Two-Dimensional Charge-Transport and Spin-Valve Effect in the Layered Antiferromagnet $Nd_{0.45}Sr_{0.55}MnO_3$

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Below the Nèel temperature (T_N) a layered antiferromagnetic (AF) $Nd_{0.45}Sr_{0.55}MnO_3$ crystal is metallic within the ferromagnetic (F) layers (*ab* plane), while insulating along the AF-coupling direction (*c* axis), showing an extremely large anisotropy in resistivity. The observed two dimensionality of charge dynamics is due to confinement of the spin-polarized carriers within the F sheets induced by the magnetic ordering as well as orbital ordering (perhaps of $d_{x^2-y^2}$ type). Large negative magnetoresistance is observed over a wide temperature region below T_N, which arises from the dimensional crossover due to the field-induced spin canting in the AF state. [S0031-9007(99)09183-8]

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Since the late rediscovery of the colossal magnetoresistance (CMR) phenomena in perovskite-type manganites, exotic phenomena arising from strong coupling among spin, charge, and orbital degrees of freedom have been attracting much interest. In spite of the vast literature on CMR manganites, however, only a few studies have so far been made on transport properties of overdoped manganites (hole-doping level $x > 0.50$). This is partly because the ferromagnetic (F) transition temperature T_c is maximal at $x = 0.3$–0.4, in which CMR is attained at relatively high temperatures at about T_c. On the other hand, the electronic phase diagrams in a wide region of the x-T plane have been reported for $(La, Sr)MnO_3$ [1], $(La, Ca)MnO_3$ [2], $(Pr, Ca)MnO_3$ [3], and so on, and unusually rich aspects in spin-charge-orbital coupled physics have begun to be unraveled. Among them, $Nd_{1-x}Sr_xMnO_3$ can be obtained in a form of high-quality crystals with a wide range of x and hence is suitable for investigating a doping dependence of the magneto-transport properties. Prior to this study, we obtained the electronic and magnetic, as well as crystal-structural, phase diagram for the $Nd_{1-x}Sr_xMnO_3$ system as a function of x ($0.30 \leq x \leq 0.80$) [4–6]. In particular, a recent neutron scattering study on the metallic compound $Nd_{0.45}Sr_{0.55}MnO_3$ has revealed that its magnetic structure is layered (*A*-type) antiferromagnetic (AF), in contrast with the F metallic state below $x < 0.50$, and the charge/orbital-ordered insulating state at $x = 0.50$ [6]. Similar *A*-type AF metallic states have recently been reported for other perovskite manganites [7]. According to the double-exchange (DE) model [8,9], the e_g conduction electron transfer t_{ij} is governed by the relation as $t_{ij} = t_0 \cos(\theta_{ij}/2)$, where θ_{ij} means the relative angle between t_{2g} spins in the neighboring sites. Therefore, t_{ij} along the AF direction is ideally expected to be zero. (The apparently metallic conduction observed for the randomly oriented samples of the *A*-type AF manganite may reflect the admixture of the conducting F layers.) From this viewpoint, we have investigated anisotropic transport properties of the *A*-type AF phase for $Nd_{0.45}Sr_{0.55}MnO_3$ crystal at various temperatures down to ~30 mK.

Optical [10], photoemission [11], and tunneling [12] spectroscopies have provided the experimental evidence for the half-metallic density of states in the F phase of perovskite manganites. Namely, the conduction band in the F state is largely split into majority and minority subbands separated by the on-site Hund's-rule energy (~1–2 eV). Therefore, electrons in the ground states are perfectly spin polarized. Tunneling magnetoresistance (TMR) based on such a highly spin-polarized character of the conduction e_g electrons has recently been demonstrated for various manganite-based systems, including trilayer junctions [13], granular polycrystals with artificial [14] and natural [15] grain boundary, and Ruddledsen-Popper phases [16]. In this context, the present layered-AF crystal of $Nd_{0.45}Sr_{0.55}MnO_3$ provides a unique opportunity for studying interplane TMR phenomena along the AF-coupling direction. In fact, we have observed 2D charge dynamics with nearly full spin polarization as well as magnetic-field-induced dimensional crossover giving rise to a gigantic TMR-like effect.

