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Oscillatory tunneling magnetoresistance caused by antiferromagnetic Mn layers

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The ballistic magnetoresistance of tunnel junctions that comprise Mn films is found to exhibit oscillations with increasing Mn-film thickness, as is investigated by means of first-principles electronic-structure and transport calculations. The period of two monolayers is directly related to the layer-wise antiferromagnetic structure of the Mn films, in particular to the alternating magnetization at the interfaces. These findings substantiate unequivocally the effect of the electronic and magnetic structure of interfaces on the conductance of tunnel junctions.

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Magnetoelectronic devices are typically composed of several parts, therefore comprising necessarily interfaces. With increasing miniaturization, structural, electronic, and magnetic properties of these interfaces become more and more important and are believed to determine essentially transport properties of spintronics devices.

In an experiment, interface structures can hardly be modified in a fully controlled manner. Typically, a series of samples needs to be fabricated under various preparation conditions, the transport properties of which are obtained subsequently. Although preparation techniques have made significant progress in the recent past, well-defined (on an atomic scale) interface structures are difficult to produce. Thus, experimentally achieved findings involve often a statistical (configurational) average.

As an example, we address Fe/MgO/Fe magnetic tunnel junctions (MTJs) that are among the in-depth investigated systems of magnetoelectronics.[1,2] In early theoretical investigations, it was assumed that MgO continues epitaxially the bcc lattice of the Fe(001) electrodes (e.g., Refs. 3 and 4). However, recent experimental investigations of the Fe/MgO interface structure revealed a partially occupied FeO layer that was found to change the theoretical tunnel magnetoresistance (TMR) drastically, with respect to that of the "ideal" structure.[5–8]

One particular issue that is not well understood to date is the influence of the magnetic structure of an interface on the TMR, in particular the effect of antiferromagnetic order. A crucial point is that the magnetic and the geometric structure are not as independent as requested, as is for example the case for Fe/MgO interfaces. As a consequence, one needs well-specified samples to extricate interface effects from transport data.

In this paper, we propose to consider layerwise antiferromagnetically (LAFM) ordered films for investigating the magnetic structure of interfaces and its effect on the ballistic TMR of planar tunnel junctions. Layerwise antiferromagnetic structures are found in epitaxial Cr and Mn films on Fe(001).[9–11]

In a recent study, the TMR of Fe/Cr(x)/Al_2O_3/FeCo tunnel junctions was investigated both experimentally and theoretically.[12] The most striking result is an oscillatory TMR with a period of two monolayers (ML) that was explained by means of the Cr band structure at $\vec{k}_\parallel=0$. However, due to the amorphous Al_2O_3 tunnel barrier and the polycrystalline FeCo electrode, the transport is likely to be diffusive ($\vec{k}_\parallel$ not conserved) and not specular ($\vec{k}_\parallel$ conserved). To obtain ballistic transport, we deliberately replaced Al_2O_3 and FeCo by vacuum and Fe, respectively. Consequently, one is not restricted to band-structure calculations but state-of-the-art methods for computing ballistic transport of MTJs can be applied as well.

The intention of the present work is to identify unequivocally the effects of a LAFM film (Mn) between a tunnel barrier (vacuum) and an electrode (Fe) on the TMR (Fig. 1). The two major magnetic configurations parallel (P) and antiparallel (AP) are defined in terms of the Fe-electrode magnetizations. As was shown recently by first-principles electronic-structure calculations,[13] Mn couples LAFM with respect to the Fe(001) substrate, provided the Mn film is sufficiently thick (>7 ML). These findings are consistent with experimental observations.[14] That Mn films on Fe(001) serve well for our purposes was also shown in a recent investigation by means of spin-resolved scanning tunneling

