Propagation dynamics of high-power femtosecond pulses in Raman-active media

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We analyze the dynamics of a high-power ultrashort optical pulse in a Raman-active medium in the case when the pulse width is small in comparison with the time Ω^{-1}, where $\hbar\Omega = E_2 - E_1$ is the energy difference of levels participating in Raman scattering. The feasibility of new nonlinear effects is shown: the existence of $2\pi n$-pulses of self-induced transparency of a Raman-active medium, generation of videopulses of electromagnetic field without carrier frequency starting from a high-frequency signal. We show that under certain conditions the pulse propagation may be accompanied by its temporal compression and at sufficiently high powers by spectrum superbroadening as well. Attention is paid to qualitatively new physics of the processes when the field dynamics is determined by the variation of the photon energy rather than the variation of the photon number.

INTRODUCTION

Stimulated Raman scattering (SRS) of light is one of the fundamental mechanisms of radiation frequency conversion exhibited in practically all states of aggregation: solids, liquids, gases, and plasmas. General regularities of SRS were recognized even in the 60s.[1,2] By the late 70s, a considerable number of experimental and theoretical works had been performed, which had revealed special features of SRS in a field of short optical pulses. These features arise from such factors as finite time of a local response of medium, effect of group retardation, etc.[3–5]

The recent progress in methods for femtosecond optical pulse generation and amplification, production of pulses having widths of order of one oscillation period of electromagnetic wave gave rise to some qualitatively new features in many nonlinear optical processes.[6] The interpretation of new wave phenomena in terms of traditional nonlinear optics dealing with the concepts of amplitude, phase, and frequency of interacting waves often proves to be impossible. The point is not only that the wave processes on a time-space scale of several field oscillations cannot be regarded as quasimonochromatic. In sufficiently strong fields, the basic assumption of weakly nonlinear and strongly dispersive medium used in traditional nonlinear optics becomes inapplicable, and so, we cannot restrict ourselves to the consideration of a finite number of interacting field harmonics.[7]

All the aforesaid holds in full measure for femtosecond pulse propagation in Raman-active media as well. It was investigated experimentally in Refs. 8–10. Indeed, typical values of the time Ω^{-1} corresponding to the Stokes (anti-Stokes) frequency shift Ω in most Raman-active media are 10^{-13}–10^{-14} s. The condition

$$\tau_p \Omega \ll 1 \qquad (1)$$

implying a qualitatively new interaction regime is thus met for for an ultrashort pulse (USP) of width $\tau_p < 10^{-14}$ s. In a field of monochromatic waves ($\tau_p \Omega \gg 1$), the presence of a seeding at the Stokes frequency is necessary for the SRS process to develop. This is spontaneous noise as a rule. In the case under study, the pulse spectrum initially contains an infinite combination of frequency components comparable in amplitude and satisfying the condition of SRS resonance. Consequently, the Raman self-scattering process becomes possible, which proceeds at field intensities much lower than the threshold ones for SRS buildup starting from spontaneous noise.[10]

The question of USP dynamics in a Raman-active medium with the condition (1) satisfied is practically intractable within the framework of the traditional Fourier analysis manipulating with quasimonochromatic Stokes and anti-Stokes field components. The development of a new approach providing adequate description of the corresponding processes is thus required to interpret experimental data and to plan experiments on femtosecond pulse propagation. This is just the purpose of the present paper.

The problem of pulse evolution in a Raman-active medium is formulated in terms of real field and real medium polarization induced by the field. This description method makes it possible to reveal a qualitatively new nature of nonlinear optical processes in the case of ultrashort and ultraintense action on matter when the field dynamics is characterized by continuous variation of the photon energy. On the basis of the approach developed, the feasibility of high-power $2\pi n$-pulse propagation under the conditions of self-induced transparency is shown in Sec. 1 of the present work. Pulses of energy less than π experience preferentially Stokes scattering, and it is possible to follow the process of effective carrier frequency reduction to formation of a videopulse, i.e. a pulse without a carrier frequency.

In Sec. 2, the evolution of high-power ($\geqslant 2\pi$) pulse spectrum is analyzed. Strong interaction of an USP with a medium is shown to result in the spectrum superbroadening effect, i.e. formation of both low- and high-frequency spectral continuum.

The effect of linear dispersion of a medium on USP dynamics is considered in Sec. 3 of the present paper. USP spectrum rearrangement due to SRS is shown to be accom-

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panied by pulse compression in the presence of normal dispersion of a medium in certain conditions.

The range of applicability of the two-level model to the SRS description and special features of this process in multilevel molecular media are discussed in Sec. 4 of the present paper.

1. SELF-INDUCED TRANSPARENCY OF $2\pi n$-PULSES; GENERATION OF VIDEOPULSES FROM HIGH-FREQUENCY SIGNALS

For a correct description of the optical pulse dynamics in a Raman-active medium in the case when the pulse width satisfies the condition (1), let us turn to exact equations for electric field $\mathscr{E}$ of the pulse and polarization $\mathscr{P}$ of a medium induced by the pulse. The Raman-active medium will be described using the well-known Bloembergen–Shen model for a nonlinear oscillator:[11]

$$\frac{\partial^2 Q}{\partial t^2}+\frac{1}{T_2}\frac{\partial Q}{\partial t}+\Omega^2 Q=-\frac{1}{2M}\left(\frac{\partial\alpha}{\partial Q}\right)\mathscr{E}^2\rho \tag{1.1}$$

$$\frac{\partial\rho}{\partial t}+\frac{\rho-\rho_0}{T_1}=\frac{1}{\hbar\Omega}\left(\frac{\partial\alpha}{\partial Q}\right)\mathscr{E}^2\frac{\partial Q}{\partial t}, \tag{1.2}$$

and the evolution of an electromagnetic pulse propagating along the Oz-axis will be described by the wave equation

$$\frac{\partial^2\mathscr{E}}{\partial Z^2}-\frac{1}{C^2}\frac{\partial^2\mathscr{E}}{\partial t^2}=\frac{4\pi}{C^2}\frac{\partial^2\mathscr{P}}{\partial t^2}, \tag{1.3}$$

in which the medium polarization $\mathscr{P}$ in the general case contains contributions both linear $\mathscr{P}_{\mathrm{L}}$ and nonlinear $\mathscr{P}_{\mathrm{NL}}$ in the field. The latter results from SRS.

