

Four-wave interaction of ultrashort light pulses propagating successively in a medium

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The interaction of a material medium and two ultrashort light pulses that traverse it successively is investigated for the case in which the frequencies of each of the pulses satisfy the conditions for two-photon resonance with some transition of the medium. It is shown that in such resonance four-photon interactions, despite the fact that the pulses are not synchronous, there exists a region in which the generated wave becomes phase locked, so that the process turns out also to be spatially coherent. Relations are derived, both for the case of large wave mismatch ($|\delta k| \rightarrow \infty$) and for the case $\delta k = 0$, that represent the area theorem for a process of the type under consideration. It is found that at large distances a subpulse of the trigger field should develop within the second pulse, and that a subpulse that satisfies the proportionality condition and whose area tends either to zero or to 2π should also develop. It is also concluded that subpulses should develop in the case of two-photon interaction of pulses that have different frequencies and do not satisfy the proportionality condition. The conditions for maximizing the conversion coefficient (for the case $\delta k = 0$) are obtained with the aid of the area theorem and approximate solutions for the first stage. It is shown that the limitation on the conversion coefficient at high pump energies is not due to an increase in the parametric transparency of the medium, as in the quasistationary case, but to the formation of a 2π subpulse. It is shown that waves that are not present at the input cannot arise in systems having large wave mismatch (i.e., in the limit $|\delta k| \rightarrow \infty$); however, the second pulse can be amplified provided both its frequency components are present at the input. The pump- and trigger-field intensities necessary for observing the effects in sodium vapor are estimated.

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The current interest in resonant four-wave parametric interactions (RFPI), which is evidenced by a considerable number of experimental papers (see, e.g., Refs. 1-9) is due to the possibility of using such interactions for frequency conversion of radiation. By now, the quasistationary theory of RFPI has been rather thoroughly worked out. However, this theory turns out to be useless for the case of pulses whose durations are comparable with or shorter than the characteristic relaxation times of the atoms (or molecules) of the medium. This fact and the widespread use of mode-locked lasers that produce pulses considerably shorter (of the order of tens of picoseconds) than the relaxation times of the atoms (or molecules) make it obviously desirable to develop a theory of RFPI for ultrashort light pulses (USLP).

The RFPI of USLP that has been studied in greatest detail is frequency degenerate and constitutes third-harmonic generation via a two-photon resonance at the pump frequency.¹⁰⁻¹³ However, RFPI that are not frequency degenerate are of no less interest; they can be used to transfer the tuning of radiation from one range to another. Below we shall examine one of these processes—one in which the frequencies ω_j of the fields E_j ($j = 1, 2, 3, 4$) satisfy the conditions

$$\omega_1 + \omega_2 = \omega_{m1}, \quad \omega_3 + \omega_4 = \omega_{m1},$$

where ω_{m1} is the transition frequency between levels 1 and m of the medium. For definiteness we assume that the input fields—those whose pulses must be fed into the medium—are the fields of frequency ω_1 and ω_2 (pump fields) and ω_3 (trigger field); the field E_4 need not be fed into the medium. The interaction of simultaneously propagating pulses of all four frequencies has been

investigated under the assumption that the fields E_3 and E_4 are weak.^{14, 15} Calculations¹⁶ have shown, however, that the polarization of the atoms (or molecules) of the medium near the frequency $\omega_4 = \omega_{m1} \pm \omega_3$ is greater when the pulse of the fields E_3 and E_4 traverse the medium after the action of the pump pulse. Experimental results¹⁷ indicate that the frequency conversion efficiency in the RFPI of USLP is also greater in this case.¹⁾ We shall accordingly investigate frequency conversion in the case of successive passage of the pump and trigger pulses through the medium. The most important aspects of the problem are the magnitude of the maximum conversion coefficient, the relations between the frequencies and energies of the pump and trigger fields necessary to achieve the maximum conversion coefficient, and the effect of wave mismatch on the interaction process. In addition, we shall obtain an expression that amounts to a formulation of the area theorem for RFPI of the type under consideration and also clarify features of the asymptotic behavior of USLP at large distances under two-photon absorption.

1. INITIAL RELATIONS

Let the fields propagating in the medium in the z direction have the form

$$E_j = \mathcal{E}_j e^{i(\omega_j t - k_j z)} + \text{c.c.}, \quad \mathcal{E}_j = A_j e^{-i\varphi_j},$$

where A_j and φ_j are slowly varying functions of z and t compared with $e^{i\omega_j t}$ and $e^{ik_j z}$, and let the lengths of the pump and trigger pulses, as well as the time between passage of the trailing edge of the first and the leading edge of the second through a given point, be much shorter than the longitudinal and transverse re-

laxation times. We assume that the pump pulse is "proportional," i. e. that the real amplitudes $A_{10}(t)$ and $A_{20}(t)$ of its components at the entrance to the medium satisfy the proportionality condition

$$n_1 \omega_1^{-1} A_{10} = n_2 \omega_2^{-1} A_{20} \quad (1)$$

(if the condition (1) is not satisfied it will not be possible to store the entire energy of the first pulse as coherent excitation: some of the energy will escape from the medium without being absorbed). Then if there is no phase modulation at the entrance to the medium (if $\varphi_1 + \varphi_2 = \text{const}$ at $z=0$), the coherent excitation of the medium after passage of the first pulse will be described by the following expressions for the population difference η^1 and the off-diagonal density-matrix element σ_{1m}^1 :

$$\eta^1 = \cos \theta(z), \quad \sigma_{1m}^1 = -\frac{1}{2} i \sin \theta(z) \exp[-i(k_1 + k_2)z - i(\varphi_1 + \varphi_2)], \quad (2)$$

in which the area of the pulse,

$$\theta(z) = 2\kappa_{12} \hbar^{-1} \int_{t_0}^{t_1} A_1 A_2 dt$$

depends on the coordinate z according to the law¹⁹

$$\text{ctg}(\theta(z)/2) = \text{ctg}(\theta_0/2) + az. \quad (3)$$

Here

$$a = 2\pi N \kappa_{12} c^{-1} (\omega_1 \omega_2 / n_1 n_2)^{1/2}, \quad \theta_0 = \theta(z=0),$$

N is the number of molecules per unit volume, κ_{12} is the polarizability of the two-photon transition under the action of the fields E_1 and E_2 , n_j is the refractive index at the frequency ω_j , and (t_0, t_1) is the time interval during which the first pulse passes the point z .

