Magnetization dynamics of non-dilute ferrofluids

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Abstract. The paper is devoted to theoretical studies of the magnetodipolar interparticle interaction effect on magnetization dynamics in moderately concentrated ferrofluids. We consider a homogeneous (without particle aggregates) ferrofluid consisting of identical spherical particles and employ a rigid dipole model, where the particle magnetic moment is fixed with respect to the particle itself. For the magnetization relaxation after an instant change of the external field, we show that the magnetodipolar interaction leads to the increase of the initial magnetization relaxation time. For the complex ac-susceptibility $\chi$ we find that the this interaction leads to an overall increase of its imaginary part $\chi''$ and shifts the $\chi''$-peak towards lower frequencies. Comparing results obtained with our analytical approach to numerical simulation data, we demonstrate that the theory provides good quantitative agreement with numerical simulations up to the particle volume fraction $\varphi \sim 6\%$ - 10%.

1. Introduction

Experiments demonstrate that magnetodipolar interparticle interaction changes significantly both the equilibrium [1] and dynamical [2] properties of ferrofluids. Theoretical models of dynamical properties of dilute ferrofluids with vanishing interparticle interactions have been proposed in [3]. These models lead to highly accurate results for very dilute ferrofluids but can not explain properties and behavior of ferrofluids where the interparticle interactions are significant. Depending on the energy of the magnetodipolar interaction, it can lead either to appearance of homogeneous long-range interparticle correlations, or to formation of chain-like, drop-like and other heterogeneous internal structures [4]. Appearance of these correlations and, especially, heterogeneous structures can significantly change dynamical properties of ferrofluids [2]. At present there is no general theory which could adequately predict an internal structure of non-dilute ferrofluids at given experimental conditions. Therefore it is reasonable to consider effects of different internal structures on dynamical phenomena in ferrofluids separately. Such idealized models can provide better insights into the influence of various internal structures on the macroscopical ferrofluids properties.
In this paper we study magnetization dynamics of a homogeneous ferrofluid. For maximal simplification of calculations, we consider a monodisperse model, consisting of identical interacting magnetic particles. It is assumed that magnetic moment of each particle has a constant magnitude and is “frozen” into the particle body. Remagnetization dynamics of this model is studied both analytically and using numerical simulations. In order to simplify the mathematical treatment, we consider an idealized ferrofluid consisting of identical particles. We assume that the magnetodipolar interaction energy is less or of the same order of magnitude as the thermal energy $kT$ (otherwise particle aggregates must appear in a ferrofluid; their treatment is beyond the scope of this paper). Next, in order to focus on magnetodipolar interaction effects, we neglect here the hydrodynamical interaction between particles. Effects of this interaction will be considered in a separate publication.

### 2. Basic equations

We consider a ferrofluid with volume $V$ containing $N$ identical spherical ferromagnetic particles with the diameter $d$. The magnitude $m$ of the particle magnetic moment $\mathbf{m}$ is constant, the moment is “frozen” into the particle body. We introduce the unit vector $\mathbf{e}_i = \mathbf{m}_i / m$ of the magnetic moment of the $i$-th particle and denote the radius-vector of this particle by $\mathbf{r}_i$. To calculate the macroscopic characteristics of this system, we must determine the $N$-particle distribution function $P_N(e_1, \ldots, e_N, r_1, \ldots, r_N)$. It can be found by solving the appropriate Fokker-Planck equation, where we have to take into account the magnetodipolar interactions between all particles. This equation reads

$$\frac{\partial P_N}{\partial t} = \sum_i I_i \left( \frac{D_i}{kT} (\nabla \cdot P_N) + \nabla_i \left( \frac{D_i}{kT} \nabla_i U \right) P_N \right) + D_r \sum_i \mathbf{r}_i \cdot \mathbf{P}_N^2 + D_t \sum_i \mathbf{r}_i^2 P_N$$

where we have used the standard notation $I_i = \mathbf{e}_i \times \frac{\partial}{\partial \mathbf{e}_i}$, $\nabla_i = \frac{\partial}{\partial \mathbf{r}_i}$. The potential energy of the system is $U = -kT \sum_i (\mathbf{r} \cdot \mathbf{e}_i) + (1/2) \sum_{i,j} w_{ij}$, where the reduced field $\mathbf{r} = \mu_0 m \mathbf{H} / kT$ is defined via the vacuum permeability $\mu_0$, and the applied magnetic field $\mathbf{H}$, term $w_{ij}$ is the magnetodipolar interaction energy of particles $i$ and $j$, $D_r$ and $D_t$ are rotational and translational diffusion coefficients of the particles. The mean magnetization of a ferrofluid can be found as

$$\mathbf{M} = mn \int e_1 P_N d\mathbf{e}_1 \ldots d\mathbf{e}_N d\mathbf{r}_1 \ldots d\mathbf{r}_N,$$

where $n$ is number of ferrofluid particles per unite volume.

