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Soft-x-ray Faraday rotation at Fe $L_{2,3}$ edges

J. B. Kortright and M. Rice
Lawrence Berkeley Laboratory, University of California, Berkeley, California 94720

R. Carr
Stanford Synchrotron Radiation Laboratory, Stanford University, Stanford, California 94309

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We measured Faraday rotation of linearly polarized soft x rays across the Fe $L_{2,3}$ edges transmitted through an Fe/Cr multilayer in an applied magnetic field; the maximum rotation of 6×10^4 deg/mm is larger than that observed for Fe in other spectral regions. Rotation data agree well with Kramers-Kronig analysis of absorption data measured using left and right circular polarization. A tunable elliptically polarizing undulator source provided linearly and circularly polarized x-ray beams. The tunable multilayer polarizer used extends optical rotation techniques into the 50–900-eV region, with element specificity from core resonances.

Magneto-optical effects were first observed by Faraday (in 1845) and Kerr (in 1876) and helped confirm the electromagnetic nature of light.[1] They have remained of fundamental scientific and practical interest, and with the development of computers are much studied in connection with data storage and retrieval technologies. Magneto-optical effects refer to various changes in the polarization state of light upon interaction with materials possessing a net magnetic moment, including rotation of the plane of linearly polarized light (Faraday, Kerr rotation), and the complementary differential absorption of left and right circularly polarized light (circular dichroism). In the near visible spectral range these effects result from excitation of electrons in the conduction band. Near x-ray-absorption edges, or resonances, magneto-optical effects can be enhanced by transitions from well-defined atomic core levels to transition symmetry selected valence-band states, and hence provide a means of obtaining element- and state-specific magnetic information in multicomponent samples.

In the x-ray range, nonresonant magneto-optical effects have been observed in transition metals[2] and rare earths.[3] While resonant x-ray magneto-optical effects in transition metals were first observed at the Fe K edge,[4,5] they are much larger at the $L_{2,3}$ edges because of the large $2p$ to $3d$ dipole transition-matrix elements. At the $L_{2,3}$ edges, large magnetic circular dichroism (MCD) using near-circular polarization has been observed,[6–8] as have magneto-optical Kerr effects using linear polarization.[9] These effects have generated great experimental and theoretical interest because they offer a means of separating electron-spin and orbital and magnetic dipole contributions to magnetism,[10–13] as well as probing element-specific properties in materials of practical interest. Until now, soft-x-ray magneto-optical studies have had to rely on effects revealed by the *intensities* of signals coming from samples, since polarization or phase-sensitive optics had not been available. We used a newly developed tunable linear polarizer to make the measurement of Faraday rotation in the 500–900-eV range that includes the $L_{2,3}$ edges of the $3d$ transition metals V, Cr, Mn, Fe, Co, and Ni. For an Fe-containing film in an applied magnetic field, Faraday rotation larger than in the visible is observed. This large rotation provides new experimental pathways to obtaining element-specific magnetic information in multicomponent magnetic thin films, and these effects can be used to produce resonant phase retarders.

Measurement of resonant optical rotation at energies up to 900 eV required the development of a continuously tunable linear polarizer for this region (Fig. 1).[14] The device utilizes

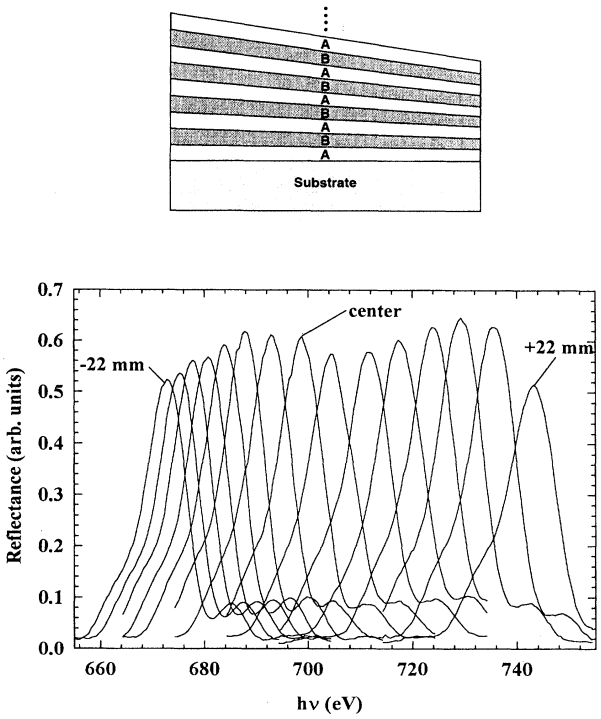

FIG. 1. A laterally graded multilayer was used to produce a tunable linear polarizer by translating the multilayer to satisfy the Bragg condition at the desired energy and the 45° polarizing angle. Measured Bragg spectra taken every 3.1 mm along the tapered multilayer demonstrate the tunability range available for these measurements.