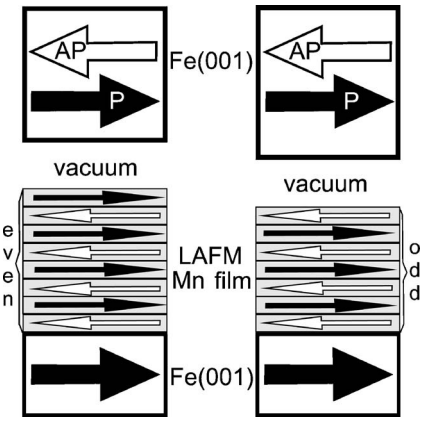

FIG. 1. Fe(001)/Mn(x)/vacuum/Fe(001) tunnel junctions for an even (left) and an odd (right) number x of Mn layers (schematic). The local magnetizations (horizontal arrows) are aligned layer-wise antiferromagnetically (LAFM) within the Mn films. The magnetic configurations P (parallel, as indicated in the top electrode) and AP (antiparallel) are defined with respect to the Fe-electrode magnetizations, with that of the bottom electrode fixed.

microscopy.[15,16] In that work, the LAFM structure was directly proven.

Consequently, we focus in this work on MTJs with Mn film thickness x ranging from 7 to 12 ML. The main difference of MTJs with an even and an odd x is the orientation of the magnetization in the Mn top layer. For even (odd) x, the latter is parallel (antiparallel) to both Fe-electrode magnetizations in the P configuration. Thus, one could expect an even-odd effect in the TMR, provided the influence of the Mn top layer is large enough.

In summary, the present theoretical *ab initio* investigation addresses the following questions: How large is the TMR, in particular is it as small as in STM experiments and in model calculations?[16] How does the TMR depend on the thickness of the Mn film? And is there an even-odd effect that is related to the respective Mn top-layer magnetization?

In a first step, *ab initio* electronic-structure calculations were performed within the framework of the local spin density approximation to density functional theory. The self-consistent potentials serve as input for the transport calculations. Both electronic-structure and transport properties were obtained with a spin-polarized layer-KKR computer code.

The computations for planar Fe(001)/Mn(x)/vacuum/Fe(001) MTJs, $x=7,\ldots,12$ ML, follow closely those for Fe(001)/Mn(x)/vacuum reported in Ref. 13. The number of vacuum layers was fixed to 3 ML (corresponding to 5.7 Å electrode separation). Thus, electronic states localized at different electrodes do not interact significantly.

The tunnel current $I(V)$ of a MTJ is calculated within Landauer-Büttiker theory.[17,18] In order to treat a nonzero bias voltage V, the potentials in one electrode were shifted rigidly by eV.[19,20] A linear voltage drop within the vacuum region is assumed.[21] This non-self-consistent treatment is corroborated by self-consistent results for Fe/FeO/MgO/Fe MTJs.[22]

The tunnel current $I(V)$ and the conductance $G(V)$,

$$I(V) = G(V)V = \frac{e^2}{h}\int_{\mu_T}^{\mu_B} T(E,V)dE, \tag{1}$$

are expressed as integral of the transmittances $T(E,V)$ in the "energy window of tunneling" given by the chemical potentials of the bottom (B) and the top (T) electrode, $eV=\mu_B-\mu_T$. $T(E,V)$ is obtained by integrating the wavevector-resolved transmittances $T(E,V;\vec{k}_\parallel)$ over the two-dimensional Brillouin zone (2BZ) (Ref. 18)

$$T(E,V) = \int_{2\mathrm{BZ}} T(E,V;\vec{k}_\parallel)dk^2. \tag{2}$$

$T(E,V;\vec{k}_\parallel)$ is the sum of the transmission probabilities of all incoming (in $\mathcal{B}$) and outgoing (in $\mathcal{T}$) Bloch states of the leads. For the 2BZ integration an adaptive-mesh algorithm was used.[23] The tunnel magnetoresistance $\delta(V)$ is defined as the asymmetry of the tunnel currents for the parallel and antiparallel magnetic configurations of the Fe electrodes (Fig. 1), $\delta(V)=[I_\mathrm{P}(V)-I_\mathrm{AP}(V)]/[I_\mathrm{P}(V)+I_\mathrm{AP}(V)]$.

