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Polarized neutron reflectometry study of an exchange biased $\text{Fe}_3\text{O}_4/\text{NiO}$ multilayer

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Polarized neutron reflectometry study of an exchange biased $\text{Fe}_3\text{O}_4/\text{NiO}$ multilayer

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Polarized neutron reflectometry was performed on a $[\text{Fe}_3\text{O}_4/\text{NiO}]_{\times 15}$ multilayer, molecular beam epitaxy grown on a (001) MgO substrate, in both exchange biased and nonexchange biased states. The Fe_3O_4 layers exhibit a depth-dependent magnetic profile characterized by a change in the magnetization near the $\text{Fe}_3\text{O}_4/\text{NiO}$ interfaces. In the exchange biased state, measurements performed in the two saturated states of the magnetic hysteresis loop reveal magnetic differences in the Fe_3O_4 , possibly due to the interfacial domain wall formation in the ferromagnetic layer.

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When a ferromagnetic (F) layer in contact with an anti-ferromagnetic (AF) layer is field-cooled through the AF Néel temperature T_N , a unidirectional anisotropy may be created, resulting in a hysteresis loop shifted along the field axis.¹ The magnitude of this shift is the exchange biasing field, H_{eb} . Recently, the interest in the exchange biasing (EB) effect has been revived²⁻⁵ due to the current application of EB in magnetoresistive sensors for recording heads and the need for materials with improved EB characteristics. More specifically, the low H_{eb} observed in many systems is a major scientific and practical problem. For example, in the extensively studied $\text{Ni}_{80}\text{Fe}_{20}/\text{FeMn}$ system, H_{eb} is about 200 times lower than predicted from the original model of Meiklejohn and Bean.^{2,3,5} They proposed that H_{eb} is the field necessary to break all the exchange couplings across a flat and uncompensated AF/F interface. Consequently, alternative models have been proposed invoking either planar⁶ or perpendicular⁷ domain walls to reduce the predicted H_{eb} value.

In principle, structures such as domain walls should be detectable by polarized neutron reflectometry (PNR) if they influence the in-plane sample magnetization.⁸ However, previous PNR studies on EB systems involving metallic $\text{Ni}_{80}\text{Fe}_{20}$ did not show any deviations from uniform sample magnetization.⁹⁻¹¹ Here we report the first results from PNR showing unusual magnetic behavior around the F/AF interface in an EB system. We chose $\text{Fe}_3\text{O}_4/\text{NiO}$ in line with our recent work on oxidic $\text{Fe}_3\text{O}_4/\text{CoO}$ systems,¹² as these systems have simple collinear spin structures. Recent progress in the synthesis of oxidic multilayers makes such studies possible.^{13,14}

In PNR, a collimated beam of polarized neutrons impinges on a flat sample at grazing incidence θ . The reflected intensity is measured versus the component of the incident neutron wave vector normal to the sample surface $q_0 = 2\pi \sin \theta / \lambda$, where λ is the neutron wavelength. The reflectivity depends on the sample scattering length density (SLD)

$\Gamma_{\pm}(z)$ which is a function of the depth z from the surface and can be expressed as the sum of two contributions: $\Gamma_{\pm}(z) = \Gamma_n(z) \pm \Gamma_m(z)$. The first, $\Gamma_n(z)$, is the nuclear SLD and depends on the atomic nature of the sample. The second, $\Gamma_m(z)$, is the magnetic SLD, proportional to the in-plane magnetization $\mu(z)$. The $\pm$ in $\Gamma_{\pm}(z)$ corresponds to the neutron spin parallel or antiparallel to $\mu(z)$, as the reflectivities of an in-plane magnetized sample are different for the two neutron spin states. The in-plane magnetization profile is determined by numerical simulation of the two reflectivities $R^{\pm}(q_0)$. In the simulation, each layer in the sample is divided into N slabs of constant $\Gamma_{\pm}(z)$ value. A least-squares algorithm is used to fit to the measured reflectivity by adjusting each $\Gamma_{\pm}(z)$ value and the slab thicknesses. The minimum value of N is chosen which allows a good fit to the measured reflectivity (here $N=3$). This then leads to the density depth profile.

