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The influence of interfacial roughness on the coherence of structure and magnetic coupling across barriers in Fe/MgO multilayers

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Abstract

Single-crystal Fe/MgO multilayers are model systems in which to study magnetic tunnel junctions. We find that the interfacial roughness leads to the loss of coherence of the crystal structure. For thick MgO layers ferromagnetic (FM) ordering is found using polarized neutron reflectivity (PNR). For thin MgO layers magnetization measurements reveal the presence of antiferromagnetic (AF) interactions, but no long-range AF order is found using PNR. After cycling in a hysteresis loop, FM correlations are found at the coercive point, and this will limit the maximum tunnelling magnetoresistance.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

The tunnel magnetoresistance (TMR) effect in magnetic tunnel junctions [1, 2] is the key to developing magnetoresistive random-access memory (MRAM), magnetic sensors and novel programmable logic devices [3–5]. Giant room-temperature magnetoresistance ratios up to 180% have been observed in single-crystal Fe/MgO/Fe magnetic tunnel junctions [6]. The results demonstrate that the coherence of wavefunctions is conserved across the tunnel barrier, and this is of key importance in making spintronic devices with novel quantum-mechanical functions, and to developing gigabit-scale MRAM. Calculations of the TMR by Mathon and Umerski for epitaxial Fe/MgO/Fe predict a ratio in excess of 1000% [8]. However, the values measured in real systems will depend sensitively on the interfacial structures. The junctions prepared by Yuasa *et al* are believed to have sharp, unoxidized interfaces [6, 7]. In contrast, Meyerheim *et al* found an interfacial FeO layer [9, 10]. *Ab initio* calculations by Heiliger *et al* using these and other interface structures show that even the sign of the TMR ratio depends on the interface structure [11]. The TMR ratio is particularly sensitive to the crystallinity at the interfaces [12, 13].

Interlayer magnetic coupling has been observed for Fe/MgO/Fe using bulk magnetometry [14]. For thin barrier thicknesses AF coupling is observed, and the results agree with theoretical models using spin-polarized quantum tunnelling of electrons between ferromagnetic layers [15, 16]. For larger spacer layer thicknesses FM coupling is observed, and this is attributed to the 'Orange peel' interaction associated with correlated roughness of the FM/insulator interfaces [17].

Here we present results for Fe/MgO multilayers using the artificial periodicity to gain detailed information on the interfacial structure and coherence using synchrotron x-rays, and the magnetic ordering using PNR. The interfacial roughness is found to have a decisive impact on both the structural and magnetic coherence, and our results have important implications for the TMR.

2. Sample growth

Single-crystal Fe/MgO multilayers were grown using ultrahigh vacuum molecular beam epitaxy (MBE) at the Clarendon Laboratory. The pressure in the chamber was less than 3×10^{-8} Pa during growth. Prior to growth the MgO(001) substrates were degreased by boiling in bath of 1,1,1-trichloroethane at 50 °C for 15 min, then in a bath of isopropyl alcohol for 15 min, and finally in a bath of methanol for 15 min [18]. A 50 Å MgO buffer layer was grown at 500 °C. Multilayers of nominal composition $[\mathrm{Fe}(50\ \text{Å})/\mathrm{MgO}(6\ \text{Å})]_{20}$ and $[\mathrm{Fe}(50\ \text{Å})/\mathrm{MgO}(20\ \text{Å})]_{10}$ were grown at ambient temperature with growth rates of 0.3 Å s^{-1} and 0.1 Å s^{-1} for the Fe and MgO, respectively. The samples were then annealed at 300 °C for half an hour. Finally, the samples were capped with 100 Å of copper to prevent oxidation. *In situ* reflection high energy diffraction was performed to monitor growth. The epitaxial relationship was determined to be Fe[100](001) ∥ MgO[110](001).

