Experimental proof of magnetic x-ray dichroism

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What we believe to be the first experimental results have been obtained on strong magnetic x-ray dichroism in the $M_{4,5}$ absorption spectra of magnetically ordered rare-earth materials, in accordance with recent predictions.

The feasibility of using x-rays to determine the magnetic structure of magnetically ordered materials by magnetic dichroism has recently been predicted theoretically.[1] Strong magnetic x-ray dichroism (MXD) is expected in the $M_{4,5}$ absorption edge structure of rare-earth-metal compounds. Polarized synchrotron radiation can therefore be used to reveal information on the local rare-earth-metal magnetic moments in solids, thin films, and surfaces. In this Brief Report we will give what we believe is the first experimental proof of this effect.

The $M_{4,5}$ absorption in rare-earth-metal compounds shows good agreement with the atomic Hartree-Fock calculations for the transitions from the $4f^n(J)$ Hund's rule ground state to the manifold of $3d^9 4f^{n+1}(J')$ final states.[2-4] Although in x-ray absorption spectroscopy (XAS) hundreds of excited levels may be involved, one can distinguish between three different types of excitations, namely for $J'-J=-1$, 0, and 1.

In the presence of a magnetic field the $2J+1$ degenerate ground state $4f^n(J)$ splits into sublevels $M_J=-J, \ldots, +J$. The relative population of these sublevels depends on the temperature. That the polarization vector of the x rays has a drastic effect on the spectrum can be seen from the simple case where only $M=-J$ is populated ($T=0$ K). With the polarization direction parallel to the magnetization only the $\Delta M=0$ transitions are allowed. The transitions $J'-J=-1$ will then vanish, because the $M'=-J$ sublevel is not present in the $J'=J-1$ state.

Here, we will illustrate in more detail the use of MXD on terbium iron garnet (TbIG), which has a rather complicated magnetic structure. The rhombohedral (or trigonal distorted cubic) cell of ferrimagnetic rare-earth-metal iron garnets ($R_3Fe_5O_{12}$) contains eight formula units.[5] The Fe^{3+} ions occupy the 24 d (tetrahedron) and the 16 a (octahedron) positions. The larger R^{3+} ions occupy the 6 c (dodecahedral) positions. Below the Néel temperature the spins of the 24 d and 16 a irons are ordered antiparallel along the [111] axis. The rare-earth-metal moments couple antiferromagnetically to the net iron moment.[6] By symmetry the 6 c sites can be divided into c_1, c_2, c_3 and c_1',c_2',c_3', where c_2 (c_2') and c_3 (c_3') are obtained from the c_1 (c_1') by rotation around the trigonal [111] axis. From neutron diffraction at 4.2 K the Tb moments in TbIG are known to form a double umbrella structure.[7,8] The magnetic moments on c_1 and c_1' are both in the $(0\bar{1}1)$ plane, having angles $\beta=30.79°$ and $\beta'=-28.07°$ with the trigonal axis, and absolute values $m=8.18\mu_B$ and $m'=8.90\mu_B$, respectively. Above 4.2 K the values of β, β', m and m' are expected to decrease.[7]

In our experiment, a single crystal of TbIG was mounted on a rotatable helium-flow cryostat in an ultrahigh vacuum of $\sim 10^{-10}$ Torr. The temperature at the surface of the sample was 55 ± 5 K. A Co_5Sm permanent magnet provided a field of ~ 2 kG parallel to the [111] surface normal, which is the easy direction of magnetization.

Synchrotron radiation from the 540-MeV storage ring ACO (Anneau de Collision d'Orsay) at the Laboratoire pour l'Utilisation du Rayonnement Electromagnétique was monochromatized with a constant-deviation double-crystal monochromator. Using beryl $(10\bar{1}0)$ crystals (morganite) the energy resolution at the Tb M_5 edge is 0.9 eV full width at half maximum.[3] In the spectral region of interest the x-ray flux is invariant with photon energy. The emitted radiation in the equatorial plane of the storage ring is linearly polarized. In the actual setup it is s polarized with respect to the beryl crystals and p polarized with respect to the sample.

XAS spectra were measured using the electron yield

method,[3] and were obtained at different angles (α) between the polarization vector of the incident radiation and the [111] magnetization direction.

The experimental M_5 spectra for various values of α are given in Fig. 1. The solid lines are theoretical curves, as discussed below. As seen, the intensities of the two major peaks are strongly polarization dependent. Parallel polarization with respect to the net magnetization enhances the left-hand peak, whereas for perpendicular polarization the right-hand peak increases in intensity.

