Pressure-Enhanced Interplane Tunneling Magnetoresistance in a Layered Manganite Crystal

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The pressure-induced variations of the anisotropic charge-transport and magnetic properties have been investigated for a bilayered manganite crystal, $La_{2-2x}Sr_{1+2x}Mn_2O_7$ ($x = 0.3$). The magnitude of the magnetic coupling between the adjacent bilayers can be weakened by application of pressure, which makes the charge dynamics more two dimensional (2D) but highly incoherent. As a result, the restoration of the interlayer hopping of the spin-polarized carriers by application of magnetic field, namely the field-induced 2D-3D crossover, gives rise to an extremely high ratio of interplane tunneling magnetoresistance, say, up to $\Delta\rho_c/\rho_c \sim 4000\%$ at 4.2 K under ~10 kbar. [S0031-9007(97)04423-2]

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Recent progress of the giant magnetoresistance study in magnetic multilayers has shed light on the study of the tunneling magnetoresistance (TMR) effect in ferromagnetic metal (FM)−insulator (I)−FM junctions. Several TMR systems with use of perovskite manganites have also been proposed and investigated, such as trilayer junctions [1,2] and granular polycrystals with intergrain boundaries [3–6]. The nearly complete exchange splitting of the spin-polarized conduction bands and the resultant high spin-polarization in the FM ground state of the perovskite manganites [7] are anticipated to drastically enhance the TMR. A recent finding of the gigantic TMR effect in a layered perovskite manganite $La_{2-2x}Sr_{1+2x}Mn_2O_7$ ($x = 0.3$) provides a novel approach to the large MR attainable at low magnetic fields [8]. The layered crystal is composed of FM MnO_2 bilayers with intervening insulating $(La,Sr)_2O_2$ blocks, as seen in the inset of Fig. 1, and can be viewed as an infinite array of FM/I/FM junctions. In such a quasi-two-dimensional (quasi-2D) FM, the interplane as well as inplane charge dynamics (and hence the MR characteristics) is expected to critically depend on the interlayer magnetic coupling between the FM MnO_2 bilayers. Here, we have utilized an external pressure to finely control such an interlayer coupling in this layered manganite, although the consequence of the pressure is highly nontrivial and unpredictable for the layered perovskite type structure with anisotropic compressibility and complex ionic-covalent bonding character [9]. In this Letter, we investigate the pressure effect on the charge-transport and magnetic properties in the layered manganite $La_{2-2x}Sr_{1+2x}Mn_2O_7$ ($x = 0.3$). In reality, the applied pressure rather weakens the interlayer coupling, realizes the 2D FM state down to low temperature, and drastically enhances the interplane TMR ratios up to $\Delta\rho_c/\rho_c \sim 4000\%$ at 4.2 K. The results have confirmed the importance of the interlayer magnetic coupling in the TMR process of the bulk crystal, and also unraveled the highly incoherent nature of the charge dynamics in the 2D FM state of the manganite.

Single crystals of $La_{2-2x}Sr_{1+2x}Mn_2O_7$ ($x = 0.3$) were grown by the floating zone method, as previously described [10]. The grown crystals were characterized by the four-circle single crystal x-ray and high-resolution transmission electron diffraction measurements to ensure single phase of the bilayer structure. The crystal was oriented using Laue x-ray diffraction patterns, and cut into a rectangular shape of typical dimension $\sim 1 \times 1 \times 0.1$ mm^2. The electrodes on the sample were formed by heat treatment type silver paint. Resistivity measurements were made by a low-frequency (15.9 Hz) four-probe technique with current parallel (ρ_{ab}) and perpendicular (ρ_c)

