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## **Influence of Nanoparticle Interactions on Their Spatial Distribution in Liquid under a Uniform Light Field**

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*The article investigates the sedimentation process of interacting nanoparticles in a liquid under the action of light pressure forces. A model is proposed that allows determining the dependence of nanoparticle concentration on the coordinate along which a constant external influence acts, taking into account the interaction forces (attraction and repulsion) between nanoparticles. The study showed that this dependence can significantly differ from the result predicted by the classical model of an ideal gas of nanoparticles. The obtained conclusion is important for developing methods to control the arrangement of nanoparticles in liquids, which is relevant for creating optical techniques for forming thin films and structures.*

**Keywords:** nanosuspension, light pressure, nanoparticle deposition, inhomogeneous concentration, particle interactions.

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### **Introduction**

The problem of forming non-uniform distributions of nanoparticles in liquid suspensions under the action of external fields, including light pressure, is a major challenge for applications in biophotonics, environmental monitoring, and deposition nanotechnology. The range of practical tasks is extremely broad: from targeted drug delivery using magnetic nanoparticles [1] and biofilm formation [2] to the purification of aqueous solutions from polyethylene microparticles [3]. Methods for controlling particle separation, including the use of optical lasers, were reviewed in [4]. Currently, experimental and theoretical studies on the effect of lasers on nanosuspensions are being actively conducted [5–7]. Most authors focus on the horizontal dynamics of particles in laser beams or thermodiffusion [8, 9], while the vertical geometry accounting for gravity has been

insufficiently studied. At the same time, the task of investigating the behavior of nanosuspensions under external influence in a vertically oriented cuvette is quite relevant. In particular, paper [10] considers the possibilities of forming various structures on a substrate, which is the bottom of the cuvette containing the nanosuspension.

Papers [11, 12] analyzed the optical deposition of nanoparticles within the ideal gas model, where the sizes of the nanoparticles and their interactions with each other were neglected. However, it is obvious that the interaction of nanoparticles cannot be neglected at their sufficiently high concentrations. Paper [14] considered the stationary distribution of nanosuspension particles with finite volume in a uniform force field, which corresponds to repulsive forces between nanoparticles.

This paper considers not only the repulsion between suspension nanoparticles, which are

acted upon by an external force (light pressure force), but also their mutual attraction.

As is known (see, for example, [15]), in the equation of state for real dilute gas state, the interaction of gas molecules in the second virial coefficient is described by two parameters  $a$  and  $b$ , where  $b$  “accounts for” the repulsion of molecules, and  $a$  – for their attraction. The parameter  $b$  is of the order of the molecular volume, while the parameter  $a$  is expressed through the integral of the potential energy of pairwise molecular interaction over the distance between molecules with a weight equal to the square of that distance. When considering a real gas with an arbitrary density, these quantities become fitting parameters in an interpolation equation (see, for example, [15, 16]).

A similar approach is applied here (as in [14]) to interacting nanoparticles that are in a liquid in equilibrium.

Indeed, nanoparticles located in a macroscopic volume  $V$ , which is small compared to the cuvette volume, can be considered as a quasi-closed and statistically independent subsystem with a variable number of particles. The probability of finding the subsystem in a certain state with an exact number of particles  $N$  is determined by the grand canonical distribution [15], which contains the potential energy of the interacting nanoparticles. If this energy can be represented as pairwise, triple, etc., interactions between particles, then the thermal equation of state  $P = P(V, T, \bar{N})$  for a sufficiently dilute “gas” of nanoparticles can be written in a virial form, just as for a conventional real gas. In this equation,  $T$  is the temperature (we will use energy units), and  $\bar{N}$  is the average number of particles. When transitioning to an arbitrary nanoparticle density, the equation of state containing virial coefficients can be replaced by one of the interpolation equations [15, 16] by analogy with a real gas. The most well-known of these equations are the van der Waals equation, the first and second Dieterici equations, and the Berthelot equation [16].

Here we will use the van der Waals equation:

$$\left( P + a \left( \frac{\bar{N}}{V} \right)^2 \right) (V - b\bar{N}) = \bar{N}T, \quad (1)$$

which allows us to determine the relationship between the nanoparticle concentration  $n$  and the coordinate  $z$ , along which a constant and uniform force  $F$  acts on the nanoparticles.

