Coherence effects in optical-frequency summation under two-photon resonance conditions

V. I. Anikin, K. N. Drabovich, and A. N. Dubovik

Moscow State Univerity
(Submitted August 5, 1976)
Zh. Eksp. Teor. Fiz. 72, 1727–1737 (May 1977)

The summation of optical frequencies in coherent pulsed excitation of a two-photon resonance is investigated. It is shown that in this case powerful ultrashort tunable pulses in the ultraviolet (including the vacuum ultraviolet) can be generated. It is found that principal conversion conditions for third-harmonic generation are the resultant 0π-pulse regime and the 2π-pulse regime. It is shown that Raman and parametric processes strongly affect the two-photon self-induced transparency effect. Cascade generation of higher harmonics under coherent resonant excitation is analyzed and its high efficiency is pointed out. Numerical estimates are given for some specific substances.

PACS numbers: 42.65.Bp, 42.65.Cq

1. INTRODUCTION

By now the steady-state theory of resonant parametric processes in which the interaction of harmonic light waves is considered has been quite thoroughly developed (see, e.g., Refs. 1–4), and the basic conclusions of this theory concerning the effects of saturation and the Stark shifts of the levels have been confirmed experimentally.[5,6] However, the applicability of the steady-state theory to the analysis of experimental situations is limited to the case in which the times characteristic of the changes in the interacting waves are considerably longer than the longitudinal and transverse relaxation times T_1 and T_2 of the resonant transition.

To realize resonant parametric conversion it is especially important to use intense tunable sources[7–9] (e.g., parametric light generators or dye lasers), which usually give pulsed emission with a broad spectrum ($\Delta\nu \gtrsim 1\ \mathrm{cm}^{-1}$). Moreover, there is a tendency to use picosecond pulses, and in the field of such pulses saturation, which greatly limits the conversion in the steady-state case, sets in at the higher intensities.[8,9] In most of these cases the steady-state theory turns out to be inapplicable, and it becomes urgently necessary to develop a nonstationary theory that would take account of the amplitude and phase modulation of the exciting radiation.

We note that some problems in the theory of the summation of optical frequencies in the pumping noise field under resonance conditions have been discussed elsewhere,[7] and that similar calculations have been made in the adiabatic-following approximation for pulsed radiation.[3,10]

Here we present a systematic discussion of the summation of optical frequencies in media having a center of inversion under two-photon resonance conditions in the field of ultrashort pulses of duration $\tau \ll T_1, T_2$. As was shown earlier,[3] in this case the parametric conversion process turns out to be closely associated with coherent resonance phenomena such as self-induced transparency.

2. BASIC EQUATIONS

Let us first consider the third-harmonic generation process. We shall assume that the nonlinear medium consists of an ensemble of atoms or molecules with eigenfrequencies ω_{mn} and that the electromagnetic field acting on the medium consists of plane waves of frequencies ω_1 and $\omega_3 = 3\omega_1$ with slowly varying amplitudes $A_1(z,t)$ and $A_3(z,t)$ propagating in the z direction.

$$E(z,t) = A_1(z,t)e^{-i(\omega_1 t - k_1 z)} + A_3(z,t)e^{-i(3\omega_1 t - k_3 z)} + \text{c.c.}$$

We shall also assume that the frequency of the fundamental radiation is close to the frequency for two-photon resonance with levels 1 and 2 ($2\omega_1 - \omega_{21} = \delta$) and that the duration τ of the pulse is short as compared with both T_1 and T_2. Then the complete set of equations for the interaction of the waves has the form[3,7]

$$\begin{gathered}
\frac{\partial A_1}{\partial z} + \frac{1}{v_1}\frac{\partial A_1}{\partial t} = i\gamma_1\left[\frac{a_1}{2}(n-n_0)A_1 + 2q\sigma_{21}A_1^* + r\sigma_{12}A_3e^{i\Delta kz}\right], \\
\frac{\partial A_3}{\partial z} + \frac{1}{v_3}\frac{\partial A_3}{\partial t} = i\gamma_3\left[\frac{a_3}{2}(n-n_0)A_3 + r\sigma_{21}A_1e^{-i\Delta kz}\right], \\
\partial\sigma_{21}/\partial t - i(\delta-\Omega_1-\Omega_3)\sigma_{21} = i(qA_1^2 + rA_3A_1^*e^{i\Delta kz})n. \\
\partial n/\partial t = -4\,\mathrm{Im}[(qA_1^{*2} + rA_1A_3^*e^{-i\Delta kz})\sigma_{21}].
\end{gathered} \tag{1}$$

