Atomic response to optical fluctuating fields: Temporal resolution on a scale less than pulse correlation time

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Excitation of a three-level atom irradiated by a sequence of two fluctuating pulses is discussed. The pulses have bandwidth τ_c^{-1}, relative delay time t_{12}, and can be correlated with one another. It is shown that, for strong excitation pulses, the level populations and atomic coherence as functions of t_{12} can vary on a time scale much smaller than τ_c. This effect may lead to a means for obtaining high temporal and spatial resolution.

I. INTRODUCTION

It is well known in relaxation studies that optical coherent transients induced by time-delayed, correlated fluctuating pulses may result in high time resolution.[1-11] For a gas of two-level atoms, it has been shown that this time resolution is given by the cross-correlation time τ_c of the applied fields rather than by the pulse duration t_p, as in the case of Fourier-transform-limited pulses. In experiments, the condition $\tau_c \ll t_p$ is usually satisfied, and subpicosecond relaxation times can be measured with nanosecond or longer pulses. Although most experiments are performed under weak-field conditions, observations are also carried out when at least one of two excitation pulses is strong,[12-15] that is, when

$$\max(\alpha_1, \alpha_2)t_p \gg 1, \tag{1.1}$$

where $\alpha_{1,2} = \langle |f_{1,2}|^2\rangle \tau_c$. The Rabi frequencies, f_1 and f_2, associated with the first and the second excitation pulses, respectively, are averaged over all possible realizations of the fluctuating fields. It has been demonstrated experimentally[13,15] and shown theoretically[16-18] that optical transient signals, as a function of relative delay time t_{12}, may vary on a time scale of order of τ_c. Moreover, it has been found[17] that, for two correlated saturating pulses, this time scale can be even smaller than τ_c. If the inequality

$$\min(\alpha_1, \alpha_2)t_p \gg 1 \tag{1.2}$$

is satisfied, the strongest signals exhibit a peak of width of order τ_c, and this peak can have a very narrow dip near its maximum of order

$$\delta t_{12} \sim \frac{1}{\eta}\tau_c \ll \tau_c, \tag{1.3}$$

where

$$\eta = \left(\frac{\alpha_1\alpha_2 t_p}{\alpha_1 + \alpha_2}\right)^{1/2}, \tag{1.4}$$

and, according to Eq.(1.2), $\eta \gg 1$. This result implies that a time resolution much better than τ_c may be achieved under appropriate conditions. Unfortunately, for two-level atoms the narrow dip is also shallow, having a relative depth of order of $\eta^{-1} \ll 1$, and this would make its experimental observation rather difficult.

The results discussed above are obtained for "two-level" atoms. It is well known, however, that many interesting phenomena that cannot exist for two-level atoms may occur in quantum systems having a number of levels $n \geq 3$. Among such effects are population trapping,[19-29] pressure-induced resonances,[30] atomic cooling below the Doppler limit,[31,32] lasers without inversion.[33]

In this paper I consider an ensemble of three-level atoms of Λ or V configuration (see Fig. 1). An atom interacts with two laser pulses having wave vectors $\mathbf{k}_1$ and $\mathbf{k}_2$, respectively. The pulses are of duration t_p and are time-delayed relative to each other by t_{12} ($t_{12} \ll t_p$). These pulses may be derived from a single laser and, thus, can be correlated. These classical incident fields have amplitudes $\mathcal{E}_1(t)$ and $\mathcal{E}_2(t - t_{12})$ and central frequencies ω_1 and ω_2, respectively. The first field drives the $|0\rangle \Rightarrow |1\rangle$ transition having frequency ω_{10}, while the second field drives the $|0\rangle \Rightarrow |2\rangle$ transition having frequency ω_{20}. It is assumed that $|\omega_{m0} - \omega_m| \ll \omega_m$, where $m = 1, 2$, is satisfied. The model can describe transitions between levels having quantum numbers J=0 and 1 which are linked by laser fields with orthogonal polarization.

The effect under consideration is related to the well-known phenomenon of population trapping. As population trapping, it is directly linked to the existence of a coherent superposition of stationary states in three-level systems of Λ and V types which, under some specific conditions, is decoupled from the excitation fields. However, in contrast to population trapping, spontaneous decay does not play an important role in the effect discussed in this paper.

Let us assume that $\omega_{10} = \omega_{20}$, and that the excitation pulses are fully correlated, have equal central frequencies, and propagate in the same direction; that is $\mathcal{E}_1(t)/\mathcal{E}_2(t)$=const, $\omega_1 = \omega_2$, and $\mathbf{k}_1 = \mathbf{k}_2$. Following a method which was exploited in studies of population trapping in Λ system,[24] one can introduce two superpositions of levels $|1\rangle$ and $|2\rangle$ given by

$$|b\rangle = \cos\beta_0|1\rangle + \sin\beta_0|2\rangle, \tag{1.5}$$

$$|c\rangle = \sin\beta_0|1\rangle - \cos\beta_0|2\rangle, \tag{1.6}$$

where

$$\tan\beta_0 = \frac{|\mu_{02}\mathcal{E}_2(t)|}{|\mu_{01}\mathcal{E}_1(t)|} = \text{const}, \tag{1.7}$$

and μ_{0m} $(m = 1, 2)$ is the dipole moment matrix element associated with a $|0\rangle \Rightarrow |m\rangle$ transition. In the absence of spontaneous decay and for zero delay time, $t_{12} = 0$, the superposition state $|c\rangle$ (a one-level subsystem) is completely decoupled from the two-level subsystem consisting of the states $|0\rangle$ and $|b\rangle$. Consequently, the population in each of the subsystems is conserved during the excitation pulses. For instance, if level $|0\rangle$ is the only one initially populated, and the first pulse is much stronger than the second one $[f_1(t) \gg f_2(t);\ \beta_0 \ll 1]$, according to Eq. (1.6) the population of level $|2\rangle$ is very small for *any* duration, intensity, and time variation of the excitation fields.

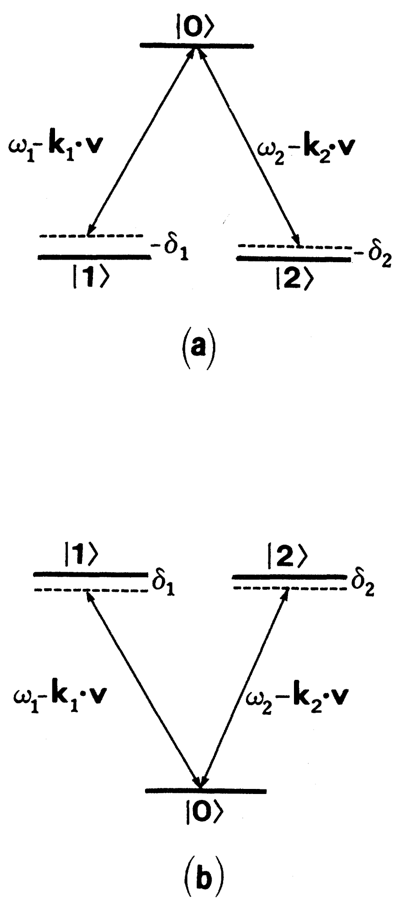

FIG. 1. Three-level configurations considered in this paper, (a) Λ system, (b) V system.

However, for nonzero delay time, $t_{12} \neq 0$, if $\mathcal{E}_{1,2}(t) \neq$const, the two subsystems are no longer decoupled from each other. Moreover, if the excitation fields are stochastic, the evolution of the three-level system changes dramatically compared to the case of zero delay time. For time $t \sim +\infty$ all level populations become equal, that is, tend to $\frac{1}{3}$, and this equilibrium distribution does not depend on initial conditions (see more detailed discussion in Sec. V). Thus, in the example considered above, the population of level $|2\rangle$ as a function of delay time may undergo significant variation from a value close to zero for $t_{12} = 0$ to a value close to $\frac{1}{3}$ which can be reached for $t_{12} \neq 0$. It is shown below that the time scale of this variation decreases with increasing pulse energy. For two strong pulses satisfying Eq. (1.2) the time scale is given by $\delta t_{12} \approx \tau_c/\eta \ll \tau_c$ and coincides with that for a two-level atom given by Eq. (1.3). However, in contrast to the two-level case, for three-level atoms this variation of the populations is a dominant effect. The significant variation of the level populations on a time scale $\delta t_{12} \approx \tau_c/\eta \ll \tau_c$ is the main result of this paper.

