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Erase/restorable asymmetric magnetization reversal in polycrystalline ferromagnetic films

S. P. Li, Santosh Kulkarni, and Saibal Roy[a)]
Tyndall National Institute, Lee Maltings, Dyke Parade, Cork, Ireland

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Asymmetric hysteresis loops are generally found in exchange-coupled ferromagnetic/antiferromagnetic layers or composite. Once the film is deposited the magnetization reversal behaviour becomes certain due to the fixed anisotropy of the film. We report an asymmetric magnetization reversal, which is erase/restorable in polycrystalline soft magnetic film. When the film is pre-saturated at a high field in the induced uniaxial easy direction, the asymmetric hysteresis loops with one branch governed by "coherent rotation" and another branch with kink induced by mixed reversal mechanism of "coherent rotation" and "rotation/180°-domain-wall-motion/rotation" are obtained. If the film is presaturated in the induced hard axis, the kink disappears and "normal" hysteresis behaviour is observed instead. Such asymmetric magnetization curve can be restored if the film is pre-saturated in the easy axis again. The observed phenomenon is originated from an embedded second magnetically hard phase which tunes the anisotropy in the film. © *2012 American Institute of Physics*. [http://dx.doi.org/10.1063/1.4765652]

The reversal of magnetization in magnetic films has attracted considerable interest in the past decades. This is mainly due to the potential novel applications in magnetic-storage and sensor technology, but also due to an interest in the complex magnetization reversal process itself. An important magnetization reversal property of magnetic thin films is the unusual reversal caused by complex anisotropy, such as asymmetric magnetization reversal,[1–4] multi-stepped switching,[5–13] etc. The asymmetric hysteresis loops are mainly observed in exchange-biased ferromagnetic-antiferromagnetic (FM/AFM) systems[14–19] and in other exchange system like exchange spring coupled bilayers.[20] The discussion and explanation has been evaded for 40 yr until the exchange bias has gained technological importance, as it pins and thus establishes a reference magnetization direction in spintronic devices.[15,21] It has been observed that the magnetization reversal is different at each branch of a hysteresis loop and the asymmetry depends on the angle between the external field and the exchange bias direction,[4,14] i.e., the difference of the two branches in a hysteresis loop varies strongly with the field directions under which the loop was taken. For instance, it has been reported that while on one side of the loop the reversal takes place by magnetization rotation, on the other side it takes place by domain wall (DW) motion or incoherent rotation, and such phenomenon is often observed in certain field directions.[14,15] The asymmetric hysteresis behaviour has been investigated by using various techniques, such as domain imaging,[1,2] polarized neutron reflectometry,[16,17] magnetoresistance,[18] Kerr magnetometry,[19] etc. Different mechanisms have been proposed including higher order FM anisotropy,[4,17] dispersion of the FM or AFM anisotropy axes,[2,3] or training effect induced irreversibility,[22,23] however, a detailed understanding of this effect is still lacking. Nevertheless, a generally accepted origin that induces such asymmetry is an exchange induced unidirectional anisotropy which breaks symmetry of magnetization reversal. In this paper, we report an asymmetric hysteresis in electroplated polycrystalline ferromagnetic films and the curious switching behaviour, which can be erased and recalled by manipulating the anisotropy of the film through magnetically presaturating the film in different directions.

The magnetic films were prepared by electroplating.[24] In brief, 200 nm–1 μm thick $Ni_{50}Fe_{50}$ films were deposited by DC electrical plating at room temperature with an applied magnetic field (200 Oe) to generate a uniaxial anisotropy (refer it as induced anisotropy). A Pt/Ti grid was used as the anode, while a metal film or silicon wafer with a sputtered metal seed layer, such as Cu/Ti, Permalloy/Ti, was used as the cathode. The surface morphology, compositions, and structures of the plated films were analyzed using scanning electron microscopy (SEM), energy dispersive x-rays (EDX), and transmission electron-microscope (TEM), respectively. The in-plane magnetization reversal was studied using hysteresis loop tracer (SHB instruments Inc, USA) equipped with two sets of orthogonal field application coils. The use of the two sets of coils allows one to investigate both the longitudinal and transverse magnetization curves, as is often done with Kerr magnetometer.[3,5,14]

Longitudinal in-plane hysteresis loop measurement in various applied field directions indicates that the plated film has uniaxial anisotropy. Figures 1(a) and 1(b) display hysteresis loops measured from 1 μm thick film in the induced easy ($\theta = 0°$) and hard ($\theta = 90°$) axes. When the field is applied in directions with angle ranging between $45° < \theta < 90°$ asymmetric hysteresis loops appear (Fig. 1(c)). A kink is observed on one branch of the loop, while another branch is a normal magnetization curve as generally observed. The observed asymmetric loops have different polarities when they are measured with fields applied in specular directions around the hard axis (Fig. 1(e)), i.e., if the kink occurs on the

[a)]E-mail: saibal.roy@Tyndall.ie.

