Reflection and transmission of ultrashort light pulses through a thin resonant medium: Local-field effects

M. G. Benedict*
Max Planck Institut für Quantenoptik, Postfach 1513, Ludwig-Prandtl-Strasse 10, D-8046 Garching bei München, Germany

V. A. Malyshev
State Optical Institute, 199034 Leningrad, U.S.S.R.

E. D. Trifonov and A. I. Zaitsev
Department of Theoretical Physics, Herzen Pedagogical Institute, 191186 Leningrad, U.S.S.R.
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We investigate the problem of resonant reflection and transmission of an ultrashort light pulse passing through a thin layer consisting of two-level atoms. The local-field correction that leads to an inversion-dependent resonance frequency generates a new mechanism of nonlinear transparency. When the excitation frequency is somewhat larger than the original resonant frequency, then the transmission of the layer exhibits a transient bistable behavior on the time scale of superradiation, which can be much shorter than the relaxation times. This type of bistability is essentially different from the ordinary stationary intrinsic bistability investigated earlier by other authors.

I. INTRODUCTION

There is a continuing interest in resonant, nonlinear optical properties of thin layers,[1-8] and particularly in their possible optical bistable behavior.[9,10] In most works, the stationary regime is the center of interest, when the duration of the external excitation T_p is much longer than the atomic relaxation times. In this context a possible new mechanism of optical bistability has been proposed and discussed in a series of papers.[3,4,11-16] It is based on the local-field correction to the electric field. As it is known,[17,18] the field acting on the atoms in a medium is different from the averaged macroscopic field (the importance of this effect in resonant interaction has been recognized long ago[19]), and this may give rise to an intrinsic optical bistability in stationary fields.

In the present work we focus our attention on the problem of reflection and transmission of ultrashort pulses: $T_p \ll T_2, T_2^*$, where T_2 and T_2^* are the homogeneous and inhomogeneous dephasing times, respectively. In contrast to our previous works[5-6] and other papers,[1,2,7,8] we take into account the effect of the local-field correction on this process. In connection with this, we derive and use a quantitative estimation for the local field which is different from that of Refs. 3, 4, and 11–14.

We will show that for a sufficiently thin layer the joint effect of the microscopic and macroscopic fields predicts a new mechanism of nonlinear transparency of the medium. This nonlinearity generates an effect which is the analog of the dispersive optical bistability in the stationary case, but it takes place on a much shorter time scale and manifests itself when $\tau_R < T_p < T_2, T_2^*$, where τ_R is the superradiation time of the medium. The reflection and transmission of such fast pulses bear a superradiant character,[5,6] and we show that the transmission of the layer can be multiple valued in this case as well. We call this effect transient bistability. Our approach is therefore different from that of Refs. 3, 4, and 11–16, where bistability was governed by the phase relaxation. We also note that the effect of the local-field correction on another type of coherent interaction of light and matter, namely, on self-induced transparency[20] has already been investigated.[21] Some preliminary results of our work have been published earlier in a short paper.[22]

II. THE EQUATIONS OF THE MODEL

We shall consider the boundary-value problem for the transmission of a light pulse through a plan parallel resonant layer consisting of two-level atoms.[23,24] Let the incoming field be a linearly polarized plane wave

$$\mathcal{E}_i(x,t)=\tfrac{1}{2}E_i(t)e^{-i(kx-\omega t)}+\text{c.c.} \tag{1}$$

with amplitude $E_i(t)$ slowly varying during one optical period $2\pi/\omega$. We restrict ourselves to the case of normal incidence and assume that the transition dipole moments of the atoms are all parallel with the direction of the exciting field. The reflected and transmitted waves will be then also linearly polarized plane waves:

$$\mathcal{E}_r(x,t)=\tfrac{1}{2}E_r(t)e^{-i(kx-\omega t)}+\text{c.c.} \tag{2a}$$

and

$$\mathcal{E}_{\text{tr}}(x,t)=\tfrac{1}{2}E_{\text{tr}}(t)e^{-i(kx-\omega t)}+\text{c.c.}\ , \tag{2b}$$

respectively, with slowly varying amplitudes in time: $E_r(t)$ and $E_{\text{tr}}(t)$. We seek the averaged macroscopic field and the polarization inside the medium in the following form:

$$\mathcal{E}(x,t)=\tfrac{1}{2}E(t)e^{i\omega t}+\text{c.c.}\ , \tag{3a}$$

$$\mathcal{P}(x,t)=\tfrac{1}{2}P(t)e^{i\omega t}+\text{c.c.} \tag{3b}$$

Here we suppose that the amplitudes E and P are also slowly varying functions of the time, but we do not require them to have this property in the space variable. The problem we investigate is one dimensional in space, thus the macroscopic field obeys the inhomogeneous wave equation

$$\left[\frac{\partial}{\partial x^2}-\frac{1}{c^2}\frac{\partial}{\partial t^2}\right]\mathcal{E}=\frac{4\pi}{c^2}\frac{\partial\mathcal{P}}{\partial t^2}\ . \tag{4}$$

The macroscopic polarization at a point x can be calculated in the usual way,[23,24] taking into account that the atoms may have different resonant frequencies ω_{21}:

$$\mathcal{P}=Np\int d\omega_{21}g(\omega_{21})(\rho_{12}+\rho_{21})\ . \tag{5}$$

Here N is the number of atoms in unit volume, p is the transition dipole moment of the atoms, and g is the line shape of the inhomogeneous broadening.

