

RESONANCE CHARGE EXCHANGE IN SLOW COLLISIONS

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The resonance charge exchange cross section is determined by the magnitude of the electron energy level splitting $\epsilon_u - \epsilon_g = f(R)$ (R is the distance between the nuclei), which is exponentially small at large values of R and hence can be determined only with an accuracy to a pre-exponential factor by perturbation-theory methods. The method employed in the present paper can be used to calculate the pre-exponential factor for $\epsilon_u - \epsilon_g$ in the case of an electron in the field of two Coulomb centers of charge Z . It is shown that for an asymptotic form $\epsilon_u - \epsilon_g = AR^n \exp(-\sqrt{-2wR})$, which is valid for a system consisting of an arbitrary atom and its ion (w is the electron energy), the relation between the collision rate and resonance charge exchange cross section can be satisfactorily described by the Firsov formula. The relation between the collision rate and resonance charge exchange cross section for a charge Z and an hydrogen-like atom in the ground and excited states is established.

1. Resonance charge exchange when an ion collides with an atom of its own kind constitutes a transition of an electron from one ion to another in the same state. The probability of resonance charge exchange is given in the adiabatic approximation by the expression [1]

$$W = \sin^2 x, \quad x = \frac{1}{2} \int_{-\infty}^{+\infty} (\epsilon_u - \epsilon_g) dt = \int_{\rho}^{\infty} (\epsilon_u - \epsilon_g) \frac{RdR}{v\sqrt{R^2 - \rho^2}}. \quad (1)$$

Here ρ —impact parameter of the collision, R —distance between nuclei, ϵ_u —electron energy level corresponding to an odd wave function, ϵ_g —electron energy level corresponding to an even wave function. We use a system of atomic units in which $\hbar = m_e l = e^2 = 1$.

At small collision velocities v , the cross section for resonance charge exchange is large, and the calculation of the cross section reduces to a determination of the asymptotic form of $\epsilon_u - \epsilon_g$ as $R \rightarrow \infty$. This quantity is exponentially small, so that by ordinary methods of perturbation theory can be determined only accurate to within a pre-exponential factor. The situation is similar to that prevailing in the problem of over-the-barrier reflection in the quasiclassical approximation [2] or in adiabatic perturbation theory [3], where the second term of perturbation theory is of the same order as the first.

In the present paper we use a method which yields the correct pre-exponential factor in the

asymptotic expansion of the quantity $\epsilon_u - \epsilon_g$ for an electron situated in the field of two Coulomb centers of charge Z . The asymptotic form of $\epsilon_u - \epsilon_g$ is obtained for large R in the case of a system consisting of an arbitrary atom and an ion of its own kind.

2. Let us calculate the energy $\epsilon_u - \epsilon_g$ of the splitting of an ion of a hydrogen-like molecule at large distances between nuclei. The variables in the Schrödinger equation for an electron situated in the field of two Coulomb centers can be separated in elliptic coordinates [4,5]

$$\xi = \frac{r_1 + r_2}{R}, \quad \eta = \frac{r_1 - r_2}{R}, \quad \varphi = \tan^{-1} \frac{y}{x},$$

where $r_{1,2}$ —distance from the electron to the corresponding center. Putting $\psi = X(\xi) Y(\eta) e^{im\varphi}$, we obtain equations for the functions $X(\xi)$ and $Y(\eta)$:

$$\frac{d}{d\xi} \left[(\xi^2 - 1) \frac{dX}{d\xi} \right] + \left[-\frac{m^2}{\xi^2 - 1} + 2RZ\xi - p^2\xi^2 + A \right] X = 0, \quad (2)$$

$$\frac{d}{d\eta} \left[(1 - \eta^2) \frac{dY}{d\eta} \right] + \left[-\frac{m^2}{1 - \eta^2} + p^2\eta^2 - A \right] Y = 0. \quad (3)$$

