

Four-Wave Mixing in a Liquid Suspension of Transparent Dielectric Microspheres

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Abstract—The process of four-wave mixing in an artificial heterogeneous nonlinear medium—a liquid suspension of transparent dielectric microspheres—is considered. The dynamics of the concentration response to gradient forces that act on microspheres in the interference field of interacting waves are investigated on the basis of the Smolukhovskii equation. Kinetic equations for the amplitudes of light-induced concentration gratings that take part in the four-wave mixing are obtained with the use of the Fourier series expansion of the distribution function of microspheres. The ratios of the microsphere radius to the grating periods are obtained under which the resultant gradient force vanishes and, hence, a suspension of dielectric microspheres does not exhibit nonlinear properties irrespective of the intensities of the interacting waves. The kinetics of the process of four-wave mixing is investigated under efficient energy exchange between reference, signal, and reversed waves. It is shown that a liquid suspension of transparent dielectric spheres is a highly effective wideband nonlinear medium for reversing the wave front of low-intensity radiation of continuous-wave lasers. © 2005 Pleiades Publishing, Inc.

1. INTRODUCTION

In this paper, we develop a theory of four-wave mixing in a liquid suspension of transparent microspheres (a heterogeneous medium) whose nonlinearity is associated with the variation of the concentration of microspheres under the action of gradient forces in the electromagnetic field of the interacting waves [1, 2]. It is well known [3] that, in a liquid suspension of microspheres, the gradient forces arising in the interference field of laser radiation draw microspheres with greater refractive index $n_0 > \bar{n}$ (where n_0 and $\bar{n}$ are the refractive indices of the microspheres and the liquid, respectively) into the region of maximal intensity (to the antinodes of the interference field). The increasing concentration of microspheres in the region with higher intensity of radiation leads to an increase in the refractive index of the suspension and to the corresponding decrease in the refractive index in the region of lower intensity (at the nodes of the interference field). In the opposite case, when $n_0 < \bar{n}$, the gradient forces draw microspheres into the region with lower intensity, thus increasing the refractive index of the suspension in the region with higher intensity of radiation. Therefore, irrespective of the ratio $\bar{m} = n_0/\bar{n}$, a liquid suspension of transparent microspheres—an artificial heterogeneous medium—behaves as a nonlinear self-focusing medium with a positive optical Kerr coefficient $n_2 > 0$ [3]. For the first time, a possible application of such heterogeneous structures as a nonlinear optical material was pointed out in [4].

The concentration nonlinearity of a heterogeneous medium associated with the spatial modulation of relatively large particles (microspheres) in a viscous liquid is characterized by much greater relaxation times compared with the nonlinearity of ordinary “atomic” media [5]. Since the size of microspheres is rather large (on the order of a few micrometers), their spatial modulation by gradient forces gives rise to abnormally large nonlinear coefficients. In [3], four-wave mixing experiments were carried out to determine the optical Kerr coefficient n_2 in a water suspension of latex microspheres of radius $a = 0.117 \mu\text{m}$ with the use of argon laser radiation beams ($\lambda = 5145 \text{ \AA}$) with a power of about 100 mW. The optical Kerr coefficient was measured to be $n_2 = 3.6 \times 10^{-3} \text{ cm}^2/\text{MW}$ for the concentration of microspheres $N_0 = 6.5 \times 10^{10} \text{ cm}^{-3}$, which turned out to be greater than the relevant coefficient in CS_2 by a factor of 10^5 . The relaxation times of the concentration gratings of microspheres for a convergence angle of 6.4° between copropagating beams were greater than 100 ms. Due to the high values of the optical Kerr coefficient n_2 , a liquid suspension of dielectric microspheres can be used as a highly effective wideband nonlinear medium for a low-intensity laser impulse of large duration.

In [6], a theory of four-wave mixing in liquid suspensions of small-size transparent microspheres was developed in a weak-field limit, when the saturation phenomena are neglected (the diffusion limit). In the diffusion limit, the gradient forces modulate the concentration of microspheres and produce two orthogonal

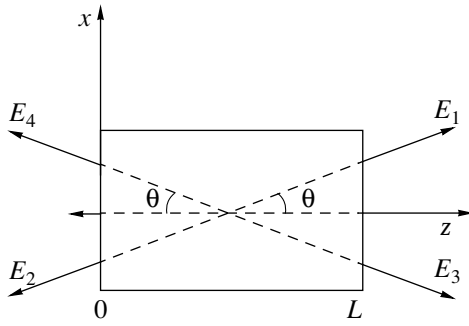


Fig. 1. Geometry of four-wave mixing: 2θ is the convergence angle of waves, and L is the length of the cell with a suspension.

concentration gratings; coherent scattering of radiation by these gratings leads to the formation of a reversed wave and the amplification of the test wave. In spite of the fact that only a small number of microspheres are involved in the four-wave mixing, the reflection coefficient of the reversed wave may reach significant values ($\eta > 1$). A factor that substantially restricts the application of the results of the theory developed in [6] is the approximation of small microspheres; in this approximation, the intensity of effective radiation inside microspheres is assumed to be constant irrespective of the position of the microspheres on the interference pattern of the field. In this case, the steady-state amplitudes of the gratings do not depend on the convergence angle of the interacting waves, and the coefficient of parametric coupling between these waves is proportional to the squared volume of a microsphere, i.e., to a value of a^6 . In particular, we will show below that the theory of four-wave mixing developed in the approximation of constant intensity of radiation inside the microspheres [6] is valid in a very narrow interval of the microsphere radii, gives a considerably overstated value of the reflection coefficient η of the reversed wave in the region of $2ka \geq 1$ (k is the wavenumber), and does not predict the oscillating behavior of the function $\eta = \eta(a)$. In this paper, we develop a four-wave mixing theory that is free of these constraints; the results obtained here include the results of [6] as a particular case.

The theory developed below is based on a simultaneous system of truncated wave equations and the two-dimensional Smolukhovskii equation for the concentration of microspheres; a solution to the latter equation is represented as a Fourier series with time-dependent amplitudes of multiply periodic concentration gratings induced by the interacting waves. The gradient forces that arise in the interference field of co- and counterpropagating waves are calculated in the Rayleigh-Gans approximation. The amplitudes of the gradient forces, which are determined by the overlap integral of a microsphere and a periodically modulated intensity of the effective radiation that takes into account the non-

uniform distribution of this radiation inside a microsphere, are obtained in the explicit form in spherical coordinates. The theory predicts the suppression of four-wave mixing due to the vanishing of the resultant components of gradient forces for certain dimensions of microspheres and the convergence angles of the interacting waves. We investigate the dynamics of the formation and relaxation of concentration gratings that are induced by the interacting light waves. We analyze the steady-state regime of four-wave mixing in the diffusion limit and determine conditions under which a parametric generation of a pair of mutually conjugate waves occurs with regard to the radiation loss due to the Rayleigh scattering by microspheres. We discuss the applicability scope of the results of [6] and the characteristic features of four-wave mixing in the diffusion limit, which are attributed to the nonuniformity of the effective radiation inside the microspheres. We present the results of the numerical simulation of the four-wave mixing process without any constraints imposed on the amplitudes of the interacting waves.

2. BASIC EQUATIONS

We will consider the process of four-wave mixing in a liquid suspension of transparent microspheres in the Rayleigh-Gans approximation [7]:

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

where λ is the radiation wavelength. Using inequalities (1), we can neglect the diffraction of radiation by microspheres [7], assuming that the effective field is specified; i.e., we can apply the so-called electrostatic approximation [8].

