STIMULATED RAMAN SCATTERING BY INFRARED-ACTIVE TRANSITIONS

B. A. AKANAEV, S. A. AKHMANOV, and Yu. G. KHRONOPULO

Moscow State University

Submitted November 2, 1967

Zh. Eksp. Teor. Fiz. 55, 1253–1265 (October, 1968)

A theory of stimulated Raman scattering (SRS) by transitions which do not obey the alternative selection rule is developed. The equations which take into account variation of the amplitudes and phases of all participating fields are derived and analyzed. Variation of the population along the direction of propagation of the waves is also taken into account. In this way the restrictions on the lifetimes of the excited particles corresponding to the optical interaction can be made apparent. On the one hand, such an analysis yields information on SRS under conditions of violation of the alternative selection rule when the rate of increase of the Stokes components and asymmetry of forward-backward SRS, etc., are affected. On the other hand, the phenomenon of SRS by infrared-active vibrations may be of importance in the problem of generation and transformation of infrared frequencies. The system considered may be an efficient infrared generator with optical pumping, as well as a converter of infrared into visible radiation. Quantitative estimates of the properties of such devices are given.

1. The purpose of this paper is the investigation of the interaction of the Stokes component of stimulated Raman scattering (SRS) with the electromagnetic wave of the scattered radiation and with a wave of frequency approximately equal to the frequency of the transition responsible for the SRS.

As is known,[1-3] such interaction can occur if this transition is not completely forbidden and in the case of interacting fields for which the condition of spatial synchronism can be fulfilled with sufficient accuracy. In this, as in the case of SRS by transitions that obey the alternative selection rule, there is an incoherent interaction (Raman), i.e., an interaction that is independent of the phase velocities of the waves; in the considered case there also arises a coherent (also called parametric) interaction of these fields. The latter can significantly influence the SRS process. In addition, there arises, as a result of this interaction under certain conditions, a wave of frequency

$$\omega_1 = \omega_{21} + \Delta = \omega_3 - \omega_2, \tag{1}$$

where ω_{21} is the frequency of the working transition, and ω_3 and ω_2 are respectively the frequencies of the incident radiation and the SRS Stokes component; the difference Δ is less than T^{-1}, the width of the absorption line for the transition 2--1 in the substance (see Fig. 1).

If optical levels are used for 1 and 2, this process is one of the means of converting the frequency of laser radiation downwards, in the given case to a frequency close to the frequency of the natural vibrations of the particles of the material system. As will be shown below, it is possible, by means of such a resonant conversion, to obtain, in principle, coherent radiation of considerable power in the frequency region where there are no powerful sources at the present time. It is also important that resonant conversion is possible in substances in which nonresonant parametric interaction of waves with frequencies ω_1, ω_2, and ω_3 is negligibly small.

In[1] generation of a resonant field and the SRS features that arise in this case were investigated in the approximation of a fixed pump field (frequency ω_3). However, this approximation does not permit obtaining a number of important characteristics of the considered process. In particular, one cannot determine the limiting magnitudes of the resonant and Raman fields, as well as the boundary values of the fields of frequency ω_3 and ω_2 and the parameters of the substance for which resonant parametric conversion is optimal. In this connection we derive and analyze below the equations describing the variation of the amplitudes and phases of all three interacting fields. We shall also take into account the variation in the population difference of the working levels in the direction of propagation of the waves; this permits finding the requirements on the lifetimes of the particles in the excited state which must be fulfilled in order that the conversion length be sufficiently small.

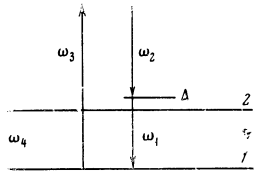

FIG. 1. Scheme for the considered nonlinear interactions. If a strong field at frequency ω_1 is applied to the medium, the system works as a generator of radiation at frequencies ω_1, ω_2. If the field at frequency ω_2 is strong, the system works as an upward frequency converter.

2. Thus, let us consider the interaction of three parametrically coupled traveling waves the frequencies of which satisfy the condition (1). We shall assume that the particles of the working substance interact with each other only through the radiation field. As a model of the working substance we shall have in mind an impurity in a dielectric. With these assumptions, in order to calculate the response of the substance at frequencies ω_l (l = 1, 2, 3) due to interaction of the fields

$$E_l e^{i\omega_l t} + \text{c.c.} \tag{2}$$

(here $E_l = A_l(z) \exp(-ik_l z)$, $k_l = 2\pi/\lambda_l$ is the wave vector of the l-th wave in the medium), we may use Eqs. (10) of[4] together with Eq. (1) from[5].

