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Orientation-dependent atomic model for electron transfer in ion-molecule collisions: Applications to $H^+ + H_2$ and $He^{2+} + H_2$

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Single-electron-capture cross sections in collisions of protons and α particles with ground-state H_2 molecules have been studied for projectile energies in the range of 1–500 keV/amu. By treating the ground-state H_2 wave function as a linear combination of atomic orbitals from the two atoms, the electron-capture amplitude in ion-molecule collisions is expressed as the coherent superposition of two single-electron-capture amplitudes in *ion-atom* collisions with the relative phase between the two amplitudes dependent on the collision velocity and the orientation of the molecule. The dependence of the capture cross sections with respect to the molecular orientation was examined. Total electron-capture cross sections averaged over the molecular orientations were obtained and found to be in good agreement with experiments.

I. INTRODUCTION

Electron capture from atoms in collisions with ions have been extensively studied both theoretically and experimentally in the last few decades. For simple target atoms with one or two active electrons, many theoretical models and elaborate computational codes have been developed in the last few years. It is now possible to predict accurate partial and total electron-capture cross sections from these theoretical calculations. From the good agreement between the few theoretical and experimental studies of the orientation and alignment parameters, as well as other multipole moments, one can argue that the scattering amplitudes from such calculations are also quite reliable.

While there have been many theoretical calculations in ion-atom collisions, rigorous theoretical models in ion-molecule collisions are very rare, particularly in the low- and intermediate-energy region. While one may argue that the theoretical models developed for ion-atom collisions, such as the atomic-orbital (AO) and the molecular-orbital (MO) expansion models, can be generalized straightforwardly to study ion-molecule collisions, in reality this has not been done except for the exploratory calculations of Kimura[1,2] using the MO expansion for H^+ and Ar^+ on H_2. The lack of calculations is primarily due to computational complexities. On the one hand, one has to treat the two-center or many-center target molecular wave functions, which makes the calculations of basis functions needed for an AO- or MO-based calculations quite large. On the other hand, one also needs to consider the effects of different molecular orientations on the electron-capture cross sections. Since almost all existing experiments for ion-molecular collisions do not explore such orientation effects, any calculations for cross sections in ion-molecule collisions have to be averaged over the molecular orientations in order to compare with experiments.

Experimental investigations of the orientation effects in ion-molecule collisions are currently underway.[3] Since the molecular ions after the collision often end up in a repulsive electronic state, one can obtain the orientation of the molecule by measuring one of the fragments from the Coulomb explosion of the molecular ion in coincidence with the scattered projectile. The sensitivity of electron-capture cross sections with respect to the orientation of the target molecule is expected to depend on the collision systems as well as collision velocities. It is thus desirable to explore such effects with a reasonably simple theoretical model to make predictions where such orientation effects are most likely to be observed. We remark that future improvement of this model is possible as will be outlined better.

Calculations of electron-capture cross sections for ion-molecule collisions under different approximations have been carried out by few authors. In the keV/amu energy region, models[4,5] based on the AO-expansion method have been employed. However, as drastic approximations were adopted in the evaluation of matrix elements, hence the results are either in poor accord[4] or only in qualitative agreement[5] with the experimental data. The most compete calculations in this energy region were carried out by Kimura[1] for $H^+ + H_2$ collisions in the energy range 0.2–20 keV. A semiclassical close-coupling model with an expansion in molecular states was used. The two lowest potential-energy surfaces of the $H_3{}^+$ ion for different orientation of the H_2 molecular with respect to the incident beam were calculated using the so-called diatoms-in-molecules[6,7] (DIM) method. [This method is similar to the linear combination of atomic orbitals (LCAO) for diatomic systems and is useful when one can treat $H_3{}^+$ as a composite system of either $H^+ + H_2$ or of $H + H_2{}^+$.] The electron-capture cross section was then obtained by solving the two-state coupled equations. His calculations allow the study of the effect of the molecular orientation and provide, for the first time, reliable theoretical results for ion-molecule collisions.

