Influence of a bias dc field and an ac field amplitude on the dynamic susceptibility of a moderately concentrated ferrofluid

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In this paper, we study the effect of a bias dc field on the dynamic response of a moderately concentrated ferrofluid to an ac magnetic field of arbitrary amplitude. The ferrofluid is modeled by an ensemble of interacting moving magnetic particles; the reaction of particle magnetic moments to ac and dc magnetic fields occurs according to the Brownian mechanism; and the ac and dc magnetic fields are parallel. Based on a numerical solution of the Fokker-Planck equation for the probability density of the orientation of the magnetic moment of a random magnetic particle, dynamic magnetization and susceptibility are determined and analyzed for various values of the ac field amplitude, the dc field strength, and the intensity of dipole-dipole interactions. It is shown that the system's magnetic response is formed under the influence of competing interactions, such as dipole-dipole, dipole-ac field, and dipole-dc field interactions. When the energies of these interactions are comparable, unexpected effects are observed: the system's susceptibility can either increase or decrease with increasing ac field amplitude. This behavior is associated with the formation of nose-to-tail dipolar structures under the action of the dc field, which can hinder or promote the system's dynamic response to the ac field. The obtained results provide a theoretical basis for predicting the dynamic properties of ferrofluids to improve their use in biomedical applications, such as, in magnetic induction hyperthermia.

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I. INTRODUCTION

Ferrofluid is a stable colloidal suspension of one-domain magnetic particles in carrier liquids [1–3]. A ferrofluid's ability to react to a magnetic field while remaining in a liquid state has led to a variety of applications [4–11], such as, targeted drug delivery, enhancing the contrast of magnetic resonance imaging, localized heating (hyperthermia), and the destruction of diseased tissue.

In an ac magnetic field $H_{ac}\cos(\omega t)$ with an amplitude H_{ac} and an angular frequency ω the ferrofluid is magnetized. Its dynamic magnetization can be written as

$$M(t)=\sum_{k=0}^{\infty} M_k'\cos(k\omega t)+M_k''\sin(k\omega t), \tag{1}$$

where t is time. The coefficients M_k' and M_k'' depend on the amplitude of the ac field and determine the kth harmonic of magnetization $\chi_k=\chi_k'+i\chi_k''$ ($\chi_k'=M_k'/H_{ac}^k$, $\chi_k''=M_k''/H_{ac}^k$), the signal of which can be filtered in the experiment. In a weak alternating magnetic field, the system's magnetic response is completely described by the first harmonic $\chi_1=\chi$, which is called dynamic susceptibility. In ac fields with a high amplitude, the higher harmonics become significant and their signal is registered in experiments.

A great variety of theoretical approaches are available to describe the dynamic susceptibility of ferrofluids. Most theoretical papers deal with an ideal system of noninteracting magnetic particles. The earliest dynamic theory of dipolar fluids was developed by Debye [12] for an ensemble of noninteracting dipolar particles under a weak ac field. Expressions for the Brownian relaxation time of the magnetic particles were derived in Refs. [13,14]. The influence of the ac field's amplitude on dynamic susceptibility and nonlinear harmonics has been studied in [14–19]. Yoshida and Enpuku [20], based on a numerical solution of the Fokker-Planck equation, have proposed simple approximation formulas for the dynamic susceptibility and third harmonic of moving noninteracting magnetic particles, which can be used for strong ac fields with an amplitude up to $\alpha_{ac}=20$ (α_{ac} is the Langevin parameter). The dc field's influence on the dynamic response and relaxation characteristics of an ensemble of moving noninteracting particles was investigated [21–23]. Zhong *et al.* [24], based on experimental data, have proposed a formula for describing the Brownian relaxation time of magnetic nanoparticles, that takes into account the relative location of the ac and dc magnetic fields. Single-particle theories of the dynamic response of an ensemble of moving noninteracting particles allow us to qualitatively describe the behavior of a ferrofluid in ac and dc fields. However, the range of applicability of these theories is limited to weakly concentrated and weakly interacting systems.

The main difficulty in predicting the properties of real ferrofluids is taking into account dipole-dipole interactions and polydispersity. The effect of polydispersity on the magnetic properties of ferrofluid has been studied in [25–31]. The influence of dipole-dipole interactions on the dynamic susceptibility has been investigated in the works [26,32–38]. Ivanov *et al.*, using the strict methods of classical statistical mechanics, have obtained an analytical expression for a dynamic response of an ensemble of interacting magnetic

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particles to a weak ac magnetic field [39]. The dependence of the dynamic response of interacting particles on the ac field's amplitude has been investigated in [40–42]. Analytical expressions are proposed for dynamic susceptibility, nonlinear harmonics, and Brownian relaxation times of magnetic particles. The dynamic response of interacting particles in a bias dc field was theoretically studied only for the case of weak ac fields [43,44].

In this paper, the dynamic response of an ensemble of moving interacting magnetic particles in a bias dc field will be studied in the case of arbitrary amplitudes of ac fields: the ac and dc magnetic fields are directed in parallel. As already noted, in strong ac fields, a nonlinear response (the higher harmonics χ_k, $k > 1$), makes a nonzero contribution to the system's magnetization. In this article, we will focus on analyzing the effect of competing interactions—dipole-dipole, dipole-ac field, and dipole-dc field—only on the first harmonics, which is a linear magnetic response of an ensemble of interacting particles.

II. MODEL AND METHODS

A. Ferrofluid model

The ferrofluid is modeled as a suspension of N hard spherical and uniformly magnetized particles with equal diameters d and dipole moments m, immersed in a structureless fluid with viscosity η at temperature T and total volume V. The short-range interactions are given by the hard sphere form

$$U_s(ij) = \begin{cases} \infty, & r_{ij} < d \\ 0, & r_{ij} \geqslant d \end{cases}, \tag{2}$$

where r_{ij} is the distance between the centers of i and j particles. The dipole-dipole interaction is modeled by the potential $U_\mathrm{d}(ij)$,

$$U_\mathrm{d}(ij) = \frac{\mu_0 m_i m_j}{4\pi r_{ij}^3}[(\hat{\mathbf{m}}_i \cdot \hat{\mathbf{m}}_j) - 3(\hat{\mathbf{m}}_i \cdot \hat{\mathbf{r}}_{ij})(\hat{\mathbf{m}}_j \cdot \hat{\mathbf{r}}_{ij})], \tag{3}$$

where μ_0 is the vacuum permeability, $\mathbf{m}_i = m\hat{\mathbf{m}}_i = m(\sin\theta_i\cos\phi_i, \sin\theta_i\sin\phi_i, \cos\theta_i)$ is the dipole moment of the particle i, $\mathbf{r}_{ij} = \mathbf{r}_i - \mathbf{r}_j = r_{ij}\hat{\mathbf{r}}_{ij}$ is the vector connecting the centers of particles i and j, $\hat{\mathbf{r}}_{ij}$ is the unit vector of $\mathbf{r}_{ij}$, and $\mathbf{r}_i = r_i(\sin\xi_i\cos\psi_i, \sin\xi_i\sin\psi_i, \cos\xi_i)$ is the radius-vector of ith particle position. The strength of the dipole-dipole interactions is characterized by the dipolar coupling constant

$$\lambda = \frac{\mu_0 m^2}{4\pi d^3 k_B T}, \tag{4}$$

where k_B is Boltzmann's constant. The particle number concentration is $\rho = N/V$; the volume particle concentration is $\varphi = \rho\pi d^3/6$. It is assumed that the particles are in a thermodynamically equilibrium state, which for the case of the absence of an external magnetic field and weak interparticle interactions corresponds to a uniform random distribution of particles in the volume V. The Langevin susceptibility $\chi_L = 8\varphi\lambda$ is used for a complex description of the sample density and the intensity of interparticle dipole-dipole interactions.

