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Two-photon excitation of a quantum system

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To analyze the crossing of the quasienergy levels of a system interacting with an intense alternating field under conditions of two-photon resonance, we propose an exactly solvable model of a field whose envelope is of the characteristic interaction switching-on type. The kinetics of the system in the field is analyzed. From the obtained general relation for the probability of two-photon excitation there follow as limits instantaneous switching-on of the field and the adiabatic limit (the Landau-Zener formulas). The dependence of the excitation probability on the field intensity and the detuning of the two-photon resonance is analyzed for different interaction switching-on regimes and with allowance for possible ionization of the system from the upper state.

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1. INTRODUCTION. FORMULATION OF THE PROBLEM

Two-photon excitation is one of the first experimentally observed effects of nonlinear optics.[1] A large number of studies have now been made in which two-photon excitation has been observed in both condensed media and in gases. The theoretical description of the probability of two-photon excitation of state 2 from state 1 is usually based on the Weisskopf–Wigner formula, which also describes single-photon excitation[2]:

$$W=|V_{12}|^2\frac{\Gamma}{(E_1-E_2+2\omega)^2+\Gamma^2/4}. \tag{1}$$

Here, V_{12} is the matrix element of the two-photon transition, E_1 and E_2 are the energies of the levels, Γ is the homogeneous line width, $\hbar=1$, and absence of saturation is also assumed: $|V_{12}|\ll|E_2-E_1-2\omega+i\Gamma/2|$. In the presence of inhomogeneous broadening, expression (1) must be appropriately averaged.

In the absence of intermediate single-photon resonance, which will be assumed in what follows, the matrix element V_{12} depends linearly on the radiation intensity I. The energy levels $E_{1,2}$ also depend linearly on the intensity because of the quadratic dynamical Stark effect.[1)] This fact can be taken into account by setting in formula (1)

$$E_{1,2}=E_{1,2}^{(0)}-\alpha_{1,2}I/4, \tag{2}$$

where $E_{1,2}^{(0)}$ are the energy levels in the absence of radiation, and $\alpha_{1,2}$ are the polarizabilities of the levels at the field frequency ω.

Equations (1) and (2) can be proved by means of Low's equations, which describe the natural width of atomic levels if one takes into account the contribution to the mass operator of not only the photon vacuum but also the field of the laser radiation.[4] For this, however, it is necessary to assume that the electromagnetic field is stationary, for otherwise the mass operator, which is a function of two four-points, becomes dependent on t and t' separately and not merely on the difference $t-t'$, and as a result Eq. (1) cannot be proved.

Since there cannot be strictly stationary laser fields (if only because of the existence of the switching-on period), formula (1) is by no means always valid. Indeed, in recent studies[5-8] it was shown that in a number of cases two-photon excitation bears a greater similarity to the transitions between molecular terms in slow collisions of atoms or adiabatic spin inversion in magnetic resonance than to the resonant absorption of a single photon.

Figure 1 explains the physical situation. Suppose, for simplicity, that the time dependence of the radiation intensity is due solely to the switching-on of the field. Then the energy levels $E_{1,2}(t)$ vary from $E_{1,2}^0$ to certain stationary values determined by the steady-state field intensity, as shown in Fig. 1. At definite values of the detuning from resonance in the absence of radiation, of the difference between the level polarizabilities, and of the intensity in the steady state, it is possible for the levels $E_1+2\omega$ and E_2 to cross at a certain time t_0. As is well known,[9] the presence of even weak interaction between states 1 and 2 leads to quasi-

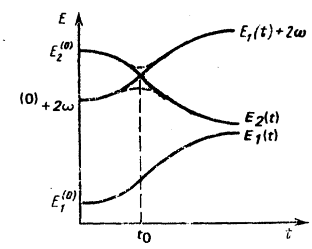

FIG. 1.

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crossing of the energy levels, as is shown in Fig. 1 by the dashed curves. In the given case, V_{12} is such an interaction. If at the same time the motion through the terms occurs sufficiently slowly, then a system in state 1 prior to the switching-on of the field will be in state 2 at $t > t_0$, i.e., the excitation probability is equal to unity. Thus, the possibility of crossing of the energy (or rather quasienergy) levels leads to results that differ strongly from those obtained in accordance with Eq. (1).

The quantitative theory of the phenomenon must make it possible to calculate the probability of adiabatic transitions between states as a function of the rate of change of the radiation intensity and the other parameters of the problem. In a number of studies,[6-8] the Landau-Zener model,[9] which is widely used in the theory of atomic collisions, was used to calculate the probabilities. In this model, it is assumed that in the region of crossing the terms can be approximated by a linear function of the time and—which is more important in the given case—the probability of an adiabatic transition is determined by the immediate neighborhood of the point of crossing. However, it is readily seen that this last condition is not satisfied in the problem of two-photon excitation. Indeed, after the crossing of the terms, the distance between them increases in proportion to the intensity, but the interaction V_{12} also increases in accordance with such a law. Therefore, if there are no special circumstances leading to anomalously small matrix elements of V_{12}, the values of $(\alpha_1 - \alpha_2)I$ and V_{12} are of the same order. In the framework of the adiabatic approximation, this situation was considered in Ref. 5 (see also Ref. 10).

