Effects of structural relaxation on calculations of the interface and transport properties of Fe/MgO(001) tunnel junctions

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The relaxation of the interface structure of Fe/MgO(100) magnetic tunnel junctions predicted by density-functional theory depends significantly on the choice of exchange and correlation functional. Bader analysis reveals that structures obtained by relaxing the cell with the local spin-density approximation (LSDA) display a different charge transfer than those relaxed with the generalized gradient approximation (GGA). As a consequence, the electronic transport is found to be extremely sensitive to the interface structure. In particular, the conductance for the LSDA-relaxed geometry is about 1 order of magnitude smaller than that of the GGA-relaxed one. Surprisingly, the effect of the exchange and correlation potential within both the LSDA and the GGA has a little effect on the calculated transmission coefficient when applied to the same Fe/MgO/Fe (001) geometry. The high sensitivity of the electronic current to the details of the relaxed interface might explain the discrepancy between the experimental and calculated values of magnetoresistance.

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I. INTRODUCTION

Tunnel magnetoresistance (TMR) is the change in the electric resistance of a magnetic trilayer made of two ferromagnetic metallic electrodes separated by an insulating spacer when the mutual orientation of the magnetizations of the electrodes changes from parallel alignment (PA) to antiparallel alignment (AA). The figure of merit for the effect is the TMR ratio, defined as

$$\mathrm{TMR} = \frac{G_{\mathrm{PA}} - G_{\mathrm{AA}}}{G_{\mathrm{AA}}},$$

where G_{PA} (G_{AA}) is the conductance for the PA (AA) configuration. Among the possible material combinations Fe/MgO(100) magnetic tunnel junctions (MTJs) have attracted much attention, since extremely large TMR ratios were theoretically predicted on the basis of symmetry-driven spin filtering from the tunnel barrier.[1] These have been now demonstrated experimentally[2–4] and the symmetry filtering effect is now widely accepted. However, the measured TMR ratios are usually significantly lower than those predicted by *ab initio* methods.

The origin of such a discrepancy is currently a matter of debate. On the theory side, interface resonance states (IRSs) located around the Fermi energy (E_F) are important for the zero-bias transport[5] and are usually difficult to describe accurately. Moreover the MgO bandgap is significantly underestimated by local spin-density approximation (LSDA) and generalized gradient approximation (GGA) so that the barrier height might not be accurately predicted. On the experimental side, the quality of Fe/MgO MTJs depends on the preparation methods. The 4% lattice mismatch between Fe and MgO produces dislocations and significant relaxation at the interface. In addition, in typical growth conditions the Fe layers close to the interface might be partially oxidized.[6–8] Many theoretical results have been published concerning the influence of interfacial structure on the electronic transport properties,[9–12] but little effort was devoted to the effect of the atomic relaxation and electron-electron correlation on the tunneling magnetoresistance. Notice, however, that in the literature whether or not the Fe/MgO interface is oxidized is controversial. Several papers claimed that the interface is not oxidized,[13–17] others claimed that the amount of oxidation at the interface can be tuned by the growth condition and that it might indeed be possible to grow nonoxidized ideal interfaces.[6,18,19] Theoretical calculations claimed that the transport properties can change drastically if such an FeO layer is present at the Fe/MgO interface.[11,20,21]

Several different geometries for the Fe/MgO interface have been proposed theoretically, based on relaxing $[\mathrm{Fe}]_n/[\mathrm{MgO}]_m$ supercells, consisting of n Fe and m MgO monolayers (MLs) subject to various constraints. In Ref. 1 LSDA relaxation for a $[\mathrm{Fe}]_5/[\mathrm{MgO}]_5$ superlattice, in which the in-plane lattice constant is held at the LSDA value for bulk Fe, yields a Fe-O distance of 2.17 Å. In contrast an early LSDA calculation for one Fe ML on MgO substrate predicted an Fe-O distance of 2.3 Å.[22] Likewise a GGA study for one MgO ML on a Fe slab returns a Fe-O distance of 2.21 Å and a separation between the first and the second Fe MLs, 6% smaller than that of bulk Fe.[10] How relevant calculations for single MLs on surfaces are for Fe/MgO MTJ is however not clear and there is still disagreement between experiments[23] and theory.[22] These controversies over the correct interface structure bring the question of how the differ-

TABLE I. The interface structure of a $[Fe]_{10}/[MgO]_6$ superlattice obtained by LSDA and GGA relaxation. The structural parameters for the unrelaxed structure are taken from Ref. 1. The middle two Fe layers are frozen at a separation of 1.43 Å and the experimental lattice constant for bulk Fe is used for the in-plane lattice constant. d_n indicates the separation (in Å) between layers from Fe/MgO the surface ($n=0$), with the index $n>0$ ($n<0$) label MgO (Fe) layers.

