Holographic method of the nanoparticles diagnostics in a fluid

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Abstract. The dynamic holography method is analyzed for the diagnostics of nanoparticles in a fluid. The concentration nonlinearities caused by electrostriction and thermodiffusion effects as well as the reorientational cubic response in nanosuspensions are considered. The results are relevant to the study of the optical diagnostics of such materials.

1. Introduction
Multicomponent nanoparticles medium (nanosuspensions, microemulsions, aerosols) are characterized by a series of specific mechanisms nonlinearities that not appear in single-component media [1-3]. In particular, these include the concentration non-linearity caused by the redistribution of the dispersed component in a two-phase medium in a laser field. The concentration flows in the medium can be caused by different mechanisms of interaction of radiation with matter. The optical nonlinear response of the nanosuspensions includes the parameters of the nanoparticles. The thermodiffusion effect (Soret effect [4]) in the binary mixtures was experimentally investigated by the optical nonlinear methods in many works [5-8]. 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 [9-12].

The purpose of this work is the theoretical analysis of the contribution of concentration and orientation nonlinear mechanisms in the formation of cubic nonlinear response in the nanosuspension.

2. Electrostrictive mechanisms of the cubic nonlinearity in nanosuspension
The coefficient of cubic nonlinearity for the concentration mechanism is determined by the concentration nonlinearity:

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

(1)

Where \( n \) the refraction index of the medium is, \( I \) is the radiation intensity, \( C \) is the concentration of nanoparticles, \( n_2^{\text{eff}} = (dn/dI) \) is the coefficient of effective cubic nonlinearity.

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 [3]:

\[ n = n_1(1 + fe) \]  

(2)
where \( \varepsilon = (n_2 - n_1)/n_1 \) and \( n_2 \) are the refractive indices of the liquid and the dispersed phase, respectively; \( f = v_0 C \) is the volume fraction of the dispersed phase, \( a_0 \) is the radius of the nanoparticle, \( v_0 = (4/3)\pi a_0^3 \) is the volume of the nanoparticle.

The system of balance equations for the concentration of particles and heat flow for this case is written as follows [6]:

\[
\frac{dC}{dt} = -\text{div}(J_d + J_{el}).
\]

Here \( (J_d + J_{el}) \) are concentration flows:

\[
J_d = -D \nabla C,
\]

\[
J_{el} = \gamma CVI,
\]

where \( J_d \) is the diffusion flow, \( D \) is the diffusion coefficient of particles, \( J_{el} \) is the electrostrictive flow, \( \gamma = (2\beta b/cn_i) \), (\( \beta \) and \( b \) are polarizability and mobility of microparticle, respectively), \( c \) is the light velocity, \( I = I_0(1 + \sin Kx) \) is the intensity spatial distribution along the layer of the medium (x-axis), \( K \) is the space wave vector of the elementary hologram.

Assuming a one-dimensional problem, the solution of equations (7) is founded in the form of:

\[
C(x,t) = C_0 + C_1(t) \sin Kx,
\]

where \( C_0 \) is the average particle concentration.

The linearized system is easily solved (with the approximation \( C_1/C_0 \ll 1 \)):

\[
C_1(t) = C_0 I_0 I_{sat}^{-1}[1 - \exp(-t/\tau_d)],
\]

\[
I_{sat} = D\gamma^{-1},
\]

\[
\tau_d = (K^2 D)^{-1},
\]

where \( I_{sat} \) is the “saturation” intensity, \( \tau_d \) is the diffusion relaxation time.

For great intensity beam ( \( I \gg I_s \) ) 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.

Using the stationary solution (13) for the amplitude of concentrations grating the expressions for nonlinear coefficients can be obtained [13]:

\[
n_2^{nr} = n_2 \varphi_0 I_{sat}^{-1},
\]

where \( \varphi_0 = (4/3)\pi a_0^3/C_0 \) is the initial volume fraction of the dispersed phase.

In opposite to the thermal nonlinearity the electrostriction cubic coefficient \( n_2^{nr} = n_2 \varphi_0 I_{sat}^{-1} \) does not depend on the relaxation time.

It was experimentally obtained the value \( 10^{-6} \text{ cm}^2/\text{kW} \) for electrostrictive nonlinearity coefficient in nanosuspensions (so-called artificial Kerr medium) [11,14].

3. Thermodiffusion effect

In two-component fluid the heat flow also can cause concentration stream arising from occurrence of thermodiffusion effect. A thermally induced mechanism of particle drift in a nonuniform temperature field is known as thermal diffusion (thermophoresis) or Soret effect in liquid-phase binary mixtures [5].

