Domain wall asymmetries in Ni$_{81}$Fe$_{19}$/NiO: proof of variable anisotropies in exchange bias systems

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Abstract. Multiple changes in the internal structure of magnetic domain walls due to alterations of the interfacial coupling across the ferromagnetic/antiferromagnetic interface are reported for Ni$_{81}$Fe$_{19}$/NiO exchange coupled films. Depending on the antiferromagnetically induced anisotropy, three different types of domain walls are observed. Cross-tie domain wall structures of decreased vortex to anti-vortex spacing develop with the addition of a thin antiferromagnetic layer. For exchange biased samples strong asymmetries in domain wall structure occur for the ascending and descending branch of the magnetization loop. For the descending branch a symmetric 180° Néel wall develops, whereas a folded cross-tie domain wall structure forms during magnetization reversal along the ascending loop branch. The novel type of ‘zig-zagged’ cross-tie wall is characterized by cross-ties reaching differently into the surrounding domain areas. The wall alterations indicate the existence of bi-modal coupling strengths in exchange coupled systems, which is in accordance with models of exchange bias that assume pinned and unpinned spins at the ferromagnetic/antiferromagnetic interface.

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1. Introduction

Exchange biased magnetic multilayers are one of the key components in current high-sensitivity magneto-resitive devices [1]. The exchange bias effect is associated with a magnetic loop shift [2] that occurs in coupled ferromagnetic/antiferromagnetic (F/AF) thin films. Despite the technological relevance of exchange biased systems in spintronic applications, and extensive experimental as well as theoretical investigations, the nature of exchange coupling between the spins of an F layer and an adjacent antiferromagnetic AF layer is still under discussion. An exchange bias field $H_{eb}$ [3]–[5] is found, the value of which can be predicted by different theories [6]–[12]. Recent experimental evidence [13, 14] indicates that for many material systems only a small fraction of uncompensated pinned spins at the interface between the F and AF contributes to the exchange bias effect. In addition, a larger remaining fraction of coupled but rotatable spins exists, which does not contribute to the magnetic loop shift. Hence, for an alignment of the F layer magnetization along the exchange bias direction, the pinned and unpinned interfacial moments are aligned in parallel. For the state of antiparallel alignment of F layer magnetization and unidirectional anisotropy, the pinned and unpinned moments are oriented antiparallel. As a consequence, the effective coupling or AF-induced anisotropy acting on the F layer should change with reversal and alignment of the F layer relative to the direction of exchange bias.

One indication of such an interaction in exchange biased samples is derived from magnetometry and magnetoresistance measurements [15]–[18] as well as magnetic domain observations [19]–[25], which reveal asymmetric remagnetization processes. Nevertheless, no direct observation of the magnetic domain wall structure itself has been made so far.

For single-layer F films, it is known that the domain wall structure depends on the F layer thickness $t_F$ and magnetic anisotropy [26]. Essentially, three different types of $180^\circ$ domain walls are observed in magnetic films: symmetric Néel walls, cross-tie walls and asymmetric Bloch walls. Cross-tie walls are characterized by an alteration of Bloch-like vortex and anti-vortex transitions along the domain wall segmented by $90^\circ$ Néel walls of opposite chirality (figure 1). As the specific energy of a $90^\circ$ Néel wall is significantly lower than that of a $180^\circ$ Néel wall, the complicated $90^\circ$ domain wall network of the cross-tie wall saves energy compared with a regular $180^\circ$ domain wall. In the cross-tie arrangement the magnetic flux of the long-range magnetic tails is partially closed in the vicinity of the walls. The occurrence of cross-tie walls in low anisotropy Ni$_{81}$Fe$_{19}$ films is limited to a narrow $t_F$ range. Below $t_F \approx 10$ nm $180^\circ$ Néel walls and above $t_F \approx 100$ nm asymmetric Bloch walls form. In the intermediate F layer thickness of
cross-tie wall stability, the cross-tie spacing $\lambda_{ct}$ scales reversely with the magnetic anisotropy energy constant $K_{u,\text{eff}}$ or anisotropy field $H_{k,F}$. This behavior is found experimentally $[27]–[29]$ and only basic features can be modeled theoretically $[27, 28]$. For a given film thickness, the experimentally found dependency of $\lambda_{ct}$ with anisotropy of a single F layer is

