High sensitivity of positive magnetoresistance in low magnetic field in perovskite oxide *p–n* junctions

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Large positive magnetoresistance (MR) and high MR sensitivity in low magnetic fields have been discovered in the Sr-doped $LaMnO_3$ and Nb-doped $SrTiO_3$ *p–n* junctions fabricated by laser molecular-beam epitaxy. The MR ratios, defined as $\Delta R/R_0$, $\Delta R = R_H - R_0$, are observed as large as 11% in 5 Oe, 23% in 100 Oe, and 26% in 1000 Oe at 290 K; 53% in 5 Oe, 80% in 100 Oe, and 94% in 1000 Oe at 255 K. The MR sensitivities are 85 Ω/Oe at 290 K, 246 Ω/Oe at 255 K, and 136 Ω/Oe at 190 K, respectively, with the applied magnetic field changed from 0 to 5 Oe. The positive MR ratios and high MR sensitivities of the *p–n* junctions are very different from that of the $LaMnO_3$ compound family. © *2005 American Institute of Physics*. [DOI: 10.1063/1.1850192]

Since the rediscovery of colossal magnetoresistance (CMR) phenomena in manganite thin films, much attention has been focused on the fabrication of artificially designed structures, such as magnetic tunnel junctions (MTJ),[1–9] and *p–n* junctions,[10–12] to verify device concepts based on oxide materials. The magnetoresistance (MR) ratio is defined as $\Delta R/R_0 = (R_H - R_0)/R_0$, where R_H is the resistance under the applied magnetic field and R_0 is the resistance in zero field. Good results of the negative MR ratio have been reported, such as 150% in $La_{0.8}Sr_{0.2}MnO_3/SrTiO_3/La_{0.8}Sr_{0.2}MnO_3$ MTJ at 5 K under a low switching field (<10 Oe),[5] and 450% in trilayer MTJ at 14 K in 200 Oe,[6] but only a few of perovskite oxide materials with positive MR were found. Ghosh *et al.* observed $\Delta R/R_0 = 25\%$ at 80 K in 4 T in a $Fe_3O_4/SrTiO_3/La_{0.7}Sr_{0.3}MnO_3$ MTJ.[4] Mitra *et al.* demonstrated a large positive MR at 48 K in 2 T in a $La_{0.7}Ca_{0.3}MnO_3/SrTiO_3/La_{0.7}Ce_{0.3}MnO_3$ MTJ.[1] And we also found the modulation effect of current and voltage, as well as the positive MR effect in systems of doped $LaMnO_3$ and Nb-doped $SrTiO_3$ *p–n* junctions and multiplayer *p–n* heterostructures.[13–15] In this Letter, we will report on the fabrication of $La_xSr_{1-x}MnO_3/SrNb_yTi_{1-y}O_3$ (LSMO/SNTO) *p–n* junctions and show the positive MR effect and high MR sensitive of the junctions in low magnetic fields.

In order to fabricate a better oxide *p–n* interface, a computer-controlled laser molecular-beam epitaxy (laser MBE) (Ref. 16) was used to deposit the LSMO/SNTO *p–n* junctions. A series of $La_{1-x}Sr_xMnO_3/SrNb_yTi_{1-y}O_3$ *p–n* junctions with various doping concentrations ($x = 0.1, 0.2; y = 0.01, 0.1$) have been elaborated. The *p–n* junctions have two different recipes: (1) 3500–5000 Å SNTO was deposited on the $SrTiO_3$ (001) substrate, followed by 3500 Å LSMO; (2) 2500–4000 Å LSMO was deposited directly on 1% Nb-doped $SrTiO_3$ (001). The preparation conditions of the junctions are described as follows: the laser (with a wavelength of 308 nm) has a repetition rate of 2 Hz and a duration 20 ns, the energy density was approximately 1 J/cm^2, the substrate temperature was kept at 630 °C, and oxygen pressure of 2×10^{-3} Pa was maintained throughout the deposition. The growth rate was about 12 Å/min for SNTO and about 13 Å/min for LSMO.

An x-ray diffraction (XRD) θ–2θ scan curve of a $La_{0.9}Sr_{0.1}MnO_3/SrNb_{0.01}Ti_{0.99}O_3$ (LS0.1MO/SN0.01TO) *p–n* heterostructure and its cross-sectional high-resolution transmission electron microscopic (HRTEM) image are shown in Fig. 1. There exist only LS0.1MO (00*l*) and SN0.01TO (00*l*) peaks, with no trace of other diffraction peaks from either the impurity phase or the randomly oriented grain. This means that the thin film of heterostructure is in single phase with *c*-axis orientation. The HRTEM image shows that the interface is perfectly oriented and the epitaxial crystalline structure shows the orientation relation of SN0.01TO(001)//LS0.1MO(001), and SN0.01TO[100]// LS0.1MO[100].

