Single Pt/Co(0.5 nm)/Pt Nano-discs: Beyond the Coherent Spin Reversal Model and thermal stability


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We investigate the dynamics of the magnetization reversal in single Pt/Co(0.5 nm)/Pt nano-discs with diameter 130 nm, fabricated by He⁺ ion irradiation. They exhibit a very narrow distribution of small switching fields and a perpendicular magnetic anisotropy. In spite of their small magnetic volume, thermally activated magnetization reversal cannot be interpreted within the usual Néel-Brown model. Non-coherent magnetization reversal proceeds here by fast nucleation at nano-disc borders and rather slow wall motion towards their center. Dynamics are perfectly accounted for by a 2D-confined droplet model, involving the wall energy rather than the anisotropy energy. On the other hand, the blocking temperature for these nano-discs is well described by the coherent spin reversal model.

Key words: magnetic nano-elements, patterned ion-irradiated structures, Néel-Brown model, superparamagnetic behavior, perpendicular magnetic recording

1. Introduction

Considerable efforts have been devoted to the investigation of mechanisms involved in the magnetization reversal of nano-particles grown by bottom-up methods or of patterned nano-elements fabricated by top-down techniques.

One challenge is to evidence and study reversal processes. The simplest one is the coherent magnetization reversal, strictly valid in ellipsoidal shaped particles, theoretically treated at $T = 0$ K a long time ago by Stoner · Wohlfarth and Néel$.^{1}$ Later, Néel and Brown$^{2}$ investigated the effect of thermal activation for such a reversal process. Only low temperature micro-SQUID investigations$^{3}$ have demonstrated the relevance of the Néel-Brown predictions for interpreting the magnetic switching in a cobalt nano-particle.

A second challenge relates to the thermal stability of discrete magnetic recording media, generally interpreted from a macrospin picture assuming valid a coherent spin reversal process in each nano-element, that is not always valid. A more realistic treatment of this problem is to consider the real shape and size of nano-elements without supposing ad-hoc activation volumes.

Unfortunately, most reported results are also limited in their analysis because they are testing assemblies of polydisperse magnetic nano-objects$^{4,9}$. As in$^{9}$, experiments on isolated nano-elements are often required to test precisely if their magnetization reversal obeys to a macrospin behavior and Stoner-Wohlfarth predictions with a switching probability of the magnetization, $P(t)$, expressed as:

$$P(t) = 1 - \exp \left( \frac{t}{\tau} \right)$$  \hspace{1cm} (1)

with $\tau = \tau_0 \exp(\Delta E/k_{\text{B}}T)$  \hspace{1cm} (2)

where $t$ represents the measurement time, $\tau_0$ is a characteristic microscopic time of the order of $10^{-11}$ s. In zero field, the energy barrier involved, $\Delta E(H = 0)$, is the product of the effective magnetic anisotropy, $K$, by the volume of the nano-element, $V$, while in a field applied along the anisotropy axis

$$\Delta E(H) = KV(1 - \frac{H}{H_A})^{1/2}$$  \hspace{1cm} (3)

if $H_A$ is the anisotropy field related to $K$ by the relation $\mu_0 H_A = 2K/M_S$.

In the case of high aspect ratio nano-elements and/or with dimensions large as compared to the exchange length, $P(t)$ is no longer an exponential function of $t$. A stretched exponential is generally more appropriate to fit the switching probability law, a characteristic feature for a reversal implying several processes. However, one can wonder if the reverse is true: “Does a time dependent exponential variation of $P(t)$ certify a coherent spin reversal?”.

2. Sample and Experimental Methods

To treat these two challenges, we have considered the case of extremely flat Pt/Co(0.5 nm)/Pt nano-discs with a diameter $D = 130$ nm exhibiting an easy magnetic anisotropy axis perpendicular to the film plane. Square arrays of these nano-discs were fabricated by uniform He⁺ irradiation 130 nm, fabricated by He⁺ ion irradiation. They exhibit a very narrow distribution of small switching fields and a perpendicular magnetic anisotropy. In spite of their small magnetic volume, thermally activated magnetization reversal cannot be interpreted within the usual Néel-Brown model. Non-coherent magnetization reversal proceeds here by fast nucleation at nano-disc borders and rather slow wall motion towards their center. Dynamics are perfectly accounted for by a 2D-confined droplet model, involving the wall energy rather than the anisotropy energy. On the other hand, the blocking temperature for these nano-discs is well described by the coherent spin reversal model.

