

Self-diffraction of the light in magnetic fluids *)

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Some results of the experimental investigation of the self-diffraction of two interfering beams in thin film samples of kerosene-based magnetic fluids are presented. The creation of a space periodic temperature distribution due to the interference field is described. Through thermo-diffusion the magnetic particles density distribution is formed. The time dependence of the diffracted beam intensity after switching off the interference field is observed. It informs us about the diffusion of magnetic particles or aggregates, forming the optical grating. The relaxation time spectrum calculation was used for the determination of particle size distribution. Also an interesting phenomenon of self structuralization of magnetic particles after application of a homogenous and strong illumination of the magnetic fluid sample was observed.

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1 Introduction

The self-diffraction may occur when two coherent light beams pass through a sample of light absorbing medium. The periodic heating caused by the intensity distribution in primary beams interference field induces the temperature changes. This temperature modulation is supposed to cause the diffraction of primary beams due to the temperature dependence of refractive index, [1], or (in colloidal liquids) due to the redistribution of colloidal particles within the temperature gradient [2, 3], known as the Soret effect. To find out the role of these two mechanisms the self-diffraction effect was studied in magnetic fluids based on fine Fe_3O_4 particles (approximately 10 nm in diameter) dispersed in various media.

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2 Theory

A simple theoretical analysis of the temperature distribution caused by a non-uniform lighting was based on the calorimetric equation. The amplitude of periodic temperature distribution in thin sample was obtained as

$$T_0 = \frac{2I_0\alpha}{(\lambda_g\Omega^2 + \lambda')}, \quad (1)$$

where I_0 is the primary beam intensity amplitude, α the light absorption coefficient in the studied liquid, λ_g the thermal conductivity coefficient, Ω the intensity distribution space frequency equal to $2\pi/D$ (D being the space period of interference field) and λ' is the heat outlet coefficient. For typical parameters of magnetic fluids the analysis gives time constant of the thermal relaxation in interference field with a space period of $D \sim 100\mu\text{m}$ being of one milisecond order. The spatial temperature modulation induces (through the thermodiffusion) a modulation of the particle space distribution. The influence of the temperature distribution upon the particle distribution can be determined by means of the diffusion equation in form

$$\frac{\partial n}{\partial t} = D_{\text{dif}} \frac{\partial^2 n}{\partial x^2} + SD_{\text{dif}} n(x, t) \frac{\partial^2 T}{\partial x^2}. \quad (2)$$

Here n is the colloidal particle concentration, D_{dif} is the particle translational diffusion coefficient and S is the thermodiffusion Soret coefficient. The diffusion of particles is much slower than the heat diffusion, so from equation (2) (when changes of n are smaller than its mean value) and from the Stokes-Einstein formula for the particle translational diffusion coefficient $D_{\text{dif}} = kT/3\pi\eta d_{\text{H}}$ it follows, that the time constant of the particle density relaxation from an inequilibrium state, created by temperature distribution, to an equilibrium one is

$$\tau = \frac{3\eta d_{\text{H}} D^2}{4\pi kT}. \quad (3)$$

The quadratic dependence of τ on a created lattice constant D is evident in this equation. Here η is the carrier viscosity and d_{H} the hydrodynamic particle diameter. As it is seen in equation (3), the relaxation time constant τ depends upon the hydrodynamic particle diameter d_{H} . The relaxation phenomena are monoexponential for monodisperse particles, but multiexponential in magnetic fluids with a real particle size distribution and can be expressed as

$$I(t)/I(0) = \int \exp(-t/\tau_d) p(\tau_d) d\tau_d, \quad (4)$$

where $p(\tau_d)$ is the distribution of time constants corresponding to the particle size distribution. The diffracted beam intensity I is proportional to n^2 , so its relaxation time $\tau_d = \tau/2$. In this sense the relaxation measurements of the diffraction intensity curves allow us to determine the particle size distribution function.

