Interaction of ultrashort light pulses with a thin layer of surface atoms in two-photon resonance

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The problem of the traversal of ultrashort light pulses through a thin layer of surface resonance atoms under conditions of two-quantum absorption is solved. Analytic solutions are given and an analog of the McCall-Hahn "energy theorem" for the optical radiation passing through or reflected by the resonance layer is formulated for the case when phase modulation is absent. An anomalous traversal of ultrashort light pulses through the resonance layer occurs for large values of the nonlinearity parameter.

1. INTRODUCTION

At the present time processes arising in propagation of ultrashort light pulses (USP) under conditions of two-quantum absorption in an extended resonance medium whose size exceeds the characteristic nonlinear-absorption length have been well studied.[1–6] In particular, it was established in Ref. 1 that under the condition $\tau_i < T_2$, where τ_i is the length of the USP, and T_2 is the polarization relaxation time, it is possible to form solitary 2π-pulses that are the analogs of single-soliton solutions of the problem of self-induced transparency (SIT) of McCall and Hahn in the theory of the one-quantum resonance.[7] The distinctive features of the mechanism of nonlinearity in the case of two-quantum absorption are manifested in the existence of the so-called energy theorem and lead to analytic solutions for the energy of the USP in the case of homogeneous broadening of the resonance transition line. This theorem describes the evolution of the energy of the USP light in an extended resonance medium under conditions of two-photon resonance and is the analog of the "area theorem" of McCall and Hahn in the one quantum case.

Interaction processes of USP light under resonance conditions in another limiting case, when the nonlinear medium is a thin layer of resonant surface atoms whose thickness is considerably less than the wavelength of the light[8–10] are no less interesting. In the case of one-quantum resonance, a similar problem can be solved exactly by use of the method of the inverse scattering problem.[8] In this case the interaction of light with the thin layer of resonance atoms reduces to the problem of scattering on a point potential, taking into account the presence of a reflected wave.

It is well known that under these conditions the nonlinear connection between the field of the light wave passing through the surface layer and the optical properties of the resonance medium play an important role. Because of the coherent character of the interaction of the USP with the resonance layer, it is possible to have nonunique solutions for the "area" of the optical pulse passing through the layer.[9] We note that this nonuniqueness of the solutions in the case of the quasi-stationary action of light is manifested in the phenomenon of optical bistability in the absence of a cavity.[10]

Therefore, it is of interest to consider coherent interaction of USP light with a thin surface layer of resonance atoms under conditions of two-quantum absorption. The simplicity of the choice of a physical model together with the specific mechanism of nonlinear interaction of light pulses with a resonance medium at the two-quantum resonance leads to analytic solutions having a sufficiently general character without resorting to the method of the inverse scattering problem. In particular, in the absence of phase modulation it is possible to "track" analytically the energy of the optical radiation passing through and reflected from a surface layer.

2. BASIC EQUATIONS AND METHOD OF SOLUTION

The system of equations for the field and the medium under conditions of two-quantum interaction can be obtained by traditional methods. The difference of our boundary-value problem from the problem of transmission of USP light in an extended resonance medium lies only in the use of appropriate electrodynamic conditions connecting the fields on the boundary between the two media rather than the Maxwell equations.

We consider a thin layer of resonance atoms situated on the boundary of two linear media ($z=0$) with dielectric constants ε_a and ε_b. We limit the detailed analysis to TE-waves only.

Let the electric field of a light wave incident from the first medium on the interface be given by the expression

$$E_y(x, z, t)=E_0(x, z, t)\exp(i\omega t-ik_x^a x-ik_z^a z), \tag{1}$$

where $E_0(x, z, t)$ is a slowly varying amplitude, k_x^a and k_z^a are the components of the wave vector in the first medium, and ω is the frequency of the incident light wave.

