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REPORTS

Role of Metal-Oxide Interface in Determining the Spin Polarization of Magnetic Tunnel Junctions

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The role of the metal-oxide interface in determining the spin polarization of electrons tunneling from or into ferromagnetic transition metals in magnetic tunnel junctions is reported. The spin polarization of cobalt in tunnel junctions with an alumina barrier is positive, but it is negative when the barrier is strontium titanate or cerium lanthanite. The results are ascribed to bonding effects at the transition metal–barrier interface. The influence of the electronic structure of metal-oxide interfaces on the spin polarization raises interesting fundamental problems and opens new ways to optimize the magnetoresistance of tunnel junctions.

A tunnel junction consists of two metallic layers (electrodes) separated by a thin insulating layer. When the electrodes are ferromagnetic, the tunneling of electrons across the insulating barrier is spin-polarized, and this polarization reflects that of the density of states (DOS) at the Fermi level (E_F) of the electrodes. This spin polarization is the origin of the tunneling magnetoresistance (TMR), which is currently a hot topic of research in magnetism (*1*) and very promising for applications (*2*). Paradoxically, even though applications have already begun to be developed, there are still gaps in our understanding of spin-polarized tunneling. For example, the physics governing the spin polarization of tunneling electrons is not clearly understood. Previously, the spin polarization P of electrons tunneling from a given ferromagnetic electrode was generally thought to reflect a characteristic intrinsic spin polarization of the DOS in the ferromagnet,

$$P = \frac{N_\uparrow(E_F) - N_\downarrow(E_F)}{N_\uparrow(E_F) + N_\downarrow(E_F)} \qquad (1)$$

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However, recent findings show that the amplitude of the spin polarization, and even its sign, depends on the choice of barrier material (*3, 4*). Here, we describe a series of TMR experiments on Co/I/$La_{0.7}Sr_{0.3}MnO_3$ (LSMO) tunnel junctions, where the barrier I can be $SrTiO_3$ (STO), $Ce_{0.69}La_{0.31}O_{1.845}$ (CLO), or Al_2O_3 (ALO). The effective polarization of Co was found to be positive (higher tunneling probability for majority spin electrons) when I is ALO, and negative (higher tunneling probability for minority spin electrons) when I is STO or CLO. Moreover, the bias dependence of the TMR is completely different in these two cases. The strong influence of the electronic structure of the barrier and barrier-electrode interface in tunnel junctions raises interesting fundamental problems and presents new ways to tailor the TMR.

The first piece of information on the spin polarization of electrons tunneling from a ferromagnetic metal (F) comes from experiments on F/I/S junctions, in which the second electrode is a superconductor (S). The spin splitting of the quasi-particle DOS of S, induced by a magnetic field, can be used to analyze the spin polarization of the tunneling current. Extensive data have been obtained with F/ALO/Al junctions, and a positive polarization has been found for all the ferromagnetic metals and alloys that have been studied (*5*). This is surprising, especially for metals like Co or Ni in which a negative polarization is expected from the smaller DOS at E_F for the majority spin direction (the majority spin d subband is below E_F). This problem has not been clearly solved, even though it is frequently argued that s-character electrons should tunnel more easily, so that the experimental positive polarization can reflect only that of the s-character DOS (*6, 7*). Some theoretical justification has been provided by ab initio calculations of the electronic structure at a Co-ALO interface. Nguyen-Mahn *et al.* (*8*) determined the DOS of the tunneling electrons on the first Al atoms at the interface and found that, because of an sp-d bonding mechanism between Al and Co, this DOS is positively polarized. This can be viewed as an interface filtering effect controlling the starting point of the polarized evanescent wave in the barrier.

In junctions with two ferromagnetic electrodes, F_1/I/F_2, spin-polarized tunneling gives rise to TMR because the resistance of the junction depends on whether the electrodes have parallel or antiparallel magnetizations. This change can be large, typically 15 to 40% at room temperature, so that TMR has great relevance for the technology of MRAM (magnetic random access memory) or read heads. The experimental results at low bias are generally interpreted according to Jullière's expression,

$$\frac{\Delta R}{R} = \frac{R_{AP} - R_P}{R_{AP}} = \frac{2P_1P_2}{1 + P_1P_2} \qquad (2)$$

where R_{AP} and R_P are the resistances in the antiparallel and parallel states, respectively, and P_1 and P_2 are the electron spin polarizations of the two electrodes.

