Spin-Polarized Vacuum Tunneling into the Exchange-Split Surface State of Gd(0001)

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(Received 17 June 1998)

Spin-polarized tunneling is demonstrated on Gd(0001) thin films using ferromagnetic probe tips in a low-temperature scanning tunneling microscope. The magnetic field dependent asymmetries in the differential tunneling conductivity are found at bias voltages which correspond to the energies of the spin components of the exchange-split Gd(0001) surface state. Maps of the spatial variation of the asymmetry reveal the magnetic structure of Gd(0001) thin films with a lateral resolution better than 20 nm. It is found that magnetic tip coatings thicker than 100 monolayers Fe may modify the sample domain structure due to the stray field of the tip. [S0031-9007(98)07602-9]

PACS numbers: 75.70.Kw, 07.79.–v, 75.25.+z

The spectroscopic capabilities of the scanning tunneling microscope (STM) [1] open up the fascinating possibility of correlating the local structural and electronic properties on the atomic scale with magnetic properties. In the past many attempts have been made to gain magnetic information by making the STM sensitive to the spin of the tunneling electrons. Basically two different concepts have been used to achieve spin-polarized vacuum tunneling.

(i) By using magnetic STM probe tips the spin-valve effect [2] can be exploited which relies on the fact that the tunneling conductance between two ferromagnetic electrodes separated by an insulating barrier depends on whether the magnetic moments are directed parallel or antiparallel. This effect has been extensively studied in planar tunneling junctions [3–5] and has been used in STM to probe the topological antiferromagnetic order of a Cr(001) surface by means of a CrO_2 tip [6].

(ii) Optically pumped GaAs tips enable spin-polarized vacuum tunneling to be observed [7] and this technique has been applied to image the magnetic domain structure of thin Co films [8].

All of the experiments performed to date are limited by the need to separate topographic, electronic, and magnetic information in the case of magnetic probe tips and to eliminate thermal or film thickness induced effects in the case of semiconducting tips.

Our experimental approach to overcome these difficulties is based on tunneling into the well-known surface state of Gd(0001) which is exchange split into a filled majority and an empty minority spin contribution [9,10]. In analogy to the low-temperature experiments performed with ferromagnet-insulator-superconductor planar tunneling junctions [11,12] where the quasiparticle density of states of superconducting aluminum is split by a magnetic field into spin-up and spin-down parts, we use two spin-polarized electronic states with opposite polarization to probe the magnetic orientation of the sample relative to the tip. We demonstrate spin-polarized tunneling by measuring the asymmetry of the differential tunneling conductivity at bias voltages corresponding to the energetic positions of the two spin contributions of the exchange-split surface state in an external magnetic field. This enables the electronic and magnetic structure information to be clearly separated. By mapping the spatial variation of the asymmetry parameter we have been able to observe the nanomagnetic domain structure of Gd(0001) ultrathin films with a spatial resolution below 20 nm.

The experiments were performed in a UHV system with a base pressure below $p = 5 \times 10^{-11}$ mbar. The preparation of clean W(110) substrates and Gd(0001) films has been described elsewhere [13,14]. In the commercial variable-temperature scanning tunneling microscope (VT-STM) used in this study the sample could be cooled down to 20 K. We used etched W-tips which were flashed *in vacuo* to remove oxide layers. The tips were magnetically coated while held at $T = 600$ K by electron beam evaporation of Fe at $p = 3 \times 10^{-10}$ mbar [15] and subsequently magnetized by fields directed perpendicular to the tip axis produced either by a permanent magnet ($H \approx 40$ mT) or by coils arranged in a Helmholtz configuration ($H \approx 8$ mT). The thickness of the magnetic tip coating Θ_{tip} is given in pseudomorphic monolayers of Fe/W(110). Spectroscopic information is gained by measuring the derivative of the tunneling current I with respect to the applied sample bias U. The so-called dI/dU signal is a measure of the local density of states of the sample below the tip apex [16]. The dI/dU signal is measured with a lock-in technique in which the feedback circuit is switched off and an ac component ($U_{\text{mod}} \leq 20$ mV, $\nu \approx 325$ Hz) is added to the gap voltage U which is ramped linearly and 80–150 values of the lock-in signal are acquired. At the end of the ramp the modulation is switched off and the feedback is reactivated. Typical stabilization parameters before each spectroscopy curve was taken were 0.8 eV and 0.2–0.5 nA. However, we confirmed that the results do *not* depend on these parameters by varying the stabilization gap voltage between $-0.8 \leq U \leq 0.8$ V and the tunneling current between $0.1 \leq I \leq 10$ nA.

