

Tunneling magnetoresistance with positive and negative sign in $\text{La}_{0.67}\text{Sr}_{0.33}\text{MnO}_3/\text{SrTiO}_3/\text{Co}$ junctions

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We have investigated the effects of modification of the SrTiO_3/Co interface as well as the SrTiO_3 barrier on the tunnel magnetoresistance (TMR) of $\text{La}_{0.67}\text{Sr}_{0.33}\text{MnO}_3/\text{SrTiO}_3/\text{Co}$ junctions. Modification was realized by the introduction of one atomic layer of either TiO_2 or SrO at the SrTiO_3/Co interface. Barriers with different oxygen content were also studied. In these structures we have observed positive as well as negative TMR, with a trend towards positive TMR for junctions with interfacial SrO and/or more oxygen-deficient barriers. This work offers more insight into the SrTiO_3/Co tunnel spin polarization and its sign.

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Magnetic tunnel junctions (MTJs) are devices consisting of two ferromagnetic metallic electrodes separated by a thin insulator barrier.¹ They have already been applied in magnetic memories and read heads and have played a crucial role in the development of spintronics.² Electrodes of half-metallic ferromagnets are attractive because they exhibit electronic transport by 100% spin-polarized carriers, as in the prototypical manganite $\text{La}_{0.67}\text{Sr}_{0.33}\text{MnO}_3$ (LSMO).³ LSMO electrodes have been incorporated in fully epitaxial MTJs using SrTiO_3 (STO) as a barrier.^{4–6} These junctions have shown the highest tunneling magnetoresistance (TMR) with a corresponding LSMO/STO tunneling spin polarization (TSP) of 95%,⁷ albeit not at room temperature. Therefore, effort has been put into engineering the LSMO/STO interface.⁸

The LSMO/STO system can also be used as a spin analyzer to study the TSP of other interfaces. An example is the work on LSMO/STO/Co MTJs, where a negative TMR indicates a negative TSP for Co with a crystalline STO barrier.^{9,10} This is opposite to the positive TSP observed for Co with amorphous Al_2O_3 barriers.¹¹ The negative TSP was ascribed to interfacial d - d bonding manifesting the d band of Co,⁹ agreeing with theoretical modeling.¹² Studies on other systems have used similar arguments.¹³ Besides d - d bonding, other origins for the negative STO/Co TSP have been proposed. First-principles calculations on Co/STO/Co structures¹⁴ assumed a TiO_2 -terminated barrier and found an induced magnetic moment on the interface Ti atoms aligned antiparallel to the Co moment, which might cause the negative TSP. More recent calculations by Velev *et al.*¹⁵ have looked at the complex band structure of STO which allows an efficient tunneling of Co d band electrons. Thus we see two kinds of arguments, pointing either to the specific interface or to the intrinsic properties of the barrier. Interestingly, for an amorphous SrTiO_3 barrier the STO/Co interface has shown positive TSP.¹⁶ The question about the origin of the negative TSP is thus still open.

In this work we aim to investigate the role of the specific interface and tunnel barrier properties on the sign of the STO/Co spin polarization. First, by selectively introducing one atomic layer of TiO_2 or SrO we define the termination of the barrier at the interface. Second, by changing the number of oxygen vacancies in the STO barrier we evaluate the effect of a defective barrier.

We have grown STO(001)/LSMO(11.3 nm)/STO(4.3–5.1 nm)/Co(11 nm)/Au(7 nm) heterostructures by pulsed-laser deposition using a stoichiometric ceramic target for LSMO and a single-crystalline target for STO. The STO (001) single-crystal substrates were chemically treated and annealed at 950 °C to obtain TiO_2 termination.¹⁷ Perovskites were deposited at 750 °C, under O_2 pressure of 0.35 mbar (0.30 mbar) and laser fluence of 2.5 J/cm² (1.0 J/cm²) for LSMO (STO). The laser repetition rate was 1 Hz, with 30-s waiting intervals for every 2 unit cells deposited. After STO barrier deposition the O_2 pressure was increased to 1 bar and kept constant during cool-down at a rate <10 °C/min, to obtain proper O content. Metals were deposited *in situ* at room temperature and 4.0 J/cm² laser fluence. Partial oxidation of the top of the Co electrode proved useful to separate its magnetic switching from the bottom LSMO electrode via exchange bias.

