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Monte Carlo study of the magnetic behavior of self-assembled nanoparticles ☆

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Abstract

The blocking behavior of quasi-two-dimensional hexagonal arrays of magnetic nanoparticles are studied by Monte Carlo simulations. Structural deviations from the perfect arrangement are considered (voids, incomplete coverage). We show that interparticle dipolar interactions are responsible for an increase of the blocking temperature with the coverage of the film and a decrease of the blocking temperature with the cube of the interparticle distance. Our results compare satisfactorily with recent experiments.

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Keywords: Nanoparticle arrays; Magnetic nanoparticles; Blocking temperature; Interparticle interactions; Monte Carlo simulation

Highly monodisperse periodic arrays of magnetic nanoparticles with diameters of the order of a few nanometers [1–4] consist ideal systems to study the magnetization reversal mechanism in ensembles of dipolar interacting nanoparticle and at the same time they are strong candidates as the next generation magnetic storage media ($\sim$1 Tb/in^2).

Due to the chemical route followed for the preparation of nanoparticle arrays, a few uncontrollable defects (voids, misplaced particles and incomplete monolayers) are always present [3]. The anisotropy of the magnetostatic interactions to the spatial arrangement of the nanoparticles makes their role of considerable importance in these systems. In this work, we study the blocking behavior of planar arrays of dipolar interacting nanoparticles.

We consider identical spherical particles with diameter D forming a two-dimensional (2-D) triangular lattice in the xy-plane and lattice constant $d \geqslant D$. We construct a nanoparticle-assembled film with finite thickness (1–4 mono-

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layers, ML) by placing particles in the upper layer above alternate interstices in the lower one [3,4]. The uppermost monolayer is incomplete and is randomly occupied. The particles are single-domain, they possess uniaxial anisotropy in a random direction and they interact via dipolar forces. The energy parameters entering the total energy of the system are the dipolar energy, $g = \mu^2/d^3$, where $\mu = M_s V$ is the particle moment, the anisotropy energy, $k = K_1 V$, and the Zeeman energy, $h = \mu H$, due to the applied field H. For Co nanoparticles with hexagonally close-packed (HCP) or FCC atomic lattice structure, typical values are $K_1 \sim 10^6$ erg/cm^3 and $M_s \sim 10^3$ emu/cm^3 that give $g/k \sim 0.52(D/d)^3$. The magnetic configuration under an applied field H and a finite temperature T is obtained by a Monte Carlo simulation, using the standard Metropolis algorithm [5]. At a given temperature and applied field, the thermalization of the system is performed within the first 10^4 Monte Carlo steps per spin and thermal averages are calculated over the subsequent 10^3 steps. Simulations were performed on a $15d \times 15d \times L_z$ cell with $d_z \leqslant L_z \leqslant 4d_z$ and $d_z = d\sqrt{2/3}$ is the inter-layer distance. We use periodic boundaries in the xy-plane and free boundaries in the z-axis.

The zero-field-cooled (ZFC) magnetization curves of a series of samples have been calculated and from the maximum value of those, the blocking temperature of the system is extracted. In Fig. 1, we show the dependence of the blocking temperature of Co nanoparticle arrays on the coverage. A clear increase of the blocking temperature with layer coverage is seen, that almost reaches saturation as soon as the first complete monolayer is formed. Below 1 ML coverage, the increase of T_b is due to the anisotropic and ferromagnetic character of dipolar interactions that introduce an additional barrier to the magnetization reversal of the particles. A similar increase of T_b with particle concentration has also been observed in 3-D random assemblies of nanoparticles, as for example, in granular films [6–8]. Increased T_b values relative to the dilute limit have been recently measured in self-assembled Co nanoparticle arrays prepared from colloidal dispersions [3] and in self-organized

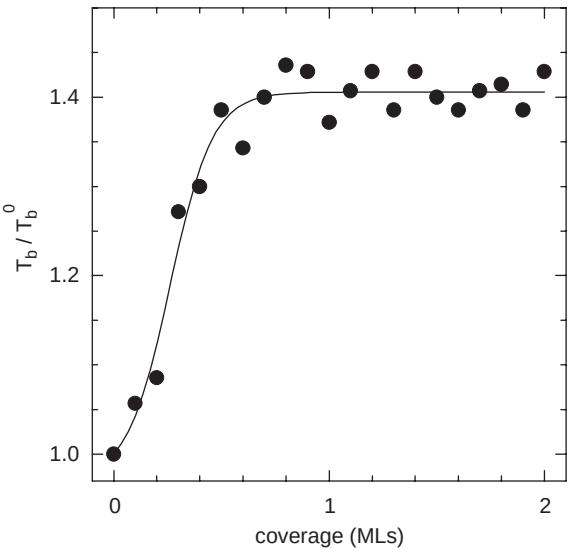

Fig. 1. Increase of blocking temperature (T_b) with layer coverage for Co nanoparticle arrays (circles) with $D = 5$ nm and $d = 6.3$ nm. Full line is a guide to the eye. In-plane applied field $H = 100$ Oe.

