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FOUR-WAVE MIXING IN A LIQUID SUSPENSION OF DIELECTRIC MICROSPHERES

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In this work a theory of the degenerate four-photon parametric scattering (FPS) is developed in a liquid suspension of transparent microspheres (heterogeneous medium), the nonlinearity of which is caused by the change in the microsphere concentration under the action of gradient forces in the electromagnetic field of interacting waves. It is shown that the water suspension of latex spheres with diameter $d = 0.234$ μm and concentration $N_0 = 6.5{\cdot}10^{10}$ cm^{-3}, in effect of the FPS process, corresponds to a cubic nonlinear medium with the optical Kerr coefficient n_2 that is larger by a factor of 10^5 than in the case of CS_2.

Keywords: gradient forces, four-wave mixing, nonlinear medium, transparent dielectric microspheres

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1. Introduction

The nonlinearity of a liquid suspension of transparent microspheres (heterogeneous medium) is explained by the fact that their concentration changes under the action of gradient forces in an electromagnetic field of interacting waves. It is known that the gradient forces arising in a liquid suspension of microspheres under the action of a laser radiation interference field draw microspheres with a large refractive index $n_0 > \bar{n}$ (where n_0 and $\bar{n}$ are refractive indices of the microspheres and the liquid, respectively) into the region with a maximum radiation intensity (interference field antinode). The increasing concentration of microspheres in the region with higher radiation intensity leads to an increase in the refractive index of the suspension and, accordingly, to a corresponding decrease in this index in the region with a lower radiation intensity (interference field nodes). In the case where $n_0 < \bar{n}$ the gradient forces draw microspheres into the region with a lower intensity and, by doing so, also increase the refractive index of the suspension in the region with higher radiation intensity. Thus, independently of ratio $M = n_0/\bar{n}$, a liquid suspension of transparent microspheres – a heterogeneous medium formed artificially – behaves like a nonlinear self-forming medium with a positive optical Kerr coefficient $n_2 > 0$ [1]. The possibility of using such heterogeneous medium as a nonlinear optical material was noted for the first time in [2].

The concentration nonlinearity of a heterogeneous medium, which is due to the spatial modulation of relatively large particles (microspheres) in the viscous liquid, is characterized by a markedly longer time of its establishment as compared to the nonlinearity of ordinary "atomic" media [3]. Since microspheres have large sizes ($\sim$ μm), their spatial modulation by gradient forces results in the appearance of unusually large nonlinear coefficient [1].

Below is the theory, developed by us, of four-wave mixing (FWM) which is based on a joint system of reduced wave equations and a two-dimensional Smoluchowski equation for the concentration of microspheres, the solution of which is presented in the form of Fourier series with time-dependent amplitudes of concentration gratings with multiple periods induced by interacting waves. The gradient forces arising in an interference field of co- and counter-propagating waves were calculated in the Rayleigh–Hans approximation. The effect of suppression of four-wave mixing because of the reduction of the resulting components of the gradient forces to zero for microspheres of certain sizes and certain angles of convergence of interacting waves has been predicted. A stationary regime of four-wave mixing has been analyzed in the diffusion

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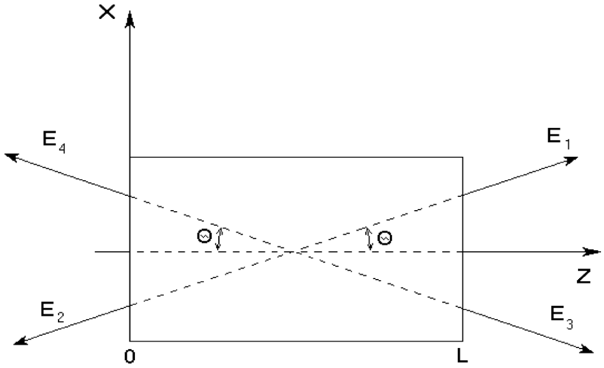

Fig. 1. Geometry of four-wave mixing: 2Θ is the angle of convergence of the interacting waves, L is the length of cell with suspension.

limit and the conditions of the appearance of parametric generation of a pair of mutually conjugate waves have been determined, with account for the radiation losses caused by the Rayleigh scattering on microspheres [4].

2. Basic equations

The process of four-wave mixing in a liquid suspension of transparent microspheres will be considered in the Rayleigh–Hans approximation [5]:

$$|m-1| \ll 1 \quad \text{and} \quad 4\pi a|m-1| \ll \lambda\,, \tag{1}$$

where a is the radius of a microsphere, λ is the radiation wavelength. If inequality (1) is fulfilled, the diffraction of radiation on the microspheres can be ignored [5] for a given acting field, i. e., the so-called electrostatic approximation can be used [6].

Let us represent the acting electromagnetic field in the form of a sum of linearly polarized plane waves with a frequency ω:

$$E = \frac{1}{2}\sum_{l=1}^{4} E_l(z,t)\mathrm{e}^{\mathrm{i}(\omega t - \vec{k}_l\vec{r})} + \text{c. c.}\,, \tag{2}$$

where $E_l(z,t)$ is the wave amplitude, $\vec{k}_l$ is the wave vector (see Fig. 1). We will assume that the interacting waves are polarized in the duration orthogonal to the plane (z,x).