The elements of the density matrix obey the equations

$$\begin{aligned}
i\hbar\frac{\partial\rho_{11}}{\partial t}&=p\mathcal{E}'(\rho_{12}-\rho_{21})+\frac{i\hbar}{T_1}\rho_{22}\ ,\\
i\hbar\frac{\partial\rho_{22}}{\partial t}&=-p\mathcal{E}'(\rho_{12}-\rho_{21})-\frac{i\hbar}{T_1}\rho_{22}\ ,\\
i\hbar\frac{\partial\rho_{12}}{\partial t}&=-\hbar\omega_{12}\rho_{12}-p\mathcal{E}'(\rho_{22}-\rho_{11})-\frac{i\hbar}{T_2}\rho_{12}\ ,\\
\rho_{21}&=\rho_{12}^*\ ,
\end{aligned} \tag{6}$$

where T_1 and T_2 are the relaxation times for the inversion and the polarization, respectively. In Eqs. (6) $\mathcal{E}'$ is the field acting on the given atom. As it is known[17,18] $\mathcal{E}'$ differs from the macroscopic field strength by a local-field correction caused by the dipole-dipole interaction

$$\mathcal{E}'=\mathcal{E}+\frac{4\pi}{3}\mathcal{P}\ . \tag{7}$$

In Ref. 4 a different form has been obtained for this correction in the thin layer limit. In Appendix A we show that even for this case, the original Lorentz result is the correct one.

In agreement with (2) and (3) we write

$$\rho_{12}(x,\omega_{21},t)=\tfrac{1}{2}R(x,\omega_{21},t)e^{i\omega t}+\text{c.c.} \tag{8}$$

Let us introduce the notations $W=\rho_{22}-\rho_{11}$ for the population difference and $E'=E+4\pi P/3$ for the amplitude of the effective field, where

$$P=Np\int d\omega_{21}g(\omega_{21})R\ . \tag{9}$$

Then from Eqs. (6) and (7) we get a system of equations for the slowly varying functions R and W:

$$\frac{\partial R}{\partial t}=\left[i\Delta-\frac{1}{T_2}\right]R+\frac{ip}{\hbar}E'W\ , \tag{10a}$$

$$\frac{\partial W}{\partial t}=\frac{ip}{2\hbar}(E'^*R-E'R^*)-\frac{1}{T_1}(1+W)\ . \tag{10b}$$

Here $\Delta=\omega_{21}-\omega$ is the detuning of the frequency of the incoming field from the atomic resonance.

Using the retarded solution of the wave equation (4), and by the help of (5) and (8) the amplitude of the macroscopic field strength can be written in the form

$$E(x,t)=E_i(x,t)-\frac{2\pi i\omega}{c}Np\int_0^L dx'e^{-i(\omega/c)|x-x'|}\int d\omega_{21}g(\omega_{21})R(x',\omega_{21},t-|x-x'|/c)\ . \tag{11}$$

In the following we shall consider the system of equations (10) and (11), which completely determine our problem. Clearly, the amplitudes of the reflected and transmitted waves can be expressed by the amplitude E in the following way:

$$E_r(t)=E(0,t)-E_i(0,t),\quad E_{\text{tr}}=E(L,t)\ . \tag{12}$$

Even without solving Eqs. (10) and (11), it is possible to draw some conclusions about the physical consequences of the local-field correction. In the case of negligible inhomogeneous broadening we have

$$P=pNR,\quad E'=E+\frac{4\pi}{3}P\ . \tag{13}$$

Inserting E' into (10) we obtain

$$\frac{\partial R}{\partial t}=\left[i\Delta'-\frac{1}{T_2}\right]R+\frac{ip}{\hbar}EW\ , \tag{14a}$$

$$\frac{\partial W}{\partial t}=\frac{ip}{2\hbar}(E^*R-ER^*)-\frac{1}{T_1}(1+W)\ , \tag{14b}$$

where

$$\Delta'=\omega_{21}'-\omega,\quad \omega_{21}'=\omega_{21}+\Delta_L W\ , \tag{15}$$

$$\Delta_L=\frac{4\pi}{3\hbar}p^2N\ . \tag{16}$$

These equations have been derived here semiclassically, a more rigorous QED derivation can be found, e.g., in Ref. 4. They are widely used in the theory of lasers,[23] bistability,[4,9,10] superradiance,[25] and many other nonlinear optical effects.[24]

We see that initially, when the system starts from its ground state $W=-1$, the local-field correction leads to a renormalization of the resonant frequency by an amount of $-4\pi p^2N/\hbar$. Later on it will cause a dynamical shift[12,19] in the resonant frequency, depending linearly on the population difference W. The amount of this frequency change is $8\pi p^2N/\hbar$, as W is ranging between -1 and $+1$. We note that $4\pi p^2N/3$ is of the same order of magnitude as the interaction energy of two equal dipoles of strength p separated by a distance $(4\pi N/3)^{-1/3}$.

As the local-field correction causes a dynamical shift of the atomic resonant frequency, its effect on the interaction will essentially depend on the relative positions of the carrier frequency of the incoming wave ω and the renormalized resonant atomic frequency $\omega'_{21}=\omega_{21}-\Delta_L$. If $\omega \leq \omega'_{21}$, then during the excitation the system will be driven out of resonance even more and will get more transparent. On the contrary, if $\omega > \omega'_{21}$, then the excitation will improve the resonance condition, and accordingly the reflection will be enhanced. We will show below that this process of pulling into resonance has a threshold character.

