Effect of an electric field on the ionization of atoms by electrons

V. P. Demkin

V. V. Kuĭbyshev State University, 634050 Tomsk, Russia

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A method is developed for calculating the probability amplitude of the ionization of atoms by electrons in an electric field. The wave functions of the initial and final scattering channels are calculated in the approximation of a "frozen" ion core. The effect of the electric field on the ionization amplitude is taken into account in the first order perturbation theory for the Green function. Estimates are made of the distortion factor $\eta_k(\mathbf{E})$, and it is shown that near the ionization threshold at a field strength E of order of 10–100 kV cm^{-1} the value of $\eta_k(\mathbf{E})$ can be large.

The kinetics of electron-atom collision processes in an electric field are due to the effects of polarization of atoms and the distortions in the waves of the impinging and scattered electrons. As shown in Ref. 1, for atomic excitation by electrons these effects separate in the expression for the scattering amplitude. The distortion factor can be calculated separately as a correction to the amplitude of scattering on a Stark state. And to calculate the scattering amplitude in the basis of Stark functions one can use methods well-known from the literature.[2]

Several difficulties emerge in the problem of electron-induced ionization of an atom because the wave function of the initial and final scattering channels belong to different Hamiltonians.[3,4] Extending the respective methods used in calculating the ionization amplitude[5] to this process in an electric field leads to incorrect accounting for the field effect.

Kang and Foland[6] developed a method for calculating the amplitude of electron-induced ionization of the hydrogen atom using the Coulomb functions as a basis. This method makes it possible to allow for exchange effects correctly and is free of the deficiencies inherent in other calculation methods.

Here we expand this method to the case of ionization of complex atoms and within the new method take consistent account of the effect of the field on ionization.

Let us consider the ionization of an atom whose initial state has a configuration $l^N l'$, where l^N is the configuration of the ion core, and l' represents a peripheral, or optical electron. We write the wave function representing the electron plus atom system as an expansion in the atomic states $\Phi_n^{(a)}(\xi)$:

$$\Psi=\sum_n \Psi_n^{(a)}(\xi)\phi_n(\mathbf{r}), \tag{1}$$

where $\phi_n(\mathbf{r})$ is the wave function of the incident electron, and the sum incorporates integration over the continuous-spectrum states.

Next in $\Psi_n^{(a)}(\xi)$ we isolate an electron in the (nl') state using the well-known formulas[7]

$$\begin{aligned}\Psi^{(a)}&=\sum_i \frac{(-1)^{N+1-i}}{\sqrt{N+1}}\,\Psi(l^N L_1 S_1, l' LS)\\ &=\sum_i\sum_m C^{LM_L}_{M_{L_1}m_{l'}} C^{SM_S}_{M_{S_1}m_{s'}} \frac{(-1)^{N+1-i}}{\sqrt{N+1}}\\ &\quad\times\Psi_{L_1S_1}(\xi_1,\ldots,\xi_{N+1},\ldots,\xi_N)\Psi_{nl'}(\xi_i),\end{aligned} \tag{2}$$

where $C^{LM_L}_{M_{L_1}m_{l'}}$ and $C^{SM_S}_{M_{S_1}m_{s'}}$ are the Clebsch–Gordan coefficients, $\Psi_{L_1S_1}(\xi_1,\ldots,\xi_{N+1},\ldots,\xi_N)$ is the wave function of the ion core, with the coordinate of the ith electron replaced by the coordinate of the $(N+1)$ electron, $\Psi_{nl'}(\xi_i)$ is the wave function of the optical electron, and the sum over m is understood to be over the quantum numbers M_{L_1}, $m_{l'}$, M_{S_1}, and $m_{s'}$.

