Reversal of wave front in four-photon processes under conditions of two-quantum resonance

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A procedure for averaging nonlinear quasioptical equations is developed and simplifies considerably the investigation of the wave-front reversal (WFR) in four-photon interactions of opposing light wave having complicated space-time structures. The effect of WFR of multimode light beams is analytically investigated with allowance for the spatial variation of all the interacting waves. It is shown that in the case of four-photon Raman interaction the coefficient of conversion into the reversed wave can be much larger than unity. The corresponding conclusions were confirmed experimentally.

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1. INTRODUCTION

Much progress was made recently in theoretical and experimental research into the phenomenon of wave-front reversal (WFR) of optical radiation in three- and four-wave parametric interactions of light waves.[1-6] Particular attention was paid here to four-wave interactions, which make possible not only WFR of low-power radiation with a conversion coefficient larger than unity, but also effective control of such reversed-radiation parameters as the frequency, polarization, duration, and waveform of the pulse. In four-wave interaction the light wave subject to WFR and one of the reference waves induce a spatial grating of perturbations of the refractive index Δn, from which the second reference wave, directed counter to the first, is scattered into a wave that is reversed relative to the first.

The experiments performed, however, dealt with certain singularities of the interaction of opposing waves only in cubic media. In such media the spatial period Λ of the Δn grating can be substantially larger as well as comparable with the length λ of the light wave. For example, when low-frequency Δn gratings are excited, owing to the interaction of the waves making small angle θ with one another, we have $\Lambda=\lambda/\theta$, and owing to the interaction of the transverse wave we have $\Lambda=\lambda/2$.

The excitation of a grating with a period $\Lambda\gg\lambda$ in inertia-less cubic media can lead to development of instabilities typical of self-focusing,[7,9] and this leads to distortions in the spatial and temporal structures of the reversed wave. On the other hand, if the interaction of the waves at small angles is negligible, then these large-scale gratings are not excited and the corresponding instabilities likewise fail to develop. An example of a process for which it is possible to suppress the self-focusing instabilities in this manner is the four-wave transformation of opposing light beams under conditions of two-photon resonance, which will be investigated below.

To clarify these conditions we consider two waves with close frequencies, propagating at small angles to each other. Assume that another pair of waves is directed counter to them. Let the sum or difference of the frequencies of the individual waves in the different pairs coincide, and let the sum within each pair not coincide with any of the frequencies of the natural oscillations of the medium. Then the interaction of the co-moving waves, due to the two-photon resonance, is insignificant. At the same time, when the corresponding waves from the opposing pairs interact, a low-frequency (when the frequencies are subtracted) or a high-frequency (when they are added) grating Δn is produced, as a result of scattering by which energy can be transferred from some waves to the others. If such an energy transfer takes place for all four waves with participation of one and the same grating Δn, then a definite relation exists between the phases of these waves and leads in final analysis to effects of the WFR type. Depending on what causes the excitation of the grating—subtraction or addition the frequencies—the discussed process reduces to Raman conversion (RC) or two-photon absorption (TA) of one pair of beams in the field of the other (opposing) pair. WFR reversal in RC was observed by us earlier,[9,10] and a corresponding theoretical analysis (as applied to stationary and nonstationary processes) was carried out there in the linear approximation (the fields of the reference waves were assumed given). As to WFR in the case of TA, although this process has not yet been observed experimentally, nonetheless its analysis is important for a clarification of the possibilities of infrared WFR, particularly at CO_2 laser wavelengths.

In the present paper we use the method of averaging over nonsinusoidal waves to investigate the WFR processes in the interaction of opposing light beams. We use these methods to carry out the second truncation of the nonlinear quasi-optical equations. In the approach used by us, the basis functions are defined in such a way that when the averaging conditions are satisfied and the boundary conditions are given the expansion of the fields of the light waves in terms of these functions contains a minimum number of terms. As a result it is possible to develop a nonlinear theory that makes it possible (in the absence of wave detuning) to investigate analytically the WFR process of multimode light beams with account taken of the spatial change of all the interaction waves. It is shown for the first time

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ever for process of similar type, that the coefficient R of conversion into the reversed wave can greatly exceed unity in the absence of self-focusing instabilities. The corresponding conditions were realized experimentally and a conversion coefficient $R\approx 7$ was obtained. In addition, the possibility of controlling the frequency and waveform of the pulse of the inverted wave was demonstrated experimentally, as well the possibility of WFR in a medium with strongly inhomogeneous fluctuations of the refractive index.

2. SECOND TRUNCATION OF NONLINEAR QUASI-OPTICAL EQUATIONS

In the general case, the WFR is based on the interaction of two opposing light waves with complex space-time structure. We examine these processes in greater detail using an example of the RC of a multimode multifrequency optical radiation.

Assume that two light waves $\mathscr{E}_+$ and $\mathscr{E}_-$, with average frequencies ω_+ and ω_- that differ by an amount equal to the stimulated-scattering (SS) shift Ω_0 characteristic of the given medium, are incident on a layer of Raman-active medium of length l:

$$\mathscr{E}_+=E_+\exp(i\omega_+t-ik_+z)+\text{c.c},\qquad \mathscr{E}_-=E_-\exp(i\omega_-t+ik_-z)+\text{c.c}\,. \tag{1}$$

