Large magnetoresistance effects near room temperature in manganite heterojunction

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Magnetoresistive property of a heterojunction composed of $Pr_{0.6}Ca_{0.4}MnO_3$ (PCMO) and Nb-doped $SrTiO_3$ (STON) has been experimentally studied. A rather complex current–voltage relation characterized by the appearance of a low-bias electric breakdown prior to the reverse current saturation process is observed. The magnetic field shows a strong depression to this electric breakdown and, as a result, leads to a great change of junction resistance. It is interesting that the magnetoresistance thus produced, remains huge, ~1600% under a field of 5 T [defined as $R(H)/R(0)-1$], in a broad temperature range well above the Curie temperature of PCMO, which indicates a different mechanism of the magnetic field effect of PCMO/STON from other manganites and manganite-based heterojunctions. A qualitative explanation is given based on an analysis about the influence of interfacial defects on Hall electric field. © *2004 American Institute of Physics*.
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One of the most remarkable features of the manganites is their strong magnetic and electronic correlation.[1] It has been well established that when the manganites undergo a paramagnetic (PM) to ferromagnetic (FM) transition, the two degenerated e_g subbands, associated, respectively, with the spin-up and spin-down states, will experience a band splitting. This assigns the manganites a magnetically tunable semiconductor character that is particularly valuable in the design of artificial material/device with unusual properties.

A typical example of manganite-based artificial materials is the *p-n* heterojunctions composed of divalent ion-doped $LaMnO_3$ and La/Nb-doped $SrTiO_3$.[2–6] In addition to the excellent rectifying property,[2–4] it has been found that the diffusion/breakdown voltage and the resistance of the junctions experience a dramatic increase at the temperature corresponding to the magnetic transition of the manganites. A great modification of magnetic field to the rectifying and resistive behaviors was also observed.[5,6] In the scenario of the buildup of interfacial potential due to the mismatch between the band structures of the manganite and $SrTiO_3$:Nb/La and the variation of this potential under external fields, these observations can be understood qualitatively.

Similar to manganites, the magnetic field effect in manganite heterojunctions is strongly temperature dependent. It mainly occurs in a narrow temperature range adjacent to the Curie temperature (T_C) of the manganite and becomes stronger with the decrease of T_C. This is consistent with the image that magnetic field affects the junction by modifying the magnetic/electronic structure of the manganite. However, as a material with special band structure, characterized by the presence of interfacial energy barrier, and corresponding electronic processes, the heterojunction is expected to exhibit distinctive properties absent in its constituents.[7] Based on this consideration, in this letter we have performed a systematic study on the heterojunction composed of a *p*-$Pr_{0.6}Ca_{0.4}MnO_3$ (PCMO) and an *n*-$SrTiO_3$ doped by 0.5 wt %Nb (STON), and special attention has been paid to the difference between PCMO/STON and PCMO. The most important discovery of the present work is the decoupling of magnetoresistance (MR) of PCMO/STON with the change of the magnetic order of PCMO. The MR, which originates from the depression of magnetic field to the electric leakage of the junction, remains huge in a wide temperature range well above the Curie temperature of PCMO, indicating a completely different MR mechanism.

The heterojunction PCMO/STON was fabricated by growing a PCMO film on a STON substrate of the (001) orientation following the procedure described elsewhere.[5] The film is 3×5 mm^2 in size and ~2000 Å in thickness. As confirmed by x-ray diffraction study, it is single phase and epitaxially grown with the (001) axis normal to the film plane.

