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Asymmetry of domain nucleation and enhanced coercivity in exchange-biased epitaxial NiO/NiFe bilayers

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Magnetization reversal processes in epitaxial NiO/NiFe bilayers were studied using the magneto-optic indicator film technique. The influence of dislocations on these processes was determined. Remagnetization parallel to the unidirectional anisotropy axis proceeds by domain nucleation and growth, with nucleation center activity being asymmetric with respect to the applied field sign. Magnetization reversal in the hard axis direction occurs by incoherent rotation. The enhanced coercivity and asymmetric nucleation can be explained by taking into account domain wall behavior in the *antiferromagnetic* layer. [S0163-1829(98)50514-0]

The unusual phenomena of unidirectional anisotropy was revealed over forty years ago in ferromagnetic (FM) Co fine particles with an oxidized antiferromagnetic (AF) shell.[1] This results in a shift of the hysteresis loop of the ferromagnet away from the zero-field axis. The same effect has since been observed in layered structures.[2–5] This effect was one of several fascinating phenomena[6–8] caused by exchange coupling between neighboring layers with different magnetic order which have opened possibilities for exciting new practical applications.[9,10]

In the first model the authors[1] proposed that the interfacial exchange coupling was comparable to the atomic exchange coupling in bulk FM or AF materials. That model, however, fails to describe the small magnitude of the measured exchange anisotropy field, H_E. Therefore, new models have been proposed which explain the measured H_E value after taking into account domain wall formation in the AF layer and the presence of a random exchange field due to monatomic steps at the AF/FM interface.[11,12]

Other drastic discrepancies between theory and experimental data have also been reported.[2,13–15] For instance, it is well known that the coercivity, H_C, of a FM film in contact with an AF layer is enhanced compared to the ''free'' FM layer. However, there is no model which describes this enhancement. This phenomenon cannot be understood in terms of a spin coherent rotation model.[1] In this latter case the interfacial exchange coupling leads only to a shift in the hysteresis loop. The coercivity of the AF/FM bilayer remains equal to the coercivity of the free FM layer: $H_C^F = 2K_F/M_S$, where K_F and M_S are the uniaxial anisotropy constant and the saturation magnetization of the ferromagnet, respectively. A model,[11] incorporating a one-dimensional planar domain wall in the AF layer, does predict the same value of H_C for both small and large interfacial exchange couplings, and even lowered H_C for intermediate coupling magnitude.

It can be thought that in an AF/FM bilayer the magnetization process in a FM film proceeds most likely by either nonuniform spin rotation or by domain wall nucleation and motion. These magnetization processes should be accompanied by inhomogeneities in the AF spin distribution both across and along the interface. We will show that the increased coercive force in the AF/FM bilayer may be explained by taking into account the nucleation of domains in the AF layer with walls having components which are both parallel and perpendicular to the AF/FM interface.

In order to study the magnetization process in epitaxial NiO/NiFe bilayers grown on single-crystal MgO (001) substrates we used the magneto-optical indicator film (MOIF) technique.[16] A transparent Bi-substituted iron garnet indicator film with in-plane anisotropy is placed on top of the sample. Polarized light is passed through the indicator film and reflected by an Al underlayer. The normal component of the magnetic stray field of the sample is detected by brightness contrast in a polarizing microscope due to the magneto-optical Faraday effect. Macroscopic hysteresis loops of the bilayers were measured with a vibrating sample magnetometer. The defect structure of the bilayers was determined by surface steps observed in an optical reflecting microscope, and internal stresses were revealed by birefringence in a polarizing microscope.[17] Bilayers of NiO(500 Å)/NiFe(100 Å) were grown by ion beam sputtering onto a (001) MgO single crystal.[18] Permalloy films grown on (001) MgO without the NiO buffers were also prepared together with the NiO/NiFe bilayers. Both uniaxial (in NiFe) and unidirectional (in NiO/NiFe) anisotropy was created during deposition in the FM layers by means of a 300 Oe uniform permanent magnet field in the plane of the substrate.

