Dichroic interference effects in circularly polarized soft-x-ray resonant magnetic scattering

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Dichroic interference effects were observed in circularly polarized soft-x-ray reflectivity at the Co $L_{2,3}$ edges of a magnetic multilayer. The data show a maximum asymmetry ratio of 80%, greater than the degree of circular polarization of the incident light, a changeover in the dichroic characteristics with increasing grazing angle, and an inverted interference pattern in the angular dependence of the asymmetry ratio. These observations are explained successfully by a classical electromagnetic model calculation which includes the complete polarization state of the incident light and spatial interference within the multilayer.

Recently, there have been extensive experimental and theoretical efforts in x-ray magnetic circular dichroism (MCD), both in the hard-x-ray[1] and soft-x-ray regions.[2] MCD effects much greater than those observed in the visible region have been reported. For example, asymmetry ratios in absorption coefficients in the range of 20–40 % have been observed at the $L_{2,3}$ edges of $3d$ transition metals and the $M_{4,5}$ edges of rare-earth elements. These large asymmetry ratios are mainly due to the strong dipole-allowed $p \rightarrow d$ and $d \rightarrow f$ transitions and the large spin-orbit coupling in the deep core levels. The large enhancement of the MCD effect coupled with the element specific nature of core electron excitation make MCD in the x-ray region an important experimental technique in the study of a wide range of magnetic systems, including impurities, alloys, as well as surfaces, thin films, and multilayers. Concurrent to the development of x-ray MCD, there has also been intensive activity over the last few years in x-ray resonant magnetic scattering using linearly polarized x rays.[13] Dramatic enhancement of the magnetic scattering cross sections and complex polarization dependence have been observed for both antiferromagnets and ferromagnets. Again, the large enhancement is ascribed to the large spin-orbit interaction, spin polarization of the conduction band, and the large resonant scattering cross section. In fact, resonant magnetic scattering and MCD are closely related through the optical theorem, which relates the absorption coefficient to the imaginary part of the forward scattering amplitude.

Despite the similarity mentioned above, x-ray MCD is essentially a spectroscopic tool with strong dichroic effect, while resonant magnetic scattering can provide structural information through angular dependence of the scattered intensity and the asymmetry ratio in the scattered intensity. Thus, it is very appealing to combine these two attributes to provide a means for the study of magnetization depth profile of magnetic thin films, multilayers, and surfaces. An analysis of the resonant magnetic scattering amplitude from a single magnetic atom using circularly polarized x rays shows that asymmetry ratios comparable to or even greater than those observed in MCD can be achieved over an extended angular range.[4]

In this paper we report the results of a circularly polarized soft-x-ray resonant magnetic scattering study of a Co-containing magnetic multilayer. Asymmetry ratios as high as 80% were observed in the specular reflectivity near the Co $L_{2,3}$ edges. These asymmetry ratios are not only much larger than the MCD effect from the same sample, but even greater than the degree of circular polarization of the incident light. In addition, there is a dramatic change in the energy dependence of the circular dichroic effects with increasing grazing angle, and an interference pattern in the angular dependence of the asymmetry ratio which is inverted from that in the reflectivity curve. Potential applications of these effects are briefly discussed.

The measurements were conducted at the AT&T Bell Laboratories Dragon beamline at the National Synchrotron Light Source. The beamline configuration for collecting circularly polarized soft x rays has been described previously.[5] The degree of circular polarization was set to 75% and the photon energy resolution was set at 1 eV. A vacuum compatible θ-2θ spectrometer was used in the experiment.[6] The angular resolution of the spectrometer, defined by a set of slits between the sample and the detector, was set to 1°. A gas proportional avalanche counter with an overall efficiency of 10% and an energy bandwidth of ~40% was used as the detector.[7] The sample was magnetized by an electromagnet capable of delivering a field up to 400 G. The sample used in the experiments was a 37-Å bcc Co(001) thin film epitaxially grown on GaAs(001) with a 560-Å buffer layer of ZnSe. In order to establish the bcc Co structure, a 5-Å Fe seed layer was deposited on the ZnSe prior to Co depositon.[8] The Co film was capped with 36 Å of Al for protecting the Co layer from oxidation. In the calculations mentioned below, the Al top layer was assumed to be fully oxidized. The thickness of each layer was determined by using an x-ray fluorescence thickness monitor. The geometric arrangement of the measurements is shown in Fig. 1. The sample was mounted with the easy axis along the magnetization direction of the electromagnetic. The magnetization vector of the sample lies both in the scattering plane and in the plane of the multilayer film. And the complete magnetization of the Co layer was checked by the element-specific magnetic hysteresis technique.[9]

Figure 2 shows a series of reflectivity curves as a function of photon energy, taken at different grazing angles where the solid and broken lines (denoted as I^+ and I^-) are reflectivity curves measurd with the spin of the incident photons parallel and antiparallel, respectively, to the sample magnetization. Also included in the figure are the absorption spectra obtained from the same sample in the same magnetization configuration. These spectra were measured by alternating between opposite applied saturating magnetic field at each photon energy, while the circular polarization was held constant. Measurements taken with constant magnetic field and alternating circular polarization showed identical results. In the following, an asymmetry ratio, defined as $(I^+-I^-)/(I^++I^-)$, is used to describe the magnitude of the dichroic effect.

