Contribution to the theory of resonant four-wave parametric interactions

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Four-wave parametric interaction of light waves in two-photon absorption is investigated. It is shown that, significantly different parametric-generation regimes may take place, depending on the ratio of the powers of the triggering and pumping fields (Fig. 1). An optimal ratio of the powers exists, for which the coefficient for conversion of the pumping into the generating field is maximal. The singularities of four-wave interactions due to phase locking of the generated fields lead to conditions under which the conversion coefficient is not restricted by population saturation due to two-photon absorption. Calculations are presented for the optimal conditions of generation at the combined and difference frequencies in the vacuum ultraviolet range. In particular, the possibility of creating a source which is tunable in the 650–656 Å range is discussed.

1. INTRODUCTION

The interest recently advanced in resonant four-wave parametric processes is due primarily to the fact that they make it possible to obtain coherent radiation sources with continuously tunable frequency, including the infrared, ultraviolet, and vacuum ultraviolet (VUV) regions of the spectrum, if tunable lasers, say liquid-state, are used for their triggering. By now, four-wave processes have been observed experimentally, based on generation of the Stokes component of stimulated Raman scattering (SRS) [1,2] and on the basis of two photon absorption (TPA) of the pump field [3] in metal vapor (see Fig. 1). Coherent radiation in the IR and VUV bands was obtained in these studies and could be tuned over a rather wide frequency range. However, there is still no sufficiently complete theoretical description of these processes, although the first discussion of one of them dates back to 1964 [4].

Four-wave resonant interactions are usually treated (see [5]) in the same manner as nonresonant parametric interactions of waves, for example third-harmonic generation in a transparent medium [6,7]. The polarization of a medium at the frequency $\omega_4 = \omega_1 + \omega_2 \pm \omega_3$ (Fig. 1) is assumed to be proportional to $\chi E_1E_2E_3$, where χ is the corresponding nonlinear susceptibility of the medium and E_j are the amplitudes of the waves with frequency ω_j. It is assumed here that resonance introduces only a change in the value of χ, and that wave interaction takes place effectively only over a length $(\delta k)^{-1}$ determined by the mismatch δk between the wave vectors of the fields. For real situations, which take place in the experiment (large radiation flux densities, reaching several gigawatts, and appreciable thicknesses of the working medium), this description is utterly inadequate because the resonant parametric processes proceed in the general case in an entirely different manner than the nonresonant ones. Thus, phase locking of the generated fields takes place in resonant processes, as a result of which the conversion can take place over lengths greatly exceeding the synchronism length; the dependence of the amplitudes of the interacting fields on the coordinates can in this case be entirely different than in the case of nonresonant processes[1)]. In addition, such a description does not take correct account of the inverse conversion of generated fields into a pump field, so that it becomes impossible to determine the maximum attainable generated fields and the optimal generation conditions.

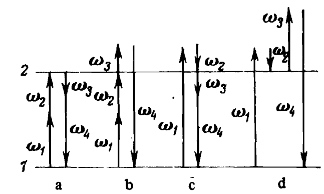

FIG. 1. Resonant four-wave parametric processes based on TPA of pump fields and on SRS; ω_1 and ω_2 are the frequencies of the pump fields (or of the pump and the SRS), ω_3 is the frequency of the triggering field, and ω_4 is the frequency of the generating field.

We consider in this paper four-wave parametric processes based on two-photon absorption of the pump field (see Fig. 1).

2. EQUATIONS FOR THE AMPLITUDES AND PHASES OF THE INTERACTING FIELDS

The procedure for calculating the polarization produced in a medium by many-frequency parametric processes was developed in [9,10]. Using this procedure, we easily obtain the medium polarization produced under the influence of the fields

$$\mathbf{e}_j[\mathscr{E}_j \exp(i\omega_j t - ik_j z) + \text{c.c.}$$

whose frequencies satisfy the condition

$$\omega_\alpha + \omega_\beta = \omega_\gamma + \omega_\delta = \omega_{21} + \Delta, \qquad (1)$$

where $j = \alpha, \beta, \gamma, \delta$; $\mathbf{e}_j$, ω_j and k_j are respectively the unit vectors of the polarization, the frequencies, and the wave vectors of the interacting fields, $\mathscr{E}_j$ are the complex amplitudes of these fields, ω_{21} is the frequency of the transition between levels 2 and 1 of the beam, and Δ is the frequency detuning. Substituting the expressions for the polarization in Maxwell's equations and considering one-dimensional time-stationary interaction of waves with zero frequency detuning, we obtain equations for the slow real amplitudes $x_j(z) = \mathscr{E}_j(z)e^{i\varphi_j(z)}$ and the phase differences $\Theta = \varphi_\alpha + \varphi_\beta - \varphi_\gamma - \varphi_\delta + z\delta k$ of the interacting fields:

$$\frac{dx_\alpha}{dz} = -\frac{2\pi\omega_\alpha nN}{\hbar^3 c}(d_1 r_1^2 x_\beta^2 x_\alpha + d_2 r_1 r_2 x_\beta x_\gamma x_\delta \cos\Theta), \qquad (2a)$$

