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Electron correlation effects in the magnetoresistance of Fe/MgO/Fe tunnel junctions: First-principles calculations

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The effect of electronic correlations on the spin-dependent ballistic transport in Fe/MgO/Fe magnetic tunnel junctions with FeO interface layers is investigated by means of first-principles calculations. The self-interaction correction to the local spin-density approximation (LSDA) is applied to electronic states localized at the FeO layers. With respect to LSDA, the electronic and magnetic properties are significantly changed; e.g. the magnetic moment at the Fe interface is increased by 23%, the conductance is in general decreased, and the tunnel magnetoresistance ratio is reduced by up to 40%. The self-interaction correction is deemed beneficial in transport calculations for systems with localized states, e.g., those with oxide layers.

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The tunnel magnetoresistance (TMR) is one key effect in the development of magnetoelectronic devices.[1] Since the early reports on TMR,[2,3] enormous experimental and theoretical efforts have provided understanding of the fundamental mechanisms in spin-dependent transport. Although sizable TMR ratios are reported for magnetic tunnel junctions (MTJs) with amorphous barriers (e.g. Refs. 4–7), MTJs with epitaxial barriers appear more promising with respect to applications. For example, Fe/MgO/Fe and FeCo/MgO/Fe junctions show large TMR ratios.[8] However, theoretical TMR ratios exceed the latter by far.[9,10] This disparity can be attributed to differences between idealized (theoretical) and real (experimental) samples. Especially structural properties, such as disorder[11,12] and noncollinear magnetism[13] at the interfaces, have a profound effect on the TMR.

A prominent example for a structural factor is a substoichiometric FeO layer at an Fe/MgO interface,[14] whose presence depends on the growth conditions.[15,16] Due to the FeO, the charge distribution at the interface is rearranged[17] and the local magnetic moments are decreased.[18] Also transport calculations find significant effects caused by FeO layers.[17–20] The partial oxidization influences the TMR as well.[12,21] Summing up, interfaces determine the TMR considerably (e.g., Ref. 22) and, therefore, have to be described properly in theory.

Transport calculations for Fe/MgO/Fe rely typically on the local spin-density approximation (LSDA) to density-functional theory which provides a good description of delocalized electronic states. In contrast, localized states are rather badly described. A reason for the latter is the interaction of an electron with itself, the so-called self-interaction (SI).[23] As a consequence of the self-repulsion of a localized electron, which can be viewed as a local positive potential, its electronic states appear at too low binding energies. For a delocalized state, the self-interaction tends to be negligible because of its outspread charge distribution. Since localized electronic states are present in FeO interface layers of Fe/MgO/Fe tunnel junctions, a question arises on how the electronic and magnetic structures—and thus the transport properties—are changed by a SI correction with respect to the conventional LSDA treatment.

The treatment of electronic correlations in the LSDA is improved on one hand by going beyond LSDA, for example, by the *GW* approximation[24,25] or by the dynamical mean-field theory,[26,27] but both are numerically demanding. On the other hand, approaches within the LSDA framework have less computational costs. On-site correlations can be improved by the successful LSDA+U method.[28] The on-site correlation energy U is regarded as an adjustable parameter which is chosen to reproduce experimental results, nevertheless leaving an ambiguity concerning its actual numerical value.

The self-interaction correction (SIC) (Ref. 29) improves the exchange-correlation functional of the LSDA by explicitly removing the self-interaction contribution from the self-consistent Kohn-Sham potentials. This leads to orbital- and spin-dependent local potentials for channels, indicated by angular-momentum and spin quantum numbers, which are associated with the localized electronic states. The optimum set of SI-corrected channels is achieved by minimization of the total energy. Hence, the SI correction is parameter-free, unlike LSDA+U. Its local character makes the SIC very attractive for multiple-scattering electronic-structure calculations.[30] A major effect of the SIC is an increase in the binding energies of the localized channels. In molecular junctions this leads to a reduced current.[31] Another example are the magnetic moments and the fundamental band gap of NiO which are considerably closer to experimental values than their LSDA counterparts.[32]

In this Brief Report, we report on an improved treatment of electronic correlations in Fe/FeO/MgO/FeO/Fe tunnel junctions. The self-interaction correction is applied to localized states in the FeO interface layers within a multiple-scattering theoretical approach. The increase in binding energy of these states has a profound effect on the electronic and magnetic properties and consequently on the spin-dependent transport. By comparing SIC with LSDA results we address which scattering channels are affected, whether the spin-dependent conductance is increased, and whether TMR ratios are improved with respect to experiment.

