COHERENCE EFFECTS IN THE PROPAGATION OF AN ULTRASHORT LIGHT PULSE IN A MEDIUM WITH TWO-PHOTON RESONANCE ABSORPTION

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The two-photon interaction of an ultrashort light pulse with a medium is examined. It is assumed that the duration of the pulse is shorter than the characteristic relaxation times of the substance. It is shown that under certain conditions the pulse passes through the medium without absorption, with a speed which can be quite different from the speed of light. The form of the field of such a pulse is found.

1. Lasers can now be used to produce powerful light pulses of duration $\tau \sim 10^{-12}$–10^{-13} sec, and methods have been proposed for the formation of pulses with duration $\tau \sim 10^{-13}$–10^{-14} sec.[1] Such pulses allow the observation of a number of new nonlinear effects, caused by coherent interaction of the light with matter. A paper by McCall and Hahn[2] deals with the increase of transparency of an absorbing medium for ultrashort light pulses when there is one-photon resonance absorption. This sort of transparency has nothing in common with the penetration of media owing to saturation, and appears only when the duration τ of the light pulse is smaller than the characteristic time T_2 of the substance for loss of coherence. The physical cause of this effect is that as the pulse is propagated the inversion between the working levels oscillates with a frequency Ω proportional to the instantaneous value of the field $\mathscr{E}(z, t)$. If the phase $\int_{-\infty}^{t} \Omega(z, t)dt$ at the end of the pulse ($t = \infty$) takes the value 2π, the pulse is propagated in the medium without absorption. In this case the pulse loses energy at the leading front, which coherently excites the molecules of the medium, and receives it at the rear front as the result of reemission by the molecules. Owing to this the speed of propagation of the pulse is less than the speed of light, and may differ from it by several orders of magnitude.

In the present paper we consider the influence of coherence effects on the propagation of an ultrashort light pulse ($\tau < T_2$) in a medium with two-photon resonance absorption. The treatment is based on the study of a system of abbreviated equations for the field and the material variables of the substance. For the case when there is only two-photon absorption these equations, derived from Maxwell's equations and the equation for the density matrix, are of the form[3,4]

$$\frac{\partial \mathscr{E}}{\partial z} + \frac{1}{c}\frac{\partial \mathscr{E}}{\partial t} = -\frac{2\pi\omega}{c}\mathscr{E}\langle P_2\rangle,$$

$$\frac{\partial \varphi}{\partial z} + \frac{1}{c}\frac{\partial \varphi}{\partial t} = -\frac{2\pi\omega}{c}\left\langle P_1 - \frac{k_{22}-k_{11}}{2}N\right\rangle$$

$$\frac{\partial P_1}{\partial t} = -\left(\Delta\omega + 2\dot{\varphi} + \frac{k_{22}-k_{11}}{4\hbar}\mathscr{E}^2\right)P_2,$$

$$\frac{\partial P_2}{\partial t} = \left(\Delta\omega + 2\dot{\varphi} + \frac{k_{22}-k_{11}}{4\hbar}\mathscr{E}^2\right)P_1 + \frac{|k_{12}|^2}{2\hbar}\mathscr{E}^2(n+N_0),$$

$$\frac{\partial n}{\partial t} = -\frac{\mathscr{E}^2}{2\hbar}P_2, \quad n = P_1 = P_2 = 0 \quad \text{при } t = -\infty.$$

In the derivation of this system it is assumed that the electromagnetic field of the pulse is given in the form $E(z, t) = \mathscr{E}(z, t)\cos[\omega t - kz + \varphi(z, t)]$, where $\mathscr{E}$ and φ are slowly varying amplitude and phase functions, ω is the frequency, k is the wave vector, and z is the direction of propagation of the pulse. The sign $\langle \ldots \rangle$ denotes averaging over the spectral distribution $g(\Delta\omega)$ of the molecules, $\Delta\omega = 2\omega - \omega_{12}$ is the resonance defect, $\mathscr{E}P_2$ and $\mathscr{E}P_1$ are the active and reactive components of the polarization of the medium at frequency ω, and n is the deviation of the difference of the populations of the working levels from its initial value. It is assumed that the dipole transition between the working levels is forbidden,

$$k_{11} = \frac{2}{\hbar}\sum_{k\geqslant 3}\frac{|\mu_{1k}|^2\omega_{k1}}{\omega_{k1}^2-\omega^2}, \quad k_{22} = \frac{2}{\hbar}\sum_{k\geqslant 3}\frac{|\mu_{2k}|^2\omega_{k2}}{\omega_{k2}^2-\omega^2},$$

$$k_{12} = k_{21}^* = \frac{1}{\hbar}\sum_{k\geqslant 3}\frac{\mu_{1k}\mu_{k2}}{\omega_{k2}+\omega},$$

where μ_{ik} and ω_{ik} are the dipole matrix element and the frequency of the transition between levels i and k of the molecule.

