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[1,2] have been used for years to investigate the magnetic structure of lanthanide[3] and actinide[4] 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[5] upon magnetization reversal initiated the ongoing search for magneto-optical (MO) effects in the *soft* x-ray regime,[6–12] as well as their application to element-specific studies of heteromagnetic systems.[13–18]

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[5,13,18] that a comparison with model calculations[19] 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[8–10] 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[20] and magnetic recording media.[21] Only recently has it been demonstrated that sizable MO signals are obtained from lanthanide elements in the soft x-ray region at the $N_{4,5}$ thresholds.[22]

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[23] of the Berliner Elektronenspeicherring für Synchrotronstrahlung (BESSY II). The photon energy resolution was set to about 100 meV (full width at half maximum) which is well below the intrinsic width of the narrow Gd $N_{4,5}$ pre-edge absorption lines.[24] 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 the total-electron yield 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.[25]

Epitaxial Gd metal films of 10 ± 1 nm thickness were prepared *in situ* by vapor deposition in ultrahigh vacuum (3×10^{-11} mbar base pressure; about 4×10^{-10} mbar during deposition) on a W(110) single-crystal substrate (for details of film preparation, see Ref. 26). For remanent *in-plane* sample magnetization, an external field was applied along the [110] direction of the substrate, i.e., parallel to the easy magnetization axis of the Gd film, using a rotatable electromagnet.[27] A compact visible-light MO Kerr-effect setup[28] was used to routinely check the state of remanent magnetization of the Gd films, revealing square-shaped hysteresis loops with about a 100-Oe coercivity.

Figure 1 displays experimental absorption spectra in the region of the Gd $N_{4,5}$ threshold. The spectra were corrected for saturation,[29,30] assuming 0.3 for the ratio of the electron escape depth to the minimal x-ray absorption length. The photon energy range of the present spectra is significantly wider than of those measured in previous studies,[24,31] including the wide asymmetric flanks of the $4d\rightarrow4f$ giant resonance (the Beutler-Fano profile). This allows one to calibrate the absorption spectra by matching both ends to the tabulated absorption coefficient[32] at photon energies where the influ-

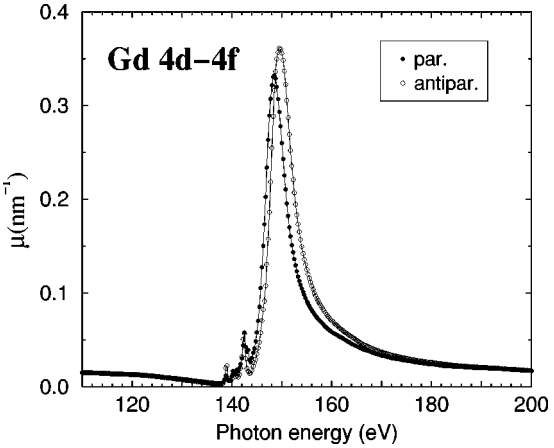

FIG. 1. Gd $N_{4,5}$ absorption spectra from remanently magnetized Gd films at $T=30$ K. CP light was incident at 30° with respect to the film plane, i.e., mainly parallel (filled symbols) and antiparallel (open symbols) to the in-plane sample magnetization.

ence of the giant resonance is expected to be negligible. To this end we fixed the absorption coefficients $\mu_{\pm}$ 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.*[32] 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 absoption coefficient is obtained with an error bar of $\pm15\%$ at the maximum, estimated from our experimental precision of $\pm1\%$ 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,[24] 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] contained the lanthanide $N_{4,5}$ absorption spectra of 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.*[32] 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 maximum absorption coefficient at the nonmagnetic Gd $N_{4,5}$ peak of $\mu\approx0.1$ nm^{-1}, corresponding to a linear x-ray absorption length of $\lambda=1/\mu\approx10$ nm.[32] By contrast, the calibrated experimental spectra from magnetized Gd in Fig. 1 reveal maximum values for the absorption coefficient of 0.33 and 0.36 nm^{-1} for nearly parallel and antiparallel orientations of the sample magnetization and photon spin, respectively. The corresponding linear absorption lengths are 3.0 and 2.8 nm, with a quoted error of $\pm15\%$; they are about three times shorter than expected.[32] 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 $\Delta\mu(\omega)\equiv[\mu_+(\omega)-\mu_-(\omega)]/\cos(30°)$, obtained from the experimental absorption spectra $\mu_{\pm}(\omega)$, is displayed in Fig. 2(a). The factor $1/\cos(30°)$ accounts for the finite experimental angle between the direc-