At high collision energies ($E > 100$ keV), charge transfer in $H^+ + H_2$ was investigated by Tuan and Gerjuoy[8] employing the Brinkman and Kramers[9] (BK) approximation to calculate the total single-electron-capture cross section for $H^+ + H_2$. They also studied the ratio of the capture cross section from H_2 to that from H as a function of collision energies and came to the conclusion that, in general, the hydrogen molecule cannot be considered as made of two *independent* hydrogen atoms as far as the collision cross sections are considered. Recently, Deb *et al.*[10] reexamined the model of Tuan and Gerjouy and calculated the differential cross section for a fixed molecular orientation. They found pronounced oscillations in the angular distributions for each fixed molecular orientation, which can be attributed to the interference of scattering amplitudes from the two atomic centers. Similar calculations based on a different version of the Born approximation were carried out by Band[11] and by Ray and Saha.[12]

In the treatment of Tuan and Gerjouy and of Deb *et al.*,[10] the charge-transfer amplitude was calculated in the full wave treatment of the heavy-particle motion and the simple first-order Born approximation was used to calculate the scattering amplitude. However, it is already well established that the BK approximation predicts an incorrect charge-transfer cross section for ion-atom collisions. Hence, the conclusion drawn from these calculations for ion-molecule collisions can be regarded as qualitative only. In view of these results, we propose the present model which is basically similar to the model of Tuan and Gerjuoy except in two aspects. (1) We formulate the scattering amplitudes in the impact-parameter approximation. (2) We approximate the ion-atom scattering amplitude from the elaborate calculations based on the AO-expansion method. Since the coupled-channel semiclassical model for ion-atom collisions using the travelling AO-expansion method is known to give reliable results, we hope that the results for collisions predicted using the present model provide a direct test of the validity of treating ion-molecule collisions as a simple extension of ion-atom collisions.

The rest of this paper is arranged as follows. In Sec. II we describe the derivation of our model in the impact-parameter approximation. The model is applied to $H^+ + H_2$ and $He^{2+} + H_2$ collision in Sec. III where the dependence of charge-transfer cross section on the orientation of the molecule is studied. The computed total capture cross sections averaged over the molecular orientation are then compared with the experimental data. It is shown that the present simple model does provide an adequate description of the existing data for the two systems studied. A discussion of the applicability and the limitation of this model and its further extension is given in Sec. IV.

II. THE THEORETICAL MODEL

Consider the collision of a projectile ion impinging on a diatomic molecule with the geometry shown in Fig. 1. The origin of the coordinate system is taken at the center C of the molecule AB. We assume only one active electron which has coordinates $\mathbf{r}_A$ ($\mathbf{r}_B$) with respect to the nucleus A (B) and $\mathbf{r}_P$ with respect to the projectile P. The electronic wave function in the initial state is written as

$$\Psi_i = \psi_i(\mathbf{r})\exp(-i\varepsilon_i t) , \tag{1a}$$

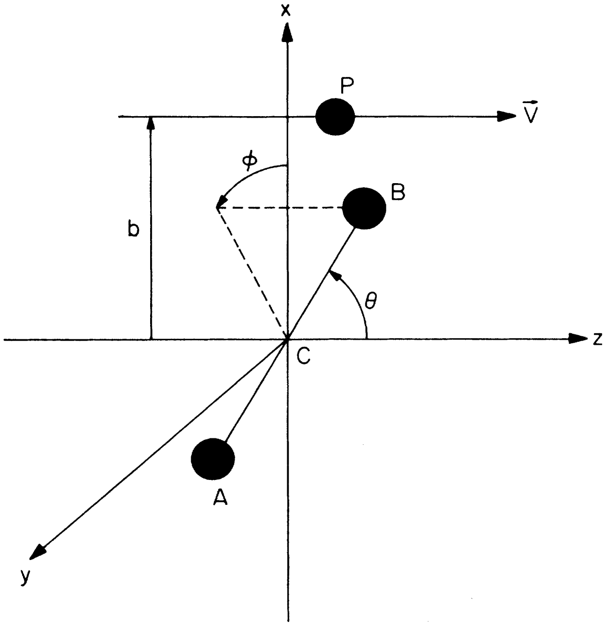

FIG. 1. Coordinate system in the laboratory frame for the ion-H_2 system.

where $\mathbf{r}$ is the position of the electron with respect to the origin. Since the vibrational motion of the molecule is much slower than the collision speed, we approximate $\psi_i(\mathbf{r})$ to be the electronic wave function at the equilibrium internuclear separation ρ ($=1.4$ a.u. for H_2) which stays fixed during the collision. In (1a), ε_i is the electronic binding energy. For the final state, the electron is moving with the projectile and the wave function is given by

$$\Psi_f = \psi_f(\mathbf{r}_P)\exp(i\mathbf{v}\cdot\mathbf{r} - iv^2 t/2 - i\varepsilon_f t) , \tag{1b}$$

where ψ_f is the electronic wave function centered at the projectile and ε_f is the corresponding binding energy. The v-dependent plane-wave translational phase factor in (1b) accounts for the motion of the projectile with respect to the chosen origin. We use atomic units throughout this article unless otherwise stated.