In Sec. II I derive equations for averaged level populations and atomic coherence of a three-level atom. Weak-field and strong-field regimes are discussed in Secs. III and IV, respectively. In Sec. V, a physical interpretation of the phenomenon is given. The implication of the results for obtaining high spatial resolution is also discussed.

II. AVERAGED EQUATIONS FOR A THREE-LEVEL ATOM

From this point, the general case of nonequal level detunings from resonance, arbitrary degree of mutual correlation of the fields, and arbitrary propagation directions of the laser beams is considered. For $\mathbf{k}_1 \neq \mathbf{k}_2$, the effective delay time of the pulses depends on the position $\mathbf{r}$ of a particular atom relative to the center of an active region in the atomic sample. For an atom characterized by a velocity $\mathbf{v}$ and a position $\mathbf{r}$ this delay time, $t_{12}(\mathbf{r})$, is given by

$$t_{12}(\mathbf{r}) = t_{12} + \frac{(\mathbf{n}_2 - \mathbf{n}_1)\cdot\mathbf{r}}{c}, \tag{2.1}$$

where $\mathbf{n}_{1,2} = \mathbf{k}_{1,2}/|\mathbf{k}_{1,2}|$, and c is the speed of light. In this paper we assume the effective delay time (2.1) to be constant for a particular atom during the excitation process. Since the atom moves during the excitation, and $\mathbf{r}(t) = \mathbf{r}(0) + \mathbf{v}t$, this assumption imposes a restriction (to be discussed in Sec. V) on the atomic velocity in the direction $\mathbf{n}_2 - \mathbf{n}_1$ across the laser beams. Further, if it is not otherwise stated, $t_{12}(\mathbf{r})$ is referred to simply as t_{12}.

The role of spontaneous decay will be considered elsewhere. In this paper, we assume that the atomic relaxation produced by sources other than the incident fields are negligible on the time scale of an experiment. In this limit one can use population amplitudes rather than a density matrix to describe the atom-field interaction. In the case of the Λ configuration [Fig. 1(a)], we represent the amplitudes in the form

$$a_0(t), \quad a_{1,2}(t)e^{i(\omega_{1,2}t+\mathbf{k}_{1,2}\cdot\mathbf{r})}. \tag{2.2}$$

Then, in the rotating-wave approximation, one obtains the following set of equations:

$$\begin{aligned} i\frac{\partial a_0}{\partial t} &= \tfrac{1}{2}f_1(t)a_1 + \tfrac{1}{2}f_2(t-t_{12})a_2, \\ i\frac{\partial a_1}{\partial t} &= \delta_1 a_1 + \tfrac{1}{2}f_1^*(t)a_0, \\ i\frac{\partial a_2}{\partial t} &= \delta_2 a_2 + \tfrac{1}{2}f_2^*(t-t_{12})a_0, \end{aligned} \tag{2.3}$$

with $f_m(t) = \mu_{0m}\mathcal{E}_m(t)\hbar^{-1}$ $(m = 1,2)$ being the Rabi frequency associated with a $|0\rangle \Rightarrow |m\rangle$ transition, and

$$\delta_m = \omega_m - \omega_{m0} - \mathbf{k}_m \cdot \mathbf{v}. \tag{2.4}$$

In the case of the V configuration [Fig. 1(b)] the coefficients in Eqs. (2.3) are obtained by substitution

$$\delta_{1,2} \Rightarrow -\delta_{1,2}, \quad f_{1,2} \Rightarrow f_{1,2}^*. \tag{2.5}$$

The Λ scheme is discussed below, and the V scheme is considered in the Appendix.

The Rabi frequencies f_1 and f_2 are treated as fully correlated complex stationary stochastic processes with zero mean values and correlation functions defined by

$$\begin{aligned} \langle f_m^*(t)f_m(t-\tau)\rangle &= \alpha_m g_{mm}(\tau), \quad m = 1,2, \\ \langle f_m(t)f_n(t-\tau)\rangle &= 0, \quad m,n = 1,2, \end{aligned} \tag{2.6}$$

and

$$\langle f_1^*(t)f_2(t-\tau)\rangle = (\Phi\alpha_1\alpha_2)^{1/2}g_{12}(\tau), \tag{2.7}$$

where $g_{mn}(\tau)$ is the normalized correlation function, i.e.,

$$g_{nm}^{-1}(0) = \tau_c^{nm}, \quad \int_0^\infty g_{nm}(\tau)d\tau = 1, \tag{2.8}$$

and the parameter Φ is a measure of the relative coherence of the pulses, which satisfies

$$0 \le \Phi \le 1. \tag{2.9}$$

For fully correlated pulses $\Phi = 1$, while for noncorrelated pulses $\Phi = 0$.

In this paper, I consider the averaged atomic density matrix

$$\rho_{mn}(t) = \langle a_m^*(t)a_n(t)\rangle \tag{2.10}$$

assuming that the correlation and delay times are sufficiently small to satisfy

$$\tau_c^{mn}, t_{12} \ll t_p, \alpha_{1,2}^{-1}, \Delta_D^{-1}, |\omega_{1,2} - \omega_{01,2}|^{-1}, \tag{2.11}$$

where Δ_D is a Doppler width of the atomic ensemble, and according to Eqs. (2.6) and (2.7)

$$\alpha_m = \langle |f_{1,2}|^2\rangle\tau_c^{mm}. \tag{2.12}$$

No restriction is imposed on a ratio τ_c^{mn}/t_{12}.

Using Eqs. (2.3) one can easily derive equations for the density matrix elements $\rho_{mn}(t)$. Owing to condition (2.11), one can apply a decorrelation approximation[34] to these equations to arrive at differential equations for the averaged level populations and the atomic coherence ρ_{21}. For pulses of arbitrary shape, one finds

$$\dot n_{01} = -\alpha_1 n_{01} - \tfrac{1}{2}\alpha_2 n_{02} + \tfrac{1}{4}\sqrt{\Phi\alpha_1\alpha_2}(3-G)(\rho_{21}+\rho_{21}^*), \tag{2.13}$$

$$\dot n_{02} = -\alpha_2 n_{02} - \tfrac{1}{2}\alpha_1 n_{01} + \tfrac{1}{4}\sqrt{\Phi\alpha_1\alpha_2}(3+G)(\rho_{21}+\rho_{21}^*), \tag{2.14}$$

$$\begin{aligned} \dot\rho_{21} = &-[\tfrac{1}{4}(\alpha_1+\alpha_2) + i\Delta]\rho_{21} \\ &+ \tfrac{1}{4}\sqrt{\Phi\alpha_1\alpha_2}[(1+G)n_{01} + (1-G)n_{02}], \end{aligned} \tag{2.15}$$

where

$$n_{0m} = \rho_{00} - \rho_{mm}, \quad m = 1,2, \tag{2.16}$$

$$\rho_{00} + \rho_{11} + \rho_{22} = 1, \tag{2.17}$$

$$\Delta = \delta_2 - \delta_1. \tag{2.18}$$

In Eqs. (2.13)–(2.15), the only dependence on the time delay of the pulses is contained in the parameter

$$G(t_{12}) = \int_0^{t_{12}} g_{12}(\tau)d\tau, \quad G(\pm\infty) = \pm 1. \tag{2.19}$$

The atom is assumed to be in its ground state(s) before the excitation pulses are applied at $t = 0$. For the Λ configuration the initial condition is

$$\begin{aligned} &n_{01}(0) = -\rho_{11}(0), \quad n_{02}(0) = -\rho_{22}(0) = \rho_{11}(0) - 1, \\ &\rho_{21}(0) = 0, \end{aligned} \tag{2.20}$$

where the initial populations $\rho_{11}(0)$ and $\rho_{22}(0)$ are not necessarily equal.

