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TRANSPORT PROPERTIES ON $La_{0.7}Sr_{0.3}MnO_3/Al_2O_3/Fe$ TUNNEL JUNCTIONS*

PAN Feng-ming (潘风明)[†], JIAO Zheng-kuan(焦正宽)

(Physics Department, Yuquan Campus of Zhejiang University, Hangzhou 310027, China)

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[†] E-mail: f.m.pan@fz-juelich.de

Abstract: Manganese oxide $La_{0.7}Sr_{0.3}MnO_3$ produced on (001) $LaAlO_3$ substrate by means of sol-gel spin-coating method was used as the base electrode of $LaSrMnO/Al_2O_3/Fe$ tunnel junctions. The I-V characteristic in the high bias region of this system was shown to be similar to that of the conventional tunnel junctions. Anomalous temperature dependence of tunneling resistance was observed to be a positive temperature coefficient of resistance. This phenomenon was attributed to the high voltage applied and was simply elucidated from the density states vs. energy diagram.

Key words: tunnel junctions, manganese oxide, transport properties
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INTRODUCTION

Much attention has been paid recently to the study of spin polarized tunneling in junctions of two ferromagnets separated by a non-magnetic tunneling barrier(Yaoi et al., 1993; Nowak et al., 1992; Suezawa et al., 1992; LeClair et al., 1994; Plaskett et al., 1994; Miyazaki et al., 1995; Tezuka et al., 1996). A significant merit of magnetic tunnels for applications is that the magnetic field needed to saturate the tunnel magnetoresistance (TMR) is considerably low, only tens of oersteds. The TMR effect, however, is not easily observable, especially at room temperature. A value of up to 24% at 4.2K and 11.8% at room temperature was obtained for $CoFe/Al_2O_3/Co$ junctions by Moodera et. al. (1995)

Magnetic manganese oxides with perovskite-type crystal structure have attracted much interest due to the very high magnetoresistance (MR) effect found in them(Jin et al., 1994). On the other hand, these oxides have nearly 100% polarizations of electron spin. This feature makes them qualified candidates as electrodes in tunnel junctions. According to Julliere's analysis (Julliere, 1975) based on the spin density of states at the Fermi level of the two ferromagnetic (FM) electrodes, the TMR can be described by the equation $TMR = 2P_1P_2/(1 + P_1P_2)$, where P_1 and P_2 are the spin polarizations of the two FM electrodes respectively. Assuming that there is no spin flip-flop of the polarized electrons when they are penetrating through the insulator, Julliere's model indicates that larger spin polarizations of FM electrodes may give rise to larger TMR. Stimulated by the above mentioned properties of tunnel junctions and manganese oxides, we produced $La_{0.7}Sr_{0.3}MnO_3/Al_2O_3/Fe$ tunnel junctions and in the present paper will discuss some transport properties of these manganese oxide tunnels.

SAMPLE PREPARATION

The manganese oxide $La_{0.7}Sr_{0.3}MnO_3$ (LaSrMnO) electrode was fabricated on (001) $LaAlO_3$ single crystal substrate by the sol-gel spin-coating method. The stock solution was prepared by mixing naphthenate of lanthanum, strontium and manganese in toluene solvent at a molar ratio of La : Sr : Mn = 0.7 : 0.3 : 1. The thickness of the manganese oxide film was about 1400Å. The crystal orientation was determined by x-ray diffraction measurements and experimental results

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showed that the LaSrMnO films had their *c*-axis oriented perpendicular to the film surface. *Φ*-scan revealed the complete epitaxial growth of the films. Magnetron sputtering through a rectangular mask was used to deposit an about 50 angstrom thick, aluminum film on the LaSrMnO base. The sputtering pressure was kept to ≃ 1 Pa of argon gas. Then the sample was taken out from the vacuum chamber and the aluminum was oxidized in air at room temperature for 24 hours. Finally an Fe counter-electrode was sputtered through a rectangular mask on the Al_2O_3 insulator film. A schematic representation of the tunnel junctions is given in Fig. 1. The purity of Al and Fe used was 99.99% and 99.9%, respectively.

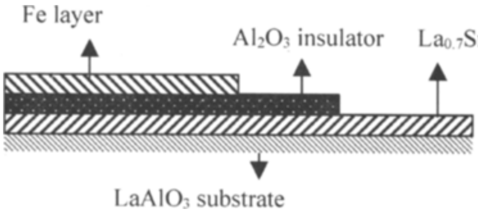

Fig. 1 Schematic representation of the tunnel junction structure for $La_{0.7}Sr_{0.3}MnO_3/Al_2O_3/Fe$ samples.

