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## Complete population inversion in a multilevel quantum system on adiabatic application of an external resonance field

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When an external resonance field is applied sufficiently slowly, a multilevel quantum system can be transferred from the initial ground state to any other higher level and the probability of this process is close to unity. However, this excitation mechanism is impossible in the case of a two-level system. The interval of external field frequencies in which a system can be excited does not increase when the field intensity is increased and, in principle, can be made as small as we please, which ensures a high excitation selectivity. This inversion mechanism acts at quite realistic values of the external field intensity and of the rate of its rise.

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### 1. INTRODUCTION

Most of the investigations of the resonance action of radiation on quantum systems have been carried out using a two-level model for which it is quite easy to obtain a clear analytic solution valid in the resonance approximation or in the approximation of a rotating wave.<sup>1-3</sup> Recently, even books have been devoted entirely to the subject of the two-level model (see, for example, Ref. 3). However, one can justifiably use

the two-level model only as long as it describes correctly all the qualitative features of real systems and the model has to be refined in quantitative features of real systems and the model has to be refined in quantitative calculations. We shall show that multilevel systems can exhibit a certain qualitatively different effect which does not occur in two-level systems. This effect is as follows: when an external resonance field is applied sufficiently slowly, a system initially in the ground quantum state can be transferred to a higher level with

a probability close to unity, i. e., a complete population inversion can be obtained on application of the field.<sup>1)</sup>

Population inversion has been considered earlier using a two-level model and it has been associated mainly with an adiabatically slow variation (scanning) of the external field frequency (see, for example, Refs. 4 and 5). Inversion as a result of change in the intensity of an external field has been discussed in several papers.<sup>6-11</sup> The mechanism proposed there reduces essentially to inversion as a result of a change in the frequency because it is based on the dynamic Stark effect, i. e., on the quadratic dependence of the effective frequency of a transition on the amplitude of an external resonance field. However, the Stark effect is of the second order in respect of the external field and in most cases it is far too weak for this purpose. We shall show that in the case of multilevel systems a population inversion on increase in the intensity is a first-order effect in respect of the external field (Autler-Townes effect<sup>12</sup>) and, therefore, it can be observed in practice in all real systems.

We shall consider quasienergies and quasistationary states<sup>13-15</sup> of a multilevel quantum system (Sec. 2). Next, we shall demonstrate the possibility of degeneracy of such quasistationary states and of lifting of this degeneracy by an external field (Sec. 3). Excitation of quantum systems expressed in the language of quasistationary states in fact involves lifting of the degeneracy of these states.<sup>16</sup>

In contrast to the majority of the earlier investigations, we shall not regard the concepts of quasienergies and quasistationary states as a terminological "decoration" of the theory but as a working tool which makes all the results extremely clear (at least to the present authors). Section 4 contains a discussion and numerical estimates. These estimates show that a population inversion can occur under typical experimental conditions. It is quite possible that the strong population of higher levels already achieved in a number of experiments is at least partly due to the adiabatic inversion effect.

In the Appendix we shall consider the excitation of multilevel systems under the action of several external fields of different frequencies.

## 2. QUASIENERGIES AND QUASISTATIONARY STATES

Let  $H$  be the Hamiltonian of a quantum system in the absence of an external field,  $|n\rangle$ —stationary states without a time factor,  $E_n$ —the energies of these states. We shall assume that the ground state energy is  $E_{n=0} = 0$ . The action of an external field can be described by supplementing the Hamiltonian with the term  $-\hat{d}E \cos \omega_1 t$ , where  $\hat{d}$  is the dipole moment operator,  $\omega_1$  is the carrier frequency, and  $E$  is the field amplitude. The values of  $E$  and  $\omega_1$  can vary slowly with time (the criterion of slowness will be formulated below). We shall consider only a system with an almost equidistant spectrum:  $E_{n+1} - E_n \ll \hbar\omega$ . This condition may be satisfied by, for example, vibrational levels of

