Propagation of short optical pulses during third-harmonic generation in the case of two-photon resonance

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(Submitted 13 July 1978)
Zh. Eksp. Teor. Fiz. 76, 856–865 (March 1979)

A numerical study has been carried out of third-harmonic generation in the field of an ultrashort pulse in the case of two-photon absorption of the fundamental radiation. This phenomenon is looked upon as a special case of a broad class of parametric resonance processes and has been found to have much in common with effects such as two-photon self-induced transparency. The medium was found to lose coherence in a time shorter than the length of the pump pulse. This was due to a shift in the resonance frequency in the strong electromagnetic field and to inhomogeneous broadening. Harmonic generation in two-photon amplifiers is discussed. Considerable phase modulation of the harmonic wave was observed in all cases.

PACS numbers: 42.50. + q, 42.65.Cq, 42.65.Gv

1. INTRODUCTION

Interactions between electromagnetic waves under resonance conditions form a branch of nonlinear optics that has recently undergone considerable development.[1-10] The current interest in such processes is connected with the considerable gain in the efficiency of frequency conversion of radiation as compared with the nonresonance case.[1,2] The progress that has already been achieved evokes the hope that tunable sources of coherent radiation in the vacuum ultraviolet[3] and the far infrared are not too far off. Third-harmonic generation (THG) in media exhibiting resonance behavior at twice the pump frequency[4-8] has attracted considerable attention. The theoretically predicted increase in the THG efficiency under these conditions[4,5] has been confirmed experimentally.[6] These papers (and the subsequent publications[1,2]) have stimulated intensive studies of the THG phenomenon as well as other methods for frequency multiplication under the conditions of two-photon resonance. The quasistationary theory of parametric resonance processes (PRP),[7-11] in which the radiation pulse length τ_p is much greater than the population relaxation time T_1 and the polarization relaxation time T_2, is the most highly developed. It has been found[7,8] that the conversion efficiency is restricted mainly by the saturation phenomenon. On the other hand, PRP in the field of ultrashort pulses ($\tau_p \ll T_1, T_2$) appears to be free from this restriction. There is, therefore, considerable interest in the propagation of ultrashort pulses (USP) during PRP.

USP propagation during two-photon resonance was first examined by Belenov and Poluéktov,[12] who predicted the two-photon self-induced transparency effect which was subsequently confirmed experimentally[13] and by numerical studies.[14,15] Another example of PRP is Raman scattering, for which solitary waves of pump and scattered radiation have been detected. Their existence is closely connected with the dispersion of the medium.[16,17] Anikin *et al.*[18] and Poluéktov[19] have discussed THG during resonance at twice the frequency of main radiation USP and have demonstrated the existence of pump and harmonic solitary waves, ensuring parametric transmission.[20] Third-harmonic generation has been investigated to a much less extent than the other PRP enumerated above. This is probably connected with difficulties encountered in the analysis of the THG equations. All this leads to a number of interesting and nontrivial examples of wave equations with solutions in the form of solitary waves.

One such case is examined in this paper, namely, the propagation of ultrashort pulses of electromagnetic radiation during third-harmonic generation under the condition of resonance at twice the pump frequency. The equations describing this process (Sec. 2) are given in the approximation of slowly-varying complex amplitudes of the interacting waves and the variables of the resonance system (components of the Bloch vector[12,21]). The equations are then solved numerically. It is found that the number of peaks in the pump pulse obeys the following general rule[12]: the initial $2\pi N$ pulse is transformed into a pulse with N well-resolved maxima (Sec. 3). In contrast to most of the publications that have appeared so far, we have taken into account inhomogeneous broadening of the resonance transition, the effect of which is discussed in Sec. 4. The propagation of ultrashort pulses of pump and harmonic radiation in an amplifying medium is also discussed (Sec. 5). This differs from the corresponding picture for the case of noninverted population.

