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Cross Type Domain in Exchange-Coupled NiO/NiFe Bilayers

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The dependences of microscopic magnetic domain on film thickness in unidirectional and isotropic exchange-coupled NiO/NiFe bilayers were investigated by magnetic force microscopy to better understand for exchange biasing. As NiO thickness increases, microscopic domain structure of unidirectional biased film changed to smaller and more complicated domains. However, for isotropic-coupled film a new *cross type domain* appeared with out-of plane magnetization orientation. The density of the cross domain is proportional to exchange biasing field, and the fact that the domain was originated by the strongest exchange coupling region was confirmed from the dynamic domain configuration during a magnetization cycle.

Key words : Cross Type Domain, Exchange-Coupled

1. Introduction

Exchange biasing (EB) at magnetically disordered ferromagnetic (FM)/antiferromagnetic (AFM) thin film interfaces has been of great interest to fundamental research as well as in practical applications. The most commonly observed properties are the shifted hysteresis loop and enhanced coercivity of the FM layer. To explain the shifted loop theoretically, several models focused on the domain structure of the AF layer for a compensated and uncompensated interfaces, assuming a single domain state or a uniform magnetization of the FM layers [1-5]. Although these theories can explain the magnitude of the loop shift in polycrystalline bilayer, they fail to account for the enhanced coercivity and the FM domain structure during magnetization reversal in terms of a spin coherent rotation model [6, 7]. Therefore, it is essential to directly observe the microscopic domain structure of the coupled FM layer.

Recently, the direct observations for magnetization reversal and domain structure in EB films have been conducted by several groups using magneto-optical indicator film imaging [8], Fresnel images [9], high-resolution interference-contrast-colloid technique [10], Kerr effects microscope [11], and magnetic force microscopy (MFM) [12-14]. Most optical measurements have an advantage for macroscopic domain configuration and magnetization reversal by large scan area of above 100 μm. These observations revealed a distinct asymmetry of the FM domain wall motion and nucleation in decreasing and increasing magnetization processes due to spiral spin rotation, acting as exchange spring [8], and a complicated and small FM domain size due to non-uniform nature of exchange coupling [10, 11]. However, from these macroscopic images, it is hard to well understand for microscopically inhomogeneous and unstable EB of NiO film with grain size of 10^1 nm. MFM image can give more detail microscopic configuration of complicated and ripple domain. However, the previous MFM images of NiO/Co bilayer [11] and small size patterned NiO/NiFe elements [12, 13] could not explain locally random-oriented coupling, because Co has a large coercivity and small EB field, and the patterned ones make a fine domains due to strong shape anisotropy.

We have revealed in this work a new *cross type domain* with magnetization of out-of plane in the isotropic exchange-coupled NiO/NiFe bilayer deposited without magnetic field, H_d. The microscopic domain structure of the isotropic coupled NiFe layer may represent more realized image for exchange coupling at interface than the unidirectional biased NiFe layer, deposited with H_d. In order to study the relation between a cross type domain and EB, we have investigated for the static and dynamic

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MFM images of the isotropic and unidirectional coupled bilayers with a different thickness and deposition rate.

2. Experimental

NiO was prepared by RF sputtering on Si/SiO at room temperature without oxygen gas, and NiFe films were deposited over the NiO film by dc sputtering with or without the H_d of 300 Oe to get in-plane unidirectional anisotropy. The structures are Si/SiO/NiO (0, 10, 30, 60 nm)/NiFe (5, 10 nm). The magnetic properties were characterized from anisotropic magnetoresistance curves using 4-point terminal method [18]. Topological and magnetic structures of NiFe and NiO films were measured by tapping mode AFM , MFM, and STM.

3. Results and Discussions

Figure 1 shows the dependences of EB field (H_{ex}) and coercivity (H_c) on NiO thickness in the isotropic and unidirectional coupled NiO(0~60 nm)/NiFe(10 nm) bilayers. As expected, the H_{ex} of the unidirectional bilayer (with H_d) was almost zero for NiO(10 nm), and increased to 75 Oe at NiO(60 nm). In generally, a thin NiO layer below a critical NiO thickness of about 30 nm, even if this thickness may be different as deposition condition, has an unstable AF grains and a weak EB with FM layer because of its low magnetocrystalline anisotropy, K_1 [5, 18]. Especially, the NiO(10 nm)/NiFe(10 nm) film revealed an unstable and noisy change of the resistance during the measuring anisotropy magnetoresistance curve. In the figure 1, we should note that the H_c increased a little as the NiO increased 30 to 60 nm, but the H_{ex} did more rapidly, and the H_c of NiO(10 nm) enhanced to 25 Oe in spite of a very weak exchange coupling, while the H_c of NiFe(10 nm) single layer is about 2 Oe.

