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Concentration nonlinearity of a suspension of transparent microspheres under the action of a gradient force in a periodically modulated laser field

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Abstract. **Based on a one-dimensional Smoluchowski equation we have developed the theory of concentration nonlinearity of a suspension of transparent microspheres under the action of a gradient force in an interference laser field. The numerical solution of a system of recurrence equations resulting from the Smoluchowski equation after expansion of the microsphere concentration $N(z, t)$ in the harmonic series has allowed us to determine the dependence of the concentration nonlinearity settling time on the intensity of the incident radiation. In the diffusion limit, we have derived the expression for the optical Kerr coefficient, which is found to be 8.5×10^{-10} cm^2 W^{-1} for an aqueous suspension of latex microspheres with a radius of 1.17 μm and a concentration of 6.5×10^{10} cm^{-3}. Diffraction of a probe wave on a light-induced concentration grating is considered as a method for studying a nonlinear concentration response of an artificial highly efficient nonlinear medium for laser radiation of long pulse duration.**

Keywords: *Smoluchowski equation, transparent microspheres, concentration nonlinearity, diffusion limit, optical Kerr coefficient, diffraction.*

1. Introduction

In this paper, based on the Smoluchowski equation [1, 2] we have developed a theory of concentration nonlinearity of a suspension of transparent microspheres under the action of a gradient force in an interference laser field. Despite the fact that each suspension component (microspheres and liquid) exhibits no nonlinearity, this artificial heterogeneous medium is a highly efficient broadband nonlinear material for cw laser radiation [3, 4]. In the experimental study of four-wave mixing (FWM) of argon laser radiation ($\lambda = 5145$ Å) in a suspension of latex microspheres of radius $R = 1.17$ μm and concentration $N_0 = 6.5 \times 10^{10}$ cm^{-3}, the measured optical Kerr coefficient n_2 was found equal to 3.6×10^{-9} cm^2 W^{-1} [3], which is 10^5 times greater than that in carbon disulfide. In this case, the formation (t_f) and decay (t_d) times responsible for FWM of concentration gratings were equal to $t_f = 320$ ms (at a pump power of ~100 mW) and $t_d = 200$ ms. A long nonlinearity settling time corresponds to a general law for nonlinear media – a linear increase in settling time with increasing n_2 (see., e.g., [5]). The theory of FWM and stimulated concentration scattering in an aqueous suspension of transparent microspheres was developed in [2, 4] and [6, 7], respectively.

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2. Basic relations

2.1. Formulation of the problem

We will consider concentration nonlinearity of a suspension of transparent microspheres under the action of a gradient force F_∇ in the field of two coherent waves of equal amplitude, converging at an angle 2Θ at the boundary of a cell with a suspension (Fig. 1). In this case, the intensity of radiation in the cell is

$$I(z,t) = I_0(t)\Big[1 + \cos\Big(2\pi\frac{z}{\Lambda}\Big)\Big], \tag{1}$$

where $\Lambda = \pi/(k\sin\Theta)$ is the modulation period, and k is the wave number. A laser pulse with an intensity of $I_0(t)$ is a rectangle and has a duration τ_p:

$$I_0(t) = I_0[Y(t) - Y(t - \tau_p)], \tag{2}$$

where $Y(t)$ is the Heaviside unit function; $I_0 = \text{const}$; and $t \geqslant 0$.

To describe the evolution of the concentration $N(z,t)$ of microspheres, we will use a one-dimensional Smoluchowski equation [8]

$$\frac{\partial N}{\partial t} = D_0\Big[\frac{\partial^2 N}{\partial z^2} - \frac{1}{k_B T}\Big(N\frac{\partial F_\nabla}{\partial z} + F_\nabla\frac{\partial N}{\partial z}\Big)\Big], \tag{3}$$

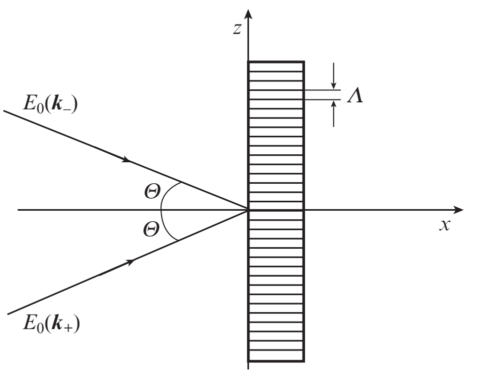

Figure 1. Scheme of excitation of a concentration grating.

