Distinct origins of magnetic-field-induced resistivity irreversibility in two manganites with similar ground states:

Pr$_{0.5}$Sr$_{0.41}$Ca$_{0.09}$MnO$_3$ and La$_{0.5}$Ca$_{0.5}$MnO$_3$

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(March 22, 2022)

Abstract

Our investigation of the magnetotransport in two charge ordered manganites with similar magnetic ground states reveals that the origin of magnetoresistance cannot be concluded from the isofield resistivity, $\rho(T, \text{constant } H)$, measurements alone. Both Pr$_{0.5}$Sr$_{0.41}$Ca$_{0.09}$MnO$_3$ (PrSrCa) and La$_{0.5}$Ca$_{0.5}$MnO$_3$ (LaCa) show a ferromagnetic transition ($T_C = 260$ K for PrSrCa, 230 K for LaCa) followed by an antiferromagnetic transition ($T_N = 170$ K for PrSrCa, 140 K for LaCa). These compounds show qualitatively similar magnetotransport: Below the irreversibility temperature $T_{IR}$, field cooled (FC) resistivity is lower than zero field cooled (ZFC) and decreases continuously with T, whereas the ZFC $\rho(T, H)$ resembles the behavior of $\rho(T, H = 0 \ T)$. The value of $\rho(\text{ZFC})/\rho(\text{FC})$ is $\approx 10^4$ at 5 K and $\mu_0H = 7 \ T$ in both compounds. However, isothermal magnetic measurements suggest distinct origins of magnetoresistance: Field cooling enhances ferromagnetic phase fraction in LaCa whereas it drives PrSrCa into a metastable state with high magnetization. The distinct origins of magnetotransport is also reflected...
in other magnetic history dependent properties.
I. INTRODUCTION

There has been a burst of interest in RE$_{1-x}$AE$_x$MnO$_3$-type manganites (RE = La$^{3+}$, Pr$^{3+}$, Nd$^{3+}$ etc., AE = Ca$^{2+}$, Sr$^{2+}$, Ba$^{2+}$, etc.,) because of their extraordinary sensitivity of resistivity ($\rho$) to internal molecular and external magnetic fields. The $\rho$(T) evolution in general shows a metallic like behavior ($d\rho/dT$ > 0) in the ferromagnetic (FM) state and an insulating like behavior ($d\rho/dT$ < 0) in the antiferromagnetic (AF) state. The essential physics of electrical transport in manganites is understood in terms of the Double Exchange (DE) interaction which allows spin conserving transfer of e$_g$ hole along Mn$^{3+}$-$t_{2g}^3$e$_g^1$-O$^{2-}$-Mn$^{4+}$-$t_{2g}^3$e$_g^0$ network if the core $t_{2g}$ spins are aligned ferromagnetically and forbids this hole exchange if the $t_{2g}$ spins are antiparallel. In addition, A-site ionic radius ($<r_A>$) and size mismatch ($\sigma^2$) of the A-site cations, orbital and charge ordering, and electron-phonon coupling can enhance the magnitude of the resistivity. A large decrease of $\rho$(T) under an external magnetic field occurs close to the magnetic phase boundary: the ferromagnetic-paramagnetic phase boundary shifts up and the charge ordered, antiferromagnetic-ferromagnetic or paramagnetic phase boundary shifts down in temperature with increasing field. There is no clear consensus about the origin of magnetoresistance (MR) although the basic mechanism can be traced to the field induced ferromagnetic alignment of $t_{2g}^3$ spins and the resulting delocalization of the e$_g$-carriers. Structural transition, electron-spin-phonon coupling, melting of charge and orbital order, and phase separation which all can be magnetically tunable appear to play an important role in determining the magnitude of the magnetoresistance.