the polarization dependence of specular reflectance from a multilayer interference structure set to reflect at 45°, the Brewster angle in the x-ray region. Operation at the Fe L_2 and L_3 edges (720 and 707 eV, respectively) required a multilayer with very small period $d=\lambda/2\ \sin(45°)\cong 12.3$ Å. We used magnetron sputtered multilayers of W/B_4C which have an s-component reflectance $R_s=0.01$ and an extinction ratio $R_s/R_p\cong 3\times 10^4$ at these energies. The ultrahigh-vacuum polarimeter rotates the multilayer about the beam with fixed incidence angle while monitoring the reflected intensity with a Si diode. Continuously tunability of the polarizer is achieved by translating the multilayer with a laterally graded period perpendicular to the beam to position the Bragg peak at the desired λ. Figure 1 shows that a tunability range of 10% was available for these measurements.

An elliptically polarizing undulator source provided both linearly polarized soft x rays for optical rotation measurements and left and right circular polarization for MCD measurements. The undulator comprises four rows of pure permanent magnets, each producing a sinusoidal field.[15] Two rows are placed above the electron beam and two below. Various states of polarization are selected by moving the rows independently in the longitudinal direction. Beam-line mirrors and a grating monochromator all operate at grazing incidence and have little effect on the polarization of the beam. Using the polarimeter we measured the degree of horizontal linear polarization to be at least 0.98 at the downstream end of the beam line.[16] Because the amount of unpolarized radiation is so small in the linear case and is only slightly dependent on polarization, we infer the degree of circular polarization to be comparable.

The transmission sample was an Fe/Cr multilayer deposited onto a 100-nm-thick silicon nitride semitransparent window. The multilayer nature of this sample is of interest for its magnetic properties,[17] but is not critical for these measurements; any Fe-containing film having a magnetization component along the beam direction will show the effects reported here. The multilayer consisted of 2.0-nm Fe and 1.9-nm Cr magnetron sputter deposited layers repeated for 40 periods with Cr on top and bottom, mounted with its surface normal parallel to the x-ray beam. A 3-kOe field produced by $Nd_xFe_{1-x}B$ blocks was applied along the beam direction and sample normal as illustrated in Fig. 2. This field strength may not be enough to force the sample's magnetization entirely out of plane, but it does result in a sizable out-of-plane component as will be demonstrated. The sample and magnet configuration rotated together through 180° so that the beam could propagate parallel and antiparallel to the applied field. Comparison measurements of optical rotation and MCD used the same sample and magnetic-field configurations, and only changed the polarization of the incident photon beam.

We observe large resonances in the rotation of the plane of horizontal linear polarization on tuning photon energy through the Fe L_3 and L_2 edges, as seen in Fig. 2. Rotation values were obtained by scanning the polarizer over a ±40° range centered about the nominal minimum (with transmission sample removed), and fitting the data to find the deviation of the minimum at each energy. A straight-line background with slope 0.063 deg/eV was subtracted from these rotation values to obtain the data in Fig. 2. This background could result from imperfect alignment of the beam-line optics or the electron trajectory through the four-quadrant undulator.[18] The sense of rotation changes sign with the direction of the applied field, showing large Faraday rotation associated with the Fe in the sample. Normalizing the maximum 4.8° rotation by the $t_{Fe}=80$ nm total Fe thickness (assuming Fe at bulk density in distinct layers) yields a Faraday rotation constant of 6.0×10^4 deg/mm. This is larger than that observed in Fe films in the visible [3.5×10^4 deg/mm at $\lambda=546$ nm (Ref. 1)] and infrared [5.1×10^4 deg/mm at $\lambda=1000$ nm (Ref. 19)] spectral regions, and three orders of magnitude greater than that observed at the Fe K edge at 7112 eV.[5] Since the magnetization of Fe may not be saturated along the beam direction, the observed rotation constant is really a lower limit to the specific rotation of Fe at its L_3 edge. This large soft-x-ray Faraday rotation, in analogy to magneto-optical effects in the visible region, implies that Kerr rotations (on reflection) should be measurable.