DFT	d_{-4}	d_{-3}	d_{-2}	d_{-1}	d_0	d_1	d_2	d_3
Unrelaxed	1.433	1.433	1.433	1.433	2.160	2.026	2.026	2.026
LSDA	1.297	1.313	1.343	1.120	2.002	2.130	2.119	2.119
GGA	1.380	1.414	1.427	1.350	2.219	2.199	2.177	2.185

ent interface geometries affect the electronic and transport properties. Our work aims at answering this question.

II. COMPUTATIONAL DETAILS

We perform structural relaxations for a $[Fe]_{10}/[MgO]_6$ superlattice with both the LSDA and GGA (Ref. 24) functionals by means of the projector augmented wave (PAW) method[25] as implemented in the VASP code.[26] Charge transfer and magnetic moments are calculated for the differently relaxed structures with Bader analysis[27] using the LSDA densities. A 12×12 in-plane **k**-point grid and an energy cutoff of 400 eV are employed to converge the charge density. As benchmark we find for bulk Fe a lattice constant of 2.75 Å and a bulk modulus of 2.68 Mbar, both in good agreement with other all-electron LSDA calculations.[28] Then the transport properties are computed with the SMEAGOL (Ref. 29) code, which combines the nonequilibrium Green's functions (NEGF) formalism with density-functional theory (DFT).[30] The GGA of Perdew, Burke, and Ernzerhof (PBE) (Ref. 31) functional and a 400×400 **k**-point grid are used for calculating the transmission coefficient.

III. ELECTRONIC STRUCTURE OF THE INTERFACE

The layer spacings at the Fe/MgO interfaces obtained after structural relaxation are summarized in Table I. These have been obtained by keeping fixed the in-plane lattice constant, set equal to the experimental value for Fe, and the atomic position of the two most internal Fe MLs. In the table we report the obtained layer spacings along the stack direction d_n, where d_0 represents the Fe-O distance at the Fe/MgO interface and $n>0$ ($n<0$) refers to separation between MgO (Fe) MLs. In the table we also report the unrelaxed structural parameters taken from Ref. 1.

LSDA relaxation gives a Fe-O distance of 2.0 Å. This is significantly different from 2.16 Å of the GGA-unrelaxed structure. The only available experimental results are for MgO on top of the Fe surface and Fe/MgO/Fe (001) with the presence of FeO at the interface,[7,8] where the FeO/MgO distance at the interface is about 2.20 Å. The LSDA relaxation also predicts that the spacing, d_{-1}, between the first two Fe layers next to the Fe/MgO interface is drastically reduced to 19% of that of bulk Fe. The changes in layer spacing of the MgO barrier, on the other hand, are relatively small. Interestingly, the GGA results are significantly different from the LSDA ones. In particular, GGA returns a Fe-O distance of 2.22 Å in agreement with experiment[7,8] and the reduction in d_{-1} is less pronounced than that for the LSDA. Our GGA-relaxed structure is in good agreement with other GGA studies[10,32] and rather close to the reference unrelaxed interface structure.

Figure 1 demonstrates that the charge transfer between Fe and MgO is very sensitive to the interface structure. The Bader analysis for the LSDA-relaxed geometry predicts that the first two Fe MLs at the Fe/MgO interface acquire a similar amount of electron charge, $\Delta\mathcal{Q}$, while for the GGA-relaxed and unrelaxed structures it indicates that the interface Fe ML loses electrons and the second Fe ML gains a significant amount of charge. In all cases MgO loses electrons to Fe. Similar charge transfer from MgO to neighboring metallic layers was predicted before for Rh/MgO interfaces.[33] For the LSDA-relaxed structure we predict a net electron transfer from MgO to Fe of up to about 0.06 electrons per atom. This is larger than the one for the other two geometries. The Bader charges for O atoms at the interface and in the middle of the MgO barrier are, respectively, 7.64 and 7.71. This indicates that charge is transferred to Fe mainly from O. Finally, since the GGA-relaxed structure is close to the unrelaxed one, the charge transfer is also similar.