The magnitude of MR is path dependent in some of the manganites: MR is lower when the sample is cooled in zero field (ZFC) prior to establishing a magnetic field at low temperature than when cooled in a field (FC) from high temperature. Only some investigators have paid attention to this aspect. However, its origin is not understood yet. Although the temperature $T_{IR}$ below which the ZFC and FC $\rho$(T) bifurcate generally shifts down in temperature with increasing H, an exceptional case of upward shift of $T_{IR}$ with H was also found.
The most elaborate study under different values of magnetic field was carried out by Xiao et al.\textsuperscript{7} on La_{0.5}Ca_{0.5}MnO_{3}. The ZFC and FC measurements reported so far were done in an isofield temperature scan mode, i.e., temperature dependence of $\rho$ under a field, but this method masks various contributions to the irreversibility. Hence, we investigated electrical and magnetic properties of La_{0.5}Ca_{0.5}MnO_{3} (LaCa) and Pr_{0.5}Sr_{0.41}Ca_{0.09}MnO_{3} (PrSrCa) in detail. Both of these compounds have similar magnetic properties: ferromagnetic transition at high temperature ($T_C = 240$ K for LaCa, 265 K for PrSrCa) and charge ordering and antiferromagnetic transition at low temperature ($T_N = 170$ K, $T_N = 140$ K $\leq T_{CO}$ for LaCa, $T_N = T_{CO} = 170$ K for PrSrCa). The $\rho(T)$ of PrSrCa compound is metallic like ($d\rho/dT > 0$) in between $T_C$ and $T_N$ whereas it is insulating in LaCa. We will show that the origin of the resistivity irreversibility in these two magnetically identical compounds are different although they look the same from an isofield temperature scan.

**II. EXPERIMENT**

The polycrystalline Pr_{0.5}Sr_{0.41}Ca_{0.09}MnO_{3} (PrSrCa) and La_{0.5}Ca_{0.5}MnO_{3} (LaCa)samples were prepared by the standard ceramic route and were characterized by neutron diffraction, electron diffraction and transport measurements.\textsuperscript{12,13} We remeasured the four probe resistivity of polycrystalline PrSrCa and LaCa using Quantum Design made Physical Property Measuring System in different field modes: In the zero field cooled (ZFC) and field cooled (FC) modes, $\rho(T)$ was measured while warming from 5 K in a field $H_1$ after cooling in $H = 0$ and $H = H_1$ respectively. Resistivity isotherms at 5 K in ZFC and FC were also measured. In the FC(7T)-FW(H) mode, the sample was cooled from $T = 300$ K to 5 K in a high field of 7 T, then the field was reduced to a lower H value at 5 K and data were taken while warming in H. In the FC(H)-FW(0T) mode, the sample was cooled from $T = 300$ K to 5 K under a high field, the field was reduced to zero at 5 K and data were taken while warming in zero field. Ac susceptibility in $H_{ac} = 1$ Oe and $f = 100$ Hz was also measured in 300 K-5
K temperature range.

III. RESULTS

Fig. 1(a) and Fig. 1(b) show the temperature dependence of $\rho(T, H = 0 \, \text{T})$ of PrSrCa, and LaCa upon cooling and warming. The insets shows the real part ($\chi'$) of the ac susceptibility. The ferromagnetic and antiferromagnetic transitions are marked. The $\rho(T, 0)$ of PrSrCa exhibits a metallic like behavior in between $T_C$ and $T_N$, and it jumps abruptly at $T_N (= 170 \, \text{K})$. While cooling, the value of $\rho$ increases by 7 orders of magnitude in between $T_N$ and 5 K. The $\rho(T, 0)$ decreases rapidly as $T_N$ is approached from below and $T_N$ shifts up to 180 K upon warming. The charge and antiferromagnetic ordering in PrSrCa occur at the same temperature. A pronounced hysteresis in $\rho(T, 0)$ and $\chi'(T)$ occur around the Neel temperature. The $\rho(T, 0)$ behavior of LaCa is similar to that reported by other groups. In contrast to PrSrCa, $\rho(T, 0)$ of LaCa does not exhibit a metallic behavior in between $T_C$ and $T_N$, which is otherwise expected according to double exchange mechanism. The $\rho(T, 0)$ increases abruptly around 140 K while cooling and increases by more than 6 orders of magnitude in between 140 K and 5 K. Below 30 K, $\rho(T, 0)$ becomes asymptotic to the temperature axis as seen previously. While warming $\rho(T, 0)$ exhibits a hysteresis over a wide temperature range (90 K-220 K) and also is the susceptibility (see the inset in Fig. 1(b)). Although the charge and antiferromagnetic orderings in LaCa are generally assumed to occur simultaneously, our results to be presented latter indicate that charge ordered clusters are present already at 220 K (i.e., in the ferromagnetic regions) even while cooling from 300 K.

