Structural, Optical, and Magnetic Properties of Erbium-Substituted Yttrium Iron Garnets

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ABSTRACT: We synthesized a series of slightly erbium-substituted yttrium iron garnets (Er:YIG), \( \text{Y}_{3-x}\text{Er}_x\text{Fe}_5\text{O}_{12} \) at different Er concentrations (\( x = 0, 0.01, 0.05, 0.10, \) and 0.20) using a solid-state reaction and investigated their structural, magnetic, and optical properties as a function of Er concentration. The volume of the unit cell slightly increased with Er concentration and Er atoms predominately replaced Y atoms in the dodecahedrons of YIG. The optical properties exhibited certain decreases in reflectance in the 1500−1600 nm wavelength range due to the presence of \( \text{Er}^{3+} \). Despite the many unpaired 4f electrons in \( \text{Er}^{3+} \), the total magnetic moments of Er:YIG showed similar trends with temperatures and magnetic fields above 30 K. An X-ray magnetic circular dichroism study confirmed the robust Fe 3d magnetic moments. However, the magnetic moments suddenly decreased to below 30 K with Er substitution, and the residual magnetism (\( M_R \)) and coercive field (\( H_C \)) in the magnetic hysteresis loops decreased to below 30 K with Er substitution. This implies that Er substitution in YIG has a negligible effect on magnetic properties over a wide temperature range except below 30 K where the Er 4f spins are coupled antiparallel to the majority Fe 3d spins. Our studies demonstrated that above 30 K the magnetic properties of YIG are retained even with Er substitution, which is evidence that the Er doping scheme is applicable for YIG-based magneto-optical devices in the mid-infrared regime.

I. INTRODUCTION

Magnetic garnets have been regarded as key materials for quantum computing and communication systems because of their intriguing quantum phenomena, such as hybridized magnon−phonon excitation,\(^1\) the conversion between magnons and phonons,\(^2\) spin pumping with elastic waves,\(^3\) and the spin Seebeck effect,\(^4\) which are based on a strong magnon and phonon interactions. Compared with the magnon−phonon interaction in dilute spin ensemble systems such as rare-earth ion-doped crystals\(^5\) or nitrogen-vacancy centers in diamonds,\(^6\) insulating ferrimagnet yttrium iron garnets (\( \text{Y}_3\text{Fe}_5\text{O}_{12}, \) YIG) have been widely used for various magneto-optical applications. Many researchers have made efforts to achieve a high spin density and a low damping rate, which are important factors for magnetic spintronic devices, as well as high-frequency microwave devices.\(^7\)−\(^10\)

It has been reported that transition-metal-substituted YIG exhibits a giant Faraday rotation or a magneto-optical Kerr effect, indicating that the interaction between magnetic and optical properties can be modulated by substitution or doping in the YIG system.\(^11\)−\(^13\) Rare-earth atoms substituted garnets have attracted much attention for optical applications because of their strong fluorescence emission in the mid-infrared regime.\(^14\) Er-substituted crystals exhibit a high radiative decay rate and a long optical coherent time, as well as mid-infrared emissions at a wavelength of 1.5 \( \mu \)m, which is the C-band for telecommunications.\(^15\) In recent years, the magnetic, optical, and structural properties of rare earth or transition-metal-substituted garnets have been widely investigated. Ce-substituted YIG has enhanced magnetic moments because of the preferred-(100) orientation.\(^16\) The coercive fields of magnetic garnets have been studied by changing the grain sizes.\(^17\),\(^18\) However, many garnets with substitutional atoms have shown degraded magnetic and optical properties due to changes of sample properties such as crystallinity, shape, size, and impurities.\(^19\)−\(^20\) Therefore, the search for appropriate substitutional atoms to enhance the magnetic properties as well as optical properties is highly demanded.