Here $p^2 = -(\frac{1}{2}) R^2 (\epsilon - Z^2/R) = -R^2 w/2$, A —separation constant that depends on the energy of the system ϵ as a parameter, and w —electron energy. The condition that the eigenvalues of (2)

and (3) coincide, $A_\xi = A_\eta$, determines the energy of the system in the given state. The energy levels in the even (ϵ_g) and odd (ϵ_u) states and the separation constants A_u and A_g coincide, apart from exponentially small terms¹⁾, when the distances between nuclei are large, so that we can assume with the same degree of accuracy that

$$\begin{aligned} \psi_{g,u} &= \frac{1}{2\sqrt{\pi}} e^{im\varphi} X(\xi) [Y(\eta) \pm Y(-\eta)] \\ &= \frac{1}{\sqrt{2}} [\psi(1) \pm \psi(2)], \end{aligned} \quad (4)$$

i.e., the functions $\psi(1)$ and $\psi(2)$ are the same for the even and odd states. The function $\psi(1)$, corresponding to the electron beam situated near the first nucleus, decreases exponentially with increasing distance from the electron to the first nucleus, while the function $\psi(2)$ increases with increasing distance from the electron to the second nucleus.

When $\xi - 1 \ll 1$ and $1 \pm \eta \ll 1$ Eqs. (2) and (3) go over into the equations of an electron situated near one Coulomb center, in parabolic coordinates^[5]. We shall assume that the electron is essentially concentrated near the Coulomb centers in the region $\xi - 1 \ll 1$, $1 \pm \eta \ll 1$. Then the state of the electron is described by the parabolic quantum numbers n_1 , n_2 , and m , while the wave function is of the form

$$\begin{aligned} \psi_{n_1 n_2 m}(1, 2) &= \frac{\sqrt{2} \gamma^{3/2} (n_1! n_2!)^{1/2} \Phi(\varphi)}{n^2 [(n_1+m)! (n_2+m)!]^{3/2}} \exp[-p(\xi \pm \eta)] \\ &\times [2p(\xi - 1)]^{m/2} [2p(1 \mp \eta)]^{m/2} L_{n_1+m}^m \\ &\times [2p(\xi - 1)] L_{n_2+m}^m [2p(1 \mp \eta)]. \end{aligned} \quad (5)$$

Here $n = n_1 + n_2 + m + 1$, $p = R\sqrt{-w/2} = \gamma R/2$, the plus sign corresponds to the electron beam situated near one nucleus ($\eta \approx -1$, $r_1 \ll R$), the minus sign near the other nucleus, $\Phi(\varphi) = (2\pi)^{-1/2} e^{\pm im\varphi}$, $m > 0$. The requirement that the electron be essentially concentrated near the Coulomb centers leads to the condition

$$p \gg n_{1,2} + m/2. \quad (6)$$

It will be shown later that to determine $\epsilon_u - \epsilon_g$ it is necessary to know the wave functions of the electron in the region between the nuclei $\xi - 1 \ll 1$, $1 \pm \eta \sim 1$. We determine the quasi-

classical solution of (3). We seek

$$Y(\eta) = e^S, \quad S = S_0 + S_1 + S_2 + \dots$$

We confine ourselves in the equation for S to the first terms of the expansion, S_0 and S_1 ($A_\eta = p^2 - 2\alpha p$, $\alpha = 2n_2 + m + 1$ ^[5]), we find

$$\begin{aligned} Y_1(\eta) &= \frac{C}{\sqrt{1-\eta^2}} \left(\frac{1-\eta}{1+\eta} \right)^{\alpha/2} e^{p\eta}, \\ Y_2(\eta) &= Y_1(-\eta) = \frac{C}{\sqrt{1-\eta^2}} \left(\frac{1+\eta}{1-\eta} \right)^{\alpha/2} e^{-p\eta}. \end{aligned} \quad (7)$$