$$\frac{dx_\gamma}{dz} = -\frac{2\pi\omega_\gamma nN}{\hbar^3 c}(d_2 r_2^2 x_\delta^2 x_\gamma + d_1 r_1 r_2 x_\alpha x_\beta x_\delta \cos\Theta), \qquad (2b)$$

$$\frac{d\Theta}{dz} = \delta k + \frac{2\pi nN r_1 r_2}{\hbar^3 c}\Big[x_\alpha x_\beta\Big(\frac{x_\delta}{x_\gamma}\omega_\gamma + \frac{x_\gamma}{x_\delta}\omega_\delta\Big)d_1 + x_\delta x_\gamma\Big(\frac{x_\alpha}{x_\beta}\omega_\beta + \frac{x_\beta}{x_\alpha}\omega_\alpha\Big)d_2\Big]\sin\Theta, \qquad (2c)$$

where $\delta k = k_\alpha + k_\beta - k_\gamma - k_\delta$, and N is the particle-number density. The equations for x_β and x_δ can be obtained from (2) and (2b) by making the substitutions $\alpha \rightleftarrows \beta$ and $\gamma \rightleftarrows \delta$.

In the case of a medium consisting of immobile molecules having the same orientation, Eqs. (2) describe the interaction of the fields with allowance for saturation of the populations of levels 1 and 2, in which case $d_1 = d_2 = T$ (T is the reciprocal of the line width of the transition 1—2),

$$r_1 = \sum_q \left[\frac{(\mathbf{p}_{1q}\mathbf{e}_\alpha)(\mathbf{p}_{q2}\mathbf{e}_\beta)}{\omega_{q2}+\omega_\beta} + \frac{(\mathbf{p}_{1q}\mathbf{e}_\beta)(\mathbf{p}_{q2}\mathbf{e}_\alpha)}{\omega_{q2}+\omega_\alpha} \right], \tag{3}$$

where p_{jq} are the dipole moments of the transitions between the levels j and q (we assume them to be real quantities); an expression for r_2 is obtained from (3) by making the substitutions $\alpha \to \gamma$ and $\beta \to \delta$. The population difference levels 1 and 2 is

$$n = n_0 f_p = n_0 \{1 + 4\hbar^{-4}\tau d[r_1^2 x_\alpha^2 x_\beta^2 + r_2^2 x_\gamma^2 x_\delta^2 + 2 r_1 r_2 x_\alpha x_\beta x_\gamma x_\delta \cos\Theta]\}^{-1} \tag{4}$$

(τ is the lifetime of the particle in the state 2).

In a gas medium we have

$$d_1 = \frac{T^{-1}}{T^{-2} + [(\mathbf{k}_\alpha + \mathbf{k}_\beta)\mathbf{v}]^2} \qquad d_2 = \frac{T^{-1}}{T^{-2} + [(\mathbf{k}_\gamma + \mathbf{k}_\delta)\mathbf{v}]^2} \tag{5a}$$

where $\mathbf{v}$ is the velocity of the molecule, and Eqs. (2) must be averaged over the orientations of the molecules and the Maxwellian velocity distribution, which reduces to an averaging of products made up of the quantities $d_{1,2} n r_1 n r_2$. In the general case this averaging is impossible, but such an averaging can be carried out for fields weaker than saturating.

Averaging over the velocities [11] yields[2)]

$$\langle d_1 \rangle = \frac{\sqrt{\pi}}{|k_\alpha + k_\beta| \bar{v}}, \quad \langle d_2 \rangle = \frac{\sqrt{\pi}}{|k_\gamma + k_\delta| \bar{v}}, \tag{5b}$$

where $\bar{v}$ is the averaged velocity of the molecules, and the quantities r_1^2, r_2^2, and $r_1 r_2$ can be averaged over the molecule orientations by using the properties of the isotropic fourth-rank tensors [12] in terms of which they can be expressed:

$$\langle r_1 r_2 \rangle = \sum_{q,j} \{[(\omega_{q2}+\omega_\beta)^{-1}(\omega_{j2}+\omega_\delta)^{-1} + (\omega_{q2}+\omega_\alpha)^{-1}(\omega_{j2}+\omega_\gamma)^{-1}] \times (g\lambda_1 + e\lambda_2 + f\lambda_3) + [(\omega_{q2}+\omega_\beta)^{-1}(\omega_{j2}+\omega_\gamma)^{-1} + (\omega_{q2}+\omega_\alpha)^{-1}(\omega_{j2}+\omega_\delta)^{-1}] \times (g\lambda_1 + f\lambda_2 + e\lambda_3)\} \tag{6}$$

where

$$\lambda_1 = {}^1/_{30}(4a-b-c), \quad \lambda_2 = {}^1/_{30}(4b-a-c), \quad \lambda_3 = (4c-a-b),$$
$$a = (\mathbf{p}_{1q}\mathbf{p}_{q2})(\mathbf{p}_{1j}\mathbf{p}_{j2}), \quad b = (\mathbf{p}_{1q}\mathbf{p}_{1j})(\mathbf{p}_{q2}\mathbf{p}_{j2}), \quad c = (\mathbf{p}_{1q}\mathbf{p}_{j2})(\mathbf{p}_{q2}\mathbf{p}_{1j}), \tag{7}$$
$$e = (\mathbf{e}_\alpha\mathbf{e}_\gamma)(\mathbf{e}_\beta\mathbf{e}_\delta), \quad f = (\mathbf{e}_\alpha\mathbf{e}_\delta)(\mathbf{e}_\beta\mathbf{e}_\gamma), \quad g = (\mathbf{e}_\alpha\mathbf{e}_\beta)(\mathbf{e}_\gamma\mathbf{e}_\delta).$$

The quantities $\langle r_1^2 \rangle$ and $\langle r_2^2 \rangle$ can be obtained from (6) by putting $\alpha = \gamma$ and $\beta = \delta$.

