

# Circular magnetic X-ray dichroism in crystalline and amorphous GdFe<sub>2</sub>

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With the advent of intense synchrotron radiation sources there has been renewed interest in utilizing x-rays to examine the magnetic properties of a variety of materials. One technique which has recently attracted great interest is Circular Magnetic X-ray Dichroism (CMXD).<sup>1</sup> CMXD is defined as the difference,  $\mu_c = \mu^+ - \mu^-$ , between the absorption of left and right circularly polarized x-rays by a magnetized sample, with  $\mu^+(\mu^-)$  representing the absorption coefficient for x-rays with the wave vector parallel (antiparallel) to the local magnetic moment of the absorbing atom. Since x-ray absorption involves transitions from well understood core levels with well defined angular momenta, observed structure in the dichroic spectra can yield information about the ground state spin polarization and spin-orbit coupling of final states.<sup>2</sup> Further, the information obtained is element and orbital specific since the technique requires scanning through a specific absorption edge. This technique can provide unique information since most magnetic measurements provide information about the total bulk moment. In the case of rare earth L<sub>2</sub> and L<sub>3</sub> edges the final states correspond to empty levels within the 5d bands which are primarily responsible for mediating the ordering among the 4f local moments.

To become a useful tool for probing electronic and magnetic structures CMXD spectra should also be material specific, i.e., sensitive to the changes in the band structure produced by different local environments. In this study, we compare the CMXD spectra of amorphous GdFe<sub>2</sub> with its crystalline counterpart and with a first principles theoretical calculation of the dichroic spectra for crystalline GdFe<sub>2</sub>. Changes in coordination and nearest neighbor distances in amorphous GdFe<sub>2</sub> should produce sufficient changes in the electronic structure to be detectable by CMXD measurements. Amorphous rare earth-transition metal materials are of considerable interest because of their many unique magnetic and magneto-optical properties.<sup>3</sup> For a better understanding of these effects, knowledge of the spin-polarization and spin-orbit coupling of individual orbitals is necessary.

CMXD analysis is, in principle, well suited to providing such information. Quantitative measurements of the degree of spin-orbit coupling are possible by employing recently derived sum rules, which relate the integrated intensity of the dichroic  $\mu_c$  and normal  $\mu_0$  absorption to the ground state values of the orbital ( $L_z$ ) and spin ( $S_z$ ) parts of the magnetic moment. Until now, a separate determination of the spin and orbital magnetic moments has been possible only by non-resonant magnetic x-ray scattering.<sup>5</sup> Owing to the small size of the magnetic cross section, however, this technique has been limited to samples with large magnetic moments (i.e. Gd  $\mu(4f) = 10\mu_B$ ; where  $\mu_B$  is the Bohr moment).<sup>6</sup> Further, this technique is not orbital specific and requires measurement of several magnetic diffraction peaks with different  $q$  values. CMXD, on the other hand, can give the values of moments on the order of  $0.01\mu_B$  with measurements at just the L<sub>2</sub> and L<sub>3</sub> absorption edges.

The CMXD measurements were made at the Cornell High-Energy Synchrotron Source bending magnet D line making use of elliptical polarization ( $P_c \approx 0.66 \pm 0.10$ ) of the synchrotron beam 0.11 mrad above the electron orbital plane. The beam was diffracted by a double crystal Si(220)

monochromator yielding an energy resolution of  $\sim 1.5$  eV in the vicinity of the Gd L edges. In order to eliminate harmonic contamination of the incident beam, the x-rays were reflected from a flat quartz mirror placed after the monochromator. The magnetization of the sample was reversed by a 3.5kG electromagnet, with the magnetic field oriented at 30° to the beam direction.

The polarization of the field was flipped every 2s at each step in an energy scan through the edges, thus producing two absorption spectra.  $I^+$  is the transmitted intensity when the magnetic moment of the sample and photon wave vector are in the same direction and  $I^-$  the transmitted intensity when the two are in opposite directions. We relate these to the dichroic signal by

$$\mu_c d = \frac{1}{M' P_c \cos \theta} \left( \ln \frac{I_0^+}{I^-} - \ln \frac{I_0^-}{I^+} \right) \quad (1)$$

where  $I_0^\pm$  are the incident intensities and  $d$  the effective sample thickness. In order to account for different experimental conditions and sample characteristics, the data were normalized by  $M'$ , the fraction of the T=0K saturation magnetization attained at room temperature with the field employed,  $P_c$  the degree of circular polarization of the incident beam, and  $\cos \theta$ , where  $\theta$  is the angle between the photon beam direction and the magnetic field direction.

The experimental spectra,  $\mu_0$  and  $\mu_c$ , taken in 0.5eV steps at the Gd L<sub>2</sub> and L<sub>3</sub> edges along with theoretical curves for crystalline GdFe<sub>2</sub> are shown in figs. 1a and 1b. A large enhancement of the amorphous dichroic spectra over the crystalline spectra is observed. This difference is believed to be due to a decreased spin-polarization of the 5d band in the amorphous material compared to the

(Figure 1) Top: Absorption of crystalline and amorphous samples. Middle: Dichroism signal  $\mu_c$  of crystalline and amorphous samples. Bottom: Theoretical curve for crystalline  $GdFe_2$ . a) Dichroism at the  $Gd L_3$  edge. b) Dichroism at the  $Gd L_2$  edge.

crystalline caused by a smaller Gd-Fe coordination. The Fe coordination around each Gd atom is  $6.5 \pm 0.6$  in amorphous  $GdFe_2$  as compared to 12 for the crystalline compound while the nearest neighbor distances remain roughly the same.<sup>7</sup> This smaller coordination number reduces the 5d(Gd)-3d(Fe) exchange thus diminishing the spin polarization of the 5d(Gd) band. This smaller degree of exchange reduces both the 5d and 3d moments, but since they are oppositely oriented their sum remains essentially constant. This results in a greater number of unoccupied holmium spin up states above the Fermi energy producing the observed enhanced signal. It should be kept in mind that CMXD is a measure of the empty state polarization. Thus, in this case, a smaller net 5d spin moment corresponds to a larger CMXD signal.

