The concentration mechanisms of cubic nonlinearity in dispersive media

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Abstract. The comparative analysis of the dynamic holograms recording efficiency in media with non-resonance mechanisms of optical nonlinearity is carried out. It is showed that the greatest values of this parameter are provided by thermodiffusion and electrostrictive mechanisms of concentration nonlinearity of liquid dispersed media.

1. Introduction
There are known many nonlinear mechanisms which are used to record dynamic holograms [1-5]. The highest values of the nonlinearity provide the resonance mechanisms - the saturation of absorption in gases, the saturation of interband transitions in semiconductors. However these nonlinearity mechanisms are limited by narrow spectral range. A non-resonant nonlinearity is free from this drawback and more promising in wide spectral range applications. The mechanisms of this type provide a sufficiently high efficiency of nonlinear interaction of light in both the visible and IR spectral ranges [1, 2].

In this paper we analyze a number of such mechanisms (and sensitive materials) in order to compare the limit characteristics of the dynamic holograms recording and identify the most promising materials for holography with the low-power laser sources.

2. The energy holographic sensitivity
It is customary to compare the different media and mechanisms of nonlinearity using a third-order nonlinear susceptibility, which characterizes the cubic nonlinear response. In the case of non-resonant non-linearity (for weakly absorbing media) is used often the other parameter - \( n_2^{\text{eff}} \, [m^2/W] \), which characterizes the change of the refractive index of the medium under the influence of incident radiation:

\[
\frac{1}{n} = \frac{1}{n_0} + n_2^{\text{eff}} I,
\]

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. However this option does not allow a valid comparison of different media and mechanisms of nonlinearity. In most cases (and especially for thermal non-linearity) this coefficient is proportional to the magnitude of quasi-stationary relaxation time of the nonlinear response \( \tau \), i.e. it depends on the geometry of the system and experimental conditions. The diffraction efficiency of the hologram also depends on its thickness, which is limited by absorption or scattering losses, and the wavelength of the radiation.

Therefore we suggest the more informative complex parameter – the energy holographic sensitivity to assess the effectiveness of the recording of dynamic holograms [2]:

\[
N_{2E} = 2\pi n_2^{\text{eff}} (\pi \alpha \lambda)^{-1} [m^2/J],
\]
where \( \alpha \) - the absorption coefficient of the medium, \( \lambda \) - light wavelength. For phase holograms the value of this parameter corresponds to the minimum energy of the recording radiation needed to change the optical thickness of the medium on the radiation wavelength and completely characterizes the diffraction efficiency of a thin hologram. Moreover this parameter dimension allows to compare the sensitivity of both volume and surface nonlinearity mechanisms \[1\]. For example, for reflection holograms the energy holographic sensitivity can be defined as:

\[ N_{2E} = (d\rho / dl)\tau^{-1}, \]

(3)

where \( \rho \) - Fresnel amplitude reflection coefficient of the boundary between two media. Similarly, the sensitivity is determined for the amplitude thin holograms – in this case the amplitude reflection coefficient is replaced by the corresponding transmittance coefficient.

The most universal non-resonant nonlinearity that exists in any absorbing medium is the heat mechanism. Let us find the value of such sensitivity for a single component material. A simplest mechanism of thermal non-linearity is due to the thermal expansion of the medium. The expression for the coefficient of effective cubic nonlinearity can be obtained by solving the one-dimensional heat equation:

\[ c_p c_T / \partial t = - \text{div} J_1 + \alpha J_1 (1 + \sin Kx), \]

(4)

where \( J_1 = -D_{11} \text{grad} T \) - heat flow, \( T \) - temperature of the material, \( D_{11} \) - the thermal conductivity of the material, \( c_p \) and \( \rho \) - the specific heat capacity and density of the medium, \( I = I_0 (1 + \sin Kx) \) - the intensity spatial distribution along the layer of the medium (x-axis), \( K \) - the space wave vector of the elementary hologram. Using the stationary solution (3) for the amplitude of thermal lattice, we obtain [2]:

\[ n_{2e}^\text{eff} = \alpha \tau_r (dn/dT)(c_p \rho)^{-1}, \]

(5)

where \((dn/dT)\) - the temperature coefficient of the refractive index, \( \tau_r = D_{11}/K^2 \) - the thermal relaxation time. The holographic sensitivity parameter is obtained from (1):

\[ N_{2E} = 2\pi (dn/dT)(c_p \rho \lambda)^{-1}. \]

(6)

Thus, the value of sensitivity for thermal nonlinearity of single-component fluids is defined by parameters \((dn/dT)\), \( c_p \) and \( \rho \). Substituting the characteristic parameters for semiconductor media \(dn/dT = 2 \cdot 10^{-4} K^4\), \( (c_p \rho) \approx 1 J/(cm^3 K)\), we obtain \( N_{2E} \approx 1 cm^2/J \) (for radiation with a wavelength of \( \lambda = 10 \mu m \)).