In order to avoid demagnetization effects, it is assumed that the magnetic particles are placed in a long cylindrical container with a height directed along the Oz axis. Note that

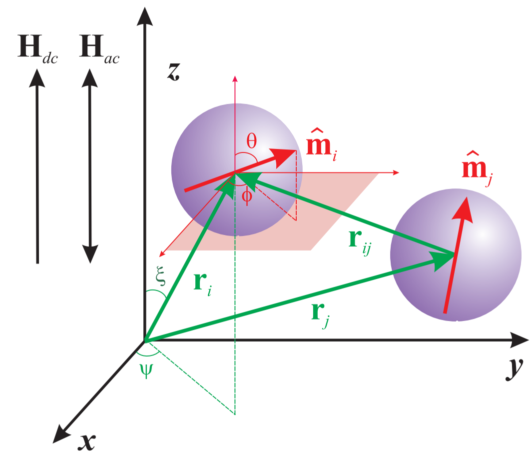

FIG. 1. Laboratory coordinate system.

this is not a significant limitation, since the demagnetizing field can be taken into account using the demagnetizing factor. An ac magnetic field $\mathbf{H}_{ac} = H_{ac}\cos(\omega t)\hat{\mathbf{H}}$ and a dc magnetic field $\mathbf{H}_{dc} = H_{dc}\hat{\mathbf{H}}$ are applied parallel to Oz axis, where H_{ac} is the amplitude of the ac field, H_{dc} is the dc field intensity, $\hat{\mathbf{H}} = (0, 0, 1)$, t is the time, and ω is an angular frequency of the ac field. In this case, the internal macroscopic field inside the sample and the external magnetic field are the same $\mathbf{H} = \mathbf{H}_{ac} + \mathbf{H}_{dc}$. This model system is shown in Fig. 1.

The interaction $U_H(i)$ between ith magnetic moment and the external magnetic fields can be written in a Zeeman form

$$U_H(i) = -\mu_0(\mathbf{m}_i \cdot \mathbf{H}) = -\mu_0 m\cos\theta_i(H_{ac}\cos(\omega t) + H_{dc}). \tag{5}$$

The strength of the dipole-field interaction can be characterized by the Langevin parameters

$$\alpha_{ac} = \frac{\mu_0 m H_{ac}}{k_B T}, \quad \alpha_{dc} = \frac{\mu_0 m H_{dc}}{k_B T}. \tag{6}$$

For the case of noninteracting magnetic particles, the potential energy of the system in units of $k_B T$ is

$$U(i) = U_{id}(i) = \frac{U_H(i)}{k_B T}. \tag{7}$$

For interacting particles, we use the approximation of the first-order modified mean-field theory (MMF1) suggested in [45]:

$$U(i) = U_\mathrm{int}(i) = \frac{1}{k_B T}(U_H(i) + \rho\langle U_d(ij)\Theta(ij)\rangle_j), \tag{8}$$

where the Heaviside step-function $\Theta(ij)$ describes the impenetrability of two particles, which is given in the model by hard-sphere potential (2); the angle brackets denote the statistical averaging over the positions of particle j and its magnetic moment orientations:

$$\langle\cdots\rangle_j = \int d\mathbf{r}_j d\hat{\mathbf{m}}_j W^{id}(j)\ldots,$$

$$\int d\hat{\mathbf{m}}_j = \frac{1}{4\pi}\int_{-1}^{1} d\cos\theta_j \int_0^{2\pi} d\phi, \tag{9}$$

$$\int d\mathbf{r}_j = \lim_{R\to\infty}\int_0^{2\pi} d\psi_j \int_{-1}^{1} d\cos\xi_j \int_0^{R/\sin\xi_j} r_j^2 dr_j, \tag{10}$$

where integration weight function $W^{id}(j)$ is the orientational probability for the magnetic moment of particle j in an ideal (noninteracting) system. Note that the integration (10) in the expression (8) assumes that the translational degrees of freedom of the particles are not limited by anything, except for the condition of mutual nonpenetration, that is, a particle can be located at an arbitrary point of the volume V at any moment of time.

B. Numerical solution of the Fokker-Planck equation

The considered system has spherical symmetry, so the probability distribution function of magnetic moment orientations $W(i)$ does not depend on the azimuth angle ϕ_i. To define $W(i) = W(t, x)$, $x = \cos\theta_i$ the Fokker-Plank equation (FPE) is used

$$2\tau_B \frac{\partial W(i)}{\partial t} = \frac{\partial}{\partial x}\left[(1-x^2)\left(\frac{\partial W(i)}{\partial x} + W(i)\frac{\partial U(i)}{\partial x}\right)\right], \quad (11)$$

where τ_B is the Brownian rotational time given by

$$\tau_B = \frac{\pi d^3 \eta}{2k_B T}. \quad (12)$$

In the absence of interparticle interactions, the potential energy $U(i)$ is simply

$$U(i) = U_{id} = -x(\alpha_{ac}\cos(\omega t) + \alpha_{dc}). \quad (13)$$

For interacting particles, considering Eqs. (9) and (10), the potential energy (8) takes the form

$$U(i) = U_{\text{int}}(i) = -x\left(\alpha_{ac}\cos(\omega t) + \alpha_{dc} + \frac{\chi_L}{2}\int_{-1}^{1} W^{id}(j)x_j dx_j\right), \quad x_j = \cos\theta_j, \quad (14)$$

where $W^{id}(j)$ is the probability density of the magnetic moment orientation of particle j in the ideal system. The FPE (11) was solved numerically. For this an unconditionally stable finite-difference scheme was used, which is suggested in [46] for the ensemble of immobilized interacting magnetic particles in applied ac field. The outline for the calculation of the function $W(i)$ for interacting particles is the following:

(i) The FPE (11) is solved numerically for a system of noninteracting particles with potential energy (13). As a result, the function $W^{id}(i)$ is found numerically.

(ii) $W^{id}(i)$ is used to determine the system's potential energy with interaction (14).

(iii) The FPE (11) is solved numerically again, using the potential energy from the previous step (14). The result is a numerical function $W(i)$, which takes into account the interparticle interactions within the framework of MMF1.