Here $\omega_+-\omega_-=\Omega_0, k_\pm=\omega_\pm/v_\pm, E_\pm$ are the complex amplitudes that satisfy in the quasi-optical approximation the following truncated equations

$$\nu_\pm\frac{\partial E_\pm}{\partial\eta}\pm\frac{\partial E_\pm}{\partial z}+\frac{i}{2k_\pm}\Delta_\perp E_\pm-\frac{ik_\pm\delta n}{2n}E_\pm=-\frac{ig_{1,2}}{2}\begin{Bmatrix}E_-Q\\ E_+Q^*\end{Bmatrix} \tag{2}$$

$$\hat{M}_{\text{av}}\left(\frac{\partial}{\partial\eta},\nabla\right)Q=-ig_3E_+E_-^*, \tag{3}$$

where

$$\eta=t-z/v_+,\quad \nu_+=0,\quad \nu_-=v_+^{-1}+v_-^{-1},$$

δ_n are the perturbations of the linear part of the refractive index, and $\hat{M}_{\text{av}}$ is an operator describing the change of the phonon coordinate Q and averaged over its high-frequency oscillations in space and in time:

$$\hat{M}_{\text{av}}=\langle\exp\{-i(\Omega_0t-qz)\}\hat{M}\exp\{i(\Omega_0t-qz)\}\rangle,\quad q=k_++k_-$$

(to simplify the exposition that follows, the phonon wave Q is assumed to be scalar, corresponding, for example, to Mandel'shtam-Brillouin scattering by hypersound or to Raman scattering by molecules with fully symmetrical vibrations).

Equations (2) and (3) retain the same form also in the case of TA in the field of opposing waves, the only difference being that in this case $\Omega_0=\omega_++\omega_-$ (but $2\omega_\pm\neq\Omega_0$), and the right-hand sides of the equations are replaced respectively by QE_-^*, QE_+^*, and E_+E_-. Although in this section the derivation is carried out only as applied to RC, there are no difficulties of principle in transferring these arguments to the case of TA. Therefore, wherever it is of interest, we shall write down the equations pertaining to TA without a detailed derivation.

We assume the frequencies ω_+ and ω_- to be close, so that we can neglect the difference of the wave vectors in Eq. (2) and put $k_+=k_-=k$. This can be done if the length of the layer L does not exceed the amplitude $(|k_+=k_-|\theta_\pm^2)^{-1}$, where $\theta_\pm$ are the angular divergences of the interacting beams. Assume that the fields E_+ and E_- have a strongly inhomogeneous transverse structure and the constant average intensity. Then, so solve Eq. (2) and (3) we use an approach whose gist consists in the assumption that at any plane z in the fields E_+ and E_- with the largest weight are represented sets of N orthogonal structures (modes) $\mathscr{E}_j$ and $\mathscr{E}_j^*$, satisfying the linear quasi-optical equations

$$\left(\frac{\partial}{\partial z}-\frac{i}{2k}\Delta_\perp-\frac{ik\delta n}{2n}\right)\mathscr{E}_j=0,\quad\left(\frac{\partial}{\partial z}+\frac{i}{2k}\Delta_\perp+\frac{ik\delta n}{2n}\right)\mathscr{E}_j^*=0, \tag{4}$$

$$\int\mathscr{E}_j\mathscr{E}_k^*\,dS=S\delta_{kj}, \tag{5}$$

where S is the area of the cross section of the interaction volume. In this case E_+ and E_- take the form

$$E_+=\sum_{j=1}^{N}c_j^+(z,\eta)\mathscr{E}_j(z,\mathbf{r}_\perp)+\tilde{E}_+,\quad E_-=\sum_{j=1}^{N}c_j^-(z,\eta)\mathscr{E}_j^*(z,\mathbf{r}_\perp)+\tilde{E}_-, \tag{6}$$

$$\int\tilde{E}_+\mathscr{E}_j^*\,dS=0,\quad\int\tilde{E}_-\mathscr{E}_j\,dS=0. \tag{7}$$

The representation (7) means that the solution of the nonlinear equations for the fields $E_\pm$ is sought in the form close to the solution of the corresponding linear problem, as is typical of the procedure of the obtaining the truncated equations.

It is convenient to choose the modes $\mathscr{E}_j$ and $\mathscr{E}_j^*$, for example, in such a way that at finite N the input values of the fields $E_+(0,r_\perp)$ and $E_-(L,r_\perp)$ can be expanded in its terms in a series with a minimum residual term. This can be attained if we stipulate that the expansion coefficients $c_j^+(t)$ satisfy the condition

$$\int_T c_j^+(t)c_k^+(t)\,dt=W_j\delta_{jk},$$

where T is the orthogonal scale and is equal to or less than the duration of the pulse. We next obtain for the field $E_+(0,r_\perp,t)$ we obtain the so-called canonical expansion of Kaarunen and Loew.[11] It is next necessary to separate in the opposing wave $E_-(L,r_\perp,t)$ the projections on the obtained unit vectors $\mathscr{E}_j^*(L,r_\perp)$ and $c_j^+(t)$, and represent the remaining part of the field again in the form of a canonical expansion. As a result of this procedure we obtain all the modes $\mathscr{E}_j$ and $\mathscr{E}_j^*$ of interest to us. If at a finite number of modes N, the residual term is negligibly small, then the corresponding $\tilde{E}_\pm$ which are orthogonal to the separated structures $\mathscr{E}_j$ and $\mathscr{E}_j^*$, vanish on the boundaries $z=0$ and $x=L$, and $\tilde{E}_+(0,r_\perp)=\tilde{E}_-(L,r_\perp)=0$ and can become different from zero only as a result of the interaction of the waves within the medium.

