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Susceptibility of the disordered system of fine magnetic particles: a Langevin-dynamics study

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Abstract

Using the Langevin dynamics, we have calculated numerically the temperature and concentration dependence of the complex alternating-current susceptibility $\chi(\omega, T)$ for disordered systems of fine magnetic particles taking into account the dipolar interparticle interaction. We demonstrate that the behaviour of the $\chi(\omega, T)$ dependencies with increasing particle concentration (which means increasing interaction strength) and with increasing frequency depends *qualitatively* on the single-particle anisotropy and on the damping parameter used in the corresponding Langevin equation.

1. Introduction

The most challenging problem in the physics of disordered systems consisting of fine magnetic particles is the investigation of their dynamical properties, such as magnetic viscosity and ac susceptibility. The major difficulty here is due to the long-range and anisotropic dipolar interparticle interaction which prevents the development of both analytical and numerical methods powerful enough to handle these systems. During the last decade some progress in analytical methods for studying the *thermodynamics* of dipolar glasses and related systems was achieved (see, e.g., [1, 2] and references therein), but adequate analytical treatment of their dynamical behaviour will hardly ever be possible.

Corresponding numerical studies can be performed using either the Monte Carlo technique or numerical solution of the stochastic Langevin equations. There has been, to our knowledge, just a single attempt to study the corresponding dynamics using a Monte Carlo approach [3], where several interesting results concerning the temperature and frequency dependence of the ac susceptibility were obtained. However, in [3] only single-particle moment flips over the energy barrier were considered. Although both the single-particle anisotropy field *and* the interaction field were taken into account when evaluating the height of the corresponding barrier, this algorithm is obviously an oversimplification of the problem—especially for the

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most interesting case of strongly interacting systems with low and moderate single-particle anisotropies. Even if it were possible to overcome these difficulties, the two inherent drawbacks of this approach would still persist: (i) the justification of the relation between the Monte Carlo step and physical time is still highly questionable; and (ii) the purely dynamical phenomena (in this case, the precession of magnetic moments) cannot be taken into account.

These difficulties are absent in the Langevin-equation formalism, which was used to study the dynamics of systems of interacting fine magnetic particles in [4, 5]. Unfortunately, as was pointed out in [6], the method used for the solution of the corresponding stochastic equations in [4,5] is not self-consistent due to the improper interpretation of the Langevin equation using the Ito stochastic calculus (instead of the Stratonovich one). In [6] it was shown once more that for systems with a *multiplicative* noise the proper interpretation of the Langevin equation as the *Stratonovich* stochastic equation is *really* important. The authors of [6] study in the framework of the corresponding formalism the ac susceptibility of a *single-particle* system only, so proper Langevin-dynamics simulation of interacting many-particle systems of this kind seems not to have been performed up to now.

In this paper we present detailed results obtained from numerical simulations using the Langevin dynamics for disordered interacting systems of fine magnetic particles (a *brief* report on this work has been accepted as a 'regular contribution' for the proceedings of ICM-2000). We have studied the temperature dependence of the ac susceptibility of such systems for various particle volume concentrations, single-particle anisotropy values and precession damping parameters. The paper is organized as follows. In section 2 we describe our simulation method, which is essentially based on (i) the Heune scheme for the solution of the stochastic differential equations [6, 7]; and (ii) the extended Lorentz-cavity method for the calculation of the interaction field for dipolar glasses. In section 3 results for various fine-particle systems are presented, and the *qualitative* difference not only between the systems with high and low single-particle anisotropy values but also between the cases of low and moderate precession damping is demonstrated. Section 4 contains a comparison with the few theoretical results available [3, 8–12] and numerous experimental results, obtained mainly for ferrofluids (see, e.g., [13–17] and references therein). Several suggestions concerning the explanation of the observed discrepancies between the 'numerical' and real experiments are discussed.

2. Simulation method

We study an interacting system of randomly placed (but non-overlapping) identical spherical fine magnetic particles which are assumed to be absolutely single domain. This means that the motion of the particle magnetization can be described as the motion of a single vector $\boldsymbol{\mu}_i$, which represents the 'rigid'-particle magnetic moment (i denotes the particle number). For this reason we use as a starting point the stochastic Landau–Lifshitz–Gilbert equation [18] for the magnetic moment motion:

$$\frac{\mathrm{d}\boldsymbol{\mu}_i}{\mathrm{d}t} = \gamma\left[\boldsymbol{\mu}_i \times (\boldsymbol{H}_i^{\mathrm{eff}} + \boldsymbol{H}_i^{\mathrm{fl}})\right] - \gamma\frac{\lambda_i}{\mu_i}\left[\boldsymbol{\mu}_i \times \left[\boldsymbol{\mu}_i \times (\boldsymbol{H}_i^{\mathrm{eff}} + \boldsymbol{H}_i^{\mathrm{fl}})\right]\right] \tag{1}$$

where the precession constant γ is equal to the gyromagnetic ratio in the limit of small damping $\lambda \ll 1$, $\boldsymbol{H}_i^{\mathrm{eff}}$ denotes the *deterministic* effective field acting on the magnetic moment of the ith particle and $\boldsymbol{H}_i^{\mathrm{fl}}$ represents the corresponding fluctuation field. Below, we consider the case of identical particles and will omit the particle index i where it cannot lead to ambiguities.

