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## Threshold features of the excitation and ionization of atoms by high-intensity electromagnetic radiation

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The step-by-step transition from the resonance excitation of the levels of a discrete spectrum by an external electromagnetic field to the ionization of the atom is studied on the basis of a unified theoretical approach. It is shown that in the intermediate region, where the interaction encompasses a large number of levels, the excitation process is on the whole irreversible, but retains to some extent the properties of the transitions between isolated discrete levels: the excitation probability depends nonmonotonically on the time. The nonexponential decay of the bound state in the near-threshold region is investigated. A quasistationary regime characterized by a constant mean probability for finding the atom in the ground state is found to exist in some interval of time. The threshold for the excitation and ionization decay processes is found to be depressed by an amount determined in a weak field by the parameter  $t^{-2/3}$  and in a strong field by the parameter  $F^{4/3}$ , where  $t$  and  $F$  are the pulse length and amplitude of the radiation intensity.

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### 1. INTRODUCTION

The probability,  $w$ , for a direct single-photon ionization of an atom, which depends linearly on the time  $t$  in the initial phase of the process, is at large  $t$  usually described by the formula<sup>1,2</sup>

$$w = 1 - e^{-\Gamma t}, \quad (1)$$

which corresponds to a simple exponential decay of the ground state. Here  $\Gamma$  is the ionization width:

$$\Gamma = 2\pi |\langle \psi_1 | V | \psi_{E_1+\omega} \rangle|^2, \quad (2)$$

$V = \frac{1}{2} \mathbf{d} \cdot \mathbf{F}$  is the atom-field interaction operator;  $\mathbf{d}$  is the dipole moment;  $\mathbf{F}$  is the intensity amplitude of the alternating electric field,  $\mathbf{F}(t) = \text{Re}(\mathbf{F}e^{-i\omega t})$ , of frequency  $\omega$ ;  $\psi_1$  and  $E_1$  are the wave function and energy of the ground state;  $\psi_{E_1+\omega}$  is the wave function of the continuous spectrum with energy  $E = E_1 + \omega$ . Here and below we use the atomic system of units. The ionization width  $\Gamma$  is a slowly varying function of the energy  $E = E_1 + \omega$ , and tends to a constant limit as  $E_1 + \omega \rightarrow 0$ , in accord with the threshold property of the cross section for the photoelectric effect in neutral atoms.<sup>3</sup> In view of the slowness

of the variation of  $\Gamma$ , below we shall consider it to be a constant.<sup>1</sup>

Let us, as usual, assume that  $F \ll 1$ . The ionization width  $\Gamma$  can, in this case, easily be estimated: for  $\omega \sim 1$ , we have  $\Gamma \sim F^2 \ll 1$ . Correspondingly, the characteristic ionization time  $\tau = \Gamma^{-1} \gg 1$ . Below we shall consider times  $t$  that are large or small compared to  $\tau$ , but always large as compared to the wave period or the characteristic intra-atomic times:  $t \gg 1$ .

The expression (1) is usually derived under the assumption that the ionization threshold is sufficiently well exceeded, i.e., that  $E_1 + \omega \gg \Gamma$ .<sup>1,2</sup> We shall show below that the conditions for the applicability of this result imposes definite limitations on the duration,  $t$ , of the interaction.

Of indubitable physical interest is the investigation of the time dependence of the processes of excitation and ionization of an atom in the near-threshold region  $|E_1 + \omega| \sim \Gamma$  and in the region,  $E_1 + \omega < 0$ ,  $|E_1 + \omega| \gg \Gamma$ , below the ionization threshold. The present paper is devoted to the study of these problems.

The quite rigid condition,  $\omega \sim 1$ , used above for the estimation of the quantity  $\Gamma$  is actually not necessary. We shall also consider frequencies significantly lower than the atomic frequencies, bearing in mind the ionization of arbitrary atoms from their excited states. The only condition that will be used is that the frequency  $\omega$  significantly exceed the ionization potential of the resonance, highly-excited, discrete levels with energy  $\sim E_1 + \omega$ .

## 2. THE GENERAL EQUATIONS

Let us consider an atom with one optical electron in the field  $F(t)$ . We shall assume that the relation between  $|E_1 + \omega|$  and  $\Gamma$  can be arbitrary, but that  $|E_1 + \omega| \ll 1$ . This means that we shall consider either the ionization of the atom in the case when the excess over the threshold is not too much (in comparison with the atomic energy), or the excitation of the high-lying levels of the discrete spectrum. In this case we can consider the spectrum of the atom in the region below the continuum threshold to be hydrogenic, and use for the values of the energy  $E_n$  the expression  $E_n = -1/2n^2$ , with  $n \gg 1$ . For  $E_1 + \omega < 0$ , it is also convenient to use the notation

$$E_1 + \omega = -\frac{1}{2n_0^2} + \frac{\beta}{n_0^3}, \quad (3)$$

where  $n_0 \gg 1$ ,  $|\beta| \leq 1$ ;  $n_0$  is the number of the discrete atomic level closest to the energy  $E_1 + \omega$ .