Let us assume that only valence electrons participate in ionization. This means that the subscript "n" in Eq. (2) is assigned only to the wave function of an optical electron. In this case, to find the discrete and continuous spectra of states $\Psi_{nl'}(\xi_i)$ it is expedient to employ the method of the "frozen" ion core,[8,9] which guarantees that the $\Psi_{nl'}(\xi_i)$ are orthogonal to the ion-core functions. We write the Hamiltonian in Eq. (1) in the form

$$H=H_i+K_{N+1}+V^*_{N+1}+K_r+V^*_r+V(\mathbf{r},\xi), \tag{3}$$

where H_i is the the ion-core Hamiltonian, K_{N+1} and K_r the kinetic-energy operators of the optical and incident electrons,

$$V^*_{n+1}=-\frac{Ze^2}{\xi_{N+1}}+\sum_{i=1}^{N}\frac{e^2}{|\xi_{N+1}-\xi_i|}$$

the potential energy of the interaction between the optical electron and the core,

$$V^*_r=-\frac{Ze^2}{r}+\sum_{i=1}^{N}\frac{e^2}{|\mathbf{r}-\xi_i|}$$

the potential energy of the interaction between the incident electron and the core, and

$$V(\mathbf{r},\xi)=\frac{e^2}{|\mathbf{r}-\xi_{N+1}|} \tag{4}$$

the potential energy of the interaction between the optical and incident electrons. We substitute (2) into the Schrödinger equation, multiply the left- and right-hand sides of (1) by the conjugate wave function of the ion core in the ground state, $\Psi^*{}_{L_1S_1}(\xi_1,\ldots,\xi_N)$, and integrate over the $\xi_1,\xi_2,\ldots,\xi_N$:

$$\begin{aligned}
&\sum_{i,n}\sum_m C^{LM_L}_{M_{L_1}m_{l'}} C^{SM_S}_{M_{S_1}m_s} \frac{(-1)^{N+1-i}}{\sqrt{N+1}} \\
&\quad\times\int d\xi_1\cdots d\xi_N\,\Psi^*{}_{L_1S_1}(\xi_1,\ldots,\xi_N) \\
&\quad\times[H_{\mathrm{i}}+K_{N+1}+V^*_{N+1}+K_r+V^*_r+V(\mathbf{r},\xi)] \\
&\quad\times\Psi_{L_1S_1}(\xi_1,\ldots,\ \xi_{N+1},\ldots,\xi_N)\Psi_{nl'}(\xi_i)\phi_n(\mathbf{r}) \\
&\quad=\mathscr{E}\sum_{i,n}\sum_m C^{LM_L}_{M_{L_1}m_{l'}} C^{SM_S}_{M_{S_1}m_{s'}} \frac{(-1)^{N+1-i}}{\sqrt{N+1}} \\
&\quad\times\int d\xi_1\cdots d\xi_N\,\Psi^*{}_{L_1S_1}(\xi_1,\ldots,\xi_N) \\
&\quad\times\Psi_{L_1S_1}(\xi_1,\ldots,\ \xi_{N+1},\ldots,\xi_N)\Psi_{nl'}(\xi_i)\phi_n(\mathbf{r}). \qquad (5)
\end{aligned}$$

Next we isolate in (5) the term with $i=N+1$:

$$\begin{aligned}
&\sum_n\sum_m C^{LM_L}_{M_{L_1}m_{l'}} C^{SM_S}_{M_{S_1}m_{s'}} \frac{1}{\sqrt{N+1}}[(\varepsilon_{L_1S_1}-\mathscr{E}) \\
&\quad\times\langle L_1S_1M'_{L_1}M'_{S_1}|L_1S_1M_{L_1}M_{S_1}\rangle\Psi_{nl'}(\xi_{N+1})\phi_n(\mathbf{r}) \\
&\quad+\langle L_1S_1M'_{L_1}M'_{S_1}|L_1S_1M_{L_1}M_{S_1}\rangle K_{N+1}\Psi_{nl'}(\xi_{N+1}) \\
&\quad\times\phi_n(\mathbf{r})+\langle L_1S_1M'_{L_1}M'_{S_1}|L_1S_1M_{L_1}M_{S_1}\rangle \\
&\quad\times K_r\Psi_{nl'}(\xi_{N+1})\phi_n(\mathbf{r})+\langle L_1S_1M'_{L_1}M'_{S_1}|V^*_{N+1}| \\
&\quad\times L_1S_1M_{L_1}M_{S_1}\rangle\Psi_{nl'}(\xi_{N+1})\phi_n(\mathbf{r}) \\
&\quad+\langle L_1S_1M'_{L_1}M'_{S_1}|V^*_r|L_1S_1M_{L_1}M_{S_1}\rangle\Psi_{nl'}(\xi_{N+1}) \\
&\quad\times\phi_n(\mathbf{r})+\langle L_1S_1M'_{L_1}M'_{S_1}|V|L_1S_1M_{L_1}M_{S_1}\rangle \\
&\quad\times\Psi_{nl'}(\xi_{N+1})\phi_n(\mathbf{r})] \\
&\quad=-\sum_{i\neq N+1}\sum_n\sum_m C^{LM_L}_{M_{L_1}m_{l'}} C^{SM_S}_{M_{S_1}m_{s'}} \frac{(-1)^{N+1-i}}{\sqrt{N+1}} \\
&\quad\times\int d\xi_1\cdots d\xi_N\,\Psi^*{}_{L_1S_1}(\xi_1,\ldots,\xi_N)(H-\mathscr{E}) \\
&\quad\times\Psi_{L_1S_1}(\xi_1,\ldots,\ \xi_{N+1},\ldots,\ \xi_N)\Psi_{nl'}(\xi_i)\phi_n(\mathbf{r}). \qquad (6)
\end{aligned}$$

