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X-641-70-206
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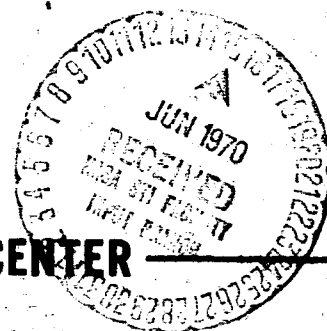
VIBRO-ROTATIONAL EXCITATIONS
OF H_2^+ BY e^+ IMPACT
A SEMI-CLASSICAL APPROACH

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JUNE 1970



— GODDARD SPACE FLIGHT CENTER —
GREENBELT, MARYLAND



N70-23688

FACILITY FORM 602

(ACCESSION NUMBER)

24

(PAGES)

Tmx-63931
(NASA CR OR TMX OR AD NUMBER)

(THRU)

(CODE)

24

(CATEGORY)

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F. H. M. Faisal

Abstract

Analogies between Coulomb excitations of nuclei and ionic molecules by charged projectiles is utilized to calculate vibro-rotational excitations of H_2^+ molecular ions by e^+ impact by a semi-classical method developed in the nuclear case.

1. Introduction

Recently there has been a considerable upsurge of interest in the study of rotational and vibrational energy loss processes in diatomic molecules by electron impact. They are of much importance not only for understanding the fundamental energy exchange processes involved, but also for their applications in such allied fields as Astrophysics and Atmospheric Physics.

In the present work we shall investigate the coupled excitation of vibro-rotational states of hydrogen molecular ions, H_2^+ , by collision with positrons, e^+ . The study of such excitations with e^+ is not only important for its intrinsic significance but also for the mathematical simplicity it introduces in the formulation of the complex excitation process itself. This is due to the fact that the Pauli exchange does not enter directly into the problem.

In this work we shall adopt a semi-classical view and make use of the analogy of Coulomb excitations of nuclei, which has been studied extensively in the past (Alder et al., 1965). The present method is semi-classical in that we shall treat the target system quantum mechanically while the motion of the projectile would be assumed to be along a classical Coulomb trajectory. In the end, however, we shall attempt to modify the classical nature of the projectile motion, which does not distinguish between the initial and final states, by demanding that the principle of ~~reciprocity~~ be satisfied by the cross-sectional expressions and invoking the Correspondence Principle to be applied to such quantities as the classical velocity of the projectile.

2. Theory

Let N , n and j denote, respectively, the electric, the vibrational and the rotational quantum numbers of the target molecule. In this paper we shall confine ourselves to the ground electronic state of the target and mostly disregard the quantum number N . One of the basic assumptions of our semiclassical approximation is that in the first approximation the incident positron moves essentially along a Rutherford trajectory around the effective positive charge of H_2^+ , situated at the cm. of the molecule. Thus, while the positron moves along its trajectory it is allowed to induce transitions in the molecular motion through the electric coupling with the various molecular charges. The transition probability, b_{if} , for the initial state i going to the final state f may then be given by the first order time dependent scattering theory of Dirac (1926). Thus we write

$$b_{if} = -i \int_{-\infty}^{\infty} e^{i\omega t} \langle f | V_{int}(t) | i \rangle dt, \quad (1)$$

where $\omega = E_i - E_f$ is the energy difference between the states i and f and $V_{int}(t)$ is the interaction potential between the incident positron and the H_2^+ target. It can easily be seen (Fig. 1) that

$$V_{int}(t) = \frac{1}{|\frac{\bar{R}}{2} - \bar{r}_2|} + \frac{1}{|\frac{\bar{R}}{2} + \bar{r}_2|} - \frac{1}{|\bar{r}_2 - \bar{r}_1|} - \frac{1}{r}, \quad (2)$$

where $\bar{r} = \bar{r}(t)$ is the trajectory of the positron and \bar{r}_1 is the position vector of the target electron, measured from the c.m. of the molecule. The vector \bar{R} stands for the separation between the two nuclei of the target.

