Nonlinear interaction of light with a thin layer of surface resonant atoms

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We consider the effects of nonlinear interaction of light with a thin layer of surface resonant atoms, allowing for the Lorentz local field in two limiting cases: the quasi-steady-state case with typical pulse lengths much greater than the transverse and the longitudinal relaxation times, and the case of ultrashort pulse lengths when the opposite inequality holds.

In connection with the development of coherent surface spectroscopy,[1,2] an interest has been shown in physical objects such as thin layers of resonant atoms.[3–24] This model of a nonlinear medium includes, on the one hand,[3–6] the problem of nonlinear surface wave propagation and on the other, the problem of ultrashort light pulses reflecting from an interface.[7–11] Under certain conditions, surface waves can be efficiently excited due to the photon echo effect.[12] Moreover, there is a purely practical interest in studying such structures in connection with the need for efficient control of laser radiation and creation of an element basis for optoelectronics.

Although the resonant layer thickness is small in comparison with optical wave length, the nonlinear relation between the field amplitude of the transmitted wave and the optical properties of the resonant medium give rise to bistability phenomena and self-pulsations under quasi-steady-state conditions.[13–19] Under certain conditions, the nonlinear dynamics of the system may exhibit optical chaos.[23,24]

The bistability properties of a thin resonant layer located at the interface of two linear media were discussed earlier on the basis of both quantum[13,15] and quasiclassical[17–22] models. In particular, in Refs. 13, 15, and 16 the need to include dipole-dipole interactions was pointed out. However, consistent implementation of this program is a laborious problem.

It was shown in Refs. 13 and 20 that effective inclusion of the dipole-dipole interaction can be performed using the local Lorentz field. It is of interest to generalize the results obtained earlier treating the Lorentz field,[11,17,19,22] although a specific model with no allowance made for the local field is valid in a number of physical situations, for instance, light propagation through a cavity with low Q in the mean-field approximation or in semiconductors, and deserves special consideration.

In the present paper, we formulate basic equations describing the nonlinear interaction of light pulses with a thin layer of resonant atoms, treating the local Lorentz field using the quasiclassical approach. We discuss further the phenomenon of cavityless optical bistability under the conditions of quasi-steady-state action of light on a thin film of atoms. Then we consider transient propagation of coherent optical pulses in the limit of ultrashort pulse widths as well as the interaction of a light pulse with a thin layer under the conditions of two-quantum absorption.

1. BASIS EQUATIONS

Let us consider a thin layer of resonant atoms, of thickness l much less than the optical wavelength. The layer is located at the interface $z=0$ of two linear media, with permittivities ε_a and ε_b. For definiteness we restrict ourselves to detailed consideration of TE waves only.

We assume that a light wave is incident on the interface from the first medium, and the electric field strength of the wave is given by the expression

$$E_y(x,z,t)=E_0(x,z,t)\exp(ik_x^a x+ik_z^a z-i\omega t),$$

where $E_0(x,z,t)$ is a slowly varying amplitude, k_x^a, k_z^a are the projections of the wave vector in the first medium ($k^a=\omega\sqrt{\varepsilon_a}/c$). For the reflected and the transmitted waves, respectively, we can write down the following expressions:

$$E_y^r(x,z,t)=E_r(x,z,t)\exp(ik_x^a x-ik_z^a z-i\omega t),$$

$$E_y^{tr}(x,z,t)=E(x,z,t)\exp(ik_x^b x-ik_z^b z-i\omega t),$$

where $E_r(x,z,t)$ and $E(x,z,t)$ are the slowly varying field amplitudes of the reflected and the refracted waves, $k^b=\omega\sqrt{\varepsilon_b}/c$, and $k_x^a=k_x^b$.

