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Diffusion mechanism of ionization of highly excited atoms in an alternating electromagnetic field

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A new mechanism is proposed for description of the ionization of highly excited atomic states by a strong low-frequency electromagnetic field. It consists of diffusion of the electron along atomic states highly perturbed by the field. The diffusion time is calculated, and the range of fields in which this mechanism is dominant is estimated. The results are compared with existing experimental data.

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1. INTRODUCTION

The ionization of atoms and molecules in a strong electromagnetic field differs substantially from the ionization process in a constant field as the result of the presence of an additional parameter–the field frequency ω. It is well known[1] that the square of the ratio γ between the external field frequency and the frequency ω_t of tunneling of the electron through the potential barrier is the dominant factor:

$$\gamma=\frac{\omega}{\omega_t}=\frac{\omega}{\mathscr{E}}(2E_n^{(0)})^{1/2},$$

where $\mathscr{E}$ is the external field strength and $E_n^{(0)}$ is the binding energy of the electron in the atom (here and below we use atomic units $m=\hbar=e=1$). The analytic solution of the problem of ionization of a particle bound by short-range forces, which was obtained by Keldysh[1] on the assumption that $\mathscr{E}\gg\mathscr{E}_{at}$ (where $\mathscr{E}_{at}=5\times10^9$ V/cm is the atomic field strength) and $E_n^{(0)}\gg\omega$, showed that for $\gamma^2>10$ the ionization process has a multiphoton nature, while for $\gamma^2<1$ it has the nature of a tunnel effect. The solution of the same problem obtained by other authors[2,3] for a circularly polarized field without the limitations on field strength and frequency mentioned above is essentially similar to the results obtained by Keldysh.

At the present time there is no general description of the process of ionization of a real atom in an alternating field. Data have been obtained only by means of perturbation theory for the multiphoton limiting case and by the quasiclassical method for the tunneling limiting case (see for example Ref. 4). Inclusion of the long-range nature of the Coulomb potential, carried out in terms of the quasiclassical method, did not result in a change in the value of γ^2 which marks the boundary between the tunnel and multiphoton regimes.[5]

However, separation of the ionization process into two limiting cases—tunneling and multiphoton—is valid only in a situation where the perturbation of the discrete atomic spectrum does not affect the transition probability. We note that, depending on the field strength and frequency of the ionizing field and also on the binding energy of the initial state, perturbation of the spectrum can play an important role for various values of the parameter γ (excluding the tunneling case).

In what follows we shall discuss the hydrogen atom, since the excited states of interest to us are always hydrogenlike. The perturbation of a bound electronic state with principal quantum number n is strong when the splitting of this state reaches a magnitude comparable with the distance to the closest shell, i.e., when we have the condition

$$\delta E_n/\Delta E_{n,\,n+1}\geqslant 1. \tag{1}$$

We note that the condition (1) is simultaneously a condition of inapplicability of the standard nonstationary perturbation-theory approach,[6] in which the unperturbed spectrum is taken as the basis.

It is an important circumstance that satisfaction of condition (1) is in no way equivalent to the condition for occurrence of the tunnel effect ($\gamma^2<1$) (Ref. 7). For this reason the problem of describing the ionization of an

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atom under conditions of strong perturbation of the spectrum arises.

In the present article we consider the ionization of an excited atom under conditions of strong perturbation of the spectrum of bound electronic states by an external alternating field of arbitrary polarization, whose frequency is $\omega \ll E_n^{(0)}$. Under these conditions, ionization occurs as the result of diffusion of the electron along the perturbed spectrum. The diffusion mechanism of increase of the energy of an atomic electron can occur when a series of successive processes of induced radiation and absorption of quanta of the external field take place. Previously a number of studies have considered the diffusion mechanism of dissociation of molecules by laser radiation. Here the possibility of realization of the diffusion mechanism is related in one way or another to the almost equidistant nature of the unperturbed spectrum of vibrational states and with the attuning of the external field frequency ω to resonance with these states.[8] The possibility of realization of the diffusion mechanism in an atom, as will be shown below, is due not to the choice of a resonant frequency ω, but to a strong perturbation of the atomic spectrum.