Let us expand the wave function of the atom in the field  $F(t)$  in terms of the wave functions of the free atom  $\psi_1$ ,  $\psi_n$ ,  $\psi_E$ :

$$\Psi(t) = A_1(t)e^{i\omega t}\psi_1 + \sum_{n=2}^{\infty} A_n(t)\psi_n + \int_0^{\infty} A_E(t)\psi_E dE. \quad (4)$$

Let us substitute the expansion (4) into the Schrödinger equation and use the following approximations based on the limitation  $F \ll 1$ .

1. We shall take into consideration the coupling of the ground state with the highly-excited discrete levels and the states of the continuum, but shall neglect the transi-

tions from these states into remote states of the continuous spectrum with energy  $\sim E_1 + 2\omega$ .

2. We shall assume that the coupling between the ground and excited states is effected by only that part of the interaction operator which has the "correct" frequency sign, which is the sign that allows the fulfillment of the energy conservation law  $E_n \approx E_1 + \omega$  or  $E \approx E_1 + \omega$ .

3. We shall take into account in the energies  $E_1$  and  $E_n$  the level shift due to the nonresonance Stark effect.<sup>2</sup> For the ground state this shift is equal to  $-\frac{1}{4}\alpha_1 F^2$ , where  $\alpha_1$  is the dynamical polarizability of the state. The shift of the highly excited (i.e.,  $n \gg 1$ ) levels does not depend on  $n$  when  $\omega \gg 1/n^2$ , and is well approximated by the vibrational energy of the electron in the wave field  $\sim F^2/\omega^2$  (Ref. 4). It should be noted that the nonresonance Stark effect can determine not only the shift, but also the mixing of neighboring levels. However, as will be shown below (see Sec. 5), this mixing and the associated splitting are small on the scale of the energies ( $\sim \Gamma$  and  $1/n_0^3$ ) and times under consideration, and will therefore not be taken into consideration below.

With allowance for the assumptions made above, the equations for the coefficients  $A_{1,n,E}$  assume the form

$$i\dot{A}_1 = (E_1 + \omega)A_1 = \sum_{n=2}^{\infty} V_{n1}^* A_n + \int_0^{\infty} V_{E1} A_E dE, \quad (5)$$

$$i\dot{A}_n - E_n A_n = V_{n1} A_1, \quad i\dot{A}_E - E A_E = V_{E1} A_1.$$

We shall assume for simplicity that the field is switched on instantaneously at  $t=0$  and that at the initial moment of time the atom is in the ground state:  $A_1(0) = 1$ ,  $A_n(0) = A_E(0) = 0$ . In problems in which the interaction is switched on instantaneously, it is convenient to use the Laplace transformation<sup>5</sup> of the coefficients  $A_{1,n,E}(t)$  (see, for example, Ref. 6).

The system of equations for the Laplace transforms,  $\tilde{A}_{1,n,E}(p)$ , of the functions  $A_{1,n,E}(t)$  has the form

$$(ip - E_1 - \omega)\tilde{A}_1(p) - i = \sum_{n=2}^{\infty} V_{n1}^* \tilde{A}_n(p) + \int_0^{\infty} V_{E1}^* \tilde{A}_E(p) dE,$$

$$(ip - E_n)\tilde{A}_n(p) = \tilde{A}_1(p)V_{n1}, \quad n=2, 3, \dots,$$

$$(ip - E)\tilde{A}_E(p) = \tilde{A}_1(p)V_{E1}. \quad (6)$$

The solution of these equations is elementary. For the function  $\tilde{A}_1(p)$ , for example, we obtain

$$\tilde{A}_1(p) = i \left[ ip - E_1 - \omega - \sum_{n=2}^{\infty} \frac{|V_{n1}|^2}{ip - E_n} - \int_0^{\infty} \frac{|V_{E1}|^2}{ip - E} dE \right]^{-1}. \quad (7)$$

The probability amplitude  $A_1(t)$  is found with the aid of the inverse Laplace transformation:

$$A_1(t) = \frac{1}{2\pi i} \int_{\sigma - i\infty}^{\sigma + i\infty} \tilde{A}_1(p) e^{pt} dp, \quad (8)$$

where the integration contour in the complex  $p$  plane should lie to the right of all the singularities of the function  $\tilde{A}_1(p)$ .<sup>5</sup> In fact, as can easily be verified, all the singularities of  $\tilde{A}_1(p)$  lie on the imaginary axis, so that we should have  $\sigma > 0$  in the formula (8).

The integral entering into the denominator of the expression (7) is the Stieltjes transform of the function  $|V_{E1}|^2$  (Ref. 7), and has a branch cut along the imagin-

ary semiaxis,  $p'' \equiv \text{Im} p < 0$ , of the  $p$  plane. On account of this, the function  $\tilde{A}_1(p)$  also has a branch cut on the imaginary negative semiaxis, its values on the shores of the cut being given by the expressions

$$\tilde{A}_1^\pm(p) = \frac{-i}{p'' + E_1 + \omega \mp i\Gamma/2}, \quad p' = \text{Re } p = \pm 0. \quad (9)$$

We have dropped in the denominator of the formula (9) the slowly varying terms having no singularities in the region  $p'' < 0$ , and determining part of the Stark shift, which, by definition, is fully allowed for in  $E_1$ . The ionization width  $\Gamma$ , which is given by the formula (2), is a slowly varying function of the energy, and is therefore approximated by a constant (see Ref. 1).

