Saturation of coherent amplification of ultrashort pulses in an inverted medium

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The propagation of an ultrashort laser pulse in a population-inverted medium is investigated taking into account the local crystal field and admixing of distant atomic levels. It is shown that these effects lead to saturation of amplification with conversion of the pulse into a unipolar dissipative soliton.

In the last few years several theoretical studies of the interaction of ultrashort pulses, which include up to one period of the light oscillations, with matter have been published.[1–3] Such investigations are of interest primarily because of the possibility of obtaining such pulses under experimental conditions.[4,5] They are also of purely theoretical interest, because it is necessary to study the system of constitutive equations and Maxwell's equations without using the approximation of slowly varying amplitudes and phases. In Refs. 1 and 3 the corresponding solutions for the electric field were obtained in the form of unipolar solitons; in addition, the coherent amplification of ultrashort pulses by an inverted, nonresonant, two-level medium was studied. It was shown that in the latter case, in addition to the compression of the pulse, the photons are transferred into the "blue" region of the spectrum. However, increasing the frequency of the field results in efficient admixing of distant atomic levels, scattering and absorption on which should impede coherent amplification. In addition, in sufficiently dense media the local electric field acting on each atom is different from the external field determined by Maxwell's equations.[6] In the case of cubic crystals the local electric field is determined by the Lorentz correction.[6] This difference arises from the fact that an electric field is induced at the location of each atom by the dipole moments of the surrounding atoms (dipole–dipole interaction). The dipole moment arising in the superposition state generates in the field of each atom an additional electric field, which gives rise to a Stark shift of the transition frequency in proportion to the population inversion.[7–9]

Summarizing what was said above, we write the system of Maxwell–Bloch equations for a two-level medium in the form

$$\frac{\partial u}{\partial t} = -(\omega_0 - Jw)v, \quad \frac{\partial v}{\partial t} = (\omega_0 - Jw)u + \Omega w,$$

$$\frac{\partial w}{\partial t} = -\Omega v,$$

$$\left(c^2 \nabla^2 - \frac{\partial^2}{\partial t^2} - \gamma \frac{\partial}{\partial t}\right)\Omega = \frac{8\pi d^2 n}{\hbar}\frac{\partial^2 u}{\partial t^2}. \tag{1}$$

Here $u=\langle\sigma_x\rangle/2$, $v=\langle\sigma_y\rangle/2$, $w=\langle\sigma_z\rangle/2$, σ_x, σ_y, and σ_z are Pauli operators, $\langle...\rangle$ denotes a quantum average, ω_0 and d are the frequency and dipole moment of the atomic transition, respectively; n is the concentration of optically active atoms, $\hbar$ is Planck's constant, c is the velocity of light, J is the effective dipole–dipole coupling constant (in the case of cubic crystals $J=4\pi d^2n/3\hbar$); $\Omega=dE/\hbar$, where E is the projection of the electric field onto the direction of the dipole moment, and γ is a phenomenological constant which accounts for the losses associated with absorption and scattering by the admixed atomic transitions.[6]

From the constitutive equations of system (1) it follows that

$$\frac{\partial^2 u}{\partial t^2}=-(\omega_0-Jw)[(\omega_0-Jw)u+\Omega w]-\frac{J}{\Omega}\left(\frac{\partial w}{\partial t}\right)^2, \tag{2}$$

$$\frac{\partial w}{\partial t}=\frac{\Omega}{\omega_0-Jw}\frac{\partial u}{\partial t}. \tag{3}$$

Following Refs. 1–3, we assume that $\Omega\ll\omega_0\ll J$. We can then write Eq. (2) approximately in the form

$$\frac{\partial^2 u}{\partial t^2}=-(\omega_0-Jw)\Omega w-\frac{J}{\Omega}\left(\frac{\partial w}{\partial t}\right)^2. \tag{4}$$

