OSCILLATIONS IN A PULSE OF RESONANCE FLUORESCENCE INITIATED BY A SMOOTH PULSE OF NONSATURATING RADIATION

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A theoretical study of oscillations occurring in a pulse of a spontaneous emission initiated in a regime of linear absorption of a smooth bell– shaped radiation pulse exciting a resonance atomic transition is presented. The regions of the oscillation existence are found numerically for different durations of the excitation pulse and for different detunings of its radiation frequency from resonance with the atomic transition. An essential dependence of the size and shape of the regions on the rate of an induced dipole moment of an atom and on the ratio of durations of leading and trailing edges of the excitation pulse is revealed.

Necessity of investigating quantum systems dynamics under the action of pulsed laser radiation is caused by a wide range of its applications. A capability of creating essential population of atoms and molecules at certain excited levels at a required time makes a physical basis for such applications as laser photo– and thermochemistry,[1,2] selective photodissociation of molecules and photoionization of atoms,[1–4] and many others. This is achieved by using short and high–power resonance laser emission pulses. Under certain conditions the use of such pulses can also cause essential coherence effects connected with excitation of polarization of resonance transitions during times that are shorter than the corresponding relaxation times.

In addition to the influence on the population dynamics observed, for example, at interference of different channels of transitions to a certain level[6] the coherence phenomena themselves are of great importance. As an example it will suffice to mention the photon echo,[2,7,8] self–induced transparency,[2] and Rabi oscillations.[7,9,10] Not aiming at reviewing a wide number of papers concerning these phenomena, we should like to note that the above–mentioned coherent and populational phenomena are usually observed at light intensity close to or exceeding the saturation level.

Among the effects of such a type the effects of intraatomic coherence at intensities significantly less than the saturation level are much less studied. Oscillations of a pulsed spontaneous emission (or, in other words, of population of the upper level combining with the field) occurring under excitation of a resonance transition by a smooth pulse with the optical frequency slightly shifted off the resonance frequency of the transition are one of such effects.

In the simplest case these oscillations can be observed in two–level atoms. Existence of such oscillations different from the Rabi ones was mentioned in Ref. 10, in which the solution of the problem of a two–level system under the action of exponentially increasing resonance radiation with the frequency shifted off the transition frequency was discussed. An exact solution of the problem for exponentially decreasing field[11] also contains these oscillations along with the population beats caused by the field intensity, however, they were not even mentioned in Ref. 11.

In this paper we present an investigation of these oscillations occurring in a pulse–response of spontaneous emission of a conservative two–level system under linear absorption of a radiation pulse with a shape close to that of real laser pulses as functions of the exciting pulse duration, its frequency shift off the transition one, and the ratio of the system relaxation rates to the slopes of the leading and trailing edges of a pulse.

The analysis is made using standard equations for density matrix $\hat{r}$ of a conservative two–level system in the model of relaxation constants and rotary wave approximation (RWA) as follows:

$$\begin{cases} \dot{R}' + \gamma R' + \Omega R'' = 0, \\ \dot{R}'' + \gamma R'' - \Omega R' + 2V(t)\,\rho_1 = V(t), \\ \dot{\rho}_1 + \gamma_1 \rho_1 - 2V(t)\,R'' = 0, \end{cases} \qquad (1)$$

$$R' + iR'' = \rho_{10}, \quad \rho_0 + \rho_1 = 1, \quad \Omega = \omega - \omega_{10},$$

$$V(t) = d_{10}\,\varepsilon(t)/2\hbar\,.$$

Here, ρ_0 and ρ_1 are the populations of the lower (0) and upper (1) levels, ρ_{10} is the off– diagonal matrix element of the density describing the field– induced polarization of the system, $\varepsilon(t)$ is the time–dependent envelope of electric field of the optical wave, Ω is the difference between the carrier frequency of light ω from the transition frequency ω_{10}, d_{10} is the matrix element of the transition $0 \to 1$ dipole moment, and γ_1 and γ are the relaxation constants of the upper level and the polarization, respectively.

