Spin-Polarized Scanning Tunneling Spectroscopy of Nanoscale Cobalt Islands on Cu(111)

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Spin-averaged and spin-polarized scanning tunneling spectroscopy at low temperature was performed on nanometer-scale triangular Co islands grown epitaxially on Cu(111) in the submonolayer coverage regime. Two structurally different island types can clearly be distinguished by their spin-averaged electronic structure. Spin-polarized measurements allow a separation of spectral contributions arising from different island stacking or from opposite magnetization states, respectively. In an applied magnetic field, both island types are found to be magnetized perpendicular to the surface, with large values of saturation field, remanence, and coercivity.

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Co/Cu multilayers are widely used in devices based on the giant magnetoresistance effect. In such applications, high quality interfaces are a crucial requirement. Ultrathin Co films on Cu in (111) orientation have attracted much attention as a system of perpendicular magnetic anisotropy (PMA) [1]. Unfortunately, it turned out extremely difficult to obtain layer-by-layer growth by epitaxy on the (111) face of Cu. Although a small lattice mismatch of -1.9% speaks in favor of a good epitaxy, the much higher surface energy of Co as compared to Cu prevents a formation of smooth Co overlayers [2]. Instead, three-dimensional island formation without any wetting layer, etching processes in the substrate surface [3], and intermixing even at room temperature [4] are found. In particular, an occurrence of stacking faults is held responsible for the lack of a desired strong oscillatory exchange coupling [5,6].

The early stages of epitaxial Co growth on Cu(111) have been the subject of intense research during the past decade. Complementary to spatially averaging techniques [2,4,7,8], scanning tunneling microscopy (STM) has been applied by several groups. In the submonolayer coverage regime, triangular islands were observed protruding to a height of two atomic layers from the Cu surface [9]. In a more recent STM study, Pedersen *et al.* [10] gave evidence that the Co islands are, in fact, three monolayers thick, with the bottom Co layer submerged into the Cu surface. Two distinct island orientations were found, rotated by 180° about the surface normal. It is generally accepted that the orientational variance is due to a stacking fault in one type of the islands, while the other type follows the fcc stacking sequence of the Cu(111) substrate.

Recently, scanning tunneling spectroscopy (STS) has been applied to Co/Cu(111) films by several groups, a local probe method capable of correlating local structural and electronic properties at the nanometer scale, thus being particularly suited for a system as inhomogeneous as Co/Cu(111). The results, however, appeared inconsistent. In a room temperature (RT) study, Vázquez de Parga *et al.* [11] found the two types of islands to be inequivalent in their electronic structures, thereby giving rise to contrasts in maps of the differential tunneling conductance dI/dV. The authors found differences in the local density of states (LDOS) above the Fermi level E_F while below E_F their curves were featureless. Okuno *et al.* [12] observed an occupied spin-polarized state centered at -0.43 eV (throughout this paper, energies are given with respect to E_F) on Co/Cu/Co sandwiches, using a ferromagnetic tunneling tip. Very recently, a combined experimental STS and *ab initio* calculation study was published by Diekhöner *et al.* [13]. Spin-averaged tunneling spectra obtained at low temperature (6 K) showed a very strong and sharp peak at -0.31 eV, which is, according to their calculations, a spin-polarized surface state of $d_{3z^2-r^2}$ symmetry with minority character. In addition, the authors reported a mostly unoccupied free-electron-like surface state of $s-p$ majority character with a parabolic dispersion giving rise to LDOS oscillations on the Co islands over a wide range of energies. With regard to the islands of different orientations, the authors reported no significant difference, except for a tendency that the main peak is centered 0.03 eV lower for one island orientation as compared to the other.