We shall describe the target molecule in terms of simple product of normalized wave-functions $\phi_N(\bar{r}_1)$, $X_n(R)$ and $Y_j^m(\hat{R})$ corresponding to the electronic, the vibrational and the rotational motions. Thus the total target wave function becomes

$$|Nnj\rangle = \phi_N(\bar{r}_1) X_n(R) Y_j^m(\hat{R}) \quad (3)$$

In view of the fact that the incident positron repels itself from the positively charged target, we may for sufficiently low energies, simplify the potential interaction (2) by expanding it for essentially non-penetrating projectile orbits. Thus we find

$$V_{int}(t) = \left\{ - \sum_{\lambda=1,2,\dots}^{\kappa=\pm\lambda} \frac{4\pi}{2\lambda+1} \frac{r_1^\lambda}{r_2^{\lambda+1}} Y_\lambda^{\kappa}(\hat{r}_1) Y_\lambda^{\kappa}(\hat{r}_2) \right. \\ \left. + \frac{1}{2} \sum_{\lambda=2,4,\dots}^{\kappa=\pm\lambda} \frac{4\pi}{2\lambda+1} \frac{(R/2)^\lambda}{r_2^{\lambda+1}} Y_\lambda^{\kappa}(\hat{R}) Y_\lambda^{\kappa}(\hat{r}_2) \right\} ; \quad (4)$$

$\kappa > \kappa_1$

Taking the matrix elements between the initial state $|N_i n_i j_i\rangle$ and the final state $|N_f n_f j_f\rangle$ we find from (3) and (4),

$$\langle f | V_{int}(t) | i \rangle = \left\{ - \sum_{\substack{\lambda=1,2,\dots \\ \kappa=\pm\lambda}}^{\kappa=\pm\lambda} \frac{4\pi}{2\lambda+1} \langle \phi_{N_f} | r_1^\lambda Y_\lambda^{\kappa}(\hat{r}_1) | \phi_{N_i} \rangle \times \right. \\ \left. \dots \right\} \quad (5)$$

$$\begin{aligned}
 & \times \langle \chi_{n_s} | \chi_{n_i} \rangle \langle Y_{j_s}^{m_s}(\hat{R}) | Y_{j_i}^{m_i}(\hat{R}) \rangle \times \frac{Y_{\lambda}^{\lambda}(\hat{R})}{R^{\lambda+1}} \\
 & + 2 \sum_{\substack{\lambda=2,4,\dots \\ \lambda=-\lambda}}^{\lambda=+\lambda} \frac{4\pi}{2\lambda+1} \langle \phi_{N_f}(\bar{r}_i) | \phi_{N_i}(\bar{r}_i) \rangle \langle \chi_{n_s} | (R/2)^{\lambda} | \chi_{n_i} \rangle \times \\
 & \times \left\langle Y_{j_s}^{m_s}(\hat{R}) | Y_{\lambda}^{\lambda}(\hat{R}) | Y_{j_i}^{m_i}(\hat{R}) \right\rangle \times \frac{Y_{\lambda}^{\lambda}(\hat{R})}{R^{\lambda+1}} \quad \} \quad (5)
 \end{aligned}$$

We find that in the present approximation, the above equation(5) controls the entire process of transitions among the various states and the various modes of motion of the target molecule. It can easily be seen that the perturbation of the electronic motion, represented by the first sum in (5) is independent of the nuclear perturbation, represented by the second sum in (5). This is, of course, a consequence of the Born-Oppenheimer separation already assumed in the product wave function (3). We note however that the transitions among any two vibrational states $|n_1\rangle$ and $|n_2\rangle$ ($n_1 \neq n_2$) or any two rotational states $|j_1\rangle$ and $|j_2\rangle$ ($j_1 \neq j_2$) or both lead to a vanishing of the first sum in (5). Thus the vibrational-rotational transition probabilities in the ground electronic state is found to be given by

$$\begin{aligned}
 b_{if} &= -4\pi i \sum_{\substack{\lambda=2,4,\dots \\ \lambda=-\lambda}}^{\lambda=+\lambda} \frac{1}{2\lambda+1} 2 \langle n_f j_f | (R/2)^{\lambda} Y_{\lambda}^{\lambda}(\hat{R}) | n_i j_i \rangle \times J_{\lambda\mu}, \\
 J_{\lambda\mu} &= \int_{-\infty}^{\infty} e^{i\omega t} \{R(t)\}^{-\lambda-1} Y_{\lambda}^{\lambda}(\hat{R}(t)) dt, \quad (6) \\
 |n_j\rangle &= | \chi_n(R) Y_j^m(\hat{R}) \rangle
 \end{aligned}$$

