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## Positive magnetoresistance in heterostructure composed of two oxides

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## Abstract

A positive magnetoresistance (MR) has been discovered in the epitaxial p–n heterostructure we fabricated with Sr-doped LaMnO<sub>3</sub> and Nb-doped SrTiO<sub>3</sub> by laser molecular-beam epitaxy. The MR dependence on the bias voltage has been displayed at the temperature of 130 and 190 K. The mechanism causing the unusual positive MR is proposed as the creation of the region near the interface with electron filling in the  $t_{2g}$  spin-down band in La<sub>0.9</sub>Sr<sub>0.1</sub>MnO<sub>3</sub>. Other puzzling MR features with bias voltage, temperature are well explained by the present scenario. © 2005 Elsevier Ltd. All rights reserved.

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Hole doped manganese oxides of general formula  $La_{1-x}B_{x}MnO_{3}$  (B=Ca, Sr, Ba, Pb) have remarkable interrelated structural, magnetic and transport properties. In particular, they exhibit very large negative magnetoresistance (MR), called colossal magnetoresistance (CMR) which value is defined as MR =  $(R_H - R_0)/R_0$  with  $R_H$ denoting the resistance under applied magnetic field H and  $R_0$  being the resistance without magnetic field. The understanding of the microscopic physics underlying the CMR properties is important and fundamental. Good results of negative MR have been reported in some magnetic tunnel junctions (MTJ) [1-4] Recently, positive MR has been found in MTJ structures [5,6] and in Oxide semiconductor p-n junction we fabricated by Laser molecular-beam epitaxy (laser MBE) [7]. It is understandable for certain MTJ to be with a positive MR property, if the tunnelling occurs between two magnetic materials with different spin carriers. However, it is very puzzling and even seems incredible for a system with the structure consisting of a nonmagnetic material (SNTO) and a negative CMR material (LSMO) to arise a large positive MR property.

The physics origin causing this abnormal phenomenon is proposed as the creation of the space-charge region where  $t_{2g}$  spin-down ( $t_{2g} \downarrow$ ) band is partially filled by electrons in LSMO. According to this scenario, the MR dependence on the bias voltage and the temperature can also be explained qualitatively.

In order to fabricate a better oxide p-n interface, a computer-controlled laser molecularbeam epitaxy (laser MBE) [8] was used to deposit the LSMO/SNTO p-n junctions. The p-n junction was made by depositing 0.1 Srdoped LaMnO<sub>3</sub> with the thickness of 4000 Å directly on 0.01 Nb-doped  $SrTiO_3$  (001) as shown in the inset of Fig. 1. An in situ reflection high-energy electron diffraction (RHEED) provided useful information on surface structure, morphology and growth mode, and the oscillation of RHEED intensity was used to control the exact number of the deposited molecular layers. Our XRD  $\theta - 2\theta$  scan curve of the LSMO/SNTO p-n heterostructure shows that there exist only LSMO (001) and SNTO (001) peaks without any trace for other diffraction peak from impurity phase or randomly oriented grain, which means that the thin films of heterostructure are in single phase with *c*-axis orientation.

The measurement was taken by a constant current with a step of 0.01 mA. The I–V behaviors of the LSMO/SNTO p–n junction in magnetic field was measured at the temperature of 130 and 190 K by a superconducting quantum interference device (SQUID, Quantum Design

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Fig. 1. The I–V curves of  $La_{0.9}Sr_{0.1}MnO_3/SrNb_{0.01}Ti_{0.99}O_3$  p–n junction without applied magnetic field at various temperatures. Inset: schematic illustration of the p–n junction.

MPMS 5.5T). A magnetic field perpendicular to the p–n interface was applied. Fig. 2 shows the dependence of the positive MR as a function of applied magnetic field at various reverse bias voltages and at the temperature of 130 and 190 K for LSMO/SNTO p–n junction. The character of increased MR with the increased magnetic field in Fig. 2 clearly demonstrates a positive MR feature of this structure. In Fig. 2 we can see that the positive MR decreases with the absolute value of the reverse bias voltage at both temperatures of 130 and 190 K. Also we find that the MR values at 190 K is much larger that that at 130 K.

