

Correlation of the magnetic and magnetotransport properties in a $\text{Nd}_{1.5}\text{Sr}_{1.5}\text{Mn}_2\text{O}_7$ single crystal

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A detailed study of the anisotropic magnetic and magnetotransport behavior has been performed on a single crystal of the layered perovskite $\text{Nd}_{1.5}\text{Sr}_{1.5}\text{Mn}_2\text{O}_7$. The magnetic properties are consistent with an antiferromagnetic (AFM) ordering below $T_N=190$ K of the manganese moments within the ab plane and a moment reorientation around ≈ 125 K towards the c direction. A spin-glass behavior develops for $T < 50$ K attributed to the freezing of the magnetic moment of the Nd(1) ions placed within the perovskite blocks, whereas the Nd(2) ions in the rocksalt layer contribute to the observed AFM order. An additional ferromagnetic contribution is observed below 275 K, ascribed to the existence of intergrowths. Low-field magnetoresistance (MR) is observed for $H \parallel c$ in both ρ_c and ρ_{ab} , reaching values up to 30% at $T=10$ K. It is associated with the reduction of the spin disorder of the Mn moments aligned parallel to the c axis. For higher-field values the sharp decrease in MR is correlated with the magnetic “flipping” transitions. Within the whole temperature range the magnetoresistance is anisotropic, depending on both the direction of the applied field and current. Evidence is provided against the existence of spin-polarized tunneling between the layers in this compound. [S0163-1829(99)03232-4]

INTRODUCTION

Perovskite manganites (AMnO_3 , with A = alkaline ion and/or rare earth ion) have been extensively studied in the last few years because they show a spectacularly large variation of their electric resistance under an applied magnetic field. The so-called “colossal magnetoresistance” (CMR) occurs in the vicinity of the ferromagnetic (FM) ordering temperature of the manganese spins. Nevertheless, large values of the magnetic fields are required to produce this effect, and more investigations needed in order that these compounds can be used in technological applications. The need for new materials exhibiting enhanced MR effects at low fields has recently motivated the investigation of the layered perovskite compounds.¹ These compounds can be considered variants of the cubic perovskite, where n layers of MnO_6 octahedra are separated along the c axis from the next n layers by an insulating block consisting of a single AO layer with the rocksalt structure (see Fig. 1). The three-dimensional perovskite structure and its layered variants belong to the Ruddlesden-Popper (RP) series, whose general formula can be written as $A_{n+1}\text{Mn}_n\text{O}_{3n+1}$ or also $(\text{AMnO}_3)_n\text{AO}$, the three-dimensional perovskite corresponding to $n=\infty$. The reduced dimensionality of the Mn-O-Mn networks in the layered compounds can introduce important modifications in the magnetic and magnetotransport properties. It is only within the MnO_2 layers, which lie within the ab plane, that the double-exchange mechanism can be expected. As for the $n=\infty$ perovskites, the magnetic ordering in the other RP manganites is accompanied by a metal-insulator transition, but the low-field MR (LFMR) effect is clearly enhanced. Nevertheless, this improvement of the MR

properties is intrinsically related to the lowering of the metal-insulator transition temperature.²

Enhanced LFMR was first observed in manganite single crystals in $\text{La}_{1.2}\text{Sr}_{1.8}\text{Mn}_2\text{O}_7$ (Ref. 1) and in $\text{La}_{1.4}\text{Sr}_{1.6}\text{Mn}_2\text{O}_7$ (Ref. 3) (layered variant $n=2$). In the last system the interplane MR is observed to be extremely large. It has been

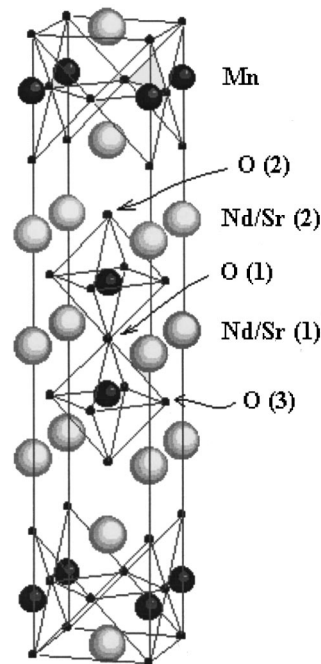


FIG. 1. Crystal structure of $\text{Nd}_{1.5}\text{Sr}_{1.5}\text{Mn}_2\text{O}_7$, belonging to the Ruddlesden-Popper series with $n=2$, displaying the different sites for the ions.

proposed that this layered perovskite behaves as a kind of multilayered system, in the sense that a current flowing perpendicular to the ab plane has to cross all the magnetic layers, maximizing the spin-dependent scattering of the carriers.³ Anisotropic transport and the appearance of two- and three-dimensional ordering are also considered a consequence of the reduced dimensionality of these systems. Arguments for^{1,4-6} and against⁷ the existence of short-range two-dimensional (2D) spin correlations in the paramagnetic phase have been reported. Whether the 2D FM is an intrinsic property of the layered compounds is still a matter for discussion. Some authors attribute this ordering to the existence of *intergrowths*.^{8,9} These intergrowths are an intrinsic defect in the RP series which is due to the disorder in the stacking sequence of the perovskite layers and the AO planes. It becomes more important for high values of n , i.e., a higher number of contiguous perovskite layers connected together which form the structural block of the cell.¹⁰

A strong effort has also been made to understand the CMR properties observed in the Nd-Sr series.¹¹⁻¹⁶ The Nd-Sr compounds do not exhibit FM ordering close to the insulator-metal transition, and, thus, the CMR observed cannot readily be explained by the double-exchange interaction. All the studies performed until now on this series have used polycrystalline samples. However, the anisotropy exhibited by the layered structures makes it desirable to study these compounds in single-crystalline form. In this paper we present the results of magnetization and magnetotransport properties on a single crystal of Nd_{1.5}Sr_{1.5}Mn₂O₇. A detailed discussion of the results is also presented.

