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Bias-crafted magnetic tunnel junctions with bistable spin-dependent states

M. Bowen^{a)}

Unité Mixte de Physique CNRS/Thales, Route départementale 128, 91767 Palaiseau Cedex, France;
 Université Paris-Sud XI, 91405 Orsay Cedex, France; and Institut de Physique et Chimie des Matériaux de
 Strasbourg (IPCMS), UMR 7504 CNRS-ULP, 23 rue du Loess BP 43, 67034 Strasbourg, France

J.-L. Maurice, A. Barthélémy, P. Prod'homme, E. Jacquet, and J.-P. Contour

Unité Mixte de Physique CNRS/Thales, Route départementale 128, 91767 Palaiseau Cedex, France
 and Université Paris-Sud XI, 91405 Orsay Cedex, France

D. Imhoff and C. Colliex

Laboratoire de Physique des Solides, Bât. 510, Université de Paris-Sud, 91405 Orsay, France

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The authors have observed stable, reversible two-resistance states with substantial tunneling magnetoresistances of opposite signs in $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3/\text{SrTiO}_3/\text{Co}_{1-x}\text{Cr}_x$ junctions. Electron energy loss spectroscopy studies reveal the segregation and oxidation of electrochemically reactive chromium at that interface, resulting in oxygen vacancies in the oxide barrier. Bias-induced switching between the two junction states is argued to reflect the incidence of these barrier defects at and near the electrically unstable $\text{SrTiO}_3/\text{Co}_{1-x}\text{Cr}_x$ interface. This affirms bias crafting as an additional lever in spintronic research across semiconducting spacers. © 2006 American Institute of Physics. [DOI: 10.1063/1.2345592]

Recent progress in the research field of magnetic tunnel junctions has hinged on understanding how symmetry-resolved wave function coupling across epitaxial systems with semiconducting spacers determines their spintronic response. In $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3/\text{SrTiO}_3(001)$ (LSMO/STO), the symmetry mismatch between tunneling carriers and the lowest barrier height was shown to lead to exotic spintronic effects and to underscore a convergence between the spintronics across metallic and semiconducting spacers.¹ The case of matching these symmetries has also been intensely studied in the Fe/MgO(001) system, thanks to a convergence between experiment^{2–5} and theory.^{6,7} Nevertheless, the measured values of tunneling magnetoresistance (TMR), which currently reach 300%, remain much lower than the 5000% predicted to result from effective spin filtering due to this case of symmetry matching, despite increasing the nominal barrier thickness⁴ or annealing the junction.⁵ This mainly reflects the presence of contaminants and defects in the epitaxial ultrathin barrier, which leads to electrical conduction through pinholes with a consequently reduced effective thickness and a lowered barrier height.⁴

To overcome this challenge, these deviations from the ideal symmetry-filtering semiconducting spacer need to be controlled. It is in principle possible to mitigate both depending on growth conditions. More interestingly, transport experiments on oxygen-deficient oxide dielectrics^{8–10} have hinted at a postgrowth *bias-crafting* solution to control defects in a tunnel junction barrier. We demonstrate this effect in $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3/\text{SrTiO}_3/\text{Co}_{1-x}\text{Cr}_x$ trilayers. In contrast to their LSMO/STO/Co brethren with stable resistance and inverse TMR,¹¹ such junctions exhibit an electric instability that leads, thanks to an applied bias, to the reversible formation of two typical junction states with substantial low-bias TMR that differ both in resistance and in the sign of TMR. Electron energy loss spectroscopy (EELS) experiments re-

veal the segregation and oxidation of Cr at the STO/CoCr interface, leading to oxygen vacancies in the STO barrier. The full tunneling spin polarization from the LSMO/STO interface^{12,13} demonstrates how bias crafting acts as a spintronic lever, resulting in a device with four addressable states.

STO(001) || LSMO(350 Å) / STO(27.8 Å) / CoCr(150 Å) / Au(50 Å) samples were grown by a combined use of pulsed laser deposition for the epitaxial oxide layers and room-temperature molecular beam epitaxy for the transition metals after temporary exposure to air during sample transfer. A large number of LSMO/STO/CoCr magnetic tunnel junctions (MTJs) with $0.04 \leq x \leq 0.2$ were studied¹⁴ from over one dozen samples processed by standard lithographic techniques.¹²

