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Analysis and contribution of stress anisotropy in epitaxial hard ferrite thin films

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Abstract

The stress anisotropy in epitaxial hard ferrites thin films ($BaFe_{12}O_{19}$, $CoFe_2O_4$) has been investigated using two methods. (a) The thickness dependence of torque curves and magnetic hysteresis loops. (b) The comparison between magnetic and magneto-optic Kerr hysteresis loops. Both analyses confirm the domination of stress in $CoFe_2O_4$ whereas in $BaFe_{12}O_{19}$ films the stress is too weak to compete with magnetocrystalline anisotropy.

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Keywords: Thin films; Magnetic anisotropy; Torque curve; Kerr effect; Hystersis loop

1. Introduction

Hard ferrites such as hexagonal ($BaFe_{12}O_{19}$) and cubic ($CoFe_2O_4$) continue to be of a significant interest for fundamental studies as well as for applied research [1–3]. Their large magnetocrystalline anisotropy and high chemical stability suggest that these materials are potential candidates for magnetic and magneto-optic recording media [4,5]. Highly oriented films are demanded to achieve a large coercivity and high remanence, which constitute the basic requirements of such applications. Despite the crucial role of the substrate in the orientation of the film texture, additional effects such as stress could prevail and influence magnetic properties of the film. The importance of stress anisotropy is related to the material magnetostriction as well as to the lattice state in the film, which is generally film-thickness dependent. A strong stress is expected at the film–substrate interface whereas the top of the film is supposed to be more relaxed. Since $BaFe_{12}O_{19}$ and $CoFe_2O_4$ exhibit large magneto-optic effects we propose to study the importance of stress anisotropy in both materials epitaxially grown by probing their surface magnetic properties with polar Kerr effect. On the other hand torque and VSM measurements are more volume or bulk related and are performed to establish the thickness dependence of magnetic anisotropy in such films.

2. Structural analyses and stress anisotropy probe

$BaFe_{12}O_{19}$ and $CoFe_2O_4$ films have been grown by pulsed laser deposition (PLD) on (0 0 1) Al_2O_3 and (1 0 0) MgO, respectively. The deposition has been

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performed at various substrate temperatures in the presence of a controlled oxygen pressure. Films with different thicknesses (50–300 nm) have been prepared by changing the number of laser shots. More details about the growth conditions are reported in Ref. [6].

The film topography is shown by the AFM images of Fig. 1. In contrast to a single-crystal structure achieved in $CoFe_2O_4$ film (Fig. 1(a)), $BaFe_{12}O_{19}$ consists of a granular structure with a smooth surface (Fig. 1(b)). The grains are circular, close to each other and exhibit a uniform size distribution (70 nm). The transition from single to polycrystalline structure illustrated in Fig. 1 could be related to the large difference in the film–substrate lattice mismatch estimated at 0.48 and 7% for $CoFe_2O_4/MgO$ and $BaFe_{12}O_{19}/Al_2O_3$, respectively. XRD measurements ($\theta/2\theta$ scan) have been performed on both ferrite films and are presented in Fig. 2. Both spectra reveal single-phase films with crystallographic orientations parallel to (1 0 0) and (0 0 1) textures for $CoFe_2O_4$ and $BaFe_{12}O_{19}$ films, respectively. However, the very narrow rocking curves (0.15° and 0.5° as FWHM for $CoFe_2O_4$ and $BaFe_{12}O_{19}$, respectively) illustrates a highly oriented structure in our films. The epitaxial growth promotes our films to be perfect samples for stress analyses.

A simple way to probe the stress in epitaxial thin films consists of investigating the thickness dependence of their magnetic properties. A good illustration of such effect is presented in Fig. 3, which shows the thickness dependence of torque curves of $CoFe_2O_4$ and $BaFe_{12}O_{19}$ films. These measurements have been performed at constant rotating field (1350 kA/m). At 0 and 90° the field lies along the normal and parallel directions to the film plane, respectively. The (a) and (b) graphs of Fig. 3 are the torque curves of 60 nm and 180 nm thick $CoFe_2O_4$ films, respectively. In contrast to the cubic anisotropy of the bulk the 180° periodicity in Fig. 3(a) confirms an oriented uniaxial anisotropy in thinner film. Moreover, the large rotational hysteresis appearing around 90° reveals a perpendicular anisotropy in the film with anisotropy field (H_k) much larger than the measurement field (1350 kA/m). It is important to notice the existence of a small periodic kink at 0 and 180° in Fig. 3(a). Upon increasing the film thickness drastic changes affect the torque curve and can be listed from Fig. 3(b) as follows. (a) The rotational hysteresis of the perpendicular anisotropy is significantly reduced. (b) The kink observed in thinner film is considerably