a molecule or by strongly excited states of a hydrogenic atom. In the Appendix we shall show that when several lasers are used, a population inversion can be achieved also in the case when the spectrum of the system is far from equidistant. We shall assume initially that  $E$  and  $\omega_1$  are independent of time. It is convenient to represent the state vector  $|\psi\rangle$  in the form

$$|\psi\rangle = \sum_n a_n(t) \exp(-in\omega_1 t) |n\rangle. \quad (1)$$

There are such solutions of the Schrödinger equation that the coefficients  $a_n(t)$  are

$$a_n(t) = b_n^\alpha \exp(-iQ_\alpha t/\hbar), \quad (2)$$

where  $Q_\alpha$  and  $b_n^\alpha$  are independent of time. Then, the state vector (1) describes quasistationary states; the quantity  $Q_\alpha$  is the quasienergy. If the external field intensity is much less than a typical atomic or molecular value of  $10^8$  V/cm, which corresponds to an intensity  $I = cE^2/8\pi = 10^{13}$  W/cm<sup>2</sup>, we find that in the case of systems with an almost equidistant spectrum the coefficients  $a_n(t)$  do not change greatly during the period  $2\pi/\omega_1$  of the external field. This makes it possible to simplify the calculations by applying the averaging method.

Substituting Eqs. (1) and (2) in the Schrödinger equation, going over to the equations for the coefficients  $b_n^\alpha$ , and averaging with respect to time, we obtain the following system of algebraic equations:<sup>2)</sup>

$$\sum_m H_{n,m} b_m^\alpha = Q^\alpha b_n^\alpha, \quad (3)$$

where

$$H_{m,n} = \hbar \begin{pmatrix} -\delta\omega_0 & f_{01} & 0 & \dots \\ f_{10} & -\delta\omega_1 & f_{12} & \dots \\ 0 & f_{21} & -\delta\omega_2 & \dots \\ \dots & \dots & \dots & \dots \end{pmatrix},$$

$$\begin{aligned} \hbar\delta\omega_n &= n\hbar\omega_1 - E_n \text{ (clearly, } \delta\omega_0 = 0), \\ f_{n,n+1} &= Ed_{n,n+1}/2\hbar \end{aligned}$$

is the field broadening of the  $n \rightarrow n+1$  transition, and  $d_{n,n+1} = \langle n | \hat{d} | n+1 \rangle$  is the dipole moment of the transition.

Equation (3) is derived ignoring the matrix elements  $\langle n | \hat{d} | m \rangle$  if  $|n-m| \geq 2$ , because these elements are much smaller than  $\langle n | \hat{d} | n \pm 1 \rangle$ . All the quantities  $f_{n,n+1}$  can be regarded as real and positive.

Each quasistationary state is characterized by a quasienergy  $Q^\alpha$  and by a set of coefficients  $b_n^\alpha$  ( $n=0, 1, 2, \dots, N$ ) which are, respectively, the eigenvalues and vectors of the matrix  $H_{n,m}$ . The quantities  $Q^\alpha$  and  $b_n^\alpha$  depend parametrically on  $\omega_1$  and  $E$ . Degenerate quasistationary states appear when one quasienergy  $Q^\alpha$  corresponds to two or more sets of coefficients  $b_n^\alpha$ . Any state can be expanded in terms of quasistationary states because the latter form a basis in the state space. We shall write down such an expansion in the form

$$|\psi\rangle = \sum_{n,\alpha} c^\alpha(t) b_n^\alpha \exp\left(-\frac{iQ^\alpha t}{\hbar} - in\omega_1 t\right) |n\rangle.$$

The coefficients  $c^\alpha$  of the above expansion are generally functions of time.

The further analysis is based on the adiabatic theo-

rem<sup>15,17</sup> according to which a system initially in some specific quasistationary state of number  $\alpha$  remains in the same state when the parameters are varied sufficiently slowly. In other words, the coefficients  $c^\alpha(t)$  are not affected by adiabatic changes in the parameters. It should be stressed that such variation can alter greatly the quantities  $Q^\alpha$  and  $b_n^\alpha$ . Changes in  $b_n^\alpha$  may be accompanied by a considerable change also in the populations  $p_n$  of the stationary states  $|n\rangle$  which are given by  $p_n = |b_n^\alpha|^2$ , if the system is in a quasistationary state of number  $\alpha$ .