It is clear that the solution of the problem when there are in fact no literal small parameters is possible to describe a particular form of the laser pulse by constructing for the time dependence of the intensity an exactly solvable model containing a sufficient number of parameters. In the present paper, we propose a general method for constructing such models and consider in detail the case of a change in intensity of the "field switching-on" type.

2. GENERAL RELATIONS

In what follows, we shall for brevity call the quantum system an "atom." In the alternating field, the amplitudes of the probabilities for finding the atom in the states 1 and 2, between which there is resonance at the frequency 2ω, satisfy the equations[11]

$$\begin{aligned} \dot{a}_1 &= {}^1/_4 i[\alpha_1 I(t) a_1 + vI(t) e^{-i\delta t} a_2], \\ \dot{a}_2 &= {}^1/_4 i[\alpha_2 I(t) a_2 + vI(t) e^{i\delta t} a_1]. \end{aligned} \tag{3}$$

Here, $I(t)$ is the envelope of the intensity, $\delta = E_2^{(0)} - E_1^{(0)} - 2\omega$ is the initial detuning from resonance corresponding to $I \to 0$, and $vI/4 = V_{12}$ is the excitation matrix element.

Explicit expressions for $\alpha_{1,2}$ and v can be readily obtained by the usual methods of perturbation theory. For example, in the dipole approximation in the case of linear polarization of the field along the z axis,

$$\begin{aligned} \alpha_k &= \sum_n |\langle k|d_z|n\rangle|^2 \left(\frac{1}{\omega_{nk}+\omega} + \frac{1}{\omega_{nk}-\omega}\right), \quad k=1,2, \\ v &= \sum_n \frac{\langle 2|d_z|n\rangle\langle n|d_z|1\rangle}{\omega_{n1}-\omega}, \end{aligned} \tag{4}$$

where d is the operator of the dipole moment, $\omega_{nk} = E_n^{(0)} - E_k^{(0)}$, and the summation in (4) is over the complete spectrum of the atom.

By choosing the phases of the wave functions, the matrix element v can be made real, as is in fact assumed in (3). Equations (3) are obtained under the condition that the field is quasimonochromatic, $|\dot{I}/I| \ll \omega$, which is usually the case.

Since spontaneous decay of the atom from the states 1 and 2 is possible, as well as ionization from these states under the influence of another stationary field, δ is complex: $\mathrm{Im}\,\delta = (\Gamma_1 - \Gamma_2)/2$, where $\Gamma_{1,2}$ are the reciprocal lifetimes of the levels; but if single-photon ionization from state 2 by a field of frequency ω is possible, then α_2 is also complex.

Equations (3) are formally analogous to the equations of the theory of slow collisions,[12] in which the part of the distance between the terms is played by the detuning $\Delta(t) = \delta - (\alpha_2 - \alpha_1)I/2$, and $vI/4$ is the interaction between them, or to the equations that describe the interaction of a resonant field of variable amplitude and frequency with a two-level system in single-photon resonance.[13] In our case, the detuning and interaction vary in time in accordance with the same law.

At $\delta = 0$, Eqs. (3) can be solved for any function $I(t)$. For the rate of excitation, we obtain in this case

$$w = \frac{d}{dt}|a_2(t)|^2 = \frac{2v^2}{(\alpha_1-\alpha_2)^2+4v^2} I(t) \int_{-\infty}^{t} I(t')\,dt'. \tag{5}$$

However, this expression cannot be directly compared with the expression (1), since the derivation of (1) presupposes that the width of the radiation spectrum is small compared with the width of the atomic line. The expression (5) is derived under the opposite assumption, $\delta = 0$. Therefore, to find the limits of applicability of the expression (1), and also to analyze the case of level crossing it is necessary to consider nonzero initial detunings.

In Eqs. (3), we replace the time by the new independent variable

$$x = \frac{1}{I_0} \int_{-\infty}^{t} I(t')\,dt'. \tag{6}$$

Here, I_0 is some characteristic intensity which ensures that x has the dimensions of time. The value of x is proportional to the area of the intensity. When t varies in the interval $(-\infty, +\infty)$, x varies in the interval $(0, x_0)$, and $x_0 \to \infty$, if the field is not switched off at large t; if the field is pulsed, then x_0 is bounded. The transformation which is the inverse of (6),

$$t(x) = I_0 \int_0^x \frac{dx}{I(x)} \tag{7}$$

is a transcendental equation for the function $x(t)$.

Substituting (6) in (3), we obtain

$$\frac{da_1}{dx}=\frac{i}{4}\{\alpha_1 I_0 a_1+vI_0 a_2 e^{-i\delta t(x)}\},$$
$$\frac{da_2}{dx}=\frac{i}{4}\{\alpha_2 I_0 a_2+vI_0 a_1 e^{i\delta t(x)}\}. \tag{8}$$

The initial conditions for this system have the form

$$a_1(0)=1,\ a_2(0)=0. \tag{9}$$

Expressing a_1 in the second of Eqs. (8) in terms of a_2 and substituting the result in the first equation, we obtain

$$I(x)\frac{d^2a_2}{dx^2}-iI_0\left[\delta+\frac{1}{4}(\alpha_1+\alpha_2)I(x)\right]\frac{da_2}{dx}$$
$$+I_0^2\left[\frac{1}{16}(v^2-\alpha_1\alpha_2)I(x)-\frac{1}{4}\delta\alpha_2\right]a_2=0. \tag{10}$$