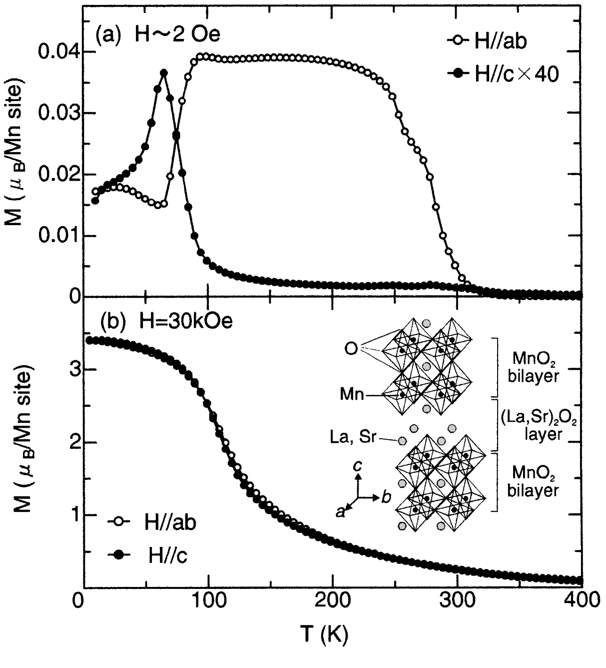

FIG. 1. Temperature dependence of zero-field cooled dc magnetization under a magnetic field H of (a) ~2 Oe and (b) 30 kOe with $H \parallel ab$ and $H \parallel c$ in the $La_{2-2x}Sr_{1+2x}Mn_2O_7$ ($x = 0.3$) crystal. The inset shows the crystal structure of $La_{2-2x}Sr_{1+2x}Mn_2O_7$.

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to MnO_2 bilayers. Magnetic fields H were provided by a superconducting magnet. Magnetization measurements at ambient pressure were performed by using a commercial SQUID magnetometer. The quasihydrostatic pressure was obtained with a clamp type pressure cell using Fluorinert as a pressure transmitting medium. The pressure effect on the magnetic susceptibility χ was investigated by use of a mutual inductance bridge at a frequency of 15.9 Hz. All the pressure values indicated in this paper are the ones calibrated by measuring the change in the superconducting transition temperature of lead [11].

Figure 1 displays the temperature dependence of the magnetization measured at low and high H. The low-H (~2 Oe) magnetization parallel (M_{ab}) and perpendicular (M_c) to MnO_2 bilayers is strikingly anisotropic. A steep rise of M_{ab} toward lower temperatures is observed at ~300 K where ρ_{ab} deviates from a thermal activation type behavior [see Fig. 2(a)], as previously reported [8]. It can be understood in terms of the evolution of the inplane short-range 2D ferromagnetic correlation below ~300 K. At temperatures around $T_c \sim 90$ K, M_c remarkably increases, while M_{ab} decreases. With further decreasing temperature, M_c exhibits a sharp anomaly centered at $T_N \sim 60$ K, somewhat similar to that found in the parallel susceptibility of an antiferromagnet. In conjunction with results of preliminary neutron diffraction measurements [12], we can infer that the 3D spin ordering takes place below T_c where the magnetic moments cant toward the c axis with the decrease of temperature. In other words, the interlayer canted antiferromagnetic order evolves with decreasing temperature down to T_N. Below T_N, the interlayer antiferromagnetic spin-ordering appears to evolve. At high H, e.g., at 30 kOe, however, the steep drop of M_c into lower temperatures below T_N as well as the anisotropy between M_{ab} and M_c disappears as shown in Fig. 1(b). The observed magnetic moment of $M \sim 3.4\mu_B$ at the lowest temperature at 30 kOe is close to that expected for the full Mn moment. These results suggest that the metamagnetic transition occurs below T_N with increasing a magnetic field.

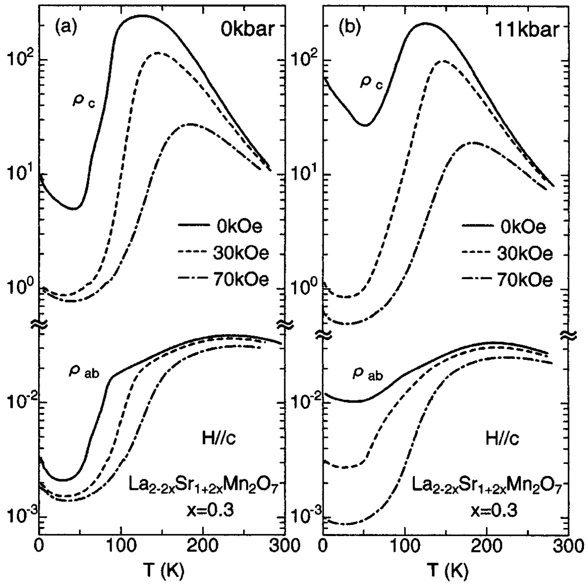

FIG. 2. Temperature dependence of inplane (ρ_{ab}) and interplane (ρ_c) resistivity under pressures of 0 kbar (a) and 11 kbar (b) at several magnetic fields with $H \parallel c$ in the $La_{2-2x}Sr_{1+2x}Mn_2O_7$ ($x = 0.3$) crystal.