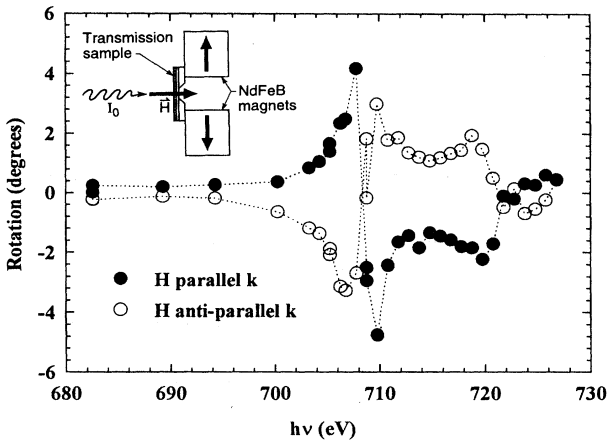

FIG. 2. Faraday rotation measured with a 3-kOe applied field parallel and antiparallel to the beam direction, which is normal to the surface of an Fe/Cr multilayer sample supported on a thin membrane. Lines connect data points.

In the x-ray range the index of refraction is $n(\lambda)=1-\delta(\lambda)-i\beta(\lambda)$ where δ and β are of order 10^{-3} at 700 eV, and are defined in terms of the atomic scattering factor $f(\lambda)$.[20] For forward scattering $f(\lambda)=Z+f'(\lambda)+if''(\lambda)$ where Z is the number of electrons in the atom. $f'(\lambda)$ and $f''(\lambda)$ are the anomalous dispersion corrections related to each other by the Kramers-Kronig transformation. For a multicomponent sample $\delta=\Sigma_i(Z_i f_i')N_i r_e\lambda^2/2\pi$ and $\beta=\Sigma_i f_i'' N_i r_e\lambda^2/2\pi$ where the sum is over atomic species, each having number density N_i, and r_e is the electron radius. The preceding description was developed to account for charge scattering, and is generalized to account for magneto-optical effects by including spin-dependent terms in the scattering cross section.[2,10–13] In the following analysis we assume all variations in δ and β are due to resonant dispersion in charge and spin-dependent scattering in Fe only, since we measure over a small energy range near the Fe edges.

Analysis of MCD absorption spectra using left and right circularly polarized incident beams predicts a Faraday rotation in good agreement with that measured. Absorption data were collected in the total electron yield mode measuring

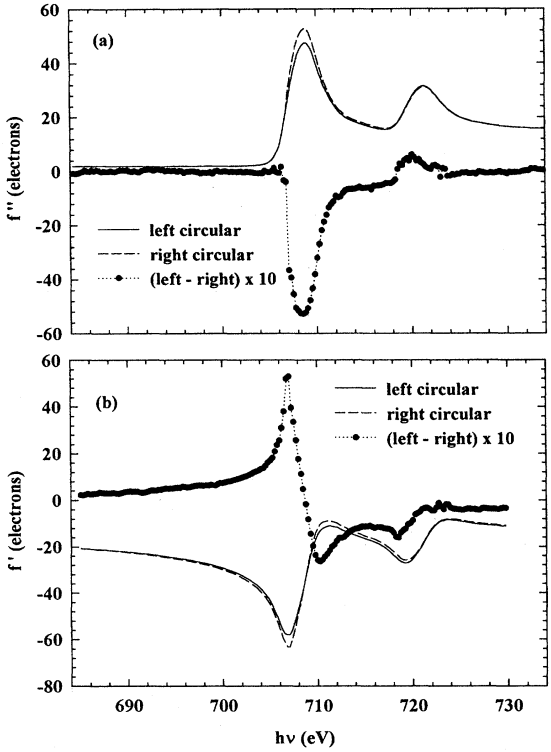

FIG. 3. Absorption data (total electron yield) taken with left and right circular polarization are normalized to f'' in (a), as is $f''_l - f''_r$ that reveals magnetic circular dichroism. The Kramers-Kronig transformation result of the absorption data are shown in f' in (b), as is $f'_l - f'_r$.