Crystals of $Nd_{0.45}Sr_{0.55}MnO_3$ were grown by the floating zone method, details of which were published in preceding papers [17]. Inductively coupled plasma spectrometry on the grown crystal indicated that the stoichiometry is nearly identical to the prescribed ratio with an accuracy of x within ± 0.01. Powder x-ray diffraction (XRD) apparatus equipped with a temperature-controllable cryostat was used to check the crystal quality and to determine the lattice parameters. Rietveld refinement of XRD patterns for the pulverized crystal indicated that all peaks are indexed without impurity phase. In order to investigate the anisotropic charge-transport properties, we have carefully cut out the samples with the edges along the pseudocubic principal directions from the melt-grown ingot, which was based on the results of the x-ray back Laue measurements: The cutting accuracy is about 0.5° or better. To eliminate a possible multidomain structure,

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we have prepared a fairly small-sized crystal, typically $1.0 \times 3.0 \times 0.1$ mm^3 or less. Resistivity measurements were performed by using the four-probe method and with the ac resistance bridge. Magnetotransport measurements in low-temperature regions down to about 30 mK were carried out in the cryostat equipped with a ^{3}He-cycle-type dilution refrigerator and with a 10-T superconducting magnet. The temperatures below 1 K are monitored by using the calibrated RuO_2 resistance thermometer. To ensure the accurate thermometry in low temperatures, we have paid maximal attention as follows: The small sample was very slowly cooled down to the He dilution temperature region, enough to maintain the thermal equilibrium to the cooling head attached to the thermometer, and used the low-excitation current to prevent the Joule heating.

Figure 1 shows the magnetization, lattice parameter, and resistivity as a function temperature for a single crystal of $Nd_{0.45}Sr_{0.55}MnO_3$. The increase of the magnetic moment above the Nèel temperature (T_N) is due to the F correlations which were evidenced by a recent inelastic neutron scattering study [18]. Such a F fluctuation is quenched at $T_N = 220$ K and sharply changed to the AF fluctuation with the layered (A-type) magnetic structure (illustrated in the inset in the top panel of Fig. 1). The thermal hysteresis was observed due to the first-order nature of this AF transition, as clearly seen in the ρ-T curves in Figs. 1 and 2. In the A-type AF state below T_N, on the other hand, the macroscopic anisotropy of magnetization was confirmed: The parallel magnetization along the [110] (or, equivalently, $[1\bar{1}0]$) direction is smaller than that of the perpendicular [001] direction. This is characteristic of the layered AF structure with the lateral spin direction, although the magnetic anisotropy appears fairly small in the present manganite.

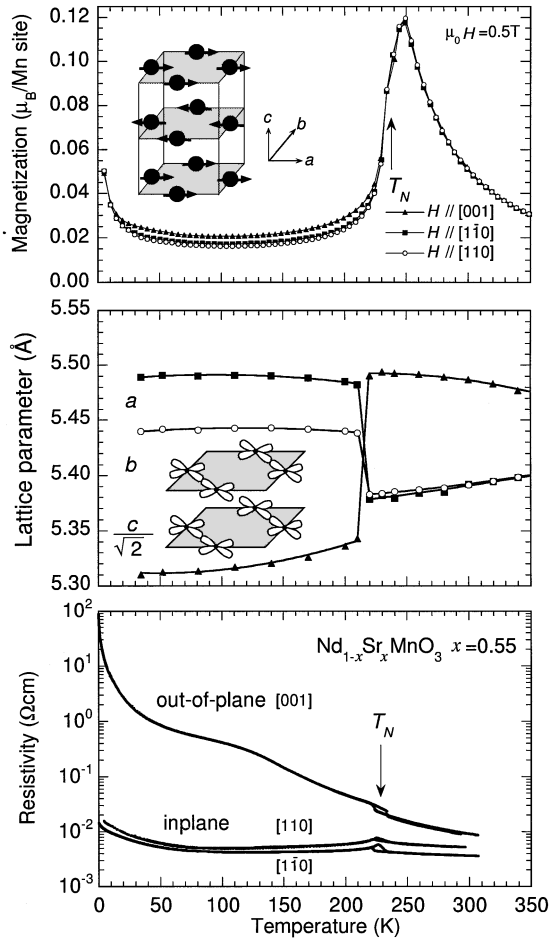

FIG. 1. Temperature dependence of magnetization (top), lattice parameters (middle), and resistivity (bottom) for a single crystal of $Nd_{0.45}Sr_{0.55}MnO_3$. Schematics of the layered (A-type) AF structure and $d_{x^2-y^2}$-type orbital ordering are illustrated in the top and middle panels, respectively. Magnetization was measured at a field of 0.5 T after cooling down the sample to 5 K in zero field.

