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LETTER TO THE EDITOR

## The large magnetoresistance of $\text{La}_{1-x}\text{Sr}_x\text{CoO}_3$ at low temperatures

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**Abstract.** We report here the resistivity ( $\rho$ ) and the magnetoresistance (MR) of the ferromagnetic perovskite oxide system  $\text{La}_{1-x}\text{Sr}_x\text{CoO}_3$  ( $0.1 < x < 0.5$ ) in a temperature range down to 4.2 K and magnetic fields up to 6 T. The MR is positive for  $x = 0.5$  and is negative for  $x \leq 0.4$ . We find that while in the metallic samples ( $x > 0.2$ ) the magnitude of the MR is typically small ( $< 10\%$ ), it is large in the composition range where  $x \leq 0.2$ , where the system is close to the metal-to-insulator transition. We compare these observations with those made for the  $\text{La}_{1-x}\text{Sr}_x\text{MnO}_3$  system.

Magnetoresistance of hole-doped 3d transition metal oxides has been a topic of interest since the discovery of very large magnetoresistance in the divalent metal-doped  $\text{La}_{1-x}\text{A}_x\text{MnO}_3$  ( $\text{A} = \text{Ca}, \text{Sr}, \text{Ba}$  etc) systems [1]. Doping results in mixed valency of Mn ( $\text{Mn}^{4+}/\text{Mn}^{3+}$ ) ions which is responsible for the unusual electronic transport and the large magnetoresistance in these materials.  $\text{LaCoO}_3$  is similar to  $\text{LaMnO}_3$  in many respects. In stoichiometric form both these materials are insulating [2] (with an increase of temperature Co ions undergo a transition to a high-spin state which leads to a metallic state in  $\text{LaCoO}_3$  for  $T > 650$  K). It is known that the substitution of divalent metals like Ca or Sr at the La site (as in  $\text{La}_{1-x}\text{Sr}_x\text{CoO}_3$ ) leads to a mixed valency of Co ions which gives rise to ferromagnetism for  $x > 0.15$  and transition to a metallic state for  $x > x_c = 0.2$  [3, 4]. Recent photoelectron spectroscopy studies have shown that doping with Sr leads to the formation of high-spin  $\text{Co}^{4+}$  ions and this induces states in the band gap with substantial oxygen 2p character due to strong hybridization [5]. However, the detailed mechanism of the ferromagnetism and the metal-to-insulator transition in these materials are not well understood. The above discussion summarizes the essential features of the  $\text{La}_{1-x}\text{Sr}_x\text{CoO}_3$  system.

In this letter we report briefly the main observations on the magnetoresistance (MR) behaviour of the  $\text{La}_{1-x}\text{Sr}_x\text{CoO}_3$  system. The motivation for the investigation is to find whether this particular system, like the manganates, exhibits large MR, or not. We find that at low temperatures the MR has an interesting dependence on the composition ( $x$ ), and close to the composition of the metal-insulator transition ( $x_c \cong 0.2$ ) the MR can be substantial.

The samples studied were polycrystalline pellets produced by the solid state reaction method [5]. The resistance was measured using a four-probe a.c. (20 Hz) or d.c. method in

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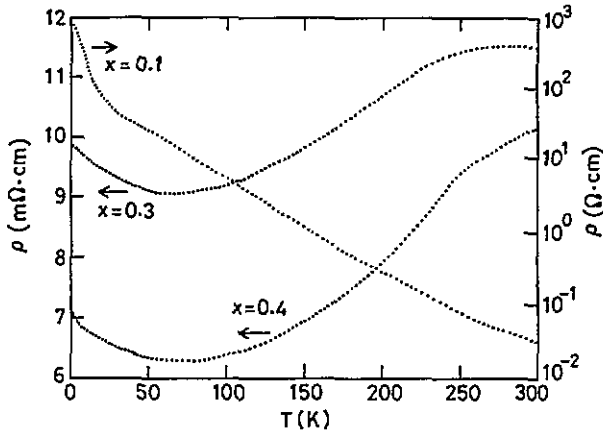


Figure 1. Resistivities ( $\rho$ ) as a function of  $T$  for the  $\text{La}_{1-x}\text{Sr}_x\text{CoO}_3$  system. (The resistivity of the  $x = 0.4$  sample has been multiplied by 2 to bring it to scale.)

the temperature range  $0.4 \text{ K} < T < 300 \text{ K}$ . The MR was measured in magnetic fields up to 6 T using a superconducting solenoid at 4.2 K.

