High-order harmonic generation by an intense laser pulse in coherent Raman transitions

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(Submitted 27 January 1995)
Zh. Éksp. Teor. Fiz. **108**, 390–403 (August 1995)

We present the results of an analytical solution of the nonlinear equations for the propagation of an ultrashort intense laser pulse through a medium described by the density matrix for two-level systems with coherent stimulated Raman self-scattering. The pulse is assumed to be much shorter than the oscillation period in the Raman transition and the relaxation times. At certain intensities of the pulse high-order harmonic generation becomes possible. We express the spectrum of such a pulse in terms of integrals of the initial field strength. We also study the dynamics of the harmonic spectrum as a function of the pulse intensity with allowance for propagation effects. Finally, we estimate the efficiency for conversion of the initial radiation into high-order harmonics. © *1995 American Institute of Physics.*

1. INTRODUCTION

The recent rapid development of laser systems capable of producing pulses of a fairly high intensity in the femtosecond range[1,2] has made it possible to study new nonlinear processes in the interaction of electromagnetic radiation with matter. One manifestation of such interaction of interest from the scientific and application viewpoints is the generation of high-order harmonics. Here the experimental spectra of the harmonics are in sharp contrast with the results obtained by perturbation-theory techniques.[3,4] Hence in this field of research there is now great interest in obtaining exact solutions for different models of the interaction of the electromagnetic field with matter.

Generation of odd harmonics up to the eleventh is observed even at low intensities ($\sim 10^{11}$ W cm^{-2}; see Ref. 5). The transition to subpicosecond pulses made it possible to substantially raise the intensity at which gas breakdown occurs and to observe the generation of higher-order harmonics: the 31st,[6] the 109th,[7], the 111th,[8] and the 143rd[2]. A detailed bibliography of the experimental work in this field is contained in Refs. 9 and 10.

A possible theoretical interpretation of high-order harmonic generation in such experiments is usually linked to above-threshold ionization of atoms caused by multiphoton transitions and the subsequent one-photon transition to the ground state or an adjacent state, which leads to emission of a hard photon whose energy is approximately the sum of the energies of the n absorbed photons.[11–19] These studies usually use the monatomic approximation (this approach can be called nonlinear Thomson scattering on a bound electron), i.e., do not allow for coherent multiparticle response of the medium and also use propagation effects; the latter, as noted in Refs. 20 and 21, lead to the strongest nonlinear effects when intense laser pulses interact with matter.

At the same time it is known that an effective mechanism of nonlinear spectrum conversion is stimulated Raman scattering (SRS), which may involve the vibrational states[20,22,23] and the purely electronic states.[23–25] An ultrashort pulse satisfying the condition $\tau_p < 1/\Omega$, where τ_p is the pulse length, and $\hbar\Omega = \epsilon_2 - \epsilon_1$ is the energy difference of the levels participating in SRS, propagates in the medium under the condition of so-called combination resonance. If the above condition is met, the pulse contains an infinite number of frequency components comparable in amplitude and satisfying the combination resonance condition, which leads to effective stimulated Raman self-scattering (SRSS) of these components.[1] And since the relaxation times of the oscillators participating in Raman scattering can be considerably longer than τ_p, the scattering processes occur in a coherent mode. In this case several phenomena may occur: 2π-pulses of self-induced transparency,[22] spectral supercontinuum lasing,[27,28] optical "rectification,"[26] and, at certain threshold intensities, efficient generation of high-order harmonics.[29]

We studied harmonic generation by numerically solving the system of nonlinear partial differential equations describing processes in SRSS conditions. However, if $\tau_p \ll 1/\Omega$, T_1, T_2, where T_1 and T_2 are the times of longitudinal and transverse relaxation, respectively, these equations can be solved analytically. The analytical approach makes it possible not only to check the numerical results but also to study the emission spectra of the harmonics in any frequency range, which is generally impossible in the numerical approach because of restrictions imposed on the accuracy of calculations and the finiteness of the calculation time. In this paper we examine a theoretical emission spectrum generated by an ultrashort pulse of laser radiation with an arbitrary envelope in the passage through a Raman-active medium under the conditions specified earlier.

