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paper 101 H
32/0030
BNL-46281 62-910903-4

Resonant X-Ray Magnetic Scattering in Holmium

BNL--46281

DE91 015190

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Abstract

We review the results of resonant x-ray magnetic scattering experiments on the rare earth metal holmium. When the incident x-ray energy is tuned near the L_{III} absorption edge, large resonant enhancements of the magnetic scattering and resonant integer harmonics are observed. These results are analyzed within the theory of x-ray resonance exchange scattering assuming electric dipole ($2p \rightarrow 5d$) and quadrupole ($2p \rightarrow 4f$) transitions among atomic orbitals.

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MASTER

INTRODUCTION

In this paper, we review the results of recent x-ray magnetic scattering experiments on the rare earth metal holmium.(1-4) Specifically, we show that when the incident x-ray energy is tuned near the L_{III} absorption edge, large resonant enhancements of the magnetic scattering, and resonant integer harmonics, are observed. Besides intrinsic interest in the x-ray scattering cross-section and its description, studies of this type are also motivated by the possibility that new structural and magnetic properties of rare earth metals might be revealed, and that new techniques may thereby be developed. As a result, interest in x-ray magnetic scattering experiments has grown rapidly during the last 10 years particularly, with the regular application of synchrotron radiation techniques. Magnetic scattering experiments have by now been performed at most of the x-ray synchrotrons around the world. In addition, insertion device beamlines dedicated to x-ray magnetic scattering experiments are planned at each of the next-generation storage rings presently under construction in the United States (APS), Europe (ESRF), and Japan (SPRING-8).

Generally, the results of x-ray magnetic scattering experiments may be analyzed within two regimes of the incident x-ray energy, namely, in the limit of high x-ray energies, when the incident x-ray energy lies above the excitation energy of any absorption edge, and at resonance, when the incident energy lies near an absorption edge. In the high energy limit, the amplitude for x-ray magnetic scattering has the simple form $[\mathbf{L}(\mathbf{Q}) \cdot \mathbf{A} + \mathbf{S}(\mathbf{Q}) \cdot \mathbf{B}]$.⁽⁵⁻⁷⁾ Here, $\mathbf{L}(\mathbf{Q})$ and $\mathbf{S}(\mathbf{Q})$ refer to the Fourier transforms of the atomic

orbital and spin magnetization densities, respectively, and the vectors **A** and **B** depend on the incident and scattered wavevectors and on the incident and scattered polarization vectors⁽⁵⁻⁷⁾. Because the vectors **A** and **B** are not identical, the polarization dependence of the orbital contribution to the magnetic cross-section differs from that of the spin contribution. This leads to the possibility that the orbital and spin magnetization densities may be distinguished in x-ray scattering experiments. The same distinction is not directly possible by neutron diffraction techniques (in dipole approximation, where the polarization dependence of the orbital contribution is identical to that of the spin contribution), and is important to a fundamental understanding of the electronic structure of magnetic materials. A particular class of experiments for which these techniques may be useful will concern quantitative studies of mixed-valence and heavy-fermion materials, for which the 4f and 5f orbital and spin magnetization densities are presently uncertain.

In the resonant regime (of interest in this paper), when the incident x-ray energy is tuned near an absorption edge, there are additional contributions to the x-ray scattering cross-section¹⁻⁴. Within a one-electron view of the electronic structure, the incident photon promotes an inner shell electron to an unoccupied orbital above the Fermi energy, which subsequently decays through the emission of an elastically scattered photon. The amplitude for resonant magnetic scattering then depends on the matrix elements which couple the ground state and the excited magnetic states allowed by the exclusion principle. In this way, the energy and polarization dependence of the magnetic scattering probes the fine

structure of magnetic states. It follows that detailed modeling of the lineshapes of the magnetic scattering may reveal the spectrum of allowed transitions near E_f , including, perhaps, the magnitude of the exchange splitting and the induced polarization within metallic conduction bands^{3,4}. It is also important to note that the polarization dependence of the resonant cross-section depends in a simple way on the directions of the local atomic moments. The existence of large resonant enhancements of the magnetic scattering then makes feasible the determination of magnetic structures by x-ray scattering techniques in a wider class of materials than was previously imagined. We believe that resonant techniques will be of special interest in studies of the magnetic properties of rare earths and actinides, particularly in application to multilayers, thin films, and surface layers. Experiments of this type are straightforward using synchrotron radiation when the incident x-ray energy lies near the L absorption edges of the rare earths ($\hbar\omega \sim 6-10\text{keV}$) and the M absorption edges of the actinides ($\hbar\omega \sim 3-5\text{keV}$). X-Ray reflectivity studies of these materials, and of transition elements, over an even wider range of energies offer still another direction for resonant studies of magnetic properties.