In figure 1 we show the resistivities ( $\rho$ ) as a function of temperature for different values of  $x$ . For  $x > x_c = 0.2$ ,  $\rho$  has a metallic temperature dependence (i.e.  $d\rho/dT > 0$ ). The ferromagnetic transition temperatures ( $T_c$ ) are in the range 200 K to 250 K depending on the Sr concentration [6, 7] and  $\rho$  shows a change in slope at around  $T_c$ , like a conventional metallic ferromagnet. From the magnetization studies reported earlier [6], we find that for  $T \ll T_c$  the magnetization ( $M$ ) of the samples with  $x \geq 0.2$  has a temperature dependence given by  $M = M_0 - \alpha T^{3/2}$ ,  $\alpha$  being a constant. The  $T^{3/2}$  term is expected to arise from the spin-wave excitation [8]. We estimated a spin-wave stiffness constant  $D_{SW} \approx 150 \text{ meV \AA}^2$  from  $\alpha$ . This value is similar to, but somewhat larger than, the value of  $D_{SW}$  estimated for the manganates [9, 10]. The electron scattering from the spin waves leads to a  $T^2$  term in the  $\rho$  of the ferromagnetic metals at low temperatures [8]. To look for a  $T^2$  term we have plotted  $\rho$  as a function of  $T^2$  for one of the metallic samples,  $\text{La}_{0.6}\text{Sr}_{0.4}\text{CoO}_3$ , in figure 2. A clear  $T^2$  term is not visible because of a rise in  $\rho$  at lower temperatures. We found that for  $0.5 \text{ K} < T < 150 \text{ K}$ ,  $\rho$  follows a relation of the type  $\rho = \rho_0 - A\sqrt{T} + BT^2$ . For the particular system,  $B \approx 2.9 \times 10^{-8} \text{ \AA cm K}^{-2}$ . The term  $A\sqrt{T}$  (with negative sign), which gives rise to a small rise in  $\rho$  at lower temperatures, has been seen for a number of metallic oxides with disorder and is caused by quantum interference effects [11].

In figure 3 we have shown the variation of the saturation magnetization  $M_s$ , resistivity ( $\rho_{4.2 \text{ K}}$ ) and the MR ( $\Delta\rho/\rho$ ) as a function of  $x$  at  $T = 4.2 \text{ K}$ . The MR data were taken in a field of 6 T. (We have defined the MR as  $\Delta\rho/\rho = \{\rho(H) - \rho(0)\}/\rho(0)$ .) In the composition region ( $x \leq 0.2$ ) where the  $\rho_{4.2 \text{ K}}$  rises as the metal-to-insulator transition is approached, the MR also becomes large and negative. It is interesting that in the same composition range  $M_s$  also shows a sharp decline. The MR remains negative and small ( $|\Delta\rho/\rho| < 10\%$ ) in the metallic regime for  $0.2 < x \leq 0.4$ . It is also interesting that for more metallic samples ( $x \approx 0.5$ ) the MR becomes positive. Such positive MR has also been seen in certain other metallic oxides [11]. However, the origin of this is not known. It should also be pointed out that the MR for all values of  $x$  is isotropic with respect to the relative orientation of the current and the field. The difference between the MRs in the two geometries (current parallel and perpendicular to the applied field) is less than 15%. Figure 3 also suggests that for certain values of  $x$ ,  $0.4 < x < 0.5$ , the MR is actually zero.  $\text{La}_{1-x}\text{Sr}_x\text{CoO}_3$  thin