Note that the distinction between this boundary-value problem and the problem of light propagation in extended resonant media is that instead of the Maxwell equations, the corresponding electrodynamic conditions are used to relate the fields at the interface:

$$\begin{aligned} &E_y(x,+0,t)-E_y(x,-0,t)=0,\\ &H_z(x,+0,t)-H_z(x,-0,t)=0,\\ &H_x(x,+0,t)-H_x(x,-0,t)=\frac{4\pi}{c}P_y(x,t), \end{aligned} \tag{1.1}$$

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

From the boundary conditions (1.1), follows a relation for the electric field amplitudes of all waves and polarizations

$$\begin{aligned} E&=\frac{2A}{A+B}E_0+i\frac{4\pi\omega}{c(A+B)}P_s,\\ E_r&=\frac{A-B}{A+B}E_0+i\frac{4\pi\omega}{c(A+B)}P_s, \end{aligned} \tag{1.2}$$

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

In the general case, the coupling conditions (1.2) should be supplemented by the Bloch equations. As a result, a closed self-consistent set of equations appears in which the field of the wave transmitted through the layer depends on the resonant properties of the surface atoms and in turn determines their dynamics.

We consider first the interaction of coherent light with a thin layer of surface atoms under the conditions of single-quantum resonance. Then

$$P_s = P_0 + P, \tag{1.3}$$

$$P = N_s\langle p_\varepsilon\rangle = N_s\int g(\varepsilon)p_\varepsilon d\varepsilon, \tag{1.4}$$

where P_0 and P are the nonresonant and resonant contributions to the polarization, and p_ε satisfies the equations

$$\begin{aligned}&\dot p_\varepsilon + \gamma_2 p_\varepsilon + i\varepsilon p_\varepsilon = -\frac{i}{\hbar}|d_{12}|^2 E n_\varepsilon,\\ &\dot n_\varepsilon + \gamma_1(n_\varepsilon - n_0) = \frac{i}{2\hbar}(p_\varepsilon^* E - p_\varepsilon E^*).\end{aligned} \tag{1.5}$$

In relations (1.4) and (1.5) p_ε and n_ε are the mean quantum mechanical values of the dipole moment and inverse population per resonant atom; d_{12} is the matrix element of the transition dipole moment; $\varepsilon = \omega_{21} - \omega$ is the detuning from resonance; N_s is the resonant atom surface density; γ_1 and γ_2 are the phenomenological population and polarization relaxation rates; and the function $g(\varepsilon)$ describes inhomogeneous broadening of the resonant energy levels.

In connection with the set of equations (1.2)–(1.5), the following point should be noted. The fields entering into the electrodynamic relations (1.2) are macroscopically average and, generally speaking, can differ from the field appearing in the Bloch equations (1.5). This is because the macroscopic average field value differs from the local field at the positions of individual resonant atoms. It is known that for a bulk medium with atoms located at cubic lattice sites and with completely random arrangement of atoms (a gaseous dielectric) the connection to the local field has the form[13,20]

$$E_{\text{loc}} = E + \frac{4\pi}{3}P,$$

where P is the volume polarization of the medium.

If the atoms are located at the surface, a sum of the form

$$\sum_i \frac{3(\mathbf{p}_i\mathbf{r}_i)\mathbf{r}_i - \mathbf{p}_i r_i^2}{r_i^5}$$

should be evaluated to calculate the field produced by all surface dipoles at the location of the given dipole.

For the geometry adopted in the present work, this expression takes the form

$$\sum_i \frac{3p_{iy}y_i^2 - p_{iy}(x_i^2+y_i^2)}{r_i^5} = \sum_i \frac{2p_{iy}y_i^2}{r_i^5}.$$

The calculation result depends on the particular arrangement of atoms at the surface. It is clear from dimensional considerations that the local field will be

$$E_{\text{loc}} = E + \mu\left(\frac{P}{a} + \frac{P_0}{a_0}\right), \tag{1.6}$$

where μ is a numerical factor of order unity, and a and a_0 are the typical average values of distances between resonant and nonresonant atoms at the surface.