### 3. DEGENERACY OF QUASISTATIONARY STATES

We shall first consider the well-known properties of a two-level system using the concept of quasistationary states in the adiabatic theorem. Elementary calculations give the eigenvalues and the eigenvectors of the matrix  $H_{n,m}$  ( $n, m = 0, 1$ ):

$$\begin{aligned} Q^{1,0} &= \hbar \left[ -\frac{\delta\omega_1}{2} \pm \left( \frac{\delta\omega_1^2}{4} + f_{01}^2 \right)^{1/2} \right], \\ b_0^{1,0} &= 2^{-1/2} \left[ 1 \pm \frac{\delta\omega_1}{(\delta\omega_1^2 + 4f_{01}^2)^{1/2}} \right]^{1/2}, \\ b_1^{1,0} &= \pm 2^{-1/2} \left[ 1 \mp \frac{\delta\omega_1}{(\delta\omega_1^2 + 4f_{01}^2)^{1/2}} \right]^{1/2}. \end{aligned} \quad (4)$$

The dependence of  $Q^\alpha$  on the frequency detuning  $\delta\omega_1$  is shown in Fig. 1. The upper curve corresponds to a quasistationary state with  $\alpha=1$  and the lower to one with  $\alpha=0$ . The dashed line is the dependence of  $Q^\alpha$  on  $\delta\omega_1$  in the absence of an external field when  $f_{01}=0$ . The horizontal dashed line corresponds to a stationary state  $n=0$ ; the dashed line at an angle corresponds to a stationary state  $n=1$ . When the curve approaches one of the dashed lines, the quasistationary state approaches the corresponding stationary state. Let us assume that initially the system is in a state identified by A in Fig. 1. Frequency scanning causes the system to evolve, on the basis of the adiabatic theorem, along the  $\alpha=0$  curve, i.e., along ABC. Consequently, the system assumes the stationary state  $n=1$ .

The adiabatic criterion is

$$\left| \frac{d}{dt} \delta\omega_1 \right| \ll f_{01}^2.$$

If this criterion is not obeyed, the evolution of the system follows approximately the path ABDE. In this case the system is not excited.

In the case of adiabatic frequency scanning the excita-

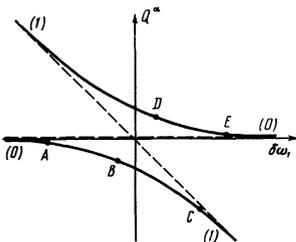


FIG. 1. Quasienergies  $Q^\alpha$  of a two-level system plotted as a function of frequency detuning  $\delta\omega_1$ . The numbers in parentheses label the stationary state to which a quasistationary state reduces on approach of the dashed and continuous lines.

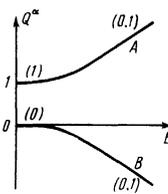


FIG. 2. Quasienergies  $Q^\alpha$  of a two-level system plotted as a function of the external field amplitude  $E$ . In Figs. 2, 3, and 4 the numbers on the left of the  $Q^\alpha$  axis identify the number  $\alpha$  of the corresponding quasistationary state. The numbers in parentheses identify stationary states which make a significant contribution in the expansion of a given quasistationary state.

tion is possible because in the absence of an external field ( $f_{01}=0$ ) the quasistationary states become degenerate for  $\delta\omega_1=0$  and the degeneracy is lifted if allowance is made for  $f_{01}$ . The dependence of  $Q^\alpha$  on the external field intensity  $I$  is shown in Fig. 2. The curves in Fig. 2 do not intersect in any approximation and, therefore, it is not possible to invert a two-level system by varying  $I$ .