We first consider the electron-capture amplitude a_{fi} in the perturbation approximation

$$a_{fi} = \langle \Psi_f | V_P | \Psi_i \rangle , \tag{2}$$

where V_P is the electron-projectile interaction. A rigorous evaluation of the matrix element (2) is to use accurate molecular wave function for Ψ_i and perform the necessary integration. In a simplified model we assume that the molecular wave function at the equilibrium distance $\rho = 1.4$ a.u. can be approximated as

$$\Psi_i = [\phi(\mathbf{r}_A) + \phi(\mathbf{r}_B)]/\sqrt{2} , \tag{3}$$

where $\phi(\mathbf{r}_j)$ is the atomic orbital placed at $j = A$ or B. In (3) we neglect the small correction due to the overlap of the two atomic orbitals from the two centers. Substituting (3) into (2), we can write the scattering amplitude as

$$a_{fi}=\frac{1}{\sqrt{2}}\left[\int d\mathbf{r}\,dt V_P\psi_f(\mathbf{r}_P)\phi(r_A)\times\exp(-i\mathbf{v}\cdot\mathbf{r}+i\omega t+iv^2t/2)+\int d\mathbf{r}\,dt V_P\psi_f(\mathbf{r}_P)\phi(\mathbf{r}_B)\times\exp(-i\mathbf{v}\cdot\mathbf{r}+i\omega t+iv^2t/2)\right], \tag{4}$$

where $\omega=\varepsilon_f-\varepsilon_i$. The quantities above are defined with respect to the origin at C. We note that the first integral is identical to the electron-capture amplitude in the perturbation theory for the projectile colliding with *atom A* except for a constant phase factor, and the second integral is also similar to the amplitude for electron capture from *atom B* except for a constant phase factor. By shifting the origin of the coordinates from C to A, the first integral can be written as an *ion-atom* electron-capture amplitude multiplied by a phase factor which depends on the orientation of the molecule. To find this phase factor, we assume that the time integration with respect to C is from $-T$ to $+T$ where $t=0$ is taken to be the position of the projectile at the distance of closest approach (we assume a straightline trajectory) with respect to C. By shifting the integration from $d\mathbf{r}$ to $d\mathbf{r}_A$, the first integral can be written as

$$\int_{-T}^{T}dt\int d\mathbf{r}_A V_P\psi_f(\mathbf{r}_P)\phi(\mathbf{r}_A)\times\exp[-i\mathbf{v}\cdot(\mathbf{r}_A-\boldsymbol{\rho}/2)+i(\omega+v^2/2)t]\ . \tag{5}$$

We next transform the time integration so that $t'=0$ when the projectile is at the distance of closest approach with respect to center A. The time τ for the wave front of the projectile to travel from A to C is $\tau=(\rho\cos\theta)/(2v)$ where θ is the angle of the molecular axis with respect to the beam direction. Such a shift results in an additional phase factor $\exp[-i(\omega+v^2/2)\tau]$. Thus, the overall phase shift due to the change of origin from C to A is $\exp[iv\rho\cos\theta/2-i(\omega+v^2/2)\tau]$. A similar argument can be applied to the second integral in (4). Retaining only the relative phase, Eq. (4) can then be written as

$$a_{fi}=\frac{1}{\sqrt{2}}\{A(\mathbf{b}_1)+A(\mathbf{b}_2)\exp[-i\rho\cos\theta(v/2-\omega/v)]\}\ , \tag{6}$$

where $\mathbf{b}_1$ and $\mathbf{b}_2$ are the impact parameters with respect to atom A and atom B, respectively. Thus, we obtain the electron-capture amplitude a_{fi} in ion-molecule collisions in terms of the electron-capture amplitudes $A(\mathbf{b})$ in ion-atom collisions. The relative phase between the two amplitudes depend on the collision velocity v, the orientation angle θ of the molecule, and the energy defect ω. In (6), we assume that the two atoms are identical, but this derivation can be easily generalized to heternuclear molecules as well as to polyatomic molecules. We remark that the result (6) can also be obtained from the wave treatment of Tuan and Gerjuoy by appling the eikonal transformation from the scattering angles of the projectile to impact parameters.[13]

Equation (6) was derived from the first-order perturbation model. However, the expression depends weakly on the definition of the scattering amplitude $A(\mathbf{b})$. We thus *generalize* Eq. (6) to include situations in which the electron-capture probability can no longer be treated perturbatively. In this case we use the $A(\mathbf{b})$ scattering amplitude computed employing the semiclassical close-coupling model for ion-atom collisions employing an expansion in traveling atomic orbitals. For example, to calculate charge-transfer cross sections in H^+ on H_2, we need only to calculate charge-transfer amplitudes $A(\mathbf{b})$ for $\mathrm{H}^++\mathrm{H}$ collisions using the close-coupling method. The results for different molecular orientations are obtained through the phase factor in (6). In this way, the charge-transfer amplitudes in ion-molecule collisions are treated as a coherent sum of charge-transfer amplitudes in ion-atom collisions with the relative phase depending on the molecular orientation and the collision velocity.

