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The effects of ZnO underlayer on microstructural and magnetic properties of $BaFe_{12}O_{19}$ thin films

A Lisfi[1] **and J C Lodder**

Information Storage Technology Group, MESA+ Research Institute, University of Twente, PO Box 217, 7500 AE Enschede, The Netherlands

E-mail: a.lisfi@el.utwente.nl

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Abstract

Barium ferrite films have been grown by pulsed laser deposition on oxidized Si (100) with and without ZnO buffer layers. The films grown directly on SiO_2/Si exhibit a granular structure with two easy axes (in-plane and perpendicular) and strong magnetic coupling. It was found that a ZnO underlayer promotes the orientation of the *c*-axis perpendicular to the film plane. Additionally, good separation between the grains and a low exchange coupling can be achieved in $BaFe_{12}O_{19}$/ZnO/SiO_2/Si films. Grain isolation and magnetic de-coupling are believed to be caused by the interdiffusion between the film and the ZnO buffer layer, which gives birth to a new non-magnetic phase ($ZnFe_2O_4$).

During recent years, magnetic recording has demonstrated an enormous progress, due to the large increase in areal density, which presently exceeds more than 10 Gb in^{-2}. The reasons for such a performance are multiple, but it is mainly due to the improvement and the introduction of new magnetic heads (MR, GMR). Additionally, new magnetic media, which can satisfy the requirements for high recording density, have become available. However, media noise and thermal stability constitute the major obstacles for increasing recording density. Because of the high sensitivity of the new heads, media noise becomes a limiting factor in reaching high performance. The origin of media noise is correlated to the magnetic coupling between grains in the media. To overcome such problems, much effort has been made to produce new alloy media with magnetic grains isolated from each other by other elements, such as Ta or Cr. Moreover, magnetic materials with high anisotropy become more attractive for magnetic recording, because magnetic stability can be achieved in very small grains.

Barium ferrite ($BaFe_{12}O_{19}$) is a magnetic oxide with high chemical stability and good mechanical hardness. Moreover, this material exhibits a hexagonal structure with a large magnetocrystalline anisotropy, parallel to the *c*-axis. Such properties make $BaFe_{12}O_{19}$ material very suitable for longitudinal as well as perpendicular recording [1–3]. Because of its hexagonal structure, sapphire is an appropriate substrate for achieving a good orientation

[1] Author to whom correspondence should be addressed.

of the anisotropy as well as a high squareness in $BaFe_{12}O_{19}$ film [4–7]. Unfortunately, the high cost of sapphire hinders its application in industry. ZnO could be an alternative, because of its hexagonal structure, which can fit with that of barium ferrite. Moreover, ZnO can grow epitaxially on SiO_2/Si [8] as well as on quartz [9] with the *c*-axis perpendicular to the film plane and can be used as a buffer layer to grow $BaFe_{12}O_{19}$.

In this paper we report on the influence of a ZnO underlayer on microstructural and magnetic properties of $BaFe_{12}O_{19}$ films. ZnO buffer layers with 250 nm thickness were grown on oxidized silicon substrates with rf planar magnetron sputtering. More details about their preparation can be found in [8]. X-ray diffraction (XRD) analysis (θ–2θ scan) revealed that the ZnO films exhibits a texture parallel to (001), which indicates that the *c*-axis is perpendicular to the film plane. However, from rocking curve measurements, the dispersion in the *c*-axis was estimated to be 4°. The $BaFe_{12}O_{19}$ films were grown by pulsed laser deposition by using a KrF excimer laser (248 nm wavelength) and a vacuum chamber [10]. In order to study the effects of the ZnO underlayer, we deposited $BaFe_{12}O_{19}$ on oxidized Si (100) with and without ZnO underlayers. The growth conditions were 1.5 J cm^{-2} and 5 Hz as energy density and laser repetition rate, respectively. During deposition, the substrate was heated to 770 °C and the O_2 pressure was kept constant at 100 mTorr. By fixing the distance between the target and substrate at 45 mm, the deposition rate was estimated to be 10 nm min^{-1}.