The region of applicability of the quasiclassical solution $S_2 \ll 1$ is of the form

$$2p(1 \pm \eta) \gg \frac{1}{4} [(a-1)^2 - m^2] = n_2(n_2 + m). \quad (8)$$

In this interval of values of η and $\xi - 1 \ll 1$, the wave function of the electron is

$$\begin{aligned} \psi_{n_1 n_2 m}(1, 2) &= \frac{C}{\sqrt{1-\eta^2}} \exp[-p(\xi - 1) \pm p\eta] \\ &\times [2p(\xi - 1)]^{m/2} \\ &\times L_{n_1+m}^m [2p(\xi - 1)] \left(\frac{1 \mp \eta}{1 \pm \eta} \right)^{\alpha/2} \Phi(\varphi), \end{aligned} \quad (9)$$

with $\psi(1)$ and $\psi(2)$ exponentially small in this region. When $2p(1 \pm \eta) \gg n_2(n_2 + m)$ we have

$$L_{n_1+m}^m [2p(1 \mp \eta)] \rightarrow \frac{(-1)^{n_2+m} (n_2 + m)!}{n_2!} [2p(1 \mp \eta)]^{n_2},$$

and if

$$2p \gg n_2(n_2 + m), \quad (10)$$

then there is a region of values of η away from the nodes of the wave function $1 \gg 1 \pm \eta \gg n_2(n_2 + m)/2p$, in which the Coulomb solution goes over continuously into the quasiclassical solution. Comparing the solutions, we determine the normalization coefficient for the quasiclassical function:

$$C = \frac{2\sqrt{2}\gamma^{3/2}}{\sqrt{n}} \frac{(n_1!)^{1/2}}{[(n_1+m)!]^{3/2}} \frac{(-1)^{n_2+m} (4p)^{n_2+m/2} e^{-p}}{[n_2!(n_2+m)!]^{1/2}}. \quad (11)$$

3. To find $\epsilon_u - \epsilon_g$ with the aid of the wave functions (8), we make use of a procedure proposed by Gor'kov and Pitaevskii^[7]. We have

$$\begin{aligned} (\epsilon_u - \epsilon_g) \int_V \psi_u \psi_g d\tau &= \frac{1}{2} \int_V (\psi_u \Delta \psi_g - \psi_g \Delta \psi_u) d\tau \\ &= \frac{1}{2} \oint_s [\psi(1) \nabla \psi(2) - \psi(2) \nabla \psi(1)] ds, \end{aligned}$$

where s —surface $1 - \eta = \text{const} \gg n_2(n_2 + m)/2p \ll 1 \pm \eta$, bounding the volume V . Inside V , the function $\psi(2)$ is exponentially small, and outside the function $\psi(1)$ is exponentially small, so that

¹⁾As can be seen from the work of Gershtein and Krivchenkov^[5], as $R \rightarrow \infty$ the quantities A and ϵ are determined by the parabolic quantum numbers n_1 , n_2 , and m of the electron in the Coulomb field, which are the same for the even and odd states.

$$\int_V \psi_u \psi_g d\tau = \frac{1}{2} \int_V [\psi^2(1) - \psi^2(2)] d\tau = \frac{1}{2},$$

$$\varepsilon_u - \varepsilon_g = \oint_s [\psi(1) \nabla \psi(2) - \psi(2) \nabla \psi(1)] d\mathcal{E}$$

$$= -R(1 - \eta^2) C^2 \left[Y(\eta) \frac{\partial Y(-\eta)}{\partial \eta} - Y(-\eta) \frac{\partial Y(\eta)}{\partial \eta} \right]$$

$$\times \int_1^\infty X^2(\xi) d\xi. \quad (12)$$

(The last formula was derived earlier by Firsov^[1] in a more complicated manner.)

Using on the surface s the quasiclassical functions $Y(\eta) = \exp(S_0 + S_1 + \dots)$, we obtain accurate to terms $\sim 1/p^2$:

$$-(1 - \eta^2) \left[Y(\eta) \frac{\partial Y(-\eta)}{\partial \eta} - Y(-\eta) \frac{\partial Y(\eta)}{\partial \eta} \right] = -(1 - \eta^2)$$

$$\times \{ [1 + S_2(-\eta) + S_2(\eta)] [S'_0(\eta) + S'_0(-\eta)]$$

$$+ [S'_1(\eta) + S'_1(-\eta)] \} = -2p,$$

and this quantity is independent of the choice of the surface s . Hence

$$\varepsilon_u - \varepsilon_g = \frac{4\gamma^3}{n} \frac{R(4p)^{2n_2+m}}{n_2!(n_2+m)!} e^{-2p} \left[1 + O\left(\frac{1}{p^2}\right) \right]. \quad (13)$$

In calculating the correction $\sim 1/p^2$ to $\varepsilon_u - \varepsilon_g$, account must be taken of the distortion of the wave function (5) of the electron near one of the nuclei, due to the action of the field of the other nucleus.