The function  $\tilde{A}_1(p)$  has on the positive imaginary semiaxis  $p'' > 0$  poles whose locations are given by the zeros of the denominator in the formula (7):

$$p'' + E_1 + \omega - \sum_{n=2}^{\infty} \frac{|V_{n1}|^2}{p'' + E_n} - \int_0^{\infty} \frac{|V_{E1}|^2}{p'' + E} dE = 0. \quad (10)$$

The sum over  $n$  in this equation has simple poles at the points  $p'' = -E_n = 1/2n^2$ , while the integral term is a slowly varying function of  $p''$ . Therefore, the poles of the function  $\tilde{A}_1(p)$  in the region  $p'' > 0$ ,  $p' = 0$  are located between two neighboring values of the energy of the free atom (with the opposite sign):  $-E_{n+1} < p'' < -E_n$ .

The contour of the integration in the complex  $p$  plane in the formula (8) can be modified in accordance with Fig. 1. The function  $A_1(t)$  can, as a result, be represented in the form of a sum of the residues at the poles and integral taken along the shores of the branch cut:

$$A_1(t) = B_1(t) + B_2(t), \quad (11)$$

$$B_1(t) = \frac{1}{2\pi i} \int_{-\infty}^0 \left( \frac{1}{y + E_1 + \omega - i\Gamma/2} - \frac{1}{y + E_1 + \omega + i\Gamma/2} \right) e^{iyt} dy, \quad (12)$$

$$B_2(t) = \sum_n \text{res } \tilde{A}_1(p_n) \exp(p_n t), \quad (13)$$

where the index  $n$  numbers the poles of the function  $\tilde{A}_1(p)$ .

The functions  $B_1(t)$  and  $B_2(t)$  determine respectively the contribution to  $A_1(t)$  from the multiple transitions into the continuum and back and the contribution from the

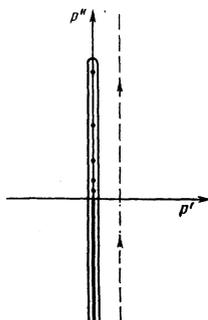


FIG. 1. Integration contour in the complex  $p$  plane. The dashed line is the original contour, the heavy straight line represents the branch cut, and the points indicate the poles of the function  $\tilde{A}_1(p)$ .

transitions involving the virtual excitation of discrete levels.

### 3. PERTURBATION THEORY

Before investigating the decay regime at large  $t$ , let us consider the case of short pulses or weak fields, i.e., the case when  $1 \ll t \ll \Gamma^{-1}$ . Under these conditions it is not difficult to find from the system of equations (5) with the aid of perturbation theory the probability for the excitation and ionization of the atom by the moment of time  $t$ :

$$w = 4 \sum_{n=2}^{\infty} \frac{|V_{n1}|^2 \sin^2((E_n - E_1 - \omega)t/2)}{(E_n - E_1 - \omega)^2} + \frac{2\Gamma}{\pi} \int_0^{\infty} \frac{\sin^2((E - E_1 - \omega)t/2)}{(E - E_1 - \omega)^2} dE. \quad (14)$$

In view of the fact that, when  $|E_1 + \omega| \ll 1$ , mainly the high-lying atomic levels with numbers  $n \gg 1$  get excited, the matrix elements  $|V_{n1}|^2$  can be written in the form  $|V_{n1}|^2 = \Gamma/2m^3$  (Ref. 8). The part of the sum over  $n$  in the formula (14) from  $n \sim t^{1/3} \gg 1$  to  $\infty$  can be transformed into an integral, similar to the second term in this expression, within the limits from  $(-\frac{1}{2}t^{-2/3})$  to zero. For  $E_1 + \omega \gg (-t^{-2/3})$ , the lower limit of the integral can be replaced by  $(-\infty)$ , which yields for the excitation and ionization probability the expression  $w = \Gamma t$ . The discarded part of the sum, i.e., the sum from  $n = 2$  to  $n \sim t^{1/3}$ , yields in this case a small contribution to  $w$  that is of the order of  $\Gamma t^{2/3} \ll \Gamma t$ . The obtained linear law of variation of the probability  $w(t)$  is similar to the usual perturbation-theory result for photo-ionization. The difference lies only in the conditions for the applicability of this formula. In the considered case we used as the "threshold" condition the inequality  $E_1 + \omega > -t^{-2/3}$  or  $n_0 > t^{1/3}$  instead of the inequality  $E_1 + \omega \geq 0$ . This means that the excitation of the high discrete levels lying above the "threshold"  $n_0 > t^{1/3} \gg 1$  occurs in the same way as the ionization, and this allows us to speak of an effective depression of the threshold by an amount  $\sim 1^{-2/3} \gg \Gamma$ . This result is qualitatively explained by the fact that, in the case of short, smooth pulses, the spectral width of the radiation,  $\Delta\omega \sim t^{-1}$ , in the "trans-threshold" region ( $n_0 > t^{1/3}$ ) exceeds the level spacing (i.e.,  $\Delta\omega > 1/n_0^3$ ) and, consequently, covers many discrete levels. Therefore, the excitation of these levels occurs in much the same way as the transitions into the continuum. In contrast, below the threshold ( $n_0 < t^{1/3}$ ),  $\Delta\omega < 1/n_0^3$ , i.e., the spectral width is less than the level spacing. The external radiation feels the discrete structure of the spectrum, and the excitation of the levels proceeds in the normal resonance fashion.