Equation (11) is solved numerically for each value of $\omega\tau_B \in (10^{-3}, 10^3)$. A feature of the realization of the numerical scheme from [46] in this work is the dynamic calculation of the time step h_t depending on the frequency of the ac field: the higher the field frequency ω, the lower h_t. This method makes it possible to obtain the convergence of the numerical solution for a high value of the dc field strength at lower computation costs. A 2D grid was used:

$$\left\{(t_k, x_i) \mid t_k = t_{k-1} + h_t,\ x_i = x_{i-1} + h_x,\ x_0 = -1 + \frac{h_x}{2},\ t_0 = 0,\ h_x = 0.01,\ h_t = \frac{2\pi}{\omega}10^{-4}\right\}.$$

This calculation was performed for the time interval $t \in [0, 4\pi/\omega]$. It should be noted that $W(i)$ at small time ($t \sim 0$) "remembers" the initial conditions, which were chosen as $W(t=0) = 1/2$. In the second period $t \in [2\pi/\omega, 4\pi/\omega]$, $W(i)$ "forgets" about the initial conditions and demonstrates the thermodynamically equilibrium behavior of the system. For this reason, the values $W(i)$ determined numerically in the second period $t \in [2\pi/\omega, 4\pi/\omega]$ were used to calculate the system's dynamic magnetic properties, such as magnetization and susceptibility. Magnetization $M(t)$ is calculated using $W(i)$ as follows:

$$M(t) = \rho m \int d\hat{\mathbf{m}}_i(\hat{\mathbf{m}}_i \cdot \hat{\mathbf{H}})W(i) = \frac{\rho m}{2}\int_{-1}^{1} xW(i)dx. \quad (15)$$

Dynamic susceptibility χ is defined as the first term in the Fourier series of $M(t)$:

$$\chi(\omega) = \frac{\omega}{2\pi H}\int_{\frac{2\pi}{\omega}}^{\frac{4\pi}{\omega}} M(t)e^{i\omega t}dt. \quad (16)$$

III. RESULTS AND DISCUSSION

A. Dynamic magnetization

Figure 2 shows the dynamic magnetization of interacting moving magnetic particles in the dc and ac magnetic fields. The figures present results for two dimensionless ac field frequencies: $\omega\tau_B = 0.1$ and $\omega\tau_B = 1$ and various combinations of values of α_{ac} and α_{dc}. At high frequencies $\omega\tau_B = 1$, the hysteresis loops are wider than at low frequencies $\omega\tau_B = 0.1$, which indicates a significant delay in the reaction of magnetic moments to the magnetic field rate at $\omega\tau_B = 1$. In the absence of dc fields, the hysteresis loops are symmetric with respect to the origin. An increase in the dc field's intensity leads to an upward shift of the hysteresis loop, narrowing, and loss of symmetry. This behavior is explained by the fact that the dc field orients and holds the magnetic moments in the dc field's direction, increasing the system's magnetization. The magnetization reaches maximum value when the ac and dc fields are co-aligned.

The effect of the dipole-dipole interaction on dynamic magnetization is shown in Fig. 3. With the increasing intensity of the ac and dc fields, the contribution of the dipole-dipole interaction to magnetization decreases. In general, the

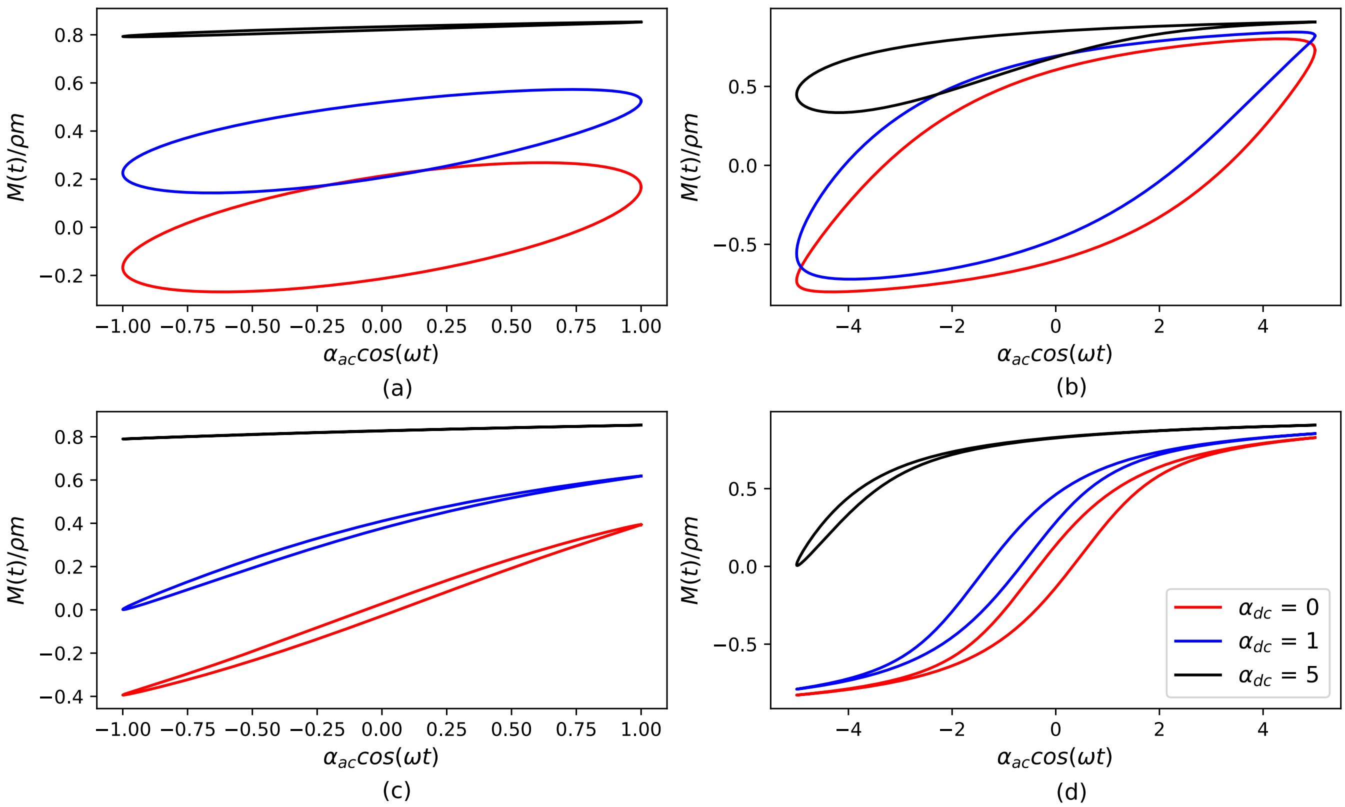

FIG. 2. Dynamic hysteresis loop of an ensemble of moving interacting magnetic particles with $\chi_L = 1$; the results are presented for four sets of frequencies $\omega\tau_B$ and ac field amplitudes α_{ac} (a) $\omega\tau_B = 1, \alpha_{ac} = 1$, (b) $\omega\tau_B = 1, \alpha_{ac} = 5$, (c) $\omega\tau_B = 0.1, \alpha_{ac} = 1$, (d) $\omega\tau_B = 0.1, \alpha_{ac} = 5$ for different values of the bias dc field $\alpha_{dc} = 0$ (red), $\alpha_{dc} = 1$ (blue), $\alpha_{dc} = 5$ (black).

characteristic behavior of the dynamic magnetization of an ensemble of interacting and noninteracting particles is similar: in the absence of the dc field, the hysteresis loop is symmetrical with respect to the origin; an increase in the intensity of the dc field leads to an increase in susceptibility, narrowing and the loss of the hysteresis loop's symmetry.