The energy of the electron situated in the field of two Coulomb centers is^[5]

$$-W = \frac{Z^2}{2n^2} + \frac{Z}{R} + \frac{3n(n_2 - n_1)}{R^2},$$

so that

$$p \approx \frac{ZR}{2n} \left[1 + \frac{n^2}{ZR} - \frac{n^4}{Z^2R^2} + \frac{3n^3}{2Z^2R^2} (n_2 - n_1) \right]. \quad (14)$$

From (13) and (14) we have for an electron in the ground state of the ion of the hydrogen molecule:

$$\varepsilon_u - \varepsilon_g = 4\gamma^3 R e^{-\gamma R} = \frac{4}{e} R e^{-R}, \quad \gamma = \sqrt{1 + \frac{2}{R}} \approx 1 + \frac{1}{R}.$$

This coincides with the result of Hering^[8], obtained by a method close to that employed here. The variation method yields^[9] $(\frac{4}{3}) \gamma^3 R \exp(-\gamma R)^2$, while formula (12) using atomic wave

functions yields $\gamma^3 R \exp(-\gamma R)$. The calculation of $\varepsilon_u - \varepsilon_g$ by the variational method is inconsistent, since ε_u and ε_g are obtained with allowance for exponentially small terms, whereas the power series terms are neglected.

4. The method employed can be used to calculate the study energy $\varepsilon_u - \varepsilon_g$ for a system consisting of an arbitrary atom and its ion, with large distances between the nuclei. Away from the nuclei the electron is acted upon by the Coulomb field of the ions so that its state is described by Eqs. (2) and (3). Consequently in the region between the nuclei $2p(1 \pm \eta) \gg \frac{1}{4}[(\alpha - 1)^2 - m^2]$ the wave function of the electron is of the form (7)³⁾. If $1 + \eta \ll 1$, corresponding to the location of the electron near one of the ions, the wave function depends only on r_1 and Z (atomic wave function), so that its dependence on η is only through the combination $R(1 + \eta)$. Therefore the connection between the normalization coefficient of the quasiclassical function C and R is given by the relation $C \sim R^{(\alpha-1)/2} e^{-\gamma R/2}$. Expressing $\varepsilon_u - \varepsilon_g$ in terms of the quasiclassical wave function of the electron by means of formula (12), we get

$$\varepsilon_u - \varepsilon_g = BR^\alpha e^{-\gamma R} [1 + O(1/R^2)]. \quad (15)$$

A criterion for the applicability of (15), connected with the possibility of expanding $\varepsilon_u - \varepsilon_g$ in powers of $1/R$, is

$$\gamma R \gg \alpha. \quad (16)$$

For low excited states, satisfaction of this criterion automatically satisfies the condition of applicability of the quasiclassical approximation (8) in the region between the nuclei.

The constants B and α cannot be obtained theoretically in the general case, since the exact function of the electron in the atom is not known, and consequently the separation constant A in (2) and (3) is unknown.

5. The cross section for resonance charge exchange is

$$\sigma = \int_0^\infty \sin^2 x \cdot 2\pi\rho d\rho. \quad (17)$$

In evaluating this integral for some forms of $[x \sim (\rho^2 + C)^{-1}, (\rho^2 + C)^{-2}, \exp(-\beta\rho^2)]$, Firsov

²⁾Usually in the calculation of $\varepsilon_u - \varepsilon_g$ by the variational method, no account is taken of the change in the electron energy Z/R due to introduction of the ion, so that the result is $(4/3)Re^{-R}$. The error incurred thereby almost cancels the inaccuracy of the variational method. As shown by calculations, the use of the atomic wave functions as trial functions for the variational methods, with allowance for their distortion as a result of the electric field of the ion and allowance for the change in the electron energy, leads to the correct result $4e^{-1}Re^{-R}$.