Expressions (2) are the initial conditions for the second pulse, whose behavior is described by the equations

$$\frac{\partial \mathcal{E}_{3i}}{\partial z} + \frac{1}{v} \frac{\partial \mathcal{E}_{3i}}{\partial t} = -\frac{2\pi i N \kappa_{34} \omega_{3i}}{n_{3i} c} \mathcal{E}_{1s} \sigma_{1m} e^{i(k_1 + k_2)z}, \quad (4)$$

$$\frac{\partial \sigma_{1m}}{\partial t} = -i \frac{\kappa_{34}}{\hbar} \mathcal{E}_{3i} \mathcal{E}_{1s} e^{-i(k_1 + k_2)z}, \quad (5)$$

and

$$\frac{\partial \eta}{\partial t} = \frac{\kappa_{34}}{\hbar} \text{Im} \{ \mathcal{E}_{3i} \mathcal{E}_{1s} \sigma_{1m} e^{i(k_1 + k_2)z} \}. \quad (6)$$

Here κ_{34} is obtained from κ_{12} by the substitution $\omega_{1,2} \rightarrow \omega_{3,4}$, we assume all the group velocities to be equal ($v_j = v$) and we neglect the difference between the polarizabilities κ_{11} and κ_{mm} .

2. FIRST FREQUENCY-CONVERSION STAGE

It is typical of most frequency-conversion experiments that the field E_4 is not present at the entrance to the medium ($A_{40} = 0$). Taking account of the fact that E_4 is small during the first stage, we find from Eq. (5) that $\sigma_{1m}(t, z) = \sigma_{1m}(t_1, z) = \sigma_{1m}^1$. Substituting this value into Eq. (4) on the right and writing the equation for the phase difference, $\Delta = \varphi_1 + \varphi_2 - \varphi_3 - \varphi_4 + z \delta k$, we have

$$\partial \Delta / \partial \zeta = \delta k + W(\xi, \zeta) \sin \Delta, \quad (7)$$

where $\delta k = k_1 + k_2 - k_3 - k_4$ is the wave mismatch, $\zeta = z$,

and $\xi = z - vt$. The quantity

$$W = \frac{\pi N \kappa_{34}}{c} \sin \theta(\zeta) \left[\frac{\omega_3 A_4(\xi, \zeta)}{n_3 A_3(\xi, \zeta)} + \frac{\omega_4 A_3(\xi, \zeta)}{n_4 A_4(\xi, \zeta)} \right] \quad (8)$$

is positive when $0 < \theta(\xi) < \pi$ and negative when $\pi < \theta(\varphi) < 2\pi$.

Using Eq. (8) and the solution

$$\text{tg} \frac{\Delta(\xi, \zeta)}{2} = \text{tg} \frac{\Delta(\xi, 0)}{2} \exp \int_0^\zeta W(\xi, \zeta) d\zeta$$

of Eq. (7) for $\delta k = 0$ and noting that $A_3 \gg A_4$ near the entrance to the medium, we easily see that the phase difference Δ quickly reaches a region close to $2k\pi$ or to $(2k+1)\pi$ in which its subsequent variation is slow and is described by the expression

$$\Delta(\xi, \zeta) = \begin{cases} \delta k/W + (2k+1)\pi & \text{if } 0 < \theta < \pi \\ -\delta k/W + 2k\pi & \text{if } \pi < \theta < 2\pi \end{cases} \quad (9)$$

Thus, phase locking of the interacting waves takes place in resonant parametric interactions of USLP that do not overlap in time and space, just as it does²⁰ in quasistationary RFPI.

Using Eqs. (4)–(6) and relation (9) (and neglecting corrections to (9) of the order of $|\delta k|W^{-1} \ll 1$), it is not difficult to obtain the following equations for the real amplitudes A_3 and A_4 :

$$\frac{\partial A_{3i}}{\partial z} + \frac{1}{v} \frac{\partial A_{3i}}{\partial t} = -\frac{\pi N \kappa_{34} \omega_{3i}}{n_{3i} c} A_{4s} \sin[\theta_i \pm \theta(z)]. \quad (10)$$

Here and below the upper sign is for the case $\pi < \theta_0 < 2\pi$, and the lower sign, for the case $0 < \theta_0 < \pi$; further,

$$\theta_i = 2\kappa_{34} \hbar^{-1} \int_{t_0}^t A_3 A_4 dt,$$

where t_2 is the time at which the second pulse appears at the point z . As long as A_4 is so small that

$$\theta_i \ll |\text{tg} \theta|, \quad \theta_i \ll 1, \quad (11)$$

Eqs. (10) have the approximate solutions

$$A_{3i} = \frac{A_{30}(t-z/v)}{2} \left(\frac{\omega_{3i} n_3}{\omega_3 n_{3i}} \right)^{1/2} \left| \left(\frac{\sin \frac{\theta(z)}{2}}{\sin \frac{\theta_0}{2}} \right)^{\alpha/\alpha} - \left(\frac{\sin \frac{\theta(z)}{2}}{\sin \frac{\theta_0}{2}} \right)^{-\alpha/\alpha} \right|, \quad (12)$$

in which $A_{30}(t) = A_3(t, z=0)$ is the amplitude of the trigger field as it enters the medium, and $\alpha = 2\pi N \kappa_{34} c^{-1} (\omega_3 \omega_4 / n_3 n_4)^{1/2}$.

