# Interaction of a strong laser pulse with the $S_{1/2}$-$P_{1/2,3/2}$ system

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The interaction of the sodium three-level system with a frequency-fixed or frequency-swept strong laser pulse is studied numerically. The validity of the secular-adiabatic approximation is analyzed. The failure of this approximation, caused by the $LS$ decoupling, for the pulses with the peak Rabi frequency greater than the fine-structure splitting is shown.

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## I. INTRODUCTION

Recently, it was shown both experimentally and theoretically that the population of the sodium ground state $S_{1/2}$ can be selectively transferred to one of the excited states $P_{1/2}$ and $P_{3/2}$ by a broadband picosecond frequency-chirped laser pulse [1]. The direction of the frequency sweeping determines which state is excited, i.e., the state whose resonance frequency is first met by a changing pulse carrier frequency. Such selective excitation takes place even when the peak Rabi frequency exceeds the fine-structure splitting. It was suggested that a careful pulse-shape control is not necessary in such an experiment.

The effect described above is explained by the adiabatic rapid passage (ARP) phenomenon [2–4]. This effect consists in the complete population transfer from the ground level of a two-state system to the upper level by sweeping the pulse adiabatically through the resonance frequency. The processes analogous to ARP also take place in the multilevel systems [3–7]. Much attention was paid to the multilevel adiabatic population inversion or redistribution schemes, which involved several time-delayed pulses. However, mainly ladder and $\Lambda$ systems were studied. The paper of Melinger *et al.* [1] is, to our knowledge, the only experimental work that treats the simple single pulse adiabatic inversion process in the atomic $V$ system.

The striking feature of the experiment performed by Melinger *et al.* [1] was that no evidence of the fine-structure breaking by the laser field was found. One can expect that the character of the excitation process should be changed when the Rabi frequency exceeds the fine-structure splitting. The $S_{1/2}$-$P_{1/2,3/2}$ system irradiated by such a strong light field transforms, roughly speaking, into an $S_0$-$P_1$ one. In order to investigate such a transformation we have performed the numerical simulation of the interaction of the $S_{1/2}$-$P_{1/2,3/2}$ sodium system with the monochromatic, linearly polarized light having constant amplitude [8]. We have shown that in the stationary regime, the $LS$ decoupling by the laser field results in the vanishing of the quantum interference effect [9]. Moreover, the character of the spectra of the scattered light is changed. In contrast to the results of [1], our calculations showed that the effect of the fine-structure breaking should be observed for the Rabi frequencies comparable with the fine-structure splitting. The time-dependent regime of the experiment is, in our opinion, the reason for this discrepancy. Nevertheless, it seems that the $LS$ decoupling should be also observed in experiments involving the pulsed excitation.

In some cases the adiabatic approximation provides analytical solutions of the equations describing the evolution of the atomic or molecular systems interacting with the light having envelope and/or carrier frequency slowly changing in time [2]. The adiabatic following effects described by these solutions are characterized by the smooth evolution of involved populations. In the present paper we intend to show the way in which the character of the interaction of the $S_{1/2}$-$P_{1/2,3/2}$ system with the relatively weak pulse with fixed or chirping frequency is changed when the pulse peak intensity increases and causes temporary $LS$ decoupling.

We use the technique introduced in [10] and adopted by us for the $S_{1/2}$-$P_{1/2,3/2}$ system in [11]. This method, presented in the example of the $S_0$-$P_1$ system in Sec. II, is based on the secular-adiabatic approximation and enables the investigation of the role of the relaxation in the adiabatic interaction of the atomic system with the light. In Sec. III we present numerical results concerning the interaction of the $S_{1/2}$-$P_{1/2,3/2}$ system with frequency-fixed pulse and in Sec. IV we show how the selectivity of the excitation can be lost in experiments similar to that performed by Melinger *et al.* [1].

## II. THE SECULAR-ADIABATIC APPROXIMATION FOR THE $S_0$-$P_1$ SYSTEM

The time evolution of the atomic system coupled to the light field and interacting collisionally with buffer gas atoms is governed by the Liouville equation for the reduced density matrix [12]

$$i\frac{d\rho}{dt} = [H, \rho] + i\Phi\rho, \tag{1}$$

where $H$ is the Hamiltonian of the system composed of the atom and the laser field and $\Phi$ is the operator describing the relaxation. In our model the matrix elements of

 

this operator are defined phenomenologically, i.e., they are obtained from the experimental spontaneous decay rates and collisional cross sections.

It is convenient to solve Eq. (1) using Liouville-space formalism [13,12]. The levels of the system considered are spatially degenerate and the appropriate basis of the Liouville space is composed of 16 elements [12]. If we assume that the incoming light has a defined polarization, the dimensionality of the problem can be reduced. For linearly polarized light the reduced basis $\{e_i\}$ of all states involved in the problem consists of 5 elements. All $e_i$ states are the spherical tensors built from the polarization vector contracted with the elements of the rotationally irreducible basis in the Liouville space (see [12] for details). The reduced density matrix is expanded in this basis according to

$$\rho = \sum_{i=1}^{5} \rho_i e_i. \tag{2}$$

In the rotating-wave approximation Eq. (1) can be written in the form of a set of the first-order differential equations for the expansion coefficients (2)

$$\frac{d\rho_i}{dt} = \sum_{j=1}^{5} A_{ij}\rho_j = \sum_{j=1}^{5} (A_{ij}^{\mathrm{SH}} + \Gamma_{ij})\rho_j, \tag{3}$$