*Three-level system.* In this case we can already see a basic difference between multilevel and two-level systems. Let us assume that the external frequency  $\omega_1$  does not vary. We shall label quasistationary states so that  $Q^{\alpha=n}(I \rightarrow 0) = -\hbar\delta\omega_n = -n\hbar\omega_1 + E_n$ . Then, a quasistationary state with  $\alpha=n$  reduces in the limit  $I \rightarrow 0$  to a stationary state  $|n\rangle$ . In the case of a three-level system we shall introduce frequencies of one- and two-photon transitions denoted by  $\omega_{01}$  and  $\omega_{02}$ , respectively; clearly, we have  $\hbar\omega_{01} = E_1$  and  $2\hbar\omega_{02} = E_2$ . We shall assume that  $\omega_{02} < \omega_{01}$  and that the frequency  $\omega_1$  lies in the interval  $\omega_{02} < \omega_1 < \omega_{01}$ . Then, in the limit  $I \rightarrow 0$ , we have  $Q^0 = 0$  and  $Q^2 < 0 < Q^1$ .

The expression for the eigenvalues and the vectors of the three-dimensional matrix  $H_{n,m}$  ( $n, m = 0, 1, 2$ ) are generally very cumbersome. Therefore, we shall assume that the upper level  $n=2$  is coupled only weakly to the level  $n=1$ ; more accurately, we shall assume that  $d_{12} \ll d_{01}$  and, therefore,  $f_{12} \ll f_{01}$ . Figure 3 shows the dependence of  $Q^\alpha$  (where  $\alpha = 0, 1, 2$ ) on  $I$ . Analytic expressions for quasienergies and quasistationary states can be found in Ref. 18. However, even without these expressions it is clear that if  $f_{12}$  is generally zero, then  $Q^{0,1}$  are given by Eq. (4), whereas we have  $Q^2 = -\hbar\delta\omega_2 = E_2 - 2\hbar\omega_1$ , irrespective of  $I$ . In this case the quasilevels  $\alpha=0$  and  $\alpha=2$ , i.e., the lines  $Q^0(I)$  and  $Q^2(I)$  may intersect (dashed lines in Fig. 3) at some

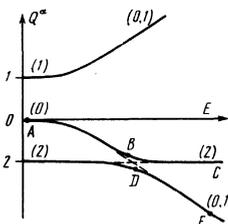


FIG. 3. Quasienergies  $Q^\alpha$  of a three-level system plotted as a function of the external field amplitude  $E$ .

value  $I=I_c$ , found from the condition

$$f_{01}^2 = \delta\omega_1(\delta\omega_2 - \delta\omega_1). \quad (5)$$

Allowance for the fact that  $f_{12} \neq 0$  lifts the degeneracy; at the same time a complete population inversion becomes possible. Let us assume that the system is initially in a state identified by  $A$  in Fig. 3; then, only the ground level  $n=0$  is significantly populated:  $p_0 \approx 1$ ,  $p_{1,2} \ll 1$ . An adiabatic increase in the intensity  $I$  causes the system to evolve along the path  $ABC$  ( $\alpha=0$ ). We can easily see that if the difference at the point  $C$  is  $Q^2 - Q^0 \gg \hbar f_{12}$ , then  $p_{0,1} \ll 1$  and  $p_2 \approx 1$ . Thus, the system is transferred to the upper level  $n=2$ . The population of this level is given, as usual, by the expression  $p_2 = |b_2^0|^2$  and in this case ( $\delta\omega_1 < 0$ ,  $\delta\omega_2 > 0$  and  $f_{01} \gg f_{12}$ ) it is equal to<sup>18</sup>

$$p_2 = \frac{1}{2} \left[ 1 - \frac{Q^0/\hbar + \delta\omega_2}{[(Q^0/\hbar + \delta\omega_2)^2 + 4f_{12}^2]^{\frac{1}{2}}} \right],$$

where  $Q^0$  is the quasienergy in the two-level approximation of Eq. (4).

