An Influence of Chain Aggregates on Magnetic Properties of Monodisperse Ferrofluids

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Recent computer simulations [1,2] have shown the microscopic structure of ferrofluids to be much more complex than previously expected. The system at a high dipolar strength and low volume fraction has proved to associate in chain aggregates, the number and length of which represent increasing functions of the ferroparticle concentration and of the strength of an external magnetic field. It is well known that magnetic fluids become optically anisotropic [3] and demonstrate an abrupt viscosity increase [4] when subjected to a magnetic field. The explanation of these phenomena is usually made in terms of chain aggregates. A lot of experimental studies, demonstrating not only chainlike aggregates existence but also their great influence upon diffusional and hydrodynamic properties of ferrofluids, are worth mentioning [5].

Magnetic properties of aggregated ferrofluids with high dipolar strength were also studied by means of molecular dynamics simulation [2]. According to these papers at low concentration the chain formation tends to increase the magnetization and induces a larger initial susceptibility. Attempts [2] to use the stiff rodlike chain model [6] have shown that the rejection of chain flexibility results in the Langevin orientational law for each rodlike chain and leads to a great overestimation of the chain response to an external field, especially for high field strengths.

In present research we use the theory of particle association in flexible chains in dilute ferrofluids, which was developed in Ref. [7] for the case of an arbitrarily strengthened magnetic field H. Chain distribution in dynamic equilibrium is obtained on the basis of free energy minimization method under the neglect of interchain interaction. The chain partition function is calculated analytically with the help of the rotation matrix technique under the condition when the interparticle dipole-dipole interaction between the nearest neighboring ferroparticles in each chain is taken into account. The modified mean field approach [8] is used for considering the dipole-dipole interaction between all particles in a ferrofluid.

The magnetic orientational response of a single flexible chain to a magnetic field appears to be rather complex. The chain demonstrates nearly a rod-like behavior at weak fields as it rotates as a highly correlated object, whereas when an applied field is strong each particle in a chain starts to obey independently a single particle Langevin law. So, the approximation of rigid rod-like chains becomes invalid in saturation conditions.

Our results for the magnetization curves of aggregated ferrofluids are demonstrated in Fig. 1. The agreement between our model and the simulation data is quite good for moderate dipolar couplings λ, but the theory underestimates the magnetization in the case of high λ. It is worth noting that the interaction only between the nearest neighboring particles in a chain is considered in the model. Evidently, if we allow for the dipole-dipole interaction between the next nearest neighboring particles, chains will become orientationally more rigid. So, this additional influence of the next nearest neighbors may be assumed as a possible explanation of the observed deviations.

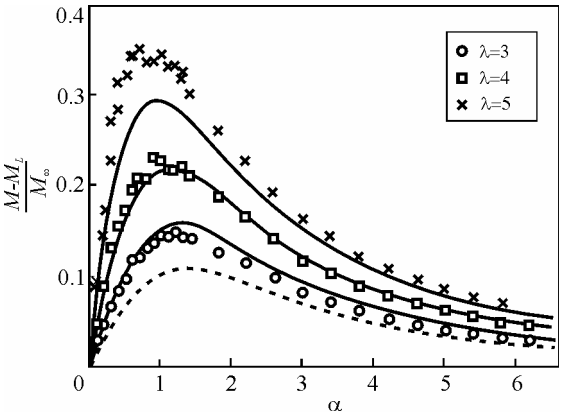

Figure 1: Magnetization curves of the aggregated ferrofluids: symbols represent the data of molecular dynamics simulations [2] for the ferrofluids with different dipolar coupling constant λ=3,4,5 with the same Langevin susceptibility $4\pi\chi_L$=1.256. Dotted line shows the result of modified mean field model [8]. The curves are calculated on the basis of the present model for corresponding values of the dipolar coupling constant. The relative values $(M-M_L)/M_\infty$ are plotted to clarify the deviations.

Both the orientational correlations of particle magnetic moments in chains and the field dependent chain lengthening result in higher magnetization and initial magnetic susceptibility of the aggregated ferrofluid. This conclusion agrees well with the molecular dynamics results [2], and the model describes well the simulated magnetization curves for monodisperse ferrofluids containing chain aggregates.

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