3 Experiment and discussion

The experimental set-up used for the self-diffraction investigation consists of the Zeiss Ar laser ILA 120, a beam splitter allowing to adjust the angle α between the beams in the range from 0.002 to 0.1 rad, a mechanical shutter allowing the interruption of the laser beam during 0.1 ms, a sample of kerosene-based magnetic fluid and a photodetector connected to PC. Used magnetic fluid samples were in form of thin films with thicknesses in the range from 10 to 30 μm . Particle diameters were estimated by magnetization measurements and transmission electron microscopy (TEM). The mean values were found to be $d_m = 9.8$ nm and $d_{\text{TEM}} = 11.8$ nm, respectively. Fig.1 presents the time dependence of diffracted beam intensity

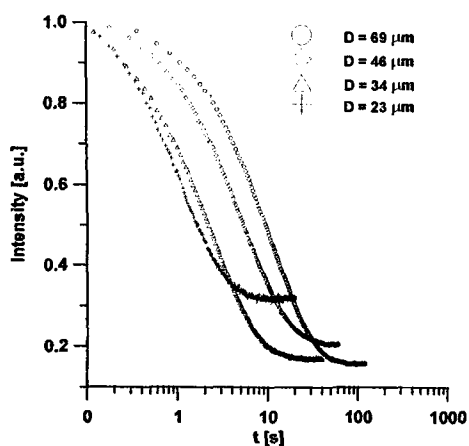


Fig. 1. The time dependence of the relative intensity $I(t)/I(0)$ of diffracted beam for different lattice constants

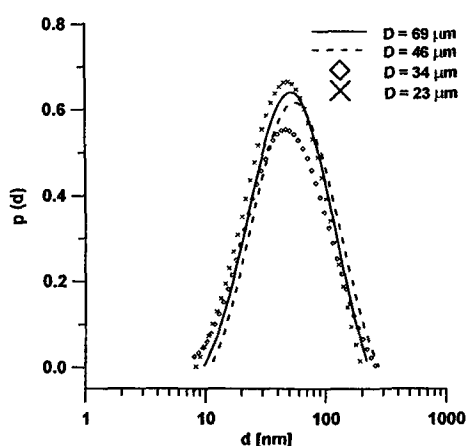


Fig. 2. The corresponding particle size distributions

for four values of lattice constants D . The corresponding particle size distributions obtained by means of equations (3) and (4) are presented in Fig.2. The obtained mean hydrodynamic diameter d_H was compared with the mean diameters estimated by TEM and by magnetization measurements. The ratios $d_H/d_{\text{TEM}} = 2.35$ and $d_H/d_m = 2.83$ were found. These ratios are quite good and fall into the range of measured values, which reaches from 2 to 15 [4]. The quadratic dependence of the mean relaxation time of diffracted beam after switching off one of interfering beams is presented in Fig.3. According to equation (2) an interesting phenomenon can be predicted for magnetic fluids with negative Soret coefficient [5]: in such a case the fluctuation of absorption at homogenous illumination may spontaneously increase because it evokes a change of heating which is connected with further changes of particle density. It can give rise to a self-structuralization of the particles density in such liquids. An example of such structuralization is given in Fig.4.

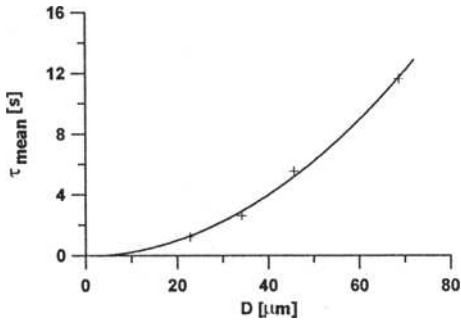


Fig. 3. The dependence of mean relaxation time of diffracted beam on the lattice constant; full line represents the best quadratic dependence

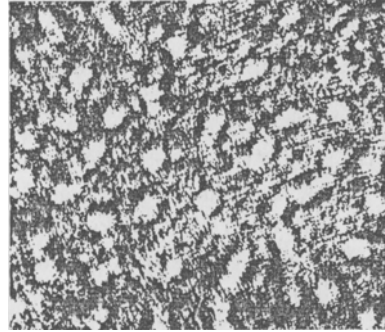


Fig. 4. Example of structuralization of particle density; $1mm$ displayed corresponds to $40\mu m$.

4 Conclusion

The performed experiments confirm, that the self-diffraction in the investigated magnetic fluids is connected with both the particles density and the refractive index thermal modulation. The quadratic dependence of mean relaxation time τ upon the created lattice constant D was confirmed according to the theoretical prediction. The observed hydrodynamic diameter of magnetic particles is approximately twice as large as the diameters obtained by TEM and the magnetic measurements. This value is very good in comparison with other measurements. The observed value of d_H confirms a good colloidal stability of used magnetic fluid, i.e. no aggregates are present. It is also shown that the self-structuralization is caused by a negative Soret constant.

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