Perpendicular Antiferromagnetic Ordering of Mn and Exchange Anisotropy in Fe/Mn Multilayers

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Fe/Mn is a model system in which to study exchange bias, since the antiferromagnetic (AF) Mn layers are believed to have uncompensated moments with all spins aligned in the plane and parallel to those of the Fe. We have determined the microscopic AF ordering at the interfaces using single-crystal neutron diffraction. An unexpected magnetic structure is obtained, with out-of-plane Mn moments perpendicular to those of Fe. This explains the low bias field and shows that the simple AF ordering assumed in a variety of exchange-biased systems may well have to be revised.

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There has been intense interest in the exchange-bias phenomenon in thin films in recent years due to its importance in technological applications, such as read heads, sensors, and magnetic random-access memories. Bias fields arise from the interaction at the interface between ferromagnetic (FM) and antiferromagnetic (AF) components [1], but a detailed understanding of this phenomenon has remained elusive [2,3]. Significant advances have been made using synchrotron x rays relating uncompensated moments at the interfaces to exchange-bias fields [4,5]. Here we determine details of the AF magnetic order at the interfaces in Fe/Mn multilayers with atomic resolution using neutron diffraction.

In a scattering experiment, such as neutron diffraction, spatial information is retained, and it is possible to determine the microscopic nature of the magnetic ordering at buried interfaces. One of the obstacles in applying this method is the difficulty in measuring the AF order from a few atomic planes in the interfacial region using a signal-limited technique. We overcome the signal problem by studying multilayers with many bilayer repeats. Furthermore, if the magnetic structure is coherent over many bilayers, it is possible to determine how the AF structure varies across an interface by modeling the intensities of the magnetic diffraction harmonics.

The phase of Mn stable at room temperature, α-Mn, has an unusually complex crystal structure, and its magnetic structure has not been solved. Much simpler cubic crystal structures are obtained at elevated temperature, and these may be stabilized at low temperature using molecular beam epitaxy (MBE), allowing the magnetic ordering to be studied. Fe/body-centered tetragonal (bct) Mn multilayers have been grown by MBE and they exhibit oscillatory exchange coupling and giant magnetoresistance [6]. Theoretical studies for Fe/Mn multilayers predict either a simple AF structure or helical structure, with the Mn moments in the plane next to the FM Fe block forming a FM sheet in the same direction as the Fe [7,8]. Thus the interfaces between Fe and Mn are expected to constitute a model uncompensated exchange-biased system, see Fig. 1(a) for a typical spin-valve structure.

Single-crystal Fe/Mn multilayers were grown at ambient temperature onto UHV annealed MgO (001) substrates with a 50 Å Fe seed layer using MBE at the Clarendon Laboratory. Growth rates of 0.2 and 0.4 Å s^{-1} were employed for Mn and Fe, respectively. Samples were capped with 50 Å of niobium to prevent oxidation of the multilayer. Figure 1(b) shows a typical hysteresis loop measured for a $[\mathrm{Fe}(24\ \text{Å})/\mathrm{Mn}(12\ \text{Å})]_{100}$ multilayer, with a much smaller bias field than expected [9], $H_{\mathrm{EB}} = 16$ Oe.

The crystal structures were determined using x-ray diffraction at the XMaS UK-CRG at the ESRF. The bct phase is stabilized, and the multilayer structure was determined from the diffraction harmonics using the model of Jehan *et al.* [10] in which the individual lattice spacings and the roughness at the interfaces were determined. The one-electron contrast between the two constituents of the multilayers means that the anomalous dispersion can be used to