Here we have used the following notation:

$$r = \frac{1}{\hbar^2}\sum_n d_{1n}d_{n2}\left(\frac{1}{\omega_{n1}+\omega_1} + \frac{1}{\omega_{n1}-3\omega_1}\right),$$

and

$$q = \frac{1}{\hbar^2}\sum_n \frac{d_{1n}d_{n2}}{\omega_{n1}-\omega_1}$$

are the matrix elements for the Raman and two-photon couplings, the $\Omega_{1,3} = a_{1,3}|A_{1,3}|^2$ are the field-induced Stark shifts, $\gamma_{1,3} = 2\pi N\omega_{1,3}^2\hbar/k_{1,3}c^2$, N is the particle density, σ_{21} is the off-diagonal element of the density matrix, n is the population difference between levels 1 and 2, n_0 is the equilibrium value of n, $\Delta k = k_3 - 3k_1$ is the wave-vector mismatch of the interacting waves,

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and the $v_{1,3}$ are the propagation velocities of the waves in the unexcited medium.

When $q\neq 0$ and $r=0$, Eqs. (1) describe the propagation of short pulses under two-photon resonance conditions and include the self-induced transparency effect, which was predicted theoretically by Belenov and Poluéktov[11] and has been observed experimentally.[12,13] When $q=0$ and $r\neq 0$, Eqs. (1) describe the Raman scattering of short light pulses,[14,15] including the coherence effects incident to the Raman interaction (see, e.g., Ref. 16). But on the whole, the interaction process is determined by the simultaneous development of three nonlinear effects: parametric generation of the third harmonic, two-photon absorption (or amplification) of the fundamental wave, and Raman interaction of the fundamental wave with the third harmonic. Then, as is evident from Eqs. (1), the equations for the density matrix have the first integral

$$4|\sigma_{21}|^2+n^2=n_0^2.$$

This integral expresses the condition for conservation of the length of the Bloch vector $\mathbf{R}$, which is frequently used in analyzing coherent resonance effects.[17,18]

Now let us make the following simplifying assumptions: the frequency of the fundamental radiation satisfies the condition $\delta=0$ for exact two-photon resonance; the effects of group delay are not important, i.e., $v_1\approx v_3=v$; and there is no mismatch of the wave vectors ($\Delta k=0$). When gaseous media are used, the last condition can be realized in practice by adding a buffer gas.[19] We shall also assume that the pumping wave is present at the entrance to the nonlinear medium, i.e. that $A_1|_{z=0}=A_{10}$ where A_{10} is real, and that the third-harmonic wave, which is absent at the entrance ($A_3|_{z=0}=0$), arises in the medium as a result of the parametric process.

It is evident from Eqs. (1) that under the above assumptions and with $a_1=a_3=0$, there is not only no phase modulation of the waves at the entrance to the nonlinear medium, but none arises in the interaction process. It is not difficult to show that in this case the phases φ_3 and φ_1 of the harmonic and pumping waves have a stable equilibrium position: $\varphi_3-3\varphi_1=\pi$ when $qr>0$ and $\varphi_3-3\varphi_1=0$ when $qr<0$. Then the interaction of the waves is described by equations for real amplitudes (see Sec. 4). A similar assertion is also valid for the more general case of summation of optical frequencies under resonance conditions, and we shall make use of it below in Sec. 5.

