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THE DYNAMICS OF THE SORET EFFECT IN THIN FILM OF MAGNETIC FLUID

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The Soret effect in interference field of two intensive laser beams in the thin film of magnetic fluid was used to create a periodical structure of density of magnetic particles. The structures obtained were indicated using the self-diffraction of the optical beam creating the structures. The relaxation phenomena after switching off the laser interference field were discussed in terms of the spectrum of relaxation times. This spectrum is proportional to hydrodynamic particle size distribution. The relaxation of optical grating consists of two well-defined channels, i.e. decay through single particles and small aggregates, respectively.

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1. Introduction

The concentration variations of particles due to the laser-beam induced by thermal diffusion in liquid mixtures (known as the Soret effect) were observed in a number of works [1–4]. The theoretical aspects of the Soret effect in fluids containing nanoparticles were studied by Tabiryan and Luo [5]. The concentration optical grating of fine magnetic particles in magnetic fluids due to their interaction with an intense laser beam was first experimentally proved by Bacri et al. [6] by means of a forced Rayleigh scattering (FRS) experiment. The dynamics of the observed effect was discussed in terms of a single relaxation time and the cooperative diffusion coefficient of the magnetic particles. This discussion of the results is possible in the situation when a size of magnetic particles is monodisperse. The size distribution of magnetic particles in magnetic fluids are never monodisperse. It means that the results should be discussed in terms of spectrum of relaxation times due to the polydispersity of particle sizes. The aim of this work was to study the dynamics of the particle density distribution formed by the Soret effect in magnetic fluids to develop the method for calculation of the spectrum of relaxation times and finally to compare this spectrum with the real particle size distribution of magnetic particles used in the experiment.

2. Experiment and methods

We have used petroleum-based magnetic fluid with fine magnetic particles of Fe_3O_4 prepared by the well-known precipitation technique. As a surfactant to prevent the aggregation of particles oleic acid was used. The volume concentration of magnetic particles was $\varphi = 0.05$. The particle size distribution was analyzed using an electron micrograph. Electron microscopy showed roughly spherical particles and the best log-normal fit of the particle size distribution has the parameters as: a mean diameter of particle size $d_{\mathrm{mm}} = 11.7$ nm and a standard deviation $\sigma = 0.3$. The magnetic measurements showed a mean size of particles $d_{\mathrm{mm}} = 9.3$ nm and $\sigma = 0.28$. For the experiment we have used an optical cell 80 μm thick filled by the magnetic fluid. The interference field in the forced Rayleigh scattering experiment was created by two coherent intensive laser beams ($\lambda_g = 0.514$ μm) crossing the above-mentioned optical cell. These beams were obtained by splitting a beam from a Zeiss argon laser ILA 120. The used laser power was 50 mW. These two pump laser beams intersect in the magnetic fluid with a definite angle α building up a spacially periodic intensity distribution in the sample $I(x) = 2I_0\,[1 + \cos(qx)]$, with $q = 2\pi/\Lambda$ and $\Lambda = \lambda_g/\,[2\sin(\alpha/2)]$. The Λ is the interfringe distance (lattice constant of the created optical grating).

When the two laser beams interfere for a few seconds an optical grating (the concentration variations of magnetic particles) was easily observed by the presence of the self-diffraction effect of the primary laser beams. If one of the two pump laser beams is switched off, the concentration optical grating smeared out in a few seconds. The dynamics of this process was observed through the intensity measurement of the first order of the diffracted pattern of the above-mentioned self-diffraction effect. The relaxation curve of this first order diffracted signal should be in principle a single decay

$$I(t)/I(0) = \exp(-t/\tau), \tag{1}$$

where τ is the relaxation time. The relaxation due to the thermal relaxation of created positions of particles to random positions is according the Perrin law [7] given by

$$\tau(d) = \pi d^3 \eta / 3k_{\mathrm{B}}T, \tag{2}$$

where d is the particle diameter and η is the carrier fluid viscosity. It should be noticed that this diameter determined from this experiment is the hydrodynamic diameter d_{H}. The above-mentioned consideration is valid in the case of monodisperse magnetic particles only. Owing to the particle size distribution, $I(t)/I(0)$ is not a simple exponential function of time. Taking into consideration polydispersity of particles, the time dependence of the intensity is given by the following formula:

$$I(t)/I(0) = \int_0^\infty \exp(-t/\tau)p(\tau)\mathrm{d}\tau, \tag{3}$$

where $p(\tau)$ is a spectrum of relaxation times. As the time constant τ is a function of d (see Eq. (2)), the function $p(\tau)$ is proportional to the hydrodynamic particle size distribution function. For the calculation of the spectrum of relaxation times $p(\tau)$, the method based on the discrete fast Fourier transformation was used [8]. This method is well known for calculation of the activation energy spectrum for the deformation processes in amorphous metals [9].

