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Garnet composite films with Au particles fabricated by repetitive formation for enhancement of Faraday effect

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Abstract
To prepare garnet (Bi : YIG) composite films with Au particles, we used a repetitive formation method to increase the number density of particles. On increasing the number of repetitions, the diameter distribution of the particles changed. After five repetitions using 5 nm Au films, the diameter distribution separated into two size groups. Shift of wavelength-excited localized surface plasmon resonance is discussed relative to the diameter distribution. In the composite films, enhancement of Faraday rotation associated with surface plasmons was observed. With six repetitions, a maximum enhanced rotation of -1.2° was obtained, which is 20 times larger than that of a single Bi : YIG film. The figures of merit for the composite films are discussed. The thickness of a Bi : YIG composite film working for enhanced Faraday rotation was examined using an ion milling method.

1. Introduction

Magneto-optical (MO) effects, that is, Faraday and Kerr effects, have been used for an optical isolator in optical communication and an MO disk in MO recording. A bismuth-substituted yttrium iron garnet (Bi : YIG) [1, 2] is an MO material which is transparent in the visible and infrared light region, on which fundamental and applied studies have been extensively performed. In a recent applied study for a high-speed optical switching device, an MO spatial light modulator with garnet pixels and small driving coils for changing the direction of magnetization of the respective pixels was fabricated [3]. However, since it is difficult to form structured pixels of thick garnet crystals on a substrate, a material is required to have a large MO rotation angle even in a thin film in order to obtain a large optical contrast. Thus, if MO materials with a large MO effect can be obtained, development of new thin film devices will be facilitated.

Surface plasmon resonance is one of the candidates used to enhance MO effects, which is an attractive method to obtain larger MO effects by introducing a nanostructure. In a noble metal particle, localized surface plasmon resonance (LSPR) is excited by light illumination; a strong near-field is generated around the particle. When the particle is surrounded by an MO material, it is expected to have an influence on the MO response. In the past, studies on the MO effect associated with plasmons have been performed. The relationship between bulk plasma and the Kerr effect was discussed by Feil and Haas in 1987 [4], and the experimental study for enhanced Kerr rotation in Fe thin films on Cu substrates by bulk plasma was reported by Katayama *et al* in 1988 [5]. With respect to MO responses associated with surface plasmon resonance, a theoretical study on the MO effect of non-magnetic particles surrounding a magnetic medium [6], a study on the MO effect for a nano-onion with multicore–shell structure [7] and a study on the change in Kerr effect of Au/YIG granular films [8] were reported. Also as an interesting phenomenon of MO response, anomalous Faraday effect near a wavelength of extraordinary transmittance in a thick metal film with MO material filled holes, was theoretically investigated [9]. Our study for MO response enhanced by LSPR started from a result of small enhancement of Faraday rotation of 0.03° in Au particles formed on a Bi : YIG films [10]. Then, with Au particles embedded in Bi : YIG thin films on quartz substrates [11] large

enhancement of Faraday rotation was observed. Later the use of Au–Ag alloy particles in composite films was attempted to shift the LSPR wavelength for enhanced Faraday rotation by changing the composition ratio [12].

In this paper, we describe an optimal structure of the Bi : YIG composite film with Au particles to have large enhancement of Faraday rotation associated with LSPR. The Au particles were fabricated by a repetitive formation method to change the Au total volume, number density and diameter distribution. We think that such a preparation of Au particles is a useful technique for other studies such as plasmon sensors, devices and so on. Optical and MO responses of the Bi : YIG composite films having Au particles with exciting LSPR are discussed.

2. Experimental

We prepared Au particles by a repetitive formation method. First, an Au thin film with a thickness of 5 nm was deposited on a fused quartz substrate using a sputtering system. The Au film was heated at a temperature of 1000 °C in an electric furnace in air for 10 min. After this first particle formation, the second Au film was deposited on the existing particles and then it was heated again. This procedure of deposition and heating was repeated until the planned number of repetitions was reached. Finally, a Bi : YIG film with a thickness of 60 nm was deposited onto the Au particles on the substrate by an RF magnetron sputtering system (Shimadzu, HSR-551S) using a $Bi_1Y_{2.5}Fe_5O_X$, target; it was heated at 750 °C for 1 h to crystallize the garnet.

To prepare some Bi : YIG composite films with rather even film surfaces, used for experiments of thickness dependence, a sol–gel method was used for the formation of Bi : YIG before the deposition by sputtering method. A gel with a composition of $Bi_{0.5}Y_{2.5}Fe_5O_{12}$ involving stoichiometric amounts of constituent metal ions [13] was coated on the Au particles on a quartz substrate by a spin-coater. The coated gel was dried at 50 °C, pre-heated at 400 °C for 20 min and heated at 750 °C for 20 min.

