Population trapping and coherent bleaching of a three-level medium by a periodic train of ultrashort pulses

O. A. Kocharovskaya and Ya. I. Khanin

Institute of Applied Physics, Academy of Sciences of the USSR, Gorki
(Submitted 15 November 1985)
Zh. Eksp. Teor. Fiz. **90**, 1610–1618 (May 1986)

An analysis is made of the propagation of a train of ultrashort pulses in a resonantly absorbing medium with two sublevels of the ground state optically coupled to a third level. It is shown that under steady-state conditions the way such a train propagates and the parameters of the medium depend resonantly on the relationship between the frequency representing the splitting of the ground state ω_{21} and the pulse repetition frequency Ω. Under resonance conditions described by $\omega_{21} = m\Omega$ ($m = 1,2,\ldots$) a sufficiently strong pulse train may result in coherent trapping of populations, i.e., it may transfer molecules to a coherent superposition state of the lower sublevels, leaving the upper sublevel almost empty. Coherent bleaching of a three-level medium by a resonant train of ultrashort pulses is predicted and it is shown that this effect is possible even when the radiation intensity is much less than that needed for saturation of an optical transition.

§1. INTRODUCTION

Resonant coherent effects in three-level media have been investigated beginning from the fifties (see, for example, Refs. 1–3 and also Ref. 4 and the literature cited there). In recent years more attention has been given to one of the variants known as the λ scheme, when two sublevels of the ground state are coupled optically to an excited level (Fig. 1).

A theoretical analysis of the λ scheme has been limited so far to situations in which a three-level medium interacts with two monochromatic fields or with two pulses, and in each case the interaction is via only one of the optical transitions which can be induced by such a field or pulse. This implies that the spectra of the fields are narrow and that the spectral lines are broadened homogeneously compared with the separation (splitting) between the sublevels ω_{21}. In the case of pulsed fields this restriction can be written in the form of the inequality $\tau_p^{-1}, T_2^{-1} \ll \omega_{21}$, where τ_p is the pulse duration and T_2 is the transverse relaxation time. In this case we can expect coherent trapping of populations by the ground-state sublevels and associated resonances of coherent nonabsorption of the optical radiation interacting with the medium.[5–7] These effects are due to resonant excitation of low-frequency coherence (represented by an off-diagonal element ρ_{21} of the density matrix) when the differences between the carrier frequencies are equal to ω_{21}. Such effects have been investigated extensively both theoretically and experimentally because of the great variety of applications in ultrahigh-resolution spectroscopy,[8] frequency stabilization,[9,10] competing two-photon ionization spectroscopy schemes,[11,12] optical bistability systems,[13,14] cooling of atoms,[15] etc. Another interesting problem is the propagation in a three-level medium of a simulton, i.e., a soliton consisting of a pair of pulses with carrier frequencies coincident with the frequencies of two optical transitions (see the review in Ref. 16).

In the opposite case, when $\tau_p^{-1}, T_2^{-1} \gg \omega_{21}$, each pulse interacts simultaneously with both optical transitions. Isolation of two carriers separated from one another by a frequency of the order of ω_{21} is then pointless. Nevertheless, it is under these conditions that we can expect manifestation of resonant coherent effects when a three-level medium interacts with optical radiation if this radiation is in the form of a periodic sequence of ultrashort pulses and if the splitting frequency ω_{21} is a multiple of the pulse repetition frequency Ω. A similar set of conditions was clearly satisfied also in the experiments described in Ref. 17, when mode-locked dye laser radiation tuned to the D_1 line traversed a cell filled with sodium vapor and the resonances of the response of the medium observed on variation of the pulse repetition frequency were used to determine the Zeeman splitting of the ground-state sublevels.

In the present paper we shall consider theoretically the resonances which accompany the interaction of a three-level medium with a periodic sequence of pulses in the case when $\tau_p^{-1}, T_2^{-1} \gg \omega_{21}$. We shall show that when the power is sufficiently high, such a resonant pulse train may cause coherent population trapping, i.e., it may transfer molecules to a coherent superposition state of the lower sublevels leaving the upper level almost empty. This makes possible propagation

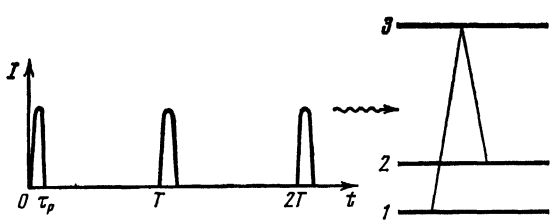

FIG. 1. Schematic representation of the propagation of a train of ultrashort pulses in a three-level medium exhibiting a frequency splitting ω_{21} which is a multiple of the pulse repetition frequency $\Omega = 2\pi/T$.