Figure 1(a) is an example of the domain structure observed during the magnetization reversal of the NiO/NiFe bilayer along the direction indicated by the arrow. In this picture, the domain configuration is entirely associated with the defect structure of the sample, revealed in reflected light [Fig. 1(b)] and in transmitted polarized light [Fig. 1(c)]. In Fig. 1(b) NiFe surface steps parallel to the [100] direction, which are associated with slip planes of screw dislocations, are revealed. Figure 1(c) shows the birefringence picture due

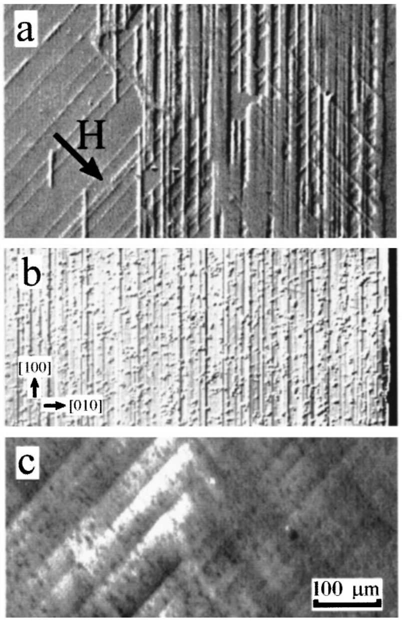

FIG. 1. (a) MOIF image of the domain structure in a NiO/NiFe bilayer (arrow indicates the direction of an applied field, $H = 6$ Oe). (b) Surface steps associated with screw dislocation slip planes revealed in reflected light at the NiFe surface. (c) Microstress fields caused by the slip planes of edge dislocations revealed by an optical birefringence. All three images are of the same region and orientation.

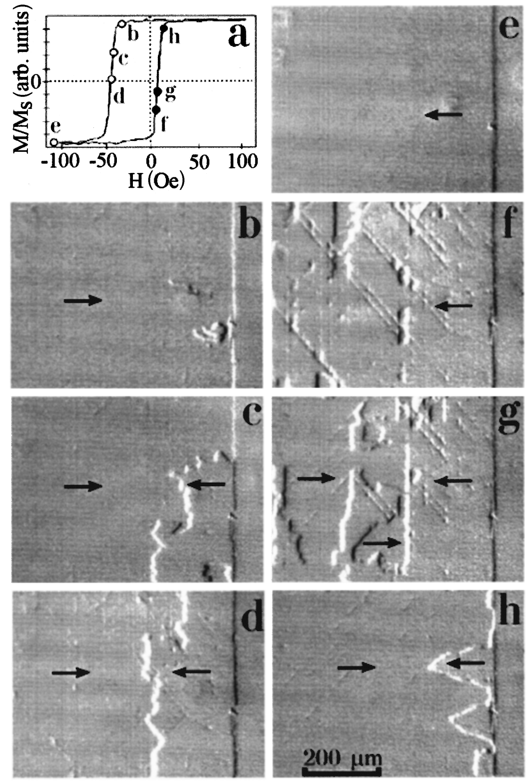

FIG. 2. Magnetic hysteresis loop (a) and MOIF images of domain structure taken during the unidirectional-axis magnetization reversal of a NiO/NiFe bilayer. (b)–(h) correspond to the conditions indicated by the circles labeled by the same letters on the hysteresis loop in (a). The vertical right-hand band perpendicular to the unidirectional axis is the edge of the bilayer, revealed due to magnetic stray fields. Arrows indicate magnetization directions in domains.

to microstresses caused by edge dislocations which are aligned along (110) and (110) slip planes. Both edge and screw dislocations were introduced into the MgO substrate during cleaving before the NiO/NiFe bilayer deposition. The screw dislocation steps shown in Fig. 1(b) indicate that these MgO dislocations propagate through the NiO and permalloy films during their epitaxial growth. It is important to note that the edge dislocations play the role of domain nucleation centers despite the fact that they do not introduce steps on the film interface.

The NiO/NiFe hysteresis loop measured along the [010] direction, which coincides with the direction of a magnetic field applied during the bilayer growth, is shown in Fig. 2(a). It exhibits an exchange shift ($H_E = 20$ Oe) and an enhanced coercive force ($H_C = 26$ Oe) compared to the coercivity of the MgO/NiFe system ($H_E^F = 0$, $H_C^F = 2$ Oe). The MOIF patterns in Figs. 2(b)–2(h) display the behavior of the domain structure of the NiO/NiFe bilayer during the magnetization reversal process. Letters on the hysteresis loop [Fig. 2(a)] refer to the conditions of the corresponding MOIF patterns. Three principal features of the bilayer magnetization reversal are worth noting. (1) The reversal occurs as a result of nucleation and subsequent growth of domains having a new magnetization orientation. Nuclei of domains with reversed magnetization form at a magnetic field close to the coercive field [Fig. 2(b)], and their growth encompasses the whole sample within a small field range of H_i, resulting in an almost square hysteresis loop. This implies that the magnetization reversal is limited by the nucleation process. (2) An asymmetry is observed in the activity of the domain nucleation centers. When the magnetic field is aligned against the unidirectional anisotropy axis, the nucleation of domains occurs at the film edges (or different chemical inhomogeneities) [Fig. 2(b)]. However, when the field is aligned along this axis, the domain nucleation takes place at dislocation slip planes and their intersections [Fig. 2(f)]. (3) The dislocations not only influence the domain nucleation but also impede the domain wall motion. As a result, the specific head-to-head domain walls consist of sections parallel to the dislocation slip plane.

