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Ordered systems of ultrafine ferromagnetic particles

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Abstract. A simple model for systems of dipolarly interacting single-domain ultrafine ferromagnetic particles is studied by Monte Carlo simulations. Zero field cooling and field cooling as well as relaxation experiments are used to compare systems with positional and orientational disorder to systems which are (i) positionally, (ii) orientationally, and (iii) positionally and orientationally ordered. It is shown that, as far as macroscopic observables are concerned, these partially [cases (i) and (ii)] or fully [case (iii)] ordered systems, despite quantitative differences, behave qualitatively very similar to the disordered one. This holds true even for the relaxation, where the decay of the magnetization $M(t)/M_{\rm S}$ (measured in units of the saturation magnetization $M_{\rm S}$) leads to an instantaneous relaxation rate $W(t) = -{\rm d}/{\rm d}t \ln[M(t)/M_{\rm S}]$ vanishing as a power-law as a function of time t, $W(t) \propto t^{-n}$. The exponent n is found to increase with increasing concentration, and becomes $n > 1$ for dense systems.

PACS. 75.50.Tt Fine-particle systems; nanocrystalline materials – 75.40.Mg Numerical simulation studies – 75.50.Lk Spin glasses and other random magnets

1 Introduction

It is generally observed that materials drastically change their macroscopic behavior if the size of the crystalline constituents becomes increasingly smaller. Many technological applications take advantage of the intriguing properties of such micro- or nanocrystalline materials, and the diverse magnetic materials consisting of ultrafine ferromagnetic particles are a very good example for this material class. As their magnetic properties can be widely tailored, for instance by changing the particles' intrinsic properties, their size distribution, the particles' positional and orientational arrangements, etc., these materials own applications in very widespread fields such as magnetic recording, permanent magnets, magnetic sensors, ferrofluids, pigments, refrigerant materials, but also in emerging fields such as quantum computation and as diagnosis and therapeutic tools in medicine (see Ref. [1] for recent comprehensive reviews).

For the same reasons that lead to their importance for real-world applications, systems of ultrafine ferromagnetic particles provide useful model systems to study quite fundamental questions of magnetism, such as the interplay of the magnetic dipolar interparticle interaction and the particles' positional and orientational arrangement. In fact, many of the intriguing features of ultrafine ferromagnetic particles, in particular those being important for the abovementioned applications, occur due to the presence of strong interparticle interactions. However, in difference to the dilute systems with vanishing interparticle interaction which are now well understood [2], the experimental results for such dense systems are still controversially discussed [3].

A particular fascinating yet quite puzzling problem in this respect are the spin-glass phases which have been reported for some systems, for example for dense samples of nanoparticles of γ-Fe_2O_3 [3(f)], ϵ-Fe_3N [3(l)], and amorphous $Fe_{1-x}C_x$ [3(m)], as well as in monolayer films of $Ni_{81}Fe_{19}$ permalloy nanoparticles [3(q)], but are absent in many others. It has been conjectured in reference [3(e)] that such cooperative behavior is due to the interplay between the random anisotropies and the dipolar interaction, whereas later experiments reported in reference [3(o)] seem to suggest that it is the particles' disordered spatial arrangement, cooperatively with the dipolar interaction, which dominate the behavior. First Monte Carlo (MC) simulations of a simple model including dipolar interaction indeed found collective phenomena at low temperatures being characteristic for spin glasses [4]. However, in later MC simulations spin-glass behavior was either not found at all [5] or only for model parameters not being realized in actual experiments [6]. These results have been questioned again by the very recent observation of slow relaxation in MC simulations [7].

One promising strategy to elucidate the behavior of dense systems of ultrafine ferromagnetic particles in general and the effect of the interparticle interaction in particular is to compare them to other related (preferentially better understood) systems. One effort in this direction

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has been made by the present author by studying the influence of various types of positional disorder [6] (with the objective of understanding the observations reported in Ref. [3(o)]) and the effect of particles' anisotropy [8]. Both studies intend to bridge the gap between the field of ultrafine ferromagnetic particles and the field of dipolar glasses [9], as the latter differ from the former mainly by the type of positional disorder and the absence of particles' anisotropy. This comparison has been quite fruitful, as it was observed in these MC simulations that, if the positional disorder becomes gradually similar to the one of dipolar glasses, the relaxation gets slower and slower until at some point a cooperative freezing at low temperatures occurs [6].