3. Experimental procedure

High-angle x-ray diffraction and low-angle x-ray reflectivity measurements for structural analysis were performed using the I16 beamline at Diamond, with an incident x-ray energy of 10 keV. All measurements were taken at room temperature.

Transmission electron microscopy (TEM) was performed at the Department of Materials in Oxford University to examine the samples in a cross-sectional view. The contrast modes were high-resolution (HREM) in a JEOL 4000EX microscope and high angle annular dark field (HAADF) in a JEOL 3000F field-emission microscope operated in a scanning mode.

SQUID measurements were performed using a Quantum Design MPMS magnetometer. All measurements were taken at room temperature with the Fe[100] in-plane direction parallel to the applied field.

Polarized neutron reflectivity, PNR, was performed on both multilayers, using the D17 reflectometer at the ILL. The samples were mounted with the Fe[100] in-plane direction parallel to the direction of the magnetic field and at right angle to the incident neutron beam, following the orientation used for the SQUID measurements. A small guide field was kept constant throughout the experiment and always had a value of 0.002 T. On- and off-specular reflectivity data were taken at the same time using a 2D detector, and a ^{3}He analyser was used with incident neutron wavelength of 5.387 Å. Both non-spin-flip and spin-flip reflectivities were measured in each given scan.

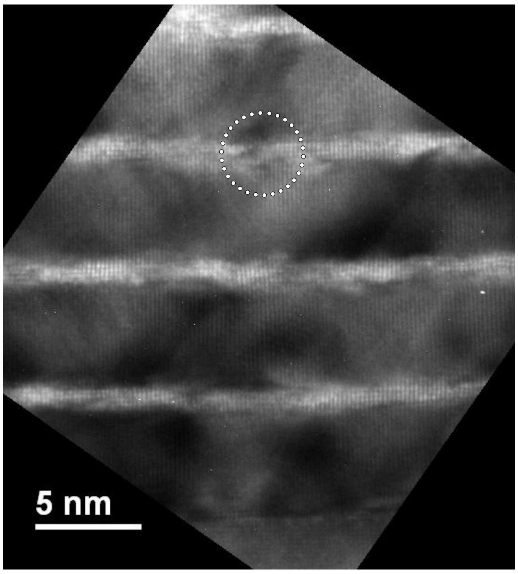

Figure 1. HREM image of the $[\mathrm{Fe}(50\ \text{Å})/\mathrm{MgO}(6\ \text{Å})]_{20}$ multilayer in cross-sectional view with zone axis Fe[100] ∥ MgO[110]. The interfaces are locally sharp and the wavy roughness varies over an in-plane length scale of a few nanometres. This leads to substantial variation in the barrier thickness (the circle highlights a pin hole).

4. Results and discussions

Figure 1 shows HREM data for the $[\mathrm{Fe}(50\ \text{Å})/\mathrm{MgO}(6\ \text{Å})]_{20}$ multilayer. These data and those obtained using scanning TEM show limited interdiffusion between layers. Previous measurements of Fe/MgO grown under these conditions show no evidence of oxidation of Fe at the interface [19]. The in-plane length scale for variations in the interfaces is over several nanometres. However, when coupled with the large out-of-plane roughness this leads to substantial variation in the barrier thickness. There is also evidence for some pin holing for the thin MgO barriers, see figure 1.

Further information on the multilayer structures was obtained from the low-angle x-ray reflectivity shown in figure 2(a) for $[\mathrm{Fe}(50\ \text{Å})/\mathrm{MgO}(6\ \text{Å})]_{20}$ determines the average bilayer composition. At low angles one is not sensitive to the crystallinity, and the SPEEDO programme by Knewtson and Suter [20] is used to model the electron density depth profile over the whole stack. The individual roughness parameters $\sigma_{\mathrm{Fe}} = 7.1$ Å and $\sigma_{\mathrm{MgO}} = 3.7$ Å are in approximate agreement with those estimated over much smaller regions on the sample using HREM.