EXPERIMENT

A polycrystalline powder of Nd_{1.5}Sr_{1.5}Mn₂O₇ was prepared from the high-purity precursors SrCO₃, Nd₂O₃, and MnO₂ using the solid state reaction method. The thoroughly mixed oxide precursors were fired at 1200 °C for 24 h with intermediate grinding. The powder was isostatically pressed into the form of rods and sintered at 1350 °C for 24 h. A single crystal was grown in air using the floating-zone technique with an IR image furnace (NEC, SC-N35HD) equipped with two halogen lamps and double hemiellipsoidal mirrors. The growth rate used was 7 to 8 mm h⁻¹ with feed and seed rods rotated in opposite directions at 30 rpm.

A large single-crystalline grain of approximate length 50 mm and diameter 8 mm was obtained. The as-grown crystal could be cleaved parallel to the growth direction, yielding the (001) surface. A piece was cut parallel to the symmetry directions in order to perform the magnetic and magnetotransport characterization in the temperature range 4 K < T < 300 K. Magnetization measurements were carried out using a commercial superconducting quantum interference device (SQUID) magnetometer (up to 5 T). The resistivity was measured using the four probe method with silver paint contacts on the sample using a steady magnetic field up to 12 T. During the measurements the sign of the current was reversed and the resulting absolute value averaged, in order to eliminate the thermoelectric voltage and the voltage due to misalignment of contacts. The magnetoresistance ratio in the present work is defined as $MR = 100 \times [\rho(H) - \rho(0)] / \rho(0)$,

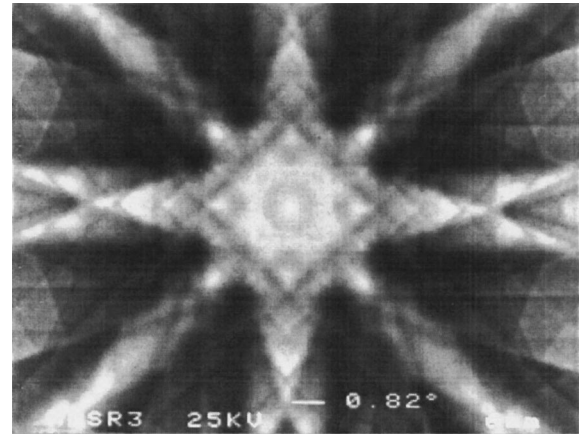


FIG. 2. Reflection electron diffraction pattern obtained with the incident beam perpendicular on the cleaved surface of the crystal. Well defined Kikuchi lines and bands can clearly be observed.

where $\rho(0)$ and $\rho(H)$ are the zero-field resistivity and the resistivity in an applied magnetic field, respectively.

RESULTS AND DISCUSSION

According to x-ray powder diffraction analysis the structure is consistent with a body-centered tetragonal unit cell ($I4/mmm; Z=2$). The lattice parameters at room temperature are $a = b = 3.8263 \text{ \AA}$ and $c = 20.3141 \text{ \AA}$. The layered nature of the compound is reflected in the powder x-ray spectra, which was observed to display features characteristic of a strong preferential orientation, making a detailed refinement of the structure difficult. Secondary phase peaks, corresponding to an $n = \infty$ perovskite, were detected in the pattern. The origin and relevance of this impurity phase will be discussed later, together with the magnetization and magnetotransport results. Electron-probe microanalysis (EPMA) revealed spatial homogeneity of the sample. X-ray Laue diffraction was performed along the length of the sample to check the orientation and the degree of crystallinity of the material. A four-fold symmetry was observed on the Laue pattern obtained for the incident beam perpendicular on the cleaved surface of the crystal, indicating that the growth direction is perpendicular to the crystallographic c axis. In Fig. 2 we show a Kikuchi electron diffraction pattern obtained by scanning electron microscopy (SEM) on the cleaved surface. A very good quality of crystallinity is necessary to obtain this kind of pattern.

In Fig. 3(a) we display the field cooling (FC) and zero-field cooling (ZFC) thermal dependencies of the magnetization measured for a magnetic field of 0.05 T applied along the c direction, M_c , and in a direction within the ab plane, M_{ab} . With cooling from room temperature we observe a sharp FM-like increase at $T^* \approx 275 \text{ K}$ for both M_{ab} and M_c magnetizations; a smooth increase of M_{ab} , with maximum slope around $T_1 \approx 200 \text{ K}$; a smooth increase of M_c , with maximum slope around $T_2 \approx 125 \text{ K}$, which is close to a freezinglike temperature for M_{ab} ; and a freezinglike temperature for the M_c magnetization at $T_3 \approx 50 \text{ K}$. We will see later in the paper that all these features are correlated with the transport behavior displayed in Fig. 3(b). A sharp increase of the magnetization with lowering of temperature around 300 K