The electrical and magnetic properties of the LSMO/SNTO *p–n* junctions are measured in the temperature range from 100 to 300 K. The surface area of the samples is about

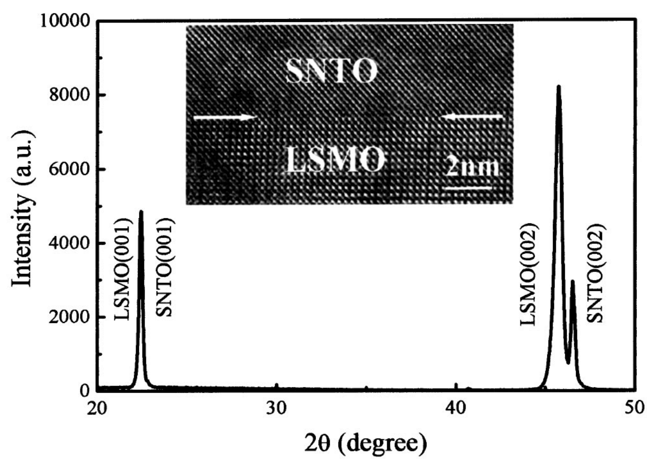

FIG. 1. The XRD θ–2θ scan curve of a LS0.1MO/SN0.01TO *p–n* heterostructure and its cross-sectional HRTEM image of the interface.

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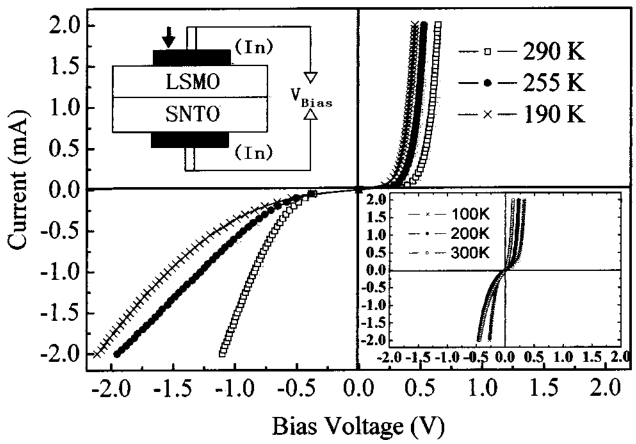

FIG. 2. The $I-V$ curves of a LS0.1MO/SN0.01TO $p-n$ junction at different temperatures. Inset: the schematic illustration of the $p-n$ junction, and the $I-V$ curves of $La_{0.8}Sr_{0.2}MnO_3$/SN0.01TO.

1.5×1.5 mm^2, and the indium (In) electrodes ($\sim$1 mm^2) are placed on the surfaces of the LSMO and SNTO. The magnetic field is applied perpendicularly to the $p-n$ interface and parallel to the current. The $p-n$ junctions can withstand current higher than 100 mA at room temperature. To minimize the heating effect at low temperature, the measurement was carried out with a pulse-modulated current source. The pulse duration was 1 s and the pulse interval was 10 s. The measuring current was kept smaller than 2 mA. We measured ten samples of different structure with two superconductive quantum interference devices (SQUID), (Quantum Design MPMS 5.5T) and observed the similar magnetic properties.

Figure 2 shows the schematic illustration and the $I-V$ curves of a LS0.1MO/SN0.01TO $p-n$ junction at 190, 255, and 290 K. The $I-V$ curves of a $La_{0.8}Sr_{0.2}MnO_3$/SN0.01TO $p-n$ junction are also shown as an inset of Fig. 2. The experimental results show that the $p-n$ junction properties of the low doping are much better than those of the higher doping not only for electrical property, but also for magnetic property. The rectifying property of the LS0.1MO/SN0.01TO $p-n$ junction is the best in the different recipes. The positive MR ratios of $La_{0.8}Sr_{0.2}MnO_3$/SN0.01TO are several times lower than that of the LS0.1MO/SN0.01TO $p-n$ junction. The Fermi level of $LaMnO_3$ locates closely under the edge of the t_{2g} spin-down band. It has been known that the minority spin (t_{2g} spin-down) carriers exist even in the hole-doped compound $La_{0.7}Sr_{0.3}MnO_3$.[17] We speculate that the positive MR phenomena of our $p-n$ heterojunction somehow relate to minority (spin-down) carriers at the t_{2g} band edge of LSMO involved in the current. Comparing the sample LS0.1MO with the sample $La_{0.8}Sr_{0.2}MnO_3$, the one with lower (10%) Sr ($La_{0.9}Sr_{0.1}MnO_3$) has a higher Fermi level, and thus provides more minority carriers at the t_{2g} band edge involved in the current. Therefore, the sample with lower doping has the with larger positive MR. Further study on the mechanism of the spin transport of such a system is under way.