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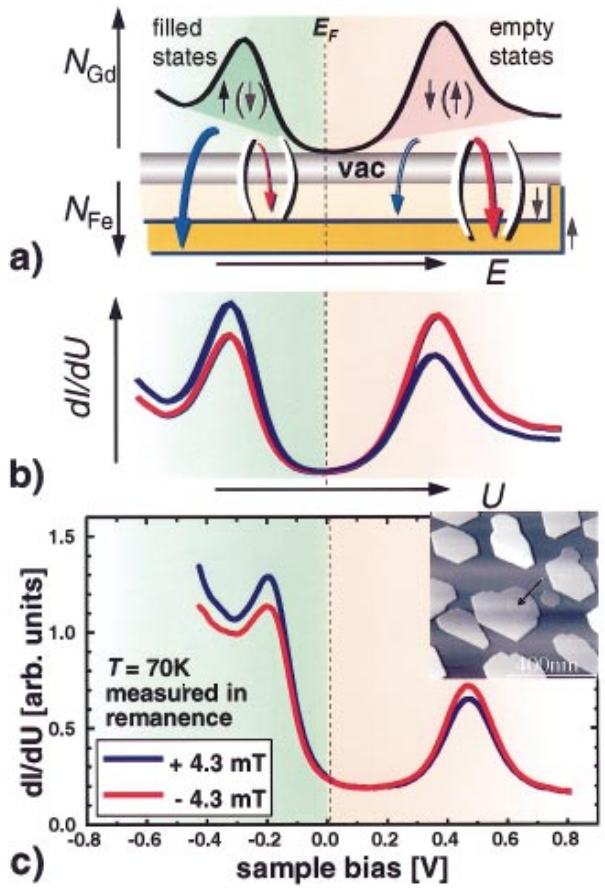

FIG. 1(color). (a) The principle of SP-STS using a sample with an exchange split surface state, e.g., Gd(0001), and a magnetic Fe tip with a constant spin polarization close to E_F: due to the spin-valve effect the tunneling current of the surface state spin component being parallel to the tip is enhanced at the expense of its spin counterpart. (b) This should lead to a reversal in the dI/dU signal at the surface state peak position upon switching the sample magnetically. (c) Exactly this behavior could be observed in the tunneling spectra measured with the tip positioned above an isolated Gd island (see arrow in the inset).

Figure 1 schematically illustrates the principle of spin-polarized scanning tunneling spectroscopy (SP-STS). A sample which exhibits an exchange-split surface state with a relatively small exchange splitting is ideally suited for our experimental approach. If the exchange splitting Δ_{ex} is too large one spin component would be too far from the Fermi level and not accessible by STS, as, e.g., in the case of Fe(001), where Δ_{ex} amounts to 2.1 eV and only the minority band appears as a peak in the dI/dU spectra just above the Fermi level [17]. In contrast, the majority (minority) part of the Gd(0001) surface state at 20 K has a binding energy of -220 meV (500 meV), i.e., the exchange splitting amounts to only 700 meV far below the Curie temperature of 293 K [15].

In the following we consider vacuum tunneling between a Gd(0001) surface [Fig. 1(a) top] and a tip material for which the sign of the spin polarization does not reverse in the energy range of interest, i.e., ± 0.5 eV around the Fermi level. This condition is fulfilled for Fe [18]. For simplicity a constant spin polarization is assumed [Fig. 1(a) bottom]. If the magnetization direction of the tip remains constant we have two possible magnetic orientational relationships between tip and sample, parallel or antiparallel. Since, however, both the majority and the minority component of the Gd(0001) surface state appear in our tunneling spectra, in any case the spins of *one* component of the surface state will be *parallel* with the tip while the *other one* will be *antiparallel*. Therefore, the spin-valve effect will act differently on the two spin components; due to the strong spin dependence of the density of states the spin component of the surface state parallel to the tip magnetization is enhanced at the expense of its counterpart being antiparallel. Consequently, by comparing tunneling dI/dU spectra measured above domains with opposite magnetization we expect a reversal in the contrast at the majority and minority peak position [Fig. 1(b)].

