Relaxation in ordered systems of ultrafine magnetic particles: effect of the exchange interaction

Stefanie Russ[1] and Armin Bunde[21, 2]

[1]1 *Institut für Theoretische Physik, Arnimallee 14, Freie Universität Berlin, 14195 Berlin, Germany*

[2]2 *Institut für Theoretische Physik III, Justus-Liebig-Universität Giessen, D-35392 Giessen, Germany*

We perform Monte Carlo simulations to study the relaxation of single-domain nanoparticles that are located on a simple cubic lattice with anisotropy axes pointing into the z-direction, under the combined influence of anisotropy energy, dipolar interaction and ferromagnetic interaction of strength J. We compare the results of classical Heisenberg systems with three-dimensional magnetic monments $\vec{\mu}_i$ to the ones of Ising systems and find that Heisenberg systems show a much richer and more complex dynamical behavior. Contrary to Heisenberg systems, Ising systems need large activation energies to turn a spin and also possess a smaller configuration space for the orientation of the $\vec{\mu}_i$. Accordingly, Heisenberg systems possess a whole landscape of different states with very close-lying energies, while Ising systems tend to get frozen in one random state far away from the groundstate. For Heisenberg systems, we identify two phase transitions, (i) at intermediate J between domain and layered states and (ii) at larger J between layered and ferromagnetic states. Between these two transitions, the layered states change their apparence and develop a sub-structure, where the orientation of the $\vec{\mu}_i$ in each layer depends on J, so that for each value of J, a new groundstate appears.

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INTRODUCTION

In the last decade, systems of ultrafine magnetic nanoparticles have received considerable interest, both due to their important technological applications (mainly in magnetic storage and recordings) and their rich and often unusual experimental behavior, which is related to their role as a complex mesoscopic system [1, 2]. The interest on the magnetic structure of these systems is twofold. The first question aims at the possible occurence of spin-glasses, as experiments on disordered magnetic materials present indications of a spin-glass phase or of a glassy-like random anisotropy system [2–5]. The second question aims at the occurence of superferromagnetic domains, since pronounced ferromagnetic states have been found in metal-insulator multilayers by ac susceptometry [2, 6], dc magnetometry [6], polarized neutron reflectivity and magnetometry studies [7] and X-ray and Kerr microscopy [8].

Ferromagnetic domains should not be expected in the simple dipolar model. Experimentally, the analysis of Mössbauer data on closely packed magnetic microcrystalline particles indicates that exchange interactions between different nanoparticles in close contact play an important role [9, 10] in the formation of ferromagnetic domains. Even in systems where the different nanoparticles are not in direct contact, an additional ferromagnetic exchange interaction is likely, which is mediated via tunnel exchange over small magnetic particles or clusters ("dark particles") in the surrounding matrix [2, 6–8]. However, also numerical relaxation simulations without exchange interaction showed final states characterized by a remanent magnetization in random [11] and in ordered systems [12] when starting from a ferromagnetic alignment.

For a realistic description of systems of ultrafine magnetic particles, it is important to include anisotropy energy, dipolar interaction and ferromagnetic exchange interactions as the relevant energy terms. Monte Carlo simulations incoporating these three energy contributions have been performed on disordered systems of ultrafine magnetic particles [13–15]. These works investigated, among others, hysterese effects, remanence and coercivity and found a quite complicated behavior based on frustration effects between the long-range dipolar interaction (favoring chains and closed-loop configurations [15] of the magnetic moments) and the short-range exchange interaction (favoring domains). Simulations incorporating only anisotropy and dipolar energy with [11, 16–18] and without [19] Ewald summation on ordered and disordered systems have been performed, regarding the question weather spin-glasses can be formed. Most works favorize a spinglass behavior or at least the occurence of ageing and memory effects [17–19], frustration [13–15] and frozen disorder [11, 17–19]. Other numerical works [20, 21] are dedicated to the competition between dipolar and exchange forces on spherical particles without the influence of the anisotropy energy. A third class of works consider Ising [22–24] or modified Ising systems [25], where only two stable positions of the magnetic moments are possible. A comparison between Heisenberg and Ising nanoparticles without dipolar interaction has been performed in [26]. For an overview, we refer to Ref. [27].

In this manuscript, we investigate the microscopic details of the occurence of frozen disorder as well as ferro-

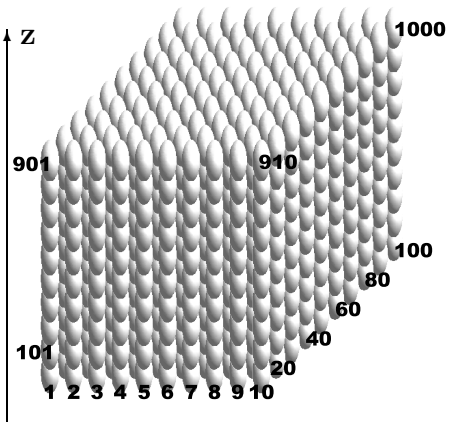

FIG. 1: Sketch of the nanoparticle system considered here, where the elongated particles are located at the sites of a simple cubic lattice with all anisotropy axes $\vec{n}_i$ (symmetry axes of the particles) pointing into the positive z- direction. The directions of the magnetic moments $\vec{\mu}_i$ that generally differ from the ones of the $\vec{n}_i$, are not shown. Note the way the sites are labeled. We will use this labeling in figure 8.

magnetic domains under the influence of local anisotropy energy, long-range dipole-dipole interaction and short-range ferromagnetic interaction between the particles. We concentrate on the most basic structure, where all three-dimensional magnetic moments $\vec{\mu}_i$ are located on the sites of a simple cubic lattice, and all anisotropy axes point into the z-direction (see figure 1), a system that nevertheless displays a large variety of complex configurations of the $\vec{\mu}_i$ and a rich relaxation dynamics. For the Ising case, the groundstates of these systems have already been studied with [23] and without [22] ferromagnetic interaction characterized by the coupling constant J. Depending on J, a scenario of very different groundstates has been found in [23], where the particle spins formed linear chains pointing into the positive or the negative z-direction. For the Heisenberg case, ordered systems have been investigated in [12, 17] for $J = 0$, but a systematic analysis for $J > 0$ has not yet been done.

Here, we consider the groundstates and the relaxation processes in Ising as well as in Heisenberg systems. Also in Heisenberg systems, quite stable chains are formed below a critical temperature that create effects of frozen disorder [17]. The groundstates for different values of J can be characterized by different arrangements of the chains. While in the "columnar antiferromagnetic state" (CAF), neighboring chains always point into the opposite direction, in the ferromagnetic state (F) all chains point into the same direction and in several kinds of layered structures the chains form layers of equally oriented $\vec{\mu}_i$.

Beside these similarities, we also find important differences between the Ising and the Heisenberg systems. First, due the two degrees of freedom of the magnetic moments $\vec{\mu}_i$ (characterized in spherical coordinates, i.e. by the angles θ_i and φ_i) and the therefore much larger configurational space of the Heisenberg systems, several configurations may possess energies very close to e.g. the CAF and the F configuration (see below). In the Ising system, in contrast, the energies of the different configurations are well distinct, and a very large activation energy is needed to break a chain (i.e. to turn one single particle inside an intact chain). As a consequence, for $J > 0$, relaxation only occurs at relatively high temperatures and freezing effects are quite dominant, an effect that was not visible in numerical works without dipolar interaction [26].