We limit the treatment to steady-state processes. We shall assume for simplicity that the medium is isotropic, the interaction of the substance with the field is dipolar, and the fields E_1, E_2, E_3 are polarized respectively along the directions a, b, c. Then the projections of the complex amplitudes of polarization on directions a, b, c may be written in the form

$$P_{1a}e^{i\omega_1 t} = Np_{a21}\sigma_{12}e^{i(\omega_1-\Delta)t},$$
$$P_{2b}e^{i\omega_2 t} = N\sum_q [p_{b1q}\rho_{q1}(\omega_2) + \rho_{2q}(\omega_2)p_{bq2}],$$
$$P_{3c}e^{i\omega_3 t} = N\sum_q [p_{cq1}\rho_{1q}(\omega_3) + \rho_{q2}(\omega_3)p_{c2q}]. \quad (3)$$

Here N is the number density of particles of the working substance; p_{amn}, p_{bmn}, p_{cmn} are the projections of the matrix elements of the dipole moment operator on the directions a, b, c;[1)] expressions for the matrix elements ρ_{1q}, ρ_{q2} of the density matrix are determined from the formulas (15) of[4], where it is necessary in accordance with Eqs. (1) to set

$$\omega_\alpha = \omega_3, \quad \omega_\beta = -\omega_2, \quad \omega_\gamma = \omega_1, \quad j = 1, \quad i = 2, \quad \Delta\omega_{\alpha\beta} = \Delta.$$

As a result we have

$$\rho_{q1} = \frac{\sigma_{21}}{\hbar}\left[\frac{p_{bq2}E_2^* e^{-i(\omega_3-\Delta)t}}{\omega_{q2}-\omega_2} + \frac{p_{cq2}E_3 e^{i(\omega_2+\Delta)t}}{\omega_{q2}+\omega_3}\right],$$
$$\rho_{2q} = \frac{\sigma_{21}}{\hbar}\left[\frac{p_{b1q}E_2^* e^{-i(\omega_2-\Delta)t}}{\omega_{q2}+\omega_3} + \frac{p_{c1q}E_2 e^{i(\omega_3+\Delta)t}}{\omega_{q2}-\omega_2}\right]; \quad (4)$$

$\sigma_{21} = \sigma_{12}^*$ is the slow amplitude of the matrix element ρ_{12}, given by (see also[4,5])

$$\sigma_{21} = \frac{i}{\hbar^2}\frac{n}{T^{-1}-i\Delta}(\hbar p_{a21}E_1^* + r^*E_2E_3^*)e^{-i\Delta t}, \quad (5)$$

where

$$r = \sum_q\left(\frac{p_{c1q}p_{bq2}}{\omega_{q2}-\omega_2} + \frac{p_{cq2}p_{b1q}}{\omega_{q2}+\omega_3}\right), \quad (6)$$

n is the population difference of levels 1 and 2 for one particle:

$$n = n_0\left\{1 + \frac{4}{\hbar^4}|\hbar p_{a12}E_1 + rE_2^*E_3|^2\frac{\tau}{(T^{-2}+\Delta^2)T}\right\}^{-1}, \quad (7)$$

n_0 is the equilibrium population difference, is the lifetime of particles in level 2. In writing down (5) and (7) it was assumed that the change of ω_{21} due to the action of the fields E_1 is small (see[6]).

Substituting (4) and (5) into (3), we obtain

$$P_{1a} = -\frac{i}{\hbar^2(T^{-1}+i\Delta)}[\hbar|p_{a21}|^2E_1 + p_{a21}rE_2^*E_3]Nn,$$
$$P_{2b} = \frac{i}{\hbar^2(T^{-1}-i\Delta)}\{p_{a21}rE_1^*E_3 + \hbar^{-1}|r|^2E_2|E_3|^2\}Nn,$$
$$P_{3c} = -\frac{i}{\hbar^2(T^{-1}+i\Delta)}\{p_{a21}r^*E_1E_2 + \hbar^{-1}|r|^2|E_2|^2E_3\}Nn. \quad (8)$$

In (8) we have neglected the contribution of that part of the polarization that is not associated with the resonant condition ($\omega_{21} = \omega_1 = \Delta$). It was shown in[6] that this is possible if the product of the matrix element p_{a21} of the transition 2—1 and the quantity r (see (6)), which determines the intensity of two-quantum transitions between levels 2 and 1, satisfies the condition

$$N|p_{a21}r| \gg \hbar^2T^{-1}|\chi|, \quad (9)$$

where χ is given by the well-known expression for the nonlinear susceptibility tensor (see (22.23) in[7]), if we exclude resonance terms from the components of the latter.

It is necessary to be aware of two possible situations. When all particles of the material system participate in the interaction, Eq. (9) may be written

$$|p_{a21}r| \gg \hbar^2T^{-1}|\chi_1|, \quad (9a)$$

where χ_1 is the nonlinear susceptibility of the substance in the calculation for a single particle. Then Eq. (9a) can be fulfilled if the two-photon transition between working levels is allowed and the line width T^{-1} is sufficiently small even for substances with high susceptibility χ_1 (thus, for example, for a magnitude of $|\chi_1| \approx 10^{-31}$ cgs esu, which occurs in KDP,[8] Eq. (9a) is fulfilled for $|p_{a21}| \geq 10^{-21}$ cgs esu, $T^{-1} \leq 10^{12}$ sec^{-1}, $|r| \approx 10^{-51}$ cgs esu). And if as a working substance one uses impurities (i.e., the condition $\omega_{21} = \omega_1 + \Delta$ is fulfilled only for the latter), then fulfillment of (9) depends, naturally, both on the quantity of the host substance and on the concentration of impurities.

Here we stop to consider cases when condition (9) occurs, since we are interested in the possibilities of these resonance processes for generating and converting frequencies (and, in particular, in the possibility of using for this purpose as a working substance, impurities in crystals which allow the condition of spatial synchronism, but whose intrinsic nonlinear susceptibility is insignificant). At the same time, solution of this problem will answer the question about the features of SRS when the condition of spatial synchronism is fulfilled. We note also that in the case $\omega_1 \ll \omega_2$, fulfillment of this condition in nonresonant parametric conversion in well known nonlinear crystals (like KDP, $LiNbO_3$) is impossible; and in resonant conversion the synchronism condition may be fulfilled on account of anomalous dispersion of the refractive index close to absorption lines.