Tunneling spectra measured in an external magnetic field with an Fe-coated probe tip [$\Theta_{tip} =$ 10 monolayers (ML)] positioned above an isolated Gd(0001) island show exactly the expected behavior [Fig. 1(c))]. After inserting the sample in the STM sample holder and cooling down to 70 K it was magnetized in a magnetic field of $+4.3$ mT applied parallel to the sample surface. Subsequently, 128 tunneling dI/dU spectra were measured in remanence with the tip positioned above the Gd island marked by an arrow in the inset of Fig. 1(c). Then the direction of the magnetic field was reversed (-4.3 mT) and further 128 tunneling dI/dU spectra were measured at the same location. This procedure was repeated several times. Figure 1(c) shows the averaged tunneling spectra measured in remanence after the application of a positive or negative field. Comparison of the spectra reveals that for positive field the differential conductivity dI/dU measured at a sample bias which corresponds to the binding energy of the occupied (majority) part of the surface state is higher than for negative field. The opposite is true for the empty (minority) part. We have chosen freestanding Gd islands on W(110) for this experiment since it is known from Kerr-effect measurements [19] that the coercivity is only 1–1.5 mT, i.e., much lower than the applied field. Therefore, one can safely conclude that the magnetization of the sample was switched by the external field while the tip magnetization remained unchanged.

The high spatial resolution down to the atomic scale is the special merit of scanning tunneling microscopy and spectroscopy. We performed spatially resolved measurements at $T = 70$ K with a W-tip coated with 5–10 ML Fe on a sample prepared by depositing 10 ML of Gd on the W(110) substrate held at 530 K. This preparation procedure leads to partially coalesced Gd islands with a Gd wetting layer on the W(110) substrate. The images in Fig. 2 were obtained on a sample in the magnetic virgin state; i.e., it was not magnetized by an external field. The scan range is 2 μm $\times$ 2 μm with 250 $\times$ 250 pixel^2

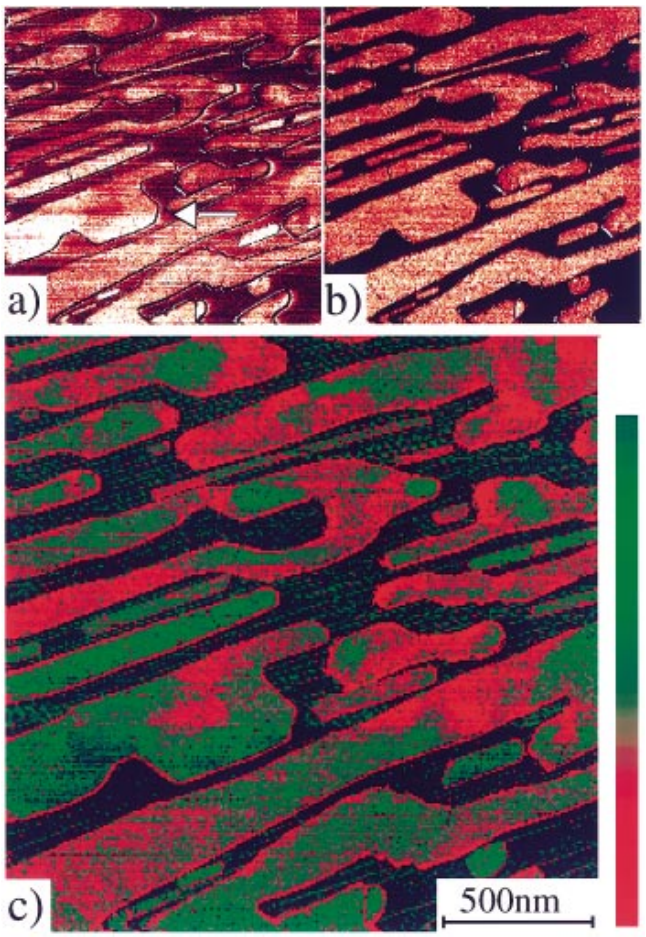

FIG. 2(color). dI/dU image at the majority [(a) $U = -0.2$ V] and the minority [(b) $U = +0.45$ V] surface state peak position. While a strong contrast within the Gd island can be recognized in (a), a weaker and opposite contrast is present in (b). The arrow indicates where an accidental tip change occurred. (c) In the asymmetry image (linear scale bar: 23%) the contrast is enhanced and tip changes less visible.