We note that all informations regarding the trajectory of the incident positron is contained in the orbit integrals $J_{\lambda\mu}$. Evaluation of these

integrals is extensively discussed in the literature (e.g. Alder et al., 1965) and we shall note here that the Coulomb trajectory of e^+ can most conveniently be given by the parametric equations

$$\begin{aligned} r(t) &= r_0 (\Sigma \cosh \tau + 1) \\ x(t) &= r_0 (\cosh \tau + \Sigma) \\ y(t) &= r_0 (\Sigma^2 - 1)^{\frac{1}{2}} \sinh \tau \\ z(t) &= 0 \\ t &= \frac{r_0}{v_0} (\Sigma \sinh \tau + \tau) \end{aligned}$$

where Σ is the eccentricity, τ is the eccentric anomaly, r_0 is half the distance of closest approach, v_0 is the projectile velocity and t is the time parameter. Substituting (7) in (6) and choosing the quantization axis along the angular momentum of the molecular rotation, it can be shown (Alder et al., 1965)

$$\begin{aligned} J_{\lambda\mu} &= \kappa_0^{-\lambda-1} \omega_0^{-1} Y_{\lambda}^{\mu}(\frac{\pi}{2}, 0) I_{\lambda\mu}(\xi, \theta), \\ I_{\lambda\mu}(\xi, \theta) &= \int_{-\infty}^{\infty} e^{i\xi(\tau + (1+\Sigma^2)^{\frac{1}{2}} \sinh \tau)} \\ &\times \frac{(\Sigma + \omega \cosh \tau + i(\Sigma^2 - 1)^{\frac{1}{2}} \sinh \tau)^{\mu}}{(1 + \Sigma \omega \cosh \tau)^{\lambda + \mu}} d\tau, \end{aligned} \quad (8)$$

$$\begin{aligned} \xi &= \frac{E_i - E_f}{\omega_0}, \\ \omega_0 &= \frac{v_0}{k_0}, \\ \kappa_0 &= \frac{1}{2E_0}. \end{aligned} \quad (9)$$

We have defined E_0 to be the incident energy in a.u. and θ as the scattering angle. To calculate the cross-sections we need the number of incident particles in a plane wave of unit flux with impact parameters between b and $b + db$. This is given by

$$2\pi b db = \frac{\kappa_0^{-2}}{4(\sin \theta/2)^4} \times d\Omega, \quad \text{where } d\Omega \text{ is an elementary solid angle ;}$$

$$b = \kappa_0 \cot \theta/2.$$

The differential cross-section for the λ^{th} multipole transition averaged over the initial substates m_i and summed over the final substates m_f , may therefore be written as

$$\left(\frac{d\sigma}{d\Omega}\right)_\lambda = \frac{1}{2j_i + 1} \sum_{m_i, m_f} |b_{if}|^2$$

$$= \frac{1}{\kappa_0^2} \kappa_0^{-2\lambda+2} 4B_\lambda(n_i j_i \rightarrow n_f j_f) \frac{df_\lambda(\xi, \theta)}{d\Omega}, \quad (11)$$

$$\frac{df_\lambda(\xi, \theta)}{d\Omega} = \frac{4\pi^2}{(2\lambda+1)^3} \sum_{\mu=-\lambda}^{\mu=\lambda} \frac{\{Y_\lambda^\mu(\frac{\pi}{2}, 0) I_{\lambda\mu}(\xi, \theta)\}^2}{(\sin \theta/2)^4} \quad (12)$$

