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RESONANCE PARAMETRIC INTERACTION OF STRONG FIELDS AT OPTICAL FREQUENCIES

G. L. GUREVICH and Yu. G. KHRONOPULO

Institute of Radio Engineering and Electronics, Academy of Sciences, U.S.S.R.

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Parametric interaction of three intense electromagnetic modes having frequencies ω_1, ω_2, and ω_3 is considered. The frequencies satisfy the condition $\omega_1 + \omega_2 = \omega_3$, the last one being the absorption frequency of the medium. Equations are derived describing the interaction both in the presence and the absence of population inversion and taking the saturation effect into account. Qualitative differences between the resonance and non-resonance interaction are considered. Generation of sum frequency fields and parametric frequency division are investigated. Analytical expressions are found for the resulting fields; the field values significantly depend upon two-photon absorption of fields $E(\omega_1)$ and $E(\omega_2)$. The maximum coefficient of conversion of the fields $E(\omega_1)$ and $E(\omega_2)$ into $E(\omega_3)$ is also determined. It is shown that the length characterizing an appreciable energy transfer between the interacting modes depends on the lifetime of particles of the working substance in the excited state. The resonance parametric interaction is found to be less critical with respect to synchronization conditions than non-resonance interaction. Numerical computation is made for the case when the working substance consists of impurities in a dielectric.

1. Parametric interaction of non-resonance monochromatic electromagnetic modes in a dispersive medium is well known in terms of theory and experiment (see, for example,[1-4]). The parametric interaction of waves having one mode frequency close to the natural frequency of the medium has been studied much less.[5-8] The mathematical treatment in all the above papers was based on a perturbation theory that is applicable only if the interacting field amplitudes are sufficiently small;[4] furthermore, neither two-photon absorption nor the population saturation effect (which may become significant in these processes, as will be shown below), were taken into account. For example, in the case of resonance at the second harmonic it is necessary to consider the two-photon absorption of the incident wave energy at the fundamental frequency as well as the absorption of the second-harmonic field. In particular, the two-photon absorption intensity determines the value of the doubled-frequency field that can be reached in generation, and the lifetime of excited particles of the working substance determines the length associated with the energy transfer between the interacting fields.

The averaging method was used in[9,10] to obtain equations that allowed for the saturation effect and described both the Raman scattering and the parametric interaction of fields when some fields had resonance frequency. These equations will be used in our work.

2. We consider an interaction of three frequencies

$$E_l \exp [i(\omega_l t - k_l z)] + \text{c.c.} \tag{1}$$

(here, $l = 1, 2, 3$, $k_l = 2\pi/\lambda_l$, λ_l is the wavelength, $\omega_3 = \omega_2 + \omega_1$; and $E_l = m_l(z) \exp[-i\varphi_l(z)]$) propagating along the z axis in a medium[1)] of particles whose energy levels contain levels 2 and 1, such that

$$\omega_3 = \omega_2 + \omega_1 = \omega_{21} + \Delta. \tag{2}$$

Here, ω_{21} is the frequency of transition $2-1$, and $0 \le \Delta \le \omega_1, \omega_2$. Henceforth we assume that level 1 corresponds to the ground state of the particles.

The complex polarization amplitude $\mathbf{P}(\omega_l)$ at the frequency ω_l is readily defined for our stationary process with the aid of Eqs. (4), (15), and (18)[2)]

[1)]The medium is considered isotropic for simplicity. Anistropy is taken into account in the same manner as in the case of the parametric interaction of nonresonance fields (see [3]).

[2)]Equation (18) of [9] contains an error: the right-hand side should be multiplied by i.

of[9] and Eq. (1) of[10]. For simplicity, a dipole field—matter interaction is considered and the fields $\mathbf{E}_3$, $\mathbf{E}_2$, and $\mathbf{E}_1$ are considered polarized in the directions a, b, and c respectively. The projections of the complex amplitudes of polarization on these directions are then determined as follows:

$$P_{c,b}(\omega_{1,2})e^{i\omega_{1,2}t} = \mathrm{Sp}_{c,b}\{\hat{p}\hat{\rho}(\omega_{1,2})\}$$
$$= \sum_q [p_{(c,b)q1}\rho_{1q}(\omega_{1,2}) + p_{(c,b)2q}\rho_{q2}(\omega_{1,2})],$$
$$P_a(\omega_3)e^{i\omega_3 t} = p_{a21}\sigma_{12}e^{i(\omega_3-\Delta)t} \tag{3}$$

