Switching Fields of Individual Co Nanoislands

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We explore the magnetization reversal process of individual Co nanoislands grown on Cu(111) by low temperature spin-polarized scanning tunneling microscopy (spin-STM) and spectroscopy (spin-STS). We measure hysteresis loops of the differential conductance of single Co islands in magnetic fields of up to 4 T. From such hysteresis loops we extract the magnetic switching field of single Co islands as a function of island size. Tentatively we analyze the size dependence of the switching field using the venerable model of thermally assisted coherent magnetization reversal. We present evidence for the failure of that model to explain our experimental results. We propose that the magnetization reversal process within individual Co nanoislands on Cu(111) is a non-coherent process.

***Index Terms*—Magnetic particles, magnetic switching, scanning probe microscopy, spin polarized transport.**

I. Introduction

THE mechanism of magnetization reversal in nanoscale particles is of utmost interest for the magnetic recording industry and for fundamental research [1], [2]. For data storage applications particles with an out-of-plane (or perpendicular) easy magnetization direction have become more and more important in the last years. Previously the magnetization reversal process of *ensembles* of nanoparticles has been investigated by averaging techniques such as magneto-optical Kerr effect (MOKE) measurements [3]. To gain access to the switching field of an *individual* nanoparticle, however, remains a true challenge. In a previous work this challenge has been tackled by micro-SQUID measurements [4], though this study is lacking a diligent characterization of the studied nanoparticles. In this work we present the first experimental study of the switching field of individual *and* well characterized nanoparticles.

We have chosen a prototype system, Co islands on Cu(111), which is known for its out-of-plane easy magnetization direction [5]. Co islands on Cu(111) are mostly of triangular shape with a baselength ranging from 5 to 30 nm. They are exactly two atomic layers high. It has been claimed before that nanoislands of very similar shape reverse their magnetization via coherent rotation [3]. In this paper we will present evidence that this picture is highly questionable for the Co islands studied here.

II. Experimental Details

We use a Cu(111) single crystal, which has been cleaned by cycles of ion bombardment (Ar^+, 2×10^{-7} mbar, 1 keV) and subsequent annealing at 700 K. We prepare Co islands by depositing a submonolayer quantity of Co onto the clean Cu(111) surface at 310 K using a calibrated e-beam evaporator. STM measurements at 8 K reveal the growth of Co islands which are two atomic layers high, i.e., 0.4 nm, and of almost triangular shape. Fig. 1 shows a constant current STM image with several Co islands close to a step edge of the Cu(111) surface. In addition, Co also decorates the step edges.

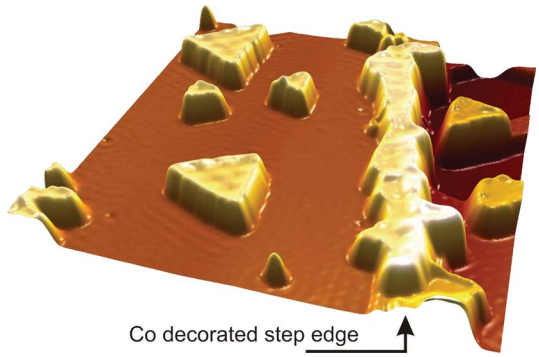

Fig. 1. Three dimensional representation of a constant current STM image of Co on Cu(111) taken at 8.3 K. Upon deposition Co decorates the step edges of the Cu(111) surface (see right part of the image). Triangular islands of Co which are two atomic layers high (i.e., 0.4 nm) are formed as well. Imaging conditions: $I_T = 1$ nA, $V_{\mathrm{Gap}} = +0.1$ V. Image size: 45 nm × 34 nm.

For spin-STM measurements we use electrochemically etched W tips, which we flash in-situ to 2500 K. To yield magnetic sensitivity we cover the tips with 20–100 atomic layers of Cr. Details of the tip preparation can be found in our earlier work [6]. To investigate the electronic and magnetic properties of individual Co islands we perform spectroscopic measurements of the differential conductance dI/dV($V_{\mathrm{Mod}} = 10 - 20$ mV, $f_{\mathrm{Mod}} = 4 - 5$ kHz). We measure dI/dV spectra on individual Co islands in magnetic fields of up to 4 T. We use the external field, which is oriented perpendicular to the surface, to manipulate the magnetization state of the islands. Changes in the magnetization state of a Co island lead to corresponding changes in the dI/dV spectra measured on the island. We can reliably distinguish these changes of the dI/dV signal from contributions due to changes in the magnetization configuration of the tip [6], [7]. From a series of spectra recorded during a sequence of field sweeps we extract dI/dV hysteresis loops of individual Co islands [7].

