Magnetic properties and CMR effect in layer type manganite LaSr$_2$Mn$_2$O$_7$ under high magnetic fields

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Abstract. Magnetoresistance and magnetization were measured for single crystals of LaSr$_2$Mn$_2$O$_7$ utilizing a nondestructive long-pulse magnet over the whole temperature region below room temperature. Application of a magnetic field gradually changes an antiferromagnetic insulating state into a forced ferromagnetic metallic state. It was found that the change of resistivity is irreversible, but no ferromagnetic domain is generated by external magnetic field.

The discovery of colossal magnetoresistance (CMR) in perovskite manganites R$_{1-x}$A$_x$MnO$_3$ (R and A being trivalent rare-earth and divalent alkaline-earth ions, respectively) has attracted considerable attention recently [1–3]. Distorted perovskite manganites such as La$_{1-x}$Sr$_x$MnO$_3$ undergo a transition from an insulating antiferromagnetic state to a ferromagnetic metallic state by doping carriers [2]. However, in some of the R$_{1-x}$A$_x$MnO$_3$ system with the ionic radii for the R and A sites being relatively small, such as La$_{1-x}$Ca$_x$MnO$_3$ and Pr$_{1-x}$Ca$_x$MnO$_3$, near $x \sim 1/2$, a charge-ordered (CO) state appears, which suppresses the ferromagnetic metallic state [4]. In ordinary charge-ordering transitions, ordering of the e$_g$ orbitals is also associated, which favours the charge-exchange-(CE-) type antiferromagnetic (AF) spin ordering [5]. These orderings are observed not only in cubic-type perovskite manganites, but also in layered-type perovskite manganites, such as La$_{1-x}$Sr$_{1+x}$MnO$_4$ ($x = 0.5$) [6].

Application of magnetic fields causes the destruction of the charge ordering. Corresponding to the charge disordering, magnetization saturates, and insulator–metal transition occurs. This transition has been observed in many CO systems [7, 8]. For the case of La$_{1-x}$Sr$_{1+x}$MnO$_4$ ($x = 0.5$), however, the temperature dependence of the resistivity shows insulator-like behaviour; nevertheless the magnetization jumps to the saturated state on applying magnetic field [9].

Recent research revealed that in some perovskite manganites no CO state is observed even if the resistivity shows insulator-like temperature dependence. For example, Pr$_{1-x}$Sr$_x$MnO$_3$ ($x = 0.5$), in which the resistivity increases with decreasing temperature at low temperature, shows no signature of the CO phase. This compound forms the a-type layered AF structure instead of the CE-type antiferromagnetic spin ordering, but A-type

layered AF structure. As a result, anisotropy of transport has been observed [10]. As the response to magnetic fields is very similar to that of the CO system, it had been believed so far that it was also a CO system.

In $LaSr_2Mn_2O_7$, which is one of the layered type perovskite manganites, charge ordering appears below around 220 K [11]. However, on decreasing temperature further, the CO state disappears. Battle *et al* assert that the ground spin structure of this system is an A-type layered AF structure [12]. However, taking into account experimental results for other compounds, it is deduced that CE-type AF spin ordering may be stable in the CO phase.

In the present work, we have investigated the magnetic and transport properties of $LaSr_2Mn_2O_7$ under high magnetic fields up to 40 T. Measurements of magnetization and magnetoresistance have been carried out over a wide temperature range. On the basis of these results, we deduce the magnetic structure of $LaSr_2Mn_2O_7$.

A single crystal of $LaSr_2Mn_2O_7$ was grown by the floating zone method. Inductively coupled plasma mass spectrometry was carried out to check the cation ratio of the crystal. The result revealed that the ratio was almost in accord with the prescribed one within an accuracy of 1%.

Using a nondestructive long-pulse magnet energized by a 300 kJ capacitor bank, we generated pulsed magnetic fields up to 40 T with a duration time of 20 ms. To remove the residual effects of magnetic fields, we increased the temperature up to room temperature and decreased the temperature monotonically down to the stabilized temperature just before shooting each magnetic field pulse. Magnetization measurements were carried out by the induction method using a couple of coaxial pickup coils. The inner coil had a diameter of 4.2 mm and 320 turns, while the outer coil had a diameter of 6.0 mm and the number of turns was adjusted to cancel the voltage induced in the inner coil by the magnetic fields. The voltage induced in the pickup coils was integrated numerically to obtain magnetization. Transport measurements were carried out by the standard four-probe method with a dc current. The directions of magnetic fields were parallel to the c axis, and those of the current were parallel (ρ_{ab}) and perpendicular (ρ_c) to the MnO_2 bilayers.

