The power of torque magnetometry: defect induced switching in hexaferrite nanostructures

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Received 4 February 2014, revised 23 June 2014
Accepted for publication 29 July 2014
Published 24 September 2014

Abstract
The presence of magnetic heterogeneities in ferromagnetic systems can reduce their performances in applications such as data storage media and permanent magnets. The detection and the full characterization of these heterogeneities is difficult especially when they are present in a very low concentration within ferromagnetic systems. Here, torque magnetometry is applied to investigate the magnetism of heterogeneities in two-dimensional hexaferrite structures. The study was conducted on a two-dimensional BaFe12O19 structure with decoupled nano-platelets grown on oxidized silicon buffered with ZnO. Measured torque curves reveal anomalous effects manifested by the presence of reversible and hysteretic kinks at large and low magnetic fields respectively. These kinks represent the contribution of magnetic heterogeneities to the global anisotropy of the film in addition to the two-fold symmetry of the major perpendicular anisotropy component. The heterogeneities consist of two types of nano-metric crystallites with the same magnitude of anisotropy as the major magnetic phase, but tilted about −74° and 74° from the normal to the film plane. These results are supported by various types of material characterization (SEM, XPS, XRD, MFM and VSM) and are well reproduced with a simple theoretical model that replicates the symmetry, switching and the easy axes alignment of the heterogeneities magnetization.

Keywords: magnetic anisotropy, torque magnetometry, crystal defects, magnetization switching, hexaferrites, two-dimensional structure

(Some figures may appear in colour only in the online journal)

1. Introduction
Magnetic anisotropy represents one of the most fascinating research topics in magnetism [1–5]. It is generally described as an angular distribution of energy within the magnetic material, which keeps its magnetization confined along a certain preferential direction called the easy axis in the absence of applied magnetic field. Because of its crucial role in many technological applications such as permanent magnets, data storage and magnetic sensing, considerable efforts are invested by the scientific community to investigate its origin as well as its magnitude in advanced magnetic materials. While highly anisotropic structures are required for permanent magnets and data storage applications, non-hysteretic magnetic materials with low anisotropy are suited for sensors application. For example, research is actively engaged in engineering new powerful permanent magnets that could replace the ones currently used in technology, which suffer from the presence of rare earth elements in their chemical
compositions [6, 7]. Metallic alloys such as FeCo and FeNi are among the potential candidates for the next generation of powerful permanent magnets due to their large magnetization, which is suitable for enhancing the energy density product $(BH)_{\text{max}}$ of the magnet. However, their cubic structure can only yield a small magnetic anisotropy, which constitutes a serious obstacle for their performances. Therefore, the big challenge of the ongoing research in this field is to develop innovative ways that can promote the stabilization of the L1$_0$ phase in these metallic alloys, which is predicted to display a huge magnetic anisotropy [8] that can push the energy density product beyond the limit currently achieved in the rare earth elements permanent magnets ($(BH)_{\text{max}} = 50 \text{ MGOe}$). In addition, the huge success enjoyed by the data storage industry over the past years has been driven by the continuous progress achieved in the synthesis of advanced magnetic materials with desirable magnetic properties and microstructure. The recent shift from the longitudinal to the perpendicular magnetic recording [9, 10] has substantially contributed to sustain the upward trend of this technology towards ultra-high recording densities. However, such technology is still facing challenges such as the thermal stability and the noise of the media, which constitute a serious obstacle for ultra-high recording densities. Magnetic coupling between the media grains is believed to be an important source of noise [11, 12], whereas grain size is a limiting factor in the magnetic stability due to the superparamagnetic limit [13, 14]. Magnetically decoupled structures with small grains, narrow switching field distribution and large magnetic anisotropy are regarded as the potential media for ultrahigh-density magnetic recording.

Hexaferrites (BaFe$_{12}$O$_{19}$) are among the potential candidates for perpendicular magnetic recording due to their large uniaxial magnetocrystalline anisotropy that can easily overcome the shape anisotropy of the two-dimensional recording media and lower the superparamagnetic limit. Their high chemical stability [15–17] can prevent any decay of the recorded information. In this article, we report on the magnetic anisotropy of two-dimensional BaFe$_{12}$O$_{19}$ nano-structures grown on an oxidized silicon substrate buffered with ZnO. Torque curves measured at different magnitudes of applied magnetic field reveal an anomalous symmetry in their magnetic structure through the presence of kinks at large field (figure 1(a)). Upon reducing the magnitude of the applied field, these kinks become hysteretic and display rotational hysteresis at specific angular regions of the torque curve. Further reduction of the magnetic field produces a continuous hysteresis that extends to all the angular regions of the torque curve, see figure 1. Based on a careful investigation of the microstructure, crystallographic texture, and the magnetic domains of the films, we were able to interpret the anomalous behaviors of the torque curves using a simple model, which also yields the symmetry of the magnetic anisotropy. The same model provides an explanation for the double switching displayed by both hysteresis loops (in-plane and out-of-plane) of the hexaferrite films.

