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A Large Faraday Effect in Rh\(^{4+}\)-Substituted Magnetic Garnets


Department of Physics, Toho University, Funabashi, Chiba 274, Japan
* Electrotechnical Laboratory, Tsukuba, Ibaraki 305, Japan

Abstract. A large Faraday rotation is reported for Rh\(^{4+}\)-substituted magnetic garnets in the near infrared wavelength region. The Faraday rotation at 0.9 \(\mu\)m reaches at about seven times as large as that in YIG by substituting Rh\(^{4+}\) ions of only 0.13 per formula unit at room temperature. From the electronic state calculations of a (Rh\(^{4+}\)O\(^{2-}\))\(^{4-}\) octahedral cluster performed by using an SCF-Unrestricted-SW X \(\alpha\) method, the large Faraday rotation is assigned to the charge transfer transitions of an electron from O 2p to Rh 4d orbitals, or \(t_{2g}^+ (2p) \rightarrow t_{2g}^+ (4d)\) and \(t_{2g}^+ (2p) \rightarrow t_{2g}^+ (4d)\) transitions.

1. Introduction

Recently, it was reported that the substitution of Ir\(^{4+}\) (5d\(^{5}\)) ions in magnetic garnets induces a large Faraday rotation at around 0.5 \(\mu\)m, and the origin of this rotation is attributable to the charge transfer transition from O 2p to Ir 5d orbitals[1]. In this paper, we report the effect of Rh\(^{4+}\) (4d\(^{5}\)) substitution on the Faraday rotation in YIG.

2. Experiment

Single crystalline films of magnetic garnet \((Y, Ca\)^{3+}\) \((Fe, Rh\)^{4+}\) \(O\)^{12}\) were grown by using an LPE method onto (111)-oriented GGG substrates by using PbO and B\(_2\)O\(_3\) fluxes. The Ca\(^{3+}\) ions in the melts were optimized so as to substitute Fe\(^{3+}\) with Rh\(^{4+}\) in a desired amount. The compositions of the films were determined by the ICPS method by using Perkins-Emer Plasma 11 type spectrometer. Rh contents were found to be less than 0.13 per formula unit. Moreover, Ca\(^{3+}\) contents were confirmed to be nearly equal to Rh\(^{4+}\) ones in all films, indicating that Rh in the films are in the tetravalent state, though Pb and Pt were incorporated into the films as impurities from the flux and the Pt crucible used in the experiment.

Faraday rotation (FR) spectra were measured by using the spectrometer (JASCO) under the magnetic field of 4.5 kOe at room temperature. Optical absorption spectra were measured by using U-3200 type spectrometer (Hitachi) at room temperature.

3. Experimental Results

Fig. 1 shows the FR spectra of \((Y, Ca\)^{3+}\) \((Fe, Rh\)^{4+}\) \(O\)^{12}\) films measured at room temperature. From this result, it is found that 1) a large negative FR peak at around 0.9 \(\mu\)m appears with the Rh\(^{4+}\)-substitution, 2) FR increases almost linearly with the Rh content in the films, and 3) FR at 0.9 \(\mu\)m

Fig. 1 Faraday rotation spectra of \(Y_{2.4}Ca_{0.6}Fe_{0.8}Rh_{0.2}O_{12}\) films at room temperature.
in the film of only 0.13 Rh per formula unit reaches about seven times as large as that in YIG. Fig.2 shows the absorption spectra of (Y,Ca) (Fe,Rh) \( \times \) O \( \times \) films measured at room temperature. It is found that 1) the absorption strength increases with Rh content in the films, and 2) there appear strong absorptions near the positions corresponding to the FR peaks.

