Magnetic properties and magneto-birefringence of magnetic fluids

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Abstract. Static magnetic properties of a large variety of magnetic fluids with magnetite particles is studied. A qualitative study of magnetization curves was performed to establish the influence of interactions or the presence of agglomerations in each sample. Improved equations for magneto-granulometric analysis, for ideal ferrofluids, were proposed. Better results for the mean magnetic diameter than in the case of using the known equations were obtained. A quantitative study using several models for ideal and interacting particles was performed to select the best method and dimensional distribution function for magneto-granulometric analysis as well as for accurately determining macroscopic quantities of samples (initial susceptibility, saturation magnetization, particle number density or magnetic volume fraction) and properties of nanoparticles (mean magnetic diameter, thickness of the nonmagnetic layer and particle distribution). A new model for magneto-birefringence was proposed and discussed as well as applied for diluted and concentrated ferrofluids. The Langevin behaviour of samples was investigated and compared with the investigation based on magnetic properties. Nanoparticles parameters like mean “magneto-optical” diameter, effective anisotropy constant, Shliomis diameter and the real part of the electrical permittivity of particles were accurately determined.

1 Introduction

Magnetic fluids (also called ferrofluids) are stable magnetic colloids in which fine particles are usually covered with a surfactant layer (sterical stabilization) to prevent agglomerations [1]. The interactions between particles may lead to several types of aggregates, usually in the shape of linear chains parallel to the magnetic field or drop-like aggregates [2]. The agglomeration process is not desired in most applications, so that a characterization of samples, based on their physical properties, is needed. If the agglomeration process is negligible, the physical properties are determined by the orientation of permanent magnetic moments of nanoparticles in the presence of an external field [3].

Both magnetization curves and induced birefringence, theoretically and experimentally analyzed in the paper, can give information about many ferrofluid macroscopic properties and particle properties (as mentioned in the abstract) and about microstructural properties of ferrofluids (the presence and type of aggregates).

2 Theoretical background

Magnetic properties. Several models based on orientation process of magnetic moments of dispersed particles are used to describe ferrofluid magnetization. The Langevin equation, \( M_L = M_s L(\xi) \) [1], where \( M_s \) is the saturation magnetization, \( L(\xi) \) the Langevin function and \( \xi = \mu_0 mH/(k_B T) \), is not in a very good agreement with the experimental data [4]. The following notations are made: \( T \) is the absolute temperature, \( k_B \) is the Boltzmann constant, \( \mu_0 \) the permeability of vacuum, \( m \) the magnetic moment of one particle and \( H \) the applied magnetic field. Corrections due to dimensional distribution and interactions are necessary. For ferrofluids with aggregates, agglomeration theories of magnetization are needed.

For dispersions of magnetite particles, the lognormal distribution function is usually used [5,6]:

\[
f(x) = \frac{1}{x S \sqrt{2\pi}} \exp \left( -\frac{\ln^2(x/D_0)}{2S^2} \right),
\]

where \( x \) is the magnetic diameter and \( D_0 \) and \( S \) are the parameters to be determined.

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Chantrell proposed the following relation for magnetization [5,7]:

\[ M_L = M_0 \int_0^\infty L(\xi) f(x) \, dx. \]  

(2)

Taking into account the dependence of magnetic moments on magnetic diameters, the magnetization can be written in the form [8,9]

\[ M_L = n \int_0^\infty m(x)L(\xi)f(x) \, dx, \]  

(3)

where \( n \) is the particle number density, \( \xi \), according to its definition, is also a function of \( x \).

Alternatively, the “gamma” distribution function was used due to the stronger decrease in the region of large diameters [8,9]:

\[ f(x) = \frac{x^S \exp(-x/D_0)}{D_0^{S+1} \Gamma(S+1)}, \]  

(4)

where \( S \) and \( D_0 \) must be determined and \( \Gamma \) is the \( \Gamma \)-Euler function.