Another fascinating class of related systems are ordered nanocrystalline materials, so-called nanocrystalline superlattices. Since they were first reported 15 years ago [10], these intriguing systems have attracted considerable attention. Multi-component nanocrystalline assemblies (the ones being important for the present considerations) could usually be achieved only in an amorphous or short-range ordered state [10,11], but very recently, two-component nanocrystalline superlattices have been reported which are fully three-dimensional and long-range ordered, consisting of ferromagnetic nanoparticles (γ-Fe_2O_3) and semiconductor quantum dots (PbSe) [12]. Besides their importance for applications, it is an interesting fundamental question how these ordered systems of ultrafine ferromagnetic particles behave magnetically. (Single crystals of magnetic molecules have already been studied, showing quite interesting magnetic behavior, such as for instance a stretched exponential relaxation with a temperature dependent exponent [13].) Furthermore, these ordered systems provide another promising candidate for a comparison to their disordered counterparts. This comparison allows for a connection to the field of ordered dipolar systems, which one might consider as the ordered counterparts of the abovementioned dipolar glasses. Systems of ordered dipoles (without anisotropy) are known to have an antiferromagnetic ground state when the dipoles are located on a simple cubic (sc) lattice, and a ferromagnetic ground state for dipoles placed on a body-centered cubic (bcc) or a face-centered cubic (fcc) lattice [14]. So, the question is whether and how these antiferromagnetic or ferromagnetic ground states show up in ordered systems of ultrafine ferromagnetic particles.

To address these questions, ordered systems of ultrafine ferromagnetic systems are investigated by the simulation of zero field cooling (ZFC) and field cooling (FC) as well as by relaxation experiments. The central and somewhat surprising result of this study is that such ordered systems, despite quantitative differences, behave qualitatively very similar to their disordered counterparts. Besides the direct implications of this observation on ordered systems of ultrafine ferromagnetic particles, it is important to note that an analytical treatment of ordered systems might turn out to be feasible (in likely difference to disordered ones). Hence, the result of a qualitatively same behavior may, first, further motivate such effort and, second, allow to draw conclusion from a successful analytical treatment of ordered systems to disordered ones.

2 Model

Analogously to the model studied previously in references [5,6,8], it is assumed that every particle i consists of a single magnetic domain with all its atomic moments rotating coherently, resulting in a constant absolute value $|\boldsymbol{\mu}_i| = M_S V_i$ of its total magnetic moment $\boldsymbol{\mu}_i$. Here, V_i denotes the volume of particle i, and M_S is the saturation magnetization which is supposed to be independent of particle volume and temperature (the latter assumption is well obeyed for temperatures much below the Curie temperature). The energy contribution of each particle i to the system's total energy is composed of three parts: (i) the anisotropy energy $E_A^{(i)}$ (either caused by the particle's shape or crystalline structure), (ii) the energy $E_H^{(i)}$ arising from the applied magnetic field, and (iii) the energies $E_D^{(i,j)}$ due to dipolar interaction with particles $j \neq i$. To keep the model simple, a temperature independent uniaxial anisotropy is considered,

$$E_A^{(i)} = -KV_i \left[\frac{\boldsymbol{\mu}_i \cdot \mathbf{n}_i}{|\boldsymbol{\mu}_i|} \right]^2, \tag{1}$$

where K denotes the anisotropy constant and the unit vector $\mathbf{n}_i$ denotes the orientation of the easy axes of particle i (the somewhat more complicated case of cubic anisotropy has been studied for instance in Ref. [15]). The coupling to the applied field $\mathbf{H}$ is described by

$$E_H^{(i)} = -\boldsymbol{\mu}_i \cdot \mathbf{H}, \tag{2}$$

and the energy due to magnetic dipolar interaction between particles i and $j \neq i$ located at $\mathbf{r}_i$ and $\mathbf{r}_j$ is given by

$$E_D^{(i,j)} = \frac{\boldsymbol{\mu}_i \cdot \boldsymbol{\mu}_j}{r_{ij}^3} - \frac{3\ (\boldsymbol{\mu}_i \cdot \mathbf{r}_{ij})\ (\boldsymbol{\mu}_j \cdot \mathbf{r}_{ij})}{r_{ij}^5} \tag{3}$$

