EXPERIMENTAL INVESTIGATIONS ON THE MICROCONVECTIVE INSTABILITY IN OPTICALLY INDUCED GRATINGS

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Introduction. Forced Rayleigh scattering experiments (FRS) are well known as a tool to measure diffusion D and Soret S_T coefficients [1, 2]. In FRS experiments the temperature grating is optically induced in a thin sample layer by focusing a periodic pattern (usually one-dimensional parallel stripes) of intense illumination. Employing ferrofluid samples, due to the Soret effect, primary induced temperature grating creates a particle concentration grating. The steady-state of the concentration grating is examined to determine S_T, whereas D may be easily calculated by knowing the half-period a of the induced grating and by measuring relaxation time τ_D of the concentration grating from its vanishing stage, after the illumination source is switched off.

Recent theoretical investigations on the microconvective instability of an inhomogeneous magnetic fluid in a Hele–Shaw cell [3, 4] predict the presence of microconvection in FRS experiments under certain conditions. Particularly, the value of a dimensionless number Cm is considered (here modified to the SI system of units):

$$\mathrm{Cm} = \mu_0 \frac{(\delta c m_0)^2}{48\pi\eta D}\, h^2 \qquad (1)$$

where δc is the modulation of the particle volume concentration, m_0 is the average particle magnetic moment component along an externally applied field, η is the viscosity and h is the cell thickness.

The author of Ref. [3] deals with an idea that the microconvective instability may develop in FRS experiments as soon as the illumination source is switched off (vanishing stage of the concentration grating). Indeed, there are evidences from FRS experiments with the applied field geometry $\nabla T \parallel \mathbf{H}$ that under certain conditions τ_D turns out to be dependent on the cell thickness h [5]. The observed dependence on h disagrees with the conception of magnetophoresis in the self-magnetic field as the only acting effect [6, 7]. Another observed effect is a notable decrease and instability of the steady-state level of concentration grating. These in the evolution of concentration grating observed side effects raise a question whether they are caused by the microconvection. The answer can be searched by proper calculation of Cm, where the responsible quantities are taken from the studied experiments. Nevertheless, there is a general problem in obtaining the values of δc and D in the presence of magnetic field, since the steady-state and the vanishing of the concentration grating are affected by the above mentioned effects. In the frame of defined problem, the only way to get correct values of δc and D in magnetic field experiments is to define them from the building stage of the concentration grating.

1. Building stage of the concentration grating. A theoretical model of building of the concentration grating with respect to δc is developed in Ref. [8]. In FRS experiments instead of δc the first order diffracted intensity I_d is measured, which has a square root link with δc [9]: $I_d \propto \delta c^2$. Simplified to the first Fourier harmonic for the one-dimensional case, one can write [8]:

$$\sqrt{I_d(t)} = A\Theta\big(1 - \exp(-\pi^2 d_\mathrm{m}\tau)\big)\,, \qquad A = 2kc_0 \frac{\mathrm{S_T} Q_0 a^2}{\lambda} \frac{s_\mathrm{m}}{d_\mathrm{m}} \frac{\sin \pi k}{\pi^2}\,, \tag{2}$$

where Θ is a photometric experimental constant, k is the proportion constant of the grating, c_0 is the initial particle volume concentration, Q_0 is the volumetric density of the heat input, λ is the heat conductivity, and Fourier time $\tau = Dt/a^2$, t being the real time. D and $\mathrm{S_T}$ are zero-field values, and the field dependent coefficients s_m and d_m stand for their respective values in the magnetic field: $D(H) = d_\mathrm{m} D$ and $\mathrm{S_T}(H) = s_\mathrm{m}\mathrm{S_T}$.

2. Processing of the experimental data. The goal of the processing is to calculate Cm from FRS experiments, performed at various magnetic field strengths, with the induced grating of two different sizes, and to compare the obtained values with the theoretically calculated neutral (threshold) curves of expected microconvection [3].

