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Structural and compositional inhomogeneities in Bismuth-Substituted rare earth iron garnet epitaxial films

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ABSTRACT

Structural and compositional inhomogeneities in the free-standing epitaxial films of rare earth iron garnet films are characterized using high-resolution X-ray diffraction and energy dispersive X-ray fluorescence. These analyses show a variation in the lattice parameters as well as in chemical compositions, displaying inconsistency from the top and the bottom of the film, suggesting an inhomogeneous distribution of rare earth content. With these variations, intricate changes often occur in the magneto-optics properties. While with conventional X-ray diffraction techniques, the films appeared to be free of gross defects, our high-resolution rocking curves reveal variations in defect density and multiple lattice parameters. These results suggest that epitaxial films presumed to be homogeneous based on conventional X-ray diffractometry, may not be so. Therefore, films found to exhibit exotic physical properties should be carefully examined for the presence of structural and compositional anomalies to optimize the magneto-optic response. If such defects are present, the experimental results may be compromised due to sample imperfections, rather than intrinsic phenomena.

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1. Introduction

Recently, there has been extensive research in magneto-optic (MO) devices based on Bismuth-doped rare earth iron garnets (Bi:RIGs) to exploit the material's large Faraday rotation [1–7]. Much work has been done to optimize compositional variation to optimize MO properties and the MO devices. While Bi:RIG materials have a large Faraday rotation response, they can be made only in thin-film or thick-film form [8]. Hence, arrays of Bi:RIG films were stacked to maximize the Faraday rotation for a high sensitivity magnetic field sensor, enabling a minimum detectable magnetic field of 1.2 $pT/Hz^{1/2}$. It has been reported that diamagnetic substitution of Fe with Ga results in an enhancement of MO responsivity at higher magnetic field whereas the responsivity under a weak field is in general unaffected [9]. In these studies, the Bi:RIG crystals were grown by liquid phase epitaxy (LPE) on (1 0 0) gadolinium gallium garnet substrates, with compositions based on the formula $(BiGdLu)_3Fe_{5-x}Ga_xO_{12}$, where the gallium content level x varied between 1.0 and 1.35. Lutetium and bismuth substitutions result in increased Curie temperature, increased lattice parameter, and improved MO responsivity. The films were cleaved from their substrates to form free standing films, and diced into 2 mm squares using a laser dicing machine. The thicknesses of these films are 180–550 μm. Because of the compositional design of Faraday rotator materials for applications in multicomponent oxide films for electronics, it is important to determine the structural homogeneity and compositional variation in the film. When a stacked array of films is arranged to maximize the Faraday rotation, adverse effects of structural inhomogeneity also increase with the number of stacked films, affecting the final outcome and compromising the detection sensitivity. In this paper, we report on the structural and compositional variations on the state-of-the-art LPE grown Bi:RIG films as determined by high-resolution X-ray diffraction and energy dispersive X-ray fluorescence.

2. Experimental

The high-resolution X-ray set-up consists of an 18 kW rotating anode with a four-circle diffractometer with a Cu $K\alpha_1$ obtained from two channel-cut Ge (2 2 0) crystals on the incident beam optics and a receiving slit/analyzer Ge(2 2 0) crystal in the

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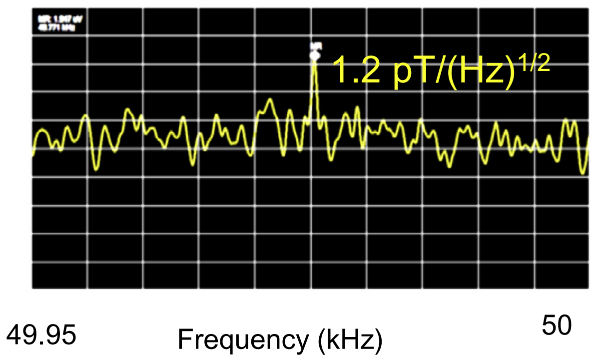

Fig. 1. A weak magnetic field (<8 pT) detected with our magneto-optic field sensor. This frequency spectrum with a bandwidth of 50 Hz shows the ac magnetic field signal at 49.975 kHz.

diffracted beam optics. This offers the best combination of counts to resolution for accurate rocking curve and two-theta measurements for obtaining lattice parameters. X-ray topography was performed on a double crystal set-up described elsewhere [10]. Magnetization measurements were performed using a vibrating sample magnetometer at temperatures between −150 °C and 300 °C and a PPMS magnetometer (Quantum Design) in the range of temperature between 2 and 250 K. The MO response of sample was measured using an ellipsometric transmission method, as described in Ref. [7]. We send a linearly polarized laser beam (probe beam) perpendicular to the sample surface to which an external magnetic field is applied. Due to Faraday effect, the polarization of probe beam rotates at an angle $\phi = Vd\boldsymbol{B} \cdot \boldsymbol{k}$. Here V is the Verdet constant, d is the MO sample thickness, $\boldsymbol{B}$ is the vector magnetic field, and $\boldsymbol{k}$ is the direction of the laser probe beam. The polarization rotation angle is measured using a cross polarization method. The same principle is used for our MO sensor. Apparently, a thicker MO material yields a larger polarization rotation and hence a higher sensitivity. So we stacked MO thick films to increase the effective thickness d. However, the probe beam attenuation increases with the number of stacked MO films. The optimum number of stacked films depends on the film properties, such as optical transmittance and magnetic domain patterns. The result shown in Fig. 1 was obtained with an MO sensor with 12 stacked films.