A field-emission scanning electron microscope (FE-SEM, JEOL, JSM-6700) was used to observe the fabricated structures of the Au particles. Transmission spectra and MO Faraday rotation were measured using a spectrophotometer (Shimadzu, UV-3150) and a MO measurement system (Neoark, BH-M600VIR-FKR-TU), respectively.

3. Results and discussion

The SEM images and diameter distributions of Au particles fabricated by repetitive formation using 5 nm Au films are shown in figure 1. The number of repetitions and the total Au thicknesses were, respectively (*a*) one (total thickness of 5 nm), (*b*) two (10 nm), (*c*) four (20 nm), (*d*) six (30 nm) and (*e*) eight (40 nm). In the one-time repetitive formation (figure 1(*a*)), the mean diameter of the Au particles was 48 nm; the full width at half maximum (FWHM) was 24 nm. In the two-time repetitive formation (*b*) with a total thickness of 10 nm, the mean diameter was the same but the diameter distribution

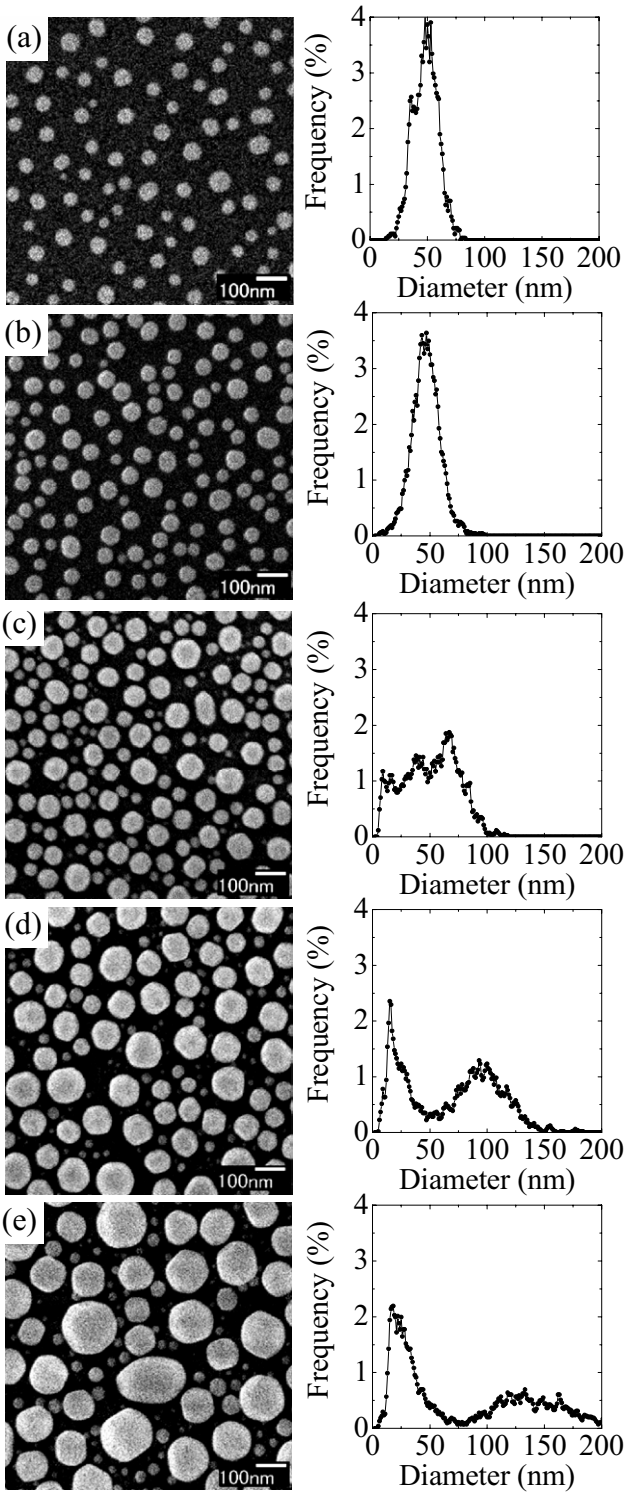

Figure 1. SEM images and diameter distributions of Au particles fabricated by the repetitive formation using 5 nm Au films. The number of repetitions were (*a*) one (total thickness: 5 nm), (*b*) two (10 nm), (*c*) four (20 nm), (*d*) six (30 nm) and (*e*) eight times (40 nm).

became slightly wider to a FWHM of 25 nm. On increasing the number of repetitions to four (figure 1(*c*)) the FWHM became wider to 51 nm; with a further increase in the number of repetitions, the diameter distribution separated into two groups with small and large diameters as shown in (*d*) and (*e*).