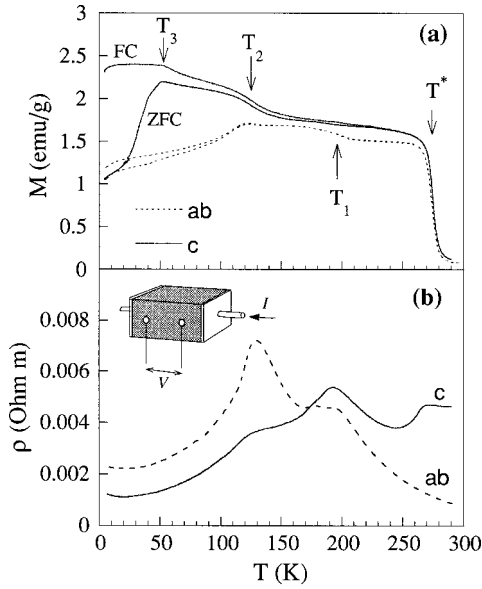


FIG. 3. Temperature dependence of the magnetization (a) and resistivity (b), measured in warming-up mode: (a) magnetization after zero-field cooling (ZFC) and after cooling in field (FC) with $H=0.05$ T applied along the c direction (solid line) and along a direction within the ab plane (dashed line); (b) resistivity at zero field for a current applied perpendicular (ρ_c) and parallel (ρ_{ab}) to the crystallographic ab plane. The electrode setup is also displayed.

has also been observed on polycrystalline samples with a composition similar to the studied one,^{15,16} but no sign of bulk FM was detected in those works. In order to understand the nature of the FM-like ordering occurring at T^* in our compound we have measured the magnetization isotherms along different crystallographic directions using a SQUID magnetometer. The results obtained are displayed in Fig. 4.

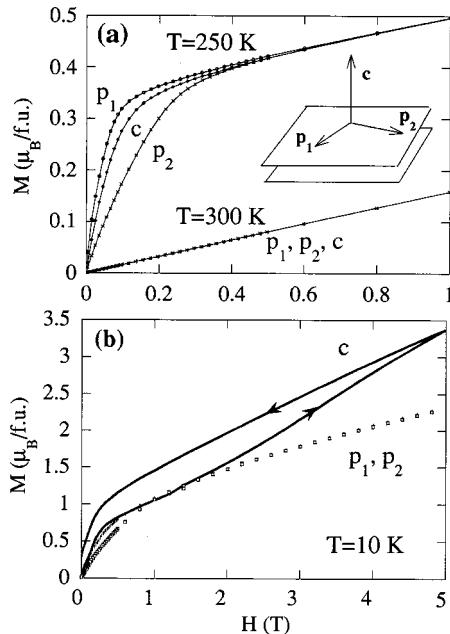


FIG. 4. Magnetization isotherms along three different crystallographic directions. The magnetization has been measured for $H\parallel c$, and H applied along two equivalent crystallographic directions (p_1, p_2) perpendicular to each other within the ab plane.

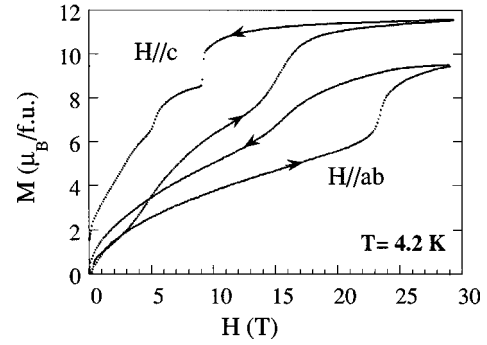


FIG. 5. Magnetization measurements performed in the pulsed high magnetic field installation at $T=4.2$ K for the field applied along the c direction, M_c , and in a direction within the ab plane, M_{ab} . The arrows indicate increasing or decreasing field mode.

The magnetization has been measured with H parallel to c and for H applied along two perpendicular directions p_1 and p_2 within the ab plane, which should be crystallographically equivalent in tetragonal symmetry. If this ordering is intrinsic to the compound, in keeping with the tetragonal symmetry, no anisotropy should be expected within the ab plane. At 300 K, temperature above the magnetic transition, we observe the expected paramagnetic (PM) behavior, the magnetization has a linear dependence with the applied field and no dependence on the direction of H . For $T=250$ K, below the FM transition, we observe an anisotropic behavior in all the three selected directions for low-field values ($H<0.4$ T). Thus, the two perpendicular directions p_1 and p_2 in the ab plane are not magnetically equivalent, reflecting a behavior which does not conform to tetragonal symmetry. As no structural change with temperature decrease has been observed in the $\text{Nd}_{2-2x}\text{Sr}_{1+2x}\text{Mn}_2\text{O}_7$ series,¹³ we conclude that this FM ordering is not intrinsic to the layered $n=2$ compound, and its possible origin will be discussed later. The magnetic behavior observed at $T=250$ K in the displayed field range corresponds to a FM magnetization superimposed on a PM magnetization curve. At lower temperatures the compound is intrinsically ordered, and at $T=10$ K instead of the linear field dependence superimposed on the FM saturation, a metamagnetic transition is observed for the magnetization measured parallel to the c axis [Fig. 4(b)]. The FM contribution to the magnetization observed is $\approx 0.5\mu_B$ at this temperature.

