Resonance excitation of atomic levels in a strong electromagnetic field

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The problem of the interaction of a classical electromagnetic field of arbitrary intensity with a two-level atom is investigated with the aid of a method which is mathematically equivalent to the quasiclassical treatment of the problem of subbarrier reflection. It is assumed that the frequency of the external field is small in comparison with the distance between the atomic levels. The resonance probability of a multiphoton transition is calculated in the linear regime. The Stark effect in a strong field is calculated. The range of validity of perturbation theory is determined for the problem under consideration. It is shown that the gauge transformation method,* which has been extensively used in recent years to solve problems of this type, is incorrect.

The goal of the present article is to investigate the interaction of a strong classical electromagnetic field with a two-level atom for the case of a multiphoton resonance. The basic assumption is the smallness of the frequency of the field in comparison with the separation between the levels. Of course, the well known results of perturbation theory [1] are not applicable for strong fields. Both of the atom's states are assumed to be nondegenerate. They are assumed to be dipoles with respect to each other. We shall use the adiabatic approximation to calculate the probability for a transition from the lower state into the upper state. As is well known,[2] it is mathematically equivalent to the quasiclassical approximation for the problem of subbarrier reflection.

Let us write down the total Hamiltonian H(t) in the form of the sum of the atom's Hamiltonian H_0 plus the dipole interaction $V = \mathbf{r}\cdot\mathbf{E}$ with the external electromagnetic field. The electron's charge is everywhere set equal to unity. The quantity $\mathbf{E} = \mathbf{E}_0 \sin\omega t$ is the electric field intensity. In the interests of simplicity, we assume that the wave is plane polarized. Let us direct the field $\mathbf{E}$ along the z axis. We seek eigenstates of the adiabatic Schrödinger equation $H(t)\Psi = \mathscr{E}(t)\Psi$ in the form of a superposition of the unperturbed lower and upper states: $\Psi = b_1\varphi_1 + b_2\varphi_2$. We denote the energy of the lower level by $-\epsilon/2$, and that of the upper level by $+\epsilon/2$. We obtain the following equations for the quantities b_1 and b_2:

$$-{}^1/_2\varepsilon b_1 + V_{12}b_2 = \mathscr{E}(t)b_1, \quad V_{21}b_1 + {}^1/_2\varepsilon b_2 = \mathscr{E}(t)b_2. \tag{1}$$

Let us denote the dipole moment of the transition by $z_{12} = \langle\varphi_2|z|\varphi_1\rangle$. From the system (1) we find that the energy eigenvalues are given by $\mathscr{E}_{1,2}(t) = \mp(1/2)\epsilon(1 + \beta^2\sin^2\omega t)^{1/2}$. We use here the notation $\beta = 2E_0|z_{12}|/\epsilon$. In analogy to the problem of subbarrier reflection, we are only interested in the complex turning points t_k which lie in the upper half-plane. They are determined from the condition $\mathscr{E}_1(t_k) = \mathscr{E}_2(t_k)$. We obtain

$$t_k = \frac{k\pi}{\omega} + \frac{i}{\omega}\operatorname{arsh}\frac{1}{\beta}, \quad k = 0, \pm1, \pm2\ldots \tag{2}$$

These points are the fundamental branch points for $\mathscr{E}_{1,2}(t)$.

First let us consider the point t_0. According to [2] we obtain the following result for the contribution to the transition probability introduced by the point t_0:

$$w_{12} = \exp\left[-2\,\mathrm{Im}\int_{t_1}^{t_0}\omega_{12}(t)\,dt\right].$$

Here $\omega_{12} = \mathscr{E}_1 - \mathscr{E}_2$, and t_1 is any arbitrary point on the real time axis. Evaluating the integral we find

$$w_{12} = \exp\left[-\frac{2n}{\sqrt{1+\beta^2}}D\left(\frac{1}{\sqrt{1+\beta^2}}\right)\right], \tag{3}$$

where $n = \epsilon/\omega$ and D is the complete elliptic integral of the third kind. We see that the quantity w_{12} is exponentially small in terms of the "adiabaticity parameter" n. Formula (3) was derived earlier (see [3,4]) in connection with the solution of the problem of pair production from vacuum in the presence of a strong field, which is mathematically quite similar to the present problem. Referring to this formula, it should also be stressed that the factor appearing in front of the exponential (equal to unity) is exact.