$$\lambda_{ct} \propto \left( K_{u,\text{eff}} \right)^{-n} = \left( H_{k,F} \right)^{-n}, \quad (n = 0.5, \ldots, 1.0), \quad (1)$$

where the exponent $n$ depends on the F layer thickness $[28]$. This relation provides merely a rough guidance for the change of $\lambda_{ct}$ with anisotropy and is only valid for small values of anisotropy. Nevertheless, $\lambda_{ct}$ serves as a sensitive measure of changes in the effective anisotropy in ferromagnetic thin films. Above a certain value of anisotropy the cross-tie wall transforms into a symmetric $180^\circ$ Néel wall $[29]$. This is due to an increased energy contribution from the Bloch lines inside the domain wall with smaller cross-tie spacing, i.e. increased Bloch line density.

In this paper, we directly analyze the magnetization structure of cross-tie domain walls with constant F layer thickness, but varying NiO layer thickness $t_{\text{NiO}}$. The measured cross-tie spacing is used as an indicator for the effective AF induced anisotropy acting on the F layer. As demonstrated, the observation of the domain wall structure opens the unique opportunity to probe changes in the effective magnetic anisotropies during reversal, simultaneously for the switched and non-switched regions. From the alterations in domain wall structure and the reversal asymmetry we are able to derive conclusions on the effective anisotropy contributions arising from the AF layer. The cross-tie domain wall structure acts as a micromagnetic sensing tool for variable anisotropy states in exchange coupled systems.

2. Experimental details

Si/SiO$_2$/Ta(4 nm)/Ni$_{81}$Fe$_{19}$(30 nm)/NiO(0–50 nm) F/AF structures were prepared by magnetron sputtering in a high vacuum sputter system with a base pressure below $2 \times 10^{-7}$ mbar at an Ar pressure of $8 \times 10^{-4}$ mbar. The Ta seed layer ensures a (111)-textured growth of the polycrystalline films. The deposition rate for the NiO layer was 1.7 nm min$^{-1}$. The F uniaxial anisotropy axis and the exchange bias direction were set in an applied magnetic in-plane field.

Figure 1. Domain model of a cross-tie domain wall structure with a characteristic cross-tie spacing $\lambda_{ct}$ [26]. The vortex ($\circ$) and anti-vortex ($\bullet$) structures, as well as the principal alignment of magnetization are indicated.
Figure 2. Hysteresis loops along the EA and HA of magnetization, respectively, easy direction and hard direction of exchange bias in selected Ni$_{81}$Fe$_{19}$ (30 nm)/NiO structures. The NiO film thickness is (a) 0 nm, (b) 5 nm, and (c) 30 nm. The coercive fields $H_{c1}$ and $H_{c2}$ are defined in (c). (d) NiO thickness $t_{\text{NiO}}$ dependence of exchange bias $H_{\text{eb,EA}}$, anisotropy field $H_{k,\text{eff}}$ and coercivity $H_{c,\text{EA}}$. The positions of $t_{\text{NiO}}$ corresponding to (a)–(c) are indicated.

of $H_{\text{dep}} = 20$ kA m$^{-1}$ during film deposition. No post-annealing was performed. Inductive magnetometry at 10 Hz was used to characterize the magnetic properties of the films. The formation of ferromagnetic domains was imaged by real-time magneto-optical Kerr microscopy [26] in the longitudinal mode through the covering NiO AF layer. All experiments were carried out at room temperature.

