Positive antiphase boundary domain wall magnetoresistance in Fe$_3$O$_4$ (110) heteroepitaxial films

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(Received 20 July 2010; revised manuscript received 21 December 2010; published 27 April 2011)

We observe a strong crystallographic direction dependence on the low-field magnetoresistance (MR) behavior of the epitaxial Fe$_3$O$_4$ (110) films grown on MgO (110) substrates. The sign of MR is positive when the current and field are parallel to [001], whereas along the [110] direction its sign is negative, similarly to that commonly observed for (100) oriented Fe$_3$O$_4$ films. We relate this effect to the presence of antipath boundaries (APB) and subsequent reduction in the width of canted spin structure in its vicinity, due to the hard axis behavior of Fe$_3$O$_4$ (110) films along this crystallographic direction. At fields greater than the anisotropy field, usual negative MR behavior related to a reduction in spin scattering at the APBs is observed. An analytical model based on the half-infinite spin chains across the APBs is provided to show that the positive MR is due to the domain walls along APBs. The temperature and film thickness dependency of the APB domain wall magnetoresistance is discussed.

DOI: 10.1103/PhysRevB.83.134436 PACS number(s): 75.70.Kw, 75.30.Gw, 75.47.Lx, 68.55.–a

I. INTRODUCTION

Electronic properties of domain walls (DWs) in ferromagnetic materials may differ from those of ferromagnetic domains. A DW is an interface between uniformly magnetized regions (domains) with different magnetization directions. The length scale over which the magnetization direction changes is determined by material parameters such as the exchange and magnetic anisotropy energies. The electrical resistance due to magnetic domain walls in metallic ferromagnets has received considerable attention for a long time. Research activity in this area has increased substantially in the recent past due to the advances in material preparation and various nanofabrication techniques which allow one to control the DW dimensions and their number. The origin of the DW resistance (DWR) in ferromagnetic metals is attributed to the mixing of up- and down-spin electrons due to the spin mistracking of the electrons on passing through the DW. However, the models proposed for the ferromagnetic metals cannot be applied generally; for example, it is not sufficient to explain the DWR observed in itinerant ferromagnets such as SrRuO$_3$. In thin films the resistivity contribution of DW is usually masked by various intrinsic and extrinsic factors such as anisotropic magnetoresistance (AMR), ordinary magnetoresistance (OMR), spin scattering related to magnetic in-homogeneities, and so on.

In recent years a lot of attention has been given to the studies of the structural, magnetic, and magnetotransport properties of heteroepitaxial thin film structures based on magnetite (Fe$_3$O$_4$) thin films, which is a very good example of a strongly correlated half-metal with significant polaronic effects. The interest is partly due to the great application potential of this material in spintronics. Fe$_3$O$_4$ is one of the most important half-metallic oxides with the highest Curie temperature ($T_C = 858$ K). There are many reports on the growth of Fe$_3$O$_4$ thin films on different types of substrates like MgO, MgAl$_2$O$_4$, sapphire, Si, or GaAs using a variety of deposition techniques. MgO (lattice constant 0.4212 nm), which has a rock salt crystal structure, is a widely used substrate for epitaxy of magnetite (lattice constant 0.83987 nm) due to the small lattice mismatch of only 0.33%. Antipath boundaries (APB) are structural defects occurring in thin films during epitaxial growth and are observed in Fe$_3$O$_4$ films when grown on a variety of substrates like MgO, MgAl$_2$O$_4$, and α-Al$_2$O$_3$. Since the Fe$_3$O$_4$ (Fd$ar{3}$m) crystal structure is of lower symmetry than MgO (Fm$ar{3}$m), there are several equivalent nucleation sites on the MgO (100) surface, which enforce the formation of APBs at the interface of the neighboring grains. An atomic step in the MgO substrate can also lead to the formation of APB in Fe$_3$O$_4$ since the lattice constant of Fe$_3$O$_4$ is almost exactly double that of MgO. The presence of APBs leads to a large difference between the physical properties of epitaxial Fe$_3$O$_4$ films and those of the bulk such as larger electrical resistivity, magnetoresistance, and magnetization which does not saturate in high magnetic fields.