If the excitation pulses are derived from two different lasers, they are mutually noncorrelated, and $\Phi = 0$ in Eqs. (2.13)–(2.15). As a result, all the t_{12}-dependent parameters in Eqs. (2.13)–(2.15) vanish, and the averaged density matrix elements do not depend on delay time. For the atomic coherence, one has $\rho_{21}(t) = 0$ for any type of three-level configuration. The behavior of a three-level system driven by noncorrelated pulses was discussed in detail in Ref. 26. In this paper we are interested mostly in analyzing the dependence of the averaged density matrix elements on delay time. I will show below how this dependence emerges for correlated pulses characterized by

$\Phi \neq 0$. In the next sections the laser pulses are assumed to have rectangular envelopes

$$\alpha_{1,2}(t) = \text{const} \neq 0 \quad \text{for} \quad 0 \leq t \leq t_p. \tag{2.21}$$

Then the coefficients in Eqs. (2.13)–(2.15) do not vary with time, and analytical solution can be obtained in some important limiting cases.

III. WEAK-FIELD REGIME

This regime is defined by

$$\alpha_1, \alpha_2 \ll t_p^{-1}. \tag{3.1}$$

For the Λ system taking into account initial conditions (2.20) and solving Eqs. (2.13)–(2.15) one gets the level populations and coherence in the form

$$\rho_{00}(t_p) = \tfrac{1}{2}[\alpha_1\rho_{11}(0) + \alpha_2\rho_{22}(0)]t_p, \tag{3.2}$$

$$\rho_{mm}(t_p) = (1 - \tfrac{1}{2}\alpha_m t_p)\rho_{mm}(0), \quad m = 1, 2, \tag{3.3}$$

$$\rho_{21}(t_p) = \frac{i\sqrt{\Phi\alpha_1\alpha_2}[\exp(-i\Delta t_p) - 1][1 + (\rho_{11}(0) - \rho_{22}(0))G(t_{12})]}{4\Delta}. \tag{3.4}$$

As one can see from Eqs. (3.2) and (3.3), in a weak-field regime the excited state is populated only slightly, and each of the pulses affects only the corresponding transition. The populations do not depend on the delay time t_{12}. However, if the initial populations of levels 1 and 2 are not equal, one can see from Eq. (3.4) that there is an asymmetric dependence of the atomic coherence ρ_{21} on the delay time. The variation of ρ_{21} occurs on a time scale of order τ_c^{12}, is given by

$$\frac{\rho_{21}(t_p; t_{12} \gg \tau_c^{12}) - \rho_{21}(t_p; t_{12} \ll -\tau_c^{12})}{\rho_{21}(t_p; t_{12} = 0)} = 2[\rho_{11}(0) - \rho_{22}(0)], \tag{3.5}$$

and can be significant for $|\rho_{11}(0) - \rho_{22}(0)| \sim 1$.

IV. STRONG-FIELD REGIME

In this section, I consider a strong-field regime which is defined by

$$(\alpha_1 + \alpha_2)t_p \gg 1. \tag{4.1}$$

To analyze this case, and for the remainder of the paper, it is assumed that

$$\alpha_1 \geq \alpha_2. \tag{4.2}$$

The results for $\alpha_2 \geq \alpha_1$ can be obtained from that for $\alpha_1 \geq \alpha_2$ by substituting

$$\alpha_1 \Leftrightarrow \alpha_2, \ \rho_{11} \Leftrightarrow \rho_{22}, \quad \rho_{21} \Leftrightarrow \rho_{12}, \ \ G(t_{12}) \to -G(t_{12}). \tag{4.3}$$

The solutions for $n_{01}(t)$, $n_{02}(t)$, and $\rho_{21}(t)$ are given by a sum of four exponentials $\exp(-\lambda_m t)$, $m = 1, \ldots, 4$, where λ_m are the roots of the equation

$$\lambda^4 - \tfrac{3}{2}(\alpha_1 + \alpha_2)\lambda^3 + \{\tfrac{9}{16}(\alpha_1 + \alpha_2)^2 + \tfrac{1}{4}\alpha_1\alpha_2[\Phi G^2 + 3(1 - \Phi)] + \Delta^2\}\lambda^2 - \tfrac{1}{16}(\alpha_1 + \alpha_2)[(\alpha_1 + \alpha_2)^2 + \alpha_1\alpha_2\Phi G^2 + 16\Delta^2]\lambda + \tfrac{3}{64}\alpha_1\alpha_2[(\alpha_1 + \alpha_2)^2(\Phi G^2 + 1 - \Phi) + 16\Delta^2] = 0. \tag{4.4}$$

If the condition

$$\sin^2(2\beta)(\Phi G^2 + 1 - \Phi + \Delta_0^2) > 4(1 + \Delta_0^2), \tag{4.5}$$

is satisfied, where

$$\beta = \arctan\sqrt{\alpha_2/\alpha_1}, \quad 0 \leq \beta \leq \pi/4, \qquad \Delta_0 = \frac{4\Delta}{\alpha_1 + \alpha_2}, \tag{4.6}$$

for all four roots λ_m it follows that

$$\lambda_m t_p \gg 1, \quad m = 1, 2, 3, 4. \tag{4.7}$$

Consequently, $n_{02}(t_p) = n_{01}(t_p) = \rho_{21}(t_p) = 0$, and at time t_p all three atomic levels are equally populated, that is

$$\rho_{mm}(t_p) = \tfrac{1}{3}, \quad m = 1, 2, 3. \tag{4.8}$$

However, if

$$\sin^2(2\beta)(\Phi G^2 + 1 - \Phi + \Delta_0^2) \ll 4(1 + \Delta_0^2), \tag{4.9}$$

one of the roots of Eq. (4.4) can satisfy the condition $\lambda t_p < 1$, and some nontrivial final distribution of the population over the levels may become possible. For pulses with very different intensities, that is for $\alpha_1 \gg \alpha_2$ ($\beta \ll 1$), condition (4.9) is satisfied for any mutual correlation of the pulses Φ ($0 \leq \Phi \leq 1$), and any t_{12} and

Δ, provided the inequality (2.11) is also satisfied. For pulses with nearly equal intensities [$\alpha_1 \sim \alpha_2$ ($\beta \sim 1$)], restrictions

$$(1-\Phi), \frac{t_{12}}{\tau_c^{12}}, \frac{4\Delta}{\alpha_1+\alpha_2} \ll 1 \tag{4.10}$$

must be imposed to satisfy Eq. (4.9), implying that the pulses have to be strongly correlated, only slightly delayed, and the difference in the detunings of levels $|1\rangle$ and $|2\rangle$ cannot be very large.