Fig. 1(c) and Fig. 1(d) show $\rho(T, H)$ in the ZFC and FC modes for PrSrCa and LaCa respectively. We can clearly see an irreversibility between the FC and ZFC curve for all field value. The irreversibility starts at a temperature ($T_{IR}$) close but below $T_N$. The $T_{IR}$ is determined from the temperature $[\rho(\text{ZFC})-\rho(\text{FC})]/\rho(\text{ZFC}) \leq 3 \%$. $T_{IR} = 160 \, \text{K}$ (155
K), 150 (150 K), 133 K (149 K) for $\mu_0 H = 2$ T, 4 T, 7 T for PrSrCa (LaCa)]. Below $T_{IR}$, the ZFC- $\rho(T,H)$ continues to rise and reaches a higher value than the FC- $\rho(T)$ which either ascends or descends with decreasing T depending upon the field strength. The temperature dependence of the ZFC and FC curves of PrSrCa and LaCa are qualitatively similar. The ZFC-$\rho(T,H))$ in both compounds mimics the temperature dependence of $\rho(T, 0)$ curve albeit. At 5 K and at 7 T the resistivity ratio $\rho(ZFC)/\rho(FC) = \approx 7.5 \times 10^4$ for PrSrCa is comparable to $\rho(ZFC)/\rho(FC) = \approx 3.8 \times 10^4$ for LaCa. Thus, from the isofield measurement alone it appears that a single mechanism is responsible for the irreversibility behavior.

In order to understand the origin of the irreversibility in details, we studied the field dependence of the resistivity, and the magnetization. Figures 2(a) and (b) show isothermal scans for PrSrCa and LaCa respectively at $T = 5$ K. When H is increased from 0 T to 7 T in the zero field cooled mode, $\rho(T = 5$ K, H) of PrSrCa decreases rather smoothly by almost 3 orders of magnitude and when H is reduced to zero $\rho(5$ K, H) does not attain the original starting value. Upon further cycling from 0 T $\rightarrow$ -7 T $\rightarrow$ 0 T, $\rho$ exhibits hysteresis but the loop is closed. The field cycling process in the FC mode is H $\rightarrow$ 0 T $\rightarrow$-H $\rightarrow$ 0 T $\rightarrow$ H. The field cooled resistivity at any value of the field is clearly lower than the corresponding zero field cooled value. The hysteresis loop is open in the positive field cycle with lower resistivity in H $\rightarrow$ 0 T branch than 0 T $\rightarrow$ H branch. In the ZFC mode, $\rho(5K, H)$ of LaCa also decreases rather smoothly up to the maximum field of $\mu_0 H = 7$ T. But the field dependence (see the curvature) is different from LaCa. It also exhibits hysteresis like the PrSrCa compound but the loop is not closed when H is increased from -5 T to 0 T. When the sample is field cooled, the hysteresis is negligible compared to PrSrCa.

