Interfacial Uncompensated Antiferromagnetic Spins: Role in Unidirectional Anisotropy in Polycrystalline $Ni_{81}Fe_{19}$/CoO Bilayers

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The uncompensated spins on the surfaces of antiferromagnetic CoO films exhibit a thermoremanent magnetization after field cooling from T_N that has the same temperature dependence as the exchange field of $Ni_{81}Fe_{19}$/CoO bilayers after field cooling. This suggests that these interfacial uncompensated spins are responsible for unidirectional anisotropy. A model based on a calculation of the density of these interfacial uncompensated spins predicts the correct magnitude of the exchange field, as well as the observed inverse dependence on interfacial grain size. [S0031-9007(97)03798-8]

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Exchange anisotropy [1] refers to the exchange interactions at an interface between ferromagnetic (FM) and antiferromagnetic (AFM) materials. Cooling a FM/AFM thin film bilayer from $T_C(\mathrm{FM}) > T > T_N(\mathrm{AFM})$ in a saturating magnetic field produces an unidirectional anisotropy that is manifested as a $\sin\theta$ component in the torque curve and a hysteresis loop displaced along the field axis. The loop shift is called the exchange field H_E. Assuming Heisenberg exchange across an epitaxial atomically smooth FM/AFM interface,

$$H_E = \frac{J_{\mathrm{ex}}\mathbf{S}_{\mathrm{FM}} \cdot \mathbf{S}_{\mathrm{AFM}}}{a^2 M_{\mathrm{FM}} t_{\mathrm{FM}}}, \qquad (1)$$

where J_{ex} is the exchange parameter, $\mathbf{S}_{\mathrm{FM}}$ and $\mathbf{S}_{\mathrm{AFM}}$ are the spins of the interfacial atoms, a is the cubic lattice parameter, and M_{FM} and t_{FM} are the magnetization and thickness of the FM layer, respectively. Observed exchange fields, however, are typically less than a few percent of the values predicted by this idealized model. A quantitatively satisfactory model for a specific exchange coupled system has not been reported.

In this Letter, we report measurements of thermoremanent moments (TRM) in CoO/MgO multilayers. This moment is interfacial and is $\sim 1\%$ of the spins in a monolayer of CoO. The TRM exhibited the same features and temperature dependence as the exchange field of $Ni_{81}Fe_{19}$/CoO bilayers. Since the TRM originates from the uncompensated interfacial AFM spins, they appear to be the spins responsible for unidirectional anisotropy. A model is presented which predicts the observed exchange field. It is based on a calculation of the density of interfacial uncompensated spins and accounts for grain size, orientation, and interfacial roughness.

CoO has a Néel temperature, $T_N = 293$ K. The AFM ordering is characterized by FM aligned (111) planes with adjacent antiparallel (111) planes. The AFM alignment results from the superexchange coupling of the cobalt cations atoms via the p orbitals of the oxygen atoms. CoO/MgO multilayers were used to determine the magnetic properties of uncoupled CoO films since MgO is nonmagnetic. CoO and MgO share the fcc NaCl structure with bulk cubic lattice parameters which differ by only 1.1%. The structural similarity permits interlayer coherent growth. A series of CoO(t_{CoO} Å)/MgO(t_{MgO} Å) multilayers were deposited with CoO thicknesses t_{CoO} ranging from 16 to 103 Å and MgO thicknesses t_{MgO} $\sim$20 Å. The structural characterization and heat capacity measurements of these multilayers are discussed in detail elsewhere [2]. Thin film bilayers of CoO and permalloy ($Ni_{81}Fe_{19}$) were used to determine the role of AFM thickness on the interfacial exchange properties. A series of films was deposited with CoO thicknesses ranging from 100 to 3000 Å; the permalloy layer thickness was fixed at 300 Å. An Ag capping layer was deposited as an oxidation barrier. The substrates were Si (100) wafers with an amorphous native oxide layer ($\sim$30 Å).

