Thermal lens spectroscopy in two-component liquid

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Abstract. In two-component fluid (binary mixture) the heat flow can cause concentration stream arising from occurrence of thermodiffusion phenomenon (Soret effect). As a result these phenomenon changes the magnitude of the transport coefficients of the mixture. In this paper the theoretical analysis of the light-induced thermodiffusion mass transfer in two-components liquid in a field of Gaussian beam was carried out. It was calculated a concentration contribution to the thermal lens response. The results are relevant to the study of the radiation self-action in the nanosuspension and optical diagnostics of such materials.

1. Introduction
Nonlinear optical diagnostics of materials involves methods based on different mechanisms of light induced modulation of optical constants of medium [1-3]. For example, nonlinear optical method is used in thermal lens spectrometry and thermo-optical diagnostics of materials [4,5]. A thermal lens response has its own peculiarities in two-component liquid, because in addition to normal thermal response associated with thermal expansion of the liquid, concentration flows can occur here arising from the phenomenon of thermal diffusion (Soret effect) and electrostriction.

The purpose of this work is the theoretical analysis of the contribution of concentration mechanisms nonlinearity in the formation of light induced response in the two component liquid.

2. Thermal lens scheme
Thermal lens technique is widely used for the optical diagnostics of materials [6-8]. The light-induced thermal lens in a homogeneous fluid is formed as a result of thermal expansion of a medium. In two-component fluid the heat flow also can cause concentration stream arising from occurrence of thermodiffusion (Soret effect). 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. A change in the concentration of dispersed components in the liquid as a result of these phenomena changes the magnitude of medium thermal lens response.

Let's take a two beam thermal lens scheme (see Figure 1). The reference (Gaussian) beam generates the thermal field in the optical cell with nanosuspension. The second (Gaussian) beam tests the thermal lens.
Figure 1. A thermal lens scheme.

Thermal lens signal shows the change of the beam intensity on the optical axis behind the screen:

$$g(t) = \frac{I(t) - I(0)}{I(0)}.$$  \hspace{1cm} (1)

To calculate the thermal lens signal we use the term for the lens transparency of the cell [5]:

$$g_u = -\frac{2(z_1/l_0)\Phi_{nl}(0)}{(1+z_1^2/l_0^2)(1+3z_1^2/l_0^2)}.$$  \hspace{1cm} (2)

Where $z_1, z_2$ are the distance from the cell center to the Gaussian beam waist and to screen, respectively (fig. 1), $l_0 = \pi r_0^2/\lambda$ is the confocal parameter, $r_0$ is the radius of the Gaussian beam waist, $\Phi_{nl}(0)$ is the nonlinear phases in optical cell on the beam axis.

Nonlinear phase consists of two deposits: the first appears due to thermal expansion of the liquid phase and the second is associated with a change in the concentration of dispersed particles:

$$\Phi_{nl}(0) = \Phi_T(0) + \Phi_C(0).$$  \hspace{1cm} (3)

$$\Phi_T(0) = k \int_0^d \frac{\partial n}{\partial T} \Delta T(z, r = 0) dz.$$  \hspace{1cm} (4)

$$\Phi_C(0) = k \int_0^d \frac{\partial n}{\partial C} \Delta C(z, r = 0) dz.$$  \hspace{1cm} (5)

Where $k = 2\pi/\lambda$ is the wave vector of the probing beam, $d$ is the thickness of a layer of liquid.
3. Light induced lens response in nanosuspension

To determine the value of thermal lens response we should consider the thermal and mass transport processes. The next system of balanced equations describes the processes that occur when a binary mixture is exposed by light field [8]:

\[ c_p \rho \frac{\partial T}{\partial t} = \chi \nabla^2 T + \alpha I_0 \] 
\[ \frac{\partial C}{\partial t} = -\text{div}(J_d + J_{el} + J_{thd}) \]  

Here \( T \) is the liquid temperature, \( C \) is concentration of the nanoparticles, \( \chi \) is coefficient of thermal conductivity; \( c_p, \rho \) are the specific heat capacity and density of the fluid, respectively; \( \alpha \) – radiation absorption coefficient, \( I_0 \) is the beam intensity.