### Problem Formulation and Main Results

Let  $N$  nanoparticles be placed in a cuvette containing a liquid. Its volume is equal to  $Sh$  ( $S$  is the base area,  $h$  is the height). Initially, the nanofluid is homogeneous, with nanoparticle concentration equal to  $\tilde{n} = N / Sh$ . At some point in time, constant external force  $\mathbf{F} = (0, 0, -F)$  begins to act on each nanoparticle. This force is directed antiparallel to the  $\bar{z}$  axis of the Cartesian coordinate system, with plane  $\bar{z} = 0$  corresponding to the cuvette base. Following the activation of the external force, the nanoparticle concentration begins to change, and thermodynamic equilibrium is established after a sufficiently long (theoretically infinite) time interval. Moreover, for a nanofluid located in a thin layer  $Sd\bar{z}$ , where it can be considered homogeneous, the equation of state (1) holds if we replace  $V$  with  $Sd\bar{z}$  and  $\bar{N}$  with  $dN$ . Then, denoting  $n = dN / Sd\bar{z}$  as the nanoparticle concentration in the cross-section of the nanofluid at coordinate  $\bar{z}$ , we rewrite (1) as

$$P = \frac{nT}{(1 - bn)} - an^2.$$

Considering that  $T = \text{const}$ , we obtain

$$dP = T \frac{dn}{(1 - bn)^2} - 2andn. \quad (2)$$

Let us introduce the following notations  $z = \bar{z} / h$ ,  $x(z) = bn(z)$ ,  $x_0 = x(0)$ ,  $x_1 = x(1)$ ,  $\tilde{x} = b\tilde{n}$ . Hereinafter,  $\tilde{n}$  is the average concentration of nanoparticles in the cuvette.

After substituting (2) into the expression

$$dP = -nFhdz,$$

we obtain

$$\frac{dx}{x(1-x)^2} - \frac{2a}{bT} dx = -\frac{Fh}{T} dz.$$

Integrating this expression from arbitrary value  $x$  to  $x_1$  on the left-hand side and from an

arbitrary  $z$  to unity on the right-hand side, after straightforward transformations, we arrive at equation:

$$z = 1 + \frac{1}{q} \left\{ \frac{1}{1-x_1} - \frac{1}{1-x} + \ln \left| \frac{1-x}{1-x_1} \right| + \ln \frac{x_1}{x} + \frac{2a}{Tb} (x-x_1) \right\}, \quad (3)$$

where  $q = Fh/T$  is dimensionless parameter.

If we set both  $a$  and  $b$  equal to zero in (3), we obtain the Boltzmann formula (for an ideal gas concentration), which can be written as:

$$\frac{1}{q} \ln \left( \frac{n_0}{n} \right) = z. \quad (4)$$

After substituting  $z = 0$  into (3), we obtain the relations between  $x_0$  and  $x_1$ :

$$1 = \frac{1}{q} \left\{ \frac{1}{1-x_0} - \frac{1}{1-x_1} + \ln \left| \frac{1-x_1}{1-x_0} \right| + \ln \frac{x_0}{x_1} - \frac{2a}{Tb} (x_0-x_1) \right\}. \quad (5)$$

To determine the concentrations of particles at  $z = 0$  and  $z = 1$  in terms of  $a$ ,  $b$  and  $q$ , we use the relation

$$Shd(zn(z)) = Shn(z)dz + Shzdn.$$

Integrating this expression over  $z$  from 0 to 1, and over  $n$  from  $n_0 = n(0)$  to  $n_1 = n(1)$ , and considering that  $N = Sh \int_0^1 n(z) dz$ , we obtain

$$\bar{x} = x_1 - \int_{x_0}^{x_1} z dx. \quad (6)$$

Substituting expression (3) into (6) and evaluating the integrals over  $x$ , we obtain the equality.

$$\bar{x} = x_1 - \frac{1}{q} \left[ \frac{x_1-x_0}{1-x_0} + x_1 \ln \frac{x_0(1-x_1)}{x_1(1-x_0)} + \frac{a}{Tb} (x_0-x_1)^2 \right]$$

or (after cancelling  $b$ )

$$\bar{n} = n_1 - \frac{1}{q} \left[ \frac{n_1-n_0}{1-bn_0} + n_1 \ln \frac{n_0(1-bn_1)}{n_1(1-bn_0)} + \frac{a}{T} (n_0-n_1)^2 \right]. \quad (7)$$

