Transmission spectra of Fe/MgO (001) double-barrier tunnel junctions at finite bias

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(Dated: August 8, 2008)

In this contribution, we calculate in a self-consistent way the ballistic transmission as a function of energy of one Fe/MgO (001) single-barrier and one double-barrier tunnel junction, relating them to their electronic structure. The transmission spectra of each kind of junction is calculated at different applied bias voltages. We focus on the impact that bias has on the resonant tunneling mediated by surface and quantum well states. The calculations are done in the coherent regime, using a combination of density functional theory and non-equilibrium Green’s functions, as implemented in the 
ab initio code SMEAGOL.

We conclude that, for both kinds of junction, the transmission functions depend on the applied bias voltage. In the single-barrier junction, transport mediated by resonant Fe minority surface states is rapidly destroyed by bias. In the double-barrier junction, the appearance of resonant tunneling through majority quantum well states is strongly affected by bias.

PACS numbers: 85.75.-d, 72.25.Mk, 73.40.Rw, 73.23.Ad

I. INTRODUCTION

Magnetic tunnel junctions (MTJs), consisting of a semiconducting barrier sandwiched by two ferromagnetic electrodes, are prototype nanoscale systems exhibiting spin-dependent electronic transport, and are nowadays being intensively investigated due to their potential applicability in spintronic devices based on the tunneling magnetoresistance effect (TMR). Besides their practical importance, MTJs are interesting heterostructures on their own since they allow us to test our electronic transport theories and models, and to understand the complex relationship that the electronic, magnetic, and interface structures have with novel spin-dependent transport phenomena (see Refs. 1 and references therein).

One of the main challenges in this field is to obtain larger TMR and V_{1/2} values (the bias voltage value at which the TMR drops to half its value at infinitesimal bias), both aspects being critical for device applications 2 3. As first suggested by Zhang et al 4 a decade ago, a possible route to accomplish these higher values is to use double-barrier MTJs (DBMTJs), in which a metallic slab (magnetic or not) is inserted in between the semiconducting spacer. In these junctions, whose magnetic and transport properties have been measured only very recently 5 6 7 8, and whose transport properties have been calculated mainly using free electron models or non-selfconsistently 8, the in-between metallic slab plays a dual role. First, it introduces quantum well states which can, in principle, couple to the evanescent states in the spacer, thus producing conductance resonances in some spin channel and therefore enhancing the TMR 4 5 6 7 8. Second, if it is magnetic, it introduces a spin-dependent potential energy profile which may act as an additional spin filter, again enhancing the TMR 5 9 10 11.

In particular, Fe/MgO (001) single- and double-barrier junctions are ideal systems to study and good candidates for applications, since they show very large TMR values and can be nowadays grown epitaxially with controlled interfaces. For example, T. Nozaki et al 3 6 have recently measured the TMR of Fe/MgO (001) SBMTJs and identically grown DBMTJs as a function of the applied voltage and found that the DBMTJs show larger TMR and V_{1/2} values. Similar findings have been reported in double-barrier junctions of other materials 2. Although a complete understanding of these features (particularly the larger V_{1/2} values) is still lacking, we have recently shown by 
ab initio calculations on Fe/MgO DBMTJs 11 that the spin-filter effect, introduced by the magnetic in-between Fe layers, is one of their origins. On the other hand, A. Iovan et al 7 have recently measured extremely large TMR values (an order of magnitude larger than those of identically grown MTJs) and conductance oscillations in Fe/MgO DBMTJs, originated from resonant tunneling processes mediated by spin-split quantum well states. Similar conductance oscillations in Fe/MgO DBMTJs have been reported by Nozaki et al as well 6. From the theoretical side, Y. Wang et al 9 have shown, from first principles, that the small conductance oscillations observed 6 in Fe/MgO DBMTJs originate from the majority ∆_{1}-symmetry QWS at the Γ point, which efficiently couples to a decaying state in the MgO spacer. From these examples it is clear that DBMTJs

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are promising candidates for spintronic applications, and consequently that further theoretical and experimental studies of these heterostructures are necessary to fully understand the role of QWSs and, especially, of the bias voltage on their spin-dependent transport properties.