The comparatively complex relaxation behavior of the Heisenberg systems that we find here can be summarized as follows: Under the influence of the dipole-dipole interaction, neighboring chains prefer to point into opposite directions, which leads without additional ferromagnetic interactions to antiferromagnetic domains. In the presence of a small ferromagnetic interaction with J exceeding a certain limit, the chainlike structures of the dipoles are maintained, but neighboring chains have a preference to point into the same direction. Now, ferromagnetic domains occur which increase their size with increasing interaction strength. We believe that this situation is close to the experimental one. When the ferromagnetic interaction strength is further increased, several ordered states appear that have the shape of layers, with the thickness of the layers depending on the system size (see below). The layered state of deepest energy consists of two ferromagnetic domains, each with half of the system size and with one layer roughly parallel ($\theta \approx 0$), the other roughly antiparallel ($\theta \approx \pi$) to the anisotropy axis. When the interaction increases further, this Ising-like behavior stops and the layers develop substructures, i.e. sub-layers with the thickness of the lattice constant, where the angles θ_i of the magnetic moments take many values between 0 and π but are roughly constant throughout one sub-layer. At even higher values of J, the ferromagnetic state becomes the groundstate. In Ising systems, this scenario is not possible. Moreover, freezing effects occur at larger values of J that suppress the relaxation behavior.

The paper is organized as follows: In section II, we describe our model systems and the numerical simulations. In section III, we calculate the energies of several selected configurations of the $\vec{\mu}_i$ for Ising and Heisenberg systems as a function of J and search for the groundstates. In sections IV and V, we investigate the microscopical details of the relaxation process and the intermediate states through which the relaxation evolves. Finally, in the Conclusion section VI, we discuss the results.

MODEL SYSTEM AND SIMULATIONS

For the numerical simulations [28], we use the same model as in Refs. [11, 12, 16, 17]. Every particle i is

considered to be a single magnetic domain with all its atomic magnetic moments rotating coherently, resulting in a three-dimensional magnetic moment $\vec{\mu}_i$ of the particle. Here, all particles are of equal volume V, resulting in a constant absolute value $|\mu_i| \equiv \mu = M_s V$ of all $\vec{\mu}_i$, where M_s is the saturation magnetization. The energy of each particle consists of three contributions: anisotropy energy, dipolar interaction energy and ferromagnetic exchange energy. We assume a temperature independent uniaxial anisotropy energy $E_A^{(i)} = -KV((\mu_i \vec{n}_i)/|\vec{\mu}_i|)^2$, where K is the anisotropy constant and the unit vectors $\vec{n}_i$ denote the easy directions. As usual, the energy of the magnetic dipolar interaction between two particles i and j separated by $\vec{r}_{ij}$ is given by $E_D^{(i,j)} = (\vec{\mu}_i\vec{\mu}_j)/r_{ij}^3 - 3(\vec{\mu}_i\vec{r}_{ij})(\vec{\mu}_j\vec{r}_{ij})/r_{ij}^5$. While these two contributions are already explained in [17], the exchange interaction between site i and j is proportional to their magnetic moments $\vec{\mu}_i$ and $\vec{\mu}_j$, $E_J^{(i,j)} = -j_{ij}(a)\vec{\mu}_i\vec{\mu}_j$ with the coupling constant $j_{ij}(a)$ that varies with the distance between them and thus with the lattice constant a. For simplicity, we concentrate on samples of $N = (L/a)^3$ particles (with $L = 10\,a$) placed on the lattice points of a simple cubic lattice with periodic boundary conditions and with all anisotropy axes $\vec{n}_i$ pointing into the positive z- direction (see figure 1). The unitless concentration c is defined as the ratio between the total volume NV occupied by the particles and the volume $L^3 = Na^3$ of the sample. Comparing different system sizes, we focus on the fixed concentration $c/c_0 = 0.4$, where $c_0 = 2K/M_s^2$ (with the anisotropy constant K) is a dimensionless material-dependent constant with $c_0 \sim 1.4$ for iron nitride and $c_0 \sim 2.1$ for maghemite nanoparticles (see also [11, 17]), so that our value of c/c_0 results in a large but not unrealistic filling factor c for ellipoidal particles. We can express all energy terms by the same unit energy E_0 by introducing the direction vector $\vec{e}_i \equiv \vec{\mu}_i/|\vec{\mu}_i|$ of the magnetic moments and with $j_{ij}(a)\vec{\mu}_i\vec{\mu}_j = j_{ij}(a)\mu^2\vec{e}_i\vec{e}_j = (j_{ij}(a)a^3c/c_0)(c_0M_s^2V)\,\vec{e}_i\vec{e}_j$. With the distance vector $\vec{r}_{ij}$ between the sites i and j, the normalized distance vector $\vec{\ell}_{ij} = \vec{r}_{ij}/a$ and $\ell_{ij} = |\vec{\ell}_{ij}|$, $r_{ij} = |\vec{r}_{ij}|$, we add the three energy contributions by summing over all N particles. Expressing all constants by the unit energy $E_0 = c_0M_s^2V = 2KV$, we obtain

$$\varepsilon \equiv \frac{E}{E_0} = \frac{c}{c_0}\frac{1}{2}\sum_{i,j\neq i}^{N}\left(\frac{\vec{e}_i\vec{e}_j}{\ell_{ij}^3} - 3\frac{(\vec{e}_i\vec{\ell}_{ij})(\vec{e}_j\vec{l}_{ij})}{\ell_{ij}^5}\right) - \\ - \frac{1}{2}\sum_{i}^{N}(\vec{e}_i\vec{n}_i)^2 - \frac{j_{ij}(a)a^3}{2}\frac{c}{c_0}\sum_{i}^{N}\sum_{k=1}^{6}\vec{e}_i\vec{e}_{i+k}, \quad (1)$$

where the 1st, 2nd and 3rd term represent the dipolar, anistropy and exchange energy, respectively and the summation index k runs over the nearest neighbors of i. In this work, we express the ferromagnetic exchange interaction by the dimensionless prefactor $J \equiv j_{ij}(a)a^3c/c_0$ that we choose as a constant value for nearest neighbors ($r_{ij} = a$) and as $J = 0$ for larger interparticle distances. Accordingly, as compared to other works, e.g. to [23], the prefactor J of this work is multiplied with the (constant) factor c/c_0 here that determines the relation of the dipole as well as the exchange interaction as compared to the anisotropy energy.

The orientation of the individual magnetic moments $\vec{\mu}_i$ is described by their two spherical coordinates θ_i and φ_i in the Heisenberg case and $\theta_i \in \{0, \pi\}$ in the Ising case. For a better comparison between Ising and Heisenberg systems, we use the energy scale of (1) for both. This means that also for Ising systems, we add the anisotropy energy term, which is, of course, only a constant term and therefore meaningless in the calculations. Our energy scale can be easily transferred to the one of Ref. [23], where the energies have been calculated in units of μ^2/a^3, which can be expressed in our units by $\mu^2/a^3 = E_0c/c_0$.