The CoO [3] and MgO films were reactively sputtered from a metal target. The permalloy was deposited by dc sputtering from an alloy target. The films were polycrystalline as determined by x-ray diffraction and transmission electron microscopy (TEM). No evidence for any phase other than CoO was detected. The cubic lattice parameter derived from the (111) peak position is 4.27 Å, which is slightly expanded (0.2%) from the bulk parameter (4.260 Å). Magnetic measurements were made with a Quantum Design SQUID magnetometer.

The CoO/MgO multilayers were cooled from 350 to 10 K in zero field (ZFC) and in +10 kOe (FC). Magnetization measurements were made in a 100 Oe field. Field cooling resulted in a stable magnetization—a TRM. Figure 1 shows the FC and ZFC measurements for the $[\mathrm{CoO}(103\ \text{Å})/\mathrm{MgO}(30\ \text{Å})]_{15}$ multilayer. The interfacial TRM density exhibits two features (i) an intermediate temperature region ($200 \geq T \geq 50$ K), where the

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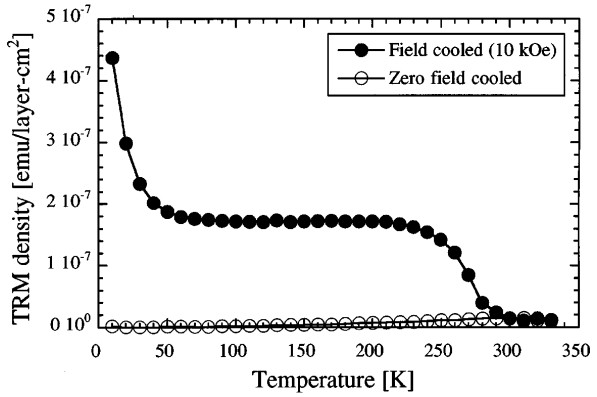

FIG. 1. FC and ZFC moment densities vs temperature of the $[\mathrm{CoO}(103\ \text{Å})/\mathrm{MgO}(30\ \text{Å})]_{15}$ multilayer.

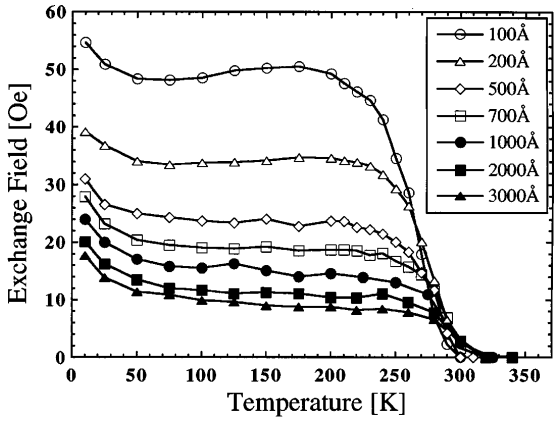

FIG. 2. Temperature dependence of H_E for permalloy (300 Å)/CoO bilayers with CoO thicknesses of 100 to 3000 Å.

magnetization is *independent* of temperature (a "plateau") and (ii) a low temperature increase ($T \leq 50$ K). The magnitudes of these two features scale closely with the number of CoO layers and have no dependence on the net CoO thickness in the multilayers. Therefore the uncompensated moment of both features is an interfacial effect and not a bulk effect. Neutron data [4] give $3.8\mu_B$ for the magnetic moment of Co^{2+} in CoO. Thus the measured interfacial uncompensated moment represents $\sim 1\%$ of the spins in a CoO monolayer.

If the CoO/MgO multilayer is field cooled to 100 K and then zero field cooled from 100 to 10 K, the low temperature increase feature is absent from the TRM. Hence the spins responsible for the low temperature increase require a moderate cooling field below 100 K to align them and will be referred to as the low freezing temperature (LFT) spins. For $T < 100$ K, we observed relaxation and hysteresis. The LFT magnetization and irreversibility disappear above 100 K which is considerably lower than T_N, so we infer that the LFT spins are weakly coupled to the spins in the core of the CoO layers. These spins may be a small fraction of surface spins that are frustrated. Similar magnetic behavior was observed in disordered surface spins in $NiFe_2O_4$ nanoparticles [5] and in isolated spin clusters in fcc $Co_xMg_{1-x}O$ [6].