We now obtain the equation for amplitudes that vary slowly along z. Substituting the amplitudes of the polarization (8) into the Maxwell equations and neglecting attenuation at frequencies ω_2, ω_3, we find

$$\frac{dA_1}{dz} + g_1(\hbar|p_{a21}|^2A_1 + p_{a21}rA_2^*A_3e^{i\delta z})n = 0, \quad (10a)$$

$$\frac{dA_2}{dz} - g_2^*(p_{a21}rA_1^*A_3e^{i\delta z} + \hbar^{-1}|r|^2A_2|A_3|^2)n = 0, \quad (10b)$$

$$\frac{dA_3}{dz} + g_3(p_{a21}r^*A_1A_2e^{-i\delta z} + \hbar^{-1}|r|^2|A_2|^2A_3)n = 0, \quad (10c)$$

where $\delta = k_1 + k_2 - k_3$ and

$$g_l = 2\pi N\omega_l^2/k_lc^2\hbar^2(T^{-1}+i\Delta). \quad (11)$$

3. It is easy to obtain the approximation of a fixed field from (10), if we leave out Eq. (10c) and substitute $A_3 = \text{const}$ in (10a) and (10b). When A_1 and A_2 are sufficiently small, so that the population difference n may be taken as the equilibrium difference, Eqs. (10a) and (10b) are analogous to the system (11) in[1]. Then

[1)] In case the transition 2—1 is magnetic dipolar, we understand by p_{21} the matrix element of the magnetic moment operator, and by E_1 the intensity of the magnetic field of the wave with frequency ω_1.

fulfillment of condition (9) corresponds to neglect of the quantity P in Eq. (34) of[1].

The solution of the system of linear equations (10a) and (10b) has the form

$$E_i = a_i e^{\varkappa_1 z} + b_i e^{\varkappa_2 z},$$

where

$$\varkappa_{1,2} = {}^1/_2\{\hbar^{-1}g_2|r|^2|A_3|^2 - \hbar g_1|p_{a21}|^2 + i\delta \pm [(\hbar^{-1}g_2|r|^2|A_3|^2 + \hbar g_1|p_{a21}|^2 + i\delta)^2 - 4g_1g_2|p_{a21}|^2|r|^2|A_3|^2]^{1/2}\}. \quad (12)$$

This expression was analyzed in[1] for the case[2] when the quantity $\hbar^{-1}g_2|r|^2|A_3|^2$, which characterize the intensity of the SRS process, is much smaller than $\hbar g_1|p_{a21}|^2$, the quantity responsible for absorption of the field at the resonant frequency. We shall show below that the case when the first of these is greater than the second is of greater interest (see Eq. (13) below).

Consider now the case of exact resonance ($\Delta = 0$). It is not difficult to show that for $\Delta = 0$, the value of δ for which κ_1 is maximum is the same as in the nonresonant parametric interaction ($\Delta \to \infty$), i.e., $\delta = 0$. Then

$$\varkappa_1 = \hbar^{-1}g_2^0|r|^2|A_3|^2 - \hbar g_1^0|p_{a21}|^2, \quad \varkappa_2 = 0, \quad (12a)$$

where $g_i^0 = g_i$ ($\Delta = 0$). From (12a) it follows that when the amplitude of the pump field is greater than critical,

$$|A_3| > |A_3|_{cr} = \hbar|p_{a21}|\omega_1 k_2^{1/2}/|r|\omega_2 k_1^{1/2}, \quad (13)$$

there is an exponential growth of the resonant and Raman fields. It will be shown below that $|A_3|_{cr}$ is not the threshold value, in the sense that amplification of fields of frequencies ω_1 and ω_2 is possible also when $|A_3| < |A_3|_{cr}$. However, when (13) is fulfilled, the limiting attainable magnitude of the resonant field sharply increases. A field intensity of the order of $|A_3|_{cr}$ is easily attained with a laser. In fact, for $\omega_1/\omega_2 = 0.1$, $|p_{a21}| \approx 10^{-21}$ cgs esu (two-photon transition allowed between levels 2 and 1), we have $|A_3|_{cr} \approx 3 \times 10^2$ cgs esu, which corresponds to a power of ~20 MW/cm^2. Thus, condition (13) can be fulfilled for transitions in which SRS is observed.

4. To determine the maximum attainable fields and the effective conversion lengths it is necessary to consider all Eqs. (10) together. We transform to real amplitudes and phases, setting

$$A_l = m_l(z)\exp(-i\varphi_l z).$$

Using the new variables

$$x = 2\hbar^{-1}p_{a21}\mu^{1/2}m_1, \quad y = \frac{2\omega_1 p_{a21}\mu^{1/2}k_2^{1/2}}{\hbar\omega_2 k_1^{1/2}}m_2,$$

$$v = \frac{\omega_2 r k_1^{1/2}}{\hbar\omega_1 p_{a21}k_2^{1/2}}m_3,$$

$$\eta = \frac{2\pi\omega_1^2 p_{a21}^2\mu N n}{c^2 k_1 \hbar\tau}, \quad \mu = \frac{\tau}{(T^{-2}+\Delta^2)T} \quad (14)$$

