Spin-Polarized Transport across Sharp Antiferromagnetic Boundaries

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We report spin-polarized transport experiments across antiphase domain boundaries which act as atomically sharp magnetic interfaces. The antiphase boundaries are prepared by growing Fe_3O_4 epitaxially on MgO, the magnetic coupling over a large fraction of these boundaries being antiferromagnetic. Magnetoresistance measurements yield linear and quadratic field dependence up to the anisotropy field for fields applied parallel and perpendicular to the film plane, respectively. This behavior can be explained by a hopping model in which spin-polarized electrons traverse an antiferromagnetic interface between two ferromagnetic chains.

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Spin-polarized transport phenomena have received increasing attention over the past several years. A prominent example is the large magnetoresistance (MR) effects observed in magnetic multilayers [1] used in magnetic field sensors such as read heads [2]. Despite its technological importance, the MR mechanism in these so-called spin valves is not well understood, because it is difficult to discriminate between the many factors that influence the MR, such as band structure, magnetic impurities, structural disorder, and interface roughness [3,4]. In this Letter we introduce a new and simpler type of spin-valve system. It consists of two ferromagnetic domains coupled antiferromagnetically at an atomically sharp interface. In the case of spin-polarized conduction electrons and assuming that spin-dependent scattering does not occur at the interface, electron transport is completely blocked. The current can be turned on by a magnetic field, because it will tilt the antiparallel spins. Note that the MR of such a system is infinitely large.

This spin-valve mechanism is in fact realized in magnetite (Fe_3O_4) layers grown epitaxially on MgO substrates. We assume that the conduction electrons are fully spin polarized (sp), which follows from band calculations which show that magnetite is a half-metallic ferrimagnet with a gap at the Fermi energy for the spin-up but not for the spin-down electrons [5]. In magnetite films the presence of so-called antiphase boundaries (APB) has been revealed by transmission electron microscopy experiments [6,7]. The APB are natural growth defects, resulting from the fact that the lattice constant of Fe_3O_4 ($a = 8.3987$ Å) is twice as large as the one of MgO ($a = 4.212$ Å). The magnetic coupling over a large fraction of these boundaries is antiferromagnetic (AF).

Because of the presence of these AF-APB, both the magnetic and transport properties of the films are different from those of the bulk material. For example, it is very difficult to fully magnetize Fe_3O_4 films [6] and films <5 nm behave superparamagnetically [8]. For thin films the resistivity is orders of magnitude larger, 0.17 and 0.016 Ω cm for 12 and 40 nm thick films, respectively, compared to the bulk value of 0.005 Ω cm. The magnetoresistance is large even at room temperature, in contrast to single crystals [9], and does not saturate in high fields [10]. The sp electrons encounter a high resistance at the AF-APB. Upon application of a magnetic field, the AF spins will align themselves to some degree with the magnetic field, thus increasing the electron transport across the boundaries. This is shown schematically in Fig. 1.

We have investigated the magnetoresistance effects for ultrathin epitaxial Fe_3O_4 films by means of resistance measurements in fields up to 5 T with the field both parallel and perpendicular to the film plane. The Fe_3O_4 ultrathin films (12 and 40 nm thick) were grown on cleaved MgO (001) substrates using molecular beam epitaxy. During growth an oxygen pressure of 10^{-6} mbar, a substrate temperature of 250 °C, and an iron flux of 1.2 Å/min were used; this procedure is known to give a good stoichiometry [8]. Resistance measurements were performed in a commercial PPMS system from Quantum Design (contacts consisting of 20 nm Ti and 40 nm Au). Linear I/V curves were obtained by applying dc voltages and measuring the current in a four-point geometry along the [100] direction of the films.

