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Thermal behavior of hard-axis magnetization in noninteracting particles with uniaxial anisotropy

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Experimental evidence is presented to support predictions made by an analytical model describing the temperature-dependent behavior of an assembly of noninteracting magnetic nanoparticles with uniaxial anisotropy under an external field. When the applied field is smaller than the anisotropy field of the particles and is oriented perpendicular to the easy axis, a maximum of the magnetization occurs at finite temperature. The theory shows good agreement with measurements of an array of CoCrPt nanoislands with uniaxial anisotropy. Deviations are discussed taking into account the thermal dependencies of the saturation magnetization and the anisotropy of the magnetic material. © *2009 American Institute of Physics*. [doi:10.1063/1.3265741]

Magnetic nanoparticle research is motivated both by the broad range of applications and by the necessity for basic understanding of physical phenomena occurring at this length scale.[1–3] For example, nanoparticle arrays have been studied as potential candidates for patterned recording media, with excellent areal density and good thermal stability.[4]

Although many experimental studies have been carried out, the theoretical models describing the collective behavior of these systems are usually made within the framework of simplified approximations. Recently, an analytical model by Vargas *et al.*[5,6] describing the behavior of an ensemble of noninteracting magnetic particles with uniaxial anisotropy was reported. They consider a system of noninteracting three-dimensional monodisperse ferromagnetic particles with uniaxial magnetic anisotropy. The model analyzes such a system under an external uniform magnetic field H, by expressing the energy of the system as simply the sum of the Zeeman and anisotropy energies. For simplicity, the saturation moment M_s and the uniaxial anisotropy K_u were taken as constant for each particle of volume V (the particle moment is $M=M_sV$). The average thermodynamic magnetization $\langle M\rangle$ of each particle along the field was expressed as

$$\langle M\rangle = kT\frac{\partial \ln Z}{\partial H},$$

where T is the temperature and Z is the partition function which depends on both energy and temperature.[6]

An important result from approach of Vargas *et al.*[5,6] is that at finite temperatures the thermodynamics of the system can be solved analytically. When the applied field H is *perpendicular* to the anisotropy axis and with magnitude smaller than the anisotropy field $H_K(=2K_u/M_s)$, a maximum in the temperature-dependent magnetization is predicted, contrary to the behavior of paramagnetic particles. At absolute zero, the moment of the particle would align at an angle $\pm\theta$ relative to the applied field. The angle θ decreases as H/H_K increases, and reaches zero when $H=H_K$. For a fixed $H/H_K<1$, an increase in temperature causes fluctuations in θ, and the asymmetric shape of the energy well leads to a decrease in the time-averaged $\langle\theta\rangle$, with a corresponding increase in the time-averaged value of $\langle M\rangle$ measured parallel to H. At higher temperatures, however, the fluctuations in θ can overcome the potential barrier between the two minima, and the time-averaged $\langle M\rangle$ decreases. This leads to a maximum in $\langle M\rangle(T)$ at a given temperature $T_{\max}$.[5]

In this letter, we present experimental evidence confirming the presence of a magnetization maximum at a finite temperature. To study this effect, arrays of magnetic nanoislands of Ti/CoCrPt with perpendicular anisotropy were fabricated by block copolymer lithography. Ti is used to promote the alignment of the cobalt c-axis (the easy magnetization axis) perpendicular to the film plane, and the alloyed platinum increases the anisotropy of the cobalt.

To prepare the samples, films of 5 nm Ti/10 nm CoCrPt/50 nm SiO_2 were sputter deposited on prime Si wafers at room temperature. A poly(styrene)-*b*-poly(ferrocenyldimethylsilane) (PS 69 kg/mol–PFS 21 kg/mol) block copolymer solution in toluene was spin-cast on top. After annealing at 180 °C a two-dimensional close-packed monolayer array of PFS spheres embedded in a PS matrix is formed. A series of dry etching steps was used to remove the PS to expose the PFS spheres, and then to transfer the PFS pattern into the silica and then into the magnetic layer forming a close-packed array of magnetic dots.[7,8] Magnetic measurements were carried out in a superconducting quantum interference device magnetometer in the temperature range 4–350 K.

