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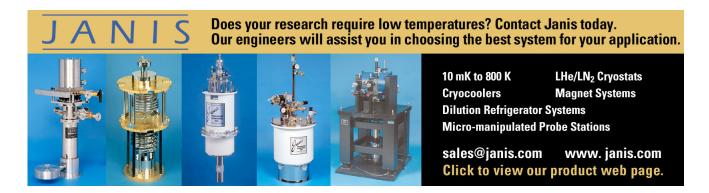
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## Colossal piezoresistance effect in Sm<sub>0.55</sub>(Sr<sub>0.5</sub>Ca<sub>0.5</sub>)<sub>0.45</sub>MnO<sub>3</sub> single crystal

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Manganites have received much attention since the discovery of colossal magnetoresistance effect, which has already made them invaluable components for various technological applications like magnetic random access memory, switching device, etc. On technological demand, the magnetotransport studies on different kinds of manganites are still going on in order to search a suitable system that will show large magnetoresistance (MR) at a relatively small magnetic field. Similar to MR, piezoresistance (PR), the change in electrical resistance in response to external pressure, can also be an important parameter for various technological purposes. Although there are several reports on the PR effect in perovskite and bilayer manganites, the observed magnitude of PR is large only at high pressure, which reduces the scope of possible technological applications. 1-7 Therefore, the challenge remains to find out new materials that exhibit a colossal piezoresistance (CPR) at a relatively low pressure. The guidelines for the choice of an appropriate material are that it should exhibit a sharp metal-insulator transition (MIT) and the transition should shift appreciably under the influence of small amount of external pressure.

From the electronic phase diagram of narrowband  $Sm_{1-x}Sr_xMnO_3$ , it is clear that the system has a multicritical point near the half doping, where three phases ferromagnetic (FM), charge/orbital ordering, and antiferromagnetic compete with each other and for the exact x = 0.49 composition, the crystal shows the special stability towards the charge/orbital ordering.<sup>8</sup> When a system is in the vicinity of such a critical point, even a weak perturbation may change the physical properties dramatically or induce a phase transition. Indeed, previous works on  $Sm_{1-x}Sr_xMnO_3$  have shown that the system exhibits a sharp first-order FM metal to paramagnetic (PM) insulator transition, and the transition is extremely sensitive to several external or internal perturbations such as magnetic field, pressure, A-site/B-site substitution or oxygen isotope exchange, etc. for x = 0.45-0.48. The bandwidth and hence the FM transition temperature  $(T_C)$  can be reduced further by substituting  $Ca^{2+}$  at  $Sr^{2+}$  site in  $Sm_{1-x}Sr_xMnO_3$ .<sup>10</sup> When the bandwidth narrows down, the system is expected to be highly susceptible to external perturbations.

In this letter, we present mainly the resistivity results on  $Sm_{1-x}(Sr_{1-y}Ca_y)_xMnO_3$  (x = 0.45 and y = 0.5) single crystal as functions of temperature and uniaxial pressure (P). We have chosen the Ca doping level at 0.5 because both the magnetic and electronic ground states are extremely sensitive to y around this doping.  $Sm_{0.55}(Sr_{1-y}Ca_y)_{0.45}MnO_3$ remains FM up to  $y \approx 0.6$ , and a FM metal to charge ordered insulator transition occurs just above this Ca content. 10 So, y = 0.6 is also a multicritical point, and the present system is in the verge of two multicritical points: one is with respect to x and another is with respect to y. Indeed, we observe that the electrical resistivity of Sm<sub>0.55</sub>(Sr<sub>0.5</sub>Ca<sub>0.5</sub>)<sub>0.45</sub>MnO<sub>3</sub> (SSCMO) crystal is very sensitive to uniaxial pressure. The MIT temperature  $(T_{MI})$  increases for uniaxial pressure applied parallel to c axis (P||c) whereas  $T_{MI}$  decreases for P perpendicular to c axis  $(P \perp c)$  at unusually high rates 79 and 77 K/GPa, respectively. Also, a huge PR (10<sup>7</sup>% at 90 MPa) is observed for P||c.