In figures 1(a) and 1(b), two SEM images are presented and show the microstructure of the two different $BaFe_{12}O_{19}$ films grown on SiO_2/Si without and with a ZnO buffer layer, respectively. Both films are 300 nm thick and were grown under the same conditions. The topography of the film deposited directly on SiO_2/Si (figure 1(a)) consists of circular as well as elongated grains, very close to each other. In a recent work [11], we showed that the microstructure of barium ferrite grown on SiO_2/Si depends strongly on the film thickness. At low thickness, less than 50 nm, the topography of $BaFe_{12}O_{19}/SiO_2$/Si film exhibits circular grains and looks close to that of films grown epitaxially on sapphire [10]. By increasing the film thickness, the proportion of elongated grains increases and induces a drastic change in the magnetic properties. We established that circular grains exhibit a perpendicular easy axis of magnetization, whereas the anisotropy of elongated grains is confined in the film plane. The strong exchange coupling between both kinds of grains (circular and elongated) induces a kink in the torque curve of the film, which deviates from that typically known for a system with uniaxial anisotropy [11]. The morphology of $BaFe_{12}O_{19}$ film, grown on a ZnO underlayer (figure 1(b)), shows a granular structure with mostly a circular shape. The grain size distribution is narrow with a mean diameter of 60 nm. Moreover, in contrast to figure 1(a), the grains in figure 1(b) are well separated from each other, which indicates a low magnetic intergranular exchange coupling in this film.

Figures 2(a) and 2(b) show XRD spectra (θ–2θ scan) of $BaFe_{12}O_{19}$ films deposited without and with a ZnO underlayer, respectively. The film grown directly on SiO_2/Si (figure 2(a)) exhibits a single $BaFe_{12}O_{19}$ phase with different crystallographic orientations such as (107), $(2\,0\,\underline{11})$ and $(2\,\underline{11}\,\underline{13})$. The orientation of the *c*-axis in each texture can be estimated and corresponds, for example, to 61° and 16° tilt from the film plane for (107) and $(2\,\underline{11}\,\underline{13})$, respectively. This result clearly shows that grains with nearly in-plane and perpendicular easy axes (*c*-axis) are present in the $BaFe_{12}O_{19}/SiO_2$/Si film. The spectrum of figure 2(b) reveals two important points: (1) $BaFe_{12}O_{19}$ is not the only phase, but coexists with a new one, which is $ZnFe_2O_4$; (2) all the crystallographic orientations of $BaFe_{12}O_{19}$ such as (006) and (008) are parallel to the (001) texture. The latter result confirms that the *c*-axis is fully perpendicular to the film plane. The origin of the $ZnFe_2O_4$ phase can be explained by the high deposition temperature (770 °C), which is able to induce a large interdiffusion between the film and the ZnO underlayer. In contrast to the hexagonal structure of barium ferrite, $ZnFe_2O_4$ is a normal spinel with a cubic structure. Because of the preferential occupation of tetrahedral sites by Zn^{+2}, the 16 Fe^{+3} ions