where $i = 1, \ldots, 5$; $A^{\mathrm{SH}}$ is the skew-Hermitian matrix, which describes the Liouvillian of the atom interacting with the electromagnetic field; and the matrix $\Gamma$ contains relaxation rates. The matrix $A$ is defined in [12]:

$$\begin{Vmatrix} 0 & \sqrt{3}\gamma & 0 & v & v \\ 0 & -\gamma & 0 & -v/\sqrt{3} & -v/\sqrt{3} \\ 0 & 0 & -\Gamma_{22}^{(2)} & -2v/\sqrt{6} & -2v/\sqrt{6} \\ -v & v/\sqrt{3} & 2v/\sqrt{6} & -\Gamma_{12}^{(1)} + i\Delta & 0 \\ -v & v/\sqrt{3} & 2v/\sqrt{6} & 0 & -\Gamma_{12}^{(1)} - i\Delta \end{Vmatrix}, \tag{4}$$

where $2v(t)$ is the time-dependent Rabi frequency and $\Delta(t) = \omega_0(t) - \omega_{21}$ denotes the detuning of the incoming light frequency $\omega_0(t)$ from the resonant frequency $\omega_{21}$. The spontaneous relaxation rate is denoted by $\gamma$ and the coherence and alignment damping rates are defined by the relations [12]

$$\Gamma_{12}^{(1)} = \gamma/2 + \gamma_{12\mathrm{coll}}^{(1)}, \quad \Gamma_{22}^{(2)} = \gamma + \gamma_{22\mathrm{coll}}^{(2)}, \tag{5}$$

where the index coll stands for the collisional contributions. The collisional line shifts are neglected [12] throughout the paper.

The first two components of the density matrix (2) are related to the populations of the ground and excited levels: $n_1 = \rho_1$ and $n_2 = \sqrt{3}\rho_2$, respectively. The third component describes the alignment in the $P_1$ state and the remaining two are the coherences between states coupled by the light. The initial condition for the solution of Eq. (3) is assumed to be $\rho_1 = 1$ and other components are equal to 0.

The solutions to (3) completely describe the evolution of the considered system. Unfortunately, for arbitrary $v(t)$ and $\omega_0(t)$ this set can be solved only numerically. However, the secular-adiabatic procedure allows analytical solutions in some cases. If we transform the equations of motion to the dressed frame [10] via the transformation

$$\rho^D(t) = U^\dagger(t)\rho(t), \quad A^D(t) = U^\dagger(t)A(t)U(t), \tag{6}$$

where $U$ diagonalizes the matrix $A^{\mathrm{SH}}$ [see (3)], then the set (3) takes the following form in the dressed frame:

$$\frac{d\rho^D(t)}{dt} = \left[A^D(t) - U^\dagger(t)\frac{dU(t)}{dt}\right]\rho^D(t). \tag{7}$$

The adiabatic approximation can be performed in the dressed frame in an easy way. In general, this approximation consists in the omission of the second term in the right-hand side of (7), which can be done when

$$\left|\left[U^\dagger(t)\frac{dU(t)}{dt}\right]_{ij}\right| \ll |A_{ii}^D(t) - A_{jj}^D(t)| \qquad (i \neq j)$$
$$\left|\left[U^\dagger(t)\frac{dU(t)}{dt}\right]_{ii}\right| \ll |A_{ii}^D(t)| \tag{8}$$

for all values of $i$ and $j$.

The matrix $A^D$ is, in general, nondiagonal since the terms describing relaxation are present in the matrix $A$ [see (4)]. However, some of the nondiagonal terms can be neglected when the absolute values of the nonzero diagonal terms (the generalized Rabi frequencies) are large. Such an approximation is called a secular one [14].

The transformation $U$ is constructed from the eigenvectors of the matrix $A^{\mathrm{SH}}$. The energy differences multiplied by the imaginary unit $i$ are the eigenvalues of this matrix. The eigenvalue 0 is degenerate. In order to obtain the transformation $U$ giving proper evolution of the density matrix, one must construct eigenvectors with the zero eigenvalue which are in agreement with the constants of motion defined as [11,15,16]

$$C_n = \mathrm{Tr}\rho(t)^n = \mathrm{Tr}\rho(0)^n, \qquad n = 1, 2, ..., N, \tag{9}$$

where $N$ is the number of the system levels. In our case $N = 2$.

The transformation $U(t)$ to the dressed frame for the $S_0$-$P_1$ system is

$$U = \frac{1}{2\Omega}\begin{Vmatrix} \Omega+\Delta & \Omega-\Delta & 0 & -2iv & 2iv \\ (\Omega-\Delta)/\sqrt{3} & (\Omega+\Delta)/\sqrt{3} & 4\Omega/\sqrt{6} & 2iv/\sqrt{3} & -2iv/\sqrt{3} \\ 2(\Omega-\Delta)/\sqrt{6} & 2(\Omega+\Delta)\sqrt{6} & -2\Omega/\sqrt{3} & 4iv/\sqrt{6} & -4iv/\sqrt{6} \\ -2iv & 2iv & 0 & \Omega+\Delta & \Omega-\Delta \\ 2iv & -2iv & 0 & \Omega-\Delta & \Omega+\Delta \end{Vmatrix}, \tag{10}$$

where the $\Omega(t) = \sqrt{4v^2(t) + \Delta^2(t)}$ is the generalized Rabi frequency for the $S_0$-$P_1$ transition. To obtain eigenvectors of the zero eigenvalue, which are the first three columns of the matrix (10), we use the constants of motion

$$\begin{aligned} \rho_1(t) + \frac{1}{\sqrt{3}}[\rho_2(t) + \sqrt{2}\rho_3(t)] &= \text{const}, \\ \rho_1(t)^2 + 2\rho_4(t)\rho_5(t) + \frac{1}{3}[\rho_2(t) + \sqrt{2}\rho_3(t)]^2 &= \text{const}, \\ \sqrt{2}\rho_2(t) - \rho_3(t) &= \text{const}. \end{aligned} \tag{11}$$

The first two are determined from (9). The third one states that the linearly polarized light cannot change the population of $P_1$, $m = \pm 1$ sublevels in the $S_0$-$P_1$ system.