3. THE WEAK-RAMAN-COUPLING CASE ($|\beta|=|r/q|\ll 1$)

If the Raman coupling is much weaker than the two-photon coupling, i.e., if $|\beta|=|r/q|\ll 1$, one can neglect the effect of the harmonic field on the amplitude of the fundamental wave and on the parameters σ_{21} and n characterizing the state of the medium. In this case the equations for A_1, σ_{21}, and n describe the two-photon self-induced transparency process, and their solution is well known.[20] It is not difficult to obtain the following expression for the intensity of the harmonic:

$$I_3=|A_3|^2=\frac{9}{4}\beta^2\left\{(A_1-A_{10})^2+\frac{a_1^2}{a_1^2+4q^2}\left[(A_1-A_{10})\operatorname{ctg}\frac{\Psi_0}{2}+2A_1Kz\right]^2\right\}, \quad (2)$$

where

$$\Psi=2q\left(1+\frac{a_1^2}{4q^2}\right)^{1/2}\int_{-\infty}^{\eta}A_1^2(\eta')\,d\eta',\qquad \Psi_0=\Psi(z=0),\qquad \eta=t-\frac{z}{v}, \quad (3)$$

while the evolution of the field of the fundamental wave is described by the formula

$$A_1^2=\frac{A_{10}^2}{1+2Kz[\sin\Psi_0+Kz(1-\cos\Psi_0)]},\qquad K=\gamma|q|\left(1+\frac{a_1^2}{4q^2}\right)^{-1/2},\qquad \gamma=\gamma_1 n_0/2. \quad (4)$$

If the Stark shift induced by the fundamental radiation can be neglected ($k_1\approx 0$), we have

$$I_3={}^9/_4\beta^2(A_1-A_{10})^2, \quad (5)$$

i.e., the intensity of the harmonic is determined solely by the difference between the pumping amplitudes at the entrance to the nonlinear medium and at the point concerned. It is evident from this that at distances of the order of the two-photon absorption length $l_n=K^{-1}$ the harmonic pulse will split into two pulses, which move with different velocities. The first pulse forms at the leading edge of the pumping pulse and propagates with velocity v, while the second pulse moves with the group velocity of the pumping wave. Now if the "area" θ_0 of the pulse at the entrance to the medium satisfies the condition

$$\theta_0=\Psi_0(\eta=\infty)>2\pi m,\quad m=1,\,2,\ldots, \quad (6)$$

then, as has been shown,[20] the fundamental pulse will break up into m subpulses, which increase in amplitude and decrease in duration. In accordance with this, the other (harmonic) pulse also breaks up into m subpulses whose peak intensity increases directly as z^2/l_n^2, with a corresponding decrease in duration, at distances $z\gg l_n$. Thus, third harmonic generation under conditions of coherent two-photon resonance interaction can be used to produce powerful ultrashort pulses in the short-wave region.

The maximum value of the conversion coefficient to the harmonic at $Kz\gg 1$ is

$$\mu\approx\frac{9}{4}\beta^2\left(1+\frac{a_1^2}{4q^2}\right),\qquad \theta_0<2\pi. \quad (7)$$

Thus, the Stark shift increases the conversion efficiency.

4. THE GENERAL CASE; THE EFFECT OF THE RAMAN PROCESS

Let us consider synchronous third-harmonic generation for arbitrary values of the ratio r/q. We shall assume for simplicity that the Stark shifts of the levels can be neglected, i.e., that $a_1=a_3\approx 0$. Then we obtain the following expressions for the density-matrix elements from Eqs. (1):

$$\sigma_{21}={}^1/_2 i n_0 \sin\Psi,\quad n=n_0\cos\Psi, \tag{8}$$

where

$$\Psi=2\int_{-\infty}^{\eta}[qA_1^2(z,\eta')+rA_1(z,\eta')A_3(z,\eta')]d\eta' \tag{9}$$

is the "rotation angle" for the variables specifying the state of the medium. Then the equations for the field amplitudes take the form

$$\frac{\partial A_1}{\partial z}=-\gamma(2qA_1-rA_3)\sin\Psi,\quad \frac{\partial A_3}{\partial z}=-3\gamma rA_1\sin\Psi. \tag{10}$$

The following generalized "energy theorem," analogous to the "energy theorem" given by Poluéktov *et al.*[18] for two-photon self-induced transparency, follows from Eqs. (10):

$$\frac{d}{dz}(W_1+W_3)=-2\gamma(1-\cos\theta),\quad W_{1,3}=\int_{-\infty}^{\infty}A_{1,3}^2\,d\eta,$$
$$\theta=\Psi(z,\infty). \tag{11}$$

Thus, when $\theta=2\pi m$ the sum of the energies of the pumping and harmonic waves is conserved during propagation, although energy exchange between the pulses can take place.

