

Microscopic Domain Structures in NiO Exchange-coupled Films

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The dependence on nickel oxide thickness and a ferromagnetic layer thickness in unidirectional and isotropic exchange-coupled NiO/NiFe(Fe) bilayer films was investigated by magnetic force microscopy to better understand the relation between magnetic domain structure and exchange biasing at microscopic length scales. As the NiO thickness increased, the domain structure of unidirectional biased films formed smaller and more complex in-plane domains. By contrast, for the isotropically coupled films, large domains generally formed with increasing NiO thickness including a cross type domain with out-of plane magnetization orientation. The density of the cross domain is proportional to exchange biasing field, and the fact that the domain mainly originated from the strongest exchange coupled region was confirmed by imaging in an applied external field during a magnetization cycle.

Key words : exchange biasing, MFM/STM, cross type domain

1. Introduction

Recently, the direct observations of magnetization reversal and domain structure in exchange coupled ferromagnetic (FM)/antiferromagnetic (AF) films have been conducted by several groups [1-7]. Most optical measurements reveal the macroscopic domain configurations and magnetization reversals of large sample areas above 100 μm on a side. These observations revealed a distinct asymmetry of the FM domain wall motion and nucleation in decreasing and increasing magnetizing fields due to spiral spin rotation, acting as exchange spring [1], as well as the complicated FM structure due to non-uniform nature of exchange coupling [3, 4]. While these macroscopic observations are valuable, it is hard to discern the behavior of microscopically inhomogeneous and often unstable exchange biasing (EB) of NiO film with grain size of 10¹ nm or less. In the last few years, magnetic force microscopy was used to provide a more detailed view of complicated ripple domain. However, in MFM measurements by others on NiO/Co bilayer [5] and patterned NiO/NiFe elements [6, 7] the effects of locally random-oriented coupling were not addressed. In the former work,

Co, which has high coercivity (H_C) and small EB field (H_{ex}) was used; while in the latter, patterning created strong shape anisotropy induced domains. Thus, those experiments could mask the intrinsic effect of EB.

In this work, we studied continuous films of NiO/NiFe(Fe) bilayers by varying the thickness of the oxide film and the ferromagnet. Furthermore, the NiFe films were deposited with and without an external magnetic field (H_d), which produced unidirectional EB and isotropic EB films, respectively. Our motivation is that the microscopic domain structure of the isotropic-coupled NiFe layer may represent a more realistic picture for exchange coupling at the interface than the unidirectional biased NiFe layer. Among other things, we observed a *cross type domain* with magnetization of out-of plane in the isotropic exchange-coupled NiO/NiFe(Fe) bilayer.

2. Experimental

NiO was prepared by rf sputtering on Si/SiO at room temperature without oxygen gas, and the NiFe film was deposited over the NiO without breaking vacuum, by dc sputtering with or without the H_d of 300 Oe to get in-plane unidirectional or isotropic EB. The structures are Si/SiO/ NiO(0, 10, 30, 60 nm)/NiFe(10 nm). The magnetic properties were characterized from anisotropic magneto-

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resistance curves using 4-point terminal method. Topological and magnetic structures of NiFe film were subsequently measured by tapping mode AFM and MFM. The images of thin Fe films grown on polycrystalline NiO are measured in-situ by UHV STM/MFM measurements.

3. Results and Discussions

The H_{ex} of the unidirectional bilayer was almost zero for 10 nm NiO, and increased to 75 Oe at 60 nm NiO. In general, a thin NiO layer below a critical thickness of about 30 nm contains unstable AF grains and weak EB with FM layer because of its low magnetocrystalline anisotropy. With the thin AF layer, as the magnetization of FM layer is rotated, the weak AF grains themselves can become unstable and switch. On the other hand, as the thickness increases, NiO film grows in fine columnar grain with a small diameter below 30 nm [3, 5], and 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.

The aforementioned statements are elucidated by the MFM images of the NiFe domain structures of the isotropic and unidirectional exchange-coupled bilayers as functions of NiO thickness. Figures 1(a)-(d) show the evolution of the microscopic *ripple* domain pattern of the unidirectional bilayer from the mesh type pattern with no NiO to a more complicated and coarse-grained structure as NiO increased to 60 nm. The mesh type ripples of EB NiFe films are the well-known to be a combination of longitudinal and transverse ripples due to small variations in local magnetization [8]. The ripples are easily removed at an applied field of 3 Oe. But the complicated ripple pattern of the 60 nm NiO bilayer persists even up to 100 Oe, with the pattern preserved until magnetization direction was reversed. These small and strongly pinned ripples almost certainly originate from the interface 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 deposited with H_d were forced to have strong unidirectional in-plane magnetization by the magnetic field. For example, we established that the H_{ex} of the unidirectional biased NiO(60 nm)/NiFe(10 nm) bilayer is strongly dependent upon NiO deposition rate. It varied from 75 Oe for 3 Å/min to 103 Oe for 12 Å/min. However, the ripple structures we observed were identical.

