Electron removal from atomic hydrogen by collisions with fully stripped carbon*

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Total cross sections for charge transfer and impact ionization in C^{+6}-H collisions have been calculated for the range of relative velocities $(0.1–10)\times 10^8$ cm/sec. At the lower velocities [$(0.1–2)\times 10^8$ cm/sec] coupled-state calculations in an impact-parameter approximation were made of the charge-transfer cross sections using a basis set of exact one-electron two-center wave functions. An investigation of the origin dependence of these perturbed stationary-state calculations showed that differences of about 30% in the cross sections occur when the origin is shifted from the target to the ion center. At the higher velocities a classical-trajectory Monte Carlo approach was employed to determine both the charge-transfer and impact-ionization cross sections. The two sets of calculations are in reasonable agreement in the overlapping energy range, and give a comprehensive picture of the overall electron-removal cross section. They show that electron capture dominates in the region between 0.1×10^8 and 3×10^8 cm/sec with a peak cross section of $\sim 5\times 10^{15}$ cm^2 at $v\approx 7\times 10^7$ cm/sec. The capture is predominantly into the $n=4$ levels of the product C^{+5} ion. Impact ionization is found to be the most important electron removal process at velocities above 4×10^8 cm/sec.

INTRODUCTION

The mechanisms of electron removal from atomic hydrogen in collisions with highly charged projectile ions are of considerable interest, not only from a theoretical point of view, but also in connection with some very practical technological problems. For example, in neutral hydrogen-beam-heated Tokamak plasmas where a small percentage of highly stripped impurity ions of C, O, N, and heavier atoms are present,[1] such electron-loss processes are believed responsible for a deleterious beam-trapping instability.[2]

The reactions leading to electron loss in collisions between a fully stripped ion A^{+q} of charge state q and a hydrogen atom can be represented as

$$A^{+q}+\mathrm{H}\rightarrow A^{+q-1}+H^{+} \quad \text{(charge transfer)} \qquad (1a)$$

and

$$A^{+q}+\mathrm{H}\rightarrow A^{+q}+H^{+}+e \quad \text{(impact ionization)}. \qquad (1b)$$

Each of these reactions is significant in a different velocity range with charge transfer most important at the lower velocities, generally below $(1 \text{ or } 2)\times 10^8$ cm/sec, and the ionization process dominating at higher energies.

In this paper we report calculations of total cross sections for both charge transfer and impact ionization for the $C^{+6}+H$ system. Previous work on the charge-transfer process has included a study using a multistate Landau-Zener method[3] and another investigation which employed an absorbing sphere model also based on the Landau-Zener approximation.[4] Recently, Bottcher[5] has calculated charge-transfer cross sections for a number of stripped ion-hydrogen atom systems including $C^{+6}+H$. His model also involves a number of crossings between diabatic states, but in this approach, a simple analytic form is found for the coupling matrix elements and the transition probabilities are constrained to preserve unitarity.

In the present work, a more rigorous approach is followed to study the capture process in the velocity range $(0.1–2)\times 10^8$ cm/sec. For one-electron diatomic molecular systems, exact adiabatic eigenenergies (potential curves), wave functions and coupling matrix elements can be calculated with comparative economy.[6,7] We have therefore made coupled-state calculations within the impact-parameter approximation using an appropriate truncated molecular basis set to calculate the total charge-transfer cross sections. A related study on the $O^{+8}+H$, $B^{+4}+H$, and $Be^{+5}+H$ systems has recently been reported by Harel and Salin.[8] Complementing the coupled -state calculations, a classical-trajectory Monte Carlo approach,[9,10] that has been found to give results which compare well with recent multicharged-ion charge-transfer experiments,[11,12] was used to calculate both the charge-transfer and the impact-ionization cross sections in the velocity range $(2–10)\times 10^8$ cm/sec.

THEORETICAL METHODS

A. Coupled-state calculations

In these cross-section calculations, we have used the rectilinear trajectory-impact parameter version of the method of perturbed stationary states. Since the method has been thoroughly discussed in the literature,[6,13] we present here only a brief description of the approach. Atomic units are used throughout.

