

Interference effects in the laser-pulse-induced autoionization spectrum of an atom

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The ionization of an atom by a smooth laser pulse is described using an adiabatic approximation developed in the theory of atomic collisions. Adiabatic quasienergy states are introduced and the process of filling them is described. It is shown that the autoionization cross section in the wings of the spectrum develops an oscillatory structure because of the interference of electrons emitted from these states at the leading and trailing edges of a pulse. At the center of this spectrum there is a resonance peak corresponding to an autoionization line. The intensity of this peak vanishes when the "condition of quantization" of the area under a pulse is satisfied; the peak profile is independent of the shape of the laser pulse. Consequently, when this quantization condition is obeyed the absorption of a pulse is minimal.

1. INTRODUCTION

The processes occurring in atoms under the influence of smooth laser pulses have begun to attract attention. Studies of these processes are related directly to the role of switching the field on and off. It is usual to assume that the field is switched on instantaneously¹⁻⁵ or adiabatically slowly.^{6,7} The hypothesis of instantaneous switching on of the field is justified only in the case of resonance processes, whereas in the case of adiabatic switching on it is presumed that the detuning from the resonance is large.⁸ Therefore, details of switching on of the field are important only in investigations of quasiresonant processes. Several papers have been published recently⁹⁻¹⁶ in which exactly soluble models are used to analyze such processes under the influence of a smooth laser pulse. It has been found that the cross sections of the various processes (such as resonant fluorescence,^{9,10} resonant ionization,^{11,12} and resonant autoionization^{13,14}) exhibit an oscillatory structure. It is usual to point out the special role of the pulses of amplitude $E_0(t)$ satisfying certain "quantization conditions" imposed on the integral $\int_{-\infty}^{\infty} E_0(t) dt$. However, a qualitative explanation of these features has not yet been provided.

We shall show that the processes induced by a smooth pulse can be described satisfactorily by the methods and results of the theory of nonadiabatic transitions in atomic collisions; the analogy is of fundamental importance. The oscillatory structure of the electron spectrum appears because of the interference of electrons leaving the system at the leading and trailing edges of a pulse. Such oscillations have been investigated earlier in the case of collisions of heavy particles¹⁷ and a detailed review of the results and methods can be found in Ref. 18. However, an important distinction is that we do not need to average over the impact parameter, so that the oscillations should be much clearer. We should mention here an earlier attempt to describe the detachment of an electron under the action of a smooth laser pulse,¹⁹ but the analogy with the theory of collisions was not pointed out there and the results obtained cannot be regarded as final.

2. ADIABATIC QUASIENERGY STATES

We shall consider a system which has a bound state $|0\rangle$ with an energy ε_0 , a continuum $|\mathbf{k}\rangle$ with a dispersion law $\varepsilon(\mathbf{k})$, and a diabatic state $(|1\rangle, \varepsilon_1)$ immersed in this contin-

uum [$\langle 0|0\rangle = \langle 1|1\rangle = 1$, $\langle \mathbf{k}|\mathbf{k}'\rangle = \delta^3(\mathbf{k} - \mathbf{k}')$]. This system is in an alternating field with a slowly varying amplitude: $E = eE_0(t) \cos \omega t$. It is assumed that the system can undergo dipole transitions $|0\rangle \rightarrow |1\rangle$ (matrix element D) and $|0\rangle \rightarrow |\mathbf{k}\rangle$ [matrix element $D(\mathbf{k})$] and autoionization $|1\rangle \rightarrow |\mathbf{k}\rangle$ [matrix element $V(\mathbf{k})$]. The wave function of the system is

$$|\psi\rangle = a_0(t) e^{i\omega t} |0\rangle + a_1(t) |1\rangle + \int d^3k b(\mathbf{k}, t) |\mathbf{k}\rangle + |\delta\psi\rangle, \quad (1)$$

where $|\delta\psi\rangle$ includes a contribution of the states corresponding to multiphoton processes (it is assumed that $\varepsilon_1 \approx \varepsilon_0 + \omega$, holds i.e., that states $|0\rangle$ and $|1\rangle$ satisfy one-photon resonance conditions). In order to determine the coefficient functions in Eq. (1) using the rotating wave approximation,⁸ we obtain (in atomic units)

$$\begin{aligned} i\dot{a}_0 &= (\varepsilon_0 + \omega) a_0 + \frac{1}{2} E_0 D a_1 + \frac{1}{2} E_0 \int D(\mathbf{k}) b(\mathbf{k}, t) d^3k, \\ i\dot{a}_1 &= \varepsilon_1 a_1 + \frac{1}{2} E_0 D a_0 + \int V(\mathbf{k}) b(\mathbf{k}, t) d^3k, \\ i\dot{b}(\mathbf{k}, t) &= \varepsilon(\mathbf{k}) b(\mathbf{k}, t) + \frac{1}{2} E_0 D(\mathbf{k}) a_0 + V(\mathbf{k}) a_1. \end{aligned} \quad (2)$$