Neglecting crystal field effects, the theoretical curves in Fig. 1 can be calculated with the theory given in Ref. 1. The Boltzmann-averaged sum of the absorption intensities for transitions between the magnetic sublevels αJM in the ground state (α labels different levels of equal J) and the excited levels $\alpha' J' M'$ is given by the line strength times a factor

$$\langle A^{\theta}_{JJ'}\rangle = \langle A^{\parallel}_{JJ'}\rangle \cos^2\theta + \langle A^{\perp}_{JJ'}\rangle \sin^2\theta \ , \tag{1}$$

where θ is the angle between the magnetic moment and the electrical polarization vector of the incident radiation.

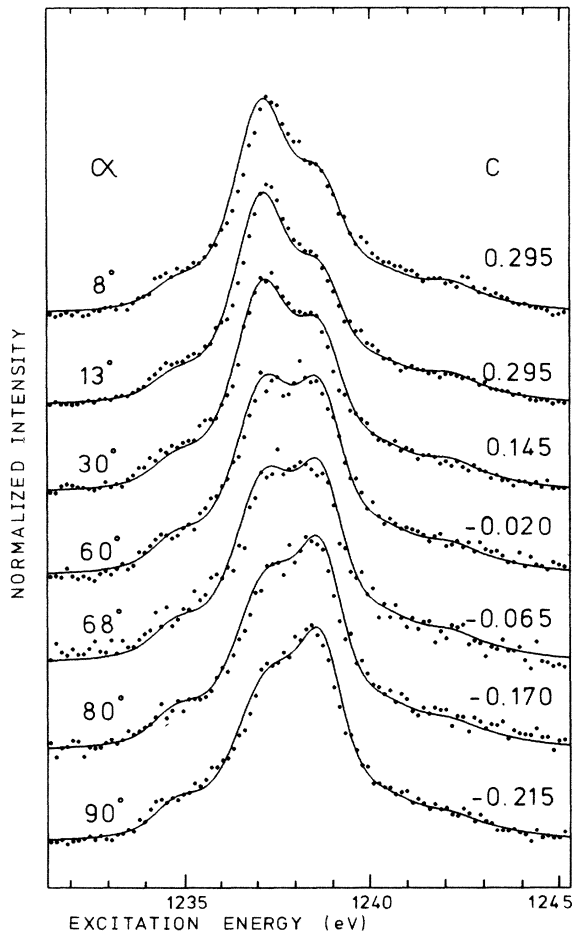

FIG. 1. Experimental M_5 absorption spectra of TbIG at various values of α, which is the angle between the polarization vector of the x rays and the [111] magnetization direction. The solid lines are fits using Eq. (5). The optimum values of C are indicated.

The temperature-dependent factors for parallel and perpendicular polarization $\langle A^{\parallel}\rangle$ and $\langle A^{\perp}\rangle$ are related as

$$\langle A^{\perp}_{JJ'}\rangle = \tfrac{1}{2}[1/(2J+1) - \langle A^{\parallel}_{JJ'}\rangle] \ . \tag{2}$$

There are only three different factors $\langle A^{\parallel}_{JJ'}\rangle$, viz. $J'-J=-1,0,+1$. They are given as a function of the temperature-dependent quantity $\langle M^2\rangle$ in Eq. (6) of Ref. 1.

If the magnetic moments are not collinear the angular average of Eq. (1) has to be taken. For a single umbrella structure we obtain

$$\langle \cos^2\theta\rangle = \tfrac{1}{3} + \tfrac{2}{3}(\tfrac{3}{2}\cos^2\alpha - \tfrac{1}{2})(\tfrac{3}{2}\cos^2\beta - \tfrac{1}{2}) \ , \tag{3}$$

where α is the angle between the [111] magnetization direction and the polarization vector, and β is the angle between the [111] direction and the magnetic moment. For the double umbrella the average of $\cos^2\beta$ has to be taken. Combination of Eq. (6) of Ref. 1 with Eqs. (1), (2), and (3) of this work yields

$$\begin{aligned}
\langle A^{\alpha}_{J,J+1}\rangle &= \frac{1}{3(2J+1)}\left[1 - \frac{J(2J-1)}{(J+1)(2J+3)}C\right] \ , \\
\langle A^{\alpha}_{JJ}\rangle &= \frac{1}{3(2J+1)}\left[1 - \frac{2J-1}{J+1}C\right] \ , \\
\langle A^{\alpha}_{J,J-1}\rangle &= \frac{1}{3(2J+1)}(1-C) \ ,
\end{aligned} \tag{4}$$

where

$$\begin{aligned}
C &= (\tfrac{3}{2}\cos^2\alpha - \tfrac{1}{2})(\tfrac{3}{2}\cos^2\beta - \tfrac{1}{2}) \\
&\quad \times \frac{\langle M^2\rangle - \frac{1}{3}J(J+1)}{J^2 - \frac{1}{3}J(J+1)} \equiv C_{\alpha} C_{\beta} C_M \ .
\end{aligned} \tag{5}$$