Here, $\hat{p}$ is the dipole moment operator, and p_{cmn}, p_{bmn}, and p_{amn} are the projections of its matrix elements on the directions of fields E_1, E_2, and E_3. The expressions for matrix elements ρ_{1q} and ρ_{q2} of the density matrix $\hat{\rho}$ are determined by Eqs. (15) of[9] (where we set j = 1, i = 2, $\alpha = 1$, $\beta = 2$, and $\Delta\omega_{\alpha,\beta} = \Delta$), and σ_{12} is obtained from Eq. (18) of[9], taking account of Eq. (1) of[10]. As a result, we have

$$P_c(\omega_1) = -d[\hbar^{-2}p_{a12}r^*E_3E_2^*e^{i(k_2-k_3)z}$$
$$+\hbar^{-3}|r|^2|E_2|^2E_1e^{-ik_1z}]Nn, \tag{4a}$$
$$P_b(\omega_2) = -d[\hbar^{-2}p_{a12}r^*E_3E_1^*e^{i(k_1-k_3)z}$$
$$+\hbar^{-3}|r|^2|E_1|^2E_2e^{-ik_2z}]Nn, \tag{4b}$$
$$P_a(\omega_3) = -d[\hbar^{-2}p_{a21}rE_1E_2e^{-i(k_1+k_2)z}$$
$$+\hbar^{-1}|p_{a21}|^2E_3e^{-ik_3z}]Nn, \tag{4c}$$

where

$$r = \sum_q \left(\frac{p_{c1q}p_{bq2}}{\omega_{q2}+\omega_2} + \frac{p_{cq2}p_{b1q}}{\omega_{q2}+\omega_1}\right)$$

$$d = \left[T^{-1} + i\left(\Delta - \sum_{l=1}^{3}\Omega_l|E_l|^2\right)\right]^{-1} \tag{5}$$

r characterizes the intensity of two-photon absorption, N is the particle density, n is the population difference between levels 1 and 2 for a single particle:

$$n = n_0\{1 + 4\hbar^{-4}[\hbar^2|p_{a21}|^2|E_3|^2$$
$$+ 2\hbar\,\mathrm{Re}\,(p_{a21}rE_1E_2E_3^*e^{i(k_3-k_1-k_2)z})$$
$$+ |r|^2|E_1|^2|E_2|^2\tau][T^{-2} + (\Delta - \Sigma\Omega_l|E_l|^2)^2]^{-1}T^{-1}\}^{-1}, \tag{6}$$

n_0 is the equilibrium population difference, τ is the particle lifetime on level 2, T^{-1} is the linewidth of transition 2—1, and $\Sigma\Omega_l|E_l|^2$ determines the change in frequency ω_{21} due to the fields E_l. The coefficients Ω_l can be found from Eq. (1) of[10]:

$$\Omega_l = \frac{2}{\hbar^2}\left\{\frac{|(\mathbf{p}_{21}\mathbf{E}_l)|^2}{\omega_{21}+\omega_l}\delta_{l3} + \sum_q\left[\frac{\omega_{q1}|(\mathbf{p}_{1q}\mathbf{E}_l)|^2}{\omega_{q1}^2-\omega_l^2} + \frac{\omega_{2q}|(\mathbf{p}_{q2}\mathbf{E}_l)|^2}{\omega_{q2}^2-\omega_l^2}\right]\right\} \tag{7}$$

(δ_{l3} is the Kronecker delta).

Let us now consider Eqs. (4a)—(4c). The first terms in the numerators of (4a)—(4c) define the part of polarization due to the parametric interaction of the fields, while the second terms in (4a) and (4b) are associated with the intensity of two-photon processes. Let us note that the quantity r defining this intensity is also included in the first part of polarization.

In a general case the "non-resonance" part of the interaction should also be included in the computation of $P(\omega_l)$; for this purpose, the corresponding terms

$$\chi_{cab}E_3E_2^*e^{i(k_2-k_3)z},\quad \chi_{bac}E_3E_1^*e^{i(k_1-k_3)z},\quad \chi_{abc}E_1E_2e^{-i(k_1+k_2)z}, \tag{8}$$

are added to (4a)—(4c). Here, χ_{abc} etc. coincide with the components of the susceptibility tensor (see expressions (22.23) in[4]) without the resonance terms.[3)]

Of interest is the case in which the resonance part of polarization plays the major role; according to (4) and (8), this calls for

$$|p_{a21}r| \gg \hbar^2T^{-1}|\tilde{\chi}|, \tag{9}$$

where $\tilde{\chi}$ is the "non-resonance" part of the susceptibility of a single particle. The above inequality is satisfied if, for example, a two-photon transition between levels 1 and 2 is well resolved (for optical frequencies $|r| \approx 10^{-51}$ cgs esu), while a single photon transition is resolved only up to the magnetic dipole approximation ($|p_{a21}| \gtrsim 10^{-21}$ cgs esu).[4)]

In fact, assuming that χ has the value of the susceptibility of KDP ($\tilde{\chi} \approx 10^{-31}$ cgs esu[3]) and that $T \approx 10^{-11}$ sec, we find that the left-hand side of (9) is more than 100 times larger than the right-hand side. Furthermore, from now on we will neglect the frequency variation of transition 2—1 as compared to the linewidth T^{-1}. It follows from (7) that this is justifiable when $E < 3 \times 10^3$ cgs esu ($\sim 10^6$ V/cm) for the parameters given above.