The APBs, which can be envisaged as the disruption of cation chains in a continuous oxygen lattice, lead to new magnetic exchange interactions, which are not present in the bulk material. There are many possible types of APBs, each characterized by its own type of atomic coordination and consequently its own type of exchange interactions. Exchange interactions occurring across some of the APBs must be antiferromagnetic (AF), and experimental evidence suggests that these AF-APBs dominate the magnetic and magnetotransport properties of Fe$_3$O$_4$ thin films. It was shown that if the spins on neighboring ions are not parallel but, instead, form an angle $\phi_{\text{nn}}$, the transfer integral is reduced to $t \cos \phi_{\text{nn}}/2$. Therefore, the transfer of the conduction electrons between ions with antiparallel spins at an AF-coupled APB is blocked and, according to the model, the conductivity reduces to zero. The conductivity becomes finite if the angle deviates from $\pi$ by the application of a magnetic field. This explains the observation of negative magnetoresistance in Fe$_3$O$_4$ thin films grown on MgO substrates. The resistivity contributions from AMR, domain wall magnetoresistance (DWMR), OMR, and so on, are expected to be negligible when compared to the much greater resistivity contribution from AF-APBs in Fe$_3$O$_4$ thin films. In the case of thin films, due to easy plane shape anisotropy, the spins far from APBs lie in plane and...
at the boundary, and the AF-coupled spins lie out of plane, forming a domain wall across APB which is distinct from the usual 180° walls due to the abruptness of the AF coupling at the boundary.32 Studies on 25-nm-thick Fe3O4 (100) film using dark-field transmission electron microscopy (TEM) and off-axis electron holography reveal that a large proportion of APBs have out-of-plane moments at the boundary which rotate to an in-plane orientation over a distance of approximately 15 to 20 nm.35 Apart from the misalignment of spins in the vicinity of APB due to strongly coupled AF spins across the boundary leading to contribution to MR, the magnetic domain walls formed along the APB (APB-DW) can also contribute to the resistivity. These APB-DW are expected to have widths smaller than conventional ones due to AF exchange at the APB. Thus it could make much greater contribution to DWMR compared to conventional DW. This contribution would be noticeable only at applied field values where the negative resistivity contribution due to the rotation of AF-coupled spins at the APB is small. It was also shown that the coarse magnetic domains are 5 to 14 times greater than the APB domains.35 Therefore, the density of the APB domain walls is much greater compared to that of conventional magnetic domain walls. In this paper, we present the results of MR studies on Fe3O4 (110) films with different thicknesses grown on MgO (110) substrates. These films show substantial in-plane magnetic anisotropy and anomalies in the characteristic behavior of MR when measured along the [001] direction. A positive longitudinal MR is observed at low fields when current and magnetic field are directed along the [001] hard magnetization axis and a typical negative MR curve is observed when measured along [1\bar{1}0]. With an analytical model of half-infinite spin chains, we show that the occurrence of this positive MR at low fields is due to APB-DW (DWMR_{APB}). To the best of our knowledge there are no reports on the crystallographic direction dependence on MR properties of Fe3O4 (110) films.

II. EXPERIMENTAL METHOD

The Fe3O4 thin films (thicknesses of 30, 60, and 90 nm) were grown on (110)-oriented MgO single crystal substrates (cut along the [110] direction within ±0.5°) using an oxygen plasma-assisted molecular beam epitaxy (MBE) system (DCA MBE M600) with a base pressure of 2 × 10−10 Torr. The substrates were annealed in situ at 600°C in 5 × 10−6 Torr oxygen for 2 h. Growth of the Fe3O4 films was carried out at a substrate temperature of 250°C from a pure metallic Fe source by means of electron beam evaporation and oxygen free radicals generated by the electron cyclotron resonance (ECR) plasma source. The plasma source was operated at a power of 80 W in an oxygen partial pressure of 1 × 10−5 Torr. Reflection high-energy electron diffraction (RHEED) (STAIB Instruments) was used to monitor the growth mode and growth rate. After the cleaning procedure, the RHEED pattern of the MgO (110) single crystalline substrate measured along the [001] and [1\bar{1}0] azimuths, respectively, showed vertical lattice rods and radial Kikuchi lines indicative of a well-ordered and flat surface. The RHEED pattern after the film growth showed half-order streaks and was accompanied by the oscillations in the intensity of specularly reflected beam. The period of oscillation corresponds to the growth rate 0.3 Å/s and confirms that the film growth occurs in a layer-by-layer mode. Auger electron spectroscopy (AES) studies and high-resolution TEM imagery (close to the atomic resolution) on Fe3O4 (100) [grown on MgO(100) substrates using same MBE setup and annealing conditions] showed that there is no signature of Mg or other impurities in these films or interfaces.11,12,24 X-ray reflectivity measurements on the films showed thickness of 30, 60, and 90 nm (±2 nm). The thickness values determined from reflectivity measurements are in good agreement with the thickness estimated from observed growth rate from RHEED intensity oscillations.