It is evident from (12) that when $0 < \theta_0 < \pi$, the fields A_3 and A_4 increase with increasing z while the pulse tends more nearly to satisfy the proportionality condition. The area ϑ_i of the pulse increases without limit, so that condition (11) must ultimately be violated. Then the first stage of the conversion ends (for the case $0 < \theta_0 < \pi$) if it has not ended earlier because of violation of the phase locking condition

$$|\delta k|W^{-1} \ll \pi.$$

If $\pi < \theta_0 < 2\pi$, the fields A_3 and A_4 also increase at first, but they reach a maximum when $\theta(z)$ comes

close to π . Although the first of the conditions (11) is violated near this point, each of the fields A_3 and A_4 is virtually the same after this point as before it provided $\vartheta_1 \ll 1$, so that one can continue to use formula (12) beyond this point. The subsequent behavior of A_3 and A_4 is the reflection in the point $\theta(z) = \pi$ of their behavior for $\pi < \theta(z) < \theta_0$. Hence we have $A_3 = A_4$ and $A_4 = 0$ at the point $\theta(z) = 2\pi - \theta_0 = \bar{\theta}$. The phase changes discontinuously by π at this point, and the subsequent behavior of the fields is the same as for pumping with an input area $\bar{\theta}$ between 0 and π .

Depending on the relation between θ_0 , α , and a , it may turn out that ϑ_1 becomes of the order of or greater than unity before the point where $\theta(z) = \pi$ is approached. In that case formula (12) is no longer applicable.

We note that the above description of the behavior of the fields for $[\pi < \theta_0 < 2\pi]$ is valid not only for systems in which the wave synchronism is exact ($\delta k = 0$), but also for systems in which $|\delta k|$ is much smaller than the reciprocal distance between the maximum [at $\theta(z) = \pi$] and the zero [at $\theta(z) = \bar{\theta}$] of the field A_4 . In the opposite case the first stage ends not because of violation of conditions (11), but because of phase breaking. In subsequent stages the character of the interaction also depends substantially on the magnitude of the wave mismatch.

3. FORMATION OF PULSES OF THE FIELDS A_3 AND A_4 IN SYSTEMS WITH WAVE SYNCHRONISM ($\delta k = 0$)

In the most typical experimental situations, A_{30} is small. In this case the subsequent behavior of the fields A_3 and A_4 can be calculated under the assumption that the second pulse nearly satisfies the proportionality condition. We introduce the following notation:

$$\psi = \text{ctg} \frac{\theta_1 \pm \theta}{2}, \quad x = az + \text{ctg} \frac{\theta_0}{2}, \quad y = -vt + \frac{x}{a}.$$

Then it is not difficult to derive the equation

$$2a \frac{\partial \psi}{\partial x} + \alpha(\psi^2 - 1) = (\psi^2 + 1) \frac{\pm 2a + \alpha(x^2 - 1)}{x^2 + 1} \quad (13)$$

from (10); this equation is related to the Riccati equation and has the particular solution $\psi = x$. The general solution of (13), expressed in terms of the variables x and y , has the form

$$\psi = \text{ctg} \frac{\theta_1 \pm \theta(x)}{2} = \pm x + \frac{(x^2 + 1)^{\pm \alpha/a}}{B(y) \mp D(x)}, \quad (14)$$

where

$$B(y) = \left(\sin^2 \frac{\theta_1}{2} \right)^{\pm \alpha/a - 1} \left[\text{ctg} \frac{S_1 \pm \theta_1}{2} \mp \text{ctg} \frac{\theta_1}{2} \right],$$

$$S_1(y) = -\frac{2\kappa_{34}}{\hbar} \int_y^{y_0} A_3(x_1, y) A_4(x_1, y) dy, \quad (15)$$

$$D(x) = \left(1 \mp \frac{\alpha}{a} \right) \int_{x_1}^x \frac{dx}{(x^2 + 1)^{\pm \alpha/a}},$$

$x_1 = az_1 + \cot(\theta_0/2) = \cot(\theta_1/2)$, and $z = z_1$ is the coordinate of the point beyond which the pulse may be assumed to be "proportional." The upper sign in Eqs. (14) and (15) is for the case $\pi < \theta_1 < 2\pi$, and the lower sign, for the case $0 < \theta_1 < \pi$.

For the first case, the solution (14) for the total area

$$\vartheta = \frac{2\kappa_{34}}{\hbar} \int_{t_1}^{\infty} A_3(x, t) A_4(x, t) dt$$

of the second pulse takes the form

$$\text{ctg} \frac{\theta + \theta(x)}{2} = x + (x^2 + 1)^{1 - \alpha/a} \left[\left(\sin^2 \frac{\theta_1}{2} \right)^{\alpha/a - 1} \times \left(\text{ctg} \frac{\theta_1 + \theta_1}{2} - \text{ctg} \frac{\theta_1}{2} \right)^{-1} - 1 - \frac{\alpha}{a} \int_{x_1}^x \frac{dx}{(x^2 + 1)^{\alpha/a}} \right]^{-1}, \quad (16)$$

where ϑ_1 is the limit of S_1 as $t \rightarrow \infty$. When $\alpha = a$ (this is the case, for example, when $\omega_3 = \omega_1$ and $\omega_4 = \omega_2$) formula (16) reduces to the area theorem for two-photon absorption of a pulse consisting of two subpulses. In the general case, (16) is the area theorem for the RFPI that we are considering. Results of an analysis of (16) are presented in Table I, from which it follows, in particular, that if $\vartheta_1 \ll 1$ (A_{30} being small, while $A_{40} = 0$) there are two limiting values $\vartheta_{1\text{lim}}$ of ϑ , namely 0 and 2π ($k = 0$).