The deterministic field $\boldsymbol{H}^{\mathrm{eff}}$ in the basic equation (1) includes the external field $\boldsymbol{H}^{\mathrm{ext}}$, the anisotropy field $\boldsymbol{H}^{\mathrm{an}}$ and the interparticle interaction field $\boldsymbol{H}^{\mathrm{dip}}$. Below, we assume that all

particles possess uniaxial anisotropy, so the anisotropy energy E^{an} is

$$E^{\mathrm{an}} = -KV(\boldsymbol{m}\cdot\boldsymbol{n})^2. \tag{2}$$

Here K denotes the anisotropy constant, V is the particle volume and the unit vectors $\boldsymbol{m}$ and $\boldsymbol{n}$ give the directions of the particle magnetic moment and the particle anisotropy axis, respectively.

The interparticle interaction field for the system under consideration is due solely to the magnetodipolar interaction and can be written as

$$\boldsymbol{H}_i^{\mathrm{dip}} = \sum_{j\neq i} \frac{3\boldsymbol{e}_{ij}(\boldsymbol{e}_{ij}\cdot\boldsymbol{\mu}_j) - \boldsymbol{\mu}_j}{r_{ij}^3} \tag{3}$$

where $\boldsymbol{e}_{ij} = \boldsymbol{r}_{ij}/r_{ij}$ and $\boldsymbol{r}_{ij}$ denotes the vector connecting the particles i and j.

Thermal fluctuations are included in the description of the magnetic moment motion via the so-called 'fluctuation field' $\boldsymbol{H}_i^{\mathrm{fl}}(t)$ whose cartesian components have the well known statistical properties [18]

$$\langle H_{\xi,i}^{\mathrm{fl}}\rangle = 0 \qquad \langle H_{\xi,i}^{\mathrm{fl}} H_{\psi,j}^{\mathrm{fl}}\rangle = 2D\delta_{ij}\delta_{\xi\psi} \tag{4}$$

where i, j are the particle numbers, ξ, $\psi = x$, y, z and the 'noise power' D is proportional to the system temperature T and depends on γ and the damping ratio λ as

$$D = \frac{\lambda}{1+\lambda^2}\frac{kT}{\gamma\mu}. \tag{5}$$

At this stage it is convenient to introduce reduced units for (i) the particle magnetic moment $\boldsymbol{m} = \boldsymbol{\mu}/M_{\mathrm{S}}V$, where M_{S} denotes the saturation magnetization of the particle material (so $|\boldsymbol{m}| = 1$), (ii) the magnetic field $\boldsymbol{h} = \boldsymbol{H}/M_{\mathrm{S}}$, (iii) the time $\tau = t\gamma M_{\mathrm{S}}$, (iv) the reduced frequency $w = \omega/\gamma M_{\mathrm{S}}$, (v) the anisotropy constant $\beta = 2K/M_{\mathrm{S}}^2$ and (vi) the distance $\rho = r/R$, where R is the particle radius. In these units the basic equation (1) reads

$$\frac{\mathrm{d}\boldsymbol{m}_i}{\mathrm{d}\tau} = \left[\boldsymbol{m}_i\times(\boldsymbol{h}_i^{\mathrm{eff}} + \boldsymbol{h}_i^{\mathrm{fl}})\right] - \lambda_i\left[\boldsymbol{m}_i\times\left[\boldsymbol{m}_i\times(\boldsymbol{h}_i^{\mathrm{eff}} + \boldsymbol{h}_i^{\mathrm{fl}})\right]\right] \tag{6}$$

and the reduced anisotropy field obtained from (2) is

$$\boldsymbol{h}_i^{\mathrm{an}} = \beta\boldsymbol{n}_i(\boldsymbol{m}_i\cdot\boldsymbol{n}_i) \tag{7}$$

and also the reduced interaction field can be expressed as

$$\boldsymbol{h}_i^{\mathrm{dip}} = \frac{4\pi}{3}\sum_{j\neq i}\frac{3\boldsymbol{e}_{ij}(\boldsymbol{e}_{ij}\cdot\boldsymbol{m}_j) - \boldsymbol{m}_j}{\rho_{ij}^3}. \tag{8}$$

The reduced temperature will be defined in units of the stray-field energy as $\tilde{T} = kT/M_{\mathrm{S}}^2V$. This definition is more convenient than the usual definition in units of the particle anisotropy energy KV because it can also be used to study particles without intrinsic anisotropy and enables a direct comparison of systems with different single-particle anisotropy values without rescaling the temperature dependencies.

Care must be taken in the evaluation of the dipolar field (8) due to the long-range nature of this interaction. In principle, for each particle the sum in (8) must be performed over all other system particles (for finite systems) or using the Ewald method for systems with periodic boundary conditions [19].

