

Variation of XMCD spectrum with temperature at R $L_{2,3}$ -edges in $R_3Fe_5O_{12}$ (R=Gd and Er)

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The variation of X-ray magnetic circular dichroism (XMCD) with temperature has been measured at R $L_{2,3}$ -edges in $R_3Fe_5O_{12}$ (R=Gd and Er). A drastic change in the spectral profile has been observed at low temperatures. The variation of XMCD peak intensity, corrected by the contribution of Fe with temperature, is in agreement with the magnetization of R sublattice. The results suggest that Fe 3d polarization plays an important role to produce the R $L_{2,3}$ -edges XMCD spectrum.

Keywords : X-ray magnetic circular dichroism, rare-earth $L_{2,3}$ -edges, rare-earth iron garnets

1. Introduction

X-ray magnetic circular dichroism (XMCD) is a powerful tool to obtain information about electronic and magnetic properties of absorbing atoms. The spectrum resulting from interplay between exchange interaction and spin-orbit coupling is very sensitive to subtle changes in the electronic states. Here, we investigate the XMCD spectrum of rare-earth (R) in the $R_3Fe_5O_{12}$ system. In particular, we focus on the appearance of a split structure in the XMCD spectrum at room temperature.

To study the role of R atoms in magnetic ordering, XMCD measurements at the R $L_{2,3}$ -edges have included intermetallic compounds such as R-Fe systems (Chaboy *et al.*, 1997, 1998; Dartyge *et al.*, 1998), R_2Co_{17} (Fischer *et al.*, 1990, 1991), $Eu_3Fe_5O_{12}$ (Schütz *et al.*, 1996) and $Ho_3Fe_5O_{12}$ (Shimomi *et al.*, 1993). However, the complicated profile of the XMCD spectrum has restricted a fuller understanding of the magnetic properties.

The XMCD spectra have been discussed using a simple model at the R $L_{2,3}$ -edges in R_2Co_{17} , in which 5d electron is assumed to be in the ground state and is polarized by 4f electrons in the Hund's rule ground state through the 4f-5d exchange interaction (Jo & Imada, 1993). However, sign of the integrated intensity was systematically inconsistent with the experimental results. To solve this problem, Matsuyama *et al.* (1997) have taken shrinkage in R 5d radial functions into account, in addition to the contributions of electric dipole (E1) and quadrupole (E2) transitions. They have successfully interpreted the overall spectral structure. However, a structure split into two negative (positive) peaks, observed at Eu L_3 -edge in $Eu_3Fe_5O_{12}$ (Schütz *et al.*, 1996) and at Ho L_3 -edge (L_2 -edge) in $Ho_3Fe_5O_{12}$ (Shimomi *et al.*, 1993) at room temperature, has never been reproduced. In order to elucidate the origin of the split structure, we have measured variation of XMCD spectrum with temperature at R $L_{2,3}$ -edges in $R_3Fe_5O_{12}$ (rare-earth iron garnet: R-IG) system.

