Exchange Field Induced Magnetoresistance in Colossal Magnetoresistance Manganites

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The effect of an exchange field on the electrical transport in thin films of metallic ferromagnetic manganites has been investigated. The exchange field was induced both by direct exchange coupling in a ferromagnet/antiferromagnet multilayer and by indirect exchange interaction in a ferromagnet/paramagnet metallic superlattice. The electrical resistance of the metallic manganite layers was found to be determined by the magnitude of the vector sum of the effective exchange field and the external magnetic field.

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Perovskite magnitudes have attracted a lot of attention because their resistance strongly depends on applied magnetic field, an effect known as colossal magnetoresistance (CMR) [1]. In this Letter, we demonstrate that the resistance of thin CMR films depends not only on external magnetic field but also on the effective exchange field that is quantum mechanical in origin. In particular, it is shown that the resistance of a manganite film is determined by the absolute value of the vector sum of the effective exchange field and the external magnetic field.

We have measured the magnetoresistance of two types of magnetic multilayer systems involving thin ferromagnetic manganite films. The first system is an antiferromagnetic/ferromagnetic/antiferromagnetic (AF/F/AF) trilayer where the F film is a metallic ferromagnetic manganite, $La_{2/3}Ca_{1/3}(Sr_{1/3})MnO_3$, and the AF films are insulating antiferromagnets $La_{1/3}Ca_{2/3}MnO_3$. The exchange field in this system is created by direct exchange coupling (exchange bias) between the F and AF layers [2,3]. The second system is a superlattice consisting of alternating ferromagnetic and paramagnetic metallic layers $(F/P)_N$, where the F layers are $La_{2/3}Ba_{1/3}MnO_3$ manganite films and the P layers are $LaNiO_3$ nickelate films [4]. The F layers are antiferromagnetically coupled via the Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction that creates an indirect exchange field acting on the ferromagnetic manganite layers [5].

All the samples investigated were single-crystal multilayers grown by ozone-assisted molecular beam epitaxy on $SrTiO_3$ (100) substrates. The details of sample preparation characterization have been given elsewhere [3,4]. The magnetoresistance was measured by a four-point ac technique with the current flowing in the plane of the heterostructures along the [010] crystallographic direction. A magnetic field of constant magnitude was rotated through 360° in the plane of a sample and the angular dependence of the resistance, which we will refer to as the rotational magnetoresistance (RMR), was measured. Throughout this paper the measured RMR amplitude for a given magnetic field is defined as the difference between the maximum and the minimum resistance divided by the maximum resistance as the field rotates through 360° in the plane of a heterostructure. The purpose of defining the RMR is to distinguish it from the conventional intrinsic anisotropic magnetoresistance (AMR); as will be seen, the RMR includes the intrinsic AMR as well as novel exchange field induced magnetoresistance.

For reference, the AMR of a single $La_{2/3}Ca_{1/3}MnO_3$ film of the same thickness (29 Å) as the F film in the AF/F/AF sample was measured (see Fig. 1a). This film exhibited an AMR similar to that observed in simple metal