Equations (2) describe the following processes:

a) Resonant four-wave parametric interactions based on two-photon absorption of the pump field [3,5] with enhancement (Fig. 1a) or absorption (Fig. 1b) of the triggering field of frequency ω_3. The process with enhancement of the triggering field can be called also generation of the difference frequency, and that with absorption of the triggering field can be called generation of the combined frequency. To obtain equations describing these processes it is necessary to put $\alpha = 1$, $\beta = 2$, $\gamma = \pm 3$, and $\delta = 4$ in (1) and (2), where the plus sign of the index γ pertains to generation of the difference frequency, and the minus sign to generation of the combined frequency (the relations $\omega_{-j} = -\omega_j$ and $k_{-j} = -k_j$ hold for the frequencies and wave vectors of the fields).

b) Parametric processes based on generation of the Stokes component of SRS [1,2,4], accompanied by enhancement (Fig. 1c) or absorption (Fig. 1d) of the triggering field. When the frequencies of the triggering field and the pump field are equal the field interaction shown in Fig. 1d is anti-Stokes stimulated Raman scattering, the singularities of which, due to phase locking, were investigated in [8]. To obtain equations describing these processes, it is necessary to put $\alpha = 1$, $\beta = -2$, and $\gamma = \pm 3$ and $\delta = 4$ in (1) and (2). We note that when the medium is exposed to the pumping and triggering fields, simultaneous generation of a difference (Figs. 1a and 1c) and combined (Figs. 1b and 1d) frequencies is possible. An analysis of the competition of these processes calls for the solution of the problem of five-wave interaction. We shall investigate the generation of the difference and combined frequencies under the assumption that only one of them takes place. This situation can be realized, for example, by introducing into the medium a substance that absorbs the corresponding frequency ω_4.

3. DIFFERENCE-FREQUENCY GENERATION

We consider the case of exact phase matching: $\delta k = 0$. The role of the mismatch of the phase velocities of the waves will be discussed later on.

As can be easily seen from (2c), at $\langle r_1 r_2 \rangle > 0$ the plane $\Theta = \pi$ is stable. If $\langle r_1 r_2 \rangle < 0$, then the stable plane is $\Theta = 0$; the equations for the field amplitudes remain unchanged in this case. For the sake of argument, we assume $\langle r_1 r_2 \rangle > 0$. Putting $\Theta = \pi$ in (2) and introducing the notation

$$\zeta = Az, \quad A = \frac{2\pi d n_0 N}{\hbar^3 c}\omega_p \langle r_1^2 \rangle, \quad s = \frac{\langle r_1 r_2 \rangle}{\langle r_1^2 \rangle},$$
$$\omega_\gamma = \omega_t, \quad \omega_\alpha = \omega_\beta = \omega_p, \quad \omega_\delta = \omega_g, \quad q_t = \frac{\omega_t}{\omega_p}s, \tag{8}$$
$$q_g = \frac{\omega_r}{\omega_p}s,$$

we write down the equations for the amplitudes of the pumping field ($x_p = x_\alpha = x_\beta$), the triggering field ($x_t = x_\gamma$), and the generated field ($x_g = x_\delta$) in the form

$$dx_p/d\zeta = -x_p(x_p^2 - s x_t x_g) f_n, \tag{9a}$$
$$dx_t/d\zeta = q_t x_g (x_p^2 - s x_t x_g) f_n, \tag{9b}$$
$$dx_g/d\zeta = q_g x_t (x_p^2 - s x_t x_g) f_n. \tag{9c}$$

In the derivation of (9) it was assumed that

$$F = \langle r_1 r_2 \rangle / \langle r_1^2 \rangle^{1/2} \langle r_2^2 \rangle^{1/2} = 1. \tag{10}$$

The equality (10) is rigorously satisfied for oriented molecules. For gaseous media, $F < 1$. It can be deduced from (6) and (7) that this parameter is close to unity in most cases. For example, (10) holds true when the interacting fields have identical polarizations, if the dipole-moment vectors $\mathbf{p}_{ij}$ participating in the formation of the composite matrix elements (3) are collinear. The influence of the deviation of F from unity on the behavior of the fields will be discussed later.