Taking the turning points with $k \neq 0$ into account (they all lie at the same distance from the real time axis) enables us to change from absolute probabilities to probabilities per unit time. According to the principle of superposition,[5] the total amplitude A_{12} for a transition into the upper state is accumulated from the summation of the amplitudes a_k associated with the individual turning points t_k. The corresponding trajectories in the complex-time plane are shown in Fig. 1. The resonance condition is that these amplitudes be added together coherently. The phase factor $-e^{iS}$ appears in the amplitude a_k in connection with the transition from a given turning point to the next. Here the quantity

$$S = n\int_0^{\pi}\sqrt{1+\beta^2\sin^2\varphi}\,d\varphi$$

represents the accumulation of the action between neighboring turning points. The minus sign appears in front of the exponential in the phase factor due to the fact that the quasiclassical wave contains a factor $E^{-1/2}$; it changes sign during the transition from the point t_k to the point t_{k+1}.

Summing the amplitudes from the N turning points and taking the absolute value squared of A_{12}, we find

$$|A_{12}|^2 = w_{12}\frac{\sin^2[N(S-\pi)/2]}{\sin^2[(S-\pi)/2]}.$$

The resonance condition has the form $(S-\pi)/2 = m\pi + \gamma$, where $\gamma \to 0$ and m is an integer. The time interval T is related to N by the formula $T = \pi N/\omega$. Finally we obtain the following expression for the transition probability per unit time, $W_{12} = |A_{12}|^2/T$:

$$W_{12} = \frac{2\omega^2}{\pi}w_{12}\delta\left[\frac{\varepsilon}{\pi}\int_0^{\pi}\sqrt{1+\beta^2\sin^2\varphi}\,d\varphi - K\omega\right], \tag{4}$$

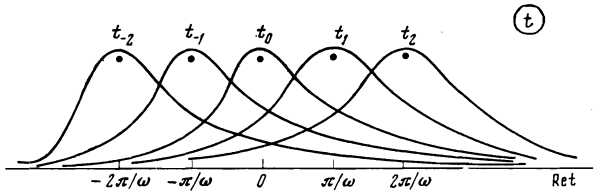

FIG. 1. Complex turning points and trajectories in the upper half of the complex t plane.

where $K = 2m + 1$. As must happen because of the dipole selection rules, absorption of the wave only occurs in the odd harmonics. The δ-function in formula (4) expresses the law of energy conservation, and its argument contains the magnitude of the energy shift due to the external field (the Stark effect).

A graph of the dependence of the function n/K on β is shown in Fig. 2. The perturbation-theory result with regard to the Stark effect, $n/K \to 1$, is obtained when $\beta \ll 1$, i.e., when the field intensity expressed in atomic units is small compared to unity. We see that the resonance ratio $n = \epsilon/\omega$ decreases with increasing intensity of the field.

Now let us proceed to an analysis of the transition probability W_{12}. Provided that $K\beta^2 \ll 1$, we obtain from formula (4) the transition probability in perturbation theory:

$$W_{12}^0 = 2\pi^{-1}\omega^2 (e\beta/4)^{2K}\delta(\varepsilon - K\omega). \qquad (5)$$

We see that the criterion for the validity of perturbation theory with regard to the transition probability is more stringent than with regard to the Stark effect. To wit, the intensity of the external field must be small in comparison with $K^{-1/2}$. On the other hand, let us write down the well known expression for the transition probability $W_{12}^{0\prime}$ which is obtained by the direct application of time-dependent perturbation theory:[1]

$$W_{12}^{0\prime} = 2\pi\omega^2 (K\beta/4)^{2K}[(K-1)!!]^{-4}\delta(\varepsilon - K\omega). \qquad (6)$$

Using Stirling's formula one can easily verify that expressions (5) and (6) agree exactly for $K \gg 1$. It turns out that the WKB approximation works reasonably well even for small values of K. For example, the values of the ratio $W_{12}^0/W_{12}^{0\prime}$ are equal to 0.75, 0.89, and 0.95 for the harmonics K equal to 1, 3, and 5, respectively.