In the remainder of this section we briefly summarize the case of weak fields, i.e., the linear limit, when we may set $W=-1$. In the stationary situation Eqs. (14a) and (13) lead to a linear relationship between the field strength and the polarization $P=\chi E$, while the index of refraction is given by

$$n=(1+4\pi\chi)^{1/2}=\left[1+\frac{2}{kL\tau_R}\frac{1}{\Delta'+i/T'_2}\right]^{1/2} . \quad (17)$$

Here we have introduced the following notations:

$$\Delta'=\Delta_0-\Delta_L, \quad \frac{1}{T'_2}=\frac{1}{T_2}+\frac{1}{T_2^*} , \quad (18)$$

where $\Delta_0=\omega_0-\omega$ is the detuning of the resonant frequency from the center of the inhomogeneous line. The latter was supposed to be a Lorentzian of linewidth $2/T_2^*$. τ_R is the superradiation time of the layer:[6]

$$\tau_R^{-1}=\frac{2\pi p^2 NkL}{\hbar}=\tfrac{3}{2}\Delta_L kL . \quad (19)$$

The laws of reflection and transmission of the medium can be deduced in the usual way,[17] or by the help[5] of the integral equation (11). If the incident pulse is ultrashort, $T_p \ll T'_2$, then the considerations above need to be refined. The spectral width of such a pulse, T_p^{-1}, is much larger than the width of the absorption line $T_2'^{-1}$. In this case the index of refraction must be considered as a function of the frequency components of the pulse. So, one has to determine the reflection and transmission coefficients for each spectral component, and then for the whole pulse. The other possibility is to remain in the time domain and solve the coupled equations (10) and (11). Going over to the nonlinear case below, we shall follow this latter route.

III. NONLINEAR TRANSMISSION AND BISTABILITY

Turning to the investigation of strong pulses already causing a significant inversion, we restrict ourselves to the case of a thin layer, for which the spatial dependence of the field and the polarization can be neglected. As it is known from linear optics, this case is determined by the inequality $|n(\omega)kL| \ll 1$. A more exact estimation will be given in Appendix B. For a thin layer the system of equations (10) and (11) takes an essentially simpler form. In this case the integral equation for the field strength reduces to a simple algebraic equation:

$$E=E_i-2\pi ikLP . \quad (20)$$

When deriving (20) from (11) we have approximated the exponential in the integrand by unity, and we have neglected the dependence of all the amplitudes on the spatial variable x. Taking into account also the local-field correction, the actual field strength acting on the atoms has the form

$$E'=E_i-2\pi ikLP+\frac{4\pi}{3}P . \quad (21)$$

The second term in this equation thus can be obtained already from the macroscopic Maxwell equations, while the local-field correction is a result of microscopic effects. A unique derivation of both terms is given in Appendix A.

To simplify notation, instead of the field amplitudes we shall use the appropriate Rabi frequencies: $\epsilon_i=pE_i/\hbar$, $\epsilon=pE/\hbar$, and $\epsilon'=pE'/\hbar$. Then our system of equations describing the interaction of the light pulse with the thin layer takes the following form:

$$\frac{\partial R}{\partial t}=\left[i\Delta'-\frac{1}{T_2}\right]R+i\epsilon' W , \quad (22a)$$

$$\frac{\partial W}{\partial t}=\frac{i}{2}(\epsilon'^* R-\epsilon' R^*)-\frac{1}{T_1}(1+W) , \quad (22b)$$

$$\epsilon'=\epsilon_i+\left[\Delta_L-\frac{i}{\tau_R}\right]\langle R\rangle . \quad (22c)$$

Here again the angular brackets denote the average over the inhomogeneous line.

If $kL \ll 1$, then from Eq. (19) it follows that $\Delta_L \gg \tau_R^{-1}$. Therefore for a thin layer ($L \ll \lambda$) the local-field contribution $\Delta_L\langle R\rangle$ to the effective field ϵ' is dominant over the radiation field $-i\langle R\rangle/\tau_R$, as far as the absolute values are concerned. Nevertheless both terms are important, because of the $\pi/2$ phase shift between them.

The local correction generates a shift in the resonant frequency, while the radiation field, as it will be shown below, induces a collective radiative relaxation of the polarization and of the inversion with a time constant τ_R.

Let us consider an ultrashort light pulse, the duration of which is less than the relaxation times, but is longer than the superradiation time: $\tau_R < T_p < T'_2, T_1$. In the linear case this relationship between the time constants has been the condition of strong reflection. What happens if we increase the intensity of the incoming pulse, so that nonlinearity is supposed to play a role? For an ultrashort pulse the system of equations (22) can be written in the form

$$\frac{\partial R}{\partial t}=\left[i\Delta'+\frac{1}{\tau_R}W\right]R+i\epsilon_i W, \quad \Delta'=\Delta+\Delta_L W , \quad (23a)$$

$$\frac{\partial W}{\partial t}=\frac{i}{2}\epsilon_i(R-R^*)-\frac{1}{\tau_R}|R|^2 , \quad (23b)$$

where the amplitude ϵ_i was taken to be real and $\langle R\rangle=R$. As it follows from Eq. (23a), if the phase

memory of the atomic system is conserved, $(T_1, T_2', T_2^* \to \infty)$, then the time scale of the radiation relaxation is $\tau_R/|W|$. This quantity as well as the shift of the resonant frequency $\Delta_L W$ depend on the inversion W. Therefore the width of the resonance will be determined by the quantity $|W|/\tau_R$.

A. Resonant excitation, nonlinear transparency

Let the incoming field be detuned from the transition frequency so that $\omega=\omega_{21}-4\pi p^2 N/3\hbar$ or $\Delta_0'=\Delta-\Delta_L=0$. If the amplitude of the exciting wave is not too large, $\epsilon_i<\tau_R^{-1}$, and the detuning is not significant during the process, $\Delta'<\tau_R^{-1}$, then the pulse is strongly reflected from the layer.[5,6] If we increase the incoming amplitude and accordingly the degree of the excitation, then the atomic system will be driven out of resonance, and instead of reflecting the layer will be transparent.