resolution. At every pixel a dI/dU spectrum was recorded. In Fig. 2 dI/dU images are shown measured at (a) $U = -0.2$ V and (b) $U = +0.45$ V, i.e., sample biases which correspond to filled and empty parts of the surface state, respectively. Since the surface state does not exist on the heavily strained first monolayer of Gd/W(110) its differential conductivity is much lower than above fully relaxed Gd(0001). Consequently, the first ML of Gd/W(110) appears black. Besides obvious tip instabilities (stripes along the fast scan direction) which are most apparent at negative sample bias and indicated by an arrow, a strong contrast on the Gd islands in the dI/dU signal at $U = -0.2$ V [Fig. 2(a)] and a weaker contrast of opposite sign at $U = +0.45$ V [Fig. 2(b)] is visible. In both cases the contrast originates from differences in tunneling spectra similar to those observed upon switching the sample magnetization by an external magnetic field [cf. Fig. 1(c)]. The image shows not always simple green and red contrast but intermediate. This is possible if some of the magnetic domains have a magnetization which is not in line with the Fe spins. We therefore conclude that we observe the magnetic domain structure of the sample.

The influence of sporadic tip changes on the image can be reduced by introducing an asymmetry parameter A which is also used in other spin-sensitive techniques in order to eliminate instrumental artifacts. Let $I_{\mathrm{maj,min}}$ be the intensity of the dI/dU signal measured at the majority (minority) peak position, then $A \equiv (I_{\mathrm{maj}} - I_{\mathrm{min}})/(I_{\mathrm{maj}} + I_{\mathrm{min}})$. Figure 2(c) shows the asymmetry image composed by calculating A at every pixel with Figs. 2(a) and 2(b) as the input data. Obviously, this procedure reduces the influence of tip instabilities and enhances the domain contrast [20].

To evaluate the signal strength and the spatial resolution obtained so far we have zoomed into a detail, i.e., a particular Gd island already visible in the top part of Fig. 2. Again the dI/dU signal for both surface state spin contributions is plotted in Figs. 3(a) (filled) and 3(b)

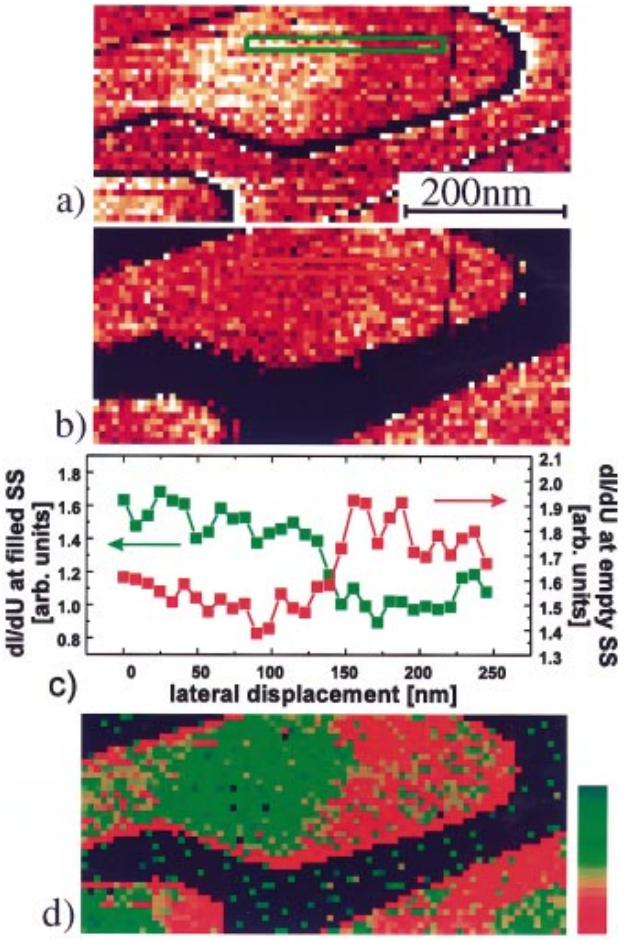

FIG. 3(color). Detail from the data set shown in Fig. 2. (a) In the dI/dU image measured at $U = -0.2$ V the left part of the island is brighter than the right part indicating a parallel alignment of the filled surface state spin part with the tip spin polarization at the Fermi level. (b) dI/dU image measured at $U = +0.45$ V. (c) Plot of the dI/dU signal drawn along the line sections indicated above. (d) Asymmetry image (linear scale bar: 23%) calculated from (a) and (b).

