Intrinsic Colossal Magnetoresistance Effect in Thin-Film $Pr_{0.5}Sr_{0.5}MnO_3$ through Dimensionality Switching

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A homogeneous colossal magnetoresistance (CMR) effect at low temperatures has been found in a thin-film perovskite manganite $Pr_{0.5}Sr_{0.5}MnO_3$. The transition is driven not by the spin alignment as in usual CMR in bulk samples but by the localization-delocalization transition switched by the change in the effective dimensionality. Two-dimensional $(x^2 - y^2)$-orbital ordering enhanced by the substrate strain is essential for the stabilization of the insulating localized state, which is on the verge of the first-order transition to the three-dimensional metallic ferromangetic state.

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The colossal magnetoresistance (CMR) in manganites is one of the most dramatic consequences of electron correlation. Up until now, CMR has been classified into two types: CMR1 and 2 [1]. CMR1 is a magnetic-field-induced transition from a charge-ordered antiferromagnetic insulator (COAFI) to a ferromagnetic metal (FMM), while CMR2 is due to the suppression of the spin fluctuation by an applied field near the Curie temperature, T_C. In CMR1, the two phases between which the transition takes place are symmetrically incompatible and hence are separated by a first-order transition. It has been argued that the ''colossal'' effect is due to the coexistence of the insulating and metallic states: opening and closure of a percolative conducting path can be brought about without too much energy cost and hence by a moderate field [2]. Accumulating evidences show that a material which exhibits CMR1 is indeed quite inhomegeneous in various scales, and that the conduction is percolative [3].

In this Letter, we present yet another CMR effect in a $Pr_{0.5}Sr_{0.5}MnO_3$ film at low temperatures, which does not involve a COAFI as an insulating phase. This is totally different from the case of $Pr_{0.5}Sr_{0.5}MnO_3$ bulk single crystals, which exhibit CMR1 [4]. We discuss the new CMR effect in terms of the localization-delocalization switching due to the magnetic-field induced dimensionality change. Here, the underlying two-dimensional (2D) character is essential and is realized by the stabilizing stress effect of the substrate in the film sample [5].

The sample was prepared by the pulsed-laser deposition technique on a (011)-oriented $(LaAlO_3)_{0.3} - (SrAl_{0.5}Ta_{0.5}O_3)_{0.7}$ (LSAT) substrate. An epitaxial film coherently grown on the substrate was obtained. Despite the firm registry of the film to the substrate, the use of the (011)-oriented substrate, as opposed to the widely used (100) substrate [6], allows the lattice distortion required at the first-order phase transition and enables us for the first time to achieve a sharp metal-insulator transition (MIT) in manganite thin films comparable to that in bulk crystals [7]. Indeed the temperature dependence of the transport and magnetization data of the film [Figs. 1(a) and 1(b)] and those of the bulk crystal [4] are qualitatively alike.

A clear difference, however, appears in the structure. X-ray diffraction study has been performed at BNL NSLS X22C and KEK-PF BL-16A1 equipped with a superconducting magnet. In Fig. 2(a) is shown the temperature dependence of several diffraction peaks in the absence of magnetic field. The lattice indices are in the pseudocubic setting. At room temperature, the film is monodomain and nearly cubic, which does not change on entering into a ferromagnetic (FM) state. At the MIT around 110 K, the film undergoes a sharp structural transformation. The (032) peak [Fig. 2(b)] splits into two peaks corresponding to the in-plane shear deformation along $[0\bar{1}1]$ and $[01\bar{1}]$ directions forming twins. The domain size must be fairly large since the peak width is not affected by the structural change. In contrast to the bulk crystal, however, there are no superlattice peaks that indicate the charge ordering (CO): neither the checkerboard-type CO pattern associated with the *CE*-type spin order nor the stripe-type CO [8] are found. The film remains in the orbital-ferro state down to 15 K.