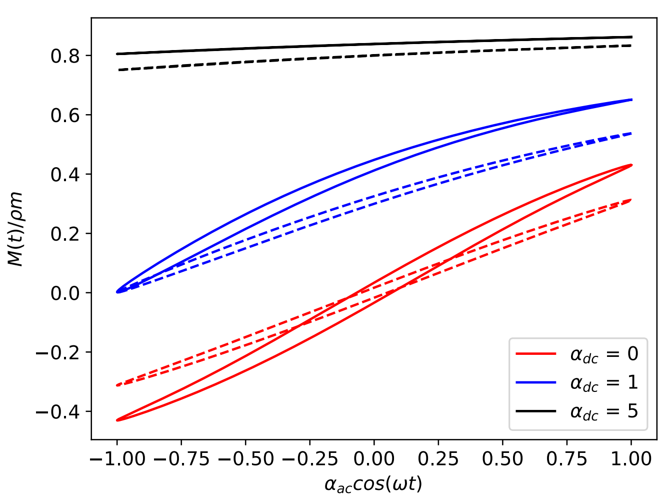

FIG. 3. The effect of the dipole-dipole interaction on the dynamic hysteresis loop of an ensemble of moving magnetic particles with $\chi_L = 1.5$ in an ac field with frequency $\omega\tau_B = 0.1$, amplitude $\alpha_{ac} = 1$ and a bias dc field $\alpha_{dc} = 0$ (red), $\alpha_{dc} = 1$ (blue), $\alpha_{dc} = 5$ (black). Solid lines are used for the system with interactions; dashed lines correspond to the system without interactions.

The results obtained in this section resemble the dynamic hysteresis loops for the system of immobilized noninteracting particles with uniaxial magnetic crystallographic anisotropy [47]. This is because the uniaxial anisotropy term has sometimes the same effect as a mean field interaction energy [48].

B. Susceptibility at low frequencies of the ac field

Experimental data and the results of theoretical studies show that if only the ac magnetic field reacts to the ferrofluid, then an increase in the ac field's amplitude reduces its dynamic magnetic response at low frequencies [20,40,42,49]. It is also known that the addition of a bias dc field directed parallel to the weak ac field reduces the ferrofluid's dynamic susceptibility in the low frequency region [24,43]. On the other hand, dipole-dipole interactions increase the ferrofluid's dynamic response in a weak ac magnetic field [50]. If high intensity ac and dc magnetic fields act simultaneously on a concentrated ferrofluid competing dipole-dipole, dipole-ac field, and dipole-dc field interactions occur in the system that lead to nonmonotonic effects in the magnetic response. In this section, the magnetic susceptibility of an ensemble of interacting particles to an ac field at $\omega \to 0$ for different values of the ac field's amplitude and the dc field's intensity are analyzed.

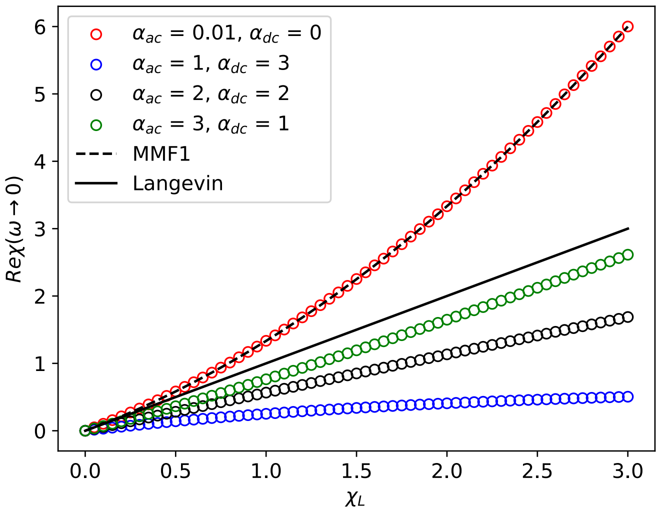

FIG. 4. Dependence of dynamic susceptibility at $\omega \to 0$ on the Langevin susceptibility χ_L. The points correspond to numerical results for $\alpha_{ac} = 0.01$ and $\alpha_{dc} = 0$ (red dots); $\alpha_{ac} = 1$ and $\alpha_{dc} = 3$ (blue dots); $\alpha_{ac} = 2$ and $\alpha_{dc} = 2$ (black dots); $\alpha_{ac} = 3$ and $\alpha_{dc} = 1$ (green dots). The black dashed line corresponds to MMF1 [51]; the black solid line corresponds to the Langevin theory.

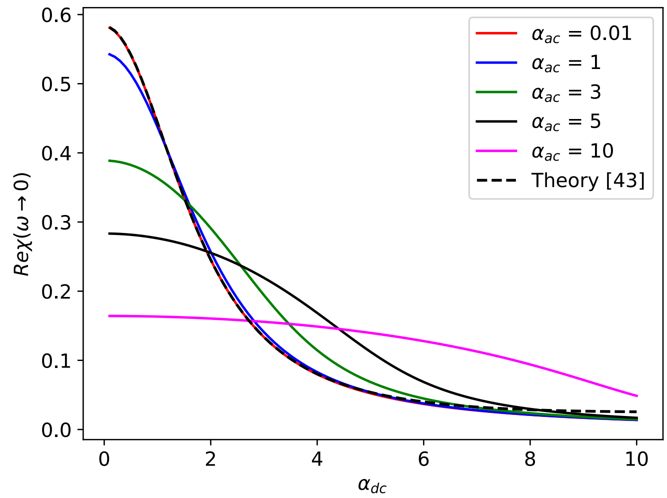

FIG. 5. The effect of the intensity of the bias dc field on the susceptibility of interacting particles with $\chi_L = 0.5$ at low frequencies of the ac field with the amplitude $\alpha_{ac} = 0.01$ (red solid line), $\alpha_{ac} = 1$ (blue solid line), $\alpha_{ac} = 3$ (green solid line), $\alpha_{ac} = 5$ (black solid line), $\alpha_{ac} = 10$ (purple solid line). Solid lines denote numerical results, dashed lines are used for theory [43].

Figure 4 shows the dependence of the dynamic susceptibility of magnetic particles at low frequencies on the Langevin susceptibility. The black solid line corresponds to the Langevin theory $\chi = \chi_L$, which describes the static initial magnetic susceptibility of noninteracting particles. The black dashed line shows the static theory MMF1 [51] $\chi = \chi_L(1 + \chi_L/3)$, valid for interacting particles. The numerical values of dynamic susceptibility determined in this paper in the region $\omega \to 0$ (red dots) at $\alpha_{ac} = 0.01$ and $\alpha_{dc} = 0$ for interacting particles are in strong agreement with the static theory of interacting particles MMF1. The blue, green, and black dots correspond to the numerical results of the dynamic susceptibility of interacting particles in the $\omega \to 0$ region, determined not for weak ac and dc fields and such that $\alpha_{ac} + \alpha_{dc} = 4$. An increase in the intensity of the ac and dc fields leads to a decrease in the system's magnetic response in the low frequency region, and the resulting susceptibility values for the given parameters lie below the Langevin curve. This behavior is explained by the fact that the reaction of magnetic moments to the ac field is hindered by a sufficiently strong dipole-dc field interaction, which leads to a decrease in the system's magnetic response to the ac field. Note that in all cases, an increase in χ_L (which means an increase in interparticle interactions) leads to an increase in the system's magnetic response.

Dependence of dynamic susceptibility at low frequencies on α_{dc} for interacting particles with $\chi_L = 0.5$ at $\alpha_{ac} =$ 0.01, 1, 3, 5, and 10 is shown in Fig. 5. The solid lines correspond to numerical results, while the dashed line shows theory [43] developed for the case of a weak ac field. The numerical results defined for $\alpha_{ac} = 0.01$ (red line) agree well with theory [43]. When $\alpha_{ac} > \alpha_{dc}$ the system's susceptibility changes slightly with the growth of α_{dc}. A sharp decline in susceptibility is observed when the values of α_{dc} begin to exceed α_{ac}. In such a system, the dipole-dc field interactions begin to dominate, the magnetic moments of the particles are held by the strong dc field and their response to the ac field becomes weak.