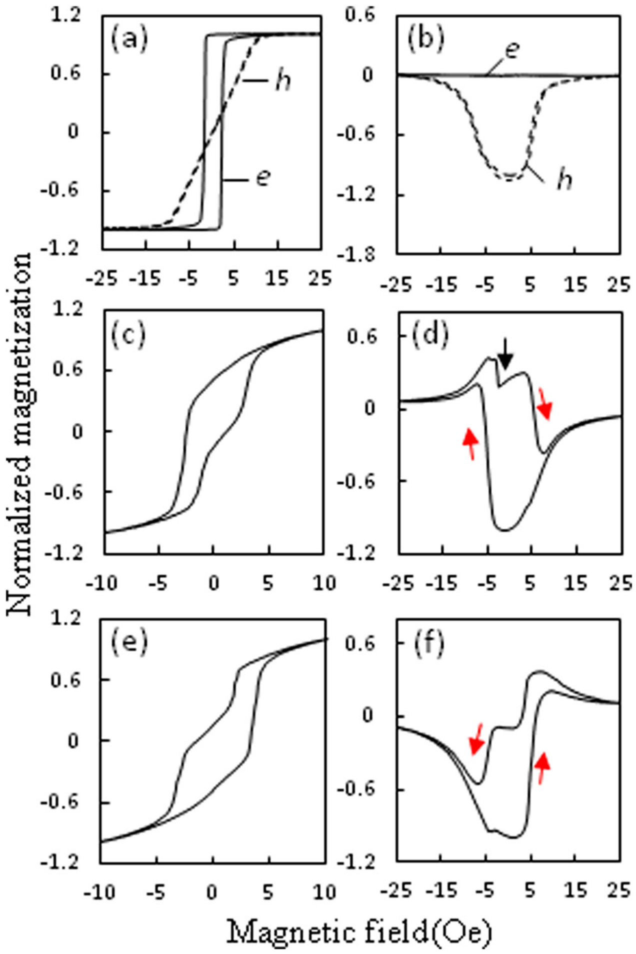

FIG. 1. Longitudinal and transverse hysteresis loops measured at angles of 0°/90° (a) and (b), 55° (c) and (d), and 125° (e) and (f).

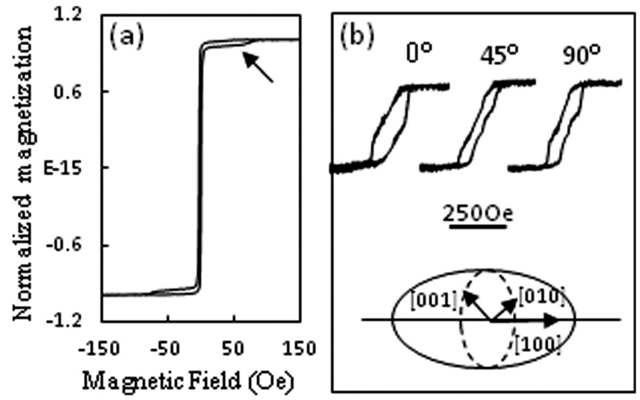

FIG. 2. Hysteresis loop taken at induced easy axis at enhanced field amplitude which shows a second phase (a) and extracted hysteresis loops of the second phase with different measurement angles and schematic illustration of proposed mechanism of biaxial formation in the hard phase (b).