Let us represent the effective electromagnetic field as a sum of linearly polarized plane waves of frequency ω :

$$E = \frac{1}{2} \sum_{l=1}^4 E_l(z, t) \exp[-i(\omega t - \mathbf{k} \cdot \mathbf{r})] + \text{c.c.}, \quad (2)$$

where $E_l(z, t)$ are the wave amplitudes and $\mathbf{k}_l$ are the wavevectors (see Fig. 1). We will assume that the interacting waves are polarized in the direction perpendicular to the plane zx .

During the four-wave mixing, the microspheres are in the field of two pairs of counterpropagating waves of comparable intensities. Therefore, one can neglect small radiative forces of light pressure in the theory in question [8, 9]. In this case, a key contribution to the light-induced formation of a concentration response is made by gradient forces, whose amplitude is given by [10]

$$\mathbf{F}_\nabla = \alpha_0 \int_V \nabla |\bar{E}|^2 dV, \quad (3)$$

where

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

is the specific polarizability of a microsphere,

$$\begin{aligned} |\bar{E}|^2 = & \frac{1}{2} \{ |E_0|^2 + [E_1 E_2^* \exp(2i(k_z z + k_x x)) \\ & + E_3 E_4^* \exp(2i(k_z z - k_x x)) \\ & + (E_1 E_4^* + E_2^* E_3) \exp(2ik_z z) + (E_1 E_3^* + E_2^* E_4) \\ & \times \exp(2ik_x x) + \text{c.c.}] \} \end{aligned} \quad (5)$$

is the time-averaged intensity of the effective radiation, $|E_0|^2 = \sum_{l=1}^4 U_l$, $V = (4\pi/3)a^3$ is the volume of a microsphere, $k_z = k \cos \theta$ and $k_x = k \sin \theta$ are the corresponding projections of the wavevector, $k = (\omega/c)\bar{n}$, and $U_l = |E_l|^2$. In fact, the integral in (3) is the overlap integral of a microsphere and the nonuniform intensity of the effective radiation.

We will assume that the products of complex conjugate amplitudes of the interacting waves are slowly varying functions of the longitudinal coordinate:

$$\frac{\partial |E_l E_r^*|}{\partial z} \ll k_z |E_l E_r^*|. \quad (6)$$

In this approximation, with regard to (5), the integral in formula (3) can be calculated in spherical coordinates exactly [11]. After the integration in (3), we obtain the following expression for the gradient force:

$$\mathbf{F}_\nabla = \mathbf{j} F_x + \mathbf{k} F_z, \quad (7)$$

where $\mathbf{j}$ and $\mathbf{k}$ are unit vectors;

$$\begin{aligned} F_\kappa = & F_{\kappa 0} \exp[2i(k_z z - k_x x)] \\ & + F_{\kappa 1} \exp[2i(k_z z + k_x x)] + F_{\kappa 2} \exp(2ik_\kappa \kappa) + \text{c.c.} \end{aligned} \quad (8)$$

are the components of the gradient-force vector $\mathbf{F}_\nabla$, and $\kappa = \{x, z\}$. The amplitudes of the harmonics of the components F_κ are defined by

$$\begin{aligned} F_{x0} = & -i\alpha_0 k_x E_3 E_4^* V_0 = -(k_x/k_z) F_{z0}, \\ F_{x1} = & i\alpha_0 k_x E_1 E_2^* V_0 = (k_x/k_z) F_{z1}, \\ F_{x2} = & i\alpha_0 k_x (E_1 E_3^* + E_2^* E_4) V_x, \\ F_{z2} = & i\alpha_0 k_z (E_1 E_4^* + E_2^* E_3) V_z, \end{aligned} \quad (9)$$

where

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

$J_{3/2}(\xi)$ is the Bessel cylindrical function, and $\Lambda_0 = \pi/k$ and $\Lambda_\kappa = \pi/k_\kappa$ are the periods of the interference patterns of the corresponding pairs of waves. The constant coefficients V_0 and V_κ in (9) result from the integration of (3) and take into account the nonuniformity of radiation inside the microspheres. One can easily show that $V_0 = V_\kappa = V$ for $\xi \ll 1$. It is obvious that $|F_x/F_z| \propto \tan \theta$; hence, in the region of small angles $\theta \ll \pi/2$, the dominant role in the formation of the concentration response is played by the longitudinal (zth) component of the gradient force. It follows from (9) that, for certain ratios of the microsphere radius to the periods Λ_κ of the interference pattern of the field, namely, for $2\pi a/\Lambda_\kappa = \xi_i$ (where ξ_i are the roots of the Bessel cylindrical function $J_{3/2}(\xi)$, $i = 1, 2, \dots$), the corresponding component of the gradient force vanishes irrespective of the positions of the microspheres. The so-called effect of “zero force” is attributed to the fact that the oppositely directed components of this field that act on the corresponding elements of a microsphere are equal. This effect was theoretically predicted independently in [8, 11]. It follows from (9) that, for certain values of the parameters of the system (for example, for $2k_x a = \xi_1$ and $2k_z a = \xi_2$), the components $F_{\kappa 2}$ of the gradient forces vanish (i.e., $V_\kappa = 0$) irrespective of the intensity of the effective radiation. Using the values of the first two roots of the Bessel function $J_{3/2}(\xi)$ [12], one can show that the condition $V_\kappa = 0$ is fulfilled for $a/\lambda = 0.709$ and

$$\tan \theta = \xi_1/\xi_2 \approx 0.58,$$

i.e., $\theta \approx 30^\circ$. It is obvious that this condition is also satisfied for $\theta = 45^\circ$ and $\sqrt{2} ka = \xi_i$. In this case, the minimal value of the ratio a/λ corresponds to $\xi_i = \xi_1$ and is equal to $a/\lambda = 0.506$. By virtue of inequalities (1), the conditions obtained above can be satisfied in suspensions with small values of the relative refractive index $|\bar{m} - 1| \leq 10^{-2}$.

Thus, due to the zero force effect, the process of four-wave mixing, to which the main contribution is made by the components $F_{\kappa 2}$, can be nearly completely suppressed for appropriate values of the microsphere sizes and the convergence angle of the interacting waves. Note also that, depending on the radii of the microspheres, the amplitudes of gradient forces (9) are alternating functions. Therefore, the microspheres of appropriate sizes may be drawn either to the antinodes (when $J_{3/2}(\xi) > 0$) or to the nodes (when $J_{3/2}(\xi) < 0$) of the interference pattern of the field. This motion of microspheres can physically be explained by the tendency to cover a maximal number of antinodes of the interference pattern of the field [8, 11].

To determine the concentration response of microspheres induced by an electromagnetic field (see (1)),

we apply the two-dimensional Smolukhovskii equation¹ where
(see, for example, [13])

$$\begin{aligned} \frac{\partial N}{\partial t} &= D\Delta_{\perp}N \\ &- b \left[N \left(\frac{\partial F_x}{\partial x} + \frac{\partial F_z}{\partial z} \right) + \left(F_x \frac{\partial N}{\partial x} + F_z \frac{\partial N}{\partial z} \right) \right], \end{aligned} \quad (11)$$

where N is the concentration of microspheres (cm^{-3}), $D = k_B T / 6\pi\eta a$ is the diffusion coefficient (cm^2/s), k_B is the Boltzmann constant, T is temperature (K), η is the viscosity of the liquid, $b = D/k_B T$ is the mobility of microspheres, and $\Delta_{\perp} = \partial^2/\partial z^2 + \partial^2/\partial x^2$. Equation (11) is valid in the domain $t > t^*$ provided that the gradient force $\mathbf{F}_V$ is a slowly varying function on the time t^* and space l^* scales defined by the relations [13, 14]

$$t^* = bm_0, \quad l^* = \sqrt{\frac{k_B T}{m_0}} t^*, \quad (12)$$

where m_0 is the mass of a microsphere. In particular, for a water suspension of latex microspheres of radius $a = 1.17 \times 10^{-5} \text{ cm}$ with density 1 g/cm^3 , which was used in the experiment in [3], we have $t^* \approx 3 \times 10^{-9} \text{ s}$ and $l^* \approx 7 \times 10^{-9} \text{ cm}$ at room temperature.