More insight into the electronic states is obtained by examining the polarized ir absorption spectra as shown in Fig. 3. In the paramagnetic insulating phase [Fig. 3(a)] and in the metallic phase below T_C [Fig. 3(b)], the electronic states are isotropic (i.e., 3D-like) and the spectra are consistent with the respective phase assignment. The metallic phase induced by the magnetic field below MIT is also isotropic [Fig. 3(c)]. The smooth continuation of the metallic phase to lower temperature field-cooled state is evident in Fig. 1(c), which shows the transmission of $[0\bar{1}1]$ polarized light at 0.35 eV. The insulating phase below MIT,

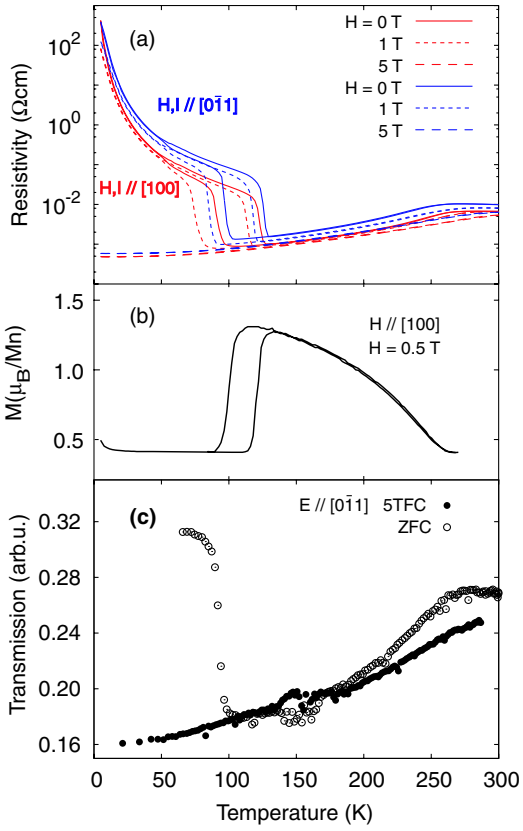

FIG. 1 (color online). (a) Resistivity vs temperature along the two orthogonal in-plane directions [100] and [0$\bar{1}$1] under various magnetic field. (b) Magnetization vs temperature with the magnetic field (0.5 T) applied along [100] direction. The data for the [0$\bar{1}$1] direction are nearly identical. (c) Transmission of [0$\bar{1}$1] polarized 0.35 eV light vs temperature at 0 T and 5 T in cooling.

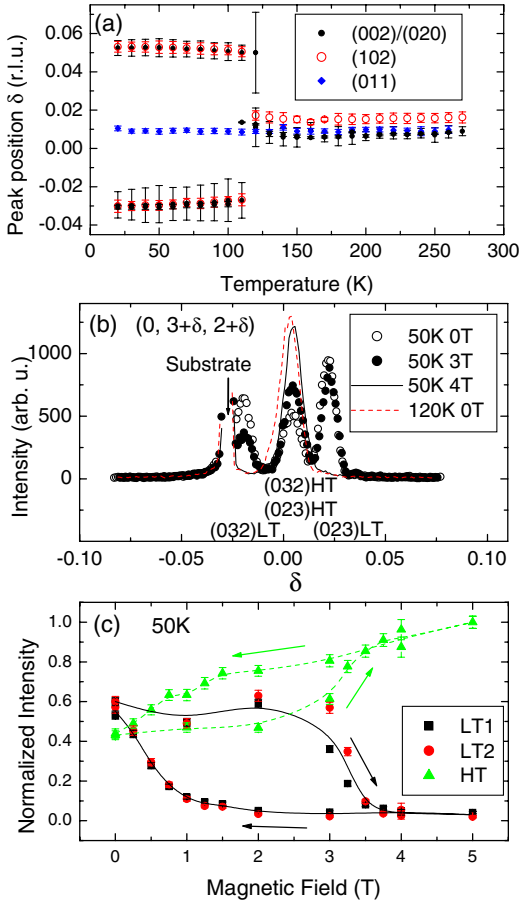

FIG. 2 (color online). (a) Temperature dependence of several diffraction spots. The diffraction indices are in pseudocubic setting. (b) (032) peak at 120 K and 50 K under magnetic field. (c) Magnetic-field dependence of the intensity of peaks at 50 K. Low-temperature peaks (LT1 and LT2) disappear around 4 T and the high-temperature (HT) structure is recovered. All peaks show hysteresis.