2. Experimental

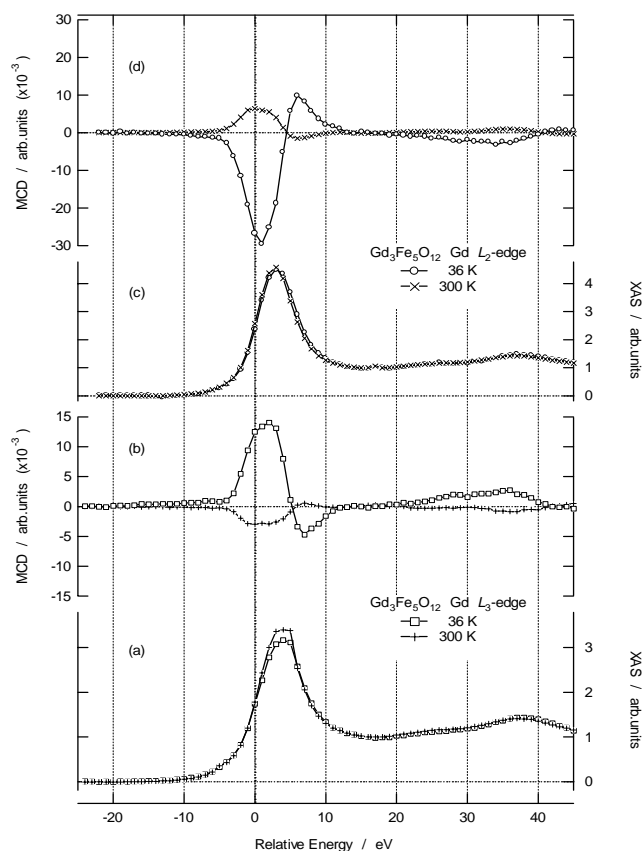
We used polycrystalline $Gd_3Fe_5O_{12}$ (Gd-IG) and $Er_3Fe_5O_{12}$ (Er-IG). In these ferrimagnets, the magnetic ions occupy three crystallographic sites: 16 Fe^{3+} ions in octahedral site (*a*-site), 24 Fe^{3+} ions in tetrahedral site (*d*-site), and 24 R^{3+} ions in dodecahedral site (*c*-site). Magnetic moments of Fe^{3+} ions in *a*-site are coupled antiferromagnetically with those of Fe^{3+} ions in *d*-site and ferromagnetically with those of R^{3+} ions in *c*-site. Gd-IG and Er-IG have a compensation temperature T_{comp} , at which bulk magnetization vanishes, corresponding to 286 K in Gd-IG and 83 K in Er-IG. At temperatures below T_{comp} , R ions play a dominant role of the magnetic properties. For X-ray absorption measurements, the powdered samples were uniformly spread on Scotch tape. $Lu_3Fe_5O_{12}$ (Lu-IG) was also used to compare with the spectrum in Gd-IG and Er-IG.

XMCD measurement in the transmission mode was made on Beamline 29XU of SPRING-8 (Tamasaku *et al.*, 2000). The beamline was composed of an in-vacuum linear undulator and a rotated-inclined double-crystal monochromator (Yabashi *et al.*, 1999) equipped with Si 111 crystal. A diamond X-ray phase retarder was used in the Laue geometry with the 220 reflection plane. The spectrum was recorded using the helicity-modulation method (Suzuki *et al.*, 1998) with a fixed magnetic field of 1.15 T. Circular polarization was alternated between plus and minus helicities using the phase retarder oscillated with 40 Hz. X-ray intensity was monitored by ionization chamber filled with N_2 gas before and after the sample. The degree of circular polarization was estimated to be 0.9 in the energy range. Temperature variation was measured in the range from 36 up to 300 K. Temperature of the sample was monitored by using a GaAlAs diode installed in the sample holder. Absorption edge energy E_0 was determined by the first inflection point of X-ray absorption near edge structure (XANES) spectrum.

3. Results and Discussion

Figure 1 and 2 show XANES and XMCD spectra as a function of energy relative to the Gd $L_{2,3}$ -edges in Gd-IG and the Er $L_{2,3}$ -edges in Er-IG, respectively. The spectra at 36 K and 300 K are compared in the figure. At room temperature, Gd-IG shows the XMCD spectrum with a negative sign at L_3 -edge and a positive one at L_2 -edge. The spectrum is split at the L_3 -edge and a shoulder structure appears at the L_2 -edge. Although both the L_3 and L_2 spectra in Gd-IG are relatively simple profile, those in Er-IG are obviously split into two adjacent peaks by a structure with an opposite sign at $(E-E_0) \sim +2$ eV, whose intensity of the L_3 (L_2) spectrum is increased (decreased) with 4f electron number (Kawamura *et al.*, 2000). The E2 contribution, labeled as A in Fig.2, is also identified at $(E-E_0) \sim -7$ eV at the L_3 -edge in Er-IG.