In the case of four-wave mixing, for microspheres the weak radiation forces of the light pressure can be ignored in the theory considered [6, 7]. In this case, a decisive contribution to the process of light-induced formation of the concentration response is made by the gradient forces, the amplitude of which is determined by expression [8]

$$\vec{F}_\nabla = \alpha_0 \int\limits_V \vec{\nabla}|\bar{E}|^2 \mathrm{d}V\,, \tag{3}$$

where

$$\alpha_0 = \frac{3}{4\pi}\bar{n}^2\frac{m^2-1}{m^2+2} \tag{4}$$

is the specific polarizability of a microsphere,

$$|\bar{E}|^2 = \frac{1}{2}\{|E_0|^2 + [E_1E_2^*\mathrm{e}^{\mathrm{i}2(k_z z + k_x x)} + E_3E_4^*\mathrm{e}^{\mathrm{i}2(k_z z - k_x x)} + (E_1E_4^* + E_2^*E_3)\mathrm{e}^{\mathrm{i}2k_z z} + (E_1E_3^* + E_2^*E_4)\mathrm{e}^{\mathrm{i}2k_x x} + \text{c. c.}]\} \tag{5}$$

is the time averaged intensity of the acting radiation, $|E_0|^2 = \sum_{l=1}^4 |E_l|^2$, $V = (4\pi/3)a^3$ is the volume of a microsphere, $k_z = k\cos\Theta$ and $k_x = k\sin\Theta$ are the corresponding projections of the wave vector $\vec{k}$, $k = (\omega/c)\bar{n}$. The integral in (3) in essence is the integral of overlap of a microsphere with the nonuniform field of the acting radiation.

We will assume that the products of the complex conjugate amplitudes of interacting waves are slow functions of the longitudinal coordinate,

$$\frac{\partial|E_lE_{l'}^*|}{\partial z} \ll k_z|E_lE_{l'}^*|\,. \tag{6}$$

In this approximation the integral in (3) can be exactly calculated in spherical coordinates with the use of (5) [9]. Integrating (3), we find the gradient force

$$\vec{F}_\nabla = \hat{\mathbf{j}}F_x + \hat{\mathbf{k}}F_z\,, \tag{7}$$

where $\hat{\mathbf{j}}$ and $\hat{\mathbf{k}}$ are orthogonal unit vectors,

$$F_\kappa = F_{\kappa 0}\mathrm{e}^{\mathrm{i}2(k_z z - k_x x)} + F_{\kappa 1}\mathrm{e}^{\mathrm{i}2(k_z z + k_x x)} + F_{\kappa 2}\mathrm{e}^{\mathrm{i}2k_\kappa \kappa} + \text{c. c.} \tag{8}$$

are the components of the gradient force vector $\vec{F}_\nabla$, $\kappa \in \{x, z\}$. The amplitudes of the harmonics of the F_κ components are determined by the relations

$$\begin{aligned} F_{x0} &= -\mathrm{i}\alpha_0 k_x E_3E_4^* V_0 = -(k_x/k_z)F_{z0}\,,\\ F_{x1} &= \mathrm{i}\alpha_0 k_x E_1E_2^* V_0 = (k_x/k_z)F_{z1}\,,\\ F_{x2} &= \mathrm{i}\alpha_0 k_x (E_1E_3^* + E_2^*E_4)V_x\,,\\ F_{z2} &= \mathrm{i}\alpha_0 k_z (E_1E_4^* + E_2^*E_3)V_z\,, \end{aligned} \tag{9}$$

where

$$\begin{aligned} V_0 &= (a\Lambda_0)^{3/2}J_{3/2}(2\pi a/\Lambda_0)\,,\\ V_\kappa &= (a\Lambda_\kappa)^{3/2}J_{3/2}(2\pi a/\Lambda_\kappa)\,. \end{aligned} \tag{10}$$

Here $J_{3/2}(\xi)$ is the cylindrical Bessel function, $\Lambda_0 = \pi/k$ and $\Lambda_\kappa = \pi/k_\kappa$ are the periods of the interference pattern of the corresponding pairs of waves. The constant coefficients V_0 and V_κ in (9) appear on integration of (3) and account for the nonuniformity of radiation inside the microspheres. It is easy to show that, at $\xi \ll 1$, $V = V_0 = V_\kappa$. It is apparent that $|F_x/F_z| \propto \tan\Theta$ and, consequently, in the region of small angles $\Theta \ll \pi/2$ a dominating part in the formation of the concentration response is played by the longitudinal component (z component) of the gradient force. As it follows from (10), for a certain ratio between the radius a of the microspheres and the period $\Lambda_{0,\kappa}$ of interference pattern, namely, at $2\pi a/\Lambda_{0,\kappa} = \xi_i$ (where ξ_i are roots of the cylindrical Bessel function $J_{3/2}(\xi)$, $i = 1, 2, \ldots$), independently of their position, the corresponding component of the gradient force is equal to zero.

The so-called "zero-force" effect is due to the equal action of its oppositely directed components acting on the corresponding elements of a microsphere. This effect has been theoretically predicted independently in [6, 9]. It follows from (10) that at certain values of parameters of the system (for example, at $2k_x a = \xi_1$, $2k_z a = \xi_2$) the components of gradient forces F_{x2} are equal to zero (i. e. $V_\kappa = 0$) independently of the intensity of the acting radiation.

Using the values of the first two roots of the Bessel function $J_{3/2}(\xi)$ [10], it can be shown that the condition $V_\kappa = 0$ is fulfilled at $a/\lambda = 0.709$ and $\tan\Theta = (\xi_1/\xi_2) \approx 0.58 \Rightarrow \Theta \approx 30^\circ$ in suspensions with a small relative refractive index, $|m - 1| \leqslant 10^{-2}$.

