Magnetic-Resonance Exchange Scattering at the Iron L_{II} and L_{III} Edges

C. Kao, J. B. Hastings, E. D. Johnson, D. P. Siddons, and G. C. Smith
Brookhaven National Laboratory, Upton, New York 11973

G. A. Prinz
Naval Research Laboratory, Washington, D.C. 20375
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Specular reflectivity of p-polarized light near the iron L_{II} and L_{III} absorption edges was measured from a single-crystal iron film with an external magnetic field perpendicular to the scattering plane. Large changes in reflectivity were observed upon reversal of the direction of the magnetic field. We attribute this resonant magnetization-sensitive effect to the interference between magnetic and nonmagnetic contribution to the resonant scattering. Similar effects can be expected in other ferromagnetic and ferrimagnetic systems.

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X-ray scattering has become an important experimental technique in the investigation of magnetic materials.[1] Of particular importance is the recently observed large resonant enhancement of the magnetic scattering cross section near an absorption threshold.[2,3] In favorable cases the magnetic scattering cross section can be comparable to that of charge scattering. For example, measurements in holmium by Gibbs *et al.*[2] have shown an enhancement of over fifty in the magnetic satellite intensities as the photon energy is tuned across the L_{III} edge ($2p_{3/2}$ to $5d$, 8.067 keV). Similar experimental studies at the M_{IV} and M_V edges of UAs ($3d_{3/2}$ to $5f$, 3.728 keV and $3d_{5/2}$ to $5f$, 3.552 keV, respectively) by Isaacs *et al.*[3] have demonstrated even larger resonant enhancements of magnetic Bragg reflections. At the M_{IV} edge the scattering intensity of the $(0,0,\frac{5}{2})$ reflection is 1% of the intensity of the (0,0,2) charge reflection, which is an enhancement of 7 orders of magnitude over the magnetic scattering intensity measured far from the edge. These observations can all be explained by resonant exchange scattering,[4] in which large resonant enhancements result from strong electric multipole transitions from atomic core levels to unoccupied states above the Fermi level, with the magnetic sensitivity arising both from the spin polarization of the partially occupied states and from exchange splitting of the unoccupied states.

The previous work[2,3] on Ho and UAs studied the energy dependence of Bragg peaks in these antiferromagnetic structures which are purely magnetic in origin. For the $3d$ transition metals large resonant enhancements are predicted for the $L_{II,III}$ edges.[4,5] In these materials, however, the transitions all occur at wavelengths which are outside the limiting sphere for Bragg reflections. However, measurement of the specular reflectivity (which is not restricted in wavelength) from ferromagnetic or ferrimagnetic samples should also be sensitive to resonant exchange scattering. In this work, we measure the specular reflectivity utilizing a linearly polarized photon beam with its polarization vector in the scattering plane (p-polarized light), and an external magnetic field perpendicular to the scattering plane.

The effect of resonant exchange scattering is manifested as changes in the specular reflectivity upon reversal of the direction of the magnetic field.[6] To illustrate the effect better, an asymmetry ratio R defined as

$$R=\frac{I^{+}-I^{-}}{I^{+}+I^{-}}$$

was derived as a function of photon energy, where I^{+} and I^{-} are the measured specular reflectivity with the applied magnetic field parallel and antiparallel to the cross product of the polarization vectors of the scattered and the incident beams. An asymmetry ratio as large as 13% was observed near the Fe L_{II} and L_{III} edges in the present configuration, and the effect should be even larger at larger scattering angles.

The results discussed in this Letter were obtained on the State University of New York, Stony Brook, beam line U-15 on the uv ring at the National Synchrotron Light Source, Brookhaven National Laboratory.[7] The optical system consists of a gold-coated toroidal grating 3 m from the source which disperses in the vertical direction and focuses down to an exit slit 9 m from the source. The beam is then redirected to the horizontal plane by a gold-coated flat mirror. The beam passes through a 1500-Å-thick Al window to a pair of slits which define the beam dimension in the horizontal direction. The spread in photon energy is about 4 eV (FWHM), and the $L_{II,III}$ edges (714 and 727 eV, respectively) are well above the 500-eV critical energy of the ring, so the beam incident on the sample is highly linearly polarized.

