Spin-polarized scanning tunneling microscopy (spin-STM) has developed into a powerful technique to investigate magnetic properties on the nanometer scale and below. In spite of the numerous results that illustrate the success of spin-STM in nanomagnetism,1–3 details regarding the decisive role of the tip in this technique have not been presented before. The main challenge of spin-STM is the characterization of the magnetic configuration of the tip, in order to draw reliable conclusion about the spin structure of the sample. The magnetic configuration of the tip is expected to be determined by the exact atomic configuration of the tip apex, which is, however, not straightforwardly accessible experimentally. Therefore, further insight into tip preparation,4 how to determine the magnetic sensitivity of a tip,5 and how to disentangle tip and sample contributions to the magnetic contrast6 is called for, and corresponding strategies are presented here.

In this letter, we propose a practical method to characterize the magnetic configuration of tips by measuring hysteresis loops of the differential conductance in an external magnetic field. This is a necessary condition for a reliable analysis of the magnetic contrast in spin-STM.

We have chosen a system for which the magnetization state and the behavior under a magnetic field have been already determined. Co islands, which form upon deposition of Co onto atomically clean Cu(111) at room temperature, present an out-of-plane magnetization direction and show switching fields at 8 K of order 1–2 T depending on island size.7,8 We have performed spin-STM studies of this system at 8 K using a W tip covered by a thin Cr film of 20–100 atomic layers (ALs), as commonly used in spin-STM studies.5 We focus in the following on a Cr thickness of 100 AL, as we found no clear-cut thickness effect on the resulting magnetic response. We have also prepared tips with a Co underlayer, i.e., W/Co(40 AL)/Cr(40 AL), in an attempt to provoke a different tip behavior in response to a magnetic field. We measure differential conductance dI/dV spectra (I: tunnel current and V: voltage tip-sample), as a function of an external field along the sample normal and obtain hysteresis loops.7

We prepare W tips as explained before in Ref. 5 by first etching in NaOH and flashing in ultrahigh vacuum to 2200 K, followed by deposition of a magnetic material onto the tip apex. However, this preparation does not directly ensure magnetic contrast routinely, and an additional microscopic preparation under tunneling conditions is applied. We apply voltage pulses between tip and sample over clean Cu(111), away from the area of interest. We find that pulses from +5 to +8 V with a duration of a few ms are suitable. Before the pulse is applied, the tunnel current is set to a larger value of 30 nA as compared to the current used for imaging of 1 nA. Subsequently the STM feedback loop is switched off for the duration of the pulse. This procedure may cause significant changes of the differential conductance spectra dI/dV. To judge the result of this microscopic preparation, we measure spectra on both Cu(111) and on the Co islands. We continue to use only those tips which show both a clear signature of the Cu surface state at ~0.44 V (Ref. 9) and a clear Co related peak at ~0.3 V.10

One might be tempted to assume that dI/dV spectra offer a sufficiently complete characterization of the tip-sample system to warrant spin-contrast in STM. We show here that this is not the case. Figure 1 presents dI/dV spectra, which were obtained above the center of Co islands with tips covered as indicated. The spectra show as a common major feature a peak around ~0.3 V. This is ascribed to a Co surface state.10 The careful inspection of the spectra in the range from 1 to ~0.3 V reveals some maxima and minima, where amplitude and position differ for curves 1 to 7. These features have not been described theoretically before and they change upon application of voltage pulses. Nevertheless high magnetic contrast is often found in this spectral range.7,8 We will see that the tips labeled 3 to 7 give magnetic contrast while the tips 1 and 2 do not. The gap voltage, where the highest magnetic contrast is observed, is indicated by a vertical marker in Fig. 1.

An important result of Fig. 1 is that tips with the same macroscopic preparation (W/Cr or W/Co/Cr) give rise to different spectral features below ~0.3 V. At these negative voltages, electrons tunnel from the sample into the tip. Thus, our observation implies that unoccupied electronic states of the tip are influenced by the microscopic tip preparation. We propose that this change of electronic and possibly spin-structure is driven by atomic re-arrangements at the tip apex, induced by voltage pulses. One might speculate that this difference in atomic configuration also gives rise to a different magnetization direction at the tip apex. To test this hypothesis, we have performed in-field measurements presented in Figs. 1–3.

Spectra obtained with tip 5 are shown in Fig. 1 at +1 and +1.8 T. The spectra change with field. A spectroscopy image taken with tip 5 of two Co islands is presented in Fig.
Figure 1. (Color) Differential conductance $dI/dV(V)$ spectra measured with seven different tips at the center of two Al high Co islands on Cu(111). Two curves are represented for tip ③, corresponding to two different magnetic configurations at $+1$ T (dotted line) and $+1.8$ T (full line). Curves ②–⑦ have been obtained at fields of $+2$, $+4$, $-2$, $-4$, and $+4$ T, respectively, which ensures parallel alignment of the sample magnetization with the external field. Curve ① has been measured at 0 T. The nominal tip composition is given for every spectrum in AL. Markers indicate the voltage for which the maximum magnetic contrast is obtained. The curves are shifted vertically and scaled differently for clarity, see Fig. 3 for an absolute scale.