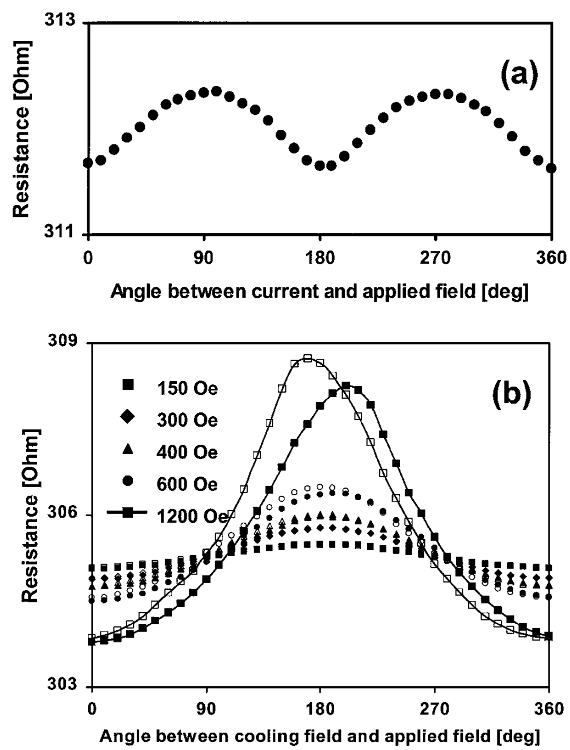

FIG. 1. (a) Resistance versus angle between the current and a saturating applied field of 1 kOe for an unbiased $La_{2/3}Ca_{1/3}MnO_3$ sample at 4.2 K. (b) Dependence of the resistance on the angle between the cooling field and the applied field directions for an exchange-biased $La_{2/3}Ca_{1/3}MnO_3$ sample at 4.2 K. The solid symbols represent the data for the clockwise rotation of the applied field while the open symbols represent the data for the counterclockwise rotation.

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ferromagnets [6,7]. In particular, (i) the AMR exhibited a 180° periodicity, (ii) sample resistance depended on the angle between the current and magnetization, and (iii) the AMR amplitude first monotonically increased and then saturated at about 0.2% with the increasing applied field (see Fig. 2). These three features define the behavior that we will refer to as the intrinsic AMR. The CMR of this sample $[R(0\text{ T}) - R(5\text{ T})]/R(5\text{ T})$ had its peak value of 80% near $T = T_C$ ($T_C = 210$ K), while at $T = 4.2$ K it was 15%.

The AF/F/AF manganite samples exhibited an effect called exchange bias that results from exchange interaction between magnetic moments of the F and AF layers at their interface [8]. It can be described as arising from an effective exchange field $\mathbf{H}_{\mathrm{EX}}$ acting on the F layer at the AF/F interface or, equivalently, from a unidirectional magnetic anisotropy. The direction of the exchange field is set by the direction of the F magnetization in a saturating field as the AF/F/AF system is cooled to a temperature below the Néel temperature of the AF ($T_N = 170$ K for $La_{1/3}Ca_{2/3}MnO_3$). The RMR data for a $La_{1/3}Ca_{2/3}MnO_3$(200 Å)/$La_{2/3}Ca_{1/3}MnO_3$(29 Å)/$La_{1/3}Ca_{2/3}MnO_3$(200 Å) trilayer are shown in Fig. 1b for several different values of the applied field H_A. The low field RMR periodicity for the exchange-biased structure is 360°, rather than the 180° expected for the AMR effect. The low field curves are fully reproducible for clockwise and counterclockwise rotations of the applied field. However, increasing the field above a threshold leads to the appearance of irreversible changes in $\mathbf{H}_{\mathrm{EX}}$ [9], which gives rise to a resistance hysteresis. For large fields ($H_A > 2$ kOe), the RMR curves exhibit two maxima while the RMR amplitude decreases and approaches the AMR amplitude of the unbiased film. Similar results were obtained for an exchange-biased $La_{2/3}Sr_{1/3}MnO_3$ sample.

In general, the exchange field modifies the measured RMR compared to that of an unbiased film for both the manganites and conventional exchange-biased ferromagnets. In both cases the magnetization points in the direction of the vector sum of the applied field and the exchange field. For a conventional ferromagnet, the resistance is determined *only by the direction of its magnetization via the intrinsic AMR effect,* and a simple analysis of the angular dependence of the resistance on the in-plane applied field direction yields the exchange field magnitude [10]. Essentially, the exchange field pins the F layer magnetization, thus reducing the amount of the magnetization rotation for modest applied fields. This results in a resistance periodicity of 360° and decreases the observed amplitude of the RMR. As the applied field is increased, the degree of rotation increases monotonically, increasing the RMR amplitude until the intrinsic AMR amplitude is reached (and the AMR periodicity crosses over from 360° to 180°). In the case of a conventional F, for no value of the applied field can the measured rotational resistance amplitude exceed the amplitude of the intrinsic AMR.

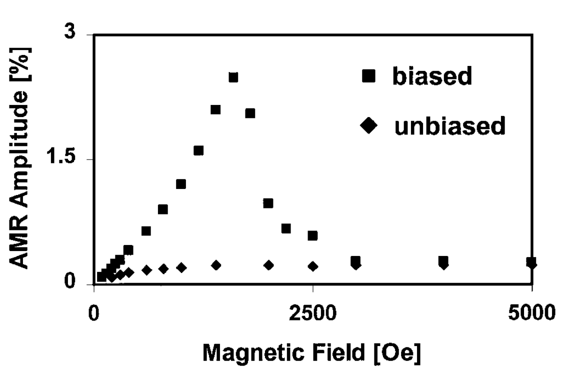

FIG. 2. The AMR amplitude at $T = 4.2$ K versus applied magnetic field magnitude for both a biased and an unbiased sample of $La_{2/3}Ca_{1/3}MnO_3$.