ascending branch when the field is applied in one side of the hard axis ($45° < \theta < 90°$) then the kink will appear in the descending branch when the measured field is in the directions of another side ($90° < \theta < 135°$). Such observed asymmetry and polarity change has been further confirmed by measuring transverse hysteresis loops (Figs. 1(d) and 1(f)). Transverse loops measured in both easy and hard axes are also shown in Fig. 1(b) indicating a domain wall propagation and coherent rotation mechanism for the two cases. The observation of such asymmetric loops in our experiment was unexpected. To observe an asymmetric magnetic curve, a unidirectional anisotropy is required, otherwise the Onsager's reciprocal relation $\boldsymbol{M}(-\boldsymbol{H}) = -\boldsymbol{M}(\boldsymbol{H})$ will be violated.[25,26] It seems that there is no reason which could cause a unidirectional anisotropy, such as exchange bias between FM/AFM layers/phases, in our samples. In order to understand the observed magnetization curves, the samples were measured at field amplitude much larger than normally used for ultrasoft magnetic materials. Fig. 2(a) shows a hysteresis loop taken in the induced easy direction with field amplitude of 250 Oe. A small additional switch at ~80 Oe is identified on the hysteresis loop which is the signature of existing a magnetically harder "second phase" formed during the plating. As the soft material is nearly saturated at ~10 Oe, one can extract the hysteresis behavior of the hard phase by removing soft phase contribution in the hysteresis curves and renormalize magnetization data from hard phase. For the ascending branch of the loops [M↑(H)], we enlarge the magnetization recorded under fields between negative saturation and −10 Oe by adding the difference of magnetization at field 10 Oe and −10 Oe, i.e., $M = M\uparrow(<-10) + [M\uparrow(10) - M\uparrow(-10)]$, then the ascending branch is normalized. Similarly, the descending curve [M↓(H)] is normalized after enlargement of the magnetization recorded under fields between −10 Oe and negative saturation by adding the difference of magnetization at field 10 Oe and −10 Oe, i.e., $M = M\downarrow(<-10) + [M\downarrow(10) - M\downarrow(-10)]$. The extracted hysteresis loops of the second phase taken from different field directions show a biaxial anisotropy with one easy direction parallels to the induced easy direction (0°–180°) and another in its perpendicular direction (90°–270°) were found (Fig. 2(b)).

The hysteresis loops in Fig. 2(b) show that the two easy axes are not equivalent and the one in the plating direction is dominant. We explain this as the inset of Fig. 2(b): The second phase with a cubic crystalline symmetry was formed during the plating. One easy direction [100] of the small grains is aligned with external field applied during film plating, while other two axes, [010] and [001], randomly distributed in the (100) plane which is perpendicular to the field direction and the average effect gives a less dominant easy axis. We explain the observed asymmetric loops as Fig. 3. The second phase is magnetized in the O-E direction which is parallel to the induced easy axis E′-E for a newly deposited film. Let us examine a magnetization reversal when the field is scanning in the direction M-M′, which has an off-set angle from induced hard axis of the film H-H′. When the field is increased from negative saturation, the magnetization vector rotates towards the nearest easy direction (O-E′) where the magnetization vector does not go to positive saturation through routine M → E′ → M′ as normally expected for loops taken in direction close to hard axis, instead the magnetization prefer jumps into O-E direction through 180° domain wall motion due to the existing demagnetization force induced by the second phase (open arrow), and then rotates into positive saturation. Thus, the magnetization reversal is completed through routine M → E′ → E → M′

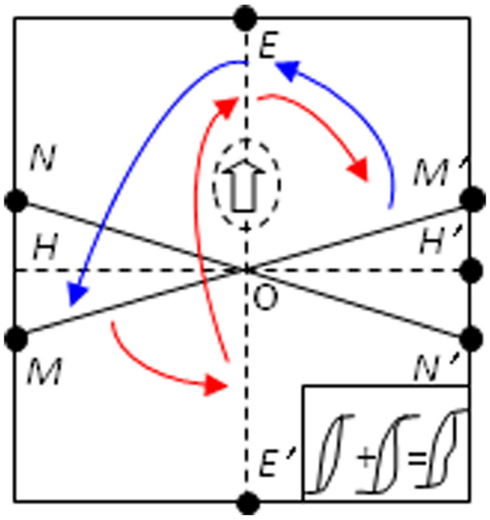

FIG. 3. Illustration of the asymmetric magnetization reversal mechanism and an inset shows a formation mechanism of kinked hysteresis loop.

(red arrows) rather than M → E′ → M′. When the field sweeps back, the vector will first adopt in easy direction O-E. As there is no demagnetizing effect from the second phase, in this case, the magnetization reversal is completed through routine M′ → E → M (blue arrows) which gives a normal magnetization curve. Similarly, if the field is applied in the direction N-N′ with an off-set angle from the hard axis in another side the reverse route will be N → E → N′ for the ascending and N′ → E′ → E → N for the descending field, respectively, which explains a changing polarity as shown in Fig. 1. The reason behind the 180° domain wall is minimization of energy of magnetization reversal. For a soft material with uniaxial anisotropy and low coercivity, the energy required for wall motion is much lower than that for the magnetization rotation. However, the wall motion is generally initiated from nucleated reverse domains and the applied field direction is close to easy axis. In our sample, the hard phase acts as nucleation centers when the magnetization of soft matrix is antiparallel to that of the hard phase and this causes a 180° domain wall motion as the field direction goes further from easy axis and an asymmetric hysteresis behavior appears.

