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Negative spin polarization in $(La,Sr)CoO_3$ probed by a magnetic tunnel junction with $(La,Sr)MnO_3$

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By using perovskite $(La,Sr)MnO_3$ (LSMO)-based magnetic tunnel junctions with $LaAlO_3$ barrier, we have experimentally revealed the spin state of $(La,Sr)CoO_3$ (LSCO) that was employed as the counterelectrode of the junctions. Inverse tunnel magnetoresistance (TMR) up to −14% was observed at 10 K in low-bias measurement. This indicates that $t_{2g\downarrow}$ electrons are the majority at the Fermi level in the LSCO band structure, taking into account the half-metallic character of LSMO. The temperature dependence of the interfacial spin polarization of LSCO as deduced from the TMR ratio indicates the reduction of the Curie temperature at the interface by 70 K. The TMR ratio exhibits an asymmetric bias dependence in qualitative agreement with the calculated spin band structure of LSCO. © *2007 American Institute of Physics*. [DOI: 10.1063/1.2806964]

The magnetic tunnel junction (MTJ) with half-metallic, i.e., fully spin polarized, materials is one of the desired electronic device elements in future spintronics. The high spin polarization of the ferromagnetic electrodes enhances the tunnel magnetoresistance (TMR) that is defined as $(R_{AP}-R_P)/R_P$, where $R_{AP(P)}$ is the junction resistance when the magnetizations of two electrodes have the antiparallel (parallel) configuration.[1–3] A half-metallic (positively spin polarized at the Fermi level E_F) perovskite oxide, $(La,Sr)MnO_3$ (LSMO), has been intensively studied for MTJ.[2,4–6] We have designed and optimized the LSMO/barrier interface ferromagnetism based on the interface engineering approach[7] and improved the fabrication processes of the junctions. As a result, reproducible and reliable fabrication technology has been established for LSMO/$LaAlO_3$ (LAO)/LSMO junctions with a TMR ratio as high as 200% at 10 K, which corresponds to an interface spin polarization of 70%,[8] according to Julliere's model.[9] By using MTJs with highly spin-polarized base electrode, the spin density of state (DOS) can be probed for an unknown ferromagnet used as a counterelectrode. De Teresa *et al.* reported on the spin-polarized tunnel junctions composed of LSMO and a ferromagnetic transition metal Co with a $SrTiO_3$ barrier.[10,11] They found that the junctions exhibited the inverse TMR responses, that is, $R_{AP}>R_P$, in low-bias measurement and showed that the bias dependence of TMR reflected the spin DOS structure of the Co-3*d* band. Hu and Suzuki reported the inverse TMR for all-oxide LSMO/$CoCr_2O_4$/Fe_3O_4 junctions.[12] This inverse TMR is in accord with the theoretical prediction that Fe_3O_4 is negatively spin polarized at E_F, although the TMR ratio of −25% was not so large as that expected for half-metallic Fe_3O_4. More recently, we have demonstrated the inverse TMR response for all-perovskite $SrRuO_3$/$SrTiO_3$/LSMO junctions, indicating a negative spin polarization in $SrRuO_3$.[13]

In this paper, we present the spin polarization of $La_{0.5}Sr_{0.5}CoO_3$ (LSCO) analyzed by the LSMO-based MTJs with LAO barriers. For perovskite Co oxides, various spin configurations are possible to be considered. For example, $LaCoO_3$ has three possible spin states: a low spin state $(t_{2g}^6, S=0)$, an intermediate spin state $(t_{2g}^5e_g^1, S=1)$, and a high spin state $(t_{2g}^4e_g^2, S=2)$. Experimentally, it is confirmed that $LaCoO_3$ is in the low spin state at low temperatures (<100 K) and eventually undergoes the crossover transition to the intermediate spin state at higher temperatures.[14] By substituting of Sr^{2+} for La^{3+} in $LaCoO_3$, the ground state changes from nonmagnetic insulator to ferromagnetic metal. The spin state for $(La,Sr)CoO_3$ is investigated by x-ray and ultraviolet photoemissions, x-ray absorption spectroscopy, and so on.[15,16] The spin band structure in $(La,Sr)CoO_3$ has been investigated by x-ray magnetic circular dichroism study[17] and first-principles calculation.[18–20] As far as we know, however, there is no report on the spin polarization in $(La,Sr)CoO_3$ by means of the spin-transport technique.

