Superselectivity of the excitation of multilevel systems by adiabatically switched-on resonance radiation

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The excitation by a near-resonance external field of a quantum system from the ground state into a band of closely spaced levels is considered. The total population $p_e(\omega_l, E)$ of the levels in the band is investigated as a function of the frequency ω_l and amplitude E of the exciting field when this field is adiabatically slowly switched on. There are three main features that distinguish the adiabatic from the instantaneous mode of application of the field. 1) The appearance of inversion in the level population: $p_e > 1/2$. Under certain conditions the band population p_e is close to unity, while the ground level is almost completely empty. 2) The ω_l dependence exhibits at certain frequencies sharp jumps, which allow the system to be excited with a high degree of selectivity. 3) If the number of levels in the band $N \geq 3$, then the Rabi oscillations (i.e., the oscillations of the population p_e as a function of the time) can, as E is increased, attenuate coherently, i.e. with neglect of all the relaxation processes. In the process the population p_e tends to unity.

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1. INTRODUCTION

There exists a certain inconsistency between the theoretical and experimental papers on the excitation of quantum systems by resonance radiation. The majority of the theoretical papers are devoted to the solution of problems in which the radiation intensity rises instantaneously at zero time from zero to the final value. At the same time, in experimental investigations performed with the aid of high-power pulsed lasers, the laser-pulse rise time is such that the problem is more adequately posed as that of excitation under conditions when the radiation is switched on adiabatically slowly. The first papers on the adiabatic excitation[1-8] have already shown that interesting qualitative effects, absent under conditions of instantaneous excitation, occur in this case. In the present paper we consider multilevel quantum systems, i.e., systems that are more complicated and, consequently, closer to reality than those considered in Refs. 1–8, and investigate for them the three most striking characteristics of adiabatic excitation: 1) the appearance of level-population inversion; 2) the very critical dependence of the total population p_e of the excited levels on the radiation frequency ω_l ("superselectivity"[1)]); 3) the coherent decay of the p_e oscillations (i.e., of the Rabi oscillations) with time toward the $p_e = 1$ level.

In considering further the function $p_e(\omega_l)$, we have in mind the following experimental setup: The system is excited by radiation having a fixed frequency ω_l and an intensity $I(t)$ that rises adiabatically from zero to some value I_m; the total population p_e of the excited levels is measured when $I(t)$ attains the value I_m; the experiment is then repeated with another frequency ω_l. This is how the $p_e(\omega_l)$ dependence arises.

Below in Sec. 2 we describe the system under investigation, and introduce a certain principle for numbering its quasi-energy and quasistationary states. This principle is not just a notation convention; it goes deeply into the physics of the matter and enables us to simplify greatly the computations in the case of a multilevel system. In Secs. 3 and 4 we consider the inversion of superselectivity in the cases of weak and strong radiation fields respectively. In Sec. 5 we consider the excitation of the system under conditions of partial adiabaticity, when transitions between the quasistationary states and the Rabi oscillations due to these transitions are possible. Section 6 contains a discussion, examples, and numerical estimates.

2. NUMBERING RULE

The system in question has a ground steady state with energy E_0 and a band of N higher-lying steady states with energies E_n ($n = 1, 2, \ldots, N$). (A system of this type is considered in Ref. 10 for the case in which the field is switched on instantaneously.) The system is excited by an external field $E = E(t)\cos\omega_l t$ such that for all n

$$|\delta\omega_n| = |\omega_l - \omega_n| \ll \omega_l, \tag{1}$$

where $\omega_n = (E_n - E_0)/\hbar$ is the resonant $0 \to n$ transition frequency and $\delta\omega_n = \omega_l - \omega_n$ is the detuning of the external-field (laser) frequency ω_l from the frequency ω_n.

We seek the wave function $|\psi(t)\rangle$ in the form

$$|\psi(t)\rangle = b_0(t)|0\rangle + \sum_{n=1}^{N} b_n(t) e^{-i\omega_l t}|n\rangle, \tag{2}$$

where the $|n\rangle$ are the steady-state wave function without the time factor and $n = 0, 1, \ldots, N$.

Substituting (2) into the Schrödinger equation and averaging over the period of the external field we obtain equations for the slow state vector $\{b_n\}$:

$$i\hbar\frac{db_n}{dt} = \sum_{m=0}^{N} H_{nm} b_m, \tag{3}$$

$$H_{nn} = -\hbar\delta\omega_n, \quad H_{0n} = H_{n0} = -\frac{Ed_{0n}}{2} = \hbar f_n. \tag{4}$$

Here d_{0n} is the dipole moment of the $|0\rangle \to |n\rangle$ transition. The phases of the states $|n\rangle$ have been chosen such that the quantities d_{0n} are real and negative; therefore, the field broadenings $f_n > 0$.