Magnetiization measurements were performed using an alternating gradient-field magnetometer (Micromag-3900, Princeton Measurements, USA) with a sensitivity of 10−8 emu. The magnetization versus field (M-H) loops were measured at room temperature by applying the magnetic field (maximum field of 1 T) in the film plane along the [001] direction and [1\bar{1}0] directions. The diamagnetic contribution from the MgO substrate was subtracted from the measured data by performing an M-H loop in the same field range of the MgO substrate of similar dimensions as that of thick film samples.

Structural characterization of the films was done using a multicrystal high-resolution x-ray diffractometer, HRXRD (Bede-D1, Bede, UK). The HRXRD (wavelength = 0.154056 nm corresponding to a Cu K-α line) in a double or triple axis configuration was performed to confirm the epitaxial relationship of the Fe3O4/MgO heteroepitaxy. The out-of-plane (a⊥) lattice parameter was determined from the analysis of a-2θ scans measured around the symmetric (220) diffraction planes common to the substrate and thin film. The in-plane lattice constant a∥ = d_{001}, was measured by performing asymmetric scans on {222} and {444} diffraction planes of the substrate and thin film. In the case of a tetragonally distorted epiplane (e.g., Fe3O4/MgO) the asymmetric diffraction plane is tilted with respect to the equivalent asymmetric diffraction plane in the substrate. This difference in interplanar angle is denoted by Δθ. Thus a convenient method for canceling out the effect of Δθ is to take measurements of both the (h k l) (gracing exit geometry, GE) and (-h k l) (gracing incidence geometry, GI) planes.1,12 This method is very effective for the precise determination of in-plane lattice parameter of thin films by removing any errors due to strain tilt. The estimated x-ray probing depth for 90% contribution of the measured intensity at GI angle of 3.969° (Fe3O4 {444} peak) is 834 nm, which is well above the thickness of the samples used in these studies.

For electrical resistivity and magnetoresistance measurements a standard dc four-probe technique was employed. The sample was mounted on a copper block fitted onto a cold finger of a closed cycle refrigerator. Temperature of the sample stage was monitored using a GaAlAs thermometer and controlled within ±0.05 K. For magnetotransport measurements, the cold finger was inserted into an electromagnet with a maximum field of 1 T (Bruker, Germany, Model:B-E10V). The magnetotransport results reported here were obtained by keeping the direction of magnetic field and current parallel to each other unless otherwise stated. The MR is defined as MR% = [(R(H)-R(0))/R(0)] × 100, where R(H) and R(0)
are the resistances of the sample with and without field, respectively.

III. RESULTS

Figure 1 shows the $\omega$-2$\theta$ scans (measured in triple-axis configuration at room temperature) for the (220) and (440) symmetric Bragg reflections of MgO and film, respectively, for all three samples. The horizontal axis in the figure is shown with reference to the Bragg angle of the (220) reflection for MgO substrate. The curves are shifted along the vertical axis for clarity. The full width at half of maximum (FWHM) of the film peak was found to decrease with increasing film thickness. The FWHM for the film peak was found to be 0.19°, 0.09°, and 0.06° for 30-, 60-, and 90-nm-thick films, respectively. The value of $a_\perp$ for the Fe$_3$O$_4$ thin films was determined from the separation of film-substrate peaks and was found to increase with increasing film thickness. From the analysis of grazing exit and grazing incidence asymmetric scans on {222/444} set of planes [shown in Figs. 2(a) and 2(b)], we obtained the in-nr lattice parameter of the films. The out-of-plane and in-plane lattice constants are shown in the Table I. The in-plane lattice constant along [001] was found to be decreasing and approaching toward the bulk value for 60- and 90-nm films. The detailed structural characterization shows that the films undergo strain relaxation above 30-nm film thickness and the amount of strain reduces (but not canceled) with increasing film thickness with increasing thickness. The observed strain is 0.345, 0.342, and 0.335% for 30-, 60-, and 90-nm films respectively. Figure 3 shows resistivity as a function temperature for various thicknesses. The resistivity of the films is found to be thickness dependent. The magnitude of resistivity increases with a decrease of film thickness. The onset value of Verwey transition temperature ($T_V$) for these films was found to be thickness dependent (108 K, 109 K, and 110.6 K ($\pm0.1$ K) for 30-, 60-, and 90-nm films, respectively). Since the Verwey transition is very sensitive to the Fe$_3$O$_4$ film stoichiometry, the presence of the Verwey transition confirms the high quality of the films. The lower value of $T_V$ in thin films compared to bulk ($\sim125$ K) could be to the reticence of long-range order of the Fe$^{2+}$ and Fe$^{3+}$ ions at the octahedral sites, which is necessary for the Verwey transition, caused by small APB domain sizes. These findings for the thickness-dependent behavior of resistivity and Verwey transition temperature are consistent with earlier transport studies on Fe$_3$O$_4$/MgO (100) heteroepitaxial system.