Let us estimate the contribution of the nonresonant polarization to the local field. In the general case the value of P_0 may be determined by nonresonant impurity atoms and is equal to $N_s^0\beta E_{\text{loc}}$ in order of magnitude, where β is the impurity atom polarizability and N_s^0 is the surface density. We assume that the local field at an impurity atom is the same as at a resonant one. Then

$$E_{\text{loc}} = \alpha_0 E + \frac{\alpha_0\mu P}{a},$$

where

$$\alpha_0 = \left(1 - \frac{\mu\beta N_s^0}{a_0} - \right)^{-1}, \quad \beta = \frac{2}{\hbar}\frac{|d_{ij}|^2\omega_{ji}}{(\omega_{ji}^2 - \omega^2)}.$$

To estimate β, we use the relation

$$\beta \sim \frac{|d_{ij}|^2}{\hbar\omega} \sim 3\cdot 10^{-25}\ \text{cm}^3.$$

For typical distances $a_0 \sim 3\cdot 10^{-8}$ cm, $N_s^0 \sim a_0^{-2}$, we have $\beta N_s^0/a_0 \sim 10^{-2}$. Thus, we may neglect the nonresonant contribution to the local field and take $a_0 \not\equiv 1$. As a result, we get the following self-consistent set of equations determining the local field:

$$\begin{cases} E = FE_0 + (\alpha' + \beta')N_s\langle p_\varepsilon\rangle,\\ \dot p_\varepsilon + \gamma_2 p_\varepsilon + i\varepsilon p_\varepsilon = -\frac{i}{\hbar}|d_{12}|^2 E n_\varepsilon,\\ \dot n_\varepsilon + \gamma_1(n_\varepsilon - n_0) = \frac{i}{2\hbar}(p_\varepsilon^* E - p_\varepsilon E^*), \end{cases} \tag{1.7}$$

where

$$\alpha' = \frac{\mu}{a}, \quad \beta' = \frac{4\pi\omega}{c(A+B)}, \quad F = \frac{2A}{A+B}. \tag{1.8}$$

We consider the solution of this set of equations in two limiting cases: quasi-steady-state, when typical lengths of optical pulses are much longer than the transverse and the longitudinal relaxation times ($\tau_p \gg \gamma_{1,2}^{-1}$), and ultrashort pulses, when the opposite inequality holds ($\tau_p \ll \gamma_2^{-1} \ll \gamma_1^{-1}$).

2. QUASI-STEADY REGIME OF LIGHT ACTING ON A THIN LAYER OF RESONANT ATOMS: OPTICAL BISTABILITY

Evidently, we may neglect the first derivatives of n_ε and p_ε in the first case. Then

$$n_\varepsilon=\frac{n_0(1+\Delta_\varepsilon^2)}{(1+\Delta_\varepsilon^2+|f_e|^2)}, \tag{2.1}$$

$$P=i\frac{N_s|d_{12}|^2}{\hbar\gamma_2}E\int g(\varepsilon)d\varepsilon\frac{(1-i\Delta_\varepsilon)}{(1+\Delta_\varepsilon^2+|f_e|^2)}, \tag{2.2}$$

where

$$\Delta_\varepsilon=\varepsilon/\gamma_2,\quad f_e=|d_{12}|E/\hbar\sqrt{\gamma_1\gamma_2}. \tag{2.3}$$

Let us examine the case of a homogeneously broadened spectral line, so that $g(\varepsilon)=\delta(\varepsilon-\Delta\omega_0)$ and

$$\int d\varepsilon g(\varepsilon)\frac{(1-i\Delta_\varepsilon)}{(1+\Delta_\varepsilon^2+|E|^2/E_s^2)}=\frac{(1-i\Delta_0)}{(1+\Delta_0^2+|E|^2/E_s^2)}, \tag{2.4}$$

where $\Delta_0=\Delta\omega_0/\gamma_2$ and $E_s=\hbar\sqrt{\gamma_1\gamma_2}/|d_{12}|$ is the saturation field.

Then in units of saturation field E_s the basic equation for the field E takes the form

$$E=FE_0+i\frac{(\alpha+i\beta)(1-i\Delta_0)}{(1+\Delta_0^2+|E|^2)}, \tag{2.5}$$

$$\alpha=\frac{\mu N_s|d_{12}|^2}{a\hbar\gamma_2},\quad \beta=\frac{4\pi\omega N_s|d_{12}|^2}{c(A+B)\hbar\gamma_2} \tag{2.6}$$

or

$$FE_0=E\left\{1+\frac{(\beta-\alpha\Delta_0)}{(1+\Delta_0^2+|E|^2)}-i\frac{(\alpha+\beta\Delta_0)}{(1+\Delta_0+|E|^2)}\right\}. \tag{2.7}$$