The physical justification of the conditions for occurrence of diffusion in an atomic spectrum is given in Sec. 2, the mathematical description of the diffusion process in Sec. 3, and the discussion of its experimental realization in Sec. 4.

2. JUSTIFICATION OF THE MODEL

It is well known that for hydrogen-like states on increase of the principal quantum number n the distance between levels is $\Delta E_{nn'} = |E_n^{(0)} - E_{n'}^{(0)}| \sim 1/n^3$ for $|n-n'| \ll n$. Each of the levels in n^2-fold degenerate in the orbital and magnetic quantum numbers l and m. Since we are interested in levels with $n \gg 1$, we shall use quasiclassical reasoning for estimates.

On turning on of the external field the atomic spectrum is perturbed and the degeneracy of the levels is lifted. We find a critical field strength at which the change of the level energy becomes comparable with the distance between neighboring atomic shells. When perturbation theory is applicable, the perturbation of the levels can be both linear and quadratic in the external field strength $\mathscr{E}$, depending on the field frequency and the value of the principal quantum number.[9] We are interested in both cases.

Linear perturbation in an alternating field has a different nature than in a constant field. In an alternating field the perturbation leads to appearance of quasi-energy levels which are a superposition of quasi-energy harmonics with energies $E_n \pm K\omega$, and $E_n \to E_n^{(0)}$ for $\mathscr{E} \to 0$. When the perturbation is linear in the externalfield strength, quasi-energy harmonics with numbers $K \sim n^2\mathscr{E}/\omega$ have maximum weight. Thus, in the case of linear perturbation we have $\delta E_n \sim n^2\mathscr{E}$. The critical value of the external field strength when the splitting of states of the shell n reaches a magnitude of the order of the distance to the closest shell $n+1$ and mixing of states of different shells occurs is

$$\mathscr{E}_{cr} \approx 1/n^5. \tag{2}$$

If the perturbation is quadratic in the external field strength $\mathscr{E}$, then for $\omega \sim 1/n^3$ in the expression for the dynamic polarizability

$$\alpha_n \approx \sum_{n'} |r_{nn'}|^2 \left(\frac{1}{\Delta E_{nn'} + \omega} + \frac{1}{\Delta E_{nn'} - \omega} \right)$$

the denominators have a value $1/n^3$ and since $r_{nn'} \sim n^2$, we have $\alpha_n \sim n^7$. This dependence for α_n corresponds to the critical field strength, which is also determined by the relation (2). We note that in the case of a constant field ($\omega \ll 1/n^3$) we obtain from the relation written above for the polarizability the well known formula $\alpha_n \sim n^6$ (Ref. 10, Sec. 77). In fact, in the sum over n' the terms with $n' = n \pm 1$ practically cancel each other, decreasing α_n by n times. Here we obtain for the critical field strength the estimate $\mathscr{E}_{cr} \sim 1/n^4$ or $1/n^5$.

It can be seen that the value of $\mathscr{E}_{cr}$ depends weakly on the specific perturbation mechanism. For $\mathscr{E} > \mathscr{E}_{cr}$, perturbation theory is inapplicable, since all terms of the series have the same order of magnitude. In what follows we shall use Eq. (2) for estimates.

For a field strength $\mathscr{E} > \mathscr{E}_{cr}$, mixing of the levels occurs. In accordance with the quasiclassical estimates (Ref. 10, Sec. 52) the average distance between these levels turns out to be approximately constant. Here the selection rules in the orbital quantum number are removed in a field of arbitrary polarization, and those in the magnetic quantum number are removed in the absence of linear or circular polarization.[11] Thus, a spectrum of highly broadened quasienergy states arises with a level density which on the average is constant and which extends to the continuum. Between these states induced transitions of the electron occur. The probability of single-photon transitions is determined by Fermi's golden rule:

$$w \approx |r_{nn'}|^2 \mathscr{E}^2 \rho_{n'}, \tag{3}$$

where $\rho_{n'}$ is the energy density of final states.