For the $S_0$-$P_1$ transition the second term on the right-hand side of Eq. (7) takes the form

$$U^+\frac{dU}{dt} = \frac{i}{\Omega^2}\left[v\frac{d\Delta}{dt} - \Delta\frac{dv}{dt}\right]\begin{Vmatrix} 0 & 0 & 0 & 1 & -1 \\ 0 & 0 & 0 & -1 & 1 \\ 0 & 0 & 0 & 0 & 0 \\ 1 & -1 & 0 & 0 & 0 \\ -1 & 1 & 0 & 0 & 0 \end{Vmatrix}. \tag{12}$$

If we neglect this term (the adiabatic approximation), which is possible when

$$\frac{|vd\Delta/dt - \Delta dv/dt|}{\Omega^3} \ll 1, \tag{13}$$

the population $(\rho_1^D, \rho_2^D, \rho_3^D)$ and the coherence $(\rho_4^D, \rho_5^D)$ parts of the dressed density operator are coupled only when the relaxation is present. If the following condition is fulfilled:

$$\frac{\gamma}{\Omega}, \frac{\Gamma_{12}^{(1)}}{\Omega}, \frac{\Gamma_{22}^{(2)}}{\Omega} \ll 1, \tag{14}$$

the nondiagonal terms coupling these two parts of the dressed density operator can be omitted (the secular approximation [14]). In the framework of the secular-adiabatic approximation, dressed populations and coherences evolve independently. Hence, for the assumed initial condition, the dressed coherences are not generated. It means that the evolution of the considered system is completely described by the evolution of the dressed populations. In our case they obey the following equations:

$$\begin{aligned} \frac{d\rho_2^D}{dt} &= \frac{1}{2\Omega^2}\left\{\left[4\Gamma_{12}^{(1)}v^2 - \gamma\Delta(\Omega - \Delta) - \frac{\gamma_{22\text{coll}}^{(2)}}{3}(\Omega^2 - \Delta^2)\right]\rho_1^D \right. \\ &\quad \left. - \left[4\Gamma_{12}^{(1)}v^2 + \gamma\Delta(\Omega + \Delta) + \frac{\gamma_{22\text{coll}}^{(2)}}{3}(\Omega + \Delta)^2\right]\rho_2^D\right\} + \frac{1}{3\sqrt{2}}\left[\gamma(\Omega - \Delta) + \gamma_{22\text{coll}}^{(2)}(\Omega + \Delta)\right]\rho_3^D, \\ \frac{d\rho_3^D}{dt} &= \frac{1}{3\sqrt{2}\Omega}\gamma_{22\text{coll}}^{(2)}[(\Omega - \Delta)\rho_1^D + (\Omega + \Delta)\rho_2^D] - \left(\gamma + \frac{\gamma_{22\text{coll}}^{(2)}}{3}\right)\rho_3^D, \\ \rho_1^D &+ \rho_2^D + \sqrt{2}\rho_3^D = 1. \end{aligned} \tag{15}$$

When the collisional relaxation is absent and when initially the only nonzero component of the density matrix is $\rho_1(0) = 1$, which in the dressed basis is equivalent to

$$\begin{aligned} \rho_2^D(0) &= \frac{1}{2}\left[1 - \frac{\Delta(0)}{|\Delta(0)|}\right], \\ \rho_3^D(0) &= 0, \end{aligned} \tag{16}$$

the solution of the set (15) is

$$\begin{aligned} \rho_2^D(t) &= \frac{1}{2} - \frac{1}{2}e^{-\lambda(t)}\left[\frac{\Delta(0)}{|\Delta(0)|} + \gamma\int_0^t \frac{\Delta(t')}{\Omega(t')}e^{\lambda(t')dt'}\right], \\ \rho_3^D(t) &= 0, \end{aligned} \tag{17}$$

where

$$\lambda(t) = \gamma\frac{2v(t)^2 + \Delta^2(t)}{\Omega(t)^2}. \tag{18}$$

Finally, after transformation back to the bare basis we obtain for the population of the upper level

$$n_2(t) = \frac{1}{2} + \frac{\Delta(t)}{\Omega(t)}\left[\rho_2^D(t) - \frac{1}{2}\right], \tag{19}$$

which for $\gamma = 0$ becomes

$$n_2(t) = \frac{1}{2}\left[1 - \frac{\Delta(0)\Delta(t)}{|\Delta(0)|\Omega(t)}\right]. \tag{20}$$

This last solution for the pulse swept through the resonance exhibits behavior typical for the ARP [2].

The analysis of the condition (13) reveals the difference between adiabatic excitation by the frequency-fixed and chirping pulses. When the detuning is fixed ($\Delta$ =const), the relation (13) becomes $|\Delta(dv/dt)|/\Omega^3 \ll 1$. The systems follows adiabatically the changes of the pulse envelope only when $|\Delta| \gg v(t)$ [2]. In such a case when the peak intensity of the pulse is increased, its duration also has to be increased in order to fulfill the adiabaticity condition.