where $D_0 = k_B T/(6\pi\eta R)$ is the diffusion coefficient of microspheres in a liquid with a viscosity η at a temperature T; and k_B is the Boltzmann constant. In the Rayleigh–Gans approximation [9] taking into account the nonuniformity of radiation (1) in a volume of a microsphere, $V = 4\pi R^3/3$, the gradient force in (3) is defined by expression [4, 10]

$$F_\nabla = 2\pi \frac{n}{c} \alpha I(t) \frac{1}{V} \int_V \nabla \cos\left(2\pi \frac{z}{\Lambda}\right) \mathrm{d}V, \qquad (4)$$

where

$$\alpha = R^3 \frac{\bar{m}^2 - 1}{\bar{m}^2 + 2} \qquad (5)$$

is the polarisability of a microsphere; and $\bar{m} = n_0/n$ is the ratio of the refractive indices of the microsphere material (n_0) and the liquid (n) at a laser wavelength λ (hereinafter we assume for certainty $\alpha > 0$).

2.2. Investigation of features of the gradient force

After integration in (4), we find

$$F_\nabla = -4\pi^2 \frac{n}{c} \frac{\alpha}{\Lambda} I(t)\, U(\Omega) \sin\left(2\pi \frac{z}{\Lambda}\right) \equiv -F_0 \sin\left(2\pi \frac{z}{\Lambda}\right), \qquad (6)$$

where

$$U(\Omega) = 3\sqrt{\frac{\pi}{2}}\, \Omega^{-3/2} J_{3/2}(\Omega) \qquad (7)$$

is a function, which takes into account the nonuniformity of radiation in the volume of a microsphere; $J_{3/2}(\Omega)$ is the Bessel function; and $\Omega = 2\pi R/\Lambda(\Theta)$.

Figure 2 shows angular dependences of the function $U(\Theta)$ at various radii R of microspheres. One can see that the inclusion of nonuniformity of the radiation intensity in the volume of a microsphere with increasing ratio $R/\Lambda(\Theta)$ leads to a decrease in the amplitude of the gradient force F_0 ($\Theta = \pi/2$ corresponds to the counterpropagation of waves, i.e. to a minimum modulation period $\Lambda = \pi/k$). Because the function $U(\Omega) \sim J_{3/2}(\Omega)$ is sign alternating, then for certain values of Ω the dependence $F_0(\Omega)$ changes its sign, and hence microspheres with $\alpha > 0$ can be localised in the nodes in the interference pattern of the field. At $\Omega = \Omega_i$ (where Ω_i are the roots of the Bessel function, $i = 1, 2, 3\ldots$), regardless of the position of a microsphere, the gradient force incident on it is $F_0(\Omega) = 0$. The so-called zero-force effect [4, 6] is caused by the same influence of its components on the corresponding elements of the microsphere volume in the region of its overlap by two neighbouring maxima (anti-nodes) of the interference pattern of the field. In particular, for the first root of the Bessel function $J_{3/2}(\Omega_1 = 4.493)$, the zero-force effect is achieved at $R/\Lambda(\Theta) = 0.3576$. In the region $\Omega_1 < \Omega < \Omega_2$, the amplitude is $F_0 < 0$, and therefore the microsphere will behave like a particle with $\alpha < 0$. The estimates show that the condition for manifestation of the zero-force effect in the Rayleigh–Gans approximation used here can be realised at $|\bar{m} - 1| \ll 1$ [4].

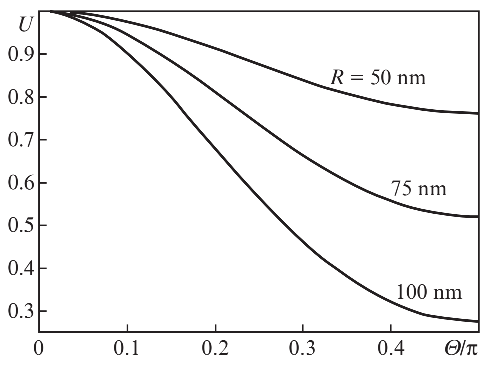

Figure 2. Angular dependences of the function $U(\Theta)$ at various values of R.

Using the well-known [11] relation

$$J_{3/2}(\Omega) = \sqrt{\frac{2}{\pi\Omega}} \left(\frac{\sin\Omega}{\Omega} - \cos\Omega\right), \qquad (8)$$

it can be shown that at $\Omega \ll 1$ taking into account the first non-vanishing term, we have

$$U(\Omega) \approx 1 - \frac{\Omega^2}{10}. \qquad (9)$$

It is obvious that the influence of nonuniformity of radiation in the volume of a microsphere can be neglected only at $R/\Lambda \ll 1$.

