

## Effect of uniaxial pressure on metal-insulator transition in $(\text{Sm}_{1-y}\text{Nd}_y)_{0.52}\text{Sr}_{0.48}\text{MnO}_3$ single crystals

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(Received 27 March 2009; accepted 3 June 2009; published online 25 June 2009)

We have investigated the effect of uniaxial pressure ( $P$ ) on resistivity along the  $ab$  plane and  $c$ -axis in single crystals of  $(\text{Sm}_{1-y}\text{Nd}_y)_{0.52}\text{Sr}_{0.48}\text{MnO}_3$  with  $y=0, 0.05$ , and  $0.3$ . The application of pressure along the  $c$ -axis shifts the metal-insulator transition (MIT) to higher temperature, while MIT temperature decreases with  $P$  when it is applied perpendicular to the  $c$ -axis. This behavior is quite different from that observed in hydrostatic pressure and can be explained by considering the  $P$  dependence change in equatorial and apical Mn–O–Mn bond angles. © 2009 American Institute of Physics. [DOI: 10.1063/1.3160019]

Narrowband manganites  $R_{1-x}A_x\text{MnO}_3$  ( $R$ : rare-earth ions,  $A$ : alkaline-earth ions) exhibit many complex and unusual phenomena due to the presence of several competitive interactions of comparable strength.<sup>1,2</sup> It has been shown that the physical properties of these systems are strongly affected by the presence of local disorder, arising mainly due to the size mismatch between  $R$  and  $A$  ions.<sup>1-3</sup> When the ionic size difference is large enough, the lattice polarons are formed, ferromagnetic (FM) metallic phase gets truncated and the nature of FM-paramagnetic (PM) transition becomes first-order with reduced metal-insulator transition (MIT) temperature ( $T_{\text{MI}}$ ). In prototypical  $\text{Sm}_{1-x}\text{Sr}_x\text{MnO}_3$ , the effect of local disorder on magnetic and electronic properties is well studied.<sup>3-5</sup> As the ionic radius of Sm differs significantly from that of Sr, the disorder is quite large and as a result this system exhibits a strong first-order FM-PM transition.<sup>4-6</sup> Several studies have shown that the nature of magnetic transition and the electronic phase diagram of  $\text{Sm}_{1-x}\text{Sr}_x\text{MnO}_3$  can be tuned by applying external perturbations such as pressure and magnetic field.<sup>4-8</sup> As a result of decrease in Mn–O bond length and increase in Mn–O–Mn bond angle,  $T_{\text{MI}}$  increases with the increase in hydrostatic pressure, leading to a pressure dependent phase boundary.<sup>5,6</sup> The unit cell under hydrostatic pressure is compressed simultaneously along all the crystallographic axes by an amount proportional to the compressibility of the respective axis. On the other hand, for uniaxial pressure ( $P$ ) the lattice compresses along the applied pressure direction but expands in other two perpendicular directions due to the Poisson effect. This provides a unique opportunity to study the physical properties both along and perpendicular to the direction of applied pressure. The measurements of physical parameters such as resistivity with current direction perpendicular to the uniaxial pressure may reveal different results from that of hydrostatic pressure.

In this letter, we have studied the uniaxial pressure dependence of  $ab$ -plane ( $\rho_{ab}$ ) and  $c$ -axis ( $\rho_c$ ) resistivities of

$(\text{Sm}_{1-y}\text{Nd}_y)_{0.52}\text{Sr}_{0.48}\text{MnO}_3$  single crystals with  $y=0, 0.05$  and  $0.3$  in order to investigate the sensitivity of transport properties on uniaxial pressure and to compare this result with that of hydrostatic pressure. We observe that uniaxial pressure applied parallel to  $c$ -axis ( $P\parallel c$ ) shifts the MIT toward higher temperature whereas  $T_{\text{MI}}$  decreases with  $P$  when it is perpendicular to  $c$ -axis ( $P\perp c$ ). This behavior is quite different from that observed for hydrostatic pressure and has not been reported earlier in FM manganites.

