Stabilization of Antiferromagnetic Order in FeO Nanolayers

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We have studied the evolution of the magnetic state of a nanometer thick antiferromagnetic (AFM) FeO layer during its formation using nuclear resonant scattering of synchrotron radiation. In contact to ferromagnetic Fe, the FeO layer does not show magnetic order at room temperature (RT). Once embedded between two Fe layers, magnetic coupling to the adjacent ferromagnets leads to a drastic increase of the Néel temperature far above RT, while the blocking temperature remains below 30 K. The presented results evidence the role that the ferromagnetic surrounding plays in modifying the magnetic state of ultrathin AFM layers.

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Magnetic data storage technology is reaching the bottom of the nanoscale. The stabilization of magnetic order in low-dimensional structures becomes a key issue. Below a critical thickness, ferromagnets undergo a superparamagnetic relaxation where spins are thermally excited, and the system does not show magnetic hysteresis anymore. Exchange bias bilayer systems composed of a ferromagnet (FM) and an antiferromagnet (AFM) have been recently reported to substantially reduce the critical thickness under which the ferromagnet undergoes superparamagnetic relaxation [1,2]. Layered FM/AFM systems, where an AFM layer (with high anisotropy) is used to pin the magnetization of the FM, are used to create reference magnetic layers in devices. Below a certain temperature (the blocking temperature T_B) the spins in the antiferromagnet are frozen and exchange coupling at the FM/AFM interface leads to a shift of the hysteresis loop and an increase of the coercivity, this phenomenon being known as the exchange bias effect [3,4].

In the ultrathin film limit, the thermal stability of these FM/AFM bilayers is strongly modified [5,6]. In exchange bias systems, one usually distinguishes the Néel (ordering) temperature T_N of the antiferromagnet and the blocking temperature T_B below which exchange bias occurs [7]. For thick AFM layers (> 50 nm), T_N and T_B are usually found to be equivalent. Field cooling the system below T_N directly leads to the freezing of spins (and the appearance of exchange bias). Early studies showed that T_B was usually decreasing with decreasing thickness of the AFM layer, and it was assumed that T_N was decreasing accordingly due to a finite size effect [5]. Recently, Vallejo-Fernandez *et al.* found out that the evolution of the exchange bias field H_e and T_B for polycrystalline films was in fact linked to the average grain volume of the AFM [6], which typically decreases with the layer thickness. This results in a decreased stability against thermal excitation and a decrease of T_B . It should be noted that there is still an underlying physics problem as data on epitaxially grown AFM/FM

bilayers also show a reduction of T_B with a decreasing AFM layer thickness [8,9]; this is still the subject of different theoretical considerations [5,10]. The evolution of T_N relative to T_B was investigated by Van der Zaag *et al.* on $\text{Fe}_3\text{O}_4/\text{CoO}/\text{Fe}_3\text{O}_4$ multilayers [9]. They found that T_N and T_B differ in the ultrathin film limit i.e., T_N is increasing and T_B is decreasing with decreasing AFM layer thickness. The increase of T_N was interpreted using a mean field theory, which describes the influence of the surrounding spins as a mean magnetic field acting on the spins in the FM and AFM layers [9].

In this Letter, we present an *in situ* study of the formation of an ultrathin antiferromagnetic FeO layer on Fe using nuclear resonant scattering (NRS) of synchrotron radiation. The isotope sensitivity of the technique is used here to study exclusively the FeO part of the system via introduction of isotopic probe layers. The sensitivity to the hyperfine interaction of the ^{57}Fe nuclei [11] allows one to follow the evolution of the chemical and magnetic environment of the ^{57}Fe atoms during the growth. We observe that the magnetically disordered FeO layer undergoes a magnetic transition only once the Fe layer deposited on its top is sufficiently thick to sustain long-range ferromagnetism. Temperature dependent measurements show a drastic increase of T_N and the occurrence of exchange bias at low temperature, as predicted in Ref. [9]. These findings demonstrate how ultrathin antiferromagnets can be stabilized by embedding them between thin ferromagnets.

The *in situ* experiment was carried out in a dedicated ultrahigh vacuum (UHV) chamber [12] at the nuclear resonance beam line of the European Synchrotron Radiation Facility ESRF (Grenoble, France) ID18 [13]. First, a 2 nm thick ^{56}Fe layer was deposited on a W(110) single crystal with a 0.5 nm ^{57}Fe layer on its top. This isotopic substitution does not modify the chemical and magnetic structure of the system but ensures that only the ^{57}Fe part of the sample is probed via NRS. At this stage the nuclear time spectrum (the time dependent deexcitation of