The reflected and transmitted wave are respectively given by

$$\begin{aligned} E_y^r(x, z, t)&=E_r(x, z, t)\exp(i\omega t-ik_x^a x+ik_z^a z),\\ E_y^{tr}(x, z, t)&=E(x, z, t)\exp(i\omega t-ik_x^b x-ik_z^b z). \end{aligned} \tag{2}$$

The boundary conditions describing the relation between the amplitude of these waves are

$$\begin{aligned} E_y(x, 0+, t)-E_y(x, 0-, t)&=0,\\ B_z(x, 0+, t)-B_z(x, 0-, t)&=0,\\ B_x(x,0+,t)-B_x(x,0-,t)&=\frac{4\pi}{c}\dot{P}_y(x,t), \end{aligned} \tag{3}$$

where $P_y(x,t)$ is the surface polarization density at the

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boundary of the media. Using boundary conditions (3), it is easy to express the slowly varying amplitudes of the electric fields E_0, E_r, and E of the incident, reflected, and transmitted light waves, respectively, in terms of the amplitude of the polarization P_F on the boundary of the media at $z = 0$:

$$E=\frac{2A}{A+B}E_0-i\frac{4\pi\omega}{c(A+B)}P_F,\qquad E_r=\frac{A-B}{A+B}E_0-i\frac{4\pi\omega}{c(A+B)}P_F, \tag{4}$$

where A and B are the Fresnel coefficients ($A = \varepsilon_a^{1/2}\cos\theta_a$, $B = \varepsilon_b^{1/2}\cos\theta_b$, and θ_a and θ_b are the angles of incidence and refraction).

The amplitude of the polarization P_F in the right-hand side of Eqs. (4) can be obtained by usual methods in the theory of the two-quantum resonance (see, e.g., Ref. 6):

$$P_F=N_0\{\langle P\rangle E^*+[k_0+(k_{11}+k_{22})/2+(k_{22}-k_{11})\langle n\rangle/2]E\}. \tag{5}$$

Here, contributions of the resonance and nonresonance types are taken into account, with n the inverted population of energy levels, referred to a single resonance atom, and N_0 is the surface density of resonance atoms in the layer. The coefficients k_{11} and k_{22} describe the Stark shift of resonance energy levels.

$$k_{ii}=\frac{2}{\hbar}\sum_j\frac{|d_{ij}|^2\omega_{ji}}{\omega_{ji}{}^2-\omega^2},\quad i=1,2;$$

k_{21} is the matrix element of the two-photon transition:

$$k_{21}=\frac{1}{\hbar}\sum_j\frac{d_{2j}d_{j1}}{\omega_{j2}+\omega};$$

the coefficient k_0 takes into account the contribution to the polarization from nonresonance impurities, and the angular brackets indicate averaging over the frequency spread within the limits of the inhomogeneously broadened line of a spectral transition:

$$\langle\ldots\rangle=\int g(\Delta\omega)(\ldots)d\Delta\omega,$$

($\Delta\omega = 2\omega - \omega_{21}$ is the resonance defect; the function $g(\Delta\omega)$ describes the form of the inhomogeneously broadened line), and the component of the Bloch vector P satisfies the following equations:

$$\dot P+i\Delta\omega P+i\frac{k_{22}-k_{11}}{4\hbar}|E|^2P=i\frac{|k_{21}|^2}{2\hbar}E^2n,\qquad \dot n=(P^*E^2-PE^{*2})/4i\hbar. \tag{6}$$

We note that, in writing the system (6), we used the approximation of ultrashort light pulses and discarded terms describing the processes of relaxation of polarization and population inversion.

In the following we consider the passage of USP of light through a resonance layer with a homogeneously broadened line of radiation $[g(\Delta\omega)\sim\delta(\Delta\omega)]$ at exact resonance ($\Delta\omega = 0$). In this case the basic system of equations has the form

$$E=\frac{2A}{A+B}E_0-i\frac{4\pi\omega}{c(A+B)}P_F,\quad E_r=\frac{A-B}{A+B}E_0-i\frac{4\pi\omega}{c(A+B)}P_F,$$
$$P_F=N_0\left\{PE^*+\left[k_0+\frac{k_{11}+k_{22}}{2}+\frac{k_{22}-k_{11}}{2}n\right]E\right\},$$
$$\dot P+i\frac{k_{22}-k_{11}}{4\hbar}|E|^2P=i\frac{|k_{21}|^2}{2\hbar}E^2n,\quad \dot n=(P^*E^2-PE^{*2})/4i\hbar. \tag{7}$$

It is necessary to note the following in connection with the use of Eqs. (7). In solving the analogous problem for conditions of one-quantum resonance, the nonresonant terms in the polarization are usually discarded because they are small (of order $\Delta\omega/\omega$, where $\Delta\omega$ is the deviation from resonance) in comparison with the resonance terms. In the case of two-quantum resonance, it follows from Eqs. (5) and (6) that the nonresonance part of the polarization is of order $N_0k_{11}E$ and is linear in the field, while the resonant part of the polarization is $N_0(|k_{21}|^2/\hbar\Delta\omega)E^3$ and is nonlinear in the field. As a result we obtain for the ratio of the resonant to nonresonant contributions

$$\frac{P_r}{P_{\rm nr}}\sim\frac{|k_{21}|^2E^2}{d^2}\frac{\omega}{\Delta\omega}\sim\frac{\omega}{\Delta\omega}\left(\frac{E}{E_a}\right)^2$$

where E_a is a characteristic intra-atom field.