In Fig. 1, the bifurcation of the FC and ZFC susceptibilities occurs at 295 ± 5 K (the bulk T_N of CoO). Thus the spins responsible for the plateau feature are magnetically coupled to the spins in the core of the CoO layers. For $T < 200$ K, the spins responsible for the plateau have anisotropy fields much larger than the LFT spins since no magnetization reversal was observed in fields as large as -55 kOe. The TRM temperature dependence (for $T > 50$ K) was similar to the temperature dependence of the sublattice magnetization for CoO as determined by neutron diffraction [7]. This correlation further suggests a strong coupling of these interfacial spins to the core spins of the CoO film.

The permalloy/CoO bilayers were cooled from 350 K ($>T_N + 50$ K) to 10 K in a $+10$ kOe field. Figure 2 shows $H_E(T)$ for some of the CoO thicknesses. $H_E(T)$ of permalloy/CoO films exhibit the plateau and low temperature increase features in identical temperature regions as the TRM of the CoO/MgO multilayers. If the TRM of the CoO(103 Å)/MgO(30 Å) multilayer and the $H_E(T)$ of the permalloy/CoO(100 Å) bilayer are normalized at 70 K, the curves overlap at all temperatures except for the region $T < 50$ K. (The lower anisotropy exhibited by the LFT spins is insufficient to pin the permalloy layer.) This correlation strongly suggests that the interfacial uncompensated spins which are responsible for the TRM play an important role in the exchange field mechanism. The correlation also suggests that the temperature dependence of H_E is consistent with strong *net* unidirectional interfacial Heisenberg exchange that is proportional to the moment density of the uncompensated interfacial AFM spins. The observation that $\sim 1\%$ of the interfacial AFM spins are uncompensated is consistent with measured permalloy/CoO exchange fields of $\sim 1\%$ of the interfacial Heisenberg exchange model [Eq. (1)].

The H_E dependence with the CoO thickness suggests a structural origin for the density of uncompensated spins. Cross-sectional TEM showed a columnar structure for all thicknesses. The sizes of the CoO crystallites at the permalloy/CoO interface were examined by TEM plan views of CoO films without any capping layers. The average CoO crystallite diameters for thicknesses of 100, 500, 1000, and 3000 Å were determined by an analysis of the dark and bright field images. Figure 3 shows the linear relationship between $H_E(50\ \text{K})$, which is representative of the plateau value, and the inverse of the CoO crystallite diameter.

The correlation between the temperature dependence of the TRM of the CoO/MgO multilayers and the exchange field of the permalloy/CoO bilayers suggests a direct relationship between the interfacial density of uncompensated spins and the strength of the exchange field. We present a model for H_E which demonstrates its connection with the interfacial density of uncompensated spins and predicts the inverse relationship between H_E and the grain diameter. We assume that each CoO crystallite is a single AFM domain. We define the normal to the FM aligned (111)-type spin plane as $\hat{\mathbf{p}}$ and define the probability of finding a

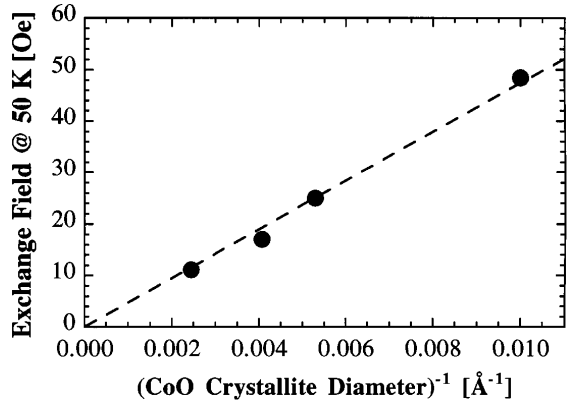

FIG. 3. Linear relationship between the plateau value of $H_E(T = 50\ \mathrm{K})$ of the permalloy/CoO bilayers and the inverse of the CoO crystallite diameter.