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Here, we synthesized a series of Er-substituted YIG, which had the chemical compositions of $Y_{1-x}Er_xFe_3O_4\downarrow$ at different Er concentrations ($x = 0, 0.01, 0.05, 0.10$, and $0.20$) using a solid-state reaction method, and studied their changed magnetic and optical properties with small concentrations of Er atoms. Fe L$_{2,3}$-XAS and XMCD spectra showed that the Er atoms seemed to replace Y atoms in the dodecahedrons of YIG. We found that the optical properties were almost invariant under Er to replace Y atoms in the dodecahedrons of YIG. We found transition lines of the Er$_{12}$ moments of Fe 3d spins. Hard XAS at the Er L$_3$-edge was conducted at the 8C beamline at the PLS. The extended X-ray absorption fine structure (EXAFS) analysis was processed using the UWXAFS package.  

III. RESULTS AND DISCUSSION

Figure 1 shows a schematic of Er-substituted Y$_{1-x}Fe_3O_4\downarrow$ (Er:YIG) with a cubic structure (space group Ia-3d). One unit cell contains Y$_{24}Fe_{40}O_{96}$ as shown in the inset of Figure 1. Since Y atoms with a trivalent state (Y$^{3+}$) have no magnetic moment, the net magnetic moment of YIG is determined by two different Fe$^{3+}$ atoms, which align antiferromagnetically along the (110) crystal direction of the YIG. For convenience, the Fe atoms in the octahedral and tetrahedral coordination are designated as Fe$_O$ and Fe$_T$, respectively. Fe$_O$ and Fe$_T$ have opposite spin directions, resulting in an expected magnetic moment of $S = 4/2 = 2$ at 0 K, which is consistent with the experimental value of 4.73 $\mu_B$/YIG. Since Er atoms have large magnetic moments and strong emission at around 1550 nm, the magnetic and optical properties of YIG might be modified by Er substitution. We synthesized polycrystalline Er-substituted YIG powders with chemical compositions of $Y_{1-x}Er_xFe_3O_4\downarrow$ ($x = 0, 0.05, 0.10, 0.15$, and $0.20$) and characterized their magnetic and optical properties as functions of the Er concentration. To prevent the unexpected formation of secondary phases, we investigated Er-substituted YIG with only a low Er concentration ($x \leq 0.20$). The actual Er concentrations were determined by ICP-OES measurements and they well matched the nominal Er concentrations of Er:YIG (see Table 1).

To obtain a solid solution of Er-substituted YIG (referred to as Er:YIG, hereafter) with a single phase, Er:YIG was sintered three times at a high temperature above 1300 °C with subsequent inter-grinding, and their crystallographic properties

Table 1. Nominal and Measured Er Concentrations ($x$) of Er:YIG

| nominal x | measured x |
|-----------|-----------|
| $x = 0$   | $x = 0.0001$ |
| $x = 0.01$| $x = 0.0088$ |
| $x = 0.05$| $x = 0.0498$ |
| $x = 0.10$| $x = 0.1003$ |
were examined using X-ray diffraction, as shown in Figure 2. The as-synthesized Er:YIG powder had a cubic structure, and the lattice constants slightly increased with increasing Er content from \( x = 0 \) to 0.20. Considering the differences in ionic radii, 0.63 Å (Fe\(^{3+}\)), 1.019 Å (Y\(^{3+}\)), and 1.144 Å (Er\(^{3+}\)), the small increase in the lattice constants of Er:YIG can be explained by Er\(^{3+}\) substitution at the Y\(^{3+}\) sites in the YIG system.

To study the optical properties of Er:YIG, we conducted FT-IR, UV–VIS, and near IR spectroscopy analyses with \( Y_{3-x}Er_xFe_5O_{12} \) powder \( (x = 0, 0.01, 0.05, \) and 0.10). The transmittance measurement of \( Y_{3-x}Er_xFe_5O_{12} \) in the region of wavenumbers from 400 to 1800 cm\(^{-1}\) showed that YIG has strong adsorption at around 450–750 cm\(^{-1}\). Figure 3a shows the FT-IR spectrum of \( Y_{3-x}Er_xFe_5O_{12} \) with the Er content \( (x = 0, 0.01, 0.05, \) and 0.10) in the region of wavenumbers from 450 to 750 cm\(^{-1}\). There were three intense absorption bands, located at around 543, 581, and 646 cm\(^{-1}\), which can be assigned to the asymmetrical stretching vibration of a Fe\(^{3+}\)–O tetrahedral in YIG.\(^{24,25}\) McDevitt et al. reported that the absorption bands in garnets could be shifted linearly with the ionic radius of rare-earth atoms.\(^{23}\)