2. Figure 2 shows a variation of the $dI/dV$ contrast on the islands at three different magnetic field values. Measurements of a complete hysteresis cycle on the smaller island, shown in curve ⑤ of Fig. 3, reveal the switching field of 1.6 T, therefore, we observe a contrast change between 1.0 and 1.75 T for the smaller island of Fig. 2. The change of contrast of the larger island is also ascribed to its magnetization reversal, which occurs at a larger field of 2.2 T as deduced from a complete hysteresis cycle (not shown). In the example of Fig. 2, the field-driven change of contrast is ascribed to a magnetization reversal of the Co islands. In general, however, also a field driven reorientation of the magnetization direction of the tip needs to be considered, as evidenced in Fig. 3.

Figure 3 shows magnetic hysteresis loops of the $dI/dV$ signal at the voltage given by the markers of Fig. 1. We use the same tips as in Fig. 1 in order to investigate the correlation between spectral features and magnetic response. W tip ① gives rise to a constant signal as a function of field, and its curve is not shown in Fig. 3. A first inspection of Fig. 3 reveals a striking unexpected result. The same macroscopic preparation of either W/Cr or W/Co/Cr gives rise to vastly different hysteresis curves, ②–⑤ and ⑥ and ⑦, respectively. To appreciate the shape of the hysteresis curves in Fig. 3 it is important to realize that the field driven change of the signals represents a corresponding change of the relative orientation between the magnetization directions of sample and tip. The sharp change of the signal in curves ⑤–⑦ around 2 T is ascribed to a switching of the magnetization direction of the islands, whereas the variation of the signal at smaller fields is due to a field driven reorientation of the magnetization direction of the tip apex. However, the W/Cr tip ② does not show any field dependent change within the noise limits, the field induced magnetization reversal of the island remains undetected. This clearly indicates that the deposition of Cr does not necessarily lead to a magnetic contrast.

We observe in Fig. 3 hysteresis curves which are either asymmetric (curve ③) or symmetric (curves ④–⑦) with respect to the y-axis. Curve ⑤ is ascribed to a fixed magnetization direction of the tip apex, which does not change in response to the external field. The symmetric hysteresis curves ④–⑦ indicate that here also the magnetization direction of the tip apex has changed in response to the external field. The hysteresis curves with a W/Co/Cr tip are symmetric ⑥–⑦. The shape can be explained by a continuous rotation of the tip magnetization direction already in very small field (curve ⑥), or by an abrupt switching of the magnetization direction of the tip (curve ⑦) at ±0.5 T. The results presented in Fig. 3 reveal a complex magnetic behavior of the tips, which even varies for tips of the same macroscopic preparation. This indicates that, in addition to the macroscopic preparation, also the necessary microscopic tip preparation by voltage pulses is a further decisive aspect which defines the magnetic contrast in spin-STM.
Note that only the tips characterized by curves ③ and ⑦ give rise to a magnetic contrast for an out of plane magnetized sample in zero field. Tips represented by curves ④–⑥ require the application of an out of plane field to do so. The results of Fig. 3 indicate that the magnitude of the $dI/dV$ signal changes differently for a change of magnetic configurations. A parallel alignment of tip and sample magnetization directions may give rise to a large, or a small $dI/dV$ signal, as compared to that of the antiparallel state. The magnitude of the $dI/dV$ signal and its magnetically induced change depend also on the voltage, at which the hysteresis curve is measured. This is apparent from the two $dI/dV$ spectra of curves ⑤ in Fig. 1, where the crossing of spectra corresponds to an inverted relation between $dI/dV$ signal magnitude and magnetic configuration.

These results are of high relevance, as it has been tacitly assumed that a high signal of the differential conductance corresponds to a parallel alignment of the magnetization direction of tip and sample, while a low signal corresponds to an antiparallel alignment.⑧,⑨,⑩ However, the results presented in Fig. 3 show that only in connection with a field sweep, a reliable deduction of the magnetic configurations from the $dI/dV$ signal is warranted.

A closer inspection of Fig. 3 reveals that the value of the differential conductance and its variation upon a change of the magnetic configuration varies from curve to curve, and it also differs for the same macroscopic preparation. The relative magnetically induced signal change can be as small as $\sim 10\%$ (curve ③), or $\sim 75\%$ (curve ⑤) for the same preparation, or as large as $\sim 100\%$ for a different preparation (curve ⑦). Therefore, a specific magnetic behavior is not linked to a certain macroscopic tip preparation.

In view of the different spectral features shown in Fig. 1 and the different magnetic response extracted from Fig. 3 it appears tempting to link a certain spectral feature with a certain magnetic response of a tip. However, we refrain from doing so. It would be questionable to establish a one-to-one correspondence between a $dI/dV(V)$ spectrum and a specific magnetic response in field. This assessment is corroborated by the curves of tip ④, which produce a hysteresis loop similar to the one of tip ⑤ in spite of a different spectrum. All these observations indicate that neither the material at the tip apex, nor its thickness, nor its spectroscopic features are sufficient parameters to determine the magnetic properties of tips, but field-dependent measurements do so.

In conclusion, we find a large variation of the magnetic behavior of tips used in spin-polarized STM studies. We show that the magnetic configuration of a tip and its change in an external field depends on both the macroscopic preparation by film coverage and on the microscopic preparation by voltage pulses. This suggests that the atomic arrangement of atoms at the tip apex, which is changed upon voltage pulses, is decisive for the spin contrast. The magnetic behavior of a tip is reliably characterized by in field measurements. This characterization of the tip is a prerequisite for a reliable analysis of the magnetic properties of the sample. A decisive benefit of measurements in field is that the magnetic origin of an observed change of contrast can be clearly verified.

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