The influences of the magnetic field on the current and voltage of the $p-n$ junction are more obvious to the negative bias than that of the positive bias because the resistance at the negative bias is larger than that at the positive bias. The dependence of the current as a function of negative bias in the LS0.1MO/SN0.01TO $p-n$ junction at 290, 255, and 190 K in an applied magnetic field varying from 0 to 1000 Oe are shown respectively in Figs. 3(a)–3(c). The absolute values of bias voltage increase with an increase in the magnetic field at a fixed current, indicating a positive MR behavior for the $p-n$ junction.

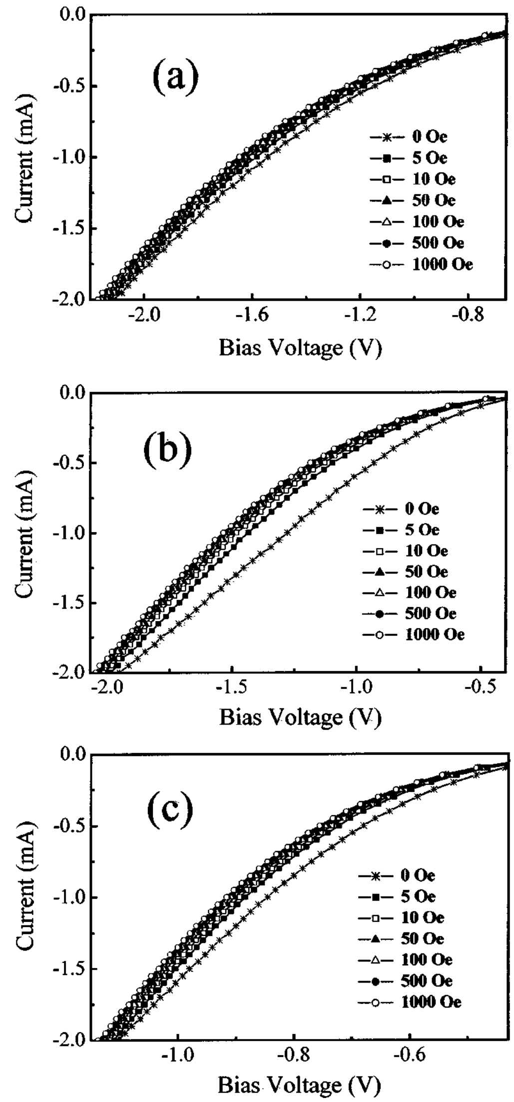

FIG. 3. Modulation of the magnetic fields on the negative bias of the $p-n$ junction at (a) 290 K, (b) 255 K, and (c) 190 K.

The MR ratios of the $p-n$ junction are plotted as a function of the applied magnetic field, as shown in the inset of Fig. 4 for the -0.73 V bias at 290 K, -0.80 V bias at 255 K, and -0.54 V bias at 190 K. The MR is always positive and increases abruptly with the applied magnetic field. For example, the MR ratios are 11% in 5 Oe, 23% in 100 Oe, and 26% in 1000 Oe at 290 K; 53% in 5 Oe, 80% in 100 Oe, and 94% in 1000 Oe at 255 K; 24% in 5 Oe, 47% in 100 Oe, and 53% in 1000 Oe at 190 K. From the point of view of application, a high MR sensitivity near room temperature and at the low field reported here is extremely interesting. Furthermore, the sensitivity of the resistance variation under the applied magnetic field is another prime important factor for potential application. Then, the MR variation versus magnetic field, defined as $(R_H-R_0)/H$, is calculated at the optimal bias voltage and shown in Fig. 4. With a small change from 0 to 5 Oe of the applied magnetic field, the MR sensitivities are 85 Ω/Oe at 290 K, 246 Ω/Oe at 255 K, and 136 Ω/Oe at 190 K. Correspondingly, the maximum change

