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Effects of magnetic field on the manganite-based bilayer junction

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An oxide bilayer junction has been fabricated by growing a $La_{0.32}Pr_{0.35}Ca_{0.33}MnO_3$ film on 0.5 wt % Nb-doped $SrTiO_3$ crystal, and its behavior under magnetic field is experimentally studied. It is found that external field greatly affected the rectifying property and the resistance of the junction, causing an extremely large magnetoresistance. The most striking observation of the present work is that the magnetoresistance of the junction can be either positive or negative, depending on temperature and applied current, and is asymmetric with respect to the direction of the bias current. These results reveal the great potential of the manganites in configuring artificial devices. © *2004 American Institute of Physics.* [DOI: 10.1063/1.1664032]

Doped manganese oxides have been a focus of intensive studies since the discovery of colossal magnetoresistance (MR) in this kind of materials. Much effort, both theoretical and experimental, has been devoted to revealing the physics underlying the colossal MR and other phenomena such as phase separation, charge-and-orbital ordering, and etc.[1] In addition to these, the practical applicability of the MR effects is also a topic of great interest. Unfortunately, the prospect for a direct application is not clear due to the difficulties that the magnetic field required for a significant MR is too high, usually exceeds several Tesla, and the maximum MR is limited to a very narrow temperature region. In contrast, it becomes more and more obvious that the great potential of the manganites may be in artificial structures. In fact, a substantial progress has been gained in the fabrication of oxide magnetic tunnel junction,[2–4] and a tunneling MR as high as 99.9% has been obtained in trilayer junction $La_{0.67}Sr_{0.33}MnO_3/SrTiO_3/La_{0.67}Sr_{0.33}MnO_3$.[4] Obviously, the magnetic tunnel junction is not the only artificial device composed of manganites. Recently, Kawai and co-workers have successfully constructed a $p-n$ junction using Nb or La-doped $SrTiO_3$ and $La_{0.9}Ba_{0.1}MnO_3$, and found a rectifying behavior and a strong bias voltage dependence of the junction resistance.[5,6] A typical feature of the manganites is the strong dependence of their property on magnetic field. Therefore, a natural anticipation is that the property of the $p-n$ junction would be magnetically tunable, which is of special interest from the viewpoint of application. It is unexpected that no such effects were observed by Kawai *et al.* This may be a result that the MR of $La_{0.9}Ba_{0.1}MnO_3$ is not large compared with other manganites: it is ~50% near the Curie temperature (T_c) and ~10% well below T_c under a field of 5 T.[7] As is well known, replacing La in $La_{0.67}Ca_{0.33}MnO_3$ by smaller atoms usually causes a dramatic enhancement of the MR effects, and sometimes a metamagnetic transition can occur in such manganites.[8] What will be the behavior of the bilayer junction composed of this kind of manganites under external field is an interesting question. In this letter, we constructed a bilayer junction using the $La_{0.32}Pr_{0.35}Ca_{0.33}MnO_3$ (LPCM) film and the 0.5 wt % Nb-doped $SrTiO_3$ (STON) crystal, and focused on its behavior under magnetic field.

The LPCM-STON junction was fabricated by growing a LPCM film on the STON crystal of the area of 3×4 mm^2 using laser ablation. The substrate was kept at ~800 °C and the O_2 pressure at 100 Pa during the deposition. The film thickness is ~500 Å, controlled by deposition time. As confirmed by the θ–2θ and ϕ scans of the x-ray diffraction, the film is epitaxial with the (001) axis aligning along the film normal. Thermomagnetization demonstrates that the ferromagnetic (FM) onset occurs at T_c~166 K and the magnetic order in the LPCM film is far from perfect (not shown). The resistivity-temperature curves in Fig. 1 indicate that the film is metallic below $T_p \approx 155$ K and activated above T_p. Magnetic field depresses the resistivity greatly. A field of 5 T causes a MR of approximately −99% at ~150 K and approximately −70% at 20 K (inset in Fig. 1), and drives the metal-to-insulator (MI) transition to ~220 K, where MR is defined by $\rho(H,I)/\rho(0,I)-1$ (I is explicitly included to express the current dependence of resistance as will be seen below). All these features, which are essentially similar to those observed in bulk materials,[8] suggest a change of the magnetic and electronic structure of the LPCM film under magnetic field as desired.