An inherent limitation of the present simple model which was derived using the perturbation theory is that the probability for ion-molecule collisions calculated using Eq. (6)

$$P(\mathbf{b})=|a_{fi}|^2 \tag{7a}$$

can become greater than unity when the charge-transfer probability $|A(\mathbf{b}_1)|^2$ [or $|A(\mathbf{b}_2)|^2$] for ion-atom collisions is close to unity. To extend the present model to the energy region where this happens, we follow the "standard" procedure by using a unitarized approximation.[14] This is done by calculating the charge-transfer probability using

$$P_u(\mathbf{b})=\sin^2\sqrt{P(\mathbf{b})}\ . \tag{7b}$$

This *ansatz* guarantees that the charge-transfer probability $P_u(\mathbf{b})$ never exceeds unity and when the probability is small, $P_u=P$. In a more elaborate calculation this limitation of the theory can be removed, as discussed in Sec. IV.

This simple model itself does not give information about the final states of the molecular ion after electron capture directly. However, it assumes that the molecule is stationary (with the given orientation) during the charge-transfer collision. The molecular ion does not relax until the charge-transfer collision is over. The distribution of the electronic and vibrational excited states of the final molecular ion is determined by the relaxation of the molecular ion from the equilibrium distance of the neutral molecule to that for the molecular ion. If the relaxation results in the breakup of the molecular ions into fragments, measurements of such fragments give direct information about the orientation of the molecule involved in the collision.

In ion-molecule collisions the impact-parameter plane no longer has cylindrical symmetry, thus the calculation of cross sections involves an integration over the impact-parameter *plane* for each molecular orientation. We choose the beam axis to be along the z direction. The impact-parameter plane is defined to be the xy plane with

$$\mathbf{b}=(b\cos\alpha,b\sin\alpha,0)\ . \tag{8a}$$

Let θ and ϕ be the azimuthal angles of the molecule, then the position of the atom A is

$$R_A=(x_A,y_A,z_A)=-\frac{\rho}{2}(\sin\theta\cos\phi,\sin\theta\sin\phi,\cos\theta) \tag{8b}$$

and $R_B = -R_A$. This relation allows us to calculate b_1 and b_2, i.e., the impact parameters needed for atoms A and B, respectively,

$$b_1^2 = (b\cos\alpha - x_A)^2 + (b\sin\alpha - y_A)^2 , \tag{9a}$$

$$b_2^2 = (b\cos\alpha + x_A)^2 + (b\sin\alpha + y_A)^2 . \tag{9b}$$

The electron-capture probability at a given b integrated over the angle α for each oriented molecule is

$$P'(b) = \int_0^{2\pi} P(b,\alpha)\,d\alpha , \tag{10}$$

where $P(b,\alpha)$ is obtained from (7a) or (7b). The total cross section for a given molecular orientation is then obtained from

$$\sigma(\theta,\phi) = \int_0^{\infty} bP'(b)\,db . \tag{11}$$

Finally the cross sections averaged over all the orientations is obtained from

$$\bar{\sigma} = \frac{1}{4\pi}\int \sigma(\theta,\phi)\,d\Omega . \tag{12}$$

III. RESULTS AND DISCUSSION

The calculations of scattering amplitudes in ion-atom collisions using the AO expansion have been described in the literature.[15,16] For $H^+ + H$ and $He^{2+} + H$ collisions, many elaborate calculations have been carried out. For the present purpose, it is not essential to do a large-scale calculation since electron capture for the former system is primarily to the ground state and for the latter is mostly to the $n=2$ states at lower energies and to the $n=1$ state for energies greater than 200 keV/amu. Within the present simple model, we thus need only to do a two-state AO-expansion calculation for $H^+ + H$ by including the 1s orbitals on each center (an effective charge of 1.09 was placed on the target atom), and for $He^{2+} + H$ we need only to include the 1s state of H and $n=1$ and $n=2$ states of He^+. Such calculations are adequate except at the higher energies where ionization becomes important. We use the electron-capture amplitudes from these coupled-channel calculations in Eq. (6) to obtain single-electron-capture amplitudes for $H^+ + H_2$ and $He^{2+} + H_2$ collisions. To compare with experiments, we employ the independent electron model where the probability for single capture $P = P(b,\alpha)$ in Eq. (10) is replaced by $2P(1-P)$ before integration over α and over b is carried out.

