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Systematic control of stress-induced anisotropy in pseudomorphic iron garnet thin films

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ABSTRACT

Iron garnets are one of the most well-studied magnetic materials that enabled magnetic bubble memories and magneto-optical devices employing films with a perpendicular easy axis. However, most studies have been conducted on rather thick films (>1 μm), and it has not been elucidated whether it is possible to align the magnetic easy axis perpendicular to the film plane for much thinner (<100 nm) films by overcoming shape anisotropy. We studied the effects of epitaxial strain and film composition on the magnetic properties of 50-nm-thick garnet thin films grown by pulsed-laser deposition. $Y_3Fe_5O_{12}$ was selected as the most prototypical garnet and $Sm_{3-x}Tm_xFe_5O_{12}$ ($x=1, 2, 3$) was selected in view of its negatively large magnetostriction constants. We employed (111) planes of single crystalline $Gd_3Ga_5O_{12}$ and $(CaGd)_3(MgGaZr)_5O_{12}$ substrates to tune the epitaxial strain. Thin films with a pseudomorphic structure were fabricated with the in-plane strain ($\varepsilon_{//}$) ranging from -1.5% to $+0.5\%$, corresponding to the stress-induced anisotropy field (H_A) ranging from -40 kOe to $+25$ kOe, respectively. The magnetization ratio of the out-of-plane to in-plane component ($M_\perp/M_{//}$) systematically varied in accord with H_A, yielding $M_\perp/M_{//}>1$ for thin films with H_A values larger than 20 kOe. Among the films grown, $Tm_3Fe_5O_{12}$ on $Gd_3Ga_5O_{12}$ showed the largest $\varepsilon_{//}$ and H_A values of $+0.5\%$ and $+25$ kOe, respectively, to realize an apparently perpendicular easy axis, confirmed by a large $M_\perp/M_{//}$ value of 7.8. Further, magnetic force microscope images showed a maze pattern typical of a perpendicularly magnetized film. These results reveal a method for tailoring the magnetic anisotropy of garnet ultrathin films by utilizing epitaxial strain. These thin films may be utilized to obtain nanoscale magnetic bubbles for use in novel devices.

1. Introduction

The structure–property relationship for magnetic iron garnets has been extensively studied decades ago [1] for such applications as magnetic bubble memories [2–5] and magneto-optical devices [6]. The control of magnetic anisotropy has been one of the key challenges for such applications. Garnets belong to the space group $Ia3d$ (O_h^{10}) and have a $\langle 111 \rangle$ magnetic easy axis in bulk crystals determined by crystallographic anisotropy while they do not exhibit any uniaxial magnetic anisotropy due to their symmetry. However, for the aforementioned applications, a perpendicular easy axis is desirable. For achieving this, uniaxial magnetic anisotropy perpendicular to the plane of (111)-oriented films is realized by overcoming shape anisotropy. In studies on relatively thick (>1 μm) garnet films grown by chemical vapor deposition, stress-induced anisotropy originating predominantly from a thermal expansion mismatch between the substrate and film has been utilized to realize perpendicular magnetization [7]. More recently, the liquid phase epitaxy (LPE) technique has been established as a useful technique for realizing perpendicular magnetization due to growth-induced anisotropy [8] and has become the most reliable manufacturing technique for practical devices. However, the uniaxial magnetic anisotropy field afforded to LPE-grown films is not sufficiently large and it is possible to realize a perpendicular easy axis only for relatively thick films for which shape anisotropy is not very large (a quantitative discussion is given in Section 3 B).

Another way to realize a large uniaxial magnetic anisotropy field for magnetic thin films is by utilizing the epitaxial strain. For such films, stress-induced anisotropy due to lattice mismatch has

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been utilized to control the easy axis. For example, the magnetic easy axis tends to be perpendicular to the film plane under a tensile epitaxial strain in (GaMn)As [9], (InMn)As [10], and (GaMn)(PN) [11]. In contrast, a compressive epitaxial strain causes the magnetic easy axis to be perpendicular to the film plane in $La_{0.7}Sr_{0.3}MnO_3$ [12,13] and $SrRuO_3$ [14]. However, the effects of epitaxial strain in garnets have not been studied systematically so far, partly because the research fields of interest require relatively thick films and it is difficult to obtain coherently strained pseudomorphic films owing to lattice relaxation in thick films [15].

Recently, pulsed-laser deposition (PLD) has been employed to fabricate relatively thin (<1 μm) garnet films, heterostructures, and multilayers for applications such as microwave devices [16], integration with semiconductor platforms [17], magnetophotonic crystals [18], and magnetoelectric functionalities [19]. The thickness controllability achieved by PLD surpasses that achieved by LPE; however, the fabrication of thin films with bulk-like properties is difficult. Kahl et al. reported that high-frequency laser repetition can substantially facilitate the growth of highly crystalline epitaxial films [20]. Further, Krockenberger et al. prepared films with saturation magnetization and a Curie temperature (TC) similar to those for LPE-grown bulk crystals by adopting higher repetition rates [21]. However, perpendicular magnetization for very thin (<100 nm) epitaxial films has not been achieved yet [16,20,22,23].

The motivation for the fabrication of very thin films with a perpendicular easy axis is to explore the possibility of manipulating nanoscale magnetic bubbles by means of an electric field [24]. The size of the magnetic domain W depends on the film thickness t as given below (p. 138 of Ref. [1]):

$$W = 2.72\sqrt{\frac{\sigma_w t}{4\pi M^2}}, \quad (1)$$

where, σ_w and M are the domain wall energy and spontaneous magnetization, respectively. For obtaining nanoscale bubbles, films with nanoscale thickness are required. A few studies have reported that the magnetic domain wall in a garnet could be moved by the application of a local electric field through a needle biased with a high voltage (>500 V) [24]. However, these studies were examined only for bulk crystals and thick films. The motion of the domain wall was confirmed by magneto-optical microscope that was possible due to the large domain size. In order to obtain nanoscale domain size for ultrathin films, the strong shape anisotropy in these films has to be overcome. If this challenge is achieved, it may become possible to design and realize new type of devices in which the magnetic domain could be controlled by applying relatively low voltages.