The determination of the adiabatic regime for the chirping pulse is more complicated. Even for the sim-

plest model of the frequency sweeping, i.e., the linear model

$$\Delta(t) = \Delta_0 + at, \tag{21}$$

with $\Delta_0$ denoting the initial detuning and $a$ the frequency sweeping rate, the relation (13) is so involved that the calculation of the parameters describing the variation of the pulse intensity and carrier frequency is practically impossible without some additional assumptions. In our considerations (compare [11]) we assume that the pulse achieves its maximum when the carrier frequency passes the resonance. In such a case we can calculate parameters giving the adiabatic regime by applying the condition (13) when the pulse is switched on and when the pulse intensity attains its maximum value. In the simplest model, two parameters describe the pulse envelope and one the frequency sweeping. Therefore, there are several methods of setting these parameters. Since in this paper we want to study the $LS$ decoupling caused by the high intensity laser field, we are interested in the pulses having a large peak intensity. The relations obtained by us (see [11]) imply two possible ways of increasing this intensity and preserving the adiabaticity of the excitation, i.e., the increase of the pulse peak intensity has to be followed only by an increase of the pulse duration or by a decrease of the pulse duration and an increase of the sweeping rate. Since the first method requires more and more energetic pulses, it seems impractical for experimental purposes and we restrict our consideration to the second method.

We have not found any analytical solutions of the set (15) when collisional relaxation rates do not vanish. It seems that in such a case, these equations can be solved only numerically. We want, however, to study the $LS$ decoupling in the $S_{1/2}$-$P_{1/2,3/2}$ system, which under the influence of the strong field should exhibit some features of the $S_0$-$P_1$ system. In particular, it is expected that the adiabatic approximation cannot describe correctly the evolution of the $S_{1/2}$-$P_{1/2,3/2}$ system excited by the strong frequency-fixed pulse tuned to the center of gravity of the $P$ multiplet. In such a case, if the pulse peak intensity is high enough, the $S_{1/2}$-$P_{1/2,3/2}$ system transforms temporarily into the resonant $S_0$-$P_1$ system. As is well known [2], the resonant two-level system does not adiabatically follow the variation of the pulse envelope. Therefore, as we show in the next section, the evolution of the $S_{1/2}$-$P_{1/2,3/2}$ system is no longer smooth.

## III. THE EXCITATION OF THE $S_{1/2}$-$P_{1/2,3/2}$ SYSTEM BY THE FREQUENCY-FIXED PULSE

The discussion of the secular-adiabatic approximation for the $S_{1/2}$-$P_{1/2,3/2}$ system was presented in [11]. Here we only want to recall that for this system the reduced basis $\{e_i\}$ consists of 10 elements [8,11,17] and that the analytical form of the transformation to the dressed frame is not known. Nevertheless, the procedure presented in [11] can supply this transformation numerically and therefore within the secular-adiabatic approximation evolution can be determined. In this section we will compare the solutions found in such a way with exact solutions obtained by the direct integration of the equations of motion.

As in [11], we assume that the system considered is excited by a linearly switched on and exponentially decaying pulse defined by

$$v(t) = 16 v_0 \frac{t}{T} \exp\left(-4\frac{t}{T}\right), \tag{22}$$

with mean coupling $v_0$, which is proportional to the mean envelope amplitude and mean pulse duration $T$ defined by [18]

$$v_0 = \int_0^\infty v(t)^2 dt \Big/ \int_0^\infty v(t) dt,$$
$$T = \left[\int_0^\infty v(t) dt\right]^2 \Big/ \int_0^\infty v(t)^2 dt. \tag{23}$$

The envelope (22) achieves a maximum equal to approximately $1.47 v_0$ for $t = T/4$ [see Fig. 1(a)]. We consider a pulse starting at $t = 0$ since it seems to be more physical than pulses which start at $-\infty$ [19].

For $v_0$ small in comparison with the fine-structure splitting and when the collisions are absent, the $S_{1/2}$-$P_{1/2,3/2}$ system can be considered as two subsystems $S_{1/2}$-$P_{1/2}$ and $S_{1/2}$-$P_{3/2}$ not coupled by light (see [11]). These subsystems are joined only by the common ground level. Let us assume that the light is tuned to the center of gravity of the upper state multiplet. This means that the detuning from the $D_1$ transition is equal to $2\omega_{32}/3$ (see [8,11]), where $\omega_{32}$ represents the fine-structure splitting. In such a case, employing conditions obtained in [11], which are similar to (13) and (14), we can find parameters $v_0$ and $T$ describing pulses which generate adiabatic evolution for both independent $S_{1/2}$-$P_{1/2}$ and $S_{1/2}$-$P_{3/2}$ systems. In general, when the mean pulse intensity increases, the longer pulse must be used to preserve the adiabaticity of the evolution.

In order to find when the secular-adiabatic approximation can be inaccurate, we perform the numerical simulation of the evolution of the whole Na $S_{1/2}$-$P_{1/2,3/2}$ system interacting with the pulse, which ensured adiabatic evolution for the $S_{1/2}$-$P_{1/2}$, and $S_{1/2}$-$P_{3/2}$ systems, respectively. As the unit of time, we chose the lifetime of $P_{1/2}$, which is equal to 16.3 ns. All other parameters are defined in relation to the spontaneous decay rate for the Na $D_1$ transition. The parameter $v_0$ denotes the mean coupling for this transition. We assume that all collisional relaxation rates vanish.