Thus, because of the "zero-force" effect, the four-wave mixing caused by a concentration nonlinearity can be practically completely suppressed at certain sizes of the microspheres and certain angles of convergence of interacting waves. Note that, depending on the radius a of the microspheres, the amplitudes of the gradient forces (9) are alternating functions. Therefore, spheres of certain sizes can be drawn into antinodes (at $J_{3/2}(\xi) > 0$) or into nodes (at $J_{3/2}(\xi) < 0$) of the interference pattern of the field. This movement of the microspheres is explained physically by their tending to overlap with a maximum number of antinodes [6, 9].

To determine the concentration response of microspheres, induced by the electromagnetic field (1), we will use the two-dimensional Smoluchowski equation (see, for example, [11]):

$$\frac{\partial N}{\partial t} = D\Delta_\perp N - b\bigg\{N\left(\frac{\partial F_x}{\partial x} + \frac{\partial F_z}{\partial z}\right) + \left(F_x\frac{\partial N}{\partial x} + F_z\frac{\partial N}{\partial z}\right)\bigg\}, \tag{11}$$

where N is the concentration of the microspheres [cm^{-3}], $D = k_\mathrm{B}T/(6\pi\nu a)$ is the diffusion coefficient [cm^2/s], k_B is the Boltzmann constant, T is the temperature, ν is the viscosity of the liquid, $b = D/(k_\mathrm{B}T)$ is the mobility of microspheres, $\Delta_\perp = \partial^2/\partial x^2 + \partial^2/\partial z^2$. Equation (11) is valid in the region $t \gg t^*$ under the condition that the gradient force $\vec{F}_\nabla$ is a slow function on the time and space scales t^* and l^* determined by the relations [11, 12]

$$t^* = bm \quad \text{and} \quad l^* = \sqrt{\frac{k_\mathrm{B}T}{m}}t^*, \tag{12}$$

where m is the mass of microsphere. In particular, $t^* \simeq 3\cdot 10^{-9}$ s and $l^* \simeq 7\cdot 10^{-9}$ cm for an aquatic suspension of latex microspheres of radius $a = 1.17\cdot 10^{-5}$ cm with the density of 1 g/cm^3 at room temperature [1].

Equation (11) is conveniently solved in the form of the harmonic series

$$N(x, z, t) = \sum_{m,n=-\infty}^{\infty} N_{mn}(t)\mathrm{e}^{\mathrm{i}2(mk_z z + nk_x x)}, \tag{13}$$

where $N_{00} = \langle N\rangle_{x,z} = N_0 = \mathrm{const}$ is the initial concentration of the microspheres, $\langle \ldots \rangle_{x,z}$ means spatial averaging, and $N^*_{mn} = N_{-m,-n}$. Substitution of (13) into (11), in view of (8), gives the following system of kinetic equations for the amplitudes $N_{mn}(t)$ of the concentration harmonics:

$$\begin{aligned}
&\left(\frac{\partial}{\partial t} + W_{mn}\right)N_{mn} \\
&= a_{mn}N_{m-1,n+1} - a^*_{mn}N_{m+1,n-1} \\
&+ b_{mn}N_{m-1,n-1} - b^*_{mn}N_{m+1,n+1} + c_n N_{m,n-1} \\
&- c^*_n N_{m,n+1} + d_m N_{m-1,n} - d^*_m N_{m+1,n}\,,
\end{aligned} \tag{14}$$

where

$$\begin{aligned}
W_{mn} &= 4D[(mk_z)^2 + (nk_x)^2]\,, \\
a_{mn} &= 2\alpha_0 b(mk_z^2 - nk_x^2)E_3E_4^*V_0\,, \\
b_{mn} &= 2\alpha_0 b(mk_z^2 + nk_x^2)E_1E_2^*V_0\,, \\
c_n &= 2\alpha_0 bnk_x^2(E_1E_3^* + E_2^*E_4)V_x\,, \\
d_m &= 2\alpha_0 bmk_z^2(E_1E_4^* + E_2^*E_3)V_z\,.
\end{aligned} \tag{15}$$

In the case where the radii of the microspheres $a \ll \Lambda_0$, the convergence angles $\Theta \ll \pi/2$ are small, and the gradient force component F_x can be ignored, the system of equations (14) takes the simpler form

$$\left(\frac{\partial}{\partial t} + W_{mn}\right) N_{mn} = 2\alpha_0 bVmk_z^2 \times [E_3E_4^*N_{m-1,n+1} - E_3^*E_4N_{m+1,n-1} + E_1E_2^*N_{m-1,n-1} - E_1^*E_2N_{m+1,n+1} + (E_1E_4^* + E_2^*E_3)N_{m-1,n} - (E_1^*E_4 + E_2E_3^*)N_{m+1,n}] . \quad (16)$$

The nonlinear polarization of the suspension of microspheres caused by a change in their concentration under the action of gradient forces is determined by relation [6]

$$P = \frac{1}{2}\alpha_0 VN(x,z,t)\sum_{l=1}^{4} E_l(z,t)\mathrm{e}^{-\mathrm{i}(\omega t - \vec{k}_l\vec{r})} + \text{c. c.} \quad (17)$$

