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2006 J. Phys.: Condens. Matter 18 L619

(http://iopscience.iop.org/0953-8984/18/50/L01)

J. Phys.: Condens. Matter **18** (2006) L619–L624 doi:10.1088/0953-8984/18/50/L01

LETTER TO THE EDITOR

Spin polarized tunnelling investigation of nanometre Co clusters by means of a Ni bulk tip

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Received 6 November 2006
Published 27 November 2006
Online at stacks.iop.org/JPhysCM/18/L619

Abstract
A massive Ni tip is used in spin polarized scanning tunnelling microscopy (SP STM) to explore the magnetization state of nanometre Co clusters, self-organized on the Au(111) surface. Constant current STM images taken at 4.6 K show a bimodal distribution of the cluster heights, accounting for the spin polarization of the STM junction. The spin polarization of the tunnel junction as a function of the bias voltage is found to depend on the local density of states of the sample examined. Changing the vacuum barrier parameters by bringing the tip closer to the surface leads to a reduction in the tunnelling magnetoresistance that may be attributed to spin flip effects.

(Some figures in this article are in colour only in the electronic version)

Spin dependent tunnelling in magnetic tunnel junctions has recently been the object of intense study due to its technological impact in a phenomenon called tunnelling magnetoresistance (TMR) [1–3]. The magnetic tunnel junction is also at the origin of spin polarized scanning tunnelling microscopy (SP STM) capable of visualizing the magnetic structure at surfaces [4] preferably in an ultrahigh vacuum (UHV) environment. As a matter of fact, SP STM has proven to be a powerful approach for studying the interplay between structural, electronic and magnetic properties at the nanometre scale. The technique has been used to detect, with a nanometre resolution, magnetic domain walls in ferromagnetic materials and magnetization orientations in small metallic islands, and even to follow the magnetization reversal in small islands on surfaces [5–8].

In this respect, it should be mentioned that vacuum tunnelling is also an ideal tool for testing fundamental TMR issues under the best conditions of surface preparation and tunnelling barrier control; the barrier width can be adjusted within a few 0.001 nm. For example the STM approach may help to disentangle effects in metal–oxide–metal junctions due to the intrinsic nature of electrodes and those due to interfaces such as sign changes in the spin polarization of the Co occurring because of interface bondings [3]. Although the SP STM technique continues to enrich our understanding, it is foreseen that it has not yet reached its full potential. For example, most of the approaches to tip material have been limited to tips with as low a magnetic stray field as possible; for an exception see the early work of [9] where a bulk CrO_2 ferromagnetic tip was used. The work, based on a detailed analysis of SP dI/dV spectroscopy, uses thin Fe, Gd or Cr film covered tungsten tips [4]. This is legitimate if the purpose is mainly to measure properties of a soft magnetic material where the probe should be as uninvasive as possible in order not to perturb the spin state of the sample. Massive ferromagnetic tips however may open new exciting perspectives in the sense that, due to the important stray field, they allow magnetic manipulation of the structures or even spin injection experiments.

In this letter, we demonstrate the feasibility of using massive Ni wire probe tips in SP STM experiments, to measure the magnetic contrasts of single Co nanoclusters, self-organized on the Au(111) surface. Massive ferromagnetic Ni tips are robust and easy to characterize. Their advantage compared to other tip materials is that the tip magnetization is always dictated by the shape anisotropy, since magnetocrystalline anisotropy is weak in Ni. Therefore, the tip will be magnetized always along its axis, leading to a magnetic configuration perpendicular to the surface in the STM experiment. In the past, similar ferromagnetic tips have been characterized by experiments on tunnelling into semiconductor surfaces [10] and by e-holography where flux quantization gives information about the magnetic stray field [11]. Magnetotransport measurements in wire constrictions, on the other hand, provide unique information on nucleation and domain wall propagation [12]. An upper value of the tip stray field can be calculated from the saturation magnetization of 0.494×10^6 A m^{-1} for Ni, which leads to a stray field of 0.62 T. A simple calculation then shows that the maximum energy shift due to the tip stray field $\mu_B B \approx 4 \times 10^{-4}$ eV is negligible.