4. Calculation

In order to make clear the origin of the observed large FR, we calculated the electronic states of a \( \text{Rh}^{3+} \text{O}^2- \) octahedral cluster by using the SCF-Unrestricted-SW-X \( \alpha \) method[2]. The result on the one electron energy levels, the electron configuration of the ground state, and the allowed charge transfer (CT) transitions (vertical arrows) in the \( \text{Rh}^{3+} \text{O}^2- \) cluster are shown in Fig.3. As seen in this figure, Rh \( 3d^8 \) \( (4d) \) \( ^2 \) ion is in the low-spin state with one hole in the down-spin \( t_{2g} \) level, and three CT transitions to the \( t_{2g} \) level are allowed from oxygen 2p-like \( t_{2g} \) or \( t_{2g} \) level by the symmetry consideration. From the Slater transition state calculations, the transition energies of these CT transitions are found to be 1.54, 2.04 and 2.84 eV, in energy order. This means that 1) the CT transitions in the \( \text{Rh}^{3+} \text{O}^2- \) \( ^2 \) cluster appear in the near infrared wavelength region, and 2) the transition energies shift to lower energies by about 1.2 eV in comparison with those in the \( \text{Ir}^{3+} \text{O}^2- \) \( ^2 \) cluster[1]. From the transition energies, we assign the observed FR to \( t_{2g} \) \( \rightarrow \) \( t_{2g} \) (2p) \( \rightarrow \) \( t_{2g} \) (4d) and \( t_{2g} \) \( \rightarrow \) \( t_{2g} \) (2p) \( \rightarrow \) \( t_{2g} \) (4d) CT transitions.

In fact, it is possible to show that the spectral shape of the large negative FR observed in Rh \( 3d^8 \)-substituted magnetic garnets can be well reproduced by assuming the two CT transitions of \( t_{2g} \) \( \rightarrow \) \( t_{2g} \) and \( t_{2g} \) \( \rightarrow \) \( t_{2g} \), as was done in the calculation of FR spectra in Ir-substituted magnetic garnets[1]. As shown in the reference 1, it should be noted that the splitting of the ground state \( t_{2g} \) \( \rightarrow \) \( t_{2g} \) in the \( \text{Rh}^{3+} \text{O}^2- \) \( ^2 \) cluster due to the spin-orbit interaction of 4d electrons of Rh \( 3d^8 \) and the molecular field from Fe sublattices in magnetic garnets are important factors in the large FR of Rh-substituted magnetic garnets, though the CT excited states \( t_{2g} \) \( \rightarrow \) \( t_{2g} \) \( \rightarrow \) \( t_{2g} \) \( \rightarrow \) \( t_{2g} \) and \( t_{2g} \) \( \rightarrow \) \( t_{2g} \) \( \rightarrow \) \( t_{2g} \) \( \rightarrow \) \( t_{2g} \) are also split by the spin-orbit interaction of 2p electrons of O \( 2p \) and the molecular field.

5. Conclusions

In conclusion, the large FR was observed at around 0.9 \( \mu \)m in Rh \( 3d^8 \)-substituted magnetic garnets. From the electronic state calculations of the \( \text{Rh}^{3+} \text{O}^2- \) \( ^2 \) cluster by using the SCF-Unrestricted-SW-X \( \alpha \) method[2], the large FR is attributable to CT transitions of an electron from O 2p to Rh 4d orbitals or \( t_{2g} \) \( \rightarrow \) \( t_{2g} \) (2p) \( \rightarrow \) \( t_{2g} \) (4d) and \( t_{2g} \) \( \rightarrow \) \( t_{2g} \) (2p) \( \rightarrow \) \( t_{2g} \) (4d). Finally, it is noted that the effects of the spin-orbit interaction of 4d electrons of Rh \( 3d^8 \) and the molecular field from Fe sublattices in the magnetic garnet on the \( t_{2g} \) \( \rightarrow \) \( t_{2g} \) ground state of the \( \text{Rh}^{3+} \text{O}^2- \) \( ^2 \) cluster are responsible for the large FR in Rh \( 3d^8 \)-substituted magnetic garnets.

References