

ON THE THEORY OF HIGH-POWER ULTRASHORT PULSE PROPAGATION IN RAMAN-ACTIVE MEDIA

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Abstract

The propagation of an intense femtosecond pulse in a Raman-active medium is analyzed. An analytic solution which describes in explicit form the evolution of the light pulse is derived. 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. As a result, the pulse of sufficiently high energy divides into stretched and compressed domains where the field decreases and increases respectively.

1 Introduction

The physics of interaction of intense ultrashort light pulses with nonlinear media has attracted interest because of progress in subpicosecond-range laser technology and the attainment of laser-beam power levels of terrawatt range (see, for example, [1]). The light pulse dynamics and the medium evolution 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 duration 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].

The Raman-active media can be effectively excited by a single femtosecond pulse because its wide spectrum initially contains intense Stokes and anti-Stokes components of the field [3, 2]. The qualitatively new interaction regime, so-called self-scattering, occurs for light intensities much lower than the threshold ones for ordinary stimulated Raman scattering developing from spontaneous noise [4]. The estimations testify to the fact that the effective excitation of high vibrational levels and even the dissociation of molecules in the field of an ultrashort electromagnetic pulse can be observed using modern femtosecond lasers [5].

On the other hand, the shape and spectrum of femtosecond pulse should undergo a substantial transformation as it propagates through the Raman-active medium. The description of pulse evolution should take into account a substantial redistribution of the medium level populations during the pulse duration that results in different interaction regimes for different pulse fractions: either absorption due to Stokes scattering or amplification due to anti-Stokes component generation become predominant. The simplest model which is widely used in the theory of stimulated

Raman scattering and includes the effect of medium saturation is the model of two-level nonlinear oscillator. Within the framework of this model we succeeded to describe in explicit form the evolution of high-power femtosecond pulse in Raman-active medium.

2 Solution of Wave Equation

We shall describe the dynamics of a Raman-active medium in the field of an ultrashort pulse by a two-level model of a nonlinear oscillator [6]:

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

Here Q is a normal coordinate, M is the reduced mass, Ω is the eigenfrequency of the equivalent nonlinear 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 $\rho_0 = \rho(t = -\infty)$ corresponds to the state of the system before the beginning of the interaction with the field).

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, the wave equation can be written

$$\frac{\partial E}{\partial z} + \frac{1}{c} \frac{\partial E}{\partial t} = -\frac{4\pi}{c} \frac{\partial P}{\partial t} \quad (2)$$

with macroscopic nonlinear polarization of the medium $P = N \left(\frac{\partial \alpha}{\partial Q} \right) EQ$ induced by the field $E(z, t)$, where N is the density of the medium. We stress that the quantity $E = E(z, t)$ in (1) is the instantaneous value of the pulse field strength, not its envelope. The reduced form of the wave equation (2) is provided by the fact that the stimulated Raman backward scattering is very weak owing to the short interaction length of pulses moving in different directions [6].

We restrict the analysis to the case of the coherent interaction, in which the pulse duration does not exceed the time scales of the response and relaxation of the medium $\tau_p \ll \Omega^{-1}, T_1, T_2$.¹

Material equations (1) can then be integrated for an arbitrary time dependence of the field $E(z, t)$ and the solution looks like rotation of material variables

$$\rho(z, t) = \rho_0 \cos \Psi(z, t), \quad \frac{\partial Q}{\partial t} = -\rho_0 \left(\frac{\hbar \Omega}{2M} \right)^{-1/2} \text{sign} \left(\frac{\partial \alpha}{\partial Q} \right) \sin \Psi(z, t). \quad (3)$$

The phase $\Psi(z, t)$ of the material variable rotation is directly proportional to the energy of the pulse fraction which has passed through the given space point z up to the given time t

$$\Psi(z, t) = \left| \frac{\partial \alpha}{\partial Q} \right| (2\hbar \Omega M)^{-1/2} \int_{-\infty}^t E^2(z, t') dt' \quad (4)$$

¹Strictly speaking the coherent regime of interaction is provided by the condition $\Omega_0 = \frac{1}{2} \left| \frac{\partial \alpha}{\partial Q} \right| (2\hbar \Omega M)^{-1/2} E_0^2 \gg \Omega, T_1^{-1}, T_2^{-1}$, i.e., the analog of Rabi frequency of the two-level oscillator is the highest frequency of the problem. For high-energy femtosecond pulses this condition as well as inequality $\tau_p \ll \Omega^{-1}, T_1, T_2$ leads to the solution (3), (4).