To estimate the detuning Δ' at which the transparency becomes significant, let us use the linear approximation: Eq. (17) with $T_2' \to \infty$. The condition of getting comparable reflection and transmission coefficients for a thin layer ($|n|kL \ll 1$) has the form $|n|^2 kL \simeq 1$. According to Eq. (17) this is equivalent to the relation $\Delta'\tau_R \simeq 1$. In this way, the transparency of the layer will be significant, when the frequency shift induced by the field is comparable or larger than the width of the resonance. As $\Delta'=(1+W)\Delta_L=2\rho_{22}\Delta_L$, we obtain an estimate for the corresponding population of the upper state: $\rho_{22}\simeq(\Delta'\tau_R)^{-1}kL \ll 1$. As it is known, for extended systems transparency is connected either with saturating fields equalizing the ground- and excited-state populations ($\rho_{22}\simeq\frac{1}{2}$), or with the effect of self-induced transparency.[20] In both cases the variation of the population is large: $\delta\rho_{22}$ is the order of unity. In contrast to this, in our case transparency appears already at a negligible inversion $\rho_{22}\simeq kL \ll 1$. (The enhancement of the resonant nonlinear susceptibility caused by dipole-dipole interaction has also been noted in Refs. 26 and 27.)

In order to obtain a more detailed picture of the response of the resonant thin layer, we have calculated numerically the time dependences of the transmission coefficient $\mathcal{T}=|\epsilon_{tr}/\epsilon_i|^2$ and of the inversion W. The results are shown in Fig. 1. This figure demonstrates the buildup of a stationary $\mathcal{T}$ and W, after switching on the external field with different constant amplitudes. *We must emphasize that stationarity is understood here, of course, on a time scale longer than τ_R, but shorter than T_2'.* As it can be seen, the numerical results justify the conclusion about the growth of the transparency caused by increasing the incident amplitude. A weak incoming wave will be totally reflected after a time τ_R needed for the buildup of the polarization (curve 1 of Fig. 1). Increasing the amplitude, the layer becomes more and more transparent (curves 2–4). This transparency arises at a negligible excitation of the atoms; the inversion remains close to its initial value -1. The calculations also show that the stationary transmission and inversion are achieved not monotonically but exhibiting damped optical nutations.

The expression for the stationary value for the reflected and transmitted waves, as well as for the transmission coefficient $\mathcal{T}$, can be found from the system of equations (23). Setting the derivatives equal to zero, we get from the first equation

$$R_{st}=-\frac{iW_{st}}{i\Delta'+W_{st}/\tau_R}\epsilon_i \ . \tag{24}$$

The second equation is satisfied identically. The system, however, has a constant of motion $|R|^2+W^2=1$, and this allows one to get an expression for W_{st}:

$$(1-W_{st}^2)\left[(\Delta')^2+\frac{W_{st}^2}{\tau_R^2}\right]-\epsilon_i^2 W_{st}^2=0 \ . \tag{25}$$

We note that this is a fourth-order equation in W_{st}, while in the case of ordinary stationary intrinsic bistability the corresponding equation is of the third order.

The amplitudes of the reflected and transmitted fields can be written in the form

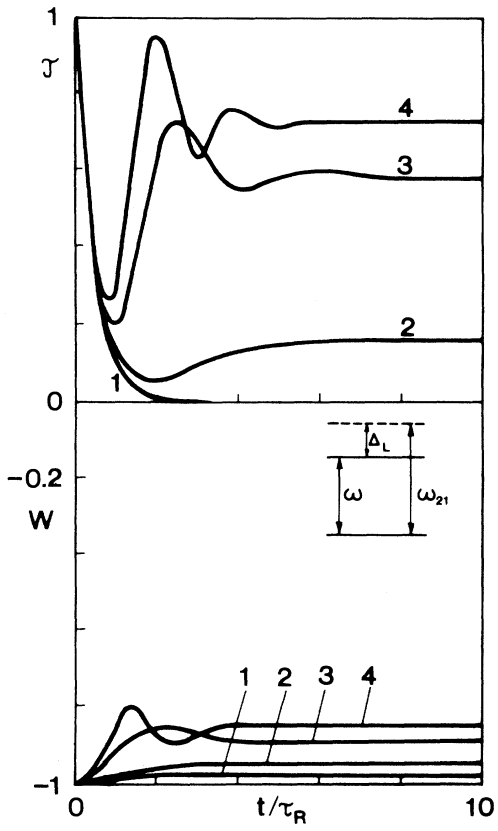

FIG. 1. Buildup of the stationary transmission coefficient $\mathcal{T}=|\epsilon_{tr}/\epsilon_i|$ and of the inversion W of a thin layer: $kL=0.066\,667$, $\Delta_L\tau_R=2/3kL=10$; after switching on a resonant ($\Delta_0'=0$) step pulse of constant amplitude: $\epsilon_i=\epsilon_0/\tau_R$. Curve 1, $\epsilon_0=0.1$; curve 2, $\epsilon_0=0.3$; curve 3, $\epsilon_0=0.7$; curve 4, $\epsilon_0=1.0$.

$$\epsilon_r = -\frac{i}{\tau_R} R_{\rm st} = -\frac{iW_{\rm st}/\tau_R}{i\Delta' + W_{\rm st}/\tau_R}\epsilon_i \ , \tag{26a}$$

$$\epsilon_{\rm tr} = \epsilon_i - \frac{i}{\tau_R} R_{\rm st} = -\frac{i\Delta'}{i\Delta' + W_{\rm st}/\tau_R}\epsilon_i \ , \tag{26b}$$

respectively. This yields the following transmission coefficient:

$$\mathcal{T} = \frac{(\Delta')^2}{(\Delta')^2 + (W_{\rm st}/\tau_R)^2} \ . \tag{27}$$

Before we turn to the detailed analysis of these results, let us first comment on their range of validity. Though they were deduced for the stationary regime $\epsilon_i = \text{const}$ and $t \to \infty$, they also can be used to describe the transmission of pulses obeying the inequality $T_2' \gg T_p \gg \tau_R$, because in such cases the polarization will follow the variation of the electric field adiabatically.

