Current-assisted annealing effect in $Nd_{0.5}(Ca,Sr)_{0.5}MnO_3$

C. W. Chang,[a] A. K. Debnath,[b] and J. G. Lin[c]
Center for Condensed Matter Science, National Taiwan University, Taipei, Taiwan 10764, Taiwan, Republic of China

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Current–voltage ($I-V$) relations of $Nd_{0.5}(Ca,Sr)_{0.5}MnO_3$ are investigated near its percolation threshold. Resistivity and $I-V$ relations reveal the unstable nature of $Nd_{0.5}(Ca,Sr)_{0.5}MnO_3$. A large slope change in the $I-V$ curve is observable for low ($I<15$ mA) applied current by manipulating the applied magnetic field at 85 K. Our results indicate that the external applied current can assist not only an insulator to metal transition but also a metal to insulator transition. © *2002 American Institute of Physics.* [DOI: 10.1063/1.1433939]

I. INTRODUCTION

Colossal magnetoresistance manganites with general formula $Re_{1-x}A_xMnO_3$ (Re=rare-earth elements and A=Ca, Sr, and Ba) exhibit complicated competition among ferromagnetism, antiferromagnetism, charge/orbital ordering for $x=0.5$.[1] By changing the average ionic size of A, the strength of various interactions can be manipulated, resulting in the changes in ferromagnetic transition temperature (T_C), charge ordering temperature (T_{co}), and metal–insulator transition temperature (T_m).[2,3] Consequently, some phenomena occur such as strong thermal hysteresis of resistivity and thermopower,[3] anomalous A-size dependence of low-field magnetoresistance,[4] and a large shift of T_{co} with A size or under high pressure.[3,5] It has been shown that the system with Re=Nd exhibits unusual A-size effects compared with other systems like Re=La or Pr.[1] These unusual features for Re=Nd can be summarized as follows.

(1) $Nd_{0.5}(Ca,Sr)_{0.5}MnO_3$ system does not show a charge ordering state for small A size, instead, the ferromagnetic metallic state persists at low temperature.[1,2]
(2) High pressure studies on $(Nd,Sm)_{0.5}Sr_{0.5}MnO_3$ and $Nd_{0.5}Sr_{0.5}MnO_3$ reveal that pressure dependence of T_{co} is opposite to that of other systems like $Pr_{0.5}Ca_{0.5}MnO_3$.[5–7]
(3) Instead of the coexistence of two phases, three different magnetic phases with different orbital ordering has been observed in $Nd_{0.5}Sr_{0.5}MnO_3$.[8]
(4) A large positive magnetoresistance at low temperature has been reported for $Nd_{0.5}Ca_{0.5}MnO_3$ with slightly different synthesis conditions from ours.[9]
(5) Large thermal instability is observed in $Nd_{0.5}(Sr,Ca)_{0.5}MnO_3$ for cyclic runs between low temperature and room temperature by adjusting the A size to the percolation threshold.[10]

Guha *et al.*[11,12] investigated the current–voltage ($I-V$) relation of $Nd_{0.5}Ca_{0.5}MnO_3$ and $Pr_{0.63}Ca_{0.037}MnO_3$ and found that both compounds exhibit nonlinear $I-V$ relation below T_{co} which suggests a current-induced depinning effect.[13] Furthermore, the magnetic field enhanced $I-V$ hysteresis was interpreted as the coexistence of different magnetic phases. It was also found that the current-induced destabilization of the charge ordering state leads to the formation of ferromagnetic domains.

Our previous observation of a jump in the $I-V$ relation for $Pr_{0.5}Sr_{0.3}Ca_{0.2}MnO_3$ below 100 K indicates a new current-induced charge ordering breakdown (COB) effect.[14] Based on the enhanced low-field magnetoresistance, this particular jump was interpreted as the existence of an intermediate state between the charge ordering state and the COB state. In this article, we present current-assisted annealing effect of $Nd_{0.5}(Sr,Ca)_{0.5}MnO_3$.