From (3) it follows a strong nonlinear dependence of polarization on the pulse field and the wave equation of the form

$$\frac{\partial E}{\partial z} + \frac{1}{c} \frac{\partial E}{\partial t} \left[1 + \beta c \int_{-\infty}^t \sin \Psi(z, t') dt' \right] = -\beta E \sin \Psi, \quad (5)$$

where $\beta = \frac{\pi}{2} N \left| \frac{\partial \alpha}{\partial Q} \right| (\hbar \Omega / 2 M c^2)^{1/2}$ is the inverse length of induced Raman self-scattering.

The nonlinear equation (5) allows an analytic solution describing in explicit form the evolution of the pulse shape and spectrum [7].

Multiplying (5) by E and integrating it with respect to time one can easily obtain the equation for phase $\Psi(z, t)$ (i.e., for current pulse energy)

$$\frac{\partial \Psi}{\partial z} + \frac{1}{c} \frac{\partial \Psi}{\partial t} \left[1 + \beta c \int_{-\infty}^t \sin \Psi(z, t') dt' \right] = \beta (\cos \Psi - 1). \quad (6)$$

Let us regard the pulse field $E(z, t)$ as a function of the spacial variable z and the phase $\Psi(z, t)$, i.e., $E(z, t) = \tilde{E}(z, \Psi)$. Taking into account that $\frac{\partial E}{\partial z} = \frac{\partial \tilde{E}}{\partial z} + \frac{\partial \tilde{E}}{\partial \Psi} \frac{\partial \Psi}{\partial z}$, $\frac{\partial E}{\partial t} = \frac{\partial \tilde{E}}{\partial \Psi} \frac{\partial \Psi}{\partial t}$, we can finally rewrite Eq. (5) in the following form:

$$\frac{\partial \tilde{E}}{\partial z} + \beta (\cos \Psi - 1) \frac{\partial \tilde{E}}{\partial \Psi} = -\beta \tilde{E} \sin \Psi. \quad (7)$$

This partial differential equation can be easily solved by integrating along characteristics on which

$$\tan \frac{\Psi(z, t)}{2} = \frac{\tan \frac{\Psi_0(\eta)}{2}}{1 + \beta z \tan \frac{\Psi_0(\eta)}{2}}, \quad (8)$$

$$E(z, t) = \frac{E_0(\eta)}{1 + \beta z \left[\sin \Psi_0(\eta) + \frac{\beta z}{2} (1 - \cos \Psi_0(\eta)) \right]}, \quad (9)$$

where $\Psi_0(\eta)$ is the given phase at the boundary of the medium (i.e., the energy of the pulse fraction which has entered the medium) which is connected with the field strength at the boundary by the relation $\Psi_0(\eta) = \left| \frac{\partial \alpha}{\partial Q} \right| (2 \hbar \Omega M)^{-1/2} \int_{-\infty}^{\eta} E_0^2(\eta') d\eta'$. According to (8) the pulse fraction which energy corresponds to the phase $\Psi_0 = 2\pi n$, $n = 1, 2, \dots$ moves through the Raman-active medium under conditions of self-induced transparency, when the energy of the leading part of 2π -pulse absorbed due to Stokes scattering completely returns to the trailing part of the pulse due to anti-Stokes scattering. In this case the spectrum of the leading part of the pulse becomes enriched by long-wavelength components of the field and the spectrum of the trailing part – by the short-wavelength components. This analysis generalizes the results found on the basis of numerical calculations [2].

The characteristics themselves are given implicitly by the expressions

$$\eta + \Phi(z, \eta) = t - z/c, \quad \Phi(z, \eta) = \beta z \int_{-\infty}^{\eta} d\eta' \left[\sin \Psi_0 + \frac{\beta z}{2} (1 - \cos \Psi_0) \right], \quad (10)$$

where $\Phi(z, \eta)$ is the nonlinear delay of the individual parts of the pulse as it moves away from the boundary.