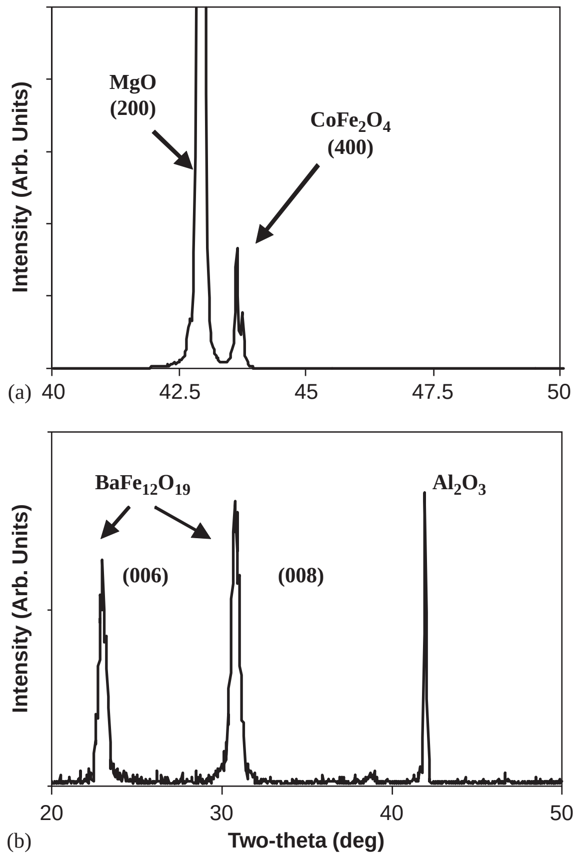

Fig. 2. $\theta/2\theta$ scan of (a) $CoFe_2O_4$ and (b) $BaFe_{12}O_{19}$ films.

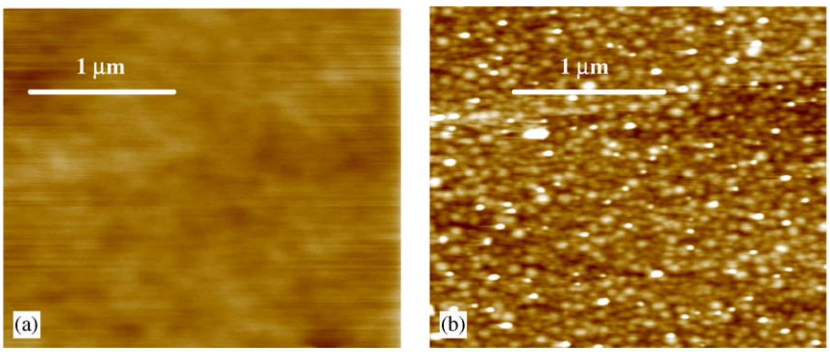

Fig. 1. (3 μm × 3 μm) AFM images of (a) $CoFe_2O_4$ and (b) $BaFe_{12}O_{19}$ thin films epitaxially grown on (1 0 0) MgO and (0 0 1) Al_2O_3, respectively.

enhanced and can be identified as an additional in-plane uniaxial anisotropy in the film. The torque curves of $CoFe_2O_4$ reveal the existence of two uniaxial anisotropies (in-plane and perpendicular) competing between each other. At low thickness the perpendicular anisotropy prevails whereas at large thickness the difference between both components (in-plane and out-of-plane) is considerably reduced. Since our $CoFe_2O_4$ films exhibit a single-crystal structure the unique way to explain such behavior is to consider the stress and magnetocrystalline

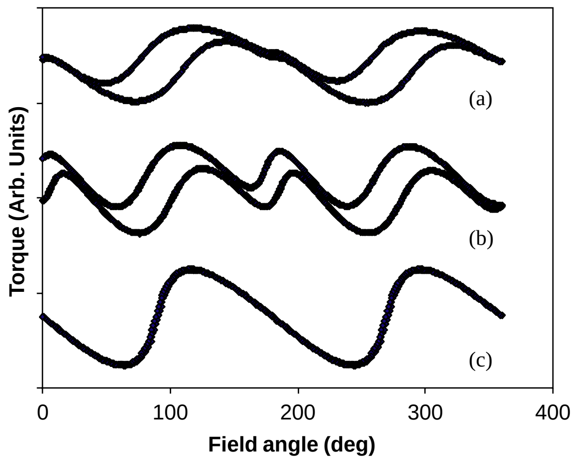

Fig. 3. Torque curves of (a) thin (60 nm), (b) thick (180 nm) $CoFe_2O_4$ films and (c) 200 nm thick $BaFe_{12}O_{19}$.