The sample is a 35-Å-thick single crystal of Fe(110) (17 atomic layers) deposited by molecular-beam epitaxy on a (110) gallium-arsinide substrate with a thin zinc-selenide overlayer. It was clamped in the jaws of an electromagnet with its [100] direction, and the direction of the applied field, collinear with the axis of a precision angle drive. The magnet supplies a field of 400 G with

no sample present. The spectrometer axis was perpendicular to the plane of polarization, and had a fixed scattering angle 2θ of 35.2° to the detector. This geometry was chosen to maximize the observable effect predicted by Hannon *et al.*,[4] within the constraints of existing equipment. After scattering from the crystal, the beam passes through a pair of horizontal slits with 1° acceptance.

A low-pressure, parallel-plate avalanche chamber[8] was used to detect the scattered x rays. The chamber has a 2.5-μm aluminized Mylar window and uses a 50% argon-ethane gas mixture at 28 Torr. At 700 eV the overall efficiency of the detector is roughly 10% and the energy resolution is about 40%. With 150 mA of stored beam the count rate in the peak of the reflectivity was 4.5 kHz.

The data shown in Fig. 1 are the sum of eight reflectivity measurements for the magnetic field applied perpendicular to the scattering plane in opposite directions (I^+ and I^-). The peak in the I^- spectrum at 714 eV is 785000 counts, and the background at 650 eV is 15400 counts. The asymmetry ratio, derived as defined above, is superimposed on the figure (with error bars set by Poisson statistics), although the effect is quite obvious in the raw data. Variations in the order in which the data are taken, or initial direction of magnetization, have no effect on the results.

For comparison with experimental results, specular reflectivity in the vicinity of Fe L_{II} and L_{III} edges was also calculated. Following Hannon *et al.*,[4] the scattering amplitude due to resonant exchange scattering in the present experimental configuration, considering only dipole transitions, is given by

$$f_{E1}^{(\mathrm{res})}=\frac{3\lambda}{8\pi}\{(\mathbf{e}_f\cdot\mathbf{e}_o)[F_{1,1}+F_{1,-1}] -i(\mathbf{e}_f\times\mathbf{e}_o)\cdot\mathbf{z}_j[F_{1,1}-F_{1,-1}]\}\,. \quad (1)$$

In Eq. (1), $\mathbf{e}_o$ and $\mathbf{e}_f$ are the polarization vectors of the incoming and the scattered beams, $\mathbf{z}_j$ is the direction of the quantization axis, and $F_{L,M}$ is the resonance response, where L is the order of the transition ($L=1$ for dipole transitions) and M is the change in angular momentum ($\Delta M=0,\pm1$). The first term is the usual resonant scattering amplitude and the second term is the linear magnetic term due to exchange scattering. To a first approximation, the $3d$ states are treated as atomic levels with an exchange splitting,[4] and the scattering amplitude becomes

$$f_{E1}^{(\mathrm{res})}=F[(\mathbf{e}_f\cdot\mathbf{e}_o)n_h+i(\mathbf{e}_f\times\mathbf{e}_o)\cdot\mathbf{z}_jP/4]\,, \quad (2)$$

with

$$P=\left[n_e-\left(\frac{\Delta}{\Gamma[x(\alpha,\eta)-i]}\right)n_h\right], \quad (3)$$

where F includes the radial part of the transition probability and the resonant denominator, $1/[x(\alpha,\eta)-i]$, n_e is the net number of spin up electrons, n_h is the total number of holes in the $3d$ band, Γ is the total width of the excited state, and Δ is the exchange splitting. In the resonant denominator x is the deviation from the absorption energy in units of half-widths, and is defined as

$$x(\alpha,\eta)=\frac{\varepsilon-(E_\eta-E_\alpha)}{\Gamma/2}\,, \quad (4)$$

where ε is the photon energy, and E_α,E_η are the energies of the ground state and the excited state, respectively. Note that the sign of the resonant denominator is opposite to the one adopted in Ref. 4. From Eq. (3), clearly the linear magnetic term is a function of both spin polarization of the partially filled states and the exchange splitting of the unoccupied states. Specular reflectivity can then be calculated using the scattering amplitude in Eq. (2).