It is readily seen that a behavior $I(t)$ of the interaction switching-on type

$$I(t)=\begin{cases}0, & t\to-\infty\\ I_0, & t\to+\infty\end{cases} \tag{11}$$

corresponds to the limits

$$I(x)=\begin{cases}0, & x=0\\ I_0, & x\to+\infty.\end{cases} \tag{12}$$

A simple case of such a dependence that ensures analytic solution of Eq. (10) is given by the function

$$I(x)=I_0x/(x+\tau), \tag{13}$$

which makes Eq. (10) hypergeometric:

$$\frac{d^2a_2}{dx^2}-i\left[\delta+\frac{\delta\tau}{x}+\frac{1}{4}(\alpha_1+\alpha_2)I_0\right]\frac{da_2}{dx}+\frac{1}{4}I_0\left[\frac{1}{4}(v^2-\alpha_1\alpha_2)I_0\right.$$
$$\left.-\delta\alpha_2-\frac{\alpha_2\delta\tau}{x}\right]a_2=0. \tag{14}$$

The parameters I_0 and τ in (13) determine the nature of the growth of the field: I_0 is the limiting value of the intensity, and τ is the switching-on time. Note that the solution of Eq. (10) can also be expressed in terms of hypergeometric functions in the more general case when

$$I(x)=(ax+b)/(cx+d).$$

The case $a=c=0$ or $b=d=0$ corresponds to a monochromatic field. For $c=0, d=1$ we obtain Nikitin's model[12]:

$$I(t)=b\exp(at/I_0).$$

Using (7), we can readily find the form of $I(t)$ corresponding to Eq. (13). For this, we express x in (13) in terms of I:

$$x=I\tau/(I_0-I)$$

and using (5) we obtain a differential equation for $I(t)$:

$$\dot{I}I_0\tau(I_0-I)^{-2}=I/I_0.$$

The solution of this equation gives implicity the dependence $I(t)$:

$$t=\tau[(1-I/I_0)^{-1}-\ln(I_0/I-1)]. \tag{15}$$

This solution is determined up to a constant term that determines the instant of switching-on of the interaction in the limit $\tau\to 0$. In (15), this instant is taken at $t=0$.

The function $I(t)$ for different values of the parameter τ is shown in Fig. 2. It can be seen that in the limit $\tau\to 0$ the $I(t)$ dependence becomes ever more pronounced in the neighborhood of the point $t=0$. The limit $\tau\to 0$ corresponds to sudden switching-on of the field, and $\tau\to\infty$ to adiabatic switching-on.

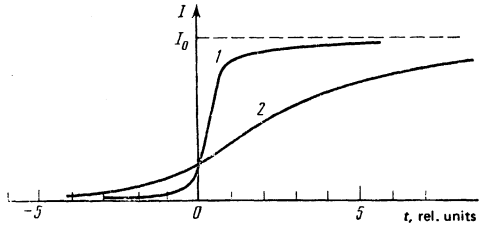

FIG. 2. Dependence $I(t)$ for different values of the parameter τ: 1) $\tau=0.1$, 2) $\tau=1$.

In what follows, we also need the $x(t)$ dependence, which can be determined from (5):

$$\frac{dx}{dt}=\frac{I(x)}{I_0}=\frac{x}{x+\tau},\quad t=x+\tau\ln\frac{x}{\tau}. \tag{16}$$

The solution of Eq. (14) with the initial conditions (9) can be written in the form

$$a_1(x)=e^{i\varepsilon x}\Phi[{}^1/{}_2 i\xi(1+\Delta/\Omega),\ i\xi,\ i\Omega x],$$
$$a_2(x)=\frac{i}{1+i\xi}\left(\frac{v\tau I_0}{4}\right)\left(\frac{x}{\tau}\right)^{1+i\xi}e^{i(\varepsilon+\delta)x}$$
$$\times\Phi\left[1+\frac{1}{2}i\xi\left(1+\frac{\Delta}{\Omega}\right),2+i\xi,i\Omega x\right], \tag{17}$$
$$\Delta=\delta+(\alpha_1-\alpha_2)I_0/4,\ \ \xi=\delta\tau,\ \ \varepsilon=-\delta+(\Delta-\Omega)/2+\alpha_2I_0/4,$$
$$\Omega=[\Delta^2+(vI_0)^2/4]^{1/2}.$$

Here Φ is the confluent hypergeometric function,[14] the parameter ξ plays the part of Massey's parameter in the theory of atomic collisions, Δ is the distance between the adiabatic terms after establishment of the stationary value of the field, and the $x(t)$ dependence is determined by (16). Thus, the parameters of the problem are the dimensionless interaction switching-on time ξ, the detuning Δ, and the Rabi frequency Ω after the field has been switched on.