For the exchange-biased manganites, however, the observed RMR behavior is different. First, the measured RMR amplitude is a nonmonotonic function of the applied field magnitude with a maximum at $H_A = 1.6$ kOe, as shown in Fig. 2. Second, the RMR amplitude at its maximum value exhibits enhancement of approximately a factor of 10 relative to the intrinsic AMR amplitude of an unbiased film. Third, the resistance of the exchange-biased film depends mainly on the angle between the cooling field and the applied field rather than on the angle between the current and magnetization. These three observations demonstrate that the RMR of exchange-biased manganites does not originate from the intrinsic AMR.

To explain this behavior, we propose a simple model that is founded on the equivalence of the exchange field and the applied magnetic field similar to that which leads to the Jaccarino-Peter effect in magnetic superconductors [11]. Figure 3 schematically shows the dependence of resistance on the magnitude of the applied field for an unbiased film due to the CMR effect. We argue that the RMR of an exchange-biased manganite film can be explained by this dependence.

In our model, the measured resistance of the AF/F/AF manganite films depends on the magnitude, H_T, of the vector sum of the applied magnetic field $\mathbf{H}_A$ and the exchange field $\mathbf{H}_{\mathrm{EX}}$ arising from the two AF layers (see the inset of Fig. 3). In the absence of the external applied field, $\mathbf{H}_T = \mathbf{H}_{\mathrm{EX}}$ and the resistance of the sample is R_0. The application of a small external magnetic field $\mathbf{H}_A$ in the opposite direction from $\mathbf{H}_{\mathrm{EX}}$ results in the decrease of the total field ($H_T = |H_{\mathrm{EX}} - H_A|$), and the resistance of the sample increases ($R = R_A$ in Fig. 3). Correspondingly, if a small field $\mathbf{H}_A$ is applied in the same direction as $\mathbf{H}_{\mathrm{EX}}$ then the total field increases ($H_T = H_{\mathrm{EX}} + H_A$) and the resistance decreases ($R = R_B$ in Fig. 3).

This model explains the single resistance maximum of the RMR in small fields, as well as the increase of its amplitude with increasing field. The shapes of the RMR curves exhibiting a relatively sharp maximum for applied fields close to the H_{EX} are also well described. The model

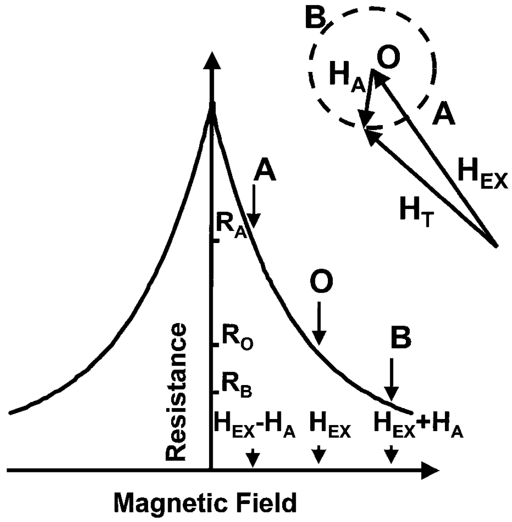

FIG. 3. A schematic dependence of resistance on magnetic field for an unbiased manganite film due to the CMR effect. Inset: Total field $\mathbf{H}_T$ is a vector sum of the applied magnetic field $\mathbf{H}_A$ and the intrinsic exchange field $\mathbf{H}_{EX}$. When an exchange field is present and the applied field is rotated through the plane of the sample, the total field varies from a minimum at point A to a maximum at point B.

also explains why the resistance depends on the angle between the applied and exchanged fields and is almost independent on the current direction in the low field regime. For $H_A > H_{EX}$, the model predicts a decreasing RMR amplitude. Although the simple application of the model for large fields is complicated by irreversible changes in H_{EX}, it is expected that the observed RMR will cross over to the intrinsic AMR behavior both in periodicity and magnitude as the applied field increases. Therefore, the magnetotransport anisotropy in exchange-biased manganites has its origin in the intrinsic *isotropic* magnetoresistance of the ferromagnet (CMR effect), and can be much larger than the intrinsic AMR of the film.