It is convenient to discuss a physical origin of the considered oscillations using a rectangular pulse $V(t) = \theta(t)\,\theta(\tau - t)V$, where $\theta(t)$ is the step–wise function. In this case the characteristic equation for system (1) for $0 < t < \tau$ takes the form

$$(\lambda + \gamma_1)(\lambda + \gamma)^2 + 4V^2(\lambda + \gamma) + \Omega^2(\lambda + \gamma_1) = 0\,. \qquad (2)$$

The cubic equation (2) relative to index λ is reduced to the quadratic one when $\Omega = 0$ (exact resonance), $V = 0$ (zero field), and $\gamma = \gamma_1$ (equal relaxation constants). When solving Eqs. (1) and other similar systems of equations with a time– dependent interaction term $V(t)$ the exact resonance is considered most frequently what allows one to proceed far

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towards analytical solution of the problem.[10] In this case the sought– for response undergoes Rabi oscillations with the frequency determined by the amplitude V while the oscillations we are interested in are absent at zero field. In the case of equal relaxation constants the solution of Eq. (1) involves temporal exponential functions with the exponents

$$\lambda_1 = -\gamma_1 , \quad \lambda_{2,3} = -\gamma_1 \pm i\,\Omega_R , \quad \Omega_R = \sqrt{\Omega_2 + 4V^2} , \quad (3)$$

where the absolute value of the imaginary parts of the second and third roots Ω_R is the classical Rabi frequency depending on the frequency detuning. It is obvious that in this case Ω and V enter into Ω_R symmetrically and, hence, equally determine the solution oscillations.

Thus, the mechanism forming the oscillations caused by both tuning the exciting field off the resonance and high– power radiation is one and the same and it is formally explained by precession[7] of an atom pseudo– spin ($R'/2$, $R''/2$, $\rho_0 - \rho_1$) around the vector of "rotational momentum" ($-2V$, 0, Ω) with the frequency Ω_R. At the same time, one can easily see from Eqs. (1) the difference in physical nature of oscillations of these two types. Really, oscillations at $\Omega = 0$ appear due to periodic migration of the population from level 0 to 1 with the period comparable with the relaxation times under saturation of the transition. The beats at $V = 0$ is caused by a relation (interference) between absorption and dispersion components of induced dipole momentum, i.e., only by intraatomic coherence with the beats period defined by the difference between the oscillation frequency of the system ω_{10} and the frequency of ω of an inducing force.

As follows from Eq. (3) the condition for observing the oscillations occurring in the regime of linear absorption, i.e., in the case of zero fields, is

$$4V^2 \ll \Omega^2 . \quad (4)$$

A solution of Eqs. (1) as $V \to 0$ for an arbitrary function $V(t)$ can be given in the form

$$\rho_1(t) = e^{-\gamma_1 t} \int_0^t dt_1 e^{(\gamma_1-\gamma)} G(t_1) \int_0^{t_1} dt_2\, e^{\gamma t_2} G(t_2) \cos \Omega(t_1 - t_2), \quad (5)$$

$$G(t) = V(t)\sqrt{2}.$$

Further analysis will be carried out using a popular pulse shape $G(t)$

$$G(t) \sim e^{-\alpha t} - e^{-\beta t} , \ \beta > \alpha > 0 . \quad (6)$$

The maximum of $G(t)$ in relation (6) is at $t = t_{max}$,

$$t_{max} = (\ln \beta - \ln \alpha)/(\beta - \alpha) ,$$

$$G(t_{max}) = r/(1 + r)^{1+1/r}, \ r = \beta/\alpha - 1 , \quad (7)$$

and FWHM is approximated with the error 0.6% by the expression

$$\tau_1 = \frac{1}{\alpha} \ln [2 + 7\,\alpha/\beta + 2.5\,(\alpha/\beta)^3] . \quad (8)$$