The $I-V$ characteristics of the LPCM-STON junction

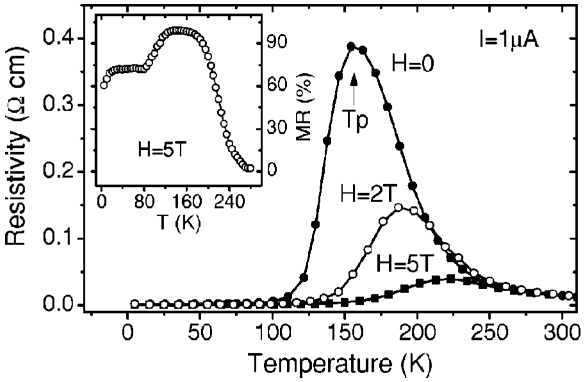

FIG. 1. Temperature-dependent resistivity of the $La_{0.32}Pr_{0.35}Ca_{0.33}MnO_3$ film measured at $H=0$, 2 T, and 5 T. Inset displays the magnetoresistance of the film under a field of 5 T.

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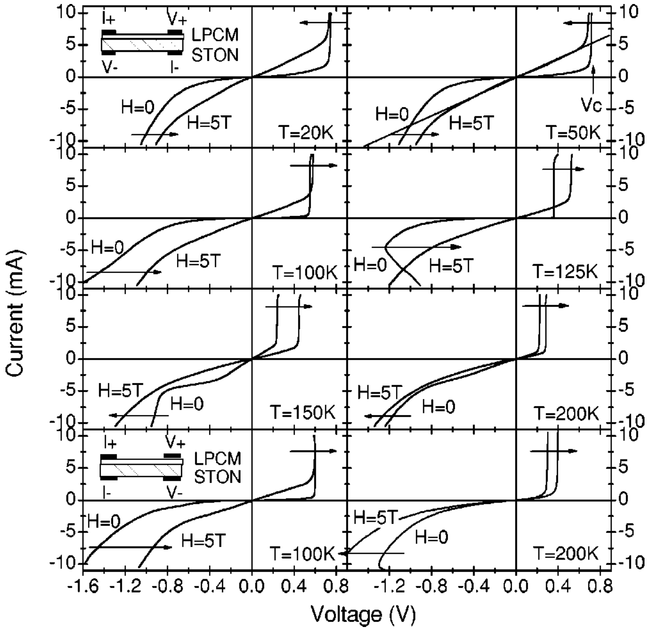

FIG. 2. Current–voltage characteristics of the LPCM-STON junction measured under the fields of $H=0$ and 5 T. Results in the top six panels are measured with the electrode settings in the top inset, and the bottom inset for the two panels; arrows indicate the change of the voltage under magnetic field.

are measured by tuning the applied current, and the typical results are presented in Fig. 2. Two different electrode settings are used for the measurements (insets of Fig. 2), and the only the results corresponding to the top setting are presented in detail because of the similarity of the two sets of data. As reported by Kawai *et al.*, the $I-V$ curves are asymmetric with respect to the origin, demonstrated by the occurrence of a voltage-saturation at V_c for the positive branch of the $I-V$ curve and the continuous growth of the voltage in the negative branch. This asymmetry persists up to room temperature despite the change of the detailed $I-V$ relation. The most remarkable observation of the present work is the strong effects of magnetic field on the $I-V$ curve and the resistance of the junction (defined as $R_{\text{jun}}=V/I$). First, the magnetic field modifies the $I-V$ relation significantly. It straightens the $I-V$ curve, inclines it to the I axis, and pushes V_c to higher voltages. The first two features are obvious in the temperature region below 125 K, while the third feature appears and develops above 100 K. Take the result at 50 K as an example. The $I-V$ curve displays as a straight line with a large slope in the range from -0.48 V to 0.63 V under a field of 5 T, whereas it is a curve close to the V axis without external field. These results imply a weakening of the rectifying ability of the LPCM-STON junction under magnetic field. Second, the effect of magnetic field is different for the positive and negative portions of the $I-V$ curve. For the former, it leads to a slight decrease of the voltage when $T<75$ K and a considerable increase when $T>100$ K. In contrast, the voltage decreases for $T<125$ K and increases for $T>150$ K for the latter.

