Enhanced Coercivity in Submicrometer-Sized Ultrathin Epitaxial Dots with In-Plane Magnetization

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We present measurements, analysis, and modeling of magnetization reversal processes in ultrathin magnetic dots with in-plane magnetization. This study is performed on a model system: Arrays of tens of millions of epitaxial (110) Fe dots. We clearly demonstrate the link existing between the increase of dot thickness and the decrease of reversal field as compared to coherent rotation (CR) predictions. For 1-nm-thick films $H_r(\theta)$ is very close to CR law, although magnetic relaxation experiments clearly show that nucleation volumes are by far smaller than an individual dot volume. This apparent discrepancy is discussed. An analytical model is proposed to describe magnetization reversal in this kind of dots. [S0031-9007(98)08338-0]

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The present trend in microelectronics and magnetic storage industries is a dramatic decrease of component size in order to reach ever higher devices integration. This integration is not merely limited by fabrication technology: the magnetic behavior of systems may change as their size is scaled down to some magnetic characteristic length scale. The superparamagnetic limit of individual grains in continuous storage media [1], demagnetizing effects in magnetic dots that may be part of spin valve components [2] and topological bits prospective recording media [1,3–5] are among the recent issues under investigation. In any case qualitative changes of magnetic properties are expected to occur [6,7] when the dimensions of dots (thickness, lateral size) are reduced down to some magnetic characteristic length scale. The present study focuses on ultrathin dots with in-plane magnetization. The motivation is the following: For in-plane magnetization magnetic poles are located on the edges of the dot, so that reducing the dot thickness lowers the magnitude of internal demagnetizing fields. In the mathematical limit of zero thickness internal dipolar fields vanish, the magnetization is uniform in the dot, and the magnetization reversal is described by the coherent rotation (CR) model [8,9], which predicts that the reversal field H_r along the easy axis of magnetization equals the anisotropy field H_a. In dots of finite thickness internal dipolar fields have to be taken into account, which generally results in H_r being decreased as compared to H_a. The relevant parameter to measure how weak internal fields are is therefore $h_r = H_r/H_a$. The closer h_r is to 1, the weaker is the influence of internal fields. In order to get rid of extrinsic sources of reversal like microstructure, roughness, etc., dots were made out of an epitaxial pulsed-laser-deposited (110)Fe layer sandwiched between two Mo or W (110) layers [10]. The dots were fabricated using successively x-ray lithography, Al mask thermal evaporation, reactive ion etching of Mo/W, and ion beam etching of Fe [11]. After etching, the dots were encapsulated *in situ* with Cu in order to prevent any oxidation during *ex situ* characterizations. Arrays of 10^8 identical particles were fabricated so that the total magnetization magnitude, approximately 10^{-8} A m^2, would allow one to perform macroscopic SQUID or vibrating sample magnetometer (VSM) measurements. Three arrays with thickness 6, 2, and 1 nm and sandwich material W (6 nm thick) or Mo (1 and 2 nm thick) were prepared, either as squares with a 500-nm edge (6 nm, with edges, respectively, parallel and perpendicular to the easy and hard axes directions) or disks with a 200-nm diameter (1 and 2 nm). As we want to point out the role of thickness, it is certainly not the best practice to vary the shape and size of dots as well. This was not a choice, but has come from historical reasons: we first studied "thick" dots and used a mask with 500-nm square-shaped dots [11]. When we turned towards studying thinner dots this mask had broken and we could use a mask with 200-nm disk-shaped dots only. The difference in lateral size is, however, not expected to play a dominant role as this size is in all cases by far larger than magnetic characteristic length scales, the exchange length $\lambda_{\text{ex.}} = \pi\sqrt{2A/\mu_0 M_s^2} \simeq 10$ nm and the Bloch wall width $\lambda_{\text{Bl.}} = \pi\sqrt{A/K} \simeq 20$–$100$ nm [12,13]. Interdots' spacing was always equal to the dots lateral size. The calculated interdot dipolar coupling was found to be negligible in this geometry [11] so that measurements over the whole array are characteristic of independent dots, but for the distribution of properties within the array. All magnetic measurements reported here were performed in plane. The anisotropy in the films was determined with an accuracy better than 10% by performing hard-axis loops. The anisotropy is fourth order in 6-nm-thick films [14], and mainly second order in 1- and 2-nm-thick films [15] due to dominant interface

contributions. No anisotropy difference between dots and films could be found for 1- and 2-nm-thick samples, whereas a slight shift was found and analyzed in the case of 6-nm-thick samples [11].