Corresponding to the layered AF spin structure, the highly anisotropic electrical property was observed (bottom panel of Fig. 1). The metallic behavior was observed within the F layers (ab plane) although the resistivity turns to increase below 80 K. By contrast, along the AF-coupling direction (c axis), the crystal remains insulating over the whole temperature region. The anisotropy ratio of resistivity of the AF to F direction, ρ_c/ρ_{ab}, is $\sim 10^4$ at 35 mK (see also Fig. 3). This value is merely a lower bound for the anisotropy, since the local defects resulting in the multidomain structure cannot thoroughly be eliminated in the present nearly cubic crystal. The observed large anisotropy, in spite of the nearly cubic lattice structure, is due to the magnetic confinement of the spin-polarized carriers within the F sheets. No spin canting along the AF direction observed in this system indicates that carriers are confined within the F plane, and

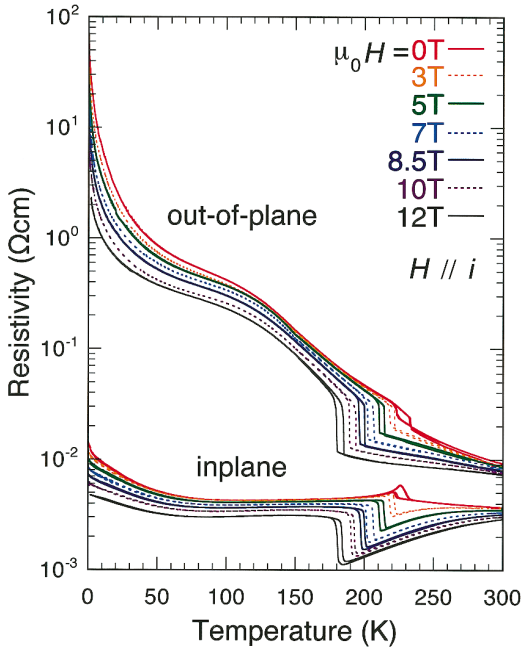

FIG. 2(color). Temperature dependence of resistivity in various H for in-plane (ab) and out-of-plane (c) directions in the $Nd_{0.45}Sr_{0.55}MnO_3$ crystal. H was applied parallel to both the current and the respective crystal axis.

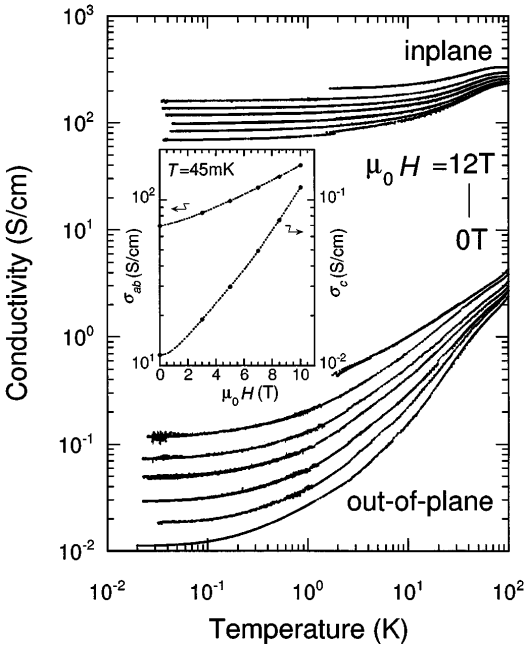

FIG. 3. Temperature dependence of conductivity along the in-plane (ab) and the out-of-plane (c) directions for the $Nd_{0.45}Sr_{0.55}MnO_3$ crystal in several H ($\mu_0 H$ = 0, 3, 5, 7, 8.5, 10, and 12 T). Note that the ordinate and the abscissa are both on a logarithmic scale. The inset shows the conductivity of each direction as a function of H at 45 mK.

the DE mechanism along the AF direction is quenched. This makes a sharp contrast to the case of the A-type AF states in the lightly doped $La_{1-x}Sr_xMnO_3$, in which a small number of mobile holes produce spin canting via $d_{3z^2-r^2}$ states along the AF-coupling direction [1,9].