**TABLE I.** Out-of-plane and in-plane lattice constant of films with various thicknesses.

<table>
<thead>
<tr>
<th>Thickness (nm)</th>
<th>$a_\perp$ Å ($\pm10^{-4}$ Å)</th>
<th>$a_\parallel$ Å ($\pm10^{-4}$ Å)</th>
</tr>
</thead>
<tbody>
<tr>
<td>30</td>
<td>8.3833</td>
<td>8.4223</td>
</tr>
<tr>
<td>60</td>
<td>8.3943</td>
<td>8.4258</td>
</tr>
<tr>
<td>90</td>
<td>8.3952</td>
<td>8.4252</td>
</tr>
</tbody>
</table>
FIG. 3. (Color online) Resistivity of films with thicknesses 30, 60, and 90 nm as a function of temperature.

Figures 4 show hysteresis loops obtained at room temperature for a 90-nm film measured at 300 K with an in-plane magnetic field aligned in [110] and [001] directions respectively (magnetization is normalized with saturation magnetization, $M_s$). The hysteresis loops obtained at 300 K in [110] is nearly square ($M_r/M_s = 0.86$) and in the [001] direction it shows hard axis behavior. All our films showed a similar behavior. Although bulk magnetite has a cubic anisotropy with a ⟨111⟩ easy axis, reports on magnetization studies of Fe$_3$O$_4$ (110) epitaxial films grown on MgO (110) substrates show a square hysteresis loop with an in-plane [110] easy axis and [001] hard axis, consistent with our measurements. Low-temperature magnetization measurements show a clear transition from hard axis to easy axis behavior below the Verwey transition when measured along [001] direction. It is well known that certain crystallographic directions can be made magnetically soft below transition by cooling the crystal through the transition with a magnetic field applied in these directions. A discussion pertaining to this issue is outside the scope of this article.

The magnetization curves can be analyzed using a simplified equation, $M(\mu_0 H)/M(\infty) = 1 - qH^{-0.5}$, described in Ref. 34. The parameter $q$ measures the difficulty of approaching saturation. The virgin magnetization curves [normalized by $M(\mu_0 H = 1 \, \text{T})$] measured along [001] at room temperature were fitted using the above equation and the values of $q$ obtained for 30-, 60-, and 90-nm films were 0.22, 0.16, and $0.097 \pm 10^{-4} T^{0.5}$, respectively. Corresponding values obtained along [110] were $0.056, 0.047$, and $0.025 \pm 10^{-4} T^{0.5}$ which are an order of magnitude less compared to the [001] direction. However, the values of the exponents for a power law fit to the thickness dependency of $q$ were $-0.71$ and $-0.7 \pm 10^{-2}$ along [001] and [110], respectively. The above analysis and the observation of similar values of the exponents along the two directions suggests that the parameter $b$ depends on APB domain size (which is thickness dependent) and the size evolution of APB domains are isotropic. The values of $q$ (measured along [001]) are of the same order but higher compared to the values reported for Fe$_3$O$_4$ (111) and Fe$_3$O$_4$ (100) films of comparable thicknesses grown on $\alpha$-Al$_2$O$_3$ and MgO substrates respectively. This could be because of the differences in the APB network geometry, the fraction of APBs exhibiting AF couplings, and the difference in the APB density.

