Production of H Atoms in the 3s State by the Dissociation of Fast H_2^+ and H_3^+ Projectiles on Impact with H_2, He, Ar, and Ne†

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Absolute cross sections have been determined for the production of hydrogen atoms in the 3s state by the dissociation of fast (20–120 keV) H_2^+ and H_3^+ projectiles on impact with H_2, He, Ar, and Ne. A cross section has also been determined for 100-keV H_2^+ impact on N_2. For 100-keV H_2^+ impact on these gases, the production of atoms in the 3s state represents about 3% of the reaction $H_2^+ \rightarrow H^+ + H$.

I. INTRODUCTION

A PREVIOUS paper[1] described an experiment in which a fast H^+ beam was passed through a differentially pumped collision chamber containing a target gas. Electron capture into excited states of H took place in the chamber and the resulting atoms emerged into an evacuated observation chamber where they decayed in flight. Capture into the long-lived 3s state of H was measured by observing the intensity of $H_\alpha(n=3 \rightarrow 2)$ radiation at a point in the observation chamber where the faster 3p and 3d states had decayed off.

This paper describes an experiment which uses the same general technique and apparatus to measure the production of hydrogen atoms in the 3s state by the dissociation of fast (20–120 keV) H_2^+ and H_3^+ projectiles on impact with H_2, He, Ar, and Ne. In this case an H_2^+ or H_3^+ beam is passed through the differentially pumped collision chamber containing the target gas and the intensity of H_α radiation is monitored in the observation chamber at a point where only the $3s \rightarrow 2p$ radiation is still present.

If cascade is neglected and if the ion-beam flux is assumed constant (low-pressure approximation), the density of observed beam atoms in an excited state, n^*, is given by

$$n^* = F\rho Q\tau(1-e^{-L/v\tau})e^{-x/v\tau},$$

where F is the ion-beam flux, ρ is the gas density in the collision chamber of length L, Q is the cross section for scattering the excited H atoms in the forward direction, τ is the lifetime of the state, x is the distance from the collision-chamber aperture, and v is the ion velocity.

The H_2^+ and H_3^+ projectiles can dissociate on collision, producing hydrogen atom fragments in the 3s state by the following reactions:

$$(1)\quad H_2^+ + A \rightarrow H(3s) + H^+ + A,$$
$$(2)\quad H_2^+ + A \rightarrow H(3s) + H + A^+,$$
$$(3)\quad H_3^+ + A \rightarrow H(3s) + [HH^+] + A,$$
$$(4)\quad H_3^+ + A \rightarrow H(3s) + [HH] + A^+,$$

where A represents the target gas. The products of reactions (1) and (3) result from the production of an excited dissociative state of the projectile ion, and the products of reactions (2) and (4), result from electron capture into an excited dissociative state of the neutral species.

II. APPARATUS

The apparatus has previously been described.[1] Chamber lengths of 5.5 and 12.5 cm were used in this investigation.

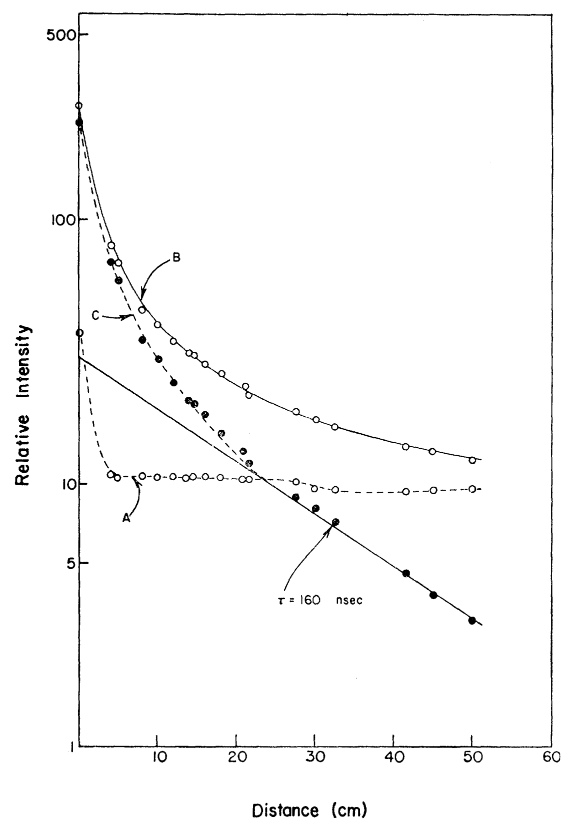

FIG. 1. Intensity of H_α radiation versus distance from the exit aperture of the collision chamber for 20-keV H_2^+ impact on He. Curve A, "background" using an evacuated collision chamber (see text). Curve B, total signal using a helium-filled collision chamber. Curve C, net signal from curve B minus curve A. The increase in the "background" curve A near the aperture is produced by the beam's striking the exit aperture, probably causing outgassing. It has been subsequently eliminated by collimating the beam so that it does not strike the aperture. It has no effect on the 3s data.