Let us turn now to the analysis of Eq. (27). It shows that the transmission will be comparable with the reflection ($\mathcal{T} \simeq \frac{1}{2}$), when $(\Delta')^2 = W_{\rm st}^2/\tau_R^2$, i.e., when the frequency shift Δ' induced by the field becomes equal to the width of the resonant frequency $|W_{\rm st}|/\tau_R$. Solving this equation, and taking into account that $\Delta = \Delta_L$, $\Delta' = (1 + W_{\rm st})/\Delta_L = 2\rho_{22}\Delta_L$, and $\Delta_L \tau_R = 2/3kL \gg 1$, we find that $W_{\rm st} = -1 + 1/(\Delta_L \tau_R)$ or $\rho_{22} = 1/(2\Delta_L \tau_R)$. In this way, the onset of the transparency begins already at small values of the inversion, which is in accordance with our previous qualitative arguments.

It is more comfortable to express the transmission coefficient by the population of the excited states ρ_{22}. Using the smallness of this quantity, we obtain from Eq. (27)

$$\mathcal{T} = \frac{\rho_{22}^2}{\rho_{22}^2 + (3kL/4)^2} \ . \tag{28}$$

The equation for ρ_{22} follows from (25), and in the limit $\rho_{22} \ll 1$ it has the form

$$\rho_{22}[\rho_{22}^2 + (3kL/4)^2] = (3kL/8)^2(\epsilon_i \tau_R)^2 \ . \tag{29}$$

With the help of this result, one can estimate the amplitude of the field inducing the transparency. Setting the limit between low and high transmission at $\mathcal{T} = \frac{1}{2}$, i.e., at $\rho_{22} = 3kL/2$, for the threshold for high transparency we get $\epsilon_i \tau_R \geq (6kL)^{1/2}$.

B. Nonresonant excitation, bistability of reflection, and transmission

We have considered above a resonant excitation. The frequency of the external field was equal to the renormalized atomic transition frequency: $\omega_{21}' = \omega_{21} - \Delta_L$. Now let us turn to the nonresonant case $\Delta_0' \neq 0$. It is clear that if the frequency of the external field ω falls into the range below the atomic transition, then the incoming wave drives the atoms further out of resonance, and therefore weakens the reflection and amplifies the transmission.

Principally new effects arise in the opposite case, when the frequency of the exciting pulse is above the atomic resonance ω_{21}' (see Fig. 2). In this case the dynamical

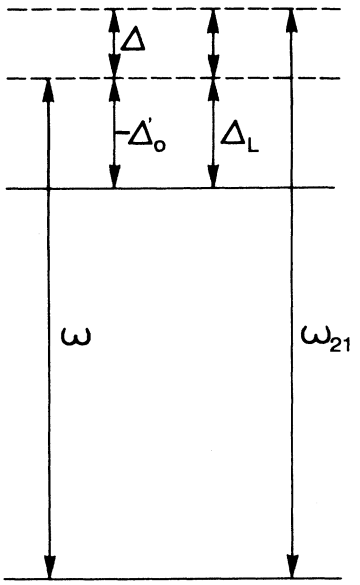

FIG. 2. The scheme of nonresonant excitation to observe optical bistability in the transmission of the resonant layer.

phase shift can compensate the initial detuning, and this leads to a stronger reflection. As it will be shown below, in this case it is possible to obtain a sudden transition between states of low and high transmissions depending either on the initial detuning or on the external field amplitude.

Suppose that the incoming wave has a constant amplitude $\epsilon_i = \epsilon_0/\tau_R$ (ϵ_0 is a dimensionless constant), and it is tuned above resonance: $\Delta_0' < 0$. Let us consider the transient stationary regime of the transmission: $\dot{R} = 0$, $\dot{W} = 0$. In this case in Eqs. (24) and (27) $\Delta' = \Delta_0' + (1 + W_{\rm st})\Delta_L$, $\Delta_0' = \Delta - \Delta_L < 0$. What are the conditions for Eq. (25) to have only real roots? We will be interested only in that range of the parameters for which the inversion remains close to its initial value, $W = -1$. We recast Eq. (25) by expressing the inversion through the population of the upper state, $\rho_{22} = (1 + W)/2$, and assume that $\rho_{22} \ll 1$. Taking into account also that $\Delta' = \Delta_0' + 2\rho_{22}\Delta_L$ we obtain

$$4\rho_{22}[(\Delta_0' + 2\Delta_L\rho_{22})^2 + (1/\tau_R)^2] = \epsilon_i^2 \ . \tag{30}$$

This equation has either a single real root or three real roots. In the first case the function $\rho_{22}(\epsilon_i)$ is single valued. In the second case, however, to some ϵ_i there corresponds three different populations of the upper level. This latter case indicates the possibility of a bistable behavior of the system.

The condition of obtaining three real roots can be found by determining the zeros of the derivative $d\epsilon_i^2/d\rho_{22}$. Introducing the notation $z = \Delta_0' + 2\rho_{22}\Delta_L$ we get

$$3z^2 + 2\Delta_0' z + (1/\tau_R)^2 = 0 \ . \tag{31}$$

Equation (40) will have three different real roots if

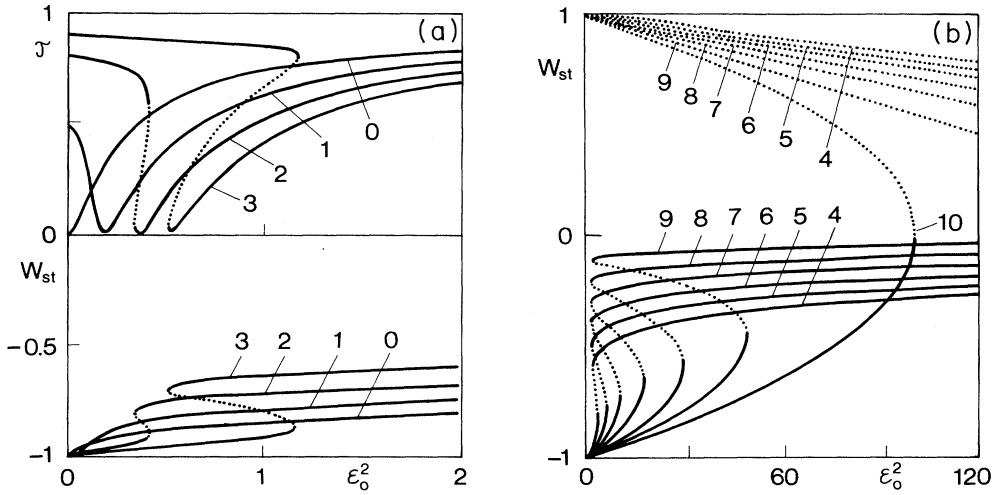

FIG. 3. The dependence of the inversion and of the transmission coefficient on the dimensionless intensity of the excitation: $\epsilon_0^2=(\epsilon_i\tau_R)^2$. The numbers denote the corresponding values of the detuning $-\Delta_0'\tau_R$.

