Prediction of huge X-ray Faraday rotation at the Gd N_{4.5} threshold

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X-ray absorption spectra in a wide energy range around the 4d–4f excitation threshold of Gd were recorded by total electron yield from in-plane magnetized Gd metal films. Matching the experimental spectra to tabulated absorption data reveals unprecedented short light absorption lengths down to 3 nm. The associated real parts of the refractive index for circularly polarized light propagating parallel or antiparallel to the Gd magnetization, determined through the Kramers-Kronig transformation, correspond to a magneto-optical Faraday rotation of 0.7° per atomic layer. This finding shall allow the study of magnetic structure and magnetization dynamics of lanthanide elements in nanosize systems and dilute alloys.

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Resonance enhancements of X-ray magnetic scattering cross sections at inner-shell absorption edges have been used for years to investigate the magnetic structure of lanthanide and actinide systems in the hard X-ray regime (above 2 keV). The experimental demonstration of large changes in the specularly reflected X-ray intensity at the Fe L_{2,3} edge upon magnetization reversal initiated the ongoing search for magneto-optical (MO) effects in the soft X-ray regime as well as their application to element-specific studies of heteromagnetic systems.

Yet, in analyzing soft X-ray MO signals from thin films and multilayer systems with thicknesses comparable to the X-ray wavelength, previous investigations have shown[1-3] that a comparison with model calculations of the reflected specular intensity (based on the Fresnel equations) is needed in order to extract a layer-resolved sample magnetization profile. Several experimental determinations of soft X-ray MO constants have been reported[4,5] for ferromagnetic transition metals in the region of the L_{2,3} thresholds, but none so far for the lanthanide elements, despite their wide recognition as, e.g., constituents of exchange-spring magnets[6] and magnetic recording media[7]. Only recently has it been demonstrated that sizeable MO signals are obtained from lanthanide elements in the soft X-ray region at the N_{4.5} thresholds[8].

Here we show that calibrated N_{4.5} absorption spectra from magnetized Gd metal, recorded with circularly polarized (CP) light in the energy interval from 110 to 200 eV, yield an X-ray absorption coefficient up to three times larger than expected. The magnetization-dependent absorption of CP light at the Gd N_{4.5} giant resonance maximum, described by the imaginary part of the refraction index, is accompanied by a huge change in light propagation speed upon magnetization reversal, a dispersive effect described by the real part of the refraction index, implying a Faraday rotation (FR) of about 0.7° per atomic layer. Thus, even very small or diluted lanthanide systems are expected to show a measurable effect.

The absorption experiments were performed at the high-resolution UE56 undulator beamline of the Berliner Elektronenspeicherring für Synchrotronstrahlung (BESSY II). The photon energy resolution was set to about 100 meV (FWHM) which is well below the intrinsic width of the narrow Gd N_{4.5} pre-edge absorption lines[4]. The photon energy interval from 110 to 200 eV was scanned at slow speed by a synchronized movement of monochromator and undulator. This synchronization is essential to properly normalize the absorption spectra and allows one to exploit the high flux of the undulator beamline of about 10^{14} photons/(s·100 mA·0.1% bandwidth) over a wide energy range. The degree of circular polarization at this Sasaki-type undulator beamline is practically 100%[23].

The absorption spectra were recorded in total-electron yield (TEY) mode using a high-current channeltron. To suppress the background of secondary electrons from the chamber walls, both the sample and a retarding grid placed in front of the channeltron, were biased with a low-voltage battery. For signal stability, high voltage was supplied to the channeltron cathode by a 3.2 kV battery box. The electron-yield current was amplified by an electrometer (set to 3 ms integration time for a scan speed of typically 0.1 eV per second). We used a light incidence angle of 30° with respect to the film plane, in order to compromise between a large projection of the CP light wave-vector onto the in-plane film magnetization and the desired small sample reflectivity[23].

Epitaxial Gd metal films of (10 ± 1) nm thickness were prepared in situ by vapor deposition in ultra-high vacuum (3 \times 10^{-11} mbar base pressure; about 4 \times 10^{-10} mbar during deposition) on a W(110) single-crystal substrate (for details of film preparation, see Ref. 28). For remnant in-plane sample magnetization, an external field was applied along [110] of the substrate, i.e., parallel to the easy magnetization axis of the Gd film, using a rotatable electromagnet[24]. A compact visible-light MO Kerr-effect setup[25] was used to routinely check the state of remnant magnetization of the Gd films, revealing square-shaped hysteresis loops with about 100 Oe coercivity.