the system of equations (10) can be reduced to the form

$$dx/dz = -[x + (\cos\theta + \alpha\sin\theta)yv]\eta, \quad (15a)$$

$$dy/dz = [(\cos\theta - \alpha\sin\theta)xv + yv^2]\eta, \quad (15b)$$

$$dv/dz = A[(\alpha\sin\theta - \cos\theta)xy - y^2 v]\eta, \quad (15c)$$

$$\frac{d\theta}{dz} = \delta + \left\{\left(\frac{yv}{x} - \frac{xv}{y} + A\frac{xy}{v}\right)\sin\theta + \alpha\left[1 + Ay^2 - v^2 - \left(\frac{yv}{x} + \frac{xv}{y} - A\frac{xy}{v}\right)\cos\theta\right]\right\}\eta, \quad (15d)$$

$$\eta = \eta_0[1 + (x^2 + 2xyv\cos\theta + y^2v^2)]^{-1}, \quad (15e)$$

where

$$\theta = \varphi_1 + \varphi_2 - \varphi_3 + \delta, \quad \eta_0 = \eta(n_0), \quad \alpha = \Delta T,$$

$$A = \frac{r^2\omega_3^2\omega_2^2(T^{-2}+\Delta^2)Tk_1^2}{4p_{a21}^4\tau\omega_1^4 k_2 k_3} \quad (16)$$

From Eqs. (15b) and (15c), it is easy to find the first integral

$$v^2 = v_0^2 + A(y_0^2 - y^2). \quad (17)$$

In spite of this, an analytical solution of the system (15) is not possible in the general case. Hence we consider the case of exact resonance ($\alpha = 0$). As noted in[6] this example permits elucidation of the basic characteristics of the resonant interactions of electromagnetic fields that we are investigating.

For $\alpha = 0$, the starting equations will have the form[3]

$$dx/dz = -(x + yv\cos\theta)\eta, \quad (18a)$$

$$dy/dz = (xv\cos\theta + yv^2)\eta, \quad (18b)$$

$$\frac{d\theta}{dz} = \delta + \left[\frac{(y^2 - x^2)v}{xy} + A\frac{xy}{v}\right]\eta\sin\theta, \quad (18c)$$

where η and v are given respectively by (15e) and (17).

Before examining (18), let us note some features of this process. As is easily seen from (18a) and (18b), the total energy of the resonant and Raman fields in the case of exact resonance can increase only on account of the Raman interaction. In fact, multiplying (18a) by x and (18b) by y and combining, we obtain

$$d(x^2 + y^2)/dz = 2(y^2v^2 - x^2)\eta.$$

The parametric interaction, on the other hand, which is described by expressions proportional to cos θ, affects only the redistribution of energy between the fields with frequencies ω_1 and ω_2. If the difference $y^2 - x^2$ is positive at the boundary ($z = 0$), it remains positive for any z, since (considering that in an uninverted system we always have $\eta > 0$)

$$d(y^2 - x^2)/dz = 2(y^2v^2 + 2xyv\cos\theta + x^2)\eta \geqslant 0. \quad (19)$$

5. We now consider the process of amplification and generation of the resonant field. Let the boundary values of the fields satisfy the condition

$$A^{-1/2}v_0, \quad y_0 > x_0 \approx 0 \quad (20)$$

or in dimensioned variables

$$\frac{\omega_2}{\omega_3}\left(\frac{k_3}{k_2}\right)^{1/2}m_{30}, \quad m_{20} > \frac{\omega_2}{\omega_1}\left(\frac{k_1}{k_2}\right)^{1/2}m_{10} \approx 0. \quad (20a)$$

In the case of exact fulfillment of the synchronism

[2] Note that in [1] the first two terms in the parentheses under the radical in (12) were assumed real. In fact, they are complex, since they contain the complex quantities g_1 and g_2 (for $\Delta \neq 0$).

[3] In what follows all matrix elements p_{mn} are assumed real.

condition ($\delta = 0$), it follows from (18c) that when $m_1 \approx 0$, the phase difference θ along z varies many times faster than the quantities x, y and tends to the stable value $\theta_0 = \pi$. The plane $\theta = \theta_0$ in the space of x, y, θ will be stable for any z, since under the initial conditions (20), the quantity $y^2 - x^2$ does not change sign along z, because of (19). Hence in (18a) and (18b), we can set $\theta = \pi$, and, considering (17), they will take the simple form

$$dx/dz = [(D - Ay^2)^{1/2}y - x]\eta, \tag{21a}$$

$$dy/dz = (D - Ay^2)^{1/2}[(D - Ay^2)^{1/2}y - x]\eta, \tag{21b}$$

where $D = v_0^2 + Ay_0^2$.

It follows from (21) that the fields in the system tend toward limiting values that satisfy the condition

$$x = (D - Ay^2)^{1/2}y, \tag{22}$$

upon fulfillment of which, as is easily shown from (22) and (15e), the population is in equilibrium: $\eta = \eta_0$ ($n = n_0$). We remark that, as with propagation of parametrically coupled waves with a resonant total frequency (see[6]), the possibility of clarifying the substance at the frequency of the resonant field follows from this. For this the values of the fields at the boundary must, in our example, satisfy the condition (22),[4] i.e., $x_0 = y_0 v_0$ and $\theta = \pi$.