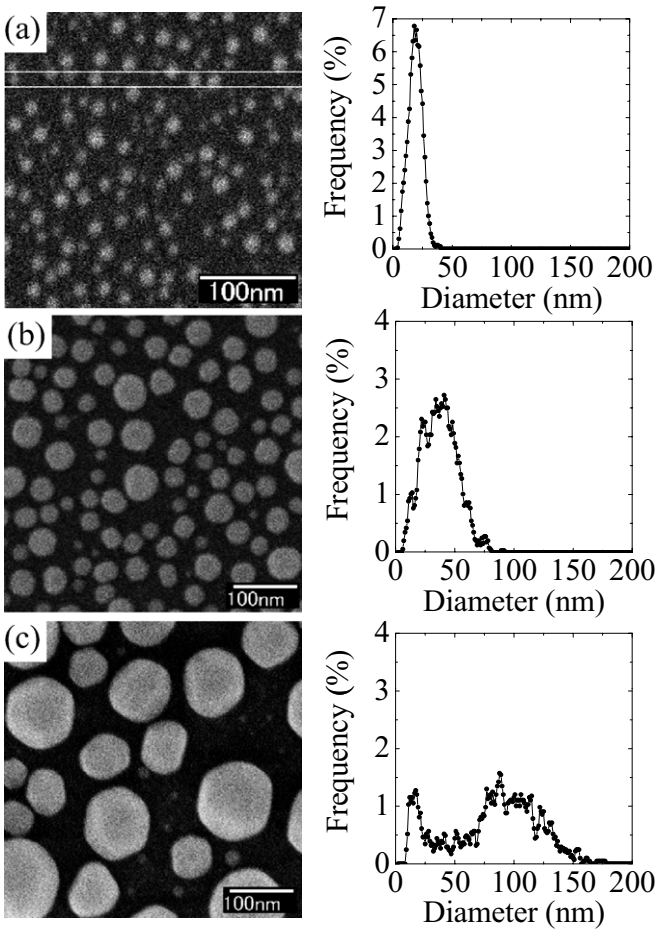

Figure 2. SEM images and diameter distributions of Au particles fabricated by repetitive formation using 2 nm Au films with number of repetitions (*a*) one (total thickness: 2 nm), (*b*) five (10 nm) and (*c*) fifteen (30 nm).

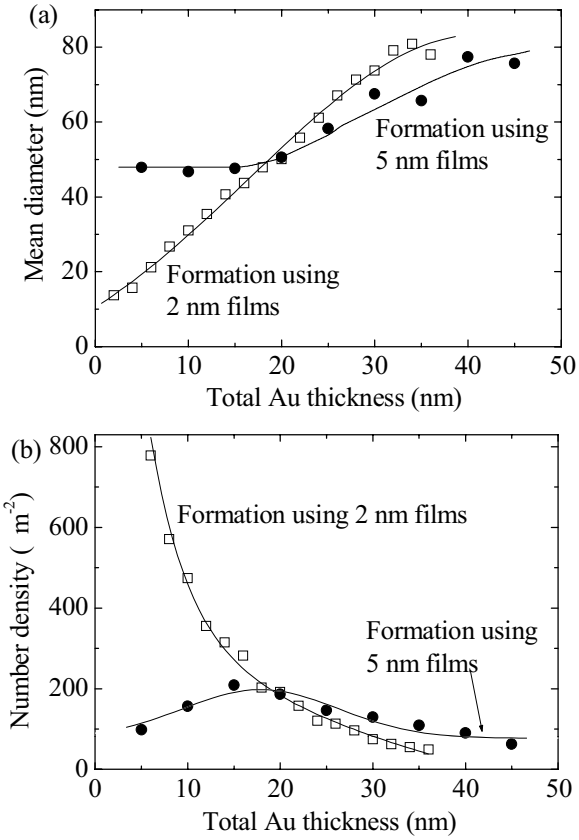

Figure 3. Total thickness of deposited Au films versus (*a*) mean diameter and (*b*) number density of fabricated Au particles.

When 2 nm Au thin films were used for the repetitive formation, a similar tendency was observed. The results of the repetitive formation with 2 nm films are shown in figure 2: (*a*) one (total thickness of 2 nm), (*b*) five (10 nm) and (*c*) fifteen times (30 nm). In the one-time repetitive formation, the mean diameter was 18 nm and the FWHM was 14 nm for the created Au particles, which were smaller than those for the one-time repetitive formation with the 5 nm film. In the case of the five-time repetitive formation (total thickness of 10 nm), the mean diameter and the FWHM increased as shown in figure 2(*b*), but this mean diameter was still smaller than that of the two-time repetitive formation with 5 nm films as shown in figure 1(*b*) which had the same total Au thickness of 10 nm. On further increasing the repetitions with fifteen-time repetitive formation (total thickness of 30 nm), the diameter distribution separated into two groups as shown in figure 2(*c*). However, the ratio of the group with small diameter was rather smaller than the case of the 5 nm film formation (figure 1(*d*)).