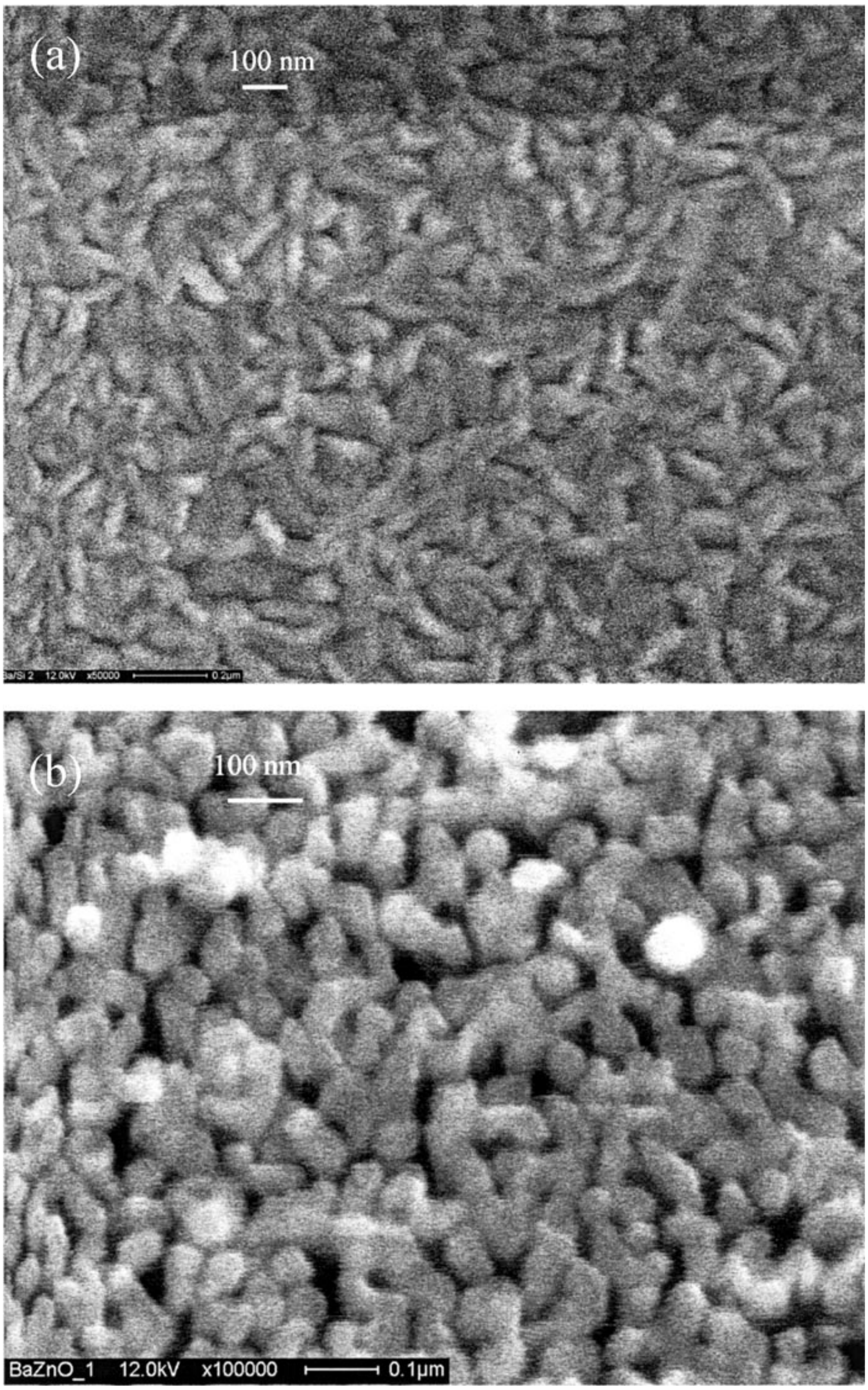
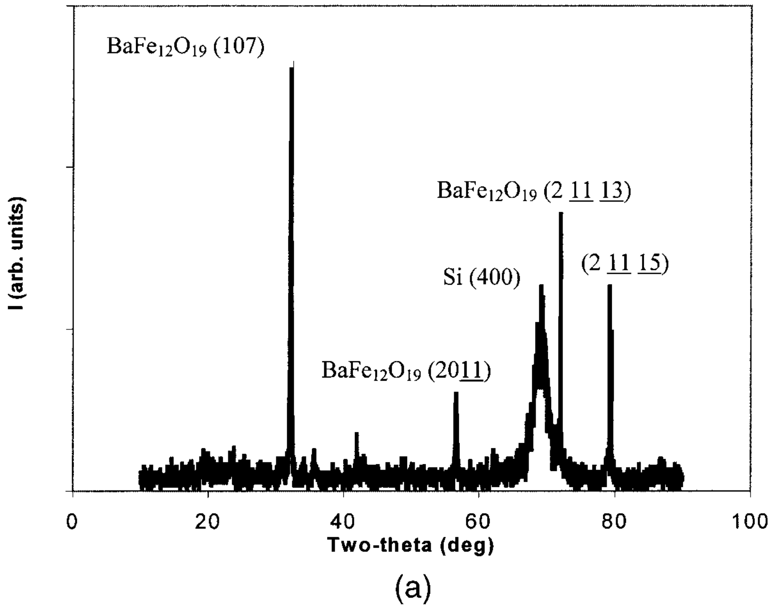


Figure 1. (a) SEM image of $BaFe_{12}O_{19}$ film (300 nm thick) grown on SiO_2/Si. Elongated and circular grains coexist and are very close to each other. (b) SEM picture of $BaFe_{12}O_{19}$ film (300 nm thick) grown on SiO_2/Si with 250 nm ZnO buffer layer. The grains are circular and well separated from each other.