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Using the eigenvalues ε^α and the eigenvectors $\{a_n^\alpha\}$ of the matrix H_{nm}, which satisfy the equation

$$\sum_{m=0}^{N} H_{nm}a_m^\alpha=\varepsilon^\alpha a_n^\alpha \quad (\alpha=0,1,\ldots,N), \tag{5}$$

we construct the quasistationary states:

$$|\alpha\rangle=\left[a_0^\alpha|0\rangle+e^{-i\omega_l t}\sum_{n=1}^{N}a_n^\alpha|n\rangle\right]\exp\left\{-\frac{i}{\hbar}\int_{-\infty}^{t}dt'\varepsilon^\alpha(t')\right\}. \tag{6}$$

The quantities ε^α are the quasi-energies of the system.[11-13] In order to avoid confusing the quasistationary and the steady states, we denote the quasistationary states by Greek and the steady states by Latin letters.

Let us introduce for the quasistationary states a numbering independent from that of the steady states; specifically, let us number the quasistationary states in ascending order of their quasi-energies:

$$\varepsilon^0<\varepsilon^1<\varepsilon^2<\ldots<\varepsilon^N. \tag{7}$$

The proposed numbering of the quasi-energies has the following physical justification. The slow variation of the intensity or frequency of the external field will be accompanied by the variation of the quasi-energies ε^α in time. But the inequalities (7) will be satisfied at all moments of time, since the quasiterms $\varepsilon^\alpha(t)$ do not, generally speaking, intersect.[2)]

The adoption of the numbering rule (7) significantly facilitates the calculations in the case of a strong field (see Secs. 4 and 5 below), but complicates them somewhat in the case of a weak field, since a special treatment is now required for the determination of the number α_n of the quasistationary state into which a given steady state with number n goes over when the field is switched on. But this is easy to do, since the quasi-energies ε^{α_n} in a weak field are $\varepsilon^{\alpha_n}=-\hbar\delta\omega_n$. Thus, to determine α_n in accordance with the numbering rule (7), we must arrange the detunings $\delta\omega_m$ in descending order. Then α_n will be equal to the number of terms preceding $\delta\omega_n$.

For the system in question $\omega_{n+1}>\omega_n$ and $\delta\omega_{n+1}<\delta\omega_n$ $(n=0,1,2,\ldots,N-1)$. Therefore, for example, the number α_0 of the quasistationary state $|\alpha_0\rangle$ into which the ground steady state goes over when the field is switched-on is equal to

$$\begin{aligned}&\alpha_0=0 &&\text{for}\quad \omega_l<\omega_1,\\ &\alpha_0=n &&\text{for}\quad \omega_n<\omega_l<\omega_{n+1},\\ &\alpha_0=N &&\text{for}\quad \omega_l>\omega_N.\end{aligned} \tag{8}$$

We see that by appropriately choosing the laser frequency ω_l we can make the system go over from the ground steady state $|0\rangle$ into and of the $N+1$ quasistationary states. This possibility, which is of paramount importance to us, is due to the fulfillment of the adiabatic theorem for the quasistationary states.[12,17] As a result of the stepwise variation of the number α_0 as a function of ω_l [see (8)], the population p_n of the steady state $|n\rangle$, regarded as a function of ω_l, will have discontinuities at the points where the laser frequency ω_l coincides with any of the resonance frequencies ω_m, since

$$p_n(\omega_l)=|\langle n|\alpha_0\rangle|^2=|a_n^{\alpha_0}|^2$$

and the coefficients $a_n^{\alpha_0}$ and $a_n^{\alpha_0+1}$ are, generally speaking, different. Below we show that a discontinuity does not occur only in a two-level $(N=1)$ system for which the equality $|a_1^0|^2=|a_1^1|^2$ is accidentally fulfilled at $\omega_l=\omega_1$.

3. WEAK FIELD

Let the maximum $E(t)$ value, equal to E_{max}, be small in the sense that the field broadenings f_n are still small compared to the level spacing in the excited band even when $E(t)=E_{max}$. Then the resonances $|0\rangle\to|n\rangle$ with different n are actually independent, and the function $p_e(\omega_l)$, where

$$p_e(\omega_l)=1-p_0(\omega_l)=\sum_{n=1}^{N}p_n(\omega_l),$$

is a set of narrow peaks corresponding to the excitation of the individual levels $|n\rangle$.