To simulate the relaxation process as a function of t (number of Monte Carlo (MC) steps) [26, 29], we use the standard Metropolis algorithm in combination with the Ewald sum method [16, 17, 30] that carefully takes the long-range nature of the dipolar interaction into account. As in earlier papers [17, 18], the temperature is expressed in the units of the reduced temperature $\tilde{T} \equiv k_BT/(2KV) = k_BT/E_0$. As the jumps for the orientations of the $\vec{\mu}_i$ are of different sizes in the Ising and the Heisenberg case [29], the time scales cannot be directly compared. In the Ising systems, one step refers to an (attempted) jump of value π, while in Heisenberg systems, the jumps are much smaller. Therefore, the "real" time should be larger in the Ising systems.

GROUND-STATE SCENARIOS

Ising spins on different kinds of ordered cubic systems have been analyzed by several authors and for several types of interactions between the spins. While Luttinger and Tisza [22] investigated pure dipolar systems by renormalization and group theory and found that the CAF state was the state with lowest energy, Kretschmer and Binder [23] extended these calculations to an additional exchange interaction $J \neq 0$ by mean-field theory and Monte Carlo simulations with Ewald techniques. They found that depending on J, a whole scenario of different groundstates appeared. For very small (positive) J, the columnar antiferromagnetic state (CAF) has the smallest energy, while for slightly larger values of J, layered states of varying thickness Δ, where the spins are arranged in planes parallel to the xz- or the yz-plane, showed lower energies than the CAF state. In the layered Ising states, all spins of one layer are oriented into the same direction, while neighboring layers have opposite spins. In the following, we will call a layered state, where all layers have the same thickness $\Delta = na$, a "layer-n" state (L-n). Additionally, in [23], many states, where the chains of up-

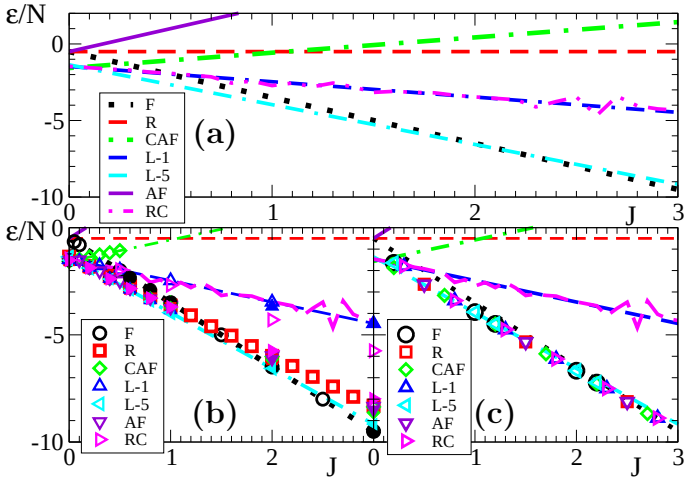

FIG. 2: (a) The (normalized) energy per spin ε/N (see Eq. (1)) is plotted versus the exchange interaction J for several configurations as specified in the legend and for systems of size $L = 10$. In (b) and (c), the symbols indicate the states reached after 10^4 MC steps when starting in these configurations for (b) Ising and (c) Heisenberg systems. Open and filled symbols stand for the reduced temperatures $\tilde{T} = 1/40$ and 1/1.25, respectively. The succession of the (colored) lines at $J = 3$ from top to bottom is: AF, CAF, R, L-1/RC, L-5, F.

and down-spins seemed to be randomly arranged showed energies close to the groundstate, so that the system normally did not relax to the groundstate but became frozen in one of these random states.

In this work, we are basically interested in Heisenberg systems, where each magnetic moment $\vec{\mu}_i$ possesses two degrees of freedom. This leads together with the smaller activation energy to a much more complex dynamics than in Ising systems, where all spins are either parallel (+) or antiparallel (-) to the z-axis. However, some similarities between Heisenberg and Ising systems exist. For example, also in Heisenberg systems, magnetic moments with all anisotropy axes aligned tend to arrange themselves in linear chains, where all dipoles inside one chain tend to be oriented either roughly parallel or roughly antiparallel to the anisotropy axes. Here, "roughly (anti-)parallel" means that the z-component of all $\vec{\mu}_i$ of a given chain is (negative) positive, but the opening angles θ_i are somewhat larger than 0 or smaller than π, respectively. This has been shown in [17] for $J = 0$ and of course, the chains become even stronger for $J > 0$.

To get a first impression about the possible dynamics and the differences between Ising and Heisenberg systems, we calculated their energies for several selected configurations and for systems of size $L = 10$. For simplicity, we denote the $\vec{\mu}_i$ also in the Heisenberg systems as "plus" (+) or "minus" (-) moments, when their z-component is parallel or antiparallel to the z-axis. The possible configurations can be described as follows: ferromagnetic state F (all $\vec{\mu}_i$ are (+) moments), columnar antiferromagnetic state CAF (see above), antiferromagnetic state AF (each (+) moment surrounded by (-) moments and vice-versa), layered states L-1 and L-5 (see above), random state R (all $\vec{\mu}_i$ completely random) and the state of random chains RC (all $\vec{\mu}_i$ arranged in chains into the z-direction with a random configuration between the chains). Figure 2(a) shows the energies per particle ε/N as a function of J, when all $\vec{\mu}_i$ are exactly in the corresponding Ising configuration (all $\theta_i \in \{0, \pi\}$). For the different ordered configurations (F, AF, CAF), we found exactly the same values as published in [23] (plus the constant anisotropy energy term), whereas for the configuration of random chains, the energy is quite high and corresponds to the energy of L-1 (same number of (+) and (-) neighbor chains). This shows that in the corresponding states of [23] of very small energies, the chains were obviously not really randomly arranged, but formed domains (see also [17]). In our units and in full agreement with [23], we find that among all investigated states, the CAF state is deepest for $J < 0.051$, while beyond this J-value, the layer-1 state has a smaller energy. In [23], layer-2 and layer-4 states were investigated for systems of sizes $L = 8$ that do not lead to periodic states in our $L = 10$ systems under periodic boundary conditions. Therefore, we find the next transition between the layer-1 and the layer-5 state (where the thickness of one layer corresponds to half the system size) at $J = 0.065$. For $J > 2.16$, the ferromagnetic configuration becomes groundstate. For comparison, we also tested systems of size $L = 6$ and $L = 8$ and found the considered energies independent of system size. The layer-2 or layer-3 state, that are not possible for $L = 10$ (under periodic boundary conditions) also show up in our calculations for systems of appropriate sizes $L = 8$ or $L = 6$, respectively.