It is convenient to represent the solution to Eq. (11) as a harmonic series

$$\begin{aligned} N(x, z, t) \\ = \sum_{m, n = -\infty}^{\infty} N_{mn}(t) \exp[2i(mk_z z + nk_x x)], \end{aligned} \quad (13)$$

where $N_{00} = \langle N \rangle_{x,z} = N_0 = \text{const}$ is the initial concentration of microspheres, $\langle \dots \rangle_{x,z}$ denotes spatial averaging, and $N_{mn}^* = N_{-m, -n}$. Substituting solution (13) into Eq. (11) and taking into account expression (8), we obtain the following system of kinetic equations for the amplitudes $N_{mn}(t)$ of concentration harmonics:

$$\begin{aligned} \left(\frac{\partial}{\partial t} + W_{mn} \right) N_{mn} &= a_{mn} N_{m-1, n+1} \\ &- a_{mn}^* N_{m+1, n-1} + b_{mn} N_{m-1, n-1} \\ &- b_{mn}^* N_{m+1, n+1} + c_n N_{m, n-1} - c_n^* N_{m, n+1} \\ &+ d_m N_{m-1, n} - d_m^* N_{m+1, n}, \end{aligned} \quad (14)$$

¹ This equation is known as the Planck–Nernst equation in English-language literature [6].

$$W_{mn} = 4D[(mk_z)^2 + (nk_x)^2],$$

$$a_{mn} = 2\alpha_0 b (mk_z^2 - nk_x^2) E_3 E_4^* V_0,$$

$$b_{mn} = 2\alpha_0 b (mk_z^2 + nk_x^2) E_1 E_2^* V_0, \quad (15)$$

$$c_{mn} = 2\alpha_0 b n k_x^2 (E_1 E_3^* + E_2^* E_4) V_x,$$

$$d_{mn} = 2\alpha_0 b m k_z^2 (E_1 E_4^* + E_2^* E_3) V_z.$$

When the radii of the microspheres and the convergence angles of the interacting waves are small, i.e., $a \ll \Lambda_0$ and $\theta \ll \pi/2$, we can neglect the component F_x of the gradient force; then, the system of equations (14) is reduced to

$$\begin{aligned} \left(\frac{\partial}{\partial t} + W_{mn} \right) N_{mn} &= 2\alpha_0 b V m k_z^2 [E_3 E_4^* N_{m-1, n+1} \\ &- E_3^* E_4 N_{m+1, n-1} + E_1 E_2^* N_{m-1, n-1} \\ &- E_1^* E_2 N_{m+1, n+1} + (E_1 E_4^* + E_2^* E_3) N_{m-1, n} \\ &- (E_1^* E_4 + E_2 E_3^*) N_{m+1, n}]. \end{aligned} \quad (16)$$

The nonlinear polarization of a suspension of microspheres induced by the variation of their concentration under gradient forces is defined by [6]

$$P = \frac{1}{2} \alpha_0 V N(x, z, t) \times \sum_{l=1}^4 E_l(z, t) \exp[-i(\omega t - \mathbf{k} \cdot \mathbf{r})] + \text{c.c.} \quad (17)$$

Substituting formulas (2) and (17) into the wave equation, we obtain the following system of truncated equations for the amplitudes of the interacting waves:

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

where $v = c/\bar{n}$ is the velocity of light in the suspension,

$$\gamma = 2\pi \frac{k}{\bar{n}^2} \alpha_0 V N_0, \quad \chi_{mn} = \frac{N_{mn}}{N_0}.$$

In Eqs. (18), we took into account the equation $N_{mn}^* = N_{-m, -n}$ and phenomenologically introduced the amplitude coefficient of loss [6, 14]

$$\rho = \frac{8\pi}{3} N_0 k^4 \left(\frac{\bar{m}^2 - 1}{\bar{m}^2 + 2} \right)^2 a^6, \quad (19)$$

due to the Rayleigh scattering by microspheres. Equations (18) imply that only two pairs of gratings from the spectrum of excited concentration gratings (13) take part in the process of four-wave mixing: one pair of orthogonal gratings,

$$N_\kappa \propto \cos(2k_\kappa \kappa), \quad \kappa = x, z,$$

leads to the parametric coupling of waves and the energy exchange among them, and the other pair,

$$N_\pm \propto \cos[2(k_z z \pm k_x x)],$$

leads to self-action phenomena. Due to the spatial averaging of the wave equations, other concentration gratings do not make a direct contribution to four-wave mixing. The indirect role of these gratings reduces to the influence on the values of the amplitudes of the main gratings N_κ and $N_\pm$.

The simultaneous system of wave equations (18) and kinetic equations (14) subject to the boundary conditions

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

and the initial conditions

$$\begin{aligned} N_{mn}(t = -\infty) &= 0, \quad m \neq 0, \quad n \neq 0, \\ N_{00}(t = -\infty) &= N_0 \end{aligned} \quad (21)$$

describes the process of four-wave mixing of waves of arbitrary intensities on the concentration nonlinearity due to the action of gradient forces on transparent microspheres.

3. KINETICS OF A CONCENTRATION GRATING OF MICROSPHERES

Based on Eqs. (14), we consider the kinetics of a concentration grating of microspheres induced by the interference field of two waves. Assume, for definiteness, that $E_2 = E_4 = 0$. Then, $a_{mn} = b_{mn} = d_m = 0$ and $c_n =$

$2\alpha_0 b n k_x^2 V_x E_1 E_3^*$; hence, the amplitudes N_{mn} of the concentration gratings and the relaxation constants W_{mn} depend only on the index n ($N_{mn} \equiv N_n$ and $W_{mn} \equiv W_n$). In this case, Eqs. (14) are significantly simplified and reduced to

$$\frac{\partial N_n}{\partial t} + W_n N_n = n(A N_{n-1} - A^* N_{n+1}), \quad (22)$$

where $W_n = 4Dn^2 k_x^2$ and $A = 2\alpha_0 b k_x^2 V_x E_1 E_3^*$.

For relatively weak waves, when higher order gratings ($n > 2$) can be neglected, Eqs. (22) yield

$$\begin{aligned} \frac{\partial N_1}{\partial t} + \Gamma N_1 &= (A N_0 - A^* N_2), \\ \frac{\partial N_2}{\partial t} + 4\Gamma N_2 &= 2A N_1, \end{aligned} \quad (23)$$

where $\Gamma = 4Dk_x^2$.