We now determine the dependence of the limiting value of the resonant field $x|_{z\to\infty} = x$ on the magnitudes of the fields at the boundary (as before, we shall assume fulfillment of condition (20a) for the latter). Dividing (21b) by (21a) and integrating the resulting equation, we find

$$x = A^{-1/2}\arcsin(A/D)^{1/2}y - F,$$

$$F = A^{-1/2}\arcsin(A/D)^{1/2}y_0 - x_0. \tag{23}$$

Using (22) and (23) we easily obtain an equation for X:

(24)

Of greatest practical interest is the parametric generation of the resonant field at small intensities of the field of the Stokes component at the boundary:

$$x_0 = 0 \quad (m_{10} = 0),$$
$$Ay_0^2 \ll v_0^2 \quad (m_{20} \ll (\omega_2/\omega_3)^{1/2} m_{30}). \tag{25}$$

With the aid of (25), (24), and (22) it is easy to see that in this case the value

$$v_0 = v_{0cr} = 1 \quad \left(m_{30} = |A_3|_{z=0} = \frac{\hbar |p_{a21}| \omega_1 k_2^{1/2}}{|r| \omega_2 k_1^{1/2}}\right)$$

corresponds to a sharp increase of the limiting value of the resonant field (see Fig. 2). However, even when $v_0 < 1$ generation of this field is possible. We recall that in the approximation of a fixed pump field with $v > 1$ we have unlimited growth of the amplitude of the field of frequency ω_1.

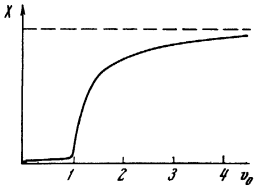

FIG. 2. Dependence of the maximum attainable resonant field on the boundary value of the pump field (in the dimensionless variables of (14) for $y_0 \ll 1$; $\lim_{v\to\infty} x_1 = \frac{\pi}{2}\frac{\hbar p_{a21}}{r}\frac{\omega_1}{(\omega_2\omega_3)^{1/2}} = \frac{\pi}{2}\left(\frac{\omega_1}{\omega_3}\right)^{1/2} (m_{30})_{cr}$. The dashed line is for $X = \pi/2A^{1/2}$.

For estimates of the magnitude of X for small excesses of v_{0cr}: $2 > v_0 > 1$, we take $X = X_1$, the value[5] of the coordinate of the apex of the curve (24):

$$X_1 = (v_0^2 + 2Ay_0^2)/2A^{1/2}. \tag{26}$$

In the general case, the equality $X = X_1$ occurs for y_0 and v_0 that satisfy the condition

$$v_0^2 + 2Ay_0^2 + 2\arcsin\left\{\left(\frac{A}{v_0^2 + Ay_0^2}\right)^{1/2} y_0\right\} - x_0 = \frac{\pi}{2}. \tag{27}$$

If (25) is fulfilled, the boundary condition relation for which $x|_{X\to\infty} \to X_1$ has the form

$$Ay_0^2 \approx \pi/2 - v_0^2. \tag{27a}$$

It follows from (26) and (27a) that already for small excess over the critical value of the field $E(\omega_3)$ at the boundary:

$$v_0 \approx (\pi/2)^{1/2} \approx 1.25,$$

the magnitude

$$X = X_1 = {}^1/_2 \lim_{v_0\to\infty} X$$

is reached; then the boundary value of the Raman field necessary for attainment of X_1 is very small.

The relations (27) and (27a) will be needed later for calculating the effective length of field conversion.

Generally speaking, the results of this section are applicable when the condition of spatial synchronism, i.e., $\delta = 0$, is exactly fulfilled. However, it will be shown below that they permit finding the optimum conditions for the generation of the resonant field also in the case when the detuning is nonzero but less than a certain critical value.

6. Now let us consider the limits of applicability of the solutions obtained in the preceding section for the case $\delta \neq 0$. In exactly the same way as in Sec. 5 of[6], it is easy to show that when

$$|\delta|^2 \ll f_{min}^2, \tag{28}$$

where f_{min} is the minimum of the coefficient of $\sin\theta$ in (18c), the phase difference θ tends toward $\theta = \pi + \varphi$, where

$$|\varphi| \leqslant |\delta|/f_{min} \ll 1. \tag{29}$$

[4]The possibility of the analogous phenomenon of clarification of the substance in the case of propagation of parametrically coupled waves with frequencies ω and 3ω in the presence of two-photon absorption of the field at the fundamental frequency was mentioned in [9].

[5]Note that the magnitude X_1 can be reached only for $v_0^2 + Ay_0^2 < 2$. In fact, for the reverse inequality, the apex of the curve of equilibrium states lies in the region $y < x$, whereas it follows from (19) that in the case of boundary conditions (20) all integral curves of the system (18) lie in the region $x < y$.