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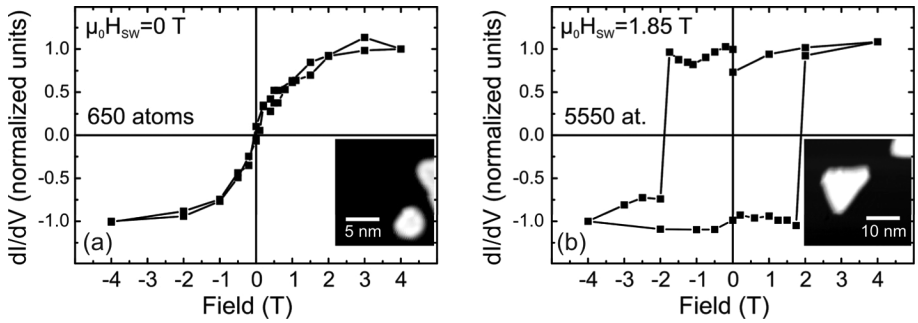

Fig. 2. Hysteresis loops of the dI/dV signal of two individual Co islands on Cu(111) recorded at 8.3 K. (b) Abrupt changes of the dI/dV signal mark the switching of the island magnetization. Resulting switching fields $\mu_0 H_{SW}$ are indicated. Insets show constant current STM images of the small (a) and large island (b) ($V_{\mathrm{Gap}} = -0.1$ V, $I_T = 1$ nA).

III. Size Dependence of the Switching Field

Our ability to extract hysteresis loops of individual Co islands allows us to quantify the switching field ($\mu_0 H_{\mathrm{SW}}$) of individual islands and to analyze its size dependence. Fig. 2 shows hysteresis loops measured on two different Co islands at 8.3 K. The dataset presented in Fig. 2(a) for an island with only 650 atoms does not show a hysteretic behavior but rather resembles an S-shape. This is ascribed to a superparamagnetic response of the island. The dataset in (b) shows a clear hysteresis of the dI/dV signal. The sudden changes of the dI/dV signal are ascribed to the switching of the out-of-plane component of the magnetization of the island. From those data we extract a switching field of 1.85 T for an island containing 5600 atoms. The hysteresis loops shown in Fig. 2 are measured with a tip having a rigid magnetization orientation with respect to the external field [6].

We have explored a wide range of island sizes ranging from 500 atoms ($\sim$5 nm^3) to 7000 atoms ($\sim$70 nm^3). Fig. 3 shows a plot of the switching field as a function of island size. Note, that all our data are extracted from dI/dV loops measured on individual Co islands. The plot of Fig. 3 is divided into two regimes. Islands in regime I which are smaller than 1000 atoms ($\sim$10 nm^3) do not show hysteresis. Islands in regime II, which are larger than 1000 atoms, exhibit a monotone increase of the switching field with size.

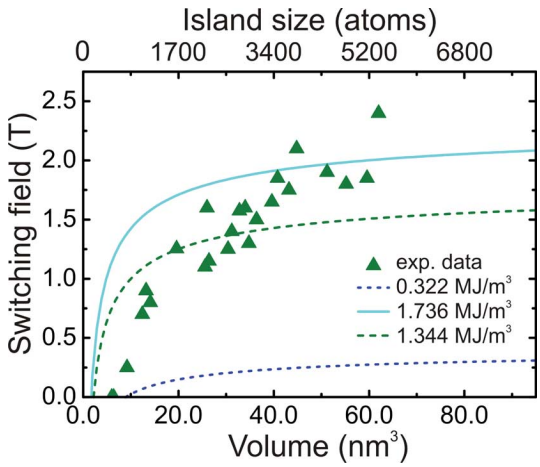

Fig. 3. Size dependence of the switching field of single Co islands on Cu(111). The data are divided into two regimes: I) Anhysteretic behavior, i.e., $\mu_0 H_{\mathrm{SW}} = 0$ T. II) Continuous increase of the switching field. Curves are calculated using the model of thermally assisted switching using different anisotropy energies. However, none of the curves gives a good description of the experimental data.

IV. Discussion

We discuss our experimental data in view of a thermally assisted magnetization reversal process. We ascribe the missing hysteresis for islands in regime I to a superparamagnetic response of those islands. In contrast, the islands in regime II show a stable magnetization, they are in the so-called blocked state.