Figure 1. Out-of-plane torque curves measured at three different magnitudes of applied magnetic field. In the (a), (b) and (c) graphs, the applied field is $1300 \text{ kA m}^{-1}$, $800 \text{ kA m}^{-1}$ and $500 \text{ kA m}^{-1}$ respectively. At 0° and 90° the field direction is perpendicular and parallel to the film plane respectively. Note the presence of reversible kinks (K1, K2, K3 and K4) at large field ($1300 \text{ kA m}^{-1}$) beside the major two-fold anisotropy of the film. These kinks become hysteretic (H1, H2, H3 and H4) upon reducing the applied field to $800 \text{ kA m}^{-1}$.

2. Fabrication method and growth conditions of the films

The sample preparation for this study has been done in two steps. First, sputtering was used to grow the buffer layer (ZnO) on thermally oxidized silicon substrate with two inches diameter and 3 μm thickness of SiO$_2$. In the second step of the synthesis, the two-inch ZnO prepared sample was cut into small squares (1 cm × 1 cm), which were used as a substrate to deposit the hexaferrite (BaFe$_{12}$O$_{19}$) film by pulsed laser deposition (PLD). Sputtering was the preferred technique for the first step of the growth since it offers a good thickness uniformity to the sputtered layer over a large area, which is difficult to achieve with PLD. The choice of oxidized silicon and ZnO to grow BaFe$_{12}$O$_{19}$ films is motivated by the following. (1) Oxidized silicon is inexpensive and ZnO can grow with well oriented texture on top of it (c-axis along the normal to the film plane). (2) ZnO exhibits a hexagonal structure that can closely match that of barium hexaferrite and promote the orientation of its c-axis, which is the preferential direction of the magnetization. Therefore, the combination of SiO$_2$/Si and ZnO is expected to produce hexaferrite films with oriented perpendicular magnetic anisotropy [18]. The preparation conditions of the ZnO layer are similar to those reported in [19]. More specifically, the ZnO seed layer was 250 nm thick and was deposited on SiO$_2$/Si by using radio frequency planar magnetron sputtering. However, the BaFe$_{12}$O$_{19}$ films have been grown by PLD under controlled oxygen pressure (100 mTorr). During the laser deposition, the substrate was heated to 770°C and the laser energy density was maintained at 1.5 J cm$^{-2}$. With 10 Hz as repetition rate of the laser shot, 30 min was the amount of time necessary to grow a 200 nm
3. Experimental results

