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Effect of interface states on spin-dependent tunneling in Fe/MgO/Fe tunnel junctions

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The electronic structure and spin-dependent tunneling in epitaxial Fe/MgO/Fe(001) tunnel junctions are studied using first-principles calculations. For small MgO barrier thickness the minority-spin resonant bands at the two interfaces make a significant contribution to the tunneling conductance for the antiparallel magnetization, whereas these bands are, in practice, mismatched by disorder and/or small applied bias for the parallel magnetization. This explains the experimentally observed decrease in tunneling magnetoresistance (TMR) for thin MgO barriers. We predict that a monolayer of Ag epitaxially deposited at the interface between Fe and MgO suppresses tunneling through the interface band and may thus be used to enhance the TMR for thin barriers.

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Magnetic tunnel junctions (MTJs) are miniature devices which consist of two ferromagnetic electrodes separated by an insulating barrier. These junctions are made in such a way that their magnetization may be switched between parallel and antiparallel states under the influence of external magnetic field. This switching is accompanied by an abrupt change of the electric conductance of the MTJ.[1] MTJs aroused much attention due to their potential application in magnetic random-access memories and magnetic field sensors. In practical terms, the figure of merit is the tunneling magnetoresistance (TMR), which is usually defined as $\mathrm{TMR}=(G_{\mathrm{P}}-G_{\mathrm{AP}})/G_{\mathrm{AP}}$, where G_{P} and G_{AP} are the conductances measured when the electrodes are magnetized parallel or antiparallel to each other. Recent reviews of spin-dependent tunneling in MTJs may be found in Refs. 2 and 3.

Since the first observation of reproducible TMR,[4] the majority of measurements were performed for amorphous or polycrystalline barriers, most commonly Al_2O_3. The highest TMR values achieved for Al_2O_3 barriers were about 70% at room temperature.[5] Meanwhile, theoretical calculations based on layer Korringa-Kohn-Rostoker (KKR) (Ref. 6) and tight-binding[7] methods predicted that much larger TMR values may be obtained for coherent tunneling in epitaxial Fe/MgO/Fe(001) junctions due to strong spin filtering. The latter is enforced by the wave-function symmetry and its relation to the complex band structure of the barrier.[8] Very large TMR values exceeding 200% were indeed measured for such junctions by Parkin *et al.*[9] and Yuasa *et al.*[10] Recently, a more accurate calculation[11] based on the full-potential linear augmented plane wave method confirmed the conclusions of Refs. 6 and 7.

For device applications it is critical to make the tunneling barrier as thin as possible in order to match the resistance of MTJs to other electronic components. Measurements for epitaxial Fe/MgO/Fe junctions show, however, that TMR decreases precipitously for barrier thickness below 2 nm.[10] A detailed characterization of the MgO structure grown on Fe(001) single crystals demonstrates a pseudomorphic growth of MgO up to 6 monolayers (ML) ($\approx$1.2 nm), with misfit dislocations being formed for thicker films.[12] The two latter experimental observations suggest that in the range of MgO thickness at which one might expect a ballistic tunneling mechanism for conduction with no contribution from defect scattering, TMR drops down with decreasing barrier thickness. The origin of this behavior is unknown. Also, these experimental facts are in disagreement with large values of TMR calculated for thin MgO barriers.[6,7]

In this paper we demonstrate that the reduction of TMR in epitaxial Fe/MgO/Fe(001) junctions at small barrier thickness is controlled by the minority-spin interface band. The presence of this band was experimentally proven by Tiusan *et al.*[13] We show that the transmission through this resonant channel is enhanced dramatically at small barrier thicknesses making a large contribution to the conductance in the antiparallel configuration and to the minority-spin conductance in the parallel configuration. The latter is, however, so sensitive to the mismatch in the potential at the two interfaces that it is, in practice, destroyed by disorder and/or applied bias. This explains the sizable decrease in TMR for thin MgO barriers which is observed experimentally.[10] We predict that a monolayer of Ag epitaxially deposited at the interface between Fe and MgO suppresses tunneling through this interface band and may thus be used to enhance TMR for thin barriers. This provides a new way to make MTJs with a low resistance and high TMR that are required for device applications. In addition, Ag interlayers protect the ferromagnetic electrodes from oxidation which is detrimental to TMR.[14]