For rectangular pulses ($E_l(t) = E_l = \text{const}$ for $t \geq 0$, $l = 1, 3$), a solution to system of equations (23) can be obtained in the explicit form (see Appendix A). It follows from solution (A.3) that, due to the excitation of the second-order grating N_2 , the amplitude of the main grating N_1 contains two components whose relaxation times are less than $t = 1/\Gamma$. The greater relaxation time of the main grating N_1 is estimated by the relation

$$\begin{aligned} \tau_1 &\approx \frac{1}{\Gamma} \left(1 - \frac{2|A|^2}{3\Gamma^2} \right) \\ &= \frac{1}{4Dk_x^2} \left[1 - \frac{1}{6} \left(\frac{\alpha_0 b V_x}{D} \right)^2 U_1 U_3 \right], \end{aligned} \quad (24)$$

which is valid for $U_1 U_3 < 6D^2/(\alpha_0 b V_x)^2$. Thus, in the approximation used, the relaxation time τ_1 of the main grating decreases linearly as the product of the intensities of the exciting waves increases. It follows from Eqs. (23) that the relaxation times $\bar{\tau}_n$ of the concentration gratings are determined by the diffusion coefficient and the periods of the gratings:

$$\bar{\tau}_n = 1/n^2 \Gamma, \quad n = 1, 2.$$

In particular, for the parameter values $D = 1.88 \times 10^{-8} \text{ cm}^2/\text{s}$ and $\Lambda_x = 3.5 \times 10 \text{ cm}^{-4}$, which correspond to the experiment of [3], we have $\bar{\tau}_1 = 165 \text{ ms}$; this result agrees well with the relaxation time $\bar{\tau}_1 = 140 \pm 40 \text{ ms}$ of the main grating N_1 measured in [3].

Figure 2 shows the time dependence of the normalized amplitude of the main grating, calculated from Eqs. (22), for various values of the intensities of the effective radiation. Figure 3 shows the relaxation time τ_1 of the main grating as a function of the radiation

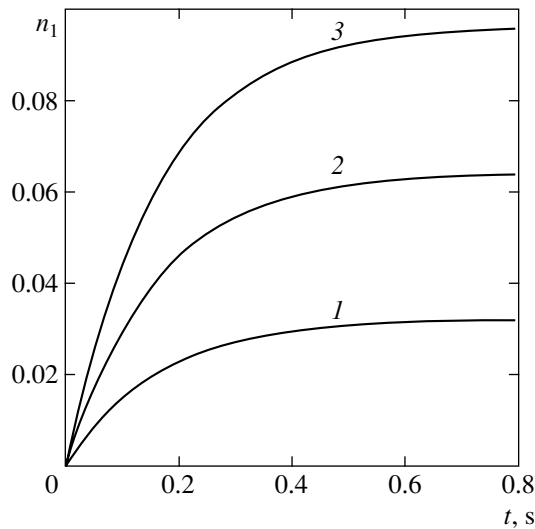


Fig. 2. Kinetics of the amplitude $n_1 = N_1/N_0$ of the main grating for $\xi = 2ka = 3.8$ and $\theta = 3.2^\circ$ for the parameter values $[\mathcal{E}_0]^2 = 2\alpha_0 V \sqrt{U_1 U_2} / k_B T = 0.2 \times 10^{-7}$ (1), 0.4×10^{-7} (2), and 0.6×10^{-7} (3).

intensity $I = (c\bar{n}/8\pi)U_0$ and the convergence angle θ of the interacting waves. The analysis shows that, for the maximal value of the intensity of the interacting waves, it suffices to restrict the consideration to gratings with $n \leq 8$ in Eqs. (22).

4. THEORY OF FOUR-WAVE MIXING IN THE DIFFUSION LIMIT

The diffusion approximation [6]

$$N(z, x, t) = N_0 + \bar{N}(z, x, t), \quad |\bar{N}| \ll N_0, \quad (25)$$

imposes a constraint on the intensities of the interacting waves and corresponds to the situation when the terms

proportional to the amplitudes of the concentration gratings N_{mn} ($m, n \neq 0$) on the right-hand sides of Eqs. (14) are neglected. In this case, Eqs. (14) imply the following relations for the components χ_{mn} that determine the process of four-wave mixing:

$$\left(\frac{\partial}{\partial t} + 4Dk_x^2\right)\chi_{01} = 2\alpha_0 V_x b k_x^2 (E_1 E_3^* + E_2^* E_4),$$

$$\left(\frac{\partial}{\partial t} + 4Dk_z^2\right)\chi_{10} = 2\alpha_0 V_z b k_z^2 (E_1 E_4^* + E_2^* E_3), \quad (26)$$

$$\left(\frac{\partial}{\partial t} + 4Dk^2\right)\chi_{11} = 2\alpha_0 V_0 b k^2 E_1 E_2^*,$$

$$\left(\frac{\partial}{\partial t} + 4Dk^2\right)\chi_{1,-1} = 2\alpha_0 V_0 b k^2 E_3 E_4^*.$$

In the diffusion limit, the relaxation times

$$\tau_0 = \frac{1}{4Dk^2} = \frac{(\Lambda_0/2\pi)^2}{D} \quad \text{and} \quad \tau_\kappa = \frac{1}{4Dk_\kappa^2} = \frac{(\Lambda_\kappa/2\pi)^2}{D},$$

of the concentration gratings do not depend on the radiation intensity and are determined by the diffusion coefficients and the periods of the interference pattern formed by the interacting waves. It follows from Eqs. (26) and (10) that, due to the nonuniformity of the radiation intensity inside the microspheres, the steady-state amplitudes of the concentration gratings depend on the convergence angle θ of the waves and take bounded values as the radius a of microspheres increases. Note that, for small radii of microspheres (for $V_0 = V_\kappa = V$) [6], the amplitudes of the concentration gratings do not depend on the angle θ and indefinitely increase as the microsphere radius increases.

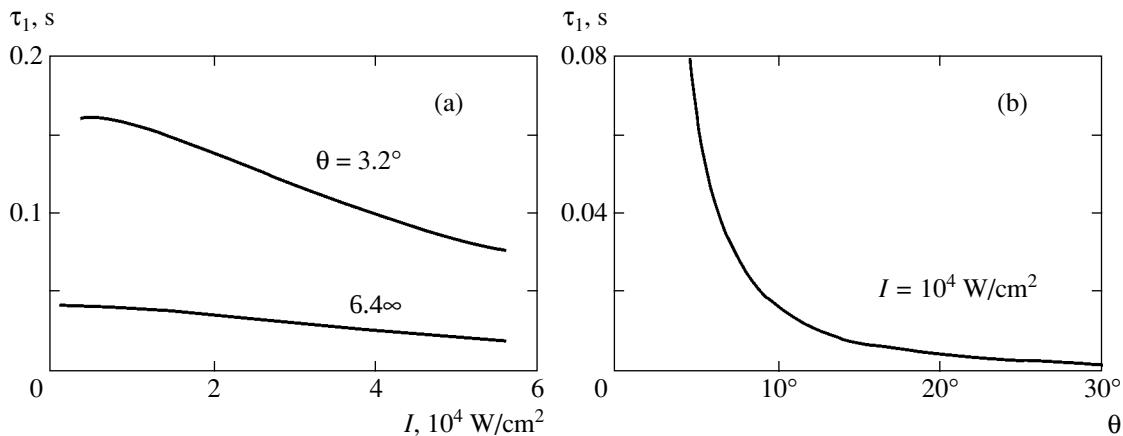


Fig. 3. Relaxation time τ_1 of the main grating as a function of (a) the intensity of effective radiation and (b) the convergence angle θ for $\xi = 2ka = 3.8$, $k = 1.6 \times 10^5 \text{ cm}^{-1}$, $\alpha_0 = 5.3 \times 10^{-2}$, and $k_B T = 4.14 \times 10^{-14} \text{ erg}$.

