Effect of antiferromagnetic layers on the spin-dependent transport in magnetic tunnel junctions

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(Received 13 January 2006; revised manuscript received 21 June 2006; published 7 August 2006)

Spin-dependent tunneling between a layer-wise antiferromagnet grown on a ferromagnetic substrate and a ferromagnetic tip electrode was studied using spin-polarized scanning tunneling microscopy (Sp-STM). The systems under investigation consist of thin Mn films deposited on an Fe(001) substrate, a vacuum barrier, and an Fe-coated STM electrode. The asymmetry of the tunnel currents, measured over a wide voltage range, is nonzero but small, only a few percent and exhibits a change of sign as a function of voltage. Surface states are found to contribute significantly to the tunnel current only for small tip-sample distances. The comparison with model calculations for ballistic transport supports that the current asymmetry can be attributed mainly to the symmetry breaking at the surface, that is to the surface layer of the Mn film.

DOI: 10.1103/PhysRevB.74.054409 PACS number(s): 75.50.Ee, 68.37.Ef, 73.20.At, 75.47.Jn

I. INTRODUCTION

Many experimental and theoretical studies were performed to investigate the tunnel magnetoresistance (TMR) in magnetic tunnel junctions (MTJs),[1,2] the latter typically consisting of two ferromagnetic electrodes separated by an insulating barrier.[3] The TMR describes the dependence of the electrical resistance on the relative orientation of the magnetizations in the two electrodes.[4] It depends significantly on details of the electronic, magnetic, and geometric structures of the interfaces between the leads and the barrier. One way to study systematically the effects of the interfaces is to prepare samples by different means, thus altering the interface structure. This approach has the drawback that the interfaces are sometimes ill-defined and, consequently, the results are difficult to interpret and to generalize. An alternative approach, followed here, is to use a spin-polarized scanning tunneling microscope (Sp-STM) instead of planar tunnel junctions to investigate the spin-dependent tunnel process. By this means, the TMR is studied for well-defined electrodes *and* controlled barriers, namely the vacuum barrier.[5] Further, Sp-STM probes the TMR locally which offers the possibility to study the transport through small and laterally confined structures which are apparently free of defects. Evidently, averaging over micron-sized MTJs, as in a setup involving planar junctions, is avoided.

One important issue of magnetoelectronics is the influence of antiferromagnetic layers on the spin-dependent tunneling in MTJs.[6] Major questions comprise the occurrence of a TMR, its magnitude, and the roles of surface states, interface states as well as bulk states. Of particular advantage for a detailed investigation are layer-wise antiferromagnets, the magnetic moments of which align ferromagnetically within one atomic layer and antiferromagnetically between adjacent atomic layers. Thus adjacent atomic layers at the film surface are oppositely magnetized. In former investigations that addressed the spin-dependent tunneling at the surface of layer-wise antiferromagnets,[7,8] the origin of the TMR was not addressed in detail. Therefore, we report in this paper on a Sp-STM investigation of Mn films on Fe(001) and focus especially on origin and size of the TMR.

The TMR has its origin in the spin polarization of the electrodes. Hence, in the following we will discuss the spin polarization in ferromagnets and antiferromagnets.

The spin polarization in ferromagnets appears due to the exchange splitting between spin-up and spin-down electronic states. This results in an imbalance in the spin-resolved density of states, causing in particular a spin polarization of the conducting electronic states close to the Fermi energy. In contrast to the situation in ferromagnets, electronic states in the bulk of layer-wise antiferromagnets are degenerate[9] and there exists no net spin polarization. The sample surface, however, breaks the translation symmetry, which lifts the degeneracy of the bulk bands resulting in a nonzero spin polarization in the Mn-surface layers. In addition it may lead to the formation of spin-polarized surface states.[10,11] The latter can be exploited to achieve a spin contrast in spin-polarized scanning tunneling spectroscopy, as was demonstrated for antiferromagnetic bulk Cr[8] and Mn films on Fe(001).[7] Even without spin-polarized surface states, however, alone the presence of the surface of layer-wise antiferromagnets leads to a spin polarization of the conduction electrons.