We have also observed exchange field dependent magnetoresistance in manganite/nickelate superlattices. It has been shown that for thin nickelate spacer layers [3 and 4 unit cells (u.c.)] the manganite layers are coupled antiferromagnetically, while for thicker nickelate spacers the coupling first becomes ferromagnetic and then vanishes [5]. The hysteresis loop and magnetoresistance of a $[La_{2/3}Ba_{1/3}MnO_3$ (10 u.c.)/$LaNiO_3$ (4 u.c.)$]_{12}$ superlattice at $T = 5$ K are shown in Figs. 4a and 4b, respectively, and are found to be almost independent on the in-plane field direction. The low field magnetoresistance of this AF-coupled superlattice is positive, in contrast to the negative giant magnetoresistance observed in AF-coupled superlattices of transition metal ferromagnets [12]. For higher applied fields, the magnetoresistance becomes negative. In contrast, ferromagnetically coupled and uncoupled superlattices as well as single films of manganites exhibit only negative magnetoresistance, such as shown in Fig. 3

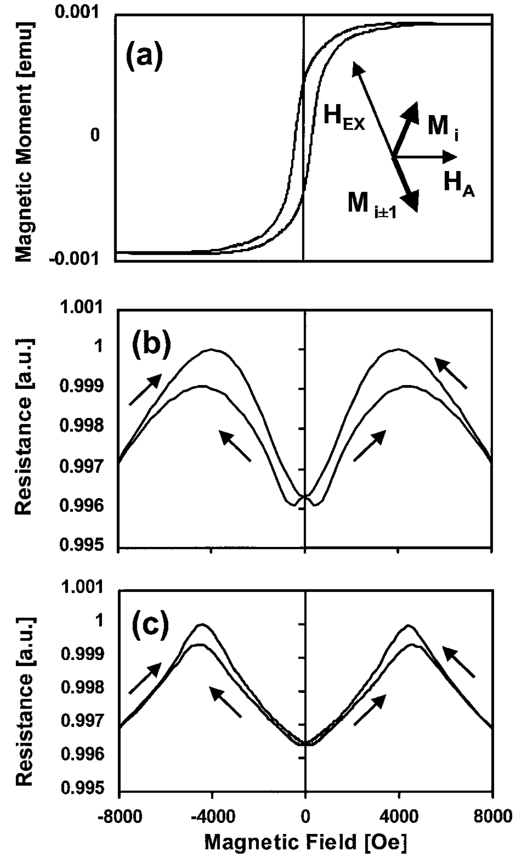

FIG. 4. (a) Hysteresis loop of the antiferromagnetically coupled $[La_{2/3}Ba_{1/3}MnO_3$ (10 u.c.)/$LaNiO_3$ (4 u.c.)$]_{12}$ superlattice at $T = 5$ K. The inset shows the external magnetic field $\mathbf{H}_A$ and the exchange field $\mathbf{H}_{EX}$ acting upon the magnetization of a manganite layer labeled $\mathbf{M}_i$. The exchange field $\mathbf{H}_{EX}$ originates from the RKKY interaction of $\mathbf{M}_i$ with the two adjacent manganite layers $\mathbf{M}_{i+1}$ and $\mathbf{M}_{i-1}$. (b) Resistance versus in-plane magnetic field for this superlattice at $T = 5$ K. (c) The model prediction for the resistance versus applied field calculated from Eq. (1) with $H_{EX} = 4.3$ kOe.

