

IONIZATION OF ATOMS IN AN ALTERNATING ELECTRICAL FIELD. III

A. M. PERELOMOV and V. S. POPOV

Institute of Theoretical and Experimental Physics

Submitted to JETP editor August 4, 1966

J. Exptl. Theoret. Phys. (U.S.S.R.) 52, 514-526 (February, 1967)

We develop further the quasiclassical approximation in the problem of the penetration of the particle through a time-varying potential barrier. We consider with the aid of this method the influence of higher harmonics of the field of the light wave on the ionization probability, and also obtain formulas for the Coulomb correction in the case of not too high frequencies ($\gamma \ll \gamma_C$, where γ_C is defined in (51)). We discuss also the physical meaning of the parameter γ_C and the possibility of taking into account the Coulomb interaction when $\gamma \gtrsim \gamma_C$.

THE ionization of atoms in the field of a strong electromagnetic wave was considered recently in a number of theoretical papers.^[1-5] The incident wave was assumed to be strictly monochromatic, and the Coulomb interaction between the emitted electron and the atomic remainder was neglected. In our preceding paper^[5] we proposed a simple method for calculating the ionization probability of a bound level under the influence of an alternating electric field; this method was an extension of the usual quasiclassical approximation to the nonstationary case. In the present paper we employ this method to consider the following questions:

1. We investigate the influence of higher harmonics in the incident wave on the ionization probability. It turns out here that in the high-frequency region of practical interest for experiments ($\gamma \gg 1$) this influence is quite significant. Thus, an admixture of $\sim 10^{-4}$ of the second harmonic or of $\sim 10^{-7}$ of the third harmonic (relative to the intensity of the fundamental frequency) is sufficient to change the ionization probability by one order of magnitude.

2. We take into account the Coulomb correction to the formula for the ionization probability. For the region of not very high frequencies ($\gamma \ll \gamma_C$, where γ_C is a characteristic quantity that depends on the external field intensity F , the level energy $\kappa^2/2$, and the force of the Coulomb interaction, see (51)), we obtain formula (45) for the ionization probability of an arbitrary atoms. The physical meaning of the parameter γ_C and the method for taking into account the Coulomb interaction when $\gamma \gtrsim \gamma_C$ are discussed.

1. INFLUENCE OF HIGHER HARMONICS ON THE IONIZATION PROBABILITY

Let us consider the ionization of a bound level ($\omega_0 = \kappa^2/2$) under the influence of an electric field $F(t)$ which is not strictly monochromatic. As shown in^[5], to find the ionization probability w it is necessary to calculate the contracted action \tilde{S} along a "classical trajectory" of the subbarrier motion. With this, the extremal classical trajectory (which minimizes $\text{Im } \tilde{S}$), corresponds to a situation in which the particle goes out from under the barrier at the instant when $F(t)$ has a maximum. Choosing the external field in the form

$$F(t) = F \sum_{k=1}^{\infty} f_k \cos k\omega t, \quad \sum_{k=1}^{\infty} f_k = 1, \quad (1)$$

we see that $t = 0$ is the point of the maximum of $F(t)$, if¹⁾

$$\sum_{k=1}^{\infty} k^2 f_k > 0. \quad (2)$$

The "time" t varies during the course of particle motion along the contour shown in Fig. 1. The classical particle trajectory is determined from the equations (see^[5])

$$\ddot{x} = F(t), \quad x(t_0) = 0, \quad \dot{x}(t_0) = i\kappa, \quad \dot{x}(0) = 0. \quad (3)$$

We are interested only in the subbarrier mo-

¹⁾Condition (2) guarantees that $F(0) > F(t)$ at sufficiently small t . If $F(t)$ has also other maxima, then it is necessary to stipulate in addition that their magnitude be smaller than $F(0)$. This will be satisfied, for, example, if all the $f_k \geq 0$, but the latter condition is not essential.

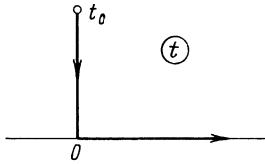


FIG. 1. The "time" t for the motion through the barrier. At the instant $t = 0$ the particle goes out from under the barrier, and the remaining part of the trajectory is realizable in classical mechanics.

tion, during the course of which²⁾ $\text{Im } \tilde{S}$ develops. Making therefore the substitution $\tau = i\omega t$ ($\tau > 0$) we have

$$x(t) = \frac{F}{\omega^2} \sum_{k=1}^{\infty} \frac{f_k}{k^2} (\text{ch } k\tau_0 - \text{ch } k\tau),$$

$$\dot{x}(t) = \frac{iF}{\omega} \sum_{k=1}^{\infty} \frac{f_k}{k} \text{sh } k\tau. \quad (4)$$

From this follows an equation for the determination of the "initial instant" τ_0 :

$$\sum_{k=1}^{\infty} \frac{f_k}{k} \text{sh } k\tau_0 = \gamma, \quad \gamma = \frac{\omega}{\omega_0} = \frac{\omega x}{F} \quad (5)$$

(γ is the adiabaticity parameter introduced in [1]). Using the equation $x\dot{x} = 0$ when $t = t_0$ and $t = 0$ (see (3)), we obtain the contracted action \tilde{S}

$$\tilde{S}(t_0, 0) = \int_{t_0}^0 \left[\frac{\dot{x}^2}{2} + F(t)x - \frac{x^2}{2} \right] dt = i \frac{\omega_0}{\omega} \int_0^{\tau_0} \left(1 + \frac{\dot{x}^2}{x^2} \right) d\tau. \quad (6)$$

