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MAGNETIC PERMEABILITY OF INVERSE FERROFLUID EMULSION: AN INFLUENCE OF INTERDROPLET INTERACTION

I.M. Subbotin

Ural Federal University, 51 Lenin ave., 620000 Ekaterinburg, Russia

A theoretical model has been developed to describe the weak-field growth of the magnetic susceptibility of inverse ferrofluid emulsion provided the interdroplet interaction is taken into account. The presented mean-field approach predicts a nonlinear dependence of the initial magnetic susceptibility on the droplet concentration and provides a good agreement with experimental data.

Introduction. The inverse ferroemulsion is a colloidal suspension of non-magnetic liquid droplets suspended in a magnetic fluid. The characteristic droplet diameter is several micrometers (1–10 μm). Under the influence of an external magnetic field, the carrier magnetic fluid is magnetized, and the droplets are deformed, stretching along the direction of the applied field. This behavior is pronounced for emulsions with rather weak values of the interfacial tension ($\sim 10^{-6}$ N/m). The effect of droplet deformation can be characterized by the minimization of the ferroemulsion free energy, which is accompanied by the decrease of the demagnetizing fields produced by nonmagnetic drops. These fields are determined by the shape of the droplets and decrease with the droplet elongation along the field. Thus, the degree of the droplet deformation can be defined from the minimization condition of the total free energy of the sample.

The experimental observations showed that the effect of droplet deformation is more pronounced under weak external magnetic fields ($H_0 \sim 0 - 0.5$ kA/m): being exposed to a weak field, the magnetic permeability of emulsions increases and reaches a maximum value [1], [2]. The subsequent decay of the magnetic permeability with the field is due to a decrease of the magnetic permeability of the magnetic fluid itself, with a practically complete stop of the droplets further elongation [5]. A similar behavior is observed for direct emulsions, in which the droplets are filled with a magnetic fluid and dispersed in a non-magnetic carrier [1]–[4].

In this paper, we present a mathematical model of the magnetic permeability of inverse ferroemulsions under the influence of a weak uniform external field.

1. Effective magnetic field. The main approaches of our model are the following:

- An external magnetic field $\mathbf{H_0}$ is assumed to be weak enough to provide the linear ferrofluid magnetization law with a constant magnetic susceptibility χ_f.
- All ferrofluid droplets are assumed to be of equal volume V_d. The shape of the stretched droplets is modelled by the elongated ellipsoid of revolution. In this case, the internal magnetic field H_in inside the droplet is uniform.

For the solution of the magnetostatic problem, the determination of the magnetic field profile inside the ferroemulsion sample demands for an ensemble of randomly displaced droplets. This problem cannot be solved analytically. However, the magnetostatic problem has an analytical solution for a single ellipsoidal droplet exposed to a uniform magnetic field H_0. For this case, further we use the subscripts 'in' and 'ex' for all variables indicating the internal volume of a droplet and the external vicinity. In terms of the ellipsoidal coordinates [6], [7], this solution for the magnetic potential ψ has the following form:

$$\psi_{\text{in}} = -H_0 ae\eta\tau \frac{1}{1+\left(\frac{\mu_{\text{in}}}{\mu_{\text{ex}}}-1\right) n_z(e)} \ , \tag{1}$$

$$\psi_{\text{ex}} = -H_0 ae\eta\tau \left(1 + \frac{\left(1-\frac{\mu_{\text{in}}}{\mu_{\text{ex}}}\right)\frac{1-e^2}{e^3}\left(Arth\frac{1}{\eta}-\frac{1}{\eta}\right)}{1+\left(\frac{\mu_{\text{in}}}{\mu_{\text{ex}}}-1\right) n_z(e)} \right) \ . \tag{2}$$

Here η, τ stand for the ellipsoidal coordinates, $\mu_{\text{in}}, \mu_{\text{ex}}$ are the magnetis permeabilities of the media inside and outside the droplet, e, a are the droplet eccentricity and the long semi-axis, and $n_z(e)$ stands for the ellipsoid demagnetization factor given by

$$n_z(e) = \frac{1-e^2}{2e^3}\left(\ln\left(\frac{1-e}{1+e}\right) - 2e\right) \ . \tag{3}$$