A. The ϕ dependence

We first discuss the impact-parameter dependence of the capture probability. Assuming the incident beam to be on the x-z plane ($\alpha = 0°$), in Fig. 2(a) we show $bP(b)$ versus b at different values of ϕ at a fixed $\theta = 30°$ for collision energy at 10 keV. We note that $P(b)$ depends very weakly on ϕ. The $bP(b)$ shows a peak at $b \cong 3$ a.u. As the collision energy increases, for example, at 100 keV as shown in Fig. 2(b), the b dependence still appears to be similar except that the peak moves inward to about 1 a.u. This weak dependence on ϕ is also reflected in the integrated cross sections. In Table I we show the normalized cross sections (in units of orientation-averaged cross section) at different ϕ angles or a fixed $\theta = 30°$ and impact energies in the range 1–400 keV. It is clear that there is very little ϕ dependence throughout the energy region studied. This conclusion is in harmony with the low-energy (1–20 keV) results of Kimura.

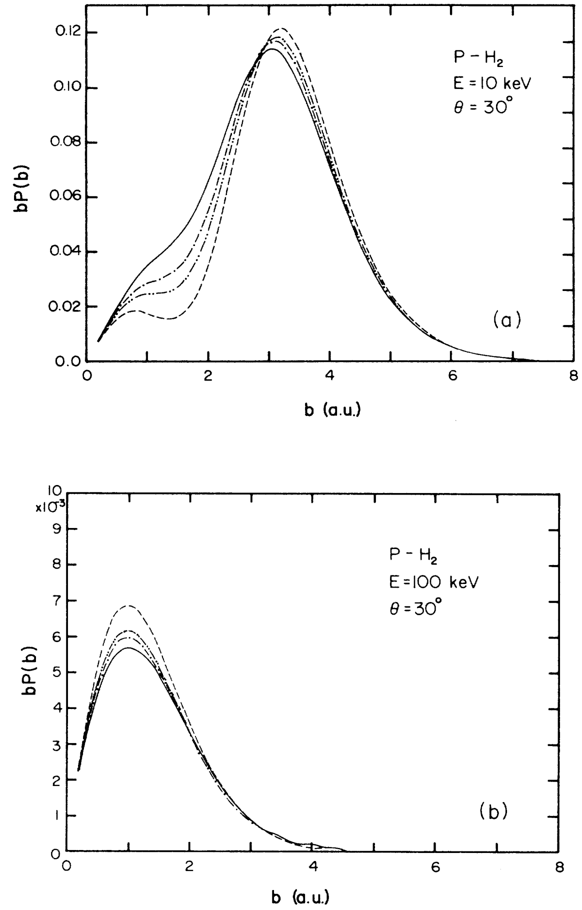

FIG. 2. (a) Probability times impact parameter as a function of impact parameter at 10-keV impact energy, $\theta = 30°$, and $\phi = 0°$ (——), 45° (—·—·—·), 60° (—··—), 90° (— — —). (b) Probability times impact parameter as a function of impact parameter at 100-keV impact energy, $\Theta = 30°$, and $\Phi = 0°$ (——), 45° (—·—·—·), 60° (—··—), 90° (— — —).

B. The θ dependence

We next examine the θ dependence. In Fig. 3(a) we show $bP(b)$ versus b for a fixed $\phi = 0°$ at various angles θ for $E = 10$ keV. At this energy we see small variations with θ angles. Kimura has investigated the same dependence at the same energy. He found a stronger θ dependence than our results indicate, although the qualitative behavior is similar. The θ dependence becomes stronger for collision energy at 100 keV, as can be seen in Fig. 3(b). This strong θ dependence is reflected in the integrated total capture cross sections, shown in Table II where the θ dependence at each energy is again referred

TABLE I. The ϕ dependence of the cross-section ratio $\sigma(\theta,\Phi)/\bar{\sigma}$ for electron capture in $H^+ + H_2$ collisions. The angle θ is fixed at 30° and $\bar{\sigma}$ is the averaged total capture cross section.