In the steady-state regime (for $t \gg \tau_k$), Eqs. (18) with regard to (26) are rewritten as

$$\begin{aligned}
 & \cos\theta \frac{dE_1}{dz} \\
 &= i\gamma \left\{ \left[1 + \frac{\alpha_0 b}{2D} (V_0 U_2 + V_x U_3 + V_z U_4) \right] E_1 \right. \\
 & \quad \left. + \frac{\alpha_0 b}{2D} (V_x + V_z) E_2^* E_3 E_4 \right\} - \rho E_1, \\
 & -\cos\theta \frac{dE_2}{dz} \\
 &= i\gamma \left\{ \left[1 + \frac{\alpha_0 b}{2D} (V_0 U_1 + V_z U_3 + V_x U_4) \right] E_2 \right. \\
 & \quad \left. + \frac{\alpha_0 b}{2D} (V_x + V_z) E_1^* E_3 E_4 \right\} - \rho E_2, \\
 & \cos\theta \frac{dE_3}{dz} \\
 &= i\gamma \left\{ \left[1 + \frac{\alpha_0 b}{2D} (V_x U_1 + V_z U_2 + V_0 U_4) \right] E_3 \right. \\
 & \quad \left. + \frac{\alpha_0 b}{2D} (V_x + V_z) E_4^* E_1 E_2 \right\} - \rho E_3, \\
 & -\cos\theta \frac{dE_4}{dz} \\
 &= i\gamma \left\{ \left[1 + \frac{\alpha_0 b}{2D} (V_x U_1 + V_z U_2 + V_0 U_3) \right] E_4 \right. \\
 & \quad \left. + \frac{\alpha_0 b}{2D} (V_x + V_z) E_3^* E_1 E_2 \right\} - \rho E_4.
 \end{aligned} \tag{27}$$

Neglecting the reaction of weak waves E_3 and E_4 to the intense reference waves E_1 and E_2 , from Eqs. (27) we obtain the following relations for $\rho = 0$:

$$\begin{aligned}
 \cos\theta \frac{dE_3}{dz} &= i\{\bar{\kappa}_3 E_3 + \beta E_4^* E_{10} E_{20} \\
 & \quad \times \exp[i\bar{\kappa}_1 \hat{z} + i\bar{\kappa}_2 (\hat{L} - \hat{z})]\}, \\
 -\cos\theta \frac{dE_4}{dz} &= -i\{\bar{\kappa}_4 E_4^* + \beta E_3 E_{10}^* E_{20}^* \\
 & \quad \times \exp[-i\bar{\kappa}_1 \hat{z} - i\bar{\kappa}_2 (\hat{L} - \hat{z})]\},
 \end{aligned} \tag{28}$$

$$\begin{aligned}
 \bar{\kappa}_1 &= \gamma \left[1 + \frac{\alpha_0 b}{2D} V_0 U_2 \right], \\
 \bar{\kappa}_2 &= \gamma \left[1 + \frac{\alpha_0 b}{2D} V_0 U_1 \right], \\
 \bar{\kappa}_3 &= \gamma \left[1 + \frac{\alpha_0 b}{2D} (V_x U_1 + V_z U_2) \right], \\
 \bar{\kappa}_4 &= \gamma \left[1 + \frac{\alpha_0 b}{2D} (V_z U_1 + V_x U_2) \right],
 \end{aligned} \tag{29}$$

$\beta = (\gamma \alpha_0 b / 2D)(V_x + V_z)$ is the coefficient of parametric coupling, $\hat{z} = z/\cos\theta$, and $\hat{L} = L/\cos\theta$. The solution to Eqs. (28) subject to the boundary conditions $E_3(0) = E_{30}$ and $E_4^*(\hat{L}) = 0$ has the form

$$\begin{aligned}
 E_3(z) &= E_{30} \exp \left[i \left(\frac{\Delta}{2} + \bar{\kappa}_3 \right) \hat{z} \right] \\
 & \times \frac{2\Gamma \cos[\Gamma(\hat{L} - \hat{z})] + i\Delta \sin[\Gamma(\hat{L} - \hat{z})]}{2\Gamma \cos\Gamma\hat{L} + i\Delta \sin\Gamma\hat{L}}, \\
 E_4^*(z) &= 2i\beta E_{30} \sqrt{U_1 U_2} \\
 & \times \exp \left[-i \left(\frac{\Delta}{2} - \bar{\kappa}_4 \right) \hat{z} + \bar{\kappa}_2 \hat{L} \right] \\
 & \times \frac{\sin[\Gamma(\hat{L} - \hat{z})]}{2\Gamma \cos\Gamma\hat{L} + i\Delta \sin\Gamma\hat{L}},
 \end{aligned} \tag{30}$$

where

$$\Gamma = \sqrt{\beta^2 U_1 U_2 + \Delta^2 / 4} \tag{31}$$

is the increment of the parametric amplification of weak radiation and

$$\begin{aligned}
 \Delta &= \bar{\kappa}_1 - \bar{\kappa}_2 - \bar{\kappa}_3 + \bar{\kappa}_4 \\
 &= \gamma \frac{\alpha_0 b}{2D} (V_0 + V_x - V_z)(U_2 - U_1)
 \end{aligned} \tag{32}$$

is the phase mismatch of the interacting waves.

It is obvious that the amplitude of the reversed wave E_4 is proportional to the coefficient β of parametric coupling. Using formulas (10), one can show that, for certain values of the microsphere radius a and the convergence angle θ of the interacting waves, the coef-

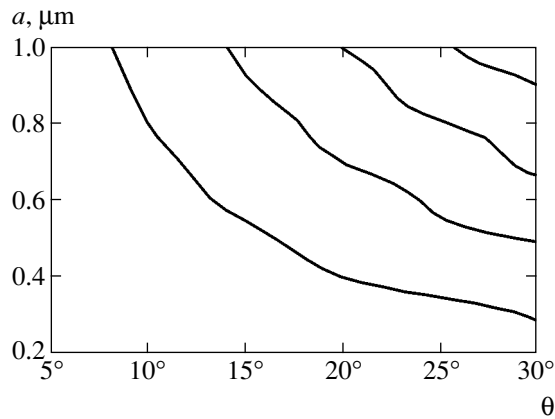


Fig. 4. Family of solutions to the equation $\beta(a, \theta) = 0$ for $k = 1.6 \times 10^5 \text{ cm}^{-1}$.

cient β vanishes. For these values of the parameters a and θ , due to the zero-force effect, concentration gratings with periods Λ_k , which are responsible for the four-wave mixing, are not induced and hence a parametric generation of the reversed wave E_4 does not occur. Figure 4 represents a family of curves on the plane $a\theta$ that shows the values of the parameters a and θ at which the coefficient of parametric coupling vanishes ($\beta = 0$).

It follows from solutions (30) that the linear theory of four-wave mixing for $\Delta = 0$ ($\Gamma = \Gamma_0 = |\beta|\sqrt{U_1 U_2}$) imposes a constraint on the intensities of the reference waves; this constraint is associated with possible parametric generation of a pair of mutually conjugate waves E_3 and E_4^* with the threshold given by the condition [15]

$$\Gamma_0 L / \cos \theta = \pi/2. \quad (33)$$

Taking into account the definition of the coupling coefficient β from (33), we obtain

$$2kN_0 \frac{\alpha_0^2 b}{\bar{n}^2 D} V |V_x + V_z| \sqrt{U_1 U_2} \frac{L}{\cos \theta} = 1. \quad (34)$$

It follows from (32) that the condition $\Delta = 0$ is satisfied in two cases: either when $U_1 = U_2$ or when $V_0 + V_x - V_z \equiv \bar{V} = 0$. Note that, $V_0 = V_k = V$ for small microspheres, $a \ll \Lambda_0$; hence, the condition $\Delta = 0$ is met, just as in the case of ordinary media with the Kerr nonlinearity [15], only for $U_1 = U_2$.

