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Thickness dependence of magnetic properties in La–Co substituted strontium hexaferrite films with perpendicular anisotropy

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ABSTRACT

The thickness dependence of magnetization reversal and coercivity behavior for La–Co substituted strontium hexaferrite (Sr-*M*) films was investigated. It is found that perpendicular anisotropy appears only when film thickness (t) is above 110 nm. With increasing t, perpendicular anisotropy energy ($K_{u\perp}$) increases gradually to its maximum of 1.76×10^6 erg/cm^3 at t=300 nm, but turns to decrease when $t > 300$ nm. Moreover, when $t > 110$ nm, those films exhibit domains pinning or Stoner–Wohlfarth reversal model, present large $K_{u\perp}$ values and a rapid increase in $H_{c\perp}$. However, while $t \leq 110$ nm, Sr-*M* films show nucleation model of magnetization reversal and perform low coercivity. The origin of the coercivity varying with thickness should be correlated with the grain size and preferred orientations in Sr-*M* films.
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1. Introduction

Magnetic thin films with high coercivity have been intensively investigated, in order to expand their applications including magnetic recording, magneto-optical recording, and microelectromechanical system devices. Among them, $Nd_2Fe_{14}B$, $SmCo_5$ and FePt 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 and high cost 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.

Recently, La–Co pair substituted strontium hexaferrite film (Sr-*M*) with perpendicular magnetization is confirmed to an effective way to enhance the intrinsic coercive force [2–4]. Moreover, the coercivity (H_c) increases with decreasing particle size (D) and turns to decrease as D decreases below single domain size, according to the thermal fluctuation of spins [5]. The single domain size for Sr-*M* is 0.5–1.0 μm [6]. It has also been reported that coercivity depends on the film thickness in the submicron range based on the physical vapor deposition processes [7,8]. Nevertheless, the strong strain possibly caused at the interface between substrate and films with a columnar structure grown *in-situ* in low ambient oxygen partial pressure limits the application of the physical vapor deposition. Experiment evidences on the research in the submicron range of Sr-*M* films have been not studied in detail. Therefore, experimental investigation of the film properties with thickness is necessary to determine due to the variation in microstructure and magnetic properties of the films.

In this work, a series of La–Co substituted strontium hexaferrite films with different thickness from 85 to 400 nm in submicroscale bulk system have been fabricated in a fixed process. Optimized radio frequency (RF) magnetron sputtering and post annealing has been explored to minimize the side effect of film growth, particularly previously mentioned lattice strain effect. The relationship between perpendicular magnetic characteristics and layer thickness has been studied.

2. Materials and methods

La–Co pair doped Sr-*M* films were deposited onto amorphous quartz glass substrates by RF magnetron sputtering system (SPF-430H) using a fixed La–Sr–Co–Fe–O target with a purity of 99.99%.

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The base pressure of sputtering chamber was 2.5×10^{-3} Pa and the sputtering power was 200 W. Substrate temperature (T_s), for the hexaferrite layers was set *in situ* substrate heating (400 °C), of which the growth rate was 4.79 nm/min. After deposition, these films were followed by a post-deposition annealing (T_a) 900 °C for 1 h in ceramic fibre muffle furnace (TM-0614P). The deposited film thickness was measured by a surface profile step instrument (KLA TENCOR P16+). The structure and crystallographic characteristics were analyzed by D8 advanced X-ray diffracto-meter (XRD, X'Pert Pro, PANalytical B. V.) diagrams with Cu*K*α radiation, the composition of films was analyzed by X-ray photoelectron spectroscopy (XPS, VGMultilab2000X). The typical value of La–Co pair doped Sr-*M* films in our experiments is $La_{0.33}Sr_{0.67}Co_{0.25}Fe_{11.75}O_{19}$. The morphology was evaluated from the atomic force microscope (AFM, Veeco NanoScope MultiMode) images and the statistical data was analyzed by nano measurer 1.2.5 software. The magnetic characteristics were measured using a physical property measurement system (PPMS, Quantum Design).

3. Results and discussion

Fig. 1(a) depicts XRD patterns of the Sr-*M* films with different thickness from 85 nm to 400 nm. Those patterns reveal that all the films are the diffraction lines corresponding to magnetoplumbite crystal structure and no other peaks are detected, which clearly demonstrates that La–Co has effectively entered the $SrFe_{12}O_{19}$ lattice structure [3]. Compared to the pattern of conventional $SrFe_{12}O_{19}$ powders (JCPDS file 79-1411), XRD patterns of these 110 nm and the thinner films show a weak peak and nearly isotropic structure, while the patterns of thickness between 150 nm and 300 nm exhibits a strong crystallo-graphic prefered texture as it almost exclusively displays strong (0 0 *l*) reflections, with $l=6$, 8, 14 of the magnetoplumbite phase. As shown in Fig. 1(b), the rocking curve of (0 0 8) plane for Sr-*M* films with 300 nm exhibits 5.7° of $\Delta\theta_{50,}$ the Gauss fitting of which is also projected. These clearly demonstrate fine texture with most grains oriented with their *c*-axis perpendicular to the film surface. However, as the thickness increases to 400 nm, the intensity of the peaks of (0 0 6) (0 0 8) (0 0 14) with (0 0 *l*) orientation decreases significantly and other randomly oriented peaks of (1 0 7) (1 1 0) (2 0 5) also appear. It is well known that Sr-*M* films have the hexagonal close-packed (*hcp*) stacking structure and the surface energy of (0 0 1) planes is lower than that of any other crystal planes, which is beneficial to form the (0 0 1) preferred orientation [9,10]. Moreover, the residual stress at the thin films exhibits compressive stress and gradually decreases with the increasing of thickness [11], which is favorable to enhance the (0 0 1) texture shown as Fig. 1(a). However, as the thickness is more than 300 nm, (0 0 1) texture of the Sr-*M* films weakens due to the more random nucleation than the perpendicular nucleation [12].

