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Magnetic field dependence of the magnetocaloric effect in magnetic nanoparticle systems: A Monte Carlo simulation

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Abstract

We study the entropy change dependence on the applied magnetic field for a dipolar interacting superparamagnetic system of fine particles. Using a Monte Carlo technique, we have obtained an enhancement of the entropy for increasing applied magnetic fields; the maximum of the entropy curves occurs at higher temperatures for larger magnetic fields. It was also observed that the blocking temperature always decreases as the magnetic field increases.

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1. Introduction

The magnetic properties of a superparamagnetic system vary as a function of the applied magnetic field. The blocking temperature has been observed to decrease with increasing magnetic field [1], and the entropy change has been reported to increase as the applied magnetic field does [2]. With the aim of a better understanding of the underlying physical origin of these behaviors we have performed some numerical simulations of these nanostructured materials using a basic nanoparticle model where the energy terms are: anisotropy energy, field energy and dipolar interaction energy.

We have used a Monte Carlo technique to simulate the magnetic field dependence of the blocking temperature (T_B) under zero field cooling (ZFC) conditions. In a complementary form we introduce also the entropy change dependence with the external magnetic field for contributing to the study of the magnetocaloric effect.

2. Monte Carlo simulation conditions

The physical model employed for our numerical simulations is the same as in Ref. [3], where the particle sample behaves as a ferrofluid without aggregations where the positions of the particles are kept fixed and the easy axes are chosen randomly. The energy of each particle is considered to have three main sources: anisotropy (E_A), Zeeman (E_H) and dipolar interaction (E_D). The results obtained by these Monte Carlo simulations were confirmed experimentally, for example see Ref. [4].

In the superparamagnetic model it is assumed single domain magnetic particles with inner coherent magnetization rotation of the atomic moments, resulting a total particle magnetic moment of constant absolute value $|\vec{\mu}_i| = M_S V_i$, where M_S is the saturation magnetization,

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considered to be independent of the particle volume V_i. According to Ref. [3], we assume a monodisperse assembly due to polydispersion has a moderate importance in our present problem. This theoretical prediction is borne out by recent experimental works [4].

For the computational treatment of the energy, the particles were placed in a liquid-like arrangement with periodic boundary conditions. The Metropolis algorithm was applied over all the particles repeating the process until the magnetization of the system reaches a stability criterium.

In our simulations, the magnetic field is introduced by means of the related parameter $h = H/H_A$, where $H_A = 2K/M_S$ depends on the material saturation magnetization M_S and the anisotropy constant K. The number of particles per unit cubic lattice was N.

The volume of the particles V is kept fixed for all the simulations, with a value $V = 0.0015L^3c_0$, where L is the edge of the cubic simulation box and c_0 is a unitless constant characteristic of the material. The generic box size is the same for all the simulations, and $t = k_BT/2KV$ is defined as the reduced temperature, where K is the anisotropy constant of the particles, k_B is the Boltzmann constant and T is the temperature.

3. Results and discussion

We simulated the ZFC curves field dependence for the sample concentrations $N = 8$, 27 and 64 in order to obtain the reduced blocking temperature ($t_B = k_BT_B/2KV$) as the maximum of the ZFC-curve. The reduced blocking temperature shifts to lower values as the magnetic field increases, according to experimental data [2], as shown in Fig. 1. It is also observed that t_B increases with sample concentration for the same applied magnetic field value, according to the results of previous works [3–5].

From Fig. 1 it is also observed the existence of the superparamagnetic phenomenon as a function of the dipolar interaction: the blocking range is larger for larger sample concentration (N), what reflects that for a higher dipolar interaction the blocked state exists even for larger applied magnetic fields. It seems to exist a limit of the blocking temperature (t_{B^*}) independently of the dipolar interaction and of the strength of the applied magnetic field; it reflects the minimum of temperature introduced by the anisotropy interaction energy, necessary for the existence of the superparamagnetic phenomena.