Next, we want to see, which energies are reached after many Monte Carlo steps, when starting in the above configurations. The results are shown by the symbols in figure 2(b) and (c) for the Ising and the Heisenberg systems, respectively. (The straight lines are the same as in figure 2(a) and show the energies of the selected states as a guideline.) In the Ising systems, for some temperatures and some values of J, the systems stay frozen in their start configuration, while this is not the case for the Heisenberg systems (except, of course, when starting in the groundstate). If relaxation takes place, the energies are lowered. In the Heisenberg systems, even for small temperatures, the systems relax until the groundstate energy is reached. Moreover, independentely from the start configuration, all final energies fall onto the same universal curve, raising the important question, whether the systems always relax to the same configuration. We will see in the following sections that this is not the case and that also in the Heisenberg systems, domain states are quite often reached. However, due to the much larger configurational space, the energies of all these configura-

tions are very close to each other. This means that we have a continuum of quasi-degenerated states with nearly identical energies but clearly distinct orientations of their magnetic moments. This scenario is one prominent feature of a spinglass.

Ising systems, on the other hand, due to their smaller configurational space show a simple freezing behavior that is different from the frozen disorder of the close-lying energies in Heisenberg systems. For large values of J, Ising systems are often frozen, while lower-lying states are existing but unavailable because of the large activation energy for a spin flip. For small values of J, as we will see in the next section, Ising systems can still relax on very large time scales (beyond the 10^4 MC steps shown in figure 2) to a more favorable state.

RELAXATION PROCESS AND FINAL STATES: VISUALIZATION

We now want to take a closer look on the relaxation process. We are interested in the ("final") states that are reached after many MC steps, and in the intermediate states through which the relaxation evolves. We first investigate the regime of small exchange interaction J, where the ferromagnetic state is not the groundstate, i.e. for $J \ll 2.15$. We start in the ferromagnetic configuration and investigate the states that are reached after 10^5 MC steps. Now, we are interested in the configuration pattern, and not in the energy.

As the magnetic moments μ_i are arranged in linear chains, oriented parallel or antiparallel to the z-axis, we follow the lines of Ref. [17] and map the system onto the xy-plane, where each of the L^2 sites stands for one specific chain. To visualize the relaxation process in Figs. 3 and 4, we call a site in the xy-plane a "+" site or a "-" site, when the z-components of *all* magnetic moments in the chain are either positive or negative, respectively. Otherwise, we call it a "0" site (gray shade).

First, in Figs. 3 and 4, we illustrate the relaxation process on several examples for different values of J, separately for Ising and for Heisenberg systems. The examples are drawn at very different temperatures in the Ising and the Heisenberg case and therefore only serve as a qualitative picture. Together with the energy calculations of the preceeding section, the observations can be summarized as follows: Ising systems (figure 3), at small temperatures $\tilde{T}$ and for intermediate and large J, simply stay in the ferromagnetic state and therefore need quite high temperatures for a relaxation process to occur in the presence of a ferromagnetic interaction. Here, we have chosen $\tilde{T} = 1/2$, where chains can still survive [17], but Ising systems are not frozen. At this temperature, Ising systems quite often either reach their groundstate (i.e. the CAF state for $J = 0$ and the appropriate layered state at $J = 0.1$ and 0.4) or still change their apparence

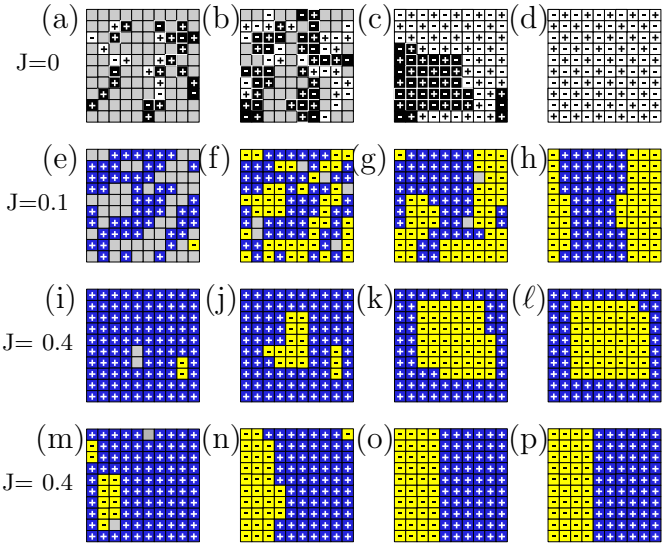

FIG. 3: Ising systems: Visualization (in the xy-plane) of representative relaxation processes from an initial ferromagnetic configuration at $t = 0$, for systems of length $L = 10a$ at temperature $\tilde{T} = 1/2$, after certain times t (number of MC steps). (a-d) $J = 0$ and $t = 2$, 20, $t = 10^4$ and $t = 10^5$, (e-h) $J = 0.1$ and $t = 10$, 20, $t = 10^4$ and $t = 10^5$ (i-ℓ) $J = 0.4$ and $t = 60$, 200, $t = 10^4$ and $t = 10^5$ and (m-p) $J = 0.4$ and $t = 200$, 2000, $t = 10^4$ and $t = 10^5$. The complete chains are indicated by "+" or "-" signs, depending on the direction of the chain. Antiferromagnetic domains (in the first row) are shown in black and white, ferromagnetic domains (in the following rows) in blue and yellow. Domains of the same color fit to each other and are allowed to emerge, whereas domain walls exist between clusters of different colors. Sites where chains have been destroyed are indicated by the gray shade.

between the 10^4th and the 10^5th MC step so that further relaxation (beyond the scope of our calculations) is very likely. Heisenberg systems, on the other hand do not seem to get frozen in some start configuration, as far as we can tell from our simulations, but find even at small $\tilde{T}$ a path to relax towards some state with an energy close to the minimum energy. However, this final state is normally not the groundstate, but either some domain state or an irregular layered state. No further chain flips were observed between the 10^4th and the 10^5th MC step, even if the final states often were very irregular, see e.g. figure 4(h). We chose $\tilde{T} = 1/20$ for the Heisenberg systems, but checked their behavior for $\tilde{T} = 1/2$ as well, to allow for a direct comparison with the Ising systems. The details can be described as follows:

In the absence of a ferromagnetic interaction ($J = 0$), (Figs. 3(a-d) and 4(a-d)), the CAF state is groundstate. Accordingly, in both cases, two kinds of antiferromagnetic domains are formed during the first 10^4 MC steps. First, in the very beginning of the process, a large fraction of the original chains are broken (i.e. many $\vec{\mu}_i$ change their direction from (+) to (-)), until an irregular pat-