Field dependency of MR shows drastically different features when measured along [001] direction as compared to [110] direction. Figure 5 shows the MR at different temperatures for 90-nm film with field and current directed along the [001] direction. Above $T_V$, films showed symmetric butterfly-shaped positive MR at low field while MR along [110] direction shows negative MR characteristics similar to the MR (with in-plane field) measured on (100) Fe$_3$O$_4$ films (see Fig. 5). At high fields MR along [001] was found to be negative and it increases with decrease in temperature for all the films and peaks near $T_V$. Figure 6 shows the MR values in a 0.9-T field as a function of temperature for all three samples. Below $T_V$ the magnitude of high-field MR...
films having different thicknesses. Measured along the \([001]\) direction as a function of temperature for different domain areas are marked as I, II, and III. Different temperature and current directed along the \([001]\) direction. Different temperature for films having different thickness at 0.9-T field. Magnetic field the Lorentz force on the carriers is generally called OMR. MR which arises in a two-band model from the action of consider various known sources of MR. A classical positive value of positive MR as a function of temperature for various difference at \(T_p\) (please see the Appendix for details). Our model takes into account an in-plane anisotropy energy density term that manifests through modification in spin transfer behavior along different crystallographic directions. All the previous reports focus on the scattering of spin polarized electrons at the AF APBs. The significant difference in our analysis is that we discuss the scenario where a positive resistivity contribution occurs due to the tapered spin chains near the boundary in addition to the scattering due to AF spins at the boundary. Prior to discussing the details of the model, we would like to mention that there is a significant difference in the strength of magnetocrystalline anisotropy between the Fe\(_3\)O\(_4\)(100) films and the Fe\(_3\)O\(_4\)(110) films, its strength being greater for the latter case. Let us consider a half-infinite spin chain on one side of the APB. In the direct vicinity of APB, the spins are misaligned due to frustration of exchange interaction at the boundary. At some distance \(\lambda\) away from APB, the effect of the exchange frustration vanishes and the spins are almost aligned either along the external field \(H\) or along the easy axis of magnetocrystalline anisotropy. Whether it is aligned along \(H\) or the easy axis depends on the direction of the field and its strength. For example, if the field is aligned along the hard axis, then at a low-field value, the spins are aligned approximately along the easy axis and at high field \((H > H_\delta)\) they are aligned along the field. We can consider two cases here. A spin chain with field applied along [001] direction (case a) and along [T10] direction (case b). In case a we can write the energy per unit area of a half-chain as

\[
\gamma(\phi, H) = \int_{-\lambda}^{\lambda} M, H(1 - \cos\varphi) + K \left( \frac{1}{4} \sin^4\varphi + \sin^2\varphi\cos^2\varphi \right) + A_F \left( \frac{d\varphi}{dx} \right)^2 dx,
\]

where \(\varphi\) is the angle between local magnetization and field \(H\) and \(\lambda\) is the distance from the APB at which spins are

FIG. 6. (Color online) Magnetoresistance as a function of temperature for films having different thickness at 0.9-T field. Magnetic field and current directed along the [001] direction. Different temperature domain areas are marked as I, II, and III.

IV. DISCUSSION

In order to understand the observed positive MR, we consider various known sources of MR. A classical positive MR which arises in a two-band model from the action of the Lorentz force on the carriers is generally called OMR. The Lorentz force–related contribution can be observed if \(\omega_L \tau > 0.1\), where \(\omega_L\) is the cyclotron frequency, \(\frac{eB}{m}\), and \(\tau\) is the scattering time. For magnetite films it was shown that \(\omega_L \tau \sim 10^{-5}\) and, therefore, Lorentz force effects can be ruled out. Another source of the positive MR could be related to the anisotropic magnetoresistance (AMR), which is caused by the spin-orbit interaction. AMR is defined as \(\rho_j = \frac{\rho_j - \rho_{\perp}}{\rho_{\parallel}}\), where \(\rho_{\parallel} = \frac{\rho_0}{\parallel} + \frac{2\Delta\rho_0}{\parallel}\), \(\rho_0\) and \(\rho_{\perp}\) denote the longitudinal \((M || j)\) and transverse \((M \perp j)\) resistivities, respectively. In the case of magnetite films these resistivities decreases with field and provide only a negative contribution to MR.

To explain the negative MR observed in Fe\(_3\)O\(_4\)(100) films grown on MgO(100) substrates, Eerenstein et al. have used a model which considers two spin chains separated by an AF-APB. Ramos et al. have used an extended version of the model given by Eerenstein et al. to validate the longitudinal MR data of Fe\(_3\)O\(_4\)(111) films grown on \(\alpha\)-Al\(_2\)O\(_3\) substrate, observed in the high-field regime. To understand and explain the spin transport in Fe\(_3\)O\(_4\)(110) films, in particular the positive MR along the [001] direction, we consider a model similar to Ref. as the APBs are expected to form in the case of Fe\(_3\)O\(_4\) films grown on MgO(100) substrates, Eerenstein et al. have used an extended version of the model given by Eerenstein et al. to validate the longitudinal MR data of Fe\(_3\)O\(_4\)(111) films grown on \(\alpha\)-Al\(_2\)O\(_3\) substrate, observed in the high-field regime. To understand and explain the spin transport in Fe\(_3\)O\(_4\)(110) films, in particular the positive MR along the [001] direction, we consider a model similar to Ref. as the APBs are expected to form in the case of Fe\(_3\)O\(_4\) films grown on MgO(100) substrates, Eerenstein et al. have used an extended version of the model given by Eerenstein et al. to validate the longitudinal MR data of Fe\(_3\)O\(_4\)(111) films grown on \(\alpha\)-Al\(_2\)O\(_3\) substrate, observed in the high-field regime. To understand and explain the spin transport in Fe\(_3\)O\(_4\)(110) films, in particular the positive MR along the [001] direction, we consider a model similar to Ref. as the APBs are expected to form in the case of Fe\(_3\)O\(_4\) films grown on MgO(100) substrates, Eerenstein et al. have used an extended version of the model given by Eerenstein et al. to validate the longitudinal MR data of Fe\(_3\)O\(_4\)(111) films grown on \(\alpha\)-Al\(_2\)O\(_3\) substrate, observed in the high-field regime. To understand and explain the spin transport in Fe\(_3\)O\(_4\)(110) films, in particular the positive MR along the [001] direction, we consider a model similar to Ref. as the APBs are expected to form in the case of Fe\(_3\)O\(_4\) films grown on MgO(100) substrates, Eerenstein et al. have used an extended version of the model given by Eerenstein et al. to validate the longitudinal MR data of Fe\(_3\)O\(_4\)(111) films grown on \(\alpha\)-Al\(_2\)O\(_3\) substrate, observed in the high-field regime.