Figure 2 shows low field MR measurements (up to 1 T) on the 12 nm thick sample measured at 125 K, with the magnetic field applied both perpendicular and parallel to

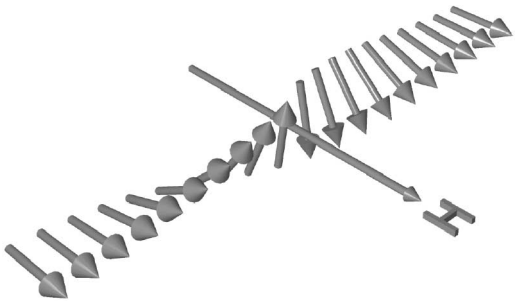

FIG. 1. Spin orientation of two ferromagnetic chains with antiferromagnetic coupling at an atomically sharp boundary subject to a magnetic field.

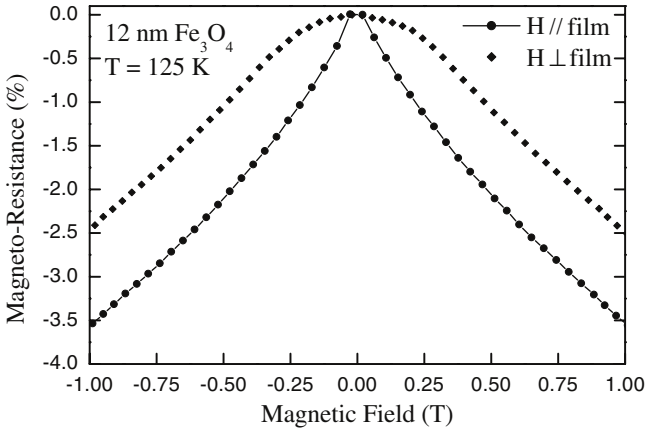

FIG. 2. Low field magnetoresistance of 12 nm thick epitaxial Fe_3O_4 films at 125 K for magnetic fields applied perpendicular and parallel to the film. Up to 0.5 T, the out-of-plane field dependence is quadratic, whereas the in-plane field dependence is linear.

the film plane. The out-of-plane MR measurements show quadratic behavior up to 0.5 T, whereas the in-plane MR behavior is linear in this regime. In this Letter we demonstrate that these results are consistent with a model of spin-polarized transport across an atomically sharp antiferromagnetically coupled boundary. The electron transport occurs through small polaron hopping [11]. By developing the Kubo formula to the first order in the transfer integral t, it was shown [12] that in the nonadiabatic limit the conductivity is proportional to t^2. If the spin on a neighboring ion is rotated over an angle $\varphi_{\rm nn}$, the transfer integral is reduced to $t = t_0 \cos\varphi_{\rm nn}/2$. Consequently, the transfer of the conduction electron between ions with antiparallel spins at an AF-APB is blocked and the conductivity is zero. On application of a magnetic field, this angle deviates from π and the conductivity becomes finite.

The magnetic field (H) dependence of the conductivity (σ) will now be calculated in a 1D model by considering a chain of spins coupled ferromagnetically with one antiferromagnetic boundary. A magnetic field aligns the spins far from the boundary, whereas the spins close to the boundary are only slightly affected by the field, as shown in Fig. 1. At weak fields the configuration can be affected by magnetic anisotropies. We take only the uniaxial anisotropy into account since in magnetite films the magnetocrystalline anisotropies are of minor importance [13]. For magnetic fields parallel to the film most of the spins remain in the plane of the film so that the uniaxial anisotropy term does not play a role. We start the calculation by considering only a half-infinite spin chain, i.e., a spin chain on one side of the antiferromagnetic boundary. The energy per unit area of such a chain, γ_1, is given by [14]

$$\gamma_1 = \int_{-\infty}^{0} \left[-M_s H \cos\varphi + A_{\rm F} \left(\frac{d\varphi}{dx} \right)^2 \right] dx \,. \qquad (1)$$

The first term in the integral is the Zeeman and the second term is the nearest neighbor exchange contribution. The angle between the saturation magnetization M_s and the magnetic field H is φ, and $A_{\rm F}$ is the exchange stiffness constant. Following standard variational calculus procedures, the second or integrated Euler condition can be written as

$$-M_s H(\cos\varphi - \cos\varphi_{-\infty}) = A_{\rm F} \left(\frac{d\varphi}{dx} \right)^2 . \qquad (2)$$

Since the spins far from the boundary are parallel to each other and to the field, $\cos\varphi_{-\infty} = 1$. It is now straightforward to calculate the equilibrium configuration of the half-infinite spin chain and the total energy from Eqs. (1) and (2) for a given value of φ.