the ^{57}Fe nucleus after pulsed excitation using synchrotron radiation [11]) shows a single frequency beating associated with a hyperfine field B_{hf} of 32.6 T [see Fig. 1(a)], typical for bcc Fe at room temperature (RT). The layer was subsequently oxidized by admission of molecular oxygen at a pressure of 1×10^{-5} mbar for 500 s. A drastic change is seen [Fig. 1(b)] in the time spectrum and the hyperfine field distribution (HFD). Two components can be identified: one with a large B_{hf} centered around 29 T (35%) and several low B_{hf} distributions accounting for nonmagnetic states. This collection of sites is consistent with the disordered oxide state reported in Ref. [14] for saturated native iron oxide layers, where it is also shown that this oxidation procedure leads to a disordered (eventually amorphous) crystalline structure. The oxide layer is then reduced to the desired FeO structure by deposition of 0.8 nm of ^{56}Fe . A 1.6 nm FeO layer enriched in ^{57}Fe is formed. It should be stressed that the study reported in [14] showed that the oxide is completely reduced to FeO after deposition of the first Fe monolayer. The additional Fe remains on top in a pure metallic state. The corresponding time spectrum [Fig. 1(c)] is fitted using a main B_{hf} component ($\sim 90\%$) centered at 5 T, attributed to nonmagnetic FeO. So one can expect that T_N does not significantly increase or is lower than the bulk value (bulk $T_N = 198$ K), although the layer

is in contact at one of its interfaces with a ferromagnetic Fe layer.

The deposition of a second 0.8 nm thick ^{56}Fe layer (which brings a nominal ^{56}Fe thickness of 1.6 nm on top of the FeO layer) drastically changes the picture [see Fig. 1(d)] when high frequency beatings, associated with large values of B_{hf} , appear in the time spectrum. It is known that this further deposition of Fe does not modify the chemical structure of the FeO nanolayer (which is already in the most reduced state [14]). The observed changes are attributed to a modification of the magnetic interaction that the FeO layer experiences. In fact, one can expect that this second deposition goes along with the transition to long-range magnetic order in the top Fe layer. One distinguishes mainly two sites in the HFD, which are attributed to Fe atoms either fully coordinated to oxygen atoms and interfacial atoms with slightly different environment. The appearance of magnetic ordering in the FeO layer indicates that T_N is effectively increased only when the AFM layer is embedded between FM layers. These findings complement very well the stabilization mechanism proposed by Van der Zaag *et al.* [9]: The Néel temperature of ultrathin AFM can be effectively increased in the case of buried layers while this seems not to be the case for a free AFM layer.

We investigated *ex situ* the temperature dependence of the magnetic state of the embedded FeO layer on a similar system which exhibits the layer sequence Si-substrate/Ta/ ^{56}Fe (2 nm)/ ^{57}FeO (1.5 nm)/ ^{56}Fe (2 nm)/Ta. NRS time spectra were recorded in the temperature range $10 \text{ K} < T < 400 \text{ K}$ to follow the evolution of the corresponding HFD. Two representative spectra recorded at 10

FIG. 1 (color online). Nuclear time spectra (left panels) and corresponding hyperfine field distributions (right panels) recorded during the growth of an Fe/native-oxide/Fe trilayer at room temperature. The solid lines on the time spectra are fits. The 1.6 nm buried FeO layer appears to be magnetically ordered only when the top Fe layer is thick enough to develop long-range magnetic order.

FIG. 2 (color online). Upper panels: Nuclear time spectra recorded on a $^{56}\text{Fe}/^{57}\text{FeO}(1.6 \text{ nm})/^{56}\text{Fe}$ trilayer at 300 K and at 10 K. The solid lines are fits. Lower panel: Hyperfine field distributions extracted from the fits of the time spectra.

and 300 K and the extracted HFD are displayed in Fig. 2. At 300 K, the HFD is similar to the one recorded *in situ* [Fig. 1(d)], proving that the magnetic state of the FeO layer is essentially the same. Taking the relative disorder of this FeO layer (created by oxidation) [14], the low hyperfine field contribution centered around 8–10 T is expected to originate from Fe atoms at grain boundaries or in interstitial positions.

The magnitude of B_{hf} is in first approximation proportional to the local magnetic moment of the atoms. The evolution of the mean value of the large $B_{\text{hf}}(\text{FeO})$ component with temperature is a good indicator of the stability of the antiferromagnetic state. It is seen in Fig. 3 that $B_{\text{hf}}(\text{FeO})$ is decreasing only slowly up to 400 K. Above this temperature, thermal diffusion started to irreversibly change the structure of the sample. A fit of the curve using a law of the form [15]

$$B_{\text{hf}} \propto m_{\text{AFM}} \propto (T_N - T)^{1/3}$$

where m_{AFM} is the magnetization of the Fe in the FeO layer, yields a T_N of 800 ± 30 K, a value much larger than the bulk value of 198 K. This large increase of T_N is in accordance with the prediction of the mean field theory used to describe the evolution of the magnetic order in ultrathin FM/AFM superlattices [9,16,17]. In the case of a FM/AFM system, the Curie temperature T_C of the FM is only weakly dependent on the relative thickness of the FM and AFM layers [17]. In contrast, the T_N of the AFM layer is gradually approaching T_C when its thickness is reduced. This mean field approach usually considers the coupling experienced by the spins in the center of each layer with its nearest neighbors under periodic boundary conditions which are compatible with a superlattice structure. As we obviously see here, this type of mechanism allows us to understand the behavior of the buried FeO layer, but cannot be used to describe the Fe/FeO bilayer system, where the FeO exhibits one free interface. In that case, the antiferromagnet seems to behave essentially as an isolated AFM layer, i.e., one can expect that T_N is not modified [9] or is eventually decreased due to the increasing relative number of interfacial (loosely coupled) spins [5].

