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Absence of Zero-Bias Anomaly in Spin-Polarized Vacuum Tunneling in Co(0001)

H. F. Ding, W. Wulfhekel, J. Henk, P. Bruno, and J. Kirschner
Max-Planck Institut für Mikrostrukturphysik, Weinberg 2, D-06120 Halle, Germany

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In a joint experimental and theoretical study, we investigate the bias-voltage dependence of the tunnel magnetoresistance (TMR) through a vacuum barrier. The TMR observed by spin-polarized scanning tunneling microscopy between an amorphous magnetic tip and a Co(0001) sample is almost independent of the bias voltage at large tip-sample separations. Whereas qualitative understanding is achieved by means of the electronic surface structure of Co, the experimental findings are compared quantitatively with bias-voltage dependent first-principles calculations for ballistic tunneling. At small tip-sample separations, a pronounced minimum in the experimental TMR was found at +200 mV bias.

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Nowadays, spin-polarized tunneling (SPT) is of particular interest due to its applications in magnetic tunnel devices and in magnetic imaging [1–3]. The field of SPT was opened in the early 1970s, when Tedrow and Meservey studied electron tunneling between a ferromagnet and a superconductor through an amorphous barrier [4] and Müller *et al.* analyzed the spin polarization of field-emitted electrons from EuS coated W tips through vacuum [5]. After Jullière had found the tunnel magnetoresistance (TMR) effect, i.e., the dependence of the tunneling resistance on the relative orientation of the lead magnetizations [6], SPT was studied intensively. Recently, magnetic tunnel junctions with reproducible characteristics at room temperature were fabricated, allowing one to elucidate the underlying physical mechanisms [3,7–15].

However, SPT is still far from being completely understood. Because of its importance for applications, a large number of studies were devoted to the so-called zero-bias anomaly, i.e., the decrease of the TMR with increasing bias voltage in planar junctions [9,16–21]. Junctions made of the same electrodes but with different insulating spacer materials or even with identical but differently prepared insulators vary considerably concerning the voltage dependence of the TMR [6,7,12,20]. With the advances in sample preparation, especially of the barriers, the bias voltage which is sufficient to halve the TMR increased from 3 mV [6] to 500–700 mV [12,20].

To explain this still puzzling behavior, several models were proposed, which have in common that they relate the TMR to the spin polarization. First, biasing of the metal-insulator-metal junctions at finite temperature leads to elastic tunneling of electrons mostly from the Fermi energy of the negative electrode into unoccupied states of the positive electrode [22,23]. The energy dependence of the density of states (DOS) in the positive electrode causes variations of the spin polarization that translate into TMR variations [24]. This DOS effect was observed in crystalline junctions [10]. Second, hot electrons from the positive electrode might be scattered in a spin-dependent way by local magnetic moments at the interfaces [9] or might create magnons [17]. Both effects reduce the spin polarization and, consequently, lower the TMR. Finally, Zhang and White suggested that incoherent tunneling due to scattering at impurities or defects located in the barrier reduces the spin polarization because the electrons tunnel via trap states [16]. This model was supported by both experiment and theory [12,18,21].

Until now, a consistent picture of the zero-bias anomaly has not been achieved. The difficulties are partly related to the complex structure of the tunnel junctions which often comprise poorly characterized amorphous barriers that bring about higher-order SPT effects [16,19,20] and complicate the theoretical treatment. However, by replacing the insulator barrier by vacuum, one can rule out defects in the spacer. Because the DOS effect, spin scattering at the interfaces, and magnon creation remain, SPT through a vacuum barrier allows one to identify the responsible mechanisms for the bias-voltage dependence.

In this Letter, we report on a joint experimental and theoretical investigation of the TMR between a Co(0001) sample and an amorphous CoFeSiB tip through a vacuum barrier as a function of bias voltage. By means of a spin-polarized STM (SP-STM), a strong drop of the experimental TMR with bias voltage is not found. To explain qualitatively this absence of the zero-bias anomaly, we performed first-principles calculations of the electronic structure of semi-infinite Co(0001). Calculations for ballistic tunneling through planar Co(0001) junctions, which take into account the bias voltage, corroborate our experimental findings quantitatively. To summarize at this point, the zero-bias anomaly can be attributed to scattering of electrons at defects in amorphous barriers. Magnon creation appears to be less important.