The separation of Au diameter distribution as seen in figures 1 and 2 can be explained as follows. The group with a large diameter may have formed by adding Au material to the pre-existing particles during the repetitive formation process, or it may have been produced by combining multiple Au particles in the process. The group with smaller diameter may have been newly created from Au fragments that remained among the pre-existing Au particles. Therefore, the mean diameter of the smaller diameter group may be kept constant because the small particle formation was repeated in every formation.

Figure 3(*a*) shows the relationship between the total thickness of Au films and mean Au diameter for the repetitive formations using 5 and 2 nm films. In the case of 5 nm film with up to three repetitions (total thickness: 15 nm), the mean diameters remained constant around 48 nm because only the FWHM increased in the diameter distribution (figures 1(*a*) and (*b*)). With four or more repetitions (total thickness of 20 nm), the mean diameter increased because the group with the larger diameter became large due to an increase in total Au thickness (figure 1(*c*)–(*e*)). In repetitive formation using 2 nm Au films, the mean diameter increased gradually with increasing total Au film thickness; with a total thickness of 20 nm the two curves for 2 and 5 nm show similar behaviour. Around this point, the obvious separation of two groups with large and small diameters also appeared.

Figure 3(*b*) shows the relationship between the total Au film thickness and number density of the fabricated Au particles using 5 and 2 nm films. With a total Au thickness of more than 20 nm, the two curves for number density were almost consistent with each other, which is the same as the case of mean diameter in (*a*). When a thin film of 2 nm was used for one formation, Au particles with small mean

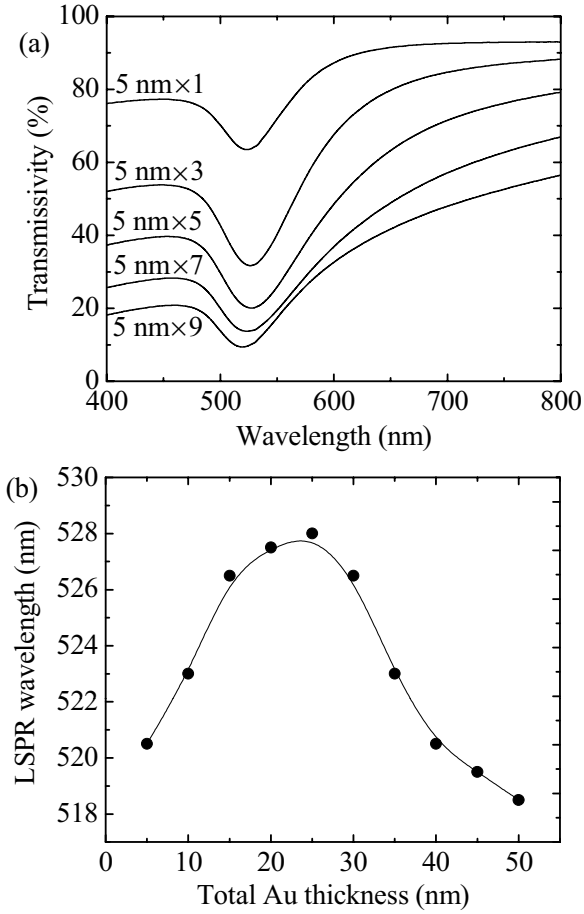

Figure 4. (*a*) Transmittance spectra of Au particles formed by the repetitive formation using 5 nm Au thin films. (*b*) LSPR wavelength versus total thickness of Au films.

diameter and enormously large number density were created; however, with increasing number of repetitions, the number density decreased drastically because combining of multiple Au particles was dominant due to small separation. In the case of 5 nm film formation, creation of new particles using Au fragments may be more dominant than combining particles because of the large separation of particles. At total thicknesses of more than 20 nm, combining particles are rather more dominant in comparison with creation, but the creation of particles by fragments in the 5 nm film repetition case may be more active than that in the 2 nm repetition case because the ratio of the group having the smaller diameter for 5 nm repetition (figure 1(*d*)) is larger than that for the 2 nm one (figure 2(*c*)).

For Au particles fabricated by repetitive formation using 5 nm Au thin films, transmittance spectra are shown in figure 4(*a*). With increasing number of repetitions, the whole transmittance curve decreased due to obstruction of illuminated light by an increase in Au volume. Absorption bands around a wavelength of 520 nm were induced by the LSPR in Au particles. As shown in figure 4(*b*), the LSPR wavelength increased to 528 nm at a total thickness of 25 nm (five repetitions) from a wavelength of 520 nm; then it decreased to a shorter wavelength. The same behaviour was seen with the result of 2 nm film repetition.