We calculate the electronic structure and tunneling conductance of Fe/MgO/Fe(001) MTJs with or without Ag interlayers using the tight-binding linear muffin-tin orbital method (TB-LMTO) in the atomic sphere approximation[15] (ASA) and the local density approximation for the exchange-correlation energy. We use the full-potential LMTO (FP-LMTO) method[16] to check the correctness of ASA in describing the band structure of the MTJ. The principal-layer Green's function technique is applied to calculate the conductance.[17] The atomic structure of the Fe/MgO/Fe junctions is taken from Ref. 6. The ASA spheres are chosen as described in Ref. 18. The quality of this choice of spheres is tested against our FP-LMTO calculations[19] for the MTJ with 4 ML of MgO. In general, we find very good agreement between the ASA and FP results. In particular, the band offset between Fe and the deep MgO layers is reproduced very well; the Fermi level E_F lies approximately 3.4 eV above the MgO valence band maximum. Because the MgO band gap is

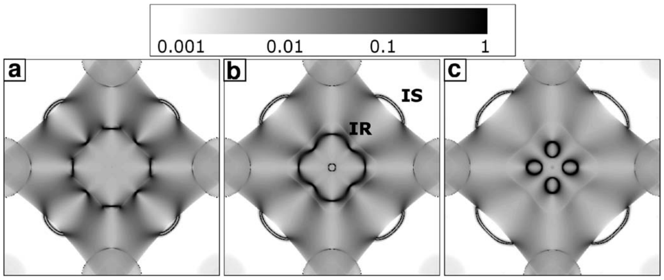

FIG. 1. Normalized minority-spin $\mathbf{k}_{\parallel}$-resolved DOS at the interfacial Fe layer in Fe/MgO/Fe(001) MTJ for three values of energy: (a) 0.02 eV below E_F, (b) at E_F, and (c) 0.02 eV above E_F. The scale is logarithmic. Interface states and resonances are marked as IS and IR in panel (b).

quite large and the metal-induced DOS quickly decays into the barrier, the band offset is almost constant in the entire range of MgO thicknesses that we studied.

The presence of the interface band can be visualized using the local density of states (DOS) resolved by transverse wave vector $\mathbf{k}_{\parallel}$. Figure 1 shows the minority-spin $\mathbf{k}_{\parallel}$-resolved DOS at and around the Fermi energy, for the interface Fe layer in a Fe/MgO/Fe(001) junction. The dark curves seen in this figure reveal the interface band, which is absent in bulk Fe. This interface band can also be seen in the energy-resolved DOS as the narrow peak near the Fermi level for minority-spin electrons (see, e.g., Fig. 1a in Ref. 13 and Fig. 3b in Ref. 6).

It is known that properties of interface (surface) states depend on whether or not they are coupled to the bulk states.[20] In Fig. 1 the interface states located about one-quarter of the Brillouin zone width from the $\bar{\Gamma}$ point are interface resonances: They lie within the continuum of bulk Bloch states and therefore have a finite linewidth. On the other hand, the states forming two parallel curves in the corners of the Brillouin zone (inside the white regions) are pure interface states. In the white regions the DOS is zero in the bulk, and the interface states have zero linewidth. To resolve these states, we added an imaginary part of 10^{-5} Ry to the energy. The two parallel bands correspond to bonding and antibonding combinations of the interface states localized at the two sides of the barrier.[21] Near the points where these bands enter the bulk continuum and become resonances one can see strong peaks in the interface DOS, similar to those predicted within a simple tight-binding model:[22]

In the single-particle approximation the interface states projected into bulk band gaps do not contribute to the tunneling conductance. Possible ways to include the contribution of such states were suggested in Refs. 23 and 24. In our case, however, the interface resonances lie much closer to the $\bar{\Gamma}$ point compared to the pure interface states. Therefore, the resonances dominate the conductance, and the use of the single-particle approximation does not lead to appreciable errors.