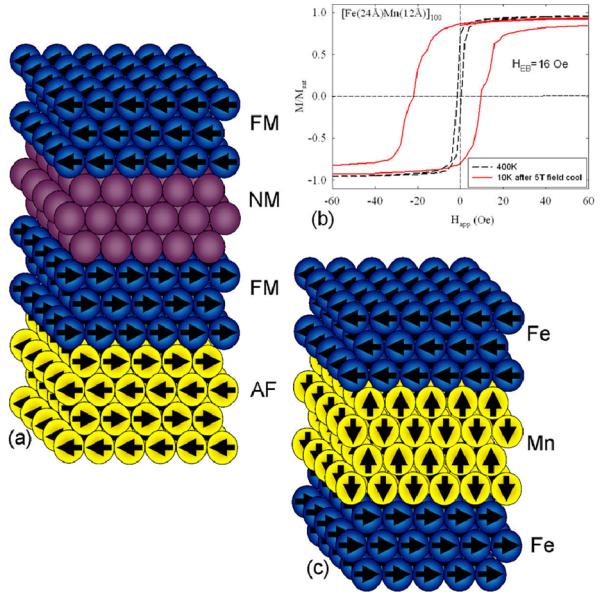

FIG. 1 (color online). (a) Schematic diagram of a spin valve with an exchange-biasing AF layer whose moments are uncompensated at the interface with the FM. An Fe/Mn spin valve would be expected to have the same moment direction for Fe and Mn ions at the interface. (b) Typical hysteresis loop for a $[\mathrm{Fe}(24\ \text{Å})/\mathrm{Mn}(12\ \text{Å})]_{100}$ multilayer after cooling to 10 K in field of $H = 5$ T. (c) The magnetic structure observed for $[\mathrm{Fe}(13.5\ \text{Å})/\mathrm{Mn}(5.4\ \text{Å})]_{300}$ using neutron diffraction.

vary the scattering contrast as a function of x-ray energy at the Fe and Mn K edges, allowing further constraint of the model. The fits to the high-angle x-ray scattering data are shown as solid lines in Fig. 2. For this sample we find a composition $[\mathrm{Fe}(13.5\ \text{Å})/\mathrm{Mn}(5.4\ \text{Å})]_{300}$, and the individual lattice parameters are $c_{\mathrm{Fe}} = 2.860(5)$ Å, $c_{\mathrm{Mn}} = 3.016(5)$ Å. The common in-plane lattice parameter is $a_{\mathrm{Fe}} = a_{\mathrm{Mn}} = 2.897(3)$ Å. Distortion of the ratio c/a for a bct lattice from 1 to $\sqrt{2}$ would lead to a change from bcc to fcc. We find a very small distortion, $c/a = 1.041(2)$, and the observed atomic volume 12.65(3) Å^3 is close to the normal value for bcc Mn. The structural roughness at the interface was found to be 1.4(5) monolayers.

The arrangement of the FM blocks of Fe was determined by polarized neutron reflectivity (PNR) using the CRISP time-of-flight reflectometer at the ISIS Facility. Figure 3 shows the data and calculated profiles for spin-up and spin-down neutrons for $[\mathrm{Fe}(50\ \text{Å})/\mathrm{Mn}(x)]$ multilayers as a function of thickness of the Mn layers, x. For $x = 6$ Å the coupling of the Fe blocks is AF, for $x = 8$ Å the correlations between successive blocks are FM, and for $x = 18$ Å a noncollinear magnetic structure is found with a coupling angle of 19° between Fe blocks. The AF peaks also appear in the spin-flip channel, and they decrease as expected upon application of a saturating magnetic field. The fact that the coupling changes from purely AF to purely FM with only a 2 Å change in Mn thickness suggests indirectly that the interfaces must be smooth and that the observed interfacial roughness arises from terracing.

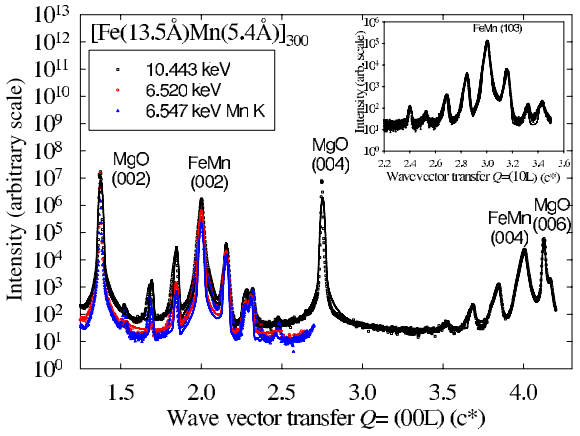

FIG. 2 (color online). The structural multilayer harmonics measured from $[\mathrm{Fe}(13.5\ \text{Å})/\mathrm{Mn}(5.4\ \text{Å})]_{300}$ using x rays at $T = 15$ K. Measurements on and off the absorption edge allow the cross section to be varied, and the measurements at higher energy allow access to more reflections.

