Topical Review

Magnetic reflectometry of heterostructures

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Abstract

Measuring the magnetic configuration at complex buried layers and interfaces is an important task, which requires especially a non-destructive probing technique. X-ray resonant magnetic reflectometry (XRMR) combines the non-destructive depth profiling potential of x-ray reflectometry with the excellent sensitivity for magnetic phenomena, utilizing the x-ray magnetic circular dichroism effect. It provides the magnetic spatial distribution with a precision down to the angstrom scale, combined with element and symmetry specificity, sub-monolayer sensitivity, and the possible separation of spin and orbital magnetic moments. This review provides an overview to the XRMR technique in a tutorial way. We focus on the introduction to the theory, measurement types, and data simulation. We provide related experimental examples and show selected applications.

Keywords: RR, XRMR, XMCD, reflectometry, magnetism, exchange bias, superconductors

(Some figures may appear in colour only in the online journal)

1. Introduction

X-rays passing the critical angle of total reflection from thin films show maxima and minima of the reflected intensity. This observation was already made in the 1930s by Kiessig [1, 2] when investigating glass coated with nickel. It was immediately evident that this technique could be used to determine thicknesses and the quality of interfaces of thin film heterostructures with very high precision. Since then the technique has been used widely for structural analysis [3–5]. With the advent of synchrotron light sources, x-rays of tuneable energies with well-defined polarization states are readily available, allowing one to match exactly the energy of electronic transitions of specific elements. Combining the reflectivity technique with the ability to tune energies to these elemental resonances leads to what is known as x-ray resonant reflectometry (XRR), a technique which provides chemical sensitivity and atomic specificity with enhanced contrast [6].

One of the major advances of using circular polarized x-rays was the rise of the x-ray magnetic circular dichroism (XMCD) effect, discovered in the year 1987 by Gisela Schütz and others [7, 8]. The effect introduces—especially at the transition metal $L_{2,3}$ and the rare earth $M_{4,5}$ edges—dramatic changes in the atomic scattering as a function of the magnetization of the sample [9–11]. In particular, the XMCD effect alters the x-ray index of refraction according to the sample magnetization, and therefore changes the reflected light intensity at an interface. Any x-ray technique can be transformed into its magnetic counterpart, just by using circular polarized light tuned in energy to a XMCD-sensitive x-ray absorption edge [12–15]. In summary, XMCD is a unique technique which provides quantitative determination of element and symmetry selective properties of the sample, even separated for spin and orbital magnetic moments, without detailed structural information [12, 15–17].

X-ray resonant magnetic reflectometry (XRMR) combines the XRR and XMCD techniques by changing the angle of circular polarized, incoming light at resonance energies and measuring the intensity of the specular reflection. As momentum transfer and scattering provide structural
information, any combination of scattering and the XMCD effect provides spatial magnetic information. In the vicinity of a resonance of a magnetic element, the same method enables us to measure the magnetic moment depth profile of the chosen element with high accuracy [18–22]. One of the first reported results is based on XRRM measured on Co performed by Kao at the NSLS Dragon beamline [23, 24]. Here, strong asymmetry ratios were observed at the Co L 2,3 edge, indicating the strong sensitivity and the relation to the XMCD effect.

A popular application of the XRRM technique is the determination of the interface magnetism in heterostructures. Such systems are important for spin injection, tunnelling magneto-resistance (TMR) and giant magneto-resistance devices (GMR), where high spin polarization and spin injection through the interface is a key issue [25, 26]. Other types of heterostructure systems are made of ferromagnetic (FM) and antiferromagnetic (AFM) or non-magnetic (NM) layers. In the case of the FM/AFM systems the phenomenon of exchange bias (EB) appears, i.e. a shift in the hysteresis loop along the external field axis, for FM/AFM heterostructures which have been cooled in an external field below the Néel temperature of the AFM. EB has been originally observed many years ago at the Co/CoO interface by Meiklejohn and Bean [27]. This effect is used in spintronic applications and in hard disc GMR sensors to pin the orientation of one FM layer [28]. It is beyond the scope of this article to give a detailed and full overview on the topic of EB. Therefore we suggest the reading of related recent review articles as [29–31]. Note that despite the fact that EB is widely used, it is not understood microscopically and quantitative models are hard to apply. Nevertheless, there exists a very intuitive model for the EB effect, based on uncompensated magnetic moments in the AFM layers, usually present at the interface to the FM layers due to the orientation of AFM domains while cooling from the paramagnetic to the AFM state with a well saturated FM in contact. In other words, the interface provides FM-like uncompensated moments in the AFM layer. Some of those uncompensated moments will rotate together with an external field driven rotation of the FM, and some will not. It is mainly assumed that the so-called rotatable uncompensated moments provide enhanced coercive fields, while the pinned ones are the source for EB. This has been studied during the last decade utilizing XMCD and XRRM [32–41].

Another class of materials are superconductor (SC)/FM heterostructures. These systems are of particular interest due to the interaction of the superconducting cooper pairs and the ferromagnetic polarized band structure of the FM. In a simplified description Cooper pairs are able to modify the FM and vice versa. Depending on the strength and charge transfer, matching of the band structure and other phenomena, electrons can either move to the other side or be reflected at the interface. This can change the ground state of the interface dramatically, providing new opportunities and effects [42–45].

The main area of interest is usually the stabilization or destabilization of the FM and/or the superconducting state close to the interface. This can yield to a FM layer in the SC or a SC and/or NM layer in the FM. In addition, charge transfer and accumulation, and orbital order can appear which are also corroborated by strain effects and interface induced imperfections [46–48]. Due to these strong relations of competing interactions, artificial SCs have been proposed, made of single layers of originally non-superconducting systems [49]. Many different heterostructures have been investigated by XMCD, XRR and XRRM so far, also consisting of insulator/SC interfaces [46, 47, 50].

In order to understand these phenomena a detailed knowledge of the magnetic structure in those interfaces is necessary which can be provided by XRRM in a unique way. Therefore, some related examples will be presented later on. The aim of the present article is to describe XRRM and show recent advances in this field. In the following section the experimental and theoretical basis is described in detail.

2. The basics behind magnetic x-ray investigations: XMCD

In order to obtain information on magnetic microscopic properties, it is necessary to achieve sensitivity to magnetic samples. As magnetism is based on angular momenta, i.e. spin and orbital moments and their expectation values [15,51], either in applied fields or in remanence, one needs to have an angular momentum sensitive probe. One well-known probe is related to the use of visible or near visible linear polarized light, where the angle of linear polarization changes as a function of sample magnetization [52, 53]. This can happen in reflection (Kerr rotation) or in transmission (Faraday rotation). As these techniques are based on the application of light, they are non-destructive and contactless. Without going into details, Kerr and Faraday effects are related to inter- or intra-band transitions, which are accessible by dipole allowed transitions utilizing photon energies in the eV range. Here, the initial and final states involved are usually band like states, which can be described, for example, in a tight binding model using a mixture of atomic orbitals. In other words, the initial and final states are not pure atomic states. For example, if two different atomic species are present in a compound, transitions from both types of atoms can be excited simultaneously and therefore element selectivity is lost. There are some rare exceptions, however, where one is able to increase or decrease the sensitivity for one atomic site due to photon energy selection [54]. Another consequence of the band like states is that they are not angular momentum eigenstates and therefore are also a mixture in terms of angular momenta. Thus, while visible optical methods are nice to use, they generally are not intrinsically element and angular momentum selective. Therefore, these methods do not provide a separation between spin and angular momenta, and they are only quantitative after comparison with other magnetometry results and they require knowledge of gauge factors [55].

Electron transitions are handled theoretically in dipole approximation by transition rules based on Fermi’s golden rule for the photoelectric effect [56]. These rules only allow a change in the angular momenta of the electrons by $\Delta l = \pm 1$, and a change in the z-component of the electronic system by $\Delta m = 0, \pm 1$, where the latter is related to linear, and left and right circular polarized light respectively [56]. Therefore, a
detailed knowledge of the initial state also fixes the symmetry of the final states, if the light has a well-defined polarization. This can be achieved nicely using core levels as an initial state [57–59]. These core levels can be resonantly excited if the photon energy matches the energy difference necessary to excite the core electron to the unoccupied density of states (DOS). This is schematically shown for Fe in the left part of figure 1. As many magnetic ions are related to the 3d transition metals, we show as an example the Fe metal 2p → 3d excitation. Here the 2p electrons are resonantly excited from the deep bound Fe 2p level. Due to the 2p spin orbit interaction the Fe 2p level is split into a less bound 2p3/2 and a more bound 2p1/2 state excitation, with 13 eV energy difference. The 2p3/2 (2p1/2) transition is also called L3 (L2) edge respectively. The related change in the absorption is shown in the right part of figure 1 but now for the magnetization parallel and antiparallel to the sample magnetization [12, 13]. The background based on the photo-excitation of all lower energy excitations has been subtracted and the intensity is normalized to the height of the ‘edge-jump’, i.e. the non-resonant enhanced absorption—here 2p—at higher energies above 740 eV for Fe. A clear magnetic effect is visible. The change in the absorption is called the XMCD signal [7, 12, 13, 15], which is plotted in the lower right part of figure 1 [60]. Note that for the case of Fe shown here, the magnetism related effect in the optical properties is about 50% of the resonant peak height at the Fe 2p3/2 transition. So the magnetic effect is quite strong and very pronounced, providing enough sensitivity to detect sub-monolayer magnetism and also paramagnetic contributions [61, 62].

The absorption can be measured by a simple transmission experiment, measuring the transmitted intensity ratio with respect to the incoming light intensity for a given sample thickness [12, 15]. The absorbed intensity is nicely described by a Lambert–Beer law with a material-specific absorption coefficient μ as sketched in figure 2. The XMCD signal is proportional to a change in the scalar product between the magnetization vector \( \vec{M} \) and the unit vector of the circular polarization \( \vec{P} \). Therefore, typical XMCD measurements are performed by switching between parallel and antiparallel orientation, because this provides the maximal change in the scalar product and therefore maximum sensitivity [12, 13]. In a typical XMCD measurement the magnetization is flipped, but also the light helicity can be reversed, with no difference if all magnetization components are rotatable by an external field.

Here we describe a very simplified model which explains the origin of the XMCD effect. If we assume a magnetized sample we find majority and minority states, where the occupation of the electrons is higher for the majority and less for the minority states. On the other hand the number of unoccupied states is reversed to the occupied ones, i.e. that the minority states provide a larger number of unoccupied states with respect to the majority ones. If we are now exciting 2p electrons with linear polarized light we will not change the \( z \)-component of the excited photoelectron and therefore we will find on average, over all possible excitation, about the same probability to excite an electron in the majority or the minority channel. If we now use right circular polarized light, we provide angular momentum and every exited electron has a \( z \)-component increased by 1. As the 2p electronic shell is filled and therefore NM these circular excited electrons have a
The XMCD effect depends on the change in the scalar product between the magnetization and the axial unit vector of the circular light polarization.

A more detailed description, which also takes advantage of the orbital magnetic moment and the different excitation channels from the L3 and the L2 edges ends up in the so-called sum rules, which have been introduced by Thole and Carra [16, 17, 60]. In the case of these sum rules, weighted sums and also differences of the integrated XMCD signal at the L3 and the L2 edges are used and normalized (taking the ratio) by the NM resonant x-ray absorption signal (XAS) [12, 13]. These integrals are simply multiplied by initial and final state related factors and multiplied by the number of holes in the final state shell [12, 13].

This XMCD/XAS ratio as used in the sum rules is valid if the radial matrix elements do not change significantly for all energy-dependent 3d band excitations (over the energy range of the XAS signal). The advantage of this ratio is that some errors in the experiment are partially cancelled and absolute values of the absorption are not necessary. This is also because radial matrix elements, which are the same for XAS and XMCD, cancel each other.

As a result spin $S$ and orbital $L$ magnetic moments can be determined quantitatively by the application of sum rules [16, 17, 60].