2.3. Analysis of the Smoluchowski equation

Introducing the variable $\xi = 2\pi z/\Lambda$, equation (3) for the function $\bar{N}(\xi, t) = N(\xi, t)/N_0$ can be written in the form

$$\frac{\partial \bar{N}}{\partial t} = \left(\frac{2\pi}{\Lambda}\right)^2 D_0 \frac{\partial^2 \bar{N}}{\partial \xi^2} + \frac{F_0}{3\Lambda R\eta}\left(\bar{N}\cos\xi + \frac{\partial N}{\partial \xi}\sin\xi\right). \qquad (10)$$

We seek the solution to equation (10) as an expansion

$$\bar{N}(\xi, t) = \sum_{\kappa=-\infty}^{\infty} \bar{N}_\kappa(t) \exp(\mathrm{i}\kappa\xi), \qquad (11)$$

in which the Hermiticity condition $\bar{N}_\kappa = \bar{N}^*_{-\kappa}$ is met. With the help of (11), equation (10) is reduced to an infinite system of recurrence equations of the form

$$\frac{\mathrm{d}\bar{N}_\kappa}{\mathrm{d}t} = -\frac{\bar{N}_\kappa}{t_\kappa} + \kappa \frac{F_0}{6\Lambda R\eta}(\bar{N}_{\kappa-1} - \bar{N}_{\kappa+1}) \qquad (12)$$

with the initial conditions $\bar{N}_0(t = 0) = 1$ and $\bar{N}_{\kappa\neq 0}(t = 0) = 0$, where $t_\kappa = 3\Lambda^2 R\eta/(2\pi\kappa^2 k_B T)$ is the time of the diffusion decay of the kth harmonic. It is obvious that in (12) odd harmonics are coupled with even harmonics and even harmonics – with odd ones. At the same time due to the fact that their amplitudes $\bar{N}_\kappa(t)$ are real, it is sufficient to consider only nonnegative values of κ ($\kappa = 1, 2, 3\ldots$).

With (11) taken into account, the solution to equation (3) is given by

$$N(z, t) = N_0\left[1 + 2\sum_{\kappa=1}^{\infty} \bar{N}_\kappa(t) \cos\left(2\pi\kappa \frac{z}{\Lambda}\right)\right]. \qquad (13)$$

For the numerical solution, it is convenient to write system (12) in the form

$$\frac{d\bar{N}_\kappa}{d\tau} = -\kappa^2\bar{N}_\kappa + \kappa G_0(\bar{N}_{\kappa-1} - \bar{N}_{\kappa+1}), \quad (14)$$

where $G_0 = F_0\Lambda/(4\pi k_B T)$; $\tau = t/t_1$; and $\kappa = 1, 2, 3\ldots$. Note that at $\Omega \ll 1$ the amplitude $F_0 \sim 1/\Lambda$, then the coefficient G_0 is independent of Λ.

In the steady state (at $\tau \gg 1$), expression (14) yields a system of algebraic equations

$$\bar{N}_\kappa = \frac{G_0}{\kappa}(\bar{N}_{\kappa-1} - \bar{N}_{\kappa+1}). \quad (15)$$

Obviously, in the steady state with increasing κ, the amplitudes of the harmonics decrease with a simultaneous weakening of their relationship with neighbouring amplitudes. At $G_0 < 1$, we can obtain from (13) and (15)

$$N(z) \approx N_0\left[1 + 2\sum_{\kappa=1}^{\infty}\frac{G_0^\kappa}{\kappa!}\cos\left(2\pi\kappa\frac{z}{\Lambda}\right)\right]. \quad (16)$$

Figure 3 shows the results of the numerical solution to system (14) for the maximum concentration response

$$\Delta\bar{N}(\tau) = 2\sum_{\kappa=1}^{\infty}\bar{N}_\kappa(\tau)$$

[in intensity antinodes, at $\cos(2\pi\kappa z/\Lambda) = 1$] as a function of the coefficient G_0 at $\Lambda(\Theta = \pi/2) = \pi/\kappa$. From the numerical solution (see Fig. 3) using the exponential approximation

$$\Delta\bar{N}(\tau) = \Delta\bar{N}_0[1 - \exp(-t/t_f)] \quad (17)$$

we obtain the dependence of the time t_f on the coefficient $G_0 \sim I_0$ (Fig. 4).