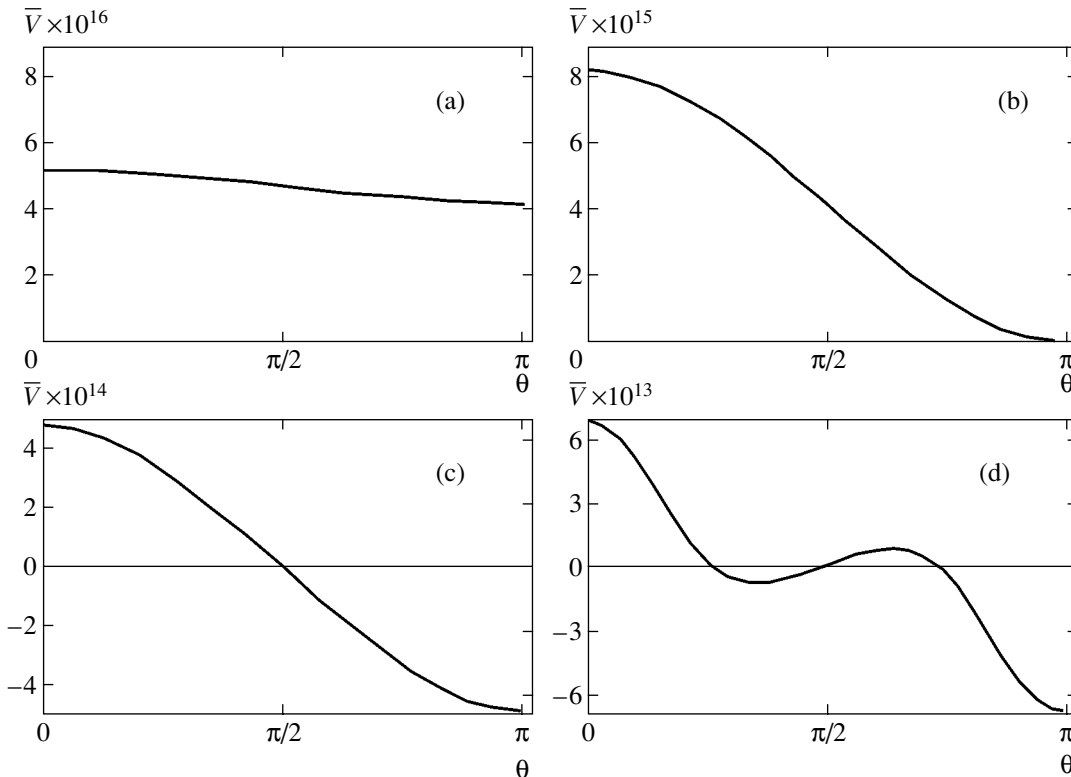


Fig. 5. Parameter $\bar{V}$ versus the angle θ for various values of the coefficient $\xi = 2ka$ for $k = 10^5 \text{ cm}^{-1}$; (a) $\xi = 1.0$, (b) $\xi = 2.5$, (c) $\xi = 4.5$, and (d) $\xi = 10.9$.

Figure 5 presents the parameter $\bar{V}$ as a function of the angle θ for various values of the coefficient $\xi = 2ka$. One can see that the parameter $\bar{V}(\theta)$ may vanish in the region of $\xi \geq 2.5$. The threshold value of the coefficient $\xi = \xi_{\text{thr}} \approx 2.5$ at which $\bar{V} = 0$ corresponds to the angle $\theta = \pi/2$. When $\xi > 2.5$, the corresponding values of θ may be much smaller. Hence, in the region of $\xi > 2.5$ ($a > 0.2\lambda$), for appropriate values of the convergence angle θ , one can implement conditions under which weak radiation is generated parametrically at nonequal values of the intensities of the reference waves, $U_1 \neq U_2$. Note that, for relatively large values of $\theta \geq \pi/6$, Eqs. (18) must contain derivatives with respect to the transverse coordinate ($\sin\theta\partial/\partial x$).

A comparison of the results obtained in the diffusion limit with the similar results of theory [6] is illustrated in Fig. 6. This figure represents the reflection coefficient η of the reversed wave, calculated by the second equation in (30), as a function of the microsphere size for $U_1 = U_2 = U_0$ and for various values of the convergence angle θ of the waves. For comparison, the dashed line represents the reflection coefficient $\eta = \eta(\xi)$ calculated by the theory of [6]. One can see that the theory of [6], in which a gradient force is determined under the assumption that the intensity of radiation inside the microspheres is uniform, $\mathbf{F}_\nabla = \alpha_0 V \nabla |\bar{\mathbf{E}}|^2$, predicts a significantly overstated value of η and is valid in the region of small $\xi \ll 1$. Within the approximations used in [6], the coefficient η is independent of the convergence angle θ of the waves and hence is the same for any values of θ . One can see in Fig. 6 that, for relatively large values of θ , when the overlap of the microspheres with the interference pattern of the field becomes significant, the reflection coefficient $\eta(\xi)$ is a nonmonotonic function of the size of microspheres and vanishes at angles of $\theta = 30^\circ$ and $\theta = 45^\circ$ (at the points $\xi \approx 8.9$ and $\xi \approx 6.36$, respectively). As pointed out above, these values of the parameters correspond to the conditions under which the process of four-wave mixing is suppressed due to the vanishing of the resultant components $F_{\kappa 2}$ of the gradient force.

One can show that the consideration of linear loss ($\rho \neq 0$) in the case of $\bar{V} = 0$ leads to the following relation, which defines a threshold of the parametric generation of weak waves (see Appendix B):

$$\tan \hat{\Gamma} \hat{L} = -\hat{\Gamma}/\rho, \quad (35)$$

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

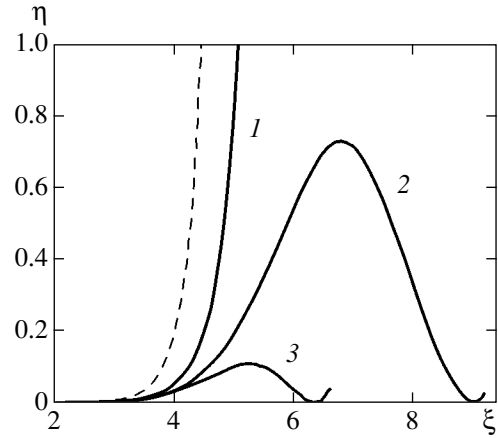


Fig. 6. Reflection coefficient η of the reversed wave as a function of the parameter ξ for $\gamma\alpha_0 V U_0 L / k_B T = 10^{-4}\xi^6$ and various values of the convergence angle θ : (1) $\theta = 3.2^\circ$, (2) $\theta = 30^\circ$, and (3) $\theta = 45^\circ$; dashed curve is calculated by using the theory presented in [6].

