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Temperature Dependence of the Rare-Earth Sublattice Magnetic Moments in $R_3Fe_5O_{12}$ (R=Sm, Gd, and Dy) investigated by X-ray Magnetic Circular Dichroism

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The exchange coupling constants J_{ca}, J_{cd}, J_{cc} have been determined in $R_3Fe_5O_{12}$ (R-IG; R=Gd, Sm, and Dy), by x-ray magnetic circular dichroism (MCD) measurements at the rare earth L_2-edge at between 2 and 300 K. The component originating from the iron sublattice was adequately removed from the measured MCD spectra, then the temperature dependence of magnetization of Gd, Dy, and Sm sublattices were obtained independently. These magnetization curves were reproduced by the molecular field model. The exchange interaction between Fe *d* site and R *c* site is much stronger than the other interactions and that the sign of the exchange coupling among R sites is positive in Sm-IG and Dy-IG and negative in Gd-IG.
Key words: magnetic circular dichroism, rare earth iron garnet, element specific magnetization

1. INTRODUCTION

Rare earth iron garnet (R-IG) has been studied through the ages [1-4] and has been applied to magneto-optical devices [5,6] and magnetic bubble memories [7,8]. The microscopic investigation of electronic states in R-IG is an essential matter from a technological aspect as well as a physical point of view. The crystal structure of R-IG belongs to the space group O^{10}_h (*Ia3d*). Three magnetic sublattices, two of iron (*a* and *d* sites) and one of rare earth (*c* site), are collinear with the orientation of their spins along [1 1 1] axis. Net magnetization of a heavy rare earth garnet falls to zero as the temperature reaches T_{comp}, where the magnetic moments of *a*, *d*, and *c* sites cancel out each other. The temperature dependence of sublattice magnetizations and the magnetic structure in R-IG are dominated by the exchange interactions among spins on each magnetic sublattice. The thermal variation of sublattice magnetizations in Gd-IG has been investigated by Litster [3] and his calculation using the molecular field model fits well to the experimental magnetization curves. However, the sublattice magnetic moments of light rare earths, such as Sm in Sm-IG [4], are difficult to estimate because it is too small to be separate from the magnetization of iron sublattices by means of conventional macroscopic measurements.

The magnetic circular dichroism (MCD) technique based on core-level x-ray spectroscopy has been used as an element specific probe of magnetic materials. Because each element has unique core-level energies, one can obtain the information of the specified element by tuning the x-ray photon energy. High brilliant synchrotron radiation sources allow us to collect MCD signals in short time with high statistical accuracy. MCD spectra in the hard x-ray region such as $L_{2,3}$-edges of rare earths or *K*-edge of *3d* metals have a complicated structure in some cases [9,10]. Asakura has theoretically pointed out that the complexities might come from an influence of the adjacent magnetic elements [11]. He reported that MCD spectra at the rare earth $L_{2,3}$-edges in $R_2Fe_{14}B$ have a component due to *3d* state of the iron sublattices through the inter-atomic *5d-3d* interaction.

In this study, we investigated temperature dependence of MCD spectra at the rare earth L_2-edge in some R-IGs to investigate those sublattice magnetization. The influence of the iron sublattices, which modifies MCD spectra, is estimated. After properly removing this influence, the exchange coupling constants among sublattices were determined by the molecular field analysis.

2. EXPERIMENTS

MCD experiments were carried out using polycrystalline samples of R-IG (R = Sm, Gd, and Dy) in the temperature range from 2 to 300 K. MCD spectra at the rare earth L_2-edge ($2p_{1/2} \rightarrow 5d$ transition) were measured at an undulator beamline BL39XU of the SPring-8 facility. MCD signals were obtained using the helicity-modulation technique [12] in the transmission. A magnetic field of 2T was applied using an electromagnet for measurements above 30 K and a superconducting magnet below 20 K.

