Journal of Magnetism and Magnetic Materials 323 (2011) 3143–3146

Contents lists available at ScienceDirect

Journal of Magnetism and Magnetic Materials

journal homepage: www.elsevier.com/locate/jmmm

Epitaxial strain-induced magnetic anisotropy in $Sm_3Fe_5O_{12}$ thin films grown by pulsed laser deposition

H. Yamahara [a,*], M. Mikami [b], M. Seki [c], H. Tabata [a,c]

[a] Department of Bioengineering, Graduate School of Engineering, University of Tokyo, 7-3-1 Hongo, Bunkyo-ku, Tokyo 113-8656, Japan
[b] Department of Materials Engineering Science, Graduate School of Engineering Science, Osaka University, 1-3 Machikaneyama-cho, Toyonaka, Osaka 560-8531, Japan
[c] Department of Electrical Engineering and Information Systems, Graduate School of Engineering, University of Tokyo, 7-3-1 Hongo, Bunkyo-ku, Tokyo 113-8656, Japan

ARTICLE INFO

Article history:
Received 29 April 2011
Received in revised form
23 June 2011
Available online 7 July 2011

Keywords:
Samarium iron garnet
Pulsed laser deposition (PLD)
Epitaxial
Thin film
Magnetic anisotropy
Lattice strain

ABSTRACT

$Sm_3Fe_5O_{12}$ thin films of various thicknesses were grown on a (0 0 1)-oriented $Gd_3Ga_5O_{12}$ substrate by pulsed laser deposition. The crystal structure of the films was strongly dependent on film thickness. The lattice was strained for thinner films due to a lattice mismatch between the film and substrate. This lattice strain was relaxed when the film thickness exceeded a critical thickness of around 660 Å. It is suggested that the epitaxial strain induces uniaxial magnetic anisotropy with an out-of-plane magnetic easy axis.

1. Introduction

Over the past decades, much attention has been paid to the magneto-optical (MO) properties of ferrimagnetic garnets from the perspective of their practical applications mainly in the field of optical communications. High-quality thin films of rare earth iron garnet (RIG) have been fabricated using various film growth techniques including liquid phase epitaxy [1], radio frequency (rf-) magnetron sputtering [2], and pulsed laser deposition (PLD) [3]. The crystal structure of RIG is cubic in the normal bulk form [4,5]. However, it is well known that the crystal structure is modified in single-crystalline thin films due to epitaxial strain originating from a lattice mismatch between the film and substrate [6]. These strained films are expected to have properties that differ from those of films without strain. For example, a shift of the magnetic easy axis [6–9] and coercivity [10] have been reported for strained magnetic thin films. Modification of magnetic properties through the epitaxial strain is quite useful for magneto-optical device applications such as magneto-optical memories, isolators, and sensors.

In this study, we investigated the MO properties of thin films of samarium iron garnet (SmIG; $a=12.530$ Å) deposited on gadolinium gallium garnet (GGG; $a=12.383$ Å) substrates using PLD. The lattice constant of SmIG is larger than that of GGG with a lattice misfit of 1.19% between the film and the substrate, which should cause compressive strain in the film.

2. Experimental

Thin films of SmIG were deposited on (0 0 1)-oriented GGG substrates using PLD. The target samples were prepared by a conventional solid state reaction. Sm_2O_3 (99.9%) and Fe_2O_3 (99.99%) powders were mixed stoichiometrically and sintered at 1200 °C for 12 h. During the PLD process, the substrate temperature and ambient oxygen pressure were maintained at 700 °C and ~0.1–1 Pa, respectively. The thickness of the prepared films was in the range ~200–4300 Å.

The crystal structures of the films were investigated using a standard X-ray diffraction (XRD) system with monochromated Cu-K_α radiation. The magneto-optical properties were investigated using magneto circular dichroism (MCD). For MCD spectra measurements, a magnetic field of 1 T was applied perpendicular to the film surface. The magnetic field dependence of MCD was measured at an excitation photon energy of around 2.88–2.99 eV. All of the MCD measurements were carried out at room temperature.

