Atomic structure of tip apex for spin-polarized scanning tunneling microscopy

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We present a high resolution transmission electron microscopy study of a Cr-coated W tip apex prepared for spin-polarized scanning tunneling microscopy (SP-STM). The characterization of the tip apex structure has been done with atomic resolution. We show that the Cr film is epitaxially grown on W and presents a monocrystalline phase. The surface analysis of the apex reveals roughness which gives rise to structures that can be considered as *nanotips*. In spite of the monocrystalline structure of these *nanotips*, we show that their spin arrangement and resulting magnetization direction cannot be controlled. SP-STM measurements on a Cr/MgO(001) sample confirm this conclusion. © *2011 American Institute of Physics*. [doi:10.1063/1.3558920]

Spin-polarized scanning tunneling microscopy (SP-STM) is a technique sensitive to the orientation of the local magnetization of a sample surface. This sensitivity relies on the magnetization direction of the apex of the STM tip. Since early SP-STM works, it is well established that proper tip preparation and characterization is a important first step to be able to measure samples reliably.[1] It has also been proposed that the tip magnetic sensitivity, namely sensitivity to the in-plane or out-of-plane component of the sample magnetization, could change depending on the thickness of the magnetic layer that composes the magnetic tip.[1] Recently, it has been shown that the magnetic properties of a tip are mainly determined by the atomic arrangement of the tip apex, i.e., the very last atoms of the tip which contribute to the tunneling current.[2–5] Indeed, the spin orientation at the apex directly results from its structural atomic arrangement. While SP-STM tip characterization is crucial to analyze spin-polarized measurements, only few studies have focused on this topic. It is intrinsically difficult to determine the tip apex atomic arrangement from STM imaging since images depend on the convolution of both the tip and sample configuration, and hence an alternative experimental characterization method is needed. Recently, field emission microscopy and field ion microscopy studies, associated with spin polarization measurements, have been made on W tips coated with ferromagnetic films.[6,7] Irisawa *et al.*[6] found that the magnetic layer deposited on W is most likely noncrystalline. The orientation of the spin polarization of the tip apex was found to be tilted, neither in-plane nor out-of-plane.

In this letter, we present a high resolution transmission electron microscopy (HRTEM) study of the apex of a Cr-coated W tip prepared for SP-STM, combined with SP-STM measurements done with another identically prepared tip. For the first time, it has been possible to visualize the atomic arrangement of crystalline Cr layers deposited on the apex of an W tip. We show that the W tip apex presents a (110) orientation and that the Cr is epitaxially grown on the apex following the same orientation. The thickness and roughness of the Cr thin film at the end of the tip were also characterized by HRTEM, enabling us to propose a microscopic description of the spin polarization of the tip. In order to assure

the validity of our tip characterization for SP-STM studies, we have performed STM measurements on a model Cr(001) surface and we show that an expected spin polarized contrast is obtained.

Our STM tips were prepared from a 0.25 mm W polycrystalline wire. They were electrochemically etched in a NaOH solution and then passivated in a HF bath. The quality of this etching procedure and the global apex shape was checked by scanning electron microscopy. The tips were then introduced into an ultrahigh vacuum (UHV) preparation chamber connected to the STM chamber. There, they were flashed by Joule heating to temperature up to white color emission. This procedure allows the removal of contaminants and oxides from the apex. Bode[1] suggests that such flashing induces melting of the apex and that a dense-packed surface of W, i.e., W(110), is probably formed at its end. After this treatment, Cr was deposited *in situ* onto the tips by electron beam evaporation at a rate of around one atomic layer per minute. One tip was transferred under UHV to the cold STM head where experiments were perform at 4 K, while the second was taken out from the UHV chamber, glued on a standard TEM copper ring and subsequently analyzed by TEM.

Electron microscopy was perform on a Jeol 2200 FS TEM-STEM microscope equipped with C_S-probe aberration correction. Figure 1(a) shows a TEM image of the overview
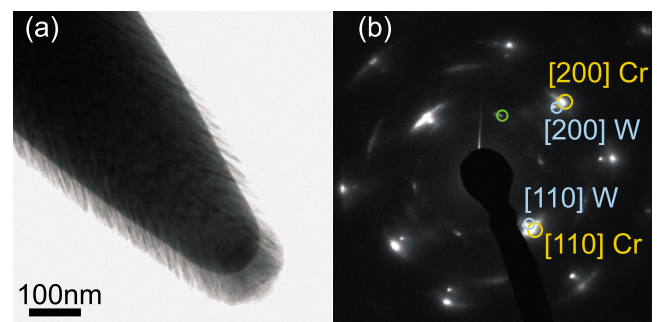

FIG. 1. (Color online) (a) TEM image of a W tip coated by Cr. The dark contrast corresponds to the W and the bright one to the Cr coating layers. (b) Electron diffraction pattern of the tip apex. Two lattices corresponding to Cr and W are visible and labeled, demonstrating the epitaxial relation Cr(110)/ W(110). An additional lattice (not labeled) is also visible and is ascribed to Cr oxide present at the Cr surface.