We now solve Eq. (3) relative to the coordinate Q:

$$Q=g_3\int_0^\eta d\eta'\,G(\eta-\eta')E_+(\eta')E_-^*(\eta'), \tag{8}$$

where $G(\eta)$ is the Green's function of the equation $\hat{M}_{yc\phi}G(\eta)=\delta(\eta)$, and substitute (6) and (8) in (2). Using the orthogonality condition (5) and (7) and the fact that the functions $\mathscr{E}_j$ satisfy Eq. (4), we obtain a system of equations equivalent to (2) and (3) (we do not write out here the equations for $\tilde{E}_\pm$ because they are too cumbersome):

$$\frac{\partial}{\partial z}c_j^+=\frac{g_1g_3}{2}\int_0^\eta d\eta' G(\eta-\eta')\sum_{k,n,m=1}^{N}A_{jknm}c_k^-(z,\eta)c_n^+(z,\eta')c_m^{-*}(z,\eta')$$
$$+f_+(\tilde E_+,\tilde E_-),\qquad (9)$$

$$\left(v_-\frac{\partial}{\partial\eta}-\frac{\partial}{\partial z}\right)c_j^-=\frac{g_2g_3}{2}\int_0^\eta d\eta' G(\eta-\eta')\sum_{k,n,m=1}^{N}\dot A_{jknm}c_k^+(z,\eta)c_n^{+*}(z,\eta')$$
$$\times c_m^-(z,\eta')+f_-(\tilde E_+,\tilde E_-).\qquad (10)$$

Here $f_\pm(\tilde E_+,\tilde E_-)$ denote the terms proportional to $E_\mp$,

$$A_{jknm}=\int \mathscr{E}_j^*\mathscr{E}_k^*\mathscr{E}_n\mathscr{E}_m\,dS\Big/\int|\mathscr{E}_j|^2\,dS$$

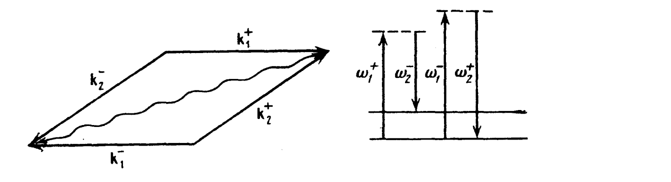
are the overlap integrals of the separated structures $\mathscr{E}_j$. To calculate A_{jknm} and to estimate the components $\tilde E_\pm$, we assume that the structures $\mathscr{E}_j$ are statistically independent, are strongly inhomogeneous in $r_\perp$, and their intensity averaged over a scale much larger than the radius ρ_j of the transverse correlation is constant in space. It is natural to assume here that the components of the matrix A_{jknm} differ from zero only for $j=n$, $k=m$, or $j=m, k=n$, and the remaining ones are sufficiently small, so that at the normal distribution law of the structures $\mathscr{E}_j$ the following representation is approximately valid.

$$A_{jknm}=A_{jkjk}\delta_{jn}\delta_{km}+A_{jkkj}\delta_{jm}\delta_{kn}-A_{jjjj}\delta_{jk}\delta_{jn}\delta_{jm}.\qquad (11)$$

To estimate the fields $\tilde E_\pm$ and their contributions to the interaction of the separated structures $\mathscr{E}_j$, we can proceed, e.g., as in problems of wave propagation in strongly inhomogeneous media in the Markov random-process approximation.[12] The indicated procedure was not used earlier to analyze the WFR in stimulated scattering.[13] Estimates show that the contribution of the fields $\tilde E_\pm$ in Eqs. (10) and (11) is negligible if the change of each of the coefficients $c_j^\pm$ is negligible over the correlation length z_{jk} of the $\mathscr{E}_j$ mode with all the modes making the main contribution to the field E_+. The value of z_{jk} is determined by the minimum of two quantities: the longitudinal interaction length $(k\theta_{jk}^2)^{-1}$ of the modes $\mathscr{E}_j$ and $\mathscr{E}_k$ (θ_{jk} is the average angle between the directions of their propagation) and the smallest of the correlation lengths $k\rho_j^2$ or $k\rho_k^2$ for each of these modes separately. In particular, at a large number of modes $\mathscr{E}_j$ with approximately equal intensities and random phases on entering the medium, each of them can have a plane wave front, but the average angle between modes should correspond to the condition indicated above.[14,15]

Assuming the condition that the fields $\tilde E_\pm$ to be satisfied and putting in (10) and (11) the terms $f_\pm(\tilde E_+,\tilde E_-)=0$, we obtain for the coefficients $c_j^\pm(z,\eta)$, with allowance for (12), a closed system of equations. On the basis of this system we can investigate various singularities of the interaction of the multicomponent waves E_+ and E_-, including the phenomenon of the WFR in stimulated scattering.[16]

In this paper, however, we limit ourselves to an analysis of the conversion of only two pairs of opposing modes. Let for the sake of argument the wave E_+ consist of two linearly polarized components $e_1c_1^+\mathscr{E}_1$ and $e_2c_2^+\mathscr{E}_2$, $(e_1,e_1)=(e_2,e_2)=1$, the first of which has a frequency coinciding with ω_1^+, and the second either a different frequency ω_2^+, or an orthogonal polarization e_2. We assume also that the opposing wave E_- also has two



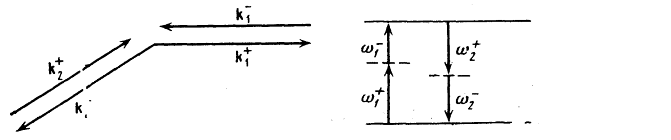


FIG. 1. Diagram of energy levels and wave vectors, illustrating the four-wave interaction in Raman conversion (a) and in two-photon absorption (b).

components $e_2c_1^-\mathscr{E}_1^*$ and $e_1c_2^-\mathscr{E}_2^*$ respectively with frequencies $\omega_1^-=\omega_2^+-\Omega$ and $\omega_2^-=\omega_1^+-\Omega$, shifted into the Stokes region by an amount $\Omega=\Omega_0-\delta\Omega$ (here $\delta\Omega$ is the detuning from exact resonance with natural oscillations of the medium). The diagram of the wave vectors corresponding to the interaction of the waves $e_{1,2}c_{1,2}^+\mathscr{E}_{1,2}$ and $e_{2,1}c_{1,2}^-\mathscr{E}_{1,2}^*$ is shown in Fig. 1a.

