Mechanism of the ordered particles arrangement in a concentration grating excited in the field of counter-propagating Gaussian beams

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In the two-particle approximation, we consider the mechanism for the formation of an ordered arrangement of transparent spherical particles of small size in a concentration grating excited by the gradient force of counter-propagating Gaussian laser beams. This mechanism is due to the joint action of the transverse gradient force and the Coulomb force arising as a result of dipole-dipole interactions between particles.

It is known that a liquid suspension of transparent particles of small size is a highly efficient nonlinear optical heterogeneous medium for continuous laser radiation. For example, as reported in Ref. [1], the optical Kerr coefficient \( n_2 \) of the water suspension of latex spherical particles with radius \( R = 0.117 \) \( \mu m \) and concentration \( N = 6.5 \cdot 10^{10} \text{ cm}^{-3} \) turned out to be \( 10^5 \) times higher than in \( \text{CS}_2 \). For the first time, the possibility of using such heterogeneous media as a nonlinear optical material was noted in Ref. [5]. Nonlinear optical phenomena in such artificially created media, in particular, four-wave mixing [1,3] and stimulated concentration scattering [6,7], are caused by energy exchange processes between the waves interacting on the concentration gratings excited by them due to the gradient component of the light pressure force.

In Refs. [8,10], the concentration gratings were recorded in an aqueous suspension of polystyrene particles with \( R = 0.7 - 3 \) \( \mu m \) with the interference field of continuous lasers. An interesting experiment on the excitation of a two-dimensional concentration grating (matrix) of polystyrene particles with \( R = 150 - 300 \) nm in water under the influence of orthogonal pairs of interfering beams of continuous laser radiation was performed in Ref. [11]. The results obtained in [11] open up the prospects of synthesizing crystal matrices that demonstrate all the characteristics of ordinary molecular crystals and are of interest for various scientific and practical applications.

In the experiments on excitation of concentration gratings by coherent beams of a He-Ne laser, an ordered arrangement of particles along its lines was observed [9,10]. Particles “collected” under the action of the longitudinal component of the gradient force \( \mathbf{F}_z \) in the maxima of the interference pattern of the field (lines of the concentration grating) were located at approximately equal distances from each other (see Fig. 1). For Gaussian beams used in these experiments, the radiation intensity along the concentration grating lines is inhomogeneous, so that the transverse component of the gradient force \( \mathbf{F}_y \) appears leading to translation of the particles in the grating. In addition, the Coulomb forces of the dipole-dipole interaction \( \mathbf{F}_{dip} \) occur due to the induced dipole moment between the particles. These forces, together with the force \( \mathbf{F}_z \), move the particles to certain stable positions in the concentration grating.

In this report, using the two-particle dipole-dipole interaction as an example, the joint action of the \( \mathbf{F}_z \) and \( \mathbf{F}_{dip} \) forces is considered resulting in the stable arrangement of the particles in the concentration grating with a certain distance between them. Analysis of the spatial distribution of particles under the action of these forces in the general case is a very complex many-body problem. Nevertheless, even consideration of two particles gives an idea about the mechanism of their ordered arrangement in the concentration grating and allows one to draw certain conclusions and numerical estimates. Although we focus on the Gaussian beams in our consideration, one can find in the literature the other realizations of optical forces using, e.g., the Airy beams [12,13].

Let us consider a mechanism leading to an ordered arrangement of transparent spherical particles of small sizes in the concentration grating excited by linearly polarized Gaussian beams oppositely propagating along the \( z \) axis. In this case, the total amplitude of the field can be represented as

\[
E = E_0 e^{-r^2/2r_0^2} \cos k z \cdot e^{-i\omega t} + c.c.,
\]

where \( r_0 \) is the beam radius, \( r = \sqrt{x^2 + y^2} \) is the radial coordinate. In the field \([1]\), the particle is affected by the gradient force given by \([2]\)

\[
\mathbf{F}_z = \frac{n^2}{2} \alpha \nabla (E^2),
\]

\[
\mathbf{F}_{dip} = \frac{e^2}{4\pi \varepsilon_0} \int d^3 r' \frac{\mathbf{r} - \mathbf{r}'}{r'^3} \mathbf{E} \cdot \mathbf{E}',
\]

\[
\mathbf{F}_y = \frac{e^2}{4\pi \varepsilon_0} \int d^3 r' \frac{\mathbf{r} - \mathbf{r}'}{r'^3} \mathbf{E} \times \mathbf{E}'.
\]
where

\[ \alpha = \frac{m^2 - 1}{m^2 + 2} R^3 = \alpha_0 R^3, \]  
\[ n = \text{the particle polarizability (} \kappa R \ll 1\), } m = n_0/n \text{ is the ratio of the refractive indices of the particle material } n_0 \text{ and the surrounding liquid } n, \]

\[ \langle E^2(z, r) \rangle_r = E_0^2 e^{-r^2/\rho_0^2} (1 + \cos 2kz). \]  

Here \( \langle \ldots \rangle \) stands for averaging over time. Further we also assume that the particles are small in comparison to the beam radius, \( d = 2R \ll r_0 \).