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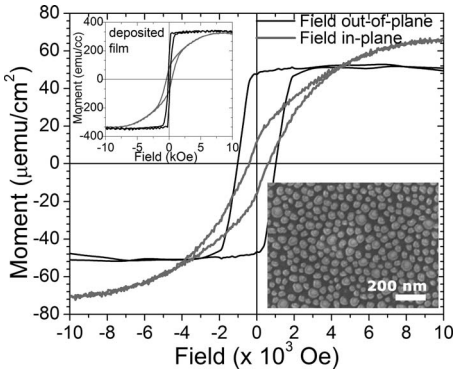

FIG. 1. $\langle M \rangle$ vs H hysteresis loop of the patterned sample, shown in the lower inset, measured with the external field oriented parallel to the easy axis of the islands (perpendicular to the substrate) at 300 K. The gray hysteresis loop was taken from a different sample (with CoCrPt islands that are 15 nm tall, with 35 nm diameter, and period of 50 nm) and shows a typical hard-axis behavior with the field parallel to the substrate. The upper inset shows in-plane and out-of-plane measurements of an as-deposited unpatterned CoCrPt/Ti film.

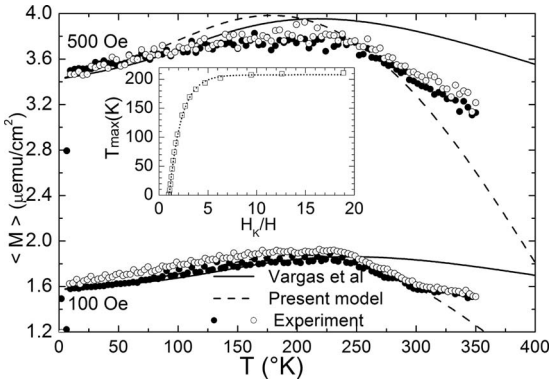

FIG. 2. $\langle M \rangle$ vs T curves of the sample measured at 100 and 500 Oe, when H is oriented perpendicular to the easy axis of the CoCrPt dots; solid circles are measured with decreasing temperature, while open circles are measured for increasing temperature. Fits from the Vargas *et al.* model (solid lines) and the present model (dashes) are shown. The inset shows the relationship between T_{max} and H, with a fit to the points that is linear for small $H_K/H - 1$.

Figure 1 shows the hysteresis curve of the sample measured at room temperature. From the hard-axis loops of the as-deposited film, we can see that $H_K \approx 8000$ Oe giving an out-of-plane anisotropy $K_U > \sim 1.4 \times 10^6$ erg/cm^3; at $T=0$, $\theta=0.7°$ at an applied field of 100 Oe, and $\theta=3.6°$ at 500 Oe. Measured values of $M_S=348$ emu/cm^3 are in agreement with literature values.[9] As can be seen from the inset of Fig. 1, the CoCrPt islands have a distribution of sizes (the average area as measured from the micrograph is 1895 nm^2 corresponding to an average diameter of around 50 nm) and their positions are poorly ordered. The diameter of the islands exceeds that of the initial PFS sphere array, which is attributed to coalescence of some of the PFS spheres during the pattern transfer process. The height of the magnetic layer of each island is 10 nm, equal to the thickness of the deposited film. Magnetostatic interactions between neighboring islands are on the order of 180 Oe (calculated by assuming a hexagonal array of CoCrPt islands with diameter of 50 nm, height of 10 nm and 60 nm center-to-center distance),[7] which is small compared to the average switching field of 1000 Oe, so that the array may be initially treated as noninteracting.

Figures 2 and 3 show the experimental field cooling (FC) magnetization curves at two different measurement fields, along with fitting curves obtained from two analytical models: the model of Vargas *et al.*[5,6] and a modified model described later in this paper. The FC measurement was carried out as follows: first, the sample was saturated at room temperature (300 K) using a field of 7.0 kOe along the easy magnetization direction (perpendicular to the substrate). This ensures all islands are magnetized in the same direction. Next, the sample was cooled from 300 to 4 K, in the presence of a magnetic field (0.1 or 0.5 kOe) applied along the hard direction (parallel to the substrate). With the field still applied, the magnetization was then measured in the hard direction as the temperature was increased. For both measurement fields, a clear maximum of magnetization is observed at finite temperatures. The maximum is a fingerprint of a second order magnetic transition and is found only when the hard-axis measurement field is lower than the anisotropy field of the particles, as predicted by the model.[5] Furthermore, we observed the same maximum of $\langle M \rangle$ both upon increasing and decreasing the temperature without saturating the sample between measurements, suggesting this effect is reversible and not a result of demagnetizing the islands, which further supports the thermodynamic nature of the effect.[5,6]

