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Citation: Appl. Phys. Lett. **71**, 1446 (1997); doi: 10.1063/1.119932 View online: http://dx.doi.org/10.1063/1.119932 View Table of Contents: http://apl.aip.org/resource/1/APPLAB/v71/i11 Published by the American Institute of Physics.

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Soft x-ray circular polarizer using magnetic circular dichroism at the Fe L_3 line

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(Received 24 February 1997; accepted for publication 9 July 1997)

Linearly polarized soft x-rays are converted to elliptical polarization at the Fe L₃ line (707 eV) using magnetic circular dichroism (MCD) on transmission through thin Fe films. A linear polarizer measured the transmitted polarization at different incidence angles to vary as expected from a model for in-plane magnetization, and also to exhibit a weak MCD effect at normal incidence interpreted to originate from perpendicular interface anisotropy. An MCD signal from a downstream Fe film was produced by switching the helicity of x-rays transmitted through an upstream circular polarizer. Practical considerations for optimizing the production of circular polarization are discussed, and synchrotron radiation applications using these circular polarizing filters are suggested. [S0003-6951(97)00737-7]

Optical production of circularly polarized photons in the soft x-ray region (150–3000 eV) using synchrotron radiation presents a challenge. Quarter-wave retarders, common in the near-visible region, efficiently convert linear to circular polarization. The associated principles can be extended up to at least 150 eV with use of multiple-reflection mirrors^{1–4} and birefringent transmission multilayers.^{5–7} Above 3 keV, quarter-wave retarders use the birefringence of near perfect crystals.⁸ The intermediate region will eventually be serviced by expensive elliptically polarizing undulator beamlines because of the lack of quarter-wave retarders.

We offer here a low-cost optical device that can be retrofitted into existing conventional undulator beamlines to convert linear to elliptical polarization at selected energies. The operational principle derives from the strong magnetic circular dichroism (MCD) at the $L_{2,3}$ edges of the 3*d* ferromagnets and the $M_{4,5}$ edges of the rare earths; the differential absorption of circular components of opposite helicities transforms an incident linearly polarized beam into an elliptically polarized beam. The principle was advanced by Goedkoop *et al.*,⁹ who considered rare earth ions in films assumed at low temperatures and in high fields to maximize the MCD effects. We believe that this letter contains the first experimental demonstration of the concept, using a Fe film at ambient temperature and in a modest field.

This resonant elliptical polarizer consists of a magnetized transmission Fe film through which linearly polarized soft x-rays propagate with wave-vector **k**. The photon energy is tuned to the maximum of the MCD response, at the peak of the FeL₃ white line. The MCD is maximized when the magnetization **M** is collinear with **k**, and varies as $\mathbf{k} \cdot \mathbf{M}$. For Fe films **M** is typically in plane, requiring non-normal incidence. A 48 nm thick Fe film was sputtered onto a 160 nm thick silicon nitride membrane, followed by a 2 nm layer of SiC to passivate the top surface from oxidation. Such polycrystalline films exhibit $\langle 110 \rangle$ texture, and grain size comparable to film thickness. Linearly polarized light from the undulator third harmonic on beamline 7.0 at the Advanced Light Source was monochromatized to $\Delta E = 0.3 \text{ eV}$ and shone through the sample with a variable incidence angle θ . NdFeB magnets provided a 70 Oe in-plane field to saturate **M** in the direction that maximized **k**·**M**. The magnetized film was placed upstream of a tunable multilayer linear polarizer. Both the MCD circular polarizer and multilayer linear polarizer were housed in an in-line polarimeter in which the linear polarizer can be retracted to pass the circularly polarized beam downstream to experiments.¹⁰

The transmission absorption spectrum at normal incidence with linearly polarized undulator radiation exhibits strong L_3 and L_2 lines at 707 and 720 eV, respectively (Fig. 1). Data were normalized to the imaginary part of the atomic scattering factor $f''(h\nu)$ above and below the edges, and then to the imaginary part of the refractive index $n(h\nu) = 1$ $-\delta(h\nu) - i \cdot \beta(h\nu)$ assuming bulk density for Fe (see Ref. 11 for details). The polarization-averaged absorption coefficient, $\mu = 4 \pi \beta / \lambda$, yields an absorption length of 18 nm at the L_3 peak, increasing to 600 nm immediately below the line. The MCD signal $\beta_+ - \beta_-$ has maxima of opposite sign at the L_3 and L_2 lines, and the larger peak at the L_3 line is useful as a circular polarizer. Here β_+ (β_-) is the absorption index for the right (left) circular component. Since linear polarization is a coherent superposition of equal parts left and right circular polarization, $\beta = (\beta_+ + \beta_-)/2$.