lattices of Co clusters in Al_2O_3 matrix [9]. However, in the latter experiments the saturation of T_b values was found after 5–7 ML, while in closed-packed hexagonal arrays we demonstrate that saturation occurs already after 2 ML. From this behavior, we conclude that in HCP spherical Co particles the collective behavior is predominantly determined by the intra-layer dipolar interactions, while inter-layer interactions play only a secondary role.

A strong dependence of the T_b on the inter-particle distance (d) is shown in Fig. 2. Our data indicate that for Co nanoparticles the blocking temperature scales as $T_b \sim (D/d)^3$. Given that the dipolar strength is $g \sim (D/d)^3$, we obtain $T_b \sim g$, which means that in hexagonal arrays of Co nanoparticles the dipolar interactions provide an additional energy barrier for magnetization reversal which is proportional to the dipolar coupling strength.

Our results for the blocking behavior are in qualitative agreement with recent measurements of ZFC/FC curves [3,9] indicating an increase of T_b relative to the dilute limit. More measurements in samples with different surfactants, which control

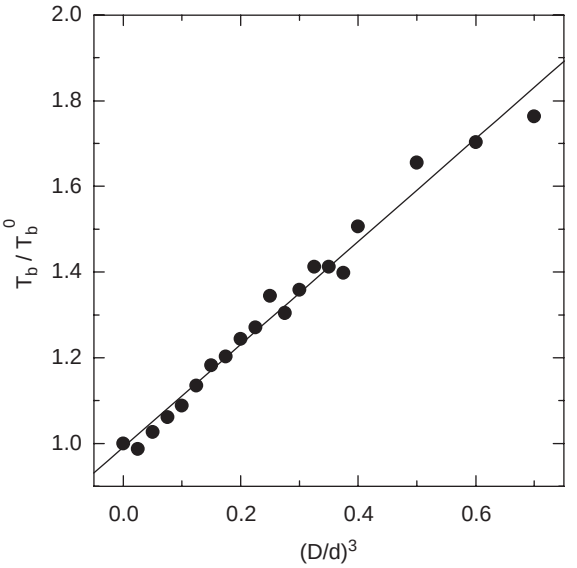

Fig. 2. Increase of blocking temperature (T_b) with decreasing particle separation (d) for an ideal hexagonal array of Co nanoparticles (circles). Straight line is a least-square fit to the simulation data. Same parameters as in Fig. 1 are used.

the interparticle distance [10], would help to verify the scaling behavior of T_b. Control of the layer coverage has so far been accomplished by variation of the colloidal dispersion concentration [4]. Our results could motivate more measurements in samples with controlled layer coverage, which will further elucidate the role of dipolar interparticle interactions in nanoparticle assemblies.

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References

[1] C. Petit, A. Taleb, M.P. Pileni, Adv. Mater. 10 (1998) 259.
[2] J.M. Harrell, S. Wang, D.E. Nikles, M. Chen, Appl. Phys. Lett. 79 (2001) 4393.
[3] C.B. Murray, S. Sun, H. Duyle, T. Betley, MRS Bull. 26 (2001) 985.
[4] V.F. Puntes, K.M. Krishnam, A.P. Alivisatos, Appl. Phys. Lett. 78 (2001) 2187;
V.F. Puntes, K.M. Krishnam, A.P. Alivisatos, Science 291 (2001) 2115.
[5] D. Kechrakos, K.N. Trohidou, Appl. Phys. Lett. 81 (2002) 4574.
[6] J.L. Dormann, D. Fiorani, E. Tronc, Adv. Chem. Phys. 98 (1997) 283.
[7] D. Kechrakos, K.N. Trohidou, Phys. Rev. B 58 (1998) 12169.
[8] R.W. Chantrell, N. Walmsey, J. Gore, M. Maylin, Phys. Rev. B 63 (2000) 024410.
[9] F. Luis, F. Petroff, J.M. Torres, L.M. García, J. Bartholomé, J. Carrey, A. Vaurès, Phys. Rev. Lett. 88 (2002) 217205.
[10] S. Sun, C.B. Murray, D. Weller, L. Folks, A. Moser, Science 287 (2000) 1989.