For simplicity, we are neglecting the contribution $|\delta\psi\rangle$. An allowance for this contribution in accordance with perturbation theory gives rise to a dynamic polarizability of the states $|1\rangle$ and $|\mathbf{k}\rangle$ and, in the final analysis, it results in redefinition of the polarizability of the state $|0\rangle$ denoted later by α . It therefore follows that if we regard α as fitting parameter, we can also include the contribution of the states $|\delta\psi\rangle$.

In view of the time dependence of $E_0(t)$, the system (2) cannot be solved analytically. However, from the practical point of view, it is sufficient to consider the case of a relatively slow change in E_0 . In this limit it is reasonable to introduce adiabatic quasienergy states dependent parametrically on E_0 by substituting $a_i = c_i \exp(-i\lambda t)$ into Eq. (2):

$$\begin{aligned} (\lambda - \varepsilon_0 - \omega) c_0 &= \frac{1}{2} E_0 D c_1 + \frac{1}{2} E_0 [F_{DV}(\lambda) c_1 + \frac{1}{2} F_{DD}(\lambda) E_0 c_0], \\ (\lambda - \varepsilon_1) c_1 &= \frac{1}{2} E_0 D c_0 + F_{VV}(\lambda) c_1 + \frac{1}{2} E_0 F_{VD}(\lambda) c_0, \end{aligned} \quad (3)$$

where

$$F_{\alpha\beta}(\lambda) = \int d^3k \alpha(\mathbf{k}) \beta(\mathbf{k}) [\lambda - \varepsilon(\mathbf{k}) + i0]^{-1}.$$

We are interested only in the case of a resonance when $|\varepsilon_0 + \omega - \varepsilon_1| \ll \omega$, and λ is quite close to ε_1 . We can then expand $F_{\alpha\beta}$ as a series in the vicinity of $\lambda = \varepsilon_1$.¹¹ We shall

introduce Z , \bar{D} , α , and ε_r , assuming that

$$z = (\partial F_{VV} / \partial \lambda)_{\lambda = \varepsilon_1}, \quad \bar{D} = [D + F_{VD}(\varepsilon_1)](1-z)^{-1/2}, \quad (4)$$

$$\alpha = F_{DD}(\varepsilon_1), \quad \varepsilon_r = \varepsilon_1 + F_{VV}(\varepsilon_1)(1-z)^{-1},$$

where α is the dynamic polarizability of the state $|0\rangle$, and ε_r represents the energy of the resonance state. We must stress that all the quantities introduced so far, generally speaking, are complex variables. Substitution of Eq. (4) into Eq. (3) gives the condition of solubility of Eq. (3):

$$\lambda_{\pm} = \pm^{1/2} \{ \varepsilon_0 + \omega + \varepsilon_r + \alpha E_0^2 \pm [(\varepsilon_0 + \omega + \alpha E_0^2 - \varepsilon_r)^2 + E_0^2 \bar{D}^2]^{1/2} \} \quad (5)$$

and the unnormalized solutions of the system (3) are

$$c_0^{\pm} = 1, \quad c_1^{\pm} = 2(\lambda_{\pm} - \varepsilon_r - \alpha E_0^2)(1-z)^{-1/2}(E_0 \bar{D})^{-1}. \quad (6)$$

The quantity which can be determined experimentally is the spectrum of the emitted electrons $S(\mathbf{k})$, which is proportional to the square of the modulus of the corresponding amplitude $d(\mathbf{k})$:

$$d(\mathbf{k}) = \lim_{t \rightarrow \infty} \exp[i\varepsilon(\mathbf{k})t] b(\mathbf{k}, t) = \int_{-\infty}^{\infty} [V(\mathbf{k}) a_0(t) + \alpha^{1/2} D(\mathbf{k}) E_0(t) a_1(t)] \exp[i\varepsilon(\mathbf{k})t] dt. \quad (7)$$