The simplest description of the switching of a small magnetic particle is the Néel-Brown model [8], [9] applied to a Stoner-Wohlfarth (S-W) reversal mechanism [10]. Within this theory the magnetization reversal is described as a thermally assisted coherent rotation of all spins in unison. This description is also known as the so-called macrospin model. This model has previously been successfully applied to nanoscale particles [4], and we apply it tentatively here; although the two layer flat Co islands of this study are not of ellipsoidal shape as requested by the S-W model. Within the framework of the Néel-Brown model the energy barrier ΔE that has to be overcome for reversing the magnetization of a single domain particle with uniaxial anisotropy is given by

$$\Delta E = KV\left(1 - \frac{\mu_0 H M_S}{2K}\right)^2 \tag{1}$$

where K is the anisotropy per unit volume, V is the volume of the particle, $\mu_0 H$ is the external magnetic field, and M_S is the saturation magnetization of the particle. This switching barrier is competing with the thermal energy of the system. For the typical time scale of our spin-STM measurements the criterion for a stable magnetization of 100 s duration is [11]:

$$\Delta E = 25 k_B T. \tag{2}$$

Here, k_B is the Boltzmann constant and T is the temperature. With zero magnetic field and at constant temperature this stability criterion determines the transition point between the superparamagnetic state and the stable magnetization state.

Particles with $V < (25 k_B T)/K$ show a superparamagnetic response, and particles with $V > (25 k_B T)/K$ show a stable

magnetization for at least 100 s. The critical volume where the transition from the superparamagnetic to the blocked state occurs is called blocking volume V_B. We determine experimentally the blocking volume from the transition point between regime I and regime II as shown in Fig. 3. For Co islands on Cu(111) we find $V_B = 8.89$ nm^3 at 8.3 K.

Within the coherent rotation model we get a first estimate of the anisotropy constant from:

$$K = \frac{25 k_B T}{V_B}. \tag{3}$$

With $T = 8.3$ K, the anisotropy is estimated to $K = 0.322$ MJ/m^3. In view of the large values reported for nanoscopic systems, e.g., 7 MJ/m^3 [12], this value is surprisingly small. This is a first indication that the model of thermally assisted coherent magnetization reversal might not be applicable. Doubts about the applicability of this model are also corroborated by considering the maximum switching field of 2.4 T that is observed in regime II. The largest possible field for reversing the magnetization direction of a single domain particle is predicted by the Stoner-Wohlfarth model. It gives a maximum switching field of $\mu_0 H_{\max} = 2\,K/M_S$. Numerous mechanism are known that can lead to smaller switching fields. However, not a single mechanism is known that would lead to larger switching fields. Hence, the Stoner-Wohlfarth model gives an upper limit for the switching field, based on the anisotropy constant and the magnetization. The magnetization of Co islands on Cu(111) has been estimated by X-Ray circular dichroism (XMCD) measurements to at least 1.6 μ_{Bohr}/atom [13]. With the estimated anisotropy constant of $K = 0.322$ MJ/m^3 this yields a switching field of $\mu_0 H_{\max} = 0.5$ T. This value is dramatically smaller than the 2.4 T observed in our experiment.

To underline the conflict raised by these first estimates for anisotropy and switching field, we perform a quantitative analysis of our data. Using the stability criterion (2) with the energy barrier (1) yields the size dependence of the switching field of a single particle, the so-called *Sharrock* equation [14]:

$$\mu_0 H_{\mathrm{SW}} = \frac{2K}{M_S}\left(1 - 5\sqrt{\frac{k_B T}{KV}}\right). \tag{4}$$

Using (4), we calculate the size dependences presented in Fig. 3. The only free parameter in (4) is the anisotropy K. We first calculate the size dependence with the anisotropy constant $K = 0.322$ MJ/m^3, that we extracted from the blocking volume V_B. The result is shown as a pointed (dark blue) curve in Fig. 3. Second, we use the anisotropy which we extract from the maximum switching field of 2.4 T and the Stoner-Wohlfarth limit: $K = (\mu_0 H_{\max} M_S)/2 = 1.736$ MJ/m^3. The result is shown as a solid (light blue) curve. Both curves clearly fail to explain the experimental findings. In a third attempt to explain our data with this model we do a least square fit of the data (broken (green) curve). The fit yields an intermediate value for the anisotropy of 1.344 MJ/m^3. However, the quality of the fit is very poor, it fails to describe the transition from the superparamagnetic to the blocked state, and we conclude that it is not applicable here.

In previous works it has been proposed to introduce an additional parameter to the coherent rotation model in order to explain experimental data [3], [4]. This parameter reduces the particle volume which is taken into account for the analysis. The reduced volume is often called "activation volume", and its consideration indeed leads to a better agreement between experiment and the coherent rotation model. However, it doesn't come as a surprise that the introduction of an additional parameter to the model leads to improved fitting results. This is a mathematical triviality. The true challenge is the interpretation of the resulting "activation volume." We think that its physical meaning should be critically questioned and that this rather formal solution most probably disguises the physics of the reversal process.