As the grazing angle increases from small angle (2°–3°) to large angle (10°–25°), the reflectivity around the absorption edges changes from dips to peaks, and the dichroic characteristic, i.e., the difference betwen I^+ and I^-, changes from shifts in the energies of the dips to variations in the peak intensities. The asymmetry ratios were found to be 44 and 80% at the peak of the L_3 edge in the 10° and 25° reflectivity spectra, respectively, while the MCD effect in the absorption spectra is only about 20% at $\theta=37°$ (equivalent to 25% at 0° measuring angle). More importantly, the asymmetry ratio of 80% is even greater than the degree of circular polarization of the incident light which was set at 75%. To further study the angular dependence of the observed effects, Fig.

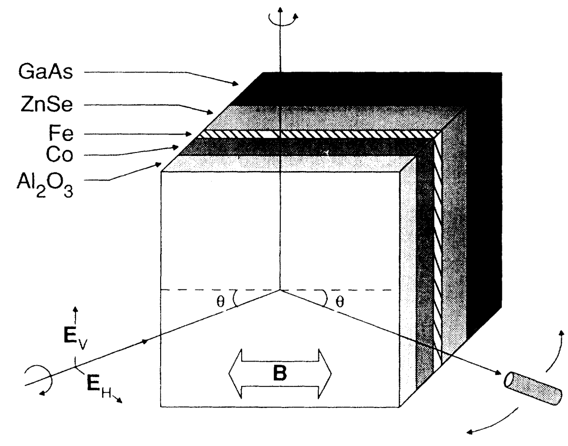

FIG. 1. Geometric arrangement of the experiment. The magnetization vector of the sample lies both in the scattering plane, defined by the incident and the scattered beams, and in the plane of the multilayer sample. E_V and E_H are the vertical and horizontal electric-field components of the incident photons.

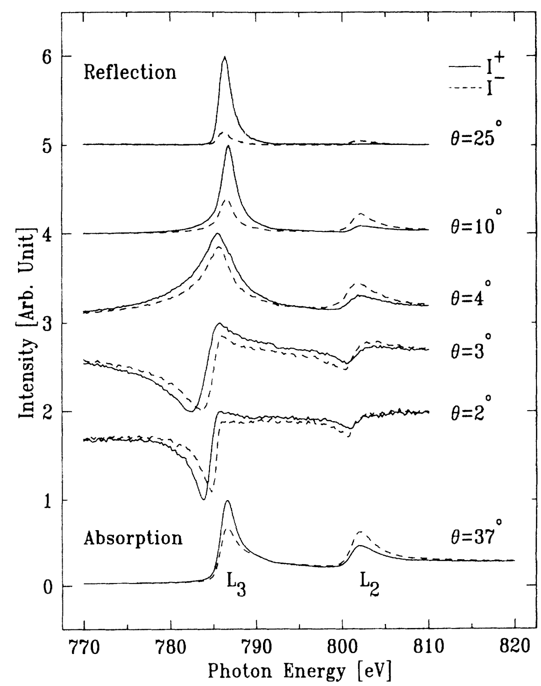

FIG. 2. Reflectivity as a function of photon energy measured at a series of grazing angles, and two opposite magnetization directions (denoted by the solid and broken lines). Also included in the figure are the two absorption spectra obtained from the same sample and same magnetization directions.

3 shows the two reflectivity curves, I^+ and I^-, as well as the asymmetry ratio measured at the photon energy of the L_3 absorption peak (786.5 eV) as a function of grazing angle. The reflectivity curves are modulated by an interference pattern, typical for thin films with thickness comaparble to the wavelength of the incident photons. In contrast to the reflectivity curves, the asymmetry ratio shows an interference pattern out of phase to that in the reflectivity curves, i.e., the maxima in the interference pattern of the asymmetry ratios coincide roughly to the minima in that of the reflectivity, and increases from a few percent to as high as 80%.