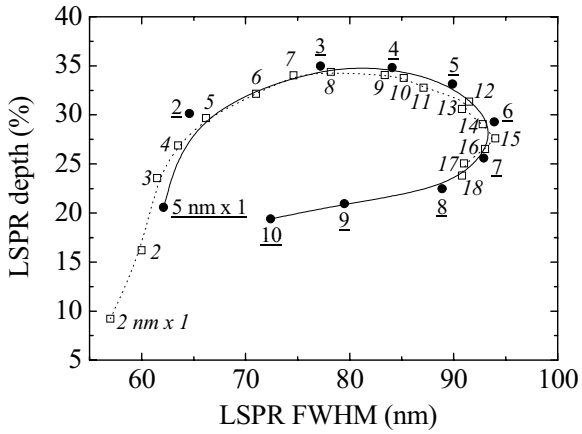

Figure 5. Relationship between FWHM and depth for LSPR absorption bands. Numbers of repetitions are denoted for 5 nm (underline) and 2 nm (italics) films.

Let us discuss the reason for the shift of the LSPR wavelength as shown in figure 4. First, we believe that the shift can be related to the diameter distribution (figure 1). Since during initial formation Au particles have a distribution with small mean diameter and small FWHM as shown in figure 1(*a*), these small Au particles excite LSPR at a shorter wavelength, e.g. 520 nm. With increasing number of repetitions, the FWHM of the diameter distribution increased, and then larger Au particles produced LSPR bands at longer wavelength. However, since there is a suitable size for a particle to induce the LSPR [14], the contribution of the separated group with larger diameter to the LSPR may become small; newly created particles will have relatively large contribution to the LSPR. Finally, newly created Au particles formed by the small fragments can play a major role in the excitation of LSPR, which have smaller diameter and FWHM than those formed by the first formation. Thus, the LSPR wavelength shifts to the shorter wavelength of 518.5 nm in comparison with that of the first formation at 520.5 nm (figure 4(*b*)).

Figure 5 shows the relationship between FWHM and depth of the LSPR absorption bands in the transmittance spectra for the Au particles formed by repetitive formation with 5 and 2 nm films. These two curves tend to be similar in a wide region.

We explain this feature of the FWHM and the depth of the LSPR absorption bands as follows. In Au particles formed by a small number of repetitions such as 2 nm × 1 time, 5 nm × 1 times (figure 5) and so on, small FWHM in the LSPR absorption band implies that the fabricated particles have a small mean diameter. Here, small LSPR depth corresponds to small Au volume. In three or four repetitions for the 5 nm film formation, the maximum depth of about 33% was obtained; the FWHM became wider because the diameter distribution became broader as seen in figure 1(*c*). On further increasing the number of repetitions to seven to ten for the 5 nm cases, the produced particles with a large diameter may not excite LSPR, but newly created small particles can produce large LSPR. Therefore, the relationship (figure 5) can also be explained by the diameter distribution of Au particles.

In the case of repetitive formation with the 2 nm films, the distinct features appear at an initial stage for the mean

diameter, the number density and also the LSPR absorption bands. However, since the LSPR itself is not so strong because of small total Au volume, it is not suitable for the enhancement of Faraday rotation. But if Au particles with small diameter and large number density are required, a thin Au film of 2 nm is of advantage. In fact, tunnel junctions of proximally positioned Au particles were tried to fabricate on a substrate using a 1 nm film [15].

After deposition of Bi : YIG films with a thickness of 60 nm on the formed Au particles on quartz substrates, transmittance and Faraday rotation spectra were measured. As shown in figure 6(*a*), with increasing number of repetitions, the LSPR absorption rapidly increased around a wavelength of 620 nm, which were shifted from around 520 nm by deposition of Bi : YIG. As is well known, a frequency ω of LSPR in the Drude model is given by

$$\omega = \frac{\omega_p}{\sqrt{1 + 2\epsilon'/\epsilon_0}},$$

where ω_p is the plasma frequency. Therefore, by deposition of Bi : YIG with permittivity ϵ', the LSPR wavelength of the Au particles will shift to longer wavelengths. In figure 6(*b*), Faraday rotation shows an increase at the LSPR wavelength. In particular, large enhancement of -1.2° was obtained by the composite film with Au particles formed by the six-time repetitive formation, which is 20 times larger than that of a single Bi : YIG thin film (figure 6(*c*)). However, transmittance became as low as 0.06 %. Accordingly, it is certain that large Faraday rotation was obtained by the influence of LSPR of the Au particles because the composite film did not have extra space around the Au particles (figure 1(*d*)) in comparison with the space around particles in (*a*). The particles with large diameter play the role in preventing insufficient light for enhanced Faraday rotation.