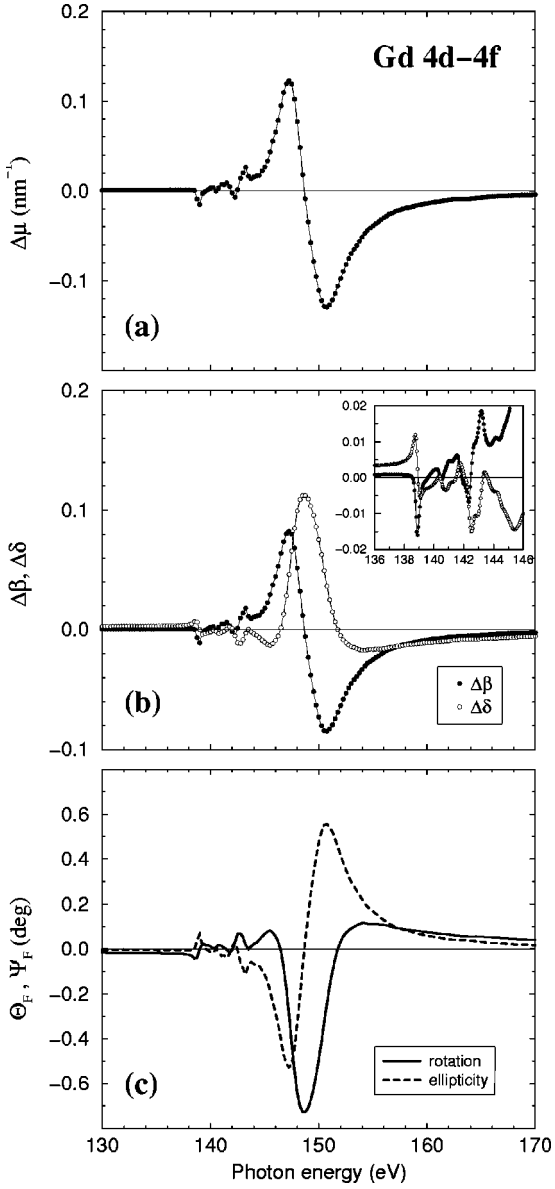

FIG. 2. (a) Difference spectrum $\Delta\mu(\omega)\equiv[\mu_+(\omega)-\mu_-(\omega)]/\cos(30°)$, the absorption spectra for opposite magnetizations given in Fig. 1. (b) Difference $\Delta\beta$ of the imaginary parts (filled symbols) and the associated difference $\Delta\delta$ of the real parts (open 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.

tions of light propagation and magnetization. Apart from minor contributions from the weaker pre-edge transitions [inset of Fig. 2(b)] around 40 eV, the $\Delta\mu(\omega)$ spectrum exhibits an S-shaped behavior, with a zero crossing near 149 eV. It originates from the very intense $4d^{10}\,4f^7[\,^8S]\rightarrow4d^9\,4f^8[\,^8P]$ transitions (dipole-allowed in *LS* coupling). For a *parallel* orientation of the photon spin and sample magnetization ($\Delta M=+1$ transitions), the intermediate $^8P_{5/2}$ 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 $^8P_{9/2}$ state at around 150 eV.[24] The large difference in absorption coefficient for opposite magnetization directions [Fig. 2(a)] corresponds to a difference in the absorptive part $\Delta\beta\equiv\beta_+-\beta_-=\Delta\mu\ \lambda/(4\pi)$ of the refractive index $n_\pm=1-\delta_\pm-i\beta_\pm$. As shown in Fig. 2(b), $\Delta\beta$ 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;[35] the result is given in Fig. 2(b) 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 $\Delta\beta$ 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 $\Delta\delta\approx0.11$ [see Fig. 2(b)].

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 ($T/T_C\ll1$). Real and imaginary parts of the FR, Θ_F and Ψ_F, respectively, are given by the expressions[36]

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

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

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 $N_{4,5}$ threshold are presented in Fig. 2(c) for linearly polarized 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 $\Theta_F=(0.73\pm0.11)^\circ$ per 0.3 nm [$(2.4\pm0.4)^\circ$/nm)] near 149 eV, right at the zero crossing of the absorption difference for CP light in Fig. 2(a). At this photon energy, Θ_F is accompanied by a vanishing Faraday ellipticity Ψ_F [cf. Fig. 2(c)]. To our knowledge this is by far the largest specific FR reported. It is nine times larger than the specific rotation maximum at the Fe L_3 threshold[10] and some 70 times (50 times) larger than in the visible (infrared) region of Fe metal.

With the predicted specific FR at $N_{4,5}$ thresholds, already some 10^{15} lanthanide atoms per cm^2 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 $N_{4,5}$ is due to the very large electric dipole ($E1$) transition probability of $4d^{10}\,4f^n\rightarrow4d^9\,4f^{n+1}$ 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 $N_{4,5}$ 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 $\pm45^\circ$ for opposite film magnetizations, one would use a 18.5-nm-thick Gd film, with an inevitable intensity reduction by a factor of 4.5×10^2. Despite this substantial loss in intensity, which leads to a transmitted flux of about 10^{11} photons/(s$\cdot$100 mA$\cdot$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 (Ref. 32) by using heavier lanthanide elements.

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