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ion irradiation at low fluence through a mask formed by cylindrical resist nano-pillars. The fluence was chosen so that, with the exception of the areas hidden from the ion beam by the nano-pillars, the film area is rendered paramagnetic. The mask was designed by electron lithography and removed before measurements, just after the ion irradiation process. Note that in contrast to a standard ion beam etching procedure, this soft patterning technique preserves film planarity, allowing refined quantitative microscopy investigations. Due to ion beam scattering and the slightly canted profile of the basis of nano-pillars, partial irradiation with a gradient profile perturbs the magnetic state at the nano-disc periphery, strongly reducing the anisotropy and the exchange constant over an estimated (25 ± 5) nm wide peripheral ring. Note also that the residual irradiation in nano-disc cores through the resist is estimated to be $D = 4 \times 10^{15}$ ions/cm$^2$.

The macrospin behavior and Néel predictions at room temperature were tested on this first patterned dot array. Under this fluence the Curie temperature is certainly higher than 900 K. In spite of the relatively small volume of the nano-discs, the hypothetical Néel blocking temperature, $T_B$, is calculated to be higher than 800 K.

Domain nucleation and subsequent wall propagation processes are presumably favored with respect to a quasi-coherent spin reversal because the nano-disc diameter exceeds much the exchange length and wall width (~ 10 to 20 nm). The nucleation field needed for reversing magnetization in the peripheral ring is far weaker than the propagation field in the core of the nano-disc. Thus, nucleation should take place first in the ring at very short time, so one expects to be only sensitive to the wall propagation process for times exceeding 50 ns. The final magnetization state of each nano-disc after applying a field ($0 < \mu_0 H < 50$ mT) pulse with variable duration $\Delta t$ ($10^{-7} < \Delta t < 10^{-2}$ s) was probed by high spatial resolution (400 nm) polar MOKE (PMOKE) microscopy using 500 nm wavelength LED light. Depending upon the time range, the field was generated either by one of the two coils (3 and 100 turns) mounted very near from the sample surface and connected to a pulse current generator delivering up to 50 A, or by an electromagnet. The rise time of the smallest coil (~ 15 ns) is fast enough to have no incidence on dynamic measurements.

Note that for such a study on individual nano-objects, micro-SQUID and SP-STM can, in principle, also be used. However, these last techniques are limited to low temperature studies of slow dynamics. Moreover, SP-STM needs in-situ nano-fabrication. Only micro-Hall measurements and Magnetic Force Microscopy have been performed so far on a unique Co/Pt multilayer nano-dot, but without testing dynamics accurately.

Several (100 µm x 100 µm) nano-element arrays with different nano-dot size were fabricated by the above-mentioned He$^+$ ion irradiation technique. We emphasize the study of single $D = (130 \pm 5)$ nm Pt/Co(0.5 nm)/Pt nano-discs arranged as a square array with 1.2 µm periodicity (Fig. 1a). The diameter $D$ was measured by both scanning electron and atomic force microscopy. The disc separation (~ 1 µm) is sufficient to suppress magnetostatic and exchange interdot interaction and to investigate the nano-discs by PMOKE microscopy independently. The square shape of the hysteresis loop (Fig. 1b) supports that magnetization reversal in a single nano-disc proceeds by fast nucleation in the peripheral ring followed by wall propagation with weakly dispersed switching fields.

Considering measurements of the anisotropy field, $H_a$, in irradiated films under variable He$^+$ fluence, we estimated $\mu_0 H_a = (230 \pm 50)$ mT in the weakly irradiated disc core. Its saturated magnetization is estimated to be $M_S = (1050 \pm 100)$ kA/m.

Considering the experimental procedure, all processed PMOKE images are differences between the sum of 64 PMOKE remnant images acquired after applying a magnetic field pulse and the sum of 64 images of the magnetized state at saturation (Fig. 1a).

A second set of measurements was performed to check the validity of the coherent reversal process close to $T_B$. For that purpose an additional controlled uniform irradiation of the nano-disc array by He$^+$ ions allowed us to reduce $T_B$ down to room temperature.