In this Letter, we will show by laterally resolved *spin-averaged* scanning tunneling spectroscopy that (i) the occurrence of a stacking fault indeed leads to a significant change in the electronic structure of the Co islands, causing strong contrasts in dI/dV maps. Further, by laterally resolved *spin-polarized* scanning tunneling spectroscopy (SP-STS) we will investigate the correlation between structural, electronic, *and magnetic* properties of the Co islands, the latter providing an additional source of contrasts. By comparing the spectral curves of islands with different stacking on the one hand and those of islands of opposite magnetic orientation on the other hand, it will be demonstrated that the two sources of contrast can clearly be distinguished. The spin polarization of the tunneling junction is found to be reversed several times within the energy range under study. By measurements with a magnetic field applied, perpendicular magnetic anisotropy of the Co islands is established.

The experiments were performed in a multichamber UHV system equipped with a homebuilt low-temperature ($T = 14 \pm 1$ K) STM described in detail elsewhere [14]. The STM operates inside the bore of a split-coil superconducting magnet capable of magnetic fields up to 2.5 T perpendicular to the sample plane. We used electrochemically etched tungsten tips. For the spin-polarized measurements, the tip was coated with a thin chromium film resulting in a sensitivity to the perpendicular sample magnetization as was verified immediately *before* and *after* the measurements reported below on the well-known Fe double layer system on W(110) [15]. The Cu(111) single crystal was cleaned by repeated cycles of Ar^+ sputtering and annealing at a typical temperature $T = 600\,^\circ$C. 0.7 ML Co was deposited at RT from an e-beam evaporator at a rate of 0.1 ML min^{-1}, with no further anneal.

Despite our efforts to bring the sample to cryogenic temperatures as quickly as possible, a certain degree of intermixing and etching during film deposition seems inevitable, especially in the vicinity of step edges. In the following, we will focus on the properties of bilayer high Co islands with preferably perfect triangular geometry; to our conviction, the interior of these islands is Co, with Cu forming a rim several atoms wide. Two distinct orientations of the triangular islands are observed, rotated by 180° with respect to each other. We have evaluated 1020 islands in a (1 μm)^2 area and found a ratio of 3.5:1. According to the literature [16], the majority of the islands continue the substrate's fcc stacking, while the minority contain a stacking fault.

In a first set of experiments, we have studied the spin-averaged electronic structure of the islands by laterally resolved tunneling spectroscopy using a nonmagnetic tungsten tip. In a scan area of (65 nm)^2, a constant current topograph was taken. Simultaneously, we measured an $I(V)$ curve and also the differential conductance $dI/dV(V)$ at every pixel of the image, i.e., 180×180 individual spectra (stabilization parameters: $I = 1.5$ nA, $V = 0.9$ V).

Results are shown in Figs. 1(a)–1(c). Numerically differentiated $I(V)$ curves are shown in (a), averaged over areas as indicated by boxes in (b). Images (b) and (c) present maps of the dI/dV signal, i.e., spectroscopic layers at voltages as indicated. The green curve (containing the second highest peak) is representative of an fcc island while the red curve (containing the highest peak) belongs to a faulted one. As a reference, also a spectrum of clean Cu is included (grey), showing the onset of the well-known Cu surface state [17] at -0.46 eV as its main characteristics. In the Co spectra, the main features are (i) a strong and sharp occupied peak, similar to the one reported in Ref. [13], and (ii) a second peak about 0.25 eV lower in energy previously not reported in STS experiments [18]. While the general curve shapes for the two types of islands are quite similar, the spectra differ most clearly by a shift in energetic position of the peaks as well as in intensity. On fcc islands, the peak is centered at -0.35 eV while it is found at -0.28 eV on faulted islands. Note that the value of -0.31 ± 0.02 eV given in Ref. [13] was obtained by averaging over a large number of spectra on many islands of different size *and orientation*. The electronic differences become even more obvious from inspecting the dI/dV maps (b) and (c). We have chosen the voltages as to maximize the contrasts between the island types. These are found at values slightly off the peak positions. While in (b) the unfaulted islands exhibit a higher LDOS (corresponding to bright areas) than the unfaulted ones, the contrasts are reversed in (c). This finding clearly proves the electronic structures to be inequivalent for the two types of islands. Similar observations of stacking-dependent electronic structure variations were reported for Gd(0001) islands on W(110) [19].