A detailed discussion of the experiments reviewed in this paper is given in reference 1. Related resonant and non-resonant x-ray magnetic scattering studies have been performed on most of the heavy rare earth metals, including Ho^{1-4,8-10}, Dy¹¹, Tm¹², and Er¹³, on two rare earth magnetic multilayers, including Gd-Y¹⁴ and Ho-Y¹⁵, and in a variety of actinides and transition metals, including UAs^{16,17}, UN,¹⁸ U Ru₂Si₂,¹⁹ UO₂,²⁰ Ni²¹, MnF₂,²² and Fe²³. The largest

resonant enhancements have been observed for incident x-ray energies near the M_{IV} absorption edges of the actinides¹⁶⁻²⁰ and near the L_{III} absorption edges of the rare earths^{1-4,11-15} and transition metals. Spin-dependent absorption measurements have been performed on a wide variety of transition metal and rare earth powdered ferromagnets (see, for example, Reference²⁴). Recently, calculations have been made of the resonant cross-sections for scattering and for absorption in ferromagnets²⁵.

The first x-ray magnetic scattering experiments were performed by deBergevin and Brunel in NiO and various Fe compounds^{26,24} following the original calculations of Platzman and Tsoar²⁸, in 1972. The first polarization sensitive x-ray magnetic scattering experiments were performed by Brunel and coworkers on Fe in 1982.²⁹ Blume suggested that interesting magnetic effects might occur near an absorption edge in 1985. ⁵Namikawa and coworkers performed early measurements of resonant magnetic effects in Ni²¹.

Metallic holmium has an hcp crystal structure with two layers per chemical unit cell and a saturation magnetic moment of $10.3\mu_B/\text{atom}$ ³⁰. In the ground state there are ten 4f electrons in a Hund's Rule 5I_8 configuration and three electrons in the 5d and 6s bands. In the metal the 5d and 6(s-p) conduction bands have energy widths between 5 and 10eV, which is typical of rare earths. The 4f bands have energy widths of about 2eV and lie less than 5eV above or below the Fermi energy. In holmium, the L_{II} absorption edge occurs at $\hbar\omega = 8067\text{eV}$, and is well separated from its L_{III} and L_I

partners at $\hbar\omega=8916$ and $\hbar\omega=9395\text{eV}$, respectively. The M_{IV} and M_V absorption edges occur at $\hbar\omega=1392$ and $\hbar\omega=1351\text{eV}$, respectively.

Immediately below the magnetic ordering temperature ($T_N=138\text{K}$), the non-resonant x-ray (neutron) magnetic diffraction pattern for holmium consists of single pairs of magnetic satellites split symmetrically around each of the main chemical (nuclear) Bragg reflections, and parallel to the (00L)-, or c-axis. This pattern is consistent with a simple spiral modulation in which the average moments are ferromagnetically aligned within the basal planes, but rotate from plane to plane with an average turn angle proportional to the magnetic wavevector $\tau^{8-10,30}$. As the temperature is decreased from $T=138\text{K}$, the magnetic wavevector decreases from about $\tau\sim 0.3c^*$ to $\tau=1/6c^*$ below $T=20\text{K}$, and may lock to rational values^{8-10,30}. At $T_C=20\text{K}$, there is a first order transition to a conical magnetic structure with a net ferromagnetic moment along the c-axis.

A schematic view of the geometry used in these experiments is shown in Figure 1. In the figure, the x-ray beam is incident onto the sample from the left, and makes an angle Θ with respect to its surface and to the (00L) Bragg planes. The coordinate system is chosen so that the σ -polarized components of the incident and scattered beams are normal to the diffraction plane (spanned by the incident and scattered wavevectors $\mathbf{k}$ and $\mathbf{k}'$), while the π -polarized components lie within the diffraction plane. The experiments were performed at the National Synchrotron Light Source (NSLS).

