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Magnetic Properties of Gd-Al-Substituted Garnet films for Microwave Devices

Nobuyasu Adachi, Yutaka Hayakwa, Takashi Okuda, Manabu Gomi
Nagoya Institute of Technology, Gokiso-cho, Showa-ku, Nagoya, 466-8555 Japan
Fax: 81-52-735-5290, e-mail: nadachi@mse.nitech.ac.jp

The magnetic properties of Gd-Al-substituted yttrium iron garnet films were investigated for microwave devices. It is well known that yttrium iron garnet is useful for low energy loss for the microwave magnetic devices, however, the magnetization change depends on temperature and induces the instability of the resonance frequencies for the case of thin films. The stability of the magnetization can be achieved by utilizing the compensating temperature dependence of rare earth magnetic garnets. We prepared Gd-Al-substituted magnetic garnet films by liquid phase epitaxy (LPE). The LPE single crystal thin film has advantages such as rapid growth with homogeneous compositions and good quality for magneto-static wave devices. The difficulty is the composition design to control both the compensation (Curie) temperature and the small mismatch between the film and the substrate. As a result of optimum growth conditions, the $Y_2Gd_1Fe_{4.65}Al_{0.35}O_{12}$ film showed almost zero mismatch to the substrate and small change of magnetization near room temperature.

Key words: magnetic garnet, microwave device, LPE, thin film

1. INTRODUCTION

Recently, magnetic rare earth garnet materials are becoming important again for microwave devices such as magnetostatic-wave (MSW) filter or MSW signal to noise ratio (S/N) enhancer with increment of demand of microwave with higher frequency region in mobile wireless communications [1-3]. The study of magnetic garnet materials had been already started for microwave devices in 1960's [4]. In 1970' and 1980', lots of work were mainly achieved for the application of magnetic bubble memory [5]. During this period, the liquid phase epitaxy (LPE) technique was developed in order to synthesize thin films. At the present stage, this technique is utilized for the application of optical isolator [6] in optical communication system and MO indicator which visualize magnetic flux as a magnetic sensor [7].

In microwave magnetic devices, it is well known that yttrium iron garnet (YIG) indicates good microwave properties in X band region because of remarkable low energy loss. However, due to the temperature change of the magnetic properties, instability of the resonance frequency occurs in the available temperature range of −10 ℃ to 50 ℃. In order to realize the stability of the resonance frequency, an idea to control the compensation point of the rare earth magnetic garnets had been proposed before 1970's [8,9]. By adjusting the compensation point and Curie temperature, it is possible to realize a flat area in the magnetization curve in the temperature range of −10 ℃ to 50 ℃. In comparison with the application of bulk materials, thin film devices have been recently improved because of the development of thin film technology. The LPE technique has an advantage for good quality films with homogeneous composition, which is necessary to completely compensate the magnetic moment of irons with the magnetic moment of rare-earth ions. The difficulty for the synthesis lies in the composition design where the lattice mismatch between the film and the substrate becomes almost zero. We report the synthesis of Gd- Al- substituted film prepared by LPE technique and optimizing process in the growth on $Gd_3Gsa_5O_{12}$ (GGG) substrate. Among rare-earth ions, Gd substitutions is considered to be suitable for adjusting a flat area of the magnetization curve near room temperature. Nonmagnetic Al ions dilute the Fe ions, which decreases the Curie temperature (T_c).

2. EXPERIMENTAL

$Y_{3-x}Gd_xFe_{5-y}Al_yO_{12}$ garnet films were prepared by LPE technique. $PbO-B_2O_3$ was used as the flux melt in a Pt crucible. The single crystal films were epitaxially grown on (111) GGG substrates (lattice constant a_s=1.238 nm). The melt composition was calculated by using *R* parameters [8] given by

$$R_1 = \frac{Fe_2O_3}{Gd_2O_3 + Y_2O_3},\ R_2 = \frac{Fe_2O_3}{Al_2O_3}, R_3 = \frac{Pb_2O_3}{B_2O_3},$$
$$R_4 = \frac{Fe_2O_3 + Al_2O_3Gd_2O_3 + Y_2O_3}{Fe_2O_3 + Al_2O_3Gd_2O_3 + Y_2O + Pb_2O + B_2O_3},\quad (1)$$
$$R_3 = \frac{Gd_2O_3}{Y_2O_3},$$

in which R_1, R_3, R_4 and R_5 were fixed to be 14, 15.6, 0.1 and 2.33, respectively. R_3 was varied from 32 to 14 for the Al substituted film. The films prepared by vertical dipping mode were used in order to determine the saturation temperature of the melt and the films prepared by horizontal dipping mode were used for characterization of magnetic properties. The growth temperature was kept between 920 ℃ and 960 ℃, where the supercooling temperature was in the vicinity of 50 ℃to 70 ℃. During the film growth, the substrate was rotated at 100 rpm. The magnetization of the films were measured by the vibrating sample magnetometer (TOEI VSM-5) in the temperature range from –193 ℃ to 300 ℃. Ferromagnetic resonance (FMR) measurements were performed in X-band region by conventional ESR magnetometer (JEOL FE1XG) at room temperature. The lattice mismatch between the film and substrate was measured by the x-ray diffractometer with Cu and the compositions of the film were determined by EDX analysis.