 0038-5646/86/050945-06$04.00

of a train of ultrashort pulses across a layer of a three-level medium practically without absorption, which can be called coherent bleaching of a three-level medium.

§2. INITIAL EQUATIONS

When a three-level medium interacts resonantly with both optical transitions, reducing the initial equations for the density matrix[18] for the field carrier frequency ω, we obtain

$$\frac{\partial\sigma_{31}}{\partial t}+i(\omega_{31}-\omega)\sigma_{31}=-\frac{\sigma_{31}}{T_2}-\frac{i}{2T_2}(n_{31}-\rho_{21})\frac{E}{E_n},$$

$$\frac{\partial\sigma_{32}}{\partial t}+i(\omega_{32}-\omega)\sigma_{32}=-\frac{\sigma_{32}}{T_2}-\frac{i}{2T_2}(n_{32}-\rho_{21}^{*})\frac{E}{E_n},$$

$$\frac{\partial\rho_{21}}{\partial t}+i\omega_{21}\rho_{21}=-\frac{\rho_{21}}{\tau_2}+\frac{i}{2T_1}\frac{(\sigma_{31}E^{*}-\sigma_{32}^{*}E)}{E_n}, \qquad (2.1)$$

$$\frac{\partial n_{31}}{\partial t}=-\frac{1+n_{31}+n_{32}}{2T_1}+\frac{i(\sigma_{31}^{*}E-\sigma_{31}E^{*})}{T_1E_n}+\frac{i(\sigma_{32}^{*}E-\sigma_{32}E^{*})}{2T_1E_n},$$

$$\frac{\partial n_{32}}{\partial t}=-\frac{1+n_{31}+n_{32}}{2T_1}+\frac{i(\sigma_{32}^{*}E-\sigma_{32}E^{*})}{T_1E_n}+\frac{i(\sigma_{31}^{*}E-\sigma_{31}E^{*})}{2T_1E_n}.$$

We shall also write down the reduced wave equation for the propagation of radiation along the z axis:

$$\frac{\partial E}{\partial z}+\frac{1}{c}\frac{\partial E}{\partial t}=i\frac{4\pi\mu^2N\omega T_2}{c\hbar}E_n(\sigma_{31}+\sigma_{32}). \qquad (2.2)$$

Here, E is the complex amplitude of the field introduced by the relationship

$$\mathscr{E}=[E\exp(-i\omega t+ikz)+\text{c.c.}]/2;$$

the quantity $E_n=\hbar/\mu(T_1T_2)^{1/2}$ is the amplitude of a monochromatic field which saturates an optical transition; σ_{31} and σ_{32} are related by $\rho_{31,32}=(T_2/T_1)^{1/2}\sigma_{31,32}\exp(-i\omega t+ikz)$ and represent the normalized amplitudes of off-diagonal elements of the density matrix of optical transitions; τ_2 is the relaxation time for low-frequency coherence; N is the density of the molecules in the medium. For simplicity, it is assumed that the spectroscopic characteristics of both optical transitions (dipole moment μ, longitudinal relaxation time T_1, transverse relaxation time T_2, and equilibrium population) are identical.

We shall assume that the widths of the optical lines are much greater than the spectral width of a pulse and also much greater than the splitting frequency of the levels, the detuning of the carrier frequency of a pulse from the optical transition frequencies, and the characteristic Rabi frequency: $T_2^{-1}\gg\tau_p^{-1},\omega_{21},|\omega_{31,32}-\omega|,\mu|E|/\hbar$. We then can assume approximately that $\partial\sigma_{31}/\partial t=\partial\sigma_{32}/\partial t=0$ and reduce the first two equations of the system (2.1) to the following algebraic relationships

$$\sigma_{31}=-i(n_{31}-\rho_{21})E/2E_n,\quad \sigma_{32}=-i(n_{32}-\rho_{21}^{*})E/2E_n. \qquad (2.3)$$