In the proposed processing, the parameters a, k, c_0, λ and Q_0 are taken as known, and $\mathrm{S_T}$ is determined by means of usual grating steady-state measurements in zero-field [1, 2, 9]. Equation (2) is rewritten in the following form:

$$\frac{1}{t} \ln\left(1 - \frac{\sqrt{I_d(t)}}{A\Theta}\right) = \frac{\pi^2 d_m D}{a^2}\,. \tag{3}$$

The experimentally taken $I_d(t)$ curve may be plotted in the co-ordinates, where the y-axis represents the left side of Eq. (3) versus time t on the x-axis. As long as the building of the concentration grating obeys Eq. (3), the curve must lie on a horizontal line (y=const) because the right side of that equation contains only the parameters, which are constant during a single FRS experiment. As the result of performed alignment (by searching the most appropriate value of $A\Theta$), the value of a constant level C and duration t' of the building of the concentration grating in accordance with Eq. (2) are found. Afterwards, the value of $d_\mathrm{m}D$ can be defined in a very simple way from Eq. (3): $d_\mathrm{m}D = -Ca^2/\pi^2$. Obtaining D demands a zero-field FRS experiment, at which $d_\mathrm{m} = s_\mathrm{m} = 1$. Moreover, a particular value of $A(d_\mathrm{m} = s_\mathrm{m} = 1)$ can be calculated, which leads to obtaining the value of the photometric constant Θ, Eqs. (2, 3). Afterwards, if Θ is known and kept constant during all performed experiments, the only unknown parameter in Eqs. (2, 3) is $s_\mathrm{m} = s_\mathrm{m}(H)$, which is calculated from each particular A.

Knowing d_m, D, s_m, $\mathrm{S_T}$ allows to calculate the modulation of the particle volume concentration δc of each particular experiment at the moment of time $t = t'$ [8]. In the end, reached Cm at t' can be calculated. Unlike the author of Ref. [3], we prefer to use in Eq. (1) the respective value of the diffusion coefficient in magnetic field, $d_m D$. Since dm is obtained from the building stage of the concentration grating, before the onset of any side effect, e.g., microconvection, we may consider $d_m D$ as the true value of the diffusion coefficient in the magnetic field.

3. Results and discussion. All experiments are performed with the same cell thickness $h = 100\ \mu$m to ensure identical illumination absorption. The suspended nanoparticles of the sample (volume concentration $c_0 = 2.3\%$) are stabilized by oleic acid coating and dissolved in tetradecane. With the examined ferrofluid sample, the steady-state measurements in zero-field indicate a typical value of $\mathrm{S_T}$ with respect to the magnetic fluid of magnetite particles – organic carrier composition: 0.16 1/K. Analysis of the building stage of the concentration grating at zero-field gives the value $D = 1.18 \cdot 10^{-11}$ m^2/s. Such value is in

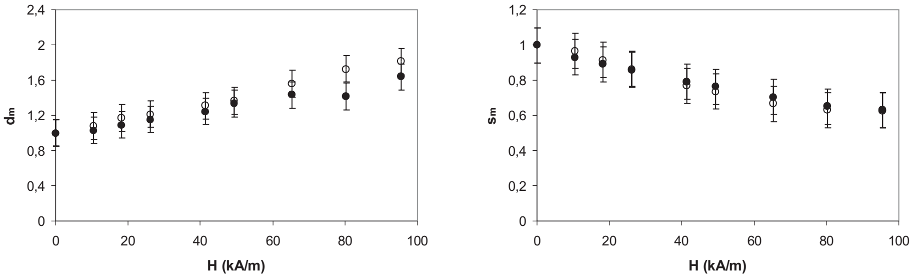

Fig. 1. The coefficients $d_{\mathrm{m}} = d_{\mathrm{m}}(H)$ and $s_{\mathrm{m}} = s_m(H)$ with the induced grating of two different sizes: hereafter, $2a = 22.5$ μm – white circles, $2a = 30$ μm – black circles.

good agreement with the Einstein–Stokes formulae, counting spherical particles, the diameter of which is $10\ldots12$ nm.