The nature of the interfaces was studied more directly using off-specular PNR. Figure 4 shows the data for $x = 18$ Å acquired using a 1D detector on CRISP (a) with a guide field of 55 Oe and (b) a saturating field of 1620 Oe. Application of the saturating field transfers all of the magnetic intensity from the AF reflection at $Q_z = 0.047$ Å^{-1} to the FM Bragg peak at $Q_z = 0.095$ Å^{-1}. The faint arc stretching through the Bragg peak from the bottom left to top right is a generalized Yoneda effect in which the scattered neutron wave vector satisfies the Bragg condition [11]. Weak vertically correlated AF and structural in-plane correlations indicate that the magnetic roughness follows the structural roughness. The in-plane widths of the AF peak in the guide field, the FM peak in the saturating field, and the structural peak are all the same within experimental uncertainty, and the in-plane domain size for the Fe magnetism is found to be at least 14 μm.

The AF ordering in the Mn layers was studied using the D10 diffractometer at the ILL. Figure 5(a) shows the purely magnetic scattering measured at $T = 2$ K for a multilayer of composition $[\mathrm{Fe}(13.5\ \text{Å})/\mathrm{Mn}(5.4\ \text{Å})]_{300}$, in the vicinity of the structural absence at $Q = (100)$. Figure 5(b) shows the order parameter, and the ordering temperature for the Mn in this multilayer is found to be $T_N = 180(4)$ K. The coupling of the Fe blocks through the Mn layers does not seem to be affected by the onset of magnetic ordering in the Mn. There is a weak magnetic reflection at (111) but, as the inset to Fig. 5(a) shows, there is no magnetic scattering at all near (001). The neutron magnetic cross section is only sensitive to the component of the magnetic moment perpendicular to the scattering wave-vector transfer. Our results show conclusively that the Mn magnetic moments are ordered in a layered (001) AF structure, but with moments perpendicular to those of the Fe and out of the plane, see Fig. 1(c). This Mn magnetic structure was found for all compositions of Fe/Mn multi-

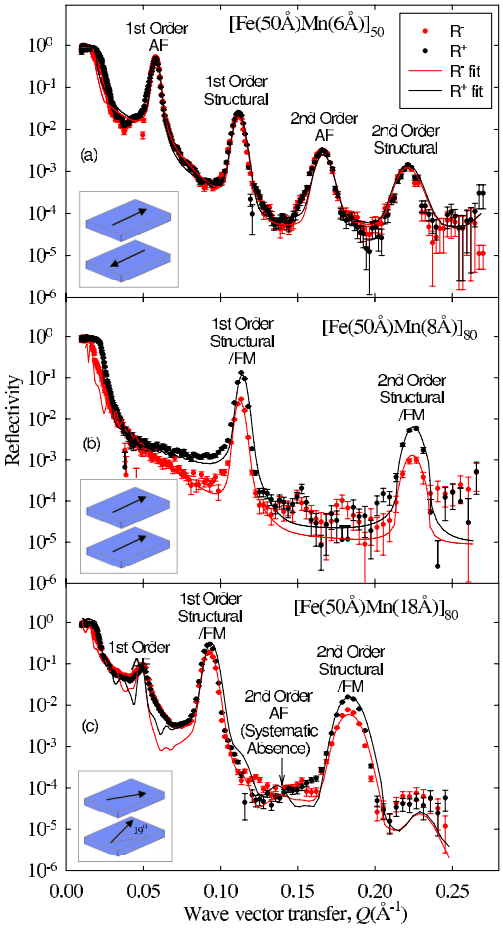

FIG. 3 (color online). The alignment of FM Fe blocks observed using specular PNR. For [Fe(50 Å)/Mn(x)] multilayers the magnetic ordering of the FM Fe blocks depends sensitively on the thickness of the Mn, x. Pure AF coupling is observed for $x = 6$ Å (a), FM exchange for $x = 8$ Å (b), and a noncollinear arrangement is found for $x = 18$ Å (c).