The total cross-section is obtained by integrating (11) over all scattering angles. Thus

$$(\sigma)_\lambda = \frac{1}{\kappa_0^2} \kappa_0^{-2\lambda+2} 4B_\lambda(n_i j_i \rightarrow n_f j_f) f_\lambda(\xi) \quad (13)$$

with

$$f_{\lambda}(\xi) = \frac{4\Gamma^2}{(2\lambda+1)^3} \sum_{\mu=-\lambda}^{\mu=+\lambda} \left\{ Y_{\lambda}^{\mu} \left(\frac{\pi}{2}, 0 \right) \right\}^2 \int_0^{\infty} I_{\lambda, \mu}(\xi, \theta) \frac{\cos \theta/2}{(\sin \theta/2)^3} d\theta \quad (14)$$

In (11) and (13) the quantity B_{λ} is the reduced transition probability which we have defined to be

$$\begin{aligned} B_{\lambda}(n_i j_i \rightarrow n_f j_f) &= \sum_{\mu} |\langle n_f j_f | (R/2)^{\lambda} Y_{\lambda}^{\mu}(\hat{R}) | n_i j_i \rangle|^2 \\ &= \frac{1}{2j_i+1} |\langle n_f j_f || (R/2)^{\lambda} Y_{\lambda} || n_i j_i \rangle|^2 \end{aligned} \quad (15)$$

where

$$\begin{aligned} \langle n_f j_f || (R/2)^{\lambda} Y_{\lambda} || n_i j_i \rangle \\ = 2^{-\lambda} A_{\lambda}(n_i \rightarrow n_f) \langle j_f || Y_{\lambda} || j_i \rangle \end{aligned} \quad (16)$$

with the vibrational matrix elements

$$A_{\lambda}(n_i \rightarrow n_f) = \int_0^{\infty} \chi_{n_f}(R) R^{\lambda} \chi_{n_i}(R) dR \quad (17)$$

and the reduced matrix element

$$\langle j_f || Y_{\lambda} || j_i \rangle = (-1)^{j_i+j_f} \left\{ \frac{2\lambda+1}{4\pi} (2j_i+1)(2j_f+1) \begin{pmatrix} j_i & \lambda & j_f \\ 0 & 0 & 0 \end{pmatrix} \right\}^{\frac{1}{2}} \quad (18)$$

3. The Symmetrization of Classical Cross-Sections

In the present approach, the projectile orbit has been described classically and consequently the cross-sectional expressions (11) and (13) do not distinguish between the initial and final states of the trajectories. One of the most successful ways of introducing the initial and final parameters for the orbits is to impose the principle of reciprocity on the cross-sectional expressions and replace various classical parameters by their corresponding quantal analogues via the correspondence principle. To satisfy the reciprocity relation between the cross-section σ_{if} for the direct process and σ_{fi} for its inverse, we must have

$$\omega_i v_i^{-1} \sigma_{if} = \omega_f v_f^{-1} \sigma_{fi} \quad (19)$$

where ω_i , v_i and ω_f and v_f are respectively the statistical weights and velocities (in a.u.) for the initial and final states of the system. For the present problem we have

$$\omega_i = (2j_i + 1) \quad \text{and} \quad \omega_f = (2j_f + 1) \quad (20)$$

From (19) and (20) we conclude that σ_{if} must have the form

$$\sigma_{if} = \frac{v_f}{v_i} \sqrt{\frac{2j_f + 1}{2j_i + 1}} \times \text{(Cross-sectional expression symmetric in } i \text{ and } f) \quad (21)$$

Extending Kramers type prescriptions for the principal quantum number $n \rightarrow n + 1$ to the positive energy continuum where n is replaced by $i\eta$, Biedenharn and Brussard (1965) obtain the correspondence

$$\eta \rightarrow (\eta^2 + 1)^{\frac{1}{2}} \quad (22)$$

for the so-called Sommerfeld number η where

$$\begin{aligned} \eta^2 &= k_0 |z_1 z_2 e^2 m| \\ &= \frac{(z_1 z_2 e^2)^2}{\hbar^2 v_0^2} \end{aligned} \quad (23)$$