Another important issue is the energy position of the excitation as function of photon energy. The resonant excitation starts to appear at an energy position close to the binding energy of the core electron, when the energy is sufficient to excite resonantly the bound electron into the desired final state. The binding energy is strongly dependent on the symmetry, the main quantum number, and the charge of the atomic nucleus [15]. Therefore, every different atom and related core electron has its specific binding energy, which is reflected in the energy position of the photon energy-dependent absorption. As observed in figure 1 the Fe L2,3 resonances appear at about 707 and 721 eV, while for example in the subsection devoted to the optical properties, we show in figure 8 the Co L2,3 resonances appearing at a higher binding energy of about 780 eV, simply because the nuclear charge is higher by one elementary charge.

In summary, the XMCD effect has as least four major advantages with respect to the Kerr and Faraday effects. With the photon energy set to an appropriate binding energy, the symmetry of the initial state can be chosen and via dipole selection rules one is able to probe any desired final state. As explained for the 3d electrons an initial 2p state (or for the 4f electrons an initial 3d state) is excited into the magnetic shell of interest. The binding energy of the initial state provides intrinsic element-specific sensitivity, and sum rules give quantitative information separated on spin and orbital moments. In the important cases of the 2p (3d) absorption probing the 3d (4f) magnetism respectively, the absorption cross sections are very strong, resulting in sub-monolayer sensitivity and also the capability to measure element-specific paramagnetism.

In terms of XRMR we simply use the strong XMCD related variation of the x-ray optical properties at the resonance conditions mentioned above. Changes in the absorption then give rise to a magnetism related change in the imaginary (or absorbing part) of the complex index of refraction, and close to the resonance also in the real or dispersive part of the index of refraction. We just want to mention, that the resonant magnetic scattering process itself could be also understood as a virtual excitation and de-excitation into the unoccupied magnetic states of interest (see for example figure 39 in [13]).

3. X-ray reflectometry—the basics

Partial beams reflected in thin films interfere with each other, resulting in constructive and destructive interference. This
phenomenon is well known in the optical reflection of soap bubbles or thin oil films on water, which usually appear coloured if illuminated by white light. In other words, for a distinct geometry one gets constructive interference only for the colour which has a wavelength that fits into the geometry-related effective phase difference. This geometry dependence makes it possible to use reflection measurements as a depth profiling technique.

Since the thickness of the soap bubbles is in the μm range, visible light provides interference patterns. In ultra-thin films (such as those as used in thin film and semiconducting technology or in solid state physics investigations of ultra-thin layers) typical thicknesses of a view nm are present. Therefore, visible light cannot provide interference patterns anymore and x-rays are required. In terms of x-rays the index of refraction is close to 1. As shown in figure 3, the condition for constructive interference can be simply calculated from the index of refraction weighted path length difference, which is

$$\Delta s = P(AD) - P(ABC) \cdot n \lambda$$

$$= 2d \cdot \sqrt{n^2 - \cos^2 \theta} = n \cdot \lambda.$$  (1)

Using \(n_1 \approx 1\) and the identity \(1 = \cos^2(\alpha) + \sin^2(\alpha)\) we get the following constructive interference condition also known as the Bragg condition:

$$\Delta s = 2d \cdot \cos \alpha = 2d \cdot \sin \theta = n \cdot \lambda.$$  (2)

For the momentum transfer \(\vec{q} = \vec{k}' - \vec{k}_0\) we get the following geometric identity:

$$q_x = 0, \quad q_y = 0, \quad \frac{q_z}{k_0} = \cos \alpha.$$  (3)

If we combine the last two equations we get

$$2d \cdot \frac{q_z}{k_0} = n \lambda \iff q_z = k_0 n \lambda \frac{1}{d} = \frac{2\pi}{\lambda} n \lambda \frac{1}{d} = \frac{2\pi}{\lambda} n$$  (4)

which is the Laue equation stating that the reciprocal space vector, here \(2\pi/d\), must be equal to the momentum transfer \(q_z\).

So far we have only described the condition for constructive interference. Also, the strength of the interference is related to the amplitudes of the interfering partial waves. If one wave has a negligible intensity with respect to the other, the amplitude will only vary slightly as a function of the angle of incidence or the momentum transfer. Therefore, it is crucial to know the absolute intensities of the waves which are interfering with each other. As known from visible light optics and from every day’s life experience, the intensity of a reflected wave is higher: (a) for grazing incidence conditions and (b) if the change in the index of refraction is larger between the two media separated by an interface. From photography one also recognizes that it is possible to reduce unwanted reflections by the use of a linear polarizer. This means that the reflection of light also depends on the polarization state of the incoming wave. This has been calculated for the reflected \((r)\) and transmitted \((t)\) light by Augustin Jean Fresnel using

$$t_r = \frac{E_r}{E_i} = \frac{2n_1 \cos \alpha}{n_1 \cos \alpha + n_2 \cos \beta}$$

$$r_r = \frac{E_r}{E_i} = \frac{n_1 \cos \alpha - n_2 \cos \beta}{n_1 \cos \alpha + n_2 \cos \beta}$$

$$t_t = \frac{E_t}{E_i} = \frac{2n_2 \cos \alpha}{n_2 \cos \alpha + n_1 \cos \beta}$$

$$r_t = \frac{E_t}{E_i} = \frac{n_1 \cos \beta - n_2 \cos \alpha}{n_2 \cos \alpha + n_1 \cos \beta}.$$  (5)

Due to energy conservation and reversibility of the optical path, the Fresnel equations are connected and imply the rules \(r_{12} = -r_{21} = r, \ t_{12} = r + 1\) and \(t_{21} = 1 - r\). The subscript defines the two film indices and the propagating direction of the incoming light, i.e. that \(\beta > \alpha\) due to the index of refraction which is usually less than 1.

continuity equations for the electrical field vector parallel (\(\tau\)) or perpendicular (\(\sigma\)) to the scattering plane defined by the incoming and outgoing \(k\)-vector plane [63, 64]:

$$t_{\tau} = \frac{E_{\tau}}{E_i} = \frac{2n_1 \cos \alpha}{n_1 \cos \alpha + n_2 \cos \beta}$$

$$r_{\tau} = \frac{E_{\tau}}{E_i} = \frac{n_1 \cos \alpha - n_2 \cos \beta}{n_1 \cos \alpha + n_2 \cos \beta}$$

$$t_{\sigma} = \frac{E_{\sigma}}{E_i} = \frac{2n_2 \cos \alpha}{n_2 \cos \alpha + n_1 \cos \beta}$$

$$r_{\sigma} = \frac{E_{\sigma}}{E_i} = \frac{n_1 \cos \beta - n_2 \cos \alpha}{n_2 \cos \alpha + n_1 \cos \beta}.$$  (5)

To be more instructive, an example for the simplest arrangement of just one Co layer on a substrate is presented in figure 4. The interference pattern is clearly visible and based on the interfering partial waves reflected at the vacuum/Co and the Co/Si interfaces. A strong and simple oscillation is visible. If we analyse the periodicity in terms of Fe layer thickness as given in the Laue equation introduced above, the thickness of the Fe can be extracted simply and reliably. It should be mentioned, however, that this simple data interpretation tends to break down at resonant conditions and for more complex layer stacks, making a quantitative simulation indispensable (see below).
4. Reflection of real systems

A quantitative model describing the reflectivity just by the interference and the Fresnel coefficients does usually not provide reasonable results. This is true especially in the important soft x-ray range, which is of special interest for magnetic investigations due to the strong XMCD effect close to absorption edges.

The most important issues for this lack of model quality are:

(1) Beams can be reflected many times, which results in a complex multiple beam interference problem, as sketched in figure 3 inside the layer.
(2) Strong magnetic effects appear close to XMCDSensitive absorption edges, the absorption can be very strong. Therefore, the polarization-dependent attenuation of the light must also be taken into account.
(3) The Fresnel coefficients are strongly reduced for rough interfaces.

Indeed, all of these extra issues can be handled quantitatively in a physical model. Thus, the existence of these issues is not only a drawback. In contrast, it allows many more experimental degrees of freedom, i.e. one can merely switch on and off reflecting interfaces, and one can focus on selected interfaces. In the following we will show some examples and provide the basic concepts to handle these issues.

4.1. Multiple beam reflection

Any reflected beam will be reflected for a second time approaching another interface. As a result the total wave field approaching or leaving an interface can be described by multiple scattering analyses, just by summing up all of the partial waves arriving from every reflection or transmission event at each interface. Nevertheless, this problem is usually not solved by simply summing up all the partial waves appearing, but rather by solving the wave field taking into account the Fresnel equations introduced above. The simplest way to do so is by solving using a recursive solution, which has been introduced for both fundamental polarization directions by Parratt in 1954 [66]. The main scheme is sketched in figure 5. At every interface one has to handle for every light polarization four partial beams, two incoming and two outgoing (from above and below) partial waves (figure 5, upper schematics). The continuity conditions of the amplitude of the partial light waves at the interface are as follows:

\[
E_i^R e^{i k_{iz}} = r \cdot E_{i+1}^T e^{-i k_{iz}} + (1 - r) E_{i+1}^R e^{i k_{iz}}, \\
E_i^T e^{-i k_{iz}} = -r \cdot E_{i+1}^R e^{i k_{iz}} + (1 + r) E_{i+1}^T e^{-i k_{iz}}.
\] (6)

The ratio of the amplitudes of the partial waves in each film is defined as \( R_i = E_i^R / E_i^T \). The equation (6) can now be written in the following recursive form:

\[
R_i = \frac{r_i + R_{i+1} e^{2i k_{iz} d_{i+1}}}{1 + r_i R_{i+1} e^{2i k_{iz} d_{i+1}}}
\] (7)

Note the difference in the definition between \( r \) and \( R \). While the first one defines the reflectivity of the electric field of exactly one light beam at an interface, the latter one defines the total electric field ratio in one layer. Therefore the numerator represents a reflection of one beam \( r \) plus the part coming from the layers below. The denominator defines the small correction for multiple scattering, which can be also derived from the geometric series of a multiple scattering solution.

One starts with the lowest interface and simplifies the problem by the reasonable assumption that no light comes back (\( R = r \)) from the thick substrate. The complex ratio between the incoming wave and the reflected wave are the \( R_i \), which are recursively calculated from the equation (7) for the \( s (= \sigma) \) and \( p (= \pi) \) polarized light waves . The \( k_i \) are the normal component of the complex wave vectors in the layer \( i \) and \( z_i \) is the depth of the \( i \)th interface, with \( d_i = \Delta z = z_i - z_{i+1} \) the thicknesses of the corresponding layers.

Figure 4. Co/Si reflectivity at photon energy of 800 eV for a film thickness of 10 nm and 20 nm. Thickness of the Fe layer is determined by the inverse reciprocal lattice vector as shown (ReMagX simulation [65]).
Figure 5. Schematic view of a multilayer system showing the light scattering of the partial waves resulting from multiple scattering. Top: definition of the electric fields of the propagating electromagnetic wave at an interface.

The measured total reflectivity is then given by

$$I_T = |I_0|^2.$$ (8)

The advantage of this method is the fast and relatively easy calculation, but s (衡 = σ) and p (衡 = π) polarized light waves have to be treated independently.

4.2. Strong absorption at the resonances

Due to the very strong absorption at the transition metal L_2,3 edges, the absorption length can be very small, for pure metals down to about 20 nm [67]. This is an important issue for XAS measurements in the total electron yield (TEY) mode at more grazing incidence conditions, where the effective x-ray penetration depth is in the same order of magnitude compared to the effective electron escape length [67]. It is obvious that the exact treatment of the absorption length is a prerequisite for reasonable and valuable simulations and related interpretations of the resonant reflectivity data. This will be briefly discussed later on in the optical constants section. Despite the necessary careful treatment of the resonant absorption, this strong absorption also reveals further prospects and options, which should be shortly discussed now.

The centre of figure 6 shows the optical constants δ and β from the complex index of refraction $n = 1 - δ + iβ$ of Fe metal. In contrast to the first example of a Co film given above, we focus now on a resonant condition, where the 2p → 3d transition of Fe is excited (Fe L_2,3 edges). We choose two different energies, with first energy set just before the absorption edge at about 702 eV and the second at the strong absorption line at about 707.5 eV. The lower energy has small absorptive part of the index of refraction β and a clear resonantly enhanced dispersive part δ, while at the higher energy the absorption has its maximum and a nearly vanishing contribution of δ. Consequently, in the first example the reflected intensity from the Fe/Si interface is only weakly damped, but the index of refraction is strongly enhanced due to the dispersive part. However, for the second energy nearly all intensity reflected from the Fe/Si interface is damped. At small momentum transfer the angle of incidence is more grazing, therefore the effective pathway through Fe is long and the Fe/Si reflected intensity experiences a stronger damping. No oscillations are visible and therefore no intensity is reflected and transmitted from the lower interface. So the reflectivity signal is dominated by the vacuum/Fe interface. Only at higher angles (high momentum transfer) are small wiggles observable.