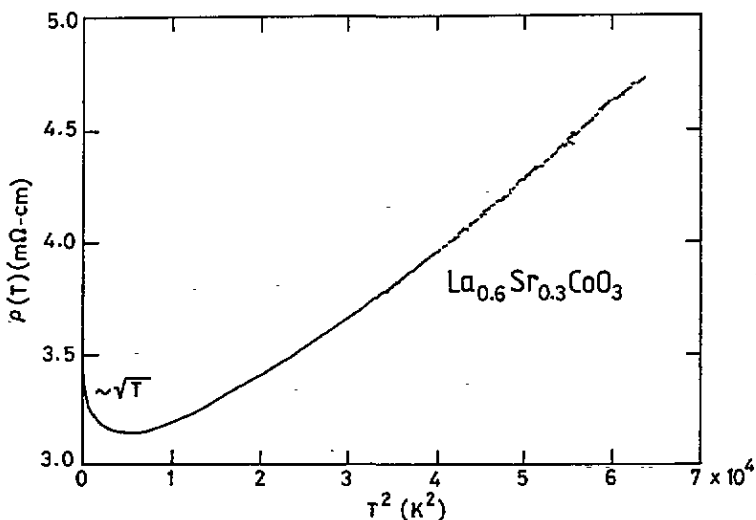


Figure 2.  $\rho$  versus  $T^2$  for the metallic sample with  $x = 0.4$ . Note the rise in  $\rho$  at low  $T$  which follows a  $\sqrt{T}$  behaviour.

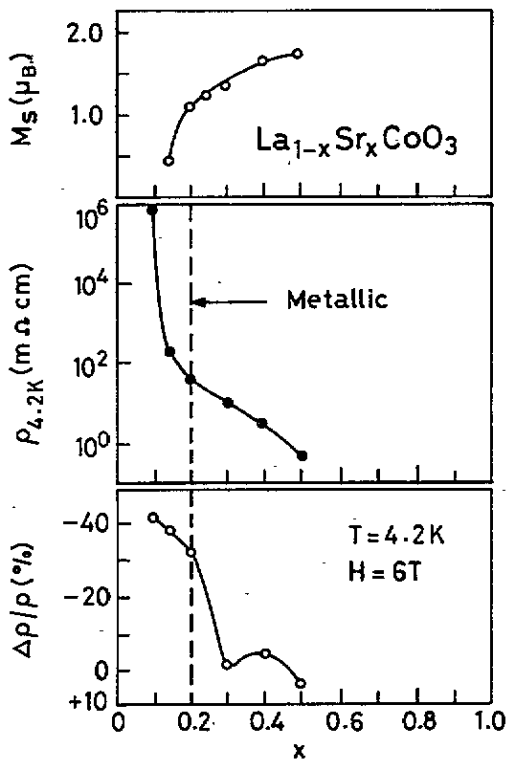


Figure 3. The dependence of the saturation magnetization  $M_s$  (in Bohr magnetons),  $\rho_{4.2\text{K}}$  and the MR ( $\Delta\rho/\rho$ ) on the composition  $x$ . The MR data were taken at  $T = 4.2\text{K}$  and  $H = 6\text{T}$ . The  $M_s$  data were obtained from [6].

films with metallic compositions are used as electrodes in devices based on perovskite oxide films. Compositions with almost zero MR are particularly attractive from the point of view of such applications [12].

The small negative MR at low temperatures for the composition range  $0.2 < x < 0.4$  can be explained as arising from the quantum interference effects which give rise to the small rise in  $\rho$  at low temperature [11]. However, the large negative MR for the composition range where  $x \approx x_c = 0.2$  cannot be explained with these ideas. This composition is not only close to the critical composition for the metal-insulator transition ( $x_c \cong 0.2$ ), but this is also in the composition range ( $x = 0.15$ ) in which ferromagnetism sets in. From the Mössbauer [4], magnetization [6] and a.c. susceptibility [7] studies, it appears that in this composition range the material is not a ferromagnet with very-long-range magnetic order. Rather, it is magnetically inhomogeneous. A simple way to look at the material would be to suppose that it has ferromagnetic clusters in a paramagnetic matrix. The resulting magnetic as well as electronic transport properties will therefore depend on the nature of these magnetic clusters and how they behave in a magnetic field. The origin of large magnetoresistance may thus be linked to the appearance of clusters of ferromagnetically aligned spins.