crystallite with that orientation at the AFM film surface as $f(\hat{\mathbf{p}})d\Omega_{\hat{\mathbf{p}}}$. We define the easy axis (known to be near the tetragonal axis in bulk CoO [4]) for a crystallite as $\hat{\mathbf{e}}$ and the probability of finding a crystallite with that easy axis as $g(\hat{\mathbf{e}})d\Omega_{\hat{e}}$ (the sign of $\hat{e}$ will be specified below). The FM aligned spin planes will intersect the interface in a series of atomic steps with spacing $d_{111}[1 - (\hat{\mathbf{n}} \cdot \hat{\mathbf{p}})^2]^{-1/2} \times (= d_{111}/\sin\theta)$, where $d_{111}(= a\sqrt{3}/3 = 0.577a)$ is the spacing of (111) planes and $\hat{\mathbf{n}}$ is the film normal, as indicated in Fig. 4. The interfacial layer of spins of each crystallite will consist of alternate rows of antiparallel spins pointing in the $\pm\hat{\mathbf{e}}$ directions. If $\hat{\mathbf{p}}$ is close to $\hat{\mathbf{n}}$, there may be two or more rows of parallel spins from each (111) spin plane at the interface. The number of adjacent rows of parallel spins may be estimated as

$$N_{\mathrm{rows}} = \mathrm{trunc}\left\{\frac{d_{111}}{d_{110}}[1 - (\hat{\mathbf{n}} \cdot \hat{\mathbf{p}})^2]^{-1/2}\right\} \qquad (2)$$

where "trunc" gives the largest integer less than the argument and d_{110} represents the in-plane spacing. The interfacial exchange energy for crystallite i is

$$U_i = -\sum_{j=1}^{N} J_{\mathrm{ex}}\mathbf{S}_{\mathrm{FM},j} \cdot \mathbf{S}_{\mathrm{AFM},j}\,.$$

If the ferromagnetic layer is in a single domain state,

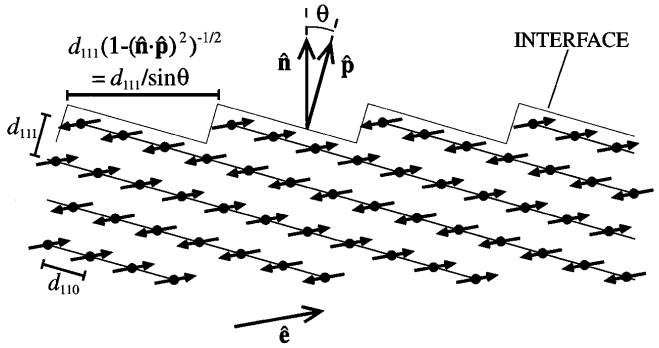

FIG. 4. Schematic of interface cross section. Film normal is $\hat{\mathbf{n}}$, $\hat{\mathbf{p}}$ is the normal to the parallel spin plane (111) of the AFM, and $\hat{\mathbf{e}}$ is the AFM spin axis (in this case $N_{\mathrm{rows}} = 4$).

$$U_i = -\mathbf{S}_{\mathrm{FM}} \cdot \left[\sum_{j=1}^{N} J_{\mathrm{ex}}\mathbf{S}_{\mathrm{AFM},j}\right] = -\mathbf{S}_{\mathrm{FM}} \cdot [J_{\mathrm{ex}}(N_{+,i} - N_{-,i})S_{\mathrm{AFM}}\hat{\mathbf{e}}],$$

where $N_{\pm}$ is the number of interfacial spins pointing in the $\pm\hat{\mathbf{e}}$ direction, and we define the sign of $\hat{\mathbf{e}}$ such that $J_{\mathrm{ex}}(N_{+,i} - N_{-,i}) \equiv J_{\mathrm{ex}}\Delta N_i$ is positive. When the film is field cooled below T_N, the AFM domain will orient itself such that $+\hat{\mathbf{e}}$ has a component in the direction of the adjacent FM spins (i.e., $\hat{H}_{\mathrm{cool}}$), therefore,

$$U_i^{\mathrm{FC}} = -|J_{\mathrm{ex}}|S_{\mathrm{AFM}}S_{\mathrm{FM}}\Delta N_i|\hat{\mathbf{e}} \cdot \hat{\mathbf{H}}_{\mathrm{cool}}|\,.$$