Under condition (4.9) the roots of Eq. (4.4) are given by

$$\lambda_1 = \tfrac{1}{4}(\alpha_1+\alpha_2)\epsilon_1, \tag{4.11}$$

$$\lambda_2 = (\alpha_1+\alpha_2)(1-\epsilon_2), \tag{4.12}$$

$$\lambda_{3,4} = \tfrac{1}{4}(\alpha_1+\alpha_2)(1+\epsilon_{3,4}), \tag{4.13}$$

where

$$\epsilon_1 = \frac{3\sin^2(2\beta)(\Phi G^2+1-\Phi+\Delta_0^2)}{4(1+\Delta_0^2)} \ll 1, \tag{4.14}$$

$$\epsilon_2 = \frac{3\sin^2(2\beta)[\Phi G^2+9(1-\Phi)+\Delta_0^2]}{16(9+\Delta_0^2)} \ll 1, \tag{4.15}$$

$$\epsilon_{3,4} = \frac{\epsilon_2-\epsilon_1}{2} \pm \left(\frac{(\epsilon_2-\epsilon_1)^2}{4} + \epsilon_1 + 3\epsilon_2 - \sin^2(2\beta)[\Phi G^2+3(1-\Phi)] - \Delta_0^2\right)^{1/2}. \tag{4.16}$$

The root λ_1 is much smaller than the real parts of the other three. Consequently, in a strong-field regime (4.1) only the term having index λ_1 can provide a contribution to the solutions of Eqs. (2.13)–(2.15) which is not exponentially small. The solution in this limit is of the form

$$\rho_{00}(t_p) = \tfrac{1}{3}[1+C_1\exp(-\lambda_1 t_p)], \tag{4.17}$$

$$\rho_{11}(t_p) = \frac{1}{3}\left(1+\frac{C_1(\alpha_1-2\alpha_2)\exp(-\lambda_1 t_p)}{\alpha_1+\alpha_2}\right), \tag{4.18}$$

$$\rho_{22}(t_p) = \frac{1}{3}\left(1+\frac{C_1(\alpha_2-2\alpha_1)\exp(-\lambda_1 t_p)}{\alpha_1+\alpha_2}\right), \tag{4.19}$$

$$\rho_{21}(t_p) = \frac{2C_1\sqrt{\Phi\alpha_1\alpha_2}[(1+G)\alpha_2+(1-G)\alpha_1]}{(\alpha_1+\alpha_2+4i\Delta)(\alpha_1+\alpha_2)} \times \exp(-\lambda_1 t_p), \tag{4.20}$$

with C_1 given by

$$C_1 = \frac{(2\alpha_2-\alpha_1)n_{01}(0)+(2\alpha_1-\alpha_2)n_{02}(0)}{2(\alpha_1+\alpha_2)}. \tag{4.21}$$

For the Λ system, taking into account Eqs. (2.20) one has

$$C_1 = -\frac{1}{4}\left(1+\rho_-(0)\frac{3(\alpha_1-\alpha_2)}{\alpha_1+\alpha_2}\right), \tag{4.22}$$

where $\rho_-(0) = \rho_{22}(0)-\rho_{11}(0)$. Below I present the results for the most important cases.

A. One strong and one weak pulse

According to conditions (4.1) and (4.2) this case is characterized by

$$\alpha_1 \gg t_p^{-1} \gg \alpha_2. \tag{4.23}$$

For the Λ configuration, taking into account Eqs. (4.23), substituting C_1 in the form (4.22) into Eqs. (4.17)–(4.20), and expanding these equations to first order of $\alpha_2 t_p$, one has

$$\rho_{00}(t_p) = \frac{\rho_{11}(0)}{2} + \frac{\alpha_2}{2\alpha_1}\rho_-(0) + \frac{\alpha_2 t_p(\Phi G^2+1-\Phi+\Delta_0^2)[1+3\rho_-(0)]}{16(1+\Delta_0^2)}, \tag{4.24}$$

$$\rho_{11}(t_p) = \frac{\rho_{11}(0)}{2} + \frac{\alpha_2}{4\alpha_1}[1+5\rho_-(0)] + \frac{\alpha_2 t_p(\Phi G^2+1-\Phi+\Delta_0^2)[1+3\rho_-(0)]}{16(1+\Delta_0^2)}, \tag{4.25}$$

$$\rho_{22}(t_p) = \rho_{22}(0) - \frac{\alpha_2}{4\alpha_1}[1+7\rho_-(0)] - \frac{\alpha_2 t_p(\Phi G^2+1-\Phi+\Delta_0^2)[1+3\rho_-(0)]}{8(1+\Delta_0^2)}, \tag{4.26}$$

$$\rho_{21}(t_p) = \sqrt{\Phi\alpha_2/\alpha_1}\frac{[1+3\rho_-(0)](G-1)}{4(1+i\Delta_0)}. \tag{4.27}$$

One can see from Eqs. (4.24)–(4.26) that level populations in a Λ system may significantly depend on delay time if an atom is prepared initially in one of the lower sublevels, $|1\rangle$ or $|2\rangle$.

For $\rho_{11}(0)=1$, the level populations are shown in Figs. 2(a)–2(c), (curve 1). Levels $|0\rangle$ and $|1\rangle$ linked by a strong field constitute a two-level system. As shown in Figs. 2(a) and 2(b), their populations are equal at time t_p and are close to 0.5. Level $|2\rangle$ is linked to the excited level, $|0\rangle$, by a weak pulse. Although the population of level $|2\rangle$ is small, in contrast to a weak-field regime, this population, as a function of delay time, consists of a background signal having value $\alpha_2 t_p/4$ and a dip of width

$$\delta t_{12} \sim \tau_c^{12}, \tag{4.28}$$

and relative depth $\Phi/(1+\Delta_0^2)$ centered at zero delay time $t_{12}=0$ [see Fig. 2(c), curve 1]. The dip vanishes for noncorrelated pulses ($\Phi=0$) or for a large difference in the detuning of excited states ($\Delta \gg \alpha_1$). Averaged atomic coherence is given by Eq. (4.27). As shown in Fig. 2(d) (curve 1), it is strongly asymmetrical function of delay time. Owing to the condition $\rho_-(0)=-1$, this asymmetry is negative, and is characterized by a time scale τ_c^{12}.

If an atom is initially pumped into level $|2\rangle$, that is, if $\rho_{22}(0)=\rho_-(0)=1$, the populations of the states $|0\rangle$ and $|1\rangle$ strongly depend on t_{12} representing similar dips of width τ_c^{12} and relative depth $\Phi/(1+\Delta_0^2)$, as shown in Figs. 3(a) and 3(b) (curve 1). The atomic coherence given by Eq. (4.27) is an asymmetric function of delay time. As one can see from Fig. 3(d) (curve 1), in contrast to the case considered above [$\rho_{11}(0)=1$], the sign of the atomic coherence is changed. This change occurs when $\rho_{22}(0)=\frac{1}{3}$ and can be already seen in Fig. 4, where an intermediate case corresponding to equal initial populations of lower levels is shown.

B. Both pulses are strong

It is shown below that the most dramatic dependence of the averaged density matrix elements on delay time occurs in a regime described by the condition

$$\alpha_1 \geq \alpha_2 \gg t_p^{-1}. \tag{4.29}$$

One can see from Eqs. (4.11) and (4.14) that the parameter $\lambda_1 t_p$ can now be large enough to satisfy $\exp(-\lambda_1 t_p) \ll 1$. Specifically, if condition (4.5)

$$\frac{3\alpha_1\alpha_2 t_p[\Phi G^2(t_{12})+1-\Phi+\Delta_0^2]}{4(\alpha_1+\alpha_2)(1+\Delta_0^2)} \gg 1 \tag{4.30}$$

is satisfied, the atomic coherence ρ_{21} vanishes, and all three atomic levels are equally populated [see Eq. (4.8)], that is,

$$\rho_{21}(t_p)=0, \quad \rho_{mm}(t_p)=\tfrac{1}{3}, \quad m=1,2,3. \tag{4.31}$$

