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Diagnostics of nanosuspension by the dynamic holography method

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Abstract. The theoretical analysis of the dynamic holograms efficiency in the nonideal nanoparticle gas is carried out. The electrostrictive mechanism of optical nonlinearity of the medium is due to the forces operating on the particles of the dispersed phase in gradient light field. The recording of the dynamic hologram is significantly non-linear process at high intensities of radiation. The amplitude of phase grating depends exponentially versus intensity but it saturates at the high filling factor due to the repulsion effect.

1. Introduction
Nonlinear optical diagnostics of materials involves methods based on different mechanisms of light induced modulation of optical constants of medium [1-3]. Multicomponent nanoparticles medium (liquid phase mixture, suspension, emulsion) is characterized by concentration perturbations caused by the redistribution component in a two-phase medium in a laser field [4-6]. The light induced change in the concentration of polymer nanoparticles provides various modifications of optical noncontact diagnostic methods in near real time. Such well known methods as the thermal lens and the dynamical holography are successfully used for the investigation of the mass transport phenomena in multicomponent liquids [7-9]. In a gradient light field the nanoparticles in the transparent medium are controlled by the electrostrictive forces, causing changes in their concentrations [10-12]. The medium is characterized by a cubic nonlinearity in this case that is correct only for small intensities of radiation [13-15]. For large radiation intensities the potential energy of particle is more than heat one and it requires consideration of non-linearity of the highest order.

The purpose of this paper is the theoretical analysis of the dynamic holograms efficiency in the nonideal nanoparticle gas in the case of the high intensity of radiation, when the concentration changing is much more than the initial value. This case is very interesting for the high intensity dynamic holography of dispersed liquid-phase media, as well as for optical diagnostics of nanomaterials [16-20].

2. Two-wave holographic scheme
The two-wave scheme is the simplest method of the holographic diagnostics. Dynamic hologram method (Forced Raleigh Scattering) based on the recording of the dynamic holograms in nonlinear material while reading it. Usually two references beams (with equal intensity) form the interference picture in the nonlinear material, thus $I = I_0(1 + \sin Kx)$ is the intensity spatial distribution along the layer of the medium (x-axis), $K = 2\pi/\Lambda$ and Λ are the space wave vector and the period of the

elementary hologram. The reading beam (with a different wavelength) is directed perpendicular to the wave vector of the hologram.

The next expression defines a diffraction efficiency of the hologram [1]:

$$\eta = I_{1p}/I_{0p} \,,$$ (1)

where I_{0p} is the intensity of the incident probe beam; I_{1p} is the intensity of the diffracted beam.

If medium is transparent and the phase modulation amplitude is small ($\varphi_1 \ll 1$) the diffraction efficiency of a thin phase hologram is [1]:

$$\eta = \left\{ 2\pi\Delta n L \lambda^{-1} \right\}^2,$$ (2)

where L is the thickness of a nanofluid layer, Δn is the amplitude of the space modulation of the refractive index.

In the case of non-resonant non-linearity (for weakly absorbing media) it is used the parameter - $n_2^{eff}[m^2/W]$, which characterizes the change of the refractive index of the medium under the influence of incident radiation (cubic nonlinear response):

$$n = n_0 + n_2^{eff} I,$$ (3)

where n_0 is the refraction index of the medium in the absence of radiation, I is the incident intensity, $n_2^{eff} = (\partial n / \partial I)$ is the coefficient of effective cubic nonlinearity.

In nanodispersive medium particle radius is much smaller than the radiation wavelength λ and the refractive index of the medium is proportional to the concentration of particles (for dilute systems):

$$n = n_1(1 + f\delta) \,,$$ (4)

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; $f = (4/3)\pi a^3 C$ is the volume fraction of the dispersed phase, a is the radius of the nanoparticle, C is the concentration of nanoparticles [13].