Taking into account the frequency detunings we make the substitution

$$c_1^+(z,\eta)=c_1^+(z),\quad c_1^-(z,\eta)=c_1^-(z)\exp\{i(\delta\omega+\delta\Omega)\eta+2i\delta k_2z\},$$
$$c_2^+(z,\eta)=c_2^+(z)e^{i\delta\omega\eta},\quad c_2^-(z,\eta)=c_2^-(z)\exp\{i\delta\Omega\eta+2i\delta k_1z\},$$
$$\delta\omega=\omega_1^+-\omega_2^-,\quad \delta k_1=\delta\Omega/v,\quad \delta k_2=(\delta\omega+\delta\Omega)/v,\quad v=v_+=v_-$$

and in the stationary case we obtain from (9) and (10) the following equations

$$\frac{dc_{1,2}^+}{dz}=-\frac{g}{2}\left\{\frac{A_{1212}}{1+i\delta\Omega/\gamma}[|c_{2,1}^-|^2c_{1,2}^++c_{2,1}^+c_{2,1}^-c_{1,2}^-e^{\mp i\delta kz}]\right.$$
$$\left.+\frac{A_{1111,2222}(e_1,e_2)}{1+i(\delta\omega+\delta\Omega)/\gamma}|c_{1,2}^-|^2c_{1,2}^+\right\},\qquad (12)$$

$$\frac{dc_{1,2}^-}{dz}=-\frac{g}{2}\left\{\frac{A_{1212}}{1-i\delta\Omega/\gamma}[|c_{2,1}^+|^2c_{1,2}^-+c_{2,1}^+c_{2,1}^-c_{1,2}^+e^{\mp i\delta kz}]\right.$$
$$\left.+\frac{A_{1111,2222}(e_1,e_2)}{1-i(\delta\omega+\delta\Omega)/\gamma}|c_{1,2}^+|^2c_{1,2}^-\right\},\qquad (13)$$

where

$$\delta k=2\delta\omega/v,\quad g=g_1g_3/\gamma\quad (g_1=g_2).$$

To estimate the influence of the last term in the right-hand sides of (12) and (13) at $\delta\omega\sim\gamma$ in the case $e_1\|e_2$, we assume that the power of the waves $c_1^\pm$ greatly exceeds the power of the weak components $e_2^\pm$. Then the change of c_1^+ and c_1^- along z is obtained from the solutions (12) and (13) at $c_2^+=c_2^-=0$. The product c_1^+ and c_1^-, which determines the coefficient of the parametric interaction of the weak components c_2^+ and c_2^-, is expressed in the form (at $\delta\Omega=0$)

$$c_1^+c_1^-=|c_1^+(z)c_1^-(z)|\exp\left\{igA_{1111}\frac{d\delta\omega/\gamma}{1+(\delta\omega/\gamma)^2}z\right\},$$

where $d=|c_1^+|^2-|c_1^-|^2=\text{const}$.

Thus, the nonlinear interaction of the waves c_1^+ and c_1^- at $\delta\omega\neq 0$ leads to the appearance of an additional detuning in the equations for c_2^+ and c_2^-

$$\delta k_{\text{eff}}=\delta k+gA_{1111}d\frac{\delta\omega/\gamma}{1+(\delta\omega/\gamma)^2}.\qquad (14)$$

The nonlinear increment to the wave detuning first increases with increasing $\delta\omega$, which should lead to a lowering of the effectiveness of the parametric conversion. With further increase of $\delta\omega$, it decreases and at $\delta\omega \gg \gamma$ it becomes negligibly small.

In the case $\delta\omega \gg \gamma$ we can neglect the interaction of the waves connected with the last terms of (12) and (13), and assumed that $A_{1212}=1$, using for the description of the RC the following system of equations

$$\frac{d}{dz}c_{1,2}^{+}=-\frac{g}{2(1+i\delta\Omega/\gamma)}[|c_{2,1}^{-}|^2c_{1,2}^{+}+c_{2,1}^{+}c_{2,1}^{-}c_{1,2}^{-*}\,e^{\mp i\delta kz}], \quad (15)$$

$$\frac{d}{dz}c_{1,2}^{-}=-\frac{g}{2(1-i\delta\Omega/\gamma)}[|c_{2,1}^{+}|^2c_{1,2}^{-}+c_{2,1}^{+}c_{2,1}^{-}c_{1,2}^{+*}\,e^{\mp i\delta kz}]. \quad (16)$$

Similar equations can be written also for the TA in the field of two pairs of opposing waves $c_{1,2}^{+}\mathscr{E}_{1,2}$ and $c_{1,2}^{-}\mathscr{E}_{1,2}^{*}$ with respective frequencies ω_1^+, ω_2^+ and $\omega_1^-=\Omega-\omega_1^+, \omega_2^-=\Omega-\omega_2^+$ $(\Omega=\Omega_0+\delta\Omega, (\delta\omega-\delta\Omega)\gg\gamma)$:

$$\frac{d}{dz}c_{1,2}^{+}=-\frac{g}{2(1+i\delta\Omega/\gamma)}[A_{1111,2222}|c_{1,2}^{-}|^2c_{1,2}^{+}+c_{2,1}^{+}c_{2,1}^{-}c_{1,2}^{-*}\,e^{\mp i\delta kz}], \quad (17)$$

$$\frac{d}{dz}c_{1,2}^{-}=+\frac{g}{2(1+i\delta\Omega/\gamma)}[A_{1111,2222}|c_{1,2}^{+}|^2c_{1,2}^{-}+c_{2,1}^{+}c_{2,1}^{-}c_{1,2}^{+*}\,e^{\mp i\delta kz}]. \quad (18)$$

The wave-vector diagram illustrating the interaction of the waves in this case is shown in Fig. 1b.

