Numerical modeling of the magnetic response of interacting superparamagnetic particles to an ac field with arbitrary amplitude

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Abstract  The dynamic magnetic response of immobilized superparamagnetic nanoparticles to an ac field with arbitrary amplitude is studied using numerical simulations. The nanoparticles are considered to be distributed randomly within an implicit solid matrix, but the easy axes of the particles are aligned parallel to the ac magnetic field. Modeling of dynamic response is based on the Fokker-Planck-Brown equation in which the interparticle dipole-dipole interactions are included within the framework of the modified mean-field theory. The effects of the magnetic crystallographic anisotropy barrier, the ac field amplitude, and the interparticle interactions on the dynamic magnetization, susceptibility, and relaxation time are analyzed. It is shown that an increase in the amplitude of the ac field significantly accelerates relaxation processes in the system under consideration whereas an increase in magnetic anisotropy of a particle and interparticle dipole-dipole interactions slows them down. The numerical results for dynamic susceptibility and relaxation time are compared against theories reliable in the weak ac field, and an excellent agreement is obtained.

1. Introduction

Embedding a large number of magnetic nanoparticles into liquid or polymer matrix makes it possible to control the properties of the composite material using an external magnetic field [1, 2, 3]. Such materials, so-called magnetic soft matter, include ferrofluids, magnetic elastomers, ferrogels, and various biocompatible magnetic filling. These complex systems attract a considerable amount of interest from researchers, medics, and engineers because they are actively used in progressive high industrial and biomedical technologies. Examples include crawling “robots”, drug delivery, sealants, heat-conduction media, separation media, gas fluidized beds, contrast agents for magnetic resonance, and magnetic hyperthermia [4, 5, 6, 7].

The response of the magnetic soft matter to the applied magnetic field is determined by two main physical mechanisms of the magnetic moment orientational relaxation in nanosized particles. They are the Brownian rotation of particles with fixed magnetic moments and the superparamagnetic Néel rotation of the magnetic moments inside the particles due to thermal fluctuations [8, 9, 10]. For an ensemble of nanoparticles, suspended in some liquid carriers, known as ferrofluids, both mechanisms take place. But in the case, when particles are embedded in some polymer matrix or biological tissues, very often the particles lose their translational and orientational degrees of freedom. In this case, the superparamagnetic Néel relaxation becomes the major mechanism determining the magnetic properties of the ensembles of such immobilized particles.

Most of the known theoretical models of the magnetic response of particles immobilized in some matrix concern noninteracting samples [8, 9, 11, 12, 13, 14]. For example, the description of the dynamic susceptibility of noninteracting particles to the weak ac magnetic field with angular frequency $\omega$ is generally performed with the Debye model [15, 16]

$$\chi_D = \chi_L \frac{1}{1 - i\omega\tau},$$

(1)

where $\chi_D$ is the dynamic susceptibility within Debye framework, $\chi_L$ is the Langevin susceptibility and $\tau$ is the effective relaxation time of the magnetic moment. In the weak ac field, relaxation time of immobilized magnetic particles with uniaxial magnetic anisotropy is determined by well-known formulas. For low potential barriers, when the particle anisotropy parameter $\sigma \ll 1$, the relaxation time is close to $\tau = \tau_D = \sigma\tau_0$, but it grows drastically with $\sigma$, and at $\sigma \gg 1$ it is very close to the Néel asymptotic expression $\tau = \tau_N = \tau_0 \exp(\sigma)$, where $\tau_0 = m/2\alpha\gamma E_A$, $m$ is the particle magnetic moment, $\alpha$ is the spin-lattice relaxation parameter, $\gamma$ is the gyromagnetic ratio. The parameter $\sigma = E_A/k_B T$ has a meaning of the energy barrier of magnetic crystallographic anisotropy $E_A$, measured in thermal energy units $k_B T$. In Ref. [18] it was proposed the approximate formula which is valid for all barrier heights $\sigma$ for the longitudinal relaxation time of a single domain ferromagnetic particle with uniaxial anisotropy

$$\tau = \tau_D e^{\sigma - \frac{1}{2\sigma}} \left[ \frac{\sigma}{\sigma + 1} \sqrt{\frac{\sigma}{\pi}} + 2^{-\sigma-1} \right]^{-1}.$$

(2)

