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Annealing Temperature Dependence of Magnetic and Optic Properties of Bi:YIG Films Deposited with Aerosol Deposition Method

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Bismuth-substituted yttrium iron garnet (Bi:YIG, $Bi_{0.5}Y_{2.5}Fe_5O_{12}$) films were deposited with aerosol deposition method and their magnetic and optical properties were investigated as a function of annealing temperature. Since the ceramic films deposited with aerosol deposition method have not a perfect crystal structure due to non-uniform internal stress occurred by mechanical collision during their deposition, the post annealing could be a key process to release its internal stress and to improve its micro structure for optimizing the magnetic and magneto-optic properties of films. The crystallinity of Bi:YIG film was improved with increase of annealing temperature, and the saturation magnetization increased up to 87 emu/cc at 800°C. The Faraday rotation increased up to 1.4 deg/μm by annealing at 700°C around the wavelength of 0.5 μm. The optical transmittance of the Bi:YIG film was also improved in visible region.

Keywords : bismuth-substituted yttrium iron garnet (Bi:YIG) film, aerosol deposition, saturation magnetization, Faraday rotation, optical transmittance

1. Introduction

Bismuth-substituted yttrium iron garnet (Bi:YIG) films, which show excellent magneto-optical properties as well as low dielectric and optical losses [1, 2], have been attracting great attention recently because of their applicability for microwave and optical signal conditioning devices such as isolator, circulator, TE-TM mode conversion and wavelength accordable filter. For applying a practical optical device, the productivity including deposition speed and low process temperature is a matter of great importance. It should be noticed that it is difficult to fabricate thick ceramic film with over a several μm by using conventional deposition methods, such as sol-gel method, sputtering [3] and thermal decomposition [4], due to crack and large stress introduced by post thermal process. From this practical viewpoint, the aerosol deposition method (ADM) [5-8] could be best choice to fabricate Bi:YIG films, especially in the case of thick films over several μm, because of its extremely high deposition speed and low process temperature. However, in ordinary case of ADM, it is not easy to make a magneto-optical thick film, such as Bi:YIG, has both good magnetic property and high transparency in spite that transparency is one of most important matters for an optical device application. This is due to the denser film shows better transparency but worse magnetic property due to internal stress and magnetoelastic coupling. In this study, we have investigated whether the magnetic and optical properties of Bi:YIG thick films deposited by ADM could be improved by optimizing their post annealing condition for the final purpose of developing optical isolator.

2. Experimental Method

ADM employs sub-micrometer particles, Bi:YIG fine particles in this study, that were accelerated with carrier gas to be ejected from a nozzle in a vacuum chamber. The accelerated particles with the speed of a few hundred m/s [9] collide onto a substrate and then formed a crystalline film. The crystalline Bi:YIG particles were prepared by a solid state reaction of the precursor powder with the composition of $Bi_{0.5}Y_{2.5}Fe_5O_{12}$, and treated successively

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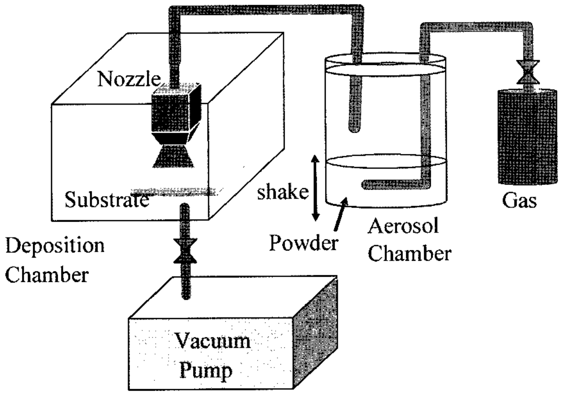
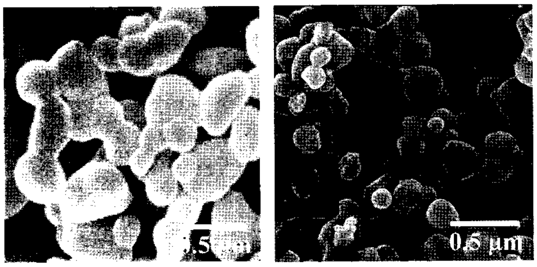


Fig. 1. Schematic illustration of aerosol deposition apparatus.

by the ball milling with 100 rpm for 2 hours in the isopropyl alcohol. The Bi:YIG films were deposited using the prepared Bi:YIG powders onto corning 1737 glass substrates by AD apparatus illustrated schematically in Fig. 1. The film depositions were performed at room temperature. The carrier gas was nitrogen with the flow rate of 9 L/min. The vacuum pressure in the deposition chamber was kept in 5 Pa during the film deposition. The fabricated films were annealed at 600~800°C for 10 minutes in air. The magnetic properties of Bi:YIG films were measured by a vibrating sample magnetometer (VSM) with the maximum magnetic field of ±5 kOe. The crystallinities of films were determined by using an X-ray diffractometer (XRD) with Cu-K radiation. The optical and magneto-optical properties were measured with an optical spectrometer and a magneto-optical spectrometer with off-crossed polarization method [10], respectively.

