

Dipolar interactions and thermal stability of two-dimensional nanoparticle arrays

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Abstract

We show results of Monte Carlo simulations of an array of monodispersed magnetic monodomain particles, in a square lattice with dipolar interactions and perpendicular uniaxial anisotropy. We first show the equilibrium phase diagram of the system, which shows three phases, superparamagnetic (SP), out-of-plane antiferromagnetic and in-plane antiferromagnetic with a reorientation transition between the last two. We then employ a recently introduced time quantified Monte Carlo method to study the relaxation of autocorrelations of the particle array for different ratios of dipolar to anisotropy energies. In the non-interacting case we show that relaxation is exponential in time with characteristic times obeying a classic result by Brown. When dipolar interactions are switched on, the relaxation is very well described by stretched exponential forms in the whole time window and in both the SP and ordered phases. Relaxation times still obey a nearly Arrhenius behaviour, with a single effective energy barrier that decreases as the dipolar interaction increases, a result that must be interpreted within the dynamics protocol. No signs of glassy behaviour were found, in agreement with the absence of disorder in the model system.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

The development of advanced experimental tools to fabricate magnetic nano-structures in a controlled way has led to an enormous growth of studies concerning the physical understanding of nanomagnetism, such as the role of different interactions and scaling behaviour. Nanostructured magnetic materials like magnetic particles and patterned magnetic alloys are very important systems in experimental and theoretical research [1], beyond their important technological applications such as magnetic data storage and other fields [2, 3]. In general, magnetic media for data storage are composed of tiny but isolated magnetic nanocrystalline grains, but the need for increasing the memory densities forces the development of non-conventional media, such as patterned media and self-

assembled magnetic nanoparticles. The idea is to replace the randomly oriented magnetic grains with magnetic particles or nanodots in which it is possible to control the magnetic anisotropy orientation [2, 3] with the advantage of reducing the noise. Whatever the magnetic support, thermal stability is a key ingredient in magnetic media for data storage. Increasing the storage density implies a reduction in the size of the magnetic grains. If the particles or grains are too small they lose thermal stability reaching the so-called *superparamagnetic (SP) limit*. For single isolated magnetic nanoparticles, modelled as giant magnetic moments, the thermal stability problem has been extensively studied from the theoretical point of view [4–6], however, when the system is a set of interacting particles there are still many open questions [7–9]. In particular, long-range dipolar interactions are unavoidable, becoming relevant when increasing the packing density of the magnetic moments. In this sense, it is very important to understand the role of dipolar interactions

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in the relaxation dynamics of a set of magnetic moments due to thermal fluctuations. Patterned alloys [2] and two-dimensional self-assembled magnetic nanoparticle arrays are systems suitable for the experimental study of thermal stability, since it is possible to tailor the magnetic anisotropy orientation of the magnetic nanoparticles with respect to the array plane. In addition, other important properties like the shape and size of the magnetic units can also be controlled. A landmark of magnetic relaxation in particle arrays is the slow, quasi-logarithmic decay of the remanent magnetization over several decades in time. The origin of this slow relaxation is still controversial. Recent simulation studies in three dimensional systems point to the necessity of considering very simplified models, with minimum ingredients, as a way to understand the mechanisms responsible for the slow relaxation and glassiness in dipolarly coupled magnetic particles [10–14]. The structure of the crystal lattice (or the absence of structure), the nature of anisotropies (whether random or not), the presence of polydispersity in the particle volumes and the volume concentration of the array, all have to be carefully considered in order to better understand the origin of magnetic behaviour in a particular array.

The relaxation behaviour of two-dimensional arrays of monodispersed magnetic moments has been studied theoretically [8, 9, 15–18]. Mean field calculations [15] show that the relaxation may be slow without the need for introducing disorder, as commonly assumed. The mean field approach does not take into account correlations between particles due to dipolar interactions and rapidly fluctuating thermal fields [9]. In fact, when dipolar interactions are considered, there are ordered states at low temperatures. Taking as example an array of particles in a square lattice and depending on the ratio between the dipolar and anisotropy strength, the system can order antiferromagnetically in the plane or out of the plane [19, 20]. The complexity of the interacting problem requires the use of numerical tools in order to go beyond mean field results [17].