We would like to address that such intermediate 180°DW sweep does not happen for whole sample, i.e., only a portion of the film is reversed by this mechanism and the remaining portion still obeys coherent rotation mechanism which is evidenced from a partial loss of magnetization on transverse magnetization curves (black arrow in Fig. 1(d)). This is caused by non uniform distribution of the size and location of the second-phase in the film. Such mixed reversal mechanism explains the kink observed on the magnetization curve, namely, the kink is induced by two independent reversal mechanisms in the film and the measured loop is formed by a superposition of normal and asymmetric reversals (inset of Fig. 3).

The 180°DW-sweeping is a common magnetization reversal mechanism when a loop is taken in/near easy directions. The striking thing observed here is that it can happen in the vicinity of hard axis which can be better understood through following discussion. In our system, the energy per unit volume can be expressed by a sum of energies of uniaxial and unidirectional anisotropies and Zeeman energy

$$E = K_U \sin^2\vartheta + K_E(1-\cos\vartheta) - MH\cos(\vartheta-\varphi), \tag{1}$$

where ϑ and φ are angles between magnetization M, magnetic field H, and induced easy axis, respectively. K_u is induced uniaxial anisotropy and K_E represents a unidirectional exchange energy contribution induced by the second phase. As we discuss the 180°-DW motion mediated magnetization switch, the behavior of spins close to domain nucleation center (i.e., those spins which are exchange coupled with the second phase) is important, thus the introduction of K_E is justified. We consider the case when a field is applied in MM′ direction as shown in Fig. 3. If the 180° DW sweeping mediated switching is originated by energetic advantage of switching E′ → E (to occupy a stable magnetic state) over E′ → H′ (to overcome a maximum energy barrier) then a ΔE (E′ → E) < ΔE(E′ → H′) is satisfied. The energies at points E, E′, and H′ can be calculated from Eq. (1)

$$E(E) = -MH_D\cos\varphi, \tag{2a}$$

$$E(E') = 2K_E + MH_D\cos\varphi, \tag{2b}$$

$$E(H') = K_U + K_E - MH_K\sin\varphi, \tag{2c}$$

where H_D is the field at which the 180° DW sweeping happens, while H_K is the field of induced anisotropy of the film. When ΔE (E′ → E) < ΔE(E′ → H′) is considered

$$\sin\varphi < \frac{1}{2} + \frac{K_E}{MH_K} + \frac{H_D}{H_K}\cos\varphi. \tag{3}$$

In Eq. (3), the third term in the right can be ignored as $H_K \gg H_D$ and $\cos\varphi$ is small, especially in the vicinity of hard axis, and then we have

$$\varphi < \sin^{-1}\left(\frac{1}{2} + \frac{K_E}{MH_K}\right). \tag{4}$$

From Eq. (4), we can see that if there is no second phase contribution ($K_E = 0$), the DW sweeping is dominated mechanism when $\vartheta < \pi/6$, i.e., in/near easy axis direction, agrees with that commonly observed. For observing the 180° DW mediated reversal at large angle, for instance $\pi/3$, the K_E need to be $\sim H_K M/3$. Although the introduction of K_E can explain the observed loop asymmetry, there are still open questions to answer, for instance, no obvious loop shift induced by hard phase has been found (see Fig. 1(a)).

To gain further insight, we have measured hysteresis loops after pre-saturating the sample at 500 Oe in the induced hard direction in which the magnetization of the second phase is setting. As expected, no kinked loops were observed for any field direction. Figs. 4(a) and 4(b) show both longitudinal and transverse loops measured at ∼55° which are normal magnetization curves rather than kinked. If we pre-saturate the sample in the induced easy axis again the kinked loops were restored. Another notable feature which is different