In the case of Er:YIG, the absorption bands \( k_1 \) and \( k_3 \) of the Fe–O tetrahedra were red-shifted while \( k_2 \) was slightly blue-shifted by increasing Er concentrations (see the trend curves of \( k_1, k_2, \) and \( k_3 \) in the inset of Figure 3a). Considering the similarity in the ionic radius of Er\(^{3+}\) and Y\(^{3+}\), the red and blue shifts in the adsorption bands might be caused by the local environment changes of the Fe–O tetrahedra, as Er atoms are substituted into Y atomic sites in the YIG system.\(^{23}\)

We also studied the optical properties of Er:YIG in the UV–VIS–NIR range. Figure 3b shows that the Er:YIG has very low reflectance in the VIS range (300–700 nm), and a steep increase in reflectance was observed at around 700 nm. Note that high reflectance exceeding 80% was observed above a wavelength of 1200 nm, which is consistent with the reported high transmittance of YIG in this wavelength range. In Er:YIG with \( x = 0.05 \) and 0.10, there are small dips in the wavelengths range of 1450–1550 nm, which correspond to the optical transition between I\(_{15/2}\) and I\(_{13/2}\) in Er\(^{3+}\) ions (see the inset figure in Figure 3b).

We checked the optical bandgaps of Er:YIG using the Tauc plot method, which has been used to estimate the optical bandgap in many semiconducting systems.\(^{26,27}\) According to the calculated band structures of YIG, YIG with a cubic structure \((Ia-3d)\) exhibits a direct bandgap.\(^{29,30}\) Therefore, we plotted \((\alpha h\nu)^{1/2}\) as a function of the photon energy as shown in Figure 3c. The optical bandgaps deduced from the Tauc plot were around 1.69–1.70 eV over all of the Er concentrations.

Figure 2. Rietveld refinements of the X-Ray diffraction data of \( Y_{3-x}Er_xFe_5O_{12} \) with the Er content \( (x = 0, 0.01, 0.05, 0.10, 0.15, \) and 0.20). The experimental and simulated X-ray diffraction are depicted as red dots and black lines, respectively. The difference between the experimental and simulated X-ray diffraction is depicted as a blue line. X-ray diffraction patterns well match the reference peaks of YIG (green bars, space group Ia-3d) with a low Er content. In \( x = 0.20 \), X-ray diffraction patterns are refined with YIG (upper green bars, space group Ia-3d) and YFeO\(_3\) (lower green bars, space group Pnma).
indicating that the optical bandgaps are almost invariant with Er substitution in a slightly Er-substituted YIG system.

The magnetization of YIG originates from the antiferromagnetically coupled 3d spins of Fe\textsuperscript{T} and Fe\textsuperscript{O}. Since Er atoms have a large magnetic moment, exceeding 9.6 \(\mu_B/\text{Er atom}\), Er substitution at the Y sites can change the magnetic moments of the YIG system. The temperature \(T\)-dependence of magnetization \(M\) in Er:YIG under a magnetic field of 0.05 T shows that \(\text{Y}_{3-x}\text{Er}_x\text{Fe}_5\text{O}_{12}\) has ferromagnetic behavior as shown in Figure 5a. The room temperature magnetic moments of \(\text{Y}_{3-x}\text{Er}_x\text{Fe}_5\text{O}_{12}\) with \(x = 0, 0.01, 0.05, \text{and} 0.10\) were \(\sim 3.0\) to \(3.3\) \(\mu_B\) per one formula unit. The temperature-dependent magnetic moments of Er:YIG showed that the magnetic moments were saturated or slightly decreased with a decrease in temperature below 100 K. Interestingly, antiferromagnet-like behaviors (dips in magnetization) appeared at low temperatures (<30 K).