Substitution of (2) and (17) into the wave equation gives the following system of reduced equations for the amplitudes of interacting waves:

$$\cos\Theta\frac{\partial E_1}{\partial z} + \frac{1}{v}\frac{\partial E_1}{\partial t} = -\rho E_1 + \mathrm{i}\gamma(E_1 + \chi_{11}E_2 + \chi_{01}E_3 + \chi_{10}E_4)\,, \quad (18a)$$

$$-\cos\Theta\frac{\partial E_2}{\partial z} + \frac{1}{v}\frac{\partial E_2}{\partial t} = -\rho E_2 + \mathrm{i}\gamma(E_2 + \chi_{11}^*E_1 + \chi_{10}^*E_3 + \chi_{01}^*E_4)\,, \quad (18b)$$

$$\cos\Theta\frac{\partial E_3}{\partial z} + \frac{1}{v}\frac{\partial E_3}{\partial t} = -\rho E_3 + \mathrm{i}\gamma(E_3 + \chi_{01}^*E_1 + \chi_{10}E_2 + \chi_{1,-1}E_4)\,, \quad (18c)$$

$$-\cos\Theta\frac{\partial E_4}{\partial z} + \frac{1}{v}\frac{\partial E_4}{\partial t} = -\rho E_4 + \mathrm{i}\gamma(E_4 + \chi_{10}^*E_1 + \chi_{01}E_2 + \chi_{1,-1}^*E_3)\,, \quad (18d)$$

where v is the velocity of light in the suspension, $\gamma = 2\pi(k/\bar{n}^2)\alpha_0 VN_0$, $\chi_{mn} = N_{mn}/N_0$. In (18) the relation $N_{mn}^* = N_{-m,-n}$ is taken into account and the factor of amplitude losses caused by the Rayleigh scattering on the microspheres is phenomenologically introduced :

$$\rho = \frac{8\pi}{3}N_0k^4\left(\frac{m^2-1}{m^2+2}\right)^2 a^6\,. \quad (19)$$

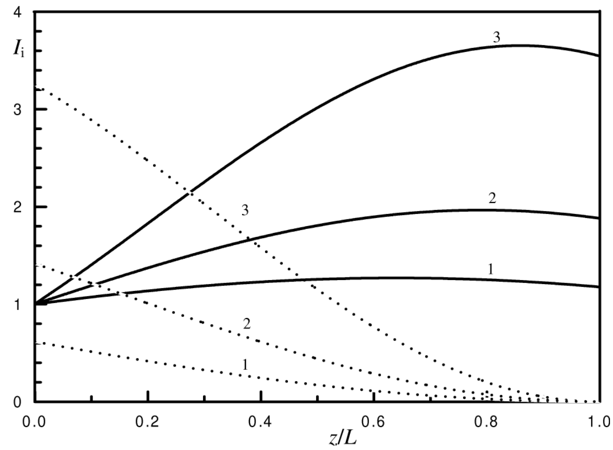

Fig. 2. Stationary distribution of normalized intensities of weak waves $I_i = |E_i|^2/|E_{30}|^2$ ($i = 3, 4$) for different values of intensity of the reference waves at $m = 1.195$, $a = 1.25{\cdot}10^{-5}$ cm, $N_0 = 6.5{\cdot}10^{-5}$ cm^{-3}, $k = 1.6{\cdot}10^5$ cm^{-1}, $T = 300$ K, $\Theta = 3.2°$, $L = 10^{-2}$ cm (I_3 are solid curves, I_4 are dotted curves): for *1* curves $2\alpha_0 VN_0/(k_\mathrm{B}T)|E_0|^2 = 3.1{\cdot}10^2$, for *2* curves it is $4.9{\cdot}10^2$, for *3* curves it is $7.0{\cdot}10^2$.

It follows from (18) that, of all the excited concentration gratings (13), two pairs of gratings participate directly in the four-wave mixing: one pair of orthogonal gratings $N_\kappa \propto \cos 2k_\kappa\kappa$ ($\kappa = x, z$) leads to a parametric connection and an energy exchange between the waves, and the other pair $N_\pm \propto \cos 2(k_z z \pm k_x x)$ leads to the self-action effects. Because of the spatial averaging of the wave equations, the other concentration gratings do not contribute directly to the FWM process. Their role reduces to the influence on the amplitudes of the main gratings, N_κ and $N_\pm$.

The joint system of wave (18) and kinetic equation (14) with the corresponding boundary

$$E_1(0,t) = E_{10}(t)\,, \quad E_2(L,t) = E_{20}(t)\,,$$
$$E_3(0,t) = E_{30}(t)\,, \quad E_4(L,t) = 0 \quad (20)$$

and initial

$$N_{mn}(t=-\infty) = 0\,, \text{ where } m \neq 0,\ n \neq 0,$$
$$N_{00}(t=-\infty) = N_0 \quad (21)$$

conditions describes the four-wave mixing of waves of arbitrary intensity which is due to the concentration nonlinearity caused by the action of gradient forces on the transparent microspheres. Figure 2 illustrates the results of the numerical solution of the joint system of Eqs. (18) and (14) in the established regime for input pulses (20) of rectangular form at $|E_{j0}|^2 = |E_0|^2 \gg |E_{30}|^2$.