Figure 1 presents the resistivity (in-plane) and magnetization of the PCMO film as functions of temperature. Similar to its bulk counterpart, the PCMO film exhibits a semiconducting behavior characterized by the exponential growth of the resistivity with the decrease of temperature. Similar to other manganites, the application of magnetic field depresses the resistivity of PCMO. This effect is not obvious above 150 K, but very strong below ~120 K. A simple calculation gives a MR of ~40% at 200 K and ~730% at 120 K, where $MR=R(0)/R(H)-1$ with $R(0)$ and $R(H)$ being resistances without and with magnetic field, respectively. A sign of charge ordering transition, though blurry, is also identifiable from the $1/M$ vs T curve (inset in the bottom panel of Fig. 1),[8] which depresses the ferromagnetic order of the PCMO film. As shown by Fig. 1, long range magnetic order in PCMO, if exists, appears below ~140 K.

The current–voltage ($I-V$) characteristics of PCMO/STON are measured in the temperature range from 20 up to 295 K with the magnetic field in film plane. Only the data above 150 K were presented here. The large resistivity of the PCMO film has disturbed the reliable determination of the $I-V$ relations below 150 K. Compared with the previously reported manganite heterojunctions, PCMO/STON exhibited

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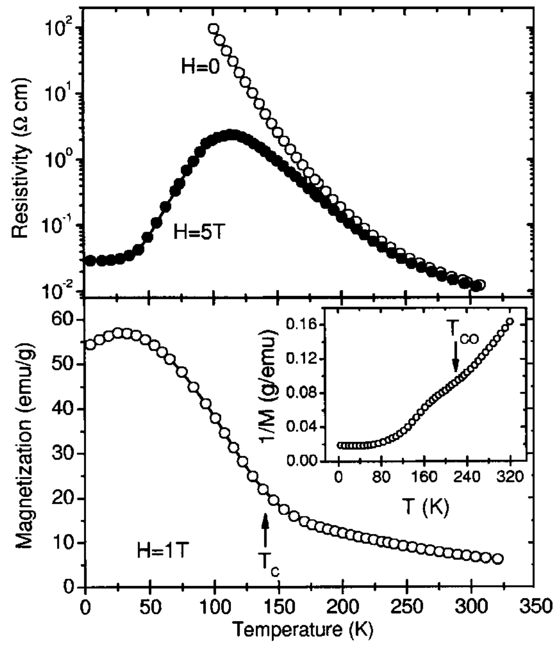

FIG. 1. In-plane resistivity (top panel) and magnetization (bottom panel) of PCMO as functions of temperature. Bottom inset displays the reciprocal magnetization vs temperature. T_{CO} marks the possible charge ordering transition.

a rather complex $I-V$ relation without external magnetic field. As shown by the $I-V$ curves recorded at $T=265$ K in Fig. 2, in addition to the two current rushes at V_b (breakdown voltage) and V_d (diffusion voltage), respectively, which are typical features of *p*-*n* junctions, the $I-V$ relation in the intermediate bias region decomposes into two distinguishable processes. The first one is the rapid growth of the current with bias voltage when the latter is small. A simple calculation indicates that in this case the junction resistance (R_J) is rather small, $\sim$10 Ω in the zero bias limit, manifesting the occurrence of significant electrical leakage. The second process begins as the reverse bias approaches $\sim$0.4 V, which causes first a gradual slowdown of the current increase then a current saturation up to the bias voltage $\sim$1.4 V.

The four-step $I-V$ variation could be a distinctive feature of PCMO/STON noting the regularity of the $I-V$ curves from 295 to 265 K and to 240 K though the complete $I-V$ relations are not obtained below 240 K due to the limit of our experiment setup, for which the upper current bound is 50 mA. Similar behaviors were also observed in the $La_{0.32}Pr_{0.35}Ca_{0.33}$ MnO_3/STON junction, though not obvious because of the presence of La (Fig. 2 in Ref. 5), which signifies the generality of this phenomenon in manganite junction.