In this paper, we describe a systematic study of the effect of epitaxial strain on magnetic anisotropy. First, we describe the effect of epitaxial strain on the magnetic properties of 50-nm-thick $Y_3Fe_5O_{12}$ films grown on $Gd_3Ga_5O_{12}$ (GGG) and $(CaGd)_3(MgGaZr)_5O_{12}$ (SGGG) substrates with nearly matched and 0.98% larger lattice constants, respectively. We intentionally modulated the lattice constant of $Y_3Fe_5O_{12}$ by modifying the film growth conditions, namely, oxygen pressure (P_{O_2}). It has been reported and we too speculate that Fe deficiency increases the lattice constant by about 1%. Magnetic anisotropy scales well with in-plane strain and the resultant stress-induced anisotropy field (H_A). However, perpendicular magnetization could not be achieved in the case of $Y_3Fe_5O_{12}$. On the basis of literature data for other garnets, we selected substituted $C_{3-x}C'_xFe_5O_{12}$ garnets where C and C' are Y, Sm, Eu, Tm, and Lu as possible candidates for realizing large H_A values necessary for perpendicular magnetization. We estimated the value of H_A by considering coherently strained pseudomorphic structures on both GGG and SGGG substrates and selected $Sm_{3-x}Tm_xFe_5O_{12}$ ($x=1, 2, 3$) as the most promising system. Systematic experiments revealed that $Tm_3Fe_5O_{12}$ on a GGG substrate exhibits magnetization hysteresis that supports the perpendicular easy axis. Indeed, magnetic force microscope (MFM) images indicate a maze pattern typical of a perpendicularly magnetized film with a submicron domain width.

2. Materials and methods

Approximately 50-nm-thick $Y_3Fe_5O_{12}$ and $Sm_{3-x}Tm_xFe_5O_{12}$ thin films were grown on the (111) planes of GGG and SGGG substrates (Saint-Gobain K. K.) by the PLD method. The lattice constants for stoichiometric $Y_3Fe_5O_{12}$, GGG, and SGGG are 1.2376 nm [25], 1.2383 nm, and 1.2497 nm, respectively. By referring to phase diagrams [26,27], single-phase ceramic targets for stoichiometric $Y_3Fe_5O_{12}$ and $Sm_{3-x}Tm_xFe_5O_{12}$ ($x=1, 2, 3$) were prepared from prescribed Fe_2O_3 (4N), Y_2O_3 (4N), Sm_2O_3 (3N), and Tm_2O_3 (3N) powders (Kojundo Chemical Laboratory Co., Ltd.) by a conventional solid-state reaction. Mixed powders were calcined, pressed into pellets, and sintered for 12 h at 1200 °C for $Y_3Fe_5O_{12}$ and at 1300 °C for $Sm_{3-x}Tm_xFe_5O_{12}$, yielding green-colored targets. Focused KrF excimer laser pulses with a wavelength of 248 nm were impinged on the target placed 40 mm away from the substrate at a frequency of 80 Hz and with a fluence of 0.6 J/cm^2. Such a high laser repetition is crucial for improving the crystallinity of the films [20,21]. Films were grown at a temperature of 800 °C with P_{O_2} ranging from 0.1 to 1 Torr. The substrate temperature was monitored using an optical pyrometer. The typical growth rate was approximately 0.002 nm/pulse (0.16 nm/s). The crystallinity and lattice parameters were examined by high-resolution X-ray diffraction (XRD) for symmetric diffraction and reciprocal space mapping (RSM). The film thickness was evaluated from Laue fringes appearing near the (444) peak in the XRD patterns. The magnetic hysteresis loop was measured using an MPMS SQUID VSM system (Quantum Design, Inc.) equipped with a vibrating sample magnetometer and a superconducting quantum interference device. The magnetic domain structure of a perpendicular easy axis film was evaluated by MFM (Asylum Research MFP-3DTM).

3. Results and discussion

3.1. Crystal structure of $Y_3Fe_5O_{12}$ films

Fig. 1 shows a series of XRD patterns for $Y_3Fe_5O_{12}$ films grown on the (111) planes of (a) GGG and (b) SGGG substrates at various P_{O_2} values. Peaks assignable to (444) diffraction by the films were clearly observed, as indicated by arrows. These films exhibited full width at half maximum values as small as 18 arcsec in X-ray rocking curves for (444) diffraction (not shown), which is close to the values for the substrate and instrument resolution. The peaks monotonically shifted to lower angles with decreasing P_{O_2}. Owing to the close lattice matching between $Y_3Fe_5O_{12}$ and GGG, an ideal heterostructure should result in an overlap of the peaks as observed for the film grown at $P_{O_2}=0.6$ Torr on GGG. The origin of peak shift can be explained by lattice expansion due to non-stoichiometry caused by the decrease in P_{O_2}. This result is consistent with the tendency reported previously for non-stoichiometric polycrystalline $Y_3Fe_5O_{12}$ films [23]. It has been reported that the lattice constant of a film grown by PLD is greater than that of a target due to Fe deficiency [16]. Representative RSM data around the (486) diffraction for films grown at $P_{O_2}=0.4$ Torr on GGG and SGGG are shown in Fig. 1(c) and (d), respectively. RSM results indicate that all $Y_3Fe_5O_{12}$ films in this study had a