The results are presented in Fig. 1, where we have plotted the evolution of the populations of the $S_{1/2}$ and $P_{1/2}$ states and the joint populations of the $P_{3/2}$, $m = \pm 1/2$ states. Since we assume that the collisional relaxation is absent and that the light is linearly polarized, the $P_{3/2}$, $m = \pm 3/2$ states are not populated and the Zeeman sublevels with the same $|m|$ are equally populated. In other words, the population of the $P_{3/2}$ state is equal to the total population of the $P_{3/2}$, $m = \pm 1/2$ states. The exact solutions are represented by dots. The continuous lines correspond to the adiabatic-secular results. For the shortest pulse analyzed [Fig. 1(a)], the evolution of the

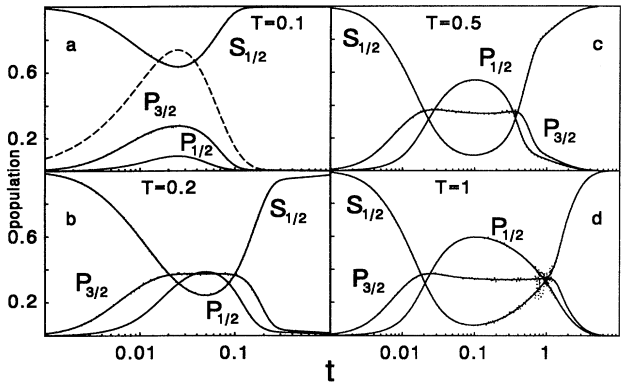


FIG. 1. The populations of the Na $S_{1/2}$, $P_{1/2}$, and $P_{3/2}$ states versus time for different frequency-fixed pulses giving adiabatic evolution. The mean coupling $v_0$ of the pulse (22) is equal to (a) $0.2\omega_{32}$, (b) $0.5\omega_{32}$, (c) $\omega_{32}$, and (d) $2\omega_{32}$. The light is tuned to the center of gravity of the $P$ multiplet. The continuous line denotes the secular-adiabatic results and the dotted line gives exact numerical results. The dashed line in (a) corresponds to the pulse envelope (22) with $v_0 = 1/2$. Here and in the following figures time is measured in lifetimes of the $D_1$ transition and other parameters are given in relation to the spontaneous decay rate for this line.

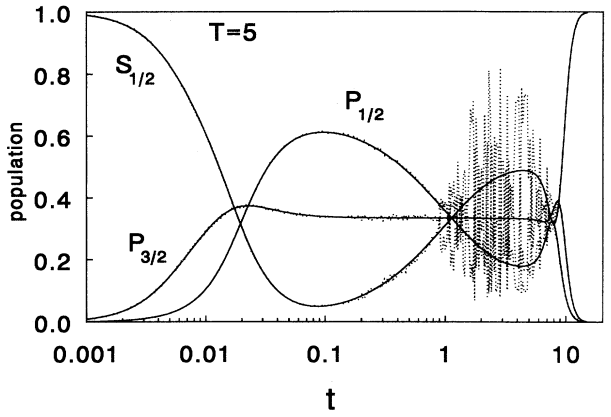


FIG. 2. The populations of the Na $S_{1/2}$, $P_{1/2}$, and $P_{3/2}$ states as functions of time. The mean coupling $v_0$ is equal to $10\omega_{32}$. The light is tuned to the center of gravity of the $P$ multiplet. The continuous line denotes the secular-adiabatic results and the dotted line gives exact numerical results.

populations of both excited states can be explained by the nonresonant excitation. Since $v_0$ is relatively small in comparison to the detunings from both transitions, these populations follow the changes of the pulse envelope. The population of the $P_{3/2}$ level is always greater than that of the $P_{1/2}$ state because the light is tuned closer to the $D_2$ transition than to the $D_1$ transition. This simple picture changes for a pulse that is both longer and stronger by a factor of 2 [Fig. 1(b)]. The population of the $P_{3/2}$ state temporarily saturates and the population inversion between both excited states and the ground state can be observed. In addition, the inversion between the $P_{1/2}$ and $P_{3/2}$ states appears. This last effect is more pronounced in Fig. 1(c).

As expected, the secular-adiabatic approximation superbly describes the evolution of the populations even for relatively strong pulses [compare Figs. 1(a)–1(c)]. However, for the strongest pulse analyzed [Fig. 1(d)], one can observe the failure of this approximation. The approximate results cannot describe strong oscillations which appear when the pulse passes its maximum. Unexpectedly, the evolution caused by the spontaneous relaxation after the passage of the pulse is described correctly. It seems that in spite of the incorrect description of the oscillations, the secular-adiabatic approximation gives the correct evolution when the oscillations are averaged out.

The discrepancy between the secular-adiabatic and exact results is better illustrated in Fig. 2, where we have plotted the evolution of the populations under investigation for long ($T = 5$) and very strong ($v_0 = 10\omega_{32}$) pulses. In the beginning the evolution is very well described by the secular-adiabatic approximation, but when the pulse achieves its maximum and the relaxation begins to influence the evolution, the exact solutions strongly oscillate, whereas the approximate ones are smooth. For such a long pulse the spontaneous relaxation becomes important. Hence the adiabatic solutions no longer follow the pulse envelope for times comparable to the lifetime.

Usually the atomic relaxation is neglected in the analysis of the adiabatic interaction of the light pulses with the atomic systems since the processes described take place in times much shorter than the relaxation times. Our results, however, presented in Figs. 1 and 2 show that the increase of the fixed-frequency pulse intensity must be followed by an increase of the pulse duration and an incorporation of the relaxation processes into the adiabatic model has to be performed.