Taking all the above considerations into account, we use Eq. (4) to find four equations for the real amplitudes $m_l(z)$ and phase differences $\varphi_1(z) + \varphi_2(z) - \varphi_3(z) + (\delta k)z = \theta(z)$[3,4], where $\delta k = k_1 + k_2 - k_3$:

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3)Expressions (8) are obtained by inserting in (3) the values of $\sigma_{mn}^{(2)}$ taken from (6) of [9].

4)In this case, p_{21} and E_3 denote the matrix element of the magnetic moment operator and the magnetic field amplitude respectively. All the p_{mn} will henceforth be considered real. Consideration of p_{mn} as a complex quantity does not yield qualitatively new results and merely renders the exposition more difficult.

$$\frac{dm_{1,2}}{dz}+\beta_{1,2}m_{1,2}=-[(B_{1,2}'\sin\theta+B_{1,2}''\cos\theta)m_{2,1}m_3+b_{1,2}''m_{2,1}^2m_{1,2}]n, \qquad (10a,b)$$

$$\frac{dm_3}{dz}+\beta_3m_3=[(B_3'\sin\theta-B_3''\cos\theta)m_1m_2-b_3''m_3]n, \qquad (10c)$$

$$\frac{d\theta}{dz}=\delta k+\left[\left(B_1''\frac{m_2m_3}{m_1}+B_2''\frac{m_1m_3}{m_2}+B_3''\frac{m_1m_2}{m_3}\right)\sin\theta+\left(B_3'\frac{m_1m_2}{m_3}-B_2'\frac{m_1m_3}{m_2}-B_1'\frac{m_2m_3}{m_1}\right)\cos\theta-(b_1'm_2^2+b_2'm_1^2)+b_3'\right]n. \qquad (10d)$$

The following notation is used in (10a)–(10d): β_l is the linear attenuation coefficient for the l-th field in the medium,

$$\begin{aligned}
&B_l=B_l'+iB_l''=\alpha_lg,\\
&\alpha_l=\pi\omega_l^2N/2c^2k_l, \quad g=4p_{a21}rT(i+T\Delta)/\hbar^2(1+T^2\Delta^2),\\
&b_l=b_l'+ib_l''=\alpha_la_l,\\
&a_1=a_2=4r^2T(i+T\Delta)/\hbar^3(1+T^2\Delta^2),\\
&a_3=4p_{a21}^2T(i+T\Delta)/\hbar(1+T^2\Delta^2).
\end{aligned} \qquad (11)$$

Let us note that the resonance absorption of field E_3 in the medium is defined by the term $b_3''m_3n$ in (10c).

Using the above notation, the population difference can be described by

$$n=n_0[1+\hbar^{-1}\tau(a_3''m_3^2+2g''m_1m_2m_3\cos\theta+a_1''m_1^2m_2^2)]^{-1}. \qquad (12)$$

It should be remembered that Eqs. (10) are valid when $\Delta\ll\omega_2,\omega_1$ and conditions (9) are satisfied.

In a general case an analytic solution of (10) is not possible. Nevertheless, the qualitative differences between resonance and non-resonance parametric interaction can be readily observed in the case of $\delta k=0$ and $\Delta=0$. Moreover, this example permits us to evaluate the maximum effectiveness of the resonance parametric conversion (the case of $\delta k\neq0$ will be considered in Sec. 5).

When $\delta k=\Delta=0$, Eq. (10d) assumes the form

$$\frac{d\theta}{dz}=\left(B_1''\frac{m_2m_3}{m_1}+B_2''\frac{m_1m_3}{m_2}+B_3''\frac{m_1m_2}{m_3}\right)n\sin\theta. \qquad (13)$$

It can be readily seen that the plane $\theta=\pi$ is stable when $n>0$ (the working level populations are not inverted).

It is useful to analyze the process of generating one of the E_l fields when its intensity at the boundary is close to zero (or, more precisely, when its intensity is determined by noise). According to (10) and (13) the rate of approach of the phase difference towards the plane $\theta=\pi$ is then much higher than the rate of change of field amplitudes m_l.

Therefore, we can assume that $\theta=\pi$ in (10a)–(10c). Introducing new variables we obtain

$$x_{1,2}=(\alpha_{2,1}a_1''\tau/\hbar\alpha_{1,2})^{1/4}m_{1,2}=(4r^2\omega_{2,1}\tau T/\hbar^4\omega_{1,2})^{1/4}m_{1,2}, \qquad (14a)$$

$$y=(\hbar^{-1}a_3''\tau)^{1/2}m_3=2\hbar^{-1}p_{a21}(\tau T)^{1/2}m_3, \qquad (14b)$$

$$\eta=(\alpha_1\alpha_2\hbar a_1''\tau^{-1})^{1/2}n=\pi r\omega_1\omega_2NT^{1/2}n/c^2\hbar(k_1k_2\tau)^{1/2} \qquad (14c)$$