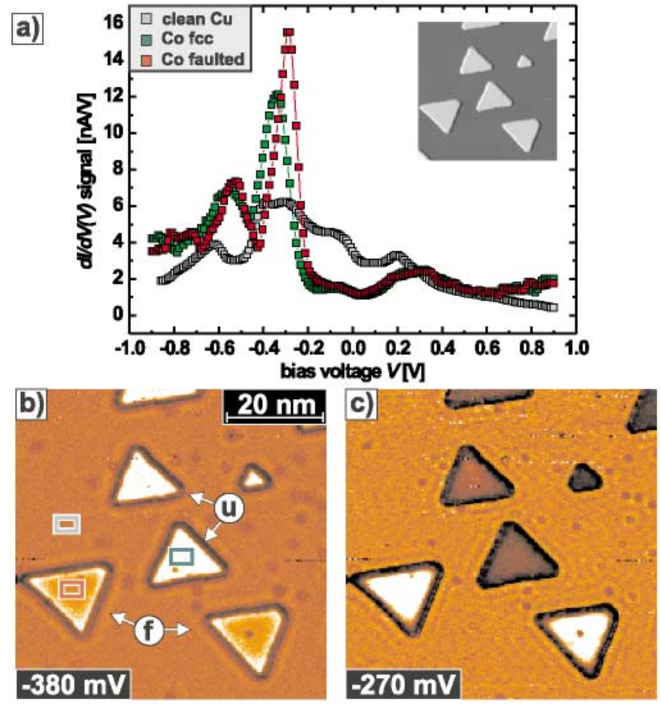

FIG. 1 (color). (a) Spin-averaged tunneling spectra, (b)–(c) dI/dV maps at voltages as indicated. The curves were averaged in the colored boxes in (b), the island types are labeled u (unfaulted) and f (faulted). Spectra of the two island types differ in peak energetic positions and intensities. At the voltages of maps (b)–(c), the contrasts are maximal. Inset in (a): Topographic image.

In a second set of experiments, we used a Cr coated tip sensitive to the perpendicular component of the sample magnetization. Applying the same procedure as described above for the spin-averaged spectroscopy, we again measured one spectrum at every pixel of the image. The results are displayed in Fig. 2. While the spectroscopic features used to discriminate the differently stacked islands in the spin-averaged measurements are found to be