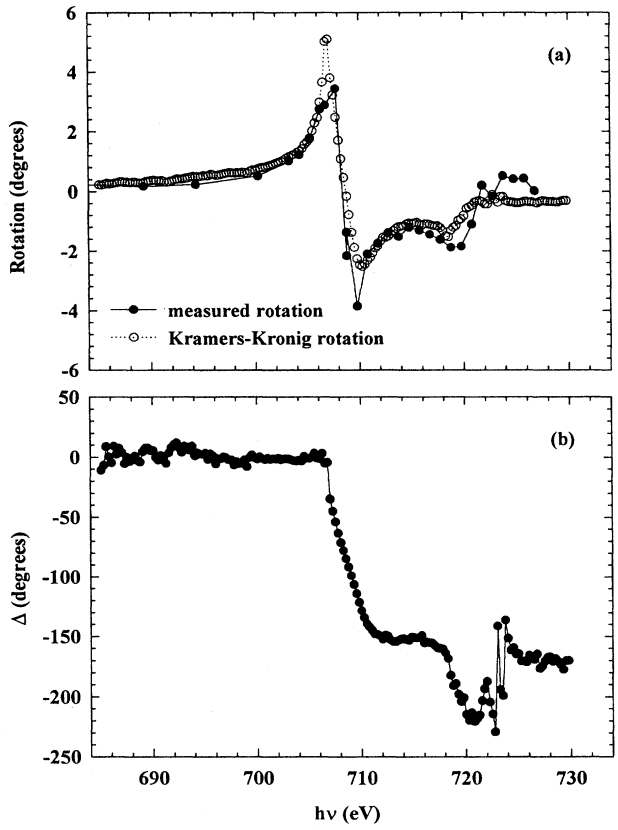

FIG. 4. Comparison of the measured rotation of linear polarization (mirror average of two curves in Fig. 2), and that predicted by the absorption data using left and right circular polarization reveals consistency between the two techniques in (a). In (b) is plotted the equivalent phase difference, or retardation, between the horizontal and vertical components describing the transmitted elliptically polarized beam.

sample current and are shown in Fig. 3(a), along with the difference that clearly shows the circular dichroism. Data were normalized to f'' by fitting and subtracting a smooth background, and scaling the data to calculated atomic values[21] away from the edge region. Direct measurement of Faraday rotation used linear polarization, which is equivalent to a coherent superposition of equal amounts of left (l) and right (r) circular polarization. Kramers-Kronig transformation of $f''_{l,r}$ yields $f'_{l,r}$ [Fig. 3(b)], from which $\delta_{l,r}$ is obtained. The transformation was calculated using the method of Ref. 22 with integration limits of 90 and 35 000 eV. Away from the measured edge region, calculated values of f'' (Ref. 21) were used in the integral, and polarization differences were ignored. The phase difference between the two components on transmission is $(\delta_l - \delta_r)2\pi t_{\mathrm{Fe}}/\lambda$ rad. The Faraday rotation angle $\alpha = (\delta_l - \delta_r)\pi t_{\mathrm{Fe}}/\lambda$. Figure 4(a) compares this calculated rotation based on MCD data with that measured directly using linear polarization. The agreement is quite good, implying that both measurements are similarly representative of the magnetic properties of the sample along the beam propagation direction.

Both the charge and spin scattering undergo extremely large resonances at the $L_{2,3}$ edges because of the efficient coupling to the partially filled $3d$ states. The f'' and f' data in Fig. 3 exhibit very large anomalous dispersion of charge scattering, independent of spin effects. $f'_{l,r}$ are reduced by more than twice the number of electrons in an Fe atom. Such large negative resonance results from the strong dipole-allowed matrix elements for transitions from the $2p_{3/2}$ and $2p_{1/2}$ core levels to a high density of empty, localized $3d$ states as expressed by the white line absorption at the edges. The spin-dependent scattering resonates at the same energies, and its resonance $(f'_l - f'_r)$ is large because of the unequal spin density in the $3d$ states. It is because the $L_{2,3}$ resonances couple directly and exclusively to the $3d$ states responsible for magnetism in Fe that we observe rotation constants larger than in the visible.