The quantity $\text{Im } \tilde{S}(t_0, 0)$ determines the principal (exponential) factor in the formula for the ionization probability

$$w \sim \exp \left\{ -\frac{2\omega_0}{\omega} f(\gamma) \right\} \quad (7)$$

where

$$f(\gamma) = \left(1 + \frac{1}{2\gamma^2} \sum_{k=1}^{\infty} \frac{f_k^2}{k^2} \right) \tau_0 - \frac{1}{4\gamma^2} \sum_{k=1}^{\infty} \frac{f_k^2}{k^3} \text{sh } 2k\tau_0 - \frac{1}{\gamma^2} \sum_{k>k'} \frac{f_k f_{k'}}{kk'} \left[\frac{\text{sh}(k+k')\tau_0}{k+k'} - \frac{\text{sh}(k-k')\tau_0}{k-k'} \right]. \quad (8)$$

When $f_k = \delta_{k1}$ Eqs. (5) and (8) yield

$$\tau_0 = \text{Arsh } \gamma, \quad f_0(\gamma) = \left(1 + \frac{1}{2\gamma^2} \right) \text{Arsh } \gamma - \frac{\sqrt{1+\gamma^2}}{2\gamma}, \quad (9)$$

which coincides with the result of Keldysh^[1] for the ionization probability in the field of a monochromatic wave. It is clear from (8) that the argument of the exponential in the formula for w is not universal and depends strongly on the concrete form of $F(t)$. This gives rise to the nonlinearity of the problem in question.

Such is the general scheme for calculating the ionization probability in the field of an arbitrary wave $F(t)$ having a period $2\pi/\omega$. Let us consider now several concrete examples.

A. Let $F(t)$ contain, besides the fundamental frequency ω , a small admixture of the k -th harmonic:³⁾

$$F(t) = F[(1 - \alpha) \cos \omega t + \alpha \cos k\omega t], \quad |\alpha| \ll 1. \quad (10)$$

In this case we can expand all quantities in terms of the small parameter α :

$$\tau_0 = \text{Arsh } \gamma - \alpha a_k + O(\alpha^2),$$

$$f(\gamma, \alpha) = f_0(\gamma) - \alpha h_k(\gamma) + O(\alpha^2), \quad (11)$$

where

$$a_k = \frac{1}{\sqrt{1+\gamma^2}} \left[\frac{\text{sh}(k \text{Arsh } \gamma)}{k} - \gamma \right], \quad (12)$$

$$h_k(\gamma) = \frac{1}{k\gamma^2} \left[\frac{\text{sh}((k+1) \text{Arsh } \gamma)}{k+1} - \frac{\text{sh}((k-1) \text{Arsh } \gamma)}{k-1} \right] - \frac{\sqrt{1+\gamma^2}}{\gamma} + \frac{\text{Arsh } \gamma}{\gamma^2}, \quad (13)$$

$$h_k(\gamma) = \begin{cases} \frac{k^2-1}{15} \gamma^3 & \text{for } \gamma \ll 1 \\ \frac{2}{k(k+1)} (2\gamma)^{k-1} & \text{for } \gamma \gg 1 \end{cases}. \quad (13a)$$

We note that $h_k(\gamma) > 0$. Therefore the probability of ionization depends essentially on the phase of the harmonic $k\omega$ relative to the fundamental frequency ω . The reason for this is easily

³⁾The cases of real interest are those with $k = 2$ and 3 . The connection between the polarization vector P and the electric field F is

$$P_t = \alpha_{ij}^{(1)} F_j + \alpha_{ijk}^{(2)} F_j F_k + \alpha_{ijkl}^{(3)} F_j F_k F_l + \dots,$$

where $\alpha^{(n)} \sim F_0^n$. Therefore, if $\alpha^{(2)} \neq 0$, then the most essential is the second-harmonic admixture. However, if the crystal has a symmetry center, then the foregoing series contains only terms of odd order (the case of so-called cubic medium), and the admixture to the fundamental frequency of the laser emission begins with the third harmonic. Generation of the second and third harmonics by a strong light wave passing through transparent media was observed experimentally [6-8]. For these cases, formulas (12) and (13) simplify somewhat:

$$a_2 = \gamma \left(1 - \frac{1}{\sqrt{1+\gamma^2}} \right), \quad a_3 = \frac{4\gamma^3}{3\sqrt{1+\gamma^2}},$$

$$h_2(\gamma) = \frac{2\gamma}{3} - \left(1 + \frac{1}{\gamma^2} \right)^{1/2} + \frac{\text{Arsh } \gamma}{\gamma^2},$$

$$h_3(\gamma) = \frac{2\gamma}{3} \frac{1}{\sqrt{1+\gamma^2}} - \left(1 + \frac{1}{\gamma^2} \right)^{1/2} + \frac{\text{Arsh } \gamma}{\gamma^2}.$$

²⁾It is easy to see that $S(t, 0)$ is a real quantity when $t > 0$.

understood in the adiabatic case ($\gamma \ll 1$), when the ionization occurs essentially at the instants when the field $F(t)$ reaches its maximum value. From (10) we get for $t \rightarrow 0$

$$F(t) = F[1 - 1/2(1 + (k^2 - 1)\alpha)\omega^2 t^2 + \dots] \approx F \cos \omega' t, \quad (14)$$

where ω' is the effective frequency:

$$\omega' = \omega[1 + (k^2 - 1)\alpha]^{1/2} \approx \omega[1 + 1/2(k^2 - 1)\alpha]. \quad (15)$$

When $\alpha > 0$ we have $\omega' > \omega$, and this leads to an increase in the ionization probability:

$$w \sim \exp\left\{-\frac{2}{3}\frac{F_0}{F}g(\gamma + \Delta\gamma)\right\} \\ = \exp\left\{-\frac{2\omega_0}{\omega}\left[f(\gamma) + \frac{(k^2 - 1)}{3}\gamma^2 g'(\gamma)\alpha\right]\right\}. \quad (16)$$

When $\gamma \ll 1$ we get $g'(\gamma) = -\gamma/5$, and (16) coincides with the exact formula (11). In the case of high frequencies (16) strongly undervalues the coefficient $h_k(\gamma)$, but the qualitative picture of the variation of w remains the same for $\alpha > 0$ and $\alpha < 0$.