and using (12), we write (10a)–(10c) for $\delta k=\Delta=0$ in the form

$$dx_{1,2}/dz+\beta_{1,2}x_{1,2}=x_{2,1}(y-x_1x_2)\eta, \qquad (15a,b)$$

$$dy/dz+\beta_3y=A(x_1x_2-y)\eta, \qquad (15c)$$

$$\eta=\eta_0/[1+(y-x_1x_2)^2], \quad \eta_0=\eta\quad(n=n_0), \qquad (15d)$$

$$A=2p_{a21}^2r^{-1}\omega_3(T\tau/\omega_1\omega_2)^{1/2}. \qquad (16)$$

3. We consider frequency doubling

$$\omega_1=\omega_2=\omega,\quad \omega_3=2\omega. \qquad (17)$$

Then $x_1\equiv x_2$.

Let us note that the plane $x_1=x_2$ is stable when $\beta_1=\beta_2$. This is readily demonstrated by multiplying (15a) and (15b) by x_1 and x_2 respectively and subtracting one equation from the other. For example, the condition $\beta_1=\beta_2$ is satisfied if ω_1 is little different from ω_2. Therefore all the relations obtained for case (17) in terms of the variables x, y, and η, are also valid for the interaction of fields with close frequencies ω_1 and ω_2. In this case the amplitudes m_1, m_2, and m_3 must be determined from (14).

Thus let us assume that $x_1=x_2=x$, neglecting linear losses from now on. Then we obtain from (15a)–(15d)

$$\frac{dx}{dz}=\frac{x(y-x^2)\eta_0}{1+(y-x^2)^2}, \qquad (18a)$$

$$\frac{dy}{dz}=A\frac{(x^2-y)\eta_0}{1+(y-x^2)^2}, \qquad (18b)$$

where, if (17) holds, x and y are determined as before by (14a) and (14b); the amplitudes of the first and second harmonic will be denoted by m and M respectively.

$$A=2p_{a21}^2r^{-1}(\tau T)^{1/2},\qquad \eta=2\pi\omega^2rNnT^{1/2}/c^2\hbar k\tau^{1/2}. \qquad (19)[5]$$

[5]A direct check will show that $P(\omega)$ derived similarly to (4 a) and (4 b) is twice as large as $P(\omega_1=\omega, E_1=E)$ obtained from (4 a) in the case of degenerate frequencies $\omega_1=\omega_2=\omega$. The coefficients in (10 a) increase in a corresponding manner, thus changing η and A. We obtain a smooth transition from (14) to (19) when $\omega_1\to\omega_2$ by allowing for the slow motions $\sim\exp[i(\omega_1-\omega_2)t]$ in the derivation of the initial shortened Eqs. (10) of [9] and Eq. (1) of [10] for $\omega_1-\omega_2\approx T^{-1}$; then $P(\omega_1)$ in (4 a), for example, will acquire the additional terms $\sim E_3E_1^*$ and $|E_1|^2E_1$ which lead to the above increase of $P(\omega)$ when $\omega_1\to\omega_2$.

Dividing (18a) by (18b) we obtain the first integral

$$y - y_0 = A \ln (x_0 / x), \quad (20)$$

where

$$x_0 = x|_{z=0}; \qquad y_0 = y|_{z=0}. \quad (21)$$

It also follows from (18) that the ratio of amplitudes of the second and first harmonics tends to the following limit,[6)] regardless of the boundary values (21):

$$y_\infty / x_\infty^2 = 1, \quad (22)$$

with $\eta = \eta_0$ ($n = n_0$). Consequently in the presence of an incident field of frequency ω the population difference n near the boundary is less than the equilibrium value because of the two-photon absorption of field $E(\omega)$ and the single-photon absorption of field $E(2\omega)$, which occur in the system as a result of the synchronization conditions. $E(2\omega)$ grows in the z direction and the parametric interaction of $E(2\omega)$ with $E(\omega)$ increases n and returns it to the equilibrium value.

Using (20) and (22) we now find the limiting values of the field amplitudes established in the course of the interaction

$$y_\infty + {}^1\!/_2 A \ln y_\infty = x_\infty^2 + A \ln x_\infty = y_0 + A \ln x_0. \quad (23)$$

Let us consider the generation of the second harmonic $y_0 = 0$ ($M_0 = 0$). From (23) we find m_0, which is the boundary value of the first-harmonic amplitude necessary to obtain M_{max}, the limit of the doubled frequency field amplitude

$$x_0 = y_\infty^{1/2} e^{y_\infty / A} \quad \text{or} \quad m_0 = (\gamma M_{max})^{1/2} e^{M_{max}/\gamma}, \quad (24)$$

$$\gamma = \hbar p_{a21} / r, \quad (25)$$

M_{max} is expressed in terms of y_∞ with the aid of (14b).