3. Macrospin reversal and Néel’s predictions

In this work we demonstrate that single nano-disc magnetization reversal cannot be interpreted within the Néel-Brown’s model. Following a previous concept introducing droplets7,14-16 for explaining magnetization reversal in thin films, an adapted 2D confined droplet model is proposed here to interpret dynamics in nano-discs extending the previously proposed spherical and planar models17,18. The wall energy is now the significant parameter, the volume $V$ in Eq. (2) being replaced by the wall section area.

![Fig. 1](image-url)
Consider first the switching probability $P(\Delta t)$ for a single nano-disc versus the pulse duration $\Delta t$ at a fixed field ($\mu_0 H = 24$ mT) applied along the anisotropy axis (Fig. 2). For each disc, $P(\Delta t)$ can be fitted by an exponential law with $\Delta t$, consistent with Eq. (1). We are sensitive here to only one mechanism because field-induced nucleation is easy and fast (< 50 ns) as compared with the probed wall propagation dynamics. From disc to disc, all $P(\Delta t)$ curves follow the same universal exponential law with a time constant $\tau_{1/2}$ distributed over less than one decade. This means that we are checking only a single reversal process in all nano-discs, the wall propagation. Unlikely, for larger nano-discs ($D = 440$ nm), the probability law can only be fitted by a stretched exponential, proving that several processes are implied during the magnetization reversal.

Second, the variation of the switching field $H_{SW}$ of a single ($D = 130$ nm) nano-disc was measured as a function of the pulse field duration $\Delta t$ (Fig. 3). Again, in view of the small distribution of $H_{SW}$ (< 10%), the $H_{SW}(\Delta t)$ renormalized curves with respect to $\tau_{1/2}$ for all nano-discs display a universal behavior. This proves again that we are probing a unique reversal mechanism in each nano-disc. For an extended pulse duration range, between 100 ns and 80 s, the switching field is always much lower than the anisotropy field ($\mu_0 H_A = 230$ mT). The variation of the activation energy $\Delta E$ with $H_{SW}$ is then obtained by the following manner. The variation of the switching time $\Delta t$ as a function of the applied magnetic field is given in Fig. 3. Since Eq. (2), links $\Delta E$ to $\tau \sim \Delta t$, and assuming $\tau_0 = 10^{-11}$ s, one can deduce the variation of $\Delta E$ with $H$, as plotted in Fig. 4 (empty circles). The experimental data show that $\Delta E(H)$ is about half the value expected for a coherent reversal in a nano-disc with a soft magnetic ring (Fig. 4). This proves that the Néel-Brown prediction cannot be extended to interpret magnetization reversal in high-aspect ratio nano-elements.

We then constructed an adapted analytical confined droplet model extending previously proposed spherical and planar treatments\cite{17, 18}, assuming nucleation in a small initial volume at the ultrathin ($h = (0.5 \pm 0.05)$ nm) nano-disc edge (in the soft peripheral ring region) and magnetization reversal by subsequent wall droplet propagation towards the center of the hard core region (with diameter $D_{hc} = (80 \pm 10)$ nm). This model describing wall propagation leads to an exponential dependence of $P(\Delta t)$, and allows us to finally deduce the $\Delta E(H)$ activation energy variation (Fig. 4). For this simple and non-coherent reversal mode, $\Delta E(H = 0) = D_{hc} h \sigma = 4 D_{hc} h (A K)^{1/2}$, is now linked to the domain wall energy, i.e. the product of the wall energy density by the cross-section area of the nano-disc, rather than to its volume, as in the Néel-Brown model. Starting from the magneto-optically measured value of $K = 1.21 \times 10^5$ J/m$^3$, considering a reasonable value for $A = 0.8 \times 10^{-11}$ J/m for a film.

Fig. 2: Magnetization reversal probability of a ($D=130$ nm) Pt/Co(0.5 nm)/Pt single nano-disc as a function of the pulse field duration, $\Delta t$.

Fig. 3: Variation of the switching field $H_{SW}$ of a single nano-disc with the field pulse duration, $\Delta t$.