(an alternative MC approach including both dipolar and exchange interactions can be found in Ref. [16]). Thereby, the inter-particle distance is $\mathbf{r}_{ij} = \mathbf{r}_i - \mathbf{r}_j$ and $r_{ij} = |\mathbf{r}_{ij}|$. Adding up the three terms in equations (1–3) and summing over all particles, the system's total energy

$$E = \sum_i E_A^{(i)} + \sum_i E_H^{(i)} + \frac{1}{2} \sum_i \sum_{j \neq i} E_D^{(i,j)} \tag{4}$$

is obtained.

To allow for the comparison with hypothetical systems for which the particle anisotropy is absent, systems characterized by the total energy

$$E' = \sum_i E_H^{(i)} + \frac{1}{2} \sum_i \sum_{j \neq i} E_D^{(i,j)} \tag{5}$$

are studied in parallel. Although these hypothetical systems are found useful to separate the relative influence of

the particle anisotropy on the system's magnetic behavior and hence study a system without orientational disorder, it should be emphasized that these hypothetical systems are somewhat unphysical, as the original model's energy scale (which is $2KV$) is not present in equation (5).

For both systems characterized by either equation (4) or equation (5), the samples considered in the following consist of between $N = 108$ and $N = 216$ particles with identical volume $V_i = V$. The unitless concentration c is defined as the ratio between the total volume $\sum_i V_i = NV$ occupied by the particles and the sample volume V_{sample}, $c = \sum_i V_i / V_{\text{sample}} = NV/V_{\text{sample}}$. The concentration will be expressed in units of $c_0 = 2K/M_S^2$ being a unitless material constant of the order of unity [17]. The long-range dipolar interaction is treated with periodic boundary conditions using Ewald's summation for an infinite sphere surrounded by vacuum [18,19] (for an alternative MC approach based on effective field treatment see for example reference [20] and references therein).

To perform the main magnetic MC simulation, a set of particle positions $\{\mathbf{r}_i\}_{\mathcal{C}}$ and, for the systems with particle anisotropy, a set of easy axes' orientations $\{\mathbf{n}_i\}_{\mathcal{C}}$ are needed for each configuration $\mathcal{C}$. The particle positions $\{\mathbf{r}_i\}_{\mathcal{C}}$ for the cases of positionally ordered systems, where the particles are located on a sc, fcc, or bcc lattice, are straightforward and identical for all configurations $\mathcal{C}$. In the case of positionally disordered systems, to obtain the sets of particle positions $\{\mathbf{r}_i\}_{\mathcal{C}}$ in a well-defined and controllable way, a preceding positional MC simulation is performed before the main magnetic MC simulation [21]. During this preceding positional MC simulation, the particles can move freely in a cubic box of linear size $L^\star$ and interact only by a standard Lennard-Jones pair potential $v^\star_{\text{LJ}}(r^\star) = 4[(1/r^\star)^{12} - (1/r^\star)^6]$ with periodic boundary conditions (quantities with * indicate reduced variables of the preceding MC simulation). The reduced density is chosen as $\varrho^\star = N[L^\star]^{-3} = 0.85$, and the system is thermalized at the desired reduced temperature $T^\star$. To rely specifically on the Lennard-Jones system to produce the disordered particle positions is mainly motivated by the fact that it is well studied, a detailed discussion is for instance given in reference [19]. It is important to note that in the case under consideration the reduced temperature $T^\star$ is *not* a real physical temperature, but just provides a scalar parameter controlling the positional disorder [22]. A detailed discussion of the influence of the positional disorder controlled by the parameter $T^\star$ on the magnetic behavior can be found in reference [8], in the following the case $T^\star = 1$ of moderate positional disorder will be used as a reference for the positionally ordered cases.