When we take out the phase factors

$$E=\mathscr{E}\exp(i\varphi),\quad E_0=\mathscr{E}_0\exp(i\varphi_0)$$

the field equation changes its form:

$$\mathscr{E}\left\{\cos(\varphi-\varphi_0)\left[1+\frac{(\beta-\alpha\Delta_0)}{(1+\Delta_0^2+|E|^2)}\right]-\sin(\varphi-\varphi_0)\frac{(\alpha+\beta\Delta_0)}{(1+\Delta_0+|E|^2)}\right\}=0, \tag{2.8}$$

$$\sin(\varphi-\varphi_0)\left[1+\frac{(\beta-\alpha\Delta_0)}{(1+\Delta_0^2+|E|^2)}\right]-\cos(\varphi-\varphi_0)\frac{(\alpha+\beta\Delta_0)}{(1+\Delta_0+|E|^2)}=0. \tag{2.9}$$

From here we easily find a solution for the phase

$$\varphi=\varphi_0+\operatorname{arctg}\left\{\frac{\alpha+\beta\Delta_0}{(1+(\beta-\alpha\Delta_0)+\Delta_0^2+X)}\right\}. \tag{2.10}$$

Upon squaring Eqs. (2.8) and (2.9) and summing them we get an expression for the intensities of the incident light and the radiation "localized in the layer":

$$Y=X\left\{\left[1+\frac{(\beta-\alpha\Delta_0)}{(1+\Delta_0^2+X)}\right]^2+\frac{(\alpha+\beta\Delta_0)^2}{(1+\Delta_0+X)^2}\right\}, \tag{2.11}$$

where $X=\mathscr{E}^2$ and $Y=F^2\mathscr{E}_0^2$.

Note that Eqs. (2.7) and (2.11) are the analogs of the equations used in the theory of absorption-dispersion optical bistability in steady state.[14]

In fact, Eq. (2.11) admits bistable regime in the solution for certain values of the parameters α, β, and Δ_0. The condition for the derivative dY/dX to vanish can be obtained from (2.11):

$$\tilde{X}^3-[\alpha^2+\beta^2-2(\beta-\alpha\Delta_0)](1+\Delta_0^2)\tilde{X}+2(\alpha^2+\beta^2)(1+\Delta_0^2)^2=0, \tag{2.12}$$

where $\tilde{X}=X+1+\Delta_0^2$.

In the general case, the last equation has three roots, two of which may be complex. One real root is of no interest for us, because for it $\tilde{X}<0$. Two other roots become real for $\tilde{X}>0$, if the inequality

$$[\alpha^2+\beta^2-2(\beta-\alpha\Delta_0)]^3>27(\alpha^2+\beta^2)(1+\Delta_0^2). \tag{2.13}$$

is fulfilled.

The last inequality is the condition for the occurrence of a bistable regime or the condition for the occurrence of an S-shaped characteristic in the dependence $X(Y)$. In the particular case $\beta=0$ and exact resonance, the condition (2.13) reduces to the inequality $\alpha>\sqrt{27}=5.2$.

Note that the properties of the solutions which are responsible for optical bistability involve the properties of the local field. To determine the amplitude of the macroscopic field along with the amplitude of the wave transmitted through the film, we must use relation (1.6) again. Then

$$E_{tr}=E-\frac{\mu P}{a}=E\left\{1-\frac{\alpha(\Delta_0+i)}{(1+\Delta_0^2+|E|^2)}\right\}. \tag{2.14}$$

Then the transmitted wave intensity $X_{tr}=E_{tr}E_{tr}^*$ and the intensity of radiation "localized in the film" $X=|E|^2$ are related by

$$X_{tr}=X\left\{\left[1-\frac{\alpha\Delta_0}{(1+\Delta_0^2+X)}\right]^2+\frac{\alpha^2}{(1+\Delta_0^2+X)^2}\right\}. \tag{2.15}$$

The relation between X and X_{tr} is of the bistability type, if the condition

$$(\alpha^2+2\alpha\Delta_0)^3>27\alpha^4(1+\Delta_0^2).$$

is met.

