## Current-voltage characteristics of phase separated La<sub>0.5</sub>Ca<sub>0.5</sub>MnO<sub>3</sub>/Nb-SrTiO<sub>3</sub> *p*-*n* junction and magnetic tunability

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The authors report the current-voltage characteristics of  $La_{0.5}Ca_{0.5}MnO_3/Nb-SrTiO_3 p-n$  junction. This junction shows remarkable thermal hysteresis, giant negative magnetoresistance (MR), remarkable differences of MR for the zero field cooling and field cooling processes, and memory effect of magnetic field. Magnetic force microscopy studies provide direct evidence of magnetic inhomogeneity in  $La_{0.5}Ca_{0.5}MnO_3$  film. These intriguing behaviors of our *p-n* junction can be explained by the phase separation in  $La_{0.5}Ca_{0.5}MnO_3$ . This work demonstrates the principle of harnessing phase separation for highly tunable device applications. © 2008 American Institute of Physics. [DOI: 10.1063/1.2944261]

Recently, the perovskite manganite-based p-n junctions have attracted much attention because they exhibit good rectifying characteristics with magnetic tunability,<sup>1</sup> indicating that manganites combined with other kinds of oxides could be a promising way for making devices. Manganites show various peculiar properties, such as the colossal magnetoresistance (MR), charge ordering (CO) and orbital ordering, and phase separation (PS). Among them, PS is one of the key issues in manganites.<sup>2</sup> The PS state can be affected by various factors, accompanied with many interesting characters such as the melting of the CO phase, hysteresis, or memory effect.<sup>2,3</sup> Although there have already been some work on manganite p-n junctions, the work on PS related behavior of *p-n* junctions based on phase separated manganites is still lacking. It is of both scientific and technological interests to study the p-n junctions with phase separated manganites because the PS phenomenon, which can be tuned by various factors, is absent in the traditional semiconductor p-njunctions.

La<sub>0.5</sub>Ca<sub>0.5</sub>MnO<sub>3</sub> (LCMO) is an interesting manganite. PS of charge ordered antiferromagnetic (COAFM) phase and ferromagnetic (FM) phase has been observed in bulk samples by various techniques.<sup>4</sup> In this letter, we study the current-voltage (*I-V*) characteristics of LCMO/Nb-SrTiO<sub>3</sub> p-n junction, which show some interesting behaviors related to the PS.

La<sub>0.5</sub>Ca<sub>0.5</sub>MnO<sub>3</sub>/Nb–SrTiO<sub>3</sub> *p-n* junctions were fabricated by growing 150 nm thick *p*-type LCMO epitaxial thin films on 0.7 wt % Nb-doped *n*-type SrTiO<sub>3</sub> (NSTO) substrates via a pulsed laser deposition technique. The substrate temperature was kept at 750 °C and the oxygen pressure at 40 Pa during deposition. After deposition, the films were cooled to room temperature in an oxygen atmosphere of 0.8 atm. The  $\theta$ -2 $\theta$  x-ray diffraction pattern of LCMO/NSTO shows only the (00*l*) peaks of the LCMO and NSTO. Magnetic force microscopy (MFM) experiments were performed at a homemade variable temperature system interfaced with a Nanoscope IIIa controller (Veeco Digital Instruments). The film was kept in cryogenic vacuum with magnetic field perpendicular to the film plane during experiments. MFM images were taken in the frequency-modulated lift mode, in which the topography and MFM scan lines are interleaved. The lift height was 20 nm.<sup>5</sup> The magnetic and electric properties were measured by using a Quantum Design magnetic property measurement system (MPMS XL7). *I-V* curves of the junctions were measured by using the two-probe method, as shown in the bottom left inset of Fig. 1. To obtain Ohmic contacts, Au and In were used as electrodes for film and NSTO, respectively.

Figure 1 is the *I-V* curves of LCMO/NSTO junction at various temperatures, showing the rectifying property. For the forward bias, the curves shift to higher voltages with decreasing temperature, which was also observed in other manganite based *p-n* junctions.<sup>1</sup> The temperature dependence of the junction resistance  $R_j$ , defined as V/I, is shown in the top inset of Fig. 1. For the forward bias voltages below the diffusion potential  $V_D$  (0.5 V) and the reverse bias voltages,  $R_j$  shows a peak at about 100 K. While for the forward bias voltages above  $V_D$ ,  $R_j$  shows a monotonic increase with decreasing temperature.