3.1. Symmetry and alignment of magnetic anisotropy, rotational hysteresis, and magnetization curves

Torque magnetometry is the most powerful method for investigating magnetic anisotropy in ferromagnetic systems since it allows the determination of all its characteristics described as follows. (1) The symmetry of the magnetic anisotropy can be directly established from the periodicity of the torque curve. (2) The anisotropy constants (first and second order) can be easily determined from the torque measurement by using Fourier analyses or the Myajima method [20], whereas the anisotropy field can be precisely measured from the field dependence of the rotational hysteresis. (3) Torque measurement facilitates the investigation of competing anisotropies [4], and can well illustrate spin reorientation phenomena when it takes place in magnetic systems. (4) Torque magnetometry can sense magnetic instabilities and predict the mechanism of magnetization reversal [21, 22]. For all these reasons, the present work relies heavily on this technique to thoroughly investigate the magnetic anisotropy of the two-dimensional Ba-ferrite nano-structures, while other structural and magnetic analyses are used to support and corroborate the anisotropy findings. Figure 1 displays typical torque curves of the synthesized hexaferite films measured at three different magnitudes of the applied magnetic field (for (a), (b) and (c) the fields are 1300 kA m\(^{-1}\), 800 kA m\(^{-1}\) and 500 kA m\(^{-1}\) respectively). In such measurement, the amplitude of the applied magnetic field is kept constant, whereas its direction is changing from 0° to 360° (clockwise) then back from 360° to 0° (counterclockwise) with a step of 5°. At 0° and 90° the field direction lies along the normal and parallel directions to the film plane respectively. At large field (1300 kA m\(^{-1}\)), the behavior of the torque curve in figure 1(a) reveals many important aspects of the magnetic anisotropy: (1) The two-fold symmetry of the torque curve (periodicity of 180°) is a clear confirmation of the uniaxial character of the magnetic anisotropy of the films. (2) The easy and hard axes of the magnetization are oriented perpendicular and parallel to the film plane respectively. Since the torque is equal to zero at both axes of the magnetization (easy and hard), the distinction between them is based on the sign of the torque slope, which is negative for the easy axis (0°) and positive for the hard axis (90°). (3) 1300 kA m\(^{-1}\) is the lowest field that produces a reversible torque without hysteresis. This gives an estimation of the anisotropy field \(H_k\) of the hexaferite films to be around 1300 kA m\(^{-1}\). It is important to point out that beside these characteristics of the magnetic anisotropy, figure 1(a) reveals an anomalous behavior in the magnetic torque illustrated by the presence of kinks at different angular positions of the applied field. These kinks denoted by K1, K2, K3 and K4 in the torque graph of figure 1(a) appear at field angles of 16°, 164°, 196° and 344° respectively. Since torque only senses the angular variation of magnetic anisotropy, it is likely that these kinks are related to the contribution of an additional magnetic anisotropy beside the major perpendicular component. With this assumption the following questions arise and need to be answered. (1) Are the four kinks in the torque curve generated by one source of anisotropy or more? (2) What is the symmetry of the magnetic anisotropy related to these kinks and what is its alignment in regard to the film plane? What is the magnitude of the kinks’ anisotropy in comparison to that of the major perpendicular component? These questions will be further discussed and answered in this paper based on the support of other analyses. Figure 1(b) represents the torque curve of the BaFe\(_{12}\)O\(_{19}\) film measured at an intermediate field (800 kA m\(^{-1}\)). Although the two-fold symmetry of the major perpendicular anisotropy is preserved, the magnetic torque becomes hysteretic and the rotational hysteresis of the major component of anisotropy appears at field angles of 90° and 270° in the graph of figure 1(b). Rotational hysteresis is an irreversible mechanism related to the switching of the magnetization, which occurs at a field angle close to the hard axis when the magnitude of the applied field is smaller than that of the anisotropy field of the magnetic system. Obviously, 90° as field angle in the graph of figure 1(b) represents the direction of the hard axis of the major component of anisotropy since its rotational hysteresis is localized at this angle. However, the most intriguing result of figure 1(b) is the hysteretic character of the four kinks described before upon reducing the applied magnetic field. The hysteretic kinks denoted by H1, H2, H3 and H4 in the torque graph of figure 1(b) represent a solid confirmation of the existence of an additional anisotropy beside the major two-fold component that it is oriented perpendicular to the film plane. Based on the number of the hysteretic kinks, their angular spacing, and their angular positions in the torque curve, the characteristics of the kinks anisotropy are: (1) Since the angular spacing between the hysteretic kinks does not equal 90°, the kinks anisotropy is not cubic but consists of two uniaxial components. The hysteretic kinks H1 and H3 belong to one uniaxial component, whereas, H2 and H4 represent the other component. (2) From the angular positions of the kinks in the torque curve, the easy axes of the (H1, H3) and (H2, H4) anisotropies are aligned along directions making angles of −74° and 74° with the normal to the film plane respectively. (3) The magnitude of the anisotropy field \(H_k\) of these two tilted anisotropies is equal to that of the major perpendicular anisotropy (\(H_k \sim 1300\) kA m\(^{-1}\)) since the rotational hysteresis of the kinks anisotropy vanishes at 1300 kA m\(^{-1}\). This result suggests that all the three components of anisotropy have the same origin (magnetocrystalline), except their directions are different; the easy axis for the major component is normal to the film plane, whereas it is tilted for the two minor components. In figure 1(c), a further
reduction of the applied magnetic to 500 kA m$^{-1}$ produces a magnetic torque with a large hysteresis that extends to all the angular regions of the measurement. The continuous hysteresis of figure 1(c) doesn’t imply a random anisotropy, rather a large increase in the rotational hysteresis of each component and their overlapping. All these informative results together with additional structural and magnetic analyses will be used to develop a simple model that can predict the behavior of the torque and its symmetry at each applied magnetic field.