Relative to the electrically stable LSMO/STO/Co junctions, grown and patterned using the same procedures, LSMO/STO/Co_{1-x}Cr_x junctions overwhelmingly exhibit electrical instability, leading to striking, reversible changes to both resistance and TMR values measured at low bias after applying a bias of given sign and amplitude. Figure 1 presents R(H) loops taken at an applied dc bias $V=500 \mu\text{V}$ on a LSMO/STO/Co₈₀Cr₂₀ junction with a surface area of $12 \mu\text{m}^2$. In its virgin state, the junction exhibited a poorly defined TMR response [see Fig. 1(a)]. After several cycles of positive and negative current sweeps to craft the junction, ending with a sweep to $I_{\text{max}}=-3.65 \mu\text{A}$, a positive TMR of +38% was measured [see Fig. 1(b)], corresponding to a junction state that we label state II. A subsequent sweep to $I_{\text{max}}=+4.3 \mu\text{A}$ led to the formation of a well-defined state I [see Fig. 1(c)] with -20% TMR, of the same sign as that for a conventional LSMO/STO/Co junction.¹¹ Interestingly, the field value $H=1 \text{ kOe}$ at which the MTJ switches from the antiparallel to the parallel configuration (see Fig. 1) corresponds to the coercivity of the 150 Å Co₈₀Cr₂₀ layer as measured by superconducting quantum interference device (data not shown). This reversible bistable switching behavior is

^{a)}Electronic mail: martin.bowen@ipcms.u-strasbg.fr

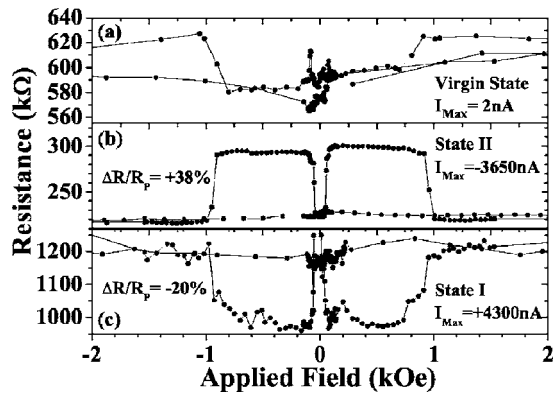


FIG. 1. LSMO/STO/Co₈₀Cr₂₀ junction at $T=4$ K. $R(H)$ loops taken at $V=500$ μ V (a) in the junction's virgin state and after preparing states (b) II and (c) I.

reproduced on other junctions of larger size and lower Cr nominal concentration, though with lower TMR values. No particular threshold bias leading to the formation of a given state could be reproduced across this extensive study. This indirectly points to the prevalence of pinholes on the transport properties across the epitaxial barrier as we will discuss later.

To pinpoint the role of Cr incorporation in our stack on spin-dependent transport, we first performed transmission electron microscopy (TEM) on a LSMO/STO/Co₈₄Cr₁₆ trilayer that also yielded clear bias-crafted states I and II after junction processing. This revealed an abrupt epitaxial LSMO/STO interface and a more disordered STO/CoCr interface (not shown), similar to that on a LSMO/STO/Co junction.¹⁵ We then performed EELS analyses by digitally scanning along lines that intersect the different layers and interfaces at right angles. Each line consisted of sixty-four 1340-channel spectra. With 0.5 eV per channel, each spectrum included the O-K, Cr-L, Mn-L, Co-L, and La-M signals. The probe diameter was about 1 nm and the interval between spectra was 0.6 nm. Our experimental and data analysis procedures are detailed in Ref. 15. Due to our fixed sample holder, the interfaces were tilted with respect to the electron beam. Thus, the effective spatial resolution of elemental profiles, defined as twice the distance from an abrupt interface at which the intensity is halved, was approximately 2.5 nm.

Figure 2(a) presents the evolution of elemental profiles across the LSMO/STO/CoCr junction. The shifted Cr profile towards STO relative to that of Co reveals a segregation of Cr at the interface. We believe that sample heating, which is similar between our TEM preparation and MTJ fabrication¹² processes, may account for this departure from the nominal alloy composition.¹⁶ The slight superposition of the Cr concentration front with the Ti concentration peak might indicate the presence of Cr within the STO barrier, but may also reflect the limited spatial resolution of our experiment.

We now discuss the fine structure of the O-K, Cr-L, and Co-L edges. The O-K edge adopts three shapes when scanning the probe from LSMO to CoCr [see Fig. 2(b)]: one typical of LSMO with a sharp peak at the edge onset, one that corresponds to the STO barrier but differs from a typical STO signal in that the peak at the edge onset should be sharper than in LSMO (Ref. 17) yet is smoothed, and one with peaks at different energies that corresponds to an unusual interfacial phase. Quite remarkably, the simplest expla-