Recently, several arguments were reported to explain the unclear phenomena for coercivity [8, 15-17, 19]. Khapikov *et al.* [17] suggested that both fluctuating and stable AF grains contribute to the H_c of coupled bilayer. Since a NiO polycrystalline film has a lower anisotropy K_1 of $\sim 10^6$ than that of CoO of $\sim 10^8$, the NiO(10 nm) bilayer contains the irreversible transitions due to an unstable AF grains. At thinner AF layer, as the magnetization of FM layer is rotated, the state of weak AF grains can become unstable and switch, and the H_c becomes dominantly enhanced by hysteretic loss in an AF grains. On the other hand, as the thickness increases, NiO film grows in fine columnar grain with a small diameter below 30 nm [10, 12], and the stable AF grains strongly couple with FM layer. The FM layer receives a locally different H_{ex} from a randomly oriented AF grains, and the magnetization reversal takes place at different external fields for different FM domains. Therefore, the breakup of the FM layer into small domains is essential for the increased coercivity. For a thicker NiO, the contribution of the inho-

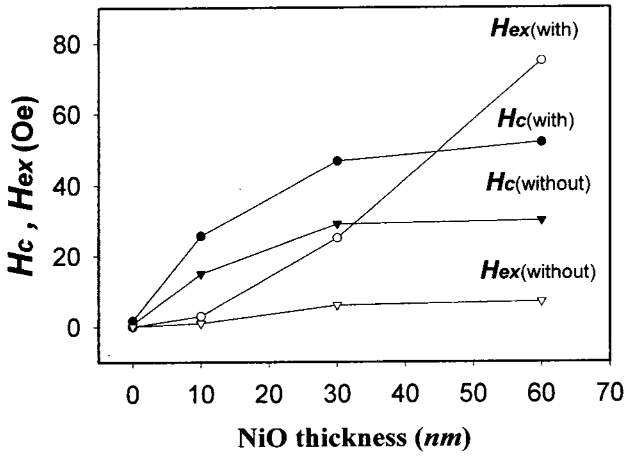


Fig. 1. The dependence of H_{ex} and H_c on NiO thickness in the isotropic (without H_d) and unidirectional (with H_d) coupled NiO(t nm)/NiFe(10 nm) bilayers, where the deposition rate of NiO is 3 Å/min.