From (7) and (5), at  $a = 0$ , it is easy to express  $n_0$  and  $n_1$  in terms of  $\bar{n}$ , as was done in [14]:

$$n_0 = \frac{q\bar{n}}{1+bq\bar{n} - \exp[-q(1-b\bar{n})]} \quad (8)$$

and

$$n_1 = \frac{q\bar{n} \exp[-q(1-b\bar{n})]}{1+(bq\bar{n}-1)\exp[-q(1-b\bar{n})]}. \quad (9)$$

However, obtaining analytical expressions for  $a \neq 0$  analogous to (8) and (9) appears to be impossible. Therefore, in this work, we will investigate the effect of nanoparticle attraction, i.e., the parameter  $a$ , on their equilibrium distribution graphically, using expression (3). We assume that at  $z = 1$ , the attraction between nanoparticles can be neglected, and consequently, the value of  $n_1$  is determined by formula (9).

## Analysis of the Results

For a graphical study of expression (3) at a finite value of parameter  $a$ , we introduce the following dimensionless quantities:  $y = n/\bar{n}$ ,  $y_1 = n_1/\bar{n}$ ,  $y_0 = n_0/\bar{n}$ ,  $g = \bar{n}b$ ,  $k = 2a\bar{n}/T$ .

Then (3) can be written in the following form:

$$z = 1 + \frac{1}{q} \left\{ \frac{1}{1-gy_1} - \frac{1}{1-gy} + \ln \left| \frac{1-gy}{1-gy_1} \right| + \ln \frac{y_1}{y} + k(y-y_1) \right\}, \quad (10)$$

that is, the concentration dependence on depth will be determined by three parameters:  $q$ ,  $g$ ,  $k$ .

In the new notation, relation (9) takes the form:

$$y_1 = \frac{q \exp[-q(1-g)]}{1+(gq-1)\exp[-q(1-g)]}. \quad (11)$$

It was shown in [14] that the dependence  $z = z(y)$  (or  $y = y(z)$ ) differs significantly from the

case when  $b = 0$  for  $q = 10^7$  and  $g = 10^{-5}$ ; therefore, it is reasonable to consider the effect of attraction between nanoparticles for the same values of  $q$  and  $g$ .

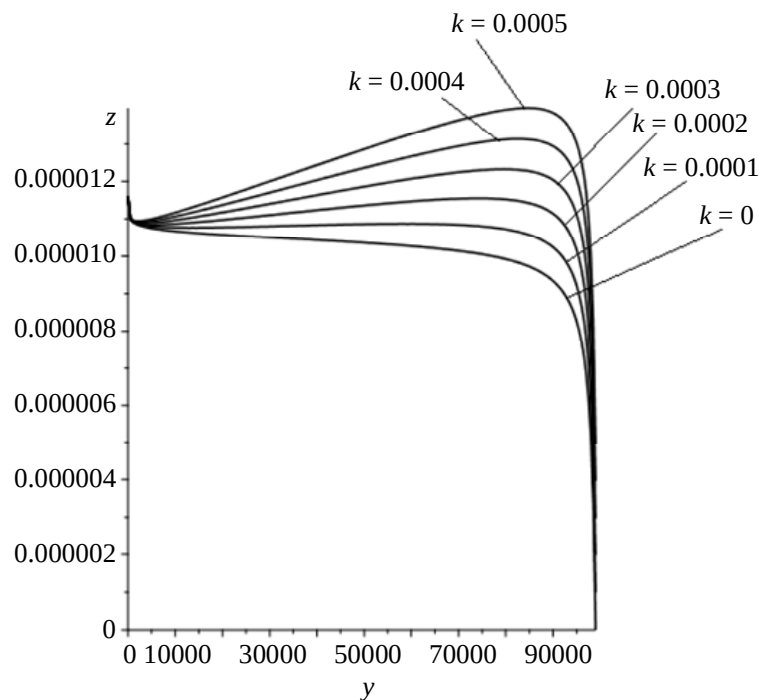
Figure 1 shows dependencies  $z = z(y)$  at  $y \in (1, y_0)$ , when  $q = 10^7$ ,  $g = 10^{-5}$ , while parameter  $k$  takes the values 0; 0.0001; 0.0002; 0.0003; 0.0004; 0.0005.