Figure 6 shows the dynamic susceptibility of interacting particles with $\chi_L = 0.5$ at low frequencies depending on α_{ac}. Solid lines correspond to the numerical results for interacting particles, dot-and-dash lines are numerical results for noninteracting particles, and the dashed line is the approximation formula from [40]. In the absence of the dc field, the

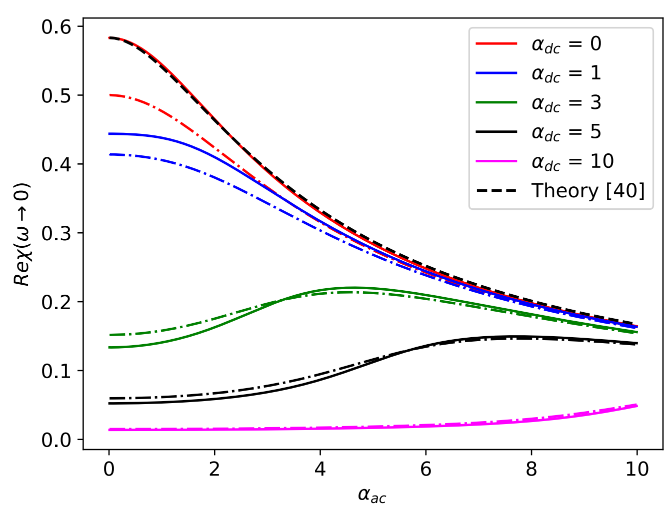

FIG. 6. The effect of the amplitude of the ac field at low frequencies ($\omega \to 0$) on the susceptibility of an ensemble of interacting (solid lines) and noninteracting (dot-and-dash lines) magnetic particles with $\chi_L = 0.5$, under the influence of the dc magnetic field with intensity: $\alpha_{dc} = 0$ (red), $\alpha_{dc} = 1$ (blue), $\alpha_{dc} = 3$ (green), $\alpha_{dc} = 5$ (black), $\alpha_{dc} = 10$ (purple). Solid and dot-and-dash lines denote numerical results, dashed line is used for the approximation formula from [40].

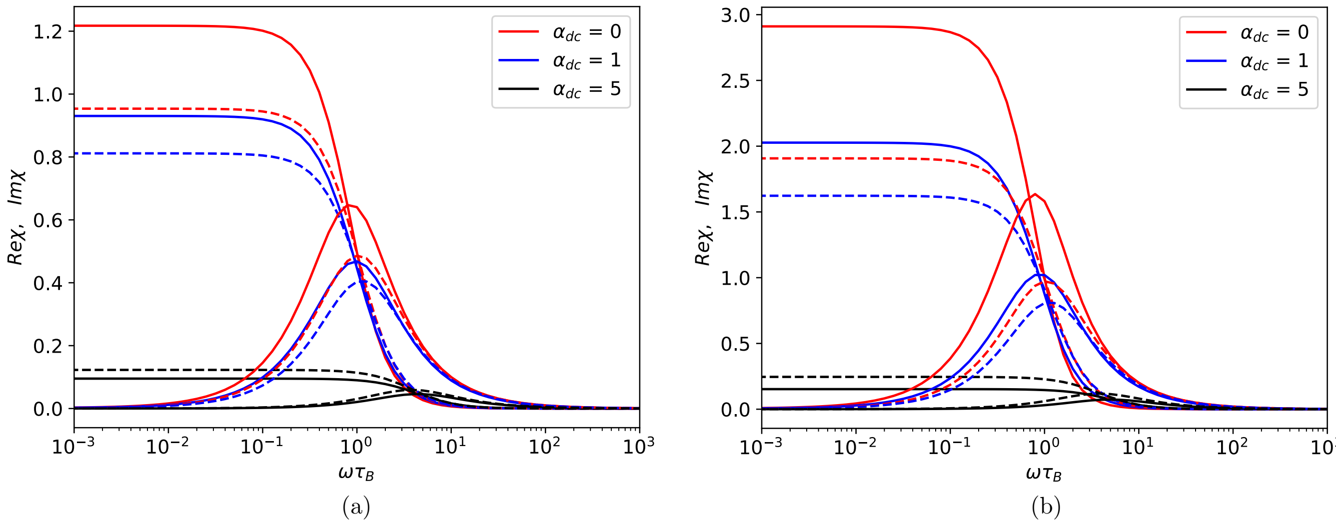

FIG. 7. Numerical results of dynamic susceptibility of magnetic particles to an ac field with amplitude $\alpha_{ac} = 1$ for different values of the Langevin susceptibility (a) $\chi_L = 1$, (b) $\chi_L = 2$. The intensities of the bias dc field are $\alpha_{dc} = 0$ (red), $\alpha_{dc} = 1$ (blue), $\alpha_{dc} = 5$ (black). The solid line corresponds to a system with interactions, the dashed line indicates a system without interactions.

numerical results for interacting particles (red solid line) are in strong agreement with [40]. In the region $\alpha_{ac} \lesssim 3$, at $\alpha_{dc} = 0$ and 1, dipole-dipole interactions increase susceptibility, but at $\alpha_{dc} = 3$ and 5, the opposite trend is observed. The explanation for this behavior is as follows. Dipole-dipole interactions in the presence of a dc field lead to the formation of nose-to-tail correlation structures. When the energy of the ac field exceeds the energy of the dc field, these dipole correlation structures react to the ac field and move with it, making a significant contribution to susceptibility. However, when the dc field strong, it captures magnetic moments, and the resulting nose-to-tail correlation structures are an additional orientation constraint that reduces susceptibility. In the region of $3 \lesssim \alpha_{ac} \lesssim 4$ at $\alpha_{dc} = 3$ and in the region of $4 \lesssim \alpha_{ac} \lesssim 6$ at $\alpha_{dc} = 5$, the ac field begins to dominate the dc field, the dipole-dipole and dipole-dc field bonds break and more and more single particles move after the ac field, which causes an increase in susceptibility. In strong fields, the dipole-dipole interaction becomes weak in comparison with the interaction between the magnetic moment and the ac field, and the system's behavior is completely determined by the dipole-ac field interaction. The change of regimes from the dominance of dipole-ac field interaction to dipole-dc field interaction and vice versa occurs near the critical point $\alpha_{dc} - \alpha_{ac} \approx 0$. This difference is not strictly equal to zero, since the magnetic field introduced by the interparticle dipole-dipole interaction contributes to the total magnetic field acting on the system.

C. Dynamic susceptibility spectrum

Figure 7 shows the effect of interparticle dipole-dipole interactions and the intensity of the dc field on the spectrum of dynamic susceptibility of a system of moving interacting magnetic particles. The susceptibility is shown for two systems with Langevin susceptibility $\chi_L = 1$ [Fig. 7(a)] and $\chi_L = 2$ [Fig. 7(b)]. At low intensities of the dc field $\alpha_{dc} = 0$ and 1, dipole-dipole interactions lead to an increase in the system's susceptibility, while for a sufficiently high intensity of the dc field $\alpha_{dc} = 5$, the opposite trend is observed. This behavior manifests itself more significantly in a system with strong interparticle interactions at $\chi_L = 2$ and is associated with the formation of nose-to-tail dipolar correlation structures, which are an additional orientation factor. Depending on the intensity of competing dipole-dipole, dipole-ac field, and dipole-dc field interactions, these structures can reduce or increase susceptibility. An increase in α_{dc} leads to a decrease in the system's susceptibility at all frequencies of the ac field and a shift to the right of the maximum of the imaginary part of susceptibility. The latter indicates a decrease in the relaxation time of magnetic moments and is explained by the fact that in a magnetized system, the ac field is not able to significantly deflect the magnetic moments held by the dc field, as a result of which their effective relaxation time is reduced.