In seeking the basic interaction regimes it is convenient to express the amplitude A_3 of the harmonic in the form

$$A_3(z,\eta)=f(z,\eta)A_1(z,\eta). \tag{12}$$

The function $f(z,\eta)$ satisfies the following equation:

$$\partial f/\partial z=-\gamma(rf^2-2qf+3r)\sin\Psi. \tag{13}$$

It is not difficult to show that the relation between the amplitudes of the interacting waves is given by the equations

$$\frac{A_1^2}{A_{10}^2}=\frac{3\beta}{\beta f^2-2f+3\beta}\exp\left[\frac{2q}{(3r^2-q^2)^{1/2}}\left(\operatorname{arctg}\frac{rf-q}{(3r^2-q^2)^{1/2}}+\operatorname{arctg}\frac{q}{(3r^2-q^2)^{1/2}}\right)\right],\quad |\beta|>\frac{1}{\sqrt{3}};$$
$$\frac{A_1^2}{A_{10}^2}=\frac{3}{(f\mp\sqrt{3})^2}\exp\left(-\frac{2f}{f\mp\sqrt{3}}\right),\quad \beta=\pm\frac{1}{\sqrt{3}}; \tag{14}$$
$$\frac{A_1^2}{A_{10}^2}=\frac{3\beta}{\beta f^2-2f+3\beta}\left(\frac{\alpha_2}{\alpha_1}\frac{rl-\alpha_1}{rf-\alpha_2}\right)^{q/(q^2-3r^2)^{1/2}},\quad |\beta|<\frac{1}{\sqrt{3}},$$

in which $\alpha_{1,2}=q\pm(q^2-3r^2)^{1/2}$. Formulas analogous to Eqs. (14) have been obtained by Afanas'ev and Manykin[1] for third-harmonic generation by two-photon resonance under steady-state conditions. It is easily seen that for the case of weak Raman coupling ($|\beta|\ll1$), Eqs. (14) reduce to Eq. (5).

The following interaction regimes are possible, depending on the ratio β of the Raman and two-photon interaction constants.

A. *Production of* 0π *pulses.* In this regime the amplitude ratio

$$f=-1/\beta,\ \text{i.e.,}\ qA_1+rA_3=0. \tag{15}$$

becomes established at distances $z\gg l_n$. Thus, the integrand in Eq. (9) vanishes throughout the pulse, and $\theta=0$. Physically, this means that two-photon excitation of the resonance transition is suppressed by the Raman interaction of the waves A_1 and A_3, which is in phase opposition to the two-photon interaction. This is the principal difference between the 0π pulses considered here and the 0π pulses arising in one-photon self-induced transparency, in which the null rotation of the Bloch vector is due to phase shifts within the pulse.[18] It is evident from Eqs. (14) and (15) that in this regime the fundamental and harmonic pulses have the same shape as the initial pumping pulse. The harmonic-conversion factor is given by

$$\mu=\frac{1}{1+\beta^2}\exp\left[-\frac{2}{(3\beta^2-1)^{1/2}}\operatorname{arctg}\frac{(3\beta^2-1)^{1/2}}{3\beta^2+1}\right],\quad |\beta|>\frac{1}{\sqrt{3}},$$
$$\mu=\frac{1}{1+\beta^2}\left(\frac{\alpha_2}{\alpha_1}\frac{q+\alpha_1}{q+\alpha_2}\right)^{q/(q^2-3r^2)^{1/2}},\quad |\beta|<\frac{1}{\sqrt{3}}. \tag{16}$$

When $|\beta|\ll1$, we have $\mu=(9/4)\beta^2$, in agreement with the estimate given in the preceding section (see Eq. (17).