2. THE BASIC EQUATIONS

We investigate the one-dimensional problem of the propagation of a pulse through a medium in SRS conditions. The electric field of the pulse is polarized along the x axis, and the wave propagates parallel to the z axis. The dynamics of the field is described by the nonlinear wave equation

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$$\frac{\partial^2 E}{\partial z^2}-\frac{1}{c^2}\frac{\partial^2 E}{\partial t^2}=\frac{4\pi}{c}\frac{\partial^2 P}{\partial t^2} \tag{1}$$

and the material equations (the model of two-level systems[20–23,30])

$$\frac{\partial^2 Q}{\partial t^2}+\frac{1}{T_2}\frac{\partial Q}{\partial t}+\Omega^2 Q=-\frac{1}{2m}\alpha_q E^2\rho, \tag{2}$$

$$\frac{\partial \rho}{\partial t}+\frac{\rho-\rho_0}{T_1}=\frac{\alpha_q}{\hbar\Omega}E^2\frac{\partial Q}{\partial t}, \tag{3}$$

where E is the electric field strength in the wave, c is the speed of light, Q is the normal coordinate of an oscillator, $\Omega=(\epsilon_2-\epsilon_1)/\hbar$ is the oscillator transition frequency, T_1 and T_2 are the phenomenological longitudinal and transverse relaxation times, $\rho=n_2-n_1$ is the normalized difference in level populations ($\rho=-1$ corresponds to the situation where all the oscillators are on the lower level), $\alpha_q=(\partial\alpha/\partial Q)_{Q=0}$ is the derivative of the polarizability with respect to the normal coordinate at zero, $P=P_l+P_n$ is the total polarization of the medium consisting of the linear (P_l) and nonlinear ($P_n=N\alpha_q QE$) parts, N is the oscillator number density, and m is the reduced mass.

To obtain an analytical solution and calculate the spectra, we examine the case of purely coherent scattering,[26] i.e., when the pulse length τ_p is much shorter than both relaxation times and the vibration oscillator period. In this case the above system of equations reduces to a single nonlinear equation,[28]

$$\frac{\partial E}{\partial z}+\frac{1}{v}\frac{\partial E}{\partial t}=\beta\frac{\partial}{\partial t}\left[E\int_{-\infty}^{t}\sin\psi(z,t')dt'\right], \tag{4}$$

where

$$\psi(z,t)=\gamma\int_{-\infty}^{t}|E(z,t')|^2dt',$$

$$\gamma=\frac{\alpha_q}{\sqrt{2\hbar\Omega m}},\quad \beta=2\pi N\rho_0\frac{v}{c}\alpha_q\sqrt{\frac{\hbar\Omega}{2mc^2}},$$

and v is the velocity due to the linear part of the polarization. The quantity β^{-1} has the dimensions of length and can be considered the Raman scattering length. As in Ref. 26, we assume that the wave propagating in the direction opposite to the direction of propagation of the primary laser radiation is considerably smaller in amplitude than the one propagating in the direction of the primary radiation. We paid special attention to this problem when studying the equations numerically with the direct and reverse waves taken into account. We found that at $\psi(z,\infty)=2\pi l$ ($l=1,2,3,\cdots$) the energy of the reverse wave is lower than that of the direct wave by five to six orders of magnitude.

The initial conditions here are as follows: the medium is in the region $z>0$ and initially is not perturbed (the latter is taken into account in deriving (4)). At $z=0$ the input pulse is fixed, $E(t,z=0)=E_0(t)$, and, generally, is of an arbitrary shape, the only restriction being that $E_0(t)\to 0$ as $t\to\pm\infty$.

3. SOLUTION OF THE EQUATIONS AND THE PULSE SPECTRA

For the independent variables we take $\zeta=\beta\times(k_0z-\omega_0t)$ and $\tau=\omega_0t$ (ω_0 is the carrier frequency of the laser pulse and k_0 is the wave vector; in the new variables $\omega_0=1$). We then return to the old notation in the equations, which should not lead to any misunderstanding. In the new variables Eq. (4) has the following form:

$$\frac{\partial E}{\partial z}=\frac{\partial E}{\partial t}\int_{-\infty}^{t}\sin\psi(t',z)dt'+E\sin\psi(t,z). \tag{5}$$

Equation (5) clearly shows that $\psi(t,z)$ satisfies a closed equation (only ψ is present in the equation). To derive this equation it is sufficient to multiply Eq. (5) by E and integrate both sides with respect to time from $-\infty$ to t using the definition of ψ:

$$\frac{\partial\psi}{\partial z}=\frac{\partial\psi}{\partial t}\int_{-\infty}^{t}\sin\psi(t',z)dt'+[1-\cos\psi(t,z)]. \tag{6}$$

Let us examine the properties of $\psi(t,z)$ as a function of t in greater detail.