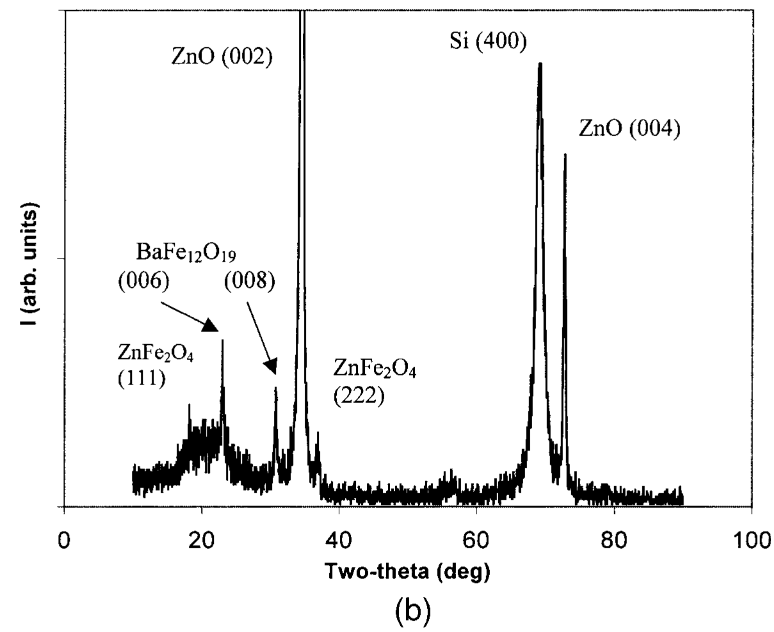


Figure 2. (a) XRD spectra of $BaFe_{12}O_{19}/SiO_2/Si$ film showing a single $BaFe_{12}O_{19}$ phase with different crystallographic orientations. (b) XRD spectra of barium ferrite film grown on oxidized silicon with ZnO buffer layer. The $BaFe_{12}O_{19}$ is highly oriented following the (001) texture and a new phase ($ZnFe_2O_4$) appears.

of the $ZnFe_2O_4$ lattice are distributed in octahedral sites. Moreover, the magnetic moments of Fe^{+3} are randomly oriented and cancel each other. Consequently $ZnFe_2O_4$ is not magnetic.

In figure 3(a), the in-plane and perpendicular magnetization loops of $BaFe_{12}O_{19}$ grown on SiO_2/Si are shown. Large coercivity ($H_c = 380$ kA m^{-1}) as well as high squareness