As is known, a total energy transfer from the first to the second harmonic is theoretically possible in the course of non-resonance frequency doubling. It readily follows from (24) that in the case under consideration the conversion factor $\alpha = M_{max}/m_0$ reaches a maximum at $m_0 \approx 1.16\,\gamma$; $\alpha_{max} \approx 0.43$. Here $M_{max} \approx 0.5\,\gamma$ and the ratio of energy converted into the second harmonic to that absorbed by the material is $W_{conv}/W_{abs}|_{z \to \infty} \approx 0.35$. If

$$M_{max} \ll \gamma, \quad (26)$$

an approximate formula can be used instead of (24):

$$M_{max} \approx m_0^2 / \gamma = (r / \hbar p_{a21}) m_0^2. \quad (27)$$

For $r \approx 10^{-51}$ cgs esu, $p_{a21} \approx 10^{-20}$ cgs esu, and $\gamma = 10^4$ cgs esu, the inequality (26) is satisfied when $m_0 \lesssim 0.3\,\gamma$ ($\sim 10^6$ V/cm). In this case the conversion factor is

$$\alpha_\infty \approx m_0 / \gamma. \quad (28)$$

Let us analyze the dependence of the doubled-frequency field amplitude upon the coordinate. From (18b) and (20) we have

$$z = (A\eta_0)^{-1} \int_{y_0}^{y} \{(x_0^2 e^{2(y_0 - t)/A} - t)^{-1} + x_0^2 e^{2(y_0-t)/A} - t\}\, dt. \quad (29)$$

In the general case the integral (29) cannot be expressed in terms of elementary functions. However, it can be computed approximately for $y_\infty < A$ ($M_{max} < \gamma$). We confine ourselves here to the case where the field of the first harmonic can be considered as given (condition (26) is satisfied). For $y_0 = 0$ we obtain

$$z = (A\eta_0)^{-1} [y x_0^2 (1 - y / x_0^2) - \ln (1 - y / x_0^2)]. \quad (30)$$

In dimensional variables we have

$$z = \frac{c^2 k}{\pi \omega^2 N n_0 p_{a21}} \left[\frac{\tau r m_0^2}{\hbar^2} \left(1 - \frac{\hbar p_{a21} M}{r m_0^2} \right) M - \frac{\hbar}{4 p_{a21} T} \ln \left(1 - \frac{\hbar p_{a21} M}{r m_0^2} \right) \right] \quad (30a)$$

The first term in the brackets of (30a) is due to saturation of the working level population difference. This term can be neglected if (see also (15d))

$$x_0^2 \ll 1 \quad \text{or} \quad m_0^2 \ll \hbar^2 / 2r(\tau T)^{1/2}. \quad (31)$$

As we know, in the case of non-resonance frequency doubling, in the approximation of a given first-harmonic field, the distance required for the second harmonic amplitude to reach a value y_1 is inversely proportional to x_0^2.[3] In the case of resonance doubling, as follows directly from (30), there is an optimum value of x_0^2

$$x_{0\,\text{opt}}^2 = y_1^2/2 + (y_1^2/4 + 1)^{1/2}, \quad (32)$$

such that $z(x_0 = x_{0\,\text{opt}}) = z_{min}$. The value of z_{min} can be found from (32) and (30).

We perform some numerical computations, assuming that an impurity dielectric is the working substance that is capable of satisfying the synchronization conditions. Let $m_0 \approx 2 \times 10^3$ cgs esu, $p_{a21} \approx 10^{-20}$ cgs esu, $r \approx 10^{-51}$ cgs esu, $\omega \approx 10^{15}$ sec^{-1}, $T \approx 10^{-11}$ sec, $Nn_0 \approx 10^{20}$, and lifetime $\tau \ll 10^{-7}$ sec. Then (31) is satisfied and (30a) yields $L \approx 10^{-2}$ cm for $M(z = L) \approx 2 \times 10^2$ cgs esu ($\alpha = 10\%$ and

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6)We will show below that for $y_0/x_0^2 \neq 1$ we have $y \to y_\infty$ and $x \to x_\infty$ as $z \to \infty$ (see (28) and (29)).

$M = 0.5\ M_{max}$). The corresponding conversion length for KDP is $L \approx 0.1$ cm (see 3.36 a in[3]).

Thus a material that is unsuitable as the working substance in a non-resonance parametric conversion (condition (9) holds) can be adequate for resonance conversion if the two-photon transition between levels 2 and 1, satisfying resonance condition (2), is allowed and the linewidth of transition 2 − 1 is sufficiently small.

Given a material of density $N \approx 10^{20}$, a conversion factor of $\alpha \approx 10\%$ can be obtained over lengths of the same order as in KDP crystals or even shorter. Let us note, however, that the above values for p_{21} and r are optimal in the sense that a considerable decrease of p_{21}[7)] from the above value (10^{-20} cgs esu) increases the conversion length; at the same time, as we noted above, α cannot exceed $\alpha_{max} = 0.43$. On the other hand, an increase of p_{a21} decreases the conversion factor (see (28)).