Figure 2 displays the hysteresis loops of the hexaferrite film measured with a VSM along two directions parallel and perpendicular to the film plane. The film magnetic moment is measured at different successive fields between $-2380$ kA m$^{-1}$ and $+2380$ kA m$^{-1}$ with an increment of 10 kA m$^{-1}$. A careful analysis of these magnetic measurements allowed us to reach the following conclusions. (1) The magnetization curves of figure 2 consist of major hysteresis loops rather than minor loops. This result is expected since the maximum applied field in the hysteresis measurement ($2380$ kA m$^{-1}$) is larger than the anisotropy field of the magnetic film ($H_k \sim 1300$ kA m$^{-1}$) estimated from the previous torque analyses. (2) The out-of-plane magnetization curve exhibits a large hysteresis with a high coercivity ($H_c \sim 300$ kA m$^{-1}$) and a large remnant magnetic moment at zero field compared to the in-plane hysteresis loop. This statement confirms the out-of-plane alignment of the magnetization easy axis, which agrees with the torque measurement result. (3) The significant hysteresis displayed by the in-plane magnetization curve and the presence of a double switching in both hysteresis loops (in-plane and out-of-plane) suggest the existence of more than one orientation of magnetic anisotropy. If the magnetization easy axis was fully confined along the normal direction to the film plane, no double switching would be observed in both loops and the in-plane magnetization curve would be reversible without hysteresis. The last magnetic aspect that has been learned from the VSM measurements is the lower magnetic moment of the BaFe$_{12}$O$_{19}$ film deposited on oxidized silicon buffered with ZnO in comparison to the one of the film directly deposited on SiO$_2$/Si without buffering layer. It was found that buffering with ZnO can reduce the magnetic moment of the hexaferrite film about 30%. This important result will be further discussed and explained based on other complementary analyses.

![Figure 2](image1.png)

Figure 2. In-plane and perpendicular hysteresis loops of the hexaferrite structure measured between $-2380$ kA m$^{-1}$ and $+2380$ kA m$^{-1}$. The perpendicular anisotropy in the film is illustrated by the large hysteresis of the perpendicular loop, which displays a high coercivity (300 kA m$^{-1}$) and a large remanence in comparison to the in-plane loop. Note the existence of a double switching in both loops.

![Figure 3](image2.png)

Figure 3. (a) SEM image showing the microstructure of the film, which consists of nano-platelets well separated from each other. The nano-platelets exhibit a hexagonal shape and their average size is 60 nm. (b) MFM picture imaged in the ac demagnetized state of the film showing the structure of magnetic domains. Dark and clear contrasts with similar hexagonal shape of the nano-platelet dominate the magnetic image. These two contrasts represent the stray field of the magnetically decoupled nano-platelets magnetized up and down due to their strong perpendicular anisotropy.
3.2. Structural analyses and magnetic imaging