Using (14) and (15), we can obtain the space-time distribution of the fields for $\pi < \theta_1 < 2\pi$:

$$A_{3,4}(x, y) = \frac{A_{3,4}(x_1, y) |\sin(\theta_1/2)|^{\alpha/a} (x^2 + 1)^{(1 - \alpha/a)/2}}{[C(y)]^{1/2} \{ [B(y) - D(x)]^2 + [B(y) - D(x)]_x + (x^2 + 1)^{1 - \alpha/a} \}^{1/2}}, \quad (17)$$

where

$$C(y) = \left[\text{ctg} \frac{S_1(y) + \theta_1}{2} - \text{ctg} \frac{\theta_1}{2} \right]^2 \sin^2 \frac{S_1(y) + \theta_1}{2}.$$

It is obvious from this that when $\alpha > a$ the fields A_3 and A_4 at large distance, near the points at which $B(y) = D(x)$, increase without limit as x increases, while the fields in the rest of the pulse decrease. If $\alpha < a$, on the other hand, the fields decrease throughout the entire pulse.

By analyzing Eq. (14) for the case $0 < \theta_1 < \pi$ in a similar manner one can show that the limiting value $\vartheta_{1\text{lim}}$ is zero (since $\vartheta_1 \ll 1$) and no subpulse whose amplitude would increase without limit is formed. This is closely evident from the computer-calculated curves in Figs. 1 and 2.

Let us discuss the more general case in which the initial stage of the conversion ends before the pulse becomes approximately "proportional." Using Eq. (10) written in terms of the coordinates ζ and ξ , we obtain

$$\frac{\partial}{\partial \zeta} \int_{\xi}^{\xi_0} A_{3,4}^2 d\xi = \frac{\pi \hbar N \omega_3 v}{n_{3,4} c} [\cos(\pm \theta_1 + \theta(\zeta)) - \cos \theta(\zeta)],$$

$$\frac{A_3^2}{\omega_3} - \frac{A_4^2}{\omega_4} = \frac{A_{30}^2(\xi)}{\omega_3} - \frac{A_{40}^2(\xi)}{\omega_4}, \quad (18)$$

TABLE I. Behavior of ϑ at large distances (as $z, x \rightarrow \infty$).

		$2k\pi < \theta_1 < 2(k+1)\pi - \theta_1$	$2(k+1)\pi - \theta_1 < \theta_1 < 2(k+1)\pi$
$\alpha > a$	$ B > D $	ϑ decreases to $2k\pi$	ϑ increases to $2(k+1)\pi$
	$ B < D $	ϑ increases to $2(k+1)\pi$	ϑ increases to $2(k+1)\pi$
$\alpha < a$	$ B > D $	ϑ decreases to $2k\pi$	ϑ increases to $2(k+1)\pi$
	$ B < D $	ϑ decreases to $2k\pi$	ϑ increases to $2k\pi$

Note. k represents any integer.

where

$$\vartheta_{\xi} = \frac{2\kappa_{34}}{\hbar} \int_{\xi}^{\xi_0} A_3 A_4 d\xi.$$

If we let ξ tend to infinity, we can see without difficulty that for arbitrary pump and trigger fields and with no input field of frequency ω_4 ($A_{40}=0$) there can be only two limiting values, namely 0 and 2π , for the total area ϑ of the pulse. Writing $\theta(\xi)=0$ in (18), we can show rigorously that a "proportional" subpulse of area 2π (or 0) develops in the second pulse at large enough distances against the background of the trigger pulse.

We note that in the case of two-photon absorption of pulses that do not satisfy the proportionality condition one can show in just the same way that "proportional" subpulses of area 2π arise, as well as a subpulse of that one of the fields A_j ($j=3,4$) for which the quantity $n_j \omega_j^{-1} A_{j0}^2$ is the larger; moreover

$$\lim_{\xi \rightarrow \infty} A_j^2(\xi, \xi) = A_{j0}^2(\xi) - \frac{\omega_3 n_4}{\omega_4 n_3} A_{40}^2(\xi).$$

Using these results together with the law of energy conservation and the first integral written down in (18), we can conclude that the limiting area ϑ_{lim} of a pulse that does not satisfy the proportionality condition as it enters the medium does not exceed the value

$$\frac{2\kappa_{34}}{\hbar} \left(\frac{\omega_4 n_3}{\omega_3 n_4} \right)^{1/2} \int_{\xi}^{\infty} A_{40}^2 dt,$$

which is the area of the "proportional" part of the input pulse and is certainly smaller than the total area

$$\frac{2\kappa_{34}}{\hbar} \int_{\xi}^{\infty} A_{30} A_{40} dt$$

of the pulse at the entrance to the medium in the case of two-photon absorption. It follows at once from what has been said that, for example, a 2π pulse that does not satisfy the proportionality condition at the entrance to the medium will damp out to zero at large distances.

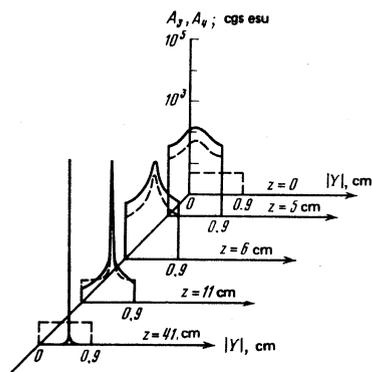


FIG. 1. Space and time evolution of the trigger-field (A_3) and generated-field (A_4) pulses as calculated for the following parameter values: $\alpha/a=2$, $\theta_0=1.9\pi$, $A_{30}=5$ cgs esu, $\lambda_3=2364$ nm, $\lambda_4=330$ nm, and $N=10^{16}$ cm $^{-3}$. $Y=x/a-vt$, and z is the distance traversed by the pulse. The trigger-field pulses are shown by the dashed curves, and the generated-field pulses, by the full curves.

