Dynamics of an intense femtosecond pulse in a Raman-active medium

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The dynamics of an intense ultrashort light pulse in a Raman-active medium is analyzed in the case in which the pulse length is short in comparison with the time Ω^{-1}, where $\hbar\Omega = E_2 - E_1$ is the energy difference between the levels participating in the Raman scattering. Some new nonlinear effects can arise: the existence of $2\pi n$ pulses of a self-induced transparency of the Raman-active medium and the generation, from a high-frequency signal, of video pulses of an electromagnetic field without a carrier frequency. The physics of the processes is new in a qualitative sense when the dynamics of the field is governed by a change in the energy of the photons rather than by a change in their number.

1. Substantial progress has been made in generating intense ultrashort light pulses with intensities $I \sim 10^{15}$ W/cm^2 and lengths of a few femtoseconds.[1] Experiments (e.g., Refs. 2 and 3) show that pulses with these properties propagate through a Raman-active medium under special conditions. The reason is that the typical value of the time Ω^{-1}, corresponding to the Stokes (or anti-Stokes) frequency shift Ω, does not exceed 10^{-13} s in most Raman-active media. For ultrashort pulses with a length $\tau_p \lesssim 10^{-14}$ s the following condition thus holds:

$$\tau_p \Omega \ll 1. \tag{1}$$

The pulse spectrum initially contains an infinite set of frequency components which are comparable in amplitude and which satisfy the condition for a Raman resonance. Accordingly, the question of the dynamics of an intense ultrashort pulse under condition (1) is essentially impossible to resolve by conventional Fourier analysis. However, one can solve this problem by formulating it in terms of the real field $\mathscr{E}$ and the real polarization $\mathscr{P}$ induced by this field. It thus becomes possible to bring out the qualitatively new nature of the nonlinear-optics processes which occur when it is no longer possible to speak in terms of definite frequencies or wavelengths of the interacting fields. The field dynamics in this case is characterized by a continuous change in the energy of the photons; i.e., it becomes a dynamics of photons.

2. We describe the evolution of an ultrashort light pulse in a two-level Raman-active medium by the system of constitutive equations

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

$$\frac{\partial \rho}{\partial t} + \frac{\rho - \rho_0}{T_1} = \frac{1}{\hbar\Omega}\left(\frac{\partial \alpha}{\partial Q}\right)\mathscr{E}^2 \frac{\partial Q}{\partial t}, \tag{3}$$

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and by a wave equation for the field,

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

where the polarization of the medium, $\mathcal{P}$, has a component which is linear in the field, $\mathcal{P}_{\mathrm{L}}$, and a component which is nonlinear in the field, $\mathcal{P}_{\mathrm{NL}}$ (a consequence of the Raman scattering).

Constitutive equations (2)–(3) describe a nonlinear oscillator with a normal coordinate Q, a reduced mass M, a frequency $\Omega = (E_2 - E_1)\hbar^{-1}$, and phenomenological relaxation times T_1 and T_2. This oscillator is excited by a driving force proportional to the product of the square of the field and the difference between the level populations, $\rho = n_2 - n_1$. The proportionality coefficient, $\partial\alpha/\partial Q$, is written in the standard form, as the derivative of the polarizability $\alpha(Q)$ with respect to the normal coordinate Q near the equilibrium value $Q = Q_0$. These equations can describe a variety of processes, depending on the particular physical situation. For example, they can describe a scattering of light by optical phonons in a solid and the interaction of ultrashort pulses with an isolated two-level system with a transition which is forbidden in the electric dipole approximation.[4]

The nonlinear polarization induced by the field, which appears in wave equation (4), is given by

$$P_{\mathrm{NL}} = N\left(\frac{\partial\alpha}{\partial Q}\right) Q\mathcal{E}, \tag{5}$$

where N is the number density of particles. The case $\rho_0 = -1$ corresponds to an interaction of the field with a noninvertible medium, and the case $\rho_0 = +1$ corresponds to a medium in which the upper level is fully populated.

Condition (1) presupposes that the interaction of the light with the medium is coherent ($\tau_p \ll T_1, T_2$) and is characterized by a very slow response of the medium to the field of the ultrashort pulse. This condition also implies that the term $\Omega^2 Q$ in (2) can be ignored in comparison with $\partial^2 Q/\partial t^2$. After these simplifications, the constitutive equations can be integrated for an arbitrary field shape. Specifically, introducing an angle through which the constitutive variables are rotated,

$$\Psi(z,t) = \left|\frac{\partial\alpha}{\partial Q}\right| (\hbar\Omega M)^{-1/2} \int_{-\infty}^{t} \mathcal{E}^2(z,t')dt', \tag{6}$$

we find

$$\rho = \rho_0 \cos\Psi, \quad \frac{\partial Q}{\partial t} = -\rho_0 \operatorname{sign}\left(\frac{\partial\alpha}{\partial Q}\right)\left(\frac{\hbar\Omega}{2M}\right)^{1/2} \sin\Psi. \tag{7}$$

For a pulse which is propagating along the positive z direction, we then have

$$\frac{\partial\mathcal{E}}{\partial z} + \frac{1}{v}\frac{\partial\mathcal{E}}{\partial t} = \beta\frac{\partial}{\partial t}(\mathcal{E}\int_{-\infty}^{t} \sin\Psi(z,t')dt), \tag{8}$$

where $\beta = 2\pi N\rho_0 |\partial\alpha/\partial Q| (\hbar\Omega/2Mc^2)^{1/2}$, and the quantity $|\beta|^{-1}$, which has the dimensionality of a length, can be called the "Raman self-scattering length." Equation (8) was derived from wave equation (4) with the help of (5)–(7) and under the condition that the field of the pulse varies only slightly over propagation distances on the order of its own length.