In this work we first show, by means of Monte Carlo simulations, the phase diagram of a system of monodispersed magnetic particles in a square lattice with perpendicular uniaxial anisotropy and dipolar interactions. With this information at hand, we then use a recently introduced time quantified Monte Carlo (TQMC) method to characterize the relaxation dynamics of the system. We characterize the time dependent relaxation near thermal equilibrium, which happens to be very well described by stretched exponential decays in the whole time window and in both ordered and disordered regions of the phase diagram. From the relaxation curves we go on to obtain the characteristic relaxation times and compare them with the predictions of the Brown–Arrhenius theory. In order to explore the influence of dipolar interactions on the effective free energy barriers that control the relaxation process, we explore a wide range of intensities of the dipolar interaction. This allows us to study the limits of validity of the Brown–Arrhenius model of relaxation and define the basic ingredients responsible for the observed slow dynamics.

2. Model and simulations

We consider a model of a two-dimensional monodispersed array of single domain magnetic nanoparticles arranged in a square lattice. The particles have uniaxial anisotropy which is oriented perpendicular to the x – y plane of the array. The classical Hamiltonian of this system can be written in the form

$$H = -D \sum_i S_{iz}^2 + g \sum_{i<j} \frac{\vec{S}_i \cdot \vec{S}_j - 3(\vec{S}_i \cdot \hat{e}_{ij})(\hat{e}_{ij} \cdot \vec{S}_j)}{r_{ij}^3}, \quad (1)$$

where $\vec{S}_i = \vec{\mu}_{np}^i / \mu_{np}$ are three-dimensional magnetic moments of unit length. μ_{np} is the value of the magnetic moment, which depends on the volume V and the magnetization M_s of the magnetic nanoparticle, $\mu_{np} = VM_s$. $\hat{e}_{ij} = \vec{r}_{ij} / r_{ij}$ are unit vectors on the plane of the array. The first term is the energy contribution of the uniaxial anisotropy pointing in the direction of the z axis. $D = K_u V$ with K_u the uniaxial anisotropy constant of the system under study. The second term in the Hamiltonian is the dipolar interaction between the magnetic particles, $g = \mu_0 \mu_{np}^2 / 4\pi a^3$ where μ_0 is the vacuum permeability and a is the lattice parameter. We consider the dipolar energy in a point dipole approximation [21]. In order to obtain the equilibrium phase diagram of the system we employ the Metropolis Monte Carlo algorithm where the spins are randomly updated in the unit sphere [22]. For the simulations of the relaxation dynamics we implemented a recently introduced time quantified Monte Carlo (TQMC) method [23–25] which allows direct comparison with experimental time scales. We considered the high damping limit in order to maximize the total time span of the simulation. The simulations were done for square lattices of linear dimensions $L = 16$ and $L = 32$. In order to minimize finite size effects due to the long ranged dipolar interactions, periodic boundary conditions were imposed by means of the Ewald method. Details of the implementation of the Ewald method can be found in [26]. Once the dipolar and anisotropy effective fields are calculated for each magnetic moment in the lattice, they are updated randomly using the TQMC [25].

3. Results

3.1. Phase diagram

Figure 1 shows the phase diagram of the system which was obtained and discussed in the context of the spin reorientation transition in [20]. Here we plot it in a way suitable for discussing the dynamical behaviour. All energy scales are in units of the Boltzmann constant, which was set to $k_B = 1$. The dashed line in the figure corresponds to the blocking temperature for our simulated time window, for the case of an array of non-interacting particles, i.e. $g/D = 0$. For high temperatures the particles are superparamagnetic (SP) phase. Upon lowering T the anisotropy and dipolar terms in the energy begin to rule the behaviour of the system, leading to two possible phases with antiferromagnetic order: for small temperatures and $g/D < 0.8$ the film orders out of plane in a checkerboard like configuration [27]. The perpendicular staggered magnetization shows a jump at the