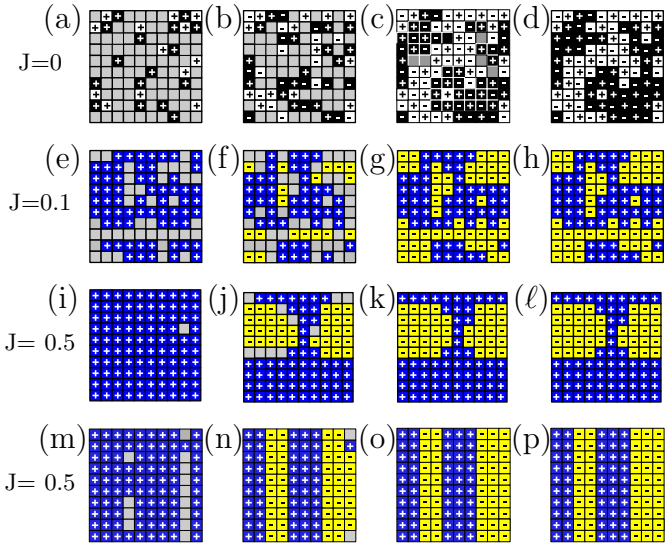

FIG. 4: Heisenberg systems: Visualization of the relaxation processes (as in figure 3) from an initial ferromagnetic configuration, for $\tilde{T} = 1/20$. (a-d) $J = 0$ and $t = 80$, 400, $t = 10^4$ and $t = 10^5$, (e-h) $J = 0.1$ and $t = 100$, 200, $t = 10^4$ and $t = 10^5$, (i-ℓ) $J = 0.5$ and $t = 200$, 400, $t = 10^4$ and $t = 10^5$ and (m-p) $J = 0.5$ and $t = 200$, 400, $t = 10^4$ and $t = 10^5$. For colors and symbols see explanations in figure 3.

tern of quite stable (+) chains has survived. Then, antiferromagnetic domains grow around these "seed" chains and many of the broken chains are now rebuilt as (-) chains, while only few of them turn back into (+) chains. When the domains grow and different domains contact each other, they either emerge and form larger domains (if they fit to each other) or establish quite stable domain walls between them. In the figures, both types of domains are shown in black and white, respectively, such that domains of the same shade fit to each other. In the Ising case, due to the quite high temperature, chains still change their direction beyond the 10^4th MC step until only one CAF domain remains. (For $\tilde{T} = 1/20$, on the other hand, a similar picture as in the Heisenberg case evolves.) In Heisenberg systems, the irregular two-domain patterns stay unchanged between the 10^4th and the 10^5th MC step for most of the temperatures. For comparision, we also tested systems at much higher temperatures in the Heisenberg case. For $\tilde{T} = 1/2$, no stable pattern arises, i.e. the chains are permanently broken and rebuilt. Already for slightly smaller temperatures, as e.g. $\tilde{T} = 1/3$ or $1/5$, the irregular two-domain patterns become stable in most systems (with some broken chains, i.e. some gray sites along the domain walls and few systems achieving the CAF state at $\tilde{T} = 1/3$). Of course, we cannot totally exclude that on much larger time scales beyond the means of current computer simulations, also in the Heisenberg case whole chains might change their direction. However, as we have already seen that these irregular states possess energies more or less identical to the minimum energy, further relaxation is not likely.

In the presence of a weak ferromagnetic interaction, $J = 0.1$ (Figs. 3(e-h) and 4(e-h)), layered states have lower energy than the CAF state. Now, less chains are broken at the beginning of the relaxation process and turned into (-) chains. Ferromagnetic (-) domains grow around these chains in a second time regime, thereby eventually destroying further chains. The ferromagnetic (+) and (-) domains are symbolized by the blue and the yellow colors of the sites. At the end, ferromagnetic domains are being formed, which can have the shape of layers. Especially in Ising systems at the high temperatures considered here, layered states are quite often reached. When the ferromagnetic interaction is increased to $J = 0.4 - 0.5$ (see Figs. 3(i-p) and 4(i-p), this picture does not change qualitatively, but the domains become more compact and in Heisenberg systems, the layered states become more frequent (sometimes still disordered, i.e. with irregular thickness of the single layers). Again, the Ising systems often change their pattern between the 10^4th and the 10^5th MC step, while this is not the case for the Heisenberg systems, where the pattern is already fixed after some 100 MC steps and further relaxation only takes place by adjustments of the single θ_i and φ_i, but not by turning (+) into (-) chains and vice versa.

RELAXATION PROCESS AND FINAL STATES: QUANTITATIVE DESCRIPTION

To see how often a given state occurs, we investigate $N_{\rm tot} = 100$ different relaxation processes for many $J \geq 0.1$ and count how many end in each of the different configurations after 10^5 MC steps. The system size is still fixed to $L = 10$. To distinguish between the different formations, we go back to the description in the xy-plane. We first define an intact row as a line into the x- or the y-direction, where all chains point into the same (positive or negative) z- direction. A state is called a layered state L, when (within a small tolerance level) either in x- or in y- direction all rows are intact. When all rows into the x- and the y-direction are simultaneously intact, we call the state a ferromagnetic state F and if a state is neither ferromagnetic nor layered, we call it a domain state D (where, for simplicity, we do not distinguish between ferro- and antiferromagnetic domains). We calculate the ratios $N_F/N_{\rm tot}$, $N_D/N_{\rm tot}$ and $N_L/N_{\rm tot}$ between the numbers N_F, N_D, N_L of F, D and L states, respectively and the total number $N_{\rm tot}$ of investigated states. The results are shown in Figs. 5 and 6 for the Ising and the Heisenberg systems, respectively, for start in (a) the ferromagnetic and (b) the random configuration. We show the relative number of systems that relax towards a layered state (blue squares), a domain state (green cir-

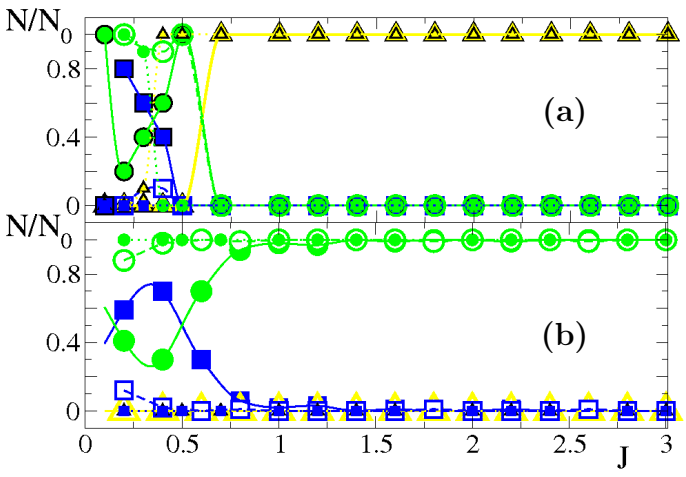

FIG. 5: Ising systems: Fraction of states in the ferromagnetic (yellow triangles), layered (blue squares) and mixed domain state (green circles) after 10^5 MC steps, when the initial configuration is (a) ferromagnetic or (b) random. The reduced temperatures are $\tilde{T} = 1/1.5$ (large filled symbols), $\tilde{T} = 1/2$ (open symbols) and 1/5 (small filled symbols).