The high-angle x-ray diffraction data were modelled in the kinematic regime, and this requires the microscopic ordering on an atomic scale. The fitted parameters are summarized in table 1. Figure 2(b) shows the x-ray diffraction intensity in the vicinity of the Fe(002) reflection for $[\mathrm{Fe}(50\ \text{Å})/\mathrm{MgO}(6\ \text{Å})]_{20}$. If the multilayer was coherent over all 20 bilayers the broad

Table 1. XRD fitted parameters.

	Lattice constant		Thickness		Structural coherence	Roughness	
Nominal composition	Fe ± 0.005 Å	MgO ± 0.005 Å	Fe ± 0.5 Å	MgO ± 0.5 Å	±1.0 Å	MgO/Fe ± 0.5 Å	Fe/MgO ± 0.5 Å
[Fe(50 Å)/MgO(6 Å)]$_{20}$	2.833	4.212	46.7	6.3	106	0.6	0.8
[Fe(50 Å)/MgO(20 Å)]$_{10}$	2.835	4.214	49.6	21.1	96	0.6	0.8

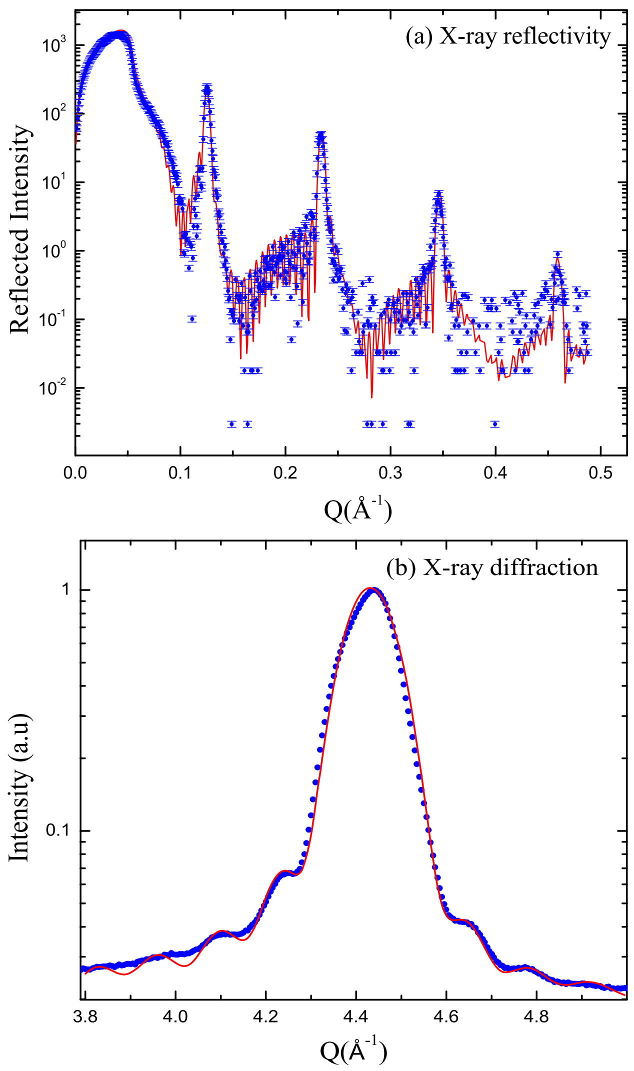

Figure 2. The average structure of the [Fe(50 Å)/MgO(6 Å)]$_{20}$ multilayer determined using synchrotron x-rays. (a) The low-angle x-ray reflectivity with a simulation over the whole multilayer stack, allowing the determination of the average bilayer composition. The interfacial roughness is in approximate agreement with the HREM. (b) The high-angle x-ray diffraction determines the coherence of the crystal structure. The sharp superlattice reflections expected for this multilayer are not detected. The simulation using just two bilayers reproduces the observed oscillations in intensity well.