3. Results and discussion

3.1. Magnetometry

Hysteresis loops for the pure F film and double layers with two different NiO thicknesses are displayed in figure 2. For the thinnest NiO layer ($t_{\text{NiO}} = 5.0$ nm, figure 2(b)) an increase in coercivity $H_{c,\text{EA}}$ along the easy axis (EA) is seen. This change is accompanied by the formation of a double-stage magnetization curve visible in the hard axis (HA) loop. A similar two-step magnetization reversal was also reported in [30]. This S-shaped HA loop was interpreted as

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an AF induced uniaxial anisotropy field $H_{k,AF}$, only existing at low fields and ‘unsnapping’ at higher magnetic fields [30, 31]. With increasing AF layer thickness, $t_{NiO} = 30$ nm is shown in figure 2(c), a magnetic loop shift is observed along the EA. The coercivity has again decreased relative to the sample with $t_{NiO} = 5$ nm. From the HA loop the effective field consisting of anisotropy field $H_{k,F}$ and exchange bias field $H_{eb}$ acting on the F layer could in principle be derived. However, comparing the effective field to the value of $H_{eb,EA}$, as derived from the EA loop shift, a discrepancy becomes obvious, which we treat as an AF induced uniaxial anisotropy field $H_{k,AF}$. From this we extract the effective anisotropy fields $H_{k,eff}$ acting on the F layer by taking the following contributions into account [31]:

$$H_{k,tot} = H_{k,F} + H_{k,AF} + H_{eb,EA} = H_{k,eff} + H_{eb,EA}. \hspace{1cm} (2)$$

The dependences of $H_{eb,EA}$, $H_{k,eff}$ and the coercivity values $H_{c,EA}$ with $t_{NiO}$ are summarized in figure 2(d). The coercivity peaks at the onset of exchange bias at an AF thickness of $t_{NiO} = 10$ nm and then decreases with the occurrence of exchange bias. This dependency can be interpreted as a transformation of a ‘mobile’ AF spin structure into an AF ‘static’ domain structure with stabilization of the AF magnetization [10, 32] with increasing AF thickness. Correlated to the coercivity is the dependency of $H_{k,eff}$, which peaks with $H_{c,EA}$ and decreases with the onset of $H_{eb,EA}$. Even for a stable exchange bias, a uniaxial-like anisotropy contributes to the magnetization reversal. A detailed discussion of the different anisotropy contributions in NiFe/NiO layers can be found in [31].

### 3.2. Domain wall observation

Additional evidence for the existence of an AF induced uniaxial-like anisotropy contribution to the reversal process is derived from the magnetic domain wall structure in the bilayer films. Domain images of a single Ni$_{81}$Fe$_{19}$ layer and a Ni$_{81}$Fe$_{19}$/NiO (5 nm) structure are shown in figures 3(a) and (b), respectively. Adding the thin NiO layer, a drastic decrease in cross-tie spacing of the F layer from $\lambda_{ct,F} = 11.5 \mu m$ down to $\lambda_{ct,F,AF} = 2.6 \mu m$ for the exchange coupled but zero loop shift sample is found. The reduction in $\lambda_{ct,F}$ is due to an increase of effective anisotropy $H_{k,eff}$ with the addition of the NiO layer and confirms the additional anisotropy.
Figure 4. Cross-tie domain wall structures in a Ni$_{81}$Fe$_{19}$/NiO bilayer with $t_{\text{NiO}} = 30$ nm. Different wall structures are observed along the descending (a) and ascending (b) loop branches at $H_{c2}$ and $H_{c1}$, respectively. The magneto-optical sensitivity is aligned vertically.

contribution to the F layer that arises from the AF layer. The narrow Bloch line spacing in the cross-tie wall is already close to the stability limit of cross-tie wall formation. This is evident in figure 3(b), where a segment of the domain wall film has already transformed into a regular symmetric 180° Néel wall. A similar lower limit of cross-tie spacing ($\lambda_{\text{ct,min}} \approx 3 \mu$m) was found for amorphous FeCoBSi single-layer films with variable uniaxial anisotropy [29]. This lower limit can be undercut for confined structures [33], in which the spreading of the Néel wall tails is limited.