A series of single crystals of  $(\text{Sm}_{1-y}\text{Nd}_y)_{0.52}\text{Sr}_{0.48}\text{MnO}_3$  with  $y=0, 0.05, 0.3$  have been prepared by floating zone technique under oxygen atmosphere.<sup>9</sup> The quality of the crystal was carefully checked by various techniques such as x-ray powder diffraction, Laue diffraction, electron dispersive x-ray analysis, scanning electron microscope, etc. Well-characterized crystals were aligned with a goniometer using Laue diffraction pattern and cut along the  $ab$  plane and  $c$ -axis with a typical dimensions of  $1.5\times 1.4\times 1.5\text{ mm}^3$  and polished. The electrical resistivity was measured by using standard four-probe technique. The sample was mounted in between the anvils. Both the anvils are 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. Here, pressure was applied through a force generator by rotating a disc micrometer and it was calculated directly from the surface area of the crystal, the rotations of the disk micrometer, and the force-constant of the spring.<sup>10</sup> The magnetization measurements were performed by using a superconducting quantum interference device magnetometer (MPMS, Quantum Design) over a wide range of temperature.

Figures 1(a) and 1(b) show the temperature dependence of  $\rho_c$  and  $\rho_{ab}$  of  $\text{Sm}_{0.52}\text{Sr}_{0.48}\text{MnO}_3$  single crystal for different uniaxial pressures.  $\rho_{ab}$  and  $\rho_c$  were measured with applied pressure parallel and perpendicular to the  $c$ -axis, respectively. With decreasing temperature, both  $\rho_{ab}$  and  $\rho_c$  increase exponentially down to  $T_{\text{MI}}$  then drop abruptly as much as

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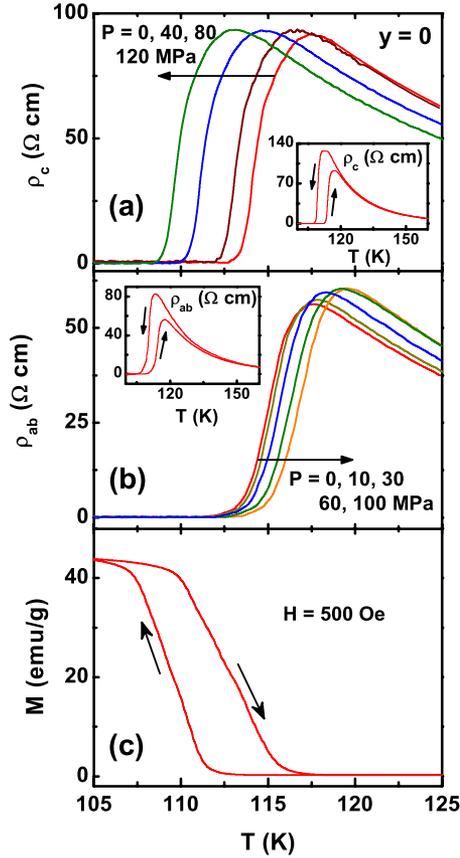


FIG. 1. (Color online) Temperature dependence of (a)  $c$ -axis resistivity ( $\rho_c$ ) of  $(\text{Sm}_{1-y}\text{Nd}_y)_{0.52}\text{Sr}_{0.48}\text{MnO}_3$  ( $y=0$ ) single crystal measured under various pressure ( $P$ ) perpendicular to  $c$ -axis (b)  $ab$ -plane resistivity ( $\rho_{ab}$ ) measured under various pressure parallel to  $c$ -axis. Data taken in the heating cycle are shown. Insets show the thermal hysteresis of  $\rho_c$  and  $\rho_{ab}$  at  $P=0$ . (c) Thermal hysteresis of magnetization ( $M$ ) at 500 Oe.

three orders of magnitude just below  $T_{\text{MI}}$ . The sharp increase in magnetization ( $M$ ) below MIT indicates that  $T_{\text{MI}}$  and  $T_C$  are close to each other [Fig. 1(c)]. Both resistivity [insets of Figs. 1(a) and 1(b)] and magnetization [Fig. 1(c)] exhibit strong thermal hysteresis. The sharp change in  $\rho_c$ ,  $\rho_{ab}$ , and  $M$  and the presence of thermal hysteresis are the indications of first-order FM to PM phase transition. The application of uniaxial pressure parallel and perpendicular to the  $c$ -axis affects MIT significantly. For  $P \perp c$ , MIT shifts toward lower temperature with the increase of  $P$ . Depending on the influence of  $P$  on  $\rho_c$ , the whole temperature region can be divided into three parts. At high temperatures well above  $T_{\text{MI}}$ ,  $\rho_c$  decreases with increasing  $P$ , whereas no appreciable change has been observed in the FM metallic state well below  $T_{\text{MI}}$ . The effect of  $P$  on  $\rho_c$  is strongest only in a narrow temperature range few kelvins below the  $T_{\text{MI}}$  where it increases rapidly with  $P$ . The peak resistivity of  $\rho_c$  remains almost unchanged with increasing pressure. In contrast to  $\rho_c$ ,  $\rho_{ab}(T)$  shows that both  $T_{\text{MI}}$  and the value of resistivity in the PM phase increase slowly with the increase of  $P$ .