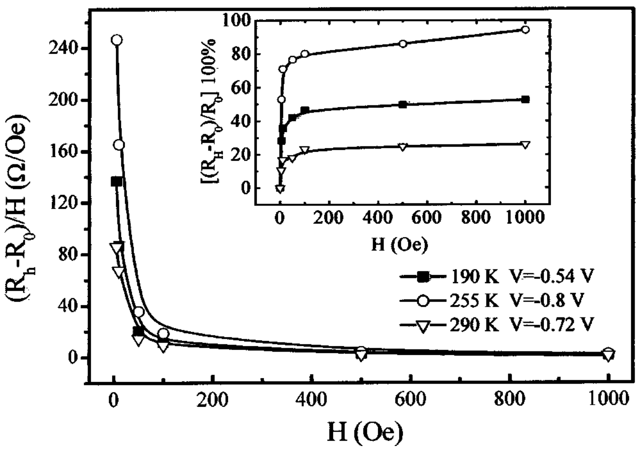

FIG. 4. The sensitivity of MR/H as a function of the magnetic field at optimal bias voltage. Inset: the MR ratios of the *p*–*n* junction as a function of the magnetic field at 290 K and $V_{bias}=-0.73$ V; 255 K and $V_{bias}=-0.80$ V; and 190 K and $V_{bias}=-0.54$ V.

of the bias voltage can be deduced as 45 mV/5 Oe in the current of −0.7 mA at 290 K, 164 mV/5 Oe in −0.6 mA at 255 K, and 50 mV/5 Oe in −0.5 mA at 190 K.

Tiwari *et al.* and Tanaka *et al.* found that the electrical characteristics of oxide heterostructures are strongly modified by the built-in electric field at the junctions.[10,12] In order to understand the mechanism of the positive MR phenomena, the resistivities of the LS0.1MO thin film and the SN0.01TO substrate are measured at room temperature by the van der Pauw method. The resistivities are 2.87 Ω cm for the LS0.1MO and 1.84×10^{-3} Ω cm for the SN0.01TO. The concentrations and mobilities of the charge carrier are determined with the Hall measurement. The hole concentration in the LS0.1MO is 1.19×10^{18} cm^{-3}, and the electron concentration in the SN0.01TO is 1.63×10^{20} cm^{-3}. The carrier mobility in the LS0.1MO is 1.8 cm^2/V s, and it is 33 cm^2/V s in the SN0.01TO. The variations of the resistance as a function of temperature of the LS0.1MO film and SN0.01TO substrate show that the LS0.1MO has a semiconductor behavior between 110 and 300 K, and the SN0.01TO exhibits a metal-like behavior between 4.2 and 300 K. However, the junction resistance is about three orders higher than that of the SN0.01TO substrate and LS0.1MO film. Therefore, the *I*–*V* characteristics and positive MR behavior reported here are certainly associated with the *p*–*n* junction.

Another peculiar behavior of the LS0.1MO/SN0.01TO *p*–*n* junction is that the positive MR ratios exist in a quite large temperature range from 190 to 290 K for magnetic fields up to 1000 Oe. Such a phenomenon is not only different from the $LaMnO_3$ compound with negative MR, but also different from the observation in the MTJ reported by Mitra *et al.*[1] They found that the MR in $La_{0.7}Ce_{0.3}MnO_3/STO/La_{0.7}Ca_{0.3}MnO_3$ MTJ grown on SNTO is negative at small bias currents, and turns positive at higher bias currents at 100 K. But the effect of the magnetic field on the resistance of the MTJ completely disappeares at the high temperature of 300 K. However, the effect of MR in our LSMO/SNTO *p*–*n* junctions still exists at a temperature of 290 K, without a crossover from positive to negative. This pronounced positive MR is indeed unusual, and could not be explained as in the framework of the MTJ. We believe the difference of the MR behavior between our *p*–*n* juction and the MTJ is due to the difference of structures between them and the difference of the microscopic physics in the spin transport of the two structures. Therefore, a further study on the mechanism of the positive MR found in this Letter is highly expected. We speculate that both the concentration of the carrier and the density of state of the electrons at E_f could be affected by the interface of the *p*–*n* junction, and a positive MR might be induced in this way.

In summary, the LSMO/SNTO *p*–*n* junctions are fabricated by laser MBE, and the positive MR ratios from 11% to 53% in the temperature range of 290–255 K can be achieved with a small change in the applied magnetic field from 0 to 5 Oe. Especially, the high MR sensitivity of about 246 Ω/Oe at 255 K and 85 Ω/Oe at 290 K can be observed in the low magnetic field. Our investigation shows the doping concentrations of Sr in LSMO and Nb in SNTO play important roles. In our case, the optimal concentrations are located about 10% for Sr and 1% for Nb in the reported LSMO/SNTO *p*–*n* junctions. It is expected that further investigation on the MR *p*–*n* junction could not only stimulate theoretical study on the mechanisms but also would open up possibilities in device developments.

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