As $r \to \infty$ in relation (6) $G(t)$ is reduced to exponentially decreasing pulse $\exp(-\alpha t)$ considered in Ref. 11. As $\beta \to \alpha\,(r \to 0)$ two– exponential pulse (6) is reduced to an exponential by power law $t\exp(-\alpha t)$ with smoother leading edge of a pulse. Experimentally recorded envelope of the intensity $\sim G^2(t)$ has the same value of t_{max} and FWHM τ_2 approximated with the error of 1.6% by the formula

$$\tau_2 = \frac{1}{\alpha} \ln [\sqrt{2} + 4.049\,\alpha/\beta] . \quad (8')$$

An analytical solution $\rho_1(t)$ for pulse (6) has the form

$$\rho_1(t) = (\gamma - \alpha)\, L_1(\alpha) \{[1/\gamma_1 - \alpha - \beta) - 1/(\gamma_1 - 2\alpha) + \delta L_2(\alpha) -$$

$$- (\delta + \alpha - \beta) L_2(\beta)]\, e^{-\gamma_1 t} + e^{-2\alpha t}/(\gamma_1 - 2\alpha) -$$

$$- e^{-(\alpha+\beta)t}/(\gamma_1 - \alpha - \beta)\} - L_2(\alpha) \{[(\gamma - \alpha)\, \delta L_1(\alpha) -$$

$$- (\gamma - \beta)(\delta - \alpha + \beta)] \cos \Omega t - \Omega[\delta L_1(\alpha) - (\delta - \alpha + \beta)\, L_1(\beta)] \times$$

$$\times \sin \Omega t\} e^{-(\gamma+\alpha)t} + (\alpha \leftrightarrow \beta) ; \quad (9)$$

$$L_1(\alpha) = 1/[(\gamma - \alpha)^2 + \Omega^2] , \ L_2(\alpha) = 1/[(\gamma_1 - \gamma - \alpha)^2 + \Omega^2] ,$$

$$\delta = \gamma_1 - 2\gamma .$$

As can be seen from Eq. (9) the pulse– response of induced fluorescence $\sim \rho_1(t)$ has a smooth (the first braces) and the oscillatory parts. The latter decays with rate determined by the polarization relaxation constant γ and excitation pulse duration. Most obviously these components of the response can be separated out as $r \to \infty$, i.e., for an exponentially decreasing excitation pulse. In this case, in the absence of collisions ($\gamma = \gamma_1/2$)

$$\rho_1(t) = \frac{1}{2} L_1(\alpha) [(e^{-\alpha t} - e^{-\gamma_1 t/2})^2 + 4e^{-(\gamma_1+2a)t/2} \sin^2(\Omega t/2)], \quad (10)$$

and the smooth part of a fluorescence pulse is proportional to $G^2(t)$ in relation (6), where $\beta = \gamma_1/2$. As $\alpha \to \gamma_1/2$ the amplitude of the smooth part decreases, approximately as $\sim (\alpha - 2\gamma_1)^2$ down to zero, what results in a maximum contrast of oscillations.

The examples of oscillations in spontaneous emission pulse calculated according to formula (9) for some values of the parameters τ_2, γ, γ_1, r are shown in Fig. 1. As can be seen from this figure the oscillation contrast defined as the ratio of the difference between half– sum of values of $\rho_1(t)$ at two adjacent maxima and the value of ρ_1 at the minimum between them to the absolute maximum of population can be sufficiently large in some cases. This means that the oscillations can be isolated against a sufficiently high background of the experimental noise.