3. Result and discussion

In the XRD (θ–2θ) patterns of SmIG on GGG, (0 0 4) and (0 0 8) reflection peaks were observed for all of the film thicknesses, suggesting that the SmIG films were grown with a (0 0 1) orientation normal to the substrate surface. Fig. 1 shows the reciprocal space mapping (RSM) around the (2 4 8) plane for films with different thicknesses. This indicates that the SmIG thin films were epitaxially

* Corresponding author. Tel.: +81358418846; fax: +81358418846.
E-mail address: tt107446@mail.ecc.u-tokyo.ac.jp (H. Yamahara).

0304-8853/$ - see front matter © 2011 Elsevier B.V. All rights reserved.
doi:10.1016/j.jmmm.2011.06.074

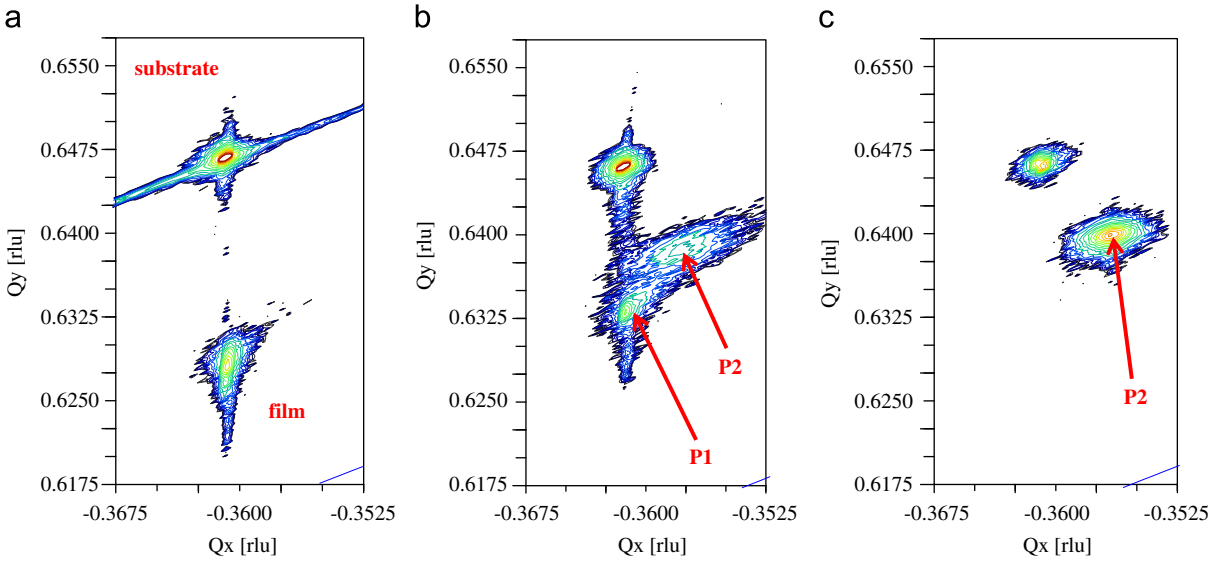

Fig. 1. Reciprocal space mapping around the (2 4 8) plane for SmIG thin films with thicknesses of (a) 580, (b) 820, and (c) 3660 Å. P1 and P2 are the strained and cubic phases, respectively (see text).

