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Influence of the interface structure on the bias dependence of tunneling magnetoresistance

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Ab initio calculations for the tunneling magnetoresistance (TMR) in planar Fe/MgO/Fe junctions are presented. The electronic and magnetic structure of the junctions are calculated self-consistently in the framework of density functional theory. The bias dependence of the tunneling conductance and the magnetoresistance is calculated in the limit of coherent tunneling. Positive and negative TMR ratios are obtained as a function of interface structure and even a sign reversal of TMR as a function of bias was found in agreement with experiments.

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The discovery of giant magnetoresistance was immediately accomplished by a theoretical explanation of the effect.[1,2] Experimental results could be explained quantitatively by *ab initio* calculations.[3,4] The revival of tunneling magnetoresistance (TMR),[5,6] however, was characterized by a discrepancy of several orders of magnitude between experimentally and theoretically obtained values of the TMR ratio.

The origin of the discrepancy is related to the sample quality. Experimental results are often obtained in the diffusive limit of tunneling that means high density of structural defects, rough interfaces, and amorphous barriers. Corresponding TMR ratios do not exceed 50% at room temperature[7,8] and could be explained by Julliere's model in terms of the spin polarization of the leads.[9] *Ab initio* calculations of TMR, however, have been performed for structurally ideal junctions in the limit of coherent tunneling.[10,11] It turned out that the results for conductance and TMR are extremely sensitive to slight structural changes. Furthermore, the phenomenon of resonance tunneling occurs for highly symmetric junctions.[10,12] As a consequence, TMR ratios up to 1000% have been predicted. These results are in contradiction to Julliere's model and to existing experimental values. Nevertheless, the results obtained by different computational schemes are in very good agreement and insight into the microscopic origin of tunneling is provided.[10,11]

A similar situation occurred for the bias dependence of TMR. Experimentally obtained bias voltage characteristics of TMR up to 1 V are similar for nearly all junctions under consideration with a general decay for increasing bias, an asymmetry concerning bias reversal, and sometimes a narrow zero bias anomaly.[8,13] Levy and Zhang have shown that magnon scattering at the electrode-barrier interfaces can be one reason for the zero bias anomaly.[14] In general, the tunneling characteristic of planar junctions is very smooth whereas vacuum tunneling shows clear indications of the electronic structure of the leads in the bias dependencies.[15]

Recent experiments based on epitaxially grown Fe/MgO/Fe samples shed light on the subject.[16–18] First, the obtained TMR ratios exceeded the predictions by Julliere's model by far.[9] Second, the bias voltage characteristic shows features that could be related to the electronic structure of the system. *Ab initio* calculations of the bias voltage characteristic of the conductance and TMR are not yet standard calculations. First attempts show again strong differences between experimentally and theoretically obtained results.[19]

The aim of this paper is to demonstrate how the interface structure can influence the TMR ratio and the corresponding bias voltage dependencies.

The role of the interface structure on tunneling was investigated in a variety of experiments.[20–22] In all cases a strong change of conductance and TMR was obtained. In Ref. 23 an inversion of TMR caused by defect states in the barrier is proposed.

All our calculations are focused on epitaxially grown Fe/MgO/Fe systems. For these junctions very accurate data of the interface structure are available.[17,24]

We studied the effect of mixed Fe/O interfaces on the electronic structure and the conductance of the Fe/MgO/Fe tunnel junction. Three types of the crystal structure (Fig. 1) were discussed. The first one has ideal Fe/MgO interfaces without mixing. This structure is possibly very close to the junctions prepared by Yuasa *et al.*[17] The in-plane lattice constant was fixed at the experimental value for the bulk bcc Fe, $a=2.866$ Å. In correspondence to the experimental data of Ref. 25 the Fe-Fe interlayer distance next to the interface is assumed to be 1.7 Å. The remaining Fe layers are separated

