Transformation of the field of an intense ultrashort pulse in a Raman-active medium

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(Submitted 2 November 1994)
Pis'ma Zh. Eksp. Teor. Fiz. **60**, No. 11, 762–765 (10 December 1994)

The propagation of an intense femtosecond pulse in a Raman-active medium is analyzed. The field of an intense light wave undergoes a substantial transformation as the wave propagates through the medium. The nature of this transformation can change over time scales comparable to the period of the optical oscillations. The transformed spectrum of the light is found for an optically thin medium. © *1994 American Institute of Physics.*

The physics of the nonlinear interactions of intense ultrashort light pulses with media has attracted interest because of progress in subpicosecond-range laser technology and the attainment of laser-beam power levels in the terawatt range (Ref. 1, for example). The dynamics of the light pulse and the evolution of the medium in an intense field differ qualitatively from the usual picture drawn by standard nonlinear-optics perturbation theory. A fundamental distinctive feature of ultrashort pulses is that their length is shorter than the time scale of the response of the medium, so the interaction definitely occurs in a coherent regime. The standard approximation of a slowly varying amplitude and a slowly varying phase of the field becomes ineffective. A description of the interaction based on the actual (instantaneous) field values is appropriate.[2,3]

In this letter we take up the problem of the propagation of an intense femtosecond pulse in a Raman-active medium. We derive an analytic solution which describes the evolution of the light pulse. As the intense light wave propagates through the medium, its field undergoes a substantial transformation. The nature of this transformation can change over time scales comparable to the period of the optical oscillations. In particular, for an optically thin medium the transformed spectrum of the light is similar to the spectrum of multiple harmonic generation in inert gases.[4]

To describe the dynamics of a Raman-active medium in the field of an ultrashort pulse, we use the model of a two-level nonlinear oscillator, as in Refs. 2 and 5:

$$\frac{\partial^2 Q}{\partial t^2}+\frac{1}{T_2}\frac{\partial Q}{\partial t}+\Omega^2 Q=-\frac{1}{2M}\left(\frac{\partial\alpha}{\partial Q}\right)E^2\rho,\qquad \frac{\partial\rho}{\partial t}+\frac{\rho-\rho_0}{T_1}=\frac{1}{\hbar\Omega}\left(\frac{\partial\alpha}{\partial Q}\right)E^2\frac{\partial Q}{\partial t}. \tag{1}$$

Here Q is a normal coordinate, M is the reduced mass, Ω is the natural transition frequency of the oscillator (the Stokes shift), T_1 and T_2 are relaxation times, the coefficient $\partial\alpha/\partial Q$ is the derivative of the polarizability at the equilibrium value $Q=Q_0$, and ρ is the difference between the populations of the upper and lower levels (the value ρ_0 corresponds to the state of the system before the beginning of the interaction with the field, at $t=-\infty$). We stress that the quantity $E=E(z,t)$ in (1) is the instantaneous value of the field of the pulse, not its amplitude.

Equations (1) are to be solved jointly with the wave equation. In the case at hand, in which the interaction of the carrier pulse with only the scattered wave propagating in the same direction is taken into account, this wave equation can be written[2]

$$\frac{\partial E}{\partial z}+\frac{1}{c}\frac{\partial E}{\partial t}=-\frac{2\pi}{c}\frac{\partial P^{NL}}{\partial t}, \quad P^{NL}=N\left(\frac{\partial \alpha}{\partial Q}\right)EQ, \tag{2}$$

where N is the density of the medium.

Equations (1) and (2), which constitute a closed, self-consistent system of equations, thus give a complete description of the nonlinear evolution of a ultrashort pulse as it propagates through a Raman-active medium.