In order to test the spin polarized contrast provided by the tips, self-organized cobalt clusters ($\sim$3 nm) on Au(111) [13] are a convenient system since their uniaxial anisotropy of the magnetization is perpendicular to the Au substrate [14, 15] with either up or down magnetization. The measurements have been performed with a low temperature STM, in an ultrahigh vacuum chamber with a base pressure of 5×10^{-11} mbar. Due to the working temperature of 4.6 K the magnetization of the Co clusters is strongly locked in the perpendicular direction, pointing either upwards or downwards. As a matter of fact, the blocking temperature of 3 nm Co clusters is measured to be about 70 K in our variable temperature experiments, in good agreement with a simple estimate[1]. The probe tips have been prepared by electrochemical etching of a Ni wire with an appropriate solution [12] and immediately introduced in the vacuum chamber to be resistively heated in order to get rid of the oxide and to be sputtered by means of Ar ions. After this treatment, the tips are magnetized *in situ* along their axis in a field of 0.3 T by means of a permanent magnet.

The constant current STM image of figure 1(a), taken with a magnetic Ni tip, shows clusters of different apparent heights as evidenced in the line scans of figures 1(b) and (c) taken under different tunnelling conditions. The vertical z scale was adjusted to show the topographic contrast of the topmost part of Co clusters. Under the same conditions, the conventional, non-magnetic W tip just reveals a unique cluster height of 4.0 Å corresponding to two atomic Co layers, and the roughness of the top does not exceed 0.1 Å [13]. The presence of two apparent heights in the case of the Ni tip is exactly what is expected, in the constant current mode, for clusters which have parallel (P) and antiparallel (AP) magnetization with respect to the magnetization alignment of the tip. This comes about because it is easier to tunnel SP electrons in an electrode with the same polarization (P) than in an electrode with opposite polarization (AP).

[1] An estimate of the blocking temperature can also be calculated from $T_b = K/k_B \ln(t/\tau_0)$, by taking the magnetic anisotropy energy from the literature. An average value of $K = 0.2$ meV/atom for bilayer Co clusters can be deduced for example from [16]. Assuming a typical measuring time $t = 10$ s, a prefactor τ_0 of the order of 10^{-10} s, and taking into account a number of atoms of about 500 in our clusters, we get $T_b \approx 44$ K.

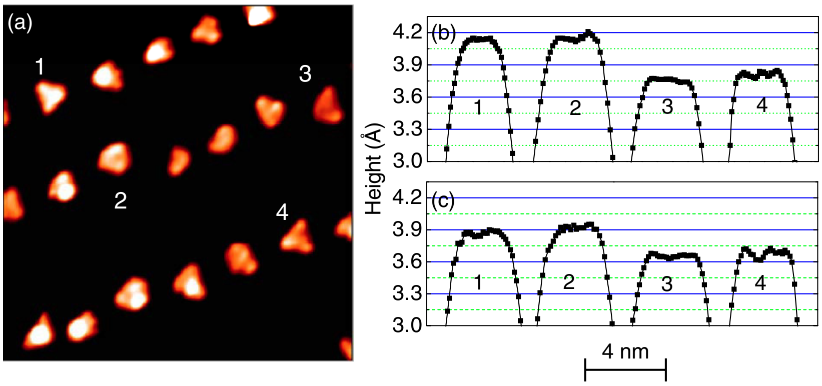

Figure 1. (a) STM image (420 × 420 Å^2) of self-organized Co clusters recorded with the magnetic tip at 4.6 K ($V = -215$ mV, $I = 250$ pA). As an example, (b) and (c) show the cross sections of the clusters numbered in (a) for ($V = -215$ mV, $I = 250$ pA) and ($V = 110$ mV, $I = 600$ pA) respectively.