As shown, despite the necessary knowledge of the optical constants, one has the great opportunity to merely switch on and off the reflected intensity below a thin layer simply by tuning the energy before and on the absorption edge of a resonant excitation of this layer. This provides higher significance and less ambiguity for the data interpretation. As shown above in the on-resonance condition at the maximum
absorption, the properties of the vac./Fe interface contributing most significantly.

4.3. Rough interfaces

It is a well-known phenomenon that rough interfaces result in a reduced reflection. To give an example: at a nice sandy beach one can only see incoming water waves without being reflected back from the sand of the shore, while the same water wave is nicely reflected at the hard wall of a harbour. The explanation for this is because the ‘water wave optical properties’, i.e. the water wave index of refraction, are changing continuously at the beach and abruptly at the harbour wall. Similar things happen in the visible optical regime, where, for example, optical elements are coated with an appropriate antireflection coating. Such a coating is usually an approximation of an effective gradual change in the optical properties, similar to the water wave at the sand beach. A rough surface is—in terms of average optical density—also a gradual change in the optical properties. The only important constraint is the lateral size of the overall rough structures, which should be smaller than the wavelength, otherwise diffuse scattering is present and results (in the optical regime) in an opaque surface. This technique has been successfully implemented by nature in order to reduce the reflection of moth eyes [68, 69], and it is successfully adopted by technology [70, 71].

To give a real reflectometry related example for the influence of the interface roughness, figure 7 shows the simulated reflectivity in an off-resonant condition (at 800 eV) for a thin Fe film deposited on a pure Si substrate. In one case the rms roughness is zero, resembling the pure Fresnel reflectivity at each interface, and in the second case the roughness at the Fe/Si interface has been set to 1 nm. It is clearly visible that the total reflected intensity is reduced dramatically by the roughness, and that the interference pattern is modified even for this small roughness.

This example clearly demonstrates that it is necessary to implement a quantitative description of the interface roughness in an appropriate way, otherwise any reflectivity fit would be orders of magnitude away from experimental results. Nevot and Croce found an analytical description of the roughness in the special case that the optical profile is an error function, which is equivalent to a Gaussian shape of the optical profile gradient [72]. In this simple case the Fresnel coefficients for the reflection are exponentially damped by the roughness of the interface

\[ r_{i,i+1}^{\text{rough}} = r_{i,i+1}^0 e^{-k_{i,i+1} \cdot \sigma^2} \]  

where the roughness factor is \( \sigma \), defined as the root mean square of the vertical interface roughness. In this description only one interface with simply modified, wave vector dependent, effective Fresnel coefficients is used. This approach is always the starting point in every simulation, because it provides
usually good fit results in a shorter time. One property of the correction term is the energy conservation in the specular direction. This is normally not true as roughnesses induces diffuse scattering and therefore a loss of energy in specular direction. The effect is negligible as long the roughness is not significantly larger than the wavelength [73].

Nevertheless, to overcome the restriction to an error function profile which will be even more important in terms of magnetism (see the example ‘Hard x-ray reflectivity for single layer systems: induced magnetism and roughness at Pt/Cu’)—one can approximate the real chemical profile by an artificial slicing of the interface in many different single layers with no roughness between each artificial layer. This method is called multi-slicing and will be briefly demonstrated in the example section.

5. Atomic magnetic scattering

The x-ray scattering of an atom is described in the far field as decaying spherical waves with wave vector \( k_0 = \omega_0 / c \) and can be written as

\[
E_f = -r_e f (q, \omega_0, \vec{e}_f, \vec{e}_i) \frac{e^{i\mathbf{q}\mathbf{r}}}{r} E_i
\]  

(10)

where the incident electric field \( E_i \) amplitude is connected with the scattered field \( E_f \) amplitude (\( r_e \) is classical electron radius). The scattering amplitude of elastic scattering is condensed in a form factor \( f \) (or scattering factor) which is dependent on the momentum transfer \( q \), the energy \( \omega_0 \) of the beam and the complex polarization vectors \( \vec{e}_f \) and \( \vec{e}_i \).

In the dipole approximation, the x-ray atomic scattering factor of a magnetic atom combines several scattering types and is described by the formula of Hannon et al [74].

\[
f (q, \omega, \vec{e}_f, \vec{e}_i) = - (\vec{e}_f \cdot \vec{e}_i) \cdot \left[ f_0(q) + f'(\omega) + f''(\omega) \right]
+ i (\vec{e}_f \times \vec{e}_i) \cdot \vec{b} \cdot \left[ m'(\omega) + im''(\omega) \right].
\]  

(11)

The isotropic part is described by the triplet \( (f_0, f', f'') \) and does not change the polarization of the scattered beam. The conventional form factor \( f_0(q) \) is related to Thomson scattering and is described for an atom as the Fourier transformation \( \tilde{r} \rightarrow \tilde{q} \) of the spatial distribution of the electron density, which is freely accessible by the x-ray photon energy. In reflectivity, \( \tilde{q} \) is typically very small in comparison of the localized electron density. Therefore, the factor is usually set to the effective electron count. In the limit of hard x-ray energies above the highest binding energy of the present electrons this number is just the atomic number \( Z \) of the corresponding atom [75]. This type of scattering is predominant in the off-resonant hard x-ray region.

The anomalous scattering factors \( (f', f'') \) can be separated into two scattering processes. The first process is the excitation of the bound states to unbound continuum states. Those lead to the typical background and absorption edges (step-like structure) in XAS measurements. This scattering type is mainly independent on the chemical environment of the atoms. Tabulated values can be found in [76–78].

The second process involved in the anomalous scattering is the excitonic-like, dipole selected, bound–bound excitation of the core electrons to an unoccupied state, which finally leads to the characteristic resonances at the absorption edges. Since this excitation typically involves the valence shell the resonance shape depends on the local environment.

The magnetic scattering with the magnetic terms \( (m', m'') \) is part of the bound–bound excitation process, but with a different polarization dependence. The scattered polarization is altered and belongs therefore to the anisotropic scattering part. The vector \( \vec{b} \) defines the direction of the magnetic moment.

A convenient method of writing equation (11) is in form of a scattering tensor \( F \) defined as

\[
f(q, \omega, \vec{e}_f, \vec{e}_i) = \vec{e}_f^{\ast} \cdot F \cdot \vec{e}_i,
\]  

(12)

with

\[
F = \left[ f_0(q) + f'(\omega) + i f''(\omega) \right] I + i \left[ m'(\omega) + im''(\omega) \right]
\times \left[ \begin{array}{ccc} 0 & \cos \theta_M & - \sin \theta_M \sin \varphi_M \\ - \cos \theta_M & 0 & \sin \theta_M \cos \varphi_M \\ \sin \theta_M \sin \varphi_M & - \sin \theta_M \cos \varphi_M & 0 \end{array} \right].
\]  

(13)

The vector \( \vec{b} \) is written in spherical coordinates as

\[
\vec{b} = \left( \begin{array}{c} \sin \theta_M \sin \varphi_M \\ \sin \theta_M \cos \varphi_M \\ \cos \theta_M \end{array} \right)
\]  

(14)

with the angles \( \theta_M \) and \( \varphi_M \). Thus, if one has a given magnetization direction, one extracts the angles \( \theta_M \) and \( \varphi_M \) and enters them into equation (13). When presented in this form, it is easy to distinguish between the isotropic scattering part formally defined as \( \frac{1}{2} \text{Tr} F \) and the anisotropic part given as \( F - \frac{1}{2} \text{Tr} F \).

The relation between the atomic form factors and the macroscopic quantities like the electric susceptibility tensor.
\( \chi \) is given by the optical theorem [79] and states that
\[
\chi = \frac{4\pi}{k_0} r_0 \sum_i \rho_i F_i. \tag{15}
\]
The sum runs over all elements \( i \) of the system given the atomic density \( \rho_i \). The dielectric permittivity tensor is then defined as
\[
\varepsilon = 1 + \chi = n^2 = (1 - \delta + i\beta)^2 \approx 1 - 2\delta + 2i\beta. \tag{16}
\]
The last three terms describe the refractive index \( n \) in the system in case of an optical isotropic system. For a description of the quantum mechanical treatment of the interaction of light with the magnetization which is finally the optical description of the XMCD effect.

By applying equations (15) and (16) the refractive index becomes
\[
\beta = \beta_0 \mp \beta_M \left[ \sin \theta \cos \theta_M - \cos \theta \sin \theta_M \sin \varphi_M \right] = \delta_0 \pm \delta_M \left[ \sin \theta \cos \theta_M - \cos \theta \sin \theta_M \sin \varphi_M \right], \tag{19}
\]
and is now a function of the direction of the incoming light connected by the angle \( \theta \). This refractive index can be used to calculate the reflectivity curves for both polarizations.
\[
\beta_M \text{ is proportional to the XMCD signal } \Delta \mu \text{ and is given by}
\beta_M = \frac{\Delta \mu}{4k_0 \left[ \sin \theta \cos \theta_M - \cos \theta \sin \theta_M \sin \varphi_M \right]}, \tag{20}
\]
where the complex index of refraction is related to the absorption coefficient by \( \mu = 2k_0\beta \).

### 6. Parratt algorithm with magnetism

The Parratt formalism as originally presented can only be used for isotropic materials. \(\sigma\) and \(\pi\) polarization are independent, leading to two separate solutions. Circular polarized light is introduced by averaging the intensities of both linear polarizations. Because of these limits the Parratt formalism is only capable to calculate the four separate light channels (in \(\rightarrow\) out) \(\sigma \rightarrow \sigma, \pi \rightarrow \pi, l \rightarrow l\) and \(r \rightarrow r\).

To understand the atomic response for these channels we can use equation (12). Using the coordinate system introduced in figure 3, the two polarization vectors for linear polarized light are
\[
\vec{e}_\sigma = \begin{pmatrix} 1 \\ 0 \\ 0 \end{pmatrix}, \quad \vec{e}_\pi = \begin{pmatrix} 0 \\ \sin \theta \\ \cos \theta \end{pmatrix}. \tag{17}
\]
The circular polarized light vectors are a combination of both linear polarized light vectors with a complex phase shift of 90° defined as
\[
\vec{e}_l = \frac{1}{\sqrt{2}} (\vec{e}_\sigma + i\vec{e}_\pi), \quad \vec{e}_r = \frac{1}{\sqrt{2}} (\vec{e}_\sigma - i\vec{e}_\pi). \tag{18}
\]
The scattering factors of an atom for different light channels can finally be calculated and are summarized in table 1.

For a typical measurement, the channels with different input and output polarizations are usually not accessible as they would require polarization filters and have small scattering amplitudes. Pure \(\sigma\) and \(\pi\) polarizations do not contain any magnetic scattering. The left and right circular light channels provide therefore the highest contrast between of the magnetic configuration with a sign change between both polarizations. Equivalent change of sign is observed by switching the

### 7. Retrieving the optical constants

The strongly absorbing resonant lines (white lines) are different for various compounds and it is difficult to calculate the real absorption profile. This becomes even more evident when the Coulomb interaction between the less bound resonantly excited final states (here 3d) and the core hole left (here 2p) is not negligible.

The easiest way to handle the problem is to measure the absorption of the material using XAS. In the soft x-ray range this is usually done in the TEY mode [13]. Another new technique is based on an energy-sensitive photon detector to measure the partial fluorescence yield of a non-resonant fluorescence line from the sample. This special way avoids self-absorption or saturation phenomena present at resonant absorption lines. The inverse of this spectrum is proportional to the x-ray absorption without any corrections due to saturation or self-absorption and can be used to get quantitatively correct optical constants [82, 83].