It must be emphasized that when $\gamma \gg 1$ the ionization probability w becomes very sensitive to small admixtures of higher harmonics in the incident radiation. Thus, for a real case^[9] with $\gamma \sim 30$ and $2\omega_0/\omega \sim 20$ we get $h_2(\gamma) \sim 20$, $h_3(\gamma) \sim 600$, and $f_0(\gamma) = 3.6$. Therefore an admixture of $\sim 10^{-4}$ of the second harmonic and of $\sim 10^{-7}$ of the third harmonic (in terms of intensity) is sufficient to change w by a factor of 10.

B. Assume that a phase shift φ exists between the k -th harmonic and the fundamental frequency:

$$F(t) = F[(1 - \alpha \cos \varphi) \cos \omega t + \alpha \cos(k\omega t + \varphi)]. \quad (17)$$

(the maximum of $F(t)$ is reached when $\omega t_m = -k\alpha \sin \varphi$, the coefficients in (17) being chosen such that $F(t_m) = F[1 + O(\alpha^2)]$). The classical trajectory is determined as before from (3), but the analysis of the subbarrier motion becomes less intuitive, since the "initial instant" τ_0 shifts with the imaginary axis into the complex plane:⁴⁾

$$\tau_0 = i\left\{\text{Arsh } \gamma - \frac{\alpha}{\sqrt{1 + \gamma^2}}\left[\left(\frac{\text{sh}(k \text{Arsh } \gamma)}{k} - \gamma\right) \cos \varphi - i \frac{\text{ch}(k \text{Arsh } \gamma)}{k} \sin \varphi\right] + O(\alpha^2)\right\}. \quad (18)$$

⁴⁾Expression (18) simplifies when $\gamma \gg 1$:

$$\tau_0 \approx i\left[\ln 2\gamma - \frac{\alpha(2\gamma)^{k-1}}{k} e^{-i\varphi} + \dots\right].$$

Calculation of the Action \tilde{S} yields

$$w \sim \exp\left\{-\frac{2\omega_0}{\omega}[f_0(\gamma) - \alpha h_k(\gamma) \cos \varphi + O(\alpha^2)]\right\}. \quad (19)$$

When $\gamma \gg 1$ and $(2\gamma)^{k-1}\alpha \gtrsim \omega/\omega_0$, an admixture of the k -th harmonic greatly changes the ionization probability.

C. In conclusion we consider an example in which the transcendental equation (5) for τ_0 can be solved in explicit form. Let

$$F(t) = F \sum_{k=0}^{\infty} (1 - \alpha) \alpha^k \cos(2k + 1)\omega t \\ = F \frac{(1 - \alpha)^2 \cos \omega t}{1 + \alpha^2 - 2\alpha \cos 2\omega t} \quad (20)$$

($0 < \alpha < 1$). With increasing α , the function $F(t)$ becomes "more peaked."⁵⁾ From (3) we get

$$\dot{x}(t) = \frac{F}{\omega} \sum_{k=0}^{\infty} \frac{(1 - \alpha) \alpha^k}{2k + 1} \sin(2k + 1)\omega t = \frac{F}{\omega \beta} \text{arctg}(\beta \sin \omega t), \quad (21)$$

$$\tau_0 = \text{Arsh}\left(\frac{\text{th}(\beta\gamma)}{\beta}\right), \quad \beta = \frac{2\sqrt{\alpha}}{1 - \alpha}. \quad (22)$$

The function $f(\gamma)$ in formula (7) for the ionization probability now takes the form

$$f(\gamma, \alpha) = \int_0^{\tau_0} \left\{1 - \left[\frac{\text{Arth}(\beta \text{sh } \tau)}{\beta\gamma}\right]^2\right\} d\tau \\ = \int_0^{x_0} \frac{dx}{\sqrt{x^2 + \beta^2}} \left[1 - \left(\frac{\text{Arth } x}{\text{Arth } x_0}\right)^2\right] \quad (23)$$

where $x_0 = \tanh(\gamma) < 1$. From this we get approximate formulas for $f(\gamma, \alpha)$ in the two limiting cases:⁶⁾

$$f(\gamma, \alpha) = \begin{cases} f_0(\gamma) + \alpha h_3(\gamma) + \dots & \text{for } \beta\gamma \ll 1 \\ \text{Arsh } \beta^{-1} - 1,05(\beta\gamma)^{-2} + \dots & \text{for } \beta\gamma \gg 1 \end{cases} \quad (24)$$

The plot of $f(\gamma, \alpha)$ against γ in Fig. 2 shows that ionization in the field (20) always exceeds ionization in the field of a monochromatic wave having the same amplitude and period. The value of w

⁵⁾As $\alpha \rightarrow 1$ we get

$$\lim_{\alpha \rightarrow 1} \frac{1 - \alpha^2}{1 + \alpha^2 - 2\alpha \cos x} = 2\pi\delta(x).$$

⁶⁾We use here the value of the integral:

$$\int_0^1 \frac{(\text{Arth } x)^2}{x} dx = \frac{7}{8} \zeta(3) = 1,052.$$

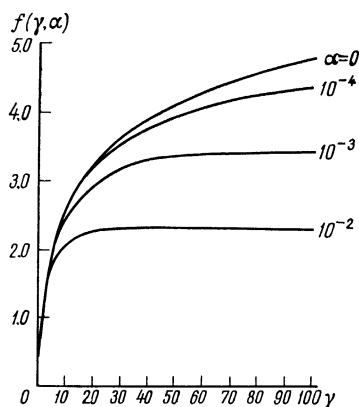


FIG. 2. The function $f(\gamma, \alpha)$ for the example (20) at different values of the parameter α . The ratio of the third harmonic intensity to that of the fundamental frequency is α^2 .

differs noticeably from that of the case $\alpha = 0$ when $\gamma \gtrsim \alpha^{-1/2}$.