For a number of applications, however, the knowledge of the relaxation times in larger magnetic fields is of vital importance. For example, in Magnetic Particle Imaging (MPI) the magnetic markers are exposed to the ac fields with different amplitudes [19, 20]. When the relaxation time is larger than the time scale of the time-varying ac magnetic field, the system does not have time to reach an equilibrium state as the magnetic field changes. In this case, one would expect a weak magnetic response since the magnetic moment would not be able to sufficiently keep pace with the ac magnetic field. However, for the ac field with a sufficiently high
amplitude a strong magnetic response can be still obtained because of the decreasing Néel relaxation time with increasing field strength, allowing the magnetic moment to better coincide with the magnetic field. The decreasing Néel relaxation time with an increasing static magnetic field strength was determined in the work [13]. The amplitude dependence of the dynamic properties of noninteracting magnetic particles in an ac field was studied in [13, 21, 22].

However, the magnetic particle response is also influenced by the dipolar interaction [23, 24, 25]. Strong interparticle dipole-dipole interactions in the sample where the particles have some degree of mobility can even lead to the formation of ferroparticle aggregates [26, 27, 28, 29]. Recent papers have shown that the dipolar interaction has a significant influence on heat production in magnetic hyperthermia treatment [30, 31]. In Refs. [24, 32, 33, 34] it has been theoretically studied the ensembles of magnetic particles with Brownian or Néel relaxation mechanism in the weak ac magnetic field to clarify the effects of dipolar interactions on the susceptibility spectrum. In all cases, dipole-dipole interactions are predicted to increase the relaxation times. In physical terms, strong correlations between particles mean that there is the collective cluster-type behavior, which is slower than the single-particle behavior. Such effects have been also detected in the experiments [35, 36, 37].

Thus, an increase of the ac field amplitude speeds up of relaxation processes in the system of immobilized superparamagnetic particles, while interparticle correlations slow down these processes. The problem is that there are no studies which take in to account the influence of both the interactions and the ac field amplitude on the magnetic response and relaxation processes in an ensemble of immobilized superparamagnetic particles. It goes without saying that it is vital to understand these effects in order to successfully using these systems in various applications and to predict correctly the properties of these media.

In this work, we will study the dynamic magnetization and susceptibility of the ensemble of interacting immobilized superparamagnetic particles with uniaxial magnetic anisotropy. Special attention will be paid to the relaxation processes of the magnetic moments of particles in the ac field with arbitrary amplitude. Modeling of magnetization processes will be based on the Fokker-Planck-Brown (FPB) equation [17, 38]. Interparticle interactions will be taken into account using the method proposed in [24]. The solution of FPB equation will be based on the unconditionally stable scheme for convection-diffusion problems proposed in [39].

2. Model and methods

2.1. Model and basic properties

We study here the dynamic magnetic response of an ensemble of identical spherical uniaxial superparamagnetic particles. Each particle contains the central uniformly magnetized core of diameter \(d\), and the saturation magnetization of the magnetic core material is \(M_0\). So, the particle magnetic moment is \(m = \pi d^3 M_0/6\). Particles are uniformly distributed and immobilized in some nonfluid matrix, so both the translational and rotational degrees of freedom of the particle bodies are "frozen", and the particle numerical concentration \(\rho\) can be considered as constant. The direction of the particle easy magnetization axis is defined by a unit vector \(\hat{n}\), and we consider the case when all particle easy axes are co-aligned and parallel to \(Oz\) axis; vector \(\hat{n} = (0, 0, 1)\) is identical for all particles. But the particle superparamagnetism means that the magnetic moments are able to rotate inside the magnetic cores due to thermal fluctuations. So, the direction of \(i\)-th magnetic moment \(\mathbf{m}_i = m\hat{m}_i = m (\sin \theta_i \cos \varphi_i, \sin \theta_i \sin \varphi_i, \cos \theta_i)\) differs from \(\hat{n}\); here \(\theta\) is polar angle, and \(\varphi\) is azimuthal angle of the unit vector \(\hat{m}\) in spherical coordinate system. The magnetic moment rotation is hindered by the Néel energy \(U_N\) of the magnetic anisotropy:
\[ U_N(i) = -Kv_m (\mathbf{m}_i \cdot \mathbf{n})^2 = -Kv_m \cos^2 \theta_i, \quad v_m = \pi d^3 / 6, \]  

(3)

where \( K \) is the magnetic crystallographic anisotropy constant.