The first and second integrals of the system (9), under the initial conditions

$$x_p|_{z=0} = x_{p0}, \quad x_t|_{z=0} = x_{t0}, \quad x_g|_{z=0} = 0$$

can be written in the form

$$x_t^2 = x_{t0}^2 + (q_t/q_g)x_g^2, \tag{11}$$

$$x_g = \frac{1}{2}\sqrt{\frac{q_g}{q_t}}\,x_{t0}\left[\left(\frac{x_{p0}}{x_p}\right)^{\xi} - \left(\frac{x_p}{x_{p0}}\right)^{\xi}\right], \quad \xi = \sqrt{q_g q_t}. \tag{12}$$

Relation (11) is valid at all values of δk.

It is easy to see from (9) that the equilibrium state

$$\bar{x}_p^2 = s\bar{x}_g\bar{x}_t \tag{13}$$

is stable. Substituting (11) and (12) in (13) we obtain an equation for the equilibrium amplitude of the pump field:

$$\bar{I}_p - \eta I_{t0}\left[\left(\frac{I_{p0}}{\bar{I}}\right)^{\xi} - \left(\frac{\bar{I}_p}{I}\right)^{\xi}\right] = 0, \tag{14}$$

$$\eta = {}^1/_4 s(\omega_g/\omega_t)^{1/2}, \quad \bar{I}_p = \bar{x}_p^2, \quad I_{t0} = x_{t0}^2.$$

The solution (14), together with (11) and (12), determines the values of the equilibrium amplitudes $\bar{x}_t$ and $\bar{x}_g$.

Figure 2 shows the integral curves (12) and the equilibrium-state curves (13) for different ratios of the limiting values of the triggering and pumping fields $\Lambda = x_{t0}/x_{p0}$. It is clear from this figure that the limiting value of the generated field is small for both small and large values of Λ. If the parameter Λ is small, the amplification process proceeds slowly, so that the pump fields reaches an equilibrium state after experiencing a strong two-photon absorption. In the case of large Λ, on the other hand, the inverse parametric conversion of the amplified fields into the pump field takes place effectively, so that the damping of the pump field and the growth of the triggering field are negligible, but the equilibrium state sets in at a small value of the generated field (see (13)). Thus, there exists an optimal ratio Λ_{opt} at which the amplification of the fields is maximal.

To find the optimal ratio of the limiting values of I_{t0} and I_{p0}, it is necessary to determine the maximum of the function $\bar{I}_g = f(I_{p0}, I_{t0})$, which is implicitly specified by the system of equations (13) and (12). The final equation for the optimal ratio of I_{t0} and I_{p0} takes the form

$$I_{p0}(2\xi\eta I_{t0})^{\xi+1} = \eta I_{t0}[I_{p0}^{2\xi} - (2\xi\eta I_{t0})^{2\xi}]. \tag{15}$$

In the general case it is impossible to obtain an analytic expression for the conversion coefficient. By way of example, we consider the optimal conversion regime in the particular case $\xi = 1$ (for example, $\xi = 1$ at $\omega_g/\omega_t = 7$ and $s = 1.5$). From (11), (12), and (13) we get

$$\bar{I}_p = I_{p0}\left(1 + \frac{I_{p0}}{\eta I_{t0}}\right)^{-1/2} \quad \bar{I} = \frac{1}{4}\frac{\omega_g}{\omega_t}\frac{I_{g0}(I_{p0} - \bar{I}_p)^2}{I_{p0}\bar{I}_p}$$
$$\bar{I}_t = \frac{1}{4}\frac{I_{t0}(I_{p0} + \bar{I}_p)^2}{I_{p0}\bar{I}_p}. \tag{16}$$

It follows from (16) that at $\Lambda_{opt} = 0.21\eta^{-1}$ there takes place an optimal conversion regime, and the optimal conversion coefficient relative to the energy of the triggering field is

$$\bar{K}_t^{opt} = \bar{I}_g^{opt}/I_{t0} \approx 0.2\omega_g/\omega_t, \tag{17}$$

and relative to the energy of the pump field is

$$\bar{K}_p^{opt} = \bar{I}_g^{opt}/I_{p0} \approx 0.17 s^{-1}\sqrt{\omega_g/\omega_t}. \tag{18}$$

At $\omega_g/\omega_t = 7$ we have $\bar{K}_t^{opt} = 1.4$ and $\bar{K}_p^{opt} = 0.31$.

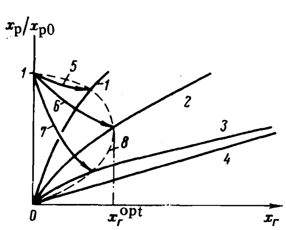

FIG. 2. Generation of the difference frequency at $\delta k = 0$. Curves 1, 2, 3, 4—curves of the states of equilibrium for $\Lambda_1 > \Lambda_2 > \Lambda_3 > \Lambda_4 = 0$, respectively ($\Lambda = I_{t0}/I_{p0}$); 5, 6, 7—integral curves for Λ_1, Λ_2, and Λ_3, respectively; 8—geometric locus of the points of equilibrium states.