1. For all values of z the function $\psi(t,z)$ is a nondecreasing function of t.

2. Since $E(t,z)$ is an oscillating function of t, the time derivative of ψ vanishes at the zeros (t_{0i}) of the field $E(t,z)$. However, the second time derivative of ψ also vanishes at these points, i.e., in the neighborhood of such a point the function ψ can be written as

$$\psi(t,z)=\psi(t_{0i})+a(t-t_{0i})^3.$$

3. Items 1 and 2 imply that ψ has an inverse function $t=t(\psi,z)$ for every value of z in the interval from $-\infty$ to $+\infty$, since between the zeros the function ψ monotonically increases and hence has a unique inverse, while in the neighborhood of a zero it has a unique inverse in the real domain in view of item 2.

This behavior of ψ makes it possible to go over to a set of new variables in Eq. (5),

$$\psi=\psi(t,z),\quad t=t(\psi,\zeta),\quad \zeta=z,\quad z=\zeta.$$

After this we can write Eq. (5) as follows:

$$\frac{\partial E}{\partial\psi}\frac{\partial\psi}{\partial z}+\frac{\partial E}{\partial z}=\frac{\partial E}{\partial\psi}\frac{\partial\psi}{\partial t}\int_{-\infty}^{t}\sin\psi(z,t')dt'$$

$$+E\sin\psi(z,t). \tag{7}$$

Combining this with Eq. (6), we get

$$\frac{\partial E}{\partial\psi}(1-\cos\psi)+\frac{\partial E}{\partial z}=E\sin\psi(z,t). \tag{8}$$

Thus, the nonlinear equation (5) has been reduced to a linear equation thanks to the above transformations. The solution of the new equation can be found in the standard way[31] and has the form

$$E(\psi,z)=(1-\cos\psi)\phi(\cot(\psi/2)+z), \tag{9}$$

where ϕ is an arbitrary function.

The form of this function can be found from the conditions at $z=0$. Then

$$E(\psi,z)=\frac{1+(u+z)^2}{1+u^2}E_0[t_0(u+z)], \tag{10}$$

where $u=\cot(\psi/2)$, and t_0 is the inverse of

$$\psi_0=\int_{-\infty}^{t_0}E_0^2(\tau)\,d\tau.$$

To determine E in the temporal region we must obtain the function $\psi(t,z)$ defined by Eq. (6). In solving this equation we again take ψ for the independent variable and t for the dependent variable, which can always be done in view of the properties of $\psi(t,z)$ discussed above. Then, if we knew $\psi(t,z)$, we would have the identity $t=t(\psi\,(t,z),z)$, which could be differentiated and hence

$$\frac{\partial t}{\partial\psi}\frac{\partial\psi}{\partial t}=1,\quad \frac{\partial t}{\partial z}+\frac{\partial t}{\partial\psi}\frac{\partial\psi}{\partial z}=0. \tag{11}$$

Using these equalities and the initial condition that $\psi(t,z)\to 0$ as $t\to-\infty$, we obtain

$$-\frac{\partial t}{\partial z}=\int_0^{\psi}\sin(\psi')\frac{\partial t}{\partial\psi'}\,d\psi'+(1-\cos\psi)\frac{\partial t}{\partial\psi}. \tag{12}$$

Finding the ψ-derivative of both sides of this equation and introducing the notation

$$f=\frac{\partial t}{\partial\psi},$$

we arrive at the linear equation

$$-\frac{\partial f}{\partial z}=2\sin\psi f+(1-\cos\psi)\frac{\partial f}{\partial\psi}. \tag{13}$$

The solution of this equation is sought in the same form as in the previous case, and it has the form

$$f=\frac{1}{(1-\cos\psi)^2}\phi_1[\cot(\psi/2)+z], \tag{14}$$

where ϕ_1 is an arbitrary function determined from the conditions at $z=0$. We then have

$$f=\frac{4}{(1-\cos\psi)^2}\,\frac{1}{(1+q(\cot(\psi/2)+z)^2)^2}\times\frac{\partial t_0(\psi_0)}{\partial\psi_0}\bigg|_{\psi_0=2\cot^{-1}(\cot(\psi/2)+z)}. \tag{15}$$

Combining this with the definition of f, we obtain

$$t=\int_0^{\psi}\frac{[1+\cot^2(\psi'/2)]}{(1+q(\cot(\psi/2)+z)^2)^2}\,\frac{\partial t_0(\psi_0)}{\partial\psi_0}\bigg|_{\psi_0=2\cot^{-1}(\cot(\psi'/2)+z)}\times d\psi'-t(0). \tag{16}$$

The expressions (10) and (16) determine the solution of Eq. (5) in parametric form. Without specifying the shape of the initial pulse it is impossible to obtain an explicit expression for the solution of Eq. (5) as a function of t and z. More than that, for the majority of realistic cases even knowing the shape of the initial pulse does not allow the function $t_0(\psi_0)$ to be found explicitly.