To avoid the demagnetization corrections we chose the macroscopic sample with an particle ensemble in a shape of highly elongated cylinder (volume \( V \)), along the \( Oz \) axis of which an ac magnetic field \( \mathbf{H} = H \cos(\omega t)\mathbf{H} \) is applied, where \( H \) is the amplitude, \( t \) is the time, and \( \mathbf{H} = (0,0,1) \). In this case an internal macroscopic field inside the sample is equal to an external magnetic field \( \mathbf{H} \), and the interaction energy \( U_m(i) \) between \( i \)-th particle magnetic moment and the magnetic field could be written in Zeeman form:

\[ U_H(i) = -\mu_0 (\mathbf{m}_i \cdot \mathbf{H}) = -\mu_0 m H \cos(\omega t) \cos \theta_i, \]  

(4)

where \( \mu_0 \) stands for the vacuum magnetic permeability. The sketch of the sample is shown in Fig. 1.

The translational position of each \( i \)-th particle is defined by its radius-vector \( \mathbf{r}_i = r_i \hat{\mathbf{r}}_i \). The magnetic interaction of two \((i\text{-th and } j\text{-th})\) uniformly magnetized spherical particles with the center-center separation vector \( \mathbf{r}_{ij} = \mathbf{r}_i - \mathbf{r}_j = r_{ij} \hat{r}_{ij} \) is described by the dipole-dipole potential energy \( U_d(i,j) \)

\[ U_d(i,j) = -\frac{\mu_0 m^2}{4\pi r_{ij}^3} [3 (\mathbf{m}_i \cdot \hat{r}_{ij}) (\mathbf{m}_j \cdot \hat{r}_{ij}) - (\mathbf{m}_i \cdot \mathbf{m}_j)]. \]  

(5)

The rotational motion of the magnetic moment of the randomly chosen particle (with number 1, for example) is described by the probability density \( W(1) \), determined by the thermal fluctuations, dipole-field, and interparticle dipole-dipole interactions. The system under consideration possesses cylindrical symmetry and so the probability density does not depend on the angle \( \varphi \), i.e., it may be written as \( W(1) = W(t,x) \), \( x = \cos \theta \). This density is the solution of FPB equation.
\[
2\tau_D \frac{\partial W(1)}{\partial t} = \frac{\partial}{\partial x} \left[ (1 - x^2) \left( \frac{\partial W(1)}{\partial x} + W(1) \frac{\partial U(1)}{\partial x} \right) \right],
\]

where \( U(1) \) is the potential energy of the dipole with number 1 in units of the thermal energy \( k_B T \). \( W(1) \) satisfies the normalization condition

\[
\int_{-1}^{1} W(t,x) dx = 1.
\]

In the ideal, noninteracting case, the potential energy \( U(1) \) includes Néel energy of the magnetic anisotropy \( U_N(1) \) and the interaction energy \( U_H(1) \) between the magnetic moment and the magnetic field:

\[
U(1) = U_{id}(1) = \frac{U_N(1) + U_H(1)}{k_B T}.
\]

To take into account dipole-dipole interactions in the FPB Eq. (6) we follow earlier work [24], where single-particle energy \( U_{id}(1) \) was extended by incorporating dipole-dipole interactions on the basis of the first order modified mean-field model (MMF1)

\[
U(1) = U_{int}(1) = \frac{1}{k_B T} \left( U_N(1) + U_H(1) + \rho \langle U_d(1,2) W_{id}(2) \Theta(1,2) \rangle \right).
\]

Here the Heaviside step-function \( \Theta(1,2) \) describes the impenetrability of the two particles, and \( W_{id}(2) \) is the orientational probability for the particle 2 in the ideal (noninteracting) system. The angled brackets denote an averaging over all possible orientations and positions of the second ferroparticle. The meaning of the last term in Eq. (9) is clear: it is the overall magnetic field produced by all other magnetic dipoles; and this dipole field influences the orientation of the 1-st magnetic moment in addition to an external magnetic field.