Let us turn to the magnetic field and pressure effects on ρ_{ab} and ρ_c. Figures 2(a) and 2(b) show the temperature profiles of ρ_{ab} and ρ_c at several magnetic fields, under pressures of 0 and 11 kbar, respectively. All these data were taken in the warming run. As seen in Fig. 2(a), ρ_{ab} and ρ_c steeply decrease into a low-temperature metallic state around T_c under ambient pressure. Taking account of the magnetization data, we can ascribe this resistive drop around T_c to the onset of the 3D spin-ordering. Common to both ρ_c-T curves at 0 and 11 kbar, the large interplane MR effect is observed above T_c. This can be interpreted in terms of the enhancement of the spin-polarized c-axis transport under a magnetic field. Comparing Fig. 2(a) with Fig. 2(b), we find that the magnetic field effects on ρ_{ab} and ρ_c under 11 kbar are essentially the same as those under ambient pressure, as far as the temperature-region above T_c is concerned.

The most striking feature is found below T_c. Both the ρ_{ab} and ρ_c are remarkably increased by applying pressure of 11 kbar at zero magnetic field, while under magnetic fields of 30 and 70 kOe the pressure effect on ρ_{ab} and ρ_c is considerably smaller below T_c. In other words, the steep drops of ρ_{ab} and ρ_c are suppressed by applying pressure in the absence of a magnetic field, which signals the pressure suppression of the 3D spin ordering. By applying magnetic fields, the 3D ferromagnetic spin arrangement should be realized even at high pressures or irrespective of pressure. This may explain the rather weak pressure dependence of resistivity at high H.

To further clarify the origin of large pressure effect on ρ_{ab} and ρ_c below T_c, we display in Fig. 3 the temperature profiles (in the warming run) of the c axis ac susceptibility χ_c with $H \parallel c$, ρ_{ab}, and ρ_c under several pressures (but without an external magnetic field throughout the measurement) in the temperature range below 160 K. The onset temperature for χ_c is apparently shifted to higher temperature with increasing pressure. The results indicate that the onset temperature for the interlayer canted antiferromagnetic spin correlation goes up with increasing pressure. On the other hand, a more conspicuous change is observed for the pressure dependence of χ_c maximum. At ambient pressure, a sharp maximum of χ_c is observed at $T_N \sim 60$ K as an indication of the onset of the antiferromagnetic interplane spin-arrangement. With increasing pressure, the anomaly is essentially shifted down in temperature, somewhat broadened and reduced in magnitude. Compared with the resistivity data, the suppression of peak structure in χ_c may have an intimate connection to the enhancement of 2D character below T_c. The steep drop of ρ_{ab} below

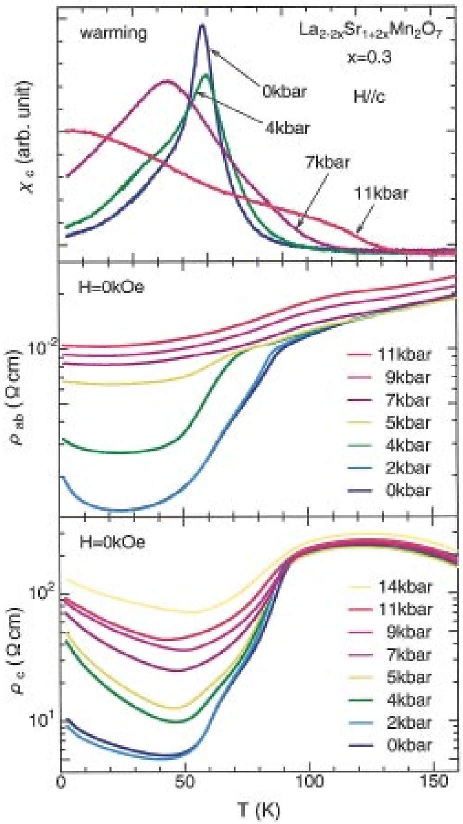

FIG. 3(color). Temperature dependence of c-axis ac susceptibility χ_c (top), ρ_{ab} (middle), and ρ_c (bottom) at several pressures in the $La_{2-2x}Sr_{1+2x}Mn_2O_7$ ($x = 0.3$) crystal.

T_c is reduced with the increase of pressure. Under the pressure of 11 kbar the ρ_{ab} shows no drop around T_c but dull temperature dependence characteristic of the 2D metallic state (like the one above T_c at 0 kbar) down to the lowest temperature. A similar feature is observed in the pressure effect on ρ_c. These results suggest that the 3D spin-ordered state at low temperatures is suppressed and changes to a more 2D-like ferromagnetic state by applying pressure. In other words, the (inherently short range) 2D spin ordering is likely to extend down to the lowest temperature under high pressures, which sensitively affects the charge transport, as observed.