Material equations (1.1)–(1.2) describe a nonlinear oscillator with a natural frequency $\Omega=(E_2-E_1)/\hbar$, where $E_{1,2}$ are the energies of the levels involved in the scattering process, a normal coordinate Q, and phenomenological relaxation times T_1 and T_2. The oscillator is excited by a driving force proportional to the product of the field squared and the difference of the level populations $\rho=n_1-n_2$. The proportionality factor $\partial\alpha/\partial Q$ is written in the traditional form as a derivative of the oscillator polarizability $\alpha(Q)$ with respect to the normal coordinate Q near the equilibrium value $Q=Q_0$, and the parameter M means the oscillator reduced mass. In various physical situations, the equations presented can describe various processes, such as light scattering by optical phonons in solids, interaction of an USP with an individual two-level system with a transition forbidden in the electric-dipole approximation, and so on.[2]

The field-induced nonlinear polarization of the medium entering into the wave equation (1.3) is given by the relation

$$\mathscr{P}_{\mathrm{NL}}=N\left(\frac{\partial\alpha}{\partial Q}\right)Q\mathscr{E}, \tag{1.4}$$

where N is the particle number density. The cases $\rho_0=-1$ and $\rho_0=+1$ correspond to the interaction of field with particles in the ground state with an energy E_1 and with a medium of totally inverted particles with an energy E_2 respectively.

In the general case, the system of self-consistent equations (1.1)–(1.4) can be studied only by numerical methods. Nevertheless, in the limit of ultrashort pulse width (1) it admits of an analytical solution, which turns out to be useful for understanding the physics of the processes under consideration. First of all, the condition of ultrashort pulse width (1) suggests that the radiation–medium interaction is coherent, i.e. $\tau_p\ll T_1$, T_2 and is characterized by a largely time-dependent response of the medium to the field. Moreover, this condition means that the item $\Omega^2 Q$ in Eq. (1.2) may be neglected in comparison with $\partial^2 Q/\partial t^2$. With the mentioned simplification made, the material equations can be integrated for a field of any form. Namely, introducing the rotation angle of the material variables

$$\Psi(z,t)=\left|\frac{\partial\alpha}{\partial Q}\right|(2\hbar\Omega M)^{-1/2}\int_{-\infty}^{t}\mathscr{E}^2(z,t')dt' \tag{1.5}$$

we find from Eqs. (1.1)–(1.2):

$$\rho=\rho_c\cos\Psi,\quad \frac{\partial Q}{\partial t}=-\rho_0\,\mathrm{sign}\left(\frac{\partial\alpha}{\partial Q}\right)\left(\frac{\hbar\Omega}{2M}\right)^{1/2}\sin\Psi. \tag{1.6}$$

Considering further pulse propagation in the positive direction of the z-axis and assuming the contribution of the nonlinear polarization $\mathscr{P}_{\mathrm{NL}}$ to be small in comparison with the contribution of the linear polarization $\mathscr{P}_{\mathrm{L}}$, we may go from Eqs. (1.3) to the first-order equation

$$\frac{\partial\mathscr{E}}{\partial z}+\frac{1}{v}\frac{\partial\mathscr{E}}{\partial t}=-\frac{2\pi v}{c^2}\frac{\partial\mathscr{P}_{\mathrm{NL}}}{\partial t}, \tag{1.7}$$

where v is the pulse velocity determined by the linear contribution to the medium's polarization. Substituting (1.6) in the expression for the nonlinear polarization (1.4), we rewrite Eq. (1.7) in the form

$$\frac{\partial\mathscr{E}}{\partial z}+\frac{1}{v}\frac{\partial\mathscr{E}}{\partial t}=\beta\frac{\partial}{\partial t}\left(\mathscr{E}\int_{-\infty}^{t}\sin\Psi(z,t')dt'\right), \tag{1.8}$$

where we introduced the coefficient $\beta=2\pi N\rho_0|\partial\alpha/\partial Q|v/c(\hbar\Omega/2Mc^2)^{1/2}$. The quantity $|\beta|^{-1}$ of length dimension may be called the length of Raman self-scattering of a pulse.