Figure 1 displays experimental absorption spectra in...
the region of the Gd N₄₋₅ threshold. The spectra were corrected for saturation, assuming 0.3 for the ratio of electron escape depth to minimal X-ray absorption length. The photon energy range of the present spectra is significantly wider than of those measured in previous studies, including the wide asymmetric flanks of the 4d → 4f giant resonance (Beutler-Fano profile). This allows one to calibrate the absorption spectra by matching both ends to the tabulated absorption coefficient at photon energies where the influence of the giant resonance is expected to be negligible. To this end we fixed the absorption coefficients µ± at the low-energy (110 eV) and high-energy sides (200 eV) of the measured spectra to the values given by the tables of Henke et al. 15.0 and 17.1 ×10⁻³ nm⁻¹ at 110 and 200 eV, respectively. This then defines the given scale of the ordinate in Fig. 1. In this way, the absorption coefficient is obtained with an error bar of ±15% at the maximum, estimated from our experimental precision of ±1% at both ends of the photon energy range, where the matching to the tabulated data was performed.

We note that the spectra in Fig. 1 show the same qualitative energy dependence as given earlier, yet the previous spectra were scaled to the same maximum value for both magnetization directions without any correction for saturation effects.

The comprehensive X-ray data tables of Henke et al. 32 contain the lanthanide N₄₋₅ absorption spectra by Zimkina et al. 33, who measured relative linear X-ray absorption lengths of nonmagnetized lanthanide samples. To obtain an absolute absorption length, Henke et al. followed Richter et al. 34 who calibrated their gas-phase photoexcitation data by matching them to calculated cross sections. In this way they arrived at a maxi-

FIG. 1: Gd N₄₋₅ absorption spectra from remanently magnetized Gd films at T = 30 K. CP light was incident at 30° with respect to the film plane, i.e. mainly parallel (filled symbols) and antiparallel (open symbols) to the in-plane sample magnetization.

FIG. 2: (a) Difference spectrum, \( \Delta \mu(\omega) \equiv (\mu_+ (\omega) - \mu_- (\omega))/\cos(30°) \), of the absorption spectra for opposite magnetizations given in Fig. 1. (b) Difference \( \Delta \beta \) of the imaginary parts (filled symbols) and the associated difference \( \Delta \delta \) of the real parts (open symbols) obtained through a Kramers-Kronig transformation. Inset: pre-edge range, measured (\( \Delta \beta \), filled symbols) and calculated (\( \Delta \delta \), open symbols) with higher point density. (c) FR and ellipticity spectra calculated for linearly polarized light transmitted normally through a 0.3 nm thick Gd metal film magnetized perpendicular to the film plane.
um absorption coefficient at the nonmagnetic Gd N₄₅₅ peak of $\mu \approx 0.1$ nm⁻¹, corresponding to a linear X-ray absorption length $\lambda = 1/\mu \approx 10$ nm. By contrast, the calibrated experimental spectra from magnetized Gd in Fig. [1] reveal maximum values for the absorption coefficient of 0.33 nm⁻¹ and 0.36 nm⁻¹, for nearly parallel and antiparallel orientation of sample magnetization and photon spin, respectively. The corresponding linear absorption lengths are 3.0 nm and 2.8 nm, with the quoted error of 7%; they are about three times shorter than expected. These soft X-ray absorption lengths are remarkably short, even compared with visible-light absorption lengths in metals of typically some 20 nm.

The difference spectrum $\Delta \mu(\omega) \equiv (\mu_+(-\omega) - \mu_+(-\omega))/\cos(30^\circ)$, obtained from the experimental absorption spectra $\mu_\pm(\omega)$, is displayed in Fig. 2a. The factor $1/\cos(30^\circ)$ accounts for the finite experimental angle between the directions of light propagation and magnetization. Apart from minor contributions from the weaker pre-edge transitions (inset of Fig. 2b) around 140 eV, the $\Delta \mu(\omega)$ spectrum exhibits an S-shape behavior, with a zero crossing near 149 eV. It originates from the very intense 4d¹⁰ 4f⁹[S]→4d⁹ 4f⁸[P] transitions (dipole-allowed in LS coupling). For parallel orientation of photon spin and sample magnetization ($\Delta M = +1$ transitions), the intermediate $8\, P_{5/2}$ state at around 148 eV is preferentially populated; for antiparallel orientation ($\Delta M = -1$ transitions), by contrast, the only allowed excitation is into the higher $8\, P_{9/2}$ state at around 150 eV. The large difference in absorption coefficient for opposite magnetization directions (Fig. 2a) corresponds to a difference in the absorptive part $\Delta \beta \equiv \beta_+ - \beta_- = \Delta \mu \lambda/(4\pi)$ of the refractive index $n_\pm = 1 - \delta_+ - i\beta_\pm$. As shown in Fig. 2b, $\Delta \beta$ changes from +0.082 to −0.085 within 3.5 eV.

