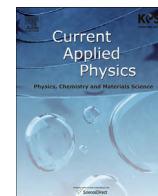




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# Effects of heating rate on the magneto-optical properties of bismuth-substituted yttrium iron garnet films prepared via modified metal-organic decomposition

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## ABSTRACT

This work investigated the effects of heating rate and annealing on the magneto-optical properties of bismuth-substituted yttrium iron garnet (Bi-YIG) thin films on glass and (111)-oriented single-crystalline gadolinium gallium garnet (GGG) substrates fabricated by metal-organic decomposition (MOD). We modified the MOD method by eliminating the pre-annealing process. We performed annealing at various temperatures to determine the optimal temperature for obtaining the Bi-YIG phase. We then annealed at the optimized temperature using various heating rates. The optimal conditions were annealing for 1 h at 750 °C at a heating rate of 30 °C/min on GGG to obtain highly crystallized fine grains. The Faraday rotation for this film was about  $-10.5^\circ/\mu\text{m}$ . The optimized heating rate enhanced the magneto-optical properties due to improved crystallinity and saturated magnetization. The Bi-YIG thin films prepared by this prescribed MOD method exhibited excellent magneto-optical performance and are potential candidates for applications in optical devices.

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## 1. Introduction

Bismuth-substituted yttrium iron garnet (Bi-YIG) is widely used for magneto-optical (MO) devices due to its superior characteristics, including the large Faraday rotation, high optical transmittance in the visible and near-infrared regions, and extremely high MO activity [1,2]. These advantages make Bi-YIG an attractive material for developing MO microscopes with high magnetic sensitivity and high time resolution compared to magnetic force microscopes (MFM), as well as applications in superconducting quantum interference devices (SQUIDS) [3]. Many studies have reported the use of Bi-YIG thin films as magnetic indicators in MO microscopes for imaging based on the MO effect. For example, real-time observations of the vortex dynamics in a superconductor and magnetic domain measurements have been reported [4]. Additionally, Bi-YIG thin films have been used as spin-thermoelectric coatings to generate the spin current in thermoelectric thin-film

technology [5].

Materials that are well suited for indicator films in MO microscopy should have a large Faraday rotation angle, low optical absorption in the given optical limit, and thin structure (thickness  $< 1 \mu\text{m}$ ). These properties directly affect the sensitivity, resolution, and quality of MO images [3]. Various methods have been used to produce high-quality Bi-YIG thin films, including liquid-phase epitaxy (LPE) [6], magnetron sputtering [7], pulsed laser deposition (PLD) [8], the sol-gel process [9], and metal-organic decomposition (MOD) [1,3,5]. Among these methods, MOD is preferable because of its simplicity and low cost. It also provides homogeneity over a large area and precise control of the composition and magnetic properties of the resulting films [1,3]. Therefore, many studies have reported the MO properties of Bi-YIG thin films prepared using the MOD method. For example, H. Lee et al. fabricated Bi-YIG thin films on glass substrates at an annealing temperature of 700 °C and reported a Faraday rotation angle of  $-2.47^\circ/\mu\text{m}$  [10]. T. Ishibashi et al. fabricated high-concentration Bi-YIG ( $\text{Bi}_{2.5}\text{Y}_1\text{Fe}_5\text{O}_{12}$ ) thin films on gadolinium gallium garnet (GGG) substrates by annealing at 750 °C and reported a Faraday

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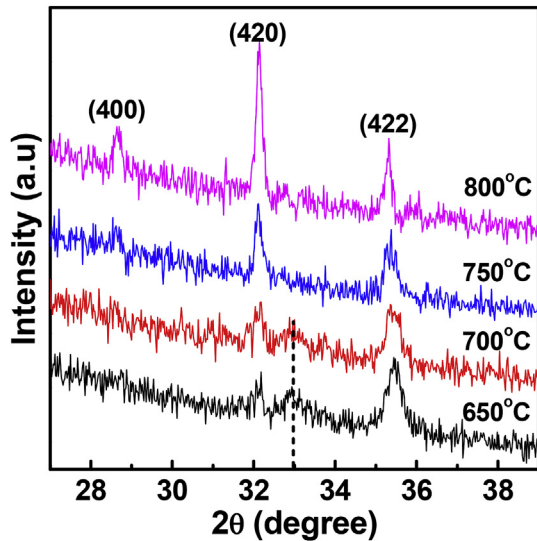


Fig. 1. X-ray diffraction (XRD) patterns of Bi-YIG thin films on GGG annealed at different temperatures (650–800 °C).

rotation angle of  $-1.55^\circ/\mu\text{m}$  [11]. O. Galstyan et al. prepared Bi-YIG thick films ( $0.8\text{--}1.2\ \mu\text{m}$ ) on glass substrates using the pre-crystallization process and reported a Faraday rotation angle of  $-3.75^\circ/\mu\text{m}$  [12]. O. Galstyan et al. also prepared Bi-YIG ( $\text{Bi}_2\text{Y}_1\text{Fe}_5\text{O}_{12}$ ) thin films on glass GGG substrates with a thickness of  $0.8\ \mu\text{m}$  [3]. They reported a Faraday rotation angle of  $-11^\circ/\mu\text{m}$  for the high-bismuth-doped Bi-YIG thick film. All of these reports confirmed that the annealing process in MOD is critical for obtaining high-quality Bi-YIG thin films [1,3,4]. The annealing temperature and heating rate strongly affect the MO and crystalline properties of the Bi-YIG thin films [10,13,14]. Additionally, the magnetic properties of the Bi-YIG films are affected by the heating rate, and this dependence has not been thoroughly investigated.