The second phenomenon associated with the ultrathin nature of the AFM is the decrease of T_B with decreasing

FIG. 3. Temperature dependence of the mean value of the large B_{hf} component displayed in Fig. 2. The solid line is a fit using a $(T_N - T)^{1/3}$ law, which yield a Néel temperature of 800 ± 30 K.

thickness. For temperatures T between T_B and T_N the spins in the AFM are free to rotate their magnetization vectors with respect to the ferromagnet. As a time-resolved technique, NRS constitutes a very sensitive probe of relaxation dynamics in magnetic systems [18]. The scattering process extends over a time scale that is determined by the lifetime τ of the excited nuclear level (141 ns in the case of ^{57}Fe). If the relaxation time of the magnetic fluctuations is significantly shorter than τ , the magnetic signal averages out and the sample appears to be nonmagnetic. With decreasing temperature the fluctuation time eventually becomes larger than the scattering time, i.e., the measurement yields a snapshot of the sample where the magnetic order seems to be frozen in. Figure 2 displays evidence for such a transition in our sample: at about 110 K a broad hyperfine field component appears around 25 T that shifts towards 28 T with decreasing temperature. It is most likely related to the freezing of spins at the interface or within the AFM. In the time spectra it manifests as an additional modulation that leads to a significant intensity enhancement between 20 and 30 ns.

For comparison, Fig. 4(a) shows temperature dependent magnetization curves recorded by vibrating-sample magnetometry (VSM) after the same sample was cooled in an external field B_{ext} of 200 mT. At 10 K, the most prominent features of the exchange bias are seen, namely, an increase of the coercive field H_c and a shift of the hysteresis loop. At 50 K, there is no exchange bias field while the coercivity remains larger than at RT. The evolution of H_c and H_e with temperature displayed in Fig. 4(b) clearly shows that the coercive field H_c starts to increase at a much higher tem-

FIG. 4 (color online). (a) Selected hysteresis curves recorded by VSM on a Fe/FeO(1.6 nm)/Fe trilayer. (b) Temperature dependence of the coercive field H_c and exchange bias field H_e extracted from the magnetization curves.

perature than the exchange bias field H_e , an effect reported to occur in ultrathin AFM layers [19]. Fernandez-Outon *et al.* have shown that spontaneous magnetization of interfacial spins in exchange bias systems can lead to a drastic increase of H_e [20], an effect which eventually depends on the amount of structural disorder in the system. In fact, the increase of H_e that we observe below 30 K is consistent with the evolution of the 28 T component in Fig. 2. The rapid broadening of this distribution for temperatures above 30 K is a sign that the corresponding atoms start to loose their fixed magnetic orientation. The fact that the distribution does not evolve much between 10 and 30 K indicates that we have reached the nominal value of the hyperfine field, i.e., that spins are not fluctuating anymore, leading to the rapidly rising exchange bias field. We conclude that the freezing of spins plays a crucial role for the appearance of exchange bias in this ultrathin disordered FeO layers grown by oxidation.

In conclusion, we have observed *in situ* the evolution of the magnetic state of a 1.6 nm thick antiferromagnetic FeO layer. We see that the presence of only one FM/AFM interface is not sufficient to stabilize the AFM ordering at RT while the deposition of a second ferromagnetic layer leads to a strong increase of T_N . This latter system then behaves as predicted by the mean field theory. The most important conclusion is that a free (ultrathin) AFM layer (even supported by a ferromagnet) does not experience an increase of its Néel temperature. While this cannot be directly generalized to the case of metallic AFM (such as those used in devices), it shows the importance the ferromagnetic surrounding plays in stabilizing AFM layers. Evidence for a similar effect in metallic systems would therefore bring insight into the mechanism that leads to the effect we observed in this oxide based antiferromagnet. We also see that the magnetization of interfacial spins plays a major role for the observation of the exchange bias effect in this ultrathin Fe/FeO systems. Additional studies on similar systems such as CoO/Co or NiO/Ni would allow us to eventually generalize this concept. Finally, these direct experimental observations show the importance of understanding the mutual influence of thin ferromagnets on thin antiferromagnets for the design of possible future magnetic systems incorporating ultrathin antiferromagnetic layers.

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