} \end{pmatrix} = IG^{-1}. \]  

(3.20)

With knowledge of these coefficients all reflected and transmitted intensities, Faraday and Kerr rotations etc. can be calculated.

3.4. Roughness and resolution

In this section it is shown how interface roughness is included in the formalism presented above. In the specular direction there is no component of the scattering vector parallel to the sample surface. Therefore graded interfaces (interdiffusion of the two materials) and interface roughness (height fluctuations) cannot be distinguished. The height distribution is simply averaged over the \( xy \) plane. The density profile of a graded interface is usually described by an error function. For a single boundary of two media in the \( xy \) plane at \( z = 0 \) with refractive indices \( n_1 \) for \( z < 0 \) and \( n_2 \) for \( z > 0 \) the refractive index profile can be written as

\[ n(z) = \frac{n_2 - n_1}{2} \text{erf} \left( \frac{z}{\sqrt{2} \sigma} \right) + \frac{n_1 + n_2}{2}. \]  

(3.21)

This is equivalent to a Gaussian interface height distribution centered at \( z = 0 \). The rms (root mean square) roughness \( \sigma \) is the width of the Gaussian distribution. An example for the error function is shown as solid line in Fig. 3.2(a). Numerically the graded interface is realized by a slicing method [96]. The interface region between \( z_1 = -2\sqrt{2} \sigma \) and \( z_2 = +2\sqrt{2} \sigma \) is divided into \( N \) different layers (typically \( N = 20 \)) of equal thickness with sharp boundaries and a constant refractive index which is calculated for the center.
3. Specular reflectivity

(a) Refractive index depth profile of a graded interface with $\sigma = 0.2$ nm separating two media with $n = 0.6$ for $z < 0$ and $n = 1$ for $z > 0$. The bars represent the slicing of the interface used in the computer code.

(b) Reflectivity of a rough MgO substrate calculated applying the slicing method and comparison to the Fresnel reflectivity.

Figure 3.2.: Implementation of roughness in the Zak formalism.

of each slice according to Eq. (3.21). At depth $z_{1,2}$ the refractive index has reached 99.99% of the bulk value $n_{1,2}$. The matrix product in Eq. (3.19) is then calculated for this stack of layers.

The generalization of Eq. (3.21) to a multilayer with $k$ interfaces yields

$$n(z) = n_1 + \sum_{i=1}^{k-1} \frac{n_{i+1} - n_i}{2} \left[ \text{erf} \left( \frac{z - z_i}{\sqrt{2}\sigma_i} \right) + 1 \right],$$

(3.22)

where $z_i$ is the position of the $i$th interface and $\sigma_i$ is the corresponding roughness. If two neighboring interfaces $i$ and $i + 1$ are separated by less than a distance of $z_{i+1} - z_i \leq 2\sqrt{2}(\sigma_i + \sigma_{i+1})$ the slicing procedure has to be modified slightly: The combined interface region limited by $z_i - 2\sqrt{2}\sigma_i < z < z_{i+1} + 2\sqrt{2}\sigma_{i+1}$ is divided into $2N$ slices of equal thickness with refractive indices according to Eq. (3.22). This procedure can be generalized for an arbitrary number of overlapping interface regions. In Fig. 3.2(b) the reflectivity of an MgO substrate with varying roughness is shown, demonstrating that the slicing method yields the same results as the approach of Nevot and Croce [88].

Since the magnetization is a vectorial quantity one can think of different kinds of roughness: At a magnetically rough interface either the absolute value of the magnetic moment might fluctuate or the magnetization direction can fluctuate similar to ripple domains. However, fluctuations of the direction will manifest themselves in off-specular scattering. In the specular reflectivity only the average magnetization direction can be measured given by the angles $\gamma$ and $\phi$ (Fig. 3.1(a)) for each layer. Magnetic interface roughness therefore is treated in the same manner as structural roughness here. The positions of the magnetic interfaces have not necessarily to coincide with those of the
3.5. Examples

In this section simple examples for specular reflectivity calculations are presented. The aim is on the one hand to check the correctness of the computer code. Therefore numerical calculations and analytical formulae are compared. On the other hand the dependence of the specular magnetic reflectivity on the incoming polarization and magnetization direction is illustrated. Different possible experimental geometries are discussed. The magneto-optical constants of Fe in the vicinity of the $L_{2,3}$ absorption edges used as input in the calculations are taken from absorption measurements in transmission geometry [75]. The real and imaginary part has been calculated as described in Sec. 1.5.

Figure 3.3.: Reflectivity of a $[\text{Fe}(1.1 \text{ nm})/\text{Cr}(0.8 \text{ nm})]_{30}$ superlattice at $E = 700$ eV depending on the resolution. For a better comparison the curves are shifted by factors $1/5$ against each other.

Chemical ones, e.g. in presence of magnetically dead layers. Therefore the depth profiles of the chemical and magnetic refractive indices are calculated independently.

Finally, the instrumental angular resolution is taken into account by

$$I_{\text{corr}}(\Theta) = \frac{1}{\Delta\Theta \sqrt{\pi}} \int d\Theta' I(\Theta') \exp\left(-\frac{(\Theta' - \Theta)^2}{\Delta\Theta^2}\right), \quad (3.23)$$

where $I(\Theta')$ is the reflected intensity for an ideal angular resolution and $I_{\text{corr}}$ is intensity including a finite resolution $\Delta\Theta$. As an example in Fig. 3.3, the reflectivity of a $[\text{Fe}(1.1 \text{ nm})/\text{Cr}(0.8 \text{ nm})]_{30}$ superlattice at $E = 700$ eV is shown in the range of the first-order Bragg reflection. With decreasing angular resolution (increasing $\Delta\Theta$) the Laue oscillations are smoothed and cannot be resolved anymore for $\Delta\Theta = 0.3^\circ$. 
3. Specular reflectivity

![Graphs showing specular reflectivity](image)

Figure 3.4.: Charge (a) and magnetic (b) contribution to the complex refractive index of Fe at the $L_{2,3}$ absorption edges (taken from Ref. [75]).

The charge and magnetic contributions to the complex refractive index of Fe are shown in Fig. 3.4.

Exact expressions for the reflection coefficients can be given only in very few cases. One of them is the polar geometry (P-MOKE), where the magnetization is perpendicular to the sample surface. For perpendicular incidence the reflection coefficient take a simple form, given by [57]

$$r_{\pm} = \frac{n_{\pm} - 1}{n_{\pm} + 1},$$

with refractive indices $n_{\pm} = N(1 \pm Q/2)$. Fig. 3.5(a) shows the reflected intensity $I_{\pm} = |r_{\pm}|^2$ of both helicities for perpendicular incidence, which were calculated both analytically and numerically. The refractive index has been taken from absorption data [75]. For other angles of incidence approximate analytical expressions for the reflection coefficients $r_{ss}$, $r_{pp}$, $r_{sp}$ and $r_{ps}$ have been derived [57]. Results of analytical and numerical calculations are plotted in Fig. 3.5(b) again yielding identical results.

Further tests of the computer code were performed, which are not shown here: For example the calculation of transmission coefficient for thin Fe layers exactly reproduces the XMCD results. The computer code is suitable for the calculation of non-resonant reflectivity in the hard x-ray range. The results are identical to data obtained with the commercial software WinGIXA [97].

Experiments of the specular magnetic reflectivity can be viewed as variants of the magneto-optical Kerr effect (MOKE) which can be realized in different geometries. The most common geometry for XRMS on ferromagnetic samples is the longitudinal MOKE (L-MOKE) geometry, where the magnetic moments are parallel to the plane of incidence and parallel to the sample surface. Circularly polarized light has to be used. As a measure for the magnetic contribution to the reflection coefficients, the intensity difference $I_+ - I_-$ or asymmetry $(I_+ - I_-)/(I_+ + I_-)$ is used. Here $\pm$ can indicate either the two helicities or two antiparallel orientations of the magnetization. In most experiments
3.5. Examples

(a) Energy-dependent reflectivity for circular polarization at normal incidence ($2\Theta = 180^\circ$).

(b) Reflectivity at $E = 706$ eV for both linear and circular incoming polarizations.

Figure 3.5.: Energy- (a) and angular-dependent (b) reflectivity in the P-MOKE geometry for an out-of-plane magnetized ($\gamma = 0^\circ$, $\phi = 180^\circ$) Fe bulk sample. Analytically calculated curves (lines) and results of the Zak computer code (symbols) are compared.

A switching of the external magnetic field is preferred, because it is easier to realize technically. Note that not the L-MOKE is measured directly, i.e. the rotation of the polarization plane of linearly polarized light, but the reflected intensity of circular light without polarization analysis in the L-MOKE geometry.

Fig. 3.6 shows the specular reflectivity for a bulk Fe substrate (A) and a 10 nm thick Fe film on MgO (B) in the L-MOKE geometry calculated for both helicities at $E = 706$ eV. The reflectivities show the typical Fresnel decay $(1 + \cos^2 \Theta)/2 \cdot q_z^{-4}$ for circularly polarized light. The thin film fringes in B not only differ in intensity but also are shifted against each other due to the real part of the Voigt parameter. Therefore the maxima in the intensity difference do not exactly coincide with the thin film oscillation maxima. The intensity difference plotted in the lower part of Fig. 3.6 has been divided by the Fresnel decay $q_z^{-4}$ for a better comparison at different angles. The largest magnetic signal is obtained at reflection angles between $2\Theta = 20^\circ$ and $40^\circ$ for A and B. The magnetic signal of the thin film can be strongly increased as compared to the bulk due to interference of amplitudes reflected from the surface and the substrate.

This example also shows that it is difficult to draw conclusions about the relative magnetization orientation of different layers in a multilayer system, because the sign of the intensity difference can change as a function of the reflection angle (e.g. at $2\Theta = 14^\circ$). The asymmetry in the inset of Fig. 3.6 is sometimes misleading because the magnetic signal in the oscillation minima is overestimated due to the normalization to the averaged intensity. It is not always advisable to measure e.g. hysteresis loops in the position of the highest asymmetry because the signal-to-noise ratio might be unsatisfactory.
Figure 3.6.: Reflectivity $I_+$ and $I_-$ and difference $I_+ - I_-$ for an Fe bulk sample (A) and 10 nm Fe on a MgO substrate (B) at a photon energy of $E = 706$ eV. For clarity reflectivity A is multiplied by a factor of 10, while the differences are kept on the same scale. The inset shows the asymmetry for B.

The transversal Kerr effect (T-MOKE) is observed when light is reflected from a sample with its magnetic moments lying parallel to the surface and perpendicular to the plane of incidence. Commonly linearly polarized light with polarization vector in the plane of incidence ($\pi$ geometry) is used. The magnetic contribution is given by the difference of reflected intensities for the two antiparallel orientations of the magnetization at fixed polarization or the corresponding asymmetry $(I(\gamma = 0^\circ) - I(\gamma = 180^\circ))/(I(\gamma = 0^\circ) + I(\gamma = 180^\circ))$. In Fig. 3.7 the reflectivity and asymmetry in the T-MOKE geometry are shown. Since T-MOKE is caused by the interference of charge and magnetic $\pi-\pi$ scattering, cf. Eq. (1.47), $\sigma$ polarized light is independent of the sample magnetization. The T-MOKE is maximum for $\pi$ polarized light close to the Brewster angle $\cot \Theta = n$, which is $\Theta = 45.06^\circ$ for Fe at a photon energy of 710 eV. For a non-magnetic sample the $\pi$ reflectivity would vanish there. Therefore the intensity is mainly caused by magnetic scattering. Since with the charge scattering the charge-magnetic interference term is vanishing as well, this angle corresponds approximately to the zero crossing of the asymmetry in Fig. 3.7(b). The largest asymmetry is obtained for intermediate angles where the charge and magnetic scattering amplitudes are in the same order of magnitude.

The T-MOKE can be measured with circular light as well, since it contains a $\pi$ component. The T-MOKE of circular light corresponds to the second term in Eq. (1.51), which does not change the sign with a reversal of the photon helicity. In contrast to the L-MOKE geometry the asymmetry is again defined as difference of the two antiparallel magnetization directions for one helicity. As can be seen in Fig. 3.7(b) the effect is much

\footnote{Note that in optics the angle of incidence is usually measured from the sample normal.}
\footnote{The Brewster angle of the magnetic sample is slightly modified by contributions second-order in $Q$ (purely magnetic $\pi-\pi$ scattering) as compared to the non-magnetic sample.}
3.5. Examples

![Graphs](image.png)

Figure 3.7.: Reflectivity (a) and asymmetry for $\pi$ linearly and right circularly polarized (RCP) light (b) in the T-MOKE geometry of an Fe substrate at $E = 710$ eV.

smaller than in the $\pi$ geometry.

To further elucidate this point, the reflected intensity at $E = 710$ eV is plotted as a function of the magnetization direction in the sample plane in Fig. 3.8(a). The reflection angle $2\Theta = 54^\circ$ corresponds to the maximum T-MOKE asymmetry of circularly polarized light in Fig. 3.7(b). The amount of magnetic scattering is seen by comparison with the intensities $I(Q = 0)$ for the non-magnetic sample (dotted lines). The magnetic contributions can be directly related to the polarization factors of the magnetic scattering amplitudes in Eqs. (1.47) and (1.51):

- Linear $\sigma$ polarized light does not show any magnetooptical effect to first order in $Q$. There is a small intensity variation $\sim \sin^2 \gamma$ due to purely magnetic $\sigma \rightarrow \pi$ scattering.

- The charge-magnetic interference term of $\pi$ polarized light is sensitive to the magnetization direction perpendicular to the scattering plane to first order in $Q$. Contributions of purely magnetic scattering $O(Q^2)$ result from $\pi \rightarrow \sigma$ and $\pi \rightarrow \pi$ scattering. The purely magnetic scattering is much smaller and can be neglected for ferromagnetic samples.

- The intensity difference of right and left circular light is sensitive to the magnetization component along the scattering plane (L-MOKE, Fig. 3.8(b)). There is also a contribution of T-MOKE independent of the helicity. In total the magnetic scattering is proportional to a term $I_m \sim \pm a \sin \gamma - b \cos \gamma$ to first order in $Q$, which can be directly identified with the two terms in Eq. (1.51).

With polarized neutron scattering (PNR) vector magnetometry is possible by measuring the non-spin-flip ($R^{++}$, $R^{--}$) and spin-flip ($R^{+-}$, $R^{-+}$) reflectivities. The above
3. Specular reflectivity

Figure 3.8.: (a) Reflected intensity as function of the in-plane magnetization direction of an Fe substrate between the Fe \( L_3 \) and \( L_2 \) edges, \( E = 710 \text{ eV} \) and \( 2\Theta = 54^\circ \). The reflected intensities obtained for \( Q = 0 \) are shown as dotted lines for each incoming polarization. (b) Asymmetry of left and right circularly polarized light.

Examples show that vector magnetometry measurements are difficult with soft x-rays, because relations between incoming polarization and magnetization direction are complicated. This aspect is further discussed in Chap. 7.

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4. Diffuse scattering

4.1. Introduction

In the previous chapter the theoretical description of scattering was restricted to the specular condition. Besides the layer thickness the specular reflectivity yields information on the rms interface roughness as well. But for example interdiffusion and roughness cannot be distinguished, since the information in the specular reflectivity is averaged in the lateral directions. An important consequence of the presence of (correlated) roughness is that the scattered beam is no longer strictly specular, as in the case of ideally flat or graded interfaces. Instead it develops a diffuse, off-specular component. Therefore the interface morphology of a layered system can be characterized by studying the diffuse scattering. This information is complementary to e.g. microscopy pictures, because diffuse scattering yields global statistical information on the interface morphology. Furthermore due to the penetration depth of x-rays it is possible to study buried interfaces. Theoretically the specular as well as the diffusely scattered intensity can be calculated within kinematical theory, i.e. the first-order Born approximation (BA). The approach is sufficient for the description of experiments, if the incidence angles are large compared to the critical angle and no multiple scattering occurs. By tuning the photon energy to an appropriate absorption edge and using the mechanism of x-ray resonant magnetic scattering (XRMS), magnetic interface properties can be determined, similar to polarized neutron scattering. If multiple-scattering effects cannot be neglected, e.g. for incidence angles in the region of the critical angle, the BA breaks down. In this case the distorted-wave Born approximation (DWBA) has to be used for a quantitative description of experiments.

In the next section BA for pure charge scattering is discussed. In section 4.3 the theory is generalized to the case of resonant magnetic scattering. The DWBA is briefly discussed in section 4.4.

4.2. Born approximation

Fig. 4.1 illustrates the scattering of an x-ray beam from a single rough surface. Within BA the amplitude of the reflected beam is calculated by summing the amplitudes of all scatterers within the illuminated volume $V$ taking into account the appropriate phase factor. If the probed length scale is large compared to the atomic radii, i.e. $qa \ll 1$, the material can be assumed to be homogeneous and the sum is replaced by an integral.
4. Diffuse scattering

Figure 4.1.: Scattering by a single rough surface.

The differential cross section in BA is then given by

$$\frac{d\sigma}{d\Omega} = \left| \rho^2 \int_V d^3r \exp(-i\vec{q} \cdot \vec{r}) \right|^2,$$

with $\rho = Nf(E)$, where $f(E)$ is the atomic scattering amplitude and $N$ is the number density of scatterers. For simplicity polarization factors are avoided in this section, all expressions are valid for $\sigma$ polarization. The absolute square of the amplitude can be written as a double integral and, using the Gauss theorem, the volume integrals can be transformed into surface integrals. To evaluate Eq. (4.1) it is assumed that the difference in heights of the surface above the $xy$ plane $[z(x',y') - z(x,y)]$ is a Gaussian random variable, which depends only on the relative difference in position $(X, Y) = (x' - x, y' - y)$.[98] This leads to

$$\frac{d\sigma}{d\Omega} = \frac{\rho^2}{q_z^2} L_x L_y \int_S dX dY e^{-q_z^2g(X,Y)} e^{i(q_x X + q_y Y)},$$

where $g(X,Y) = \langle [z(x',y') - z(x,y)]^2 \rangle$ and $S = L_x L_y$ is the illuminated area. For $g(X,Y)$ various models can be assumed. Here only the model of a self-affine, correlated, isotropic rough surface including a cut-off length is presented, which ensures finite height fluctuations for $R \to \infty$, where $R = (X^2 + Y^2)^{1/2}$. In this model the representation for $g(R)$ is

$$g(R) = 2\sigma^2 \left[ 1 - \exp \left( -\frac{R}{\xi} \right) \right].$$

In Eq (4.3) several parameters have been introduced: the rms roughness $\sigma = \langle z^2 \rangle$ is the average height fluctuation of the surface, the correlation length $\xi$ has the function of an effective cut-off. For in-plane distances larger than $\xi$ the surface appears smooth while for distances smaller than $\xi$ it appears as a rough surface with fractal behavior. The surface fractal dimension $D$ is connected to the hurst parameter $h$ $(0 < h < 1)$ by $D = 3 - h$. Small values of $h$ produce extremely jagged surfaces, while values of $h$ approaching 1 appear to have smooth hills and valleys. If Eq. (4.3) is inserted into Eq. (4.2), the differential cross section naturally splits up into a specular part restricted to $q_x = q_y = 0$ and a diffuse part. As shown in Ref. [98] the final result for the diffuse
scattering of a single surface is

\[
\left( \frac{d\sigma}{d\Omega} \right)_{\text{diffuse}} = \frac{L_x L_y}{q_x^2} \rho^2 e^{-q_x^2 \sigma^2} \int_S dX dY \left( e^{i q_x C(X,Y)} - 1 \right) e^{-i(q_x X + q_y Y)},
\]

where the height-height correlation function \( C(R) =< z(0) z(R) > \) has been introduced. It is connected to \( g(R) \) via

\[
C(R) = \sigma^2 - \frac{1}{2} g(R) = \sigma^2 \exp \left( -\frac{R}{\xi} \right).
\]

Eq. (4.4) can only be solved numerically in general. The parameters \( \sigma, \xi \) and \( h \) are then determined by fitting the model curve to measured data.

So far only a single surface has been considered. For multilayer systems Eq. (4.4) can easily be generalized by introducing height-height correlation functions for each pair of interfaces,

\[
C_{ij}(R) = \frac{\sigma_i \sigma_j}{2} \left( \exp \left( -\frac{R}{\xi_i} \right) + \exp \left( -\frac{R}{\xi_j} \right) \right) e^{-|z_i - z_j|/\xi},
\]

and summing the contribution of each interface (4.4) over all interfaces \( i \) and \( j \) [99,100],

\[
\left( \frac{d\sigma}{d\Omega} \right)_{\text{diffuse}} = \frac{L_x L_y}{q_x^2} \sum_{ij} \Delta \rho_i^2 \Delta \rho_j^2 e^{-q_x^2 (\sigma_i^2 + \sigma_j^2)/2} e^{i q_x (z_i - z_j)} S_{ij}(\vec{q}).
\]

Here \( S_{ij}(\vec{q}) \) represents the integral in Eq. (4.4) with \( C \) replaced by \( C_{ij} \), \( \Delta \rho_i \) is the difference in \( \rho \) across the \( i \)th interface. The out-of-plane roughness correlation of the different interfaces is described by the perpendicular correlation length \( \xi_\perp \). One can think of two extreme cases [101]: If the different interfaces are completely uncorrelated, \( \xi_\perp = 0 \), then the height-height correlation function \( C_{ij} \) is zero except for \( i = j \) and the diffuse intensity is the incoherent superposition of the diffuse scattering from each interface. In this case the off-specular intensity is smeared over the full reciprocal space, since the phase factor \( \exp(iq_x(z_i - z_j)) = 1 \) is now independent of \( q_x \). If on the other hand all interfaces in a periodic multilayer are vertically correlated, i.e. if \( \xi_\perp \) is larger than the total layer thickness, the phase factor leads to a concentration of the diffuse intensity at the superlattice peak position \( q_x = 2\pi/\Lambda \), due to a coherent superposition of all interface contributions, where \( \Lambda = z_{i+1} - z_i \) is the bilayer thickness.

All diffuse scattering measurements presented in this thesis are obtained by using a slit in front of the detector, which is perpendicular to the scattering plane. In this way the off-specular intensity is integrated in \( q_y \) direction. Typically two different kind of scans are used to map the diffuse intensity in reciprocal \( q_x - q_z \) space. A transverse \( q_x \) scan at fixed \( q_z \) is useful to determine the in-plane correlation length \( \xi \) and the hurst parameter \( h \). Sometimes a rocking scan is carried out instead, where the sample is rotated at a fixed detector angle \( 2\Theta \) [9]. In a longitudinal off-specular scan \( q_z \) is varied at a small, constant \( q_x \) value close to the specular ridge. In this way information on the correlation of interfaces among each other is obtained. Again a \((\Theta + \Delta \Theta, 2\Theta)\) scan can be alternatively used.

For a fit of Eq. (4.7) to experimental data, it has to be taken into account that the illuminated surface area depends on the angle of incidence \( \omega \). If the full incoming beam
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hits the sample, this effect can be corrected by multiplying the model curve [4.7] by a factor \( \sin \Theta / \sin \omega \) [102].

Due to a change in path length of the x-rays in the sample with \( \omega \), the beam attenuation depends on the incidence and reflection angles. Therefore, if the absorption is too large to be neglected, an additional correction factor has to be multiplied, which can be written as [102]

\[
F(2\Theta, \omega) = \frac{1 + \exp \left[ -N\alpha(2\Theta, \omega) \right] - 2 \exp \left[ -N\alpha(2\Theta, \omega)/2 \right]}{1 + \exp \left[ -\alpha(2\Theta, \omega) \right] - 2 \exp \left[ -\alpha(2\Theta, \omega)/2 \right]},
\]

with

\[
\alpha(2\Theta, \omega) = (d_1\mu_1 + d_2\mu_2) \left( \frac{1}{\sin \omega} + \frac{1}{\sin(2\Theta - \omega)} \right),
\]

where \( \mu_{1,2} \) and \( d_{1,2} \) are the absorption coefficients and layer thicknesses of material 1 and 2 and \( N \) is the number of bilayers in the sample. This correction factor is especially important close to absorption edges in the soft x-ray range.

