Self-focusing of the light in transparent nanosuspension

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Abstract. It was analyzed the self-focusing regime of the Gaussian beam in transparent nanosuspension with electrostrictive nonlinearity. The theoretical analysis of the light-induced mass transfer in the nanosuspension was carried out for large intensities of the Gaussian beam radiation, when the concentration change is comparable to the primary value. The nonlinear lens in this mode is the nonlinear function of the incident light intensity. It is shown that the critical power value decreases significantly for high intensity beam. The results are relevant to the study of the radiation self-action in the nanosuspension and optical diagnostics of such materials.

1. Introduction
The self-focusing (defocusing) of the light beam is well known nonlinear effect [1-3]. The light-induced thermal lens in a homogeneous fluid is formed as a result of thermal expansion of a medium. In two-component liquid the heat flow also can cause concentration stream arising from occurrence of thermodiffusion (Soret effect [2]). Another mechanism of optical nonlinearity of the medium is due to the forces operating on the particles of the dispersed phase in gradient light field [3-5]. Gradient electrostrictive forces may cause concentration streams in nanosuspension (artificial Kerr media) [5]. Depending on the sign of polarizability nanoparticles drift from areas with higher intensity of the electromagnetic wave (if their refractive index is more than one for liquid). This optical nonlinearity was studied experimentally in nanosuspensions and microemulsions [6-7]. The theoretical analysis is restricted usually by the case of weak intensities of radiation.

The aim of this work is to analyze the self-focusing regime in transparent nanosuspension at high intensities of radiation.

2. Electrostrictive mechanisms of cubic nonlinearity in nanosuspension
Usually in the case of non-resonant mechanism (for weakly absorbing media) is used the parameter of cubic nonlinearity - \( n_2^{\text{eff}} \left[ \frac{m^2}{W} \right] \), which characterizes the change of the refractive index \( n \) of the medium under the influence of incident radiation:

\[
n = n_0 + n_2^{\text{eff}} I,
\]

where \( n_0 \) - the refraction index of the medium in the absence of radiation, \( I \) - radiation intensity, \( n_2^{\text{eff}} = (dn/dI) \) - the coefficient of effective cubic nonlinearity.
The coefficient of cubic nonlinearity for the electrostrictive mechanism is determined by the concentration nonlinearity:

\[
 n_2^{\text{eff}} = (dn / dC)(dC/dI)
\]  

(2)

where \( C \) is the concentration of nanoparticles.

In nanosuspension the particle radius is much smaller than the radiation wavelength \( \lambda \), therefore the refractive index of the medium is proportional to the concentration of particles [8]:

\[
 n = n_1(1 + f\delta),
\]  

(3)

where \( \delta = (n_2 - n_1)/n_1 \); \( n_1 \) and \( n_2 \) are the refractive indices of the liquid and the dispersed phase, respectively; \( f = v_0C \) is the volume fraction of the dispersed phase, \( r \) is the radius of the nanoparticle, \( v_0 = (4/3)\pi r^3 \) is the volume of the nanoparticle.

Balanced equation describing the dynamics of concentration of nanoparticles considering diffusion and electrostrictive flows can be written as [4]:

\[
 \frac{\partial C}{\partial t} = D\Delta C - \text{div}(\gamma C\nabla I),
\]  

(4)

where \( D \) is the diffusion coefficient, \( \gamma = 4\pi\beta(\tilde{c}nk_BT)^{-1} \), \( \beta \) is the particle polarizability, \( k_B \) is the Boltzmann constant, \( \tilde{c} \) is the velocity of light in vacuum, \( I(r) \) is the intensity of radiation.

In the stationary mode equation (4) takes the next form (diffusion and electrostrictive flows are equal):

\[
 0 = -D\nabla C + \gamma C\nabla I.
\]  

(5)

This equation can be linearized in the case of low concentrations (\( C \ll 1 \)) and small it changes:

\[
 (C(r,t) = C_0(1 + C'(r,t)), C'(r,t) \ll 1).
\]  

(6)

\[
 0 = -D\nabla C' + \gamma\nabla I.
\]  

(7)

The exact solution of the equation (7) is:

\[
 C' = I(r)I_s^{-1}.
\]  

(8)

where \( I_s = \gamma^{-1}D \) is the “saturation” intensity.

Thus the changing of the particle concentration (and effective refractive index) is directly proportional to radiation intensity. It is a case of the classic cubic nonlinear response of the material. The coefficient of effective cubic nonlinearity of nanosuspension can be calculated using expression (2-3) and (8) [7]:

\[
 n_2^{\text{eff}} = n_1\delta f_0I_s^{-1},
\]  

(9)

where \( f_0 = v_0C \) is the initial volume fraction of the dispersed phase.