³⁾From the comparison of the solution of the Schrödinger equation for electrons in a field that decreases at large distances from the nuclei, and from the solution of (2) and (3) away from the nuclei, we find that expansion of A in powers of p begins with p^2 . From this we obtain in the general case $A = p^2 - 2ap$.

proposed to seek the cross section in the form

$$\sigma = \pi R_0^2/2, \quad x(R_0) = 1/\pi. \quad (18)$$

Bates and Boyd [10] checked the correctness of this method of determining the cross section for a dependence $x \sim 1/\rho^n$. It is shown in the appendix that if the cross section for resonance charge exchange is large ($R_0 \sim \sqrt{\sigma} \gg 1$), then it is determined completely by the asymptotic form of $x(\rho)$. If the asymptotic form of $\epsilon_u - \epsilon_g$ is described by (15), then the cross section, apart from terms proportional to a constant, is [see (A.2)]

$$\sigma = \frac{\pi}{2} R_0^2 - \frac{\pi^3}{24\gamma^2}, \quad x(R_0) = \frac{11}{40}. \quad (19)$$

In calculating the cross section with accuracy to terms $\sim R_0$, the result obtained coincides with that determined by the Firsov method (18), with slight modification of the constant.

Allowance for the constant term $-\pi^3/24\gamma^2$ in the cross section (19) is correct if two terms are used in the asymptotic expansion of $\epsilon_u - \epsilon_g$. The second term of the asymptotic expansion arises when account is taken of the Coulomb term in the electron energy (14). For the nondegenerate state of the electron we have $w = -\gamma_0^2/2 - Z/R + O(1/R^4)$, where $-\gamma_0^2/2$ is the energy of the electron in the atom and Z is the charge of the ion. Expanding γ in powers of $1/R$ we obtain

$$\sigma = \frac{\pi R_0^2}{2} + \frac{\pi}{2\gamma_0^2} \left[\frac{2ZB'(\gamma_0)}{B(\gamma_0)} + \frac{Z^2}{\gamma_0^2} - \frac{Z}{\gamma_0} - \frac{\pi^2}{12} \right], \quad (20a)$$

where R_0 is determined by the relation

$$(\epsilon_u - \epsilon_g) R_0^{1/2} = 0.22v\gamma_0^{1/2}e^{Z/\gamma_0}. \quad (20b)$$

6. On the basis of (19)–(20) we establish, using (13) and (14), a connection between the cross section for the resonant charge exchange in the relative velocity of collision of the Coulomb charge Z with a hydrogen like atom situated in a state n_1, n_2 , and m :

$$\sigma = \frac{\pi R_0^2}{2} + \frac{\pi n^2}{2Z^2} \left[n(4n_1 + 2n_2 + 3m + 6) - \frac{\pi^2}{12} \right], \quad (21a)$$

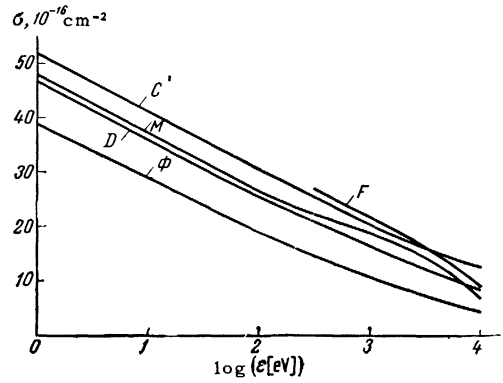
with R_0 determined from the equation

$$\frac{(2ZR_0/n)^{2n_2+m+3/2} e^{-ZR_0/n}}{n_2!(n_2+m)!} = 0.155 \frac{n^2 v}{Z} e^n, \quad (21b)$$

the criterion for the applicability of these relations being

$$ZR_0/n \gg \max \{n; n_2(n_2+m)\}. \quad (21c)$$

A comparison of (21) with experiment [11] and with the results of the calculations of the cross section for the resonance charge exchange of the



F—data of [11], D—data of [12], M—data of [13], C—results of the present work; Φ —corresponds to $\epsilon_u - \epsilon_g$ calculated by formula (12) using atomic wave functions.

proton on a hydrogen atom in the ground state [12,13] is shown in the figure.