Equations (3) and (4) can be integrated with an arbitrary function $\Omega(\vec{r},t)$:

$$w=w_\infty\cos\theta,\quad \theta=\int_{-\infty}^{t}\Omega(\vec{r},t')dt',$$

$$\frac{\partial u}{\partial t}=-\omega_0 w_\infty\sin\theta+\frac{1}{2}Jw_\infty^2\sin 2\theta. \tag{5}$$

Here w_∞ is the inversion of an atom before the action of the electromagnetic pulse. Using Eq. (5) and the last equation in system (1), we find

$$\left(c^2\nabla^2-\frac{\partial^2}{\partial t^2}-\gamma\frac{\partial}{\partial t}\right)\theta=-\frac{8\pi d^2 n}{\hbar}\left(\omega_0 w_\infty\sin\theta-\frac{1}{2}Jw_\infty^2\sin 2\theta\right). \tag{6}$$

An analogous equation is encountered in the theory of the motion of a domain wall in a ferromagnet in an external magnetic field.[10] Assuming that the pulse propagates along the z axis in a steady fashion with a velocity v_p, we obtain the corresponding solutions for the field and population inversion as follows:

$$E=\frac{\hbar}{d\tau}\operatorname{sech}\frac{t-z/v_p}{\tau}, \tag{7}$$

$$w=-w_\infty\tanh\frac{t-z/v_p}{\tau}, \tag{8}$$

where $\tau^{-1}=8\pi d^2 n\omega_0 w_\infty/\hbar\gamma$, $v_p=c/\sqrt{1+q}$, $q=\hbar J\gamma^2/(8\pi d^2\omega_0^2 n)$ (for a cubic crystal we have $q=\gamma^2/6\omega_0^2$).

Solution (7), (8) describes a stationary localized electromagnetic pulse, which as it propagates removes energy in the initially inverted medium and dissipates it as a result of losses on the admixed atomic transitions. As expected, the area of the pulse is equal to π. The velocity, amplitude, and duration of the pulse (7) are not related to one another, as in integrable soliton models, but rather they are determined solely by the parameters of the medium. Such formations are sometimes called dissipative solitons (dissipative structures). Thus the increase in the frequency of the photons in ultrashort pulses[1] should reach saturation point and be replaced by the stationary state [Eqs. (7) and (8)]. From Eqs. (7) and (8) it is evident that as $\gamma \to 0$, the increase in the compression and amplification is unbounded. The condition of applicability of this approximation, $\Omega \ll \omega_0$, shows that $\tau^{-1} \gg \omega_0$ or $\gamma \ll 4\pi d^2 n/\hbar$ ($w_\infty = 1/2$). After substituting here $n \sim 10^{17}$ cm^{-3} and $d \sim 5 \times 10^{-18}$ absolute units,[1] the last condition becomes $\gamma \ll 3 \times 10^{10}$ s^{-1}. Setting $\omega_0 \sim 10^{14}$ s^{-1}, we find that for cubic crystals $q \ll 10^{-8}$, and with high accuracy $v_p = c$. In addition, for $\gamma \simeq 10^9$ s^{-1} $\tau \sim 1$ fs, $E \sim 10^8$ V/cm, and the intensity $I \sim 10^{13}$ W/cm^2. In Ref. 1 it is shown that for the adopted parameters the frequency of photons in ultrashort pulses is $\omega(z) \gg \omega_0$ at distances $z \gg 1$ cm. The spectral width of the pulse (7) is equal, in order of magnitude, to τ^{-1}. If $\omega_0 \sim 10^{14}$ s^{-1} and $\tau^{-1} \sim 10^{15}$ s^{-1}, then saturation of amplification can occur at distances $z \simeq 10$ cm, which corresponds to the propagation time $\tau \simeq 3 \times 10^{-10}$ s. This value is much shorter than the characteristic spontaneous emission times. Saturation of amplification of ultrashort pulses can therefore be easily observed experimentally. By measuring the width and amplitude of a dissipative soliton at the exit from the sample it is possible to extract information about the parameters of the medium; for example, it is possible to determine γ and J.

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