In order to determine the relevance of the secondary phase FM contribution to the magnetic properties we need to determine the value of the saturation magnetization of the whole system at the same temperature. For this reason we have performed magnetization measurements in pulsed high magnetic fields up to 30 T, whose detailed analysis will be presented elsewhere.¹⁷ The compound reveals an intrinsic antiferromagnetic behavior with field-induced metamagnetic transitions due to “flipping” of the moments from antiparallel to parallel configuration. In Fig. 5 we display the results for $M_c(H)$ and $M_{ab}(H)$ obtained at $T=4.2$ K. For $H\parallel c$ these transitions are induced at lower fields than for the field applied within the ab plane. For $H\parallel c$ two metamagnetic transitions are observed, the first around 3.4 T and a sharper one at 13.8 T, after which saturation is reached. For $H\parallel ab$ a sharp metamagnetic transition takes place at ≈ 22.8 T. The

highest saturation value of magnetization ($\approx 11.5\mu_B$) is obtained for $H\parallel c$. The saturation value obtained for $H\parallel ab$ is much lower than this, indicating that the canting of moments is still present at the maximum field for this direction. With decreasing magnetic field a large hysteresis of the order of 7 T is observed in both curves. For $H\parallel c$ the magnetization drops in two steps, sharply at 9 T and in a smoother way around 5.0 T. The decrease of magnetization along this direction has also been observed to happen in a single step, the shape of the curve for decreasing fields varying from measurement to measurement, which indicates that the system enters a metastable state. At high temperatures where the FM of the secondary phase is present, only Mn moments can contribute to the signal, and we estimate the secondary phase contribution to be less than 9%.

According to the above discussion, the compound displays an intrinsic paramagnetic behavior at high temperature and an AFM configuration at 4.2 K. It has also been observed that the magnetization vs field measurements performed at different temperatures can be explained if we assume that the AFM ordering already exists at temperatures higher than 100 K. Neutron diffraction results on polycrystalline samples of $\text{Nd}_{2-2x}\text{Sr}_{1+2x}\text{Mn}_2\text{O}_7$ with higher Sr content ($x=0.5, 0.45$) have revealed a long-range antiferromagnetic ordering of the Mn ions at 140 K along the ab plane.¹² For these compositions, the Nd(2) ions in the rocksalt site order along the c direction at 30 K, producing either a complete (for $x=0.45$) or partial rotation (for $x=0.5$) of the Mn sublattice towards the same direction. The Nd(1) ions placed within the perovskite blocks are magnetically frustrated, forming a spin-glass (SG) layer. This picture is consistent with our results on $\text{Nd}_{1.5}\text{Sr}_{1.5}\text{Mn}_2\text{O}_7$, where the Mn ions would order within the ab plane at a higher temperature, $T_1=200$ K, giving the extra contribution observed in the M_{ab} vs T curve [see Fig. 3(a)]. No extra contribution is observed along the c direction in the temperature range $T_2 < T < T_1$, for which the magnetization starts to increase around 125 K. At this temperature the component of the magnetization parallel to the ab plane decreases, indicating that reorientation of the Mn moments from the plane to the perpendicular direction is taking place. Below T_3 , M_c decreases. The rotation of the Mn moments from the ab plane towards the c direction at T_2 has been ascribed previously to the appearance of a magnetic moment on the Nd ions at this temperature.¹² As the rare earth ion has a strong magnetocrystalline anisotropy it would force the rotation of the Mn moments. However, if this is the case for our compound, the deduced temperature for the ordering of the Nd moments, ≈ 125 K, is relatively high and thus the origin for this moment reorientation is still not clear.

The fact that we observe an intrinsic small contribution to the bulk magnetization for relatively low-field values, allowing us to observe all the features described in the $M(T)$ curve at 200 K and below, is readily explained assuming that the AFM ordering in this compound is not perfect. It is reasonable to expect that as in three-dimensional perovskites there exists competition between FM double-exchange and AFM superexchange interactions. As a consequence, canted AFM or weak FM order can appear with decreasing x , as suggested by others.¹⁶ Another possible explanation can be given assuming the existence of uncompensated moments in

an AFM matrix arising due to defects. Nevertheless, it has been demonstrated that even in perfect AFM superlattices the presence of domain walls can produce a finite moment that can be measured at vanishing field.¹⁸ A weak contribution to the magnetization arising from the presence of domain walls cannot be disregarded.

We have thus concluded that $\text{Nd}_{1.5}\text{Sr}_{1.5}\text{Mn}_2\text{O}_7$ not only exhibits intrinsic AFM ordering at low temperatures but also an additional FM ordering at high temperature close to 300 K. A similar transition has also been observed in single-crystalline samples of $\text{La}_{1.2}\text{Sr}_{1.8}\text{Mn}_2\text{O}_7$.¹⁹ Surprisingly, in the La-Sr system which exhibits an intrinsic FM ordering at 100 K, neutron scattering experiments have not been able to detect any Mn moments above this temperature.²⁰ The magnetic transition observed around 300 K in the La-Sr compounds has been attributed in the works of Potter *et al.*⁸ and Bader *et al.*⁹ to the two-dimensional FM ordering of trace amounts of intergrowths whose volume fraction might be too small to be detected by neutron scattering, or whose magnetic scattering length might be too short to be detected. Seshadri *et al.* have demonstrated the presence of intergrowths in these materials using transmission electron microscopy (TEM).²¹ Taking into account these results, it is reasonable to attribute the FM ordering observed in our sample around 275 K to the existence of defects in the crystal structure.

The temperature dependence of the resistivity at zero field for a current (I) applied perpendicular to the ab plane (ρ_c) and parallel to it (ρ_{ab}) is shown in Fig. 3(b), where the electrode setup used is also displayed. The following features can be observed: in the whole temperature range both ρ_{ab} and ρ_c show the same order of magnitude; for sufficiently high temperatures the conduction is activated, with a resistivity which increases with decreasing temperature; two broad maxima can be observed in both the $\rho_{ab}(T)$ and $\rho_c(T)$ curves at temperatures $T=190$ and 130 K; and a sharp decrease of ρ along the c direction is observed around $T^*=275$ K, but no effect can be detected at this temperature for the resistivity measured along the ab plane.