We employ periodic boundary conditions to reduce finite-size effects, so the Ewald method would be the most appropriate choice. However, we have found that for all kinds of simulation presented in this report it is sufficient to use the much simpler extended Lorentz-cavity method as suggested and improved by many authors (see, e.g., [3, 20, 21]). In this method the

contributions to the interaction field acting on the given particle from all the particles inside the sphere with the 'critical radius' R_c around this particle are taken into account exactly (the summation (3) is performed only over those particles j for which $r_{ij} \leqslant R_c$). The field from the region outside this R_c-sphere is treated as the field of the homogeneously magnetized media (with the corresponding average magnetization) inside the spherical cavity. The critical radius R_c should be determined 'experimentally' using the obvious idea that for the appropriate choice of R_c its further increase should not affect the results. We have found that the value $R_c = 2\langle\Delta r\rangle$, where $\langle\Delta r\rangle$ is the average interparticle distance, is sufficiently large for all the particle concentrations used in our study. A typical example of the test simulations used to determine the critical radius R_c is presented in figure 1, where the temperature dependence of the susceptibility calculated for various R_c-values is displayed. It can be seen that on changing the ratio $R_c/\langle\Delta r\rangle$ from 1.0 to 1.5 a substantial and *systematic* shift of the $\chi''(T)$ is observed, whereby the differences between the curves for $R_c/\langle\Delta r\rangle = 1.5$ and $R_c/\langle\Delta r\rangle = 2.0$ are random and lie within the statistical errors. The results for $R_c/\langle\Delta r\rangle = 3.0$ are indistinguishable from those obtained for $R_c/\langle\Delta r\rangle = 3.0$ and are not shown.

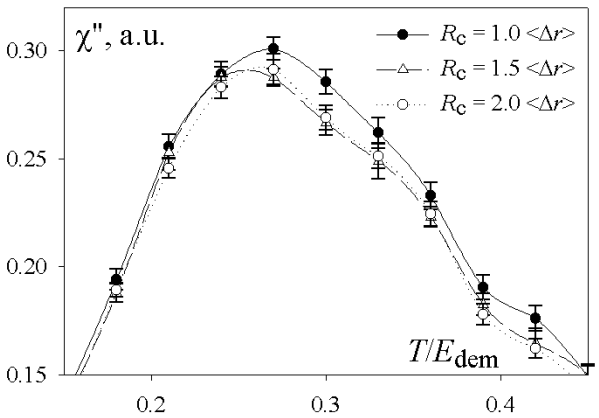

Figure 1. A test example for the choice of the cut-off radius (a system of particles with $\beta = 2.0$, $\lambda = 0.1$ and concentration $c = 0.08$; reduced frequency $w = 0.03$). Susceptibility curves are calculated using various values of the cut-off radius R_c as shown in the key.

To obtain physically consistent results one should interpret the basic Langevin equation (6) in the Stratonovich sense [7]. It was recently pointed out once more [6] that for the case of multiplicative noise (as in (6)) it *is* important which stochastic calculus (Ito or Stratonovich) is used in the solution of the corresponding stochastic differential equation and that the *Stratonovich* calculus should be used to obtain correct physical properties of magnetic systems. For this reason we have used for the solution of (6) the stochastic Heune scheme [7] which is known to converge quadratically to the Stratonovich solution of the corresponding stochastic equation [22]. After trying several methods we have found that this scheme seems to represent the optimal compromise when trying to achieve the maximal possible accuracy while keeping the programming effort reasonably moderate.

The time step $\Delta\tau$ used in the numerical solution of (6) was chosen (as usual) as large as possible in order to perform simulations over sufficiently long times, but small enough to keep discretization errors in the frames of the statistical accuracy of the results. We have found that the value $\Delta\tau = 0.05$ is an appropriate choice for all parameter sets used in our simulations.

The real ($\chi'(\omega, T)$) and imaginary ($\chi''(\omega, T)$) parts of the ac susceptibility of the system were calculated in a standard way. First of all, several thousand time steps L_{th} were performed

for the thermalization of the system starting from a random initial state. The corresponding number of time steps $L_{\rm th}$ was determined from the requirement that after the thermalization the system energy should remain constant in frames of thermodynamical fluctuations (no systematic changes in the system energy should be observed). Afterwards an oscillating field

$$h_z = h_0 \cos w\tau \tag{9}$$

was applied to the system and the real and imaginary susceptibility parts were calculated as

$$\chi'(w, \tilde{T}) = \frac{1}{h_0}\frac{1}{L_\tau}\sum_{l=1}^{L_\tau}\langle m_{l,z}\rangle \cos w\tau_l \tag{10}$$

$$\chi''(w, \tilde{T}) = \frac{1}{h_0}\frac{1}{L_\tau}\sum_{l=1}^{L_\tau}\langle m_{l,z}\rangle \sin w\tau_l \tag{11}$$

where $\langle m_{l,z}\rangle$ is the z-projection of the system magnetization per particle at the time step l and $\tau_l = l\,\Delta\tau$. The total number of steps L_τ was chosen such that for the given time step $\Delta\tau$ the measurements were performed during an *integer* number of field cycles N_c ($L_\tau = 2\pi N_c/w\,\Delta\tau$).