In order to provide a quantitative explanation for the circular dichroic effects observed in this work and to predict circular dichroism in resonant magnetic scattering under various experimental conditions, a model calculation for magnetic multilayer reflectivity was developed. To summarize briefly, within classical electromagnetic theory, the propagation of light in an ideal multilayer medium, i.e., one in which the interface planes are perfectly flat, smooth, and parallel to one another, can be solved exactly if the dielectric and magnetic permeability tensors are known.[10] In this particular case, a five-layer system composed of 36 Å of Al_2O_3, 37 Å of Co, 5 Å of Fe, 560 Å of ZnSe, and a GaAs substrate was considered. For photon energies around the Co $L_{2,3}$ edges, the dielectric tensors of all layers other than Co are diagonal with an isotropic dielectric parameter, i.e., $\varepsilon_{xx}=\varepsilon_{yy}=\varepsilon_{zz}=\varepsilon$. Both the real and imaginary parts of these dielectric constants were obtained from the calculated atomic scattering factors.[11] For the Co layer, an additional parameter q is needed to account for the magnetic circular dichroic effects, as manifested through the nonvanishing off-diagonal elements ($\varepsilon_{xy}=-\varepsilon_{yx}=iq$). Since no reliable calculated values for both ε and q near the $L_{2,3}$ edges of Co are available, the following approach was taken: the imaginary parts of ε and q were set to be proportional to the sum, $(I^+ + I^-)$, and the difference, $(I^+ - I^-)$, of the two absorption spectra, respectively. Calculated atomic photoabsorption cross sections of Co for photon energies below the L_3 edge and above the L_2 edge were used to scale the measured spectra to absolute cross sections. The real parts of ε and q were then calculated by using the Kramers-Kronig transformation. The magnetic permeability tensors were set to unity in the calculation.

Figure 4 shows the calculated I^+ and I^- spectra corresponding to the measured spectra shown in Fig. 2. Except for some discrepancy in the L_3 to L_2 peak (or dip) intensity ratios, the calculated spectra are in very good agreement with the measured ones. The two different types of dichroic characteristic, i.e., the energy shifts in the dip positions for small grazing angles and the intensity variations in the peaks for large angles, have been accounted for quantitatively. The calculated reflectivity and asymmetry ratio as a function of grazing angle are shown in Fig. 5. Again, the agreement between the calculated and the measured spectra is very good, except that the measured interference patterns are not as sharp as the calculated ones. The smearing-out of the interference pattern in the measured spectra might be due to structural roughness or other imperfections at the surface or the interfaces of the sample. In fact, model calculations have shown that these interference patterns are very sensitive to the layer thickness, even at the level of a few Å.

We also have found from experiment (by collecting synchrotron radiation at different vertical angles) and from calculation that the asymmetry ratio in reflection is in general nor proportional to the degree of circular polarization of the incident light, in contrast to the MCD effects in absorption. The calculations here were per-

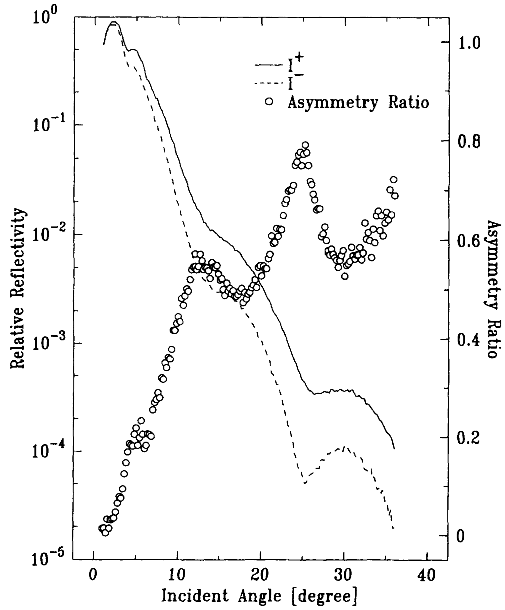

FIG. 3. Reflectivity as a function of grazing angle measured at the peak of the Co L_3 edge (786.5 eV). The solid and broken lines denote the two opposite magnetization directions used in the measurements. Also included in the figure is the asymmetry ratio derived from the two reflectivity curves.

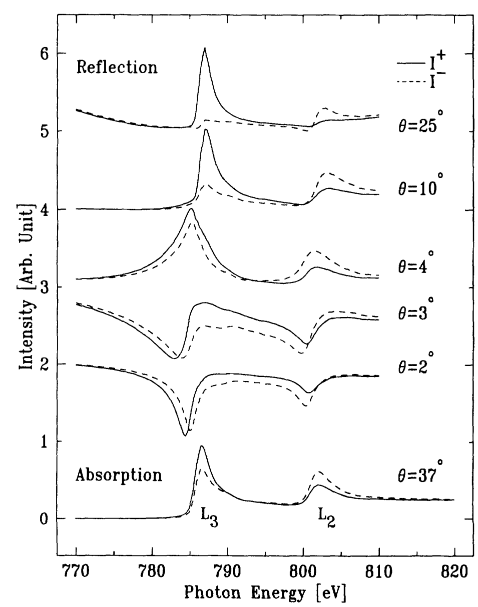

FIG. 4. Calculated reflectivity and absorption as a function of photon energy. The notation for the two opposite magnetization directions and the grazing angles are the same as those shown in Fig. 2.