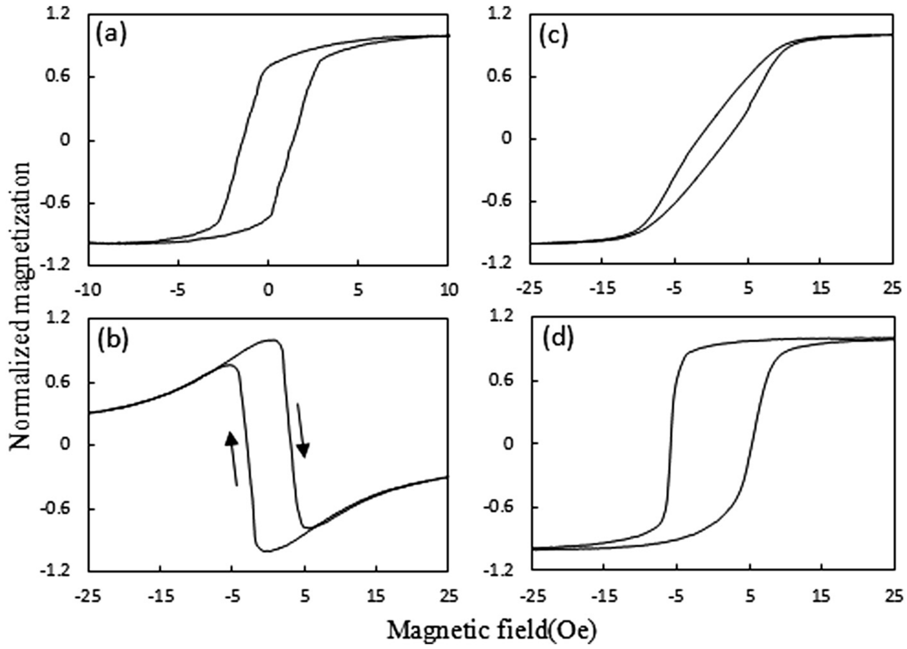

FIG. 4. Hysteresis loops taken from same sample (as Fig. 1) after saturation in its induced hard axis (a)–(c). (a) and (b) longitudinal and transverse loops taken in 55° direction. (c) Hysteresis loop taken in the induced hard direction. (d) A hysteresis loop taken from a sample with ~3 mm × 3 mm size and 250 nm thick which shows asymmetric switch behaviour.

from the case of easy-axis-presaturation is the appearance of obvious coercivity (~2 Oe) and remanence (~0.2) with the hard axis loop (Fig. 4(c)), in contrast to the non-hysteresis hard axis loops for the easy-axis presaturated case (Fig. 1(a)), and this further confirms the biaxial behavior of the second phase. We did not see obvious loop shift for both easy and hard axis hysteresis loops after saturating samples in hard axis.

The samples we plated and measured in this work were 3 cm by 3 cm. If we dice the samples into smaller sizes (less than 3 mm by 3 mm), we have found that the kink disappeared in most of the samples, where asymmetric loop with one branch switched by 180° wall motion (Fig. 4(d)) and standard hysteresis (similar to Fig. 4(a)) can be observed instead. This has further proved two different switching mechanisms coexist in the samples.

We have also attempted to understand the reason that might cause the second phase, and a TEM and EDX analyses were performed. The result shows that the plated film is polycrystalline NiFe alloy with Ni-element-rich regions (dark area, Fig. 5) with average film composition of 64%Ni + 36%Fe. The diffraction shows identical crystalline structure (FCC) for both NiFe matrix and Ni rich regions, which is more likely Ni_3Fe.[27] Although the coercivity of Ni_3Fe nanoparticle assembly is in the range of ~80 Oe (Ref. 28), it should be much lower when they are embedded in soft magnetic matrix. The reason of getting the second phase in the electroplating process is not clear at the moment.

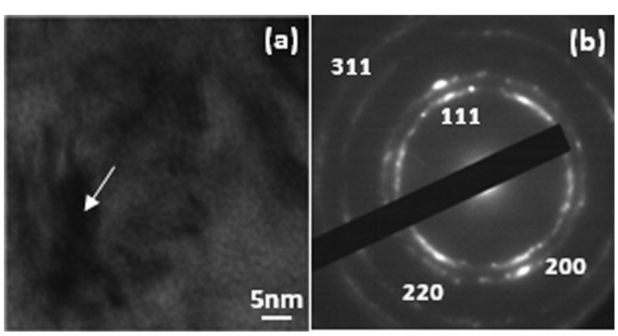

FIG. 5. TEM image of the plated film (a) and diffraction pattern (b).

In summary, we have observed an erase/restorable asymmetric magnetization reversal in polycrystalline soft magnetic film. If the film is pre-saturated at a high field in the induced uniaxial easy direction, one branch of the hysteresis loop is governed by "coherent rotation" mechanism and another branch with kink is induced by a mixed reversal mechanism of "coherent rotation" and "rotation/180°DW-swiping/rotation." The kinked loop can be erased by saturating the film in hard axis and restored by saturating the film in the induced easy axis again. The observed phenomenon opens a new avenue in engineering magnetization switch by artificially generating unidirectional anisotropy with designed direction, symmetry, strength, etc., for developing new type of magnetic devices such as sensor that enables to sense both field direction and amplitude.

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