In order to study a more realistic representation of the local coupling distribution, the MFM images of the isotropic coupled bilayers were obtained. Some representative

results are shown in figures 1(e)-(h). We note that as the NiO thickness was increased, the magnetization direction rotated from in-plane to out-of-plane. Even at 10 nm NiO, the mesh type ripples have all but disappeared. At 30 nm NiO thickness, high contrast and large domains appear in stark contrast with the unidirectional films. Upon closer inspection, we found that the magnetization patterns contained unusual domains, which we refer to as “*cross-type domains*”. These are most evident in the 60 nm NiO bilayer as shown in the marked circles of figure 1(h). A high-resolution image of the cross-type domain is shown in figure 2 where we rescanned a section of figure 1(h) amplified 4x and 10x. The bright areas and dark areas that comprise the horizontal and vertical legs of the cross are from the magnetic poles of the out-of-plane magnetization. The domains became smaller but had stronger contrast as NiFe thickness was decreased. We investigated the difference in cross domain structure as a function of deposition rate and hence, H_{ex} , and found that the density of these cross-type is proportional to the value of the H_{ex} . Thus, we believe that these domains are responsible for H_{ex} .

To explain the origin and structure of the cross-type domain a schematic of the interfacial cross section of the unidirectional and isotropic coupled FM layer over an

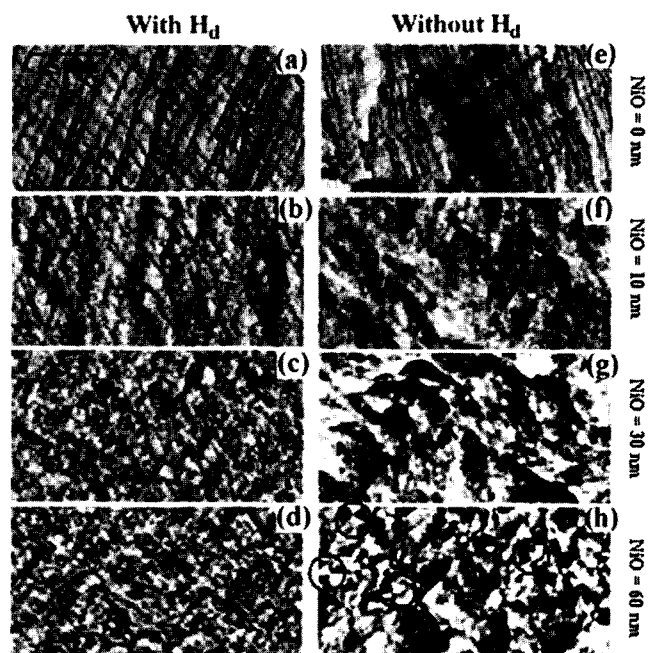


Fig. 1. The dependences of microscopic MFM images on NiO thickness in the NiO(t nm)/NiFe(10 nm) bilayer deposited with and without H_d , where t is (a, e) 0 nm, (b, f) 10 nm, (c, g) 30 nm, and (d, h) 60 nm. The (a)-(d) images were measured from the bilayers with H_d and the (e)-(h) ones without H_d . The marked circles show the cross-type domains.

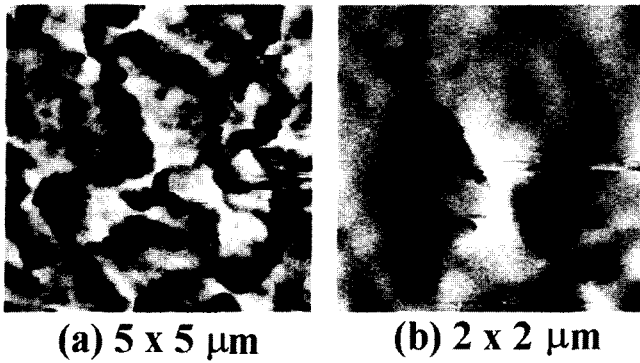


Fig. 2. The (a) $5 \times 5 \mu\text{m}$ and (b) $2 \times 2 \mu\text{m}$ scanned high-resolution image of cross-type domain in the NiO(60 nm)/NiFe(10 nm) bilayer without H_d .

(111) oriented and tilted AF grain are suggested in figure 3. The schematic is based on high-resolution TEM micrographs of NiO/Co interfaces measured by others [3]. The NiFe film deposited without H_d over randomly oriented NiO grains 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 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 1(a)~(d). The complex ripple pattern observed for the exchanged biased films arise from the local variation 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 10^1 nm, the strong ferromagnetic coupling between NiFe causes the formation of domains with out-of-plane magnetization. These domains are most likely 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 3(c). These cross-type domains are quite different from other systems that have out-of-plane magnetizations such those seen in multilayer films used in perpendicular