After the particle positions $\{\mathbf{r}_i\}_{\mathcal{C}}$ for the current configuration $\mathcal{C}$ are either given by a lattice or have been provided by the preceding MC simulation, the respective orientations $\{\mathbf{n}_i\}_{\mathcal{C}}$ of the particles' easy axes are chosen for the systems with particle anisotropy. These orientations are either taken aligned with the field ($\mathbf{n}_i$ parallel to $\mathbf{H}$) and hence identical for all configurations $\mathcal{C}$, or are chosen randomly with uniform probability. Both the positions and the orientations remain constant during the following main magnetic MC simulation. It should be noted, as there are four different choices concerning the particle positions $\{\mathbf{r}_i\}_{\mathcal{C}}$ (disordered particles' positions with $T^\star = 1$ as well as particles' positions chosen as sc, bcc, and fcc lattice) and three different choices concerning the anisotropy (no anisotropy energy, as well as the orientations $\{\mathbf{n}_i\}_{\mathcal{C}}$ of the particles' easy axes being disordered or aligned with the applied field), there are twelve different combinations to be studied.

During the main MC simulation, the temperature T and the applied field H are changed as discussed below, and the magnetization M in the direction of the applied field is recorded in certain time intervals (for further details concerning the application of the Metropolis MC algorithm [21] to the present model see Refs. [5,6,8,15,23]). The whole procedure of choosing the particles' position, choosing the easy axes, and main MC simulation is repeated for each configuration $\mathcal{C}$, and an ensemble average is performed by averaging over 650 to 1250 independent configurations.

3 Results

The comparison between the twelve cases of positional and orientational order and disorder, respectively, is done by performing simulations of ZFC/FC experiments. During such an experiment, the system is first demagnetized at (super-)paramagnetic temperatures and cooled in zero external magnetic field, afterwards a small external field is applied, the system is heated until reaching (super-)paramagnetic temperatures again (yielding the ZFC magnetization) and then cooled again (yielding the FC magnetization). For the applied field $H/H_A = 0.1$ is used in the cases discussed below, where $H_A = 2K/M_S$ denotes the anisotropy field, and the system is cooled and heated using a constant rate of $k_B|\Delta T|/(2KV) = 0.01225$ every 8000 MC steps.

The comparison will be exemplified by a single moderate concentration $c/c_0 = 0.13$, but the behavior shown below is typical for the rather large range of concentrations for which the dipolar interaction and the anisotropy are both important. In the dilute limit $c \to 0$, the dipolar interaction and hence the particles' positional arrangement becomes irrelevant, and the system's behavior is solely governed by the anisotropy energy.

The first part of the study done focuses on the three cases of positionally disordered systems, either with randomly oriented easy axes, without anisotropy energy, or with aligned easy axes, see Figure 1. These three cases with $T^\star = 1$ will serve as reference for the remaining nine positionally ordered cases. As already discussed in reference [8], the blocking temperature T_B is drastically lowered in systems without anisotropy energy, equation (5), and hence the particles anisotropy, cooperatively with the dipolar interaction, is a major mechanism for the blocking of the magnetic moments for medium and large concentrations. This can also be seen in the partially (i.e., orientationally) ordered systems with aligned particles'

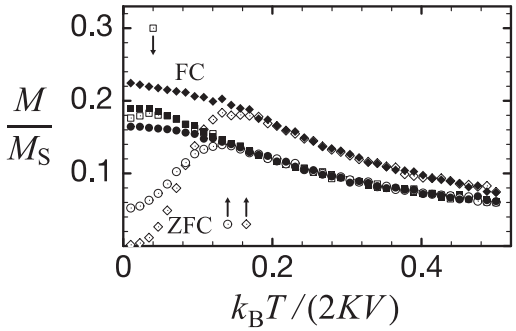

Fig. 1. Plot of the magnetization $M/M_{\rm S}$ vs. temperature $k_{\rm B}T/(2KV)$ obtained by ZFC/FC with a cooling/heating rate of $k_{\rm B}|\Delta T|/(2KV) = 0.01225$ every 8000 MC steps, for concentration $c/c_0 = 0.13$ and positional disorder $T^\star = 1$ ($N = 125$ particles). The open and full circles show the data for systems with randomly oriented easy axes, open and full squares are for systems without anisotropy energy, and open and full diamonds are for systems for which the easy axes are aligned with the field. The arrows indicate the respective blocking temperature $T_{\rm B}$, and the error bars are about the size of the symbols.