II. EXPERIMENTAL DETAILS

Samples of $Nd_{0.5}Ca_{0.5-x}Sr_xMnO_3$ were synthesized by the standard solid-state reaction method using high purity powders of Nd_2O_3, $SrCO_3$, $CaCO_3$ and $MnCO_3$. Nd_2O_3 powder was first dried at 900 °C for 1 h prior to the preparation of the stoichiometric mixture of the constituents. Thoroughly mixed powders were calcined at ambient pressure at 1200 °C for 16 h with a heating rate of 5 °C/min. Material was cooled to 150 °C with same rate as used for the heating schedule. Thoroughly grinded calcined material was pressed into pellets and sintered at 1400 °C for 16 h with the same heating/cooling rate as used for calcination. X-ray diffraction (XRD) data had confirmed that all compounds were single phase. Sintered pellets were cut into rectangular shapes with typical dimension of ($8.0\times2.0\times1.0$ mm^3) by a diamond wheel. Four contact pads of indium were soldered on to the sample and resistivity of the samples were measured using a helium cryostat (average cooling rate ~-2.2 K/min) by the standard dc four-probe technique with a computer controlled program. Low temperature $I-V$ measurements were taken cooling the samples by liquid nitrogen passing through a setup attached with an alternating gradient magnetometer (Applied Magnetics Laboratory Inc. Baltimore, MD 21209, USA) and the data were taken with a computer controlled program by

[a]New address: Department of Physics, University of California at Berkeley, Berkeley, California 94720.
[b]Permanent address: Technical Physics and Prototype Engineering Division, Bhabha Atomic Research Centre, Trombay, Mumbai-400 085, India.
[c]Author to whom all correspondence should be addressed at: Center for Condensed Matter Science, National Taiwan University No. 1, Sec. 4, Rd. Roosevelt, Taipei 106, Taiwan; electronic mail: jglin@ccms.ntu.edu.tw

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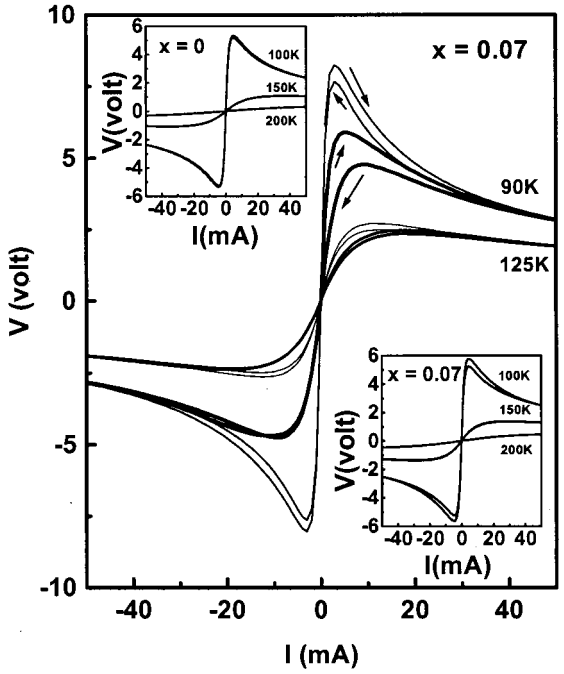

FIG. 1. Insets are the $I-V$ relations for $x=0$ (left-hand side top corner) and 0.07 (right-hand side bottom corner). Compared are the $I-V$ relation for $x=0.07$ under zero field (thin lines) and 1.5 T (bold lines).