In accordance with these magnetic and electronic transitions, the crystallographic structural change occurs: The orthorhombic $O^{\ddagger}$ structure with the lattice constants $a \sim b < c/\sqrt{2}$ changes to O′ with $c/\sqrt{2} < b < a$ below T_N, and, consequently, the ab (F) plane expands and the c axis (AF) shrinks (middle panel of Fig. 1). These changes in lattice parameters imply that the $d_{x^2-y^2}$-type orbitals lie in the F plane and the transfer interaction along the AF direction almost vanishes (depicted in the inset in the middle panel of Fig. 1). Namely, the $d_{x^2-y^2}$ orbitals are stabilized to maximize the transfer interaction or the kinetic exchange of carriers within the ab plane while fully keeping the AF superexchange interaction between the t_{2g} local spins along the c axis. A recent theoretical calculation based on the mean field approximation [4] also predicts that coherent (uniform) ordering of $d_{x^2-y^2}$ orbitals should be realized in the A-type spin structure in overdoped manganites, in contrast to the $d_{3x^2-r^2}/d_{3y^2-r^2}$-type orbital ordering observed in the insulating parent material $LaMnO_3$ with the same A-type spin structure [19,20]. The existence of the cooperative or competitive order parameters, such as F, AF, and orbital-ordering (or collective Jahn-Teller) interactions, may result in the first-order spin-lattice coupled phase transition as observed. In addition, no sign of superlattice due to the orbital ordering is observed within the F plane below T_N by electron-beam [21] and neutron [18] diffraction measurements, being also consistent with the above mentioned orbital-ordering accompanying no charge ordering: The orbital arrangement is not necessarily static and the carriers can move only by using the $d_{x^2-y^2}$ orbitals.

We show in Fig. 2 the magnetic field (H) effect on the anisotropic charge-transport properties. The AF-transition temperature T_N can be decreased with an external H. This is due to the stabilization of the F state by application of H, which causes negative MR above T_N as a result of the suppression of spin scattering. Over a wide temperature region below T_N, the large negative MR was also observed, especially for the AF direction. The H forcedly aligns opposite spins in the adjacent ab planes to the H direction, which results in the spin canting (see the schematic in the inset of Fig. 4). This causes an increment of the transfer interaction along the AF-coupling direction or at least removes the carrier confinement due to the collinear AF coupling. Such MR phenomena below T_N will be discussed in detail by using the results of the isothermal scans (Fig. 4).

Let us turn to the anisotropic magnetotransport at very low temperatures (Fig. 3). As clearly seen in the figure, the conductivity in the F plane (σ_{ab}) decreases gently and saturates below ~1 K. On the other hand, conductivity along the AF direction (σ_c) decreases with the decrease of

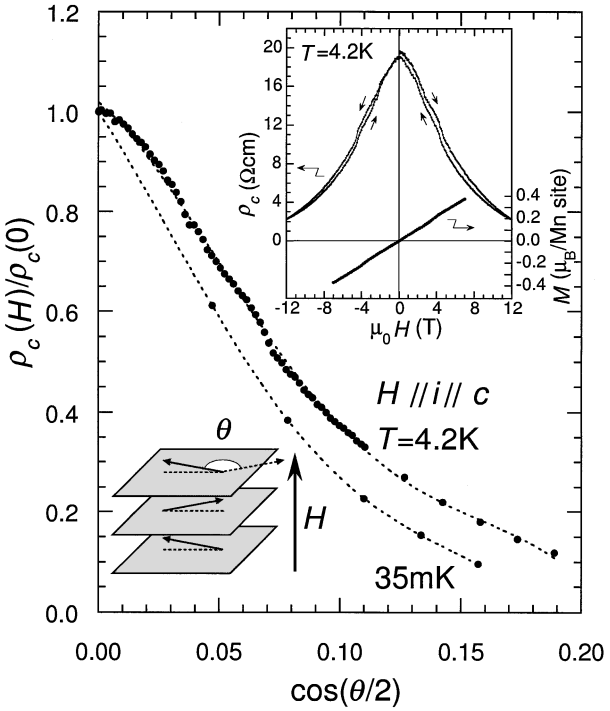

FIG. 4. Normalized magnetoresistance, $\rho_c(H)/\rho_c(0)$, for the c-axis direction in the $Nd_{0.45}Sr_{0.55}MnO_3$ crystal as a function of $\cos(\theta/2)$, where θ is the relative angle between the ordered spins on the neighboring ab planes. The θ values are determined by the M-H curves at 4.2 K. The data of 35 mK are plotted from the ρ-T curves in Fig. 2, provided that the M-H curve is identical with that at 4.2 K. The inset shows isothermal magnetoresistance (left ordinate) and corresponding M-H curve (right ordinate) at 4.2 K for the same sample.