The time-dependent Schrödinger equation for the colliding system

$$\left(H_{\rm el} - i\frac{\partial}{\partial t}\right)\psi = 0 \tag{2}$$

is solved for each given impact parameter. Here ψ is the time-dependent molecular wave function and $H_{\rm el}$ is the electronic Hamiltonian of the collision system. With ψ expanded in a suitable basis set of one-electron two-center wave functions calculated from

$$[H_{\rm el} - \epsilon_j(R)]\Phi_j = 0, \tag{3}$$

we obtain from Eq. (2) the standard set of coupled equations for the amplitudes a_j,

$$\frac{d}{dt}a_j(t) = -\sum_{j'} a_{j'}(t)\left\langle \Phi_j \left| \frac{\partial}{\partial t} \right| \Phi_{j'} \right\rangle \times \exp\{-i[S_{j'}(t) - S_j(t)]\}. \tag{4}$$

In these equations, Φ_j are the adiabatic molecular wave functions, ϵ_j the corresponding energy eigenvalues obtained as a function of the nuclear separation R, (adiabatic potential curves), j denotes the molecular quantum state (usually the united atom quantum numbers), and the phase factors S_j are given by

$$S_j(t) = \int_{-\infty}^{t} \epsilon_j(R)\, dt' \tag{5}$$

Determination of the matrix elements of Eq. (4) necessitates the calculation of either the radial coupling $\langle \Phi_j | \partial/\partial R | \Phi_{j'} \rangle$ or rotational coupling $\langle \Phi_j | iL_y | \Phi_{j'} \rangle$ matrix elements depending on the symmetries of the states involved.

Power's code[14,15] is used to compute the relevant potential curves for this system. The Schrödinger Eq. (3) for the one-electron two-center system is separable in prolate-spheroidal coordinates such that

$$\Phi_j = L(\xi)M(\eta)\Omega(\varphi), \tag{6}$$

where

$$\xi = (r_1 + r_2)/R, \tag{7}$$

$$\eta = (r_1 - r_2)/R, \tag{8}$$

and $\vec{r}_1$ and $\vec{r}_2$ are the position vectors of the electron with respect to the target proton and the projectile, respectively. A very efficient continued-fraction algorithm is employed giving the energy eigenvalues and separation constants as a function of R.

The matrix elements required in the calculations are determined using a code developed by Salin and his associates in Bordeaux.[6] In this program, the series representations for $L(\xi)$ and $M(\eta)$ are identical to that used by Hunter and Pritchard,[16] and the radial and rotational coupling matrix elements between states Φ_j and $\Phi_{j'}$ are obtained using the operators

$$\frac{\partial}{\partial R} = \frac{-(2/R^2)}{\epsilon_j - \epsilon_{j'}} \frac{(q+1)\xi + (q-1)\eta}{(\xi^2 - \eta^2)} + \frac{1}{R} \frac{\xi(1-\xi^2)\frac{\partial}{\partial \xi} + \eta(\eta^2 - 1)\frac{\partial}{\partial \eta}}{\xi^2 - \eta^2} \tag{9}$$

and

$$iL_y = \frac{(\xi^2-1)^{1/2}(1-\eta^2)^{1/2}}{\xi^2 - \eta^2}\cos\varphi\left(\eta\frac{\partial}{\partial \xi} - \xi\frac{\partial}{\partial \eta}\right) - \xi\eta\sin\varphi\,(\xi^2-1)^{-1/2}(1-\eta^2)^{-1/2}\frac{\partial}{\partial \varphi}, \tag{10}$$

where q is the projectile charge. (The target atom is assumed to be hydrogen.)

The above operator expressions were derived for an origin of coordinates chosen at the midpoint between the nuclei, but they can be easily generalized for any arbitrary origin.