4.3. X-ray resonant magnetic diffuse scattering

Conventional diffuse x-ray scattering measurements, as discussed in the previous section, are not sensitive to the magnetic sample properties. The magnetic analogue of such experiments is diffuse XRMS. Experiments in the soft x-ray region have been carried out on ferromagnetic films with circular polarization measuring the \( q \) component perpendicular to the scattering plane [35] or in the scattering plane [36, 103] similar to the longitudinal Kerr (L-MOKE) geometry for specular reflectivity measurements (cf. Chap. 1), and on antiferromagnetically-coupled (AF) superlattices using linear \( \sigma \) polarization [37].

In experiments performed in the L-MOKE geometry usually the intensity difference of both photon helicities is measured (or equivalently the difference of the parallel and antiparallel magnetization saturation direction). It has been pointed out that this difference is not purely magnetic in nature and contains both charge and magnetic contributions, as will be shown below [103]. Only for AF-coupled superlattices a separation of charge and magnetic scattering has been possible so far.

At least three different mechanisms can be distinguished, leading to a magnetic contribution to the off-specular intensity [37, 103]. The first, in which the moment direction does not change, is directly connected to the chemical composition across the interface and should be correlated with the structural roughness. A second contribution to magnetic roughness arises from fluctuations in the magnetization direction, e.g. ripple domains. The third contribution to diffuse scattering is caused by magnetic domains, even in the absence of magnetic interface roughness.

In this section expressions for off-specular XRMS are presented, which are used in Part III of the present thesis to analyze experimental data. The theoretical description of diffuse XRMS is given on the level of the first-order BA as it has been developed in Refs. [95, 104, 105]. This seems to be sufficient since in the soft x-ray range the scattering angles are generally large compared to the critical angle and multiple scattering effects are negligible at least for the measurements discussed in the present work.
General expressions for diffuse XRMS are tedious due to the complex form of the resonant magnetic scattering amplitude (1.39). Only expressions are given for special geometries here: First, the diffuse intensity difference in L-MOKE geometry using circular polarization is considered as it will be used in Chap. 9. Subsequently, the diffuse scattering by AF-coupled superlattices and AF domains is discussed (Chap. 7).

Osgood et al. \[104\] have developed a model for diffuse XRMS by a magnetic surface in BA. This model has been generalized by Nelson et al. \[105\] to the case of magnetic multilayers. The derivation is completely analogous to the model of conventional diffuse x-ray scattering outlined in the previous section. First, a single magnetic surface is considered. The resonant magnetic scattering amplitude (1.39) is inserted into Eq. (4.1). If terms corresponding to linear dichroism are neglected, the differential cross section is

\[
\frac{d\sigma}{d\Omega} = \left| \left( N_e(-Zr_0) + N_r \frac{3\lambda}{8\pi} [F_1^i + F_{-1}^i] \right) \bar{\epsilon}_f \cdot \bar{\epsilon}_i \int_{V_e} d^3r \exp (-i\mathbf{q} \cdot \mathbf{r}) + iN_r \frac{3\lambda}{8\pi} [F_{-1}^i - F_1^i] (\bar{\epsilon}_f^* \times \bar{\epsilon}_i) \cdot \mathbf{m} \int_{V_m} d^3r \exp (-i\mathbf{q} \cdot \mathbf{r}) \right|^2,
\]

where \( N_e \) and \( N_r \) are the number densities of all atoms and the resonant atoms, respectively. It is assumed that the sample is composed of two parts: a charge \( (V_e) \) and magnetic volume \( (V_m) \), which overlap considerably but which are in general separate entities due to the presence of magnetic dead layers, etc. These two volumes will have different roughnesses in general, so that the integral of \( \exp(-i\mathbf{q} \cdot \mathbf{r}) \) over the charge and magnetic volumes will be different. When multiplied out, Eq. (4.10) can be grouped into three terms: a pure charge term, a pure magnetic term, and a charge-magnetic interference term. The only term which is sensitive to a reversal of photon helicity or magnetic field, is the interference term

\[
\left( \frac{d\sigma}{d\Omega} \right)_{\text{int}} = \left\{ \left( N_e(-Zr_0) + N_r \frac{3\lambda}{8\pi} [F_1^i + F_{-1}^i] \right) (\bar{\epsilon}_f^* \times \bar{\epsilon}_i) \right. \times \left( \bar{\epsilon}_f^* \times \bar{\epsilon}_i \right) \cdot \mathbf{m} + \text{c.c.} \left\} \int_{V_e} \int_{V_m} d^3r d^3r' \exp (-i\mathbf{q} \cdot (\mathbf{r} - \mathbf{r}')),
\]

where c.c. is the complex conjugate of the previous term. To evaluate the integral in Eq. (4.11), again the approach of Sinha et. al \[98\] is applied: The volume integrals are transformed into surface integrals. The heights of the structural and magnetic surfaces above the \( xy \)-plane are given by the functions \( z_e(x, y) \) and \( z_m(x, y) \), respectively. Both, the structural and magnetic surfaces are assumed to show self-affine, fractal behavior and \( z_e(x, y) - z_m(x', y') \) to be a Gaussian random variable. The further derivation is completely analogous to that in Sec. 4.2 and only the generalized result for the difference in diffuse intensity of left and right circularly polarized light by a magnetic multilayer is stated here:

\[
\Delta \left( \frac{d\sigma}{d\Omega} \right)_{\text{diffuse}} = P_c \frac{L_x L_y}{q_z^2} \left[ \mathbf{k}_f \cdot \mathbf{m} + \cos(\alpha + \beta) \mathbf{k}_i \cdot \mathbf{m} \right] \times
\]

\[
\sum_{i,j}^N \Delta \rho_{ij} e^{i\mathbf{q}_z (z_i - z_j)} e^{-q_z^2 (\sigma_{z,i}^2 + \sigma_{z,j}^2)/2} S_{ij}^{\text{em}} (\mathbf{q}) + \Delta \rho_{ij} e^{i\mathbf{q}_z (z_i - z_j)} e^{-q_z^2 (\sigma_{z,i}^2 + \sigma_{z,j}^2)/2} S_{ij}^{\text{em}} (\mathbf{q})
\]

(4.13)
4. Diffuse scattering

with

\[ S^{em}_{ij}(\vec{q}) = \int_S dX dY \left( e^{i\vec{q} \cdot \vec{Z} + q_x X + q_y Y} - 1 \right) e^{i(q_x X + q_y Y)}, \]  

(4.14)

where \( C^{em}_{ij}(X, Y) \) is the height-height correlation function of the structural interface \( i \) and the magnetic interface \( j \). The function is identical to Eq. (4.6), but the correlation length \( \xi \) and the hurst parameter \( h \) should be replaced by \( \xi^{em} \) and \( h^{em} \), characterizing the charge-magnetic correlation. The factor in front of the sum in Eq. (4.12) contains the amount of circular polarization \( P_c \) and the geometrical dependence on the angles of incidence and reflection \( \alpha_i \) and \( \alpha_f \). From this geometrical factor it can be seen that only magnetic moments in the scattering plane contribute to the diffuse intensity difference. Due to the scalar product \( \vec{k}_i, f \cdot \vec{m} \) out-of-plane magnetic moments contribute only for large scattering angles. The function

\[ \Delta \rho^{em}_{ij} = \Delta \left\{ N_c(-Zr_0) + N_r \frac{3\lambda}{8\pi} \left[ F_{1-1}^1 + F_1^1 \right]^\ast \right\}_i \Delta \left\{ N_r \frac{3\lambda}{8\pi} \left[ F_{1-1}^1 - F_1^1 \right] \right\}_j \]  

(4.15)
displays the difference in the charge scattering amplitude across the \( i \)th interface times the difference in the magnetic scattering across the \( j \)th interface.

From Eq. (4.12), it is obvious that the diffuse scattering difference \( \Delta I \) between right- and left circularly polarized light is a term that depends on both the structural and magnetic interfaces. If the structural and magnetic interfaces are completely uncorrelated (i.e., \( C^{em}_{ij} = 0 \)) \( S^{em}_{ij}(\vec{q}) \) vanishes, and \( \Delta I \) does not contain a diffuse term to leading order in the magnetization.

In AF-coupled superlattices a peak of purely magnetic origin can be observed at half the \( q_z \) position of the structural peak. The magnetic intensity is mainly caused by scattering second-order in the magnetization, i.e. \( \sigma \to \pi, \pi \to \sigma \) and \( \pi \to \pi \) scattering. The first two channels are sensitive to the magnetization direction in the scattering plane, while the \( \pi \to \pi \) channel is sensitive to the perpendicular component. Since the scattering is of purely magnetic origin, the magnetic roughness has not necessarily to be correlated to structural roughness and the real magnetic correlation length \( \xi_m \) and hurst parameter \( h_m \) can be determined from a fit to the experiment instead of the charge-magnetic parameters. Osgood et al. [104] have given explicit expressions for these differential cross sections of a single surface, as well. However, the line shape of the intensity curve as a function of \( q_x \) is identical to that in Eq. (4.4) and the differential cross sections are not stated here.

A model for off-specular x-ray magnetic scattering by domains has been developed in Refs. [95, 104]. Similar models exist for neutron scattering [106]. There are two cases to consider: If the typical lateral domain size is larger than the lateral coherence length of the x-rays, the scattered intensity is the incoherent sum of the scattering from the magnetized regions. If there is no net magnetization, the contributions to the interference term will cancel out leading to \( \Delta I = 0 \). Second-order contributions to the off-specular scattering will hardly be seen, since the length scale involved will be too large to be resolved in \( q_x \) from the specular beam. If, on the other hand, the lateral size of the domains is smaller than the lateral coherence length, domain scattering will manifest itself in the off-specular scattering. In analogy to the height-height correlation function \( C(X, Y) \) a domain correlation function

\[ \gamma_d(X, Y) = < p(0, 0) p(X, Y) > \]  

(4.16)
4.4. Distorted-wave Born approximation

is introduced, where \( p(x, y) = \pm 1 \) is the probability that the domain at position \((x, y)\) is aligned parallel (antiparallel) to the average magnetization direction, which is assumed to be uncorrelated with structural features at the interface. A model for random domains and sharp walls, \( \gamma_d(R) = \exp(-R/a) \), where \( a \) is the average domain size, yields \[95\]

\[
S^{mm}(\vec{q}) = \int dX dY \gamma_d(X, Y) e^{-i(q_x X + q_y Y)} = \frac{a^2}{\left[1 + (q_x^2 + q_y^2)a^2\right]^{3/2}}, \tag{4.17}
\]

where \( mm \) indicates purely magnetic scattering. If the \( q \) component perpendicular to the scattering plane is integrated out, (4.17) has a Lorentzian line shape.

### 4.4. Distorted-wave Born approximation

The BA breaks down for small scattering angles, where total external or multiple internal reflections occur. In this case the distorted-wave Born approximation (DWBA) has to be applied. Within this approach the illuminating amplitudes in each layer and their respective interferences are calculated dynamically, as obtained e.g. by the Parratt formalism \[87\] or the Zak formalism \[59\] presented in the previous chapter for ideally flat interfaces. On the other hand, the diffuse scattering within each layer is still calculated kinematically as in the BA. The differential cross section for scattering by a rough surface in DWBA has been firstly calculated by Sinha et al. \[98\]:

\[
\left(\frac{d\sigma}{d\Omega}\right)_{\text{diffuse}} = \frac{L_x L_y}{16\pi^2} k_0^2 1 - n^2)^2 \left| T(\alpha_i) \right|^2 \left| T(\alpha_f) \right|^2 S(\vec{q}_t), \tag{4.18}
\]

where \( T(\alpha_{i,f}) \) are the transmission coefficients of the surface for angles \( \alpha_{i,f} \) and

\[
S(\vec{q}_t) = \frac{1}{q_x^2 q_y^2} e^{\sigma^2(q_x^2 + q_y^2)/2} \int_S dX dY \left( e^{i\vec{q}_t \cdot \vec{C}(X,Y)} - 1 \right) e^{-i(q_x X + q_y Y)}. \tag{4.19}
\]

The wave vector transfer inside the reflecting medium is given by \( \vec{q}_t = n \vec{q} \). While the in-plane components of the scattering vector are constant during transmission, the \( z \) component even might be purely imaginary for incident angles below the critical angle. The height-height correlation function \( \vec{C}(X,Y) \) is the same as introduced previously.

The generalization of the DWBA formalism for multilayer systems rapidly becomes very complicated \[99, 101, 107, 108\]. An interface for these calculations is available via internet \[109\], which has been used to analyze the small-angle hard x-ray diffuse scattering intensities in Chaps. \[7\] and \[9\]. General expressions for x-ray resonant magnetic diffuse scattering from multilayers within DWBA were recently derived by Lee et al. \[95\]. However, due to the complexity of the resulting expressions, a discussion of both, the non-resonant and resonant magnetic diffuse scattering would go beyond the scope of this thesis.
4. Diffuse scattering
Part II.

Instrumentation for soft x-ray resonant magnetic scattering
5. Synchrotron radiation

5.1. Introduction

The development of magneto-optical x-ray measurement techniques like x-ray magnetic circular dichroism (XMCD) or x-ray resonant magnetic scattering (XRMS) would have been impossible without the availability of high-brilliance synchrotron sources. Only then it was possible to overcome the drawbacks of standard x-ray tubes such as the relatively low radiation power and - even more important for resonant scattering - the missing tunability of the photon energy. Originally synchrotron radiation was observed as parasitic effect in circular particle accelerators dedicated to nuclear and elementary particle physics. Nowadays electron and positron storage rings are built especially for the creation of synchrotron radiation at dipole bending magnets, which are necessary to keep the particles in a closed orbit, and at insertion devices such as wigglers or undulators. With free-electron lasers (FEL) completely coherent x-ray sources might be available in future with a brilliance many orders of magnitude higher than the radiation from third-generation storage rings. In this chapter the basic properties of synchrotron radiation from bending magnets and insertion devices are briefly illustrated. Subsequently the experimental conditions at bending magnet beamline PM3 and undulator beamlines UE56/1 and UE56/2 at BESSY II (Berliner Elektronenspeicherring-Gesellschaft für Synchrotron Strahlung m.b.H.) are discussed.

5.2. Creation of synchrotron radiation

Synchrotron radiation is emitted by charges accelerated transverse to their propagation direction. In the constant field $B$ of a dipole bending magnet electrons or positrons are moving on a circular arc with radius $r = m_e v c / e B$ and angular frequency $\omega_0 = v / r$. For slowly moving charges, i.e. in the electron rest frame, the angular distribution of the electromagnetic radiation is given by the dipole characteristics. At relativistic electron velocities $v \approx c$ the emitted intensity is concentrated to a small cone of opening angle $1 / \gamma$ due to the Lorentz transformation, where $\gamma = E_e / m_e c^2$ is the electron energy in units of its rest mass energy $m_e c^2 = 511$ keV. The radiation patterns for $\gamma = 1$ and $\gamma = 2$ are depicted in Fig. 5.1. Realistic values of $\gamma$ are in the order of $10^3$ to $10^4$ for GeV electron energies present at modern synchrotron storage rings. The radiation cone is seen as a sweeping search light with maximum intensity when the observer is directly facing into the oncoming electron. The radiation frequency depends on the state of motion of the observer due to the Doppler effect. In the electron rest frame the energy...
Figure 5.1.: Radiation pattern for charges accelerated transverse to the direction of motion. The relativistic pattern \( (\gamma = 2) \) is scaled by a factor of \( 10^{-2} \) for the same acceleration \( \ddot{v} \).

of the emitted radiation \( \hbar \omega_0 \) is typically in the neV range. In the laboratory frame the wavelengths are shifted to the x-ray range.

A strict derivation of the radiated intensity per unit frequency interval per unit solid angle for an electron moving on a circular arc yields [110]

\[
\frac{d^2 I}{d\omega d\Omega} = \frac{3e^2}{4\pi^2 c} \left( \frac{\omega}{\omega_c} \right)^2 \gamma^2 \left( 1 + \frac{\gamma^2}{\Theta^2} \right)^2 \left[ K_{2/3}^2(\xi) + \frac{\gamma^2 \Theta^2}{1 + \gamma^2 \Theta^2} K_{1/3}^2(\xi) \right],
\]

with

\[
x = \frac{\omega}{2\omega_c} \left( 1 + \frac{\Theta^2}{\gamma^2} \right)^{3/2},
\]

where \( \Theta \) is the emission angle out of the plane of the electron orbit, \( \omega_c = 3\gamma^3 \omega_0/2 \) is a characteristic frequency and \( K_{1/3}^2(\xi) \) and \( K_{2/3}^2(\xi) \) are modified Bessel functions. For \( \xi \gg 1 \) the radiated intensity is negligible. This will occur at frequencies larger than \( \omega_c \) and at large angles showing again that the radiation is mostly confined to the synchrotron plane. For frequencies \( \omega \ll \omega_c \) the intensity spectrum is proportional to \( \omega^{2/3} \) while for \( \omega \gg \omega_c \) the intensity decays exponentially. In practical units the characteristic photon energy is given by

\[
E_c[eV] = \hbar \omega_c = 665E_c^2[GeV]B[T].
\]

Fig. [5.2(a)] shows the intensity spectrum of a bending magnet at BESSY II integrated over \( \Theta \). The electron energy is 1.7 GeV. The magnetic field of 1.3 T forces the electrons to a circular arc of \( r = 4.35 \) m radius.

The first term in Eq. (5.1) corresponds to a polarization parallel to the orbital plane, and the second to perpendicular polarization. Since the second term vanishes for \( \Theta = 0 \), the polarization is fully linear in the synchrotron plane and becomes more and more circular above and below as shown in Fig. [5.2(b)]. This polarization dependence can be utilized to perform experiments with circular polarization even at bending magnet beamlines. However, a high degree of circular polarization is only achievable at the expense of intensity.

5.3. Insertion devices

X-ray beams of much higher brightness can be produced by so-called insertion devices installed in the straight sections of a storage ring between the dipole bending magnets.
5.3. Insertion devices

An insertion device consists of a periodic array of magnets with the field direction alternately up and down perpendicular to the electron propagation direction, forcing the electron to oscillations as it traverses through the section. Insertion devices can be divided into two types, wigglers and undulators, depending on the parameter

\[ K = \frac{e\lambda_u B_0}{2\pi m_e c} = 0.934\lambda_u [\text{cm}] B_0 [\text{T}], \]  

(5.4)

where \( \lambda_u \) is the magnetic period and \( B_0 \) the field of the insertion device. \( K \) is a measure of the deflection of the direction of electron motion away from the forward direction. The maximum angular deviation is given by \( \delta = K/\gamma \).

For \( K \gg 1 \) (typically \( K = 20 \)) the insertion device is called a wiggler. It can be viewed as a series of circular arcs, turning successively to the left and to the right. Since the opening angle of the radiation for a single circular arc is of the order \( \gamma^{-1} \ll \delta \), an observer detects a series of incoherent flicks of the searchlight beam. Therefore the spectrum generated by a wiggler is the same as that from a bending magnet of the same field strength. The intensity is increased by a factor \( 2N \), where \( N \) is the number of periods of the wiggler.

In case of an undulator with \( K \leq 1 \) the searchlight beam of radiation moves negligible compared to its own angular width. In consequence the radiation detected by an observer is an almost coherent superposition of the contributions from all oscillations. Therefore the intensity from an undulator scales with \( N^2 \) leading to a large increase in brightness compared to that of a bending magnet or wiggler. Due to the requirement of constructive interference the intensity spectrum is sharply peaked at a fundamental wavelength depending on the spatial undulator period \( \lambda_u \). This wavelength and its

Figure 5.2.: Energy spectrum and polarization of synchrotron radiation from a bending magnet at BESSY II.

(a) Energy spectrum of the bending magnet. The energy is given both, in eV and in units of the critical energy.

(b) Degree of circular polarization as a function of the emission angle \( \Theta \) for different energies.
higher harmonics are given by

\[ \lambda_n = \frac{\lambda_u}{2n\gamma^2} \left(1 + \frac{K^2}{2} + \gamma^2\Theta^2\right), \]  

(5.5)

where \( n \) is an odd integer number. In the electron rest frame the electron performs a harmonic, sinusoidal motion. But it is not harmonic seen from the on-axis observer. Due to the Doppler shift the sinusoidal curve is distorted towards a triangular shape, which can be resolved into Fourier components leading to the higher harmonics. The distortion and therewith the intensity of the higher harmonics increases with \( K \). The spectral intensity distribution of the first, third and fifth harmonic is shown in Fig. 5.3 with the maximum intensity normalized to unity. Experimentally a broad energy range is accessible, because the parameter \( K \) can be tuned by changing the vertical gap between the magnetic poles and therewith the magnetic field \( B_0 \) in Eq. (5.4). Another feature of the radiation from an undulator is that the intrinsic angular divergence with a width of approximately \( 1/(\sqrt{N}\gamma) \) is much smaller than that of a bending magnet or wiggler.

So far only linear undulators have been discussed with an arrangement of magnetic fields as shown in Fig. 5.4(a). The radiation is linearly polarized in the synchrotron plane. In a helical undulator the magnet rows above and below the synchrotron plane can be horizontally shifted relative to each other as shown in Fig. 5.4(b). For a shift of \( \lambda_u/4 \) a helical magnetic field is generated. The electron trajectory describes a spiral leading to circular polarization. By further increasing the shift to \( \lambda_u/2 \) the electrons oscillate in the vertical direction and correspondingly perpendicular linear polarization is produced.
5.4. Measurements at BESSY II

BESSY II is a third-generation synchrotron radiation facility starting its operation in 1998, which is especially dedicated to vacuum ultra violet (VUV) and soft x-ray radiation. The measurements presented in the present work have been carried out at the bending magnet beamline PM3 (partly Chap. 7 and Chap. 8) and at undulator beamlines UE56/1-PGM and UE56/2-PGM2 (Chaps. 7 and 9).

According to Eq. (5.3) the critical energy of the bending magnets at BESSY II is 2.5 keV leading to the intensity spectrum shown in Fig. 5.2(a). At beamline PM3, an energy range of 20 eV to 1.2 keV is accessible with a maximum photon flux between 500 eV and 900 eV, approximately, ideally suited for measurements at the $L_{2,3}$ absorption edges of 3$d$ transition metals. By means of a vertical aperture it is possible to select linear or circular polarization up to $P_c = \pm 95\%$, respectively. The monochromator is a plane grating mirror with an energy resolution of approximately $\Delta E/E = 1 \cdot 10^{-4}$ at $E = 800$ eV, depending on the exact setting of the exit slit. The position of the horizontal focus is 500 mm behind the exit slit. Vertically the beam is focused to the exit slit. Therefore the spot size of the beam is roughly $0.5 \times 1$ mm$^2$ (horizontal $\times$ vertical). This configuration enables an efficient use of the incoming intensity with the ALICE diffractometer, which is described in detail in the next chapter: Since the scattering plane is horizontal, vertical entrance and detector slits define the angular resolution, while the intensity is integrated in the vertical direction. The direct beam causes a current of typically 50 nA in the photodiode used as detector. Since the dark current of the photodiode is less than 0.01 pA, reflectivity measurements over five orders of magnitude are possible, which is sufficient for most multilayer systems. A strong advantage of beamline PM3 is that the x-ray beam is available 24 hours/day and has not to be shared with other users.

