Tunneling magnetoresistance of Fe/ZnSe (001) single- and double-barrier junctions as a function of interface structure

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Abstract

In this contribution, we calculate the spin-dependent ballistic and coherent transport through epitaxial Fe/ZnSe (001) simple and double magnetic tunnel junctions with two different interface terminations: Zn-terminated and Se-terminated. The electronic structure of the junctions is modeled by a second-nearest neighbors \textit{spd} tight-binding Hamiltonian parametrized to \textit{ab initio} calculated band structures, while the conductances and the tunneling magnetoresistance are calculated within Landauer’s formalism. The calculations are done at zero bias voltage and as a function of energy. We show and discuss the influence of the interface structure on the spin-dependent transport through simple and double tunnel junctions.

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I. INTRODUCTION

Mixed ferromagnetic/semiconductor nanostructures are gaining an increasing interest due to their potentialities for spintronic applications, such as magnetic random access memories and magnetic field sensors (see, for example, Ref. 1). It is known that the magnetic and electronic properties of these heterostructures depend on the nature of the metal/semiconductor interface, which affects their magnetotransport properties in a significant way. 2

For example, M. Eddrief et al have recently shown in photoemission experiments that the Fe $\Delta_1$ spin-up band along the (001) direction, which is the band that couples most efficiently to the ZnSe complex bands in Fe/ZnSe (001) magnetic tunnel junctions (MTJs), is strongly modified due to Zn interdiffusion into the Fe electrodes, and that these modifications may be the origin of the very low tunneling magnetoresistance (TMR) measured in these systems. On the theoretical side, M. Freyss et al have shown that the spin polarization $P_S = (N_{up} - N_{dn})/(N_{up} + N_{dn})$ (being $N_{up/dn}$ the density of states of the majority/minority electrons) at the Fermi level of Fe/ZnSe (001) interfaces is more negative for the Zn-terminated interface than for the Se-terminated interface, and the transport calculations on Fe/ZnSe (001) single-barrier junctions done by H. C. Herper et al clearly indicate that the TMR is higher for Se-terminated interfaces than for Zn-terminated ones. These examples concerning Fe/ZnSe hybrid nanostructures, together with those of Ref. 2 for other hybrid junctions, demonstrate that the metal/barrier interfaces play a major role in determining the magnetotransport properties of MTJs.

In this contribution, we analyse the transport properties of some examples of Fe/ZnSe (001) double-barrier tunnel junctions (DMTJs) as compared to MTJs, as a function of interface termination. To do this, we calculate the conductances and the TMR of epitaxial Fe/ZnSe (001) tunnel junctions with two different interface structures: (a) Zn-terminated (both interfaces contain Fe and Zn atoms), and (b) Se-terminated (both interfaces contain Fe and Se atoms). The results obtained could be relevant for the practical use of MTJs and of DMTJs in spintronic devices.
II. SYSTEMS UNDER STUDY AND CALCULATION METHODS

Our single-barrier magnetic tunnel junctions consist of \( n \) layers of zincblende ZnSe (001) sandwiched by two BCC Fe (001) semi-infinite electrodes. We form DMTJs by inserting \( m \) layers of BCC Fe (001) in between the \( 2n \) layers of ZnSe, so that the Fe midlayers are sandwiched by \( n \) identical ZnSe layers at each side. In what follows, we will call the active region (AR) to whatever is sandwiched by the left and right semi-infinite Fe electrodes. Each ZnSe layer and Fe midlayer has a width of 0.567 nm and 0.287 nm, respectively. The junctions are periodic in the \( x-y \) plane, being \( z \) the transport direction. Fig. 1 shows, as an example, the interface structure of a Zn-terminated MTJ with \( n=1 \). The Se-terminated junctions are formed by interchanging the Zn and the Se atoms. We note that the junctions are epitaxial and that interface relaxation is not taken into account.