From Eq. (1.8) an equation follows for the variation of the total pulse energy $W(z)=\Psi(z,\infty)$ as a function of z

$$\frac{dW}{dz}=\beta(1-\cos W). \tag{1.9}$$

The pulse energy variation law (1.9) is seen to be practically the same as in the case of coherent two-photon interaction of field with a resonant medium.[12] In the case of a noninverted medium ($\rho_0<0,\beta<0$), pulses of energy $W(0)=2\pi k$, $k=1, 2,\ldots$ do not experience absorption and propagate under the conditions of self-induced transparency. The result obtained is not unexpected, since both processes are two-quantum ones with the same selection rules. In the case of two-photon absorption, the interpretation of the

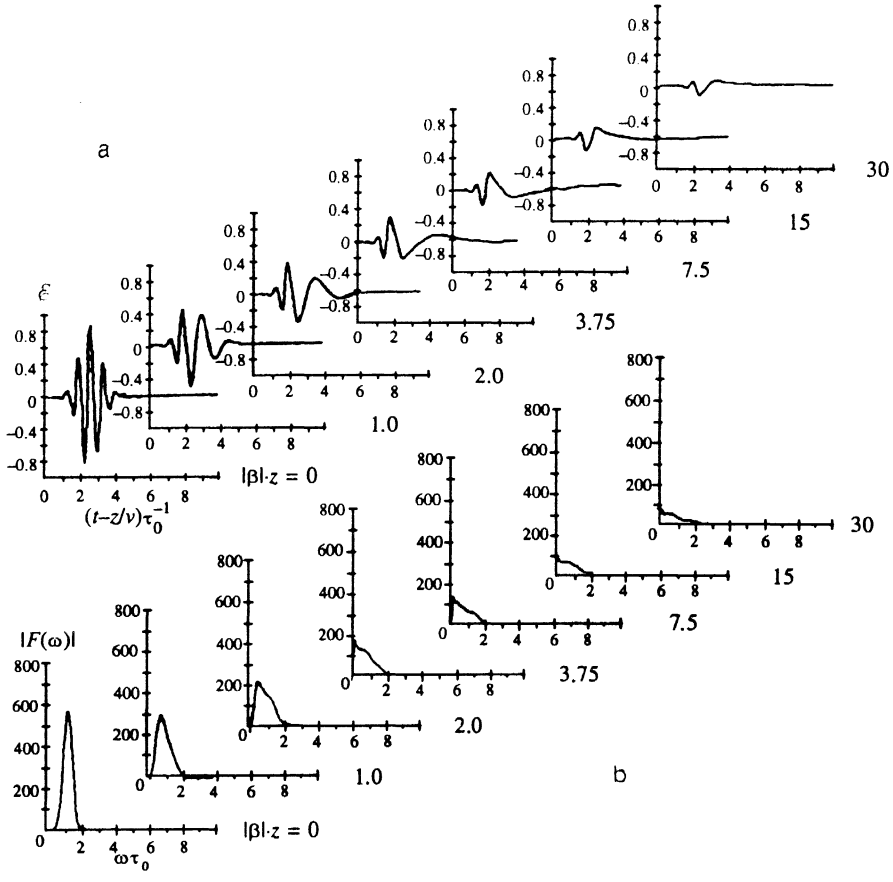

FIG. 1. Videopulse generation from a high-frequency signal in a Raman-active medium; evolution of electric field $\mathcal{E}(z,t)$ (a) and spectrum $F(\omega,z)$ (b) of a pulse. At the medium input the field has the form $\mathcal{E}(0,t)\sim\sin(\omega t)\exp\{-(t/\tau_0)\}^2$; $(\omega/2\pi)\tau_0=4$; $W(0)=\pi/2$; $\Omega\tau_0=0.1$. $\mathcal{E}$ and F are given in arbitrary units.

effect is in essence the same as in the McCall and Hahn model of the single-quantum effect. Raman scattering proceeds in accordance with a more complicated scheme. In a noninverted medium ($\beta<0$), the leading edge of a 2π-pulse is absorbed when Stokes field components are generated, while anti-Stokes components return the energy to the pulse "tail." As this takes place, the pulse itself is not stationary in shape and spectrum. Its trailing edge becomes enriched in high-frequency components, where the energy of the leading edge is transferred. The spectrum evolution of high-power ($W(0)\geqslant 2\pi$) pulses will be considered at greater length in Sec. 2. Here, we shall follow qualitatively the dynamics of 2π-pulse spectrum in the quasimonochromatic approximation valid at least at propagation lengths $z\ll|\beta|^{-1}$.

Representing the USP electric field in the form of a quasimonochromatic wave of amplitude $E(z,t)$ and frequency $\omega(z,t)=\omega+\dot{\varphi}(z,t)$, we can go from Eq. (1.8) to an equation for the "slow" phase φ of the wave. The self-phase modulation of the USP appearing in a Raman-active medium is then described by the expression

$$\dot{\varphi}(z,t)=\beta z\omega\sin\tilde{\Psi}(z,t), \tag{1.10}$$

where $\tilde{\Psi}$ is defined from (1.5) with the substitution $\mathcal{E}^2\to 1/2E^2$. From (1.10) it follows that in a noninverted medium ($\beta<0$) the leading part of the pulse, for which $\tilde{\Psi}(z,t)<\pi$, experiences a red frequency shift, while the trailing part, for which $\pi<\tilde{\Psi}(z,t)<2\pi$, shifts to blue.

Let us consider now at greater length the propagation of an USP of energy $W(0)<\pi$ for lengths $z\geqslant|\beta|^{-1}$ when its frequency variation is large and the process cannot be assumed to be quasimonochromatic. Owing to preferentially Stokes scattering, the effective frequency $\omega(z)$ of the pulse will continuously shift to red, and this process cannot stop until the USP energy is totally depleted because of excitation of oscillations in a medium. It is pertinent to compare this scattering mode with ordinary Stokes scattering of monochromatic field, in which the transfer is limited by the conditions of conservation of the total photon number. One might expect that with the USP frequency decreasing to $\omega(z)\leqslant\tau_p^{-1}$, the wavelength of the pulse formed becomes equal to its own spatial dimensions. What this would mean is the initial pulse of the visible range ($\omega(0)\tau_p\gg 1$) converts to a videopulse, i.e. a pulse without a carrier frequency.[1)] These qualitative reasonings are completely supported by the results of the numerical solution of Eqs. (1.1)–(1.4) presented in Fig. 1. The dynamics of videopulse formation is convenient to track by observing the evolution of the USP spectrum shown in Fig. 1b. The center of gravity of the spectrum is seen to shift to red, while the spectrum itself is broadened toward the long-wave wing down to zero frequencies.

