Journal of Magnetism and Magnetic Materials 342 (2013) 134–138

Contents lists available at SciVerse ScienceDirect

Journal of Magnetism and Magnetic Materials

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

Microstructure and magnetic properties of La–Co substituted strontium hexaferrite films prepared by pulsed laser deposition

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ARTICLE INFO

Article history:
Received 4 November 2012
Received in revised form
16 March 2013
Available online 28 April 2013

Keywords:
Pulsed laser deposition
Strontium hexaferrite film
La–Co substitution
Perpendicular magnetic anisotropy

ABSTRACT

Microstructure and magnetic properties of La–Co substituted strontium hexaferrite films ($Sr_{1-x}La_xFe_{12-x}Co_xO_{19}$) fabricated by pulsed laser deposition on Si(100)/Pt(111) substrate were investigated. The coercivities of the films in perpendicular direction were higher than those in in-plane direction which confirms the perpendicular magnetic anisotropy of the films. Atomic force microscopy images of the films revealed decreasing of the plate-like grains size, from 300 to 110 nm with increasing the La–Co contents. The saturation magnetization increased slightly till $x=0.2$ and then decreased from $x=0.2$ to $x=0.4$. However, the coercivity increased from 2.3 kOe for the $SrFe_{12}O_{19}$ film to 4.1 kOe for the $Sr_{0.6}La_{0.4}Fe_{11.6}Co_{0.4}O_{19}$ film, because of the decrease of the grain size and increase of the magnetic anisotropy field.

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1. Introduction

Recently, permanent magnetic films have been widely studied for the application in microelectrical mechanical systems (MEMS). $Nd_2Fe_{14}B$ and $SmCo_5$ thin films are the most promising materials to be used to miniaturize the system on account of their high energy product [1]. However, poor corrosion resistance against humidity limits their application in some circumstances. The virtues of barium and strontium hexaferrite films derived from their corrosion resistance and acceptable magnetic properties have drawn much attention of the designing engineers. Furthermore, they were also used as microwave materials [2] and perpendicular recording media [3] due to their relatively high uniaxial magnetocrystalline anisotropy and moderate saturation magnetization. On the other hand, the magnetic performance of the M-type hexaferrites must be improved, e.g. through ion substitution techniques for various applications. It was reported that the saturation magnetization as well as the magnetocrystalline anisotropy constant of hexaferrite particles increased and the value of dH_c/dT also reduced by the La–Co substitution, which are needed for high density magnetic recording media [4].

The $SrFe_{12}O_{19}$ (SrM) films have been prepared by various methods such as sputtering [5], pulsed laser deposition [6], metallorganic chemical vapor deposition [7] and sol–gel method [8]. However, pulsed laser deposition technique is suitable for growing the films with complex compound and structure, due to its ability to transfer the stoichiometry of complex multi-elemental targets to different substrates. This unique feature has been successfully used to deposit Co/Ti [9], Sc [10] and Al [11] substituted hexaferrite films.

In this investigation, we report a study of the microstructure and magnetic properties of $Sr_{1-x}La_xFe_{12-x}Co_xO_{19}$ ($x=0$–0.4) thin films pulse laser deposited on the commercial $Si(100)/SiO_2/Ti/Pt(111)$ substrate.

2. Experimental procedure

The $Sr_{1-x}La_xFe_{12-x}Co_xO_{19}$ ($x=0$–0.4) targets were prepared by the sol–gel technique. Lanthanum nitrate, cobalt nitrate, iron nitrate, strontium nitrate and citric acid were dissolved in the distilled water at which the molar ratio of citric acid to the total metallic ions was 1:1 [12]. After homogenization, the pH was adjusted to 7 with ammonia under continuous stirring. The final mixture was slowly evaporated at 80 °C until a highly viscous gel was formed. The resulted gel was heated to 200 °C till ignited in a self-propagating process. The final residue was calcined at 1000 °C for 2 h. Powders were pressed into pellets and sintered in air at 1200 °C for 2 h, followed by cooling in the furnace. The resulting target was homogeneous and single phase with a density greater than 90% with respect to the theoretical amount.