If the external field is weak and the absolute value of the detuning $\delta = \varepsilon_r - \varepsilon_0 - \omega$ is relatively large, the only quasi-energy state which becomes populated is that which reduces to the state $|0\rangle$ in the zero-field limit, i.e.,

$$a_0 \approx 1, \quad a_1 \approx -\alpha^{1/2} E_0 \bar{D} / \delta, \quad \lambda = \varepsilon_0 + \omega + \alpha^{1/2} E_0^2 (\alpha - 2\bar{D}^2 / \delta).$$

In this case we have

$$d(\mathbf{k}) = \frac{1}{2} \left[D(\mathbf{k}) - \frac{\bar{D} V(\mathbf{k})}{\delta} \right] \int_{-\infty}^{\infty} E_0(t) \exp\{i[\varepsilon(\mathbf{k}) - \varepsilon_0 - \omega]t\} dt,$$

which makes it possible to determine the main parameters of the theory $V(\mathbf{k})\bar{D}$ and $D(\mathbf{k})$ from the experiments on conventional (nonlaser) photoionization.

3. ADIABATIC APPROXIMATION AND NONADIABATIC TRANSITIONS

In the adiabatic approximation the solution of the system (2) is

$$a_i(t) = A_+ c_i^+(t) \exp\left[-i \int_t^{\infty} \lambda_+(t) d\tau\right] + A_- c_i^-(t) \exp\left[-i \int_t^{\infty} \lambda_-(t) d\tau\right], \quad (8)$$

where $\lambda_{\pm}(t)$ and $c_i^{\pm}(t)$ are given by Eqs. (5) and (6), i.e., they depend on time via the dependence $E_0(t)$. In our case, because of the complex nature of the main characteristics of the system λ_{\pm} in the case of real values of t , we have to determine the lower limit of integration in Eq. (8). We shall consider symmetric laser pulses characterized by $E_0(t) = E_0(-t)$; it is then convenient to take $t_1 = 0$.

The adiabatic representation of Eq. (8) postulates that the coefficients A_{\pm} are independent of time, which is not true in the vicinity of the points of degeneracy of the system (3) when $\lambda_+ = \lambda_-$. Such degeneracy occurs in a critical

field

$$E_{cr} = \pm 2i\alpha^{-1} [\bar{D} \pm (\bar{D}^2 - \alpha\delta)^{1/2}].$$

If the detuning is small, only the values

$$E_{cr1} = \pm (2i/\alpha) [\bar{D} - (\bar{D}^2 - \alpha\delta)^{1/2}] \quad (9)$$

are relatively small when $\lambda \approx (\varepsilon_0 + \omega + \varepsilon_r)/2$ is close to ε_1 and lies within the range of validity of the expansion of $F_{\alpha\beta}$. Other values of the critical field are very large, of the order of the atomic field, and the behavior of the system under these conditions is outside the scope of the present study.

The value of the critical field determines when the adiabatic approximation of Eq. (8) breaks down: $E_0(t_{cr}) = E_{cr1}$. In the case of exponential switching of the field on or off, which is the most important process in our problem, these moments form two vertical series in the complex t plane, similar to the series in the model of Nikitin.²⁰ In this case the standard problem for the description of nonadiabatic transitions corresponds to a system of equations

$$i\dot{a}_0 = (\varepsilon_0 + \alpha E_0^2) a_0 + \alpha^{1/2} E_0 \bar{D} a_1, \quad (10)$$

$$i\dot{a}_1 = \varepsilon_r a_1 + \alpha^{1/2} E_0 \bar{D} a_0$$

if $E_0 = A \exp(\gamma t)$. The solution of this system subject to the initial conditions $a_0(-\infty) = 1$ and $a_1(-\infty) = 0$ can be expressed in terms of the confluent hypergeometric functions:

$$a_0(t) = {}_1F_1\left(\frac{1}{2} + i\frac{\delta}{2\gamma} - i\frac{\bar{D}^2}{2\gamma\alpha}, \frac{1}{2} + i\frac{\delta}{2\gamma}; -i\alpha \frac{E_0^2(t)}{8\gamma}\right) \cdot \exp[-i(\varepsilon_0 + \omega)t] \quad (11)$$