however, is very anisotropic [Fig. 3(d)]. The gap in the [0$\bar{1}$1] direction is well above 1 eV while, in the [100] direction, it is only 0.5 eV. From the orbital-ferro order, abrupt shrinkage of $d(001)$ [Fig. 2(a)] at MIT, magnetization, and the optical anisotropy, it is clear that the film is in the $x^2 - y^2$-type orbital and the A-type spin arrangement; the ordering plane is (001). However, the interplane spin arrangement is not completely antiparallel but canted and appreciable amount of ferromagnetic component is obvious as shown in Fig. 1(b) [see also Fig. 4(c) below]. Perovskite manganites with these orbital and spin order are known to exhibit saturating in-plane resistivity (if not metallic) toward low temperature, since the in-plane ferromagnetic and $x^2 - y^2$-type ferro-orbital order allow in-plane conduction while the out-of-plane resistivity is much higher [9]. This is in stark contrast to the nearly isotropic and diverging resistivity shown in Fig. 1(a), which suggests that the nature of the insulating phase in the film is different from that in bulk. Pure 2D conduction is of course not stable against localization driven by disorder; in the bulk crystals, slight 3D interaction may stabilize the conducting state making the resistivity not strongly diverging in any direction. The situation may differ in a film, since the mode of structural deformation is restricted due to the substrate stress and hence the dimensionality of the interaction.

The insulating state is not robust, however. Indeed metallic conduction is easily restored by relatively low magnetic field. Typical CMR effect is shown in Fig. 4. At 60 K [Fig. 4(b)], the sample was initially cooled in a magnetic field of 5 T. As is shown in Fig. 1(a), it was in the FMM state. As the field was reduced to zero, it underwent MIT. The FMM state was again recovered around 4 T in the subsequent application of the magnetic field. The MIT can be repeated afterwards by magnetic field cycling with some hysteresis. The simultaneously taken ir transmission completely matches with the resistivity. This fact alone makes it clear that the CMR effect is not due to the percolative conduction through the FMM phase coexisting with COI. If it were the case, the conduction would have exhibited a steep increase at the percolation threshold [10] while the transmittance should be linearly related to the

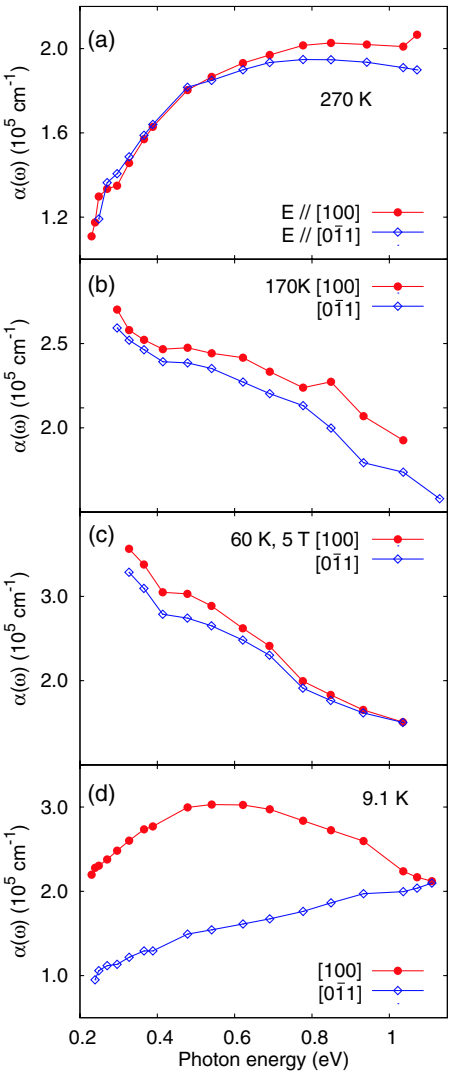

FIG. 3 (color online). Polarized ir absorption spectra. (a) In the paramagnetic insulating phase. (b) At 170 K in the FM phase. (c) At 60 K in the FM phase induced by the magnetic field of 5 T. (d) At 9.1 K in the insulating phase without magnetic field.