Films were deposited on the commercial $Si/SiO_2/Ti$ (400 nm)/Pt (400 nm) substrate by focusing 260 mJ, 23 ns pulses from a KrF

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http://dx.doi.org/10.1016/j.jmmm.2013.04.066

excimer laser (COMPex 200 Fluorine from Lambda Physik) with a wavelength of 248 nm onto the target to give a fluence of 3 J/cm^2 per pulse at 5 Hz. The laser beam was incident on the target at an angle of 45° from the target and the substrate was set at a distance of 4 cm from the target. The oxygen pressure and substrate temperature were kept in 0.13 mbar at 700 °C [13]. By deposition for 30 min the films were produced with thickness of 200 nm, according to the cross sectional scanning electron microscope (SEM) micrographs. The as-deposited film was cooled down slowly to room temperature at 1 atm O_2 pressure.

The crystal structure of the films was checked by X-ray diffraction (XRD) in θ–2θ geometry, using CuKα radiation (Philips PW-1730). *c*-axis lattice parameter was also obtained by fitting the most significant (00*l*) diffraction peaks to Bragg's law. The thickness was determined using a SEM (CamScan MV2300).

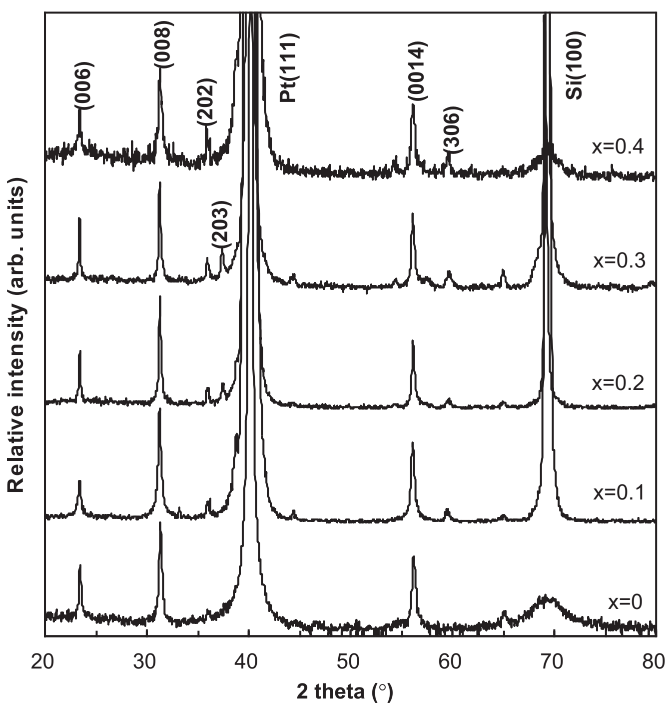

Fig. 1. XRD patterns of the $Sr_{1-x}La_xFe_{12-x}Co_xO_{19}$ films with various compositions.

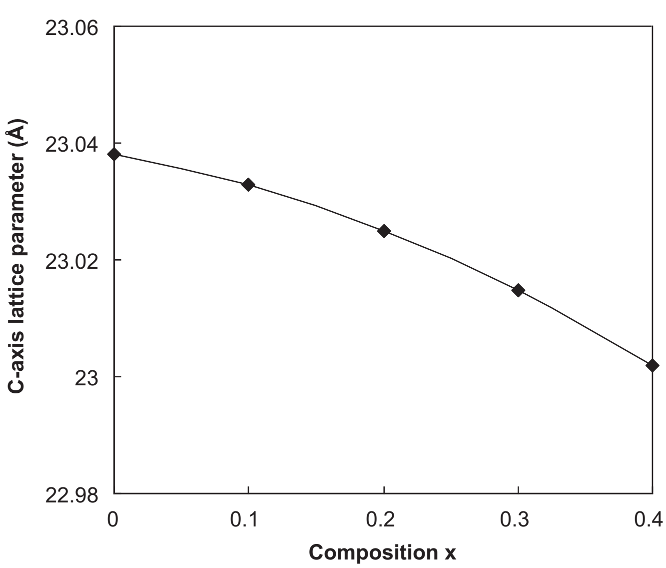

Fig. 2. Variation of *c*-axis lattice parameter as a function of composition (*x*).