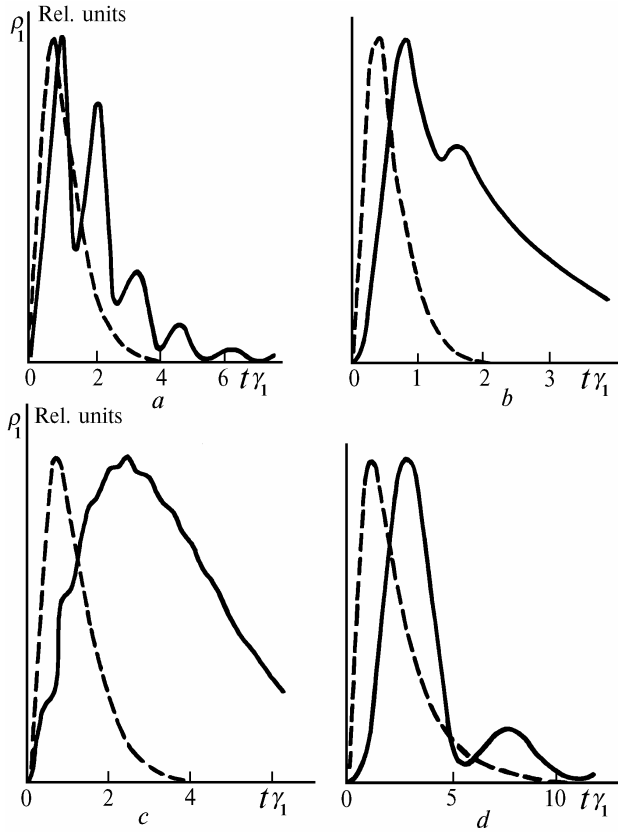

FIG. 1. The shape of a pulse– response of resonance fluorescence having oscillations in a linear regime of absorption caused by the carrier frequency shift of the exciting pulse off the transition frequency (solid lines):

a) $\tau_2\gamma_1 = 1.2$, $\Omega/\gamma_1 = 10$, $\gamma/\gamma_1 = 0.5$, *and* $r = 0.1$;

b) $\tau_2\gamma_1 = 0.6$, $\Omega/\gamma_1 = 12$, $\gamma/\gamma_1 = 2$, *and* $r = 0.1$;

c) $\tau_2\gamma_1 = 1.3$, $\Omega/\gamma_1 = 24$, $\gamma/\gamma_1 = 2$, *and* $r = 0.1$; *and*,

d) $\tau_2\gamma_1 = 2.2$, $\Omega/\gamma_1 = 2.5$, $\gamma/\gamma_1 = 0.5$, *and* $r = 9$.

The exciting pulse $G^2(t)$ *in Eq. (6) is shown by dashed lines.*

From Eqs. (9) and (10), as well as from the general considerations, it follows that the condition necessary for observing oscillations is long enough pulse duration τ_2 and a large time of the polarization decay γ^{-1} compared to the period of oscillations $\tau_{osc} = 2\pi/\Omega$

$$\tau_{osc} \lesssim \min(\tau_2, \gamma^{-1}) . \qquad (11)$$

Relation (11) obviously limits the region of existence of oscillations in a pulse– response from the short durations of pulses. From the side of large τ_2 values there also exists a limitation on the oscillations, since in this case the oscillations with maximum amplitude are observed on a limited interval $0 < t < 1/\gamma$ and, as a result, they are suppressed due to the steepness of a pulse front and because of smallness of the response amplitude $\sim t^2$ at $t \ll \tau_2$ (see Fig. 1*c*). Thus, the domain of values τ_2, in which the oscillations are pronounced, is bounded from the side of large and small times and is determined by the parameters γ, γ_1, and r.

More complete information about the existence of the domain of essential oscillations depending on the pulse duration τ_2 and the frequency shift Ω gives Fig. 2, which shows calculated isolines of equal numbers of oscillations in a response with the contrast exceeding 5%. The number of oscillations equal to the number of local minima of $\rho_1(t)$ between the maxima which satisfy the condition of certain contrast is shown by the figures near lines.