The magnetization \( M \) of the ferroparticle ensemble is defined as the projection of the randomly chosen particle magnetic moment onto the magnetic field direction, weighted with the one-particle probability density \( W(1) \), averaged over all possible orientations:

\[
M(t) = \rho m \int d\hat{m}_1 (\hat{m}_1 \cdot \hat{H}) W(1) = \rho m \int_{-1}^{1} x W(1) dx.
\]

Dynamic susceptibility \( \chi \) is defined as the first term in the Fourier series of \( M(t) \)

\[
\chi(\omega) = \frac{\omega}{2\pi \hat{H}} \int_{0}^{2\pi} M(t) e^{i\omega t} dt.
\]

2.2. Theoretical approach for a small ac field amplitude

The theory of the dynamic magnetic response of an ensemble of immobilized superparamagnetic ferroparticles on the weak ac magnetic field has been developed recently in [32]. The theory is based on the FPB equation for the case when the particle easy magnetization axes are aligned in one direction and the ac field is oriented with some given angle to the easy axes. Following the article [32] the susceptibility of the noninteracting \( \chi^{id} \) and the interacting \( \chi \) particles for the system described above can be written in the form

\[
\chi^{id} = A_1^{id} \chi_L, \quad \chi = \chi_L A_1^{id} \left( 1 + \frac{\chi_L A_1^{id}}{3} \right), \quad \chi_L = \frac{\mu_0 \rho m^2}{3k_B T},
\]
where \( A_{1}^{id} \) is found from the solution of the set of equations

\[
\left[ 2\tau \omega + n(n + 1) - 2\sigma \frac{n(n + 1)}{(2n + 3)(2n - 1)} \right] A_{n}^{id} - 2\sigma \frac{(n - 1)n(n + 1)}{(2n - 3)(2n - 1)} A_{n-2}^{id} + 2\sigma \frac{n(n + 1)(n + 2)}{(2n + 3)(2n + 5)} A_{n+2}^{id} = (2n + 1) \left( \int_{-1}^{1} e^{\sigma x^2} dx \right)^{-1} \int_{-1}^{1} e^{\sigma x^2} (1 - \sigma + \sigma x^2) P_n(x) dx.
\]

with \( A_{n}^{id} \leq 0 \). Here \( P_n(x) \) is the Legendre polynomials. Explicit expressions for \( A_{1}^{id} \) can be determined by truncating at some arbitrary order \( n = k \), setting \( A_{n>k}^{id} = 0 \), and solving the set of \( k \) algebraic equations.

It is worth noting again that the formulae (12), (13) are valid only for small field amplitudes. Subsequently, they will be used to test numerical results in the limit of the weak ac fields.

2.3. Numerical approach

Approximation methods are usually used to solve Eq. (6) since the exact solution can be obtained only for a number of particular cases. There are several methods, and the first one is based on representing the solution as unknown function expanded in eigenfunctions of the differential operator [21, 40]. From the practical point of view, it is necessary to use a finite number of terms of the expansion and, as a rule, this number depends on parameters of the equation in an undetermined way. This means that the error of approximation can be underestimated. The second method is based on the numerical solution of the corresponding system of stochastic equations. In this paper the finite difference scheme suggested in [39] is used for the direct numerical solution of the Eq. (6). This is a fast way to construct an approximation. The proposed in [39] stable scheme is developed for solving convection-diffusion problems and can be used for solution of Eq. (6) because the first term in the right-hand side of Eq. (6) can be considered as a diffusion term and the second one is a convection term. The criteria of convergence of the numerical approximation to the solution were proved in [39]. The advantage of the method is numerical stability even in case of the convection term predominance in the FPB equation.

Eq. (6) is discretised on the uniform Cartesian grid in two-dimensional space \((t, x)\). Let \( h_t, h_x \) be the grid spacing in \( t \) and \( x \)-direction. The values \( W_{k,i} \) are values of unknown function \( W(t, x) \) at the grid nodes with the coordinates \( t_k = kh_t, x_i = -1 + (i + 0.5)h_x \), i.e., \( W_{k,i} = W(t_k, x_i) \). Indexes \( k \) and \( i \) vary from 0 to their final values \( N_t, N_x \) that are equal to \( N_t = T_f/h_t, N_x = 2/h_x - 1 \) correspondingly, where \( T_f \) is the final moment of the considered time interval for simulation.