Substituting them into the remaining equations, we find that Eqs. (2.1) and (2.2) yield the following self-consistent system of nonlinear equations:

$$\frac{\partial I}{\partial z}+\frac{1}{c}\frac{\partial I}{\partial t}=-a(n+\operatorname{Re}\rho_{21})I, \qquad (2.4)$$

$$\frac{\partial\rho_{21}}{\partial t}+i\omega_{21}\rho_{21}=-\frac{\rho_{21}}{\tau_2}-\frac{1}{2T_1}(n+\rho_{21})\frac{I}{I_n}, \qquad (2.5)$$

$$\frac{\partial n}{\partial t}=-\frac{2n-1}{2T_1}-\frac{3}{2T_1}(n+\operatorname{Re}\rho_{21})\frac{I}{I_n}. \qquad (2.6)$$

Here, $I=c|E|^2/8\pi$ is the field intensity; $I_n=c|E_n|^2/8\pi$; $n=(n_{13}+n_{23})/2$ is the half-sum of the differences between the populations at the levels participating in the optical transition; and $a=8\pi\mu^2N\omega T_2/c\hbar$ is the linear absorption coefficient.

The system (2.4)–(2.6) differs from the corresponding rate equations for a two-level medium given in Ref. 18 by the presence of an additional equation (2.5) representing the complex dynamic variable ρ_{21}. This gives rise to a specific "coherent" nonlinearity, different from the nonlinearity of saturation and generally of a resonant nature.

§3. COHERENT TRAPPING OF POPULATIONS BY A TRAIN OF ULTRASHORT PULSES

The problem of describing the behavior of a three-level medium in a given field of a periodic sequence of ultrashort pulses reduces to finding a solution of a system of three real linear equations (2.4)–(2.6) with coefficients which are periodic functions of time. We shall consider a steady-state solution describing periodic oscillations which occur in the medium at the pulse repetition frequency. The task becomes easier because we can distinguish two intervals in each period. During the short action of a pulse [$t\in(0,\tau_p)$], when relaxation and natural oscillations of the medium can be ignored, the variables $\rho_{21}=u+iv$ and n vary only under the action of the field:

$$\begin{vmatrix}u\\v\\n\end{vmatrix}_t=\begin{pmatrix}1-[1-b(t)]/4 & 0 & -[1-b(t)]/4\\ 0 & b^{1/4}(t) & 0\\ -3[1-b(t)]/4 & 0 & 1-3[1-b(t)]/4\end{pmatrix}\begin{vmatrix}u\\v\\n\end{vmatrix}_{t=0}, \qquad (3.1)$$

$$b(t)=\exp\left[-(2/I_nT_1)\int_0^t I(t,z)\,dt\right].$$

In the interval between the pulses [$t\in(\tau_p,T)$] the field is zero. Three damped oscillations of ρ_{21} and relaxation of n to the equilibrium value occur during this interval:

$$\begin{vmatrix}u\\v\\n\end{vmatrix}_t=\begin{pmatrix}e^{-t/\tau_2}\cos\omega_{21}t & e^{-t/\tau_2}\sin\omega_{21}t & 0\\ -e^{-t/\tau_2}\sin\omega_{21}t & e^{-t/\tau_2}\cos\omega_{21}t & 0\\ 0 & 0 & e^{-t/T_1}\end{pmatrix}\begin{vmatrix}u\\v\\n\end{vmatrix}_{t=\tau_p}+\begin{vmatrix}0\\0\\(1-e^{-t/\tau_2})/2\end{vmatrix}. \qquad (3.2)$$

The amplitudes of steady-state oscillations of the parameters of the medium are determined by the fixed points of the superposition of the transformations (3.1) (during the action of a pulse, $t=\tau_p$) and (3.2) (during the interval between the pulses, $t=T$). Hence, we can readily show that the amplitudes at the end of a pulse can be found from the following inhomogeneous linear system of algebraic equations:

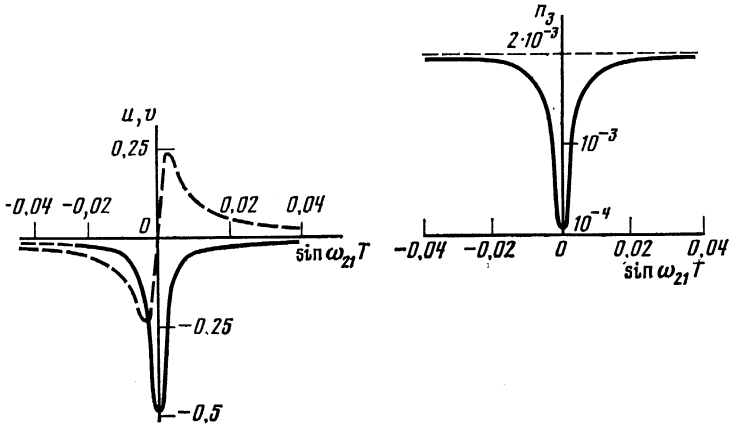

FIG. 2. Coherent population trapping. Resonant dependences of the amplitudes of oscillations [Eqs. (3.4)–(3.7)] of a low-frequency coherence $\rho_{21} = u + iv$ and of the population of the upper level n_3 on the parameter $\sin\omega_{21}T$, representing the detuning of the pulse repetition frequency $\Omega = 2\pi/T$ from the resonance value $\Omega_0 = \omega_{21}/m$ ($m = 1,2,\ldots$; $\sin\omega_{21}T \approx 2\pi(\Omega - \Omega_0)/\omega_{21}$ when $|\Omega - \Omega_0| \ll \omega_{21}$), plotted on the basis of numerical calculations carried out for the parameters $\tau_2/T_1 = 10^5$, $T_1/T = 10^{-1}$, $\bar{I} = 10^{-3}I_s = 10^3 I_c$.

$$u\left[1-\left(1-\frac{1-b_p}{4}\right)\theta_2\cos\omega_{21}T\right]$$
$$+v\theta_2\left(\frac{1-b_p}{4}-1\right)\sin\omega_{21}T+n\theta_1\frac{1-b_p}{4}$$
$$=-\frac{(1-\theta_1)(1-b_p)}{8},$$
$$u\theta_2 b_p^{1/4}\sin\omega_{21}T+v(1-\theta_2 b_p^{1/4}\cos\omega_{21}T)=0, \qquad (3.3)$$
$$^3/_4 u\theta_2(1-b_p)\cos\omega_{21}T+{}^3/_4 v\theta_2(1-b_p)\sin\omega_{21}T$$
$$+n\{1-\theta_1[1-{}^3/_4(1-b_p)]\}={}^1/_2(1-\theta_1)[1-{}^3/_4(1-b_p)].$$

The solution of this system of equations is described by

$$u=-(1-\theta_1)(1-b_p)(1-\theta_2 b_p^{1/4}\cos\omega_{21}T)/8\Delta, \qquad (3.4)$$

$$v=\theta_2(1-\theta_1)(1-b_p)b_p^{1/4}\sin\omega_{21}T/8\Delta, \qquad (3.5)$$

$$n_3=(1-b_p)[1+\theta_2^2 b_p^{1/4}-\theta_2(1+b_p^{1/4})\cos\omega_{21}T]/4\Delta, \qquad (3.6)$$

$$\Delta=1-\theta_1 b_p-\theta_1(1-b_p)/4+\theta_2^2 b_p^{1/4}[1-\theta_1 b_p-(1-b_p)/4]$$
$$-\theta_2[(1-\theta_1 b_p)(1+b_p^{1/4})-(1-b_p)(1+\theta_1 b_p^{1/4})/4]\cos\omega_{21}T. \qquad (3.7)$$

Here, $n_3 = (1-2n)/3$ is the population of the upper level at the end of a pulse:

$$t=\tau_p, \quad b_p=b(t=\tau_p), \quad \theta_1=\exp(-T/T_1), \quad \theta_2=\exp(-T/\tau_2).$$

An analysis of these expressions shows that if $\tau_2 \gg T$, when the low-frequency coherence does not have sufficient time to relax in the interval between two consecutive excitations, the amplitudes of the oscillations of ρ_{21} and n_3 depend resonantly on the pulse repetition frequency (Fig. 2). The resonances occur when the splitting frequency is a multiple of the pulse repetition frequency: $\omega_{21} = 2\pi m/T$, where $m = 1,2,\ldots$ or when $\sin\omega_{21}T = 0$. Since $\mathrm{Re}\,\rho_{21} < 0$, excitation of a low-frequency coherence reduces the term on the right-hand side of Eq. (2.6) which is proportional to $n + \mathrm{Re}\,\rho_{21}$ and, consequently, it reduces n_3. Therefore, we can speak of resonant suppression of the saturation effect.

