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Lorentz transmission electron microscopy and magnetic force microscopy characterization of NiFe/Al-oxide/Co films

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Magnetization reversal process of NiFe/Al-oxide/Co junction films was observed directly using Lorentz transmission electron microscopy (LTEM) and magnetic force microscopy (MFM). *In situ* magnetizing experiments performed in both LTEM and MFM were facilitated by a pair of electromagnets, which were mounted on the sample stages. A two-stage magnetization reversal process for the junction film was clearly observed in LTEM with NiFe magnetization reversed first via domain wall motion followed by Co magnetization reversal via moment rotation and domain wall motion. Reversal mechanism and domain characteristics of the NiFe and Co layers showed very distinctive features. The magnetization curve of the junction film measured using alternating gradient force magnetometry showed a nonzero slope at the antiparallel magnetization configuration region, which implies that magnetization directions of the NiFe and Co layers were not exactly antiparallel due to Co moment rotation existed in that region. After the magnetization reversal of the Co was complete, MFM images revealed some magnetic contrast, which suggests that an out-of-plane magnetization component remained in the Co layer. Such magnetic contrast disappeared at higher magnetic fields when the Co moments further rotated and aligned parallel to the applied field direction. [DOI: 10.1063/1.1427142]

I. INTRODUCTION

Magnetic tunnel junction (MTJ) has attracted much attention for both fundamental and applied physics research,[1,2] as it possesses promising application potential in nonvolatile magnetic random access memory and magnetoresistive read head technologies. An MTJ basically consists of two ferromagnetic layers separated by an insulator. Tunneling resistance between the ferromagnets depends strongly on the relative orientation of the magnetizations of the ferromagnets because of the asymmetry in the density of states of the majority and minority energy bands in a ferromagnet.[3] Generally speaking, in parallel magnetization configuration, the tunneling resistance is minimum, while in antiparallel magnetization configuration, the tunneling resistance is maximum.[4] It is therefore important for an MTJ to possess a clear two-stage magnetization reversal process for application purposes. The aim of this work is to directly observe and hence to obtain a better understanding of the magnetization reversal process of NiFe/Al-oxide/Co junction films using Lorentz transmission electron microscopy (LTEM) and magnetic force microscopy (MFM). The successful application of LTEM for the characterization of magnetoresistive multilayer systems has been reported.[5,6] MFM-based techniques have been exploited to characterize properties and performance of magnetoresisitive devices such as the effect of shield on magnetoresistive read-head performance[7] and the magnetoresistive response of patterned giant magnetoresistance sensors with different edge stabilization schemes.[8] The coercivity of NiFe is lower than that of Co, hence it is expected that a two-stage magnetization reversal process can be observed. The magnetic moments in the junction film are believed to be mainly oriented in plane because of the thin-film geometry, therefore, it is very useful to use the LTEM technique to observe the magnetic domains and the reversal process of the junction films, as LTEM is sensitive to in-

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plane magnetization of magnetic specimens. As a supplementary technique to further characterize the reversal process of the junction films, MFM, which is sensitive to stray field from magnetic specimens, was employed to examine the activity of the out-of-plane magnetization components in the junction film during the reversal process.

II. EXPERIMENTAL DETAILS

The NiFe/Al-oxide/Co (17/5/21 nm) films were fabricated using magnetron sputtering. The Al-oxide layer was deposited by direct sputtering from a pure alumina target. Microstructure of the film was characterized using high-resolution electron microscopy (HREM). *In situ* magnetizing LTEM experiment was performed in a JEOL 4000EX transmission electron microscope fitted with a low-field objective pole piece.[9] A pair of electromagnets, which can produce in-plane fields up to 400 Oe *in situ*, were mounted on two sides of the sample stage. LTEM was performed in the Fresnel imaging mode (i.e., the imaging lens is simply defocused so that the object plane is no longer coincident with the specimen), thus domain walls appeared as narrow dark and bright bands.[10] Furthermore, magnetization ripple, which is useful for indicating the magnetization direction of domain, was also observed. The defocusing value of the imaging lens was kept constant throughout the *in situ* magnetizing experiment, therefore, the change of magnetic contrast in the Fresnel images observed was not due to the change of defocusing value. In order to obtain an overview of the two-stage magnetization reversal process of the junction films and to confirm that the magnetization in the NiFe layer reversed first followed by the magnetization reversal of the Co layer, magnetization curves were measured for the junction films using alternating gradient force magnetometry (AGFM) with magnetic field applied in plane.

