Decay of highly excited atoms in an electric field

M. B. Kadomtsev and B. M. Smirnov

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A calculation is made of the critical electric field strength at which the barrier disappears for an electron in the field of a Coulomb center with given parabolic quantum numbers. The lifetimes of highly excited atoms are found as functions of the difference between the field intensity and the critical value. A study is made of the decay dynamics of highly excited atoms when a beam of such atoms passes through an electric field region. Experimental data on the ionization of highly excited atoms in an electric field are analyzed.

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1. Some years ago, experimental methods were developed that make it possible to produce and investigate highly excited atoms in given highly excited states. These methods, which are based on a tunable laser, yielded much information about processes involving highly excited atoms (see the review of Ref. 1). These methods are currently being developed and perfected. The best method of detecting highly excited atoms with large values of the principal quantum number n of the excited electron ($n > 20$) is based on ionization of these atoms in an external electric field and detection of the resulting ions. The main advantage of the method is its selectivity, since the decay time of a highly excited atom in a field in the parameter range of practical interest depends strongly on both the electric field strength and the quantum numbers of the excited electron. Therefore, for the diagnosis of highly excited states we require a rigorous theory that establishes the connection between the lifetime of a given highly excited state of an atom and the electric field strength. The present paper is devoted to these questions.

The decay mechanism of an atom in an electric field associated with below-barrier tunneling of the electron from the field of the atomic core into the continuum was already elucidated in the first stage of the investigation of this process.[2] The asymptotic theory subsequently developed for the decay of atomic particles in a constant electric field (see, for example, Ref. 3) is not suitable for a highly excited atom. In such a case, the basic assumption of the asymptotic theory is not satisfied, since the barrier width is comparable with the diameter of the atom.

The possibilities of the asymptotic theory can be estimated on the basis of the papers of Damburg and Kolosov,[4,5] who found the first two terms of the asymptotic series for the level width of a highly excited atom in an electric field. In particular, for $n_1 = n$ the expression has the form (here and in what follows we use the system of atomic units: $\hbar = m_e = e = 1$)

$$\Gamma = \frac{4}{Fn^6}\exp\left(3n - \frac{2}{3Fn^3}\right)(1-4n^3F), \quad n \gg 1.$$

We determine formally the maximal value of this expression, which is

$$\Gamma_{max} = 6n^{-3}\exp(-8n^2/3+3n-1).$$

It can be seen that Γ_{max} decreases strongly with increasing n and at large n becomes less than the actually measured level widths. Since the asymptotic theory is invalid at the field strengths corresponding to the obtained maximum of the width, it is not suitable for describing the observed decays at large n. For this reason the asymptotic theory of the decay of highly excited atoms is valid for n which are not large.

The large barrier width in the case of the decay of a highly excited atom in an electric field has the consequence that the decay of the atom during measurable times occurs at an electric field strength close to F_0, the field strength at which the barrier disappears at a point. Therefore, the first task in the investigation of the process is the determination of the critical field.

2. The simplest determination of the critical field can be done as follows.[6-9] The potential containing the highly excited electron is

$$U = -r^{-1} - Fz,$$

where r is the distance from the electron to the atomic core, z is the coordinate along the field, and F is the field strength; the electron excitation energy is $\varepsilon = -1/2n^2$. Equating these quantities and their derivatives, we find that the barrier disappears at the point $z = 4n^2$ on the axis at the field strength

$$F_0 = 1/16n^4. \tag{1}$$

This method of finding the critical field cannot stand up to serious criticism. First, we have used the assumptions of perturbation theory, according to which the electron binding energy in the excited atom is the same as in the absence of the field. Second, in this method of finding the critical field we assume that it does not depend on the distribution of the electron in space, i.e., does not depend on the other quantum numbers of the electron apart from n. Despite the rough approximation used in obtaining the result, the expression (1) has proved itself well in the evaluation of experimental results and has been widely used.

Another approach[10] uses a purely classical description of the motion of an electron in the field of a Coulomb center in an external field. At the critical value of the electric field, the motion of the electron in the field of the ion ceases to be finite. In Ref. 10, expressions are found for the critical fields F_0 and the energies ε of an excited electron in these fields for limiting cases of the electron motion (the values for $n_2 = n$, $m = n$ here and in what follows are to be understood in the limit $n_2, m \to n, n_1 \gg 1$):

$$n_1=n, \quad n^4F_0=2^{10}/3^4\pi^4=0.130, \quad |\varepsilon|n^2=2^8/3^2\pi^2=0.720; \tag{2a}$$

$$n_2=n, \quad n^4F_0=0.3834, \quad \varepsilon=0; \tag{2b}$$

$$m=n, \quad n^4F_0=2^{12}/3^9=0.208, \quad |\varepsilon|n^2=2^7/3^5=0.527. \tag{2c}$$

Here, n, n_1, n_2, m are the parabolic quantum numbers of the electron, and $n=n_1+n_2+m$ ($n\gg 1$).

Our first task is to find the critical field for an electron in the field of a Coulomb center and the frequencies of decay of a highly excited atom in electric fields near the critical field. We shall analyze the Schrödinger equation for the electron wave function in this case. Its variables separate in parabolic coordinates.[11,12] The electron wave function is

$$\psi=\frac{e^{im\varphi}}{\sqrt{2\pi}}\frac{\chi_1(\xi)\chi_2(\eta)}{\sqrt{\xi\eta}} \tag{3}$$

where ξ, η, φ are the parabolic coordinates, m is the projection of the electron's angular momentum onto the field direction, and the equations for the functions χ_1 and χ_2 are[11,12]

$$\frac{d^2\chi_1}{d\xi^2}+\left(\frac{\varepsilon}{2}+\frac{\beta_1}{\xi}-\frac{m^2-1}{4\xi^2}+\frac{F}{4}\xi\right)\chi_1=0,$$
$$\frac{d^2\chi_2}{d\eta^2}+\left(\frac{\varepsilon}{2}+\frac{\beta_2}{\eta}-\frac{m^2-1}{4\eta^2}-\frac{F}{4}\eta\right)\chi_2=0. \tag{4}$$

Here, ε is the energy of the electron state, F is the electric field strength, and β_1 and β_2 are separation constants connected by

$$\beta_1+\beta_2=1. \tag{5}$$

We shall use the circumstance that the state is quasiclassical. This leads to the Bohr quantization conditions

$$\int_{\xi_1}^{\xi_2} p_\xi\, d\xi=\pi(n_1+{}^1/_2), \tag{6}$$

$$\int_{\eta_1}^{\eta_2} p_\eta\, d\eta=\pi(n_2+{}^1/_2). \tag{7}$$

Here,

$$p_\xi=[\varepsilon/2+\beta_1/\xi-(m^2-1)/4\xi^2+F\xi/4]^{1/2}$$

is the electron momentum in the ξ space, and p_η is defined similarly; ξ_1 and ξ_2 are the zeros of p_ξ [$p_\xi(\xi_{1,2})=0$]; η_1 and η_2 are the zeros of p_η; and n_1 and n_2 are parabolic quantum numbers. In accordance with the quasiclassical conditions (6) and (7), we introduce parabolic quantum numbers, this being valid for $n_1\gg 1$, $n_2\gg 1$.

We now determine the critical field at which the barrier disappears. The condition corresponding to entry of the considered level into the continuum takes the form that at the right-hand turning point ξ_2 the effective potential energy has a maximum, so that

$$\left.\frac{dp_\xi}{d\xi}\right|_{\xi_2}=0. \tag{8}$$

The four equations (4)–(8) establish a unique connection between β_1, β_2, the electron energy ε, and the field strength F_0 at which the given level enters the continuum. The solution of this system of equations in the simplest case $m=0$ after introduction of the parameter $y=4F_0/|\varepsilon|^2-1=\beta_2/\beta_1$ can be conveniently represented in the form

$$|\varepsilon|n^2=\frac{64}{9\pi^2(n_1/n)^2(1+y)^2}, \tag{9a}$$

$$F_0n^4=\frac{2^{10}}{(3\pi n_1/n)^4(1+y)^3}, \tag{9b}$$

where the parameter y is the solution of the transcendental equation

$$\frac{n_2}{n_1}=\frac{3}{2}(\sqrt{1+y}-1)^{3/2}\int_0^1 dt\left[(1-t^2)\left(t^2+\frac{\sqrt{1+y}+1}{\sqrt{1+y}-1}\right)\right]^{1/2}. \tag{9c}$$

We consider limiting cases. As $n_2\to 0$, we have $y\to 0$ and $n_2/n_1=(3\pi/8\sqrt{2})y=0.833y$ from Eq. (9c). Equations (9a) and (9b) in this case give

$$F_0n^4=\frac{2^{10}}{3^4\pi^4}\left(1+0.4\frac{n_2}{n}\right), \tag{10a}$$

$$|\varepsilon|n^2=\frac{2^7}{3^2\pi^2}\left(1-0.4\frac{n_2}{n}\right), \tag{10b}$$

the zeroth term of the expansion agreeing completely with the expression (2a). In the other limiting case $n_1\to 0$, $y\to\infty$ we obtain

$$\frac{n_2}{n_1}=\frac{3\sqrt{\pi}}{4}\frac{\Gamma({}^5/_4)}{\Gamma({}^7/_4)}y^{3/4}, \tag{11a}$$

and on the basis of Eqs. (9a) and (9b) we find

$$F_0n^4=\frac{4}{\pi^2}\frac{\Gamma^4({}^5/_4)}{\Gamma^4({}^7/_4)}=0.383,$$

$$|\varepsilon|n^2=\frac{64}{9\pi^2}\left(\frac{n_1}{n}\right)^{2/3}\left[\frac{\Gamma({}^7/_4)}{3\sqrt{\pi}\,\Gamma({}^5/_4)}\right]^{8/3}=1.48\left(\frac{n_1}{n}\right)^{2/3}. \tag{11b}$$

In the limiting case, these expressions agree with (2b).

We now turn to the general case $m\neq 0$. Equations (5)–(7) are transformed in the general case to the following form, which makes it possible to find the critical field F_0 and the electron binding energy ε_0 at this value of the field strength:

$$F_0n^4=\frac{2^{10}f_2^4(\alpha)}{(3\pi n_1/n)^4(1+y)^3}, \tag{12a}$$

$$|\varepsilon_0|n^2=\frac{64f_1(\alpha)f_2^2(\alpha)}{(3\pi n_1/n)^2(1+y)^2}. \tag{12b}$$

The functions in these expressions are given by

$$f_1=\frac{1}{2}\left(x+\frac{1}{x}-\frac{\alpha}{x^2}\right), \quad x=\frac{2}{\sqrt{3}}\sin\left[\frac{\pi}{3}-\frac{1}{3}\sin^{-1}(3\sqrt{3}\alpha)\right], \tag{13a}$$

$$f_2=\frac{3}{4}\int_{\alpha/x^2}^{x}\left(\frac{x}{\tau}-1\right)\left(\tau-\frac{\alpha}{x^2}\right)^{1/2}d\tau. \tag{13b}$$

The values of the parameters α and $y=\beta_2/\beta_1$ in (12) and (13) are related to the electron quantum numbers n_1 and n_2 by

$$\frac{n_1}{m}=\frac{2}{3\pi}\frac{f_2(\alpha)}{\sqrt{\alpha}}, \tag{14a}$$

$$\frac{n_2}{m}=\frac{f_1^{3/2}(\alpha)}{\pi\sqrt{\alpha}}(\sqrt{1+y}-1)^{3/2}\int_{t_1}^{t_2}\left[\frac{\sqrt{1+y}+1}{\sqrt{1+y}-1}\right.$$
$$\left.\times\left(\frac{1}{f_1^2(\alpha)}-t^2\right)+t^2(1-t^2)-\frac{\alpha}{f_1^3(\alpha)(\sqrt{1+y}-1)^3t^2}\right]^{1/2}dt, \tag{14b}$$

the limits of integration (t_1, t_2) in the last integral being the zeros of the integrand.

The relations (12)–(14) make it possible to find the critical field and binding energy of an electron in the field of a Coulomb center. The right-hand side of the relations (12) does not depend on the principal quantum number, and is determined solely by n_1/n, n_2/n, m/n. This gives a scaling law for the critical field and electron binding energy for different values of the principal quantum number of the electron. Further, the parameter α in (12)–(14) varies from 0 to $\sqrt{3}/9$, and the parameter $y=\beta_2/\beta_1$ from $y_{\min}(\alpha)$ to infinity. The value $y_{\min}(\alpha)$ corresponds to the condition that the maximum of the integrand in (14b) is zero, i.e., the integral (14b) vanishes, which corresponds to $n_2=0$. For $\alpha=0$, $y_{\min}=0$, and for $\alpha=\sqrt{3}/9$, $y_{\min}=5/4$.

We consider limiting cases of the relations (12)–(14). In the case $\alpha=0$, we obtain $f_1=f_2=1$ from Eqs. (13), and on the basis of Eqs. (14) we obtain $n_1/m=\infty$, $n_2/m=\infty$, i.e., $m=0$. In this case, Eqs. (12a) and (12b) go over into (9a) and (9b), and the ratio of the expressions (14b) and (14a) into the expression (9c). Another limiting case corresponds to $\alpha=\sqrt{3}/9$. In this case, the lower and upper limits in the expression (13b) coincide, i.e., $f_2=0$, and then in accordance with (13a) we have $f_1=\sqrt{3}/2$. In accordance with (14a), it follows that $n_1=0$. If at the same time $y=y_{\min}=5/4$, then the integral (14b) vanishes, i.e., $n_2=0$ and $n=m$. Substituting the value $y=5/4$ and $f_2(n_1/n)^{-1}=3\pi/2\sqrt{\alpha}=\pi 3^{1/4}/2$ in accordance with the expression (14a) in Eqs. (12a) and (12b), we arrive at Eqs. (2c).

Figure 1 shows the values of the critical field and the corresponding electron energies found in accordance with the expressions (12) and the relations (13) and (14).

3. We now determine the decay frequency of a highly excited hydrogen atom in a constant electric field. Since we are interested in problems associated with the detection of highly excited atoms in an electric field, we shall consider the case of a below-barrier electron transition, when the decay time is appreciably longer than the characteristic time of revolution of the electron in the atom. On the other hand, because of the strong exponential dependence of the decay time on the field strength, the case when the field strength does not differ appreciably from the critical value $|F_0-F|\ll F_0$ is the most interesting.

We consider first the decay of a highly excited atom with $m=0$. The frequency of below-barrier tunneling of the electron is by definition

$$w=\int_s |\psi|^2 v_\xi ds, \qquad (15)$$

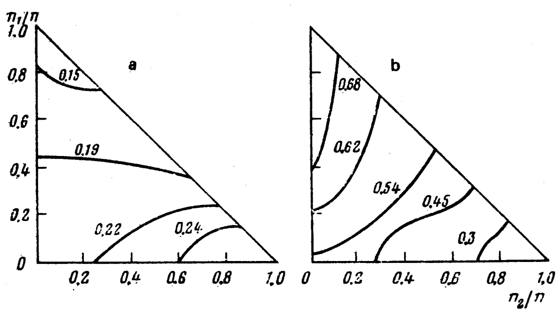

FIG. 1. Reduced values of the critical electric field strengths (a) and the energies corresponding to them (b) on the electric field strength.

where $|\psi|^2$ is the electron density in the classically allowed region of motion (see Fig. 2), v_ξ is the electron velocity, and ds is the element of area. Taking the plane of integration far from the turning point, we obtain $dS=\pi\xi d\eta$. The electron wave function is determined by Eq. (3), and far from the turning point the quasiclassical wave function $\chi_1(\xi)$ has the form

$$\chi_1(\xi)=\frac{c}{\sqrt{v_\xi}}\exp\left(i\int_{\xi_0}^{\xi} v_\xi d\xi\right), \qquad (16)$$

where v_ξ, as in Eq. (15), is the velocity component along the coordinate ξ of the quasiclassical electron. On the basis of the above relations, we obtain for the probability of the atom's decaying in unit time ($\xi\gg\eta$)

$$w=\frac{1}{2}|c|^2\int|\chi_2(\eta)|^2\frac{d\eta}{\eta}=\frac{|c|^2}{2} \qquad (16a)$$

subject to the following normalization condition of the wave function χ_2:

$$\int|\chi_2(\eta)|^2\frac{d\eta}{\eta}=1. \qquad (16b)$$

The quantity $|c|^2$ is the probability of below-barrier tunneling of the electron in the one-dimensional case. In what follows, we shall be interested in the tunneling of an electron at a field strength near the critical value. Then in the region of the well near the turning point, where the electron momentum vanishes, the derivative of the momentum with respect to the coordinate is also near zero. This raises the probability of finding an electron in a well that makes the pre-exponential factor vanish in the expression for the probability of a below-barrier transition at the point where the barrier disappears. This conclusion is valid for a sufficient width of the barrier, when the regions of the well and the regions of the classical motion of the electron are separated. If this condition is not satisfied, the pre-exponential factor in the probability of below-barrier tunneling will contain a small quantity corresponding to the large value of the electron's quantum number.

The solution of the considered problem by the usual method[12] involving matching of the solutions of the Schrödinger equation in the different regions of Fig. 2 with allowance for the quasiclassical motion of the bound electron with respect to both coordinates leads to the following expression for the decay time τ of an atom in an electric field ($\tau=w^{-1}$):

$$\tau=\tau_0 D^{-1}\ln X. \qquad (17)$$

Here, τ_0 is in order of magnitude equal to the revolution time of the bound electron in its orbit, and is

$$\tau_0=1/\sqrt{2}F_0^{3/4}, \qquad (18a)$$

so that F_0 is the field at which the given level disappears. In (17), D is the coefficient of below-barrier tunneling for a parabolic barrier and in accordance with Kemble's formula[12] is equal to

$$D=\left[1+\exp\left(\frac{8\beta_1\Delta\varepsilon f_1^{3/4}x^{3/2}}{|\varepsilon|^{3/2}\sqrt{1-3\alpha/x}}\right)\right]^{-1}. \qquad (18b)$$

Here, $\Delta\varepsilon$ is the energy difference between the top of the barrier and the electron energy level ($\Delta\varepsilon$ is positive

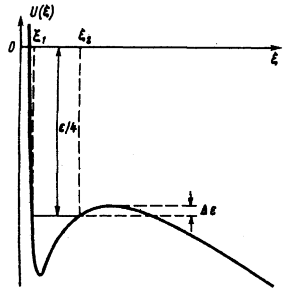

FIG. 2. Coordinate dependence of the effective potential for an electron in the field of a Coulomb center and in a constant electric field.

for a below-barrier transition; see Fig. 2); the remaining notation is the same as in Eqs. (13). The quantity under the logarithm in (17) characterizes the normalization of the wave function in the well. Its value is

$$X=\begin{cases}\dfrac{2\beta_1 x^{1/2} f_1^{3/4}}{|\varepsilon|^{1/2}}\left(1-\dfrac{3\alpha}{x}\right)^{1/2}; & |\varepsilon|\gg\Delta\varepsilon>-\dfrac{|\varepsilon|^{3/2}(1-3\alpha/x)^{1/2}}{16\beta_1 f_1^{3/4}x^{3/2}},\\[2ex] \dfrac{(1-3\alpha/x)|\varepsilon| f_1^{1/2}}{8|\Delta\varepsilon|x}; & \Delta\varepsilon<-\dfrac{\varepsilon^{3/2}(1-3\alpha/x)^{1/2}}{16\beta_1 f_1^{3/4}x^{3/2}}.\end{cases} \tag{18c}$$

The obtained result is valid for a parabolic form of the barrier, which requires fulfillment of the condition

$$\frac{|\Delta\varepsilon|}{|\varepsilon|}\ll\left(1-\frac{3\alpha}{x}\right)^{3}. \tag{19}$$

The main difficulty in finding the lifetime of a highly excited atom in an electric field consists of determining the connection between the field strength F and the barrier height $\Delta\varepsilon$. In the neighborhood of the critical field strength, in the linear approximation in the difference between the field strength and the critical value, $\Delta\varepsilon$ vanishes. To see this, we write the Schrödinger equation (4) in the form

$$\frac{\varepsilon}{4}\chi_1=\left[-\frac{1}{2}\frac{\partial^2}{\partial\xi^2}+V(\xi)\right]\chi_1.$$

In the first order of perturbation theory, the change in the energy is

$$\Delta E/4=\langle\chi_1|\delta V|\chi_1\rangle,$$

where δV is the change in the effective potential associated with the change in the field. In accordance with Eq. (4), it is

$$\delta V=\delta\beta_1/2\xi+\delta F\xi/8,$$

where $\delta\beta_1$ and δF are the deviations of the parameters from their values at the point where $\Delta\varepsilon=0$. For these parameter values, the electron is concentrated mainly in the neighborhood of the turning point ξ_0, which is simultaneously the top of the barrier, i.e., $|\chi_1|\sim\delta(\xi-\xi_0)$. Therefore, in the first order of perturbation theory $\Delta E/4=\delta V(\xi_0)$, i.e., $\Delta\varepsilon=\varepsilon/4-V(\xi_0)=0$.

We determine $\Delta\varepsilon$ near the critical field strength in the following approximation for $|m|=0$. We introduce the notation $\alpha=(F_0-F)/F_0$, $\gamma=4\Delta\varepsilon/\varepsilon=1-(4\beta_1F/\varepsilon^2)^{1/2}$, and, by virtue of what we have said above, $\gamma\ll\alpha$. We represent β_1 in the form $\beta_1=\beta_1^0(1+C\alpha)$, where $\beta_1^0=\varepsilon_0^2/4F_0$, and C is a coefficient that depends on β_1^0 and is determined from Eq. (7):

$$C=(1-\beta_1^0)/(3\beta_1^0+\sqrt{\beta_1^0}).$$

From (6) we now readily obtain an equation for γ:

$$^3/_2\gamma\ln(1.6/\gamma)=\alpha(1+3C). \tag{20}$$

The lifetime of the highly excited state is

$$\tau=\frac{1}{\sqrt{2}}\left(\frac{1}{F_0}\right)^{3/4}\ln(2/\varepsilon_0^{1/2})\left[1+\exp\left(\frac{3\pi^2\gamma n_1}{2}\right)\right]. \tag{21}$$

We rewrite (21) in the case when $n_1=n$ and the barrier penetrability is low:

$$\tau=3.27n^3\ln(3\pi n/4)\exp(3\pi^2\gamma n/2). \tag{22}$$

Note that in the considered region of field strengths, which is outside the region of applicability of the asymptotic theory, this theory gives a different dependence of the lifetime on the principal quantum number. Thus, for $n_1=n$ at the critical field value (2a) the first term of the asymptotic series[13] gives the value $\tau=0.0325n^2\exp(2.128n)$.

In investigating the decay of a highly excited atom in an electric field, we have assumed that the relative width of the level is not large, which made it possible to determine the position of the level ignoring its width. The expressions we have obtained make it possible to estimate the accuracy of this procedure. In accordance with Eqs. (2a) and (21), the product of the level width Γ and the energy at the point of entry of the level into the continuum for $n_1=n$ is

$$\Gamma|\varepsilon_0|=\left[\frac{3\pi n}{4}\ln\left(\frac{3\pi n}{4}\right)\right]^{-1}.$$

Hence, for $n=50$ the ratio of the uncertainty of the level energy to the energy itself is 1.8×10^{-3}, while for $n=20$ it is 5.5×10^{-3}. In the region of below-barrier decay of the atom, in which the processes in which we are interested take place, this ratio is significantly smaller, which means that we can ignore the level width.

We now demonstrate the selectivity of the method of detection of highly excited atoms as a result of their ionization in an electric field on the basis of (22). The ionization probability when a beam of highly excited atoms passes through an electric field region is

$$P=1-\exp(-\tau_{soj}/\tau),$$

where τ_{soj} is the time of sojourn of the atom between the capacitor plates, this being $\sim10^{-6}$ sec under the experimental conditions. We shall assume that highly excited atoms with $n_1=n$ are created selectively. Figure 3 shows the dependence of the measured signal, which is proportional to dP/dF, on the electric field strength, which can be varied smoothly under the experimental conditions. We have here used $\tau_{soj}=10^{-6}$ sec. It can be seen that the method of detecting highly excited atoms using their ionization in an electric field makes it possible to separate reliably highly excited atoms with different values n of the principal quantum number. This circumstance is used experimentally.

In Table I, we give the field strengths at which decay of highly excited atoms has been observed. Under the experimental conditions, the time corresponding to the decay is 10^{-8}–10^{-10} sec. Note that the expression (1) gives the value $F_0n^4=3.2\times10^8$ V/cm.

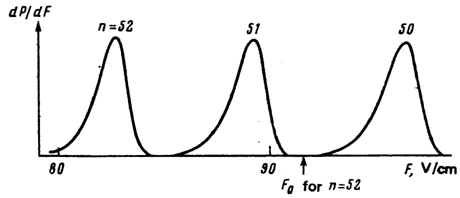

FIG. 3. Dependence of dP/dF on the field strength (P is the probability of the atom's decaying when it passes through a field region at the given strength of the field).

4. Hitherto, we have investigated adiabatic decay of an atom, when the state of the atom with the field switched on is characterized by the same quantum numbers as in the absence of the field. Two decay paths of atoms in an electric field are observed—adiabatic and diabatic.[20,21] The latter is determined by transitions between the highly excited states while the field is being switched on.

Let us now follow the dynamics of the decay of a highly excited atom as it enters an electric field. In reality, a beam of highly excited atoms is passed through the plates of a capacitor, so that ionization of the highly excited atoms is used for their detection. In the absence of the electric field, the highly excited atoms (except for hydrogen) are described by spherical quantum numbers, the electron binding energy being $1/2(n-\delta_l)^2$, where δ_l is the quantum defect of the state with the given angular momentum. It can be seen that the influence of the deviation of the field of the atomic core from the Coulomb field, which makes a contribution δ_l/n^3 to the electron energy, is small compared with the influence of the electric field at a near critical strength. In such a case, the contribution due to the influence of the electric field to the electron energy is $\sim 1/n^2$. Therefore, when we investigate the decay of a highly excited atom in an electric field, it would appear that we should re-expand the wave function of an electron in the state nl with respect to parabolic quantum numbers and for each parabolic state consider the decay independently.

This would be correct if the electric field is switched on suddenly. We shall now write down a criterion for sudden switching on of the field. In the case of slow variation of the field strength, the individual energy levels of an electron in the field of the atomic core and in the electric field have pseudocrossings. If the field of the atomic core were a purely Coulomb field (as in the hydrogen atom), there would be strict crossing of the levels. The minimal distance between the levels, which is determined by the short-range part of the interaction, is

$$\Delta=2\langle\psi_1|V|\psi_2\rangle=2\sum_{l=|m|}^{n-1}\frac{\delta_l}{(nn')^{3/2}}C_l(n_1,n_2,m)C_l(n_1',n_2',m), \tag{23}$$

where $\psi_1\equiv\psi_{n_1n_2m}$, $\psi_2\equiv\psi_{n_1'n_2'm}$ are the unperturbed wave functions of the interacting levels; $n=n_1+n_2+|m|+1$, $n'=n_1'+n_2'+|m|+1$ are the values of the principal quantum number corresponding to these levels; and $C_l(n_1,n_2,m)=\langle\Phi_{nlm}|\psi_{n_1,n_2,m}\rangle$ are the coefficients in the expansion of the parabolic functions $\psi_{n_1n_2m}$ with respect to the basis of the spherical functions Φ_{nlm}.

The condition of sudden switching on of the electric field corresponds to a small value of the Massey parameter, which in the Landau–Zener form becomes[12]

$$\Delta^2\left(\frac{d\Delta E}{dt}\right)^{-1}\ll 1, \tag{24}$$

where ΔE is the energy difference of the corresponding states for the terms unperturbed by the short-range interaction. Since $\Delta E\sim Fn^2$, it is convenient to rewrite condition (24) using the relation (23) in the form

$$dF/dt\gg\Delta^2/n^2. \tag{25}$$

If the condition (25) is satisfied, the pseudocrossing of the energy levels of the excited atom during the variation of the electric field is taken as crossing.

In the cases $|m|=0$, $|m|=1$, and $|m|=2$, the expansion coefficients in (23) are

$$C_2(n_1,n_2,2)=\frac{\sqrt{30}}{n^{5/2}}[(n-n_2-1)(n-n_2-2)(n_2+1)(n_2+2)]^{1/2}, \tag{26a}$$

$$C_1(n_1,n_2,1)=\frac{\sqrt{6}}{n^{3/2}}[(n-n_2-1)(n_2+1)]^{1/2}, \tag{26b}$$

$$C_0(n_1,n_2,0)=1/\sqrt{n}. \tag{26c}$$

Substitution of these expressions in (23) gives

$$\Delta_2=\frac{60\delta_0}{(nn')^4}[(n-n_2-1)(n-n_2-2)(n_2+1)(n_2+2)(n'-n_2'-1)\times(n-n_2'-2)(n_2'+1)(n_2'+2)]^{1/2}, \tag{27a}$$

$$\Delta_1=\frac{12\delta_1}{(nn')^3}[(n-n_2-1)(n_2+1)(n'-n_2'-1)(n_2'+1)]^{1/2}, \tag{27b}$$

$$\Delta_0=2\delta_2/(nn')^2 \tag{27c}$$

(we also assume $\delta_0\gg\delta_1\gg\delta_2$, $\delta_l=0$, $l\geq 3$), where n and n' are the principal quantum numbers of the pseudocrossing levels. In the case of the first crossing of the nearest levels $n'=n+1$ and under the assumption that all the quantum energy defects except the zeroth are small, this formula gives the expression $2\delta_0/n^2(n+1)^2$, which is close to the one obtained in Refs. 22 and 23. As an example, let us consider sodium, for which $\delta_0=1.35$, $\delta_1=0.86$, $\delta_2=0.015$, $\delta_{l\geq 3}\approx 0$.

Analysis shows that at a typical rate of switching on of the electric field, $dF/dt\sim 10^9-10^{10}$ V·cm^{-1}·sec^{-1}, and for $n\sim 20-50$ the transition between the states with $|m|=2$ take place in the diabatic manner, whereas in the cases $|m|=0$ and 1 (see Refs. 20 and 21) the tran-

TABLE I. Electric Field strengths F_0 at which decay of the highly excited state with the given n occurred in the corresponding experiment.

Excited atom	Method of producing the highly excited atoms	Range of principal quantum numbers	F_0n^4, 10^8 V/cm
H Ref. 14	Charge exchange	9–16	5.8
He Ref. 14	»	9–17	5.8
H Ref. 15	»	13–28	6.9
Na Ref. 6	Laser excitation	26–37	3.6
Rb Ref. 7	»	28–78	3.2
Na Ref. 8	»	16–21, $l=m=0$	3.4
		16–19, $l=1$, $m=0$	3.0
		16–19, $l=m=1$	3.1
		15–20, $l=2$, $m=0$	3.6
		15–20, $l=2$, $m=1$	3.7
		15–20, $l=m=2$	4.0
Rb Ref. 16	»	28–60	3.2
Xe Ref. 17	»	24–40	4.0
Na Ref. 18	»	12–14	3.2
Na Ref. 19	»	22–31, $l=0$	3.5
		26–30, $l=1$	3.8
		20–30, $l=2$	3.8

sitions take place adiabatically. At the same time, the self-splitting between the levels is comparable to the distance between them, i.e., in these cases the two-level approximation is invalid. This nature of the development of the system (diabatic and adiabatic) when the electric field is switched on was observed in Ref. 20.

Note that the nature of the transitions when the electric field is switched on is also reflected in the ionization of the atoms in the electric field. When the field switching on is diabatic, the critical field at which decay of the state is observed is determined by (12a). If the system develops in accordance with the adiabatic law, the field energy changes little with the variation of the field because of the large pseudocrossing. Then the decay of the level takes place because of admixture of the level $(n'-1, 0, 0)$, and in accordance with (2a) the value of n' must be found from the relation $0.5n^{-2} = 0.72n'^{-2}$. This gives [in accordance with (2a)] the critical field strength $F_0 = 0.13/n'^4 = 1/16n^4$, which is close to the value obtained from the expression (1).

[1]B. M. Smirnov, Usp. Fiz. Nauk 131, 577 (1980) [Sov. Phys. Usp. 24, 450 (1980)].

[2]C. Lanczos, Z. Phys. 62, 518 (1930); 68, 204 (1931).

[3]B. M. Smirnov, Atomnye stolknoveniya i élementarnye protsessy b plazme (Atomic Collisions and Elementary Processes in Plasma), Atomizdat, Moscow (1968), p. 244.

[4]R. Ja. Damburg and V. V. Kolosov, J. Phys. B 11, 1921 (1978).

[5]R. Ja. Damburg and V. V. Kolosov, J. Phys. B 12, 2637 (1979).

[6]T. W. Ducas, M. G. Littman, R. R. Freeman, and D. Kleppner, Phys. Rev. Lett. 35, 366 (1975).

[7]H. T. Duong, S. Liberman, and J. Pinard, Opt. Commun. 18, 533 (1976).

[8]T. F. Gallagher, L. M. Humphrey, W. E. Cooke, R. M. Hill, and S. A. Edelstein, Phys. Rev. A 16, 1098 (1977).

[9]M. G. Littman, M. M. Kash, and D. Kleppner, Phys. Rev. Lett. 41, 103 (1978).

[10]D. Banks and J. G. Leopold, J. Phys. 118, L. 5, 37 (1978).

[11]H. A. Bethe and E. E. Salpeter, Quantum Mechanics of One and Two Electron Atoms, Springer, Berlin (1957) [the Russian reference is to p. 48 of the Russian translation published by IL, Moscow (1960)].

[12]L. D. Landau and E. M. Lifshitz, Kvantovaya mechanika, Nauka, Moscow (1974), p. 337 [English translation: Quantum Mechanics. Non-Relativistic Theory, 3rd ed., Pergamon Press, Oxford (1977)].

[13]S. Y. Slavyanov, Problemy matematicheskoǐ fiziki (Problems of Mathematical Physics), Izd. LGU (1970), p. 125.

[14]R. N. Il'in, V. A. Oparin, N. T. Serenkov, E. S. Solov'ev, and N. V. Fedorenko, Zh. Eksp. Teor. Fiz. 59, 103 (1970) [Sov. Phys. JETP 32, 59 (1971)].

[15]J. E. Bayfield, G. A. Khrynallah, and P. M. Koch, Phys. Rev. A 9, 209 (1974).

[16]S. Liberman and J. Pinard, Phys. Rev. A 2, 507 (1979).

[17]R. F. Stebbings, C. J. Latimen, W. P. West, F. B. Dunning, and T. B. Cook, Phys. Rev. A 12, 1453 (1975).

[18]M. G. Littman, M. L. Zimmerman, D. Kleppner, Phys. Rev. Lett. 37, 486 (1976).

[19]J. L. Vialle and H. T. Duong, J. Phys. B 12, 1407 (1979).

[20]P. H. Jeyes, G. W. Foltz, K. A. Smith, E. J. Beiting, F. G. Kellert, F. B. Dunning, and R. F. Stebbings, Phys. Rev. Lett. 44, 390 (1980).

[21]F. G. Kellert, K. A. Smith, R. D. Rundel, F. B. Dunning, and R. F. Stebbings, J. Chem. Phys. 72, 3179 (1980).

[22]M. L. Zimmerman, M. G. Littman, M. M. Kash, and D. Kleppner, Phys. Rev. A 20, 2251 (1979).

[23]T. V. Komarov, T. P. Grozdanov, and K. R. Janev, J. Phys. B 13, L573 (1980).

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