Next we determine the equilibrium configuration of the full chain. This is done by minimizing the total energy of the full chain, consisting of the contribution γ_1 and γ_2 of two half-infinite chains on either side of the boundary and an antiferromagnetic coupling energy at the boundary [14],

$$\gamma_{\rm total} = \gamma_1 + \gamma_2 + \frac{A_{\rm AF}}{d} [1 - \cos(\varphi_2 - \varphi_1)], \qquad (3)$$

where φ_1 and φ_2 are the angles between the spins at the left- and the right-hand side of the boundary and the magnetic field, respectively. $A_{\rm AF}$ is the (negative) exchange stiffness constant for the AF exchange interaction at the boundary and d is the distance between two neighboring spin chains along the boundary. The total energy has to be minimized with respect to φ_1 and φ_2. The derivatives of the energy for the half-infinite chains can be calculated using the relation $d\gamma/d\varphi = 2A(d\varphi/dx)$, which can be derived from Eqs. (1) and (2). The final result of the minimization is $\varphi_1 = -\varphi_2 = \varphi_{\rm nn}/2 = \varphi_{\rm AF}$ and

$$HM_s = W_{\rm AF}(\cos^2\varphi_{\rm AF} + \cos^3\varphi_{\rm AF}), \qquad (4)$$

where $W_{\rm AF} = A_{\rm AF}^2/A_{\rm F} d^2$. Taking the literature values for $A_{\rm F}$ and the antiferromagnetic coupling in magnetite [6,8], in a field of 1 T, $W_{\rm AF}/HM_s \geq 10$. For such a strong AF coupling the angle $\varphi_{\rm AF}$ will still be close to $\pi/2$, and the cubic term may be neglected.

In order to calculate the conductivity we recall that $\sigma \propto t^2 = t_0^2 \cos^2\varphi_{\rm AF}$. As long as the alignment at the boundary is nearly antiparallel, by far the largest mobility drop will be experienced at the interface. Based on Eq. (4) we expect that the transport through an AF-APB without magnetic anisotropy is blocked if no magnetic field is present, and it increases approximately linearly with a magnetic field.

If a weak field is applied perpendicular to the plane of the film, the uniaxial anisotropy has to be taken into account. Adding the uniaxial anisotropy energy density $K\cos^2\varphi$ to the integral of Eq. (1) two different regimes can be distinguished for the field dependence. For external magnetic fields smaller or larger than the uniaxial anisotropy field $H_{\rm an}$ the following approximate results are

obtained:

$$I\text{: } H < H_{\text{an}} \rightarrow \cos^2\varphi = \frac{(M_s H)^2}{4KW_{\text{AF}}}, \tag{5}$$

$$II\text{: } H > H_{\text{an}} \rightarrow \cos^2\varphi = \frac{M_s H - K}{W_{\text{AF}}}. \tag{6}$$

Following the argument used previously, we expect that the conductivity of an AF-APB changes quadratically with a magnetic field along a hard magnetization axis until it equals the anisotropy field and is approximately linear for higher fields.