Figures 2(a) and 2(b) represent the temperature dependence of  $\rho_c$  and  $\rho_{ab}$  for  $(\text{Sm}_{1-y}\text{Nd}_y)_{0.52}\text{Sr}_{0.48}\text{MnO}_3$  single crystals with  $y=0.05$  and  $0.3$ , respectively. It is clear that resistivity decreases, MIT shifts toward higher temperature and the discontinuity in resistivity at  $T_{\text{MI}}$  reduces with increasing  $y$ . At ambient pressure,  $T_{\text{MI}}$  increases almost linearly with  $y$  at the rate of  $\sim 159$  K, while the width of ther-

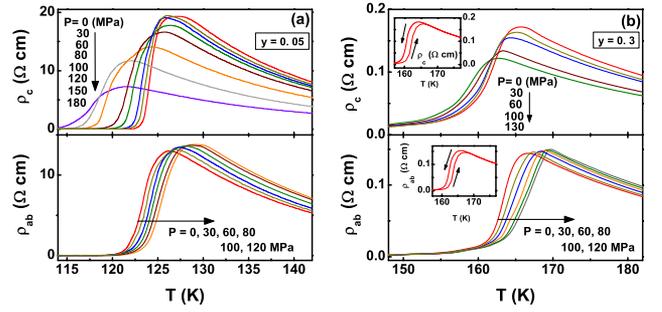


FIG. 2. (Color online) Temperature dependence of  $\rho_c$  (top) and  $\rho_{ab}$  (bottom) for different uniaxial pressure for  $(\text{Sm}_{1-y}\text{Nd}_y)_{0.52}\text{Sr}_{0.48}\text{MnO}_3$  single crystals: (a)  $y=0.05$  and (b)  $y=0.3$ . Insets show the thermal hysteresis of  $\rho_c$  and  $\rho_{ab}$  for  $y=0.3$  at  $P=0$ .

mal hysteresis decreases at the rate of  $\sim 10$  K. The hysteresis width is about 4.5 K for  $y=0$  (insets of Fig. 1) and it becomes  $\sim 1.5$  K for  $y=0.3$  [insets of Fig. 2(b)]. We have also analyzed the temperature dependence of resistivity for different  $y$ . In the temperature range  $T_{\text{MI}} < T < 300$  K, both  $\rho_{ab}$  and  $\rho_c$  obey an activated behavior  $\rho \sim \exp[E_g/kT]$ , where  $E_g$  is the activation energy.  $E_g$  decreases monotonically from 102 to 87 meV as  $y$  increases from 0 to 0.3. This suggests that substitution of Nd at Sm site suppresses the formation of polaronic state, increases the carrier mobility and stabilizes the metallic phase; thus, both the conductivity and  $T_{\text{MI}}$  are enhanced. The dependence of  $\rho_{ab}$  and  $\rho_c$  on  $P$  for  $y=0.05$  and  $0.3$  are qualitatively similar to that for undoped sample ( $y=0$ ).