The prepared Au particles by repetitive formation had the size distribution as shown in figure 1. Here, the preferable size of the particles to enhance Faraday rotation should be mentioned. In the case of Ag particles, it is reported that the maximum near-field is excited by a particle with a diameter around 40 nm; the penetration length of the near-field increases with increasing diameter, for example, the penetration length is about 10 nm for a diameter of 40 nm [14]. Therefore, we guess that the preferable Au particle size may be around 40 nm or more to excite the large near-field with the long penetration to the surrounding Bi : YIG material. Our prepared samples contained Au particles in this size range, but these do not have an optimum distribution. To understand this problem for the preferable size, studies on Au particles with a narrow size distribution formed by a chemical method, electron beam lithography and so on will be required.

The performance of MO effects is quantified by a figure of merit. Here we use the definition $|\theta_F|T^{1/2}$, where θ_F is the Faraday rotation angle and T is transmissivity. Using θ_F and T at wavelengths for the maximum enhanced Faraday rotation in figure 6, the figures of merit were obtained, as shown in figure 7. That is, the figures of merit of the composite films increased with increasing number of repetitions. The maximum value of 0.08° was obtained in

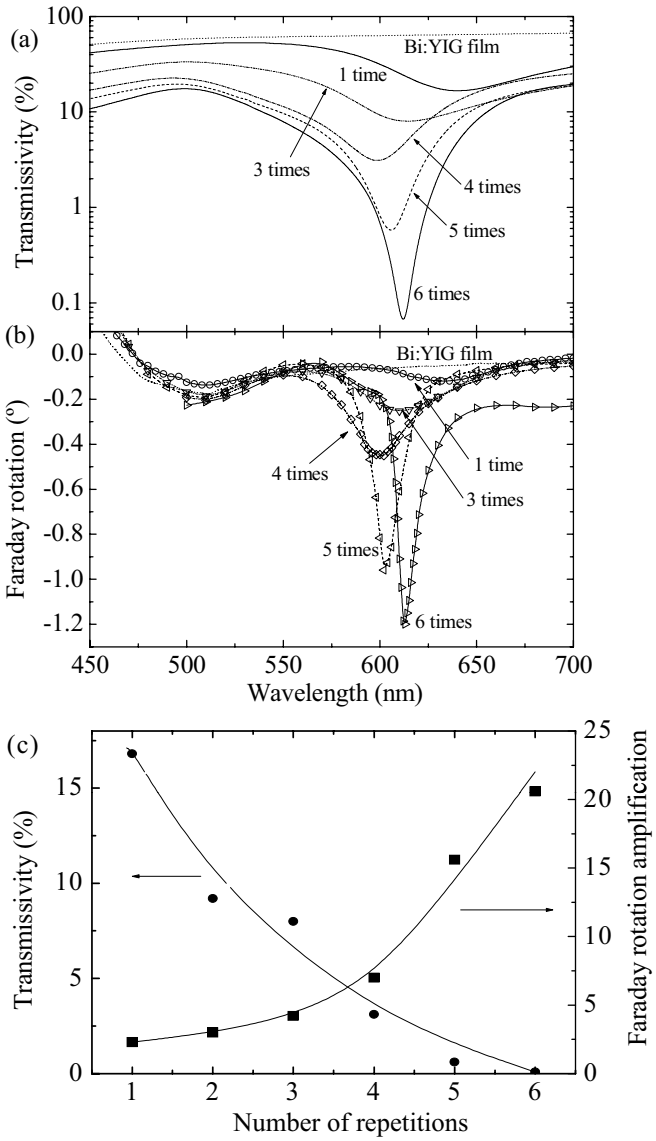

Figure 6. (*a*) Transmittance and (*b*) Faraday rotation spectra of the Bi : YIG composite films with Au particles fabricated by repetitive formation using 5 nm Au films. A single Bi : YIG film is also shown as a reference. (*c*) Transmissivity and Faraday rotation amplification versus number of repetitions.

the four-time repetitive formation; however, in the six-time repetitive formation the value decreased to 0.03°, which is smaller than that of a single Bi : YIG film because of the very small transmissivity of 0.06%.

