Light-induced thermodiffusion in two-component media

V Ivanov¹, G Ivanova², K Okishev³ and V Khe⁴

¹ Professor, FESTU, Khabarovsk, Russia
² Senior Lecturer, FESTU, Khabarovsk, Russia
³ Associate Professor, FESTU, Khabarovsk, Russia
⁴ Engineer, FESTU, Khabarovsk, Russia

E-mail: khe@ngs.ru

Abstract. We have theoretically studied the optical transmittance response of thin cell with liquid containing absorbing nanoparticles in a Gaussian beam field. The transmittance spatial changing is caused by thermal diffusion phenomenon (Soret effect) which produces the variations of concentration of absorbing nanoparticles. The thickness of optical cell (including windows) is significantly less than the size of the beam. As a result, an exact analytical expression for the one-dimensional thermal task is derived, taking into account the Soret feedback that leads to the temperature rising on the axis of a Gaussian beam. We have experimentally studied this phenomenon in carbon nanosuspension.

1. Introduction

The light-induced phenomena in nanosuspensions are widely used in nonlinear optics [1-2], dynamic holography [3-4], optical diagnostics of materials [5]. There are special mechanisms of optical nonlinearity in such media associated with the concentration flows due to light-induced phenomena [2-4]. The redistribution of the components concentration results in a corresponding change in the refractive index (and absorption) of the liquid. The thermal diffusion (Soret effect) is one of such phenomenon which was investigated theoretically and experimentally [1, 5]. We proposed a new thermodiffusion scheme for a thin-layer cell [6]. In this work we have theoretically studied the optical transmittance response of thin cell with liquid containing absorbing nanoparticles. The experimental results are represented for carbon nanosuspension.

2. Theoretical model

We consider one-beam scheme measurement of the thermal diffusion response in the two-component liquid layer (Fig. 1). Let’s the absorption coefficient \( \alpha \) is determined entirely by one component of liquid with mass concentration \( C (\alpha = \beta C) \), where \( \beta = (\partial \beta / \partial C) \) – constant) and liquid is located in a thin cell with thickness \( d \).

We have for a Gaussian beam distribution in the cell plane perpendicular to the optical axis \( z \):

\[
I = I_0 \exp\left(-r^2 / \omega^2\right),
\]

where \( \omega \) is the radius of the beam, \( r \) is the distance from the axis of the beam, \( I_0 \) is intensity on the axis of a Gaussian beam.

We assume a small thicknesses of the liquid layer \( d \) and of the cell window \( L : (d + 2L) \ll \omega \). In this case the radial heat flux is negligible and we get one-dimensional heat propagation task:
where \( c_m, \rho_m \) are the specific heat capacity and density of the liquid medium, \( T_m \) is the liquid temperature, \( \chi_m \) is the heat conductivity coefficient. The absorption is low (\( ad \ll 1 \)).

The similar thermal task was also considered for the temperature change in the window of the cell \( T_w \):
\[
c_m \rho_m \frac{\partial T_w}{\partial t} = \chi_m \frac{\partial^2 T_w}{\partial z^2},
\]
where \( c_m, \rho_m, \chi_m \) are thermo-physical parameters of the window material.

**Figure 1.** The scheme for the thermal task in “Soret cell”: 1 – lens, 2 – cell with liquid, 4 – photodetector, \( l_1 \) – distance between beam waist and cell center.

Boundary conditions are as following:

\[
\chi_m \left( \frac{\partial T_m}{\partial z} \right)_{z = \frac{d}{2} + \frac{dL}{2}} = -\gamma (T_m - T_0),
\]
\[
T_m (d/2) = T_m (d/2),
\]
\[
\chi_m \left( \frac{\partial T_m}{\partial z} \right)_{z = \frac{d}{2} + dL/2} = \chi_m \left( \frac{\partial T_m}{\partial z} \right)_{z = \frac{d}{2} + \frac{dL}{2}}.
\]

where \( \gamma, T_0 \) are the convective heat transfer coefficient and ambient temperature accordingly, \( T_0 = T_m (L + d/2) \).

In the stationary mode we have the following solution of the system (2-6) for temperature distribution in the liquid \( T_m^s (z) \) and the cell window \( T_w^s (z) \):

\[
T_m^s = T_0 + T (d/2) + a \Gamma (r) \chi_m^{-1} \left\{ \left( d/2 \right)^2 - z^2 \right\},
\]
\[
T_w^s = T_0 + T (d/2) + a \delta \Gamma (r) \chi_m^{-1} \left\{ \left( d/2 \right)^2 - z^2 \right\},
\]
\[
T (d/2) = a \Gamma (r) d \left\{ \gamma^{-1} + L \chi_m^{-1} \right\}.
\]

For the layer thicknesses \( d \ll L \) we can neglect the temperature change along the thickness of the cell and take it equal \( T_m^s (r) \):
$T_n(r) = T_0 + \alpha dA_l \exp(-r^2/\omega^2) \tag{10}$

where $A = (Lz_m^{-1} + \gamma^{-1} + dz_m^{-1}/2)$.