The self-focusing regime in the cubic nonlinearity medium is characterized by the critical beam power [5]:

\[
 P_s = \frac{\lambda^2}{64\pi^2 n_2^{\text{eff}}}.
\]  

(10)

where \( \lambda \) is the radiation wavelength.
Using (3-4) one can found the critical beam power for nanosuspension with electrostrictive response:

\[ P_{s0}^{el} = \frac{\lambda^2 I_s}{64\pi^2 n_1^2 f_0}. \]  

(11)

For ordinary parameters of nanosuspension formula (5) gives the values \( P_{s0}^{el} \approx 10^2 \) W.

The above analysis is valid only for small intensities of radiation (\( I \ll I_s \)), when the coefficient of effective cubic nonlinearity is a constant parameter of the medium.

3. Self-focusing of the power light beam

For great intensity beam (\( I >> I_s \)) the nonlinear response of the nanosuspension does not match to the cubic nonlinearity, because the concentration is not a linear function of the light intensity. The task of mass transfer in the light field requires the solving of the equation (5), because the concentration change is comparable to the primary value in this case.

We will consider the transparent nanosuspension under the influence of laser radiation with Gaussian intensity profile [9]:

\[ I = I_0 e^{-r^2/\rho^2}, \]

(12)

where \( I_0 \) is the radiation intensity on the beam axis, \( r \) is the radial distance from the beam axis, \( r_0 \) is the beam radius.

The general solution of the equation (5) is looking for in the form of:

\[ C_s = B \exp \{I_0 I_s^{-1} e^{-r^2/\rho^2}\}. \]

(13)

where \( B \) is the normalization constant.

The constant of normalization \( B \) is given from conservation of particle number:

\[ \int_0^\infty 2\pi r dr = \pi R^2 C_0. \]

(14)

where \( R \) is the radius of the cylindrical cell, \( C_0 \) is the initial concentration of dispersed particles.

Let’s introduce dimensionless parameter of light intensity \( \alpha = I_0 I_s^{-1} \). Figure 1 shows the concentration of the nanoparticles versus the normalized distance from the axis of the beam \( \rho = (r/r_0) \) for the different intensity values \( \alpha \) (it was calculated for \( R/r_0 = 5 \)).

The change of the concentration \( \Delta C_{st} \) on the axis of the beam:

\[ \Delta C_{st} = B(\exp \alpha - 1). \]

(15)

where \( \Delta C_{st} = (C_{st} - 1) \).

Figure 2 plots the change of the concentration \( \Delta C_{st} \) on the axis of the beam (\( \rho = 0 \)) versus normalized intensity \( \alpha \). The change of the particle concentration is not proportional to the radiation intensity (as opposed to the usual cubic nonlinearity).

The self-focusing regime is analyzed usually in paraxial approximation [5]. In our case the refractive index profile in the nanosuspension near the axis beam is expressed from (3) and (13):

\[ n = n_1 + n_0 \delta n_0 B e^\alpha [1 - \alpha (r^2/r_0^2)] \]

(16)

The analysis of the critical power for this case gives the next expression:
\[ P'_s = P_s / P'_{s0} = \alpha (e^\alpha - 1)^{-1}. \] (17)

**Figure 1.** The concentration of nanoparticles \( C_{st} = C / C_0 \) (a.u.) versus distance \( \rho \) (a.u.) from the axis of the beam for different values of the intensity of the light: \( \alpha_1 = 0.5 \) ( ); \( \alpha_2 = 1 \) ( ); \( \alpha_3 = 2 \) ( ).

**Figure 2.** The change of the concentration \( \Delta C_{st} \) on the axis of the beam versus normalized intensity \( \alpha \) ( ). The linear dependence \( \Delta C_{st} = \alpha \) is shown for comparison ( ).

Figure 3 plots the critical power of the beam versus normalized intensity \( \alpha \) (according to the formula (17)).

**Figure 3.** The critical power of the beam (arb. un.) versus normalized intensity \( \alpha \).

We can see the sharply decreasing of the critical power value when \( \alpha \gg 1 \). Thus this region is more convenient for the experimental realization.
4. Conclusions
We have analyzed the two-dimensional diffusion in the nanosuspension with electrostrictive nonlinearity in a Gaussian beam of radiation. In case of high intensity beam the nonlinear response of the nanosuspension does not match to the cubic nonlinearity. The expression was achieved for light induced stationary concentration response in nanosuspension. The resulting expression shows that the critical power value decreases significantly for high value α.

The results are relevant to nonlinear optics of nanosuspension [10-12], including the optical diagnostics of such nano-materials [13-14].

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