Microscopic Imaging of Fe Magnetic Domains Exchange Coupled with Those in a NiO(001) Surface

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(Received 18 October 1999)

We revealed the micromagnetic structure of an Fe thin film exchange interacting with the spins of a fully compensated (001) surface of antiferromagnetic NiO. The interface exchange interaction causes the Fe domains to follow the NiO domains. The Fe spin polarization is in plane and the spin polarization in each domain is roughly perpendicular to an easy-spin axis of the NiO. These results agree with numerically calculated spin directions. Our numerical results also show that the NiO spins at the interface cant from the easy-spin axis towards the Fe spin because of exchange coupling.

PACS numbers: 75.70.Ak, 75.30.Gw, 75.60.Ch, 75.70.Cn

Multilayer systems formed by thin films of ferromagnetic (F), antiferromagnetic (AF), and nonmagnetic materials show new magnetic properties which cannot be obtained by bulk materials. These systems have attracted much attention in the field of basic research as well as technological applications. Giant magnetoresistance (GMR) [1] of magnetic superlattices and exchange anisotropy [2] of F and AF bilayered films are two of the most remarkable phenomena in these systems, and both have been successfully used in high-performance magnetic-sensor devices of magnetic storage systems. In both systems, the magnetic interaction between the films plays an important role in their magnetic properties, and the mechanism of the magnetic interaction must be clarified in order to understand the origin of their phenomena and improve the device performances. Extensive experimental and theoretical studies have revealed the interaction mechanism (interlayer coupling) in the GMR films. On the other hand, although four decades have passed since the first discovery of exchange anisotropy [2], the magnetic interaction in F/AF bilayers is still unclear.

It has been generally accepted that the net moment appearing on an uncompensated AF surface that is ferromagnetically or antiferromagnetically coupled with the F moment is responsible for the exchange anisotropy [2,3]. This coupling, however, means that the net exchange coupling energy between the F film and the compensated AF surface is independent of the relative angle between the F and AF spins; consequently, the AF film does not cause the exchange anisotropy of the F/AF bilayer. However, the results of recent experiments suggest that (i) the hysteresis loop shift of the Fe film on the compensated $FeF_2(110)$ surface can be reduced by increasing the surface roughness of the compensated plane [4] and that (ii) the F spin of the NiFe films on the compensated and uncompensated CoO planes seems to have a tendency to be directed perpendicular to the AF easy axis [5]. Moreover, Koon reported simulations indicating that the F spins exchange coupled with the spins of the compensated AF plane tend to be directed in the perpendicular direction to the AF easy axis and that the AF spins cant towards the F spin [6]. Recent experiments on Fe_3O_4/CoO superlattices [7] and on Fe/FeF_2 [8] have supported his simulations. However, in the former experiment the magnetic lattice parameter of ferrimagnetic Fe_3O_4 was about twice that of CoO, so the Fe_3O_4 spins are probably coupled to *ferromagnetically* aligned spins belonging to one of the CoO sublattices [9]. The CoO(001) plane is thus not a compensated surface for the Fe_3O_4. In the latter experiment, the authors pointed out that perpendicular coupling between the Fe and FeF_2 spins could occur, rather than exchange coupling, because of the magnetoelastic effects. Furthermore, since the AF surface roughness was not estimated in either of these experiments, the possibility that the net AF moment at the interface cannot be excluded.

In the present study, we perform a microscopic investigation of the relationship between the spin polarization of an Fe film and the spins on a compensated NiO(001) plane by using a spin-polarized scanning electron microscope (spin SEM) [10] to observe the Fe domains. To obtain an almost perfectly compensated and clean NiO(001) surface, we cleaved a NiO single crystal then evaporated Fe onto it in an ultrahigh-vacuum (UHV) chamber. We used an atomic force microscope (AFM) to confirm that its surface consists of several-hundred-nanometer-wide atomically flat surfaces. We found that the exchange interaction at the interface forces the Fe spin-polarization distribution to follow the NiO spin configuration even at the compensated NiO(001) surface. By taking into account the spin canting of the NiO, we calculated the spin configurations of Fe/NiO(001). This calculation shows that the Fe and NiO spins at the interface are almost perpendicularly coupled.

Our experiments—except the AFM measurements—were conducted in a spin SEM whose base pressure was less than 2.7×10^{-8} Pa. Shortly after a NiO single crystal was cleaved, a thin film of Fe was evaporated (at 0.03 nm/min) onto the NiO(001) surface at room temperature under no magnetic field. During the evaporation, the vacuum pressure of the chamber was kept below 6.7×10^{-8} Pa. The total film thickness, calculated from

0031-9007/00/85(3)/646(4)$15.00

the evaporation time and the ion current of the evaporating beam, was about 0.9 nm. Then the Fe magnetic domains were observed. Finally, an AFM was used to observe the atomic steps of the sample surface at atmospheric pressure.