However, in our case of a homogeneously broadened line and exact resonance, $\Delta\omega$ should be taken to mean the Stark frequency shift which, in order of magnitude, is

$$\Delta\omega_E\approx\frac{k_{22}-k_{11}}{4\hbar}|E|^2\sim\frac{d^2|E|^2}{(\hbar\omega)^2}\omega\sim\omega\left(\frac{E}{E_a}\right)^2.$$

Thus, the resonance and nonresonance terms are of the same order.

It follows from the boundary conditions (3) that the amplitudes of the field of the transmitted and reflected waves are connected by the simple relationship

$$E-E_r=E_0. \tag{8}$$

Therefore, we consider in detail the characteristics of the field of the transmitted wave. We take for the amplitude of the electric field of the USP light incident on the resonance layer

$$E_0=\mathscr{E}_0(t)\exp[i\varphi_0(t)].$$

We seek then the slowly varying amplitudes of the field of the transmitted wave and the component of the Bloch vector in a form that allows phase modulation

$$E=\mathscr{E}(t)\exp[i\varphi(t)],\quad P=[U(t)+iV(t)]\exp[2i\varphi(t)].$$

We obtain after separation of the variables

$$\begin{gathered}(1-\beta V)\mathscr{E}=F\mathscr{E}_0\cos(\varphi_0-\varphi),\\ [\xi_0+\beta(U+{}^1/_2(k_{22}-k_{11})\delta n)]\mathscr{E}=F\mathscr{E}_0\sin(\varphi_0-\varphi),\\ \dot U-(4\hbar)^{-1}(k_{22}-k_{11})\mathscr{E}^2V-2\dot\varphi V=0,\\ \dot V+(4\hbar)^{-1}(k_{22}-k_{11})\mathscr{E}^2U+2\dot\varphi U\\ =(2\hbar)^{-1}|k_{21}|^2\mathscr{E}^2(\delta n+n_0),\qquad \delta\dot n=-V\mathscr{E}^2/2\hbar,\end{gathered} \tag{9}$$

where the constants ξ_0 and β are determined by the nonresonant polarization and by the parameters of the resonance layer:

$$\xi_0=\beta[k_0+{}^1/_2(k_{11}+k_{22})+{}^1/_2(k_{22}-k_{11})n_0],\quad \beta=4\pi\omega N_0/c(A+B),$$

n_0 is the initial value of the inversion (in the absence of a field), and F is expressed thorugh the Fresnel coefficients A and B: $F = 2A/(A+B)$.

Finding a general solution of the system (9) is quite difficult because of the possible phase modulation. We note that by obtaining analytic solutions valid for an extended resonance medium it is possible, by renormalization of the wave number, to exclude the term due to the nonresonant part of the polarization.[1]

We will solve the system (9) approximately, neglecting the derivative $\dot{\varphi}$. This is obviously possible for the conditions

$$\dot{\varphi} \ll \tau_i^{-1} \text{ or } \dot{\varphi} \ll \frac{(k_{22}-k_{11})}{4\hbar}\mathscr{E}^2.$$

It is easy to obtain from the third and fifth equations of system (9)

$$\dot{U}+{}^1/_2(k_{22}-k_{11})\delta\dot{n}=0.$$

Thus, the quantity $U(t) + (1/2)(k_{22} - k_{11})\delta n(t)$ is an integral of the motion, which can be taken to be zero from the conditions $U(-\infty) = 0$ and $\delta n(-\infty) = 0$. The solution of the last equations of system (9) follows easily:

$$V=\frac{2|k_{21}|^2 n_0}{[(k_{22}-k_{11})^2+4|k_{21}|^2]^{1/2}}\sin\Psi,$$
$$\delta n=\frac{4|k_{21}|n_0}{(k_{22}-k_{11})^2+4|k_{21}|^2}(\cos\Psi-1), \tag{10}$$

where

$$\Psi=\frac{[(k_{22}-k_{11})^2+4|k_{21}|^2]^{1/2}}{4\hbar}\int_{-\infty}^{t}\mathscr{E}^2(t)\,dt.$$

