H^- formation in proton-helium collisions

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The process of formation of H^- as a result of double electron capture by protons from helium is investigated using an atomic-state expansion in which three traveling atomic orbitals have been retained. The cross sections have been presented for incident proton energy ranging from 10 to 200 keV. The results are compared with the previous experimental and theoretical values. Our results show a maximum in the cross-section energy curve in qualitative agreement with the experimental findings.

INTRODUCTION

The process in which a proton captures a single electron from a helium atom has been studied in detail both theoretically[1] and experimentally[2] by several workers. The cross sections for the formation of H^- as a result of double electron capture by a proton passing through helium have been measured experimentally for low incident proton energy (<65 keV) by Fogel *et al.*,[3] Williams,[4] and Schryber.[5] Recently Toburen and Nakai[6] have measured the same cross sections for higher incident energies of proton, viz., in the energy region of 75 to 200 keV. Though quite a large number of experiments have been performed to measure the double-electron-capture cross section in the proton-helium collision, no reliable theoretical calculations are available for comparison with the experimental findings.

The only theoretical attempt so far made in this direction is Gerasimenko's.[7] He used the Born approximation and calculated the double-capture cross section for the incident proton energy varying between 150 and 750 keV. The results of Gerasimenko are orders of magnitude too large compared to the experimental results. Furthermore, with a decrease in the incident energy, the behavior of the cross sections calculated by Gerasimenko is quite different from that experimentally observed. The experimental cross section reaches a maximum at about 35 keV and steadily decreases with further decrease in the incident ion energy. Gerasimenko did not calculate the Born cross sections for such incident energies. However, from the trend of his results (Fig. 1) it may be inferred that if the Born calculations are carried out to lower energies, any maximum will have a value several orders of magnitude greater than the experimental findings.

The occurrence of a maximum in the observed double-capture cross section and the absence of any theoretical calculation in the corresponding energy region have led us to make a fresh theoretical investigation into this problem. In this paper we propose to investigate the above collision process in a three-state approximation[8]; we have considered the following possibilities:

$$H^+ + He \rightarrow H^+ + He \tag{1a}$$

$$\rightarrow H + He^+ \tag{1b}$$

$$\rightarrow H^- + He^{++}, \tag{1c}$$

where He, He^+, H, and H^- are the ground states of the helium atom, the helium ion, the hydrogen atom and the negative hydrogen ion, respectively. This problem with two active electrons is especially complicated and the associated computational difficulty is enormous. The matrix elements involve oscillatory momentum transfer terms and their evaluation is formidably difficult. Consequently we have to limit our investigation to calculations with the inclusion of the initial channel, the final channel, and only one intermediate channel. The intermediate channel (1b) we have considered is that in which a single electron has been captured by the proton in the $1s$ state and the helium ion remains in its ground state. From previous calculations,[1] it has been found that the cross sections for the single capture into the excited states are much smaller than those for capture in the ground state. Hence the intermediate channel (1b) is likely to play a more important role than those in which single capture into excited states occurs.

THEORY

The wave function Ψ for the proton-helium system is approximated by the expansion

$$\Psi = a_1(t)\psi_1 + [a_2(t)/\sqrt{2}](\psi_{21} + \psi_{22}) + a_3(t)\psi_3, \tag{2}$$

where the $a_i(t)$'s are the coefficients of expansion dependent on time t only and ψ_1, ψ_{21}, ψ_{22}, and ψ_3 are the traveling orbitals, given by (atomic units