The effect of ac field amplitude on the dynamic susceptibility spectrum of an ensemble of moving interacting magnetic particles under a bias dc field is shown in Fig. 8. In the case of $\alpha_{dc} = 1$ [Fig. 8(a)], an increase in α_{ac} leads to a decrease in susceptibility for all field frequencies. If the dc field is strong $\alpha_{dc} = 5$ [Fig. 8(c)], then the system's susceptibility increases with the growth of α_{ac} at all frequencies of the ac field. When $\alpha_{dc} = 3$ [Fig. 8(b)], a nonmonotonic change in susceptibility is observed with an increase in α_{ac} due to competing dipole-ac field, dipole-dc field, and dipole-dipole interactions.

It is possible to show a clear dependence of dynamic susceptibility on temperature (Fig. 9). For magnetite particles (Fe_3O_4), which are typical for real ferrofluids, the mean diameter is $d = 10$ nm, saturation magnetization is $M_s = 4.8 \times 10^5$ A m^{-1}, and particle dipole moment is $m = M_s \pi d^3/6 = 2.5 \times 10^{-19}$ Am^2. For a system with a volume concentration $\varphi = 0.08$ in applied ac and dc magnetic fields with $H_{ac} = H_{dc} = 12$ kAm^{-1}, an increase in temperature leads to a decrease in the susceptibility at low frequencies. This behavior is due to an increase in the kinetic energy of the system, which contributes to the disordering of the magnetic moments of the ferroparticles.

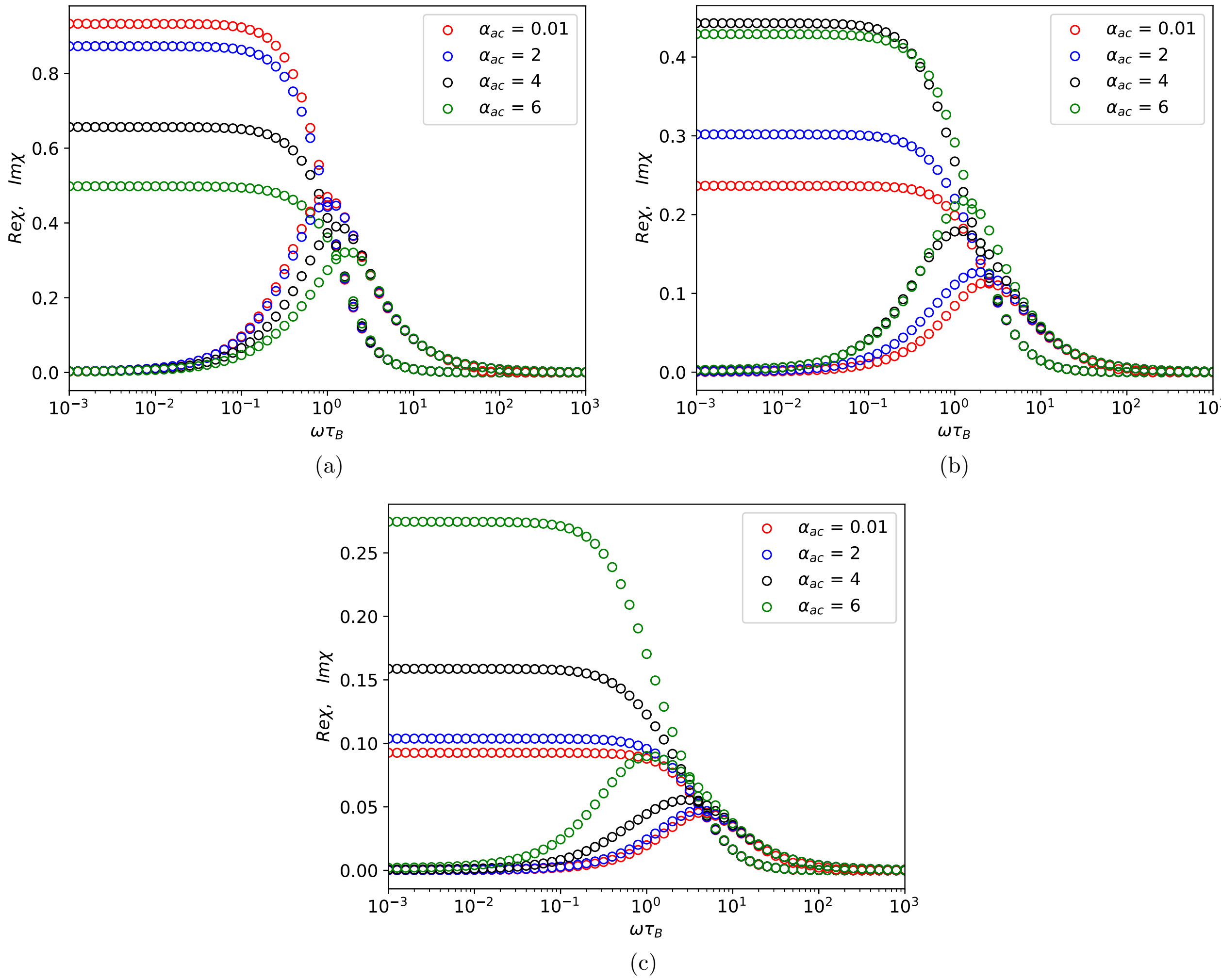

FIG. 8. Numerical results of dynamic susceptibility of an ensemble of interacting magnetic particles with $\chi_L = 1$ to an ac field with amplitude $\alpha_{ac} = 0.01$ (red), $\alpha_{ac} = 2$ (blue), $\alpha_{ac} = 4$ (black), and $\alpha_{ac} = 6$ (green). The intensities of the bias dc field are (a) $\alpha_{dc} = 1$, (b) $\alpha_{dc} = 3$, (c) $\alpha_{dc} = 5$.

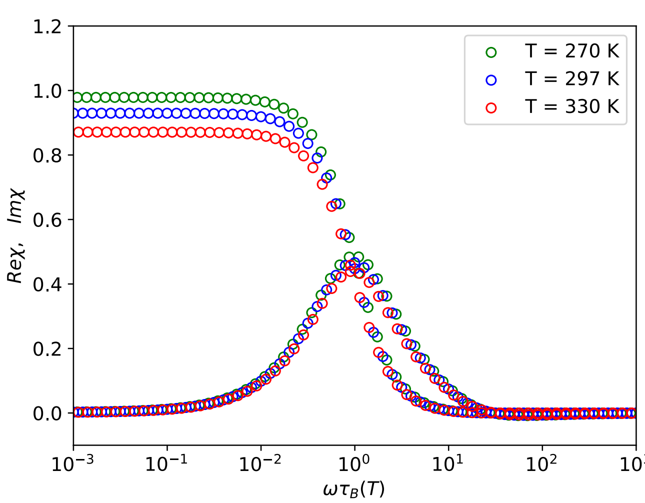

FIG. 9. Numerical results of dynamic susceptibility for a suspension of the magnetite particles with diameter $d = 10$ nm and volume concentration $\varphi = 0.08$ in applied ac and dc magnetic fields with $H_{ac} = H_{dc} = 12$ kAm^{-1} at $T = 270$ K (green), $T = 297$ K (blue), and $T = 330$ K (red).