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[1] R. H. Hughes, H. R. Dawson, B. M. Doughty, and C. A. Stigers, Phys. Rev. 146, 53 (1966).

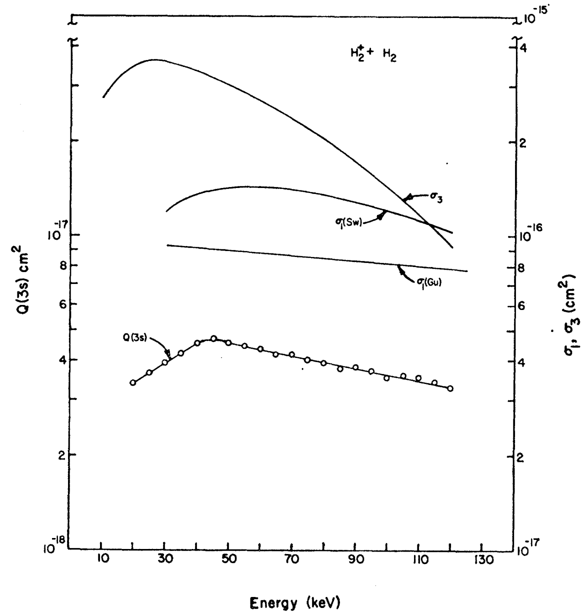

FIG. 2. Cross sections for the reaction $H_2^+ \to H + H^+$ as measured by Guidini, σ_1(Gu) (Ref. 3), and by Sweetman, σ_1(Sw) (Ref. 4); cross sections for the reaction $H_2^+ + e \to H + H$, σ_3 (McClure, Ref. 2); and cross sections for the production of H(3s) atoms by H_2^+ impact on H_2, $\alpha(3s)$ versus energy.

III. TREATMENT OF DATA

Figure 1 shows a plot of H_α intensity versus distance from the exit aperture of the collision chamber for 20-keV H_2^+ impact on He. As can be seen, a considerable "background" exists from radiation produced by the interaction of the beam with the residual gas in the observation chamber. This background (curve A, Fig. 1) is obtained by monitoring the radiation when the collision chamber is evacuated but with the observation chamber at the same pressure as during an actual run; the background is then subtracted from the raw data (curve B) taken with the collision chamber filled with the target gas. The difference leads to the final decay curve (curve C) from which the measurements are obtained.

The decay curve (C) shows that at a sufficiently great distance from the aperture the shorter-lived $3p$ and $3d$ states have decayed off and only the 160-nsec $3s$ decay is left. All data were obtained at a distance from the exit aperture equal to or greater than the velocity times the lifetime of the $3s$ state. It was ascertained that at these distances the radiation fell on an $\exp[-x/v\tau_{3s}]$ curve for all gases.

Collision-chamber pressures were in the range of 10^{-3}–10^{-4} Torr. In this range, the signals were found to be proportional to both current and pressure. However, there was a slight beam-current attenuation produced either by charge transfer or scattering of charged particles to the inside of the collision chamber. The beam attenuation was held to 10% or less. The mean ion flux

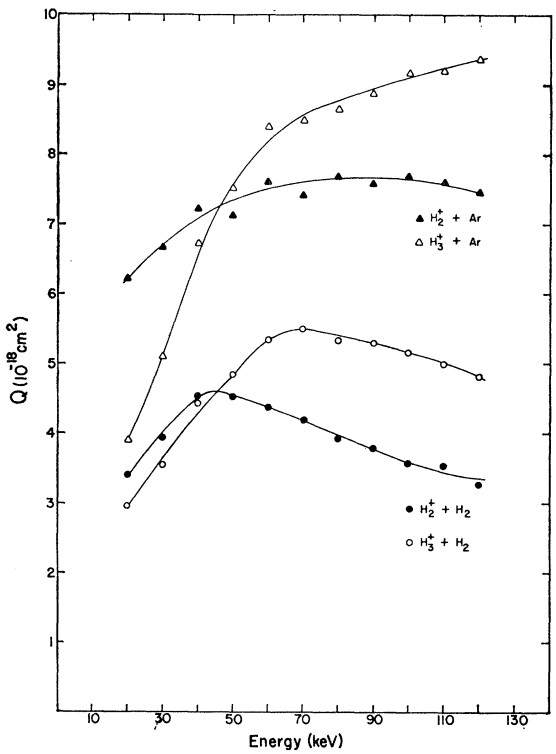

FIG. 3. Cross sections for the production of H(3s) atoms by H_2^+ and H_3^+ impact on Ar and H_2 versus energy.

was used in determining the cross section for the long-lived $3s$ state.