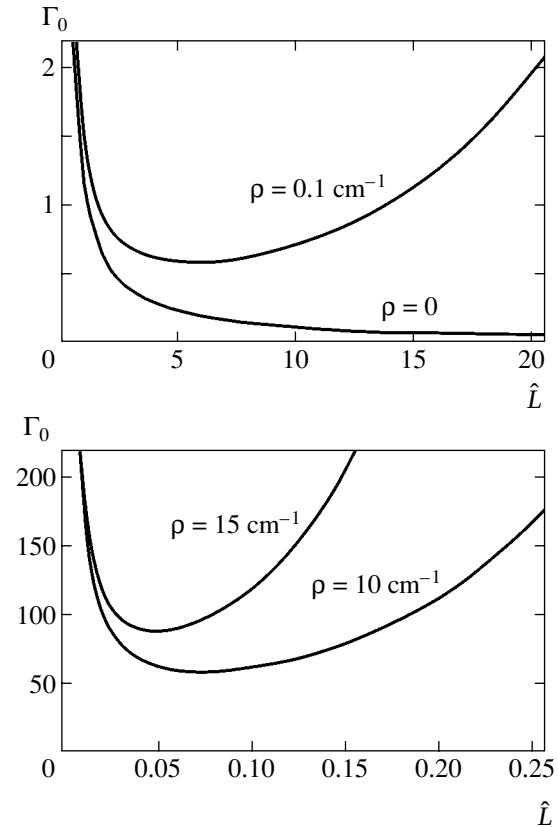


Fig. 7. Normalized threshold intensity Γ_0 of reference waves versus the length $\hat{L}$ for various values of the coefficient ρ .

Figure 7 shows the threshold value $\Gamma_0 = |\beta| \sqrt{U_1 U_2}$ as a function of the length $\hat{L}$ for various values of the amplitude coefficient of loss ρ . One can see that, for $\rho \neq 0$, the optimal value of $\hat{L}$ is equal to $(\rho \hat{L})_{\text{opt}} \approx 0.74$.

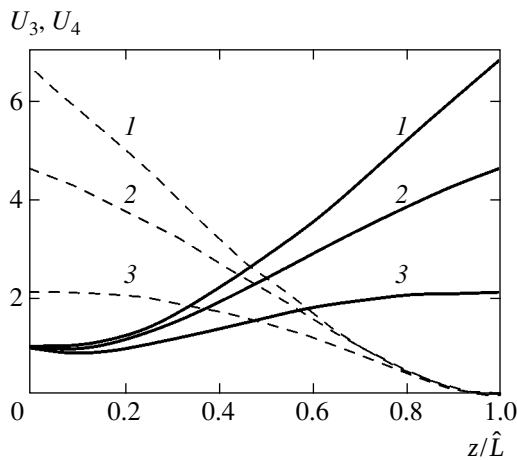


Fig. 8. Steady-state distributions of the intensities of the signal U_3 (solid lines) and reversed U_4 (dashed lines) waves for various microsphere sizes a : (1) 0.1 μm , (2) 0.11 μm , and (3) 0.12 μm .

For comparison, note that, for an ordinary medium with cubic nonlinearity, approximate analytic estimates yield a value of $(\rho\hat{L})_{\text{opt}} \approx 0.69$ [16].

5. CONCLUSIONS

The results obtained above show that, within the approximations used in [6], the concentration nonlinearity of a liquid suspension of transparent microspheres cannot be correctly calculated for a wide range of parameters. In particular, a rigorous calculation of the concentration nonlinearity yields much smaller values of the reflection coefficient of a reversed wave compared to those predicted by the theory of [6]. Nevertheless, estimates show that a liquid suspension of transparent microspheres—an artificial heterogeneous medium each of whose components taken separately does not exhibit nonlinear optical properties—may serve as a prospective wideband material for reversing the wave front of low-intensity radiation of continuous-wave lasers. The theoretical results obtained in the present paper are in good agreement with the experimental values measured in [3], of the reflection coefficient η of a reversed wave and the relaxation time τ_1 of a concentration grating versus the intensity of reference waves.

Figure 8 presents the spatial distributions of the intensities of the signal U_3 and reversed U_4 waves that is obtained by the numerical simulation of the simultaneous system of the wave (18) and kinetic (14) equations for the four-wave mixing of argon laser radiation ($E_{10}(t) = E_{20}(t) = E_0$ for $t \geq 0$, $E_{30}(t) = 1$) in a water suspension of latex microspheres [3] at room temperature: $n_0 = 1.59$, $\bar{n} = 1.33$, $N_0 = 6.5 \times 10^{10} \text{ cm}^{-3}$, $L = 10^{-2} \text{ cm}$, $\theta = 3.2^\circ$, $D = 2.2 \times 10^{-13} \text{ a}^{-1} \text{ cm}^2/\text{s}$, and the intensities $I = 7.5 \times 10^4 \text{ W/cm}^2$ of the reference waves. One can

see in Fig. 8 that, for the chosen range of the parameters, the efficiency of the four-wave mixing is quite sensitive to the size of microspheres. In this case, an increase in the microsphere radius by 20% leads to about a threefold increase in the reflection coefficient η of the reversed wave.

In this study, by analogy with [6], we have taken into account the radiation damping due to the Rayleigh scattering phenomenologically, by adding a linear loss factor $\rho \propto N_0$ to the wave equations (18). Strictly speaking, the spatial modulation of microspheres by the effective radiation leads to the dependence of the loss factor on the radiation intensity, $\rho = \rho(|E|^2)$. A rigorous consideration of the nonlinearity of the loss factor is guaranteed by an appropriate modification of formula (17), which has the form

$$P = \frac{\alpha_0 V}{2} N(x, z, t) \left\{ 1 + i \frac{8}{3} k \left(\frac{\omega}{c} \right)^2 \alpha_0 V \right\} \times \sum_{l=1}^4 E_l \exp[-i(\omega t - \mathbf{k} \cdot \mathbf{r})] + \text{c.c.}$$

in the general case. Hence, generally speaking, the processes of Rayleigh scattering also modulate the imaginary part of the dielectric constant of a suspension; i.e., amplitude-phase gratings are induced in the suspension whose amplitude component should be taken into account provided that $\|\alpha_0\|V \geq k^{-3}$.

Under relatively high intensities of the interacting waves, high concentrations of microspheres at the antinodes (nodes) of the interference field may lead to the formation of clusters even in a rarefied suspension [3]; naturally, this fact leads to a qualitative change in the process of four-wave mixing. Moreover, in this case, one should also take into account that the size of microspheres is finite, which restricts the limiting concentrations to $N_{\text{lim}} = 1/V$.

Thus, a further development of the theory of four-wave mixing in liquid suspensions of transparent microspheres should be carried out with regard to the nonlinearity of the imaginary part of the dielectric constant of the suspension, the formation of clusters of microspheres, and the saturation of the amplitudes of the concentration gratings due to the finiteness of the volume of microspheres.