Figures 1(a) and 1(b), respectively, are images ($70 \times 71\ \mu\mathrm{m}^2$) of the magnetic domains and the surface morphology of an as-grown Fe film obtained simultaneously by the spin SEM. The color of the domain image shows the direction of the spin polarization, which is given by the radial direction corresponding to the same color in the color wheel shown below the domain image. The perpendicular spin-polarization-component image (not shown) indicates that over the entire observed area the Fe spin polarization is in the film plane. The colors of the magnetic domains are mainly yellow and red, and the corresponding spin-polarization directions are close to $[\bar{1}10]$ and $[\bar{1}\bar{1}0]$ but incline a little towards $[\bar{1}00]$. The arrows in Fig. 1(a) show the spin-polarization directions averaged over an area of $2.4 \times 2.4\ \mu\mathrm{m}^2$. The mean spin-polarization directions averaged over almost the entire regions of the yellow and red domains were, respectively, $15° \pm 11°$ from $[\bar{1}10]$ and $-14° \pm 15°$ from $[\bar{1}\bar{1}0]$. These domains roughly extend along [100] and [010]. There are also fine dark-blue domains inside the yellow domains and fine light-blue domains inside the red domains. The spin-polarization directions of these fine domains are roughly antiparallel to those of the surrounding domains.

The domain structure shown in Fig. 1(a) is related to the crystalline axes of the NiO. Thus, it is quite natural that the exchange interaction at the interface forces the Fe domains to follow the domains of the underlying NiO. Indeed, there are two types of domains in NiO, *T* and *S* domains, and their boundaries are determined by the crystalline axes [11]. The directions of the domain boundaries in Fig. 1(a) indicate that the yellow and red domains represent *T* domains, not *S* domains, formed in the NiO(001) surface. NiO has a rock-salt structure, slightly rhombohedrally deformed below the Néel temperature (523 K). Because of contraction along one of the $\langle 111 \rangle$ axes, the NiO has a crystallographic twin structure. One of the $\{111\}$ planes perpendicular to the contracting axis is a magnetically easy plane. The crystallographic domains twinned with each other coincide with the magnetic domains (*T* domains). The directions of the intersections of the *T*-domain walls with the (001) surface were easily obtained as [100], [010], [110], and $[\bar{1}10]$ [12], which are consistent with those of the domain walls in Fig. 1(a).

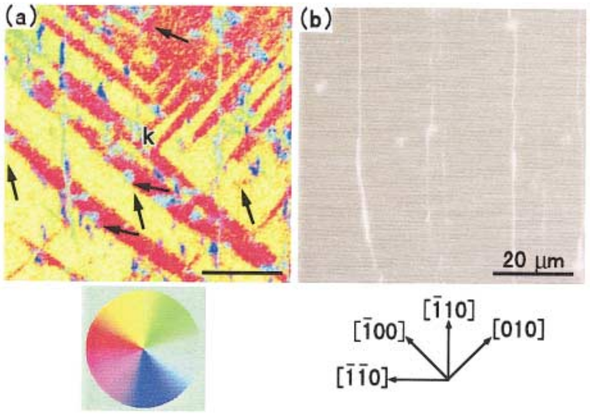

FIG. 1 (color). (a) Magnetic domains and (b) surface morphology of Fe/NiO(001). The color of the domain image shows the Fe spin-polarization direction given by the radial direction of the same color in the color wheel shown below (a). Arrows in (a) represent the spin-polarization directions averaged over about $2.4 \times 2.4\ \mu\mathrm{m}^2$.

The domain structure shown in Fig. 1(a) is very different from the domain structures of F thin films with in-plane spin polarization grown on nonmagnetic substrates. One of these latter structures is nearly a single domain [13,14] and the other consists of irregular-shaped multidomains with bent or rough domain boundaries [14,15]. These previously observed domain structures of the thin films seem common to all thin films grown on nonmagnetic substrates. This also supports the fact that the multidomain structure with straight domain boundaries observed in Fig. 1(a) should reflect the domain structure of the underlying NiO.