The local magnetic moments of the Fe atoms [Fig. 2(a)] are calculated by integrating the spin densities in the domains determined by charge densities resulting from the Bader analysis. These are similar for the GGA-relaxed and unrelaxed structures, both presenting a significant enhancement of the Fe magnetic moment at the MgO interface. For

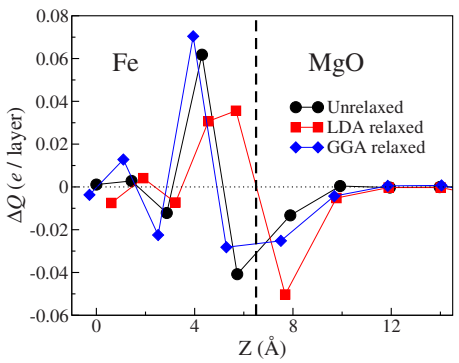

FIG. 1. (Color online) Charge transfer, $\Delta\mathcal{Q}$, for a $[Fe]_{10}/[MgO]_6$ superlattice as calculated from Bader analysis. Net charge transfer is obtained by subtracting the atomic charges of bulk Fe and bulk MgO from the Bader atomic charges in the Fe/MgO superlattice. The vertical dashed line denotes the Fe/MgO interface.

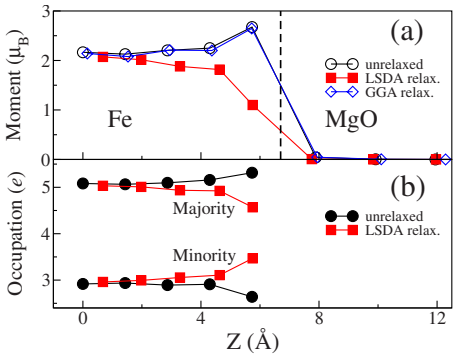

FIG. 2. (Color online) (a) The magnetic moment and (b) spin-decomposed electron occupation for a $[Fe]_{10}/[MgO]_6$ superlattice. These are calculated using Bader analysis for the valence electrons. The vertical dashed line in the top panel denotes the Fe/MgO interface.

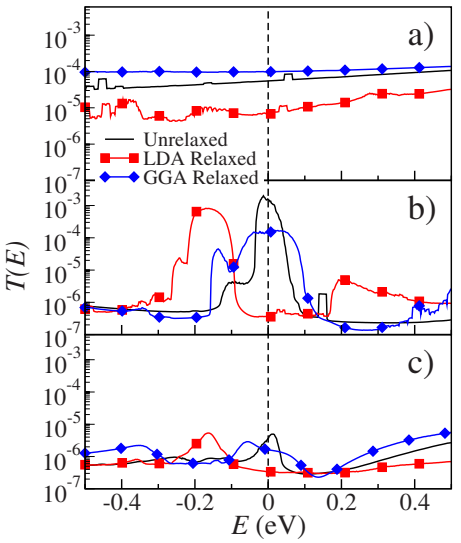

FIG. 3. (Color online) Transmission $T(E)$ for (a) PA majority spins, (b) PA minority spins, and (c) for the AA of a Fe/MgO/Fe MTJ. The transmission $T(E)$ for the unrelaxed structure is calculated within the GGA. The Fermi energy is set to zero.

the LSDA-relaxed structure however such interfacial magnetic moment is dramatically suppressed. This is only $1.10\mu_B$, to be compared with $2.65\mu_B$ and $2.68\mu_B$, respectively, for the GGA-relaxed and unrelaxed geometries. Since ΔQ is much smaller than the change in the magnetic moment, the dramatic decrease in interface magnetic moment in the LSDA calculations is caused by an internal electron redistribution between the majority- and minority-spin subbands. This is demonstrated by the spin-decomposed electron occupation [Fig. 2(b)]. Since in the LSDA relaxation d_{-1} is significantly shorter than that for GGA relaxation, the suppression of the magnetic moment is expected, based on the fact that many magnetic materials undergo magnetic collapse under pressure.[34]