For noncorrelated pulses ($\Phi=0$) this result holds for any delay time. However, even for fully correlated pulses ($\Phi=1$) this situation occurs for a delay time

$$|t_{12}| \gg \tau_c^{12}\eta^{-1}, \tag{4.32}$$

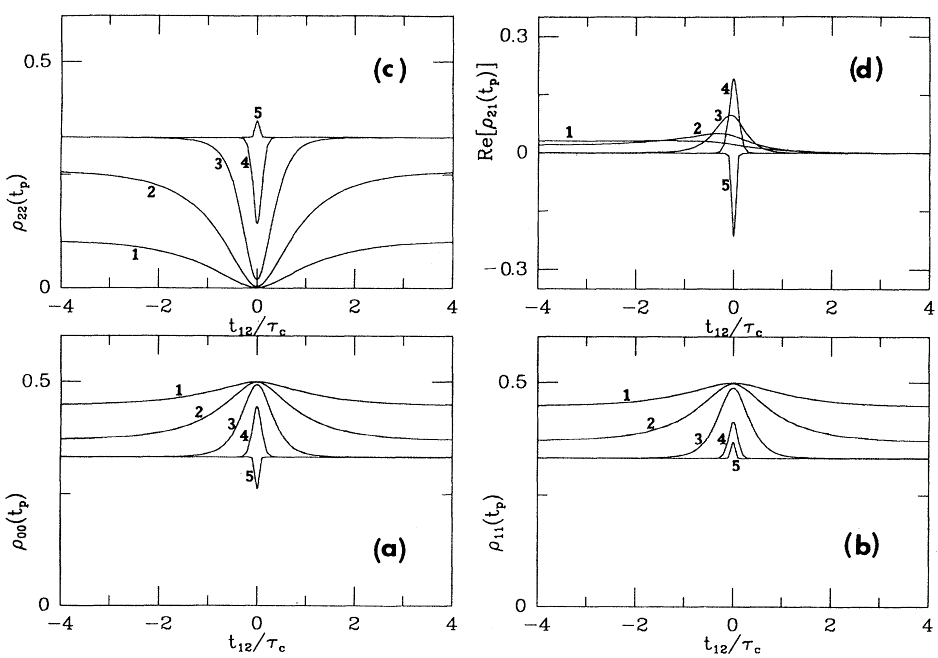

FIG. 2. The averaged populations and atomic coherence of a Λ system as functions of delay time t_{12} for $\rho_{11}(0)=1$. The populations $\rho_{00}(t_p)$, $\rho_{11}(t_p)$, and $\rho_{22}(t_p)$ are shown in (a), (b), and (c), respectively. The atomic coherence ρ_{21} is presented in (d). The first of two fully correlated pulses ($\Phi=1$) is strong ($\alpha_1 t_p=10^3$), while an intensity of the second pulse varies: $\alpha_2 t_p=$ 0.5 (curve 1), 2 (curve 2), 10 (curve 3), 10^2 (curve 4), 10^3 (curve 5). The calculations are carried out for $\Delta t_p=10$.

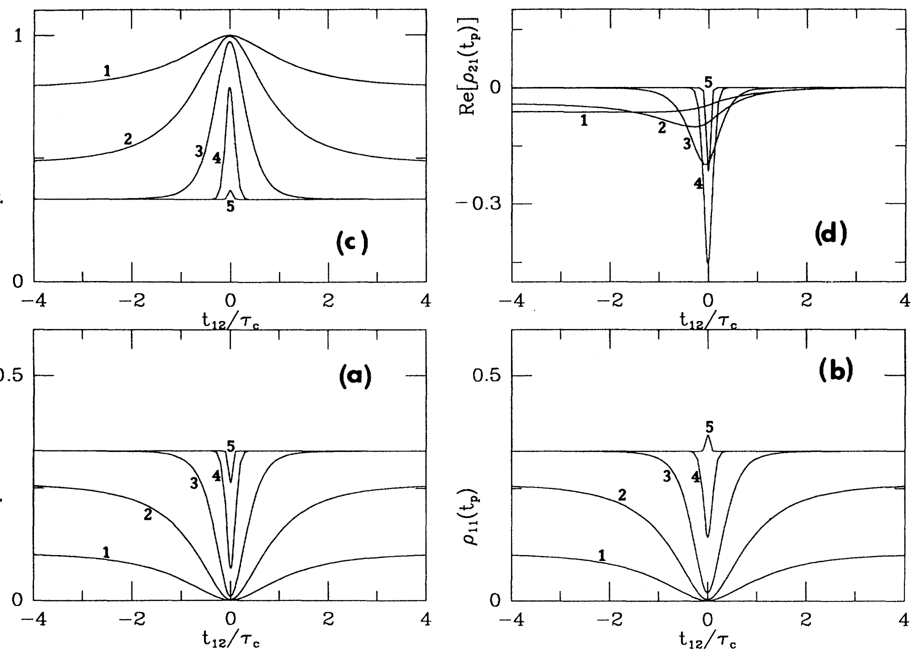

FIG. 3. The same as Fig. 3, but $\rho_{22}(0)=1$.

where η is given by Eq. (1.4), and for strong pulses (4.29) $\eta \gg 1$. Thus, background values of density matrix elements which are reached for relatively large delay times correspond to equal populations and vanishing atomic coherence.

If the pulses are strongly correlated, and the difference of the detunings of levels $|1\rangle$ and $|2\rangle$ is relatively small, that is, if

$$(1-\Phi), \Delta_0^2 < \eta^{-2} \ll 1. \tag{4.33}$$

the condition $\lambda_1 t_p < 1$ can be satisfied, provided the pulses are delayed only slightly

$$G^2(t_{12}) < \eta^{-2} \ll 1. \tag{4.34}$$

Consequently, in the limit (4.33) significant dependence of populations and atomic coherence on delay time may

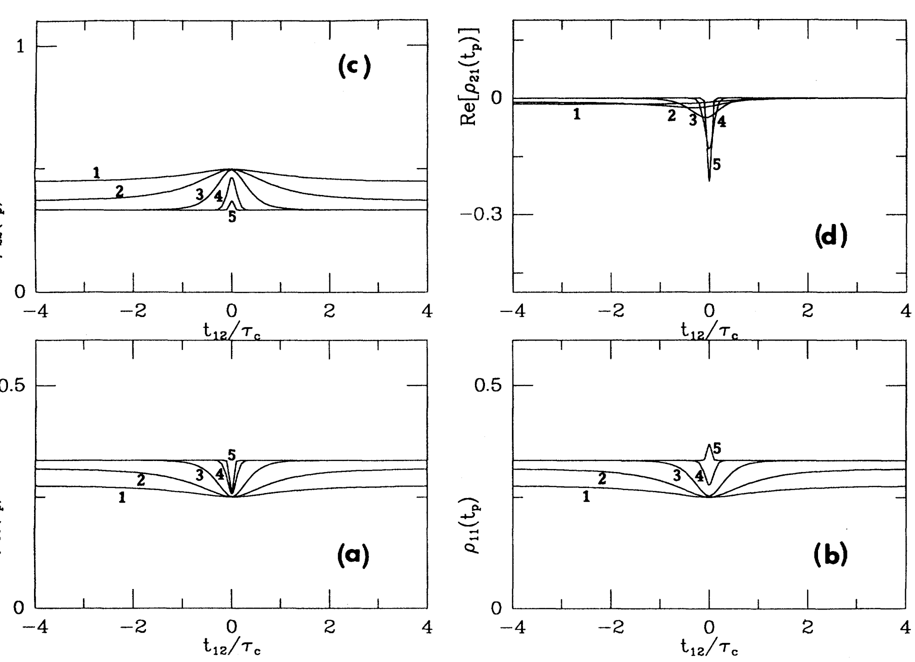

FIG. 4. The same as Fig. 3, but $\rho_{11}(0) = \rho_{22}(0) = \frac{1}{2}$.

occur. According to Eq. (4.34) the time scale of this dependence is given by

$$\delta t_{12} \approx \tau_c^{12}/\eta \ll \tau_c^{12}, \tag{4.35}$$

implying that level populations and atomic coherence of a three-level atom as functions of delay time may vary on a time scale which is much smaller than cross-correlation time of the excitation pulses. The effect resembles that for a two-level atom[17] [see Eq. (1.3)]. However, for a three-level atom the effect can be much more significant.

