Electrical-modulated magnetoresistance in multi-*p*-*n* heterojunctions of $La_{0.9}Sr_{0.1}MnO_3$ and oxygen-vacant $SrTiO_{3-\delta}$ on Si substrates

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The electrical modulation of the magnetoresistance (MR) from −70% to 80% under a small magnetic field of 200 Oe near room temperature is found in multi-*p*-*n* heterostructures of $SrTiO_{3-\delta}/La_{0.9}Sr_{0.1}MnO_3/SrTiO_{3-\delta}/La_{0.9}Sr_{0.1}MnO_3/Si$ we fabricated. The mechanism causing the modulation of MR by applied bias is proposed as the interface competition effect in the multilayer heterojunctions. Our results of the present structure are expected to meet the high desire for the application of large electronic modulation of MR near room temperature. © *2008 American Institute of Physics*. [DOI: 10.1063/1.3054343]

Electrical modulation of resistance, magnetoresistance (MR), and magnetism in multifunctional films and heterostructures, therefore, has attracted a lot of research activities to find a material system showing magnetoelectric coupling at room temperature, and to explore the opportunity of writing magnetic data with an electric field for data storage applications.[1–5] Current induced switching of resistive states in magnetoresistive manganites has been also explored.[6–9] In particular, the electrical modulation of double exchange ferromagnetism on *p*-*n* junction consisting of $La_{0.9}Ba_{0.1}MnO_3$ and Nb-doped $SrTiO_3$ and a large positive MR in a $La_{0.7}Ce_{0.3}MnO_3/SrTiO_3/La_{0.7}Ca_{0.3}MnO_3$ tunnel junction were reported in 2001 and 2003,[10,11] respectively. Although we recently reported that large positive MR ratios have been created at high temperature and low applied magnetic field, 23% in 100 Oe at 290 K for $La_{0.9}Sr_{0.1}MnO_3$ (LSMO)/ $SrNb_{0.01}Ti_{0.99}O_3$ *p*-*n* junction, 6.4% in 100 Oe at 300 K for $[LSMO/SrNb_{0.01}Ti_{0.99}O_3]_n$ multilayer structure we fabricated by the laser molecular-beam epitaxy (laser MBE),[12] large electrical modulation of MR property has not been found until our present study. In this letter we will present an electric field modulation of large low-field magnetoresistance (LFMR) near room temperature, which was almost a dream lots of physicists in this area are working for. The bias dependence of the MR shows a maximum positive MR at −3.0 V, and a crossover from negative to positive MR occurs at 0.07 V, which offers an approach to control and switch magnetic properties using an electric field. This electric-field-driven MR effect strongly indicates the potential application in low-power high-speed magnetoresistance random access memory (MRAM) technology and beyond based on spintronics.

The junction deposition experiments were directly carried out on a *n*-type silicon substrate by a laser MBE system.[13] First, after wet-chemical cleaning, the Si substrates were dipped in ~5% HF solution for 20~30 s to remove the native silicon oxide on the surface and meanwhile, to form a hydrogen-terminated surface. The Si substrates then were transferred into the epitaxial chamber immediately. The initial deposition of about three unit cells LSMO film was under the base pressure of 5×10^{-6} Pa at room temperature to prevent the formation of the SiO_2 interface layer. After that, the substrate temperature was raised to 620 °C. The thicknesses of the LSMO and $SrTiO_{3-\delta}$ (STO) layers were 100 and 20 nm, respectively. Since $SrTiO_3$ thin films prepared by vacuum processes usually contain oxygen vacancies and possess some characteristics of the *n*-type semiconductors,[14,15] the junction STO/LSMO/STO/LSMO is formed as a *n*-*p*-*n*-*p* heterostructure on *n*−Si substrate. The *in situ* reflection high-energy electron diffraction (RHEED) was used to characterize the crystallization of the junctions. The sharp streak RHEED patterns were observed after the growth of each layer, which is similar to our previous reports, indicating that the junctions have good crystallized structures and smooth interfaces and surfaces.[12,16] The *ex situ* x-ray diffraction patterns show that only a particular set of lines (00*l*) in the spectrum, without any trace for other diffraction peak from impurity phase or randomly oriented grain, is present for all the layers of the structure. The Hall coefficient measurement of the LSMO (100 nm) and STO (20 nm) single layer films confirmed that the carrier concentrations of the LSMO and STO were 1.19×10^{18} and -4.83×10^{19} cm^{-3}, respectively.