The 0π-pulse regime exists for all values of β and, as is easily shown, is stable.

B. *The "proportional" regime.* This regime is possible only when $|\beta|<1/\sqrt{3}$ and is characterized by the relation

$$f_{1,2}=\alpha_{1,2}/r. \tag{17}$$

It is evident from Eq. (13) that in this case the amplitude ratio f does not change with distance, even though Ψ may still depend on η and z. The amplitudes A_1 and A_3 evolve according to identical equations, which have the same form as the equations for two-photon self-induced transparency,[18,20] but with the substitution

$$K\to K_1=\frac{\gamma|q|}{2}[1+(1-3\beta^2)^{1/2}],\quad \Psi=2(q+\alpha_{1,2})\int_{-\infty}^{\eta}A_1^2\,d\eta'. \tag{18}$$

Consequently, 2π pulses, whose peak intensity increases with distance and whose duration decreases in proportion to $(K_1z)^2$, can be formed in the "proportional" regime.

The "proportional" regime can be realized by feeding both pumping and harmonic pulses, satisfying the condition $A_{30}=f_{1,2}A_{10}$, into the nonlinear medium. Numerical solution of Eqs. (10) on a computer shows that this regime can also be realized under ordinary conditions, when $A_{30}=0$; in this case $f=\beta^{-1}(1-(1-3\beta^2)^{1/2})$. In this regime the two-photon and Raman processes contribute proportionally to the excitation of the two-photon transition. The equations for $A_{1,3}$ admit a soliton solution having the Lorentz shape, which is analogous to the soliton solution for two-photon self-induced transparency with the appropriate substitution (18).

The development of the interaction process in the case $|\beta|<1/\sqrt{3}$ presents the following over-all appearance: at the leading edge of the fundamental pulse there arises a harmonic pulse, which moves with velocity v, and the resultant 0π pulse discussed above is formed at distances $z \gg l_n$.

If the initial pulse $A_{10}(\eta)$ satisfies the condition

$$\theta_0 \geqslant 2\pi\left\{\frac{1}{2-(1-3\beta^2)^{1/2}}\left[1-\left(\frac{\alpha_2}{\alpha_1}\,\frac{q+\alpha_1}{q+\alpha_2}\right)^{q/(q^2-3r^2)^{1/2}}\right]^{-1}\right\}, \qquad (19)$$

the trailing parts of the pulse reach the "proportional" regime. Formula (19) is actually the condition for the existence of 2π pulses in the process under consideration.

When the Raman coupling is weak ($|\beta|\ll 1$) the 2π-pulse threshold is slightly higher than in the case of "pure" two-photon interaction: $\theta_0 \geqslant 2\pi(1+(9/4)\beta^2)$. The θ_0 threshold increases with increasing $|\beta|$ and becomes infinite at $|\beta| = 1/\sqrt{3}$, Thus, when $|\beta| \geqslant 1/\sqrt{3}$ the formation of 2π pulses (the "proportional" regime) is not possible at any initial-pulse intensities: equilibrium between the two-photon and Raman interactions is always reached and 0π pulses are formed at distances $z \gg l_n$. A fraction of the energy W_0 of the initial pulse, given by

$$W=W_0\left[1-\exp\left(-\frac{2}{(3\beta^2-1)^{1/2}}\operatorname{arctg}\frac{(3\beta^2-1)^{1/2}}{3\beta^2+1}\right)\right], \qquad (20)$$

is absorbed by the medium during the process.

Numerical solution of Eqs. (10) on a computer confirms the results of the above analysis. Figure 1 shows the evolution of the pulses when condition (19) is satisfied. It is evident that short pulses, which increase in intensity and decrease in duration, are formed at the trailing edge; the resultant 0π pulse is formed at the leading edge.