The measured XAS signal at the relevant edges is then adapted to the slope and the edge height (jump) of tabulated non-resonant \(\beta\) values above and below the edges [76–78], providing the absorptive part of the resonant optical properties. The dispersive part is then calculated using Kramers–Kronig relations [84]:
\[
f'(\omega) = \frac{2}{\pi} \int_0^\infty \frac{\omega f''(\omega')}{\omega^2 - \omega'^2} d\omega'. \tag{21}
\]
The integral can be solved by various methods, usually by direct numerical integration or by a Fourier transformation [85].
In terms of the magnetic part the same procedure has to be done. One measures the XMCD absorption, for example for reversed magnetization with fixed circular polarization. Both curves are adapted to tabulated values as sketched above. This provides an XMCD absorption signal, which gives the dispersive part using Kramers–Kronig relations. This procedure has been done by many groups so far. To find some more examples please see [18, 35, 86].

As mentioned above, XAS is usually measured indirectly by using TEY or by fluorescence yield. Both methods rely strongly on secondary processes and suffer from self-absorption phenomena. The absorption coefficient measured is then a non-linear function of the real absorption coefficient. Corrections are possible [67] but need additional knowledge like the electron escape depth. This could be a severe problem, especially in terms of magnetism, when one wants to use the XAS signal from buried films [87].

Another way to retrieve the optical constants of some material of interest is based on measuring the grazing incidence reflectivity of bare and clean substrate like flat bars, made of the material of interest. The bars need to be thick enough, that no intensity is reflected back from the bottom of the bar, in order to prevent any type of interference phenomena which could disturb the determination of the optical constants. This is not an essential prerequisite, but it helps to simplify the determination of the constants and minimize possible conflicts. The reflected intensity follows the Fresnel equations in which \( \delta \) and \( \beta \) of the substrate are the only unknowns. By Kramers–Kronig consistent fitting of the fixed \( q_x \)-scans the quantitative correct optical constants can be retrieved [88–91]. This approach is still new and under development. Therefore a detailed comparison to the other XAS based methods is not really possible.

As shown, the detailed knowledge of the optical properties is a prerequisite to obtain quantitative chemical and magnetic profiles. On the other hand, interfaces are usually related to a change in the chemistry and therefore also in the resonant excitable states. This can alter the local chemistry of the interface atoms significantly and therefore also their optical profiles at the resonance. This has been discussed in detail elsewhere [92]. Nevertheless, if the changes are known, these can be taken into account, providing better magnetic moment determinations. Usually thickness-dependent TEY mode XMCD measurements and/or density functional theory results are a possible source of knowledge which can help solving this problem.

In summary we would emphasize that it is absolutely necessary to measure the XAS/XMCD signals together with the XRMR measurements. This is because all optical constants are indeed only effective optical constants convoluted with the present energy resolution of the beamline in its present state. In addition, the photon energy used at the synchrotron must be clearly identified and the exact values must be used. As shown for Co this is quite dramatic close to absorption edge maximum, because the dispersive part shows intense and derivative-like variations. Small errors in the absolute energy are a strong source for systematic variations in the determined magnetization profiles.

8. Matrix formalism

In reality the polarization of the light is not fixed, but varies while propagating through the film. That is for initially \( \pi \)-light one can observe \( \sigma \)-light components and vice versa as shown in section 6. This polarization transition can be handled quantitatively by the so-called magnetic matrix formalisms, introduced by Mansuripur [93] and Zak et al [52, 94]. Similar derivations have since been made in [21, 95, 96]. In the matrix formalisms, reflection and transmission at every interface is handled by \( 4 \times 4 \) matrices. In addition, the propagation of the light waves from one interface to the next, which results in phase differences and intensity modulations due to absorption, is also handled by \( 4 \times 4 \) matrices. The recursive algorithm introduced by Parratt is replaced by iterative matrix products and the solution of the corresponding linear equation system. The main advantage of this formalism is the full treatment of all partial waves including the change of the polarization state. It enables the treatment of anisotropic magnetic and NM media.

8.1. Derivation of the differential equation for light propagation

The derivation starts by transforming the differential Maxwell equations. In SI units the macroscopic Maxwell–Faraday equation and the Ampère’s law are defined as:

\[
\nabla \times \vec{H} = \frac{\partial \vec{D}}{\partial t} + \mu_0 \vec{j} \quad (22)
\]

\[
\nabla \times \vec{E} = -\frac{\partial \vec{B}}{\partial t} \quad (23)
\]

with \( \vec{D} = \varepsilon_0 \varepsilon \vec{E}, \vec{B} = \mu_0 \vec{H} \) and \( \vec{j} = \sigma \vec{E} \). Magnetic and charge distributions are neglected or are part of the dielectric tensor \( \varepsilon_r \). Multiplying equation (23) with \( \nabla \times \) leads to

\[
\nabla \times \left( \nabla \times \vec{E} \right) = \frac{\partial^2 \vec{E}}{\partial t^2} - \mu_0 \varepsilon_0 \varepsilon_r \frac{\partial^2 \vec{E}}{\partial t^2} + \mu_0 \sigma \frac{\partial \vec{E}}{\partial t} \quad (24)
\]

and is simplified to

\[
\nabla^2 \vec{E} - \nabla \left( \nabla \cdot \vec{E} \right) = \frac{1}{c^2} \varepsilon_r \frac{\partial^2 \vec{E}}{\partial t^2} + \mu_0 \sigma \frac{\partial \vec{E}}{\partial t}. \quad (25)
\]

As result we would like to obtain the final state \( t \to \infty \), when a stable wave field has been created in the sample, i.e. after the multiple scattering effects. Because we are dealing with one frequency \( \omega \) of the light the formalism is best described in the frequency domain. Applying the ansatz \( \vec{E}(r, t) = \tilde{\vec{E}}(r) e^{-i \omega t} \) leads to

\[
\nabla^2 \tilde{\vec{E}} - \nabla \left( \nabla \cdot \tilde{\vec{E}} \right) = \left( \frac{\omega^2}{c^2} \varepsilon_r - i \omega \mu_0 \sigma \right) \tilde{\vec{E}}. \quad (26)
\]

Here we have divided both sides of the equation by the time-dependent factor. Thus, in the following we only discuss the spatially dependent part of \( \tilde{\vec{E}}(r) \).
Mathematically equation (27) becomes a coupled linear differential equation system with constant coefficients. This type of differential equation is analytically solvable [97]. Its solution space can be described with plane waves of the form

$$\vec{E} = E_{0}(\vec{r}) e^{i\vec{k}\cdot\vec{r}}. \quad (29)$$

Due to the non-azimuthal dependence of the problem one parameter of the wave vector can be chosen freely

$$\vec{k} = k_{0} \begin{pmatrix} 0 \\ k_{y} \\ k_{z} \end{pmatrix}. \quad (30)$$

The scattering plane is located in the $y$–$z$-axis (figure 3) and $k_{y} = \cos \theta$ is a material independent quantity defining the angle of the incident beam.

The differential equation system results in a polynomial eigensystem

$$\begin{pmatrix} k_{y}^{2} + k_{z}^{2} - \epsilon_{xx} & -\epsilon_{xy} & -\epsilon_{xz} \\
-\epsilon_{yx} & k_{y}^{2} - \epsilon_{yy} & -k_{z} \epsilon_{yz} - \epsilon_{zy} \\
-\epsilon_{zx} & -k_{z} \epsilon_{zy} & k_{y}^{2} - \epsilon_{zz} \end{pmatrix} \vec{E}_{0} = 0. \quad (31)$$

This eigenproblem for $k_{z}$ has non-zero solutions if the determinant is zero and leads to a complicated polynomial of 4th degree of the type

$$A k_{z}^{4} + B k_{z}^{3} + C k_{z}^{2} + D k_{z} + E = 0. \quad (32)$$

but can be analytical solved, e.g. by Ferrari’s method [98]. The coefficients $A - E$ can be found in [99]. For every layer, four eigensolutions $k_{z}$ and electric fields $\vec{E}$ are retrieved.

For most anisotropic cases the eigenvectors are given by [100]

$$\vec{e}_{i} = C \cdot \begin{pmatrix} -\epsilon_{xy} (\epsilon_{zz} - k_{z}^{2}) + \epsilon_{xz} (\epsilon_{zy} + k_{y} k_{z}) \\
(\epsilon_{zz} - k_{z}^{2}) (\epsilon_{xx} - k_{x}^{2} - \epsilon_{xy} \epsilon_{xz}) \\
- (\epsilon_{xx} - k_{x}^{2} - \epsilon_{xy} \epsilon_{xz}) (\epsilon_{zy} + k_{y} k_{z}) + \epsilon_{yx} \epsilon_{xy} \end{pmatrix} \quad (33)$$

with an arbitrary scaling factor $C$. 

---

**Figure 8.** Charge terms $\delta$ and $\beta$ (left) and magnetic terms $\Delta \delta$ and $\Delta \beta$ (right) of the complex scattering amplitude of Co. The energy position of the reflectivity measurement (see below) is marked by the hollow circles and the vertical lines. (Reproduced with permission from [12], © Carl Hanser Verlag, München.)
A special case is the isotropic film where $\varepsilon = n^2 \mathbf{I}$ and leads to two degenerate eigenvalues
\[ k_{c,1,2} = -\sqrt{n^2 - \cos^2 \theta}, \quad k_{c,3,4} = \sqrt{n^2 - \cos^2 \theta} \quad (34) \]
which is a wave vector representation of Snell’s law. The solution defines two propagating wave vectors in positive and negative $z$-direction respectively. The degenerate eigenvalues span a plane of possible eigenvectors which includes the $\sigma$ and $\pi$ polarized vectors. A convenient definition for the eigenvectors is therefore (equation (17))
\[ \vec{e}_{1,3} = \begin{pmatrix} 1 \\ 0 \\ 0 \end{pmatrix}, \quad \vec{e}_{2,4} = \begin{pmatrix} 0 \\ k_{c,2,4} \\ k_y \end{pmatrix}. \quad (35) \]
This degeneracy is lifted for the magnetic case.

The electric and magnetic field inside a layer $j$ emerge from a linear combination of all elementary plane wave solutions of the corresponding fields and are given by
\[
\vec{E}^{(j)}(z) = \sum_{i=1}^{4} E_i^{(j)} \cdot \vec{e}_i^{(j)} \cdot e^{ik_{c,i}z}, \\
\vec{B}^{(j)}(z) = -\frac{k}{\omega} \times \vec{E}^{(j)}(z) \cdot \vec{e}_i^{(j)} \cdot e^{-ik_{c,i}z}
\]
(36)
with $\vec{b}_i = 1/\omega \cdot \vec{k} \times \vec{e}_i$. The solution of the differential equation is now simplified to the question of the unknown complex valued electrical field amplitudes $E_i^{(j)}$. To obtain this electric field of the layered system, interface and boundary conditions have to be applied.

### 8.3. Matrix formalism for arbitrary dielectric tensors

The interface conditions, as usually used for the derivation of the Fresnel equations introduced earlier, are defined by six equations, one for each component of the electric and magnetic field [101]. Because there are only four unknown parameters per layer, four conditions can be selected. A common choice is the continuity conditions of the in-plane components of the electric field $\vec{E}$ and magnetic field $\vec{B}$.
\[
E_x^{(j)b} = E_x^{(j+1)t}, \quad E_y^{(j)b} = E_y^{(j+1)t} \\
B_x^{(j)b} = B_x^{(j+1)t}, \quad B_y^{(j)b} = B_y^{(j+1)t}.
\quad (37)
\]
The index $t$ and $b$ define the top (t) and bottom (b) electric fields in one layer.

The system is finally described by a linear system of equations with $4N$ unknowns where $N$ is the number of total layers including the substrate and vacuum ‘layer’. These four unknowns per layer are simply the downward and upward propagating beams in the expansion of the two polarization eigenstates. The interface conditions connect neighbouring layers. This matrix form is basically a band matrix with a $4 \times 4$ block structure.

This symmetry allows us to solve the linear equation system by local inversion of the $4 \times 4$ matrices. The method is called $4 \times 4$ matrix formalism and is closely related to the known matrix methods in optics like Abeles matrix formalism [102], Mueller or Jones calculus [103] used for the calculation of a set of optical devices like lenses, mirrors and absorbers. An overview is given in [63, 64, 104].