Additional transformations in the domain wall structure are observed with further increasing NiO thickness. For the two magnetization reversal paths along EA an asymmetry in domain wall type is found. In figures 4(a) and (b), the domain wall structures in a Ni$_{81}$Fe$_{19}$/NiO (30 nm) bilayer during the magnetic switching events at $H_{c1}$ and $H_{c2}$ (see EA loop in figure 2(c)) are shown. During reversal, neither signs of coherent or incoherent rotation of magnetization, nor signs of loop asymmetry are found. However, for the descending loop branch (figure 4(a)) no cross-tie domain wall structure is visible. Due to a high effective anisotropy acting on the F layer, a symmetric 180° Néel wall instead of a cross-tie wall develops. This observation is consistent with an increase in effective anisotropy beyond the case shown in figure 3(b). For the ascending loop branch, however, the reversal process takes place by the movement of an almost regular cross-tie wall. The cross-tie period of $\lambda_{\text{ct,F-AF}} = 7.5 \mu$m lies between the low and high anisotropy cases shown in figures 3(a) and (b), respectively. This clearly indicates that the average effective anisotropy, which acts on the F layer along the ascending loop, branch is lower as for the descending loop branch. Nevertheless it is still higher than for the single F layer case with narrow cross-tie spacing. The alteration in domain wall structure in dependence of the magnetic history verifies the existence of asymmetric AF anisotropy contributions that change with the reversal of the ferromagnetic layer and which lead to the observed asymmetric domain wall behavior.

In addition, the cross-tie wall structure is further modified as compared with the regular wall presented in figure 1. Notably, the cross-ties extend further into the surrounding domain of the non-switched portion of the film (bottom domain in figure 5(a)), indicating different effective anisotropies at both sides of the domain wall. A detailed sketch of the asymmetrically shaped cross-tie wall similar to figure 1 is given in figure 5(b). The asymmetrically shaped domain
Figure 5. (a) Magnification of the folded cross-tie domain wall of figure 4(b) together with (b) a schematic sketch of the domain wall structure (see figure 1 for comparison).

wall proves that with the switching of the F layer the effective coupling field also changes. The asymmetric magnetization and effective anisotropy distribution in the distorted cross-tie structure is also evident from the ‘zig-zagging’ of the domain wall. The wall is folded by an angle $\Theta \approx \pm 10^\circ$ relative to the net direction of the wall. The asymmetry in the domain wall structure is qualitatively illustrated by differently tilted magnetization vectors on both sides of the wall in figure 5(b). The values of the effective anisotropy before and after switching, respectively on both sides of the domain walls, cannot be determined directly from the images. However, from simple micromagnetic arguments, by minimizing the magnetic charges across the domain wall segments and assuming a symmetric anisotropy energy contribution from both sides of the domain wall, the effective magnetic anisotropy ratio in the domains can be derived. It is expressed by

$$
\frac{H_{k_{\text{eff1}}}}{H_{k_{\text{eff2}}}} = \frac{K_{u_{\text{eff1}}}}{K_{u_{\text{eff2}}}} = \frac{\sin^2(\Theta_2)}{\sin^2(\Theta_1)} = \frac{\sin^2(\pi/4 - \Theta)}{\sin^2(\pi/4 + \Theta)} = 2.04 \approx 2, 
$$

where $K_{u_{\text{eff1}}}$, $K_{u_{\text{eff2}}}$, and $H_{k_{\text{eff1}}}$, $H_{k_{\text{eff2}}}$ are the effective anisotropy energy constants, respectively magnetic anisotropy fields, of the regions surrounding the domain wall. $\Theta_1$ and $\Theta_2$ are the corresponding angles of magnetization of the cross-ties as labeled in figure 5. With a folding angle $\Theta = 10^\circ$, i.e. a change in magnetization angle of $\pm \Theta$ relative to the regular cross-tie wall configuration ($\Theta_1 = \Theta_2 = \pi/4$), a change of anisotropy during switching by a factor of 2 is derived. Hence, the novel type of folded cross-tie wall directly evidences the modification of the effective anisotropy and the simultaneous occurrence of bi-modal anisotropy states during the switching of magnetization in the exchange biased ferromagnet.