3. RESULTS AND DISCUSSION

Fig.1 shows the MCD spectra of the rare earth L_2-edges in (a) Gd-IG, (b) Dy-IG and, (c) Sm-IG taken at several temperatures. In the figure, MCD spectra are normalized by the edge jumps of XAS. With increasing

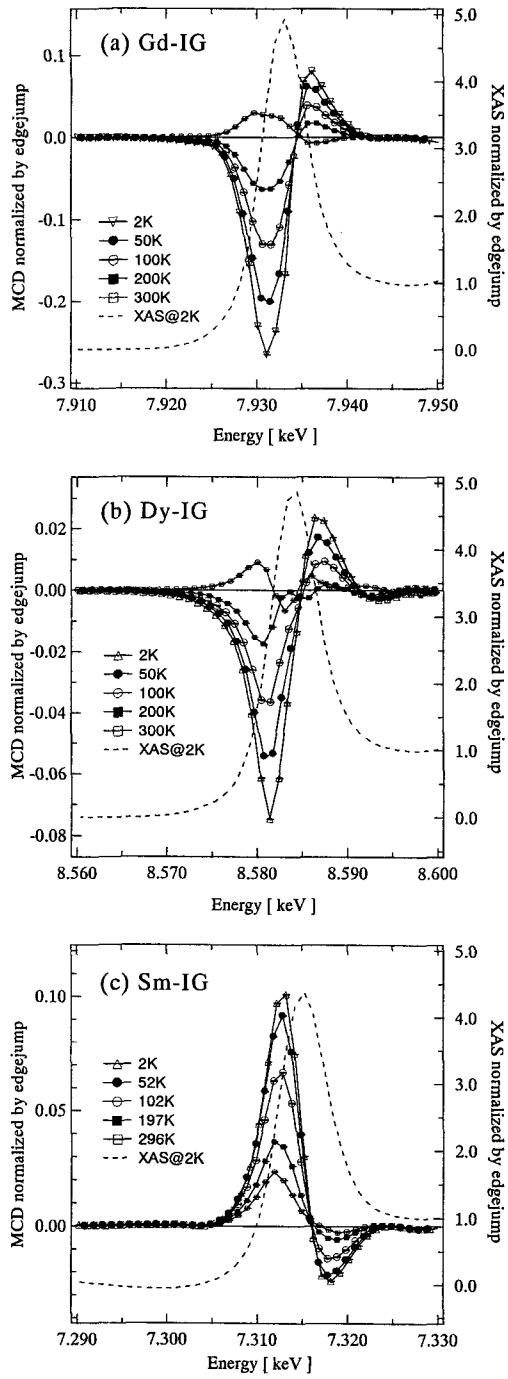

Fig.1 Rare earth L_2-edge MCD spectra in (a)Gd-IG, (b) Dy-IG, and (C)Sm-IG at several temperatures. Dashed line shows XAS of each sample. All MCD spectra are normalized by the edge jumps of XAS.

temperature, the MCD spectrum of each sample smoothly changes its shape and its amplitude reduces. For Gd-IG and Dy-IG, there are sign reversing at T_{comp} = 282K and 222K, respectively. The sign of MCD of Gd-IG and Dy-IG is negative below T_{comp}, on the other hand, that of Sm-IG is positive. This suggests that the direction of magnetization of iron *d* site is parallel to the external magnetic field in Sm-IG whereas antiparallel in Gd-IG and Dy-IG below T_{comp}. The temperature dependence of the intensity in Gd-IG is not consistent with the Gd sublattice magnetization reported in Ref. 2. The rare earth $L_{2,3}$-edge MCD in rare earth-transition metal compounds is thought to be composed of two components, one originates from the magnetic moment in rare earth *4f* shell and the other come from the magnetic moment of the adjacent iron sites. We have performed a fitting analysis to separate the MCD spectra into these two components. In the analysis, the fitting was based on the difference in temperature dependence of the sublattice magnetization on Gd and Fe sites, and reported magnetization data [2]. The resulting MCD spectra of Gd and Fe component are shown in Fig.2. The sum of those is compared with the experimental spectrum at 280 K. The Fe component has a considerable intensity. This means that the magnetization of iron sublattices affects the spin polarization of R *5d* shell perhaps via oxide ions.

The component of iron sublattice magnetization was removed from the measured MCD spectra of Dy-IG and Sm-IG in order to obtain the MCD components of contributed purely from Dy or Sm sublattice. Thermal variations of those MCD components, which should be proportional to the magnetization of R sublattices, are plotted in Fig.3. In this figure, Gd sublattice magnetization deduced by Litster [3] is shown by a solid line. The values of all components are normalized to be unity at 2 K. In Fig.2, Sm magnetic moment in Sm-IG shows slower reduction than Gd or Dy with increasing temperature.