The UE56/1 undulator ($\lambda_u = 56$ mm) is an elliptically polarizing insertion device. In the photon energy range between 500 eV and 900 eV the third harmonic of the undulator spectrum is used. The maximum degree of circular polarization is approximately 90% at a photon energy of 700 eV. The beamline has a plane grating monochromator (PGM). The 1200 mm$^{-1}$ grid has been used for the measurements described in the present thesis. An exit slit width of typically 100 $\mu$m provides an energy resolution of $E/\Delta E = 4000$ at 700 eV. In an energy scan the monochromator and the undulator (gap and shift)
are driven simultaneously in order to receive the maximum flux at each photon energy (c.f. Fig. 5.3). Depending on the slit settings the photo diode current in the direct beam is several µA, i.e. at least two orders of magnitude larger than at the bending magnet beamline. At beamline UE56/2-PGM2 the x-ray radiation is created by two undulators (upstream and downstream) with the possibility to either rapidly switch between the beams (e.g. switching between different helicities in an XMCD experiment) or add the intensities of both undulators. Apart from that the design of beamlines UE56/1-PGM and UE56/2-PGM2 is identical. For more details see Ref. [113].
6. The ALICE diffractometer

6.1. Introduction

In order to carry out soft XRMS experiments with the possibility of low-temperature measurements in the presence of a variable magnetic field, a new vacuum-compatible diffractometer has been designed and constructed within the framework of a project funded by the BMBF (Bundesministerium für Bildung und Forschung) starting in April 2001. Several instruments for soft x-ray resonant magnetic scattering were available at that time at different synchrotron facilities worldwide [114, 115, 116, 117, 118]. However, these instruments are limited either in temperature, field or angular range and for most of them it is not possible to combine all these features at the same time. The ALICE diffractometer covers a temperature range of 30 K - 380 K. Simultaneously a variable magnetic field of ±2.7 kOe can be applied. This large parameter range is unique at least in Germany. XRMS is still a rapidly evolving scientific field, and in the meantime other new instruments with an extended range of parameters dedicated to soft XRMS have started operation [119]. After one and a half year of construction, the ALICE diffractometer was successfully tested at beamline PM3 at BESSY II in December 2002. In 2003/2004 the instrument was installed at different beamlines during 20 weeks of beamtime in total. Ten weeks of beamtime are approved in 2005 and the funding period has been extended for three additional years by the BMBF. The instrument was planned to be open to other users from the very beginning. Groups from Durham/England, Uppsala/Sweden, Potsdam, Berlin and Jülich have used the ALICE diffractometer for soft x-ray measurements. A variety of different physical systems has been investigated. Two of them are presented in Part III. A complete list of publications resulting from measurements with the ALICE diffractometer can be found at the end of the thesis.

In this chapter the new diffractometer ALICE for soft x-ray resonant magnetic scattering is described, whose design and construction were the main work content underlying the present thesis

6.2. The diffractometer

Due to the strong air absorption of x-rays in the considered energy range (500 eV - 1500 eV), XRMS experiments have to be carried out under vacuum conditions. Therefore

1For the origin of the name ALICE see Ref. [120].
2This chapter is based on the article Diffractometer for soft x-ray resonant magnetic scattering, see Ref. [121]. Some technical details and recent modifications of the experimental setup have been added.
6. The ALICE diffractometer

![Schematic diagram of the diffractometer](image)

Figure 6.1.: Schematic topview of the diffractometer. For details see the main text.

The two-circle goniometer is installed in a cylindrical vacuum chamber of 400 mm inner diameter and 310 mm height with the cylinder axis being vertical. The chamber is pumped by a 260 l/s turbo-molecular pump, backed by a scroll-type dry roughing pump. The sample can be changed through a 200 mm-diameter load-lock window. Although the chamber is UHV-compatible, high vacuum conditions \( < 1 \times 10^{-6} \text{ mbar} \) are sufficient \[114] for the measurements carried out within this study and therefore the pumping time after sample change is below 30 minutes.

At synchrotron sources like BESSY II, an oil-free vacuum of typically \( < 5 \times 10^{-9} \text{ mbar} \) is required at the last valve of the beamline to ensure the ultrahigh vacuum environment of the beamline optics and the storage ring. To fulfill this condition a differential pumping stage consisting of a pinhole P1 (1.5 mm diameter) and a turbo-molecular pump between the chamber and the beamline was installed (Fig. 6.1). For vacuum analysis a quadrupole mass spectrometer is included in the setup.

The sample and the detector are driven by two differentially pumped 100 mm-bore rotating platforms (RP) with stepper motors and gears external to the vacuum. During the assembly of the setup the rotation axes of the RPs for detector (2Θ) and sample (ω) have been checked to have an offset and a tilt of less than 0.05 mm and 0.05°, respectively. The angular resolution for sample and detector rotation is 0.005°. The rotation range is ±180° for the sample and approximately −170° to +145° for the detector.

Both RPs are concentrically mounted on top of the chamber so that the scattering geometry is horizontal. For linearly polarized light with polarization vector in the orbit plane of the synchrotron this arrangement corresponds to a transversal Kerr effect (T-MOKE) geometry sensitive to magnetic moments lying parallel to the sample surface and perpendicular to the plane of incidence. Using circularly polarized light the longitudinal Kerr effect (L-MOKE) geometry can be realized, where the magnetic moments are both
6.2. The diffractometer

The scattered beam is detected by a Hamamatsu G1127-04 GaAsP photodiode with a dark current below \(10 \text{ fA}\). The glass window protecting the diode surface has to be removed for soft x-ray measurements. Typical photocurrents are 100 nA-10 µA in the direct beam, depending on whether the radiation is produced by a bending magnet or insertion device and depending on the slit widths defining the energy and angular resolution. The signal might decrease to \(< 10 \text{ fA}\) at high scattering angles limited by the dark current of the photodiode. The corresponding photocurrent is measured by a Keithley 6517A or Keithley 6514 electrometer. Depending on the application both instruments have their individual advantages: While the 6517A has a lower noise level and low-current measurements down to 1 fA are possible, the Keithley 6514 is much faster - with a good accuracy for currents down to 50 fA. Additionally the Keithley 6517A electrometer contains a programmable, bipolar 1000 V voltage supply.

In order to measure such small currents the electrical circuit must be perfectly shielded against externally induced noise. Outside the vacuum a triax cable is used. The outermost conductor is grounded. The inner coaxial wires carrying the diode signal are connected to a floating shield BNC vacuum feedthrough without electrical contact to the chamber. In vacuum the diode is connected to a Kapton-insulated coaxial cable. The grounded vacuum chamber serves as additional shielding. To avoid ground loops the chamber and the grounded triax conductor are not connected. For low-level current measurements it is imperative to minimize leakage and noise currents \[122\]. Leakage currents can be eliminated through guarding. Basically, guarding uses a conductor (the inner shield of the triax cable) at the same potential as the sensitive current path to totally surround the input leads carrying the signal. Noise currents may arise from triboelectric or piezoelectric effects: Triboelectric currents are generated by charges created at the interface between a conductor and an insulator due to friction. Piezoelectric currents are generated when mechanical stress is applied to certain insulating materials. These effects are minimized by using special low-noise triax cables. Another source of noise currents is dielectric absorption that occurs when a voltage applied across an insulator causes positive and negative charges within that insulator to polarize. When the voltage is removed, the separated charges generate a decaying current through external circuitry as they recombine. The dissipation of these currents may take even hours. For this reason all cables should be connected several hours before low-current measurements.

It is possible to measure x-ray absorption and XMCD spectra by means of the total electron yield (TEY) method (see Chap. \[2\]). For this purpose the sample is mounted electrically insulated on the sample holder. The wiring is almost identical to that of the photodiode described above. Only the inner wire of the coaxial cable is connected to the sample. The shielding is kept around the wire as close to the sample as possible. A bias voltage of 50-100 V is applied using the Keithley 6517A voltage supply or a series of commercial 9 V batteries.

There are two linear feedthroughs F1 and F2 installed in the chamber in front of the sample as indicated in Fig. 6.1. By means of these feedthroughs several components can

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\[3\] In the past the dark current increased to the pA range after several weeks of beamtime necessitating a replacement of the diode. The origin of this increase is not clear at the moment.
be moved into the position of the direct beam, which are successively described below.

Thin metallic Fe and Ni foils are installed at linear feedthrough F2. For energy calibration the transmission spectrum of these foils is measured. Two slits S1 and S2 are installed in the vacuum chamber in order to define the angular resolution. S1 is installed on linear feedthrough F2 approximately 100 mm before the sample. S2 is installed in front of the detector. Both slits are variable and were set to 300 µm during most of the experiments. With these settings an angular resolution of 0.13° is obtained.

The intensity $I_0$ of the incident beam can be monitored by means of the photocurrent of a gold mesh, which can be installed on feedthrough F1. Nevertheless, several aspects have to be considered regarding the $I_0$ measurement: The most simple way to measure $I_0$ is to monitor the storage ring current. This method is sufficient as long as experiments are performed at a fixed energy. Otherwise the energy-dependence of the emitted intensity and the efficiency of optical elements have to be taken into account (see e.g. Fig. 5.2(a)). Therefore an $I_0$ monitor behind all optical elements and slits is desirable as it would be realized by the gold mesh on F1. However, in order to measure the incoming intensity accurately, the number of Au wires per unit area has to be large compared to the beam spot size. The mesh position in the chamber is close to the focus of the x-ray beam, which is optimally at the sample position. Correspondingly the necessary wire density is large and a huge part of the intensity is absorbed. In our experiments the photoelectric current of the refocusing mirror in the beamline optics has been used as $I_0$ monitor. In this way no intensity is lost. The current might be energy-dependent due to a surface contamination of the mirror, which is a problem especially at energies around the carbon K absorption edges (284 eV). Furthermore the slit S1 is still behind the $I_0$ monitor, which leads to errors if the beam position is slightly changing in the experiment as discussed below.

The sample can be linearly moved by a manipulator consisting of a XY table (5 µm resolution) and a linear Z translator in all directions relative to the beam. It is mounted on a cradle and can be manually tilted with two screws. The sample environment consists of a He closed-cycle cryostat with the possibility of heating (temperature range of 30-380 K) and a rotatable electromagnet from the company AMACC, Sweden. The cryostat is controlled by a Lakeshore 340 temperature controller. For temperatures higher than 320 K, the cold head and the heater can be thermally decoupled by evacuating an intermediate cavity, which is filled with 800 mbar of He for low temperature measurements. In order to construct a suitable magnet for XRMS experiments several aspects had to be considered: The magnetic field should be applied in the scattering plane along the sample surface (L-MOKE geometry). In order to perform low-temperature measurements the magnet must be thermally and therewith mechanically decoupled from the sample. Therefore it has to be independently rotatable in angle-dependent reflectivity measurements. The maximum field should be at least 0.1 T, for exchange-bias systems larger fields are desirable (for a review of exchange bias phenomena see e.g. Ref. [123]). On the one hand, for the study of element-specific remagnetization processes a tuneable electromagnet is necessary. On the other hand, it should be possible to permanently apply large magnetic fields, e.g. if an exchange-bias system is field cooled. As a consequence there must be a possibility to carry off the heat load. A water-cooled electromagnet is difficult to realize in vacuum. Therefore an alternative approach was chosen: The magnet coils on a horseshoe yoke are placed in a cylindrical tube outside of the vacuum.
6.2. The diffractometer

Figure 6.2.: View into the vacuum chamber through the loadlock window. The synchrotron beam coming from the left-hand side is reflected from the sample, which is located between the two magnetic poles. The detector housing on the right-hand side protects the photodiode from parasitic scattering.

A water-cooled copper plate is installed below the coils. The tube is mounted on a 150 mm ID flange at the bottom side of the chamber with the cylinder axis parallel to the diffractometer axes. The yoke is magnetically coupled through a thin stainless steel plate to a set of pole pieces in vacuum. The slitted poles and the cylindrical tube below can be seen in Fig. 6.2.

Originally the idea behind the rotation of the magnet was as follows: The poles should be rotated on a low-friction ball bearing denoted by the dotted circle in Fig. 6.1. When the horseshoe yoke below was rotated the poles on top should follow the motion due to the magnetic coupling. This principle was successfully tested out of the vacuum. However, it turned out that the friction is dramatically increased in the vacuum. Several attempts to improve the mechanical stability of the system were without success. Finally another differentially pumped RP was installed for the rotation of the complete cylindrical tube.

The magnet has been calibrated ex-situ before the experiments. With two sets of poles, which are different in gap size $G$, a maximum field of approximately 0.1 T ($G = 40$ mm) and 0.27 T ($G = 15$ mm, shown in Fig. 6.2) can be reached, respectively. For measurements in transmission geometry another set of poles with $G = 5$ mm has been manufactured. Due to the very small gap size the maximum field is approximately 0.8 T. The pole pieces are made of iron and nickel-plated by an electro-chemical procedure \cite{124} in order to prevent oxidation.
Stepper motors, temperature and magnetic field are controlled via GPIB interface from a Linux PC. The bipolar power supply (±35 V, ±5 A) for the magnetic field is controlled by a Delta Electronika PSC44M interface, which converts the digital GPIB signal to two analog voltages in a range of 0-10 V.

Data acquisition is performed running standard SPEC software. The software had to be modified in several respects in order to come up to the specialties of the diffractometer. First, the rotation of sample and detector are not independent. This problem was overcome by introducing the standard motors \( \text{th} \) and \( \text{tth} \) as virtual motors leading to a combined motion of both RPs. Second, the measurement of the diode current is very different from photon counting in standard hard x-ray scattering. In addition to the statistical errors, systematic drifts in the detector signal have to be considered. The diode current is measured \( N \) times at each point in a scan. For high intensities \( N = 10 \) is sufficient while at high scattering angles \( N = 500 \) might be necessary. Depending on the Keithley settings each measurement takes 0.01 s - 0.1 s. To visualize a possible drift of the detector signal all measurements are plotted in a separate window versus time. The window is updated for each point in a scan. A typical systematic error is an exponential-like approaching to the "real" value. In that case a simple averaging over all \( N \) points would yield the wrong result. In the software it is now possible to define a number of first points, which are ignored in the averaging procedure in order to yield the correct value.

A photograph of the diffractometer is shown in Fig. 6.3.

6.3. Alignment

The instrument was designed to be operational at different beamlines or even at different synchrotron facilities like BESSY II or the Swiss Light Source (SLS). The setup is usually located at the Institute of Solid State Physics at the Ruhr-University Bochum for maintenance and improvements. Hence transportability, flexibility and a quick alignment procedure were important considerations for the construction.

The goniometer chamber and the differential pumping stage are mounted on two independently adjustable frames similar to the six-strut alignment systems commonly used e.g. at the Advanced Light Source (ALS) in Berkeley. These frames are fixed to a rack each by seven struts - four vertical and three horizontal - with ball jointed end connections. By turning a strut, the distance between the two ball joints can be varied. That way it is possible to move the frames in all six degrees of freedom. Each strut has a travel of 50 mm. The struts can be mounted in different position to the bottom rack, so that the instrument is suited for a beam height from approximately 1200 mm to 1600 mm above the floor without the necessity of any additional platform.

The alignment procedure in the reflection geometry is as follows: First, the beam is checked to pass the pinhole P1 and hit the middle of the vacuum window coated with fluorescent pigments (Fig. 6.1). After the vertical struts V1 and V2 have been rotated until the beam strikes the center of the photodiode the other vertical struts are adjusted with a precision spirit level to ensure that the rotation axis is exactly vertical. Next the rotation axis is checked to lie in the direct beam. The sample is positioned to cut half of the direct beam by means of the XY table. A quick rocking scan ensures that
the sample surface is exactly parallel to the beam direction. The corresponding sample position \( y_1 \) can be read from the micrometer screw. If the rotation axis is exactly in the middle of the direct beam, again half of the intensity should be measured after rotating the sample by 180°, because the sample cuts half of the direct beam in both positions. Otherwise the new half-intensity position \( y_2 \) is determined. Now the complete chamber is moved by means of the horizontal struts H1 and H2 by a distance \( (y_1 - y_2)/2 \). Since the detector is moved either in this way, it has to be realigned. The alignment can be checked again after the sample is rotated back to \( \Theta = 180^\circ \). Usually only one iteration step is necessary to align the rotation axes within an accuracy of < 0.05 mm, which is sufficient for reflectivity measurements.

After the rotation axes are aligned relative to the center of the direct beam once, the zero positions of the angular scales of the sample and detector rotation are calibrated. The direct beam might change its position during an experiment due to a slightly different electron trajectory in the storage ring after injection or due to heating of the optical elements. The heat load of the optical elements might vary in time, because it depends on the intensity of the incoming beam and therefore on the ring current. This deviation from the original position can be measured by a detector scan. The shift in mm is
calculated from the angular deviation and is simply corrected by moving the complete diffractometer with the horizontal struts H1 and H2. Therefore a renewed alignment is very precise and can be done within a few minutes.

For the alignment of the sample tilt there is a laser system consisting of a pentaprism and another pinhole mounted on two linear feedthroughs. The pentaprism in front of the pinhole can reflect a laser beam along the path of the x-ray beam. This provides a method of aligning the sample tilt angle with two screws by hand before the vacuum system is pumped [116].

6.4. Instrumental outlook

The project was originally funded for a period of three years. After a positive evaluation of the project three more years of funding were granted by the BMBF. Therefore it is possible to improve the experimental setup. Experiences of the first measurements and new ideas can be realized. Below a list of planned and desirable technical improvements is given:

- Since circular light is sensitive to the magnetization component in the scattering plane (L-MOKE) and linearly $\pi$ polarized light to the perpendicular component (T-MOKE), vector-magnetometry measurements are possible with the existing experimental setup in principle. An example is given in Chap. [7]. However, a quantitative comparison of the longitudinal and transverse MOKE is difficult, since both effects differently depend on the angle of incidence. Therefore it is preferable to apply the magnetic field in the longitudinal and transverse direction and perform measurements with a fixed polarization instead. At the moment it is not possible to apply a transverse field. A suitable magnet has to be constructed.

- For the study of systems revealing magneto-crystalline anisotropy it is planned to install the possibility to rotate the sample around the sample normal. So far it is not possible to change the sample tilt and the slit widths in-situ as well. This problem will be solved by installing vacuum-compatible stepper motors.

- The lowest-possible temperature with closed-cycle He cryostat is 30 K. Other disadvantages of the present cryostat are the unavoidable sample vibrations caused by the expander module of the cold head and the long cooling time until the lowest temperature is reached. In order to solve the mentioned problems it is planned to install a continuous-flow liquid He cryostat (Janis research, model ST-100). The lowest temperature possible with the new cryostat will be $T = 1.5$ K.

- X-ray absorption spectroscopy (XAS) in the TEY mode is highly surface sensitive. An ion gun is needed to clean the surface from oxide layers or other contaminations. Additionally it will be possible to measure XAS via the fluorescence yield, which is less surface sensitive, by means of a second photodiode installed close to the sample above the scattering plane.

- So far the motor position is only set by the stepper motor control but the exact mechanical motor position is not monitored. In case of a malfunction, for example
6.4. Instrumental outlook

if a motor loses steps, the complete alignment has to be redone. The zero positions of $\omega$ and $2\Theta$ have to be realigned as described above. Even worse, if some error occurs in the interplay of sample and magnet rotation, sample holder or magnet yoke might be damaged in a crash. So far this scenario did not happen. However, it would be desirable to install a stepper motor position control to avoid damages and time-consuming realignment procedures.

- A collaboration for measurements of incoherent [126, 127] and coherent small-angle scattering on membrane substrates in transmission geometry has already been started, where characteristic stripe domains in magnetic multilayers with perpendicular anisotropy (such as Co/Pt or Co/Pd) provide an excellent model system. Especially a new approach for lensless imaging of magnetic nanostructures using spectro holography [128, 129, 130] is very promising, since this technique is transferable to a wide variety of different samples, appears scalable to diffraction limited resolution, and is well suited for ultrafast single-shot imaging with future coherent x-ray free-electron laser sources. Recently in a first experiment the ALICE setup was used to demonstrate that spectro holography is also easily compatible with applying external perpendicular fields up to 7 kOe to the membrane sample, thus being suitable to monitor magnetization reversal processes similar as in transmission x-ray microscopy [47]. After this successful start within the collaboration, it is now necessary to provide an own new CCD camera that is permanently installed in the ALICE chamber for speckle and spectro holography measurements.

- With an increasing number of external users the requirements to the computer control and the general handling of the diffractometer gain importance. Regarding the SPEC software, a graphical user interface (GUI) has been developed by the Beamline Instrument Software Support (BLISS) group at the European Synchrotron Radiation Facility (ESRF). The computer code can be downloaded from the ESRF homepage [131]. The implementation of a GUI software is one step to organize the experimental setup more user friendly. Another simplification of the handling could be the motorization of most degrees of freedom such as the linear sample manipulation, which are operated manually at the present time.

It is quite clear that it is extremely difficult to install all improvements at the same time. For instance it might be impossible to reach low temperatures when motors for sample tilt and sample rotation are installed. Therefore it is conceivable in future that different components of the setup are individually assembled depending on the technical demands of the specific experiment before a beamtime.
6. The ALICE diffractometer
Part III.

Results and discussion
7. Exchange-coupled Fe/Cr superlattices

7.1. Introduction

During the last decade a growing number of experiments has been carried out using x-ray resonant scattering to study magnetic superlattices, i.e. periodic heterostructures of alternating ferromagnetic (FM) layers and non-FM spacer layers. Among these systems the class of antiferromagnetically (AF) exchange-coupled superlattices plays an important role. Due to the doubling of the magnetic period as compared to the structural period a reflection of purely magnetic origin can be observed at half $q_z$ position of the structural Bragg peak in reciprocal space. Since the long photon wavelengths at the $L_{2,3}$ absorption edges of 3$d$ transition metals (TM) do not allow diffraction experiments for regular crystalline materials, artificial structures with long periods yield the only possibility to get such a purely magnetic Bragg reflection at an accessible reciprocal space vector. Therefore AF exchange-coupled superlattices are ideal model systems to demonstrate the capability of x-ray resonant magnetic scattering (XRMS). So far AF satellite peaks have been observed in Ag/Ni \cite{33} and Co/Cu \cite{37, 132, 133, 134} systems.

The requirements to the experimental environment are relatively low at first sight. The magnetic peak can be observed with any kind of incoming polarization. Therefore a bending magnet beamline is sufficient to perform the experiment. Since the AF peak occurs in the remanent state, an external magnetic field is not needed. Furthermore the amount of magnetic material in multilayer systems is usually large enough to evoke a high scattering intensity. Therefore the measurements can easily be performed.

On the other hand the magnetic structure of interlayer exchange-coupled (IEC) multilayer systems can be very complicated and the quantitative interpretation of experimental data is not easy. So far a quantitative analysis of XRMS data is only reported for FM structures. Tonnerre et al. demonstrated the capability of XRMS to give information similar to that derived from XMCD \cite{33}. In particular they determined the spin and orbital magnetic moments of Ni in a Ag/Ni multilayer by fitting the asymmetry ratio of left and right circularly polarized light at a superlattice reflection using a kinematical approach. Beside the spectroscopic information of x-ray magnetic circular dichroism (XMCD) resonant scattering reveals depth-selective information as well. With the same approach layer-averaged depth profiles of induced 5$d$ magnetic moments in La/Fe and Ce/Fe multilayers could be determined using hard x-rays \cite{89, 135}. The depth profile of induced Pt magnetic moments at a single Pt/Co interface in a spin-valve system has been determined using hard x-rays in the vicinity of the Pt $L_3$ edge \cite{90}. A modified Parratt algorithm was applied for the data analysis. In the recent past an algorithm for
7. Exchange-coupled Fe/Cr superlattices

the calculation of specular reflectivity \[94\] and diffuse scattering \[95\] has been presented, utilizing the distorted-wave Born approximation (DWBA).

