

Observation of the X-Ray Magneto-Optical Voigt Effect

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The existence of the x-ray magneto-optical Voigt effect is demonstrated. By means of polarization analysis the Voigt rotation and ellipticity of linearly polarized synchrotron radiation are measured at the Co L_3 edge upon transmission through an amorphous Co film. The observed x-ray Voigt rotation is about $7.5^\circ/\mu\text{m}$. On the basis of *ab initio* calculations it is shown that the x-ray Voigt effect follows sensitively the amount of spin polarization of the $2p$ core states. Therefore it provides a unique measure of the spin splitting of the core states.

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The magnetic state of individual elements in a material can be selectively probed with x-ray magneto-optical (MO) spectroscopies, making them leading-edge research methods in the investigation of magnetism [1–4]. To the recent outstanding achievements in this area belong the demonstration of the anisotropy of the orbital moment with regard to the magnetization direction, and its relationship to the magnetocrystalline anisotropy energy [1,2], and the imaging of anomalous ferromagnetic domain structures [3]. Magneto-x-ray spectroscopies which have become popular in the last few years, due to the availability of brilliant and polarized synchrotron radiation, can be classified according to three criteria. The first criterion is the polarization state of the incident x-ray light being used, whether it is linearly or circularly polarized; the second criterion is whether an intensity measurement (e.g., the x-ray magnetic circular dichroism) or a polarization state analysis of the light after its interaction with the material is performed (e.g., the x-ray Faraday effect [5,6]). The third one is the dependence of the MO effect on the magnetization M , i.e., if the effect is to lowest order proportional to the expectation value of the magnetization, $\langle M \rangle$, or to the square of the magnetization, $\langle M^2 \rangle$ [7]. Up to now mainly intensity measurements of MO effects that are linear in $\langle M \rangle$ are performed (e.g., [1,3,8]). X-ray MO spectroscopies that are sensitive to $\langle M^2 \rangle$ hold great potential for the future investigation of antiferromagnetic (AFM) materials, something which is not possible with x-ray spectroscopies that are linear in M [9,10]. Technologically important materials of broad current interest, as magnetoresistive, spin-valve, and exchange-bias materials [11], could be investigated by such new types of x-ray spectroscopies on an element selective level [12].

A new x-ray MO phenomenon that is quadratic in M is the Voigt effect. The Voigt effect [13], which is sometimes denoted as linear magnetic birefringence, describes the rotation of the polarization plane of linearly polarized light when passing through a sample that has magnetic moments perpendicular to the propagation direction of the

light. In addition the state of polarization changes to elliptical. The Voigt effect is distinct from the x-ray magnetic linear dichroism (XMLD), an effect which is also sensitive to $\langle M^2 \rangle$: Whereas in the Voigt experiment a polarization analysis is performed, the XMLD experiment is a plain intensity measurement [14–16], revealing thus no information on the polarization state of light.

In this Letter, we demonstrate for the first time the existence of the x-ray MO Voigt rotation and ellipticity, with spectra measured at the L_3 edge of an amorphous Co film at room temperature. We show that the measured MO effect is indeed the x-ray Voigt effect, by presenting three pieces of evidence: (1) we show that the measured MO quantities obey all symmetry relations expected for the Voigt effect, as the dependence on the magnetization and polarization directions; (2) we have performed additional measurements of the XMLD asymmetry, which can be related directly to the Voigt rotation; and (3) we have made *ab initio* calculations of the x-ray Voigt effect spectra, which are in good agreement with the measured spectra. As little is yet known about the physical origin of MO effects that scale with $\langle M^2 \rangle$, we show on the basis of the *ab initio* calculations that the x-ray Voigt effect depends crucially on the spin splitting of the $2p$ core states. The spin splitting of the $2p$ core states is small (<1 eV), and therefore omissible for the modeling of x-ray MO effects that are linear in $\langle M \rangle$. As an unexpected outcome, our calculations show the Voigt effect to *vanish* when the $2p$ spin splitting is neglected.