3. KINETICS OF THE SYSTEM IN THE FIELD

Using Eq. (17), we now consider some features of the behavior of the level populations $n_{1,2}(t)$ of the system and also the total excitation probability:

$$n_{1,2}(t)=|a_{1,2}(t)|^2,\quad W=\lim_{t\to\infty}n_2(t). \tag{18}$$

The results of numerical calculations in accordance with the relations (17) and (18) are given in Fig. 3 for different values of the parameters of the problem corresponding to different field switching-on regimes. At small values of ξ, the time dependence of n_2 corresponds to Rabi oscillations of the populations, and the mean value $\langle n_2(t)\rangle$ cannot exceed $\frac{1}{2}$, i.e., inversion is impossible. At larger values of ξ, as can be seen in the figure, the amplitude of the oscillation decreases, and the mean value $\langle n_2(t)\rangle$ can become greater than $\frac{1}{2}$ because of the possibility, noted in the Introduction, of adiabatic inversion when the levels cross. This is confirmed by the different values of the population of level 2 for different signs of Δ: crossing occurs for $\Delta<0$ but not for $\Delta>0$, since the sign of δ is chosen to be

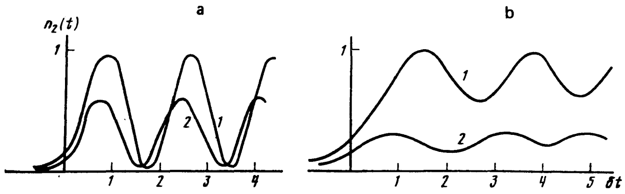

FIG. 3. Time dependence of the population of the upper level for different interaction switching-on regimes when $v/|\alpha_2-\alpha_1|\lambda=0.1$: a) $\pi\xi=1$, b) $\pi\xi=10$. In both figures, curve 1 corresponds to $\Delta<0$ and curve 2 to $\Delta>0$.

positive.

It is obvious that these features in the behavior of the system arise if the Stark shift of the levels can compensate for the initial detuning: $(\alpha_2-\alpha_1)I_0/4 \gtrsim \delta$. The quantity

$$I_c=|4\delta/(\alpha_2-\alpha_1)| \tag{19}$$

is, thus, the threshold intensity for excitation of the system. For example, for atoms of alkali metals in the optical range of frequencies the polarizabilities of the lower levels have a value ~10^3 atomic units.[11] In this case, the threshold intensity of the electric field of the wave corresponding to I_c is ~10^5 V/cm for δ ~0.1 cm^{-1}. This order of magnitude of δ is characteristic of the Doppler frequency detuning of the atoms of a gas at temperature ~10^3 °K.

4. THE LIMITS OF INSTANTANEOUS AND ADIABATIC SWITCHING-ON

These limiting cases can be obtained from the general relations (17). For simplicity, we assume here, as in the previous section, that δ and Δ are real. In the limit $\xi\to 0$, which corresponds to instaneous switching-on of the interaction, we use the equation

$$\lim_{s\to 0}\Phi(as, cs, z)=(1+a/c)(e^z-1),$$

which can be readily obtained from the expansion of the confluent hypergeometric function in a power series.[14] Noting also that in the limit $\tau\to 0$ the relation $x(t)\sim t$ follows from (16), we obtain

$$\begin{gathered} a_1(t)=\exp\{{}^1/_2 i[-\delta+{}^1/_4(\alpha_1+\alpha_2)I_0]t\} \\ \times[\cos(\Omega t/2)+i(\Delta/\Omega)\sin(\Omega t/2)], \\ a_2(t)=i(vI_0/2\Omega)\exp\{{}^1/_2 i[\delta+{}^1/_4(\alpha_1+\alpha_2)I_0]t\}\sin(\Omega t/2), \\ \langle n_{1,2}(t)\rangle=(1\pm\Delta^2/\Omega^2)/2. \end{gathered} \tag{20}$$

The dependence of $\langle n_2\rangle$ on I_0 is shown in Fig. 4 by the dashed curve. The nonmonotonic dependence of the population of the upper level on the intensity is due to the possibility of tuning the levels to resonance for corresponding parameters of the problem.[2)] The maximum on the curve at the point $I_0=I_c$ will be sharper the more important the resonance for the excitation of the system, i.e., the smaller the nondiagonal element v of the interaction compared with the diagonal $\alpha_2-\alpha_1$. In the opposite case $\alpha_1=\alpha_2$, $\delta=\Delta$ and the dependence of $\langle n_2\rangle$ on I_0 determines the usual monotonic saturation curve.[16]

If the interaction switching-on time τ is appreciably longer than the other characteristic times of the problem, the amplitudes $a_{1,2}(t)$ go over into the well-known expressions for a monochromatic field corresponding

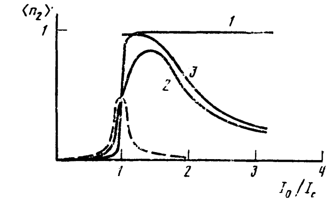

FIG. 4. Dependence of the population of the upper level on the field intensity for $v/|\alpha_2-\alpha_1|=0$ and 1. The dashed curve is the dependence corresponding to instantaneous switching-on of the interaction. Curve 1 is the result of calculation in accordance with Eq. (22 (adiabatic limit), curve 2 corresponds to the exact expression (24), and curve 3 to the Landau-Zener approximation (27). For curves 2 and 3, $\xi=10$, $\xi v^2/(\alpha_2-\alpha_1)^2=0.1$. The result of the calculation in accordance with the expression for $\xi=500$, $\xi v^2/(\alpha_2-\alpha_2)^2=5$ agrees with the result of the adiabatic limit (curve 1) and is approximated well by the step function (30).