EXPERIMENTAL RESULTS

The I-V characteristic is usually used to verify the quality of a tunnel junction and to prove that tunneling is the main source of the conductance. Fig. 2 shows the voltage dependence of tunneling current of a $LaSrMnO/Al_2O_3/Fe$ junction at both low temperature and room temperature. When the bias applied was lower than 300 mV, the tunneling current was too small to be distinguished. This behavior is expected for a good tunnel junction and can be qualitatively understood in the concept of states density of electron energy, which will be discussed later. At high bias, the current increased monotonously with variations of applied voltage but deviated from its extrapolate values at higher bias and showed lower percentage change. For the manganese oxide junctions in the present study, the variation behavior of tunneling current vs bias was different from that of junctions with two metallic FM electrodes where a parabolic dependence was observed in the I-V plot. At low temperature and with bias less than 10 V, the percentage increase of current with variations of voltage was much higher than that at room temperature. This behavior is similar to the conductance dip observed at low temperature in previous reports(Moodera et al., 1995; 1996). Several causative factors have been considered for such anomalies, including localization effect in amorphous materials, magnetic scattering in the interface or in the barrier, and multistep tunneling through an intermediate state.

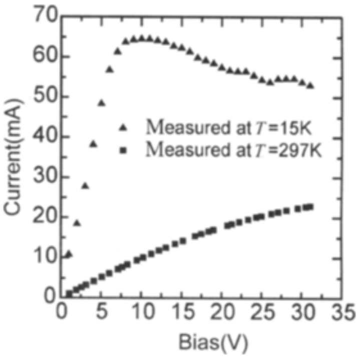

Fig. 2 The tunneling current vs. applied voltage for $La_{0.7}Sr_{0.3}MnO_3/Al_2O_3/Fe$ junction at $T = 15K$ and 297K respectively.

Fig. 3 shows the temperature dependence of junction resistance (R_j) for $LaSrMnO/Al_2O_3/Fe$. In our experiments a constant voltage of 10V was applied. Contrary to theoretical predictions and previous reported results, the tunnel resistance exhibited metallic behavior and R_j increased with temperature. According to Simmon's and Stratton's tunneling theory(Simmons, 1964;

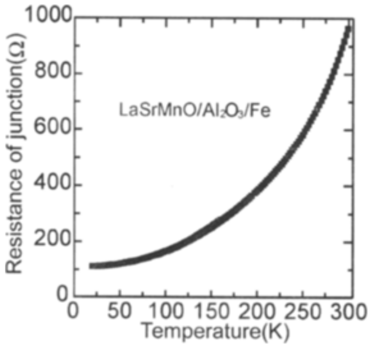

Fig. 3 Temperature dependence of junction esistance for $La_{0.7}Sr_{0.3}MnO_3/Al_2O_3/Fe$ samples. A constant voltage of 10V was applied in measurement.

Stratton, 1962), the tunnel current varies linearly with T^2. We attribute this anomaly to the high bias applied to samples and simply interpret it in terms of the density of states vs. energy diagram, as shown in Fig. 4. To describe tunneling of electrons in the manganese oxide junctions, the "semiconductor picture", i. e., the band structure of semiconductor, is used. When no voltage is applied or the voltage applied is less than Δ/e with 2Δ the energy gap of semiconductor, electrons from metal electrode (A) cannot tunnel into the semiconductor (B) because of the energy gap (2Δ) in the electron states. When a bias is applied to raise the potential of A relative to B by an amount eV which is just greater than Δ, a few electrons from A can tunnel into the density of electron states in B. If the potential difference eV is larger than the barrier height h, the barrier becomes very distorted in shape, as shown in Fig. 4, and some electrons actually tunnel into allowed states in the insulator, rather than into B. In such a situation, the barrier is lost, to some extent. In other words, the conductance should be considered to be the contribution of total combinations of junctions, but not only the tunneling effect from electrode A to B. The R-T curve of LaSrMnO on $LaAlO_3$ substrate was measured and metallic behavior of temperature dependence of the resistance was found to be below the room temperature. As a result, the junction resistance vs. temperature curve becomes like that of metals. It should be noted that the simple diagram of Fig. 4 is for $T = 0K$ and the real situation is complex. In the present study it is just a schematic representation of the theoretical analysis.