The Voigt effect is detected in the geometry shown in Fig. 1, in which the incident light is normal to the surface, and is linearly polarized at an angle α with respect to the magnetization. For $\alpha = \pi/4$ one has the standard Voigt geometry as known from the visible range [13]. The magnetization lies in the plane of the film. Upon transmission, the polarization plane becomes rotated over the Voigt angle θ_V , and, in addition, the light has become elliptically polarized by an amount ε_V . The two refractive indices of the material are n_{\parallel} and n_{\perp} , corresponding to the propagation of linear modes which have the electrical field vector

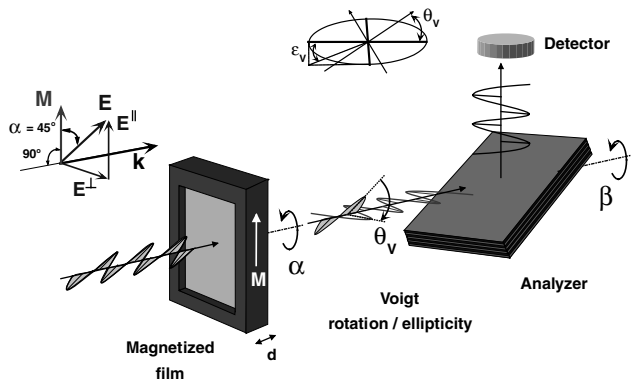


FIG. 1. Experimental setup for observing the x-ray Voigt effect on magnetic films. The film is magnetized in plane and the linearly polarized x-ray beam is at normal incidence to the film. The polarization plane and magnetization are at an angle α . In the standard Voigt configuration, α is set at 45° . The polarization plane of the transmitted light is tilted over an angle, which is the Voigt rotation θ_V , and also the transmitted light has become elliptically polarized.

parallel, perpendicular, respectively, to the magnetization. For $\alpha = \pi/4$ the incident light can be decomposed in two equal components, one parallel and one normal to the magnetization. When the two refractive indices are unequal, which happens in an isotropic material *only* because of the magnetization, the complex Voigt effect occurs upon transmission. It can be expressed by

$$\begin{aligned} \theta_V - i\varepsilon_V &\approx \frac{\omega d}{2ic} [n_{\parallel} - n_{\perp}] \\ &\approx \frac{\omega d}{4ic\bar{n}} \left[\epsilon_{\parallel} - \epsilon_{\perp} - \frac{\epsilon_{od}^2}{\epsilon_{\perp}} \right], \end{aligned} \quad (1)$$

where d is the thickness of the film, ϵ_{\parallel} , ϵ_{\perp} , are the diagonal components of the permittivity tensor for orientations of the E vector parallel, normal, respectively, to the magnetization, \bar{n} is the average refractive index, and ϵ_{od} is the off-diagonal permittivity of the ferromagnetic material [17,18]. It follows from Eq. (1) that the Voigt effect is even in the magnetization: The Onsager relations demand that the diagonal components of the permittivity be even in M , $\epsilon_{\parallel,\perp}(-M) = \epsilon_{\parallel,\perp}(M)$, whereas the off-diagonal permittivity is to be odd, $\epsilon_{od}(-M) = -\epsilon_{od}(M)$. Substituted into (1), these relations show that the Voigt effect is to lowest order quadratic in M . For AFM materials ϵ_{od} vanishes, but $\epsilon_{\parallel} - \epsilon_{\perp}$ remains. Therefore the Voigt effect has the advantage over the Faraday and Kerr effects that it is suitable for the study of antiferromagnets as well [19]. Both for ferro- and antiferromagnets, Voigt effect measurements provide information on the magnetocrystalline anisotropy Δn in the refractive index, i.e., $\Delta n = n_{\parallel} - n_{\perp}$.

The Voigt effect can be measured at any angle α between the polarization and magnetization. The expected α dependence of the Voigt rotation is $\theta(\alpha) = \theta_V \sin 2\alpha$, and similar for the ellipticity [17]. Thus, when $\alpha = 0$ or $\pi/2$ the Voigt effect is anticipated to disappear, whereas

at an angle $\alpha = \pi/4, 3\pi/4$, the maximal values are attained, which are the rotation θ_V and ellipticity ε_V . The α and M characteristics of the Voigt effect are indispensable to identify the x-ray Voigt effect.