area fraction of the FMM phase relative to the insulating phase. The CMR effect is observed throughout the insulating phase although 5 T is not enough for restoring the metallic phase completely at low temperature after reducing the field to zero as shown in Fig. 4(a); the close correlation between the conductivity and transmittance still holds here.

Let us note that the maximum resistivity that satisfies the metallic conduction [the Ioffe-Regel condition [11]] is about 0.9 m Ω cm assuming the lattice constant $a = 0.38$ nm and the half-filled half-metallic band. Figure 1(a) shows that the film is barely 3D metallic even under 5 T. Therefore, as soon as the dimensionality is reduced, the electrons fall into strong localization.

The CMR is not directly related to the magnetization; another distinct feature different from CMR1. To the best of our knowledge, all CMR phenomena reported so far at low temperatures show close correlation between the magnetization and the conductivity. The M-H curve at 60 K shown in Fig. 4(c) shows that the magnetization quickly saturates at low field and behaves nearly ferromagnetically. The weak metamagnetic feature in increasing H appears to correlate with the CMR but it does not compare with the typical CMR1 [e.g., Fig. 3 of Ref. [4]] and no hint of MIT is found in magnetization on reducing the field. On the other hand, there is a large structural change induced by the magnetic field as shown in Fig. 2(c). Although taken at slightly lower temperature, the recovery of the high-temperature monodomain structure with elongated $d(001)$ is clearly related to the CMR effect.

The observations so far suggest that we should look for mechanism other than the double-exchange interaction for the CMR effect seen here. One possible scenario is as follows. Below MIT, the large reduction in c axis promotes 2D character with $x^2 - y^2$ orbital-ferromagnetic order [12]. However, a 2D electronic state is susceptible to localization. Once localized, the electronic state is further stabilized by a relatively large energy barrier [the pseudogap [13]] as is evidenced by the fact that the dc conductivity keeps track with the optical conductivity (i.e., the transmittance) at as high as 0.35 eV [Figs. 4(a) and 4(b)]. In fact, the optical conductivity around 0.3 eV is calculated to