Then $\cos\theta = -(1 - \varphi^2/2)$, and from Eqs. (18) we find

$$dx/dz = \{y(D - Ay^2)^{1/2}(1 - \varphi^2/2) - x\}\eta, \quad (30a)$$

$$dy/dz = \{y(D - Ay^2)^{1/2} - x(1 - \varphi^2/2)\}(D - Ay^2)\eta. \quad (30b)$$

It is obvious that in case (28) we can neglect $\varphi^2/2$ compared to 1 far from curve (22) in the (x, y) plane and we can assume phase trajectories that coincide with those which occur for $\delta = 0$. From (21) and (23) we can find the length z_1 in which the quantity x changes from x_0 to x_1, where x_1 is defined as the point of intersection of the considered phase trajectory with the boundary of the region (see Fig. 2):

$$|(D - Ay^2)^{1/2}y - x| \approx \varphi^2; \quad (31)$$

$$\eta_0 z_1 = 2\int_{x_0}^{x_1} \frac{dx}{DA^{-1/2}\sin 2A^{1/2}(x+F) - 2x} + \frac{x_0^2 - x_1^2}{2} + \frac{D}{4A}[1 - 2AD^{-1}y_0^2 - \cos 2A^{1/2}(x_1 + F)]. \quad (32)$$

When the imaging point approaches the region of (31), it is necessary to consider Eqs. (30) instead of (21). It follows from (30) that instead of the equilibrium curve (24) there exists for $\delta \neq 0$ only one stable equilibrium state–the point with coordinates $y = (D/A)^{1/2}$, $x = 0$.

Thus, the field of the Raman frequency tends to the same value $y = (D/A)^{1/2}$ as it would have tended even in the absence of the condition of spatial synchronism (and consequently in the absence of the resonant field). However, in this case, beginning from a field magnitude y_1 (see Fig. 3), the rate of change of $y(z)$ is at least φ^{-2} times less than in a medium that does not allow fulfillment of the condition of spatial synchronism for the interacting fields. In fact, Eqs. (30) permit one to find approximately the dependence of the amplitudes of the resonant and Raman fields on the coordinates in the region of (31); for an insignificant change of the populations, i.e., $\eta = \eta_0$, we have from (30)[6]

$$y^2 \approx \frac{Dy_1^2 \exp(2D\varphi^2\eta_0 z)}{v_1^2 + Ay_1^2\exp(2B\varphi^2\eta_0 z)}, \quad (33a)$$

$$x \approx \frac{Dy_1 v_1 \exp(D\varphi^2\eta_0 z)}{v_1^2 + Ay_1^2\exp(2D\varphi^2\eta_0 z)} \quad (33b)$$

where $v_1^2 = D - Ay_1^2$. The validity of the assertion made above about the rate of change of the Raman field is easy to see by comparing (33a) with the solution of Eq. (18b) in the absence of the spatial synchronism condition.

We now discuss the question of the length in which significant transfer of the energies of the interacting fields occurs. We consider first the case $D > 1$. The conversion length may be found from (32) and (33b), if we set $x_1 = X - \varphi^2$, where X is the solution of (24). It is obvious that the length L in which a maximum in x is attained is (to the accuracy of the order φ^2)

$$L = z_1 + z_2,$$

where z_1 is determined from (32) and z_2 from (33b),

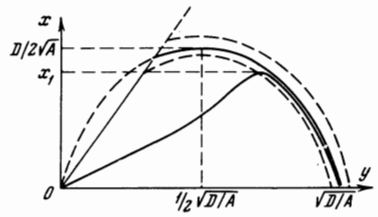

FIG. 3. Phase plane in the case $v_0 > v_{0cr} = 1$.

(17), and (23):

$$\eta_0 z_2 = \frac{1}{2D\varphi^2}\ln\frac{v_1^2}{Ay_1^2} = \frac{1}{2D\varphi^2}\ln\frac{D - Ay_1^2}{Ay_1^2} \approx \frac{1}{2D\varphi^2}\ln \operatorname{tg}^2 A^{1/2}(X + F - \varphi^2); \quad (34a)$$

here X, as before, is the solution of (24) for boundary values y_0 and v_0 satisfying the conditions (20). If the boundary conditions are such that the left hand side of (27) is greater than or equal to the right, then $L_1 = z_1$; then as x_1 it is sufficient to take $x_1 \approx 0.9X$. With further increase of z the resonant field decreases (see Figs. 3 and 4).

Here we make the following remark. The first term in (32) is independent of τ, the lifetime of particles of the working substance in the excited state; the remaining terms depend on this time. Hence τ strongly influences the magnitude of z_1 if the first term of (32) is less than the sum of the others. In Sec. 8 we shall estimate the values of τ for which such influence occurs in a concrete example.

When $D < 1$ and the magnitude of the Raman field at the boundary is sufficiently small ($A^{1/2}y_0 \lesssim \varphi^2$), all changes in the fields occur in the region of (31) already at the boundary itself. Then the dependence $x(z)$ is determined by (33b),[7] in which it is necessary to set $x_1 = x_0$, $y_1 = y_0$, while

$$L = z_2 = \frac{1}{2D\eta_0\varphi^2}\ln\frac{v_0^2}{Ay_0^2}. \quad (34b)$$

In the conclusion we shall estimate the magnitude of δ for which condition (28) is fulfilled. With the aid of (17), (18c), (23), and (25), it is possible to show

$$|f_{min}| \gtrsim |f(X_1)| = \eta_0. \quad (35)$$

7. For large $|\delta|$ it is possible to assume that in first approximation the Raman field increases, as in ordinary SRS. We restrict ourselves to the case when the population difference of the working levels changes little with coordinate. Then it is easy to find $y(z)$ from (17) and (18b). Substituting this solution into (18a), we obtain

$$x = -Dy_0 v_0 \eta_0 \exp(-\eta_0 z)\cdot\int_0^z \frac{\exp[(D+1)\eta_0 z]\cos\delta z}{v_0^2 + Ay_0^2\exp(2D\eta_0 z)}dz. \quad (36)$$

When $dy/dz \ll \delta$, which is the case when

$$2D^{3/2}\eta_0/3\sqrt{3}A^{1/2} \ll \delta, \quad (37)$$

then the coefficient of $\cos\delta z$ in (36) can be taken out; taking into account that as $x \to 0$ the phase difference

[6] It is easy to establish from (15e) and (28) that if it is impossible to neglect the change of the population difference along z, then the rate of change of x and y is still less than in (33).