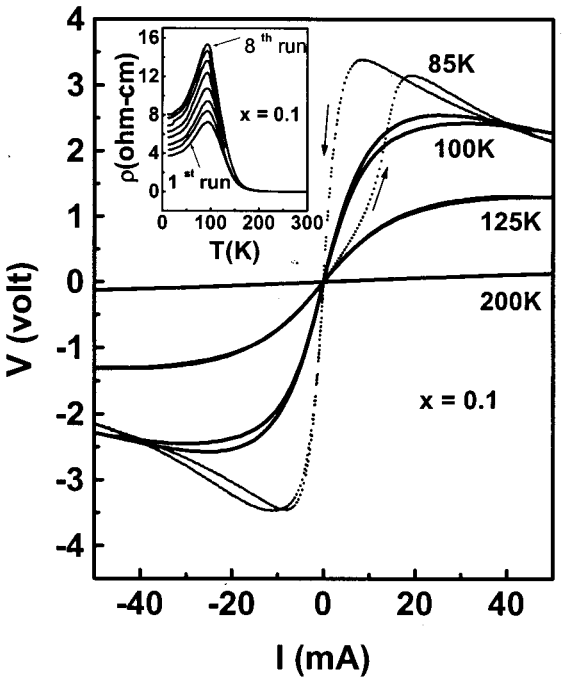

FIG. 2. $I-V$ relations for $x=0.1$ at different temperatures. The $I-V$ relation for $T=85$ K (dot line) shows pronounced difference from those above $T_m \sim 110$ K. Inset shows the resistivity change (warming curve) for successive eight cyclic runs between 295 and 10 K. T_m (slightly shifts for cyclic runs) is determined as peak position of the resistivity warming curve shown in the inset.

slowly sweeping the dc current and simultaneously measuring the voltage across the sample with temperature variation less than 0.1 K. In $I-V$ measurements, the current step was 200 μA and we obtained 499 voltage values for $I=0$ to 50 mA and back to $I=0$ (Fig. 3 and 4). To calculate the (effective) resistance, we divided each voltage value by the corresponding current value. To eliminate the memory effect, the sample was always heated to room temperature before next $I-V$ measurement.

III. RESULTS AND DISCUSSION

The insets of Fig. 1 display the temperature dependence of the $I-V$ relation at zero field for $x=0$ and 0.07. At $T<150$ K, the $I-V$ relation shows apparent nonlinear behavior accompanied by hysteresis for both $x=0$ and 0.07. For $x=0$, the charge order state appears at 250 K.[1] For $x=0.07$, although the magnetization data indicates a small fraction of ferromagnetic domains,[10] the resistivity data shows insulating behavior below room temperature which suggests that the charge ordering state still dominates the low temperature behavior and, as a result, the $I-V$ relations are similar to that of $x=0$. Figure 1 displays the $I-V$ relation for $x=0.07$ under 1.5 T. The magnetic field of 1.5 T has a negligible effect on the $I-V$ relations of $x=0$ (not shown), however, for $x=0.07$, the magnetic field not only shifts the position of the $I-V$ curves but also enhances the $I-V$ hysteresis. Considering the percolation concentration at $x=0.08$,[10] it is natural to attribute the magnetic field effect on $x=0.07$ to a field-induced percolative transition, i.e., more ferromagnetic/metallic domains are connected under magnetic field. Our results are also consistent with Guha *et al.* where phase segregation has been correlated with the enhanced $I-V$ hysteresis.[11]

The inset of Fig. 2 shows the resistivity plots of $x=0.1$ for eight successive cyclic runs measured by a helium cryostat (only the warming curves are shown for clarity). For $x=0.1$, the Sr concentration exceeds the percolation threshold of $x=0.08$, and it shows ferromagnetic metallic behavior at low temperature with $T_m \sim 110$ K. The increase of resistivity with cyclic runs observed in $Nd_{0.5}(Sr,Ca)_{0.5}MnO_3$ was attributed to the thermal instability. Details of the thermal instability were discussed elsewhere.[10] In Fig. 2, the unusual behavior of the Re=Nd system is more pronounced in the $I-V$ relation below T_m. At 85 K, initially the sample is in the metallic state, as reflected in the $I-V$ slope which is smaller than that of $T=100$ K. While decreasing the current (in backward scan), the $I-V$ hysteresis behaves differently from those of $T>100$ K. Further sweeping of the current to negative direction does not show a corresponding dramatic slope change, which suggests that the current-assisted transition is irreversible in character. Actually, after a complete scan, we have found that resistivity of the sample increases by 13 times of its initial value.