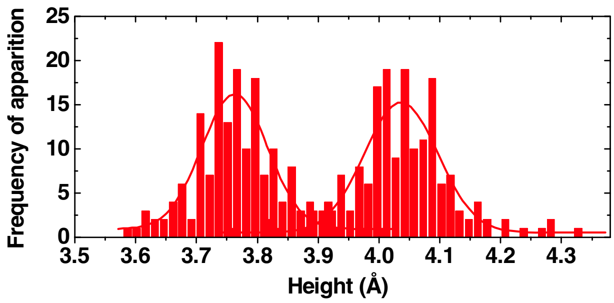

Figure 2. Example of a histogram over many images of the height distribution obtained with the magnetic tip at $I = 450$ pA and $V = 60$ mV, clearly showing two peaks separated by $\Delta z \approx 0.28$ Å. As indicated in the text, the spin polarization, and thus Δz, depends on the tunnelling parameters, current and bias voltage.

Furthermore, for as prepared clusters and in the absence of an external field, we expect as many clusters pointing upwards as downwards. This must be reflected in the statistics of greyscale levels obtained from a great many SP STM images. Figure 2 shows such a statistics where the frequency of occurrence of a given height has been reported for particular values of current and bias voltage. A bimodal distribution is clearly visible, corresponding to equal intensity *parallel* and *antiparallel* states separated, in this case, by $\Delta z = 0.28$ Å. As will be shown below, Δz is directly related to the spin polarization of the tunnelling gap. It must be stressed however that the apparent height strongly depends on the tunnelling parameters, in particular the tunnelling current and the bias voltage.

From SP constant current STM data the polarization of the tunnelling junction can easily be calculated in a simple picture from the distribution of clusters heights. In the topographic mode of the STM, $I_0 = I_P(z_P) = I_A(z_P - \Delta z)$, where I_0 is the feedback current of the STM controller. In order to calculate the polarization of the STM junction, we need to compare *parallel* and *antiparallel* currents for same heights, for example $z_P = z_0$. In other words, we need to calculate $I_A(z_P)$ from $I_0 = I_A(z_P - \Delta z)$:

$$I_A(z_P) = I_0 \exp[-A\Phi^{1/2}\Delta z]. \quad (1)$$

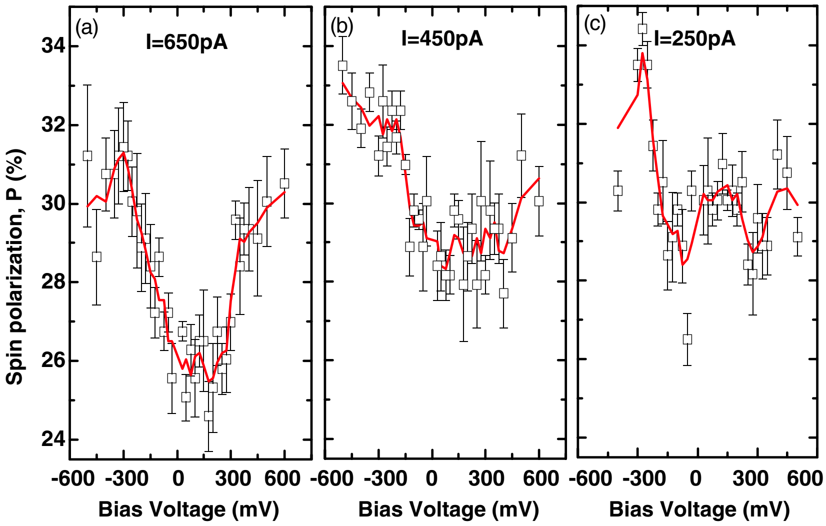

Figure 3. Experimental polarization of the STM tunnel junction, as defined in equation (2), as a function of the bias voltage, for $I = 650$, 450 and 250 pA.

For the calculation we took $\Phi = 4.0$ eV as obtained from the I–z measurement and $A = 1.025\ (\text{eV})^{-1/2}\ \text{Å}^{-1}$. The polarization of the STM junction is then given by[2]