According to [39] the Eq. (6) can be written in a discrete form

\[
\frac{\exp(-\delta h_t) W_{k,i} - W_{k-1,i}}{h_t} + (C_2 + D + \delta)(\exp(-\delta h_t) W_k(x)) = 0, \quad i = 0..N_x. \tag{14}
\]

\( D \) and \( C_2 \) are discrete operators for diffusion and convection terms accordingly. For operator \( D \) the standard second-order central difference is used

\[
DW_k(x) = \frac{1}{h_x^2} \left( f(x_i + \frac{h_x}{2}) (W_{k,i+1} - W_{k,i}) + f(x_i - \frac{h_x}{2}) (W_{k,i} - W_{k,i-1}) \right),
\]

where \( f(x) = (1 - x^2) \). Operator \( C_2 \) is constructed to satisfy stability criteria of numerical solution and is defined as follows.
\[
C_2W_k(x) = \frac{1}{2h_x}(v(t^*, x + 0.5h_x)(W_{k,i+1} + W_{k,i})) - \frac{1}{2h_x}v(t^*, x - 0.5h_x)(W_{k,i} + W_{k,i-1}),
\]

\[
t^* = 0.5(t_{n+1} + t_n), \quad v(x) = \frac{\partial U(1)}{\partial x}.
\]

In Eq. (14) \( \delta \) can be considered as a regularization parameter that is used to make numerical scheme unconditionally stable and it is equal to \( 0.5 \max |v'_x| \).

At every \( t = t_k \) Eq. (14) is linear algebraic systems with unknown variables \( W_{k,0}, W_{k,1}, ..., W_{k,N} \) that are found by the tridiagonal matrix algorithm. To satisfy the condition (7) the normalization is carried out

\[
W_{k,i}^{\text{norm}} = \frac{W_{k,i}}{h_x \sum_{i=0}^{N-1} W_{k,i}},
\]

therefore

\[
h_x \sum_{i=0}^{N-1} W_{k,i}^{\text{norm}} = 1.
\]

The last equation is condition (7) in discrete form, determined by the method of rectangles.

Magnetization \( M \) is calculated by integrating with the trapezoidal rule expression

\[
M(t_k) = \rho m \int_{-1}^{1} W_{k,i}^{\text{norm}}(t_k, x) x \, dx, \quad x \in \{x_i\},
\]

(15)

the real \( \text{Re}(\chi) \) and imaginary \( \text{Im}(\chi) \) part of susceptibility can be found by numerical integration over \( \{t_k\} \)

\[
\text{Re}(\chi) = \frac{\omega}{H\pi} \int_{0}^{2\pi} M(t) \cos(\omega t) dt, \quad \text{Im}(\chi) = \frac{\omega}{H\pi} \int_{0}^{2\pi} M(t) \sin(\omega t) dt, \quad t \in \{t_k\}.
\]

(16)

### 3. Results and Discussion

#### 3.1. Magnetization and susceptibility

Figure 2 shows numerical results of the magnetization curves as the functions of time for the system of immobilized particles with \( \chi_L = 1 \) at the ac field frequency \( \omega \tau_D = 1 \). There are two sets of lines in Fig.2, solid lines correspond to the noninteracting particles, and the dashed ones describe the interacting particles. An increase in the dimensionless ac field amplitude \( \alpha = \mu_0 mH/k_BT \), also known as the Langevin parameter, leads to growth of the magnetization modulus. This tendency is a result of growing orientational interactions between the magnetic moment and the ac field. A change in the magnetic anisotropy constant \( \sigma \) leads to a shift of the extrema of the time-dependent magnetization curves that indicates a transformation of the relaxation processes in the system.

In order to validate the numerical methodology of dynamic susceptibility calculation, the numerical tests were run at the small ac field amplitude \( \alpha = 0.01 \), where the theory (12) is reliable. The susceptibility spectra of three systems with \( \chi_L = 1 \) and \( \sigma = 0, 3, \) and 5 are shown in Fig. 3. The agreement between numerical data (symbols) and theory (lines) is excellent. Dashed lines and open symbols demonstrate the behavior of noninteracting particles. In the weak ac field the increase of the magnetic anisotropy constant
Figure 2. Normalized magnetization $M^*(t) = M(t)/\rho m$ of noninteracting (blue solid lines) and interacting (red dashed lines) particles with $\chi_L = 1$ at $\omega \tau_D = 1$ from numerical simulations. The Langevin parameters $\alpha$ and anisotropy constants $\sigma$ are (a) $\alpha = 1, \sigma = 0.1$, (b) $\alpha = 5, \sigma = 0.1$, (c) $\alpha = 1, \sigma = 3$, (d) $\alpha = 5, \sigma = 3$.