The relative width of a resonance of half-maximum $\delta = |\Omega - \Omega_0|/\Omega_0$, where $\Omega_0 = 2\pi/T = \omega_{21}/m$ is the resonance value of the pulse repetition frequency, is found from the condition

$$n_3(\sin\omega_{21}T) = [n_3(\sin\omega_{21}T=1)+n_3(\sin\omega_{21}T=0)]/2.$$

In the interesting case of a narrow resonance, $\delta \ll 1$, when $\sin\omega_{21}T \approx 2\pi(\Omega-\Omega_0)/\omega_{21}$ is true within a resonance peak, it follows from Eqs. (3.6) and (3.7) that

$$\delta^2 = 2(1-\theta_2)(1-\theta_2 b_p^{1/4})h/(1+\theta_2^2 b_p^{1/4}). \qquad (3.8)$$

The ratio of the populations $h = n_3(\sin\omega_{21}T = 1)/n_3(\sin\omega_{21}T = 0)$ describing the depth of a resonance is given by the formula

$$h=[1+(1-b_p)\nu]\Big/\left[1+\frac{1-b_p}{4}\left(\frac{4\theta_1-1}{1-\theta_1}+\frac{1}{1+\theta_2^2 b_p^{1/4}}\right)\right], \qquad (3.9)$$

where

$$\nu=(3\theta_1+\theta_2-4\theta_1\theta_2)/4(1-\theta_1)(1-\theta_2). \qquad (3.10)$$

The important parameters of the problem which determine the resonance characteristics are the coherent intensity

$$I_c = 2I_n T_1/\tau_2 \qquad (3.11)$$

and the saturation intensity

$$I_s = I_n T_1(1-\theta_1)/T, \qquad (3.12)$$

where $I_n = c\hbar^2/8\pi\mu^2 T_1 T_2$. These quantities are related respectively to the excitation of a low frequency coherence and equalization of the populations. In the case of a very low intensity,

$$\bar{I} \ll \min\{I_s, I_c\}, \quad \bar{I} = \int_0^{\tau_p} I(t,z)\,dt/T,$$

resonance is shallow $h \approx 1 + \bar{I}/I_c$, and the width of a resonance is determined by the width of a low-frequency transition: $\delta \approx 2\pi m/\omega_{21}\tau_2$. An increase in the intensity increases the depth and width of a resonance.

The most interesting is the case when the lifetime of a low-frequency coherence exceeds the lifetime of the upper level ($\tau_2 \gg T_1$) and the intensity exceeds the characteristic coherent value given by Eq. (3.11). Then, a train of ultrashort pulses induces coherent trapping of populations. In this effect the population of the upper level under resonance conditions is many times smaller than away from resonance in a two-level medium exhibiting an optical transition with the same parameters. Then, the low-frequency coherence is described by $\rho_{21} \approx -1/2$, i.e., its modulus is close to the

maximum possible value. In other words, molecules are captured into a superposition state of the lower sublevels and the upper level is practically empty. It is important to note that this effect may appear even at an intensity much lower than the saturation value, because $I_c \ll I_s$ holds due to the condition $\tau_2 \gg T_1, T$. If $I_c \lesssim \bar{I} \ll I_s$, it then follows from Eqs. (3.8) and (3.9) that the depth and width of a resonance increase as functions of the intensity in accordance with the law

$$h \approx 1+\bar{I}/I_c, \quad \delta \approx 2\pi m(1+\bar{I}/I_c)/\omega_{21}\tau_2.$$

This means that the coherent trapping of populations first becomes more and more prominent under resonance conditions and, second, appears in a wider range of the values of the detuning of the pulse repetition frequency from a resonance.

The effect is observed also at intensities much higher than the saturation value [$\bar{I} \gg I_s$, see Eq. (3.12)]. In spite of the fact that outside a resonance the population of the upper level is close to 1/3, i.e., in spite of the populations of the three levels being practically equal, the saturation effect is suppressed at a resonance. In this case the width and depth of a resonance reach very high values. In the case of a medium with an instantaneous response when $T_1 \ll T$, we have $h \approx \tau_2/4T$ and $\delta \sim 2^{-1/2}$. For a medium with a finite response time, when $T_1 \gg T$, we have

$$h \approx \tau_2/3T_1,$$

$$\delta \approx (2\pi m/\omega_{21}\tau_2)\,[\tau_2(1+\bar{I}/I_c)/3T_1]^{1/2} \text{ for } I_s \ll \bar{I} \ll I_n T_1/T,$$

$$\delta \approx 2(\pi m/3\omega_{21}T_1)^{1/2} \text{ for } \bar{I} \gg I_n T_1/T.$$

We can see that coherent trapping of populations in a medium with an instantaneous response is stronger because the ratio of the time constants τ_2/T_1 is greater.

