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Magnetically tunable properties related with carriers density in self-doped $La_{1-x}MnO_3/y$ wt %Nb–$SrTiO_3$ heteroepitaxial junctions

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The self-doped $La_{1-x}MnO_3$ (x=0.1 and 0.3) thin films deposited on Nb-doped (wt % y) $SrTiO_3$ (y=0.05 and 0.8) crystals to form heteroepitaxial junctions have been prepared by the pulse laser deposition method. The current-voltage loops of junction were measured at several fixed magnetic fields for the temperature from 10 to 300 K. We have focused on the effects of doping level and annealing time on the magnetically tunable property of the junction. The results show that these junctions have a typical temperature-dependent rectifying characteristics and asymmetrical hysteresis. The magnetically tunable property of the junction was related with the annealing time for the self-doped $La_{1-x}MnO_{3-\delta}$ thin film and the doping level in the Nb-doped $SrTiO_3$ (STON) crystal. In the self-doped $La_{0.9}MnO_3$/0.05-STON junction annealed at 900 °C for 5 h, the relative ratio of voltage $[V_b(0)-V_b(H)]/V_b(0)$ is about 70% at H=6 T and T=70 K for I=0.1 mA, showing a large magnetically tunable property. These results reveal the great potential of the manganites in configuring artificial devices. © *2010 American Institute of Physics*. [doi:10.1063/1.3358597]

I. INTRODUCTION

The manganite heteroepitaxial junctions have been a focus of intensive studies since the discovery of a good rectifying behavior in this kind of *p*-*n* junction.[1–5] Due to the high sensitivity of electronic and magnetic properties of manganese to external magnetic field and electric field, the diffusion potential may make these simple *p*-*n* junctions promising for various novel applications. Therefore, the magnetically tunable property of *p*-*n* junction is of special interest from the viewpoint of application. It was also known that the forward and reverse current characteristics of diode were improved greatly by the changing in carriers' density of two sides. In these manganite heteroepitaxial junctions, the conducting $La_{1-x}MnO_3$ thin films were used as the *p*-type region, while the electron conducting Nb-doped (wt % y) $SrTiO_3$ (y-STON) substrates as *n*-type region. What will be the effects of the doping level in the STON crystals and the annealing time for the self-doped $La_{1-x}MnO_{3-\delta}$ thin films is an interesting question for us. In this paper, we constructed a bilayer junction using self-doped $La_{1-x}MnO_{3-\delta}$ thin film and y-STON crystal, and focused on the current-voltage loops under various magnetic fields for the temperature from 10 to 300 K. We observed a large magnetically tunable property in the self-doped $La_{0.9}MnO_3$/0.05-STON junction annealed at 900 °C for 5 h.

II. SAMPLES AND EXPERIMENTAL

The epitaxial self-doped $La_{1-x}MnO_{3-\delta}$ (x=0.1 and/or 0.3) thin film (~100 nm) were deposited on heating y-STON (y=0.05 and/or 0.8) single crystal and a $SrTiO_3$ substrate (~720 °C) in 1 mbar pure oxygen gas by pulse laser deposition technique. The $La_{1-x}MnO_{3-\delta}/SrTiO_3$ sample as a reference is used to study the property of $La_{1-x}MnO_{3-\delta}$ layer. The heteroepitaxial junction area is about 5×3 mm^2. The isothermal current-voltage (*I*-*V*) loops from 10 to 300 K with an interval of 10 K in the applied magnetic field up to 9 T were performed on a Quantum Design physical property measurement system (PPMS-9) by a four-probe method shown in the upper inset of Fig. 1(a). In order to get the isothermal *I*-*V* loops at any temperature and any magnetic field, we must keep the measuring current below 0.1 mA in the voltage limit of PPMS. The measuring current increases from −0.1 to 0.1 mA then from 0.1 to −0.1 mA in a period of 11.2 s for 256 points. The magnetic field was applied perpendicular to the surface of thin film.