E_{lab} (keV/amu)	$\phi=0°$	$\phi=30°$	$\sigma(\theta,\phi)/\bar{\sigma}$ $\phi=45°$	$\phi=60°$	$\phi=90°$	$\bar{\sigma}$ (10^{-16} cm^2)
1	0.69	0.77	0.88	1.03	1.25	8.13
10	1.03	1.01	0.99	0.97	0.93	6.78
25	0.93	0.93	0.93	0.94	0.95	4.33
50	0.80	0.81	0.83	0.85	0.90	1.89
75	0.72	0.73	0.74	0.77	0.83	0.78
100	0.69	0.69	0.69	0.71	0.77	0.35
125	0.65	0.64	0.65	0.65	0.70	0.17
150	0.60	0.60	0.60	0.60	0.65	0.09
175	0.58	0.56	0.56	0.56	0.60	0.05
200	0.55	0.55	0.55	0.52	0.55	0.029
250	0.55	0.45	0.45	0.45	0.45	0.011
300	0.39	0.39	0.39	0.33	0.39	0.0051
400	0.38	0.31	0.23	0.23	0.15	0.0013

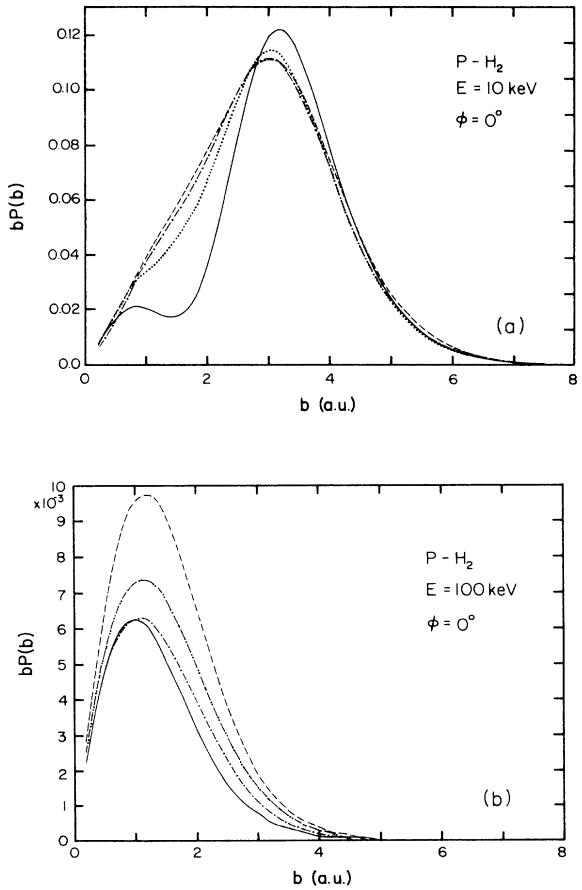

FIG. 3. (a) Probability times impact parameter as a function of impact parameter at 10-keV impact energy, $\Phi=0°$, and $\Theta=0°$ (———), 30° (· · · ·), 45° (—·—·—·), 90° (— — —). (b) Probability times impact parameter as a function of impact parameter at 100-keV impact energy, $\Phi=0°$, and $\Theta=0°$ (———), 45° (—·—·—·), 60°, (–··–), 90° (— — —).

to the orientation averaged total capture cross sections. We also note the strong θ dependence for $E=200-400$ keV, where the electron-capture cross sections from molecules parallel to the beam ($\theta=0°$) is 5–20 times less than from molecules perpendicular to the beam ($\theta=90°$). In the lower-energy region, the effect of molecular orientation is very small. The lower-energy results are in agreement with the conclusion of Kimura.

Our results suggest that the search of molecular orientation effect for the electron-capture process for the $H^+ + H_2$ system should be carried out in the 200–400-keV region. However, this is not a desirable system for experimental studies since the H_2^+ formed after the capture is mostly in the stable $1s\,\sigma_g$ state which cannot be detected by the Coulomb-explosion technique. Nevertheless our results indicate that the orientation effect depends on the collision energies. Work is in progress in studying the orientation effects from N_2 by heavy-ion impact. For example, if the incident ions are bare nuclei of oxygen or flourine, electron capture from the K shell of N_2 is the dominant process. The stablization of the K-shell vacancy by Auger emission is followed most likely by the dissociation of the molecular ion which can be detected using the Coulomb-explosion technique.

