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Breakdown by magnetic field in a $La_{0.7}Sr_{0.3}MnO_3$/MgO/Fe spin valve

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A $La_{0.7}Sr_{0.3}MnO_3$/MgO/Fe spin valve with inverse tunneling magnetoresistance (TMR) was fabricated on a (100) $SrTiO_3$ substrate by radio frequency magnetron sputtering. Giant TMR ratios up to 540% were obtained. The breakdown of the spin valve was observed at high magnetic field, which was attributed to the joint action of the invalidation of MgO barrier and the shift of Fermi energy in $La_{0.7}Sr_{0.3}MnO_3$ at high magnetic field. © *2012 American Institute of Physics*. [http://dx.doi.org/10.1063/1.3696670]

Spin valves have attracted much attention due to their applications in magnetic storage and magnetic field sensor. Magnetic tunneling junctions (MTJs) are an important branch of the spin valve family, and they possess high magnetoresistance (MR) ratio at low field. Spin source materials with high spin polarization are necessary for high-performance devices. Some ferromagnetic metals and half-metal materials, such as iron and $La_{0.7}Sr_{0.3}MnO_3$ (LSMO), have shown their satisfying capability as the spin source. Worthy of mention is that the barrier material also plays an important role in the spin valve devices. Obtained in iron based MTJs,[1,2] the spin filter effect of MgO barrier in MTJs demonstrated the tunneling MR (TMR) ratios higher than 1000%. In LSMO/MgO/$SrRuO_3$ MTJs, theoretically, it was predicted that a spin rectification effect will arise due to the symmetry-filtering properties of the MgO barrier.[3] Although the lattice mismatch is up to 8%, epitaxial LSMO layer on MgO has been realized.[4,5] The above facts suggest that high MR ratio might also be obtained in LSMO/MgO/Fe system.

However, there exist inevitably some defect levels in the bandgap of MgO due to the intrinsic defects and unintentionally doped impurities. These levels can provide channels for carriers, which will results in the degradation of the barrier effect. Especially, in the strongly correlated materials such as LSMO, the Fermi level position depends on the external magnetic field remarkably.[6] The shift of Fermi level with external field and defect levels in MgO increase the breakdown possibility of the MTJ devices. Therefore, the defects in barrier materials and the electronic state change in electrodes caused by the external conditions should be considered during designing and fabricating MTJ devices.

In this work, we fabricated a LSMO/MgO/Fe spin valve. The MR ratio up to 540% was obtained at 190 K. An inverse TMR effect with low resistant states at high field was observed, which was ascribed to the breakdown effect mentioned above.

The LSMO(150 nm)/MgO(2–3 nm)/Fe(80 nm) spin valve was fabricated on a (100) $SrTiO_3$ substrate by magnetron sputtering. To facilitate characterization, a multilayer film with the same structure was fabricated. The deposition temperature and the chamber pressure were kept at 750 °C and 6 Pa, 600 °C and 1 Pa, 500 °C and 0.5 Pa for LSMO, MgO, and Fe layers, respectively. For the growth of LSMO and MgO layers, Ar-O_2 mixtures with flow ratios 2:1 and 3:1 were used as sputtering and reaction gas, respectively. For the deposition of Fe layer, H_2-Ar mixture with a ratio of 1:5 was used as sputtering gas to provide a reducing atmosphere. The junction area is $200 \times 200\ \mu m^2$.

X-ray diffraction (XRD) with Cu Kα radiation ($\lambda = 0.154$ nm) and x-ray photoelectron spectroscopy (XPS) were used to characterize the crystal quality and electronic states. The magnetic properties were measured using a MPMS-XL superconducting quantum interference device (SQUID). The MR curves were measured using a HMS7707 Hall measurement system.

Fig. 1 shows the XRD pattern of the LSMO/MgO/Fe multilayer film on the (100) $SrTiO_3$ substrate. The diffraction intensity is shown in a logarithmic scale. Besides the $SrTiO_3$ (200) peak at $2\theta = 46.5°$, three peaks located at $2\theta = 43.1°$,