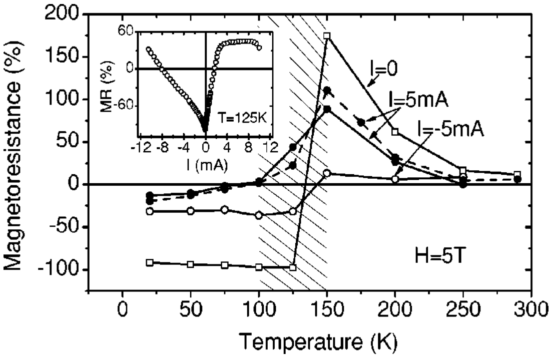

FIG. 3. Magnetoresistance of the LPCM-STON junction measured under a field of 5 T for selected current $I=-5$, 0, and 5 mA. Inset displays the magnetoresistance of the LPCM-STON junction as a function of bias current ($H=5$ T and $T=125$ K); solid lines are guides for the eye; dashed line shows the data obtained with the electrode setting in the bottom inset of Fig. 2. Hatched area indicates the temperature region within which the sign of the magnetoresistance is strongly current dependent.

All these effects are clearly demonstrated by the variation of the junction resistance. Figure 3 presents the typical MR of the junction for selected currents $I=-5$ mA, 5 mA, and 0 (obtained by extrapolating the results to the zero-current limit). Corresponding to the inclining of the $I-V$ curve under magnetic field, the junction resistance exhibits a significant reduction, and the MR ratio is as large as $\sim-96\%$ for $H=5$ T. This is a value comparable to that of the manganites. Different from the conventional manganites, however, the MR of the LPCM-STON junction can be positive. As shown in Fig. 3, with the increase of temperature, the MR undergoes a dramatic negative-to-positive transition, and a huge positive MR of ~180% is observed when $I=0$. In the intermediate temperature range between 100 and 150 K (hatched area in Fig. 3), the sign of MR depends on the applied current, and a large positive bias current usually yields a positive MR. One point worthy of special attention is that the MR is asymmetric with respect to the direction of the applied current. It is, for example, ~0 for $I=1.76$ mA and -67% for $I=-1.76$ mA at $T=125$ K. The MR is modulated simply by the direction of the bias current. This result indicates that the asymmetric character of the $p-n$ junction is an advantage to obtain unexpected effects. The inset in Fig. 3 illustrates the variation of MR against applied current at $T=125$ K.

Negative MR has been a focus of recent studies, and the well known phenomena are the giant MR in metallic multilayers, the colossal MR in manganites, and the tunneling MR in spin tunnel junction. Different from the negative MR, the conventional MR in metals and semiconductors is positive and usually small. To our knowledge, the rich variation of MR versus temperature, current, and magnetic field in the LPCM-STON junction has not been observed before in either artificial or natural materials. As a primary explanation, we prefer to relate it to the variation of the electronic structure of LPCM. In Fig. 4 we schematically show the band structure of LPCM and STON.[9] In the hole-doped manganites, three Mn $3d$ electrons form the localized t_{2g} band (not shown). The remaining electrons occupy the conducting e_g band which is energetically higher than the t_{2g} band. The e_g band further splits into two subbands of spin-up and spin-down ($e_g\uparrow$ and $e_g\downarrow$) due to Hund's rule coupling,[10] and the Fermi level locates in the $e_g\uparrow$ subband. An energy barrier could be formed at the LPCM-STON interface noting that the Fermi energy of the degenerated n-type semiconductor STON is near the conducting band.[11] When the voltage on the junction exceeds V_c, this energy barrier is overcome, and