2. BASIC EQUATIONS

The equations describing third-harmonic generation in our case have frequently been derived in the literature.[10,11,18,19] Consider the interaction of two plane waves propagating in the z direction with complex amplitudes $\mathscr{E}_i = A_i e^{i\varphi_i}$ ($i = 1, 3$, pump wave $\omega_1 = \omega$, harmonic $\omega_3 = 3\omega$) in a medium exhibiting resonance properties at the frequency $\omega_{21} \approx 2\omega$. Let us suppose that the phase-locking condition is satisfied, and no other energy levels participate in the resonance. The evolution of the medium can then be described by the reduced density matrix[21] ρ_{ij}, where

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$$w=(\rho_{11}-\rho_{22}),\quad v=\sigma\rho_{21}\exp\left[2i(\omega t-k_1 z)\right],\tag{1}$$

are slowly-varying functions of position and of time. In this approximation, the complete set of THG equations is

$$\begin{aligned}&\partial v/\partial\tau=-i(\delta+\alpha_n)v+iw(\varepsilon_1^2+\beta\varepsilon_1^*\varepsilon_3),\\&\partial w/\partial\tau=\mathrm{Im}\left[v^*(\varepsilon_1^2+\beta\varepsilon_1^*\varepsilon_3)\right],\end{aligned}\tag{2a}$$

$$\begin{aligned}&\partial\varepsilon_1/\partial\zeta=i[\alpha_1\langle w-w_0\rangle\varepsilon_1+2\langle v\rangle\varepsilon_1^*+\beta\langle v^*\rangle\varepsilon_3],\\&\partial\varepsilon_3/\partial\zeta=i3[\alpha_3\langle w-w_0\rangle\varepsilon_3+\beta\langle v\rangle\varepsilon_1]\end{aligned}\tag{2b}$$

where

$$\varepsilon_i=\mathscr{E}_i/\Gamma,\quad \Gamma=\max\mathscr{E}_1(t,z=0),\quad \beta=r/q,\quad \zeta=z/L,$$
$$\tau=\Omega(t-z/c),\quad \delta=(\omega_{21}-2\omega)/\Omega,\quad \Omega=2\Gamma^2|q|,$$
$$L^{-1}=(2\pi n\omega^2\hbar|q|/k_1c^2),$$

n is the density of resonance atoms, and the parameters q and r which describe the interaction between the fields and the medium have the following form:

$$r=2\hbar^{-2}\sum_j d_{1j}d_{j2}(\omega_{j1}-2\omega)/(\omega_{j1}+\omega)(\omega_{j1}-3\omega),$$

$$q=\hbar^{-2}\sum_j d_{1j}d_{j2}/(\omega_{j1}-\omega).$$

The symbol σ introduced in (1) represents $q/|q|$. The shift of the resonance energy levels in the fields $\mathscr{E}_1$ and $\mathscr{E}_3$ is represented by the coefficients α_1 and α_3 which can be expressed in terms of the Stark constant[18] as follows:

$$\alpha_i=a_i/|q|,\quad i=1,3,$$
$$\alpha_n=(\alpha_1|\varepsilon_1|^2+\alpha_3|\varepsilon_3|^2)/2.$$

The angle brackets in (2b) represent averaging over the resonance frequency ω_{21}, the spread of which is responsible for the inhomogeneous broadening of the resonance line:

$$\langle\ldots\rangle=\int_{-\infty}^{\infty}g(\Delta\omega)\ldots d\Delta\omega,$$

where $g(\Delta\omega)$ is the Gaussian function normalized to unity with a full width of $1/T_2^*$.

Equations (2a) and (2b) were solved numerically for different values of β, τ_p/T_2^*, and Ω^2. The reference values were chosen to correspond to the $3s-4s$ transition in sodium[18]: $\beta=0.06$, $\alpha_1=\alpha_3=2$, $T_2^*=10^{-9}$ sec, $q=-4.5\times10^4$ cgs, $2\omega=10^{15}$ sec$^{-1}\cdot$deg, $n=10^6$ cm^{-3}; Γ^2 is equal to 10^5 cgs, which corresponds to an electric field of the order of 10^5 V/cm. It is assumed that the medium is initially in the ground state ($\langle w_0\rangle=1$), and is illuminated by a Gaussian pulse of the fundamental radiation; the harmonic is absent.

The parameter Ω can be interpreted as the frequency of the two-photon Rabi precession[22] in an electromagnetic field of constant amplitude (equal to Γ). In the present case, $\Omega^{-1}\sim10^{-10}$ sec. The length L is given in terms of the nonlinear absorption length L_{nl} as follows[12]

$$L=L_{\mathrm{n}}(1+\alpha_1^2/4)^{-1/2}$$

and is of the same order of magnitude.

A numerical solution of Eqs. (2a)–(2b) was performed by the Runge-Kutta method to a precision of the order of 1%. Further subdivision of the computational step resulted in a smaller error, showing that the chosen difference scheme was stable. The integration in (2b) was performed by the trapezium method with an accuracy of better than 0.1%.

