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X-ray resonance exchange scattering in dysprosium

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Resonant enhancement of the scattering intensity of the magnetic satellite resulting from the basal-plane spiral antiferromagnetic structure is observed in dysprosium metal at the $L_{\rm III}$ absorption edge. The polarization dependence and enhancement agree with theory and are similar to those observed previously in holmium metal.

Resonances in the intensity of magnetic x-ray scattering when the incident x rays are tuned through absorption edges has been shown in several recent experiments. These resonances can be used to study the magnetic properties of materials and may provide information about Fermi edge structure. The first such effects were observed in ferromagnetic nickel at the K absorption edge.[1] This was followed by the observation of a 50-fold enhancement of the first-order harmonic and the appearance of higher harmonics in the basal-plane, spiral antiferromagnetic structure of holmium. These measurements revealed two resonance peaks about the $L_{\rm III}$ absorption edge which were separated in energy by about 6 eV.[2] A model which explained the two peaks and the polarization dependence of all the observed higher harmonics was developed. This model for "x-ray resonance exchange scattering" attributed the enhancement to electric dipole and quadrupole atomiclike transitions which are sensitive to the magnetization via an exchange splitting of the intermediate states and the exclusion principle.[3] Larger resonances were predicted at the $M_{\rm IV}$ and $M_{\rm V}$ absorption edges of rare-earth and actinide elements and compounds. This prediction was confirmed in studies of the type-I and type-IA antiferromagnetic structures of UAs, where the amplitude of the antiferromagnetic reflections were a remarkable $9r_0$, where r_0 is the classical electron radius.[4]

In this Rapid Communication, we describe the study of the resonance enhancement of the magnetic scattering about the $L_{\rm III}$ absorption edge at the first-harmonic $(0,0,2+\tau)$ with a total intensity that was 3×10^{-5} weaker than the charge peak at the (0,0,2). The polarization dependence of the resonance peaks agree with the theory. The results described here are qualitatively similar to those found for holmium (see Ref. 2).

The scattering amplitude for dipole transitions that gives rise to magnetic scattering is given by

$$f_{\rm dip}^{\rm xres}(\omega,\varepsilon_f,\varepsilon_i)=\frac{3}{8\pi}\lambda((\varepsilon_f^*\cdot\varepsilon_i)[F_{11}(\omega)+F_{1-1}(\omega)]-i(\varepsilon_f^*\times\varepsilon_i)\cdot\mathbf{z}_J[F_{11}(\omega)-F_{1-1}(\omega)]$$
$$+(\varepsilon_f^*\cdot\mathbf{z}_J)(\varepsilon_i\cdot\mathbf{z}_J)[2F_{10}(\omega)-F_{11}(\omega)-F_{1-1}(\omega)])\,,$$

where ε_i and ε_f are the polarization vectors of the incident and scattered beams, respectively, and $\mathbf{z}_J$ is the local magnetization direction (i.e., axis quantization for the atomic moment). The $F_{1m}(\omega)$'s, which are proportional to the atomic polarizability, include the resonant denominator and are a function of the incident x-ray energy and not the polarization. The first term is independent of the magnetization direction and gives rise to scattering at the zeroth harmonic. The second term gives rise to first harmonic satellites for an antiferromagnet. In a spiral antiferromagnet, second-harmonic (as well as zeroth harmonic) satellites arise from the third term, which depends quadratically on the magnetization direction. The polarization dependence of these terms, which arises from the atomic selection rules, is clearly observed in this experiment and is demonstrated below.

The experiments were carried out with the AT&T Bell Laboratories beamline X16B at the National Synchrotron Light Source. A singly bent, asymmetrically cut Ge(111) monochromator focuses 2 mrad of radiation in the horizontal. The x-ray energy is continuously tuned over a 200-eV range by simultaneously rotating the monochromator crystal, translating the diffractometer normal to the incident beam, and finally rotating the diffractometer to keep the θ and $2-\theta$ axes normal to the incident beam. This horizontal plane monochromator geometry provides high-quality energy scans as is evidenced by the absorption profile for a dysprosium foil shown in the top of Fig. 1. The width of the white-line spectrum seen at the $L_{\rm III}$ absorption edge is similar to that observed with a double-crystal, vertical-plane, Si(111) monochromator in holmium metal. (The holmium measurements were made on beamline A2 at the Cornell High Energy Synchrotron Source.[2])

The polarization of an x-ray beam from a bending magnet source is linearly polarized in the plane of the electron

orbit and becomes increasingly more right- or left-handed elliptically polarized above or below the plane. Horizontal slits upstream of the monochromator can be varied in width and translated vertically to tune the degree of linear polarization of the incident beam. On center, the degree of linear polarization is determined by the angular acceptance of the slits and fluctuations of the source. Previous measurements at beamline X16B have shown that the degree of linear polarization varies from 95% at the center of the beam to 50% 5 mm above with 0.5 mm slits. The degree of linear polarization is defined as $P=(I_\sigma - I_\pi)/(I_\sigma + I_\pi)$, where I_σ and I_π are the intensity of the component of linear polarization parallel and perpendicular to the plane of the electron orbit, respectively. In the present experiment, P was measured to be 90%.

