Coherent propagation of high-power light pulses through a medium under conditions of two-quantum interaction

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We consider the evolution of high-power laser-radiation pulses under conditions of coherent two-quantum interaction with resonant media. We show that coherent effects are produced in the evolution of such pulses with frequencies ω_L and ω_S, satisfying the equation $\omega_L \pm \omega_S = \omega_{21}$ (ω_{21} is the resonant frequency of the medium), and lead to a breakdown into individual subpulses with subsequent decrease in their duration and increase in power. The effectiveness of such a process depends on the initial ratio of the amplitudes $\mathcal{E}_L$ and $\mathcal{E}_S$.

The singularities of coherent interaction between an absorbing medium and an ultrashort laser pulse, when the doubled pulse carrier frequency 2ω coincides with the transition frequency ω_{21}, was considered in our paper.[1] It was shown that under these conditions pulses of sufficiently high power break up into subpulses whose energy remains practically constant during the course of propagation, but their duration decreases and the power increases. The limiting duration and power of the subpulses are determined by the optical strength of the medium (by cascade and multiphoton ionization).

In this paper we examine the possibility of similar effects in the more general case of two-quantum interaction, such as $\omega_L + \omega_S = \omega_{21}$ (two-photon "absorption" of a pair of pulses with frequencies ω_L and ω_S) or $\omega_L - \omega_S = \omega_{21}$ (Raman interaction of such pulses with the medium). It is assumed, as in[1], that the inhomogeneous broadening is small and that the pulse durations τ_L and τ_S are smaller than the polarization relaxation time T_2. Under these conditions, the system of material equations and Maxwell's equations can be reduced to equations for the amplitudes of the fields of both pulses:

$$\frac{\partial \mathcal{E}_L}{\partial z} + \frac{n}{c}\frac{\partial \mathcal{E}_L}{\partial t} = -k_L \mathcal{E}_S \sin \Psi$$
$$\frac{\partial \mathcal{E}_S}{\partial z} + \frac{n}{c}\frac{\partial \mathcal{E}_S}{\partial t} = \pm k_S \mathcal{E}_L \sin \Psi \, . \tag{1}$$

Here n is the refractive index of the medium, $k_{L,S} = (2\pi\omega_{L,S}/nc)N\tau_{12}$, τ_{12} is the two-photon composite matrix element, and N is the density of the particles of the medium.

The quantity $\Psi(z, t) = (\tau_{12}/2\hbar)\int_{-\infty}^{t}\mathcal{E}_L(z, t')\mathcal{E}_S(z, t')dt'$ is an important characteristic of the coherence and of the nonlinearity of the interaction, and is analogous to the quantity $(\tau_{12}/2\hbar)\int_{-\infty}^{t}\mathcal{E}'^2(z, t')dt'$ for the case $2\omega = \omega_{21}$ (see[1]) and takes the "overlap" of $\mathcal{E}_L$ and $\mathcal{E}_S$ into account.

The signs (−) and (+) in the second equation of (1) correspond to the processes of two-photon "absorption" and Raman interaction, which we shall consider separately.

1. Two-photon absorption

In this case (1) leads to a conservation law for the difference between the numbers of the quanta:

$$\frac{\mathcal{E}_L^2(z, t)}{\omega_L} - \frac{\mathcal{E}_S^2(z, t)}{\omega_S} = \frac{\mathcal{E}_L^2(0, \tau)}{\omega_L} - \frac{\mathcal{E}_S^2(0, \tau)}{\omega_S} \qquad \tau = t - \frac{nz}{c} \, . \tag{2}$$

Pulse propagation in accordance with (1)–(2) was investigated numerically with a computer for a Gaussian initial pulse waveform with various initial values $\theta_0 = \Psi(z = 0, t = \infty)$ and $\omega_S = 0.8\omega_L$. It turned out that for initial pulses with close amplitudes the results differ little from those obtained in[1] for the case of coinciding frequencies $\omega_L = \omega_S$, leading to a breakdown into individual high-power ultrashort subpulses. In the case of strongly differing amplitudes (Fig. 1), this process is much more effective for the pulse with the lower initial energy (see also[2]). Thus, the condition $\omega_L = \omega_S$ is not necessary for the onset of similar coherent effects; moreover, by combining pulses of different amplitudes and frequencies (with the condition $\omega_L + \omega_S = \omega_{21}$ satisfied), one can simplify the conditions for their observation.