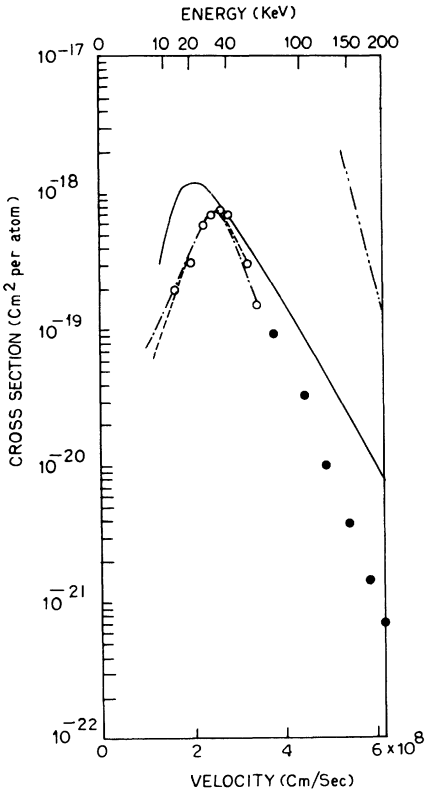

FIG. 1. Cross sections for formation of H^- as a result of double electron capture by protons from helium compared with previous experimental and theoretical results. Experimental: –•–, Ref. 3; ---, Ref. 4; ○, Ref. 5; ●, Ref. 6. Theoretical: ———, this work; – – – –, Ref. 7.

are used throughout)

$$\psi_1 = \phi_{He}(r_{1A}, r_{2A}) \times \exp\left[-i\epsilon_{He}t - \tfrac{1}{4}iv^2t - \tfrac{1}{2}i\vec{v}\cdot(\vec{r}_1+\vec{r}_2)\right], \quad (3)$$

$$\psi_{21} = \phi_H(r_{1B})\phi_{He^+}(r_{2A}) \times \exp\left[-i(\epsilon_H+\epsilon_{He^+})t - \tfrac{1}{4}iv^2t - \tfrac{1}{2}i\vec{v}\cdot(\vec{r}_2-\vec{r}_1)\right], \quad (4)$$

$$\psi_{22} = \phi_H(r_{2B})\phi_{He^+}(r_{1A}) \times \exp\left[-i(\epsilon_H+\epsilon_{He^+})t - \tfrac{1}{4}iv^2t - \tfrac{1}{2}i\vec{v}\cdot(\vec{r}_1-\vec{r}_2)\right], \quad (5)$$

$$\psi_3 = \phi_{H^-}(r_{1B}, r_{2B}) \times \exp\left[-i\epsilon_{H^-}t - \tfrac{1}{4}iv^2t + \tfrac{1}{2}i\vec{v}\cdot(\vec{r}_1+\vec{r}_2)\right], \quad (6)$$

where ϕ_{He}, ϕ_H, ϕ_{He^+}, and ϕ_{H^-} are the normalized wave functions for helium, hydrogen, ionized helium, and the negative hydrogen ion, respectively; $\vec{r}_{1A}$, $\vec{r}_{2A}$, $\vec{r}_{1B}$, and $\vec{r}_{2B}$ are the position vectors of electrons 1 and 2 from the helium nucleus A and the proton B, respectively, and ϵ_{He}, ϵ_H, ϵ_{He^+}, and ϵ_{H^-} are the eigenenergies of the respective atoms and ions. The vectors $\vec{r}_1$ and $\vec{r}_2$ are the position vectors of the two electrons from the midpoint of the line joining A and B. For the helium and the negative hydrogen ion we have used the wave functions of Shull and Löwdin[9] and Löwdin,[10] respectively.

The equations for finding the coefficients a_i ($i = 1$ to 3) are obtained by using a variational principle.[11] Utilizing the symmetry properties of the wave functions, the final set of coupled differential equations in a_i is written as

$$i(\dot{a}_1 + \sqrt{2}f_{1,21}\dot{a}_2 + f_{13}\dot{a}_3) - F_{11}a_1 - \sqrt{2}(F_{1,21} - \tfrac{1}{2}i\dot{f}_{1,21})a_2 - (F_{13} - \tfrac{1}{2}i\dot{f}_{13})a_3 = 0, \quad (7a)$$

$$i[\sqrt{2}\bar{f}_{1,21}\dot{a}_1 + (1+f_{21,22})\dot{a}_2 + \sqrt{2}f_{21,3}\dot{a}_3] - \sqrt{2}(\bar{F}_{1,21} - \tfrac{1}{2}i\dot{\bar{f}}_{1,21})a_1 - (F_{21,21} + F_{21,22} - \tfrac{1}{2}i\dot{f}_{21,22})a_2 - \sqrt{2}(F_{21,3} - \tfrac{1}{2}i\dot{f}_{21,3})a_3 = 0, \quad (7b)$$

$$i(\bar{f}_{13}\dot{a}_1 + \sqrt{2}\bar{f}_{21,3}\dot{a}_2 + \dot{a}_3) - (\bar{F}_{13} - \tfrac{1}{2}i\dot{\bar{f}}_{13})a_1 - \sqrt{2}(\bar{F}_{21,3} - \tfrac{1}{2}i\dot{\bar{f}}_{21,3})a_2 - F_{33}a_3 = 0 \quad (7c)$$