$$\Delta_0' < -\sqrt{3}/\tau_R \ . \tag{32}$$

In this case the plot of the function $\rho_{22}(\epsilon_i)$ shows a wrinkle. The boundaries of the interval where this function is three-valued can be determined by substituting the roots of Eq. (31) into Eq. (30). The smallness of the roots of Eq. (30) is ensured by demanding the inequality $|\Delta_0'| \ll \Delta_L$. According to (32) this means that the shift $|\Delta_0'|\tau_R$ can be of the order of unity.

Figure 3 shows some plots of the function $W_{st}(\epsilon_0^2,\Delta_0')$ illustrating the above analytical considerations. *The figures were drawn using the exact equation* (25), which is of fourth order, in contrast to the approximate Eq. (30), which is only of third order. Therefore in a certain range of the variables ϵ_0^2 and Δ_0' the function W_{st} is four-valued. The instable values have been plotted with dotted lines.

The dependence of the transmission coefficient $\mathcal{T}$ on the dimensionless intensity ϵ_0^2 is shown in the upper half of Fig. 3(a). It can be seen that beginning from some value of Δ_0' (in the figure at $\Delta_0'=-2/\tau_R$), there is a hysteresis in the transmission.

The S-shaped form of the function W_{st} means a multivalued (bistable) response of the system. As an illustration, in Fig. 4 we show the buildup of the stationary transmission coefficient $\mathcal{T}$ and inversion W_{st} for a thin layer ($kL=0.066\,67$, $\Delta_L\tau_R=10$), after switching on a nonresonant field ($\Delta_0'\tau_R=-2.2$) with a constant amplitude $\epsilon_0=\epsilon_i\tau_R$. As it can be seen from Fig. 4, when ϵ_0 gets larger than a certain value (for the parameters we have chosen if $\epsilon_0>0.725$) the character of the response suddenly changes from an almost total transmission to a strong reflection. The corresponding jump can be observed also in the behavior of the inversion W. We can calculate numerically the dynamical frequency shift during the process. In the range above the threshold (in our case when $\epsilon_0>0.725$) the initial detuning has been almost perfectly compensated by the dynamical phase shift; for

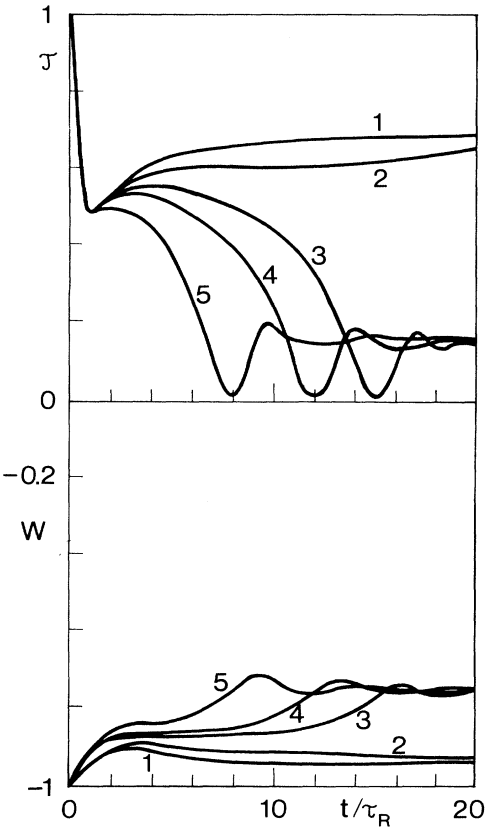

FIG. 4. The sudden change of the kinetics of the transmission coefficient of the thin layer ($kL=0.066\,667$) for a nonresonant excitation ($\Delta_0'\tau_R=-2.2$) of constant amplitude $\epsilon_i=\epsilon_0/\tau_R$. Curve 1, $\epsilon_0=0.72$; curve 2, $\epsilon_0=0.725$; curve 3, $\epsilon_0=0.7275$; curve 4, $\epsilon_0=0.73$; curve 5, $\epsilon_0=0.74$.

the stationary value of $(\Delta'-\Delta_0)\tau_R$ we have obtained 2.3. This means that in the stationary regime the atoms are in resonance with the external field, which results in a strong reflection. In between, the departure of the inversion from its initial value remained relatively small.

The bistability in the transmission of the resonant layer considered here bears a purely dispersive character, because we did not take into account the relaxation processes: $1/T_1$ and $1/T_2'$ were set equal to zero. Our approach is therefore different from that of Refs. 3, 4, and 11–16, where bistability was studied on the time scale of the phase relaxation T_2. In order to realize bistability in that case one needs much stronger, saturating fields.

All that has been stated is valid for light pulses which are longer than the time of stationary response τ_R, but shorter than the relaxation times T_1 and T_2'. Let us remind the reader that there is another restriction we have been using: the incident wave can be reflected only if $\epsilon_i < \tau_R^{-1}$. This is connected with the fact that the induced polarization of the layer cannot generate a secondary field with an amplitude larger than τ_R^{-1}. Therefore the contrast in bistability will be high only if $\epsilon_i < \tau_R^{-1}$.

