Journal of Magnetism and Magnetic Materials 324 (2012) 2894–2898

Contents lists available at SciVerse ScienceDirect

Journal of Magnetism and Magnetic Materials

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

Preparation of strontium hexaferrite film by pulsed laser deposition with *in situ* heating and post annealing

S.M. Masoudpanah [a,*], S.A. Seyyed Ebrahimi [a], C.K. Ong [b]

[a] Center of Excellence for Magnetic Materials, School of Metallurgy and Materials,College of Engineering, University of Tehran, Tehran, Iran
[b] Center for Superconducting and Magnetic Materials, Department of Physics, National University of Singapore, 2 Science Drive 3, Singapore 117542, Singapore

ARTICLE INFO

Article history:
Received 4 February 2012
Received in revised form
7 April 2012
Available online 4 May 2012

Keywords:
Strontium hexaferrite film
Perpendicular magnetic anisotropy
In situ heat treatment
Post annealing heat treatment

ABSTRACT

Strontium hexaferrite ($SrFe_{12}O_{19}$) films have been fabricated by pulsed laser deposition on Si(1 0 0) substrate with Pt(1 1 1) underlayer through *in situ* and post annealing heat treatments. *C*-axis perpendicular oriented $SrFe_{12}O_{19}$ films have been confirmed by X-ray diffraction patterns for both of the *in situ* heated and post annealed films. The cluster-like single domain structures are recognized by magnetic force microscopy. Higher coercivity in perpendicular direction than that for the in-plane direction shows that the films have perpendicular magnetic anisotropy. High perpendicular coercivity, around 3.8 kOe, has been achieved after post annealing at 500 °C. Higher coercivity of the post annealed $SrFe_{12}O_{19}$ films was found to be related to nanosized grain of about 50–80 nm.

1. Introduction

With respect to applications in microwave circuits, strontium and barium hexaferrite films with epitaxial quality are widely studied. It is well known that the *c*-axis perpendicular oriented M-type hexaferrite films exhibit large perpendicular magnetic anisotropy field (Ha) of 17 kOe, thus could keep magnetization perpendicular to the film plane by overcoming demagnetization in plane [1]. M-type hexaferrites have also been identified as one of the candidates for high density magnetic recording media due to their large coercivity and good mechanical and chemical stability [2,3]. For the preparation of $SrFe_{12}O_{19}$ (SrM) thin film as a high density perpendicular magnetic recording medium, much effort has been concentrated on the *c*-axis perpendicular orientation [4] and the decrease of substrate temperature during deposition [5]. The substrate temperature necessary for the crystallization of SrM films is generally in the range of 500–600 °C and it is strongly desired to lower this temperature, because the interdiffusion between magnetic layer and substrate at high temperatures strongly affects the magnetic properties [6].

Some researchers have been studying post annealing heat treatment of SrM films deposited at low temperatures in order to develop a new process which is suitable for practical fabrication of recording media [7]. It is reported that the post annealing of strontium hexaferrite films prepared by sputtering results in much larger coercivity than that of the films obtained by *in situ* heat treatment. However, the post annealed SrM films exhibit a large dispersion on the *c*-axis orientation [7].

It is well known that pulsed laser deposition (PLD) is a powerful method to deposit oxide films. Some works on laser deposition of strontium hexaferrite thin films have been reported. The coercivity of strontium hexaferrite films prepared by laser deposition is much lower than that of conventional sputtering [8]. However, the combination of PLD and different heat treatment processes may be a promising method to fabricate SrM films with controllable magnetic properties.

In this work, we report the microstructure and magnetic properties of strontium hexaferrite films synthesized by laser deposition on commercial Pt(400 nm)/Ti(400 nm)/SiO_2/Si substrate with *in situ* and post annealing heat treatments.