Several steps running in the [110] direction are evident in Fig. 1(b). Although such steps nucleate the fine domains, they do not significantly influence the overall domain structure. Figure 2 shows an AFM image of the same sample. The several-atom-high steps run mainly in the [010], [110], and [100] directions. Steps **p** and **q** in the image show, respectively, one-atom-high and two-atom-high steps. The terrace width is typically several hundred nanometers, and there could be many steps between the terraces in Fig. 1(b). Since the Fe was evaporated just after the cleaving in UHV, an ideal exchange-coupled system should be obtained within the atomically flat terraces. Moreover, the spin-polarization configuration within the yellow and red domains in Fig. 1(a) is similar everywhere; therefore, the atomic steps have little effect on the Fe spin-polarization configuration. We can thus conclude that Fig. 1(a) shows the Fe spin-polarization configuration coupled with a perfectly compensated NiO(001) surface.

Because we cannot observe the NiO spin configuration directly with our spin SEM, we estimated the Fe and NiO spin configurations by using a simple model similar to the

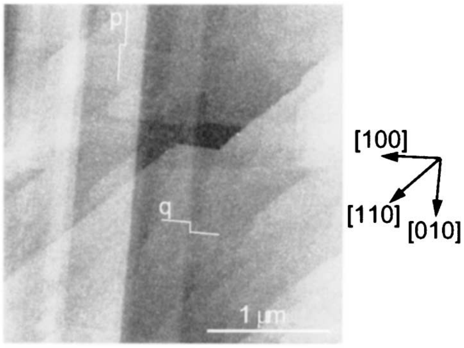

FIG. 2. AFM image of Fe/NiO(001). Steps **p** and **q**, respectively, show one-atom-high and two-atom-high steps.

one Mauri [3] introduced. Our model is based on three assumptions: (i) the Fe spins at the interface are in the same direction, (ii) the anisotropy at the NiO surface is the same as that in the bulk, and (iii) each Fe spin is exchange coupled with its corresponding NiO spin and the lattice mismatch is not considered. Our model and the following calculations are discussed in detail elsewhere. The magnetic energy per unit area of the Fe/NiO(001) system is given by

$$E_t = -JN\mathbf{s} \cdot (\mathbf{s}_1 + \mathbf{s}_2) + (tM^2/\mu_0)\cos^2\Theta + E_w\,, \tag{1}$$

where J is the exchange integral between Fe and Ni atoms, N ($= 2/a^2$, a: lattice constant) is the number of magnetic atoms per unit area at the interface, $\mathbf{s}$ is a Fe spin, $\mathbf{s}_1$ and $\mathbf{s}_2$ are the spins belonging to the NiO sublattices, t ($= 0.9$ nm) is the Fe film thickness, M is the Fe saturation magnetization, Θ is a polar angle of the Fe spin [see Fig. 3(a)], and E_w is the energy of the NiO domain wall created at the interface. The first term of Eq. (1) is the exchange interaction energy at the interface, and the second term is the magnetostatic energy induced by the magnetic charge on the Fe film surface. The Fe domain wall parallel to the film is not created because the Fe film thickness of 0.9 nm is much thinner than the ordinary 180°-wall thickness ($\approx$40 nm) of Fe. We ignored the anisotropy energy of the Fe film, i.e., $|-tK_f\cos^2\theta| \lesssim 4 \times 10^{-5}$ J/m^2 (K_f: anisotropy of Fe), whose effect on E_t is very small.

The exchange interaction at the interface makes the NiO-sublattice spins in the topmost layer rotate at certain angles generally different from each other. The NiO domain wall created at the interface is thus not an ordinary wall with coherent rotation of each sublattice spin. The rotation angles of the pair of NiO sublattice spins can be expressed by the linear combination of the coherent rotation angle and the canting angle of the spin pair. The domain wall energy is therefore approximated by $E_w = E_r + E_c$ (E_r: wall energy due to the coherent rotation; E_c: wall energy due to the canting). Note that one of the {111} planes is the magnetically easy plane and there are three equivalent easy axes (sixfold anisotropy) within an easy plane, e.g., the $\langle 11\bar{2}\rangle$ axes in the (111) easy plane [see Fig. 3(a)]. Therefore, E_r can be approximated as the sum of the two wall energies depending on the out-of-plane anisotropy and in-plane anisotropy. Then, E_r and E_c are given, respectively, by $-2\sqrt{A_{af}K_1}\cos\alpha - (2/3)\sqrt{2A_{af}K_2}\cos 6(\beta - \pi/3)$ and $(4\sqrt{3}A_{af}/a)\cos\eta$ [16] [A_{af}: exchange stiffness; K_1: anisotropy out of (111) plane; K_2: anisotropy in plane of (111); α: coherent rotation angle out of (111) plane; β: coherent rotation angle from $[\bar{1}2\bar{1}]$ within (111); η: canting angle of $\mathbf{s}_1$ and $\mathbf{s}_2$]. To substitute the above expressions of wall energies into Eq. (1), we numerically calculated the energy minimum state of Fe/NiO(001). In the calculation, when $a = 0.42$ nm, $A_{af} = 3 \times 10^{-11}$ J/m (calculated from $J = 19$ meV for bulk NiO), $K_1 = 2.9 \times 10^5$ J/m^3, and $K_2 = 1.5 \times 10^4$ J/m^3 [17], we get $2\sqrt{A_{af}K_1} = 5.9 \times 10^{-3}$, $(2/3)\sqrt{2A_{af}K_2} = 6.3 \times 10^{-4}$, and $4\sqrt{3}A_{af}/a = 0.49$ J/m^2. Since J at the interface in our sample was unknown, we performed numerical calculations varying J from 0.5 to 20 meV. Most of the values of the exchange integrals between 3d- and 3d-metal atoms were between 0.5 and 20 meV [18,19].