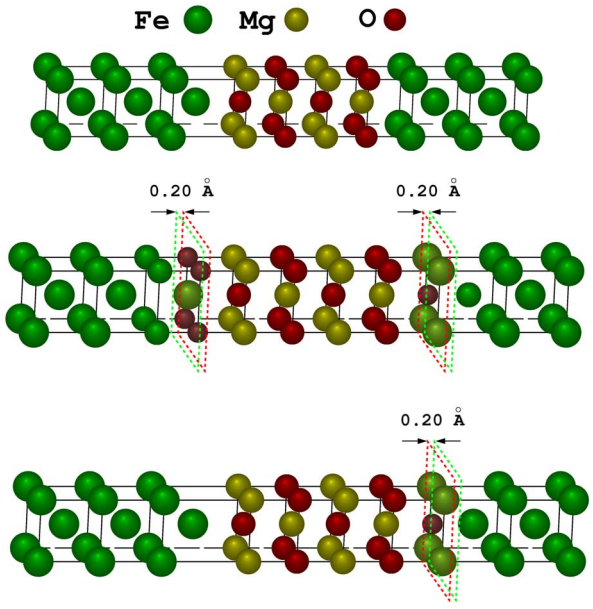

FIG. 1. (Color online) Geometric structures of investigated junctions.

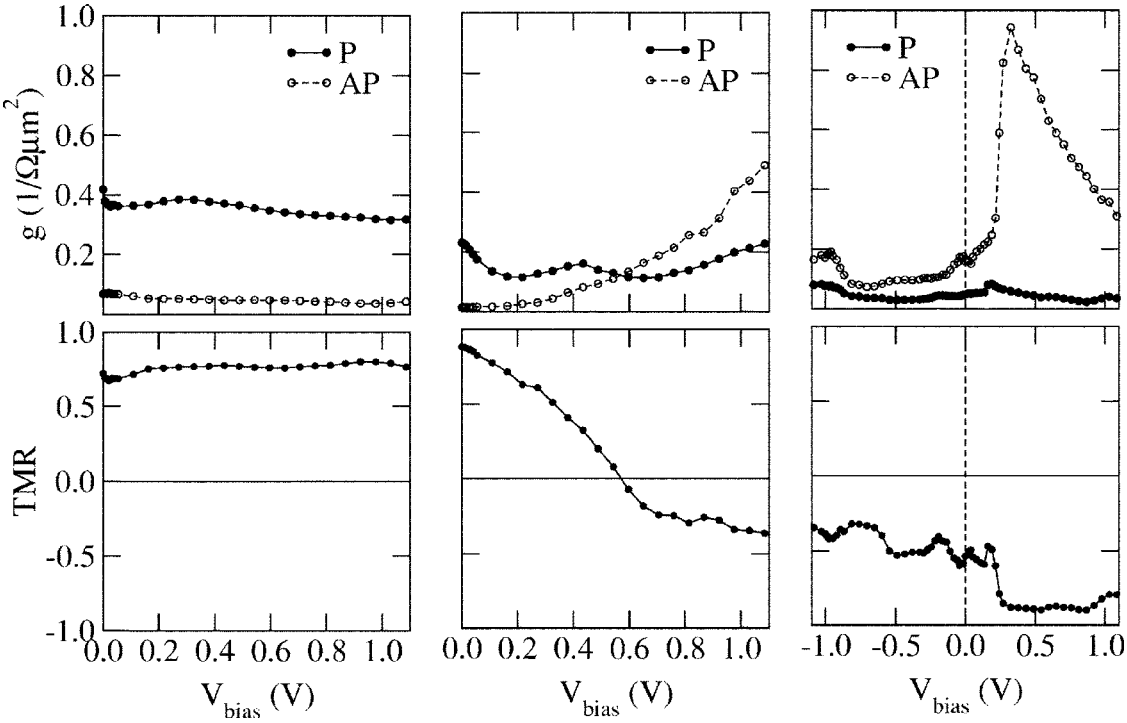

FIG. 2. Bias dependence of conductance and TMR. Left: ideal; middle: symmetric; right: asymmetric.

as in Fe bulk (1.43 Å). The distance between the interface Fe layer and the O in the the first MgO layer is fixed at 2.35 Å. The first and the second MgO layer are separated by 2.25 Å, whereas the distance between the second and third MgO layer is 2.15 Å, which is close to the bulk value of MgO.