The total interfacial energy is then

$$U_{\mathrm{tot}}^{\mathrm{FC}} = -|J_{\mathrm{ex}}|S_{\mathrm{AFM}}S_{\mathrm{FM}}N_c \times \left[\int d\Omega_{\hat{\mathbf{p}}}\, f(\hat{\mathbf{p}})\Delta N_i\right]\left[\int d\Omega_{\hat{\mathbf{e}}}\, g(\hat{\mathbf{e}})|\hat{\mathbf{e}} \cdot \hat{\mathbf{H}}_{\mathrm{cool}}|\right],$$

where $N_c = \mathrm{area}/L^2 =$ total number of crystallites at the interface. If we assume that the AFM spins are frozen below T_N and that $g(\hat{\mathbf{e}})$ is azimuthally symmetric, we find that the interfacial energy as a function of $\hat{\mathbf{S}}_{\mathrm{FM}}$ is

$$\frac{U_{\mathrm{tot}}^{\mathrm{FC}}(\hat{\mathbf{S}}_{\mathrm{FM}})}{\mathrm{area}} = -(1/L^2)\,|J_{\mathrm{ex}}|S_{\mathrm{AFM}}S_{\mathrm{FM}}(\hat{\mathbf{H}}_{\mathrm{cool}} \cdot \hat{\mathbf{S}}_{\mathrm{FM}}) \times \left[\int d\Omega_{\hat{\mathbf{p}}}\, f(\hat{\mathbf{p}})\Delta N_i\right] \times \left[\int d\Omega_{\hat{\mathbf{e}}}\, g(\hat{\mathbf{e}})|\hat{\mathbf{e}} \cdot \hat{\mathbf{H}}_{\mathrm{cool}}|\right], \qquad (3)$$

Finally, following Malozemoff [8], we have

$$H_E = \frac{[U_{\mathrm{tot}}^{\mathrm{FC}}(-\hat{\mathbf{S}}_{\mathrm{FM}}) - U_{\mathrm{tot}}^{\mathrm{FC}}(\hat{\mathbf{S}}_{\mathrm{FM}})]/\mathrm{area}}{2M_{\mathrm{FM}}t_{\mathrm{FM}}}\,,$$

where M_{FM} and t_{FM} are the magnetization and thickness of the FM layer.

To compute the first integral in Eq. (3), we numerically calculated the average number of uncompensated spins for elliptical crystallites as a function of N_{rows}, which is related to the orientation $\hat{\mathbf{p}}$ by Eq. (2). We incorporated interfacial roughness as specified by the average lateral dimension of interfacial topographic features. We first mapped the interfacial plane onto a square lattice for computational convenience (this mapping slightly changes the shape of the model crystallites, but this is not significant). As described above, the interfacial plane consists of a periodic pattern of N_{rows} rows of spins in the $+\hat{\mathbf{e}}$ direction, followed by N_{rows} rows of spins in the $-\hat{\mathbf{e}}$ direction. To simulate roughness, we superimposed elliptical "islands" of monolayer thickness on the spin map. The effect of adding one atomic layer is to reverse the direction of the spin at each site covered by the island, since successive layers of CoO spins have opposite direction. The major axis of these islands was kept fixed (denoted "feature diameter"), while the position, orientation of the major axes, and aspect ratio (from 1:1 to 1:1.5) were varied. Islands were superimposed sequentially, allowing overlap between them, until