Although the accurate knowledge about the spin structure must be obtained by neutron scattering measurements, we may have, from the aforementioned results, the following scenario for the interrelation between the interplane magnetic coupling and the charge transport. At ambient pressure, the interlayer canted spin correlation between adjacent MnO_2 bilayers evolves at $T_c \sim 90$ K. With further decrease of temperature, the antiferromagnetic (AF) interplane coupling appears at $T_N \sim 60$ K, dominating over the interplane ferromagnetic (F) coupling. The low-temperature phase of this system may consist of mostly AF and some F static order between the adjacent MnO_2 bilayers. The interlayer coupling, in particular the AF one, is appreciably suppressed by application of pressure, which results in the realization of the 2D-like electronic and magnetic system down to low temperatures.

For an origin of the pressure effect on the interlayer coupling, we may need to consider the orbital degrees of freedom of the e_g-like conduction electrons. Since the orbital characters strongly couple with lattice, the structural change by the application of pressure may reflect on the change in the orbital state. In a $La_{2-2x}Sr_{1+2x}Mn_2O_7$ ($x = 0.4$) sample, Argyriou *et al.* [9] observed a considerably larger change in the Mn-O apical bond length than that in the Mn-O inplane bond length of MnO_6 octahedron by the application of hydrostatic pressure. The results suggest that the application of hydrostatic pressure changes the orbital state through such an anisotropic change in the Mn-O bond length in bilayered manganites. Recently, Ishihara *et al.* [13] have studied theoretically the pressure effect on the spin and orbital states in the bilayered manganite, taking account of the anisotropic change in the Mn-O bond length. Considering the energy difference between $3d_{x^2-y^2}$ and $3d_{3z^2-r^2}$ orbitals in the pressure-deformed lattice, they showed that the applied pressure can weaken the interlayer charge and spin couplings through the stabilization of the $3d_{x^2-y^2}$ orbital. The change of the orbital character may thus be responsible for the observed pressure effect on the interlayer coupling in the bilayered manganite.

The field dependencies of ρ_{ab} and ρ_c have been measured with $H \parallel c$ at 4.2 K under several pressures, as displayed in the upper and lower panels of Fig. 4, respectively. For these measurements, the crystals were slowly cooled from room temperature to 4.2 K at 70 kOe, and then magnetic fields were swept cyclically. Both the ρ_{ab} and ρ_c rapidly decrease with increasing magnetic fields, and become nearly constant above the saturation field $H_{\mathrm{sat}} \sim 2$ kOe. The large difference between the interplane (ρ_c) and inplane (ρ_{ab}) MR at ambient pressure, $[\rho_c(0\ \mathrm{kOe}) - \rho_c(8\ \mathrm{kOe})]/\rho_c(8\ \mathrm{kOe}) \sim 490\%$ for the CPP (current-perpendicular-plane) geometry vs $[\rho_{ab}(0\ \mathrm{kOe}) - \rho_{ab}(8\ \mathrm{kOe})]/\rho_{ab}(8\ \mathrm{kOe}) \sim 25\%$ for the CIP (current-in-plane) geometry, has been ascribed to the presence of the interplane TMR process for the CPP configuration [8]. Namely, the c-axis transport of the spin-polarized electron is blocked at the insulating block accompanying the AF-type coupling between the adjacent MnO_2 bilayers, but the magnetization process removes such AF-coupling boundaries and allows the interplane tunneling of spin-polarized electrons. In addition, we have observed the nonlinear I-V characteristics in ρ_c [14], suggestive of a FM/I/FM tunneling process.

The most striking feature presently observed is that the magnitude of the interplane MR is drastically enhanced up to ~4000% by applying pressure (Fig. 4). This is because ρ_c values at zero field remarkably increase by high pressure while remaining nearly pressure independent at $H \geq H_{\mathrm{sat}}$ [see and compare Fig. 3(a) with (b)]. H_{sat} is increased slightly (up to ≈4 kOe) with the increase of pressure. Taking account of the above discussion, such an