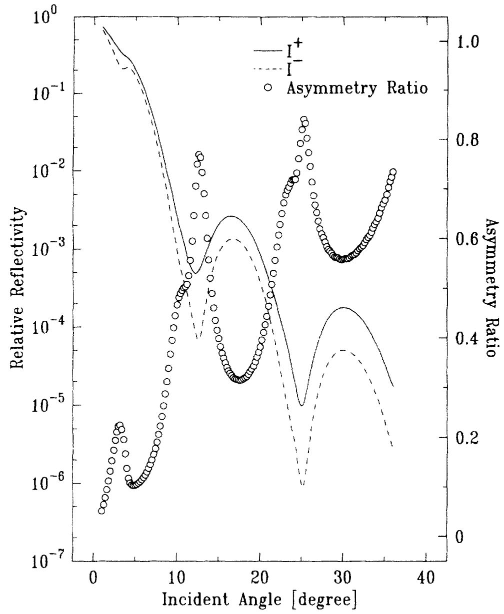

FIG. 5. Calculated reflectivity and asymmetry ratios at the peak of the Co L_3 edge. The notations are the same as those shown in Fig. 3.

formed with the polarization vectors set at $E_h \pm 0.45 i E_v$ (equivalent to 75% degree of circular polarization), to simulate the polarization state of the incident light used in the measurements. Asymmetry ratios calculated with 100% circular polarization, and then multiplied by the degree of circular polarization gave rather poor agreement with the measurements. In fact, in this particular case, at $\theta = 25°$ 75% circular polarization gives nearly the same calculated asymmetry ratios as 100% circular polarizaation.[12]

In summary, we report circularly polarized resonant magnetic scattering measurements. Surprisingly large magnetic circular dichroic effects and interesting interference patterns were observed, and successfully explained by a model classical electromagnetic calculation. The large enhancement of the dichroic effect in circularly polarized reflectivity can be used to extend the sensitivity of MCD measurements to magnetic systems with even smaller magnetic moments. This work also demonstrates that these effects are very sensitive to the complete polarization state of the photons and to spatial interference within the multilayers. The high sensitivity of these effects to the polarization state of the photons can be utilized to exploit the new capability offered by the recent development in variable-polarization synchrotron-radiation insertion devices. The large dichroic effects exhibited over a wide angular range, and its sensitivity to layer thickness and interface roughness, can provide a means for the study of element-specific magnetic depth porfile of surfaces, thin films, and multilayers. It should also be possible to separate magnetic rougness from nonmagnetic roughness of interfaces in magnetic multilayers by detailed measurement and modeling of the interference patterns in reflectivity and asymmetry ratio.

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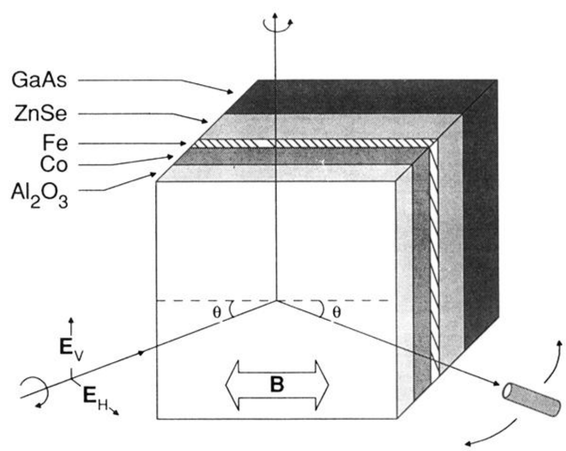
[12]A general equation for the dependence of the asymmetry ratio in reflection on the degree of circular polarization is found to be $(I_1 \times P_c)/(I_2 + I_3 \times \sqrt{1-P_c^2})$, where I_1, I_2, and I_3 are functions of the diagonal and off-diagonal elements of the dielectric tensor, and the incident angle. In this particular case, I_3 is of the same order of magnitude as I_2 and opposite in sign, which leads to the nonlinear dependence of the asymmetry ratio on P_c, and an asymmetry ratio greater than P_c at certain incident angle. A detailed account of the derivation of this relation will be published elsewhere.



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