The most remarkable observation of the present work is the strong effects of magnetic field (H) on the $I-V$ dependence, especially in the low bias regime. As shown in Fig. 2, magnetic field dramatically depresses the electrical leakage and drives the system directly into the current saturation state as a reverse bias is applied, producing the typical $I-V$ relation of the conventional *p*-*n* junctions. This process occurs in the entire temperature range investigated. It is interesting to note that, though the significant declining of the $I-V$ slope, changes in diffusion voltage are not obvious, and V_d is actually unaffected above 200 K. This result implies the different origins of the magnetic field effects on electrical leakage and diffusion voltage.

Figure 3 presents the zero-bias junction resistance and the corresponding magnetoresistance as functions of temperature. As mentioned in the previous paragraph, R_J is quite small, varying between $\sim$3.6 and $\sim$30 Ω without magnetic field. This reveals the fact that the junction is at the edge of electrical breakdown. The application of magnetic field causes an obvious upward shift of the $\ln R_J - T$ curve, producing a huge and positive MR. The maximum MR occurs at $T=150$ K, where R_J increases from $\sim$3.6 Ω for $H=0$ to 66.2 Ω for $H=5$ T, and the resulting MR $[R_J(H)/R_J(0)-1]$ is $\sim$1800%.

It is instructive to compare the MR of PCMO/STON with that of PCMO. According to Fig. 3, the MR is of the

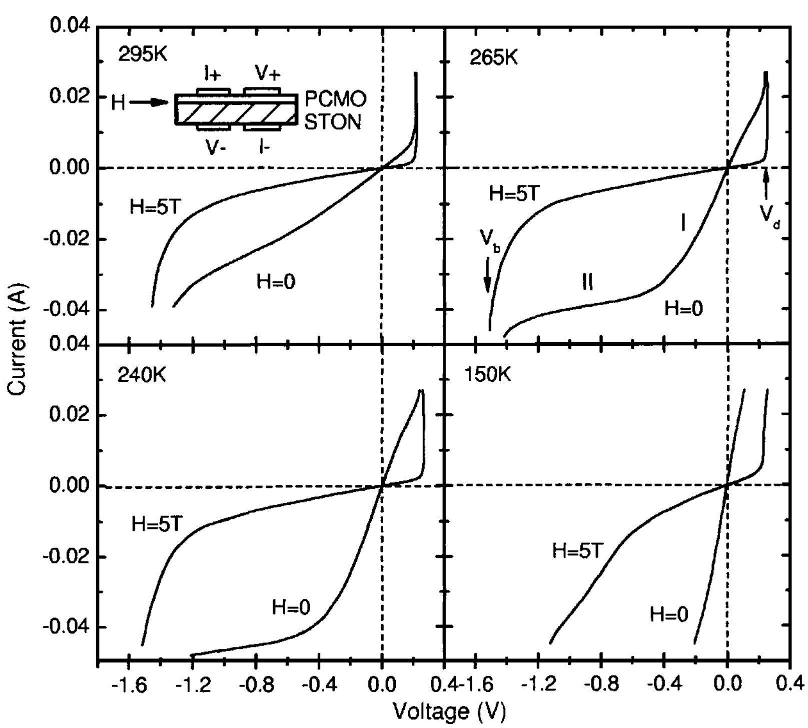

FIG. 2. Current vs voltage characteristics of PCMO/STON measured by tuning bias current. The top inset is a schematic electrode setting for the resistive measurement. Roman numbers in the figure ($T=265$ K) mark the two resistive processes.