§4. COHERENT BLEACHING OF A THREE-LEVEL MEDIUM

We shall now consider the self-consistent problem of the propagation of a periodic train of ultrashort pulses in a three-level medium. Substituting the above solution (3.1) into Eq. (2.4) and integrating the latter over the duration of a pulse, we obtain the following equation for the change in the energy of a pulse

$$W_p = \int_0^{\tau_p} I(t, z)\,dt$$

in the course of propagation:

$$dW_p/dz = -(1-\theta_1)\hbar\omega N n_3. \tag{4.1}$$

This equation has a simple physical meaning: the change in the energy of a pulse due to its steady-state passage across a layer of a medium dz thick is equal to that fraction of the energy stored in the medium as a result of filling of the upper level which has been dissipated during the interpulse interval as a result of relaxation of the population of the upper level at a rate T_1^{-1}.

It is clear from Eq. (4.1) that the absorption coefficient and the absorbed power are both proportional to n_3. Therefore, they decrease resonantly under conditions of coherent population trapping.

The substitution of Eq. (3.6) into Eq. (4.1) yields a nonlinear first-order differential equation for the energy of a pulse, which is generally integrable in quadratures. In particular, in the case of a strictly resonant sequence of pulses the integral can be calculated explicitly and the result can be presented in the form of a relationship governing the change in the average intensity of the pulses as they propagate in the medium:

$$e^{-\alpha z} = \left[\frac{1-b_p(z)}{1-b_p(0)}\right]\left[\frac{b_p(0)}{b_p(z)}\right]^{\nu+1}. \tag{4.2}$$

According to Eq. (4.2), the nonlinearity of the absorption law is described by the average intensity

$$I^{NL} = I_n T_1/2T(\nu+1) \sim \min\{I_s,\ I_c\}. \tag{4.3}$$

If the intensity is much less than this value, i.e., if it is much less than the saturation (3.12) and coherent (3.11) values $\bar{I} \ll \min\{I_s, I_c\}$), the nonlinearity is not manifested and an exponential decay occurs in accordance with the law $\bar{I}(z)/\bar{I}(0) = \exp(-\alpha z)$. However, if the intensity at the entry to the medium exceeds I^{NL} $\bar{I} \gg \min\{I_s, I_c\}$), then during the initial stage the intensity decreases proportionally to a coefficient which itself is inversely proportional to a parameter $\nu + 1 = [3/(1-\theta_1) + 1/(1-\theta_2)]/4$ (see Fig. 3):

$$\bar{I}(z)/\bar{I}(0) \approx 1 - \alpha z I_n T_1/2(\nu+1)\bar{I}(0)T. \tag{4.4}$$

Only at high values of z when the intensity decreases to such an extent that the nonlinearity becomes unimportant [$\bar{I}(z) \ll I^{NL}$], does the decay law become exponential, as found in the linear theory of propagation of radiation. The characteristic length in which the intensity decreases by the factor $e \approx 2.7$ is

$$L \sim \frac{1}{\alpha}\left[1+(\nu+1)\frac{2\bar{I}(0)T}{I_n T_1}\right]. \tag{4.5}$$

We can show that in the case of a two-level medium with the same optical transition parameters the law describing the decay of the average intensity of the pulses as they propagate differs from Eq. (4.2) by the replacement of the parameter $\nu + 1$ with $1/(1-\theta_1)$. This is due to the fact that in a two-level medium the coherent nonlinearity is absent and only the absorption nonlinearity may be manifested (when $\bar{I} \gg I_s$). Qualitative differences of the propagation of a resonant train of ultrashort pulses in a three-level medium

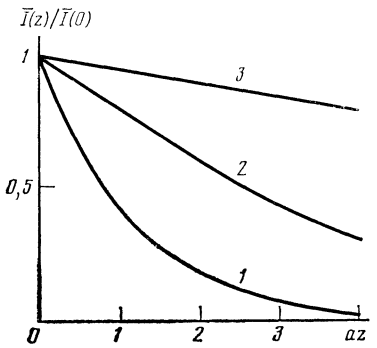

FIG. 3. Coherent bleaching of a three-level medium by a train of ultrashort pulses. The curves represent nonlinear decay of the intensity of the pulses as they propagate in a three-level medium, found by numerical calculation for the following values of the parameters ν and $2\bar{I}(0)T/I_n T_1$: 0.25 and 0.25 for curve 1; 0.25 and 4 for curve 2; 4 and 4 for curve 3.