The case of a Λ configuration is illustrated in Figs. 2–4, curves 3–5. The population of excited level, ρ_{00}, as a function of t_{12}, exhibits a peak [see Fig. 2(a)] if

$$\alpha_1 > 2\alpha_2 \tag{4.36}$$

and

$$\rho_{11}(0) - \rho_{22}(0) > \frac{\alpha_1 + \alpha_2}{3(\alpha_1 - \alpha_2)}. \tag{4.37}$$

The width of this peak decreases with increasing intensity of the second pulse and is given by Eq. (4.35). As one can see from Fig. 2(a) (curve 5), and Figs. 3(a) and 4(a), if any of conditions (4.36) and (4.37) is not satisfied, the peak inverts into a dip having width (4.35). The relative depth of this dip is given by

$$\frac{\rho_{00}(t_p; t_{12} \gg \tau_c^{12}\eta^{-1}) - \rho_{00}(t_p; t_{12} = 0)}{\rho_{00}(t_p; t_{12} \gg \tau_c^{12}\eta^{-1})}$$

$$= \frac{1}{4}\left(1 + \frac{3[\rho_{22}(0) - \rho_{11}(0)](\alpha_1 - \alpha_2)}{\alpha_1 + \alpha_2}\right)\xi, \tag{4.38}$$

where

$$\xi = \exp[-3\eta^2(1 - \Phi + \Delta_0^2)/4]. \tag{4.39}$$

The depth (4.38) reaches its maximum value ξ for

$$\rho_{22}(0) = 1 \quad \text{and} \quad \alpha_1 \gg \alpha_2, \tag{4.40}$$

and can be close to unity for fully correlated pulses [see Fig. 3(a)].

Behavior of the level population $\rho_{22}(t_p)$, as a function of delay time, is just the opposite of $\rho_{00}(t_p)$. As shown in Figs. 2–4(c), if $\rho_{00}(t_p)$ exhibits a peak, $\rho_{22}(t_p)$ exhibits a dip, and vice versa.

Population of level $|1\rangle$, as a function of t_{12}, exhibits a dip only if condition (4.36) is satisfied, while (4.37) is violated [Figs. 2–4(b)]. As one can see from Fig. 3(b), the dip acquires its maximum relative depth ξ under conditions (4.40).

Averaged atomic coherence given by Eqs. (4.20) and (4.22), as a function of delay time, exhibits a narrow peak with a zero background [see Figs. 2–4(d)]. If conditions (4.36) and (4.37) are violated, the peak changes its sign [for $\mathrm{Re}(\rho_{21})$ it changes from positive to negative]. The peak acquires its maximum amplitude

$$\rho_{21}^{\max}(t_p) = -\frac{0.56\xi\sqrt{\Phi}}{1 + i\Delta_0} \tag{4.41}$$

for $\rho_{22}(0) = 1$ and for the pulse intensities $\alpha_2 = 0.228\alpha_1$.

The most important feature of the results presented above is a very rapid variation of the averaged density matrix elements with delay time. The temporal width of peaks and dips is of order of $\tau_c^{12}\eta^{-1}$ which is much smaller than the cross-correlation time of the pulses. It

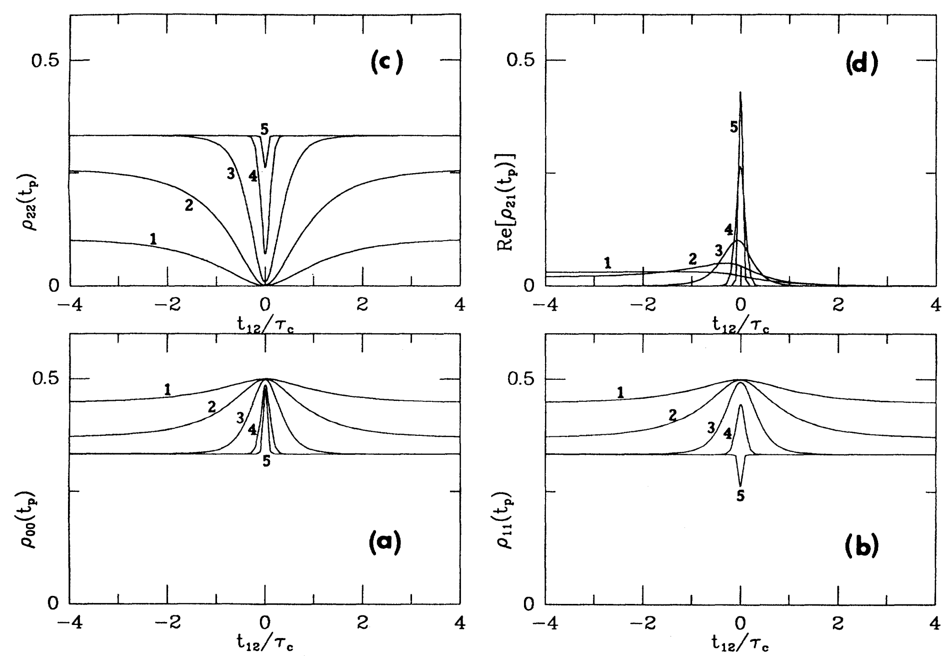

FIG. 5. The averaged populations and atomic coherence of a V system as functions of delay time t_{12} for $\rho_{00}(0) = 1$. The populations $\rho_{00}(t_p)$, $\rho_{11}(t_p)$, and $\rho_{22}(t_p)$ are shown in (a), (b), and (c), respectively. The atomic coherence ρ_{21} is presented in (d). The parameters of the pulse are the same as in Fig. 2.

is also important that for fully correlated pulses with very different intensities, the relative depth of dips can be close to unity, and the peak for atomic coherence has zero background.

The case of the V system is illustrated in Fig. 5 and is discussed in detail in the Appendix. Comparing Figs. 5 and 2, one can see that, for the V configuration, the dependence of the density matrix elements on delay time is very similar to that of density matrix elements of Λ system initially pumped into level $|1\rangle$. A significant difference emerges only for $2\alpha_2 > \alpha_1$. In this case, for populations, peaks obtained for the Λ system correspond to dips for the V system, and vice versa, and the peaks for atomic coherence are of opposite signs.

V. DISCUSSION

To give a qualitative explanation of the strong-field results, one can define the set of states (1.5) and (1.6) introduced in Sec. I in a more general way as

$$|b\rangle = \cos\beta|1\rangle + \sin\beta|2\rangle,$$
$$|c\rangle = \sin\beta|1\rangle - \cos\beta|2\rangle, \qquad (5.1)$$

where β is given by Eq. (4.6). The probability amplitudes of levels $|1\rangle$ and $|2\rangle$ can be expressed in terms of probability amplitudes of states $|b\rangle$ and $|c\rangle$ as

$$a_1 = a_b\cos\beta + a_c\sin\beta,$$
$$a_2 = a_b\sin\beta - a_c\cos\beta. \qquad (5.2)$$

The qualitative consideration for the Λ and V systems is similar. In this section, for the sake of simplicity, I consider the V configuration [the initial condition for this scheme is always given by $a_0(0) = 1$]. Using representation (5.2) and taking into account Eqs. (2.5) one obtains the following equations for the population amplitudes of the "new" states (5.1):

$$i\dot{a}_0 = \tfrac{1}{2}\{[f_1^*(t)\cos\beta + f_2^*(t-t_{12})\sin\beta]a_b + [f_1^*(t)\sin\beta - f_2^*(t-t_{12})\cos\beta]a_c\}, \qquad (5.3)$$

$$i\dot{a}_b = -(\delta_1\cos^2\beta + \delta_2\sin^2\beta)a_b + \sin\beta\cos\beta(\delta_1-\delta_2)a_c + \tfrac{1}{2}[f_1(t)\cos\beta + f_2(t-t_{12})\sin\beta]a_0, \qquad (5.4)$$

$$i\dot{a}_c = -(\delta_1\sin^2\beta + \delta_2\cos^2\beta)a_c + \sin\beta\cos\beta(\delta_1-\delta_2)a_b + \tfrac{1}{2}[f_1(t)\sin\beta - f_2(t-t_{12})\cos\beta]a_0. \qquad (5.5)$$

One can see from Eq. (5.5) that state $|c\rangle$ is completely decoupled from other states of the system if

$$\Delta = \delta_2 - \delta_1 = 0 \qquad (5.6)$$

and

$$f_1(t)\sin\beta = f_2(t-t_{12})\cos\beta,$$

or (5.7)

$$f_1(t)\sqrt{\alpha_2} = f_2(t-t_{12})\sqrt{\alpha_1}.$$

Condition (5.6) holds for equal detunings of levels $|1\rangle$ and $|2\rangle$. The second condition, (5.7), is satisfied for any monochromatic fields $f_1(t)$=const and $f_2(t)$=const and does not depend on delay time. Specifically, the effect of decoupling leads to population trapping in Λ systems for Δ=0.