3. Theory of four-wave mixing in the diffusion limit

The diffusion approximation [4]

$$N(z,x,t) = N_0 + \bar{N}(z,x,t)\,,\ |\bar{N}| \ll N_0\,, \quad (22)$$

imposes a restriction on the intensity of the interacting waves and allows the terms proportional to the amplitudes of the concentration gratings N_{mn} $(m,n \neq 0)$ on the right-hand side of (14) to be neglected. In this case, from (14) we obtain the following equations for the components χ_{mn} determining the FWM process:

$$\left(\frac{\partial}{\partial t} + 4Dk_x^2\right)\chi_{01} = 2\alpha_0 V_x b k_x^2 (E_1E_3^* + E_2^*E_4)\,, \quad (23a)$$

$$\left(\frac{\partial}{\partial t} + 4Dk_z^2\right)\chi_{10} = 2\alpha_0 V_z b k_z^2 (E_1E_4^* + E_2^*E_3)\,, \quad (23b)$$

$$\left(\frac{\partial}{\partial t} + 4Dk^2\right)\chi_{11} = 2\alpha_0 V_0 b k^2 E_1E_2^*\,, \quad (23c)$$

$$\left(\frac{\partial}{\partial t} + 4Dk^2\right)\chi_{1,-1} = 2\alpha_0 V_0 b k^2 E_3E_4^*\,. \quad (23d)$$

Evidently, in the diffusion limit, the times of establishment of concentration gratings $\tau_0 = 1/(4Dk^2) = (\Lambda_0/2\pi)^2/D$ and $\tau_\kappa = 1/(4Dk_\kappa^2) = (\Lambda_\kappa/2\pi)^2/D$ are independent of the radiation intensity and are determined by the diffusion coefficient and the periods of diffractional pattern formed by the interacting waves. Note that for microspheres having small radii (at $V_0 = V_\kappa = V$) the stationary values of the amplitudes of the concentration gratings are independent of the angle of convergence of the interacting waves.

In the stationary regime at $t \gg \tau_\kappa$, the Eqs. (18) in view of (23) take the form

$$\cos\Theta\frac{\partial E_1}{\partial z} = -\rho E_1 + \mathrm{i}\gamma\left\{\frac{a_0 b}{2D}(V_x+V_z)E_2^*E_3E_4 + \left[1 + \frac{a_0 b}{2D}(V_0U_2 + V_xU_3 + V_zU_4)\right]E_1\right\}, \quad (24a)$$

$$-\cos\Theta\frac{\partial E_2}{\partial z} = -\rho E_2 + \mathrm{i}\gamma\left\{\frac{a_0 b}{2D}(V_x+V_z)E_1^*E_3E_4 + \left[1 + \frac{a_0 b}{2D}(V_0U_1 + V_zU_3 + V_xU_4)\right]E_2\right\}, \quad (24b)$$

$$\cos\Theta\frac{\partial E_3}{\partial z} = -\rho E_3 + \mathrm{i}\gamma\left\{\frac{a_0 b}{2D}(V_x+V_z)E_4^*E_1E_2 + \left[1 + \frac{a_0 b}{2D}(V_xU_1 + V_zU_2 + V_0U_4)\right]E_3\right\}, \quad (24c)$$

$$-\cos\Theta\frac{\partial E_4}{\partial z} = -\rho E_4 + \mathrm{i}\gamma\left\{\frac{a_0 b}{2D}(V_x+V_z)E_3^*E_1E_2 + \left[1 + \frac{a_0 b}{2D}(V_zU_1 + V_xU_2 + V_0U_3)\right]E_4\right\}, \quad (24d)$$

where $U_l = |E_l|^2$. Ignoring the influence of the weak waves E_3 and E_4 on the powerful reference waves E_1 and E_2, from (24) at $\rho = 0$ we find

$$\cos\Theta\frac{\partial E_3}{\partial z} = \mathrm{i}\left\{\bar{\kappa}_3E_3 + \beta E_4^*E_{10}E_{20}\mathrm{e}^{\mathrm{i}\bar{\kappa}_2\hat{L} + \mathrm{i}(\bar{\kappa}_1 - \bar{\kappa}_2)\hat{z}}\right\}, \quad (25a)$$

$$-\cos\Theta\frac{\partial E_4^*}{\partial z} = \mathrm{i}\left\{\bar{\kappa}_4E_4 + \beta E_3E_{10}^*E_{20}^*\mathrm{e}^{-\mathrm{i}\bar{\kappa}_2\hat{L} + \mathrm{i}(\bar{\kappa}_2 - \bar{\kappa}_1)\hat{z}}\right\}, \quad (25b)$$

where

$$\bar{\kappa}_1 = \gamma\left(1 + \frac{a_0 b}{2D}V_0U_2\right), \quad \bar{\kappa}_2 = \gamma\left(1 + \frac{a_0 b}{2D}V_0U_1\right), \quad (26a)$$

$$\bar{\kappa}_3 = \gamma\left[1 + \frac{a_0 b}{2D}(V_xU_1 + V_zU_2)\right], \quad \bar{\kappa}_4 = \gamma\left[1 + \frac{a_0 b}{2D}(V_zU_1 + V_xU_2)\right], \quad (26b)$$

$\beta = \gamma a_0 b/(2D)(V_x + V_z)$ is the parametric coupling coefficient $\hat{z} = z/\cos\Theta$, $\hat{L} = L/\cos\Theta$. The solution of (25) with the boundary conditions $E_3(0) = E_{30}$ and $E_4^*(\hat{L}) = 0$ has the form