In our case, specular reflectivity including only the usual resonant term is first calculated and compared with the experimental results to determine the values of the absorption-edge energies, absorption coefficients at the edges, core-hole lifetime, and total width of the excited state.[9,10] Interference effects due to the vacuum-Fe and Fe-substrate interfaces are also included.[11] Very good agreement between the calculated and measured reflectivity was obtained with values typical of $L_{II,III}$ edges of $3d$ transition metals. Next, the linear magnetic term is included to calculate the reflectivity in the presence of the magnetic field, and the asymmetry ratios. Note that Eq. (3) is derived in the limit of $\Delta/\Gamma\ll1$, which is not satisfied in the case of Fe because of the

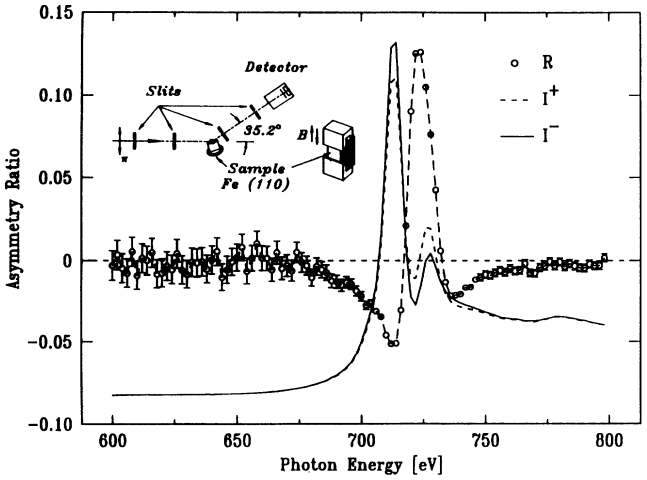

FIG. 1. Specular reflectivity of p-polarized light, I^+ and I^-, and asymmetry ratios R measured from a 35-Å Fe(110) crystal. The line through the asymmetry ratios is only to serve as a guide to the eye. Inset: Scattering geometry. The polarization of the radiation is in the scattering plane, the applied magnetic field is perpendicular to the scattering plane, and the scattering angle 2θ is 35.2°.

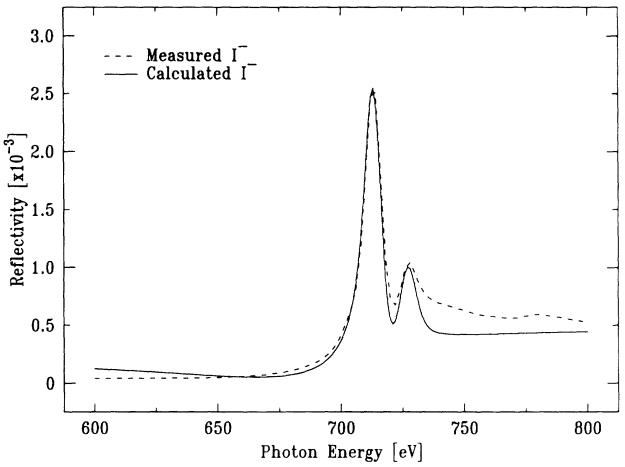

FIG. 2. Comparison between the measured and the calculated reflectivity. The following parameters are used in the calculation: $n_e=1$, $n_h=4$, $\Delta=1.5$ eV, $\Gamma=5.0$ eV.