The role of multiphoton transitions in a strongly perturbed spectrum turns out to be less important. We shall show this in the case of two-photon transitions. The amplitude of a two-photon transition is described by the relation

$$\sum_m \frac{r_{nm} r_{mn'}}{E_n - E_m \mp \omega} \mathscr{E}^2,$$

where $E_{n,m}$ are the energies of the perturbed levels. From this sum over m it is necessary to exclue the principal terms corresponding to cascade transitions $n \to m \to n'$, for which the energy denominator $E_n - E_m \mp \omega$ is very small, since $E_n - E_m$ and ω are of order n^{-3}. These terms have the same order of magnitude as the amplitude of the one-photon transition. The reason for the exception cited is that such cascade transitions already are established in the diffusion equations discussed below if second iterations are carried out in these equations. Discarding of the remaining terms,

for which $E_m \sim n^{-2}$ and which have random phases, leads to an error $\lesssim 1/n$. This procedure corresponds to use of balance equations for description of kinetics.[12]

We emphasize again that the external field is assumed to be arbitrarily polarized. This corresponds to the actual arrangement of the experiments discussed below. In the general case the form of polarization of the field has a weak effect on the diffusion mechanism.

Since the probabilities of absorption and radiation of a photon in induced transitions are approximately the same, in the case of low frequencies where

$$\omega \ll E_n^{(0)} \approx 1/n^2, \tag{4}$$

the electron can take on energy only with a statistically large number of absorption-radiation events. This process has the nature of diffusion along the energy axis and describes ionization in the multiphoton limiting case under conditions in which the spectrum of the atom is highly perturbed by the external field.

Let us clarify the conditions in which the tunnel effect plays an important role under conditions of strong perturbation of the spectrum. For this purpose we shall formulate a criterion of the occurrence of tunneling ($\gamma^2 < 1$) in a form similar to Eq. (2). In the case of a constant field the experimental data obtained for $10 < n < 50$ show that the field strength $\mathscr{E}_t$ at which ionization occurs in atomic times has a magnitude[13-15]

$$\mathscr{E}_t \approx 1/16n^4. \tag{5}$$

This value is in reasonable agreement with the theoretical estimates.[16,17]

In an alternating low-frequency field when the condition for tunnel ionization is satisfied, the result depends on the nature of the field polarization. In the case of circular polarization the estimate (5) is obviously unchanged. In the other limiting case of linear polarization of the field the tunneling probability increases by a factor $\sqrt{\mathscr{E}n^3}$. This quantity can be obtained if we assume that the field changes adiabatically slowly and if we use the saddle-point method in averaging over time of the formulas obtained for a constant field. In particular, for $\mathscr{E} = \mathscr{E}_{cr} = 1/n^5$ the tunneling probability decreases by n times in comparison with the case of a constant field. However, in view of the sharp threshold dependence of the ionization probability on the field, this change of the probability makes practically no change in the estimate (5).

Thus, the value of $\mathscr{E}_t$ in the case of an alternating field is practically unchanged from the value for a constant field. From comparison of Eq. (5) with Eq. (2) it is evident that for $\gamma^2 < 1$ the diffusion process of ionization also can occur, but only in a very narrow region:

$$1/n^5 < \mathscr{E} < 1/16n^4,$$

i.e., only for extremely large values of n. Thus, if $\mathscr{E} > \mathscr{E}_t$ and $\gamma^2 < 1$, ionization of atoms from a highly excited state, as a rule, will occur as a consequence of the tunnel effect.