The solution given in equation (36) can be written in form of a matrix
\[
\begin{pmatrix}
E_1^{(j)} \\
E_2^{(j)} \\
E_3^{(j)} \\
E_4^{(j)}
\end{pmatrix} = \begin{pmatrix}
E_1^{(j)b} \\
E_2^{(j)b} \\
E_3^{(j)b} \\
E_4^{(j)b}
\end{pmatrix} \begin{pmatrix}
P^{(j)}
\end{pmatrix} = A^{(j)} \begin{pmatrix}
P^{(j)}
\end{pmatrix}
\quad (38)
\]
with the propagating matrix
\[
P^{(j)} = \begin{pmatrix}
e^{ik_{c,1}d^{(j)}} & 0 & 0 & 0 \\
0 & e^{ik_{c,2}d^{(j)}} & 0 & 0 \\
0 & 0 & e^{ik_{c,3}d^{(j)}} & 0 \\
0 & 0 & 0 & e^{ik_{c,4}d^{(j)}}
\end{pmatrix}
\quad (39)
\]
which connects the top and bottom electric field of a film and the dynamical (or boundary) matrix of eigenvectors
\[
A^{(j)} = \begin{pmatrix}
e_{c_1} & e_{c_2} & e_{c_3} & e_{c_4} \\
e_{s_1} & e_{s_2} & e_{s_3} & e_{s_4} \\
b_{c_1} & b_{c_2} & b_{c_3} & b_{c_4} \\
b_{s_1} & b_{s_2} & b_{s_3} & b_{s_4}
\end{pmatrix}
\quad (40)
\]
which connects the electric field with the eigenvectors. In the isotropic case the dynamical matrix is written as
\[
A^{(j)} = \begin{pmatrix}
1 & 0 & 1 & 0 \\
0 & k_{c,2}/n & 0 & -k_{c,3}/n \\
0 & -n & 0 & -n \\
k_{c,3} & 0 & -k_{c,3} & 0
\end{pmatrix}.
\quad (41)
\]
Despite this important result for the special case of isotropic media, we derive in the remaining part the final solution of the problem for the common case of any possible matrix $A$. Note that due to the scaling properties of the eigenvectors and the linearity of the interface conditions some pre factors are omitted. By applying the interface conditions of equation (40) the combined electric and magnetic field vectors are eliminated:
\[
A^{-1} \begin{pmatrix}
E_1^{(j)} \\
E_2^{(j)} \\
E_3^{(j)} \\
E_4^{(j)}
\end{pmatrix} = A^{(j-1)} P^{(j-1)} \begin{pmatrix}
E_1^{(j-1)} \\
E_2^{(j-1)} \\
E_3^{(j-1)} \\
E_4^{(j-1)}
\end{pmatrix}.
\quad (42)
\]
Equation (42) shows these connections between the coefficients of two layers. A recursive approach eliminates finally most of the coefficients $E_i^{(j)}$ and results in
\[
\begin{pmatrix}
E_1^{(0)} \\
E_2^{(0)} \\
E_3^{(0)} \\
E_4^{(0)}
\end{pmatrix} = \left[ \prod_j A^{-1(j)} A^{(j-1)} P^{(j-1)} \right] \begin{pmatrix}
E_1^{(j)} \\
E_2^{(j)} \\
E_3^{(j)} \\
E_4^{(j)}
\end{pmatrix} = B \begin{pmatrix}
E_1^{(0)} \\
E_2^{(0)} \\
E_3^{(0)} \\
E_4^{(0)}
\end{pmatrix}
\quad (43)
where the indices (s) and (v) define the substrate and the vacuum respectively. B is the total transmission and reflection matrix of the whole layer stack, which is of course a function of the angle of incidence and is also energy dependent, due the energy dependence of the wave vector and dielectric tensor.

The term $A^{-1(j)} A^{(−1)}$ takes the role of the Fresnel coefficients and tends to the same solution as in equation (5) for the isotropic case. Therefore roughness can be applied directly, as described by Elzo et al [96].

The final form combines the remaining eight coefficients $E_{1}^{(v)}$ and $E_{2}^{(v)}$ of the boundaries by the matrix B and therefore consists of four equations. To solve these four equations, boundary conditions have to be applied. These conditions are the same as for the Parrett formalism and are defined as a given incident wave and a non-reflecting wave in the substrate.

Additionally, the final eigensolutions of the elementary plane wave solutions have to be sorted according to their propagating direction. By definition the top vacuum layer is isotropic (refractive index $n = 1$), and therefore the solution of equation (41) is taken. Hence $e_{1}^{(v)}$ and $e_{2}^{(v)}$ are the amplitudes for $\sigma$ and $\pi$ polarization of the incident beam, and $e_{3}^{(v)}$ and $e_{4}^{(v)}$ must be set to zero. The remaining four unknowns of the amplitude of the reflected beam and transmitted beam are determined by the four equations.

The reflectivity $R$, defined as the intensity of the reflected beam $I_R$ divided by the intensity of the incident beam $I_0$, is then given by

$$R = \frac{I_R}{I_0} = \left| \frac{r_{11} e_{1}^{(v)} + r_{12} e_{2}^{(v)}}{r_{21} e_{1}^{(v)} + r_{22} e_{2}^{(v)}} \right|^2 \quad (44)$$

with

$$\begin{pmatrix} e_{1}^{(v)} \\ e_{2}^{(v)} \end{pmatrix} = \begin{pmatrix} r_{11} & r_{12} \\ r_{21} & r_{22} \end{pmatrix} \begin{pmatrix} e_{1}^{(v)} \\ e_{2}^{(v)} \end{pmatrix} \quad (45)$$

$$r_{11} = \frac{B_{22} B_{31} - B_{32} B_{21}}{B_{22} B_{11} - B_{12} B_{21}}, \quad r_{12} = \frac{B_{11} B_{32} - B_{31} B_{12}}{B_{22} B_{11} - B_{12} B_{21}}$$

$$r_{21} = \frac{B_{22} B_{31} - B_{32} B_{21}}{B_{22} B_{11} - B_{12} B_{21}}, \quad r_{22} = \frac{B_{11} B_{32} - B_{31} B_{12}}{B_{22} B_{11} - B_{12} B_{21}}$$

### 8.4. Matrix formalism for magnetic dielectric tensor

The matrix formalism works for all types of dielectric tensors. However, the algorithm used to solve the problem is not numerically stable. The problematic cases are the degenerate and nearly degenerate eigensolutions of equation (40) [105]. Nearly degenerate eigenvalues result in eigenvectors which are almost linearly dependent, i.e. they are almost parallel. In this case the inverse of equation (4) could contain large values and the algorithm has to handle numbers which could differ by several orders of magnitudes. The finite precision of the number format however causes rounding errors and finally leads to wrong results. The problems can be avoided by defining a limit in which two eigenvalues are treated as degenerate and special solutions of all degenerate types.

For the magnetic case, the properties of the dielectric tensor can be used to stabilize the algorithm. The off diagonal part of the matrix is antisymmetric $\varepsilon$ and very small compared to the on diagonal elements. The first property can be used to reduce the number of cases to handle while the second can lead to a reduction of computational cost by neglecting higher order terms and by calculating an analytical solution of the inverse boundary matrix.

The handling of the degenerate cases cannot be avoided. However, the second property enables us to perform a basis transformation which is described by Zak et al in [52]. Instead of taking the tangential components of the electric and magnetic field, the electric fields perpendicular and parallel to the plane of incidence are used.

In the following, the boundary and propagating matrices for a numerically stable solution for the most common case of an in-plane aligned magnetization in the scattering plane ($\theta_M = 90^{\circ}$, $\varphi_M = 90^{\circ}$) with the dielectric tensor

$$\varepsilon = n^2 \begin{pmatrix} 1 & -iQ \\ 0 & 1 \end{pmatrix}$$

are given. The magneto-optical constant is defined as $Q = 2 \cdot (\delta_M + i \beta_M)$ and is assumed to be small in comparison to the refractive index $n$:

The dynamical matrix becomes

$$A^{(j)} = \begin{pmatrix} 1 & 0 & -iQ/k_y & 0 \\ iQ/k_y & 0 & 1 & 0 \\ -iQ/k_y & 1 & 0 & -k_z/n \\ iQ/k_y & 0 & -k_z/n & 1 \end{pmatrix}$$

(46)

(47)

with

$$k_y = \cos \theta, \quad k_z = \sqrt{n^2 - \cos^2 \theta}.$$ (48)

In the limit $Q \to 0$, equation (47) becomes the isotropic case shown in equation (41). The problem with the degeneracy is then solved. Since the algorithm is no longer used in the eigenbasis, the propagating matrix has to be transformed into

$$D^{(j)} = \begin{pmatrix} U \cos \sigma & U \sin \sigma & 0 & 0 \\ -U \sin \sigma & U \cos \sigma & 0 & 0 \\ 0 & 0 & -U^{-1} \cos \sigma & -U^{-1} \sin \sigma \\ 0 & 0 & -U^{-1} \sin \sigma & U^{-1} \cos \sigma \end{pmatrix}$$

(49)

with

$$U = e^{-i k_y h_{d}}, \quad \sigma = \frac{1}{2} k_0 n Q d \frac{k_y}{k_z}.$$ (50)

For further details and expansions to arbitrary magnetization directions we would like to refer to the original literature [52, 94]. Such a specialization of the matrix formalism to magnetic cases can be done in many ways including also different approximations. However, the basic structure of the $4 \times 4$ matrix formalism is always preserved [21, 93, 95, 96, 99].

### 9. Limitations, remarks and comparison to other techniques

The limitation of XRMR is given by the applicability of the model to real samples. High quality samples and
especially very homogeneous in-plane order are therefore the most important requirements for the heterostructure. The reflectivity signal is always averaged over the plane. Hence the roughness model is usually sufficient to represent real interfaces but it cannot differ between interface types. A chemically diffuse planar interface and a rough interface in the absence of chemical interdiffusion will provide the same results, since both atomic density profiles are close to each other [106]. Complementary methods like transmission electron microscopy (TEM) or simple surface roughness information are necessary to provide more details solving this issue. For example see the CrO$_2$/RuO$_2$ system, where TEM and AFM have shown that interdiffusion was not important [107]. Especially magnetic interfaces can be best studied if the chemical profile is quite sharp, as demonstrated in section 11.3. In this study it was very important to have nearly atomically flat interfaces, otherwise no spatial difference between the magnetic profile of the rotatable and pinned magnetic moments would have been observed. Nevertheless, even for very dramatic variation of the interface properties XRMR is able to identify the correlation between magnetic and structural profiles as shown is section 11.2.

The case of reduced symmetry, for example below cubic, does not provide a general problem to the method itself. If known any symmetry can be handled and simulated quantitatively via the $4 \times 4$ matrix formalisms discussed in section 8. Nevertheless, there are possible issues which need to be discussed. The simplest modification to a cubic system is of course the symmetry breaking at an interface. This means that at least for the atoms directly located at the interface the symmetry is always less than cubic. Another possible interface induced change is the presence of compressive or tensile stress. Both can give rise to changes to the orbital moment with respect to the bulk. The first reports on this issue are the spin reorientation transition of Co in Au as a function of Co thickness, where a clear orbital moment variation and anisotropy has been observed [108, 109]. In addition the reduced symmetry, can give rise to the so-called $T_z$ term, which causes an anisotropy of the effective spin moment and therefore an anisotropy of the magnetic optical constants [16]. In the case of strong magneto-crystalline anisotropy and unknown tilted easy axis behaviour similar implications to the magnetic optical constants can arise. The pragmatic way to handle this problem is the following: If there is no indication for an extra term in anisotropy, for example from MOKE or SQUID measurements one tries to fit the XRMR data with an isotropic optical tensor. If this does not work or one already knows that a strong anisotropy is present one adopts the optical tensor to the expected reduced symmetry and again optimizes the fits of the XRMR data, as partially demonstrated in section11.6. As discussed for XRR above, orbital reflectometry and valence sensitivity is usually strongly reduced with respect to XRMR. In addition, measurement time is much longer for neutrons (days and hours compared to 10–20 min for XRMR) and also the momentum transfer is usually strongly reduced with respect to XRR/XRMR. On the other hand the soft x-ray magnetic cross section is so strong, that XRMR is able to quantitatively detect the magnetization of sub monolayers or strongly diluted ions or even the paramagnetic moment in small applied fields [107], which is not possible by neutron reflectivity.