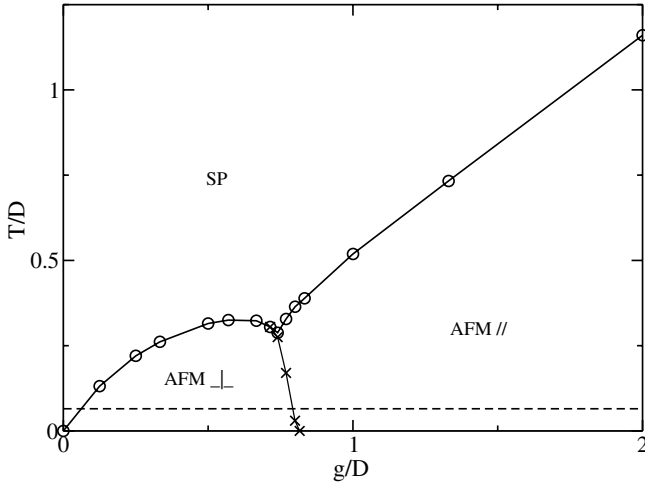


Figure 1. Phase diagram obtained from Monte Carlo simulations for a system size $L = 32$. The dashed line indicates the blocking temperature for the case of non-interacting particles.

transition temperature, suggesting that the transition to the perpendicular antiferromagnetic phase is first order. Of course, a precise determination of the order of the transition would only be possible considering much larger system sizes, which is beyond the scope of this work. Our results in this study all refer to this region of the phase diagram. For comparison, from an array of 4 nm Co particles we obtain $g/D = 0.67$ or $g/D = 0.084$, if the particles are in contact or are separated by a distance equal to the diameter, respectively. Here we use the Co parameters of [28]. With growing dipolar coupling, the system goes through a spin reorientation transition to an in-plane antiferromagnet as described in [20].

The knowledge of the different phases in an interacting system is essential to correctly interpret the relaxation behaviour which will be the subject of the following sections.

3.2. Near equilibrium relaxation

For the dynamical simulations we employed the recently introduced TQMC method [23–25] which allows one to map Monte Carlo time steps to real time scales for any values of the damping parameter usually introduced in micromagnetic simulations. This new TQMC is very efficient and much easier to implement and control than a micromagnetic simulation. The new approach is based on a controlled map between the stochastic Monte Carlo dynamics and the corresponding stochastic Fokker–Planck equation. By means of a detailed correspondence between the two approaches to the same stochastic dynamics, the real time scale (in units of a characteristic *damping time*) and MC time steps become related by

$$\Delta t[\tau_K] = \alpha \frac{R^2}{20} \Delta t[\text{MCS}], \quad (2)$$

where the damping time is given by

$$\tau_K = \frac{1}{\gamma H_k} \frac{(1+a^2)}{a}. \quad (3)$$

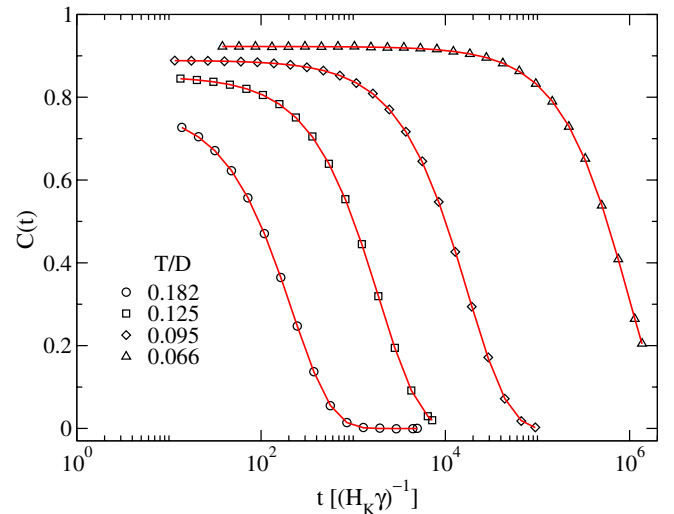


Figure 2. Autocorrelations for a system of noninteracting particles. Continuous lines are exponential fits.