All formalisms presented above are limited in several respects. Either the equations are explicitly derived for scattering vectors which fulfill the Bragg condition, the magnetization is fixed to the scattering plane or small angle approximations are used. Most reflectivity simulations so far dealt with magnetically saturated samples. In Chap. 3 the Zak formalism was presented \[59, 92\], which describes all magneto-optical effects to first order in the Voigt parameter \(Q\), including resonant magnetic scattering. Apart from the specular condition there are no restrictions to the magnetization direction, wavelength or angle of incidence. This approach has been successfully applied to describe e.g. the Kerr rotation \[21, 55\] and specular reflectivity \[93\] in the soft x-ray range. Furthermore, it has been shown in Chap. 3 how interface roughness can be included in this formalism by a slicing method.

In comparison to methods like SQUID (superconducting quantum interference device) magnetometry, which measure the integral sample magnetic moment, field-dependent XRMS additionally offers the possibility to measure element-specific hysteresis loops, i.e. the field-dependence of the scattering intensity, in a certain reciprocal space vector. Therefore XRMS can directly probe the field-dependence of magnetic order in a sample. Several groups have studied magnetization reversal processes using XRMS. Idzerda et al. were able to quantify magnetic domain correlations in a exchange-coupled Co/Cr/Co trilayer system with strong uniaxial anisotropy by measuring several hysteresis loops at different angles in the specular reflectivity \[136\]. Freeland et al. measured the field-dependence of specular and off-specular intensities and therewith could differentiate the magnetization reversal of bulk and interface spins in CoFe thin films \[36\], respectively. In the case of weakly AF-coupled Co/Cu multilayers it could be shown that, choosing the AF or structural Bragg peak position, one can draw hysteresis loops of antiferromagnetic as well as ferromagnetic order similar to GMR loops and SQUID hysteresis loops, respectively \[134, 137\].

However, it has been pointed out by several authors that these hysteresis loops are not identical with SQUID or GMR measurements. Bragg peak positions and Kiessing fringes can be shifted as a function of the applied magnetic field due to the change in the real part of the magneto-optical constants. Therefore the reflected intensity depends not only on overall magnetization but on the details of the reflectivity curve as well. Most conclusions of XRMS measurements on exchange-coupled multilayers are drawn on a qualitative basis so far, and appropriate tools for a quantitative data analysis are necessary.

Fe/Cr(001) represents an ideal model system to study the capability of XRMS. On the one hand it has been one of the most heavily studied systems in the field of exchange coupled thin films. On the other hand it exhibits a large variety of effects and is therefore still a subject of current research. Astonishingly, there is no literature available about soft x-ray resonant magnetic scattering on exchange-coupled Fe/Cr superlattices, although oscillatory IEC has been observed at first in this TM system.

A series of Fe/Cr(001) superlattices with varying thicknesses was prepared in order to perform XRMS studies. The results for two samples are discussed here, which are called samples \(A\) and \(B\) in the following. The Cr layer thickness of sample \(A\), an \([\text{Fe}(1.2\, \text{nm})/\text{Cr}(0.8\, \text{nm})]_{20}\) superlattice, corresponds to the first AF-coupling maximum. This
sample clearly shows a purely magnetic Bragg reflection due to the AF coupling. However, magnetization reversal experiments are difficult to interpret, since the maximum field during XRMS experiments is approximately five times smaller than the saturation field of the sample and only minor loops can be measured. Therefore a second [Fe(1.5nm)/Cr(2.5nm)]\textsuperscript{10} superlattice \(B\) was grown close to the second, weaker coupling maximum, which can be fully saturated during XRMS experiments in the setup. Beside the first observation of such an AF peak in the Fe/Cr(001) system, a detailed quantitative analysis of soft XRMS data is demonstrated on this model system. The specular reflectivity has been simulated using the Zak formalism including interface roughness. XRMS hysteresis loops could be understood on a quantitative basis. For this purpose the magnetization measurements have been fitted by minimizing the free energy of the Fe/Cr system yielding the coupling angles between the Fe layers for each magnetic field value. These angles have been taken as input parameters for the Zak formalism leading to a good agreement with the measured data. These results illustrate that so-called optical effects can be understood on the basis of the Zak formalism and that a quantitative analysis of complex magnetic structures is possible with XRMS similar to polarized neutron reflectivity (PNR)\cite{9, 138}.

The chapter\cite{1} is structured as follows. In the next section a short summary of the basic facts of IEC with focus on the Fe/Cr system is given. A series of Fe/Cr superlattices with varying thicknesses was prepared in order to perform XRMS studies. The sample preparation is described in section 7.3. Subsequently experimental results for the two samples are discussed in detail. The chapter is concluded by summarizing the results.

### 7.2. Interlayer exchange coupling

Starting in 1986, the discovery of exchange coupling between FM films separated by non-FM spacer layers evoked great interest in the field of thin film magnetism\cite{6, 141, 142}. It could be shown that the phenomenon is independent of the FM layer material and can be observed in both rare earths and TMs. The spacer layer can consist of any paramagnetic or antiferromagnetic transition or noble metal. Since its discovery the effect has been observed in a wide range of materials\cite{143, 144}. Furthermore it was discovered that the IEC can oscillate between FM and AF, depending on the interlayer thickness\cite{144}. If the relative orientation of the two FM layers is antiparallel, the electrical resistance is significantly enhanced as compared to the parallel case due to spin-dependent electron scattering. This so-called giant magnetoresistance (GMR) effect is now widely used in applications such as magnetic sensors and hard disk read heads. Among the TM systems Fe/Cr(001) played an important role. It was the first TM system which was shown to exhibit AF and FM exchange-coupling. The GMR effect was also found in Fe/Cr at first\cite{7, 143}.

For the transition region between parallel (FM) and antiparallel (AF) coupling, where the coupling is weak, Rühig et al. observed a non-collinear 90\(^\circ\) orientation between the magnetization directions in a Fe/Cr/Fe trilayer\cite{8}. This phenomenon was attributed\cite{139, 140}.

1The chapter is partly based on the articles \textit{X-ray resonant magnetic scattering of Fe/Cr superlattices} and \textit{X-ray resonant magnetic scattering on non-collinearly coupled Cr/Fe superlattices}. Several model calculations have been added.
to the existence of another, biquadratic, exchange coupling additionally to the bilinear coupling causing AF alignment. Altogether the contribution of IEC to the free energy density in a system of two FM layers separated by a diamagnetic or paramagnetic spacer layer can be described phenomenologically by

\[ E(\Delta \phi) = -\frac{J_{AF}}{d_{FM}} \cos \Delta \phi - \frac{J_{BQ}}{d_{FM}} \cos^2 \Delta \phi, \]  

(7.1)

where \( J_{AF} \) and \( J_{BQ} \) are called bilinear and biquadratic coupling constants and \( \Delta \phi \) is the angle between the two magnetization directions. The FM layer thickness \( d_{FM} \) appearing in the denominator of Eq. (7.1) indicates that IEC is an interface-dependent phenomenon. If the first term in Eq. (7.1) dominates, the system is AF-coupled for \( J_{AF} < 0 \) and FM-coupled for \( J_{AF} > 0 \). If the second term dominates, 90°-coupling is obtained for \( J_{BQ} < 0 \).

In case of an AF spacer layer another coupling mechanism is present, which can be expressed phenomenologically as

\[ E(\Delta \phi) = \frac{J_{+}}{d_{FM}} (\Delta \phi)^2 + \frac{J_{-}}{d_{FM}} (\Delta \phi - \pi)^2, \]  

(7.2)

where \( 0 \leq \Delta \phi \leq \pi \) and \( J_{\pm} \geq 0 \) \[146\], the so-called proximity magnetism model. In an ideal system with smooth interfaces one gets \( J_{+} = 0 \) for an even number of monolayers (ML) and \( J_{-} = 0 \) for an odd number of MLs in the spacer layer, leading to an IEC oscillation with a 2ML period, which has been observed in Fe/Cr/Fe trilayer systems \[147\]. In case of monatomic steps at the interface a mixing of both coupling terms is possible. This mixing can result in any intermediate coupling angle between 0° and 180°. It has to be pointed out that in case of metallic antiferromagnets like Mn or Cr both types of coupling, (7.1) and (7.2), can coexist. For example an oscillation period of approximately 1.8 nm can be observed in Fe/Cr additionally to the 2ML oscillations.

The earliest attempt to explain IEC microscopically was made in the framework of the Rudermann-Kittel-Kasuya-Yoshida (RKKY) theory. While the 1/D^2 decay of exchange coupling depending on the interlayer thickness \( D \) \[148\] and oscillation periods \[149, 150\] were predicted correctly in this picture, the RKKY theory failed to describe phases and amplitudes quantitatively. Other theories like the quantum well model \[151\] and the sd-mixing model \[152, 153\] yielded the same oscillatory behavior and decay as the RKKY model with more realistic quantitative results for amplitudes and phases. However, the coexistence of a variety of apparently different mechanisms predicting essentially similar behavior was unsatisfactory. The puzzle was finally solved by Bruno \[154\] and by Stiles \[155\]. It could be shown that the different pictures corresponded to different approximations of a unified model in terms of quantum interferences \[156\]. The basic ideas of this model are outlined below.

An electron propagating in a paramagnetic layer sandwiched between two potential barriers is partially reflected at barriers \( A \) and \( B \) with reflection coefficients \( r_A \) and \( r_B \), respectively. Depending on the phase shift after a complete round trip, the interference of the electron wave is constructive or destructive. Standing waves are formed up for certain electron wave vectors. Therefore the density of states is modified as compared to the bulk due to quantum interferences. If now the potential barriers are FM, the
7.2. Interlayer exchange coupling

Figure 7.1.: Cross section of the Cu Fermi surface along the (110) direction. The dotted line indicates the first Brillouin zone. The nesting vectors $q_{\perp F}$ correspond to the oscillation periods in the given growth directions.

reflection coefficients depend on the relative orientation of electron spin and magnetization. From the changes in the density of states the energy difference between FM and AF configuration can be calculated, yielding

$$E_F - E_{AF} \approx -\frac{1}{\pi^3} \text{Im} \int d^2k_{\parallel} \int_{-\infty}^{\infty} f(\epsilon) \Delta r_A \Delta r_B e^{iq_{\perp D} d\epsilon} d\epsilon,$$

where $\Delta r_{A,B}$ are the differences of reflection coefficients for both spin directions, $k_{\parallel}$ is the in-plane wave vector, $q_{\perp}$ is the difference of wave vectors perpendicular to the barriers and $f(\epsilon)$ is the Fermi function. The integrations over parallel wave vectors $k_{\parallel}$ and over energy show that all electrons in the conduction band contribute to the IEC. The coupling depends on the spin asymmetry of the confinement because of the magnetic layers $A$ and $B$, expressed by the two factors $\Delta r_A$ and $\Delta r_B$, respectively, and on the electron propagation in the spacer layer, which is responsible for the interference effect. The oscillatory behavior of IEC can be directly seen from the exponential function $\exp(iq_{\perp} D)$.

The integrals in (7.3) can be analytically evaluated in the approximation of large spacer thicknesses. Then only electronic states at the Fermi level give predominant contributions to the IEC and the wave vectors can be expanded around $\epsilon_F$. The integration over in-plane wave vectors is calculated in the stationary phase approximation. In this case the only remaining contributions arise from the neighborhood of states having in-plane wave vectors such that the spanning vector of the Fermi surface $q_{\perp F}$ is stationary with respect to $k_{\parallel}$. The spanning vector connects points with antiparallel Fermi velocity. This selection rule was derived in the context of the RKKY model at first [149]. The corresponding contribution oscillates with a period $\Lambda = 2\pi/q_{\perp F}$. There may be several of such stationary spanning vectors and, hence, several oscillation periods. In Fig. 7.1...
the Fermi surface of Cu is shown indicating the stationary spanning vectors for different growth directions. The theoretically predicted oscillation periods are in excellent agreement with experiments [157, 158, 159]. In real samples the interfaces will not be ideally flat and roughness has to be taken into account. The coupling strength might be reduced as compared to theoretically predicted value, because it induces varying effective layer thicknesses, whose phases interfere destructively. In case of Fe/Cr(001) roughness can even suppress the 2ML-period oscillation [152]. More generally Bruno and Chappert have shown that short-period oscillations are more sensitive to roughness than long-period oscillations [150].

So far only bilinear coupling has been considered. Biquadratic coupling may occur as a second-order term in the quantum confinement model. However, this intrinsic effect is much too small to explain the experimentally observed biquadratic coupling. Additionally, the predicted oscillation of \( J_{BQ} \) around zero cannot be brought into agreement with experiments, where \( J_{BQ} < 0 \) has always been found. Instead Slonczewski suggested that biquadratic coupling is a secondary effect, which can be explained by fluctuations of the spacer layer thickness [160]. He showed that a lateral thickness variation by one monolayer can induce fluctuations of \( 2\Delta J_{BL} \) between AF \( (J_{BL} < 0) \) and FM \( (J_{BL} > 0) \) coupling in the transition region, where \( J_{BL} \approx 0 \). Since the exchange stiffness caused by the exchange coupling within the FM layer prevents a variation between AF and FM coupled areas on the lateral length scale of the spacer thickness variations, an intermediate coupling angle results.

As mentioned above the situation is different, if the spacer layer is antiferromagnetic. Slonczewski proposed the existence of helicoidally twisted quasi-antiferromagnetic proximity states in the spacer, which lead him to the coupling energy given in Eq. \( 7.2 \) [146]. The mechanism of non-collinear coupling in presence of spacer thickness variations is displayed in Fig. 7.2. The effect increases with larger lateral length scales of thickness.
Figure 7.3.: X-ray penetration depth of Fe at the $L_{2,3}$ edges (taken from Ref. [75]). The dashed lines indicate the sample thicknesses without the Cr buffer layer. The inset shows tabulated data over a larger energy range for comparison of soft and hard x-ray penetration depths [161].

variations $l$. On the other hand the model holds only for small values of $l$, so that the exchange stiffness preserves sufficient spatial uniformity of each of the magnetic moments in the FM layers. This condition is typically satisfied for $l < 10$ nm.

### 7.3. Sample preparation and experimental

Both Fe/Cr(001) superlattices were prepared under the same conditions by molecular beam epitaxy (MBE) using thermal effusion cells for Fe and Cr with rates of $1.5 \times 10^{-3}$ nm/s and $1.3 \times 10^{-3}$ nm/s, respectively. The superlattices were grown on MgO(001) substrates. To induce high-quality crystalline growth a seed layer of approximately 30 nm Cr was deposited at a rate of $1.5 \times 10^{-2}$ nm/s and a temperature of 450°C. The seed layer was annealed for 30 min. at the same temperature. A 2.5 nm thick Cr capping layer provided protection against oxidation of the underlying superlattice. The base pressure of the MBE chamber was $4 \times 10^{-11}$ mbar before starting the deposition.

The Cr layer thickness of sample A is 0.8 nm corresponding to the first maximum in the AF interlayer exchange coupling [147]. The Cr layer thickness of sample B is close to the second AF coupling maximum (2.5 nm). The number of repeats ($N = 20$ and $N = 10$ for samples A and B, respectively) and the Fe layer thicknesses were chosen to not exceed the total penetration depth of soft x-rays, which is about 50 nm at an energy just below the Fe $L_3$ edge, where we performed most of our XRMS experiments (cf. Fig. 7.3). The epitaxial relationship between the superlattice and substrate was
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MgO(001)
35 nm Cr buffer
→
Fe1.2 nm
0.8 nm Cr
←
Fe1.2 nm
0.8 nm Cr
2.5 nm Cr cap

\{ \Lambda \}
MgO(001)
35 nm Cr buffer
→
1.5 Fe nm
2.5 nm Cr
←
1.5 Fe nm
2.5 nm Cr
2.5 nm Cr cap
\{ 2 \Lambda \}

(a) (b)

Figure 7.4.: Schematic view of the Fe/Cr superlattices with Cr layer thicknesses corresponding to the first (a) and second (b) AF coupling maximum. The structural and the magnetic periods are denoted by \( \Lambda \) and \( 2\Lambda \), respectively.

determined to be MgO(001){110} \parallel Fe(001){100}, which is the usually observed 45° in-plane relationship between bcc Fe and fcc MgO. Both sample designs including the nominal layer thicknesses are schematically depicted in Fig. 7.4.

The samples have been structurally characterized with a standard rotating anode Cu \( \text{K}_\alpha \) x-ray tube in the laboratory in Bochum and with synchrotron radiation at wiggler beamline W1 at HASYLAB, Hamburg. The magnetic properties were determined by means of the standard longitudinal Kerr effect, including rotation measurements in order to investigate the samples magnetic anisotropy, and SQUID magnetometry.

The XRMS experiments were carried out at the bending magnet beamline PM3 and at the undulator beamlines UE56/1 and UE56/2 of the Berlin storage ring for synchrotron radiation (BESSY II). The beamline optics allow to vary the energies in the range from 20 eV to 1300 eV with the possibility of tuning the polarization from linear to circular. The energy resolution was set to approximately \( \Delta E/E = 1 \cdot 10^{-4} \). For XRMS experiments the ALICE two-circle diffractometer was used. The incident and exit slits were set to 300 µm giving an angular resolution of 0.13°. As described in Chap. 6, the sample environment allows to apply magnetic fields in the range ±2.7 kOe and to control the temperature in the range from 30 K to 350 K. The incident photons were either about 90% circularly polarized with positive helicity or 100% linearly polarized with the electric field vector parallel to the horizontal scattering plane (\( \pi \) polarization). The magnetic field was applied in the scattering plane and parallel to the sample surface. This corresponds to the longitudinal magneto-optical Kerr effect geometry.

In Chap. 3 the interplay of the incident photon polarization and the magnetization direction has been explained in detail for ferromagnetic samples. In case of AF-coupled superlattices the situation is more complex, since purely magnetic second-order contributions to the scattered intensity can be dominant at the reciprocal space position corresponding to the magnetic periodicity. The question, which component of the magnetization direction is measured for a specific incoming polarization, can be judged by examining the polarization factors for linear and circular light in Eqs. (1.47) and (1.51), respectively. However, it is difficult to estimate, which contribution is dominant at a certain angle of incidence. Model calculations are helpful in this respect. In the top panel of Fig. 7.5 the reflectivity of an AF-coupled [Fe(1.52 nm)/Cr(2.56 nm)]\text{400} superlattice with
Figure 7.5.: Model calculations of the reflected intensity from an ideal \([\text{Fe}(1.52 \text{ nm})/\text{Cr}(2.56 \text{ nm})]_{400}\) superlattice. In the top panel the specular reflectivity of \(\sigma\) polarized light is shown for \(\gamma_1 = +90^\circ\) and \(\gamma_2 = -90^\circ\). Below, the magnetic signal is shown for different incoming polarizations at the first four AF peaks (1/2, 3/2, 5/2 and 7/2) depending on the direction of the magnetization axis.
Ideally smooth interfaces is shown for linear $\sigma$ polarized light. The magnetization axis is in the scattering plane and in the surface plane. The photon energy is 705.5 eV, slightly below the Fe $L_3$ absorption edge. Fe and Cr layer thicknesses correspond to those of superlattice $B$. The number of 400 bilayers is chosen in order to suppress total thickness oscillations and to increase the magnetic signal. In the shown angular range three charge Bragg reflections can be observed, corresponding to the structural periodicity. The reflectivity curve exhibits four magnetic satellite peaks at the half-order positions, which are denoted by $1/2$ AF ($2\Theta = 12^\circ$), $3/2$ AF ($2\Theta = 38^\circ$), $5/2$ AF ($2\Theta = 65^\circ$) and $7/2$ AF ($2\Theta = 98^\circ$) in the following. Due to the high charge scattering at small incidence angles the $1/2$ AF peak is very weak. The dashed line shows the pure charge scattering, i.e. the Voigt parameter was set to $Q = 0$ in the calculation. In the lower part of Fig. 7.5 the reflectivity is plotted for different polarizations (rows) in the vicinity of the four AF peaks (columns) as a function of the direction of the magnetization axis $\gamma$ and the scattering angle $2\Theta$. In order to visualize the magnetic signal the pure charge scattering $I(Q = 0)$ is subtracted. The magnetization axis is varied with respect to the angle $\gamma$, which is defined in Fig. 7.6. The magnetization vectors of all odd-numbered layers ($\gamma_1$) point in direction of $\gamma$ while the even-numbered layers ($\gamma_2$) are aligned antiparallel.

When the beam is linearly polarized in the vertical plane ($\sigma$, first row in Fig. 7.5), the only contribution stems from purely magnetic $\sigma \rightarrow \pi$ scattering and the experiment is sensitive to the components of magnetization in the scattering plane, i.e. $\gamma = 90^\circ$, $270^\circ$. The situation is more complicated when the beam is linearly $\pi$ polarized (second row). Three components contribute to the magnetic scattering: While the $\pi \rightarrow \pi$ charge-magnetic scattering (T-MOKE) and the second-order $\pi \rightarrow \pi$ scattering are sensitive to the vertical component of magnetization, second-order $\pi \rightarrow \sigma$ scattering measures the component in the scattering plane. Due to the finite penetration depth of the soft x-ray beam the topmost Fe layer contributes to a larger extent to the scattered intensity than the following layers. Therefore a ferromagnetic T-MOKE signal can be observed even at the AF peak positions, which is maximal at $\gamma = 180^\circ$. The relative contribution of the T-MOKE decreases at the higher order AF peaks due to the increasing penetration depth. While the purely magnetic $\pi \rightarrow \sigma$ scattering is almost independent of the angle of incidence, the $\pi \rightarrow \pi$ scattering increases with $2\Theta$ and is the dominant contribution.
7.4. \([\text{Fe}(1.2\text{nm})/\text{Cr}(0.8\text{nm})]_{20}\) superlattice

at the 7/2 AF peak. Similarly, when right circularly polarized (RCP) light is used (third row), the component of ferromagnetic magnetization in the scattering plane is measured (L-MOKE geometry). Since circularly polarized light is a superposition of linear \(\sigma\) and \(\pi\) polarization, all second-order contributions described above can also be observed with circularly polarized light.

Usually a real AF coupled superlattice will not form a single-domain state in remanence. Since the magnetization configuration with \(\gamma_1\) and \(\gamma_2\) permuted is energetically equivalent, both configurations will be present. Typical domain sizes are in the order of several \(\mu\text{m}^2\), much smaller than the illuminated sample area. If the average domain diameter is larger than the longitudinal coherence length of the synchrotron radiation, the scattering from different domains will add incoherently\(^2\). Therefore the intensities \(I(\gamma)\) and \(I(\gamma + 180^\circ)\) are summed for \(\pi\) and RCP in the fourth and fifth row of Fig. 7.5, respectively. The ferromagnetic contributions cancel out in this way. For \(\pi\) polarized light the magnetic intensity at the different AF peaks shows a transition from \(\pi \rightarrow \sigma\) scattering at small \(2\Theta\) to \(\pi \rightarrow \pi\) scattering at large angles. For circularly polarized light the scattered intensity becomes almost independent of the magnetization direction at the 7/2 AF peak. However, even for AF-coupled samples in a multidomain state the ferromagnetic contributions to scattering become important in the presence of an external magnetic field as discussed in the next sections.