### 10. Fitting of magnetic reflectivity curves

The most common way to perform a quantitative analysis of the measured reflectivity curves is by fitting of layer models with the formalisms described in the previous sections. The challenge here lies in the fact that the corresponding equations are non-linear in terms of thickness, roughness and optical constants. For example, by changing the optical constants of a film the main term influenced is the wave vector $k_z$, which by itself is part of the Fresnel coefficients and the propagating wave.

Hence it is not easily predictable, what happens to the reflected beam intensity. The Fresnel coefficients at each interface and the propagating wave lead to a change in phase and amplitude but in different ways. A common mistake is to rely fully on the propagating wave for the analysis. One might argue that on a resonance the increased absorption might...
lead to fewer details, reduced reflected intensity, and reduced depth sensitivity. The latter might be true for either very thick films or strong resonances like the M₅ resonance of LaB₆. However, the general observation is a significant increase of the reflected intensity. The reason is simply the reflected intensity at the interfaces. The amplitude of the reflected intensity is proportional to the gradient of the optical constants and finally also responsible for an increased sensitivity to sample details. Reflectivity is especially sensitive to changes in the optical constants.

Another common dispute concerns the assumptions on the maximum obtainable resolution based on the incoming wavelength. In fact the resolution reachable exceeds the wavelength by far. The first trivial reason is the fact, that the wave propagates two times through the layers which leads to the effective wavelength \( \lambda = 2\pi / q_z \). The other reason regards the phase shift induced by the Fresnel coefficients. Near and on resonances the phase shift could alter between +90° and −90° and give an additional increase in depth resolution. A mapping of the resonances is therefore recommended.

The final reason for achievable resolutions beyond the wavelength limit is the fit itself. The full shape analysis allows the determination of details far beyond the Abbe diffraction limit. To give an example: to determine the thickness of a film there is no need to measure a whole thickness-oscillation of the film. The measurement contains enough details to determine the thickness also with a quarter oscillation or even less, but with a reduced accuracy. The accuracy one typically finds for the (statistical continuum) thickness of a layer is usually sub-crystalline 0.5 Å, even for typical wavelengths of the L-edges of transition metals (10–30 Å). This is one of the biggest strengths of x-ray reflectometry. This seems to be contradictory, but it is a well-known physical principal used in ellipsometry, where the thickness resolution can be much less than one nm, using visible light or even IR with a wavelength of more than three orders of magnitude higher compared to the thickness resolution achievable [104].

To improve the understanding of the model it is also advantageous to understand different regions of a measured ‘fixed-energy’ reflectivity curve. Beginning from the lowest angle the reflectivity is governed by the total reflection described by Fresnel coefficients of the surface. The propagating wave inside the sample is reduced to the absorptive part leading to an exponential decay of the light intensity inside the sample, which is also called evanesence wave. For these small angles, the reflectivity is fully surface sensitive and can be used to check the optical constants of the surface layer. This region is also influenced by the reduction of the total reflected intensity because of the limited size of the sample and the effective beam size, where only a part of the total beam is reflected by the sample while the other part passes the surface of the sample. If the geometry is known, a correction factor can be applied [111].

The intermediate angle part of the reflectivity curve is governed by a decaying intensity proportional to \( 1/q_z^2 \) (multiplied by an exponential decay caused by the roughness) induced by the Fresnel coefficients. In first approximation the thickness oscillations are just added to that line resulting from the interference of the propagating wave parts. By increasing the angle the absorptive part of the wave vector decreases. This gives rise to the assumption, that one is more sensitive to deep buried samples at high angles. For most grown samples, this effect is negligible as the change in absorption is not large enough to make any difference. The incoming light usually reaches the substrate at any angle in this region. The exception of this rule is again a very strong resonance in addition with quite thick layers.

For higher angles the more important effect is related to the real part of the wave vector \( k_z \). Larger wavelengths lead to an averaging effect of sample details. Hence, higher angles and therefore lowered effective wavelength in \( z \)-direction contain more information about the structure and especially about the interfaces of the film. Usually this effect could be observed in the increasing amount of details in ‘fixed-\( q_z \)’ or ‘fixed-angle’ energy scans measured at resonances.

Additionally with increasing angle the crystallinity of the sample becomes important. As long as the waves reflected from the atomic layers interfere randomly one can rely on the continuum approach. However if \( q_z \) is reasonable close to a Bragg peak the switch to a crystalline layered model becomes mandatory.

Keeping all of these details in mind, it becomes obvious that fitting is a difficult task and consequently a lot of research has been performed on this topic [112–114].

In the hard x-ray regime the theory of reflectometry can be further simplified by ignoring multiple scattering and absorption leading to the kinematic approximation. In first order the reflectivity is then just a Fourier transformation of the optical profile \( \delta(z) \propto \rho(z) \) where \( \rho(z) : \) electron density, which can be solved by a simple inverse Fourier transform. But due to the lack of phase detectors for x-rays, half of the necessary data is missing. Methods to perform a reverse Fourier transformation with specific constraints (quasi-model-independent) tend to be underdetermined and oversimplify the real spectra, assuming that the measured curve does not contain any experimental errors.

From the layer growth procedure parameters are usually already fixed in some range, for example the thickness with some error bars. Therefore an \textit{a posteriori} fit limiting the possible parameters to the most reasonable parameters (thicknesses, roughnesses) seems to be the best choice, at least in terms of practicability, especially in terms of finite time to find the best solution. In this way one is able to allow only the ‘most reasonable’ models for the sample by using the Occam’s razor principle.

To determine the quality of the fit, usually fitting algorithms for reflectivity provide the effective error as the sum of the squared error of every logarithmic data point \( R_{\text{meas}} \) (least-squares):

\[
\chi^2 = \sum_i \left( \log R_{\text{meas},i} - \log R_{\text{theory},i} \right)^2.
\]  

The logarithm is chosen to weight the error of different data points in a comparable way, because of the strongly decaying intensity for higher angles. It should be mentioned here that
the error definition is far from being optimal as the ‘goodness’ of the fit as defined by humans and $\chi^2$ can differ strongly. To mention an extreme example, a straight line through the middle of an oscillation is a very stable minima in terms of $\chi^2$. Therefore, the ‘best (coarse) fitter’ is usually the human intellect, who chooses reasonable boundaries for the fit parameters.

Additionally the multi-dimensional landscape of $\chi^2$ can be very ‘rough’ and contains usually a rather infinite number of local minima. Hence simple downhill algorithms like methods based on the Nelder–Mead algorithm [115] are not optimal but usually work fine as the final fitting step. Fast converging methods like the Levenberg–Marquardt algorithm are not optimal as they usually need derivations of the spectra regarding the fit parameters, which have to be determined numerically and they easily stuck in local error minima.

To overcome these non-linear problems computer science has developed some heuristic optimization algorithms based on natural phenomena such as simulated annealing and genetic algorithms.

The basic idea of annealing methods is the simulation of the annealing processes in crystals to optimize their crystallinity and lower their total energy. To reproduce this effect here an effective fluctuation term is added to the fitted parameters. While minimizing the fit with an iterative downhill method the parameters fluctuate jumping over small error minima and staying in good minima. The fluctuation is reduced each iteration, so that the probability jumping out of a good error minima is reduced [115].

The second heuristic optimization algorithm is based on the mimicking the process of natural selection. An initial population is chosen with different fitting parameters constrained only by the user to a reasonable configuration space. The fitness, namely $\chi^2$ is determined for each candidate. In the selection step only the future parents with the best fitness survive. They are used to determine the new generation by mixing and mutation processes of the fit parameters [116–120].

The advantage of the second method is the sampling of the whole configuration space and the great parallelizability of the algorithm on modern computers. A population of several tens of thousands of individuals are possible to simulate within reasonable time. Therefore the genetic algorithms seem to be the favourite method.

To focus on the magnetic part of the reflectivity, the asymmetry between the intensities of left circular $R_l$ light and right circular $R_r$ light is usually calculated by

$$ A = \frac{R_l - R_r}{R_l + R_r}. \quad (52) $$

The denominator ensures that the asymmetry is always normed to the absolute intensity of the signal, otherwise less important high reflectivity parts at lower angles are over-represented. Another benefit of this equation is the reduction of errors introduced by the fits of the absolute intensity as the normalized difference already eliminates such errors. A perfect quantitative analysis of the absolute reflectivity intensity is therefore not required.

To summarize, the step by step guide to analyse the reflectivity measurement is

(1) Fit the measured XAS data of the resonances to the tabulated off-resonant tables of the refractive index and determine the XMCD signal. Perform a Kramers–Kronig transformation to determine the real part of the XAS related refractive index and of the XMCD signal.

(2) Start to model the film with a good guess of the heterostructure, which is usually known from the details of the growth process. Parameters like the thickness can be often determined by calculating the approximate thickness from the oscillations of reflectivity spectra ($t = 2\pi/\Delta q_z$).

(3) Perform a manual coarse fit of the structure. Determine physical relevant fitting intervals for the different roughnesses and thicknesses and experimental parameters like energy resolution, angular resolution and the scaling factor for the measured intensity.

(4) Use all fitting algorithms described above to determine the best parameters for the sample. Rely also on off-resonant reflectivity curves, where the refractive index is known from tabulated values.

(5) Check the reliability of the optical constants. Significant contaminations, like carbon hydrates or water on top could alter the shape of the reflectivity curve and must be included as an additional top film.

(6) After the structural model is converged and a satisfying quality has been achieved, one starts to assume a magnetic model usually beginning with a homogenous magnetization of the FM layers and fitting of the magnetic asymmetry signal. Because of speed issues one uses first the (magnetic-) Parratt formalism and for further model improvements at the end the advanced 4×4 matrix formalism. Usually the magnetic structure profile will differ from the chemical profile, for example due to magnetically dead layers or enhanced saturation magnetization at the interfaces. Therefore structural and magnetic models must be independent with each other, which can be achieved by an intelligent slicing of the chemical model. In order to allow arbitrary functions as magnetization profiles the sample can be sliced into very small layers (multi-slicing). A parameterized separate magnetic model relying also on the same parameter types like the chemical model (thicknesses, roughnesses) is preferable.

In our case we have successfully developed a software package called ReMagX, which has implemented all the described algorithms and fitting procedures and is available online [65]. The program is very versatile, speed optimized and contains a graphical user interface. For example, we can nicely switch between precisions and speed by changing from matrix to Parratt algorithm. Our code is very fast allowing multilayer segmentation, allowing complex chemical profiles and arbitrary magnetic profiles. Also the presence of variety of fitting algorithms and methods provides optimal algorithms for different parts of the fitting problem.

11. Examples

In this section we provide a short overview on the exceptional possibilities of magnetism related soft x-ray scattering in the
Figure 9 shows the experimental NM resonant reflectivity curves of a Ce/Fe multilayer system. Clear superstructure Bragg reflections are directly observable. For the magnetic case, the magnetic asymmetry ratios as defined above are plotted as a function of photon energy in figure 10, where the angle has been chosen at various superstructure Bragg peaks, indicated by the diffraction order. Without going into details of the analysis, the chemical profile has been extracted from the NM spectra, and the magnetic counterpart from the magnetic asymmetry.

Multilayer systems provide the unique opportunity to extract NM and magnetic multilayer information in an analytical way, averaged over all accessed individual layers [19]. Here, simply the position, width and the height ratios of the magnetic and NM superstructure peaks give directly the periodicity and the magnetic profiles. This is also valid for the magnetic asymmetry ratio.

Due to the overall averaging effect of multilayers, thickness and roughness variations and their dependence on the distance of an individual layer from the substrate cannot be addressed analytically. On the other hand, these variations are often important, especially in systems with large lattice mismatch at the interfaces. Furthermore, diffuse XRMS in the soft x-ray region can be applied to investigate correlations between chemical and magnetic roughness in thin films and multilayers [122, 123]. Magnetic resonant reflectivity measurements in the soft x-ray region have been performed [20, 23, 24, 124, 125] to measure the energy dependence of the reflected intensity for parallel and antiparallel alignment of the photon helicity and the direction of the magnetization.