FIG. 7. (Color online) DWMR\(_{APB}\) of epitaxial Fe\(_3\)O\(_4\) films measured along the [001] direction as a function of temperature for films having different thicknesses.
aligned either along the field or the easy axis. The first term in the integral is the Zeeman and the second term is the cubic anisotropy energy density. The third term is the exchange energy density. \( K \) is the magnetocrystalline anisotropy constant and \( A_F \) is the exchange stiffness constant. The shape anisotropy of demagnetizing field is not relevant here, as in both cases the field is directed in the film plane. With variational calculus the condition for minimum energy can be derived as

\[
M_s H \sin \phi + K \cos \phi \sin \psi (3\cos^2 \phi - 1) = -2A_F \left( \frac{d^2 \psi}{dx^2} \right),
\]

(2)

which states that torque is zero everywhere along the spin chain. When the field is zero, the spins far from the APB will be aligned in the easy axis direction and with increasing field they will gradually rotate toward the direction of the field. Now we can further consider two situations here, where the applied field is above and below the magnetocrystalline anisotropy field \( H_A \). Above \( H_A \), the spins far from the APB will be aligned with the in-plane field while AF-coupled spin near APB will make an angle \( \phi_o \) with the field [see Figs. 8(a) and 8(c)]. Below \( H_A \) spins far from the APB orient approximately in the easy axis directions (\([\overline{1}10]\)) and spin near APB makes an angle \( \phi_o \) with the field [see Fig. 8(b)].

With these conditions, from Eq. (2) we can show that, for the case of \( H \) parallel to [001], when \( H < H_A \)

\[
\sqrt{A_F} \left( \frac{d\psi}{dx} \right) = \left[ -M_s H \cos \phi_o - \frac{K}{32}(1 + 4\cos^2 \phi_o + 3\cos\psi \phi_o) \right]^{\frac{1}{2}},
\]

(3)

\[
\frac{d\psi}{d\phi} = 2\sqrt{A_F} \left[ -M_s H \cos \phi_o - \frac{K}{32}(24\cos^4 \phi_o - 16\cos^2 \phi_o) \right]^{\frac{1}{2}}.
\]

(4)

and when \( H > H_A \)

\[
\sqrt{A_F} \left( \frac{d\psi}{dx} \right) = \left[ M_s H(1 - \cos \phi_o) + \frac{K}{32}(7 - 4\cos 2\phi_o - 3\cos 4\phi_o) \right]^{\frac{1}{2}}
\]

\[
\frac{d\psi}{d\phi} = 2\sqrt{K A_F} \left[ \frac{M_s H}{K}(1 - \cos \phi_o) \right]^{\frac{1}{2}} + \frac{1}{32}(7 - 4\cos 2\phi_o - 3\cos 4\phi_o) \right]^{\frac{1}{2}}.
\]

(5)

Now considering the spin chains on both sides of the APB with an angle \( \phi_o = \phi_1 \) and \( \phi_o = \phi_2 \) for the left and right side chains, the energy density of antiferromagnetic coupling at the boundary can be written as

\[
\gamma_{AF-APB} = \frac{A_{AF}}{d}(1 - \cos(\phi_2 - \phi_1)),
\]

(7)

where \( A_{AF} \) is the negative exchange stiffness constant for the AF exchange interaction at the boundary and \( d \) is the distance between the neighboring chains along the boundary. The final approximate result of minimization is \( \phi_1 = -\phi_2 = \frac{\pi}{2} = \phi_{AF} \) and

\[
\cos^2 \phi_{AF} = \frac{(M_s H)^2}{K(W - 2\sqrt{2K W + K})} \quad H < H_A,
\]

(8)

\[
\cos^2 \phi_{AF} = \frac{(4M_s H + K)}{(4W - 12K)} \quad H > H_A,
\]

(9)

where \( W = A_{AF}^2 / A_F d^2 \).