3. Results and discussion

The substitution of Al ions decreased the T_c and increases the compensation point. Taking it into consideration the magnetic properties of bulk materials of $Y_{3-x}Gd_xFe_{5-y}Al_yO_{12}$, we investigate $Y_2Gd_1Fe_{5-y}Al_yO_{12}$ in detail in order to adjust the flat area of magnetization curve near room temperature. Without Al ions, the compensation point lies below –200 ℃. The growth conditions and characterization of the films are shown in Table I. The substitution of Al was varied from y=0 to y=0.7 in this experiment. The mismatch between the film and the substrate of the specimen B was smallest among the films, where the composition was y=0.35. The temperature dependence of the saturation magnetization is shown in Fig. 1. With increasing the content of Al ions, the T_c shifts to lower temperature and the T_c of the specimen E is approximately 150 ℃. On the contrary, the compensation point shifts to higher temperature and that of the specimen E is approximately –100 ℃. The saturation magnetization remarkably decreased with substitution of Al ions. Without Al substitution, the saturation magnetization at room temperature shows $4\pi M_s$ value of approximately 1000 gauss, whereas the specimen E with y=0.7 shows approximately 160 gauss, which is one-fifth as large as the value of the specimen A. The reason comes from the selective substitution of Al ions with Fe ions on tetrahedral sites. The net magnetization of a magnetic garnet is due to the tetrahedral Fe ions.

Table 1. Growth Condition and Characterizations of $Y_2Gd_1Fe_{5-x}Al_xO_{12}$.

	A	B	C	D	E
Saturation Temp. (℃)	984	996	1004	1015	1025
Growth Temp. (℃)	928	932	946	945	956
Supercooling (℃)	56	64	58	70	69
Dipping Time (min)	5	2	2	1.5	1.5
Rotation (rpm)	100	100	100	100	100
Thickness (mm)	13.1	6.7	6.1	5.3	5.1
a_f (×10 nm)	12.411	12.386	12.370	12.360	12.362
Δa (×10^{-2} nm)	2.8	0.3	-1.3	-2.3	-2.1
x (mol)	0	0.35	0.46	0.54	0.72

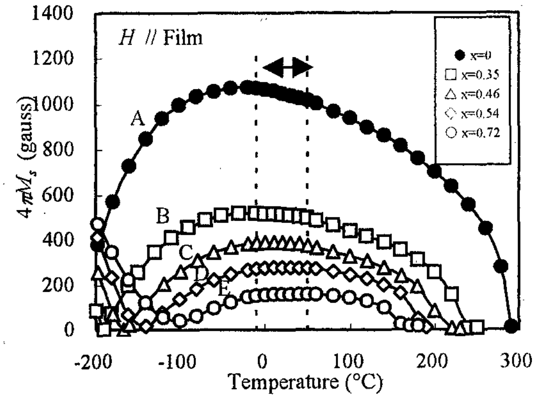

Fig.1 Temperature dependence of saturation magnetization for $Y_2Gd_1Fe_{5-x}Al_xO_{12}$

The tip of the magnetization curve between the compensation point and the T_c shifts to room temperature and the change of the saturation magnetization $\Delta(4\pi M_s)$ in the temperature range of –10 ℃ to 50 ℃ decrease with increment of Al substitution. As shown in Table II, the smallest value of $\Delta(4\pi M_s)$ is 6 gauss where the flat area is almost realized in magnetization curve.

Table 2 Magnetic Properties of $Y_2Gd_1Fe_{5-x}Al_xO_{12}$.

	A	B	C	D	E
$(4\pi Ms)_{max}$ (gauss)	1073	517	390	278	157
$\Delta(4\pi Ms)$ (gauss)	53	21	15	6	6
ΔH (Oe)	232	77	111	153	133

The FMR spectra of the specimens are shown in Figure 2. The magnetic field was applied perpendicular to the film plane at room temperature. With substitutions of Al ions, the resonance field shifts to lower field. The sharp resonance signals were observed in the specimens A and B. With increment of Al content, the intensity of the resonance signal becomes weak and spurious signals appear. The decrease of the resonance field and signal intensity comes from the reduced magnetization and magnetic interaction due to the dilution of Fe ions. One of the possible reasons of the spurious mode observed in the specimen E is the shape effect. The specimens were cut into approximately 5×5 mm rectangular shapes. The demagnetization effect of the edge area of the specimen causes the inhomogeneous resonance field.