Let us compare now formulas (7) and (8) for w with the result of the exact quantum mechanical calculation. The expression for the ionization probability of a bound level under the influence of an arbitrary electric field $F(t)$ with period $T = 2\pi/\omega$ can be obtained⁷⁾ by the method described in [4] for a monochromatic wave. Confining ourselves for simplicity to the case of a wave with linear polarization, we have

$$w = \sum_{n \geq \nu} w_n(F, \omega), \quad (25)$$

$$\nu = \frac{\omega_0}{\omega} \left(1 + \frac{1}{2\gamma^2} \sum_{k=1}^{\infty} \frac{f_k^2}{k^2} \right);$$

$$w_n(F, \omega) = 2\pi \int d\mathbf{p} \delta \left(\frac{p^2 - p_n^2}{2} \right) |F_n(\mathbf{p})|^2.$$

$$p_n = \sqrt{2\omega(n - \nu)}. \quad (26)$$

The coefficients $F_n(\mathbf{p})$, which determine the partial probability of ionization with absorption of n quanta, is obtained from the Fourier-series expansion

$$\begin{aligned} \chi_{lm}(\boldsymbol{\pi}(t)) \exp \left\{ \frac{i}{2} \left[(p^2 + \kappa^2)t + 2\mathbf{p}\boldsymbol{\xi}(t) + \int_0^t A^2(t') dt' \right] \right\} \\ = \exp \left\{ \frac{i}{2} (p^2 + \kappa^2 + \bar{A}^2)t \right\} \sum_{n=-\infty}^{\infty} F_n(\mathbf{p}) e^{-in\omega t}, \end{aligned} \quad (27)$$

$$\bar{A}^2 = \lim_{T \rightarrow \infty} \frac{1}{T} \int_0^T A^2(t') dt' = \frac{\kappa^2}{2\gamma^2} \sum_{k=1}^{\infty} \frac{f_k^2}{k^2} \quad (28)$$

(see [4, 5] for the notation).

From this we can easily obtain for $F_n(\mathbf{p})$ a formula similar to formula (45) of [5]:

$$\begin{aligned} F_n(\mathbf{p})|_{\nu=p_n} = \frac{e^{i\mathbf{p}\boldsymbol{\xi}(0)}}{2\pi} \int_{-\pi}^{\pi} d(\omega t) \chi_{lm}(\boldsymbol{\pi}(t)) \\ \times \exp \left\{ i \frac{\omega_0}{\omega} \int_0^{\omega t} \left[1 + \frac{\pi^2(\alpha)}{\kappa^2} \right] d\alpha \right\}. \end{aligned} \quad (29)$$

Owing to the rapidly oscillating exponential (when $\omega \ll \omega_0$), the integral in (29) is "concentrated" at the saddle point:⁸⁾ $\pi^2(t_0) + \kappa^2 = 0$ or

$$A(t_0) = -\frac{F}{\omega} \sum_{k=1}^{\infty} \frac{f_k}{k} \sin k\omega t_0 = \mp i(\kappa^2 + p_{\perp}^2)^{1/2} + p_z \quad (30)$$

(here $\mathbf{p} = (p_{\perp}, p_z)$) is the time-averaged momentum of the emitted electron. Comparison of (29) and (30) with (6) and (7) shows that the factor $\chi_{lm}(\boldsymbol{\pi}(t))$, which contributes to the multiplier preceding the exponential, is neglected in the quasiclassical method. In addition, we have considered above only the extremal classical trajectory ($\mathbf{p} = 0$). This limitation is not essential: as shown in [5], the contribution of trajectories close to the extremal one can be taken into account within the framework of the quasiclassical method, and a correct law for the decrease of w_n with increasing n can be obtained. Thus, the calculation of $w_n(F, \omega)$ by the saddle point method corresponds fully⁹⁾ to the quasiclassical approximation in the ionization problem. The latter, however, introduces into the consideration classical trajectories and has the advantage of being clearer.

2. ALLOWANCE FOR THE COULOMB INTERACTION

In the earlier calculations^[3-5] we neglected the Coulomb interaction between the electron and the atomic remainder, i.e., the obtained formulas pertained essentially to the case of ionization of negative ions:

$$A^- + n\hbar\omega \rightarrow A + e^-.$$

However, it is clear already from the formula for the ionization probability in the constant field^[10] that it is important to take into account the Coulomb correction:

⁸⁾We note that the saddle point t_0 coincides with the "initial instant of time" for the subbarrier motion (see Fig. 1).

⁹⁾It is important here to take also into account the fact that owing to the quantum character of the light absorption, emitted electrons can only have discrete momenta $p = p_n = [2\omega(n - \nu)]^{1/2}$.

⁷⁾Neglecting the Coulomb interaction between the electron and the atomic remainder.

$$w_{\text{stat}}(F) = \omega_0 |C_{kl}|^2 \left(\frac{2F_0}{F} \right)^{2\lambda-1} e^{-2F_0/3F},$$

$$F_0 = \kappa^2, \quad \lambda = \kappa_c / \kappa. \quad (31)$$

Neglect of the Coulomb interaction corresponds to $\lambda = 0$. In real experiments¹⁰⁾ $2F_0/F \sim 10^3$ and $\lambda \sim 1$, i.e., allowance for the Coulomb interaction introduces a factor $\sim 10^6$ into $w_{\text{stat}}(F)$.

In the region $\kappa r \gg 1$, the intraatomic potential reduces to the Coulomb potential: $V(r) \sim -\kappa_c/r$, with $|V(r)| \ll \omega_0$. This makes it possible to take into account the influence of the Coulomb interaction by perturbation theory (see a similar analysis for the case of a constant field^[12]). We represent the contracted action \tilde{S} in the form

$$\tilde{S}(t_0, 0) = \int_{t_0}^0 \left[\frac{\dot{x}^2}{2} - V(x, t) - \frac{\kappa^2}{2} \right] dt = S_0 + \delta S;$$

$$V(x, t) = -Fx \cos \omega t + \delta V(x),$$

$$\delta V(x) \approx -\kappa_c/x \text{ for } \kappa x \gg 1. \quad (32)$$

Here \tilde{S}_0 denotes the action calculated for the short-range potential ($\lambda = 0$), and $\delta\tilde{S}$ is the Coulomb correction. Making allowances in (32) for the first-order variations in δV , we get

$$\delta\tilde{S} = \int_{t_0}^0 \left(\dot{x} \delta \dot{x} - \frac{\partial V_0}{\partial x} \delta x - \delta V \right) dt - \left[\frac{\dot{x}^2}{2} - V_0 - \frac{\kappa^2}{2} \right]_{t=t_0} \delta t_0. \quad (33)$$

Using the equation of the unperturbed trajectory $\ddot{x} = -\partial V_0(x, t)/\partial x$ and the condition $\dot{x}(0) = 0$, we transform (33) into

$$\delta\tilde{S} = - \int_{t_0}^0 \delta V(x(t)) dt$$

$$- \left[\dot{x} \delta \dot{x} + \left(\frac{\dot{x}^2 - \kappa^2}{2} - V_0(x, t) \right) \delta t \right]_{t=t_0}. \quad (33a)$$

The perturbed trajectory $\tilde{x}(t) = x(t) + \delta x(t)$ satisfies the condition $\tilde{x}(t_0 + \delta t_0) = 0$, which leads to a connection between the variation δx and δt :

$$\delta x(t_0) = -\dot{x}(t_0) \delta t_0. \quad (34)$$

Substituting this in (33a), we get

$$\left[\dot{x} \delta \dot{x} + \left(\frac{\dot{x}^2 - \kappa^2}{2} - V_0(x, t) \right) \delta t \right]_{t=t_0}$$

$$= [E_0 - H(x, t)]_{t=t_0} \delta t_0 = 0,$$

where account is taken of the equation for the determination of the "initial instant" t_0 (see formula (39) of [5]). Ultimately we get

$$\delta\tilde{S} = - \int_{t_0}^0 \delta V(x(t)) dt, \quad (35)$$

where $x(t)$ is the unperturbed trajectory (without allowance for δV).

Let us consider first the case of linear polarization (the electric field $F(t) = F \cos \omega t$ is directed along the x axis). Then^[5]

$$x(t) = \frac{F}{\omega^2} (\cos \omega t_0 - \cos \omega t), \quad \omega t_0 = i \operatorname{Arsh} \gamma. \quad (36)$$

Formula (35) can be used if the perturbing potential $\delta V(r)$ is small along the entire classical trajectory. In our case this is not so when $\kappa r \lesssim 1$; this region of values of r cannot be considered quasiclassically at all.

We therefore use a joining-together procedure: we introduce x_1 such that^[11]

$$1 \ll \kappa x_1 \ll \kappa x_0 = F_0/F (1 + \sqrt{1 + \gamma^2}) \quad (37)$$

(x_0 is the "dynamic" width of the barrier, see [5]). When $1 \ll \kappa r \ll \kappa x_0$, the intraatomic potential is already small, but the external field can still be neglected. Under these conditions, the wave function $\psi(r)$ coincides with the asymptotic wave function of the free atom:

$$\psi(r) \sim C_{\kappa} \kappa^{3/2} (\kappa r)^{\lambda-1} e^{-\kappa r} \sim r^{-1} \exp\{-\operatorname{Im} \tilde{S}(r)\}, \quad (38)$$

where

$$\operatorname{Im} \tilde{S}(r) = \kappa r - \lambda \ln \kappa r$$

(see the Appendix in [5]). Therefore (35) can be transformed into

$$\delta\tilde{S} = -i\lambda \ln \kappa x_1 - \int_{t_1}^0 \delta V(x(t)) dt, \quad x_1 = x(t_1) \quad (39)$$

(the arbitrary length x_1 drops out of the final answer). The integral in (39), in the case of a Coulomb perturbation, can be evaluated exactly:

$$\int_{t_1}^0 \delta V(x(t)) dt = i\lambda \gamma \int_0^{\omega t_1} \frac{d\tau}{\operatorname{ch} \tau_0 - \operatorname{ch} \tau}$$

$$= i\lambda \ln \frac{\operatorname{th}(\tau_0/2) + \operatorname{th}(\tau_1/2)}{\operatorname{th}(\tau_0/2) - \operatorname{th}(\tau_1/2)},$$

¹⁰⁾We present the values of the parameter λ for atoms of several transparent gases: $\lambda = 1.00$ for H, 0.74 for He, 0.93 for Ar, and 1.06 for Xe, etc. (see the table in [1]).

¹¹⁾ x_0 decreases monotonically with increasing γ . When $\gamma \gg 1$ we have $\kappa x_0 \approx F_0/F \gamma = 2\omega_0/\omega$. Therefore the condition $\omega \ll \omega_0$ makes it possible to choose a joining point x_1 satisfying the inequalities (37).

where $\tau_1 = \omega t_1$ and $\tau_0 = \omega t_0 = \sinh^{-1} \gamma$. Hence

$$\delta \mathcal{S}(t_0, 0) = -i\lambda \ln \left[\kappa x_1 \operatorname{sh} \left(\frac{\tau_0 + \tau_1}{2} \right) \middle/ \operatorname{sh} \left(\frac{\tau_0 + \tau_1}{2} \right) \right]. \quad (40)$$

From (36) we get as $t \rightarrow t_0$

$$\kappa x(t) = \frac{\kappa F}{\omega^2} \operatorname{sh} \tau_0 (\tau_0 - \tau) + \dots = \frac{2\omega_0}{\omega} (\tau_0 - \tau) \quad (41)$$

