ENERGY DEPENDENCE OF THE TOTAL DIFFERENTIAL CROSS SECTIONS FOR H⁺ + Ar AND H⁺ + Kr

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Total differential cross sections, the sum of the differential cross sections for all processes, including charge transfer, were measured for H⁺ + Ar and H⁺ + Kr. The results are compared to two theoretical models, one employing a single, average trajectory and the other based on semi-classical wave analysis. These models give very different results in the rainbow region of the ground state molecular ion potential when the probability of a transition becomes significant. The second method gives better qualitative agreement with the present data.

1. Introduction

The impact parameter method for describing collisions between atomic systems is based on the assumption that for each impact parameter the nuclei follow a single classical trajectory [1]. This allows a great simplification in the description of scattering phenomena and is useful for obtaining transition probabilities. If these trajectories have physical significance then their differential cross section would correspond to the sum of the measured differential cross sections for all processes, the total differential cross section. At high energies Everhart et al. [2] have determined that the total differential cross section can be characterized over a broad range of angle and energy by elastic scattering from a screened Coulomb potential. For proton–rare gas collisions Champion et al. [3] have shown that at very low energies, where only elastic scattering occurs, the differential cross section shows rainbow structure characteristic of the adiabatic ground state curve of the rare-gas–hydride ion. Therefore as one increases the incident ion energy the adiabatic structure is replaced by a diabatic, screened Coulomb differential cross section [4]. This change in the total differential cross section with increasing energy is due to the inability of the electrons to adjust rapidly enough to the nuclear motion, which results in electronic transitions as well as changes in the forces between the colliding systems.

In order to unravel to what extent one can still use a single potential (which may be energy dependent) to describe differential cross sections for collisions which result in several final states we have measured absolute total differential cross sections for H⁺ + Ar and H⁺ + Kr in the energy range 500–5000 eV. In this letter we compare results of these measurements to two simple theoretical descriptions, one based on a single energy-dependent interaction, the other based on a semi-classical wave method. Delos et al. have discussed the regions of validity of such descriptions [5].

2. Experiments

The experimental results discussed here were obtained by scattering a mass-selected proton beam from Ar and Kr target gases and detecting the scattered protons and H atoms separately as a function of scattering angle [6]. The neutral particles were detected

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by an electron multiplier in line with the slits defining the scattering angle, and the ions were detected by a second multiplier after being separated from the neutral particles by electrostatic deflection. In order to obtain absolute differential cross sections it was necessary to determine the absolute sensitivity of both detectors. For the ion detector this was done by comparing the multiplier output to the weakest ion currents measurable with an electrometer. Determining the sensitivity of the neutral particle detector was more difficult and was accomplished by measuring the total (integrated over angle) charge transfer cross section by collecting the slow target ions in the scattering cell, setting this total equal to the integral over angle of the measured relative differential charge transfer cross section determined by the detected neutral particle current. The experimental curves displayed in the figures represent the total differential cross sections (the sum of the ion and neutral cross sections).

The detectors were 15 stage Be-Cu electron multipliers (Dumont SPM03-412) and the angular resolution of the detection system for these measurements was 0.25 degrees. It is estimated that the absolute experimental cross sections reported here are accurate to ±40 percent with a reproducibility of ±15 percent.

3. Models

In $^7\text{Ar}^{11}$ and $^7\text{Kr}^{11}$ the lowest $^1\Sigma$ charge transfer state, which is close in energy to the incoming ground state channel, is the most accessible inelastic state at the collision energies investigated. For these systems the ground state attractive potential wells can be thought of as primarily due to the charge transfer interaction associated with the $^1\Sigma$ states. It appears reasonable to assume, based on available potential curves [7], that this interaction is greater than the energy defect for those internuclear distances, $R$, of importance in the present scattering experiment, roughly 1 to 4 au. Therefore we treat these collisions in the strong-coupling, two state approximation for which the charge transfer probability can be written [8]

$$
\mathcal{P} = 4P(1 - P) \sin^2 \Omega .
$$

Here $P$ and $\Omega$, slowly varying functions of the impact parameter, $b$, can be estimated from knowledge of an interaction potential, $V(R)$, the velocity, $v$, and the energy defect, $\epsilon$. Applying the semi-classical analysis of Ford and Wheeler [9] the strong coupling approximation yields a total differential cross section, here a sum of the elastic and charge transfer cross sections, of the form [10]

$$
\sigma_+(\theta) = [1 - P(\theta)] \sigma_0(\theta) + P(\theta) \sigma_1(\theta) ,
$$

where $\sigma_0(\theta)$ and $\sigma_1(\theta)$ are the elastic scattering cross sections for the two $^1\Sigma$ adiabatic potentials, $V_0(R)$ and $V_1(R)$, and $b_0(\theta)$ and $b_1(\theta)$ give the dependence of impact parameter on scattering angle for these potentials. In eq. (1a) we have dropped small oscillatory terms due to the difference between $P(b_0(\theta))$ and $P(b_1(\theta))$. Further, when one of the elastic scattering cross sections has more than one impact parameter contributing at any angle, each branch should be weighted according to the appropriate $P(b)$.

