Ionization of highly excited states of a hydrogen atom by a strong low-frequency field

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(Received 6 July 1983)
Zh. Eksp. Teor. Fiz. **86**, 860–865 (March 1984)

The probability of ionization of highly excited states of a hydrogen atom by a low-frequency field is estimated by using the previously derived quasi-classical wave function of an electron in a Coulomb field and in a radiation field. The expression obtained predicts an ionization threshold at field intensities approximately equal to those observed experimentally, but predicts an increase in ionization probability that is approximately ten times the increase observed experimentally when the field intensity in the threshold region is increased. The approximations underlying the derivation of the equation for the ionization probability are discussed.

I. INTRODUCTION

As was first shown by Keldysh,[1] multiphoton and tunnel ionizations are the limiting cases of the same process of ionization by an alternating field. However, his study and those made by other authors[2,3] actually pertained to the case of ionization of a system bound by a short-range potential and having only one level. For real atoms, the presence of a long-range Coulomb potential and of a large number of bound states leads to a substantial change in ionization probability as compared with the case of a short-range potential. This accounts for the extensive development of the high-order perturbation theory for atoms,[4–6] which is in satisfactory agreement with experimental data on multiphoton ionization of atoms by visible and infrared radiation.

Exceptions are experiments on ionization of highly excited states of the hydrogen atom by a microwave field,[7,8] as well as analogous experiments recently conducted on He (Ref. 9) and Na.[10] A high-order perturbation theory can hardly be developed to explain these experiments, since the number of absorbed photons in the latter is several hundred. Apparently, it cannot as yet be stated that there exists a complete quantitative description of the process of multiphoton ionization of highly excited atomic states by a strong low-frequency field, even though several approaches to this problem have been proposed.

Since the adiabaticity parameter $\gamma = \omega/nF_0$ introduced by Keldysh is less than or of the order of unity in these experiments, the ionization probability can be estimated[11] on the basis of the corresponding expression for tunnel ionization in a constant field if F_0 is replaced by $F_0 \cos \omega t$ and averaged over the period of the field. Here and below, the atomic system of units is used, ω and F_0 are the frequency and intensity of the field, and n is the principal quantum number. The expression obtained predicts a marked increase (threshold) of the ionization probability at fields F_0 equal to several tenths of n^{-4}, this being approximately equal to what is observed experimentally. A disadvantage of this expression is that it is independent of the frequency of the field and differs by an insignificant factor from the corresponding expression for the tunneling probability in a constant field. Both the tunneling process itself and the expression obtained are essentially quantum-mechanical.

A second approach is a purely classical one[12,13] and consists in solving the classical equations of motion with the aid of the Monte Carlo method. Although the authors note the good agreement between the calculated results and experimental data, there still remains the question, is it possible to account for the ionization of highly excited states of atoms by a low-frequency field solely on the basis of classical mechanics? If so, it would be of definite interest to explain in what region of frequencies and fields classical mechanics could be used, since both limiting cases, i.e., multiphoton and tunnel ionization, are essentially quantum-mechanical. Of interest is another classical mechanism of ionization,[14–16] based on the onset of stochastic instability in nonlinear oscillations of an electron acted upon by a strong wave.

This paper will give a rough estimate of the probability of ionization of highly excited states of a hydrogen atom by a strong low-frequency field on the basis of a previously[17] derived quasi-classical wave function.

1. QUASI-CLASSICAL ESTIMATE OF IONIZATION PROBABILITY

The author[17] derived a quasi-classical wave function for highly excited states of a hydrogen atom in a strong alternating field of frequency less than or of the order of the Kepler frequency. The wave function constitutes a double series: a Fourier series in the alternating field and a series in spherical functions. In the radial wave functions of all the channels, a common, rapidly oscillating part related to the classical elliptical motion of the electron, was separated from the slowly changing part which is the excitation probability amplitude of the given state. The square of the probability-amplitude at $u = \pi$ (u being the variable describing the elliptical motion of the electron) give the excitation probability of the given state per period. Dividing this quantity by the period of orbital revolution $T = 2\pi n^3$ and summing over all the states located above the ionization threshold, we obtain the ionization probability per unit time:

$$W = \frac{1}{2\pi n^3} \sum_{N=N_0+1}^{\infty} \sum_{s=-\infty}^{\infty} |a_{Ns}(\pi)|^2, \qquad N_0 = (2n^2\omega)^{-1}, \quad s = (l-L-N)/2. \tag{1}$$