It is important to note that the NiFe film grown on MgO without an AF NiO layer has a similar dislocation structure, but its domain structure behaves differently. In this latter film, there is no shift in its hysteresis loop, its coercive force is very small ($H_C^F = 2$ Oe), and the dislocation structure did not exhibit much influence on the domain wall behavior. In the case of the MgO/NiFe film, the formation of domains with reversed magnetization was observed at film edges for both applied field directions parallel to the easy axis, and there was no asymmetry in the activity of its nucleation centers. These observations imply that the dislocations in the NiO/NiFe bilayer influence primarily the spin configuration statics and dynamics in the AF layer which is exchange coupled with the spins in the FM layer.

The hysteresis loop and MOIF images taken during mag-

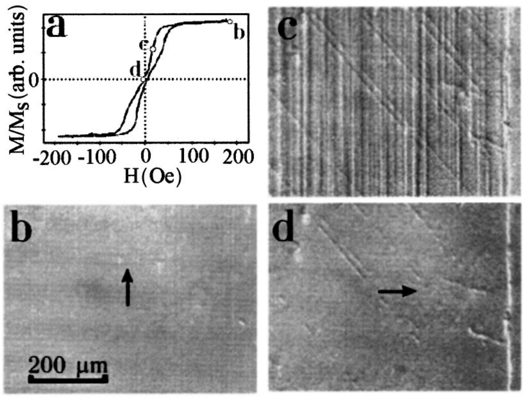

FIG. 3. Hysteresis loop (a) and MOIF images of magnetic structure [(b)–(d)] observed during hard axis magnetization reversal of a NiO/NiFe bilayer.

netization reversal along a direction perpendicular to the unidirectional anisotropy axis in the NiO/NiFe bilayer are shown in Fig. 3. In this case, a much different reversal mechanism was found. The hard axis magnetization curve was nearly linear and saturated near $2H_E$. The hysteresis loop displayed almost zero coercivity and no field shift. The MOIF images show that the reversal proceeds by incoherent magnetization rotation. Figure 3(b) shows the MOIF image for the bilayer when in the saturated state. Note, there is no vertical component to the magnetostatic field in this case at the sample edge (which is parallel to the right side of the picture frame, but indented $\sim 20\ \mu$m). When the magnetic field was reduced, the nonhomogeneous MOIF image [Fig. 3(c)] corresponding to the dislocation structure (Fig. 1) appeared. Since the directions of the magnetization vectors in Fig. 3(c) are not parallel to the unidirectional anisotropy axis, it is obvious that the dislocation microstresses have changed the effective magnetic anisotropy. However, at zero applied field the magnetization becomes nearly uniform and is aligned along the unidirectional anisotropy axis. This alignment is revealed by the presence of the stray field induced vertical white stripe at the right-hand edge of the film [Fig. 3(d)].

To explain the enhanced coercivity we propose the model that is a generalization of the model given in Ref. 11. Direct experimental observations of the magnetization reversal processes showed that the film remagnetization in the easy direction proceeds by domain-wall nucleation and motion. Therefore, spin variations along the AF/FM interface need to be considered. We include these spin variations in Eq. (1) below (describing the energy density of the bilayer averaged over its thickness), which is simply an extension of Eq. (1) of Ref. 11:

$$\sigma = A_F d\left(\frac{d\phi}{dx}\right)^2 + K_F d\sin^2\phi - H_x M_S d\cos\phi - H_x^m(x) M_S d\cos\phi + A_A\Delta_A\left(\frac{d\psi}{dx}\right)^2 + K_A\Delta_A\sin^2\psi + 2\sqrt{A_A K_A}(1-\cos\psi) - J\cos(\phi-\psi), \tag{1}$$

where A_F, K_F, A_A, and K_A are the exchange and anisotropy constants of the FM and AF layers, respectively, d is the FM thickness, $\Delta_A = \sqrt{A_A/K_A}$ is related to the domain wall thickness in the AF layer, H_x is the external magnetic field, J is the AF/FM interfacial exchange constant, and $H_x^m(x) = -2M_S d\cos\phi(0)/x$ is the magnetostatic field at the FM edge. ϕ and ψ are, respectively, the directions of the FM layer magnetization and AF layer spin vectors with respect to the x axis (defined perpendicular to the bilayer edge).