In the previous relations $\alpha = D/T$, R is the size of a cone where the spin is updated, $H_k = 2K/M_s$, M_s is the saturation magnetization and a is the damping parameter. For details on the method and notation see [24, 25, 29]. In the present simulations $0.03 < R < 0.1$ in order to optimize the time span of the dynamics for the different temperatures.

In order to characterize the relaxation dynamics of the system as it approaches equilibrium, we measured time dependent autocorrelation functions by first relaxing the system for 10^5 MC steps from an initially disordered configuration of the spins. Then an initial configuration was stored $\{\vec{S}_i(0)\}$ and subsequent autocorrelations were defined as:

$$C(t) = \frac{1}{N} \sum_{i=1}^N \vec{S}_i(0) \cdot \vec{S}_i(t). \quad (4)$$

Statistical averages of this function for many realizations of initial conditions were done. Autocorrelations relax similarly to the magnetization and are related to the linear response function by the fluctuation–dissipation theorem.

Our reference system is an ensemble of non-interacting particles with uniaxial anisotropy. This corresponds to the vertical axis of figure 1. In figure 2 we show four correlation curves for this case, each curve corresponding to a low temperature in the high energy barrier regime, $D/T \gg 1$. The correlation curves show a rapid decay to a rather high value where a plateau is developed as the temperature is lowered. This regime corresponds to the relaxation inside the initial basin of the energy minimum near the initial condition. At long times the particles are able to overcome the anisotropy barriers and a final relaxation takes place where the particles decorrelate from their initial state. In this case of noninteracting particles the relaxations are exponential in time, as can be seen in the fits in figure 2:

$$C(t) = C_0 e^{-t/\tau}. \quad (5)$$

Note that the horizontal time scale is given in units of $(\gamma H_k)^{-1}$. Taking as reference cobalt with a high damping constant $a = 1$, then $\tau_K = 4.82 \times 10^{-11}$ s using $\gamma = 0.2212 \times 10^6$ m A $^{-1}$ s $^{-2}$

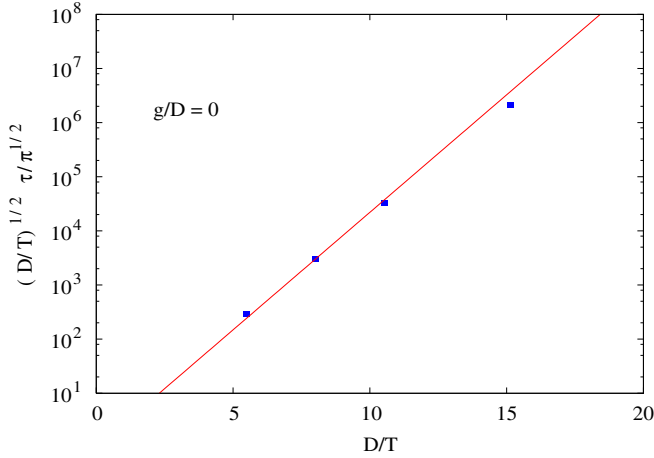


Figure 3. Relaxation times for a system of noninteracting particles. The solid line is the analytic approximation by Brown.

for the gyromagnetic ratio and $H_k = 168 \text{ kA m}^{-1}$ for the anisotropy field [28].