III. RESULTS AND DISCUSSIONS

Before we explore the rectifying behavior of $La_{1-x}MnO_3$/y-STON junction under the magnetic field, we should know the effect of annealing time on the colossal magnetoresistance (CMR) behavior of $La_{1-x}MnO_3$ thin films. Therefore we have measured the temperature dependence of resistance for these annealed $La_{1-x}MnO_3$ thin films. The results show that the as-grown sample has a semiconducting behavior and a metal-insulator transition observed in annealed sample. The metal-insulator transition temperature depends on the annealing temperature and time. We think 900 °C and 5 h was a good condition to get high metal-insulator transition temperature. In this study, the metal-insulator transition temperature of $La_{1-x}MnO_3$ layer at H=0 occurs at 306.5 K for the $La_{0.7}MnO_{3-\delta}$/0.8-STON junction annealed at 900 °C for 1 h (sample A), at 338 K for the $La_{0.7}MnO_{3-\delta}$/0.05-STON junction annealed at 900 °C for 5

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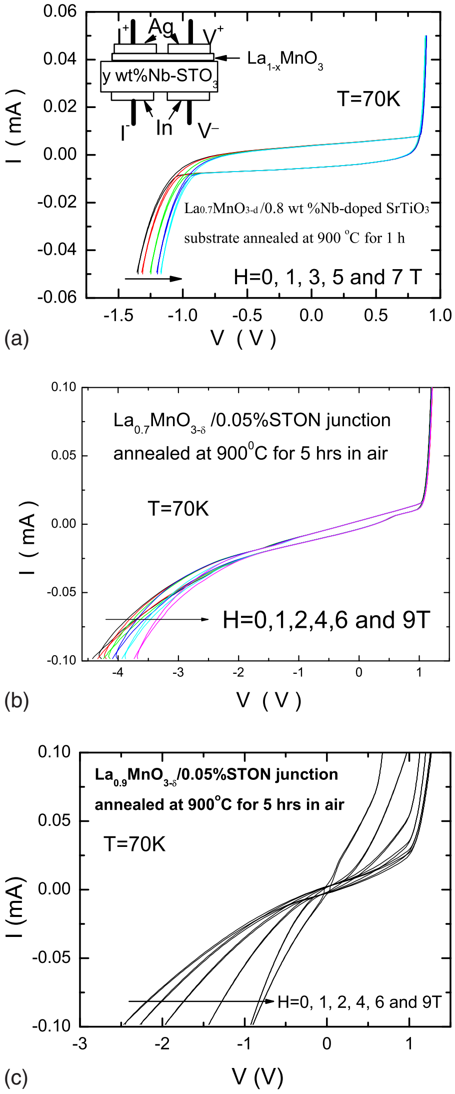

FIG. 1. (Color online) Isothermal current-voltage loops of the heteroepitaxial $La_{1-x}MnO_{3-\delta}$/y-STON junction at $T=70$ K for various applied magnetic fields. (a) H=0, 1, 3, 5, and 7 T for sample A; (b) H=0, 1, 2, 4, 6, and 9 T for sample B; and (c) H=0, 1, 2, 4, 6, and 9 T for sample C; respectively. The contacts configuration was shown in the upper inset of (a).

h (sample B) and at 305 K for the $La_{0.9}MnO_{3-\delta}$/0.05-STON junction annealed at 900 °C for 5 h (sample C), respectively. Due to the limit of paper length, we do not present the CMR behavior of these annealed $La_{1-x}MnO_3$ thin films here.

Our aim in this paper is to explore the magnetically tunable property of these heteroepitaxial $La_{1-x}MnO_3$/y-STON junctions affected by the annealing time for the self-doped $La_{1-x}MnO_{3-\delta}$ thin film and the doping level in the Nb-doped $SrTiO_3$ crystal. Therefore, we have measured the isothermal current-voltage loops for these heteroepitaxial $La_{1-x}MnO_3$/y-STON junctions at various magnetic fields for temperature from 300 to 10 K with an interval of 10 K. All of *I*-*V* loops show good rectifying characteristic, but they do not come from the initial point ($I=0$, $V=0$), showing a large asymmetrical hysteretic loop. For the clarity, we just show typical *I*-*V* curves at 70K for various magnetic fields in Fig. 1. The observed asymmetrically hysteretic *I*-*V* loops can be quantitatively simulated by a resistance capacitance shunted junctionlike model $I=I_0[\exp(bV/T)-1]+CdV/dt+V/R$.[5] From Fig. 1, we can observe that the asymmetrical hysteretic *I*-*V* loops depend really on the annealing time for the self-doped $La_{1-x}MnO_{3-\delta}$ thin film and the doping level in the Nb-doped $SrTiO_3$ crystal. The diffuse voltage V_d and breakdown voltage V_b are different for these samples. $V_d \approx 0.75$ V and $V_b \approx 1.4$ V in sample A, $V_d \approx 1.1$ V and $V_b \approx 4$ V in sample B and $V_d > 1.25$ V and $V_b > 3$ V in sample C indicate that the rectifying characteristic depends on the carriers density in the self-doped $La_{1-x}MnO_{3-\delta}$ thin films and the STON substrates.