Additional checks were performed to verify the linearity of the system response by performing the simulations with various oscillating-field amplitudes h_0 and checking that the susceptibilities obtained do not depend on the h_0-value. Performing such tests, we have looked for a universal criterion which would enable us not to have to perform the linearity check for each set of the system parameters (e.g., single-particle anisotropy β and particle volume concentration c). For an *interacting* system it is not possible to use for such a universal criterion just the maximal value of a single-particle anisotropy field, $h_K = H_K/M_{\rm S} = 2K/M_{\rm S}^2$ (as was done in [6] for a single-particle case). The problem is that for various β- and c-values the relation between the anisotropy and interaction field acting on a single-particle magnetic moment may be very different. For this reason the h_0-value chosen on the basis of just the h_K-value would be too small for systems with weak single-particle anisotropy and high volume concentration. This, in turn, would result in unnecessarily large statistical errors in the χ-'measurements'.

For this reason we have adopted the following criterion for the h_0-choice. For each pair of (β, c) values and for a random orientation of single-particle moments, we have evaluated the average magnitude $h^{\rm tot}$ of the total (anisotropy plus interaction) deterministic field acting on each particle in the absence of an external field. Then we have chosen the oscillating-field amplitude as a fraction of this average magnitude: $h_0 = ah^{\rm tot}$. The value $a = 0.2$ was found to be large enough to provide reasonably small statistical errors but still small enough to ensure the linearity of the system response.

Successful tests of our program code were performed on a system of non-interacting particles with aligned and randomly oriented anisotropy axes: results obtained with our program were in quantitative agreement (in the frames of the statistical errors) with known analytical results as well as with recent numerical calculations performed by Garcia-Palacios and co-workers (see [6] and references therein).

3. Results and discussion

As was mentioned in the introduction, we have studied the dependence of the ac susceptibility $\chi(\omega, T)$ on various system parameters: (i) particle concentration c (which controls the interaction strength); (ii) the single-particle anisotropy strength β; (iii) the damping parameter λ (see basic equation (6)); and (iv) the frequency of the oscillating field ω.

For most of the results presented below, simulations were carried out on a system consisting of $N_p = 256$ particles positioned randomly (but not overlapping) inside a cubic volume. Periodic boundary conditions were assumed. For some key parameter sets, simulations were repeated for $N_p = 512$ particles to ensure that the results were independent of the system size. For all parameter sets, susceptibility values were sampled for at least $N_c = 20$ oscillating-field cycles, so for the typical reduced frequency studied, $w = 0.03$, about $L_\tau \sim 10^5$ time steps were required. Averaging over at least $N_{conf} = 16$ (mostly $N_{conf} = 32$) independently generated spatial particle configurations was performed. A typical simulation run time for obtaining a single $\chi(T)$ dependence for a system with the parameters listed above (which means also the averaging over 16–32 system configurations) on an Alpha 500 MHz workstation using a program code written in Fortran90 was about 20–40 h.

The most interesting question concerning the behaviour of the systems under study is that of the influence of the *interparticle dipolar interaction* on the system dynamics—in particular, on its ac susceptibility $\chi(T)$. To study this problem we have performed simulations for various particle volume concentrations keeping all other system parameters fixed. The corresponding results are shown in figures 2–6, where it can be clearly seen that changes in the $\chi(w, \tilde{T})$ curves with the increasing particle concentration depend *qualitatively* on the single-particle anisotropy value β and on the damping parameter λ.

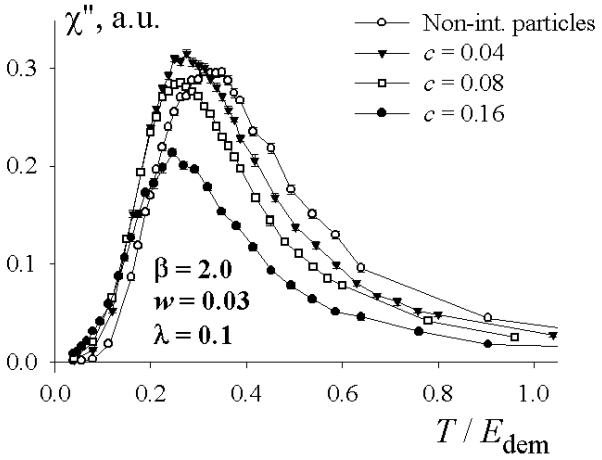

Figure 2. $\chi''(w, T)$ for *large* single-particle anisotropy $\beta = 2.0$ and *low* damping $\lambda = 0.1$.

We are interested mainly in the distribution of the free-energy barriers in the systems under study, so below we shall show results concerning the imaginary part of the ac susceptibility only.