to adiabatic switching-on of the interaction.[5,10] The wave function of the atom describes in this case a state with definite quasienergy:

$$\begin{gathered} \Psi(t)=a_1(t)|1\rangle+a_2(t)|2\rangle \\ =e^{-i\mathscr{E}t}\left\{\frac{vI}{4\Omega}\left[\frac{1}{2}\left(1-\frac{\Delta}{\Omega}\right)\right]^{-1/2}|1\rangle-\left[\frac{1}{2}\left(1-\frac{\Delta}{\Omega}\right)\right]^{1/2}e^{-i2\omega t}|2\rangle\right\}, \\ \mathscr{E}=E_1^{(0)}+{}^1/_4\alpha_1 I(t)+{}^1/_2(\Delta-\Omega), \end{gathered} \tag{21}$$

where $\mathscr{E}$ is the quasienergy, which depends adiabatically on the time, and Δ and Ω are determined by Eqs. (17), in which it is necessary to replace I_0 by $I(t)$. For $\delta<0$, it is necessary to reverse the sign of Ω in (21).

The populations of the upper and lower levels change adiabatically in time in this case in accordance with the expressions

$$\langle n_{1,2}(t)\rangle=(1\pm\Delta/\Omega)/2. \tag{22}$$

The dependence of $\langle n_2\rangle$ on the intensity determined by Eq. (22) is now different from the case of instantaneous switching-on, and is shown by curve 1 in Fig. 4. The threshold nature of the dependence of $\langle n_2\rangle$ on I_0 is due to the possibility of crossing of the quasienergy levels. It corresponds to the possibility of complete population of state 2 when the field is switched on and is analogous to spin inversion in the case of a slow passage through resonance.[17] As in the case of instantaneous switch-on of the interaction, the curve is steeper the smaller v.

5. ASYMPTOTIC POPULATION OF THE LEVELS

We now consider the total probability of excitation of the system without any assumption about the value of the parameter ξ. This can be done using the asymptotic behavior of the confluent hypergeometric function,[14] since as $t\to\infty$ and $x\to\infty$

$$\begin{gathered} \lim_{t\to\infty}|a_2(t)|^2=\frac{1}{2}+\frac{\Delta}{2\Omega}\frac{\exp(-\pi\xi\Delta/\Omega)-\operatorname{ch}\pi\xi}{\operatorname{sh}\pi\xi} \\ -\operatorname{Re}\left\{A\exp\left[i\left(\Omega x+\frac{\Delta}{\Omega}\xi\ln(x/\tau)\right)\right]\right\}, \\ A=\frac{2\pi}{\xi\operatorname{sh}\pi\xi}\frac{\exp(-\pi\xi\Delta/2\Omega)(\Omega\tau)^{i\Delta\xi/\Omega}}{\Gamma[i\xi(1+\Delta/\Omega)/2]\Gamma[i\xi(1-\Delta/\Omega)/2]}. \end{gathered} \tag{23}$$

It can be seen that the population is a sum of two terms: one constant in time and another that oscillates with a frequency equal up to a logarithmic term to the Rabi frequency. The amplitude of the oscillating term A is rapidly damped with increasing ξ. Averaging over these oscillations, we obtain finally

$$\langle W\rangle=\lim_{t\to\infty}\langle n_2(t)\rangle=\frac{1}{2}+\frac{\Delta}{2\Omega}\frac{\exp(-\pi\xi\Delta/\Omega)-\operatorname{ch}\pi\xi}{\operatorname{sh}\pi\xi}. \qquad (24)$$

This formula gives the exact value (in the framework of the chosen model) of the probability of two-photon excitation. Let us consider its limiting cases, assuming, as before, $\delta > 0$.

In the case of instantaneous switching on of the interaction, $\xi \to 0$, we obtain

$$\langle W\rangle=(1-\Delta^2/\Omega^2)/2,$$

which agrees with formula (20).

In the case of slow switching-on, $\xi\to\infty$, retaining in (24) only the increasing exponentials, we obtain

$$\langle W\rangle=\frac{1}{2}\left\{1-\frac{\Delta}{\Omega}\left[1-2\exp\left(-\pi\xi\frac{1+\Delta}{\Omega}\right)\right]\right\}. \qquad (25)$$

The exponential retained in (25) can take arbitrary values irrespective of the value of ξ. The expression (25) thus gives the probability of excitation of the upper level in the case of slow switching-on of the field. Comparing (25) with (22), we see that the latter holds only when

$$\xi(1+\Delta/\Omega)\gg 1, \qquad (26)$$

which signifies either the absence of level crossing or that vI_0 is not small compared with $|\Delta|$.

But if the inequality (26) is not satisfied, i.e., $\Delta \approx -\Omega$, which can occur in the case of level crossing and, in addition, for small value of vI_0/Δ, then

$$\langle W\rangle=1-\exp[-2\pi\xi(vI_0/4\Delta)^2]. \qquad (27)$$

Equation (27) corresponds to the Landau–Zener approximation for the given problem. Thus, the Landau–Zener approximation is valid in the region of parameters values determined by the inequalities (sign Δ = −sign δ)

$$\xi\gg 1, \quad \xi(vI_0/\Delta)^2\lesssim 1. \qquad (28)$$

Let us consider in more detail the dependence of the excitation probability on the field intensity in the case of smooth switching-on, $\xi\gg 1$. The regions of I_0 values in which the different approximations are valid are determined by the ratio $v/|\alpha_2-\alpha_1|$.