However, the complex-valued Fourier spectrum of the signal can be expressed in terms of an integral of the initial field $E_0(\tau)$.

By definition,

$$E(\omega,z)=\int_{-\infty}^{\infty}\exp(-i\omega t)E(t,z)\,dt.$$

Substituting into this equation the values of E and t from Eqs. (10) and (16) and performing time-consuming but otherwise simple transformations of the integrand involving transformations to other integration variables, we obtain

$$E(\omega,z)=\int_{-\infty}^{\infty}\exp(-i\omega t)E_0(\tau)\,d\tau, \tag{17}$$

where

$$t=\tau-z\int_{-\infty}^{\tau}\sin\left[\gamma\int_{-\infty}^{\tau'}E_0^2(\tau'')\,d\tau''\right]d\tau'+(z^2/2)\int_{-\infty}^{\tau}\left\{1-\cos\left[\gamma\int_{-\infty}^{\tau'}E_0^2(\tau'')\,d\tau''\right]\right\}d\tau'. \tag{18}$$

If we take the case $z\ll 1$, then to within a linear term we have

$$E(\omega,z)=\int_{-\infty}^{\infty}\exp(-i\omega\tau)E_0(\tau)\times\left\{1-i\omega z\int_{-\infty}^{\tau}\sin\left[\gamma\int_{-\infty}^{\tau'}E_0^2(\tau'')d\tau''\right]d\tau'\right\}d\tau, \tag{19}$$

which coincides with the spectrum of the solution obtained if Eq. (5) is solved by the method of successive approximations. Thus, Eqs. (17) and (18) provide a clear picture of the spectrum of the solution in terms of integrals of the initial pulse. It appears, however, that for realistic shapes of the initial signal these integrals cannot be expressed analytically, while numerically they can easily be calculated.

4. ANALYTICAL SPECTRA IN THE SIMPLEST CASES

Case 1. The initial pulse is specified on the interval $(-t_m,t_m)$ by the function

$$E_0(\tau)=-A_0\,\mathrm{sign}\,(\tau),$$

where $2t_m$ is the length of the pulse, and A_0 is the pulse amplitude. In this case all the integrals can be evaluated explicitly and the complex-valued spectrum of the signal is

$$E(\omega,z)=-2A_0\sum J_n(\omega\xi)\exp[in(\omega_2 t_m+\varphi-\pi/2)]\times\frac{1-\exp(i\psi_n)}{in\omega_2-i\omega(1+z^2/2)}, \tag{20}$$

where $\omega_2=\gamma A_0^2$, $\xi=z\sqrt{1+z^2/4}/\omega_2$, $J_n(x)$ is the Bessel function, and

$$\sin\varphi=\frac{z/2}{\sqrt{1+(z/2)^2}},\qquad \psi_n=[\omega(1+z^2/2)-n\omega_2]t_m.$$

The above formula shows that the spectrum of the pulse is continuous with peaks lying near the zeros of the denominator; the frequency of such a peak is

$$\omega_n=\frac{n\omega_2}{(1+z^2/2)}. \tag{21}$$

Thus, the spectrum contains quasiharmonics of frequency ω_2 that shift toward the red region as the signal moves deeper into the medium.

Case 2. Now we take a more realistic initial pulse, specified on the same interval by the function

$$E(\tau)=A_0\sin\tau. \tag{22}$$

The complex-valued spectrum of the signal can be expressed as

$$E(\omega,z)=A_0\int_{-t_m}^{t_m}\exp\{-i\omega[\tau(1+z^2/2)-z\sqrt{1}+(z/2)^2\psi_\alpha]\}\sin\tau\,d\tau, \tag{23}$$

where

$$\psi_\alpha=\int_{-t_m}^{\tau}\sin[\omega_2(\tau'-\sin(2\tau')/2)+\varphi_1]d\tau',$$

$$\omega_2=\gamma A_0^2/2\ ,\quad \varphi_1=\omega_2 t_m+\varphi,\quad \sin\varphi=\frac{z/2}{\sqrt{1+(z/2)^2}}.$$

For

$$\psi_0=\gamma\int_{-t_m}^{t_m}E_0^2(\tau)\,d\tau=2\pi k,$$

with k an integer, and if s periods of the field oscillations fit into one pulse, then $\omega_2=k/s$.