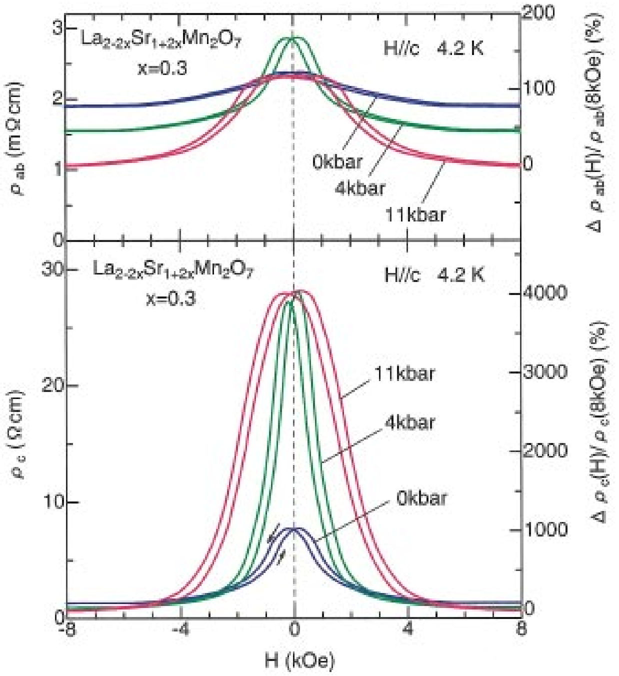

FIG. 4(color). Inplane resistivity ρ_{ab} (upper) and interplane resistivity ρ_c (lower) as a function of a magnetic field with $H \parallel c$ at 4.2 K under several pressures in the $La_{2-2x}Sr_{1+2x}Mn_2O_7$ ($x = 0.3$) crystal. The measurements were performed after the sample was once cooled under 70 kOe. The scale of the right ordinate is the MR ratio for 11 kbar data, as given by $[\rho(0\text{ kOe}) - \rho(8\text{ kOe})]/\rho(8\text{ kOe})$.

enhanced MR effect can be closely related to the reduction of the interplane magnetic coupling. The application of pressure appears to suppress not only the interplane F coupling but also the interplane AF one, in contrast to the previous observation in magnetic multilayers [15] that the interlayer AF coupling enhances the CPP-MR effect. The weakly pressure-dependent ρ_c at $H \geq H_{\mathrm{sat}}$ suggests that the pressure effect on ρ_c is small in the absence of the magnetic domain boundaries. By contrast, at low temperatures ($\leq T_c$) ρ_c shows strong pressure enhancement at zero field. This indicates that conduction electrons moving along the c axis in the pressure-induced 2D FM state suffer from even stronger scattering than in the weakly AF-coupled state at ambient pressure.

In the real material, magnetic domains are present within each ferromagnetic MnO_2 bilayer below T_N at ambient pressure. The regions of the interplane AF alignment are therefore incorporated with local regions of the interplane F alignment. The regions of the interplane F alignment which remain as defects in the interplane AF alignment, are hence likely to reduce ρ_c and lead to the relative decrease in the interplane MR. (The microscopic current path for the measurement of ρ_c must be highly inhomogeneous.) The suppression of the interplane coupling by applying pressure, may decouple these defects of the F alignment, and cause the increase of the resistivity at zero field. At $H \geq H_{\mathrm{sat}}$ the decoupled spin domain of each MnO_2 bilayer is aligned along the field direction, which can be viewed as the transition from the 2D ferromagnetic (paramagnetic along the c axis) to the 3D ferromagnetic state. In fact, such an electronic dimensional crossover is also manifested in an enhanced inplane MR (CIP-MR) under pressures (see the upper panel of Fig. 4). This arises from the fact that the 2D FM state is a highly diffuse metal as seen in ρ_{ab}-T curves at 11 kbar of Fig. 3. This enhanced inplane MR thus reflects the deconfinement transition of the spin-polarized carrier, being reminiscent of the spin-valve effect [16].

In summary, we have performed magnetic and transport measurements under pressures to investigate the effect of the interplane coupling in layered manganites, $La_{2-2x}Sr_{1+2x}Mn_2O_7$ ($x = 0.3$) with the quasi-2D ferromagnetic metallic state. In this system, the antiferromagnetic interplane coupling is dominant at low temperatures under ambient pressure. However, the application of pressure has enabled us to systematically weaken the interplane magnetic coupling, making the charge-transport more 2D-like. As a result, the interplane tunneling MR is enhanced up to ~4000% at ≈10 kbar and 4.2 K, reflecting the field-induced 2D-3D crossover of the ferromagnetic metallic state. This is because the 2D-like conduction of the nearly fully spin-polarized carriers within the magnetically interplane-decoupled MnO_2 bilayers is highly diffuse or incoherent perhaps due to the coupling with phononic and/or orbital excitations.

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