Easy-axis hysteresis loops of continuous films and dots for thickness 6, 2, and 1 nm are shown in Fig. 1a. The loops were performed at low temperature (10 K) so that thermal activation plays no role. In this figure the extremum values on the x axis were set to the anisotropy field H_a (respectively, 55, 300, and 550 mT) to allow a straightforward comparison of h_r values (the magnitude of H_a increases as thickness decreases due to the interplay of interface vs volume sources of anisotropy [14]). First note that h_r increases from continuous films to dots for a given thickness. This increase is a direct consequence of patterning: continuous films reverse by free propagation of a unique domain wall after nucleation on a major defect somewhere on the film, so that h_r do not reflect the film intrinsic properties (the increase of h_r in continuous films below 1 nm results from domain walls pinning due to the dramatic relative importance of atomic statistical roughness on films of less than 5 atomic layers [16]). In a dot the probability to find a major defect is strongly reduced so that h_r is now intrinsic to the film properties and dots geometry. The demagnetizing influence is then evidenced as h_r ranges from 0.25 for 6-nm thickness to 0.73 for 1-nm thickness. The physical meaning of this increase can be better understood by plotting in-plane energy profiles of a spin at the reversal field $h = h_r$, taking into account the anisotropy energy and the Zeeman energy (Fig. 1b). One immediately sees that by patterning systems were stabilized in a highly metastable state, both in terms of local well shallowness and small angular extension. As thermal activation is negligible the depth of the well just before the reversal can be seen as an approximate measure of the dipolar energy in a nucleation volume. The weak influence of this energy was also revealed by in-plane angular $h_r(\theta)$ measurements which prove to be quite close to the Stoner-Wohlfarth (SW) CR law (see Fig. 2 and Ref. [15]).

In the following we focus on 1-nm-thick dots whose behavior looks the closest to CR. It is generally thought that the CR law may be observed only in the case of systems smaller than $\lambda_{\text{ex.}}$ and $\lambda_{\text{Bl.}}$, in which nonuniform configuration are obviously energetically unfavorable even in the case of strong internal dipolar fields. In the present case the reversal nearly *looks like* CR, although the dot's lateral size (200 nm) is still well above $\lambda_{\text{ex.}}$ ($\simeq$ 10 nm) and $\lambda_{\text{Bl.}}$ ($\simeq$ 20 nm). In order to address this question we determined the activation volume in dots. The determination was done independently by two thermal activation analyses between 2 and 300 K: the first one is that of the thermal dependence of H_r and the other one that of the temperature