($\tau = -i\omega t$). Owing to the factor $2\omega_0/\omega \gg 1$, there exist instants τ_1 such that $(\tau_0 - \tau_1) \ll 1$ and $\kappa x_1 \gg 1$. It is therefore possible to take in (40) a limit as $\tau_1 \rightarrow \tau_0$. This yields

$$\begin{aligned} \delta \mathcal{S}(t_0, 0) &= -i\lambda \ln \left(\frac{2F_0}{F} \right), \\ \exp\{-2 \operatorname{Im} \delta \mathcal{S}\} &= \left(\frac{2F_0}{F} \right)^{2\lambda}. \end{aligned} \quad (42)$$

The dependence of $\delta \tilde{S}$ on the frequency has dropped out, i.e., the Coulomb correction in the ionization probability $w(F, \omega)$ has the same form as for a static field (see (31)).

The Coulomb correction for the ionization by elliptically polarized light is calculated in similar fashion. In this case the potential $\delta V(r) = -\kappa_C/r$ must be averaged along the two-dimensional trajectory

$$\begin{aligned} x(t) &= \frac{F}{\omega^2} (\cos \omega t_0 - \cos \omega t), \\ y(t) &= \frac{F}{\omega^2} \left(\gamma \frac{k_0}{\kappa} \omega t - \varepsilon \sin \omega t \right) \end{aligned} \quad (43)$$

(here k_0 is the average momentum of the outgoing electrons; its value depends on γ and on the ellipticity ε —see Fig. 2 of [5]). We note that when $t = i\tau$ the coordinate $y(t)$ becomes purely imaginary, and therefore $\delta V(r)$ must be defined as follows:

$$\begin{aligned} \delta V(r(t)) &= -\kappa_C (x^2 + y^2)^{-1/2} \\ &= -\lambda \gamma \omega \left[(\operatorname{ch} \tau_0 - \operatorname{ch} \tau)^2 - \varepsilon^2 \tau^2 \left(\frac{\operatorname{sh} \tau_0}{\tau_0} - \frac{\operatorname{sh} \tau}{\tau} \right)^2 \right]^{-1/2}. \end{aligned} \quad (44)$$

It can be shown that the radicand in (44) is positive. It is now necessary to calculate the contribution made to $\delta \operatorname{Im} \tilde{S}$ by the region $\kappa r \gg 1$. To this end, we choose τ_1 such that $(\tau_0 - \tau_1) \ll 1$ and $\kappa r_1 \gg 1$, and join at the point τ_1 to the internal solution (38). The arbitrary point τ_1 drops out of the final answer; as a result we arrive at the following formula for the ionization probability with allowance for the Coulomb correction:

$$w(F, \omega, \varepsilon) = \left[\frac{2F_0}{F} C(\gamma, \varepsilon) \right]^{2\lambda} w_{\text{s.r.}}(F, \omega, \varepsilon), \quad (45)$$

where $w_{\text{s.r.}}$ is the ionization probability for a short-range potential ($\lambda = 0$),

$$C(\gamma, \varepsilon) = \frac{\tau_0}{2\gamma} \exp \left\{ \int_0^{\tau_0} d\tau \left[\frac{\gamma}{F(\gamma, \varepsilon)} - \frac{1}{\tau_0 - \tau} \right] \right\}, \quad (46)$$

$$F(\gamma, \varepsilon) = \left[(\operatorname{ch} \tau_0 - \operatorname{ch} \tau)^2 - \varepsilon^2 \tau^2 \left(\frac{\operatorname{sh} \tau_0}{\tau_0} - \frac{\operatorname{sh} \tau}{\tau} \right)^2 \right]^{1/2}. \quad (47)$$

The integral in (46) converges. The ‘‘initial instant’’ $t_0 = \tau_0/\omega$ is determined from the transcendental equation

$$\operatorname{sh}^2 \tau_0 - \varepsilon^2 \left(\operatorname{ch} \tau_0 - \frac{\operatorname{sh} \tau_0}{\tau_0} \right)^2 = \gamma^2, \quad (48)$$

which can be obtained from Eqs. (18) of [5]. The quantity t_0 has the meaning of the total time of electron motion under the barrier. A plot of $t_0(\gamma, \varepsilon)$ is shown in Fig. 3.

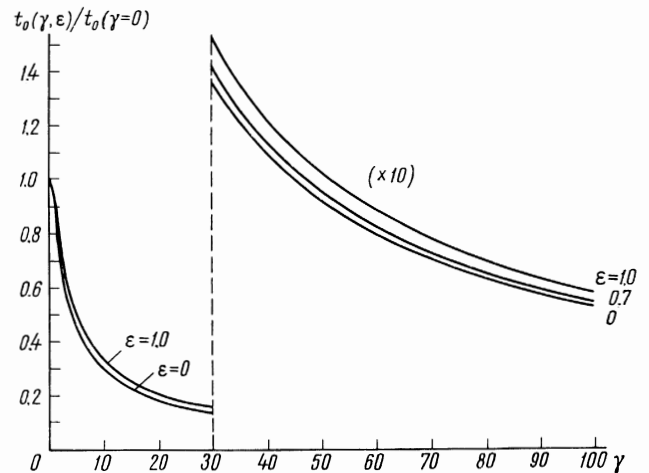


FIG. 3. Ratio $t_0(\gamma, \varepsilon)/t_0(\gamma=0)$ vs. γ . With increasing γ , the time of flight t_0 through the barrier becomes shorter. The ordinate scale is increased by a factor of 10 for $\gamma > 30$.