Figure 3(a) shows the dependence of  $T_{\text{MI}}$  on  $P$  for  $y=0$ . Neglecting the weak nonlinear feature in  $T_{\text{MI}}(P)$ , we find that  $T_{\text{MI}}$  decreases at the rate of 39 K/GPa for  $P \perp c$  while it increases at the rate of 17 K/GPa for  $P \parallel c$ . In Fig. 3(b), the variation in room temperature resistivity ( $y=0$ ) with pressure is shown.  $\rho_{ab}$  increases sharply followed by a saturationlike behavior, while  $\rho_c$  decreases monotonically with increasing  $P$ . To illustrate the effect of uniaxial pressure on peak resistivity ( $\rho_{\text{peak}}$ ), we have calculated the change in peak resistivity  $\Delta\rho_{\text{peak}}/\rho_{\text{peak}} = [\rho_{\text{peak}}(P) - \rho_{\text{peak}}(0)]/\rho_{\text{peak}}(0)$ . In Fig. 3(c),  $\Delta\rho_{\text{peak}}/\rho_{\text{peak}}$  has been plotted as a function of  $P$  for different  $y$ . The figure shows that  $\Delta\rho_{\text{peak}}/\rho_{\text{peak}}$  increases with  $P$  for all  $y$  in the case of  $\rho_{ab}$  but decreases for  $\rho_c$ . With increasing  $y$ , the dependence of  $\Delta\rho_{\text{peak}}/\rho_{\text{peak}}$  on  $P$  diminishes for  $\rho_{ab}$  while strongly increases for  $\rho_c$ . The rate of change of  $T_{\text{MI}}$  with  $P$ , i.e.,  $dT_{\text{MI}}/dP$  increases slowly for  $P \parallel c$  and decreases faster for  $P \perp c$  with increasing  $y$  [inset of Fig. 3(c)].

For a first-order magnetic phase transition, pressure can shift the transition temperature, leading to a phase transition line along which two thermodynamic phases coexist. To analyze the metal-insulator or FM-PM phase boundary, we use the Clausius-Clapeyron equation. The Clausius-Clapeyron relation can be modified as,  $dT_{\text{MI}}/dP = (bc\Delta a)/\Delta S$  or  $dT_{\text{MI}}/dP = (ac\Delta b)/\Delta S$  for  $P \perp c$  and  $dT_{\text{MI}}/dP = (ab\Delta c)/\Delta S$  for  $P \parallel c$ .<sup>11</sup> Here,  $S$  denotes the entropy per unit cell and  $a$ ,  $b$ ,  $c$  are the cell parameters. Using the observed values of  $dT_{\text{MI}}/dP$  and the reported data on cell parameters for  $y=0$ ,<sup>12,13</sup> the entropy difference  $\Delta S$  associated with the FM-PM transition is found to be  $\sim 2.3$  J/mol K for  $P \parallel c$  and  $\sim 0.6$  J/mol K for  $P \perp c$ . In both the cases,  $\Delta S$  is positive because  $dT_{\text{MI}}/dP$  and  $\Delta a$  (or  $\Delta b$ ) are negative for  $P \perp c$ , whereas  $dT_{\text{MI}}/dP$  and  $\Delta c$  are positive for  $P \parallel c$ .<sup>13</sup> As  $y$  increases, the discontinuity in the cell parameters at  $T_{\text{MI}}$  con-

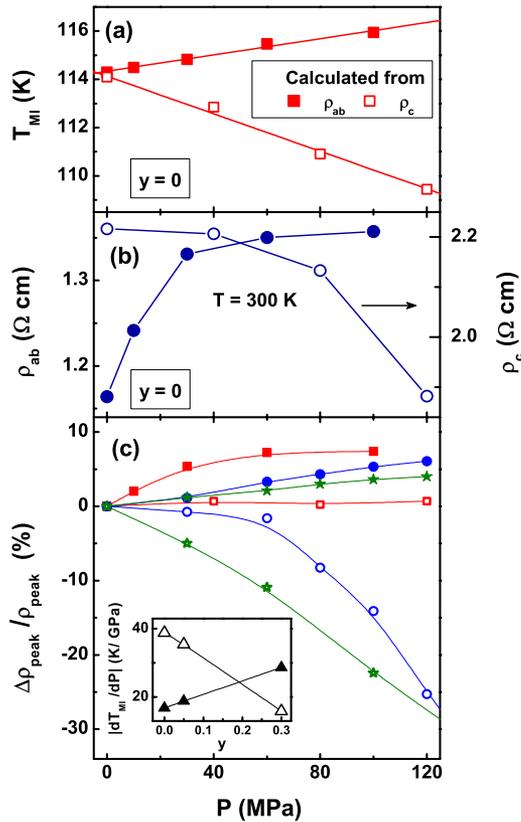


FIG. 3. (Color online) Pressure dependence of (a)  $T_{MI}$  and (b) room temperature resistivity of  $(\text{Sm}_{1-y}\text{Nd}_y)_{0.52}\text{Sr}_{0.48}\text{MnO}_3$  ( $y=0$ ) single crystal. (c) Pressure dependence of  $\Delta\rho_{\text{peak}}$  of  $(\text{Sm}_{1-y}\text{Nd}_y)_{0.52}\text{Sr}_{0.48}\text{MnO}_3$  with  $y=0$  (square), 0.05 (circle), and 0.3 (star) both for  $\rho_{ab}$  (solid symbol) and  $\rho_c$  (open symbol). Inset shows  $dT_{MI}/dP$  vs  $y$  for  $\rho_{ab}$  (solid symbol) and  $\rho_c$  (open symbol).