3.1. High- and moderate-anisotropy cases

For moderate and large anisotropies ($\beta \geqslant 1$; see figures 2–4) the peak in the $\chi''(\tilde{T})$ dependencies shifts towards *lower* temperatures with the growing particle concentration (increasing interaction strength). This shift can be clearly seen especially for systems with low damping: results for $\beta = 2.0$ and $\lambda = 0.1$ are shown in figure 2, and details of the corresponding $\chi''(\tilde{T})$ peaks are displayed in figure 3. When the dissipation increases, the peak shift gets weaker and it can hardly be seen for the moderate damping (figure 4, $\beta = 2.0$ and $\lambda = 1.0$). These results mean that the dipolar interaction leads to a *decrease* of the free-energy barriers in fine-particle

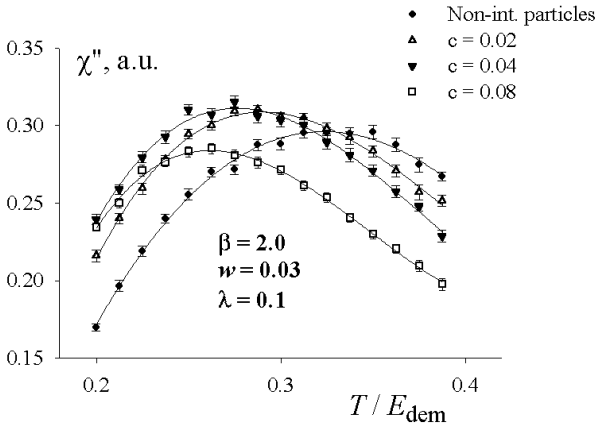

Figure 3. Peak details for figure 2.

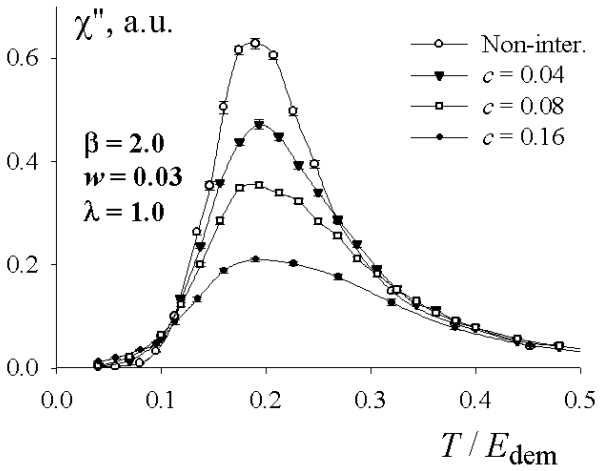

Figure 4. $\chi''(w, T)$ for *large* single-particle anisotropy $\beta = 2.0$ and *moderate* damping $\lambda = 1.0$.

systems with high and moderate anisotropies and this effect is more pronounced in systems with low damping.

We would also like to point out another interesting feature of the $\chi''(\tilde{T})$ curves for the moderate-anisotropy case and *low* damping—the *non-monotonic* dependence of the peak *height* on the particle concentration. To display this feature more clearly, we show the enlargement of the corresponding peaks in figure 3, where $\chi''(\tilde{T})$ dependencies for a non-interacting system and for interacting systems with the particle volume concentrations $c = 0.02$, 0.04 and 0.08 are shown. The peak height of the $\chi''(\tilde{T})$ curves increases from the non-interacting system up to the particle volume fraction $c \approx 0.04$, after which it starts to decrease (see the curve for $c = 0.08$ in the same diagram, figure 3, and the curve for $c = 0.16$ in figure 2). The effect is small, but clearly beyond our statistical errors. We have observed this feature for systems with moderate single-particle anisotropy and *low* damping only—for the moderate-damping case the peak height already decreases monotonically with increasing concentration (figure 4). At present we do not have any qualitative physical explanation for this phenomenon.

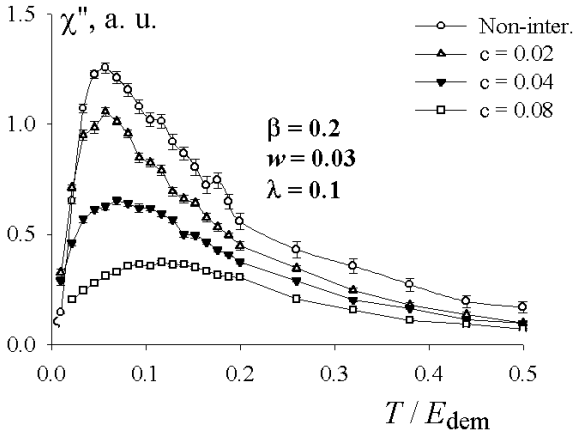

Figure 5. $\chi''(w, T)$ for *small* single-particle anisotropy $\beta = 0.2$ and *low* damping $\lambda = 0.1$.

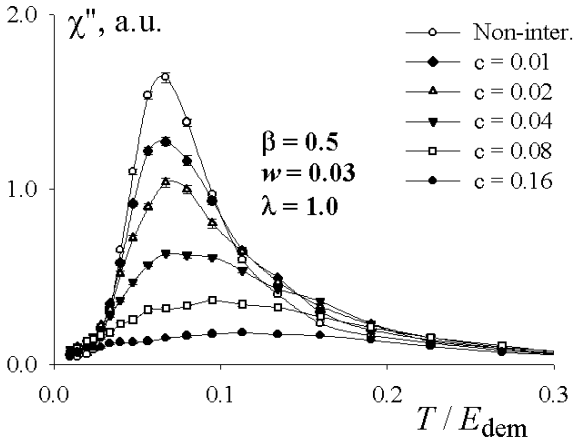

Figure 6. $\chi''(w, T)$ for *small* single-particle anisotropy $\beta = 0.5$ and *moderate* damping $\lambda = 1.0$.