### 4. STRONG FIELD. TRANSITIONS FROM THE GROUND STATE INTO THE CONTINUUM AND BACK

Let us now proceed to the analysis of the decay regimes for the ground state in a strong field  $F(t)$  in the case when the interaction duration  $t$  is sufficiently long, assuming that  $\Gamma t \geq 1$ . Let us first consider the contribution to the amplitude,  $A_1(t)$ , of the probability of finding the atom in the ground state from the transitions between the ground state and the continuum (the function  $B_1(t)$  in

the formula (11)). As a result of simple mathematical transformations, the expression (12) for  $B_1(t)$  can be transformed to the form

$$B_1(t) = \frac{1}{2\pi i} \exp[-i(E_1 + \omega)t] \left\{ e^{-\Gamma t/2} \left[ \text{Ei} \left( \frac{\Gamma t}{2} + i(E_1 + \omega)t \right) + 2\pi i \theta(E_1 + \omega) \right] - e^{\Gamma t/2} \text{Ei} \left( -\frac{\Gamma t}{2} + i(E_1 + \omega)t \right) \right\}, \quad (15)$$

where  $\text{Ei}(x)$  is an exponential integral function<sup>9</sup>;  $\theta(x) = 1$  for  $x > 0$  and  $\theta(x) = 0$  for  $x < 0 \dots$

For a long interaction duration, or sufficiently far from the threshold, when  $|E_1 + \omega|t \gg 1$ , the expression (15) is significantly simplified:

$$B_1(t) = -\frac{i\Gamma}{2\pi t} \frac{1}{(E_1 + \omega)^2 + \Gamma^2/4} + \theta(E_1 + \omega) \exp \left[ -i(E_1 + \omega)t - \frac{\Gamma t}{2} \right]. \quad (16)$$

If then the contribution from the discrete levels can be neglected, i.e., if  $|B_2| \ll |B_1|$ , which is possible for large excesses over the threshold (Sec. 5), then  $A_1(t) \approx B_1(t)$ , and the formula (16) describes the decay of the ground state at large  $t$ . It can be seen from this that the conditions for the applicability of the exponential-decay model impose limitations not only on the excess over the threshold,  $E + \omega$ , but also on the interaction time  $t$ . The ionization process is characterized by an exponential dependence  $w(t)$  provided

$$\Gamma^{-1} \ln \frac{E_1 + \omega}{\Gamma} > t \gg \Gamma^{-1}.$$

If

$$t > \Gamma^{-1} \ln \frac{E_1 + \omega}{\Gamma} \gg \Gamma^{-1},$$

then the exponential decay is replaced by the more slowly varying power-law decay:

$$|A_1(t)|^2 \sim \Gamma^{-2} (E_1 + \omega)^{-2t}. \quad (17)$$

This result is due to the fact that the continuous electron spectrum is bounded from below, and agrees with the general theorems on the decay laws for physical systems,<sup>10,11</sup> which theorems forbid strictly exponential decay at large  $t$ . As will be shown below, the formulas (16) and (17) for  $E_1 + \omega \gg \Gamma$  remain valid also when the transitions into the highly-excited states of the discrete spectrum are taken into account (see Sec. 5). According to (16), when  $E_1 + \omega < 0$ ,  $|E_1 + \omega| \gg \Gamma$ , the contribution to  $A_1(t)$  of the transitions into the continuum decreases, and can be neglected (cf. formulas (28) and (29) of Sec. 5).

In the region close to the edge of the continuum spectrum, i.e., for  $|E_1 + \omega| \ll \Gamma$ , the asymptotic representation of the function  $B_1(t)$  for  $\Gamma t \gg 1$  has the form

$$B_1(t) = -2i/\pi \Gamma t. \quad (18)$$

This result also corresponds to a power-law decay of the ground state:  $|B_1(t)|^2 \sim (\Gamma t)^{-2}$ . However, we cannot in this near-threshold region neglect the contribution to  $A_1(t)$  from the discrete levels. This contribution is determined by the function  $B_2(t)$ , (13), which we now proceed to investigate.

## 5. STRONG FIELD. TRANSITIONS BETWEEN THE STATES OF THE DISCRETE SPECTRUM

To compute  $B_2(t)$ , it is necessary to find the positions of the poles of the function  $\tilde{A}_1(p)$ , which are the solutions to Eq. (10), and compute the residues at these points.

On account of the fact that  $|V_{E_1}|^2$  is a slowly varying function, the integral term in Eq. (10) is easily estimated:

$$\int_0^\infty \frac{|V_{E_1}|^2}{p'' + E} dE \approx \frac{\Gamma}{2\pi} \ln \frac{b}{p''}, \quad (19)$$

where  $b = \text{const} \sim 1$ .