The magnetic properties were measured by a superconducting quantum interference device. Figure 1 shows the magnetic hysteresis loops of the sample with *H* parallel to the film plane, and magnetic coercivities are ~75 and 40 Oe for 260 and 350 K, respectively, which clearly demonstrates that the ferromagnetic phase exists near room temperature in the heterostructure.

The current-voltage (*I*-*V*) characteristics of the sample, measured with a pulse-modulated current source, display the typical rectifying property of a *p*-*n* junction over a wide temperature range of 350–80 K, as shown in Fig. 2. The asymmetric *I*-*V* curves are very clear in forward- and reverse-bias measurements. The slope of the *I*-*V* curve becomes steeper in the forward-bias case and the junction resistance decreases with increasing temperature.

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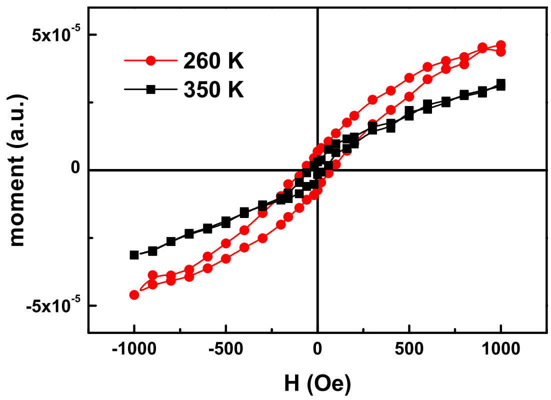

FIG. 1. (Color online) Magnetic hysteresis loops of a typical STO/LSMO/STO/LSMO/Si multi-*p*-*n* junction with magnetic field parallel to the film plane for various temperatures.

The LFMR in perovskite manganite oxides is of special interests due to the fact that this effect may find potential applications in next generation of digital recording and sensing devices. However, LFMR is too small and has a steep decay with temperature in general structures, which is unfavorable for device applications at room temperature. In contrast, the present multi-*p*-*n* junction of LSMO and STO on Si exhibits large LFMR effect near room temperature. Figure 3(a) shows the dependence of MR of the multi-*p*-*n* heterostructure as a function of reverse-bias voltage under $H=200$ Oe, with H applied parallel to the interface of the heterostructure. Here, the 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 H and R_0 is the resistance without a magnetic field. The MR decreases with the increasing temperature, and the maximum value of each curve occurs at about −3.0 V. The mechanism causing the positive MR can be understood by the electron filling in the t_{2g} spin-down band in the space-charge region of LSMO near the interfaces and the tunneling process of carriers from the LSMO homogeneous region into the LSMO space-charge region under reverse bias voltages.[17]

The most interesting phenomenon is the positive bias voltage dependence of LFMR, which shows that a crossover from negative to positive values occurs at ∼0.07 V in the bias increasing process, as shown in Fig. 3(b). The peak with large value of ∼80% and the dip of −70% of the LFMR at 260 K in a very low magnetic field of 200 Oe occur at about 0.075 and 0.05 V, respectively. Furthermore the MRs are ∼−37% and 45% at 300 K, ∼−20% and 20% at 350 K under a magnetic field of 200 Oe. To clearly demonstrate the electric and magnetic fields modulation on the spin polarized transport property of the multi-*p*-*n* heterostructures, we measured the dependence of the resistance and corresponding MR ratio of the multi-*p*-*n* junction on the temperature between 80 and 350 K for the above special bias, −3, 0.05, and 0.075 V shown in Fig. 4. From Fig. 4(a), we can clearly see