We restrict the analysis to the case of the coherent interaction, in which the pulse length does not exceed the time scales of the response and relaxation of the medium: $\tau_p \ll \Omega^{-1}$, T_1, T_2. These conditions hold in the femtosecond range. Constitutive equations (1) can then be integrated for an arbitrary time dependence of the field $E(z,t)$. The dynamics of the medium is determined exclusively by the "phase" of the oscillator:[2]

$$\Psi(z,t)=\left|\frac{\partial \alpha}{\partial Q}\right|(2\hbar\Omega M)^{-1/2}\int_{-\infty}^{t} dt' E^2(z,t'). \tag{3}$$

A solution of system (2), (3) can be found by integrating along characteristics on which the change in "phase" is

$$\tan(\Psi/2)=\frac{\tan(\Psi_0/2)}{1+\beta z\tan(\Psi_0/2)}, \tag{4}$$

where $\Psi_0=\Psi(0,\eta)$ is the given phase at the boundary of the medium. The characteristics themselves are given implicitly by the expressions

$$\eta+\Phi(z,\eta)=t-z/c, \quad \Phi(z,\eta)=\beta z\int_{-\infty}^{\eta} d\eta'\left(\sin\Psi_0+\frac{\beta z}{2}(1-\cos\Psi_0)\right), \tag{5}$$

where $\Phi(z,\eta)$ is the nonlinear delay of the individual parts of the pulse as it moves away from the boundary, and $\beta=(\pi/2)N|\partial\alpha/\partial Q|(\hbar\Omega/2Mc^2)^{1/2}$ is the reciprocal length of induced Raman self-scattering.[2] Equation (4) describes the change in the energy of the pulse as it propagates through the medium, while relation (5) actually determines the transformation of the shape of the pulse. The two processes develop over the same characteristic distance, which is proportional to β^{-1}. For the field in the medium we find from (4) and (5)

$$E(z,t)=\frac{E(0,\eta)}{\left(1+\dfrac{\partial\Phi(z,\eta)}{\partial\eta}\right)}. \tag{6}$$

If the field at the boundary of the medium can be characterized by a frequency $\omega(0,t)$, which may itself vary in time, then as the pulse propagates this frequency transforms in accordance with

$$\omega(z,t)=\frac{\omega(0,\eta)}{\left(1+\frac{\partial\Phi(z,\eta)}{\partial\eta}\right)}. \tag{7}$$

Equations (4)–(6) constitute a complete solution of our problem. It can be seen from (6) and (7) that the changes in the field and the characteristic frequency over space occur identically. At the beginning of the pulse, when the phase of the two-level oscillator satisfies $\Psi_0 \ll 1$ (i.e., when the energy of the part of the pulse which has entered the medium is small), there are decreases in the field and in the field oscillation frequency:

$$E(z,t)=E(0,\eta)/\left(1+\frac{\beta z\Psi_0}{2}\right)^2, \quad \omega(z,t)=\omega(0,\eta)/\left(1+\frac{\beta z\Psi_0}{2}\right)^2.$$

This case corresponds to Stokes scattering. Later, when the phase becomes greater than $\pi/2$ and reaches the value $\arctan(-2/\beta z)$, the field and its frequency increase. The generation of anti-Stokes components of the field thus becomes predominant. At $\Psi_0=\pi$, the nature of the pulse transformation changes again. At a given point in space at different times, corresponding to different characteristics (5), we thus observe oscillations of regimes of compression and stretching of the field oscillation periods. The physical reason for these features is that, as in the generation of $2\pi n$ pulses under conditions of coherent two-photon transitions, the field of the pulse is either in phase or out of phase with the polarization as the pulse propagates through the medium. Correspondingly, the field is either absorbed or amplified. This analysis generalizes the results found on the basis of numerical calculations in Refs. 2 and 3.

The sequence of regimes of stretching and compression of the pulse with increasing value of the incident energy reverses when we switch from an originally absorbing medium to an originally inverted one.