In the opposite case $\gamma^2 > 10$ there is no tunnel ionization, and when the criterion (2) is satisfied the diffusion mechanism is dominant; for lower field strengths, when condition (2) is not satisfied, ionization of the atom from a highly excited state has a K_0-photon nature. From the condition $\gamma^2 > 10$ and Eq. (4) we can obtain an estimate of the interval of field frequencies in which the ionization has a diffusion nature:

$$1/n^2 \gg \omega > \mathscr{E}n > 1/n^4. \tag{6}$$

As can be seen from Eq. (6), the diffusion mechanism of ionization of neutral atoms can occur for frequencies $\omega \sim 1/n^3$, i.e., for $n \gtrsim 10$, beginning in the far infrared region and extending into the longer-wavelength region. For example, at the short-wavelength limit of this region for $\omega \approx 10^{-3}$ atomic units $\approx 4 \times 10^{-5}$eV, ionization from a state with $n = 10$ occurs as the result of the diffusion mechanism for field strengths in the range 5×10^5 V/cm $< \mathscr{E} < 5 \times 10^6$ V/cm. At higher field strengths the tunnel effect is dominant. As n increases, the region of realization of the diffusion mechanism of ionization broadens.

It is evident that for positive ions the diffusion mechanism can occur also in the optical region of frequencies.

The diffusion process determines the ionization probability value averaged over small energy intervals. Here statistical fluctuations of the ionization probability as a function of the radiation frequency are possible; these are similar to Ericson fluctuations in the cross sections of nuclear reactions.[18]

3. MATHEMATICAL DESCRIPTION OF THE DIFFUSION PROCESS

The kinetic equation which determines the change with time of the distribution of atoms along the energy axis has the form

$$dN(E,t)/dt = -W_{\text{rad}}(E \to E-\omega)N(E,t) - W_{\text{abs}}(E \to E+\omega)N(E,t) + W_{\text{rad}}(E+\omega \to E)N(E+\omega,t) + W_{\text{abs}}(E-\omega \to E)N(E-\omega,t). \tag{7}$$

Here $N(E,t)$ is the probability of finding an atom in a state with energy E: W_{rad} and W_{abs} are the probabilities per unit time of transitions with radiation or absorption of a photon of frequency ω. In accordance with the arguments given in Sec. 2, processes involving absorption and radiation of several photons in one event are not taken into account. Neglecting spontaneous transitions, $W_{\text{rad}} \approx W_{\text{abs}} \approx w$, where w is determined by Eq. (3).

Assuming $E \gg \omega$, we reduce the difference equation (7) to differential form:

$$\frac{\partial N(E,t)}{\partial t} = \frac{\partial}{\partial E}\left[w \frac{\partial N(E,t)}{\partial E} \right] \omega^2. \tag{8}$$

In an external field with constant frequency and amplitude the value of w does not depend on time. According to Eq. (3) we have $w \sim n^7 \mathscr{E}^2 \sim \mathscr{E}^2 E^{-7/2}$. Here we have used the estimates $\rho \sim n^3$ and $r_{nn'} \sim n^2$ which are characteristic of the unperturbed spectrum of the atom. Despite the fact that the levels of the atom are split in the field and

have a quasienergy structure, the estimate given does not depend on this, since on increase of the density of final states the matrix element of the transition between fixed states decreases in proportion. However, this reasoning is valid only for the averaged probabilities and does not exclude the possibility of relatively small statistical fluctuations of w with change of the field frequency ω.

Equation (8) is an equation of the diffusion type, and from dimensional considerations it is possible to obtain the characteristic time in which the diffusion occurs:

$$\tau \approx E_0^2/w\omega^2 \approx E_0^{11/3}/\omega^2\mathscr{E}^2,$$

where E_0 is the characteristic energy interval. Taking for this quantity its maximum value compatible with the condition (2) for existence of the diffusion mechanism, we find that $E_0 \sim \mathscr{E}_{cr}^{2/5}$ and

$$\tau \approx \mathscr{E}_{cr}^{11/3}/\omega^2\mathscr{E}^2 = 1/n^{11}\omega^2\mathscr{E}^2. \tag{9}$$

As we shall see, this estimate is confirmed by the solution of Eq. (8).

