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Rectifying properties and colossal magnetoresistance in $La_{0.9}Hf_{0.1}MnO_3$ /Nb-0.7 wt%-doped $SrTiO_3$ heterojunction

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ABSTRACT

A heterojunction with good rectifying properties in a wide temperature range from 20 K to 300 K was fabricated simply by depositing an as-grown $La_{0.9}Hf_{0.1}MnO_3$ (LHMO) film on a commercial 0.7 wt% Nb-doped $SrTiO_3$ single crystal substrate using pulsed laser deposition technique. The current-voltage behavior of the LHMO/STON is measured under applied magnetic fields varying between 0 and 5 T. The heterojunction shows a remarkable magnetoresistance which depends on both the temperature and bias voltages. The sign of the magnetoresistance as function of temperature at either forward or reverse bias voltage is extensively studied by the filling of electrons in the e_g and t_{2g} band. The good rectifying behaviors, the magnetic tunable properties and the maximum magnetoresistance obtained at room temperature make this simple heterojunction promising for practical applications.

Keywords: magnetoresistance, manganite, heterojunction

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I. INTRODUCTION

Perovskite manganites have been extensively studied for their huge magnetoresistance (MR) and many other interesting properties.[1] Recently much attention has been paid to the manganite-based heterojunctions for their good rectifying properties similar to traditional semiconductor junctions. In addition to these, since a typical feature of the manganites is the strong dependence of their properties on magnetic filed, a spontaneous expectation is that the properties of these manganite-based heterojunctions can be modulated by the magnetic field, which is of special interest from the viewpoint of application. In fact, much effort has been devoted to this field. Although good results of negative MR have been reported in some magnetic tunnel junctions (MTJ),[2,3] an extremely large positive MR (~180%) was found in $La_{0.32}Pr_{0.35}Ca_{0.33}MnO_3$ based p-n junction.[4] Furthermore, a crossover from negative to positive MR of $La_{0.7}Ce_{0.3}MnO_3$-$SrTiO_3$-Nb heterojunction was observed by Sheng *et al*.[5] It is understandable for certain manganite based artificial structures to be with a negative MR property since it may originate from the negative MR of the manganites. However it is very interesting and unbelievable that a structure consisting of a negative colossal MR material and a nonmagnetic material (such as Nb-doped $SrTiO_3$) can show a large positive MR property. Besides, the maximum MR is usually observed at very low temperature or in the vicinity of transition temperature. This is inapplicable in semiconductor devices which must be operated at higher temperature even near room temperature. Thus it is highly desirable to find new materials and simple structures with a large MR at high temperature. Perovskite $La_{0.9}Hf_{0.1}MnO_3$ (LHMO) is a tetravalent cation-doped manganite in which the La^{3+} ions of the parent compound $LaMnO_3$ are substituted by Hf^{4+} ions. Our previous reports indicate that the LHMO film is in a mixed valence state of Mn^{2+} and Mn^{3+}, implying an electron-doped conduction mechanism, and it show similar MR and electroresistance (ER) effect as hole-doped manganites.[6,7] However, studies on this electron-doped manganite based heterojunction have seldom been reported so far.

In this piece of work, we successfully constructed a bilayer heterojunction by combining a manganite LHMO with a 0.7 wt % Nd-doped $SrTiO_3$ (STON). The heterojunction shows excellent rectifying properties and a remarkable MR in a wide temperature range from 20 to 300 K. It is found that the filling of electrons in the interface of the heterojunction, which is highly affected by the temperature, plays a crucial role in the sign of MR.

II. EXPERIMENTAL

The epitaxial LHMO film was grown on a 0.7 wt% Nb-doped $SrTiO_3$ single crystal substrate (LHMO/STON) by using pulsed laser deposition technique. A disk-shaped ceramic target of the stoichiometric $La_{0.9}Hf_{0.1}MnO_3$ was synthesized by the standard solid reaction technique using high-purity powdered La_2O_3, HfO_2 and MnO.[8] The film deposition took place in pure oxygen with a pressure of ~0.5 mbar. The energy of the laser beam was ~400 mJ with a wavelength of 248 nm and a pulse frequency of 5 Hz. The substrate temperature was kept at 700 ℃ as measured by a k-type thermocouple inserted into the heater block. The thickness of the films (~280 nm) was measured by surface profile measuring system (DEKTAK3). After deposition, the chamber was vented to 1 bar with high purity oxygen and the sample was cooled down to room temperature. As confirmed by X-ray diffraction (SIEMENS D5000 with Cu-Kα radiation), they are single phase and epitaxially grown with the (001) axis normal to the films plane. The current-voltage behavior of LHMO/STON heterojunction was measured for temperature ranging from 20 to 300 K, and the measuring current was kept below 1 mA to minimize the heating effect.