The quantity

$$f = 2^{-\frac{1}{2}} f_{12} \left[ 1 + \frac{\delta\omega_1}{(\delta\omega_1^2 + 4f_{01}^2)^{\frac{1}{2}}} \right]^{\frac{1}{2}}$$

determines the width of the region where the transition of the system to the  $n=2$  state takes place. Inversion occurs for  $Q^0 < -\hbar\delta\omega_2$  and hence we obtain the condition (5).

Thus, in the case of adiabatic application of an external field we can expect 100% inversion of the  $0-2$  transition. This result differs basically from the possibility of inversion as a result of instantaneous application of the field, when for the optimal selection of  $f_{01}/f_{12}$  the maximum attainable inversion is only 2% (see Ref. 2, §9.4).

It can easily be shown that in the present case the adiabatic condition is

$$\left| \frac{df_{01}}{dt} \right| \ll \left| \left\langle f_{01} \left| \delta\omega_2 \right| \left( \frac{d_{12}}{d_{01}} \right)^2 \right\rangle \right|, \quad |d_{01}|^2 \quad (6)$$

for  $I=I_c$  corresponding to Eq. (5). If the condition (6) is not satisfied, the system evolves along the path  $ABDE$ . At the point  $E$  the populations  $p_n$  are  $p_0 \approx p_1 \approx \frac{1}{2}$ ,  $p_2 \ll 1$ .

It is interesting to note that the smallness of the dipole moment  $d_{12}$  helps in a sense to populate the level  $n=2$ . Although when  $d_{12}$  is small, it is more difficult to satisfy the adiabatic condition (6), the quasistationary state  $\alpha=0$  at the point  $C$  almost coincides with the stationary state  $n=2$ . However, if  $d_{12} \sim d_{01}$ , then the quasistationary state  $\alpha=0$  at the point  $C$  includes considerable admixtures of the stationary states  $n=0$  and  $n=1$ , so that the population is distributed more or less uniformly between the three levels  $n=0, 1, 2$ .

It is worth noting one feature of the population of the levels resulting from an adiabatic increase in  $I$ . Let us assume that the external field is so strong that  $f_{01}$ ,  $f_{12} \gg \delta\omega$ ,  $\delta\omega_2$  and we still have  $\delta\omega_1 < 0 < \delta\omega_2$ , i.e., that the frequency  $\omega_1$  lies in the interval  $(\omega_{02}, \omega_{01})$ . Then the quasienergy  $Q^0$  and the vector  $b_n^0$  are ( $n=0, 1, 2$ )

$$Q^0 = \hbar\delta\omega_2 \frac{f_{01}^2}{f_{01}^2 + f_{12}^2},$$

$$b_n^0 = (f_{01}^2 + f_{12}^2)^{-\frac{1}{2}} \left( f_{12}, \delta\omega_2 \frac{f_{01}f_{12}}{f_{01}^2 + f_{12}^2}, f_{01} \right).$$

If, as before, we have  $f_{01} \gg f_{12}$ , then almost the whole population is at the upper level  $n=2$ , in spite of the fact that the field broadening of  $f_{01}$  and  $f_{12}$  is much greater than the characteristic detuning  $\delta\omega_1$  and  $\delta\omega_2$ . In the case of instantaneous application of such a strong field the population would have been distributed uniformly between all three levels  $n=0, 1$ , and  $2$ .

It should be now stressed that adiabatic application of the field makes it possible to attain any desired excitation selectivity which is not limited by the field broadening  $f_{01}$  or  $f_{12}$ . We have shown that, irrespective of the final intensity of the radiation, the system goes over to the state  $n=2$  if  $\omega_1$  lies in the interval  $(\omega_{02}, \omega_{01})$ . On the other hand, outside this interval the level  $n=2$  cannot be excited. In fact, in this case when  $I=0$  the signs of  $Q^1$  and  $Q^2$  are the same and the application of the field simply increases  $|Q^0 - Q^2|$ , so that intersection of the curves  $Q^0(I)$  and  $Q^2(I)$  is impossible even for  $f_{12}=0$ . Thus, the width of the interval in which the external field frequency  $\omega_1$  should lie to ensure a transition of the system to the upper level 2 is  $|\omega_{01} - \omega_{02}|$  and can be as small as we please irrespective of the external field intensity. It should be stressed that in the case of instantaneous application of the field the width of the interval of effective excitation of the level 2 is  $f_{12}/\sqrt{2}$  (Ref. 18), which is much greater than  $|\omega_{01} - \omega_{02}|$ . This may be of interest in ultrahigh-resolution spectroscopy and in the separation of isotopes when the isotopes when the isotopic shift is small.<sup>19</sup>