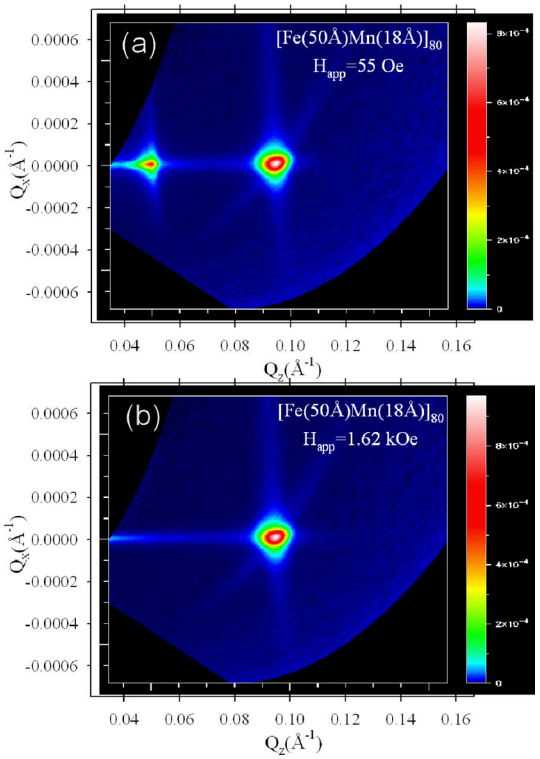

FIG. 4 (color online). The in-plane correlations at the interface studied using off-specular PNR. The panels show the data from the [Fe(50 Å)/Mn(18 Å)]$_{80}$ multilayer (a) in a guide field of 55 Oe and (b) in a saturating field of 1620 Oe. The AF and FM positions are $Q_z = 0.047$ and 0.095 Å^{-1}, respectively. A faint arc passing through the Bragg peak from a generalized Yoneda effect is also observed. The streaks of scattering through the Bragg reflections parallel to Q_x indicate vertically correlated magnetic and structural roughness.

layers studied, and the results readily explain why the bias fields are so low.

The sharpness of the magnetic peaks in scans of Q in the [10L] direction, together with the presence of magnetic multilayer harmonics in Fig. 5(a), clearly demonstrates that the Mn magnetic order propagates coherently through the Fe layers. This is one of the main reasons why it is possible to detect the AF Mn reflections at all. The magnetic multilayer peak intensities calculated using the structural parameters determined independently using x rays and the magnetic structure shown in Fig. 1(c) are in excellent agreement with the data, with the magnitude of the AF moment following the concentration of Mn in each layer. This shows that there is no additional magnetic roughness at the interfaces or modulation within the Mn layers. The individual moments per site determined by comparison with the structural scattering are 1.85(5)μ_B for the Fe and 3.0(3)μ_B for the Mn. There is no evidence for any in-plane magnetic reconstruction of the Mn moments next to the Fe.

The most recent *ab initio* calculations for bulk bct Mn predict an in-plane $c(2 \times 2)$ AF in the (001) planes with a transition to layered AF in (001) planes in an expanded state closer to fct Mn [12]. Around the equilibrium density, a FM phase is intermediate in energy between these phases, and earlier studies [13,14] had ignored the possibility of in-plane AF and, therefore, FM is often reported as the ground state for bct Mn in the literature. More complex ferrimagnetic [15] and helimagnetic [16] structures have also been found to be competitive in energy. Calculations of the effect of the tetragonal distortion give the in-plane $c(2 \times 2)$ AF, for bct, then a (110) layer-by-layer AF, and then a (001) layer-by-layer AF as the distortion tends towards fct [17]. We can rule out any of the structures predicted for bulk bct Mn. Our results are instead consistent with that predicted for fct Mn. Thus, the magnetic ordering in our nanostructures differs markedly from the bulk.