It is also of interest to determine the distance in which the second harmonic field amounts to a considerable fraction of M_{max}, such as $M = 0.9\ M_{max}$ ($y = 0.9 y_\infty$). Let $M_{max} = s\gamma$ ($y_\infty = sA$), where $s \ll 1$; then according to (27) and (30)

$$x_0 = (sA)^{1/2}, \quad L_{0,9} = (A\eta_0)^{-1}(2.3 + 0.09\ s^2A^2), \quad (33)$$

or

$$m_0 = s^{1/2}\gamma; \quad L_{0.9} \approx \frac{\hbar c^2 k}{\pi\omega^2 N n_0}\left(\frac{0.57}{p_{a21}^2 T} + 0.09 s^2 p_{a21}^2 \frac{\tau}{r^2}\right). \quad (33a)$$

It follows from (33a) that $L_{0.9}$ depends significantly upon the working-substance particle lifetime at level 2 if

$$\tau \gtrsim \tau_{crit} \approx 6r^2/s^2 p_{a21}^4 T. \quad (34)$$

For the above parameters of the material and for $m_0 \approx 2 \times 10^3$ cgs esu, we obtain $\alpha_\infty \approx 0.2$, $s = 4 \times 10^{-2}$, and $\tau_{crit} \approx 4 \times 10^{-8}$ sec. For $\tau \approx \tau_{crit}$ we get $L_{0.9} \approx 0.1$ cm.

4. Let us consider the problem of the parametric frequency division. According to (20) a field of frequency ω can be amplified only if $y_0 > A$ ($M_0 > \gamma$). This is difficult to achieve if $\gamma \gtrsim 10^4$ cgs esu (the limiting values of the fields when $y_0 > A$ can be found from (23)). The conversion factor can be increased considerably by placing the working substance in a resonator with a frequency ω. We consider here parametric frequency division in a traveling-wave resonator.

7)Note that $p_{\alpha 21}$ can be reduced by suitably orienting the field E_3 with respect to the direction of p_{21}.

The walls of the resonator are assumed transparent to fields of frequency 2ω (or ω_3). The phase difference is again assumed to be π, for during the establishment of the stationary distribution of the field $E(\omega)$ the growing resonator fluctuations will be just those characterized by the stable value $\theta = \pi$. Using (18a) and (20), we obtain

$$\frac{dx^2}{dz} = 2\frac{x^2\left[y_0 - \frac{1}{2}A\ln(x/x_0)^2 - x^2\right]\eta_0}{1 + \left[y_0 - \frac{1}{2}A\ln(x/x_0)^2 - x^2\right]^2}. \quad (35)$$

Assuming that the reflection coefficient R satisfies the condition $R^{-1} - 1 \ll 1$ for $z = l$ (l is the length of the resonator), and $R = 1$ for $z = 0$, we can expand $\ln(x/x_0)$ in a series. Using only the linear terms of the expansion and integrating (35), we obtain

$$z + \Gamma = \eta_0^{-1}\left[\ln\frac{(x^2v)^{(u+u^{-1})}}{u - x^2v^{1/u}} - x^2v\right], \quad (36)$$

where $v = 1 + A/2x_0^2$, $u = y_0 + A/2$, and Γ is the integration constant.

The boundary conditions can be written as follows:[8)]

$$\Gamma = \varphi(x_0), \qquad l + \Gamma = \varphi(x_l),$$

where $\varphi(x_l)$ is the right-hand side of (36) and x_l is the field in the resonator for $z = l$. Subtracting the first equality from the second, we find the resonator length necessary to obtain x_l

$$\eta_0 l = \left[y_0 + \left(y_0 + \frac{A}{2}\right)^{-1} - x_l^2\right](1 - R) + \left(y_0 + \frac{A}{2}\right)^{-1}\ln\frac{y_0 - Rx_l^2}{y_0 + (A(1-R)/2) - x_l^2}. \quad (37)$$

It follows from (37) that the limiting value of the field in the resonator is

$$x_{l\,max} = [y_0 - A(1-R)/2]^{1/2}. \quad (38)$$

Therefore, intensive conversion requires an input field of $y_0 \gg A(1 - R)/2$; it should be noted that the field in the absence of a resonator should be $y_0 \gg A$.

The field at the output from the resonator is $x_b = x_l(1 - R)$. Using (38) we find it to have a maximum for $R \approx 1 - 4y_0/3A$. Since we have assumed that R is close to unity, the last statement is valid for $4y_0/3A \ll 1$. Then

$$x_{b\,max} = 4y_0^{3/2}/3\sqrt{3}A. \quad (39)$$

8)Only amplitude relations are considered here. To determine the frequency spectrum of the resonator it is necessary to consider the field-phase equations in addition to (35).