With this aim in mind, here we calculate the coherent transmission spectra of ideal Fe/MgO (001) SBMTJs and DBMTJs at different values of the applied bias voltages. The focus is put on the impact that this bias voltage has on the transmission resonances mediated by quantum well states in DBMTJs and by surface states in SBMTJs [11, 12]. To this end, we employ the self-consistent \textit{ab initio} SMEAGOL code [13, 14], which is based on a non-equilibrium Green’s function formalism [16]. By extending the work of Y. Wang \textit{et al.} [9] that considered only the Γ point, we take into account a large number of wave vectors inside the surface Brillouin Zone. Furthermore, the transport calculations are performed self-consistently, taking full account of the non-equilibrium electronic population inside the junction induced by a finite applied bias voltage to the Fe electrodes. We have already presented the calculated characteristic curves of Fe/MgO (001) SBMTJs and DBMTJs and discussed the different transport mechanisms elsewhere [11], reaching qualitative agreement with available experimental results. Here, we would like to focus on the impact that a bias voltage has on these mechanisms, emphasizing the differences between the transmission functions in the single- and double-barrier cases.

II. CALCULATION DETAILS

Our SBMTJs consist of 2 monolayers (MLs) of MgO (001), representing 4 Å, sandwiched by two \textit{semi-infinite} bcc Fe (001) electrodes, while our DBMTJs are multilayers of the type (MgO)$_2$/Fe$_m$/MgO (001) sandwiched by the same electrodes. The number of in-between Fe monolayers is set equal to $m=2$ MLs, representing 2.87 Å. In both cases, the junctions are assumed to be periodic in the $x$-$y$ plane, being $z$ the transport direction. In this work, we restrict to the parallel magnetic configuration of the junctions (hereafter denoted $P$), in which the magnetization vector in every magnetic region is parallel to each other.

In order to account for the charge transfer and to correctly reproduce the band offset between Fe and MgO, we include in the cell for self-consistent calculations four Fe MLs belonging to the electrodes at both sides of the junction. This in enough to correctly account for charge screening inside the ferromagnet. Similar to previous calculations [8, 12, 17] the lattice constant of the electrodes is fixed to 2.87 Å and that of MgO is taken to be $\sqrt{2}$ larger. This, together with a 45° rotation of the Fe unit cell, allows epitaxial matching between Fe and MgO. In this work, the possible appearance of FeO interfacial layers as well as atomic relaxation and disorder, are not considered.

For the electronic structure of the junctions, we use norm-conserving pseudopotentials, double-zeta basis set for all the angular momenta and the generalized gradient approximation (GGA) [18] to the exchange and correlation potential. We have thoroughly checked that the band structure and the density of states of bulk Fe, bulk MgO and Fe/MgO multilayers, as well as the charge transfer and magnetic moments in the last case, are very well reproduced as compared to FP-LAPW results obtained using the highly accurate WIEN2k code [19]. We obtain a band offset (the difference between the Fermi energy $E_F$ of Fe and the valence band of MgO) of 3.51 eV, in very good agreement with previous theoretical [20] and experimental reports [21]. As well-known, density functional calculations using semi-local exchange and correlation functionals underestimate the band gap and ours are not an exception. We obtain a band gap of 5.4 eV (as compared to the experimental value of 7.8 eV [21, 22]), which agrees well with what expected from GGA [17, 20].

The ballistic transmission coefficient $T^\sigma(E, V)$ is calculated for each bias and it is given by

$$T^\sigma(E, V) = \frac{1}{V_{\text{BZ}}} \int dk_x dk_y T^\sigma(E, V, k_x, k_y)$$  \hspace{1cm} (1)

where $V_{\text{BZ}}$ is the area of the surface Brillouin zone orthogonal to the transport direction $z$. Here we assume that both spin and transverse momentum $k_z$ are conserved, an approximation that is valid for relatively thin epitaxial junctions. The $k_{\parallel}$-\textit{resolved} transmission coefficient appearing in Eq. 1 is calculated self-consistently from the non-equilibrium Green’s functions formalism in the usual way [13, 14, 16]. It is given by $T = Tr[H_\parallel G^\sigma R^\sigma]$, where for simplicity we omit the spin label $\sigma$. Here, $\Gamma_{L,R}$ are the broadening matrices describing the interaction (thus the finite lifetime) of the scattering region’s energy levels due to the interaction with the left and right electrodes, and $G^\sigma (G^0)$ is the associated retarded (advanced) Green’s function describing the one-electron dynamics inside the scattering region. The broadening matrices are calculated from the self-energies $\Sigma_{L,R}$ as

$$\Gamma_{L,R} = i \langle \Sigma_{L,R} - \Sigma_{L,R}^\dagger \rangle.$$  These in turn are obtained with the semi-analytic method described in reference [14].