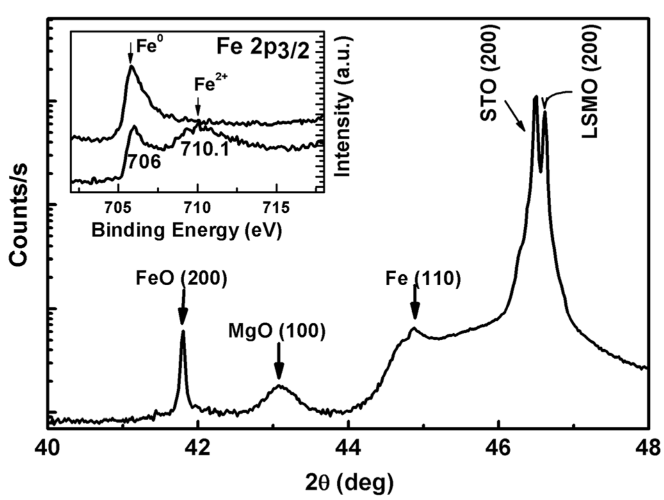

FIG. 1. The XRD pattern of the LSMO/MgO/Fe multilayer structure. The y-axis is shown logarithmically. The inset is Fe $2P_{3/2}$ XPS of the multilayer samples with (upper curve) and without (lower curve) Mg-MgO layer.

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44.7°, and 46.6° were observed, which are in accordance with MgO (100), Fe (110), and LSMO (200), respectively. The peak at 41.8° agrees well with FeO (200). The inset in Fig. 1 shows the Fe $2p_{3/2}$ XPS spectra of the multilayer films with (upper curve) and without 2–3-nm-thick Mg-MgO protective layer (lower curve). After deposited a Mg-MgO layer, the 710-eV-signals from Fe^{2+} $2p_{3/2}$ in XPS disappeared. It indicates that the FeO is formed by surface oxidation.

Fig. 2(a) sketches the MR measurement configuration. The external field was perpendicular to the film plane. The injected current is 500 nA. In early studies, it has been found that the TMR effect decreases rapidly on increasing temperatures and vanishes around T = 200 K, despite the high Curie temperature up to 330 K for $La_{0.7}Sr_{0.3}MnO_3$.[7] Therefore, the measurements were performed at 190 K, which can be obtained easily by semiconductor refrigeration. In addition, this temperature is lower than the Nell temperature of FeO (198 K). In this case, strong exchange coupling will exists at the Fe/FeO interface due to the antiferromagnetism of FeO. By M-H measurement for a Fe/FeO thin film, remarkable increase of coercive force and exchange bias field were observed with decreasing temperature (not shown here). It means a significant pinning effect below 200 K.

It had been reported that Fe/($BaTiO_3$ or $SrTiO_3$)/LSMO MTJs show an inverse TMR character due to the negative spin polarization of Fe.[8–10] Here, the LSMO-MgO-Fe MTJ should also possess an inverse TMR due to the positive and the negative polarizations in LSMO and Fe electrodes, respectively.

Figs. 2(b)–2(d) show the MR curves of the LSMO/MgO/Fe MTJ in different field ranges. The MR ratio is defined as $\Delta R/R_0$, where ΔR and R_0 are the change and minimum values of the resistance, respectively. Fig. 2(b) shows the MR curve in the field range of ±3.5 kOe. The MR loop shows an unsymmetrical shape. It is similar with the MR behavior of spin valves at low field.[11] This is because the external field is not large enough to reverse the magnetic moment of the iron layer due to the pinning effect from FeO layer.

Fig. 2(c) shows the MR curve in the field range of ±5.5 kOe. Here, the magnetic moments of both the two electrodes were reversed at the field maximum. The junction resistance varies in the range of 45–290 Ω and the corresponding MR ratio is up to 540%. However, the MR shows a certain decrease at the maximal field, which is different from common inverse TMR curves. In order to study MR variation at high field, MR curve in the field range up to ±12 kOe was measured. For a clear observation, only the MR variation with varying field from −12 kOe to 12 kOe is shown in Fig. 2(d). As observed, the MR ratio decreases abruptly to about zero as the field exceeds 7–8 kOe, just like a breakdown effect. It is distinct from a simple inverse TMR curve with high resistance states at high fields. The carrier spin in the electrodes is not able to reverse again with varying the field monotonically. Therefore, the invalidation of the barrier is the most possible reason of the low MR state.