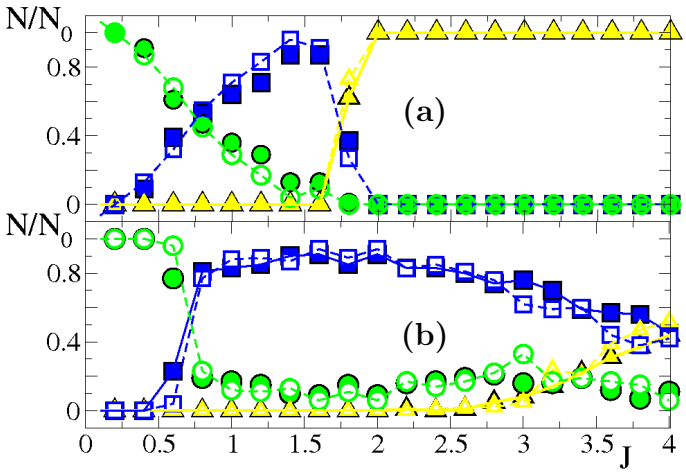

FIG. 6: Heisenberg systems: Fraction of states in the ferromagnetic (yellow triangles), layered (blue squares) and in the mixed domain state (green circles) after 10^5 MC steps, when the initial configuration is (a) ferromagnetic or (b) random. The symbols are the same as in figure 5 and the reduced temperatures are $\tilde{T} = 1/5$ (large filled symbols) and $\tilde{T} = 1/20$ (open symbols).

cles) or the ferromagnetic state (yellow triangles) as a function of J for several temperatures. Remember that among the states of section III, for $J < 0.0654$, the CAF state, for $0.0654 < J < 2.16$, the layer-5 state and for $J > 2.16$, the ferromagnetic state is the state of lowest energy. Again, a very different behavior occurs for the Ising and the Heisenberg systems:

In the Heisenberg systems, a temperature-independent phase transition takes place for $J = J_{c_1}$ between the domain states and the layered states, where exactly at J_{c_1} half of the systems end in the domain state and half of the systems in layered configurations. For $J < J_{c_1}$, domain states are dominant, while layered states are more frequent for $J > J_{c_1}$. J_{c_1} depends slightly on the start configuration, i.e. $J_{c_1} \approx 0.75$ when starting in the ferromagnetic state and slightly smaller when starting in the random configuration. Ferromagnetic (final) states do not occur in the regime of small J. (For a discussion of the regime of larger J see below). In the Ising systems, in contrast, depending on the start configuration, either domain or ferromagnetic states are dominating, while layered states – even if energetically favorable – are negligable except for very small J. The behavior of the Ising systems looks quite unsystematic and does not lead to a well-defined phase transition that is quite obviously supressed by trivial freezing effects. Especially for start in the ferromagnetic configuration, the system simply stays there for $J \gtrapprox 0.5$ (yellow triangles in figure 5(a)), while for start in a random configuration, frozen domain states dominate. By comparison with figure 2, we can see that these frozen domain states (red squares in figure 2) possess energies that are indeed clearly higher than the groundstate energies. Quite obviously, even if present, states of lower energy are unreachable because of the high activation energy of the spin flips, so that the final configuration of the $\vec{\mu}_i$ is quite arbitrary.

Therefore, in the following, we concentrate on Heisenberg systems. It is likely that the different behavior found in Figs. 5 and 6 is due to a different microscopic behavior and that many intermediate orientations of the magnetic moments relative to their easy axes play a strong role in Heisenberg systems, not only for the energies of the final states but also for the relaxation process itself. Therefore, for a more microscopic description, we now investigate the time development of the distribution $P(\theta, t)$ of the angles θ_i between the z-axis and $\vec{\mu}_i$, when starting in the ferromagnetic configuration. Figure 7 shows $P(\theta, t)$ at several times t (number of MC steps) for the reduced temperature $\tilde{T} = 1/20$ for several values of J between 0 and 1. Again, we can observe the two time regimes of figure 4 and now take a closer look at their microscopic details (left and right columns of figure 7).

In the first regime (Figs. 7(a,c,e)), $P(\theta, t)$ shows one single maximum that moves towards larger values of θ with increasing time and broadens. Accordingly, the magnetic moments $\vec{\mu}_i$ relax in a quite collective fashion towards the xy-plane (away from their anisotropy axes $\vec{n}_i$) with some of them changing their orientation. Obviously, the relaxation is governed by the dipole-dipole interaction and therefore neighboring chains do not stay in parallel orientations, which leads to a weakening and even breaking of most of the chains along the z-direction (see also figure 4). Clearly, the ferromagnetic interaction

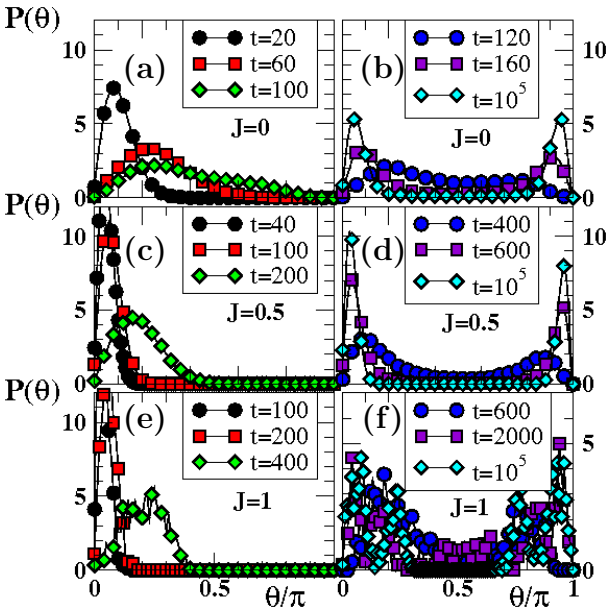

FIG. 7: Heisenberg systems: Distribution $P(\theta)$ of the angle θ_i between $\vec{\mu}_i$ and the z-axis, averaged over 100 systems of size $N = 10^3$ for $\tilde{T} = 1/20$ and (a, b) $J = 0$, (c, d) $J = 0.5$ and (e,f) $J = 1$ for several t (number of MC steps) as indicated in the legend during the 1st and the 2nd regime of the relaxation process (1st and 2nd column, respectively).

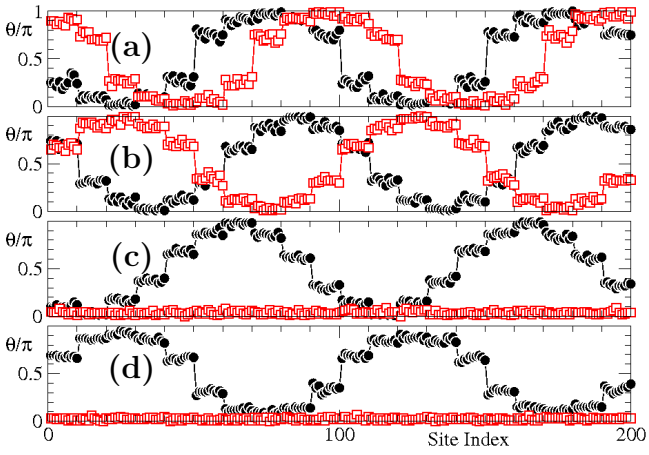

FIG. 8: Heisenberg systems: The normalized values of the angles θ_i of the $\vec{\mu}_i$ are plotted for several typical configurations after 10^5 MC steps versus the site index i in the order as shown in figure 1 for the first 2 planes of the cube. The values are shown for (a) $J = 1.2$, (b) $J = 1.5$, (c) $J = 2$ and (d) $J = 3$ and for start in (i) a random configuration (black filled circles) and (ii) the ferromagnetic configuration (red open squares). The arising pattern is repeated for each plane (therefore, the other 8 planes are omitted). The shift between the red and the black curves is meaningless because of the periodic boundary conditions.