We focus here on Fe/MgO/Fe tunnel junctions with four MgO spacer layers and a stoichiometric FeO layer at either side of the barrier, i.e., a Fe/FeO/(MgO)$_4$/FeO/Fe junction in the [001] orientation. The geometry is adopted from experiment.[19] The electronic structure is calculated from first principles within the scalar-relativistic Korringa-Kohn-Rostoker (KKR) method (for details see Refs. 19 and 33). The charge density is normalized by Lloyd's formula[34] using a maximum angular momentum $l_{\max}=3$. The self-interaction correction, which is implemented according to Ref. 30 in terms of local orbital- and spin-dependent potentials, is applied to the electronic states in the FeO interface layers. The total-energy minimization yields that all *d* electrons with majority-spin orientation of Fe have to be SI corrected.

The ballistic conductance $C(E)$ at energy E is computed within Landauer-Büttiker theory,[35]

$$C(E) = \frac{e^2}{h}\int_{2\mathrm{BZ}} T(E,\boldsymbol{k}_{\|})dk^2. \tag{1}$$

The transmittance $T(E,\boldsymbol{k}_{\|})$ is obtained by layer KKR within an *S*-matrix approach[36] and integrated over the two-dimensional Brillouin zone (2BZ) using special-point meshes[37] with at least 40 000 $\boldsymbol{k}_{\|}$. The TMR ratio

$$\delta(E) \equiv \frac{C_{\mathrm{P}}(E) - C_{\mathrm{AP}}(E)}{C_{\mathrm{P}}(E) + C_{\mathrm{AP}}(E)} \tag{2}$$

is computed from the conductances C_{P} and C_{AP} for the parallel (P) and the antiparallel (AP) configurations of the electrode magnetizations. The chosen range of ±1 eV about the Fermi level E_{F} covers energies which are involved in the transport for typical bias voltages; we consider zero bias here.

The electronic and transport properties are conveniently analyzed by means of spectral densities, momentum distributions, and transmittance maps. The former are computed from the Green's function G_{al} of site a and layer l,

$$N_{al}(E,\boldsymbol{k}_{\|}) = -\frac{1}{\pi}\mathrm{Im}\,\mathrm{Tr}\,G_{al}^{+}(E,\boldsymbol{k}_{\|}). \tag{3}$$

Momentum distributions display $N_{al}(E,\boldsymbol{k}_{\|})$ versus $\boldsymbol{k}_{\|}$ at given E, similar to transmittance maps which show $T(E,\boldsymbol{k}_{\|})$.

Computed within LSDA, the *d*-majority states at Fe sites in an FeO layer occupy the energy range from −5.5 to −0.5 eV (Fig. 1). Sharp maxima provide evidence for their localization at this layer.[38] Application of the SIC increases their binding energies by about 8 eV. The *sp* states hybridize weakly with the *d*-majority states and are thus mildly affected by the SIC (not shown); *d*-minority states are unchanged.

The energetic rearrangement of the *d*-majority states is accompanied by changes in the charge distribution and in the local magnetic moments in the FeO layer and, to a smaller extent, in the adjacent layers. The magnetic moment of Fe is increased from 2.83 μ_B (LSDA) to 3.49 μ_B (SIC), i.e., a gain by 23%. An Fe interface magnetic moment of $(3.3\pm0.3)\mu_B$ has been estimated from x-ray photoelectron spectroscopy experiments.[39] This value agrees (within the error) with the SIC result but deviates significantly from the LSDA result, hence suggesting that the Fe/FeO/MgO interface is adequately described by the SIC.