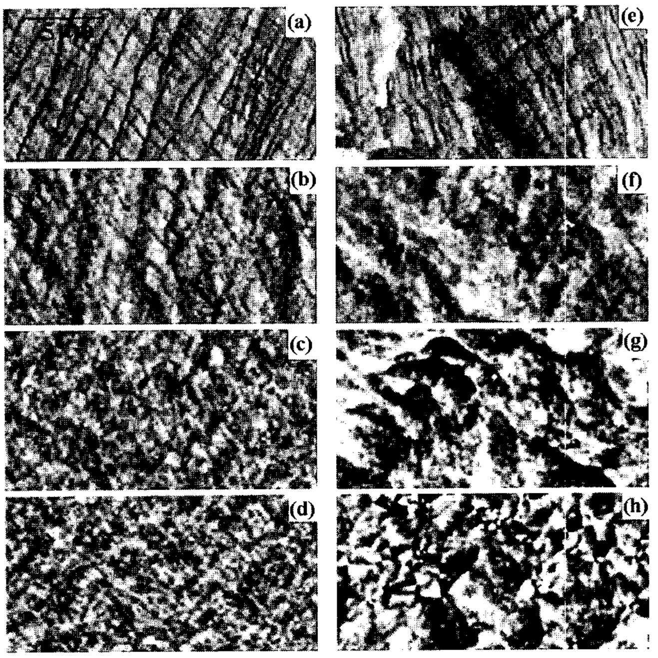


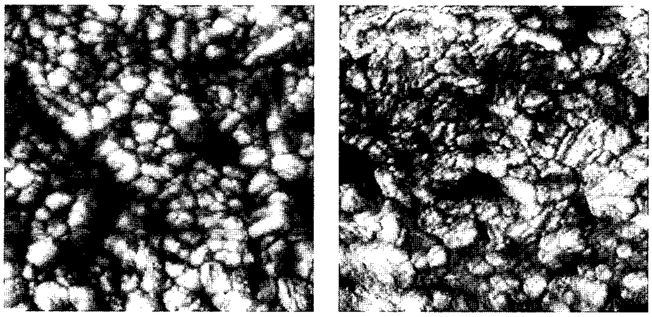
Fig. 2. The dependences of microscopic MFM images on NiO thickness in the NiO(t nm)/NiFe(10 nm) bilayer deposited with and without H_d, where t is (a, e) 0, (b, f) 10 nm, (c, g) 30 nm, and (d, h) 60 nm. The (a)~(d) images were measured from the bilayers with H_d and the (e)~(h) ones without H_d. The scan size of width is 20 μm. The deposition rate of NiO is 3 Å/min.

mogeneous reversal to coercivity becomes much larger than that of irreversible transition.

The microscopic NiFe domain structures in the isotropic and unidirectional exchange-coupled bilayers as increasing NiO thickness were measured. Figures 2(a) ~(d) show the evolution of the microscopic *ripple* domain pattern of the unidirectional bilayer from mesh shape with on NiO to a more complicated and coarse-grained structure as NiO increased to 60 nm. The mesh type ripples of NiFe film are the well-known longitudinal and transverse ripple due to small orientation variation in magnetization [20]. The ripple are easily removed at an applied field of 3 Oe. But the complicated ripple pattern of the NiO(60 nm) bilayer persists even up to 100 Oe, and its pattern preserved until magnetization direction was reversed, only its contrast changed. The small and strongly pinned ripples are certainly originated from the interfaces coupling of randomly oriented AF grains. However, it is difficult to distinguish the ripple structures between the bilayers having a different H_{ex}, because ones with H_d were forced to have a strong unidirectional magnetization to in-plane by the deposition magnetic field. For example, the H_{ex} of the unidirectional exchange-biased bilayers of NiO(60 nm)/NiFe(10 nm) is strongly dependent upon NiO deposition rate and NiFe thickness. It varied from 75 Oe for NiO(60 nm, 3 Å/min) to 103 Oe for NiO(60 nm, 12 Å/min). The STM images of the NiO(60 nm) films with the rates of 3 and 12 Å/min were observed at scan size of 200 nm, as shown in Figure 3. The average grain size is about 20 nm, and the NiO film deposited at 12 Å/min has a smoother surface than that of 3 Å/min. The enhancement of exchange biasing as increasing deposition rate is from smooth surface. Also as NiFe thickness decreases from 10 nm 5 nm, the H_{ex} was increased to 123 Oe. However, the ripple structures were identical.

To obtain more real domain configuration representing

scan size-200 nm



(a) NiO (60 nm, 3 A/min) **(b) NiO (60 nm, 12 A/min)**

Fig. 3. The STM images of NiO(60 nm, 3 Å/min) and NiO(60 nm, 12 Å/min). Scan size is 200 nm.