It is convenient to describe the evolution of the pulse shape by an effective frequency $\omega(z, t)$ of the pulse field oscillations. The value $\omega(z, t)$ characterizes the density of field oscillations for different parts of the pulse and its variation as the pulse propagates through the medium. It follows from (10) that if we fix a small fraction of the pulse its duration $\Delta\tau$ at the space point z is connected with that one $\Delta\tau_0$ at the boundary by the relation $\Delta\tau = \Delta\tau_0 (1 + \partial\Phi/\partial\eta)$. The value $\Delta\tau$ determines the field oscillation period at the given space point z , hence the effective frequency of the field oscillations transforms in accordance with

$$\omega(z, t) = \frac{\omega_0(\eta)}{1 + \beta z \left[\sin \Psi_0 + \frac{\beta z}{2} (1 - \cos \Psi_0) \right]}. \quad (11)$$

3 Femtosecond Pulse Evolution in Raman-active Medium

It can be seen from Eqs. (9) and (11) that the changes in the field $E(z, t)$ and the characteristic frequency $\omega(z, t)$ 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 pulse fraction which has entered the medium is small), there are decreases in the field strength and the oscillation frequency: $E(z, t) = E_0(\eta) (1 + \beta z \Psi_0/2)^{-2}$, $\omega(z, t) = \omega_0(\eta) (1 + \beta z \Psi_0/2)^{-2}$. This case corresponds to Stocks scattering. Later, when the phase becomes greater than π 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 (10), we thus observe oscillations of regimes of compression and stretching of the field oscillation periods. 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.

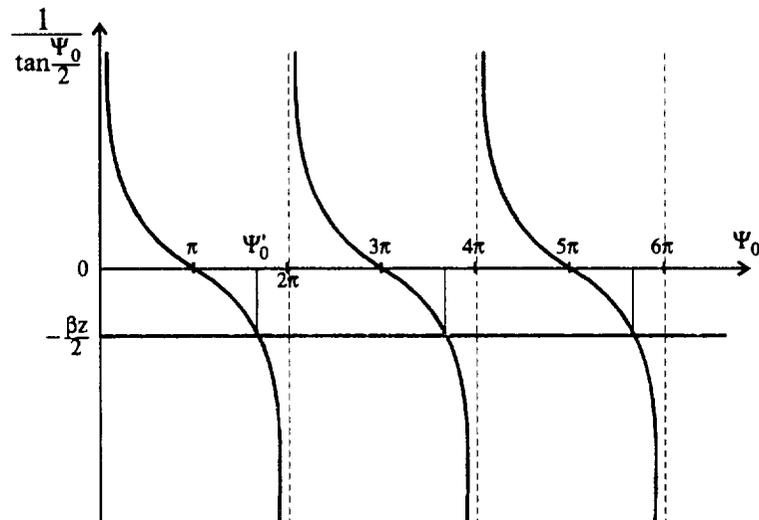


FIG. 1.

A descriptive way to analyze the pulse evolution is provided by Fig. 1. It is seen from (9) and (11) that the nature of pulse transformation is determined by the sign of the expression in brackets of the denominator $\sin \Psi_0 + \frac{\beta z}{2}(1 - \cos \Psi_0) = 2 \sin^2 \frac{\Psi_0}{2} \left[\left(\tan \frac{\Psi_0}{2} \right)^{-1} + \frac{\beta z}{2} \right]$. If $\Psi_0 < \pi$, we have $\left(\tan \frac{\Psi_0}{2} \right)^{-1} > -\frac{\beta z}{2}$ for any distance z from the medium boundary. It means that the energy of such pulses can be only absorbed and the absorption is accompanied by the increase of the field oscillation periods for any pulse fraction (and as a result, by the increase of pulse duration) and by the shift of the pulse spectrum into the low-frequency region due to Stokes scattering.

If $\pi < \Psi_0 < 2\pi$, there are two different regimes of pulse transformation. The leading pulse fraction $0 < \Psi_0 < \Psi'_0 = \arctan(-2/\beta z)$ undergoes energy absorption and stretching of the field oscillation periods and its spectrum becomes enriched with low-frequency components of the field. The field in the trailing pulse fraction amplifies during the pulse propagation, the oscillation periods decrease (that results in the compression of this pulse fraction) and the spectrum is enriched with high-frequency components. For 2π -pulse all the energy concentrates in the trailing edge which is compressed as the pulse propagates through the medium. The same picture takes place for pulse fractions $2\pi n < \Psi_0 < 2\pi(n + 1)$.