If the excitation fields vary with time, Eq. (5.7) can be satisfied only for $f_1(t)/f_2(t)$=const, that is, if one field is an exact replica of another. For stochastic fields this means that they have to be fully correlated (Φ=1) with zero delay time, $t_{12} = 0$. In this particular case the existence of population trapping was shown by Dalton and Knight.[27] Under conditions (5.6) and (5.7) in the absence of relaxation, the probability amplitudes of states $|1\rangle$ and $|2\rangle$ are linked to each other by a relation

$$[a_1(t) - a_1(0)]\sin\beta = [a_2(t) - a_2(0)]\cos\beta, \qquad (5.8)$$

that is, amplitude $a_1(t)$ is proportional to $a_2(t)$ with an additional constant shift, although for fluctuating pulses both amplitudes are now stochastic functions of time. For instance, let us consider the case when the first pulse is much stronger than the second one, that is, $f_1(t) \gg f_2(t)$, and level $|0\rangle$ is initially populated. Then, the parameter β is small ($\beta \ll 1$), and from Eq. (5.8) one has $a_2(t)/a_1(t) = \beta \ll 1$, that is, level $|2\rangle$ is populated much less than level $|1\rangle$ for *any* given time.

As soon as condition (5.6) or (5.7) do not hold,[35] state $|c\rangle$ is coupled to the rest of the system, and the temporal evolution of a three-level atom driven by stochastic fields, changes dramatically. For large time $t \sim +\infty$ all the atomic coherences $\langle a_m^*(t)a_n(t)\rangle$, ($n \neq m$ and $n,m = 0,1,2$ or $n,m = 0,b,c$) tend to zero, while all the populations $\langle a_m^*(t)a_m(t)\rangle$ tend to $\frac{1}{3}$. This steady-state distribution does not depend on initial conditions and is a manifestation of a more general result concerned with dynamics of a many-level quantum system under the influence of a stochastic field characterized by a spectrum with power wings. One can prove[36] that if spontaneous decay is neglected, the density matrix of a system with N nondegenerate levels tends to $\langle a_m^*(t)a_n(t)\rangle = N^{-1}\delta_{mn}$ for $t \sim +\infty$. The physical origin of this result can be traced to the white-noise type of the spectrum of a stochastic laser field or one of its time derivatives. In this case the laser field can be considered as a reservoir characterized by an infinite temperature. A quantum system eventually comes to thermal equilibrium with this reservoir, which results in equal populations of all the nondegenerate levels. However, for a given time t_p under different conditions the system can be at different stages of a transient process leading to this equilibrium distribution. For instance, for fully correlated pulses ($\Phi = 1$) and equal detunings $\delta_1 = \delta_2$, one can estimate the rate of this process in a V system [$a_0(0) = 1$] by examining the population of state $|c\rangle$. For $t_{12} = 0$, one has $|a_c(t)|^2 = 0$. To calculate $\langle|a_c(t_p)|^2\rangle$ for $t_{12} \neq 0$, one can consider the case, when an excitation process has two stages characterized by different rates. First, the population is quickly redistributed between states $|0\rangle$ and $|b\rangle$. Then, slow "leakage" to state $|c\rangle$ comes into play and leads to equal distribution of population among all three levels. To estimate the rate of this slow process one can consider times for which state $|c\rangle$ is still populated only slightly, that is $a_c(t) \ll 1$. Then from Eqs. (5.3)–(5.5) one has

$$a_c(t) = -\tfrac{1}{4} \int_0^t \exp[i\delta_1(t-t')][f_1(t')\sin\beta - f_2(t'-t_{12})\cos\beta] \int_0^{t'} [f_1^*(t'')\cos\beta + f_2^*(t''-t_{12})\sin\beta] a_b(t'') dt' dt''. \tag{5.9}$$

To calculate the population of state $|c\rangle$, one takes into account Eqs. (2.6)–(2.8) and condition (2.11). Using Eq. (5.9), and averaging the resulting equation for $|a_c(t)|^2$ over field fluctuations, one arrives at

$$\langle |a_c(t)|^2 \rangle = \frac{\alpha_1\alpha_2 G^2(t_{12})}{16} \times \int \int_0^t \langle a_b^*(t') a_b(t'') \rangle e^{i\delta_1(t'-t'')} dt' dt'', \tag{5.10}$$

where $G(t_{12})$ is given by Eq. (2.19). The correlation function of the probability amplitude $a_b(t)$ in Eq. (5.10) can be estimated in the framework of a two-level system $|0\rangle \Leftrightarrow |b\rangle$.[37] Using Eqs. (5.3) and (5.4), applying a decorrelation approximation, and taking into account that, for a two-level system driven by a strong field, for $t \gg (\alpha_1+\alpha_2)^{-1}$ the relation $\langle |a_b(t')|^2 \rangle = \frac{1}{2}$ is valid, one arrives at

$$\langle a_b^*(t') a_b(t'') \rangle = \tfrac{1}{2} \exp\{-[i\delta_1(t'-t'') + \tfrac{1}{4}(\alpha_1+\alpha_2)|t'-t''|]\}, \tag{5.11}$$

where $t', t'' \gg (\alpha_1+\alpha_2)^{-1}$. Substituting Eq. (5.11) into Eq. (5.10) and taking the integral one arrives at

$$\langle |a_c(t_p)|^2 \rangle = \frac{\alpha_1\alpha_2 G^2(t_{12}) t_p}{4(\alpha_1+\alpha_2)} = \frac{\lambda_1 t_p}{3}. \tag{5.12}$$

Equation (5.12) gives a quantitatively correct excitation rate λ_1 [see Eqs. (4.11) and (4.14)] of a slow process that eventually leads to equal populations of all the states, and strongly depends on delay time.

The temporal resolution that one can obtain using the dependence of populations on delay time in a strong-field regime is given by Eq. (4.35). Under conditions considered in this paper, the theoretical limit for this resolution can be found by assuming that $\alpha_1^{-1} = \alpha_2^{-1} = \tau_c^{12}$, and $t_p = T_1$, where T_1 is the lifetime of the excited level(s). Then, one has

$$(\delta t_{12})_{\text{lim}} = 2 \left(\frac{2(\tau_c^{12})^3}{3T_1} \right)^{1/2}. \tag{5.13}$$

The time resolution given by Eq. (5.13) may be extremely high. For τ_c^{12}=1 ps, and $T_1 = 2.1 \times 10^{-5}$ s ($5\,^1S_0$–$5\,^3P_1$ transition in Sr) one arrives at $(\delta t_{12})_{\text{lim}} = 3.6 \times 10^{-16}$ s. For currently obtainable laser pulses, the achievable time resolution is not quite so good. For pulses with equal energies of 2 mJ, t_p=10 ns, τ_c^{12}=100 fs, and a laser beam diameter of 1 mm, one obtains $\alpha_1 = \alpha_2 = 1.7 \times 10^{11}$ s^{-1}, and $\delta t_{12} = 4 \times 10^{-15}$ s for the $4\,^3P_1$–$5\,^3S_1$ transition in Ca (Λ configuration). Thus, a time resolution equal to a few optical periods might be achieved.

Recently, considerable attention has been given to different techniques leading to high spatial resolution of atomic particles.[38–43] In particular, methods using optical Raman transitions in a highly inhomogeneous magnetic field or optical standing wave were suggested[43] to obtain submicrometer accuracy in position measurements.