To investigate interface effects on the conductance of a MTJ, it is highly desirable that only properties of a single interface layer change while those of the remaining layers do not. For Fe(001)/Mn(x)/vacuum, this was shown by first-principles calculations to be the case to a good approximation,[13] provided the number of Mn layers x is large enough ($x\geqslant 7$ ML). If so, the main difference of thick Mn films with an even and an odd number of Mn layers is the opposite alignment of the top-layer magnetizations (Fig. 1).

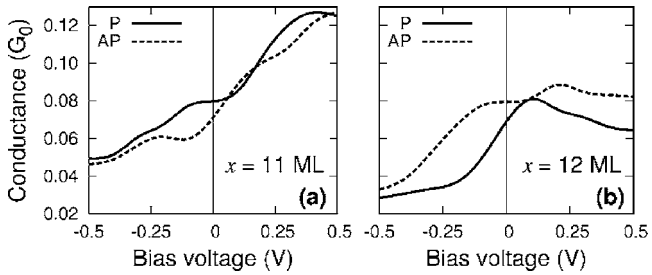

FIG. 2. Conductances of Fe/Mn(x)/vacuum/Fe magnetic tunnel junctions for $x=11$ ML (a) and $x=12$ ML (b) of Mn for parallel (P, solid) and antiparallel (AP, dashed) magnetic configurations versus bias voltage. $G_0=e^2/h$ is the quantum of conductance.

For the bias dependence of the conductances with $x=11$ ML [Fig. 2(a)], one finds $G_\mathrm{P}(V)>G_\mathrm{AP}(V)$ almost in the entire range of voltages. Increasing x by 1 ML reverses the order to $G_\mathrm{P}(V)<G_\mathrm{AP}(V)$. Note further that $G_\mathrm{P}(V)$ and $G_\mathrm{AP}(V)$ exhibit a rather similar bias dependence for both x, indicating a small TMR $\delta(V)$ (in absolute value). The variation upon increasing x indicates that there are still differences in the electronic structure of the Mn films, although x is rather large.

These findings suggest that the Mn top layer acts mainly as a spin filter which suppresses the transmission of electrons of one spin orientation with respect to that of the other (for oxidized Co surface, see Ref. 24). It essentially determines the order of the conductances $G_\mathrm{P}(V)$ and $G_\mathrm{AP}(V)$. Consequently, the TMR $\delta(V)$ changes sign as a function of the Mn-film thickness (Fig. 3). This even-odd effect shows up clearly for negative bias voltages V (i.e., for tunneling into the bottom electrode). For an even x, $\delta(V)$ is negative (blue) whereas for odd x, it is positive (red). For positive V, the thickness dependence is more complicated but also shows 2-ML oscillations.

The TMR of the paradigm of MTJs, Fe(001)/MgO(x)/Fe(001), approaches 1 with increasing MgO thickness (see, e.g., Ref. 5). For Fe/Mn(x)/vacuum/Fe, however, the TMR is comparatively small, with a maximum of about 0.3. On average, $|\delta(V)|$ is even smaller. In particular, it shows no unequivocal trend (i.e., decrease or increase) upon increasing x, as is explained by the fact that a Mn film is conducting (in contrast to an insulating MgO film). These findings agree nicely with experimental data and with those of a model calculation.[16]

Recapitulating at this point, the TMR of Fe/Mn(x)/vacuum/Fe MTJs shows an even-odd effect, prominently at negative bias voltages [Fig. 3(a)]. The tunnel magnetoresistance agrees nicely with the experimental one for Fe/Cr(x)/Al_2O_3/FeCo MTJs, which also exhibit a 2-ML period in particular voltage ranges.[12] These findings suggest that the oscillations are explained by the same mechanism,