7.4. \([\text{Fe}(1.2\text{nm})/\text{Cr}(0.8\text{nm})]_{20}\) superlattice

In this section the experimental results of sample \(A\) are discussed. Fig. 7.7(a) shows a specular reflectivity of the \([\text{Fe}(1.2\text{nm})/\text{Cr}(0.8\text{nm})]_{20}\) superlattice measured with hard x-rays (\(E = 8048\) eV) at beamline W1, HASYLAB. In addition to the two superlattice Bragg peaks at \(q_z = 3.3\) nm\(^{-1}\) and \(q_z = 6.5\) nm\(^{-1}\) two types of thickness oscillations are observed: The faster oscillation period reflects the total thickness of the full stack including the buffer layer and the slower one corresponds to the thickness of the superlattice stack alone. From the analysis of these periods a bilayer thickness of 1.93 nm and a total thickness of 76 nm can be determined. The presence of thickness oscillations beyond \(q_z = 8\) nm\(^{-1}\) (\(2\Theta = 12^\circ\)) indicates smooth interfaces. Since the non-resonant scattering factor in the hard x-ray range depends mainly on the electron number of the elements, the contrast between Fe (\(Z = 26\)) and Cr (\(Z = 24\)) is low. Therefore the superlattice peaks are not very pronounced. The magnitude of the second-order Bragg peak is comparable to that of the total thickness oscillations. Due to the small bilayer thickness a third superlattice peak is too far out in \(q_z\) and cannot be observed.

To determine layer thickness and interface roughness parameters more precisely, the reflectivity has been simulated. The fit result, also shown in Fig. 7.7(a), is obtained with the same computer program based on the Zak formalism, which is used for the simulation of soft x-ray resonant magnetic scattering data. The curves are identical to those obtained with commercial software using the Parratt formalism \cite{87} or alternative codes available in the internet \cite{109}. This finding again underpins the validity of the Zak formalism for the interpretation of x-ray data. The thickness and roughness parameters are

\(^2\)For coherent scattering of domains the scattering amplitudes are summed, and the AF peak in the specular reflectivity vanishes.
7. Exchange-coupled Fe/Cr superlattices

Figure 7.7.: Structural characterization of the \([\text{Fe}(1.2 \text{ nm})/\text{Cr}(0.8 \text{ nm})]_{20}\) superlattice with hard x-rays: Specular reflectivity (a) and transverse scan at the first-order superlattice Bragg peak (b). The measurements (dots) are fitted (lines) as discussed in the main text.

for the Fe \((d_{\text{Fe}}, \sigma_{\text{Fe}})\) and Cr \((d_{\text{Cr}}, \sigma_{\text{Cr}})\) layers are given in Tab. 7.1. However, due to the low contrast of Fe and Cr, fitting is difficult and the error bars of the obtained thickness and roughness parameters are rather large.

In Fig. 7.7(b) a transverse off-specular scan at the first-order structural Bragg peak position is shown measured at the same photon energy. The diffuse scattering has been fitted by a superposition of two different curves. The broad diffuse component has been simulated using the distorted wave Born approximation (DWBA) \[109\], assuming perfect vertical correlation of all interfaces \((\xi_{\perp} = \infty)\). The best fit is obtained with an in-plane correlation length of \(\xi_{1} = 20 \text{ nm}\) and a jaggedness of \(h_{1} = 1\) (for a definition of \(h\) see Sec. 4.2). The roughness parameters have been taken from the reflectivity fit. The specular peak has to be convoluted with a Gaussian profile corresponding to the instrumental resolution. There is a second long in-plane correlation length present, which can best be fitted by a Lorentzian profile (i.e. \(h_{2} = 0.5\)) with a widths of \(\xi_{2} = 1.7 \mu\text{m}\). The parameters are summarized in Tab. 7.1.

The magnetic properties of the Fe/Cr superlattice have been studied by MOKE and SQUID magnetometry. The MOKE measurements exhibit a four-fold in-plane anisotropy. Fig. 7.8 shows a magnetization curve of the sample measured at \(T = 100 \text{ K}\)

Table 7.1.: Structural properties of the \([\text{Fe}(1.2 \text{ nm})/\text{Cr}(0.8 \text{ nm})]_{20}\) superlattice as determined by fits to the hard x-ray scattering. All values are given in nm.

<table>
<thead>
<tr>
<th>(d_{\text{Fe}})</th>
<th>(\sigma_{\text{Fe}})</th>
<th>(d_{\text{Cr}})</th>
<th>(\sigma_{\text{Cr}})</th>
<th>(\xi_{1})</th>
<th>(h_{1})</th>
<th>(\xi_{2})</th>
<th>(h_{2})</th>
<th>(\xi_{\perp})</th>
</tr>
</thead>
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<tr>
<td>1.15</td>
<td>0.26</td>
<td>0.78</td>
<td>0.19</td>
<td>20</td>
<td>1</td>
<td>1700</td>
<td>0.5</td>
<td>(\infty)</td>
</tr>
</tbody>
</table>
Figure 7.8.: Magnetic hysteresis loop at $T = 100$ K (dots) and comparison to the model calculations according to Eqs. (7.4) (dashed line) and (7.5) (solid line). The inset shows the magnetization at small fields enlarged.

by SQUID magnetometry with the external field along the easy direction. The sample exhibits almost perfect AF coupling. The superlattice is still not fully saturated at the maximum field of 5 T, which is technically possible. The remanent magnetization is 20% of the saturation magnetization. If it is assumed that all magnetization reversal is due to coherent rotation, it is possible to simulate the hysteresis loop by minimizing the free energy. According to the phenomenological model of Eq. (7.1) the free energy for a superlattice of $N$ ferromagnetic layers with a four-fold magneto-crystalline anisotropy in presence of an external magnetic field $H$ is given by

$$E(\phi_1, \ldots, \phi_n) = -\sum_{n=1}^{N-1} \frac{J_{AF}}{d_{Fe}} \cos(\phi_n - \phi_{n+1}) - \sum_{n=1}^{N-1} \frac{J_{BQ}}{d_{Fe}} \cos^2(\phi_n - \phi_{n+1}) + \sum_{n=1}^{N} \frac{1}{4} K_1 \sin^2(2\phi_n) - \sum_{n=1}^{N} \mu_0 M_S H \cos(\phi_n - \phi_H),$$  

(7.4)

where the first and second term describe bilinear and biquadratic coupling with coupling constants $J_{AF}$ and $J_{BQ}$, respectively. Here the sign convention is used that negative $J_{AF}$ corresponds to antiferromagnetic coupling and negative $J_{BQ}$ corresponds to orthogonal coupling. The angles $\phi_n$ define the magnetization direction of the $n$th layer, $\phi_H$ is the direction of the external field. Alternatively, the free energy of the multilayer in the proximity model according to Eq. (7.2) is

$$E(\phi_1, \ldots, \phi_n) = \sum_{n=1}^{N-1} \frac{J_+}{d_{Fe}} \left( \frac{\phi_n - \phi_{n+1}}{\pi} \right)^2 + \sum_{n=1}^{N-1} \frac{J_-}{d_{Fe}} \left( \frac{\pi - (\phi_n - \phi_{n+1})}{\pi} \right)^2.$$
with the coupling constants $J_+$ and $J_-$. In order to determine the coupling constants the free energy is minimized for each value of the external field. The resulting hysteresis loop is compared to the experimental curve. The best fits for both models are shown as lines in Fig. 7.8. The fit parameters are summarized in Tab. 7.2. It turns out that the four-fold in-plane anisotropy can be neglected in the fit. While the bilinear/biquadratic model leads to small discrepancies, the proximity model is in perfect agreement with experimental curve for external fields larger than 0.2 T.

The angles $\phi_n$ as a function of the external field are depicted in Figs. 7.9(a) and 7.9(b) for positive magnetic fields with $\phi_H = 0^\circ$. Both models yield similar results: In remanence the coupling angle between adjacent layers is approximately $\Delta \phi = 140^\circ$. Since the Fe layers at the bottom ($n = 1$) and the top ($n = 20$) of the superlattice stack are exchange-coupled only to a single neighboring Fe layer, these layers can be easier rotated in direction of the external field. At small fields the exchange-coupling dominates the free energy and the system tries to maintain the coupling angle present in remanence. Therefore layers $n = 2$ and $n = 19$ even rotate in the direction antiparallel to $H$ first. While all magnetization vectors are aligned along the field direction for fields larger than a finite saturation field $H_S$ in the model of bilinear and biquadratic coupling, the angles $\phi_n$ asymptotically converge at the field direction in the proximity model.

At external fields $H$ below 0.3 T (inset in Fig. 7.8) both the bilinear/biquadratic model and the proximity model fail to reproduce the experimental curve. The remanent magnetization is overestimated by a factor of 1.6 in the model calculations. This indicates that the assumption of coherent rotation of each layer is no longer valid. At small fields the exchange stiffness is not sufficient to keep the layers uniformly magnetized and they break into domains [162]. Due to the fluctuations in the Cr layer thickness, AF-coupled, ferromagnetically and non-collinear coupled domains might coexist. The magnetization reversal is probably caused by domain wall movement rather than by coherent rotation in this regime.

The soft XRMS measurements on sample A revealed the presence of a purely magnetic half-order AF reflection in the specular reflectivity for the first time in the Fe/Cr system. A specular reflectivity and a longitudinal off-specular scan with the sample angle mistuned by $\Delta \Theta = 0.4^\circ$ are depicted in Fig. 7.10. The in-plane Fe(100) direction, i.e. magnetically easy axis of the multilayer, has been aligned parallel to the scattering plane. When the energy of the circularly polarized incident photons is tuned just

<table>
<thead>
<tr>
<th>$M^BL/BQ_{\text{m}}$ [A m$^{-1}$]</th>
<th>$H_S$ [T]</th>
<th>$J_{AF}$ [mJ m$^{-2}$]</th>
<th>$J_{BQ}$ [mJ m$^{-2}$]</th>
<th>$M^\text{prox}_{\text{m}}$</th>
<th>$J_+$ [mJ m$^{-2}$]</th>
<th>$J_-$ [mJ m$^{-2}$]</th>
</tr>
</thead>
<tbody>
<tr>
<td>$1.33 \cdot 10^6$</td>
<td>3.5</td>
<td>-0.62</td>
<td>-0.4</td>
<td>1.4 $\cdot 10^6$</td>
<td>0.85</td>
<td>3.1</td>
</tr>
</tbody>
</table>

Table 7.2.: Parameters resulting from the model calculations according to Eqs. (7.4) and (7.5) for the [Fe(1.2 nm)/Cr(0.8 nm)]$_{20}$ superlattice. Note that the saturation magnetization is asymptotically approached in the proximity model. Therefore a saturation field does not exist.
below the Fe $L_3$ edge, strong magnetic Bragg peaks at the 1/2-order ($2\Theta = 24.6^\circ$) and 3/2-order ($2\Theta = 83^\circ$) positions in reciprocal space are observed in the specular reflectivity additionally to the structural superlattice peak at $2\Theta = 51.2^\circ$. The optimal energy, where the magnetic contrast reaches its maximum, is experimentally determined by performing a scan of the photon energy at the $q_z$ position of the charge and half-order magnetic Bragg peaks in reciprocal space, respectively (Fig. 7.11). The scattered intensity shows a huge resonant enhancement, which is maximal at 705.5 eV for both scans, i.e. approximately 2 eV below the Fe $L_3$ absorption edge (707.5 eV). All soft x-ray measurements of multilayer A were performed at this photon energy.

Figs. 7.10 and 7.11 contain model calculations of the specular reflected intensity based on the Zak formalism, which has been discussed in detail in Chap. 3. In the simulation the magneto-optical (MO) constants of Fe across the $L_{2,3}$ absorption edges have been used as determined from x-ray absorption and XMCD measurements in transmission geometry [75] (see Fig. 3.4). The real part of the MO constants has been calculated from the corresponding imaginary part by means of the Kramers-Kronig transformations (see Sec. 1.5). For the refractive index of Cr tabulated values have been used [161]. In order to fit the experimental data it is advisable to fix all free fit parameters step by step. First, the energy-dependent charge Bragg peak intensity is simulated using the structural parameters from the hard x-ray scattering analysis. In this way the instrumental energy scale can be calibrated, which is calculated from the monochromator settings and might exhibit an offset of several eV. After the adjustment of the theoretical and experimental energy scales the photon energy is fixed in the simulation of the specular reflectivity. In principle the charge profile of the sample is already known from the fit to the hard x-ray data. However, as mentioned before, the contrast between Fe and Cr is low and therefore the ratio of Fe and Cr layer thickness in a bilayer cannot be determined precisely. For photon energies close to the Fe $L_{2,3}$ absorption edges the contrast is much higher. The parameters determined from the hard x-ray and soft x-ray data analysis are generally

Figure 7.9.: Angle of the magnetization direction of each layer in the bilinear/biquadratic model (a) and the proximity model (b).
Figure 7.10.: Specular reflectivity and longitudinal offset scan ($\Delta \Theta = 0.4^\circ$) measured with circularly polarized light at $E = 705.5$ eV.

Figure 7.11.: Reflected intensity as a function of energy in the range of Fe $L_{2,3}$ edges at the structural Bragg peak (triangles) and the 1/2-order AF peak (circles). The solid lines are simulations with the same set of parameters as in Fig. 7.10.
in good agreement. Astonishingly, the specular reflectivity decays faster at soft x-ray energies. Therefore the Fe and Cr charge roughness parameters have to be increased by approximately 0.1 nm each to fit the experimental curve. The origin of this discrepancy is not clear so far. The strong total thickness oscillations observed experimentally cannot be reproduced by the fit, indicating that the absorption of Fe or Cr is assumed too large. For angles larger than $2\Theta = 60^\circ$ the calculated and measured total thickness oscillations run out of phase. The 3/2 AF peak positions also deviate slightly.

The 1/2 and 3/2 AF peaks are modelled by assuming perfect AF coupling with the magnetization axis in the scattering plane. While domains with the magnetization axis perpendicular to the scattering plane might be present as well, they do not contribute to the scattered intensity at the 1/2 AF reflection according to Fig. 7.5. The best fit is obtained by assuming an incoherent superposition of domains with the stacking sequences $(\gamma_{2n-1} = 90^\circ, \gamma_{2n} = 270^\circ)$ and $(\gamma_{2n-1} = 270^\circ, \gamma_{2n} = 90^\circ)$. The magnetic roughness parameters in the multilayer are about 65% of the values of the structural roughnesses. The charge and magnetic depth profiles of the refractive index are shown in Fig. 7.12.

The field dependence of the scattered intensity has been investigated at the first-order Bragg peak and the magnetic half-order peak. The magnetic contribution to the structural Bragg reflection is sensitive to the total magnetization of the sample, that at the half-order peak is sensitive to the antiferromagnetic coupling. Fig. 7.13(a) shows
the hysteresis loop measured at the position of the first-order Bragg peak. Although the data is a bit noisy, the shape of the hysteresis loop is similar to the one measured by SQUID magnetometry as shown by a solid line in Fig. 7.13(a). The SQUID hysteresis reveals a biquadratic contribution to the antiferromagnetic coupling, which cannot be recognized from the magnetic x-ray data. This could be explained by the fact that only a minor loop is measured with soft x-rays, since the maximum field available in the diffractometer ($H = 0.27$ T) is much too small to saturate the sample.

The hysteresis loop at the half-order magnetic peak (Fig. 7.13(b)) has a maximum in the remanent state and decreases with increasing magnetic field. Since circularly polarized x-rays are sensitive to the in-plane sample magnetization in the scattering plane, an additional ferromagnetic contribution to the total scattering amplitude is observed. This explains the difference of the intensity for positive and negative fields.

The presence of a half-order peak in the off-specular scattering in Fig. 7.10 indicates that the ferromagnetic layers are strictly antiferromagnetically correlated from top to bottom. The magnetic diffuse scattering is caused both by magnetic domains and height fluctuations in the magnetic layer thickness, i.e. magnetic roughness as will be discussed now. Fig. 7.14(b) shows a soft x-ray off-specular transverse scan using $\pi$ polarized light at the $q_z$ value of the first-order structural Bragg peak (circles). For comparison the same scan measured at $E = 8048$ eV (see Fig. 7.7(b)) is plotted in the same graph (triangles). Comparing both curves the most prominent feature is the extended range in reciprocal space, which can be probed with soft x-rays. This range is limited by the sample horizon, where either the incident or scattered beam is parallel to the sample surface. Due to the long wavelengths at soft x-ray energies the reflection angle for the structural Bragg peak is large ($2\Theta = 51.4^\circ$) and correspondingly the sample horizon is far out in a transverse scan. Multiple scattering effects such as Yoneda wings are not visible. Therefore the
Figure 7.14.: Transverse scan at the structural Bragg peak position with soft and hard x-rays (a) and at the AF peak position (b).

Born approximation (see Chap. 4) can be applied for the data analysis, which leads to a considerable simplification as compared to the DWBA. A simulation of the soft x-ray data in Born approximation is shown in Fig. 7.14(a) (line). The calculated curve has been corrected for illumination and attenuation effects as already described in Chap. 4. As result the in-plane correlation length of $\xi = 17$ nm and a jaggedness parameter of $h = 0.88$ are obtained in good agreement with the hard x-ray simulation (cf. Tab. 7.1). The specular peak again was simulated by a Gaussian curve with a width corresponding to the angular resolution of the instrument.

The dependence of the diffuse scattering on the applied magnetic field provides information on the domain structure in the film plane and on the spin disorder at the interfaces [36, 104, 105]. Hase et al. have shown that the diffuse scattering around the half-order magnetic peak in AF-coupled magnetic superlattices is of purely magnetic origin [37]. In order to avoid the additional ferromagnetic contribution of the background, these measurements were carried out with linearly $\pi$ polarized x-rays. Fig. 7.14(b) shows transverse scans through the half-order peak measured in the remanent state (squares) and in a field of $H = +0.27$ T (triangles). The transverse scans measured for positive and negative field values are identical. Therefore only one of them is shown in Fig. 7.14(b). Both curves exhibit a narrow component (see inset in Fig. 7.14(b)) and a broad diffuse shoulder. The width of the narrow component drops by a factor of two from remanence to the maximum field value of $H = 0.27$ T. Similarly, the intensity of the diffuse shoulder also drops by a factor of two over the same field range. Such change of the intensity can only be observed at the $q_z$ value of the half-order peak. Otherwise a field dependence can no longer be detected. It is assumed that the shape and intensity of the central peak is due to the magnetic domain structure, while the broad component might result from height fluctuations of the magnetic interface or misalignments of the magnetic spins at the interfaces between the Fe and Cr layers (magnetic roughness). All changes of the rocking curves are completely reversible.
7. Exchange-coupled Fe/Cr superlattices

Figure 7.15.: Hard x-ray reflectivity of the \([Fe(1.5\text{nm})/Cr(2.5\text{nm})]_{10}\) superlattice, measured with a standard \(\text{Cu} \ K_\alpha\) rotating anode x-ray tube, and fit.

The curves have been modelled within the frame of Born approximation (lines). The best fit to the broad diffuse shoulder yields an in-plane correlation length of \(15\) nm and a jaggedness parameter \(h = 1\) independent of the magnetic field, indicating a strong correlation to the structural short-range interface height fluctuations. The narrow component originating from magnetic domains has a Lorentzian line shape. It has been modelled using Eq. (4.17) \[95\]. The average domain size increases from \(280\) nm in remanence to \(680\) nm at \(H = 0.27\) T. The Lorentzian line shape of the rocking curves has already been reported for Fe/Cr multilayers measured by synchrotron Mössbauer and polarized neutron reflectometry \[163\] and for Co/Cu multilayers measured by neutron reflectometry \[106\]. It was explained by a multidomain state of the magnetic superlattice in the remanent state.

7.5. \([Fe(1.5\text{nm})/Cr(2.5\text{nm})]_{10}\) superlattice

The Fe/Cr multilayer \(A\) with the Cr layer thickness in the first AF coupling maximum was ideally suited to demonstrate the occurrence of the half-order magnetic peak due to the doubling of the magnetic period as compared to the structural one. However, because of the strong AF interlayer exchange-coupling it was not possible to fully saturate the sample in the external magnetic field during the soft x-ray measurements. Therefore the Fe/Cr superlattice \(B\) with the Cr layer thickness close to the weaker second AF coupling maximum (\(\sim 2.5\) nm) was prepared in order to investigate the full magnetization reversal by means of XRMS. The focus is on the question whether an XRMS vector-magnetometry is possible by using different polarizations of the incoming light. The
Table 7.3.: Parameters resulting from the model calculations according to Eqs. (7.4) and (7.5) for the [Fe(1.2 nm)/Cr(2.5 nm)]$_{10}$ superlattice.

<table>
<thead>
<tr>
<th>$M_S^{HL/BQ}$ $[\text{A/m}]$</th>
<th>$H_S$ $[\text{T}]$</th>
<th>$J_{AF}$ $[\text{mJ/m}^2]$</th>
<th>$J_{BQ}$ $[\text{mJ/m}^2]$</th>
<th>$M_S^{\text{prox.}}$ $[\text{A/m}]$</th>
<th>$J_+$ $[\text{mJ/m}^2]$</th>
<th>$J_-$ $[\text{mJ/m}^2]$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$1.4 \cdot 10^6$</td>
<td>0.28</td>
<td>-0.037</td>
<td>-0.058</td>
<td>1.43 $\cdot 10^6$</td>
<td>0.17</td>
<td>0.27</td>
</tr>
</tbody>
</table>

price to be paid when studying sample B is that the superlattice exhibits non-collinear IEC, and the AF reflections in XRMS are less pronounced. The experimental results of superlattice B are discussed in the following.

The structural composition of the sample has been determined by hard x-ray reflectometry using a standard Cu $K_\alpha$ rotating anode x-ray tube (Fig. 7.15). From a fit (line) to the measured data (symbols) the layer sequence of the superlattice has been determined to be MgO/Cr(29 nm)/[Fe (1.55 nm)/Cr(2.5 nm)]$_{10}$/Cr(1.0 nm)/Cr$_2$O$_3$(1.9 nm). The roughness parameters of the Fe and Cr layers in the superlattice stack are 0.34 nm and 0.2 nm, respectively.

A hysteresis loop measured by SQUID magnetometry is shown in Fig. 7.16. In remanence the magnetization is 56% of the saturation value. The magnetic field necessary to saturate the sample is below 0.3 T. The hysteresis loop has been simulated in the bilinear/biquadratic and the proximity model according to Eqs. (7.4) and (7.5), respectively. Again the proximity model yields a better agreement to the experimental data. The calculated curve perfectly reproduces the experiment almost over the full field range. Only for very small fields below 5 mT discrepancies can be observed. The fit parameters

![Figure 7.16.](image-url)
7. Exchange-coupled Fe/Cr superlattices

Figure 7.17.: Angle of the magnetization direction of each layer in the bilinear/biquadratic model (a) and the proximity model (b).

for both models are given in Tab. 7.3. In comparison to sample A the bilinear coupling constants $J_{AF}$ and $J_-$ are one order of magnitude smaller. The dominating contribution to the IEC is biquadratic. The resulting rotation angles $\phi_n$ for the Fe layers are depicted in Fig. 7.17. The calculated non-collinear coupling angle in remanence $\Delta \phi \approx 110^\circ$ is close to purely biquadratic coupling. MOKE measurements with varying rotational angles between the crystallographic axes and the external magnetic field revealed the same in-plane anisotropy as for sample A.