(bars denote complex conjugates), where

$$f_{1,21} = \int \phi_{He}(r_{1A}, r_{2A})\phi_H(r_{1B})\phi_{He^+}(r_{2A}) \exp\left[i(\epsilon_{He} - \epsilon_H - \epsilon_{He^+})t + i\vec{v}\cdot\vec{r}_1\right] d\vec{r}_1\, d\vec{r}_2,$$

$$f_{13} = \int \phi_{He}(r_{1A}, r_{2A})\phi_{H^-}(r_{1B}, r_{2B}) \exp\left[i(\epsilon_{He} - \epsilon_{H^-})t + i\vec{v}\cdot(\vec{r}_1+\vec{r}_2)\right] d\vec{r}_1\, d\vec{r}_2,$$

$$f_{21,22} = \int \phi_H(r_{1B})\phi_{He^+}(r_{2A})\phi_H(r_{2B})\phi_{He^+}(r_{1A}) \exp\left[i\vec{v}\cdot(\vec{r}_2 - \vec{r}_1)\right] d\vec{r}_1\, d\vec{r}_2,$$

$$f_{21,3} = \int \phi_H(r_{1B})\phi_{He^+}(r_{2A})\phi_{H^-}(r_{1B}, r_{2B}) \exp\left[i(\epsilon_H + \epsilon_{He^+} - \epsilon_{H^-})t + i\vec{v}\cdot\vec{r}_2\right] d\vec{r}_1\, d\vec{r}_2 ,$$

$$F_{11} = \int \phi_{He}(r_{1A}, r_{2A}) V_1 \phi_{He}(r_{1A}, r_{2A})\, d\vec{r}_1\, d\vec{r}_2 ,$$

$$F_{1,21} = \int \phi_{He}(r_{1A}, r_{2A}) \tfrac{1}{2}(V_1 + V_{21}) \phi_H(r_{1B})\phi_{He^+}(r_{2A}) \exp\left[i(\epsilon_{He} - \epsilon_H - \epsilon_{He^+})t + i\vec{v}\cdot\vec{r}_1\right] d\vec{r}_1, d\vec{r}_2 ,$$

$$F_{13} = \int \phi_{He}(r_{1A}, r_{2A}) \tfrac{1}{2}(V_1 + V_3) \phi_{H^-}(r_{1B}, r_{2B}) \exp\left[i(\epsilon_{He} - \epsilon_{H^-})t + i\vec{v}\cdot(\vec{r}_1 + \vec{r}_2)\right] d\vec{r}_1\, d\vec{r}_2 ,$$

$$F_{21,21} = \int \phi_H(r_{1B})\phi_{He^+}(r_{2A}) V_{21} \phi_H(r_{1B})\phi_{He^+}(r_{2A})\, d\vec{r}_1\, d\vec{r}_2 ,$$

$$F_{21,22} = \int \phi_H(r_{1B})\phi_{He^+}(r_{2A}) \tfrac{1}{2}(V_{21} + V_{22}) \phi_H(r_{2B})\phi_{He^+}(r_{1A}) \exp\left[i\vec{v}\cdot(\vec{r}_2 - \vec{r}_1)\right] d\vec{r}_1\, d\vec{r}_2 ,$$

$$F_{21,3} = \int \phi_H(r_{1B})\phi_{He^+}(r_{2A}) \tfrac{1}{2}(V_{21} + V_3) \phi_{H^-}(r_{1B}, r_{2B}) \exp\left[i(\epsilon_H + \epsilon_{He^+} - \epsilon_{H^-})t + i\vec{v}\cdot\vec{r}_2\right] d\vec{r}_1\, d\vec{r}_2 ,$$

and

$$F_{33} = \int \phi_{H^-}(r_{1B}, r_{2B}) V_3 \phi_{H^-}(r_{1B}, r_{2B})\, d\vec{r}_1\, d\vec{r}_2 ,$$

with

$$V_1 = -\frac{1}{r_{1B}} - \frac{1}{r_{2B}}, \quad V_{21} = -\frac{2}{r_{1A}} + \frac{1}{r_{12}} - \frac{1}{r_{2B}},$$

$$V_{22} = -\frac{2}{r_{2A}} + \frac{1}{r_{12}} - \frac{1}{r_{1B}}, \quad \text{and} \quad V_3 = -\frac{2}{r_{1A}} - \frac{2}{r_{2A}}.$$