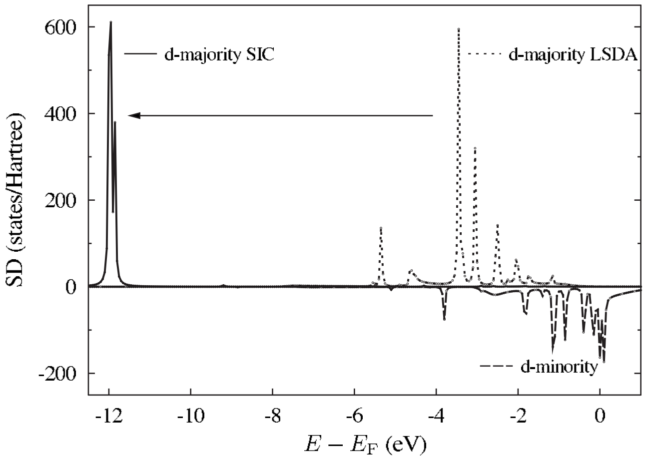

FIG. 1. Spectral density (SD) of *d* states at an Fe site of an FeO interface layer for $\boldsymbol{k}_{\|}=0$. The binding energies of majority states (positive SD) are increased by the SIC (solid) as compared to the LSDA (dotted); cf. the arrow. Minority states (shown as negative SD, dashed) are not affected by SIC.

In the following, we concentrate on two energies for which the analysis is uncomplex. The electronic structure is less influenced by the SIC at energies above E_{F} than below E_{F}. Accordingly, there is hardly any visible change in the momentum distribution for $E_{\mathrm{F}}+0.4$ eV upon application of the SIC [compare Fig. 2(a) with Fig. 2(b)]. One can thus expect that the conductance at this energy is also marginally affected.

Also the momentum distributions at $E_{\mathrm{F}}-0.4$ eV as obtained within LSDA and SIC [Figs. 2(c) and 2(d)] agree in large parts of the 2BZ. The striking difference is that a feature of roughly square shape [indicated by (1) in Fig. 2(c)] is missing in the SIC case [Fig. 2(d)]. We note in passing that momentum distributions at similar energies behave accordingly. Although features in the momentum distributions, in

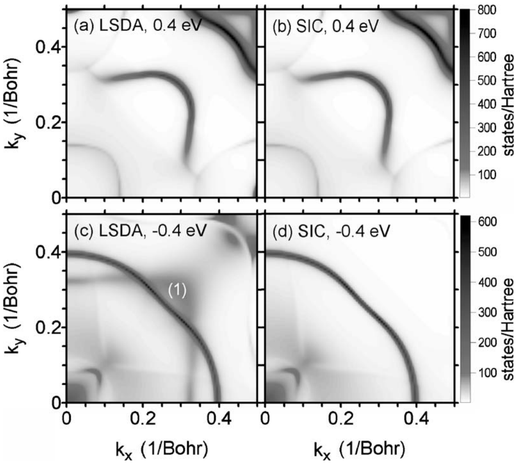

FIG. 2. Momentum distributions of an Fe site in an FeO interface layer calculated within the LSDA (left row) and within the SIC (right row) at energies 0.4 eV (top column) and −0.4 eV (bottom column) with respect to the Fermi level. In each panel a quarter of the two-dimensional Brillouin zone is shown. The region indicated by (1) in (c) is discussed in the text.

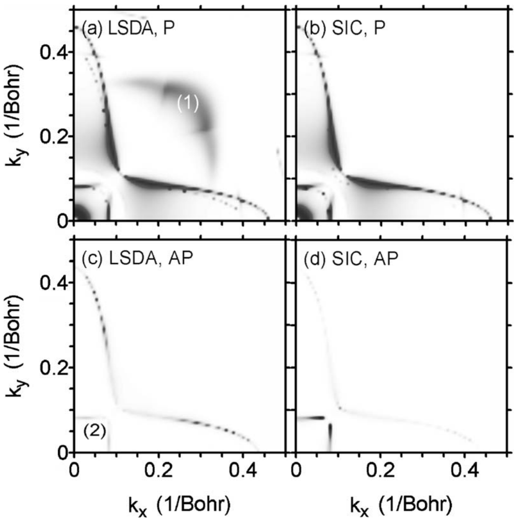

FIG. 3. Transmittance maps for the P (top row) and AP (bottom row) configurations obtained within the LSDA (left column) and within the SIC (right column). The energy is 0.4 eV below the Fermi level. The gray scale covers a transmittance range from 0 (white) to 1.5 (black). In each panel a quarter of the two-dimensional Brillouin zone is shown.