Figures 2(c) and 2(d) compare M(H) of PrSrCa and LaCa respectively. In the ZFC mode, M(H) of PrSrCa shows a weak ferromagnetic like behavior for $\mu_0 H < 0.5$ T and then increases with a constant slope up to the maximum field of 5 T. The maximum magnetic moment at 5 T in the ZFC mode is 0.185 $\mu_B$ which is only 5.3 % of the maximum moment of 3.5 $\mu_B$. 
expected if the whole sample were ferromagnetic. When the sample is field cooled under 5 T, M at 5 T increases to 0.483 $\mu_B$ which is nearly 2.6 times larger than the corresponding field value in the ZFC mode. The M(H) loop is symmetric about the origin when the field is cycled between -5 T and +5 T after the virgin 5 T $\rightarrow$ 0 T curve. But, the value at $\mu_0H = 5$ T decreases to 0.3 $\mu_B$ when the field is increased from -5 T to +0.5 T. We have carried out M(H) under FC mode for $\mu_0H = 4$ T, and 3 T. Since the behaviors are qualitatively the same as 5 T data we do not show them here. The M(H) of LaCa (Fig. 2d) is in many sense contrasting to PrSrCa. The ZFC curve exhibits a ferromagnetic like behavior but with a large moment at 0.5 T. The value M at 0.5 T is 0.18 $\mu_B$ for LaCa whereas it is only 0.04 $\mu_B$ for PrSrCa. At 5 T, M of LaCa is 0.38 $\mu_B$ which is twice as large as PrSrCa. A small hysteresis is seen only in the positive field cycle. When LaCa is field cooled under 5 T, M at 5 T increases to 0.85 $\mu_B$ however, unlike in PrSrCa, M does not reduce to low value at 5 T after $5T \rightarrow 0T \rightarrow -5T \rightarrow 5T$ cycle. More importantly we see a large enhancement of the low field magnetization in LaCa compared to PrSrCa. The spontaneous magnetic moment $M_0 = 0.68 \mu_B$ (determined by extrapolating the linear high field part to $H = 0$) decreases with the strength of the cooling field. The presence of a ferromagnetic phase below the charge ordering temperature is in agreement with the results inferred from other techniques$^{13,17}$ and supports the phase separation scenario proposed theoretically$^6$. The volume fraction of the ferromagnetic phase in LaCa can be calculated from $f_m = M_0/M_S$ where $M_S = 3.5 \mu_B$ and $M_0$ is the spontaneous magnetic moment. The $f_m$ in LaCa increases from 4.95 % in the ZFC mode to 19.69 % when field cooled in 5 T. These are only rough estimates without considering canting of moments in the antiferromagnetic phase. When extrapolated from the recently published data on Pr$_{0.5}$Sr$_{0.5-x}$Ca$_x$MnO$_3$ (x = 0.1, 0.2)$^5$, the low field behavior M(H) in our PrSrCa compound can be ascribed to few ten nanometer size ferromagnetic clusters ($f_m < 0.8$ %) which can behave like a superparamagnetic entity. These observations underline distinct origins of the path dependent magnetoresistance in these two magnetically identical compounds.
The above presented zero field cooled data in both compounds do not indicate any evidence of a field induced antiferromagnetic-ferromagnetic (metamagnetic) transition up to 5 T which is otherwise expected. To know whether we are dealing in the field range far below the critical field for metamagnetic transition, we carried out M(H) measurement at different temperatures and for higher field values (µ₀H >5 T) in particularly for PrSrCa. Magnetization up to 12 T for PrSrCa sample was measured using a vibrating sample magnetometer at University of Zaragoza, Spain. Fig. 3 (a) shows M(H) data for PrSrCa. The measurements were done in zero field cooled mode. The M(H) curve at 210 K is a typical of a long range ferromagnet. But for lower temperatures, the M(H) , for example at T = 150 K, initially increases linearly but then jumps abruptly to a higher value around the threshold field Hₐ = 4 T and at H >>Hₐ, M(H) appears to saturate. The transition is first order as can be guessed from the hysteretic behavior. The abrupt jump characterize the metamagnetic transition from the antiferromagnetic state to either a spin flop or a spin flip state. For H >>Hₐ, the sample is ferromagnetic and for H <Hₐ, the increase of M(H) is due to canting of antiferromagnetic moment away form the direction of the spin axis towards the field direction. The critical field for the metamagnetic transition increases from 4 T at 150 K to 8 T at 75 K and to more than 11 T at 25 K.

We also carried out M(H) for LaCa but the field was restricted to the maximum available field of 5 T in the SQUID magnetometer at Laboratoire CRISMAT, Caen, but the available field range (0 T -5 T) is sufficient for the purpose of this paper. The results are shown in Fig. 3(b). At T = 150 K ≈ T_N, M(H) behaves like a ferromagnet until µ₀H = 2 T, but then increases rapidly above 3.5 T due to the metamagnetic transition. The metamagnetic transition is not completed at 5 T. At 5 T, the magnetic state can be characterized as a mixture of magnetic field induced ferromagnetic domains (in addition to ferromagnetic phase present in zero field) and antiferromagnetic domains with canted moments. From the spontaneous magnetization M₀ = 0.6 µ_B we calculate the volume fraction of the ferromagnetic phase fₘ = 17.15 %. At 125 K, fₘ decreases to ≈ 5.7 % and
\[ \approx 4.95 \% \text{ at } 5 \text{ K.} \] The M(H) curve of LaCa at 5 K is more like ferromagnet because the metamagnetic transition takes place at much higher fields.