[the expression for $a_1(t)$ can be obtained from Eq. (11) and one of the equations in the system (10); we shall not need it later]. This solution should be matched to the adiabatic representation of Eq. (8). However, the matching in the limit $t \rightarrow \infty$ is unsatisfactory because such an asymptote postulates passage through both nonadiabatic regions, whereas the second region is physically meaningless. The correct solution of the problem can be obtained by constructing a uniform asymptotic representation for the hypergeometric functions in Eq. (11), which however leads to very cumbersome expressions. We shall adopt a simpler procedure: we shall consider the asymptotic expression (11) in the case when $|\bar{D}|^2 / \alpha \gg \gamma$, but $\delta \sim \gamma$:

$$a_0 = \Gamma\left(\frac{1}{2} + i\frac{\delta}{2\gamma}\right) \left(\frac{E_0 \bar{D}}{4\gamma}\right)^{(\gamma - i\delta)/2\gamma} J_{-(\gamma - i\delta)/2\gamma}\left(\frac{E_0 \bar{D}}{2\gamma}\right) \cdot \exp\left[-i(\varepsilon_0 + \omega)t - i\frac{\alpha E_0^2}{16\gamma}\right], \quad (12)$$

where $\Gamma(x)$ is the gamma function, $J_{\beta}(u)$ is a Bessel function. Representation described by Eq. (12) corresponds to the solution of the Demkov problem²¹ describing the passage through one nonadiabatic region. Therefore, comparing Eqs. (12) and (8), we obtain

$$A_{\pm} = \pi^{-1/2} \Gamma(1/2 + iq) (A\bar{D}/4\gamma)^{-iq} \exp[\mp^{1/2} \pi q - i^{1/2} i(Y \pm Z)], \quad (13)$$

where

$$q = \frac{\delta}{2\gamma}, \quad Y = \frac{1}{8} \alpha \int_{-\infty}^{\infty} E_0^2(t) dt,$$

$$Z = i\pi q + \frac{1}{2} \int_{-t_{cr}}^{t_{cr}} (\lambda_+ - \lambda_-) dt.$$

If the detuning is small, so that $\delta \ll \max |E_0(t)\tilde{D}|$, then

$$Z \approx \frac{1}{2} \tilde{D} \int_{-\infty}^{\infty} E_0(t) dt. \quad (14)$$

By analogy with Eq. (12), we can obtain expressions describing the process of switching off the field. The solution of interest to us is then

$$a_0 = \Gamma^{(1/2+iq)} \exp(-iY) (E_0\tilde{D}/4\gamma)^{1/2-iq} \{ \cos Z - i \operatorname{th} \pi q \sin Z \} J_{-1/2-iq}(E_0\tilde{D}/2\gamma) + \sin Z J_{1/2+iq}(E_0\tilde{D}/2\gamma) \} \exp(-i\epsilon_r t). \quad (15)$$

The probability that the system does not become ionized by a single pulse,

$$W = |a_0(+\infty)|^2 = \left| \frac{\Gamma^{(1/2+iq)} (A\tilde{D})^{-2iq}}{\Gamma^{(1/2-iq)}} (\cos Z + i \operatorname{th} \pi q \sin Z) \right|^2,$$

can be described more simply if the imaginary parts of the main characteristics are small:

$$W = 1 - \sin^2 Z / \operatorname{ch}^2 \pi q. \quad (16)$$

For a fixed value of the detuning δ the probability W oscillates as a function of Z ; if $Z = \pi n$, the ionization probability is minimal. It should be pointed out that expressions similar to Eq. (16) are known from the theory of nonadiabatic transitions occurring in atomic collisions (see Refs. 21 and 22 and also Ref. 23).

4. SPECTRUM OF EMITTED ELECTRONS

The amplitude of the spectrum of emitted electrons $d(\mathbf{k})$ of Eq. (7) can be transformed, subject to Eq. (10), to the following form where a_1 does not occur:

$$d(\mathbf{k}) = \frac{1}{2} \left[D(\mathbf{k}) + \frac{\tilde{D}V(\mathbf{k})}{\epsilon(\mathbf{k}) - \epsilon_r} \right] \int_{-\infty}^{\infty} e^{i\epsilon(\mathbf{k})t} a_0(t) E_0(t) dt. \quad (17)$$

We shall assume that a laser pulse has an envelope shown in Fig. 1. For a given detuning δ from a resonance we can obtain E_{cr} from Eq. (9) and then the nonadiabatic regions lie in the vicinity of the points $t = \pm \operatorname{Re} t_{cr}$. We can identify three natural regions in the electron spectrum (Fig. 2): the wings of a line (I and III) and the line center (II). Electrons corresponding to the line wings are emitted from adiabatic quasienergy states and the line center is due to regions $|t| > \operatorname{Re} t_{cr}$.