from the propagation in a two-level medium, and also from the nonresonance case appear in a situation when the coherent nonlinearity is important ($\bar{I} \gg I_c$) and the saturation nonlinearity is weak compared with the former [$I_c \ll I_s$, see Eqs. (3.11) and (3.12)]. In other words, the differences occur in the case of coherent population trapping. Then, under resonance conditions the characteristic length L of penetration of radiation into the medium described by Eq. (4.5) is many times greater than under off-resonance conditions or in a two-level medium: if $I_c \ll \bar{I}(0) \ll I_s$ the difference is a factor of $\bar{I}(0)/I_c$, whereas for $\bar{I}(0) \gg I_s$ the difference is a factor of τ_2/T_1 for a medium with a finite response time and a factor of τ_2/T for a medium with an instantaneous response.

In this way a three-level medium is bleached coherently by a train of ultrashort pulses: a strong resonance train of ultrashort pulses propagates in a three-level medium over large distances practically without absorption.

This coherent bleaching effect and the simulton propagation[16] involve resonant excitation of a low-frequency coherence so that the pulses propagate practically without energy losses. However, it must be stressed that the conditions for coherent bleaching $\omega_{21} \ll \tau_p^{-1} \ll T_2^{-1}$ are opposite to the conditions for the simulton propagation $\omega_{21} \gg \tau_p^{-1} \gg T_2^{-1}$, and the characteristic value of the coherent intensity of Eq. (3.11) in the case when $\tau_2 \gg T$ is much less than the intensity necessary for the simulton propagation which is $\sim I_n T_1/T_2$.

We shall conclude with some comments on the possible applications of coherent population trapping and coherent bleaching of a three-level medium by a train ultrashort pulses.

A series of experimental investigations was recently reported[19,20] in which the resonant response of a three-level medium to a periodic train of ultrashort pulses was used in ultrahigh-resolution spectroscopy. Resonances with widths of the order of 30 Hz were recorded.[20] This method makes it possible to determine small and large frequency splitting, because the condition $\tau_p^{-1} \gg \omega_{21}$ can currently be satisfied right up to splitting of the order of 10^{13} Hz. Moreover, the technique for generation of the required ultrashort pulse trains has recently become reasonably accessible and popular.[21] The promising nature of this method is mentioned in a recent review on laser spectroscopy.[22]

However, in all the experimental investigations (with the evident exception of Ref. 17) the condition of wide optical lines $T_2^{-1} \gg \omega_{21}$ has not been satisfied. In some cases the homogeneous width has been even less than the pulse repetition frequency, i.e., less than the interval between consecutive pulses in a train. Under these conditions each atom interacts only with one pair of spectral components which are in resonance with the optical lines. In such a case the phase relationships between the modes are unimportant, so that there is no need to ensure mode locking of laser radiation. Such spectroscopy involving the use of a train of ultrashort pulses is closely related to the familiar variant of the spectroscopy of quantum beats based on mode crossing resonances.[23]

On the condition $T_2^{-1} \gg \omega_{21}$ ensures that each atom interacts with all the spectral components of laser radiation, i.e., that interacts with a train of pulses as a whole and, consequently, this condition ensures the fullest spectroscopic utilization of a sequence of ultrashort pulses. Resonances which occur in the presence of wide-band optical transitions ($T_2^{-1} \gg \omega_{21}$), when the low-frequency structure is completely submerged under the homogeneous broadening of the optical lines, provides an approach to spectroscopy which is free not only of the Doppler line broadening, but also of the homogeneous (including natural) broadening. The possibility of such "subhomogeneous" spectroscopy depends on the resonant nature of the coherent nonlinearity of a three-level medium which is manifested on interaction with a periodic train of ultrashort pulses.

It should also be mentioned that, in spite of several experimental investigations, the theoretical discussions of the interaction between a train of ultrashort pulses and a three-level medium have been limited so far to the statement that the sum of single, decaying at a rate characterized by the time τ_2 and oscillating at a natural frequency ω_{21}, responses of a low-frequency coherence to the individual pulses in a train should increase resonantly under conditions when the natural frequency is a multiple of the interaction frequency.[17] However, the law describing the change in the intensity of the pulses as they propagate in the medium and an analytic expression for the absorption profile (dependence of the absorption coefficient on the pulse repetition frequency), particularly the dependence of the resonance width and depth on the intensity, have not been hitherto determined. The knowledge of these dependences described by Eqs. (4.2), (3.6), (3.8), and (3.9) makes it possible not only to find the splitting frequency ω_{21}, but also the low-frequency coherence relaxation time τ_2 and, if the parameters of the three-level medium are known and the splitting frequency can be varied (for example, by the application of a magnetic field in the case of the Zeeman sublevels), these equations can be used to find the parameters of an ultrashort pulse train.