MFM study was made on the junction films using silicon pyramidal tips coated with CrCo thin films. All MFM images were taken in phase imaging mode. The image contrast corresponds to variations in the phase shift of the cantilever oscillation that are caused by the magnetic force gradients above the sample surface. To study the magnetization reversal process in the sample, a pair of electromagnets capable of producing in-plane fields *in situ* up to about 600 Oe was mounted on the sample stage. During the *in situ* magnetizing MFM experiment, images were taken under various fields up to about 400 Oe applied along one direction. Repeatable MFM images were obtained by rescanning the same area, indicating that the domain structure of the sample was not affected by the stray field of the tip. The magnetic images obtained in the remanent states before and after the experiment were found to have comparable image contrast. This ascertained that the magnetic moment of the tip was not altered by the applied field.

III. RESULTS AND DISCUSSION

Figure 1 shows a typical HREM cross sectional image of the NiFe/Al-oxide/Co junction film. The NiFe and Co layers were clearly separated by the Al-oxide layer which appeared amorphous homogeneously. Both NiFe and Co layers showed crystallographic texture, however the grains were randomly oriented. Furthermore, the interfaces between the ferromagnetic layers and the Al-oxide layer were not perfectly flat, which was due to the surface roughness of the bottom NiFe layer, thus the surface of the Al-oxide layer and, therefore, the top interface also exhibited some roughness.

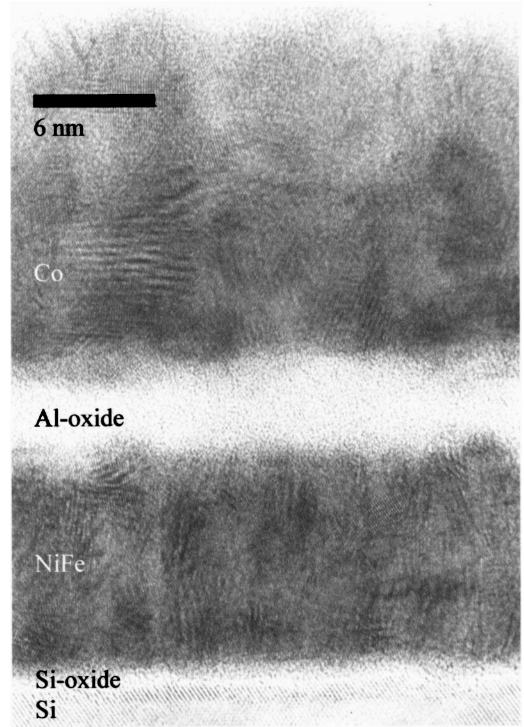

FIG. 1. Typical HREM cross sectional image of the NiFe/Al-oxide/Co junction film.