3. PROPAGATION OF PULSES OF DIFFERENT ENERGY

It is convenient to use a geometric treatment of the evolution of the resonance system, namely, the two-photon vector model,[21,22] as a means of qualitative description of the effects arising during the propagation of ultrashort pump and harmonic pulses under the condition of two-photon resonance. In this model, the Bloch vector **P** with components

$$P_1+iP_2=ve^{-i2\varphi_1},\quad P_3=w,\tag{3}$$

rotates around the effective field **R** with instantaneous frequency equal to $|\mathbf{R}|$, where

$$\begin{aligned}&R_1=S_1^2+\beta S_1S_3\cos\Phi,\quad R_2=\beta S_1S_3\sin\Phi,\\&R_3=\delta+\alpha_n+2\partial\varphi_1/\partial\tau,\quad S_i=|\varepsilon_i|\end{aligned}\tag{4}$$

and $\Phi=\varphi_3-3\varphi_1$. While the pulse is on, the vector **P** rotates through an angle

$$\Psi(\delta,\zeta)=\int_{-\infty}^{\infty}|\mathbf{R}(\delta,\zeta,\tau)|d\tau.\tag{5}$$

Whenever $\Psi=2\pi N$, the medium returns to its original state after the transmission of the pulse. This is analogous to the self-induced transparency in the case of single-photon resonance.[23] The vector model can always be introduced, whatever the order of the resonance, provided only the two-level model is valid, so that the transparency phenomenon is expected for all multiphoton processes during the propagation of the ultrashort pulses.

When harmonic conversion is small, $A_1\gg A_3$, and the pump is strong enough, $a_1A_1^2\gg1/T_2^*$, the direction of the effective field vector makes an angle with the P_3 axis, whose maximum value can be estimated from the formula

$$\theta=\arccos\left[a_1/(a_1^2+4q^2)^{1/2}\right]=\arccos\left[(1+\alpha_1^2/4)^{-1/2}\alpha_1/2\right].\tag{6}$$

Having rotated around **R** through the angle π, the Bloch vector assumes a third component, given by

$$P_3=w_0[2\cos^2\theta-1].$$

The presence of the Stark shift of the resonance level is thus seen to prevent the complete inversion of the population in the case of two-photon resonance. This qualitative result is valid not only in the limit of strong and rapidly-varying fields. It is also valid in the adiabatic sequence,[22] and is in agreement with the behavior of the population obtained by the numerical solution of the problem.

In general, the Bloch vector depends on the frequency detuning δ. Let us take **P** with $\delta=0$. The corresponding angle of rotation for $\zeta=0$ will be looked upon as the characteristic indicating the possibility or otherwise of the transparency effect. Using the initial conditions and (5), we find that

$$\Psi_0=\Psi(\delta=0,\zeta=0)=(1+\alpha_1^2/4)^{1/2}\int_{-\infty}^{\infty}S_1^2(\tau,\zeta=0)\,d\tau$$

where Ψ_0 is the energy of the incident pump pulse to within a factor.

When $\Psi_0=1.33$, the propagation of the fundamental and harmonic pulses turns out to be similar to the propagation of pulses with "energy" less than 2π in the theory of two-photon self-induced transparency[12-15] with the one difference that the attenuation of the pump pulse is accompanied by the transfer of energy to the harmonic pulse. The difference between the populations of the resonance levels is found to vary only slightly.

A more interesting behavior of the pulses of interacting waves is found to occur for $\Psi_0 = 2.66\pi$ (Fig. 1). The medium absorbs energy on both the leading and trailing edges of the pump pulse. Some of it is returned to the harmonic and the pump, and some remains in the medium. At a certain time, the Bloch vector assumes a position perpendicular to the P_3 axis, and the population difference is zero. This is so because the coefficient α_1 [see (6)] is chosen to be equal to 2, i.e., the Stark coefficient a_1 is equal to twice the modulus of the combined matrix element of the two-photon coupling. The maximum possible polarization is induced in the medium. Since the only mechanism for the relaxation of a coherent state of the medium is the dephasing of the radiators through inhomogeneous broadening, and $\tau_p/T_2^* = 0.2$, this superradiant state persists for the duration of the pulses, and the return of the energy from the medium to the field occurs exceedingly rapidly, i.e., in a time of the order of Ω^{-1}. A sharp peak is produced in the central part of the pulses, the amplitude of which grows and the width decreases in the course of propagation. At the same time, there is a change in the phase difference Φ due to the real part of the polarization.