0038-5646/84/030502-04$04.00

where N is the number of absorbed photons, l is the orbital quantum number, and L is the orbital quantum number of the unperturbed state. For amplitudes $a_{Ns}(\pi)$, the following expression was obtained in the form of a product of Bessel functions:

$$a_{Ns}(\pi)=J_{N+s}\left(\frac{c_-}{2\sin\pi\omega n^3}\right)J_s\left(\frac{c_+}{2\sin\pi\omega n^3}\right)$$
$$\times\exp\{i\pi N(1-\omega n^3)+i\pi s\}, \tag{2}$$

$$c_\pm=\frac{\nu^5F_0}{2}\left(1-\frac{M^2}{L^2}\right)^{1/2}\int_{-\pi}^{\pi}du(1-\varepsilon\cos u)[(\cos u-\varepsilon)\cos\omega t$$
$$\mp(1-\varepsilon^2)^{1/2}\sin u\sin\omega t], \tag{3}$$

where

$$t=\nu^3(u-\varepsilon\sin u),\qquad \varepsilon=(1-L^2/\nu^2)^{1/2},$$

M being the magnetic quantum number, and ν the mean value of the principal quantum number for all the channels; in Ref. 17, this value was chosen equal to the principal quantum number n of the unperturbed state. For $\omega\nu^3\ll 1$, we have

$$c_\pm=-{}^3/{}_2\pi\nu^5F_0\varepsilon(1-M^2/L^2)^{1/2}. \tag{4}$$

For $\omega\nu^3\gg 1$ and $L\ll\nu$, the quantities $c_\pm$ are expressed[6] in terms of the Airy function $\mathrm{Ai}(y)$ and its derivative $\mathrm{Ai}'(y)$:

$$c_\pm=\frac{2^{2/3}\pi F_0}{\omega^{5/3}}\left(1-\frac{M^2}{L^2}\right)^{1/2}[-\mathrm{Ai}'(y)\mp y^{1/2}\mathrm{Ai}(y)],$$
$$y=\left(\frac{\omega L^3}{2}\right)^{2/3}. \tag{5}$$

Let us note that expression (5) is independent of the principal quantum number, i.e., of the electron energy.

Since, $\omega n^3<1$ in the experiments of Refs. 7–9, we shall use henceforth for our estimate will Eq. (4) with ν replaced by n, and consider only states with $M=L=0$. Representing squared Bessel functions as an integral, and the sum of squared Bessel functions over N also as an integral,[18] after simple transformations we obtain

$$W=\frac{x}{\pi^2n^3}\int_0^{\pi/2}d\vartheta\,\vartheta\sin\vartheta\,J_{N_0}(x\cos\vartheta)J_{N_0+1}(x\cos\vartheta),$$
$$x=\frac{3n^2F_0}{2\omega}. \tag{6}$$

The parameter x is the ratio of the maximum Stark energy to the photon energy. Since the index of the Bessel function N_0 is a large quantity, W will be appreciable only when the argument is approximately equal to the index. Again representing the product of Bessel functions as an integral and using an asymptotic expression of the Bessel function, we finally obtain

$$W=\frac{1}{4\pi^2n^3N_0^{2/3}}\left[-\mathrm{Ai}'(a)-a\int_a^\infty dv\,\mathrm{Ai}(v)\right], \tag{7}$$

$$a=\frac{2(N_0-x)}{N_0^{1/3}}=\frac{2}{(2n^2\omega)^{2/3}}(1-3n^4F_0). \tag{8}$$