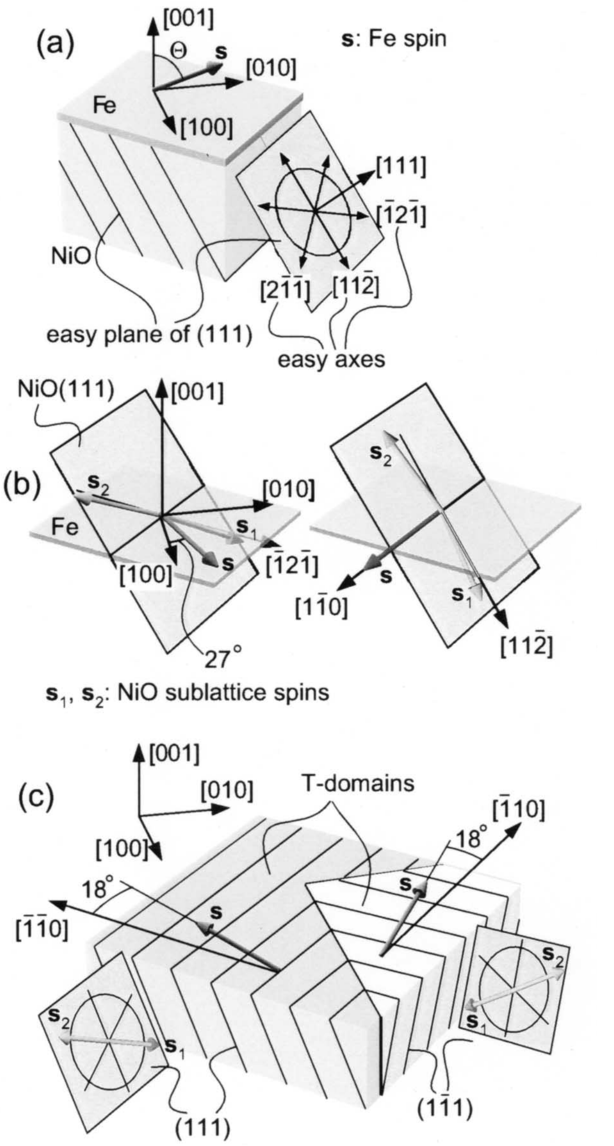

FIG. 3. (a) Easy axes of bulk NiO. (b) The left- and right-hand illustrations represent calculated spin configurations at the Fe/NiO(001) interface, where the NiO spins in the bulk are directed along the easy axes of $[\bar{1}2\bar{1}]$ and $[11\bar{2}]$, respectively. (c) Schematic illustration of the Fe and NiO spin configuration including the T domain structure around $\mathbf{k}$ in Fig. 1(a).

The Fe and NiO spin configurations at the interface ($J = 5$ meV), where the NiO spins in the bulk are directed along the easy axes of $[\bar{1}2\bar{1}]$ and $[11\bar{2}]$, are schematically illustrated in Fig. 3(b). The Fe spins are always in the (001) plane. When the initial spin axis is in the $[\bar{1}2\bar{1}]$ direction, the Fe spin is directed 27° or -153° from [100]; that is, perpendicular to the NiO easy axis of $[\bar{1}2\bar{1}]$. The left-hand illustration of Fig. 3(b) corresponds to the angle of 27°. The NiO spins do not coherently rotate and cant only about 2° from $[\bar{1}2\bar{1}]$ towards the Fe spin. When the initial spin axis is in the $[11\bar{2}]$ direction, the Fe spin is directed −45° ($[1\bar{1}0]$) or 135° ($[\bar{1}10]$) from [100]; that is, also perpendicular to the NiO easy axis of $[11\bar{2}]$. The right-hand illustration of Fig. 3(b) corresponds to the initial spin axis of $[11\bar{2}]$. The NiO spins also do not coherently rotate and cant only about 2° from $[11\bar{2}]$ towards the Fe spin. Canting the NiO spins towards the Fe spin decreases the exchange coupling energy and, consequently, reduces the total magnetic energy instead of increasing the wall energy. When the initial NiO spins are in other easy axes, additional calculations showed that the relationship between Fe and NiO spins is similar to that in Fig. 3(b).