Single crystals of Sm<sub>0.55</sub>(Sr<sub>0.5</sub>Ca<sub>0.5</sub>)<sub>0.45</sub>MnO<sub>3</sub> were grown by the floating zone technique under oxygen atmosphere. 22 No trace of impurity was detected in powder x-ray diffraction (XRD). Besides XRD, the quality of the crystal was carefully checked by various techniques such as Laue diffraction, scanning electron microscope, etc. Well-characterized crystals were then aligned with a goniometer using Laue diffraction pattern and cut along the ab plane and c axis with a typical dimension of  $2.5 \times 2 \times 2.5 \,\mathrm{mm}^3$  and polished carefully with nice velvet to get parallel and flat surfaces. The electrical resistivity was measured by using standard four-probe technique. The sample was mounted in between the Be-Cu anvils. Both the anvils were electrically insulated from the sample by coating with GE varnish. Uniaxial pressure was applied both along the ab plane and c axis. A calibrated cernox sensor was used to measure the sample temperature. The pressure was applied through a force generator by rotating a disc micrometer, and its magnitude was calculated directly from the surface area of the

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crystal, the rotations of the disc micrometer, and the forceconstant of the spring.<sup>23</sup> The magnetization measurements were performed by using a superconducting quantum interference device magnetometer (MPMS, Quantum Design).

Figure 1 shows the temperature dependence of *ab*-plane  $(\rho_{ab})$  and c-axis  $(\rho_c)$  resistivities of SSCMO single crystal for different uniaxial pressures.  $\rho_{ab}$  and  $\rho_c$  were measured with current flow perpendicular and parallel to the c axis, respectively, while the applied pressure is always perpendicular to the direction of current flow. Both  $\rho_{ab}$  and  $\rho_c$  increase exponentially upon cooling from high temperatures and then suddenly drops as much as six decades just below  $T_{MI}$  $(\sim 82.5 \,\mathrm{K})$ . The temperature dependence of magnetization (M) [inset of Fig. 1(a)] shows that M increases sharply just below  $T_{MI}$ , which indicates that  $T_{MI}$  and  $T_{C}$  are very close to each other. M exhibits a strong thermal hysteresis, whose width is  $\sim 13 \, \text{K}$  at ambient pressure. The temperature dependence of resistivity at ambient pressure also shows similar hysteresis (not shown). The sharp change in  $\rho_{ab}$ ,  $\rho_c$ , and M at  $T_C$  and the presence of thermal hysteresis suggest that FM metal to PM insulator transition in SSCMO is first order in nature. In case of Sm<sub>0.55</sub>Sr<sub>0.45</sub>MnO<sub>3</sub> (SSMO) single crystal, resistivity drops by three orders of magnitude at  $T_{MI}$  $(\sim 135 \,\mathrm{K})$  and the width of the thermal hysteresis in resistivity and magnetization is about 1.7 K.16 So both the order of the resistivity drop at  $T_{MI}$  and the hysteresis width are much

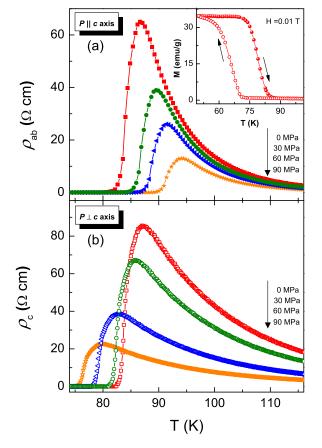


FIG. 1. Temperature dependence of (a) ab-plane resistivity ( $\rho_{ab}$ ) of  $Sm_{0.55}(Sr_{0.5}Ca_{0.5})_{0.45}MnO_3$  single crystal measured under various uniaxial pressure parallel to c axis (P||c) (b) c-axis resistivity  $(\rho_c)$  measured for different pressure, applied perpendicular to the c axis  $(P \perp c)$ . Data taken in the heating cycle are shown. Inset shows the temperature dependence of magnetization (M) both in heating and cooling cycles.