slows this process down, i.e. the respective relaxation times increase considerably with J. At the end of this 1st relaxation process, the systems are balanced with respect to the dipolar interaction, but unbalanced with respect to the anisotropy energy that favors a parallel or an antiparallel alignment of the dipoles with respect to the anisotropy axes (that point into the z-direction for the systems considered here). Therefore, at the beginning of the second relaxation process, the anisotropy energy starts to govern the relaxation and causes the dipoles to align again in chains that can point either into the positive or into the negative z-direction. As a result, for not too large J, the single maximum splits into two maxima (Figs. 7(b,d)). With increasing time, both maxima separate from each other moving towards $\theta = 0 + \delta$ and $\theta = \pi - \delta$, where δ decreases with T and approaches zero in the limit of $T \to 0$. During this second time regime, the additional ferromagnetic interaction favors a parallel instead of an antiparallel alignment of the chains. Especially for small and intermediate values of J, where the spin flips occur very fast and randomly, this leads to the observed disordered domain structure, where in response to the orientation at $t = 0$, the positive domains are slightly larger (note the different height of the peaks in figure 7(d)). For larger J, the relaxation process is slowed down and less chains are destroyed during the first time regime and randomly rebuilt, leading to more ordered structures, i.e. to a dominance of the layered structures.

A new behavior arises for $J \geq 1$, as we can see from Figs. 7(e,f). Due to the larger contribution of the ferromagnetic interaction energy (compared with the anisotropy energy), many different values of θ are found in the final states of the histogram. Already for $J = 1$, we can see a double-structure of the maximum during the first time regime in figure 7(e). In the second time regime of figure 7(f), many different peaks appear in the histogram, i.e. the condition of an (anti)-parallel orientation of the $\vec{\mu}_i$ relative to the z-axis is now relieved. Obviously, the anisotropy energy term is compensated by the larger value of the ferromagnetic interaction, so that the larger configurational space of the $\vec{\mu}_i$ becomes relevant not only for the relaxation process but also for the final states.

In figure 8, we plot the angles θ_i for some typical systems after 10^5 MC steps versus the site index i (defined in figure 1), for $J = 1.2$, 1.5, 2, and 3. The black filled circles refer to a random and the red open squares to a ferromagnetic start configuration. As the curves are periodic and repeat after 100 sites (one layer in the xy-plane), we only show the first two planes, i.e. from site index $i = 1$ to $i = 200$. The figure shows a new kind of layered state that can be described as a "modified layer-5 state". Contrary to the layered states at small J, the $\vec{\mu}_i$ are now arranged in planes, where the inclination angles between the planes can accept many different values (not only 0 and π as in the case of small J). Accordingly, a new sub-structure of many planes of thickness $\Delta = a$ appears. For different values of J, we find different values

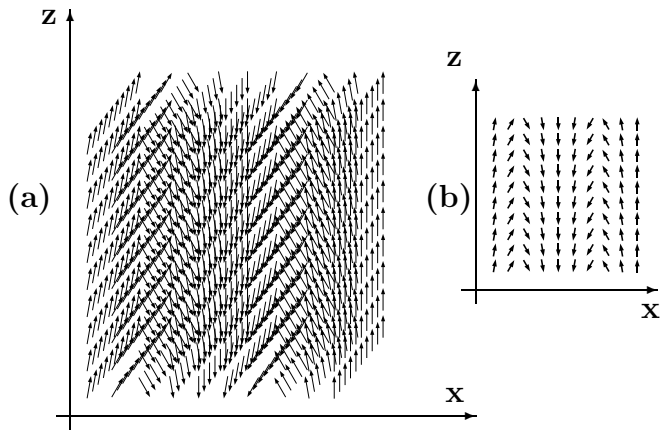

FIG. 9: Sketch of the $\vec{\mu}_i$, when the system (for the geometry of the particles see figure 1) is in the "modified layer-5" state, i.e. when the $\vec{\mu}_i$ are arranged in planes of similar moments (here in yz-planes) and the inclination angles of the individual planes accept many different moments. In (a), the three-dimensional structure of the system is shown and in (b) the two-dimensional front side (1st chain of each plane).

of the inclination angles and thus for the values of θ, i.e., the shape of the groundstate depends on J. A sketch of a typical modified layer-5 is shown in figure 9

A new phase transition to ferromagnetic states occurs at still larger values of J, as we have already seen in figure 6. The transition point between layered and ferromagnetic states depends strongly on the initial conditions. When we start in the ferromagnetic configuration, the transition J_{c_2} where ferromagnetic and layered states occur in equal number, takes place at $J_{c_2} \approx 1.7$ and for $J \gtrapprox 2.15$, when the energy of the ferromagnetic configuration becomes smaller than the energy of the layer-5 state, all final states are ferromagnetic. When we start in a random configuration, on the other hand, $J_{c_2} \approx 4.00$ so that layered states still dominate even for quite large values of J. Therefore, the final states as well as the details of the phase transition between layered and ferromagnetic states are strongly history-dependent. We know already from the energy calculations of section III that the energies of these different final states are more or less identical. This means that again, a competition of energetically very close-lying states leads to this scenario.

SUMMARY AND CONCLUSIONS

In summary, we have investigated the role of the ferromagnetic exchange interaction, characterized by the strength of the coupling constant J, on magnetic relaxation in systems of ultrafine magnetic particles, where also the magnetic dipole interaction and the anisotropy energy are taken into account. For transparency, we have considered the most basic structure, where all dipoles are located on the sides of a simple cubic lattice, with anisotropy axes pointing into the z-direction and compared Ising systems with Heisenberg systems. We have found that in both kinds of systems, essentially dipolar chains are formed, but that the details of the relaxation process are quite different in both cases.

Ising systems have a tendency to get frozen in configurations with relatively high energies. The freezing effects can be so strong that no relaxation could be observed during our simulations (even when starting at energies much higher than the groundstate energy). But even when – at smaller values of J and larger temperatures T – relaxation takes place, the groundstate energies are quite often not reached and the system gets frozen in some domain state of higher energy. Heisenberg systems, on the other hand, always show relaxation (except, when starting in the groundstate). Contrary to the Ising systems, they possess a landscape of states with different configuration of the magnetic moments but with close-lying energies, typical for spinglasses.

Consequently, in Heisenberg systems a more complex relaxation scenario takes place: For J close to zero, the chains form antiferromagnetic domains, while in the presence of a larger ferromagnetic interaction, the chains prefer ferromagnetic ordering between them and thus form ferromagnetic domains that grow with J. Around a critical J_{c_1}, a phase transition between domain states and layered states occurs where the exact value of J_{c_1} depends slightly on the start configuration. For even larger J, these layers develop a substructure where many individual layers are inclined against each other with the inclination angle depending on J. Finally, at a second phase transition at $J = J_{c_2} > J_{c_1}$, the system becomes ferromagnetic.

