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Interfacial tension of magnetic fluids in the mean spherical approximation

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Abstract

The interfacial tension coefficient for a plane and spherical interface between weakly and highly concentrate phases of magnetic fluid was evaluated in square-gradient expansion for the mean spherical approximation of dipole–dipole interaction. The method based on the minimization of free energy functional was used for solution. The density profile in the interface layer, the influence factor, Tolman length and the interfacial tension coefficient were calculated as functions of temperature and concentration using the tail of pair correlation function. The anisotropy of interfacial tension for a plane interface was calculated far from critical point in weak magnetic field. The estimations for arbitrary angles between magnetic field and interface normal were made for interfacial tension. It was shown that dependence of the interfacial tension on radius of macrodrop is negligible. Giving the expression for interfacial tension coefficient is possible and can be presented as functions of physical measured parameters: magnetic susceptibility, density and compressibility. That opens up the possibility of using these expressions for description of polydisperse magnetic fluids. The theoretical results agree well with experimental estimations.

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1. Introduction

The first statistical theory of the fluid surface was elaborated by Fowler who attempted to treat the fluid properties in terms of intermolecular forces acting at the interface of two phases [\[1\]](#page--1-0). The theory is based on the density jump approximation used by Fowler for description of transient region. This makes the theory inapplicable to description of the fluid surface close to the critical point where the transient region is large in size. Kirkwood and Buff [\[2\]](#page--1-0) developed a formal statistical theory of the fluid surface free of this mathematical artifice, although these authors, too, were faced with the necessity to restrict the transient region to the interface where the jump of density

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occurs. In the absence of rigorous, clearly formulated theory of anisotropic two-particle distribution at the liquid interface, much efforts have been spent for developing a variety of approximate descriptions that allow statistical calculation of the stress tensor at the interface [\[3–5\].](#page--1-0) In these works, the radial part of the pair distribution function is supposed to be invariable along the interface. A comparison with the experiment suggests fairly good agreement with theoretical results for liquids with radial potential of pair interaction [\[6,7\].](#page--1-0)

In the last years the density functional formalism (DFF) [\[8\]](#page--1-0) has become recognized as a powerful tool in the study of non-uniform fluids. In particular, it has proved useful in the understanding of interface phenomena in fluids. DFF theories often make use of the so-called gradient expansion. In this expansion the free energy functional is replaced by an expansion that involves gradient in density

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valid for small gradients and small departures from homogeneity. In practice, however, it seems that the behavior of the non-homogeneous fluid is quite well described even if these conditions are not satisfied well. A further ingredient of the theory is knowledge of the local free energy density and some correlation functions of the bulk homogeneous fluid. The calculation of surface tension of dipolar fluid in the context of the DFF was made for modified van der Waals model [\[9,10\]](#page--1-0) and modified mean field theory [\[11\]](#page--1-0).

In spite of the fact that there has been a number of welldefined analytical models, no experimental attempts have been made to measure the contribution of dipole interaction to surface tension of dipole liquids. One of the reasons is that in dipole liquids (water being the most demonstrative example) there exist other dispersion interactions whose contribution to the surface tension is considerably greater than that of dipole interactions. The other reason is that contribution of the dipole interaction to the surface energy can be determined experimentally only in the presence of the external field. However, in real liquids the energy of dipole moment interaction with the field in one molecule exceeds the heat energy in abnormally large fields (such fields cannot be generated by experiment). Therefore it was only the advent of man-made materials which enables scientists to estimate the contribution of dipole interactions to the surface tension. Among these the magnetic fluids (MFs) seem to be ideally suited to this purpose.

With the appearance of MFs in mid-1960s, the problem of phase transition has been the subject of heated discussion in the literature. However, some fundamental aspects of the problem are still open. At present the most comprehensively studied effect is the phase transition of the first kind—stratification of MF into weakly and highly concentrate phases under the action of magneto dipole interparticle interactions [\[12–15\].](#page--1-0) In this case, the concentrated phase is represented by the so-called drop aggregates with characteristic dimensions of a few tens of mkm. In the absence of external fields the aggregate takes the spherical shape due to the interfacial tension between the drop aggregate and environment. A nearly spherical shape is also preserved in a weak magnetic field.

However, despite a great body of experimental data [\[16–18\]](#page--1-0) the evaluation of the surface tension coefficient even in the framework of density jump [\[19\],](#page--1-0) modified Cahn–Hilliard theory [\[20\],](#page--1-0) the mean field model (MFM) [\[21–23\]](#page--1-0) is a challenging task, which has been accomplished not long ago. The calculations of this kind in the context of the microscopic mean spherical approximation (MSA) [\[24\]](#page--1-0) are still to be done. There are few valid reasons why the investigation of interfacial tension in MF is of great importance.

Firstly, MFs are the best analog to water, the former having two points in its favor—the well-known interaction potential and the spherical shape of particles.

Secondly, MF is the medium with continuously varying and controlled energy of inter-particle interactions. The concentration and mean size of colloidal particles are variable quantities in experiments.

Thirdly, the interfacial tension of MFs is one of the effects of magnetodipole interactions. The investigation of magnetodipole interactions is not accomplished without the surface properties ferrocolloid solution.

Therefore, the objective of the present investigation is evaluation of the interfacial tension coefficient, the influence factor, density profile in the transient layer between weakly and highly concentrate phases of MF and its dependence on the particle density, temperature and weak magnetic field for a plane and spherical interface of MF in square-gradient expansion based on the MSA.

The paper is organized as follows: In Section 2, we outline the calculation of dipole–dipole interaction in the MSA and MFs thermodynamics within the framework of MSA. The influence factor is derived using the tail of pair correlation function. In Section 3, the density profile in the interface layer, the Tolman length and the interfacial tension coefficient were calculated as functions of temperature and concentration for flat interface. In the case of spherical interface (Section 4) we used the Tolmen formula to describe the dependence of the interfacial surface tension on the radius of the spherical drop aggregate. In Section 5, the anisotropic response of interfacial surface tension in weak magnetic field was calculated far from critical point for flat interface.

2. Thermodynamics of magnetic fluid in MSA

The MSA was introduced by Lebowitz and Percus [\[25\]](#page--1-0) as a generalization of the MSA for spin systems of Lewis and Wannier [\[26\]](#page--1-0). Wertheim [\[24\]](#page--1-0) realized the pure microscopic approach for polar fluids within the framework MSA. For such fluids the instantaneous dipole moment depends on the configuration of all other molecules. Clearly, the existence of a many-body molecular dipole moment greatly complicates the statistical mechanics of fluids, i.e., real fluids. Despite this, Wertheim has succeeded in formulating a remarkably elegant theory for these systems. The results of his theory provide the natural generalizations of the rigid polar fluid results. Later Morozov and co-workers [\[15\]](#page--1-0) successfully used Wertheim generalization of MSA to describe properties of MFs. Moreover, Morozov [\[27\]](#page--1-0) was the first to extend boundary of polar fluids MSA for arbitrary external fields and considered the isotropic–ferromagnetic transitions theory of dipolar hard-sphere fluids based on MSA [\[28\]](#page--1-0).

2.1. Statistical background of MSA

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