3. Results and discussion

Fig. 2a shows the magnetization M as a function of H-field taken from a 2 mm × 2mm film sample, revealing that our sample has an in-plane easy axis parallel to the surface. Fig. 2b and c show the temperature dependence of magnetization M, exhibiting the compensation temperatures and Curie temperatures for two different

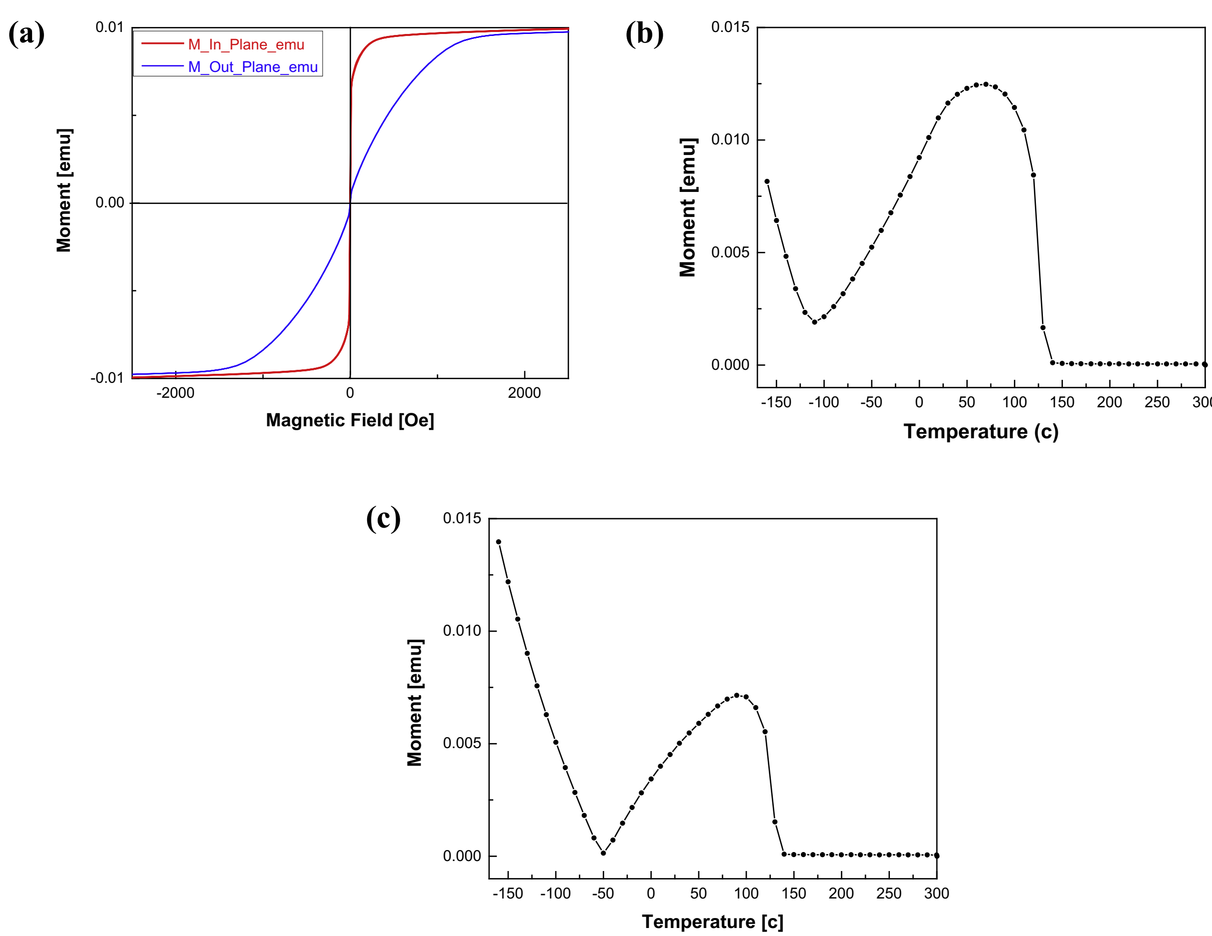

Fig. 2. (a). Magnetization both in-plane and out-of-plane as a function of magnetic field H measured from our $(BiGdLu)_3Fe_{5-x}Ga_xO_{12}$ thick film at room temperature. (b,c) The temperature dependence of magnetization for two different pieces from the same film of $(BiGdLu)_3Fe_{5-x}Ga_xO_{12}$ showing different compensation temperatures although the Curie temperatures appear to be the same.