Tables are available for the derivative and integral of the Airy function.[18] For $a\gg 1$,

$$W=(8\pi^{5/2}n^3N_0^{2/3}a^{5/4})^{-1}\exp(-{}^2/{}_3a^{3/2}). \tag{9}$$

If the perturbation acts on the atom during a time $t=2\pi m/\omega$, where m is the number of periods of the field, the probability of its ionization is

$$R=1-\exp(-Wt). \tag{10}$$

Let us compare the predictions of Eqs. (7)–(10) with experiment. Since N_0 amounts to several hundred in the experiment, a is very large, and the ionization probability W is low, provided the field F_0 does not approach the value $1/3n^4$, at which a sharp increases in ionization probability should be observed. Such a sharp increase in W (threshold) was observed for the hydrogen atom in fields F_0 that were 2.6 times lower than this value in Ref. 9 and 3.5 to 4.5 times lower in Ref. 8. The fact that in the experiment the ionization threshold is observed at fields several times lower than given by Eq. (8) is partly accounted for, since in Eq. (4) the mean value of the principal quantum number ν was replaced by the smaller quantity n. Actually, Eq. (8) should have ν^5/n instead of n^4, where ν can be found from the definition of the mean energy

$$1/2\nu^2=(1/2n^2+|E_f|)/2. \tag{11}$$

Since the final state is close to the ionization threshold, the energy E_f corresponding to it can be made to approach zero. The $\nu=2^{1/2}n$, and in Eq. (8) the factor 3 will be replaced by $3\cdot 2^{5/2}\approx 17$. It is obvious that the experiment lies between these two extreme definitions of ν. Let us note that for states with nonzero values of L and M an additional factor $(1-L^2/\nu^2)^{1/2}(1-M^2/L^2)^{1/2}$, smaller than unity, will appear in front of n^4 in Eq. (8).

Equation (7) predicts in the threshold region an increase in $W(F_0)$ ten times faster than observed experimentally. Contributing to the leveling of the increase of $W(F_0)$ in the experiment is the inhomogeneity of the field and especially the presence of a whole set of initial states for which the thresholds are different. It is still unclear whether a more exact solution of the fundamental quasi-classical equations (see below) will result in a different dependence of W of F_0 in the threshold region than predicted by Eq. (7). According to Eqs. (7) and (8), this dependence is determined by the quantum-mechanical quantity n^4F_0.

2. SIMPLIFICATION OF QUASI-CLASSICAL EQUATIONS AND DISCUSSION OF THE APPROXIMATIONS

We shall now discuss the quasi-classical approximations on the basis of which expressions were derived for the amplitudes $a_{Ns}(\pi)$ and the ionization probability W. As shown in Ref. 17, the amplitudes are

$$a_{Ns}(\pi)=\sum_{N',s'=-\infty}^{\infty}i^{N-N'}J_{N-N'+s-s'}(c_-)J_{s'-s}(c_+)b_{N's'}, \tag{12}$$

where the constants b_{Ns} must satisfy the system of homogeneous algebraic equations

$$b_{Ns}e^{-2i\pi\nu_N} = \sum_{N'=-\infty}^{N_0} \sum_{s=-\infty}^{\infty} J_{N-N'+s-s'}(c_-)J_{s'-s}(c_+)b_{N's'}, \quad (13)$$

$$\nu_N = (-2E-2N\omega)^{-1/2} \quad (14)$$

Let us note first that the system (13) can be substantially simplified if the following periodic functions are introduced:

$$d_N(\beta) = \sum_{s=-\infty}^{\infty} e^{is\beta} b_{Ns}, \quad d_N(\beta+2\pi) = d_N(\beta) \quad (15)$$

and the summation theorem for Bessel functions[18] is used. After some trivial changes in notation, we obtain the following system instead of Eq. (13):

$$d_N e^{-2i\pi\nu_N} = \sum_{N'=-\infty}^{N_0} J_{N-N'}(w)d_{N'}, \quad w = (c_-^2+c_+^2+2c_-c_+\cos\beta)^{1/2}. \quad (16)$$

If the function

$$F(\varphi) = \sum_{N=-\infty}^{N_0} e^{iN\varphi} d_N, \quad F(\varphi+2\pi) = F(\varphi) \quad (17)$$

is further introduced, and the integral representation of Ref. 18 is used for the Bessel functions, the system (16) can be reduced to the homogeneous integral equation

$$\int_{-\pi}^{\pi} d\alpha F(\alpha) \sum_{N=-\infty}^{N_0} e^{iN(\varphi-\alpha)}[e^{-2i\pi\nu_N} - e^{iw\sin\alpha}] = 0, \quad (18)$$

which should be satisfied for any φ.