It is convenient to separate out from the sum over  $n$  in Eq. (10) the terms corresponding to  $E_n$ -energy values that are closest to  $(-p'')$ . The entire remaining part of the sum can, on account of the fact that  $p'' \ll 1$  and  $n \gg 1$ , be transformed into an integral, and is also easy to estimate. As a result, it is easy to verify that the contribution from the remote terms in the sum over  $n$  exactly cancels out the logarithmic contribution from the integral term, (19). This means that the  $dE$  integral in Eq. (10) can, for all practical purposes, be dropped, and that it is sufficient to retain in the sum over  $n$  one or two terms corresponding to the values of the energy  $E_n$  that are closest to  $(-p'')$ .

The solutions in Eq. (10), the form of the function  $B_2(t)$ , and the nature of the excitation process essentially depend on the relation between the parameters  $\Gamma$  and  $E_1 + \omega$ , or  $\Gamma$  and  $n_0$ . Let us consider a number of the most typical cases, beginning with a relatively low value of the frequency  $\omega$  and gradually increasing it with the object of following all the distinctive features that arise as the threshold region is crossed.

1. First, let  $\Gamma n_0^2 \ll \Gamma n_0^3 \ll 1$ , i.e., let  $E_1 + \omega < 0$  and  $|E_1 + \omega| \gg \Gamma$ , and let the level spacing  $\delta E \sim 1/n_0^3$  in the vicinity of the value of  $E_1 + \omega$  be large compared to  $\Gamma$ . This is a resonance case, when the  $E_1$  and  $E_{n_0}$  levels are strongly coupled to each other through the interaction with the field and form a two-level system.<sup>12</sup> The resonance condition is  $|\beta| \ll 1$ , where the quantity  $\beta$  is given by the formula (3), and plays the role of a detuning. The roots of Eq. (10) are close to  $-E_{n_0} = 1/2n_0^2$ . To find them, it is sufficient to retain in the sum over  $n$  in (10) only the term with  $n = n_0$ . Solving the resulting quadratic equation, and verifying that the contribution of the remaining poles is small, we represent the function  $A_1(t)$  in the form

$$A_1(t) = \exp \left[ i \left( \frac{\varepsilon}{2} - E_1 - \omega \right) t \right] \left( \cos \Omega t - \frac{i\varepsilon}{2\Omega} \sin \Omega t \right), \quad (20)$$

where

$$\varepsilon = \frac{\beta}{n_0}, \quad \Omega = \left( \frac{\varepsilon^2}{4} + \frac{\Gamma}{2\pi n_0^3} \right)^{1/2} = \left( \frac{\varepsilon^2}{4} + |V_{n_0}|^2 \right)^{1/2}$$

is the Rabi frequency of a two-level system.

Thus, the system oscillates between two levels, and, in the resonance approximation, transitions into other states do not occur, i.e., the ground state does not decay. At exact resonance ( $|\varepsilon| \ll \Omega$ ,  $|\beta| \ll (\Gamma n_0^3)^{1/2}$ )  $A_1$

$\sim \cos \omega t$ ; the system goes over completely from the lower level into the upper level and back:  $|A_1(t)|^2 = \frac{1}{2}$ . The decay of the atom in this case is due to ionization from the resonance state  $n_0$ , and occurs within a time  $\sim n_0^3/\Gamma \gg 1/\Omega$  (see Refs. 1, 13, and 14).

Let us now proceed to the region of higher frequencies, where  $\Gamma n_0^3 \gg 1$  (but, as before,  $E_1 + \omega < 0$ ). This condition implies that the ionization width covers many levels in the vicinity of the value of the energy  $E_1 + \omega$ . For this reason, the excitation of the atom can no longer be described within the framework of the two-level model, or, in other words, the residues of many poles contribute to the functions  $B_2(t)$  and  $A_1(t)$ .

In the general case it is not possible to solve Eq. (10) and exactly find the positions of all the poles. For a qualitative description, however, it turns out to be sufficient to divide the entire region of summation over  $n$  in the formula (13) into two principal subregions characterized by two types of roots of Eq. (10): I) poles close to the values of the ionization energy,  $-E_n$ , of the free atom and II) poles separated from the nearest  $-E_n$  value by distances of the order of the distance to the neighboring level,  $E_{n+1} - E_n = 1/n^3$ . To find the type-I roots of Eq. (10), we retain in the sum over  $n$  a single term and seek corrections to the value  $p_n'' = -E_n = 1/2n^2$  with the aid of perturbation theory, which yields

$$p_n'' = \frac{1}{2n^2} + \frac{\Gamma/2\pi n^2}{E_1 + \omega + 1/2n^2}, \quad p_n' = 0. \quad (21)$$

This result is valid provided

$$n^2 \left| p_n'' - \frac{1}{2n^2} \right| = \frac{\Gamma/2\pi}{E_1 + \omega + 1/2n^2} \ll 1. \quad (22)$$

The residue of the function  $\tilde{A}_1(p)$  at these poles is equal to

$$\text{res } \tilde{A}_1(p_n) = \left[ 1 + \frac{2\pi n^2}{\Gamma} \left( E_1 + \omega + \frac{1}{2n^2} \right)^{-1} \right]^{-1}. \quad (23)$$