In order to establish our model to interpret the origin of the intriguing positive MR phenomenon, we first present, some useful information about the heterojunction and its SNTO substrate, as shown in Fig. 3. From the *T*-dependent resistance curve in Fig. 3(a), it can be found that the LSMO/ SNTO heterojunction exhibits the semiconductor behavior, similar to the property of LSMO in its phase diagram. On the other hand, the curve of resistance vs. temperature in



Fig. 2. The variation of MR values at 130 and 190 K of the system with the applied magnetic field at various values of bias voltage.



Fig. 3. The curves of resistance vs. temperature for LSMO/SNTO heterojunction (a) and SNTO substrate (b), respectively.

Fig. 3(b) indicates that the SNTO substrate displays metallic characteristic, suggesting the SNTO can be regarded as a metallic-like substrate with the Fermi level locating above its bottom of the conduction band.

On the base of the above analysis, we present our model based on the band structures of LSMO and SNTO to understand the physics inducing the abnormal positive CMR property of this p-n junction structure consisting of a nonmagnetic material (SNTO) and a negative CMR material (LSMO). If SNTO is connected with LSMO in an applied magnetic field, the electrons from *n* type SNTO will leak out into the adjacent p type LSMO, even partially fill the  $t_{2g} \downarrow$  band after filling up the  $e_g^1$  spin-up  $(e_g^1 \uparrow)$  band of LSMO near the interface, then the Diffusion Barrier will be built up around the interface to stop the further defusing of electrons. On the LSMO side close to the interface, a spacecharge region where the electron density of states (DOS) is larger than that of the homogeneous regions of LSMO is created, as shown in Fig. 4. Therefore, the  $t_{2g} \downarrow$  band edge is closer to the Fermi level in the space-charge region near the interface than that in the homogeneous region far from the interface. The electrons in  $t_{2g} \downarrow$  band where the magnetization is anti-parallel to the spin are minority spin carriers, and the electrons (or holes) in  $e_g^1 \uparrow$  and  $e_g^2 \uparrow$  band where the magnetization is parallel to the spin are majority spin carriers. The existence of minority spin carriers in the hole doped compound La<sub>0.7</sub>Sr<sub>0.3</sub>MnO<sub>3</sub> and in MTJ system has been proposed [5,9]. The same as previous work [5], weak Hund's rule coupling (the splitting of  $t_{2g} \uparrow$  and  $t_{2g} \downarrow$ being smaller than the sum of the crystal field splitting energy between  $e_{g}$  and  $t_{2g}$  bands and Jahn-Teller splitting energy between two  $e_g$  bands) is proposed here as



Fig. 4. The schematic DOS of the p–n junction (a), and the corresponding band diagram for each region (b).

a hypothesis on which the existing of the minority spin carriers in the system is founded. For a system with a positive MR, the  $t_{2g} \downarrow$  band of the space-charge region has to be filled by some electrons.

In order to show different regions in the structure in an applied magnetic filed, we plot Fig. 4 schematically for the system in equilibrium. Region I denotes the LSMO homogeneous region far away from the interface, region II denotes the LSMO space-charge region close to the interface, region III is the SNTO space-charge region close to the interface, and region IV is the SNTO homogeneous region far away from the interface. The DOS is schematically shown for each region in Fig. 4(a), and the corresponding band diagram for each region is schematically plotted in Fig. 4(b). In Fig. 4,  $E_F$  denoting the Fermi level in the system without bias voltage,  $E_g$  denoting the band gap between  $t_{2g} \downarrow$  and  $e_g^1 \uparrow$ , and  $\Delta E$  being the energy deference between  $e_g^2 \uparrow$  band edge and  $t_{2g} \downarrow$  band edge in LSMO.  $E_g$  is smaller than the band gap between the two bands of  $e_g \uparrow (e_g^1 \uparrow \text{ and } e_g^2 \uparrow) (\approx 1.0 \text{ eV})$  due to the weak Hund's rule coupling and the Fermi level locates slightly above the valence band [5,10-12]. Let's reasonably assume  $E_{\rm g}$  being in the range of 0.5–1.0 eV. The Fermi level of homogeneous SNTO locats slightly above the bottom of Ti 3d conduction band, and the band gap of SNTO is much narrow than that of SrTiO<sub>3</sub> (3.2 eV) [13-17] due to the Nb doping, which is in consistent with the metallic behavior of SNTO resulted from our resistance vs. temperature measurement. The MR across such a p-n junction of the ferromagnetic and a nonmagnetic compound depends on the relative spin orientation of electrons around the Fermi level in each region where the spin polarized carriers pass trough.