The first two equations of system (9) reduce to

$$(1+\xi\sin\Psi)\mathscr{E}=F\mathscr{E}_0\cos(\varphi_0-\varphi),$$
$$\xi_0\mathscr{E}=F\mathscr{E}_0\sin(\varphi_0-\varphi), \tag{11}$$

where

$$\xi=-\frac{8\pi\omega N_0|k_{21}|^2 n_0}{c(A+B)[(k_{22}-k_{11})^2+4|k_{21}|^2]^{1/2}}, \tag{12}$$

from which it follows that

$$\varphi=\varphi_0-\operatorname{arctg}\{\xi_0/(1+\xi\sin\Psi)\}. \tag{13}$$

It is possible to finally formulate the condition for the neglect of phase modulation by using Eq. (13): the first is absence of phase modulation at the entrance to the resonance medium, and the second is

$$\left|\frac{\xi_0\xi\cos\Psi}{(1+\xi\sin\Psi)^2+\xi_0^2}\right|\ll 1. \tag{14}$$

3. ENERGIES OF REFLECTED AND TRANSMITTED USP LIGHT IN A THIN RESONANCE LAYER IN THE ABSENCE OF PHASE MODULATION

We begin by formulating an "energy theorem" for optical radiation passing through a resonance layer. Squaring each of Eqs. (11) and dropping the trigonometric functions containing $\varphi_0(t)$ and $\varphi(t)$, we obtain

$$(1+\xi\sin\Psi)^2\mathscr{E}^2+\xi_0^2\mathscr{E}^2=F^2\mathscr{E}_0^2.$$

Integrating over time and changing to the "rotation angles" Ψ and Ψ_0 leads to an "energy theorem" connecting the quantities Ψ and Ψ_0 with the parameters of the resonance layer:

$$\left(1+\xi_0^2+\frac{\xi^2}{2}\right)\Psi+4\xi\sin^2\frac{\Psi}{2}-\frac{\xi^2}{4}\sin 2\Psi=F^2\Psi_0. \tag{15}$$

In the same way we obtain an expression connecting the "rotation angle" (and with it the energy) of the reflected USP light with the parameters of the resonance layer. It follows from Eq. (8) that

$$\mathscr{E}_r^2=\mathscr{E}^2+\mathscr{E}_0^2-2\mathscr{E}\mathscr{E}_0\cos(\varphi_0-\varphi). \tag{16}$$

Integrating over time and using the first equation of system (11) leads to the relation

$$\Psi_r=\Psi+\Psi_0-2F^{-1}\{\Psi+\xi(1-\cos\Psi)\}. \tag{17}$$

We note that Eq. (17) is the law of conservation of energy in the interaction of USP light with a thin surface layer. One can easily be convinced of this by using the obvious properties of the Fresnel coefficients: $2B/(A+B) = 2-F$. Thus,

$$A(\Psi_0-\Psi_r)=B\Psi+\xi(A+B)(1-\cos\Psi), \tag{18}$$

i.e., the energy of the incident radiation is distributed between the energies of the reflected and transmitted waves and the energy absorbed in the resonance layer.

Hence, Eqs. (15) and (18) allow a determination of the energies of the reflected and transmitted USP light for two-quantum resonance in the absence of phase modulation.

We analyze the possible cases of absence of phase modulation. The inequalities (14) can be satisfied for the following conditions: a) $\xi_0, \xi \ll 1$, b) $\xi \lesssim 1, \xi_0 \ll 1$, c) $\xi_0 \sim 1, \xi \ll 1$, and d) $\xi \sim 1, \xi_0 \gg \xi$. In these limiting situations the "energy theorem" takes the following forms:

a) $\Psi=F^2\Psi_0$,
b) $F^2\Psi_0=(1+\xi^2/2)\Psi+4\xi\sin^2(\Psi/2)-(\xi^2/4)\sin 2\Psi$,
c) $(1+\xi_0^2)\Psi=F^2\Psi_0$,
d) $\xi_0^2\Psi=F^2\Psi_0$.