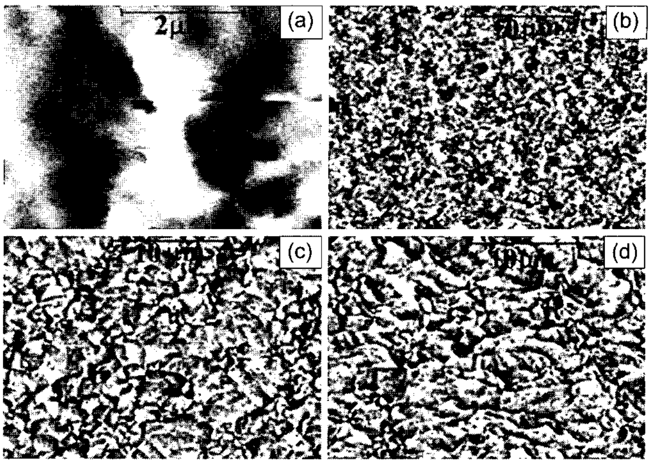


Fig. 4. (a) A typical cross-type domain (2 μm × 2 μm), the 10 mm scanned MFM images of (b) NiFe(10 nm) and (c) NiFe(5 nm) film over NiO(60 nm, 3 Å/min), and the 30 mm scanned MFM images of the NiO(60 nm)/NiFe(10 nm) bilayers with a different deposition rates of NiO as (d) 3 Å/min and (e) 12 Å/min. All samples were deposited without H_d.

a local coupling distribution, the MFM images of the isotropic coupled bilayers as increasing NiO thickness were measured, as shown in Figure 2(e)~(h). Note that as the NiO thickness increases, the magnetization direction was transferred from in plane to out-of plane, and we found that the magnetization domain in the NiO(60 nm) bilayer contained extraordinary domains, which we refer to as cross-type domain. The cross-type domain, as shown in the 2 μm scanned MFM image of Figure 4(a), has the strong magnetic poles to out-of plane and the sharp contrast at pole boundary, and irregularly spread in whole NiFe surface. In the Figure 4(b) and (c) for the NiFe(10 nm, 5 nm), the cross domains became smaller size and increased more contrast between poles as decreasing NiFe thickness. From the 30 μm scanned images of the bilayers with a different deposition rate in Figure 4(d), we can confirm the existence of the domain reflects a strong coupled regions, because the number of these domains is proportional to a macroscopically measured H_{ex}.

To explain the origin and structure of the cross-type domain a schematic diagrams of the interfacial cross section of the unidirectional and isotropic coupled FM layer over an (111) oriented and tilted AF grain are suggested in Figure 5. The schematic view of the tilted AF grain is based on the high-resolution TEM micrograph of NiO/Co interfaces measured by others Chopra *et al.* [10]. The NiFe film deposited without H_d over randomly oriented NiO grains would preferentially contain a higher density of out-of-plane moments in comparison with those deposited with an in-plane H_d field. The in-plane H_d field

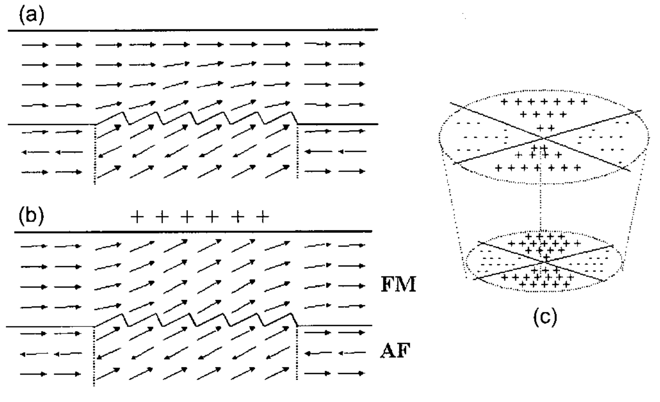
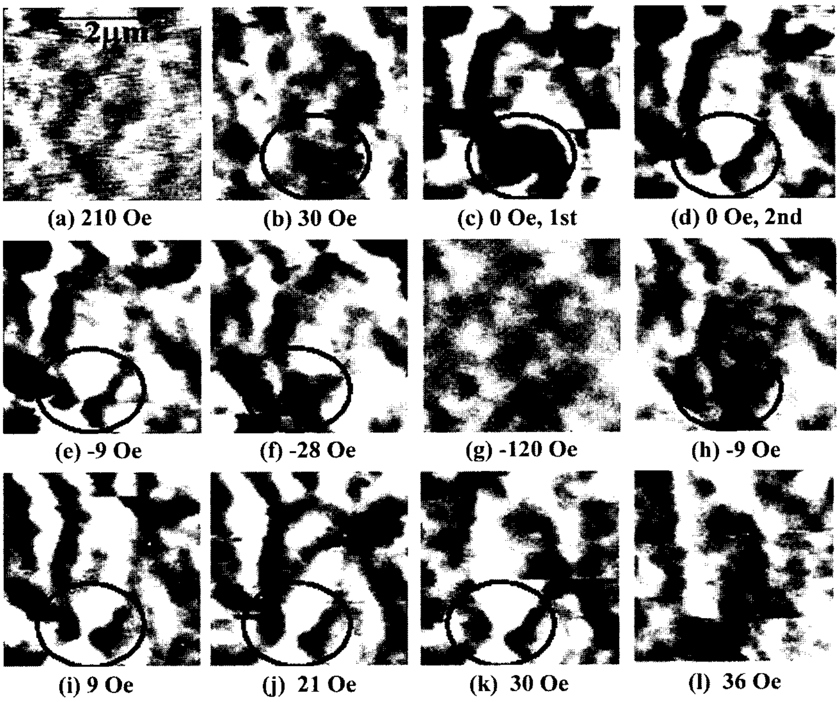