The characteristic length of "rectification" of the initial USP to a videopulse can be estimated from formula

$$z_{\text{rect}}\sim\omega\tau_p[2\pi|\beta|W(0)]^{-1}, \tag{1.11}$$

It follows from (1.11) that the rectification length decreases with the initial pulse energy $W(0)$ increasing.

The analysis of USP propagation in a Raman-active medium presented in this Section is thus indicative of qualitatively new nature of nonlinear optical processes in the range of ultrashort pulsewidths ($\tau_p \ll \Omega^{-1}$) and superintense ($W(0) \sim 2\pi$) electromagnetic fields. The dynamics of a field interacting with a medium is characterized by variation of the photon energy rather than variation of the photon number in the field.

2. SUPERBROADENING OF THE HIGH-POWER USP SPECTRUM

In the present section, we shall consider the dynamics of SRS of high-power USPs in connection with the problem of superbroadening of subpicosecond pulse spectrum. It should be noted that after the discovery of spectral continuum generation[14,15] and its demonstration in various media including solids, liquids, and gases, the understanding of this phenomenon is still far from being complete. In particular, none of the theoretical models developed for spectrum superbroadening (see, for instance, Ref. 16) is universal enough to explain the regularities observed in such a variety of media.

At the same time, as early as in the paper by Smith, Liu, and Bloembergen[17] it was noted that many properties of a superbroadened spectrum cannot be explained without involving the SRS mechanism exhibited in the majority of materials. Indeed, along with scattering by optical phonons, the considered model of nonlinear medium (1.1)–(1.2) describes spin-flip SRS processes, SRS in multicomponent plasmas, scattering by coupled electron-phonon excitations, SRS in amorphous semiconductors, liquids, glasses, Brillouin scattering (as a particular case of scattering by acoustic phonons) in one or other approximation. Moreover, the model is applicable to the description of scattering from sublevels of atomic or ionic fine structure and sublevels formed by energy state splitting in crystalline field as well. So, there are reasons to turn once more to Ref. 17 concluding that the superbroadened spectrum formation in various media is related to SRS process in one way or another.

We suppose below that the SRS mechanism is the basic mechanism for nonlinear transformation of USP spectrum and consider the evolution of a pulse whose duration is comparable to or smaller than the characteristic time Ω^{-1}. The analytic investigation of Eqs. (1.1)–(1.4) [see formulas (1.6)] as well as their numerical solution show that the main parameter governing the evolution of USP spectrum is its energy $W(0)=\Psi(0,\infty)$. There are ranges of $W(0)$ values characterized by qualitatively different dynamics of the spectrum $F(\omega)$. If the USP input energy is smaller than π, then as shown before (Fig. 1) the pulse spectrum is broadened to the long-wave region. Physically, this is associated with the fact that the spectrum in this case is formed by induced transitions of the system from the lower to the upper level under the action of USP, i.e. $\partial\rho(z,t)/\partial t > 0$, and consequently Stokes scattering prevails. Note in particular that this result agrees with the experiments on transformation of optical soliton spectra in waveguides and on long-wave continuum generation in relatively low fields ($\leqslant 10^{12}$ W/cm^2).[10]

The situation is fundamentally different if the USP input energy $W(0)$ is larger than 2π. In this case, two distinct groups of intervals may be selected in the space-time structure of the field $\mathscr{E}(z,t)$. The first group of intervals corresponds to the values of the instantaneous phase $\Psi(z,t)$ such that $2\pi k < \Psi(z,t) < (2k+1)\pi$, $k=0, 1, 2,...$ For these $\Psi(z,t)$ values, Stokes scattering prevails in the SRS process. The second group of intervals in the USP field structure is characterized by instantaneous phase values $(2k+1)\pi < \Psi(z,t) < (2k+2)\pi$ for which the process of medium-field interaction is accompanied by the inverse transition of the system from the upper to the lower level ($\partial\rho(z,t)/\partial t < 0$). This means that anti-Stokes scattering of the pulse field prevails in these intervals. The propagation of a pulse of energy $W(0) > 2\pi$ is thus accompanied by concurrent processes of generating both Stokes and anti-Stokes frequency components. The results of the numerical investigation of this effect for the case of a pulse of energy $W(0)=2\pi$ are shown in Fig. 2. It is seen that the pulse spectrum starts broadening to both low- and high-frequency regions simultaneously as opposed to a SRS pulse of energy $W(0) < \pi$. For lengths $|\beta| z \geqslant 0.55$, the USP spectrum extends from zero frequencies to frequencies exceeding the initial carrier frequency of the pulse by a factor of 2–3, i.e. generation of spectral continuum is observed. This effect may be interpreted as a result of multiple Stokes scattering of one group of photons and multiple anti-Stokes scattering of other group of photons. It is pertinent to stress once more the qualitatively new features in the interaction between electromagnetic field and medium, when the dynamics of a pulse is characterized by a continuous frequency variation to both low-frequency (in the leading part of a 2π-pulse) and high-frequency (in its trailing part) spectrum regions.

