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Negative differential resistance in $La_{0.67}Ca_{0.33}MnO_{3-\delta}$/Nb–$SrTiO_3$ *p-n* junction

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$La_{0.67}Ca_{0.33}MnO_{3-\delta}$/Nb–$SrTiO_3$ *p-n* junction was fabricated by pulsed laser deposition. The *I-V* curves of this junction show rectifying behavior. Negative differential resistance (NDR) was observed at low temperatures under large bias voltages and NDR becomes more remarkable with decreasing temperature. In addition to NDR, the *I-V* curves also show remarkable hysteresis. The results were explained in terms of the effect of local Joule heating on the phase separation in the strained ultrathin $La_{0.67}Ca_{0.33}MnO_{3-\delta}$ thin film. © *2007 American Institute of Physics*.
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Colossal magnetoresistive perovskite manganites have attracted much attention because of their rich physics and potential applications.[1] It has been shown that phase separation together with concomitant percolation conductivity play an important role in manganites.[2–5] Phase separation can be in the nanometer scale or micrometer scale. Recently, there are some reports that in the phase separated manganites, local Joule heating due to the electric current can affect the phase separation in $(La_{0.3}Pr_{0.7})_{0.7}Ca_{0.3}MnO_3$ single crystal and size confined $Pr_{0.65}(Ca_{0.75}Sr_{0.25})_{0.35}MnO_3$ thin film, leading to the negative differential resistance (NDR).[6,7] By using the magneto-optical imaging technique, it was observed that at low temperatures large current results in the collapse of the phase separation through a local Joule heating. In $Pr_{0.65}(Ca_{0.75}Sr_{0.25})_{0.35}MnO_3$ system, NDR was only observed in the patterned size confined film, while the bulk and the unpatterned films did not show NDR. This indicates that NDR only appears with some conditions in the phase separated manganite thin films. It should be pointed out that NDR is a well-known concept in device physics, and it has been found in many physical systems with various mechanisms, such as the electron trapping, Gunn effect, electrochemical redox reaction, electric field induced structure change, sequential tunneling in superlattices, etc.[8–12] So it is interesting to see whether the local Joule heating or other mechanisms can lead to NDR in other phase separated manganites.

For ultrathin $La_{0.67}Ca_{0.33}MnO_3$ (LCMO) thin films, it has been shown that the strain, induced by the lattice mismatch between the film and the substrate, can result in a charge ordered insulating (COI) state.[13] In fact, the strain tends to make the film become COI; however, the release or partial release of the strain in some regions make ferromagnetic metallic (FMM) state recover.[13] Because the strain is not homogeneous in the films, COI state exists in the regions with large strain, while FMM state exists in the regions with small or no strain. Therefore, below a certain temperature phase separation occurs with the coexistence of FMM state, and the COI state in the ultrathin LCMO and the FMM domains are separated by the COI domains.[13] Ultrathin LCMO is an interesting system to see whether NDR exists. The high resistance of the ultrathin LCMO is not favorable for the study of the electric current effect, so LCMO was grown on the conducting Nb doped $SrTiO_3$ (NSTO) substrate. In this case, we can measure the current-voltage curves perpendicular to the surface of the ultrathin LCMO film. NDR was observed in LCMO/NSTO heterostructure.

LCMO/NSTO heterostructure was fabricated by growing $La_{0.67}Ca_{0.33}MnO_{3-\delta}$ film on 0.1 wt % NSTO substrate. For comparison, LCMO film was also deposited on $SrTiO_3$ substrate at the same time. The substrate temperature was kept at 650 °C and O_2 pressure was 40 Pa during the deposition. The samples were cooled in a 40 Pa oxygen environment to control the oxygen content. The thickness of LCMO is about 15 nm, and the strain effect is expected to be strong for this thickness. The temperature dependence of magnetization was measured for LCMO/NSTO under a 100 Oe magnetic field by using Quantum Design magnetic property measurement system. The *I-V* curves of LCMO/NSTO heterostructure were measured at different temperatures by using a Keithley 2400 source meter with the two-point method, and the sketch is shown in the inset of Fig. 1. An increasing pulsed dc voltage with a width of 0.5 s and an interval of 3 s was applied to measure the *I-V* curves. Au was used for the electrodes, and they were annealed to make Ohmic contact.