Perovskite layer-by-layer growth was monitored by *in situ* reflection high-energy electron diffraction (RHEED).¹⁸ Transmission electron microscopy (not shown) confirmed that both LSMO and STO grow epitaxially, whereas the top Co electrode is polycrystalline. Standard lithographic techniques are used to define junctions with circular active areas of 150 μm unless stated otherwise. Junctions fabricated according to the process described above will be referenced to as standard junctions. The TMR ratio is defined as $(R_{ap} - R_p)/R_p$, where R_p and R_{ap} are the resistances for parallel and antiparallel magnetization of the two electrodes, respectively. Measurements were done in four-point cross geometry, with all results presented here satisfying that junction resistance be at least 10 times the electrode square resistance.^{19,20}

The TMR in the standard junctions [see Fig. 1(a)] is negative (so-called inverse TMR) and has an asymmetric bias dependence, with a maximum absolute value at ca. +300 mV and vanishing TMR for high negative bias. This result is similar to that reported by De Teresa *et al.*⁹ and indicates a negative tunneling spin polarization for the STO/Co interface. The temperature dependence of junction resistance is shown in Fig. 1(b). Resistance shows a maximum at 260–280 K and a 2–3 times decrease with lowering temperature. This temperature dependence was also observed in similar structures²¹ and is usually attributed to a reduced ef

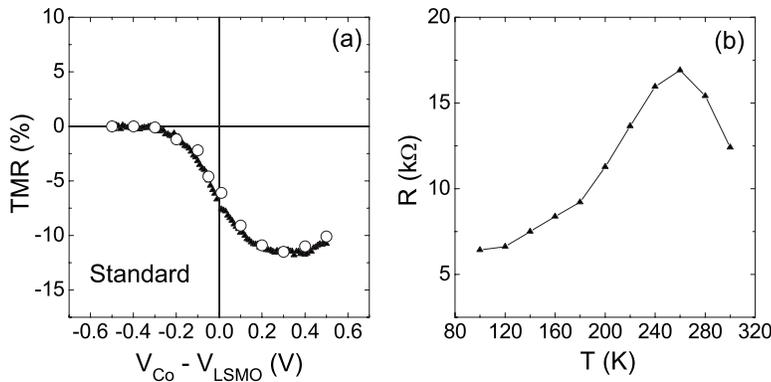


FIG. 1. Standard junctions. (a) TMR at 90 K and (b) R vs T at 40 mV for representative junctions. TMR solid symbols are from I - V curves in parallel and antiparallel states, and open symbols are from magnetic field sweeps at a given bias.

+fective ordering temperature of the LSMO/STO interface.⁶ The TMR temperature dependence was also studied (not shown). For standard junctions the TMR decreased roughly linear with temperature and vanished at about 280 K.

Next we studied the effect of the specific STO/Co interface on the TSP. In principle, by considering the TiO₂ termination of the substrate and assuming a perfect cube-on-cube growth, one would expect the barrier to already have TiO₂ termination. Theoretical calculations point out this termination at the STO/Co interface to be more stable,¹⁴ and there is experimental evidence for the presence of this interface.^{22,23} Still, we decided to observe the result of depositing interfacial TiO₂. Calibration of the amount of laser pulses needed for one atomic layer was extracted from XRD measurements on thick TiO₂ films. Deposition conditions were similar to those of LSMO, except that the O₂ pressure was 0.10 mbar. Just after depositing the STO barrier the TiO₂ deposition was done, followed by O₂ pressure increase to 1 bar and sample

cool-down. The TMR of TiO₂ modified junctions was also negative [see Fig. 2(a)] and did not show any drastic departure from the case of standard junctions. This is in agreement with studies on junctions with complete TiO₂ barriers.²⁴ Junction resistance [see Fig. 2(b)] also showed a temperature dependence similar to the standard case.