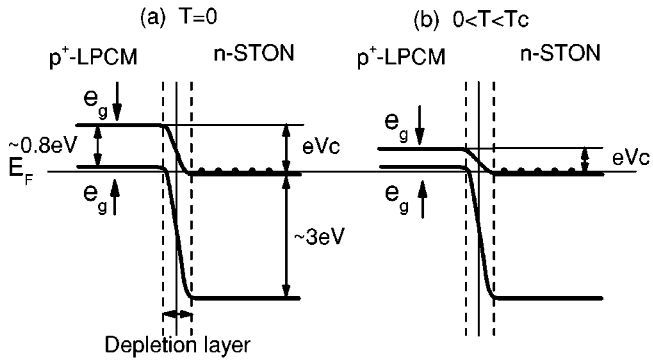

FIG. 4. Schematic diagrams for the band structure of LPCM and STON at (a) $T=0$ and (b) $0<T<T_c$.

the snowslide-like current increase appears. The $e_g\uparrow-e_g\downarrow$ band gap is expected to vary with the magnetic state of LPCM. In fact, when the spins of the Mn^{4+} ions deviates from a fully ferromagnetic alignment, an exact parallel or antiparallel arrangement between the spins of the e_g electrons and the Mn^{4+} ions is impossible. This implies a reduction in the energy cost for the e_g electron to reverse its direction. Therefore, with the increase of temperature, the energy gap between e_g bands will decrease and, finally, vanish when $T>T_c$.[12] As a result, V_c decreases with temperature. This analysis is consistent with the experiment observation. As shown in Fig. 5, the most drastic decrease of V_C takes place near the Curie temperature of the LPCM film. It is in this temperature regime that the FM order of the film changes rapidly. On the contrary, when the FM order of the LPCM film is improved by an external field, the band gap reopens up, yielding an increase of V_c (Fig. 5).

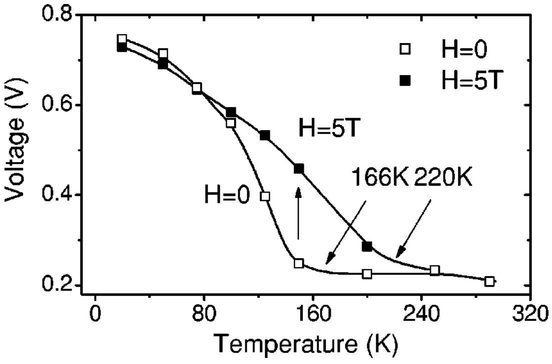

FIG. 5. Variation of the saturation voltage (V_c) as a function of temperature. The arrow marks the increase of V_c under magnetic field. The maximum energy barrier at the LPCM-STON interface ($eVc\sim0.75$ eV) is the same order of the $e_g\uparrow-e_g\downarrow$ band gap (~0.8 eV).

The finite zero-bias resistance of the junction could be a result of electron tunneling from the conducting band of STON to the $e_g\uparrow$ band of LPCM. It exhibits a rapid decrease above 150 K (Fig. 2), which is probably due to the increase of the density of state near the Fermi level after the overlap of the two e_g subbands. In this image, when external field drives the system back to the FM state, R_{jun} will increase (positive MR) due to the resplitting of the e_g subbands. While the negative MR in the low temperature regime may be attributed to the variation of the depletion layer. It becomes thinner when the magnetic field derives LPCM into a well metallic state, which enhances the electron tunneling. Obviously, what we give here is only a possible explanation. A through understanding of the present observation requires a full knowledge on the band structure of LPCM.

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