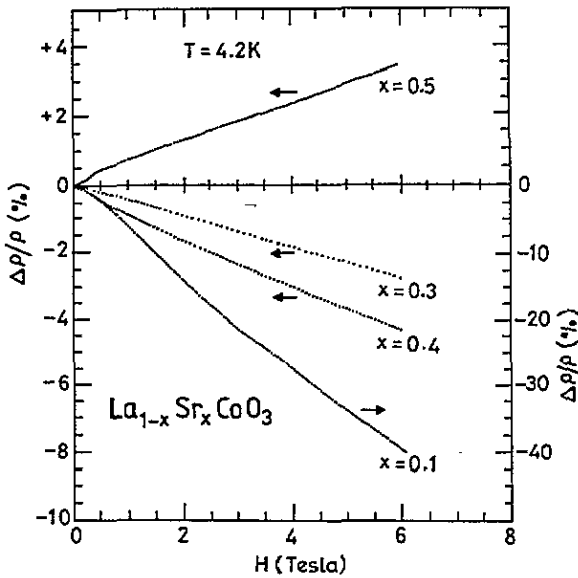


Figure 4. The dependence of the MR on  $H$  at  $T = 4.2$  K for different compositions.

The change of the resistance  $\rho$  as a function of  $H$  is shown in figure 4. For the metallic compositions  $x = 0.3-0.5$ , the MR shows little sign of saturation with  $H$ . Similar trends in the variation of the MR with  $H$  persist for samples with lower  $x$ . This type of dependence of the MR on  $H$  is unlike that which one observes in ferromagnetic oxides like the  $\text{La}_{1-x}\text{Sr}_x\text{MnO}_3$  system. In this system, for the temperature range  $T < T_c$ , the MR as a function of  $H$  shows two regimes—it is very similar to the variation of the magnetization  $M$  with  $H$ . In low-field regions ( $H < 1$  T)  $\Delta\rho/\rho$  changes rapidly with  $H$  and then beyond the saturation ( $H > 1$  T) the change is gradual.

We now briefly point out the main differences between the doped cobaltates and the manganates [1, 9]. In addition to the difference between the field dependences of the MR, the two systems have other important differences. One noteworthy difference is between their zero-field resistivities. In the manganate system ( $\text{La}_{1-x}\text{A}_x\text{MnO}_3$ ) the resistivity is activated

in the paramagnetic state (i.e., for  $T > T_c$ ,  $d\rho/dT < 0$ ) and as  $T$  is decreased it shows a peak at  $T \approx T_c$ , and for  $T < T_c$ ,  $\rho$  shows metallic behaviour (i.e.,  $d\rho/dT > 0$ ). This peak in  $\rho$  is taken as a signature of an insulator-to-metal transition. In the cobaltate system no such peak is seen in  $\rho$  as a function of  $T$ , implying that there is no temperature-driven insulator-metal transition in the system. In this system the insulator-to-metal transition is brought about by changing the composition and occurs when the system is already ferromagnetic. Both these systems are ferromagnetic as well as metallic due to the mixed valency of the transition metal ions. However, there are important qualitative differences in their electronic transport properties. The main difference is likely to arise from the electronic/spin structure and the coupling of the lattice distortions to the spins.  $Mn^{4+}$  has a  $t_{2g}^3 e_g^0$  configuration, while  $Co^{4+}$  in the high-spin state has a  $t_{2g}^3 e_g^2$  configuration. The existence of empty  $e_g$  orbitals in  $Mn^{4+}$  and half-filled  $e_g$  orbitals in  $Co^{4+}$  may be responsible for the qualitative difference in the electronic transport behaviour. A detailed investigation of the MR and magnetic properties of the  $La_{1-x}Sr_xCoO_3$  system is in progress.

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