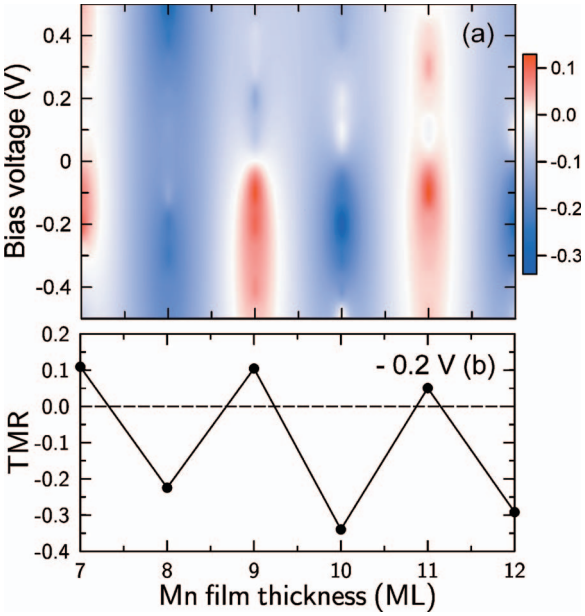

FIG. 3. (Color) (a) Tunnel magnetoresistance (TMR) in Fe/Mn(x)/vacuum/Fe magnetic tunnel junctions versus number x of Mn layers (abscissa) and bias voltage (ordinate), depicted as color scale (right). Data are interpolated to obtain a smooth color gradient. (b) TMR versus x at −0.2 V bias voltage.

irrespectively of the transport being specular or diffusive.

The TMR oscillations cannot be explained by the electronic structure of a MTJ at a single $\vec{k}_{\|}$. Electronic states in the entire 2BZ can contribute to the transmittance [Eq. (2)], although typically a few small regions in reciprocal space outweigh by far the contribution of the remainder. First evidence for the 2-ML oscillations being related to the Mn top layer is provided by the observation that the $\vec{k}_{\|}$-resolved zero-bias transmittance $T(E,V;\vec{k}_{\|})$ for P (AP) and $x=11$ ML agrees well with that for AP (P) and $x=12$ ML (not shown). Further support is given by comparing the layer-, spin-, and angular-momentum-resolved Bloch spectral densities (SD) with $T(E,V;\vec{k}_{\|})$, being aware that there is no one-to-one correspondence of these quantities.

Figure 4 shows that structures in the transmittances (top row) have counterparts in the *sp* spectral densities of the surface layer of the Fe top electrode (c), (d) and of the Mn top layer (e), (f). We would like to draw the reader's attention to the selected features, as indicated in black. For the other layers, there is no such clear correspondence, as is evident, for instance, for the Mn layer at the Fe/Mn interface of the bottom electrode (g), (h) and for an Fe bulk layer (i), (j). Thus, these layers play a minor role concerning the electronic transport. From this finding, one may conclude that layers close to the tunnel barrier govern the transport properties. Please note that the change of the potential is largest at these layers.[31]

The *d*-state spectral densities do not fit well to the transmittances, which implies that these states contribute much less to the conductance than *sp* states, regardless of their much larger SD. This observation is consistent with the