Rotating the linear polarizer about the transmitted beam yields the first three of four Stokes parameters describing its polarization. The multilayer polarizer suppresses possible harmonics and out-of-band stray radiation by at least $10^{-2}-10^{-3}$ to yield a clean measurement of the polarization of the transmitted fundamental radiation, even with the fundamental strongly absorbed by the L_3 line as required in the operation of this polarizing filter. The intensity measured in these azimuthal scans is

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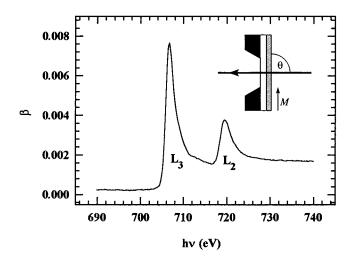


FIG. 1. The transmission absorption spectrum of a magnetized 48 nm Fe film measured with linear incident polarization.

$$I(\alpha) = S_0(R_S + R_P)/2 + 0.5(R_S - R_P)(S_1 \cos 2\alpha) + S_2 \sin 2\alpha),$$
(1)

where α is the azimuthal angle, $R_S(\approx 0.01)$ and $R_P(\approx 1 \times 10^{-5})$ are the reflectivities for *s* and *p* linear components, and S_0 , S_1 , S_2 are the first three Stokes parameters. The degree of linear polarization $P_L \equiv (S_1^2 + S_2^2)^{1/2}/S_0$ is measured directly, and we deduce the degree of circular polarization $P_C \equiv S_3/S_0$ from $P_L^2 + P_C^2 = 1$ assuming no unpolarized radiation is present. Polarization measurements through the film at normal incidence and 691 eV (well below the L_3 line) yielded $P_L^2 = 0.99$, consistent with the undulator design to produce linear polarization. The difference of this P_L^2 from 1.00 is near the limit of measurement accuracy, but could also include any circular or unpolarized radiation present in the beam, and thus sets an upper limit to a possible unpolarized component in the incident radiation.

Figure 2 shows polarimetry data at several angles, together with fits of Eq. (1) to these data. Data and fits were normalized by $S_0(R_S+R_P)$ to a common scale from 0 to 1 on which perfect circular polarization would yield a flat line at 0.5. Decreasing linear polarization is evident as θ decreases from 90°. The P_C^2 values obtained from these data are plotted in Fig. 3. At normal incidence $P_C^2=0.055$ at the MCD peak (707 eV), while $P_C^2 \leq 0.01$ below the peak (691 eV).

To explain this increase in normal incidence P_C^2 when tuned to the MCD peak, we must assume either a small normal component of **M**, or a resonant depolarization of the transmitted beam. A normal component of **M** is not unexpected. Perpendicular anisotropy could arise from inhomogeneities internal to the film.¹² Interface effects could also yield this perpendicular anisotropy through roughness,¹³ reduced symmetry,^{14–16} or microstructural effects (intermixing, strain) that might alter interatomic structure and hence magnetization through magnetocrystalline or other anisotropy mechanisms.¹⁷ Resonant magneto-optical depolarization of the transmitted beam is not expected for saturated films, and is not consistent with transmission MCD¹⁸ and Faraday rotation¹¹ measurements through Fe films. Nor would depo-

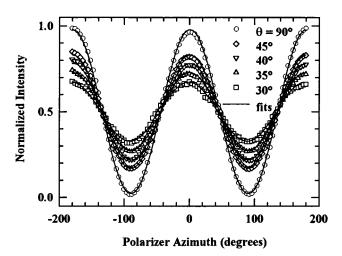


FIG. 2. Polarizer scans of the beam transmitted through the Fe film at different incidence angles at the L_3 peak.

larization be expected to exhibit the specific angular dependence in Figs. 2 and 3 that is well described by a MCD model assuming in-plane **M**.

Away from 90°, P_C^2 varies according to the expected $\mathbf{k} \cdot \mathbf{M}$ behavior for in-plane \mathbf{M} , verifying that iron moments have predominantly this orientation and providing the magnitude of the MCD. Several calculated quantities plotted in Fig. 3 derive from the measured absorption index β (Fig. 1) for linear incident polarization. The absorption coefficients for right (+) and left (-) circular polarizations are $4\pi(\beta)$ $-\Delta \cos \theta / \lambda$ and $4\pi (\beta + \Delta \cos \theta) / \lambda$, respectively, where Δ $=(\beta_{+}-\beta_{-})/2$ is the MCD deviation from the polarizationaveraged absorption, and the cosine factor accounts for inplane magnetization (assumed complete). The pathlength is $t_0/\sin\theta$ with t_0 the film thickness. For this 48 nm film at the L_3 peak $\mu t_0 = 2.66$. The transmission versus θ for the $T_{+/-} = \exp[-4\pi(\beta - /$ opposite helical components $+\Delta \cos \theta t_0/(\lambda \sin \theta)$]. Figure 3 shows the relative transmissions, which are these expressions normalized by their value

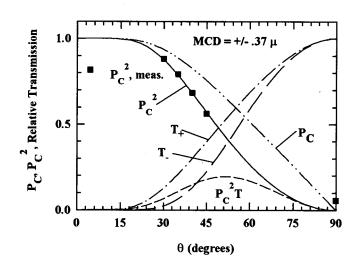


FIG. 3. For $h\nu$ at the L_3 MCD peak, several quantities are displayed on the same vertical scale as they depend on the incidence angle to the circular polarizer. Symbols represent measured values for P_C^2 . Lines represent calculated quantities described in the text.