3. Results

Figs. 2(a) and (b) show SEM images of the powders (a) annealed at 1000°C and (b) ball-milled for 10 hours after annealing. Powder preparation, especially thermal treatment, is an important pre-process in ADM, since adequate crystal structure of particle allows the film having appropriate crystal structure even if the film is deposited at relatively low temperature. The annealing temperature was decided in terms of micro-structure and saturation magnetization of the powders.



(a) (b)

Fig. 2. SEM images of Bi:YIG powders (a) annealed at 1000°C and (b) ball-milled for 10 hours.

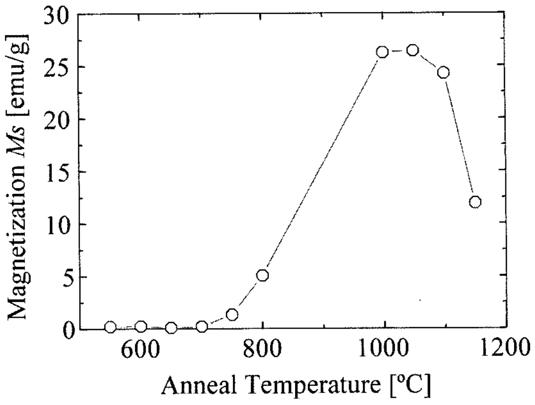


Fig. 3. Annealing temperature dependence of saturation magnetization of the powders.

Fig. 3 shows the annealing temperature dependence of saturation magnetization of the powder. The saturation magnetization of the powders increased with increase of annealing temperature up to 1050°C, but extraordinary grain-growths were observed over 1000°C. Indeed, neck-connections of particles for grain growth appeared in the film annealed at 1000°C as shown in Fig. 2(a). This is why the process to control of particle size, which should be carried out after annealing successively, is needed to obtain the films having both excellent microstructure and magnetic properties in ADM. We could obtain the particles with suitable size and shape through the ball milling process after annealing. The average particle diameter of the powder prepared finally was ~240 nm.

Fig. 4 shows SEM image of the cross section of as-

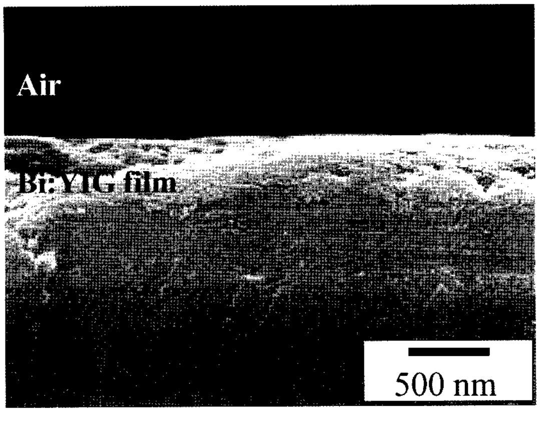


Fig. 4. SEM image of cross section of the Bi:YIG film deposited by ADM apparatus.

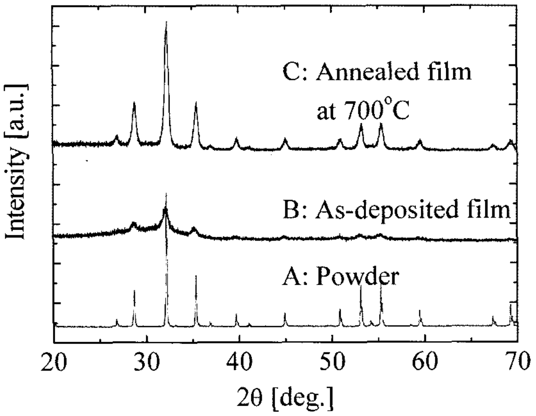


Fig. 5. XRD patterns of Bi:YIG particles and films.

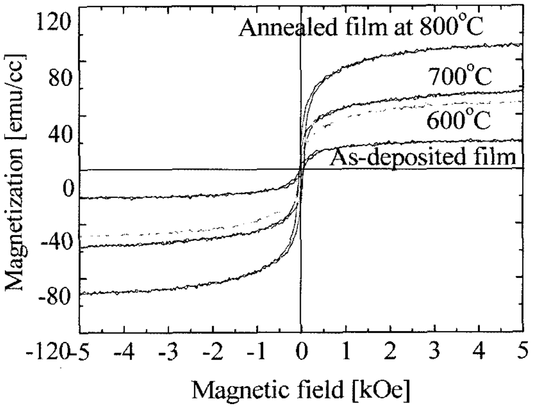


Fig. 6. Magnetization curves of the Bi:YIG film.