Figure 3(a) is a SEM image showing the typical topography of an as-deposited BaFe12O19 film grown on oxidized silicon buffered with ZnO. The morphology of the film consists of a unique microstructure characterized by nano-platelets well separated from each other. The nano-platelets exhibit a hexagonal shape rather than circular, and their average size is estimated to be 60 nm. Such microstructure strongly deviates from that typically known for BaFe12O19 films directly deposited on SiO2/Si [23], which displays highly packed elongated grains with a strong magnetic coupling. The low magnetic coupling between the nano-platelets of the synthesized microstructure is also illustrated by the magnetic image shown in figure 3(b). The magnetic domains structure was imaged in the ac demagnetized state of the hexaferrite sample by using a commercial MFM (Digital Instruments, DI 3100). The magnetic structure observed in figure 3(b) looks very different from the stripe domain structure reported in post-annealed BaFe12O19 film grown on sapphire substrate with a perpendicular magnetic anisotropy [24]. As displayed by figure 3(b), two strong magnetic contrasts with dark and clear colors characterize the magnetic structure. A dark contrast represents an attractive force between the magnetic tip and magnetic domain magnetized vertically down. However, a clear contrast is the result of a repulsive force between the magnetic tip and magnetic domain with a magnetic moment aligned vertically up. These two strong contrasts in the magnetic picture of figure 3(b) are surrounded by a weak contrast and exhibit a shape similar to the one of the nano-platelets revealed by the microstructure. These two strong contrasts represent the magnetic stray field of the single domain nano-platelets since their magnetic moments can only be aligned vertically up or down due to their large perpendicular anisotropy. Moreover, the single domain character of the nano-platelets can be understood from their small sizes (60 nm), which cannot sustain a multi-domain structure. The critical size for the transition from single to multi-domains for BaFe12O19 is estimated to be 1 μm [25]. The origin of the weak magnetic contrast can have two potential sources. (1) The tilted anisotropy previously reported in the torque analyses, which weakly interacts with the magnetic tip during the MFM imaging. (2) The presence of a secondary non-magnetic phase on top of the film, which has been clarified by further XRD and XPS investigations; figure 4(a) shows a typical XRD spectrum (θ/2θ scan) of hexaferrite film grown on oxidized silicon buffered with ZnO in the conditions reported before. The XRD scan reveals many interesting crystallographic aspects of the two-dimensional structure. (1) The ZnO buffer layer exhibits a well-oriented texture illustrated by the (002) and (004) reflections. (2) The presence of (006) and (008) peaks of BaFe12O19 indicate that their c-axis is perpendicular to the film plane. (3) A new phase identified as ZnFe2O4 with (111) texture is observed in the spectra. According to Scherrer equation \( D = \lambda / (2 \Delta \omega \times \cos(\theta)) \), where \( D \) is the average crystal size, \( \lambda \) is the x-ray wavelength, \( \Delta \omega \) is the full width at the half maximum of the diffraction peak and \( \theta \) is half of the peak position in the θ/2θ scan spectra), the domain size for this new phase is estimated to be 31 nm. It is suggested that a significant interdiffusion occurs between the film and the buffer layer leading to the formation of this phase since the growth is performed at high temperature (770 °C). It is important to point out that ZnFe2O4 is commonly known among the ferrites family as a non-magnetic spinel phase. To reinforce our claim about the diffusion of Zn, we performed XPS measurement with binding energy range that extends from 0 to 1400 eV on all the samples of interest. As displayed by figure 5, surface analyzes carried out with XPS reveal the existence of Zn at the top of the film, which is an indication that ZnFe2O4 is not uniquely localized at the interface region between the film and the buffer layer. These analyses suggest that the origin of the magnetic decoupling could be related to the ZnFe2O4 phase, which may surround the BaFe12O19 nano-platelets. However, the non-magnetic character of the new spinel phase (ZnFe2O4) can well explain the large reduction of the magnetic moment of the hexaferrite film grown on the
top of the ZnO buffer layer in comparison to the one of the film directly deposited on oxidized silicon.

So far, the result of the XRD spectra of figure 4(a) has only revealed the major perpendicular component of anisotropy, which is related to the (006) and (008) BaFe₁₂O₁₉ textures. However, the textures of the two minor components of the anisotropy reported by the previous torque measurement do not figure in the XRD spectra of figure 4(a). A potential explanation for the missing texture of the minor components of anisotropy could be its overlapping with the ZnO reflections in the XRD spectra. Therefore, it is important to investigate the XRD spectra of BaFe₁₂O₁₉ directly deposited on oxidized silicon without ZnO buffer layer, which is displayed in figure 4(b). This XRD scan is characterized by the presence of several BaFe₁₂O₁₉ textures such as (107), which displays a tilted c-axis of 61° from the film plane. However, after a careful look at both spectra of figure 4, one may realize that the (2 11 13) BaFe₁₂O₁₉ peak appears almost at the same two-theta angle as the (400) ZnO reflection. It is important to mention that the (2 11 13) BaFe₁₂O₁₉ texture represents crystallites with tilted c-axes of +74° and −74° from the normal to the film plane, which coincide exactly with the two minor components of anisotropy revealed by torque analyses before. It is more likely that the (2 11 13) BaFe₁₂O₁₉ and the (400) ZnO reflections coexist and overlap in the XRD scan of figure 4(a).

Based on the support of all these analyses, the full map of the hexaferrite film texture is now complete; it consists of one major component with a perpendicular c-axis and heterogeneities represented by two minor components with tilted c-axes making angles of −74° and 74° with the normal to the film plane. A fundamental question arises: can the symmetry, the kinks, and the rotational hysteresis of the torque curves reported in figure 1 be simulated and reproduced under the established crystallographic textures? If so, what is the fraction of the crystallites with perpendicular c-axis and that of the heterogeneities with tilted c-axes that can reproduce the behavior of the torque curves at different fields displayed by figure 1? These questions are the subject of a detailed study in the next section, which will provide answers based on a simple model.

4. Simulation and modeling of the magnetic anisotropy and its rotational hysteresis

In order to predict the symmetry of the magnetic anisotropy and the rotational hysteresis of its different components reported before, we performed simulations of the torque curves under different applied fields based on a simple model. In this model we assume that the film consists of three kinds of crystallites with three different c-axis orientations as presented in figure 6. \( \alpha_1 \) represents the magnetic fraction of crystallites with normal c-axis to the film plane, whereas \( \alpha_2 \) and \( \alpha_3 \) are those of the crystallites with tilted c-axis (−74° and 74° from the normal to the film plane normal). Moreover, based on the

Figure 5. XPS spectra showing the presence of Zn at the top of film in addition to the chemical elements of the hexaferrite structure such as Ba, Fe and O. The surface analysis character of XPS is a direct proof of the diffusion of Zn to the top of the film.