4. CONVERSION COEFFICIENT AND THRESHOLD CONDITION FOR SYSTEMS HAVING WAVE SYNCHRONISM ($\delta k = 0$)

Let us find the limiting values (as $z \rightarrow \infty$) of the coefficients $\beta_{3,4}$ for conversion of pump energy to energy of the fields A_3 and A_4 . Making use of the fact that a "proportional" subpulse develops against the background of the trigger pulse, we can easily express the limiting energies $U_{3,4}^{lim}$ of the fields $A_{3,4}$ in terms of the pulse area ϑ_{lim} :

$$U_3^{lim} = \int_{\xi}^{\infty} A_{30}^2 dt = U_{30} + \frac{\hbar}{2\kappa_{34}} \left(\frac{\omega_3 n_4}{\omega_4 n_3} \right)^{1/2} \vartheta_{lim} \quad (19)$$

$$U_4^{lim} = \int_{\xi}^{\infty} A_{40}^2 dt = \frac{\hbar}{2\kappa_{34}} \left(\frac{\omega_4 n_3}{\omega_3 n_4} \right)^{1/2} \vartheta_{lim}.$$

From this it follows that

$$\beta_3^{lim} = \frac{U_3^{lim} - U_{30}}{U_{10} + U_{20}} = \frac{\omega_3}{n_3} \left(\frac{\omega_1}{n_1} + \frac{\omega_2}{n_2} \right) \frac{a}{\alpha} \frac{\vartheta_{lim}}{\theta_0} \approx \frac{\omega_3}{\omega_{21}} \frac{a}{\alpha} \frac{\vartheta_{lim}}{\theta_0}, \quad (20)$$

$$\beta_4^{lim} = \frac{U_4^{lim}}{U_{10} + U_{20}} \approx \frac{\omega_4}{\omega_{21}} \frac{a}{\alpha} \frac{\vartheta_{lim}}{\theta_0}$$

(U_{10} and U_{20} are the input energies of the pump fields A_1 and A_2). The fact that the refractive indices do not differ by more than 1% is taken into account in (20).

The complete limiting conversion coefficient is

$$\beta_{lim} = \beta_3^{lim} + \beta_4^{lim} = \frac{a}{\alpha} \frac{\vartheta_{lim}}{\theta_0}. \quad (21)$$

It is evident from Eqs. (2) and (4)–(6) that the energies U_3 and U_4 of the fields A_3 and A_4 are periodic functions of θ_0 with the period 2π (this is shown in Ref. 16 under the assumption that the fields A_3 and A_4 are weak). Hence ϑ_{lim} is also a periodic function of θ_0 , and according to (20) and (21) the use of a pump pulse with area $\theta_0 > 2\pi$ will reduce the conversion coefficient.

It was shown in the preceding section that ϑ_{lim} can assume either of the two values 0 and 2π . In the first case $\beta_{lim} = 0$, and in the second case β_{lim} differs from

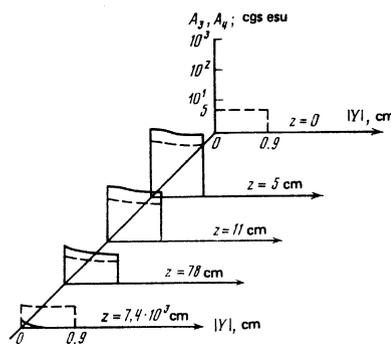


FIG. 2. Space and time evolution of the trigger-field (A_3) and generated-field (A_4) pulses as calculated for the following parameter values: $\alpha/a=1/2$, $\theta_0=1.9\pi$, $A_{30}=5$ cgs esu, $\lambda_3=2364$ nm, $\lambda_4=330$ nm, and $N=10^{16}$ cm $^{-3}$. $Y=x/a-vt$, and z is the distance traversed by the pulse. The trigger-field pulses are shown by the dashed curves, and the generated-field pulses, by the full curves.

zero and is given by

$$\beta_{\text{lim}} = 2\pi a / \theta_0 \alpha. \quad (22)$$

It is of interest to find the conditions that the trigger field must satisfy in order for the value $\vartheta_{1\text{lim}} = 2\pi$ to be reached. To do this we must determine the point x_1 at which the solution for ϑ in the first stage of the conversion (12) fits the value of ϑ from (16) smoothly, i. e., where the functions $\vartheta(x)$ themselves and their first derivations with respect to x coincide. Expressing ϑ_1 , B , and D in terms of the value found for x_1 , we obtain the threshold condition

$$\int_{x_1}^{\infty} A_{30}^2 dt > \frac{4\hbar a}{\kappa_{31}(\alpha - a)} \left(\frac{\omega_3 n_4}{\omega_3 n_3} \right)^{1/2} \left(\sin^2 \frac{\theta_0}{2} \right)^{\alpha/a} \left[\int_{-1}^{\infty} \frac{dx}{(x^2 + 1)^{\alpha/a}} \right]^{-1} = U_{30}^{\text{thr}}. \quad (23)$$

The condition that the pulse satisfies the proportionality condition at the point x_1 turns out to be even simpler:

$$[2 \sin^2(\theta_0/2)]^{2\alpha/a} \ll 1. \quad (24)$$

This pulse can be regarded as small when

$$\frac{\kappa_{31}}{\hbar} \left(\frac{\omega_3 n_3}{\omega_3 n_4} \right)^{1/2} \left(\sin^2 \frac{\theta_0}{2} \right)^{-\alpha/a} \int_{x_1}^{\infty} A_{30}^2 dt \ll 2^{\alpha/a - 1}. \quad (25)$$