The function ψ_α can be calculated via the standard series expansion[32]

$$\exp(iz\sin\Theta)=\sum_{n=-\infty}^{\infty}J_n(z)\exp(in\Theta) \tag{24}$$

and is specified by the following expression:

$$\psi_\alpha=\sum_{n=-\infty}^{\infty}\frac{J_n(\omega_2/2)}{2n-\omega_2}\cos(2n\tau-\omega_2\tau-\varphi_1). \tag{25}$$

To illustrate the theoretical spectrum we limit ourselves to the case of $\omega\leqslant 10$, $z\leqslant 0.5$, and $\omega_2\leqslant 1$. Then the complex-valued spectrum of the system is

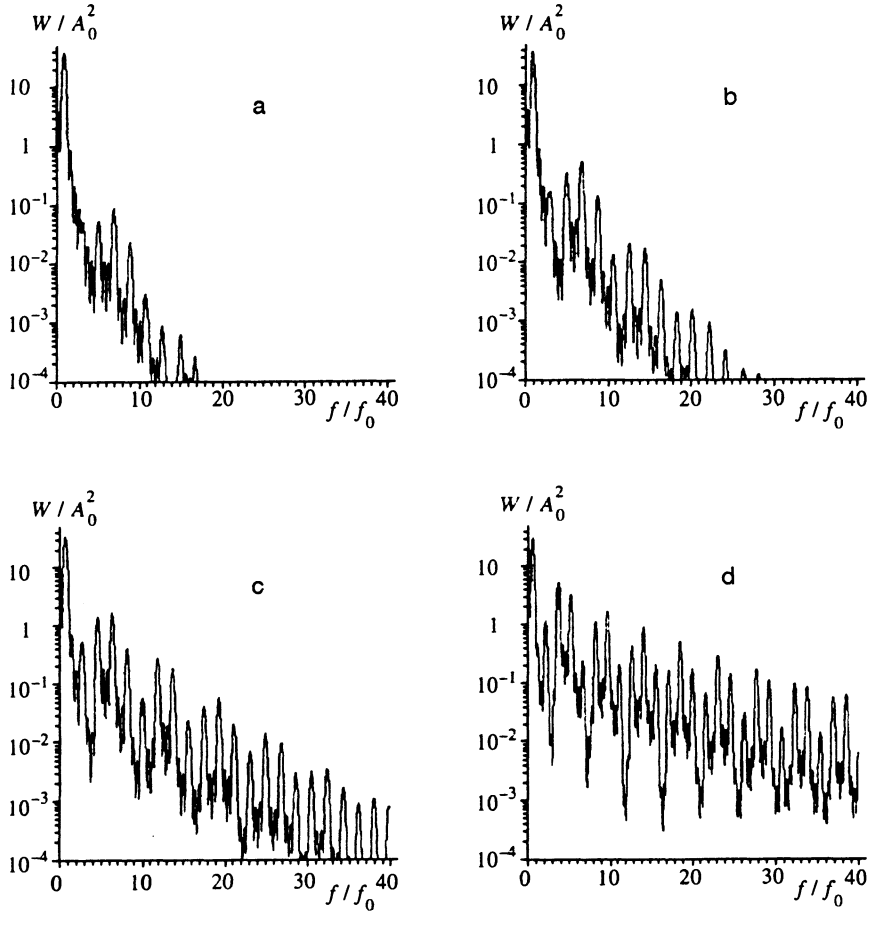

FIG. 1. Dynamics of the pulse power spectrum (a rectangular envelope) as a function of the ratio of the frequency to the frequency of the initial pulse in the passage of the pulse through the Raman-active medium. Here W is the power of a harmonic, A_0 is the amplitude of the initial signal, $\tau_p=2$, $k=8$, (a) $z=0.1$, (b) $z=0.25$, (c) $z=0.5$, (d) $z=1.0$.

$$E(\omega,z)=(A_0/i)\sum_{m,k,l=-\infty}^{\infty} J_m(\zeta_0)J_k(\zeta_+)J_l(\zeta_-)$$
$$\times\exp(-iM\varphi_2)R_{LM}, \qquad (26)$$

where

$$L=k-l, \qquad M=m+k+l, \qquad \varphi_2=\varphi_1-\pi/2,$$
$$\zeta_0=-\omega t_z J_0(\omega_2/2)/\omega_2, \qquad \zeta_+=\omega t_z J_1(\omega_2/2)/(2-\omega_2),$$
$$\zeta_-=\omega t_z J_1(\omega_2/2)/(2+\omega_2), \qquad t_z=z\sqrt{1+(z/2)^2},$$
$$R_{LM}=\frac{\sin[(\omega(1+z^2/2)-M\omega_2+1)t_m]}{2L-M\omega_2+1+\omega(1+z^2/2)}$$
$$-\frac{\sin[(\omega(1+z^2/2)-M\omega_2-1)t_m]}{2L-M\omega_2-1+\omega(1+z^2/2)}. \qquad (27)$$

Thus, we again have a quasiharmonic spectrum whose peaks lie close to the zeros of the denominators; the frequencies of the quasiharmonics are

$$\omega_{LM}=\frac{2L-M\omega_2\pm1}{1+z^2/2}. \qquad (28)$$

In this case the frequencies of the quasiharmonics are labeled by two integers L and M, where L refers to the carrier frequency of the laser radiation and M to the frequency $\omega_2=k/s=\psi_0/\tau_p$, where $\psi_0=\gamma\int_{-\infty}^{\infty}E_0^2(\tau)\,d\tau$. The characteristic feature of this case, as in the previous case, is the shift of the quasiharmonic frequencies toward the red region as the signal moves deeper into the medium.