In conclusion, the author expresses his gratitude to A. M. Dykhne and O. B. Firsov for valuable advice and interest in the work.

APPENDIX

Let us calculate the integral (17)⁴. Since $\epsilon_u - \epsilon_g > 0$ for any R , we get that

$$x = \int_{\rho}^{\infty} \frac{(\epsilon_u - \epsilon_g) R dR}{v \sqrt{R^2 - \rho^2}}$$

is a monotonic function of ρ . We put $x_{\max} = x(0) \sim 1/v \gg 1$. The cross section for resonance charge exchange is

$$\begin{aligned} \sigma &= \pi \int_{x_{\max}}^0 \sin^2 x(\rho^2)' dx = \pi \int_{x_{\max}}^{\pi k} \sin^2 x(\rho^2) dx \\ &+ \int_0^{\pi k} \rho^2 \sin 2x dx = I_1 + I_2. \end{aligned}$$

Putting $x_{\max} = \pi n + \Delta$, $0 < \Delta < \pi$, and neglecting the integral from $\pi n + \Delta$ to πn , which is of the order of v^2 , we get

$$\begin{aligned} I_1 &= \pi \int_{\pi n}^{\pi k} (\rho^2)' \sin^2 x dx \\ &= -\pi \int_0^{\pi} \sin^2 x dx \sum_{m=k}^{n-1} (\rho^2)'(x + \pi m). \end{aligned}$$

Let us expand $(\rho^2)'(x + y)$ in a Taylor series in the region $\pi(m - 1/2) < y < \pi(m + 1/2)$ and take the integral over this region. We get

⁴The idea of these calculations is due to A. M. Dykhne.

$$\sum_{m=k}^{n-1} (\rho^2)'(x + \pi k) = \frac{1}{\pi} \int_{\pi(k-1/2)}^{\pi(n-1/2)} dy (\rho^2)'(x + y) - \frac{1}{\pi} \frac{(\pi/2)^3}{3!} \int_{\pi(k-1/2)}^{\pi(n-1/2)} (\rho^2)''' dy + \dots,$$

so that when $v \ll 1$

$$I_1 = \int_0^{\pi} \sin^2 x dx \rho^2 \left(x + \pi k - \frac{\pi}{2} \right) - \frac{\pi^3}{24} \rho(\pi k) \rho''(\pi k) + \dots \quad (\text{A.1})$$

If $\pi k \gg 1$, only the first term of (A.1) is significant. It follows from (A.1) that if $\rho(\pi k) \gg 1$, the cross section is determined by the asymptotic form of $\varepsilon_u - \varepsilon_g$.

Let us calculate the cross section for the resonant charge exchange in the case when the asymptotic form of $\varepsilon_u - \varepsilon_g$ is given by (11), so that $x(\rho) \rightarrow D\rho^{\alpha+1/2} e^{-\gamma\rho}$. We introduce ρ_1 in such a way that $x(\rho_1) = 1$ and $\rho_1 \sim \ln(1/v) \gg 1$. For $\rho \gg 1$ we have

$$\gamma(\rho - \rho_1) = -\ln x + (\alpha + 1/2) \ln(\rho/\rho_1),$$

and $(\rho - \rho_1) \ll \rho_1$

$$\rho = \rho_1 - \frac{\ln x}{\gamma} - \frac{(\alpha + 1/2) \ln^2 x}{\gamma^2 \rho^2},$$

where $\gamma' = \gamma - (\alpha + 1/2)/\rho_1$. This relation is satisfied when $|\ln x| \ll \gamma\rho_1$, and since $\gamma\rho_1 \gg 1$ we can, by choosing $\ln \pi k \ll \gamma\rho_1$, ensure the correctness of this relation in the main region of integration of I_1 and I_2 . Using as the small expansion parameter $1/\gamma\rho_1 \ll 1$, we have accurate

to constant terms

$$\sigma = \frac{\pi}{2} \rho^2 \Big|_{x=\pi/4} - \frac{\pi^3}{24\gamma^2} + O\left(\frac{1}{\gamma\rho_1}, \frac{\rho_1}{\pi^2 k^2}\right). \quad (\text{A.2})$$

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