Typical magnetic field–resistivity (R–H) curves are shown in figure 1. In this figure, the direction of current was parallel to the MnO_2 bilayers. Above $T_{CO} \sim 220$ K, there is no hysteresis in the R–H curve, and the MR ratio is relatively small. However, below T_{CO}, a giant negative magnetoresistance appears with large hysteresis. Although the change of resistivity is giant, there is no jump of resistivity. The resistance after applying magnetic field is always smaller than that before applying it. The size of hysteresis strongly depends on the duration time, especially around 160 K. From the R–H measurements shown in figure 1, some points are picked up and replotted as the temperature dependence of resistivity under magnetic fields (figure 2). All of the points adopted are at increasing magnetic fields. With increasing magnetic field, the temperature dependence of the resistivity becomes metallic, which is different from that of a 214 system such as $La_{0.5}Sr_{1.5}MnO_4$ [9]. The magnetoresistance with a current perpendicular to the MnO_2 bilayers (ρ_c) is shown in figure 3. Most of the features are the same as the curve for ρ_{ab}.

Magnetization curves are shown in figure 4. For the case of the magnetic field direction parallel to the c axis (M_c), above T_{CO}, M increases monotonically without hysteresis with increasing H, and dM/dH decreases with increasing H. Below T_{CO}, on the other hand, there is an area where dM/dH decreases with increasing H, and a small hysteresis appears in the M–H curve. This curve is very similar to that of $La_{0.5}Sr_{1.5}MnO_4$ [9]. However, on decreasing temperature further, the hysteresis disappears. Such features were observed also in the case where the magnetic field direction is perpendicular to the c axis (M_{ab}),
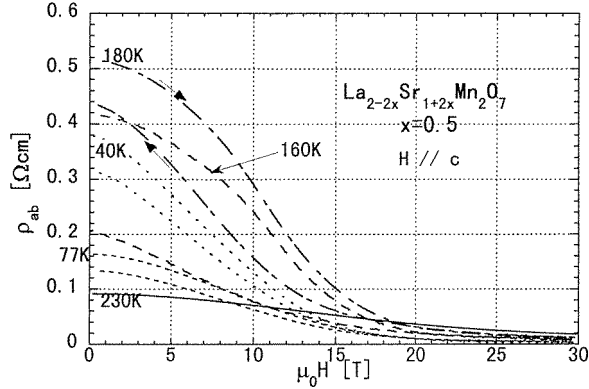
Figure 1. Magnetoresistance of LaSr$_2$Mn$_2$O$_7$ with a current parallel to the MnO$_2$ bilayers. Magnetic fields are perpendicular to the bilayers ($H \| c$).
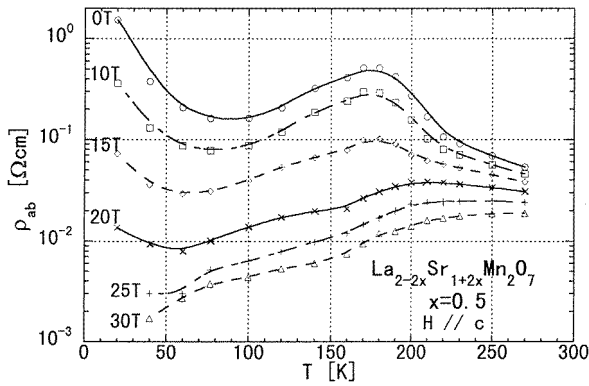

Figure 2. Temperature dependence of resistivity under magnetic fields with a current parallel to the MnO$_2$ bilayers. Magnetic fields are perpendicular to the bilayers ($H \| c$).

although the size of hysteresis was different. However, for H parallel to the ab plane, the hysteresis tends to disappear at around 80 K, and the hysteresis becomes larger again with decreasing temperature. At around 4 K the hysteresis is enhanced and M shows a jump around 25 T as a first-order transition. It is worth noting that, for both increasing and decreasing H, M shows antiferromagnetic properties around 0 T from room temperature to liquid He temperature.

As mentioned before, the resistivity for both directions after applying magnetic field is lower than before for both directions. If we suppose that the orbital structure of the e$_g$ electrons was always tetragonal and the reduction of resistivity was caused by the change of orbital character (say from $x^2 - y^2$ to $3z^2 - r^2$, or vice versa), the result of the experiment would not be explained, because such orbital change should bring the opposite effect to resistivities for each direction. If ferromagnetic domains appear in the sample after applying the magnetic field, the change of the resistivity might be explained. However, in the $M-H$ curve we can see no ferromagnetic features.