III. RESULTS AND DISCUSSIONS

The I-V curves of LHMO/STON, measured at selected temperature (60 K,120 K, 180 K, 240 K and 300 K) under the fields of H = 0 and 5 T, are presented in Fig. 1. A schematic illustration of this heterojunction is also shown in the left top of Fig. 1. The positive bias is defined as the current flows from the LHMO film to the STON substrate. A magnetic filed was applied perpendicularly to the interface of the heterojunction. It is notable that the I-V curves exhibit an excellent rectifying property, whatever with or without magnetic fields, in a wide temperature range from 60 to 300 K which mimics the conventional bipolar junction. In the I-V curves, two particular voltages called diffusion voltage (V_d) and breakdown voltage (V_b) respectively are defined at which the current $|A|$=1 mA, as marked in the right bottom of Fig. 1 by arrows.

In order to analyze the junction more straightforward in the later part of this paper, the density of state (DOS) of

LHMO/STON heterojunction is plotted in Fig. 2. The gray areas mark the states being occupied by electrons. In this sketch, region Ⅰ denotes the LHMO homogenous region far away from the LHMO-STON interface, region Ⅱ the LHMO depletion layer closer to the interface, region Ⅲ the STON depletion layer closer to the interface, and region Ⅳ the STON homogenous region far away from the interface. E_g denotes the band gap between the t_{2g} spin-down ($t_{2g}\downarrow$) band and e_g^1 spin-up ($e_g^1\uparrow$) band. E_F is near the bottom of $t_{2g}\downarrow$ band for region Ⅰ due to the low doping of tetravalent Hf ions in LHMO and slightly above the bottom of the Ti 3d conduction band for region Ⅳ. Noting that the E_F of STON is generally higher than that of LHMO,[9] when they are attached together, some electrons in STON will diffuse to LHMO to line up the Fermi lever, as shown in Fig. 2(a).

The V_d as function of temperature for LHMO/STON heterojunction measured under the fields of H = 0 and 5 T are shown in Fig. 3. Whatever with or without magnetic fields, V_d decreases monotonously as the temperature increasing. Here the decreasing V_d means a weakening built-in electric filed in the junction region or a lowering energy barrier at the LHMO-STON interface. When temperature increased, the depletion region becomes thinner due to thermionic emission of carriers, similar to the case of conventional bipolar junction, which accounts for the smaller V_d at high temperature.

The temperature dependence of the MR ratio (I = 1 mA) is shown in the inset of Fig. 3. The most remarkable observation in our sample is that the positive MR increases with increasing temperature and the maximum (about 50%) are obtained at 300 K which is of much favorable to practical applications. As the schematic DOS shown in Fig. 2, both LHMO and STON should be regarded as degenerated semiconductor with corresponding band gap. When a forward bias is applied to the junction, the number of electrons filled in the $t_{2g}\downarrow$ band in the region Ⅱ decreases, as shown in Fig. 2(b). The positive MR shown in our LHMO/STON heterojunction can also be explained by the electron filling in the $t_{2g}\downarrow$ band in the region Ⅱ.[10] As for the maximum MR existing at 300 K, which is different from ref. 9, is understandable when considering the difference of band diagram between LHMO and $La_{0.9}Sr_{0.1}MnO_3$. For the LHMO/STON junction, with the increased temperature, the electron filling of the $t_{2g}\downarrow$ band in the region Ⅱ increases, as well as the positive MR in the inset of Fig. 3. However even when the temperature reach 300 K which is our maximum testing temperature (room temperature), the filling of electrons in region Ⅱ still could not reach the band edge of $e_g^2\uparrow$ and the minority channel of $t_{2g}\downarrow$ still dominates the transport, as shown in Fig. 2(c). When a magnetic field is applied to the heterojunction, the spin polarization of $e_g^1\uparrow$ band and $t_{2g}\downarrow$ band are both increased, thus less and less carriers can tunneling between them due to the scattering between carriers with antiparallel spins, which means larger and larger resistance, and positive MR are caused in the heterojunction.