The dc conductivity in Fe3O4 can be explained by the superposition of a small polaron band and hopping conduction. It was shown that in the nonadiabatic limit the conductivity \( \sigma \propto t^2 \), where the transfer integral \( t \) can be expressed as \( t = t_0 \cos^2 \phi_{AF} \). Eerenstein et al. have shown that MR = \( -\cos^2 \phi_{AF} C \), where \( C \) is a constant which depends on the volume fraction of the APB domains. From Eq. (8), it is evident that for a field below the anisotropy field, \( H_A \), the conductivity across the antiferromagnetically coupled APB will decrease quadratically (small change) with the field and is linear (large change) above \( H_A \). Therefore in the low-field regime the MR contribution from other factors should dominate over the MR contribution due to AF-APBs.

Similar analysis for case b, with the field along the easy axis \([\overline{1}10]\) direction and an anisotropy energy density \( \gamma_{\sigma} = K[\frac{1}{2}\cos^2 2\phi] \), gives

\[
\cos^2 \phi_{AF} = \left[ \frac{M_s H}{W + K} \right],
\]

(10)

which suggests that the field dependence of MR is linear only. DWMR predicted by the Levy and Zhang (LZ) model for ferromagnetic metals is given by \( \frac{\Delta \rho}{\rho} = \frac{\xi}{m^2} \frac{F(\alpha)}{\rho} \), where \( \xi \) is the spin misstarking parameter and is given by \( \xi = \frac{\pi}{2} \hbar^2 k_F / 4 m w J \), where \( k_F \) is the Fermi wave vector, \( m \) is the effective mass, \( J \) the Stoner exchange splitting, and \( w \) is the domain wall width. \( \alpha \) is the spin asymmetry of the current. For our case, we can take the essential predication of this model, which is DWMR \( \propto \omega^{-2} \).
For the case of \( \varphi_0 = \pi \), from Eq. (3) we can deduce \( \lambda \propto 1/\sqrt{H} \), which means the width of the canted spin chain \( \lambda \) reduces with increasing field. The same dependency of \( \lambda \) on \( H \) can be derived in the case where \( H > H_A \) and in case b. But the essential difference between these two cases and the case where \( H < H_A \) is that, as \( \lambda \) decreases with increasing field, the angle between the spins (within the distance \( \lambda \)) decreases in the former case while it increases in the latter case. This is due to the differences in the rotation of AF-coupled APB spins with the field as explained earlier. Therefore the formation of highly canted spin structure below \( H_A \) should contribute some resistance to the traversing spins. Considering the model of DWMR we can write \( \text{DWMR}_{\text{APB}} \propto \beta H \), where \( \beta \) is a constant. This additional positive MR is visible because of the low negative MR contribution from APBs at low-field values (\( H < H_A \)). Figure 9 shows the variation of \( \beta \), which is extracted from the linear fit to the positive region of the MR curves of various film thicknesses with temperature. \( \beta \) follows the same trend of DWMR\(_{\text{APB}}\) shown in Fig. 7 which peaks at temperature \( T_p \). In the case of negative MR as a function of temperature, we can identify three different regimes as shown in Fig. 6 (regimes I, II, and III are marked only for the 90-nm film for clarity). There is a clear change in the slope from regime I to regime II. These regimes are found to be thickness dependent and \( T_p \) observed in the case of DWMR\(_{\text{APB}}\) matches with the center of regime II. The difference between the spin-dependent transport mechanisms in these regions could be related to the peaking of DWMR\(_{\text{APB}}\) at \( T_p \). Below \( T_V \) DWMR\(_{\text{APB}}\) vanishes. This is related to the hard axis to easy axis transition below \( T_V \) in the [001] direction. This will lead to a situation similar to the analysis of MR behavior along the [110] direction, where MR is dominated by the APB contribution only.