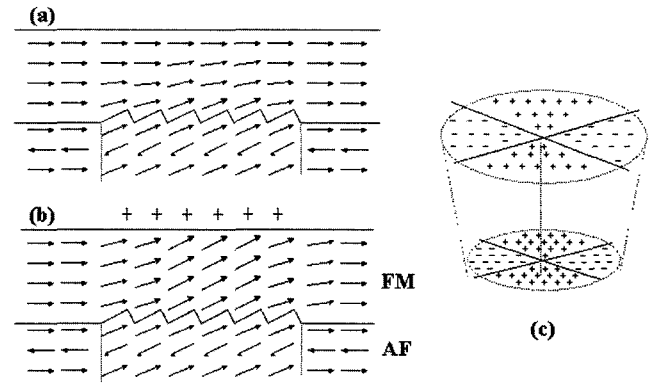


Fig. 3. A schematic views of the interfacial cross sections of an (a) unidirectional and (b) isotropic exchange-coupled FM layer over a (111)-oriented and tilted AF grain. Each arrow represents average magnetization vector of several spins. (c) A schematic view of a typical cross-type domain with out-of-plane magnetic poles

recording. In those systems, one invariably finds labyrinth or serpentine patterns that are expected to form 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 observed grain sizes of polycrystalline NiO film were reported to be a few ten nanometers [3, 5] but the sizes of these domains were about $1\text{--}2 \mu\text{m}$ 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 settle this apparent inconsistency, our model can be supplemented from Takano's model for the uncompensated interfacial AF spins [9]. The cross domain can be regarded as caused by the uncompensated poles of several NiO grains rather than one tilted grain. Also, even if the local out-of-plane pole was from each tilted (111) grain, the macroscopically observed pole density will be reconstructed by a strong exchange coupling between FM spins. Conclusively, one can argue that the preponderance of the cross-type domains reflect a high density of uncompensated moments at FM/AF interface; and these uncompensated moments generate a strong exchange biasing.

Figure 4 shows a possible process on the creation and field induced variations of cross-type domain in NiO(60 nm, $3 \text{ \AA}/\text{min}$)/NiFe(10 nm) bilayer during a magnetization cycle. As the external field is released from positive saturation, a strong contrast out-of-plane pole was created at zero field as shown in figure 4(c). Note that the scanned image (c) is different from (d) despite being obtained in a same region and field. Prior to the creation of the cross-type domain at (d), a single out-of-plane

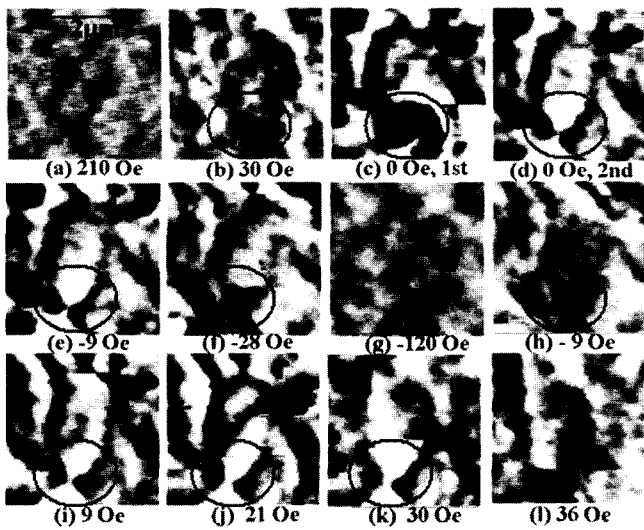


Fig. 4. MFM images of cross type domain changing as applied field decreases (a) to (g), and increases (g) to (k). The cross domain is at the marked circles, and the scratched lines in the images of (c), (f), and (k) were influenced by MFM tip.

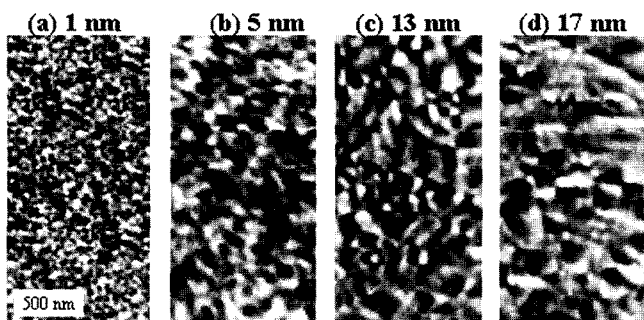


Fig. 5. MFM images of exchange-coupled NiO/Fe films as Fe thickness increase (a) 1 nm to (d) 17 nm.

domain was concentrated at the same region. The field from the MFM tip provided that additional perturbation to induce the transformation into a cross-type domain which reduces the pole energy. 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 same region and with nearly identical structure. It is strong evidence that the domain was induced from the strongest exchange coupling of FM/AM interface.

Figure 5 shows the MFM images of thin Fe films grown on polycrystalline NiO using in-situ UHV MFM/

STM measurements. As the Fe thickness increases 1 nm to 17 nm, the magnetic domain structure changed from the fine complex structure of 1 nm Fe film to the cross type domain structure above 10 nm Fe.

4. Conclusion

We observed a new cross-type domain with strong out-of poles in isotropic exchange-coupled NiO/NiFe film. From the data on the dependence on thickness and deposition rate, the existence of the domain has been directly linked with the local exchange biasing field. The fact that the domain originated from the strongest exchange-coupling region was confirmed by dynamic magnetic domain configurations during a magnetization cycle and the MFM images of different Fe thickness.

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