3. NONLINEAR DYNAMICS OF THE PROCESS OF FOUR-WAVE CONVERSION

We proceed to an investigation of Eqs. (15), (16) and (17), (18) which describe the conversion of the wave $c_2^-\mathscr{E}_2^*$ into a wave with reversed front $c_2^+\mathscr{E}_2$ in the field of two opposing complex-conjugate waves $c_1^+\mathscr{E}_1$ and $c_2^-\mathscr{E}_1^*$. We write down first the solution of these equations in the approximation where the values of c_1^+ and c_1^- are given, assuming that the wave subjected to the WFR is a sufficiently weak wave $c_2^-(L)\mathscr{E}_2^*(L, r_1)$ specified on the boundary $Z=L$, and there is no wave $c_2^+\mathscr{E}_2$ on the boundary $z=0$. Then the amplitude coefficients of the conversion in the reversed wave $R_a=c_2^+(L)/c_2^-(L)$ does not depend on the incident wave C_2^-. For RC in the absence of wave detuning $(\delta k<\min(g|c_1^{\pm}|^2, L^{-1}))$ we have

$$R_a=c_1^+c_1^-\left[1-\exp\left(\frac{1}{2}g(\delta\Omega)L|c_1^+|^2+\frac{1}{2}g^*(\delta\Omega)L|c_1^-|^2\right)\right]$$

$$\cdot\left[\frac{g(\delta\Omega)}{g^*(\delta\Omega)}|c_1^+|^2+|c_1^-|^2\exp\left(\frac{1}{2}g(\delta\Omega)L|c_1^+|^2+\frac{1}{2}g^*(\delta\Omega)L|c_1^-|^2\right)\right]^{-1},$$

$$g(\delta\Omega)=g/(1-i\delta\Omega/\gamma). \quad (19)$$

As seen from (19), at $\delta\Omega\neq 0$ there exist values of $|c_1^-|^2$ and $|c_1^+|^2$ at which $R_a\to\infty$. These values are shown in Fig. 2 for $g>0$. The reason why the conversion coefficient becomes infinite is that upon detuning from resonance the phonon wave has a phase such that the wave is excited not only by the pair of waves c_1^+ and c_1^- but also by the pair c_2^+ and c_2^-. In this case the reversal process becomes unstable and both waves c_2^+ and c_2^- are generated in the field of the opposing waves c_1^+ and c_1^-, even if their values on the boundaries $z=0$ and $z=L$ were equal to zero.

At $\delta\Omega=0$, the reversal coefficient is always finite, but can exceed unity substantially. Thus, at $g>0$ the maximum value $R_{\max}=|R_{a\max}|^2=\frac{1}{4}e^{M/2}$ is reached when $|c_1^-|^2=|c_1^+|^2e^{-M/2}$, and at $g<0$ it is reached when $|c_1^-|^2=|c_1^+|^2e^{+M/2}$ $(M=g(|c_1^-|^2+|c_1^+|^2)$.

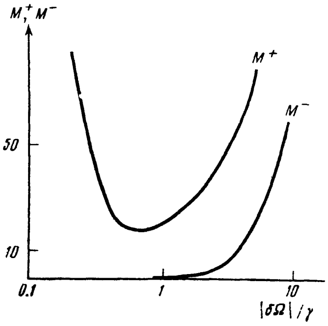


FIG. 2. Dependence of the values of the increments $M^+=g|c_1^+|^2L$ and $M^-=g|c_1^-|^2L$, at which the coefficient R becomes infinite on the normalized detuning.

In the case of TA, in the absence of wave detuning δk, the value of R_a

$$R_a=\operatorname{tg}\left\{\frac{1}{2}\int_0^L g(\delta\Omega)c_1^+c_1^-dz\right\}. \quad (20)$$

It is easily seen that R_a remains finite at finite wave amplitudes c_1^+ and c_1^-. The reason is that as a result of the two-photon absorption

$$\frac{1}{2}\int_0^L g(\delta\Omega)c_1^+c_1^-dz\to\frac{\pi}{2}$$

only asymptotically with increasing values of c_1^+ and c_1^- on the boundaries of the medium.

Figure 3 shows (at $\delta\Omega=0$) the values of the conversion coefficient for two cases: $c_1^-(L)=c_1^+(0)$ (curve a) and $c_1^-(L)=c_1^+(L)$ (curve b). The latter boundary condition is realized when the opposing wave $c_1^-\mathscr{E}_1^*$ is produced by reflection of the $c_1^+\mathscr{E}_1$ wave from of a wave-front-reversing mirror.