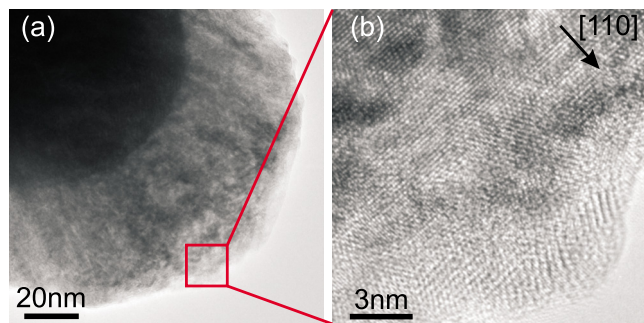a)Electronic mail: guillemin.rodary@lpn.cnrs.fr.

FIG. 2. (Color online) (a) HRTEM image of the W tip apex where the Cr/W interface is clearly visible. The last Cr layers present roughness of few nanometers. (b) Magnification of the red square depicted in (a). Atomic rows are observed until the last Cr layers. The arrow indicates the [110] crystalline orientation.

of the tip end. Due to their large mass difference and relative thickness, different electron transmissions result from W and Cr. The W tip is thus well identifiable by the black contrast and the Cr coating by a brighter layer. This image displays the quality of the Cr deposition at the very end of the tip but also shows columnar structures on the tip sides due to shadow effects during evaporation. The thickness of the Cr layer is determined by TEM imaging to be 50 ± 5 nm, consistent with our evaporation calibration. In Fig. 2(a) we present a HRTEM image of the tip apex and in Fig. 2(b) a magnification of an apex area where atomic layers are clearly observable. Figures 1 and 2(a) evidence the low roughness of the W, while it is obvious from Fig. 2 that the last Cr layers present three-dimensional structures of few nanometers. Figure 1(b) presents an electron diffraction pattern of the tip apex containing both W and Cr. Two neighboring spot families are clearly observable and are indexed on Fig. 1(b). The first one corresponds to the bulk W lattice parameter and the second to bulk Cr, indicating a fully relaxed structure. The diffraction pattern shows unambiguously that an epitaxial relationship exists between Cr and W, with both axial and radial parallel directions: [110]Cr∥[110]W and [100]Cr∥[100]W. This shows that the treated W tip presents a monocrystalline phase at its apex, in spite of the use of an usual polycrystalline W wire, as previously suggested,[1,6] and that the Cr is epitaxially grown on it. The atomic arrangement along the [110] direction is visible in Fig. 2(b), which confirms that the crystalline order remains until the last Cr layers. Chemical analysis have been performed by energy-dispersive x-ray spectroscopy at different zones along the tip axis. Only W and Cr species were detected by this technique (in exception to oxygen near the surface, originating from natural in-air oxidation, in agreement with electron diffraction), confirming the absence of contamination on the tip apex.

The analysis of the HRTEM results implies several considerations on the apex structure important for SP-STM. We can conclude from the observation of the monocrystalline phase of the tip apex that the Cr atoms which contribute to the spin polarized tunneling current forms a Cr(110) surface. This finding is of major importance as an input for first-principle calculations which simulate SP-STM results by taking into account tips structure.[3,8] Furthermore, an epitaxial surface on the tip apex gives rise to a clearly defined electronic structure that contributes to the tunneling effect, while a flatter and smoothed density of states arises from polycrystalline apex.

From this Cr apex atomic arrangement it is naturally tempting to deduce also a spin structure of the atoms contributing to the tunnel current. The (110) Cr surface presents an antiferromagnetic order with alternative atomic rows of opposite spins along the [001] direction.[9] If the tip apex would present such an ideal surface, no clear magnetic contrast would be observed by SP-STM since the same quantities of up and down spins would contribute to the tunneling current. However, this ideal surface does not seem to exist in our case. Looking at the overall structure of the apex shown in Fig. 2, the foremost atoms of the apex are not organized as a flat surface but rather in three-dimensional clusters. These 10 nm clusters probably form the real tip, i.e., the ensemble of atoms on which the spin resolved density of states that contributes to the tunneling process depends. Even if these clusters present a monocrystalline phase, as they appear to in Figs. 1(b) and 2(b), the absolute orientation of this phase as compared to the tip axis cannot be known from one tip to another. Therefore, the foremost spin of the apex which dictates the spin orientation of the tip can probably take any direction independent of the macroscopic crystalline arrangement of the apex. To clean our tip *in situ*, we pulse the tip-sample bias voltage, which frequently can remove material from the tip, such as one of these clusters. The spin structure, and therefore also the spin sensitivity of the tip, can thus be changed by this procedure. An important conclusion is deduced from this result: the spin structure of the apex cannot be controlled by the tip preparation. The determination of the spin sensitivity of the tip should not rely on the tip preparation but rather on *in situ* characterization of tip magnetization by techniques like in field measurement as previously proposed.[2]