Once the relevant truncated basis set is determined and the associated potential-energy curves and matrix elements computed, the coupled equations (4) are integrated using standard methods. The initial condition corresponds to the electron bound to the proton in ground-state hydrogen.

In this study we have adopted a procedure related to that discussed by Piacentini and Salin[6] which is designed to avoid the problems associated with momentum transfer to the electron in the capture process. In this prescription, the origin for computing the matrix elements is placed on the target proton.[17] Under these conditions, the charge-exchange probability, corresponding to a given impact parameter ρ is given by

$$|b_{CX}(\rho)|^2 = 1 - \sum_{j'} |a_{j'}(+\infty)|^2, \tag{11}$$

where the summation is taken over all states j' for which the electron asymptotically remains on the proton. The total charge-transfer cross section is then obtained by an integration over impact parameters.

B. Classical-trajectory calculations

The classical-trajectory Monte Carlo method used in this work is based on the procedures developed by Abrines and Percival[9] for $H^+ + H$ collisions and by Karplus *et al.*,[10] and has been described in detail in recent papers.[11,12] The general approach involves the solution of the classical equations of motion for a three body system (in the present case, the electron, proton and the C^{+6} projectile). Hamilton's equations (12 coupled first-order differential equations) are solved numerically for a large number of trajectories which includes a random selection of impact parameters. Following Abrines and Percival,[9] the spherically

symmetric hydrogen ground states is represented classically by a microcanonical momentum distribution corresponding to a bound-state energy of 0.5 atomic units.

For each trajectory, the projectile is placed at a large (effectively asymptotic) distance from the target atom. The equations of motion are integrated and the projectile is followed as it approaches, interacts strongly with and then separates from the target atom. The integration is halted when once again the projectile is far enough from the target so that the interaction is negligible. If, at the termination of the trajectory, the electron is still bound to the proton, then no reaction has occurred. If, at termination, the electron is bound to the projectile, then charge transfer has occurred. If the electron is not bound either to the target or to the projectile, then ionization has occurred. The number of ionizations and electron captures occurring during approximately two thousand trajectories at each collision energy are separately counted and the cross sections and associated statistical errors calculated. The computer time required to calculate each pair of charge exchange and ionization cross sections was approximately 0.4 min on a CDC 7600 computer.

RESULTS

Potential curves for a number of the molecular states for the C^{+6}-H collision system were computed as discussed and the relevant curves are given in Fig. 1. Each state is labeled by its set of united atom quantum numbers $n\,l\,m$. The initial system corresponding to H($1s$) and C^{+6} at infinite separation correlates to a $6h\sigma$ state. However, a pseudocrossing exists at about $R = 16$ between the $6h\sigma$ and the $5g\sigma$ states which is "diabatic" in the velocity range considered here. Hence, we formulate a model in which we represent the initial state by a diabatic curve obtained by smoothly joining the $6h\sigma$ to the $5g\sigma$ curves through the avoided crossing.

In the spirit of this model, the calculations are made as follows. The integration of the coupled equations (4) begins and ends at $R = 14.5$ just within the outer pseudocrossing. We set $a_{5g\sigma} = 1$ as the approximate initial condition assuming this is equivalent to $a_{6h\sigma} = 1$ as $t \to -\infty$ because of the diabatic crossing at $R \approx 16$. Similarly, upon termination of the integration, $a_{5g\sigma}$ is regarded as the probability amplitude corresponding to the electron remaining with the proton. The choice of the origin on the proton seems appropriate here since just within the outer crossing, the $5g\sigma$ state has strong H($1s$) character.

In the previous Landau-Zener study, where only radial couplings were considered, the dominant transition was found to be associated with a pseudocrossing between the $5g\sigma$ and $4f\sigma$ potential curves close to $R = 8.0$. However, as shown in Fig. 1, over a considerable range of R, the $5g\sigma$ curve is close to the $4p\pi$, $4d\pi$, and $4f\pi$ curves suggesting the possibility of significant rotational coupling to these π levels. Other close-lying states such as the $5g\pi$ level may also have some influence on the overall electron-transfer process and so a systematic investigation was made to study the effects of these various couplings.