IV. TRANSPORT PROPERTIES

The effects of the interface structure on the electronic transport are studied with SMEAGOL.[29] At zero bias the conductance, G, is simply $G=e^2/hT(E_F)$, where e is the electron charge, h the Planck's constant, and $T(E_F)$ is the transmission coefficient calculated at E_F. The transmission coefficient $T(E_F)$ calculated within the GGA for the unrelaxed structure and for the LSDA and GGA-relaxed structures is presented in Fig. 3. The transmission for majority spins in the PA depends weakly on the energy but it is reduced by about 1 order of magnitude for the LSDA-relaxed structure when compared to both the GGA and the unrelaxed ones. In contrast the minority-spin channel for PA is dominated by a peak at around E_F. This originates from IRSs at the Fe/MgO interface.[35] For the LSDA-relaxed structure these peaks are shifted by about 0.2 eV towards lower energies with respect to the GGA-relaxed case. A similar shift is found for both spins in the AA configuration. This shift leads to a large change in the low bias conductance. As is shown in Table II, the total conductance of the LSDA-relaxed structure is about 30 times smaller than that of the GGA-relaxed structure for the PA. Although the magnetic moment and charge transfer are similar for the GGA-relaxed structure and the unrelaxed structure, the transmission coefficients for the two structures are significantly different. The GGA-relaxed structure shows less pronounced and wider peaks in the transmission coefficient than those of the unrelaxed structure. This results in the conductance of GGA-relaxed structure being five times smaller than that of the unrelaxed one.

Since the peak in transmission at around E_F originates from the IRSs at the Fe/MgO interface, the shift in energy of the transmission coefficient for the LSDA-relaxed structure relative to that of the GGA-relaxed one is the result of a high sensitivity of IRSs to the interface geometry. In Fig. 4 the density of states (DOS), projected on the interface Fe layer, is shown for both spins. For the LSDA-relaxed structure this is shifted by about 0.2 eV with respect to that of the GGA-relaxed structure, which causes the corresponding energy shift of $T(E)$. Since the unrelaxed and GGA-relaxed structures are close to each other, the general features of the projected DOS for the two are also similar. In order to get a better insight into the role of relaxation, in Fig. 5 we show the **k**-resolved projected DOS at E_F for the interfacial Fe layer. The results for the GGA-unrelaxed and GGA-relaxed structures are rather similar, but they differ remarkably from that for the LSDA-relaxed case. In general the LSDA-

TABLE II. The TMR and zero-bias conductance for PA and AA Fe/MgO/Fe MTJ. GGA PBE is used for the unrelaxed and relaxed structures. The results are shown for the unrelaxed, the LSDA-relaxed, and for the GGA-relaxed structures. The experimental TMR is for a Fe/MgO/Fe MTJ with a 2.3-nm-thick MgO barrier and a temperature of 20 K (Ref. 3).

	G_{PA}	G_{AA}	TMR (%)
GGA unrelaxed	1.21×10^{-3}	7.18×10^{-6}	17300
LSDA relaxed	7.32×10^{-6}	6.74×10^{-7}	986
GGA relaxed	2.38×10^{-4}	3.23×10^{-6}	7270
Expt.			247

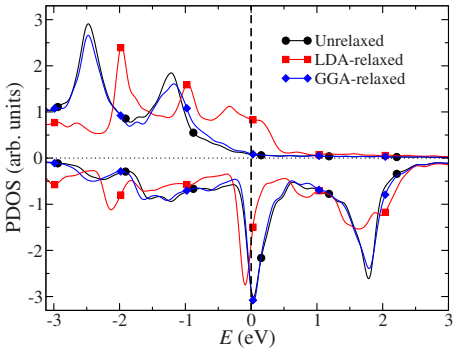

FIG. 4. (Color online) The projected density of states (PDOS) of the interface Fe 3*d* orbitals in a $[Fe]_{10}/[MgO]_6$ junction. Positive- and negative-projected DOSs represent the majority and minority components, respectively. The PDOS for the unrelaxed structure is calculated within the GGA.

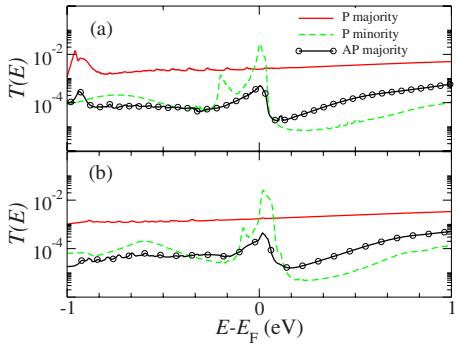

FIG. 6. (Color online) Comparison of the transmission coefficient $T(E)$ for unrelaxed atomic positions of $[Fe]_8/[MgO]_4/[Fe]_8$ calculated both within (a) the LSDA and (b) the GGA. It can be seen that the effect of the different treatments of the correlation is small at the vicinity of the Fermi level.

relaxed geometry shows a substantial reduction in the spin-polarization, demonstrated by the fact that the **k**-resolved DOS is similar for the majority and minority spins. This is expected to produce a reduction in spin filtering and as a consequence a reduction in TMR. It is also important to note that the main differences between LSDA-relaxed and GGA-relaxed (and GGA-unrelaxed) geometries are more evident around Brillouin zone center, i.e., for electrons with a large linear momentum perpendicular to the MgO barrier and therefore with a larger transmission probability.

