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Efficient Calculation of Low Energy Configurations of Nanoparticle Ensembles for Magnetoresistive Sensor Devices by Means of Stochastic Spin Dynamicsand Monte Carlo Methods

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We present results of Monte Carlo and stochastic spin dynamics simulations of a magnetic nanoparticle model system based on experimentally produced samples. Thermodynamic investigations as well as spin dynamics studies show characteristic features, both resembling magnetic dipole glass behaviour. While spin dynamics studies at $T = 0$ yield a multitude of low energy configurations, thermodynamic simulations show a clear transition between a paramagnetic and a frozen magnetic state. Moreover, we demonstrate the application of experimentally inspired demagnetization protocols to compute low energy configurations of the systems under consideration efficiently.

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1. Introduction

By dispersing magnetic nanoparticles in conductive gel-like matrices, magnetoresistive sensors with promising features can be developed. It has been shown [1] that in systems of commercially available cobalt nanoparticles and novel silica [2] or agarose gels, giant magnetoresistance effect amplitudes of more than 60% can be observed in transport measurements, yielding enhanced sensor sensitivity. The use of this technique opens up a wide field for applications because of the mechanical flexibility of gels compared to conventionally prepared sensor setups and reduced production costs.

In this paper we show that theoretical investigations of these magnetic nanoparticle ensembles reveal sophisticated magnetic behaviour as a result of dipolar coupling between the nanoparticles and spatial disorder reminiscent of magnetic dipole glass behaviour. We first present results from our stochastic spin dynamics simulations at $T = 0$ K that reveal a multitude of low energy configurations instead of a unique ground state. These low energy configurations can be obtained very efficiently by simulating experimentally inspired demagnetization protocols for artificial spin ice systems [3]. Furthermore, we show results of our Monte Carlo simulations that reveal a transition temperature of the temperature dependent susceptibility separating between a low temperature frozen magnetic state and a high temperature paramagnetic state.

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2. Model structure and simulation methods

For our investigations we have used a model system that resembles a nanoparticle arrangement which is typical for experimentally produced structures. This model system, named AH41, consists of 200 monodisperse cobalt nanoparticles with equal diameters of 20 nm, distributed on a substrate with a size of 350 nm $\times$ 350 nm as shown in Fig. 1. In our model calculations we assign to each nanoparticle a constant effective magnetic moment of $\mu_{\text{eff}} = 641371.18$ [μ_{B}], according to the following equation, with M_{sat}, the saturation magnetization in J T^{-1} m^{-3}, μ_{B}, the Bohr magneton in J T^{-1} and d, the particle's diameter in m:

$$\mu_{\text{eff}} = \frac{M_{\text{sat}} \pi d^3}{6\mu_B}. \tag{1}$$

Furthermore, due to the large distances between the particles we only take dipole–dipole interactions among particles into account. We have used a standard Monte Carlo Metropolis algorithm in order to investigate the thermodynamic properties [4]. Starting from random initial condition we have performed 5×10^7 discarded Monte Carlo steps (MCS) in order to relax the system. After that we have performed 5×10^4 MCS for producing thermal averages. In every MCS, the orientation of each magnetic moment is randomly changed. The total energies of the configurations before and after each change are compared. According to a certain probability the new configuration is accepted or rejected. For a detailed description of the algorithm see [4].

In order to study the spin dynamics of our model system we have used a stochastic spin dynamics algorithm based on a Langevin-approach where the heat bath

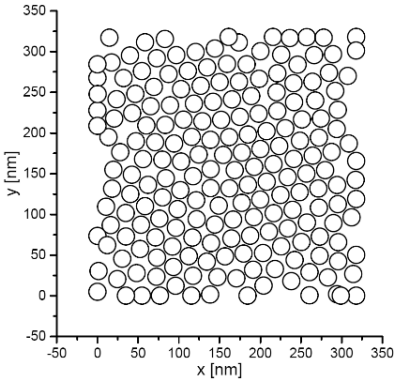

Fig. 1. Sample structure AH41 consisting of 200 cobalt particles.

contact is modelled by the competition of frictional and fluctuational fields [4].