A notable feature of the interface band at the Fe/MgO interface is its weak dispersion. This causes a significant change in the location of this band within the first Brillouin zone when energy is shifted by a tiny amount of 0.02 eV, as is seen in Fig. 1. This feature makes any calculations of the interface states in Fe/MgO/Fe unreliable in terms of their Fermi level intercepts. It is very likely that this particular feature of the interface states is the reason why earlier calculations based on different methods[6,7,11] result in very dissimilar shapes of the minority-spin conductance plotted as a function of $\mathbf{k}_{\parallel}$.

Panels (a)–(c) in Fig. 2 show the spin-resolved transmission for the MTJ with 4 ML of MgO for parallel and antiparallel magnetization. As is clearly seen from panel (b), the resonant interface band enhances the transmission in the minority-spin channel. This enhancement is most pronounced for small barrier thicknesses, because the interface band lies away from the $\bar{\Gamma}$ point, and therefore the resonant contribution to the transmission decays faster with barrier thickness compared to the nonresonant contribution. We find that for MgO thicknesses smaller than 6 ML the contribution from minority-spin electrons in the parallel configuration becomes higher than that from majority-spin electrons. We note

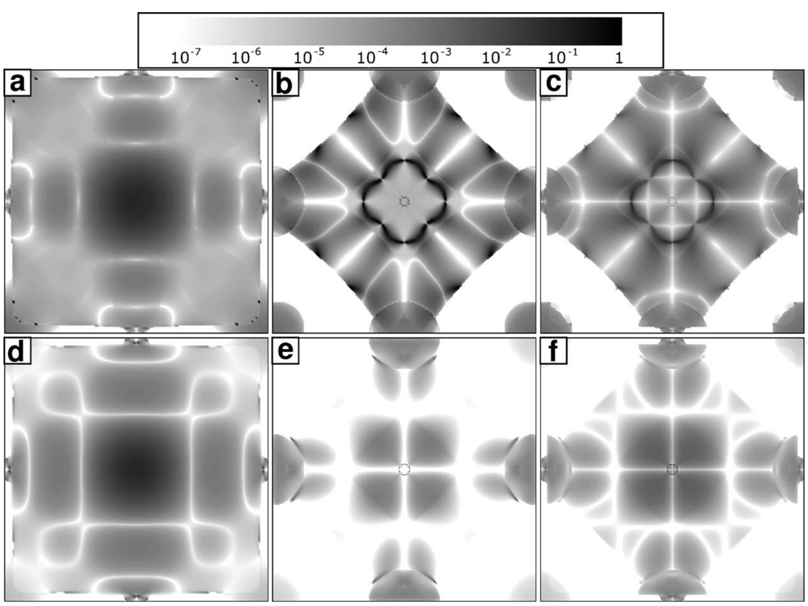

FIG. 2. Transmission probability as a function of $\mathbf{k}_{\parallel}$. (a)–(c), Fe/MgO/Fe junction with 4 MgO ML; (d)–(f), same junction with Ag interlayers; (a) and (d), majority spins; (b) and (e), minority spins; (c) and (f), antiparallel configuration (per spin).

that in the calculation by Butler *et al.*[6] this crossover does not occur down to 4 ML of MgO, although the similar tendency is clearly seen from Fig. 16 in that paper. This disagreement likely results from the interface band crossing of the Fermi level at a larger distance from the $\bar{\Gamma}$ point.

An important property of the minority-spin interface resonances is that they strongly contribute to the conductance in the parallel configuration *only for ideal, symmetric junctions, and only at zero bias.* Indeed, it is seen in Fig. 1 that the interface DOS for these resonances exceeds the DOS for neighboring regions of the surface Brillouin zone by one to two orders of magnitude. Therefore, the interface resonances generate large tunneling current only if they match similar resonances at the other side of the barrier. As follows from Fig. 1, a bias voltage of the order of 0.01 eV is sufficient to destroy this matching even for ideal epitaxy. We checked this by calculating the conductance for a small bias voltage using the surface transmission function method introduced in Ref. 25. As expected, at 0.02 eV bias voltage the conductance becomes fully dominated by majority-spin electrons. Disorder would also tend to break the matching of the interface resonances even at zero bias. Therefore, we argue that in real Fe/MgO/Fe MTJs the minority-spin channel in the parallel configuration is closed.