In order to obtain the magnetocaloric effect we have calculated the corresponding entropy change. By the usual Maxwell's thermodynamics relation [6] for a discrete magnetic field and temperature variation, the entropy evolves approximately as a discrete summation [7], which converted to our reduced magnitudes results in

$$\Delta s_i = \frac{m(t_{i+1}, h) - m(t_i, h)}{t_{i+1} - t_i} \Delta h, \tag{1}$$

where $m = 0.0015\sum_i \cos\theta_i$ is the reduced magnetization and s stands for the resulting reduced entropy. In Fig. 2, the magnetic field dependence of the negative entropy change is plotted for temperatures above t_B, fitting the computational results to a Langevin-like function.

The negative entropy change undergoes a temperature increase as the applied magnetic field does, according to Ref. [2]. At the same time, the enhancement of the negative entropy shifts to higher temperatures for increasing magnetic fields, as indicated by the arrows. The behavior is analogous for the $N = 8$ and $N = 27$ samples.

The variation of the negative entropy change is proportional to the applied field, reflecting the linear Zeeman energy dependence of the system. It does not show any particular value for a larger negative entropy change, in a similar way as shown to occur with the dipolar interaction energy [5].

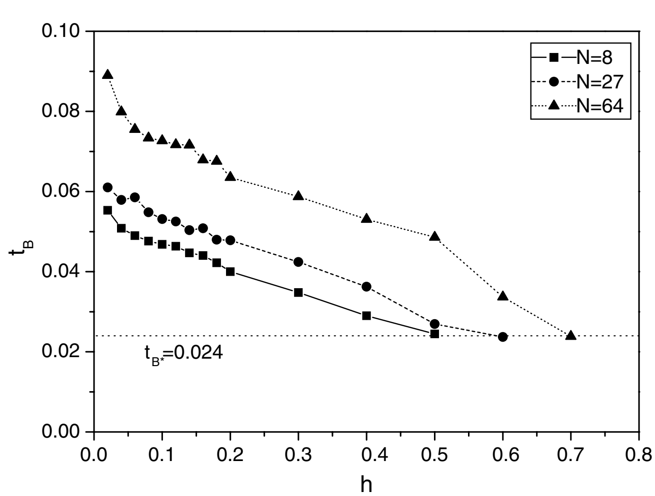

Fig. 1. Reduced blocking temperature versus the reduced magnetic field for different sample concentrations (N).

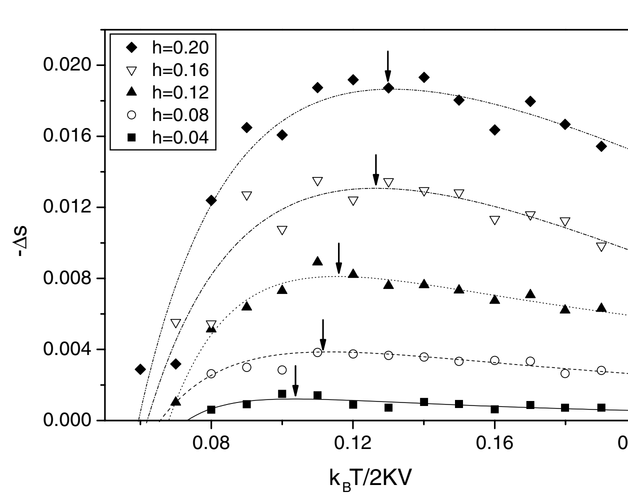

Fig. 2. Negative entropy change plotted against the reduced temperature for different values of the reduced magnetic field (h). The arrows indicate the maximum of the entropy curves.

4. Conclusions

When dealing with assemblies of fine magnetic particles the strength of the applied magnetic field is a main magnitude to evaluate. Above the blocking temperature, larger applied magnetic fields will lead to higher entropy changes, but the change will occur at higher temperatures for larger fields. Fitting the appropriate strength of the field would be a previous task for any magnetocaloric experiment. A possible guide could be the simulation of the better conditions using for example a Monte Carlo technique.

These calculations complement the preliminary ones started in Ref. [5] about the magnetocaloric effect for achieving the most appropriate conditions.

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