Thus, the temporal form of the transmitted and reflected USP light can differ significantly from the form of the incident USP light only in case b). In the remaining cases neglect of phase modulation leads to conservation of the temporal form of all light pulses.

A graph of the dependence of the energy of the incident USP as a function of the energy of the USP light passing through the resonance layer for the conditions $\xi \ll 1$, and $\xi_0 \ll 1$ is given in Fig. 1. We note the monotonic character of this dependence, since the time derivatives of the functions Ψ_0 and Ψ define essentially positive quantities. We note also that the region for which the derivative $d\Psi_0/d\Psi$ is close to zero broadens as ξ approaches unity. Physically, this means that, in those regions, an insignificant increase in the energy of the incident light pulse can lead to a significant increase in the energy of the radiation passing through the resonant layer; i.e., anomalous transmission occurs.

We estimate the orders of magnitude of the corresponding quantities. The coefficients of the Stark shift and the matrix element of the two-photon transition are in general of the same order of magnitude: $k_{11}\sim k_{22}\sim k_{21}\sim d^2/\hbar\omega\sim 3\times 10^{-25}$ cgs esu with $d\sim 10^{-18}$ esu and $\omega\sim 3\times 10^{15}$

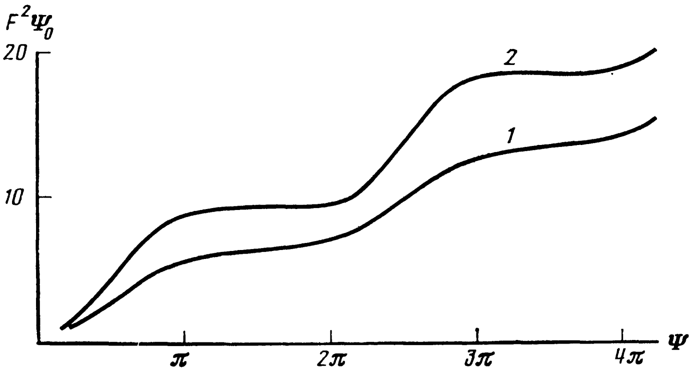

FIG. 1. The dependence of the energy of the incident USP light on the energy of the USP light passing through the resonance layer for values of the parameters $\xi_0 \ll 1$, $\xi = 0.5$ (curve 1), or $\xi = 1$ (curve 2).

s^{-1}. It is clear that nonlinear effects in the resonance layer will occur for $\xi_0 \sim \xi \sim 1$. Hence, $N_0 \sim \lambda (A + B)/8\pi^2 k \sim 10^{18}$ cm^{-2}, which means approximately 10^2 atom layers in films based on LaF_3. We note that in a number of cases (e.g., in metal atoms such as Na, Rb) k can exceed the given value by several orders of magnitude due to the influence of an intermediate resonance level, and can reach $\sim 3 \times 10^{-21}$ cgs esu (Ref. 11). In this case $N_0 \sim 10^{14}$ cm^{-2}. We estimate the necessary levels of intensity from the condition that the "rotation angles" Ψ should be of order $\pi/2$. Hence, $k_{21}E^2\delta_p/\pi\hbar \sim 1$ and for the intensity $I = cE^2/8\pi$, we obtain $I \sim \hbar c/8k_{21}\delta_p \sim 10$ $MW \cdot cm^{-2}$ for pulse lengths $\delta_p \sim 10^{-12}$ s and $k_{21} \sim 3 \times 10^{-21}$ esu.

Note added in proof (17 January 1989). We emphasize that the noted features of the interaction of USP light with a thin layer of surface resonance atoms were obtained neglecting the local field. In the general case the quantity E in the system of Eqs. (7) should be taken as the local field acting on the resonance atoms in the film: $E_{\rm loc} = E + \mu P_F/a$, where a is the characteristic distance between the atoms on the surface and μ is a numerical coefficient of order unity. The "energy theorem" (15) remains valid also with allowance for the local field if we take in Eq. (15) for the values 1; ξ_0, ξ^2, ξ, the corresponding values

$$(1-\xi_\alpha^0);\ \xi_\beta^0;\ \xi_\alpha^2+\xi_\beta^2;\ \xi_\beta^0\xi_\alpha+\xi_\beta(1-\xi_\alpha^0),$$

where

$$\xi_\beta^{(0)}=\xi_{(0)},\ \xi_\alpha^{(0)}=\xi_\beta^{(0)}\mu N_0/\beta a.$$

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