Effect of hydrostatic pressure and magnetic field on electrical resistance of the Kondo-like perovskite manganese oxide, \( \text{La}_{0.12} \text{Ce}_{0.4} \text{Sr}_{0.5} \text{MnO}_3 \) with a ferrimagnetic ground state, have been investigated up to 2.1 GPa and 9 T. In this compound, the Mn-moments undergo double exchange mediated ferromagnetic ordering at \( T_C \sim 280 \) K and there is a resistance maximum, \( T_{\text{max}} \) at about 130 K which is correlated with an antiferromagnetic ordering of cerium with respect to the Mn-sublattice moments. Under pressure, the \( T_{\text{max}} \) shifts to lower temperature at a rate of \( -dT_{\text{max}}/dP = -162 \text{ K/GPa} \) and disappears at a critical pressure \( P_c = 0.9 \text{ GPa} \). Further, the coefficient, \( m \) of \( -\log T_{\text{max}} \) term due to Kondo scattering decreases linearly with increase of pressure showing an inflection point in the vicinity of \( P_c \). These results suggest that cerium undergoes a transition from Ce\(^{4+} \) state to Ce\(^{3+} / \text{Ce}^{4+} \) mixed valence state under pressure. In contrast to pressure effect, the applied magnetic field shifts \( T_{\text{max}} \) to higher temperature presumably due to enhanced ferromagnetic Mn moments.

PACS numbers: 72.64.Fj, 72.15.Eb, 75.30.Mb
teractions between the localized Ce:4f and ferromagnetic Mn moments, we have investigated a simultaneous and individual effects of hydrostatic pressure and magnetic field on the behavior of electrical resistance in La$_{0.1}$Ce$_{0.4}$Sr$_{0.5}$MnO$_3$ within a temperature range of 4.2 K – 300 K. We use this approach because our earlier ambient-pressure work clearly established that the resistivity features are correlated with magnetization and neutron diffraction data. From this study, we substantiate that the anomalous temperature dependence of electrical resistance is due to Kondo-like scattering of Mn:Ce$^4$ electrons by localized Ce:4f moments. Further, we infer that there is a valence change of cerium under pressure from Ce$^{3+}$ state to Ce$^{4+}$/Ce$^{5+}$ mixed valence state.

Polycrystalline sample of La$_{0.1}$Ce$_{0.4}$Sr$_{0.5}$MnO$_3$ was prepared by calcining stoichiometric mixtures of La$_2$O$_3$, CeO$_2$, SrCO$_3$, Mn$_2$O$_3$ at 1100°C and sintering at 1500°C. Phase purity, nuclear and magnetic structures were determined by powder x-ray and neutron diffraction methods. The electrical resistance was measured by a standard four-probe method with a direct current of 1 mA down to 4.2 K. The electrical contact was made with a silver paint of heat treatment type. Hydrostatic pressure up to 2.1 GPa was generated by using a CuBe piston-cylinder device and 1:1 mixture of Fluorinert FC70 and FC77 as a pressure medium. The pressure was changed at RT to minimize internal strain in the specimen and the load was controlled to within ±1 % through-out the measurement. The magnetic field was applied by a superconducting magnet up to 9 T. The details of the present apparatus at multi-extreme conditions have been reported previously.

Temperature dependence of electrical resistance at various pressures is shown in Fig. 1. In this figure, there is no indication for the DE ferromagnetic metallic transition near $T_C \sim 280$ K. However, we have already reported$^{2}$ from the temperature derivative of $R$ that, in this system, the $T_C$ increases with pressure at a rate of $dT_C/dP = +1.9$ K/GPa, as shown in Fig.2. The small increase of $T_C$ is consistent with the fact that this system is in a weak coupling region due to a large filling of conduction band, where the system is less sensitive to pressure or change in $W$. At 0.1 GPa, $R$ increases logarithmically ($-\log T$) with decreasing temperature in the range $\sim 180 - 230$ K as shown by solid line in Fig. 1(a) and exhibits a broad maximum at about 130 K ($T_{max}$) and a minimum at about 30 K ($T_{min}$) which are shown by arrows in Fig.1. As discussed earlier, the $T_{max}$ is due to the onset of antiferromagnetic ordering of Ce$^{3+}$ moments with respect to the ferromagnetic Mn moments. This $R(T)$ behavior is reminiscent of the typical concentrated Kondo compounds.$^{12,13}$ However, the origin of $T_{max}$ in the present case should be compared and contrasted with the concentrated Kondo systems where the origin of resistivity maximum is due to crystal field effects and Kondo coherence, and the $T_{max}$ has a large positive pressure coefficient$^{13,14}$ Further, the decrease of resistivity below Ce ordering is not sharp such as one normally observes at a magnetic ordering of Ce in intermetallics. This is because, the antiferromagnetic coupling in the present system is between Ce and Mn moments and the antiparallel arrangement of Ce is induced by Mn moments. In fact, a recent theoretical analysis on this system reproduce the magnetic behavior of Mn and Ce in this system.$^{16}$