FIG. 1. (Color online) *I-V* curves of the junction at various temperatures. The top inset shows the temperature dependence of the junction resistance at various bias voltages. The bottom left inset displays the schematic of the *p-n* junction and the electrodes setting. The bottom right inset shows the current difference  $D_I$  between the cooling and warming measurements at the bias voltage of 0.3 V.

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FIG. 2. (Color online) [(a) and (b)] Variation of ZFC MR with the bias voltage at different temperatures with an in-plane magnetic field of  $\mu_0 H$ =0.2 T. (c) Magnetization vs temperature under an in-plane magnetic field of 0.2 T for the FC and ZFC processes. (d) Temperature dependence of MR under a 0.2 T in-plane magnetic field at the bias voltage of 0.3 V for the FC and ZFC processes, respectively. The inset of (d) is the temperature dependence of the difference between MR<sub>ZFC</sub> and MR<sub>FC</sub>,  $\Delta$ MR=MR<sub>ZFC</sub>-MR<sub>FC</sub>.

It has been reported that near the CO temperature, the electrical resistance of LCMO film shows a thermal hysteresis between the cooling and warming curves,<sup>6</sup> which is due to the first-order phase transition between COAFM phase and FM phase in LCMO. The I-V curves of LCMO/NSTO junction show that currents measured in the warming process  $(I_W)$  are larger than those in the cooling process  $(I_C)$  under the same bias voltage, consistent with the thermal hysteresis of LCMO film resistance.<sup>6</sup> The difference of the currents is defined as  $D_I = (I_W - I_C) / I_C \times 100\%$ . The bottom right inset of Fig. 1 is the temperature dependence of  $D_I$ . It is very small at both high temperatures and very low temperatures, but with decreasing temperature it starts to increase at around 100 K, which is consistent with the peak temperature in the  $R_i$ -T curves, and becomes remarkable near 75 K.

Figures 2(a) and 2(b) display the temperature dependence of MR of the junction in the zero field cooling (ZFC) process, defined as  $MR_{ZFC} = (R_H - R_0) / R_0 \times 100\%$ , where  $R_H$ and  $R_0$  stand for the junction resistance with and without an in-plane magnetic field, respectively. MR<sub>ZFC</sub> reaches a maximum value of about 23% at 75 K, which is consistent with the characteristic temperature at which the thermal hysteresis of the junction shows a maximum. As seen from Figs. 2(a) and 2(b), MR<sub>ZFC</sub> also changes with the bias voltage and shows a peak for both the forward and reverse biases at different temperatures. For the forward bias region, the depletion layer vanishes when the voltage is larger than  $V_D$ , leading to a remarkable decrease of MR because of the domination of the highly tensile-strained LCMO layer near the interface of LCMO/NSTO.<sup>6</sup> For the reverse bias region, MR first increases with the increase of the bias voltage and begins to decrease at a certain bias voltage. This decrease may be related to the gradual breakdown of the junction manifesting as the remarkable increase of the current.

Furthermore, the current increase induced by a 0.2 T magnetic field in the field cooling (FC) process is larger than that in the ZFC process. Figure 2(d) shows the temperature dependence of MR<sub>ZFC</sub> and MR<sub>FC</sub> with a bias voltage of 0.3 V and the inset shows the difference between them, which becomes more and more significant with decreasing temperature. This phenomenon is consistent with the M-T curve of the junction under a 0.2 T magnetic field, as shown



FIG. 3. (Color online) I-V curves of the junction with an in-plane magnetic field  $\mu_0 H=0$ , 5.0, and 0 T, successively. The upper left inset is the magnetic field dependence of resistance for LCMO film grown on SrTiO<sub>3</sub> substrate. The upper right inset is the schematic of the barrier between the COAFM and FM states. The bottom inset shows the schematic of the coexistence of COAFM and FM phases in the junction.

in Fig. 2(c), which shows the separation between the FC and ZFC M-T curves increases with decreasing temperature. Since the FM region is metallic and the COAFM phase is insulating, the transport in this inhomogeneous system is mainly dominated by the spin-dependent tunneling or percolation. Therefore, the magnetization alignment and the volume increment of the FM regions in the FC process induce a much larger MR than in the ZFC process. Thus, the difference between the MR<sub>ZFC</sub> and MR<sub>FC</sub> is more remarkable at low temperatures.