5. NUMERICAL ESTIMATES OF THE SPECTRA

Using Eqs. (17) and (19), we can easily calculate the laser pulse spectrum for any distance and any shape of the pulse as the pulse travels through the nonlinear Raman-active medium. The results of such calculations are presented in diagrammatic form for different values of z and $k=\gamma\int_{-\infty}^{\infty}d\tau\,E_0^2(\tau)/2\pi$.

Here we do not give numerical estimates of the spectrum of the signal described by the equation $E(\tau)=\operatorname{sgn}\tau$ because the structure of the spectrum is clear from the exact analytical expression (20). We limit ourselves to the spectra of pulses whose envelopes are rectangular and Gaussian.

1. *Rectangular envelope.* The shape of the signal is specified by the equation $E_0=A_0\sin\tau$. The results of spectrum calculations are shown in Fig. 1 for $\tau_p=2$ and $k=8$ at $z=0.1$, 0.25, 0.5, and 1.0. The peaks in the spectrum usually correspond to the odd harmonics of the carrier frequency.

An analysis of the other cases shows that as k grows the harmonic spectrum acquires a plateau with small maxima; the plateau shifts to higher frequencies as k grows. The high-harmonic generation efficiency (the ratio of the power at the peak of the harmonic to that in the first harmonic) increases with k. At $z=1.0$ and $k=16$ the efficiency of conversion into the first harmonic is $\eta=10^{-3}$.

2. *Gaussian envelope.* More interesting from the practical viewpoint is the case of a pulse with a Gaussian envelope,

$$E_0(\tau)=A_0\exp\{-(\tau/\tau_p)^2\}\sin\tau$$

(in what follows τ_p is expressed in fractions of the period of the laser carrier frequency).

Figure 2 displays the dynamics of the spectrum as a function of z for $\tau_p=2$ and $k=8$ at $z=0.1$, 0.25, 0.5, and 1.0. The quasiharmonics are clearly visible in the spectrum. The efficiency of conversion into the 31st harmonic is 10^{-6} at $z=0.25$, 10^{-4} at $z=0.5$, and 2.0×10^{-3} at $z=1.0$. Clearly, as the pulse travels through the Raman-active medium, the pulse gets "enriched" with high-order harmonics.

The dynamics of the spectrum of a Gaussian pulse at fixed $z=0.5$ and $\tau_p=2$ and different energies k of the input pulse is demonstrated by Fig. 3. For $k=1$ the spectrum resembles a supercontinuum,[27] and effective generation of harmonics begins as k grows, with the conversion efficiency increasing with k.

An analysis of the results of numerous calculations demonstrates the possibility of generating high-order harmonics by the above mechanism with a fairly high efficiency. For instance, harmonics close to the 30th can be generated with an efficiency of 10^{-2} to 10^{-5}, depending on the length and intensity of the pulse and on z.

The general structure of the spectrum for high harmonics is shown in Fig. 4 (at $k=8$ and $\tau_p=2$). The spectrum clearly exhibits a band nature, with each band consisting of discrete quasiharmonics differing little in amplitude. An estimate of the conversion efficiency of this emission mechanism in the 100–200 Å wavelength range shows that for the harmonics of the frequency of a Ti:sapphire laser it is 5.0×10^{-6}–5.0×10^{-5}, which is of the same order of magnitude or higher than the conversion efficiency of an x-ray laser.[33] For laser radiation in the water "window" ($\sim$40 Å) with the same pump laser the conversion efficiency is 10^{-8}–10^{-7}, and the mechanism can be used for practical generation of pulses of coherent radiation in this wavelength range.

