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Simulations of ferrofluid dynamics: Rigid dipoles model versus particles with internal degrees of freedom

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Abstract

For numerical studies of a ferrofluid dynamics we have developed a model which includes internal magnetic degrees of freedom of ferrofluid particles. Contrary to standard models, we take into account that the magnetocrystalline anisotropy of a ferrofluid particle material is finite, so that the particle moment is allowed to rotate with respect to the particle itself. Simulating magnetization relaxation of a ferrofluid after switching off the external field and comparing results with those obtained for rigid dipoles model, we demonstrate that for anisotropy typical for commonly used ferrofluid materials inclusion of 'magnetic' degrees of freedom is essential for a correct description of ferrofluid dynamics.

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Keywords: Ferrofluid; Numerical simulations; Dipolar systems

In standard ferrofluid models (see, e.g., Refs. [1–4]) magnetic moment of a ferrofluid particle is supposed to be fixed with respect to the particle itself (rigid dipoles model), which corresponds to the limit of an infinitely high single-particle magnetic anisotropy. For real ferrofluids, however, the magnetocrystalline anisotropy K of a ferrofluid particle material is finite and usually even not large, so that the reduced anisotropy (which gives the relation of the anisotropy field to the maximal dipolar interaction field) is $\beta = 2K/M_S^2 \sim 1$ and the anisotropy energy KV (V is the magnetic particle volume) can be of the same order of magnitude as the thermal energy kT. For this reason, we have implemented a model where particle magnetic moments are allowed to rotate with respect to the particle crystallographic axes. Our model results in a system of differential equations describing both magnetic and mechanical degrees of freedom, where the 'magnetic' equations are coupled with the 'mechanic' equations via (i) the interparticle distances which determine the dipolar interaction fields and (ii) relative orientations of the particle anisotropy axes and magnetic moments which define the mechanical torque acting on the particle. Detailed discussion of these equations will be given elsewhere [5].

In this contribution we compare simulation results obtained using this 'flexible' dipoles model with those from the rigid dipoles system, taking as a physical example the relaxation of a ferrofluid magnetization after a ferrofluid is magnetized in an external field and this field is then switched off. Such a relaxation can be conveniently simulated using the Langevin dynamics, because the corresponding relaxation time is of the same order of magnitude as the rotational diffusion time $\tau_B^{rot} = 3\eta V_p/kT \sim 10^{-5} - 10^{-6}$s. We have simulated a water-based (viscosity $\eta = 10^{-2}$ P) ferrofluid consisting of identical particles with the magnetic kernel radius $R_{mag} = 10$ nm, shell thickness $h = 2$ nm and saturation magnetization $M_S = 400$ G (magnetite). We have studied the effects of the anisotropy strength β and magnetodipolar interparticle interaction; the latter can be controlled by the changing the particle volume fraction (concentration) c. All relaxation curves presented below were obtained by averaging over 8 independent runs on systems containing $N = 1000$ particles each.

In Fig. 1 we compare magnetization relaxation for the rigid dipole model and a ferrofluid with 'flexible' magnetic moments. For particles with a relatively large anisotropy $\beta = 2.0$ chosen

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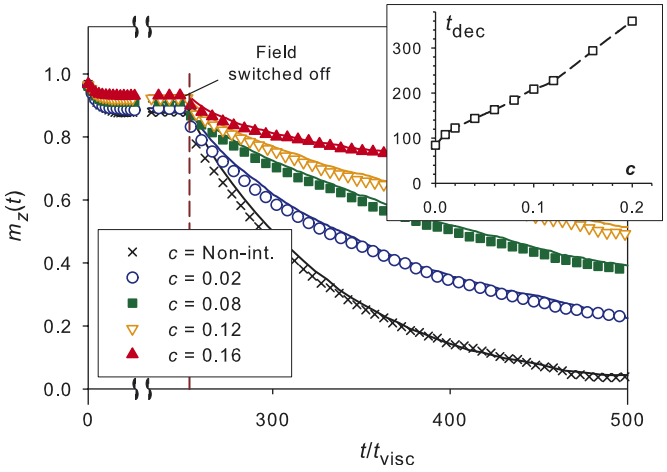

Fig. 1. Magnetization relaxation after switching off the external field for ferrofluids with different particle concentrations c (shown in the legend). Solid lines are relaxation curves for the rigid dipole model, symbols-for 'flexible' magnetic moments with $\beta = 2.0$. The inset shows the concentration dependence of the relaxation time.

