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Faraday rotation spectra at shallow core levels: 3*p* edges of Fe, Co, and Ni

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Abstract. We present magneto-optical (MO) Faraday spectra measured at the 3p edges of Fe, Co, and Ni. A polarization analysis of the final state of the transmitted radiation is employed to determine the Faraday rotation at these edges. The MO effect becomes resonantly enhanced at the 3p edges and accordingly large values for the intrinsic rotation constant k of 2.2×10^5 , 1.5×10^5 and 0.8×10^5 deg/mm for the three ferromagnetic elements Fe, Co, and Ni, respectively, have been measured. These values are large and comparable to those observed at the 2p edges. This remarkable feature emphasizes the importance of the exchange (EX) splitting over the spin–orbit (SO) interaction of the core levels in determining MO effects in the extreme ultra-violet (EUV) region. From these measurements the complete set of MO constants at the 3p edges has been derived.

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1. Introduction

Magneto-optical (MO) spectroscopy using polarized soft x-rays [1] allows element selective investigations due to the specific energies of the absorption edges, which are characteristic for every element. Furthermore, a pronounced enhancement of the transition probabilities at the onset of absorption edges occurs, which leads to MO responses two orders of magnitude larger than those in the visible range. These features are mainly responsible for the recent popularity of x-ray MO spectroscopies. Currently exploited experimental techniques are the x-ray magnetic circular dichroism (XMCD) [2, 3], the x-ray magnetic linear dichroism (XMLD) [4, 5], the resonant magnetic reflectometry [6, 7], the x-ray Voigt effect [8] and the x-ray Faraday effect [9, 10]. These MO effects are observable in various geometries and modes such as transmission, reflection or absorption. The size of the MO effects relates both to the spin–orbit (SO) interaction and exchange (EX) interaction of the core levels *and* of the valence bands of the transition metals or rare earths [11].

So far most of the magneto-x-ray spectroscopic investigations were carried out at the 2p absorption edges of 3d transition-metal atoms. Here the SO coupling yields a splitting of the $2p_{3/2}$ and $2p_{1/2}$ states of $\Delta_{SO} \sim 13-16 \text{ eV}$, which is larger than the EX interaction of the 3d valence states ($\Delta_{EX} \sim 1-2 \text{ eV}$) [12], and much larger than the EX interaction of the 2p core levels ($\Delta_{EX} \sim 0.3-0.9 \text{ eV}$) [13]. At core levels with lower binding energies e.g., at the 3p states, however, the SO splitting is reduced by about a factor of ten, while the EX splitting of the 3p states increases, making these interaction energies comparable ($\sim 1 \text{ eV}$) [14]–[17]. Thus, a comparison of data obtained from MO spectroscopies at the 3p edges with those measured at the 2p edges might give insight into the relative role played by the SO and EX interactions in the MO effects observed at the different edges.

Up to now only a few x-ray MO investigations have been carried out at the transition metal 3p edges. These include measurements of the transversal magneto-optical Kerr effect (T-MOKE) [18]–[20], the XMCD [21], dichroism in reflection with circular [22, 23] and linear [24] polarized light, and the so-called magnetic linear dichroism in the angular distribution (MLDAD) of photoelectrons [25]. Two experiments on the Faraday rotation of Ni and Co at the 3p edges were reported [26, 27]. However, an exhaustive investigation of the Faraday effect providing the complete set of MO constants for ferromagnetic Fe, Co, and Ni is missing. These MO constants are those parts of the index of refraction *n* describing the modification of the refractive index due to the macroscopic magnetization.

In this paper, we present a full set of Faraday rotation data measured at the shallow 3p core levels of Fe, Co, and Ni. From these Faraday data we determine the complete set of MO constants

for the EUV region. An important result of our investigations is that the intrinsic Faraday rotation constants at the 3p edges are found to be *as large as* those at the 2p edges, which emphasizes the enhanced importance of the role played by the EX interaction over the SO interaction at the 3p edges.

In the following, we first provide the theoretical model underlying the MO Faraday effect and specify the formalism used for deriving the intrinsic MO constants. In section 3, we outline the employed experimental technique. Our experimental data are presented and analysed in section 4, in which we also compile the complete set of deduced MO constants for the transition metal elements.