To explain the magnetoresistance of the heterostructures, the antiferromagnetic RKKY coupling in the manganite/nickelate superlattices can be represented by effective exchange field $\mathbf{H}_{EX}$ of constant magnitude acting upon each manganite layer in the superlattice. The two manganite layers adjacent to a given layer produce this exchange field that points in the direction opposite from the magnetization of these two adjacent layers (see the inset of Fig. 4a). We assume, as in the case of the direct exchange field in the AF/F/AF trilayer, that this indirect exchange field alters the resistance of the manganite layer. Again, the resistance is a monotonically decreasing function of the absolute value of the total field, which is a vector sum of the applied magnetic field, and the indirect exchange field. In this case, the total field is given by

$$H_T = \sqrt{H_{EX}^2 + H_A^2 - 2H_{EX}H_A m(H_A)}, \qquad (1)$$

where $m(H_A)$ is the normalized projection of the superlattice magnetization onto the applied field direction

[$m(H_A) = M_Z(H_A)/M_S$, where $M_Z(H_A)$ is shown in Fig. 4a, and M_S is the saturation magnetization of the superlattice]. By using Eq. (1), the $R(H_T)$ shown in Fig. 3, and the experimentally measured $m(H_A)$, we can calculate the magnetoresistance of the AF-coupled superlattice with the magnitude of the exchange field H_{EX} as a fitting parameter. As shown in Fig. 4c for $H_{\mathrm{EX}} = 4.3$ kOe, the calculated magnetoresistance is in good qualitative agreement with the experimentally measured magnetoresistance curve shown in Fig. 4b. We also note that the positions of the resistance maxima occur at the minima of H_T and, therefore, are independent of the exact shape of the magnetoresistance curve shown in Fig. 3 as long as the magnetoresistance is negative. Similar results were obtained for a superlattice with a 3-u.c.-thick nickelate spacer layer. The resistance maxima in this superlattice occur at higher values of the applied field which is consistent with a larger AF-coupling constant observed for a 3-u.c.-thick spacer [5].

This exchange field induced magnetoresistance allows us to extract the approximate magnitude of the exchange field from the magnetoresistance data both in the exchange biased structure and in the AF-coupled superlattice. It is easy to show that the RMR of the exchange-biased film is largest if the magnitude of the applied field is equal to that of the exchange field, and thus the field at which the largest RMR is observed gives the approximate magnitude of the exchange field [13]. In a temperature range from 4.2 to 170 K, the exchange field determined by the hysteresis loop method (0.6 kOe at $T = 4.2$ K was smaller than that given by this magnetoresistance technique (1.6 kOe at $T = 4.2$ K). This is not surprising, because the reversal of the ferromagnet magnetization may significantly decrease the initial magnitude of the exchange field [10]. Therefore, the hysteresis loop shift does not give the *initial* value of the exchange field. On the contrary, MR measurements employing only a small rotation of the magnetization away from the exchange field direction do not alter the exchange field and can yield its initial value.

However, in the case of exchange-biased manganite films, there is an alternative explanation for the large difference between the hysteresis loop shift and exchange field obtained from the magnetoresistance data. It has been argued that the *low temperature* magnetoresistance in CMR manganite films is a surface or an interfacial effect [14] since it not observed in bulk manganites. This means that, although the whole CMR films is magnetic, only a thin layer near the interface is magnetoresistive. We also note that the direct exchange field, being an interfacial effect, is strongest near the interface and rapidly decays in the interior of the CMR film. The combination of these two factors may be responsible for the larger exchange field obtained from the magnetoresistance data than from the hysteresis loop data. Indeed, magnetoresistance measurements probe only the total field in the thin magnetoresistive layer near the interface where the exchange field is the largest. The exchange field extracted from the hysteresis loop data is an average of the microscopic exchange field taken over the entire F layer thickness.

Magnetoresistance of the manganite/nickelate superlattices also supports the interfacial character of the low temperature magnetoresistance in manganite films. The approximate value of the exchange field (proportional to the bilinear coupling constant) obtained by a procedure described in Ref. [5] from the hysteresis loop shown in Fig. 4a is 1.5 kOe which is significantly less than 4.3 kOe obtained from the magnetoresistance data. In this case, the indirect exchange field is also strongest near the manganite film interface due to a short mean-free path of the conduction electrons in manganite films.

In conclusion, the quantum mechanical exchange field alters the resistance of thin films of metallic ferromagnetic manganites. The electrical resistance of these manganite films is determined by the vector sum of the effective exchange field and the external magnetic field. In addition, the magnetoresistance data support the interfacial origin of the low temperature magnetoresistance in single-crystalline thin films of these materials.

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