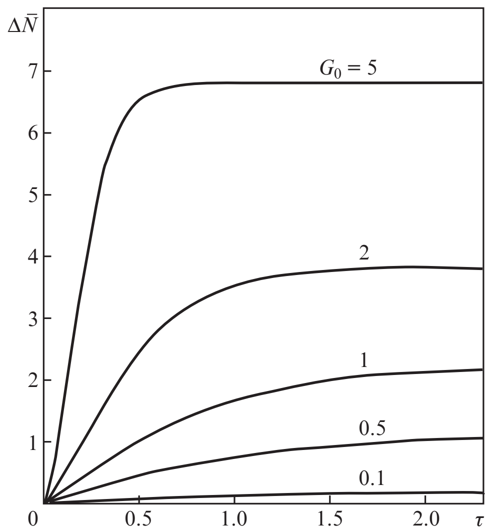

Figure 3. Time dependences of the concentration response at $\Lambda = \pi/\kappa$ and different values of G_0.

One can see from Fig. 4 that with increasing intensity I_0 the time t_f decreases, and accordingly with decreasing I_0 the time $t_f \to t_1$. A decrease in t_f with increasing I_0 was observed in the experimental study of FWM in an aqueous suspension of latex microspheres [3].

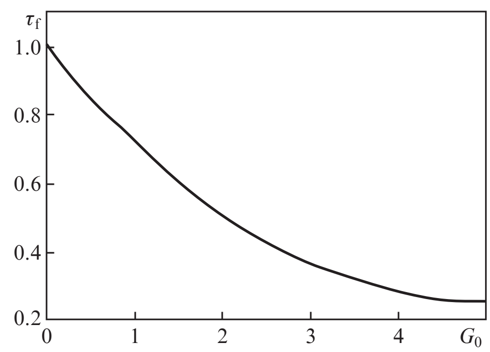

Figure 4. Dependence of the normalised time $\tau_f = t_f/t_1$ on the coefficient G_0.

2.4. Diffusion limit

In the diffusion limit $G_0 \ll 1$ [2, 4], when it is possible to take into account only one harmonic $\bar{N}_1(t)$, from (14) we find

$$\bar{N}_1(t) = G_0[1 - \exp(-t/t_0)], \quad (18)$$

where $t_0 = t_f = t_1$.

In this case (13) takes the form

$$N(z,t) = N_0\left\{1 + 2G_0[1 - \exp(-t/t_0)]\cos\left(2\pi\frac{z}{\Lambda}\right)\right\}. \quad (19)$$

Estimates show that in an aqueous suspension of latex microspheres with $R \approx 10^{-5}$ cm, formula (19) is valid at $I_0 < 10^2$ W cm^{-2}. When the radiation is turned off, the process of relaxation of the main grating is described by the expression

$$\bar{N}_1(t) = 2G_0[1 - \exp(-\tau_p/t_0)]\exp[-(t - \tau_p)/t_0] \text{ at } t \geqslant t_p. \quad (20)$$

3. Raman–Nath diffraction on the main concentration grating

Taking into account (19) and the expression for the polarisation of a weak diffracting wave $E_c\exp[i(\boldsymbol{k}_c\boldsymbol{r} - \omega t)]$

$$P_c = [\varepsilon_0 + \alpha N(z,t)]E_c[-i(\omega t - \boldsymbol{k}_c\boldsymbol{r})] \quad (21)$$

for the amplitude E_c we can obtain an equation

$$\cos\Theta_c\frac{\partial E_c}{\partial x} = i\frac{\omega}{c}n_2^0[1 - \exp(-t/t_0)]I_0\cos\left(2\pi\frac{z}{\Lambda}\right)E_c, \quad (22)$$

where ε_0 is the dielectric constant of the liquid; Θ_c is the angle between the vector $\boldsymbol{k}_c$ and the normal to the layer of the suspension;