In summary, the experimentally observed size dependence of the switching field of individual Co islands on Cu(111) cannot be reliably described using the model of thermally assisted coherent magnetization rotation. From this failure we conclude, that the magnetization reversal process within the Co islands is more complicated, and that it might involve noncoherent processes.

This conclusion calls for a more sophisticated analysis of our experimental data. However, a state-of-the art theoretical description of the reversal process presents a true challenge. Decisive physical quantities, which have been treated as constants previously, are expected to show distinct spatial variations. This expectation is based on our spatially resolved spin-STM study of the electronic properties of an individual Co island. Our earlier work revealed a spatial variation of both the differential conductance and the spin-polarization within one island [15], [16]. In addition,we have convincing indications that also the anisotropy cannot be described by one constant value. Preliminary calculations by Etz show that for a Co island with 16 atoms the anisotropy can vary substantially from atom to atom [17].

A local variation of the anisotropy will lead to different energy barriers for switching the local magnetic moments within one island. In that case a coherent rotation of all spins in unison would become less favorable. In contrast, local minima of the anisotropy could favor nucleation of so-called droplets. Droplets are, like domains, areas which are oppositely magnetized with respect to the rest of the particle. Unlike domains, droplets are non-equilibrium entities which either grow or shrink but never reach a stable size [18].

We think that droplet nucleation is a plausible magnetization reversal mechanism since it is favorable for systems with high anisotropy [19]. The reason is that a high anisotropy leads to very thin domain walls between the droplets of down to one lattice constant [19]. This theoretical estimate is corroborated by an experimental study of Fe stripes on a W(110) surface [20], which reports domain walls in the Fe stripes of only 0.6 nm width. However, the model of droplet nucleation is describing the magnetization reversal process of single domain particles. Hence, the "domain" walls between the droplets, i.e., droplet walls, are considerably thinner than a "classical" domain wall between two equilibrium domains. We estimate the domain wall width for our Co islands using:

$$\delta_0 = \sqrt{A/K}. \tag{5}$$

Here, A is the exchange stiffness [21]. From the maximum switching field of 2.4 T we yield a lower limit for the anisotropy for the Co islands of $K > 1.7$ MJ/m^3. This large anisotropy, in combination with a typical value for $A = 10^{-11}$ J/m, leads to $\delta_0 < 2.5$ nm. This value is smaller than the base lengths of the islands in region II of Fig. 3 of 9–20 nm. Hence, such a domain wall could exist within these islands, making droplet nucleation a plausible reversal mechanism. Experimentally we never observed a domain wall. However, due to the nonequilibrium character of the droplets this does not exclude a reversal via droplet nucleation.

Furthermore dynamic phenomena are very likely to play a crucial role for the magnetization reversal in nanoparticles. A recent study [22] reports that the reversal rate of Co nanodots on Au(111) is strongly influenced by the thermal excitation of spin-waves within the Co nanodots, and that this leads to distinct deviations from the macrospin model. Although such dynamical processes are not accessible by the current experiment, we believe that they are an additional justification for questioning the reversal via coherent rotation.

Recently the magnetization reversal process of individual superparamagnetic Fe islands on W(110) has been studied by spin-STM [12]. This investigation did not elaborate the magnitude of the switching field of thermally stable islands, but rather focused on the statistical analysis of the switching probability of the islands. The authors report a very large anisotropy of $\sim$7 MJ/m^3 for this system and find that the reversal process of islands with 30–150 atoms, i.e., with diameters of 2–6 nm, is best described by a nucleation model. Hence, this previous work supports our picture that nanoscopic particles with a large anisotropy can exhibit extremely narrow domain walls, and that the magnetization reversal process can be described by a nucleation process. The authors [12] are focusing on the size dependence of the *switching probability* of superparamagnetic particles. Their work does not provide any insight into the size dependence of the *switching field* of thermally stable particles which are investigated here.

V. Conclusion

Our analysis of the size dependence of the switching field of individual nanoparticles strongly suggests that the model of thermally assisted coherent magnetization reversal cannot be applied in the case of Co islands on Cu(111). We conclude that the magnetization reversal process might include non-coherent processes like the nucleation of droplets and the excitation of spin-waves. The large anisotropy which we estimate for the Co islands supports this view, since it makes the formation of the extremely narrow domain walls, needed for the nucleation process, plausible.

Our findings indicate that a state-of-the art theoretical description of the reversal process would have to include spatial variations of the anisotropy and most likely also of the magnetic moments within the islands. At present such a treatment remains a true challenge for theory. However, we are confident that our experiments will trigger some new developments in this field.

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