It was necessary to use RHEED to control the number of pulses for one SrO atomic layer, as thick films of SrO did not grow epitaxially on STO. Monitoring of the RHEED intensity in the Bragg condition produced only one oscillation.²⁵⁻²⁷ SrO was deposited at 0.13 mbar O₂, 1.1 J/cm² laser fluence, and at a temperature of 800 °C to obtain clearer initial RHEED oscillation. Epitaxial growth was confirmed by observing more than one oscillation in anti-Bragg condition.²⁸ To avoid exposing the SrO atomic layer to the increased O₂ pressure, as the SrO termination is chemically less stable,²⁹ while still keeping proper O content in the STO barrier, we adopted the following procedure: First

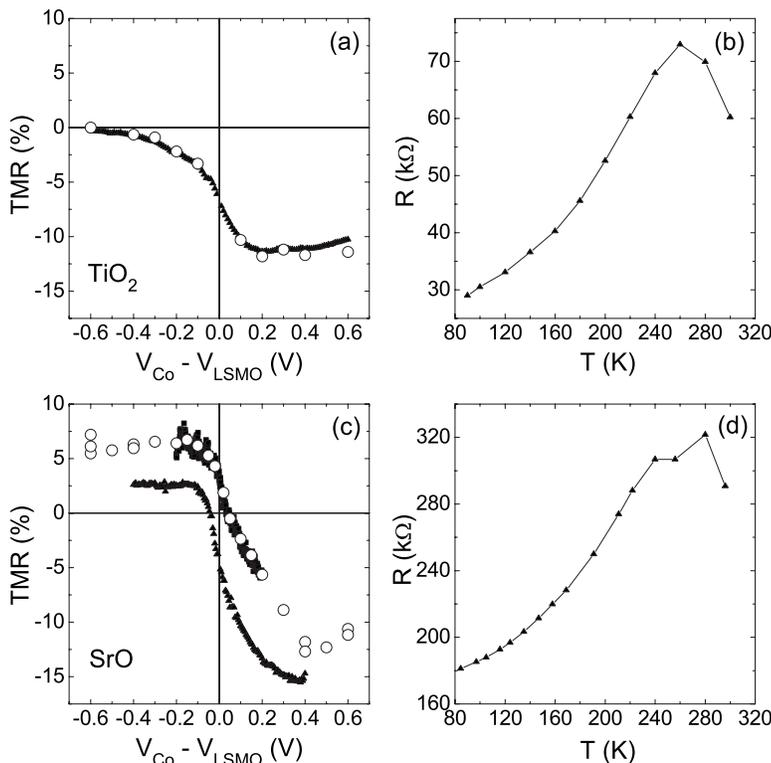


FIG. 2. Interface modified junctions. (a) TMR at 90 K and (b) R vs T at 50 mV for junctions with TiO₂ at the STO/Co interface. (c) TMR at 85 K and (d) R vs T at 10 mV for junctions with SrO at the STO/Co interface. In (c) data for two different junctions are given, where TMR solid symbols are from I - V curves in parallel and anti-parallel states, and open symbols (one junction only) are from magnetic field sweeps at a given bias.