The function $C(\gamma, \varepsilon)$ was calculated numerically by means of (46)–(48). The results are shown in Fig. 4 for different values of the ellipticity ε . In the adiabatic region ($\gamma \ll 1$) the Coulomb correction does not depend on ε .¹²⁾ For the ionization probability of unpolarized atoms we obtain the following formula:

$$\begin{aligned} w(F, \omega, \varepsilon) &= \omega_0 |C_{\kappa i}|^2 \left[\frac{6}{\pi(1-\varepsilon^2)} \right]^{1/2} \left(\frac{2F_0}{F} \right)^{2\lambda-3/2} \\ &\times \exp \left\{ -\frac{2}{3} \frac{F_0}{F} \left[1 - \frac{1}{10} \left(1 - \frac{\varepsilon^2}{3} \right) \gamma^2 + \dots \right] \right\}. \end{aligned} \quad (49)$$

¹²⁾The reason is that, regardless of the value of ε , the electron trajectory does not have time to bend during the time of flight of the particle through the barrier.

Process	ω_0 , eV	ω_0/ω	λ	F , 10^7 V/cm	γ	γ_c	σ	Experiment
$\text{Xe} \rightarrow \text{Xe}^+ + e$	12.13	6.81	1.06	1.3 ± 0.3	24.5	17.8	1.90	[9]
$\text{H}_2 \rightarrow \text{H}_2^+ + e$	15.43	8.65	0.94	1.1 ± 0.3	32.7	24.6	1.77	[13]

If the polarization of the light is very close to circular, the factor $[3F/\pi(1-\epsilon^2)F_0]^{1/2}$ should be replaced by $A(F, \epsilon)$ (see formula (10) of [4]).

In the region $\gamma \gtrsim 1$ it is necessary to take into account in the pre-exponential factor the function $C(\gamma, \epsilon)$. However, as seen from Fig. 4, this yields only a numerical factor on the order of unity up to $\gamma = 30$, which can be neglected, since the formula for w contains at any rate the unknown constant C_{KL} . Thus, the influence of the ellipticity ϵ of the light on the pre-exponential factor is insignificant (unlike the argument of the exponential, in which the dependence on ϵ is strong and must be taken into account—see [5]).

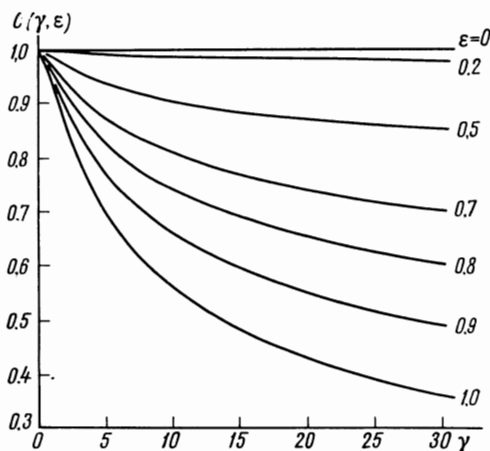


FIG. 4. Plot of the function $C(\gamma, \epsilon)$ for different values of the ellipticity ϵ .

We note now that the method used above for taking into account the Coulomb interaction is not always exact. We started from the assumption that since the Coulomb potential is small when $\kappa r \gg 1$, its influence on the trajectory of the subbarrier motion is also small (in determining this trajectory we took into account only the external field $F(t)$, see (36) and (43)). Let us compare the Coulomb force and the intensity of the external field in the final point of the trajectory:

$$\frac{F_c}{F} \Big|_{x=x_0} = \frac{\lambda F}{F_0} (1 + \sqrt{1 + \gamma^2})^2. \quad (50)$$

For a constant field

$$\frac{F_c}{F} \Big|_{x=x_0} = \frac{4\lambda F}{F_0} \ll 1, \quad (50a)$$

therefore the main part of the subbarrier trajectory passes through a region where the external field is stronger than the Coulomb field, so that the Coulomb potential can be regarded as a small perturbation. The same holds also in the adiabatic case $\gamma \ll 1$.

However, with increasing γ , the dynamic barrier width x_0 decreases, and the ratio (50) increases. We denote by γ_c that value of the parameter γ at which (50) becomes equal to unity:

$$\gamma_c = \sqrt{F_0/\lambda F} \quad (\gamma_c \gg 1). \quad (51)$$

To obtain the subbarrier trajectory when $\gamma \gtrsim \gamma_c$, it is necessary to solve the nonlinear equation¹³⁾

$$\ddot{x} = -\kappa_c/x^2 + F \cos \omega t \quad (52)$$

in the region $x > x_1$ (x_1 is the joining point, $\kappa x_1 \gg 1$). Going over to the dimensionless variables $\tau = -i\omega t$ and $\xi = \omega x/\kappa$, we transform (52) into

$$\frac{d^2\xi}{d\tau^2} = \frac{1}{\gamma} \left(\frac{\sigma}{\xi^2} - \text{ch } \tau \right), \quad \sigma = \left(\frac{\gamma}{\gamma_c} \right)^2 = \lambda \left(\frac{\omega}{2\omega_0} \right)^2 \frac{F_0}{F}. \quad (53)$$

For the unperturbed trajectory ($\sigma \rightarrow 0$) the dynamic barrier width is

$$\xi_0 = \frac{\gamma}{1 + \sqrt{1 + \gamma^2}} \rightarrow 1 \quad \text{for } \gamma \gg 1. \quad (54)$$

A numerical solution of (53), obtained by A. S. Kronrod for several values of the parameters γ and σ , has shown that when $s \sim 0.9-1.0$ a qualitative change takes place in the form of the subbarrier trajectory, in that a section of "retrograde motion" appears on it (see Fig. 5). Therefore allowance for the Coulomb interaction when $\gamma \gtrsim \gamma_c$ calls for a separate analysis.