In the limiting case of strong fields when  $f_{01}, f_{12} \gg \delta\omega_1, \delta\omega_2$ , we can easily find the vector  $b_n^0$  even when the external frequency is outside the interval  $(\omega_{02}, \omega_{01})$ . The populations  $p_n$  are given by

$$p_0 = \frac{f_{01}^2}{2(f_{01}^2 + f_{12}^2)}, \quad p_1 = \frac{1}{2}, \quad p_2 = \frac{f_{12}^2}{2(f_{01}^2 + f_{12}^2)}.$$

It should be noted that  $p_1 > p_0$ , i.e., the transition  $0-1$  always ceases inversion. If  $f_{12} \gg f_{01}$ , then  $p_0 \ll 1$ ,  $p_1 = p_2 = \frac{1}{2}$ , i.e., we obtain inversion also as a result of the  $0-2$  transition.

*Multilevel systems.* In the quantitative sense such systems differ little from the three-level case in the sense that again the system may be transferred to any one of the higher levels. We shall not analyze in detail the great variety of possibilities in the case of multilevel systems. Only by way of example we shall consider the case when  $f_{01} \gg f_{n-1,n}$ ,  $n=2, 3, \dots, N$ . To be specific, we shall assume that  $\omega_{0n} < \omega_{0,n-1}$  ( $n\hbar\omega_{0n} = E_n$ ); this case is encountered in the vibrational modes of the majority of molecules. Let the laser frequency  $\omega_1$  lie within the interval  $(\omega_{0,N-2}, \omega_{0,N-1})$  (see Fig. 4, which shows the  $N=5$  case). If all the values of  $f_{n-1,n}$ , except  $f_{01}$  vanish, then an increase in the intensity  $I$  reduces the quantity  $Q^0(I)$  because of repulsion between the quasilevels  $\alpha=0$  and  $\alpha=1$ , and if  $I$  is sufficiently high, the curve  $Q^0(I)$  can intersect lines  $Q^1(I)$  and  $Q^2(I)$  (Fig. 4).

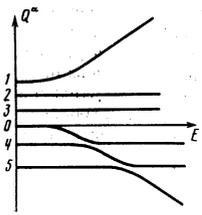


FIG. 4. Quasienergies  $Q^\alpha$  of a six-level system plotted as a function of the external field amplitude  $E$ .

Allowance for the fact that  $f_{n-1,n} \neq 0$  makes the intersection impossible, exactly as in the case of a three-level system. When the quasilevels  $Q^0(I)$  and  $Q^{\alpha=n}(I)$  come closer together, all the formulas obtained for the three-level system remain valid if the matrix element  $d_{12}$  is replaced with  $d_{1n}$ , which is the effective matrix element for an  $(n-1)$ -photon transition. The system then goes over to that level  $k$  which is the first to satisfy, on increase in  $I$ , the adiabatic condition (6) when the  $Q^0(I)$  and  $Q^{\alpha=k}(I)$  curves approach one another (Fig. 4). In this example the matrix elements  $d_{1k}$  decrease on increase  $k$  because of an increase in the number of absorbed photons and, therefore, the system either goes to the level  $k=4$  on the first approach between the  $Q^0(I)$  and  $Q^\alpha(I)$  curves or the adiabatic condition is not satisfied for any approach.