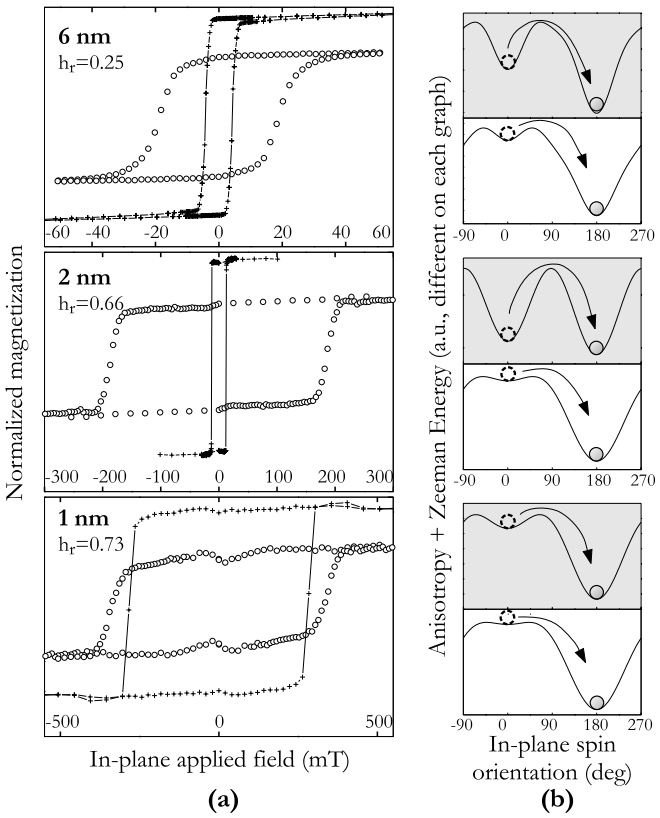

FIG. 1. (a) In-plane hysteresis loops of continuous films (+) and dots (○) as a function of dot thickness. Two different normalizations have been used for clarity. (b) Anisotropy plus Zeeman energy of a spin, as a function of the spin in-plane angle. Curves were calculated at reversal fields of films (grey background) and at that of dots (white background). The initial state is $\varphi = 0°$ and the external field is applied along $\varphi = 180°$.

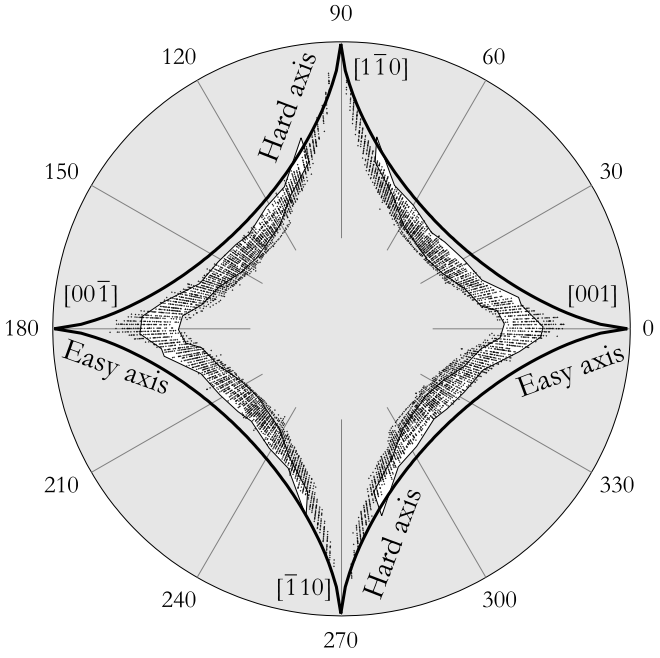

FIG. 2. Polar plot of in-plane reversal field $H_r(\theta)$ in 1-nm-thick dots measured microscopically using a micro-SQUID (each dot on the plot stands for the reversal of a single dot in the array [15]) and macroscopically using a VSM (nonshaded area, behind dots). The CR law is plotted as a black line.

and field dependence of magnetic viscosity [17]. Both analyses are based on similar Brown relaxation equations: the energy barrier ΔE to be overcome to reverse the magnetization is

$$\Delta E = \frac{1}{2}\,\mu_0 M_s H_r^0 V_n (1 - H/H_r^0)^\alpha, \qquad (1)$$

where H_r^0 is the reversal field extrapolated at $T = 0$ K, α is an adjustable exponent around 2, V_n is the activation volume, and M_s is the saturation volume magnetization. Because of the fact that not all dots are identical, one must also take into account a distribution function of reversal fields $\rho(H_r^0)$ at $T = 0$ K. The mean reversal time for a given H_r^0 is

$$\tau = \tau_0 \exp\left[\frac{\mu_0 M_s H_r^0 V_n}{2kT}(1 - H/H_r^0)^\alpha\right], \qquad (2)$$

where $\tau_0 \simeq 10^{-9}$ s. Using the conventional "step approximation" [18] the mean reversal field $\overline{H}_r(T)$ is given by the following expression:

$$\ln\left(1 - \frac{\overline{H_r}(T)}{\overline{H_r^0}}\right) = \frac{1}{\alpha}\ln T + \frac{1}{\alpha}\ln\left[\frac{2k}{\mu_0 M_s \overline{H_r^0} V_n}\ln\left(\frac{2\tau_{\rm acq}}{\tau}\right)\right], \qquad (3)$$