From (8) we find an equation for the change in the total energy $W(z) = \Psi(z,\infty)$ of the pulse along the coordinate z:

$$\frac{dW}{dz} = \beta(1 - \cos W). \tag{9}$$

We see that the behavior of the energy in (9) is essentially the same as in the case of a coherent two-photon interaction of a pulse with a medium:[5] Pulses with an energy $W = 2\pi k (k = 1,2,...)$ propagate under conditions of an induced self-transparency. This result is not surprising, since both of these processes are two-quantum processes, and they obey the same selection rules. In the case of a two-photon interaction, the interpretation of the effect is essentially the same as that in the model of the one-quantum effect of McCall and Hahn. For the Raman scattering under consideration here, the process is more complicated. Specifically, for $\rho_0 = -1$ the front of a 2π pulse is absorbed during the generation of the Stokes components of the field; the anti-Stokes components restore energy to the tail of the pulse. The pulse itself is time-varying in terms of shape and spectrum: The trailing edge of the pulse becomes enriched in high-frequency components, into which the energy of the leading edge is pumped. Consequently, the pulse becomes compressed with increasing z. We will not reproduce here the results of a numerical solution of Eqs. (1)–(3). Qualitatively, the same dynamics of the spectrum of a 2π pulse can be seen in a quasimonochromatic approximation, which is valid at least for propagation distances $z < |\beta|^{-1}$.

We write the electric field of the ultrashort pulse as a quasimonochromatic wave with an amplitude $E(z,t)$ and a frequency $\omega(z,t) = \omega + \dot{\varphi}(z,t)$. We can then switch from Eq. (8) to equations for the "slow" amplitude E and the "slow" phase φ. The phase self-modulation of the ultrashort pulse which occurs in the Raman-active medium is then described by

$$\dot{\varphi}(z,t) \approx \beta z \omega \sin \tilde{\Psi}, \tag{10}$$

where $\tilde{\Psi}$ is given by (6) after the substitution $\mathscr{E}^2 \to \frac{1}{2}E^2$. For a low-energy pulse ($\sin \tilde{\psi} \approx \tilde{\psi}$) in an absorbing medium ($\rho_0 = -1$), we find immediately from (10) that the instantaneous frequency $\omega(t)$ of the pulse decreases from the leading edge to the trailing edge. This decrease is a consequence of the coherent nature of the interaction of the Raman-active medium with the field. The maximum frequency shift, corresponding to the trailing edge of the pulse, is proportional to the energy of the pulse. If the Raman scattering occurs in a medium with a normal group-velocity dispersion, the phase self-modulation will cause a compression of the pulse.[6]

We now consider the propagation of an ultrashort pulse of low energy $\Psi(z,\infty) < 1$ over distances $z > |\beta|^{-1}$, such that the change in the frequency of the pulse is large, and the process cannot be regarded as quasimonochromatic. Because of the predominant Stokes scattering, the effective frequency of the pulse, $\omega(z)$, shifts continuously in the red direction. This process can come to a halt only when the energy of the pulse is completely exhausted as a result of the excitation of the medium (compare this situation with ordinary Stokes scattering of a monochromatic field, in which case the pumping is limited by conservation of the total number of photons). One might expect that as the frequency of the ultrashort pulse decreased to $\omega(z) \lesssim \tau_p^{-1}$, the wavelength of the pulse which forms would become comparable to the