Let us note that according to (39) $x(l)$ becomes a two-valued function when

$$y_0 > Y \approx \frac{A(1-R)}{4} + \left[\frac{A^2(1-R)^2}{16} + 1\right]^{1/2}. \quad (40)$$

Consequently, if (40) holds, two different field distributions of the first harmonic are possible for the same length l in the resonator.[9)] The qualitative dependence of x_l on l for this case is given in Fig. 1 (curve 1). The value of $x_{l\,min}$ is readily determined from (37)

$$2Rx_{l\,min}^2 = y_0(1+R) - AR(1-R) - \{[y_0(1+R) - AR(1-R)]^2 - 4R[y_0^2 - Ay_0(1-R) - (y_0 + AR)/(y_0 + A)]\}^{1/2}. \quad (41)$$

The values of l_{min} are found by substituting (41) into (37). It should be noted that when (40) holds, the value of l corresponding to $x_l = 0$ does not correspond to the threshold: stationary solutions are also possible for lower l (see Fig. 1). This means that the excitation conditions cannot be obtained in our case with the aid of a linear approximation in terms of x_l^2.

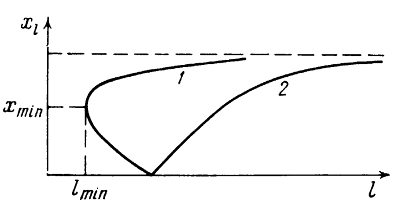

FIG. 1. Parametric frequency division. Field as a function of resonator length; curve 1—$y_0 > Y$; curve 2—$y_0 < Y$.

We conclude this section with numerical computations. Setting $\gamma = 10^4$ cgs esu, $M_0 \approx 10^3$ cgs esu, $R \approx 0.96$, $T \approx 10^{-11}$ sec, $\tau \approx 10^{-8}$ sec, and $N \approx 10^{20}$, we find from (38) and (14) $m_{l\,max} \approx 2.5 \times 10^3$ cgs esu, and $m_{b\,max} \approx 10^2$ cgs esu. Let us note that (40) does not hold here and a unique field distribution of the first harmonic is possible in the resonator (Fig. 1, curve 2). The stationary-solution criterion is found from (37): $l > 0.1$ cm. A resonator length $l \approx 0.25$ cm is necessary to obtain $m_l \approx 10^3$ cgs esu.

5. We consider the qualitative description of the phenomena in the case of incomplete synchronization $\delta k \neq 0$. The detuning Δ is assumed equal to zero as before. The equation for $\theta = 2\varphi_1(z) - \varphi_2(z) + (\delta k)z$ is obtained from (13) by adding δk to the right-hand side. Let us introduce a new variable $\varphi = \theta - \pi$. Then (13) assumes the form

$$d\varphi / dz = -f(z) \sin\varphi + \delta k, \quad (42)$$

where $f(z)$ is the coefficient of $\sin\theta$ in (13).

We now show that the function $\theta(z)$ has an upper bound when $|\delta k| < \min f(z)$. To consider an actual case, we assume that $\delta k > 0$ (the case of negative δk is considered in a similar manner, replacing φ by $-\varphi$).

Let us introduce two functions $\psi_1(z)$ and $\psi_2(z)$ such that

$$d\psi_1 / dz \leqslant -f(z) \sin\psi_1 + \delta k,$$
$$\delta\psi_2 / dz \geqslant -f(z) \sin\psi_2 + \delta k. \quad (43)$$

Then, if $\psi_1(0) \le \varphi(0) \le \psi_2(0)$ we have $\psi_1(z) \le \varphi(z) \le \psi_2(z)$.[11] We set $\psi_1 \equiv 0$ and ψ_2 equal to the solution of the second equation of (43) in which $f(z) = f_{min}$. Here

$$d\psi_2 / dz = -f_{min} \sin\psi_2 + \delta k \geqslant -f(z) \sin\psi_2 + \delta k,$$

$$\psi_2(z) = 2\,\mathrm{arctg}\frac{f_{min} + q - (f_{min} - q)Ke^{zq}}{\delta k(1 - Ke^{zq})} \quad (44)^*$$

where

$$q = \sqrt{f_{min}^2 - (\delta k)^2},$$

$$K = \frac{\delta k\,\mathrm{tg}\,{}^1/{}_2\psi_2(0) - f_{min} - q}{\delta k\,\mathrm{tg}\,{}^1/{}_2\psi_2(0) - f_{min} + q},$$

It follows from (44) that ψ_2 tends to the limit

$$\psi_2(\infty) = 2\,\mathrm{arctg}\frac{f_{min} - q}{\delta k}.$$

If

$$(\delta k)^2 \ll f_{min}^2, \quad (45)$$

$\psi_2(\infty) \approx \delta k/f_{min}$. Thus, if (45) holds, φ tends to the region

$$0 \leqslant \varphi \leqslant \delta k / f_{min} \quad (46)$$

and $\theta = \pi + \delta k/f_{min}$. Here, the rate of change of φ along the z axis, equal to $[f^2 - (\delta k)^2]^{1/2}$ (see (44)), is the minimum rate. In fact, as noted above, this rate, which is proportional to $m(z)/M_0$ (or to m_1m_2/m_3), is much larger than the minimum in the case of frequency doubling (or frequency addition) when $M_0 \approx 0$ ($m_3 \approx 0$ for $z = 0$) (see (42) and (13)).

