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Multiple scattering approach to XMCD for Bi substituted Gd iron garnet

Ikuko Hojo\textsuperscript{1}, Shin-ichi Nagamatsu\textsuperscript{1}, Takashi Maruyama\textsuperscript{1}, Hiroshi Maruyama\textsuperscript{2}, Takashi Fujikawa\textsuperscript{1}

\textsuperscript{1}Graduate School of Advanced Integration Science, Chiba University, Yayoi-cho 1-33, Inage, Chiba 263-8522, Japan
\textsuperscript{2}Department of Physical Sciences, Graduate School of Science, Hiroshima University, 1-3-1 Kagamiyama, Higashi-Hiroshima, Hiroshima 739-8562, Japan

E-mail: hojo@graduate.chiba-u.jp

Abstract. We have studied Bi $L$-edge X-ray magnetic circular dichroism (XMCD) of Bi substituted Gd iron garnet (BiGd-IG) by using the relativistic multiple scattering calculations. BiGd-IG has giant magneto-optical response with substituted Bi. The XMCD analysis is suitable to understand the interesting behavior; it is important to understand the local magnetic and electronic structure around absorption atom Bi. Comparing the experimental spectra, we find that the proportion of magnetic atoms to nonmagnetic ones around Bi strongly influence the Bi XMCD spectra. We also investigate the effects of the spin polarization of the Bi $6p$ states.

1. Introduction

Bi substituted Gd iron garnet (BiGd-IG), which is a ferrimagnetic insulator, is well known as magnetic-optical material because it strongly enhances Faraday rotation in visible and near-infrared regions with increase of Bi [1], and is applied to the optical communication. According to Shinagawa \textit{et al}., these effects are related to the charge transfer transitions in iron-oxygen molecular complexes modified by covalent mixing of Bi $6p$ orbitals with oxygen $2p$ orbitals due to large spin-orbit interaction in Bi [2].

To understand the interesting properties, it is crucial to obtain detailed information on electronic and geometric structures around substituted Bi in garnet. For that purpose XMCD should be one of the promising tools because the substituted Bi are randomly distributed on Gd sites. A powerful technique to study magnetic order, neutron scattering has to be applied to ordered magnetic systems. XMCD is now widely used [3], and applied to related systems, rare earth iron garnets [4], and rare earth intermetallic materials $RFe_{11}Ti$ [5], and $R_2Fe_{17}$, $R_2Fe_{17}H_x$, $R_2Fe_{17}N_x$ [6].

In this paper, we study Bi $L$-edge XMCD of BiGd-IG and (BiYd)(FeGa)O by use of relativistic full multiple scattering calculations. In particular the influence caused by substituted Bi on nearby magnetic atoms are investigated in detail. We also study the effects of the spin polarization on Bi.
2. Theory

The XMCD theory, used in this paper, is based on the one electron relativistic multiple scattering theory proposed by Fujikawa and Nagamatsu [7]. The 4-spinor core state \( |c\rangle \) can be written in terms of the large component \(|\varphi_c\rangle\) and the small component \(|\chi_c\rangle\). By using full relativistic Green’s function \( G_D \), the X-ray absorption intensity is described as

\[
I(\omega) = -\text{Im}(\langle c|\Delta^* G_D(\varepsilon)\Delta|c\rangle) = T_{11}(\omega) + T_{12}(\omega) + T_{21}(\omega) + U_{11}(\omega) + \cdots.
\]

(1)

\( T_{11} \) is the most important term and \( T_{12}, T_{21}, U_{11} \) are relativistic correction terms which vanish in the nonrelativistic limit (\( c \to \infty \)). In the case of \( L_{2,3}\)-edge XMCD, \( \Delta T_{11} \), is dominant and is given by

\[
\Delta T_{11} = -2\text{Im}(\langle \varphi_c|\Delta^+ G(\varepsilon)\Delta_+|\varphi_c\rangle - \langle \varphi_c|\Delta^+ G(\varepsilon)\Delta_-|\varphi_c\rangle),
\]

where \( g(\varepsilon) \) is a full one-electron nonrelativistic Green’s function for the photoelectron energy \( \varepsilon \), and \( \Delta_\pm \) is electron-photon interaction operator with \( \pm \) circular polarization.

In the case of \( L_1\)-edge, the term of \( T_{11} \) has no contribution to XMCD because the core state is never influenced by spin-orbit coupling. For the randomlly oriented systems as considered here, the XMCD spectra are described by

\[
\Delta T = \Delta U_{11}
\]

(3)

where \( \Delta U_{11} \) is XMCD caused by spin-orbit coupling for photoelectrons,

\[
\Delta U_{11} = -2\text{Im}(\langle \varphi_c|\Delta^+ G \chi Q(V - \varepsilon)QG\Delta_+|\varphi_c\rangle - \langle \varphi_c|\Delta^+ G \chi Q(V - \varepsilon)QG\Delta_-|\varphi_c\rangle).
\]

(4)

3. Experiment

XMCD spectra at Bi \( L\)-edge for the powdered sample, \((\text{Gd}_{0.97}\text{Y}_{0.03}\text{Bi}_{1.14})(\text{Fe}_{4.15}\text{Ga}_{0.85})\text{O}_{12}\), are measured on the 39XU beamlines at SPring-8 Facility using the helicity-modulation method [8]. The XMCD measurements are performed at room temperature and under an applied magnetic field of 0.6 T.