We shall now consider the wings of a line. We shall find the spectrum by calculating the integral of Eq. (17) by the method of stationary phase. The stationary points t_s^\pm obtained in such calculations satisfy the conditions $\lambda_\pm(t_s^\pm) = \epsilon(\mathbf{k})$ and represent the times at which an electron of energy $\epsilon(\mathbf{k})$ becomes detached. It is clear from Fig. 1 that if $\epsilon(\mathbf{k}) \leq \operatorname{Re} \lambda_+(0)$ or $\epsilon(\mathbf{k}) \geq \operatorname{Re} \lambda_-(0)$, there are two such points; addition of the corresponding contributions

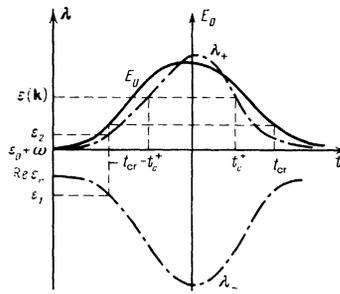


FIG. 1. Time dependence of the real parts of the pulse amplitude $E_0(t)$ and of the adiabatic quasienergies $\lambda_\pm(t)$. The following notation is used: $\pm t_c$ are the moments of detachment of an electron of energy $\epsilon(\mathbf{k})$ in the adiabatic region; $\pm t_{cr}$ are the moments in the vicinity of which there is a nonadiabatic transition characterized by $E_0 = E_{cr}$ of Eq. (9); ϵ_1 and ϵ_2 represent the energies separating the wings from the center of the spectrum.

gives rise to an oscillatory structure of the spectrum. The right-hand wing of the line [$d_+(\mathbf{k})$] is created by an adiabatic quasienergy state $\lambda_+(t)$ and the left-hand wing [$d_-(\mathbf{k})$] by an adiabatic quasienergy state $\lambda_-(t)$:

$$d_\pm(\mathbf{k}) = \left[D(\mathbf{k}) + \frac{\tilde{D}V(\mathbf{k})}{\epsilon(\mathbf{k}) - \epsilon_r} \right] \left(\frac{A\tilde{D}}{4\gamma} \right)^{-iq} \Gamma\left(\frac{1}{2} + iq\right) \cdot \exp\left[-\frac{1}{2}i(Y+Z)\right] \exp\left(\mp \frac{1}{2}\pi q\right) E_0(t_c^\pm) \cdot \left(\pm 2 \frac{d\lambda_\pm}{dt} \Big|_{t_c^\pm} \right)^{-1/2} \cos\left\{ \pm \int_{t_c^\pm}^0 [\lambda_\pm(t) - \epsilon(\mathbf{k})] dt - \frac{\pi}{4} \right\}. \quad (18)$$

It is clear from Eq. (18) that in the case of large values of the parameter q which represents the product of the detuning and the time at which the field is switched on) one of the wings is suppressed exponentially because the state which reduces to the state $|0\rangle$ at the moment the field is switched off is populated preferentially. The total number of oscillations N can be obtained from Eq. (18):

$$N = Z - 2q [\ln(A\tilde{D}/4\gamma q) - 1],$$

i.e., the number of oscillations is governed by the area Z under a pulse, as already pointed out in Refs. 9–12. Naturally, oscillations in the spectrum appear only if the decay of adiabatic quasienergy states is relatively weak in the course

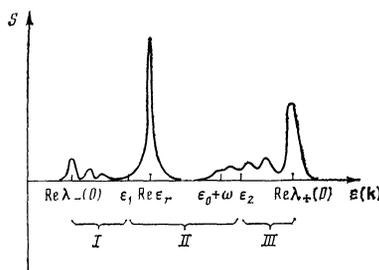


FIG. 2. Spectrum of the emitted electrons. Regions I and III (wings of the spectrum) are due to the detachment of an electron in the adiabatic region, whereas II is the center of the spectrum. The central peak corresponds to the autoionization of a free atom.

of motion from one point of detachment to another. In any case, such decay makes the oscillations incomplete.