DWMR\(_{\text{APB}}\) is found to increase with the film thickness. It was shown that the APB domain size \( D \) is proportional to \( \sqrt{t} \), where \( t \) is the film thickness. Micro magnetic simulations reported by Bataille et al. show that APBs can no longer be considered independent when the distance between them is less than 40 nm and the blocking field \( H_B \) increases as film thickness.
thickness decreases. 34 This is due to the reason that, when two AF-APBs come closer the spins within the boundary can be pinned, which allows for less rotational freedom compared to spins in larger domains. This could be the reason for the dependency of DWMR_{APB} on film thickness. The field corresponding to maximum value of DWMR_{APB} is found to be 0.26 T at room temperature (see Fig. 5). This value is almost near but smaller than the $H_A (\sim 0.36 \text{T})$ measured through magnetization measurements (see Fig. 4). This suggests that decoupling of the AF spins at the boundary starts at a field value lower than the anisotropy field and therefore negative MR starts to dominate. Eerenstein et al. have used a similar model to predict quadratic field dependence for the low-field MR, when field is applied perpendicular to the current and film plane of Fe$_3$O$_4$(100) films. 32,45 It is not possible to have a direct comparison of our model and results with theirs due to the following reasons. Fe$_3$O$_4$(100) epitaxial thin film has small in-plane anisotropy and therefore the modeling is done by considering only the uniaxial shape anisotropy. 32,45 In the Fe$_3$O$_4$ (110) thin film case there exists a strong in-plane anisotropy on top of the shape anisotropy. This suggests that the rotations of the spin chains in the applied field differ in these two cases. Another reason is the difference in the way Fe-O-Fe ions meet at the APBs in both these cases due to symmetry differences. For example, one such difference is depicted in Figs. 10(b) and 10(c). Here APB formation along the [-110] direction on (110)- and (100)-oriented MgO surfaces are compared. 10(b) relates to the (110) surface and shows a perfect 180° AF super exchange while Fig. 10(c) relates to the (100) surface and shows superposition of 180° AF and 90° ferromagnetic superexchanges. This superposition of two opposite types of exchanges in the latter case could reduce the AF coupling strength leading to the domination of negative MR and hindering the observation of positive DWMR_{APB}. The MR measurement configuration used by Eerenstein et al. differs from ours. What we measure is longitudinal magnetoresistance (where the current $J$ and field $H$ are parallel to each other) while their measurement is for transversal MR (where the current $J$ and field $H$ are perpendicular to $H$), which again makes it difficult for a direct comparison. From the above analysis it is evident that positive MR observed is due to the DWMR_{APB}. Since the formation of APBs is not confined to a specific system like Fe$_3$O$_4$/MgO heteroepitaxy, the findings could be applied to other systems as well.

V. CONCLUSIONS

In conclusion we have performed detailed studies of structural and magnetotransport properties of Fe$_3$O$_4$ (110) films grown on MgO (110) substrate. Fe$_3$O$_4$(110) films show a significant in-plane magnetocrystalline anisotropy which is not present in Fe$_3$O$_4$(100) films grown on MgO (100) substrates. We found characteristic differences in the nature of MR between the longitudinal MR along [\langle110\rangle] and [\langle001\rangle] directions. Longitudinal MR along [\langle001\rangle] is positive at fields below the magnetocrystalline anisotropy field. With an analytical model based on the half-infinite spin chains across APBs, we have shown that this positive MR at low fields is due to the reduction in the width of the canted spin structure across APBs and associated increase in the spin canting. This DWMR_{APB} peaks at a particular temperature $T_p$ and vanishes below $T_F$. We associate this with the difference between the spin-dependent transport mechanisms and hard-to-easy axis transition across $T_F$. We have also shown that DWMR_{APB} depends on the size of the APB domains.

ACKNOWLEDGMENTS

This work was supported by the Science Foundation of Ireland (SFI) under contract no. 00/PI.1/C042.

APPENDIX: FORMATION OF APBs IN Fe$_3$O$_4$(110) FILMS GROWN ON MgO(110) SUBSTRATES

During the early stages of growth, adjacent Fe$_3$O$_4$ nucleation islands may be shifted by a $\sqrt{3}2a_0\{\langle111\rangle\}$ or $\sqrt{3}2a_0\{\langle\bar{1}1\bar{1}\rangle\}$ and a 1/2 a_0 (001) shift vector. The islands separated by these shift vectors can eventually form APBs when they coalesce. Figures 10(a) and 10(b) show the possible APB formation with the boundary along (001) and (110) directions. The direction of the boundary could be along other directions as well, not necessarily along (001) and (110). However, it is clear that shift vectors $\sqrt{3}2a_0\{\langle111\rangle\}$ or $\sqrt{3}2a_0\{\langle\bar{1}1\bar{1}\rangle\}$ lead predominantly to the formation of APB along low index direction (001) and likewise the shift vector 1/2 a_0 (001) leads prominently to APB along a low index direction (110). Figures 10(b) and 10(c) show the possible antiferromagnetic and ferromagnetic superexchanges (SE) across APBs formed on the (110) and (100) surfaces.

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