From the present data, the associated difference in the real part $\Delta \delta \equiv \delta_+ - \delta_-$, for CP light propagating (exactly) parallel or antiparallel to the Gd magnetization, was derived using the Kramers-Kronig (KK) transformation for magnetic systems. The result is given in Fig. 3b by open symbols. The accuracy of this integral transformation depends mainly on the spectral range available for integration. Within the extended photon energy range of 110 - 200 eV, the absorption spectra recorded for opposite magnetization directions appear to become asymptotically equal at both ends of the experimental photon-energy range (see Fig. 1). Hence the difference $\Delta \delta$ vanishes at the two boundaries, so that the result of the KK transformation is not affected by the choice of the experimental photon energy range. $\Delta \delta$ peaks right at the zero crossing of the absorptive part, where it amounts to $\Delta \delta \approx 0.11$ (see Fig. 2b).

For future applications, we use the experimental difference in the absorptive part, $\Delta \beta$, together with the calculated phase difference, $\Delta \delta$, to calculate the complex FR of Gd metal. Here we assume fully oriented 4f magnetic moments as existing, e.g., in the ferromagnetic phase at low temperatures ($T/T_C \ll 1$). Real and imaginary parts of the FR, $\Theta_F$ and $\Psi_F$, respectively, are given by the expressions:

$$\tan(2\Theta_F) = 2\, \text{Re}[a]/(1 - |a|^2), \quad (1)$$

$$\sin(2\Psi_F) = 2\, \text{Im}[a]/(1 + |a|^2), \quad (2)$$

where $d$ is the film thickness, $a = \tan(\Delta n\omega d/c)$ and $\Delta n = (n_+ - n_-)/2$. The $\Theta_F(\omega)$ and $\Psi_F(\omega)$ spectra of Gd metal at the N₄₅₅ threshold are presented in Fig. 2 for linearly polarized (LP) light transmitted in normal direction through a 0.3 nm (1 monolayer) thick Gd metal film magnetized perpendicular to the film plane, either parallel or antiparallel to the light propagation direction. The spectra predict a FR of $\Theta_F = (0.73 \pm 0.11)\degree$ per 0.3 nm ((2.4 ± 0.4)°/nm) near 149 eV, right at the zero crossing of the absorption difference for CP light in Fig. 2a. At this photon energy, $\Theta_F$ is accompanied by a vanishing Faraday ellipticity $\Psi_F$ (cf. Fig. 2b). To our knowledge this is by far the largest specific FR reported. It is 9 times larger than the specific rotation maximum at the Fe L₃ threshold and some 70 times (50 times) larger than in the visible (infrared) region of Fe metal.

With the predicted specific FR at N₄₅₅ thresholds, already some 10¹⁵ lanthanide atoms per cm² as in, e.g., a single atomic layer, a very dilute film, or nanosize particles, should be sufficient to show a measurable rotation. Note that it is not at all evident that continuum classical electrodynamics, as used in this work for the 10 nm thick Gd metal films, will still be appropriate when approaching atomic dimensions.

The huge FR at Gd N₄₅₅ is due to the very large electric dipole (E1) transition probability of 4d¹⁰ 4f⁹ → 4d⁹ 4f⁸⁺¹ transitions ($n = 7$ for Gd). Hence, when applying magnetized Gd films as a method to rotate the plane of light polarization at the fixed photon energy of 149 eV, the strong absorption at the Gd N₄₅₅ maximum (cf. Fig. 1) leads to a very short penetration length of the order of only 3 nm. In order to obtain, e.g., a FR of ±45° for opposite film magnetizations, one would use a 18.5 nm thick Gd film, with an inevitable intensity loss in intensity, which leads to a transmitted flux of about 10¹¹ photons/(s·100 mA·0.1% bandwidth) at a typical third-generation undulator beamline, Gd films might well be useful in differential (lock-in technique) experiments, where fast switching of the X-ray polarization plane (ns time scale) is required. One could extend the photon energy range of this method to about 180 eV by using heavier lanthanide elements.

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