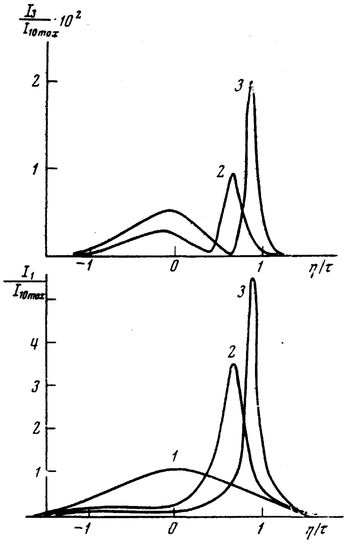

FIG. 1. Evolution of the pumping and third-harmonic pulses at $\theta=2.2\pi$ and $|\beta|=0.06$: 1—pumping pulse at the entrance; 2, 3—pulses at $z=l_n$ and $z=2l_n$.

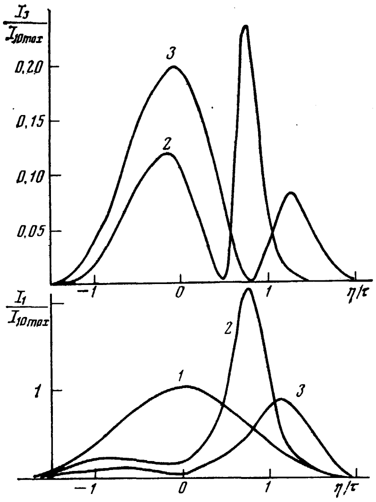

FIG. 2. Evolution of the pumping and third-harmonic pulses at $\theta_0=2.2\pi$ and $|\beta|=0.5$: 1—pulses at the entrance; 2, 3—pulses at $z=l_n$ and $z=2l_n$.

When $|\beta|<1/\sqrt{3}$, however, and condition (19) is not satisfied, the formation of short pulses at the pumping and harmonic frequencies, which begins at short distances, subsequently ceases (Fig. 2). When $|\beta|\geqslant 1/\sqrt{3}$, on the other hand, no short pulses are formed at the trailing edge at all (Fig. 3). Separate intensity pulsations can arise during the formation of the 0π pulse.

In concluding this section we note that the effects discussed can also occur when $\Delta k \neq 0$, but for this it is necessary that the synchronous interaction length be considerably greater than the two-photon absorption length, i.e., that $\Delta k l_n \ll 1$.

5. SUMMATION OF OPTICAL FREQUENCIES

Let us briefly consider the more general case in which which there are two pulses of frequencies ω_1 and ω_2 and real amplitudes $A_{10}(\eta)$ and $A_{20}(\eta)$ at the entrance to the nonlinear medium, the frequency ω_1 satisfying the condition $\delta=0$ for exact two-photon resonance. In this case resonance polarization arises at both the third-

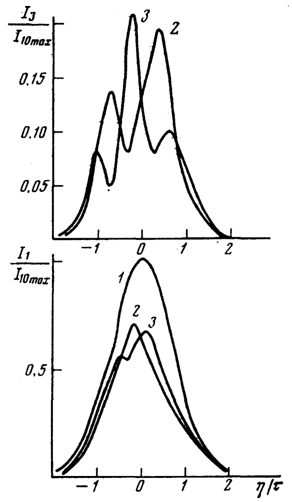

FIG. 3. Evolution of the pumping and third-harmonic pulses at $\theta_0=2.2\pi$ and $|\beta|=2$: 1—pumping pulse at the entrance; 2, 3—pulses at $z=l_n$ and $z=2l_n$.

harmonic frequency and the summation frequency $\omega_4 = 2\omega_1 + \omega_2$. To simplify the problem we shall assume that the condition for synchronism is satisfied only for the summation frequency ($k_4 = 2k_1 + k_2$), while the wave-vector mismatch for third-harmonic generation is large ($\Delta k l_n \gg 1$). Then we need consider only the interactions of three waves with amplitudes A_1, A_2, and A_4, where A_4 is the amplitude of the wave at the summation frequency. Neglecting the Stark shifts of the levels, we can write the equations for this interaction in the form

$$\partial A_1/\partial z = -2\gamma q A_1 \sin \Psi_1, \quad \partial A_2/\partial z = \gamma_2 r_2 A_4 \sin \Psi_1, \quad \partial A_4/\partial z = -\gamma_4 r_2 A_2 \sin \Psi_1, \tag{21}$$

where

$$\gamma_{2,4} = \frac{\pi N \hbar n_0 \omega_{2,4}^2}{k_{2,4} c^2}, \quad r_2 = \frac{1}{\hbar^2} \sum_n d_{1n} d_{n2} \left(\frac{1}{\omega_{n1}+\omega_2} + \frac{1}{\omega_{n1}-\omega_4} \right), \quad \Psi_1 = 2 \int_{-\infty}^{\eta} (q A_1^2 + r_2 A_2 A_4)\, d\eta'. \tag{22}$$