The V_b as function of temperature for LHMO/STON heterojunction measured under the fields of H = 0 and 5 T are presented in Fig. 4. The temperature dependence of the MR ratio (I = –1 mA) is presented in the inset of Fig. 4. It is notable that the MR undergo a crossover from positive to negative, and to positive again when the temperature rising from 60 K to 300 K. When a reverse bias is applied to the junction, the number of electrons filled in the $t_{2g}\downarrow$ band in the region Ⅱ increases, as shown in Fig. 2(d). As we mentioned before, at low temperature (60 K), there are electrons filling of the $t_{2g}\downarrow$ band in the region Ⅱ which caused the positive MR in the junction. When the temperature increases (100 K – 200 K), the electrons could start to fill the $e_g^2\uparrow$ band in region Ⅱ and the majority channel dominates the transport, which causes the negative MR in the junction, as shown in Fig. 2(e). When we keep increasing the temperature (220 K – 300 K), as presented in Fig. 2(f), the $t_{2g}\downarrow$ band is almost full filled while the $e_g^2\uparrow$ band is half filled. At this condition, the minority channel dominates the transport again, as well as the positive MR.

Based on our experiment results and explanations, it is obvious that the filling of electrons in the e_g and t_{2g} band, as well as the sign of MR, are highly affected by the temperature. Furthermore, it is possible to make the temperature at which the maximum MR happens near to the room temperature by precise picking the manganites and STON with suitable doping lever.

IV. CONCLUSION

A LHMO/STON heterojunction, which shows excellent rectifying properties in a wide temperature range from 20 to 300 K, has been fabricated successfully. The sign of the MR as function of temperature at either forward or reverse bias voltage is extensively studied by the filling of electrons in the e_g and t_{2g} band. Our results reveal a possibility in controlling the temperature at which the maximum MR happens near the room temperature by precise picking the manganites and STON with suitable doping lever, which is of special importance in practical applications.

V. ACKNOWLEDGEMENTS

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Fig. 1. I-V curves of LHMO/STON heterojunction measured at selected temperature (60 K, 120 K, 180 K, 240 K and 300 K) under the fields of H=0 (solid symbol) and 5T (hollow symbol). The left top shows a schematic illustration of the present heterojunction.

Fig. 2. The schematic DOS of the LHMO/STON heterojunction at zero bias voltage (a), forward bias voltage (b-c) and reverse bias voltage (d-f), respectively. The gray areas mark the states being occupied by electrons.

Fig. 3. Diffusion voltage (V_d) as function of temperature for LHMO/STON heterojunction under the fields of H=0 and 5 T. The inset shows temperature dependence of MR (I = 1 mA).

Fig. 4. Breakdown voltage (V_b) as function of temperature for LHMO/STON heterojunction under the fields of H=0 and 5 T. The inset shows temperature dependence of MR (I = –1 mA).

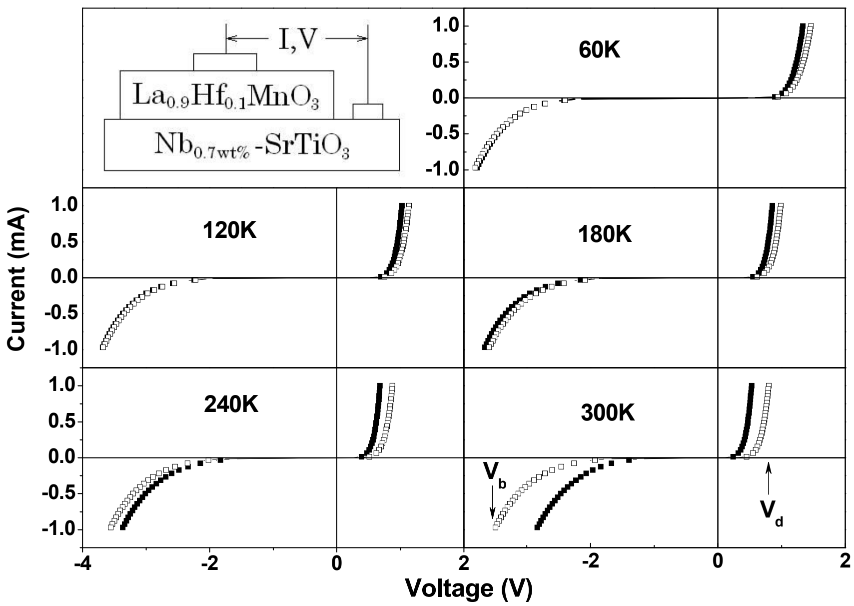
I,V
$La_{0.9}Hf_{0.1}MnO_3$
$Nb_{0.7wt\%}$-$SrTiO_3$
60K
120K
180K
240K
300K
V_b
V_d
Current (mA)
Voltage (V)

Fig. 1.