3. Properties at $T = 0$ K

Using our stochastic spin dynamics method we have first performed simple relaxation simulations at $T = 0$ K starting from random initial configurations. This procedure is commonly used in order to find the ground state of a classical spin system. However, instead of a single ground state, we have found a multitude of low energy configurations which are apparently separated by energy barriers that cannot be overcome by the simple relaxation procedure. The differences of the total energies of these configurations lie within about 2% however the spin orientations are completely different as shown in Fig. 2.

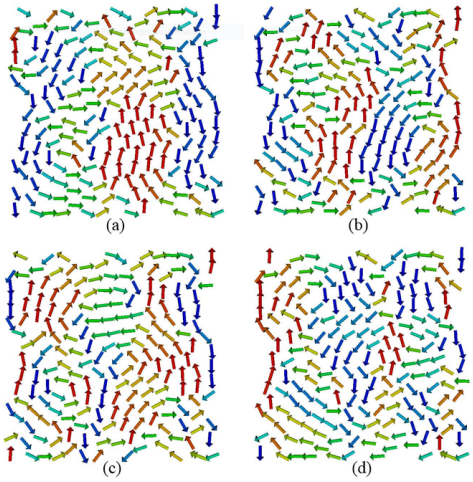

Fig. 2. Examples of low energy configurations of AH41 as obtained by stochastic spin dynamics simulations. Starting from random initial conditions the system is relaxed by setting $T = 0$ K. The coloring of the arrows represents the in-plane component of the magnetic moment. The resulting total energies are (a) -3.78467×10^{-16} eV, (b) -3.75635×10^{-16} eV, (c) -3.79583×10^{-16} eV and (d) -3.72457×10^{-16} eV.

While the preceding simulations show that numerous low energy configurations exist, it is not yet clear whether configurations of even lower total energy exist

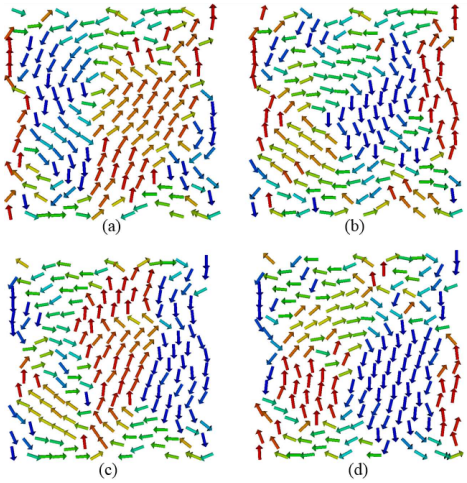

Fig. 3. Low energy configurations of AH41, computed via an experimentally inspired demagnetization process at $T = 0$ K. The coloring of the arrows indicates the in-plane component of the magnetic moment. The resulting total energies are (a) -3.82643×10^{-16} eV, (b) -3.83821×10^{-16} eV, (c) -3.81961×10^{-16} and (d) -3.83900×10^{-16} eV.

that just cannot be found by a simple relaxation procedure. Experimentally, demagnetization protocols have been used to drive magnetic ensembles into a low energy state. As reported in [3], such demagnetization routines have also been successfully applied numerically to search for lowest energy configurations of artificial spin ice systems. As a consequence of that, we have modified our relaxation algorithm by a demagnetization protocol which uses sinusoidally varying rotating and damped magnetic fields. Some results of our new relaxation procedure are shown in Fig. 3. In general, one observes lower total energies as compared to the simple relaxation procedure. Moreover, the differences of the total energies of these configurations lie within about 0.5% and therefore are much closer to each other in energy as compared to the configurations obtained by the simple relaxation algorithm. By comparison, larger vortices can be observed in the case of using a demagnetization protocol as compared to the case of the simple relaxation. A qualitative explanation of the advantage of the demagnetization protocol over the simple relaxation is the following. The simple relaxation simulations cannot change the relative orientation of closely placed nanoparticles even if changing their orientation would result in a lower total energy due to their strong dipolar interaction. By using the implemented demagnetization protocol, these strong couplings could partially be shifted to lower the total energy due to the agitation of the oscillating component of the applied magnetic field. As a result, low energy configurations of magnetic nanoparticle ensembles can be computed more efficiently using the above described demagnetization protocol.