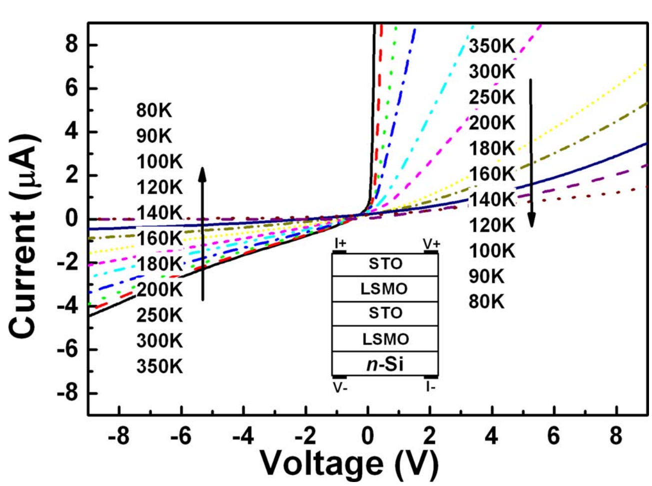

FIG. 2. (Color online) *I*-*V* characteristics of the STO/LSMO/STO/LSMO/Si multi-*p*-*n* junction, taken at various temperatures in zero field. Inset: contact configuration of the junction and the indium electrodes of 0.5 mm^2 were placed on the surfaces of the film and substrate.

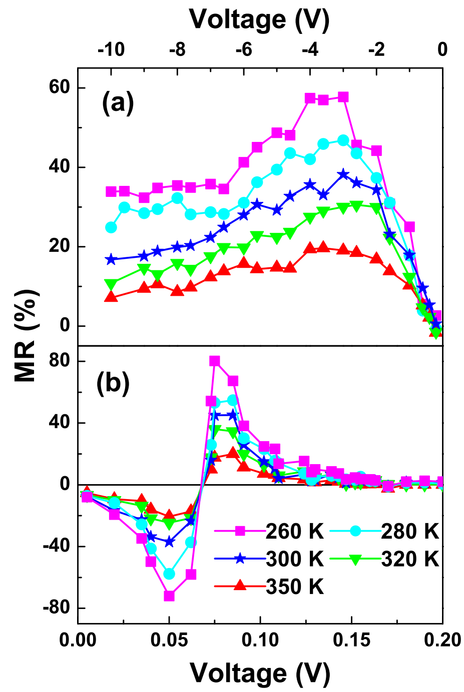

FIG. 3. (Color online) (a) Negative and (b) positive bias voltage dependences of MR with the field $H=200$ Oe applied parallel to the plane at various temperatures.

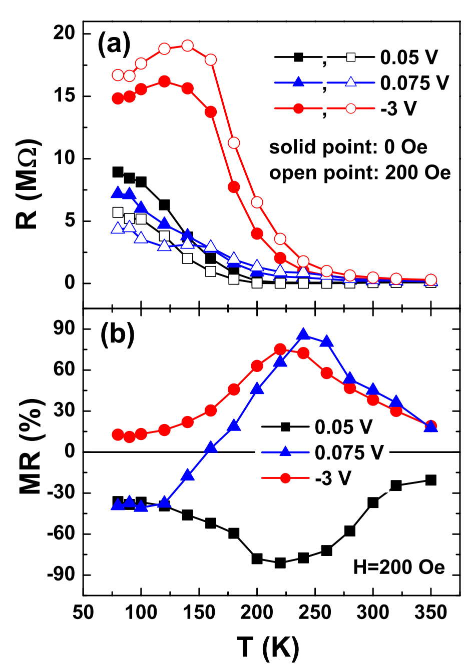

FIG. 4. (Color online) The dependence of (a) the resistance and (b) corresponding MR ratio of the multi-*p*-*n* junction on the temperature between 80 and 350 K for various biases, −3, 0.05, and 0.075 V.