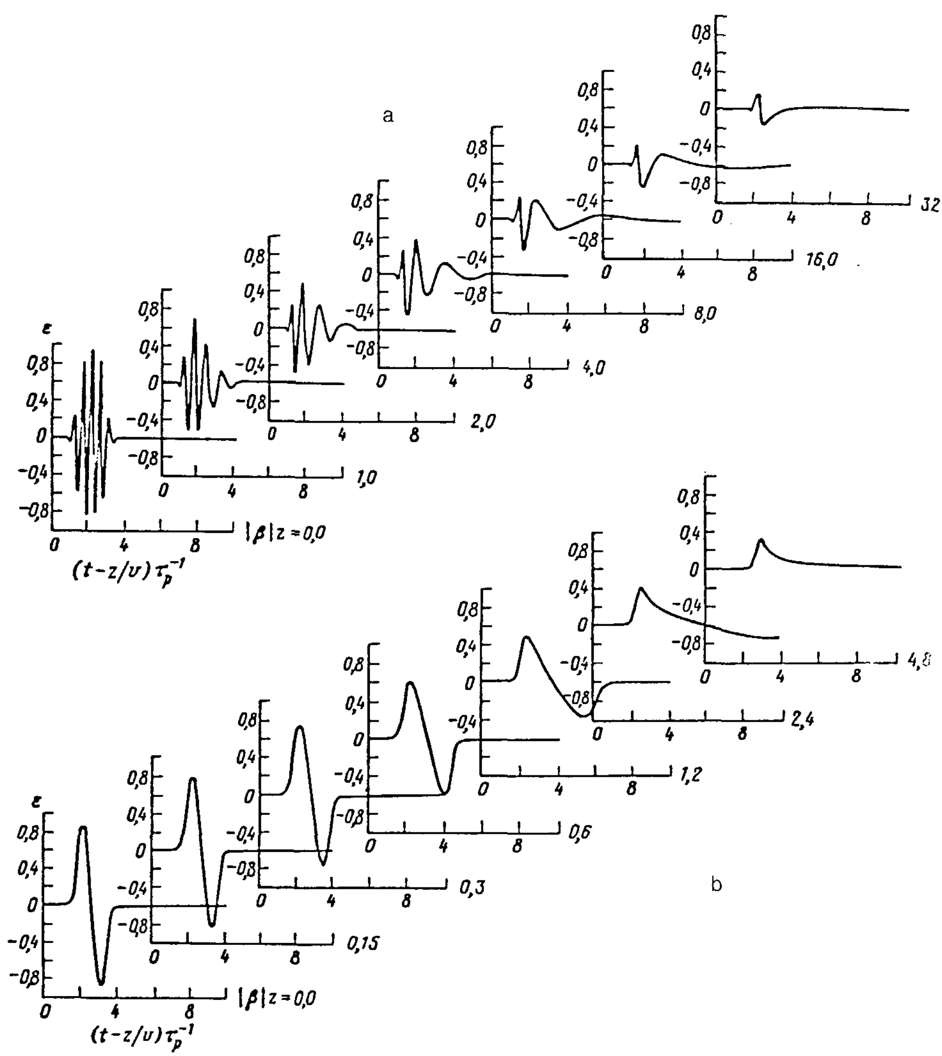

FIG. 1. Generation of video pulses, either (a) bipolar or (b) unipolar, during the propagation of an ultrashort pulse through a Raman-active medium. As it enters the medium, the field of the ultrashort pulses is $\mathscr{E}(0,t) \sim \sin(\omega t) e^{-(t/\tau_p)^2}$: a—$(\omega/2\pi)\tau_p = 4$, $\Omega\tau_p = 0.1$; b—$(\omega/2\pi)\tau_p = 1$, $\Omega\tau_p = 0.1$. The field of the ultrashort pulses is polarized along the x axis and is propagating through a slab $|x| < a$ of a Raman-active medium. The volume $|x| > a$ is filled with a medium with a conductivity $\sigma = \infty$.

spatial size of the pulse. This situation would imply that an initial pulse in the visible range $[\omega(0)\tau_p \gg 1]$ would convert into a video pulse, i.e., a pulse without a carrier frequency.[1)] These qualitative arguments are supported completely by the results of a numerical solution of Eqs. (2)–(4); these results are shown in Fig. 1. The length scale for the "rectification" of the original ultrashort pulse to a video pulse is inversely proportional to the energy of the initial pulse. It can be found from the expression $L_{\rm rect} \sim \omega r_p(\beta W(0))^{-1}$. Assuming $\tau_p = 20$ Fs, $\omega = 2\times 10^{15}$ rad/s, $\Omega = 10^{13}$ rad/s, $\partial\alpha/\partial Q \sim 10^{-15}$ cm^2, and $N \sim 10^{21}$ cm^{-3} for estimates, we find that at an intensity $I = 3\times 10^{13}$ W/cm^2 this length would be $L_{\rm rect} \sim 1$ cm. The intensity of the video pulse would be $\sim 10^{12}$ W/cm^2.

This analysis of the propagation of ultrashort pulses through a Raman-active medium thus indicates that the nonlinear-optics processes which occur at ultrashort pulse lengths ($\tau_p \ll \Omega^{-1}$) and at ultraintense fields $[\Psi(z,\infty) \sim 1]$ are of a qualitatively new nature. The dynamics of a field which is interacting with a medium is characterized by a change in the energy of the photons themselves, rather than by a change in the number of photons.

We note in conclusion that, according to Ref. 8, diffraction effects would lead to the requirement that the area under the electric field of the pulse must vanish; i.e., a pulse with a limited aperture could not have a constant component. This condition could be satisfied by, for example, a train of two or more temporally separated video pulses with fields differing in sign.

[1)] To the best of our knowledge, video pulses with $\tau_p \sim 10^{-12}$ are produced by shorting charged striplines by means of beams from femtosecond lasers.[7] During optical "rectification" of an ultrashort light pulse in a medium with a quadratic nonlinearity, bipolar Čerenkov pulses ($\tau_p \sim 10^{-13}$ s) appear.[1]

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