In the XRMS experiments the sample was installed with the in-plane hard axis parallel to the scattering plane. The Fe/Cr multilayer was characterized with circularly and linearly $\pi$ polarized radiation in order to measure the longitudinal (L-MOKE) and perpendicular (T-MOKE) magnetization components, respectively. First, the specular reflectivity measured with $\pi$ polarization is discussed. By tuning the photon energy to $E = 705.5$ eV just below the Fe $L_3$ edge (see Sec. 7.4), structural Bragg peaks are observed at the even positions ($2\Theta = 24.6^\circ, 50.9^\circ, 80.0^\circ$) and magnetic Bragg peaks at the half-order positions ($2\Theta = 36.7^\circ, 65.0^\circ, 97.2^\circ$) in units of the reciprocal lattice vector associated with the superlattice periodicity (Fig. 7.18). Because of the high charge-scattering intensity at small angles the 1/2 AF peak is not observed. The line represents the data of a simulation procedure based on the Zak formalism with the optical constants parameters taken from Ref. 75 as above. All structural parameters (layer thicknesses and interface roughnesses) are taken from the fit of the hard x-ray data. Similar as in sample A the magnetic roughness in the multilayer is about 65% of the value of the structural roughness. The best agreement between the experimental data and the calculated values is achieved for the magnetic structure corresponding to biquadratic coupling in the multilayer with the magnetization vector in the iron layer parallel to the easy axis of the in-plane magnetic anisotropy. The contributions of two different domains are added ($\gamma_1 = 45^\circ$, $\gamma_2 = 135^\circ$ and the permutation of $\gamma_1$ and $\gamma_2$), i.e. the net magnetic moment is aligned parallel to the external field. The fit is in overall

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Figure 7.18.: Specular reflectivity of the [Fe(1.52 nm)/Cr(2.56 nm)]_{10} superlattice measured with linear π polarized light (symbols) and fit (line). The inset shows the underlying depth-dependent charge (β_{c}) and magnetic (β_{m}) imaginary part of the refractive index. Each dot represents an individual layer in the sample model including roughness. The alternating magnetization direction is not shown.

Figure 7.18.: Specular reflectivity of the [Fe(1.5nm)/Cr(2.5nm)]_{10} superlattice measured with linear π polarized light (symbols) and fit (line). The inset shows the underlying depth-dependent charge (β_{c}) and magnetic (β_{m}) imaginary part of the refractive index. Each dot represents an individual layer in the sample model including roughness. The alternating magnetization direction is not shown.

good agreement with the experiment. Only the calculated angular position of the 5/2 AF reflection deviates from the measured one. The reason is not quite clear, the same effect already has been observed in the reflectivity of superlattice A (see Fig. 7.10). The structural and magnetic depths profiles for the topmost four bilayers are shown as inset in Fig. 7.18.

The reflected intensity in dependence of the external magnetic field measured with linearly π and right circularly polarized radiation at the position of the third-order structural (2Θ = 80.4°) Bragg peak and the 7/2 AF peak (2Θ = 97.2°) are reproduced in Figs. 7.19 and 7.20, respectively. The intensity at the structural Bragg peak is sensitive to ferromagnetic order. At the half-order reflection the antiferromagnetic order in the sample is measured. However, the specularly reflected intensity might be sensitive to the net magnetization also at angles other than the structural Bragg reflection. Therefore a ferromagnetic signal at 2Θ = 97.2° cannot be excluded. While the intensity of the RCP light at the structural Bragg peak has exactly the same shape as the magnetization curve measured by SQUID magnetometry, the other field-dependent intensities cannot be related directly to a specific magnetization component in the sample. It has been pointed out by several authors that care must be taken in drawing quantitative conclusions from that kind of measurements, since the choice of the scattering angle corresponding to e.g., a Bragg peak or Kiessig fringes influences the exact line shape of the hysteresis [134, 136].

Model calculations have been carried out in order to elucidate the mechanisms un-
derlying line shape of the field-dependent reflected intensity. For the simulation of the experimental data (lines in Figs. 7.19 and 7.20) the angles calculated in the proximity model (Fig. 7.17(b)) were taken as input parameters. The contributions of the two different stacking sequences are added incoherently. It turns out that interface roughness does not have any influence on the shape of the hysteresis loops, there is an intensity scaling effect only. Therefore in order to simplify the simulation procedure calculations are performed assuming ideally flat interfaces. The calculated curves are scaled to the measured intensity for better comparison.

According to Fig. 7.5 the hysteresis loops using circular light are sensitive to the ferromagnetic component along the field direction and to contributions second order

Figure 7.20.: Hysteresis loops at the 7/2 AF peak.

(a) (b)
7.6. Summary and conclusion

in the magnetization parallel as well as perpendicular to the scattering plane. The shape of the hysteresis loop at the structural peak in Fig. 7.19(a) is identical to the SQUID measurements. The calculated curve perfectly fits the data. The field-dependent intensity measured with circular light at the 7/2 AF peak in Fig. 7.20(a) consists of two components: In addition to the ferromagnetic background the curve exhibits an intensity increase at low external fields due to the antiferromagnetic order.

With $\pi$ polarized light the T-MOKE is measured, which is sensitive to the magnetization component perpendicular to the scattering plane. The model calculations for the third-order Bragg peak reproduce the somewhat counterintuitive reduction in intensity for decreasing magnetic fields as well as the intensity increase at the 7/2 AF peak, which is very similar to that measured with RCP light. From the step in intensity at zero external magnetic field present in both curves it has to be concluded that a ferromagnetic component in the sample is switching from the $\gamma = 0^\circ$ direction to $\gamma = 180^\circ$. This step cannot be reproduced by the model calculation. However, assuming that approximately 10% of the multilayer consist of ferromagnetically coupled domains yields a natural explanation of the observed step: The hard axis of the sample has been aligned with a precision of only $\pm 5^\circ$ along the field direction in the experiment. Therefore the easy direction forming the smaller angle with the external field is preferred and a net magnetization component perpendicular to the external field occurs. It is difficult to include this effect in the model of coherent rotation. In principle a microscopic model of the domain configuration in the sample is necessary. Here for simplicity an ad-hoc step function has been added to the hysteresis loop.

7.6. Summary and conclusion

Two different Fe/Cr superlattices with the Cr layer thickness in the first (sample A) and second (sample B) AF interlayer exchange-coupling maximum have been studied by means of x-ray resonant magnetic scattering. SQUID magnetometry reveals almost purely bilinear AF coupling in sample A and non-collinear coupling in sample B with a coupling angle close to 90°. Both multilayers exhibit purely magnetic Bragg reflections at the half-order positions in units of the reciprocal lattice vector of the superlattice periodicity, when the photon energy is tuned close to the Fe $L_3$ absorption edge. The half-order peaks reflect the antiparallel orientation of the Fe magnetization vectors of the adjacent layers in remanence.

The off-specular scattering at the $q_z$ position of the half-order peak reveals two different components: a short-range in-plane correlation of the magnetic roughness on the same length scale as the structural roughness and a narrow diffuse component due to magnetic domains. With increasing magnetic field the width in $q_x$ of the narrow component drops by roughly a factor of two. A multidomain state in remanence is inferred from the Lorentzian line shape of the central half-order peak, which diminishes with increasing field.

The specular reflectivities of both samples are simulated using the Zak formalism including interface roughness by a slicing method. From a fit to the experimental data the coupling angle of adjacent Fe layers can be determined. While it is not possible to saturate sample A in the external magnetic field during the soft x-ray measurements because
of the strong AF-coupling, sample B can be fully saturated during XRMS experiments in the setup. The XRMS hysteresis loops on superlattice B, i.e. the reflected intensity at a constant scattering angle as a function of the external field, can be understood on a quantitative basis as well. For this purpose the magnetization measurements are fitted by minimizing the free energy of the Fe/Cr system yielding the coupling angles between the Fe layers for each magnetic field value. These angles are taken as input parameters for the Zak formalism leading to a good agreement with the measured data.

In conclusion, it has been shown that layer-resolved vector-magnetometry measurements are possible by using XRMS with different polarizations of the incoming light. Polarized neutron reflectometry (PNR) is maybe the only other technique capable of such an analysis \[9, 164\]. The techniques yield complementary information on the magnetic structure. PNR has the advantage of a high penetration depth. The simple relation between the directions of the neutron polarization and the sample’s magnetic induction drastically simplifies interpretation of the experimental data as compared to XRMS. On the other hand with XRMS it is possible to investigate the magnetic structure element-specific. Due to the high brilliance of present-day synchrotron sources the sample volume can be much smaller than in PNR experiments. Therefore it is possible to study the surface magnetism and lateral structures or time-dependent phenomena. A disadvantage of XRMS might be the complex relation between the photon polarization and the direction of the sample magnetization. However, with a proper choice of the experimental geometry and the numerical data analysis available these difficulties can be overcome.
8. Element-specific x-ray circular magnetic dichroism of a Co$_2$MnGe Heusler thin film

8.1. Introduction

Ferromagnetic metals with 100% spin polarization at the Fermi level, so called ferromagnetic half metals, attract increasing attention in the literature, since they have the potential to play an important role in the rapidly emerging field of spintronics [165, 166, 167]. Spintronic devices are designed to control both, the charge current and the spin current of the electrons in one single functional unit and will offer the realization of new concepts of data storage and data processing. Ferromagnetic half metals having only one spin direction at the Fermi level are ideally suited as electrodes in these devices e.g. for spin injection [11, 168, 169], spin filtering [12] or in tunnelling magnetoresistance (TMR) applications [13].

Among the ferromagnetic half metals known in the literature there is an increasing number of Heusler alloys, these are ternary metallic compounds with the general composition $A_2BX$, $A$ and $B$ being transition metal atoms and $X$ a non-magnetic atom [170]. Until now theorists detected about 20 different Heusler phases as half metallic in their band structure calculations [10, 171, 172, 173, 174, 175, 176, 177, 178]. The most popular among them are the classical ferromagnetic half metals PtMnSb and NiMnSb [179], so-called half Heusler compounds, since one of the $A$ sublattices is empty, the new pseudo ternary phase Co$_2$(Cr$_{1-x}$Fe$_x$)Al [178] and the phases Co$_2$MnSi and Co$_2$MnGe [10]. The latter compound is the subject of the present investigation. Co$_2$MnSi and Co$_2$MnGe are considered as excellent candidates for spintronic applications, since the volume magnetization and the ferromagnetic Curie temperatures are high (985 K and 905 K, respectively) and there is a good lattice matching with the GaAs semiconductor family [180]. It has been shown recently that high-quality epitaxial layers of Co$_2$MnGe can be grown on GaAs [181, 182].

The Heusler alloy Co$_2$MnGe grows in the $L2_1$ structure – a cubic lattice combining four fcc sublattices occupied by the Co, Mn and Ge atoms, respectively. Since the narrow gap in the minority spin band at the Fermi energy is induced by symmetry [175], any deviation from perfect site order in the Heusler unit cell can severely affect the degree of spin polarization of the system [14, 15]. Perfectly ordered $L2_1$ symmetry is difficult to achieve even in bulk single crystals of the Heusler phase. In thin film heterostructures, which must be processed at rather low temperatures in order to prevent
excessive interdiffusion at the interface, site disorder can hardly be avoided completely. Thus the presence of site disorder has been suggested to be the main reason for the moderate performance of GMR and TMR devices based on the fully spin polarized Heusler compounds achieved until now [183, 184].

Recent theoretical model calculations of the electronic and magnetic properties of typical point defects in the Co$_2$MnSi and Co$_2$MnGe phase have elucidated this point in detail [185]. A Mn antisite defect, i.e. a Mn atom at a regular Co position in the ordered L2$_1$ lattice, has its magnetic moment antiparallel to the nearest neighbor Mn and Co moments, due to the antiferromagnetic exchange coupling between nearest neighbor Mn atoms. Since in this case the Mn moments count twice, this type of defects will drastically reduce the ferromagnetic magnetization. Co antisite defects, on the other hand, retain their ferromagnetic spin orientation and the magnetic moment remains virtually unchanged [185]. However, these defects contribute to electronic states in the minority spin gap, i.e. very effectively destroy the half metallicity of the films.

These calculations offer a reasonable explanation for the reduced ferromagnetic saturation magnetization observed in thin films of the Co$_2$MnGe phase when prepared at low temperatures [186].

An ideal tool for studying the element specific magnetic moments directly is x-ray magnetic circular dichroism (XMCD). There is only one previous XMCD measurement on the Co$_2$MnGe phase published in the literature until now, which is dealing, however, with a bulk sample with a saturation magnetization close to the theoretical value for ideal L2$_1$ order [187]. In this chapter the results of x-ray absorption measurements and XMCD are discussed, which were performed on a thin film of the ferromagnetic Heusler compound Co$_2$MnGe at the $L_2$, $L_3$ edges of Mn and Co. While the Co spectra are similar to those of metallic Co, the Mn spectra indicate partly localized 3$d$ electrons, which are attributed to Mn atoms at the interface with the Au protection layer. The XMCD spectra are analyzed using sum rules in order to estimate the element-specific magnetic moments. The decrease of the thin film magnetization as compared to the bulk is mainly caused by a reduction of the Mn moments.

8.2. Preparation and experimental

A thin film with a nominal thickness of 11 nm of the Co$_2$MnGe phase has been prepared on a sapphire a-plane substrate, as described in detail elsewhere [186, 189]. To induce a high-quality (110) textured growth a vanadium seed layer with a thickness of 2 nm was sputtered before the deposition of the Co$_2$MnGe film. The film was covered by a 2nm gold cap layer. Au is known to wet the Co$_2$MnGe surface [186], and this thickness is sufficient to provide a perfect protection against oxidation of the surface. This is essential for the analysis below, since x-ray absorption spectroscopy (XAS) measured in total electron yield mode (TEY) probes a surface layer of several nm total thickness due to the electron escape depth, i.e. the method is rather surface sensitive [85].

The Co$_2$MnGe film of the present study was prepared at a substrate temperature of 300°C. Whereas, at a growth temperature of 500°C Co$_2$MnGe films develop about 95%
of the theoretical saturation magnetization (corresponding to 4.95 $\mu_B$/formula unit), at 300$^\circ$C there is a strong reduction of the saturation magnetization to about 60% of the theoretical value $^{[89]}$. Since the main interest of the present study is to investigate the microscopic origin of this moment reduction, the film for the XMCD study has been prepared at 300$^\circ$C. The film had a saturation magnetization of 2.98 $\mu_B$/formula unit at 5 K as measured by SQUID magnetometry. At room temperature, where the XAS and the XMCD spectra were taken, the saturation magnetization was 2.32 $\mu_B$/formula unit.

The structural quality of the film was characterized by hard x-ray scattering using a Cu $K_\alpha$ rotating anode x-ray tube. A low angle hard x-ray reflectivity scan is shown in Fig. 8.1. Film thickness oscillations observed up to $2\Theta = 10^\circ$ reveal the smooth surfaces and interfaces of the film. The parameters given in the subscript of Fig. 8.1 and obtained by fitting the reflectivity using the Parratt formalism $^{[87]}$ corroborate this statement. Fig. 8.2 shows an atomic force microscopy (AFM) picture at the surface of the sample. The Au surface on the top of the film is atomically flat with an rms roughness of 0.1-0.2 nm. In large angle x-ray diffraction only the (220) and (440) Bragg reflections occur proving that the sample has grown with pure (110) out-of-plane texture.

XAS and XMCD were measured at the bending magnet beamline PM3 at BESSY II using the ALICE diffractometer (see Chap. 6). The energy resolution was set to approximately $\Delta E/E = 1 \cdot 10^{-4}$. All absorption spectra were taken by the total electron yield (TEY) method, i.e. by measuring the sample drain current. XMCD spectra were measured with fixed helicity and alternating magnetic field $H = \pm 1.1$ kOe parallel to the sample surface. The degree of circular polarization $P_c$ was approximately 95%.
8. XMCD of a Co$_2$MnGe Heusler thin film

![AFM picture of the sample surface. The Au cap layer is atomically flat with a rms roughness of about 0.1-0.2 nm.](image)

The angle of incidence was chosen to be 40° with respect to the surface. At this angle saturation effects are supposed to be small [85, 86]. The TEY signal is therefore assumed to be proportional to the absorption coefficient, $Y_{\pm} \sim \hbar \omega \mu_{\pm}(\hbar \omega)$, where $\pm$ denotes the magnetic field direction parallel (+) or antiparallel (−) to the photon helicity. The $Y_+$ and $Y_-$ spectra have been corrected for a small offset (< 1%) in the pre-$L_3$ and post-$L_2$ edge region due to different electron trajectories in the external magnetic field [190]. The spectra were normalized to the incoming photon flux. All measurements were performed at room temperature.

8.3. Results and discussion

The averaged x-ray absorption spectra $(Y_+ + Y_-)/2$ at the Mn and Co $L_{2,3}$ edges are shown in Fig. 8.3. The Mn spectrum shows a multiplet structure at the $L_3$ edge with peaks at 1.5 eV and 3.5 eV above the absorption edge maximum and a doublet structure at the $L_2$ absorption edge. These features have not been observed in bulk Co$_2$MnGe Heusler samples with surfaces prepared in situ [187]. The multiplet structure is a clear sign of localized 3$d$ electrons. Calculated absorption spectra of Mn in the $6S_{5/2}$ ground state, the atomic high-spin ground state of Mn, show the same fine structure [187, 191, 192]. The transition from the localized high-spin ground state to the itinerant low-spin ground state of metallic Mn has been described by Dürre et al. [79]. A strong localization of Mn 3$d$ electrons has been observed mainly in two systems: Mn atoms in a noble metal environment with completely filled 3$d$ shells [191, 193] and oxidized Mn [80, 194]. The first interpretation holds in the present case, since the film is protected by an Au cap layer preventing oxidation but allowing some intermixing of Au and Mn at the interface.

The Co XAS shown in Fig. 8.3 is much broader. The shape is very similar to that of pure metallic Co, multiplet structures as typical for CoO are not present [195]. A small shoulder-like structure is seen 1 eV above the $L_3$ absorption edge maximum. A more pronounced structure at the same energy has been observed by Miyamoto et al. and has
Figure 8.3.: XAS spectra at the Mn and Co $L_{2,3}$ edges. The photon energy is defined with respect to the $L_3$ peak position.

been interpreted as an optical transition to the unoccupied minority $t_{2g}$ states in the Co partial density of states (PDOS) [175, 187].

The localized (itinerant) character of the Mn (Co) 3$d$ states can be further quantified by the branching ratio $R = A(2p_{3/2})/(A(2p_{1/2}) + A(2p_{3/2}))$, which is the ratio of background-corrected integrated intensities at the corresponding absorption edges [79, 196]. To subtract the step-like increase of intensity, an arctan function has been used [76]. The function mimics a step of 2/3 and 1/3 of the total step height at the $L_3$ and $L_2$ edge energy, respectively, corresponding to the number of electrons in the core state. The steps are broadened over an energy range $\Delta E=0.2$ eV due to finite instru-
mental resolution, temperature and final state lifetime. For Mn one finds a branching ratio of $R = 0.74$, close to the value for atomic Mn. The Co branching ratio is $R = 0.68$. Within the error bars of the measurement this corresponds to the statistical value of $2/3$ expected for a metal with neither core-valence-electron correlation nor ground state spin-orbit splitting of the conduction band.

XMCD spectra $Y_+ - Y_-$ at the Mn and Co $2p \rightarrow 3d$ absorption edges are displayed in Fig. 8.4. The difference signal has been corrected for the degree of circular polarization and the angle of incidence. Within the noise level of the experiment no fine structure can be resolved neither at the Mn nor at the Co absorption edges. This might be an indication that the XAS results from a superposition of non-magnetic Mn atoms at the Au/Co$_2$MnGe interface and magnetic Mn with itinerant 3$d$ electrons in the Co$_2$MnGe bulk. While interface Mn atoms show the XAS multiplet structure, only the magnetically ordered Mn atoms in the Co$_2$MnGe film contribute to the XMCD signal.

The XMCD measurements are quantitatively analyzed by performing a sum rule analysis in order to determine the element-specific orbital and spin magnetic moments [2, 3]. This procedure might evoke large systematic errors for several reasons: First, the number of 3$d$ holes $n_h$ is experimentally inaccessible and has to be taken from band structure calculations, yielding 2.196 and 4.488 for Co and Mn, respectively [175]. Second, the spin magnetic dipole term $\langle T_z \rangle$ is assumed to be negligible. Principally this assumption seems justified for Co$_2$MnGe because of the cubic symmetry of the system. However, due to the symmetry breaking at the interface the magnetic dipole term could be of importance here. Furthermore, in case of Mn the spin sum rule analysis is problematic because $2p - 3d$ electrostatic interactions are large compared to $2p$ spin-orbit coupling leading to an overlap of $L_3$ and $L_2$ edges and a mixing between the two $j$ levels. Dürre et al. gave a correction factor $\xi = 1.5$ for $jj$ mixing in Mn [79, 80]. Keeping these systematic errors in mind, the sum rule analysis of the Co XMCD spectra yields $m_{\text{spin}} = 0.55\ \mu_B$/atom.
Table 8.1.: Element-specific magnetic moments determined by XMCD, magnetometry and band structure calculations. For a better comparison of XMCD experiments and theory, the element-specific moments measured at room temperature (RT) have been extrapolated to 4.2K. The range of values given for the Mn magnetic moment corresponds to the upper and lower limit of the $jj$ mixing correction factor.

<table>
<thead>
<tr>
<th>Element</th>
<th>Theoretical value</th>
<th>XMCD at RT</th>
<th>Extrapolation to 5 K</th>
</tr>
</thead>
<tbody>
<tr>
<td>Mn</td>
<td>3.04</td>
<td>1.04-1.53</td>
<td>1.36-1.97</td>
</tr>
<tr>
<td>Co</td>
<td>0.98</td>
<td>0.58</td>
<td>0.75</td>
</tr>
<tr>
<td>total</td>
<td>5</td>
<td>2.2-2.69</td>
<td>2.83-3.46</td>
</tr>
</tbody>
</table>

$m_{\text{orb}} = 0.028 \ \mu_B/\text{atom}$. The Mn moments are $m_{\text{spin}} = 0.98 - 1.47 \ \mu_B/\text{atom}$, depending on the value assumed for $\xi$ and $m_{\text{orb}} = 0.056 \ \mu_B/\text{atom}$.

The magnetic moments obtained from the XMCD spectra can be compared to the integral magnetic moment measured by SQUID-based magnetometry. SQUID measurements at room temperature yield a magnetic moment of $m = 2.32 \ \mu_B/\text{formula unit}$. The total magnetic moment at 5 K is $2.98 \ \mu_B/\text{formula unit}$. Correcting the atomic magnetic moments determined by XMCD at room temperature by the same factor, the magnetic saturation moments of Co and Mn given in the last column of Tab. 8.1 are obtained, which can be compared directly to the theoretical moments.

Both, the magnetic moments of Co and Mn are found to be reduced compared to the theoretical moment for a fully ordered Heusler compound. However, the Mn magnetic moment seems to be significantly affected, whereas the value of the Co moment coincides reasonably with the theoretical value within the experimental error. This result is consistent with the model calculations by Picozzi et al. [185]. From these calculations it is expected that an antisite Mn atom has a spin orientation antiparallel to the neighboring Mn spins and contributes a moment of about $-1.4 \ \mu_B$ instead of $+3.04 \ \mu_B$ on a regular Mn position.

When comparing the total magnetic saturation moment of the Co$_2$MnGe molecule of $2.83-3.46 \ \mu_B$ from XMCD with the total saturation magnetic moment of $2.98 \ \mu_B$ from SQUID magnetometry, one finds that average volume magnetic moment nearly coincides with the lower boundary of the magnetic moment determined by XMCD. The magnetization at the Au/Co$_2$MnGe interface might be reduced as compared to the film due to symmetry breaking and interdiffusion. The interface layer thickness is on the order of the rms roughness $\sigma = 0.33$ nm, which is still small compared to the region probed by XMCD. This systematic error in the magnetic moments is estimated to be approximately 15%. Neglecting the difference between the surface layer and the bulk, the total reduction of the Mn moment in Tab. 8.1 can be explained by assuming about 30% of Mn sitting on Co positions. This high degree of site disorder seems not unreasonable considering low preparation temperature and the chemical similarity of Co and Mn.
Mn position, the film has most probably lost its half metallicity [185], and will exhibit a spin polarization similar to other normal ferromagnetic transition metals.