The balance equations for the concentration of nanoparticles for this case is written as follows
\[ c_p \rho dT / dt = -\text{div} J_1 + \alpha I_0 (1 + \sin Kx), \]  
\[ dC / dt = -\text{div}(-D \nabla C - C(1 - C)D_T \nabla T), \]
where \( J_1 = -\chi \text{grad} T \) is the heat flow, \( T \) is temperature of the material, \( \chi \) is the thermal conductivity of the material, \( c_p \) and \( \rho \) are the specific heat capacity and density of the medium, \( \alpha \) is the absorption coefficient of the medium.

The solving of the one-dimensional problem for the temperature amplitude is founded in the next form:
\[ T_1(t) = T_1(t) \sin Kx, \]
The solution (11) for the temperature amplitude \( T_1 \) is:
\[ T_1 = T_1' (1 - \exp[-t / \tau_1]), \]
\[ T_1' = a I_0 \tau_1 (c_p \rho)^{-1}, \]
\[ \tau_1 = c_p \rho K^{-2}, \]
where \( \tau_1 \) is the thermal relaxation time, \( T_1' \) is the temperature amplitude in the steady state regime.

Let’s consider an expression for the holographic sensitivity of the dispersion medium, taking into account both (thermodiffusion and electrostriction) concentration mechanisms.

The linearized equation (12) is easily solved by taking into account (15):
\[ C_1 = C_1' (1 - \exp[-t / \tau_1]), \]
\[ C_1' = \alpha \xi C_s s T_1 K^2, \]
where \( C_1' \) is the concentration amplitude in the steady state regime.

The expression for nonlinear coefficients for this case is the next:
\[ n_2^{m2} = n_1 \exp(\phi_2) \alpha_\xi^{-1} s \tau K^{-2}, \]
It was experimentally obtained the value \( 10^{-7} \text{ cm}^2/\text{kW} \) for nonlinearity coefficient in nanosuspensions [5].

4. Orientation nonlinearity
The optically induced birefringence in the nanosuspension with non-spherical particles presents the orientation mechanism of the cubic nonlinearity [1]. In the classical model the macroscopic polarization can be obtained by performing an average on the microellipsoid angular displacement of the ensemble of identical microellipsoidal particles [15]. The induced dipole appears due to light field and the opposite action of the rotational diffusion. The model gives the next expression for cubic nonlinearity coefficient \( n_2^{m2} \):
\[ n_2^{m2} = 8 \pi^2 C (\alpha_{\mu} - \alpha_\perp)^2 / 15 n_0^b k_T T, \text{[cm}^2/\text{kW}], \]
where \( C \) is the particle concentration, \( k_T \) is the Boltzman factor, \( \alpha_{\mu} \) and \( \alpha_\perp \) are the values of microellipsoid polarizability along the semi-major and the semi-minor axes \( a \) and \( b \). The numerical estimation gives the theoretical value about \( 10^{-10} \text{ cm}^2/\text{kW} \) for \( n_2^{m2} \).

The values of the particle polarizabilities are given by the expressions [15]:
\[ \alpha_{\mu} = (a b^2 / 3) e_a (e_a - 1) / [1 + (a b^2 / 2) (e_a - 1) A_1 (a, b)], \]
\[ \alpha_\perp = (a b^2 / 3) e_\perp (e_\perp - 1) / [1 + (a b^2 / 2) (e_\perp - 1) A_1 (a, b)], \]
\[ \alpha_n = \left( \frac{ab^2}{3} \right) \epsilon_n (\epsilon_n - 1) \left[ 1 + \left( \frac{ab^2}{2} \right) (\epsilon_n - 1) A_1 (a, b) \right], \]  
(22)

where \( A_1 \) and \( A_2 \) are shape dependent coefficients, \( a \) and \( b \) are the semi-major and the semi-minor axis length, \( \epsilon_n \) is the fluid dielectric constant, \( \epsilon_r \) is the ratio of dielectric constant of the nanoparticle material to \( \epsilon_n \).

The dynamics of the induced birefringence is described by a Debye-like rotational diffusion equation. In the case of cylindrically symmetric particles one obtains the next solution:

\[ \Delta n_{\nu}^\alpha(t) = \Delta n_{\nu}^{\alpha s}[1 - \exp(-t/\tau_{\alpha})], \]

(23)

where \( \Delta n_{\nu} \) is the steady state value of induced birefringence, \( \tau_{\alpha} \) is the Debye’s response time, \( \eta \) is the fluid viscosity, \( G(b/a) \) is a form factor which depends on the \((b/a)\) ratio.

\[ \tau_{\alpha} = \frac{8\pi\eta a^3 \left( 1 - (b/a)^4 \right) / (9k_B T (2 - (b/a)^2)) G(b/a - 1)}{\left( b/a \right)^2}, \]

(24)

This type of nonlinearity has been observed experimentally in nanosuspensions [15]. The measured cubic coefficient \( n_{\nu}^{\alpha} \) is \( 10^{-7} \) cm²/kW. This value was interpreted in the model for the particles with intrinsic material birefringence [15].