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**Figure 3.** (a) FT-IR spectrum of \(\text{Y}_{3-x}\text{Er}_x\text{Fe}_5\text{O}_{12}\) with the Er content \((x = 0, 0.01, 0.05, \text{and} 0.10)\) with three absorption bands, \(k_1, k_2, \text{and} k_3\). The inset shows the shifts of \(k_1, k_2, \text{and} k_3\) as a function of Er contents \((x)\). (b) UV–VIS–NIR spectrum of \(\text{Y}_{3-x}\text{Er}_x\text{Fe}_5\text{O}_{12}\) with the Er content \((x = 0, 0.01, 0.05, \text{and} 0.10)\). The inset shows the optical transition of \(\text{Er}^{3+}\) in a wavelength range of 1450–1550 nm. (c) Tauc plot of \(\text{Y}_{3-x}\text{Er}_x\text{Fe}_5\text{O}_{12}\) with the Er content \((x = 0, 0.01, 0.05, \text{and} 0.10)\) where \(\alpha\) is the absorbance and \(h\nu\) is an incident photon energy.

**Figure 4.** (a) Temperature-dependent magnetic moment of Er:YIG \((x = 0, 0.01, 0.05, 0.10, \text{and} 0.20)\) from 2 to 300 K. (b) DSC measurements of \(\text{Y}_{3-x}\text{Er}_x\text{Fe}_5\text{O}_{12}\) with Er content \((x = 0, 0.05, 0.10, \text{and} 0.20)\) with a changing temperature between 300 and 700 K upon heating and cooling.
for all of the samples, except for the undoped YIG, which is likely due to the ordering of Er 4f spins in the low-temperature range. The contribution of the Er 4f spins should be dominated by the interactions between Er and the YIG lattice rather than a direct Er–Er interaction because Er 4f orbitals should be much more localized than Fe 3d orbitals and furthermore much farther apart from each other (∼3.8 Å) compared to Er–FeT (∼3.10 Å) or Er–FeO (∼3.46 Å). The onset temperatures of the dips (referred to as $T_{Er}$) increased and the amount of decrease in the magnetic moment below $T_{Er}$ increased with increasing Er concentration, which confirms the conclusion that the dips originated from the weak Er 4f spin ordering. Nevertheless, the presence of Er atoms did not significantly alter the magnetic moments at temperatures above $T_{Er}$ (<30 K), clearly showing that Er 4f spins had a negligible impact on room temperature magnetism.

We estimated the Curie temperatures ($T_C$) where the ferromagnetic ordering disappeared in Er:YIG using DSC measurements. Upon cooling and heating Er:YIG, an exothermic and endothermic transition occurs at certain temperatures, which are referred to as Curie temperatures ($T_C$) of Er:YIG. Figure 4 shows the changes of $T_C$ during heating and cooling and are marked with black dashed lines; $T_C$ decreases by roughly 10 K with increasing content of Er from $x = 0$ to 0.20. Considering a reported $T_C$ of YIG at around 560 K, the $T_C$ of the Er:YIG slightly decreases by substituting Er into YIG.

The magnetic field ($H$) dependence of the magnetization of Er:YIG was measured at various temperatures. Figure 5a shows that the magnetic moments of $Y_{3-x}Er_xFe_5O_{12}$ were saturated for $x = 0, 0.01, 0.05, 0.10$, and 0.20, clearly showing that Er 4f spins had a negligible impact on room temperature magnetism.

