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ATOM CAPTURE AND LOSS IN ION MOLECULE COLLISIONS

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Progress is reported in measuring the energy and angular distribution of protons emerging with velocity close to the beam velocity from the target region when Ar^+ beams collide with a CH_4 target and ArH^+ beams collide with a He target at asymptotically high speeds. The protons result from the transfer of a target constituent to the projectile (atom capture) or from the dissociation of the projectile molecule in the collision (atom loss). For atom capture processes the Thomas peak is clearly observed.

1. INTRODUCTION

Ion-atom collisions at asymptotically high speeds involving electronic charge transfer, projectile ionization or target ionization are the subject of many recent theoretical investigations. In the asymptotic velocity regime, where the projectile speed v is substantially greater than the characteristic orbital speed of the electron in a target or projectile atom, a perturbation expansion is supposed to provide an increasingly accurate description of these processes. For charge transfer, interesting ties to classical descriptions exist.

In current electron capture and loss experiments, however, the asymptotic velocity regime can only be reached if the projectiles have velocities $\gtrsim 10$ au even for light targets. Furthermore, angular distributions measurements for electronic charge transfer and projectile ionization processes become increasingly difficult at projectile speeds over 10 au because of the small scattering angles involved.

A simple physical picture of such three-body scattering processes may emerge from alternative experiments, involving the capture of a whole atom from a molecule by a projectile ion or of the loss of an atom from a projectile molecular ion. Projectiles with kinetic energies on the order of 100 eV/u have asymptotically high speeds, substantially greater than the characteristic vibrational speeds of atoms in molecules, while the laboratory scattering angles for atom capture and atom loss events are measurably different from 0 deg. Doubly and triply differential cross section measurements become possible, promising to reveal completely new information about the associated scattering amplitudes.

We are investigating collisions at asymptotically high speeds between singly charged diatomic hydride ions (H_2^+, HeH^+, ArH^+, KrH^+) and neutral atoms (He, Ne, Ar, Kr) and between singly charged atomic projectiles (Ar^+, Kr^+) and neutral target molecules with one or more hydrogen atom constituents (H_2, H_2O, CH_4). We focus on the dissociation of the projectile molecule into states in which the relative velocity of the fragments is small compared to the projectile velocity and on the transfer of target constituents into projectile centered continuum states, i.e. states in the vibrational continuum of the electronic ground state of the projectile hydride ion.

DISCLAIMER

2. ATOM LOSS

In a collision between a diatomic hydride ion and a target atom, low lying states in the vibrational continuum of the projectile molecule can be excited. The molecule dissociates, most often into a neutral fragment and a proton. In the laboratory frame the velocity spectrum of protons produced in such atom loss events peaks at the projectile velocity $\vec{v}$. We have observed this peak (1) by measuring the velocity distribution of protons emerging from the target into a cone of half angle Θ_0 between 0.6 deg and 2.6 deg about the beam direction. However, at asymptotically high speeds excitation to low-lying states in the vibrational continuum of the ground states is not the only mechanism for collisional dissociation. Our spectra show prominent features in the wings of the proton loss peaks at the proton velocity $\vec{v}_p = \vec{v}$. Symmetric structure appears in both wings, growing more intense as the projectile energy increases. We observe peaks corresponding to proton velocities $\vec{v}_s$, such that $|\vec{v}_s - \vec{v}|$ is constant, independent of projectile energy and target gas. The intensities of the features in the wings are, however, strongly dependent on the nature of the target atoms and are most intense for He targets. For ArH^+ projectiles at the higher projectile energies, we observe two well-resolved lines at $|\vec{v}_s - \vec{v}| = (7.2 \pm .3) \cdot 10^{-3}$ au ($1.3 \pm .1$ eV), and $(15.9 \pm .3) \cdot 10^{-3}$ au ($6.3 \pm .3$ eV), respectively; and one weak structure between these lines at $|\vec{v}_s - \vec{v}| = (11.9 \pm .4) \cdot 10^{-3}$ au ($3.5 \pm .3$ eV), as shown in Figure 1. We assume the observed lines