In this study, we prepared Bi-YIG thin films on amorphous glass and single-crystalline GGG substrates by modifying the MOD method and investigated the heating rate and annealing effects on MO properties. X-ray diffraction (XRD) and high-resolution transmission electron microscopy (HRTEM) revealed that the heating

rate significantly affected densification, crystallization, and magnetic properties of the Bi-YIG thin films.

## 2. Experimental

The MOD solution for thin-film preparation was purchased from Kojundo Chemical Laboratory, and the Bi:Y:Fe ratio was 1:2:5. First, the MOD solution was spin-coated onto a clean substrate using a spin coater at 500 rpm for 5 s and 1000 rpm for 30 s. The substrates were cleaned using standard cleaning procedures. The coated film was dried at 100 °C for 30 min on a hot plate to evaporate the solvent. Next, the film was annealed for 1 h in a tube furnace under an oxygen atmosphere at temperatures of 650–800 °C at a heating rate of 10 °C/min. Annealing was also performed at various heating rates (10–40 °C/min) at the optimal temperature for the garnet phase. These steps were repeated until the desired thickness was achieved; one process cycle resulted in a film thickness of approximately 75 nm. We prepared two sets of samples using different heating rates to achieve a thickness of 150 nm. One set was prepared on single-crystalline GGG substrates and the other set on Corning's Eagle XG glass.

The setup for measuring the Faraday rotation angle has been described in a previous report [10]. We used an Ocean optics HL-2000 as the light source, with a wavelength range of 360–2400 nm. The cross-sectional views of the films were obtained by TEM using an FEI (Japan) Tecnai F-30 microscope operated at 300 kV. The crystal structure and magnetization were characterized using a high-resolution X-ray diffractometer (Bruker D8 Discover) and a vibrating sample magnetometer (VSM, Lakeshore 7304), respectively.

## 3. Results and discussion

Fig. 1 presents the XRD patterns of the Bi-YIG thin films prepared on (111)-oriented GGG substrates at different annealing temperatures and heating rates. The films annealed at 650 °C and 700 °C exhibited a secondary phase with low crystallinity. The secondary phase was identified as orthorhombic yttrium ortho-ferrite ( $\text{o-YFeO}_3$ ), and it is shown as the dashed line in Fig. 1 [15]. The films annealed at 750 °C and 800 °C exhibited the garnet phase only. The peaks correspond to the (400), (420), and (422) planes, and the (420) peak was the strongest at  $2\theta = 32^\circ$ . Thus, an annealing

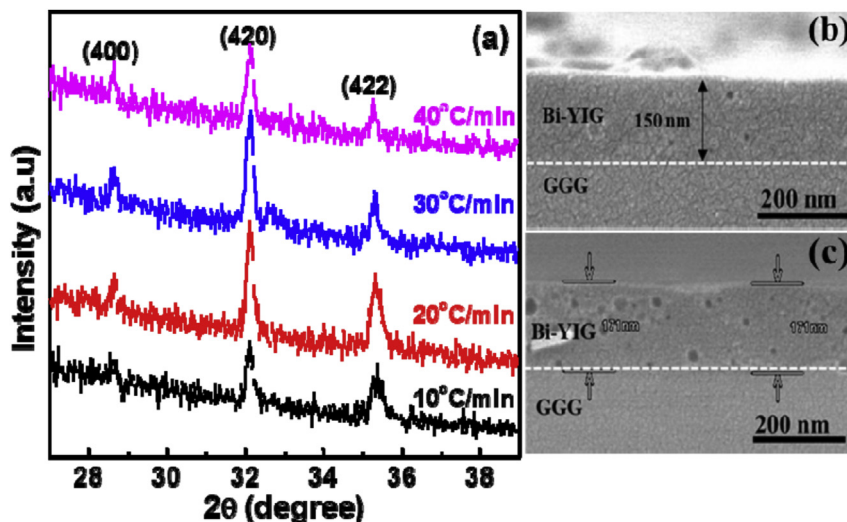


Fig. 2. (a) XRD patterns of Bi-YIG thin films on GGG annealed at different heating rates (10–40 °C/min) at 750 °C. Cross-sectional scanning electron microscopy (SEM) images of films annealed at (b) 30 °C/min and (c) 10 °C/min.

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