Let us consider one of such peaks, say, the $n=1$ peak. To begin with, we shall completely ignore the effect of the other levels, i.e., we shall consider a two-level $(N=1)$ system. The quasi-energies ε^α and the eigenvectors a_m^α in this case are easily found from Eq. (5):

$$\begin{aligned}\varepsilon^{0,1}&=\frac{\hbar}{2}[-\delta\omega_1\mp(\delta\omega_1^2+4f_1^2)^{1/2}],\\ a_0^{0,1}&=\frac{1}{\sqrt{2}}\left[1\mp\frac{\delta\omega_1}{(\delta\omega_1^2+4f_1^2)^{1/2}}\right]^{1/2}\\ a_1^{0,1}&=\mp\frac{1}{\sqrt{2}}\left[1\pm\frac{\delta\omega_1}{(\delta\omega_1^2+4f_1^2)^{1/2}}\right]^{1/2},\qquad \delta\omega_1=\omega_l-\omega_1.\end{aligned} \tag{9}$$

Here and below the first exponent corresponds to the upper sign. When the system is in the quasistationary states $\alpha=0$ and 1 the populations p_1 of the excited $n=1$ level are respectively

$$p_1^{\alpha=0}=|a_1^0|^2=\frac{1}{2}\left(1+\frac{\delta\omega_1}{(\delta\omega_1^2+4f_1^2)^{1/2}}\right), \tag{10a}$$

$$p_1^{\alpha=1}=|a_1^1|^2=\frac{1}{2}\left(1-\frac{\delta\omega_1}{(\delta\omega_1^2+4f_1^2)^{1/2}}\right). \tag{10b}$$

These curves are shown dashed in the upper part of Fig. 1a. As the field is adiabatically switched on, the system goes over from the ground steady state into the quasistationary state with the number α_0; in this case, according to the numbering rule [see (8)], $\alpha_0=0$ when $\delta\omega_1<0$ and $\alpha_0=1$ when $\delta\omega_1>0$. As a result, we obtain

$$p_e(\omega_l)=p_1(\omega_l)=\frac{1}{2}\left(1-\frac{|\delta\omega_1|}{(\delta\omega_1^2+4f_1^2)^{1/2}}\right). \tag{11}$$

The parts of the curves (10a) and (10b) that make up together the curve (11) are represented in Fig. 1a by the continuous curve. Notice that (11) has a kink (a derivative discontinuity) at the exact-resonance point $\delta\omega_1=0$ (see Fig. 1a). For comparison, we also show in Fig. 1a the time-averaged population p_1^{inst} of the upper level (the dotted curve) as computed in standard fashion in the case when the field is instantaneously switched on:

$$p_1^{inst}=2f_1^2/(\delta\omega_1^2+4f_1^2). \tag{12}$$

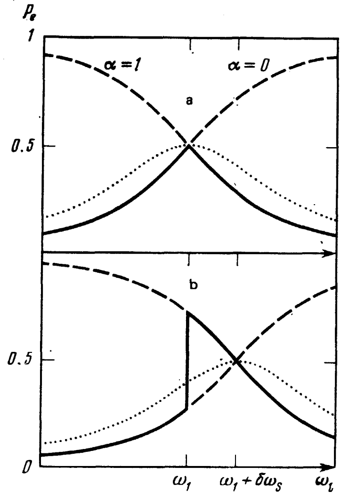

FIG. 1. Two-level system ($p_e = p_1$). The dashed curves show the populations of the quasistationary states $\alpha = 0$ and $\alpha = 1$. In the lower part the dashed curves coincide with the continuous curve. The continuous and dotted curves are respectively plots of the population p_e of the excitable levels vs the field frequency ω_l when the field is switched on adiabatically and instantaneously. The upper figure shows the p_e when all the other levels except the $\alpha = 0$ and $\alpha = 1$ ones are completely neglected. The lower figure shows the p_e when the effect of the other levels is taken into account with the aid of perturbation theory; $\delta\omega_S$ is the Stark resonance-frequency shift due to this effect. The field broadening $f = \delta\omega_S$. For clarity, the discontinuity points in the dependence $p_e(\omega_l)$ in the bottom figure have been joined by a vertical segment.

Notice that even in the case of the two-level system a higher selectivity is achieved when the field is adiabatically switched on than when it is instantaneously switched on, a fact which stems from two characteristics of the dependence (11). First, the curve (11) is narrower than (12): its width (at half-maximum) is smaller by a factor of $\sqrt{3}$; second, the presence of the kink at the point $\delta\omega_1 = 0$ in (11) in a sense also increases the selectivity.