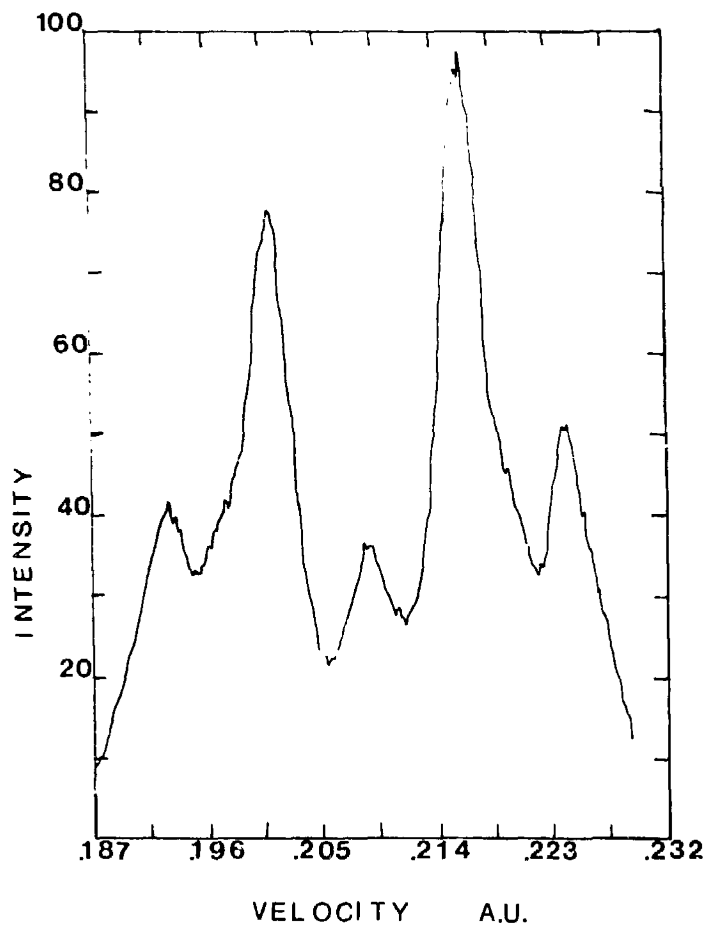


Figure 1. Velocity spectrum of protons emerging into a cone of half angle $\Theta_0 = 1.6$ deg about the beam direction for 44.8 keV ArH^+ ions traversing a He target. The beam velocity is .209 au.

to be due to sudden electronic rearrangements of the molecular projectile in the collision, leaving its nuclear constituents relatively static (Frank-Condon transitions). The projectile undergoes a sudden vertical transition from the electronic ground state to some excited electronic state, which then dissociates, liberating a total CM kinetic energy $U(r) - U(\infty)$, where $U(r)$ is the potential energy corresponding to the internuclear separation r for the particular electronic state excited. The distribution of internuclear separations in the electronic ground state from which the excitation proceeds is of course determined by the populations of the various vibrational states present in the incident beam. In addition to measuring the proton velocity distribution along the beam direction we are now making detailed angular distribution measurements of energy-analyzed protons produced in atom loss events. This will allow us to map the complete velocity distribution of these protons in the projectile rest frame.

3. ATOM CAPTURE

In 1927 Thomas (2) gave a classical treatment of the transfer of a light target constituent to a heavy projectile valid in the asymptotic regime. Thomas scattering is a two step process. The light particle is first scattered by the projectile and then by a heavy target constituent in such a manner that projectile and captured particle emerge with almost the same velocity at a critical angle Θ_c with respect to the incident beam direction. For electron capture by protons from He $\Theta_c = \sqrt{3m/2M} = 0.47$ mrad. (m and M are the electron and proton mass respectively). It is now widely understood that any quantum treatment of capture and high energies must take this process into account (3). In a perturbation expansion, it corresponds to a second order Born process that dominates over the first order Born term in the limit of high projectile velocity. Only recently, the first observation of the Thomas peak in the differential cross section for high energy electron capture by protons from He has been reported (4), and experiments involving heavy ion projectiles would be even more difficult. For atom capture at asymtotically high velocities however, we have observed the Thomas peak at $\Theta_c = 1.3$ deg with Ar^+ projectiles and a CH_4 target (5). In two recent theoretical papers Shakeshaft and Spruch (6,7) have focussed attention on the connection between classical and quantum mechanical scattering descriptions, and atom capture experiments are among the most suitable to reveal such ties.

In our early measurements we have recorded the number of protons emerging into a cone of half angle Θ_0 about the forward direction as a function of proton energy for 100-300 eV/u Ar^+ and Kr^+ projectiles incident on CH_4. States in the vibrational continuum of ArH^+ and KrH^+ dissociate into $Ar + H^+$ and $Kr + H^+$, respectively (8). After a continuum capture event the electron follows the projectile ion and the proton emerges with velocity $v_p \cong v$. If atom capture proceeds via the double scattering mechanism, then the projectile and the captured atom emerge at a critical angle Θ_c. For Ar^+ on CH_4 $\Theta_c = 1.3$ deg and for Kr^+ on CH_4 $\Theta_c = 0.6$ deg. For Ar^+ projectiles we have varied Θ_0 from being smaller to being larger than the critical angle while still collecting protons emerging at all azimuthal angles. For acceptance angles $\Theta_0 > \Theta_c$ we observe a central peak in the proton spectra centered at a proton velocity $v_p = v'$ where v' is close to but slightly less than the beam velocity v. This peak vanishes for $\Theta_0 < \Theta_c$. We interpret this central peak as the atom capture to the continuum peak, produced via the double scattering mechanism. For 100-300 eV/u Ar^+ and Kr^+ projectiles on the CH_4 measured integrated cross sections are small; they lie between 5 and 100 barn.