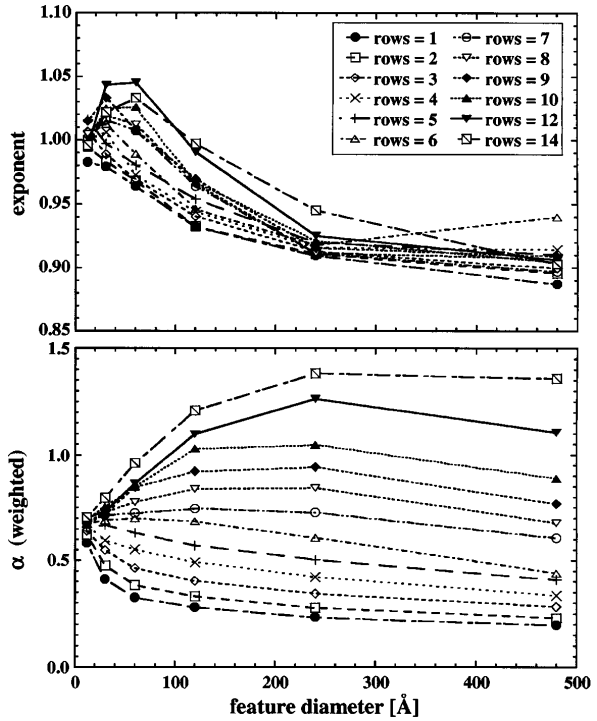

FIG. 5. Calculated values of the prefactor α and the exponent in power-law fits of $\langle \Delta N \rangle$ vs feature diameter and N_{rows}. $N_{\text{rows}} = 1$ corresponds to $\theta > 24.1°$ and $N_{\text{rows}} = 14$ corresponds to $3.3° > \theta > 3.1°$. α is weighted based on equal population of domains with the four different (111)-type parallel spin planes.

the total area of the islands equaled the total area of the spin map. To compute the number of uncompensated spins for a model crystallite, ΔN_i, we simply added the total number of spins in each direction within an elliptical region, having major axis equal to L, on the spin map. We created a spin map which was several times larger than L, so a statistical average could be taken by varying the position, orientation of the major axis, and aspect ratio of the model crystallite (similar to the above procedure for generating roughness). Averages were taken by computing ΔN_i for 10^6 model crystallites.

The results of these calculations are summarized as follows: (1) a perfectly regular interface, with equally spaced atomic terraces, results in $\langle \Delta N \rangle = \alpha (L/d_{110})^{0.5}$, (2) the addition of roughness results in $\langle \Delta N \rangle \simeq \alpha (L/d_{110})^{0.90\sim1.04}$, (3) for small values of feature diameter $\langle \Delta N \rangle = 0.6(L/d_{110})$, independent of N_{rows}, and (4) the prefactor α increases as N_{rows} increases. These results are shown in Fig. 5. Since the result of Eq. (3) is proportional to $\langle \Delta N \rangle / L^2$, the rough case gives $H_E \sim L^{-1}$, in agreement with the experimental results of Fig. 3.

As an example, we calculate H_E for the 100 Å thick sample as follows. Recent work by the authors suggests [9], via a mean field analysis of exchange coupled bilayers having different FM layers, that the interfacial exchange is a direct exchange between the metal ions. Following this approach, we estimate that the constant $J_{\text{ex}} S_{\text{AFM}} S_{\text{FM}} = \overline{T}_{\text{C}}/8 = 143$ K, where $\overline{T}_{\text{C}}$ is the Curie temperature of the interfacial alloy $(Ni_{81}Fe_{19})_{0.5}Co_{0.5}$ [10]. X-ray diffraction rocking curves of the CoO (222) peak are nearly flat, indicating the absence of a strong crystallographic texture. Therefore we assume uniform distributions of crystallite orientations [i.e., $f(\hat{\mathbf{p}})$ and $g(\hat{\mathbf{e}})$ constant]. With these parameters, we calculate exchange fields of 107, 80, 63, 50, 40, 31 Oe for topographic feature diameters of 12, 30, 60, 120, 240, 480 Å, respectively. The experimental result of 48 Oe is consistent with a topographic feature diameter of approximately 120 Å. This result is consistent with roughness of only a few "extra" atomic steps across the face of each crystallite.

In summary, we measured the interfacial uncompensated moment on an AFM surface as a TRM of a field cooled CoO/MgO multilayer. The temperature dependence of these uncompensated spins parallels that of the exchange field of permalloy/CoO bilayers. A model for unidirectional anisotropy correctly predicts the inverse dependence of the uncompensated spins on grain size and the correct magnitude of H_E.

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