where $\tau_{\rm acq} \simeq 30$ s is the approximate acquisition time for one single point and $\overline{H_r^0}$ is the mean reversal field of the array at $T = 0$ K. In the case of magnetic relaxation the maximum magnetic viscosity occurs for $H = \overline{H_r}(T)$ and equals

$$S_{\max}(T) = \frac{1}{\alpha[\ln(\tau_{\rm acq}/\tau_0)]^{(\alpha-1)/\alpha}}\left(\frac{kT}{V_n}\right)^{1/\alpha}\rho_{\max}, \qquad (4)$$

where $\rho_{\max}$ is the maximum value of the reversal field distribution function over the array. Experimental magnetic viscosity data between 2 and 300 K were fitted using Eqs. (3) and (4) to estimate α and V_n. We found $\alpha = 1.75$ and $\alpha = 1.56$ with the mean reversal field and the viscosity analyses, respectively, which is close to the predictions of CR [9,19], i.e., and to first order: 2 when **h** is applied exactly along the anisotropy axis and 1.5 when **h** is applied in any other direction. V_n can be expressed in terms of an in-plane activation length ξ ($V_n = t\xi^2$) as the magnetization is uniform throughout the dots due to the small thickness t. We found $\xi = 20.8$ and 20.2 nm, respectively, or 1.02 and 0.99 in terms of $\lambda_{\rm Bl.}$. It is reasonable to find that $\xi \sim \lambda_{\rm Bl.}$ as both have the same meaning, i.e., the characteristic length scale of in-plane magnetization rotation, viewed from the dynamical and statical points of view, respectively. However, V_n is still only around 1% of the dot total volume. This indicates that the reversal is indeed *not* coherent. There is, however, no discrepancy between this statement and the nearly CR law as obtained from $h_r(\theta)$ experiments (Fig. 2): nucleation volumes are expected to grow near the edges where demagnetizing fields are maximum. As the demagnetizing energy is weak nucleation volumes reverse for an $h_r(\theta)$ value which is not very far from $h_{\rm CR}(\theta)$. In other words, the nucleation is not coherent within the dots, but as the nucleation volumes behave close to uniform magnetization the $h_r(\theta)$ law is very similar to that of CR.

We developed an analytical model called "torque model," whose hypotheses rely on the experimental observations described above. In the present Letter we restrict ourselves to introducing the model and its major conclusions. A more detailed description will be reported elsewhere [13].

The torque model relies on the following four hypotheses.

(1) The magnetization lies in plane and is uniform throughout the dot thickness.

(2) The dot geometry is simplified to a half-infinite thin film whose edge is perpendicular to the in-plane easy axis of magnetization. This was motivated because corners generally hinder magnetization reversal, so that nucleation sites should be located on edges and away from corners. As ξ is by far smaller than the dot edge length nucleation volumes should not feel the influence of corners and the reversal should be determined by the edges' geometry only.

(3) Dipolar fields in 2D systems are short ranged [11], with a characteristic length scale of the order of the system thickness t. Here $t \ll \lambda_{\rm ex.}$ and $t \ll \lambda_{\rm Bl.}$ so that the dot self-demagnetizing effect is nearly pinpointlike as compared to ξ. This demagnetizing effect was accordingly taken into account as an edge torque in the edge micromagnetic Brown equation.

(4) Low order expansion terms only were kept in equations. This is justified by the quasisingle domain state of nucleation volumes.

Within this framework equilibrium states are found by solving the following equation:

$$1 + h - \omega_0^2/4 = D^2(h)(1 - \omega_0^2 + \omega_0^4/4), \qquad (5)$$

where ω_0 is the magnetization rotation at the dot edge, with $\omega_0 = 0$ for single domain state, and $D(h)$ is defined as

$$D(h) = \frac{\mu_0 M_s^2 t}{4\pi\sqrt{AK}}\left[1 - C - \ln\left(\frac{t}{2}\sqrt{1+h}\sqrt{K/A}\right)\right], \qquad (6)$$

where $C = 0.577\ldots$ is Euler's number. Note that h has negative values in the last two equations ($h = -1$ stands for CR). Equations (5) and (6) were solved numerically to determine stable and unstable equilibrium states for each applied field. The resulting reversal diagram calculated using the low-temperature parameters of 1-nm-thick dots $H_a = 0.55$ T and $M_s = 1.7 \times 10^6$ A m^{-1} is shown in Fig. 3.