grown on the GGG substrate. At a film thickness of 580 Å, the in-plane lattice constant of the film was almost equal to that of the substrate, while the out-of-plane lattice constant of the film was larger than that of the substrate. This indicates that the lattice of the thinner SmIG film is elongated perpendicular to the film surface due to the epitaxial strain. The tetragonality (the ratio of the *a*- and *c*-axis; *c*/*a*) of the film is 1.028 for the film with a thickness of 580 Å. When the film thickness was increased to 820 Å, the value of the in-plane reciprocal lattice vector (Q_x) of the SmIG did not match that of GGG and the tetragonality of SmIG decreased. Furthermore, another phase with an almost cubic structure (tetragonality: 1.002) was also found. These results indicate the partial relaxation of the epitaxial strain. For thicker films, the strain was fully relaxed, as shown in Fig. 1(c). The thickness dependence of the *c*-axis length of the SmIG films is shown in Fig. 2. In the range 200–2200 Å, the *c*-axis length of the strained phase (P1) decreased with increasing film thickness, indicating the relaxation of stress in the film from the substrate. In contrast, the *c*-axis length of the relaxed phase (P2) was larger than that of the P1 phase and was almost independent of film thickness. The larger lattice length in the thicker films is probably due to the oxygen vacancies. In oxide materials, it is known that oxygen vacancies cause lattice expansion [11]. To confirm the influence of oxygen vacancies, the prepared films were post-annealed at 800 °C in air for 10 h. A significant decrease in the *c*-axis length after post-annealing, especially at 3300 and 4300 Å, was observed only for the P2 phase, implying the contribution of oxygen vacancies. In contrast, the *c*-length of the P1 phase did not change after annealing. This indicates that the lattice of the film was bound to the substrate and was therefore not affected by oxygen vacancies. In the garnet films, the cracks do not propagate when the film thickness is smaller than the critical film thickness (h_c) [12], which is expressed as follows:

$$h_c = \frac{a_0(1-\nu)^2}{5\pi f^2}$$

where a_0 is the interatomic distance, ν is the Poisson ratio, and f is the misfit between the film and the substrate. Using the parameters $a_0 = 3$ Å and $\nu = 0.30$ for general rare earth garnets [13] and $f = 0.0119$ (calculated from $a_f = 12.530$ Å and $a_s = 12.383$ Å), a value of 660 Å is obtained for h_c. This value explains why the peaks at around 740 Å began to separate drastically in the range ~580–740 Å in the XRD results.

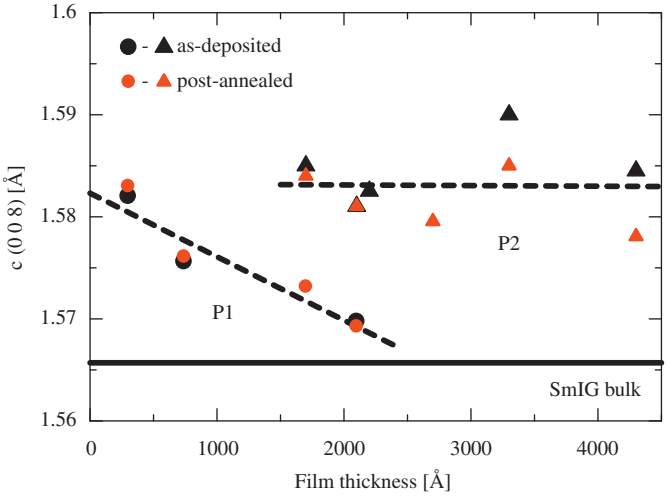

Fig. 2. Dependence of *c*-axis length on the thickness of SmIG films. The circular and triangular symbols indicate the strained (P1) and cubic (P2) phases, respectively.

Fig. 3(a) shows the MCD spectra for the SmIG films with various thicknesses. An external magnetic field of 1 T was applied perpendicular to the film surface. It is well known that RIG shows strong optical absorption in the region $E > 2.9$ eV [14]. Decrease in MCD signal approximately above 3.2 eV observed in the 4300 Å thick film was attributed to the decrease in optical transmission. *M*–*H* loops were measured with a wavelength of 415–430 nm, which corresponds to a photon energy equivalent to the excitation energy for the $O^{2-}(2p) \rightarrow Fe^{3+}(3d)$ charge transfer energy peaks [15]. As shown in Fig. 3(b), all of the samples exhibited clear hysteresis loops and the shapes of the *M*–*H* loops changed depending on film thickness. To evaluate the magnetic anisotropy, the change in the squareness, which is defined as the value of remnant magnetization (M_r) divided by the saturated magnetization (M_s) with the variation of the film thickness, is shown in Fig. 4. When the thickness was within 200–300 Å (below h_c), the squareness showed high values around 0.93. This indicates the presence of an out-of-plane easy axis of magnetization. However, when the thickness exceeded h_c, the squareness