IV. CONCLUSIONS

The dynamic magnetization and susceptibility of an ensemble of moving interacting magnetic particles in applied dc field and ac field with an arbitrary amplitude are studied. The following configuration of magnetic fields is considered: ac and dc fields are directed along the Oz axis of the coordinate system. Dynamic magnetization and susceptibility were obtained based on a numerical solution of the Fokker-Planck equation for the probability density of the orientation of the magnetic moment of a random particle. In the FPE, dipole-dipole interactions are taken into account within the framework of the first-order modified mean-field theory. The obtained numerical results were tested on known theoretical results for two limiting cases: (a) there is a dc field and the ac field's amplitude is low [43]; (b) there is no dc field and the ac field is of an arbitrary amplitude [40]. Strong agreement with the literature data was obtained.

The dynamic response of an ensemble of moving interacting particles is formed as a result of competing dipole-dipole, dipole-ac field, and dipole-dc field interactions. In a region where the energy of these three interactions is comparable, an increase in α_{ac} at a fixed α_{dc} can cause both an increase and decrease in susceptibility.

At low dc field intensities, interparticle dipole-dipole interactions increase the magnetic response of an ensemble of moving magnetic particles, while at high α_{dc} dipole interactions decrease the system's dynamic response. This behavior is associated with the formation of nose-to-tail dipolar structures under the dc field. When the dipole-ac field energy exceeds the dipole-dc field energy, nose-to-tail dipolar structures move under the action of the ac field, significantly increasing the system's susceptibility. When the dipole-ac field energy is less than the dipole-dc field energy, nose-to-tail structures are held in the direction of the dc field, being an additional orientation constraint that reduces susceptibility.

The theory developed in the article allows us to numerically predict the magnetic response of an ensemble of free moving magnetic particles taking into account competing dipole-dipole, dipole-ac field, and dipole-dc field interactions. Since the theory is built within the framework of the first-order modified mean-field theory, it has limitations on the concentration and intensity of dipole-dipole interactions $\chi_L \lesssim 3$. The numerical scheme implemented for the FPE solution does not impose any restrictions on the intensity of the dc field or the amplitude of the ac field.

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[1] R. E. Rosensweig, *Ferrohydrodynamics* (Courier Corporation, 2013).

[2] V. Socoliuc, M. V. Avdeev, V. Kuncser, R. Turcu, E. Tombácz, and L. Vékás, Ferrofluids and bio-ferrofluids: Looking back and stepping forward, Nanoscale **14**, 4786 (2022).

[3] J. Philip, Magnetic nanofluids (ferrofluids): Recent advances, applications, challenges, and future directions, Adv. Colloid Interface Sci. **311**, 102810 (2023).

[4] Q. A. Pankhurst, N. T. K. Thanh, S. K. Jones, and J. Dobson, Progress in applications of magnetic nanoparticles in biomedicine, J. Phys. D **42**, 224001 (2009).

[5] V. M. Wu, E. Huynh, S. Tang, and V. Uskoković, Brain and bone cancer targeting by a ferrofluid composed of superparamagnetic iron-oxide/silica/carbon nanoparticles (earthicles), Acta Biomater. **88**, 422 (2019).

[6] A. Sohail, M. Fatima, R. Ellahi, and K. B. Akram, A videographic assessment of ferrofluid during magnetic drug targeting: An application of artificial intelligence in nanomedicine, J. Mol. Liq. **285**, 47 (2019).

[7] G. Kandasamy, A. Sudame, P. Bhati, A. Chakrabarty, and D. Maity, Systematic investigations on heating effects of carboxyl-amine functionalized superparamagnetic iron oxide nanoparticles (spions) based ferrofluids for in vitro cancer hyperthermia therapy, J. Mol. Liq. **256**, 224 (2018).

[8] M. M. Nosan, A. S. Omelyanchik, I. G. Samusev, N. M. Myslitskaya, R. Y. Borkunov, I. I. Lyatun, V. V. Rodionova, and V. V. Bryukhanov, Magnetic properties of nanoparticles prepared from -Fe target by laser ablation in liquids, *2017 Progress in Electromagnetics Research Symposium - Fall (PIERS - FALL), Singapore* (IEEE, Piscataway, NJ, 2017).

[9] M. S. Gopika, B. B. Lahiri, B. Anju, J. Philip, and S. Savitha Pillai, Magnetic hyperthermia studies in magnetite ferrofluids based on bio-friendly oils extracted from calophyllum inophyllum, brassica juncea, ricinus communis and madhuca longifolia, J. Magn. Magn. Mater. **537**, 168134 (2021).

[10] E. A. Sokolov, D. A. Kalyuzhnaya, A. O. Vasilyeva, and P. A. Ryapolov, Microfluidic devices with integrated controlled magnetic field sources, *2022 Conference of Russian Young Researchers in Electrical and Electronic Engineering (ElConRus), Saint Petersburg, Russian Federation* (IEEE, Piscataway, NJ, 2022).

[11] A. A. Zakinyan, S. S. Belykh, A. R. Zakinyan, and K. V. Yerin, Structured media based on magnetic colloids as a promising material for magnetically controllable optical elements, J. Opt. Technol. **88**, 158 (2021).

[12] P. J. W. Debye, *Polar Molecules* (Chemical Catalog Company, New York, 1929).

[13] M. A. Martsenyuk, Y. L. Raikher, and M. I. Shliomis, On the kinetics of magnetization of suspensions of ferromagnetic particles, Sov. Phys. JETP **38**, 413 (1974).

[14] Y. L. Raikher and V. I. Stepanov, Nonlinear dynamic susceptibilities and field-induced birefringence in magnetic particle assemblies, Adv. Chem. Phys. **129**, 419 (2004).

[15] V. Zverev, A. Dobroserdova, A. Kuznetsov, A. Ivanov, and E. Elfimova, Computer simulations of dynamic response of ferrofluids on an alternating magnetic field with high amplitude, Mathematics **9**, 2581 (2021).

[16] S. Draack, N. Lucht, H. Remmer, M. Martens, B. Fischer, M. Schilling, F. Ludwig, and T. Viereck, Multiparametric Magnetic particle spectroscopy of $CoFe_2O_4$ nanoparticles in viscous Media, J. Phys. Chem. C **123**, 6787 (2019).

[17] W. T. Coffey and Y. P. Kalmykov, *The Langevin Equation*, Vol. 27 of World Scientific Series in Contemporary Chemical Physics (World Scientific, Singapore, 2012).

[18] R. J. Deissler, Y. Wu, and M. A. Martens, Dependence of Brownian and Néel relaxation times on magnetic field strength, Med. Phys. **41**, 012301 (2014).

[19] A. A. Kuznetsov and A. F. Pshenichnikov, Nonlinear response of a dilute ferrofluid to an alternating magnetic field, J. Mol. Liq. **346**, 117449 (2022).

[20] T. Yoshida and K. Enpuku, Simulation and quantitative clarification of AC susceptibility of magnetic fluid in nonlinear Brownian relaxation region, Jpn. J. Appl. Phys. **48**, 127002 (2009).

[21] T. M. Batrudinov, A. V. Ambarov, E. A. Elfimova, V. S. Zverev, and A. O. Ivanov, Theoretical study of the dynamic magnetic response of ferrofluid to static and alternating magnetic fields, J. Magn. Magn. Mater. **431**, 180 (2017).