In junctions studied up to now, mostly with ferromagnetic transition-metal electrodes and ALO barriers, a normal TMR has been found; that is, the tunnel resistance is smaller when the magnetizations of F_1 and F_2 are parallel. This behavior is expected when the sign of the polarization coefficient P is the same for both electrodes and is consistent with the aforementioned uniformly positive spin polarization found for various transition metals in F/ALO/Al junctions. However, two recent results have indicated that, with types of barrier other than ALO, the spin polarization of electrons tunneling from Co or NiFe (permalloy) can also be negative. Sharma *et* *al.* (*3*) have shown an inversion of the TMR ($R_{AP} < R_P$) in NiFe/Ta_2O_5/ALO/NiFe double-barrier junctions, which indicates opposite polarizations at the NiFe-Ta_2O_5 and ALO-NiFe interfaces. De Teresa *et al.* (*4*) have also obtained an inverse TMR effect in Co/STO/LSMO tunnel junctions. In LSMO, spin-resolved photoemission experiments at low temperature have confirmed the strong positive polarization of the DOS at the Fermi level expected from the conduction mechanism by hopping of only majority spin electrons between Mn^{3+} and Mn^{4+} (*9*). With a positive polarization for LSMO, the inverse TMR of Co/STO/LSMO junctions implies that, with a STO barrier, the polarization of Co is negative. This is in contrast to what is found with ALO but is in agreement with the DOS of the d band of Co. Such a result is confirmed by the bias dependence of the TMR, which reflects the structure of the DOS of the Co d band (*4*).

The junctions we have studied are composed of 35 nm of LSMO deposited on a STO(001) substrate for the bottom electrode; 2.5 nm of STO or CLO, or 3.0 nm of ALO, for the barrier; and 30 nm of Co for the top electrode. We have also studied a Co/ALO/STO/LSMO junction in which the barrier is composed of 1.5 nm of ALO and 1 nm of STO. The samples are capped with a 5-nm Au layer. The LSMO, STO, and CLO thin films were grown using pulsed laser deposition with an oxygen pressure of 0.35 torr and at a temperature of around 700°C. Co and Au were deposited by sputtering. High-resolution transmission electron microscopy images show that the growth of LSMO on the STO substrate and that of STO and CLO on LSMO are epitaxial. In contrast, the Co layers are polycrystalline. The ALO insulating barrier was obtained by radio-frequency sputter etching of an Al layer in an Ar-O_2 plasma (*10*). The trilayers were etched using a conventional ultraviolet lithography technique to define mesa structures (with diameters of 10, 20, or 50 μm) for tunneling experiments. Details on the preparation and on the magnetic characterization have been published elsewhere (*4*). Here, we compare the properties of junctions with approximately the same barrier thickness, 2.5 to 3.0 nm. The smaller resistance of the junctions with STO and CLO barriers is consistent with the smaller gap of STO and CLO relative to that of ALO and also with the slightly smaller thickness of the STO and CLO barriers. In measurements with mesa structures, it is important that the tunnel resistance be much larger than the resistances of the electrodes between the barrier and the voltage contacts at the top and bottom of the mesa. According to our direct measurements of the resistance of the electrodes, these resistances never exceed 0.02% of the tunnel resistance (*4*). This rules out any contribution from the magnetoresistance of the electrodes

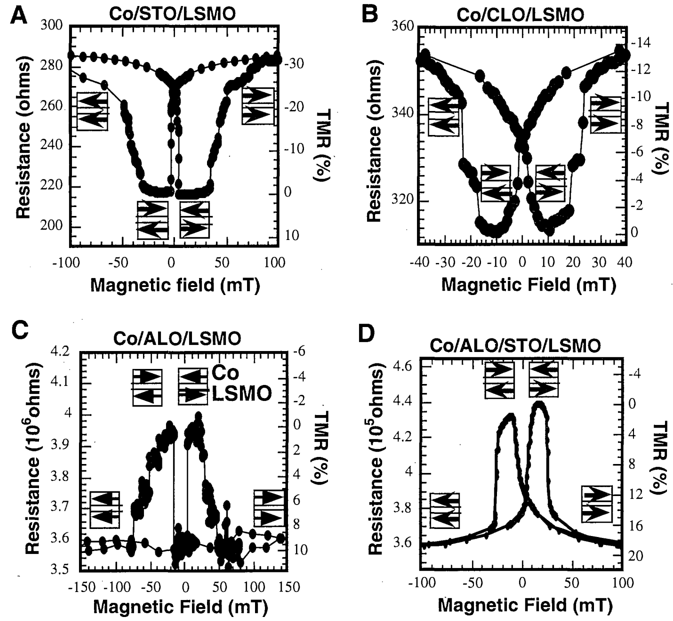

Fig. 1. TMR curves recorded at 40 K with a bias of −10 mV for (**A**) Co/STO/LSMO, (**B**) Co/CLO/LSMO, (**C**) Co/ALO/LSMO, and (**D**) Co/ALO/STO/LSMO junctions. The field ranges of parallel and antiparallel magnetic alignment are schematically indicated by arrows and are consistent with magnetization measurements (*4*). With our definition, the electrons tunnel from LSMO to Co at negative bias.