A first-order perturbation theory (Ivanov’s model), takes into account the dipole-dipole interactions and gives a correction term to the Langevin magnetization \( M_L \), according to the following equation [9,10]:

\[ M = M_L \left( 1 + \frac{1}{3} \frac{\partial M_L}{\partial H} \right). \]  

(5)

Taking into account the dimensional distribution, the magnetization becomes:

\[ M = n \int_0^\infty m(x) \left( \coth \frac{1}{\xi} \right) f(x) \, dx \times \left[ 1 + \frac{n\mu_0}{3k_B T} \int_0^\infty m^2(x) \left( \frac{1}{\xi^2} - \frac{1}{\sinh^2 \xi} \right) f(x) \, dx \right]. \]  

(6)

The initial susceptibility is given by

\[ \chi_i = \chi_{iL} \left( 1 + \frac{1}{3} \chi_{iL} \right). \]  

(7)

The area of applicability of this model is determined by the width of the distribution or dimensions of the particles (in fact by the ratio between the dipole-dipole energy and thermal energy, which should be smaller than unity).

It usually applies to moderately concentrated ferrofluids. For high concentrations, van der Waals and steric (or electrical) interactions and agglomerate formation respectively must be taken into account.

A simpler method of taking into account interactions, replaces the applied field with the effective field according to Weiss’ model (mean effective field model):

\[ H_e = H + \eta M, \]  

(8)

where the Lorentz value of 1/3 can be considered for \( \eta \). \( H \) is the applied field, corrected with the demagnetization field if necessary and \( H_e \) the mean effective field. This model can be applied for weakly and moderately concentrated ferrofluids, since the initial susceptibility is given by

\[ \chi_i = \frac{\chi_{iL}}{1 - \frac{3}{2} \chi_{iL}}, \]  

(9)

where \( \chi_{iL} \) is the Langevin initial susceptibility, and no phase transition occurs experimentally for \( \chi_{iL} \geq 3 \).

In the case of ferrofluids containing identical spherical particles which form linear chains with variable length, the Zubarev model [11] gives an equation for magnetization at low fields:

\[ M \approx \frac{x_0 m \xi}{3v_0} \exp(-\epsilon) \frac{1 + x_0}{(1 - x_0)^3}, \]  

(10)

where \( v_0 \) is the volume of one particle, \( \epsilon \) the coupling parameter (the ratio of maximum dipole-dipole energy to thermal energy) and \( x_0 \) is related to the mean number of particles per chain at zero field by the following equation:

\[ (k) = \frac{\Phi \exp(1 - x_0)}{x_0}. \]  

(11)

The volume fraction of solid particles is denoted by \( \Phi \). This model applies when \( \epsilon \gg 1 \) and \( \epsilon \gg \xi \).

**Magneto-granulometric analysis.** The mean magnetic diameter and the standard deviation of particles can be determined from magnetization curves if the parameters of the dimensional distribution are known. Thus, for lognormal distribution, one obtains

\[ \langle D_m^p \rangle = D_0^p \exp \left( \frac{p^2 S^2}{2} \right), \]  

(12)

\[ \sigma = \sqrt{\langle D_m^2 \rangle - \langle D_m \rangle^2} = \frac{D_0 \exp \left( \frac{S^2}{2} \right)}{\langle \exp S^2 - 1 \rangle^{1/2}}. \]  

(13)

Here \( \langle D_m^p \rangle \) is the mean value of the magnetic diameter \( D_m \) to the \( p \)-th power, \( \sigma \) is the standard deviation from the mean magnetic diameter \( \langle D_m \rangle \) and \( D_0 \) and \( S \) are the parameters of the lognormal distribution. In the case of the “gamma” distribution, there results

\[ \langle D_m^p \rangle = D_0^p (S + 1)(S + 2) \cdots (S + p), \]  

(14)

\[ \sigma = D_0 (S + 1)^{1/2}. \]  

(15)

Here \( D_0 \) and \( S \) are the parameters of the “gamma” distribution function.

The Chantrell method [5] (based on Eq. (2)), for determining the parameters of the lognormal distribution in the case of noninteracting particles, is well known and widely used [6,7,12,13]:

\[ S = \frac{1}{3} \ln \frac{3 \chi H_0}{M_S}, \]  

(16)

\[ D_0^3 = \frac{18k_B T}{\mu_0 \pi M_d} \frac{\chi_i}{3H_0 M_e}. \]  

(17)