From the results for the correlation functions we can obtain the relaxation times for the particles. In a classic paper, Brown obtained approximate expressions for the relaxation time of single domain magnetic particles. In the case of zero applied field and high energy barriers the result of Brown is [4]

$$\frac{\tau}{\tau_K} \approx \frac{\sqrt{\pi}}{2} \alpha^{-1/2} e^\alpha. \quad (6)$$

In figure 3 we show the relaxation times obtained from the exponential fits of figure 2 together with the analytic approximation by Brown in a log–linear plot. The agreement is very good except for the point corresponding to the lowest temperature. This departure may be due to insufficient relaxation of the particles for this very high energy barrier. This explanation can be supported by calculating the *blocking temperature* for the ensemble of particles, defined as the temperature for which the experimental time scale equals the relaxation time. This T_B is usually calculated assuming an exponential or Arrhenius behaviour of the relaxation time. This is not completely true in this case, where the prefactor of the exponential in (6) depends on temperature. Nevertheless, a fit with a purely Arrhenius behaviour of the form $\tau = \tau_0 \exp(\Delta E/T)$ works reasonably well and allows a rough determination of the blocking temperature:

$$T_B = \frac{D\Delta E}{\ln(t_{\text{exp}}/\tau_0)}, \quad (7)$$

where in our simulations $t_{\text{exp}} \simeq 10^6$. From the fitting values of $\Delta E = 0.85$ and $\tau_0 = 2.27$, we obtain $T_B/D \approx 0.065$, which is near the value of the lowest temperature simulated. Then it is probable that the curve corresponding to $T = 0.066$ in figure 2 is still subject to considerable fluctuations.

Upon switching the dipolar interaction the main observation is that the relaxation is no longer exponential. In figure 4 we show correlation curves for a small intensity of the dipolar interactions $g/D = 0.0625$ and different temperatures,

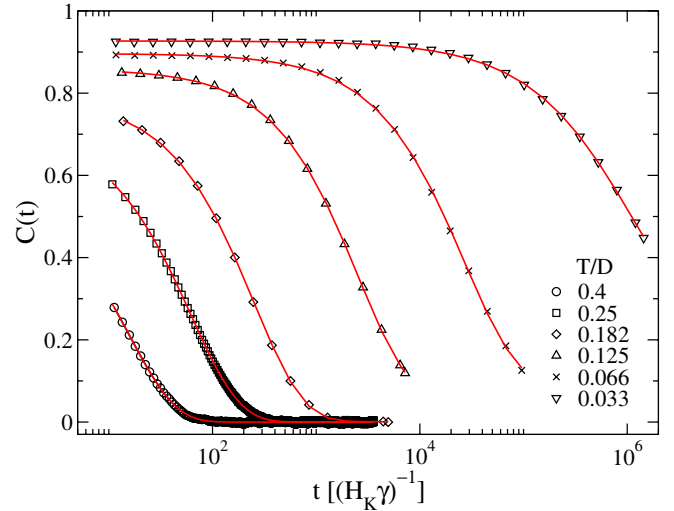


Figure 4. Correlations for $g/D = 0.0625$. Solid lines are fits to stretched exponential forms (see text).

all in the SP phase (see figure 1). Together with the data points, the solid lines represent fits with a *stretched exponential form*:

$$C(t) = C_0 e^{-(\frac{t}{\tau})^\beta}. \quad (8)$$

In this case of weak dipolar interaction the values of the exponent β are around 0.9. In fact we found that the typical β values diminish as the intensity of the dipolar coupling grows. Moreover, the stretched exponential form can describe the whole time span of the relaxation for any value of the dipolar intensity. Nevertheless, to our knowledge, there is no analytic prediction for this particular form of the time dependent relaxation in systems of interacting particles.

A quasi-logarithmic time decay is widely employed to fit relaxation data in systems with some kind of disorder. Nevertheless some time ago Dahlberg *et al* [30] showed that this slow relaxation need not be associated with disorder in the system and can as well be present due to the long ranged dipolar interactions. They verified this by solving numerically a mean field model of two state particles. More recently Denisov *et al* [9] obtained analytic expressions for the relaxation of the remanent magnetization in a 2D system of interacting dipoles with strong perpendicular anisotropy from an initially saturated state. They find a crossover from slow relaxation at intermediate times to an exponential one at very long times. Although the models are similar, a direct comparison of their results with ours is not straightforward due to the different protocols used in each case.