2. Description of the Faraday effect

The starting point for a description of the Faraday effect is the so-called polar geometry, which is adopted in standard Faraday measurements [28]. The sample's magnetization as well as the incident light are both normal to the sample surface. The incident linearly polarized light can be decomposed into two circularly polarized waves, having equal amplitude, but opposite helicity. These two modes match continuously to the two possible, circularly polarized eigenmodes in the magnetic material. The propagation of the radiation through the material is described by the complex refractive indices n_{\pm} for the two circularly polarized eigenmodes:

$$n_{\pm} = 1 - (\delta \pm \Delta \delta) + \mathbf{i}(\beta \pm \Delta \beta), \tag{1}$$

where the subscript +(-) refers to the parallel (antiparallel) orientation of the photon helicity with respect to the magnetization direction of the sample. The real part ($\delta \pm \Delta \delta$) describes the dispersion of the radiation, while the imaginary part ($\beta \pm \Delta \beta$) describes the absorption. The effect of the magnetization is given by the respective MO constants $\Delta \delta$ and $\Delta \beta$, which are nonzero for magnetized ferromagnetic materials.

The magnetic contribution $\Delta\beta$ leads to a difference in the absorption of left and right circularly polarized light components. $\Delta\delta$ leads to a difference in the phase of the two circularly polarized waves traversing the sample. As a results, the initially linearly polarized light ($P_{\text{Lin}} = 1$) becomes elliptically polarized ($P_{\text{Lin}} < 1$) behind the material, giving rise to the Faraday ellipticity ε_{F} . The polarization plane is rotated by φ_{F} (Faraday rotation) (shown schematically in figure 1).

We measure the Faraday effect in a geometry differing from the pure polar geometry (see figure 1). The light impinges not normally on the surface, but at an oblique angle of incidence θ_i (defined with respect to the sample's surface). Also, the magnetic foils are magnetized within the film plane rather than perpendicular to it, which is the so-called longitudinal configuration. In this geometry, the Faraday effect can be measured as long as the magnetization direction has a nonzero component parallel to the light's wave vector. Previously we derived expressions [10] for the Faraday rotation and ellipticity in this geometry

$$\varphi_{\rm F} = -\frac{E_{\rm phot}d_O}{\hbar c \tan \theta_{\rm t}} \Delta \delta, \qquad \tan \varepsilon_{\rm F} = \frac{E_{\rm phot}d_O}{\hbar c \tan \theta_{\rm t}} \Delta \beta, \tag{2}$$

where E_{phot} is the photon energy, d_0 is the thickness of the foil and θ_i is the angle of refraction.

At the 2p edges, due to the small value of the optical constants (ca. 10^{-3}), i.e. $n \approx 1$, it is usually assumed that $\tan \theta_i \approx \tan \theta_i$. Thus, according to equation (2), both $\Delta \delta$ and $\Delta \beta$ can directly

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Figure 1. Schematic picture of the experimental set-up used for the detection of the Faraday effect. An inclined angle of incidence θ_i is used and the magnetic field (*H*) set to ± 1000 e is applied longitudinally in the plane of the sample.

be obtained from experiment with rather small error. At the 3p edges, things are different. The optical constants are larger by a factor 10–100 and a more exact calculation of the tan θ_i factor shows that to first order in δ and β

$$\Delta \delta \approx \frac{\hbar c \tan \theta_{\rm i}}{E_{\rm phot} d_O} \left[-\varphi_{\rm F} \left(1 - \frac{\delta}{\sin^2 \theta_{\rm i}} \right) + \varepsilon_{\rm F} \left(\frac{\beta}{\sin^2 \theta_{\rm i}} \right) \right]$$

$$\Delta \beta \approx \frac{\hbar c \tan \theta_{\rm i}}{E_{\rm phot} d_O} \left[\varepsilon_{\rm F} \left(1 - \frac{\delta}{\sin^2 \theta_{\rm i}} \right) + \varphi_{\rm F} \left(\frac{\beta}{\sin^2 \theta_{\rm i}} \right) \right].$$
(3)

Therefore the *experimental determination* of $\Delta\delta$ and $\Delta\beta$ complicates due to the difficulty in obtaining an errorless spectrum for the dispersion. This is usually computed from a Kramers–Kronig transformation [28] of the absorption, leading to appreciable errors since theoretically its spectrum needs to be known from 0 eV to infinity.