Trilayer epitaxial films consisting of bottom $La_{0.6}Sr_{0.4}MnO_3$ (50 nm)/barrier LAO (1.5 nm)/top LSCO (50 nm) layers were grown on $(LaAlO_3)_{0.3}-(Sr_2AlTaO_6)_{0.7}$ substrates by a pulsed-laser deposition method. The epitaxy processes were *in situ* monitored and controlled by a reflection high-energy electron diffraction. The bottom LSMO and LAO barrier layers were deposited at an oxygen pressure P_{O_2} of 1 mTorr and at a substrate temperature T_s of 870 °C. The top LSCO layer was then grown under the condition of $P_{O_2}=0.8$ mTorr and $T_s=680$ °C. The atomically flat surfaces of the bottom electrode and the barrier layer are very important to ensure the uniform transport in a junction. An atomic force microscopy measurement on separate films revealed

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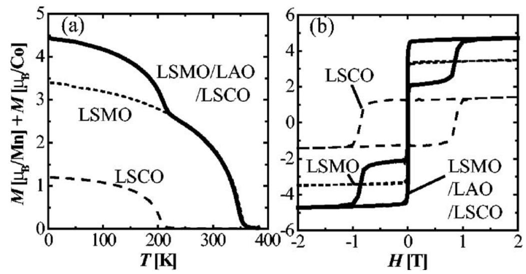

FIG. 1. (a) Temperature dependence of magnetization M during cooling in a magnetic field H of 50 mT and (b) M-H curve hysteresis measured at 5 K for an unpatterned LSMO (50 nm)/LAO (1.5 nm)/LSCO (50 nm) trilayer film. M is normalized by the LSMO and LSCO layer thicknesses. The results for LSMO and LSCO single layer films are also shown by broken lines for comparison.

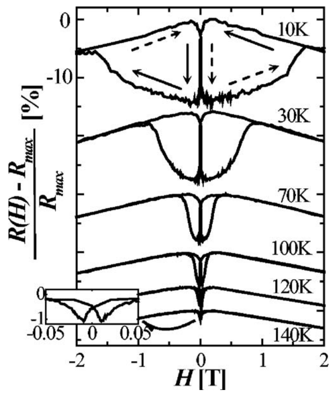

FIG. 2. Magnetic field dependence of measured junction resistance $R(H)$ as a function of the temperature in a low-bias measurement for a LSMO/LAO/LSCO junction. The junction resistance is normalized as $[R(H) - R_{\max}]/R_{\max}$, where $R_{\max}$ is the maximum value of $R(H)$ during the field scan, corresponding to R_P. The traces are shifted vertically for clarity.

that LSMO/LAO layers were grown with an atomically flat surface keeping steps of one unit-cell height and wide terraces, whereas the LSCO film showed a rather rough surface with precipitates. After the deposition of the trilayer film, a protection layer of Au film (50 nm) was *ex situ* deposited to prevent the chemical reaction during the fabrication process of the MTJs. The formations of the junction electrode and the junction isolation were carried out by using the conventional photolithography and Ar ion milling. A self-aligning lithographic process was used for the definition of the junction area and the formation of SiO_2 layer (350 nm) for insulating the top and bottom electrodes. Finally, a film of Au (500 nm) was deposited to make connection to the top electrodes.

Figures 1(a) and 1(b) show the temperature dependence of the magnetization M under field cooling with a magnetic field H of 50 mT and a typical M-H curve at 5 K for an unpatterned LSMO/LAO/LSCO trilayer film, respectively. The magnetization for LSMO and LSCO single layer films is also plotted for comparison with that of the trilayer, where M is normalized by the LSMO and LSCO layer thicknesses. In all the measurements, the magnetic field was applied parallel to the film plane, which is along the magnetic easy axis for LSMO and LSCO. The two Curie temperatures T_C and coercive fields H_c observed for the trilayer junction agree with the respective T_C and H_c for LSMO and LSCO single layer films. This indicates that the bottom LSMO and top LSCO in the trilayer are magnetically decoupled and have excellent quality comparable to the single layer films.