The overlap integrals f_{mn} cannot be evaluated analytically but are expressed in the form of one-dimensional time integrals.[12] The analytical evaluation of the interaction integrals F_{11}, $F_{21,21}$, and F_{33} are always possible; the integrals $F_{1,21}$, F_{13}, and $F_{21,3}$ are, however, expressed as one-dimensional time integrals as in the case of f_{mn}. The two-centered exchange interaction integral $F_{21,22}$ contains a momentum transfer term and is not easily tractable. A Mulliken-type approximation[11] has been used for its determination. For the evaluation of the matrix elements involving the momentum transfer terms we use a method similar to that of Cheshire,[13] Chatterjee *et al.*,[14] and Mukherjee *et al.*[12]

We finally solve numerically the set of equations (7) using the initial conditions $a_1 = 1$ and $a_i(i \neq 1) = 0$ at $t = -\infty$. The values of the three quantities $|a_i|^2$ for $i = 1$ to 3 at $t = +\infty$ give the probabilities for the three processes [Eq. (1)] for the given velocity v and the impact parameter p. Multiplying the probabilities by $2\pi p\, dp$ and integrating over p from zero to infinity, the total cross sections for the corresponding processes are obtained.

RESULTS AND DISCUSSIONS

In Fig. 1, we have shown our calculated results for the cross sections for formation of H^- as a result of double electron capture by protons from helium for the incident energy 10–200 keV, as functions of incident proton energy. For comparison, we have also given in the same figure the experimental[3–6] and the previous theoretical results.[7] Our results for the double-capture cross section are in fair agreement with the measured cross sections of Fogel *et al.*,[3] Williams,[4] and Schryber.[5] The only existing theoretical results for the problem under consideration are Gerasimenko's[7]; they are for the high-energy region and are more than two orders of magnitude greater than the experimental values. In the low-energy region, with the increase of incident energy, the experimental results[3–5] show a peak in the cross sections and then gradually decrease. In our present calculation we also find the same nature, as is clear from our curve shown in Fig. 1. The maximum value of the cross section (1.3×10^{-18} cm^2) occurs in our calculation near the incident ion energy of 20 keV, whereas the experimental cross section (0.7×10^{-18} cm^2) reaches a maximum at about 35 keV. The calculated value of the cross section is somewhat higher than that observed experimentally. In the higher-energy region covered by Toburen and Nakai,[6] i.e., for 75–200 keV, our theoretical results show a much better trend as com-

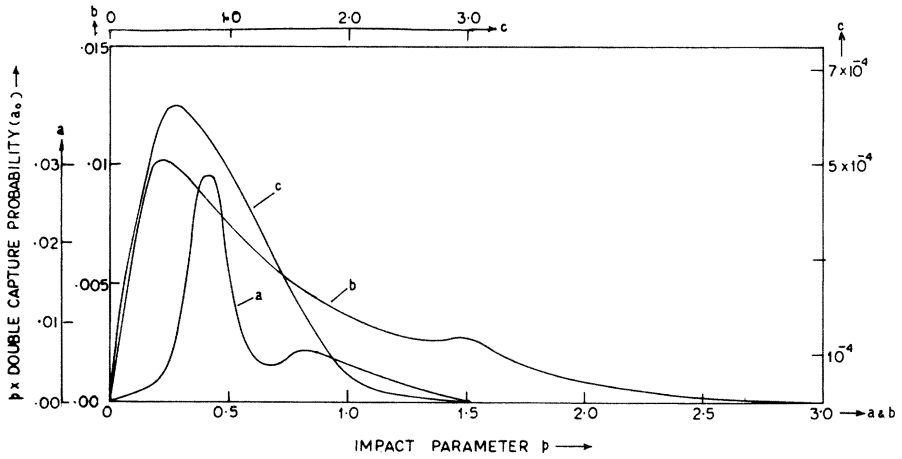

FIG. 2. Product of the impact parameter p and the probability for double electron capture at (a) 10, (b) 20, and (c) 100 keV as a function of p.

pared to the Born results of Gerasimenko but do not agree quantitatively with the experimental findings. The discrepancy between the present theoretical values and the experimental findings may be attributed to the effect of continuum states ignored in our calculation.