2. Let us consider the propagation of a pulse in a substance with a uniformly broadened emission line [$g(\Delta\omega) \sim \delta(\Delta\omega)$] at exact resonance ($\Delta\omega = 0$). In this case the system (1) takes the form

$$\frac{\partial \mathscr{E}}{\partial z} = -\frac{1}{c}\frac{\partial \mathscr{E}}{\partial t} - \frac{2\pi\omega}{c}P_2\mathscr{E},$$

$$\frac{\partial \varphi}{\partial z} = -\frac{1}{c}\frac{\partial \varphi}{\partial t} - \frac{2\pi\omega}{c}\left(P_1 - \frac{k_{22}-k_{11}}{2}n\right),$$

$$\frac{\partial P_1}{\partial t} = -\left(2\dot{\varphi} + \frac{k_{22}-k_{11}}{4\hbar}\mathscr{E}^2\right)P_2,$$

$$\frac{\partial P_2}{\partial t} = \left(2\dot{\varphi} + \frac{k_{22}-k_{11}}{4\hbar}\mathscr{E}^2\right)P_1 + \frac{|k_{12}|^2}{2\hbar}(n+N_0),$$

$$\frac{\partial n}{\partial t} = -\frac{\mathscr{E}^2}{2\hbar}P_2. \qquad (2)$$

From the system (2) one can find the general expression for the quantity $x = 2\dot{\varphi}$, i.e., the change of the carrier frequency of the pulse. Differentiating the

second equation in (2) with respect to the time, we get

$$\frac{\partial x}{\partial z}+\frac{1}{c}\frac{\partial x}{\partial t}=-\frac{4\pi\omega}{c}\frac{\partial}{\partial t}\left(P_1-\frac{k_{22}-k_{11}}{2}n\right).$$

When we next multiply the equation for n by $(k_{11} - k_{22})/2$ and combine it with the equation for P_1, we have

$$\frac{\partial}{\partial t}\left(P_1-\frac{k_{22}-k_{11}}{2}n\right)=-xP_2,$$

that is,

$$\frac{\partial x}{\partial z}+\frac{1}{c}\frac{\partial x}{\partial t}=\frac{4\pi\omega}{c}xP_2. \tag{3}$$

Comparing (3) with the first equation of the system (2), we get

$$x\left(\frac{\partial\mathscr{E}^2}{\partial z}+\frac{1}{c}\frac{\partial\mathscr{E}^2}{\partial t}\right)=-\mathscr{E}^2\left(\frac{\partial x}{\partial z}+\frac{1}{c}\frac{\partial x}{\partial t}\right). \tag{4}$$

From (4) we get the relation

$$x(z,t)=f(z-ct)/\mathscr{E}^2(z,t), \tag{5}$$

where $f(z - ct)$ is an arbitrary function. From physical considerations it is obvious that $f(z - ct) = 0$ $[\mathscr{E}(z, \pm\infty) = 0]$, i.e., $x = 0$ for all z and t. Owing to this the material equations of the system (2) admit an exact solution for arbitrary form of the field $\mathscr{E}(z, t)$. Introducing the quantity

$$\psi(z,t)=\frac{\sqrt{(k_{22}-k_{11})^2+4|k_{12}|^2}}{4\hbar}\int_{-\infty}^{t}\mathscr{E}^2(z,t)\,dt, \tag{6}$$

which defines the phase angle of the material variables, and using the initial conditions, we have from (2)

$$P_1=\frac{2(k_{22}-k_{11})|k_{12}|^2}{(k_{22}-k_{11})^2+4|k_{12}|^2}N_0(\cos\psi-1),$$

$$P_2=-\frac{2|k_{12}|^2}{\sqrt{(k_{22}-k_{11})^2+4|k_{12}|^2}}N_0\sin\psi,$$

$$n=\frac{4|k_{12}|^2}{(k_{22}-k_{11})^2+4|k_{12}|^2}N_0(\cos\psi-1). \tag{7}$$

The quantity $\theta(z) = \psi(z, \infty)$, which describes the phase change of the material variables at the point z during the action of the pulse, is proportional to the total energy of the pulse at the given point. Using the first equation of the system (2) and the quantity P_2 from (7), we find an equation for θ:

$$\frac{d\theta}{dz}=-\alpha(1-\cos\theta),\quad \alpha=\frac{8\pi\omega}{c}\frac{|k_{12}|^2}{\sqrt{(k_{22}-k_{11})^2+4|k_{12}|^2}}. \tag{8}$$

For small θ

$$\frac{d\theta}{dz}=-\frac{\alpha}{2}\theta^2, \tag{8a}$$

which corresponds to ordinary two-photon absorption of light in the medium. For $\theta = 2\pi m$, where m is an integer, $d\theta/dz = 0$, and the pulse is propagated without absorption. If $2\pi m < \theta < 2\pi(m + 1)$, the energy of the pulse will be absorbed up to the value $\theta = 2\pi m$, and thereafter the pulse will be propagated without absorption.