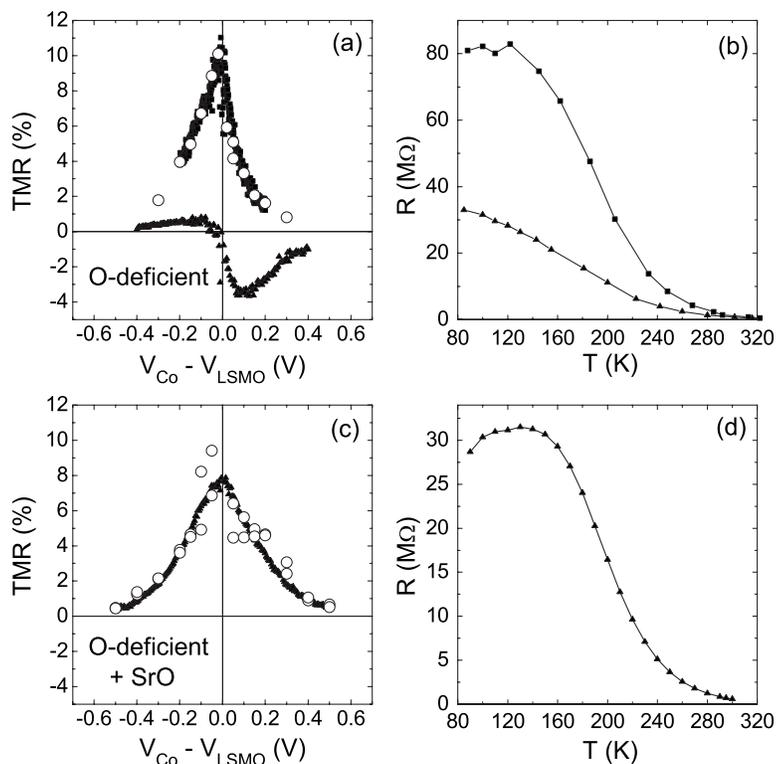


FIG. 3. Oxygen-deficient junctions. (a) TMR at 85 K and (b) R vs T at 10 mV for two junctions with O-deficient barrier. (c) TMR at 90 K and (d) R vs T at 20 mV for a junction with O-deficient barrier as well as SrO at the STO/Co interface. The junction with triangle symbols in (a) and (b) has a diameter of 100 μm .

we deposit the STO barrier and cool-down under 1 bar O_2 , as the standard case. Then we raise the temperature to 800 $^\circ\text{C}$ to deposit the SrO and cool-down using the same pressure as for deposition (0.13 mbar).

Junctions with interfacial SrO showed either positive or negative TMR at zero bias [see Fig. 2(c)]. All junctions exhibited TMR reversal at small bias, with negative TMR at large positive bias and positive TMR for large negative bias. So SrO termination resulted in a trend towards positive TMR, while keeping a similar asymmetry on bias as the standard junctions. The temperature dependence of junction resistance [see Fig. 2(d)] was similar to the standard junctions, indicating similar quality barriers for all junctions discussed so far.

We now consider another kind of modification to the system: the O content. It was pointed out that the O_2 pressure after perovskites deposition was increased to 1 bar to achieve proper O content. We have also fabricated junctions where this step has been omitted; instead, the O_2 pressure has been kept at 0.13 mbar while cooling down from 800 $^\circ\text{C}$. This way we evaluate the result of increasing the number of O vacancies in the barrier.

The properties of O-deficient junctions proved quite different. Figure 3(a) shows the TMR bias dependence for two O-deficient junctions. Here we observe that one junction shows small positive TMR at negative bias and appreciable negative TMR for positive bias, while the other junction shows positive TMR in the whole bias range. We observe again a trend towards positive TMR, only in this case much stronger than for the junctions with interfacial SrO. Considering the positive TMR observed it is interesting to study the result of combining both types of modifications—i.e., SrO termination and O vacancies. Therefore we fabricated junc-

tions where interfacial SrO was introduced just after STO deposition (to change the termination), followed by cool-down in only 0.13 mbar O_2 in order to increase O vacancies. The result of this combined modification on TMR is depicted in Fig. 3(c) as a fully positive TMR in the whole bias range and symmetric bias dependence. Furthermore, both types of O-deficient junctions show a R vs T behavior with resistance increasing drastically (by about 2 orders of magnitude) as the temperature is reduced [see Figs. 3(b) and 3(d)]. This is strikingly different from the junctions of Figs. 1 and 2, which have higher oxygen content. It is worth noting that the O-deficient junctions had higher resistance values than the O-rich junctions at low temperature (MΩ vs kΩ). Compared to the standard junctions, the TMR for O-deficient junctions decreased faster with temperature, vanishing at about 220 K, roughly corresponding to the range where the junction resistance has dropped significantly.

Thus, introducing O deficiencies produces drastic modifications of the junction properties. Both the positive TMR and the symmetrical bias dependence are commonly observed in amorphous Al_2O_3 junctions.³⁰ An important point is that here we have used a crystalline, epitaxial barrier and yet we observe a similar behavior. The effect of an O-deficient barrier on a similar system was previously studied³¹ and found to produce a more symmetrical bias dependence, although no TMR sign reversal was observed. The effect of O vacancies was indirectly studied by using bias crafting in electrically unstable LSMO/STO/CoCr junctions.³² Bias crafting produced positive TMR, though no bias dependence was presented. Similar bias effects had been observed irreversibly with unstable CoFe electrodes.²¹ Here we observe TMR reversal through direct control of O content of the STO barrier and/or the STO/Co interface termination.