IV. CONCLUSIONS

Describing the nonlinear optical properties of optically thin resonant layers it is principally important to take into account the dipole-dipole interaction of the atoms. This interaction can be effectively taken into account in the optical Bloch equations by replacing the macroscopic mean-field $\mathcal{E}$ with the effective local-field $\mathcal{E}'=\mathcal{E}+4\pi\mathcal{P}/3$. This correction may lead to several interesting physical effects. We were dealing especially with the following ones, which can be important with respect to possible future applications: (i) the nonlinear transparency of a thin resonant layer remaining near to its ground state, and (ii) the transient bistability of the transmission. Both effects are induced by the dynamical frequency shift with a switching time shorter than the relaxation times. They manifest themselves when certain relations between the parameters of the atomic system and the incident field are fulfilled. The duration of the excitation T_p must be larger than the superradiation time of the layer τ_R and at the same time it is supposed to be shorter than the relaxation times of the atomic system. In addition, the Rabi frequency of the incident pulse must be less than τ_R^{-1}. Thus the conditions to observe the predicted effects are $\tau_R < T_p < T_2^*, T_2'$ and $\epsilon_i < \tau_R^{-1}$. We propose that the effects predicted above might be realized experimentally using excitonic lines of aromatic compounds[28] (such as naphtalene and anthracene), or in materials[29] containing unoccupied d or f orbitals as Cr_2O_3 or MnO_2. The lowest electronic excitations in these materials are excitons of small radius. The presence of the excitonic lines shows that the dipole-dipole interaction is larger than the homogeneous and inhomogeneous broadenings, (which is impossible in gases[15]) and this makes us hopeful that the required inequalities will be satisfied.

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APPENDIX A

Following Ref. 4, we calculate the effective field $\mathcal{E}'$ at the place of the ith atom generated by all the other dipoles. We suppose that each dipole points into the x direction and we neglect the inhomogeneous broadening. We shall calculate the quantity $\epsilon'=\mathbf{p}\cdot\mathbf{E}'/\hbar$, where $\mathbf{E}'$ is the slowly varying amplitude of the field $\mathcal{E}'$.

We consider a plane cylindrical layer of thickness $L \ll \lambda$ and radius R_0. The origin of coordinates we choose in the center of the cylinder. Let us find the field acting on an atom on the symmetry axis (z direction) by the familiar procedure.

We cut out a spherical volume of a small but macroscopic radius δ around the location of the ith atom. The field of the dipoles generated by the atoms within the sphere is zero[17,18] at the point $\mathbf{r}_i$. The field of the dipoles outside the sphere can be calculated in the continuous approximation. Then

$$\epsilon' = -i\Gamma R \ , \tag{A1}$$

$$\Gamma = \sum_{j(\neq i)} B_{ij} = N\int_{V-V_\delta} B(r_{ij})dV_j \ , \tag{A2}$$

where the integration is to be performed over the volume of the cylindrical volume with the spherical hole V_δ in it. The matrix B_{ij} is given by the expression[4]

$$B_{ij} = \tfrac{3}{2}\beta\{[(\boldsymbol{l}_i\cdot\boldsymbol{l}_j)-(\boldsymbol{l}_i\cdot\mathbf{n}_{ij})(\boldsymbol{l}_j\cdot\mathbf{n}_{ij})]F_1(kr_{ij}) + [(\boldsymbol{l}_i\cdot\mathbf{n}_{ij})(\boldsymbol{l}_j\cdot\mathbf{n}_{ij})]F_2(kr_{ij})\} \ , \tag{A3}$$

$$F_1(\xi) = e^{-i\xi}\left(\frac{i}{\xi}-\frac{i}{\xi^3}+\frac{1}{\xi^2}\right) , \tag{A4}$$

$$F_2(\xi) = e^{-i\xi}\left(\frac{2i}{\xi^3}-\frac{2}{\xi^2}\right) . \tag{A5}$$

Here $\beta=2p^2k^3/3\hbar$, $\boldsymbol{l}_i$ is the unit vector pointing into the direction of $\mathbf{p}_i$ (in our case $\boldsymbol{l}_i=\boldsymbol{l}_j=\boldsymbol{l}$), $\mathbf{r}_{ij}=\mathbf{r}_i-\mathbf{r}_j$, $\mathbf{n}_{ij}=\mathbf{r}_{ij}/r_{ij}$.

In cylindrical coordinates (ρ,φ,z)

$$\boldsymbol{l}_i\cdot\mathbf{n}_{ij} = -\frac{\rho_j}{r_{ij}}\cos\varphi_j, \quad r_{ij}^2=\rho_j^2+(z_j-z_i)^2 \ . \tag{A6}$$

The ρ_i coordinate does not enter expression (A6), because the ith atom is supposed to be on the axis of the layer, $\rho_i=0$. It is appropriate here to point out an error made in Ref. 4, where the factor ρ_j/r_{ij} in (A6) has been omitted in the calculation. This inaccuracy, as it will be shown below, leads to a substantial difference in the result for Γ.