Figure 6. Angular orientations of the nano-platelets in regard to the film plane. P1 represents the platelets with normal c-axis to the film plane, which represent the major anisotropy component of the film. P2 and P3 represent the heterogeneities with tilted c-axes (−74° and +74° from the normal to the film plane).
support of the microstructure and the magnetic domains imaged by SEM and MFM in figure 3, we assume that the magnetic interactions between the film crystallites can be neglected. However, under the assumption of a non-interacting system, the net torque of the hexaferrite film will be a linear superposition of the torques produced by the three different oriented c-axes. Before simulating the torque curves of the hexaferrite film, it will be interesting to establish the behavior of the torque curves under different applied fields for uniaxial anisotropy. As illustrated by figure 7, when a magnetic system is under the application of magnetic field at an angle \( \psi \) from its easy axis, there is competition between the uniaxial anisotropy energy \( E_a = K_a \sin^2(\theta) \) \( K_a \) is the anisotropy constant and \( \theta \) is the magnetization angle measured from the easy axis) and the Zeeman energy \( E_z = -HM \cos (\psi - \theta) \) where \( H \) is the magnitude of the applied field, and \( M \) is the system magnetization) to control the alignment of the magnetization direction. For each field angle \( \psi \), the compromise between these two forms of energy \( E_a \) and \( E_z \) allows the determination of the stable state of the magnetization angle \( \theta \), and the torque exerted by the anisotropy on the magnetization is given by \( L(\psi) = -\partial E_a / \partial \theta = -K_a \sin(2\theta(\psi)) \). The shape of the torque curve is defined by the relationship between \( \theta \) and \( \psi \), which is strongly dependent on the reduced field \( h = H / H_k \). \( H_k = 2K_a / M \) is the anisotropy field of the magnetic system. Calculations have been performed to illustrate the torque behavior at different reduced fields. The summary of the results presented in figure 8 reveals that different shapes of torque are possible upon varying the reduced field. The simplest case, which is illustrated by figure 8(a), occurs when the reduced field \( h \) is infinite and the Zeeman energy \( E_z \) completely dominates the anisotropy energy \( E_a \). Here the magnetization will be aligned along the direction of the applied magnetic field. Therefore, \( \theta \) and \( \psi \) will be equal, and the torque curve will be given by \( L(\psi) = -K_a \sin (2\psi) \). The torque is then reversible (no hysteresis) and the angular positions of the easy and hard axes of the magnetization correspond to zero torque with negative and positive torque slopes respectively. It is important to note that the absolute values of the torque slopes at the easy and hard axes are equal for infinite reduced field. Upon decreasing the reduced field \( h \) to 1, a significant change occurs in the torque behavior although its reversible character with two-fold symmetry persists as displayed by figure 8(b). This change can be clearly seen in the torque slopes at both axes (easy and hard) of the magnetization. The torque slope at the hard axis is infinitely large due to the rapid variation of the magnetization direction when the field angle is close to the hard axis. The slow variation of the torque at field angles close to the easy axis direction is an indication of the smooth rotation of the magnetization in this angular region. However, the most drastic change in the torque character occurs when \( 0.5 \leq h < 1 \). In this range of reduced field, the torque displays hysteretic behavior as revealed by the graphs (c) and (d) of figure 8. While the torque is reversible around field angles close to the easy axis (0°), it is characterized by a vertical jump with hysteresis when the field direction is close to the hard axis (90°). The rotational hysteresis observed is characteristic of the jump of the magnetization between its two metastable states. The rotational hysteresis is maximum at \( h = 0.5 \) and exhibits a fast decrease versus \( h \) to vanish at \( h = 1 \). It is
important to note that for $h$ smaller than 0.5, the torque displays a reversible character with one-fold symmetry. Now, since the behavior of the torque for oriented uniaxial anisotropy has been established for different reduced fields, it will be much easier to simulate the torque of the hexaferrite film, which basically consists of three anisotropies with normal and tilted easy axes from the film plane. Moreover, the reduced field is the same for the three film anisotropies since they have the same magnetocrystalline origin ($H_{K}$ and $K_u$ are the same for the three anisotropies). Simulations have been performed for the hexaferrite film torque by superposing the torques of the three anisotropies. In the calculation, the magnetic moment of the crystallites with perpendicular $c$-axis represents 80% of the total magnetic moment of the film ($\alpha_1 = 0.8$), whereas the remaining 20% magnetic moment is equally distributed among the heterogeneities represented by the two kinds of crystallites with tilted $c$-axes ($\alpha_2 = \alpha_3 = 0.1$). Figure 9 displays the torque curves of the hexaferrite film simulated at three different values of the reduced field ($h = 1$, 0.8 and 0.5 for the (a), (b) and (c) graphs respectively). The results of the simulation can be summarized as follow. (1) The symmetry of the magnetic anisotropy and the absence of hysteresis are well reproduced in the simulated torque at high field, which perfectly matches the experimental result reported in figure 1(a). The four kinks are well predicted in the simulated high field torque curve ($h = 1$) and their angular positions perfectly coincide with those observed in the measured torque of figure 1(a). (2) The hysteretic character of the torque at intermediate field is well illustrated in the simulated graph of figure 9(b). The simulation reveals three localized components of rotational hysteresis that are similar to those revealed by the experiment in figure 1(b). Moreover, the hysteresis areas and the angular positions of the three rotational hysteresis components are well predicted in the simulated torque curve of figure 9(b) and show good agreement with the measurement reported in figure 1(b). (3) The large continuous hysteresis of figure 1(c) is well reproduced in the simulated torque of figure 9(c). The simulation at low field ($h = 0.5$) shows overlap of the three components of rotational hysteresis. It is important to note that the simulation shows kinks and rotational hysteresis with a vertical jump, whereas in the experiment the kinks and the rotational hysteresis display a smooth behavior with the field angle. This difference is mainly due to the demagnetizing field induced during the measurement and not being considered in the simulation. Because of the out-of-plane configuration of the measurement, the intrinsic field, which is the sum of the applied and the demagnetizing fields is not constant during the torque measurement. This produces a shearing in the torque, which displays a smooth variation at angular regions around the hard axes of the three anisotropies, which is not the case for the simulation.