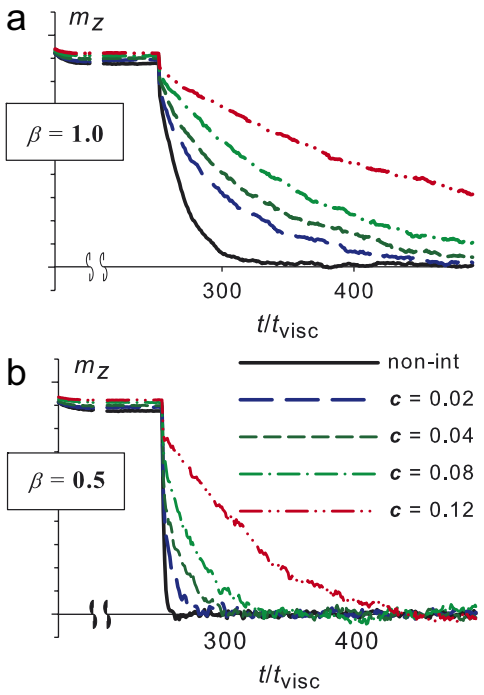

Fig. 2. The same magnetization relaxation as shown in Fig. 1 for ferrofluids with 'flexible' magnetic moments and various single-particle anisotropy constants β as shown in the figure. It can be clearly seen that (i) already for the moderate anisotropy $\beta = 1.0$ the relaxation is much faster than for the rigid dipole model and (ii) the influence of the magnetodipolar interaction is much stronger for a system of particles with smaller anisotropy.

for these simulations the relation of the anisotropy energy barrier to the thermal energy $\beta M_S^2 V_{mag}/2kT \approx 16$ is high. Hence we expect that the Neel relaxation can be neglected and the two models should give equivalent results. This is indeed the case, as it can be clearly seen comparing the relaxation curves in Fig. 1 for one and the same concentration obtained from the rigid dipole model (lines) and ferrofluids with 'flexible' dipoles (symbols). The only difference between the models is a slightly lower initial magnetization for the model with 'flexible' dipoles, which is due to additional thermal fluctuations of magnetic moments around the anisotropy axis which decrease the equilibrium magnetization.

However, already for the anisotropy $\beta = 1.0$ which is only two times smaller than used in the previous simulations, magnetization relaxation is substantially faster than for the rigid dipoles due to a significant contribution of the Neel relaxation: compare, e.g., black solid lines in Figs. 1 and 2a which show magnetization relaxation for systems of non-interacting particles with $\beta = 2.0$ and 1.0. For the still lower anisotropy $\beta = 0.5$ the Neel relaxation for the non-interacting system is so fast that it is practically instantaneous on the time scale of the viscous Brownian motion (solid black line in Fig. 2b). This happens because although the relation of the anisotropy energy barrier to the thermal energy $\beta M_S^2 V_{mag}/2kT \approx 4$ is still relatively large, the prefactor before the exponent in the expression for the Neel relaxation time $\tau_{Neel} = \tau_0 \cdot \exp(KV/kT)$ is so small that the corresponding transition time is much smaller than the rotational Brownian motion time τ_B^{rot}.

Another important effect clearly visible from our simulation results is a much stronger influence of the magnetodipolar interparticle interaction for systems with smaller anisotropy values. Whereas for $\beta = 2.0$ the decay time for, e.g., the particle concentration $c = 0.12$ is only about three times larger than for the non-interacting system (see inset to Fig. 1), for a system with $\beta = 0.5$ the relation of corresponding relaxation times exceeds $t^{dec}_{c=0.12}/t^{dec}_{non\text{-}int} > 10^3$. The reason for such an enhancement of the interaction influence is its qualitatively different role in ferrofluids consisting of particles with high and low anisotropy barriers. For particles with high anisotropy values the magnetodipolar interaction 'only' causes substantial correlations of magnetic moments for neighboring particles, thus increasing somewhat the corresponding relaxation time (such a 'dynamic' cluster relaxes slower than a single particle). In contrast to the previous situation, in systems with low-anisotropy particles the interaction field strongly increases the energy barriers for the Neel relaxation [6], thus leading to the exponential increase of the magnetization decay time.

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