4. Summary

In conclusion, we identified drastic changes of the internal domain wall structure in exchange coupled thin films. The observed changes in the internal ferromagnetic domain wall structure are concurrent with varying anisotropy contributions from the AF layer in Ni$_{81}$Fe$_{19}$/NiO films. Different types of characteristic wall structures form during magnetization reversal along the forward and recoil branches of the hysteresis loop for films exhibiting exchange bias. Asymmetric anisotropy fields acting on the F layer during reversal induce an asymmetrically jagged type of cross-tie domain wall structure. This wall folding indicates the existence of unstable AF magnetization contributions, resulting in changes in the effective anisotropy.
This is the first direct observation of domain wall transformations during magnetization reversal due to antiferromagnetic exchange coupling. The domain walls act as an indicator for variable anisotropy changes in F/AF thin film systems.

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References

[1] Gurney B A, Speriosu V S, Wilholt D R, Lefakis H, Fontana R E Jr, Heim D E and Dovek M 1997 Can spin valves be reliably deposited for magnetic recording applications? J. Appl. Phys. 81 3998–4003
[2] Meiklejohn W H and Bean C P 1956 New magnetic anisotropy Phys. Rev. 102 1413–4
[3] Berkowitz A E and Takano K 1999 Exchange anisotropy—a review J. Magn. Magn. Mater. 200 552–70
[4] Noguès J and Schuller I K 1999 Exchange bias J. Magn. Magn. Mater. 192 203–32
[5] Stamps R L 2000 Mechanisms for exchange bias J. Phys. D: Appl. Phys. 33 R247–68
[6] Malozemoff A P 1988 Mechanisms of exchange anisotropy J. Appl. Phys. 63 3874–79
[7] Takano K, Kodama R H, Berkowitz A E, Cao W and Thomas G 1997 Phys. Rev. Lett. 79 1130
[8] Koon N C 1996 Calculations of exchange bias in thin films with ferromagnetic/antiferromagnetic interfaces Phys. Rev. Lett. 78 4865–67
[9] Stiles M D and McMichael R D 1999 Model for exchange bias in polycrystalline ferromagnet–antiferromagnet bilayers Phys. Rev. B 59 3722–33
[10] Nowak U, Misra A and Usadel K D 2001 Domain state model for exchange bias J. Appl. Phys. 89 7269–71
[11] Suess D, Kirschner M, Schrefl T, Fidler J and Stamps R L 2003 Exchange bias of polycrystalline magnets with perfectly compensated interfaces Phys. Rev. B 67 054419
[12] Radu F and Zabel H 2007 Exchange bias effect of ferro-/antiferromagnetic heterostructures Magnetic Heterostructures (Springer Tracts in Modern Physics vol 227) (Berlin: Springer) pp 97–184
[13] Ohldag H, Scholl A, Nolting F, Arendt E, Maat S, Young A T, Carey M and Stöhr J 2003 Correlation between exchange bias and pinned interfacial spins Phys. Rev. Lett. 91 017203
[14] Schlagel R, Rohlsberger R, Klein T, Burkel E, Strohm C and Ruffer R 2009 Spatially resolved magnetic reversal in a multilayered exchange bias system New J. Phys. 11 013043
[15] Fitzsimmons M R, Yashar P, Leighton C, Schuller I K, Nogués J, Majkrzak C F and Dura J A 2000 Asymmetric magnetization reversal in exchange-biased hysteresis loops Phys. Rev. Lett. 84 3986–9
[16] Leighton C, Song M, Nogués J, Cyrille M C and Shuller I K 2000 Using magnetoresistance to probe reversal asymmetry in exchange biased bilayers J. Appl. Phys. 88 344–7
[17] McCord J, Mattheis R and Elefant D 2004 Dynamic magnetic anisotropy at the onset of exchange bias: The NiFe/IrMn ferromagnet/antiferromagnet system Phys. Rev. B 70 094420
[18] Camarero J, Sort J, Hoffmann A, García-Martín J, Dieny B, Miranda R and Nogués J 2005 Origin of the asymmetric magnetization reversal behavior in exchange-biased systems: competing anisotropies Phys. Rev. Lett. 95 057204
[19] Nikitenko V I, Gornakov V S, Shapiro A J, Shull R D, Kai Liu, Zhou S M and Chien C L 2000 Asymmetry in elementary events of magnetization reversal in a ferromagnetic/antiferromagnetic bilayer Phys. Rev. Lett. 84 765–8
[20] Wang Y G, Petford-Long A K, Hughes T, Laidler H, O’Grady K and Kief M T 2002 Magnetisation reversal of the ferromagnetic layer in IrMn/CoFe bilayer films J. Magn. Magn. Mater. 242–245 1073–6