However a light with an intensity of several \( mW/cm^2 \) is required the greater value of the coefficient of nonlinearity for effective dynamic hologram recording. Since the relaxation time (e.g., time diffusion concentration) increases with particle size increasing, it is natural to expect high nonlinearity parameters in nanodispersive media (colloids, suspensions, micro-emulsions) the particle size of which is limited by the wavelength of the radiation due to light scattering. The “nanodispersive” trend can be traced quite easily in the searching of large non-linearities. If in the first papers was used the nonlinear medium with a typical value \( n_{2e}^\text{eff} \approx 10^{-13} cm^2/W \) (molecular Kerr nonlinearity), than was experimentally obtained values \( n_{2e}^\text{eff} \approx 10^{-8} cm^2/W \) (artificial Kerr-like nonlinearity) \[3\].
3. The thermodiffusion and electrostrictive mechanisms of concentration nonlinearity

Multicomponent nanoparticulate medium (liquid phase mixture, suspension, emulsion) are characterized by a series of specific mechanisms nonlinearities that are not in single-component media. In particular, these include the concentration of non-linearity caused by the redistribution 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.

A thermally induced mechanism of particle drift in a nonuniform temperature field is known as thermal diffusion (thermophoresis) in gases or Soret effect in liquid-phase binary mixtures [4, 5]. The microheterogeneous medium with different refractive index of components in the electromagnetic field is forced by electrostriction forces, which can also be the cause of concentration streams [2]. Depending on the sign of the polarizability the microparticles 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.

Consider setting holographic sensitivity of the concentration non-linearity. In nanodispersive medium particle radius is much smaller than the radiation wavelength \( \lambda \), the refractive index of the medium is proportional to the concentration of particles (for dilute systems):

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

where and \( \delta = (n_2 - n_1)/n_1 \); \( n_1 \) and \( n_2 \) - the refractive indices of the substance and the dispersion medium of the dispersed phase, respectively; \( \phi = (4/3)\pi r^3 C \) - the volume fraction of the dispersed phase, \( r \) - the radius of the microparticle, \( C \) - the concentration of nanoparticles. Then we have for each concentration mechanism of nonlinearity:

\[
n_{eff}^2 = (d/dC)(dC/dI).
\]

Let’s consider an expression for the holographic sensitivity of the dispersion medium, taking into account both (thermodiffusion and electrostriction) concentration mechanisms. The system of balance equations for the concentration of particles and heat flow for this case is written as follows [2]:

\[
c_p \rho dT/dt = -\text{div} J_1 + \alpha I_0 (1 + \sin Kx), \quad (9)
\]

\[
dC/dt = \text{div}(J_2 + J_3). \quad (10)
\]

Here \( J_1 \) and \( (J_2 + J_3) \) - thermal and concentration flows, respectively:

\[
J_1 = -D_{11} \nabla T - D_{12} \nabla C, \quad (11)
\]

\[
J_2 = -D_{21} C \nabla T - D_{22} \nabla C, \quad (12)
\]

\[
J_3 = \gamma C \nabla I, \quad (13)
\]

where \( D_{22} \) - diffusion coefficient of particles, \( D_{21} \) and \( D_{12} \) - coefficients describing cross-flows (the effect of thermal diffusion and Dufour-effect, respectively), \( J_3 \) - electrostrictive flow, \( \gamma = (2\beta b/c n_1) \) (\( \beta \) and \( b \) - polarizability and mobility of microparticle, respectively), \( c \) - the light velocity[2]. Assuming a one-dimensional problem, we seek the solution of equations (9-13) in the form of:
\[ C( x, t ) = C_0 + C_1(t) \sin Kx, \]  
(14)  
\[ T(x, t) = T_0(t) + T_1(t) \sin Kx. \]  
(15)  
Here \( C_0 \) and \( T_0 \) are the average particle concentration and temperature of the medium. Let’s the amplitudes of the thermal \( T_1 \) and concentration \( C_1 \) profiles are small - (\( C_1/C_0 \) << 1, \( T_1/T_0 \) << 1). The linearized system is easily solved and we have by taking into account (2) and (9-15):

\[ \mathcal{N}_{2E} = 2m_1\phi_0\delta[D_{11}D_{21}(c_{\mu}\rho)^{1/3} - \gamma\alpha^{1/4}K^2]^{1/4}. \]  
(16)  

The sensitivities of the concentration nonlinearity mechanisms of liquid-phase two-component medium are determined by the coefficient of thermal diffusion \( D_{21} \) (1-st term) and by microparticle polarizability in the case of the electrostrictive mechanism (2-nd term). For a non-absorbing nanodispersive medium the absorption coefficient should be replaced by the extinction coefficient. As can be seen from (16), both mechanisms may either strengthen or weaken each other depending on the sign of the thermal diffusion coefficient and polarizability of the dispersed particles. The evaluations from (6) and results of experimental studies [4-6] shows the value \( \mathcal{N}_{2E} \approx 10^2 \text{ cm}^2 / \text{J} \) for both concentration nonlinear holographic sensitivity in the vicinity of the critical point of the micro-emulsion.

4. Conclusions

Thus, our analysis shows that the nanoparticulate liquid phases are characterized by high values of the holographic sensitivity. More over the dispersion media have another advantage. It is known that the main parameters of the nonlinear medium are usually interrelated: for example, the value of nonlinearity usually corresponds to poor temporal resolution, spatial resolution and sensitivity of the holographic media are inversely related. Because the parameters of known materials form a discrete set, there is a problem of selection medium with optimized characteristics for particular applications. The most promising materials in this sense are also the nano-materials, because one can change the volume fraction of the various components and their composition, i.e. control (including in real time) of parameters.

Since there are known same mechanisms of nonlinearity in the nano-dispersive systems with the largest holographic sensitivity we can distinguish them as a promising new class of media for dynamic holography [2].

5. References

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