The double switching observed in the hysteresis loops of figure 2 can be well understood from the independent switching of the magnetizations of the three types of crystallite in the magnetically decoupled nano-structure of the hexaferrite film. It is well known that the switching field varies with the angle between the magnetization easy axis and the direction of the applied field. When the magnetic hysteresis loop is measured along the normal to the film plane, the two types of crystallites with tilted $c$-axes possess the same switching field, but different from that of crystallites with normal $c$-axis, leading to a double switching. The same effect is produced for the in-plane magnetic measurement.

5. Conclusions

Magnetic heterogeneities have been successfully characterized in nano-structured two-dimensional hexaferrites with decoupled magnetic nano-platelets by using torque magnetometry. It has been established that the anomalous effects reported in the experimental torque curves are the result of the contribution of these heterogeneities to the global anisotropy of the film beside its major perpendicular component. These heterogeneities consist of nano-meter crystallites with tilted $c$-axes ($-74^\circ$ and $74^\circ$) from the normal to the film plane and display the same anisotropy field as the major perpendicular component of anisotropy. Based on the magnetically decoupled structure provided by MFM and other structural analyses carried out with XRD, XPS and SEM, a simple theoretical model has been developed to predict the magnetic anisotropy of the hexaferrite film. This model provides a good estimation of the heterogeneities concentration in the film and perfectly reproduces the symmetry and the magnitude of their anisotropy as well as the switching of their magnetization.
Acknowledgments

The authors gratefully acknowledge the financial support from the National Science Foundation under research grants NSF DMR 1206380, DMR 1206397, and DMR 1337916. The authors are very thankful to Dr Karen Gaskell for her contribution with the XPS measurement.

Appendix A

This section provides the calculation detail for determining the c-axis alignment for the heterogeneities crystallites as well as for the major texture in the hexaferrite films. The calculation method is based on the BaFe12O19 textures revealed by the XRD measurement through $\theta$/2$\theta$ scan spectra. It is established that for two reticular planes indexed as $(h_1, k_1, l_1)$ and $(h_2, k_2, l_2)$ in the same hexagonal crystal with $a$ and $c$ axes, the relative angle $\phi$ between both crystallographic planes is expressed according to the following formula:

$$\cos (\phi) = \left[\frac{(h_1h_2 + k_1k_2)}{a^2} + \frac{(l_1l_2)}{c^2}\right]^{1/2} \times \left[\frac{(h_1^2 + k_1^2)}{a^2} + \frac{l_1^2}{c^2}\right]^{1/2} \times \left[\frac{(h_2^2 + k_2^2)}{a^2} + \frac{l_2^2}{c^2}\right]^{1/2}.$$ 

