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Faraday effect of polycrystalline bismuth iron garnet thin film prepared by mist chemical vapor deposition method

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ABSTRACT

We have synthesized polycrystalline thin film composed of a single phase of metastable bismuth iron garnet, $Bi_3Fe_5O_{12}$, on a fused silica substrate, one of the most widely utilized substrates in the solid-state electronics, by using mist chemical vapor deposition (mist CVD) method. The phase purity and stoichiometry are confirmed by X-ray diffraction and Rutherford backscattering spectrometry. The resultant thin film shows a small surface roughness of 3.251 nm. The saturation magnetization at room temperature is 1200 G, and the Faraday rotation angle at 633 nm reaches −5.2 deg/μm. Both the magnetization and the Faraday rotation angles are somewhat higher than those of polycrystalline BIG thin films prepared by other methods.

1. Introduction

Garnet-type ferrites such as $Y_3Fe_5O_{12}$ (YIG) and $Gd_3Fe_5O_{12}$ (GIG) are known to show large Faraday effect in a wide wavelength range covering the ultraviolet, visible, and infrared regions. In particular, they are highly transparent as well as possess large Faraday rotation angle in the infrared region, and consequently, those garnet-type ferrites are nowadays practically utilized as nonreciprocal photonics devices such as optical isolators and modulators in the optical telecommunications, which break the time-reversal symmetry of light propagation. It is also known that the replacement of the rare-earth ion by Bi^{3+} ion at the eight-coordinated site in the garnet-type structure efficiently enhances the Faraday effect while keeping the transparency to the infrared light rather high [1]. Although the concentration of Bi^{3+} which can replace the rare-earth ion in a stable phase of garnet-type ferrite is restricted, $Bi_3Fe_5O_{12}$ (bismuth iron garnet, BIG), a garnet-type ferrite with the eight-coordinated sites occupied only by Bi^{3+} ions, can be synthesized as a metastable phase, as first demonstrated by Okuda et al. [2] BIG manifests extraordinary large Faraday effect and high magneto-optical figure of merit in the visible and near-infrared region. This metastable compound can be epitaxially grown onto only a single-crystalline garnet substrate because it forms in a non-equilibrium way. Actually, if a non-garnet material such as silica glass is used as a substrate, $BiFeO_3$ and $Bi_2Fe_4O_9$ that are thermodynamically stable phases are readily grown. Therefore, it is not easy to practically apply this excellent magneto-optical material to integrated photonics system, where construction of tiny photonics devices on ubiquitous substrates such as silica glass is strongly required. To overcome this problem, buffer layers with garnet-type structure have been introduced for the deposition of BIG thin film on non-garnet-type substrates, and several attempts have been made to prepare polycrystalline BIG thin films onto silica glass substrates by using several deposition techniques including reactive radio-frequency (RRF) sputtering, reactive ion beam (RIB) sputtering, and pulsed laser deposition (PLD) [3–7].

In the previous studies, we have demonstrated that mist chemical vapor deposition (mist CVD) method, one of the emerging techniques for thin film deposition, is an efficient process to achieve high quality garnet-type ferrite thin films [8–10]. BIG thin film epitaxially grown on a single-crystallinegadolinium gallium garnet substrate by means of mist CVD method exhibits magneto-optical functionalities comparable to those of BIG thin films prepared by other deposition techniques. For instance, magneto-optical figure of merit for the mist CVD-derived epitaxial BIG thin film is 1.5 times larger than that of PLD-derived thin film, due to its excellent optical properties which stem from a relatively flat surface and better morphology [9]. In the present work, we report on the successful preparation of polycrystalline thin film composed of a single phase of BIG on a fused silica (amorphous SiO_2) substrate by using the mist CVD method. Buffer layers utilized in our study are YIG and yttrium aluminum garnet (YAG, with the chemical formula of $Y_3Al_5O_{12}$). We also discuss physical properties including optical transmittance, magnetization, and Faraday effect for the resultant thin film.