As a way to consider the collective influence of droplets on the field inside the ferrofluid, we suggest an iteration procedure similar to the one presented in [4]. The one-drop solution (1)–(2) is considered to be the zero iteration step. According to Eq. (1), the zero-iteration internal field H_{in}^0 is related to the external field as

$$H_{\text{in}}^0 + \left(\frac{\mu_{\text{in}}}{\mu_{\text{ex}}}-1\right) n_z(e^0) H_{\text{in}}^0 = H_0 \ . \tag{4}$$

Next we summarize the magnetic field perturbations produced by all randomly positioned droplets at an arbitrary point of the ferrofluid volume and add this averaged collective perturbations to the external field to obtain an effective magnetic field inside the ferrofluid volume:

$$H_{\text{ex}}^1 = H_0 + \varphi\left(\frac{\mu_{\text{in}}}{\mu_{\text{ex}}}-1\right) n_z(e^0) H_{\text{in}}^0 \ , \tag{5}$$

where φ stands for the droplet volume concentration. The next iterations are based on the idea that a random droplet is influenced by the effective field H_{ex}, instead of H_0. The j-th iteration step is

$$H_{\text{in}}^j + \left(\frac{\mu_i}{\mu_e}-1\right) n_z(e^j) H_{\text{in}}^j = H_0 + \varphi\left(\frac{\mu_{\text{in}}}{\mu_{\text{ex}}}-1\right) n_z(e^{j-1}) H_{\text{in}}^{j-1} \ , \tag{6}$$

$$H_{\text{ex}}^{j+1} = H_0 + \varphi\left(\frac{\mu_{\text{in}}}{\mu_{\text{ex}}}-1\right) n_z(e^j) H_{\text{in}}^j \ . \tag{7}$$

This iterative procedure is proved to be converging, and the limit is

$$H_{\text{in}} + (1-\varphi)\left(\frac{\mu_{\text{in}}}{\mu_{\text{ex}}}-1\right) n_z(e) H_{\text{in}} = H_0 \ , \tag{8}$$

$$H_{\text{ex}} = H_0 \left(1 + \frac{\varphi\left(\frac{\mu_{\text{in}}}{\mu_{\text{ex}}}-1\right) n_z(e)}{1+(1-\varphi)\left(\frac{\mu_{\text{in}}}{\mu_{\text{ex}}}-1\right) n_z(e)} \right) \ . \tag{9}$$

These equations define the magnetic field H_{in} inside the droplets and the mean field H_{ex} inside the carrier ferrofluid as functions of the external field H_0 and droplet eccentricity e. It is worth mentioning that the mean field is uniform and it is align with the external one.

2. Droplet elongation. To close the set (8)–(9), the droplet eccentricity should be calculated independently for each given value of H_0. We use here the minimization of the ferroemulsion free energy,

$$F_{\rm sys} = N\gamma 2\pi \left(\frac{3V_{\rm d}}{4\pi}\right)^{2/3} S_{\rm d}^{*}(e) - NV_{\rm d}\int\limits_{0}^{H_0}\chi_{\rm in}H_{\rm in}{\rm d}H_0 - (V - NV_{\rm d})\int\limits_{0}^{H_0}\chi_{\rm ex}H_{\rm ex}{\rm d}H_0\ , \quad (10)$$

where V stands for the sample volume, N is the number of droplets, obviously, $\varphi = NV_{\rm d}/V$, $2\pi\,(3V_{\rm d}/4\pi)^{2/3}\,S_{\rm d}^{*}(e)$ is the surface area of the ellipsoidal droplet, and γ stands for the interfacial tension at the droplet boundary. The emulsion free energy (10) is defined for arbitrary values of the magnetic susceptibilities of the dispersed fluid ($\chi_{\rm in}$) and carrier liquid ($\chi_{\rm ex}$). The free energy minimum yields

$$R_{\rm d}\frac{H_0^2}{3\gamma} = -4\pi\frac{\dfrac{\partial S_{\rm d}^{*}(e)}{\partial e}}{\dfrac{\partial n_z(e)}{\partial e}}\frac{(\mu_{\rm ex} + (1-\varphi)(\mu_{\rm in} - \mu_{\rm ex})n_z(e))^2}{(1-\varphi)(\mu_{\rm in} - \mu_{\rm ex})^2\mu_{\rm ex}}, \quad (11)$$

where $R_{\rm d}$ stands for the radius of a non-deformed spherical droplet.