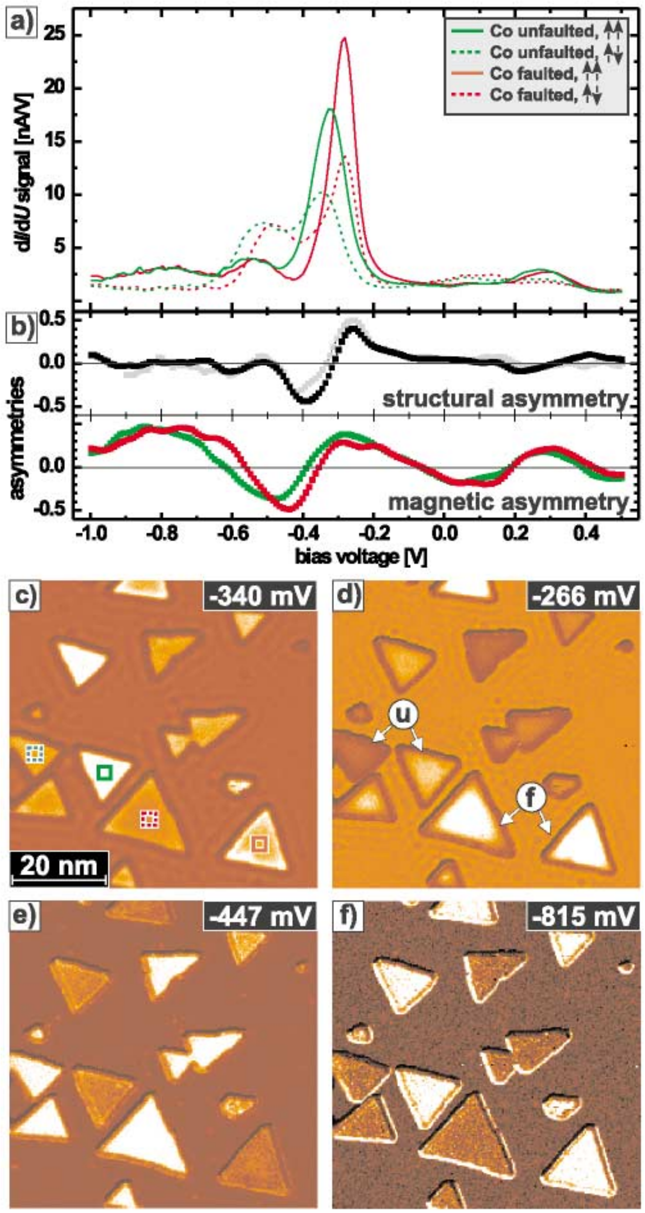

FIG. 2 (color). (a) Spin-resolved tunneling spectra. Arrows ⇈ (⇅) refer to parallel (antiparallel) magnetization alignment of sample and tip. (b) Asymmetries arising from different stacking [upper panel; grey (black) spin averaged (spin polarized)], and from opposite magnetization (lower panel). (c)–(f) dI/dV maps at bias voltages as indicated. Maps (c)–(d) allow a direct comparison to the spin-averaged case in Figs. 1(b) and 1(c). As exemplified by maps (e)–(f), the spin-dependent contribution is clearly dominant in most parts of the energy range. Stabilization parameters: $I = 1$ nA, $V = +0.6$ V.

preserved, we now observe two distinct curves for each island type (solid vs dotted curves). The latter differences are caused by the *magnetic* state of the islands which are magnetized either parallel or antiparallel to the tip magnetization, respectively. This source of contrast was unavailable in the spin-averaged measurements. In Figs. 2(c)–2(f), dI/dV maps at selected energies are displayed. Panels (c)–(d) represent the spin-polarized analogues to Figs. 1(b) and 1(c), emphasizing contrasts due to different stacking, while the voltages for panels (e)–(f) are chosen such that the magnetic contrasts are particularly strong. Comparing Figs. 1(b) and 1(c) to 2(c) and 2(d), we notice that the stacking-dependent contrasts are less clear than in the spin-averaged case. The reason is that, competing with the structural contrast, a spin-dependent LDOS contribution of comparable strength is superimposed. In most parts of the energetic range under study, however, the contrasts are clearly dominated by the magnetic signal. This is exemplified in Figs. 2(e) and 2(f). We note that the contrasts are reversed in the two images. Thus, it is not immediately clear if bright or dark is indicative of a parallel alignment of tip and sample magnetization. SP-STS is sensitive to the spin polarization of the tunneling junction $P_{ts}(E)$, which is a convolution of the polarizations of both electrodes involved, i.e., tip and sample. It is well known that Cr (the tip material) is negatively polarized around the Fermi level [20]. Taking the curves of higher intensities at the Co d_z surface state as indicative of a parallel magnetization alignment (solid curves), we conclude that this sample state is also negatively polarized, in agreement with Ref. [13]; thus P_{ts} is positive in this energetic region. As indicated by intersections of the solid curves with the dotted ones, a sign reversal happens at -0.38 eV (faulted: -0.35 eV) and again at -0.62 eV (faulted: -0.57 eV), i.e., in the region of the second peak. According to recent spin-density functional calculations, this peak has also minority character [21]. Thus, to account for the observed sign reversals of P_{ts}, the additional assumption is required that the tip polarization is reversed at roughly -0.38 eV.