The polarization of the scattered beam was measured by choosing an analyzer crystal with a reflection for which the Bragg angle was 45° near the L_{III} edge at $E=7.796$ keV. With this condition, only a single polarization component of the beam incident on the analyzer was passed into the detector, at 90° from the beam scattered from the sample. The other component was measured by rotating the analyzer by 90° about the axis of the scattered beam. For the holmium experiments, the (0,0,6) reflection of graphite was employed at the L_{III} edge, but the reflectivity was less than 5%. For the L_{III} edge of dysprosium the (1,1,0) of beryllium, with $\theta=45°$ at $E=7.67$ keV has a much larger reflectivity of 30%.

The dysprosium sample was a [0,0,1], 0.2-mm-thick single-crystal plate, grown by Beaudry at the Ames Laboratory. Dysprosium, like holmium, has a large free-ion magnetic moment of $10.6\mu_B$ arising from a Hund's rule $^6H_{15/2}$ ground state. The moments form a basal-plane spiral antiferromagnetic structure for temperatures between $83 \leq T \leq 176$ K. The wave vector describing the spiral varies continuously from $\tau=0.24\mathbf{c}^*$ at $T=176$ K to $\tau=0.15\mathbf{c}^*$ at $T=83$ K. At $T=83$ K a transition into a ferromagnetic phase is observed. In the spiral phase there are symmetric magnetic satellite peaks about each reciprocal lattice vector $\mathbf{G}$ at $\mathbf{G}\pm\tau$. The experiments described here were carried out at $T=105$ K, where $\tau=0.163\mathbf{c}^*$, with the sample mounted on the cold finger of a closed-cycle ^{4}He refrigerator and centered on a standard, four-circle Huber diffractometer.

Figure 1 shows the integrated intensity of the (0,0,2 $+\tau$) magnetic satellite for both polarization components in the scattered beam, about the L_{III} absorption edge of Dy. At the peak of the resonance, $E=7.798$ keV, the integrated intensity of the sum of both polarization components at the $(0,0,2+\tau)$ is 3×10^{-5} times weaker than the intensity at the (0,0,2) charge peak. The intensity of the magnetic satellite drops by a factor of 90 when the incident x-ray energy is tuned below the absorption edge to $E=7.668$ keV. This resonance enhancement is similar in magnitude to the factor of 50 found at the L_{III} edge in

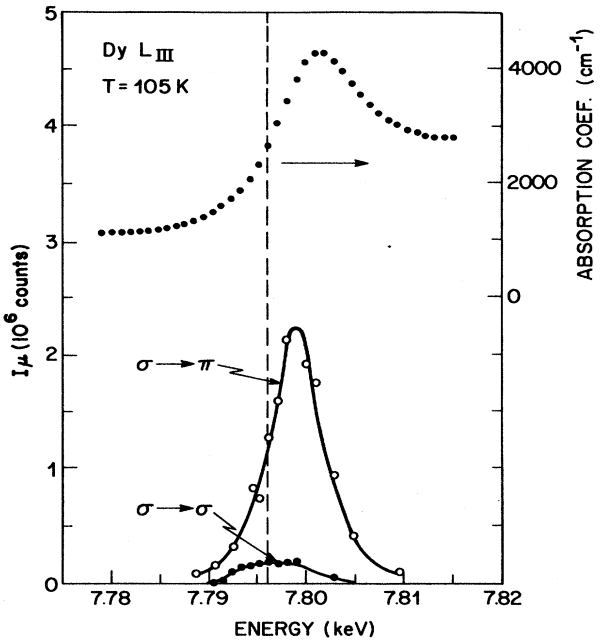

FIG. 1. Upper curve shows the absorption profile for a dysprosium foil taken with a singly bent, asymmetrically cut Ge(111) monochromator. Lower curves show the integrated intensity of the $(0,0,2+\tau)$ magnetic satellite for both scattered polarizations; σ to π (open circles) and σ to σ (filled circles).

holmium. As in holmium, the intensity of the σ to π scattering is the strongest and is peaked just below the peak in the white line spectrum. The σ to σ scattering peaks at a lower energy and is weaker than observed in Ho. In Ho the second harmonic is principally σ to σ at the energy where the σ to π scattering of the first harmonic is peaked (see Fig. 2 of Ref. 2). This behavior is similar in dysprosium, where the ratio of intensities

$$I_{\sigma\rightarrow\pi}(0,0,2+\tau)/I_{\sigma\rightarrow\sigma}(0,0,2+2\tau)\sim 8\,.$$

All of these results are consistent with the dipole and quadrupole selection rules described by Hannon *et al.* in Ref. 3.

The general features of the resonant magnetic scattering in Dy are similar to those in Ho. This suggests that resonances should be observed in other rare-earth metals. It will be of particular interest to study the resonant behavior in mixed valence materials such as TmSe. TmSe, which orders antiferromagnetically, shows a clear splitting in the white line spectrum which is attributed to the Tm^{3+} and Tm^{2+} configurations, respectively.

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