A quantity related to the Voigt effect is the XMLD asymmetry parameter A , which is measured as an intensity difference of light transmitted with polarization perpendicular (T_{\perp}) and parallel (T_{\parallel}) to the magnetization. We shall show experimentally that

$$A \equiv (T_{\parallel} - T_{\perp}) / (T_{\parallel} + T_{\perp}) \approx 2\theta_V, \quad (2)$$

as is theoretically expected. Although the XMLD parameter A and Voigt rotation θ_V are connected, they are measured in entirely different methods. Previously it was shown in the visible range that the Voigt effect is the most suitable method to visualize AFM coupled domains [20].

The experiments, schematically shown in Fig. 1, have been performed at the undulator beam line UE56/1 PGM 1 of BESSY II [21]. The spectral resolution at about 780 eV was $E/\Delta E = 2500$, and the degree of linear polarization was practical unity ($P_L > 0.99$). We used the BESSY ultrahigh vacuum polarimeter chamber [22], in which the angle α can be varied by rotating the sample. Magnetic coils supply variable magnetic fields between ± 500 Oe that are oriented in the sample's plane, and that are sufficient to obtain magnetically saturated films. The linear polarization of the incident or transmitted beam was analyzed by rotating a W/B₄C reflection multilayer (300 periods of 1.2 nm each, angle of incidence close to the Brewster angle) around the azimuthal angle β (see Fig. 1), while the reflected intensity is monitored by a GaAs:P diode. *In situ* exchange and removal of samples enable a quasisimultaneous polarization analysis of the incident and of the transmitted beam. The transmission sample was an amorphous, magnetron-sputter deposited Co layer (200 nm) on a Si₃N₄ foil of 100 nm thickness, capped with 3 nm V to prevent oxidation. Note that the sample must be amorphous or cubic for the Voigt experiment to guarantee that $\epsilon_{\parallel} \equiv \epsilon_{\perp}$ in the absence of magnetization.

To measure the Voigt effect we have monitored the intensity at the detector (see Fig. 1) as a function of α at a fixed analyzer angle β for each photon energy across the Co $2p_{3/2}$ edge (see Fig. 2, inset). At $\beta = \pi/4$ the intensity reflected from the analyzer is most sensitive to small rotations θ of the plane of polarization of the transmitted light, while for $\beta = 0$ it is most sensitive to changes of ε . The detected normalized intensity is the product of the incident intensity, the transmission T of the sample, and the reflection R of the analyzer. We obtain the ellipsometric quantities by fitting the measured intensity $TR(\alpha)$ (see Fig. 2) using the following functional forms for T and R . The transmission T can be expressed as $T = \bar{T}[1 + A \cos(2\alpha)]$ with A the XMLD parameter and \bar{T} the average transmission, $\bar{T} = (T_{\parallel} + T_{\perp})/2$. The reflectivity is given by $R = R_0 + (R_s - R_p)/(R_s + R_p)P'_L \cos(2\beta + 2\theta)$, where R_0 is the average reflectance, R_s , R_p are the

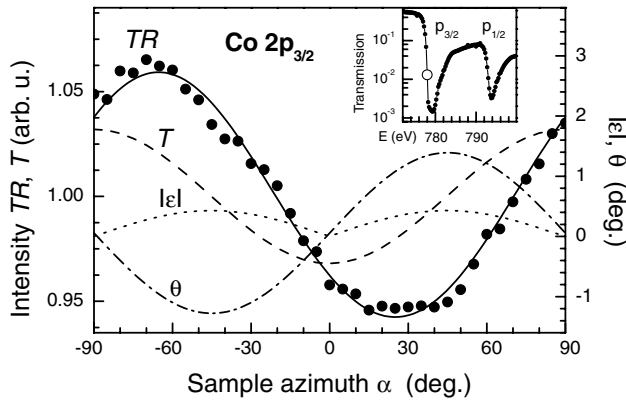


FIG. 2. Normalized intensity TR at 778 eV as a function of α (\bullet) and the fit (solid curve, left ordinate). The deduced MO rotation θ , the ellipticity $|\epsilon|$, and the transmission T are shown by the dash-dotted, dotted, and dashed curves, respectively. The inset shows the transmission spectrum T_{\perp} across the Co $2p$ absorption edge.