The results obtained in this paper imply that using time-delayed, correlated, fluctuating laser pulses one may achieve high spatial resolution in the absence of any external potentials characterized by large gradients. According to Eq. (2.1), the delay time actually depends on location of an atom through

$$t_{12}(\mathbf{r}) = t_{12} + \frac{(\mathbf{n}_2 - \mathbf{n}_1)\cdot\mathbf{r}}{c}.$$

Hence, a temporal resolution given by Eq. (4.35) leads to a spatial resolution

$$\delta x = \frac{c\,\delta t_{12}}{2\sin(\theta/2)} = \frac{c\tau_c^{12}}{2\eta\sin(\theta/2)}, \tag{5.14}$$

where x is a coordinate in a direction $(\mathbf{n}_1 - \mathbf{n}_2)$, and $0 \le \theta \le \pi$ is an angle between the wave vectors $\mathbf{k}_1$ and $\mathbf{k}_2$. Atoms located in the vicinity δx around the point in the atomic sample where $t_{12}(\mathbf{r}) = 0$ end up with very different distribution of populations compared to those located outside that region. Using an estimate made above for δt_{12} one can see that a spatial resolution of order of 1 μm can be achieved for counterpropagating pulses.

In this paper, all the results are obtained under the assumption that, for a given atom, the delay time, $t_{12}(\mathbf{r})$, does not vary during the pulses. Since atoms move in the x direction, however, only the atom which does not leave the region δx during the excitation, is driven by the pulses with a delay time smaller than δt_{12}, and, consequently, acquires highly nonequal level populations at time t_p. The velocity v_x of this atom satisfies condition

$$\frac{2\sin(\theta/2)v_x t_p}{c} < \delta t_{12} . \qquad (5.15)$$

For all other atoms the results obtained for $|t_{12}| > \delta t_{12}$ can be applied, and all the level populations are equal to $\frac{1}{3}$. This velocity dependence of the populations leads to a velocity resolution δv_x given by

$$\delta v_x = \frac{c\delta t_{12}}{2t_p\sin(\theta/2)} = \frac{c\tau_c^{12}}{2\eta t_p \sin(\theta/2)}. \qquad (5.16)$$

An interesting feature of this velocity resolution is that it can be obtained *simultaneously* with a spatial resolution δx.

In this paper an atomic motion has been considered classically. This assumption means that the condition

$$m\delta v_x \delta x \gg 2\pi\hbar, \qquad (5.17)$$

where m is atomic mass, must be satisfied. Substituting Eqs. (5.14) and (5.16) into Eq. (5.17) one obtains a restriction on time resolution δt_{12} in the form

$$\begin{aligned}\delta t_{12} &\gg \frac{2\sin(\theta/2)}{c}\left(\frac{2\pi\hbar t_p}{m}\right)^{1/2} \\ &= 1.3\times 10^{-16}\sin(\theta/2)\left(\frac{t_p\ (\mathrm{ns})}{A\ (\mathrm{amu})}\right)^{1/2}\ \mathrm{s}, \end{aligned}\qquad (5.18)$$

where A is atomic mass in atomic mass units. For realistic laser parameters condition (5.18) is satisfied. However, for hypothetically stronger and longer pulses, the theoretical limit for δt_{12} given by Eq. (5.13) would violate the restriction (5.18), and the fully quantum treatment of the phenomenon might be necessary.

Conventional detection methods such as photoionization, absorption, transient, and propagation effects can be used to probe the final distribution of atomic level populations. One can consider observational schemes using cells as well as atomic beams. In the latter case even cw-laser radiation sources can be used to observe the effect. Since for zero delay time the resulting level populations do not depend on fluctuations, the effect under consideration can be observed on a shot to shot basis which does not require statistical averaging, in sharp contrast to most phenomena induced by fluctuating light. Atomic systems of particular interest include such atoms as Ca and Sr which have an excited state $J=1$ characterized by relatively long lifetime.

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APPENDIX: THE V CONFIGURATION

To consider the case of the V system, one can use Eqs. (2.13)–(2.15) with initial condition

$$n_{01}(0) = n_{02}(0) = \rho_{00}(0) = 1, \quad \rho_{21}(0) = 0. \qquad (A1)$$

In the weak-field regime (3.1), taking into account initial conditions (A1) and solving Eqs. (2.13)–(2.15) one gets the level populations and coherence in the form

$$\begin{aligned}\rho_{00}(t_p) &= 1 - \tfrac{1}{2}(\alpha_1+\alpha_2)t_p, \\ \rho_{mm}(t_p) &= \tfrac{1}{2}\alpha_m t_p, \quad m=1,2, \\ \rho_{21}(t_p) &= \frac{i\sqrt{\Phi\alpha_1\alpha_2}[\exp(-i\Delta t_p)-1]}{2\Delta}.\end{aligned} \qquad (A2)$$

As one can see from Eqs. (A2), in a weak-field regime the excited states are populated only slightly, and the matrix elements do not depend on the delay time t_{12}.

In the strong-field regime (4.1), the solutions (4.17)–(4.20) are still applicable, with C_1 given by

$$C_1 = \tfrac{1}{2}. \qquad (A3)$$

In the case (4.23) of one strong and one weak pulse, populations and atomic coherence are given by

$$\rho_{00}(t_p) = \rho_{11}(t_p) \approx \tfrac{1}{2}, \qquad (A4)$$

$$\rho_{22}(t_p) = \frac{\alpha_2}{2\alpha_1} + \frac{\alpha_2 t_p[\Phi G^2 + 1 - \Phi + \Delta_0^2]}{4(1+\Delta_0^2)} \ll 1, \qquad (A5)$$

$$\rho_{21}(t_p) = \sqrt{\Phi\alpha_2/\alpha_1}\,\frac{1-G}{1+i\Delta_0}. \qquad (A6)$$

Levels $|0\rangle$ and $|1\rangle$ are linked by a strong field and constitute a two-level system. At time t_p, their populations are close to $\frac{1}{2}$. The population of level $|2\rangle$ is small. As a function of delay time, it consists of a background signal of value $\alpha_2 t_p/4$ and of a dip having width τ_c^{12}. For large delay time $|t_{12}| \gg \tau_c^{12}$ the population of level $|2\rangle$ is twice as small as that given by Eq. (A2) in a weak-field regime. This result is quite understandable, since the population of the ground state is now equal to $\frac{1}{2}$. Averaged atomic coherence is given by Eq. (A6) and, as a function of delay time, exhibits a significant dependence on delay time in the form of strong negative asymmetry.

If both pulses are strong [see Eq. (4.29)], the ground-state population ρ_{00}, as a function of delay time, exhibits a peak centered at $t_{12}=0$ [see Fig. 5(a), curves 3 and 4]. The temporal width of this peak is given by Eq. (4.35), and its relative height is equal to 0.5ξ, where ξ is given by Eq. (4.39). The population of level $|2\rangle$, as a function of t_{12}, consists of a background signal (4.31) having a dip of width (4.35) and relative depth

$$\frac{\rho_{22}(t_p;t_{12}\gg\tau_c^{12}\eta^{-1}) - \rho_{22}(t_p;t_{12}=0)}{\rho_{22}(t_p;t_{12}\gg\tau_c^{12}\eta^{-1})} = \frac{2\alpha_1-\alpha_2}{2(\alpha_1+\alpha_2)}\xi, \qquad (A7)$$

which for fully correlated pulses with very different intensities can be close to unity [see Fig. 5(c), curves 3 and 4]. Population of level $|1\rangle$, as a function of delay time, for $\alpha_1 > 2\alpha_2$ exhibits a peak which inverts into a dip for $\alpha_1 < 2\alpha_2$ [see Fig. 5(b), curves 3–5, respectively]. The averaged atomic coherence given by Eqs. (4.20) and (A3) exhibits a narrow peak with no background [see Fig. 5(d), curves 3–5]. The peak acquires its maximum amplitude given by Eq. (4.41) for the pulses with equal intensities, $\alpha_1 = \alpha_2$.

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