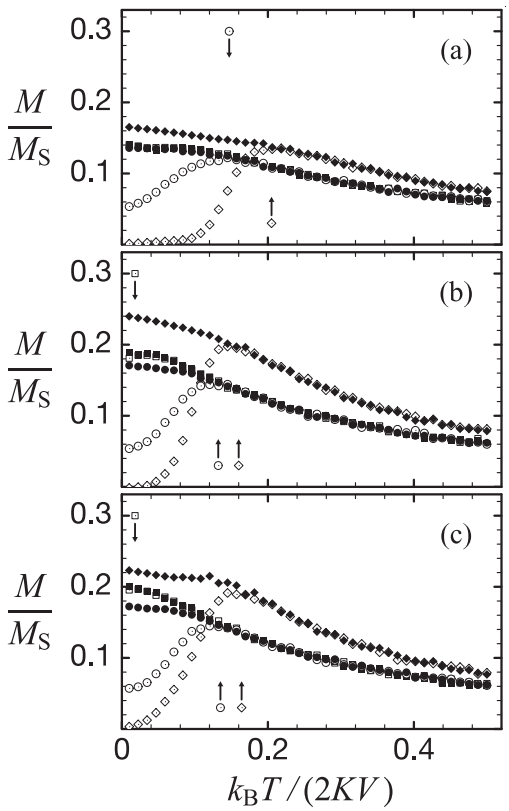

Fig. 2. Plot of the magnetization $M/M_{\rm S}$ vs. temperature $k_{\rm B}T/(2KV)$ obtained by ZFC/FC with a cooling/heating rate of $k_{\rm B}|\Delta T|/(2KV) = 0.01225$ every 8000 MC steps, for concentration $c/c_0 = 0.13$. The N particles are arranged on a (a) sc lattice ($N = 216$), (b) bcc lattice ($N = 128$), and (c) fcc lattice ($N = 108$). In all three cases, the open and full circles show the data for systems with randomly oriented easy axes, open and full squares are for systems without anisotropy energy, and open and full diamonds are for systems with easy axes aligned with the field. The arrows indicate the respective blocking temperature $T_{\rm B}$ (in (a), for the systems without anisotropy energy, $k_{\rm B}T_{\rm B}/(2KV) < 0.01$ and could not be obtained precisely) and the error bars are about the size of the symbols.

anisotropy axes, for which the blocking temperature $T_{\rm B}$ is significantly increased.

The central part of this study are the nine cases of positionally ordered systems, focusing on the sc, bcc, and fcc lattices, see Figure 2. For systems consisting solely of interacting dipoles, the case where the dipoles are located on a sc lattice has an 'columnar' antiferromagnetic ground state, whereas dipoles being located on bcc and fcc lattices have a ferromagnetic ground state [14]. This twofold behavior can be seen when comparing the data obtained for the sc lattice shown in Figure 2a with the data obtained for the bcc and fcc lattice shown in Figures 2b and c. The applied field $H/H_{\rm A} = 0.1$ is chosen to be rather large to ensure an accurate MC simulation, nevertheless the differences are quite obvious when looking on the cases of systems without anisotropy energy and of systems with aligned anisotropy axes, as both the blocking temperature and the low temperature FC magnetization differ by around 25%. The dichotomy is somewhat 'smeared out' by the orientational disorder, and the differences drop to below 10%. It is also interesting that for the case of the particles being located on a sc lattice, the field cooling curves for systems with disordered orientations of the anisotropy axes and for systems without anisotropy energy are essentially identical, indicating that the disordered orientations of the particles' easy axes average out for this positional arrangement. Note that for the case of particles being located on a sc lattice without anisotropy energy, the blocking temperature $k_{\rm B}T_{\rm B}/(2KV) < 0.01$ and could not be obtained precisely. In general one observes that (i) the blocking temperatures $T_{\rm B}$ for the case of orientational disorder is almost identical when comparing the cases of positionally disordered systems and the three cases of ordered systems, so that positional order or disorder has hardly any effect if orientational disorder is present. However, there is a quantitatively different behavior (ii) for systems without anisotropy energy and (iii) for systems with aligned anisotropy axes. The systems for which the particles are located on bcc and fcc lattices still behave similar to the positional disordered case, whereas the system of sc lattice behaves quite different: In the system without anisotropy energy, the blocking temperature $T_{\rm B}$ is decreased, whereas for aligned anisotropy axes a pronounced increase of the blocking temperature $T_{\rm B}$ is observed. As the system of particles located on a sc lattice with aligned anisotropy axes has a significantly larger blocking temperature $T_{\rm B}$, see Figure 2c, and is hence somewhat particular, it is suggestive to have a more detailed look on this specific case. In Figure 3, the data shown in Figure 2a for concentration $c/c_0 = 0.13$ is compared with two larger concentrations $c/c_0 = 0.195$ and $c/c_0 = 0.26$. It is observed that the blocking temperature increases in this concentration range roughly linear with the concentration.