4. GENERATION OF COMBINED FREQUENCY

Just as in the generation of the difference frequency, we consider first the case of exact phase matching of the interacting waves. From (2.c) we see readily that a value $\Theta = \pi$ is established at $z = 0$. However, with increasing field x_g, the term proportional to x_g/x_t and entering with a negative sign in the coefficient W of $\sin\Theta$ can exceed the sum of the remaining terms in this coefficient, and this changes the phase difference from $\Theta = \pi$ to $\Theta = 0$; then the inverse process of parametric conversion of the field x_g into x_t will set in.

A rigorous solution of the problem calls for simultaneous solution of Eqs. (2), which is impossible. We seek an approximate solution of (2), assuming $\Theta = \pi$, and then use this solution to find the values of the fields at which the jump of Θ takes place.

The equations for the field amplitudes at $\Theta = \pi$ take the form (9), but the sign of the right-hand side of (9b) must be reversed. The first and second integrals and the curves for the stable equilibrium states take the form

$$I_t = I_{t0} - \frac{\omega_t}{\omega_g}I_g, \tag{19}$$

$$I_g = \frac{\omega_g}{\omega_t}I_{t0}\sin^2\ln\left(\frac{I_{p0}}{I_p}\right)^{\xi/2}, \tag{20}$$

$$\bar{I}_p = s\left[I_g\left(I_{t0} - \frac{\omega_t}{\omega_g}I_g\right)\right]^{1/2}. \tag{21}$$

Substituting (19) and (20) in Eqs. (2c) for the phase difference, we obtain the following expression for the coefficient W of $\sin\Theta$:

$$W = Af_n s\sqrt{\frac{\omega_g}{\omega}}I_{t0}\left[\sin\ln\left(\frac{I_{p0}}{I_p}\right)^{\xi} + \xi\frac{I_p}{2\eta I_{t0}}\operatorname{ctg}\ln\left(\frac{I_{p0}}{I_p}\right)^{\xi}\right]. \tag{22}$$

The function W changes from $+\infty$ to $-\infty$ when $\ln(I_{p0}/I_p)^{\xi}$ changes from zero to π, and passes through zero in the region of values $\pi/2 < \ln(I_{p0}/I_p)^{\xi} < \pi$. In the region $W > 0$ there exist two different conversion regimes, which can be easily determined by plotting the integral

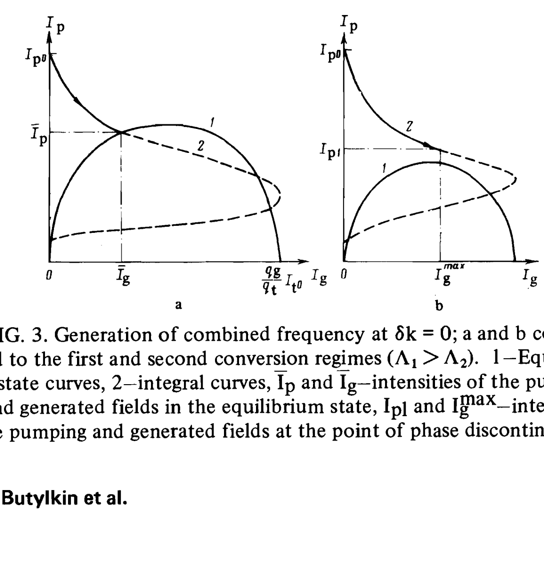

FIG. 3. Generation of combined frequency at $\delta k = 0$; a and b correspond to the first and second conversion regimes ($\Lambda_1 > \Lambda_2$). 1—Equilibrium-state curves, 2—integral curves, $\bar{I}_p$ and $\bar{I}_g$—intensities of the pumping and generated fields in the equilibrium state, I_{pl} and I_g^{max}—intensities of the pumping and generated fields at the point of phase discontinuity.

curves (20) and the stationary-state curves (21) at different ratios $\Lambda = I_{t0}/I_{p0}$ (Fig. 3).

A. The first regime takes place if the following condition is satisfied:

$$2\eta I_{t0} > I_{p0} \exp(-\pi/2\xi). \qquad (23)$$

In this regime, the interacting fields reach a stationary state, and the process in question evolves in analogy with the generation of the difference frequency. The equilibrium value of the pump field is determined from the equation

$$\bar{I}_n - 2\eta I_{t0} \sin \ln (I_{p0}/\bar{I}_p)^{\xi} = 0. \qquad (24)$$

It can be seen from (20) and (21) that the maximum conversion coefficients in terms of the triggering-field energy

$$K_t = \bar{I}_g / I_{t0} = \omega_g / 2\omega_t \qquad (25)$$

and in terms of the combined energy of the triggering and pumping fields

$$K = \frac{\bar{I}_g}{I_{t0} + I_{p0}} = \frac{\omega_g}{2\omega_t} \frac{\exp(-\pi/2\xi)}{2\eta + \exp(-\pi/2\xi)} \qquad (26)$$

occur at a definite value of Λ:

$$\Lambda_{opt} = \exp(-\pi/2\xi)/2\eta \qquad (27)$$

B. At

$$2\eta I_{t0} < I_{p0} \exp(-\pi/2\xi) \qquad (28)$$

the condition W = 0 is reached before the interacting fields reach the equilibrium state. The generated field is maximal at a value $I_p = I_{p1}$ determined by solving the equation W = 0. If the inequality (28) is strong (weak triggering fields), then the inverse parametric conversion of the generated wave into a pump field is negligible, so that the interaction can be described by assuming that the pump is changed only as a result of the two-phonon absorption. Only in this case, as can be easily shown, is the maximum conversion coefficient K_t independent of I_{t0}. In fact, in this situation the main contribution to W is made by the second term of (22), and the phase discontinuity, and consequently also the maximum field, occur at a value $\ln(I_{p0}/I_p)^{\xi} = \pi/2$; the maximum conversion coefficients are determined here by expressions (25) and (26) at arbitrary values of I_{t0}, with $K \ll 1$. We shall show below that only under this situation are the results obtained in[5] valid.