The propagation of the pump pulse with $\Psi_0 = 5.32\pi$ and $\tau_p/T_2^* = 0.4$ has some of the features of the propagation of ultrashort pulses under the conditions of only the two-

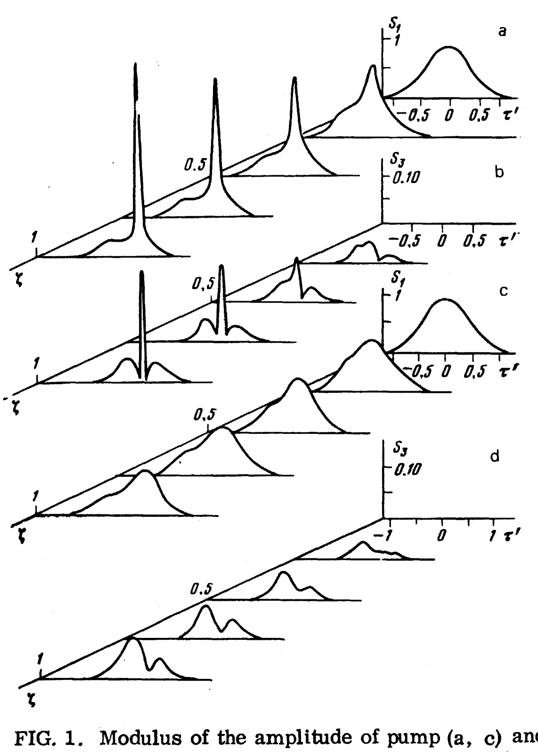

FIG. 1. Modulus of the amplitude of pump (a, c) and harmonic (b, d) as functions of position ζ and of time $\tau' = t/\tau_p$ for different inhomogeneous relaxation times T_2^*.

photon absorption, namely, the Bloch vector passes through its original position ($\Psi_0 > 4\pi$) twice, and two maxima appear on the profile of the fundamental pulse. The more complicated structure of the modulus of the harmonic-pulse envelope is due to the presence of the two sharp peaks on the pump pulse, but, otherwise, the THG picture is qualitatively similar to the preceding case.

It is important to note that while we have everywhere the propagation of one pump pulse and one harmonic pulse, the harmonic envelope has positive and negative segments as a result of the phase modulation of the $\mathscr{E}_3$ wave. It is precisely this that leads to the pattern resembling the subdivision of a pulse into subpulses. The true envelope is

$$E_3 = \mathrm{Re}\,\{\mathscr{E}_3 \exp[i\omega_3 t - ik_3 z]\}$$
$$= \Gamma[\mathrm{Re}\,\varepsilon_3 \cos(\omega_3 t - k_3 z) - \mathrm{Im}\,\varepsilon_3 \sin(\omega_3 t - k_3 z)].$$

Thus, whereas in the case of the single-photon resonance the $2\pi N$-pulse splits into N 2π-pulses that propagate in general with different velocities, in the present case the subdivision of the pulse signifies amplitude and phase modulation of one solitary wave. The "quasipulses" produced in this way have equal propagation velocities, and we have a situation similar to that observed in the case of two-photon self-induced transparency.

4. ROLE OF INHOMOGENEOUS BROADENING AND THE HIGH-FREQUENCY STARK EFFECT

To achieve a better understanding of the effect of inhomogeneous broadening on the propagation of ultrashort pulses during two-photon resonance, it is useful to compare the associated phenomena with the case of single-photon resonance propagation. In the latter case, if the pulse length is large is enough ($\tau_p \gg T_2^*$), the spectrum of an inhomogeneously broadened line will involve the excitation of radiators with frequency spread $\Delta\omega_b \lesssim 1/\tau_p$. The dephasing of the atoms due to this spread occurs in a time $\sim\tau_p$, so that energy transfer between the medium and the electromagnetic field is still not coherent. In the reverse situation ($\tau_p \ll T_2^*$), all the radiators are excited and the dephasing time is determined by T_2^*, so that the coherent state again persists during the time of the pulse.

If the coherent state is excited with the aid of two-photon resonance (or multiphoton resonance), the simultaneous change in the resonance frequency in the strong field of the incident radiation must be taken into account. Suppose that $\tau_p \gg T_2^*$ and the Stark shift is

$$\Delta\omega_s = \sum_i a_i |\mathscr{E}_i|^2 .$$

We now have excited atoms for which the frequency spread is $\Delta\omega_b \lesssim 1/\tau_p + \Delta\omega$. The dephasing of the radiators occurs in a time shorter than τ_p, the polarization decays before the end of the pulse, and some of the field energy remains in the medium. If $\tau_p \ll T_2^*$, the entire inhomogeneously broadened line is excited and, whatever the Stark shift, the dephasing occurs in a time T_2^*, i.e., the coherent state of the medium persists during the operation of the radiation pulse.