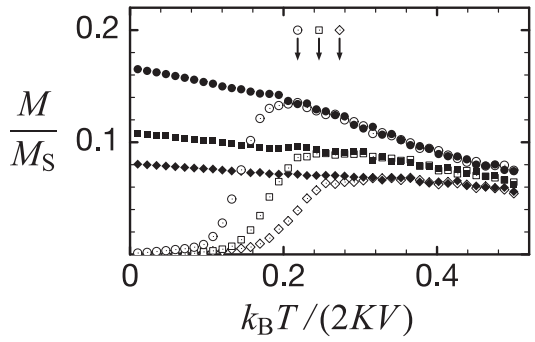

Fig. 3. Plot of the magnetization $M/M_{\rm S}$ vs. temperature $k_{\rm B}T/(2KV)$ obtained by ZFC/FC with a cooling/heating rate of $k_{\rm B}|\Delta T|/(2KV) = 0.01225$ every 8000 MC steps, for $N = 216$ particles arranged on a sc lattice with easy axes aligned with the field. The symbols refer to a concentration $c/c_0 = 0.13$ (open and full circles), $c/c_0 = 0.195$ (open and full squares), and $c/c_0 = 0.26$ (open and full diamonds). The arrows indicate the respective blocking temperature $T_{\rm B}$, and the error bars are about the size of the symbols.

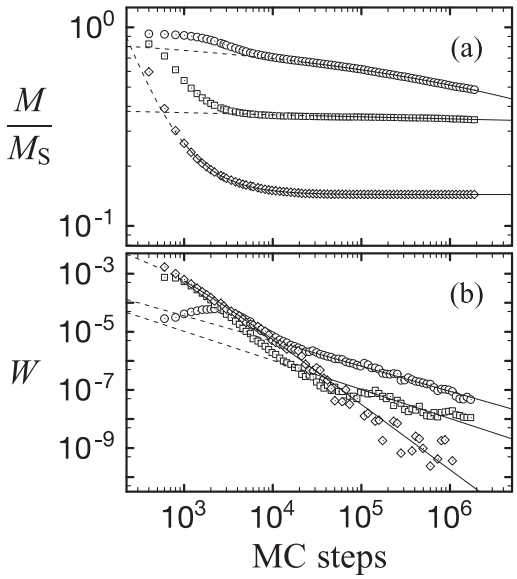

Fig. 4. (a) Plot of the magnetization $M/M_{\rm S}$ vs. time t (in number of MC steps) for $N = 216$ particles arranged on a sc lattice with easy axes aligned with the field. The symbols refer to concentration $c/c_0 = 0.13$ and temperature $k_{\rm B}T/(2KV) = 0.04435$ (open circles), $c/c_0 = 0.195$ and $k_{\rm B}T/(2KV) = 0.05$ (open squares), and $c/c_0 = 0.26$ and $k_{\rm B}T/(2KV) = 0.05543$ (open diamonds). The lines show fits of the form of equation (7) for $t > t_0$, with $t_0 = 2 \times 10^4$, $t_0 = 3 \times 10^4$, and $t_0 = 10^3$ (from top to bottom), as described in the main text (the dashed part is for $t \leq t_0$). The error bars are smaller than the size of the symbols. (b) Plot of the instantaneous relaxation rate W vs. time t (in number of MC steps), as obtained from the data shown in (a). The lines show the same fits as in (a) after being translated into instantaneous relaxation rates (the dashed part is for $t \leq t_0$).