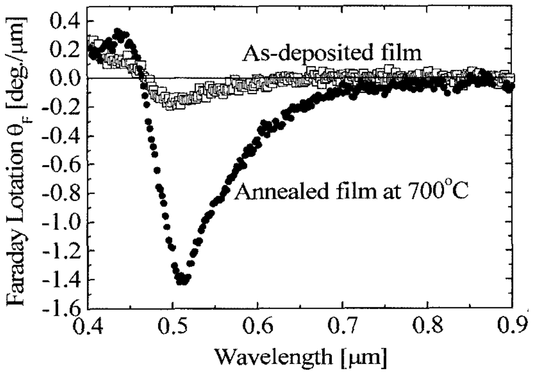


Fig. 7. Spectra of Faraday rotation of the Bi:YIG films in wavelength from 0.4 to 0.8 μm.

deposited Bi:YIG film with the thickness of 1 μm. As shown in Fig. 4, there was few void space inside film resulting the highly dense film, although the film was deposited with extremely high deposition rate (~1 μm/min).

Fig. 5 shows the XRD patterns of the Bi:YIG powder, the as-deposited film and the film annealed for 10 minutes at 700°C. It should be noticed that the as-deposited Bi:YIG film (curve B) had polycrystal garnet structure without any thermal treatment. However, the intensities of peaks were much lower than that of the original powder (curve A), which means the particles were smashed by mechanical collision onto substrate during their deposition. Moreover, the widths of peaks became more broaden after deposition, indicating the crystalline size decreased and crystal lattices were strained. The stress could be formed during not only deposition but also ball milling process. But, it was considered that stress was formed mainly during deposition process, because the XRD peaks of the film were largely broaden after deposition. In addition to this, the shift of peaks, which is proportional to lattice strain, indicated strongly the crystal lattices had been strained. However, intensities of peaks of the annealed Bi:YIG film (curve C) increased notably, indicating that the micro-structure could be recovered by the suitable annealing process.

Fig. 6 shows the magnetization curves of as-deposited Bi:YIG film and annealed films at 600, 700 and 800°C for 10 minutes. From these results, it is easy to find that the magnetic properties were improved by annealing. The saturation magnetization M_s increased by annealing at over 600°C, and reached 87 emu/cc at 800°C. However, when the Bi:YIG film was annealed over 800°C, the film roughness was increased because of the grain growth. So, it was estimated that the optimum annealing temperature might be around 700°C in a viewpoint of magneto-optic property in spite that M_s increased with increase of the annealing temperature up to 800°C.

Fig. 7 shows the Faraday rotation spectra of Bi:YIG films. At the wavelength around 500 nm, the Faraday rotation angle of as-deposited film was only 0.2 deg/μm. But, the Faraday rotation was also improved by annealing, which was due to the recovery of crystalline structure and magnetic property of the film. The Faraday rotation angle reached 1.4 deg/μm by annealing at 700°C. The films annealed at over 800°C had so rough surface, as mentioned above, that it was difficult to obtain sufficient precision in the measurement of magneto-optic properties (Faraday rotation) and optical transmission.

Fig. 8 shows the optical transmission spectra of Bi:YIG film with 1.5 μm thick. The transmittance increased with a crystalline improvement, which was achieved by annealing. In particular, the transmittance of the film annealed at 700°C increased greatly in visible region. It was confirmed by considering experimental results that the magnetic and optical properties of Bi:YIG films deposed by ADM were improved by annealing in relatively low temperature. This fact provides the evidence that the

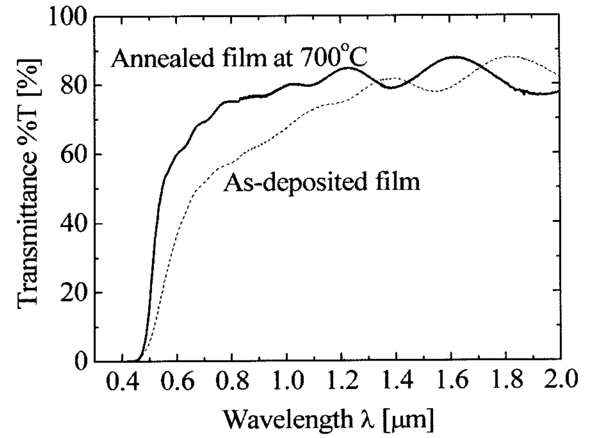


Fig. 8. Transmission spectra of the Bi:YIG films.

strain was induced in the crystal structure when crystalline powder was deposited on substrate. It was supposed that the BiYIG films deposited by ADM could have good magnetic and magneto-optic properties despite of lower annealing temperature compare with conventional sintering process ~1000°C, because the film was sintered by not only thermal energy of annealing but also mechanical energy of the internal strain, which was formed during deposition.

4. Conclusion

Highly dense Bi:YIG films were deposited on glass substrates by ADM at room temperature to investigate the annealing temperature dependence on their magnetic and magneto-optic properties. The experimental results indicated these properties were improved largely by annealing. The saturation magnetization increased with increase of annealing temperature, however, which made the film roughness increased at over 800°C. The Faraday rotation was improved with annealing at 700°C from 0.2 deg/μm to 1.4 deg/μm. It should be mentioned again that the films deposited by ADM can show notable magnetic and magneto-optic properties in spite of relatively low annealing temperature ~700°C. This fact suggests that the Bi:YIG film deposited by ADM might be promising to construct an optical component with an inexpensive glass substrate, such as borosilicate glass of which softening temperature is around 800°C.

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