Stretched exponential relaxation is also observed in the low temperature regime of the present system, when it relaxes towards a state with long-range antiferromagnetic order out of the plane. In figure 5 we show autocorrelation curves for temperatures corresponding to the ordered phase. For this value of $g/D = 0.25$ the stretching exponent β is around 0.6, i.e. the relaxation is much slower than in the non-interacting case. Nevertheless, although not shown in the figure, the behaviour of relaxation times does not present any anomaly when crossing the phase transition temperature. Due to the first

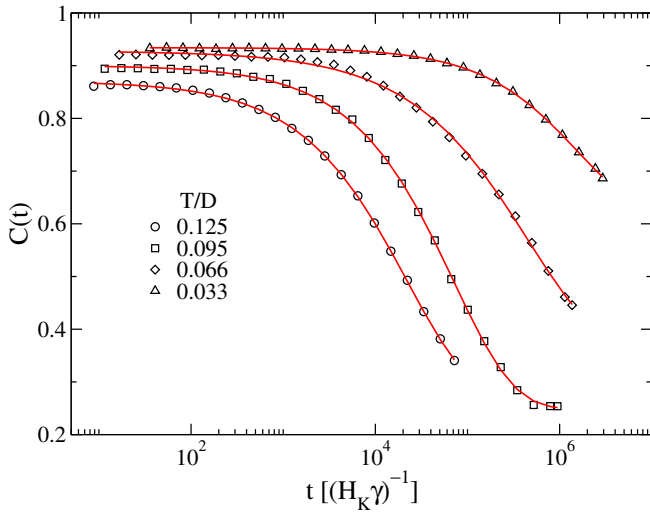


Figure 5. Correlations for $g/D = 0.25$. Solid lines are fits to stretched exponential forms (see text).

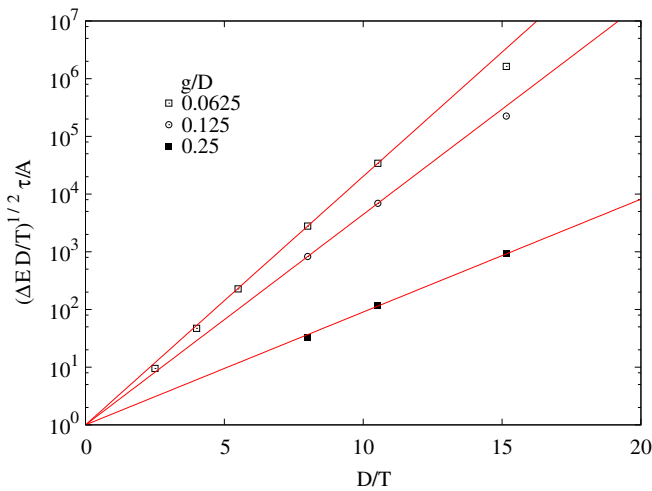


Figure 6. Relaxation times for a system with three different dipolar to anisotropy interaction ratios. The solid lines are fits with expression (9) as described in the text.

order nature of the transition (and the small size of the system) only a gradual change of stability between the disordered and the ordered phases takes place, with no evident effect on the typical relaxation times.

Together with the departure from exponential decay, when interactions are switched on the relaxation times also behave differently from the independent particle relation equation (6). In figure 6 the characteristic relaxation times corresponding to dipolar to anisotropy ratios $g/D = 0.0625, 0.125$ and 0.25 are shown. The values of τ were obtained from the corresponding stretched exponential fits and the data were fitted with the functional form:

$$\frac{\tau}{\tau_K} \approx \frac{A}{2} (\alpha \Delta E)^{-1/2} e^{\alpha \Delta E}, \quad (9)$$

which is a natural modification of Brown's relation (6), where now $D\Delta E$ represents an effective free energy barrier (we fixed $D = 1$ in the simulations whenever $g \neq 0$). Typical values of