PS systems generally show memory effect of magnetic field. The I-V curves of the junction at 10 K with 0 T, 5.0 T and removal of the 5.0 T in-plane magnetic field (0 T2) were measured successively and the result is shown in Fig. 3. The current increases notably under a 5.0 T magnetic field, showing a giant negative MR. When the magnetic field was removed, the current decreases to a value larger than the previous value at 0 T, demonstrating the magnetic memory effect of the LCMO/NSTO junction. For LCMO thin film, a 5.0 T magnetic field can convert the COAFM phase to FM phase, resulting in a remarkable decrease of resistivity.<sup>9</sup> The magnetic field dependence of resistance at 10 K for LCMO grown on SrTiO<sub>3</sub> substrate is shown in the upper left inset of Fig. 3. It displays a significant MR memory effect which can account for the memory effect of the junction. It has been proposed that COAFM phase is the ground state for the halfdoped LCMO but FM phase can be trapped in the AFM host at low temperatures.<sup>7</sup> Upon applying a 5.0 T magnetic field, the COAFM phase begins to convert to the FM phase, which is aligned by the magnetic field, leading to a much lower resistance. After removing the field, most of these FM domains are frozen and cannot go back to the original COAFM phase because the potential barrier between the FM and COAFM phases (see the upper right inset of Fig. 3) is likely much larger than the thermal fluctuation. This scenario can account for the memory effect in LCMO film or LCMO/ NSTO junction.

Although there have been many evidences of PS in LCMO bulk samples,<sup>4</sup> it is crucial to show that in LCMO thin film. Figure 4(a) shows the MFM image of LCMO taken at 10 K with  $\mu_0 H=1.0$  T ( $\perp$  film) after ZFC. As indicated in the M(H) loop in Fig. 4(d), the FM phase is saturated for the  $\mu_0 H > 0.8$  T, i.e., any FM region is single domain. Thus, the MFM data for  $\mu_0 H > 0.8$  T provide direct evidence of mag-Author complimentary copy. Redistribution subject to AIP license or copyright, see http://apl.aip.org/apl/copyright.jsp



FIG. 4. (Color online) (a)–(c) are MFM and AFM images of the same area of the sample  $(3 \times 3 \ \mu m^2)$  taken during an isothermal magnetic field sweep at 10 K after ZFC. The field values (color scales) of the images in consecutive sequence are (a) 1.0 T (5 Hz) and (b) 8.0 T (5 Hz). The scale of the topographic image (c) is 30 nm. (d) The out-of-plane M(H) curve of the LCMO film at T=10 K.

netic PS in our LCMO film. In 1.0 T field, the magnetizations of both FM regions and the MFM tip are parallel to the external magnetic field, giving only attractive force between them. The darker places, indicating stronger attractive interaction, are regions with larger local magnetization averaged over the film thickness. Regions with intermediate contrast are possibly the places with overlapping of FM phase and nonmagnetic matrix (presumably COAFM phase). Figure 4(b) shows the MFM image at the identical location in 8.0 T field after an isothermal field sweep. FM regions grow gradually upon increasing the magnetic field. This is consistent with the rather gradual reduction of the resistivity shown in the upper left inset of Fig. 3. Upon decreasing the magnetic field to 1.0 after 8.0 T, no visible change could be found in MFM images (not shown for brevity), supporting the aforementioned memory effect. Details of MFM studies will be published elsewhere.<sup>8</sup> No correlation between MFM images and topographic image [Fig. 4(c)] confirms that topographic defects are not responsible for the observed pinning/memory effects.

The characteristics of the LCMO/NSTO junction mentioned above are mainly determined by the properties of the LCMO side of the junction and can be understood in terms of the PS. In this scenario, at the interface between LCMO and NSTO, there are FM and COAFM regions alternately in contact with NSTO. Because the COAFM regions are insulating, the behavior of the junction is determined by the FM regions and many micro p-n junctions in parallel are formed as shown schematically in the right bottom inset of Fig. 3. The proportions of the FM and COAFM regions change with temperature and magnetic field, etc. Since PS can be tuned by various factors, devices based on the PS materials would have a good tunability. Our work demonstrates the rich phenomena in p-n junctions based on the phase separated manganites and will activate the exploration of PS controlled devices and related physics.

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