larger in SSCMO than SSMO, indicating that the first-order FM transition in SSCMO is much stronger than that for SSMO. With the substitution of Ca at the Sr site in SSMO, both the bandwidth (determined by the average A-site cation radius,  $\langle r_A \rangle$ ) and quenched disorder (measured as the variance of ionic radii,  $\sigma^2$  (Ref. 24)) decrease. The decrease of  $\langle r_A \rangle$  tends to reduce  $T_C$  whereas  $\sigma^2$  plays an opposite role.<sup>24</sup> As the effect of  $\langle r_A \rangle$  on  $T_C$  is stronger than the effect of  $\sigma^2$ on the same,  $T_C$  in SSCMO reduces with the increase of Ca content. According to tight binding approximation, electronic bandwidth (W) depends on the Mn–O–Mn bond angle  $(\phi)$  and Mn–O bond length  $(d_{Mn-O})$  and can be written as  $W = \cos \omega / d_{Mn-O}^{3.5}$ , where the tilt angle  $\omega$  is defined as  $\omega = (\pi - \phi)/2$ . Radaelli *et al.* have shown that with decreasing  $\langle r_A \rangle$  below the critical value  $\sim 1.24$  Å,  $d_{Mn-O}$  increases whereas  $\varphi$  decreases, and the combined effect contributes to the rapid decrease of  $W^{25}$ . They have also observed that the evolution of  $T_C$  with  $\langle r_A \rangle$  is remarkably similar to that of W. In manganites, the FM double exchange interaction (J) is proportional to the bandwidth or the charge transfer integral.  $^{26}$  As  $T_C$  is proportional to J, the decrease of bandwidth causes the suppression of  $T_C$ .  $^{26}$  When the bandwidth of a system narrows down, some of the charge carriers can be trapped by the lattice distortion, which gives rise to the formation of polarons, and it has been shown by using the mean-field and variational Lang-Firsov approximations that  $T_C$  decreases with increasing electron-phonon coupling strength.<sup>27</sup> Several studies have shown the direct evidences of the existence of polarons in perovskite and bilayer manganites by using neutron diffraction, electron paramagnetic resonance, optical conductivity spectra, etc. 28-32 The polaronic nature of charge conduction can also be inferred from electronic and thermal transport.<sup>33–36</sup> The possibility for the existence of polarons primarily comes from the activated temperature dependence of  $\rho$  [ $\rho \sim \exp(E_{\varrho}/kT)$ , where  $E_{\varrho}$  is the activation energy] above  $T_C$ . Previous works have shown that  $\rho$  of SSMO obeys the activated behavior with  $E_g \sim 102$  meV. The resistivity of SSCMO also exhibits similar behavior but with higher value of  $E_g$  (~119 meV). The partial substitution of Ca at Sr site in SSMO reduces  $\langle r_A \rangle$  from 1.21 to 1.18 Å, and hence SSCMO becomes more favorable for the formation of polarons. When these polarons form, FM is truncated, self-trapping becomes more evident, and drives the material towards a first-order phase transition.

The application of uniaxial pressure both parallel and perpendicular to the c axis of SSCMO affects MIT significantly (Fig. 1). For  $P||c, \rho_{ab}$  decreases and MIT shifts monotonically towards higher temperature with increasing P. The effect of P on reducing  $\rho_{ab}$  is quite strong in a narrow temperature range of few Kelvins above and below  $T_{MI}(0)$ whereas much smaller change in  $\rho_{ab}$  with P has been observed both at high temperatures well above  $T_{MI}$  as well as in the metallic state below  $T_{MI}$ . In contrast to  $\rho_{ab}$ , P dependence of  $\rho_c$  is quite unusual and  $T_{MI}$  decreases sharply with P. Here, the effect of P on  $\rho_c$  is not confined around  $T_{MI}$  but extended over a wider range of temperature above MIT. Figure 2(a) shows the dependence of  $T_{MI}$  [defined as the maximum in  $d\rho/dT$ ] on P for SSCMO crystal. Neglecting the weak nonlinear dependence of  $T_{MI}$  on P, we find that  $T_{MI}$ increases at the rate of  $\sim$ 79 K/GPa for P||c while it decreases

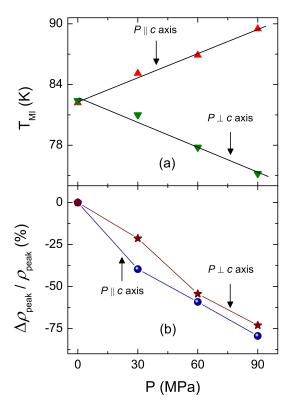


FIG. 2. Uniaxial pressure dependence of (a) metal-insulator transition temperature ( $T_{MI}$ ) and (b)  $\Delta \rho_{peak}/\rho_{peak}$  of SSCMO.