We have three concentration flows:

\[ J_d = -D \nabla C , \]  
\[ J_{el} = \gamma C \nabla I , \]  
\[ J_{thd} = D_T C (1 - C) \nabla T . \]  

here \( J_d \) is diffusion flow, \( J_{el} \) is electrostrictive flow, \( J_{thd} \) is thermal diffusion flow. \( D \) и \( D_T \) are the diffusion and thermal diffusion coefficients, respectively, \( \gamma = (2 \beta / c n_1) \), (\( \beta \) and \( b \) are polarizability and mobility of nanoparticle, respectively), \( c \) is the light velocity [9].

We will consider the case of low concentrations ( \( C << 1 \) and small it changes. This approximation allows the presenting of the desired concentration in the form of sum of unperturbed parts \( C_0 \) and perturbed \( C_N \):

\[ C(r,t) = C_0 + C_N(r,t) = C_0 (1 + C'(r,t)) \]  
\[ C'(r,t) = \frac{C_N(r,t)}{C_0} << 1 . \]  

We have next boundary (13) and initial (14) conditions:

\[ C(R,t) = C_0 , \]  
\[ T(R,t) = T_0 , \]  
\[ C(r,0) = C_0 , \]  
\[ T(r,0) = T_0 , \]  

where \( R \) is the radius of the cylindrical cell, \( T_0 \) is the temperature at the border cell.

After the necessary integration we obtain an exact solution:

\[ T(r,t) = T_0 + \alpha d_0 \chi \sum_{n=1}^{\infty} J_n \left( \frac{\mu_n r / R}{\tau_{nh}} \right) \left( \frac{\mu_n}{\tau_{nh}} \right)^2 J_n^2 \left( \frac{\mu_n}{\tau_{nh}} \right) \left[ 1 - \exp \left( -\frac{\mu_n^2 r^2 / 4 R^2}{\tau_{nh}} \right) \right] \]  
\[ \tau_{nh} = 4 R^2 c_p \rho (\mu_n \chi)^{-1} , \]  

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where \( \mu_n \) is the positive roots of the equation \( J_0(\mu_n) = 0 \); \( \tau_{th} \) is relaxation time of the thermal response.

Further it is assumed that the temperature field is set in the system faster than the particle concentration distribution. It allows solving the task in terms of stationary temperature [10].

Decision of the task (6-7) can be obtained by using the Green's function:

\[
C(r, t) = g_1 r_1^2 J_0(\mu_n) \sum_{n=1}^\infty \frac{\sqrt{\mu_n^4 r_1^2 / (4 R^2)}}{\mu_n^2 J_0^4(\mu_n)} \exp\left[-\pi \mu_n^2 r_1^2 / 4 R^2\right] \left(1 - \exp\left[-\nu \mu_n^2 / \tau_{th}^d\right]\right) + \gamma C_0 D^4 \exp\left[-r^2 / r_1^2\right] \left(1 - \exp\left(-t / \tau_{thd}\right)\right) \]  

(17)

\[
\tau_{thd} = R^2 D^{-1} \]  

(18)

\[
\tau_d = r_1^2 D^{-1} \]  

(19)

where \( \tau_{thd} \) is relaxation time of the thermodiffusion response, \( \tau_d \) is relaxation time of the diffusion response.

First and second terms describe the thermal diffusion and electrostrictive effects, respectively.

The obtained results allow us to calculate the summarized light induced lens response (for example, in stationary case):

\[
S^\text{tot}_n = \frac{z_1 k I_0 d}{4} \left( \frac{\alpha \chi^{-1} S \gamma \rho n^2 \left( \frac{\partial n}{\partial T} \right) - \alpha \chi^{-1} n^2 \left( \frac{\partial n}{\partial C} \right) + \gamma C_0 D^{-1} \left( \frac{\partial n}{\partial C} \right)}{1 + \frac{z_1^2}{l_0^2} \left(1 + 3 \frac{z_1^2}{l_0^2}\right)} \right) \]  

(20)

Thus, the expression was achieved for the light induced lens response in the binary mixture.

The most interesting case was achieved for the light induced lens response of the nanosuspension. In nanodispersive medium particle radius is much smaller than the radiation wavelength \( \lambda \) and the refractive index of the medium is proportional to the concentration of particles (for dilute systems):

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

(21)

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

The experimental results show that for nanosuspension all three components in (20) can be compared by size [11].

4. Conclusions

We have analyzed the two-dimensional diffusion in the nanosuspension with two concentration nonlinearities (thermodiffusion and electrostrictive) in a Gaussian beam radiation field. As a result the exact expressions were achieved for the light induced lens response in the binary mixture. We get the exactly expression describing the kinetics of the thermal expansion, thermal diffusion and electrostrictive effect [6-7].

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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