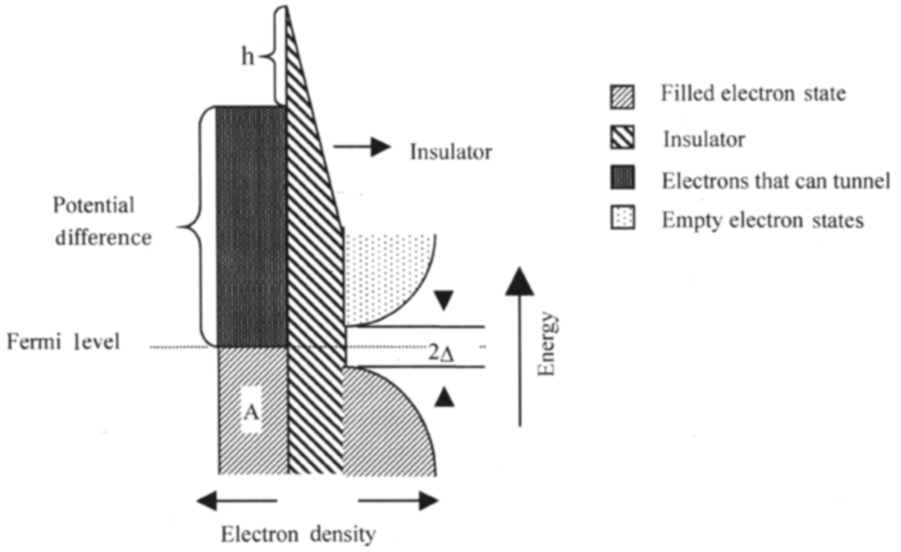

Fig.4 Electron density of states vs. energy diagram for a semiconductor-insulator-metal junction, with A of normal metal and B of semiconductor.

According to previous reports that a high bias may significantly lower the tunneling magnetoresistance (Moodera et al., 1995; 1996), we should try our best to lower the bias needed to produce the tunneling in manganese oxide junctions, such as $LaSrMnO/Al_2O_3/Fe$ in the present study, in order to take advantages of both magnetic tunnels and manganese oxides.

CONCLUSIONS

In summary, we have produced tunnel junctions with one electrode of manganese oxide $La_{0.7}Sr_{0.3}MnO_3$ and have investigated their transport properties. The I-V characteristic showed tunneling behavior but the bias needed to produce tunneling was higher than that of junctions with two electrodes of normal FM metals. This feature may affect the tunneling magnetoresistance effect. Due to the high bias applied, the tunneling effect covered up the temperature dependence of resistance and a metallic R-T curve was found. This phenomenon can be roughly interpreted using diagrams of density of states vs. energy.

References

Jin, S., Tiefel, T. H., McCormack, M., et al., 1994. Thousandfold change in resistivity in magnetoresistive La-Ca-Mn-O Films. *Science* **264**: 413

Julliere, M., 1975. Tunneling between ferromagnetic films. *hys. Lett.* **54A**: 225

LeClair, P., Moodera, J. S. and Meservey, R., 1994. Ferromagnetic-ferromagnetic tunneling and the spin filter effect. *J. Appl. Phys.* **76**: 6546

Miyazaki, T. and Tezuka, N., 1995. Spin polarized tunneling in FM/I/FM junctions. *J. Magn. Magn. Mater.* **151**: 403

Moodera, J. S., Kinder, L. R., Wong, T. M. et al., 1995. Large magnetoresistance at room temperature in ferromagnetic thin film tunnel junctions. *Phys. Rev. Lett.* **74**: 3273

Moodera, J.S. and Kinder, L.R., 1996. FM-I-FM tunneling: Spin dependent tunneling and large magnetoresistance in trilayer Junctions. *J. Appl. Phys.* **79**: 4725

Nowak, J. and Rauluszkiewicz, J., 1992. Spin dependent electron tunneling between Ferromagnetic Films. *J. Magn. Magn. Mater.* **109**:79

Plaskett, T. S., Freitas, P. P., Barradas, N. P., et al. 1994. Magnetoresistance and magnetic properties of NiFe/oxide/Co junctions prepared by magnetron sputtering. *J. Appl. Phys.* **76**: 6104

Simmons, J.G., 1964. Generalized thermal J-V characteristic for the electric tunnel effect. *J. Appl. Phys.* **35**: 2655

Stratton, R., 1962. Voltage-Current characteristics for tunneling through insulating films. *J. Phys. Chem. Solids*, **23**: 1177

Suezawa, Y., Takahashi, F. and Gondo, Y., 1992. Spin-polarized Tunneling in Ni/Al_2O_3/Co Junctions and Large Magnetoresistance of Ni/Co Double Layers. *Jpn. J. Appl. Phys.* **31**: L1415

Tezuka, N. and Miyazaki, T., 1996. Magnetic tunneling effect in Fe/Al_2O_3/Ni_{1-x}Fex Junctions. *J. Appl. Phys.* **79**: 6262

Yaoi, T., Ishio, S., and Miyazaki, T., 1993. Dependence of magnetoresistance on temperature and Applied voltage in 82Ni-Fe/Al-Al_2O_3/Co tunneling junctions. *J. Magn. Magn. Mater.* **126**: 430