Thus for the sake of simplicity we have measured the spectral dependence of the Faraday rotation and calculate $\Delta\delta$ according equation (2) assuming $\tan \theta_t \approx \tan \theta_i$. This introduces an error of approximately 15%, similar to what would be obtained by the above commented procedure. $\Delta\beta$ has been calculated using the Kramers–Kronig relation

$$\Delta\beta = \frac{2}{\pi} P \int_0^\infty \frac{\omega'}{\omega'^2 - \omega^2} \Delta\delta(\omega') \,\mathrm{d}\omega',\tag{4}$$

which in this case can be exactly computed since $\Delta \delta = 0$ away from the edges.

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3. Experimental technique

The room temperature experiments were performed at the U125/PGM beamline [29, 30] at the synchrotron radiation source BESSY using the BESSY polarimeter chamber [31]. The spectral resolution $E/\Delta E$ was set to 5000 and the degree of linear polarization of the incident light was assumed to be $P_{\text{Lin}} = 1$ because of beam optics [29, 30]. An amorphous Fe₂₀Ni₈₀ alloy film of 100 nm thickness, a 50 nm Co layer, and a 30 nm Ni layer were investigated. The films were sputter deposited on to 100 nm Si₃N₄ substrates and covered by a 3 nm cap layer (Cu for the alloy and Al for Co) to prevent oxidation. The analyser, a gold mirror on silicon, was operated at an angle of incidence close to the Brewster angle. *In situ* exchange and removal of samples allowed for quasi-simultaneous polarization analysis of the incident and the transmitted beam.

Figure 1 shows a schematic picture of the set-up used for the Faraday measurements. The angle of incidence was set to $\theta_i = 60^\circ$ for the Fe₂₀Ni₈₀ alloy and $\theta_i = 50^\circ$ for the pure Co and Ni samples. The films were magnetically saturated parallel and antiparallel to the direction of the scattering plane and of the surface of the sample by a magnetic coil system, which allowed the application of magnetic fields up to $H^{\pm} = \pm 500$ Oe.

In such a configuration, the total transmitted intensity (I) at the detector depends on the direction of the applied magnetic field $(I^+ \text{ and } I^- \text{ for } H^+ \text{ and } H^-, \text{ respectively})$, the photon energy and the azimuthal orientation of the analyser α (see figures 1 and 2) and is given by

$$I^{\pm} = I_0 T R_{\rm An} [1 + P_{\rm Lin} P \cos(2\alpha \pm 2\varphi_{\rm F})], \tag{5}$$

where the change in the ellipticity after transmission is contained in P_{Lin} , and φ_{F} accounts for the rotation of the polarization plane. I_{o} is the incident intensity, T the transmittance of the sample, R_{An} the reflectance of the analyser and P the polarizing power of the analyser defined by $P = (R_{\text{s}} - R_{\text{p}})/(R_{\text{s}} + R_{\text{p}})$, where $(R_{\text{s}} + R_{\text{p}})$ is the reflection coefficient of the analyser measured at $\alpha = 0^{\circ}(90^{\circ})$.

The polarization analysis was carried out in an energy dispersive way. The intensity behind the analyser mirror was monitored with the azimuthal angle fixed to $\alpha = 45^{\circ}$, while scanning the energy (figure 3). In this mode, the intensity reflected from the analyser is mainly sensitive to the rotation of the plane of polarization φ_F (see below equation (6)). This method has the great advantage over the classical polarization analysis (scanning α at different energies) that it permits one to obtain the Faraday rotation with very high spectral resolution as well as density of data points in a single measurement.

For the determination of the Faraday rotation, we measured the asymmetry parameter $A_{\alpha=45} = (I^+ - I^-)/(I^+ + I^-)$ of the intensity behind the analyser upon inverting the magnetization (by means of an external magnetic field $H^{\pm} = \pm 100 \text{ Oe}$) of the transmission sample at an analyser angle $\alpha = 45^{\circ}$. At this angle the difference between I^+ and I^- is maximal (see figure 2). Using equation (5), we obtain the Faraday rotation

$$\frac{A_{\alpha=45}}{P} = P_{\rm Lin} \sin 2\varphi_{\rm F},\tag{6}$$

where P_{Lin} is set to $P_{\text{Lin}} = 1$ since changes in the degree of linear polarization affect the transmission curve for $\alpha = 45^{\circ}$ only slightly [8].