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2. Experimental procedure

The mist CVD setup utilized in this study is a fine-channel type mist sourced film former (TOUKI Co. Ltd., Type: MSFF-FC). Fused silica substrates were ultrasonically cleaned before deposition of thin films. First, a buffer layer of polycrystalline YIG was deposited on the fused silica substrate by using the mist CVD method. The precursor solution was prepared by dissolving tris(acetylacetonato)iron(III), $Fe(C_5H_7O_2)_3$, and tris(acetylacetonato)yttrium(III) hydrate, $Y(C_5H_7O_2)_3 \cdot nH_2O$, in acetone. Details of the preparation method for YIG layer were described elsewhere [8]. A buffer layer of polycrystalline YAG was also prepared by the mist CVD method to evaluate adequately magnetic and magneto-optical properties of polycrystalline BIG thin film. The precursor solution was prepared by dissolving tris(acetylacetonato)yttrium(III) hydrate, $Y(C_5H_7O_2)_3 \cdot nH_2O$ and tris(acetylacetonato)aluminum(III), $Al(C_5H_7O_2)_3$, in *N*,*N*-dimethylformamide (DMF). It should be noted that aluminum oxide had an extremely low deposition rate, which was about one twelfth of iron oxide and one fifth of yttrium oxide at a substrate temperature of 500 °C. Thus, when the molar ratio of $Y(C_5H_7O_2)_3/Al(C_5H_7O_2)_3$ in the precursor solution was equal to 0.125, a single phase of YAG without any impurity phases could be obtained. For the process, the substrate temperature and the post-annealing temperature were 500 and 800 °C, respectively.

For the preparation of polycrystalline BIG thin film, the precursor solution was prepared by dissolving tris(acetylacetonato) iron(III), $Fe(C_5H_7O_2)_3$ in DMF and mixing it with 2-ethylhexanoic acid solution containing 25 wt% of bismuth(III) 2-ethylhexanoate, $Bi(C_8H_{15}O_2)_3$. The molar ratio of $Bi(C_8H_{15}O_2)_3$ to $Fe(C_5H_7O_2)_3$ was kept to be 0.463 and the total concentration was 0.050 mol/L. The precursor solution was ultrasonically atomized by using a 2.4 MHz transducer, and the resultant mist droplets were transferred to a reaction vessel with nitrogen gas at a flow rate of 6 L/min. The substrate temperature was set as 300 °C and single-phase of BIG was grown at a post-annealing temperature of 530 °C for 30 min. All of these process conditions are the same as those described in Ref. [9], in which the conditions proved to be optimal.

The crystalline phase was identified by using X-ray diffraction (XRD) with CuK_α radiation. The composition of the thin films was determined by Rutherford backscattering spectrometry (RBS) using a 2 MeV He^+ ion beam, which was produced by a Pelletron-type accelerator located at the heavy-ion accelerator facility at the Quantum Science and Engineering Center of Kyoto University. The thickness of BIG thin films and buffer layers was evaluated by a high-sensitivity surface profiler and confirmed by the analysis of Rutherford backscattering spectra. The surface morphology of the thin films was observed by atomic force microscopy (AFM).

Magnetic field dependence of magnetization was measured at room temperature by using a superconducting quantum interference device magnetometer (SQUID). Optical transmittance measurements were carried out at room temperature in a wavelength range from 500 to 2000 nm. Faraday rotation angle was measured at room temperature by a polarization modulation technique with a Xe lamp as a light source. The thin film sample was placed in a static magnetic field of 15 kOe applied in a direction perpendicular to the surface of the thin film, and the rotation angle was measured in a wavelength range from 350 to 850 nm. The Faraday rotation angles of fused silica substrate with YAG buffer layer were measured before the deposition of BIG thin film and subtracted from those of the sample composed of BIG thin film, YAG buffer layer and the fused silica substrate to obtain the values for the BIG thin film.