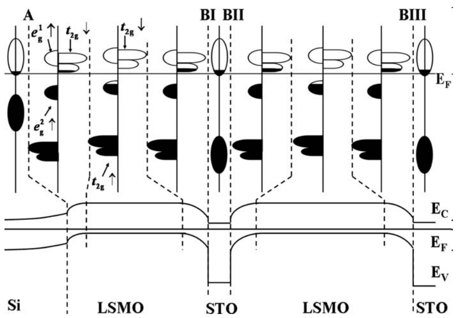

FIG. 5. The schematic electron DOS of the multi-*p*-*n* junction near room temperature and the band diagram for each region. A, BI, BII, and BIII denote the interfaces.

that the resistance reaches its maximum around the temperature of 150 K at the reverse bias of −3 V. Figure 4(b) displays that the multi-*p*-*n* heterostructure shows a positive MR property at −3 V and 0.075 eV, but a negative MR property at 0.05 V in almost all temperatures.

It is well known that near room temperature the intrinsic MR of LSMO film is small and negative based on its phase diagram and transport properties,[18–20] and STO itself is almost without any MR property. So the MR feature of the multi-*p*-*n* heterojunction is to be unequivocally attributed to the heterostructure aspect of the sample and field-induced spin modulation and interface phenomena.[17] To understand the mechanism of the forward electrical field modulated MR reversion (from negative to positive MR) shown in Fig. 3(b), similar to the model we reported previously,[17] we schematically plotted the band structure (lower one) and the spin polarized density of states (DOS) for carriers (upper one) in each region of the system, as shown in Fig. 5. With reverse-bias voltage, the positive MR occurs in all interface regions of A and BI, BII, and BIII so that the system shows positive MR in all values of reverse bias, as shown in Fig. 3(a). As demonstrated in our previous work, positive MR only occurs when carriers flowing in the system are with antiparallel spin states at different regions.[17] At forward bias voltage, interface A region shows negative MR, meanwhile, interface BI, BII, and BIII regions show positive MR. Actually there is a competition mechanism of positive and negative MR in the system mainly caused by the four interfaces of A, BI, BII, and BIII. As we have reported in our previous work,[15–17] the carrier density in Si is much lower than that in LSMO so that the charge region of Si at the interface A is much larger than that of LSMO in the interface B, as well as the resistance. Therefore the resistance at region A is much larger than the sum of the resistances in regions BI, BII, and BIII. At very small forward bias, the resistance in A region is still quite large due to the barrier in interface A for both electrons and hole so that the resistance at interface A can still be dominant in the system. In this case, the system shows the intrinsic negative MR property of LSMO, which is the case of MR curve at bias smaller than 0.07 V shown in Fig. 3(b). With bigger bias, tunneling of carriers occurs at the interface of BI and BIII, thus the doping holes in t_{2g} spin-down states at interface region BI (BIII) of LSMO are more involved in the transport, while e_g^1 spin up carrier play an important role for transport in the region far from the interface of LSMO.[17] Meanwhile, the resistance of interface A is getting smaller with the increasing of the forward bias so that the positive MR dominates in the system, which is the case of MR curve at larger bias than 0.07 V shown in Fig. 3(b). The competition of the transport properties at interfaces A, BI, BII, and BIII results in the modulation of MR values by bias. In much larger absolute values of bias, the e_g^2 spin up carriers in interface regions of LSMO start to get involved in the transport, and the spin parallel carriers between the interface region and the homogeneous region of LSMO play a more and more important role in transport,[17] which should be the reason for the decay of positive MR with very large bias voltages shown in both Figs. 3(a) and 3(b). Based on the present scenario, we predict that the variation of the structure should cause the variation of electrical modulation on MR behavior, which is expected to be proved by further study.

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