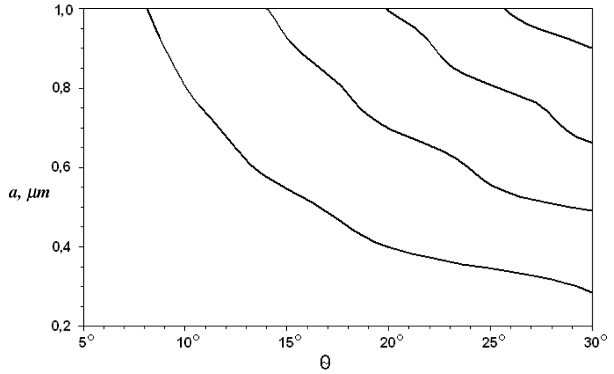

Fig. 3. Series of solutions of the equation $\beta(a, \Theta) = 0$ at $k = 1.6{\cdot}10^5$ cm^{-1}.

$$E_3(z) = E_{30} \cdot \mathrm{e}^{\mathrm{i}(\Delta/2+\bar{\kappa}_3)\hat{z}} \times \frac{2\Gamma\cos[\Gamma\cdot(\hat{L}-\hat{z})] + \mathrm{i}\Delta\sin[\Gamma\cdot(\hat{L}-\hat{z})]}{2\Gamma\cos[\Gamma\cdot\hat{L}] + \mathrm{i}\Delta\sin[\Gamma\cdot\hat{L}]}, \tag{27a}$$

$$E_4^*(z) = \mathrm{i}2\beta E_{30}\sqrt{U_1U_2} \cdot \mathrm{e}^{-\mathrm{i}(\Delta/2-\bar{\kappa}_4)\hat{z}+\bar{\kappa}_2\hat{L}} \times \frac{\sin[\Gamma\cdot(\hat{L}-\hat{z})]}{2\Gamma\cos[\Gamma\cdot\hat{L}] + \mathrm{i}\Delta\sin[\Gamma\cdot\hat{L}]}, \tag{27b}$$

where

$$\Gamma = \sqrt{\beta^2 U_1 U_2 + \Delta^2/4} \tag{28}$$

is the parametric gain increment of weak radiation,

$$\Delta = (\bar{\kappa}_1 - \bar{\kappa}_2 - \bar{\kappa}_3 + \bar{\kappa}_4) = \gamma\frac{a_0 b}{2D}(V_0 + V_x - V_z)\cdot(U_2 - U_1) \tag{29}$$

is the phase mismatch of the interacting waves.

Clearly, the amplitude of the inverse wave E_4 is proportional to the coefficient β of parametric coupling. Using relation (10), it can be shown that the coefficient β reduces to zero at certain values of the microsphere radius and the interacting wave convergence angle Θ. At these values of the parameters a and Θ, because of "zero-force" effect, the concentration gratings with period Λ_κ that are responsible for the four-wave mixing are not excited, and so the parametric generation of the inverse wave E_4 does not occur. Figure 3 shows a series of curves on the plane (a, Θ) that demonstrate the values of a and Θ at which the parametric coupling coefficient β is equal to zero.

It follows from (27) that at $\Delta = 0$, $\Gamma = \Gamma_0 = |\beta|\sqrt{U_1U_2}$ the linear FWM theory imposes a restriction on the intensities of reference waves because of the possibility of parametric generation of a pair of mutually conjugate waves E_3 and E_4^*, the threshold of which is determined from condition [13]

$$\frac{\Gamma_0 L}{\cos\Theta} = \frac{\pi}{2}\,. \tag{30}$$

In view of the definition of the coupling coefficient β, we obtain from (30)

$$2kN_0\frac{a_0^2 b}{\bar{n}^2 D}V\,|V_x + V_z|\sqrt{U_1U_2}\frac{L}{\cos\Theta} = 1\,. \tag{31}$$

It follows from (29) that the condition $\Delta = 0$ is fulfilled in two cases, where either $U_1 = U_2$ or $(V_0 + V_x - V_z) \equiv \bar{V} = 0$. Note that for microspheres of small sizes, for which $a \ll \Lambda_0$ and $V_0 = V_\kappa = V$, the condition $\Delta = 0$ is fulfilled only at $U_j = U$, $j = 1, 2$, as in the case of ordinary media with a Kerr nonlinearity [13].

Figure 4 shows the dependence of the parameter $\bar{V}$ on the angle Θ for different values of the coefficient $\xi = 2ka$. It is seen from Fig. 4 that the parameter $\bar{V}(\Theta)$ can be equal to zero in the region $\xi \geqslant 2.5$. The angle $\Theta = \pi/2$ corresponds to the threshold value of the coefficient $\xi = \xi_{\mathrm{thr}} \approx 2.5$, at which $\bar{V} = 0$. At $\xi > 2.5$ the corresponding values of the angle Θ can be much smaller. Consequently, in the region $\xi > 2.5$ ($a > 0.2\lambda$) the conditions of parametric generation of a weak radiation at non-equal intensities of the reference waves ($U_1 \neq U_2$) can be realized at corresponding values of the convergence angle Θ. Note that at relatively large angles (18) should contain derivatives with respect to the transverse coordinate ($\sin\Theta\partial/\partial x$).

It can be shown that account of the linear losses ($\rho \neq 0$) in the case where $\bar{V} = 0$ leads to the following relation determining the threshold of parametric generation of weak waves:

$$\tan(\hat{\Gamma}\cdot\hat{L}) = -\frac{\hat{\Gamma}}{\rho}\,, \tag{32}$$

where $\hat{\Gamma} = \sqrt{\beta^2 U_1 U_2 e^{-2\rho\hat{L}} - \rho^2} > 0$.