Experimental.—All experiments were performed in an ultrahigh vacuum chamber ($p = 5 \times 10^{-11}$ mbar) equipped with a SP-STM and standard surface characterization techniques [2,25,26]. The single-crystalline Co(0001) sample and the magnetic tip were cleaned *in situ* by Ar^+ sputtering. During scanning, the longitudinal magnetization of the tip was switched periodically. The resulting variations of the tunneling current were detected with a phase-sensitive lock-in amplifier, allowing one to map the magnetic structure of the sample. Measuring this way the average tunneling current and its modulation between opposite tip magnetizations, the TMR was determined *directly*, in analogy to the experiments by Jullière. This approach differs from recent SP-STM experiments by Bode *et al.* [1], in which the differential conductance was measured. Their dI/dV spectra contain information on the spin polarization as well as on the local DOS and stress the role of spin-polarized surface states in vacuum tunneling.

Tunneling images of both the topography and the magnetic structure were recorded simultaneously at room temperature. The typical dendriticlike perpendicular domain pattern of Co(0001) was observed, similar to that seen with standard magnetic-imaging techniques [27–29]. Applying an external magnetic field, the domain pattern could be displaced while the topography image did not move, thus proving the magnetic origin of the contrast. Magnetic contrast due to mechanisms similar to magnetic force microscopy was ruled out [28] and changes of the tip-sample distance due to magnetostriction were experimentally and theoretically shown to be less than 0.1 pm [30]. Exposure of a few Langmuir of oxygen or deposition of a few monolayers of Au on Co(0001) caused a fading of the magnetic contrast, as was observed for SPT in Au seeded planar tunnel junctions [31].

To study the TMR as a function of the applied bias voltage, we zoomed into a small area and recorded the magnetic contrast (which is proportional to the TMR). In each pixel of the images, the feedback loop was opened for a short time such that the tip position was fixed. The bias-voltage U was ramped while measuring the averaged and the modulated tunneling currents $I_t(U)$ and $\Delta I(U)$, respectively. The TMR, defined as the asymmetry δ of the tunneling currents for opposite tip magnetizations ($\uparrow$ and $\downarrow$), was calculated from the ratio of these two currents averaged over about 1000 pixels,

$$\delta(U) = \frac{I_\uparrow(U) - I_\downarrow(U)}{I_\uparrow(U) + I_\downarrow(U)} = \frac{\Delta I(U)}{2I_t(U)}. \tag{1}$$

Since the TMR is proportional to the scalar product of the tip and the sample magnetizations [32,33], the relative change of the magnetic contrast in small-scale images fully reflects the TMR obtained from parallel (P) and antiparallel (AP) magnetic configurations, although the sample magnetization is oriented only slightly out of the surface plane (typically 10°) [27,29].

Theoretical.—To explain qualitatively and quantitatively the experimental results, we performed first-principles relativistic Korringa-Kohn-Rostoker (KKR) calculations for bulk Co and semi-infinite Co(0001). Besides band structures and layer-resolved spectral densities that provide qualitative insight, we obtained quantitative support by TMR calculations for a planar junction of two Co(0001) surfaces. Conductances G for P and AP magnetic configurations were computed within the layer-KKR scheme of tunneling proposed by MacLaren and co-workers [34]. The time-consuming Brillouin-zone integration of the transmission was carried out by adaptive mesh refinement [35]. In order to treat a bias voltage between the leads in a simple model, the inner potentials of the leads were kept fixed but differed by the voltage drop ΔE. The potential of the tunnel barrier was taken as a superposition of two surface barriers [36]. Hence, this barrier interpolates smoothly between the respective surfaces. Note that the height of the tunnel barrier is determined by the distance d between the leads and by the bias-voltage ΔE. This model should be valid in first approximation for small ΔE and for large d. The TMR is defined in analogy to Eq. (1) in terms of the averaged conductances $G_{\mathrm{av}} = \int_{\Delta E} G(E) dE/\Delta E$. Note that the theoretical TMR is symmetric with respect to the bias voltage due to identical leads, in contrast to the experiment (surface vs STM tip).

Results and discussion.—The measured TMR [37] [Fig. 1(a)] obtained with the tip stabilized at 1 V, 1 nA ($\approx$ 7 Å above the sample surface) appears to be almost constant for bias voltages up to ±1 V. This absence of the zero-bias anomaly is in clear contrast to the case of planar tunnel junctions with insulating spacers. If spin-dependent scattering at the interface magnetic moments and at magnons were the prominent mechanisms for the decrease of the TMR, the latter should also be present in our case. Its absence, however, indicates that these mechanisms are not dominant.