An "energy theorem" analogous to (11) follows from Eqs. (21):

$$\frac{d}{dz}(W_1 + W_2 + W_4) = -2\gamma(1-\cos\theta_1), \quad \theta_1 = \Psi_1(z, \infty). \tag{23}$$

From this it is evident that when $\theta_1 = 2\pi m$ ($m = 0, 1, 2, \ldots$) the total energy of the pulses does not change with distance, although the pulses may exchange energy with each other.

The first and second integrals of Eqs. (21) agree in form with the integrals obtained by Butylkin *et al.*[2] for the steady-state interaction:

$$A_4^2 = \frac{\omega_4}{\omega_2}(A_{20}^2 - A_2^2), \tag{24}$$

$$A_4 = \left(\frac{\omega_4}{\omega_2}\right)^{1/2} A_{20} \sin\left[\frac{(\omega_2\omega_4)^{1/2}}{2\omega_1} \beta_2 \ln \frac{A_1}{A_{10}}\right], \quad \beta_2 = \frac{r_2}{q}. \tag{25}$$

It is evident from Eqs. (21)–(25) that at small distances $z \ll l_n$ the first pulse interacts with the medium almost independently of the presence of the second pulse. With the appearance of the wave at the summation frequency the Raman process comes into play; then the "overlap integral" of pulses A_2 and A_4 (see Eq. (22)) increases and may considerably affect the "rotation angle" Ψ_1 of the variables specifying the state of the medium and thereby affect the entire process of coherent interaction of the waves with the medium.

If the condition

$$\left| \beta_2 \frac{A_{20}(\eta)}{A_{10}(\eta)} \right| \ll 1, \tag{26}$$

is satisfied within the limits of the pumping pulse $A_{10}(\eta)$, the Raman process will have virtually no effect on the propagation of the first pulse, which in this case will evolve in accordance with Eq. (4). Then if $\theta_0 > 2\pi$, powerful ultrashort pulses, similar to those discussed above, will be formed at frequencies ω_1 and ω_4; as a rule, moreover, there will be more subpulses at the converted frequency than at the fundamental frequency ω_1. In fact, when $|\beta_2| \ll 1$, it follows from Eq. (25) that

$$I_4 = A_4^2 \approx \frac{\omega_4^2}{4\omega_2^2} \beta_2^2 I_{20} \ln^2 \frac{A_1}{A_{10}}. \tag{27}$$

If the pulse at frequency ω_2 completely "covers" the pumping pulse, the maxima of the converted radiation will actually coincide with the extrema of A_1. In this case we shall have

$$I_{4max} \approx \frac{\omega_4^2}{4\omega_2^2} \beta_2^2 I_{20} \ln^2(2Kz)$$

at large distances $z \gg l_n$.

On the whole, the frequency-summation process is much more complicated than the process of third harmonic generation. Nevertheless, it is very important to investigate the summation process, first, because in this case ultrashort frequency-tunable pulses can be generated in the ultraviolet and the vacuum ultraviolet, and second, because the pulses at frequency ω_2 can be used as probe pulses to investigate the effects of two-photon self-induced transparency. For example, if condition (26) is satisfied and $\tau_2 \lesssim \tau$, where τ_2 is the duration of the second pulse, then, if the pulse of converted radiation can be detected, it will be possible to obtain virtually complete information about the evolution of the pumping pulse (see Eqs. (25) and (27)) by varying the time delay of the second pulse with respect to the first one. We note that just such a setup has been employed[21] in experiments on the coherent superposition of quantum states under two-photon resonance conditions.