In the layered compounds it should be expected that the conductivity measured for the current applied within the ab plane is much higher than for the current applied perpendicular to the perovskite layers. This two-dimensional behavior has in fact been observed on $\text{La}_{2-2x}\text{Sr}_{1+2x}\text{Mn}_2\text{O}_7$ with $x=0.4, 0.3$,^{1,2} where the value of ρ_c is between two and three orders of magnitude larger than ρ_{ab} . The similar order of magnitude observed here for ρ_{ab} and ρ_c is a reflection of the three-dimensional character of the transport properties in $\text{Nd}_{1.5}\text{Sr}_{1.5}\text{Mn}_2\text{O}_7$. In order to understand the puzzling behavior described above, we have proceeded to study the magnetic properties of this layered manganite.

As noted before, the magnetic properties show a strong correlation with the transport properties in this compound. Comparing Figs. 3(a) and 3(b) the different features observed with decreasing temperature can be related in the following way.

(1) The steep decrease of resistivity along the c direction taking place at T^* is related to the FM-like ordering observed at this temperature. The FM ordering, attributed to the existence of intergrowths, induces a reduction of the spin scattering disorder along the c direction. No decrease is ob-

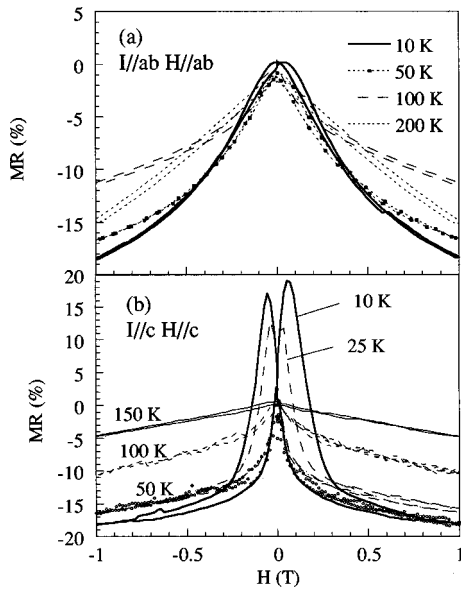


FIG. 6. MR measurements under magnetic field up to 1 T at some selected temperatures for two configurations of current and field, as indicated in the figures.

served at this temperature in the resistivity within the ab plane, indicating that there is no change on the correlation of the moments in the direction parallel to the planes at the magnetic transition. This result can be understood assuming a distribution of point or small defects spread within the ab plane.

(2) The cusps observed in both the ρ_{ab} and ρ_c curves at temperatures 190 and 130 K are related to the intrinsic magnetic ordering temperatures T_1 [anomaly observed in $M_{ab}(T)$] and T_2 [observed in $M_c(T)$]. The AFM ordering of the Mn moments along the ab plane at T_1 produces a smooth reduction of the spin scattering, leading to the corresponding decrease of the resistivity. Such a decrease occurs for both current directions indicating that the observed AFM ordering is of three-dimensional nature. Going down in temperature, when T_2 is approached, the Mn moments rotate from the ab plane to the c axis. The resistivity increase in this range can be explained assuming a noncoherent rotation of the moments. With decreasing temperature the ordering is stronger and the spin scattering is reduced, leading to a monotonous decrease of resistivity. The freezing of the Nd moments at temperature T_3 does not, as expected, produce any change on the conductivity of the system.

Isothermal MR measurements have been performed at different temperatures in the range 4–300 K under magnetic fields up to 12 T. Some of the MR curves show the existence of LFMR, whose origin is different from the mechanism giving rise to the MR observed at higher-field values. We will analyze first the low-field behavior. In order to determine the LFMR effect of the compound a set of low-field ($H < 1$ T) measurements has been performed, using a sweep rate of 0.2 T/min. To avoid initial history-dependent effects the initial increase and decrease of the field was carried out for all the isotherms, which will not be displayed in the figures. Figures 6(a) and 6(b) show the MR isotherms at some selected temperatures, measured for H applied along the ab plane parallel to the current, i.e., $H \parallel ab$ $I \parallel ab$ (a), and for $H \parallel c$

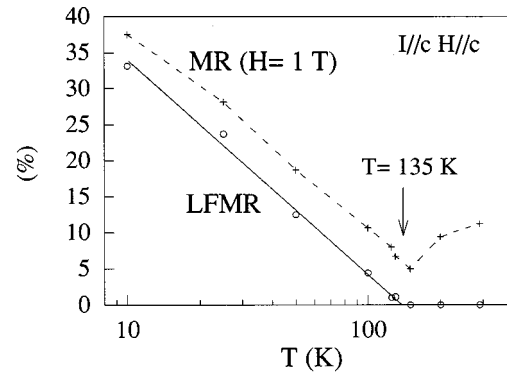


FIG. 7. Absolute value of the MR vs temperature at $H=1$ T (crosses) and LFMR (circles) measured with the current applied along the c direction for $H \parallel c$. Note the logarithmic scale used for the temperature axis. The continuous line represents a linear fit of the LFMR data for $T < 150$ K. The arrow indicates the temperature at which the fit crosses the ordinate axis. The dashed line is a guide to the eye.