Fig. 5. A schematic views of the interfacial cross section of a (a) unidirectional and (b) isotropic exchange-coupled FM layer over a (111)-oriented and tilted AF grain. Each arrow represents average magnetization vector of several spins. (c) A schematic view of a typical cross-type domain with out-of poles.

forces the successive atomic layers to orient parallel to the surface, so that the number of vertical moments decreases with increasing thickness. As a consequence, the moments near at the surface of the 10 nm thick NiFe films are preferentially in-plane and lead to the MFM image in Figure 2(a)-(d). The complex ripple pattern observed for the exchanged biased films arise from the divergence of the in-plane magnetization, Div*M, possibly reflecting the grains of the underlying NiO films. A similar explanation is presumably valid for the case when the grown films are magnetically annealed. For the case isotropic films, the aligning field is absent during growth so that the ferromagnetic exchange interaction forces the NiFe film to follow the underlying moment of the EB layer which consequently lead to a large out-of-plane component. Despite the fact that the NiO grains are on the order of 10 nm, the strong ferromagnetic coupling between NiFe causes the formation of domains with out-of-plane magnetization. These domains are most like circular by symmetry considerations, and the center is quite possibly pinned by regions of locally strong exchange coupling with the NiO. As the NiFe thickness increases the size of the domains similarly increase as observed in our experiments. These domains possess high magnetostatic energy which is then reduced by the break-up of domains in the cross-type configuration as shown in the sketch in Figure 5. These cross-type domains are quite different from other systems that have out-of-plane magnetizations such as multiplayers used in perpendicular recording. In those systems, one invariably finds laryrinth or serpentine patterns which are expected in the absence of underlying pinning centers afforded by the ferromagnetic exchange coupling.

Finally, an alternative explanation for the observed effect is as follows. The grain sizes of 60 nm NiO films



(a) 210 Oe (b) 30 Oe (c) 0 Oe, 1st (d) 0 Oe, 2nd

(e) -9 Oe (f) -28 Oe (g) -120 Oe (h) -9 Oe

(i) 9 Oe (j) 21 Oe (k) 30 Oe (l) 36 Oe

Fig. 6. MFM images of cross type domain changing as applied field decreases (a) to (f), and increases (g) to (k). The cross domain is at the bottom of images, and the scratched lines in the images of (c), (e), and (k) were influenced by MFM tip.

observed by STM as shown in Figure 3 are about 20 nm, but the sizes of cross-type domains were about 1~2 μm in NiFe(10 nm). Although the domain size was reduced with decreasing NiFe thickness, it could not explain that each domain has originated from one tilted NiO grain. To overcome the controversy, our model can be supplemented from Takanos model for the uncompensated interfacial AF spins [4]. The cross domain will be caused by uncompensated pole of several NiO grains rather than one tilted grain. Also, even if the local out-of pole was from each tilted (111) grain, the macroscopically observed pole density will be reconstructed by a strong exchange coupling between FM spins. Conclusively, it is distinct that many cross-type domains reflect a high density of uncompensated moments at FM/AF interface, and these uncompensated moments generate a strong exchange biasing.