Suppose $v\ll|\alpha_2-\alpha_1|$. Then in the region $I_0<I_c$ the inequality (26) is satisfied; therefore, the relation (22) holds. With further increase in the field, the parameter vI_0/Δ decreases rapidly, and if $\xi v^2\lesssim(\alpha_2-\alpha_1)^2$, then the probability of excitation is determined by Eq. (27). The limiting value of the population of the upper level for $I_0\gg I_c$ is

$$\langle W\rangle\approx 1-\exp[-2\pi\xi v^2/(\alpha_2-\alpha_1)^2] \qquad (29)$$

and does not depend on I_0.

But if $\xi v^2\gg(\alpha_2-\alpha_1)^2$, then the excitation probability is determined by (22), and since $v\ll|\alpha_2-\alpha_1|$, $\langle W\rangle$ is a step function in I_0:

$$\langle W(I_0)\rangle=\begin{cases}0, & I_0<I_c-\Delta I\\ 1, & I_0>I_c+\Delta I\end{cases} \qquad (30)$$

where ΔI is small compared with I_c.

We estimate ΔI:

$$\Delta I\sim|n_2(I_0)\Big/\frac{\partial n_2}{\partial I_0}\Big|_{I_0=I_c}\sim\frac{vI_c}{|\alpha_2-\alpha_1|}\sim\frac{v\delta}{(\alpha_2-\alpha_1)^2}\quad \frac{\Delta I}{I_c}\sim\frac{v}{|\alpha_2-\alpha_1|}.$$

Thus, if the conditions

$$\xi\gg 1,\quad \xi v^2/(\alpha_2-\alpha_1)^2\gg 1,\quad v\ll|\alpha_2-\alpha_1|,$$

are simultaneously satisfied, ΔI can be omitted in (30) and $\langle W(I_0)\rangle$ can be assumed to be a step function.

Now suppose $v\gtrsim|\alpha_2-\alpha_1|$. Then the inequality (26) is satisfied; therefore Eq. (22) holds for all I_0. However, the approximation (30) for the excitation probability is now invalid. Graphs of $\langle W(I_0)\rangle$ corresponding to the above cases are shown in Fig. 4. Curves 2 and 3 describe the excitation for $\xi v^2<(\alpha_2-\alpha_1)^2$. It can be seen that for $I_0>I_c$ the Landau–Zener approximation (curve 3) is virtually identical with the exact result (curve 2). The result of calculations in accordance with the exact formula for $\xi v^2>(\alpha_2-\alpha_1)^2$ is close to the adiabatic approximation and is well described by a step function.

We now consider the dispersion dependence of the excitation probability. This dependence is determined by the expression (24), in which the independent variable is the dimensionless detuning ξ. As the independent variable, it takes values from $-\infty$ to $+\infty$ and, therefore, for the same switching-on time τ there are corresponding field switching-on regimes for the different regions of δ values.

It is obvious that the singularities in the dispersion dependence associated with the crossing of the levels are in the region of δ values where δ and Δ have opposite signs. In accordance with the previously adopted notation, $\Delta=\delta+\delta_s$, where $\delta_s=(\alpha_1-\alpha_2)I_0/4$ is the resulting Stark shift ($\sigma_s<0$). Then the region of crossing is $0\leqslant\delta\leqslant|\delta_s|$.

We consider slow switching-on $\xi_s=|\delta_s|\tau\gg 1$. Then there is a region of δ values such that $\delta\tau\gg 1$, namely, $\tau^{-1}\ll\delta\leqslant|\delta_s|$, for which the expression (25) is valid. If in addition the inequality (26) is satisfied in this region, then the formula of the adiabatic approximation (24) is valid for the excitation probability. The graph of this function is shown in Fig. 5a by the dashed curve. It can be seen that the maximal population holds in the limit $\delta\to 0$. Curves 2 and 1 of this figure, which have a similar nature to the one mentioned above, are constructed in accordance with the general expression (24) for $vI_0/\Delta\gtrsim 1$. One can say that these curves describe the dispersion curve in the adiabatic inversion regime (see Ref. 10).

But if the inequality (26) is not satisfied (for $vI_0/\Delta\ll 1$), then in the region $\tau^{-1}\ll\delta<|\delta_s|$ the Landau–Zener relation (27) holds. If δ is sufficiently close to $|\delta_s|$, Δ decreases so much that the conditions (26) are satisfied and inversion occurs in this region, too. Graphs of the excitation probability corresponding to this case are shown in Fig. 5b. It can be seen that the maximal