We have used the Mulliken-type approximation for the calculation of the integral $F_{21,22}$ occurring in Eq. (7). This approximation gives a good estimate of the integral when the colliding nuclei are close to each other. With an increase in the internuclear distance the percentage error increases but the value of the integral itself becomes small. It may be noted that the integral $F_{21,22}$ occurs only in Eq. (7b) of our coupled set of differential equations. This indicates that any change in the double-capture cross section by protons from helium owing to uncertainty in the integral $F_{21,22}$ is caused by the coupling with the state of single capture. Furthermore, the single-capture cross section computed in the present work is in agreement with those obtained by other workers.[1] Hence we may conclude that the use of the Mulliken-type approximation does not cause any serious error in the present calculation.

We have presented in Fig. 2 our results for the probabilities of the double capture at incident protor energies of 10, 20, and 100 keV, respectively. At an incident ion energy of 10 keV, the probability curve has a maximum at impact parameter $p = 0.4$ and then diminishes rapidly but shows a hump near $p = 0.8$. For the incident ion energy of 20 keV, the probability curve has a maximum at $p = 0.2$ and does not diminish as rapidly as in the case of 10 keV. This curve also shows a hump near $p = 1.5$. In the 100-keV case, the probability curve shows a maximum at $p = 0.5$. Unfortunately, no experimental or theoretical results exist to compare our calculated values of these probabilities.

The calculated values of our single-capture cross sections almost coincide with the previous theoretical values[1] and as such these results are not presented here. All numerical computations have been performed on the IBM 370 at the Indian Institute of Technology, Madras.

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[1]L. T. Sin FaiLam, Proc. Phys. Soc. Lond. 92, 67 (1967); B. H. Bransden and L. T. Sin FaiLam, Proc. Phys. Soc. Lond. 87, 653 (1966); T. A. Green, H. E. Stanley and Y. C. Chiang, Helv. Phys. Acta. 38, 109 (1965); Thomas G. Winter and Chun C. Lin, Phys. Rev. A 10, 2141 (1974).

[2]P. M. Stier and C. F. Barnett, Phys. Rev. 103, 896 (1956); C. F. Barnett and H. K. Reynolds, Phys. Rev. 109, 355 (1958); F. P. Ziemba, G. J. Lockwood, G. H. Morgan, and E. Everhart, Phys. Rev. 118, 1552 (1960); H. F. Helbig and E. Everhart, Phys. Rev. 136, A674 (1964); S. K. Allison and M. Garcia-Munoz, in *Atomic and Molecular Processes*, edited by D. R. Bates (Academic, New York, 1962), p. 753.

[3]Ya. M. Fogel, R. V. Mitin, V. G. Kozlov, and N. D. Romashko, Zh. Eksp. Teor. Fiz. 35, 565 (1958) [Sov. Phys.—JETP 8, 390 (1959)].

[4]J. F. Williams, Phys. Rev. 150, 7 (1966).

[5]Von. U. Schryber, Helv. Phys. Acta. 39, 562 (1966).

[6]L. H. Toburen and M. Y. Nakai, Phys. Rev. 177, 191

(1969).
[7]V. I. Gerasimenko, Zh. Eksp. Teor. Fiz. 41, 1104 (1961). [Sov. Phys.—JETP 14, 789 (1962)].
[8]A short communication with results for a single incident energy has already been published in Phys. Lett. 51A, 539 (1975).
[9]H. Shull and P. O. Löwdin, J. Chem. Phys. 25, 1035 (1956).
[10]P. O. Löwdin, Phys. Rev. 90, 120 (1953).
[11]K. Roy and S. C. Mukherjee, Phys. Rev. A 7, 130 (1973).
[12]S. C. Mukherjee, K. Roy, and N. C. Sil, J. Phys. B 6, 467 (1973).
[13]I. M. Cheshire, Proc. Phys. Soc. Lond. 92, 862 (1967).
[14]G. Chatterjee, D. M. Bhattacharya, and N. C. Sil, Indian J. Phys. 41, 934 (1967).