In earlier experiments Cook et al (9) observed the Thomas peak in the formation of H_2^+ by fast proton impact on CH_4. The position of the Thomas peak can be predicted classically. The shape of the Thomas peak, however, depends on the detailed nature of the interaction. To extract this shape information we are now making the first doubly differential cross section measurements for atom capture to continuum states, differential in captured particle scattering angle and energy.

4. APPARATUS

Measurements differential in proton scattering angle and energy are made using a two dimensional position sensitive particle detection system which is described in detail in this volume (10).

Our beams pass through a 1 cm long target gas cell located at the entrance focus of a doubly focussing spherical sector electrostatic analyzer accepting protons emitted into a cone of half angle $\Theta \leq 6$ deg about the beam direction. A 1 mm aperture in the exit focus of the analyzer sets the energy resolution to $\Delta E/E = 1\%$ FWHM. The position sensitive detector system is mounted 15 cm away from the exit aperture. We record an atom capture to the continuum or atom loss event by detecting a proton emerging from the collision with velocity $\vec{v}_p$ close to the beam velocity $\vec{v}$. We measure the angular and energy distribution of the protons emerging from the collision, i.e., we measure $d^2\sigma/d\Omega_p dE_p$. The angular distribution of the protons with $\vec{v}_p$ close to $\vec{v}$ in the scattering region is imaged one to one into the detection region. The entire distribution in polar angle Θ and azimuthal angle ϕ can be acquired simultaneously. A microchannel electron multiplier array of 25 mm diameter active area is the primary event detector. Output pulses are collected on a resistive anode, and position decoding utilizes the charge division method. Computer assisted data acquisition is implemented using the modular CAMAC standard and a multitasking control program.

5. RESULTS

Measurements using the position sensitive detector system have just begun and only preliminary results can be presented here.

Figure 2 shows the angular distribution of protons emerging from the target region with speed equal to the projectile speed v, for 7 keV Ar^+ projectiles on CH_4. It can clearly be seen that the protons emerge preferentially at a nonzero angle $\Theta \cong 1.4$ deg relative to the beam direction. This measured angle agrees well with $\Theta_c = 1.3$ deg predicted for atom capture via Thomas double scattering. Figure 2 shows raw data, not yet corrected for slight distortions due to the imperfect focussing properties of the 160 deg spherical sector proton energy analyzer. Because atom capture cross sections are so small (on the order of barn) great care must be taken to eliminate background. Here we have not yet been completely successful and the ring structure in the angular distribution, which is the Thomas peak, does not have perfect symmetry. Background problems are now being corrected. The results presented in Figure 2 clearly show that atom capture to the continuum at asymptotically high speeds proceeds via the Thomas double scattering mechanism. To investigate the detailed shape of the Thomas peak, measurements are now being made of the angular distribution of the protons emerging from the target region as a function of proton energy. We will then be able to map the complete velocity distribution of the captured particles. The velocity of a detected proton along the beam direction is determined by the analyzer field and the velocity transverse to the beam direction from the measured position of arrival on the channel plate.

For atom loss processes complete velocity distributions will also be obtained from measurement of proton spectra differential in proton energy and scattering angle.