known s , p reflectances of the analyzer that are independent of α , and P_L' is the degree of linear polarization after the sample. P_L' is given by $P_L' \approx 1 - 2\epsilon^2$, thus to first order in ϵ we have $P_L' \approx 1$. We first evaluate the spectra taken at $\beta = 45^\circ$ for fixed $P_L' = 1$, which gives us the α dependence of θ . This yields directly $\theta(\alpha) \propto \sin 2\alpha$ (see Fig. 2), which shows that the measured MO rotation obeys the theoretically expected dependence of the Voigt rotation on the angle α . This excludes that the measured MO angle is a Faraday rotation induced by misalignment, because, if there were a magnetization component along the propagation direction, a constant α dependence would result. In a second step we evaluated the data taken at $\beta = 0^\circ$ in which we determine $|\epsilon|$, since only ϵ^2 enters the fit formula. We note that $|\epsilon|$ obviously cannot be fitted with the same accuracy as θ . We obtain for ϵ the best fit result with a $\sin(2\alpha)$ dependence (Fig. 2), which is exactly what is expected for the Voigt ellipticity.

The measured soft x-ray Voigt rotation spectrum is shown in Fig. 3. Each data point for θ_V is the result of an α scan for a fixed photon energy. Because of the resonant excitation from $2p$ to $3d$ levels, the rotation data display resonant maxima immediately below and above the absorption edge. The maximum values of up to $7.5^\circ/\mu\text{m}$ are almost an order of magnitude larger than those observed in the visible region [23]. As a further test we measured under inverted magnetic field the Voigt rotation, which we found to be independent of the magnetization direction (see Fig. 3). These experimental observations unambiguously show that the measured x-ray MO effect is the Voigt effect.

For comparison to the x-ray Voigt rotation, we determined the XMLD asymmetry by measuring the transmission spectra T_{\perp} (see Fig. 2, inset) and T_{\parallel} . The result is shown in Fig. 3. The two quantities agree well, in accordance with Eq. (2), except for the high-energy side of the $2p_{3/2}$ absorption edge. The XMLD experiment is dis-

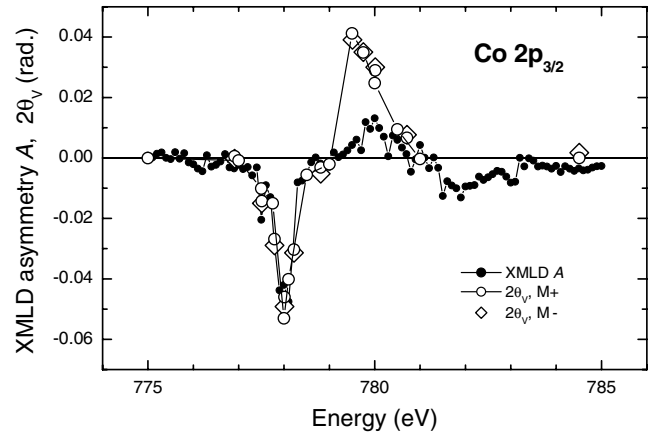


FIG. 3. The x-ray Voigt rotation θ_V measured at the Co L_3 edge with two antiparallel directions of the magnetization (\circ , \diamond). Their equivalence shows the quadratic dependence of the Voigt rotation on the magnetization. For comparison the XMLD asymmetry A (\bullet) is shown. Both spectra ought to agree according to theory, but do not around 780 eV because the XMLD is diminished by fluorescence noise.

turbed by fluorescence background noise, which occurs at the high-energy side of the Co $2p_{3/2}$ edge. Thereby the XMLD asymmetry becomes diminished. The measurement of the Voigt rotation is less liable to fluorescence disturbances, because the analyzer is energy selective and set to select the incident light, while the fluorescence light with its different wavelength is suppressed.