Thus, X_{tr} is found in two steps. First, X is determined from Eq. (2.11), and then Eq. (2.15) is used to find X_{tr}. Note that for $\beta\ll\alpha$, the quantities X, X_{tr}, and Y are related by

$$Y=X_{tr}+\frac{2\beta X}{(1+\Delta_0^2+X)}. \tag{2.16}$$

In the limit of small β we can neglect the ambiguity in the solution for X giving rise to the bistable regime. Then we find that the transmitted radiation intensity is independent of the resonant layer properties, $\mathscr{E}_{tr}^2=F^2\mathscr{E}_0^2$.

3. TIME-DEPENDENT TRANSMISSION OF COHERENT OPTICAL PULSES IN THE ULTRASHORT LIMIT; PHOTON ECHO.

By ultrashort optical pulses we mean pulses much shorter than the typical polarization relaxation times γ_2^{-1}. In this limit, system (1.7) simplifies:

$$\begin{cases} E=FE_0+(\alpha'+\beta')N_s\langle p_\varepsilon\rangle, \\ \dot p_\varepsilon+i\varepsilon p_\varepsilon=-\dfrac{i}{\hbar}|d_{12}|^2En_\varepsilon, \\ \dot n_\varepsilon=\dfrac{i}{2\hbar}(p_\varepsilon^*E-p_\varepsilon E^*). \end{cases} \tag{3.1}$$

When the phase factors are removed, $E_0=\mathscr{E}_0\exp(i\varphi_0)$, $E=\mathscr{E}\exp(i\varphi)$, $p_\varepsilon=(u_\varepsilon+iv_\varepsilon)\exp(i\varphi)$, Eqs. (3.1) take the form

$$\begin{aligned} &\mathscr{E}=\beta'N_s\langle v_\varepsilon\rangle-\alpha'N_s\langle u_\varepsilon\rangle=F\mathscr{E}_0\cos(\varphi-\varphi_0), \\ &\alpha'N_s\langle v_\varepsilon\rangle+\beta'N_s\langle u_\varepsilon\rangle=F\mathscr{E}_0\sin(\varphi-\varphi_0), \\ &\dot u_\varepsilon-(\varepsilon+\dot\varphi)v_\varepsilon=0, \\ &\dot v_\varepsilon+(\varepsilon+\dot\varphi)u_\varepsilon=-\frac{|d_{12}|^2}{\hbar}\mathscr{E}n_\varepsilon,\quad \dot n_\mathscr{E}=\frac{v_\varepsilon\mathscr{E}}{\hbar}. \end{aligned} \tag{3.2}$$

Let us consider approximate solutions of this set of equations corresponding to the absence of phase modulation ($\dot\varphi=0$). We also neglect $\langle u_\varepsilon\rangle$. In the case of a symmetric inhomogeneously broadened line, as well as that of exact resonance, this quantity is exactly zero.

Under these conditions, the system (3.2) admits an approximate solution analogous to the "area theorem" by McCall and Hahn.[19]

Thus, integrating the first two equations of (3.2) over time and using the relations

$$\frac{|d_{12}|}{\hbar}\int_{-\infty}^{\infty}\mathscr{E}_0(t)dt=\Theta_0,\quad \frac{|d_{12}|}{\hbar}\int_{-\infty}^{\infty}\langle v_\varepsilon(t)\rangle dt=\frac{|d_{12}|^2}{\hbar}\pi g(0)\sin\Theta,$$

we get

$$\begin{aligned} &\Theta+\xi_\beta\sin\Theta=F\Theta_0\cos(\varphi-\varphi_0), \\ &\xi_\alpha\sin\Theta=F\Theta_0\sin(\varphi-\varphi_0), \end{aligned} \tag{3.3}$$

where

$$\xi_\alpha=\frac{\alpha'N_s|d_{12}|^2\pi g(0)}{\hbar},\quad \xi_\beta=\frac{\beta'N_s|d_{12}|^2\pi g(0)}{\hbar}. \tag{3.4}$$

From this we easily get

$$\Theta_0^2F^2=\Theta^2+(\xi_\alpha^2+\xi_\beta^2)\sin^2\Theta+2\xi_\beta\Theta\sin\Theta, \tag{3.5}$$

$$\varphi=\varphi_0+\operatorname{arctg}\frac{\xi_\alpha\sin\Theta}{(\Theta+\xi_\beta\sin\Theta)}. \tag{3.6}$$