The magnetoresistance measurements were performed on ultrathin layers of magnetite containing many AF-APB. Transmission electron microscopy analysis shows that the average domain size for a 12.5 nm and a 40 nm thick film are 30 nm and 50 nm, respectively. To make a connection between the single boundary calculations and the macroscopic experiments, we assume that the layer consists of sheets of highly resistive material dispersed in a matrix of ordinary bulk material. The total conductivity then is an (unknown) function of a field independent σ_{bulk} (as a single crystal without AF-APB's does not show MR [9]) and a field dependent σ_{AF}, which can be developed into powers of the latter, or

$$\sigma(H) = \sigma_0 + \left(\frac{d\sigma}{d\sigma_{\text{AF}}}\right)_0 \sigma_{\text{AF}}(H) + \ldots. \tag{7}$$

Assuming that the field dependent part is small, only the term linear in $\sigma_{\text{AF}}(H)$ contributes to the magnetoresistance, and

$$\text{MR} = \frac{\rho_H - \rho_0}{\rho_H} \approx -\left(\frac{d\sigma}{d\sigma_{\text{AF}}}\right)_0 \frac{\sigma_{\text{AF}}(H)}{\sigma_0}. \tag{8}$$

Thus in first order the magnetoresistance should be proportional to the average (negative) single boundary conductivity.

The model gives a good qualitative description for the observed features of the MR behavior. Figure 2 shows that the low in-plane field behavior of the MR curve is linear in the applied field, as expected from Eq. (4). This figure also shows that for the out-of-plane measurements the MR has a quadratic field dependence for fields smaller than 0.5 T. This value is in good agreement with the uniaxial anisotropy field of 0.53 T obtained from magnetization loops for a 115 nm thick Fe_3O_4 film [13].

For larger fields, a linear dependence on the magnetic field is expected. The full field MR behavior (up to 5 T) for 12 and 40 nm thick films measured at 125 K (magnetic field applied both parallel and perpendicular to the film plane) is shown in Fig. 3. The high field behavior starts to deviate from linearity. These deviations can partly be explained by taking the full field dependence of Eq. (4) into account. For larger fields, the cubic term in Eq. (4) cannot be neglected (as in 5 T $W_{\text{AF}}/HM_s \geq 2$) and causes the MR to deviate from linearity. Another contribution to the nonlinearity is the distribution of AF exchange coupling strengths for the boundary regions. This results from the fact that different types of boundaries [6] do not have the same number of AF coupled neighbors in the boundary region. In regions where more than two boundaries meet, the AF coupling might be frustrated [8], which also changes the effective AF coupling strength.

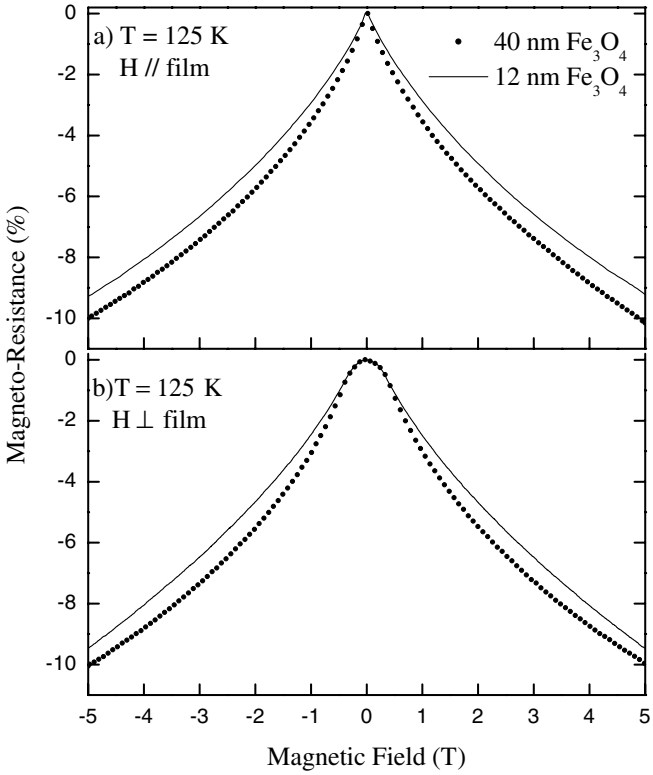

FIG. 3. High field magnetoresistance of 12 and 40 nm thick epitaxial Fe_3O_4 films at 125 K for magnetic fields applied (a) parallel to the film and (b) perpendicular to the film.

