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## EXPERIMENTAL EVIDENCE FOR LARGE NONADIABATIC EFFECTS IN CLOSE PROTON COLLISIONS WITH HYDROGEN ATOMS*


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Excitation to the 2$s$ state is important in keV-energy $H^+$-H(1$s$) charge-transfer collisions having reduced scattering angles near 3 keV degrees.


The differential charge-transfer cross section data for large-angle $H^+$-H scattering obtained by Everhart and co-workers[1] has been a most sensitive test of theoretical calculations for this fundamental collision system. As their measurements provided no direct information on excited-state production, it was initially hoped that electronic transitions induced by the nuclear motion could be neglected in the calculations. However, the importance of such nonadiabatic effects for close collisions has been predicted by Bates and Williams[2] and re-emphasized more recently by Knudson and Thorson[3] and by Russek and co-workers.[4] These authors have predicted theoretically that dynamic coupling effects can lead to the production of hydrogen atoms with principal quantum number $n = 2$. The present angular-distribution measurements for 2$s$-state production show directly that such nonadiabatic effects are very important in close collisions for some energy range below about 12 keV.

The fractional probability $P(2s)$ for H(2$s$) production in a charge-transfer collision was measured for proton energies between 6 and 30 keV and for laboratory scattering angles out to 0.35 deg. The apparatus used for the measurements has been described in earlier reports of total cross-section measurements for charge transfer into the 2$s$ state.[5,6] Scattered fast metastable atoms were selectively detected by Stark-effect quenching in a dc electric field, followed by observation of the resultant Lyman alpha emission; scattered atoms were also detected without state selection by using secondary electron emission from a metallic surface. In the present study, a small circular aperture was placed in front of the atom detectors and translated with them across the scattered-atom angular distributions. A scan of the detector assembly over the incident proton beam resulted in a beam profile of 0.10 deg full width at half-maximum.

The data obtained are shown in Fig. 1, where $P(2s)$ is plotted versus the laboratory scattering angle $\theta$ for several fixed laboratory energies $E$. The possible fractional error of about ±40% in the values for $P(2s)$ arises principally from uncertainties in atom detection efficiencies. The significant feature in the data is the large rise with scattering angle observed at the lower energies. At higher energies the mechanism responsible for this behavior becomes inoperative, and production of H(2$s$) appears primarily at angles very close to the forward direction. It is only this latter large impact-parameter mechanism that results in a sizeable total H(2$s$) production cross section.[6] The data of Fig. 1 also indicate the existence of a maximum in $P(2s)$ lying near a reduced scattering angle $\tau \equiv E\theta$ of about 3 keV deg. The shape and the exact position of this structure remain to be determined by experiments utilizing higher angular resolution.

The connection[7] between $\tau$ and the impact parameter $b$ requires knowledge of the effective nonadiabatic potential-energy curves involved in the collision processes, quantities not yet available. The limit of pure Coulomb scattering for

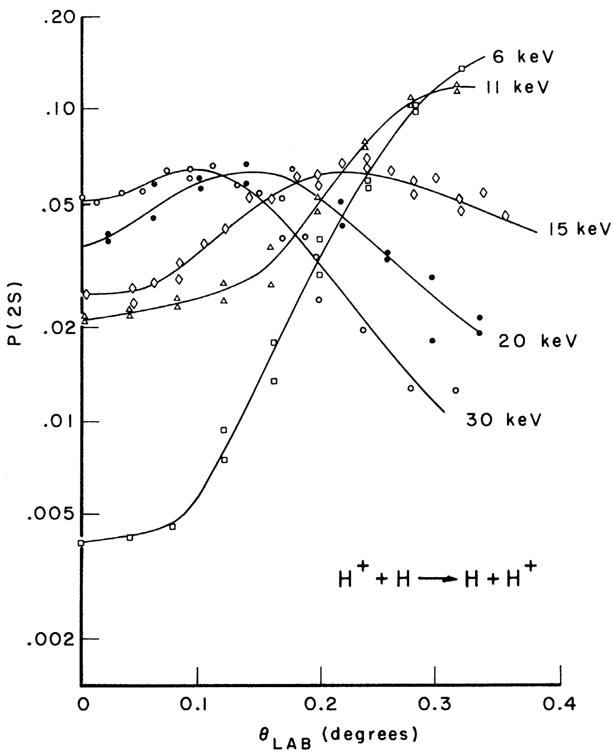


FIG. 1. The measured fractional probability $P(2s)$ for H(2$s$) production in a $H^+$-H(1$s$) charge-transfer collision, as a function of laboratory scattering angle and laboratory incident proton energy. The uncertainty in the vertical scale is about ±40%; the angular resolution is about 0.10 deg full width at half-maximum.

the two protons would yield $b=\tau^{-1}$ in atomic units; this takes on the value 0.5 a.u. for $\tau=3$ keV deg. For the true $H^+$-H collision, electron screening produces a smaller value of $b$ for a given $\theta$. Thus the collisions that produce the observed large values of $P(2s)$ at low energies are for impact parameters less than 0.5 a.u.

Coupled-channel scattering cross section calculations such as those made for $H^+$-H by Wilets and Gallaher[8] can exhibit structure in $P(2s)$ qualitatively similar to that presently observed. Their calculations indicate a maximum in the dependence of $P(2s)$ on impact parameter, reaching a value 0.4 at $b=1$ a.u. for $E=2$ keV, and changing to a value 0.3 at $b=2.3$ a.u. for $E=25$ keV. Thus the present data show maxima at considerably smaller impact parameters than predicted by their four-state theory employing a symmetrized hydrogenic traveling-wave expansion of the collision-system wave function.