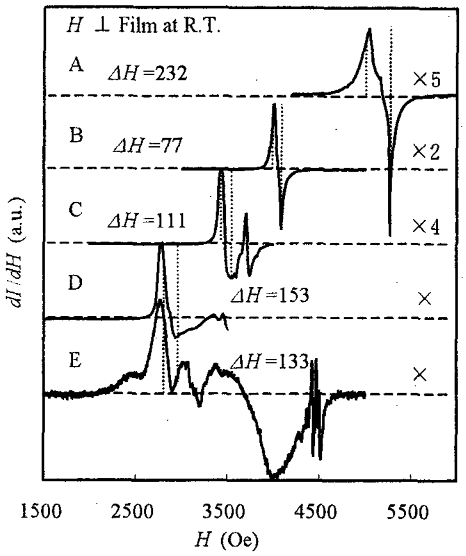

Fig.2. FMR spectra of $Y_2Gd_1Fe_{5-x}Al_xO_{12}$ at X band region in the magnetic field perpendicular to the film plane.

With decreasing intensity of the main resonance signal, the spurious mode probably appears.

The ferromagnetic resonance frequency ω in the applied magnetic field is

$$\omega^2 = \gamma^2\,[H_{res} + (N_x - N_z)\times 4\pi M_s]\times [H_{res} + (N_y - N_z)\times 4\pi M_s], \qquad (2)$$

where, γ =2.8 MHz/Oe is the gyromagnetic ratio, H_{res} is the resonance field, N_x, N_y, and N_z are the demagnetization factors, M_s is the saturation magnetization [9]. If the specimen is a sphere, the resonance frequency is independent of the saturation magnetization because $N_z = N_x = N_y = 1/3$. For the case of thin films, the demagnetization factors can be approximated to be N_z =1, N_x = N_y =0, then the resonance frequency is written

$$\omega = \gamma(H_{res} - 4\pi M_s), \qquad (3)$$

which depends on the saturation magnetization. The change of the saturation magnetization induces the instability of the resonance frequency. From the point of view of stability of the resonance frequency, the specimen D and E are better than others because of the smaller $\Delta(4\pi M_s)$. However, the resonance linewidths ΔH of the specimen D and E are more than 100 Oe (see Table II). At the present stage, the smallest value of ΔH is 77 Oe which was obtained in the specimen B.

The equation of motion of ferromagnetic moment including damping term is given by

$$\begin{aligned}\mathbf{M} &= \gamma\,(\mathbf{M}\times\mathbf{H}) - \frac{\alpha}{\mathbf{M}}(\mathbf{M}\times\mathbf{M}) \\ &= \gamma\,\mathbf{M}\times\left(\mathbf{H} - \frac{\alpha}{\gamma\mathbf{M}}\mathbf{M}\right),\end{aligned} \qquad (4)$$

where α is a measure of the strength of the damping [10]. The α is described using the FMR linewidth as

$$\alpha = \frac{\gamma\,\Delta H}{2\omega}. \qquad (5)$$

From the point of view of low loss by damping term, the specimen B is better than the others. There are several factors which induce the broadening of the resonance width. Tanno et. al. reported that the ΔH of Ga substituted YIG showed a remarkable increment when the $4\pi M_s$ was less than 500 gauss [11]. It is also well known that the stress due to the mismatch between the film and the substrate increases ΔH. In the results of our experiment, the value of $4\pi M_s$ of the specimen B is approximately 500 gauss and the mismatch Δa is almost zero. These properties are expected to reduce the value of ΔH.

In order to realize the small $\Delta 4\pi M_s$ and ΔH simultaneously, one of the possible ideas is the selective substitution with the Fe ions on octahedral ions which prevent from the reduction of saturation magnetization. The substitution sites depend on the kinds of ions [12]. Further investigations are necessary.

3. Summary

The magnetic garnet $Y_{3-x}Gd_xFe_{5-y}Al_yO_{12}$ films were grown on (111) GGG by LPE technique. The substitution of Gd ions has a roll of generating a compensation point and Al substitution has a roll of shifting the temperature of both compensation point and T_c. Under the condition of x=1, the flat area of the magnetization curve was adjusted in the temperature range between −10 ℃ to 50 ℃ by optimizing the content of Al ions to be y=0.54 or 0.72. On the other hand, the smallest FMR linewidth was obtained in the condition of y=0.35. In order to realize both the temperature stability resonance frequency and the low energy loss, it was revealed that the values of M_s and ΔM_s should be optimized simultaneously.

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