ΔE are 0.99, 0.83 and 0.45 for $g/D = 0.0625, 0.125$ and 0.25 respectively. It was not possible to fit the data for the interacting systems fixing ΔE to one, as in the non-interacting case, even considering the exponent of the temperature dependent prefactor as a fitting parameter different from $1/2$. It is important to note that while the data for $g/D = 0.0625$ all correspond to the SP phase, those corresponding to the two larger values of dipolar interaction probe the system relaxing towards an ordered phase. In spite of this, it can be seen in figure 6 that in all cases the data are compatible with a single free energy barrier as represented by the nearly Arrhenius form (9). From this behaviour it can be concluded that glassy behaviour is not present in the dipolar interacting system, as this case should imply a non-Arrhenius dependence of relaxation times. Of course, some kind of disorder not considered in the present simulations, e.g. random anisotropies or polydispersity, may lead to spin glass like behaviour (see, for example, discussions in [11, 12] and references therein). Instead, we observe that dipolar interactions are responsible for an effective reduction of the free energy barriers which control the relaxation process, even with an initial state where demagnetization fields are significantly reduced. This energy barrier reduction is related to the growth of the ordered state as can be realized by looking at the order parameter evolution during the relaxation. It is worth stressing that the growth of the ordered phase does not affect the Arrhenius character of the relaxation process.

4. Conclusions

We have studied the relaxation dynamics of an array of dipolar interacting magnetic particles. Instead of looking at the remanent magnetization from a saturated state, we have first let the system relax near equilibrium and the subsequent relaxation was studied. Our model of a monodispersed array of single domain particles with uniform perpendicular anisotropy, while clearly simplified, is a first step towards a more systematic study of the origin of slow dynamics in realistic nanoparticles arrays. We obtained two main results: first, regarding the time dependence of relaxation functions, we found that while the relaxation is exponential in the non-interacting case, it changes to a slower relaxation form as the dipolar interaction is switched on, already for a very weak interaction strength. A stretched exponential form fits the data remarkably well over the whole time span of the relaxation, in contrast to the known limitations of a logarithmic fit [15, 31, 32]. While this may indicate a fundamental role of the stretched exponential form as a consequence of the microscopic interactions [10–12], this remains to be proved on a theoretical basis. We also found that the slow dynamics is present in the interacting system regardless of the temperature regime, while relaxing towards the SP or ordered phases. Second, we showed that the Brown–Arrhenius result for the temperature dependence of relaxation times works very well for independent particles, but needs to be modified when interactions are present. We proposed a slight modification of the original Brown's results in order to fit the data. Within this form, the behaviour of relaxation times shows that

effective free energy barriers are reduced with growing dipolar interactions, at least in this case where the system is initially already in a demagnetized state.

The TQMC method turned out to be a convenient and efficient way to perform simulations which can be directly confronted with experimental data. We used the example of cobalt nanoparticles in which we showed that realistic time scales can be reached within reasonable model parameters.

Monte Carlo simulations of three-dimensional arrays of particles, including both configurational and anisotropy disorder [10–14], have shown that anisotropy disorder smears out the effect of configurational disorder [12], and that this disorder is particularly important when the ground state is a columnar antiferromagnet [12, 13]. Moreover, glassy behaviour, present in these cases, seems to be enhanced in the configurationally ordered case [14]. Then, future studies of two-dimensional systems should add the effects of dipolar interactions with the influence of disorder on relaxation, present for example when the anisotropy axes are randomly oriented, which is the usual case in self-assembled or epitaxial nanoparticle systems [32–34]. For the important case of perpendicular anisotropy, relevant for data storage, disorder can also be present in the intensities of local anisotropies [35, 34]. Polydispersed assemblies with a distribution of grain volumes can also be straightforwardly considered within the present model [32, 33]. We plan to address these issues in future works, together with different dynamical protocols more directly related to experiments, such as for example, the behaviour of the thermoremanent magnetization in field cooled protocols and the influence of external fields after a zero field cooled process.

Acknowledgments

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