In order to estimate the exchange interactions of the rare earth *c* site, that is, *a-c*, *d-c*, *c-c* interactions, we have performed fitting based on the molecular field model for the present ferri-magnetic system consisting of three magnetic sublattices, according to the expressions,

$$\sigma_c = B_{S_c}(x_c),$$

$$x_c = \frac{g^2\mu_B^2}{k_BT} S_c\left(N_a\lambda_{ca}S_a\sigma_a + N_d\lambda_{cd}S_d\sigma_d + N_c\lambda_{cc}S_c\sigma_c\right).$$

Here, σ_i is the magnetization of *i* sublattice, which is

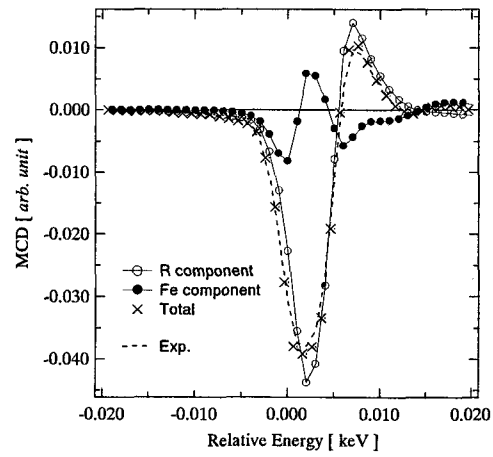

Fig.2 Rare earth component and iron component that contribute to MCD spectra at rare earth L_2-edge in Gd-IG at 280 K. The sum of two components is shown by cross, and dashed line is the measured spectrum.

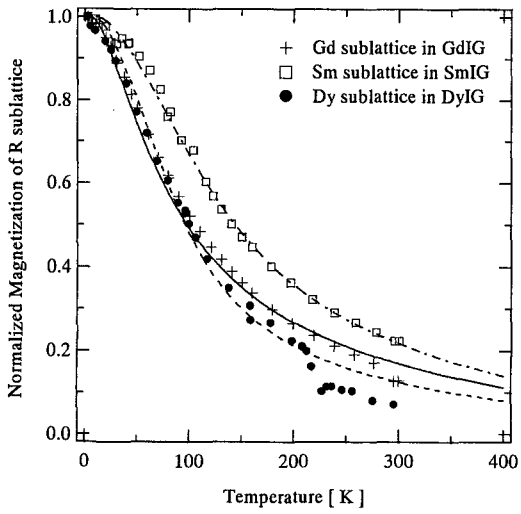

Fig.3 Temperature dependences of R sublattices in R-IG estimated from MCD components of Gd, Dy and Sm. Lines are the fitting curves of the molecular field analysis.

normalized by the magnetization at 0 K. The g is Lande's g factor. The S_i is the spin quantum number; $S_a = S_d = 5/2$ for the iron sites, and $S_c = 5/2$ for the Dy or Sm site, and N_i is number of magnetic ions in unit volume. λ_{ij} is the molecular field constants, which is combined to the exchange integral as,

$$\lambda_{ij} = \frac{2z_{ij}}{N_j g^2 \mu_B{}^2} J_{ij},$$

where z_{ij} is the coordination number of j ions around i ion. The values σ_a and σ_d in [3] are used as the magnetization of iron sublattices. The fitting curves of the Dy and Sm sublattice magnetization are shown in Fig.3 by a broken line and a dashed line, respectively, and Table I gives the obtained parameters J_{ca}, J_{cd}, and J_{cc}. This result shows that the exchange constant between Fe *d* site and R *c* site is much stronger than others, and the negative sign supports antiparallel coupling. The signs of *c-c* interaction and *d-c* interaction depend on the atomic number of R ion. In addiction, the temperature dependence of Sm sublattice magnetic moment in Sm-IG was estimated to be reliable values in this study using MCD technique as a probe of element specific information.

4. CONCLUSION

We have measured the MCD spectra at rare earth L_2-edge in Gd-IG, Dy-IG, and Sm-IG in the temperature range between 2 and 300 K. The temperature dependences of rare earth components in MCD spectra of Dy-IG and Sm-IG were obtained by subtracting the iron component that was estimated from MCD spectra in Gd-IG comparing the macroscopic magnetization data. The exchange constants deduced based on molecular field analysis show that the interaction between iron *d* site and rare earth *c* site is much stronger than other interactions.

Table I Exchange constants in R-IG deduced from the temperature dependence of the R sublattice magnetization.

	J_{ca} (cm^{-1})	J_{cd} (cm^{-1})	J_{cc} (cm^{-1})
Gd-IG[a]	-0.22	-3.46	-0.13
Dy-IG	0.64	-4.34	1.29
Sm-IG	-0.33	-4.98	1.01

[a] Ref. 3

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