$$P = \frac{I_P - I_A}{I_P + I_A}. \tag{2}$$

In figure 3 the polarization of the STM junction as a function of the bias voltage has been reported from the Δz measurements. As can be seen in figure 3, the polarization as a function of bias voltage is not a monotonic function. Depending on tunnelling current I and bias voltage V (note that on varying V the barrier width varies as well), the polarization of the tunnelling junction is found to vary between 25% and 34%. The many features that appear in the polarization P as a function of V cannot be attributed to an effect of voltage dependent barrier width which normally induces an even variation with respect to the bias voltage. They must rather be assigned to structures in the sample's local density of states (LDOS). A good estimate of the LDOS can be obtained from scanning tunnelling spectroscopy (STS) measurements performed on the same system at 4.6 K [19]. Two peaks dominated by minority d electrons are found respectively above (+0.3 eV) and below (−0.15 eV) the Fermi energy. These spin polarized features lead to a steep increase of the spin polarization in the corresponding energy ranges. Furthermore, when the SP current is changed from 250 to 650 pA, a dip appears in the curves of figure 3 close to zero bias, indicating a strong influence of barrier narrowing on the polarization of the Ni tip–vacuum–Co cluster junction. A similar behaviour has also been observed in spin polarized tunnelling experiments performed on a Co single crystal as a function of tip distance when the tip comes closer to the surface [20].

The polarization P of the junction can be related to the polarization P_i of the electrodes where $i = 1, 2$. Using the phenomenological Jullière model [21], under the assumption of low bias, and utilizing $P = P_1 P_2$, the TMR expressed as $\text{TMR} = 2P/(1 - P)$ can then be calculated. Due to the fact that the spin polarization drastically depends on the LDOS, TMR values can only be compared for well defined bias voltages, for example in the vicinity of the Fermi level. The low bias spin polarization P and the TMR are reported in table 1 for

[2] In STM experiments this formula was used for the first time in [9].

Table 1. Spin polarization P and tunnelling magnetoresistance (TMR), calculated from images taken at three different tunnelling currents I and a bias of 30 mV. See the text for details.

I (pA)	P	TMR
250	0.30 ± 0.01	0.86 ± 0.01
450	0.29 ± 0.02	0.82 ± 0.03
650	0.26 ± 0.01	0.71 ± 0.02

three values of the tunnelling current I. As can be observed, P as well as the TMR decreases for increasing current. It should be recalled here that in the normal STM mode, a current increase brings the tip closer to the surface and therefore reduces the tunnel barrier width. As a matter of fact, spin flip effects, originating from electrode interactions, have been suggested as an explanation for the decrease in TMR at small barrier widths [17, 18]. In a free electron treatment of this problem, that takes into account the barrier parameters, the TMR is given by $\text{TMR} = 2(1-\gamma)P/(1-P+\gamma(1+P))$, where $0 \leqslant \gamma \leqslant 1$ represents the ratio between the square matrix elements of the spin flips and spin-conserved tunnelling. In the case of spin conservation, γ is zero and the above formula reduces to Jullière's one. In this model, the γ factors calculated from the TMR values of table 1 increase as a function of current (decreasing barrier width) but remain small. An upper value of $\gamma = 0.06$ is obtained for a tunnelling current of 650 pA. From $I(z)$ measurements a rough value of the vacuum barrier width can be evaluated; for 250 pA and bias voltages of a few tens of mV a barrier width of 6.5 Å is found. Therefore, by changing I from 250 to 450 and to 650 pA, we calculate a decrease of the barrier width of $\Delta z = 0.38$ and 0.55 Å, respectively. These small Δz variations clearly reveal the high sensitivity of the spin polarization and TMR to small changes in the width of thin vacuum barriers.

In conclusion, we analysed the magnetization state, at low temperature, of self-organized nanometre Co clusters by SP STM using massive Ni wire probe tips. A bimodal cluster height distribution was found, due to the spin polarized tunnelling currents in *parallel* and *antiparallel* configurations, leading to tunnelling magnetoresistance of the junction. Although the SP STM junctions are dominated by features of the LDOS it is found that the geometry of the junction plays a significant role. In particular, by decreasing the vacuum barrier width it was found that the polarization and the tunnelling magnetoresistance at the Fermi level decrease as well. The findings are important for understanding the magnetotransport in tunnel junctions and may have an impact in the development of new spin electronic devices.

The authors acknowledge technical support from J G Faullumel and G Biechel. This work was supported by the Growth Project No G5RD-CT-2001-00478 from the EC.

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