Figure 2 shows an example of pulse evolution described by the solution (8)-(11) for $\Psi_0 = 4\pi$. The input pulse represents a two-period fraction of a sine-shaped signal. Figure illustrates the time dependence of the pulse field at different distances from the medium boundary. Thus, the high-energy pulse is divided into the set of compressed powerful 2π -subpulses the maximum field strength and inverse width of which at large distances increase $\sim (\beta z)^2$ according to (8)-(10).

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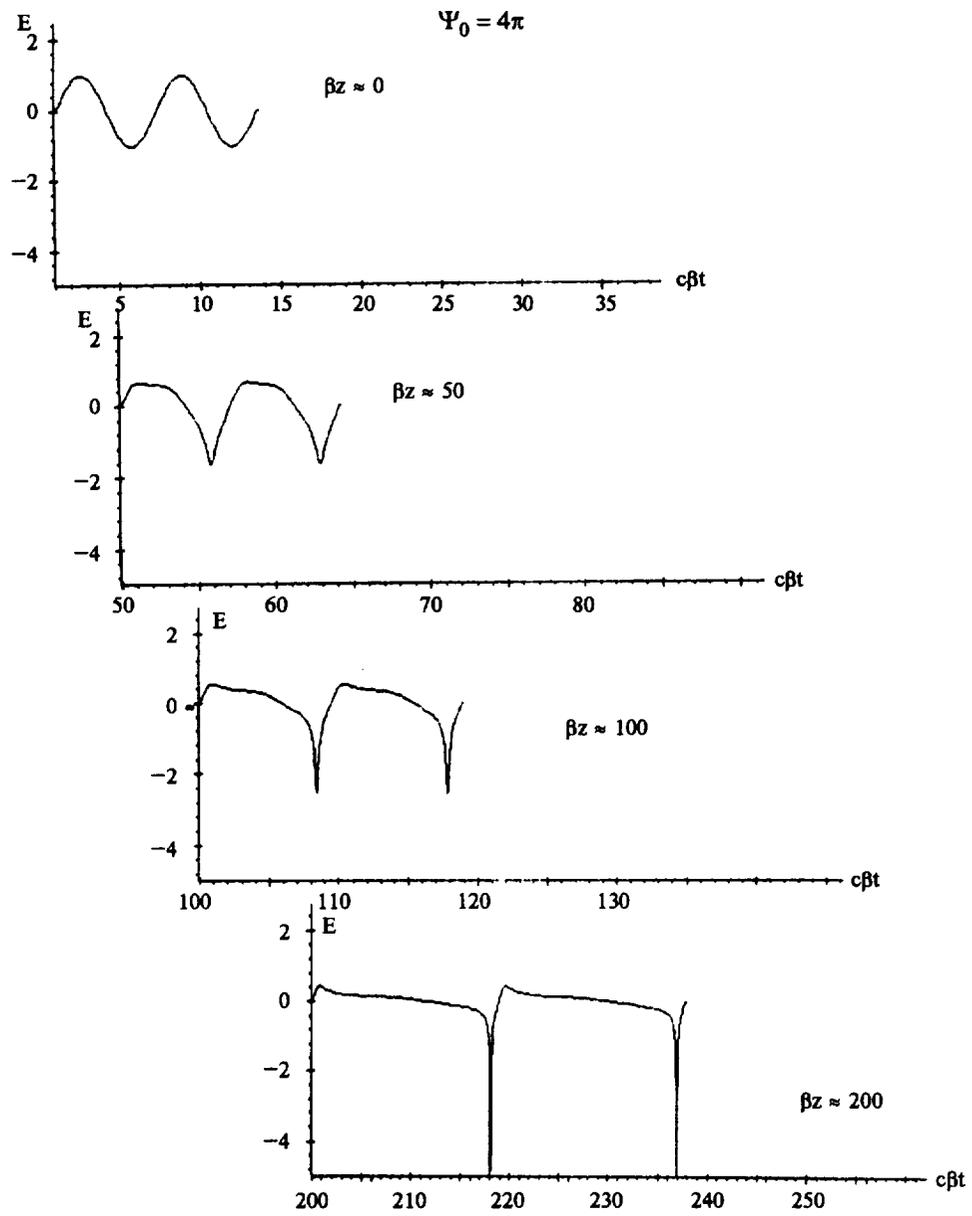


FIG. 2.