As well known, inevitably there are some defect levels in the bandgap of MgO, which were caused by intrinsic vacancies, interstitials, and interfacial states. Their existence was supported by luminescence bands with various energies.[12] These defect levels can provide some conducting channels in the nanometer-thick MgO layer. Furthermore, the Fermi level in double exchange materials can be influenced by external magnetic field remarkably.[13,14] In $La_{0.7}Sr_{0.3}MnO_3$, the e_g orbits of more than half the Mn ions are filled, so the Fermi level would be elevated by external magnetic field.[6] When the Fermi level approaches a certain defect level in MgO, the electrons can cross the MgO barrier by the hopping mode instead of the tunneling mode. Thus,

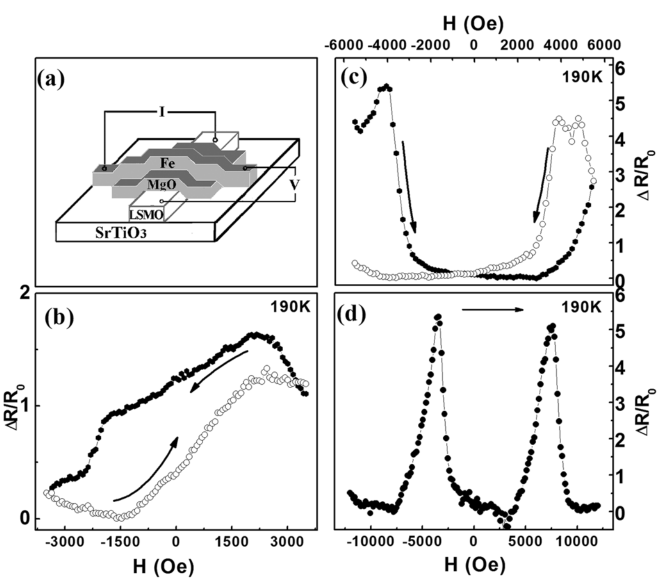

FIG. 2. (a) Schematic representation of a typical device consisted of LSMO bottom electrodes and Fe top electrodes and a MgO barrier. (b)-(d) MR curves of the spin valve device. The magnetic field ranges are (b) ±3.5 kOe, (c) ±5.5 kOe, and (d) ±12 kOe, respectively. All the MR curves were measured at 190 K.

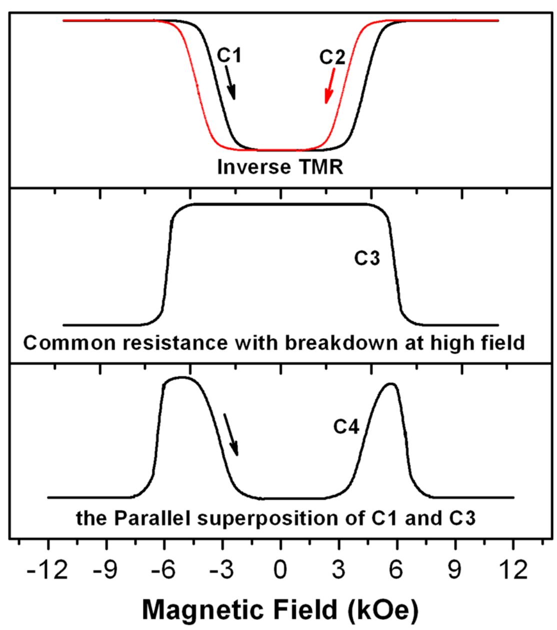

FIG. 3. The sketch for parallel connection superposition of an inverse TMR curve and a common resistance with breakdown at high magnetic field. C1 and C2 are forward part (from −12 to 12 kOe) and backward part (from 12 to −12 kOe) of an inverse TMR curve. C3 depicts a resistance with taking a breakdown effect at high magnetic field into account. C4 is the parallel superposition of C1 and C3.

the antiparallel alignment between LSMO and Fe does not block the conducting electrons any longer, so that the barrier effect of MgO is weakened remarkably. The MR behavior in Figs. 2(c) and 2(d) can be treated as a parallel connection of an inverse TMR and a common resistance accompanied with breakdown at high magnetic field, as sketched in Fig. 3. The top curves C1 and C2 depict an inverse TMR effect. The middle curve C3 sketches the resistance variation of the device without considering TMR but with taking into account the breakdown at high magnetic field. The bottom curve C4 is the parallel combination of the curves C1 and C3, which is in good agreement with the MR curve shown in Fig. 2(d).

In summary, a $La_{0.7}Sr_{0.3}MnO_3$/MgO/Fe spin valve was fabricated on a $SrTiO_3$ substrate. The device shows an inverse TMR, and a MR ratio up to 540% was obtained at 190 K. It indicates that MgO barrier benefits a high MR ratio in $La_{0.7}Sr_{0.3}MnO_3$/MgO/Fe MTJs. At high magnetic field, the breakdown effect was observed, which was attributed to the joint action of defect levels in MgO and Fermi level shift in LSMO. Therefore, during designing and fabricating TMR devices, the defects in barrier material and electronic state change of electrode caused by external conditions should be considered.

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