To ascertain the dynamics of the processes under saturation conditions, we obtain the nonlinear solutions of Eqs. (15), (16) and (17), (18) at exact resonance $(\delta\Omega=0)$ and in the absence of wave detuning $(\delta k=0)$. If we change over in Eqs. (15), (16) and (17), (18) to the amplitudes $|c_j^{\pm}|$ and to the phases $\varphi_j^{\pm}$ then it is easily seen that under the condition $c_2^+(0)=0$ the phase differ-

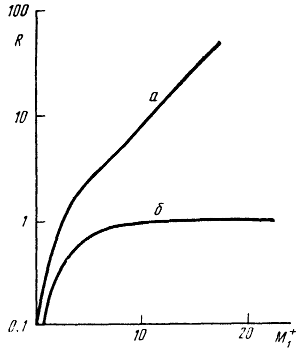


FIG. 3. Dependence of the coefficient $R=|R_a|^2$ on the power of the pump wave $M^+=g|c_1^+(0)|^2L$ for different boundary conditions.

ence $\Phi=\varphi_1^+ + \varphi_1^- - \varphi_2^+ - \varphi_2^-$ settles at a value π. We then obtain for the amplitudes $|c_j^\pm|$ equations in closed form, which have the following first integrals (in the TA case it is assumed that $A_{1111}=A_{2222}=1$): for the RC case

$$|c_1^+|^2-|c_2^-|^2=J_1,\quad |c_1^-|^2-|c_2^+|^2=J_2,\quad |c_1^+c_1^-|-|c_2^+c_2^-|=J_3. \tag{21}$$

For the TA case

$$|c_1^+|^2+|c_1^-|^2=J_1,\quad |c_2^+|^2+|c_2^-|^2=J_2,\quad |c_1^+c_2^-|+|c_1^-c_2^+|=J_3. \tag{22}$$

Using these relations, we obtain first-order differential equations whose solution at nonzero values of $|c_1^+(0)|$, $|c_1^-(L)|, c_2^-(L)$ leads to a transcendental equation for $|c_1^+(L)|$. For the RC this equation takes the form

$$\frac{[b+(b^2-a^2)^{1/2}]\zeta(L)+a}{a\zeta(L)+b+(b^2-a^2)^{1/2}}=\frac{[b+(b^2-a^2)^{1/2}]\zeta(0)+a}{a\zeta(0)+b+(b^2-a^2)^{1/2}}\exp\left[\frac{1}{2}gL(b^2-a^2)^{1/2}\right], \tag{23}$$

where $\zeta=|c_2^-|/c_1^+|$. At $g>0$

$$a=-2|c_1^+(L)|^2|c_2^-(L)|(|c_1^+(L)|^2-|c_2^-(L)|^2)^{-1},$$
$$b=|c_1^+(L)|^2-|c_2^-(L)|^2(|c_1^+(L)|^2+|c_2^+(L)|^2)(|c_1^+(L)|^2-|c_2^-(L)|^2)^{-1},$$

and for $g<0$

$$a=-2|c_1^+(0)||c_2^-(0)||c_1^-(0)|^2(|c_1^+(0)|^2-|c_2^-(0)|^2)^{-1},$$
$$b=|c_1^+(0)|^2-|c_2^-(0)|^2+|c_1^-(0)|^2(|c_1^+(0)|^2-|c_2^-(0)|^2)(|c_1^+(0)|^2-|c_2^-(0)|^2)^{-1}.$$

For the TA case

$$\frac{bR_a+a}{b+R_a a}=\frac{a}{b}\exp\left[-\frac{1}{2}gL(a^2+b^2)^{1/2}\right], \tag{24}$$

where

$$a=-2|c_1^-(0)||c_1^+(0)|,\quad b=|c_2^-(0)|^2-|c_1^-(0)|^2.$$

Figures 4 and 5 show the values of $R=|R_a|^2$ for the RC and TA respectively and for different values of the power of the wave $c_2^-(L)$ subject to WFR. The obtained solutions in the absence of wave detuning and at exact resonance complete the investigation of the stationary regime of the corresponding four-photon conversion processes.

4. EXPERIMENTAL INVESTIGATION OF FOUR-WAVE RAMAN CONVERSION

An experimental investigation of the WFR phenomenon in four-wave interaction under conditions of two-

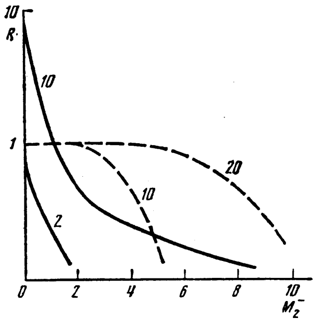


FIG. 5. Dependence of the coefficient R on the intensity of the incident wave $M_2^-=g|c_2^-|^2L$ at different values of the pump wave intensity $M_1^+=g|c_1^+|^2L$ (numbers on the curves). The solid curves pertain to the boundary condition $c_1^+(0)=c_1^-(L)$, and the dashed ones to $c_1^+(L)=c_1^-(L)$.

photon resonance was carried out for SMBS conversion of a Stokes wave into the anti-Stokes component in the field of the opposing pump waves (see Fig. 6). The WFR process proceeds in this case in the following manner. The pump wave $c_1^-\mathscr{E}_1^*$ is scattered into the reversed wave $c_2^+\mathscr{E}_2$ by a hypersonic grating excited by a pump $c_1^+\mathscr{E}_1$ and a signal wave $c_2^-\mathscr{E}_2^*$, the latter having with respect to the wave $c_1^+\mathscr{E}_1$ a Stokes shift equal to the hypersound frequency (Fig. 1a). The pump $c_1^+\mathscr{E}_1$ was the second harmonic of a neodymium-glass laser ($\lambda=0.53$ μm, $\tau_{\text{pulse}}=25$ nsec, $d=3$ mm, $P=1$–10 MW). The opposing pump wave $c_1^-\mathscr{E}_1^*$ was produced by reflection of the wave $c_1^+\mathscr{E}_1$ either from a flat mirror 3 (in the case of plane pump waves) or from a SMBS mirror (a lens that focuses the radiation into a cell with a scattering medium). The two pump waves intersected in a cell 1 ($L=5$ cm) with acetone. Part of the radiation diverted from the pump $c_1^+\mathscr{E}_1$ was focused into an additional cell 2, also filled with acetone, where the SMBS was excited. The Stokes wave, which had the required phase shift Ω_0 relative to the pump wave $c_1^+\mathscr{E}_1$, served as the $c_2^-\mathscr{E}_2^*$ was subject to the WFR.