It follows from (3.7) that phase modulation may be neglected only in the small area limit. Then

$$\varphi=\varphi_0+\operatorname{arctg}\frac{\xi_\alpha}{(1+\xi_\beta)}, \tag{3.7}$$

and from (3.5) we obtain

$$\Theta_0F=\Theta\sqrt{(1+\xi_\beta)^2+\xi_\alpha^2}. \tag{3.8}$$

The optical pulse area Θ determined by the local field in the film can be arbitrary in the limit $\xi_\alpha\ll\xi_\beta$ or if the density of resonant atoms in the film is small ($\alpha\gg\lambda$). It can easily be shown that $\xi_\beta\ll1$ holds in this case, and we find $\Theta_0F=\Theta$.

In the general case, we come to the conclusion that the phenomenon of phase modulation is important when ultrashort optical pulses interact with a thin layer of resonant atoms, and the "area theorem" of McCall and Hahn loses its physical meaning.

Note also that the original equations (3.1) describing the interaction of an ultrashort optical pulse with a thin layer of resonant atoms whn the Lorentz field is included cannot be integrated by the inverse scattering method.

The field amplitude of the transmitted wave can be determined similarly. Using relation (1.6) we get

$$E_{tr}=E-\alpha'N_s\langle p_\varepsilon\rangle, \tag{3.9}$$

and integrating over time, we arrive at the following expressions:

$$\Theta_{tr}^2=\xi_\alpha^2\sin^2\Theta+\Theta^2; \tag{3.10}$$

$$\varphi-\varphi_0=\operatorname{arctg}\frac{\Theta}{\xi_\alpha\sin\Theta}. \tag{3.11}$$

Again, the condition of zero phase modulation can be met for moderate Θ values ($\Theta\leqslant1$). In the limit of small "areas," we get

$$\Theta_{tr}=\sqrt{\xi_\alpha^2+1}\,\Theta. \tag{3.12}$$

Consider now the formation of photon echo signals induced in a resonant layer by a sequence of two ultrashort optical pulses with time separation τ_0. The feasibility of obtaining analytical solutions in this region is associated with relations (3.7), (3.8), and (3.12), signifying essentially the "small-area" approximation $\Theta<1$. It is known that in this approximation we may set

$$n_\varepsilon(t)=n_\varepsilon(0)=-1$$

and find from Eqs. (3.1) that

$$\langle p_\varepsilon\rangle=-\frac{i}{\hbar}|d_{12}|^2\int_{-\infty}^{t}dt'\int_{-\infty}^{\infty}d\varepsilon g(\varepsilon)E(t')\times\exp[-\varepsilon(t-t')].$$

In the limit $\tau_p\gg T_2^*$ where τ_p is the excitation pulse width and T_2^* characterizes the spectral width of the function $g(\varepsilon)$, we can obtain

$$\langle p_\varepsilon\rangle=-\frac{i}{\hbar}|d_{12}|^2\pi g(0)E(t).$$

Then using (3.1), we arrive at expression

$$E=\frac{FE_0}{(1+\xi_\beta-i\xi_\alpha)}. \tag{3.13}$$

This expression should be regarded as the field acting on the resonant atoms in a thin layer. Then, using the results of the photon echo theory for disperse resonant media, we write down the following expression for the polarization when the photon echo signal occurs:

$$P(t)=-\frac{i|d_{12}|^4N_s}{4\hbar^3}\kappa^2\kappa^*\langle\mathscr{E}_{20}^2(\varepsilon)\mathscr{E}_{10}^*(\varepsilon)$$

$$\times\exp[-i(t-\tau_\theta)]\rangle, \tag{3.14}$$

where

$$\kappa=\frac{F}{(1+\xi_\beta-i\xi_\alpha)},$$

and $\mathscr{E}_{1,20}(\varepsilon_2)$ are the Fourier transforms of the slow envelopes of the incident optical pulses and τ_θ is the typical time of photon echo signal formation. It is assumed that the excitation pulse widths τ_p satisfy the inequality $\tau_{p1,2}\ll\tau_0$.