The magnetization reversal process of the junction film observed using LTEM is shown in Fig. 2. Magnetization ripple, which is due to anisotropy dispersion, can be seen in the junction film. When there is a variation, from place to place in the film, of the direction of the easy axis and/or the magnitude of the anisotropy constant, because of inhomogeneities in the structure of the film (i.e., anisotropy dispersion), the direction of the local magnetization varies slightly from one point to another even within a domain. The nonparallelism of the local magnetization increases the exchange energy of the system, while free poles are created within the domain because of the finite divergence of magnetization causing stray fields and magnetostatic energy. Magnetization ripple thus forms for use to minimize the exchange and magnetostatic energy. Magnetization ripple is normal everywhere to the local magnetization direction. A field of −400 Oe was applied to the junction film initially in order to saturate the NiFe and Co layers. The field was then decreased [Fig. 2(a)] and when it was reduced to zero, magnetization ripple with slightly higher contrast was visible in the junction film [Fig. 2(b)]. Some domains [e.g., marked D in Figs. 2(b) and 2(c)] began to nucleate around some defects, which could be some dust particles or microscratches residing on the substrate surface in the film. When the field was increased in a reverse direction, the domains marked D grew and domain walls were clearly observed [Fig. 2(c)]. As the field increased, the

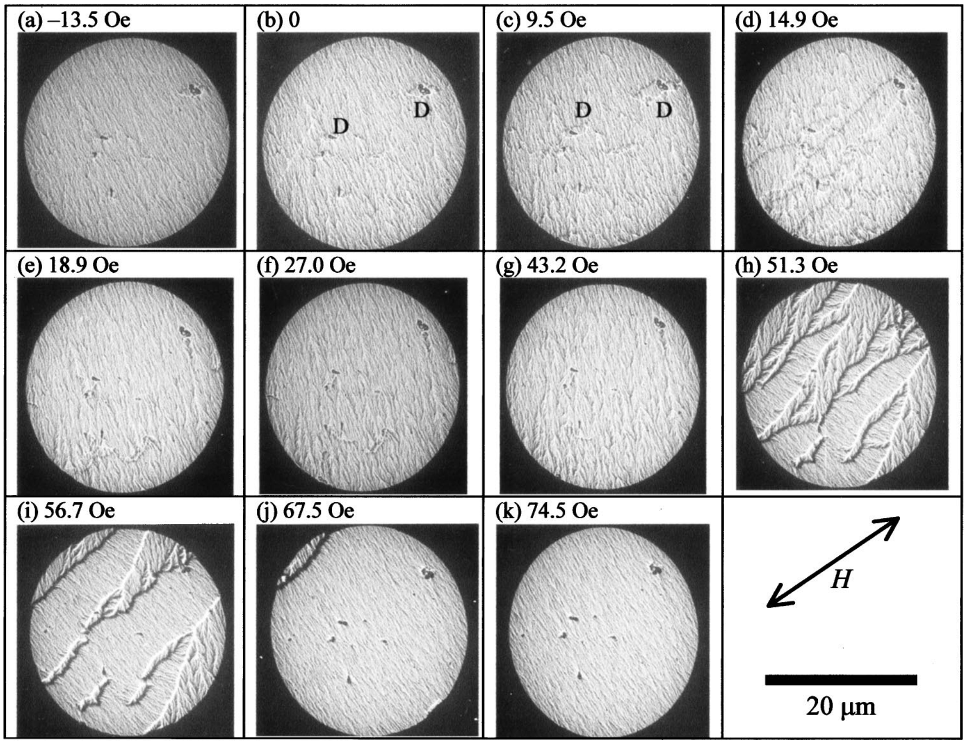

FIG. 2. LTEM Fresnel images of the magnetization process for NiFe/Al-oxide/Co junction film. The direction of the applied field, *H*, is indicated. All images are of the same area.

domains grew quickly via domain wall motion [Fig. 2(d)]. The domain walls visible in Figs. 2(c) and 2(d) are expected to be in the NiFe layer because NiFe has a lower coercivity than Co which suggests that the magnetization reversal process of the NiFe layer should occur in a lower field than that of the Co layer. The domain walls in the NiFe layer mostly disappeared when the field applied was 18.9 Oe [Fig. 2(e)], indicating that the magnetization reversal of the NiFe layer was complete. The LTEM image remained almost the same when the field increased to 27.0 Oe [Fig. 2(f)]. It is noticed that the magnetization ripple did not rotate much from Figs. 2(a)–2(d), which implies that there was no significant moment rotation in the junction film before the magnetization reversal of the NiFe layer was complete, and that the magnetization reversal of the NiFe layer occurred mainly via domain wall motion.