For convenience in registration of the WFR effect, a phase plate (PP) was placed in the path of the $C_2^-\mathscr{E}_2^*$ wave; this was a glass plate etched in fluoric acid and increasing the beam divergence to $\theta=1.2\times10^{-2}$ rad. The quantity registered in the experiment was the distribution of the intensity in the near field and the angular spectrum of the anti-Stokes beam, as well as oscillograms of the Stokes and anti-Stokes pulses.

After passing through the PP, the radiation from the WFR again became single-mode and its angle sepctrum duplicated the spectrum of the initial Stokes beam.

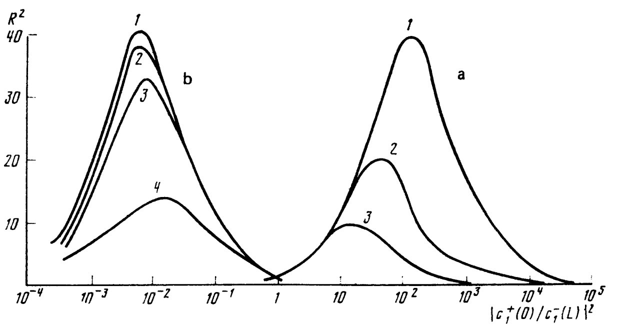


FIG. 4. Dependence of the coefficient $|R|^2$ on the ratio of the pump intensities $|c_1^+(0)|^2/|c_1^-(L)|^2$ for different values of the incident wave intensity $c_2^-(L)$ for the cases $g>0$ (a) and $g<0$ (b). The fixed values are $g|c_1^+(0)|^2L=10$, for case a and $g|c_1^-(L)|^2L=10$ for case b. The curves are marked by the values of the power of the incident wave $|c_2^-(L)/c_1^+(0)|^2$ (a) and $|c_2^-(L)/c_1^-(L)|^2$ (b): 1—4.2×10^{-4}, 2—1.1×10^{-2}, 3—3.3×10^{-2}, 4—0.14.

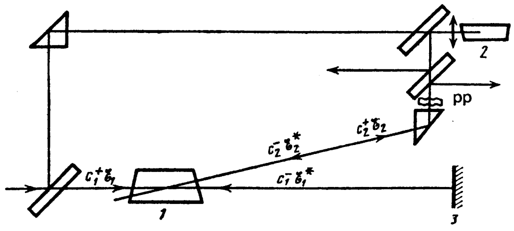


FIG. 6. Block diagram of experimental setup.

Mirror	Acetone	Carbon tetra-chloride	Water	Flat mirror
$\lvert\nu_1-\nu_3\rvert/\gamma=\lvert\delta\omega\rvert/\gamma$	0	2.1	8.7	37.5
Relative pump power	1	4.1	1.3	1.5

The effectiveness of the WFR process depended on the wave detuning $\delta k = 4\pi n\delta\nu$ (cm^{-1}), where $\delta\nu = \nu_1 - \nu_3$ is the difference of the SMBS shifts in cells 1 and 2 respectively, and n is the refractive index. The coefficient of conversion R into the reversed wave was largest at $\delta\nu = 0$. At a small difference between the frequency of the opposing pump and the frequency of the wave $c_2^-\mathscr{E}_2^*$ subject to the WFR, the conversion effectiveness first decreased rapidly, and then again increased (see the table, which shows data on the relative values of the pump corresponding to one and the same value of the intensity of the inverted wave $c_2^+\mathscr{E}_2$ when the role of the mirror 3 is assumed by SMBS cells with different frequency shifts). This change in the effectiveness of the conversion took place under conditions when the linear detuning $\delta k = 4\pi n\delta\nu$ was small compared with the local increment $g|c_1^+|^2$. The considerable decrease of the effectiveness of conversion at $\delta\nu = 0.012$ cm^{-1} is due to the fact that in this case the ratio $\delta\omega/\gamma \sim 2$ and the wave detuning (14) increased to a value comparable with that of the local increment.

In the case of plane pump waves, the WFR effect was observed if the wave $c_1\mathscr{E}_1$ was reflected both by a flat mirror and by SMBS mirrors with different frequency shifts. However, if the wave $c_1\mathscr{E}_1$ is passed through a PP, then when the wave is reflected by an ordinary mir-mirror the WFR effect disappeared (there was no reflected anti-Stokes signal at all in the case of sufficiently small-scale modulation of the pump). On the other hand, if the reflection was produced by an SMBS mirror in such a way that the opposing pump turned out to be complex conjugate to $c_1\mathscr{E}_1$, then the WFR of the $c_2^-\mathscr{E}_2^*$ wave was quite distinctly observed.

Thus, the use of a SMBS mirror makes it possible to effect WFR in four-wave interactions in multimode pump beams. This result is of interest for the realization of a WFR scheme in a cubic medium, where introduction of small-scale modulation into the pump beam suppresses the self-focusing effect,[17,18] a fact that can increase the coefficient of reflection into the inverted wave.