The first step involved the computation of the coupling matrix elements corresponding to transitions between the various states which might be involved. From an examination of these quantities, it became clear that the radial coupling between the $5g\sigma$ and $4f\sigma$ states was indeed very important, and that the rotational coupling between the $5g\sigma$ and $4f\pi$ and $4f\sigma$ and $4f\pi$ levels could be expected to have a significant effect. Moreover, couplings involving the $4d\pi$, $4p\pi$, and $5g\pi$ states could not be discounted. The most important matrix elements (anticipating the results of the coupled-state calculations which are discussed further on) are presented (solid line curves) in Fig. 2. Note the significant maximum in the radial coupling matrix element curve at the position corresponding to the pseudocrossing at $R \approx 8$.

Three sets of calculations of the total cross sections were then made using progressively larger

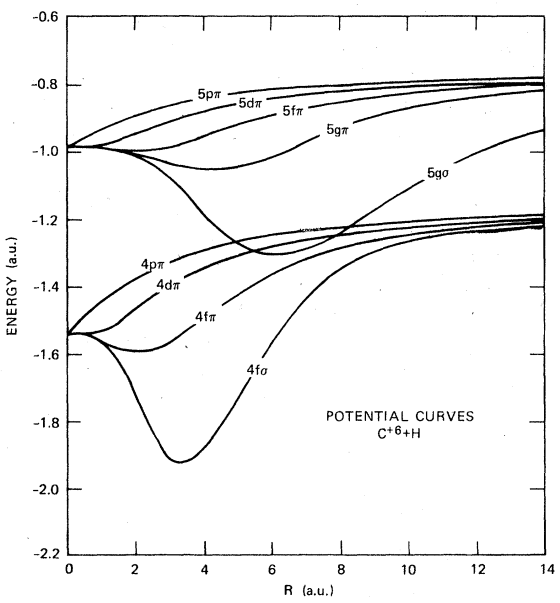

FIG. 1. Relevant adiabatic potential curves for the C^{+6}-H collision system. Each curve is labeled by its associated united atom quantum numbers. The nuclear repulsion energies have not been included in these curves.

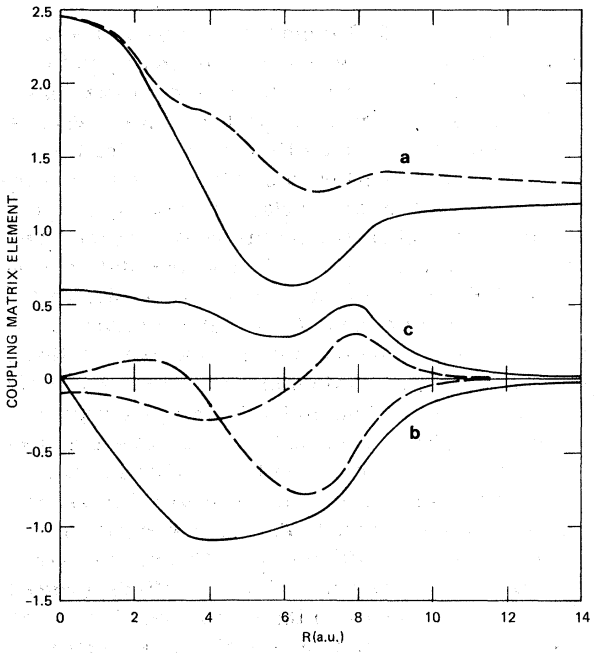

FIG. 2. The three important rotational and radial coupling matrix elements for the $C^{+6}+H$ collision system, computed both with origin of coordinates on proton (solid lines) and on C^{+6} projectile (dashed lines). (a) $\langle 4f\pi|iL_y|4f\sigma\rangle$; (b) $\langle 4f\pi|iL_y|5g\sigma\rangle$; (c) $\langle 4f\sigma|\partial/\partial R|5g\sigma\rangle$.