Let us now consider how the $p_e(\omega_l)$ dependence will change when the effect of the other levels is taken into account. Since the external field is weak, all the remaining states of the upper band (except the $|1\rangle$ state) are practically not excited: $p_n \ll 1$, since $f_n \ll \delta\omega_n$, $n = 2, 3, \ldots, N$. It is clear that the shape of the curves (10) will not change in this case. The only difference will be a Stark shift of the transition frequency: instead of ω_1, we should use in (10) the frequency

$$\omega_1{}^S = \omega_1 + \delta\omega_S, \qquad \delta\omega_S = -\frac{E^2}{\hbar^2}\sum_{n=2}^{N}\frac{d_{0n}{}^2}{4\delta\omega_n}.$$

Notice that $\delta\omega_S > 0$, since all the $\delta\omega_n < 0$. The Stark shift will, as shown in Fig. 1b, lead to some general shift of the curves (10). At the same time the number of the quasistationary state into which the system goes over as the field is switched on is given as before by the expression (8). Therefore the transition from one curve to the other, when ω_l is varied, occurs as before at the point where $\omega_l = \omega_1$, and not at the point of intersection of the curves, where $\omega_l = \omega_1 + \delta\omega_S$. Hence we can easily see how the discontinuity at $\omega_l = \omega_1$ appears in the $p_e(\omega_l)$ dependence. We have

$$p_e(\omega_l) = \frac{1}{2}\left\{1 + \frac{\delta\omega_1 - \delta\omega_S}{[(\delta\omega_1 - \delta\omega_S)^2 + 4f_1{}^2]^{1/2}}\right\}, \quad \delta\omega_1 < 0,$$
$$p_e(\omega_l) = \frac{1}{2}\left\{1 - \frac{\delta\omega_1 - \delta\omega_S}{[(\delta\omega_1 - \delta\omega_S)^2 + 4f_1{}^2]^{1/2}}\right\}, \quad \delta\omega_1 > 0, \qquad (13)$$

where $\delta\omega_1 = \omega_l - \omega_1$. The jump

$$\Delta p_e = p_e(\omega_1 + 0) - p_e(\omega_1 - 0)$$

is equal to

$$\Delta p_e = \delta\omega_S/(\delta\omega_S{}^2 + 4f_1{}^2)^{1/2}. \qquad (14)$$

Notice that in a weak field $\Delta p_e \propto E \propto \sqrt{I}$. Discontinuities of this type make it possible for quantum systems to be excited with a very high selectivity (see Ref. 9).

The other qualitative characteristic of the excitation in the case when the field is adiabatically switched on—the occurrence of inversion ($p_1 > p_0$)—can also be seen from (13) and Fig. 1b. The inversion is maximal at the frequency $\omega_l = \omega_1 + 0$, where it is given by the expression (14):

$$p_1(\omega_1 + 0) - p_0(\omega_1 + 0) = \Delta p_e > 0.$$

For the sake of clarity, let us briefly discuss the case in which the upper band consists of two levels (i.e., in which $N = 2$). In this case the secular equation following from (5) is cubic, which allows us to find analytic expressions for ε^α and a_n^α with the aid of the Cardan formula. We do not give these expressions, in view of their unwieldiness. Instead, we show in Fig. 2 the plots of

$$p_e(\omega_l) = p_1(\omega_l) + p_2(\omega_l)$$

constructed with the aid of the exact solution for the case $d_{02} = 4d_{01}$. Since the dipole moments enter into all the formulas only in the combination $d_{0n}E$, the transition to a greater d_{0n} is, in a sense, a transition to a higher field intensity. Therefore the evolution of the individual peaks of $p_e(\omega_l)$ as E_{max} increases is also clear from Fig. 2. If E_{max} is not yet too high, and the criterion

$$f_{n\,max} = E_{max} d_{0n}/\hbar \ll \Delta, \qquad n = 1, 2,$$

(where $\Delta = \omega_2 - \omega_1$ is the band width) for a weak field is fulfilled, then, the jumps Δp_e occurring near the resonance frequencies ω_1 and ω_2 decrease in our case as

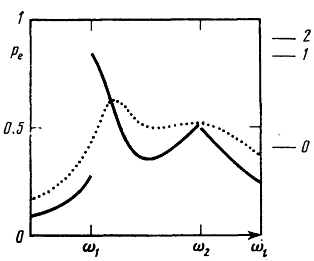

FIG. 2. A three-level system ($p_e = p_1 + p_2$; $N = 2$) in a weak field. The level diagram is shown on the right. The dipole moment $d_{02} = 4d_{01}$. The total field broadening $f^2 = f_1^2 + f_2^2 = \frac{17}{80}\Delta^2$, where $\Delta = \omega_2 - \omega_1$. The notation is the same as in Fig. 1.