Finally, we have investigated the time evolution of the eigenvalues of the matrix $iA^{\mathrm{SH}}$ for the Na $S_{1/2}$-$P_{1/2,3/2}$ system (see [11]). These eigenvalues are equal to the differences of the corresponding eigenenergies. Four of them are equal to 0 and six others form pairs having opposite signs. Therefore three quantities $|E_2 - (E_1 + \omega_0)|$, $|E_3-(E_1+\omega_0)|$, and $|E_3-E_2|$ describe the evolution of these energy differences ($E_1$ denotes the energy of the ground level, whereas $E_2$ and $E_3$ are the energies of the excited states $\hbar = 1$). As seen in Fig. 3, these quantities follow the variation of the pulse envelope.

These eigenvalues can be interpreted as the frequencies of the temporarily emitted light having polarization parallel to the driving one. Therefore, there are the following contributions to the time-dependent spectrum [20]: the central line with frequency $\omega_0$ and three pairs of lines with varying frequencies (see Fig. 3) placed symmetrically around $\omega_0$. When the incoming field is strong enough, but the fine structure is not destroyed, these sidelines can be identified as components of the Mollow spectrum on $D_1$ and $D_2$ transitions and as lines connected with the Raman processes between $P_{1/2}$ and $P_{3/2}$ via the ground state [22].

It seems that the time-dependent spectrum can be a tool giving insight into the $LS$ decoupling process. We

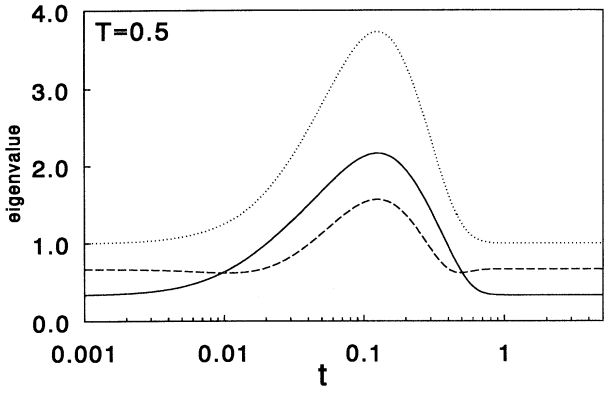


FIG. 3. The evolution of the positive eigenvalues of the matrix $iA^{\mathrm{SH}}$ [see (3)] for the Na $S_{1/2}$-$P_{1/2,3/2}$ system interacting with the frequency-fixed pulse (22) tuned to the center of gravity of the upper state multiplet. The mean pulse intensity $v_0 = \omega_{32}$. The continuous line corresponds to $|E_2-(E_1+\omega_0)|$, the dashed line to $|E_3-(E_1+\omega_0)|$, and the dotted line to $|E_3-E_2|$ eigenvalues, respectively. The magnitudes of the eigenvalues are divided by the fine-structure splitting $w_{32}$.

expect that this process should be reflected at least in the energy spectrum [20] of the light scattered by the system.

## IV. THE CHIRPING-PULSE EXCITATION OF THE $S_{1/2}$-$P_{1/2,3/2}$ SYSTEM

In this section we present results of the numerical simulation of the experiment performed by Melinger *et al.* [1]. The frequency sweeping in this experiment is nearly linear. It is also suggested that the observed effects weakly depend on the shape of the pulse envelope [1]. In our simulation, we follow these statements and use the envelope (22) and the linear sweep described by the rate $a$ [see (21)]. In order to simplify the analysis we assume that the pulse achieves a maximum when the frequency passes the resonance with one of the $D_1$ or $D_2$ transitions. In such a case we can use conditions derived in [11] to construct frequency-swept pulses giving ARP on one of these transitions. More precisely, we find parameters $T$, $v_0$, and $a$ giving adiabatic evolution for the $S_{1/2}$-$P_{1/2}$ and $S_{1/2}$-$P_{3/2}$ systems. These conditions, based on the assumption that the pulse must be adiabatic when it starts and when it passes the maximum, imply that the increase of the pulse peak intensity can be followed by a shortening of the pulse and an increase of the velocity of the pulse's frequency sweeping (see Sec. II).

First we perform calculations for the pulse lasting approximately as long as the one used in the experiment [1] ($T$=0.001 lifetimes of the $D_1$ transition, i.e., about 16.3 ps) and having a maximum equal approximately to $1.18\omega_{32}$, when the light is in resonance with the $S_{1/2}$-$P_{1/2}$ transition. The driving frequency is swept with the rate $a = 1.88 \times 10^4\omega_{32}$ per $D_1$ lifetime (19.6 $\mathrm{cm}^{-1}$/ps). We obtain results similar to those registered in the experiment, i.e., when the sweeping begins on the red side of the $D_1$ transition, the population is transferred from the ground level to the excited state $P_{1/2}$ [Fig. 4(a)]; in the opposite case for initial detuning on the blue side of the $P_{3/2}$ level, the population is transferred to the $P_{3/2}$ state [Fig. 4(b)]. The third state, $P_{3/2}$ in the first case and $P_{1/2}$ in the second, is populated only temporarily. The exact results obtained by numerical integration of the equations of motion and approximate solutions obtained in the framework of the secular-adiabatic approximation coincide.

In spite of the fact that the Rabi frequency is, in this case, greater than the fine-structure splitting, there is no evidence of the $LS$ decoupling. In our opinion, since the excitation is time dependent, the pulse with a bigger peak intensity must be used to observe this effect. In order to use a stronger pulse and obtain smooth evolution, we make the pulse shorter and the sweeping faster. In Figs. 4(c) and 4(d) we present the results of calculations performed for the pulse with $T = 0.0001$ (1.63 ps), $v_0 = 8\omega_{32}$, and $a = 1.88 \times 10^6\omega_{32}$. One can observe big discrepancy between exact and secular-adiabatic results, especially in Fig. 4(c), where the sweeping is assumed to start from the red side of the $P_{1/2}$ level. Moreover, the selectivity of the excitation predicted by the secular-adiabatic approximation does not take place. Both excited states are populated. However, the level that starts its interaction with the pulse earlier is more populated. The ground level is completely emptied.