Before presenting our results, we sketch scenarios on the origin and size of the TMR. In a first scenario, we use the Jullière model[4] for the description of the resulting TMR. In this model, the spin polarization of the electrodes is considered as a bulk property, any interface or surface effects are neglected. This means that in principle this model is not sufficient to describe the behavior of a TMR of an electrode consisting of a thin magnetic film on a ferromagnetic substrate. Taking, however, this model literally one could assume for our system the bulk properties of the Fe substrate, while neglecting the Mn film or the bulk properties of the Mn while neglecting the Fe underneath. In the first case, one is mainly left with the sizable TMR between two joint Fe electrodes.[12] If we consider the Mn film as a bulklike layer-wise antiferromagnet, the electronic states are degenerated as described above. According to the Jullière model this would result in a zero TMR.

In a second scenario we will consider the improved Jullière model by Maekawa and Gafvert.[13] In this model, the spin polarization at surfaces and interfaces are important. The interface between Fe and Mn is the same for an even and odd number of Mn layers. Thus, only the spin polarization at the Mn surface layers (with magnetic moment of about

$3.7\mu_{\mathrm{Bohr}}$) are essential for the considered MTJ. This implies an even-odd effect due to the number of Mn layers. Hence, the TMR should be small and changes sign upon adding a monolayer of Mn (again, the Fe substrate is solely used to stabilize the layer-wise antiferromagnetic structure of the Mn film).

The above scenarios describe two extreme cases, one considers bulk properties and the other surface and interface properties. The latter model, however, describes the experiments more appropriate since it has been observed experimentally that interfaces play a major role in MTJ (see, for example, the effect of FeO layers in Fe/MgO/Fe tunnel junctions[14–16]). Therefore, we expect a small TMR with alternating sign upon changing the Mn-film thickness by one layer.

In the present study, the investigated MTJs consist of an Fe-coated ferromagnetic STM electrode, a vacuum barrier, and thin Mn films deposited on an Fe(001) substrate. It will be shown in the following that irrespective of the presence of surface states the breaking of the translational symmetry of the bulk states by the surface of a layer-wise antiferromagnet alone causes a spin-dependent tunnel current and, consequently, a tunnel magnetoresistance. The experimental results are supported with model calculations for ballistic electronic transport that include the key ingredients of the experimentally investigated MTJ and are based on first-principles electronic-structure calculations.[10]

II. EXPERIMENT

The Sp-STM experiments were performed in ultrahigh vacuum (base pressure $p < 10^{-10}$ mbar) and at room temperature. Mn films were deposited on clean flat Fe(001) surfaces of Fe whiskers by electron-beam evaporation at a substrate temperature of about 400 K. The film thickness was monitored by medium-energy electron diffraction.[17] The magnetization of the Fe(001) surface is in-plane, and the magnetic configuration of the Fe substrate was controlled by in-situ Kerr microscopy measurements. All Sp-STM data shown in this paper were taken in regions where the Fe substrate was homogeneously magnetized along the [001] direction. Using ferromagnetic rings as tip electrodes of the Sp-STM,[18] the spin component of the spin polarization of the Mn films collinear to the Fe substrate magnetization was investigated. In Sp-STM, spin sensitivity is achieved using ferromagnetic STM electrodes and analyzing the spin-dependent tunnel current between the STM electrode and a magnetic surface. Fast switching of the magnetization of the tip electrode with a frequency much higher than the cutoff frequency of the STM feedback loop allows to separate topographic from spin information (the latter is correlated to the magnetization) and, thus, both can be imaged simultaneously.[19]

The quantities discussed later are defined as follows. The tunnel current I is measured for opposite tip magnetizations ($\uparrow$, $\downarrow$). The current asymmetry is then given by

$$A(M_{\mathrm{Mn}}) \equiv \frac{I(\uparrow) - I(\downarrow)}{I(\uparrow) + I(\downarrow)}. \quad (1)$$

Here, M_{Mn} indicates that the asymmetry A depends on the thickness and thus on the magnetization within the Mn film, in particular on that of the surface layer. The spin contrast is defined as $C \equiv A(+M_{\mathrm{Mn}}) - A(-M_{\mathrm{Mn}})$ with $\pm M_{\mathrm{Mn}}$ for oppositely magnetized Mn top layers (i.e., the local Mn-film thickness differs by 1 ML for $\pm M_{\mathrm{Mn}}$).