We investigated the effective thickness for the enhanced MO effect associated with LSPR in the Bi : YIG composite film with Au particles formed by five repetitions with 5 nm films. To make a smooth surface on the composite film, a Bi : YIG film was formed by the sol–gel method on the Au particles before deposition of the sputtered Bi : YIG film. It is because the shape of the Au particles remains on a composite film surface using only a sputtered film. However, although the sol–gel Bi : YIG film does not have exactly the same MO response as the sputtered film, we believe that the result concerning effective thickness does not change in the layered Bi : YIG film by the sol–gel and the sputtering methods. Then, the

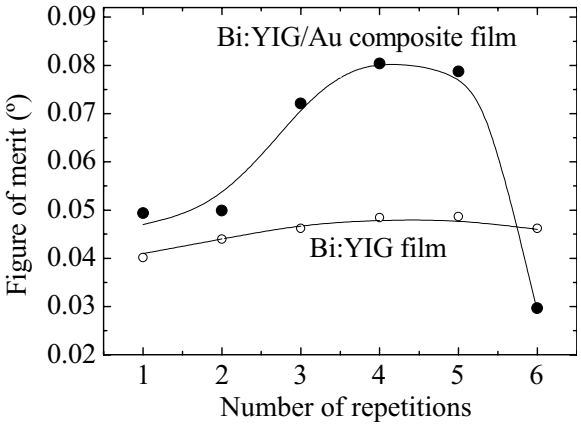

Figure 7. Figures of merit $|\theta_F|T^{1/2}$ for the Bi : YIG composite films with Au particles. Data for a single Bi : YIG film is shown as a reference.

Bi : YIG surface was gradually etched by an Ar ion milling method to measure the changes in transmittance and Faraday rotation. Figure 8(*a*) shows the cross-sectional SEM image of the fabricated composite film with a thickness of 185 nm. It is seen that the Au particles are embedded in the Bi : YIG. After ion milling to a thickness of 97 nm (*b*), the Bi : YIG was thinned to nearly the height of the large diameter Au particles. Upon ion milling to 62 nm, the Au particles were also ground as seen in figure 8(*c*). Finally, Faraday rotation was reduced by ion milling of the composite film. In figure 8(*d*), one can see that native Faraday rotation of Bi : YIG around 520 nm decreased with decreasing film thickness from 185 nm, however; large Faraday rotation associated with the LSPR did not change significantly until a thickness of 97 nm. This feature can be seen clearly from the thickness dependences of wavelength and Faraday rotation shown in figure 8(*e*). Namely, the Faraday rotation decreased from around a thickness of about 100 nm, while the LSPR wavelength shifted to shorter wavelengths. These shifts mean that the influence of the LSPR on the enhancement of Faraday rotation is limited within 100 nm Bi : YIG thickness. However, in the composite film fabricated by five-time repetitive formation Au particles separated into two diameter groups of about 20 and 95 nm. Although it is not clear which size of the Au particles induced this effective thickness of 100 nm in Faraday rotation, the result shows that the enhanced Faraday rotation was produced by the localized area around Au particles.

4. Conclusions

A repetitive formation method of Au particles was used to fabricate Bi : YIG composite films. This method is suitable to control Au volume and number density of particles; furthermore, it has the great advantage of a simple fabrication process. The Bi : YIG composite films with the Au particles were able to produce enhanced Faraday rotation. In composite film with six repetitions, a Faraday rotation angle of -1.2° was obtained, which is almost 20 times larger than that of a Bi : YIG film. Although it is a considerably large enhancement of MO effect, the figure of merit was not so large because of very small