The relaxation behavior for the three cases shown in Figure 3 is displayed in Figure 4a, where the magnetization $M/M_{\rm S}$ is shown as a function of time t on a log-log scale. The comparison is done for temperatures having a fixed ratio $T/T_{\rm B} = 0.203045$, where $T_{\rm B}$ is obtained from Figure 3. The setup is such that the systems are cooled in the saturated field $H/H_{\rm A} = 4$ from (super-)paramagnetic temperatures to the desired temperature, and the applied field is cut at $t = 0$, where the time t is measured in MC steps. One can clearly distinguish three different behaviors: The top curve for the smallest concentration $c/c_0 = 0.13$ shows a pronounced downward curvature, so that the magnetization decays faster than a power-law. The middle curve for the medium concentration $c/c_0 = 0.195$ shows a rather clear power-law decay, whereas the bottom curve for the largest concentration $c/c_0 = 0.26$ seems to approach a finite value with no further decay.

As there is no analytical form for the relaxation behavior known, many different functional forms have been suggested [24,25]. One way to analyze the three-fold behavior seen in Figure 4a more quantitatively is by looking on the instantaneous relaxation rate [7,25]. Denoting the magnetization at time t as $M(t)$, one defines the instantaneous relaxation rate $W(t)$ at time t as $W(t) = -{\rm d}/{\rm d}t \ln[M(t)/M_{\rm S}]$, so that one can write

$$\frac{M(t)}{M_{\rm S}} = \exp\left[-\int_0^t W(t')\,{\rm d}t'\right]. \qquad (6)$$

For disordered systems, one typically finds (true or effective) power-law decays for the instantaneous relaxation rate of the form $W(t) = W_0\, t^{-n}$ for large times t [26]. Under the assumption that $W(t)$ decays as a power-law for $t > t_0$, depending on the value of the exponent n, there are three cases to be distinguished for the behavior of $M(t)$, namely

$$\frac{M(t)}{M_{\rm S}} \simeq \begin{cases} \exp\left[-c_0\left(\dfrac{t}{t_0}\right)^{1-n}\right], & 0 \leq n < 1, \\ \left(\dfrac{t}{t_0}\right)^{-W_0}, & n = 1, \\ \exp(-c_0)\left[1 + c_0\left(\dfrac{t}{t_0}\right)^{1-n}\right], & n > 1, \end{cases} \qquad (7)$$

with the constant c_0 given by $c_0 = W_0\, t_0^{1-n}/|1-n|$ for $n \neq 1$. It should be noted that for $n > 1$, the magnetization in equation (7) approaches a finite value in the limit $t \to \infty$, $M_\infty = \lim_{t\to\infty} M(t) = M(t_0)\exp[-W_0\, t_0^{1-n}/|1-n|]$, and the system does not further relax. This is different from the cases $n \leq 1$, for which the magnetization eventually reaches the equilibrium value in the limit $t \to \infty$ (although the decay is very slow in the case $n = 1$).

In fact, the data shown in Figure 4a can be rather accurately fitted with equation (7), so that the instantaneous relaxation rate is very well described by a power-law after

some time t_0, even for this fully ordered system. For the case of the temperature at which relaxation takes place having a fixed ratio $T/T_B = 0.203045$, the system with the smallest concentration $c/c_0 = 0.13$ follows the first scenario with $0 \leq n \simeq 0.87 < 1$ for $t > t_0 = 2 \times 10^4$, the system with the medium concentration $c/c_0 = 0.195$ follows the second scenario with $n \simeq 1$ for $t > t_0 = 3 \times 10^4$, and the system with the largest concentration $c/c_0 = 0.26$ follows the third scenario with $n \simeq 2.25 > 1$ already for $t > t_0 = 10^3$. In Figure 4a, the three fits are shown as lines, which are drawn dashed below t_0. The numerically obtained instantaneous relaxation rates $W(t)$ are shown in Figure 4b, where the three different scenarios can be seen more directly, as $W(t)$ decays as $W(t) \propto t^{-0.87}$ for $c/c_0 = 0.13$, $W(t) \propto t^{-1}$ for $c/c_0 = 0.195$, and $W(t) \propto t^{-2.25}$ for $c/c_0 = 0.26$. The three fits of Figure 4a, after their translation into instantaneous relaxation rates, are shown as lines in Figure 4b, which are again drawn dashed below t_0. In the case of the largest concentration $c/c_0 = 0.26$, a finite value of the magnetization $M_\infty/M_S \simeq 0.144$ is obtained in the limit of $t \to \infty$. The present results allow an interesting comparison to a recent study [7], as the present systems are fully ordered, so that one would not expect the slow or vanishing relaxation being related to any glassy behavior. Therefore, an important conclusion of studying ordered systems of ultrafine ferromagnetic particles is that one might generally not take an instantaneous relaxation rate $W(t)$ decaying as $W(t) \propto t^{-n}$ with an exponent $n > 1$ as an indication of some type of glassy phase.