$I \parallel c$ (b). The results obtained for $I \parallel ab$ $H \parallel c$ are observed to be qualitatively very similar to those for $I \parallel c$ $H \parallel c$. In these figures we note that, while for the measurements with $H \parallel ab$ the MR curves show a smooth variation in the whole temperature range, the MR obtained for $H \parallel c$ is characterized by the presence of a sharp decrease at low-field values $H < 0.5$ T (LFMR). There is a crossover from the high-temperature behavior, $T > 150$ K, where no LFMR is observed, to the low-temperature range with a LFMR component. Below 50 K the MR loops adopt the characteristic *butterfly shape*, with increasing *hysteresis* with decreasing temperature. As the magnetic field increases, ρ increases sharply, showing a peak at particular field value. With a further increase in H the resistance drops quickly and displays a saturation behavior. We have estimated the absolute value of the LFMR at each temperature considering the MR at the peak minus the value at the technical saturation of the curves. The LFMR contribution increases logarithmically as the temperature is lowered, as displayed in Fig. 7, reaching a value higher than 30% at 10 K. This logarithmic increase of the LFMR represents a radically different behavior with respect to the one observed on single-crystalline $\text{La}_{1.4}\text{Sr}_{1.6}\text{Mn}_2\text{O}_7$,³ where the LFMR for the current running perpendicular to the layers is nearly T independent, and also from the temperature dependence observed in thin films of La-Ca layered perovskites.²² We can exclude the possibility that the intergrowths which are ferromagnetic below T^* are contributing to the LFMR, as no qualitative change has been observed comparing the low-field MR isotherms taken at $T=290$ K—above T^* —and at $T=200$ K—below T^* (not displayed in the figure). If the LFMR was associated with the spontaneous FM magnetization it should be observed also in the range 150–275 K, which is not the case. This contribution has to be associated with an intrinsic ordering of the compound as will be noted later.

A peculiar feature of the magnetotransport properties of the $\text{Nd}_{1.5}\text{Sr}_{1.5}\text{Mn}_2\text{O}_7$ single crystal can be observed by comparing the MR loops obtained for different directions of the applied field and current, as displayed in Fig. 8 for $T=10$ K. It is commonly accepted that the value of the magnetic field at which the LFMR peaks corresponds to the co-

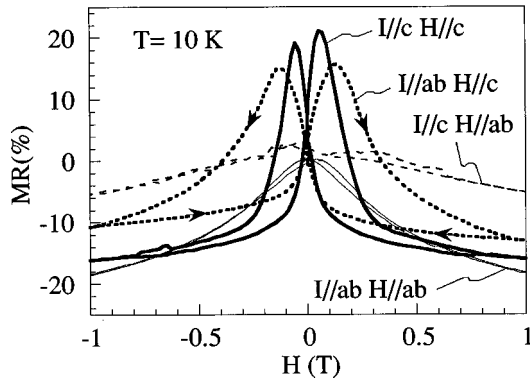


FIG. 8. MR isotherms measured at 10 K for different directions of the applied current and field up to 1 T.

ervecive field (H_C) of the system (zero magnetization),²³ and presents saturationlike behavior at the field value where magnetization reaches technical saturation. However, we should note that in our measurements on $\text{Nd}_{1.5}\text{Sr}_{1.5}\text{Mn}_2\text{O}_7$ the MR peak for $I\parallel c H\parallel c$ occurs at a smaller magnetic field value than for $I\parallel ab H\parallel c$. The loops are wider for the latter measurement and saturation is also reached at higher-field values. In contrast to bulk samples, epitaxial thin films of three-dimensional perovskite also exhibit dependence of the MR on the angle between the current and applied field.²⁴ The difference in the MR-peak field value observed here cannot be ascribed to differences in the sample shape that could lead to different demagnetizing field values, as the coercive field and the MR peak would not be affected. The complete disorder of the moments along the c direction for increasing $H\parallel c$ deduced from the measurement with $I\parallel c$ is reached at 580 Oe, which is within the experimental error in agreement with the H_C obtained at the same temperature in magnetization measurements (500 Oe). Nevertheless, from the MR results on $I\parallel ab H\parallel ab$ we deduce that the moments are still correlated along the ab plane at this field value, and the resistivity has not reached the maximum. In this way, the peak observed in the MR curves cannot not be associated to a zero-magnetization state of the bulk compound. The complete disorder of the moments along the ab plane is only reached for a higher-field value, 1280 Oe but, at this field, the moments are correlated along the c direction. From the higher-field value observed of the MR peak with $I\parallel ab$ we conclude that the magnetic moments in this system are more correlated within the ab plane than with respect to the c direction. This anisotropy in the MR is thus an intrinsic effect associated to the layered nature of the compound.

The LFMR is negligible for $I\parallel c H\parallel ab$ and, although the MR measured with $I\parallel ab H\parallel ab$ drops relatively fast with increasing field, the lack of hysteresis in all these curves at low temperature reveals a completely different nature for the LFMR obtained in the measurements with $H\parallel ab$ with respect to those with $H\parallel c$. Thus, the low-field changes observed in the MR for $H\parallel ab$ have probably the same origin to those observed at higher-field values. Returning now to Fig. 7 we would like to note that the LFMR contribution appears at some temperature below 150 K. The linear fit of the thermal decrease displayed in this figure cuts the ordinate axis at $T \approx 135$ K, close to the temperature at which the Mn moments reorient towards the c direction. The LFMR effect is