Let us examine the spectral composition of the transformed pulse. From (6) we find the following result for the Fourier components of the field:

$$E_\omega(z)=\int_{-\infty}^{\infty} E(0,\eta)\exp[-i\omega t(z,\eta)]d\eta. \tag{8}$$

We assume $E(0,t)=E_0\sin(\omega_0 t)$, and we restrict the discussion to the case of an optically thin medium, $\beta L \ll 1$, so the phase-matching condition holds for the pump pulse and the harmonics. From (8) we find that the spectrum of the pulse, transformed as a result of stimulated Raman self-scattering, is made up of the components

$$\omega_n=\Omega_0\pm(2n-1)\omega_0. \tag{9}$$

Here $\Omega_0=(1/2)|\partial\alpha/\partial Q|(2\hbar\Omega M)^{-1/2}E_0^2$ is the analog of the Rabi frequency of the two-level oscillator.

For the intensities of the harmonics which are generated, I_n, at the exit from the interaction region, $z=L$, we have the simple relation

$$\frac{I_n}{I_0}=C_n(\beta L)^2, \tag{10}$$

where I_0 is the intensity of the incident pulse. When dispersion of the medium is taken into account accurately, we find an additional factor $\kappa(\omega_n)(\sin\mu_n/\mu_n)^2$ on the right side of Eq. (10), where $\kappa(\omega_n)$ is the refractive index of the medium at the frequency of the nth harmonic, $\mu_n = \Delta k_n L/2$, $\Delta k_n = [k_n - (2n-1)k_0]$ is a parameter of the dispersive mismatch of the phases of the harmonic and of the corresponding component of the nonlinear polarization, and k_n and k_0 are the wave numbers of the harmonic and of the original wave. The coefficients C_n depend on only the normalized intensity $A = \Omega_0/2\omega_0$:

$$C_n \approx [A \pm (n-1/2)]^{1/2} \left(\frac{J_{n-1}(A)}{A \pm (n-1)} \pm \frac{J_n(A)}{A \pm n} \right)^2 . \tag{11}$$

The choice of sign in (11) is dictated by relation (9) for the frequency of the harmonic under consideration. The linewidth is determined primarily by the profile of the envelope of the ultrashort pulse and is described by $\Delta\omega/\omega_0 \sim (A\omega_0)^{-1} dA/dt \sim (\omega_0 \tau_p)^{-1}$.

It is simple to see that the spectrum of the coherent stimulated Raman self-scattering of an intense femtosecond laser pulse as described by Eqs. (9)–(11) retains the basic features characteristic of the spectrum of higher harmonics observed in inert gases.[4] Specifically, the spectrum has a wide plateau of components with intensities which are comparable in magnitude, beyond which, with increasing harmonic index, there is a sharp decay [in accordance with the asymptotic formula $J_n(x) \sim (2\pi n)^{-1/2}(ex/2n)^n$]. We thus have the estimate $\omega^* \sim (1+e/2)\Omega_0$ for the long-wave boundary of the spectrum $(\Omega_0 \gg \omega_0)$.

The spectrum which is generated is nevertheless qualitatively different from the usual spectrum in inert gases, in which one observes exclusively odd harmonics of the carrier frequency. In the case under discussion here, the transformed spectrum of the stimulated Raman scattering acquires an "additional" shift $\sim \Omega_0$, which is generally arbitrary and not determined by any selection rules. This circumstance has certain advantages: It becomes possible to continuously tune the frequency of short-wave light by varying the intensity of the incident pulse.

We conclude with some numerical estimates. Adopting the typical values $\partial\alpha/\partial Q \sim 10^{-15}$ cm^2, $\Omega \sim 10^{13}$ s^{-1}, and $M \sim 5 \times 10^{-23}$ g, we find the energy density corresponding to the change in the pulse transformation regime to be ~ 1 J/cm^2. Consequently, oscillations of the field structure can be observed by using advanced subpicosecond pulse generators. For a carrier frequency $\omega_0 \approx 10^{15}$ rad/s and a pulse intensity $I_0 \sim 10^{16}$ W/cm^2, we find $A \sim 10$, and the short-wave boundary of the spectrum is at the $\sim$50th harmonic. The frequency-transformation mechanism discussed here thus looks extremely promising from the standpoint of developing a source of coherent soft x radiation.

This study had financial support from the Russian Fund for Fundamental Research (Project 93-02-14271) and the International Science Foundation.

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Translated by D. Parsons