E_{max} increases, but then the widths of the corresponding peaks increase (see Fig. 2). Notice that even in a weak field the magnitude Δp_e of the jump near the frequency ω_1 will be close to unity when the field E_{max} is such that $\delta\omega_S \gg f_1$. This condition is consistent with the criterion for a weak field if the dipole moments of the transitions differ greatly from each other, i.e., if $d_{02} \gg d_{01}$.

4. STRONG FIELD

Let us consider the case of a strong field, when

$$f \gg \Delta, \tag{15}$$

where $f^2 = f_1^2 + f_2^2 + \cdots + f_N^2$ and $\Delta = (E_N - E_1)/\hbar$ is the band width. Let us first write out the expressions for the quasi-energies $\varepsilon^{0,N}$ and the eigenvectors $\{a_n^{0,N}\}$ with the minimum ($\alpha = 0$) and maximum ($\alpha = N$) quasi-energies. In the broad frequency range where $|\delta\omega_{1,N}| \ll f^2/\Delta$ we have from Eq. (5) the expressions

$$\varepsilon^{0,N} = \frac{\hbar}{2}[-\delta\omega_{1,N} \mp (\delta\omega_{1,N}^2 + 4f^2)^{1/2}],$$

$$a_0^{0,N} = \frac{1}{\sqrt{2}}\left[1 \mp \frac{\delta\omega_{1,N}}{(\delta\omega_{1,N}^2 + 4f^2)^{1/2}}\right]^{1/2}, \tag{16}$$

$$a_n^{0,N} = \pm\frac{f_n}{f\sqrt{2}}\left[1 \pm \frac{\delta\omega_{1,N}}{(\delta\omega_{1,N}^2 + 4f^2)^{1/2}}\right]^{1/2}, \quad n = 1, 2, \ldots, N;$$

an $(N+1)$-level system possesses, besides these two quasistationary states, $N-1$ other such states with numbers $\alpha = 1, \ldots, N-1$.

In the case of an arbitrary $N \geq 2$, it is impossible to obtain lucid analytic expressions for the $\alpha = 1, 2, \ldots, N-1$ states. Nevertheless, we can prove the following two important assertions in their general form: 1) All these states are, in the case of strong fields, i.e., when the condition (15) is fulfilled, inverse states (in the sense that $p_e \approx 1$, $p_0 \approx 0$); 2) their quasi-energies ε^α ($\alpha = 1, 2, \ldots, N-1$) are close to zero (i.e., $|\varepsilon^\alpha| \ll f$). These assertions are valid in the case of detunings $|\delta\omega_n| \approx \Delta$, particularly in the frequency region $\omega_1 < \omega_l < \omega_N$, where the excitation of the states in question occurs [see (8)]. Indeed, in the case when $|\delta\omega_n| \approx \Delta$, the diagonal terms in H, (4) ($\sim\Delta$), turn out to be small compared to the off-diagonal terms ($\sim f$); therefore, in the first approximation the secular equation that follows from (5) assumes the form

$$\varepsilon^{N-1}(\varepsilon^2 - \hbar^2 f^2) = 0. \tag{17}$$

Equation (17) possesses, besides the two quasi-energies $\varepsilon^{0,N} = \mp\hbar f$ corresponding to the states (16), an $(N-1)$-tuple root $\varepsilon^\alpha = 0$, $\alpha = 1, 2, \ldots, N-1$. In the next order in Δ/f this degeneracy is lifted, and all the quasi-energies turn out to be of the order of Δ. The structure of the corresponding states is clear from (5): the n-th row in (5) has the form

$$f_n a_0^\alpha - \delta\omega_n a_n^\alpha = \varepsilon^\alpha a_n^\alpha.$$

Since

$$\varepsilon^\alpha \sim \delta\omega_n \sim \Delta, \quad a_0^\alpha / a_n^\alpha \sim \Delta/f_n \sim \Delta/f.$$

The criterion (15) for a strong field guarantees the smallness of this ratio. Thus, $a_0^\alpha \ll 1$, which completes the proof of the two assertions about the states with $\alpha = 1, 2, \ldots, N-1$. Below we shall call these states inverse states.