It would be of interest to describe the profile of the outermost line in the wings of the spectrum. This line is due to electrons which leave the system in the vicinity of extrema of the quasienergies λ_+ and λ_- . If a pulse has a clear peak, we can use the quadratic approximation so that the calculation of Eq. (17) yields an Airy function (by analogy with Ref. 17). However, if the pulse has a long plateau, during which the system decays significantly, then it is preferable to use a different approximation: $E_0 = E_m - B \exp(-\gamma_1 t)$ or $E_0 = E_m - B t^{-\beta}$ (Ref. 17).

We shall consider the spectrum of electrons in the central part III shown in Fig. 2. We shall do this by calculating the integral (17) and use the standard solutions of Eqs. (12) and (15). We then obtain

$$d_{\text{III}}(\mathbf{k}) = \left[D(\mathbf{k}) + \frac{\bar{D}V(\mathbf{k})}{\varepsilon(\mathbf{k}) - \varepsilon_r} \right] \frac{\Gamma(1/2 + iq)}{\bar{D}} \left\{ \left(\frac{4\gamma}{A\bar{D}} \right)^{i[\varepsilon(\mathbf{k}) - \varepsilon_0 - \omega]/\gamma} \cdot \Gamma\left(\frac{1}{2} - i \frac{\varepsilon_0 + \omega - \varepsilon(\mathbf{k})}{2\gamma} \right) \left[\Gamma\left(i \frac{\varepsilon_r - \varepsilon(\mathbf{k})}{2\gamma} \right) \right]^{-1} + \left(\frac{4\gamma}{A\bar{D}} \right)^{i[\varepsilon_r - \varepsilon(\mathbf{k})]/\gamma} e^{-iX} \Gamma\left(\frac{1}{2} + i \frac{\varepsilon_0 + \omega - \varepsilon(\mathbf{k})}{2\gamma} \right) \cdot \left[\Gamma\left(-i \frac{\varepsilon_r - \varepsilon(\mathbf{k})}{2\gamma} \right) \right]^{-1} \cdot \left[\cos Z + i \sin Z \operatorname{cth} \frac{1/2\pi[\varepsilon_r - \varepsilon(\mathbf{k})]}{\gamma} \right] \right\}. \quad (19)$$

The first term in braces represents the contribution of the leading edge of a pulse, whereas the second term corresponds to the trailing edge. The factor

$$\left| \left(\frac{4\gamma}{A\bar{D}} \right)^{i[\varepsilon_r - \varepsilon(\mathbf{k})]/\gamma} e^{-iX} \right|$$

describes the process of decay of the system during its motion in the adiabatic region. Naturally, the contribution of the trailing edge is significant only if this factor is not too small. The most important feature of the spectrum at the center is the presence of a pole contribution due to decay of an autoionizing state. It follows from Eq. (19) that this contribution is proportional to $\sin Z$ and its profile depends weakly on the nature of the process of switching on the field, governed by the parameter γ . It should be pointed out particularly that when the condition $Z = \pi n$ is satisfied, the line corresponding to decay of an autoionizing state is missing after the passage of a pulse, in agreement with Eq. (16).

5. CONCLUSIONS

We shall now summarize the results. A smooth laser pulse induces a transition of an atomic system to a superposition of adiabatic quasienergy states. This transition is described by the usual theory of nonadiabatic processes in

atomic collisions. According to this theory, the probability that the system returns to its initial state after the passage of a pulse oscillates as a function of the pulse area and can be close to unity. For pulses of this kind the absorption is minimal. Autoionization of electrons with a specific energy occurs (in the adiabatic part of a pulse) at two points where the corresponding contributions interfere, creating an oscillatory structure of the spectrum. In the central part of the spectrum there is a resonant pole maximum corresponding to decay of an autoionizing state filled after the passage of a pulse. If the probability of survival of the system is close to unity, the intensity of this maximum is close to zero.

It should be stressed that our analysis of one-photon processes far from the threshold can be extended also to multiphoton processes.

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¹⁾ We are assuming that ε_1 lies far from the threshold. Near-threshold processes can be considered using our approach, but a separate analysis is then required.

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