5. EFFECT OF WAVE MISMATCH

We have considered above a model with $\delta k = 0$. We have shown within the framework of this model that maximum conversion coefficients can be obtained if the interaction length is sufficient to attain fields close to stationary. In real media usually $\delta k \neq 0$ and it may turn out that the linear-synchronism length $(\delta k)^{-1}$ is smaller than the distance over which the stationary state would be reached at $\delta k = 0$. It turns out that, under certain definite conditions coherent interaction of the light waves can be effected in resonant four-wave processes over lengths exceeding the linear-synchronism length; consequently, fields close to the stationary values (14) and (24) can be reached also when $\delta k \neq 0$.

In fact, at $W > \delta k$ (W is the coefficient of $\sin \Theta$ in the phase equation (2c)), phase locking of the interacting fields takes place in the system[13]; if $W|_{z=0} \gg \delta k$, the system arrives rapidly at a state with $\Theta \approx \pi + \delta k/W$, and the process can be analyzed approximately by assuming $\delta k = 0$ in the first stage. The conditions under which phase locking ensures the attainment of fields close to stationary can be obtained for the case of generation of the combined frequency by using expression (22) for $W(I_p)$.

When the difference frequency is generated, an expression for $W(I_p)$ can be obtained by substituting x_g and x_t from (11) and (12) in Eq. (2c):

$$W(I_p) = L_{TPA}^{-1} f_n \left(\frac{1-y^2}{y} \eta\Lambda + \xi^{1/2} \frac{1+y^2}{1-y^2} y^{-1/\xi} \right). \qquad (29)$$

Here $L_{TPA} = (2AI_{p0})^{-1}$ is the characteristic two-photon absorption length, and $y = (I_p/I_{p0})^{\xi}$. At the point of incidence (z = 0) the function (29), just like (22), becomes infinite. It follows therefore that phase locking always takes place during the initial stage of the interaction. The phase locking can stop either before or after the equilibrium state for $\delta k = 0$ is reached, depending on the value of δk and on the initial field intensities. An approximate value of the pump intensity at which the stopping takes place is determined from the equation

$$W(I_p^{stop}) = \delta k.$$

Obviously, the stopping of the phase locking will not occur before the stationary state is reached if $W(\bar{I}_p) > \delta k$.

Let us consider by way of example a situation where $\xi = 1$. The expression for $W(I_p)$ in the case of difference-frequency generation can be expressed in terms of the limiting intensities I_{p0} and I_{t0}, by using (16) and (29). Then the range of values of I_{p0} and I_{t0} at which phase locking ensures fields close to the equilibrium values (16) is given by the inequality

$$W(\bar{I}_p) = 2L_{TPA}^{-1} [\eta\Lambda(1+\eta\Lambda)]^{1/2} \geqslant \delta k. \qquad (30)$$

After the fields attain values close to equilibrium, they will attenuate slowly. Indeed, inasmuch as in this region $\Theta \approx \pi + (\delta k)/W(\bar{I}_p)$, we can easily find from (2a) that the pump varies like

$$I_p \approx \bar{I}_p \left[1 + \left(\frac{\delta k}{W} \right)^2 \frac{z}{2\bar{L}_{TPA}} \right]^{-1}, \qquad (31)$$

i.e., the characteristic length of its damping is $(W/\delta k)^2$ times larger than the length of two-photon absorption $\bar{L}_{TPA} = (2A\bar{I}_p)^{-1}$. With the aid of (31), (11), and (2b) we can easily show that the generated and triggering fields will also be damped in the region of values $x_g \approx \bar{x}_g$ and $x_t \approx \bar{x}_t$, but somewhat more slowly than the pump field. The fields behave slowly when the combined frequency is generated.

At large wave mismatches δk and at low intensities of the incidence fields, when (30) is replaced by a strong inverse inequality, the stopping of the phase locking occurs far from the equilibrium state. In this case one can neglect the reverse transfer of the generated and triggering fields to the pump field, and conversion takes place over a length $\sim \pi|\delta k|^{-1}$. If furthermore the two-photon absorption of the pump field (and consequently also of the fields x_g and x_t) is small over this length, then the field intensities behave in the same manner as in nonresonant four-wave interactions. Only in this case are the results of[5] valid. We note that the efficiency of generation of both the combined and the different frequencies is low in this case (K_p, $K_t \ll 1$).