[22] W. T. Coffey, Y. P. Kalmykov, and N. Wei, Nonlinear normal and anomalous response of non-interacting electric and magnetic dipoles subjected to strong AC and DC bias fields, Nonlinear Dyn. **80**, 1861 (2015).

[23] S. V. Titov, P.-M. Déjardin, H. El Mrabti, and Y. P. Kalmykov, Nonlinear magnetization relaxation of superparamagnetic nanoparticles in superimposed ac and dc magnetic bias fields, Phys. Rev. B **82**, 100413(R) (2010).

[24] J. Zhong, N. Lucht, B. Hankiewicz, M. Schilling, and F. Ludwig, Magnetic field orientation dependent dynamic susceptibility and Brownian relaxation time of magnetic nanoparticles, Appl. Phys. Lett. **115**, 133102 (2019).

[25] A. V. Lebedev, S. S. Kantorovich, A. O. Ivanov, I. M. Arefyev, and A. F. Pshenichnikov, Weakening of magnetic response experimentally observed for ferrofluids with strongly interacting magnetic nanoparticles, J. Mol. Liq. **277**, 762 (2019).

[26] A. O. Ivanov and P. J. Camp, Effects of interactions, structure formation, and polydispersity on the dynamic magnetic susceptibility and magnetic relaxation of ferrofluids, J. Mol. Liq. **356**, 119034 (2022).

[27] A. S. Ivanov, A. Y. Solovyova, V. S. Zverev, and E. A. Elfimova, Distribution functions of magnetic moments and relaxation times for magnetic fluids exhibiting controllable microstructure evolution, J. Mol. Liq. **367**, 120550 (2022).

[28] E. A. Elfimova, A. O. Ivanov, E. V. Lakhtina, A. F. Pshenichnikov, and P. J. Camp, Sedimentation equilibria in polydisperse ferrofluids: Critical comparisons between experiment, theory, and computer simulation, Soft Matter **12**, 4103 (2016).

[29] A. Y. Solovyova, O. A. Goldina, A. O. Ivanov, A. V. Lebedev, and E. A. Elfimova, The initial magnetic susceptibility of polydisperse ferrofluids: A comparison between experiment and theory over a wide range of concentration, J. Chem. Phys. **145**, 084909 (2016).

[30] A. O. Ivanov and E. A. Elfimova, Low-temperature magnetic susceptibility of concentrated ferrofluids: The influence of polydispersity, J. Magn. Magn. Mater. **374**, 327 (2015).

[31] I. Szalai and S. Dietrich, Magnetization of multicomponent ferrofluids, J. Phys.: Condens. Matter **23**, 326004 (2011).

[32] B. U. Felderhof and R. B. Jones, Mean field theory of the nonlinear response of an interacting dipolar system with rotational diffusion to an oscillating field, J. Phys.: Condens. Matter **15**, 4011 (2003).

[33] P. Ilg and S. Hess, Nonequilibrium dynamics and magnetoviscosity of moderately concentrated magnetic liquids: A dynamic mean-field study, Z. Naturforsch., A **58**, 589 (2003).

[34] D. V. Berkov, L. Y. Iskakova, and A. Y. Zubarev, Theoretical study of the magnetization dynamics of nondilute ferrofluids, Phys. Rev. E **79**, 021407 (2009).

[35] P. M. Déjardin and F. Ladieu, Nonlinear susceptibilities of interacting polar molecules in the self-consistent field approximation, J. Chem. Phys. **140**, 034506 (2014).

[36] A. V. Lebedev, V. I. Stepanov, A. A. Kuznetsov, A. O. Ivanov, and A. F. Pshenichnikov, Dynamic susceptibility of a concentrated ferrofluid: The role of interparticle interactions, Phys. Rev. E **100**, 032605 (2019).

[37] A. O. Ivanov and P. J. Camp, Theory of the dynamic magnetic susceptibility of ferrofluids, Phys. Rev. E **98**, 050602(R) (2018).

[38] A. O. Ivanov, S. S. Kantorovich, E. A. Elfimova, V. S. Zverev, J. O. Sindt, and P. J. Camp, The influence of interparticle correlations and self-assembly on the dynamic initial magnetic susceptibility spectra of ferrofluids, J. Magn. Magn. Mater. **431**, 141 (2017).

[39] A. O. Ivanov and O. B. Kuznetsova, Magnetic properties of dense ferrofluids: An influence of interparticle correlations, Phys. Rev. E **64**, 041405 (2001).

[40] M. S. Rusanov, V. S. Zverev, and E. A. Elfimova, The dynamic magnetic susceptibility of a ferrofluid: The influence of interparticle interactions and AC field amplitude, Phys. Rev. E **104**, 044604 (2021).

[41] A. Y. Zubarev and A. V. Yushkov, Dynamic properties of moderately concentrated magnetic liquids, J. Exp. Theor. Phys. **87**, 484 (1998).

[42] B. Horváth, P. Decsi, and I. Szalai, Nonlinear contributions to the dynamic magnetic susceptibility of magnetic fluids, J. Mol. Liq. **359**, 119279 (2022).

[43] T. M. Batrudinov, Y. E. Nekhoroshkova, E. I. Paramonov, V. S. Zverev, E. A. Elfimova, A. O. Ivanov, and P. J. Camp, Dynamic magnetic response of a ferrofluid in a static uniform magnetic field, Phys. Rev. E **98**, 052602 (2018).

[44] A. F. Pshenichnikov, A. V. Lebedev, and A. O. Ivanov, Dynamics of magnetic fluids in crossed DC and AC magnetic fields, Nanomaterials **9**, 1711 (2019).

[45] A. O. Ivanov, V. S. Zverev, and S. S. Kantorovich, Revealing the signature of dipolar interactions in dynamic spectra of polydisperse magnetic nanoparticles, Soft Matter **12**, 3507 (2016).

[46] A. V. Ambarov, V. S. Zverev, and E. A. Elfimova, Numerical modeling of the magnetic response of interacting superparamagnetic particles to an ac field with arbitrary amplitude, Modell. Simul. Mater. Sci. Eng. **28**, 085009 (2020).

[47] P. Déjardin, Y. P. Kalmykov, B. Kashevsky, H. El Mrabti, I. Poperechny, Y. L. Raikher, and S. Titov, Effect of a dc bias field on the dynamic hysteresis of single-domain ferromagnetic particles, J. Appl. Phys. **107**, 073914 (2010).

[48] W. T. Coffey, D. Crothers, Y. P. Kalmykov, and P. Déjardin, Nonlinear response of permanent dipoles in a uniaxial potential to alternating fields, Phys. Rev. E **71**, 062102 (2005).

[49] J. Dieckhoff, D. Eberbeck, M. Schilling, and F. Ludwig, Magnetic-field dependence of Brownian and Néel relaxation times, J. Appl. Phys. **119**, 043903 (2016).

[50] J. O. Sindt, P. J. Camp, S. S. Kantorovich, E. A. Elfimova, and A. O. Ivanov, Influence of dipolar interactions on the magnetic susceptibility spectra of ferrofluids, Phys. Rev. E **93**, 063117 (2016).

[51] A. F. Pshenichnikov, V. V. Mekhonoshin, and A. V. Lebedev, Magneto-granulometric analysis of concentrated ferrocolloids, J. Magn. Magn. Mater. **161**, 94 (1996).