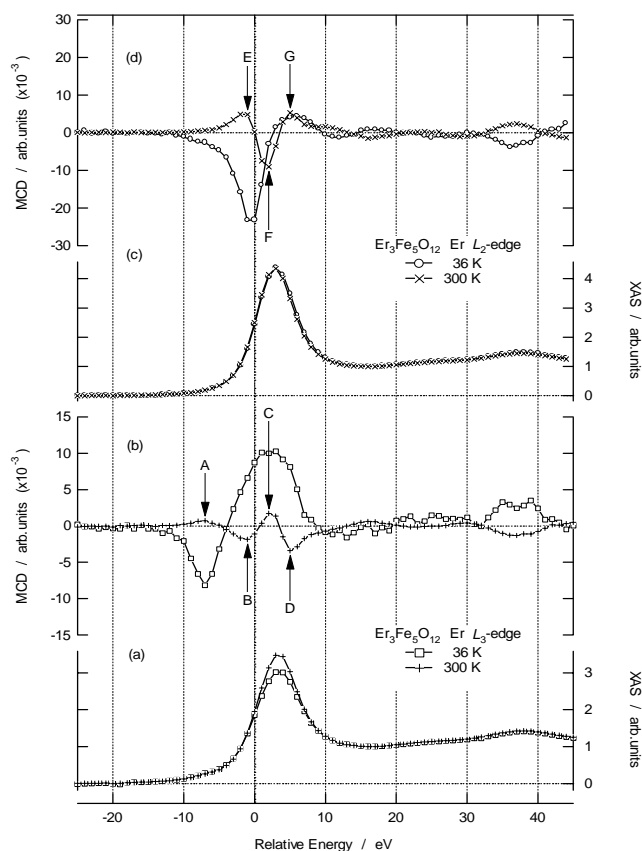
In the L_3 -edge XANES spectrum, the white-line intensity is reduced at 36 K in both Gd-IG and Er-IG, whereas the L_2 -edge does not show such reduction. Ratio of the edge-jump at 36 K to that at 300 K is evaluated to be 93% in Gd-IG and 87% in Er-IG. This result suggests that the density of states of R 5d-bands is changed by 4f-5d exchange interaction or hybridization between O 2p and Fe 3d orbitals when the temperature is decreased. On the other hand, the XMCD intensity is drastically enhanced at 36 K compared with that at 300 K, and the sign is reversed because of the compensation phenomenon. It appears that the split structure in Er-IG is covered by some large effect that becomes predominant at low temperatures. This phenomenon may be interpreted as the contribution of R ions, which is drastically

**Figure 1**

Gd L_3 -edge, (a) XANES and (b) XMCD spectra, and Gd L_2 -edge, (c) XANES and (d) XMCD spectra in Gd-IG. Absorption edge energy was determined from the first inflection point in the XANES spectrum.

enhanced at low temperatures and eventually exceeds the component of Fe ions. The present observations indicate that Fe 3d states strongly affect on the R L -edge XMCD spectrum and play an important role at room temperature.

To confirm this speculation, we considered the correlation between the XMCD peak intensity and bulk magnetization in Er-IG and paid attention four peaks in the L_3 -XMCD spectrum, labeled as A, B, C, and D in Fig.2 (b), and three peaks in the L_2 -XMCD spectrum, labeled as E, F, and G in Fig.2 (d). The peak intensity was plotted as a function of temperature in Fig.3. The magnetization of Er^{3+} sublattice, denoted as M_{Er} , is also shown in the figure to compare with the XMCD intensity. The value of M_{Er} was evaluated from the difference in bulk magnetization between Er-IG and Lu-IG, because the magnetization in Lu-IG originates in only Fe^{3+} ions. In general, the XMCD intensity is proportional to magnetization of the absorbing magnetic atom. Therefore, it is expected that the XMCD intensity varies with temperature in the same manner of the temperature variation of magnetization. For all of the peaks A – G, the XMCD intensity was subjected to correction by the value of magnetization at 36 K. As shown in Fig.3, an agreement between the XMCD intensity and M_{Er} curve is poor at higher temperatures for the L_3 -edge and over the measured range for the L_2 -edge. This result indicates that some additional effect should contribute to the XMCD intensity at the R $L_{2,3}$ -edges. This may be consistent with

**Figure 2**

Er L_3 -edge, (a) XANES and (b) XMCD spectra, and Er L_2 -edge, (c) XANES and (d) XMCD spectra in Er-IG.

observation of the Lu $L_{2,3}$ -edges XMCD spectra in magnetic compounds including Lu (Baudelet *et al.*, 1990; Chaboy *et al.*, 1997; Kawamura *et al.*, 2000).