Figure 1 shows the *I-V* curves of LCMO/NSTO heterostructure at different temperatures. It can be seen that the heterostructure shows good rectifying property which can be attributed to the *p-n* junction nature of the heterostructure.[14] But *p-n* junction is not the topic of this letter. Instead we focus on the NDR shown in LCMO/NSTO heterostructure with forward bias voltage exceeding the threshold value. For large forward bias voltages, the depletion layer of a *p-n* junc-

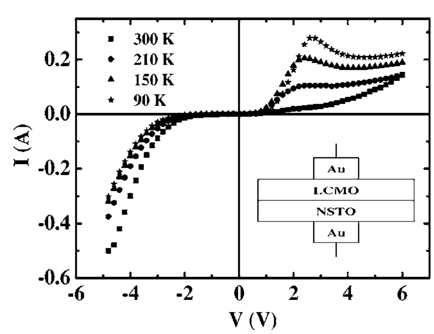

FIG. 1. *I-V* curves measured at different temperatures. The inset is the sketch of the two-point method.

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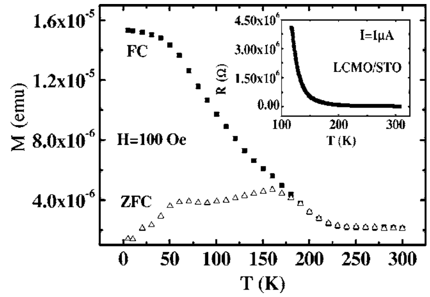

FIG. 2. Temperature dependence of magnetization measured under a 100 Oe magnetic field. The inset is the temperature dependence of resistance of LCMO thin film grown on $SrTiO_3$ substrate.

tion vanishes, so the *p-n* junction does not contribute to the resistance of the heterostructure. It can be seen from Fig. 1 that the forward current of LCMO/NSTO heterostructure increases dramatically with decreasing temperature for large bias voltages. This behavior was described in our previous paper.[14] NDR can be observed for the large forward bias voltages when the temperature is below 210 K and becomes more remarkable at low temperatures. The occurrence of NDR can be understood by considering the phase separation and the resultant filamentary electronic transport in ultrathin LCMO thin film. At low temperatures, phase separation takes place in LCMO film with the coexistence of the COI and FMM phases. Figure 2 shows the temperature dependence of the magnetization for LCMO/NSTO with zero-field cooled (ZFC) and field cooled (FC) measured at 100 Oe magnetic field. The width of the magnetic transition is quite large, and the ZFC and FC curves show large difference, which is consistent with the coexistence of FMM domains and COI domains in the phase separated ultrathin LCMO film. ZFC magnetization nearly vanishes at low temperatures indicating randomly oriented magnetic domains with high barriers for the domain alignment. So at the interface between LCMO and NSTO, there are FMM and COI domains in contact with NSTO. As a result, many micro *p-n* junctions are formed in parallel at the interface of LCMO/NSTO heterostructures with FMM and COI domains as *p* layers, as sketched in the upper inset of Fig. 3. In this inset, the open ovals stand for the FMM domains and the black squares are the COI domains. Since COI regions are insulating, the behavior of the junction is determined by the micro *p-n* junctions of FMM *p* layers. For the in-plane (parallel to the film

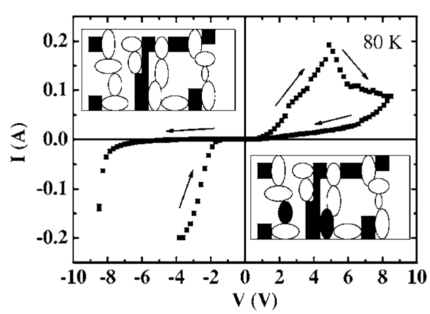

FIG. 3. *I-V* curve measured at $T = 80$ K with the up sweeping and down sweeping measurements. The arrows indicate the measurement direction. The upper-left inset is the sketch of the filamentary transport model. The open ovals stand for the FMM phases and the black squares are the COI phases. In the lower inset, some open ovals change to black ovals (black means insulating) due to the local Joule heating, leading to the decrease of the conducting path number.