To get a clearer picture of the relevance of the two major sources of contrasts, i.e., different stacking and opposite magnetization, we have plotted the asymmetries $A = (a - b)/(a + b)$ of the respective curves in Fig. 2(b). The upper panel clearly shows the similarity of the structurally caused asymmetries for the spin-averaged (grey) and the spin-polarized (black) case. On the other hand, the magnetic asymmetries as displayed for both island types in the lower panel of (b) follow characteristics being very different from the structural asymmetries. Strong oscillatory behavior is observed over the whole energy range caused by the sign reversal of the spin polarization as discussed above. Guided by these asymmetry curves, any contrasts observed in spin-resolved dI/dV maps at various energies can unambiguously be interpreted.

In order to verify the *magnetic* origin of the observed contrasts, we recorded dI/dV maps in variable magnetic fields applied along the surface normal. In Fig. 3, a selection is presented, $(150\ \text{nm})^2$ in size, acquired at $V_{bias} = -0.18$ V. In the virgin state islands of both orientations are found either dark or bright, corresponding to their magnetization being either parallel or antiparallel to the tip magnetization. A field as strong as 1 T causes only a small fraction of the dark islands, exclusively those

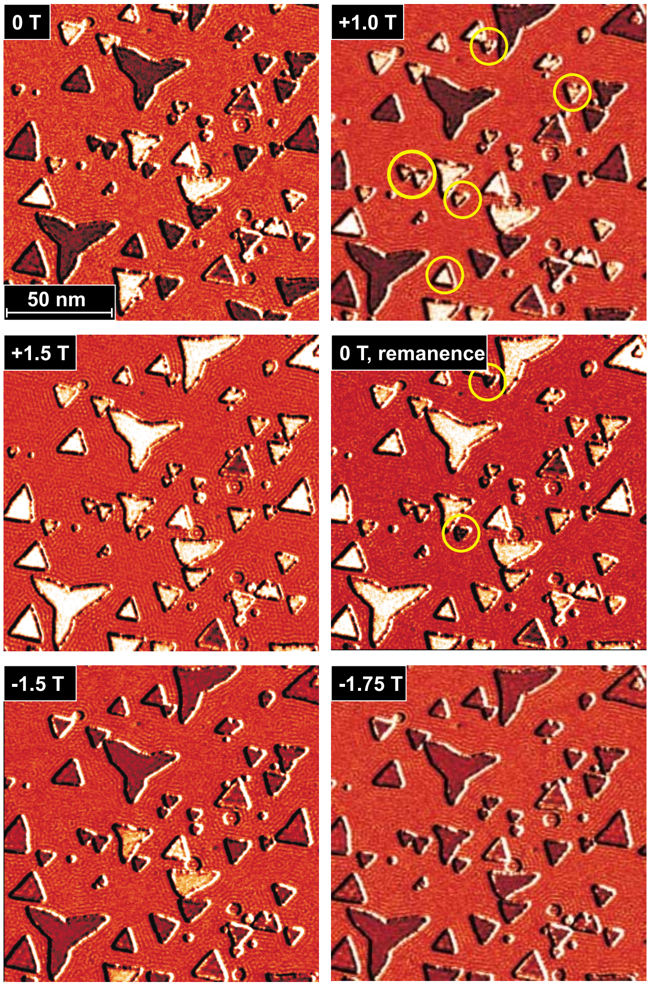

FIG. 3 (color). Spin-polarized dI/dV maps ($V = -0.18$ V) of Co islands in variable perpendicular magnetic fields. At zero field, islands of both stacking types are found bright or dark, corresponding to parallel or antiparallel alignment of sample and tip magnetization, respectively. A field of 1 T switches only a few of the smaller islands (circles). 1.5 T is not sufficient to achieve saturation. After field removal, only two small islands reverse their magnetization (circles), indicating a remanence M_r close to saturation M_s.