The Fokker-Planck equation (1) can not be solved exactly for two reasons. The first one is the well-known problem of statistical physics – interparticle interaction in a many-particle system does not allow (nearly always) to solve the governing equation for a many-particle distribution function or to calculate the Gibbs statistical integral. The second reason is the purely mathematical difficulty arising by the solution of the Fokker-Planck equation even for a single particle.

In order to take into account interaction between particles as accurately as possible and to avoid unjustified intuitive assumptions, we use the mathematically regular method of virial expansion over the particle concentration. To overcome the second problem, we use the effective-field approach, suggested by M. Shliomis [3], which is a version of the trial function method. According to this approach, we write the function $P_N$ in the form of an equilibrium Gibbs function in some effective magnetic field $\mathbf{H}_e$, which is to be determined, instead of the real field $\mathbf{H}$. To find the components of $\mathbf{H}_e$ we multiply Eq. (1) by the orientation vector $\mathbf{e}$ and integrate the result over all $\mathbf{e}$, and $\mathbf{r}$. This way we obtain an equation, which includes the first and second statistical moments of
the function $P_N$. By using in these moments the Gibbs function with the effective field $H_e$, we arrive at ordinary differential equations with respect to the components of this field. These equations can be solved much easier than the basic equation (1). Having found the field $H_e$ and using the Gibbs function with $H_e$ in Eq.(2), we determine the macroscopical magnetization $M$.

Detailed derivation of the analytical equation for $H_e$ will be given elsewhere [6].

Numerical simulations are based on the Langevin dynamics formalism, where the equations of motion for the relevant degrees of freedom characterizing the system under study are solved taking into account thermal fluctuations (see [5] for details).

3. Results and discussion

Main results of our analytical and numerical calculations are presented below.

![Graph](image1)

**Fig. 1.** Time dependence of the reduced magnetization $\mu = M/\mu_{mn}$ after the step-wise change of the external field from 200 Oe to 0 (left), and from 0 to 200 Oe (right). Lines – analytical theory, circles – numerical simulations. Figures near the curves show volume concentrations $\phi$ of the magnetic phase.

![Graph](image2)

**Fig. 2.** Relaxation time $\tau$ of the magnetization onset after the instant increase of the external field from 0 to 200 Oe vs volume concentration $\phi$.

Fig. 1. shows the effect of the interaction between particles on their mean magnetic moment $\mu$.

Results presented in Fig. 2 demonstrate that the magnetodipolar interaction between particles leads to the increase of the initial rate of the ferrofluid magnetization.
The frequency dependencies of real $\chi'$ and imaginary $\chi''$ parts of the reduced complex susceptibility of a ferrofluid $\chi = M_0/(H_0 \phi)$ are presented in Fig. 3. Here index 0 marks the amplitudes of the magnetization and applied oscillating field. These results show that interparticle interaction decreases the frequency of $\chi''$-maximum and increases the value of $\chi''$ at this maximum.

**Conclusion**

An analytical model of the magnetization dynamics of idealized monodisperse ferrofluids with interacting particles is presented. The model does not contain any fitting parameters and leads to a good agreement with computer simulations up to high (~10%) volume concentrations of the magnetic phase. Magnetodipolar interaction between particles decreases the rate of the ferrofluid remagnetization, decreases the peak frequency of the imaginary part of the ferrofluid complex susceptibility and increases the height of this peak. Suggested model can be also applied for investigation of other properties (e.g., rheological behavior) of non-dilute ferrofluids and generalized to polydisperse systems.

This work has been supported by RFFI, grants N 06-01-00125, 07-02-00079, 07-01-960769Ural, 08-02-00647, Fund CRDF, PG07-005-02.

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