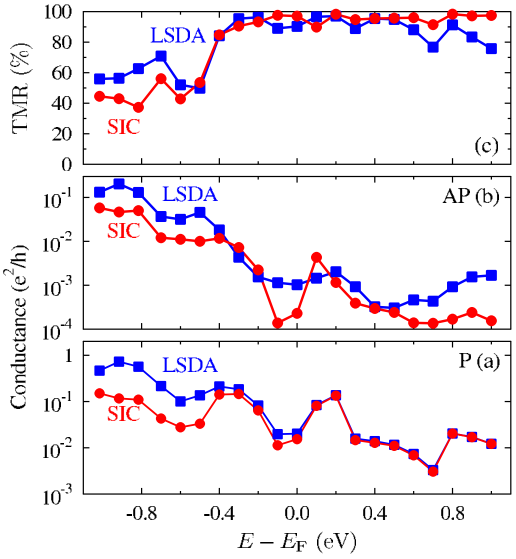

FIG. 4. (Color online) Conductances for (a) P and (b) AP alignments calculated within the LSDA (blue, squares) and the SIC (red, dots), shown in logarithmic scale. (c) The TMR ratio is computed from the conductances in (a) and (b) according to Eq. (2).

particular, those of interface layers, can have counterparts in the transmittance maps (e.g., Refs. 22 and 33), the above findings cannot be extrapolated in a straightforward manner to transport properties. The reason is that transmittances are, roughly speaking, global in the sense that they are determined by the electronic states of the entire junction. In contrast, momentum distributions capture local properties.

To demonstrate the effect of the SIC on the spin-dependent transport, we discuss transmittance maps for $E_F - 0.4$ eV (Fig. 3). For the P configuration, the application of the SIC removes d states with sizable transmittance, especially in the region (1) in Fig. 3(a). The associated conductance is decreased from 0.212 e^2/h for LSDA to 0.140 e^2/h for SIC, that is, by about 34%. Inspection of the momentum distributions corroborates that this finding originates from the d-majority states of Fe in the FeO interface layer.

That SIC affects also layers adjacent to the FeO layer is supported by analyzing region (2) for the AP configuration [Figs. 3(c) and 3(d)]. Here, the transmittance is slightly increased by SIC. The same trend holds also for the P configuration but to a lesser extent and is attributed to the Fe layer next to the FeO layer, as follows from the momentum distributions (not shown here). The above findings show that the SIC can impose contrarian trends in the transmittance. The change in the latter depends especially on the electronic states close-by the SI-corrected sites and on the in-plane wave vector. It appears thus hardly possible to predict changes in the TMR ratios.

As expected from the above discussion, the conductances $C(E)$ are affected most by SIC at low energies, i.e., in an energy range from which the d-majority states in the FeO layer are removed. For both the P and the AP configurations, the conductance is significantly reduced upon application of SIC [Figs. 4(a) and 4(b)]. Since the same trend is found for P and AP, the TMR ratio is moderately affected. More specifically, it is reduced by about 12% on average at energies less than −0.5 eV with a maximum relative decrease of 40% at −0.8 eV. Although there are no d-majority states of Fe at energies larger than E_F, the TMR ratio is increased by about 10% [Fig. 4(c)] which is attributed to the change in C_{AP} by up to 1 order of magnitude [Fig. 2(b), $E \approx E_F + 0.9$ eV].

The first-principles electronic-structure and transport calculations reported here evidence that a proper treatment of localized states can have a profound effect on the spin-dependent transport in magnetic tunnel junctions. It turns out that the SIC, applied to electronic states localized in the FeO interface layers of Fe/MgO/Fe junctions is a suitable method for transport calculations. Although it has less impact on the tunnel magnetoresistance than geometric relaxations (e.g., Ref. 19) or disorder at the interfaces,[12] it nevertheless could be necessary for a complete and proper theoretical description of ballistic transport in junctions with oxide layers (for example, in multiferroic heterojunctions in which ferroelectric perovskites, such as $BaTiO_3$ or $PbTiO_3$, are essential ingredients).

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