Unequal attenuation of the circular components of the initially linearly polarized beam in the rotation measurement induces an ellipticity in the transmitted beam. The ellipticity angle $\omega = (\beta_l - \beta_r)\pi t_{\mathrm{Fe}}/\lambda$ rad. From the data in Fig. 3(a), the maximum $\omega = 5.1°$ at the peak of the L_3 resonance; $\tan(\omega)$ gives the ratio of the minor to major axes of the ellipse described by the transmitted beam, and the ellipse is rotated by α from the horizontal plane of incident linear polarization. An equivalent description of the elliptically polarized transmitted beam is obtained by resolving it into horizontal and vertical linear components that have a relative phase difference, or retardation, Δ where $\tan(\Delta) = \tan(2\omega)/\sin(2\alpha)$.[23] We find large values of Δ in the reso-

nance region as shown in Fig. 4(b). This resonant phase retardation is much larger than that expected to be produced by transmission multilayer interference retarders at this energy,[14] and may provide useful phase retarding elements, albeit with a very limited energy range.

Optical rotation using linear polarization and MCD using circular polarization are self-consistent for the sample studied, and we are led to consider the applicability of the different measurements as element-specific magnetic probes. From a classical optics viewpoint, Faraday rotation measures $(\delta_l - \delta_r)$ and MCD measures $(\beta_l - \beta_r)$ along the propagation direction; the two quantities are related by the Kramers-Kronig transformation. Thus measurement of one yields the other, assuming $\delta_{l,r}(\lambda)$ and $\beta_{l,r}(\lambda)$ are sufficiently well known away from the energy region of interest so that transformation errors are negligible. Kerr effects only measuring reflected intensity do not separate δ and β, but measure a signal influenced by both quantities. MCD measurements in the soft x ray have required incident light with a large degree of circular polarization, while optical rotation requires only linearly polarized beams. MCD measured using electron yield techniques is limited by electron escape depths to near surface sensitivity. Optical rotation measured in transmission is sensitive to the bulk of thin films, and in reflection has penetration depths ranging from 3 to hundreds of nm with the incident angle below and above the critical angle, respectively. Optical detection is more compatible than electron detection with measurements in varying magnetic fields.

To summarize, we have measured resonant Faraday rotation on transmission through Fe-containing films in the soft x ray and found rotation constants larger than found in other spectral regions. We have used left and right circular polarization to measure the absorptive part of the magneto-optical response from the same sample, and found the two measurements in good agreement for the sample studied. Soft-x-ray optical rotation in both transmission and reflection, using linear incident polarization, thus provides a complementary experimental pathway to obtaining element-specific magnetic information on a wide variety of materials. The newly developed optics enabling these measurements are applicable over at least the 50–900-eV region, covering absorption resonances of numerous elements. The extension of various polarization-resolved spectroscopic techniques into the soft-x-ray region is thus possible.

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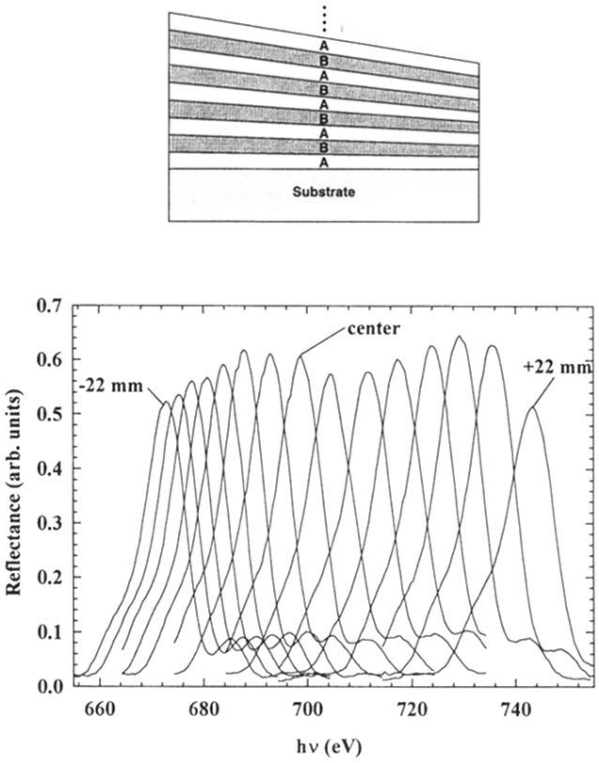

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