Figure 5 shows dependence of the threshold value of $\Gamma_0 = |\beta|\sqrt{U_1U_2}$ on the length $\hat{L}$ for different values of the amplitude loss factor ρ. It is seen from Fig. 5 that at $\rho \neq 0$ the optimum value of the length $\hat{L}$ is equal to $(\rho\hat{L})_{\mathrm{opt}} = 0.74$. For comparison, we note that approximate analytical estimations give the value of $(\rho\hat{L})_{\mathrm{opt}} = 0.69$ for an ordinary medium with a cubic nonlinearity [14]. Comparison of solution (27) with the ones obtained for a medium with the phase cubic nonlinearity shows that the optical Kerr coefficient is $n_2 \approx 4{\cdot}10^{-3}$ cm^2/MW for an aquatic suspension of latex microspheres ($m = 1.195$, $N = 6.5{\cdot}10^{10}$ cm^{-3}, and $a = 0.117\,\mu$m) at the wavelength $\lambda = 5145$ Å (an

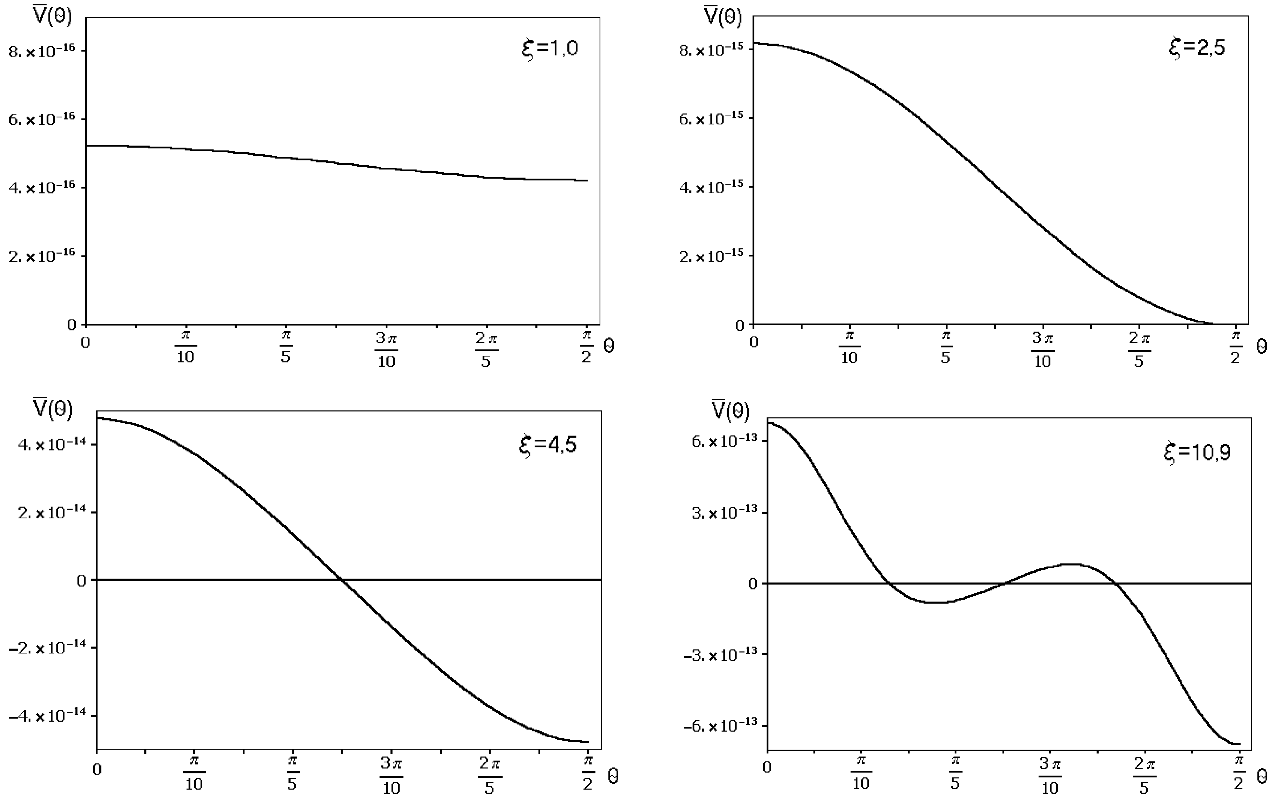

Fig. 4. Dependence of parameter $\bar{V}$ on the angle Θ for different values of the coefficient $\xi = 2ka$ at $k = 10^5$ cm^{-1}.