is given in terms of half the distance of closest approach r_0 , or the incident velocity v_0 . In (23) η and z_1 are the mass and charge of the projectile and z_2 stands for the effective charge of the target. From the fundamental correspondence between the quantum mechanical matrix elements and the classical Fourier components and from the relation (22), it can be shown (for a concise derivation and an elegant discussion see, Biedenharn and Brussard, 1965) that the classical η yields the final correspondence

$$\eta \rightleftharpoons \left\{ \frac{2(\eta_i^2 + 1)(\eta_f^2 + 1)}{(\eta_i^2 + 1)^{\frac{1}{2}} + (\eta_f^2 + 1)^{\frac{1}{2}}} \right\}^{\frac{1}{3}} \quad (24)$$

We note that in terms of the initial and final parameter η_i and η_f the quantity ξ defined in (10) reduces to

$$\xi = \eta_i - \eta_f \quad (25)$$

For the cross-sectional expression to be inserted in equation (21) we shall adopt the classical expression (13) and symmetrise it through the use of equations (22), (23) and (24) as follows:

$$\frac{1}{v_i^2} \Rightarrow \{(\eta_i^2+1)(\eta_f^2+1)\}^{\frac{1}{2}} \quad (26)$$

$$\lambda_0^{-2\lambda+2} \Rightarrow \left\{ \frac{2(\eta_i^2+1)(\eta_f^2+1)}{(\eta_i^2+1)^{\frac{1}{2}} + (\eta_f^2+1)^{\frac{1}{2}}} \right\}^{\frac{4-4\lambda}{3}} \quad (27)$$

Finally, substituting (26) and (27) in (13) and combining the result with (21) we obtain

$$\sigma_{if} = \frac{\eta_i}{\eta_f} \{(\eta_i^2+1)(\eta_f^2+1)\}^{\frac{1}{2}} \left\{ \frac{2(\eta_i^2+1)(\eta_f^2+1)}{(\eta_i^2+1)^{\frac{1}{2}} + (\eta_f^2+1)^{\frac{1}{2}}} \right\}^{\frac{4-4\lambda}{3}} \times$$

$$\times 4B_\lambda (n_i j_i \rightarrow n_f j_f) f_\lambda (\xi = \eta_i - \eta_f) \quad (28)$$

An expression similar to (28) is obtained for the differential cross-section with $f_\lambda(\xi)$ replaced by $\frac{df_\lambda(\xi, \theta)}{d\Omega}$

We shall emphasize the fact that the symmetrization procedure adopted here for the classical cross-sections is by no means unique in character.

Nevertheless it has been found that an essentially similar procedure adopted by Biedenharn and Brussaard (1965) in nuclear excitation problems, produced excellent agreement with the corresponding quantal calculations. We note, however, that our symmetrization procedure has the advantage over that of the previous authors in that it yields the correct threshold law: $\sigma_{if} \approx e^{-2\pi\eta_f}$

(Wigner, 1968) for the repulsive Coulomb scattering while the other expression yields

$\sigma_{ij} \approx \frac{1}{\eta^3} e^{-2\pi\eta}$ near the threshold. The two procedures, however, differ little numerically somewhat away from the threshold.

4. Numerical Calculations

The vibrational matrix elements are calculated by using Morse functions (Morse, 1929) for the target H_2^+ molecular ions:

$$\psi_n(R) = N_n e^{-\frac{1}{2}z} z^{(k-2n-1)} F_n(z)$$

where

$$F_n(z) = \sum_{i=0}^n \frac{(-1)^i \binom{n}{i} z^i}{(k-2n)(k-2n+1)(k-2n+i+1)} \quad (29)$$

$$z = k e^{-\beta(R-R_0)}, \quad R_0 = R \text{ at equilibrium,}$$

$$k = \frac{l}{x_0},$$

$$N_n^2 = \frac{(k-n-1)(k-n-2)\dots(k-2n)}{n! \Gamma(k-2n-1)} \beta, \quad \beta^2 = 2M_0 \omega_0 x_0, \text{ and } \omega_0 = D_0 - \sqrt{D_0^2 - 2E_{01}D_0},$$

where $x_0 = \frac{\omega_0}{8\pi D_0}$ is the spectroscopic parameter, M_0 is the reduced mass, D_0 is the dissociation energy and E_{01} is the energy difference of the first-excited state from the ground state.