Representative rocking curves of the magnetic satellite of the (002) reflection (002)+ obtained as the incident photon energy is tuned through the L_{III} absorption edge, are shown in Figure 2. In these scans the sample temperature was fixed at $T=32K$, at which temperature holmium exhibits a simple spiral magnetic phase with $\tau=0.189c^*$. The results for $\hbar\omega=800eV$ are shown in the lower right of the figure, where a small magnetic peak is visible on top of a flat background. Typical count rates in the magnetic peak at this energy were about 1000/sec (on backgrounds of about 1/sec) at an NSLS electron storage ring current of 100ma. As the incident photon energy is increased, the scattering angle (2θ) decreases and the peak intensity increases, until about $\hbar\omega=8070eV$, where the magnetic intensity reaches a maximum. At this energy the measured count rate in the magnetic peak is about 45,000/sec (at a ring current of 100ma) on a background of about 2000/sec. Thus, the measured peak intensity increases by nearly a factor fifty by tuning the incident x-ray energy from $\hbar\omega=8000eV$ to an energy near the L_{III} absorption edge. Relative to the intensity of the charge scattering obtained at the (0,0,2) reflection, the resonant magnetic scattering at the (0,0,2)+ reflection is reduced by a factor of order 10^4 . The increased background above the edge originates in the holmium L_{III} fluorescence, and may be reduced by the use an energy dispersive detector. Above $\hbar\omega=8070eV$, the magnetic intensity again decreases, but remains on top of the large fluorescent background.

Integrated intensities measured for the (0,0,2)+ magnetic satellite are shown plotted on a linear scale from $\hbar\omega=7700eV$ to $\hbar\omega=8100eV$ in the lower part of Figure 3. Although the intensity

obtained near $\hbar\omega=7700\text{eV}$ appears small when plotted on this scale, the measured signal at that energy was several hundred count/sec. The data in the figure have been corrected at each energy for the holmium L_{III} absorption, which is shown plotted in the top panel of Figure 3. For simplicity, we have assumed the infinite flat plate geometry, in which the integrated intensity is multiplied by the absorption coefficient μ at each incident photon energy. After correction for the absorption, the integrated intensity measured at the L_{III} absorption edge for the (0,0,2)+ reflection is greater by a factor of about one hundred fifty than the integrated intensity measured at $\hbar\omega=7850\text{eV}$.

In addition to the resonant enhancement observed for the magnetic scattering at (0,0, $L=2+\tau$), we have also observed resonant harmonics of the magnetic scattering at $L=2+2\tau, 2+3\tau$, and $2+4\tau$. The energy dependence of the integrated intensity, corrected for the absorption, of the resonant magnetic scattering at τ and its harmonics is shown plotted on a linear scale in Figure 4. At the top of the figure are the results at τ . The maximum integrated intensity occurs a few electron volts above the inflection point of the absorption at an incident photon energy $\hbar\omega=8070\text{eV}$ and the energy full-width-half-maximum (FWHM) is about 10eV. The second panel of Figure 4 shows the energy dependence of the scattering at 2τ . In this case, no scattering is observed below $\hbar\omega=8040\text{eV}$, nor above $\hbar\omega=8082\text{eV}$. Similar to the magnetic scattering at τ , the maximum intensity again occurs near 8070eV, but is reduced by a factor of about twenty five. In contrast to the scattering at τ , the lineshape for the scattering at 2τ exhibits a shoulder near $\hbar\omega=8062\text{eV}$, which

leads to a broadening of the FWHM to about 14eV. The energy dependence of the integrated intensity of the scattering at $L=2+3\tau$ is shown in the third panel of Figure 4. To within the experimental statistics, no scattering is observed below $\hbar\omega=8054\text{eV}$, nor above $\hbar\omega=8080\text{eV}$. The profile of the scattering at 3τ takes a maximum at $\hbar\omega=8063\text{eV}$, which is about 4eV below the inflection point of the absorption. The energy FWHM for the scattering at 3τ is 6eV, which is the instrumental resolution at this energy. The profile for the scattering at $L=2+4\tau$, shown in the fourth panel, also takes a maximum at $\hbar\omega=8063\text{eV}$ and has a similar energy width.