temperature even below 1 K and then saturates below 100 mK. This saturation at an extremely low value ($\approx 10^{-2}$ S/cm) might be suspected to be due to the leak of conducting current along the in-plane direction (ab) which might be contained in the out-of-plane direction (c), namely due to the local lattice defects leading to a multidomain structure. However, σ_c is observed to increase by an order of magnitude upon application of H (≥ 10 T) even at the lowest temperature (35 mK) of the present study. The field increase of σ_{ab} is much less than σ_c, as seen in the inset of Fig. 3, and hence the simple scenario of the admixture of the in-plane component to the c-axis component is not easily acceptable. Another possibility is that the ordered orbitals are not exactly of $d_{x^2-y^2}$ type but have the $d_{3z^2-r^2}$ component due to the tilting of MnO_6 octahedra in the actual orthorhombically distorted lattice structure. The ground state of this system was found to be a diffuse metal with an extremely large anisotropy. Under high H, the anisotropy σ_{ab}/σ_c decreases conspicuously, e.g., ≈ 7000 in zero field to 1300 in 10-T field at 45 mK, and the system is more 3D-like due to the increase of σ_c.

We show in Fig. 4 the isothermal MR for the c-axis direction at a low temperature (4.2 K) as a function of $\cos(\theta/2)$. Here, θ represents the relative angle between the ordered spins on the neighboring ab planes and was deduced from the magnetization curve (the inset of Fig. 4) by assuming that the field-induced spin canting gives the observed magnetization: Namely, $M = M_s \cos(\theta/2)$, $M_s \approx 3.45\ \mu_B$/Mn being the saturated magnetization. The large negative isothermal-MR, e.g., $\rho_c(H)/\rho_c(0) \approx 0.1$ at 12 T and 4.2 K, was observed (the inset of Fig. 4) as expected from the ρ-T curves in the field (Figs. 2 and 3). The MR effect is nearly isotropic with respect to the H direction in spite of the anisotropic spin structures, but in accordance with the nearly isotropic M-H curves for both the parallel and the perpendicular H directions to the F plane. The observed MR arises from quite a different mechanism from that of the conventional MR near T_c in the manganites. In the present case of the A-type AF phase, a small F moment appears to induce such a large MR, as observed. For example, the spin-canting angle is about 10° in the field of 12 T at 4.2 K. Such a field-induced spin canting should give rise to the revival of the electron hopping along the AF-coupling direction. Without explicit consideration of the orbital degree of freedom, the revived hopping interaction along the c axis would be given by the relation that $t^{(c)} \propto \cos(\theta/2)$, as in the simple DE model. In this context, the layered AF state of the present compound may be viewed as an infinite ordered stack of the spin valves. In fact, the large MR was observed in a wide temperature range below T_N, in contrast to the conventional MR occurring in a limited temperature window near T_c. Here, it is worthwhile to recall the twofold nature of the 2D carrier confinement; one is the AF-coupled spin structure along the c axis, and the other is the "ferromagnetic" ordering of $d_{x^2-y^2}$-type orbitals. Therefore, the fairly large c-axis TMR implies the dimensional crossover due to the field-induced canting of not only the spin but also the orbital, namely, the field-induced admixture of a $d_{3z^2-r^2}$-type orbital with a majority $d_{x^2-y^2}$-type orbital.

In summary, we have revealed anisotropic magnetotransport properties in pseudocubic perovskite manganite, $Nd_{0.45}Sr_{0.55}MnO_3$ crystal, with the layered AF spin structure. The observed huge anisotropy implies, together with the result of recent theoretical consideration, the magnetic and orbital-ordering-induced confinement of the spin-polarized carriers within the F plane. The change in lattice parameters at T_N suggests that the $d_{x^2-y^2}$-type orbitals uniformly ("ferromagnetically") order in the F plane, and the transfer interaction along the AF direction is quenched. Moreover, relatively large negative MR was observed over a broad range of temperature below T_N, which arises from the dimensional crossover due to the spin canting of AF ordering induced by a magnetic field and subsequent interplane tunneling of the spin-polarized carrier. By analogy of GMR (giant MR) effects for the AF coupled metallic superlattices [22], this layered AF state of perovskite-type manganite may be viewed as a prototype for the intrinsic spin-valve system.

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