The variation of the TMR sign by changing the termination layer at the STO/Co interface suggests that the specific interface properties play a role. A change to positive TMR upon introducing interfacial SrO might support the idea that the negative TMR of standard junctions is due to an antiparallel moment induced on Ti atoms¹⁴ or due to the specific d bonding to Ti. In this sense, the positive TSP for Co with amorphous STO barriers¹⁶ could be related to an unclear or mixed barrier termination. Instead, one can also focus on the bonding between the Co and the O ions of the barrier material,³³ as there is evidence for this type of Co-O bonding.²³ Indeed, it has been pointed out theoretically that interfacial O may be responsible for the positive TSP of Co/Al₂O₃ due to Co-O bonding.³⁴ Although these arguments strengthen the importance of the interface structure and bonding, explicit calculations to explain the difference observed between SrO and TiO₂ termination are not yet available.

Another possibility is that each termination has different influence on interface states. Calculations by Velev *et al.*¹⁵ evidenced interface resonant states in the minority channel which dominate the conductance in Co/STO/Co junctions. If these would be diminished for interfacial SrO, then one would expect a trend towards positive TSP. From the literature on Fe/MgO/Fe junctions it is known that a monolayer of Ag (Ref. 35) or C (Ref. 36) can suppress interfacial resonances. Theoretical calculations for SrO-terminated STO/Co are thus needed. We note that interface disorder due to the introduction of any of the atomic layers cannot be strictly ruled out. This may decrease momentum conservation or destroy interface states,³⁷ possibly changing the TSP. Nevertheless, good growth of TiO₂ films and RHEED oscillations for SrO suggest slight or no interface disorder.

The effect of O vacancies is a trend towards positive TMR and stronger temperature dependence. The stronger decay of both junction resistance and TMR with temperature is consistent with thermal activation of a spin-independent conduction channel via defect states (O vacancies) in the barrier^{38,39} and possibly a lower T_c of the interfacial LSMO.⁶ One mechanism which may account for TMR reversal via these defect states is resonant tunneling⁴⁰ if the states are located close to Fermi level. Recent calculations⁴¹ have shown that O vacancy levels in STO are located 0.5 eV below the conduction band bottom when in the bulk and 0.25 eV when at the surface. If we consider a barrier height of 1.2 eV, which

is that reported by De Teresa *et al.*^{9,42} and close to half the STO band gap, then O deficiencies would be located far enough from the Fermi level to not consider resonant tunneling. In this case, nonresonant scattering by these states must be considered, as recently described for O vacancies in MgO-based junctions.⁴⁵

Nonresonant scattering would affect the conductance and reduce momentum selectivity. In the simplest picture, such scattering would redistribute tunneling electrons among different wave vectors, thereby connecting states for which coherent tunneling is not possible. In Ref. 15 it was calculated for crystalline Co/STO/Co junctions that whereas minority-spin bands have sharp states close to the Γ point which dominate the conductance, the majority-spin bands are mostly featureless. Therefore, scattering and loss of parallel momentum conservation would have a major impact on the minority-spin channel, while probably not affecting the majority-spin channel in a significant manner. While this would produce a less negative TSP of the STO/Co interface, it does not seem likely to result in a sign reversal. The latter feature of our experimental observations therefore remains unexplained. We note that nonresonant scattering is expected to reduce junction conductivity, as was observed in our O-deficient junctions [see Figs. 3(b) and 3(d)]. Similar effects have been predicted for Fe/MgO/Fe junctions.⁴⁵ Finally, it is worth noting that a more subtle effect on TMR can take place through changes in the potential profile.⁴⁶

In conclusion, it has been shown that the TMR within the half epitaxial system LSMO/STO/Co can have positive or negative sign depending on the termination layer of the barrier at the STO/Co interface. Also O vacancies proved to promote positive TMR. Fully positive and symmetric TMR was obtained for O-deficient barriers with SrO termination at the STO/Co interface. These sign changes observed within the same material system highlight the subtle physics and reopen the discussion on the precise origin of the inverse TMR of standard LSMO/STO/Co junctions. Our results provide a more stringent set of experimental observations to help narrow down possible theoretical explanations, and we hope it stimulates further work in that direction.

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