Now let’s consider the effect of thermal diffusion. Since the diffusion processes are much orders of magnitude slower than the heat transfer processes, we can assume that thermal diffusion of absorbing components occurs in stationary temperature field that is determined by formulas (7-9), but with a concentration-dependent absorption coefficient.

Balanced equation for the concentration of absorbing particles $C$ is as following:

$$\frac{\partial C}{\partial t} = D\Delta C + D_r C(1-C)\Delta_r T, \tag{11}$$

here: $D$ is the diffusion coefficient of absorbing particles, and $D_r$ is the thermal diffusion coefficient, $\Delta_r$ is the Laplacian operator in cylindrical coordinates.

The change in the concentration of the absorbing components is small compared to the primary one: $C = C_0(1+C_1) \approx C_1$, where $C_1 << 1$.

Let us initial concentration of the absorbing particles is small too $(C_0 << 1)$, thus the task is linearised. We have the following equation with boundary conditions:

$$\frac{\partial C_i}{\partial t} = D\Delta C_i + D_r \Delta_r T, \tag{12}$$

$$\nabla C_i(r) |_{r=0} = 0, \tag{13}$$

$$C_i(r = \infty) = 0. \tag{14}$$

The solution is taken by using the Green function method:

$$C_i(r,t) = \frac{I(r)}{I_s} \left\{ \exp \left[ \frac{-r^2}{\omega^2} \right] - \left(1 + \frac{t}{\tau} \right)^{-1} \exp \left[ \frac{-r^2}{\omega^2} \left(1 + \frac{t}{\tau} \right)^{-1} \right] \right\}, \tag{15}$$

where $I_s = (C_0 \beta dA_l s_e^{-1})^{-1}$, $S_r = D_r D^{-1}$ is the Soret coefficient, $\tau = \omega^2 (4D)^{-1}$ is the relaxation time.

Then we get for the intensity of the passed radiation $I_{\text{pass}}(r)$:

$$I_{\text{pass}}(r) = \left\{ 1 - \beta C_0 \right\} \left[ 1 + C_i(r,t) \right] \frac{I_0}{I_s} \exp \left[ \frac{-r^2}{\omega^2} \right], \tag{16}$$

We use this term to calculate the total passed power:

$$P_{\text{pass}}(t) = (1 - \alpha_0 d) P_0 + \left( \frac{I_0}{I_s} \right) \frac{\alpha_0 d P_0}{2(\tau + t)}, \tag{17}$$

where first term is the passed radiation without Soret effect. We neglect the Fresnel reflections for simplicity.

Using (10,15) we get for the temperature of the nanosuspension:

$$T_n(r) = T_0 + S_r^{-1}(1+C_i(r)) \left[ \frac{I(r)}{I_s} \right]. \tag{18}$$

The last expression describes a so-called ‘Soret feedback’ that leads to the temperature rising near the axis of a Gaussian beam.

3. Experimental results and discussions

We used the suspension of carbon nanoparticles (with a diameter of 0.1-0.2 µm) in water. The initial mass fraction of nanoparticles was $C_0 = 6 \cdot 10^{-3}$. The vertical laser beam (He-Ne laser, wavelength 0.63 µm, 60 mW power) was directed on the horizontal optical cell. The thickness of the cell window (130 µm) and liquid laer (70 µm) was smaller than the light beam radius (1200 µm). The temperature of the upper window was controlled by the infrared thermograph “IRTIS-2000”.
Figure 2a shows the time evolution of the total signal (passed beam power) after beginning of the cell illumination. The disperse phase concentration and absorption coefficient decline both as a result of action of thermal diffusion. Figure 2b shows the time evolution of surface temperature in the center of the window.

![Figure 2](image-url)

**Figure 2.** The time evolution of the total signal (passed beam power) after beginning of the cell illumination (2a); the time evolution of surface temperature in the center of the window (2b).

The enlightenment time corresponds to the diffusion processes. The real value of the absorbing factor was sufficiently high ($a d \approx 2$). Thus the theoretical model corresponds to the experimental dates only approximately. We have the evaluation for the Soret coefficient $S_T \approx 2 \cdot 10^{-3} K^{-1}$ which corresponds to the characteristic values for nanosuspensions.

### 4. Conclusions

It is shown that thermal diffusion response in the two-component liquid contains a Soret feedback contribution due to thermodiffusion concentration change of absorbing components. The magnitude of this contribution can be quite large for the low radiation intensity. Thus, a modulation of the self-induced absorption coefficient should be taken into account in the thermal lens spectrometry analysis of multi-component liquids [4-6]. Received expressions can be used for experimental determination of the heat and mass transfer coefficients in such multi-component materials [7-9].

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