Surprisingly, M(H) data at \( T = 175 \text{ K} \) which is above \( T_N = 140 \text{ K} \) but below \( T_C = 230 \text{ K} \) also show a metamagnetic transition above \( \mu_0H = 1 \text{ T} \) with hysteresis. However, the metamagnetic transition is not as sharp at 150 K or as in PrSrCa compound. The M(H) data at 200 K also closely resemble the 175 K but with a very small hysteresis. We suggest that charge and orbital ordered domains exist above the Neel temperature in contrast to the assumption that charge ordering and antiferromagnetic orderings occur simultaneously.\[ \] The metamagnetic transition in this temperature range is caused by the field induced destruction of the charge and orbital order. The delocalization of \( e_g \)- carriers enhances ferromagnetic ordering of \( t_{2g}^3 \) spins and so the magnetization increases. The broad metamagnetic transition suggests that domains of variable sizes are present. Some indirect evidence for charge ordering in the ferromagnetic region can be quoted from the very recent literature: A large positive volume magnetostriction above \( T_N \) in \( \text{La}_{0.5}\text{Ca}_{0.5}\text{MnO}_3 \) was reported earlier by us\[ \] and it was suggested that charge ordered phase of lower unit cell volume coexist with a ferromagnetic phase of higher unit cell volume in between \( T_C \) and \( T_N \). The ferromagnetic phase having higher unit cell volume than the charge ordered phase was recently confirmed by Huang et al.\[ \] (see Fig. 7 in ref. 10) who also found formation of a new structural phase around 220 K in \( \text{La}_{0.5}\text{Ca}_{0.5}\text{MnO}_3 \). The new structural phase is most likely charge and orbital ordered although Huang et al.\[ \] did not explicitly mention it. The charge-orbital domains are most likely paramagnetic\[ \] in this temperature range since no antiferromagnetic signal was detected in between 220 K and 150 K.\[ \] As the temperature is lowered the charge ordered domains grow in size coalesce around \( T = 140 \text{ K} \) below which \( \rho(T = 0) \) starts to increase rapidly. A long range antiferromagnetic ordering also takes place around 140 K. A question remains to be unanswered is why the resistivity does not show metallic behavior in between \( T_C \) and \( T_N \). A possibility is that charge ordered domains above \( T_N \) but below \( T_C \) are sandwiched between ferromagnetic domains and hence impedes charge transfer be-
tween ferromagnetic domains. However, we cannot arrive any definitive conclusion from the present study alone and future work should address this issue.