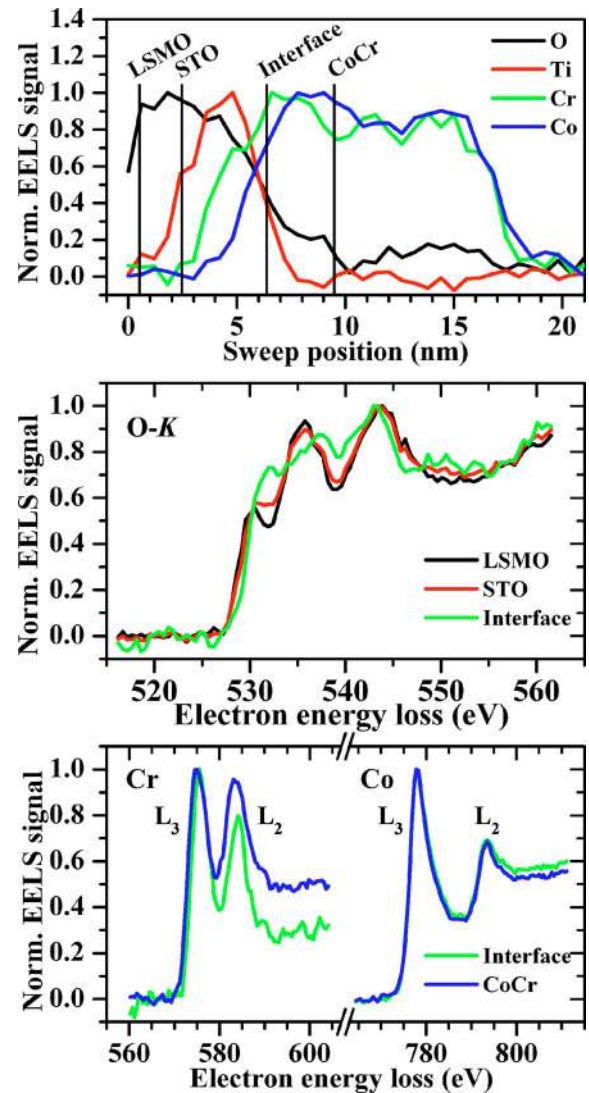


FIG. 2. (Color online) EELS analysis of the LSMO/STO/Co₈₄Cr₁₆ junction stack. (a) Normalized elemental profiles from the LSMO layer across the STO barrier and into CoCr. Vertical lines define sweep positions for spectra in (b) and (c). (b) Oxygen K edge in LSMO, STO, and at the STO/CoCr interface. (c) Cr-L and Co-L edges at the STO/CoCr interface and in bulk CoCr.

nation of the smoothing of the peak in the STO barrier is the presence of several percent of oxygen vacancies.¹⁸ Turning to the Cr spectra [see Fig. 2(c)], the Cr- L_2 and Cr- L_3 peaks have about the same maxima inside the CoCr layer. However, at the interface, the edge onset is shifted towards higher energy losses, and the Cr- L_2 peak is much smaller. These changes in transition metal edges are typical of oxidation.^{17,19} On the other hand, the Co-L edges measured at the interface and in the CoCr bulk are almost identical [see Fig. 2(c)], implying that the interfacial Co is not oxidized here, in contrast to the STO/Co interface.¹⁵

Since oxygen vacancies have appeared in the STO barrier, it is then quite clear that the segregated, oxidized chromium has somehow pumped out oxygen from the STO ultrathin layer. This phenomenon resembles that described by Watanabe *et al.* in the case of Cr-doped STO,⁸ and that very recently discussed by Szot *et al.* as an effect of oxygen trapping and release by structural defects in STO.¹⁰ Furthermore, both articles describe electric-field induced changes to defects along micron-long filamentary paths of conduction that

result in bistable device resistances. Across an ultrathin epitaxial barrier, these paths become conduction hot spots that channel the bulk of the junction current, and defects may therefore be easily modified due to the large $E > 10^6$ V/cm electric field across the ultrathin STO dielectric even at low bias.

Given the nearly total spin polarization at the LSMO/STO interface,^{12,13} we deduce that the improvement and manipulation of the spintronic properties at the STO/CoCr tunneling interface result from a defect-driven mechanism that is bias induced. This may either reflect changes to the interfacial potential profile²⁰⁻²² or resonant tunneling²³ through these bias-crafted oxygen vacancies that, according to that theory, are favorably located at one interface of the junction.

In conclusion, we have crafted the spin-dependent properties of an electrochemically unstable and spintronically dead MTJ interface, thanks to an applied bias. This leads in our LSMO/STO/CoCr MTJs to reversible, stable two-resistance states associated with sizable TMR amplitudes of opposite signs, i.e., to a device with four addressable states. Our method and results suggest that bias crafting not only be taken into account in bias-dependent tunnel junction studies, much like thermal annealing, but also be harnessed as a systematic method to engineer a junction with improved barrier/interface quality, and to control its spintronic response. This should lead to the electrochemical engineering of an effective interface with interesting spintronic properties from the elemental constituents of the nominal one.

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