For H_2^+ molecular ions we choose

$$D_0 = 0.2053 \text{ Ryd.}$$

$$\omega_0 = 0.0212 \text{ Ryd.}$$

$$M_0 = 918.334 \text{ a.u.}$$

The energy difference E_{01} is calculated by Cohen et al. (1960) and is very nearly equal to 0.02 Ryd.

Following Heaps and Herzberg (1952) the vibrational matrix elements are found to be

$$\int \Psi_{n_f}(R) R^2 \Psi_{n_i}(R) dR = \frac{(-1)^{n_i+n_f+1}}{\beta^2 n_f!} \frac{2 C_{n_f n_i} H_{n_f n_i}}{\{1 - (n_f+1)x_e\}}$$

where

$$C_{n_f n_i} = \left(\frac{\{1 - (2n_i+1)x_e\} \{1 - (2n_f+1)x_e\} x_e^{n_f-n_i} n_f!}{\{1 - (n_i+1)x_e\} \{1 - (n_i+2)x_e\} \dots \{1 - n_f x_e\} n_i!} \right)^{\frac{1}{2}}$$

and $H_{n_f n_i}$ is the polynomial the first few of which are

$$H_{n_f 0} = (n_f - 1)! \ln \left(1 - \left(n_f + \frac{3}{2} \right) x_e \right) + a_{n_f}$$

with

$$a_1 = 0, \quad a_2 = 1, \quad a_3 = 3, \quad a_4 = 11, \quad a_5 = 50, \dots$$

5. Results and Discussions

We present here both the differential as well as the total cross-sections for vibro-rotational transitions between the ground state and the first few excited vibrational-rotational states. In figures 2 and 3 we compare the difference between different rotational excitations and a fixed vibrational transition $n_i = 0 \rightarrow n_f = 1$. We find that the cross-sections for $j_i = 0 \rightarrow j_f = 2$ is by far the largest compared to the rest and those between $j_i = 2, 4, 6 \rightarrow j_f = 4, 6, 8$ respectively are comparable with each other. In figures 4 and 5 we have similar results between the same set of rotational states but with a different vibrational transition: $n_i = 0 \rightarrow n_f = 2$. Comparing Fig. 4 and 5 with Fig. 2 and 3 we find that the magnitude of all the $0 \rightarrow 2$ cross sections are an order of magnitude or more larger than those for the $0 \rightarrow 1$ transitions. Finally in Fig. 6 we compare the total cross-sections for a fixed rotational excitation $j_i = 0 \rightarrow j_f = 2$, with various vibrational excitations: $n_i = 0 \rightarrow n_f = 1$, $n_i = 0 \rightarrow n_f = 2$, and $n_i = 0 \rightarrow n_f = 3$.

This result again shows that the $n_i = 0 \rightarrow n_f = 2$ vibrational transition is much larger than those for $n_i = 0 \rightarrow n_f = 1$ and $n_i = 0 \rightarrow n_f = 3$; while the last two cross-sections are comparable in magnitude. Although we have not plotted the cross sections for $n_i = 0 \rightarrow n_f = 4$, we noted that it was somewhat smaller than that for $n_i = 0 \rightarrow n_f = 3$ in the energy range under consideration.

We believe that the present method is particularly suitable for the energy range $E_i = 0.0$ to $E_i \leq 0.5$ Ryd. for the following reasons.

- (1) In this energy range the distance of closest approach is $\geq 2 a_0$ and is expected to be large enough to allow essentially non-penetrating orbits for the projectiles.

(ii) The threshold for positronium formation being at 0.5 Ryd., we may neglect such channels for positron energies below 0.5 Ryd.

We conclude by noting that the calculated absolute transition probabilities are all very small so that the application of a first order theory is generally satisfactory.

Acknowledgments

I would like to thank Dr. A. Temkin for his helpful comments and
Drs. A. B. Ritchie and D. Morgan for their interest in this work.

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