In the second junction both interfaces consist of an FeO layer. The system remains symmetric and in the following this label will be used to distinguish between the three geometries, despite that the ideal junction geometry is symmetric, too. The oxygen atoms are placed close to the octahedral vacancy position shifted outward by 0.2 Å, so that the distance between these atoms and the next Mg atoms is 2.15 Å. In our study, all oxygen sites were occupied, and the in-plane periodicity was kept. Partial occupancy of the FeO layer by the oxygen atoms[25] is not discussed in this paper. The third crystal structure under consideration contains both the ideal and the FeO interface and is, of course, asymmetric. This way, we model structural differences between the left and right interface due to specific growth conditions.[24,25] The interlayer distances for all three structural models correspond to the experimental data of Ref. 25 for 4.65 monolayer coverage of MgO.

The electronic structure of the sytems was calculated self-consistently within the framework of density functional theory using a screened KKR (Korringa-Kohn-Rostoker) Green's function method well suited to treat systems of dimensions comparable to experimentally investigated systems.[26,27] For the self-consistent calculations the superlattice geometry with four MgO layers sandwiched by 10 Fe layers was used. For the conductance calculation a system with infinite Fe layers is constructed, which means that two semi-infinite Fe electrodes are effectively taken into account. In the ideal system, the moment of the Fe interface layer was $2.83\mu_B$ similar to the Fe(001) surface. Introducing oxygen at the interface, the Fe moment is quenched to about $2.46\mu_B$ because of charge transfer between Fe and O. In the majority channel the Fe_{3d} states hybridize with the O_{sp} states and in the minority channel the position of the surface state close to E_F is shifted upwards by about 0.3 eV. A similar shift was confirmed in Ref. 28.

Due to the in-plane translational invariance the eigenstates of the electrodes are labeled by the wave vector $\mathbf{k}_\parallel$. The transmission probability as introduced by Landauer[29] was computed using a Kubo formalism expressed in terms of the Green's function of the semi-infinite system.[30] The conductance g is obtained by a two-dimensional integration over the surface Brillouin zone and the assumption of conduction in parallel by the two spin channels

$$T(E) = \sum_\sigma \int d^2\mathbf{k}_\parallel T^\sigma_{\mathbf{k}_\parallel}(E), \tag{1}$$

with the transmission probability $T^\sigma_{\mathbf{k}_\parallel}(E) = \mathrm{Tr}[J^\sigma_L(E)G^\sigma_{LR}(\mathbf{k}_\parallel,E)J^\sigma_R(E)G^\sigma_{RL}(\mathbf{k}_\parallel,E)]$. The planes L and R are situated on both sides of the barrier in the unperturbed electrode regions. $J^\sigma_{L,R}(E)$ are the current operator matrices and $G^\sigma_{LR}(\mathbf{k}_\parallel,E)$ are the Green's function elements connecting both sides of the junction.

Applying an external bias voltage V the chemical potentials of the electrodes μ_L and $\mu_R = \mu_L + eV$ are shifted with respect to each other. Due to the small transmission we assumed a linear voltage drop inside the MgO barrier, which was confirmed by self-consistent calculations.[31] The conductance $g(V)$ is obtained by an energy integration between μ_L and μ_R to cover all tunneling states

$$g(V) = \frac{I}{V} = \frac{e^2}{h}\frac{1}{eV}\int_{\mu_L}^{\mu_R} dET(E). \quad (2)$$

Convergence with respect to $\mathbf{k}_\parallel$ and concerning the energy integration was better than 2%.

The conductance was calculated for parallel (P) and antiparallel (AP) alignment of the moments in the Fe electrodes. Due to the weak magnetic interaction between electrodes a frozen potential approximation was applied to construct the effective potential for AP configuration from P configuration without a self-consistent cycle.[32]

The TMR ratio is the conductance difference for parallel and antiparallel alignment of the moments in the Fe electrodes normalized to the sum of both

$$\frac{g^P - g^{AP}}{g^P + g^{AP}}. \quad (3)$$

This definition differs from the one mostly used. It was chosen to limit the TMR ratio for both, the positive and at the same time the negative, or inverse TMR effect.