In our view, all of the data presented in Figures 3-4 may be most simply understood on the basis of electric multipole contributions to the x-ray scattering cross-section through f' and f'' . Specifically, in application to holmium, it may be shown¹⁻⁴ that dipole-allowed transitions, coupling 2p core electrons with 5d-derived conduction band states, lead to only two harmonics: at τ and 2τ . Similarly, the quadrupole allowed transitions, coupling 2p core electrons to 4f-derived states, lead to four harmonics: at τ , 2τ , 3τ , and 4τ .

Referring to Figure 4, the scattering located at $\hbar\omega=8063\text{eV}$ for the first 2 harmonics at τ and 2τ and all the scattering at 3τ and 4τ may be associated with quadrupole excitations. The extra energy width of the scattering at τ and 2τ , i.e. the additional scattering observed at $\hbar\omega=8067\text{eV}$, is then associated with dipole excitations. This designation gains particular credibility in polarization sensitive experiments where the dipole and quadrupole contributions

have a distinct polarization dependence and may be uniquely identified¹⁻⁴.

To investigate the dependence of the lineshapes on electric multipole transitions, on the induced polarization and exchange splitting within the d-band, and on the non-resonant magnetic scattering, systematic studies of the integrated magnetic intensity of the (0,0,2)+, (0,0,4)+, and (0,0,6)+ magnetic reflections were undertaken between $\hbar\omega=7900\text{eV}$ and 8150eV . All of the intensities reported here are integrated intensities, and have been corrected for the absorption and by the Lorentz factor. Except for a single scale factor (amp1, below), these intensities may be regarded as absolute. In the analysis, however, no account has been taken of the contributions to the scattering from non-linearly polarized components of the incident or scattered beams. Although we believe these additional contributions are probably small ($P=0.9\pm.1$ at NSLS), they certainly exist, and they may distort the lineshapes. For this reason, our interpretation of the lineshapes is limited mainly to their qualitative features. Polarization sensitive experiments were performed at the same time as these and are described in reference 1.

The total integrated intensity obtained at the magnetic wavevector τ for the (0,0,2)+, (0,0,4)+, and (0,0,6)+ reflections is plotted on a logarithmic scale versus incident x-ray energy in Figure 5. For clarity of presentation, the results for the (0,0,2)+ and (0,0,4)+ reflections have been scaled by factors of 200 and 10, respectively. The solid lines are the results of fits, which are discussed below. The main features of the lineshapes in Figure 5 are

clear. In each case the profile of the magnetic scattering exhibits a long, asymmetric tail which extends toward lower energy, and a large enhancement of the scattering in the neighborhood of the L_{III} edge. The asymmetry is weakest, and the enhancement largest, for the $(0,0,2)^+$ satellite. As the scattering angle increases, the relative asymmetry also increases. It is interesting that the resonance width for the $(0,0,6)^+$ reflection is broader near the L_{III} edge than are the corresponding widths for the $(0,0,4)^+$ or $(0,0,2)^+$ reflections. This behaviour suggests the possibility of additional fine structure. Above the L_{III} absorption edge, the scattering is again reduced, and the fluorescent background is large.

We have modelled the energy dependence of the magnetic scattering for the $(0,0,2)^+$, $(0,0,4)^+$ and $(0,0,6)^+$ reflections using non-resonant contributions taken from reference 6 and resonant contributions taken from reference 4. In the model, we assume that the resonant contributions arise mainly from electric dipole transitions coupling 2p core states and 5d conduction band states and from electric quadrupole transitions coupling 2p core states and 4f states. For simplicity, we also assume that the degree of linear polarization of the incident beam $P=1$. Then, the amplitude for the magnetic scattering is:

$$|f\sigma - \pi|^2 = amp1 \left| J_{eff} \frac{\hbar\omega}{mc^2} \sin(2\theta)\sin(\theta) + \frac{\cos(\theta)}{\Gamma(x-i)} \left[A - \frac{B}{\Gamma(x-i)} \right] + \frac{\cos(\theta)\cos(2\theta)C}{\Gamma(x'-i)} \right|^2, \quad (1)$$

where

$$J_{eff} = \begin{cases} 5.5, (0,0,2)^+ \\ [3/4 f_L + 1/5 f_s] J = 4.0, (0,0,4)^+, \\ 2.3(0,0,6)^+ \end{cases}$$

$$A = \frac{Fn_c}{4}, \quad B = \frac{(\Delta)n_h F}{4}, \quad \text{and} \quad C = F'$$