This lack of selectivity can be explained qualitatively in the following way. The strong field transforms the $S_{1/2}$-$P_{1/2,3/2}$ system into an $S_0$-$P_1$ one in which the ARP process described in Sec. II takes place. Then the population is redistributed between $P_{1/2}$ and $P_{3/2}$, $m = \pm 1/2$ states by the $LS$ coupling.

We obtain similar results when we assume that the pulse reaches its maximum when the frequency passes

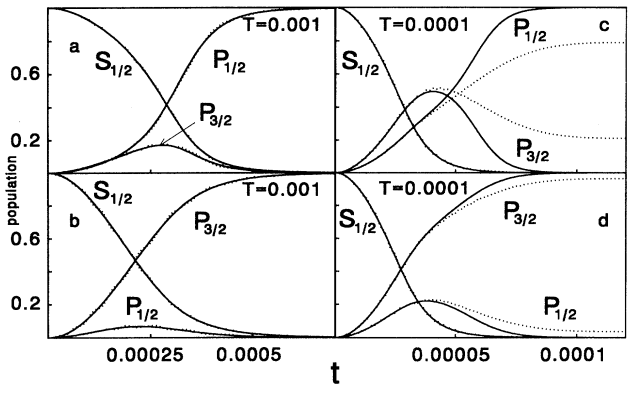


FIG. 4. The populations of the Na $S_{1/2}$, $P_{1/2}$, and $P_{3/2}$ states versus time for different frequency-swept pulses giving adiabatic evolution. The mean coupling $v_0$ of the pulse (22) and frequency sweeping rate $a$ are equal to, respectively, (a) and (b) $0.8\omega_{32}$ and $1.88\times10^4\omega_{32}$ per $D_1$ lifetime, (c) and (d) $8\omega_{32}$ and $1.88\times10^6\omega_{32}$. In (a) and (c) sweeping starts from the red side of the $P_{1/2}$ level and in (b) and (d) from the blue side of $P_{3/2}$. In all cases the pulse achieves a maximum when the driving frequency passes the resonance with the $D_1$ transition. The continuous line denotes the secular-adiabatic results and the dotted line gives exact numerical results.

the $D_2$ transition or the center of gravity of the $P$ multiplet. Therefore the precise adjustment of the frequency sweeping is not important.

We conclude that when the mean pulse intensity $v_0$ is much greater than the fine-structure splitting, the system is so strongly coupled to the electromagnetic field that the secular-adiabatic approximation, which gives correct results for the uncoupled transitions $S_{1/2}$-$P_{1/2}$ and $S_{1/2}$-$P_{3/2}$, cannot describe the behavior of the composed $S_{1/2}$-$P_{1/2,3/2}$ system. Nevertheless, this approximation gives a very good description of the excitation process when the mean intensity is comparable to the fine-structure splitting [see Fig. 4(a) and compare [1]].

If we go beyond the adiabatic approximation, the effects described above can be still present, but the population evolution becomes highly oscillatory. Moreover, the population of the ground level after passage of the pulse can be quite large.

In Fig. 5 we give the evolution of the energy differences of the system considered for the case presented in Fig. 4(a). As a consequence of the frequency sweeping the interchange of the these energy differences takes place. The difference $|E_3 - E_2|$ evolves into $|E_3 - (E_1 + \omega_0)|$, $|E_2 - (E_1 + \omega_0)|$ into $|E_3 - E_2|$, and $|E_3 - (E_1 + \omega_0)|$ into $|E_2 - (E_1 + \omega_0)|$. In other words, the dressed state $|1\rangle$ finishes its evolution as state $|2\rangle$, $|2\rangle$ as $|3\rangle$, and $|3\rangle$ as $|1\rangle$ (compare [1]).

We also investigate the dependence of the considered effect on the shape of the pulse envelope. Apart from the envelope (22), we also perform calculations for the pulse composed of two exponents (2Ex)

$$v(t) = 6v_0 \left[\exp\left(-3\frac{t}{T}\right) - \exp\left(-6\frac{t}{T}\right)\right] \tag{24}$$

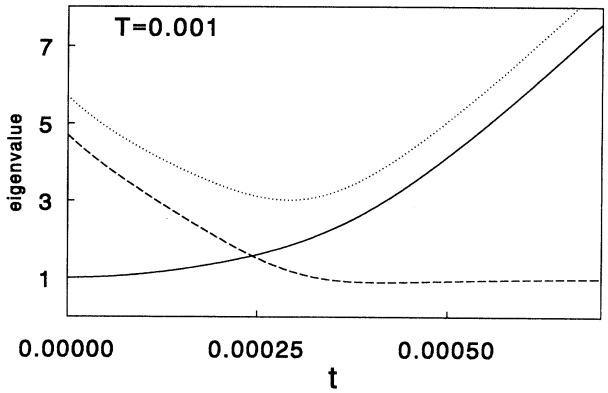


FIG. 5. The evolution of the positive eigenvalues of the matrix $iA^{\mathrm{SH}}$ for the Na $S_{1/2}$-$P_{1/2,3/2}$ system interacting with the frequency-swept pulse (22) having a maximum when the frequency passes the $D_1$ transition frequency. The mean pulse intensity $v_0 = \omega_{32}$. The frequency sweeping starts from the red side of $P_{1/2}$ state with the rate $a = 1.88 \times 10^4 \omega_{32}$ per $D_1$ lifetime. The continuous line corresponds to the eigenvalue being initially equal to $|E_3 - E_2|$, the dashed line to $|E_3 - (E_1 + \omega_0)|$, and the dotted line to $|E_2 - (E_1 + \omega_0)|$. The eigenvalues are divided by the fine-structure splitting $w_{32}$ as in Fig. 3.