To obtain an equation for $\phi_n(\mathbf{r})$, we multiply (6) by $\Psi^*{}_{ql''}(\xi_{N+1})$ and integrate with respect to ξ_{N+1}. The result is

$$\begin{aligned}
&C^{LM_L}_{M'{}_{L_1}m'{}_{l'}} C^{SM_S}_{M'{}_{S_1}m'{}_{s'}}[\varepsilon_{L_1S_1}+\varepsilon_q-\mathscr{E}+K_r \\
&\quad+\langle L_1S_1M'_{L_1}M'_{S_1}|V^*_r|L_1S_1M_{L_1}M_{S_1}\rangle]\phi_q(\mathbf{r}) \\
&\quad+\sum_n\sum_m C^{LM_L}_{M_{L_1}m_{l'}} C^{SM_S}_{M_{S_1}m_{s'}}\langle\Psi_q|V|\Psi_n\rangle\phi_n(\mathbf{r})=0, \qquad (7)
\end{aligned}$$

where ε_q is the eigenvalue for state $\Psi_{ql''}$ satisfying the equation

$$\begin{aligned}
&\Big[C^{LM_L}_{M'{}_{L_1}m'{}_{l'}} C^{SM_S}_{M'{}_{S_1}m'{}_{s'}} K_{N+1}+\sum_m C^{LM_L}_{M_{L_1}m_{l'}} C^{SM_S}_{M_{S_1}m_{s'}} \\
&\quad\times\langle L_1S_1M'_{L_1}M'_{S_1}|V^*_{N+1}|L_1S_1M_{L_1}M_{S_1}\rangle-\varepsilon_q\Big] \\
&\quad\times\Psi_{ql''}(\xi_{N+1})=-\sum_n\sum_m\sum_{i\neq N+1}(-1)^{N+1-i} \\
&\quad\times C^{LM_L}_{M_{L_1}m_{l'}} C^{SM_S}_{M_{S_1}m_{s'}}\int d\xi_1\cdots d\xi_N\,\Psi^*{}_{L_1S_1}(\xi_1,\ldots,\xi_N) \\
&\quad\times\left\{\sum_{p=1}^{N}\frac{e^2}{|\xi_{N+1}-\xi_p|}\right\} \\
&\quad\times\Psi_{L_1S_1}(\xi_1,\ldots,\xi_{N+1},\ldots,\xi_N)\Psi_{nl'}(\xi_i). \qquad (8)
\end{aligned}$$

Let us denote the energy of the incident particle by $\varepsilon_r=\varepsilon_{L_1S_1}+\varepsilon_q-E$. As a result from (7) we obtain the following equation:

$$\begin{aligned}
&[K_r-\varepsilon_r+\langle L_1S_1M_{L_1}M_{S_1}|V^*_r|L_1S_1M_{L_1}M_{S_1}\rangle]\phi_q(\mathbf{r}) \\
&\quad=-\sum_n\langle\Psi_{ql''}|V|\Psi_{nl'}\rangle\phi_n(\mathbf{r}). \qquad (9)
\end{aligned}$$

The Clebsch–Gordan coefficients disappear when the initial state of the atomic electron is specified.