IV. DISCUSSION

Now we turn our attention again towards the origin of the field induced irreversibility in resistivity. The magnetization study suggests that the ferromagnetic phase fraction at 5 K in LaCa (fm $\approx 4.5\%$) is larger than in PrSrCa (fm $\approx 0.8\%$). It is likely that the ferromagnetic phase is concentrated in some regions of the sample in LaCa and is randomly distributed in PrSrCa. The difference in the curvature of the virgin ZFC - $\rho$(H) curves below 2 T in Fig. 2(a) and 2(b) suggests such a possibility. When the sample is zero field cooled, the magnetization of the ferromagnetic domains is randomly oriented in LaCa and as H is increased from 0 T to 2 T, after domain wall motion, ferromagnetic domains reorient in the field direction. This causes a rapid decrease of the resistivity in LaCa in this field range. The spin canting of the antiferromagnetic sublattices at higher fields (H $> 2$ T) can also contribute to the magnetization. The main source of the magnetoresistance in this field regime in the zero field is tunnelling of the spin polarized carriers between the ferromagnetic domains and between ferromagnetic domains and canted antiferromagnetic phase. In PrSrCa, the dominant contribution to MR in the zero field mode is the canting of antiferromagnetic sublattice. The ZFC behavior of $\rho$(H) at 5 K in both PrCa and LaCa suggests that metamagnetic transition does not take place up to 7 T. Field cooling has two effects. First, it gives a preferential orientation of M of ferromagnetic domains which already exist in LaCa. Second, new ferromagnetic domains are formed at high temperature due to the destruction of the charge ordered domains. The second contribution is more effective in LaCa than in PrSrCa because charge ordered domains of various sizes and hence different critical fields (H$_C$) are already present above the Neel temperature in LaCa. This new ferromagnetic phase continues to exist down to the lowest temperature in LaCa. Hence, the ferromagnetic phase fraction increases with the strength of the cooling field in LaCa and
so, $\rho$ is lowered in the field cooled mode. If the ferromagnetic domains form a percolating network ($H \geq 2\ T\$), not necessarily to be a 3 dimensional network but can be filamentary\textsuperscript{6}, then the temperature dependence of $\rho$ is mainly determined by the thermal dependence of the magnetization of the ferromagnetic phase and so $\rho(T)$ increases with increasing temperature. There are two possible scenarios in the case of PrSrCa. It appears that field cooling does not enhance the ferromagnetic phase fraction in PrSrCa if one interprets the magnetization data (Fig. 2(a)). So, the first possibility is that the high field state is metastable with a majority antiferromagnetic domains with nonzero net magnetisations coexisting with nanometric ferromagnetic clusters. In the double exchange picture\textsuperscript{2}, the magnetization due to canting between two neighboring Mn$^{3+}$ and Mn$^{4+}$ sites is $M = M_s \cos(\theta/2)$ where $\theta$ is the canting angle and $M_s$ is the saturation magnetization. From the observed value of $M = 0.483\ \mu_B$ at 5 T and at 5 K in the virgin FC curve (see Fig. 2(c)), we calculate $\theta/2 = 82.06^\circ$ between the antiferromagnetically coupled sublattice. which is reduced from $\theta/2 = 90^\circ$ in the uncanted scenario. After field cycling $M$ reduces to 0.2903 $\mu_B$, and the canting angle increases to $\theta/2 = 85.25^\circ$ and in the zero field cooled mode the canting angle is still higher ($\theta/2 = 86.97^\circ$). Thus, the low resistivity in the field cooled mode in PrSrCa is due to charge transport through the canted antiferromagnetic domains rather than by the increase of ferromagnetic phase fraction. Another possibility is that field cooling ($H \geq H_C$) partially destroys the antiferromagnetic and charge ordering and creates bigger ferromagnetic clusters and antiferromagnetic, charge ordered domains. The low resistivity in the field cooled mode can be understood as the result of the percolation of the ferromagnetic clusters. When the field is reduced from 5 T, the magnetization of ferromagnetic clusters becomes random and hence the spontaneous magnetization is not apparent in the $M(H)$ curve. From the magnetization measurement alone it is not possible to distinguish between the above two possibilities.

These subtle differences between these two compounds are also reflected in other mea-
surements. Figures 4(a) and 4(b) compare the ρ(T) behaviors of PrSrCa and LaCa, respectively, in the FC (H)-FW(0T) mode. The samples were first cooled rapidly from 300 K to 5 K in a field H, H was reduced to zero rapidly (250 Oe/sec) and data were taken while warming in zero field. It can be seen that after reducing H from 7 T to 0 T, the value of ρ at 5 K in PrSrCa is more than 3 orders of magnitude higher than in LaCa. This reflects the fact that the field cooled state has a longer relaxation time (more stable) in LaCa than in PrSrCa which is in agreement with the low field magnetic behavior. When LaCa is heated from 5 K, the FC (7 T)-FW(0T) curve exhibits a maximum at $T_{\text{max}} = 75$ K, below this temperature, ρ(T) keeps 'memory' of the low resistive state at 7 T and lost its 'memory' above 75 K. The $T_{\text{max}}$ shifts down ($T_{\text{max}} = 75$ K, 70, 64 K for H = 7 T, 4 T and 2 T respectively) with decreasing H. The FC(7T)-FW(0T) curve of PrSrCa exhibits a broad maximum at a higher temperature $T_{\text{max}} = 111$ K and we do not find a clear shift of $T_{\text{max}}$ with H. In contrast to LaCa, the FC(2 T)-FW(0 T) curve does not exhibit a maximum. Figures 4(c) and 4(d) compare the ρ(T) behavior respectively for PrSrCa and LaCa in the FC(7 T)-FW(H) mode. In the FC(7 T)-FW(H) mode, the sample was cooled each time from 300 K to 5K at a constant field of H = 7 T, the field was reduced to a H value at 5 K and then ρ(T) in the field (H) was measured for different values of H. So for each set of curves presented, the initial state of the sample at 5 K is the same, but a new configuration of domains and/or canted state is reached by reducing the field from 7 T to the prescribed H. The resistivity values at 5 K for different H values remain nearly the same for LaCa but not for PrSrCa which shows a factor of 100 increase between the FC(7 T)-FW(7 T) and the FC(7 T)-FW(1 T) data. The FC(7 T)-FW(H) curve for each H value, except for the FC(7T)-FW(7T) curve in LaCa, shows a maximum at $T_{\text{max}}$ which shifts down continuously with decreasing H for LaCa but there is a tendency to shift up below 5 T in PrSrCa. In addition, ρ(T) suddenly jumps from a low to high value at the temperature $T_J << T_{\text{max}}$. The jump is sharper in LaCa, than in PrSrCa, (for example see the FW(1T) curve). We suggest that the low resistivity below $T_J$ (= 12 K in LaCa and 60 K in PrSrCa for FW(1T)) is caused by the 'memory' of the low resistive state at FC (7 T). The downward shift of $T_{\text{max}}$
in LaCa can be understood as follows: The ferromagnetic phase fraction decreases with decrease in \( H \) and hence percolation occurs at a lower temperature. However, the dominant effect of reducing \( H \) from 7 T in PrCa is the decrease in the canting angle. The peak in the FC(7 T)-FC(H) occurs when the loss of the Zeeman energy due to increase in canting angles is compensated by an increase in thermal energy. Presently no standard formula is available to fit these data to extract a quantitative information. Nevertheless, these data indicate that the origins of magnetoresistance in these two compounds are different.