It follows from the foregoing qualitative analysis that the influence of inhomogeneous broadening on the propagation of ultrashort pulses during two-photon (multiphoton) resonance is stimulated by the Stark shift, i.e., a phenomenon whose contribution is small in the case of the single-quantum interaction. Inhomogeneous broadening was taken into account by Tan-no *et al.*[24] and Hanamura[25] in the absence of a relative shift between resonance levels in the field of the ultrashort pulses, so that their treatment is incorrect.

The results obtained through a numerical solution of (2a)–(2b) serve as an illustration of the foregoing discussion. The values of β, α_1, α_3, and Γ^2 were the same as in Sec. 3. The pump pulse with $\Psi_0=2.66$, $\tau_p/T_2^*=8$ tended to change its shape during the propagation process (Fig. 1) to a lesser extent than in the case of $\tau_p/T_2^*=0.2$. The harmonic, which was a phase-modulated solitary wave, did not show much change either. The time dependence of the phase difference was more monotonic than in the case of a narrow inhomogeneously broadened line. The influence of inhomogeneous broadening on the propagation of the pump and harmonic waves was particularly appreciable when $\Psi_0=5.32\pi$. The energy in the leading edge of the pump pulse was used to generate the harmonic and to excite the medium. The dephasing of the radiators due to inhomogeneous broadening meant that the coherent return of the energy to the pump and harmonic fields was not very effective, so that the trailing edge of the pump pulse remained practically the same and only a slight "tail" of the harmonic was found to persist.

5. PROPAGATION OF ULTRASHORT PULSES OF INTERACTING WAVES IN AN AMPLIFYING MEDIUM

In addition to the phenomena occurring during the interaction between very short optical pulses and an absorbing resonance medium it is interesting to consider the case of an amplifying medium.[16,26] There is already published experimental work on two-photon amplification of picosecond pulses in coherently inverted potassium vapor.[27] However, the theory of this process is still rather fragmentary. There is only the "energy theorem" obtained in the absence of inhomogeneous broadening. This theorem can be used for the two-photon amplifier[25] to show that a low-energy pulse can be amplified up to the 2π-pulse, but nothing can be said about its shape. If this solitary wave is established, it will leave behind an inverted medium because the Bloch vector returns to its original position after a rotation through 2π. A similar result for the energy of the fundamental radiation and its third harmonic is predicted by the "generalized energy theorem"[18] obtained in the limit of an infinitely narrow resonance line and zero Stark shift of resonance levels.

Let us consider third-harmonic generation in the medium in which all the atoms were first inverted, i.e., $\langle w_0\rangle=-1$. All the other parameters are the same as in Sec. 2 and $\tau_p/T_2^*=0.2$. Numerical solution of Eqs. (2a)–(2b) has shown that a Gaussian pump pulse with $\Psi_0=2.66$ propagates in a way that is qualitatively different from the case of a noninverted medium (Fig. 2). Amplification produces a sharp peak on the leading edge, which shifts in the direction of propagation. Harmonic generation occurs at the same time. The population dif-

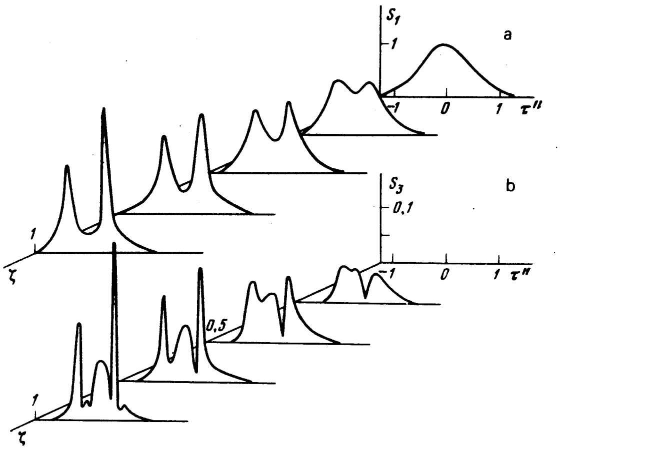

FIG. 2. Propagation of pump (a) and harmonic (b) pulses in the two-photon amplifier, $\tau''=\Omega^{-1}\tau/\tau_p$.

ference reaches its zero value, and the medium assumes a state that can be identified with the superradiant state. For the chosen value of τ_p/T_2^*, this state will not decay because of the dephasing of the resonance atoms produced by the inhomogeneous broadening.