In the present case, with increasing pressure, $T_{max}$

![FIG. 1: Temperature dependence of the electrical resistance $R(T)$ of La$_{0.1}$Ce$_{0.4}$Sr$_{0.5}$MnO$_3$ at high pressures. The downward and upward arrows indicate $T_{max}$ and $T_{min}$, respectively. $T_C$ is nearly 280 K. Inset shows the shift of $T_{max}$ to a higher temperature under a magnetic field of 7 T and P = 0 GPa.](image)

![FIG. 2: Pressure dependence of $T_{max}$ and $T_{min}$ at B = 0 T as shown by solid circle and triangle, respectively. The solid, dashed and dotted lines are guides for the eye. $P_c$ is the value of pressure corresponds to the extrapolation of $T_{max}$ to zero temperature.](image)
cerium corresponds to the ordering temperature of 15 cerium indicates a valence change of cerium. In manganites, for example, CeAl$_2$ shows variation of R(T) with pressure up to 0.8 GPa, close to $P_c$, and then decreases with further increase of pressure. Since the pressure effect on the DE ferromagnet with larger W does not have any contribution to R at low temperatures, the increase of R suggests that the strength of J, the exchange integral, which depends on the hybridization between the conduction electron and f-electron, increases with pressure. The decrease of R above $P_c$ may be due to decrease of $T_{max}$. In the intermediate region (90 $\leq T$ $\leq$ 250) the large decrease of R with increase of pressure is mainly due to suppression of spin dependent scattering that results from the valence change of cerium. The fact that the R(T) does not become a metallic like even at 2.1 GPa suggests that the cerium has not completely changed to Ce$^{4+}$ state and rather exists in Ce$^{4+}$/Ce$^{3+}$ mixed valence state. Fig. 3 shows variation of R at various pressures as a function of temperature in logarithmic scale. It can be seen that there is a region of $-\log T$ dependence of R at high temperatures (> 100 K) as shown by solid line at all pressures. This is a signature of Kondo scattering of conduction electrons by the localized moments. The coefficients of $-\log T$ term is termed as m. In Fig. 3(b), normalized coefficient $m(P)/m(0.1)$ plotted as a function of pressure, where $m(P)$ is defined as $m=-\partial R/\partial \log T$, and $m(0.1)$ is that at 0.1 GPa. The coefficient, $m(P)/m(0.1)$ decreases linearly with increase of pressure because of the change in valence state of cerium as discussed above. This behavior is also different qualitatively from that of the typical Kondo compounds. Near $P_c$ $\sim$ 0.9 GPa, a small inflec-

It is intriguing and important to note the change of R(T) at various pressures as a function of temperature (see Fig. 4). Near $T_c$, the decrease of R with increase of pressure is due to: (i) widening of $\epsilon_g$ band as reported for La$_{1-x}$Sr$_x$MnO$_3$ and (ii) valence change of cerium. On the other hand, at the lowest temperature measured (4.2 K), R increases with pressure up to 0.8 GPa, which is in sharp contrast to the previous data of magnetic field (B) shifts the $T_{max}$ decreases to a lower temperature at a rate of $dT_{max}/dP$ = -162 K/GPa, which is in sharp contrast to the typical concentrated Kondo systems, for example, CeAl$_2$ or CeInCu$_2$ where the $T_{max}$ increases with pressure. On the other hand, $T_{min}$ seems to shift to higher temperature as shown in Fig. 4(a). At pressures higher than 0.8 GPa both $T_{max}$ and $T_{min}$ are not seen down to 4.2 K. It should be noticed from the Fig. 4(a) that the extrapolated value of pressure, where the $T_{max}$ equals to zero, is 0.9 GPa, which we termed as critical pressure $P_c$. Since the $T_{max}$ corresponds to the ordering temperature of cerium, the $P_c$ might indicate the suppression of magnetic ordering of cerium. According to the effect of hydrostatic pressure on DE ferromagnetism, we would expect that the $T_{max}$ associated with Ce ordering should increase with pressure. The observed decrease of $T_{max}$ or the magnetic ordering temperature of cerium with increase of pressure rather indicates a valence change of cerium from Ce$^{3+}$ towards Ce$^{4+}$ state. In contrast to pressure effect, the application of magnetic field (B) shifts the $T_{max}$ to higher temperature due to enhanced ferromagnetic Mn moments, as shown in the inset of Fig. 4(b) for an applied field of 7 T. The presence of resistivity minimum in the vicinity of 25 K in manganites is well known and has been attributed to the effect of electron-electron interactions.

**FIG. 3:** (a) Temperature dependence of R, in which the abscissa is selected as a logarithmic scale and $-\log T$ dependence is shown by solid lines. (b) Pressure dependence of the normalized coefficient $m(P)/m(0.1)$ of $-\log T$ term. The previous data of $m(P)/m(0.1)$ is described as closed circle. $P_c$ is defined as an inflection point of the coefficient. The dotted and solid lines are guides for the eye.

**FIG. 4:** Pressure dependence of negative MR ratio ($=\Delta R/R(B) \times 100 \%$) at 4.2 K. The MR ratio at 9 T is plotted in the inset as a function of pressure. A solid curve is guide for the eye.
tion is seen and above which the effect of pressure on $m$ becomes smaller, indicating a decrease in the strength of the Kondo scattering above $P_c$. This is consistent with the observation that the $T_{\text{max}}$ disappears at $P_c$, where one would expect a change in the interaction between the Ce:4$f$ local and the conduction electron moments.