In the case when the upper band consists of two levels (i.e., when $N = 2$), we have a single inverse ($\alpha = 1$) state. Its quasi-energy ε^1 and eigenvector $a^1 = (a_0^1, a_1^1, a_2^1)$ are equal in a fairly broad range of detunings $|\delta\omega_{1,2}| \ll f^2/\Delta$ to

$$\varepsilon^1 = -\frac{\hbar}{2}(\delta\omega_1 + \delta\omega_2) + \frac{\hbar\Delta}{2}\frac{f_1^2 - f_2^2}{f^2},$$

$$a^1 = \left(\Delta\frac{f_1 f_2}{f^3}, \frac{f_2}{f}, \frac{-f_1}{f}\right). \tag{18}$$

According to (8), the system goes over into one of the inverse states if the laser frequency lies in the interval $\omega_1 < \omega_l < \omega_N$. Outside this interval the states (16) with $\alpha = 0$ and N are excited. Thus, the $p_e(\omega_l)$ dependence for an arbitrary $(N+1)$-level level-band system has, in the case of a strong field, a universal shape, which is shown in Fig. 3.

Notice also that it is precisely the proved fact (i.e., that $a_0^\alpha \ll 1, \alpha = 1, 2, \ldots, N-1$) which indicates that inverse states cannot be significantly excited when the field is instantaneously switched on. Indeed, in this case the fraction C^α of a given quasistationary state with number α in the solution is equal to $C^\alpha = (a_0^\alpha)^*$, i.e., $|C^\alpha|^2 \ll 1$. Therefore, no differences will be observed between the two-level ($N = 1$) and multilevel ($N \geq 2$) cases when a strong field satisfying the condition (15) is switched on instantaneously: in the case when the field is switched on instantaneously $p_e(\omega_l)$ always has the shape of the dotted curve in Fig. 3. The spike of the $p_e(\omega_l)$ curve in the region $\omega_1 < \omega_l < \omega_N$, which arises despite the fact that the field broadening "covers" this interval [see (15)], is a clear manifestation of the superselectivity that obtains in the case when the external field is switched on adiabatically. Notice that the jumps Δp_e that occur at the frequencies $\omega_l = \omega_1$ and $\omega_l = \omega_N$ are, in the present case, not small, but close to $\Delta p_e = \frac{1}{2}$. In the frequency range $\omega_1 < \omega_l < \omega_N$ we have a completely empty ground state and, as a result total population inversion: $p_0 \approx 0$, $p_e \approx 1$.

5. TRANSITIONS BETWEEN THE QUASISTATIONARY STATES

Thus far we have considered the case in which the field is switched on infinitely slowly, when only one

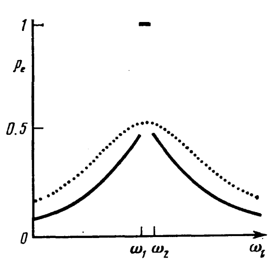

FIG. 3. Multilevel system ($p_e = p_1 + p_2 + \ldots + p_N$; $N \geq 2$) in a strong field. The $p_e(\omega_l)$ plot is for a three-level system ($N = 2$; $d_{01} = d_{02}$) with $f^2 = f_1^2 + f_2^2 = 12.5\Delta^2$. For all $N \geq 2$ systems of the type under consideration the function $p_e(\omega_l)$ has qualitatively the same shape as the function plotted in this figure. The notation is the same as in Figs. 1 and 2.

of the coefficients in the expansion

$$|\psi(t)\rangle=\sum_{\alpha}C^{\alpha}|\alpha\rangle \tag{19}$$

is nonzero: $|C^{\alpha_0}|=1$. In the general case there can occur transitions between the quasistationary states, and the C^{α} can be nonzero for all α. Substituting the expansion (19) into the Schrödinger equation, we obtain the usual system of equations for the coefficients C^{α}:

$$\frac{dC^{\alpha}}{dt}=\sum_{\beta}T^{\alpha\beta}\exp\left\{-\frac{i}{\hbar}\int_{-\infty}^{t}dt'\,(\varepsilon^{\beta}-\varepsilon^{\alpha})\right\}, \tag{20}$$

$$T^{\alpha\beta}=-\sum_{n}a_n^{\alpha*}\frac{da_n^{\beta}}{dt}.$$

To find out whether transitions can occur between the adiabatic states $|\alpha\rangle$ and $|\beta\rangle$, we must compare the characteristic time t_f of variation of $T^{\alpha\beta}$ with the frequency $|\varepsilon^{\alpha}-\varepsilon^{\beta}|/\hbar$. Let the quasi-energies of the system under investigation be such that, for all substantial field intensities