We have also compared the temperature dependencies of the ac susceptibilities for various *frequencies* keeping the particle concentration constant. The corresponding result for $\beta = 2.0$ is presented in figure 7, where $\chi''(w, \tilde{T})$ curves for the particle volume fraction $c = 0.08$ and reduced frequencies $w = 0.03$, 0.1 and 0.3 are shown. It can be clearly seen that with increasing frequency the peak position is shifted towards *higher* temperatures, whereby the peak height *decreases* (see below for a comparison with experimental results).

3.2. Low-anisotropy case

For sufficiently small anisotropy values (how small depends on the dissipation parameter), the $\chi''(w, \tilde{T})$ peak shifts towards *higher* temperatures when the particle concentration increases—see figure 5 ($\beta = 0.2$, $\lambda = 0.1$) and figure 6 ($\beta = 0.5$ and $\lambda = 1.0$). Such a shift can be easily understood, because the low single-particle anisotropy β means that the interparticle interaction already makes the dominant contribution to the effective field for moderate particle concentration, thus leading to the increase of the average energy barrier height when increasing the interaction strength (particle concentration).

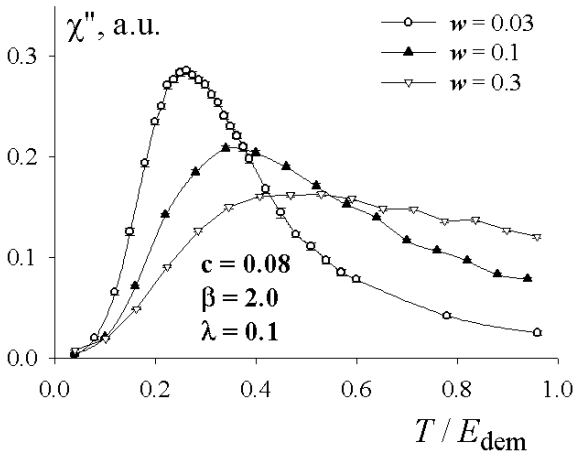

Figure 7. The frequency shift of the $\chi''(w, T)$ dependencies for *large* anisotropy $\beta = 2.0$.

The frequency shift of $\chi''(\tilde{T}, w)$ for the particle system with the low anisotropy $\beta = 0.5$ is shown in figure 8, where corresponding χ''-dependencies for the frequencies $w = 0.003$, 0.01 and 0.03 are displayed. The peak position moves towards *higher* temperatures with increasing frequency for this system also; for the physical explanation for this—probably almost universal—behaviour, see, e.g., [6]. Note, however, that the peak height now *increases* when the frequency increases. This behaviour is qualitatively different from that in the high-anisotropy case and thus is very important when comparing our results with experimental data (see below).

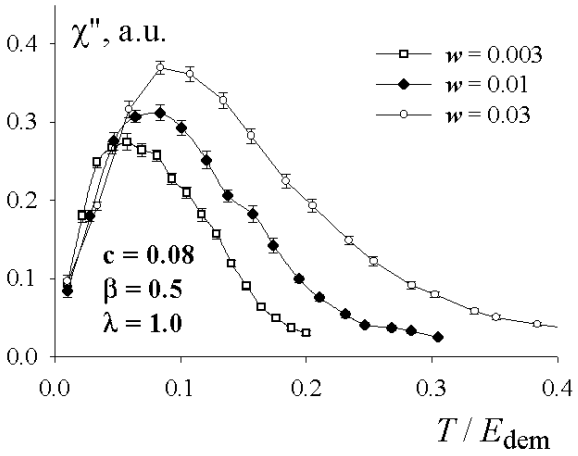

Figure 8. As in figure 8, but for the system with the *small* anisotropy $\beta = 0.5$.

3.3. Comparison with other theoretical results

There is a substantial body of theoretical work done on this subject, but very few reliable results for *interacting* dipolar magnetic systems have been obtained up to now—mainly due to the already mentioned long-range and anisotropic nature of the dipolar interaction.

To our knowledge, all analytical approaches applied for the calculation of the ac susceptibility in such systems are based on the energy barrier approximation [23]. This approximation assumes that the system can be considered as a set of more or less independent subsystems each of which has its own relaxation time. The distribution of these relaxation times is due to the different heights of the energy barriers E for corresponding subsystems. If the distribution of the energy barriers $\rho(E)$ is known, then the calculation of the corresponding relaxation time distribution $f(\tau_{\rm rel})$ is a straightforward task—at least in the Arrhenius approximation ($\tau_{\rm rel} \sim \exp(E/T)$). The last (and also trivial) step in this approach is the convolution of the $f(\tau_{\rm rel})$ distribution obtained with, e.g., the kernel $\omega\tau_{\rm rel}/(1 + (\omega\tau_{\rm rel})^2)$ to obtain the imaginary part of the ac susceptibility $\chi''(\omega, T)$.