A single-crystalline $[\text{Fe}_3\text{O}_4 (16 \text{ nm})/\text{NiO} (16 \text{ nm})]_{\times 15}$ superlattice was deposited on a (001) MgO substrate using oxygen plasma-assisted molecular beam epitaxy.¹⁴ During growth, the substrate temperature was 548 K, above the NiO bulk T_N of 521 K. Subsequently, the sample was cooled to room temperature in a magnetic field of 8 kA/m, applied along the [100] in-plane crystalline direction to induce unidirectional anisotropy. The sample was characterized by reflection high-energy electron diffraction (RHEED) and x-ray diffraction and found to be single crystalline with all layers aligned with the substrate lattice, and showing long-range lattice coherence.^{14,15} A detailed x-ray reflectometry (XRR), analysis¹⁶ (Fig. 1) showed that the superlattice periodicity was $32.4 \pm 0.5 \text{ nm}$, with a rms roughness of 0.8 nm on either side of the interfaces, and that a variation in the layer densities occurred. The Fe_3O_4 layer density is minimum in the layer center and increases towards the interfaces by 5%.

Magnetometry studies were performed in a SQUID magnetometer (Quantum Design, MPMS5). The saturation magnetization at 300 K was 350 kA/m, 73% of the bulk value of Fe_3O_4 of 480 kA/m. This low value agrees with previous

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FIG. 1. The intensity of x-ray reflectivity I in counts per second (cps) vs the perpendicular x-ray wave vector, q_0 . The solid line corresponds to the best fit.

observations in $\text{Fe}_3\text{O}_4/\text{NiO}$ ¹⁷ and $\text{Fe}_3\text{O}_4/\text{CoO}$.^{12,13} The biasing H_{eb} was 5 kA/m. The PNR measurements were performed on the time-of-flight (TOF) reflectometer ROG of the IRI in Delft.¹⁸ In the TOF method, variation of q_0 is obtained by reflecting neutrons of all wavelengths, with λ determined by measuring the neutron velocity. All measurements were performed at room temperature. An electromagnet created a maximum field of 710 kA/m.

Our purpose is to understand what modifications appear when the AF/F system is made unidirectionally anisotropic (EB state). The sample was therefore measured in both EB and nonexchange biased (NEB) states. To simplify measurement interpretation, we determined the structural properties of the sample by XRR and, independently, by PNR when magnetizing the sample perpendicularly to the surface. Since at a perpendicular field $H=710$ kA/m the in-plane magnetization is zero, the neutron reflectivity depends solely on $\Gamma_n(z)$ and is independent of the neutron spin state. This confirmed the structural information obtained by XRR. Figure 2 shows this measurement under the perpendicular field. The peaks at 0.14 and 0.31 nm^{-1} correspond to first- and third-order Bragg reflections from the superlattice periodicity. The weak third-order peak indicate that the NiO and Fe_3O_4 layers have nonuniform $\Gamma_n(z)$ profile which reduce the reflective potential barrier seen by the neutrons at each interface.

The previously described simulation procedure leads to the fitted reflectivity (solid line in Fig. 2) and to the nuclear density profile of Fig. 3. Due to the large number of parameters, the profile is not unambiguous. However, all solutions found were similar and exhibit the same global behavior, consistent with x-ray reflectometry analysis. Throughout the sample, $\Gamma_n(z)$ has a minimum value at the center of the Fe_3O_4 layers and increases near the interfaces. From XRR it is concluded that this variation can be attributed to a density variation within the layers. Oppositely, $\Gamma_n(z)$ is maximum at the layer center in NiO. For comparison, the dashed line in Fig. 2 represents the calculated reflectivity assuming the layers have a constant $\Gamma_n(z)$ value, equal to the average of the layer profile found.