Unlike the parallel configuration, the interface resonances *do* contribute to the conductance in the antiparallel configuration, where they tunnel into majority-spin states of the other electrode. The latter have no fine structure in the Brillouin zone, and hence the conductance is insensitive to a potential mismatch at the two interfaces which may occur in reality. The enhanced contribution of interface resonances, which is clearly seen in Fig. 2(c), leads to the decrease of TMR at low barrier thickness. We emphasize that although the exact location of the interface resonances is not determined accurately due to intrinsic limitations of the density functional theory, their presence at the Fermi level[13] inevitably results in the reduced TMR at small barrier thickness.[10]

These features are evident in Fig. 3 which shows the conductance and TMR as a function of barrier thickness. To make the figure clearer, we used the definition of TMR that varies between −1 and 1: $R=(G_P-G_{AP})/(G_P+G_{AP})$. In the parallel configuration the majority-spin conductance is controlled by the Δ_1 band which dominates at large barrier thickness making TMR very large.[6] Below 6 ML of MgO, however, minority-spin electrons overcome the contribution from majority-spin electrons due to the contribution from the interface resonances. In the antiparallel configuration the spin conductance decreases faster than the majority-spin conductance in the parallel configuration, because it is dominated by the same interface resonances located away from the $\bar{\Gamma}$ point (see Fig. 1). As was justified above, for real MTJs the minority-spin conductance in the parallel configuration can be disregarded in the calculation of TMR. This leads to the increase of TMR with increasing the barrier thickness. A similar behavior is observed experimentally until the barrier thickness exceeds approximately 1.5 nm[10] which corresponds to 7–8 ML of MgO. At larger thicknesses the rate of decay for the parallel and antiparallel conductance becomes essentially identical. This crossover may be due to the loss of

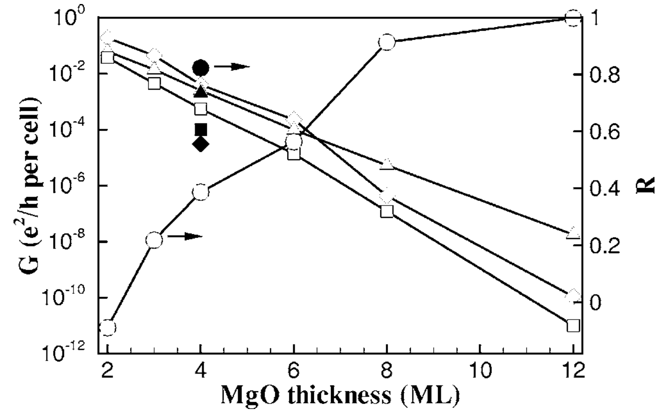

FIG. 3. Conductance (left axis) and TMR (right axis) vs barrier thickness for Fe/MgO/Fe junctions (open symbols). Triangles, majority spin, parallel configuration; diamonds, minority spin, parallel configuration; squares, each spin, antiparallel configuration; circles, TMR ratio *R*, calculated disregarding minority spin in the parallel configuration (see text). Solid symbols: conductance and TMR for a Fe/Ag/MgO/Ag/Fe junction.

$\mathbf{k}_\parallel$ conservation induced by subbarrier scattering on defects, which causes tunneling electrons to diffuse over the surface Brillouin zone. The epitaxial junction model is inapplicable in this regime.