4. Thermodynamic properties

We have performed Monte Carlo simulations in order to investigate the temperature dependent magnetic

susceptibility. For the sake of simplicity and for reducing computation time to a reasonable time span of approximately 2 h, we only used one section of the structure AH41 measuring 50 nm × 50 nm. The results are shown in Fig. 4.

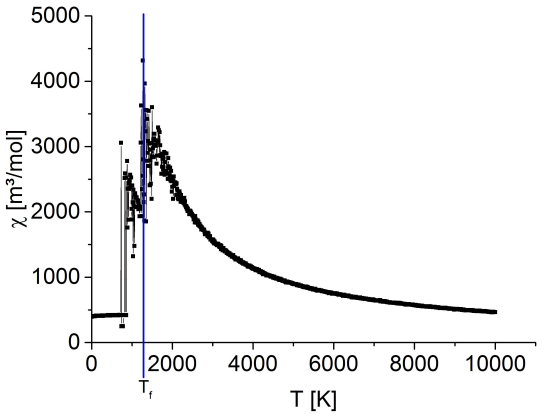

Fig. 4. Plot of the temperature-dependent magnetic susceptibility χ of one section of AH41 showing the transition between a frozen magnetic and a paramagnetic behavior at $T_f = 1650$ K. Furthermore, it is observable that within the frozen regime simulation times are way too small to establish ergodic behavior [5].

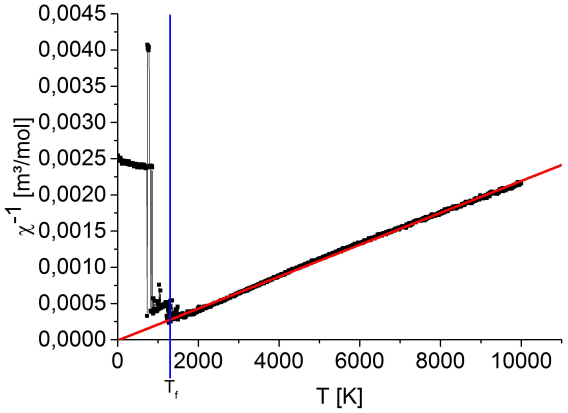

Fig. 5. Linear fit of the inverse susceptibility above the transition temperature clearly showing the paramagnetic regime above about 1650 K. The linear fit between the freezing temperature $T_f = 1650$ K and 10000 K results in a y-intercept of 1.00336×10^{-5} with a standard error of 1.82401×10^{-6}.

It is clearly visible that the statistical quality of the obtained data is changing dramatically at about $T_f = 1650$ K. Below T_f the Monte Carlo algorithm produces data of very poor statistical quality. Above T_f the quality is acceptable. The zero field susceptibility exhibits two local maxima within the temperature range. The second maximum at $T_f = 1650$ K indicates the transition temperature between the frozen magnetic state and the paramagnetic state of the system. Above T_f the system's frozen state disappears because thermal fluctuations dominate over the dipolar interactions. In this region one expects Curie's law for independent paramagnets to be valid. In fact, we show in Fig. 5 that the inverse susceptibility T_f shows strictly linear behaviour down to T_f and goes right through the origin. The reason for the poor statistics at low temperatures can be attributed to a dramatic increase in the relaxation times below T_f which is typical for spin glass systems [5]. Here, our simulation times turn out to be way too small to establish ergodic behaviour.

5. Conclusions

We have shown that a model system that resembles a nanoparticle arrangement which is typical for experimentally produced structures shows dipolar glass behaviour. By stochastic spin dynamics simulations we have found that a unique ground state cannot be identified, instead a multitude of low energy spin configurations exists of nearly the same energy. These spin configurations can be computed very efficiently by applying an experimentally inspired demagnetization protocol. In addition to that, the analogy to magnetic dipole glasses has been investigated by thermodynamic simulations. Here, we found a transition temperature in the temperature dependent susceptibility which is a distinctive feature of magnetic dipole glasses. Our simulations show paramagnetic behaviour above the transition temperature and freezing of the magnetic moments below the transition temperature. Future work includes the improvement of the demagnetization protocol by investigating different profiles. Furthermore, we will expand our spin dynamics simulation method by molecular dynamics methods in order to include the structuring process of magnetic nanoparticles in a surrounding medium due to interparticular interactions and interactions with external fields.

Acknowledgments

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