These findings concerning control of the tip apex spin structure and magnetic sensitivity should have consequences on SP-STM measurements. In order to confirm this point, we performed STM measurements using a coated Cr tip. As a test sample, we chose the Cr(001) surface whose spin structure is suitable for easy magnetic contrast detection.[10,11] But instead of using bulk Cr monocrystal, whose surface preparation is experimentally very demanding,[12] we rather chose an original and new way to prepare Cr(001) by thin film deposition on a MgO(001) substrate.[13,14] In order to minimize surface contamination originating from the substrate, we cleave a 200 μm thick MgO(001) substrate in UHV, following the semiconductor cleavage technique we used for cross-sectional STM experiments.[15] We then deposit on the cleaved side a 30 nm thick Cr film by thermal evaporation at 300 °C.

An STM structural study of the Cr sample reveals the presence of three-dimensional structures on the surface, such as square base pyramids or spirals.[13] These structures are formed by stacking of small monoatomic terraces which present a statistically predominant [110] orientation, as expected from the epitaxial relation [110](001)bcc-Cr∥[100] ×(001)MgO.[14] STM images are made using a Cr tip at a temperature of 4 K. If dI/dV images do not present any spin polarized contrast, voltage pulses are performed in order to change the atomic scale structure of the tip apex until obtaining the expected magnetic contrast. The success of this procedure confirms the validity of the image described above of
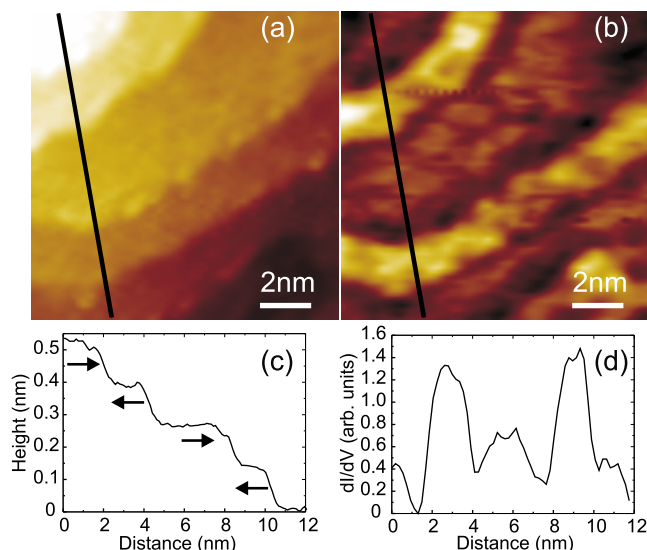
FIG. 3. (Color online) Images of successive monoatomic Cr(001) terraces. Left: topographic image and scan line done on the black line. Right: dI/dV mapping of the same area. Images are performed at 4 K and with stabilization parameters of 1 nA and 0.15 V. Bright and dark alternative contrast is ascribed to spin polarized sensitivity. Arrows show schematically the alternative spin orientation of each terrace.

a cluster-tip that can change under voltages pulses. Figure 3 shows topographic and differential conductance dI/dV maps of the same area presenting a succession of narrow monoatomic terraces. The dI/dV map presents alternative bright and dark contrast from one terrace to the adjacent one, which is expected for spin resolved mapping.[10,11]

In conclusion, we show by HRTEM that Cr deposited on the apex of a flashed W tip is monocrystalline and presents a well defined epitaxial relation with the W apex surface. In spite of this well ordered tip, the apex surface presents nano-

clusters, or *nanotips*, which determine the properties of the tunneling current, such as the spin sensitivity. The foremost spins of the apex that contribute to the tunneling current are therefore most probably always in a canted direction as compare to the tip axis, and should thus result in both in-plane and out-of-plane sensitivity. SP-STM experiments show that such a tip can go through spin sensitive or insensitive states under pulse induced atomic rearrangement.

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