It was observed that very slight ripple rotation began to occur at 18.9 Oe [Fig. 2(e)], and the rotation process continued as the applied field increased to 43.2 Oe [Fig. 2(g)]. Such ripple rotation is expected to be due to the moment rotation in the Co layer. When the applied field increased to 43.2 Oe, the ripple contrast increased and higher angular distribution of the ripple was observed, but no domain wall was visible [Fig. 2(g)]. The magnetization directions of the NiFe and Co layers were almost antiparallel to each other between 18.9 Oe [Fig. 2(e)] and 51.3 Oe [Fig. 2(h)]. However, it is believed that the magnetization of the Co layer was rotating toward the reverse field direction throughout the ''antiparallel'' magnetization configuration region resulting from the ripple rotation observed between Figs. 2(e) and 2(g). As the field increased further, domain walls (which are expected to be domain walls in the Co layer) appeared at 51.3 Oe [Fig. 2(h)]. Comparing Figs. 2(g) and 2(h), one can observe that there is significant ripple rotation, which implies that the domains were mainly nucleated by the process of moment rotation. Domains with magnetization parallel to the reverse field direction (magnetization ripple in these domains show very low angular distribution) grew via domain wall motion as the field increased [Fig. 2(i)]. Almost all of the domain walls in the Co layer disappeared at 67.5 Oe [Fig. 2(j)] which indicated that the magnetization of the Co layer had reversed generally to the reverse field direction. After magnetization reversal of the Co layer, the magnetization directions of the NiFe and Co layers were parallel and aligned in the reverse field direction, and only weak magnetization ripple was observed [Fig. 2(k)]. Magnetization ripple still existed at field values higher than 75 Oe and the ripple contrast faded as the field value increased (note: there is an instrumentation limitation on observing LTEM images at field values higher than 120 Oe). The existence of ripple contrast in relatively high fields may confirm the presence of the out-of-plane magnetization component as observed in the MFM experiment described next.

In the Fresnel mode LTEM images, the domain walls in the NiFe layer appeared narrower than those in the Co layer. It is because the Co layer was thicker and its saturation magnetization was higher than the NiFe layer, thus the electrons were deflected more when passing through the Co layer, therefore the domain wall images in the Co layer appeared wider than those in the NiFe layer.[11] When the Fresnel mode LTEM images were studied, it was difficult to conclude whether the ripple contrast was contributed by the NiFe layer, by the Co layer, or by both layers because plan-view images were observed, so that a projection of the NiFe and Co layers were superimposed in a single image. When the junction film was in low fields, the ripple contrast was due to both NiFe and Co layers, therefore, it was very difficult to distinguish the ripple contrast provided by the two layers. At

