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Optical method for formation of crystals in nanosuspension

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Abstract. It is proposed to use light pressure forces to form crystals from nanoparticles in a transparent nanosuspension. The paper discussed theoretical model of formation of crystal from nanoparticles on a bottom of cuvette by using the laser effect in liquid. The solution of one-dimensional task of the light induced mass transfer was received as depending on intensity of laser beam.

1. Introduction

Chemical methods for the formation of nanostructures (including photonic crystals) from the liquid phase are widely known. However, recent studies show a number of new opportunities. For example, Duhr and Braun show how the light-induced thermal diffusion of latex microparticles in a fluid leads to the formation of a two-dimensional crystal on a cooling surface in a convective fluid flow [1]. The concentration of microparticles increased in the zone of the light beam by several orders of magnitude. For nanoparticles, the realization of such method is difficult, since the coefficient of thermal diffusion is much less for them than for microparticles [2].

In this paper, it is proposed to use light pressure forces to form crystals from nanoparticles in a transparent nanosuspension.

2. Light-induced formation of crystals

Consider the liquid phase medium with the nanoparticles (dispersed phase) which is under the influence of the reference laser beam with a uniform intensity profile I (figure 1).

Light pressure force acting on the nanoparticle from the high-power reference beam is equal to [3]:

$$F_{p} = \frac{128\pi^{5}a^{6}n_{1}}{3c_{0}\lambda^{4}} \left[\frac{m^{2}-1}{m^{2}+2}\right]I, \qquad (1)$$

where $m = n_2 / n_1$, n_1 , n_2 are the refractive substance indices of the dispersion medium and the dispersed phase respectively, c_0 is the velocity of light, *a* is the particle radius, λ is the light wavelength.

Balanced one-dimensional equation describing the dynamics of the concentration of the nanoparticles in a liquid phase medium with diffusion [3]:

$$\frac{\partial C}{\partial t} = D\nabla^2 C - V\nabla C \,. \tag{2}$$

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Figure 1. The light-induced sedimentation scheme: 1 - cell with liquid, I - laser beam

here C(z,t) is volume concentration of particulate matter, axis z is aligned with the reference beam I, D is diffusion coefficient; particle velocity $V = \left(64\pi^4 a^5 n_1 \left(m^2 - 1\right) \left(m^2 + 2\right)^{-1} \left(9c_0 \lambda^4 \eta\right)^{-1}\right) I$, where η is viscosity of the fluid

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The relevant boundary conditions:

$$-D\nabla C + VC = 0, \text{ when } z = 0 \text{ and } z = l, \qquad (3)$$

where l - the height of the cell along the propagation of the reference beam. Initial conditions:

$$C = C_0, \text{ when } t = 0, \tag{4}$$

where C_0 - the initial concentration of nanoparticles.

The exact solution of the equations (2) - (4):

$$C(z,t) = C_0 \left\{ 1 - u e^{0.5uz} \sum_{m=1}^{\infty} \frac{2(\pi m)^2 \left[1 - (-1)^m e^{-0.5u} \right]}{\left[(\pi m)^2 + u^2 / 4 \right]^2} y_m \left[1 - e^{-((\pi m)^2 + u^2 / 4) \frac{D}{L^2} t} \right] \right\},$$
(5)

where $y_m = (u/2\pi n)\sin(\pi nz) + \cos(\pi nz)$, u = VL/D.

In Figures 2 and 3 the solutions for u=3 and various intervals of times are shown. It is seen that the equilibrium actually occurs at t=1.

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Figure 2. Function $C(z,t)/C_0$, if u=3 and $t \in [0;0.1]$



Figure 3. Function $C(z,t)/C_0$, if u=3 and $t \in [0;10]$

For the steady-state the expression (5) takes the next form:

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$$C(z,I) = C_0 \gamma I D^{-1} l \frac{e^{\gamma I D^{-1} z}}{e^{\gamma I D^{-1} l} - 1},$$
(6)

where $\gamma = 64\pi^4 a^5 n_1 (m^2 - 1) (m^2 + 2)^{-1} (9c_0 \lambda^4 \eta)^{-1}$.

The expression (6) we can write as:

$$C(z, l_{ph}) = C_0 l / l_{ph} \frac{e^{z / l_{ph}}}{e^{l / l_{ph}} - 1},$$
(7)

where we introduce parameter $l_{ph} = D/\gamma I$, that shows the depth at which the particle concentration changes by a factor of e at a given intensity.

The figure 4 illustrates the dependence of the parameter l_{ph} on the radiation intensity for the following values: a=10⁻⁷ m, n₁=1.1, n₂=1.33, λ =632 nm, η =1.004 mPa·s.



Figure 4. Dependence of the parameter l_{ph} on the radiation intensity.

Also the expression (6) can be written as:

$$C_{r.u.}(z', I_{r.u.}) = I_{r.u.} \frac{\exp(I_{r.u.} z')}{\exp(I_{r.u.}) - 1},$$
(8)

where $C_{r.u.}(z', I_{r.u.}) = C(z', I) / C_0$ – relative concentration, $I_{r.u.} = I / I_{sat}$ – relative intensity of radiation, $I_{sat} = D / \gamma l$ – saturation intensity, z' = z/l.

3. Conclusions

Expression (8) gives a direct relationship between the change in the relative concentration and the relative intensity of radiation at the bottom of the cell. As estimates show, an essential increase in the relative concentration requires an intensity of about some MW/m^2 , which is achievable for transparent nanomaterials using continuous laser sources. These light intensities form the conditions for the growth of the crystal structure.

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Thus, an optical method for the formation of crystals from nanosuspension is proposed. A distinctive feature of the method is the use of light pressure forces, with the help of which it is possible to carry out effective sedimentation of nanoparticles in a transparent medium [4-6].

The proposed method for the formation of a colloidal crystal from dilute nanosuspension is also of interest for the formation of photonic crystals and chemical sensors [7-10]. The methods for efficient accumulation of nanoparticles from solution are of great interest for increasing the sensitivity of diffusion-limited surface biosensors [1]. The results can also be useful for the development of methods for optical diagnostics of nanomaterials [11-14].

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