Inhomogeneities can be introduced into the light beams not only by phase plates, but also by the very medium in which nonlinear interaction takes place. In the experiment, inhomogeneities of the refractive index were produced artificially, by heating the cell 1. The convection of the liquid led in this case to strong large-scale distrotions of the pump beam passing through the cell. If the acetone was heated to the boiling point, then passage of the laser pulse caused, apparently as a result of heating of the microinclusions, strong boiling in the entire volume. Small-scale scattering of the pump radiation was then observed, as well as an increase of the beam divergence. Nonetheless, when the pump $c_1\mathscr{E}_1$ was reflected by an SMBS mirror, WFR of the Stokes wave was reliably registered in both cases, although in a number of flashes its quality was slightly worse. Thus, in our scheme the WFR can be realized not only with strongly inhomogeneous pump beams, but also in an optically inhomogeneous medium.

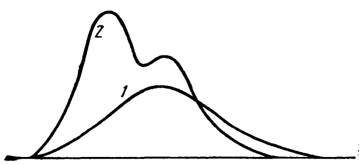


FIG. 7. Characteristic waveforms of the pulses of the incident Stokes wave 1 and of the reversed anti-Stokes wave 2.

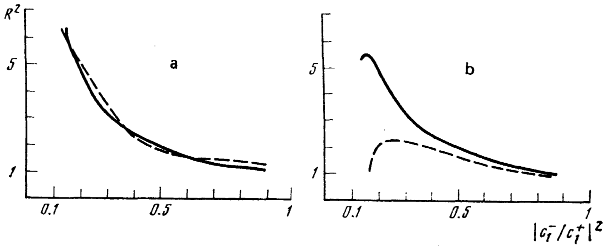


FIG. 8. Experimental (dashed) and theoretical (solid) plots of $|R|^2$ on the pump-wave intensity ratio at $|c_{1\max}^-|^2/|c_{1\max}^+|^2 = 0.35$ and at the values of the increment $g|c_{1\max}^+|^2L$, equal to 20 (a) and 13 (b).

Other important features of the investigated process are the possibility of controlling the parameters of the radiation with the WFR, and of obtaining a larger-than-unity coefficient of conversion into the reversed wave. Thus, for example, the shape and duration of the pulse as well as the conversion coefficient R can be controlled by varying the ratio of the intensities of the pump waves. In the experiments, the pump pulses were delayed relative to each other, and consequently the conversion coefficient varied with time and, as seen from Fig. 7, the shape of the pulse of the reversed wave differed strongly from that of the incident wave. Since the ratio c_1^-/c_1^+ varied with time, we were able to measure $R(c_1^-/c_1^+)$ in one laser shot. The measured experimental plots of $R(c_1^-/c_1^+)$ for different values of $gL|c_{1\max}^+|^2$ are in qualitatively good agreement with the theoretical plots based on formula (23) for Gaussian pump pulses with durations and delays that are in accord with the experiment. Figure 8 shows by way of example one of these plots. The maximum conversion coefficient measured in the experiment was $R = 7$.

We note in conclusion that in WFR as a result of four-photon interaction it is possible also to control other parameters of the reversed radiation—the polarization, duration, and waveform of the pulse, the carrier frequency, and others. Experimentally, however, these features still remain little investigated (see Ref. 10 concerning some aspects of this problem).

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Translational nonequilibrium of a gas in a resonant optical field

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It is shown that a traveling electromagnetic wave can give rise to a strong translational nonequilibrium of a resonant impurity gas. Allowance is made for two nonequilibrium mechanisms, one of which is associated with a resonant radiation pressure and the other with the difference between the cross sections for elastic collisions of excited and normal atoms with a buffer gas. A study is made of kinetic, macroscopic, and optical manifestations of an induced translational nonequilibrium.

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1. INTRODUCTION

A translational nonequilibrium of a gas due to a resonant radiation pressure[1] has been investigated by us earlier.[2-4] A study of the diffusion of resonant atoms in the field of a traveling electromagnetic wave is reported in Ref. 4 with calculations of the macroscopic fluxes of atoms resulting in an inhomogeneous distribution of particles in a bounded region. It is assumed there implicitly that the elastic scattering frequencies are the same for excited and normal atoms. Gel'mukhanov and Shalagin[5] and Kalyazin and Sazonov[6] were the first to demonstrate the appearance of a flux of resonant atoms due to the difference between the scattering cross sections and to carry out calculations[5] under conditions such that the velocity distribution function of resonant particles differs little from the equilibrium form (homogeneous broadening).

We shall take into account each of these nonequilibrium factors, consider strongly nonequilibrium states of a gas (inhomogeneous braodening), and study their macroscopic manifestations.

2. DISTRIBUTION FUNCTION

In the quasiclassical limit the state of a resonant gas in the field of a traveling electromagnetic wave can be described by the kinetic equation for the density matrix in the mixed representation $\rho(\mathbf{r},\mathbf{v},t)$. We shall assume that the gas is sufficiently dilute and that the elastic scattering frequencies at the upper and lower levels ν_i are considerably less than the longitudinal and transverse relaxation rates $\nu_i \ll \gamma, \gamma_\perp$. In the case when the mass of a resonant atom is much greater than the mass of a buffer gas atom ($m \gg M$) the collision integrals can be written in the form

$$\mathrm{St}(\rho_{ii}) = \nu_i \left[\operatorname{div}_{\mathbf{v}} \mathbf{v}\rho_{ii} + \frac{u_0^2}{2}\frac{\partial^2\rho_{ii}}{\partial \mathbf{v}^2}\right] = \nu_i \hat{L}\rho_{ii}, \tag{1}$$

where u_0 is the most probable velocity of the resonant gas atoms. Then, the equations for the density matrix under steady-state conditions obtained ignoring the terms $\sim(\hbar k/mv)^2 \ll 1$ give the following kinetic equation for the distribution function of the atomic velocities $f = \mathrm{Tr}(\rho)$:

