Threshold singularities of the photodetachment of an electron from a negative ion by a strong electromagnetic field

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(Submitted 13 July 1982)
Zh. Eksp. Teor. Fiz. **83**, 2035–2047 (December 1982)

A solution is obtained for the problem of threshold singularities of the photodetachment of an electron from a negative ion at a finite pulse duration, with account taken of the level shift due to the dynamic Stark effect, both for steplike pulses and for pulses with smooth envelopes. It is shown that in the latter case qualitatively new singularities can arise, namely, the ionization probability vanishes below the ionization threshold at a sufficiently long pulse duration, and in the entire near-threshold region the ionization probability saturates in a definite range of parameters, i.e., becomes independent of the external field intensity. Possible ionization regimes are investigated and the explicit form of the electron-photodetachment probability is obtained for all cases.

PACS numbers: 32.80.Fb, 32.60. + i

1. INTRODUCTION

It is well known that at a sufficiently large excess of a photoelectron energy above the photoionization threshold, the dependence of the ionization probability $w_i(t)$ on the time t has a simple exponential character, $w_i = 1 - \exp(-\Gamma_i t)$, where Γ_i is the ionization width of the ground state of the atom and is equal to the probability of its ionization per unit time. The decay of the atom does not have an exponential character near the ionization threshold.[1]

The spectrum of the neutral atoms is known to contain a system of discrete levels that condense as the threshold is approached. The presence of these subthreshold levels is quite important for the threshold singularities of photoionization of atoms.[1] In negative ions, on the contrary, there is only one discrete level as a rule.[2] Another difference of negative ions from neutral atoms is that in negative ions the matrix element of the transition into the continuum vanishes at the threshold, i.e., at zero photoelectron energy $E = 0$; $|v_{E_0}|^2 \propto E^{1/2}$ or $|v_{E_0}|^2 \propto E^{3/2}$, depending on whether the ground state is a P or S state (in neutral atoms, $|v_{E_0}|^2 = \text{const}$ as $E \to 0$).

The indicated difference between neutral atoms and negative ions requires an independent study of the threshold singularities of the photodetachment of an electron from a negative ion as a function of the frequency ω and of the external-field intensity $\mathscr{E}_0$, as well as of the pulse duration τ and of the shape of its envelope $\mathscr{E}_0(t)$. For pulses with smooth envelopes, a substantial role can be played by the time-dependent shift $\delta E_0(t)$ of the level E_0 of the ground state in an external high-frequency field.

The foregoing problems were considered to some degree in the literature.[3,4] Kumekov and Perel',[3] however, did not investigate[1)] the character of the decay of a negative ion and determined correctly only the asymptotic residual probability at infinite pulse duration in a model in which the interaction is turned on instantaneously (see Sec. 3 below). A nonexponential decay law $w \propto \tau^{-3}$, which is one of the many possible decay regimes considered below, was obtained in the instantaneous-switching model in Ref. 4, which contains also some results of numerical calculations. The results of Refs. 3 and 4 are in no way exhaustive, particularly when models more or less close to reality are considered with a smooth envelope of the radiation pulse. We shall investigate below decay regimes of negative ions in the near-threshold region, which are realized in the model of instantaneous application of the interaction (Secs. 2 and 3) and in the case of pulses with smooth envelopes (Secs. 4 and 5).

2. GENERAL EQUATIONS. INSTANTANEOUS APPLICATION OF INTERACTION

Thus, we consider a quantum system having one discrete level and a continuous spectrum, and interacting with an electromagnetic field $\mathscr{E} = \mathscr{E}_0 f(t)\cos\omega t$, where $f(t)$ is the interaction switch-on function. Let the field frequency be close to the ionization threshold $\omega \approx -E_0$, where $-E_0$ is the binding energy of the electron in the negative ion and $\hbar = 1$. We assume that the field intensity E_0 is much lower than the characteristic value $\mathscr{E}_c$ which is the analog of the intra-atomic field:

$$\mathscr{E}_0 < \mathscr{E}_c \sim |E_0|/e\langle r\rangle \sim 10^8 \text{ V/cm}, \tag{1}$$

where $\langle r\rangle$ is the average dimension of the negative ion.[2]

By virtue of the condition (1), both the shift δE_0 of the level E_0 in the field $\mathscr{E}$ and its ionization width Γ_i are small compared with $|E_0|$. The condition (1) makes it also possible to neglect transitions $E \leftrightarrow E'$ (Ref. 5).

The wave function of a negative ion in a field $\mathscr{E}$ has the form of the superposition

$$\Psi(t) = C_0(t)\varphi_0 + \int_0^\infty dE C_E(t)\varphi_E, \tag{2}$$

where φ_{0E} are the unperturbed stationary wave functions of the negative ion, and the coefficients $C_{0E}(t)$ satisfy the equations

$$i\dot{C}_0=E_0C_0+2f(t)\cos\omega t\int_0^\infty dE\, v_{0E}C_E,$$
$$i\dot{C}_E=EC_E+2f(t)\,v_{E0}C_0\cos\omega t. \tag{3}$$

Here $v=-(1/2)\mathbf{d}\cdot\mathscr{E}_0$, $\mathbf{d}$ is the dipole moment of the system,

$$C_0(t\to-\infty)=e^{-iE_0t},\quad C_E(-\infty)=0. \tag{4}$$

In the model of instantaneous switching of the interaction $f(t)=0$ at $t<0$ and $t>\tau$, and $f(t)=1$ at $0<t<\tau$. The interaction energy of the negative ion with the field consists of two parts, which can be arbitrary called "resonant" and "nonresonant." The nonresonant terms in the first and second equations of the system (3) are proportional respectively to $\exp(-i\omega t)$ and $\exp(e\omega t)$. These terms contribute to the small rapidly oscillating components of the amplitude $C_0\exp(iE_0t)$, which we shall neglect, and also determine that part of the time-independent (in the case of instantaneous switching) Stark shift of the ground level, which by assumption is taken into account from the very beginning in the notation used for the energy of the discrete level E_0. In terms of these approximations, Eqs. (3) take the form

$$i\dot{C}_0=E_0C_0+\int_0^\infty dE v_{0E}\tilde{C}_E,\qquad i\dot{\tilde{C}}_E=(E-\omega)\tilde{C}_E+v_{E0}C_0, \tag{5}$$

where $\tilde{C}_E=e^{-i\omega t}C_E$.

The system (5) can be solved both directly with the aid of a Laplace transformation and by the method of quasienergy functions (or by the Fano method), used earlier in Ref. 5. We represent a brief description of the solution of the problem on the basis of the second approach, without assuming (in contrast to Ref. 5) a large excess above threshold.

The quasienergy (stationary) solutions of Eqs. (5) are sought in the form

$C_0,\ \tilde{C}_E\propto e^{-i\gamma t}$,

where γ is the quasienergy. At $\gamma+\omega>0$ these solutions take the form

$$\psi_\gamma=e^{-i\gamma t}|v_{\gamma+\omega,0}|^{-2}(\pi^2+z_\gamma^2)^{-1/2}$$
$$\times\left\{\varphi_0+e^{-i\omega t}\int_0^\infty dEv_{E0}\varphi_E[P/(\gamma-E+\omega)+z_\gamma\delta(\gamma-E+\omega)]\right\},$$
$$z_\gamma=|v_{\gamma+\omega,0}|^{-2}\left[\gamma-E_0-\int_0^\infty dE\frac{|v_{E0}|^2}{\gamma-E+\omega}\right]. \tag{6}$$

The spectrum of the quasienergy states (6) is continuous.

At $\gamma+\omega<0$ the system (5) has one solution that determines the quasienergy wave function of the bound state, normalized to unity:

$$\psi_0=e^{-i\gamma_0 t}\left[1+\int_0^\infty dE\frac{|v_{E0}|^2}{(\gamma_0-E+\omega)^2}\right]^{-1/2}$$
$$\times\left\{\varphi_0+e^{-i\omega t}\int_0^\infty dEv_{E0}\varphi_E(\gamma_0-E+\omega)^{-1}\right\}, \tag{7}$$

where the quasienergy γ_0 of the discrete level should be obtained from the equation

$$\gamma_0-E_0=\int_0^\infty dE\frac{|v_{E0}|^2}{\gamma_0-E+\omega}\approx-\int_0^\infty dE\frac{|v_{E0}|^2}{E}\equiv\Delta-E_0-\omega \tag{8}$$

under the condition $\Delta<0$.

Accurate to small corrections $\sim|E_0+\omega|/E_0$, the difference between γ_0 and E_0 is determined by that part of the quadratic dynamic Stark effect which is connected with the resonant part of the interaction energy. Taking this contribution into account, γ_0 is the true value of the ground-state energy, shifted relative to its unperturbed energy by an amount $-(1/4)\alpha(\omega)\mathscr{E}_0^2$, where $\alpha(\omega)$ is the dynamic polarizability of the level E_0 at the field frequency ω.

At $\Delta<0$ the system of functions (ψ_0,ψ_γ) is complete in the basis (φ_0,φ_E), while at $\Delta>0$ the system (6) of functions ψ_γ is complete. Using the completeness condition, we can obtain the time-dependent wave function $\Psi(t)$ of the system and with its aid, say the probability amplitude of finding a negative ion in the ground state $A_0(\tau)$ after turning off the field at $t=\tau$. At $\Delta<0$, the form of $A_0(\tau)$ is

$$A_0(\tau)=e^{-i(\gamma_0+\omega)\tau}\left[1+\int_0^\infty dE\frac{|v_{E0}|^2}{(\gamma_0-E+\omega)^2}\right]^{-1}$$
$$+\int_0^\infty dEe^{-iE\tau}|v_{E0}|^2\left[\pi^2|v_{E0}|^4+\left(E-E_0-\omega-\int_0^\infty dE'\frac{|v_{E'0}|^2}{E-E'}\right)^2\right]^{-1}. \tag{9}$$

The second term in (9) is connected with the contribution of the quasienergy wave functions ψ_γ(6) of the continuous spectrum. In the general case the contribution of the integral term in (9) decrease with increasing τ. As $\tau\to\infty$ the probability amplitude $A_0(\tau)$ is determined by only the first term in (9), and this agrees with the result of Ref. 3. At finite τ, however, the contribution of the integral term is not small and it is this term which determines the character of the system decay. We present below results that follow from Eq. (9) and determine the different realized ionization regimes. At $\Delta>0$ the first term in (9) should be left out, for in this case no bound quasienergy state is produced.

3. IONIZATION REGIMES IN THE INSTANTANEOUS-APPLICATION MODEL

Under the condition (1) and when ω differs little from $-E_0$, $|E_0+\omega|\ll|E_0|$, the main contribution to the integrals with respect to energy in (9) is made by the region of small E, $E\ll|E_0|$. This enables us to use in place of the function $|v_{E_0}|^2$ its approximates expression as $E\to0$, namely $|v_{E_0}|^2=\beta E^{1/2}$ or $|v_{E_0}|^2=\beta E^{3/2}$, where β is a constant. We consider these two cases separately.

1. Ionization from P states $|vE_0|^2=\beta E^{1/2}$

We obtain first a more exact solution of Eq. (8) for γ_0. Using, e.g., different model functions $|v_{E_0}|^2$ with a correct asymptotic form $|v_{E_0}|^2$ as $E\to0$, we easily verify that independently of the model,

$$\int_0^\infty\frac{dE|v_{E_0}|^2}{E-\gamma_0-\omega}\approx\int_0^\infty\frac{dE|v_{E_0}|^2}{E}-\pi\beta|\gamma_0+\omega|^{1/2}. \tag{10}$$

After substitution in the first of Eqs. (8), this yields

$$\gamma_0+\omega=-{}^1/_2\pi^2\beta^2-|\Delta|+{}^1/_2\pi\beta(\pi^2\beta^2+4|\Delta|)^{1/2}, \tag{11}$$

where Δ is determined by the last equation in (8).

The contribution of a discrete quasienergy level to the probability amplitude $A_0(\tau)$ (9) is determined by the integral

$$\int_0^\infty \frac{dE|v_{E_0}|^2}{(E-\gamma_0-\omega)^2}\approx\frac{\pi\beta}{2|\gamma_0+\omega|^{1/2}} \tag{12}$$

and its value, with allowance for (11), is (at $\Delta<0$)

$$A_0^{(1)}=e^{-i\Delta\tau}(1-\pi\beta/(\pi^2\beta^2+4|\Delta|)^{1/2}). \tag{13}$$

At $\Delta>0$ we have $A_0^{(1)}=0$.

The contribution of the continuum of quasienergy states to $A_0(\tau)$ is determined by the integral

$$A_0^{(2)}=\beta\int_0^\infty \frac{dEe^{-iE\tau}E^{1/2}}{\pi^2\beta^2E+(E-\Delta)^2}. \tag{14}$$

The substitution $E=x^2$ and expansion of the integrand in partial fractions make it possible to express (14) in terms of tabulated integrals and express $A_0^{(2)}(\tau)$ in terms of a Fresnel integral $\Phi(u)$ (Ref. 6):

$$A_0^{(2)}=(\pi^2\beta^2-4\Delta)^{-1/2}\{x_1e^{i\tau x_1^2}(1-\Phi(\sqrt{i\tau}x_1)) -x_2e^{i\tau x_2^2}(1-\Phi(\sqrt{i\tau}x_2))\}, \tag{15}$$

$$x_1={}^1/_2[\pi\beta+(\pi^2\beta^2-4\Delta)^{1/2}],\quad x_2={}^1/_2\,\mathrm{sign}\,\Delta\,[\pi\beta-(\pi^2\beta^2-4\Delta)^{1/2}]. \tag{16}$$

Expression (15) for the probability amplitude $A_0^{(2)}(\tau)$ becomes simpler in limiting cases when the arguments of the Fresnel integrals are small or large, so that one can use for $\Phi(u)$ either a series expansion at $|u|\ll1$, or an asymptotic representation at $|u|\gg1$ can be used directly if $|\arg u|<\pi/2$. This condition is satisfied for both Fresnel integrals in (15), so long as $\Delta<(1/2)\pi^2\beta^2$. At $\Delta>(1/2)\pi^2\beta^2$ we have $\arg(\sqrt{i}x_1)>\pi/2$. In this case it is necessary to use for the first Fresnel integral $\Phi(\sqrt{i\tau}x_1)$ in (15) the relation $\Phi(u)=-\Phi(-u)$, which transfers its arguments to the right-hand half-plane of the complex variable u. After this one can use again the usual asymptotic formulas for $\Phi(u)$ at $u\gg u$, $|\arg|u<\pi/2$ (Ref. 6).

The total probability amplitude $A_0(\tau)$ is determined by the sum of $A_0^{(2)}$ (13) and $A_0^{(2)}$ (15) at $\Delta<0$, and coincides with $A_0^{(2)}$ at $\Delta>0$. We present approximate expressions for $A_0(\tau)$ and $w_0(\tau)=|A_0(\tau)|^2$, which follow from (13) and (15) at different relations between the parameters Δ, $\pi^2\beta^2$, and τ^{-1}.

1) Let first $\Delta<(1/4)\pi^2\beta^2$, i.e., let the radiation frequency by quite close to the threshold. In this case the following ionization regimes are possible:

(a) Short pulse duration:

$$\tau\ll(\pi\beta)^{-2}\ll\pi^2\beta^2/\Delta^2. \tag{17}$$

In this case $\sqrt{\tau}x_{1,2}\ll1$ and, independently of the sign of Δ, we have

$$A_0^{(1)}+A_0^{(2)}\approx1-2\beta(i\pi\tau)^{1/2},$$
$$w_0\approx1-2\beta(\pi\tau)^{1/2},\ w_i=1-w_0\approx2\beta(\pi\tau)^{1/2}. \tag{18}$$

The ionization probablity w_i is small and is characterized by a square-root dependence on the pulse duration τ and by a quadratic dependence on the field intensity ($\varphi\propto\mathscr{E}_0^2$).

(b) The range of intermediate pulse durations:

$$(\pi\beta)^{-2}\ll\tau\ll\pi^2\beta^2/\Delta^2. \tag{19}$$

In this case $\sqrt{\tau}x_1\gg1$ but $\sqrt{\tau}x_2\ll1$,

$$A_0\approx\pi^{-3/2}\beta^{-1}(i\tau)^{-1/2},\quad w_0\approx\pi^{-3}\beta^{-2}\tau^{-1}\ll1. \tag{20}$$

The ionization probability $w_i=1-w_0$ under the conditions (19) is close to unity (independently of the sign of Δ, i.e., both above and below the threshold). The residual probability w_0 decrease with increasing τ and $\mathscr{E}_0^2$: $w_0\propto\mathscr{E}_0^{-4}\tau^{-1}$.

(c) Large pulse duration:

$$\tau\gg\pi^2\beta^2/\Delta^2\gg(\pi\beta)^{-2},\quad \sqrt{\tau}|x_{1,2}|\gg1. \tag{21}$$

At $\Delta>0$ we have

$$A_0=A_0^{(2)}\approx\sqrt{\pi}\beta/2\Delta^2(i\tau)^{3/2},\quad w_0=\pi\beta^2/4\Delta^4\tau^3\ll1. \tag{22}$$

The relation $w_0\propto\tau^{-3}$ agrees with the conclusion of Ref. 4. At $\Delta<0$ we have

$$A_0=A_0^{(1)}+A_0^{(2)}\approx\frac{2|\Delta|}{\pi^2\beta^2}+\frac{\sqrt{\pi}\beta}{2\Delta^2(i\tau)^{3/2}},$$
$$w_0=\frac{4\Delta^2}{\pi^4\beta^4}-\frac{\sqrt{2}}{|\Delta|\beta(\pi\tau)^{3/2}}. \tag{23}$$

In this case the difference between the decay laws at $\Delta>0$ and $\Delta<0$ becomes already noticeable. At $\Delta<0$ (below the threshold) the residual probability is low, but as $\tau\to\infty$ it tends to a certain constant value. At $\Delta>0$, the residual probability tends to zero with increasing τ.

2) Let now $|\Delta|\gtrsim\pi^2\beta^2/4$, i.e., let the difference between the field frequency and its threshold value be more appreciable. The arguments of the Fresnel integrals in Eq. (15) are determined in this case by the parameter $(|\Delta|\tau)^{1/2}$ and therefore two different ionization regimes are possible: $|\Delta|\tau\ll1$ and $|\Delta|\tau\gg1$. At $|\Delta|\ll1$, i.e., at a short pulse duration, the arguments of the Fresnel integrals in (15) are small. The initial decay stage, characterized by Eqs. (18), is again realized. This result is consequently independent of the relation between Δ and $\pi^2\beta^2$. The condition for the applicability of Eqs. (18) is the restriction on the pulse duration:

$$\tau\ll\min\{|\Delta|^{-1},\ \pi^{-2}\beta^{-2}\}. \tag{24}$$

At large pulse duration $|\Delta|\tau\gg1$ and at $|\Delta|\gtrsim\pi^2\beta^2/4$ the ionization regime is different for positive and negative Δ.

(a) $\Delta<0$, $|\Delta|\gg\pi^2\beta^2/4$, $|\Delta|\tau\gg1$:

$$A_0=e^{-i\Delta\tau}(1-\pi\beta/2|\Delta|^{1/2})+\sqrt{\pi}\beta/2\Delta^2(i\tau)^{3/2},$$
$$w_0=1-\frac{\pi\beta}{|\Delta|^{1/2}}+\frac{\sqrt{\pi}\beta}{\Delta^2\tau^{3/2}}\sin\left(\tau\Delta-\frac{\pi}{4}\right). \tag{25}$$

With increasing τ the probability w_0 tends in this case, in oscillatory fashion, to its asymptotic value which differs less from unity the larger $|\Delta|$, i.e., the more the field frequency differs from the threshold value.

(b) $\Delta > 0$, $\Delta\tau \gg 1$. At $\Delta > \pi^2\beta^2/2$, arg $(\sqrt{i\tau}x_1)$ becomes larger than $\pi/2$, and the Fresnel-integral transformation $\Phi(u) = -\Phi(-u)$ becomes necessary for a transition to the right-hand half-plane, after which the asymptotic form of $\Phi(u)$ at $|u| \gg 1$ can be used. The result is

$$A_0 \approx \exp\{-i\Delta\tau - \tau\pi\beta\sqrt{\Delta}\} + \frac{\sqrt{\pi}\beta}{2\Delta^2(i\tau)^{3/2}},$$

$$w_0 = e^{-2\pi\beta\tau\sqrt{\Delta}} + e^{-\pi\beta\tau\sqrt{\Delta}}\frac{\sqrt{\pi}\beta}{\Delta^2\tau^{3/2}}\sin\left(\tau\Delta - \frac{\pi}{4}\right) + \frac{\pi\beta^2}{4\Delta^4\tau^3}. \quad (26)$$

The first term in each of the equations in (26) corresponds to an exponential decay constant $\Gamma_i = 2\pi\beta\sqrt{\Delta}$ equal to the ionization width. At a large excess above threshold, the ionization, as expected, follows mainly an exponential law. The nonexponential terms in Eqs. (26) are significant at not too large values of Δ, and also at a very long pulse duration, when $\Gamma_i\tau \gg 1$ and the exponential $\exp(-\Gamma_i\tau)$ vanishes. This result agrees with the general properties of the decay of quantum systems.[7]

Thus, on the whole the ionization probability (or the residual probability) depends substantially on the pulse duration τ. At short τ the system is practically insensitive to the threshold frequency [the ionization regimes determined by Eqs. (18) and (20) are the same for $\Delta < 0$ and $\Delta > 0$]. At large τ above the threshold ($\Delta > 1$) a considerable ionization takes place (total ionization in the limit as $\tau \to \infty$), and below the threshold ($\Delta < 0$) a finite residual probability $w_0(\tau)$ is preserved. The residual probability w_0 is closer to its asymptotic value

$$\omega_0(\infty) = |A_0^{(1)}(\tau)|^2,$$

the larger τ, Depending on the deviation from the threshold Δ, the asymptotic residual probability $w_0(\infty)$ starts out with zero at $\Delta = 0$ and increases with decreasing Δ in accordance with Eqs. (13).

The characteristic times at which a transition from the region of "short" pulses to the region of "long" pulses is effected are determined by the right-hand side of the inequality (24).

2. Ionization from S states, $|v_{E_0}|^2 = \beta E^{3/2}$

The contribution from the continuum of the quasienergy states is determined in this case by the integral

$$A_0^{(2)}(\tau) = \tilde{\beta}\int_0^\infty \frac{dE e^{-iE\tau}E^{3/2}}{\pi^2\tilde{\beta}^2E^3 + (E-\Delta)^2}, \quad (27)$$

$$\tilde{\beta} \approx \beta\left(1 - \int_0^\infty \frac{dE|v_{E_0}|^2}{E^2}\right). \quad (28)$$

The energy region that makes a noticeable contribution to the integral (27) should be substantially lower than the characteristic energy scale $\sim |E_0|$ over which the matrix element $|v_{E_0}|^2$ deviates noticeably from the relation $|v_{E_0}|^2 = \beta E^{3/2}$. It can be easily seen that by virtue of this condition the first term in the denominator of the integrand of (27) is always small, except in the vicinity of the point $E \approx \Delta$ at $\Delta > 0$. This makes it possible to replace the integral (27) by its equivalent

$$A_0^{(2)}(\tau) \approx \tilde{\beta}\int_0^\infty \frac{dE e^{-iE\tau}E^{3/2}}{\pi^2\tilde{\beta}^2|\Delta|^3 + (E-\Delta)^2}. \quad (29)$$

This integral is again expressed in terms of Fresnel integrals and can be represented in a form similar to (15):

$$A_0^{(2)}(\tau) = \tilde{\beta}(\pi/i\tau)^{1/2} + \frac{i}{2|\Delta|^{3/2}} \times\{x_1^3 e^{i\tau x_1^2}(1 - \Phi(\sqrt{i\tau}x_1)) - x_2^3 e^{i\tau x_2^2}(1 - \Phi(\sqrt{i\tau}x_2))\}, \quad (30)$$

$$x_{1,2}^2 = -\Delta \pm i\pi\beta|\Delta|^{3/2}. \quad (31)$$

The contribution from a discrete quasienergy level to the amplitude $A_0(\tau)$ takes at $\Delta < 0$ the form

$$A_0^{(1)} \approx e^{-i\Delta\tau}\left(1 - \int_0^\infty dE\frac{|v_{E_0}|^2}{E^2}\right), \quad (32)$$

$A_0^{(1)} = 0$ at $\Delta > 0$.

Formula (30) is again substantially simplified at short and long pulse durations τ, and in contrast to Sec. 1 the only parameter that determines the scale τ is now the deviation from the threshold Δ. The expression for the total probability amplitude $A_0(\tau)$ at all Δ can be written in the form

$$A_0(\tau) \approx \left(1 - \int_0^\infty dE\frac{|v_{E_0}|^2}{E^2}\right)\exp\{-i\Delta\tau - \pi\beta\tau\Delta^{3/2}\theta(+\Delta)\} + \begin{cases}\beta(\pi/i\tau)^{1/2} & \text{at } |\Delta|\tau \ll 1\\ -3\beta\sqrt{\pi}/4\sqrt{i}\Delta^2\tau^{5/2} & \text{at } |\Delta|\tau \gg 1\end{cases}, \quad (33)$$

where $\theta(x > 0) = 1$ and $\theta(x < 0) = 0$.

The first term in (33) at $\Delta > 0$ describes an exponential decay, and the second a nonexponential one. The exponential-decay constant is the ionization width $\Gamma_i = 2\pi\beta\Delta^{3/2}$. At small τ and $\Gamma_i\tau \ll 1$ the system again behaves in like manner both above and below the threshold ($\Delta \gtrless 0$). The amplitude $A_0(\tau)$ and the residual probability $w_0 = |A_0(\tau)|^2$ at finite τ are continuous functions of the deviation from the threshold Δ. With increasing τ, the $w_0(\tau)$ dependence in the near-threshold region becomes more and more abrupt and takes a steplike form as $\tau \to \infty$. In this case the residual probability assume its asymptotic value

$$w_0(\tau \to \infty) = |A_0(\infty)|^2 \approx \left(1 - 2\int_0^\infty dE\frac{|v_{E_0}|^2}{E^2}\right)\theta(-\Delta). \quad (34)$$

Above the threshold ($\Delta > 0$) the asymptotic residual probability is equal to zero. Below the threshold ($\Delta < 0$) the residual probability as $\tau \to \infty$ is practically independent of and differs little from unity.

The steplike character of the asymptotic residual probability (as $\tau \to \infty$) in the case of ionization from S states distinguishes this case from the case of ionization from P states, where the asymptotic residual probability is continuous (as a function of Δ). Whereas in the case of ionization from P states in the near-threshold region $\Delta < 0$ the asymptotic residual probability is small (as $\tau \to \infty$ and at $|\Delta| \ll \pi^2\beta^2$), in ionization from S states the deviation of $w_0(\infty)$ from unity is small at all $\Delta > 0$. In both cases, however, the asymptotic probability of the ionization $w_i(\tau \to \infty)$ at $\Delta > 0$ differs from zero (although it can be small) and is equal to unity at $\Delta > 0$. We shall investigate below which of these results, and to what degree, remain valid on going to more realistic models that admit of a smooth time dependence of the radiation-pulse envelope $\mathscr{E}_0(t)$.

4. IONIZATION BY A RADIATION PULSE WITH A SMOOTH ENVELOPE. GENERAL EQUATIONS

Thus, let the switching function $f(t)$, normalized by the condition $f_{max} = f(0) = 1$, be a smooth function of the time satisfying the condition $f(\pm\infty) = 0$ and $f(-t) = f(t)$. The electron photodetachment is described by the general equations (3), which are equivalent to a single integrodifferential equation for the function $C_0(t)$:

$$i\dot{C}_0 = E_0 C_0 - 4i \int_0^\infty dE |v_{E_0}|^2 f(t) \cos \omega t \int_{-\infty}^t dt' e^{iE(t'-t)} f(t') C_0(t') \times \cos \omega t'. \quad (35)$$

The characteristic rate of change of the function $C_0(t)$ in Eqs. (3) and (35) is determined by the ground-state energy E_0: The funtion $C_0(t)\exp(iE_0 t)$ is slow compared with $\exp(-iE_0 t)$. In this sense, the switching function $f(t)$ is also slow if the pulse duration τ satisfies the condition $\tau \gg 1/|E_0|$. The nonresonant part of the interaction is responsible for that part of the integrand with respect to t' in (35) which is certainly rapidly oscillating. Integrating in this term with respect to t' (in analogy with Ref. 8) and retaining in (35) only the terms whose rate of change can be $\sim E_0$, we reduce (35) to the form

$$i\dot{C}_0 = (E_0 + \delta E_0'(t)) C_0 - if(t) \int_0^\infty dE |v_{E_0}|^2 \times \int_{-\infty}^t dt' \exp[i(E-\omega)(t'-t)] f(t') C_0(t'), \quad (36)$$

where

$$\delta E_0'(t) = -f^2(t) \int_0^\infty \frac{dE |v_{E_0}|^2}{E - E_0 + \omega} \quad (37)$$

is that part of the shift of the level E_0 due to the dynamic Stark effect, which is caused by the nonresonant part of the interaction energy.

Let $\delta E_0''(t)$ be the additional (as yet unknown) part of the shift of the level E_0 due to the resonant part of the interaction energy, and let

$$E_0(t) = E_0 + \delta E_0'(t) + \delta E_0''(t) \equiv E_0 + \delta E_0(t).$$

The substitution

$$C_0(t) = \exp\left\{-i \int_0^t E_0(t') dt'\right\} b_0(t) \quad (38)$$

reduces (36) to the form

$$i\dot{b}_0 = -\delta E_0''(t) b_0 - i \int_0^\infty dE |v_{E_0}|^2 f(t) \times \int_0^\infty dt' \exp\left\{-i(E-\omega)t' + i \int_{t-t'}^t E_0(t'') dt''\right\} f(t-t') b_0(t-t'). \quad (39)$$

The two independent functions $\delta_{E''}(t)$ and $b_0(t)$ determine the phase and the amplitude of the complex function $C_0(t)$, so that $\delta E_0''(t)$ and $b_0(t)$ can be regarded as real, and Eq. (39) can be rewritten in the form

$$\dot{b}_0 = -\int_0^\infty dE |v_{E_0}|^2 f(t) \int_0^\infty dt' \cos\left[(E-\omega)t' - \int_{t-t'}^t E_0(t'') dt''\right] \times f(t-t') b_0(t-t'), \quad (40)$$

$$\delta E_0''(t) b_0(t) = -f(t) \int_0^\infty dE |v_{E_0}|^2 \times \int_0^\infty dt' \sin\left[(E-\omega)t' - \int_{t-t'}^t E_0(t'') dt''\right] f(t-t') b_0(t-t'). \quad (41)$$

Equation (41) admits of further simplification if it is assumed that sin [...] is a rapidly oscillating function of t'. The last statement is qualitatively justified by the fact that in the region of small E, where the sinusoidal factor in (41) can be a slow function of t', this factor is itself small, therefore the region of small E makes a small contribution to the integral with respect to E. We present now a more rigorous proof. We introduce in place of t' the new variable

$$\Theta = [E - \omega - E_0(t)]^{-1} \int_{t-t'}^t dt'' [E - \omega - E_0(t'')].$$

We multiply the integrand in (41) by $\exp(\lambda\Theta)\exp(-\lambda\Theta)$, where λ^{-1} is of the order of the characteristic times of variation of the slow functions $f(t-t')$ and $b_0(t-t')$. We expand the product

$$R(\Theta) \equiv e^{\lambda\Theta} f(t-t') b_0(t-t') \frac{E-\omega-E_0(t)}{E-\omega-E_0(t-t')} \quad (42)$$

in powers of Θ, obtaining in lowest order $R(\Theta) = f(t) b_0(t)$. The integral with respect to t' takes in this approximation the form

$$\mathrm{Im} \int_0^\infty d\Theta \exp[-\lambda\Theta + i(E-\omega-E_0(t))\Theta] = \frac{E-\omega-E_0(t)}{\lambda^2 + (E-\omega-E_0(t))^2} \approx P \frac{1}{E-\omega-E_0(t)}, \quad (43)$$

so that we get as a result

$$\delta E_0''(t) = -f^2(t) \int_0^\infty dE \frac{|v_{E_0}|^2}{E-\omega-E_0(t)}. \quad (44)$$

It can be verified that the corrections connected with the series expansion (42) make a contribution that is small in the parameter $(\lambda/|E_0|)^2 \ll 1$, where $|E_0|$ determines the characteristic energy scale over which the function $|v_{E_0}|^2$ reaches a maximum.

Accurate to small corrections $\sim d^2\mathscr{E}_0^2/E_0^2$ we can put $E_0(t)\approx E_0$ in the denominator of (44). Jointly with (37), Eq. (44) determines, just as in the case of a large excess above threshold,[8] the total shift of the level on account of the quadratic Stark effect

$$\delta E_0(t)\approx -\tfrac{1}{4}\alpha(\omega)\mathscr{E}_0^2 f^2(t),$$

where $\alpha(\omega)$ is the dynamic polarizability of the level E_0.

Equation (40) determines the probability amplitude of the transition $b_0(t)$. It can hardly be solved in the general form. We shall consider below a situation wherein the ionization process (but, of course, not the level shift) can be described by perturbation theory. Putting in the right-hand side of (40) $b_0=1$, we obtain after simple transformations an expression for the ionization probability

$$w_i=1-|b_0(\infty)|^2=4\int_0^\infty dE|v_{E_0}|^2 \times\left\{\mathrm{Re}\int_0^\infty dt f(t)\exp\left[i(E-\Delta)t-i\int_0^t \delta E_0(t'')dt''\right]\right\}^2, \tag{45}$$

where $\Delta=E_0+\omega$ (is the field is turned on and off smoothly, the system is characterized by the asymptotic value of the deviation from the threshold at $t=\pm\infty$).

In the next section we shall describe the results obtained from Eq. (45) for a model switching function $f(t)=\exp(-\lambda|t|)$, where $\lambda=1/\tau$, with account taken of the level shift $\delta E_0(t)=-\alpha e^{-2\lambda|t|}$ $[\alpha=(1/4)\alpha(\omega)\mathscr{E}_0^2]$. We shall analyze both the ensuing ionization regimes and the conditions for the applicability of perturbation theory, which are determined by the inequality $w_i<1$ and at which Eq. (45) is valid.

5. REGIMES OF IONIZATION BY A RADIATION PULSE WITH A SMOOTH ENVELOPE

For the model switching function $f(t)=\exp(-|\lambda|t)$, the integral with respect to the time t in Eq. (45) is expressed in terms of the incomplete gamma function $\gamma(a,x)$ (Ref. 6):

$$w_i=\lambda^{-2}\int_0^\infty dE|v_{E_0}|^2\left\{\mathrm{Re}\left[e^{-i\alpha/2\lambda}\right.\right.$$
$$\left.\left.\times(-i\alpha/2\lambda)^{-\frac{1}{2}+i(\Delta-E)/2\lambda}\gamma\left(\frac{1}{2}-i\frac{\Delta-E}{2\lambda},\ -i\frac{\alpha}{2\lambda}\right)\right]\right\}^2. \tag{46}$$

Integration with respect to the energy E can be carried out in different limiting cases if the matrix element v_{E_0} is in the form of its approximate representation at small E.

1. Ionization from P states, $|v_{E_0}|^2=E^{1/2}$

1) Let at first the Stark shift of the levels be small:

$$\alpha\ll\lambda,\quad {}^1/_4\tau|\alpha(\omega)|\mathscr{E}_0^2\ll 1. \tag{47}$$

Formula (46) takes in this case the simplest form

$$w_i=4\lambda^2\int_0^\infty\frac{dE|v_{E_0}|^2}{[(\Delta-E)^2+\lambda^2]^2}. \tag{48}$$

It follows therefore that under the condition (47), depending on the deviation form the threshold Δ, the following three ionization regimes are possible:

(a) $\Delta\gg\lambda$, wherein

$$w_i=2\pi\beta\Delta^{1/2}/\lambda\propto\tau. \tag{49}$$

The linear $w_i(\tau)$ dependence indicates that Eqs. (49) corresponds to the ordinary exponential decay during its initial stage, when the product $\Gamma_i\tau$ is small.

Equation (49) is valid so long as $\beta>\lambda/\Delta^{1/2}$. In strong fields, however, it can be generalized in elementary fashion: $w_i=\Gamma_i\tau$ is replaced by $w_i=1-\exp(-\Gamma_i\tau)$.

(b) $|\Delta|\ll\lambda$, in this case

$$w_i=\pi\beta/\sqrt{2\lambda}\propto\sqrt{\tau}. \tag{50}$$

This ionization regime is characterized by a square-root dependence of $w_i(\tau)$ and is analogous to the ionization regime that occurs under similar conditions in the model of in stantaneous application of the interaction and is described by Eqs. (18).

The condition for the applicability of perturbation theory takes in this case the form $\beta\ll\sqrt{\lambda}$.

(c) $|\Delta|\gg\lambda$, $\Delta<0$,

$$w_i=\pi\lambda^2\beta/4|\Delta|^{3/2}\propto\tau^{-2}. \tag{51}$$

With increasing τ or with decreasing λ, i.e., in the adiabatic limit, at the ionization probability vanishes at $\Delta<0$. This result, as will be shown later, is quite general. It is due to the smoothness of the switching the interaction on and off and differs from the corresponding conclusion in the instantaneous switching model, where the residual probability w_0 is less than unity ($w_i\neq 0$) even in the limit of an infinite pulse duration.

2) Let now, on the contrary, the Stark shift of the level E_0 be large: $\alpha\gg\lambda$. The use of the asymptotic form of the γ function for large values of the second argument, and the averaging of the integrand in (46) over the fast oscillations, yield

$$w_i=\frac{2\pi}{\alpha\lambda}\int_0^\infty dE|v_{E_0}|^2\left[1+\exp\left(-\pi\frac{\Delta-E}{\lambda}\right)\right]^{-1}. \tag{52}$$

In all the preceding cases (48)–(51) the ionization probability was obtained under conditions of applicability of ordinary perturbation theory, and was therefore proportional to the square of the field, $w_i\propto\mathscr{E}_0^2$. In contrast to this, there is no field dependence in (52), since $\alpha\propto\mathscr{E}_0^2$ and $|v_{E_0}|^2\propto\mathscr{E}_0^2$. In this respect Eq. (52), as well as the corollaries below, corresponds to a certain probability saturation due to a shift of the quasienergy level on account of the dynamic Stark effect in a feild of variable amplitude.

In full analogy with the preceding analysis, depending on the relation between Δ and λ, the following three ionization regimes occur:

(a) $\Delta\gg\lambda$,

$$w_i=4\pi\beta\Delta^{3/2}/3\alpha\lambda\propto\tau. \tag{53}$$

This result is valid so long as $w_i < 1$ or

$\beta < \alpha\lambda / \Delta^{3/2}$.

Equation (53) is a reflection of the linear dependence of the ionization probability on the pulse duration τ, and is similar in this respect to (49). However, in view of the considerable variable Stark shift, the coefficient of τ has an entirely different dependence on Δ and does not depend on $\mathscr{E}_0$.

(b) $|\Delta| \ll \lambda$,

$$w_i = \frac{\beta}{\alpha}\lambda^{1/2}\left(1-\frac{1}{\sqrt{2}}\right)\zeta\left(\frac{3}{2}\right) \propto \frac{1}{\sqrt{\tau}}; \qquad (54)$$

under the additional condition

$\beta \ll \alpha/\sqrt{\lambda}$.

Here $\zeta(x)$ is the Riemann zeta function[6] [$\zeta e/2 \approx 2.61$].

The result described by Eq. (54) is essentially connected with the strong dynamic Stark effect in a field of variable amplitude and has no analog in either the instantaneous switching model or under conditions of smooth switching of the interaction when the Stark effect is small.

(c) $\Delta < 0$, $|\Delta| \gg \lambda$,

$$w_i = (\beta/\alpha)\sqrt{\lambda}e^{-\pi|\Delta|/\lambda}. \qquad (55)$$

The ionization probability is exponentially small and in the adiabatic limit ($\lambda \to 0$, $\tau \to \infty$) it vanishes at all[2)] $\Delta < 0$.

2. Ionization form S states, $|v_{E_0}|^2 = \beta E^{3/2}$

In this case, Eqs. (48) and (52) remain valid as before. We present here the results that follow from them and the conditions for their applicability.

1) $\alpha \ll \lambda$; a) $\Delta \gg \lambda$,

$$w_i = 2\pi\beta\Delta^{3/2}\lambda^{-1} \propto \tau \qquad (56)$$

— a linear dependence on τ, corresponding to the initial stage of the exponential decay.

b) $|\Delta| \ll \lambda$,

$$w_i = \pi\beta\sqrt{\lambda}/\sqrt{2} \propto 1/\sqrt{\tau} \qquad (57)$$

— a result analogous to the consequences of Eq. (33) at $|\Delta| \ll \tau 1$ in the instantaneous-switching model.

c) $\Delta < 0$, $|\Delta| \gg \lambda$,

$$w_i = \pi\beta\lambda^2/4|\Delta|^{1/2} \propto 1/\tau^2. \qquad (58)$$

2) $\alpha \gg \lambda$; a) $\Delta \gg \lambda$,

$$w_i = (4\pi\beta/5\alpha\lambda)\Delta^{5/2} \propto \tau. \qquad (59)$$

b) $|\Delta| \ll \lambda$,

$$w_i = \frac{3\beta}{4\pi\alpha}\lambda^{3/2}\left(2-\frac{1}{\sqrt{2}}\right)\zeta\left(\frac{5}{2}\right) \propto \tau^{-3/2}, \quad \zeta\left(\frac{5}{2}\right) \approx 1.34. \qquad (60)$$

c) $\Delta < 0$, $|\Delta| \gg \lambda$,

$$w_i = \frac{3}{2\pi}\frac{\beta\lambda^{3/2}}{\alpha}e^{-\pi|\Delta|/\lambda}. \qquad (61)$$

Again, just as in ionization from P states, in the case of a large variable Stark shift [Eqs. (59)–(61)] the ionization probability saturates, i.e., is independent of the field $\mathscr{E}_0$. At all $\Delta < 0$, including the adiabatic limit ($\lambda \to 0$, $\tau \to \infty$) the ionization probability tends to zero exponentially [Eq. (61)].

CONCLUSION

Let us formulate briefly the main conclusions.

1. At a finite pulse duration in the near-threshold region ($|\Delta| \ll |E_0|$) the ionization probability is finite but is not equal to unity at all $\Delta \gtrless 0$.

2. Directly in the near-threshold region, at a small value of the Stark shift of the level E_0 and under conditions when perturbation theory is valid, the ionization probability is qualitatively independent of the shape of the pulse [Eqs. (18) and (50), (33) and (57)]. The same holds also for exponential decay (at large Δ and τ). Some near-threshold ionization regimes which occur in transitions into the continuum from the P state correspond to appreciable ionization and cannot be described within the framework of perturbation theory [Eqs. (20) and (22)]. (These ionization regimes were investigated in the present paper only in the instantaneous-switching model, and it is therefore not clear to what extent they are universal and whether they remain in force for another pulse shape.)

3. Below the ionization threshold $\Delta < 0$ the character of the process can be substantially different for pulses of step-like shape and for pulses with smooth envelopes. In the latter case, in the adiabatic limit $\tau \to \infty$ [Eqs. (55) and (61)] the ionization probability vanishes, whereas in the case of instantaneous switching the probability w_i is finite at all $\tau < 0$ and $\Delta < 0$. The criterion for the transition to the adiabatic limit in the case of a smooth envelope is that the pulse duration τ be considerably longer than all the characteristic times of the problem and, in particular, we must have $\tau \gg + /|\Delta|$. It is clear therefore that directly near the threshold ($\Delta \to 0$) the adiabatic approximation does not hold, and it is necessary to use the expressions obtained for the ionization probability at a finite pulse duration.

4. In the presence of a strong dynamic Stark effect in a field of alternating amplitude, qualitatively new singularities appear in the ionization near the threshold. A number of new regimes set in near the threshold (items 1–2 and 2–2 of Sec. 5), which differ from the ionization regimes in the instantaneous-switching or in the case of a small Stark shift. In the entire near-threshold region the ionization probability is saturated, i.e., is independent of the field intensity $\mathscr{E}_0$ (up to the transition to the exponential decay at $\Delta > 0$). At a large deviation from the threshold Δ, the ionization probability becomes exponentially small at $\Delta < 0$ and approaches unity at $\Delta > 0$.

1)No account is taken in the cited paper of the subthreshold discrete levels, so that its results pertain only to negative ions but not to atoms.

2)We note that it was found in Ref. 3 that in the adiabatic regime the residual probability $w_0 \neq 0$ at $\Delta > 0$, but this is incorrect.

[1] A. I. Andryushin, A. E. Kazakov, and M. V. Fedorov, Zh. Eksp. Teor. Fiz. **76**, 1907 (1979) [Sov. Phys. JETP **49**, 966 (1979)].

[2] H. S. W. Massey, Negative Ions (Russ. Transl.), Mir, 1979.

[3] S. E. Kumekov and V. I. Perel', Zh Eksp. Teor. Fiz. **81**, 1693 (1981) [Sov. Phys. JETP **54**, 899 (1981)].

[4] K. Rzazevski, M. Lewenstein, and J. H. Eberly, J. Phys. B (1982).

[5] A. E. Kazakov, V. P. Makarov, and M. V. Fedorov, Zh. Eksp. Teor. Fiz. **70**, 38 (1976) [Sov. Phys. JETP **43**, 20 (1976)].

[6] I. S. Gradshtein and I. M. Ryzhik, *Tables of Integrals, Sums, Series, and Products*, Academic, 1965.

[7] L. A. Khalfin, Zh. Eksp. Teor. Fiz. **36**, 1080 (1959) [Sov. Phys. JETP **9**, 767 (1959)].

[8] N. B. Delone and M. V. Fedorov, in Multiphoton Ionization of Atoms, Trudy FIAN **115**, 42 (1980).

Translated by J. G. Adashko