In Fig. 1 the coefficients d_{m} and s_{m}, obtained by the above described method, are plotted. We see a rather good coincidence of the obtained values between two different sizes of the induced grating. Fig. 2 displays the multiplication $dmsm$, proving to be very weak dependent on the magnetic field. This agrees with the developed theoretical model [10], which predicts the shift of $dmsm$ at the field strength of 100 kA/m only ca. 1%. Thus the present experimental work confirms the theoretical foundings of a weak dependence of the thermodiffusion coefficient, which is widely used and defined as $D_T = D(H) \cdot \mathrm{S_T}(H)$, on the magnetic field strength. Based on this, one can conclude that respectable changes of the Soret coefficient $\mathrm{S_T}$ in the presence of magnetic field must be taken as the response to that of the diffusion coefficient D (the latter is proved theoretically as well as experimentally, e.g., Ref. [7]). As the final result of the processing, the left plot in Fig. 3 shows Cm calculated from the performed FRS experiments. It is seen that the congruence between experiments and theory is better in the case of the induced grating of larger period ($2a = 30$ μm). Experiments at the weakest applied magnetic field ($H = 10$ kA/m) provide the values of Cm, which are obviously below a certain level, reached in stronger fields. It can be easily explained by supposing that in such a weak field the building of the concentration grating ends mainly due to the same reason as in zero-field experiment: the equilibrium between the induced by the Soret effect concentration difference and particle diffusion sets in: $\mathrm{S_T}c_0(1-c_0)\nabla T = -\nabla c$. The same evidence gives inspecting the experimentally taken curves I_d: in weak fields the decrease of the steady-state level of concentration grating is smaller (in zero-field experiments there is no decrease at all).

The most important experimental drawback seems to be a non uniform heating over the sample thickness due to absorption of illumination. The employed

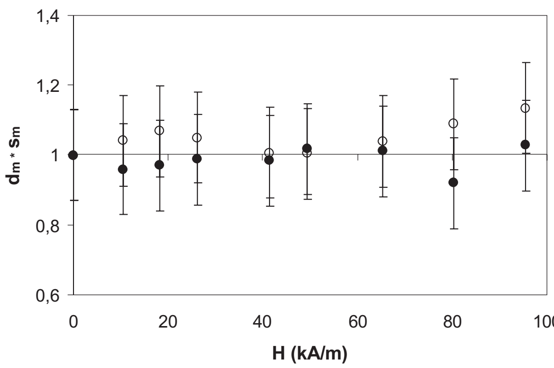

Fig. 2. The multiplication $d_{\mathrm{m}}s_{\mathrm{m}}$ at different magnetic field strengths.

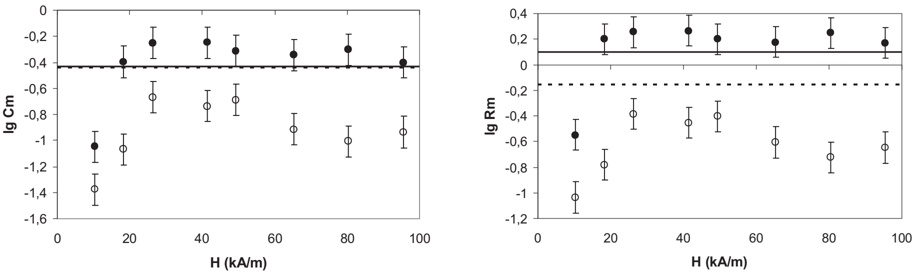

Fig. 3. Calculated Cm and Rm vs. the applied magnetic field. The lines represent theoretically calculated neutral curves, Ref. [3]: $2a = 22.5$ μm – dashed line, $2a = 30$ μm – solid line.

magnetic fluid sample absorbs ca. 6% of the illumination, it means that the heating at the exit from a 100 μm thick layer is less than a half of the initial one. With regard to this, one may consider the half-period a of the induced grating instead of the cell thickness h as the characteristic space scaling in the dimensionless number responsible for the onset of microconvection (there is an incorrectness in translation the theoretical solution with regard to non-slip boundaries to free ones, though). Thus the given above Cm turns to the magnetic Rayleigh number $\mathrm{Rm} = \mu_0(\delta c m_0 a)^2/(\eta d_x D)$, Ref. [10].

All data from the left plot of Fig. 3 are recalculated to Rm and shown in the right plot. Although the theoretically calculated neutral curves alienate themselves, the congruence between experiments and theory remains approximately the same.

4. Conclusion. The performed FRS experiments with an applied magnetic field ($\nabla T \parallel \mathbf{H}$) are characterized by some notable side effects: the decrease and the instability of the steady-state level and non-exponential decay in the vanishing stage of the concentration grating. The comparison between experiments and theory of the dimensionless numbers Cm and Rm, which are responsible for the onset of microconvection, can be taken as quite successful to conclude that the above mentioned side effects are caused by the presence of microconvection. An excellent fit between experiments and theory in the present work cannot be achieved due to some significant discrepancy: e.g., the theory considers a Hele–Shaw cell, whereas in experiments the third dimension is of particular importance.

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