6. CONCLUSION

We have obtained an analytical solution of the problem of propagation of a short intense laser pulse through a Raman- active medium on the assumption that the medium is described by the model of two-level systems and that the pulse length is much shorter than the oscillation period and the transverse and longitudinal relaxation times. On the basis of this solution the spectrum of the pulse is expressed in terms of integrals of the electric field of the initial pulse. The representation obtained makes it possible to analyze the general structure of the spectrum. Generally, the spectrum contains many peaks, which can be interpreted as the contribution of the quasiharmonics present in the pulse that propagates through the medium; the energy of the quasiharmonics depends both on the properties of the medium and on the length, shape, and intensity of the pulse. At low intensities ($\psi_0=2\pi$) the spectrum resembles a supercontinuum, while at higher intensities peaks corresponding to quasiharmonics emerge, and the height of these peaks grows both in absolute value (the conversion efficiency increases) and relative to the continuous background. The quasiharmonic fre-

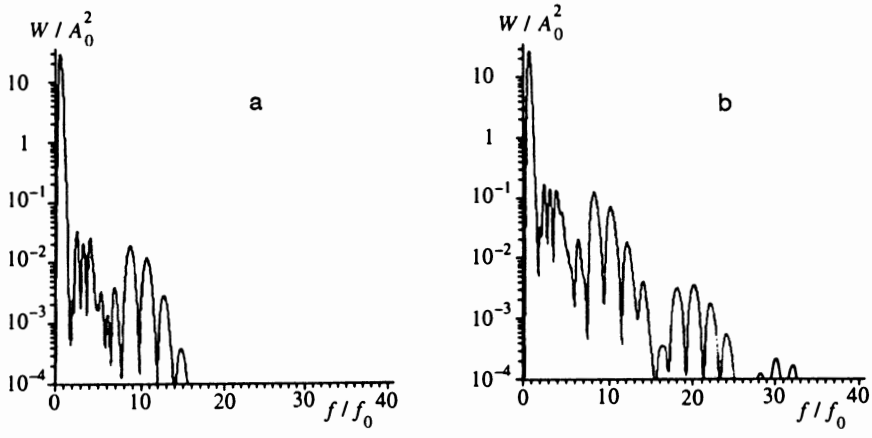

FIG. 2. As in Fig. 1, but with a pulse having a Gaussian envelope.

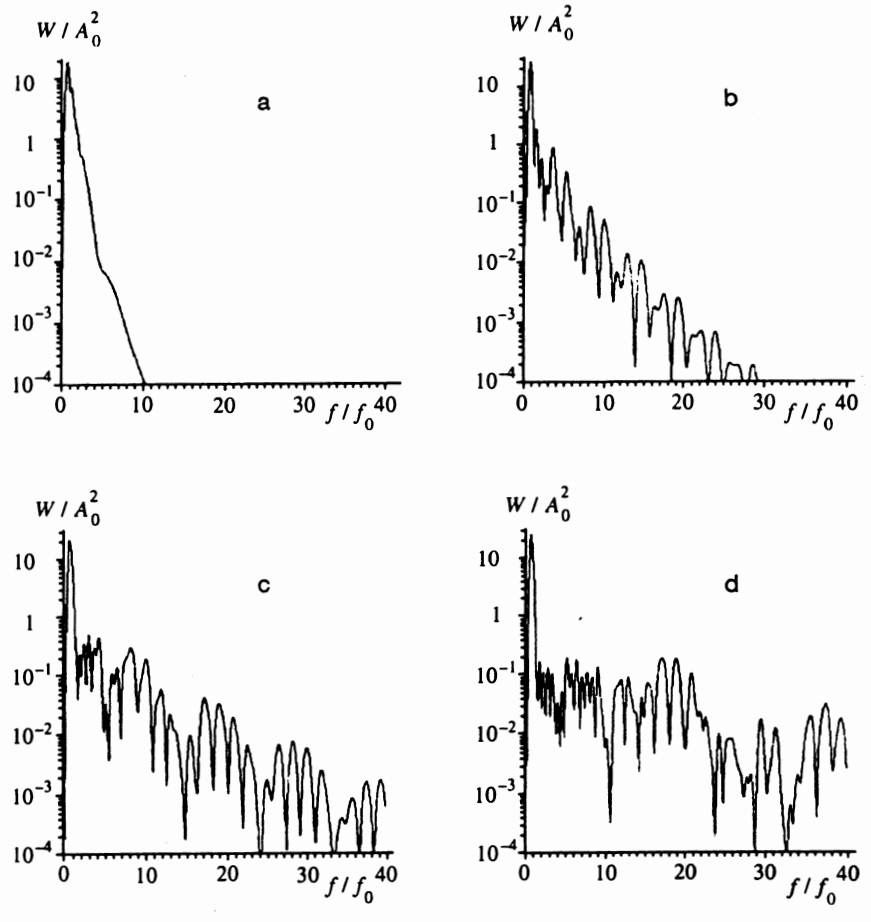

FIG. 3. The power spectrum as a function of the intensity of the initial pulse ((a) $k=1$, (b) $k=4$, (c) $k=8$, and (d) $k=12$) for a pulse with a Gaussian envelope with $\tau_p=2$ at $z=0.5$.