Atomic/magnetic force microscope (AFM/MFM) (Veeco Dimension 3100) was applied to image the topography and magnetic structure of the films. The average grain size of the films was determined by lineal intercept method. Magnetic properties were also measured by a vibrating sample magnetometer (Lakeshore 7400) at a maximum applied field of 12 kOe.

3. Results and discussion

Fig. 1 shows the XRD patterns of the La–Co substituted SrM films deposited on the Si(100)/Pt(111) substrate. The only pure strontium hexaferrite phase was formed in all samples, and the impurity phases were not observed. Furthermore, strong (00*l*) basal reflections confirm that the films are aligned with the *c*-axis perpendicular to the film plane. However, the other nonbasal reflections such as (202), (203) and (306) are also observed in the patterns of the substituted films. The *c*-axis growth of SrM film is due to the small mismatch between SrM(001) and Pt(111) planes [14,15]. The *c*-axis lattice parameter versus the La–Co additions is shown in Fig. 2. The *c*-axis length decreased with La–Co additions due to the substitution of Sr^{2+} (1.17 Å) by La^{3+} (1.13 Å) with the smaller radius, while the substitution of Fe^{3+} by Co^{2+} did not affect the length of *c*-axis because of the similar radius of Co^{2+} (0.65 Å) and Fe^{3+} (0.69 Å) [16].

AFM and MFM were used to evaluate the grain size, surface morphology and magnetic structure. AFM and MFM images of the $SrFe_{12}O_{19}$ and $Sr_{0.8}La_{0.2}Fe_{11.8}Co_{0.2}O_{19}$ films and AFM micrographs of the $Sr_{1-x}La_xFe_{12-x}Co_xO_{19}$ films as a function of composition are shown in Figs. 3 and 4, respectively. The figures showed the plate-like hexagonal shaped grains in the $SrFe_{12}O_{19}$ film, related to the growth of the *c*-axis perpendicular to the film plane and some grains deviated from the *c*-axis perpendicular orientation in the substituted films in which their hexagonal shape have not been seen completely, in consistence with XRD results. Compared to the unsubstituted film, the average grain size becomes smaller in the range of 300–110 nm with the increase of La–Co additions (Fig. 4 and Table 1). The La–Co substitutions can cause lattice distortion and internal stress which hinder the grain growth. Root mean square (RMS) surface roughness of the films is in the range of 20–50 nm which increases with La–Co additions.

Fig. 3 also compares MFM images of the $SrFe_{12}O_{19}$ and $Sr_{0.8}La_{0.2}Fe_{11.8}Co_{0.2}O_{19}$ films. The bright and dark contrasts are representative of a domain pattern with different magnetic orientation along the out of plane direction, because the MFM data correspond to the out of plane magnetization component only. The figures showed that the domain structure of the $Sr_{0.8}La_{0.2}Fe_{11.8}Co_{0.2}O_{19}$ films is finer than that of the $SrFe_{12}O_{19}$ film, due to the smaller grains [17].

Out of plane and in-plane magnetic hysteresis loops for the $SrFe_{12}O_{19}$ and $Sr_{0.6}La_{0.4}Fe_{11.6}Co_{0.4}O_{19}$ films are presented in Fig. 5. Higher coercivities in perpendicular direction rather than in-plane direction are due to the *c*-axis perpendicular orientation. The easy loops also seem to be sheared due to the effects of the film demagnetizing fields. Furthermore, observation of a wider in-plane magnetization curve of the $Sr_{0.6}La_{0.4}Fe_{11.6}Co_{0.4}O_{19}$ film (Fig. 5b) in comparison with that of the $SrFe_{12}O_{19}$ film (Fig. 5a), indicates higher deviation from the perpendicular *c*-axis orientation as shown in Fig. 1.

It is well known that in a unit cell of M-type hexaferrite, there are five distinct crystallographic sites or sublattices for the cations, three octahedral (2a, 12k, $4f_2$), one tetrahedral ($4f_1$) and one trigonal bipyramid (2b). The spin directions for the 2a, 12k and 2b sublattices are parallel to each other (in the direction of the crystallographic *c*-axis), whereas those of $4f_1$ and $4f_2$ sublattices are in the opposite direction, which are coupled by superexchange interactions through the O^{2-} ions [18]. Consequently, the magnetic properties of the substituted M-type strontium hexaferrite are strongly dependent on

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