Note in conclusion that for a fixed USP width, the spectrum superbroadening effect will appear if the radiation intensity exceeds the threshold value

$$I_{\text{th}} = c(\hbar\Omega M)^{1/2}\left(\frac{\partial\alpha}{\partial Q}\right)^{-1}\tau_p^{-1}, \tag{2.1}$$

which is determined from the condition that the energy density of the pulse exceeds that of a 2π-pulse.

3. USP TEMPORAL COMPRESSION IN RAMAN-ACTIVE MEDIUM WITH LINEAR DISPERSION

In the analysis of the USP propagation dynamics we have not assumed yet that the dominating mechanism in the process of field–medium interaction is the excitation of the phonon subsystem. In actual experimental conditions, the impact of other processes, linear and nonlinear in field, on the pulse evolution may turn out to be fundamental. In what follows, we dwell on the analysis of linear dispersion effect on the USP propagation in a Raman-active medium and show that a joint action of these two processes results in new effects in the pulse propagation dynamics.

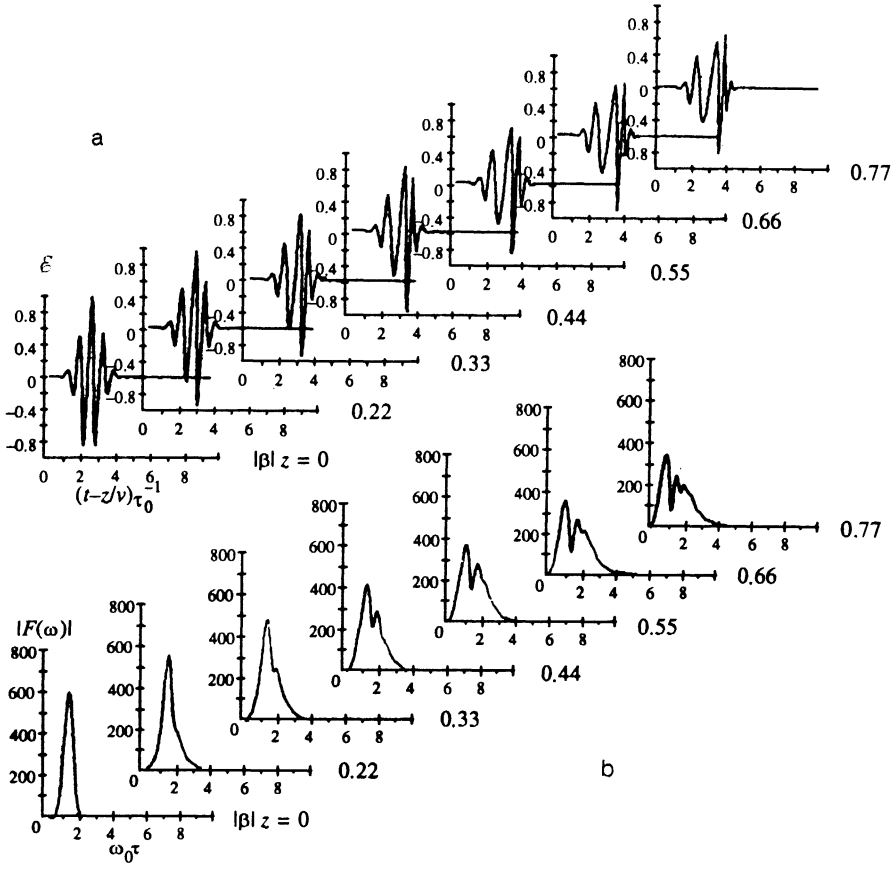

FIG. 2. Generation of spectral continuum in the process of high-power pulse propagation in a Raman-active medium; evolution of electric field $\mathscr{E}(z,t)$ (a) and spectrum $F(\omega,t)$ (b) of a pulse. At the medium input, $\mathscr{E}(0,t)\sim\sin(\omega,t)\exp\{-(t/\tau_0)^2\}$; $(\omega/2\pi)\tau_0=4$; $W(0)=2\pi$; $\Omega\tau_0=0.1$. $\mathscr{E}$ and F are given in arbitrary units.

We assume that an USP whose duration satisfies the condition (1) may be regarded as a quasimonochromatic signal of frequency $\omega\gg\tau_p^{-1}$ for the propagation lengths under consideration, and its field may be represented in the form

$$\mathscr{E}(t)=\tfrac{1}{2}\{\tilde{E}e^{i(\omega t-kz)}+\text{c.c.}\}, \tag{3.1}$$

where $\tilde{E}=Ee^{i\varphi}$ is the complex amplitude ($E=|\tilde{E}|$), φ is the phase. As already mentioned in Sec. 2, in this case the influence of SRS on the USP spectrum evolution results in self-phase modulation of the field, which is described by formula (1.10) in the absence of dispersion. Furthermore, let the pulse energy be small, i.e. $W(0)\ll 1$. For small energies, $\sin\tilde{\Psi}\simeq\tilde{\Psi}$, and we can rewrite (1.10) in the form

$$\dot{\varphi}(z,t)=\beta z\omega\tilde{\Psi}(z,t), \tag{3.2}$$

whence it follows that the addition to the field carrier frequency is proportional to the pulse energy that has passed through a given cross section z by some instant of time t. Since $\beta<0$ in a noninverted medium, Eq. (3.2) implies that the pulse instantaneous frequency decreases when going from the leading to the trailing edge. Note that the integral form of field intensity dependence of the instantaneous frequency results, in fact, from coherent nature of the interaction between a pulse and a Raman-active medium.