Equation (9) differs from the ordinary equations of multichannel scattering theory in that it contains the matrix element of the operator V^*_r sandwiched between ion-core wave functions.

If we allow for exchange between the incident electron and the core, the homogeneous part of Eq. (9) contains an exchange term:

$$\begin{aligned}
&[K_r-\varepsilon_r+\langle L_1S_1M_{L_1}M_{S_1}|V^*_r|L_1S_1M_{L_1}M_{S_1}\rangle]\phi_q(\mathbf{r}) \\
&\quad+\sum_{i=1}^{N}(-1)^{N+1-i}\int d\xi_1\cdots d\xi_N\,\Psi^*{}_{L_1S_1} \\
&\quad\times(\xi_1,\ldots,\ \xi_N)\left\{\sum_{p=1}^{N}\frac{e^2}{|\xi_p-\mathbf{r}|}\right\} \\
&\quad\times\Psi_{L_1S_1}(\xi_1,\ldots,\mathbf{r},\ldots,\xi_N)\phi_q(\xi_i)=0. \qquad (10)
\end{aligned}$$

Thus, the movements of the incident and atomic electrons in the ion-core field are described by a single Hamiltonian and, hence their wave functions form a orthonormalized system.

The solution to Eq. (9) in integral form can be written in the usual way:

$$\begin{aligned}
\phi_q(\mathbf{r})&=\phi_q^{(0)}(\mathbf{r})+\sum_n\int d\mathbf{r}'\,G_q^{(0)}(\mathbf{r},\mathbf{r}') \\
&\quad\times\langle\Psi_{ql''}|V|\Psi_{nl'}\rangle\phi_n(\mathbf{r}'), \qquad (11)
\end{aligned}$$

where $\phi_q^{(0)}(\mathbf{r})$ and the Green's function $G_q^{(0)}(\mathbf{r},\mathbf{r}')$ are the solutions of appropriate homogeneous equations with the potential $\langle L_1S_1M_{L_1}M_{S_1}|V_r^*|L_1S_1M_{L_1}M_{S_1}\rangle$. The asymptotic behavior of $G_q^{(0)}(\mathbf{r},\mathbf{r}')$ for $r\gg r'$ is given by the function

$$-\frac{2\pi\mu}{h^2}\phi_q^{(-)*}\frac{\exp ik_q r}{r}$$

(see Ref. 2), with $\phi_q^{(-)*}=\phi_{-q}^{(+)}$.

The inelastic scattering amplitude can be expressed in the following way:

$$A_{nq}^{kk'}=-\frac{2\pi\mu}{h^2}\int\phi_{k'}^{(-)*}(\mathbf{r}')\langle\Psi_{ql''}|V|\Psi_{nl'}\rangle\phi_k(\mathbf{r}')d\mathbf{r}'. \tag{12}$$

Here the upper and lower indices designate the initial and final states of the incident and atomic electrons, respectively. This formula differs from the one in Ref. 6 in that here the $\phi_n(\mathbf{r}')$ are not Coulomb functions.

Exchange between the impinging and valence electrons can be allowed for in the same way as is done in Ref. 6. In this case instead of $\Psi_{ql''}(\xi_{N+1})$ and $\phi_n(\mathbf{r})$ we must take

$$\Psi(\xi,\mathbf{r})=\frac{1}{\sqrt{2}}[\Psi_{ql''}(\xi_{N+1})\phi_n(\mathbf{r})\pm\Psi_{ql''}(\mathbf{r})\phi_n(\xi_{N+1})]. \tag{13}$$

Then Eq. (9) transforms into

$$\begin{aligned}&[K_r-\varepsilon_r+\langle L_1S_1M_{L_1}M_{S_1}|V_r^*|L_1S_1M_{L_1}M_{S_1}\rangle]\phi_q(\mathbf{r})\\&=-\sum_n[\langle\Psi_{ql''}|V|\Psi_{nl'}\rangle\phi_n(\mathbf{r})\pm\Psi_{nl'}(\mathbf{r})\\&\quad\times\langle\Psi_{ql''}|V|\phi_n\rangle].\end{aligned} \tag{14}$$