The reflectivities $R^\pm(q_0)$ of the in-plane saturated NEB sample were measured in a field of 640 kA/m, at the satu-

FIG. 2. Polarized neutron reflectivity vs perpendicular neutron wave vector component q_0 . In the top curve, the sample was magnetized perpendicularly to the film plane. In the two lower curves (corresponding to + and - neutron spin states), the sample was magnetized in-plane in the nonexchange biased (NEB) state. All curves have been offset for clarity. The solid lines are the calculated fits. The dashed line corresponds to a constant nuclear scattering length profile in each layer.

rated part of the hysteresis loop (Fig. 2). The magnetic depth profile (Fig. 3, solid line) was obtained using the nuclear profile. The result is a very pronounced variation in the Fe_3O_4 layer magnetization. Generally, the magnetization is maximum in the center of the layer where it is close to bulk value, but drops sharply towards the interfaces. The average magnetization is 340 kA/m, very close to the 350 kA/m obtained by SQUID magnetometry. Adjustment of the NiO layer parameters showed no evidence for a noticeable magnetic contribution from this layer.

In the EB state, $R^\pm(q_0)$ were measured in both left and right saturated states of the F hysteresis loop for $H=\pm 640$ kA/m (denoted states EBL and EBR). $R^\pm(q_0)$ for EBR differ only very slightly from the NEB case and the profiles are experimentally indistinguishable. In contrast, EBL exhibits

FIG. 3. Nuclear and magnetic profiles deduced from reflectivity fits vs depth for the top 170 nm of the sample. The top curve is the nuclear scattering length density. The bottom curves are the in-plane magnetization profiles of the saturated sample for: (a) NEB and exchange-biased EBR (solid line), (b) exchange-biased EBL (dotted line).

FIG. 4. Polarized neutron reflectivity for both neutron spin states, R^+ and R^- , for the exchange biased EBL state. The solid line is the calculated fit. Note the difference with the dotted line which is the calculated reflectivity R^+ for the EBR case.

clear differences with EBR. This is demonstrated by the comparison with the fitted reflectivity (Fig. 4) for the EBR measurement. The rate of decay of the reflectivity between the Bragg reflections is clearly different. The magnetic depth profile for EBL is shown in Fig. 3. The magnetization profiles for the EBR and EBL state appear quite different, particularly near the interfaces, varying as much as 40%. The calculated average sample magnetization for EBL is 325 kA/m, the same as EBR to within 5%.

Thus there is a clear deviation from uniform magnetization in the Fe_3O_4 layers, but no observable contribution from the NiO. A possible explanation for this change in the magnetization near the interface could be vacancies in the Fe_3O_4 at the interface. However, Mössbauer spectroscopy¹⁹ shows that thin, MBE-grown, Fe_3O_4 films retain the magnetic features of the bulk material, except very close to the surface (the first 0.5 nm). A “dead” layer of 0.5 nm on each interface would only lead to an overall magnetization reduction of 6.7%. The 27% reduction observed in this study is clearly much larger. Moreover, XRR showed that the roughness or indeterminacy of the interface in this sample is only 0.8 nm. A recent Mössbauer study of similar MBE-grown $\text{Fe}_3\text{O}_4/\text{NiO}$ superlattices²⁰ showed that at each interface the Fe octahedral-site moments are canted between 51° and 60° out of the plane over a region of 1.5–2.0 nm to align with the adjacent Ni moments across the F/AF interface.²⁰ This correlates with the reduction of the F in-plane moments at the interfaces observed in PNR. Under rotation of the applied F moments pinned to the Ni moments are, depending on their distance from the interface, not able to rotate, leading to a region of progressive moment rotation or an F domain wall when the field transits from the EBL value to that at EBR, creating the observed differences between the two states. A domain wall in the F layer would reduce the effective inter-

face exchange coupling, leading to a reduction in H_{eb} . Interestingly, we suggest that domain wall formation appears in the F layer, whereas current models for exchange biasing^{6,7} consider this only in the AF. This result is logical because the condition for exchange biasing is that the AF must have the largest anisotropy, making domain wall formation energetically preferable in the F layer. Some extension of the F domain wall into the AF cannot be ruled out, as such a parallel AF domain wall may be below what is detectable by PNR.

In conclusion, we have established the existence of a strong deviation from uniform magnetization in thin Fe_3O_4 layers exchange coupled to AF layers of NiO. When the sample is exchange biased, differences, located near the interfaces, are observed between the magnetization depth profiles of the opposite saturated states of the F hysteresis loop. These results suggest the formation of a domain wall in the F layer parallel to the interface, and should lead to an adaptation of current models for exchange biasing, as this domain wall might be responsible for the reduced H_{eb} values found, at least, in oxides systems.¹²

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