Figure 2. Principal sketch of the polarization analysis. Classically, azimuthal α -scan of the analyser are performed at several photon energies across the edge. The Faraday rotation (φ_F) is obtained from the angular shift between curves measured for H > 0 and H < 0. The Faraday ellipticity (ε_F) is deduced from the decrement on the amplitude of this curves with respect to that of H = 0 Oe. Alternatively, in the energy dispersive mode the asymmetry $A_{\alpha} = (I^+ - I^-)/(I^+ + I^-)$ as a function of the photon energy is measured for fixed α allowing the spectral determination of ϕ_F from a single measurement. At $\alpha = 45^\circ$, $A_{\alpha=45}$ is maximum and the detector is most sensitive to the MO rotation since $(I^+ - I^-)$ is proportional to $P_{\text{Lin}} \sin(2\varphi_F)$.

4. Results and discussion

Figure 3 (top panel) shows the intensity data (I^+, I^-) at the Fe and Ni edges of the Fe₂₀Ni₈₀ film obtained for $\alpha = 45^\circ$ for two inverted magnetic fields. The deduced asymmetry is plotted in the bottom panel. From these data, the energy dependence of the Faraday rotation is determined using the experimentally obtained spectra of the polarizing power. In order to compare the MO rotation for all three elements on an equal footing we deduce the intrinsic MO rotation constants *k* using

$$\varphi_{\rm F} = k \, \frac{d_0}{\tan \theta_{\rm t}} \tag{7}$$

where the geometrical term accounts for the projection of the magnetic field and the thickness (d_0) of the layer. The Fe–Ni alloy film is treated as a double layer system of thicknesses 20 nm Fe and 80 nm Ni (we will examine the effects of alloy formation on the 3p MO response of the 3d transition metals in more detail in the near future) in order to determine the individual contributions to the MO constants of Fe and Ni. This separation of the individual structures at 60 eV is justified. The magnetic response of Ni practically vanishes for energies below 60 eV as can be seen from the bottom panel of figure 4.

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Figure 3. Top panel: the normalized intensities I^+/I_0 and I/I_0 measured at room temperature at the detector after the interaction of the radiation with the Fe₂₀Ni₈₀ sample. Bottom panel: the resulting asymmetry $A_{a=45} = (I^+ - I^-)/(I^+ + I^-)$, from which the magnetic contributions to the optical constants can directly be determined.



Figure 4. Absorption and MO constants obtained for Ni at the 3p edges. Top panel: the measured absorption constant β . Bottom panel: spectra of the MO constants $\Delta\delta$, $\Delta\beta$ and Faraday rotation constant *k*.



Figure 5. Absorption and MO constants obtained for Co at the 3p edges. Top panel: the measured absorption constant β . Bottom panel: spectra of the MO constants $\Delta\delta$, $\Delta\beta$ and Faraday rotation constant *k*.

The results are shown in the bottom panels of figures 4 and 5 for the pure Ni and Co samples, respectively whereas those of Fe and Ni obtained from the alloy are shown in the lower panel of figure 6. As can be seen by comparison of figures 4 and 6, the measured rotation spectrum at the Ni edge for the $Fe_{20}Ni_{80}$ film is identical to that measured for the pure Ni sample, further supporting the separation of magnetic structures for the alloy. The maximum values of the rotation constant *k* reach 2.1×10^5 , 1.5×10^5 , and 0.8×10^5 deg/mm at the 3*p* edges of Fe, Co, and Ni, respectively. These values are about one order of magnitude larger than those obtained in the visible range [32] and agree with those deduced from [25, 26] for the cases of Ni and Co at the 3*p* edges.

The energy dependence of the absorption constant β was obtained from the transmitted intensity and is shown in figures 4, 5 and 6 (top panels). For the Fe–Ni alloy, the individual contributions from Fe and Ni are not separated due to the proximity of the broad edges.