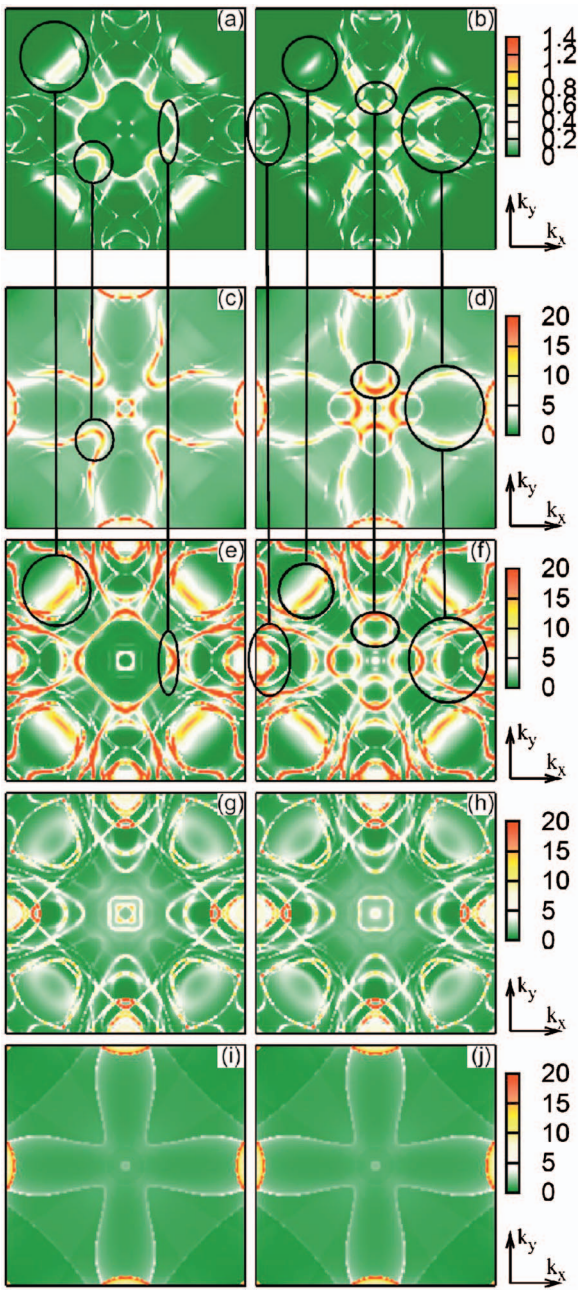

FIG. 4. (Color) Transport properties of a Fe/Mn(11)/vacuum/Fe tunnel junctions in P (left column) and AP (right column) configuration. Top row: transmittances $T(E,V;\vec{k}_{\|})$ in the 2BZ at zero bias, shown as color scale (right). Bottom rows: spectral densities (SDs) at the Fermi level for *sp* electronic states (color scales in states/Hartree) for the surface layer of the top Fe electrode (c), (d), the Mn top layer (e), (f), the Mn layer at the Mn/Fe interface of the bottom electrode (g), (h), and of an Fe bulk layer (i), (j). Selected features are indicated in black.

Slonczewski model.[25] Also spectral densities decomposed with respect to point-group representations at the 2BZ center[12] do not agree well with the transmittances.

A conducting spacer can give rise to quantum-well states which may determine the transport properties considerably, as was shown for thin Cu(001) films in a MTJ.[26,27] The oscillations in the TMR as a function of Cu thickness have a

period that is given by the nesting vectors of the constant-energy surface. In contrast to Cu, with its highly conducting *sp* states at the Fermi level, Mn quantum-well states do not show up in the transport properties.[30,32] Therefore, they cannot be responsible for the 2-ML oscillations.

Spin-polarized surface states would increase the transmission in one spin channel, thereby reducing or increasing the TMR. Consequently, they would affect the amplitude of the TMR oscillation but not its 2-ML period. Surface states were indeed found in experiments on Mn/Fe(001).[14,16] However, the most prominent one, at +0.8 eV relative to the Fermi level,[13] is not within the range of bias voltages considered here.

In a recent theoretical investigation, the tunneling anisotropic magnetoresistance in Fe(001)/vacuum/Cu(001) was attributed mainly to a minority-spin surface resonance in Fe(001).[28] For the Fermi energy, it shows up close to the 2BZ center.[29] In that system, the Cu electrode provides conducting channels in the entire 2BZ, in contrast to a Mn/Fe electrode. Consequently, these surface resonances—showing up in our calculations at the top electrode [e.g., the square-shaped structure at the center of Fig. 4(c)]—have little effect on the TMR studied in this paper.

The electronic and magnetic structure of layers close to the tunnel barrier determine essentially the transport properties in MTJs, for instance, the TMR. This conclusion is obtained by first-principles calculations for Fe(001)/Mn(x)/vacuum/Fe(001) MTJs, $x=7,\ldots,12$ ML. The LAFM order in the Mn films results in TMR oscillations with a period of 2 ML (even-odd effect), as is explained by the alternating orientation of the top-layer magnetization upon increasing x by 1 ML. These theoretical results suggest to investigate experimentally similar systems, e.g., by spin-resolved STM.

The LAFM order of conducting spacers and the associated even-odd effect in the TMR provides an additional degree of freedom for spin-dependent transport in MTJs, especially in connection with advanced preparation techniques. Hence, the presented results might be important for technological applications.

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[31] For Fe/MgO/Fe junctions these are obviously the layers at the Fe/MgO interfaces.
[32] Resonant tunneling would show up as so-called hot spots in the transmittance (Ref. 30).