In the parallel configuration (\( P \)), the magnetizations of all the magnetic regions are parallel to each other. In the antiparallel configuration (\( AP \)), in MTJs the electrodes’ magnetizations are antiparallel to each other, while in DMTJs they remain parallel to each other and the Fe midlayer’s magnetization is antiparallel. We note that, since the coercive fields of the electrodes and of the midlayer are different, these magnetic configurations are experimentally achievable.\(^7\)

The electronic structure of the junctions is modeled by a second nearest neighbors \( spd \) tight-binding Hamiltonian fitted to \textit{ab initio} band structure calculations for bulk Fe and bulk ZnSe.\(^8\) When forming the junctions, the ZnSe tight-binding on-site energies are rigidly shifted to make the Fe Fermi level fall 1.1 eV below its conduction band minimum, as indicated in photoemission experiments performed on Fe/ZnSe junctions.\(^9\) Further details can be found in.\(^10\)

The ballistic conductances \( \Gamma \) are calculated using Landauer’s formalism expressed in terms of Green’s functions (see\(^10,11\)). The self-energies describing the influence of the electrodes on the active region are calculated from the electrodes’ surface Green’s functions (SGFs) in the usual way,\(^11\) while the SGFs are obtained using the semi-analytical method described in\(^12\). The tunneling magnetoresistance coefficient is defined as \( \text{TMR}=100 \times (\Gamma_P - \Gamma_{AP})/\Gamma_P \), where \( \Gamma_P \) and \( \Gamma_{AP} \) are the conductances in the \( P \) and in the \( AP \) magnetic configurations, respectively. With this definition, the TMR ranges from \(-\infty\) to 100 %. By calculating \( \Gamma \) using different numbers of parallel-to-the-interface wavevectors \( \mathbf{k}_{//} = k_x\hat{x} + k_y\hat{y} \) (see Fig.
1), we find that 5000 is enough to reach convergence. More details on the method used to calculate conductances can be found in\textsuperscript{10}.

In this work, we restrict ourselves to zero temperature, to infinitesimal bias voltage and to the coherent regime (see Ref.\textsuperscript{11}). We assume that the electron’s $k_{//}$ and spin are conserved during tunneling, since the junctions are epitaxial and the Fe midlayer is thin ($<1.8$ nm) and ordered.

III. RESULTS AND DISCUSSION

Fig. 2 shows the tunneling magnetoresistance of Se- and of Zn-terminated MTJs with $n=2$ (1.13 nm), and of DMTJs with $n=2$ and $m=6$ (1.72 nm), as a function of energy (referred to the Fermi level $E_F$). For single-barrier junctions, it is seen that the Se-terminated MTJ has a large TMR, near 80 \% on the average, while the Zn-terminated MTJ has a much lower one of 40 \%, in qualitative agreement with the results of\textsuperscript{6}. For both terminations, the dependence of the TMR on energy is rather smooth.

For double-barrier junctions, it is seen that the DMTJs’ TMR versus energy behaviors are quite different for each termination. The Se-terminated double junction shows an almost constant TMR enhancement of 20 \% with respect to the MTJ, except for certain energies at which the TMR drops abruptly. The TMR of the Zn-terminated DMTJ is also, in general, larger than the corresponding MTJ, but the TMR versus energy behavior is not as smooth as in the Se-terminated DMTJ.