Let us consider now the impact of linear dispersion of medium on the pulse evolution. If the field carrier frequency ω lies in the region of normal dispersion of group velocities, i.e. $\partial^2k(\omega)/\partial\omega^2>0$, where $k(\omega)=\omega n(\omega)/c$, then the low-frequency components of the pulse spectrum will travel faster than the high-frequency ones, and for a certain propagation length temporal pulse compression may be expected.

To investigate this possibility, let us go from Eq. (1.8) to an equation for the slowly varying complex amplitude E allowing for dispersive spreading:

$$\frac{\partial\tilde{E}}{\partial z}-\frac{i}{2}k_{\omega\omega}\frac{\partial^2\tilde{E}}{\partial\tau^2}=-i\gamma\tilde{E}\int_{-\infty}^{\tau}d\tau'\int_{-\infty}^{\tau'}|\tilde{E}(\tau'')|^2d\tau''. \tag{3.3}$$

Here, $\gamma=(\pi\omega N|\rho_0|/2cnM)(\partial\alpha/\partial Q)^2$; $k_{\omega\omega}=\partial^2k/\partial\omega^2$; $\tau=(t-z/v)$ is the retarded time. The parameters of Eq. (3.3) governing the pulse dynamics are the dispersive spreading length z_d and the self-phase modulation length z_{spm}:

$$z_d=\tau_0^2K_{\omega\omega}^{-1},\quad z_{spm}=(\gamma E_0^2\tau_0^2)^{-1}, \tag{3.4}$$

where E_0 and τ_0 are the characteristic values of the field strength and the pulsewidth at the medium input. The length z_{spm} determines the scale for which the nonlinear variation $\dot{\varphi}$ becomes equal to the inverse pulsewidth τ_0^{-1}.

Before we turn to the numerical analysis of (3.3), several qualitative estimates for the optimum compression length z_{opt} and the pulse compression factor s_{opt} can be made based on the considerations employed to estimate these parameters in media with Kerr nonlinearity.[6] The value of pulse spectrum nonlinear broadening for the prop-

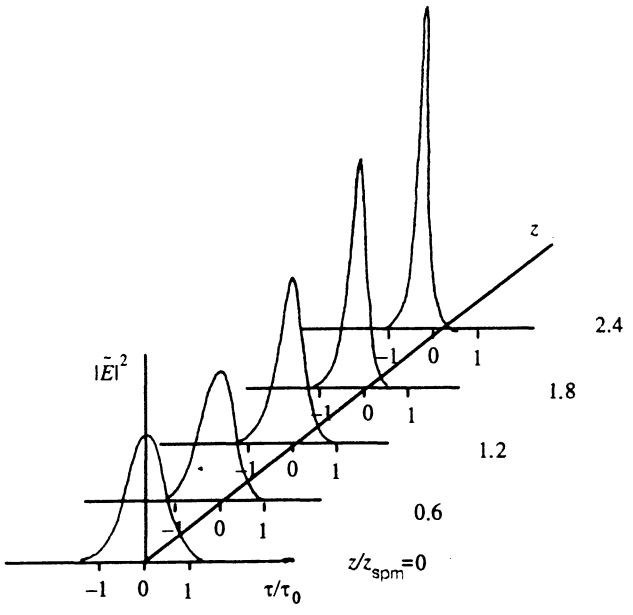

FIG. 3. Compression of a quasimonochromatic pulse in the process of propagation in a Raman-active medium with normal dispersion. At the medium input, the pulse envelope is $\tilde{E}(0,\tau)\sim\exp\{-(\tau/\tau_0)^2\}$; $z_d/z_{spm}=8$; $\Omega\tau_0=0.1$.

agation length z is $\Delta\omega\simeq\tau_0^{-1}(z/z_{spm})$. The dispersion-induced relative time delay of the high-frequency and the low-frequency components corresponding to this spectrum width may be estimated as $\Delta t_d\simeq zk_{\omega\omega}\Delta\omega$. Obviously the maximum pulse compression occurs when the Stokes components localized in the pulse "tail" overtake the components of the leading edge, which have not yet been scattered, i.e. when $\Delta_d\simeq\tau_0$. From this condition, we find that the optimum compression length to order of magnitude is equal to

$$z_{opt}\sim(z_d z_{spm})^{1/2}=E_0^{-1}(\gamma K_{\omega\omega})^{-1/2} \tag{3.5}$$

and does not depend on the pulsewidth τ_0. The compression factor will be determined from the ratio of the pulse spectrum $\Delta\omega(z_{opt})$ broadened for the length z_{opt} and the input spectrum width $\Delta\omega(0)\simeq\tau_0^{-1}$. So, we find

$$s_{opt}\sim(z_d/z_{spm})^{1/2}=\tau_0^2 E_0(\gamma/k_{\omega\omega})^{1/2}. \tag{3.6}$$

Thus, it is necessary that the self-phase modulation length z_{spm} is shorter than the dispersion spreading length z_d in order to realize pulse compression.

The numerical analysis of the USP dynamics performed on the basis of Eq. (3.3) showed that formulas (3.5) and (3.6) satisfactorily describe the dependencies of s_{opt} and z_{opt} on parameters of a medium and a pulse. One of representative calculations of pulse time compression for the optimum compression length z_{opt} is presented in Fig. 3. One feature of the pulse dynamics in a dispersive Raman-active medium (see Fig. 3) is an asymmetry of the compression process in the (z,τ) coordinate system. It is associated with the fact that at $W(0)\ll 1$ nonlinear spectrum broadening proceeds to the Stokes region only and increases when going to the pulse "tail." As a consequence, pulse compression is a result of its energy concentration near the leading front.