From Fig. 1, we can also observe a strong asymmetrical field effect on the rectifying property. For sample A and B, the diffusive voltage V_d is not affected by the applied magnetic field, but the breakdown voltage V_b decreases with the increasing of magnetic field. The breakdown voltage V_b in sample B is higher than 4 V at 70 K at H=0 T. In the sample C, both the diffusive voltage V_d and the breakdown voltage V_b decrease with the increasing of magnetic field. The breakdown and diffusive voltages should be determined from the *I*-*V* curve and respond to increasing of the downward and forward currents rapidly, respectively. Due to the voltage limit of PPMS ($V=5$ V) for *I*-*V* measurement, we cannot get the accurate breakdown voltage V_b of sample C. If we defined the voltage at $I=50$ μA for sample A and at $I=100$ μA for sample B and C, respectively, as the breakdown voltage V_b, we can obtain the field dependence of the breakdown voltage V_b at $T=150$, 100, 50, and 10 K, respectively, as shown in Fig. 2. From Fig. 2(a), we can see that the absolute breakdown voltage V_b decreases linearly with the applied magnetic field, namely, the breakdown voltage V_b is tuned by the applied magnetic field. At a fixed magnetic field, the breakdown voltage V_b increases with decreasing of temperature above 50 K. When the temperature is lower than 50 K, the breakdown voltage V_b is independent of the temperature. For sample B shown in Fig. 2(b), the field dependence of the breakdown voltage V_b is similar to that in sample A, but the absolute value of breakdown voltage V_b is higher than that of sample A. From Fig. 2(b), the temperature dependence of breakdown voltage V_b at a fixed magnetic field for sample B is different that of sample A. The breakdown voltage V_b has a peak near 100 K. The field dependence of breakdown voltage V_b in sample C shown in Fig. 2(c) decreases linearly with the increasing of applied magnetic field for $H \leq 6$ T, then displays a platform for $H > 6$ T. The temperature dependence of breakdown voltage V_b for sample C is similar to that for sample B. There is a peak near 100 K.

In order to show the magnetically tunable properties of these heteroepitaxial $La_{1-x}MnO_3$/y-STON junction clearly, we calculate the relative ratio of breakdown voltage $[V_b(0)-V_b(\mathrm{H})]/V_b(0)$ at given temperatures for various magnetic fields from the data of Fig. 2. The field dependence of relative ratio of breakdown voltage $[V_b(0)-V_b(\mathrm{H})]/V_b(0)^{*}100\%$ is shown in Fig. 3. At a fixed temperature, the relative ratio of voltage $[V_b(0)-V_b(\mathrm{H})]/V_b(0)$ increases linearly with the increasing of applied magnetic field up to 6T for all of the $La_{0.7}MnO_{3-\delta}$/y-STON junctions. The maximum of the relative ratio of voltage $[V_b(0)-V_b(\mathrm{H})]/V_b(0)$ is different. The maximum of the relative ratio of voltage $[V_b(0)-V_b(\mathrm{H})]/V_b(0)$ is about 14% at T=10 K and H=7 T for the