as sources of anisotropy. Although a small misfit (0.48%) exists between the MgO and $CoFe_2O_4$ lattices a strong stress anisotropy could be induced especially at low film thickness due to the large magnetostriction of cobalt ferrite [7]. Since $CoFe_2O_4$ exhibits a negative magnetostriction the tensile stress detected by XRD in our films [8,9] induces a large perpendicular anisotropy ($5 \times 10^6\,\mathrm{erg/cm^3}$) at low thickness. The magnitude and orientation of stress anisotropy estimated from XRD show a good agreement with the torque measurement. At low thickness the perpendicular stress anisotropy dominates the in-plane magnetocrystalline component as illustrated by Fig. 3(a). Upon increasing the film thickness the stress is progressively released due to the lattice relaxation, [9] which allows the in-plane magnetocrystalline anisotropy to become more competitive (Fig. 3(b)). On the other hand the torque curve of $BaFe_{12}O_{19}$ (Fig. 3(c)) reveals the existence of magnetic anisotropy with the following characteristics. (a) The anisotropy is uniaxial and oriented perpendicular to the film plane. (b) The very small rotational hysteresis localized at 90° indicates that H_k in such films is close to the measurement field (1350 kA/m). (c) The torque curve behavior is constant regardless the film thickness. The latter result suggests that stress is not active in the control of the magnetic anisotropy of $BaFe_{12}O_{19}$ films despite their large lattice mismatch (7%) to Al_2O_3. The only prevailing form of anisotropy in such structure is magnetocrystalline.

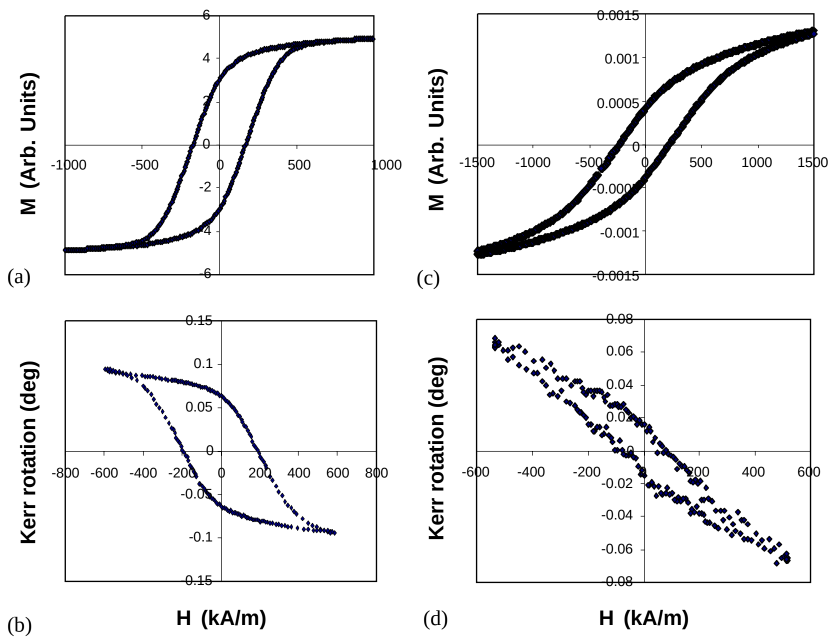

Fig. 4. (a) Magnetic and (b) Kerr loops of $BaFe_{12}O_{19}$ film (200 nm thick). (c) Magnetic and (d) Kerr loops of $CoFe_2O_4$ film (180 nm thick). Note the difference in the respective scales of the loops.

An alternative technique to investigate the stress consists of a direct comparison between the surface and volume properties of the film. As magneto-optic effects are large in both ferrites films their surface magnetism can be probed using Kerr effect. On the other hand, the volume magnetic properties can be accessible from VSM hysteresis loops. Both wavelength spectra and hysteresis loop measurements have been carried out with polar Kerr effect. The $BaFe_{12}O_{19}$ Kerr spectra reveals the existence of strong peaks in the UV wavelength whereas the maximum Kerr rotation in $CoFe_2O_4$ occurs around 600 nm. The (a) and (b) graphs of Fig. 4 are the loops of $BaFe_{12}O_{19}$ film (200 nm thick) measured by VSM and Kerr at 390 nm wavelength, respectively. Except the inversion of magneto-optic loop (Fig. 4(b)) due to the negative Kerr rotation both curves (a) and (b) exhibit the same behavior including the loop shape, the coercivity (200 kA/m) and the remanence (0.7). Since Kerr is a surface effect and mainly probes the magnetization of the film surface (10–20 nm) it is easier to confirm the similarity between the volume and surface properties in $BaFe_{12}O_{19}$ regardless the film thickness. Such result illustrates the weakness of stress in such films and supports the analyses reported before on the thickness dependence of magnetic anisotropy. The (c) and (d) graphs of Fig. 4 are the magnetic and magneto-optic loops of 180 nm thick $CoFe_2O_4$ film. The coercivity and hysteresis of Kerr loop (100 kA/m) (Fig. 4(d)) are considerably reduced in comparison to those of magnetic loop (250 kA/m) (Fig. 4(c)). Such result can be explained by the weakness of the perpendicular stress anisotropy at the top of the film due to the lattice relaxation. The difference between magnetic and Kerr loops is very significant for both minor as well as major loops. It is clear that the properties of the surface are completely different from those of the volume despite the single-crystal structure of $CoFe_2O_4$ films. Such result seems to comfort the investigation reported before. The difference in behavior of stress anisotropy in $BaFe_{12}O_{19}$ and $CoFe_2O_4$ is certainly related to the large difference in magnetostriction of both materials as well as to the film microstructure.

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