4. SPECIAL FEATURES OF USP PROPAGATION IN MULTILEVEL RAMAN-ACTIVE MEDIA

Stimulated light scattering from molecule vibrations is one of traditional and most efficient mechanisms for frequency conversion in Raman-active media. At the same time, a number of special features are exhibited when a molecule is excited by an USP satisfying the condition (1). Namely, a variety of vibrational quantum states, instead of single one, can be excited by an USP of a sufficiently high power. So, the two-level model cannot be applied. In the present section, we examine USP propagation in a multilevel molecular medium and show that nonlinear effects arising here are qualitatively analogous to the ones considered earlier for a two-level phonon subsystem.

In the limit of ultrashort action (1), it becomes physically justified and convenient, as we see later, to describe a molecular system directly in terms of wave function of the vibrational Hamiltonian rather than in terms of probability amplitudes for vibrational sublevels. Restricting ourselves to the case of two-level molecules, we write the Schrödinger equation for the wave functions of its electron states Φ_n depending on the internuclear distance R:[18]

$$i\hbar\frac{\partial\Phi_n}{\partial t}+\frac{\hbar^2}{2M}\nabla^2\Phi_n-U_n(R)\Phi_n=-\sum_{k\neq n}\mathscr{E}\mu_{nk}\Phi_k. \tag{4.1}$$

Here, $U_n(R)$ is the adiabatic energy for the nth electron state, M is the reduced molecular mass, $\mu_{nk}(R)$ is the matrix element of the dipole moment operator of the electron transition. A molecule is assumed to have a zero dipole moment in (4.1) for the sake of simplicity, i.e. $\mu_{nm}=0$.

Let the molecule be in the state Φ_0 ($n=0$) prior to the interaction with an USP field ($t=-\infty$), while for the remaining states with $n\neq 0$ we have $\Phi_n=0$. If none of the field spectral components is in resonance with the frequencies of electron transition $(U_n-U_0)/\hbar$, then the perturbation theory gives the wave functions Φ_n with $n\neq 0$ to first order in the field $\mathscr{E}$. We shall present the corresponding expressions for the case when the frequencies of all the spectral components of the pulse are much lower than the characteristic frequencies of transitions between electron levels.[2)] In this limit, we get an expression for $\Phi_n^{(1)}$

$$\Phi_n^{(1)}\approx\frac{\mu_{n0}\mathscr{E}}{U_n-U_0}\Phi_0. \tag{4.2}$$

The substitution of (4.2) in Eq. (4.1) gives a closed equation for the wave function Φ_0:

$$i\hbar\frac{\partial\Phi_0}{\partial t}+\frac{\hbar^2}{2M}\nabla^2\Phi_0-U_0(R)\Phi_0=-\frac{1}{2}\alpha_{\gamma\delta}(R)\mathscr{E}_\gamma\mathscr{E}_\delta\Phi_0. \tag{4.3}$$

The factor multiplying Φ_0 in the right side of (4.3) has a meaning of energy of interaction between the induced dipole moment and the field. The quantity

$$\alpha_{\gamma\delta}(R)=2\sum_{n\neq 0}\frac{(\mu^{\gamma}(R))_{0n}(\mu^{\delta}(R))_{n0}}{U_n(R)-U_0(R)} \tag{4.4}$$

is merely the electron polarizability of a molecule in the low-frequency limit $\omega\to 0$.

The field-induced deviation of the internuclear distance from the equilibrium value $x=R-R_0$ will be assumed to be negligible, so the potential of the electron ground state may be approximated by the harmonic potential $U_0(R)=1/2M\Omega^2X^2$.

Expanding the polarizability $\alpha_{\gamma\delta}(R)$ in series in small X, we go from Eq. (4.3) to the equation

$$i\hbar\frac{\partial\Phi_0}{\partial t}+\frac{\hbar^2}{2M}\frac{\partial^2}{\partial X^2}\Phi_0-\frac{1}{2}\left[M\Omega^2X^2-\left(\frac{\partial\alpha}{\partial X}\right)_{\gamma\delta}\mathscr{E}_{\gamma}\mathscr{E}_{\delta}X\right]$$

$$\Phi_0=0. \tag{4.5}$$

Prior to the action of an USP, let the molecule be at the lowest vibrational level, i.e.

$$\Phi_0(t=-\infty)=\varphi_{v=0}=\left(\frac{M\Omega}{\pi\hbar}\right)^{1/4}\exp\{-M\Omega X^2/2\hbar\}. \tag{4.6}$$

Using a formal analogy between the problem under study and propagation of Gaussian beams in a waveguide with a parabolic profile of refractive index,[19] we can try a solution of (4.5) in the form of a Gaussian wave packet

$$\Phi_0(X,t)=\exp\left\{-i\left(\frac{1}{2}qX^2+sX+p\right)\right\} \tag{4.7}$$

with time-dependent parameters q, s, and p. For the parameter $\bar{X}(t)=-\mathrm{Im}(s)/\mathrm{Im}(q)$ determining, according to (4.7), the center of the wave packet $|\Phi_0(X,t)|^2\sim\exp\{-M\Omega(X-\bar{X})^2/\hbar\}$ we easily get an equation:

$$\frac{\partial^2\bar{X}}{\partial t^2}+\Omega^2\bar{X}=\frac{1}{2M}\left(\frac{\partial\alpha}{\partial X}\right)_{\gamma\delta}\mathscr{E}_{\gamma}\mathscr{E}_{\delta}. \tag{4.8}$$