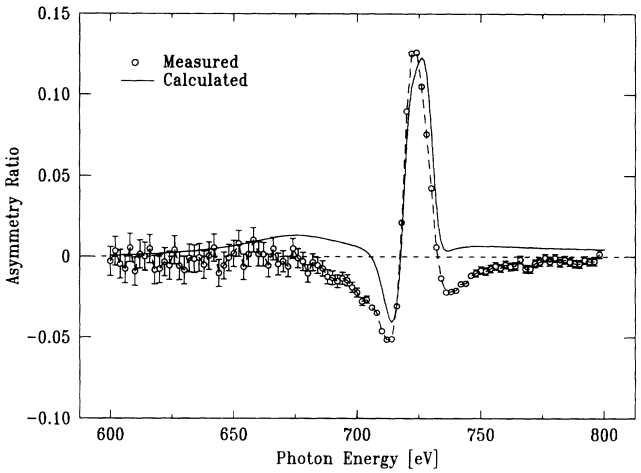

FIG. 3. Comparison between the measured and the calculated asymmetry ratios. The following parameters are used in the calculation: $n_e=1$, $n_h=4$, $\Delta=1.5$ eV, $\Gamma=5.0$ eV.

large exchange splitting and narrow $3d$ band. Therefore we have extended the calculation to include second-order terms in Δ/Γ, although no significant correction was introduced by these terms.

Figure 2 shows both the measured I^- reflectivity and the calculated I^- reflectivity using the following parameters: $n_e=1$, $n_h=4$, $\Delta=1.5$ eV, $\Gamma=5.0$ eV. Figure 3 compares the asymmetry ratios calculated using the same parameters, and the measured asymmetry ratios. From Figs. 2 and 3, it is evident that good quantitative agreement between the calculated reflectivity and asymmetry curves, and the experimental results is achieved. Better agreement might be obtained by including the effect of band structure in more detailed calculations, which are in progress. However, even within the simple atomiclike model, all the major features in the measured asymmetry curve are reproduced by the calculation. Moreover, these features are sensitive to the parameters in the linear magnetic term, n_e , n_h, Δ, and Γ. This sensitivity can be exploited to determine the values of these important parameters. For example, to obtain a good fit to the data, an exchange splitting of 1.5 eV is used in Fig. 2, which is within the range of calculated exchange splitting for Fe, from 1.36 eV (near the bottom of the d band) to 2.24 eV (near the top of the d band);[12] whereas the largest asymmetry ratio would be reduced to less than 10% if an exchange splitting of 1.0 eV is used instead. It should also be noted that the observed asymmetry ratio is roughly proportional to $\tan(2\theta)$. Thus, as mentioned above, much larger effects are expected at higher scattering angles, although the absolute reflectivity would be smaller.

In summary, we have observed large asymmetry ratios in the specular reflectivity in the vicinity of Fe L_{II} and L_{III} edges due to resonant exchange scattering. Similar resonant enhanced, magnetization sensitive effects should also be observable in other ferromagnetic and ferrimagnetic systems. The results reported here demonstrate that important information about the electronic structure of these ferromagnetic systems can be extracted from these measurements. In comparison with other experimental techniques which are used to study the unoccupied states near the Fermi level of magnetic materials, such as spin-polarized inverse photoemission,[13] magneto-optical Kerr effect measurements,[14] and photoabsorption using circularly polarized light,[15] resonant exchange scattering has the following unique features. It is a coherent elastic-scattering process which has the advantage of having no complicated final-state effects. The localized nature of core excitation is particularly useful in the study of impurity effects and of multicomponent systems. Also, because of the relatively long penetration depth of soft-x-ray photons, the condition of the sample surface is not necessarily important, although surface sensitivity can be enhanced by reducing the scattering angle.

Finally, the large resonant enhancement combined with the high flux of synchrotron radiation makes this technique especially attractive for the study of surface, interface, and thin-film magnetism, and for the characterization of dilute magnetic samples. Thus, we believe specular reflectivity will be an important addition to existing experimental techniques for the study of magnetism and magnetic materials characterization.

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