11.2. Hard x-ray reflectivity for single layer systems: induced magnetism and roughness at Pt/Co

Platinum is close to the Stoner criterion of a ferromagnet and tends to be FM by itself in contact with another ferromagnet. Pt is also a heavy metal with higher nuclear charges, compared to the 3d TM, which gives rise to an enhanced spin–orbit coupling. This is the reason why systems like FePt in its layered L10 phase provide high magneto-crystalline-anisotropy, and making this system promising for future high density recording media [126]. In order to study this interfacial behaviour, bilayer systems like CoPt or FePt have been studied, providing better understanding of the induced interface coupling [127]. One important question is the strength of the induced FM moments in Pt and the typical distance where FM is still present. This usually cannot be investigated by conventional methods like SQUID, because the small induced magnetic moment in Pt cannot be separated from the strong FM signal of the 3d TM. If ever a small additional fraction, related to Pt, is observed one cannot distinguish this additional part from an interface induced enhanced magnetism moment in the TM. Therefore, XRMR is an ideal tool to study in an element-specific manner the magnetization of Pt, which has been performed at the Pt L3 edge [18, 86].

Figure 11 shows our results performed at different bilayers of Pt/Co where, due to various buffer layers and preparation procedures, the roughness of the CoPt interface has been varied systematically [86]. Figure 11(a) shows the NM and non-resonant reflection profiles, measured with a Cu anode. Black circles are the measured intensities and the red lines are the related Parratt-based fits, providing excellent agreement and therefore accurate structure profiles and interface roughness.

Figure 11(b) shows the magnetic asymmetry ratio combined with the magnetic simulations. Again, nearly perfect agreement between experiment and simulation is achieved. For both simulations, the chemical and the magnetic profiles have been fitted in order to give best agreement with the experimental results. In figure 11(c), the blue and magenta reflection mode. This section is not intended to give a complete overview on the full history of XRMR, but it will provide an idea what kind of unique scientific goals can be achieved using this technique.

11.1. Hard x-ray reflectivity for multilayer systems: CeFe

We start with the analysis of multilayer systems. The reason for this is given in the presence of the Bragg peaks related to the superstructure of the multilayer. As most readers are more familiar with Bragg peaks, this approach seems to be more intuitive. Superlattice reflections in multilayers contain information of layer-averaged properties, for example magnetization depth profiles for one component of the whole multilayer system [19, 20, 121].
curves represent the related chemical profile of Co and Pt respectively, while the orange bars represent the magneto-optical Pt profile. Here the roughness and therefore the profiles have been approximated by the multi-slicing method, which are directly visible as the depth dependence of the vertical bars in figures 11(c) and (d).

The magnetic profiles show strong variations as a function of roughness—the induced Pt magnetization tends to reach deeper into the Pt layer in conditions where the Co ‘reaches’ chemically deeper into Pt. From XMCD sum rules and reference sample normalization, the relation between the magneto-optical effect at the Pt L3 edge and the induced Pt magnetic moment can be used to provide absolute Pt magnetization values. By normalizing the magneto-optical profile to the chemical profile, the depth-dependent magnetic moment per Pt atom has been determined as shown in figure 11(d). The result shows that Pt magnetic moment tends to saturate at about $0.21 \mu_B$, when the coordination with Co is high enough. This decreases gradually with reduced Co concentration in a non-trivial manner. We would like to mention that the Pt L3 edge XRMR at about 11.565 eV gives an accessible momentum transfer range up to 2.5 nm$^{-1}$ which is accessible only at very grazing incidence conditions up to about 1.1$^\circ$.

It should be emphasized that other magnetic depth profiling methods like neutron reflectivity are only sensitive to the total magnetic moment. In this case neutrons cannot distinguish between the small magnetic Pt moment or an enhanced Co moment or just slightly more Co which is positioned more deeply inside the Pt due to roughness and/or diffusion.

Another question to be answered is a possible change in the Co magnetization with Pt on one side and NM Cu at the other. Figure 12 shows the XRMR results from the 0.7 nm interface roughness sample already introduced before. This sample has a Cu layer below Co. In order to probe Co we switch to the soft x-ray range enabling the Co 2p $\rightarrow$ 3d excitation at the Co L$_{2,3}$ edges. This edge reveals a very strong NM (XAS) and magnetic (XMCD) resonance effect. For the simulation and fitting of the stack we use the optical properties of Co introduced in the optical properties section and shown in figure 8. Again, the magnetic profile can be nicely fitted. The interface position is indicated by the dotted line in figures 12(c) and (d), where the concentration is half/half of the two constituents. Two effects are directly visible. In the case of the Cu interface a significant reduction of the Cu moment is detected, while on the Pt site the Co moment is enhanced.

This example shows that even small magnetic moment variations can be detected and investigated quantitatively by means of XRMR. Absolute changes are traceable, if the optical properties and their XMCD effect with respect to the absolute magnetic moments are well known. Also in GMR systems XRMR was able to provide detailed knowledge on the interface magnetism, which is necessary to understand, for example, the nearly vanishing GMR effect in the high spin polarized CrO$_2$/RuO$_2$/CrO$_2$ layer system [107].
11.3. EB and rotatable and pinned magnetic moments in the MnPd/Fe system

A MnPd/Fe bilayer is a typical metallic EB system [29]. The antiferromagnet MnPd has a Néel temperature at about 500 K and crystallizes in the CuAu-I type structure. Originally MnPd has been identified as an archetype for compensated and uncompensated AFM interfaces depending on the chosen growth direction of the AFM, which can be controlled...
as we will see later on [35]. This allows us to study pinned and nearly perfect interfaces with extraordinary small roughness, a of this oriented MnPd film grown on MgO (0 0 1) will be presented with roughness phenomena. In the following, an unpinned uncompensated magnetic moments, not entangled left inset in figure13, the NM reflectivity curve and its quality structural and magnetic profiles. In the upper previous sections, these curves are necessary to obtain high quality changes significantly for the different growth direction, by the substrate temperature. Unfortunately, the interface quality changes significantly for the different growth direction, preventing somehow a detailed analysis of the EB dependence for ideal compensated and uncompensated interfaces of the AFM [128]. Nevertheless, a-axis oriented MnPd/Fe provides nearly perfect interfaces with extraordinary small roughness, as we will see later on [35]. This allows us to study pinned and unpinned uncompensated magnetic moments, not entangled with roughness phenomena. In the following, an a-axis oriented MnPd film grown on MgO (0 0 1) will be presented and discussed. 

Figure 13 shows reflectivity curves and asymmetries of this a-axis oriented MnPd/Fe film. As discussed in previous sections, these curves are necessary to obtain high quality structural and magnetic profiles. In the upper left inset in figure 13, the NM reflectivity curve and its simulation are shown, performed resonantly at the Mn L3 edge. The interference pattern is quite simple and easy to understand intuitively. Only one periodicity is observable, which corresponds only to reflections of a single layer. The periodicity fits nicely to the total thickness sum of the Fe layer with the capping Pt on top. The very thick MnPd layer below provides strong absorption at the Mn L3 edge, which guarantees that no significant intensity from the MgO/MnPd interface contributes to the interference pattern. This is a nice example of a desired ‘switching-off’ of lower interfaces, as already discussed in the introduction to real systems above in figure 6. The corresponding reflectivity fit provides the chemical profile with enhanced Mn sensitivity, which is shown in figure 14(a) as the dispersive and absorptive part of the optical constants (red and black lines). This nicely shows a very narrow transition from the MnPd to Fe with a FWHM of only 0.4 nm. In addition, figure 14(b) shows a sketch of the sample stack, where the MnO substrate has been omitted.

As discussed in the XMCD section, circular polarized light and a change in the scalar product of the axial vector of the circular polarization and the magnetization vector provides changes in the optical properties, if the incoming photon energy is tuned close to the Mn L2,3 edges. In order to be sensitive to the rotatable part of the sample magnetization one simply switches the sample magnetization by an external field and leaves the sample orientation and the light helicity constant. In this case only the scalar product between the light helicity and the rotatable moments changes and provides a magnetic anisotropy. This is sketched in the upper right inset in figure 13. The related magnetic reflection asymmetry and fit results are shown in the upper part of figure 13, thus providing the magnetic profile. The Mn magnetization profile of the rotatable magnetic moments is shown in figure 14(a) as the magneto-optical profile (green and blue dashed lines). From the comparison of the blue dashed with the red straight line one clearly sees that the Mn atoms in contact with the Fe are rotating. In addition the sign of the absorptive part of the magneto-optical constants is positive, indicating an antiparallel magnetic orientation and interaction between Mn and Fe. This gives a clear and simple picture: the Mn atoms with a high Fe coordination are strongly exchange coupled antiparallel to the Fe. This directly tells us that the simple exchange coupling model of the EB has to be modified to magnetically strongly coupled ions, which are originally related to the AFM, and now behave like a FM with antiparallel orientation to the external field.

This example shows well the very strong sensitivity of XRRM to magnetic configurations. A clear and nearly noise free magnetic interference pattern, as shown in the upper part of figure 13, can be observed, which is only related to fractions of a monolayer of Mn that is only present at the interface. For details of the optical constants and the validity of the results please refer to [35].

In order to get an effect from the pinned moments, one has to change the scalar product between the pinned moments and the light helicity, while the rotatable moments have to fix their scalar product with the light helicity. This can be done by flipping the external field (which flips the rotatable moments)
Figure 14. Shown are the $z$-dependent optical profiles for the real and imaginary parts $\delta$ and $\beta$ separated in (a); red and black lines represent the NM optical profile based on the chemical profile. The green and blue dashed lines (magenta and cyan dashed–dotted lines) are the magneto-optical constants for the rotatable (non-rotatable) uncompensated Mn moments respectively. (b) Sketch of the layered stack. (c) Element-specific field dependent magneto-optical curve along and perpendicular to EB direction and for Fe and Mn. (Reproduced with permission from [36].)

and also flipping the light helicity at the same time. In this case the scalar product between the rotatable moments and the light helicity is always parallel (or antiparallel) while the pinned moments change from parallel to antiparallel orientation with respect to the circular polarization. This is sketched in the lower right inset in figure 13. Therefore, only the pinned moments provide an asymmetry as expressed now by an extended asymmetry function,

$$A = \frac{R^{++} - R^{--}}{R^{++} + R^{--}}. \quad (53)$$

Here the first ± in the superscript is related to the change in sample magnetization, while the second one represents the change in light helicity.

The related so-called pinned asymmetry together with the fit is shown in the lower part of figure 13, which is about to be 5 times smaller compared to the rotatable moment asymmetry shown above. The related magneto-optical profile is also plotted in figure 14(a) as the corresponding dashed–dotted magenta and cyan lines. Comparing the absorptive profile between the rotatable and the pinned moments, clear changes are present. As already suggested by the intensity, the rotatable moment profile has a much stronger effect with respect to the pinned one. We would like to mention here that there is no exact one to one correspondence between the MO-profile height and the asymmetry observed, because the interference intensity is also convoluted by the magnetic roughness, which is here just the width of the profile. Nevertheless, a clear change in width is observable, showing that the pinned magnetic moments related to EB are not a simple interface phenomenon and rather extend into the AFM layer over a 3–4 times larger range compared to the interface roughness. One also sees an opposite orientation of the uncompensated magnetic pinned moments with respect to the rotatable ones. This means that on average the pinned uncompensated moments are parallel oriented to the bias field saturation direction.

In figure 14(c), element-specific hysteresis loops measured in reflection mode are shown. This is more precisely described as the external field dependent magneto-optical asymmetry at a fixed energy and fixed angle. These measurements have been performed by choosing energies close to an XMCD-sensitive edge, here the Fe and Mn L3 edges. In addition one has to choose an appropriate angle of incidence, usually selected by a closer look to the asymmetry curve shown for example in the upper (rotatable) part of figure 13. Our case shows nicely that the EB is directly observable in the Fe L3 loop. Also, rotatable Mn moments rotate exactly in the same way as the Fe does on top. For the perpendicular alignment of the EB direction with respect to the scattering plane no EB is observed, as expected, and indeed also no uncompensated magnetic moments are observed as evidenced by the almost absence of a pinned asymmetry. This is shown by the green curve in the lower part of figure 14(c). We would like to mention here that the sign of the observed hysteresis loops is completely arbitrary depending on the momentum transfer of the magnetic asymmetry curve. As the sign changes in figure 13 also the hysteresis loop can go from high to low or from lower to higher signal. It is also important to mention that the exact momentum transfer can also change the shape of the element-specific magneto-optical hysteresis loop due to magnetization related changes in the interference phase. Usually one chooses a momentum transfer where an energy-dependent scan with fixed momentum transfer (constant $q_z$ scan) provides a profile close to a L2,3 absorption edge, slightly modified by the wings
related to the dispersive part. This procedure is usually enough to get reasonable ‘hysteresis loops’. Nevertheless, in principle this complex interference modulated shape variation of the ‘element-specific hysteresis loops’ can also be simulated and fitted. This provides reliable determination of observable differences between element-specific XRMR-based hysteresis loops and those from complementary methods like SQUID or VSM.