[7] Of course, in this case the approximate solution (33) has meaning only for $\varphi^2 \ll D/2A^{1/2}$.

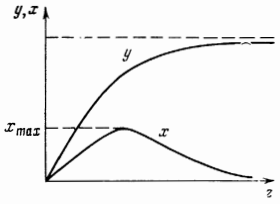

FIG. 4

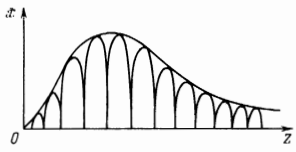

FIG. 5

FIG. 4. Qualitative form of the dependence of field intensity: y—at the Stokes frequency ω_2 and x—the resonant field at frequency ω_1, on the coordinate z, with fulfillment of the condition $|\varphi| < \varphi_{cr}$.

FIG. 5. Qualitative dependence of the magnitude of the resonant field on coordinate for large mismatches of the phase velocities.

$\theta \to \pi$ (see (18b)), we finally obtain (see Fig. 5)

$$x = \frac{Dy_0 v_0 \eta_0 \exp(D\eta_0 z)}{v_0^2 + Ay_0^2 \exp(2D\eta_0 z)} \frac{|\sin \delta z|}{\delta}. \quad (38)$$

The length in which the amplitude of the resonant field attains a maximum is determined (to the accuracy of magnitudes of the order δ^{-1}) by the expression

$$L = (2D\eta_0)^{-1} \ln (v_0^2 / Ay_0^2) \quad (39a)$$

or in dimensioned variables

$$L = \frac{c^2\hbar^3 k_2}{4\pi\omega_2^2 r^2 T n_0 N m_{30}^2} \ln \frac{\omega_2^2 k_3 m_{30}^2}{\omega_3^2 k_2 m_{20}^2}, \quad (39b)$$

and the magnitude of this maximum is

$$x_{max} = D\eta_0 / 2A^{1/2}\delta \quad (40)$$

or

$$m_{1\,max} = \frac{\pi\omega_1^2 (k_2 k_3)^{1/2} p_{a\,21} r T N n_0}{\hbar^2 c^2 \omega_2 \omega_3 k_1 \delta} \left(\frac{\omega_2^2}{k_2} m_{30}^2 + \frac{\omega_3^2}{k_3} m_{20}^2 \right). \quad (40a)$$

8. Let us consider the process of frequency combination, limiting ourselves to the case of a fixed field at frequency ω_2 ($y_0 \gg x, v$) and, as before, assuming that $\Delta = 0$. From Eqs. (18) it is easy to determine the limiting field at the sum frequency for $\delta = 0$, $v_0 = 0$:

$$\lim_{z\to\infty} v = Ay_0 x_0 / (1 + Ay_0^2). \quad (41a)$$

Transforming to dimensioned quantities using (14) and (16), we find

$$\lim_{z\to\infty} m_3 = \frac{\hbar p_{a\,21} r \omega_3^2 k_1 m_{10} m_{20}}{p_{a21}^2 \omega_1^2 k_3 \hbar^2 + r^2 \omega_3^2 k_1 m_{20}^2}. \quad (41b)$$

The approximation of a fixed field is applicable if $\lim v \ll y_0$ as $z \to \infty$, i.e., (see (41a)),

$$y_0^2 \gg x_0 - A^{-1},$$

or

$$m_{20}^2 \gg \frac{\hbar\omega_2^2 k_1 m_{10}}{2\omega_1^2 p_{a\,21} \mu^{1/2} k_2} - \left(\frac{\hbar p}{r}\right)^2 \frac{\omega_1^2 k_3}{\omega_3^2 k_1}.$$

We give also the form of the function $z(v)$:

$$\eta_0 z = (1 + Ay^2)^{-1} \ln \frac{Ay_0 x_0}{v_0 + Ay_0 x_0 - v(1 + Ay^2)} + \frac{2Ay_0 x_0 v - v^2(1 + Ay_0^2)}{A^2 y_0^2}. \quad (42)$$

The length L in which the field at the sum frequency becomes 0.9 lim v (for $z \to \infty$) is

$$L_{0.9} = \frac{4.6 + x_0^2}{2\eta_0 (1 + Ay_0^2)} \quad (43a)$$

or

$$L_{0.9} = \frac{\hbar^2 c^2 k_1 k_3}{2\pi N n_0} \left(\frac{2{,}3\hbar^2 T^{-1} + 2p_{a21}^2 \tau m_{10}^2}{\hbar^2 k_3 p_{a21}^2 \omega_1^2 + r^2 \omega_3^2 k_1 m_{20}^2} \right). \quad (43b)$$