In concluding this section, we note that the small deviation of the parameter F (see (10)) from unity results in the same modification of the equations for the field amplitudes as the deviation of the phase difference Θ from π as a result of incomplete synchronism. Therefore at $1 - F \ll 1$ the solution of Eqs. (2a) and (2b) changes little in a region far from the stationary solutions of Eqs. (9). After the fields reach values close to equilibrium, the deviation of F from unity leads to a damping of the fields even in the case when $\delta k = 0$[3).

6. CONVERSION LENGTH AND THE INFLUENCE OF POPULATION SATURATION

We define the conversion length L as the distance over which a specified conversion coefficient (or a specified value of I_g, say $I_g = 0.9\bar{I}_g$) is reached. It follows from (9a) that

$$L=\frac{1}{2A}\int_{I_{p1}}^{I_{p0}}\frac{dI_p}{I_p(I_p-s\sqrt{I_gI_t})f_n(I_p,I_g,I_t)} \qquad (32)$$

(here I_{p1} corresponds to a specified value I_g). The integral in (32) can be calculated by using the integrals of motion (11) and (12) or (19) and (20) for the respective cases of generation of the difference and combined frequencies.

Let us consider, for example, the generation of the difference frequency in a medium consisting of immobile oriented molecules. In such a medium we have

$$f_n=[1+I_{sat}^{-2}(I_p-s\sqrt{I_gI_t})^2]^{-1}, \qquad (33)$$

where

$$I_{sat}^{1/2}=\hbar(2|r_1|)^{-1/2}(\tau d)^{-1/4} \qquad (34)$$

is the amplitude of the saturating field. Substituting (11), (12), and (33) in (32) and integrating, we get in the particular case $\xi = 1$

$$L=L_1+L_2=\frac{L_{TPA}}{2\sqrt{\eta\Lambda}(1+\eta\Lambda)}\ln\left[\frac{I_{p0}-\bar{I}_p}{I_{p0}+\bar{I}_p}\frac{I_{p1}+\bar{I}_p}{I_{p1}-\bar{I}_p}\right]+L_{TPA}\eta\Lambda\frac{I_{p0}^2(I_{p0}-I_{p1})(I_{p1}I_{p0}-\bar{I}_p^2)}{I_{sat}^2\bar{I}_p^2I_{p1}}. \qquad (35)$$

The quantity $\bar{I}_p$ in this equation is determined from (16). The first term (L_1) of (35) is the conversion length in a medium without saturation, while the second (L_2) describes the increase of the conversion length as a result of the saturation of the populations. The contribution of L_2 to the conversion length is appreciable if $I_{p0} \gg I_{sat}$.

Obviously, the influence of population saturation on the four-wave interaction process in gaseous media is qualitatively the same as the medium of oriented molecules. For gases, however, it is possible to use (32) to calculate the conversion length only when the fields do not exceed the saturation value and $f_n \approx 1$; in this case the conversion length coincides with L_1.

It was shown above that $\delta k \neq 0$ it is possible to obtain fields close to $\bar{I}_p$, $\bar{I}_g$, and $\bar{I}_t$ in the presence of phase locking. In this case, the conversion length is also described by expression (32) (or (35)). We can therefore state that phase locking is realized, then the population saturation does not change the conversion coefficient, but leads only to an increase of the conversion length. The latter may turn out much larger than the linear-synchronism length.

It is stated in [5] that the conversion coefficient is limited by saturation. This statement is valid only when the phase locking is stopped long before the stationary state is reached. In this case the population saturation decreases the local effectiveness of the interaction, and the conversion takes place only over the linear-synchronism length.

7. NUMERICAL ESTIMATES AND CONCLUSIONS

Let us stop to discuss the optimal generation of the combined and different frequencies in vapors and gases. We formulate first the conditions that must be satisfied to realize such a regime.

The ratio of the incident values of the triggering and pumping fields x_{t0}/x_{p0} should satisfy the conditions (15) and (27) respectively in the case of generation of the difference and combined frequencies. The field values x_{t0} and x_{p0} should be sufficient to ensure phase locking up to a generated-field value close to equilibrium in the case of exact synchronism ($\delta k = 0$). To this end it is necessary to satisfy the condition $W(I_{t0}, I_{p0}, \bar{I}_p) > (\delta k)$, where W is determined by expressions (29) and (22), while $\bar{I}_p = \bar{x}_p^2$ is a solution of Eqs. (14) and (24).

If simultaneous satisfaction of the conditions for the optimal ratio x_{t0}/x_{p0} and phase locking is impossible because the power of one of these fields is insufficient, then it is desirable to increase the power of the second field to a level at which $W \approx \delta k$.