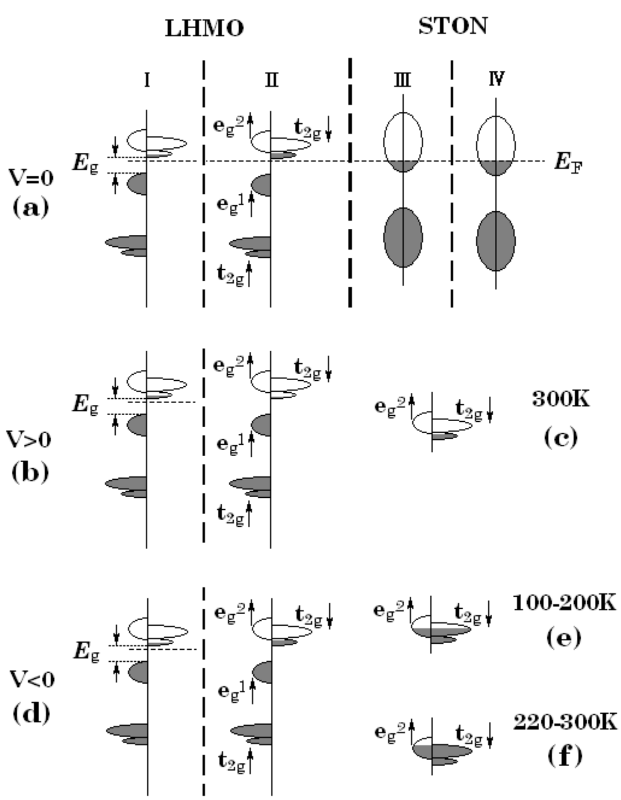
LHMO
STON
I
II
III
IV
V=0
(a)
V>0
(b)
V<0
(d)
E_g
E_F
e_g^2↑
t_{2g}↓
e_g^1↑
t_{2g}↑
300K
(c)
100-200K
(e)
220-300K
(f)

Fig. 2.

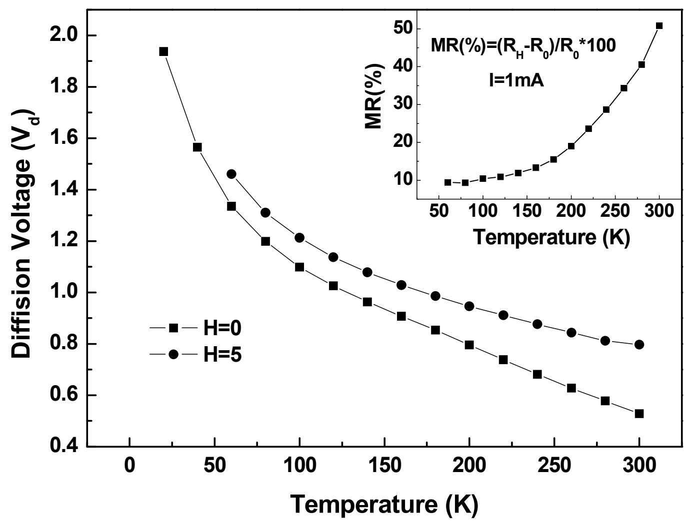
Diffision Voltage (V_d)
Temperature (K)
H=0
H=5
MR(%)=(R_H-R_0)/R_0*100
I=1mA
MR(%)
Temperature (K)

Fig. 3.

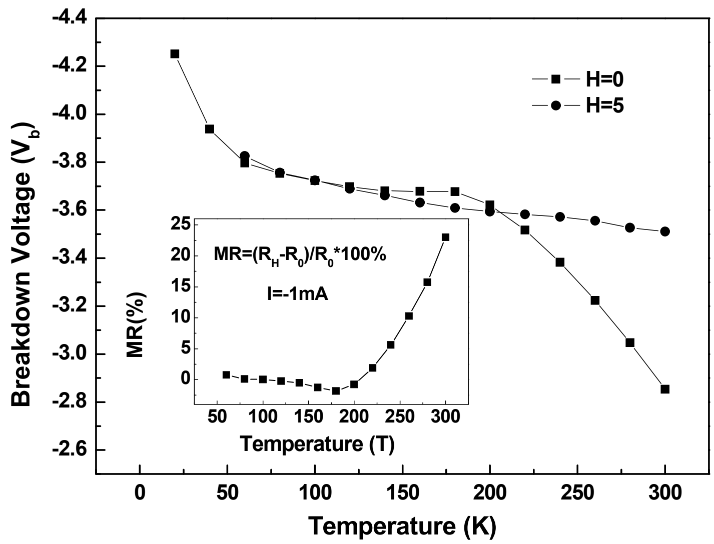
Breakdown Voltage (V_b)
Temperature (K)
H=0
H=5
MR=(R_H-R_0)/R_0*100%
I=-1mA
MR(%)
Temperature (T)

Fig. 4.