and for linearly (LG) and quadratically (QG) switched Gaussian pulses

$$v(t) = \frac{16}{\pi} v_0 \frac{t}{T} \exp\left[-\frac{8}{\pi}\left(\frac{t}{T}\right)^2\right], \tag{25}$$

$$v(t) = \frac{64\pi\sqrt{2}}{27} v_0 \left(\frac{t}{T}\right)^2 \exp\left[-\frac{8\pi}{9}\left(\frac{t}{T}\right)^2\right]. \tag{26}$$

These envelopes fulfill relations (23). The envelopes (22) and (24)–(26) having the same $v_0$ and $T$ have the same energy and area in the sense that they can be considered equivalent. The plots of the functions (24)–(26) are presented in [21].

As before, we assume that each of these pulses passes maximum when its driving frequency coincides with the $D_1$ transition. The results obtained for the same parameters as in the case plotted in Fig. 4(c) are presented in Fig. 6. Since each of the envelopes considered achieves a maximum at a different time (2Ex at $0.231T$, LG at $0.443T$, and QG at $0.598T$), the plots of the population evolution for different pulses are shifted, but they are qualitatively similar. The final populations after the passage of the pulse for different envelopes differ slightly. In that sense the excitation process can be treated as independent of the shape of the pulse envelope (compare [1]).

## V. FINAL REMARKS

We have investigated numerically the Na system interacting with the short laser pulse. The cases of the frequency-fixed and frequency-swept pulses have been studied. The relevance of the secular-adiabatic approx-

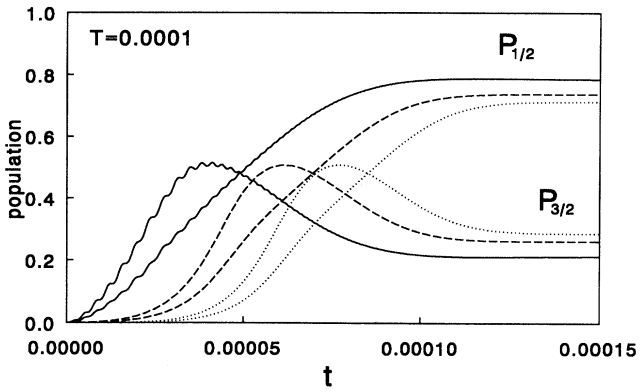


FIG. 6. The exact evolution of the populations of the Na $P_{1/2}$ and $P_{3/2}$ states for different frequency-swept pulses giving adiabatic evolution. The mean coupling $v_0 = 8\omega_{32}$ and frequency sweeping rate $a = 1.88 \times 10^6 \omega_{32}$. The sweeping starts from the red side of the $P_{1/2}$ level. The pulses achieve a maximum when the driving frequency passes the resonance with the $D_1$ transition. The continuous line corresponds to the 2Ex envelope, the dashed line to LG, and the dotted line to QG.

imation has been tested. Since the analytical condition for the adiabaticity of the interaction for the $S_{1/2}$-$P_{1/2,3/2}$ system cannot be found, we have constructed the appropriate pulses, demanding that they should give the adiabatic evolution of the systems $S_{1/2}$-$P_{1/2}$ and $S_{1/2}$-$P_{3/2}$ taken separately. It was shown that such an approach is correct for the pulses having a mean intensity smaller or comparable to the fine-structure splitting. It seems that the interaction of the stronger pulses with the $S_{1/2}$-$P_{1/2,3/2}$ system cannot be described in the framework of the secular-adiabatic approximation. However, this approximation provides an accurate account of the evolution in the initial region when the field intensity is smaller than the energy gap between excited levels.

The pulse's frequency sweeping is a very useful technique which enables the creation of the population inversion in atomic and molecular systems. Usually the sequence of pulses is used to produce inversions in the multilevel systems. Melinger *et al.* have shown [1] experimentally and theoretically that one strong and short chirping pulse can transfer the population of the ground level in the $S_{1/2}$-$P_{1/2,3/2}$ system to one of the excited levels. However, the experiment and the numerical simulation were performed in a regime in which the breaking of the fine-structure coupling does not influence the results of the sweeping. We have shown that for fields one order of magnitude stronger and pulses one order shorter than those used in the experiment, the selectivity of the excitation, which is the most striking experimental result, can be removed.

Our calculations have demonstrated that the process of population transfer to the upper level, selective or unselective, weakly depends on the pulse shape and frequency-sweeping adjustment. Such robustness of this process [1] allows easy experimental applications.

We have shown in [8] that the $S_{1/2}$-$P_{1/2,3/2}$ system under the influence of the strong stationary field exhibits some features of the $S_0$-$P_1$ system. When the excitation has pulsed character, the fine structure is broken only temporarily. Nevertheless, the evolution is influenced by the effects associated with the $S_0$-$P_1$ system and the perturbation of the smooth adiabatic evolution presented in Sec. III or the population redistribution by the $LS$ interaction described in Sec. IV should be observed.

## ACKNOWLEDGMENTS

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