Let us return to finding the solution of (8). We use the designation

$$\omega^2 w = a^2 E^{-7/2},$$

where the constant a is given by $a \sim \omega\mathscr{E}$. We construct a solution of Eq. (8) by the Fourier method:

$$N_\lambda(E, t) = \exp(-\lambda^2 t) N_\lambda(E), \quad N_\lambda(E) = E^{1/4} y_\lambda(E),$$
$$\frac{d^2 y_\lambda}{dE^2} = -\left(\frac{\lambda^2}{a^2} E^{1/2} - \frac{77}{16} E^{-2}\right) y_\lambda, \tag{10}$$

where λ is the variable-separation constant in Eq. (8).

For solution of Eq. (10) we use the fact that for $\lambda \gtrsim 1/\tau, E \approx E_0$ we have

$$(\lambda/a)^2 E^{11/4} \gg 1. \tag{11}$$

The inequality (11) is not satisfied for $t \gg \tau$. However, this region is not of interest, since for large times the diffusion stops. When this is taken into account the solution of Eq. (10) is written in terms of cylindrical functions

$$y_\lambda(E) = C_1 E^{1/2} J_{2/11}\left(\frac{4\lambda}{11a} E^{11/4}\right) + C_2 E^{1/2} J_{-2/11}\left(\frac{4\lambda}{11a} E^{11/4}\right),$$

where $C_{1,2}$ are constants between which a relation can be established from the condition of vanishing of the probability flux along the energy axis for $E = E_0$:

$$\left.\frac{\partial N}{\partial E}\right|_{E_0} = 0. \tag{12}$$

The condition (12) corresponds to disappearance of the diffusion mechanism as a consequence of violation of criterion (2).

The solution of Eq. (10) satisfying Eq. (12) can be written in the form

$$N_\lambda(E) = C(\lambda) \frac{d}{dE_0}\left\{ (EE_0)^{1/4}\left[J_{-2/11}\left(\frac{4\lambda}{11a} E_0^{11/4}\right) J_{2/11}\left(\frac{4\lambda}{11a} E^{11/4}\right) - J_{2/11}\left(\frac{4\lambda}{11a} E_0^{11/4}\right) J_{-2/11}\left(\frac{4\lambda}{11a} E^{11/4}\right)\right]\right\}. \tag{13}$$

The function $C(\lambda)$ must be determined from the initial condition. For this purpose we shall qualitatively approximate the cylindrical functions in Eq. (13) by the first terms of their asymptotic expansion:

$$N_\lambda(E) \approx C'(\lambda) E^{1/4} \cos\left[\frac{4\lambda}{11a}(E_0^{11/4} - E^{11/4})\right], \tag{14}$$

where $C'(\lambda)$ is some new unknown function. Expression (14) now satisfies the condition (12) with accuracy to quantities of first order in the large parameter $\lambda E_0^{11/4}/a$.

The general solution of Eq. (8) has the form

$$N(E, t) = \int_0^\infty d\lambda C'(\lambda) e^{-\lambda^2 t} E^{1/4} \cos\left[\frac{4\lambda}{11a}(E_0^{11/4} - E^{11/4})\right].$$

It can be shown that $C'(\lambda)$ is determined here by the formula

$$C'(\lambda) = \frac{2}{\pi a} \int_0^{E_0} N(E, 0) E^{1/4} \cos\left[\frac{14\lambda}{11a}(E_0^{11/4} - E^{11/4})\right] dE.$$

For the initial condition $N(E, 0) = \delta(E - E_0)$ we obtain

$$N(E, t) = \frac{(EE_0)^{1/4}}{a(\pi t)^{1/2}} \exp\left\{ -\frac{4}{121 a^2 t}(E_0^{11/4} - E^{11/4})^2\right\}. \tag{15}$$

As follows from Eq. (15), with accuracy to a numerical factor 1/30 the diffusion time is determined by Eq. (9). The approximation (14) used above is not valid for $t \gg \tau$. Obviously this region is not of interest, in view of the fact that the diffusion process terminates at such times. From (15) it is also evident that the function $N(E, t)$ has a sharp maximum at $E = E_0$. Therefore an electron appreciably removed from the initial state with energy E_0 rapidly goes into the continuum. This fact follows from the sharp rise of the diffusion rate with decrease of the binding energy E, which follows from Eq. (9).