Further information follows from our *ab initio* calculations, which were performed before the experiment. To our knowledge, no calculations of the Voigt effect existed prior to ours. We computed the components of the permittivity tensor, for which we used the Kubo linear-response expressions in the single-particle approximation [24,25]. The required single-particle states and energies are computed within the local spin-density approximation to density-functional theory [26]. In our numerical evaluation of the permittivities we employ the full-potential, linearized augmented plane-wave method as implemented in the WIEN97 code [27]. The core states are computed as solutions of the Kohn-Sham-Dirac equation in a spherical potential, with spin polarization treated perturbatively. The relativistic valence states are computed within the full, nonspherical potential, using the second variational approach to include self-consistently the spin-orbit interaction [25]. *Ab initio* calculations can provide insight into physical aspects that are not accessible experimentally: First our calculations of the ϵ components showed that the third term in (1), $\epsilon_{od}^2/\epsilon_{\perp}$, is about 100 times smaller than $\epsilon_{\parallel} - \epsilon_{\perp}$, and therefore plays no role at all. Second, our calculations revealed an unprecedented sensitivity of the x-ray Voigt effect to the spin polarization of the core states. This is shown in Fig. 4 where we plotted the experimental Voigt spectra together with the *ab initio* calculated spectra of fcc Co that were computed both with and without the core spin polarization. We chose fcc Co for

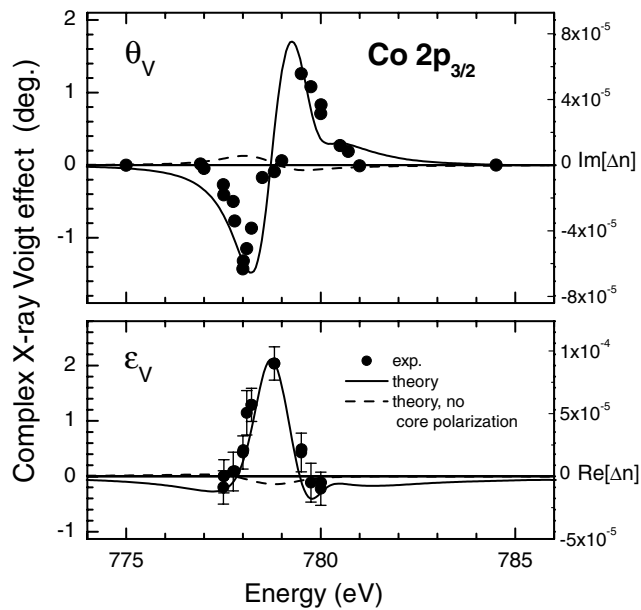


FIG. 4. Calculated and measured x-ray Voigt effect at the L_3 edge of 200 nm Co. Top panel: The experimental Voigt rotation θ_V (●) of amorphous Co and the calculated Voigt rotation of fcc Co. The full curve gives the Voigt rotation computed with spin polarization of the $2p_{3/2}$ core states, while the dashed curve shows the Voigt rotation obtained without core polarization. Bottom panel: Likewise, but for the Voigt ellipticity ε_V . The right ordinates show the magnetocrystalline anisotropy Δn in the refractive index n .

the calculation, because it is more isotropic and thus better comparable to amorphous Co than hcp Co. The negative ε values in Fig. 4 were extrapolated in accordance with the Kramers-Kronig relations. A lifetime broadening of 0.8 eV was applied in the calculations. Obviously, the Voigt rotation predicted with the $2p_{3/2}$ spin polarization included and the measured spectrum are in excellent agreement, astonishing in view of the smallness of the MO quantity. Conversely, the Voigt effect calculated *without* core spin polarization, but with exchange-split valence states, is practically zero and bears no correspondence to the experiment. This demonstrates that the x-ray Voigt effect depends critically on the core spin splitting, which is unexpected, since the spin splitting of the $3/2$, $-3/2$ sub-levels of Co is only 0.79 eV. In Fig. 4 we also present the magnetocrystalline anisotropy Δn in the refractive index as determined via Eq. (1) from the data (right ordinates).

To conclude, we have for the first time proven the existence of the x-ray magneto-optical Voigt effect. In the presence of fluorescence the x-ray Voigt effect can be measured more precisely than the related XMLD. Our investigations open up new possible avenues for magneto-x-ray studies of antiferromagnetic materials. Utilizing the x-ray Voigt effect, element selective MO studies of the local magnetization in AFM materials become feasible. We also expect

the x-ray Voigt effect to hold potential for the visualization of AFM domains. For cubic ferromagnetic and AFM materials the magnetocrystalline anisotropy in the refractive index n can be probed using x-ray Voigt spectroscopy. In combination with *ab initio* calculations, x-ray Voigt effect measurements provide a powerful new instrument to investigate the core spin splitting.

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