APPENDICES

APPENDIX A

Upon the substitution of $N_n = \hat{N}_n \exp(-4\Gamma t)$, the system of equations (23) is reduced to the following second-order equation for $\hat{N}_1$:

$$\frac{d^2 \hat{N}_1}{dt^2} - 3\Gamma \frac{d\hat{N}_1}{dt} + 2|A|^2 \hat{N}_1 = 4\Gamma A N_0 \exp(4\Gamma t). \quad (\text{A.1})$$

A solution to this equation for $\lambda^2 = 9\Gamma^2 - 8|A|^2 > 0$ is given by

$$\begin{aligned} \hat{N}_1(t) = & \exp\left(\frac{3\Gamma}{2}t\right) \left\{ C_+ \exp\left(\frac{\lambda}{2}t\right) \right. \\ & + C_- \exp\left(-\frac{\lambda}{2}t\right) + 8\frac{\Gamma}{\lambda} AN_0 \int_0^t \exp\left(-\frac{3\Gamma}{2}t'\right) \\ & \left. \times \sinh\left[\frac{\lambda}{2}(t-t')dt'\right] \right\}, \end{aligned} \quad (\text{A.2})$$

where $C_{\pm}$ are integration constants. The initial condition $\hat{N}_1(0) = 0$ implies the relation $C_+ = -C_-$. The coefficient C_+ is determined from the condition that

$$\hat{N}_2 \rightarrow 0, \quad \hat{N}_1 \rightarrow \frac{AN_0}{\Gamma} [1 - \exp(-\Gamma t)] \exp(4\Gamma t),$$

as $A \rightarrow 0$. It can easily be shown that this passage to the limit is attained for $C_+ = N_0 A / 3\Gamma$. Taking this fact into account, from (A.2) we find

$$\begin{aligned} N_1(t) = & \frac{2AN_0}{3\Gamma} \exp\left(-\frac{5\Gamma}{2}t\right) \\ & \times \left\{ \sinh\left(\frac{\lambda}{2}t\right) + \frac{3\Gamma^2}{2\Gamma^2 + |A|^2} \right. \\ & \left. \times \left[\exp\left(\frac{5\Gamma}{2}t\right) - \cosh\left(\frac{\lambda}{2}t\right) - 5\frac{\Gamma}{\lambda} \sinh\left(\frac{\lambda}{2}t\right) \right] \right\}. \end{aligned} \quad (\text{A.3})$$

Similarly, for $N_2(t)$, the second equation in (23) yields

$$N_2(t) = 2A \int_0^t N_1(t') \exp[-4\Gamma(t-t')] dt'. \quad (\text{A.4})$$

APPENDIX B

For $\rho \neq 0$, the first two equations in (27) yield the following equations for the amplitudes of the reference waves:

$$\begin{aligned} \frac{dE_1}{d\hat{z}} = & i\gamma \left(1 + \frac{\alpha_0 b}{2D} V_0 |E_2|^2 \right) E_1 - \rho E_1, \\ -\frac{dE_2}{d\hat{z}} = & i\gamma \left(1 + \frac{\alpha_0 b}{2D} V_0 |E_1|^2 \right) E_2 - \rho E_2. \end{aligned} \quad (\text{B.1})$$

A solution to these equations is given by

$$E_1(\hat{z}) = E_{10} \exp \left\{ (i\gamma - \rho)\hat{z} + i\gamma \frac{\alpha_0 b}{2D} V_0 |E_{20}|^2 \right.$$

$$\left. \times \exp(-2\rho\hat{L}) \int_0^{\hat{L}} \exp(2\rho z) dz \right\}, \quad (\text{B.2})$$

$$\begin{aligned} E_2(\hat{z}) = & E_{20} \exp \left\{ (i\gamma - \rho)(\hat{L} - \hat{z}) - i\gamma \frac{\alpha_0 b}{2D} \right. \\ & \left. \times V_0 |E_{10}|^2 \int_{\hat{L}}^{\hat{z}} \exp(-2\rho z) dz \right\}. \end{aligned}$$

The substitution of solutions (B.2) into the last two equations in (28) yields

$$\begin{aligned} \frac{dE_3}{d\hat{z}} = & i\{\bar{\kappa}_3(\hat{z}) + \beta E_{10} E_{20} E_4^* \\ & \times \exp[(i\gamma - \rho)\hat{z} + i\varphi(\hat{z})]\} - \rho E_3, \\ -\frac{dE_4^*}{d\hat{z}} = & -i\{\bar{\kappa}_4(\hat{z}) + \beta E_{10}^* E_{20}^* E_3 \\ & \times \exp[-(i\gamma + \rho)\hat{z} - i\varphi(\hat{z})]\} - \rho E_4^*, \end{aligned} \quad (\text{B.3})$$

where

$$\begin{aligned} \bar{\kappa}_3(\hat{z}) = & \gamma \left\{ 1 + \frac{\alpha_0 b}{2D} [V_x |E_{10}|^2 \exp(-2\rho\hat{z}) \right. \\ & \left. + V_z |E_{20}|^2 \exp(-2\rho(\hat{L} - \hat{z}))] \right\}, \\ \bar{\kappa}_4(\hat{z}) = & \gamma \left\{ 1 + \frac{\alpha_0 b}{2D} [V_x |E_{10}|^2 \exp(-2\rho\hat{z}) \right. \\ & \left. + V_z |E_{20}|^2 \exp(-2\rho(\hat{L} - \hat{z}))] \right\}, \end{aligned}$$

$$\begin{aligned} \varphi(\hat{z}) = & \gamma \frac{\alpha_0 b}{2D} V_0 \left[|E_{20}|^2 \exp(-2\rho\hat{L}) \int_0^{\hat{z}} \exp(2\rho z) dz \right. \\ & \left. - |E_{10}|^2 \int_0^{\hat{z}} \exp(-2\rho z) dz \right]. \end{aligned}$$

The substitutions

$$\begin{aligned} E_3(\hat{z}) = & \mathcal{E}_3(\hat{z}) \exp \left(-\rho\hat{z} + i \int_0^{\hat{z}} \bar{\kappa}_3 dz \right), \\ E_4^*(\hat{z}) = & \mathcal{E}_4^*(\hat{z}) \exp \left(-\rho(L - \hat{z}) + i \int_{\hat{L}}^{\hat{z}} \bar{\kappa}_4 dz \right) \end{aligned} \quad (\text{B.4})$$

reduce the system of equations (B.3) to the following second-order equation for the amplitude $\mathcal{E}_3(\hat{z})$:

$$\frac{d^2 \mathcal{E}_3}{d\hat{z}^2} - [2\rho + i\Delta(\hat{z})] \frac{d\mathcal{E}_3}{d\hat{z}} + \beta^2 |E_{10}|^2 |E_{20}|^2 \exp(-2\rho \hat{L}) \mathcal{E}_3 = 0, \quad (\text{B.5})$$

where

$$\Delta(\hat{z}) = \gamma \frac{\alpha_0 b}{2D} \bar{V}(\theta) \{ |E_{20}|^2 \exp[-2\rho(\hat{L} - \hat{z})] - |E_{10}|^2 \exp(-2\rho \hat{z}) \}.$$

It follows from (B.5) that, for $\bar{V}(\theta) = 0$, the roots of the characteristic equation

$$\lambda^2 - 2\rho\lambda + \beta^2 |E_{10}|^2 |E_{20}|^2 \exp(-2\rho \hat{L}) = 0 \quad (\text{B.6})$$

are given by

$$\lambda_{\pm} = \rho \pm i\sqrt{\beta^2 |E_{10}|^2 |E_{20}|^2 \exp(-2\rho \hat{L}) - \rho^2} \equiv \rho \pm i\hat{\Gamma}. \quad (\text{B.7})$$

Applying the boundary conditions $E_3(0) = E_{30}$ and $E_4^*(\hat{L}) = 0$, we determine the integration constants $C_{\pm}$ of the system of equations (B.3):

$$C_{\pm} = \pm E_{30} \frac{\lambda_{\mp} \exp(\mp i\hat{\Gamma}\hat{L})}{\lambda_{-} \exp(-i\hat{\Gamma}\hat{L}) - \lambda_{+} \exp(i\hat{\Gamma}\hat{L})}. \quad (\text{B.8})$$

The condition $\lambda_{-} \exp(-i\hat{\Gamma}\hat{L}) - \lambda_{+} \exp(i\hat{\Gamma}\hat{L}) = 0$ implies formula (35).

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