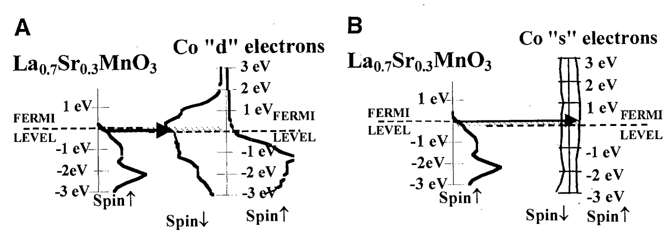

Fig. 2. (**A** and **B**) The left profiles represent the half-metallic DOS of the LSMO derived from photoemission experiments of Park *et al.* (*9*). The right profiles represent the majority (spin ↑) and minority (spin ↓) DOS of the d-character (A) and s-character (B) electrons calculated for the (001) surface of Co by Wang (*12*). The arrows represent the most probable tunneling transitions at a small negative bias, in (A) from spin ↑ states of LSMO to spin ↓ d-states of Co in the antiparallel configuration, in (B) from spin ↑ to spin ↑ states in the parallel configuration.

and any geometrical effect of the type described by Moodera (*11*). We have also measured Co/STO/LSMO junctions with thicker barriers, which exhibit much higher resistances but still the same TMR behavior.

TMR curves were obtained on Co/I/LSMO tunnel junctions with I = ALO, STO, CLO, or ALO/STO at a small applied bias of −10 mV (Fig. 1). An inverse TMR (with $R_{AP} < R_P$) was obtained for STO (Fig. 1A) and CLO (Fig. 1B) barriers, whereas a normal TMR ($R_{AP} > R_P$) was found for ALO (Fig. 1C) and ALO/STO (Fig. 1D) barriers. From Eq. 2 with a positive polarization of LSMO, we can determine that the effective polarization of Co at the Fermi level is negative when the barrier is STO or CLO, and positive, as generally found, when the barrier is ALO. The negative polarization of Co when the barrier is STO or CLO can be viewed as a preferential transmission of electrons of d character at the Co-STO and Co-CLO interfaces. The positive polarization when the barrier is ALO has been ascribed to the selection of the s-character electrons by bonding effects at the Co-ALO interface (*8*). Inserting a STO layer between ALO and LSMO does not result in a change from the behavior of an ALO barrier alone (Fig. 1D), which suggests that the positive sign of the Co polarization is associated with the electronic structure at the Co-ALO interface rather than with the propagation through the barrier.

The relative position of the DOS for LSMO [majority spin band derived from photoemission experiments (*9*)] and the DOS for the d-character (Fig. 2A) or s-character (Fig. 2B) electrons calculated for the (100) surface of Co (*12*) is shown for an applied bias around zero (–10 mV) corresponding to the measurements reported in Fig. 1. At this bias value, the band structure of Co in the vicinity of the Fermi level is probed. When the tunneling of d-character electrons is predominant (STO or CLO barriers), given that the d-band DOS at the Fermi level of Co is larger for the minority spin direction (see Fig. 2A), the most probable transitions are those between the majority spin-up (spin ↑) band of LSMO and the minority spin-down (spin ↓) d-states of Co occurring in the antiparallel configuration, which accounts for the observed inverse TMR. At larger bias, one expects to probe the relatively fine structure of the d-band DOS. For negative bias, the Fermi

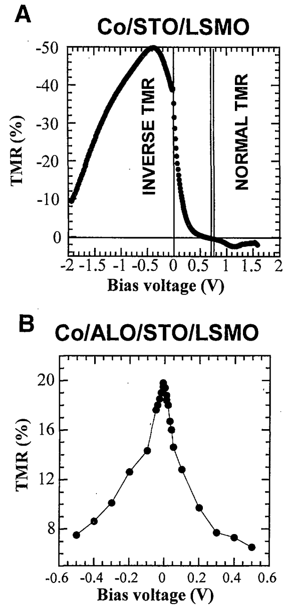

Fig. 3. Bias dependence of the TMR ratio in (**A**) Co/STO/LSMO and (**B**) Co/ALO/STO/LSMO tunnel junctions.