Figure 6 shows the dynamic variations of cross-type domain in NiO(60 nm, 3 Å/min)/NiFe(10 nm) bilayer during a magnetization cycle. As the external field is released from positive saturation, the strong density of out-of pole was created at zero field, as shown in Figure 6(c). Note that the scanned image (c) is different from (d) despite being obtained in a same region and field. Before creating cross-type domain at (d), a strong monopole density was concentrated at the same region, and the domain was created with some assistance from the MFM tip to reduce the pole energy. Also, when the cross domain disappears as increasing field, its pole intensity was stronger than that of other region, as shown in Figure 6(f). More importantly, we observed that despite difference elsewhere in the domain configurations with increasing and decreasing fields, the cross type domain invariably appeared at the some region and with nearly identical structure. It is a strong evidence that the domain was induced from the strongest exchange coupling of FM/AM interface.

In summary, we have observed a new cross-type domain with strong out-of poles in isotropic exchange-coupled NiO/NiFe film. From the dependences of thickness and deposition rate, the existence of the domain was directly related with exchange biasing field. The fact that the domain is originated from the strongest exchange-coupling region was confirmed by dynamic magnetic domain configurations during a magnetization cycle.

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References

[1] D. Mauri, H. C. Siegmann, P. S. Bagus, and E. Kay, J. Appl. Phys. **62**, 3047 (1987).

[2] A. P. Malozemoff, Phys. Rev. B **35**, 3679 (1987); J. Appl. Phys. **63**, 3874 (1988).

[3] N. C. Koon, Phys. Rev. Lett. **78**, 4865 (1997).

[4] T. C. Schulthess and W. H. Butler, Phys. Rev. Lett. **18**, 4516 (1998)

[5] A. E. Berkowitz and K. Takano, J. Mag. Mag. Mat. **200**, 552 (1999); Phys. Rev. Lett. **70**, 1130(1997).

[6] Z. Lie and S. Zhang, Phys. Rev. B **61**, R14 897 (2000); Appl. Phys. Lett. 77, 423 (2000).

[7] M. S. Stiles and R. D. Mcmichael, Phys. Rev. B **63**, 064405 (2001).

[8] V. I. Nikitenko et al., Phys. Rev. Lett. **84**, 765 (2000); Phys. Rev. B **57**, R8111 (1998); J. Appl. Phys. **83**, 6828 (1998); J. Magn. Magn. Mater. **198-199**, 500 (1999).

[9] X. Portier, A. K. Pettiford-Long, S. Mao, A.M. Goodman, H. Laidly, and K. OGrady, IEEE Trans. Magn. **35**, 3091(1999).

[10] H. D. Chopra, D. X. Yang, P. J. Chen, H. J. Brown, L. J. Swartzendruber, and W. F. Egelhoff, Jr, Phys. Rev. B **61**, 15312 (2000); J. Appl. Phys. **87**, 4942 (2000)

[11] Z. Qian, M. T. Kief, P. K. George, J. M. Sivertsen, and J. H. Judy, J. App. Phys. **85**, 5525 (1999).

[12] M. Cartier, S. Auffret, Y. Samson, P. Bayle-Guillemaud, B. Dieny, J. Magn. Magn. Mater. **223**, 63 (2001).

[13] J. Yu, A. D. Kent, and S. S. Parkin, J. Appl. Phys. **87**, 5049 (2000)

[14] J. Ding and J. Zhu, J. Appl. Phys. **79**, 5892 (1996); J. C. Wu, H. W. Huang, C. H. Lai, and T. H. Wu, J. Appl. Phys. **87**, 4948 (2000).

[15] M. D. Stiles and R. D. McMichael, Phys Rev. B **63**, 064405 (2001).

[16] Z. Li and S. Zhang, Appl. Phys. Lett. **77**, 423 (2000); Phys. Rev. B **61**, R14897 (2000).

[17] A. F. Khapikov, J. W. Harrell, H. Fujiwara, and C. Hou, J. Appl. Phys. **87**, 4954 (2000); Fujiwara *et al.*,.IEEE Trans. Magn. **35**, 3082 (1999).

[18] S. Soeya, S. Tadokoro, T. Imagawa, and M. Fuyama, J. Appl. Phys. **74**, 6297 (1993); G. H. Yu, *et al.*, J. Magn. Magn. Mater. **224**, 61 (2001).

[19] F. Y. Yang and C. L. Chien, Phys. Rev. Lett. **85**, 2597 (2000).

[20] M. Prutton, Thin ferromagnetic films (Butterworth, Washington D.C., 1964), p.162.