The first term in this expression is the non-resonant contribution to the magnetic scattering from a magnetic spiral⁶. J_{eff} is the total effective magnetic moment, and depends on the form factors for the orbital- and spin-magnetization densities f_L and f_S . The values for J_{eff} are calculated from published values. The terms containing A and B are the resonant contributions from electric dipole transitions coupling 2p and spin-down and spin-up 5d orbitals. n_c and n_h are, respectively, the net induced polarization and the number of holes in the 5-d band of holmium, and Δ is the energy of the exchange splitting between spin down and spin up states in the d-band. $\bar{r}$ depends on the overlap of radial 2p and 5d electronic wavefunctions of holmium. The principal effect of the exchange splitting is to introduce a second-order pole with amplitude proportional to B. Γ is the width of the excited resonant state, and $x=2(E_a-E_b-\hbar\omega)/\Gamma$ is the resonant denominator. The term containing C is the resonant contribution from electric quadrupole transitions coupling 2p and 4f states. The 6eV shift which appears in the resonant denominator of the quadrupole term $x'=2(E_a-E_b-\hbar\omega-6)/\Gamma$ accounts for the 6eV splitting which separates the dipole and quadrupole excitations (see reference 1). The trigonometric functions give the polarization dependence of the resonant and non-resonant cross-sections for a magnetic spiral, and depend on the scattering angle. amp1 is a global scale factor, independent of scattering angle 2θ , which accounts for the structure factor, the Debye-Waller factor, the incident intensity, etc.

Self-consistent fits of the data to the model are shown by the solid lines in Figure 5. Because these parameters are highly correlated, it has not been possible to obtain "best fits" of the results, which would result from arbitrary variations of the parameters. Instead, these values have emerged after repeated trials starting from a variety of initial values. Small variations of the parameters may also give reasonable fits. Although the results are approximate, certain features of the resulting lineshapes are clear, and suggest that the basic elements of the description are correct. In particular, it is seen that the asymmetric tail and the large enhancement near the L_{III} absorption edge for the total cross-section in Figure 5 is closely reproduced. On the basis of the model, it is unambiguous that this asymmetry arises from the interference of the resonant and non-resonant cross-sections in Equation 1. It may also be deduced that the decreased asymmetry in the lineshape for the $(0,0,2)+$ reflection results from the decrease of the non-resonant magnetic cross-section scattering at small momentum transfers.

A second result of the fitting concerns the apparent broadening or fine structure of the lineshapes of the $(0,0,6)+$ reflections, relative to the lineshapes of the $(0,0,4)+$ and $(0,0,2)+$ reflections, in Figure 5. Although the agreement is not precise, it seems clear from the model that the weak shoulder which becomes visible at higher momentum transfers arises from quadrupole contributions to the magnetic scattering (that is, from the term containing C in Equation 1). This manifestation of the quadrupole contributions at higher momentum transfers is a consequence of the polarization

dependence of the resonant cross-section. It is noteworthy that these same ideas may also be used to explain the results of polarization sensitive magnetic scattering experiments (see Ref. 1).