If we do not determine x_1 from the condition that ϑ and its first derivative match at this point, but rather from the requirement that the pulse satisfy the proportionality condition to a certain degree of approximation, then the threshold condition (23) will remain virtually unchanged, while the region within which the analysis is applicable, which is determined by (25), will become considerably broader. If condition (23) is satisfied, then, as is evident from (22), the conversion coefficient for a given pump pulse ($\theta_0 = \text{const}$) will not depend on the strength of the trigger field. When θ_0 is not much smaller than 2π but $\vartheta_{1\text{lim}} = 2\pi$ (the threshold being exceeded) then $\beta \approx a/\alpha < 1$ for systems in which $\alpha > a$. The largest value of β will be reached when α is only slightly larger than a , but in this case a large trigger field must be fed in [see (23)]. It is possible that by increasing the strength of the trigger field one might be able to assure passage to a 2π pulse at a lower pump-pulse area θ_0 and thereby increase the limiting energy-conversion coefficient in systems in which α is considerably larger than a .

If both of the fields A_{30} and A_{40} are fed into the medium the conversion coefficients will not be obtained from Eqs. (20) and (21) directly, but by the equations obtained by substituting $\vartheta_{1\text{lim}} - \vartheta_0$ for $\vartheta_{1\text{lim}}$ in them, where

$$\vartheta_0 = \frac{2\kappa_{31}}{\hbar} \left(\frac{\omega_3 n_4}{\omega_3 n_3} \right)^{1/2} \int_{x_1}^{\infty} A_{40}^2 dt \quad (26)$$

(for definiteness we assume that $(n_3/\omega_3)^{1/2} A_{30} > (n_4/\omega_4)^{1/2} A_{40}$). The threshold conditions for the amplification of pulses of the fields A_{30} and A_{40} that satisfy the proportionality conditions at the entrance to the medium will already be determined not by (23), but by the relations between ϑ_1 , B , and D in accordance with Table I. A pulse of the arbitrary fields A_{30} and A_{40} will be amplified if the threshold conditions according to Table I are satisfied for the "proportional" part of the input pulse of area ϑ_0 [see Eq. (26)]. Then the limiting conversion coefficient will not depend on whether or not

the second pulse is "proportional" on entering the medium.

When $a > \alpha$, the result $\vartheta_{1\text{lim}} = 2\pi$ would violate energy conservation [it would imply that $\beta_{1\text{lim}} > 1$, see Eq. (21)] for arbitrary values of θ_0 and A_{30} (with $A_{40} = 0$). Hence the only possible limiting value for the pulse area in systems having $a > \alpha$ is $\vartheta_{1\text{lim}} = 0$, i. e., in this case it is impossible to obtain a 2π pulse in the limit. However, this does not mean that no frequency conversion is possible at all when $a > \alpha$. Let us consider the case $a = 2\alpha$ as an example. We determine the maximum pulse area ϑ_{max} from (16), while the position x_m of the maximum is a root of the equation

$$\begin{aligned} \sin \frac{\theta_0}{2} \left[\text{ctg} \frac{\theta_1 + \theta_1}{2} - \text{ctg} \frac{\theta_1}{2} \right] + \frac{1}{2} \ln \left| \text{ctg} \frac{\theta_0}{4} \right| \\ = -\frac{1}{2} \left(\frac{(x_m^2 + 1)^{1/2}}{x_m} - \ln |x_m + (x_m^2 + 1)^{1/2}| \right). \end{aligned}$$

On substituting $x_1 = -1$ here and assuming that

$$\frac{\kappa_{31}}{\hbar} \left(\frac{\omega_3 n_3}{\omega_3 n_4} \right)^{1/2} U_{30} \left| \sin \frac{\theta_0}{2} \right|^{-1} = 0.47,$$

we find that $x_m \approx 0.1$, and that $\vartheta_{\text{max}} = 2\pi - 4 \arccot(x_m) \approx 0.13\pi$, which corresponds to a conversion coefficient of $\beta \approx 0.13$. It should be noted that it is not the entire energy of the pump pulse (of area θ_0) that is converted, but only the part of it that is stored in the medium on the interval $(0, z_m)$, where $z_m = a^{-1}[x_m - \cot(\theta_0/2)]$. The entire energy of the pump pulse can be completely stored only over an infinitely long distance, so complete conversion of the pump-pulse energy is possible only at large distances.

It is of interest to estimate the limiting values $\beta_{1\text{lim}}$ of the conversion coefficients and the threshold intensities I_{thr} of the trigger field necessary to realize them. Figure 3 shows the relation between $\beta_{1\text{lim}}$ and I_{thr} in sodium vapor (the variable parameter is α/a). The calculations were carried through for the $3S_{1/2}$ and $4D_{5/2}$ levels as the working levels and the following values of the fixed parameters: $N = 10^{16} \text{ cm}^{-3}$; $\kappa_{12} = 2 \times 10^{-22} \text{ cgs esu}$; pump frequencies $\nu_{1,2} = 17275 \text{ cm}^{-1}$; trigger- and generated-field frequencies varying near $\nu_3 = 4260 \text{ cm}^{-1}$ and $\nu_4 = 30290 \text{ cm}^{-1}$, respectively; and pump pulse area $\theta_0 = 1.9\pi$. As is evident from the figure, a trigger-pulse field intensity of $I_{\text{thr}} = 100 \text{ kW/cm}^2$ is required to achieve a conversion coefficient of $\beta_{1\text{lim}} = 75\%$, while $\beta_{1\text{lim}}$ reaches 90% at $I_{\text{thr}} = 500 \text{ kW/cm}^2$.