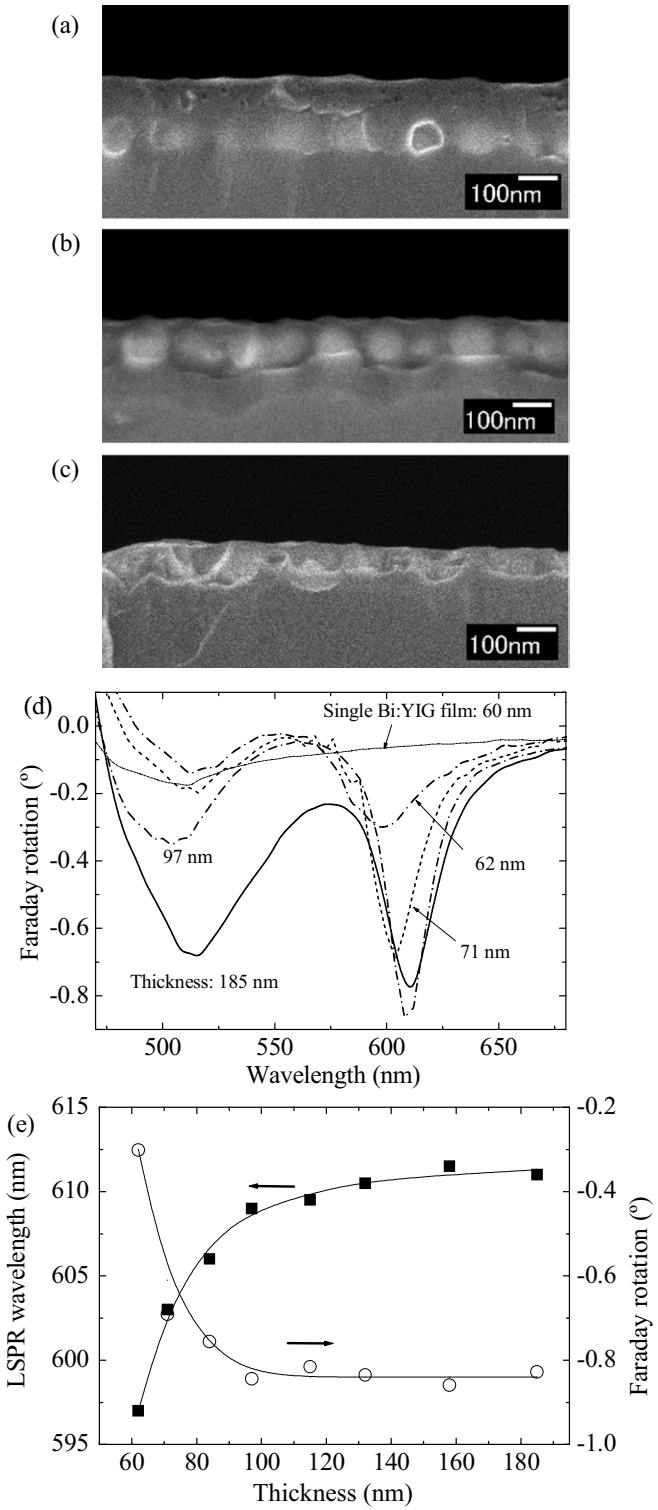

Figure 8. Ar ion milling of the Bi : YIG/Au particles composite film (five repetitions with 5 nm films). Cross-sectional SEM images of the composite film with a thickness of (*a*) 185 nm, (*b*) 97 nm and (*c*) 62 nm. (*d*) Changes in transmissivity spectra of the composite film. A reference of single Bi : YIG film is added. (*e*) Film thickness dependences of LSPR wavelength and Faraday rotation.

transmissivity of 0.06%. For details, the prepared Au particles by the repetitive formation had the size distribution. However, all sizes of Au particles are not effective in enhancing the Faraday rotation; there must be a preferable size and distance

between them. We believe that the Bi : YIG/Au composite film has a large possibility to show further enhancement of Faraday rotation by an optimum structure.

Acknowledgments

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References

[1] Kahn F J, Pershan P S and Remeika J P 1969 *Phys. Rev.* **186** 891
[2] Scott G B, Lacklison D E, Ralph H I and Page J L 1975 *Phys. Rev.* B **12** 2562
[3] Park J H, Takagi H, Cho J K, Nishimura K, Uchida H and Inoue M 2003 *Japan. J. Appl. Phys.* **42** 2332
[4] Feil H and Haas C 1987 *Phys. Rev. Lett.* **58** 65
[5] Katayama T, Suzuki Y, Awano H, Nishihara Y and Koshizuka N 1988 *Phys. Rev. Lett.* **60** 1426
[6] Kosobukin V A 1998 *Surf. Sci.* **406** 32
[7] Abe M and Suwa T 2004 *Phys. Rev.* B **70** 235103
[8] Tomita S, Kato T, Tsunashima S, Iwata S, Fujii M and Hayashi S 2006 *Phys. Rev. Lett.* **96** 167402
[9] Khanikaev A B, Fedyanin A A, Baryshev A V, Inoue M and Granovsky A B 2007 *Opt. Express* **15** 6612
[10] Okubo T, Fujikawa R, Masuda Y, Fedyanin A, Baryshev A, Khanikaev A, Shin K H, Uchida H, Lim P B and Inoue M 2006 *Techical Report Inst. Electr. Eng. Japan* MAG-06-167 1 (in Japanese)
[11] Uchida H, Masuda Y, Fujikawa R, Baryshev A V and Inoue M 2009 *J. Magn. Magn. Mater.* **321** 843
[12] Mizutani Y, Uchida H, Masuda Y, Baryshev A V and Inoue M 2009 *J. Magn. Soc. Japan* **33** 481
[13] Matsumoto K, Yamamoto S, Yamanobe Y, Ueno A, Yamaguchi K and Fujii T 1991 *Japan. J. Appl. Phys.* **30** 1696
[14] Fukui M and Ohtsu G 2003 *Fundamental of Optical Technology* (Tokyo: Ohmsha) chapter 6 p 107 (in Japanese)
[15] Uchida H, Fumoto H, Baryshev A, Kim J, and Inoue M 2008 *IEEJ Trans. Electr. Electron. Eng.* **3** 660