Fig. 4: Field dependence of the energy barriers calculated from the Néel-Brown and refined confined droplet models assuming the existence of a soft magnetic ring at the nano-disc periphery in both cases. Comparison with experimental data.
irradiated under a residual $0.4 \times 10^{16}$ He$^+$ ions/cm$^2$ fluence, one deduces $\sigma = 3.94 \times 10^3$ J/cm$^2$, i.e. $\Delta E(H = 0) = 1.57 \times 10^{-19}$ J = 0.97 eV.

4. Superparamagnetic behavior and Néel’s prediction

The previous nano-disc array was submitted again to an additional uniform He$^+$ ion irradiation with a fluence of $8 \times 10^{15}$ ions/cm$^2$. This meant that the core was irradiated by a total fluence of $1.2 \times 10^{16}$ He$^+$ ions/cm$^2$, the width of the peripheral weak ferromagnetic ring being reduced to about half of its previous value, i.e. 12 nm. $T_c$ was estimated to 480 K from the disappearance of the magneto-optical Kerr effect in a uniformly irradiated virgin film at the same fluence. Consequently, room temperature measurements were performed far below $T_c$.

Consequently, the effective anisotropy, $K$, and the exchange stiffness, $A$, of the nano-disc cores were significantly reduced ($A \approx 3 \times 10^{-12}$ J/m, $K = 4.95 \times 10^4$ J/m$^3$). This decreases the energy barrier $\Delta E$, bringing the blocking temperature $T_B$ close to $T = 298$ K, i.e. our measurement temperature. For the confined droplet model, we deduced $\Delta E(H = 0) = 4\Phi_{	ext{w}} h (A K)^{1/2} = (6.2 \pm 1.2) \times 10^{-20}$ J. Using Eq. (2):

$$T_B(t) = \Delta E(H=0)k_B \ln(t/t_0)$$

(4)

t = 10^2$ s being the typical time for 64-images averaging. Assuming $t_0 = 10^{-11}$ s, we find $T_B(10^2$ s) = $(150 \pm 30)$ K, a value far below room temperature which would imply unrealistically large exchange stiffness or anisotropy constant. Whereas, starting from the Néel-Brown prediction and still considering the volume of the core, $\Delta E(H = 0) = KV = (1.06 \pm 0.18) \times 10^{-19}$ J, the blocking temperature is then estimated to $T_B(10^2$ s) = $(330 \pm 50)$ K, in good agreement with measurements at room temperature. This probably means that, for this last system, the magnetization reversal mode becomes quasi-uniform inside the core of the nano-discs. Indeed, well localized nucleation regions no longer survive since the estimated width (~12 nm) of the magnetically soft ring becomes smaller than the domain wall width (~25 nm). Close to the blocking superparamagnetic temperature, $T_B$, large fluctuations in zero field generate an expected telegraphic noise$^3$, i.e. nano-disc magnetization fluctuates between the two « up » and « down » stable macrospin states. Such a telegraphic noise was directly visualized in real time on our P/MOKE microscopy snapshots for this irradiated nano-disc array. The found value of $T_B$ is also consistent with the fact we observed telegraphic noise with a mean time constant of $10^8$ s at room temperature. As expected from the field-dependent expression of the energy barrier, $\Delta E(H) = KV(1-\langle H/H_A \rangle)^2$, the telegraphic noise disappears under weak applied fields (~0.5 mT).

So, the coherent reversal model is adequate for explaining reversal close to $T_B$, confirming the expectation that strong fluctuations restore a more coherent spin reversal behavior inside nano-discs.

5. Conclusion

The magnetization reversal in a flat single domain nano-disc with perpendicular anisotropy cannot be interpreted within the usual Néel-Brown prediction. A proposed confined droplet model is better adapted, as shown here in the case of Pt/Co(0.5 nm)/Pt nano-discs patterned by He$^+$ ion beam irradiation through a mask. By this fabrication procedure a soft magnetic ring region surrounds each nano-disc, favoring field-induced nucleation at their edge. We thus realize a narrow distribution of switching fields only controlled by wall propagation$^{10,20}$. Another advantage of such patterned system is a reduction of the switching field while the temperature stability relates to the full volume of the nano-disc. This last property is analogous to that exhibited by spring magnets$^{21}$, hybrid soft-hard layers medium$^{22}$, or hybrid nano-particles$^{23}$

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References


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