Finally, the magneto-optical (MO) constants of Co$_2$MnGe are determined from the absorption spectra, i.e. the real part $\delta$ and imaginary part $\beta$ of the complex refractive index $n_{\pm} = 1 - \delta_{\pm} + i\beta_{\pm}$ for right and left circularly polarized light at the Mn and Co 2$p$ absorption edges. According to the optical theorem, the imaginary part is directly proportional to the absorption coefficient, $\beta_{\pm} = \mu_{\pm}/(2k)$, where $k$ is the photon wave vector (see Sec. 1.5). To obtain absolute values of $\beta$, the background of the Au cap layer must be subtracted. Therefore the absorption spectra have been normalized to tabulated values [161] in the pre-$L_3$ and post-$L_2$ energy regions.

The associated real part $\delta_{\pm}$ can be calculated by applying modified Kramers-Kronig (KK) relations [69, 67]. The values of $\beta$ outside the measured energy range, which are necessary for the calculation of KK relations, were taken from Henke et al. [161]. The results for both real and imaginary part in the energy region of Mn and Co 2$p$ absorption

---

Figure 8.5.: Charge and magnetic absorption corrections (a,b) and dispersion corrections (c,d) to the refractive index at the $L_{2,3}$ edges of Mn and Co and a comparison to the tabulated refractive index (open symbols) taken from Ref. [161].
The knowledge of the MO constants is essential for the quantitative analysis of the specular reflectivity of a [Co$_2$MnGe/Au]$_{50}$ superlattice as discussed in the next chapter.

### 8.4. Summary and conclusion

The analysis of the element-specific soft x-ray absorption and soft XMCD on the Co and Mn $L_{2,3}$ edges of a Co$_2$MnGe thin film revealed a rather different behavior of the Co and Mn atoms. The XAS of Mn presents a pronounced multiplet structure which is attributed to Mn atoms with partly localized wave functions at the Au interface. The sum rule analysis of the XMCD spectra indicates that the reduced total saturation magnetization is caused solely by a reduction of the Mn moments, as predicted by theoretical model calculations of Co$_2$MnGe with site disorder. Although the validity of the sum rule analysis for Mn has sometimes been doubted, the results are consistent when comparing the magnetization measurements and the element-specific magnetic moments. The high degree of site disorder in the film which is suggested by the strongly reduced Mn magnetic moments is qualitatively consistent with magnetotransport data\cite{189, 197} revealing the small electronic mean free path of the order of 2 nm at 4 K and a strong isotropic spin disorder magnetoresistance. The half-metallicity of the ideal Co$_2$MnGe compound cannot be expected for the film under study here, because the electronic states of point defects will fill the gap in the density of states of the minority spin band. The results presented here point out the difficulties one will encounter when trying to use the full spin polarization of the Heusler compounds in TMR and GMR devices. Without high temperature processing providing a high degree of metallurgical order of the Heusler phase the performance will not be superior to conventional ferromagnetic transition metals.
8. XMCD of a $\text{Co}_2\text{MnGe}$ Heusler thin film
9. Element-specific characterization of the interface magnetism in [Co$_2$MnGe/Au]$_n$ multilayers

9.1. Introduction

The magnetism of the ferromagnetic half-metallic Heusler compounds at the interface with other metals, insulators and semiconductors is a critical issue when judging the perspective of these materials to be used in future spintronic devices. Detailed experimental investigations of the Heusler compounds in the past few years revealed, how difficult it is to realize the theoretically predicted full spin polarization in thin film devices or even in single crystals. Irrespective of the experimental method applied to determine the degree of spin polarization quantitatively, whether by spin-resolved photoelectron spectroscopy [198, 199], Andreev reflection at a superconductor surface [200] or by TMR [181], the degree of spin polarization is always definitely less than 100%. One important reason for this failure, which is often stressed by theorists, is the fact that the experiments determine the spin polarization at the surface and not in the bulk and due to the change of symmetry at the surface the spin polarization might be lost. This principally can be overcome by the right choice of the crystallographic direction and termination at the surface [201].

On a more elementary basis, as discussed in the previous chapter, even in bulk single crystals of the Heusler alloys the full spin polarization might be lost if the crystal structure does not coincide perfectly with the ideal $L_2_1$ structure assumed in the band structure calculations. As usual in ordered metallic alloys, antisite disorder frequently occurs even in well annealed single crystals [202, 203]. Theoretical model calculations taking antisite atoms into consideration show that the electronic states of these defects might fill the gap in the minority spin band and destroy the half metallicity. In Co$_2$MnGe, e.g., Co atoms on the regular Mn positions create electronic states just at the Fermi energy of the minority spin band [14, 185]. In this context the interfaces between the Heusler alloys and semiconductors, insulators or non-magnetic metals, which typically exist in all spintronic devices, need special attention. Even in thermodynamic equilibrium it is not clear a priori that the ordered $L_2_1$ phase is established for the first few monolayers of the film growing on the substrate. The delicate balance of the energy and entropy contributions driving the metallurgical order might be sensitively disturbed by interactions with the substrate and epitaxial strain. Since in many spintronic applications the spin polarization for the first few monolayers is of utmost importance, this
is an essential issue for a critical assessment of the perspective of the Heusler alloys to be used in future spintronic devices.

The x-ray absorption measurements presented in the previous chapter cannot provide quantitative information on the interface magnetism, since the signal is integrated over a volume of several nm thickness at the sample surface. Therefore details of the magnetization distribution within the Heusler layers cannot be resolved. In the present chapter, this problem is tackled by applying the method of soft x-ray resonant magnetic scattering (XRMS) on a high-quality multilayer \([\text{Co}_2\text{MnGe}/\text{Au}]_{50}\). Circularly polarized x-ray radiation is used in the energy range of the Co and Mn \(L_{2,3}\) edges. By observing the difference in the specular reflectivity for the two magnetization directions parallel and antiparallel to the photon helicity in an energy scan across the \(L_{2,3}\) edge of a magnetic element, one can precisely determine the magnetization profile of a ferromagnetic thin film. This has been demonstrated convincingly for single thin films and superlattices by other authors before [89, 90, 135]. Since there are two magnetic elements in the \([\text{Co}_2\text{MnGe}/\text{Au}]_{50}\) multilayer, the analysis can be carried out separately for Mn and Co. It should be emphasized already here that unfortunately in XRMS little can be learned by a mere qualitative inspection of the spectra. Only a sophisticated computer based data analysis and fitting gives the relevant quantitative information. However, with the magneto-optical formalism presented in Chap. 3, a corresponding analysis is possible and reliable. The resulting magnetization depth profile is definitely different for Mn and Co spins and asymmetric with respect to the growth direction. At room temperature non-ferromagnetic interface layers with a thickness of about 0.45 nm at the bottom and 0.3 nm at the top of the \(\text{Co}_2\text{MnGe}\) layers exist.

In addition to the specular reflected intensity, off-specular scattering has been studied. The comparison of non-resonant and resonant magnetic diffuse scattering reveals that the correlated structural and magnetic roughness is almost identical, the corresponding length scale being the in-plane crystallite size.

The subsequent central part of the chapter is organized as follows: After shortly reviewing the bulk magnetic properties of \([\text{Co}_2\text{MnGe}/\text{Au}]_{n}\) multilayers, the conventional hard x-ray structural analysis of the multilayers is presented, including off-specular (diffuse) scattering providing information about the interface morphology and the roughness correlations. Subsequently the main issue of the chapter is discussed, namely the XRMS at the \(L_{2,3}\) edge of Mn and Co. Off-specular XRMS being sensitive to correlations between the chemical and magnetic roughness concludes the experimental section.

### 9.2. Sample preparation and experimental

The superlattice of the present study has a nominal composition \([\text{Co}_2\text{MnGe}(3\text{nm})/\text{Au}(2.2\text{nm})]_{50}\) and has been deposited by r.f. sputtering on sapphire \(a\)-plane at a substrate temperature of 300°C, as described in detail in Refs. [186, 189].

The structural characterization was carried out using non-resonant hard x-ray scattering \((E = 8 \text{ keV})\) at wiggler beamline W1 at the HASYLAB in Hamburg. Two types of scans were performed as schematically depicted in Fig. 9.1. In the specular reflectiv-

\[^1\text{This chapter is based on the article }\text{Element-specific characterization of the interface magnetism in } [\text{Co}_2\text{MnGe}/\text{Au}]_n \text{ multilayers by x-ray resonant magnetic scattering, see Ref. [204].}\]
Figure 9.1.: Scattering geometry in reciprocal space for (a) specular reflectivity ($\Theta - 2\Theta$ scan, $q_x = 0$) and (b) diffuse scattering ($q_x$ scan, $q_z$ constant). Bragg peaks in the specular reflectivity and diffuse Bragg sheets are schematically shown as dots and dotted lines, respectively.

ity (a) the scattering vector $\vec{q}$ is perpendicular to the sample surface. Based on these measurements, information on layer thicknesses and the electron density gradient perpendicular to the surface, i.e. the structural roughness, can be obtained. In a transverse scan $q_z$ is kept constant and $q_x$ is varied. The scattering vector contains an in-plane component and then yields information about correlation lengths in the sample plane.

The soft XRMS experiments were performed with the diffractometer ALICE [121] at the undulator beamlines UE56/1-PGM and UE56/2-PGM2 at BESSY II. The diffractometer comprises a two-circle goniometer and works in horizontal scattering geometry (see Chap. 6). The vertical entrance and detector slits were set to 300 $\mu$m each, resulting in an instrument resolution of 0.14°. Circularly polarized light in the energy range of 600 eV - 900 eV was used with an energy resolution of approximately $\Delta E/E = 1 \cdot 10^{-4}$.

A magnetic field can be applied in the scattering plane along the sample surface either parallel or antiparallel to the photon helicity, which corresponds to the longitudinal magneto-optical Kerr effect (L-MOKE) geometry. The maximum field of ±0.11 T was high enough to fully saturate the sample. The magnetic contribution to the scattered intensity was always measured by switching the magnetic field at fixed photon helicity.

Photon energies were tuned to the regions of Mn and Co $L_{2,3}$ absorption edges in order to determine the element-specific magnetic structure. The resonant magnetic scattering length has been discussed in Chap. 1. It is given by

$$f = (\vec{\epsilon}_i^* \cdot \vec{\epsilon}_f) F_c + i(\vec{\epsilon}_i^* \times \vec{\epsilon}_f) \cdot \vec{m} F^{(1)} + (\vec{\epsilon}_i^* \cdot \vec{m}) (\vec{\epsilon}_f \cdot \vec{m}) F^{(2)},$$

where $\vec{\epsilon}_i$ and $\vec{\epsilon}_f$ are the polarization vectors of incident and scattered x-rays and $\vec{m}$ is a unit vector pointing along the magnetization direction. The scattering amplitude consists of three terms with distinct polarization dependence, where $F_c = -r_e Z + F^{(0)}$ describes the non-resonant (Thomson) and resonant charge scattering and $F^{(1)}$ and $F^{(2)}$ are the first-order and second-order magnetic amplitudes, respectively. In the L-MOKE geometry using circularly polarized light the leading magnetic contribution to scattering
Figure 9.2.: Relative saturation magnetization versus the thickness of the Co$_2$MnGe layer for multilayers [Co$_2$MnGe(d)/Au(3nm)]$_{30}$ (a) and magnetic hysteresis loop measured at 4 K (squares) and 14 K (circles) for a [Co$_2$MnGe/Au]$_{30}$ multilayer after field-cooling in $H = 0.02$ T [205] (b).

arises from the $F^c F^{(1)}$ interference term. Since $F^{(2)}$ is generally small compared to $F^{(1)}$ and second-order contributions do not change with a magnetization reversal, these contributions are neglected in the data analysis.

As in hard x-ray measurements, both, the specular reflectivity and diffuse scattering were measured. In order to separate the structural and magnetic contribution to the scattered intensity it is appropriate to measure the energy-dependent intensity at a fixed scattering angle $2\Theta$ as well. All hard and soft x-ray spectra shown below have been taken at room temperature.

9.3. Results and discussion

9.3.1. Magnetic order of [Co$_2$MnGe/Au]$_n$ multilayers

Fig. 9.2(a) shows the magnetic saturation magnetization measured at 5 K via SQUID magnetometry for [Co$_2$MnGe/Au]$_n$ multilayers with variable layer thickness $d_{\text{Heusler}}$ of the Co$_2$MnGe Heusler layers and the Au layer thickness kept constant at $d_{\text{Au}} = 3$ nm [186]. The saturation magnetization breaks down to very small values below 20% of the theoretical saturation magnetization corresponding to 5$\mu_B$ per Co$_2$MnGe formula unit below a thickness of 1.5 nm. Actually the Co$_2$MnGe layers with $d_{\text{Heusler}} < 1.5$ nm are no longer ferromagnetic but exhibit spin glass order with a spin glass freezing temperature $T_G$ of about 20 K [189]. Above $d_{\text{Heusler}} = 1.5$ nm ferromagnetic order sets in, although with a strongly reduced ferromagnetic saturation magnetization of only about 50% of the theoretical value. The ferromagnetic Curie temperatures reach about 500 K for $d_{\text{Heusler}} = 3$ nm compared to 900 K for the bulk Co$_2$MnGe sample. Interestingly after field cooling the ferromagnetic hysteresis loops of the [Co$_2$MnGe/Au]$_n$ layers exhibit
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Figure 9.3.: Reflectivity of the Co$_2$MnGe/Au superlattice measured with linearly $\sigma$ polarized hard x-rays ($E = 8048$ eV) at wiggler beamline W1 at HASYLAB and simulation (solid line).

a low temperature anomaly, namely a hysteresis loop shifted along the magnetic field axis by an exchange bias field $H_{eb}$ (Fig. 9.2(b)). This is indicative of the existence of a unidirectional exchange anisotropy characterized quantitatively by the exchange bias field $H_{eb}$ [123]. The exchange bias shift of the hysteresis loop sets in at about $T_B = 20$ K, i.e. at about the spin glass transition temperature for the very thin Co$_2$MnGe Heusler layers. Thus it appears natural to attribute the origin of the exchange bias field to a spin glass order of the Co$_2$MnGe/Au interface layers at a blocking temperature $T_B = 20$ K. From this hypothesis if follows that at room temperature the interface layers should be essentially paramagnetic i.e. contribute little to the ferromagnetic saturation magnetization. However, macroscopic magnetization measurements alone can neither really prove this hypothesis nor do they allow to derive details of the magnetization profile within the Heusler layers. This is the starting point and the main aim of the present XRMS study.

9.3.2. Non-magnetic x-ray reflectivity

In Fig. 9.3 an x-ray specular reflectivity of the $[\text{Co}_2\text{MnGe/Au}]_{50}$ superlattice of the present study is shown, measured at a photon energy of 8048 eV. Due to the large difference between the electron densities of Au and Co$_2$MnGe, the contrast between both materials is large even in non-resonant scattering. Superlattice peaks up to the ninth order are visible indicating smooth interfaces. Total thickness oscillations cannot be seen since their period is smaller than the instrumental resolution. A theoretical fit to the data by the Parratt formalism [87] yields thicknesses of 2.29 nm and 2.93 nm for the
Figure 9.4: $q_x - q_z$ map of the reflected intensity in reciprocal space. The Bragg sheets indicate the vertical correlation of the interface roughness. The bar on the right hand side defines the intensity scale in arbitrary units.

Au and Co$_2$MnGe layers, respectively. While the superlattice peak positions are mainly sensitive to the bilayer thickness $\Lambda$, the relative peak intensities strongly depend on the ratio of Co$_2$MnGe and Au layer thicknesses. Therefore by varying the fit parameters, the error bar to the thicknesses given above can be estimated to be 0.05 nm. The rms interface roughnesses $\sigma$ of the Au and the Co$_2$MnGe layer resulting from the fit are $\sigma_{\text{Au}} = 0.4 \pm 0.1$ nm and $\sigma_{\text{Heusler}} = 0.3 \pm 0.1$ nm, respectively.

A complete reciprocal space map of the scattered intensity is shown in Fig. 9.4. The vertical line of high intensity at $q_x = 0$ corresponds to the specular reflectivity as shown in Fig. 9.3. The $q_z$ range covers the first four superlattice peaks. The accessible $q_x$ values are limited by the sample horizon, where either the incident or the scattered beam is parallel to the sample surface (cf. Fig. 9.1). Two different types of diffuse scattering appear: The first type is smeared over the whole reciprocal space and corresponds to vertically uncorrelated roughness. The second type of diffuse scattering, caused by vertically correlated roughness, appears at both sides of the superlattice peak $q_z$ positions.

In Fig. 9.5 a single transverse $q_x$ scan is shown at the $q_z$ position of the third-order superlattice peak. Only at this position in reciprocal space the sample horizon allows to see the full width of diffuse scattering indicating a small in-plane roughness correlation length. The intensity of the diffuse scattering is more than two orders of magnitude smaller than the specular intensity. The additional peaks at $q_x = \pm 0.05$ nm$^{-1}$ and $\pm 0.1$ nm$^{-1}$ arise, because either the incident or the scattered beam satisfies the Bragg condition for the first-order and second-order superlattice peak. The weak shoulder at $q_x = \pm 0.14$ nm$^{-1}$ coincides with the critical angle of Au (Yoneda wings).

The diffuse scattering spectrum can be modelled within the frame of distorted-wave
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Figure 9.5: Transverse $q_x$ scan at the position of the third-order Bragg peak at $q_z = 3.66$ nm$^{-1}$ (dots) and fit within distorted-wave Born approximation (line).

Born approximation (DWBA) \cite{98,109,206}. The result is shown as solid line in Fig. 9.5. The measured curve is not perfectly symmetric around $q_x = 0$, because the sample volume probed is changing with the angle of incidence \cite{102}. To take this asymmetry into account, the theoretical curve has been multiplied by the factor $\sin \Theta / \sin \omega$, where $\Theta$ is half of the detector angle and $\omega$ is the angle of incidence. For incident angles smaller than $\omega_0 = 0.4^\circ$ it has also been taken into account, that part of the incident beam does not hit the sample, giving a correction factor $\sin \omega / \sin \omega_0$. The best fit to the experimental data is obtained for a lateral roughness correlation length of $\xi_l = 18$ nm and a vertical correlation length of $\xi_v = 30$ nm with a hurst parameter $h = 1$ \cite{98}, where $0 < h < 1$ determines how smooth or jagged the surface is. The lateral correlation length can naturally be assigned to the Co$_2$MnGe crystallite size, the vertical correlation length is in good agreement with the thickness of coherently scattering lattice planes obtained from high-angle x-ray diffraction (see below). An additional component of diffuse scattering can be seen at small $q_x$ values close to the specular peak. Since this $q$ position is far away from the critical angles, it can be fitted within kinematical approximation by a Lorentzian curve (corresponding to $h = 0.5$). From the width a correlation length of 0.5 $\mu$m is deduced, which corresponds to the typical terrace length of the Al$_2$O$_3$ substrate due to its miscut angle.

It should be noted that the roughness parameters derived from the specular reflectivity in Fig. 9.3 also are the best parameters to fit the diffuse scattering spectra. This indicates that the rms roughness is caused by topological height fluctuations rather than by interdiffusion of Au and the Heusler components Co and Mn. This is not unexpected since the atomic radii are very different.
9. Interface magnetism in Co$_2$MnGe/Au multilayers

9.3.3. High-angle Bragg scattering

The [Co$_2$MnGe/Au]$_n$ superlattices grow with perfect Co$_2$MnGe(110)/Au(111) texture in the out-of-plane direction. In plane they are polycrystalline [189]. In Fig. 9.6 a scan across the Co$_2$MnGe(220)/Au(111) Bragg peak is shown. Reflections up to the 4th order can be observed, evidencing a good coherence of the out-of-plane growth. The vertical coherence length resulting from the width of the satellite peaks is $\xi_v = 30$ nm, the positions of the satellite peaks yield the same superlattice period as the small-angle reflectivity above.

9.3.4. X-ray resonant magnetic reflectivity

Fig. 9.7 compares the specular reflectivities of the [Co$_2$MnGe/Au]$_{50}$ superlattice up to the third-order Bragg peak, measured with hard x-rays and with circularly polarized soft x-rays at the Co and Mn $L_3$ edge in magnetic remanence, respectively. One should note that the Bragg peak positions in $q_z$ do not exactly coincide for the different energies. This is due to the strong variation of dispersion corrections $\delta(E)$ at the Co and Mn absorption edges according to the modified Bragg law $2A\sin\Theta(1 - \bar{\delta}/\sin^2\Theta) = n\lambda$, where $\bar{\delta}$ is the bilayer-averaged dispersion correction, $\lambda$ is the wavelength and $n$ is an integer number. The peak width depends on the absorption, as can be seen in the Co reflectivity, which is measured at 780 eV, i.e. two eV above the Co $L_3$ edge. The effect is largest for small angles due to the increased photon penetration length in the sample. If the exact structural and magnetic sample composition is known, the dispersive and absorptive corrections to the refractive index can be measured from the energy
### 9.3. Results and discussion

The reflected intensity of circularly polarized x-rays has been measured after magnetic saturation in the directions parallel ($I_+$) and antiparallel ($I_-$) to the photon helicity at the angular position of the first three Bragg peaks. The position is indicated by the arrows in Fig. 9.7. The reflectivity spectra are shown in Figs. 9.8 and 9.9 for the Co and Mn $L_{2,3}$ edges, respectively.

The sum of the intensities for both magnetization directions ($I_+ + I_-)/2$ reflects the pure charge scattering and is independent of the magnetization of the sample. Since the momentum transfer $q_z$ is proportional to the photon energy, the energy scan does not exactly measure the energy-dependent peak intensity but a segment of the specular reflectivity. The scanned $q_z$ range is indicated by two vertical lines for each scan in Fig. 9.7, respectively. This explains the intensity increase for small and intensity drop for large energies, especially in the spectra of the second and third Bragg peak. On this line shape, strong intensity variations due to absorption are superimposed, when the energy is passing the $L_3$ and $L_2$ absorption edges of Co and Mn. Other effects of resonant scattering, such as a shift in peak position due to dispersion (see above) are difficult to assign qualitatively to specific features in the charge intensity spectra.

The best way of visualizing the magnetic contribution to the resonant scattering is to plot the asymmetry $(I_+ - I_-)/(I_+ + I_-)$ (second row in Figs. 9.8 and 9.9). The asymmetry of the first-order Bragg peak of Co is similar to the real part of the magneto-
Figure 9.8.: Charge intensities \((I_+ + I_-)/2\) (top) and asymmetries \((I_+ - I_-)/(I_+ + I_-)\) at the first (a), second (b) and third (c) order Bragg peaks at the Co \(L_{2,3}\) absorption edges. The dots represent measured data, the lines are model calculations. The theoretical charge intensity curves are shifted by a factor 1/5.

optical constant \(\delta_m\) as shown in Fig. 8.5 with a slow increase starting approximately 10 eV below the \(L_3\) edge, a zero-crossing at the \(L_3\) edge, a plateau between the \(L_3\) and \(L_2\) edges, and a second zero-crossing. The asymmetry of the first-order Mn peak

Figure 9.9.: Charge intensities (top) and asymmetries at the first (a), second (b) and third (c) order Bragg peaks at the Mn \(L_{2,3}\) absorption edges. The dots represent measured data, the lines are model calculations. The theoretical charge intensity curves are shifted by a factor 1/5.
looks more like the corresponding imaginary part $\beta_m$ of the optical constant, i.e. the XMCD signal, because the plateau between the $L_3$ and $L_2$ edges is almost absent. It is conspicuous that the overall sign of the asymmetry appears inverted for the second-order Co Bragg peak compared to the first- and third-order peak. In case of Mn the asymmetry of the second- and third-order Bragg peak is inverted. This behavior already indicates on a qualitative level that the ferromagnetic magnetization introduces new length scales which are different for Co and Mn.