Finally, the set of Eqs. (8)–(9) and Eq. (11) result in an expression for the magnetic permeability μ_{em} of the ferrofluid emulsion in the weak-field limit:

$$\mu_{em} = \mu_{\rm ex} + \mu_{\rm ex}\frac{\varphi\,(\mu_{\rm in} - \mu_{\rm ex})}{\mu_{\rm ex} + (1-\varphi)\,(\mu_{\rm in} - \mu_{\rm ex})\,n_z(e)}\ . \quad (12)$$

For the case of spherical droplets ($n_z(e=0)$=1/3), this expression (12) coincides with the known Maxwell–Wagner formulae [8]–[9] derived first for dielectric suspensions. We may assume this coincidence as a substantiation of the suggested effective field method.

The comparison of the presented theory and experimental observations [1], [2] is illustrated in Figs. 1 and 2 for a ferrofluid emulsions characterized by the following parameters: $4\pi\chi_{\rm f} = 5.4$, $\chi_{\rm ex} = \chi_{\rm f}$, $\mu_{\rm in} = 1$, $R_{\rm d} = 5\,\mu m$.

The dependence of the initial magnetic susceptibility of inverse ferrofluid emulsion on the volume concentration of droplets is presented in Fig. 1. The weak-field dependence of the ferrofluid emulsion magnetic susceptibility is illustrated in

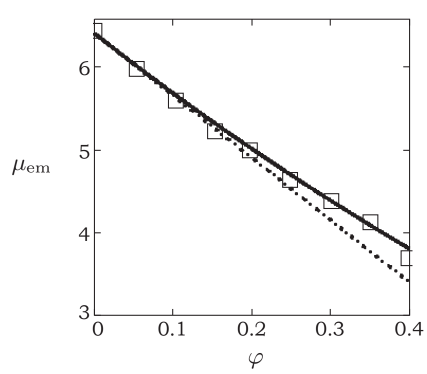

Fig. 1. Initial magnetic susceptibility of inverse ferroemulsion. Experimental data [1]–[2] are indicated with boxes. The dotted line characterizes the linear concentration dependence predicted by the model of non-interacting droplets. The nonlinear behaviour obtained on the basis of the effectively interacting droplet model Eq. (12) is given by the solid line.

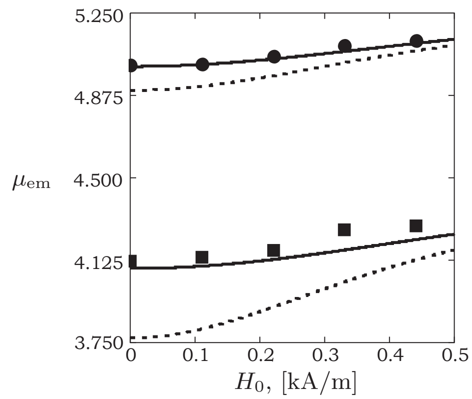

Fig. 2. Field dependence of the ferrofluid emulsion magnetic susceptibility. Experimental data [1]–[2] are presented for emulsions with the droplet volume fraction $\varphi = 0.2$ (circles) and 0.35 (boxes). Theoretical predictions are shown with lines for the same emulsions: dotted lines are for non-interacting droplets; solid lines correspond to the model.

Fig. 2. In both graphics, dots correspond to the experimental data [1],[2], dotted lines stand for the model of non-interacting droplets (zero step of the presented procedure in Eqs. (4), (5)), and solid lines illustrate the results of the present model (Eqs. (12), (11)). As it is clear from the figures, if compare with the approximation of non-interacting droplets, the consideration of the interdroplet interaction improves the agreement of theory and experimental data.

3. Conclusion.

- The nonlinear behaviour of the initial susceptibility of inverse ferroemulsions is caused by the influence of interdroplet interactions.
- The presented simple model describes qualitatively good the dependence of the growing magnetic permeability of ferrofluid emulsion on the weak external magnetic field.
- The results of the presented mean-field approach to consider the interdroplet interaction provide a good agreement with the experimental data.

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