$$|\varepsilon^{\alpha_n}-\varepsilon^{\alpha_0}|\gg\hbar|\delta\omega_n|$$

(we are considering the evolution of the system from the ground state). Thus, the adiabaticity condition has the form

$$t_f\gg|\delta\omega_n|^{-1},\qquad n=1,2,\ldots,N. \tag{21}$$

The time t_f is the characteristic time of variation of $E(t)$ when the field broadening $f_n(t)\approx|\delta\omega_n|$. In the majority of cases, t_f coincides with the characteristic laser-pulse rise time.

Let us note another interesting characteristic of the case in which the field is switched on adiabatically: the Rabi oscillations, which arise upon the violation of the adiabaticity condition (21) for one or several (but not all!) transitions, can attenuate as a result of the increase of the field intensity even in the absence of all the relaxation processes.

It can be shown that this property is characteristic of all multilevel systems of the type in question if $N\geq 3$. Let us illustrate this property in the particular case of a four-level ($N=3$) system, for which $\delta\omega_2=\omega_l-\omega_2=0$, and, thus, the adiabaticity condition (21) for the $0\to 2$ transition is clearly not fulfilled. But if the adiabaticity condition (21) is fulfilled for the remaining transitions $0\to 1$ and $0\to 3$, it follows from the system (20) that $C^0=C^3=0$, $C^1=C^2=1/\sqrt{2}$. In the case

$$f_1=f_2=f_3=f/\sqrt{3}\text{ and }E_2-E_1=E_3-E_2=\Delta/2$$

we obtain from (5) and (6) the quasistationary states $|\alpha=1\rangle$ and $|\alpha=2\rangle$ and their quasi-energies ε^1 and ε^2, with $\varepsilon^1=-\varepsilon^2$. Substituting these quantities into (19), we find $|\psi(t)\rangle$, whence we find

$$p_e(t)=1-2(a_0^{2})^2\cos^2\left(\int_{-\infty}^{t}dt'\varepsilon^2(t')\right); \tag{22}$$

$$\varepsilon^2=\sqrt{\frac{2}{3}}f\left[1+\frac{4f^2}{\Delta^2}+\left(1+\frac{8f^2}{3\Delta^2}+\frac{16f^4}{\Delta^4}\right)^{1/2}\right]^{-1/2},$$

$$a_0^{2}=\left[1+\frac{f^2}{3(\varepsilon^2)^2}+\frac{8f^2[4(\varepsilon^2)^2+\Delta^2]}{3[4(\varepsilon^2)^2-\Delta^2]^2}\right]^{-1/2},$$

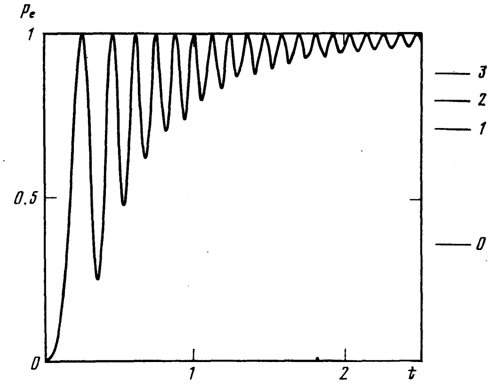

FIG. 4. The time dependence of p_e for the four-level system depicted on the right. The dipole moments $d_{01}=d_{02}=d_{03}=d$. The external-field frequency $\omega_l=\omega_2$. The field amplitude $E(t)$ increases linearly in time. Plotted along the t axis is the ratio $E(t)d/\hbar\Delta$. This parameter attains the value 1 over a period of time equal to t_f, which has the meaning of the characteristic switching-on time of the field. The plot is for the case in which $t_f=100\ \Delta^{-1}=50|\delta\omega_1|^{-1}=50|\delta\omega_3|^{-1}$; the detuning $\delta\omega_2=0$ [see the adiabaticity condition (21)].

and $f=f(t)$. The $p_e(t)$ oscillation amplitude, which is equal to $(a_0^2)^2$, tends to zero when $f(t)$ rises above Δ. The curve $p_e(t)$ is shown in Fig. 4. Let us emphasize again the difference between the present effect and the usual (relaxational) attenuation of the Rabi oscillations. In the latter case the oscillations usually decay to some level $\bar{p}<1$; for a two-level system $\bar{p}=\frac{1}{2}$. The decay of the Rabi oscillations to the level $\bar{p}=1$ (see Fig. 4) on account of relaxation or as a result of an inhomogeneous broadening is impossible.