(empty). Both images show a domain wall crossing the island from top to bottom. A closer inspection of line sections drawn along the green and red box from left to right across this domain wall reveals that both the filled and the empty surface state spin part increase and decrease in intensity on a lateral scale below 20 nm, respectively. The spin polarization P of the majority (minority) part of the surface state can be estimated by

$$P = \frac{1}{S} \frac{(I^{\uparrow\ (\uparrow)}_{\mathrm{maj,(min)}} - I^{\downarrow\ (\downarrow)}_{\mathrm{maj,(min)}})}{(I^{\uparrow\ (\uparrow)}_{\mathrm{maj,(min)}} + I^{\downarrow\ (\downarrow)}_{\mathrm{maj,(min)}})},$$

where $I^{\uparrow\ (\uparrow)}_{\mathrm{maj,(min)}}$ and $I^{\downarrow\ (\downarrow)}_{\mathrm{maj,(min)}}$ are the intensity of the dI/dU signal measured at the majority (minority) peak position above different magnetic domains $(\uparrow,\downarrow)$ and S is the spin polarization of the tip. Assuming $S = 0.44$ [12,21] we obtain up to $P \approx 0.45$ (-0.24) for the majority (minority) part of the surface state, which is in good agreement with temperature dependent spin-resolved (inverse) photoelectron spectroscopy data [9,10]. We observed higher spin polarization values when using Fe coated tips which were magnetized by a permanent magnet instead of Helmholtz coils pointing to the fact that a magnetic field of $H \approx 8$ mT as produced by the latter is not sufficient to magnetize the tip to its saturation value.

The influence of the stray field of the probe tip on the domain structure of the sample is a critical parameter which has to be assumed in order to determine the significance of the experimental images. We never observed any indication of tip induced changes of the sample domain structure for Fe coatings with $\Theta_{\mathrm{tip}} \leq 10$ ML. If, however, $\Theta_{\mathrm{tip}} \geq 100$ ML tip-induced magnetic modifications were observed. Figure 4 shows the asymmetry image of Gd(0001) islands grown on a W(110) substrate. The sample was scanned from bottom to top with a tip coated with an Fe film of 100–200 ML thickness. Two blue circles indicate locations where a checkerboard pattern was observed which is characteristic for a magnetization reversal of single Gd islands. Magnetic switching of the tip can be ruled out since other Gd islands are imaged unchanged in subsequent scan lines.

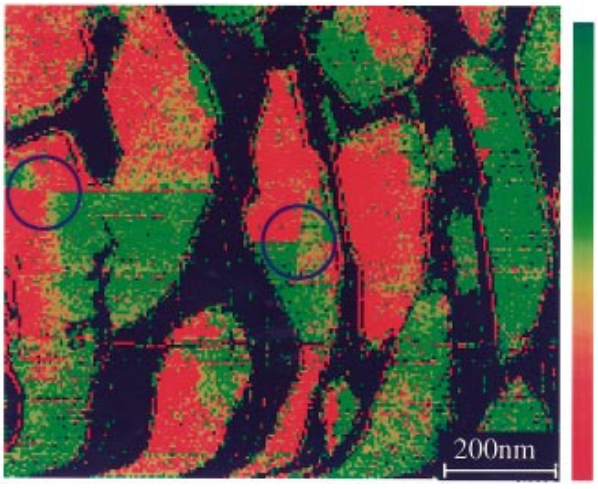

FIG. 4(color). Asymmetry image (linear scale bar: 19%) of a Gd sample measured with a 100–200 ML thick Fe coating on the tip. The blue circles indicate two Gd islands whose magnetization were accidentally switched probably due to interaction between tip and sample mediated via the stray field.

In summary, we have observed spin-polarized tunneling into the exchange split surface state of ferromagnetic Gd(0001). The spin-dependent nature of the signal could be proven by switching the observed contrast on Gd(0001) in an external magnetic field. We could image the domain structure of Gd(0001) islands with a resolution below 20 nm. The thickness of the magnetic tip coating plays an important role for a nondestructive imaging of the sample domain structure.

We gratefully acknowledge financial support from the BMBF and the DFG and R. L. Johnson for critically reading the manuscript.

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