Correspondingly, instead of (11), the solution to Eq. (14) is given by the function

$$\begin{aligned}\phi_q(\mathbf{r})=\phi_q^{(0)}(\mathbf{r})+\sum_n\Big[&\int d\mathbf{r}'\,G_q^{(0)}(\mathbf{r},\mathbf{r}')\\&\times\langle\Psi_{ql''}|V|\Psi_{nl'}\rangle\phi_n(\mathbf{r}')\\&\pm\int d\mathbf{r}'\,G_q^{(0)}(\mathbf{r},\mathbf{r}')\Psi_{nl'}(\mathbf{r}')\langle\Psi_{ql''}|V|\phi_n\rangle\Big].\end{aligned} \tag{15}$$

Then the exchange part of the ionization amplitude can be written as

$$\begin{aligned}A_{nq\,\mathrm{exch}}^{kk'}=-(\pm1)\frac{2\pi\mu}{h^2}\int&\phi_{k'}^{(-)*}(\mathbf{r}')\\&\times\langle\Psi_{ql''}|V|\phi_n\rangle\Psi_{nl'}(\mathbf{r}')d\mathbf{r}'.\end{aligned} \tag{16}$$

We can allow for the effect of the field on the ionization amplitude in the same way as is done in the case of excitation.[1] In first-order perturbation theory the Green's function in the field is

$$G_q(\mathbf{r},\mathbf{r}')=G_q^{(0)}(\mathbf{r},\mathbf{r}')\left[1+\int d\mathbf{r}''\,V_f(\mathbf{r}'')G_q^{(0)}(\mathbf{r}'',\mathbf{r}')\right], \tag{17}$$

where

$$V_f(\mathbf{r}'')=-\frac{\alpha e\mathbf{E}\mathbf{r}}{r^3},$$

with α the dipole polarizability of the ion, and $\mathbf{E}$ the electric field strength.

As shown in Ref. 1, the field contributes considerably to the final scattering channel, where in the case of ionization there is the ion core and two electrons in a continuous-spectrum state.

Owing to the long-range nature of V_f, the field provides its greatest contribution to the distortion of the scattered-electron wave at distances r'' greater than the range of the scattering potential $V(r'')$. Then for $G_q^{(0)}\times(\mathbf{r}'',\mathbf{r}')$ we can employ an asymptotic expression in the limit $r''\gg r'$ similar to that for $G_q^{(0)}(\mathbf{r},\mathbf{r}')$ in the limit $r\gg r'$. As a result the Green's function in the field has the form

$$G_{k'}(\mathbf{r},\mathbf{r}')=-\frac{2\pi\mu}{h^2}\phi_{k'}^{(-)*}\frac{\exp(ik'r)}{r}[1+\eta_{k'}(\mathbf{E})], \tag{18}$$

where

$$\eta_{k'}(\mathbf{E})=-\frac{2\pi\mu}{h^2}\int\phi_q^{(-)*}(\mathbf{r})\langle ql''|V_f|ql'\rangle\frac{\exp(ik'r)}{r}d\mathbf{r}. \tag{19}$$

In the formula for the distortion factor $\eta_{k'}(\mathbf{E})$ the difference from the case of excitation[1] is that the matrix element of the scattering matrix is taken over the Stark functions of the continuous spectrum and instead of a plane wave the integrand contains $\phi_{k'}^{(-)*}$.

For the amplitude of ionization in the field we arrive at the following formula:

$$\begin{aligned}A_{nq,\mathrm{tot}}^{kk'}=-\frac{2\pi\mu}{h^2}[1+\eta_{k'}(\mathbf{E})]\Big[&\int\phi_{k'}^{(-)*}(\mathbf{r}')\\&\times\langle\Psi_{ql''}|V|\Psi_{nl'}\rangle\phi_k(\mathbf{r}')d\mathbf{r}'\\&\pm\int\phi_{k'}^{(-)*}(\mathbf{r}')\langle\Psi_{ql''}|V|\phi_k\rangle\Psi_{nl'}(\mathbf{r}')d\mathbf{r}'\Big],\end{aligned} \tag{20}$$

where the integrals are evaluated in the basis of Stark functions.