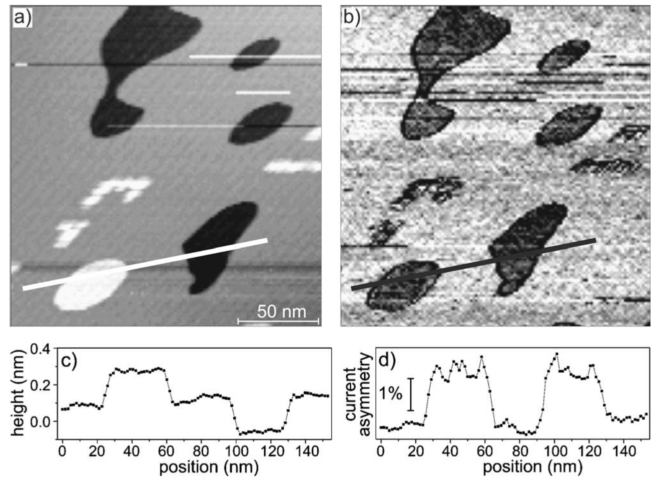

FIG. 1. STM images of 11.9 ML (±1 ML) Mn on Fe(001), taken at 0.1 V bias voltage and 3 nA tunnel current. The topography (a) and the current asymmetry (b) are displayed for the same region of the sample. The line profiles in (c) and (d), taken along the lines in (a) and (b), show clearly the layer-wise antiferromagnetic order between three different Mn layers.

In order to obtain the current asymmetry as a function of bias voltage, the tip was stabilized at a chosen voltage and a chosen average tunnel current. Subsequently, the feedback loop of the STM was opened and the bias voltage was ramped, implying that the tip-sample distance was fixed while the average tunnel current and the spin-dependent tunnel current changed. This procedure was repeated at each lateral point of the image. Note that the applied voltage between the electrodes of a MTJ determines the number of electronic states that contribute to the tunnel current, thus also the spin polarization of the tunnel current.[26]

III. DISCUSSION OF EXPERIMENTAL RESULTS

Figure 1(a) shows the topography of an 11.9 ML (±1 ML) thick Mn film. The top layer n (grey) is almost closed but in addition to layer n, layer $n+1$ (islands, white) and $n-1$ (holes, black) are visible at the surface. The different surface layers are separated in height by monatomic Mn steps, as is evident by the line profile [Fig. 1(c)].

The surface magnetic structure is depicted in Fig. 1(b). Clearly, an alternating contrast is visible in the current asymmetry, showing the layer-wise antiferromagnetic order of adjacent Mn layers.[7] Thus, holes and islands show the same current asymmetry but opposite to the one of the layer in between them [cf. the line profiles in Fig. 1(c) and 1(d)].[17]

In the following, we will consider the investigations of the current asymmetry as a function of the bias voltage. The current asymmetry, Eq. (1), was measured for bias voltages ranging from +1 V to −1 V. To determine the spin contrast C, A was taken on two oppositely magnetized Mn top layers (for example, closed layer—hole or island—closed layer).

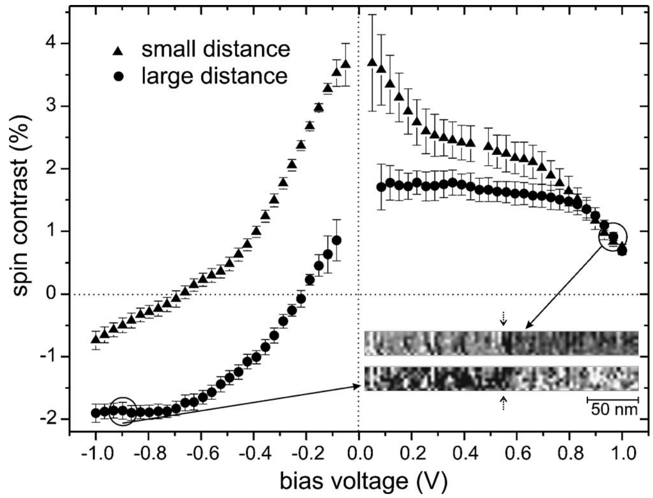

FIG. 2. Spin contrast vs bias voltage for Mn films 6 ML and 7 ML (±1 ML) thick. The STM tip was stabilized at two different distances from the Mn top layer: for 3 nA and 1 V (●; large tip-sample distance) and for 24 nA and 1 V (▲; small tip-sample distance). Each data point corresponds to an average over about 300 pixels. The insets show the current asymmetries for +0.97 V and −0.9 V. At the position of the dotted arrows, a step of monatomic height is present in the topography.

From theory it is known that for Mn films thicker than about 4 ML the electronic structure does not depend significantly on the Mn film thickness,[10] as is supported by experiments.[7,20] Thus, the measured spin contrast C is expected to be nearly independent on the Mn film thickness, provided that the Mn film is thick enough (as is the case in the present investigation).