#### 4. DISCUSSION

We shall now obtain some numerical estimates demonstrating the possibility of observing the effect under discussion. We shall note first of all that in order to observe inversion (if  $d_{01} > d_{12}$ ), we must have

$$f_{01} t_f \gg (d_{01}/d_{12})^2, \quad (7)$$

where  $t_f$  is the duration of the pulse front. When the laser frequency is optimal from the point of view of satisfying simultaneously the two adiabatic conditions in Eq. (6) [so that  $\delta\omega_1 = -(d_{12}/d_{01})\delta\omega_2$ ] and when  $|E_2 - 2E_1| < \hbar f_{01}$ , the condition (7) ensures that the excitation is adiabatic. This condition is close to the conditions for the observation of other coherent effects (self-induced transparency, photon echo, etc.<sup>3</sup>) which appear when the area under a pulse  $f_{01} t_f$  ( $t_f$  is the pulse duration) exceeds unity. Clearly, the investigated effect of adiabatic inversion of multilevel systems should be observed when the same techniques and the same conditions are used as for the other coherent effects.

We shall now obtain the conditions for inversion of transitions in vibrational-rotational molecular modes whose spectrum is nearly equidistant. Let us assume that

$$E(v, J)/\hbar c = \omega_e \left( v + \frac{1}{2} \right) - x_e \omega_e \left( v + \frac{1}{2} \right)^2 + B_e J(J+1),$$

where  $(v, J)$  is a vibrational-rotational level of the molecule,  $x_e \omega_e$  is the anharmonic constant, and  $B_e$  is the rotational constant. We shall consider the transition  $(0, J) \rightarrow (1, J-1) \rightarrow (2, J)$ . Then, adiabatic application of an external field can transfer the system to an upper level  $(2, J)$  provided the laser frequency  $\omega_l$  lies between  $\omega_e - 2x_e \omega_e - 2B_e J$  and  $\omega_e - 3x_e \omega_e$ , and if the con-

dition  $f_{01}^2 \gg f_{12}^2$  is satisfied. This last condition can be satisfied by circular polarization of the laser radiation because this reduced the rotational component of the matrix elements.

In fact, in the case of a  $(v, J, M) \rightarrow (v+1, J-1, M+1)$  ( $M$  is the projection of the momentum along the wave vector direction), we have

$$f_{01} = \frac{E d_{v,v+1}^{\text{vib}}}{2} \left[ \frac{(J-M)(J-M-1)}{4J^2-1} \right]^{1/2},$$

and for the next step  $(v+1, J-1, M+1) \rightarrow (v+2, J, M+2)$

$$f_{12} = \frac{E d_{v+1,v+2}^{\text{vib}}}{2} \left[ \frac{(J+M+2)(J+M+1)}{4J^2-1} \right]^{1/2}.$$

The vibrational components of the matrix elements are related by

$$d_{v-1,v}^{\text{vib}} = (v)^{1/2} d_{01}^{\text{vib}}.$$

Then, in the case when  $v=0, M=-J$ , we obtain

$$f_{12}^2/f_{01}^2 = 1/J(J+1/2),$$

and the population of the upper level  $(2, J, -J+2)$  is equal to  $(1 + f_{12}^2/f_{01}^2)^{-1}$  after adiabatic application of the field and on increase in  $J$  this population rapidly approaches unity.

We shall next assume that the laser frequency  $\omega_l$  satisfies the optimal condition so that  $\delta\omega_1 = -(d_{12}/d_{01})\delta\omega_2$ . Then, a population inversion occurs if

$$I > I_c = \frac{4\hbar^2 c^3 (x_e \omega_e - 2B_e J)^2}{\pi (d_{01})^2} \frac{4J^2 - 1}{(J-M)(J-M-1)}. \quad (8)$$

The equality  $I_c$  in the condition (8) is valid if  $d_{12} \ll d_{01}$ , whereas for  $d_{12} \sim d_{01}$  it gives an order-of-magnitude estimate of  $I_c$ . The adiabatic condition (6) becomes  $(dI/dt) \sim I/t_f$

$$t_f \gg \frac{1}{4c|x_e \omega_e - 2B_e J|} \frac{(J-M)(J-M-1)}{(J+M+2)(J+M+1)}. \quad (9)$$