Figure 3 displays the resistivity, voltage, and differential resistance versus the applied current at 85 K. In this case, the sample was zero-field cooled (ZFC) to 85 K and before performing the $I-V$ experiment at 1.5 T applied field sample was kept at that temperature for half an hour to reach an equilibrium. It took about a half an hour for a complete $I-V$

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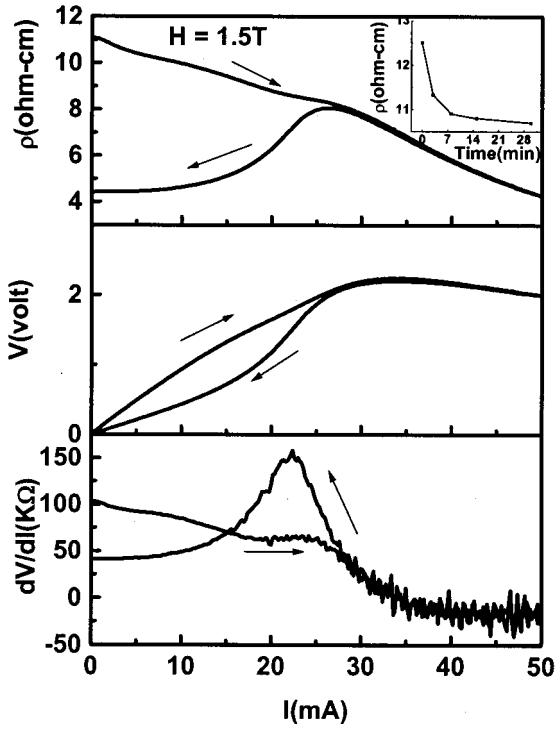

FIG. 3. Resistivity (upper panel), voltage (middle panel), and differential resistance (bottom panel) vs applied current for $x=0.1$ at 85 K under 1.5 T. Sample was ZFC to 85 K before performing the $I-V$ experiment under 1.5 T. Inset displays the relaxation effect after applying the magnetic field.

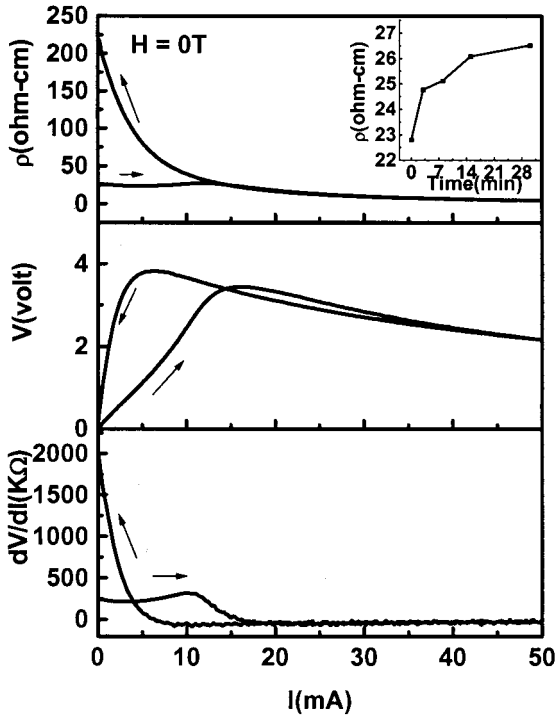

FIG. 4. Resistivity (upper panel), voltage (middle panel), and differential resistance (bottom panel) vs applied current for $x=0.1$ at 85 K under zero field. Sample was FC to 85 K under 1.5 T before performing the $I-V$ experiment. Inset displays the relaxation effect after switching off the magnetic field.

scan from $I=0$ to 50 mA and back to $I=0$. Comparing the upper panel to the inset of Fig. 3, it is seen that the change of resistivity for a complete scan is much higher than that obtained for half an hour to reach at equilibrium. From the middle and the bottom panels of Fig. 3, it is observed that the slope changes in the backward scan (decreasing I), which implies the sample undergoes a current-assisted insulator metal transition at 0–20 mA applied current range. Overall, the sample resistivity reduces to about 1/2.5 of its initial value after a complete scan.