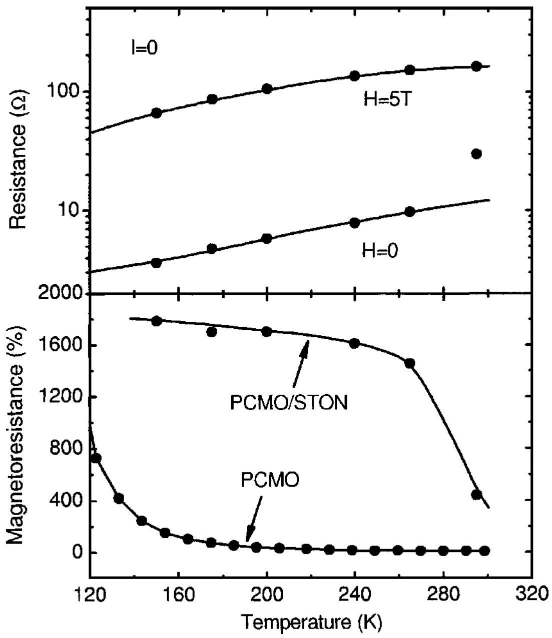

FIG. 3. Top panel: Junction resistance as functions of temperature for PCMO/STON obtained in the zero-bias limit. Bottom panel: Temperature-dependent magnetoresistance of PCMO/STON calculated for $H=5$ T. Results for PCMO are also shown for comparison. Solid lines are guides for the eye.

order of ~1500%–1800% in magnitude for the former while below 150% for the latter between 150 and 265 K, that is, the MR of the junction can be much greater than that of its constituents. This result is interesting in the sense that it demonstrates the possibility to gain great MR in a broad temperature range by combining materials without or with minor MR.

In fact, a significant magnetic field effect has also been observed previously in manganites or manganite-based heterojunctions.[5,6] It usually exhibited as a simultaneous variation of junction resistance and diffusion voltage in the vicinity of the Curie temperature of the manganites, and was ascribed to the change of the magnetic order, or band structure, of the manganites under external field. It is interesting to note that the MR of PCMO/STON remains huge in a wide temperature range. Only a slight MR degeneration is observed when the temperature increases from 150 to 265 K, and the MR is still considerably large even at room temperature (~440%). This means the absence of a direct relation between the MR of PCMO/STON and the change of the magnetic order of PCMO noting the fact that the Curie temperature of the latter is quite low (~140 K).

It is obvious that the low-bias electrical leakage is basically important for the occurrence of huge MR in PCMO/STON. This electrical leakage can be a result of a soft electric breakdown in the sense that it yields merely a limited current growth, and the initial state is restorable after the removing of bias voltage. The magnetic field effect exhibits mainly as a depression of the soft breakdown, and a field of 5 T actually eliminates this process completely. We noticed that a similar result has recently been reported for a Schottky junction composed of Au and AsGa.[9] Similar to PCMO/STON, this junction experienced first an electric breakdown then a slow current growth with the increase of bias voltage. A field of 0.5 T prevents the electric breakdown effectively. The underlying physics for these phenomena is still not very clear at present. A possible reason may be the presence of interfacial defects. According to the semiconductor theory,[7] a high resistivity interface layer will be formed when PCMO and STON are brought into contact due to the mismatch of band structure, carrier type/density of the two materials. This resistive layer will be uneven if interfacial pits or pinhole-like defects exist, which is possible noting the large junction area of PCMO/STON (~15 mm^2), and the soft electric breakdown may take place at these points. When a magnetic field is applied in parallel to the film plane, a Hall electric field will be established in PCMO and STON to balance the Lorentz force experienced by moving charges. The Hall electric field can be different, both in strength and direction, for the two layers of the junction because of their different carrier type/density, and insulated by the high resistivity interface. It is obvious that the presence of interfacial defects could weaken the insulating barrier, leading to a redistribution of the Hall field near these defects. This actually implies the presence of inhomogeneous Hall field adjacent to the interface of the junction. As a result, the Hall field cannot exactly balance the Lorentz force at these defects, and the charge carriers will be swept away from the defects either by the former or by the latter, leading to the depression of electric breakdown. In this scenario, the magnetic field effect should be sensitive to the interfacial state of the junction. Indeed, it is found that the soft electrical breakdown disappears after a postannealing at 700 °C at 20 min in air, and the typical $I-V$ relations of the p-n junctions are resulted. The present work reveals the great potential of the heterojunction as an artificial material with unusual magnetic and resistive properties.

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