It is sufficient to calculate the amplitude of the concentration grating C_1 for defining the amplitude of the space modulation of the refractive index in the material:

$$\Delta n = (\partial n/\partial C)C_1.$$ (5)

3. Electrostrictive mechanism of optical nonlinearity

The electrostrictive mechanism of optical nonlinearity of the medium is due to the forces operating on the particles of the dispersed phase in gradient light field. Depending on the sign of the polarizability the nanoparticles may be drawn (if the refractive index of the dispersed phase material is greater than the dispersion medium) or pushed out (otherwise) from areas with greater intensity of the electromagnetic wave [1].

The system of balance equations for the concentration of particles for this case is written as follows [13]:

$$dC/dt = -\operatorname{div}(-D\nabla C + \gamma C(1 - \nu C)\nabla I).$$ (6)

where D is the diffusion coefficient of particle, $J = \gamma C \nabla I$ is electrostrictive flow, $\gamma = (2\beta b / c n_1)$, ($\beta$ and b - polarizability and mobility of nanoparticle, respectively), c is the light velocity, ν is the constant of the particle interaction.

The proposed phenomenological model involves the particles interacting (repulsion). This model gives the value $\nu = 1$ for the simple model of the hard sphere potential [11].

In case of substantial changes of concentrations of particles the original equation (6) can be solved in the stationary mode:

$$- D \nabla C + \gamma C (1 - \nu C) \nabla I = 0 . \tag{7}$$

The solution of the equation (7) can be written as ($\nu = 1$):

$$C_R = (f_0 + \exp[-\alpha SinX])^{-1}, \tag{8}$$

where $C_R = C / C_0$ is normalized concentration of nanoparticles, $X = x / \Lambda$ is dimensionless coordinate, $\alpha = I_0 / I_{th}$ is normalised intensity, $I_{th} = D\gamma^{-1}$ is some "thermal" intensity of radiation.

The case of the ideal gas of nanoparticles (without repulsion $\nu = 0$) gives the next expression:

$$C_R = \exp[\alpha SinX], \tag{9}$$

The amplitude of the concentration grating depends exponentially versus intensity for low values of filling factor f_0. The expressions (8-9) give the same result for both cases (Figure 1).
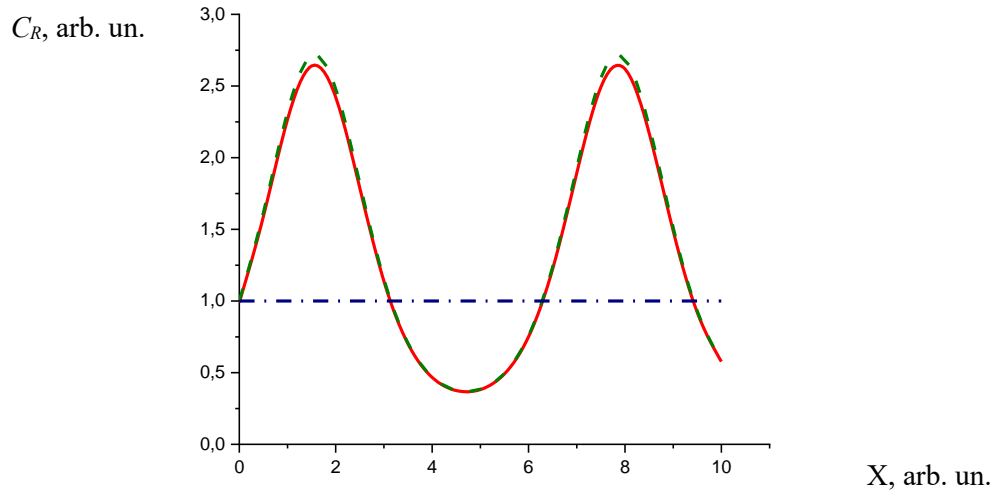

Figure 1. Concentration gratings of nanoparticles for intensity α=1 ($\alpha = I_0 / I_{th}$ is dimensionless intensity of radiation): in ideal gas (━ ━ ━) according to (9); in the non-ideal gas (━━━) according to (8). The initial concentration is constant for α=0 (━ · ━); $f_0 = 0.01$ (low concentration).