The latter expression permits elucidation of the effect of the lifetime of the particles of the working substance on the effective conversion length. Obviously, the contribution of the term in (43b) that depends on τ is significant if

$$\tau \gtrsim \tau_{cr} = \hbar^2 / T p_{a\,21}^2 m_{10}^2. \quad (44)$$

We make some numerical estimates in conclusion. For

$$|p_{a21}| = 10^{-20} - 10^{-21} \text{ cgs esu},$$
$$N = 10^{20} \text{ cm}^{-3}, \quad T = 10^{-11} \text{ sec},$$
$$r = 10^{-51} \text{ cgs esu}, \tau \ll \tau_{crit}$$

and for $m_{10} = 1$ cgs esu (corresponding to a radiated power at ω_1 of the order of 200 W/cm^2), we have

$$\lim_{z\to\infty} m_3 \approx 1 - 0.1 \text{ cgs esu},$$
$$L_{0.9} \approx 10 - 100 \text{ cm}.$$

9. We now discuss these results.

A. First of all we remark that in the framework of our model, taken after the authors of[1] (the particles of the working substance interact only via the radiation field), the limiting magnitude of the Raman field for nonzero detuning of the wave vectors δ is just the same as in the absence of the spatial synchronism condition. However, for a sufficiently small δ (see (18)), the growth of this field along z is markedly slower than in the latter case (see Eq. (41)).

B. For sufficiently small δ (see (28)) the part of the energy which is lost by the pump in ordinary SRS can go over into induced emission at the frequency of the working transition $\omega_1 = \omega_{21}$. The process by which the resonant field arises can be described as a mixing of the fields of the pump (frequency ω_3) and of the field at frequency ω_2 arising in the SRS process by resonant nonlinearity with losses (see Eq. (10)). The maximum of the field $E(\omega_1)$ is attained in a length determined by Eqs. (32), (34a), and (34b). The magnitude of this maximum increases sharply when the amplitude of the pumping field satisfies the condition (13). It should be noted that for fulfillment of condition (13) with realizable intensities of the pump field $E(\omega_3)$, the condition $P_{a21} < 10^{-20}$ cgs esu ought to be fulfilled. However, as p_{a21} decreases the length $L_{0.9}$ sharply increases. As seen from the table, the magnitude of $L_{0.9}$ varies from ~10 to ~100 cm for values of p_{a21} from 3×10^{-21} to 10^{-22}. Thus, generation of a resonant field with significant power is possible only over a rather narrow interval of values of the parameters p_{a21} and r. However, it should be remembered that in the general case the conversion length decreases

Dependence of effective conversion length on p for $N = 10^{21}$ cm^{-3}

x/X	$p = 3\cdot10^{-21}$, $A = 2.5$	$p = 10^{-21}$, $A = 250$	$p = 3\cdot10^{-22}$, $A = 2500$
	z_1, cm	z_2, cm	z_3, cm
0.1	2,5	10	35,6
0.2	3,0	15	78,9
0.3	3.7	18	103
0.4	4,2	21	124
0.5	4,7	24	144
0.6	5,3	26	163
0.7	5,8	28	184
0.8	6,6	31	209
0.9	8,0	32	230

In all cases $v_0 = 1.25$, $y_0 = 10^{-3}$.

as the line narrows (in this it is important that the time τ be less than critical (see, for example, (44)).

C. The considered parametric interaction is less sensitive to the exactness of fulfillment of the condition of spatial synchronism that the nonresonant parametric interaction. In fact, in the latter case L is a length of coherent interaction of the order δ^{-1}; in the former process, on the other hand, L equals z_1 when $\delta < \delta_{cr}$, from Eq. (32). As follows from (28), (35), and (14), $\delta_{cr} \approx 2\pi\omega_1 p_{a21} NT/c\hbar$. For the values of the parameters used in calculating the values of L given in the first column of the table, we have $(\delta_{cr})^{-1} \approx 0.5$ cm, whereas $L = z_1 = 10$ cm.

D. The treatment we have carried out of the resonant parametric interaction is strictly valid only in the case when the working substance is an impurity in a solid. However, since the effects discussed can arise in compressed gases (pressure ~10 to 100 atm), we may suppose that a resonant field can arise in the case of SRS in a gas, if a constant field strong enough to permit the working transition to a slight degree is applied.

We thank B S. Butylkin, A. E. Kaplan, and V. M. Faĭn for helpful discussions.

[1] P. N. Butcher, R. W. Loudon, and T. P. McLean, Proc. Phys. Soc. (London) 85, 565 (1965).

[2] Y. R. Shen, Phys. Rev. 138, A1741 (1965).

[3] F. De Martini, J. Appl. Phys. 37, 4503 (1966).

[4] Yu. G. Khronopulo, Izv. Vysshikh. Uchebn. Zavedenii, Radiofiz., 7, 675 (1964) [Izv. Vuzov, Radiophys., 7, No. 4, 118 (1964)].

[5] G. L. Gurevich and Yu. G. Khronopulo, Izv. Vysshikh. Uchebn. Zavedenii, Radiofiz., 8, 493 (1965).

[6] G. L. Gurevich and Yu. G. Khronopulo, Zh. Eksp. Teor. Fiz. 51, 1499 (1966) [Sov. Phys. JETP 24, 1012 (1967)].

[7] V. M. Faĭn and Ya. I. Khanin, Kvantovaya radiofizika (Quantum Radiophysics), Sov. Radio, 1965.

[8] S. A. Akhmanov and R. V. Khokhlov, Problemy nelineĭnoĭ optiki (Problems in Nonlinear Optics), VINITI, 1964.

[9] É. A. Manykin and A. M. Afanas'ev, Zh. Eksp. Teor. Fiz. 48, 931 (1965) [Sov. Phys. JETP 21, 619 (1965)].

Translated by L. M. Matarrese
140