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High sensitivity magnetization measurements of nanoscale cobalt clusters

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Presented is a novel high sensitivity magnetometer allowing us to measure the magnetization reversal of about $10^4 \mu_B$ corresponding to a sensitivity of about 10^{-16} emu. The detector is a niobium micro-bridge DC superconducting quantum interference device (SQUID), fabricated using electron-beam lithography. It is operational in the temperature range of 0–7 K. Furthermore, we present a method to deposit on the SQUID loop a small number of Co clusters of about 2–5 nm in diameter. The first results obtained on these samples show that there is still a ferromagnetic coupling between the clusters and the magnetization reversal takes place by small avalanches. © 1995 American Institute of Physics.

I. INTRODUCTION

Despite considerable work, the understanding of the process of magnetization reversal in a single-domain particle constitutes an important and still challenging goal in magnetism. This goal is important because the knowledge of this process is of fundamental interest, in particular for the problem of magnetization reversal in complex systems (assemblies of particles, bulk materials, etc.). It also has essential implications in recording media applications. The rapidly increasing density of magnetic storage poses the question of how far the reduction of volume to surface ratio can continue without inducing new sources of information errors (as those resulting from particles surface roughness or, at low temperature, from quantum tunneling of the magnetization) besides those due to thermal activation only.^{1–5}

In this letter we report on a novel high-sensitivity magnetometer allowing the first studies of magnetization reversal of a small number of crystalline single-domain Co clusters of 2–5 nm in diameter. These studies constitute the first step towards magnetization measurements of an individual nanoscale particle.

II. MEASUREMENT TECHNIQUE

As magnetometer we are using planar micro-bridge DC superconducting quantum interference devices (SQUID),⁶ fabricated by electron-beam lithography, using a JEOL 5DIII *e*-beam writer. The SQUID consists of a square loop, about 1–2 μm wide and interrupted by two micro-bridge Josephson junctions. The dimensions of the SQUID arms are between 0.1 and 0.4 μm [Fig. 1(a)]. The SQUID loops are made of Al or Nb, operational at temperatures as high as 0.8 or 7 K, respectively. Our method consists in measuring the critical current of the SQUID. As the critical current is a

periodic function of the flux going through the SQUID loop, one can easily deduce the flux change in the SQUID loop. The resolution of our SQUIDs is about $10^{-4} \Phi_0$ ($\Phi_0 = h/2e = 2 \times 10^{-15}$ Wb). On these SQUIDs, we deposit a small number of ferromagnetic clusters by low-energy cluster beam deposition⁷ [Figs. 1(a) and 1(b)]. Due to the close proximity between sample and SQUID we have a very efficient and direct flux coupling. When the external field is applied in the plane of the SQUID, the SQUID is only sensitive to the flux induced by the stray field of the sample's magnetization. In the case of Nb-SQUIDs, we can apply in-plane magnetic fields as high as 1 T. At these fields we observe a smooth change of the absolute value of the critical current, which we take into account. The field coils are wound with a highly multifilamentary (14496 filaments of 0.55 μm each) superconducting wire, minimizing remanence and flux creep. Today, in this configuration, we can detect magnetization reversals corresponding to $10^4 \mu_B$.^{8,9}

For the $M(H)$ measurements we record the measured flux change in the SQUID loop as a function of the in-plane applied magnetic field. The parallelism between the magnetic field and the SQUID plane is adjusted by two independent methods: (i) saturating the sample in high fields (1 T) and (ii) by subtracting the magnetization signal detected by an empty SQUID next to the SQUIDs with sample (at a distance of some μm). In the figures we indicate the flux change measured as well as the change of the magnetic moment. Flux is converted in magnetic moments using a SQUID sensitivity of about $4 \times 10^7 \mu_B / \Phi_0$. This value is obtained from measurements of Co, Ni, or Fe particles patterned by electron-beam lithography out of sputtered thin films and having in-plane dimensions of 50 nm–1 μm and thicknesses of 5–50 nm.^{8,9} At the moment we cannot be sure to what extent this value is exact in the case of nanoscale clusters. It should be considered as a rough order of magnitude.

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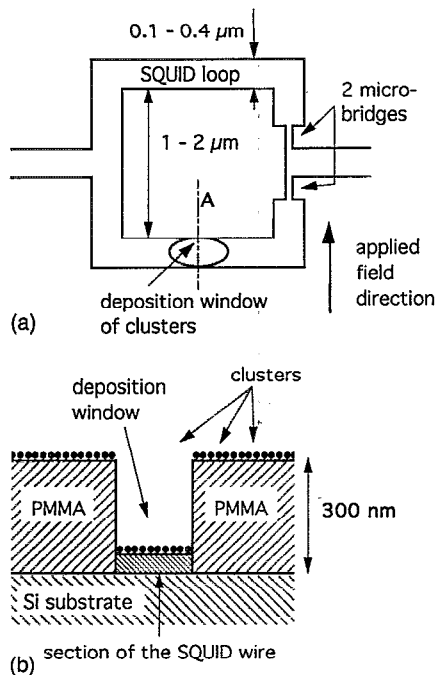


FIG. 1. (a) Schematic drawing of our micro-bridge dc-SQUID showing the SQUID loop with the two micro-bridge junctions. The deposition of the clusters is made on the SQUID loop (best flux coupling) using a PMMA masque. The direction of the in-plane applied magnetic field is indicated. (b) Cross section of the deposition window following the line A in (a).

III. PREPARATION AND DEPOSITION OF CLUSTERS

The clusters are elaborated using low-energy cluster beam deposition techniques.⁷ We use laser vaporization (Nd-YAG laser light) to create a plasma in a vacuum cavity. A laser shot ionizes the target material and a synchronized high-pressure helium pulse thermalizes this plasma, provoking cluster growth. These clusters are then rapidly quenched during isentropic expansion in vacuum, and they are deposited on substrates at room temperature. Limited diffusion and recrystallization will then occur, leading to typical supported particle sizes between 2 and 6 nm for Ni, Co, or Fe clusters, independent from the deposited thickness.¹⁰ As an example, Fig. 2 shows a crystalline Ni cluster of about 3 nm. Electrical percolation begins for equivalent film thicknesses thicker than about 2 nm of supported particles. These films are nanocrystallized (random stacking of clusters).¹⁰

Our samples are patterned by electron-beam lithography on a silicon substrate. To test the reproducibility of our measurements, each chip consists of about 100 micro-SQUIDs. The chip is covered with 300 nm poly(methylmethacrylate) (PMMA) resist with windows in it of variable shapes. The windows are placed on the arms of the SQUIDs, using electron-beam lithography, as shown in Fig. 1(b). The clusters are deposited directly on the whole chip. For cluster depositions thinner than 1 nm of equivalent film thickness, the SQUIDs are only sensitive to clusters which fall into the windows. Clusters sitting directly on the wire are best coupled to the SQUID loop. The clusters on top of the PMMA mask are too far away from the SQUID to give a detectable signal. For cluster depositions thicker than 1 nm,

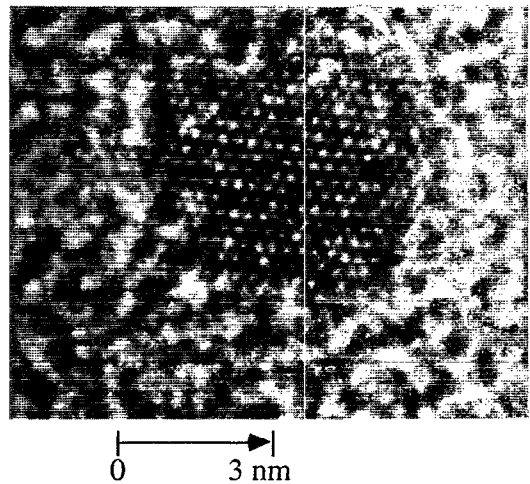


FIG. 2. Scanning transmission electron micrograph of a crystalline Ni cluster of about 3 nm.

we dissolve the PMMA in a solvent after the cluster deposition, removing from the chip all clusters stuck on the top of the PMMA which give a background signal.

IV. RESULTS

In the following, we present four hysteresis loops showing the ferromagnetic coupling between the clusters which depends strongly on the deposition thickness.

A. Below the electrical percolation thickness

Figures 3(a) and 3(b) show typical hysteresis loops of a small number of clusters which are deposited into two different deposition windows, as seen in the insets, but with the same equivalent film thickness of about 0.2 nm. This thickness is much smaller than the percolation film thickness (2 nm). Nevertheless, there is still a ferromagnetic coupling between clusters as the observed magnetization jumps are larger than those expected for individual clusters of 2–5 nm. The overall amplitude of the magnetization change is ten times bigger for the sample with the square window. This is in agreement with the geometry as its surface is ten times the surface of the elliptic window. The insets of Figs. 3(a) and 3(b) show the largest magnetization jumps corresponding to spin flips of about $5 \cdot 10^5 \mu_B$ in Fig. 3(a) and $5 \cdot 10^4 \mu_B$ in Fig. 3(b). As one round Co cluster of 5 nm has about $10^4 \mu_B$, there are at least 50 clusters which flip together in Fig. 3(a) and five clusters in Fig. 3(b). We can mention that the shape of these curves is typical for a material with a huge random anisotropy as encountered in rare-earth based particles.⁸ But in our case, the origin of the random anisotropy comes from the random stacking of our nanocrystallized particles on the substrate.¹¹ Near the percolation threshold, the ferromagnetic correlation length is between 1 and 10 nm. The switching field is independent of the window area. The hysteresis loops show very small and not reproducible magnetization jumps corresponding to the relaxation of a few nanometric particles. In summary, for thicknesses below the electrical percolation thickness, the particles have no macroscopic shape dependence.

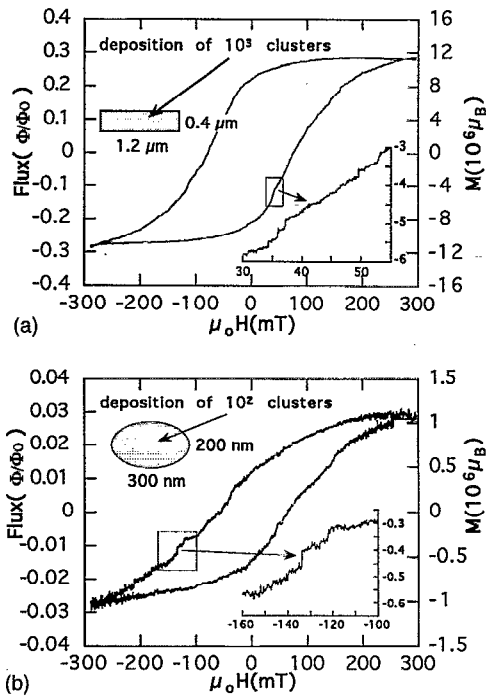


FIG. 3. Hysteresis loops of about (a) 10^3 and (b) 10^2 Co clusters which are deposited on the SQUID wire. The equivalent film thicknesses of the deposited clusters is about 0.2 nm. The measured flux change is given in units of Φ_0 and approximately in units of μ_B . The insets, presenting a zoom of the hysteresis loops, reveal small magnetization jumps coming from several clusters which are strongly coupled (one round Co cluster of 5 nm has about $10^4 \mu_B$). $T=0.2$ K.

B. Above the electrical percolation thickness

Figures 4(a) and 4(b) show the hysteresis loop of an equivalent film thicknesses of 5 and 15 nm, thicker than the percolation thickness (2 nm). In this case, the clusters form a nanocrystallized film, i.e., they are strongly coupled by dipole and exchange interactions and the hysteresis loop becomes dependent on the shape of the window. These hysteresis loops are very similar to those obtained on submicronic Co particles patterned by electron-beam lithography out of sputtered films.^{8,9} First, the switching field decreases with the thickness increases (from 0.2 to 5 nm equivalent thicknesses) and the hysteresis loop tends to a rectangular shape as in single-domain Co particles. Then, for thicker samples (15 nm equivalent thickness), the hysteresis loop is similar to hysteresis loops of multi-domain particles. However, in the case of nanocrystallized films, the switching field distributions of the magnetization jumps are about ten times broader [see inset of Fig. 4(b)]. This comes from the fact that nanocrystallized films are porous and slightly oxidized at cluster boundaries. This leads to a distribution of reduced cluster-to-cluster exchange couplings (with respect to a bulk film¹¹), which increases the switching field distribution. As in this case, the cluster-to-cluster coupling is much stronger than in the previous one (below the threshold percolation thickness), so one expects much larger avalanches. This is effectively observed as can be seen in Figs. 4(a) and 4(b). The magnetization reverses mainly in two or three big avalanches, whereas in

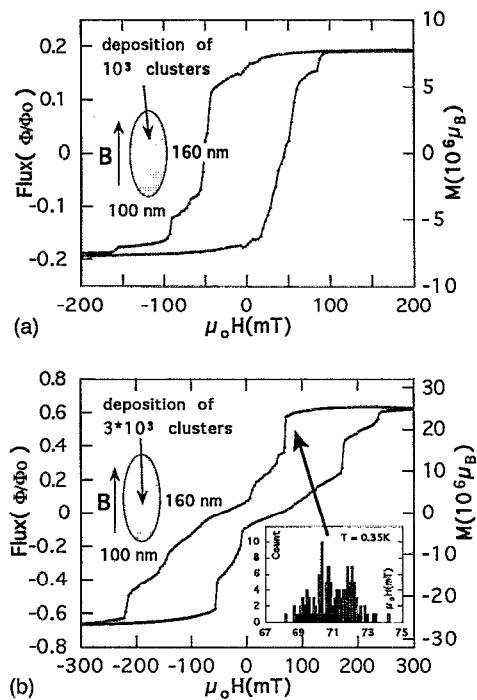


FIG. 4. Hysteresis loops of two elliptic Co particles composed of thick layers of Co clusters forming nanocrystallized films with equivalent film thicknesses of (a) 5 and (b) 15 nm. The measured flux change is given in units of Φ_0 and approximately in units of μ_B . $T=0.2$ K. The insets on the left present the shape of the deposition window. The inset on the right of (b) shows a switching field histogram for the first avalanche drawn from 130 hysteresis loops which were measured at a constant field sweeping rate of 20 mT/s.

smaller equivalent film thicknesses [see Figs. 3(a) and 3(b)] there are a large number of small avalanches.

V. CONCLUSION

We present a new method of magnetization measurements allowing us to measure the dynamical behavior of single-domain particles with a resolution of $10^4 \mu_B$ (10^{-16} emu). In the future, we want to make *in situ* measurements to minimize cluster oxidation and to follow the magnetic percolation threshold *in situ* during the deposition. Improvements of our SQUID technique (shunted dc-SQUID) will increase the resolution by two orders of magnitude. Then, individual cluster studies will be possible

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