Figure 6. (a) $M (H)$ loops for Er:YIG measured at 2 and 300 K. (b) The coercive force and residual magnetization of Er:YIG as a function of the Er content.
above 0.1 T at a value of around 4.0 $\mu_B$/YIG, which is similar to the theoretically predicted net magnetic moment of YIG ($5.0 \mu_B$/YIG). In a temperature range between 2 and 300 K, the saturated magnetic moments of Y$_{1-x}$Er$_x$FeO$_{12}$ under a high magnetic field changed slightly with temperature and Er concentrations; however, there were no significant changes in the magnetic properties. The magnetic hysteresis curves of Y$_{1-x}$Er$_x$FeO$_{12}$ ($x = 0.01, 0.05,$ and $0.10$) taken at 2 and 300 K were compared as shown in Figure 6a. The magnetic hysteresis curves of Y$_{1-x}$Er$_x$FeO$_{12}$ were retained up to room temperature with the coercive field ($H_C$) and residual magnetization ($M_R$) of Y$_{1-x}$Er$_x$FeO$_{12}$. The changes in $H_C$ and $M_R$ are explained by impulsive spin ordering at the surface of the Er:YIG powder or the formation of secondary phases. The XAS lineshapes reflect details of the Fe O coordination, such as its symmetry and the strength of orbital hybridizations. The spectra are very similar to each other. This suggests identical coordination symmetry of the Fe ions regardless of the Er concentration. The presence of a secondary phase, o-YFeO$_3$ (see Figure 2), might also contribute to the XAS data. However, no significant changes in the lineshapes were observed, even for the Er-rich sample ($x = 0.2$).

The XMCD spectra are displayed in Figure 7b. The intensities of the dips and peaks in the five spectra are almost the same, again indicating negligible evolution of the Fe magnetism with increasing Er concentration. Since o-YFeO$_3$ is almost antiferromagnetic and Er$_3$O$_5$ has no Fe ions, the XMCD spectra may be solely due to the local structure and magnetism of the Er:YIG. This indicates that the structure and magnetism in Er:YIG is robust with the inclusion of Er up to 0.2. Overall, the XMCD lineshapes reflect details of the Fe–O coordination, such as its symmetry and the strength of orbital hybridizations. The spectra are very similar to each other. This suggests identical coordination symmetry of the Fe ions regardless of the Er concentration. The presence of a secondary phase, o-YFeO$_3$ (see Figure 2), might also contribute to the XAS data. However, no significant changes in the lineshapes were observed, even for the Er-rich sample ($x = 0.2$).

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The results of the Fe 2p–3d atomic multiplet calculations, which have taken into account the configuration interactions under either the O₈ or T₄O ligand field, successfully reproduce the experimental data of YIG.46

Thus, each of the peaks and dips in Figure 7b can be due to the signatures of Fe³⁺–O₈ (Fe₃O) and Fe³⁺–T₄ (Fe₄) for the two peaks at ∼707 and ∼710 eV, and Fe³⁺–T₄ for the dips at ∼709 eV in the L₃-edge region. The values of FeO at the L₃-edge are overall positive (peaks) while those of Fe₄ are overall negative (dips). This is because the spins of the FeO and Fe₄ sites tend to align antiferromagnetically to each other due to super-exchange interactions. The sum of the two XMCDs reproduces the experimental data of Er:YIG, as well as the YIG crystal. Therefore, this confirms that the ratios of the Fe³⁺ ions, Fe₄/FeO = 3/2, are indeed maintained in Er:YIG.

To elucidate the Y replacement by Er, we conducted Er L₃-edge XAS for Er:YIG with x = 0.2 as a representative. Figure 8 shows the Fourier-transform (FT) magnitudes of the extended X-ray absorption fine structures (EXAFS) of the x = 0.2 sample and a reference Er₂O₃ powder. The FT was processed on k²-weighted (k: electron momentum) EXAFS in a range of 2–10 Å⁻¹ using a Hanning window.33 The FT EXAFS magnitudes show the bonding distributions of Er ions in both samples. The overall FT spectra are very different from each other, suggesting that the local environment of Er in the x = 0.2 sample is fundamentally different from that of Er₂O₃. This confirms that the presence of Er₂O₃ in Er:YIG samples is negligible.