The width of a domain of the oscillation existence in τ_2 units is proportional to the polarization relaxation time $1/\gamma$ (compare Figs. 2*a* and 2*b*). In both cases at a fixed γ the width increases at a steeper leading and smoother and longer trailing edges of a pulse. A periodical saw– tooth structure of the isolines at large τ_2 is explained by a successive crawl of the oscillations over a maximum of the smooth part of the pulse and by suppression of them at the leading edge of the pulse. Smooth and regular isolines at small τ_2 correspond to oscillations at the trailing edge of the pulse when the first oscillation maximum occurs at t larger than that of the maximum of the smooth part of the response pulse. Crooks of the curves in Fig. 2*a* in the region of mean values of τ_2 are caused by the dependence of the oscillation phase on Ω and the pulse– response shape near its maximum.

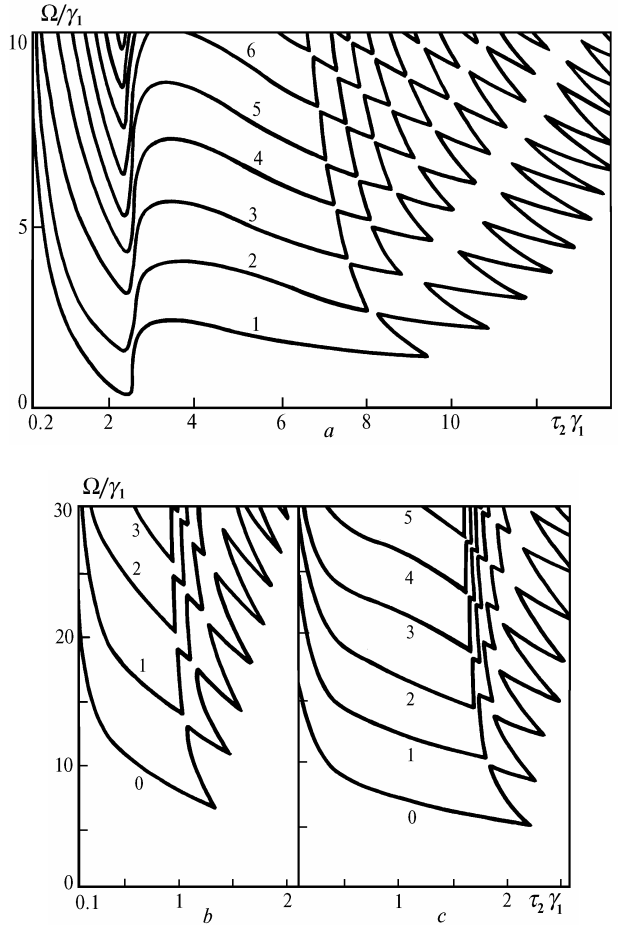

FIG. 2. The domains of existence of oscillations in dependence on the exciting pulse duration τ_2 *and on the frequency detuning* Ω *for some values of* γ *and* r: *a)* $\gamma/\gamma_1 = 0.5$, $r = 0.1$; *b)* $\gamma/\gamma_1 = 2$, $r = 0.1$; *and*, *c)* $\gamma/\gamma_1 = 2$ *and* $r = 9$.

Calculations for oscillations with higher contrast made in similar way as the calculations of isolines shown in

Fig. 2, have demonstrated that, on the whole, the view and behavior of isolines remain the same and the width of the existence domain (in τ_2 units) decreases only slightly with increasing contrast. Rareness of isolines along the Ω axis is more noticeable, especially, at small τ_2. Thus, for example, a change of the contrast from 5 to 50% for the case shown in Fig. 2*a* causes a decrease of a number of oscillations taken into account from 9 to 6 at $\Omega = 10\gamma_1$ and $\tau_2 \sim 2/\gamma_1$. At $\tau_2 > 3/\gamma_1$ the changes are much less.

In conclusion it should be noted that essential dependence of specific configuration of the domain of the oscillation existence on the ratio of γ/γ_1, shape, and the ratio of the durations of the exciting pulse fronts makes a good basis for a very intelligent experimental investigation of these characteristics, especially, at low gas pressures when the values of γ are smallest.

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