Now we propose an assumption that without magnetic field, the charge-ordered CE-type AF phase and the charge-disordered A-type AF phase coexist, and that the former phase is
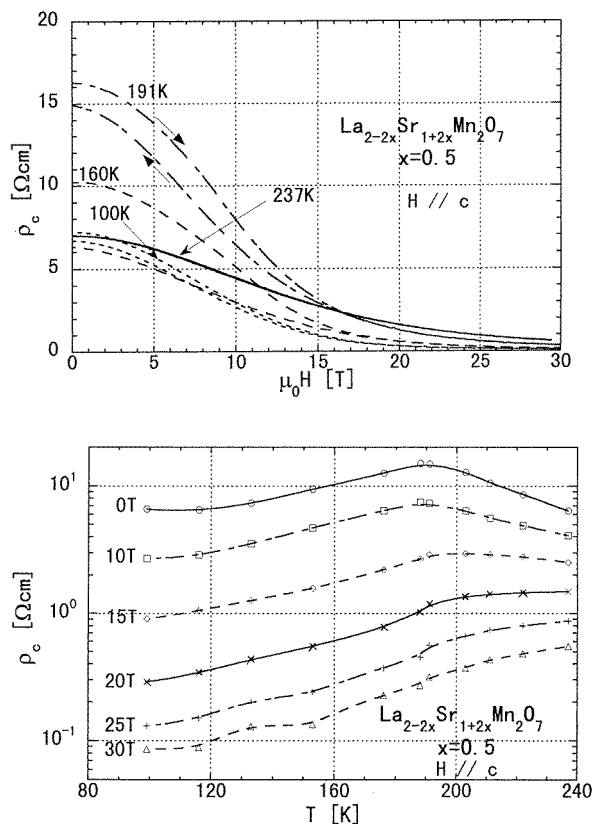
Figure 3. Magnetoresistance (upper panel) and temperature dependence of resistivity under magnetic fields (lower panel) with a current and magnetic fields perpendicular to the MnO$_2$ bilayers.

destroyed by magnetic fields, and that after applying magnetic field the proportion of the latter increases. At very low temperatures the latter phase dominates from the beginning, so the hysteresis of the R–H curve is small. The change of the temperature dependence of the resistivity at very low temperatures can be explained such that without magnetic field there is the A-type AF phase, where e$_g$ electrons can move only in each MnO$_2$ plane, which causes carrier localization. However, under high magnetic fields carriers become movable to the neighbouring layers and the dimensionality of transport becomes 3D-like, which breaks the carrier localization. Of course these are just conjectures, and it is necessary to perform neutron scattering experiments to obtain a reliable model for the spin structures.

There are still many problems. First, there is an abnormal jump in the M–H curve at low temperatures with H parallel to the ab plane. This jump cannot be explained by the magnetic anisotropy. Second compared with other systems which favour, the A-type AF structure, such as Pr$_{1-x}$Sr$_x$MnO$_3$($x = 0.5$), the resistivity along the c axis is too small. Moreover, as mentioned before, in the M–H curve there is an area where dM/dH increases with increasing H, which cannot be explained by the spin polarization.

In summary, we have investigated magnetic and transport properties of the layered perovskite manganite LaSr$_2$Mn$_2$O$_7$ under high magnetic field. In high magnetic fields, there was no charge-ordered phase, and the transport properties were metallic. The effect of
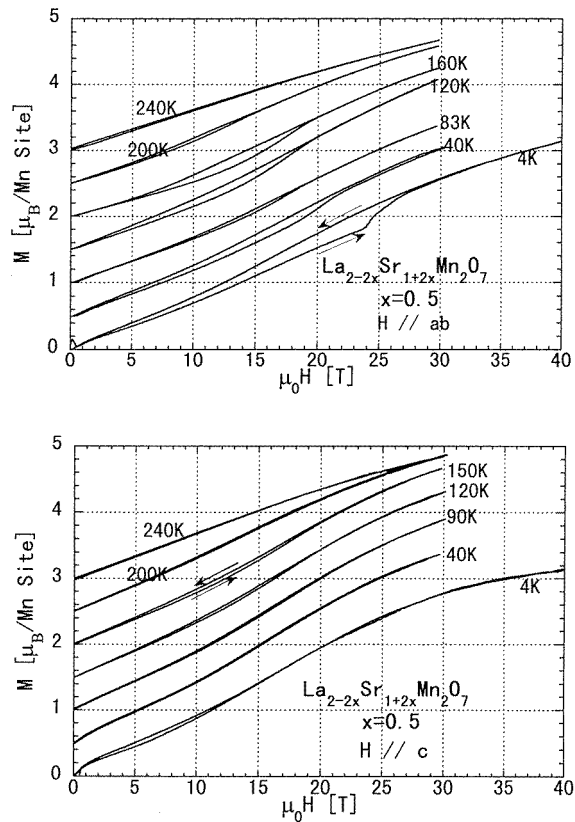
Figure 4. Magnetization with magnetic fields parallel (upper panel) and perpendicular (lower panel) to the MnO$_2$ bilayers.

the magnetic field did not disappear completely even when magnetic field was decreased, which implies that spin and orbital structure are changed by external magnetic field.

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