The probability for an electron to tunnel coherently through a barrier with height V_b is proportional to $\exp(-2d\sqrt{2V_b + \vec{k}_\parallel^2})$ [23,38]. Because it decays stronger with tip-sample distance d for large transverse crystal momentum $\vec{k}_\parallel$ than for a small one, it gets "focused" at the Brillouin-zone center for large d. Therefore, we concentrate in the following on the electronic structure of the Γ–Δ–A direction ($\vec{k}_\parallel = 0$). The band structure [Fig. 2(a)] shows two very close but spin-orbit

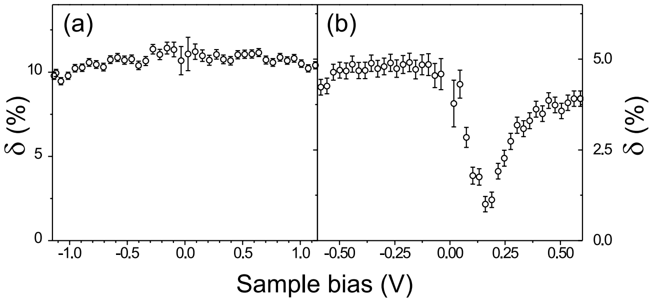

FIG. 1. Tunnel magnetoresistance δ and its error of a clean Co(0001) surface vs bias voltage U, obtained with a magnetic tip stabilized at 1 V, 1 nA (a) and at 100 mV, 1 nA (b).

split minority bands (white) ranging from slightly below the Fermi energy (0 eV) up to about 1 eV. Since their spin polarization P is almost constant ($P \approx -0.99$ with variation less than 0.01), an almost constant TMR is expected, in agreement with the experimental findings. For negative bias, the TMR is expected constant as well because the electrons tunnel into the amorphous tip, the spectral density of which should possess no sharp features. Summarizing, the bulk electronic structure corroborates qualitatively our experimental findings for large d.

To support quantitatively the experimental finding, we present in Fig. 3(a) the calculated averaged tunneling conductances G_{av} integrated over the whole Brillouin zone. As expected for a symmetric junction, these are larger for the P than for the AP configuration. Further, they decrease by almost 2 orders of magnitude when increasing d from $2d_0 = 4.07$ Å to $3d_0 = 6.11$ Å ($d_0 = 2.035$ Å, the Co interlayer distance). In agreement with the preceding arguments, the TMR is almost constant [Fig. 3(b)].

The TMR can be considerably changed for small barrier widths because electronic states with large $\vec{k}_\parallel$ can contribute significantly to the tunneling current (cf. the argument above) and tunneling via surface states can become important also (for STM experiments, see, e.g., Ref. [38]). Figure 1(b) presents the experimental TMR vs bias voltage obtained with the same tip as used for large separations, but at a smaller tip-sample separation (≈ 5 Å; feedback conditions: 100 mV, 1 nA). In this case, the bias voltage was limited to a smaller range (from −600 mV up to +600 mV) to avoid saturation of the $I(V)$ spectra. For negative bias, a constant TMR was still observed, in agreement with the expected tip electronic structure. For positive bias, however, the TMR showed a strong minimum at 200 mV and was reduced above 400 mV. As the bulk states along the Γ–Δ–A direction ($\vec{k}_\parallel = 0$) are mostly of minority character, one can speculate whether the dip is related to majority states with large $\vec{k}_\parallel$ or to a majority surface state reducing the TMR. Indeed, inverse photoemission measurements revealed a surface state in Co(0001) at about 200 meV [39,40] which is also found in our calculations [black arrow in Fig. 2(b)] [41]. At small tip-sample separations, the tunneling probability through this surface state might be enhanced, so as to decrease significantly the TMR. This mechanism could qualitatively explain the constant TMR for large tip-sample separations and the minimum at small tip-sample separations. Our tunneling calculations cannot provide direct support because surface states lie in a bulk band gap and, therefore, do not contribute to the ballistic conductance. Nevertheless, it is conceivable that the surface state contributes via scattering at steps or other defects at the surface which breaks the $\vec{k}_\parallel$ conservation in ballistic tunneling [42] and by this reduce the TMR. We note in passing that the TMR dip reported here is not related to a drop in the TMR asymmetry between forward and backward biasing observed by LeClair *et al.* for a Co tunnel junction with insulating spacer [43]. That feature was related to bulk states and not to a