After integrating (A2) over φ_j and using (A6) we get

$$\Gamma=\tfrac{3}{2}\pi N\beta\left[\left[\int_{-L/2}^{z_i-\delta}dz_j+\int_{z_i-\delta}^{L/2}dz_j\right]\int_0^{R_0}\rho_j d\rho_j + \int_{z_i-\delta}^{z_i+\delta}dz_j\int_{[\delta^2+(z_i-z_j)^2]^{1/2}}^{R_0}\rho_j d\rho_j\right] \times\left[F_1(kr_{ij})+F_2(kr_{ij}) + \frac{(z_i-z_j)^2}{r_{ij}^2}[F_1(kr_{ij})-F_2(kr_{ij})]\right] . \quad \text{(A7)}$$

Substituting $\rho_j^2=r_{ij}^2-(z_j-z_i)^2$ and using the relations[4]

$$\xi[F_1(\xi)+F_2(\xi)]=-\frac{d}{d\xi}\left[e^{-i\xi}\left[1+\frac{i}{\xi}\right]\right] , \quad \text{(A8)}$$

$$\frac{1}{\xi}[F_1(\xi)-F_2(\xi)]=\frac{d}{d\xi}\left[e^{-i\xi}\left[\frac{i}{\xi^3}-\frac{1}{\xi^2}\right]\right] , \quad \text{(A9)}$$

one can perform the integration over ρ_j in (A7). We obtain

$$\Gamma=\tfrac{3}{2}\pi\beta\frac{N}{k^2}\left[\int_{-L/2}^{z_i-\delta}dz_j+\int_{z_i+\delta}^{L/2}dz_j\right][\Phi(k[R_0^2+(z_i-z_j)^2]^{1/2},k(z_j-z_i))-\Phi(k|z_j-z_i|,k(z_j-z_i))] + \tfrac{3}{2}\pi\beta\frac{N}{k^2}\int_{z_i-\delta}^{z_i+\delta}dz_j[\Phi(k[R_0^2+(z_i-z_j)^2]^{1/2},k(z_j-z_i))-\Phi(k\delta,k(z_j-z_i))] , \quad \text{(A10)}$$

where

$$\Phi(\xi,\eta)=-\left[1+\frac{i}{\xi}+\frac{\eta^2}{\xi^2}\left[1-\frac{i}{\xi}\right]\right]e^{-i\xi} . \quad \text{(A11)}$$

The integrations of the functions $\Phi(k|z_j-z_i|,k(z_j-z_i))$ and $\Phi(k\delta,k(z_j-z_i))$ also can be performed in explicit form, so that for Γ we obtain

$$\Gamma=\frac{3}{2}\pi\beta\frac{N}{k^2}\left[-\frac{8}{3k}(i-k\delta)e^{-ik\delta}+\frac{4i}{k}e^{-ikL/2}\cos kz_i+\int_{-L/2}^{-L/2}dz_j\Phi(k[R_0^2+(z_i-z_j)^2]^{1/2},k(z_j-z_i))\right] . \quad \text{(A12)}$$

So far we have not used any approximation in the calculations, therefore the expression for Γ is exact in the framework of the present model. Let us turn attention to the fact that Γ remains finite even in the limit $\delta\to 0$:

$$\Gamma=\tfrac{3}{2}\pi\beta\frac{N}{k^2}\left[-\frac{8i}{3k}+\frac{4i}{k}e^{-ikL/2}\cos kz_i+\int_{-L/2}^{-L/2}dz_j\Phi(k[R_0^2+(z_i-z_j)^2]^{1/2},k(z_j-z_i))\right] . \quad \text{(A13)}$$

From this equation in the limit of a thin layer $kL\ll 1$, $k|z_i|\ll 1$ it follows that

$$\Gamma=i\frac{2\pi\beta N}{k^3}+\frac{3\pi\beta NL}{k^2}+\frac{3\pi\beta N}{2k^2}\int_{-L/2}^{-L/2}dz_j\Phi(k[R_0^2+(z_i-z_j)^2]^{1/2},k(z_j-z_i)) . \quad \text{(A14)}$$

For the sake of comparison we quote the corresponding result of Ref. 4:

$$\Gamma=i\frac{3\pi\beta N}{k^3}\left[1+\ln\frac{(L^2/4-z_i^2)^{1/2}}{\delta}\right] . \quad \text{(A15)}$$

Here the terms proportional to kL have already been omitted, because in Ref. 4 the case $kL\to 0$ has been considered. Therefore we have to compare (A15) with the first term of (A14). This is the term which embodies the difference between the effective and the macroscopic field. We see that the important difference between our result and (A15) is that the latter is divergent when δ, the radius of the hollow sphere, goes to zero. This singularity is the consequence of the inaccuracy pointed out above.

Remembering that $\beta=2p^2k^3/3\hbar$, the first term in (A14) can be written in the form $4\pi ip^2N/3\hbar$. The corresponding contribution to the effective field ϵ' is $4\pi p^2NR/3\hbar$. This expression is in full accordance with the standard result for the local-field correction $4\pi P/3=4\pi pN/3$. The second term in (A14) is the reaction field $-2\pi ikLP$, while the third term can be shown to be negligible in the limit $R_0\to\infty$.

APPENDIX B

As it has been noted in the beginning of Sec. III, in the linear approximation the resonant layer can be regarded to be thin if its thickness L is less than the wavelength of the light within the medium divided by 2π, i.e., if $|n|kL\ll 1$, where n is the complex index of refraction. Here we will obtain a more general result being valid for the nonlinear case as well.

In considering the limit of the thin layer, we have used the algebraic expression (20) for the field, instead of the integral equation (11). At first sight the condition that $L<\lambda/2\pi$ (and not $L<\lambda/2\pi n$), seems to be sufficient to validate this approximation. Replacing the phase factor by unity in the integrand of Eq. (11) leads to a uniform field strength in the medium, and via the material equa-

tions (10) also to the uniformity of the inversion W and the polarization R. In order to understand the situation, let us estimate the omitted terms. It is clear that they are of the order of $2\pi pN(kL)^2\langle R\rangle$, and they must be small compared with the terms kept in the equation. Therefore the condition we seek has the form

$$kL\left|\frac{\langle R\rangle}{\epsilon_i\tau_R-\langle R\rangle}\right|\ll 1\ . \tag{B1}$$

It is not difficult to prove that in the linear case, when $\langle R\rangle=(\chi/pN)E$ ($\chi\gg 1$), the condition (B1) is equivalent to $|n|kL\ll 1$.

*Present address: Department of Theoretical Physics, József Attila University, H-6701 Szeged, P.O. Box 428, Hungary.

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