Fig. 2 demonstrates hysteresis loops of Sr-*M* films with $t=85$ nm (a), 110 nm (b), 150 nm (c), 200 nm (d), 300 nm (e), 400 nm (f). The filled and open circles show the hysteresis loops in the perpendicular and in-plane directions to the film, respectively. When the thickness is less than 110 nm, the hysteresis loops of out of plane and in-plane overlap, and the field (*H*) dependent magnetization presents soft ferromagnetic character due to the incomplete development of the (0 0 1) preferred orientation and the insufficient crystallization during the film growth [13]. However, as the thickness is 150 nm, the hysteresis loops of out of plane and in-plane start to deviate, and show significant differences in Fig. 2 (c). At 150–300 nm, the presence of perpendicular magnetic anisotropy is confirmed as the fact that coercivity and remanence squareness ratio (*S*) of Sr-*M* films in perpendicular direction are much higher than those in in-plane, which originates from a preferred (0 0 1) texture as shown in Fig. 1. At 400 nm, the loops display a narrow shape width and show a weakened perpendicular anisotropy.

The relationships between magnetic properties and thickness are detailed in Figs. 3 and 4. As shown in Fig. 3, the change of M_s as a function of strontium hexaferrite film thickness is monotonously increased and *S* is improved sharply at first and then shows a little fluctuation near 0.8. However, the change of coercivity is complex, although the in-plane coercivity ($H_{c\parallel}$) is always below 500 Oe as shown in Fig. 4, the coercivity out of plane ($H_{c\perp}$) increases rapidly at first and then decreases gradually between the thickness range of 85–400 nm. Notably, $H_{c\perp}$ reaches a maximum of 2870 Oe at 300 nm, lower than H_c of the bulk materials (5503 Oe) [14]. Here the perpendicular anisotropy fields (H_k) of Sr-*M* films

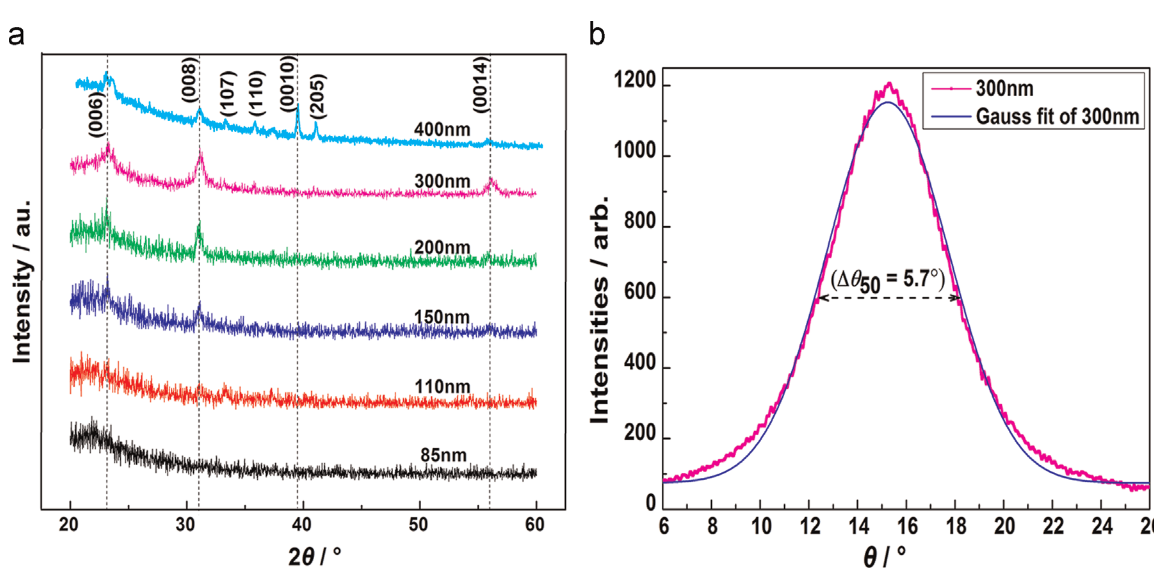

Fig. 1. XRD patterns of Sr-*M* films with different thickness: 85 nm, 110 nm, 150 nm, 200 nm, 300 nm, 400 nm, respectively (a) Rocking curve of (0 0 8) plane for the Sr-*M* films with 300 nm and its Gauss fitting, respectively.

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