C. Total charge-transfer cross sections for H^+ collisions with H_2

Experimental measurements of total charge-transfer cross sections in ion-molecule collisions usually do not provide information about the orientation effect. Thus we compare the experimental data with our orientation-averaged cross sections. The results are shown in Fig. 4, together with the data from different experimental groups adopted from the compilation of Tawara *et al.*[17] Calculations from Kimura[1] below 20 keV are also shown. We note that the results from our calculations are in good agreement with the experimental data[18–25] over the energy region shown. At low energy (below 2 keV) our re-

TABLE II. The θ dependence of the cross-section ratio $\sigma(\theta,\phi)/\bar{\sigma}$ for electron capture in H^+-H_2 collision. The angle ϕ is fixed at 0° and $\bar{\sigma}$ is the averaged total capture cross section.

E_{lab} (keV/amu)	$\sigma(\theta,\phi)/\bar{\sigma}$ $\theta=0°$	$\theta=30°$	$\theta=45°$	$\theta=60°$	$\theta=90°$	$\bar{\sigma}$ (10^{-16} cm^2)
1	1.22	0.69	0.62	0.66	0.72	8.13
10	0.94	1.03	1.07	1.05	1.16	6.78
25	0.95	0.93	0.95	0.98	1.07	4.33
50	0.88	0.80	0.83	0.90	1.07	1.89
75	0.78	0.73	0.79	0.91	1.14	0.78
100	0.71	0.69	0.77	0.94	1.23	0.35
125	0.65	0.65	0.77	1.00	1.29	0.17
150	0.55	0.55	0.78	1.00	1.44	0.09
175	0.40	0.58	0.80	1.00	1.40	0.05
200	0.34	0.55	0.79	1.10	1.72	0.029
250	0.27	0.45	0.82	1.18	1.82	0.011
300	0.18	0.39	0.78	1.18	1.96	0.0051
400	0.05	0.38	0.85	1.38	2.31	0.0013

sults do not decrease with decreasing collision energies as indicated by the MO calculations of Kimura and the experimental data. This could be attributed to the breakdown of the simple model and the unitarized approximation used. A possible improvement of the model is discussed in Sec. IV. Our results in the 10–100-keV region agree quite well with the experimental data. The gradual increasing discrepancy for energies above 100 keV is partly due to the failure of the two-state atomic model for describing electron-capture processes in ion-atom collisions at high energies.

In estimating cross sections for ion-H_2 collisions at higher energies, it is customary to assume that the cross section at a given energy is twice the cross section of ion-H collisions at the same energy. This is equivalent to the assumption of complete random phases in Eq. (6) such that the scattering probabilities from each atom can be added incoherently. Our model provides a direct estimate of the validity of this assumption. In Table III we show the calculated cross sections for orientation-averaged molecular target, atomic target, and the cross-section ratio. We note that this ratio varies with scattering energies, ranging from less than 2 to greater than 2.

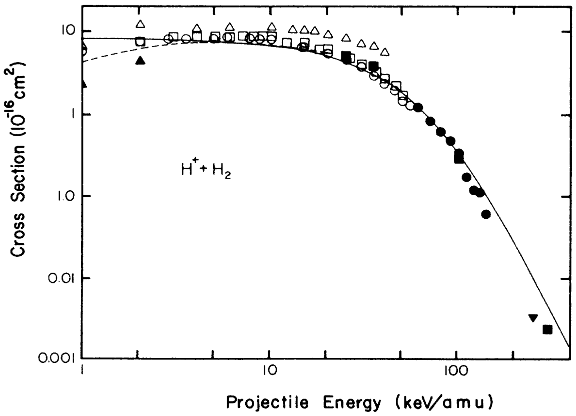

FIG. 4. Single-capture cross section for $H^+ + H_2 \rightarrow H(1s) + H_2^+$. Theory: ——; present, — — —, Ref. 1. Experiment: ▲, Ref. 18; ○, Ref. 20; ●, Ref. 21; △, Ref. 22, ■, Ref. 23; □, Ref. 24; ▼, Ref. 25.

D. Total charge-transfer cross sections for He^{2+} on H_2

We have also studied the total charge-transfer cross sections for the collisions of He^{2+} on H_2 in the 10–400-keV/amu energy region. The total capture cross sections averaged over the molecular orientation are shown in Fig. 5. Note that the agreement between our calculations and the experimental results[23,26] is again quite good except at the higher energies (> 200 keV/amu). In the calculation, capture to the $n=1$ and $n=2$ states are included. In Table IV we compare the total capture cross sections from H_2 and H targets along with their ratio.

IV. SUMMARY

In this paper we show that electron-capture cross sections for ion-molecule collisions can be obtained from the

TABLE III. Capture cross sections $\bar{\sigma}$ and σ^A for $H^+ + H_2$ and $H^+ + H$ collisions, respectively.