One can observe that the measurement agrees rather well with the Vargas *et al.* model at low temperatures, but at temperatures higher than that of the maximum in $\langle M \rangle(T)$, the agreement is poor. This could result from simplifying assumptions in the Vargas *et al.* model. Vargas *et al.* considers each particle as a single magnetic moment of a fixed value independent of temperature whose magnetization reverses by coherent rotation, and neglects the temperature dependence of the uniaxial anisotropy K_u. The actual sample has temperature dependent M_s and K_u, and a rather broad distribution of particle sizes. The c-axis distribution has a width of approximately 5°, according to x-ray diffraction measurements, so dispersion in the easy axis directions is not considered. As a first approach we consider the temperature dependencies of both the magnetic moment and the magnetic anisotropy. As in the Vargas *et al.* model,[5] dipolar interactions between neighboring dots are neglected as they are small compared to the average switching field. If dipolar interactions were significant in the sample, the signature

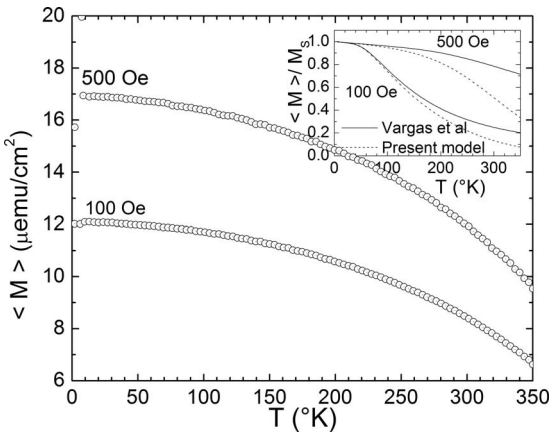

FIG. 3. Magnetization vs temperature curves of the nonsaturated sample, when H is applied parallel to the easy axis of the CoCrPt dots. The inset shows the model behavior in which $\langle M \rangle$ decreases monotonically with T.

maximum of $\langle M\rangle(T)$ would not be present in the measurement.

Data for the temperature dependence of the magnetic moment $M_s(T)$ and anisotropy $K_u(T)$ for CoCrPt were fitted as follows:

$$\frac{K_u(T)}{K_u(0)} = \kappa(T) = a - b\ \tanh[c(T-d)],$$

where $a=0.3956$, $b=0.6275$, $c=0.0054$, and $d=373.4$, and

$$\frac{M_S(T)}{M_S(0)} = \left[1 - \left(\frac{T}{T_C}\right)^{2.5}\right],$$

where $M_S=350$ emu/cm^3 (measured at room temperature) and is in agreement with previously reported values,[9,10] $T_C=570$ K is the Curie temperature as measured for this sample, and K_u and all temperature dependencies were obtained from literature.[9,10]

The results of the modified model (which takes into account temperature-dependent anisotropy and magnetization) are shown in Fig. 2 as a dashed line. It is clear that in the measured temperature range the modified model fits the experimental data better than the previous model for both measurement fields (100 and 500 Oe). The relationship between the temperature $T_{\max}$ where the maximum in $\langle M\rangle$ occurs and the field is a nonlinear, monotonically increasing function of H_K/H that reaches a constant value for $H_K/H>6$ (see inset of Fig. 2). For small values of $((H_K/H)-1)$ (near to the phase transition) the relationship is approximately linear, and can be approximated by $T_{\max}=128.76((H_K/H)-1)$.

Figure 3 shows the magnetic behavior of the sample during field cooling with an applied field of 100 or 500 Oe parallel to the easy axis of the dots (i.e., perpendicular to the sample plane). In this measurement, the sample was not saturated between measurements. The moment decreases monotonically with temperature when the field is applied parallel to the easy axis, in contrast to the observed behavior when the field was applied perpendicular to the easy axis. This is consistent with the model that predicts a maximum in $\langle M\rangle \times(T)$ only when the field is applied perpendicular to the easy axis. The predicted theoretical behavior when the field is applied parallel to the easy axis is plotted as an inset to Fig. 3. We see that both for the measurement and the model the magnetization decreases monotonically. The difference in the rate of decrease between the model and measurement demonstrate the need for further refinement of the model. The reason that the measured $\langle M\rangle$ at zero K differs between the two fields is that the sample was not saturated between the two measurements, so the 100 Oe data reaches a lower maximum magnetization value.

In conclusion, this report experimentally verifies that an ensemble of magnetic particles with collinear uniaxial anisotropy axes has a magnetization maximum at finite temperature when an external field $<H_K$ is applied perpendicular to the easy axis, but not when the field is parallel to the easy axis. This hard-axis behavior is a fingerprint of a second order phase transition in which the order parameter is the magnetization along the applied field. In this letter we show an experimental confirmation of this phenomenon, using an improved model which includes temperature-dependent anisotropy and magnetization. This result is relevant in applications where the temperature-dependent behavior of uniaxial magnetic particles is important, such as in high density magnetic memory.

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