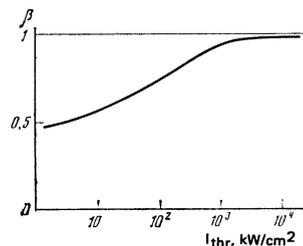


FIG. 3. Relation between the complete limiting conversion coefficient $\beta_{1\text{lim}}$ for sodium vapor and the trigger-field threshold intensity I_{thr} necessary to reach it (the variable parameter is the ratio α/a). The characteristics of sodium vapor used in the calculations were taken from Ref. 21.

5. SYSTEMS HAVING LARGE WAVE MISMATCH

Let us write Eqs. (4)–(6) in the form

$$\frac{\partial \mathcal{E}_{3,4}}{\partial z} + \frac{1}{v} \frac{\partial \mathcal{E}_{3,4}}{\partial t} = -\frac{\pi N \kappa_{34} \omega_{3,4}}{n_{3,c}} \mathcal{E}_{i,3} w, \quad (27)$$

$$\frac{\partial w}{\partial t} = \frac{2\kappa_{34}}{\hbar} \mathcal{E}_3 \mathcal{E}_4 \eta, \quad \frac{\partial \eta}{\partial t} = -\frac{2\kappa_{34}}{\hbar} \text{Re}(\mathcal{E}_3 \mathcal{E}_4^* w)$$

and the initial conditions, in the form

$$\eta_{in} = \cos \theta(z), \quad w_{in} = \sin \theta(z) \exp[-i(z\delta k + \varphi_1 + \varphi_2)]. \quad (28)$$

Here w is defined by the relation $\sigma_{1m} = -(1/2)iv \cdot \exp[-i(k_3 + k_4)z]$. We express the fields E_3 and E_4 and the density-matrix elements σ_{1m} and η in the form

$$\mathcal{E}_{3,4} = \bar{\mathcal{E}}_{3,4} + \tilde{\mathcal{E}}_{3,4}, \quad \eta = \bar{\eta} + \tilde{\eta}, \quad w = \bar{w} + \tilde{w},$$

with $|\tilde{\mathcal{E}}_{3,4}| \ll |\bar{\mathcal{E}}_{3,4}|$. Let us average Eqs. (27) and the initial conditions (28) over an interval that is short as compared with the characteristic length in which the fields $\bar{\mathcal{E}}_3$ and $\bar{\mathcal{E}}_4$ vary appreciably but much longer than $2\pi|\delta k|^{-1}$ (here $\bar{\mathcal{E}}_{3,4}$, $\bar{\eta}$, and \bar{w} are slowly varying functions of z (as compared with $e^{i\delta k}$), while

$$\tilde{\mathcal{E}}_{3,4} = \sum_{\nu} \mathcal{E}_{3,4}^{(\nu)} \exp(-i\nu z \delta k),$$

$$\tilde{\eta} = \sum_{\nu} \eta^{(\nu)} \exp(-i\nu z \delta k), \quad \tilde{w} = \sum_{\nu} w^{(\nu)} \exp(-i\nu z \delta k)$$

are rapidly varying functions). The averaged equations agree with (27) under the substitutions

$$\mathcal{E}_{3,4} \rightarrow \bar{\mathcal{E}}_{3,4}, \quad \eta \rightarrow \bar{\eta}, \quad w \rightarrow \bar{w}.$$

The initial values of the slowly varying components are

$$\bar{w}_{in} = 0, \quad \eta_{in} = \cos \theta(z).$$

On calculating the rapidly varying components of the fields it is not difficult to convince oneself that they are much smaller than the slowly varying components when the $|\delta k|$ are large enough.

Turning to the equations for the real amplitudes and phases ($\mathcal{E}_j = \bar{A}_j e^{-i\theta_j}$) we find that they are just the equations for two-photon interaction of the fields \mathcal{E}_3 and \mathcal{E}_4 with a medium in which the initial population difference is distributed according to the law $\eta_{in} = \cos \theta(z)$ and $\sigma_{1m} = 0$. The slowly varying real amplitudes satisfy the equations

$$\frac{\partial \bar{A}_{3,4}}{\partial z} + \frac{1}{v} \frac{\partial \bar{A}_{3,4}}{\partial t} = -\frac{\pi N \kappa_{34} \omega_{3,4}}{n_{3,c}} \bar{A}_{3,4} \cos \theta(z) \sin \left[\frac{2\kappa_{34}}{\hbar} \int_{z_0}^z \bar{A}_3 \bar{A}_4 dt \right]. \quad (29)$$

The boundary conditions for the amplitudes are $\bar{A}_{30} \approx A_{30}$ and $\bar{A}_{40} \approx A_{40}$ provided both the fields A_{30} and A_{40} are present at the input. If $A_{40} = 0$, Eqs. (27) must be used, beginning at the point z_c at which the discontinuous phase change takes place (see Sec. 2); in this case the boundary values \bar{A}_{jc} are determined with the aid of the condition

$$\mathcal{E}_j(z_c) = \bar{\mathcal{E}}_{jc}(z_c) + \tilde{\mathcal{E}}_j(z_c),$$

and are given approximately by the formulas

$$\bar{A}_{3c} \approx A_{30}, \quad \bar{A}_{4c} \approx A_{30} \frac{\pi N \kappa_{34} \omega_4 |\sin \theta_0|}{|\delta k| c n_4}. \quad (30)$$