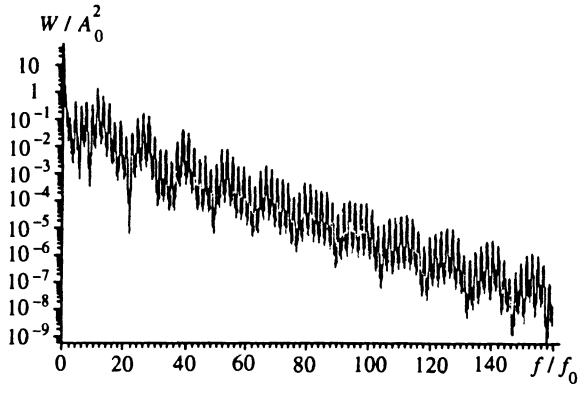

FIG. 4. The spectrum structure over a broad frequency range for a pulse with a Gaussian envelope: $k = 8$ and $z = 0.5$.

quencies are specified by two integers (see Eq. (28)), one of which refers to the carrier frequency of the initial pulse and the other to a certain effective frequency proportional to the ratio of the total energy of the initial pulse to the pulse length. Although the latter was demonstrated for the case of a pulse with a rectangular envelope, one can expect that this is true for any short pulse. The height and width of the peaks in the spectrum change as the pulse propagates through the medium. For short paths, when the conversion efficiency is still moderate, odd harmonics of the carrier frequency are usually observed.

The analytical solution enables estimating the behavior of the spectrum over a broad frequency range, which is difficult or even impossible to do numerically. We have shown that the spectrum has a band nature, i.e., the heights of the peaks are modulated in magnitude (see Fig. 4). Studies of the spectra for different values of the parameter k, which characterizes the pulse energy, and for different pulse lengths allow us to estimate the efficiency of conversion of the initial radiation into various spectral ranges; for one thing, the emission of a Ti:sapphire laser can be converted into the 100–200 Å wavelength range with an efficiency of 10^{-6}–10^{-5}, which is of the same order of magnitude as in X-ray lasers in the same range. This means that the mechanism considered here is unquestionably of interest for generating short pulses of coherent soft X-ray radiation.

Let us estimate the possible values of the parameters γ and β for several realistic media. For instance, in benzene at the transition frequency ($\Omega/2\pi c = 992\ \mathrm{cm}^{-1}$), for the differential forward SRS cross section ($\sigma = (\omega_0/c)^4(\hbar/2m\Omega)\alpha_q^2$) we have $\sigma = 3\times10^{-29}$ cm^2, and $1/T_2\pi c = 2\ \mathrm{cm}^{-1}$ (see Ref. 30). Here for pulses with $\tau_p = 10$ fs the conditions $\tau_p < \Omega^{-1}, T_1, T_2$ are met, and for $N \sim 10^{18}\ \mathrm{cm}^{-3}$ and a pulse intensity $I_0 \sim 10^{13}\ \mathrm{W\ cm}^{-2}$ we have $\beta \sim 1\ \mathrm{cm}^{-1}$ and $\psi_0 \sim 2\pi$. Clearly, higher intensities and densities make it possible to shift to the range of parameters discussed in this paper. Note, however, that benzene is not quite suitable in representing the two-level system considered here because its spectrum has a multilevel nature, which makes it impossible to limit ourselves to the two-level approximation at high intensities of the initial pulse.

The model can be applied with greater success to ions of inert gases. For instance, in the Ne^+ the two states in the structure of the ground level ($^2P_{1/2}$ and $^2P_{3/2}$) form a two-level system with a transition with $\Omega/2\pi c = 780.4\ \mathrm{cm}^{-1}$, which is parity-forbidden in the electrodipole approximation; all the other states are separated in frequency from those just mentioned by more than $2\times10^5\ \mathrm{cm}^{-1}$. A similar pattern is observed in Ar^+, Kr^+, and Xe^+ ions.[34] We also note that in experiments with multiphoton ionization of Kr it was shown that laser pulses with an initial intensity of roughly $10^{13}\ \mathrm{W\ cm}^{-2}$ can effectively alter the populations of the states $^2P_{1/2}$ and $^2P_{3/2}$ in Kr^+ (see Ref. 35). The situation is approximately the same with halogen atoms. Hence these gases can be used, for one thing, to study the generation of high-order harmonics by the mechanism elaborated in this paper.

This work was supported by Department of public Education of Republic of Belorussia and the International Science Foundation (Grant No. N9T000)

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Translated by Eugene Yankovsky