High value of the initial filling factor f_0 leads to the essential difference for these two models. The amplitude of the concentration grating in the non-ideal gas is noticeably less than in the case of ideal gase (Figure 2).
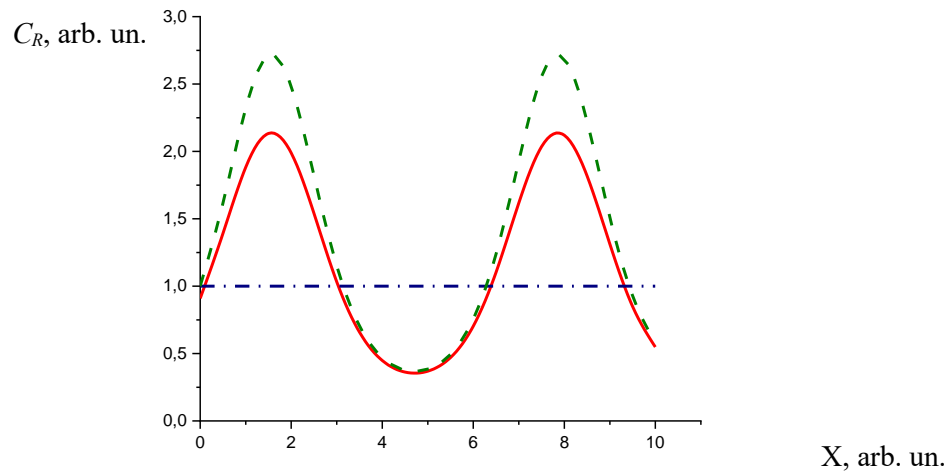

Figure 2. Concentration gratings of nanoparticles for intensity α=1: in ideal gas (– – –) according to (9); in the non-ideal gas (——) according to (8). The initial concentration is constant for α=0 (– · –); $f_0 = 0.1$ (large concentration).

The difference grows dramatically with the value of the intensity. The concentrations gratings are significantly nonsinusoidal for large initial concentration and high intensity. The ideal gas model (formula (8)) gives the high value of the concentration grating, but the hard sphere gas model gives much less nonlinear response (Figures 3).
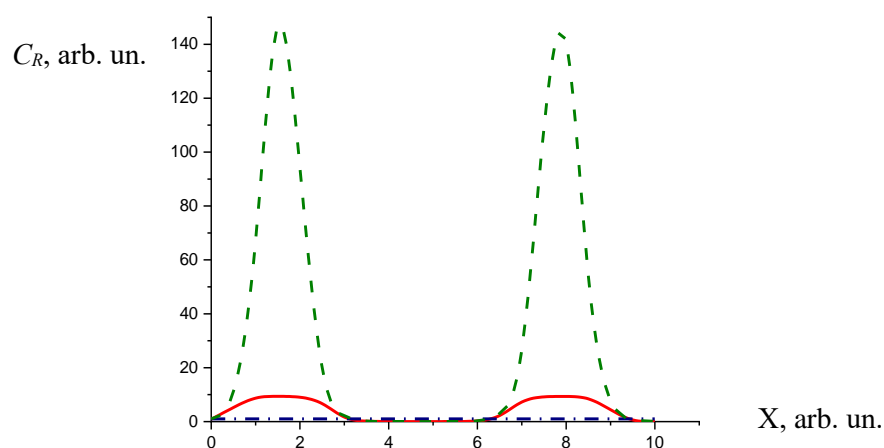

Figure 3. Concentration gratings of nanoparticles for intensity α=5: in the ideal gas (– – –) according to (9); in the non-ideal gas (——) according to (8). The initial concentration is constant for α=0 (– · –); $f_0 = 0.1$ (large concentration).

Thus the value of the nonlinearity (and the dynamic holograms efficiency) for the high intensity radiation is restricted by the repulsion effect in the case of the initial large particle concentrations.

4. Conclusions

The electrostrictive nonlinearity in the hard sphere gas model was theoretically investigated. The amplitude of the concentration grating depends exponentially versus intensity only for the low values of the intensity and initial filling factor and saturates for the high values of the both parameters.

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