3. Let us now proceed to find the form of the stationary pulse. We introduce the variable $t' = t - z/v$, where v is the group velocity of propagation of the pulse $(v < c)$. The equation for $\mathscr{E}(t')$ then takes the form

$$\frac{d\mathscr{E}^2}{dt'}=b\mathscr{E}^2\sin\frac{\sqrt{(k_{22}-k_{11})^2+4|k_{12}|^2}}{4\hbar}\int_{-\infty}^{t'}\mathscr{E}^2\,dt',$$

$$b=\frac{8\pi\omega v}{c-v}\frac{|k_{12}|^2N_0}{\sqrt{(k_{22}-k_{11})^2+4|k_{12}|^2}}. \tag{9}$$

Integrating (9), we get

$$\mathscr{E}^2(t')=a\left(\cos\frac{\sqrt{(k_{22}-k_{11})^2+4|k_{12}|^2}}{4\hbar}\int_{-\infty}^{t'}\mathscr{E}^2\,dt'-1\right),$$

$$a=\frac{32\pi\omega v\hbar}{c-v}\frac{|k_{12}|^2N_0}{[(k_{22}-k_{11})^2+4|k_{12}|^2]^{1/2}}$$

so that

$$\frac{d}{dt'}\mathscr{E}^2(t')=\pm b\mathscr{E}^2\sqrt{\frac{2\mathscr{E}^2}{a}-\frac{\mathscr{E}^4}{a^2}}. \tag{10}$$

The solution of (10) is a pulse of the Lorentz shape:

$$\mathscr{E}^2(t')=\frac{2a}{1+(bt')^2}. \tag{11}$$

According to (11) the speed of the pulse is connected with its duration by the relation

$$b(v)=\frac{1}{\tau},\quad v=c\Big/\left[1+\frac{8\pi|k_{12}|^2\omega\tau N_0}{\sqrt{(k_{22}-k_{11})^2+4|k_{12}|^2}}\right]. \tag{12}$$

It is easily verified that the pulse found in (8) is a 2π pulse and is propagated in the medium without absorption.

Let us now discuss briefly the case of a nonuniformly broadened line of the material. The presence of a Stark effect shift $(k_{11} - k_{22})\mathscr{E}^2/4\hbar$ of the working levels destroys the symmetry of the system of material equations with respect to $\Delta\omega$. The result is that the frequency shift $x = 2\dot{\varphi}$ becomes time-dependent. In fact, by combining the equations for P_1, P_2, and n in (2) we can derive the following relation

$$\frac{d}{dt'}\left\langle P_1-\frac{k_{22}-k_{11}}{2}n\right\rangle=-\langle\Delta\omega P_2\rangle-x\langle P_2\rangle. \tag{13}$$

At the same time the equations for the field $\mathscr{E}$ and for x take the forms

$$\frac{d}{dt'}\mathscr{E}^2=\frac{4\pi\omega v}{c-v}\mathscr{E}^2\langle P_2\rangle,$$

$$x=\frac{4\pi\omega v}{c-v}\left\langle P_1-\frac{k_{22}-k_{11}}{2}n\right\rangle. \tag{14}$$

From the last equation of the system (2) it follows that

$$\mathscr{E}^2\langle P_2\rangle=-2\hbar\frac{d\langle n\rangle}{dt'},\quad \langle\Delta\omega P_2\rangle=-\frac{2\hbar}{\mathscr{E}^2}\frac{d}{dt'}\langle\Delta\omega n\rangle.$$

The result is that

$$\frac{d\mathscr{E}^2}{dt'}=-\frac{8\pi\omega v\hbar}{c-v}\frac{d\langle n\rangle}{dt'}$$

$$\mathscr{E}^2=-\frac{8\pi\omega v\hbar}{c-v}\langle n\rangle,\quad \langle\Delta\omega P_2\rangle=\left(\frac{d}{dt'}\langle\Delta\omega n\rangle\right)\Big/\frac{d}{dt'}\langle n\rangle. \tag{15}$$

Combining Eqs. (10), (11), and (12), we get

$$\frac{dx}{dt'}+\frac{x}{\langle n\rangle}\frac{d\langle n\rangle}{dt'}+\frac{1}{\langle n\rangle}\frac{d\langle\Delta\omega n\rangle}{dt'}=0. \tag{16}$$

The exact solution of (16), satisfying the initial conditions, is

$$x = -\frac{\langle \Delta\omega n \rangle}{\langle n \rangle}. \quad (17)$$

Since n is not symmetric with respect to $\Delta\omega$, the quantity x in general has a significant time dependence; the system (2) becomes much more complicated, and it is now evidently possible to determine the quantities P_1, P_2, n, x, and $\mathcal{E}^2$ only by means of numerical integration.

In conclusion we note that the effect described here can also occur for two-photon interzone absorption in a semiconductor at low temperatures, when the relaxation-time times are rather large. Of course, a quantitative calculation of the effect in a semiconductor requires a special approach, taking into account specific features (for example, the line shape) of the semiconductor.

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