The conditions (8) and (9) can easily be satisfied for the following selection of  $J$  applicable to many molecules. Specifically, in the case of  ${}^6\text{LiF}$  [ $\omega_e = 964.07 \text{ cm}^{-1}$ ,  $x_e \omega_e = 8.895 \text{ cm}^{-1}$ ,  $B_e = 1.5087 \text{ cm}^{-1}$  (Ref. 20), and  $d_{01}^{\text{vib}} = 0.27 D$  (Ref. 21)] the  $(v, J, m) = (0, 3, -3) \rightarrow (1, 2, -2) \rightarrow (2, 3, -1)$  transition is characterized by  $x_e \omega_e - 2B_e J = -0.16 \text{ cm}^{-1}$  and the conditions (8) and (9) become  $I > I_c = 1 \text{ MW/cm}^2$ ,  $t_f \gg 1 \text{ nsec}$ . The laser frequency should then be  $937.02 \text{ cm}^{-1}$ , which lies in the region of continuous tuning of a  $\text{CO}_2$  laser, and the final population of the upper level  $(v', J', M') = (2, 3, -1)$  is

$$p = \{1 + 1/J(J+1/2)\}^{-1} = 0.87.$$

We shall conclude by noting an interesting possibility that the system remains excited even after the end of a laser pulse. In this case one needs to use an asymmetric laser excitation pulse with a steep trailing edge.<sup>9,10</sup> If the leading edge of this pulse satisfies the adiabatic condition and transfers the system to an excited state, whereas the trailing edge satisfies the opposite condition (instantaneous termination), the system cannot return to the ground state and remains excited until the next collision.

## APPENDIX

We shall now consider the case when the spectrum of the excited system is far from equidistant and the condition  $E_{n+1} - E_n \ll \hbar\omega_1$  cannot be satisfied. A population inversion can be achieved in such a system using several lasers each of which is close to resonance with a given transition  $E_n - E_{n-1} \ll \hbar\omega_1^{(n)}$  (the field of the  $n$ -th laser has the form  $E^{(n)} \cos\omega_1^{(n)} t$ ). In this case we again obtain Eq. (3) where the parameters  $f_{n-1,n}$  and  $\delta\omega_n$  are given by

$$\delta\omega_n = \sum_{m=1}^n \omega_1^{(m)} - \frac{E_n}{\hbar}, \quad f_{n-1,n} = \frac{E^{(n)} d_{n-1,n}}{2\hbar}.$$

Then, Eq. (3) becomes valid provided

$$f_{n,n\pm 1}, \delta\omega_n \ll |\omega_1^{(n)} - \omega_1^{(n\pm 1)}|.$$

Thus, all the results of the present investigation obtained for systems with an almost equidistant spectrum can be extended in a natural manner to arbitrary multi-level systems. It should be noted that in this case all the parameters  $f_{n,n-1}$  and  $\delta\omega_n$  of the effective Hamiltonian can be varied independently selecting the intensities and frequencies of the appropriate lasers. This differs greatly from the above case of excitation with one laser when a change in the intensity alters simultaneously all the values of  $f_{n,n-1}$  and a change in the frequency of the laser alters  $\delta\omega_n$ . Consequently, inversion is possible only for a limited number of systems. On the other hand, any three-level system can be inverted by two lasers and it is then necessary to select only their intensities so as to satisfy the conditions  $f_{01} t_f \gg 1$  and  $f_{01} > f_{12}$ , selecting the laser frequencies from the conditions

$$f_{01}^2 > \delta\omega_2 (\delta\omega_2 - \delta\omega_1), \quad \delta\omega_1 = -(d_{12}/d_{01}) \delta\omega_2.$$

<sup>1)</sup> It is necessary to stress the difference between the proposed method from the excitation by a  $\pi$  pulse, when an inversion appears only after the end of the laser pulse of a very specific area (see Ref. 3).

<sup>2)</sup> Here and later summation is not carried out over repeated indices unless the summation sign is given. The Greek indices refer to different quasistationary states; the Latin indices identify various stationary states.

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