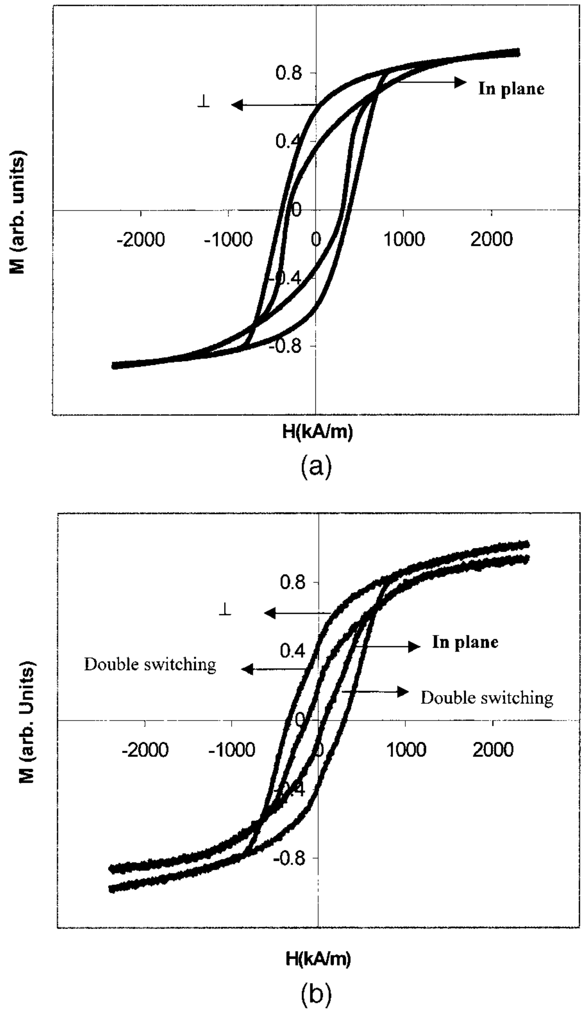


Figure 3. (a) In-plane and perpendicular hysteresis loops of $BaFe_{12}O_{19}$ film deposited directly on oxidized silicon. The anisotropy is perpendicular and strong coupling exists between the grains (smooth loops). (b) In-plane and perpendicular hysteresis loops of film grown on the ZnO underlayer. Double switching characterizes both loops because of the low magnetic coupling.

characterize the perpendicular loop. The large hysteresis in the in-plane magnetization can be related to the textures $(2\,\underline{11}\,\underline{13})$ and $(2\,\underline{11}\,\underline{15})$ with nearly in-plane c-axis. The in-plane loop looks similar to that of a system with a random distribution of easy axes. With a simple comparison between the two loops, we can confirm that the magnetic anisotropy is oriented perpendicular to the film plane. This result can be explained as follows. The film grown on SiO_2/Si consists of grains with in-plane and perpendicular easy axis, but most of the grains exhibit a perpendicular anisotropy. As revealed by XRD (figure 2(a)), the film is polycrystalline and the c-axis of grains has only certain orientations. If we consider a low exchange coupling between grains, multiple switching should be observed in the magnetization loop due to the angular dependence of the switching field. If the magnetization reverses by coherent rotation mode as in the Stoner–Wohlfarth model [12], the angular dependence of the switching field is well known and it is given by

$$H_s(\psi) = H_k/(\sin^{2/3}(\psi) + \cos^{2/3}(\psi))^{3/2}.$$

H_s is the switching field, ψ is the field angle measured from the easy axis and H_k is the anisotropy field of the particle. This is not the case for the $BaFe_{12}O_{19}/SiO_2$/Si film, which exhibits smooth hysteresis loops (figure 3(a)). This confirms the existence of strong exchange coupling between grains, which seems also to be supported by the SEM image of figure 1(a) (grains very close to each other). Figure 3(b) shows both magnetization loops (in-plane and perpendicular) for the $BaFe_{12}O_{19}$ film grown on a ZnO buffer layer. The easy axis is perpendicular to the film plane. However, the hysteresis of the in-plane loop is largely reduced in comparison to that of the film grown without the buffer layer. This can be understood from the XRD measurement of figure 2(b), which shows only textures with a c-axis perpendicular to the film plane. However, the hysteresis in the in-plane loop is too large to be induced only by the dispersion of the c-axis. To explain this effect, it is interesting to refer to the work of Kreisel *et al* [13]. With Raman spectroscopy, Kreisel studied the Raman-active vibrations in $BaFe_{12}O_{19}$ films grown on various substrates such as Al_2O_3 (001), $Gd_3Ga_2O_{12}$ (111) and Si (100). This study suggested the coexistence of well oriented regions and randomly oriented microcrystallites in epitaxial films. Moreover, with XRD, it was not possible to detect any signature of random crystallites. This result may explain the contradiction between the XRD measurement (figure 2(b)) and the large hysteresis in the in-plane loop (figure 3(b)). Another interesting point concerns the existence of double switching in the loops of figure 3(b). This is clear evidence of the existence of two kinds of grains with different orientations of the c-axis and low exchange coupling. The magnetic decoupling can clearly be understood from the SEM image of figure 1(b) and could be related to the $ZnFe_2O_4$ phase, which is not magnetic and may surround the $BaFe_{12}O_{19}$ grains. Additional analysis such as x-ray photon spectroscopy revealed the presence of Zn at the surface of the film, which is a clear confirmation of the existence of $ZnFe_2O_4$ not only at the interface, but also at the top of the film. It is important to point out the low magnetic moment possessed by the $BaFe_{12}O_{19}$ film deposited on ZnO in comparison to that directly grown on SiO_2/Si. As revealed by XRD (figure 2(b)), the existence of an additional phase ($ZnFe_2O_4$), which is not magnetic, can explain the reduction in the magnetization of the film.

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