Figure 2 shows the experimental result of the spin contrast.

In the following, we focus on two representative data sets, one obtained for a large tip-sample distance (stabilization conditions of 1 V and 3 nA, ● in Fig. 2) and the other one for a small tip-sample distance (1 V and 24 nA, ▲). Both stabilization conditions result in similar dependencies of the spin contrast on the bias voltage.[27] In case of the large tip-sample distance, a positive spin contrast is measured for bias voltages larger than −0.2 V and a negative otherwise. The change of sign is also demonstrated by the reversed current asymmetries shown in the two images in the inset of Fig. 2. The upper image shows the current asymmetry taken at 0.97 V and the lower one the current asymmetry taken at −0.9 V. In case of the smaller tip-sample distance (▲), however, a peak shows up close to 0 V which shifts the change of sign to −0.7 V. This difference can be explained by the fact that for smaller distances electronic states with large momentum $\hbar\vec{k}_{\parallel}$ in the surface plane have an enhanced transmission probability through the barrier than at larger distances where typically the center of the two-dimensional Brillouin zone contributes, as will be discussed later. For different STM tips, the same general shape of the voltage dependence was observed, only the size of the spin contrast changed slightly.

Summarizing at this point, the experiments show that a spin contrast can be observed at the surface of antiferromagnetic Mn films on Fe(001). The detected contrast occurs practically in the whole investigated range of bias voltages, indicating that it is not related to spin-polarized surface states only, as previously claimed for Mn on Fe and Cr.[7,8] It is rather small, i.e., a few percent (in absolute value) which is much less than recently found in optimized ferromagnet/insulator/ferromagnet MTJs.

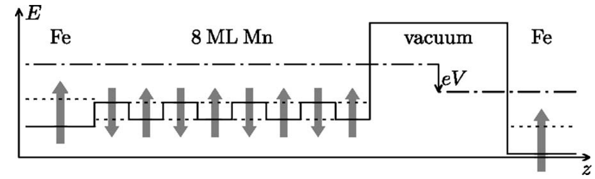

FIG. 3. Model of the magnetic tunnel junction in parallel configuration (schematic). The spin-dependent potentials (spin-up, solid; spin-down, dashed) are shown for Fe electrodes separated by 8 ML of Mn and a vacuum barrier. Arrows indicate the magnetization orientation in each slice. The Fermi level (dashed-dotted) of the right Fe electrode is shifted by the bias voltage V with respect to that of the left electrode.

IV. THEORETICAL

In order to explain qualitatively the experimental findings, model calculations for the ballistic tunnel current were performed, based on *ab initio* electronic-structure calculations. A model was chosen in favor over sophisticated *ab initio* transport calculations in order to pinpoint the physical origin of the effect.

Ab initio electronic-structure calculations for Mn/Fe(001) with film thicknesses up to 12 ML (for details see Ref. 10) provide the input for the model calculations. The one-dimensional model for the MTJ comprises an Fe electrode (left electrode in Fig. 3), the Mn film, the vacuum barrier, and a second Fe electrode (right electrode). The interlayer spacings were taken as in the *ab initio* calculations. The spin-split electronic states in the electrodes were obtained within the nearly free electron model, fitting the effective electron mass and the band offsets to the Fe band structure. Taking, however, the mass of free electrons instead of the fitted ones, changed the shape of the spin contrast. In other words, the Fe bands are poorly described by free-electron parabolas. For the layer-wise antiferromagnetic Mn film, a Kronig-Penney model was adopted[21] (the potential steps were chosen as to reproduce representative Mn bands). The vacuum barrier was taken as a nonmagnetic step-shaped potential. In order to keep the model as simple as possible a sloping barrier (due to a nonzero bias voltage) was abandoned. We are aware that an improved barrier shape would slightly change the bias dependence of the TMR. It is, however, irrelevant for the general explanation of the effect.

It is known that essential contributions to the conductance of MTJs with Fe electrodes arise from nonzero in-plane wave vectors (see, for example, Refs. 14 and 15). Therefore, instead of taking the Fe and Mn band structures at the center of the two-dimensional Brillouin zone ($\vec{k}_{\parallel}=0$), bands at nonzero $\vec{k}_{\parallel}$ were taken for the fit. Here, we choose $\vec{k}_{\parallel}$ from a ring with 0.15 Bohr^{-1} radius since for these $\vec{k}_{\parallel}$ values the highest transmission was found (cf. Refs. 14, 15, and 22). We note in passing that the presented results do not change significantly with the actual value of $\vec{k}_{\parallel}$ within this ring. Further note that the fits of the electronic structure are valid only in a comparably small energy range around the Fermi energy (about -1.0 eV to $+0.6$ eV). Hence, the range of bias voltages of the experiment could not be covered fully.