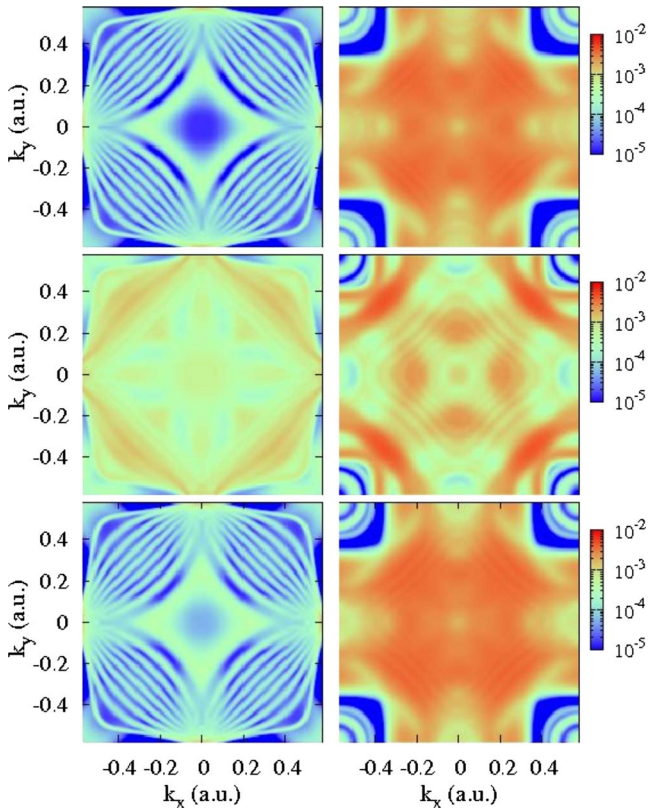

FIG. 5. (Color online) The **k**-resolved projected density of states for the interface Fe ML in a Fe/MgO/Fe junction. GGA-unrelaxed, LSDA-relaxed, and GGA-relaxed structures are shown in the top, middle, and bottom panels, respectively. The left and right panels are for majority and minority spins, respectively.

The changes in conductance lead to very different values for the TMR. Our calculated values for the TMR are shown in Table II. The LSDA-relaxed structure has a TMR of about 10^3, which is much smaller than the TMRs of the other two structures and in much closer agreement to experiments.[3] The dramatic reduction in TMR for the LSDA-relaxed structure originates from two features: the shift to lower energies of the IRSs transmission peak and the large reduction in the majority-spin transmission for PA. These can be both associated with a loss of spin polarization of the first Fe layer at the Fe/MgO interface in the LSDA-relaxed case. To show whether the huge effect on the TMR is due to the atomic relaxation alone or to the combined effect of the different functionals and relaxation, we have calculated the transmission coefficient of $[Fe]_8/[MgO]_4/[Fe]_8$ for the unrelaxed atomic positions both within the GGA and the LSDA as shown in Fig. 6. It is clear from this figure that the transmission coefficient remains almost unchanged at the vicinity of the Fermi level. The change in the TMR is less than 1%. The small difference in the peak positions of the LSDA and GGA results is due to the small differences in the electronic structure calculated using the two functionals. We are therefore confident that most of the change in the transmission coefficients is due to atomic relaxation effects on the interface electronic structure as can be seen in Figs. 2 and 4 where the magnetic properties at the interface and Fermi surface are altered due to atomic relaxations.

V. CONCLUSION

Motivated by the significant difference between theory and experiments on the reported TMR values in Fe/MgO/Fe(100) MTJs, we studied the interface structure and its effects on the electronic and transport properties by means of DFT and the NEGF formalism. In general LSDA- and GGA-relaxed interfacial geometries are rather different, yielding different charge transfer and interfacial magnetism. In particular the local magnetic moment of the interfacial Fe ML is severely suppressed for the LSDA-relaxed structure and en-

hanced for GGA-relaxed ones. These differences are reflected in the transport properties. In particular the differences at the interface determine the energy position of resonances in the transmission for the minority spins. These resonances are caused by IRSs and largely determine the zero-bias conductance. As a consequence the TMR is rather sensitive to the interfacial structure. These features can partially explain the disagreement between theory and experiments and are expected to apply to many other systems relevant for spintronics. As a by-product, we have shown that for the same structure of the interface, the GGA and the LDA functionals produce very similar transmission coefficients and TMR.

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