3. Results and discussion

Fig. 1(a) shows the XRD patterns of as-deposited (amorphous) and post-annealed (crystallized) thin films. For the as-deposited thin film, all the diffraction lines are ascribable to YIG, suggesting that the YIG buffer layer is thick enough to be clearly detected by XRD; the thicknesses of the YIG layer evaluated by the high-sensitivity surface profiler is 45 nm. On the other hand, the diffraction lines observed for the post-annealed thin film are assigned to BIG in addition to YIG, indicating that the thin film is composed of a single phase of BIG without any impurity phases. The measurement by high-sensitivity surface profiler indicates that the BIG thin film is 220 nm thick. The average grain size of BIG can be evaluated by the Williamson–Hall method:

$$\frac{\beta \cos\theta}{\lambda} = 4\xi\frac{\sin\theta}{\lambda} + \frac{1}{t},$$

where β, λ, ξ, and t indicate the full width at half maximum (FWHM), the wavelength of X-ray (1.54060 Å), the strain, and the grain size, respectively. By using the relation, the average grain size of the polycrystalline BIG thin film prepared in the present study is estimated to be 50.8 nm. The lattice parameter is obtainable by using the relation between the 2θ peak positions and the Nelson–Riley function: $1/2[(\cos^2\theta/\sin\theta)+(\cos^2\theta/\theta)]$ [11] and extrapolating the data to $\theta=\pi/2$. The lattice parameter thus obtained is $a=12.631$ Å, in excellent agreement with values reported previously [9,12–14].

XRD pattern of post-annealed thin film prepared on a YAG buffer layer/fused silica substrate is shown in Fig. 1(b). Each of the diffraction lines is ascribable to BIG or YAG, indicating that single phase of BIG is grown also on the YAG layer/fused silica substrate. The average grain size and the lattice parameter of the BIG thin film are evaluated to be 54.3 nm and 12.636 Å, respectively. These values are almost identical to those obtained for the BIG thin film

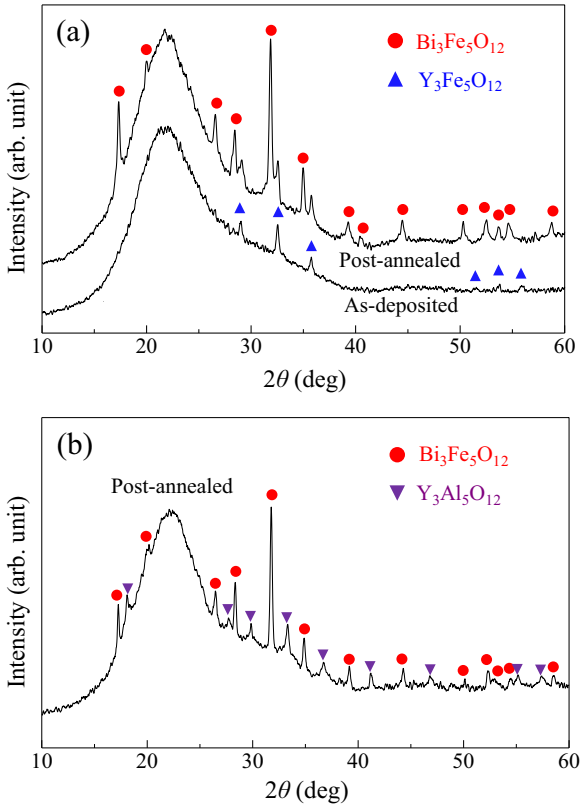

Fig. 1. X-ray diffraction patterns of (a) as-deposited and post-annealed thin films grown on YIG buffer layer/fused silica substrates and (b) post-annealed thin film grown on YAG buffer layer/fused silica substrate. The closed circles, triangles, and upside-down triangles denote X-ray diffraction lines assigned to BIG, YIG, and YAG phases, respectively.