Equations (13) or the substantially simpler (16) and (18) are the fundamental quasi-classical equations for describing multiphoton transitions in hydrogenlike atoms acted upon by a strong field of an electromagnetic wave. In their derivation it is only required that, in all the channels making a substantial contribution to the multiphoton process under consideration, one be able distinguish in the electron motion the classical motion due to the Coulomb field. When $\omega n^3 \gg 1$, this motion is parabolic. The quantities $c_\pm$ in this case are determined by expression (5) and are independent of energy, and therefore Eqs. (16) or (18) should satisfactorily describe the ionization process.

In the case $\omega n^3 \lesssim 1$, this motion is elliptical, and $c_\pm$ is determined in accordance with Eq. (3) or (4). At such frequencies, ν_N can be replaced by $\nu_0 + N\omega\nu_0^3$, where $\nu_0 = (-2E)^{-1/2}$, and the upper limit of the sum in Eq. (18) can be made to approach infinity. This approximation corresponds to the approximation in which the levels are equidistant. The sum over N then reduces to a δ function. If the function $F(\alpha)$ is sought in the form $F(\alpha) = \exp[if(\alpha)]$, one finds that $f(\alpha)$ must satisfy the functional equation

$$f(\varphi - 2\pi\omega\nu_0^3) = f(\varphi) + w\sin\varphi + 2\pi(\nu_0 - n), \quad (19)$$

which has the solution

$$f(\varphi) = \frac{n-\nu_0}{\omega\nu_0^3}\varphi + \frac{w\cos(\varphi+\pi\omega\nu_0^3)}{2\sin\pi\omega\nu_0^3}, \quad (20)$$

where n is an integer. It follows from the condition that $F(\varphi)$ be periodic that

$$\nu_0 = n + k\omega\nu_0^3, \quad (21)$$

where k is also an interger. Taking k equal to zero, we find that $\nu_0 = n$, and expanding $F(\varphi)$ in Fourier series in φ and β, we arrive at the solution of Eqs. (13), guessed in Ref. 17, in the approximation of equidistant levels and hence, at expression (2) for amplitudes $a_{Ns}(\pi)$. Since $k\omega\nu_0^3$ is small compared to n, $E \approx -1/2n^2 + k\omega$, i.e., different k simply correspond to different quasi-energy levels.

The solution found above should adequately describe multiphoton transitions in the region of equidistance of levels. This solution should give a poorer description of ionization, since the electron acquires energy it leaves at the end of the ionization process the region of equidistant levels and of ellipticity of its motion and enters the region of parabolic motion and a nonequidistant spectrum. Nevertheless, we assume that when $\omega n^3 \lesssim 1$ the main contribution to the determination of the wave function and hence to the ionization is made by the region of equidistant levels, and the solution obtained can give a rough estimate of the ionization probability.

Equations (16) and (18) are equations for eigenvalues. The fact that the summation in them extends to N_0 is due to the boundary conditions in open channels,[6] which reduce to the absence of converging waves from them. The energy obtained is therefore complex, and its imaginary part determines the width of the level. When N_0 is replaced by infinity, the energy becomes real, and the ionization probability can be approximately determined, as above, by means of Eq. (1). It follows from Eq. (18) that the energy depends on β and φ as parameters, i.e., each level in the presence of a field is converted into a band. This is due to the fact that in the presence of a field, the wave function is a mixture of an infinite number of states with different orbital quantum numbers and different numbers of absorbed (emitted) photons. At frequencies ω that are multiples of n^{-3}, a resonant structure appears in the solution (20). However, the hydrogen spectrum is not equidistant, and its influence on the ionization probability has not yet been elucidated.

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Translated by A. Peiperl