central peak would be replaced by a sharp central peak with narrow superlattice peaks either side. Instead we observe a single broad peak here, with further broad oscillations further away from the main peak. The solid line in figure 2(b) shows the simulated scattering for two bilayers of the Fe/MgO, with the individual lattice spacings 1.416 and 2.106 Å close to those of bulk Fe and MgO in the centre of the blocks, and allowing the roughness and strain at the interfaces to vary according to the model of Jehan *et al* [21]. The excellent agreement between the model and the data shows that the crystal structure is only coherent over a couple of bilayers. The block thicknesses are consistent with the low-angle data. However, the fact that the roughnesses $\sigma_{\mathrm{Fe}} = 0.6$ Å and $\sigma_{\mathrm{MgO}} = 0.8$ Å are less than those obtained using low-angle x-rays presumably arises because the in-plane coherence of the crystallites is much smaller than the region sampled using reflectivity.

Figure 3(a) shows that the [Fe(50 Å)/MgO(6 Å)]$_{20}$ multilayer requires a field of 0.2 T to become fully saturated. This indicates the presence of AF interactions in agreement with bulk magnetization studies of tunnel junctions with this barrier thickness [14]. The jump in the hysteresis loop at very small applied field may also indicate a FM component. The ordering of the FM Fe blocks was investigated by PNR. Given the SQUID results in figure 3(a), the PNR results for the virgin state of [Fe(50 Å)/MgO(6 Å)]$_{20}$ with a guide field of 0.002 T in figure 3(b) are rather surprising. First, there are no indications of an AF peak at half the wavevector transfer of the structural peak. Secondly, there is no sign of FM correlations since there is no difference between the non-spin-flip reflectivities R^{++} and R^{--}. Finally, there is no correlated component perpendicular to the guide field, since the spin-flip reflectivities R^{+-} and R^{-+} are at background level. There are no long-range magnetic correlations between the Fe blocks at all, and the data are reproduced using the POLLY fitting programme by Langridge [22] using simply the structural model determined independently using x-ray reflectivity with the data in figure 2(a).

The PNR data in a saturating field for [Fe(50 Å)/MgO (6 Å)]$_{20}$ in figure 3(c) are fit with an Fe moment of 1.9 μ_{B} in the direction of the applied field and the same structural model. The sample was then saturated in the opposite direction, and then returned to its coercive state with a field of 0.009 T. Figure 3(d) shows that R^{++} and R^{--} are still different in the coercive state, and the fit to the data gave an ordered moment of 0.4 μ_{B}, indicating the presence of FM domains whose net moments cancel. From figure 3(a) we deduce that the observed moment cannot be explained by an offset in the applied field. According to the simulations, and comparison with the width of the FM Bragg peak in figure 3(c), the FM correlations extend over the whole stack in the coercive state.

The hysteresis loop for the [Fe(50 Å)/MgO(20 Å)]$_{10}$ multilayer shown in figure 4(a) indicates FM coupling, again in agreement with the results for tunnel junctions with thicker barrier layers [14]. The PNR data in the saturated state and the virgin state in a guide field of 0.002 T, figure 4(b), are both described with FM correlations and the structure determined