The selfconsistent loop consists in starting from an initial density matrix of the scattering region (for example, the equilibrium \textit{no bias} density matrix), $\rho^{(i)}$, which gives an initial Kohn-Sham Hamiltonian $H^{(i)}[15]$. With this, the $G^{r,s}$ can be obtained [13, 16], since $\Sigma_{L,R}$ are calculated at equilibrium ($V=0$) and then rigidly shifted in energy. That is, the voltage-dependent selfenergies $\Sigma_{L,R}(E, V)$ are therefore given by $\Sigma_{L,R}(E \mp \epsilon V/2, V = 0)$. The upper (lower) sign in the first argument refers to the left (right) electrode. These retarded/advanced Green’s functions are then fed into Keldysh equation for the lesser correlation $G^< = G^r \Sigma^< G^s$ [16], from which a new density matrix is obtained. In this expression, the lesser self-energy $\Sigma^< = \Sigma^<_L + \Sigma^<_R$ is obtained from the local equilibrium
hypothesis: \( \Sigma_{E,R} = i f_{L,R} \Gamma_{L,R} \), where \( f \) is the Fermi-Dirac distribution function corresponding to the left and right electrodes. With this new density matrix obtained via the Keldysh equation, the loop starts again, until convergence is achieved. We note that the selfconsistent loop must be performed at each bias voltage (because the electron population inside the scattering region, induced by the electrodes, depends on \( V \)).

In our calculations, we use a \( 8 \times 8 \times 8 \) k-point mesh in reciprocal space to calculate the density matrix of the scattering region and a \( 150 \times 150 \times 1 \) mesh to evaluate the transmission at each bias voltage. We have carefully verified that these meshes are sufficient for converging the density matrix and the transmission.

**III. RESULTS AND DISCUSSION**

Starting with the single-barrier junction, in Fig. 1 we show the transmission spectra at zero bias voltage (upper panel) and at \( V=0.1 \) Volt (lower panel). It is seen that, at zero bias, the \( P \) minority channel presents a broad peak centered around \( E_F+0.1 \) eV. This conductance peak is the signature of a resonance mediated by the Fe-minority surface state located at that energy. We note that this surface state resonance is also present in a SBMTJ with \( n=4 \) MLs. At zero bias (or low enough bias), the Fe surface states located at both sides of the MgO barrier are approximately aligned in energy, and therefore can resonate through it. As it can be seen from the lower panel of Fig. 1, this surface state resonance is washed out by applying even a small bias voltage of 0.1 Volt. In such a situation, due to the applied bias voltage the surface states at each electrode are no longer aligned in energy. As a consequence, the resonance condition is lost and the transmission peak disappears. Therefore, as has been already shown theoretically by Runger et al [12] in Fe/MgO SBMTJs, transport calculations in which the transmission function is bias-independent (often taken as the zero-bias transmission) overestimate the contribution of surface states to the current. Here, we see a first example showing the importance of taking into account the dependence of the transmission function on the applied bias voltage.

Another interesting feature from Fig. 1 is that the \( P \) majority transmission is almost independent of bias, in marked contrast to the \( P \) minority transmission. This difference is a direct consequence of the different transport mechanisms that govern the transmission of each of the two \( P \) channels. The \( P \) minority conductance is dominated (at least for SBMTJs with thin barriers) by Fe surface states that can couple to each other directly through the MgO slab. In contrast, the \( P \) majority transmission is governed by the MgO slowly-decaying complex band of symmetry \( \Delta_1 \), that can couple efficiently to the Fe Bloch states of the same symmetry [13]. The complex bands are smooth functions of the energy (just as real bands are), and therefore the application of moderate bias voltages has no significant influence on them.