For simplicity, the expression for the polarization is given at the origin of the coordinate resonant layer system, i.e., at $x=0$.

4. INTERACTION OF AN ULTRASHORT OPTICAL PULSE WITH A THIN LAYER OF SURFACE ATOMS UNDER TWO-PHOTON RESONANCE CONDITIONS

In the case of two photon resonance, the basic equations (1.2) remain valid, but the polarization P_s should be taken to be the expression

$$P_s=N_sPE^*+N_s\left[\frac{N_0}{N_s}K_0+\frac{K_{11}+K_{22}}{2}+\frac{K_{22}-K_{11}}{2}n\right]E. \tag{4.1}$$

The resonant and nonresonant contributions are taken into account here; n is the inverse population of resonant energy levels per atom; N_s and N_0 are the surface densities of resonant and nonresonant atoms in the layer; K_{11} and K_{22} describet he Stark shift of energy levels; K_{21} is the matrix element of the two-photon transition; the factor K_0 allows for the contribution of nonresonant impurities into polarization; and the the P component of the Bloch vector satisfies

$$\dot{P}-i\frac{K_{22}-K_{11}}{4\hbar}|E|^2P=-i\frac{|K_{21}|^2}{2\hbar}E^2n,$$

$$\dot{n}=\frac{(PE^{*2}-P^*E^*)}{4i\hbar}. \tag{4.2}$$

Note that in writing down Eqs. (4.2) we have used the ultrashort-width approximation and treated the case of exact resonance ($2\omega=\omega_{21}$) and homogeneously broadened lines.

We assume that the ultrashort optical pulse is incident on a resonant layer, and the electric field amplitude of the pulse is represented by the expression

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

If the quantity E entering into the Eqs. (4.2) is taken to mean the local field acting on a resonant atom, Eq. (1.2) becomes

$$E=FE_0+(\alpha'+i\beta')P_s, \tag{4.3}$$

Then for the local field amplitude E and the Bloch vector component we use the representation analogous to E_0:

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

Separating the variables, we get

$$(1+\beta v-\xi_\alpha^0)\mathscr{E}-\alpha\left[u+\frac{K_{22}-K_{11}}{2}\delta n\right]\mathscr{E}$$

$$=F\mathscr{E}_0\cos(\varphi-\varphi_0),$$

$$(\xi_\beta^0+\alpha v)\mathscr{E}+\beta\left[u+\frac{K_{22}-K_{11}}{2}\delta n\right]\mathscr{E}=F\mathscr{E}_0\sin(\varphi-\varphi_0),$$

$$\dot{u}+\frac{K_{22}-K_{11}}{4\hbar}\mathscr{E}^2v-2\dot{\varphi}v=0, \tag{4.4}$$

$$\dot{v}-\frac{K_{22}-K_{11}}{4\hbar}\mathscr{E}^2u+2\dot{\varphi}u=-\frac{|K_{12}|^2}{2\hbar}\mathscr{E}^2(\delta n+n_0),$$

$$\delta\dot{n}=\frac{v\mathscr{E}^2}{2\hbar}.$$

where $\alpha=\alpha'N_s$, $\beta=\beta'N_s$;

$$\xi_\alpha^0(\beta)=\alpha(\beta)\left\{\frac{N_0}{N_s}K_0+\frac{K_{11}+K_{22}}{2}+\frac{K_{22}-K_{11}}{2}n_0\right\}. \tag{4.5}$$

Neglecting phase modulation, from the first and the fifth equations of system (4.4) we find

$$\dot{u}+\frac{K_{22}-K_{11}}{2}\delta\dot{n}=0,$$

so the second terms in the first two equations (4.4) can be omitted. Then we present the solution of Eqs. (4.4) in the following form:

$$v=\frac{-2|K_{21}|^2n_0}{\sqrt{(K_{22}-K_{11})^2+4|K_{21}|^2}}\sin\psi,$$

$$\psi=\frac{\sqrt{(K_{22}-K_{11})^2+4|K_{21}|^2}}{4\hbar}\int_{-\infty}^{t}\mathscr{E}^2(t)dt,$$

$$(1-\xi_\alpha^0+\xi_\beta\sin\psi)\mathscr{E}=F\mathscr{E}_0\cos(\varphi-\varphi_0),$$

$$(\xi_\beta^0+\xi_\alpha\sin\psi)\mathscr{E}=F\mathscr{E}_0\sin(\varphi-\varphi_0), \tag{4.6}$$

where

$$\xi_{\alpha,\beta}=\alpha(\beta)\frac{-2|K_{21}|^2n_0}{\sqrt{(K_{22}-K_{11})^2+4|K_{21}|^2}}. \tag{4.7}$$