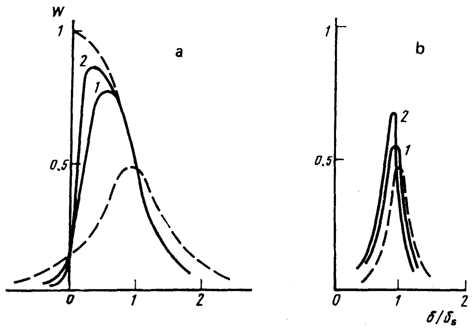

FIG. 5. Dispersion curves for probability of excitation (ionization). a) Adiabatic inversion regime ($v/|\alpha_2-\alpha_1| = 1$): curve 1 corresponds to $\xi_s = 5$, $\xi_s v^2/(\alpha_2-\alpha_1)^2 = 5$, curve 2 to $\xi_s = 10$, $\xi_s v^2/(\alpha_a-\alpha_1)^2 = 10$; the dashed curves show the limits of instantaneous ($\xi_s = 0$) and adiabatic ($\xi_s \to \infty$) switching-on of the interaction. b) Landau–Zener regime ($v/|\alpha_2-\alpha_1| = 0.1$): curve 1 corresponds to $\xi_s = 5$, $\xi_s v^2/(\alpha_2-\alpha_1)^2 = 0.05$, curve 2 to $\xi_s = 10$, $\xi_s v^2/(\alpha_2-\alpha_1)^2 = 0.1$; the dashed curve, to $\xi_s = 0$.

value of the population is in the region[3)] $\delta \sim -\delta_s$. It is natural to call this regime the Landau–Zener inversion regime.

For sufficiently large $I_0 \gg I_c$, the nature of the excitation regime is determined by the ratio $v/|\alpha_2-\alpha_1|$, namely, for $\xi_s v^2/(\alpha_1-\alpha_2)^2 \gg 1$ we have the adiabatic regime and for $\xi_s v^2(\alpha_2-\alpha_1)^2 \lesssim 1$ the Landau–Zener regime, but if $I_0 \gtrsim I_c$, then the nature of the regime is determined by the ratio vI_0/Δ.

6. DISPERSION DEPENDENCE OF THE PROBABILITY OF THREE-PHOTON RESONANCE IONIZATION

Now suppose that ionization can take place from the upper state of the atom under the influence of the considered field. This possibility can be taken into account by assuming α_2 in Eqs. (3) is complex. Then the width of the upper level is determined by

$$\gamma_2 = I\,\mathrm{Im}\,\alpha_2/2.$$

We determine the frequency dependence of the rate of ionization w_i of the atom under conditions of two-photon resonance in the time interval $\tau \ll t \ll \gamma_2^{-1}$, which corresponds to a stationary regime:

$$w_i = \frac{d}{dt}(|a_1|^2+|a_2|^2) = \gamma_2|a_2|^2. \tag{31}$$

In addition, we shall assume that $t \gg |\Delta|^{-1}$, $|\Omega|^{-1}$. These inequalities make it possible to average the ionization probability over the Rabi oscillations. In fact, there are often many reasons leading to the averaging of such oscillations without the assumptions we have made; as examples, we may mention transverse relaxation processes and spatial inhomogeneity of the field.

For the considered time interval, the asymptotic population of the upper level with allowance for the possibility of its decay plays the main part in the expression (31). In the theory of atomic collisions, the problem of nonadiabatic transitions between decaying levels has been considered on a number of occasions.[18] Formally, our formulation of the problem is similar to the problem of taking into account coherent interaction in charge exchange processes in atomic collisions.

To calculate the ionization probability (31), we use the relations (17), assuming now that Δ and Ω are complex. If the obtained states are to describe states of decay type, it is necessary that $\mathrm{Im}(\Delta, \Omega) < 0$. Using the asymptotic behavior of the confluent hypergeometric function, we obtain for $|\Omega| x \gg 1$

$$\langle n_2(t)\rangle = \left|\frac{vI_0}{4\Omega}\right|^2 \frac{\pi\xi}{\mathrm{sh}\,\pi\xi}\exp[-2x(t)\,\mathrm{Im}\,\varepsilon] \times\left\{\left|\frac{(\Omega\tau)^{-\frac{1}{2}i\xi/(1+\Delta/\Omega)}}{\Gamma[1-\frac{1}{2}i\xi(1-\Delta/\Omega)]}\right|^2 \exp\left[-\frac{\pi\xi}{2}\left(1+\mathrm{Re}\frac{\Delta}{\Omega}\right)-\delta(x(t)-t)\,\mathrm{Im}\frac{\Delta}{\Omega}\right] + \left|\frac{(\Omega\tau)^{\frac{1}{2}i\xi(1-\Delta/\Omega)}}{\Gamma[1+\frac{1}{2}i\xi(1+\Delta/\Omega)]}\right|^2 \exp\left[\frac{\pi\xi}{2}\left(1-\mathrm{Re}\frac{\Delta}{\Omega}\right)+2x(t)\,\mathrm{Im}\,\Omega\right]\right\}. \tag{32}$$

In the region $\gamma_2^{-1} \gg t \gg \tau$, we have $x(t) \approx t$, and we therefore finally obtain

$$\langle w_i\rangle = \gamma_2\left|\frac{vI_0}{4\Omega}\right|^2 \frac{\pi\xi}{\mathrm{sh}\,\pi\xi}\left\{\left|\frac{(\Omega\tau)^{-\frac{1}{2}i\xi(1+\Delta/\Omega)}}{\Gamma[1-\frac{1}{2}i\xi(1-\Delta/\Omega)]}\right|^2 \times\exp\left[-\frac{\pi\xi}{2}\left(1+\mathrm{Re}\frac{\Delta}{\Omega}\right)\right] + \left|\frac{(\Omega\tau)^{\frac{1}{2}i\xi(1-\Delta/\Omega)}}{\Gamma[1+\frac{1}{2}i\xi(1+\Delta/\Omega)]}\right|^2 \exp\left[\frac{\pi\xi}{2}\left(1-\mathrm{Re}\frac{\Delta}{\Omega}\right)\right]\right\}. \tag{33}$$

If in this formula we set $\xi = 0$ (instantaneous switching-on of the interaction), we obtain the formula

$$\langle w_i\rangle = 2\gamma_2\left|\frac{vI_0}{4\Omega}\right|^2, \tag{34}$$

which can also be obtained directly from (31) and the expression (20), which determines the mean population of the upper level with allowance for the fact that Ω is now complex.