![FIG. 1: Transmission spectra of a single-barrier junction with \( n=2 \) MLs (4 Å) at zero bias (upper panel) and at \( V=0.1 \) Volt (lower panel).](image)

Going over to double-barrier junctions, in Figs. 2-4 we show the transmission spectra of the DBMTJ with \( n=2 \) MLs (4 Å) and \( m=2 \) MLs (2.87 Å), at \( V=0.005 \) Volt, 0.1 Volt and 0.15 Volt, respectively. At \( V=0.005 \) Volt (Fig. 2) the surface state resonance in the \( P \) minority channel is still observed, although the peak height is smaller than the one at zero bias shown in Fig. 1 for the single-barrier junction. That is to say, the resonant mechanism mediated by the Fe surface states can occur even in DBMTJs with very thin in-between Fe slabs, as long as the applied bias is small. As it happens in the single-barrier junction, the application of a bias voltage rapidly destroys this resonance, as it can be seen from Figs. 3 and 4 where the transmission resonance is no longer present. The origin of this feature is exactly the same as in SBMTJs, namely, the misalignment in energy of the Fe surface states at each side of the barrier.

At \( V=0.005 \) Volt (Fig. 2) and at \( V=0.15 \) Volt (Fig. 4), the \( P \) majority transmissions are very similar to each other, being increasing linear functions of the energy. Again, this feature reflects the fact that the \( P \) majority transmission is essentially governed by the complex bands of MgO. But at \( V=0.1 \) Volt (Fig. 3) the \( P \) majority transmission is radically different. It is seen that it presents a sharp peak located near \( E_F+0.075 \) eV. This peak is characteristic of resonant tunneling through a quantum well state inside the in-between Fe slab. This quantum well state has already been shown to be present by Wang et al [9], and we have studied its impact on the \( I−V \) curves of Fe/MgO (001) DBMTJs [11], reaching quantitative agreement with recent experimental results [4,7]. Therefore, from Figs. 2-4 we see that the appear-
FIG. 2: Transmission spectra of a double-barrier junction with \( n = 2 \) MLs (4 Å) and \( m = 2 \) MLs (2.87 Å) at \( V = 0.005 \) Volt.

FIG. 3: Transmission spectra of a double-barrier junction with \( n = 2 \) MLs (4 Å) and \( m = 2 \) MLs (2.87 Å) at \( V = 0.1 \) Volt.

FIG. 4: Transmission spectra of a double-barrier junction with \( n = 2 \) MLs (4 Å) and \( m = 2 \) MLs (2.87 Å) at \( V = 0.15 \) Volt.

The appearance of a transmission resonance mediated by a quantum well state is strongly dependent on the applied bias voltage. It is very interesting to note that the resonance disappears even with very slight changes in the applied bias voltage. A change in bias of 0.05 Volt is already sufficient to suppress the quantum well state resonance.

These results clearly show that, in general, it is not possible to assume that the transmission function of tunneling barriers is independent of bias, as it is usually done in non-selfconsistent calculations [8]. Both in single- and in double-barrier junctions these functions depend on bias, although the dominant transport mechanisms are completely different. In particular, in the double-barrier junction that we consider, the \( P \) minority channel behaves, qualitatively, as it does in single-barrier ones, being dominated by the resonances mediated by Fe surface states at each side of the MgO barrier. The application of a bias voltage shifts these surface states in energy, thus forbidding their direct coupling to each other. On the other hand, the \( P \) majority channel behaves differently because in double-barrier junctions, in addition to transport mediated by the complex bands of MgO, another mechanism comes into play, namely, resonant tunneling through quantum well states. Our results are just an example of how bias-dependent is this last transport mechanism. We are currently investigating these issues in more detail. In particular, we are trying to understand the origin of the bias dependence of quantum well states resonances and its relation to the potential profile inside the scattering region.

IV. CONCLUSIONS

Using realistic models of electronic structure and self-consistent coherent transport calculations based on non-equilibrium Green’s functions formalism, we have obtained the transmission spectra of a single-barrier and of a double-barrier Fe/MgO tunnel junction, at different values of the applied bias voltage. We have shown that, both in single- and double-barrier junctions, the transmission spectra depend on bias. In particular, in single-barrier junctions the application of a small bias voltage is already sufficient to suppress resonances between Fe minority surface states across the insulating barrier, a fact already discussed by Rungger et al [12]. In double-barrier junctions, the appearance of resonances
mediated by quantum well states in the majority channel is strongly dependent on the applied bias. These results bring some insight into the phenomena of resonant tunneling through epitaxial magnetic tunnel junctions at finite bias, and may be useful in the design and development of future spin-electronic devices.

This work was partially funded by UBACyT-X115, PIP-CONICET 6016, PICT 05-33304 and PME 06-117. A. M. Llois belongs to CONICET. (Argentina).

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