Figure 2 shows typical TMR characteristics as a function of the temperature in a low-bias measurement for a junction with a dimension of $5 \times 10\ \mu m^2$. The junction resistance in the parallel configuration is 20.5 kΩ at 10 K, which is two orders of magnitude larger than the spreading resistance of the LSMO electrode. The junction exhibits a clear inverse TMR response up to 140 K, and the TMR ratio is about −14% at 10 K. With increasing temperature, the amplitude of the TMR response and the value of H_c are decreased. All the fabricated junctions had such an inverse TMR effect with good reproducibility. Since the conduction electrons of LSMO are almost fully spin-polarized $e_{g\uparrow}$ electrons, this inverse TMR implies that ↓ spin electrons are the majority at E_F in LSCO. The interface spin polarization P of the electrode can be deduced from the TMR ratio by Julliere's formula.[9] We have similarly fabricated LSMO/LAO/LSMO MTJs to evaluate the interface spin polarization of LSMO (P_{LSMO}) and obtained $P_{LSMO} \sim +70\%$ at 10 K.[8] From the experimental value of P_{LSMO} and the observed TMR ratio of −14%, the interface spin polarization of LSCO (P_{LSCO}) can be deduced as −11% at 10 K.

Figure 3(a) shows the temperature dependences of the TMR ratio for LSMO/LAO/LSCO (closed circles) and LSMO/LAO/LSMO (open squares) junctions. Here, each TMR ratio is taken as an average value for three typical junctions and normalized by the value at 10 K. The temperature dependences of the normalized magnetization M/M_0 for LSMO and LSCO single layer films, where M_0 is M at 10 K, are also plotted for reference. Although the reduction of interfacial T_C is observed in both of the junctions, the normalized TMR value for the LSMO/LAO/LSCO junction appears to reduce rather gradually with the increasing temperature, as compared with that for the LSMO/LAO/LSMO junction. In order to make these behaviors more clear, we plot the normalized spin polarization P/P_0 against the normalized temperature T/T_C^{TMR} in Fig. 3(b), where P_0 is P at 10 K and T_C^{TMR} is the temperature at which the TMR vanishes. The T/T_C dependence of M/M_0 is also plotted for comparison. For evaluating P_{LSCO} at each temperature, we use the experimental data of P_{LSMO} (T) shown in Fig. 3(a). Although

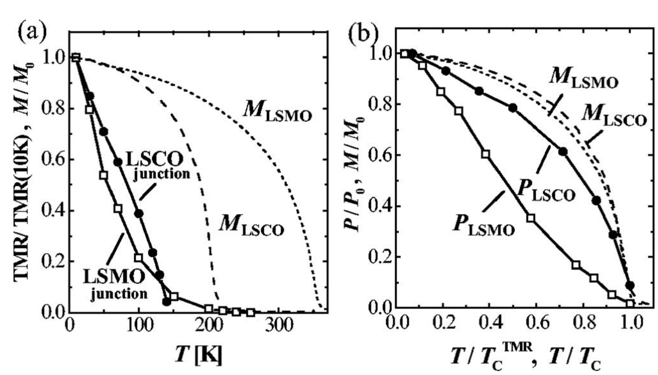

FIG. 3. (a) Temperature dependences of normalized TMR ratio (averages of typical three junctions) for LSMO/LAO/LSCO junction (●) and LSMO/LAO/LSMO junction (□). For comparison, normalized M-T curves for LSMO and LSCO single layer films are also shown by broken lines. Here, TMR ratio and M are normalized by the values at 10 K. (b) Normalized temperature (T/T_C^{TMR}) dependence of interface spin polarization P deduced from TMR for LSCO (●) and LSMO (□), where T_C^{TMR} is the temperature at which TMR vanishes. P is also normalized by the value at 10 K. The T/T_C dependences of normalized M for LSMO and LSCO, where T_C for M is the Curie temperature defined from M-T curve, are also shown.