The longitudinal component of the gradient force,

\[ F_z = -n^2 \frac{\pi}{\Lambda} \alpha E_0^2 e^{-r^2/\rho_0^2} \sin 2\pi \frac{z}{\Lambda}, \]

forms the concentration grating with the period \( \Lambda = \pi/k \), whereas the transversal one,

\[ F_r = -n^2 \frac{r}{\rho_0^2} \alpha E_0^2 e^{-r^2/\rho_0^2} \left( 1 + \cos 2\pi \frac{z}{\Lambda} \right), \]

together with the Coulomb dipole-dipole force \( F_{dip} \) results in the ordered arrangement of particles in the grating (at \( z = m\Lambda, m = 0, \pm 1, \pm 2, \ldots \)).

The dipole moment of the particle induced by the field can be written as follows,

\[ p(r, t) = q(r) d \cdot \cos \omega t, \]

where \( q(r) = \frac{2\alpha E_0}{d} e^{-r^2/\rho_0^2} \) is the dipole charge.

To simplify calculations, we assume that the first particle is located at the beams center \( (r = 0) \), whereas the second particle is in the arbitrary point \( r \) (see Fig. 2). Since these coordinates correspond to the particles centers, then \( r \gg d \).

Using the geometrical scheme shown in Fig. 2 and the results of Ref. [14], one can obtain for our case the expressions for the Coulomb forces of repulsion \( (F_+ > 0) \) and attraction \( (F_- < 0) \),

\[ F_+ = \frac{8 \alpha E_0}{rd} e^{-r^2/\rho_0^2}, \]

\[ F_- = -\frac{F_+}{(1 + d^2/r^2)^{3/2}}. \]  

Thus, the resulting Coulomb force is

\[ F_{dip} = F_+ + F_- = 8 \left( \frac{\alpha E_0}{rd} \right)^2 e^{-r^2/\rho_0^2} \left[ 1 - \frac{1}{(1 + d^2/r^2)^{3/2}} \right] > 0. \]

From Eq. (6) at \( z = m\Lambda \) and Eq. (9), we have the transcendental equation for the equilibrium distance \( r_p \) between the particles,

\[ \frac{\alpha_d r_p^2}{2n^2 r_p^3} \left[ 1 - \frac{1}{(1 + d^2/r_p^2)^{3/2}} \right] e^{r_p^2/\rho_0^2} = 1. \]

Obviously, the distance \( r_p \) does not depend on the radiation intensity. It is determined by the polarizability and particle size as well as the radius of laser beams. In particular, for \( r_p \ll r_0 \) and \( (d/r_p)^2 \ll 1 \), we find from Eq. (10) that

\[ r_p \approx \sqrt[3]{6 \alpha_0 (r_0/n)^2}. \]

For example, for polystyrene particles with \( R = 100 \text{ nm} \) in water \( (n = 1.33, \alpha_0 = 0.12) \) and beams radius \( r_0 = 4 \cdot 10^{-2} \) mm, we obtain \( r_p \approx 0.9 \cdot 10^{-3} \) mm. At the same time, both inequalities determining the range of applicability of Eq. (11) are satisfied. With an increase in the particle radius by 2 times, the distance \( r_p \) increases by about 1.5 times.

In conclusion, we note that in the experiments [9, 10], the small-particle approximation \( (\kappa R \ll 1) \) used here is not fulfilled and, accordingly, the formalism of the Maxwell stress tensor [15] must be used to calculate the gradient force. This approach allows us to calculate the gradient force in the case of particles of arbitrary radius [16]. However, such calculations are very time consuming and require numerical simulations. For this brief report, which explains only the mechanism of the ordered arrangement of particles in the concentration gratings, these cumbersome calculations are not given.

Figure 2. Geometric scheme for calculating the Coulomb forces in the interaction of two dipoles induced by the linearly polarized field \( E_0 \).
here. We also note that when a concentration grating is excited by plane waves, the arrangement of the particles in its lines will be determined by the Coulomb forces of the dipole-dipole interaction and the transverse dimensions of the cell containing the suspension. The considered mechanism based on competition of the gradient force and dipole-dipole interaction should be taken into account upon creation of molecular-crystal-like structures [11] from liquid suspensions of small particles with the optical methods.

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