The curve corresponding to  $k = 0$  coincides with the curve obtained in [14].

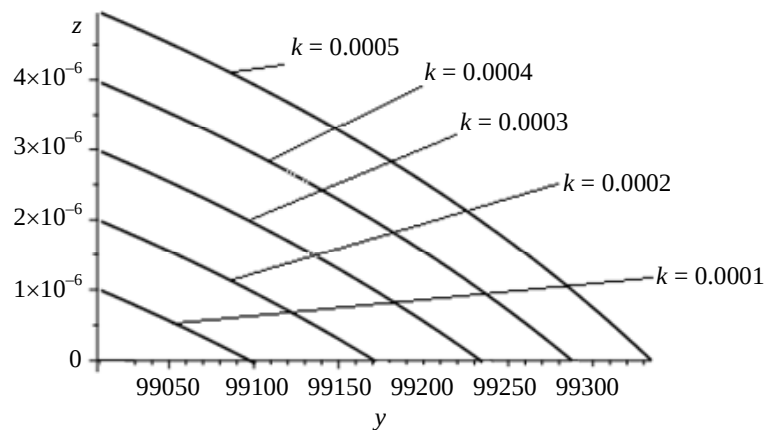
From the obtained curves, it can be seen that a relative concentration of particles with an extremely large gradient is created in a very thin

layer near the cuvette bottom. With  $a$  increasing, the downward migration of particles intensifies, and the curves reach a greater maximum at a longer distance from the cuvette bottom.

Figure 2 shows the same dependencies  $z = z(y)$ , as in Figure 1, but at  $y \in [y_0, 99334, 51654]$ . Analysis of the results shows that, with  $a$  increasing, the concentration of nanoparticles at the cuvette bottom increases (the authors do not show the functions  $z = z(y)$  for negative  $z$ , which have no physical meaning in the chosen formulation of the problem).