E_{lab} (keV/amu)	σ^A (10^{-16} cm^2)	$\bar{\sigma}$ (10^{-16} cm^2)	$\bar{\sigma}/\sigma^A$
1.0	17.18 (14.2[a])	8.13 (4.22[a])	0.47
10.0	8.54 (7.4[a])	6.78 (6.82[a])	0.79
25.0	3.93	4.33	1.10
50.0	0.73	1.89	2.58
100.0	0.12	0.35	2.99
125.0	0.058	0.170	2.95
150.0	0.032	0.090	2.87
175.0	0.018	0.05	2.78
200.0	0.011	0.029	2.68
250.0	0.0046	0.011	2.50
300.0	0.0022	0.0051	2.33
400.0	0.0006	0.0013	2.08

[a] Corresponding cross sections from Kimura (Ref. 1).

TABLE IV. Capture cross sections $\bar{\sigma}$ and σ^A (in units of 10^{-16} cm^2) for $He^{2+}+H_2$ and $He^{2+}+H$ collisions, respectively.

E_{lab} (keV/amu)	σ^A (10^{-16} cm^2)	$\bar{\sigma}$ (10^{-16} cm^2)	$\bar{\sigma}/\sigma^A$
12.5	13.6	16.87	1.2
25.0	9.74	12.17	1.2
50.0	3.48	6.69	1.9
75.0	1.27	3.03	2.4
100.0	0.54	1.36	2.5
150.0	0.13	0.33	2.5
200.0	0.043	0.100	2.3
250.0	0.018	0.040	2.2
325.0	0.0056	0.011	1.96
375.0	0.0029	0.0056	1.91
500.0	0.0008	0.0015	1.88

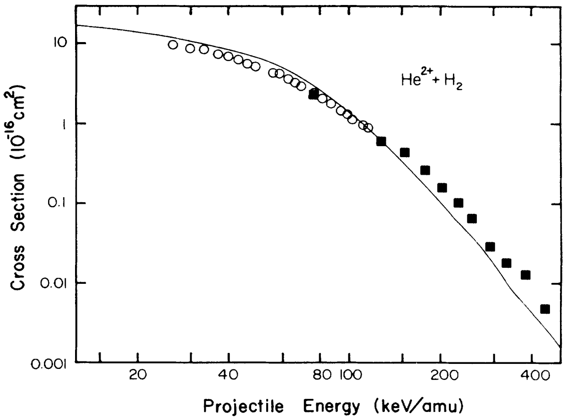

FIG. 5. Single-capture cross section for $He^{2+}+H_2\rightarrow He^+(nl)+H_2^+$. Theory: ——. Experiment (see text): ■, Ref. 23; ○, Ref. 26.

electron-capture amplitudes in ion-atom collisions using the proposed simple model. Basic to the present model is the assumption that the molecular target wave function can be written as the linear combination of simple atomic orbitals and that the charge-transfer amplitude can be written in the first-order perturbation form. Both approximations can be removed in a more detailed calculation.

It appears that generalization of the atomic-orbital expansion method to ion-molecular collisions may not be that difficult computationally. By representing the molecular wave function at the equilibrium internuclear separation in terms of linear combination of atomic orbitals from the two atomic centers of the molecular, the evaluation of all the matrix elements in a coupled-channel calculation for ion-molecular collision can follow the procedure presented in Sec. III. Thus the matrix elements are similar to those encountered in ion-atom collisions and the effect of molecular orientation enters only in the phase factor of the form of Eq. (6). Such problems may be suitable for parallel processors, as are the evaluation of the matrix elements in ion-atom collision, since all the molecular orientations can be calculated at the same time. In such close-coupling calculations, the unitarity of the scattering amplitude is preserved.

The simple model presented here for ion-molecular collisions is useful for checking the validity of the even simpler models used in the literature[27,28] for electron capture from more complex atoms. Total electron-capture cross sections from a range of molecules of the type C_nH_m for $n\leq 4$ and $m\leq 8$ by protons in the energy range of 0.8–3 MeV have been reported by various workers. It was found that in this energy region, the contribution to the capture cross sections from H is very small, but the total capture cross sections per carbon atom is not a constant, but rather decreases with increasing n. It would be interesting to see if these experimental results can be explained by the present simple model.

In summary the present model provides a first step toward the understanding of electron-capture processes in ion-molecular collisions. Without going into detailed complicated calculations, the present model affords the possibility of understanding electron capture from the molecular target and explore its orientation effects. Work is in progress for calculating the orientation-averaged capture cross sections for a number of other ion-molecule collision systems, however, experiments which explore the orientation effect of the molecule are definitely most desirable.

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