Thus, the condition under which formula (45) for the Coulomb correction is applicable is $\sigma \ll 1$. Inasmuch as the light emitted by a laser has a

¹³⁾Equation (52) gives the extremal classical trajectory for motion in the field of a linearly polarized wave. (We assume that $p_{\perp} = 0$, so that the motion reduces to one-dimensional. Allowance for p_{\perp} does not change the principal term of the exponential for the ionization probability).

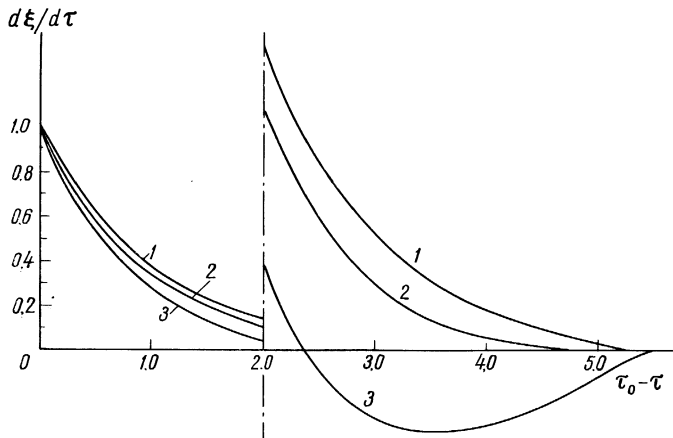


FIG. 5. Plot of the velocity of the subbarrier motion at $\gamma = 100$, $\sigma = 0$ (curve 1), $\sigma = 0.92$ (curve 2), and $\sigma = 1.90$ (curve 3). The values of τ_0 for curves 1, 2, and 3 respectively are 5.30, 5.35, and 5.46. The ordinate scale for $\tau_0 - \tau > 2$ has been increased ten times.

fixed frequency, this condition is best rewritten in the form

$$F \gg \lambda (\omega/\omega_0)^2 F_0 \quad (55)$$

which shows that (45) pertains to the case of sufficiently strong fields. The values of the parameters γ , γ_C , etc. for the presently performed experiments on ionization of atoms by laser light are listed in the table. It is clear from these data that for a detailed comparison of theory and experiment it is necessary to take into account exactly the Coulomb interaction with the aid of (52) and (53).

M. V. Terent'ev took part in the initial stage of the present work. The authors are sincerely grateful to him for numerous useful discussions, and also for a check on the calculations of Sec. 2.

We also wish to thank A. S. Kronrod and L. Borisov for the numerical calculations.

Note added in proof (10 January 1967). Formula (35) makes it possible to take into account the Coulomb correction not only to the total ionization probability, but also to the momentum spectrum of the emitted electrons. Substituting in (39) trajectory $x(t)$ for $p_x \neq 0$ (see formula (48) of [5]), we arrive at an integral of the type (46), where $F(\gamma, \epsilon)$ must be replaced by $\cosh \tau_0 - \cosh \tau - i\gamma p \kappa^{-1}(\tau_0 - \tau)$, where $\tau_0 = \sinh^{-1} \gamma(1 + i p \kappa^{-1})$. This integral can be calculated in the limiting cases $\gamma \ll 1$ and $\gamma \gg 1$. In the case $\gamma \gg 1$, the distribution over p_x is of the form

$$w(p_x) \sim \exp \left\{ -\frac{2\omega_0}{\omega} \tau_0 \left[1 + \frac{\sigma}{\gamma} \left(\frac{2}{3} \tau_0^2 - 2\tau_0 + 4 \right) \right] \frac{p_x^2}{\kappa^2} \right\},$$

where $\tau_0 = \ln 2\gamma$ and σ is defined in (53). The Coulomb interaction leads to a small decrease in the width of the momentum spectrum. The most probable value of the momentum, $p_x = 0$, remains unchanged here.

¹ L. V. Keldysh, JETP 47, 1945 (1964), Soviet Phys. JETP 20, 1307 (1965).

² A. Gold and H. B. Bebb, Phys. Rev. Lett. 14, 60 (1965); Phys. Rev. 143, 1 (1966).

³ A. I. Nikishov and V. I. Ritus, JETP 50, 255 (1966), Soviet Phys. JETP 23, 168 (1966).

⁴ A. M. Perelomov, V. S. Popov, and M. V. Terent'ev, JETP 50, 1393 (1966), Soviet Phys. JETP 23, 924 (1966).

⁵ A. M. Perelomov, V. S. Popov, and M. V. Terent'ev, JETP 51, 309 (1966), Soviet Phys. JETP 24, 207 (1967).

⁶ S. A. Akhmanov and R. V. Khokhlov, Problemy nelineinoi optiki (Problems of Nonlinear Optics), Published by Information Institute, 1965. N. Bloembergen, Nonlinear Optics, Benjamin, 1965.

⁷ R. C. Miller, D. A. Kleinman, and A. Savage, Phys. Rev. Lett. 11, 146 (1963); C. K. N. Patel, *ibid.* 16, 613 (1966).

⁸ R. W. Terhune, P. D. Maker, and C. M. Savage, Phys. Rev. Lett. 8, 404 (1962).

⁹ G. S. Voronov, and N. B. Delone, JETP Letters 1, No. 2, 42 (1965), transl. 1, 66 (1965); JETP 50, 78 (1966), Soviet Phys. JETP 23, 54 (1966).

¹⁰ B. M. Smirnov and M. I. Chibisov, JETP 49, 841 (1965), Soviet Phys. 22, 585 (1966).

¹¹ A. M. Perelomov, V. S. Popov, and M. V. Terent'ev, Preprint, Institute of Theoretical and Experimental Physics, No. 409 (1966).

¹² L. D. Landau and E. M. Lifshitz, Kvantovaya mekhanika (Quantum Mechanics), Fizmatgiz, 1963.

¹³ G. S. Voronov, G. A. Delone, N. B. Delone, and O. V. Kudrevatova, JETP Letters 2, 377 (1965), transl. p. 237.

Translated by J. G. Adashko