2. Raman scattering

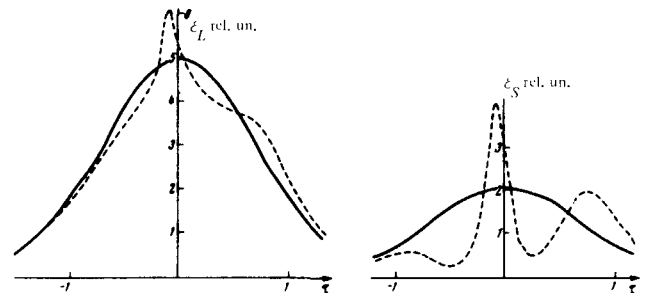In this case there follows from (1) the law of conservation of the sum of the number of quanta (the Manley-Rowe rule):

FIG. 1. Two-photon absorption ($\theta_0 = 4\pi$, initial energy ratio $r = 0.2$): solid curves — $k_2 z = 0$, dashed — $k_2 z = 0.5$.
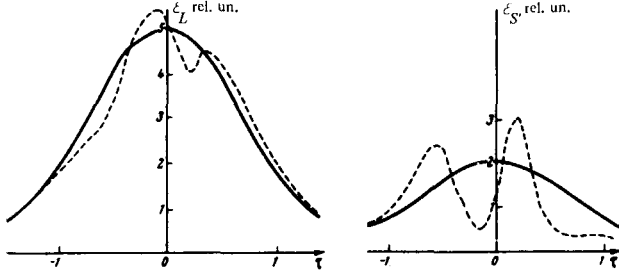
FIG. 2. Raman interaction ($\theta_0 \approx 4\pi$, $r \approx 0.2$): solid lines $- k_2 z = 0$, dashed $- k_2 z = 0.35$.

FIG. 3. Raman interaction ($\theta_0 \approx 4\pi$, $r = 1$): solid lines $- k_2 z = 0$, dashed $- k_2 z = 0.5$.

$$\frac{\mathscr{E}^2_L(z,t)}{\omega_L} + \frac{\mathscr{E}^2_S(z,t)}{\omega_S} = \frac{\mathscr{E}^2_L(0,r)}{\omega_L} + \frac{\mathscr{E}^2_S(0,r)}{\omega_S}, \qquad (3)$$

It follows from this, in particular, that the growth of the individual "bursts" inside the "signal" and "Stokes" pulses is limited by the initial value (3), but a numerical calculation shows that the redistribution of the energy can be appreciable, with formation of abrupt oscillations (Figs. 2 and 3). It is important that the process proceeds here much more effectively for the pulse with the smaller initial energy, leading to an appreciable growth of the power of individual subpulses.

At the same time, a continuous coherent pumping of energy from the "signal" to the "Stokes" pulse and back takes place during the course of propagation. By way of example, let us consider the Raman interaction in molecular nitrogen using laser pulses and its Stokes component with duration $\approx 10^{-8}$ sec and power $\approx 10^8 - 10^9$ W/cm^2, ($\theta_0 = 10 - 100$), with a Raman-scattering cross section $\sigma \approx 10^{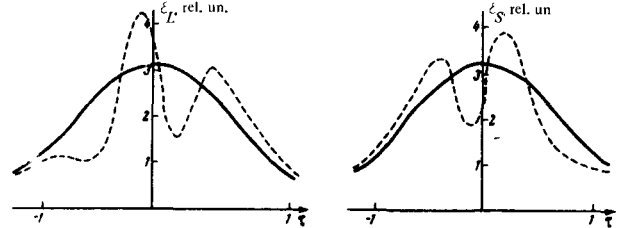-31}$ cm^2 ($r_{12} = 10^{-25}$ cm^3). At a particle density N on the order of $\sim 10^{17}$ cm^{-3}, the characteristic distance over which the pulse subdivision takes place is $z_0 \approx k_{L,S}^{-1} \approx 10^2$ cm. Thus, the use of two pulses with different amplitudes and frequencies uncovers greater possibilities for the observation of the indicated coherent effect and for their practical application.

We note also that similar effects should take place also in the case of an appreciable inhomogeneous width, inasmuch as the physical causes of the phenomenon do not depend on the nature of the emission line. In addition, these effects can appear in the case when the pulse duration is $\tau \approx T_2$, although their character will be less strongly pronounced. A confirmation of these assumptions is provided by a numerical calculation performed by us for the case of two-photon interaction of light with a semiconductor. [3]

[1] I. A. Poluéktov, Yu. M. Popov, and V. S. Roĭtberg, ZhETF Pis. Red. 18, 638 (1973) [JETP Lett. 18, 373 (1973)].
[2] N. Tan-no, K. Yokoto, and H. Inaba, Phys. Rev. Lett. 29, 1211 (1972).
[3] T. L. Gvardzhaladze, I. A. Poluéktov, and V. S. Roĭtberg. Kratkie soobshcheniya po fizike, FIAN SSSR, No. 3, 7, 1973.