The positions of the poles that do not satisfy the condition (22) are determined by the type-II roots of Eq. (10):

$$p_n'' = \frac{1}{2n^2} + \frac{\alpha_1}{n^2}, \quad p_n' = 0. \quad (24)$$

The residue at these poles is, in order of magnitude, equal to

$$\text{res } \tilde{A}_1(p_n) = \alpha_2/\Gamma n^2. \quad (25)$$

The constants  $\alpha_{1,2}$  in the formulas (24) and (25) are determined up to a factor of the order of unity ( $\alpha_1 \sim 1$ ,  $\alpha_2 \sim 1$ ). The splitting of the region of summation over  $n$  into subregions of the types I and II also constitutes a fairly crude procedure. Needless to say, there exist intermediate parameter ranges in which the transition from (21) to (24) occurs. Nevertheless, such a computational procedure turns out to be sufficient for a qualitative description of the excitation processes. Here the theory allows us to predict all the dependences on the principal parameters of the problem, but makes no pretensions to the correct determination of the numerical coefficients. In all the formulas that follow, as well as in (24) and (25), the coefficients have been determined up to factors of the order of unity. In view of this, below we shall of-

ten not write out the numerical coefficients explicitly, assuming them to be equal to unity.

The specific splitting into subregions of the types I and II and the specific results depend on the relation between the physical parameters  $\Gamma$  and  $E_1 + \omega$ , i.e., on the parameter  $\Gamma n_0^2$ . Let us consider the cases  $\Gamma n_0^2 \ll 1$  and  $\Gamma n_0^2 \gg 1$ , one after the other in order of increasing frequency (and  $n_0$ ).

2. Let  $\Gamma n_0^3 \gg 1 \gg \Gamma n_0^2$ , i.e., let  $E_1 + \omega < 0$ ,  $|E_1 + \omega| \gg \Gamma$ , but  $\Gamma \gg \delta E_{n_0} \sim 1/n_0^3$ . The splitting of the summation region into subregions of the types I and II yields

$$B_2(t) = \sum_{n=2}^{C_1 n_0} \Gamma n e^{i/2n^2} + \sum_{n=n_0-\Delta n}^{n_0+\Delta n} \frac{1}{\Gamma n^2} e^{i/2n^2} + \sum_{n=C_2 n_0}^{\infty} \frac{\Gamma n_0^4}{n^2} e^{i/2n^2} + \Gamma n_0^3 \left( \sum_{n=C_1 n_0}^{n_0-\Delta n} \frac{1}{(n-n_0)^2} e^{i/2n^2} + \sum_{n=n_0+\Delta n}^{C_2 n_0} \frac{1}{(n-n_0)^2} e^{i/2n^2} \right), \quad (26)$$

where  $\Delta n \equiv \Gamma n_0^3 \gg 1$ ,  $\Delta n \ll n_0$ ,  $C_{1,2} \sim 1$ , with  $C_1 < 1$  and  $C_2 > 1$ . Those parts of the sums entering into this expression whose term numbers  $n > t^{1/3}$  can be replaced by integrals. As to the remaining parts of the sums ( $n < t^{1/3}$ ), we can estimate their contribution to the mean probability  $|A_1|^2$  by replacing

$$\left| \sum_{n=2}^{t^{1/3}} a_n \exp\left(\frac{it}{2n^2}\right) \right|^2$$

by  $\sum |a_n|^2$ , after which we can again go over from summation to integration. Such a replacement of the square of a sum by a sum of squares corresponds to averaging over rapid oscillations, and allows the description of the slow variations of the mean probability  $w_1(t) \equiv |A_1(t)|^2$ . Using this procedure, and evaluating the integrals for  $n_0^3 \gg t \gg n_0^2$ , we obtain from (26) the expression

$$w_1(t) = \Gamma^2 t + \frac{1}{\Gamma^2 t^2}, \quad (27)$$

where we have, in accord with the remark made earlier, dropped the coefficients of the order of unity.

For  $t \sim n_0^3$ , but  $t^{1/3} < n_0 - \Gamma n_0^3$ , instead of the formula (27), we find in similar fashion the formula

$$w_1(t) = \frac{1}{\Gamma^2 t^2} + \frac{\Gamma^2 n_0^4}{(n_0 - t^{1/3})^3}. \quad (28)$$

Finally, for  $t^{1/3} > n_0 - \Gamma n_0^3$  (and, in particular, for  $t > n_0^3$ ) we have

$$w_1 = w_{1, \text{min}} = |E_1 + \omega|^{1/2} / \Gamma = \text{const} \ll 1. \quad (29)$$

It is easy to verify that the formulas (27)–(29) join each other at the boundaries of the regions of their applicability. The qualitative form of the dependence  $w_1(t)$  is shown in Fig. 2. The minimum value of this function can be determined from either the formula (27), or the formula (28). Thus, for  $\Gamma^{4/3} n_0^3 > 1$  (which is not at variance with all the other assumptions made), the behavior of  $w_1(t)$  in the vicinity of the minimum is described by the formula (27). The value of  $t$  at which  $w_1(t)$  is a minimum is  $t_1 = \Gamma^{-4/3}$ , with  $w_{1, \text{min}} = \Gamma^{2/3}$ .