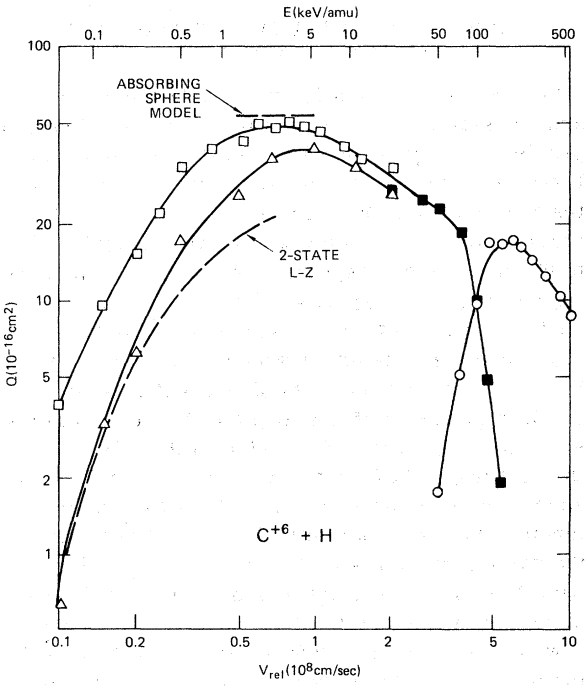

FIG. 3. Calculated electron removal cross sections for the C^{+6}-H collision system. Two-state close-coupling charge-exchange values Δ; six-state close-coupling charge-exchange values □; Monte Carlo charge-exchange values ■; Monte Carlo impact ionization values ○. The lines drawn in for each set of points are intended to guide the eye. The Landau-Zener (Ref. 3) and absorbing sphere (Ref. 4) results are indicated in the figure.

basis sets. The first was a two-state calculation involving only the radially coupled $5g\sigma$ and $4f\sigma$ states. The second was a three-state calculation which included these two and also the $4f\pi$ state. The last and most comprehensive calculation was a six-state calculation which involved all of the previous plus the $5g\pi$, $4d\pi$, and $4p\pi$ states.

The results of the two-state and six-state calculations are shown in Fig. 3 where they are compared with the previous multistate Landau-Zener and absorbing sphere calculations. (In the case of the $C^{+6}-H$ system, the Landau-Zener calculation actually involved only two states.) The three-state calculations which are not included in the figure were in close agreement with the six-state results (within ±10%) showing that coupling to the $5g\pi$, $4d\pi$ and $4p\pi$ states had negligible effect on the total cross sections.

The two-state cross sections are in reasonable agreement with the Landau-Zener results in the threshold region, but appear to reach a peak value 60% to 80% higher than that projected from the Landau-Zener values.[3,18] The inclusion of rotational coupling to the $4f\pi$ state clearly has (in the six- or three-state calculations) a very important influence on the cross sections giving results in the 10^7 cm/sec velocity region which are 1.5 to 3 times larger than the two-state calculations and with a slight shift to lower velocities of the peak cross section. It is interesting to note that the absorbing sphere model[4] gives results in its region of validity $v \sim 7\times10^7$ cm/sec which agree well with the six-state calculations. This is not surprising when one considers that the conceptually simple absorbing sphere method implicitly includes the effects of rotational coupling in its formulation.

Classical-trajectory cross sections for ionization and charge transfer were calculated in the higher velocity range $[(2-10)\times10^8$ cm/sec] and are also plotted in Fig. 3. The Monte Carlo charge-transfer cross sections join quite smoothly to the molecular results in the intermediate region and the overall curve gives a comprehensive picture of the electron-loss process over the complete range $(0.1-10)\times10^8$ cm/sec. Clearly, electron capture of the H-atom electron dominates the electron removal process in the lower velocity region up to about 4×10^8 cm/sec, reaching a peak value of close to 5×10^{-15} cm^2 at a velocity $\sim0.7\times10^8$ cm/sec. The impact-ionization process becomes appreciable at about 3×10^8 cm/sec and becomes dominant at velocities above 4×10^8 cm/sec.