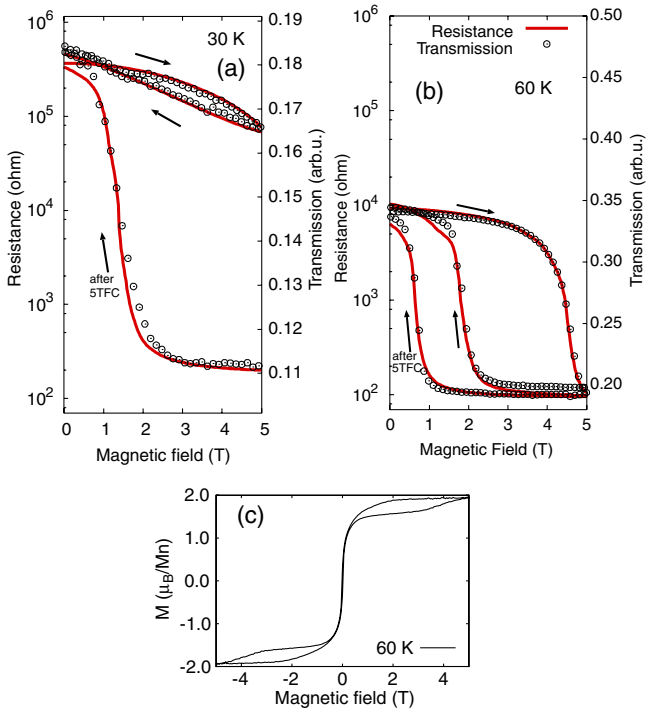

FIG. 4 (color online). (a) Magnetoresistance and magnetotransmittance for 0.35 eV light polarized along [0$\bar{1}$1] direction at 30 K. This is the interplane direction of the orbital order. For the sake of clarity, only every 5 data points are shown for the transmittance. (b) Magnetoresistance and magnetotransmittance at 60 K. (c) Magnetization curve at 60 K.

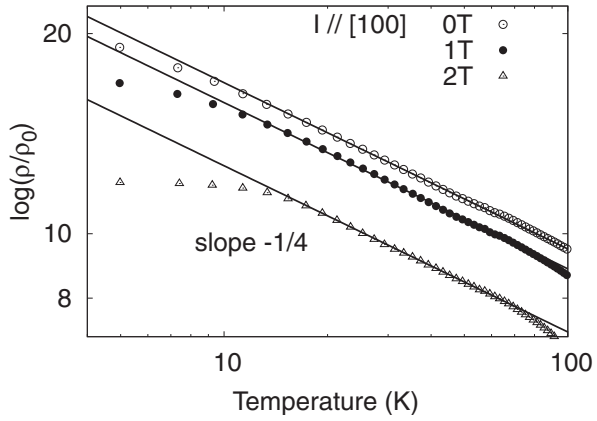

FIG. 5. $\log(\rho)$ vs T in the logarithmic scale. The variable-range hopping model should yield a linear relationship and the slope is the exponent. The lines are slope of $-1/4$ and drawn for the guide of the eye. The fit along $[0\bar{1}1]$ direction is qualitatively similar.

be about $2 \times 10^3\ \Omega^{-1}\,\mathrm{cm}^{-1}$ for the case of Fig. 3(c) and extrapolates reasonably well to the observed dc value [Fig. 1(a)]. Two possible mechanisms that can create a pseudogap are Coulomb gap and polaronic gap: both are expected to be in the range of 0.5–1 eV in manganites. As shown in Fig. 5, the low-temperature conduction is well described by 3D variable-range hopping model $\log(\rho/\rho_0) \propto T^{\alpha}$ [11]. The exponent ($\alpha \sim -1/4$) indicates that the Coulomb gap is not at work here, which should result in a larger exponent ($\alpha \sim -1/2$) [14]. The isotropic (3D-like) and the diverging resistance in the presence of strong 2D interaction are thus not contradicting each other. The magnetic field governs the dimensionality and hence the degree of localization while the temperature dependence of the resistivity is isotropic because it is dominated by the variable-range hopping of small polaron.

The idea of the insulating behavior by strong localization in manganites is, of course, nothing new [15] and the importance of localization in layered systems has been discussed [16]. The role of the electron-phonon interaction in determining the large energy scale landscape of the electronic states in layered manganites has been pointed out from angle resolved photoemission spectra (ARPES) [13], which is so strong that the density of states at the Fermi energy is nearly depleted even in the metallic state [17,18]. This tendency should be of less importance in 3D and hence the effect is easily visible in the layered manganites.

A thin film formed on a substrate with appropriate symmetry and in-plane lattice constant is a system that can go between 2D and 3D. In the present example, a (011) substrate can easily accommodate the change in $d(001)$ [Fig. 2(c)], which governs the dimensionality of the electronic state in perovskite manganites [12]. When it shrinks, the 2D character sets in. However, the exact role the magnetic field plays in the CMR presented here seems extremely subtle and is not obvious. Although the magnetostrictive effect clearly causes the major part of the dimensionality switching, this alone seems to be not enough to fully account for the phenomena.

In conclusion, we have presented a new CMR effect that does not scale with the magnetization and does not involve phase separation. Although the effect is clearly visible in a thin-film sample, it may have consequence in bulk CMR manganites as well [19].

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