6. DISCUSSION

Since in real experiments the time $t_f<\infty$, all the discontinuities in the function $p_e(\omega_l)$ disappear. This, however, does not imply the disappearance of the superselectivity, since the very strong frequency dependence of $p_e(\omega_l)$ in the vicinity of the transition frequencies ω_n is preserved. Indeed, according to (21), the adiabaticity condition is violated only in the case of detunings $|\delta\omega_n|$ smaller than t_f^{-1}. Therefore, instead of discontinuities at the frequencies ω_n, we obtain smooth curves with transition-region widths of the order of t_f^{-1}. The derivative $dp_e/d\omega_l$, which determines the spectral selectivity, is of the order of t_f at $\omega_l=\omega_n$. It can be made arbitrarily large by increasing t_f.

Let us also emphasize that the maximum selectivity equal to $dp_e/d\omega_l\approx t_f$ in the case when the field is switched on adiabatically, does not depend on the radiation intensity I. This differs qualitatively from the excitation under conditions when the field is instantaneously switched on, in which case the selectivity is bounded by the magnitude of the field broadening f; it inevitably decreases with increasing intensity I, since $f\propto I^{1/2}$.

As applied to the optical region, the appearance of inversion in the level population was first noted in Refs.

1 and 2; the population jumps and other characteristics of the system as functions of the frequency were first considered[3)] in Ref. 5. In Refs. 9 and 14 and in the present paper it is shown, in particular, that the features found in Refs. 1–8 are characteristic not only for the systems considered there, but have a universal character, and can be observed in a broad range of frequencies. Furthermore, in the present paper we have found and investigated another (i.e., a third) characteristic that occurs in the case when the field is switched on adiabatically: the coherent attenuation of the Rabi oscillations in a system with four or more levels (see Sec. 5). This attenuation can be readily investigated in experiment.

For typical pulsed lasers the adiabaticity condition (21) is usually fulfilled very easily. As an example, let us consider the 6^1S_0–6^3P_1 transition ($\lambda = 7911$ Å) in a Ba atom located in a $B = 2$ kG magnetic field. The Zeeman splitting of the upper 6^3P_1 state attains in this case a value of the order of $\Delta \approx 0.3$ cm^{-1}. To observe significant jumps Δp_e, we must have $f \approx \Delta$, a value which is attained even in fields of intensity $I \approx 200$ kW/cm^2. The adiabaticity condition $t_f \gg \Delta^{-1} \approx 0.1$ nsec is fulfilled for pulses in the nanosecond range. The lifetime of the upper 6^3P_1 state is of the order of 300 nsec, which is sufficient for observation.

Observation of the superselectivity of the adiabatic excitation and the resulting inversion is, naturally, possible for other systems. To observe them, it is only necessary for the radiation to induce at least two resonance transitions from some level. This condition is easily satisfied when the band is made up of fine- or hyperfine-structure components in atoms, or of rotational-vibrational levels in molecules.

All the considered effects are coherent in the sense that they were computed without allowance for the relaxation processes. Therefore, in order for them to be observed, it is necessary that the excitation time be short compared to the phase-relaxation time T_2', as well as to the radiation-coherence time $\Delta\omega_l^{-1}$. As in the case of the observation of other coherence effects (such as self-induced transparency, photon echo, etc.[18]), these conditions can be satisfied by working with gases at low pressures ($p \lesssim 1$ Torr), and with molecular or atomic beams.

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[1)]We use the term "superselectivity," since the critical dependence of p_e on ω_l allows quantum systems to be excited with a significantly higher selectivity than in the case when a field of the same intensity is switched on instantaneously. (See in this connection Ref. 9.)

[2)]The problem of the classification of the quasistationary states and the corresponding numbering of their quasi-energies is not quite trivial. Another numbering scheme is usually used (see, for example, Refs. 1, 4, and 14): the number of quasistationary state coincides with a number of that steady state that the quasistationary state goes over into when the field is switched on. But this scheme has a serious disadvantage: As the frequency is varied, the quasi-energies ε^α go through discontinuities, to which no measurements in the state of the system correspond (see, for example, Ref. 15). By using the numbering rule (7), we assume that the set of levels does not possess any hidden symmetry that makes the intersection of the quasiterms possible (see in this connection Ref. 16).

[3)]Also considered in Ref. 5, besides the jumps, is the appearance of inversion, this phenomenon being called there "self-induced adiabatic inversion."

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