The polarization induced in a molecule by the external field is

$$\mathscr{P}_{\gamma}(t)=\sum_{m\neq n}\int\Phi_m\mu_{mn}\bar{\Phi}_n^{*}dR$$

$$=\int|\Phi_0(X,t)|^2\alpha_{\gamma\delta}(X)\mathscr{E}_{\delta}(t)dX, \tag{4.9}$$

or, using representation (4.7) for Φ_0,

$$\mathscr{P}_{\gamma}=\alpha_{\gamma\delta}(0)\mathscr{E}_{\delta}+\left(\frac{\partial\alpha}{\partial X}\right)_{\gamma\delta}\mathscr{E}_{\delta}\bar{X}, \tag{4.10}$$

i.e. the nonlinear part of the response due to SRS is expressed in terms of the coordinate of the wave packet center. It is easy to see from the comparison of Eqs. (1.1), (1.4) and (4.8), (4.10) that the coordinate of the wave packet center $\bar{X}$ in a multilevel system plays the same role as the normal coordinate in the two-level model. Equation (4.8) coincides with linearized Eq. (1.1) ($n\simeq -1$) of a two-level oscillator.

So, the nonlinear effects considered above within the framework of the two-level model for weak saturation of the transition ($W(0)<1$), such as videopulse generation from high-frequency signals, spectrum superbroadening to the Stokes region, temporal pulse compression in a medium with linear dispersion, can be observed in molecular media as well. Note in conclusion that the harmonic potential approximation does not make it possible to describe the effects of pulse spectrum broadening to the anti-Stokes region. These effects can probably be included in consideration as a consequence of saturation of the molecule response to the external field, when anharmonicity is allowed for.

CONCLUSION

In the present paper, the propagation dynamics of high-power femtosecond optical pulses in a Raman-active medium is studied under the conditions when the pulse width is small compared to the period of characteristic natural phonon, molecular, etc. vibrations in matter. Based on the solution of exact equations for electromagnetic field and polarization of a Raman-active medium, the feasibility of new nonlinear effects is shown, such as self-induced transparency of $2\pi n$-pulses of electromagnetic field, generation of videopulses (i.e. pulses without a carrier frequency) from high-frequency signals. The propagation of pulses of sufficiently high power is shown to be accompanied by spectrum superbroadening to both Stokes and anti-Stokes regions. The rearrangement of the pulse spectrum due to SRS leads to temporal pulse compression under the conditions of normal dispersion in a medium.

Attention is paid to qualitatively new physics of nonlinear optical processes in the range of ultrashort pulsewidths ($\tau_p\ll\Omega^{-1}$) and ultraintense fields ($I\geqslant I_{\mathrm{th}}$). The dynamics of the field interacting with the medium is characterized by the variation of the photon energy itself rather than the variation of the photon number.

Let's estimate now the possibility of experimental observation of the effects under discussion. Assuming the optical phonon frequency in a substance $\Omega\sim 10^{13}$ rad/s, effective molecular mass $M\sim 10^{-22}$ g, polarizability $\partial\alpha/\partial Q\sim 10^{-15}$ cm^2, and concentration $N|\rho_0|\sim 10^{22}$ cm^{-3}, we estimate the rectification length from high-frequency field to videopulse from (1.11). For instance, for a pulsewidth $\tau_p=20$ fs, a carrier frequency $\omega=2\cdot 10^{15}$ s^{-1}, and an intensity $I=10^{13}$ W/cm^2, we find that $z_{\mathrm{rect}}\simeq 0.3$ cm. The field strength in the videopulse will be $\mathscr{E}\sim 10^7$ V/cm. This suggests that Stokes scattering of femtosecond optical pulses in a Raman-active medium can be a highly efficient method for intense videopulse generation. Note that in comparison with the technique of videopulse production based on generation of IR field in quadratic crystals (the Cerenkov pulses), where the conversion efficiency is low in view of the lack of phase matching between the optical and the IR fields, the proposed approach does not rely upon phase-matched interaction of waves at all.

According to (2.1), the spectrum superbroadening to both the Stokes and anti-Stokes regions will be observed at pulse intensities $I>10^{14}$ W/cm^2, and for the propagation

length cited above the spectral broadening will become comparable to the carrier frequency of the initial pulse to order of magnitude.

Finally, we estimate the conditions of pulse compression in a Raman-active medium. For example, for a pulse of width $\tau_p = 40$ fs, the dispersion spreading length is $z_d \simeq 16$ cm at a value of normal group velocity dispersion $k_{\omega\omega} \sim 10^{-28}$ s^2/cm.[6] Then we find from (3.6) that a fourfold pulse compression to a width $\tau_p = 10$ fs will occur at an input intensity $I \simeq 10^{11}$ W/cm^2 for a propagation length $z_{opt} \simeq 4$ cm.

The presented estimates thus show that the experimental realization of the effects under discussion is quite feasible with the help of modern femtosecond laser sources.

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[1)]To the best of our knowledge videopulses with $\tau_p \sim 10^{-12}$ s are produced when shorting charged strip lines by radiation of femtosecond lasers;[13] in the process of optical "rectification" of an USP in a medium with quadratic nonlinearity, Cerenkov bipolar pulses appear ($\tau_p \leqslant 10^{-12}$ s).[5]

[2)]It should be noted that the equations of the Bloembergen–Shen model (1.1), (1.2), in which the oscillator polarizability does not depend on spectral composition of the excitation field $\mathscr{E}$, actually corresponds to this low-frequency limit.

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