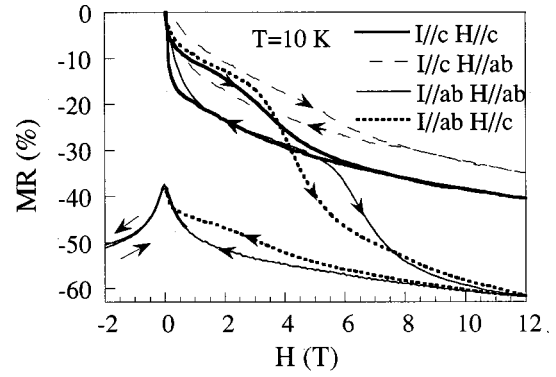


FIG. 9. MR isotherms measured at $T=10$ K for different directions of the applied field and current up to 12 T. The arrows indicate increasing or decreasing field mode.

associated in this compound with the reduction by the field of the disorder of the spins away from the c axis. A field $H\parallel c$ reduces this disorder more effectively than $H\parallel ab$, which is easily understood by taking into account the axial anisotropy of the system in this temperature range. Both the conduction perpendicular and parallel to the layers show this effect and thus the tunneling of carriers between different layers separated by insulating rocksalt layer, used to explain the observed results in the La-Sr system, cannot be applied here.

From Fig. 8 we note that the loops are unexpectedly *asymmetric*. In principle, there are no factors that can break the symmetry with respect to the sign of the applied field direction. A similar kind of asymmetry has also been detected in measurements on spin-valve systems. It has been observed that a deviation in the direction of the applied field of only $\theta=0.1^\circ$ from the perpendicular direction to the layers can lead to an apparent asymmetric field dependence in the MR loops.²⁵ Such an effect has been explained taking into account the geometrical anisotropy accompanied by a large demagnetizing field. In this respect the system is behaving like a spin valve for low values of the applied field.

In Fig. 9 we display the MR vs H curves measured at $T=10$ K for different directions of the applied field and the current in fields up to 12 T. The MR at high fields is clearly correlated to the changes in the initial AFM configuration of the cell. The magnetic field induces a canting of the AFM spins in the direction parallel to it. This canting is produced in a smooth manner for some field ranges and by jumps when some critical field values are reached. The clearest transition occurs at $H_c^* \approx 2.5$ T for $H\parallel c$, and $H_{ab}^* \approx 5.5$ T for $H\parallel ab$. The transition at H_c^* , although less pronounced, is also clearly observed in the magnetization measurements displayed in Figs. 4(b). Nevertheless no sharp change in M_{ab} is observed at H_{ab}^* . Magnetoresistance measurement is thus a more sensitive tool than the magnetization for detecting the changes in the configuration of the moments for the field applied along the plane. A final remark should be made about the hysteretic behavior observed in these high-field MR measurements, where we observe a very large hysteresis in the MR obtained with $I\parallel ab$ and a negligible hysteresis for $I\parallel c$. This indicates that after a field loop (increase followed by decrease of the field) the system returns to the initial spin correlations state only for I along the c direction, whereas the resistivity remains low at $H=0$ for $I\parallel ab$, indicating that

strong spontaneous spin correlations are present within the *ab* plane.

CONCLUSIONS

We have presented the magnetic and transport study on a $\text{Nd}_{1.5}\text{Sr}_{1.5}\text{Mn}_2\text{O}_7$ single crystal. The compound displays an intrinsic PM behavior at temperatures higher than $T \approx 200$ K, with an additional FM contribution for $T < 275$ K ascribed to the presence of intergrowths. The intrinsic magnetic behavior with decreasing temperature can be explained assuming the AFM ordering of the Mn moments within the plane, with a spontaneous spin reorientation transition around 125 K towards the *c* direction. Under high magnetic fields “flipping” transitions are induced at low temperature, taking place at 13.8 T for $H \parallel c$ and at 22.8 T for $H \parallel ab$. We would like to note that these transitions are associated with very large changes in the lattice, as observed in high-field magnetostriction measurements.²⁶

The features observed in the magnetoresistance are strongly correlated with the magnetic behavior. At low temperatures large changes are observed upon field-induced “flipping” of the AFM configuration. Within the whole temperature range the magnetoresistance is anisotropic, depending on the direction of the applied field and current. Low-field magnetoresistance is observed below 150 K in the MR isotherms measured with $H \parallel c$. It increases logarithmically with decreasing temperature, and gives rise to hysteretic butterfly-shaped loops for $T < 50$ K. This contribution is present in both ρ_c and ρ_{ab} resistivities with the same order of magnitude, which excludes the existence of spin-polarized tunneling between the layers in this compound. The LFM is associated with the reduction of the spin scattering of the moments with respect to the *c* axis. The large hysteresis observed for the high-field magnetoresistance measurements with the current applied perpendicular to the *c* axis reflects the presence of strong spin correlations along the *ab* plane at low temperatures.

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¹Y. Moritomo, A. Asamitsu, H. Kuwahara, and Y. Tokura, *Nature* (London) **380**, 141 (1996).

²R. Mahesh, R. Mahendiran, A. K. Raychaudhuri, and C. N. R. Rao, *Solid State Chem.* **122**, 448 (1996).

³T. Kimura, H. Tomioka, H. Kuwahara, A. Asamitsu, M. Tamura, and Y. Tokura, *Science* **274**, 1699 (1996).

⁴J. B. MacChesney, J. F. Potter, and R. C. Sherwood, *J. Appl. Phys.* **40**, 1243 (1969).

⁵D. N. Argyriou, T. M. Kelley, J. F. Mitchell, R. A. Robinson, R. Osborn, S. Rosenkranz, R. I. Sheldon, and J. D. Jorgensen, *J. Appl. Phys.* **83**, 6374 (1998).