Subsequently, the total wave function of the MTJ with scattering boundary conditions was constructed by matching of the Fe, Mn, and vacuum wave functions at the respective interfaces.[23] Note that by this means all parameters of the model are fixed by the geometry and the *ab initio* results for the electronic structure. The spin-resolved conductances of the magnetic tunnel junction in parallel and antiparallel magnetic configuration of the electrodes were computed within the Landauer-Büttiker theory from the transmission probabilities of the scattering channels in the Fe electrodes.[24] The bias voltage was taken into account by shifting the potential, and hence the Fermi energy, of the right Fe electrode. The spin-resolved current was eventually obtained by integrating the conductance over the "energy window of tunneling" between the Fermi levels of the two electrodes.[25]

We would like to emphasize that the purpose of the model calculations is only to explain the main statements of the experiment, namely a small but nonzero TMR in the entire range of bias voltages and a change of sign upon increasing the number of Mn layers by one. Details of the experiment cannot be reproduced this way.

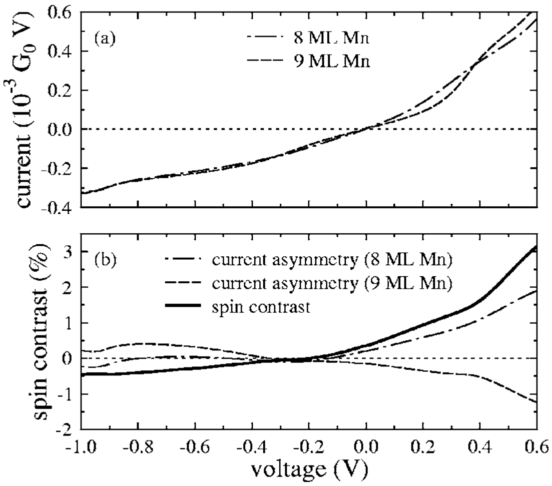

FIG. 4. Spin-dependent tunneling, obtained from the model calculations. (a) Averaged current I_{av} vs bias voltage for MTJs with Mn films of 8 ML (dashed-dotted) and 9 ML (dashed) thickness. G_0 denotes the quantum of conductance, e^2/h. (b) Current asymmetry for 8 ML (dashed-dotted) and 9 ML (dashed) as well as the spin contrast SC (solid).

V. EXPLANATION OF THE TMR

As typical examples, theoretical results are presented for 8 ML and 9 ML Mn films, i.e., in the range of the film thicknesses in the experiment. In a first approximation, the current-voltage characteristics $I_{\text{av}}(V)=[I_{\text{P}}(V)+I_{\text{AP}}(V)]/2$ are almost linear for both Mn film thicknesses [Fig. 4(a)] but reveal a slight increase of the slope for positive voltages. P and AP denote the parallel and the antiparallel orientation of the magnetization in the Fe-tip electrode as compared to the magnetization direction of the Fe substrate. The current asymmetry obtained from the currents for P and AP configurations are very small [Fig. 4(b)]. Further, both spectra are similar except for a change of sign, and both current asymmetries are zero at about -0.3 V. The spin contrast C [solid in Fig. 4(b)] is defined as the difference of the current asymmetry calculated for 8 ML and 9 ML Mn, as in the experiment. It displays the same main features as in the experiment, especially compared to the data obtained for the large tip-sample distance (Fig. 2, ●). In particular, the same order of magnitude is found (a few percent) and it changes sign at -0.3 V.

We note in passing that the theoretical results are almost independent of both the vacuum-barrier thickness and of the Mn film thickness, unless for films very few ML thick. The shape of the spin contrast depends on the Fe bands: e. g., free electrons (instead of nearly free electrons with the fitted band offsets and effective masses) give a different shape and, hence, fail to explain the experimental findings. The general conclusion, however, to observe a TMR at layer-wise antiferromagnets that is caused by the symmetry breaking at the surface is independent of the used model description for the electrons.

From the above it is evident that the model reproduces the essential findings of the experiment and, hence, contains expectedly the ingredients that explain these. We now provide an explanation of our findings.