surface) electronic transport in LCMO film, no FMM percolation path is formed, i.e., the FMM domains are separated by the COI regions, so the in-plane electronic transport is dominated by the COI regions, and the temperature dependence of resistivity shows the insulating behavior, as shown in the inset of Fig. 2, which is the temperature dependence of the resistance for LCMO film grown on $SrTiO_3$ substrate along the direction parallel to the film surface. But in the vertical direction, some FMM percolation paths may be formed and the current can transport in these filamentary paths. Electric current with moderate value can expand the FMM regions due to the spin polarized current effect,[15] and increases the number of conducting paths. For large forward bias voltages, however, the value of the current is very large, so the heating power determined by *VI* is very large, resulting in strong Joule heating, which can change the FMM domains into nonferromagnetic insulating (NFI) domains because of the abrupt temperature increase since the sample is in the NFI state at high temperatrues.[6] So the resistance of the junction increases dramatically, leading to NDR. The initial state of the interface is very important for the observation of NDR. If the FMM domains are connected to form the transport paths, NDR occurs at higher voltages with the FMM domains changing into NFI domains to form an open circuit. This picture has been shown in the sketch in the lower inset of Fig. 3. In this inset, some open ovals change to black ovals (black means insulating) due to the local Joule heating, leading to the decrease of the conducting path number and the resultant dramatic increase of the junction resistance. This is the key point to understand the NDR phenomenon. It is also expected that Joule heating induced variation of the phase separation at low temperatures may not recover quickly when the current decreases quickly. This should be reflected in the hysteresis of *I-V* curves for the up sweeping and down sweeping measurements.

Figure 3 shows the *I-V* curve of the LCMO/NSTO *p-n* junction measured at 80 K with the up sweeping and down sweeping measurements. Interestingly, NDR does not appear in the down sweeping curve, and the breakdown voltage for the reverse bias increases remarkably. This can be understood based on the above model that some electronic transport chains have been broken to form open circuits due to the local Joule heating, and these broken chains cannot recover quickly, so LCMO/NSTO heterostructure remains in the high resistance state. This also leads to the increase of the breakdown voltage from −2 V (up sweeping) to −7 V (down sweeping).

From the temperature dependence of the junction resistance ($R_{junction}$) with different bias voltages, defined as V/I, one can get some information on the temperature dependence of the different contributions of resistance. The main contributions to $R_{junction}$ are the contact resistances between Au and LCMO and between Au and NSTO, the resistances of LCMO and NSTO, and the resistance of the depletion layer at the interface of LCMO/NSTO. Among them, the contact resistances between Au and LCMO, and between Au and NSTO, and the resistance of NSTO can be neglected compared to $R_{junction}$. So only the resistances of LCMO and the resistance of the depletion layer at the interface of LCMO/NSTO need to be considered. Figure 4 shows the temperature dependence of $R_{junction}$ at different forward bias voltages. $R_{junction}$ increases with decreasing temperature for small bias voltages (inset of Fig. 4), showing the semicon-

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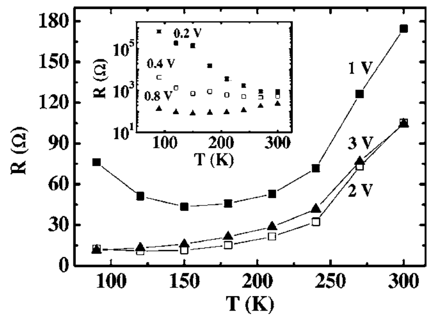

FIG. 4. Temperature dependence of R_{junction} measured at different forward bias voltages. The inset shows the R_{junction}-T curves measured at small bias voltages.