The values of the parameters used to generate the fits shown in Figure 5 are $\text{amp1}=0.03$, $A=2$, $B=4$, $-C=C'=3/4$, and $\Gamma=9\text{eV}$. The fitted level width $\Gamma=9\text{eV}$ is close to the atomic value calculated for holmium of about 7eV . It is interesting that the ratio of the fitted constants $A/B=1/2$ is of the same order of magnitude as may be estimated from reasonable values of the parameters for holmium: $A/B=\pi_e/\pi_h\Delta=1/8$ with $\pi_h\sim 8$, $\pi_e\sim 0.2$, and $\Delta=0.2\text{eV}$. Similarly, the fitted values of the constants A and B each fall within a factor of two of the values obtained using one-electron orbitals in a calculation of the electronic structure of $(\text{Ho})^{++}$. Using the same estimates for π_h , π_e , and Δ as above, these calculations³⁻⁴ yield $A\sim 1$ and $B\sim 8$. Thus, the fitted parameters A and B are qualitatively consistent with one-electron predictions for the electronic structure. The discrepancies between the fitted parameters and their calculated values suggest that more sophisticated calculations of the electronic structure may be required before a quantitative comparison is possible. In the calculation, for example, the influence of the core hole on the wavefunctions of the excited state has been ignored. It should be clearly noted, in addition, that the effect of the exchange splitting on the model may be entirely removed by setting $B=0$ in Equation 7 above. Fits of the model with $B=0$ have also been successfully performed, leading to results of nearly the same quality as those shown in Figure 5. For this range of the parameters, the main effect of the exchange splitting on the lineshapes is to introduce small

corrections to the maximum intensity. Therefore, while the present results are consistent with an orbital model of the electronic structure which includes exchange splitting of the d-band, these fits are not unique. Finally, we note the values of the constant C and C' are of the same order of magnitude as A and B, but that C has opposite sign to the results expected from one-electron calculations. It seems possible that this difference may reflect the influence of the crystal field, which has also been ignored in the calculation³⁻⁴.

In this paper, we have summarized some resonance properties of the x-ray magnetic cross-section of the spiral antiferromagnet holmium for incident x-ray energies near the L_{III} absorption edge. The results are qualitatively consistent with the theory of x-ray resonance exchange scattering, including electric dipole and quadrupole transitions among atomic orbitals. The most striking feature of the lineshapes is their asymmetry, which arises from the interference of the resonant and non-resonant contributions. It remains to explain the quantitative deviations of the fitted parameters for the L_{III} resonance behavior from simple one-electron estimates and to explain the weakness of the enhancement observed near the L_{II} absorption edge relative to that observed near the L_{III} edge. In our view, x-ray magnetic scattering techniques will find continued, fruitful application in studies of rare earths, actinides, transition elements, and their compounds-particularly in the extensions of these studies to thin films, multilayers, and surface layers. It also seems clear that these techniques will benefit directly from the development of polarization- and energy-tunable

insertion-device beamlines at the high brightness storage rings under construction in the United States, Europe, and Japan.

Acknowledgements

The work reviewed in this paper was performed in collaboration with G. Grübel, D.R. Harshman, E.D. Isaacs, D.B. McWhan, D. Mills, and C. Vettier. Their contributions are gratefully acknowledged. We have also benefitted from discussions with M. Blume, J.P. Hannon, G.H. Lander, W.G. Stirling, and G.T. Trammell.

Support of the IQF Magnetism Group and of Elsevier North-Holland during the 1991 International Conference on Magnetism held on 2-6 Sept. in Edinburgh, Scotland is also gratefully acknowledged.

Work performed at Brookhaven National Laboratory is supported by the U.S. D.O.E. under contract No. DE-AC02-76CH00016.

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Figure Captions

1. Schematic view of geometry used in these experiments.
2. Rocking curves of the x-ray magnetic scattering for holmium at the $(0,0,2)^+$ reflection plotted as the incident x-ray energy is tuned through the L_{III} absorption edge at $\hbar\omega=8067\text{eV}$.
3. Top: The absorption measured from a 5 micron holmium film as the incident x-ray energy is tuned through the L_{III} edge. Bottom: The integrated magnetic intensity of the $(0,0,2)^+$ reflection of holmium for the same range of incident photon energies. Solid lines are drawn only to guide the eye.
4. Integrated magnetic intensity of the $(0,0, L=2+\tau)$ magnetic reflection and of its harmonics located at $L=2+2\tau, 2+3\tau$, and $2+4\tau$, plotted as the incident x-ray energy is tuned through the L_{III} absorption edge. Solid lines are drawn only to guide the eye.
5. Total integrated magnetic intensity for the $(0,0,2)^+, (0,0,4)^+$, and $(0,0,6)^+$ reflections plotted on a logarithmic scale as the incident x-ray energy is tuned through the L_{III} absorption edge. The solid lines are the results of fits, which are discussed in the text.

Fig 2

Fig 3

Fig 4