To investigate the system with the largest concentration $c/c_0 = 0.26$ in somewhat more detail and to see whether the observed vanishing relaxation is robust with respect to finite applied fields, the relaxation behavior in an applied field $H/H_A = 0.1$ is studied. Similarly as before, the system is cooled in an applied field from (super-)paramagnetic temperatures to the desired temperature, at which the field is changed to a finite value $H/H_A = 0.1$. Here, three cases are studied, namely cooling in the applied field $H/H_A = 0.1$ (so no change of the applied field is done at $t = 0$) and cooling in the saturated fields $H/H_A = 4$ and $H/H_A = -4$. As can be seen in Figure 5, these three initial conditions at $t = 0$ yield three distinct values of the magnetization. It is important to note that the instantaneous relaxation rates for the two cases of cooling in saturated fields decay as $W(t) \propto t^{-n}$ with $n > 1$, so that the magnetization will remain distinct in the limit $t \to \infty$. Hence, the observation of a slow or vanishing relaxation is robust even if the relaxation occurs in a finite applied field.

4 Conclusions

The central and somewhat surprising conclusion of this paper is that, as far as macroscopic observables are concerned, the cases of positional order are not that different from the positionally disordered case as long as orientational disorder is still present. The reason seems to be that the orientational disorder 'smears out' to a large extend the underlying positional order. Even if the system is fully ordered, the behavior is qualitatively still similar, despite quantitative differences. Probably the most interesting case studied is the system where the particles are located on a sc lattice and have the anisotropy axes aligned with the applied field, both from the point of view of addressing fundamental questions of magnetism such as relaxation behavior, as well as concerning possible applications. The latter, in particular, as the blocking temperature is in this case much larger than the one of the other (ordered or disordered) systems, for fixed concentration. However, the relaxation behavior is still quite similar to disordered systems, as the instantaneous relaxation rate $W(t) = -\mathrm{d}/\mathrm{d}t \ln[M(t)/M_S]$ decays as a power-law, $W(t) \propto t^{-n}$, for $t > t_0$. For the largest concentration $c/c_0 = 0.26$ studied, when the system is cooled in a saturated applied field and the field is cut at $t = 0$, $W(t) \propto t^{-2.25}$ is observed for $t > t_0 = 10^3$, so that there is a finite magnetization in the limit of $t \to \infty$. This effect is found to be robust even if the applied field is not cut to zero but reduced to a finite value. As these systems are fully ordered, one would not expect the slow or vanishing relaxation being related to any glassy behavior. Therefore, an important conclusion is that an instantaneous relaxation rate $W(t)$ decaying as $W(t) \propto t^{-n}$ with an exponent $n > 1$ might generally not be taken as an indication of some type of glassy phase.

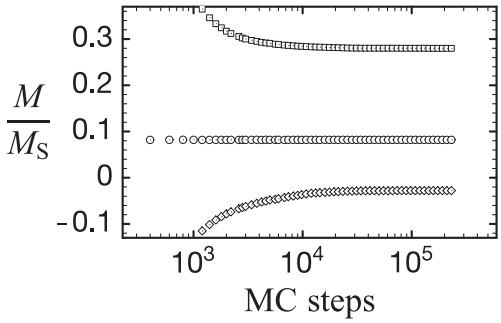

Fig. 5. Plot of the magnetization M/M_S vs. time t (in number of MC steps) for $N = 216$ particles arranged on a sc lattice with easy axes aligned with the field, concentration $c/c_0 = 0.26$, temperature $k_BT/(2KV) = 0.05543$, and applied field $H/H_A = 0.1$. The symbols refer to cooling in applied field $H/H_A = 0.1$ (open circles), and to cooling in saturated fields $H/H_A = 4$ (open squares) and $H/H_A = -4$ (open diamonds). The error bars are smaller than the size of the symbols.

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