In the single trajectory description one obtains $\sigma_1(\theta)$ from elastic scattering from an effective potential which depends on the electronic state during the collision [11]. In the strong coupling limit the probability of being in a particular quasi-molecular state during the collision is very nearly constant for intermolecular separations less that the effective "crossing" point, $R_c$, where, according to Stueckelberg [8], $V_0(R_c) \approx \epsilon/2$. In this region, the effective potential for each impact parameter can be well represented by

$$
V(R) = [1 - P(b)] V_0(R) + P(b) V_1(R) .
$$

To estimate the cross sections we use a simple model for the potentials consisting of a common adiabatic term of the form

$$
V_0(R) = Z \epsilon e^{-CR} / R
$$

and a coupling term

$$
V_1(R) = Z \epsilon e^{-CR} / R_0 ,
$$

where $Z$ is the nuclear charge and $C$ is the average of screening constants determined from the ionization potentials of the two neutrals [11]. The parameter $R_0$, corresponding approximately to the zero of the ground state potential, was chosen to reproduce, roughly, the rainbow angles for low energy elastic scattering as found by Champion et al. [3]. For $^7\text{Ar}^{11}$ and $^7\text{Kr}^{11}$ we have chosen $R_0$ to be 1.9 and 2.4 au, respectively, yielding effective "crossing" points, $R_c$, of 5.2 and 7.1 au. At these values of $R_c$ the contribu-
Fig. 1. The probability of being in the excited $1\Sigma$ molecular ion state of ArH$^+$ during a single passage at impact parameter, $b = 2$ a.u. Results were obtained by integrating the two-state impact parameter equations with a straight line trajectory. The internuclear separation is $R = (r^2 + b^2)^{1/2}$ and $R_e$ indicates the effective 'crossing' $V_1(R_e) = \omega/2$.

Fig. 2. The reduced, total differential cross sections for H$^+$ + Ar are plotted versus $\theta_T$; $I$ is the incident ion energy and $\theta$ is the laboratory scattering angle. We compare the present experiments to the theoretical models presented here. $\theta_R$ indicates the rainbow angle for the $1\Sigma$ ground state of ArH$^+$ as found from low energy elastic scattering [3]. Numbers indicate incident ion energies in keV.

rainbow region [9] and elsewhere they were obtained classically.

It is apparent that both models have the correct, general energy dependence. However, they predict quite different results in the rainbow region of the low energy elastic scattering data of Champion et al. [3] indicated by $\theta_R$. For the single trajectory model of eq. (1b) the rainbow effect is shifted continuously to lower values of $\tau$ as one increases ion energy, or increases $P$, whereas eq. (1a) yields damped rainbow structure at approximately the same $\tau$ value for all energies [13]. In going from 0.2 to 5 keV, $P$ goes from about 0.05 to 0.4 for the impact parameters of interest.

For H$^+$ + Kr, where the energy defect is smaller, $P$ ranges from about 0.3 to 0.45 for the values of energy in the range 0.5 to 3 keV. For all of the energies eq. (1b) would yield total differential cross sections with no rainbow structure at the $\tau$ values of interest.

\[ \Delta P(1 - P) = \left| \int_{-\infty}^{\infty} V_1 e^{-i\omega r} \, dr \right|^2 \int_{-\infty}^{\infty} V_1 \, dr, \]

yields values of $P$ in reasonable agreement with the above numerically integrated results except at the lowest energies where $\omega/2 \approx 0$.

In fig. 2, we compare, on reduced cross section plots, $\rho(\neq \tau)$, the experimental data for H$^+$ + Ar with the models based on eqs. (1a) and (1b) respectively, $P$ being obtained from eq. (4). An Airy function was used to describe the differential cross sections in the

* For impact parameters much less than $R_e$, the transition probability, $P$, is relatively insensitive to the choice of trajectory.

† The Rosen-Zener expression for $\rho$ when used in a semi-classical wave analysis as in ref. [10] would also yield $\sigma_T(\theta)$ as in eq. (1a).
Fig. 3. The reduced total differential cross sections for H\(^+\) + Kr are plotted versus \(\theta T\); \(T\) is the incident ion energy and \(\theta\) is the laboratory scattering angle. We compare the present experiments to the two theoretical models: (a) based on eq. (1a) and (b) based on eq. (1b). \(\tau_R\) indicates the rainbow angle for the \(1\Sigma\) ground state of KrI\(^+\) as found from low energy elastic scattering [3].

As can be seen from the results at 0.5 keV in fig. 3, however, the experimental data for this system as given in fig. 3 seem to quite clearly indicate a rainbow effect in the same region as found in the low energy elastic scattering data [3] indicated by \(\tau_R\).

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References

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