Now we focus on the positive MR dependence on the applied reverse (negative) bias V under which the Fermi level of region I (p-side) is raised with respect to that of region IV (n-side). With enough large negative bias voltage, electrons in region I can tunnel into region II. We can approximately present the current decreasing part  $\Delta I_+$ 

(causing the positive MR) with the magnetic field H and the current increasing part  $\Delta I_{-}$  (causing the negative MR) with H in the following way,

$$\Delta I_{+} = I_{+}^{0} - I_{+}^{H} \propto \text{DOS}_{\text{I}}(E_{e^{1}\uparrow}) \text{DOS}_{\text{II}}(E_{t\downarrow}), \tag{1}$$

$$\Delta I_{-} = I_{-}^{H} - I_{-}^{0} \propto \text{DOS}_{\text{I}}(E_{e^{1}\uparrow}) \text{DOS}_{\text{II}}(E_{e^{2}\uparrow}), \qquad (2)$$

where  $\text{DOS}_{\text{I}}(E_{e^{1}\uparrow})$  denotes the DOS at the electron filling level of  $e_g^1 \uparrow$  band in region I,  $\text{DOS}_{II}(E_{e^2\uparrow})$  denotes the DOS of carries involved in the current in  $e_g^2 \uparrow$  band in region II, and  $DOS_{II}(E_{t\downarrow})$  denotes the DOS of carries involved in the current in  $t_{2g} \downarrow$  band in region II under the bias V. Only if  $\Delta I_+ > \Delta I_-$ , the system can show a positive MR property. From above equations, it can be clearly seen that the electron filling in  $t_{2g} \downarrow$  band in region II and the tunneling process are the origin causing the positive MR, and the competition between two sources of currents (Eqs. (1) and (2)) plays a crucial role in the MR evolution with various measuring conditions of the system. At the bias |V| much smaller than  $E_{\rm g}$ , the electrons in the region I hardly tunnel the barrier  $E_{g}$  to flow to the region II, thus almost no current flow in the system. This corresponds to what we observed at small negative bias voltage in Fig. 1. With |V| being larger than a certain value, the Fermi level of region I shifted up enough to reach the bottom of  $e_g^2 \uparrow$  band of region II, tunneling current occurs, so that both the majority channel of  $e_g^2 \uparrow$  and the minority channel of  $t_{2g} \downarrow$  in region starts to be available for transport. The scattering of spin up electrons in  $e_g^1 \uparrow$  of region I with spin down electrons in  $t_{2g} \downarrow$  of II increases with the applied magnetic field, which causes the positive MR in the system. The increasing of the reverse bias voltage increases the Fermi level in region I, and thus makes more and more electrons filling of  $e_g^2 \uparrow$  in region II and being involved in the transport, which causes the MR value starts to decrease with the increasing of |V|. This behavior of MR depending on the bias voltage agrees well with the phenomenon shown in Fig. 2.

The temperature dependence of the positive MR in Fig. 2 can also be understood by this scenario as the following. With the increased temperature, the electron filling of the  $t_{2g}$   $\downarrow$  band in region II increases, as well as the positive MR in the system.

To obtain a quantitatively explanation, a detailed calculation of the band structure and the temperature evolution of the band in LSMO would be expected. In the realistic system, the four regions are not clearly divided as what are shown in Fig. 4, but merge one to the next smoothly from region I to IV. Thus sharp interfaces between different regions do not exist, but the origin of the MR is in principle the same as we have presented above.

In summary, positive MR properties of LSMO/SNTO p-n junction have been reported and the origin of the puzzling phenomena has been proposed by the electron filling in the spin-down band in the space-charge region of LSMO close to the interface and the tunneling process in

different region of LSMO. Meanwhile, spin-up  $(e_g^2 \uparrow)$  carriers of region II in transport play a crucial role in the MR evolution at various measuring conditions. We believe the results and their physics origin we present in this letter are important not only from a practical viewpoint, but also as a potential of a new insight into the microscopic physics of the p–n junction consisting of the ferromagnetic and nonmagnetic materials.

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