In the $(\theta$/2$\theta$) scan geometry, the Bragg condition $2d\sin \theta = n\lambda$, where $d$ is the spacing distance between the diffracting planes, $\lambda$ is the x-ray wavelength, and $\theta$ represents half of the angular peak position, which represents the diffraction requirement can only be satisfied for reticular planes parallel to the film surface. Consequently, all XRD reflections in the $(\theta$/2$\theta$) spectra correspond to crystallographic planes parallel to the film plane. If we consider two reflections $(h_1, k_1, l_1)$ and $(h_2, k_2, l_2)$ in the $(\theta$/2$\theta$) spectra coming from two different crystals, both planes are parallel to the $(100)$ plane. Consequently, all XRD reflections in the $(\theta$/2$\theta$) spectra correspond to crystallographic planes parallel to the film plane. If we consider two reflections $(h_1, k_1, l_1)$ and $(h_2, k_2, l_2)$ in the $(\theta$/2$\theta$) spectra coming from two different crystals, both planes are parallel to the $(100)$ plane. The relative angle $\phi$ between the $c$-axes of both crystals can be determined according to the last formula. For the (001) reflection the $c$-axis is perpendicular to the film plane and can be used as a reference angle for the calculation. Since for barium ferrite the lattice parameters are $a=0.58945$ nm and $c=2.32$ nm, the angle between the $c$-axes of both crystals relative to the reflections (2 1 1) and (0 0 1) can be easily estimated by using the last formula:

$$\cos (\phi) = \left[-13/(2.32)^2\right]\left\{\left[1/(2.32)^2\right]^{1/2} \times \left[125/(0.58945)^2 + 169/(2.32)^2\right]^{1/2}\right\} = -0.2833 = \cos (\pm 106\)°).

Because the $c$-axis of (001) is perpendicular to the film plane, those of the (2 1 1) texture which represents the heterogeneities crystallites make angles $\theta_1$ and $\theta_2$ with the normal to the film plane equal to $-106\)° + $180\)° = 74\)° and $106\)° − $180\)° = $-74\)° respectively.

Appendix B

As reported in this paper, the investigated hexaferrite films display hysteresis loops that exhibit a double switching and a reduced squareness in comparison to a fully oriented magnetic system. In this section we use the angular dependence of the coercivity and the remanance in hysteresis loop to explain the involvement of the heterogeneities crystallites in the observed double switching character as well as in the reduction of the remanence of the hexaferrite film hysteresis loop. It is important to point out that the reduction of the perpendicular loop squareness is mainly produced through two different sources. (1) The demagnetizing field, which is generated through the measurement geometry and induces a shearing in the perpendicular loop with a reduced remanence. (2) The nearly in-plane alignment of the $c$-axis of the heterogeneities crystallites. As revealed in this article, the heterogeneities crystallites exhibit tilted $c$-axes about $-74\)° and $+74\)° from the normal to the film plane. It is well known that for magnetization reversal mechanism controlled by coherent rotation mode, the coercivity, the switching field and the remanence are strongly dependent on the applied field angle in regard to the direction of the easy axis. Figure A.1(1)
depicts the angular variation of the coercivity, which shows a maximum value when the applied field is parallel to the easy axis (0°) and vanishes at the hard axis ($H_c = 0$ at 90°) due to the reversible character of the magnetization along this direction. A similar decreasing behavior is predicted in the remanence, which drops from 1 at the easy axis (0°) to zero at the hard axis ($H_c = 0$ at 90°) as shown in figure A.1(2). As illustrated by figure A.1(1), it becomes clear that that the coercivity for the majority of the platelets with normal $c$-axis will be different and much larger than that of the heterogeneities crystallites with tilted $c$-axes when the measurement is performed along the normal direction to the film plane. Consequently, a double switching will be observed in both hysteresis loops (out-of-plane and in-plane) due to the magnetic decoupling in this structure. However, as displayed by figure A.1(2) the reduction in the remanence of the perpendicular loop can be well understood from the very small remanence of the heterogeneities crystallites with nearly in-plane $c$-axes. These angular analyses demonstrate that the heterogeneities crystallites are responsible in one hand for the double switching observed in the major hysteresis loop and in the other hand significantly contribute to the reduction of the remanence due to the tilt of their $c$-axes.

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