The following points can be inferred.

(1) As expected the reversal field absolute value is found to increase with decreasing thickness. Ultimately coherent rotation occurs for $t = 0$. This can be understood as self-demagnetizing fields have then completely vanished. The model yields $|h| = 0.84$ for $t = 1$ nm while $|h| = 0.73$ is the experimental value. The

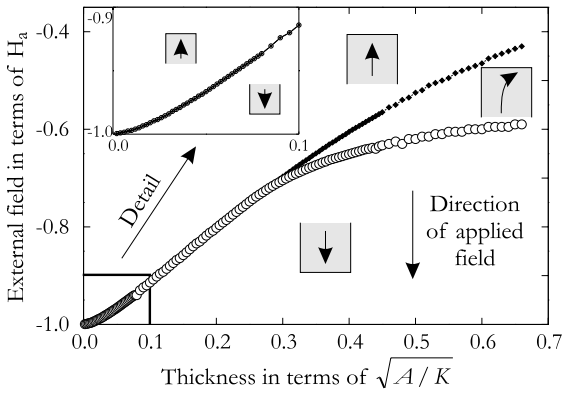

FIG. 3. Magnetization state of a dot vs thickness and negative applied field, calculated with the torque model for $H_a = 0.55$ T. Uniform-to-nonuniform second-order transition line (◆) and first-order reversal line (○) are displayed. Calculated data close to the origin are shown on an enlarged scale in the inset.

agreement is satisfactory. Reversal field values yielded by micromagnetic calculations on disk- or square-shaped dots are in very good agreement with these predicted by the model [13], so that the statistical surface/edge roughness and slight nominal thickness underestimation could be blamed for the discrepancy with experiments.

(2) Static nonuniform magnetization states are never favored below a critical thickness t_c. This could not be checked experimentally due to the weakness of the overall magnetization.

(3) The torque model eventually allows the calculation of the exponent α of Eq. (1). We found $\alpha = 2$ for $t < t_c$ and $\alpha = 1.5$ for $t > t_c$. Experimental data are available for 1-nm-thick dots only (which are predicted to lie in the range $t < t_c$). We found $\alpha = 1.65 \pm 0.1$ (1.56 or 1.75 depending on the method; see above) in the range 2–300 K. As $\alpha = 2$ and 1.5 result from first order expansions, direct comparison is not possible. Instead one has to fit a polynomial to the energy well predicted by the model in the energy range 2–300 K. This yielded $\alpha = 1.65 \pm 0.02$, in excellent agreement with the experiment.

To conclude, we fabricated submicrometer-sized dots out of ultrathin epitaxial (110)Fe films with in-plane magnetization, with negligible interdots dipolar coupling. We demonstrated the correlation between the reduction of thickness and the increase of coercive fields. This behavior was ascribed to the fact that for in-plane magnetization magnetic poles are located on dots' edges, so that reducing the dot thickness reduces the dots' internal demagnetizing fields. This effect is further enhanced by the fact that dipolar fields are very short ranged in 2D systems. We also showed that in the thinnest ($t \simeq 1$ nm) dots the experimental angular dependence of the reversal field $[h_r(\theta)]$ was close to that predicted by the CR model, although activation volumes equal approximately 1% only of the total volume of one dot, i.e., although magnetization reversal is clearly *not* coherent. This demonstrates that a nucleation-propagation reversal may nearly follow the CR law if nucleation sites environments show conditions close to uniform magnetization, e.g., with negligible demagnetizing fields here. Finally, we proposed an analytical model (torque model) of magnetization reversal in such dots. The model outputs are in quantitative agreement with experimental results, both in terms of reversal field and α exponent of the field dependence of the relaxation barrier height.

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