Figure 3. The susceptibility spectra of noninteracting (dashed lines and open symbols) and interacting (solid lines and filled symbols) particles with $\chi_L = 1$ at $\alpha = 0.01$. Lines correspond to analytical expression (12) from [32] obtained for small field amplitudes. Symbols are from numerical simulations. The anisotropy constants are $\sigma = 0$ (blue squares), $\sigma = 3$ (red triangles), $\sigma = 5$ (black circles). The coefficient $A_1^{id}$ in the dynamic susceptibilities (12) is calculated from the set of 10 algebraic equations (13).
\( \sigma \) results in the shift up of the susceptibility spectra and shift to lower frequencies \( \omega \tau_D \) of the maximum of the imaginary part of the susceptibility. These tendencies indicate that an increase of the internal magnetic anisotropy of particles leads to an increase of the magnetic moment relaxation time and a growth of the system magnetic response to the ac field. Note, that the latter is valid only for the model under consideration when the easy magnetization axes and the field are aligned. Interactions lead to the increases in the imaginary part of susceptibility (solid lines and filled symbols) and shift of the maximum to lower frequencies for all values of \( \sigma \). The shift signals the onset of dipolar nose-to-tail correlations and the concomitant increase in the characteristic rotation time. At low frequencies, dipole-dipole interactions increase the in-phase magnetic response of the system \( Re(\chi) \) whereas at moderate frequencies (0.1 \( \lesssim \omega \tau_D \lesssim 1 \) for the parameters from Fig.3) the real part of the susceptibility for noninteracting particles exceeds one for interacting particles. In this region, dipole-dipole interactions in combination with high particle magnetic anisotropy can block the response of the system to the ac field.

An increase in the magnetic response with increasing magnetic anisotropy of particles in a system of interacting immobilized superparamagnetic particles is also observed in the static case. This effect was studied in [41]. It was shown that the initial magnetic susceptibility of interacting superparamagnetic particles, the easy magnetization axes of which are directed along the \( Oz \) axis, increases from \( \chi = \chi_L(1 + \chi_L/3) \) for \( \sigma = 0 \) to \( \chi = 3\chi_L(1 + \chi_L) \) for \( \sigma \to \infty \). This behavior was explained in [41] in terms of the long-range nature of the dipole–dipole interaction and the role of orientational averaging. With large energy barriers (\( \sigma \gg 1 \)), the magnetic moments appear to be strongly correlated, which results in a strong enhancement of the magnetic response.

The susceptibility spectra for three systems with \( \chi_L = 0.1, 1 \) and \( \sigma = 1 \) at \( \alpha = 3 \) are shown in Fig. 4. Dashed lines correspond to the noninteracting system, solid lines are interacting immobilized superparamagnetic particles. For low concentrated sample with small value of the Langevin susceptibility \( \chi_L = 0.1 \) the solid lines coincide with the dashed ones that indicate the insignificant role of interparticle interactions in the system. In moderately concentrated samples with the Langevin susceptibility \( \chi_L = 1 \) and 1.5 the dipole-dipole interactions have a considerable effect on the dynamic spectrum. As the Langevin parameter \( \chi_L \) is increased, the susceptibility shifts upward for both interacting and noninteracting particles reflecting the growth of the magnetic response of the sample. The position of the maximum of \( Im(\chi) \) for noninteracting particles remains unchanged with increasing \( \chi_L \), while for interacting particles a small shift of the maximum to the left is observed. This is because the dipolar nose-to-tail correlations lead to the increase in the characteristic rotation time whereas the relaxation time of the magnetic moment of noninteracting particles does not depend on the \( \chi_L \).

Figure 5 shows the susceptibility spectra for system with \( \chi_L = 0.5 \) and \( \sigma = 1 \) at different values of ac field amplitude \( \alpha = 0.1, 1 \) and 5. At low to moderate frequencies, both real and imaginary parts of susceptibility decrease with increasing field strength. This shows that there is an additional orientational constraint arising in the strong field from the nose-to-tail dipolar correlations which reduces the susceptibility. As the field strength is increased, the peak in the imaginary part of susceptibility shifts to higher frequencies. For \( \alpha = 5 \), the imaginary part reaches the maximum at a frequency \( \omega \tau_D > 1 \), it means that in the strong ac field the relaxation processes proceed faster than in an ideal system of immobilized magnetic particles. At high frequencies, the magnetic response of the immobilized superparamagnetic particles is almost independent of the ac field amplitude. Note that the decrease in the susceptibility with increasing external field is also observed in a static case. Moreover, this behavior is quite natural from a mathematical point of view: the static magnetic susceptibility can be defined as a derivative of the magnetization \( dM/dH \). An inherent property of the static magnetization curve \( M(H) \) is its upward convexity. Hence \( d^2M/d^2H < 0 \), then \( dM/dH \)
Figure 4. The susceptibility spectra of noninteracting (dashed lines) and interacting (solid lines) particles with $\sigma = 1$ at $\alpha = 3$ from numerical simulations. The interaction parameters are $\chi_L = 1.5$ (curves a), $\chi_L = 1$ (curves b), $\chi_L = 0.1$ (curves c).