The calculated bias dependence of the conductance g and the TMR ratio are shown in Fig. 2 for all considered junctions. For the ideal junction we obtain only small variations of g with the voltage for both magnetic configurations. Therefore the tunneling magnetoresistance is nearly constant over the whole voltage range. Using the conventional definition of TMR as $(g^P - g^{AP})/g^{AP}$ the values are between 400–800%, which is the same order of magnitude as the experimental results of 247%.[17]

The same behavior of g was found for all junctions in the P configuration. Strong variations of the conductance as a function of bias are obtained in the AP configuration of the symmetric and asymmetric junction. The symmetric junction shows a strong increase, whereas in the asymmetric case the broken symmetry causes a different behavior of conductance for positive and negative voltage. These features are reflected in the TMR ratio. For the symmetric geometry the increase of g in the AP configuration causes a decreasing TMR with bias, which is normally observed in experiments. TMR even changes sign at 0.57 V.

In the asymmetric junction the TMR is negative over the whole voltage range because of the higher conductance in the AP configuration. Negative TMR ratios are in contradiction to Julliere's model for junctions with identical leads. Nevertheless, negative TMR was predicted theoretically and seems to be an indication of coherent tunneling.[31] An interesting feature is the resonance in the conductance of the AP configuration at about 0.33 V. This resonance is related to the density of states at the interface layers and their symmetry, which is decisive for the tunneling matrix elements, and will be discussed in detail in a forthcoming paper. At a voltage of about 0.3 V unoccupied interface resonance states at the FeO interface of majority character start to contribute to the tunneling current. These states are located close to the Γ point of the Brillouin zone and have the same symmetry as the corresponding occupied minority states in the opposite electrode with the ideal interface. In contrast, these unoccupied majority states at the FeO interface have no counterparts in the occupied minority states and no resonance appears for the conductance of the symmetric junction geometry.

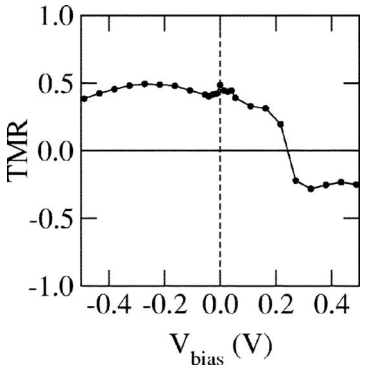

FIG. 3. Bias dependence of TMR of a junction with inhomogeneous interfaces (for details see text).

As can be seen from Fig. 2 the TMR characteristics of a given electrode-barrier combination can vary in a wide range driven by the structure of the interfaces. Up to here a homogeneous structure of the interface was considered. The following example demonstrates how the TMR characteristic can be influenced by inhomogeneous structures deviating from the previous ones.

Assuming that the interfaces have inhomogeneous FeO decoration, the whole junction can be considered as a system of columns conducting in parallel with different interface structures (Fig. 1). The conductance behavior of every column is described above (Fig. 2). A simple structural model would be to combine the ideal, symmetric, and asymmetric junction geometry with equal weight. A resulting different oxygen occupation of the left and right interface can be expected from specific growth conditions. The corresponding TMR characteristic obtained for this model junction, which is shown in Fig. 3, is very similar to an experimentally obtained one.[28] Especially, the sign reversal of the TMR occurs nearly at the same voltage as in the experiment. The reason is the resonance in the conductance in the AP configuration of the asymmetric junction geometry. Therefore the sign reversal in the experiment is a clear indication of the influence of the density of states at the FeO interface layer and is related to a fraction of the junction with asymmetric geometry.

In conclusion, the conductance of Fe/MgO/Fe tunnel junctions was calculated without adjustable parameters in the limit of coherent tunneling for different interface geometries as a function of the applied bias voltage. The modification of the interfaces allows the generation of positive and negative TMR ratios. The bias characteristic is strongly influenced by the interface geometry since it is related to the local densities of states and the symmetry of the eigenstates at the interface layers. A bias dependence close to the experiment[28] with clear indications of coherent tunneling is obtained for a simple structural model.

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