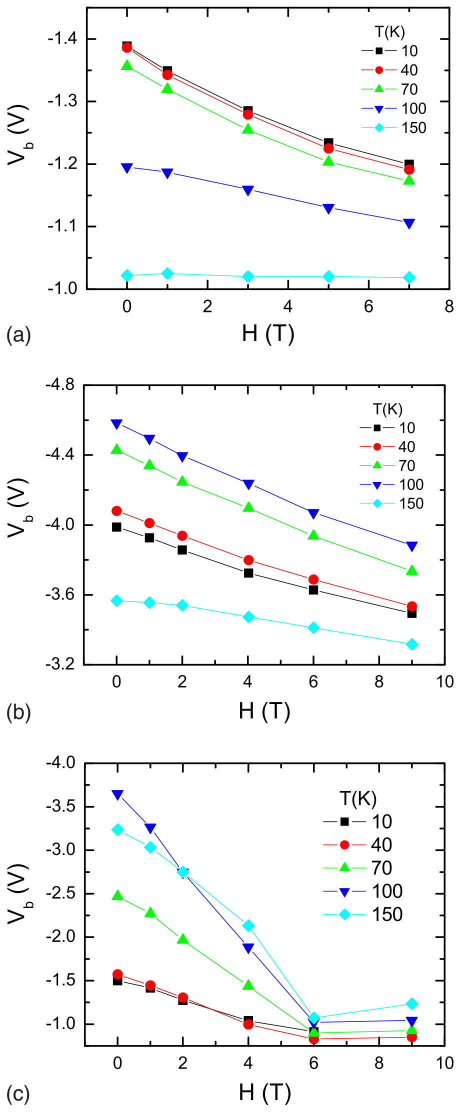

FIG. 2. (Color online) Field dependence of the breakdown voltage V_b at T=150, 100, 70, 40 and 10 K, respectively. (a) Sample A, (b) sample B, and (c) sample C.

sample A and 15% at T=70–100 K and H=9 T for sample B. However, we find a large relative ratio of voltage $[V_b(0) - V_b(\mathrm{H})]/V_b(0)$ in sample C. The maximum of the relative ratio of voltage $[V_b(0)-V_b(\mathrm{H})]/V_b(0)$ is about 70% at T =70~100 K and H=6 T. These results reveal the great potential of the manganites in configuring artificial devices.

IV. CONCLUSIONS

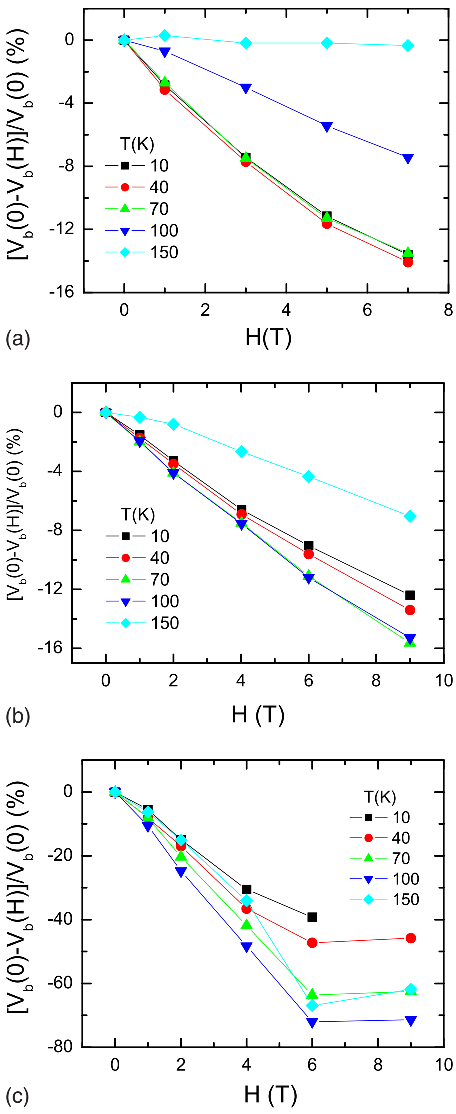

FIG. 3. (Color online) Field dependence of the breakdown voltage V_b ratio $[V_b(0)-V_b(\mathrm{H})]/V_b(0)*100\%$ at T=150, 100, 70, 40 and 10 K, respectively. (a) Sample A, (b) sample B, and (c) sample C.

The bilayer $La_{1-x}MnO_{3-\delta}$/y-STON heteroepitaxial junctions show typical temperature-dependent rectifying characteristics and asymmetrical hysteresis. The asymmetrical *I-V* curve depends on the density of carriers in the self-doped $La_{1-x}MnO_{3-\delta}$ thin films and that in the Nb-doped $SrTiO_3$ substrates. We observed a larger magnetically tunable property in the self-doped $La_{0.9}MnO_3$/0.05-STON junction annealed at 900 °C for 5 h. The relative ratio of voltage $[V_b(0)-V_b(\mathrm{H})]/V_b(0)$ is about 70% at H=6 T and T =70 K for I=0.1 mA.

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