ORIGIN DEPENDENCE OF COUPLED-STATE CALCULATIONS

Because of the diabatic crossing between the $6h\sigma$ and $5g\sigma$ states at $R=16$, it was found, as discussed previously, that the collision system could be analyzed in terms of a three-state expansion involving the $4f\sigma$ and $4f\pi$ states and an initial state which corresponds to $5g\sigma$ below the $R=16$ crossing and to $6h\sigma$ above the crossing. The location of the origin on the proton appears to be a reasonable choice in these total cross-section calculations in light of the arguments advanced by Piacentini and Salin.[6] Nevertheless, it is of interest to study the origin dependence of the coupled-state calculations since this dependence is related to the failure of the perturbed stationary-state method to take into account momentum transfer. This was done by making two additional sets of computations using the same three-state molecular expansion, but with different coordinate origins. In one set, the origin was placed on the C^{+6} ion, and in the other, it was located on the center-of-mass of the colliding particles. The matrix elements computed in the former case are plotted as the dashed curves in Fig. 2.

The cross sections obtained with the origin located on the ion Q_{II} and on the center of mass Q_{III} are compared in Table I with the corresponding values Q_I obtained in the initial calculations. The Q_{II} and Q_{III} values are in general lower than Q_I and the differences increase significantly with increase in relative velocity. One should perhaps view these deviations as an indication of the uncertainties in the calculated cross sections which seem to be on the order of 30%.

SUMMARY AND CONCLUDING REMARKS

We have calculated electron-removal cross sections (electron capture and impact ionization) for C^{+6}-H collisions in the range of relative velocities $(0.1-10)\times10^8$ cm/sec using two fundamentally different approaches. Below 2×10^8 cm/sec, we have used a semiclassical close-coupling method with a basis set of one-electron two-center molecular orbitals to determine the electron-capture cross sections. Above 2×10^8 cm/sec, a classical-trajectory Monte Carlo technique was employed to determine both electron-capture and impact-ionization cross sections. (Impact ionization appears to be negligible below 2×10^8 cm/sec.) The two sets of charge-transfer calculations are in reasonable agreement in the overlapping velocity range giving a comprehensive picture of the overall electron removal processes. The coupled-state calculations indicate that a three-state molecular expansion ($5g\sigma$, $4f\sigma$, and $4f\pi$) is sufficient for characterizing the collision system, and that rotational coupling to the $4f\pi$ state significantly increases the capture cross sections in the 10^7 cm/sec region of velocities. Thus, the coupled-state determinations are a factor of two or three larger than the previous Landau-Zener results, although they are in excellent agreement with the absorbing sphere model values at 7×10^7 cm/sec. The capture process is shown to be very selective with the product C^{+5} ion left primarily in the $n=4$ level. In general, charge transfer dominates the electron removal processes below about 4×10^8 cm/sec with impact ionization becoming most important at the higher velocities.

At the present time, there are no experimental data with which to compare these calculations. The only other theoretical values are the charge-transfer cross sections of Bottcher[5] which are in significant disagreement with our results with respect to magnitude, velocity dependence, and the presence of oscillations. It is noted that Bottcher's theory does not take into account rotational coupling which appears to have an important influence in these collisions. It is surprising, however, that for those calculations in the present study where only radial coupling is considered such as the two-state Landau-Zener or the two-state close-coupling curves shown in Fig. 3, the disagreement with Bottcher's theory is still so significant. The reason for such a discrepancy is not understood.

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TABLE I. Comparison of three-state C^{+6}-H charge-transfer cross sections computed with differing coordinate origins.

v_{rel} (10^7 cm/sec)	Q_I (origin on H) (10^{-16} cm^2)	Q_{II} (origin on C) (10^{-16} cm^2)	Q_{III} (origin on c.m.) (10^{-16} cm^2)
2	15.2	13.9	14.9
5	43.2	34.9	36.0
10	46.3	34.3	36.0
20	34.3	22.3	25.1

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