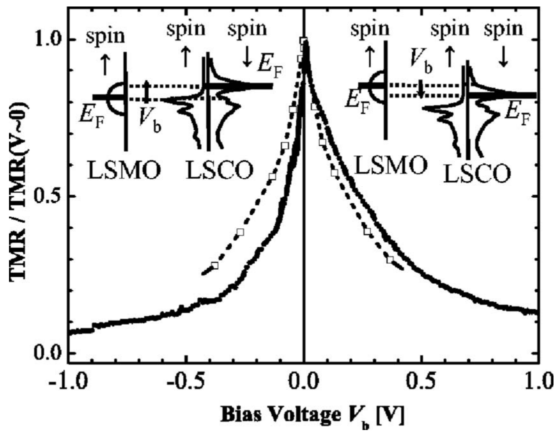

FIG. 4. Bias voltage V_b dependence of normalized TMR ratio for LSMO/LAO/LSCO junction (solid line) and LSMO/LAO/LSMO junction (broken line). The positive bias is defined as a current flow from LSCO to LSMO electrodes. The TMR ratio is normalized by the value at the low-bias ($V_b \sim 10$ mV) measurement. Inset: schematics for tunneling transport between the DOS in spin-polarized LSMO and LSCO (after Ref. 19) for $V_b \sim -1$ V (left) and $V_b \sim +1$ V (right).

P_{LSMO} is rapidly reduced with the increasing temperature, compared to M_{LSMO}, P_{LSCO} has almost the same temperature dependence as M_{LSCO}. This fact may indicate that the LSCO/LAO interface is more robust in terms of the spin polarization, compared to the LSMO/LAO one.

The bias voltage V_b dependence of the TMR ratio for the LSMO/LAO/LSCO junction at 10 K is shown by the solid line in Fig. 4, where the positive bias is defined as the current flow from LSCO to LSMO. Although the amplitude of the negative TMR response was decreased as increasing $|V_b|$, the junction did not show such a reversal of the TMR response from negative to positive at $|V_b| < 1.5$ V, as reported for the LSMO/$SrTiO_3$/Co junction.[10,11] However, the V_b dependence of the TMR ratio for the LSMO/LAO/LSCO junction exhibits an asymmetrical behavior, which is contrastive with a symmetrical V_b dependence of the LSMO/LAO/LSMO junction as denoted by the broken line in Fig. 4. The asymmetrical V_b dependence might be explained qualitatively in terms of the calculated spectrum of the spin-polarized DOS for LSCO.[19,20] For simplicity, LSMO is considered to be completely spin polarized in the following argument. Since the DOS of LSCO $t_{2g\downarrow}$ spin band at E_F is much higher than that of ↑ spin band at $V_b \sim 0$, as shown in the inset of Fig. 4, the tunneling probability for the antiparallel M configuration exceeds that for the parallel M configuration, leading to the inverse TMR response. When $|V_b|$ is increased in $V_b < 0$ region, the contribution of the tunneling process between LSMO $e_{g\uparrow}$ and LSCO $t_{2g\uparrow}$ increases rapidly (left diagram of the inset), resulting in the suppressed contribution of negatively spin-polarized LSCO electrons in the tunneling process. Thus, the negative TMR ratio is expected to drop sharply. In $V_b > 0$ region, on the other hand, since the contribution of LSCO ↑ spin band does not change so much (right diagram of the inset), the reduction of the TMR ratio in $V_b > 0$ region is expected to be more gradual, compared to that in $V_b < 0$ region.

In conclusion, we have investigated the spin state of LSCO using LSMO-based magnetic tunnel junctions with LAO barrier. These junctions show the inverse TMR response, which indicates that $t_{2g\downarrow}$ electrons are the majority at E_F in LSCO. From the TMR ratio of −14% and $P_{(LSMO)}$ deduced from LSMO/LAO/LSMO junctions, P_{LSCO} is estimated to be −11%. The temperature dependence of interfacial spin polarization of LSCO, as deduced from the TMR ratio, is similar to that of bulk magnetization in contrast to the case of LSMO. In addition, the TMR ratio exhibits asymmetric bias dependence, which can be qualitatively explained by the calculated spin DOS structure of LSCO.

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