Within the Jullière model,[4] introduced in the introduction, the experimental finding of a small but nonzero TMR cannot be explained. In the improved model by Maekawa and Gafvert[13] interface and surface properties become more important. For the present case of MTJ's involving Fe/Mn, the question arises what interface is the most important. From first-principles calculations it is known that for thicker Mn films the electronic structure at the Fe-Mn interface and in the interior of the Mn films does not change significantly with Mn-film thickness.[10] In addition, the Mn/vacuum and the vacuum/Fe interfaces are also expected not to change with increase of the Mn-film thickness. These arguments suggest to explain our findings by the outermost Mn layer, the most important property of which is to reverse the magnetization orientation upon altering the Mn film thickness by one monolayer.

Two completely spin-polarized currents $I_{\uparrow}$ and $I_{\downarrow}$ (two-current model) flow from the Fe electrode towards the Mn film. In each Mn layer, both currents become damped in a spin-dependent manner such that after an even number of Mn layers both currents are damped identically. Hence, one would observe the same TMR for any Mn film with an even number of Mn layers. However, for an odd number of Mn layers this symmetry is broken, leading to an even-odd effect in the TMR. In other words, the TMR can be regarded as defined for the magnetization orientations of the Mn top layer and of the STM tip, in contrast as being defined by the magnetization orientations of the Fe electrodes. Thus,

$G_{\text{even}}(\text{P})=G_{\text{odd}}(\text{AP})$ and $G_{\text{even}}(\text{AP})=G_{\text{odd}}(\text{P})$ for Mn films with an even and odd number of layers, where G is the conductance. The same relations hold for the currents and, therefore, the current asymmetry changes sign upon altering the magnetization orientation of the topmost Mn layer.

In the Landauer-Büttiker theory for ballistic transport, especially in the model used here, surface states or states localized in the top Mn layer do not contribute to the tunnel process (because these states are orthogonal to the scattering states in the electrodes). Thus, the TMR is not due to surface states and shows up at all bias voltages. But the qualitative agreement between theory and experiment for large tip-sample distances, keeping in mind the simplicity of the model, suggests that the ballistic transport is governed by electronic states near the Brillouin zone center.

For smaller tip-sample distances however (Fig. 2, ▲), the experimental data show an enhancement of the spin contrast at about zero bias that is not observed in the model calculations. This finding suggests to explain the enhancement of the experimental spin contrast by a spin-polarized surface state. *Ab initio* calculations of the spin-resolved spectral density of state show a spin-polarized surface state close to the Fermi level that consists mainly of p_x and p_y orbitals.[10] Due to the strong localization of this state within the plane of the sample surface (i. e., the Mn film), the influence of this surface state may be significant only at small tip-sample distances.

VI. CONCLUSION AND OUTLOOK

Spin-polarized scanning tunneling microscopy was used to study the magnetization-dependent tunnel process in a MTJ including an antiferromagnetic film. Because of the possibility to probe the tunneling locally, the spin-dependent tunnel current on oppositely magnetized layers of a layer-wise antiferromagnet could be studied. Practically over the whole investigated voltage range a nonzero spin contrast was observed. A change of sign of the spin contrast is present at about −0.3 V. For small tip-sample distances, an enhancement of the spin contrast is found close to 0 V.

The experimental findings are interpreted on the basis of model calculations. The calculations reproduce the size and shape of the bias dependence of the spin contrast explaining the results by means of bulk electronic states in the Mn film and by the symmetry breaking at the Mn surface. In particular, the TMR is essentially due to the magnetization in the outermost Mn layers. Spin-polarized surface states as source for spin contrast become relevant only at small tip-sample distances.

The model calculations presented here can only provide a qualitative understanding of the observed TMR. For a quantitative understanding, however, sophisticated transport calculations that are based on first-principles electronic-structure calculations are desirable. Assumptions on which the model calculations are based can be checked this way.

ACKNOWLEDGMENTS

The authors are grateful to Peter Bose (University Halle and International Max Planck Research School for Science and Technology of Nanostructures) and Ingrid Mertig (University Halle) for fruitful discussions.

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[26] The magnetization of the tip was switched with a high frequency so that the average tunnel current was independent of the direction of the magnetization of the sample. Therefore, the distance between tip and sample was not influenced by the direction of the magnetizations.
[27] The topographic STM images show no significant dependence on the value of the tunnel current.