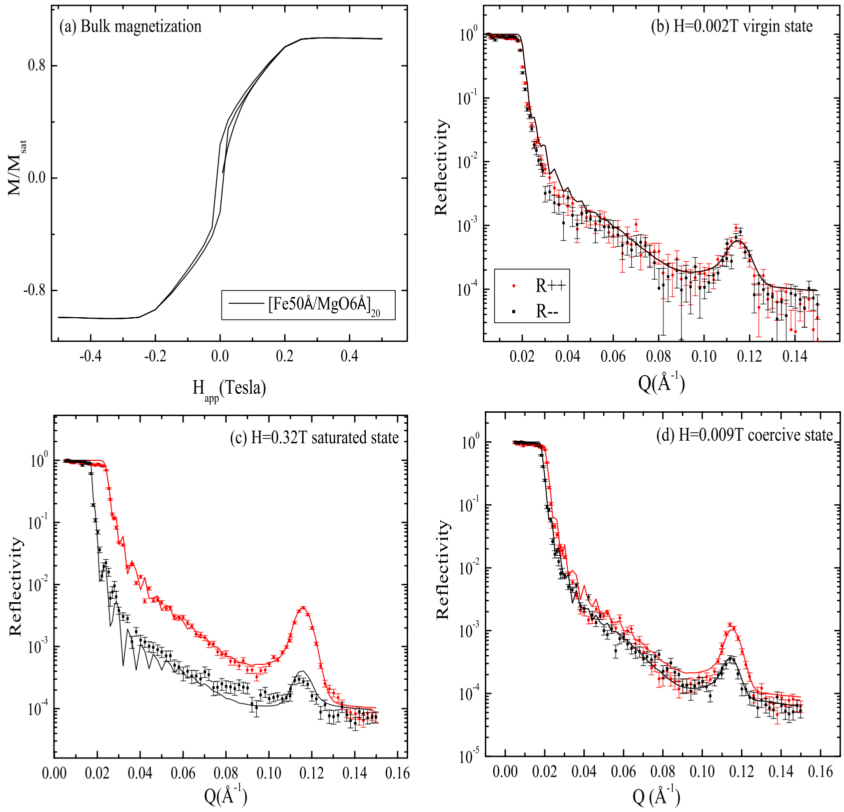

Figure 3. The magnetic properties of the [Fe(50 Å)/MgO(6 Å)]$_{20}$ multilayer at room temperature. (a) The bulk magnetization measurements with the saturation field of 0.2 T indicating the presence of AF interactions, and the jump at zero applied field suggesting a FM component. (b) In the virgin state with a guide field of 0.002 T there is no AF peak, no difference in the non-spin-flip PNR, and no signal in the spin-flip channels, showing that there are no long-range magnetic correlations. (c) In a saturating field the PNR data can be described using a model with all Fe moments pointing along the field direction and the structural parameters fixed at those determined separately using the x-ray data in figure 2(a). (d) At the coercive point in a field of 0.009 T the sample does not return to the virgin state. There remain substantial FM correlations, and this will ultimately limit the TMR.

separately using x-ray reflectivity. In this case the saturation moment for Fe is 2.2 μ_B. In the virgin state the moment components perpendicular to the guide field are comparable to those parallel to the field, giving rise to the observation of signal in the R^{+-} and R^{-+} channels, see figure 4(c). In all cases the coherence of the FM structure is across the whole multilayer stack.

The rather limited crystalline coherence can be understood in terms of the observed interlayer roughness, since it results in a thickness of the barrier layers that varies through the multilayer stack. In many multilayers the individual interplanar spacings of each component of the multilayer are rather similar, and this effect would not disrupt the structural coherence. However, the Fe and MgO have very different spacings between individual planes and, therefore, if successive blocks contain randomly varying numbers of planes the crystalline coherence is lost after only one or two bilayers.

The absence of magnetic coherence for the virgin state of [Fe(50 Å)/MgO(6 Å)]$_{20}$ cannot be attributed to this limited crystalline coherence, since the PNR measurements are in the low wavevector transfer regime and are not sensitive to the microscopic magnetic ordering on an atomic scale. In contrast the [Fe(50 Å)/MgO(20 Å)]$_{10}$ multilayer does exhibit long-range FM coherence in the virgin state. Furthermore, the absence of long-range magnetic order for [Fe(50 Å)/MgO(6 Å)]$_{20}$ cannot be explained by weak coupling since the magnetization measurements indicate a substantial AF interaction, and the sample with a much thicker barrier layer is coherent. A possible explanation is that the randomly varying thickness of the barrier layer arising from the interfacial roughness could lead to successive Fe blocks being AF and FM coupled. Barriers of 6 Å thickness would make successive Fe blocks AF coupled. However, pin holing for thinner barriers may lead to FM coupling, and blocks with thicker barrier layer are expected to be FM coupled. This would lead to static magnetic disorder.