3. Results and discussion

The self-diffraction experiment was performed for three values of angle α corresponding to various lattice constants of the optical grating obtained, i.e. $\Lambda =$ 16 μm, 25 μm, and 49 μm, respectively. Figure 1 illustrates the experimental relaxation signal of the first order of the diffracted pattern after switching off the laser interference field. By means of the above-described technique we calculated the corresponding spectrum of relaxation times (Fig. 2). Two well-defined maxima are seen in this figure.

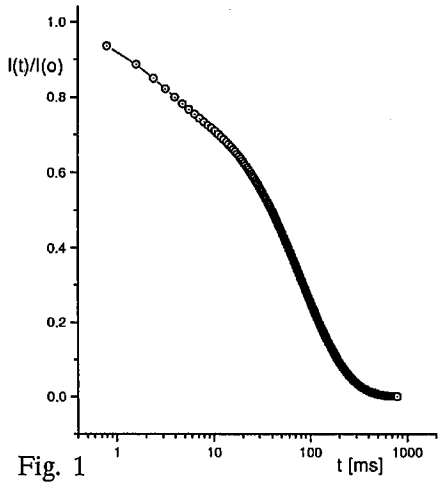

Fig. 1

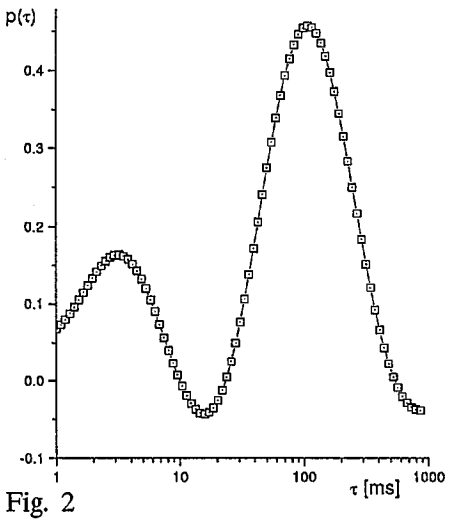

Fig. 2

Fig. 1. The relaxation signal of the relative intensity $I(t)/I(0)$ of the first order of diffracted pattern for the lattice constant $\Lambda = 25$ μm.

Fig. 2. The corresponding spectrum of relaxation times.

TABLE

The parameters of the spectrum relaxation times for studied lattice constants.

Λ [μm]	τ_1 [ms]	d_{H1}/d_{mp}	τ_2 [ms]	d_{H2}/d_{mp}
16	2.4	15.0	88.8	50.0
25	3.0	16.1	104.1	52.7
49	2.8	15.7	91.5	50.5

Experimental results are summarized in Table, where the ratios of the hydrodynamic diameters of the calculated maxima to the mean particle size diameter ($d_{mp} = 11.7$ nm) are given. These values for the calculated maxima are nearly the same for various lattice constants being in agreement with the fact that we used one magnetic fluid in the experiment. The hydrodynamic sizes d_H deduced from

these measurements are much longer than the sizes determined from electron microscopy or magnetization measurement d_m. The disparity in diameter d_H/d_m may vary by a factor from 2 to 15 [10]. The experimental values of d_H/d_m were measured ($d_H/d_m = 15$) for kerosene [11] and ($d_H/d_m = 11$) for mineral oil [12] based on magnetic fluids for example. In the experiment we have used the petroleum-based magnetic fluid which is similar to the kerosene-based magnetic fluid, i.e. the observed ratio of the first maximum ($d_H/d_m = 15.0 \div 16.1$) is in agreement with the previous experiments. The obtained value of the second maximum d_H/d_m is in the range of 50.0–52.7. This situation can correspond to small clusters of magnetic particles. The main result of this work is to develop a new method of calculation of the hydrodynamic particle size distribution function in magnetic fluid by means of the study of relaxation phenomena. From the results we can conclude, which processes are responsible for the relaxation phenomena.

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