The theoretical [8] and experimental [18–29] situation for the magnetic structure and coupling of Mn overlayers on bcc Fe is controversial. An in-plane AF with two inequivalent Mn sites [18–20], a layered AF with AF coupling at the interface [21,22], FM coupling at the surface [23], and a FM Mn film coupled parallel to the

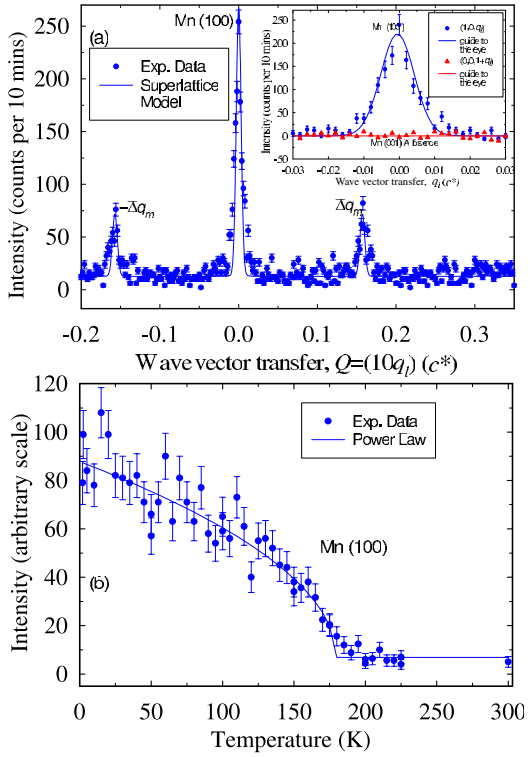

FIG. 5 (color online). Magnetic Bragg peaks at forbidden bct reflections. (a) There is a strong magnetic peak at (100) at low temperature, but no magnetic scattering at (001), as shown in the inset. The solid line through the magnetic reflection and its multilayer harmonics is calculated for the AF Mn structure shown in Fig. 1(c) with the structural parameters fixed at those determined using the x-ray data and a $3\mu_B$ moment for all Mn ions across the layer. (b) The Mn order parameter determined from the integrated intensity at $Q = (100)$. A fit of a power law gives $T_N = 180(4)$ K.

Fe for up to three monolayers, becoming AF at higher coverages [24,25], have been reported. The magnetic properties are very sensitive to coverage and surface contamination [26]. Recently, spin-polarized scanning tunneling microscopy suggests in-plane layered magnetism [27,28], and x-ray magnetic circular dichroism and linear dichroism spectroscopies reveal an in-plane AF with almost perpendicular coupling [29]. None of the magnetic structures found in Mn overlayers on bcc Fe are observed in our Fe/Mn multilayers.

The theoretical prediction for Fe/Mn multilayers is a layered AF in the Mn with in-plane moments and FM coupling at the interfaces with the Fe [7,8]. Helical phases can also satisfy the interfacial interaction, and these are also predicted to be stable for Fe/Mn. In fact, similar spiral and commensurate structures with in-plane moments are observed in Fe/Cr [30,31] multilayers. In-plane moments are also found for more complicated, fully compensated Fe_3O_4/CoO multilayers, and the fact that the AF CoO moments are perpendicular to the ferromagnetic component of Fe_3O_4 is attributed to a spin-flop transition [32]. An out-of-plane component in the AF spin structure was found for $Fe/FeSn_2$ bilayers [33]. However, the absence of any magnetic scattering along the $[00L]$ direction completely rules out scenarios with any in-plane Mn moments for our Fe/Mn multilayers. The observation of perpendicular moments in the Mn is consistent with the symmetry of the bct lattice and may arise from frustration of the interfacial interaction at terraces.

In summary, we have found a layered (001) AF structure in bct Mn stabilized in Fe/Mn multilayers with out-of-plane moments perpendicular to those of Fe. The results clearly demonstrate the importance of measuring the AF structures at buried interfaces in magnetic nanostructures using neutron diffraction.

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