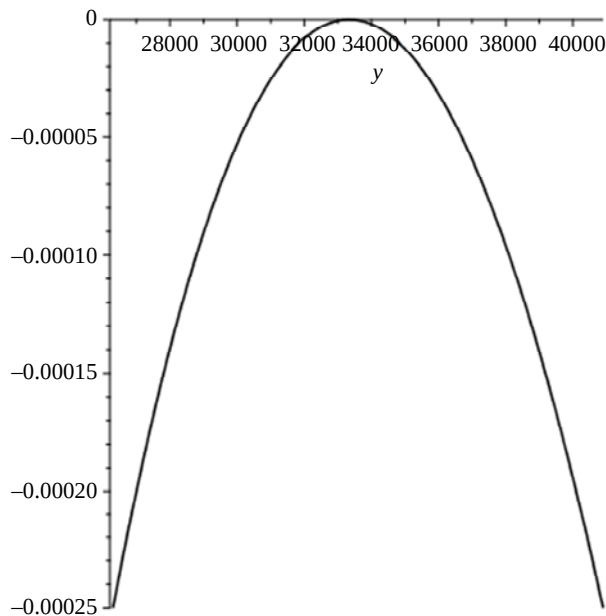


**Fig. 1.** Functions  $z = z(y)$  at  $g = 10^{-5}$  and  $q = 10^7$ ,  $y \in (1, y_0)$

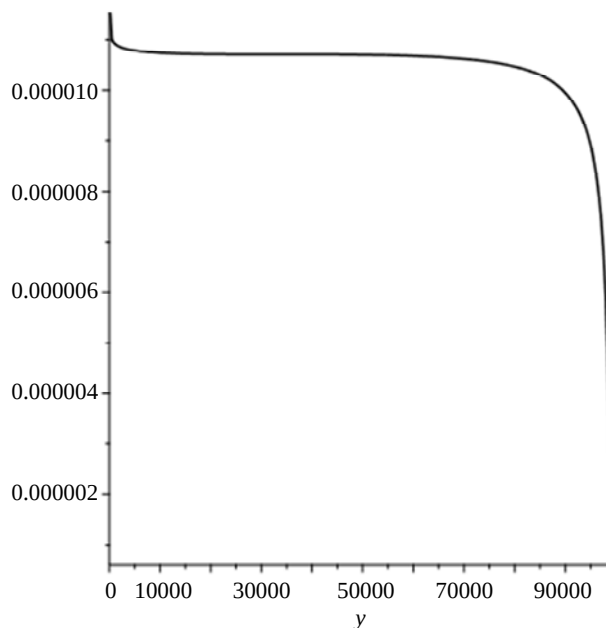


**Fig. 2.** Functions  $z = z(y)$  at  $g = 10^{-5}$  and  $q = 10^7$ ,  $y \in (y_0, 99334, 51654)$

Figure 3 shows the derivative  $dz/dy = u(y)$  (multiplied by  $10^9$ ) at  $k = 0.0000675$ . The equation  $u(y) = 0$  has two multiple real roots (in the region of non-negative  $z$ ) equal to 33333.3333. Consequently, for such a value of  $k$ , the function  $z = z(y)$  has an inflection point (Fig. 4).



**Fig. 3.** Functions  $u = u(y)$  at  $k = 0.0000675$ ,  $g = 10^{-5}$  and  $q = 10^7$

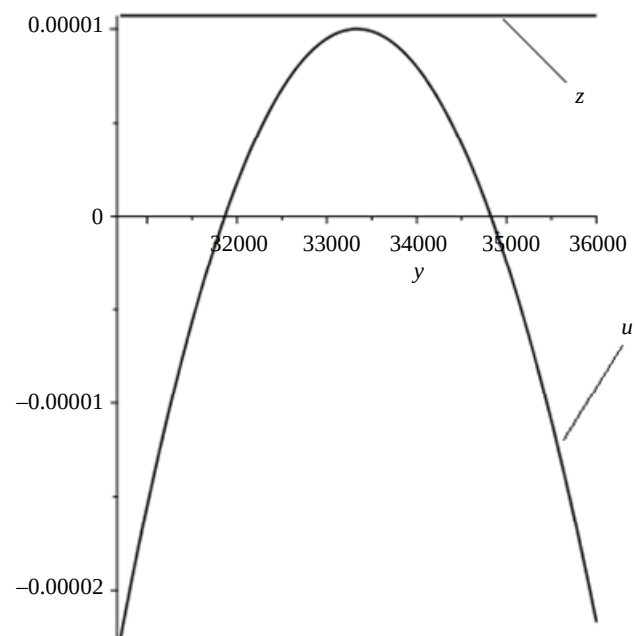


**Fig. 4.** Functions  $z = z(y)$  at  $g = 10^{-5}$  and  $q = 10^7$ ,  $k = 0.0000675$ ,  $y \in (1, y_0)$

If, for the chosen values of  $q$  and  $g$ , the parameter  $k$  exceeds 0.0000675, then the equation  $u = 0$  will have two distinct roots, with the

smaller root corresponding to the minimum of the function  $z(y)$ , and the larger root to its maximum. Figure 5 shows the functions  $z$  and  $u$  (for clarity, the latter is multiplied by  $10^9$ ) for  $k = 0.0000676$ . In this figure, the function  $z = z(y)$  is shown as a horizontal segment; in reality, this is an illusion. It is simply that  $z(y_{\max}) - z(y_{\min}) \ll 0.00001$ .

Thus, we find that for sufficiently large  $q$ , and starting from certain values of  $k$ , the function  $y = y(z)$  (the inverse of  $z = z(y)$ ) becomes multivalued over some intervals of the coordinate  $z$ . Physically, this may correspond to different “phases” of the nanosuspension, by analogy with the van der Waals gas.



**Fig. 5.** Functions  $z = z(y)$  and  $u = u(y)$  at  $g = 10^{-5}$  and  $q = 10^7$ ,  $k = 0.0000675$ ,  $y \in (26302.32..40899.1)$

## Conclusion

The problem of nanoparticle deposition in a liquid under external impact is of significant scientific interest due to a wide range of practical applications. The results obtained show that taking into account nanoparticle interactions fundamentally changes the pattern of their distribution in the liquid. Based on the performed studies, it can be expected that in a nonuniform suspension of interacting nanoparticles subjected to intense laser radiation, spatial regions may exist in which different phases of the suspension coexist, similar to the different phases of the van der Waals gas.

Moreover, it offers a possibility of the targeted formation of layers with increased particle concentration under laser irradiation. The obtained expressions (3) and (10) show that it is possible to control nanoparticles in a liquid to form thin-film structures, which is of interest for the development of optical methods for particle manipulation, as well as for methods of nonlinear optics and diagnostics of liquid-phase media.

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