The function  $w_1(t)$  is nonmonotonic. The initial decrease of the probability  $w_1(t)$  is due to the nonexponential decay of the ground state (according to the law  $1/t^2$ ). The increase of  $w_1(t)$  is due to the discrete structure of

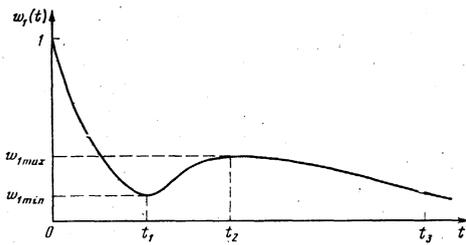


FIG. 2. Qualitative shape of the time dependence of the mean probability,  $w_1$ , for finding an atom in the ground state.

the excited levels, and reflects the partial reversibility of the decay process. Also connected with this is the appearance of the quasistationary regime with a nonzero "residual" probability, given by the formula (29), for finding the atom in the ground state. The transition to this regime in the case under consideration occurs at the moment  $t_2 \sim n_0^3$ . The appearance of the residual probability is in accord with the Fock-Krylov theorem, according to which a system with a discrete spectrum does not decay to the end.<sup>10,11,15</sup> This result is, however, valid only within the framework of the model used. When allowance is made for the two-photon transitions into high-lying states of the continuum, the quasienergy spectrum of the system becomes continuous, and complete decay of the ground state (i.e., complete ionization of the atom) occurs. It is easy to estimate the times at which the two-photon transitions become important. The probability of such a transition is determined by the two-photon composite resonance matrix element

$$V_{iE}^{(2)} \sim \sum_n \frac{V_{in} V_{nE}}{E_1 + \omega - E_n}, \quad E \approx E_1 + 2\omega.$$

Here it is necessary to take into account the above considered resonance mixing of the levels, which gives rise to some effective width,  $\gamma$ , of the resonance denominator. The number of levels that make a substantial contribution to the sum over  $n$  is, in order of magnitude, clearly equal to  $\gamma n^3$ . Allowing also for the fact that  $V_{in} \sim V_{nE} \sim (\Gamma/n^3)^{1/2}$ , we obtain for  $V_{iE}^{(2)}$  the estimate  $V_{iE}^{(2)} \sim \Gamma$ . The time for the complete ionization of the atom on account of the two-photon transition is

$$t_3 \sim 1/|V_{iE}^{(2)}|^2 \sim 1/\Gamma^2 \gg t_2. \quad (30)$$

This estimate for the complete-ionization time does not depend on the position of the resonance with respect to the continuum edge, and is also valid in all cases considered above.

Generally speaking, at large times, besides the resonance two-photon transitions, it may also be necessary to take into account the nonresonance two-photon mixing of the levels. The mixing of neighboring, highly-excited, discrete levels is described by the nonresonance composite matrix elements  $V_{nm}^{(2)}$ . At frequencies significantly exceeding the ionization potential of the excited levels (i.e., for  $\omega \gg 1/n_0^2$ ), the diagonal part,  $V_{nn}^{(2)}$ , of the matrix elements corresponds, according to Ref. 4, to a constant shift of the levels by an amount equal to the vibrational energy of the electron in the wave field ( $\sim F^2/\omega^2$ ). The off-diagonal (with respect to the principal and orbital quantum numbers) matrix elements determine the mixing of the near-degenerate, highly-excited

levels. The magnitude of the off-diagonal matrix elements can easily be estimated:  $V_{nm}^{(2)} \sim V_{n1} V_{1m} \sim \Gamma/(\gamma n^3)^{3/2} \sim \Gamma n_0^3$  (Ref. 8), which is considerably less than the characteristic energy scales of the problem,  $\Gamma$  or  $1/n_0^3$ . The discrete-level mixing time is  $n_0^3/\Gamma \gg t_3$ , and, consequently, this effect can be neglected on the time scale under consideration.

Similarly, we can estimate the time required for the mixing of the highly-excited discrete levels with the neighboring continuum states. For this purpose, it is necessary to add to the second of the equations (5) the term

$$\int V_{nE}^{(2)} A_E dE, \quad (31)$$

where  $V_{nE}^{(2)}$  is the nonresonance composite matrix element:  $V_{nE}^{(2)} \sim V_{n1} V_{1E}$ . Estimating the quantity  $A_E$  with the aid of the third of the equations (5), and comparing the nonresonance term (31) with the resonance term  $V_{n1} A_1$ , we find the time at which this mixing effect becomes important:

$$t \sim V_{n1}/V_{E1} V_{nE}^{(2)} \Gamma \sim 1/\Gamma^2,$$

where we have assumed that the range of energy values that contributes substantially to the integral (31) is, in order of magnitude, equal to  $\Gamma$ . The obtained estimate shows that the mixing of the discrete levels with the states of the continuum becomes important at the same times at which complete ionization of the atom occurs as a result of real two-photon transitions:  $t \sim t_3$ . Therefore, within the framework of the considered model, this effect can also be neglected.

Thus, on the whole, we can distinguish four phases in the ground-state decay process: 1) an initial decay in the single-photon transition model; 2) a partial return to the ground state; 3) a quasistationary regime characterized by some nonzero constant mean probability  $w_{1max}$ ; 4) an irreversible decay (ionization) on account of two-photon processes. Qualitatively, this picture remains valid in the entire near-threshold region (provided many levels are excited at once). Only the quantitative values of the parameters determining the limits of the various regions change.