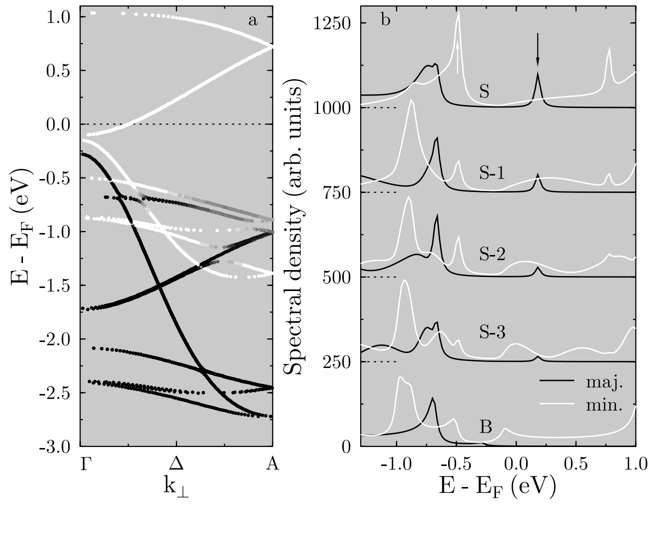

FIG. 2. (a) Spin-resolved relativistic band structure of bulk hcp Co along the Γ–Δ–A direction (i.e., $\vec{k}_\parallel = 0$). The sliding grey scale of the filled circles reflects the spin polarization P: "majority" ($P \approx 1$) black; "minority" ($P \approx -1$) white. (b) Spin- and layer-resolved spectral density of Co(0001) at $\vec{k}_\parallel = 0$ for the first four surface layers (S, S-1, …, S-3) and a bulk layer (B). The arrows mark surface states.

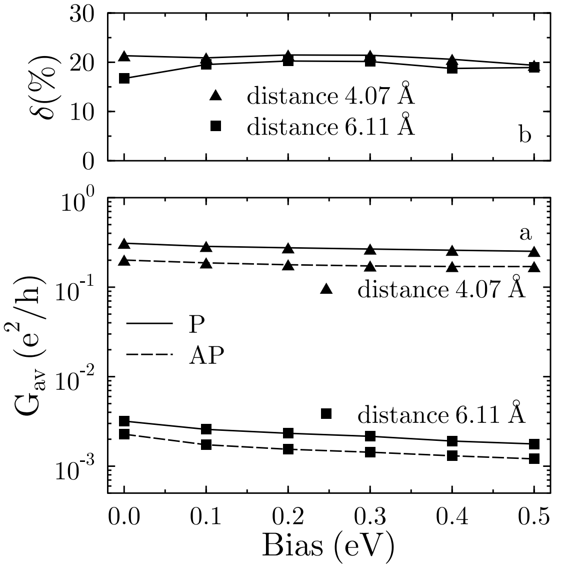

FIG. 3. Theoretical spin-dependent tunneling through Co(0001) planar junctions. (a) Averaged conductances G_{av} in logarithmic scale for P (solid lines) and AP (dashed) configurations at various barrier widths d (triangles: $d = 2d_0$, squares: $d = 3d_0$; in units of the Co interlayer distance d_0) vs bias voltage. (b) Tunnel magnetoresistance δ obtained from the data shown in (a).

surface state. Eventually, the minority surface state at about -430 meV which accounted for a considerable increase of the TMR in spin-polarized scanning tunneling spectroscopy [44] is not expected to change significantly the TMR of the total tunneling current because its spin polarization is similar to that in the bulk states [white arrow in Fig. 2(b)]. This is a striking difference to the surface state at 200 meV.

Conclusions.—Investigating the electron tunneling between a Co(0001) surface and an amorphous tip across a vacuum barrier with a spin-polarized STM, we observed an almost constant tunneling magnetoresistance with bias voltage for large tip-sample separations, i.e., no zero-bias anomaly. Thus, the zero-bias anomaly in planar tunnel junctions with insulator barriers can be attributed to defect scattering in the barrier, rather than to magnon creation or spin excitations at the interfaces. First-principles calculations for the electronic structure and for ballistic tunneling including the bias voltage corroborate our experimental finding qualitatively and quantitatively. For small barrier width, a drop in the TMR occurred at $+200$ mV bias voltage, which is likely related to a majority surface state of Co(0001).

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