It is also convenient to define a transition probability expressed in units of the probability W_{12}^0: $F \equiv W_{12}/W_{12}^0$. The quantity F characterizes the deviation of the transition probability from the value predicted by perturbation theory. From Eqs. (4) and (5) we find $F \cong e^{-(1/2)K\beta^2}$ for small values of β. This can be written as an effective reduction in the value of the exponent characterizing the dependence of W_{12} on the field intensity: $K' = K(1-\beta^2/2)$, which agrees with the experimental data on the excitation of multiphoton resonances in strong fields.[6]

The universal dependence of the quantity

$$\mathcal{Q} = \frac{1}{2K}\ln\left[\frac{\pi W_{12}}{2\omega^2}\right].$$

on β is shown in Fig. 3. We see that the rate of growth of W_{12} slows down as the intensity of the field increases. The found solution loses its validity for $\beta > 1$ (when w_{12} is of the order of unity), i.e., for field intensities of the order of unity. The WKB approximation used here is not

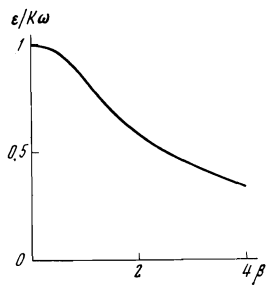

FIG. 2

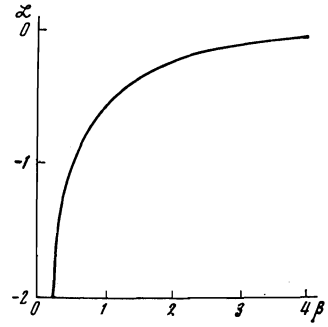

FIG. 3

FIG. 2. The dependence of the ratio of the transition energy ϵ to the energy $K\omega$ of the absorbed quanta on the parameter β, which characterizes the intensity of the external field (Stark effect).

FIG. 3. The universal dependence of the logarithm of the transition probability on β.

applicable for large fields. The solution is also not suitable for too large times T of interaction with the field. The condition $|A_{12}|^2 \ll 1$ implies that one must have $T \ll 1/\omega\sqrt{w_{12}}$. Saturation sets in at large times, in which case transitions from the upper state to the lower state also occur, and both levels are characterized by average populations. At the same time one must satisfy $T \gg 1/\omega$; otherwise it will not be possible to form the resonance. The saturation regime does not appear in a real situation, owing to the large ionization width of the upper level, and therefore it is not treated here.

One can show that taking other atomic levels into consideration basically amounts to a renormalization of the quantity β. For example, one can take the existence of a level with higher energy ϵ_3, which is coupled to the ground state by the dipole matrix element z_{13}, into account by making the substitution $\beta \to \beta(1+\gamma)$ in all formulas, where $\gamma = (\epsilon/8\epsilon_3)|z_{13}/z_{12}|^2$. In the general case the quantity β_{eff}, which is defined by the atomic Green's function, appears in the formulas.

The so-called* gauge transformation method[7] has recently been proposed for the solution of the problem under consideration. This method is based on the fact that in the presence of a slowly varying field A(t) the wave function $\Psi(t)$ can be written in the form $\Psi(t)$ $e^{i\mathbf{A}\cdot\mathbf{r}}\Phi(t)$, where $\Phi(t)$ is the unperturbed wave function. An expression of this form would be exact in the case of a constant field. Now we shall show that this method leads to erroneous results when it is applied to the problem under investigation.

Of course, the corrections to the function $\Psi(t)$ are proportional to the adiabaticity parameter and are therefore small. However, they give a large contribution to the transition amplitude, which is determined by the elements of the T-matrix:

$$T_{if} = \int [\Phi_i(t)\mathbf{r}\mathbf{E}(t)\Psi_f(t)]\,dt.$$

Actually the integrand of the expression for Tif is rapidly oscillating. One can easily verify that when the corrections of the next order in the adiabatic parameter are added to the function $\Psi_f(t)$, the integrand oscillates more weakly. Consequently the corrections introduce a contribution to the integral which is of the same order as the fundamental term. Mistakes of this type were made in [8] in connection with an investigation of transi-

tions into the continuum due to the influence of a strong field; these mistakes were corrected in [9].

Ionization from the upper level into the continuum appears substantially following the resonance multiphoton absorption. In addition, direct ionization from the lower level due to the influence of the field competes with the given process.

In conclusion the authors express their gratitude to V. S. Popov for a discussion of this article.

*Note by translator: Reiss's method is called the momentum translation approximation.

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