The total probability of ionization of an atom up to a moment of time t is determined by the expression

$$W(t) = 1 - \int_0^{E_0} N(E, t) dE.$$

Estimating this quantity for $t \ll \tau$, we obtain $W(t) \sim \sqrt{t/\tau}$. From this and also from Eq. (15) it is evident that for the mechanism of ionization considered the concept of probability per unit time has no meaning.

4. DISCUSSION OF RESULTS AND ANALYSIS OF EXPERIMENTS

Let us now turn to analysis of the existing experimental data of Bayfield, Koch, and Gardner on the process of ionization from highly excited states of the hydrogen atom by fields in the radio-frequency region.[19-21] These authors observed ionization of atomic hydrogen states with $n \approx 50$ in fields of strength $\mathscr{E} \lesssim 100$ V/cm with frequency $\omega \approx 10$ GHz. They also determined the critical field strength at which ionization occurs in a time less than 10^{-8} sec. In Ref. 20 they observed a change in the probability of ionization from states with $n = 45-57$ by

two to three times for a small change of the external field frequency in the range $\omega = 9.6–11.4$ GHz. The nature of the dependence $w(\omega)$ is related to the number n of the excited state and varies irregularly with n. However, the average width of the observed maxima are approximately the same: ~1 GHz.

Under the experimental conditions realized in the studies of these authors, the parameter γ^2 varied from 10^2 to 2 as the field strength changed from 10 to 100 V/cm, i.e., the tunnel effect does not play an important role in the ionization process. The experimental observation of a frequency dependence of the probability in Ref. 20 confirms this conclusion, as the authors of this work correctly point out. Nevertheless, estimates show that the perturbation both of the initial states ($n \approx 50$) and of the higher states was of the order of or greater than the distance to the closest unperturbed levels, i.e., the perturbation was strong. This fact also is noted by the authors of these studies.

From our point of view, ionization under the conditions realized in the experiments of Bayfield, Gardner, and Koch can occur only as the result of electron diffusion along a highly perturbed spectrum. All of the characteristics of the observed ionization process—threshold field intensity, absolute ionization probability, and dependence of the probability on frequency—are well described in terms of the theory developed above. For example, in accordance with the condition (2) the critical field for $n \approx 50$ is $\mathscr{E}_{cr} \approx 15$ V/cm. This value corresponds to the observed threshold for formation of ions. The relation (6) is also satisfied here. Thus, it is necessary to assume that the ionization has a diffusion nature. The theoretical diffusion time is $\tau \approx 10^{-8}$ sec for a field strength $\mathscr{E} = 100$ V/cm. This value is in good agreement with the experimental data on ionization probability.[20] From the τ value given it is possible to obtain a lower limit of the width of the quasistationary states: $\Gamma > 100$ MHz. This also agrees with the experimental data on the width of the maxima in the frequency dependence of the ionization probability. As a whole the nature of the frequency dependence of the ionization probability observed in Ref. 20 is in reasonable agreement with the statistical nature of the diffusion ionization process (see Ref. 18). Finally, the fact that in Ref. 21 ionization was not observed in a field $\mathscr{E} < 5$ V/cm without tuning to a resonance also agrees with the diffusion model, since the field strength indicated is less than the critical value (2).

Further studies of the diffusion ionization process present obvious interest. On the one hand, this is a rare example of a case in which the dynamics of the transitions in an atomic spectrum is determined by statistical regularities. On the other hand, this process can have substantial value in various phenomena involving selective excitation of high states.

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