The TMR versus energy behavior shown in Fig. 2 can be understood from Figs. 3 and 4, where we show the conductances in the $P$ and $AP$ configurations of MTJs (with $n=2$) and of DMTJs (with $n=2$ and $m=6$) with Se termination (Fig. 3) and with Zn termination (Fig. 4). From the lower panel of Fig. 3 (Se-terminated DMTJ) it can be observed the $AP$ conductance resonance which produces the TMR drop at $E = E_F - 0.05$ eV shown in Fig. 2. It can also be seen that the conductances exhibit an oscillatory behavior with energy, indicating the presence of resonances. Going over to the lower panel of Fig. 4 (Zn-terminated DMTJ) it is seen that the TMR drop at $E = E_F - 0.1$ eV does not have its origin in a resonance in the $AP$ conductance but that it is due to a drop in the $P$ majority conductance. It is also interesting to note that the $P$ majority conductance is a smooth function of the energy (except for the already mentioned drop), in contrast to the
$P$ majority conductance of the Se-terminated DMTJ, which shows two peaks for energies above $E_F$. These results suggest that, in the Zn-terminated DMTJ that we are considering, the spin-up quantum well states of the Fe midlayer do not couple to the evanescent states in the semiconductor, while in the Se-terminated DMTJ they do couple.

Another interesting feature, that can be observed comparing the upper panels of Figs. 3 and 4, is that the $P$ minority resonance occurring in the Se-terminated MTJ (Fig. 3), does not appear in the Zn-terminated one (Fig. 4). This conductance peak is due to the resonant coupling of the well-known Fe spin-down interface states (which are pinned at $E_F$+0.2 eV) at each interface (see, for example, Refs. 2,3,4,5). Since it is known that this interface state is also present in the Zn-terminated Fe/ZnSe interfaces5, our results indicate that in the Zn-terminated junction the interface states at each side of the barrier do not couple to each other, while in the Se-terminated case they do. We have checked that this $P$ minority conductance peak is also present in the Se-terminated MTJ with $n=4$ (2.27 nm), but that it is absent in the same MTJ with Zn termination. The $P$ minority conductances of the Se- and of the Zn-terminated junctions with $n=4$ are shown in Fig. 5. Note that, as expected, the conductance peak in the Se-terminated MTJ is not as sharp as that of the one with $n=2$. We are at present investigating, from first principles, the origin of these two behaviors, namely the absence of $P$ minority and of $P$ majority conductance peaks in the Zn-terminated MTJs and DMTJs, respectively.

Continuing with the analysis of DMTJs, we show in Table 1 the TMR values, evaluated at $E_F$ and at $E_F$±0.05 eV, of DMTJs with $n=2$ (1.13 nm) and different values of $m$, and of the MTJs with $n=2$ (and $m=0$), for the two different interface terminations considered. It is seen that, in general, the TMR values of the Se-terminated DMTJs are positive and rather large (the maximum is 98.9 %), while the ones corresponding to the Zn-terminated DMTJs are either positive (but not so large) or negative and very large (reaching −639 %). These features may result in a more pronounced bias voltage dependence of the TMR in Zn-terminated DMTJs than in Se-terminated ones, although further studies are desirable.

IV. SUMMARY

Using a realistic model for the electronic structure and accurate conductance calculations in the coherent, zero bias and ballistic regime, we showed that the spin-dependent transport
TABLE I: TMR values evaluated at $E_F$ and at $E_F \pm 0.05$ eV of DMTJs with $n=2$ (1.13 nm) and different values of $m$, together with those of the MTJ with $n=2$ (and $m=0$), for two different interface structures. TMR values are given in % and refer to $E_F−0.05\text{eV}/E_F/E_F+0.05\text{eV}$. The number in brackets is the TMR value averaged over the three energies considered.

| $m$ (n=2) | Zn-terminated | Se-terminated |
|-----------|----------------|---------------|
| 0         | 21.1/29.8/40.9 (30.6) | 66.3/73.3/67 (68.9) |
| 2         | $−639/−88.8/27.5 (−233.4)$ | $−66.1/67.3/87.8 (29.7)$ |
| 4         | 69.6/−98.9/36.2 (2.3) | 92.9/95.9/98.9 (95.9) |
| 6         | 67/31.4/65.4 (54.6) | 33.5/87.7/90.9 (70.7) |

properties of Fe/ZnSe (001) single- and double-barrier tunnel junctions are very sensitive to the Fe/ZnSe interface structure. In particular, we found that in the Se-terminated single-barrier junctions considered, the conductance of minority electrons has a peak that is absent in the Zn-terminated junctions, indicating that in the latter case the Fe spin-down interface states at each side of the ZnSe barrier do not couple to each other. We also found that the tunneling magnetoresistance of double-barrier junctions reaches higher values in the Se-terminated double junctions than in the Zn-terminated ones, in which case the TMR can reach very large and negative values.