V. SUMMARY

In summary, we have shown that magnetotransport in two magnetically identical compounds \( \text{La}_{0.5}\text{Ca}_{0.5}\text{MnO}_3 \) (LaCa) and \( \text{Pr}_{0.5}\text{Sr}_{0.41}\text{Ca}_{0.09}\text{MnO}_3 \) (PrSrCa) are strongly path dependent. Field cooling is more effective in lowering the resistivity than zero field cooling. The isofield temperature scan data are qualitatively similar in both compounds. However, from the isofield temperature scan alone it is impossible to understand the origin of the magnetic history dependent irreversibility in resistivity. From the isothermal magnetic measurements, we have shown that the dominant effect of field cooling is to increase the ferromagnetic phase fraction in LaCa whereas it creates metastable canted antiferromagnetic domains and/or ferromagnetic clusters in PrSrCa. If LaCa is cooled under the maximum field of 7 T, and \( H \) is reduced to a different value at 5 K, the resistivity value is nearly the same for different values of \( H \) at 5 K whereas it increases by two orders of magnitude in PrSrCa. The resistivity of LaCa when warmed in a field after field cooling in \( \mu_0H = 7 \) T, shows a monotonic downward shift of the position of the resistivity peak but PrSrCa shows a different behavior. These two compounds also exhibit different resistivity behaviors if samples are heated in zero field after field cooling in different \( H \). A quantitative understanding of these results are still lacking. In view of these results, it will be interesting to study the irreversibility behavior of the resistivity in other half doped manganites like \( \text{Pr}_{0.5}\text{Sr}_{0.5}\text{MnO}_3 \) and \( \text{Nd}_{0.5}\text{Sr}_{0.5}\text{MnO}_3 \). The future work should also address whether the irreversibility in the resistivity can be observed...
in a non phase separated manganite.

VI. ACKNOWLEDGMENTS

R. M thanks MENRT (France) for financial assistance and thanks Professor M. R. Ibarra for allowing us to use the vibrating sample magnetometer for the high field magnetic measurement on one of the samples.
Figure captions

**FIG. 1** Temperature dependence of resistivity at $H = 0$ T for (a) Pr$_{0.5}$Sr$_{0.41}$Ca$_{0.09}$MnO$_3$ (PrSrCa) and (b) La$_{0.5}$Ca$_{0.5}$MnO$_3$ (LaCa). Insets show the real part of the ac susceptibility. $\rho(T)$ in zero field cooled (ZFC) and field cooled (FC) modes while warming for (c) PrSrCa (d) LaCa.

**FIG. 2** $\rho(H)$ isotherms at 5 K in the ZFC and FC modes for different cooling field in (a) PrSrCa (b) LaCa. M(H) isotherms at 5 K in the ZFC and FC modes in (a) PrSrCa, (b) LaCa.