2. Experimental procedure

The $SrFe_{12}O_{19}$ target was prepared by sol–gel technique. The required proportion of iron nitrate, strontium nitrate and citric acid were dissolved in the distilled water at which the molar ratio of citric acid to total metal ions was 1:1 and the Fe/Sr molar ratio was 10 [9]. 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 resulting gel was heated at 200 °C till it ignited in a self-propagated 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 a slow cooling in the furnace. The

* Corresponding author. Tel.: +98 21 61114132, fax: +98 21 8800 6076.
E-mail address: masoodpanah@ut.ac.ir (S.M. Masoudpanah).

0304-8853/$ - see front matter © 2012 Elsevier B.V. All rights reserved.
http://dx.doi.org/10.1016/j.jmmm.2012.04.034

resulting target was homogeneous and single phase with a density greater than 90% with respect to the theoretical one.

Films were deposited by focusing 260 mJ, 23 ns pulses from a KrF excimer laser (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. One group of samples was deposited by *in situ* heating at 450–800 °C and in oxygen pressure of 0.13 mbar during film growth [10]. The as-deposited film was cooled down slowly to room temperature at 1 atm O_2 pressure. Another group of films was prepared by post annealing, that is, films deposited at 450 °C and in oxygen pressure of 0.13 mbar and followed by annealing at 500–800 °C in the same pressure of flowing oxygen. After 30 min annealing, the films were cooled down slowly to room temperature at 1 atm oxygen pressure. The deposition for 30 min produces films with thickness of about 200 nm, according to the cross sectional scanning electron microscope (SEM) micrograph [11].

The crystal structure of the films was checked by X-ray diffraction (XRD) in θ–2θ geometry, using CuKα radiation (Philips PW-1730). The thickness was determined using a SEM (CamScan MV2300). Atomic/magnetic force microscope (AFM/MFM) (Veeco Dimension 3100) was employed to image the topography and magnetic structure of the films. 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 XRD patterns of the SrM films deposited on Si(1 0 0) substrate with Pt(1 1 1) underlayer after post annealing and *in situ* heat treatment. For deposition temperature of 450 °C, the deposited film appeared to be amorphous. Whereas, the as-grown film at 450 °C shows no crystallinity in XRD; after post annealing for 30 min (Fig. 1b), the film shows significant reflections of the SrM (0 0 1) planes in the XRD pattern, which indicates that its *c*-axis is perpendicular to the film plane. Presumably, there are small *c*-axis oriented "nuclei" in the as-deposited film at 450 °C, which grow during the subsequent heat treatment. The XRD patterns of the SrM films with *in situ* heating also show only (0 0 1) reflections at all deposition temperatures (Fig. 1c). The lattice mismatch between SrM (0 0 1) and Pt (1 1 1) is about 6.2%, while that for the SrM (0 0 1) and Si (0 0 1) is ~9% [12]. According to Frank and Van der Merwe's theory [13], if the value of the mismatch parameters is larger than ~9%, hetero-epitaxy cannot be formed between the two layers. Therefore, the *c*-axis growth of SrM is due to the small mismatch between *c*-axis oriented SrM film and Pt (1 1 1) underlayer. Dispersion of the *c*-axis orientation of the films can be determined by the full width at half-maximum ($\Delta\theta_{50}$) of rocking curves (ω-scan) taken on the (0 0 8) diffraction peak. The rocking curves for the SrM film grown with *in situ* and post annealing at 700 °C are shown in Fig. 2. The film prepared with *in situ* heating exhibits smaller $\Delta\theta_{50}$ due to the higher mobility of the adatoms for arranging in proper crystal structure during deposition. Furthermore, as we shall see, post annealed films are more granular with smaller grains; hence a larger $\Delta\theta_{50}$ is not surprising.