For these reasons most authors attempt to calculate the distribution of the energy barriers. Corresponding analytical methods are available only for the weak-interaction case—either in the form of the local field theory [8, 9] or as the pair approximation (for the latest and most detailed work, see [10]). The latter implicitly assumes that the pair interaction, i.e., interaction with the nearest neighbour, plays the dominant role and thus is valid for small particle concentrations and hence in the weak-interaction regime only. Both methods lead to the conclusion that the dipolar interaction leads on average to the *decrease* of the system energy barriers—after averaging over the local field distribution [8,9] or the pair orientations [10] (see, however, the discussion in [11]).

These conclusions seem to support our result that for *large* single-particle anisotropy (the weak-interaction case; see figure 2) the $\chi''(w, \tilde{T})$ peak shifts towards *lower* temperatures which means that the energy barrier distribution moves towards *lower* barriers. However, it should be noted that theories of this kind are not very well suited for checking numerical or experimental results concerning the irreversible processes in magnetic systems in general and ac susceptibility data in particular, because (i) the effect predicted by such theories is weak by the very definition of the perturbation approach, (ii) the temperature dependence of the energy barrier heights and the features of the energy landscape near the saddle point are ignored and (iii) the precession of magnetic moments cannot be taken into account (which may be quite important: compare, e.g., figures 2 and 4).

The distribution of the energy barriers can be evaluated rigorously—i.e., beyond the perturbation approach—using the numerical minimization of the thermodynamical action for the transition between the two metastable states given [12]. Using this method, we were able to show that in systems of fine magnetic particles with high and moderate anisotropies, the energy barrier distribution does indeed move towards *lower* barriers with increasing particle concentration [12]. But it is still quite difficult to draw conclusions concerning the ac susceptibility based on this fact alone, for the reasons (ii) and (iii) mentioned above.

The ac susceptibility itself for an *interacting* system of fine magnetic particles was studied, to our knowledge, only in [3], using the Monte Carlo method. Bearing in mind that this method is not able to take into account the *precession* of magnetic moments, we can try to compare results from [3] with our data obtained in the moderate-damping regime, where the precession is expected to play a minor role (we have also performed simulations in the high-damping case—for $\lambda = 10$—and could not find any qualitative differences between the high- and moderate-damping cases). Simulations in [3] were carried out for particles with the anisotropy $K = 1.6 \times 10^5$ erg cm^{-3} and saturation magnetization $M_{\rm S} = 420$ G, so the reduced anisotropy constant for these particles is $\beta = 2K/M_{\rm S}^2 \approx 1.8$ and the results of [3] should correspond to our data for $\beta = 2.0$.

Indeed, the shift of the $\chi''(\omega, T)$ peak with increasing particle concentration found in [3] is qualitatively similar to our results (compare figure 2 from [3] with our figure 4). However, the relative peak *heights* for various concentrations are considerably different. The same

applies to the *frequency* shift of the $\chi''(\omega, T)$ curves (see figure 3 in [3] and our figure 7): the $\chi''(T)$ peak moves towards higher temperatures with increasing frequency for both result sets, but the peak height *increases* with ω in [3], whereby according to our simulations it should *decrease* with growing frequency for the high and moderate anisotropies β (figure 7). The most probable reason for this discrepancy is that the Monte Carlo approach used in [3] takes into account single-particle flips *only*, thus neglecting collective remagnetization processes. Such processes are not expected to play a dominant role in systems with moderate and high single-particle anisotropies, but for moderate anisotropy values can nevertheless affect the results quantitatively.

3.4. Comparison with experimental data

The ac susceptibility of magnetic nanocomposites is one of their most important features both from the fundamental point of view and from that of evaluation of their suitability for numerous technical applications [24, 25]. For these reasons there exist a huge number of experimental papers where $\chi(\omega, T)$ has been measured. From experimental data available for various nanocomposites we have chosen mainly results obtained on frozen ferrofluids, because (i) these systems can be characterized reasonably well and (ii) very accurate and reliable results on ferrofluids are available (for the best experiments, see [13–15, 17]).

Corresponding measurements are usually performed on ferrofluids consisting of magnetite or maghemite particles. In most of the studies, the following system behaviour was observed [13–17]:

(i) Concentration shift of $\chi(\omega, T)$: the peak in the $\chi''(\omega, T)$ curves moves towards *higher* temperatures with increasing concentration c, whereby the height of this peak decreases *monotonically* with growing concentration.
(ii) Frequency shift of $\chi(\omega, T)$: the peak in the $\chi''(\omega, T)$ curves moves towards *higher* temperatures with increasing frequency ω, whereby the height of this peak *increases* with growing frequency.

Such behaviour agrees very well (at least qualitatively) with our results for systems with low single-particle anisotropies $\beta = 0.2$–0.5 (see figures 5, 6 and 8). However, there is apparently an important disagreement between our simulation results and the experiments cited above: the anisotropy $\beta = 2K/M_S^2$ (calculated from the K- and M_S-values reported, e.g., for particles studied in [13, 15]) for these measurements is about $\beta \approx 1.5$–2.0, so the opposite shift of the $\chi''(T)$ peak with the particle concentration should be observed according to our simulations (see figure 2 for the low-damping case) or this shift should be very weak (figure 4, moderate damping). Below, we discuss two possible reasons for this discrepancy.