level of LSMO is situated above the Fermi level of Co and a maximum of inverse TMR is expected when the Fermi level of LSMO is approximately at the maximum of the spin ↓ DOS of Co. This is consistent with the maximum of inverse TMR observed at −0.4 V for Co/STO/LSMO junctions (Fig. 3A). For a positive bias, the TMR is expected to change sign and become normal above 1 V when the Fermi level of LSMO goes down into the energy range of the majority spin d-band of Co. This is also observed in Fig. 3A.

For ALO and ALO/STO barriers, a predominant tunneling of s-character electrons (see arrow in Fig. 2B) is the usual explanation of the positive polarization (*6–8*). The rapid drop with bias (Fig. 3B) is similar to what has been observed in most junctions with ALO barriers, and completely different from what is obtained when the tunneling is predominantly by d-character electrons (Fig. 3A). The origin of this rapid decrease of the TMR at relatively small bias has never been clearly explained. This is roughly consistent with the energy dependence of the DOS induced by sp-d bonding effects on the first atomic layer of ALO in the calculation of Nguyen-Mahn *et al.* (*8*) for the Co-ALO interface. But Zhang *et al.* (*13*) have also shown that a large part of the TMR drop can be attributed to the excitation of spin waves.

The experiments reported here and in several recent publications (*3, 4*) demonstrate the important role of the electronic structure of the metal-oxide interface in determining the spin polarization of the tunneling electrons. The negative polarization for the Co-STO interface has been ascribed to d-d bonding effects between Al and Ti (*4*). This interpretation is similar to that proposed to explain, in terms of sp-d bonding, the positive polarization at the Co-ALO interface (*8*). However, there is no general theory predicting the trend of the experimental results for Co—that is, a negative polarization with oxides of d elements (STO, CLO, Ta_2O_5) and a positive one when there are only s and p states (ALO). It is likely that the spin polarization should also depend on the position of the Fermi level with respect to the electronic levels of each character above and below the gap of the insulator. In addition, as an evanescent wave in an insulator is a Bloch wave with an imaginary wave vector, one can expect different decay lengths for Bloch waves of different character. This means that the final polarization could also depend on the thickness of the barrier, as illustrated by the calculations of MacLaren *et al.* for Fe/ZnSe/Fe junctions (*14*).

The influence of the barrier on the spin polarization opens new ways to shape and optimize the TMR. Interesting bias dependencies can be obtained with barriers selecting the d electrons and probing the fine structure of the d-DOS, as in Fig. 3A. The DOS of a d-band can also be easily tailored by alloying (for example, by introduction of virtual bound states) to produce specific bias dependencies. Although here we concentrated on the problem of the spin polarization of the Co electrode and regarded the strongly spin-polarized LSMO only as a useful spin analyzer, the large TMR ratios obtained by combining Co and LSMO electrodes (50% with a STO barrier) are also an interesting result. The drawback arising from the low Curie temperature of LSMO (~350 K) is the reduction of the TMR at room temperature, down to about 5% at 300 K in Co/STO/LSMO (*4*). However, other types of oxides of the double-perovskite family (for example, Sr_2FeMoO_6) combine electronic properties similar to those of manganites with a definitely higher Curie temperature (*15*). Their use in magnetic tunnel junctions is promising for a new generation of tunnel junctions with very high magnetoresistance for room-temperature applications.

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Emergence of Scaling in Random Networks

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Systems as diverse as genetic networks or the World Wide Web are best described as networks with complex topology. A common property of many large networks is that the vertex connectivities follow a scale-free power-law distribution. This feature was found to be a consequence of two generic mechanisms: (i) networks expand continuously by the addition of new vertices, and (ii) new vertices attach preferentially to sites that are already well connected. A model based on these two ingredients reproduces the observed stationary scale-free distributions, which indicates that the development of large networks is governed by robust self-organizing phenomena that go beyond the particulars of the individual systems.

The inability of contemporary science to describe systems composed of nonidentical elements that have diverse and nonlocal interactions currently limits advances in many disciplines, ranging from molecular biology to computer science (*1*). The difficulty of describing these systems lies partly in their topology: Many of them form rather complex networks whose vertices are the elements of the system and whose edges represent the interactions between them. For example, liv-

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