Our Heisenberg systems, even if very simple and geometrically ordered, clearly show history-dependence, frozen disorder and a rich landscape of states of close-lying energies, as it is typical for spinglasses. Frozen disorder occurs in the regime of small and intermediate J, where the system often relaxes into quite stable domain states of (+) and (-) domains with energies very close to the groundstate energy. At larger values of J, the modified layered states again show energies very close to the energy of the ferromagnetic state, so that again, two different states are in competition for the groundstate energy, leading to a history-dependence of the relaxation. Contrary to the situation at smaller J, both states are now ordered.

It could be interesting to investigate the change of J_{c_1} and J_{c_2} in response to system parameters as e.g. the system size L and the constant c/c_0 that determines the ratio between the dipole-dipole as well as the exchange energy as compared to the anisotropy energy (see Eq. (1)). While a thorough investigation would require additional time-consuming simulations, some qualitative considerations can be drawn from our present results. First of all,

as mentioned in section III, the ground state energies of the ordered Ising configurations were independent of the system sizes for $L \geq 6$, which means that the Ewald summation technique provides reliable energy values. There are, however minor effects, as e.g. the participation of states of different symmetry, as e.g. the layer-2 state in systems of side length $L = 8$ or the layer-3 state for $L = 6$ or $L = 12$ (as also mentioned in section III). Whenever these states are possible, they have some influence on the transition point to the ferromagnetic state, by serving as intermediate states through which the system can evolve. Moreover, the energy of the layer-$L/2$-state that is in direct competition to the ferromagnetic state, decreases with L. However, these can be considered as finite-size effects that should disappear in very large systems where very many different layered states states exist. For the dependence of the transitions with c/c_0, more simulations will be necessary in the future. The parameter J as well as the strength of the dipole interaction, both scale with c/c_0. Therefore, the transition value J_{c_1} should in first approximation also scale with c/c_0. The second transition point, J_{c_2}, on the other hand, strongly depends on the anisotropy energy. Therefore, its behavior under a change of c/c_0 cannot be determined on the basis of the current simulations.

Remanent magnetization could at best occur in the regime, where domains are formed. Due to the development of domains of different sizes for small and intermediate J, the numbers of positive and negative chains may differ, leading to a small finite magnetization. Since the relaxation depends on the history, there exists a slight excess of (+) chains, when starting in the ferromagnetic configuration, resulting in a small remanent magnetization and the appearance of a spurious ferromagnetic state. In the range of larger J, where the domain states disappear and transform into ordered layered states, the remanent magnetization naturally disappears, so that – in a counterintuitive way – the remanent magnetization disappears, when the ferromagnetic exchange interaction is increased. However, on the basis of the present simulations we cannot decide if the remanent magnetization at small J is a finite-size effect or not.

ACKNOWLEDGEMENTS

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[1] Batlle X and Labarta A 2002 *J. Phys. D* **35** R15
[2] Chen X, Sahoo S, Kleemann W, Cardoso S and Freitas P P 2004 *Phys. Rev. B* **70** 172411
[3] Jonsson T, Mattsson J, Djurberg C, Khan F A, Nordblad P, and Svedlindh P 1995 *Phys. Rev. Lett.* **75** 4138
[4] Chantrell R W, El-Hilo M, and O Grady K 1991 *IEEE Trans. Magn.* **27** 3570
[5] Luo W, Nagel S R, Rosenbaum T F, and Rosensweig R E 1991 *Phys. Rev. Lett.* **67** 2721
[6] Kleemann W, Petracic O, Binek C, Kakazei G N, Pogorelov Y G, Sousa J B, Cardoso S and Freitas P P 2001 *Phys. Rev. B* **63** 134423
[7] Bedanta S, Petracic O, Kentzinger E, Kleemann W, Rücker U, Paul A, Brückel T, Cardoso S and Freitas P P 2005 *Phys. Rev. B* **72** 024419
[8] Bedanta S, Eimüller T, Kleemann W, Rhensius J, Stromberg F, Amaladass E, Cardoso S and Freitas P P 2007 *Phys. Rev. Lett.* **98**, 176601
[9] Morup S, Madsen M B and Franck J J 1983 *Mag. Mag. Mat.* **40** 163
[10] Hansen M F, Koch C B and Morup S 2000 *Phys. Rev. B* **62** 1124
[11] Ulrich M, Garcia-Otero J, Rivas J and Bunde A 2003 *Phys. Rev. B* **67** 024416
[12] Porto M 2005 *Eur. Phys. J. B* **45** 369
[13] Verdes C, Ruiz-Diaz B, Thompson S M, Chantrell R W and Stancu A 2002 *Phys. Rev. B* **65** 174417
[14] Chubykalo-Fesenko O, Guslienko K, Klemmer T J, Wu X W, Chantrell R W and Weller D 2006 *Physica B* **382** 235
[15] El-Hilo M, Chantrell R W and O'Grady K 1998 *J. Appl. Phys.* **84** 5114
[16] Garcia-Otero J, Porto M, Rivas J, and Bunde A 2000 *Phys. Rev. Lett.* **84** 167
[17] Russ S, Bunde A 2006 *Phys. Rev. B* **74** 064426
[18] Russ S, Bunde A 2007 *Phys. Rev. B* **75** 174445
[19] Andersson J-O et al. 1997 *Phys. Rev. B* **56** 13983
[20] Kechrakos D and Trohidou K N 2003 *J. Mag. and Mag. Mat.* **262** 107
[21] Vargas P, d'Albuquerque e Castro J and Altbir 1999 *D Phys. Rev. B* **60** 6541 and Altbir D, Vargas P and d'Albuquerque e Castro 2001 *J Phys. Rev. B* **64** 012410
[22] Luttinger J M and Tisza L 1946 Phys. Rev. **70** 954
[23] Kretschmer R and Binder K *Z. Phys. B* 1979 **34** 375
[24] A. D. Liu and H. N. Bertram 2001 *J. Appl. Phys.* **89** 2861
[25] Brinzanik R, Jensen P J and Bennemann K H *Phys. Rev. B* **68** 174414 (2003).
[26] Hinzke D and Nowak U 1998 Phys. Rev. B **58** 265
[27] Majetich S A and Sachan M 2006 J. Phys. D **39** R407
[28] Nowak U Chantrell R W and Kennedy E C 2000 *Phys. Rev. Lett.* **84** 163
[29] In every step, we select a particle i at random and generate an attempted random change of the orientation of its magnetization vector $\vec{\mu}_i$. In the Heisenberg case, the attempted new $\vec{\mu}_i$ is chosen randomly in a spherical segment with an aperture angle $\Delta\alpha = 0.1$ around the present orientation of $\vec{\mu}_i$ (for details see Ref. [16, 17]). We define a "Monte Carlo (MC) step" as a number of $N = L^3$ such trials, so that per average each particle is chosen once per MC step.
[30] Allen M P and Tildesley D J *Computer Simulation of Liquids* (Clarendon, Oxford, 1987).