3.4.1. Particle aggregation. First of all, such a disagreement could be caused by the particle aggregation in real ferrofluids. Such an aggregation would lead not only to the *spatially* inhomogeneous particle distribution, but also to the correlations between the anisotropy axis *directions* of the neighbouring particles (due to the particle rotation during the aggregation process). These correlations would obviously increase the energy barriers in the system under study, thus leading to the shift of the $\chi''(\tilde{T})$ peak towards *higher* temperatures with increasing particle concentration.

Whether such an aggregation takes place can be estimated by comparing the thermal energy $E_T = kT$ (at the *freezing temperature* T_{freeze} of the carrier fluid) with the energy of the magnetodipolar interparticle interaction $E_{\text{dip}} \sim (VM_S)^2/r_{\text{min}}^3$ for two particles with the minimal possible separation between them, r_{min} (which, in turn, can be estimated as

$r_{\min} = d_{\rm p} + 2h_{\rm surf}$ when the particle diameter $d_{\rm p}$ and the thickness of the surfactant layer $h_{\rm surf}$ surrounding the particles are known). Corresponding estimations for the parameter values typical for oil-based magnetite and maghemite ferrofluids ($d_{\rm p} \approx 10$ nm, $M_{\rm S} \approx 400$ G, $h_{\rm surf} \approx 5$ nm, $T_{\rm freeze} \approx 200$ K) lead to the conclusion that the thermal energy is several times larger than the maximal dipolar interaction energy, so the aggregation is quite unlikely. However, one should keep it in mind that such estimations are very sensitive to the assumed particle sizes and the thickness of the surfactant layer. This means that, due to the unavoidable size distribution of ferrofluid particles and 'quality distribution' of the surfactant layer, some aggregates may be present in any ferrofluid—the only question is how many of them are there. Another point is that for the *water*-based ferrofluids considerable aggregation due to the concentration of particles on the boundaries between the growing ice crystals may occur.

3.4.2. Size-dependent anisotropy. The more probable reason for the discrepancy under discussion is the following. The value of the anisotropy constant K reported for the given ferrofluid sample is usually the *average* value obtained from the corresponding low-temperature hysteresis loop of a sample [26]. Different particles contribute to this hysteresis loop—and hence to the average K-value—with the weight proportional to their magnetic moment μ. According to the susceptibility measurements, however, the particle contributions are proportional to the *square* of their moment μ^2 (see, e.g., [27]), which means that the contribution of larger particles is much more significant in the latter case.

On the other hand, it is well known that the total effective anisotropy (as well as other magnetic properties) of fine magnetic particles depends strongly on the particle size [28–30]. The reason is that this total anisotropy is the sum of the contributions arising from the crystallographic, surface and shape anisotropies (the latter occurring due to the deviation of the particle form from a perfect sphere). The last two contributions depend on the particle size—the surface anisotropy due to the decreasing fraction of surface atoms with the growing particle size and the shape anisotropy because the shape fluctuations may decrease when the particle size and hence the number of particle atoms increases.

For all experimentally studied systems it was found that the total effective anisotropy decreases with growing particle size [28–30]. Hence larger particles with larger magnetic moments (which contribute significantly more to the susceptibility than to the magnetization itself) have much smaller values of the effective anisotropy β, thus leading to the shift of the $\chi''(T)$ peaks towards *higher* temperatures with increasing particle volume fraction; this is as it should be for the low-anisotropy case (see figures 5 and 6). The increase of the particle saturation magnetization $M_{\rm S}$ with the growing particle size, which was recently confirmed experimentally once more [31], would enhance this effect.

4. Conclusions

We have performed Langevin-dynamics simulations of the irreversible remagnetization processes in systems of fine magnetic particles and were able to measure the corresponding ac susceptibility as a function of the particle concentration, single-particle anisotropy, damping parameter and frequency of the applied field. We have demonstrated that the ac susceptibility peak can move with the increasing particle concentration (dipolar interaction strength) both towards lower and towards higher temperatures, depending on (i) the single-particle anisotropy and (ii) the damping parameter. This means, in turn, that the dipolar interaction can either decrease the (free-) energy barriers in the system under study—if such barriers are already

created by a relatively high single-particle anisotropy—or increase these barriers—if they are mainly due to the dipolar interaction itself.

We point out that our method takes the dipolar interparticle interaction fully into account, incorporating all the dynamic and static correlations between the particle magnetic moments. For this reason, the discrepancy found between our results and real experiments performed on ferrofluids with apparently the same single-particle parameters shows that *some additional features* of the fine-particle systems should be taken into account to understand their dynamical properties adequately. We propose that such a feature could be (leaving aside trivial reasons like the particle aggregation) the dependence of the total single-particle anisotropy and the particle saturation magnetization on its *size*. Further experimental studies of such dependencies are clearly necessary to provide the corresponding quantitative input for numerical simulations which could verify this idea.

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