is a decreasing function of $H$. Thus, the concave property of the static magnetization curve guarantees that the static susceptibility decreases with increasing field.

Figure 5. The susceptibility spectra of interacting particles with $\chi_L = 0.5$ and $\sigma = 1$ from numerical simulations. The Langevin parameters are $\alpha = 0.1$ (curves a), $\alpha = 1$ (curves b), $\alpha = 5$ (curves c).

3.2. Relaxation times

The position of the maximum $\omega_{\text{max}}\tau_D$ of the imaginary part of the susceptibility determines the characteristic relaxation time $\tau$ of the particle magnetic moment in an ac magnetic field. For an ideal system of immobilized superparamagnetic particles in the weak ac field when there is no intraparticle energy barrier, the characteristic relaxation time of the magnetic moment is $\tau = \tau_D = 1/\omega_{\text{max}}$. An increase in the field amplitude, the intensity of the interparticle interactions, and internal magnetic anisotropy of particles lead to a slowdown or acceleration of relaxation processes; the value of the position of the maximum of the imaginary part of the susceptibility determines the following ratio of characteristic relaxation times: $\omega_{\text{max}}\tau_D = \tau_D/\tau$. Thus, a shift of the position of the maximum from 1 demonstrates the changes in relaxation processes in comparison
with the ideal system. Fig. 6 shows the position of the maximum of the imaginary part of the susceptibility as a function of the field amplitude $\alpha$ and magnetic anisotropy constant $\sigma$. The left panel shows a system of noninteracting particles, the right one demonstrates a system with $\chi_L = 0.5$ in which the interparticle dipole-dipole interactions are taken into account. An increase in $\alpha$ decreases the characteristic relaxation time of the particle magnetic moment, while an increase in $\sigma$ leads to an increase in the relaxation time. This behavior can be qualitatively explained by analyzing the potential energy of the particles. It is energetically favorable for magnetic moments to orient themselves so that a minimum of potential energy is achieved; this orientation coincides with the direction of the field $\cos \theta_i = 1$ for noninteracting particles. Therefore, in a dynamic system, magnetic moments, which are directed opposite to the applied field $\cos \theta_i = -1$ have the longest relaxation time. In thermodynamic equilibrium and when $\cos \theta_i = -1$ the particle energy (8) is determined by $U(i) = -\sigma + \alpha$. With increasing $\alpha$, the energy increases, while increasing $\sigma$ leads to a decrease in the energy. Therefore, with an increase in $\alpha$, the orientation of the magnetic moment $\cos \theta_i = -1$ becomes more and more disadvantageous in terms of energy. So, the larger $\alpha$, the faster the magnetic moment deviates from the orientation $\cos \theta_i = -1$, hence relaxation processes are accelerated. An increase in $\sigma$ has the opposite effect, slowing down the relaxation processes.

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{figure6.png}
\caption{Contour plots of the position of the maximum of the imaginary part of susceptibility depending on the field amplitude $\alpha$ and magnetic anisotropy constant $\sigma$. The left panel corresponds to noninteracting system, the right panel takes into account interparticle dipole-dipole interactions, $\chi_L = 0.5$. The plots were calculated numerically with $h_i = h_x = 0.01$ for every $\alpha$ and $\sigma$ from $\{(\alpha_p, \sigma_q) : \alpha_p = 0.02p, \sigma_q = 0.02q, p = 0\ldots250, q = 0\ldots250\}$.}
\end{figure}