**FIG. 3** M(H) isotherms for (a) PrSrCa (b) LaCa. The curves were recorded after cooling the sample from 300 K to a prescribed temperature in zero field.

**FIG. 4** $\rho(T)$ measured in zero field while warming after cooling in a field (H) for (a) PrSrCa and (b) LaCa. $\rho(T)$ measured in a field H while warming after cooling in 7 T field for (c) PrSrCa and (d) LaCa.
REFERENCES

1 For reviews see Colossal magnetoresistance, charge ordering and related properties of manganese oxides' Editors C. N. R. Rao and B. Raveau World Scientific Singapore (1998); Mixed valence manganites J. M. D. Coey, M. Viret and S. von Molnàr, Adv. Physics 48, 167 (1999).

2 C. Zener, Phys. Rev. 82, 403 (1951).

3 L. M. Rodriguez-Martinez and J. P. Attfield, Phys. Rev. B 54, R15622 (1996).

4 A. J. Millis, Nature (London), 392, 147 (1998).

5 A. Asamitsu, Y. Moritomo, Y. Tomioka, T. Arima, and Y. Tokura, Nature (London), 373, 407 (1995); J. M. De Teresa, M. R. Ibarra, P. A. Algarabel, C. Ritter, C. Marquina, J. Blasco, J. Garcia, and A. del Moral, Nature 386, 256 (1997); Y. Tokura and Y. Tomioka, J. Magn. Magn. Mater. 200, 1 (1999); C. Ritter, R. Mahendiran, M. R. Ibarra, L. Morellon, A. Maignan, B. Raveau, and C. N. R. Rao, Phys. Rev. B 61. R9229 (2000).

6 E. Dagotto, T. Hotta, and A. Moreo, Phys. Rep. 344, 1 (2001).

7 Gang Xiao, G. Q. Gong, C. L. Canedy, E. J. McNiff. Jr., and A. Gupta, J. Appl. Phys. 81, 5324 (1997).

8 H. Kuwahara, Y. Moritomo, Y. Tomioka, A. Asamitsu, M. Kasai, R. Kumai, and Y. Tokura, Phy. Rev. B 56, 9386 (1997).

9 M. Hervieu, R. Mahendiran, A. Maignan, C. Martin, and B. Raveau, Appl. Phys. Lett. 77, 3254 (2000).

10 Q. Huang, J. W. Lynn, R. W. Erwin, A. Santoro, D. C. Dender, V. N. Smolyaninova, K. Ghosh, and R. L. Greene, Phys. Rev. B. 61, 8895 (2000).

11 R. Mahendiran, A. Maignan, C. Martin, M. Hervieu, and B. Raveau (unpublished).
12 F. Damay, C. Martin, A. Maignan, M. Hervieu, B. Raveau, Z. Jirák, G. André, and F. Bourée, Chem. Mater. 11, 536 (1999).

13 M. Roy, J. F. Mitchell, A. P. Ramirez, and P. Schiffer, J. Phys.: Condens. Matter 11, 4843 (1999); M. Roy, P. Schiffer, J. F. Mitchell, and A. P. Ramirez, Phys. Rev. B 58, 5185 (1998).

14 J. B. Goodenough, Phys. Rev. 82, 100, 564 (1955)

15 D. Niebieskikwiat, R. D. Sánchez, A. Caneiro, and B. Alascio, Phys. Rev. B 63, 212402.

16 R. Mahendiran, M. R. Ibarra, A. Maignan, C. Martin, B. Raveau, and A. Hernando, Solid State Commun. 111, 525 (1999).

17 S. Mori, C. H. Chen and S. -W. Cheong, Rev. Lett. 81, 3972 (1998); G. Allodi, R. De Renzi, F. Licci, and M. W. Pieper, Phys. Rev. Lett. 81, 4736 (1998); P. Levy, F. Parisi, G. Polla, D. Vega, G. Leyva, H. Lanza, R. S. Freitas, and L. Ghivelder, Phys. Rev. B. 62, 6437 (2000); F. Parisi, P. Levy, L.Ghivelder, G. Polla, and D. Vega, Phys. Rev. B 63, 144419 (2001).

18 F. Rivadulla et al., cond-mat/0105088.
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