at the rate of  $\sim$ 77 K/GPa for  $P\perp c$ . In our previous work on  $Sm_{0.52}Sr_{0.48}MnO_3$ , the values of  $dT_{MI}/dP$  were 17 and -39 K/GPa for P||c and  $P\perp c$ , respectively. <sup>18</sup> Apart from 3D perovskite manganites, there are few reports on the effect of uniaxial pressure on the charge transport in layered manganites.<sup>6,37</sup> In La<sub>1.25</sub>Sr<sub>1.75</sub>Mn<sub>2</sub>O<sub>7</sub> crystal, the application of uniaxial pressure raises the  $T_{MI}$  at the rate of 46.33 K/GPa for P||c, and  $T_{MI}$  decreases at the rate of 16.58 K/GPa for  $P\perp c$ . From literature survey, we understand that the above values of  $dT_{MI}/dP$  for SSCMO are much higher than that observed in other metallic FM perovskite and bilayer manganites. To illustrate the effect of uniaxial pressure on peak resistivity  $(\rho_{peak})$ , we have calculated the change in peak resistivity as 
$$\begin{split} & \Delta \dot{\rho}_{peak}/\rho_{peak} = [\rho_{peak}(P) - \rho_{peak}(0)]/\rho_{peak}(0). \quad \text{Figure} \quad \text{2(b)} \\ & \text{shows the } P \text{ dependence of } \Delta \rho_{peak}/\rho_{peak} \text{ for } P||c \text{ and } P \bot c. \end{split}$$
In both the cases,  $\Delta \rho_{peak}$  decreases almost linearly with P, but the rate of decrease is slightly larger for P||c.

External pressure, whether it is hydrostatic or uniaxial, changes electrical and magnetic properties of manganites mainly through lattice parameters. From the pressure dependent neutron diffraction measurements on  $\text{La}_{1-x}\text{Sr}_x\text{MnO}_3$ , it has been observed that  $d_{Mn-O}$  compresses under hydrostatic pressure whereas  $\phi$  increases. As both of these changes increase the charge transfer process, the double exchange favoring FM metallic state is enhanced and hence  $T_{MI}$  or  $T_C$  increases. But in case of uniaxial pressure, one has to consider the pressure dependent change in  $\phi$  both in the apical (i.e., along c axis) and equatorial (i.e., in ab plane) direction separately. When  $\rho_c$  is measured with  $P \perp c$ , Mn–O–Mn bond angle along equatorial direction ( $\phi_{eq}$ ) is expected to increase, and at the same time Mn–O–Mn bond angle along apical direction ( $\phi_{ap}$ ) will decrease and  $d_{Mn-O}$  along the c axis will

increase due to the Poisson ratio. For orthorhombic structure, the departure of  $\phi$  from collinearity is larger along the apical direction as compared to the equatorial direction. So the physical properties would be more sensitive to the change in  $\phi_{ap}$  than  $\phi_{eq}$ , and hence  $T_{MI}$  decreases with P for  $P \perp c$ . Similarly, for  $\rho_{ab}$  with P||c,  $\phi_{ap}$  increases and Mn–O bond length along c axis decreases whereas  $\phi_{eq}$  decreases and the in plane Mn–O bond length increases with P. As the effect of the change of c axis parameters on physical properties is larger than that along the ab plane,  $T_{MI}$  increases for P||c. However, the studies of the crystal structure under uniaxial pressure are necessary for clear understanding.