In order to investigate the $I-V$ relation below the transition temperature and to eliminate the effect of the thermal instability, the following procedure has been carried out:

(i) the sample was field cooled (FC) to 85 K under 1.5 T to assure the reappearance of transition at ~110 K,

(ii) applied magnetic field was then switched off at 85 K, and the sample was kept at same temperature for half an hour and,

(iii) during that half hour, the resistivity increment resulting from the relaxation effect is recorded (inset of Fig. 4)

(iv) before performing the $I-V$ measurement.

In spite of the reappearance of T_m at 110 K under 1.5 T, the initial value of sample resistivity (see Fig. 4) is found higher than those of Figs. 2 and 3. However, the main features of Fig. 2 are also found in Fig. 4. The final values of the sample resistivity became eight times higher than its initial value as displayed in the upper panel of Fig. 4. Comparing the upper panel of Fig. 4 with the inset, the increment of resistivity can be attributed to the current-assisted annealing effect since the contribution from the relaxation effect is found negligible. In the middle and bottom panels of Fig. 4, it is clearly seen that the $I-V$ slope changes in a forward scan (increasing I), which indicates a metal to insulator transition. The irrelevance of the Joule heating can be clearly seen in the low current range (0–5 mA). Here, for a given current in the backward scan, resistivity is found higher than that of the forward scan, which is opposite to the Joule heating effect. It is therefore reasonable to assume that the Joule heating is irrelevant to our transport results, qualitatively at least. For $I>20$ mA, the sample resistance decreases similar to the common current-induced COB. While reducing the applied current, the sample goes from a COB state to a CO state as indicated by the final high resistance value. These dramatic behaviors of the sample resistivity with the applied current not only reflects the unstable nature of $x=0.1$, but also indicates the current-assisted annealing effect. Similarly, the ''magnetic annealing'' effect has been observed in other systems like $Pr_{0.5}Sr_{0.5}MnO_3$ and $La_{0.5}Ca_{0.5}MnO_3$.[15,16] In both cases, the present current-assisted metal to insulator transition and the reported magnetic annealing effect, the resistance increases after a complete scan of current/magnetic field, suggesting that a more perfect charge ordering state is energetically favorable.

Considering evidences of the coexistence of ferromagnetic(metallic) and antiferromagnetic(insulating) domains,[8,11]

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we suggest the overall results in Figs. 3 and 4 to be explained by a relative change of the free energy of different magnetic phases. As demonstrated in Fig. 3, initially the sample is in the insulating (higher sensitivity) state, after applying magnetic field, the free energy of the ferromagnetic phases is lowered; therefore, the applied current easily assists the growth of ferromagnetic/metallic phases. On the contrary, in Fig. 4, the initial state is a metallic one, switching off the field lifts the free energy of the ferromagnetic (metallic) phases, as a result, current assists in the depletion of the metallic population. It is worth noting that despite the appearance of the low resistance COB state at high current ($I > 20$ mA), the current-assisted metal to insulator transistor persists after a complete scan. This means that the free energy of the insulating state is energetically more favorable than the ferromagnetic (metallic) state under zero field. This energetically favored insulating state is the origin of the thermal instability observed in $Nd_{0.5}(Ca,Sr)_{0.5}MnO_3$ as shown in the inset of Fig. 2.

IV. CONCLUSION

In summary, we have investigated the $I-V$ relations of $Nd_{0.5}Ca_{0.5-x}Sr_xMnO_3$. By adjusting x near the percolation threshold, the magnetic field enhanced $I-V$ hysteresis indicates the coexistence of different magnetic domains. A current-assisted transition is observed below the metal-insulator transition of 110 K. Our results demonstrate that the applied current can assist not only an insulating to metal transition but also a metal to insulator transition. This particular effect results from the strong competition among different magnetic phases present in $Nd_{0.5}Ca_{0.5-x}Sr_xMnO_3$.

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