Let us now consider the region of still higher values of the frequency  $\omega$ .

3.  $|E_1 + \omega| \ll \Gamma$ , and  $E_1 + \omega$  can have any sign. For  $E_1 + \omega < 0$ , the condition  $|E_1 + \omega| \ll \Gamma$  implies that  $\Gamma n_0^3 \gg \Gamma n_0^2 \gg 1$ . All the computations and the qualitative nature of the dependence  $w_1(t)$  are similar to the preceding case. The formula (27) also remains valid, it remaining unchanged when allowance is made for the transitions, described by Eq. (18), into the continuum. Only the parameters  $t_2$  and  $w_{1max}$  change quantitatively, and are now equal to  $t_2 = \Gamma^{-3/2}$ ,  $w_{1max} = \Gamma^{1/2}$ . An existence domain does not exist for a formula of the type (28) in the case under consideration: the transition to the quasistationary regime occurs directly from the formula (27). A comparison with the preceding case shows that the non-monotonic nature of the variation of the probability  $w_1(t)$  gets smoothed over as the frequency increases:  $w_{1max}$  decreases, while  $w_{1min}$  does not change.

4. Finally, in the case of a large excess over the threshold, i.e., for  $E_1 + \omega \gg \Gamma$  (but  $E_1 + \omega \ll 1$ ), the principal relationships are, as before, similar to those considered above. For  $(E_1 + \omega)^{-1} \ll t \ll (E_1 + \omega)^{-3/2}$ ,

$$w_1(t) = \Gamma^2 t + \Gamma^2 / (E_1 + \omega)^4 t^2 + e^{-t}, \quad (32)$$

where we have taken into account both the transitions into the continuum, (16), and the transitions into the states of the discrete spectrum (which are responsible for the first term in the formula (32)).

The transition from exponential to power-law decay occurs at  $t \geq t_0$ , where

$$t_0 = \frac{1}{\Gamma} \ln \frac{E_1 + \omega}{\Gamma}. \quad (33)$$

The moment of time  $t_1$  (Fig. 2) and the minimum value of the function  $w_1(t)$  are equal to

$$t_1 = (E_1 + \omega)^{-1/2}, \quad w_{1, \min} = \Gamma^2 / (E_1 + \omega)^{3/2}. \quad (34)$$

The transition to the quasistationary regime occurs at  $t \sim t_2$ , with

$$t_2 = (E_1 + \omega)^{-3/2}, \quad w_{1, \max} = \Gamma^2 / (E_1 + \omega)^{3/2}. \quad (35)$$

The complete decay of the system occurs at  $t \geq t_3$ , where  $t_3$  is determined as before by the formula (30). As is easy to see, we can have  $t_0 > t_2$  in the case of a sufficiently large excess over the threshold. There is then no region in which the function  $w_1(t)$  is nonmonotonic. A direct transition from the exponential decay in the single-photon ionization model<sup>1</sup> to the quasistationary regime characterized by the residual probability  $w_{1, \max}$  occurs as  $t$  increases.

As the excess over the threshold increases, the quantity  $w_{1, \max}$  decreases.

## 6. THE MAIN CONCLUSIONS

In conclusion, let us briefly state the principal results of the investigation.

The analysis performed allows us to follow the step-by-step transition from the resonance excitation of the discrete levels in a two-level system to the irreversible process of ionization related to transitions into the continuum. We have shown that, in the intermediate region, where the interaction encompasses many discrete levels and part of the continuum, the ground-state decay probability is, on the whole, irreversible, but bears traces of transitions into the discrete spectrum. The probability, averaged over the rapid oscillations, for finding the atom in the ground state depends nonmonotonically on time. A partial return of the atom into the ground state occurs at some stage. We have shown that, as the light frequency,  $\omega$ , increases, this partial reversibility and the nonmonotonic character get smoothed over, and disappear ultimately.

An interesting distinctive feature of the decay of the ground state in the near-threshold region is its nonexponential character: the probability for finding the atom in the ground state decreases like  $1/t^2$ . Another inter-

esting feature of the excitation and ionization of atoms is the appearance in some time interval of a quasistationary regime characterized by a nonzero constant mean probability for finding the atom in the ground state.

In view of the great similarity between the processes of ionization and excitation of high-lying discrete levels, we can speak of an effective depression of the threshold for these decay processes. In the case of a weak field and a short pulse length ( $\Gamma t \ll 1$ ), the threshold is depressed by an amount  $\sim t^{-2/3}$ ; in the opposite case of a strong field ( $\Gamma t \gg 1$ ), by an amount  $\sim \Gamma^{2/3} \sim F^{4/3}$ . Qualitatively, this result follows from the argument that the excitation decay regime is possible until the spectral width of the radiation  $\Delta \omega \sim 1/t$ , or the interaction energy, determined by the ionization width  $\Gamma$ , exceeds the spacing,  $1/n_0^3$ , of the highly excited levels. It is easy to see that in both cases the threshold is depressed by an amount significantly greater than the Stark shift (equal to the electron vibration energy,  $F^2/\omega^2 \sim F^2 \sim \Gamma$ ) of the highly excited levels of the atom.

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