⁶T. G. Perring, G. Aeppli, Y. Moritomo, and Y. Tokura, *Phys. Rev. Lett.* **78**, 3197 (1997).

⁷R. H. Heffener, D. E. MacLaughlin, G. J. Nieuwenhuys, T. Kimura, G. M. Luke, Y. Tokura, and Y. J. Uemura, *Phys. Rev. Lett.* **81**, 1706 (1998).

⁸C. D. Potter, Maribeth Swiatek, S. D. Bader, D. N. Argyriou, J. F. Mitchell, J. Miller, D. G. Hinks, and J. D. Jorgensen, *Phys. Rev. B* **57**, 72 (1998).

⁹S. D. Bader, R. M. Osgood, D. J. Miller, J. F. Mitchell, and J. S. Jiang, *J. Appl. Phys.* **83**, 6385 (1998).

¹⁰C. N. R. Rao and B. Raveau, *Transition Metal Oxides* (Wiley-VCH, New York, 1998).

¹¹P. D. Battle, S. J. Blundell, M. A. Green, W. Hayes, M. Honold, A. K. Klehe, N. S. Laskey, J. E. Millburn, L. Murphy, M. J. Rosseinsky, N. A. Samarin, J. Singleton, N. E. Sluchanko, S. P. Sullivan, and J. F. Vente, *J. Phys.: Condens. Matter* **8**, L427 (1996); P. D. Battle, M. A. Green, N. S. Laskey, J. E. Millburn, L. Murphy, M. J. Rosseinsky, S. P. Sullivan, and J. F. Vente, *Chem. Mater.* **9**, 552 (1997); P. D. Battle, M. A. Green, N. S. Laskey, N. Kasmir, J. E. Millburn, L. E. Spring, S. P. Sullivan, M. J. Rosseinsky, and J. F. Vente, *J. Mater. Chem.* **7**, 997 (1997).

¹²P. D. Battle, M. A. Green, N. S. Laskey, and J. E. Millburn, *Phys. Rev. B* **54**, 15 967 (1996).

¹³P. D. Battle, S. J. Blundell, D. E. Cox, M. A. Green, J. E. Millburn, P. G. Radaelli, M. J. Rosseinsky, J. Singleton, L. E.

Spring, and J. F. Vente, in *Solid-State Chemistry of Inorganic Materials*, edited by A. K. Davies, A. J. Jacobson, C. C. Torardi, and T. A. Vanderah, MRS Symposia Proceedings No. 453 (Materials Research Society, Pittsburgh, 1997), p. 331.

¹⁴P. D. Battle, N. Kasmir, J. E. Millburn, M. J. Rosseinsky, R. T. Patel, L. E. Spring, S. J. Blundell, W. Hayes, A. K. Klehe, A. Mihut, and J. Singleton, *J. Appl. Phys.* **83**, 6379 (1997).

¹⁵N. H. Hur, Jin-Tae Kim, K. H. Yoo, Y. K. Park, and J.-C. Park, *Phys. Rev. B* **57**, 10 740 (1998).

¹⁶Jun Zhang, Fangwei Wang, Panlin Zhang, Xiandong Sun, and Qiwei Yan, *J. Magn. Magn. Mater.* **190**, 166 (1998).

¹⁷B. García-Landa, M. R. Ibarra, M. Hilbers, P. A. Algarabel, A. del Moral, G. Balakrishnan, M. R. Lees, and D. McK Paul (unpublished).

¹⁸N. Papanicolaou, *J. Phys.: Condens. Matter* **10**, L131 (1998).

¹⁹J. F. Mitchell, D. N. Argyriou, J. D. Jorgensen, D. G. Hinks, C. D. Potter, and S. D. Bader, *Phys. Rev. B* **55**, 63 (1997).

²⁰D. N. Argyriou, J. F. Mitchell, J. B. Goodenough, O. Chmaissem, S. Short, and J. D. Jorgensen, *Phys. Rev. Lett.* **78**, 1568 (1997).

²¹R. Seshadri, M. Hervieu, C. Martin, A. Maignan, B. Domenges, B. Raveau, and A. N. Fitch, *Chem. Mater.* **9**, 1778 (1997).

²²H. Asano, J. Hayakawa, and M. Matsui, *Phys. Rev. B* **57**, 1052 (1998).

²³See, for instance, R. M. Bozorth, *Ferromagnetism* (van Nostrand, New York, 1951), p. 745; M. B. Stearns, in *High Temperature ordered Intermetallic Alloys III*, edited by C. T. Liu, A. I. Taub, N. S. Stoloff, and C. C. Koch, MRS Symposia Proceedings No. 133 (Materials Research Society, Pittsburgh, 1989), p. 553; J. M. D. Coey, A. E. Berkowitz, L. Balcells, F. F. Putris, and F. T. Parker, *Appl. Phys. Lett.* **72**, 734 (1998).

²⁴J. N. Eckstein, I. Bozovic, J. O'Donnell, M. Onellion, and M. S. Rzechowski, *Appl. Phys. Lett.* **69**, 1312 (1996); J. O'Donnell, M. Onellion, M. S. Rzechowski, J. N. Eckstein, and I. Bozovic, *Phys. Rev. B* **55**, 5873 (1997).

²⁵H. Sato, S. Miya, Y. Kobayashi, Y. Aoki, H. Yamamoto, and M. Nakada, *J. Appl. Phys.* **83**, 5927 (1998).

²⁶B. García-Landa, C. Marquina, M. R. Ibarra, M. Hilbers, G. Balakrishnan, M. R. Lees, and D. McK Paul (unpublished).