5. Diffraction efficiency of the dynamic hologram

The dynamic holography method (Forced Raleigh Scattering) based on the recording of the dynamic holograms in nonlinear material while reading it. The next expression defines a diffraction efficiency of the hologram [1]:

\[ \eta = I_r/I_0, \]

(25)

where \( I_0 \) is the intensity of the incident beam; \( I_r \) is the intensity of the diffracted beam. The diffraction efficiency of a thin phase hologram is:

\[ \eta = t_0^2 J_n^2(\varphi_1), \]

(26)

where \( t_0 \) is amplitude transmittance of the unlighting hologram; \( \varphi_1 \) is amplitude modulation after light exposition; \( J_n \) is the Bessel function of the n-th order.

If medium is transparent and the modulation amplitude is small (\( \varphi_1 \ll 1 \)), the diffraction efficiency is equal:

\[ \eta = \left( 2\pi n t_0 L \right)^{-1}, \]

(27)

where \( L \) is the thickness of a nanofluid layer.

The summarized expression for diffraction efficiency is the next:

\[ \eta = \left( 2\pi n t_0 \right)^{-1} \left[ \alpha \left( \varphi_1 - \varphi_2 \right)^{-1} \left( \partial n / \partial T \right) + n_0 \varphi_2 (a_1^{\perp} S_1 K - I_0^2) + 8\pi^2 C (a_{IIi} - a_{II})^2 / 15n_0^2 k_B T^2 \right]. \]

(28)

where \( \left( \partial n / \partial T \right) \) is the thermal refractive index coefficient of the fluid.

This expression includes the thermal expansion (first term), the concentration nonlinearity (second term) and the orientation nonlinearity (third term). All types of the nonlinearities are characterised by the various relaxation times (formulas \((16), (9)\) and \((24)\) accordingly).

6. Conclusions
The dynamic holography method for the nanosuspension diagnostics was analyzed taking into account the concentration and orientation nonlinearities. The expression was received for the time dependence of the diffraction efficiency of the dynamic hologram in nanosuspension. The results demonstrate the extensive possibilities of the optical diagnostics of such nano-materials [16-20].

References
[1] Shen Y R 1984 The principles of nonlinear optics (Wiley, New York) p 112-119
[2] Malacarne L C, Astrath N G C, Medina A N, Herculano L S, Baesso M L, Pedreira P R B, Shen J, Wen Q, Michaelian K H and Fairbridge C 2011 Optics Express 19 4058
[3] Ivanov V I, Ivanova G D, Kirjushina S I and Mjagotin A V 2016 Journal of Physics: Conference Series 735 012013
[4] Morozov K I 2009 Phys. Rev. E 79 031204
[5] Leplla C and Wiegand S 2003 Philosoph. Mag. 83 1989
[6] Ivanov V I, Ivanova G D and Khe V K 2017 Proc. SPIE 10176 1017607
[7] Lee W, El-Ganainy R, Christodoulides D, Dholakia K and Wright E 2009 Optics Express 17 10277
[8] Piazza R 2009 Soft Matter 4 1740–1744
[9] Vicary L 2002 Philos. Mag. B. 82 447-452
[10] Ivanov V, Ivanova G, Okishev K and Khe V 2017 IOP Conf. Ser.: Mater. Sci. Eng. 168 012045
[11] El-Ganainy R, Christodoulides D, Rotschild C and Segev M 2007 Opt. Express 15 10207
[12] Chintamani P, Shalini M, Agnel P, Meera V, Tejas I H and Radha S 2014 International Journal of Chemical and Physical Sciences 3(5) 44
[13] Ovseychook O O, Ivanov V I, Mjagotin A V and Ivanova G D 2018 IOP Conf. Series: Journal of Physics: Conf. Series 1038 012091
[14] Philip J and Nisha M 2010 Journal of Physics: Conference Series 214 012035
[15] Pizzoferrato R, Marinelly M, Zammit U, Scudiery F, Martelluchi S and Romagnoli M 1988 Optics Communications 68 231.
[16] Kamanina V, Kuzhakov P V, Serov S V, Kukharchik A A, Petlitsyn A A, Barinov O V, Borkovskii M F, Kozhevnikov N M and Kajzar F 2013 Proc. SPIE 8622 86221B
[17] Ivanov V I, Ivanova G D, Okishev K N and Khe V K 2016 Proc. SPIE 10035 100354Y
[18] Ishaaya A A, Vuong L T, Grow T D and Gaeta A L 2008 Opt. Lett. 33 13-15
[19] Mjagotin A V, Ivanov V I and Ivanova G D 2017 Proc. SPIE 10176 101761Z.
[20] Polyakov P, Luetttmer-Strathmann J and Wiegand S 2006 Journal of Physical Chemistry B 110 26215-26224