Here again we have the propagation of a single harmonic plus pump solitary wave but, owing to the considerable phase modulation, the true envelope of the third harmonic has both positive and negative segments. This is responsible for the complex structure of the modulus of the amplitude of this wave.

6. DISCUSSION

The main difference between the present result and those already published is that inhomogeneous broadening of the resonance line has been taken into account together with the high-frequency Stark effect. It was found that, if the spectral width of the pump pulse ($\sim 1/\tau_p$) exceeded the width of the inhomogeneously broadened line, the propagation of the ultrashort pulses of parametrically coupled waves was not very different from the corresponding process for $T_2^*=\infty$. However, in the opposite limiting case, the propagation of such waves is different from the propagation of ultrashort pulses in the absence of a spread in the resonance frequency of the radiators. The reason for this is that the two-level system loses coherence in a time $(\tau_p^{-1}+\Delta\omega_S)^{-1}$, which is less than the length τ_p of the radiation pulse. Numerical solution of (2a)–(2b) for zero Stark shift and $\tau_p \gg T_2^*$ has confirmed this mechanism. The pulse amplitude and the effective number of excited atoms were found to decrease for a given set of common pulse characteristics.

Another feature of the propagation of ultrashort pulses during third-harmonic generation is the parametric transmission effect. The corresponding solitary waves, named 0π pulses,[18] are specific for PRP. As they move through the resonance medium, the effective field vector has no transverse components ($R_1=R_2=0$) and the Bloch vector remains fixed, executing a "rotation" through zero angle. Figure 3 illustrates the evolution of parametric transmission during third-harmonic generation. In this figure, $\tau_p/T_2^*=0.2$, the "energy" of the incident pulse is $\Psi_0=2.66\pi$, the coefficient governing the combin-

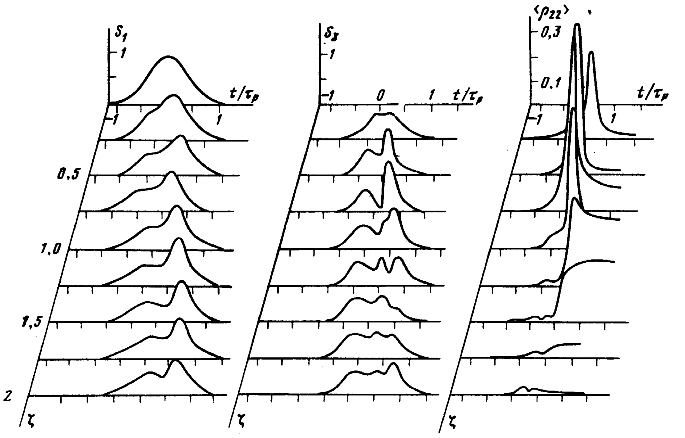

FIG. 3. Evolution of parametric transmission during third-harmonic generation.

ational coupling with respect to two-photon absorption is $\beta = 1.2$, and all the other parameters are the same as in Fig. 1. It is important to note that the harmonic pulse does not repeat the shape of the pump pulse, as expected from the "generalized energy theorem," obtained in the absence of phase modulation.

The foregoing results of the numerical analysis of the THG process in the field of an ultrashort pump pulse under the conditions of two-photon resonance suggest that the above equations form a new class of wave equations with solutions in the form of solitary waves. As in the case of single-photon resonance, several questions have to be examined, namely, possible types of pulse solutions, the existence among them of solitons, i.e., pulses retaining stability after collisions with one another, solutions in the form of an infinite sequence of pulses, the effects of the propagation of parametrically coupled waves in an amplifying medium, and so on. Phase modulation can lead to the formation of exceedingly narrow peaks on the pulse envelope, and it may be necessary to go beyond the framework of the approximation of slowly-varying complex amplitudes. Numerical studies of the equations describing third-harmonic generation and other parametric resonance processes will undoubtedly facilitate such investigations. Among analytic methods, we note the well-tested method used for the converse scattering problem[28] which has recently been applied to the Raman scattering problem.[29]

The authors are greatly indebted to S. O. Elyutin for many discussions relating to the foregoing problems.

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Translated by S. Chomet