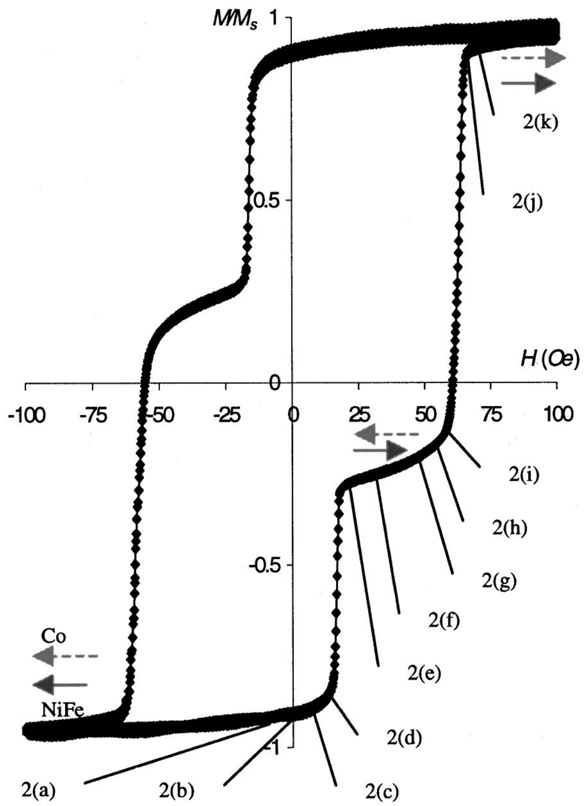

FIG. 3. Normalized magnetization vs applied field for the NiFe/Al-oxide/Co junction film. The corresponding domain structure at different field values along the hysteresis loop is shown in Fig. 2.

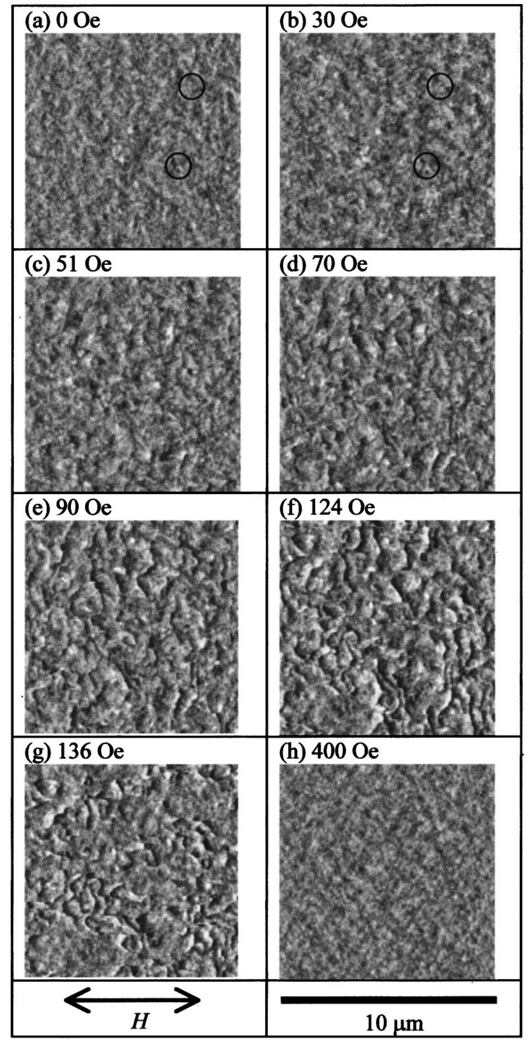

FIG. 4. MFM images of the magnetization process for NiFe/Al-oxide/Co junction film. All images are of the same area. Circled regions in (a) and (b) are examples of local switching of image contrast observed in this field range.

fields above the magnetization reversal of the NiFe layer occurred, the ripple contrast was expected to be mainly due to the Co layer because the magnetic moments in the NiFe layer were almost saturated and aligned in the reverse field direction while the magnetic moments in the Co layer were still rotating. When the junction film was in high fields, the ripple contrast was low because the magnetic moments in both NiFe and Co layers were saturated and aligned parallel to the applied field direction.