tinuously decreases and vanishes above a critical concentration where  $\Delta S$  will be zero and the FM transition becomes second-order in nature.<sup>12</sup>

Pressure, whether it is hydrostatic or uniaxial, changes electrical and magnetic properties of manganites mainly through lattice parameters of the system. So we try to give a plausible explanation for the unusual  $P$  dependence of  $\rho_{ab}$  and  $\rho_c$  of  $(\text{Sm}_{1-y}\text{Nd}_y)_{0.52}\text{Sr}_{0.48}\text{MnO}_3$  single crystals in terms of structural change with pressure. As the present system is very close to charge ordering insulating state, a small change in lattice parameter due to the external pressure may affect the electrical transport dramatically.<sup>12</sup> With the application of hydrostatic pressure, Mn–O bond length decreases and Mn–O–Mn bond angle increases. As these changes increase the charge transfer process, the double exchange favoring FM metallic state is enhanced. In the case of uniaxial pressure, one has to consider the small anisotropy between equatorial (Mn–O<sub>eq</sub>–Mn) and apical (Mn–O<sub>ap</sub>–Mn) bond angles. For orthorhombic structure, the departure of Mn–O–Mn bond angle from colinearity is larger along apical direction as compared to  $ab$  plane.<sup>14</sup> This is also reflected in the resistivity anisotropy at ambient pressure. So the physical properties

would be more sensitive to the change in Mn–O<sub>ap</sub>–Mn than Mn–O<sub>eq</sub>–Mn. When  $\rho_c$  is measured with  $P \perp c$ , the Mn–O<sub>eq</sub>–Mn increases and at the same time Mn–O<sub>ap</sub>–Mn decreases and Mn–O bond length along the  $c$ -axis increases due to the Poisson ratio. As the effect of change along  $c$ -axis is large and the measurement is done along the same direction, we observe quite large decrease of  $T_{MI}$ . However, there is no straightforward explanation for the increase of  $T_{MI}$  in  $\rho_{ab}(T)$  with  $P$ . For  $\rho_{ab}$  with  $P \parallel c$ , Mn–O<sub>ap</sub>–Mn increases and Mn–O bond length along  $c$ -axis decreases rapidly whereas Mn–O<sub>eq</sub>–Mn decreases and the inplane Mn–O bond length increases slowly with  $P$ . As the change along  $c$ -axis is larger than along the  $ab$  plane, the anisotropy of the system reduces with the application of pressure in the  $c$  direction. The effect of change in lattice parameters along  $c$ -axis suppresses the effect due to the decrease of Mn–O<sub>eq</sub>–Mn. As a result,  $T_{MI}$  of  $\rho_{ab}$  increases with  $P$  but at a slower rate than that for  $\rho_c$ . However, it is important to study the crystal structure under uniaxial pressure to reveal the correlation between structural and transport properties.

In summary, we have studied the temperature and uniaxial pressure dependence of  $ab$ -plane and  $c$ -axis resistivity in single crystals of  $(\text{Sm}_{1-y}\text{Nd}_y)_{0.52}\text{Sr}_{0.48}\text{MnO}_3$  with  $y=0$ , 0.05, and 0.3. For  $y=0$ , with increasing pressure along the  $c$ -axis,  $T_{MI}$  shifts toward the higher temperature at the rate of 17 K/GPa, whereas  $T_{MI}$  decreases at the rate of 39 K/GPa, when  $P$  is applied perpendicular to the  $c$ -axis. As  $y$  increases,  $dT_{MI}/dP$  increases for  $P \parallel c$  and decreases for  $P \perp c$ . These behaviors are very different from that observed in hydrostatic pressure.

One of us (A.M.) acknowledges the financial support through the fellowship from CSIR, India.

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