We can clarify the characteristic features of the be-

havior of the fields of frequencies ω_3 and ω_4 by examining the propagation of a square pulse of length τ_2 . As long as

$$\theta_i = \frac{2\kappa_{34}}{\hbar} \int_{z_c}^z \bar{A}_3 \bar{A}_4 dt \ll 1,$$

we can replace the sine in (29) by its argument and then solve the equation; the solution, expressed in terms of the variables $M_j = \bar{A}_j^2 n_j \omega_j^{-1}$, has the form

$$\frac{M_4}{M_{4c}} = M \left\{ M_{3c} \exp \left[M \gamma \left(z - z_c + \frac{\theta - \theta_c}{a} \right) \right] - M_{4c} \right\}^{-1}, \quad (31)$$

$$\frac{M_3}{M_{3c}} = M \left\{ M_{3c} - M_{4c} \exp \left[-M \gamma \left(z - z_c + \frac{\theta - \theta_c}{a} \right) \right] \right\}^{-1},$$

where

$$M = M_{3c} + M_{4c}, \quad \gamma = 2\pi N \kappa_{34}^2 \omega_3 \omega_4 \tau_2 / n_3 n_4 c \hbar, \\ \theta_c = \theta(z_c).$$

If both A_{30} and A_{40} are different from zero, we must write

$$M_{jc} = \bar{A}_{j0}^2 n_j / \omega_j, \quad \theta_c = \theta_0, \quad z_c = 0.$$

It follows from (31) and (30) that as $|\delta k| \rightarrow \infty$, M_{4c} and \bar{A}_{4c} tend to zero and the field A_4 does not arise. It is also evident from (31) that if $M_{4c} \neq 0$ but $2\pi > \theta > 3\pi/2$, M_4 decreases with increasing distance and subsequently, when $3\pi/2 > \theta > \pi/2$, M_4 increases, after which the radiation of frequency ω_4 finally dies out. The greatest rise occurs when $\theta_c = 3\pi/2$ and $\theta = \pi/2$ (this corresponds to $z - z_c = 2/a$). As M_4 increases, the ratio M_4/M_3 approaches unity, and if

$$M \gamma \geq a \ln(M_{4c}/M_{3c}),$$

the pulse becomes "proportional" (within the limitations of the present approximation). After that (and sometimes earlier) the condition $\bar{\mathcal{E}}_i \ll 1$ may be violated.

The subsequent behavior of the pulse after it has become approximately "proportional" can be traced with the aid of the formula

$$\text{ctg} \frac{\bar{\theta}}{2} = \text{ctg} \frac{\bar{\theta}_p}{2} + \alpha(z - z_p) + \frac{\alpha}{a} [\theta(z) - \theta_p]. \quad (32)$$

which expresses the area theorem for our system ($\bar{\theta}$ is the limit of $\bar{\theta}_i$ as $t \rightarrow \infty$, z_p is the coordinate of the point beyond which the pulse can be regarded as "proportional", $\theta_p = \theta(z_p)$, and $\bar{\theta}_p = \bar{\theta}(z_p)$). It follows from (32) that when $2k\pi < \bar{\theta}_p < 2(k+1)\pi$, $\bar{\theta} \rightarrow 2k\pi$ as $z \rightarrow \infty$. When $A_{40} = 0$ the area $\bar{\theta}_p$ is less than 2π , so the pulse of the field A_4 must obviously damp out to zero ($\bar{\theta} \rightarrow 0$) at large distances. At finite distances, however, a considerable fraction of the energy stored in the medium may turn up in the "proportional" pulse. For example, for $\theta_0 = 3\pi/2$, $\theta = \pi/2$, and $z = 2/a$, the energy-conversion coefficient

$$\beta(z) = \frac{a}{\alpha} [\bar{\theta}(z) - \bar{\theta}_0] [\theta_0 - \theta(z)]^{-1},$$

becomes

$$\beta(z) = \frac{a}{\alpha \pi} \left\{ 2 \arccot \left[\frac{\alpha}{a} (2 - \pi) + \text{ctg} \frac{\bar{\theta}_0}{2} \right] - \bar{\theta}_0 \right\},$$

and reaches the value $\beta \approx 0.6$ if $\bar{\theta}_0 = \pi$ and $\alpha = a$.

6. CONCLUSION

Thus, the interaction of bichromatic USLP propagating successively in a medium under conditions of two-photon resonance with one of the transitions between energy levels of the medium retains a number of features of quasistationary RFPI: the generated wave (not present at the input) becomes phase locked in the first stage; the region in which phase locking takes place narrows spatially with increasing wave mismatch; and an increase in the wave mismatch leads to a decrease in the energy of the generated wave, this energy falling to zero in the limit $|\delta k| \rightarrow \infty$. On considering the interaction of USLP that do not coincide in space and time, however, we find considerable changes in the nature of the process. These changes lead, first of all, to amplification of a bichromatic pulse at a finite distance, even in the case of arbitrarily large wave mismatch. At the same time, the pulse may damp out to zero at large enough distances even in systems with exact wave synchronism (in quasistationary RFPI at $\delta k = 0$ a steady-state intensity is established at large distances as a result of parametric transparency (see Ref. 20 and the literature cited there)). The amplification at large distances is a threshold effect: the strength of the trigger field must exceed a certain threshold value. The present case differs from that of quasistationary RFPI in that the limiting conversion coefficient is independent of the strength of the trigger field (provided the threshold is exceeded) and the final energy of the generated field is established, not as a result of parametric transparency of the medium, but as a result of parametric transparency of the medium, but as a result of the development against the background of the trigger-field pulse of a subpulse of area 2π that satisfies the proportionality condition.

¹⁾This result can evidently be attributed to the parametric transparency that arises when all the fields interact simultaneously and limits the frequency conversion efficiency (see Ref. 18 for quasistationary RFPI of the type under consideration and Refs. 11 and 12 for the generation of the third harmonic of the ultrashort pump pulse under two-photon resonance).

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