We have already shown that the resistivity of SSCMO changes drastically with uniaxial pressure. In order to quantify these changes, we have calculated the piezoresistance as  $\Delta \rho / \rho(P) = [\rho(0) - \rho(P)] / \rho(P)$ , where  $\rho(0)$  and  $\rho(P)$  are the resistivity values at ambient and under uniaxial pressure, respectively. Figures 3(a) and 3(b) show the temperature dependence of PR of SSCMO crystal for different P, applied both parallel and perpendicular to the c axis, respectively. For P||c, PR initially does not change significantly with the decrease of temperature, but it increases very rapidly as T approaches close to  $T_{MI}$ , passes through a maximum, and then decreases very slowly at low temperatures. For  $P = 30 \,\mathrm{MPa}$ , the peak in PR vs T curve appears at around  $T_{MI}(0)$  and the value of PR at peak is  $\sim 20400\%$ . As P increases, the position of the peak shifts very slowly towards higher temperature whereas the value of PR at peak increases very rapidly and exceeds  $10^7\%$  for P = 90 MPa. We believe that such kind of

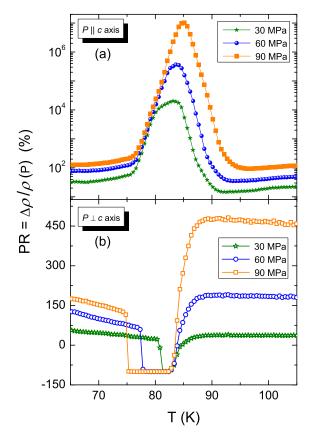


FIG. 3. Temperature dependence of piezoresistance  $\Delta \rho/\rho(P) = [\rho(0) - \rho(P)]/\rho(P)$  of SSCMO for various uniaxial pressure, applied (a) parallel and (b) perpendicular to the c axis. For clarity, PR is plotted in semi-log scale for P||c.

CPR effect has not been observed in any FM manganite. This CPR effect may be viewed as a switching-like phenomenon between high-resistive and low-resistive states, which may have some technological applications. On the other hand, for  $P \perp c$ , both positive and negative PR have been observed which can be easily understood from the decrease and increase of  $\rho_c$  with P in the different temperature regions [Fig. 1(b)]. With decreasing temperature, PR at  $P = 30 \,\mathrm{MPa}$  does not change appreciably down to 87 K, but PR changes its sign from positive to negative around 85 K and shows a narrow square-well type minimum at around  $T_{MI}(0)$ . With further lowering of temperature below  $T_{MI}(0)$ , PR becomes positive and almost T independent as in the case of PM state. Also, the values of PR in the PM and FM states are almost same. It is interesting to note that with further increase of P above 30 MPa although the width of the minimum increases rapidly, the value of PR within the minimum does not change at all up to 90 MPa, i.e., PR remains pinned at  $\sim$ 100%. Also, as  $\rho_c$  is more sensitive to P in the PM state as compared to FM state for  $P \perp c$ , PR vs T curve becomes more and more asymmetric with increasing P. In this context, it may be useful to compare the present results with that of hydrostatic pressure effect on resistivity in manganites. In general, hydrostatic pressure always suppresses resistivity both in the PM and FM states, but its effect is much stronger in the vicinity of  $T_C$  and, as a result, PR exhibits a peak close to  $T_C$  as in the case of MR. In this sense, the effect of uniaxial pressure on resistivity close to  $T_{MI}$  for P||c is similar to that for hydrostatic pressure. However, for  $P \perp c$ , the temperature dependence of PR is very unusual. We would like to mention here that the first-order nature of the FM transition is strongest in Eu<sub>1-x</sub>Sr<sub>x</sub>MnO<sub>3</sub>  $(x \approx 0.4)$  with lowest  $T_C$  ( $\sim 40$  K) among the manganite family. 16,39 The hysteresis width is around 14 K, and the resistivity jump across the transition is  $\sim 9$  orders of magnitude. As the resistivity change at  $T_{MI}$  in this system is two orders of magnitude larger than in SSCMO, Eu<sub>1-x</sub>Sr<sub>x</sub>MnO<sub>3</sub> may show much larger PR effect close to MIT.

In summary, we have studied the influence of uniaxial pressure on resistivity in a single crystal of Sm<sub>0.55</sub>  $(Sr_{0.5}Ca_{0.5})_{0.45}MnO_3$ . With increasing pressure,  $T_{MI}$  increases for P||c and  $T_{MI}$  decreases for  $P\perp c$  at the rates 79 and 77K/GPa, respectively. This system also exhibits a true colossal piezoresistance effect at a relatively low value of applied pressure along c axis which opens up a broad range of possible applications with pressure as a control parameter.

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