ducting behavior. This reflects the characteristic of the depletion layer of the junction. With increasing bias voltage, R_{junction} decreases because the thickness of the depletion layer of the junction decreases with the forward bias voltage. When the forward voltage exceeds the diffusion voltage V_D (at this voltage, the depletion layer of the junction vanishes) at about 1 V (see Fig. 1), R_{junction} shows a metallic behavior. The small resistance upturn at low temperatures for the 1 V curve is related to the increase of V_D at low temperatures. For $V > V_D$, R_{junction} reflects the electronic transport of LCMO perpendicular to the film, and the percolation of FMM domains determines R_{junction}. Moderate voltage (current) can reduce R_{junction} by the spin polarized current effect.[15] When voltage (current) is large, local Joule heating effect will dominate, leading to the high resistance state and NDR. It is interesting that Tang and Wang reported the NDR in the cluster superlattice of tellurium in zeolite, and they showed that the NDR is related to the electric field induced structure change. This phenomenon is similar in nature with NDR in manganites induced by local Joule heating.

In summary, NDR was observed in LCMO/NSTO heterojunction at low temperatures. Hysteresis was also observed for the *I*-*V* curves of up sweeping and down sweeping. The results can be attributed to the local Joule heating effect on the phase separated ultrathin LCMO. This work demonstrates that local Joule heating can result in some interesting phenomena in the phase separated manganites, which may have potential applications.

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[1] J. M. D. Coey, M. Viret, and S. von Molnar, Adv. Phys. **48**, 167 (1999); *Colossal Magnetoresistive Oxides*, edited by Y. Tokura (Gordon & Breach, London, 1999).
[2] A. Moreo, S. Yunoki, and E. Dagotto, Science **283**, 2034 (1999).
[3] M. Fäth, S. Freisem, A. A. Menovsky, Y. Tomioka, J. Aarts, and J. A. Mydosh, Science **285**, 1540 (1999).
[4] M. Uehara, S. Mori, C. Chen, and S.-W. Cheong, Nature (London) **399**, 560 (1999).
[5] E. Dagotto, Science **309**, 257 (2005).
[6] M. Tokunaga, Y. Tokunaga, and T. Tamegai, Phys. Rev. Lett. **93**, 037203 (2004); M. Tokunaga, H. Song, Y. Tokunaga, and T. Tamegai, *ibid.* **94**, 157203 (2005).
[7] T. Wu and J. F. Mitchell, Appl. Phys. Lett. **86**, 252505 (2005).
[8] K. Kamakura and N. Mutoh, Appl. Phys. Lett. **27**, 214 (1975).
[9] R. G. Cope and A. W. Penn, Br. J. Appl. Phys., J. Phys. D **1**, 161 (1968).
[10] Z. K. Tang and X. R. Wang, Appl. Phys. Lett. **68**, 3449 (1996); J. N. Wang, B. Q. Sun, X. R. Wang, Y. Q. Wang, and W. K. Ge, *ibid.* **75**, 2620 (1999).
[11] X. R. Wang and Q. Niu, Phys. Rev. B **59**, R12755 (1999).
[12] J. Chen, M. A. Red, A. M. Rawlett, and J. M. Tour, Science **286**, 1550 (1999).
[13] A. Biswas, M. Rajeswari, R. C. Srivastava, T. Venkatesan, and R. L. Greece, Phys. Rev. B **63**, 184424 (2001); A. Biswas, M. Rajeswari, R. C. Srivastava, Y. H. Li, T. Venkatesan, and R. L. Greene, *ibid.* **61**, 9665 (2000).
[14] P. L. Lang, Y. G. Zhao, B. Yang, X. L. Zhang, J. Li, P. Wang, and D. N. Zheng, Appl. Phys. Lett. **87**, 053502 (2005); C. M. Xiong, Y. G. Zhao, B. T. Xie, P. L. Lang, and K. J. Jin, *ibid.* **88**, 193507 (2006); Y. S. Xiao, X. P. Zhang, and Y. G. Zhao, *ibid.* **88**, 213501 (2006).
[15] Y. G. Zhao, Y. H. Wang, G. M. Zhang, B. Zhang, X. P. Zhang, C. X. Yang, P. L. Lang, M. H. Zhu, and P. C. Guan, Appl. Phys. Lett. **86**, 122502 (2005).

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