As the first example, we consider the possibility of generation of a difference frequency in HgI vapor under the influence of radiation of a neodymium laser tuned to a wavelength 1.075 μ [15] and its fourth harmonic; for the latter, two-photon resonance is obtained between the ground state $6s^2S$ and $8s'S$ ($\hbar\omega_{21} = 74404.6$ cm^{-1} [16]). We assume for the estimates the values $\langle r_1^2\rangle \approx 10^{-100}$ cgs esu and s = 1.5. Then the solution (16) is valid. At a pressure 10 Torr (t = 200° C, N = 2.7×10^{17} cm^{-3} [17]) and $\tau = 5 \times 10^{-9}$ sec we have $d = 2 \times 10^{-10}$ sec and $A = 4 \times 10^{-6}$ cgs esu [see (8) and (5b)]. If k = 0.2 cm^{-1}, then at an incident pump power 75 MW/cm^2 the optimal generation regime is obtained at a triggering field power 15 MW/cm^2 and the phase-locking condition (30) is satisfied. The pump conversion coefficient (18) is equal to $K_p = 0.9\bar{K}_p^{opt} \approx 0.28$, and the triggering-field coefficient is $K_t = 0.9\bar{K}_t^{opt} \approx 1.25$. We note that in this case the pump intensity is smaller by a factor 2.2 than the saturation value.

Let us consider the generation of a summary frequency in MgI under the influence of the radiation from two dye lasers [5] (at λ_p = 4597 Å, two photon absorption takes place between the states $3s^2S$ and $4s'S$). If $\omega_g/\omega_t = 10$, then the quantities $\langle r_1^2\rangle$ and $\langle r_2^2\rangle$, estimated from formulas (6) and (7), with allowance for the contribution made to the two-photon absorption and the SRS by the states $3p'P^0$ and $4p'P^0$, are equal to $\sim 1.4 \times 10^{-99}$ and 2×10^{-98} cgs esu, respectively; then s = 3.8 and $\xi = 2.7$. At a vapor pressure 10 Torr and $\tau \approx 5 \times 10^{-9}$ we get $d \approx 8 \times 10^{-11}$ sec and $A = 3.6 \times 10^{-6}$ cgs esu. At[4) $\delta k = 0.3$ cm^{-1}, I_{p0} = 25 MW/cm^2 and $I_{t0} \approx 2.7$ MW/cm^2, the first conversion regime is realized (see Sec. 3), phase locking ensures a state close to equilibrium, and the conversion coefficients with respect to the combined energy of the triggering field and the pump are $\bar{K} = 0.31$

Conversion coefficients and conversion and stopping lengths in the generation of the difference frequency in NeI exposed to the ninth harmonic of a neodymium laser and to a laser with a rhodamine-6Zh solution

Λ	$\bar{K}_t$, %	$\bar{K}_p$, %	K_t, %	K_p, %	L, cm	L_{stop}, cm
0.167	184	31	38	6.5	0.96	0.21
0.66	41.8	27.5	38	24	0.77	0.77
1.33	11.5	15	10	13.5	0.40	1.85
2	6.5	12	5.8	10.8	0.25	6.25

Here $\Lambda = I_{t0}/I_{p0}$; L is the length in which the conversion coefficient $K_t = 0.9\bar{K}_t$ ($K_p = 0.9\bar{K}_p$) is reached; t_{stop} is the length over which phase locking stops.

and $\bar{K}_t = 3.5$. The conversion length corresponding to $K = 0.9\bar{K}$ is 15 cm.

In conclusion, let us estimate the possibility of generation of the difference frequency in NeI ($2p^{6\prime}S \to 5p''[1\frac{1}{2}]$) exposed to the ninth harmonic of a neodymium laser and to a laser with a solution of rhodamine-6Zh (to satisfy the two-photon resonance condition it is necessary to change the neodymium laser tuning by 32 cm^{-1} [15].) We put $N = 10^{20}$ cm^{-3}, $d \approx 10^{-11}$ sec, $\langle r_1^2\rangle \approx 10^{-102}$ cgs esu, and $s = 1.65$. Then $\xi = 1$ and the solution (16) is valid. We calculate the conversion coefficients $\bar{K}_p$ and $\bar{K}_t$ at $\delta k = 0$, and K_p and K_t at $\delta k = 5$ cm^{-1} and a pump power 375 MW/cm^2, and also the conversion length L and the stopping length of the phase locking L_{stop} for different ratios $\Lambda = x_{t0}/x_{p0}$. The results of the calculations are summarized in the table. It is seen from the table that at $\Lambda = 0.167$ the stationary value of x_g is maximal, but it is not reached, since the phase-locking is stopped before that. The optimal regime at $\delta k = 5$ cm^{-1} corresponds to $\Lambda = 0.66$. Further increase of Λ leads to a decrease of the conversion coefficient; the phase locking then makes it possible to realize coherent interaction at lengths greatly exceeding $(\delta k)^{-1}$. We note that the conversion coefficient in this regime depends little on the length of the chamber with the working gas.

Thus, the last example demonstrates the possibility of producing a tunable source in the range 650—656 Å. To trigger it (at $\Lambda = 0.66$) in a system with confocal focusing [18], the required pump power is 1.6 kW and the required triggering field power is 6 kW.

[1]The role of phase locking in parametric generation of the anti-Stokes component of SRS was demonstrated theoretically and experimentally in [8].

[2]Since practical interest attaches to the interaction of waves with small wave mismatch $|\delta_k| \ll |k_j|$, it is assumed in (2) and henceforth that $\langle d_1\rangle = \langle d_2\rangle = d$.

[3]A similar phenomenon takes place in resonant frequency doubling in gases [14].

[4]The wave mismatch can be varied in certain limits by introducing inert gases into the working medium.

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