These results have implications for the properties of the technologically important Fe/MgO/Fe tunnel junctions. The rather large interfacial roughness will certainly limit the maximum TMR ratio. The presence of FM interactions in both the virgin and coercive states for thin barrier layers will also limit the practically attainable TMR ratios compared to those possible for junctions with pure AF interactions.

5. Conclusions

In summary, we find that Fe/MgO multilayers interfaces are locally sharply, but with a wavy roughness that leads to

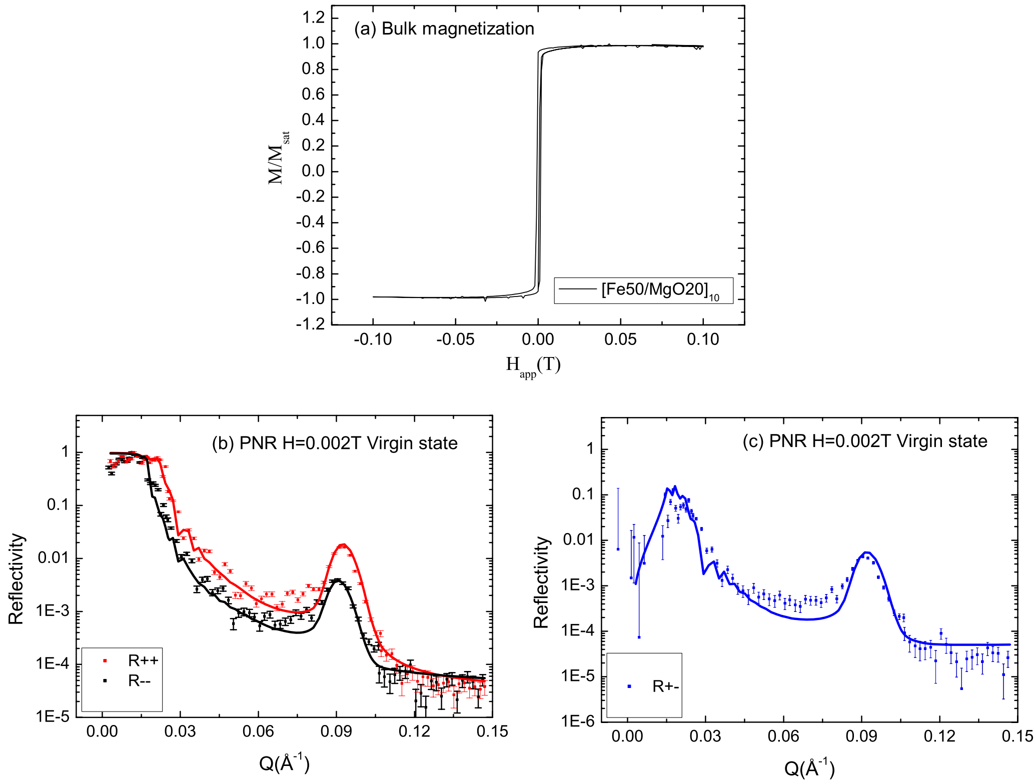

Figure 4. The magnetic properties of the [Fe(50 Å)/MgO(20 Å)]$_{10}$ multilayer at room temperature. (a) The bulk magnetization measurements suggest a purely FM response, and in the virgin state the (b) non-spin-flip and (c) spin-flip PNR data can be fitted using a model with FM correlations extending over the whole multilayer stack, but with in-plane domains with comparable components parallel and perpendicular to the guide field.

substantial variation in the thickness of the barrier layers, severely limiting the coherence of the crystalline structure and the magnetic order.

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