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1 E. Y. Tsymbal, O. N. Mryasov, and P. R. LeClair J. Phys.: Condens. Matter **15**, 109 (2003)
2 C. Tiusan et al, J. Phys.: Condens. Matter **18**, 941 (2006); C. Tiusan et al, Phys. Rev. Lett. **93**, 106602 (2004); M. E. Eames and J. C. Inkson, Appl. Phys. Lett. **88**, 252511 (2006); C. Heiliger et al, Phys. Rev. B **73**, 214441 (2006); K. D. Belashchenko, J. Velev, and E. Y. Tsymbal, cond-mat 0505348 v1, 13 May 2005; N. Papanikolaou et al, Phys. Rev. B **62**, 11118 (2000)
3 M. Eddrief et al, Phys. Rev. B **73**, 115315 (2006)
4 Ph. Mavropoulos, N. Papanikolaou, and P. H. Dederichs, Phys. Rev. Lett. **85**, 1088 (2000); J.
M. MacLaren et al, Phys. Rev. B 59, 5470 (1999)

5 M. Freyss et al, Phys. Rev. B 66, 014445 (2002)

6 H. C. Herper et al, Phys. Rev. B 64, 184442 (2001)

7 T. Nozaki et al, Appl. Phys. Lett. 86, 082501 (2005); Z. M. Zeng et al, J. Magn. Magn. Mater. 303, 219 (2006)

8 D. A. Papaconstantopoulos, Handbook of the band structure of elemental solids (Plenum Press, New York, 1986); R. Viswanatha, S. Sapra, B. Satpati, P.V Satyam, B.N Dev, and D.D Sarma, cond-mat 0505451 v1, 18 May 2005

9 M. Eddrief et al, Appl. Phys. Lett. 81, 4553 (2002)

10 J. Peralta-Ramos and A. M. Llouis, Phys. Rev. B 73, 214422 (2006)

11 S. Datta, Electronic transport in mesoscopic systems (Cambridge University Press, Cambridge, 1999)

12 S. Sanvito, C. J. Lambert, J. H. Jefferson, and A. M. Bratkovsky, Phys. Rev. B 59, 11936 (1999)
FIG. 1: Interface structure along the $z$ direction of a Zn-terminated Fe/ZnSe (001) single-barrier junction with a ZnSe thickness of 0.567 nm ($n=1$). The junction is periodic in the $x$-$y$ plane and the Fe electrodes are semi-infinite.
FIG. 2: Tunneling magnetoresistance as a function of energy of MTJs with \( n=2 \) (1.13 nm) and of DMTJs with \( n=2 \) and \( m=6 \) (1.72 nm), for two different terminations. Upper panel: Se-terminated. Lower panel: Zn-terminated.
FIG. 3: Conductances in the $P$ and in the $AP$ configurations of a Se-terminated single-barrier junction with $n=2$ (upper panel), together with those of a Se-terminated double-barrier junction with $n=2$ and $m=6$ (lower panel).
MTJ $n = 2$, Zn–terminated

DMTJ $n = 2$, $m = 6$, Zn–terminated

FIG. 4: Conductances in the $P$ and in the $AP$ configuration of a Zn-terminated single-barrier junction with $n=2$ (upper panel), together with those of a Zn-terminated double-barrier junction with $n=2$ and $m=6$ (lower panel).
FIG. 5: Conductance of minority electrons in the parallel configuration of Se- and of Zn-terminated single-barrier junctions with $n=4$ (2.27 nm).