Fig. 7 shows the dependence of the ratio of characteristic relaxation times $\tau/\tau_D$ at $\alpha = 0.01, 1.5, 3,$ and 5 for a system of noninteracting particles. The dashed line corresponds to the theoretical expression (2), the solid lines are from numerical simulations. The theoretical expression (2) is in good agreement with the numerical result calculated for $\alpha = 0.01$. It emphasized that Eq. (2) is reliable for a system of noninteracting particles in a weak ac field. With an increase in the magnetic field amplitude, the characteristic relaxation time of the magnetic moment decreases, and in the region of the parameters $\alpha \gtrsim 3$ and $0 \leq \sigma \lesssim 1.5$ (taken separately in insert), the relation $\tau < \tau_D$ is valid, i.e., the relaxation of the magnetic moment in the system under consideration occurs faster than in the ideal system of immobilized superparamagnetic particles without
intraparticle energy barrier. Interparticle dipole-dipole interactions lead to a significant increase in relaxation times at small field amplitudes ($\alpha \lesssim 3$) and have almost no effect on relaxation processes at large field amplitudes ($\alpha \gtrsim 3$)(Fig. 8).

**Figure 7.** The ratio $\tau/\tau_D$ for the noninteracting particles as a function of anisotropy constant $\sigma$ at the Langevin parameters $\alpha = 0.01$ (curve a), 1.5 (curve b), 3 (curve c), and 5 (curve d). Solid lines are from numerical simulations, the dashed line is analytical expression (2) from [18]. Insert shows a zoom of the region $0 \leq \sigma \leq 2.5$ of numerical results for $\alpha = 3, 5$ and the theory (2).

**Figure 8.** The same as in Fig. 7 for the system of interacting particles with $\chi_L = 1$. The dashed line corresponds to the noninteracting particles (2) as in Fig. 7.

**Fig. 9** shows the ratio of characteristic relaxation times $\tau/\tau_D$ of the systems with different values $\chi_L = 0.5, 1$ at $\alpha = 0.01, 3$ and $0 \leq \sigma \leq 5$. For the strong magnetic field the interactions between magnetic moments and the field dominate over the interparticle interactions therefore the relaxation times do not depend on $\chi_L$; only the value of magnetic field and internal magnetic anisotropy affect the relaxation processes in the system (Fig. 9 (b)). In a weak field, an increase in $\chi_L$ leads to an increase in relaxation times (Fig. 9 (a)).
Figure 9. The ratio $\tau/\tau_D$ as a function of anisotropy constant $\sigma$ at the Langevin susceptibility $\chi_L = 0.5$, 1 and for the noninteracting particles. The Langevin parameters are (a) $\alpha = 0.01$ (b) $\alpha = 3$.

4. Conclusions

A numerical study of the magnetic response of immobilized interacting superparamagnetic particles in the ac field of arbitrary amplitude was carried out in the article. The modeling was based on the FPB equation, which was solved using the finite difference scheme proposed in [39]. To solve the problem studied in the article, the method [39] was modified, namely instead of boundary conditions the normalization relation for the required function was used. The magnetization, susceptibility, and relaxation time were numerically investigated. It was shown that an increase in the amplitude of the alternating field contributes to a decrease in susceptibility, but accelerates relaxation processes that occur in the system. With an increase in the internal magnetic anisotropy of the particles, the susceptibility of the system increases, but the relaxation times of the magnetic moments also increase. For sufficiently large field amplitudes and small values of the magnetic anisotropy constant ($\alpha \gtrsim 3$ and $0 \leq \sigma \lesssim 1.5$), the relaxation time of interacting superparamagnetic particles is less than $\tau_D$ – the characteristic relaxation time of an ideal system. It should be emphasized, that the conclusions above are valid only for the system under consideration, in which the axes of easy magnetization of particles are aligned with the field. Dipole-dipole interactions have a significant effect on magnetic and relaxation processes in systems in a weak field, but in a moderate and strong magnetic field, relaxation processes are determined only by the magnitude of the magnetic field and internal magnetic anisotropy of particles.

For small field amplitudes, the numerical results were tested on known theoretical data and showed their consistency. There are no theoretical data on the dynamic magnetic properties for immobilized interacting superparamagnetic particles in a field of arbitrary amplitude in the scientific literature, therefore, the numerical results obtained in this paper fill this gap. The investigation of the behavior of immobilized interacting superparamagnetic particles in a field of arbitrary amplitude is of practical importance and can be applied to design systems with specified properties and use them in novel technologies.
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References