At the rare earth  $L_{2,3}$  edges, with initial p ( $l=1$ ) states, final d ( $l=2$ ) states, and 5d electron occupancy  $n \approx 1.8$  (from the band theory), the sum rules for the orbital ( $\langle L_z \rangle$ ) and spin ( $\langle S_z \rangle$ ) moments reduce to the following simple expressions,

$$\int_{L_2-L_3} d\omega \mu_c \Big/ \int_{L_2-L_3} d\omega 3\mu_o = 16.4 \langle L_z \rangle \quad (2)$$

and,

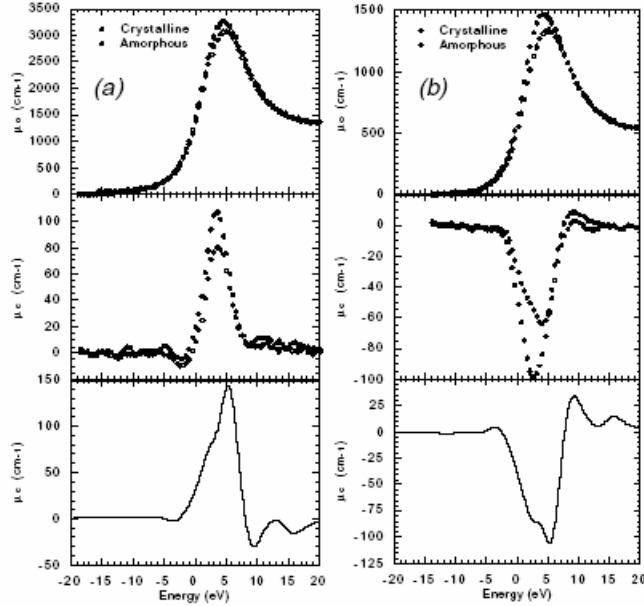
$$\left\{ \int_{L_3} d\omega \mu_c - 2 \int_{L_2} d\omega \mu_c \right\} \Big/ \int_{L_2-L_3} d\omega 3\mu_o = 12.3 \left( \langle S_z \rangle + \frac{7}{2} \langle T_z \rangle \right), \quad (3)$$

with  $3\mu_o = \mu^+ + \mu^- + \mu_o$ . In the expression above  $\langle T_z \rangle$  is the spatial average of the magnetic dipole operator<sup>8</sup> which is generally not  $\langle S_z \rangle$  in the expression for rare earth  $L_{2,3}$  edges. Thus, for the rare earth 5d states a quantitative value of only the orbital moment can be obtained.

Using these expressions, orbital moment values of  $0.018 \pm 0.005\mu_B$  and  $0.005 \pm 0.003\mu_B$  were obtained for the crystalline and amorphous samples respectively. The uncertainty in the size of the moments arises primarily from the uncertainty in the degree of circular polarization. Since both the amorphous and crystalline data are scaled by this value, the error in relative difference between the amorphous and crystalline samples is  $\sim 15\%$  less. The smaller value of the 5d band spin polarization in the amorphous sample is also responsible, in part, for the substantially smaller  $\langle L_z \rangle$  as compared to the crystalline sample, since a smaller net 5d moment implies a smaller orbital moment. The domi-

value of the 5d orbital moment observed by experiment,  $0.018 \pm 0.005\mu_B$  for crystalline  $GdFe_2$ , agrees well with the value obtained from theory,  $0.014\mu_B$ .

This study has demonstrated that the differences in the spin dependent band structure between amorphous and crystalline materials are easily observable by CMXD. Further, it has been shown that the differences can be quantified using the recently derived sum rules to obtain the size of the orbital moments. This information is unique since other methods provide information on the bulk moment and not that of individual orbit-



nant mechanism responsible for the quenching of the orbital moment, however, is believed to be the more random crystal field symmetry present in the amorphous sample. The average crystal field at a particular Gd site should be substantially more asymmetric in the amorphous compound as compared to the crystalline sample leading to decreased effectiveness of the spin-orbit coupling to produce an orbital polarization of the 5d conduction bands.

The theoretical dichro spectra at the  $L_2$  and  $L_3$  edges reproduce the general features observed in the experimental spectra, however they possess more pronounced structure. It is believed that this is due to the neglect of core-hole effects. The inclusion of core hole effects would draw in, and narrow, the 5d band compressing the signal near the Fermi energy, as observed in experiment. The sum rules are not affected by the core hole, since they involve an integration over all states. The

als and generally can not separate the orbital and spin contributions. This is important in materials such as rare earth transition metal compounds where changes in the 5d-3d spin polarization can cancel in a bulk measurement. This information should prove valuable in analyzing the magnetic and magneto-optical properties of amorphous rare earth-transition metal materials. It is expected that, with the advent of third generation synchrotron sources, CMXD should become an increasingly important tool to probe the magnetic properties of a variety of materials.

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