4. Results and discussion

In BiGd-IG, Bi ions occupy substituted Gd sites surrounded by twelve \( \text{O}^{2-} \) ions. We have two different types of Fe atoms; \((\text{Fe})_a\) is octahedrally surrounded by eight \( \text{O}^{2-} \) ions and \((\text{Fe})_d\) is tetragonally surrounded by four \( \text{O}^{2-} \) ions. In these solids, \((\text{Fe})_a\) and \((\text{Fe})_d\) are strongly spin polarized, whereas other atoms are nonmagnetic. Magnetic moments on \((\text{Fe})_a\) and \((\text{Fe})_d\) are parallel and anti-parallel to Gd magnetic moment. We take 89 atoms (up to 6.24 Å) into account to obtain good convergence in regard to the cluster size. We refer to the electronic structure calculated by FEFF8 and estimate each magnetic moment as saturated magnetic moment.

In this paper, we only show Bi \( L_3 \) and \( L_1\)-edge XANES (X-ray absorption near edge structure)/XMCD because \( L_3\)-edge XMCD is similar to \( L_2\)-edge XMCD as explained by

\[
\Delta T_{11}(L_2) = -\Delta T_{11}(L_3).
\]

First we calculate BiGd-IG Bi \( L_3 \) and \( L_1\)-edge XANES. Figure 1 shows that the calculated spectra are in good agreement with the experimental spectra. This result confirms that the electronic structures are appropriate in both cases.

4.1. The spin polarization on Bi

We study the dependence of the spin polarization on Bi ions. Three different models (nonmagnetic, parallel/ anti-parallel magnetic moment to the Gd spin polarization) are used. Figure 2 shows the Bi \( L_3(a) \) and \( L_1(b) \)-edge XMCD spectra for different spin polarization on Bi atom compared with the experimental spectra: + (plus) and - (minus) correspond to the parallel
and anti-parallel magnetic moment. We find the spectral change in the energy region from -5 eV to 20 eV. In low energy region (<20 eV), the calculated spectra are sensitive to the spin polarization on Bi 6p orbital. This result shows that even though Bi is nonmagnetic, we can observe finite Bi XMCD. Assuming that Bi has anti-parallel magnetic moment to Gd, we obtain considerably good agreement. The agreement is, however, not sufficiently good, because we cannot fully take the random distribution into account in our calculations.

Figure 2 shows that the calculated XMCD intensity is about twenty times as high as that of the observed one. The XMCD signal is strongly influenced by temperature for these systems because each Gd spin orientation is sensitive to temperature [4,9,10,11]: In the case of Gd L2-edge in Gd-IG, the XMCD intensity at 20 K is about ten times as high as that of the one at 300 K [11]. In the case of Dy L2 and L3-edge in Dy-IG, we find the similar tendency[4]. In the present calculations, we assume that all magnetic moments on Gd are parallel as expected at T=0 K. Actually the spin orientation of Gd ions is far from this simple model.

4.2. The effect of nonmagnetic atoms around Bi

We next study the effect of the nonmagnetic atoms such as Y and Ga around Bi. For that purpose two models are considered : One is BiGd-IG, where some Gd are substituted by Bi in

Figure 1. Calculated Bi L3(a) and L1(b) -edge XANES spectra for BiGd-IG and observed ones for (BiYGd)(FeGa)O.
Gd-IG. The other is (BiYGd)(FeGa)O, where some Gd ions are substituted by Bi and Y ions, and some Fe ions by Ga ions. We calculate Bi $L_3$ and $L_1$-edge XMCD for different substituted models, and take average over them. Of course we only consider some representative models. The averaged XMCD spectra are compared with the experimental ones for (BiYGd)(FeGa)O in Figure 3. With increase of substitution ratio of Y and Ga, the XMCD spectra show large deviation from those for BiGd-IG. The calculated XMCD is sensitive to the substitution in particular near the absorption edge. Although the substitution takes place beyond the nearest neighbors, we find considerable influence on the calculated spectra.

![Figure 3](image_url)

**Figure 3.** Calculated Bi $L_3$(a) and $L_1$(b)-edge XMCD spectra for BiGd-IG and (BiYGd)(FeGa)O compared with the experimental ones for (BiYGd)(FeGa)O.

5. Conclusion
The present relativistic full multiple scattering calculations demonstrate that even though Bi ions are nonmagnetic, finite XMCD is expected because of spin-dependent exchange scatterings from surrounding magnetic ions (Gd and Fe in the present case). A simple rule, finite Bi XMCD implies spin polarization at Bi sites, can be misleading. The Bi XMCD spectra are also influenced by the substitution of Gd by Y, and of Fe by Ga although they are beyond first coordination shell of Bi ions. Unfortunately the detailed magnetic and structural information cannot be obtained because the XMCD spectra reflect local magnetic and geometric structure up to 6.2 Å.

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