Magnetoresistance (MR) ratio $(\Delta R/R(B) = (R(0) - R(B))/R(B))$ at various pressures at 4.2 K is plotted as a function of $B^2$ in Fig. 4. The negative CMR effect in perovskite manganese oxides is well known as reported in previous experiments. In the present experiment, the negative MR monotonically increases with applying $B$ which is mainly due to suppression of Kondo-like scattering and a small contribution from grain boundary scattering, as the DE interactions in large $W$ materials results in MR effect only near $T_c$. Above 5 T, MR lineary increases with respect to $B^2$, revealing the existence of spin-dependent scattering. The inset shows MR ratio at 4.2 K and at a field of 9 T as a function of pressure: MR increases with pressure and a peak is clearly seen near $P_c \sim 0.9$ GPa. This is consistent with the observation that the pressure dependence of $R$ at 4.2 K has similar behavior, indicating the electronic changes associated with the valence change of cerium.

The pressure dependence of $R(T)$ ($T < 100$ K) under a magnetic field of $B = 5$ T is shown in Fig. 5. By applying 5 T, $T_{\text{min}}$ shifts from 30 K to 40 K at 0.1 GPa. This phenomenon was explained by the enhancement of cerium ordering induced by the ferromagnetic manganese moments. In contrast to zero applied field, $R$ at 4.2 K and 5 T increases with increasing pressure without showing any maximum in the present pressure range. It is possible that $P_c$ exceeds the maximum pressure limit in our experiment because the ordering temperature of cerium increases with applied magnetic field. $T_{\text{min}}$ increases with pressure similar to that observed under zero field as shown Fig. 4. It would be interesting to carry out neutron diffraction experiment under pressure to see the changes in the ordering temperature of cerium.

In conclusion, we have found that under zero applied magnetic field, the $T_{\text{max}}$ due to magnetic ordering of cerium and normalized coefficient of $-\log T$ term, $m(P)/m(0.1)$ originated from Kondo-like scattering of conduction electrons decrease with increasing pressure. Further, the $T_{\text{max}}$ disappear at a critical pressure ($P_c \sim 0.9$ GPa) where there is a corresponding change in the slope of the coefficient of the $-\log T$ term, indicating a decrease in the strength of the Kondo-like scattering. The suppression of $T_{\text{max}}$ has been attributed to a valence change of cerium from Ce$^{3+}$ state to Ce$^{4+}$/Ce$^{3+}$ mixed valence state under pressure.

---

* Electronic address: sundaresan@jncasr.ac.in

1. Colossal Magnetoresistance, Charge Ordering and Related Properties of Managanese Oxides, edited by C. N. R. Rao and B. Raveau (World Scientific, Singapore, 1998).
2. A. P. Ramirez, J. Phys. Cond. Matter. 9, 8171 (1997), and references therein.
3. G. H. Jonker, Physica (Amsterdam) XVI, 337 (1950).
4. C. Zener, Phys. Rev. 82, 403 (1951).
5. P. W. Anderson and H. Hasegawa, Phys. Rev. 100, 675 (1955).
6. P. -G. de Gennes, Phys. Rev. 118, 141 (1955).
7. A. Sundaresan, V. Caignaert, A. Maignan, B. Raveau, E. Suard, Phys. Rev. B60, 533 (1999).
8. A. Sundaresan, J. L. Tholence, A. Maignan, B. Raveau, E. Suard, P. Bordet, J. Magn. Magn. Mater. 226-230, 777 (2001).
9. T. Eto, G. Oomi, F. Honda, A. Sundaresan, J. Magn. Magn. Mater. 226-230, 879 (2001).
10. F. Honda, S. Kaji, I. Minamitake, M. Ohashi, G. Oomi, T. Eto, and T. Kagayama, J. Phys. Cond. Matter. 14, 11501 (2002).
11. F. Hanzawa, K. Yamada, K. Yoshida, J. Magn. Magn. Mater. 47 & 48, 357 (1985).
12. T. Kagayama, G. Oomi, H. Takahashi, N. Möri, Y. Önuki, T. Komatsubara, Phys. Rev. B44, 7690 (1991).
13. T. Kagayama, G. Oomi, J. Phys. Soc. Jpn. 65, Suppl. B 42 (1996).
14. G. Oomi, T. Kagayama, Y. Önuki, H. Mitamura, T. Goto, Proc. Moscow Inter. Sympo. Magn (MISM’99), 351 (1999).
15. Y. Morimoto, A. Asamitsu, and Y. Tokura, Phys. Rev. B 51, 16491 (1995).
16. G. Martinez, J. R. Iglesias, A. R. Ruppenthal, J. M. Avigno, C. Lacroix, J. Magn. Magn. Mater. 226-230, 214 (2001).
17. D. Kumar, J. Sankar, J. Narayan, Rajiv K. Singh, A. K. Majumdar, Phys. Rev. B 65, 94407 (2002).