$$n_2^0 = (2\pi\alpha)^2 N_0\frac{U(\Omega)}{ck_B T} \quad (23)$$

is the optical Kerr coefficient, which in the general case depends not only on the suspension parameters (α, N_0, T), but also on the modulation period Λ of laser radiation. Note that at $U(\Omega) \approx 1$, regardless of the sign of α, the coefficient is $n_2^0 > 0$ and determined only by the parameters of the suspension, similarly to media with a cubic nonlinearity. For latex microspheres in water at room temperature and argon laser radiation with $\lambda_0 = 5145$ Å under the experimental conditions [3] ($N_0 = 6.5 \times 10^{10}$ cm^{-3}, $n_0 = 1.59$, $n = 1.33$, $R = 1.17 \times 10^{-5}$ cm, $k_\mathrm{B}T = 4.05 \times 10^{-14}$ erg) of (23), from (23) at $U(\Omega) \approx 1$ we find $n_2^0 = 8.5 \times 10^{10}$ cm^2 W^{-1}, which is 2.8×10^4 times greater than n_2^0 in carbon disulfide ($n_2^0(\mathrm{CS_2}) = 3 \times 10^{-14}$ cm^2 W^{-1} [5]). The experimentally measured value of n_2^0 in the study of the concentration FWM was 3.6×10^{-9} cm^2 W^{-1} [3]. This difference in the values of the Kerr coefficient n_2^0 is probably associated with the use of the plane wave approximation in this paper. For the above parameters at $\Lambda = \pi/k$ and $\eta = 10^{-2}$ P, $t_1 = 0.5 \times 10^{-3}$ s.

From the solution to equation (22)

$$E_\mathrm{c}(L) = E_\mathrm{c}(0)\exp[\mathrm{i}\delta(t)\cos(2\pi z/L)] \tag{24}$$

using a known relation

$$\exp[\mathrm{i}\delta\sin(2\pi z/L)] = \sum_{m=-\infty}^{\infty} J_m(\delta)\exp(\mathrm{i}m2\pi z/L)$$

for the diffraction efficiency of the mth order we find (see, e.g., [12])

$$\eta_m = J_m^2(\delta), \tag{25}$$

where $\delta(t) = (n_2^0\omega/c)[1 - \exp(-t/t_0)]I_0(t)L/\cos\Theta_\mathrm{c}$; and L is the thickness of the suspension layer.

In this case, the direction in the diffraction maxima will be determined by the grating equation [5]

$$\cos\Theta_m = m\frac{\lambda}{\Lambda} + \cos\Theta_\mathrm{c}, \tag{26}$$

where $m = 0, \pm1, \pm2\ldots$. Note that from the identity

$$\sum_{m=0}^{\infty} J_m^2 = 1$$

follows the law of conservation of energy for the diffracted wave. The diffraction efficiency η_m can be used to experimentally measure the time t_0 of formation and decay of the concentration grating as a function of the period t_0 and optical Kerr coefficient n_2^0. In the case of high intensities I_0 (at $G_0 \geqslant 1$) one can also measure the dependence of the nonlinearity settling time on the modulation period Λ and the intensity of the incident radiation.

4. Conclusions

We have developed the theory of concentration nonlinearity of a suspension of transparent microspheres under the action of a gradient force F_∇ in a periodically modulated laser field. We have studied the gradient force, taking into account the nonuniformity of radiation in the volume of a microsphere. The conditions have been determined for observing the zero-force effect, when, regardless of the microsphere position, the force acting on the microsphere is equal to zero: $F_\nabla = 0$.

Based on a one-dimensional Smoluchowski equation represented as a superposition of harmonics of the concentration of microspheres with multiple periods, we have analysed the kinetics of concentration nonlinearity of a suspension. The resulting system of steady-state recurrence equations has been solved numerically. As a result, we have determined the dependence of the concentration nonlinearity formation time t_f on the intensity of the incident radiation. A decrease in t_f with increasing intensity is associated with an increase in velocity $v \sim F_\nabla$ of microspheres ($t_\mathrm{f} \sim \Lambda/v$). It is shown that in the diffusion limit the time t_f coincides with the diffusion decay time of concentration nonlinearity, $t_\mathrm{f} = t_0$. In this approximation, we have obtained an expression for the optical Kerr coefficient $n_2^0 \sim \alpha^2 N_0 U(\Omega)/(k_\mathrm{B}T)$, from which it follows that at $\Omega = 2\pi R/\Lambda \ll 1$, irrespective of the sign of the polarisability α, $n_2^0 > 0$. It is shown that for the suspension parameters used for the study of FWM [3], $n_2^0 = 8.5 \times 10^{-10}$ cm^2 W^{-1}, which is 3×10^{-4} times greater than n_2^0 in carbon disulfide. Thus, the suspension of transparent microspheres – an artificial heterogeneous medium, whose each component alone does not exhibit nonlinear optical properties, may serve as a promising broadband nonlinear material for low-intensity laser radiation of long pulse duration.

To study the kinetics of concentration nonlinearity we present expressions for the diffraction efficiencies of arbitrary orders of a weak wave on a concentration grating of microspheres induced by periodically modulated laser radiation.

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