Note that the factors $\langle A\rangle$ have axial symmetry around the [111] axis.

In order to observe an anisotropic absorption ($C \neq 0$), three conditions must be met: α and β have to be different from the magic angle (54.7°), and $\langle M^2\rangle$ must be unequal to its isotropic value $\frac{1}{3}J(J+1)$. This value is reached when all sublevels are equally populated. The largest anisotropy is expected if β is zero and $\Theta = kT/g\,|\mu_B|\,H$ is small.

Line strengths and energies were calculated with an atomic Hartree-Fock program.[9,10] The theoretical spectra for values of C which gave the best fit to the experimental data are displayed in Fig. 1. As seen, there is a good agreement between theory and experiment for all the structures present in the spectrum.

The optimized values of C are shown in Fig. 2. The solid lines give C as a function of α for various constant values of $C_\beta C_M$. The least-squares value for $C_\beta C_M$ is equal to 0.312, with an error of ~ 0.05. We can verify this value of $C_\beta C_M$ by results from magnetization measurements. At $T = 55 \pm 5$ K the Tb contribution to the magnetization in TbIG is equal to $m = (18.0 \mp 0.8)\mu_B$ per formula unit.[6] Using $m = 3(\cos\beta) g \langle M\rangle \mu_B$ and Eq. (5) in Ref. 1 this gives $C_\beta C_M = 0.30 \mp 0.03$ for $\beta = 30$, or $C_\beta C_M = 0.34 \mp 0.03$ for $\beta = 0°$. We obtain a reasonable value for $C_\beta C_M$, which shows that the magnetic dichroism

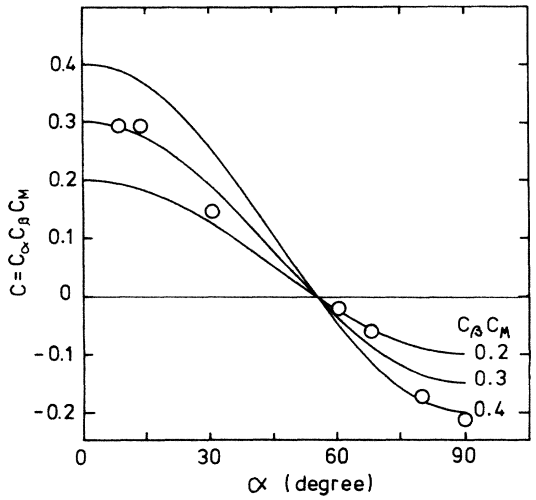

FIG. 2. The optimum values of C, obtained by fitting the experimental spectra (○). C as a function of α for various constant values of $C_\beta C_M$ is given by the solid lines.

effect is in agreement with other magnetic measurements. It is also seen that, with the given error bars in the numbers, the experiment cannot accurately predict the value of β. However, the large scatter in the values for $C_\beta C_M$ can be reduced by improving the statistics in the experimental spectra and by regulating the temperature more precisely. In order to obtain more detailed information about the magnetic structure it is also important to measure at a lower temperature. For $\Theta = kT/g\,|\mu_B|\,H \lesssim 0.3$, only the lowest magnetic sublevel is filled, then C_M goes to one, and the temperature dependence disappears. We are planning improvements on our experimental setup in order to achieve this.

In summary, we conclude that the M_5 absorption spectrum of TbIG in the presence of a magnetic field shows strong polarization-dependent effects. These effects are in accord with the magnetic structure, multiplet calculations, and the predictions of Ref. 1.

MXD is complementary to other techniques, such as neutron diffraction and Mössbauer, because it can be applied to magnetically ordered thin films and surfaces, and because it is applicable for all magnetic rare-earth-metal compounds and probably can be extended to transition-metal compounds. Because MXD yields information on $\langle M^2 \rangle$ this technique can also be used for antiferromagnets where $\langle M \rangle = 0$.

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