A three-level medium can also be applied as a nonlinear filter for the passive mode locking of laser radiation. This possibility has been mentioned earlier[24] using the mode approach. The above space-time description makes clear the physical mechanism of passive mode locking by a three-level filter. In contrast to the usual saturable absorbers which operate on the basis of a nonlinear reduction in the absorption because of the saturation effect,[18] a three-level filter operates by means of coherent bleaching. Since this effect is of resonant nature, mode locking should break down when the condition that the splitting frequency is a multiple of the intermode interval is no longer obeyed. This circumstance can be used in intracavity laser spectroscopy.

[1]A. Javan, Phys. Rev. **107**, 1579 (1957).
[2]A. M. Clogston, J. Phys. Chem. Solids **4**, 271 (1958).
[3]V. M. Kontorovich and A. M. Prokhorov, Zh. Eksp. Teor. Fiz. **33**, 1428 (1957) [Sov. Phys. JETP **6**, 1100 (1958)].
[4]V. M. Faĭn and Ya. I. Khanin, Kvantovaya radiofizika, Sovetskoe Radio, Moscow, 1965 (Quantum Electronics, 2 vols., MIT Press, Cam-

bridge, Mass, 1968; Pergamon Press, Oxford, 1969).
[5]G. Alzetta, L. Moi, and G. Orriols, Nuovo Cimento B **52**, 209 (1979).
[6]G. Orriols, Nuovo Cimento B **53**, 1 (1979).
[7]P. M. Radmore and P. L. Knight, J. Phys. B **15**, 561 (1982).
[8]M. Kaivola, P. Thorsen, and O. Poulsen, Phys. Rev. A **32**, 207 (1985).
[9]P. R. Hemmer, S. Ezekiel, and C. C. Leiby, Jr., Opt. Lett. **8**, 440 (1983).
[10]B. J. Dalton, R. McDuff, and P. L. Knight, Opt. Acta **32**, 61 (1985).
[11]P. M. Radmore and P. L. Knight, Phys. Lett. A **102**, 180 (1984).
[12]Z. Deng, Phys. Lett. A **105**, 43 (1984).
[13]G. P. Agrawal, Phys. Rev. A **24**, 1399 (1981).
[14]J. Mlynek, F. Mitschke, R. Deserno, and W. Lange, Phys. Rev. A **29**, 1297 (1984).
[15]V. G. Minogin and Yu. V. Rozhdestvenskiĭ, Zh. Eksp. Teor. Fiz. **88**, 1950 (1985) [Sov. Phys. JETP **61**, 1156 (1985)].
[16]L. A. Bol'shov and V. V. Likhanskiĭ, Kvantovaya Elektron. (Moscow) **12**, 1339 (1985) [Sov. J. Quantum Electron. **15**, 889 (1985)].
[17]J. Mlynek, W. Lange, H. Harde, and H. Burggraf, Phys. Rev. A **24**, 1099 (1981).
[18]Ya. I. Khanin, Dinamika kvantovykh generatorov (Dynamics of Lasers), Sovetskoe Radio, M., 1975.
[19]H. Harde and H. Burggraf, Opt. Commun. **40**, 441 (1982).
[20]H. Harde and H. Burggraf, Laser Spectroscopy VI (Proc. Sixth Intern. Conf., Interlaken, Switzerland, 1983, ed. by H. P. Weber and W. Lüthy), Springer Verlag, Berlin, 1983, p. 117.
[21]E. M. Dianov, A. Ya. Karasik, P. V. Mamyshev, A. M. Prokhorov, and V. N. Serkin, Zh. Eksp. Teor. Fiz. **89**, 781 (1985) [Sov. Phys. JETP **62**, 448 (1985)].
[22]R. C. Thompson, Rep. Prog. Phys. **48**, 531 (1985).
[23]E. B. Aleksandrov, Usp. Fiz. Nauk **107**, 595 (1972) [Sov. Phys. Usp. **15**, 436 (1973)].
[24]O. A. Kocharovskaya, Ya. I. Khanin, and V. B. Tsaregradskiĭ, Kvantovaya Elektron. (Moscow) **12**, 1227 (1985) [Sov. J. Quantum Electron. **15**, 810 (1985)].

Translated by A. Tybulewicz