In order to enhance TMR for thin MgO barriers we propose to use thin epitaxial Ag interlayers deposited at the Fe/MgO interfaces. Since the lattice parameter of Ag is close to both Fe and MgO lattice parameters, Ag can be deposited epitaxially on Fe(001),[26] Fe can be grown on Ag,[28] and Ag on MgO.[29] Therefore, epitaxial Fe/Ag/MgO/Ag/Fe(001) tunnel junctions are feasible. It is known that an epitaxial Ag overlayer on Fe(001) surface notably modifies the electronic structure of the surface states,[26] and it is natural to expect similar changes for the Fe/MgO interfaces where Fe and MgO interact only weakly. If the minority-spin interface DOS is reduced by Ag, the antiparallel conductance will be suppressed. On the other hand, the majority-spin conductance should not strongly be affected due to almost perfect transmission through the Fe/Ag(001) interface.[27] This is the rationale for using Ag interlayers.[30]

We place 1 ML of Ag atoms on each Fe(001) electrode in the fourfold hollow sites. The 4 ML MgO barrier is inserted between Ag-terminated electrodes so that O atoms at the interfacial ML of MgO lie above the Ag atoms. This interface structure is considered the most stable for Fe/Ag(001) and Ag/MgO(001) interfaces.[26,31] To find the equilibrium interlayer distances, we relax the atomic structure of the MTJ using the pseudopotential plane-wave method[32] implemented within the Vienna Ab Initio Simulation Package (VASP).[33] The generalized gradient approximation[34] is used for the exchange-correlation energy. We find a 5.2% reduction in the Fe interlayer distance at the interface, the distance between the interface Fe and Ag layers being 1.88 Å, and the distance between Ag and MgO layers being 2.76 Å.

Figures 2(d)–2(f) show the $\mathbf{k}_\parallel$- and spin-resolved conductance of Fe/MgO/Fe junctions with Ag interlayers. Not unexpectedly, the majority-spin conductance is weakly affected by the Ag interlayers, whereas the minority-spin conductance and the spin conductance in the antiparallel configuration change dramatically. The most pronounced difference for the latter two is the disappearance of the interface resonances that dominated the conductance of the Fe/MgO/Fe junction with no Ag interlayers [compare Figs. 2(b) and 2(c) and Figs. 2(e) and 2(f)]. This strong change occurs due to the Fe–Ag hybridization which makes the interface resonant band more dispersive and removes the Fermi level crossing responsible for the highly conductive resonant states. A careful examination of the band structure shows that the interface resonant band still crosses the Fermi level very close to the $\bar{\Gamma}$ point [an obscure circular feature in Figs. 2(e) and 2(f)], but due to its dispersive nature the interface DOS is small. As a result, this band crossing contributes 30% of the total minority-spin conductance in the parallel configuration, and only about 7% of the conductance in the antiparallel configuration. The significant reduction of the conductance in the antiparallel configuration leads to a dramatic enhancement of the TMR ratio from 0.39 to 0.82 (see Fig. 3). Thus, Ag interlayers practically eliminate the contribution from the interface resonances and enhance TMR for thin barriers.

In conclusion, we have found that interface resonant states in Fe/MgO/Fe(001) tunnel junctions contribute to the conductance in the antiparallel configuration and are responsible for the decrease of TMR at a small barrier thickness, which explains the experimental results of Yuasa *et al.*[10] Depositing thin Ag interlayers at the Fe/MgO interfaces is an efficient and practical way to suppress the tunneling through these resonant states and thereby to enhance the TMR for thin barriers.

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[18] For all the Fe atoms we use the sphere radius of 2.667 a.u. providing space filling in bulk Fe. Inside MgO we insert an empty sphere (ES) in the center of each cubic pore formed by 4 Mg and 4 O atoms. The radii are taken to be 2.202 a.u. for Mg, 1.811 a.u. for O and 1.721 a.u. for ES, providing space filling in bulk MgO. ESs are also placed at the Fe/MgO interface exactly above the ESs inside MgO; their position is adjusted to minimize overlaps. The radii of these interfacial ESs are set to be 1.761 a.u., as required for global space filling.
[19] All atomic potential parameters were obtained using a supercell TB-LMTO calculation with 12 layers of Fe. To achieve better agreement with the Green's function calculations, the combined correction term was not used in TB-LMTO calculations. Our choice of atomic spheres gives excellent agreement with FP-LMTO calculation for the band gap [5.86 eV vs 5.82 eV for bulk MgO with the lattice parameter reduced to match the Fe(001) surface].
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