Figure 3(c) shows a schematic illustration of the Fe and NiO spin configuration corresponding to the *T*-domain structure around **k** in Fig. 1(a). The obtained Fe spin corresponding to one in the yellow domain in Fig. 1(a) is directed 18° from $[\bar{1}10]$. Underlying NiO spins are in $(1\bar{1}1)$ and the NiO spins at the interface cant towards the Fe spin from [121]. The Fe spin corresponding to one in the red domain in Fig. 1(a) is directed 18° from $[\bar{1}\bar{1}0]$, and the NiO spins at the interface cant from $[\bar{1}2\bar{1}]$ towards the Fe spin, where the NiO spins in the bulk are in (111), which is twinned with $(1\bar{1}1)$. These Fe spin directions reasonably fit the observed Fe spin-polarization directions, which are 15° ± 11° from $[\bar{1}10]$ in the yellow domain and −14° ± 15° from $[\bar{1}\bar{1}0]$ in the red domain. But there remains ambiguity in the *T*-domain structure. The calculated Fe-spin directions are also the same as in the other three twinned pairs of *T* domains, $(1\bar{1}1)$ and $(\bar{1}\bar{1}1)$, $(\bar{1}11)$ and (111), and $(\bar{1}11)$ and $(\bar{1}\bar{1}1)$. In all cases, the NiO spins are directed along the easy axis but cant towards the Fe spin. This relationship is similar to that shown in Fig. 3(c). In addition, within the range of $0.5 < J < 20$ meV, the obtained spin configurations at the interface are, except for the canting angles, almost the same as those when $J = 5$ meV. The canting angle increases with increasing J (0.2° at $J = 0.5$ meV and 8° at $J = 20$ meV). If $J < 0$ (antiferromagnetic coupling), the NiO spins cant away from the Fe spin.

Using x-ray magnetic linear dichroism, Stöhr *et al.* [20] showed that the NiO spins at the (001) surface are perpendicular to the surface plane. This seems to be not consistent with our calculated NiO spin configuration at the Fe/NiO interface. However, the NiO anisotropy at the interface is different from that at the surface because Fe atoms in Fe/NiO are attached to the Ni atoms at the interface. Ijiri *et al.* [7] obtained a spin configuration of exchange-biased (001) Fe_3O_4/CoO superlattices by using neutron diffraction. They found that the CoO spins are not directed in the surface normal direction but lie in the film plane. The CoO has the same lattice structure as the NiO and a magnetic structure similar to that of NiO. We thus conclude that the large perpendicular anisotropy of NiO is not induced at the Fe/NiO interface.

In summary, we examined how the micromagnetic structure of a 0.9-nm-thick Fe film exchange interacted with the spins of a fully compensated NiO(001) surface. The Fe domains represent the *T* domains of the NiO, and the Fe spin polarization in each domain is in plane and is inclined about 15° (or 14°) from $[\bar{1}10]$ (or $[\bar{1}\bar{1}0]$) to $[\bar{1}00]$. This angle is roughly perpendicular to one of the easy spin axes of NiO. Calculated directions obtained by means of a simple model agree with the observed Fe spin-polarization directions. It is thus concluded that near-perpendicular coupling between the Fe spin and the NiO sublattice spins is realized at the interface. This coupling is similar to that reported by Koon, except that the NiO spins are not parallel to the interface.

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[16] We can obtain the expression of the wall energy of spin canting according to the analogy of the ordinary wall energy of F material. When all spins in NiO cant at an angle of η from the easy axis, the energy increase per unit volume is written as $-(12JS^2/a^3)\cos 2\eta - K_{af}\cos^2\eta \approx (6A_{af}/a^2)(2\sin^2\eta - 1)$, which corresponds to the anisotropy energy, $K_{af}\sin^2\eta$, of the ordinary domain wall. Thus, we substitute $12A_{af}/a^2$ for K_{af} in the ordinary energy expression of $-2\sqrt{A_{af}K_{af}}\cos\eta$. We obtain $-(4\sqrt{3}A_{af}/a)\cos\eta$.

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