To estimate the distortion factor $\eta_k(\mathbf{E})$ we proceed as follows. Since integration in (19) is done over the outer region, the function ϕ can be chosen as the solution of the equation with a Coulomb potential:[10]

$$\begin{aligned}\phi_k(\mathbf{r})=e^{-n\pi/2}\sum_l&\frac{\Gamma(l+1+in)}{(2l)!}(2ikr)^le^{ikr}\\&\times F(l+1+in,\,2l+2,-2ikr)P_l(\cos\vartheta_{\mathbf{kr}}),\end{aligned} \tag{21}$$

where $n=-\mu Ze^2/h^2k$. Then the formula (19) for the distortion factor yields

$$\begin{aligned}\eta_k(\mathbf{E})=-\frac{2\pi\mu}{h^2}e^{-n\pi/2}\frac{\Gamma(2+in)}{2!}\int&e^{2ikr}(\alpha eE)\\&\times F(2+in,\,4,\,-2ikr)P_1(\cos\vartheta_{\mathbf{kr}})d(2ikr).\end{aligned} \tag{22}$$

Using the properties of hypergeometric functions,[11] we can evaluate the integral in (22) analytically. Then

$$\eta_k(\mathbf{E}) = -\frac{\mu}{\hbar^2}\, e^{n\pi/2}\, \frac{|\Gamma(2-in)|}{1+in}\, (\alpha e E) \times F(1-in,3,2ikr_0)\, P_1(\cos\vartheta_{\mathbf{kr}}). \qquad (23)$$

Here we have allowed for the fact that $n<0$. Next we represent the hypergeometric function in the form of a series and use the properties of gamma functions.[11] As a result we arrive at the following expression for the distortion factor::

$$\eta_k(\mathbf{E}) = 6\frac{\mu}{\hbar^2}\, e^{n\pi/2} (\alpha e E) P_1(\cos\vartheta_{\mathbf{kr}}) \sqrt{n^2+1}\, \sqrt{in} \times \left(\frac{\pi}{\sin(in\pi n)}\right)^{1/2} \left[\frac{z_0^0}{0!\,\Gamma(3)} + \frac{(1+in)z_0^1}{1!\,\Gamma(4)} + \frac{(2+in)(1+in)z_0^2}{3!\,\Gamma(5)} + \cdots\right], \qquad (24)$$

where $z_0 = 2ikr_0$.

Let us consider the extreme cases of slow and fast scattered electrons and estimate the distortion factors in both.

(a) $n\to 0$ $(k\to\infty)$. In this case formula (24) implies

$$\eta_k(\mathbf{E}) = 6\frac{\mu}{\hbar^2}\, (\alpha e E) P_1(\cos\vartheta_{\mathbf{kr}})\, \frac{1}{\Gamma(3)}\,. \qquad (25)$$

Since the polarizability of the ion core is insignificant, $\eta_k(\mathbf{E}) \sim 10^{-8}E$, and at $E\sim 10$–$100\ \mathrm{kV\,cm^{-1}}$ we have $\eta_k \ll 1$.

(b) $n\to\infty$ $(k\to 0)$. In (18) we introduce the notation $in \equiv x$. Then the definition of n implies $z_0 = -2r_0k_0/x$, with $k_0 = 4\pi^2\mu e^2/h^2$. The series within the square brackets in (24) can be written in the form of a sum,

$$\sum_m \frac{(1+m/x)\beta^m}{m!\,\Gamma(3+m)},$$

where $\beta = 2r_0k_0$. In the limit of $x\to\infty$ the sum is equal to $\beta^{-1}J_2(\sqrt{8k_0r_0})$, where $J_2(\sqrt{8k_0r_0})$ is a Bessel function.[12]

As a result we arrive at the following formula for the distortion factor:

$$\eta_k(\mathbf{E}) = 6\frac{\mu}{\hbar^2}\, (\alpha e E) P_1(\cos\vartheta_{\mathbf{kr}}) \times (2\pi)^{1/2} n^{3/2} \beta^{-1} J_2(\sqrt{8k_0r_0}). \qquad (26)$$

When the scattered-electron momentum is low, $ka_0 \leqslant 0.01$, the factor $n^{3/2}$ can balance the insignificant value of the ion-core polarizability α. In this case the distortion factor $\eta(\mathbf{E})$ has a strong effect on the ionization amplitude.

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