From this we find

$$\tilde{A}\psi-\tilde{B}\sin 2\psi+\tilde{C}\sin^2\frac{\psi}{2}=F^2\psi_0. \tag{4.8}$$

Here

$$\tilde{A}=\left[\xi_\beta^{02}+(1-\xi_\alpha^0)^2+\frac{\xi_\alpha^2+\xi_\beta^2}{2}\right],$$

$$\tilde{B}=(\xi_\alpha^2+\xi_\beta^2)/4, \tag{4.9}$$

$$\tilde{C}=4\xi_\beta^0\xi_\alpha+4\xi_\beta(1-\xi_\alpha^0).$$

Note that integral of motion (4.8) was obtained neglecting phase modulation. It follows from the last two equations of the solution (4.6) that

$$\varphi-\varphi_0=\operatorname{arctg}\frac{(\xi_\beta^0+\xi_\alpha\sin\psi)}{(1-\xi_\alpha^0+\xi_\beta\sin\psi)}, \tag{4.10}$$

and since $\alpha\gg\beta$ usually holds, we have $\xi_\alpha\gg\xi_\beta$ ($\xi_\alpha^0\gg\xi_\beta^0$) and phase modulation may be neglected if $\xi_\alpha^0\simeq 1$. Then we may set $\varphi-\varphi_0=\pi/2$.

Thus, relation (4.8) determines the "rotation angle" ψ and the energy of the electromagnetic field localized in a thin layer of resonant atoms along with it. To determine the energy of an ultrashort optical pulse transmitted through the layer we should use the relation between the transmitted macroscopic wave amplitude E_{tr} and the local field E:

$$E_{tr}=E-\alpha' P_s=(1-\xi_\alpha^0-i\alpha v)E. \tag{4.11}$$

Using solution (4.6) and separating the variables in expression (4.11) we get

$$\begin{aligned}&\mathscr{E}_{tr}\cos(\varphi_{tr}-\varphi)=\mathscr{E}(1-\xi_\alpha^0),\\ &\mathscr{E}_{tr}\sin(\varphi_{tr}-\varphi)=-\xi_\alpha\sin\psi\mathscr{E},\end{aligned} \tag{4.12}$$

whence it follows that for $\xi_\alpha^0\simeq 1$ we have $\varphi_{tr}-\varphi=-\pi/2$ and

$$\psi_{tr}=\frac{\xi_\alpha^2}{2}\psi-\frac{\xi_\alpha^2}{4}\sin 2\psi,$$

then

$$\psi_{tr}=F^2\psi_0-4\xi_\beta^0\xi_\alpha\sin^2\frac{\psi}{2}-\left(\xi_\beta^{02}+\frac{\xi_\beta^2}{2}\right)\psi+\frac{\xi_\beta^2}{4}\sin 2\psi. \tag{4.13}$$

As a result expressions (4.8), (4.9), and (4.13) allow us to determine the energy of an ultrashort optical pulse transmitted through a thin resonant layer as a function of the incident pulse energy.

CONCLUSION

Thus, the inclusion of the dipole-dipole interaction in the effective local Lorentz field does not alter qualitatively the character of optical bistability in the problem of an optical pulse passing through a thin surface layer of resonant atoms for the case of single-quantum resonance in the quasi-steady regime. In the limit of ultrashort pulse lengths, much shorter than the typical times for irreversible relaxation of the polarization, a consistent treatment of phase modulation is needed. Analytical solutions can be obtained in the "small-area" approximation only. In the case of coherent propagation of optical pulses under the conditions of two-quantum resonance, the phenomenon of phase modulation is essential as well. Analytical solutions are obtained only in particular cases.

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