of small size, to switch their magnetization so that they are imaged bright (a few of them are marked by circles). A field of 1.5 T is required to switch most islands, still leaving two islands in their initial dark state. Field removal reveals a nearly 100% remanence; only two very small islands are switched (see circles), presumably due to stray fields being particularly strong from close-by larger islands. A reversed field of -1.5 T is required to again switch most of the islands into the opposite direction. Even at -1.75 T the sample is not completely saturated as is found in a larger scan [(350 nm)^2, not shown], where still a few islands are observed being magnetized opposite to the applied field. This finding confirms that the islands are ferromagnetic with strong PMA, exhibiting a remanence-to-saturation ratio M_r/M_s of nearly 1 and a surprisingly high coercivity 1.0 T $< \mu_0 H_c <$ 1.5 T. While Gradmann's discovery of PMA was originally made on Co films capped with Cu [1,22], our observation of PMA for uncovered Co islands is not unexpected in view of the trend reported recently by Camarero *et al.* [23] and earlier by Huang *et al.* [24], who studied Co films of 2 ML and less nominal coverage (i.e., local coverage between 2 and 5 ML) at RT by (laterally averaging) Kerr effect measurements.

In summary, we have studied the spin-averaged and the spin-polarized electronic structure of nanometer-scale Co islands on Cu(111) by scanning tunneling spectroscopy. Two island types of different stacking were clearly recognized by their inequivalent band structures. A detailed analysis of spin-polarized spectral curves provided the information necessary to discriminate between structural and magnetic contrasts in dI/dV maps. Ferromagnetism was established for both island types by measurements in variable magnetic fields, revealing perpendicular magnetization and large values of the saturation field and coercivity.

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[1] U. Gradmann and J. Müller, Z. Angew. Phys. **30**, 87 (1970).
[2] M. T. Kief and W. F. Egelhoff, Jr., Phys. Rev. B **47**, 10 785 (1993).
[3] J. de la Figuera *et al.*, Surf. Sci. **307–309**, 538 (1994).
[4] A. Rabe *et al.*, Phys. Rev. Lett. **73**, 2728 (1994).
[5] S. S. P. Parkin *et al.*, Phys. Rev. B **46**, 9262 (1992).
[6] M. Zheng *et al.*, J. Phys. Condens. Matter **12**, 783 (2000).
[7] B. P. Tonner, Z.-L. Han, and J. Zhang, Phys. Rev. B **47**, 9723 (1993).
[8] V. Scheuch *et al.*, Surf. Sci. **318**, 115 (1994).
[9] J. de la Figuera *et al.*, Phys. Rev. B **47**, 13 043 (1993).
[10] M. Ø. Pedersen *et al.*, Surf. Sci. **387**, 86 (1997).
[11] A. L. Vázquez de Parga, F. J. García-Vidal, and R. Miranda, Phys. Rev. Lett. **85**, 4365 (2000).
[12] S. N. Okuno, T. Kishi, and K. Tanaka, Phys. Rev. Lett. **88**, 066803 (2002).
[13] L. Diekhöner *et al.*, Phys. Rev. Lett. **90**, 236801 (2003).
[14] O. Pietzsch *et al.*, Rev. Sci. Instrum. **71**, 424 (2000).
[15] A. Kubetzka *et al.*, Phys. Rev. Lett. **88**, 057201 (2002).
[16] C. Rath *et al.*, Phys. Rev. B **55**, 10 791 (1997).
[17] M. F. Crommie, C. P. Lutz, and D. M. Eigler, Nature (London) **363**, 524 (1993).
[18] LDOS oscillations similar to those reported in Ref. [13] are also resolved in our dI/dV maps but are not discussed in this paper.
[19] M. Bode *et al.*, Acta Phys. Pol. A **93**, 273 (1998).
[20] C. Rau and S. Eichner, Phys. Rev. Lett. **47**, 939 (1981).
[21] S. Heinze (private communication).
[22] J. Kohlhepp, H. J. Elmers, and U. Gradmann, J. Magn. Magn. Mater. **121**, 487 (1993).
[23] J. Camarero *et al.*, Phys. Rev. B **64**, 125406 (2001).
[24] F. Huang *et al.*, Phys. Rev. B **49**, 3962 (1994).