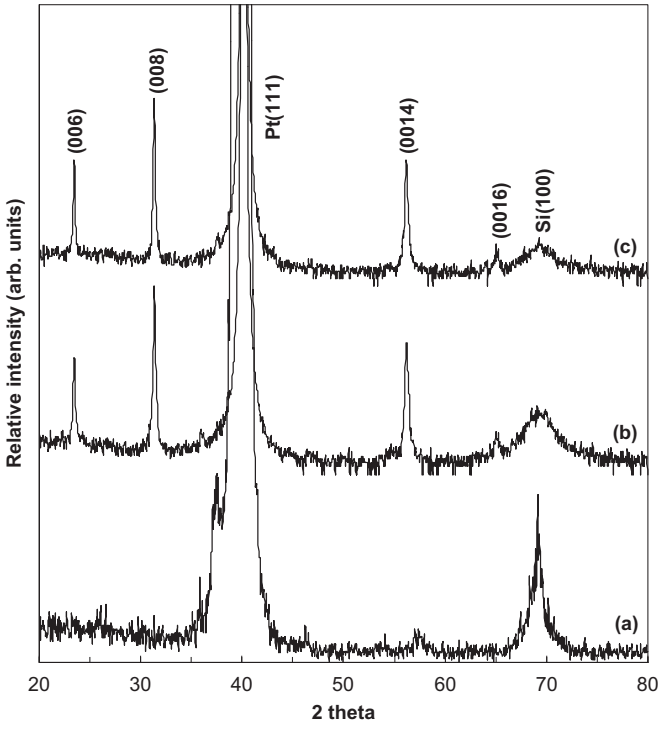

Fig. 1. XRD patterns of $SrFe_{12}O_{19}$ films deposited on Si(1 0 0) substrate with Pt(1 1 1) underlayer, (a) at 450 °C, (b) after post annealing at 700 °C, and (c) with *in situ* heating at 700 °C.

Atomic/magnetic force microscopy was used to evaluate the grain size, surface morphology and magnetic structure. AFM morphologies of the SrM films with *in situ* and post annealing are shown in Fig. 3. It is very obvious that the films with *in situ* heating have hexagonal shape grains. This indicates that the films are grown with *c*-axis normal to the film plane, which is consistent with XRD results. The films have also an average grain size in the range of 100–250 nm with rough surface of the root mean squared roughness (RMS) value of 10–20 nm. In contrast, the post annealed SrM films have smaller grain size of 50–80 nm along with smooth surface with the RMS value of 5–10 nm, which is a very promising parameter for high density magnetic recording media. The possible reason for larger grain size with *in situ* heat treatment is higher mobility of active atoms on heated substrate during film growth [14].

Fig. 4 shows AFM and MFM images of the SrM film with *in situ* heat treatment and after post annealing at 700 °C. The observed bright and dark contrast images are representative of a domain

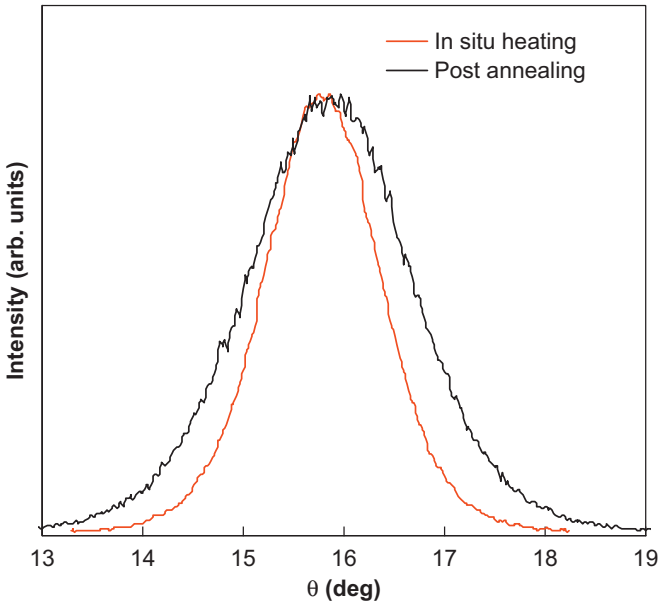

Fig. 2. Rocking curves of (0 0 8) plane for the SrM film prepared with *in situ* and post annealing at 700 °C.