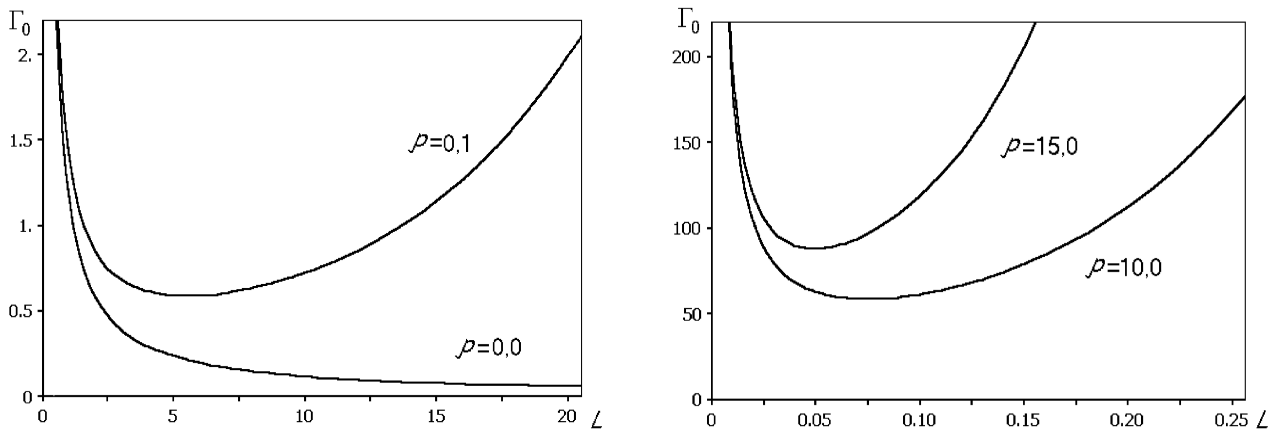

Fig. 5. Dependence of the normalized threshold intensity Γ_0 of the reference waves on the length $\hat{L}$ for different values of the coefficient ρ.

argon laser). The value $n_2 = 3.6{\cdot}10^{-3}$ cm^2/MW [1] that is measured experimentally for the same values of the suspension parameters is close to the theoretical estimate. Note that this value of n_2 is larger by a factor of 10^5 than that for the nonlinear liquid CS_2 [13].

4. Conclusion

In this work, a theory of four-wave mixing in a liquid suspension of transparent microspheres is developed, the nonlinearity of which is due to a change in microsphere concentration under the action of gradient forces in the electromagnetic field of interacting waves. The effect of suppression of four-wave mixing because of the reduction of the resulting components of the gradient forces to zero for microspheres of certain sizes and certain angles of convergence of interacting waves has been predicted. A stationary regime of four-wave mixing has been analyzed in the diffusion limit, and the conditions of the appearance of parametric generation of a pair of mutually conjugate waves have been determined with account for the radiation losses caused by the Rayleigh scattering on microspheres. It is shown that a liquid suspension of dielectric microspheres – an artifically created heterogeneous medium, each component of which does not exhibit nonlinear optical properties – can be used as a highly effective wideband nonlinear medium for the low-intensity laser radiation of long duration.

References

[1] P.W. Smith, A. Ashkin, and W.J. Tomlinson, Four-wave mixing in an artificial Kerr medium, Opt. Lett. **6**(6), 284–286, (1981).

[2] A.J. Palmer, Nonlinear optics in aerosols, Opt. Lett. **5**, 54–56, (1980).

[3] Ya. Yariv, *Kvantovaya Elektronika* (Moscow, Sovetskoe Radio, 1980), 488 p. [in Russian].

[4] D. Rogovin and O. Sari, Phase conjugation in liquid suspensions of microspheres in the diffusive limit, Phys. Rev. A **31**(4), 2375–2389, (1985).

[5] G. van de Hulst, *Scattering of the Light by Small Particles* (Moscow, 1961), 536 p. [in Russian].

[6] P. Zemánek, A. Jonáš, and M. Liška, Simplified description of optical forces acting on a nanoparticle in the Gaussian standing wave, J. Opt. Soc. Am. A **19**(5), 1025–1034, (2002).

[7] S.A. Akhmanov and S.Yu. Nikitin, *Physical Optics* (Moscow, 1998), 656 p. [in Russian].

[8] Rohrbach and E.H.K. Stelzer, Optical trapping of dielectric particles in arbitrary fields, J. Opt. Soc. Am. A **18**(4), 839–853, (2001).

[9] A.A. Afanas'ev, A.N. Rubinov, Yu.A. Kurochkin, S.Yu. Mikhnevich, and I.E. Ermolaev, Localisation of spherical particles under the action of a gradient force in an interference field of laser radiation, Quantum Electron. **33**(3), 250–254 (2003).

[10] G. Korn and T. Korn, *Reference Book on Mathematics* (Moscow, 1973), 831 p. [in Russian].

[11] A.I. Akhiezer and S.V. Peletminskii, *Methods of Statistical Physics* (Moscow, 1977), 368 p. [in Russian].

[12] L.D. Landau and E.M. Livshits, *Electrodyndmics of Continuous Media* (Moscow, 1982), 620 p. [in Russian].

[13] B.Ya. Zel'dovich, N.F. Pilipetskii, and V.V. Shkunov, *Conversion of Wave Front* (Moscow, 1985), 247 p. [in Russian].

[14] B.Ya. Zel'dovich and T.V. Yakovleva, Influence of linear absorption and reflection on the characteristics of the four-wave mixing of reflected waves, Kvant. Elektron. (Moscow) **8**(9), 1891–1898 (1981) [in Russian].

KETURBANGIS MAIŠYMAS SKYSTOJE DIELEKTRINIŲ MIKROSFERŲ SUSPENSIJOJE

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Santrauka

Pateikta teorija, aprašanti išsigimusią keturfotonę parametrinę sklaidą (KPS) skystoje skaidrių mikrosferų suspensijoje (įvairialytėje medžiagoje), kurios netiesiškumą nulemia mikrosferų koncentracijos pokytis, susidaręs veikiant gradientinėms jėgoms sąveikaujančių bangų elektromagnetiniuose laukuose. Parodyta, kad latekso sferų, kurių diametras $d = 0,234$ μm ir koncentracija $N_0 = 6,5{\cdot}10^{10}$ cm^{-3}, vandens suspensijoje vykstančios KPS efektyvumas atitinka kubinę netiesinę terpę, kurios optinis Kero koeficientas n_2 daugiau kaip 10^5 kartų didesnis už atitinkamą CS_2 koeficientą.