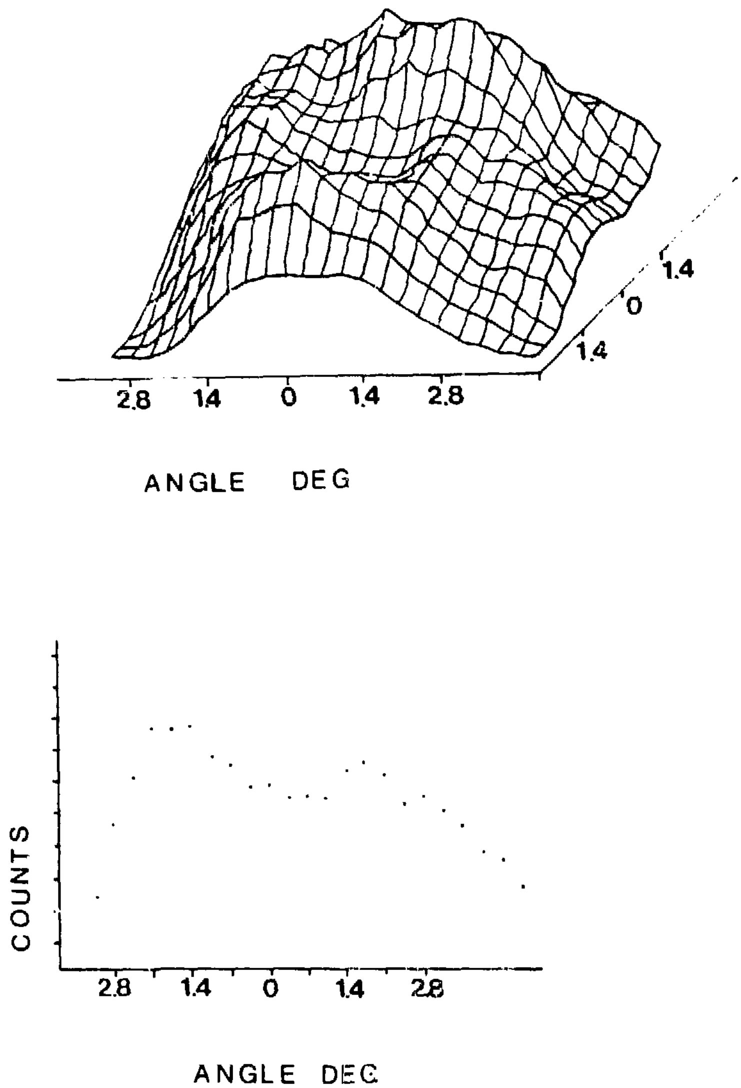
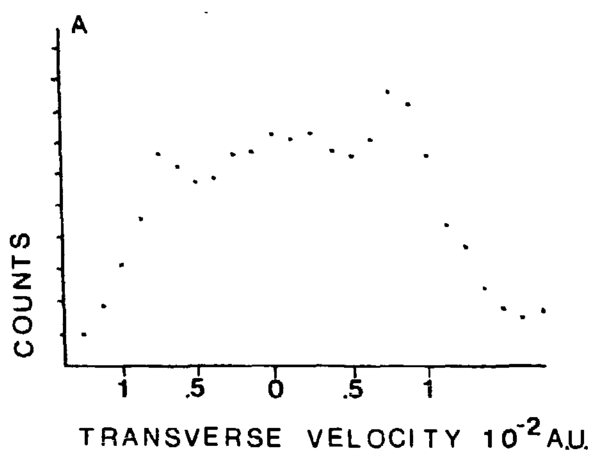


Figure 2. Angular distribution of protons emerging from the target region with speed equal to the projectile speed v for 7 keV Ar^+ projectiles on CH_4. The protons emerge preferentially at a nonzero angle $\Theta \cong 1.4$ deg relative to the beam direction as predicted for continuum capture proceeding via the Thomas double scattering mechanism. The lower graph shows a cut through the channel plate image shown in the upper graph.

Figure 3a shows a cut through a channel plate image measured with 40 keV ArH^+ projectiles on He. The proton velocity along the beam direction equals the projectile velocity v = .1975 au. We observe a peak at zero transverse velocity due to dissociation of ArH^+ after excitation to low lying states in the vibrational continuum of the ground state. Peaks at ~7.5 x 10^{-3} au transverse velocity are due to protons being emitted in the projectile rest frame with this speed transverse to the beam direction.

In Figure 3b the proton velocity along the beam direction equals .1940 au. The central peak has disappeared and the peaks at ~7×10^{-3} au are due to protons being emitted in the projectile rest frame with the speed ~7.5 x 10^{-3} au as above but ~25 deg backwards from the direction transverse to the beam. When angular distributions at proton velocities between v ± .02 au have been measured, a map of the complete velocity distribution in the projectile rest frame can be assembled.



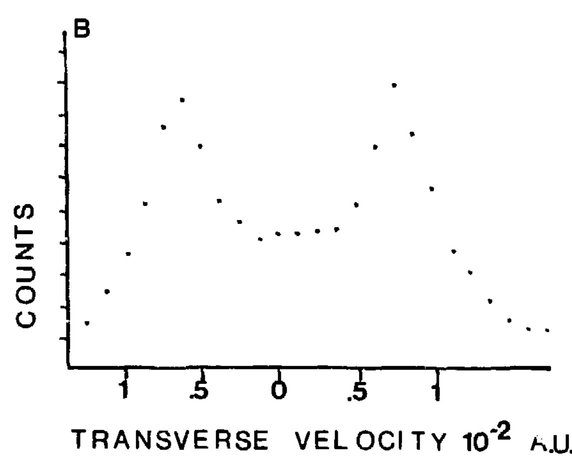


Figure 3. Cuts through channel plate images (angular distributions) measured for 40 keV ArH^+ on He. The protons result from the dissociation of the ArH^+ molecule in the collision. a) The proton velocity equals the beam velocity v = .1975 au b) The proton velocity is .1940 au

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