New Journal of Physics 11 (2009) 083016 (http://www.njp.org/)
[21] de Haas O, Schäfer R, Schultz L, Schneider C M, Chang Y M and Lin M T 2003 Critical angle for irreversible switching of the exchange-bias direction in NiO-Cu-Ni$_{81}$Fe$_{19}$ films Phys. Rev. B 67 054405
[22] McCord J, Mattheis R and Schäfer R 2003 Kerr observations of asymmetric magnetization reversal processes in CoFe/IrMn bi-layer systems J. Appl. Phys. 93 5491–7
[23] Blomqvist P, Krishnan K M and Ohldag H 2005 Direct imaging of asymmetric magnetization reversal in exchange-biased Fe/MnPd bilayers by x-ray photoemission electron microscopy Phys. Rev. Lett. 94 107203
[24] Widuch S, Celinski Z, Balin K, Schäfer R, Schultz L, Skrzypek D and McCord J 2008 Variation in ferromagnetic domain density and domain asymmetry in Fe/FeF$_2$ exchange bias structures Phys. Rev. B 77 184433
[25] McCord J, Hamann C, Schäfer R, Schultz L and Mattheis R 2008 Nonlinear exchange coupling and magnetic domain asymmetry in ferromagnetic/IrMn thin films Phys. Rev. B 78 094419
[26] Hubert A and Schäfer R 2008 Magnetic Domains 3rd edn (Berlin: Springer)
[27] Middelhoeck S 1963 Domain walls in thin Ni-Fe films J. Appl. Phys. 34 1054–9
[28] DeSimone A, Kohn R, Müller S and Otto F 2005 Recent analytical developments in micromagnetics The Science of Hysteresis vol 2 ed G Bertotti and I Mayergoyz (Amsterdam: Elsevier) chapter 4 pp 269–381
[29] Schäffel F 2004 Observation of domain walls in amorphous CoZrTa and FeCoBSi thin films Diploma Thesis Technical University of Dresden
[30] Zhao T, Fujiwara H, Zhang K, Hou C and Kai T 2001 Enhanced uniaxial anisotropy and two-step magnetization process along the hard axis of polycrystalline NiFe/NiO bilayers Phys. Rev. B 65 014431
[31] McCord J, Kaltofen R, Gemming T, Hühne R and Schultz L 2007 Aspects of static and dynamic magnetic anisotropy in Ni$_{81}$Fe$_{19}$-NiO films Phys. Rev. B 75 134418
[32] Stiles M D and McMichael R D 2001 Coercivity in exchange-bias bilayers Phys. Rev. B 63 064405
[33] Wiese N, McVitie S, Chapman J N, Capella-Kort A and Otto F 2007 On the scaling behaviour of cross-tie domain wall structures in patterned NiFe elements Europhys. Lett. 80 57003