11.4. EB in the Py/CoO system

Here we describe an example performed mainly by helicity switching of the incoming soft x-ray beam by Roy [34], Blackburn [129] and co-workers, showing the presence of rotatable and pinned moments for the Permalloy (Py) CoO EB system. In contrast to the system in the previous section, the AFM is now a classical oxide CoO, which has also played the key role in the first reported EB system Co/CoO [27]. In this case also oxidization of the metallic Py layer on top must be taken into account. The nice advantage of these systems stems from the strong temperature dependence of the EB effect. While the EB vanishes almost entirely at RT, it is quite prominent at about 150 K. Again this property can be monitored by the two hysteresis curves reproduced in the upper left part of figure 15 [34]. For RT measurements, the magnetic asymmetry has been determined and fitted using the distorted Born wave approximation (DBWA) [21]. Similar to the results shown before, a clear signature for a non-vanishing magnetization in the AFM has been observed. Here a thin layer of Co, at the interface to the FM, is also ferromagnetically ordered with respect to the Py layer. In order to study the pinned magnetic moments the sample has been field cooled to 150 K, giving a clear signature for EB (see hysteresis loop in figure 15). This should be also related to pinned magnetic moments, in particular the Co moments. Therefore, the authors performed a magnetic XRMR measurement where the magnetic asymmetry stems from changes in the light helicity, while the magnetization was held fixed, but parallel and antiparallel oriented with respect to the beam direction. By flipping the external field the relative orientation between the pinned and the rotatable moments could be changed from parallel to antiparallel. The total magnetic moment is therefore enhanced for parallel and reduced for antiparallel orientation. The related XRMR measurements for flipped light helicity are presented in the upper right part of figure 15 and clearly show deviations in the strength of the magnetic effect. The resulting Co interface magnetization is also shown in the DBWA-based fit results presented in the lower right part of figure 15.

Figure 15. Left: upper part shows a TEM viewgraph of a Py film on top a CoO layer and the RT (150 K) hysteresis loops with vanishing (present) EB. Magnetization profiles determined from different XRMR measurements performed at the Ni and Co L2,3 edges. (Reproduced with permission from [34], copyright 2007 American Physical Society). Right: upper part shows the asymmetry signals for switched light helicity with the magnetization held fixed parallel and antiparallel to the photon beam direction. Lower part shows the magnetization profiles related to the upper two XRMR curves. (Reproduced with permission from [129]. Copyright 2008 American Physical Society.)
of the difference between the red and the black curves is related to the pinned moments, simply added (subtracted) to the rotatable part.

Thus, the order of magnitude for the ratio of the uncompensated pinned to the rotatable moments in the AFM studied here in the Py/CoO system is close to the results observed for the MnPd/Fe systems presented in the previous section.

11.5. Interface magnetism in SC/FM heterostructures

In this section we will briefly demonstrate the capability of XRMR for SC/FM heterostructures. This is an important issue in modern solid state physics, due to the strong interaction of magnetic and electronic degrees of freedom. Especially the interdiffusion of electrons at the SC/FM interface is a nice playground for detailed studies of the interaction between such systems.

In the system shown here, the general question was: is there a SC-induced superconductivity in the FM, or just a reduction of the magnetic moment in the FM top layer, or opposite, do we find a FM layer in the SC? Here we show in figure 16(a) the XRR results performed at a La$_2$Ca$_{3/2}$MnO$_3$ (LCMO) film optimally doped to obtain FM with a Curie temperature around 240 K, where YBa$_2$Cu$_3$O$_{7-\delta}$ (YBCO) has been grown on top [50]. SrTiO$_3$ (STO) with (0 0 1) orientation has been used as a substrate. In the XRR curves no visible temperature dependence is observable. On the other hand the XRR results were able to be fitted almost perfectly, providing the optical profile shown in figure 16(b). It is worthwhile to note that the STO/LCMO interface is nearly perfect with a roughness of only 0.1 nm, while the LCMO/YBCO interface roughness is still low but about one order of magnitude higher.

Figure 16(c) shows the corresponding Mn based magnetic XRMR asymmetry. Again, a nearly perfect fit has been obtained. Figure 16(d) shows the comparison between the magneto-optical and the NM (structural) optical profiles, normalized to each other. A clear shift of the magnetic profile with respect to the chemical profile can be observed. This indicates a magnetic dead layer of Mn close to the YBCO interface. Indeed, it could also be that a thin layer of an AFM LCMO phase is present, which is in terms of XRMR measurements equivalently NM.

Interestingly, no low temperature dependence, especially at the superconducting phase transition, could be observed at the LCMO/YBCO interface. In addition, the XRMR results also provide magnetic profile information at the bottom, i.e. the STO/LCMO interface, and reveal a peculiar anomaly, as
shown in the left part of figure 17. A strong increase of the Mn magnetization can be observed, which is about double the height of the saturation magnetization. The temperature dependence of this effect is shown in the right part of figure 17. Here the temperature-dependent XRMR curves are shown, together with the simulation based fits. Again a nearly perfect agreement between the fits and the XRMR asymmetry could be achieved.

Above the XRMR curves the height of the anomaly from the left part is plotted. This temperature dependence is nicely correlated to a structural phase transition of the STO substrate from a cubic to a tetragonal, which appears below 105 K [130–133].

Indeed the results shown here and similar results have been discussed controversially in the literature. Growth mode and quality related strain, strain relaxation, interface diffusion, electrical conductivity, layer thickness, and also the layer stacking sequence are important issues in this field. It is beyond the scope of this article providing deep insight into this interesting part of solid state physics, but it shows that XRMR measurements provide unique and element-specific insight into interface dominated physics.

11.6. Perpendicular magnetic EB coupled system

So far only in-plane magnetized systems have been shown here, related to the inherent shape anisotropy present in ultrathin film systems, which usually forces the magnetization into the film plane. On the other hand, many interesting magnetic materials like FePt, CoPt or FM with layer thicknesses close to one atomic layer exhibit perpendicular magnetic anisotropy. Also, materials with special microcrystalline structure are materials of choice for high density hard disc recording technologies [134, 135].

As we have already discussed in section 2, the magneto-optical effect for circular polarized light is proportional to the scalar product between the corresponding magnetization of interest and the direction of the incoming photon beam. In terms of XRMR measurements for in-plane oriented sample magnetization, with projection along the photon beam direction, one gets a strong magnetic asymmetry effect at lower incidence angle conditions, while the asymmetry clearly vanishes at normal incidence because of the vanishing scalar product. This behaviour changes completely in the case of perpendicular magnetized samples. Here the scalar product becomes zero at grazing incidence and has its maximum at normal incidence. This is described in detail in the universal magnetic scattering tensor equation (13) (for details of a two-dimensional basis we refer to [41]).

In principle, this behaviour of the scalar product is the only important change in the simulation of the reflectivity curves for perpendicular magnetized samples. It has been demonstrated by Tonnerre and co-workers on a perpendicular magnetized PtCo film as an FM grown on Pt(1 1 1) substrate, and a multilayer structure of [NiO(3ML)/CoO(3ML)]3 acting as an antiferromagnet [41]. This system shows nice perpendicular anisotropy, and also perpendicular EB behaviour below 320 K. figure 18 shows the XRMR results performed at two different Co L3 edge energies and in the lower part of figure 18 the corresponding magnetic asymmetries are presented. Only

4 This is also the case for many in-plane oriented systems at grazing incidence conditions, especially with non-magnetic protective layers on top. In this case the XRMR effects are reduced due to total reflection conditions.
Figure 18. Upper part: experimental and calculated XRR intensities, for left and right circular polarized light, performed at two different Co L$_3$ edge related photon energies. Lower part: experimental and simulated asymmetry ratios utilizing various magnetic profile models. Only the red coloured pnn0 model reproduces the experimental results. (Reproduced with permission from [41], copyright 2008 American Physical Society.)

The red curves provide good agreement between the magnetic model and the experimental results.

The major result is the following: the Co magnetic moment is parallel or FM out of plane aligned to the external field in the CoPt layer with a slightly enhanced magnetic moment of 2.07$\mu_B$ with respect to bulk Co [60]. Then the first Co layer in CoO is 0.3 nm in thickness and also parallel aligned (p), with a subsequent antiparallel aligned Co inside the CoO with a thickness of 0.7 nm (n). The remaining slice of 0.2 nm has no net magnetization (0).

12. Concluding remarks

Without doubt, reflectometry and especially XRMR are very promising but also very sophisticated techniques. The combination of x-ray circular magnetic dichroism and reflectometry is the perfect way for non-destructive element-specific magnetic depth-analysis. Reflectometry is thereby one of the rare x-ray techniques in which a quantitative full shape analysis of the measured spectra is carried out. Because of this quantitative analysis, it is a unique and powerful tool, providing quantitative results. Key ingredients to the success of reflectometry are the large dynamic range in intensity of more than 7 orders of magnitude and an exceptional sensitivity to the electronic properties.

This sensitivity allows not only the analysis of magnetic configuration. Transition metal oxides (TMOs) bear a great variety of effects as their correlated electronic structure induces many interesting phenomena like charge, spin, and orbital orderings dependent on temperature, external fields, strain or composition. Quantitative and structural analyses of those physical phenomena are the key for understanding macroscopic properties like electrical and thermal transport, magnetism, optical responses, metal–insulator transitions and superconductivity of these materials. Especially the reconstruction phenomena at interfaces of TMOs could provide new ground for completely novel physical phenomena [42–44]. Some of these phenomena have already been demonstrated, including 2D-electron gases [136–138], interface superconductivity [139] or interface magnetism [140]. Resonant reflectometry is able to give new insights into these systems. Based on resonant reflectometry the orbital polarization of the interfaces of LNO/ LAO superlattices were determined quantitatively [46, 47].

At this final end we would like to give a probably too short comparison to neutron reflectivity experiments, where XRMR provides dramatic advantages. The most important point is the element and symmetry selectivity, which enables a quantitative determination of sample magnetization, even in the background of strong magnetic counterparts. Especially
the examples shown in figures 13, 16 and 17 have been performed in about 20 min with a quite broad momentum transfer range of about 0.3–0.4 Å⁻¹. Conventional neutron reflectometry results are much smaller in momentum transfer and take considerably longer, especially with the same signal to noise ratio. Another advantage of x-rays is the opportunity to choose between structural and magnetic effects, simply by changing to the appropriate light polarization. On the other hand, neutron data can be simulated more easily by utilizing tabulated neutron optical constants, while the XRM results critically need the spectroscopic determination of the optical properties. In addition, spin flip channels can be evaluated, enabling further directional magnetization information.

As described in this review article, the exact knowledge of the optical constants is an important prerequisite for the XRM analysis. In addition, the majority of XRM results have been obtained at fixed photon energies with varied momentum transfer. On the other hand one can perform energy-dependent XRM measurements at constant momentum transfer. These two approaches can be combined, producing a 3d landscape, or reflectivity map. In terms of known optical constants this introduces a dramatic increase in over determination, which allows solving of the scattering phase problem and gives an exact solution [114]. This allows in principle the partial determination of changes in the optical properties and therefore XAS/XMCD spectroscopy of buried systems performed in a non-destructive way. The combination of Kramers–Kronig relations with the reflectivity landscape provides the possibility of an extra constraint between the dispersive- and imaginary parts, which increases the significance and reliability of such XRM-based new spectroscopy results. This method is indeed unique and very powerful. It allows in principle the spatial determination and separation of spin and orbital magnetic moments.

One can expect a greater variety of physical effects analysed quantitatively with resonant reflectivity in the future, including the magnetism and complex interface phenomena in solid state research.

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