To determine f_{min} we can use the results of the analysis for $\delta k = 0$. Then, according to (13) and (24), if (26) and (31) hold,

$$\min f \approx \min B_1'' \frac{m_0^2}{M(z)} = B_1'' \frac{m_0^2}{M(\infty)} = B_1''\gamma.$$

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9)We are not concerned here with the time stability of these distributions.

*arctg ≡ $\tan^{-1}$, tg ≡ tan.

Using the above parameters of the medium we find that $f_{min} \approx 10-100$. Consequently, $\cos\theta \approx -[1-(\delta k/f_{min})^2]$ for $\delta k < (3-30)$ cm^{-1} [10)] in the range defined by (46).

In the case of insignificant saturation Eqs. (18) have the form

$$\frac{\partial y}{dz} = A\left[x^2\left(1-\frac{\varphi^2}{2}\right)-y\right]\eta_0,$$

$$\frac{dx^2}{dz} = 2x^2\left[y\left(1-\frac{\varphi^2}{2}\right)-x^2\right]\eta_0. \qquad (47)$$

If $A \gg 1$, y rapidly reaches the range of slow motions

$$y = x^2(1-\varphi^2/2) \qquad (48)$$

and remains there. Therefore

$$dx^2/dz \approx -2x^4\varphi^2, \quad x^2 \approx \frac{(x^0)^2}{1+2(x^0)^2\varphi^2 z}. \qquad (49)$$

Here, x^0 is the value of x for which the representative point falls within the range $(x^2 - y) \approx \varphi^2/2$ (see Fig. 2). According to (48) and (49), x and y tend towards a stable equilibrium condition $y = x = 0$, rather than towards the singular curve $x^2 = y$

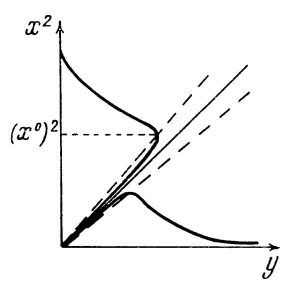

FIG. 2. Phase plane in the case of incomplete synchronization (explanation in text).

10)Let us note that in the case of the parametric frequency division the requirements imposed on δk are more rigorous, since f_{min} depends on the population difference n which, owing to saturation, can be small near the boundary in the presence of a strong resonance field m_3. Furthermore, $d\theta|dz|_{z=0} \sim m_3 n|_{z=0}$ is in this case smaller, generally speaking, than in the case of frequency doubling.

as in the case of $\delta k = 0$. The rate of this motion, according to (49), does not exceed $2x_0^4\varphi^2$.

In conclusion let us note the following. If we specify the ratio of the first and second harmonic fields at the boundary as $\gamma M_0/m_0^2 \approx 1$ ($M_0 \ll m_0$), then the resonance field of the frequency 2ω at $\delta k = 0$ propagates without absorption (neglecting linear attenuation), as follows from (22) and (23). For $0 < \delta k < f_{min}$, Eqs. (47) are valid; it follows from (49) that the second harmonic field attenuates by less than a factor of two at a distance of $z_1 \approx 1/2\varphi^2 x_0^2$. For $m_0 \approx 2 \times 10^3$ cgs esu, $\gamma \approx 10^4$ cgs esu, and $\varphi \approx 0.1$, we have $z_1 \approx 20$ cm.

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[1] R. V. Khokhlov, Radiotekhnika i élektronika **6**, 1116 (1961).

[2] J. A. Armstrong, N. Bloembergen, J. Ducing, and P. S. Pershan, Phys. Rev. **127**, 1918 (1962).

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

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

[5] D. A. Kleiman, Phys. Rev. 128, 1761 (1962).

[6] B. Lax, J. G. Mavroides, and D. F. Edwards, Phys. Rev. Lett. 8, 166 (1962).

[7] R. Laudon, Proc. Phys. Soc. 80, 952 (1962).

[8] A. M. Afanas'ev and E. A. Manykin, JETP 48, 483 (1965), Soviet Phys. JETP 21, 323 (1965).

[9] Yu. G. Khronopulo, Izv. Vuzov, Radiofizika 7, 674 (1964).

[10] V. S. Butylkin, G. L. Gurevich, M. I. Kheĭfets, and Yu. G. Khronopulo, Izv. Vuzov, Radiofizika 9, 538 (1966).

[11] E. Kamke, Differentialgleichungen, Losungsmetoden und Losungen, Pt. I, Chelsea, 1948.

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