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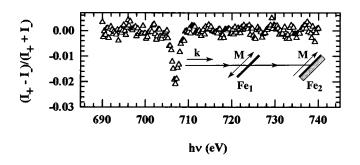


FIG. 4. The asymmetry ratio in photocurrent spectra I_+ and I_- from a downstream Fe film (Fe₂) taken in the beam transmitted through an upstream Fe film (Fe₁) set with opposite signs of $\mathbf{k} \cdot \mathbf{M}$ to produce resonant elliptical polarization with opposite helicities (primarily at the L_3 line).

at 90°. Absorption by the silicon nitride membrane is included using a measured $\mu t_0 = 0.33$. The polarizationaveraged transmission $T = (T_+ + T_-)/2$. The degree of circular polarization $P_C = (T_+ - T_-)/(T_+ + T_-)$. Varying only Δ to fit the measured polarization data yields $\Delta = 0.37\beta$. This represents the magnitude of the MCD effect for **k**||**M**, and agrees with transmission MCD measurements of Ref. 18.¹⁹

To further confirm transmitted elliptical polarization, MCD from a downstream Fe film was measured using radiation transmitted through an Fe elliptical polarizer. The MCD polarizer was a magnetized 20 nm Fe film (Fe₁) at 45° , and absorption spectra via photocurrent were measured from a downstream magnetized 44 nm Fe film (Fe₂) also at 45°. Figure 4 shows the experimental geometry and the asymmetry ratio $(I_+ - I_-)/(I_+ + I_-)$ where I_+ and I_- are the raw spectra taken with opposite signs of $\mathbf{k} \cdot \mathbf{M}$ at Fe₁. The peak in the asymmetry ratio at the L_3 peak demonstrates that transmission of linear polarization through Fe₁ yields elliptical polarization. The measured asymmetry ratio is 0.022, while the above model with the same Δ predicts 0.045. Removal of roughly half of the smooth pre-edge background in the individual spectra brings the measured MCD into agreement with that predicted by the model. Background contributions are common in photocurrent spectra, and are expected from higher order or stray radiation from the grating. No MCD is seen at the L_2 edge, consistent with the reduced absorption and MCD at the L_2 peak.

Utilization of these elliptical polarizing line filters will require careful consideration of experimental details. In Fig. $3 P_C^2 T$ is maximum near 50°, providing an operating point assuming this figure of merit is relevant. Operating here, T is calculated to be 0.046; more than 90% of the beam is absorbed in the 48 nm polarizing filter. For M in-plane efficiency improves for thinner films operating at more grazing angles. Reducing thickness by half and operating at θ = 22.5° yields the same pathlength and increases in T to 0.060 and in P_C from 0.67 at 50° to 0.83. Even more efficient would be films dominated by perpendicular anisotropy operating at normal incidence. Absorption losses are unavoidable, and can never be less than 50% when Δ has its maximum value of $1.0 \cdot \beta$. Since absorption losses greater than 90% for Fe and other materials can be expected at real MCD lines, less attenuated higher harmonics, scattered radiation, or other background signals can significantly reduce polarization contrast. Maximizing circular polarization requires high enough energy resolution to saturate the MCD signal. While the sense of helicity can be switched mechanically as done here, an ac applied field could rapidly modulate the transmitted helicity for various measurements.

These results demonstrate magneto-optical elliptical polarizing filters in a region of the soft x-ray spectrum previously devoid of such optics. The resonant nature of soft x-ray MCD limits the applicability of these polarizers to specific energies spanning at least 707-1500 eV utilizing magnetic transition and rare earth metals. This apparent drawback is no disadvantage for applications relying on MCD contrast such as magnetic x-ray microscopes,²⁰ where these are precisely the energies desired for operation. Other applications that may benefit include photoemission spectroscopy, photoelectron diffraction, and any other measurements requiring monoenergetic, nontunable circular polarization. The weak MCD observed at normal incidence indicates that resonant soft x-ray magneto-optical effects are large enough to reveal signals originating from only one or two buried interfaces. This work demonstrates that direct soft x-ray polarization measurements are quite sensitive and useful in such studies.

S.K.K. acknowledges support from the Korea Research Foundation 1996 Postdoctoral Fellowship Program. This work was supported by the Director, Office of Energy Research, Office of Basic Energy Sciences, Division of Materials Science, of the U.S. Department of Energy under Contract No. DE-AC03-76SF00098.

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