In the case of adiabatic switching-on of the interaction, $\xi \to \infty$, we consider, as before, two cases. If the values of $1 \pm \mathrm{Re}(\Delta, \Omega)$ are not small, then from (33) we obtain relations analogous to (22):

$$\langle w_i\rangle = \frac{\gamma_2}{2}\left|1-\frac{\Delta}{\Omega}\right|. \tag{35}$$

To obtain this expression, one must use Stirling's formula for the Γ function and note that in the formulation of the problem considered here $\mathrm{Im}(\Delta, \Omega) \ll \mathrm{Re}(\Delta, \Omega)$, since the opposite inequality corresponds to a trivial case of perturbation theory. Therefore, we can assume that

$$\mathrm{Re}\frac{\Delta}{\Omega} \sim \frac{\mathrm{Re}\,\Delta}{\mathrm{Re}\,\Omega}, \quad \mathrm{Im}\frac{\Delta}{\Omega} \sim \frac{\mathrm{Im}\,\Delta}{\mathrm{Re}\,\Omega} \ll 1, \quad \frac{\mathrm{Re}\,\Delta}{\mathrm{Re}\,\Omega}. \tag{36}$$

If $|vI_0/4\Delta| \ll 1$, then the Weisskopf–Wigner formula follows from (34).[4)]

The case of crossing of the levels when $\mathrm{Re}(\Delta/\Omega) \approx -1$ leads to the following expression for the ionization probability of the atom:

$$\langle w_i\rangle = \gamma_2\left|\frac{vI_0}{4\Omega}\right|^2\left\{2\frac{\exp[-\pi\xi(1+\mathrm{Re}(\Delta/\Omega))]}{1-\mathrm{Re}(\Delta/\Omega)} \times|(\Omega\tau)^{i\xi(1+\Delta/\Omega)}|^2\left[\frac{\xi}{2}(1-\mathrm{Re}(\Delta/\Omega))\right]^{-\xi\,\mathrm{Im}(\Delta/\Omega)} + 2\pi\xi\frac{\exp[-\frac{1}{2}\pi\xi(1+\mathrm{Re}(\Delta/\Omega))-\xi(1-\mathrm{Re}(\Delta/\Omega))\arg\Omega]}{|\Gamma[1+\frac{1}{2}i\xi(1+\Delta/\Omega)]|^2}\right\}. \tag{37}$$

From this expression for $\mathrm{Im}\Delta = \mathrm{Im}\,\Omega = 0$ we obtain the expression (27) and the Landau–Zener formulas, which are, thus, a special case of the relation (37) without allowance for the width of the level.

The form of the dispersion curves under conditions of not too large ionization width is close to that shown in Fig. 5. As in the case of excitation, fulfillment of the inequalities permits one to distinguish the adiabatic regime with maximum at $\delta \sim 0$ and the Landau–Zener regime, when $\xi_s v^2/(\alpha_2 - \alpha_1)^2 \lesssim 1$, with narrow maximum at $\delta \sim |\delta_s|$.

1) The linear Stark effect in an alternating field can occur in some cases in systems which are not centrally symmetric,[3] but this possibility is not taken into account here.

2) Nonmonotonicity of the saturation curve in the case of single-photon resonance due to rearrangement of the multiplet structure of the atomic levels in a monochromatic field was considered in Ref. 15.

3) The possibility of a maximum of the dispersion curve at a point other than $\delta = 0$ was noted by Fedorov.[10]

4) To obtain Eq. (1) in the case of instantaneous switching-on of the interaction, it is necessary to use the smallness of $|vI_0/4\Delta|$ already in Eq. (32) and then set $t \gg \gamma_2^{-1}$; this is the usual procedure when one considers decay with instantaneous switching-on of the interaction (cf. Ref. 19).

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Self-focusing of laser beams at various spatial profiles of the incident radiation

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The character of formation of a nonlinear focus was investigated for a media with a Kerr type nonlinearity, under subthreshold conditions as well as in the regime of developed self-focusing at various spatial profiles of the incident radiation. A numerical experiment was used to determine the influence of the profile of the incident radiation and of its intensity on the character of the field distribution in the region of the nonlinear focus and on the power flowing into the first nonlinear focus in the case of beams of the supergaussian type. The dependence of the self-focusing threshold on the initial beam divergence is obtained for both bounded and unbounded media.

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INTRODUCTION

Even though the main laws governing the self-focusing of laser radiation have by now been sufficiently well-investigated, many aspects of this phenomenon remain unclear. Thus, the process investigated in greatest detail was self-focusing of light beams with Gaussian intensity profiles, which were found to have a multi-focus structure.[1] The question of deviation of the incident radiation from gaussian as it affects the main characteristics of wave propagation in a medium with Kerr type of nonlinearity remains open. Thus, for

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