In order to evaluate a magnetization depth profile for the $[\text{Co}_2\text{MnGe}/\text{Au}]_{50}$ superlattice from the magnetic part of the reflectivity spectra in Figs. 9.8 and 9.9, the energy-dependent intensities and asymmetries have been modelled within a magneto-optical matrix formalism developed by Zak et al. using the classical dielectric tensor [59, 92]. Within this formalism it is possible to calculate the reflectivity for electromagnetic radiation of arbitrary incidence angle and polarization on layered structures with an arbitrary magnetization depth profile. The formalism is not limited to Bragg reflections and has been used to model magneto-optical effects at soft x-ray energies in a number of publications before [21, 55, 93]. The presence of structural and magnetic interface roughness can be included by dividing the interface into $N$ discrete layers (typically $N = 20$) with constant refractive index, which has been shown to yield the same results as the Nevot-Croce approach [88, 208].

The analysis following Ref. [59, 92] requires the knowledge of the energy dependence of the refractive index $n = 1 - (\delta_c + \delta_m) + i(\beta_c + \beta_m)$ with the charge contributions $\delta_c$ and $\beta_c$ and the magnetic contributions $\delta_m$ and $\beta_m$. This has been determined for a Co$_2$MnGe film in a separate x-ray absorption experiment as described in Chap. 8. The magneto-optical constants are shown in Fig. 8.5.

In the upper panels of Figs. 9.8 and 9.9 the measured and the calculated charge scattering intensities $(I_+ + I_-)/2$ are compared. The set of parameters characterizing the chemical structure, i.e. the thickness and roughness parameters, which has been determined from the previous analysis by hard x-ray small angle reflectivity, is kept fixed in the fit of the spectra. There is an overall good agreement of the spectra, looking apart from details for the third-order Bragg peaks where the largest $q_z$ range is scanned.

In the next essential step the asymmetry is fitted with the same set of fixed parameters (lower panels in Figs. 9.8 and 9.9), assuming a profile for the depth dependence of the ferromagnetic magnetization as discussed in detail below. It should be stressed that the shape of the magnetic profile is the only free parameter during the fit.

If it is assumed that the magnetization profile follows exactly the chemical profile, i.e. is given by the ideal step profile plus a smearing at the interfaces given by the chemical roughness parameter, even the qualitative features of the asymmetry spectra cannot be reproduced. The first, second and third order Bragg peak asymmetries of Co and Mn then approximately have the same shape, similar to that of the first-order Bragg peak of Co. Only if non-ferromagnetic interlayers at the interface of Co$_2$MnGe/Au are allowed, the complex features of the experimental spectra can be reproduced. The best fits are shown as solid lines in the lower panels of Figs. 9.8 and 9.9 and reproduce the experimental spectra perfectly. The corresponding magnetization profiles for Co and Mn are shown in Fig. 9.10.

For a better comparison of charge and magnetic profiles the refractive indices are
normalized according to

\[
\rho_c(z) = \frac{|\delta_c(z) - \delta_{c,Au}|}{|\delta_{c,Co_2MnGe} - \delta_{c,Au}|}, \quad \rho_m(z) = \frac{\delta_m(z)}{\delta_{m,Co_2MnGe}},
\]  

so that \(\rho(z) = 1\) (\(\rho(z) = 0\)) if the refractive index corresponds to the bulk Co\(_2\)MnGe (Au) value. The corresponding imaginary part \(\beta_{c,m}\) has of course the same \(z\) dependence as the real part. In case of Co the best fit is obtained for non-ferromagnetic layer thicknesses of \(d_t = 0.3\) nm at the top and \(d_b = 0.45\) nm at the bottom of the Co\(_2\)MnGe layer. For Mn the fit yields thicknesses of 0.58 nm and 1.08 nm for the upper and lower non-ferromagnetic layers, respectively. The magnetic roughness parameters are almost identical to the structural ones – \(\sigma_m = 0.28\) nm for Co and Mn – and are assumed to be identical for the upper and lower magnetic interface.

The shape of the asymmetries at the Bragg peaks of different order is very sensitive to the magnetization profile, as shown exemplarily in Fig. 9.11. In the model calculation in Fig. 9.11(a) the thickness of the non-ferromagnetic layer at the top and at the bottom of the Co\(_2\)MnGe layer is taken as identical and varied between 0 and 1.3 nm. The asymmetry is calculated at photon energies of 775 eV and 635 eV, i.e. a few eV below the Co and Mn \(L_3\) edges. The results are very similar for both energies: While the asymmetry of the first-order Bragg peak is almost not affected by the presence of non-ferromagnetic interlayers, the magnitude and even the sign of the asymmetry at the second- and third-order peaks is drastically changed. The non-ferromagnetic thicknesses consistent with the experimentally observed sign of the asymmetry are marked by grey bars for both elements. Thus, qualitatively by just matching the sign of the asymmetry observed in the experiment, one can define that the non-ferromagnetic interface layer
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(a) Asymmetry at the first three BPs with constant energy below the Mn (left) and Co (right) $L_3$ edges, respectively, as a function of the non-ferromagnetic layer thickness. The non-ferromagnetic layers at bottom and top of the Co$_2$MnGe layers are assumed to be equal in thickness.

(b) Energy-dependent asymmetry at the Mn $L_{2,3}$ edge for the second-order BP. The ratio of bottom and top non-ferromagnetic layer thickness is varied with the total thickness kept constant. 1: $d_t = d_b = 0.83$ nm, 2: $d_t = 0.58$ nm, $d_b = 1.08$ nm, 3: $d_t = 0.38$ nm, $d_b = 1.30$ nm, 4: $d_t = 1.08$ nm, $d_b = 0.58$ nm.

Figure 9.11.: Symmetric (a) and asymmetric (b) variation of the non-ferromagnetic layer thicknesses $d_b$ and $d_t$.

thickness for Co must be in the limits $0.2$ nm $< d_{t,b} < 0.6$ nm, for Mn the lower limit is $0.6$ nm $< d_{t,b}$.

Also the spectacular and rather puzzling asymmetry of the magnetization profile can be determined precisely: As a result of the fit e.g. Mn possesses a non-ferromagnetic layer at the bottom, whose thickness is nearly a factor of two larger than that at the top. In order to demonstrate the sensitivity of the spectra to the distribution between $d_t$ and $d_b$, a model calculation is shown for the second-order Bragg peak of Mn assuming a constant total non-ferromagnetic thickness $d_{nm} = d_t + d_b = 1.63$ nm with a different distribution between $d_t$ and $d_b$ in Fig. 9.11(b). Only distribution 2 with $d_t = 0.58$ nm and $d_b = 1.08$ nm can reproduce the experimental spectrum. It should be stressed that this set of parameters yields the best fit to all three spectra in the lower panel of Fig. 9.9.

The magnetic roughness parameter is not critical in the fitting procedure. Any roughness parameter in the range $\sigma_m = 0.15$ nm and 0.35 nm yields a good fit without changing the other parameters.

Finally, a simple model is presented which naturally explains the overall sign of the asymmetry observed at the different Bragg reflections. If the optical reflection from the periodic multilayer is considered as diffraction from a one-dimensional crystal, the envelope curve of the superlattice Bragg peaks is given by the absolute square of the bilayer form factor \[209\]. For non-resonant charge scattering the form factor $F_{\text{charge}}(q_z)$ is the Fourier transform of the electron density depth profile. Correspondingly, since in the L-MOKE geometry the resonant charge-magnetic interference term is measured, the
9. Interface magnetism in Co$_2$MnGe/Au multilayers

Figure 9.12.: Sketch of the simplified sample model as described in the main text (a) and the resulting magnetic form factors as a function of the magnetic layer thickness according to Eq. (9.5) (b).

The magnetic form factor $F_{\text{mag}}(q_z)$ is now the Fourier transform of the magnetization depth profile,

$$F_{\text{mag}}(q_z) \sim \int_{-\Lambda/2}^{\Lambda/2} m(z) \exp(iq_zz) \, dz,$$

(9.4)

where $m(z)$ is the fraction of saturation magnetization at depth $z$. Since the charge form factor is independent of the magnetic structure, it is a constant for each Bragg peak. In the simplest model $m(z)$ is one for $|z| < d_{\text{mag}}/2$ and zero for $|z| > d_{\text{mag}}/2$ (Fig. 9.12(a)).

Additionally, for simplicity of the argument only the real part is considered in the following. Then the integration in Eq. (9.4) can easily performed, yielding

$$F_{\text{mag}}(q_{nz}) \sim \sin \left( n\pi \frac{d_{\text{mag}}}{\Lambda} \right),$$

(9.5)

where $q_{nz} = 2\pi n/\Lambda$ is the reciprocal space vector of the nth superlattice peak. In Fig. 9.12 the sinusoidal functions for $n = 1, 2, 3$ are shown. The signs of the three form factors are chosen to be positive, when the structural and magnetic thickness coincide. In this case both, the charge and the magnetic form factor should be either positive or negative and correspondingly their product is always positive. The result is in perfect agreement with the simulations shown in Fig. 9.11(a). It should be kept in mind that this model is oversimplified and a simulation of the measured data in the frame of the magneto-optical approach as described above is necessary to draw quantitative conclusions. However, while this model is not suitable for the quantitative
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Figure 9.13.: Diffuse scattering at the Co (a) and Mn (b) edges. The measured charge (circles) and magnetic (triangles) intensities are modelled within the framework of Born approximation (lines) as described in the main text.

data analysis, it clarifies the mechanism behind the inversion of the overall sign in the asymmetry spectra.

9.3.5. Magnetic diffuse scattering

Similar to the case of the non-magnetic off-specular scattering, it is possible to study the off-specular magnetic scattering and to derive information about the correlation of the magnetic roughness [35, 95, 104, 105]. Fig. 9.13 shows transverse scans at the $q_z$ position of the first-order superlattice peak for Co and Mn, respectively. Since the sample horizon is at larger $q_x$ values due to the larger soft x-ray wavelengths as compared to hard x-rays, the full width of diffuse scattering is already visible at the first-order Bragg peak. For each element the transverse scans were measured at an energy slightly below the $L_3$ edge where the absorption is relatively low and the asymmetry has a maximum (cf. Figs. 9.8(a) and 9.9(a)). The corresponding charge $(I_+ + I_-)/2$ and magnetic $(I_+ - I_-)$ transverse scans are shown in Figs. 9.13(a) and 9.13(b). Both charge and magnetic scans exhibit the same three components as the hard x-ray transverse scan in Fig. 9.5: the specular peak at $q_x = 0$, a narrow component due to the substrate roughness ($\xi_1, h_1$), and a broad diffuse component corresponding to a short-range in-plane correlation ($\xi_2, h_2$). The experimental data do not show any signs of multiple reflections like Yoneda wings. Therefore the Born approximation is sufficient to describe the diffuse scattering [104, 105]. The parameters for the best fits to the four transverse scans are listed in Tab. 9.1.

For simplicity the vertical correlation of interfaces is assumed to be perfect in the model. This assumption is justified by the fact that the vertical correlation length as determined from the hard x-ray data is larger than the penetration depth of the soft x-rays at these energies and incidence angles. The roughness parameters for the charge
Table 9.1.: Parameters for the fit to transverse scans at both Co and Mn absorption edges.

<table>
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<th>$\xi_2$ [nm]</th>
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and the magnetization are taken from the simulation of the reflectivity. As for hard x-ray diffuse scattering the theoretical curve is multiplied by a factor $\sin \Theta / \sin \omega$ correcting the varying illumination. Due to absorption the penetration length of the soft x-rays in the sample changes with $\omega$, and the beam attenuation depends on the incidence and reflection angles. Savage et al. have presented a correction factor taking into account this effect [102], which is also included in the calculations of the Co and Mn transverse scans.

Since the intensity difference is proportional to the charge-magnetic interference term, the existence of magnetic diffuse scattering at the Bragg peaks already indicates that the chemical and magnetic interfaces are correlated. Within the experimental error bars the magnetic interface morphology follows exactly the chemical one. This observation confirms again that the presence of non-ferromagnetic layers is intrinsically correlated to chemical disorder in the sample. Both chemical and magnetic interfaces have a correlated roughness on the scale of the crystallite size.

9.4. Summary and conclusion

The main aim of the present chapter was to derive the profile of the ferromagnetic magnetization inside a $[\text{Co}_2\text{MnGe}/\text{Au}]_n$ multilayer by the method of element-specific soft x-ray resonant magnetic scattering. In order to increase the reliability of the fitting procedure of the magnetic x-ray scattering spectra the structural parameters determined by conventional hard x-ray scattering have been used as fixed input parameters.

It turned out that the asymmetry spectra at the Mn and Co $L_{2,3}$ edges have a rich internal structure which react sensitively on the magnetization profile. This sensitivity originates from the interference of waves scattered by the charge distribution and the magnetization distribution. Qualitatively the superlattice Bragg reflection of $n$th order depends on the $n$th Fourier component of the magnetization distribution and by this is sensitive to details of the magnetization profile. For a reliable determination of the magnetization profile it was essential to use a high quality multilayer instead of a simpler Au/Co$_2$MnGe bi- or trilayer system, since the change of the asymmetry spectra at different superlattice Bragg peaks contains the most important information.

The resulting ferromagnetic magnetization profile within the Co$_2$MnGe layers gives clear evidence for the existence of non-ferromagnetic interlayers, as has been hypothesized from the temperature dependence of the exchange bias field already. The thickness of the non-ferromagnetic interlayer is larger at the bottom than at the top of the
Co$_2$MnGe layer and, even more remarkable, the shape of the magnetization profile determined for Mn and Co is definitely different.

The latter result seems unreasonable at the first glance, since one would expect that only one ferromagnetic layer in the core of the Co$_2$MnGe layer can exist. However, it gets a plausible explanation, when the complex correlation between the chemical structure and the ferromagnetism of the Co$_2$MnGe Heusler alloys are taken into account:

Metallurgically strongly disordered Co$_2$MnGe is non-ferromagnetic and has a spin glass type of magnetic ground state [215]. Moderately disordered Co$_2$MnGe with, say, the Ge sublattice intact but with a large number of antisite defects in the Co and Mn sublattices is ferromagnetic but with a reduced ferromagnetic saturation magnetization [188]. Theoretical model calculations show that a Co spin on a regular Mn position keeps its ferromagnetic spin orientation and its full moment of about 0.7 $\mu_B$ per Co atom [185]. Mn on a Co position, however, has an antiparallel spin orientation and a reduced moment. Thus the Mn spin is much more affected by the site disorder than the Co spin. This theoretical prediction was confirmed by the XMCD measurements of the single Co$_2$MnGe film in the previous chapter [188].

The different experimental magnetization profiles for Co and Mn can then be explained by assuming a gradual transition of the metallurgical order of the Co$_2$MnGe film from strongly disordered (bcc type) for the first few monolayers grown on Au to well ordered ($L_2_1$ type) in the core of the Co$_2$MnGe layer. The edge of the Co profile (see Fig. 9.10) indicates the onset of the ferromagnetic magnetization, the edge of the (narrower) Mn profile indicates the position inside the ferromagnetic layer where the degree of site disorder is low enough to give a resultant ferromagnetic magnetization of the Mn spins. Thus the details in the Mn and Co profiles in Fig. 9.10 can be interpreted as follows:

At the bottom of the Co$_2$MnGe layer there is a non-ferromagnetic interlayer of 0.45 nm thickness. Between 0.45 nm and 1.08 nm there is ferromagnetic order, however with a low magnetization resulting essentially from the Co spins, since the Mn spins are frozen in random parallel and antiparallel orientations. Between 1.08 nm and 2.3 nm the chemical structure approaches the ordered $L_2_1$ structure and the Mn moments are oriented ferromagnetically. Approaching the top interface with Au, a metallurgically disordered layer without a ferromagnetic magnetization from the Mn spins ranging from 2.3 nm to 2.7 nm is followed by a non-ferromagnetic layer of 0.3 nm thickness at the top of the Co$_2$MnGe layer.

Translating this model into absolute values for the magnetic moments, the integration over the magnetization profiles in Fig. 9.10 shows that only about 45% of the Mn spins and 70% of the Co spins contribute to the ferromagnetic magnetization. The total magnetic moment measured by SQUID based magnetometry is about 47% of the theoretical moment of 5$\mu_B$ per formula unit expected if all Mn and Co spins would contribute to the ferromagnetic magnetization. Referring the experimentally determined total magnetic moment to the ferromagnetically ordered Mn and Co spins within the Co$_2$MnGe layer only, a saturation magnetic moment of 4.5 $\mu_B$ per formula unit is derived, which is pretty close to the theoretical full saturation moment. This indicates that in the core of the Co$_2$MnGe layers the magnetization is high and the chemical structure is well ordered $L_2_1$.

The asymmetry of the profile in the growth direction with a thicker non ferromagnetic layer at the bottom than at the top of the Co$_2$MnGe layer is a further point of interest.
which needs discussion. When deposited on the Au surface the first few monolayers of Co$_2$MnGe grow in a strongly disordered structure, maybe in the bcc structure with random occupation of all sublattices. Only after reaching a thickness of about 1 nm the ordered $L2_1$ structure is being established. The reason for this could be interdiffusion of Au and Co$_2$MnGe, however, from the analysis of the diffuse x-ray scattering this can be excluded. The interfaces are sharp and the roughness only originates from correlated topological fluctuations. Thus, the disorder seems to be induced by the Au interface, e.g. by the non-perfect lattice matching and corresponding lattice strain or by a tendency towards surface segregation of one component of the ternary Heusler compound.

The growth conditions at the other interface, namely at the top of the Co$_2$MnGe layers are very different, since here the Au film grows on a metallurgically well ordered Heusler film. Nevertheless, strong disorder and a non-ferromagnetic interlayer occur again. This disorder could be induced by the bombardment of the Heusler surface by Au atoms in the sputter discharge or again by interactions with the Au surface.

Finally, coming back to the important question concerning the applicability of the fully spin polarized Heusler alloys in the field of spintronics, the present results indicate that the magnetism at the interfaces with other materials is a very delicate point. Non ferromagnetic interlayers as existing at the Co$_2$MnGe/Au interfaces can be detrimental for spintronic applications. So one is either limited to applications which can tolerate a low or vanishing spin polarization of a thin interlayer, or one must carefully search for surface combinations of the Heusler compounds and other materials without loss of ferromagnetism and chemical order.
10. Conclusions

Soft x-ray resonant magnetic scattering allows to determine element-specific and depth-resolving information of the local magnetic order of thin films, multilayers, and alloys. Within the framework of the present study a new diffractometer for XRMS experiments with an extended range of parameters has been constructed and successfully implemented. Several external user groups have already used the instrument for XRMS measurements on a wide spectrum of different physical systems. A positive evaluation of the project by the BMBF resulted in a second funding period allowing to continuously improve the experimental setup. Planned upgrades of the setup include a new continuous-flow He cryostat, the installation of a magnetic field perpendicular to the scattering plane and vacuum-compatible stepper motors for slits, sample tilt and in-plane rotation. First experiments in transmission geometry (incoherent and coherent small angle scattering, spectro holography) have been performed on multilayers exhibiting perpendicular magnetic anisotropy. For this kind of experiments it is planned to provide a new CCD camera that is permanently installed in the chamber.

It turns out that a mere qualitative analysis of XRMS spectra is often not sufficient. In order to gain quantitative information several numerical tools were developed. The calculation of the specular reflected intensity for radiation of arbitrary polarization and incidence angle onto structures having arbitrary magnetization distributions was performed using the Zak matrix formalism. Roughness was included by slicing the interface into a series of infinitesimal layers. The diffuse scattering was calculated by means of the Born approximation. This treatment is sufficient, since the transverse scans are generally measured at large scattering angles compared to the total reflection edge and the spectra do not exhibit effects of multiple scattering.

Two different multilayer systems have been studied in this work. The main results are briefly summarized in the following:

Interlayer exchange-coupled Fe/Cr(001) superlattices are ideally suited to demonstrate the capabilities of XRMS. Two different samples have been studied with the Cr spacer layer thickness in the first and second maximum of antiferromagnetic coupling. The first sample reveals almost purely antiferromagnetic coupling while the second sample is non-collinearly coupled with a coupling angle close to 90°. When the energy of the incoming photons is tuned close to the Fe $L_3$ absorption edge, both superlattices show half-order peaks reflecting the antiparallel orientation of the Fe magnetization vectors of the adjacent layers in remanence. The antiferromagnetic and ferromagnetic order in the samples has been studied as a function of the applied field by measuring the reflected intensity at different positions in reciprocal space (half-order and structural Bragg reflection). The experiment is sensitive to the magnetization components parallel...
and perpendicular to the external field, when circularly and linearly $\pi$ polarized light is used, respectively. Therefore the determination of a vectorized magnetization profile is possible. It has been demonstrated that the measured spectra can be well reproduced assuming coherent rotation of magnetization in each layer and using the Zak matrix formalism for the calculation of the specular reflectivity.

Thin films and multilayers of the Heusler compound Co$_2$MnGe have been studied by means of soft x-ray absorption spectroscopy, magnetic circular dichroism and resonant magnetic scattering. The analysis of the element-specific XAS and XMCD on the Co and Mn $L_{2,3}$ edges of a Co$_2$MnGe thin film revealed a rather different behavior of the Co and Mn atoms. The XAS of Mn presents a pronounced multiplet structure which is attributed to Mn atoms with partly localized wave functions at the Au interface. The sum rule analysis of the XMCD spectra indicates that the reduced total saturation magnetization is caused solely by a reduction of the Mn moments, as predicted by theoretical model calculations of Co$_2$MnGe with site disorder. Although the validity of the sum rule analysis for Mn has sometimes been doubted, the results are consistent when comparing the magnetization measurements and the element-specific magnetic moments.

Another aim of the XAS measurements was the determination of the magneto-optical constants at the Co and Mn $L_{2,3}$ edges, which have been used to derive the profile of the ferromagnetic magnetization inside a [Co$_2$MnGe/Au]$_{50}$ multilayer from XRMS spectra. The resulting magnetization depth profile within the Co$_2$MnGe layers gives clear evidence for the existence of non-ferromagnetic interlayers at the interface, as has already been hypothesized from the temperature dependence of the exchange bias field. The thickness of the non-ferromagnetic interlayer is larger at the bottom than at the top of the Co$_2$MnGe layer and, even more remarkable, the shape of the magnetization profile determined for Mn and Co is definitely different. The latter result can be understood qualitatively by comparison with band structure calculations indicating that Mn spins are much more affected by site disorder than the Co spins. Therefore, the different experimental magnetization profiles for Co and Mn can be explained by assuming a gradual transition of the metallurgical order of the Co$_2$MnGe film from strongly disordered (bcc type) for the first few monolayers grown on Au to well ordered ($L_21$ type) in the core of the Co$_2$MnGe layer. The disorder at the bottom of the Co$_2$MnGe layer seems to be induced by the Au interface, e.g. by the non-perfect lattice matching and corresponding lattice strain or by a tendency towards surface segregation of one component of the ternary Heusler compound. The thinner non-ferromagnetic layer at the top of the Co$_2$MnGe films might be caused by disorder due to the bombardment of the Heusler surface by Au atoms in the sputter discharge or again due to interactions with the Au surface.

Regarding the applicability of the Heusler alloys in the field of spintronics, the results of the soft x-ray measurements show that the half-metallicity of the ideal Co$_2$MnGe compound cannot be expected for the thin films and multilayers studied here, because of site disorder in the Co$_2$MnGe films. Non-ferromagnetic interlayers as existing at the Co$_2$MnGe/Au interfaces further complicate the situation. These results point out the difficulties one will encounter when trying to use the full spin polarization of the Heusler compounds in TMR and GMR devices.
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List of publications which have resulted from this work


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# Lebenslauf

<table>
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<tr>
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<th>Johannes Grabis</th>
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<tr>
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<tr>
<td>2000 - 2001</td>
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<td>Wissenschaftlicher Mitarbeiter am Lehrstuhl für Experimentalphysik/ Festkörperphysik, Ruhr-Universität Bochum (Prof. Dr. Dr. h.c. H. Zabel)</td>
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