Near the interface, if $J/2\sqrt{A_A K_A} \gg 1$ one can assume that ϕ and ψ change coherently. The coercivity is determined from the stability criteria for nonuniform magnetization reversal modes near the film edge. For the free FM layer this yields[19]

$$H_C^F = \frac{2K_F}{M_s} - \frac{M_S^3 d^2}{8A_F}. \tag{2}$$

In a similar manner, for the AF/FM bilayer we have

$$H_C^- = -\frac{2K_F}{M_s} - \frac{2K_A}{M_S}\frac{2\Delta_A}{d} + \frac{M_S^3 d^3}{8(A_F d + A_A\Delta_A)} = -\frac{2K_F}{M_s} - \frac{\sigma_A}{M_S d} + \frac{M_S^3 d^3}{8(A_F d + A_A\Delta_A)}, \tag{3}$$

$$H_C^+ = \frac{2K_F}{M_s} - \frac{M_S^3 d^3}{8(A_F d + A_A\Delta_A)}, \tag{4}$$

where H_C^+ and H_C^- are the coercivities at the magnetic field orientations parallel and antiparallel to the unidirectional anisotropy axis, respectively, and $\sigma_A = 4\sqrt{A_A K_A}$. It follows that the bilayer exchange anisotropy field and coercivity are

$$H_E = \frac{H_C^+ + H_C^-}{2} = -\frac{2K_A}{M_S}\frac{\Delta_A}{d} = -\frac{\sigma_A}{2M_S d}, \tag{5}$$

$$H_C = \frac{H_C^+ - H_C^-}{2} = \frac{2K_F}{M_s} + \frac{2K_A}{M_S}\frac{\Delta_A}{d} - \frac{M_S^3 d^3}{8(A_F d + A_A\Delta_A)}. \tag{6}$$

So, the exchange shift of the hysteresis loop H_E given by Eq. (6) is the same as obtained in Ref. 11. In addition, according to our model, the enhanced coercivity H_C appears as a fundamental property of an AF/FM sandwich associated with spin variations along the interface. The main point of our treatment of the bilayer reversal is that we decomposed a two-dimensional spin distribution of the AF into two one-dimensional distributions: one parallel and one perpendicular to the interface. The difference between coercivities during remagnetization in opposite directions of the bilayer [see Eqs. (3) and (4)] occurs because in one case it is necessary to overcome the energy of the spin distribution in the AF both along and across the interface. However, when a field of opposite polarity is applied there is no necessity to overcome the energy of the spin distribution perpendicular to the AF/FM interface. Moreover, the energy stored in a planar domain wall compensates for the energy cost of nucleating the spin inhomogeneity along the interface. Therefore, the term which is proportional to σ_A disappears from the expression for H_C^+ [compare Eqs. (3) and (4)]. In both cases we assume the nucleation process at the edge of the sample is due to the magnetostatic fields.

Clearly, the asymmetry in the activity of domain nucleation centers observed cannot be explained by the above model. To understand this asymmetry it is necessary to take into account local variations of the antiferromagnetic anisotropy. The physical origin for these variations is most likely crystal lattice defects. In our bilayers, due to the low magnetostriction of permalloy we neglect the anisotropy variations due to stresses around crystal defects (like dislocations) in the ferromagnet. Regions with enhanced anisotropy in the antiferromagnet play no role when the antiferromagnet does not have a Mauri-like[11] planar domain wall, but they play a crucial role when there is such a domain wall consisting of twisted spins in the NiO. The energy associated with spin twisting is of the order of $\sqrt{A_A K_A}$. Therefore, spins at places with enhanced K_A tend to untwist at a lower magnetic field similar to the untwisting of a torsional spring when the externally applied torque is relaxed. This easier spin rotation in the antiferromagnet results then in an easier local magnetization reversal in the ferromagnet. Similar asymmetry in domain nucleation has been observed earlier in the CoO/Co system.[2] We suggest that our model can explain those observations as well, thereby suggesting this phenomenon may be generic for all AF/FM bilayers.

The remagnetization experiment of the bilayer in the hard direction supports the correctness of our assumption that the anisotropy distribution in the AF layer is inhomogeneous. As one can see from Fig. 3(c), when the field is decreased, the inhomogeneous rotation of M_S is determined by the dislocations. More detailed analysis shows that the spins rotate at lower fields near these dislocations. Only when the field is switched to zero will all the spins orient along the unidirectional anisotropy axis [Fig. 3(d)].

In summary, we have studied experimentally the magnetization reversal of epitaxial NiO/NiFe bilayers. We have extended the model of exchange biasing[11] to describe the measured enhanced coercivity and we observed in these bilayers an asymmetry in the activity of various domain nucleation centers.

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