We regard Fe molecular field as the additional effect. Then, to estimate intrinsic Er component, contribution of Fe ions should be removed from the Er L -edge XMCD spectrum in Er-IG. The contribution of Fe ions was assumed to be the spectrum in Lu-IG. The corrected XMCD spectrum in Er-IG could be obtained by subtracting the spectrum in Lu-IG from the original spectrum in Er-IG. As a result, the peak intensity of the corrected XMCD signal is plotted as a function of temperature in Fig. 3, indicated by closed square and circle. The XMCD peak intensity is in better agreement with the M_{Er} curve. From this analysis, we conclude that the split structure, labeled as C and F, results from the contribution of Fe 3d polarization, although R L -edge XMCD spectrum is mainly explained by the combination of 4f-5d exchange interaction and the shrinkage in R 5d radial functions. Therefore, in R-Fe system the Fe molecular field may influence on magnetic polarization in the R 5d states.

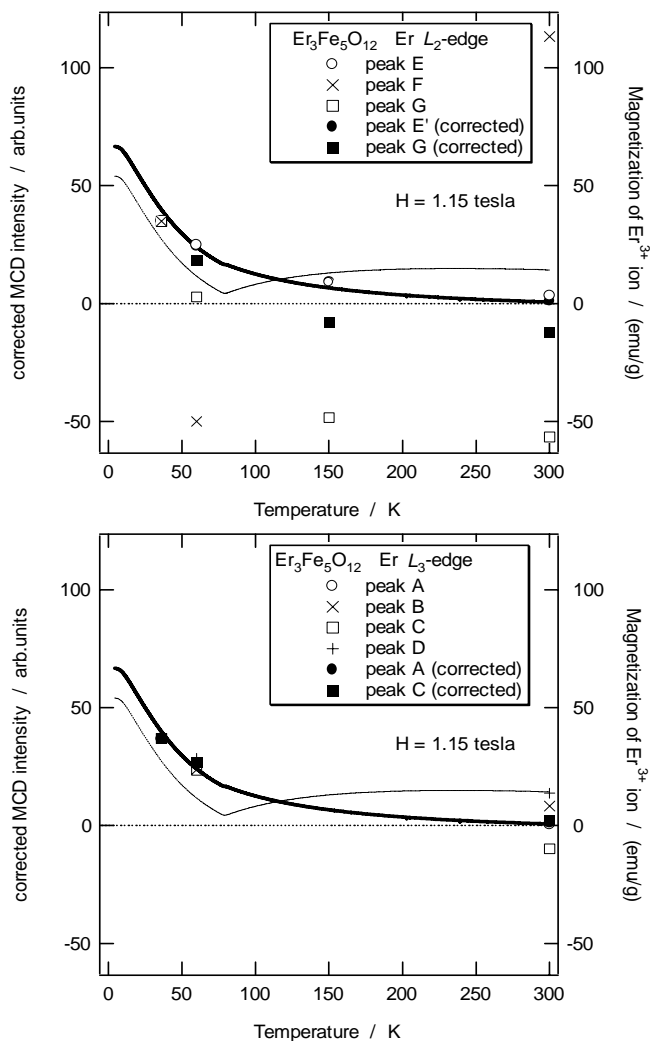


Figure 3

Temperature dependence of XMCD intensity associated with the peaks A-G shown in Fig.2. The intensity was normalized by the value at 36 K. Closed symbols indicate the corrected value of the XMCD intensity in Er-IG. The solid line displays the Er sublattice magnetization. The dotted line displays the magnetization in Er-IG.

4. Conclusion

We have measured the variation of XANES and XMCD spectra with temperature at the Gd and Er $L_{2,3}$ -edges. At low temperatures, a decrease in the white-line peak intensity has been observed at the L_3 -edge only. For the first time, a drastic change in the XMCD spectrum has been also observed at both the L_3 - and L_2 -edge in R-IG. We conclude that the Fe $3d$ polarization plays an important role to the XMCD spectrum at the R $L_{2,3}$ -edges at room temperature.

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