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LETTER TO THE EDITOR

Half-metallicity proven using fully spin-polarized tunnelling

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Abstract
A half-metal has been defined as a material with propagating electron states at the Fermi energy only for one of the spin directions. But is it fully half-metallic, that is without electrons with opposite spin at that energy? We have studied the spin-conserving process of tunnelling between $La_{0.7}Sr_{0.3}MnO_3$ half-metallic electrodes across an ultrathin $SrTiO_3$ insulator. This experiment demonstrates that the class of half-metallic materials indeed exists at non-zero temperatures, even at interfaces. It also shows that a fully spin-polarized tunnelling current may persist at large bias.

The discovery in 1983 of a new class of materials called half-metals [1], for which conduction is ensured by electrons with only one of two possible values of quantum mechanical spin, has fuelled much research on their synthesis and integration into spintronic devices. However, despite its namesake, a controversy has arisen over whether such materials are indeed devoid of electron states with opposite spin at the Fermi level, thereby calling into question the nature of this class of materials. Indeed, the presence of such states has been argued to arise from finite temperature effects [2], the spin–orbit interaction, symmetry breaking and disorder at interfaces, and more recently, in the form of non-quasiparticle (NQP) states (see for example [3]), from electronic correlation. Though such electronic states have thus far not been measured experimentally through the techniques of spin-polarized photoemission and inverse photoemission spectroscopy, in particular regarding the prototypical half-metallic manganite $La_{0.7}Sr_{0.3}MnO_3$ (LSMO) [4, 5], the relative accuracy of such techniques has not settled this issue.

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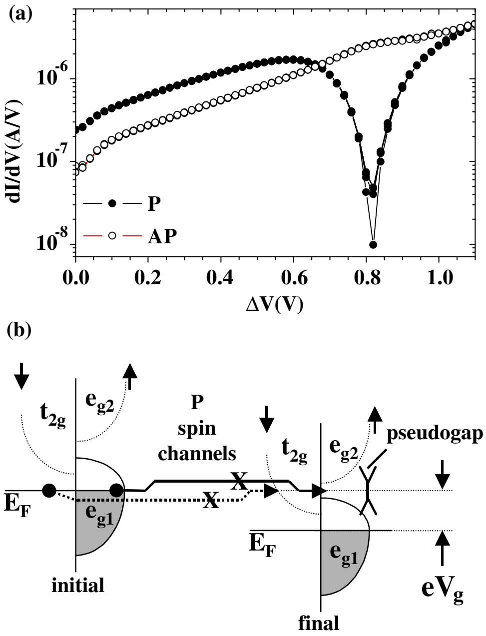

Figure 1. (a) Conductance dI/dV versus applied bias ΔV across an LSMO(350 Å)/STO (27.8 Å)/LSMO(100 Å) junction grown epitaxially on STO(001) in the parallel (P) and antiparallel (AP) configurations of electrode magnetization. (b) Schematic diagram of the densities of states of the two electrodes for an applied bias $V_g = 0.82$ V, with arrows representing the two spin channels in the parallel (P) configuration, which are blocked (crosses).

(This figure is in colour only in the electronic version)

Another such spectroscopic approach is that of spin-polarized tunnelling [5] between half-metallic electrodes across an insulating barrier. Previous measurements at a low value of applied bias between LSMO electrodes across an ultrathin $SrTiO_3$ (STO) layer, which acts as the tunnel barrier, have yielded a spin-up polarization of the two spin channels across this epitaxial magnetic tunnel junction exceeding 95% [6]. This value is limited both by difficulties in obtaining two perfect junction interfaces, and by the excitation of spin waves in a low-bias regime spanning about 150 mV [5], which may reflect the density of NQP states above the Fermi level [3]. Further measurements at a higher applied bias outside this transport regime now conclusively affirm the total spin polarization of states at the Fermi level of the LSMO/STO(001) interface.

LSMO/STO/LSMO trilayers were epitaxially grown onto STO(001) substrates using pulsed laser deposition at 700 °C in a 350 mTorr oxygen ambient. Junctions were then defined using conventional UV photolithography, and measurements performed in four-point DC mode at $T = 10$ K using a closed-cycle cryostat. Additional details regarding growth and patterning may be found in [6].

Figure 1(a) presents the evolution of conductance $G = \mathrm{d}I/\mathrm{d}V$ as a function of applied bias ΔV between the lower and upper electrodes at $T = 10$ K for a $2 \times 6\ \mu\mathrm{m}^2$ junction with a barrier height of 1.85 eV. In the parallel (P) configuration of the electrodes' magnetization, both initial spin-up and (if any) spin-down electrons at the LSMO Fermi level of the injecting electrode may tunnel into respective final spin-up and spin-down states of the collecting electrode. In the antiparallel (AP) configuration, the spin channels are crossed, such that initial spin-up electrons tunnel toward final spin-down states. Once the applied bias allows initial spin-up electrons to reach final states at the bottom of the spin-down t_{2g} band, the conductance G_{AP} in the AP configuration increases more quickly than G_P [5]. The striking observation is that, past $V = 0.6$ V, while G_{AP} continues to increase, G_P decreases and reaches a minimum at $V_g = 0.82$ V, which is one order of magnitude lower than the conductance found at $V = 0$. As shown schematically in figure 1(b), this absence of conduction in the P configuration at V_g implies that both spin channels are blocked. (i) P spin-up channel: initial spin-up electrons are available in LSMO for tunnelling, but at energy $E = E_F + eV_g$ no final spin-up states are present to tunnel into. Such a pseudogap in the density of states has been predicted to result from a Jahn–Teller distortion of the oxygen octahedral environment of Mn in manganites [7], and such changes can be expected at an interface as probed by tunnelling. (ii) P spin-down channel: since spin-down final states are available at $E = E_F + eV_g$ [5], the blocking of this channel implies, within experimental limits, that no spin-down initial states exist in LSMO at E_F.

The extent to which both spin channels are blocked may be discussed more quantitatively: differential TMR, defined as the difference between G_P and G_{AP} relative to G_{AP}, reaches -99% at V_g, i.e. within 1% of the theoretical maximum which defines fully half-metallic behaviour. At V_g in the AP configuration, there is a fully spin-polarized tunnelling current from majority spin initial states to minority spin final states.

To summarize, the collapse at $V_g = 0.82$ V of the parallel conductance, which is comprised of both the spin-up and spin-down channels, offers two strong conclusions. First, it substantiates the fully half-metallic property of LSMO. Indeed, the absence of conduction in the P spin-down channel, despite the presence of spin-down final states at $E = E_F + eV_g$, implies that no initial spin-down states, whether propagating or not, are present at the Fermi level of the LSMO/STO interface. Second, the blocking of the P spin-up channel due to the absence of final spin-up states at $E = E_F + eV_g$, and despite the presence of final spin-down states at that energy, proves that a tunnelling current through a solid-state insulating barrier can be *fully* spin polarized at large bias despite possible spin-flip relaxation mechanisms—an important result for future computer memory design [8]. Thus, our experimental results confirm that half-metals really do exist at non-zero temperatures and can maintain this property at interfaces. Further efforts toward the design of novel half-metallic materials with more robust properties are now needed.

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