The major peaks were assigned according to the phase uncorrected atomic distance, R, as Er–O, Er–Fe₄, and Er–FeO for the x = 0.2 sample, and as Er–O and Er–Er for Er₂O₃. The roughly estimated bond lengths (R + Δ) after the scattering phase corrections (Δ) were ∼2.2 to 2.4 Å (Er–O), 2.84 Å (Er–Fe₄), and 3.21 Å (Er–FeO), a bit reduced from the bond lengths of 2.37–2.43 Å (Y–O), 3.09 Å (Y–Fe₄), and 3.46 Å (Y–FeO) in pristine YIG.34 The reduction in bond lengths (by 0.1–0.2 Å) appears contradictory to the slightly different size (radius) of Er³⁺ ions (1.144 Å) compared to Y³⁺ ions (1.019 Å). Perhaps, certain local lattice shrinkage is accompanied near Er³⁺ in substituted YIG. We also considered how Er substitution influences three absorption bands in the FT-IR spectra, which correspond to the vibration of the FeO−O tetrahedra. However, the composition dependence of the absorption wavenumbers is rather weak, implying the structural shrinkage occurs only locally.

Meanwhile, the average valence of the Er ions was the same as in Er₂O₃, which was confirmed because the first peaks had the same energy (highlighted by the dashed vertical line) in the X-ray absorption near-edge structures (XANES) displayed in the inset. The overall similarity in the Er local structure (EXAFS) as well as in the average valence of Er (XANES) strongly suggests that the Er³⁺ ions indeed occupy the Y sites in the YIG structure. This is consistent with the finding of robustness of the local environment (and thereby the magnetism) of the Fe ions in Figure 7.

The Er³⁺ ions substituting the Y sites can have limited influence on the magnetism in YIG because the magnetism is dominated by the spin interactions of the relatively dispersed Fe 3d orbital states, not the localized Er 4f orbital state. If Er³⁺ occupied Fe₄ or FeO sites instead, the Fe⁴⁺ ions leaching from Er:YIG should have constituted a different Fe compound with a distinct local structure (as well as magnetism) from YIG. This is contradictory to the findings of XAS and XMCD in Figure 7, where there are no noticeable changes in the lineshapes observed upon Er doping. Therefore, we concluded that Er doping indeed occurred in YIG (apart from the formation of YFeO₃), replacing the Y ions, while the electronic or spin configurations of the Fe ions hardly changed.

Therefore, the decrease in magnetic moments with an increasing Er concentration cannot account for the degradation in Fe 3d ferrimagnetism in YIG. The temperature-dependent magnetization data in Figure 5a show that the magnetic moment decreases gradually as the temperature decreases, even at temperatures above Tₐ, in the low-temperature regime (<150 K). Although the Er 4f spins may contribute to the magnetic moments at low temperatures, as in the case of Er₂O₃,42 the Er 4f electron states should be highly localized in contrast to the delocalized Fe 3d states, so as to be less relevant to the spin transfer, which is the key ingredient in spin devices. This indicates that the loss in spin-transfer functionality induced by the Er substitution would not be substantial.

On the other hand, there is a small but clear evolution in the optical properties (reflectance in the wavelength range of 1450–1550 nm in Figure 3b) due to optical transitions within the Er³⁺ 4f states. Absorption will lead to radiation in the C-band, which is useful for telecommunications. Thus, adding Er offers a promising way of improving optical properties while preserving the magnetic properties in YIG. Further studies are needed to optimize the efficiency of the dopant-related radiation to use YIG-based materials in magneto-optical devices.

IV. CONCLUSIONS

YₓErₓFe₂O₅ was successfully synthesized with various Er concentrations up to x = 0.20 using a solid-state reaction method, and the effects of the Er substitution on the magnetic and optical properties of YIG were scrutinized using various techniques. The Er atoms replacing the Y sites in YIG seemed to modify the optical properties, whereas the magnetic properties were barely modified by the Er substitution except for the contribution of Er 4f spins in the low-temperature regime (Tₐ < 30 K). The magnetism near room temperature was retained even for the Er-substituted YIG, suggesting that rare-earth ion doping is an effective way of controlling
optoelectronic properties in the mid-infrared range, which is useful for magneto-optical device applications.

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**Notes**

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