The individual magnetic contribution to the optical constant $\Delta\delta$ has been extracted directly from the Faraday rotation constant according to the following relation obtained from equations (2) and (7)

$$k = \frac{2\pi}{\lambda} \Delta \delta, \tag{8}$$

where for the evaluation the approximation $\theta_i \approx \theta_t$ was used. The result is shown in the bottom panels of figures 4–6 for Ni, Co, and for the Fe–Ni alloy, respectively. The $\Delta\beta$ spectra have been computed by the Kramers–Kronig transformation (equation (4)). Table 1 contains the maximum values obtained for Fe, Co, and Ni, respectively, at the 3*p* edges together with those reported at the 2*p* region [3, 9, 10].



Figure 6. Absorption and MO constants obtained for the Fe₂₀Ni₈₀ alloy sample at the 3*p* edges. Top panel: the measured absorption constant β . Bottom panel: spectra of the MO constants $\Delta\delta$, $\Delta\beta$ and Faraday rotation constant *k*. $\Delta\delta$, $\Delta\beta$ and *k* are normalized to the respective effective Fe and Ni thicknesses.

Table 1. Maximum values obtained for the MO constants $\Delta\delta$ and $\Delta\beta$ at the 3*p* and 2*p* edges of Fe, Co, and Ni. Data for the 2*p* edges and magnetic moments are taken from [8].

	Fe		Со		N	i
Edge	3 <i>p</i>	2 <i>p</i>	3 <i>p</i>	2 <i>p</i>	3 <i>p</i>	2p
$\Delta\delta(10^{-2})$	1.4	0.11	1.1	0.05	0.45	0.045
$-\Delta\beta(10^{-2})$	1.4	0.19	1.1	0.13	0.5	0.06
$\mu_{\rm B}/{\rm atom}$	2.23		1.64		0.62	

From table 1, we note that the absolute maximum values of $\Delta\delta$ and $\Delta\beta$ increase with the magnetic moment of the 3*d* element. Such behaviour is understood for the 2*p* edges, for which it was shown that the XMCD and Faraday spectra are directly proportional to the difference between the spin-up and spin-down 3*d* densities of states [13]. For the 3*p* core states such proportionality apparently also holds (see table 1). However, a significant difference between the 2*p* and 3*p* core levels is the relative extent of the EX and SO splitting. For the 2*p* levels the SO splitting is of the order of 15 eV, while the core level EX splitting can be ignored to a good approximation [10]. Thus, the large MO effects found at the 2*p* edges are due to the large SO interaction in the initial 2*p* states and the EX splitting of the final 3*d* states. The situation is different for the 3*p* core levels; here both the SO and EX splitting of the 3*p* levels are of

the same size [14] and consequently, contribute both to bringing about the sizable MO effect at the 3p edges.

Earlier magneto-x-ray experiments proved the existence of MO effects at the 3*p* edges, but did not highlight their attainable size. A T-MOKE reaching up to 17% was previously reported [18] for the 3*p* edges of Co and a value of 7% for the 3*p* threshold of Fe [19]. However, in a recent investigation we showed that at the 3*p* edges of Fe and Co huge T-MOKE signals reaching up to 75 and 94%, respectively can be achieved for $\theta_i \approx 45^\circ$ [20] where the maximum T-MOKE is expected. Thus, while the x-ray MO effects at the 3*p* edges have often been disregarded over those at the 2*p* edges, the present Faraday effect study as well as the recent T-MOKE study [21] prove that the MO effects at the 3*p* edges can in fact be *as large as* those at the 2*p* edges.

5. Conclusions

We have measured resonantly enhanced MO Faraday rotation spectra across the 3*p* levels of Fe, Co, and Ni. An advantage of Faraday measurements over classical XMCD experiments lies in the fact that linearly polarized light (with $P_{\text{Lin}} = 1$) can be employed. The polarization plane rotation after the interaction with the material has been determined using an energy dispersive mode in the photon energy range of interest (45–75 eV) from only one measurement. From these data, the complete set of MO constants and $\Delta\beta$ has been determined for Fe, Ni, and Co at the respective 3*p* edges.

Finally, large values for the Faraday rotation constant k have been found for Fe, Co, and Ni, similar to those found at their respective 2p edges [9]. We attribute this finding to the much more pronounced influence of the EX splitting of the 3p core levels on determining the size of the MO effects, thus showing clear differences with respect to MO effects at the 2p edges, where the effect of the EX splitting of the core levels is ignored because of the larger influence of the SO interaction.

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