Figure 3 shows a normalized magnetization curve for the junction film. The two-stage magnetization reversal characteristic of the junction film is clearly revealed in the magnetization curve. The field values at which the corresponding domain structure images were recorded during the LTEM *in situ* magnetizing experiment are indicated [e.g., 2(a) in Fig. 3 corresponds to image (a) in Fig. 2]. The normalized magnetization of the junction film is not zero when the NiFe and Co layers were in the antiparallel magnetization configuration. It is because the saturation magnetization of NiFe is smaller than that of Co, besides that the NiFe layer was thinner than the Co layer. Thus, it can be confirmed that the NiFe layer reversed first followed by the reversal of the Co layer. The magnetization reversal of the NiFe layer occurred between Figs. 2(c) and 2(e). It is expected that the magnetization of the NiFe and Co layers were almost antiparallel to each other between Figs. 2(e) and 2(h). However, the nonzero slope between Figs. 2(e) and 2(h) indicates that the magnetization of the NiFe and Co layers were not exactly antiparallel to each other over that field range. The existence of such a nonzero slope agrees with the LTEM observations of ripple rotation over that field range [e.g., compare Figs. 2(e) and 2(g)], which implies that moment rotation began to occur in the Co layer after the magnetization reversal of the NiFe layer was complete. The magnetization of the Co layer mainly reversed between Figs. 2(h) and 2(k). After Fig. 2(k), the magnetization directions of the NiFe and Co layers were parallel and aligned in the reverse field direction. The very small slope of the magnetization curve at the fields higher than 75 Oe could be induced by the out-of-plane magnetization components observed between 90 and 136 Oe in the MFM experiment.

Figure 4 shows the MFM images obtained at various stages of the hysteresis cycle. The sample was first magnetized to saturation by applying a field of −400 Oe. The field was then decreased to zero and a fine domain structure was observed [Fig. 4(a)]. The observed image contrast arises either from divergence of magnetization at the domain walls, or from the variations in the out-of-plane magnetization components of the top Co layer. When increasing the reverse field to 30 Oe, local switching of image contrast occurred [examples are highlighted by the circled regions in Figs. 4(a) and 4(b)]. The *in situ* magnetizing LTEM study revealed that in this field range the magnetization reversal involved mainly domain wall motion in the NiFe layer. This could induce the observed local changes in the magnetization component of the Co layer, because the NiFe and Co layers are ferromagnetically coupled due to ''orange-peel'' coupling effect, which is caused by the interface roughness.[12] As the field was increased from 50 Oe to 70 Oe [Figs. 4(c) and 4(d)], a zig-zag pattern running normal to the field direction appeared. In this field range, moment rotation and domain wall motion in the Co layer were observed in the LTEM study. The image contrast increased with applied field up to about 124 Oe [Figs. 4(d)–4(f)]. When increasing the field to about 136 Oe, local switching of the image contrast occurred again, resulting in a disruption of the zig-zag domain pattern [Fig. 4(g)]. The magnetic contrast appeared between Figs. 4(e) and 4(g) which suggests that the Co moments were not aligning exactly in plane, however with out-of-plane components, after the magnetization reversal of Co was complete. As LTEM is not sensitive to magnetic field normal to plane, so no significant magnetic contrast was observed in LTEM in that field range. On the contrary, MFM images indicate that before the Co layer was fully saturated, the Co moments alignment varied normal to plane, but not completely in plane. Further increase in field caused the image contrast to decrease as the in-plane magnetization component of the Co layer along the field direction increased toward saturation [Fig. 4(h)]. On reducing the field from a saturation value to zero, only little local switching of image contrast was observed.

IV. CONCLUSIONS

The magnetization reversal process of NiFe/Al-oxide/Co junction films was studied by performing *in situ* LTEM and MFM experiments. Magnetization of the NiFe layer first reversed via wall motion followed by the Co magnetization reversal via initial moment rotation and then wall motion in the two-stage magnetization reversal process of the junction film. The magnetization curve measured using AGFM showed a nonzero slope at the antiparallel magnetization configuration region indicating that the magnetization directions of the NiFe and the Co layers were not exactly antiparallel in that region. Co moment rotation began to occur immediately after the reversal of the NiFe was complete; it was consistent with the ripple rotation observed in LTEM. The MFM results revealed the presence of an out-of- plane magnetization component in the Co layer after the magnetization reversal of the Co layer was generally complete. When the applied field was increased to higher values, the out-of-plane magnetization component in the Co layer diminished as the Co moment rotated further in order to align parallel to the field direction.

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