Excitation of the state H(2$p$) and its effect on $H^+$-H charge-transfer collisions has been investigated theoretically by Knudson and Thorson.[3] Their calculations employed an eigenfunction expansion in $1s\sigma_g$, $2p\sigma_u$, and $2p\pi_u$ molecular states, a basis set believed adequate for energies below 0.5 keV. A single maximum in the fractional excitation probability $P(2p)$ was predicted for energies from 75 up to 250 eV, the highest energy considered. This maximum was found to lie at reduced scattering angles between 2.2 and 1.3 keV deg. The structure in $P(2s)$ presently observed at higher energies where more molecular states have important coupling matrix elements may well be closely related to that predicted for $P(2p)$ at roughly half the observed reduced scattering angle.

One explicit molecular mechanism for the observed behavior of $P(2s)$ has recently been proposed by Russek and co-workers.[4] They performed numerical calculations for 2-keV $H^+$-H scattering, utilizing wave function expansions in terms of variously truncated molecular basis sets. They found that inclusion of the $3p\sigma_u$ state of $H_2^+$ was important for close collisions, and predicted probabilities for H(2$s$) production as large as $\frac{1}{3}$ for $b=0.1$ a.u. The coupling to this state was shown to be from the $2p\sigma_u$ state, via the intermediate $2p\pi_u$ state. Thus the rotationally induced transition $2p\sigma_u \to 2p\pi_u$ is followed by another dynamically induced transition $2p\pi_u \to 3p\sigma_u$, whenever the collision is fairly close, and the internuclear line rotates not too fast and not too slowly.

The present data then indicate that some kind of nonadiabatic coupling to the $3p\sigma_u$ or other states [i.e., H(2$s$) production] is indeed important below about 12 keV, and must in general be included in calculations of close $H^+$-H collisions. The inclusion of the $3p\sigma_u$ state generally reduces the repletion of the $2p\sigma_u$ state through the intermediate $2p\pi_u$ state; it therefore increases the damping of the overall charge-transfer probability $P_0$ below 12 keV, as is needed to bring theory into closer agreement with Everhart's data at these energies.

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## EXCITATION OF Ar I AND He I IN LOW-ENERGY $He^+ + Ar$ COLLISIONS*

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Previous work on investigations of spectral lines emitted from low-energy $He^+ + Ar$ collisions has indicated the presence of emission solely from Ar II. We have extended these investigations to show that both Ar I and He I spectra are also present and have measured the relative excitation cross sections for the 7635- and 5876-Å lines of these two species for energies less than 700 eV.

The excitation of rare-gas atoms by collisions with low-energy (<1000 eV) ions has been the subject of several recent spectroscopic experiments. In previous investigations of excitation caused by $He^+ + Ar$ collisions, Lipeles, Novick, and Tolk[1] have presented spectra which revealed only the presence of lines of Ar II. Schlumbohm[2] has also reported similar spectra from his experiments and has measured the excitation cross sections for several lines. Most recently, Lipeles et al.[3] have published cross sections for the excitation of the 4610- and 4765-Å lines of Ar II; these results reveal much more structure than the cross sections measured by Schlumbohm. A qualitative understanding of the origin of this structure in the excitation cross sections can be inferred from the complicated behavior of the potential curves of the system as a function of internuclear separation. Rosenthal and Foley[4] have shown that pseudocrossings of the adiabatic energy curves of excited states of the $He^+ + He$ system are responsible for oscillations in the excitation total cross sections of the $^1S$ and $^3S$ states. The same mechanism may well give rise to the structure observed in the $He^+ + Ar$ cross sections and in addition might be expected to produce excited products other than Ar II. In the present paper we demonstrate the results of $He^+ + Ar$ reactions which indeed show that both excited He I and Ar I are also produced by collisions of this system. It appears that a thorough study of the excitation of these two species as well as the excitation of Ar II is necessary in order to construct a complete picture of the experimental reactions which take place.

Our optical observations are made in a collision chamber in which the pressure of argon is maintained at $7\times10^{-4}$ Torr. The helium-ion beam is produced by bombardment of neutral helium with 50-V electrons and the spread in energy of the beam is 3 V full width at half-maximum intensity. Light is focused onto the entrance slit of a Jarrell-Ash model No. 82-410, 0.25-m monochromator. The slit is oriented perpendicular to the ion beam. Although the cross-sectional dimension of the beam varies with energy, the optical image is always much narrower than the length of the entrance slit of the monochromator, and the relative line intensities as a function of energy are not distorted by the variation in geometry of the beam. A cooled EMI 9558Q photomultiplier is used as a detector, and typical dark counting rates are 5 pulses per second.

At wavelengths less than 6000 Å the Ar I lines are generally obscured by the strong Ar II spectrum, and only at longer wavelengths can unmistakable identifications be made. Figure 1 shows a portion of the spectrum in the region from 7500 to 7800 Å which has been produced by collisions; a comparison spectrum from an argon discharge lamp has also been plotted in order to emphasize the identification. Lines which are seen in the collision spectrum, but not in the lamp spectrum, are second-order lines from excited argon ions. The line at 7635 Å is the strongest feature detected in the Ar I spectrum. Other lines at longer wavelengths may actually be more intense, but the quantum efficiency of the photomultiplier decreases rapidly toward longer wavelengths and is almost zero at 8000 Å. Many lines which are strong in gas discharges lie above 8000 Å and may well be excited by collisions also.

The relative excitation cross sections have been measured for both the 7635-Å line of Ar I