The MR decreases with increasing temperature. At 300 K, the MR is reduced to 3% (not shown here). However, the shape of the MR curves is independent of temperature. This is shown in Fig. 4, where the curves have been normalized by taking the value at 5 T to be -1. This observation is in excellent agreement with the model presented above since the field dependent part of the conductivity is a single boundary property and does not depend on temperature. The shape does change with film thickness and the deviations from linearity at high field increase for the thicker samples. Figures 3 and 4 both show that the MR curves are still not saturated in applied fields of 5 T. Margulies *et al.* [6] observed that the same holds for the magnetization, which has not saturated in applied fields of 7 T. This has also been related to the strong antiferromagnetic exchange coupling at the APB.

Similar observations of the MR of Fe_3O_4 grown on MgO [15] and $SrTiO_3$ [16] have been reported. Ziese and Blyte [15] modeled the MR by considering two ferromagnetic chains (with fixed spin orientation) coupled through one spin at the boundary that is allowed to change its

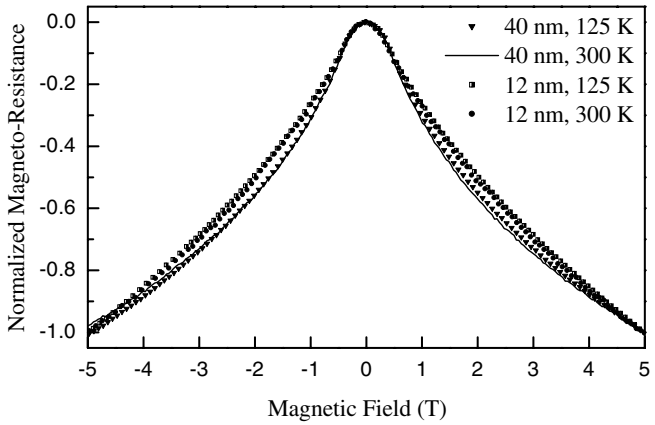

FIG. 4. Normalized magnetoresistance (at 125 and 300 K) by taking the value at 5 T to be -1, for 12 and 40 nm thick epitaxial Fe_3O_4 films and magnetic fields applied perpendicular to the film. The shape is independent of temperature but depends on the film thickness.

orientation. Our model takes the full field dependence of all spins in the chain into account, including the AF coupling and thus gives a more realistic picture of the spin configuration near the boundary.

For the films grown on $SrTiO_3$ [16] the MR data show a similar field dependence as has been observed in the manganites. The transport properties of the manganites are in many respects similar to those of Fe_3O_4. Whereas the colossal magnetoresistance (CMR) in the manganites is associated with the field dependence of the metal-insulator transition taking place at T_C, our results on Fe_3O_4 are related to the MR behavior of the manganites below T_C. For the manganites, it was shown that a dominant MR term can be generated by introducing many grain boundaries, i.e., in powdered, sintered samples. Spin-polarized tunneling between antiferromagnetically oriented grains is the dominant term for MR at low fields [17]. For these polycrystalline samples, the high field MR is also independent of temperature and mainly linear in the applied magnetic field and is associated with the alignment of spins in a magnetically disordered region [17,18]. We think that this mesoscopic description of the conduction mechanism between different grains is in several respects analogous to the localized hopping model presented by us. However, in contrast to the grain boundaries, our films consist of domains that are structurally intergrown, and the magnetic properties of adjacent domains are strongly coupled. Therefore, an atomically sharp interface without a disordered region with switchable magnetic orientations is obtained.

In summary, we have performed magnetoresistance measurements on Fe_3O_4 films containing atomically sharp antiferromagnetic boundaries. The shape of the MR curves is independent of temperature. The MR curves can be described by a model of spin-polarized transport across an atomically sharp AF coupled boundary. The MR measurements yield linear and quadratic field dependence up to the anisotropy field for fields applied parallel and perpendicular to the film plane, respectively, in agreement with the model. The measurements have been performed over many AF boundaries. In order to fully compare the MR data with the model it is of interest to study the MR of a single boundary, as the MR is expected to be very large.

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