6. HIGHER-HARMONIC GENERATION

Coherent two-photon resonant excitation can also be used to generate higher harmonics. Here cascade processes of the type

$$\omega_1 + \omega_1 + \omega_1 \to \omega_3 \quad (2\omega_1 = \omega_{21}), \qquad \omega_3 + \omega_1 + \omega_1 \to \omega_5 = 5\omega_1, \tag{28}$$

which are strongly suppressed by the saturation effect in a field of long pulses, may prove to be effective.

It is not difficult to show that the nonlinear resonant polarization at the fifth-harmonic frequency has the form

$$P(5\omega_1) = N\hbar r_5 \sigma_{21} A_3 e^{-i5\omega_1 t} + \text{c.c.}$$

where

$$r_5 = \frac{1}{\hbar^2} \sum_n d_{1n} d_{n2} \left(\frac{1}{\omega_{n1}+3\omega_1} + \frac{1}{\omega_{n1}-5\omega_1} \right).$$

Under conditions of spatial synchronism ($\Delta k = 0$ and $k_3 + 2k_1 = k_5$) the amplitude of the fifth harmonic is given by

$$A_5 = i\gamma_5 r_5 \int_0^z \sigma_{21}(z', \eta) A_3(z', \eta)\,dz'. \quad (29)$$

On neglecting the effect of the Raman process, we easily find that

$$A_5 = -\frac{15}{4}\frac{r_5 r}{q^2}\left[A_{10}\left(\ln\frac{A_1}{A_{10}}+1\right)-A_1\right], \quad (30)$$

in which A_1 is given by Eq. (4). Consequently, ultrashort pulses analogous to those obtained in third-harmonic generation, which increase in intensity and decrease in duration, can be generated at the fifth-harmonic frequency. The maximum intensity $I_{5\,\max}$ of these pulses at $z \gg l_n$ is given by

$$I_{5\max} \approx \frac{225}{4}\left(\frac{r_5 r}{q^2}\right)^2 (Kz)^2. \quad (31)$$

In conclusion, we note that nondegenerate cascade processes of the type

$$2\omega_1+\omega_2\to\omega_4, \quad 2\omega_1+\omega_4\to\omega_5=4\omega_1+\omega_2,$$

are also effective in coherent excitation and make it possible to obtain ultrashort tunable pulses in the ultraviolet and the vacuum ultraviolet.

7. CONCLUSION

Now let us present a few numerical estimates for specific substances. For the $3s$–$4s$ transition in sodium we have $q \approx -4.5\cdot 10^4$, $r \approx -2.7\cdot 10^3$, and $a_1 \approx 9\cdot 10^4$ (cgs esu), and according to Eq. (2) a conversion coefficient to the third harmonic ($\lambda_3 \approx 2590$ Å) of ~1.5% can be achieved. With $N \approx 10^{16}$ cm^{-3}, effective narrowing of the pulses takes place in lengths of 10–20 cm.

Similarly, for two-photon resonance with the $2s$–$5s$ transition in lithium we have $q \approx 3.8\cdot 10^3$, $r \approx 7\cdot 10^2$, and $r_5 \approx 1.2\cdot 10^2$ (cgs esu), and the conversion to the third harmonic ($\lambda_3 \approx 1730$ Å) under coherent interaction amounts to ~8%, and the conversion to the fifth harmonic ($\lambda_5 \approx 1040$ Å) may accordingly reach ~1%.

We note that four-photon parametric processes under conditions of coherent two-photon excitation may also prove to be useful for generating frequency-tunable pulses in the infrared. Raman scattering from transitions excited by a two-photon process when two pulses with frequencies ω_1 and ω_2 satisfying the conditions $2\omega_1 - \omega_{21} \approx 0$ and $\omega_2 > \omega_{21}$ are fed into the nonlinear medium would seem to be promising for this purpose. In this case the resultant frequency $\omega_4 = 2\omega_1 - \omega_2$ may lie in the far infrared. We note that this process has been examined by Venkin *et al.*[22] for the steady-state case without allowance for population changes.

The authors are deeply grateful to S. A. Akhmanov for his interest in the work and for valuable discussions.

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Translated by E. Brunner