In general, the apparent cross sections derived from the two chambers were in close agreement at the higher energies. However, a systematic discrepancy developed

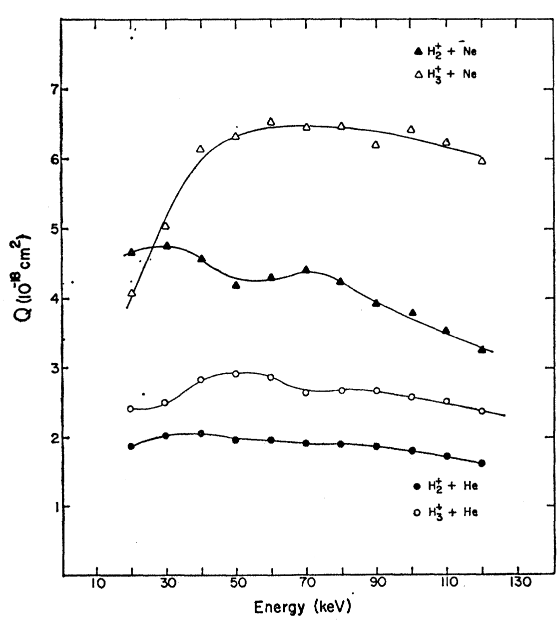

FIG. 4. Cross sections for the production of H(3s) atoms by H_2^+ and H_3^+ impact on Ne and He versus energy.

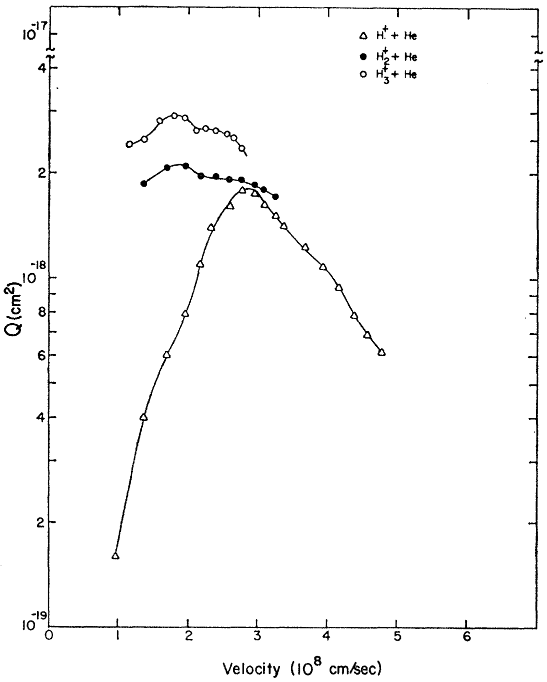

FIG. 5 Cross sections for the production of H(3*s*) atoms by H^+ (Ref. 1), H_2^+ and H_3^+ impact on He versus projectile velocity.

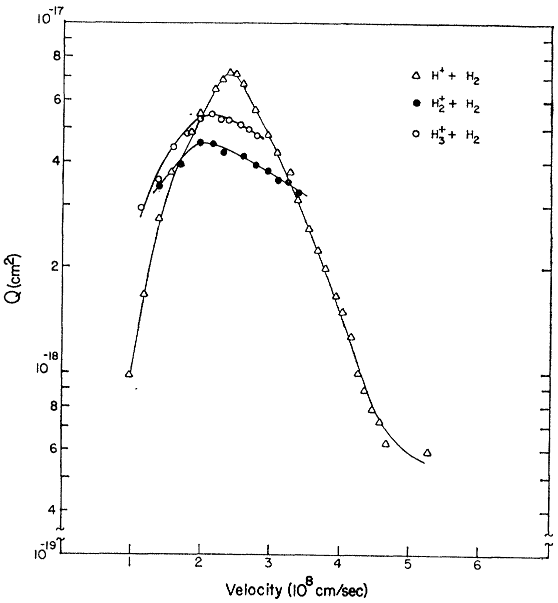

FIG. 6. Cross sections for the production of H(3*s*) atoms by H^+ (Ref. 1), H_2^+ and H_3^+ impact on H_2 versus projectile velocity.

between the two sets of data at the lower energies, the long chamber tending to give the smaller cross section. This discrepancy reached a maximum of 5–15% at 20 keV, depending on the gas, and is attributed to large-angle scattering of excited atoms, which results in the loss of excited atoms to the inside of the collision chamber.

Since the large-angle-scattering loss is expected to be much more severe in the long chamber as compared with the short chamber, the approximate agreement between the two chambers indicates that large-angle scattering is not a particularly important process in these reactions. A rough calculation was performed, using the angular dependence of the differential cross section for H atom scattering as measured by McClure[2] for 20-keV H_2^+ on H_2. The calculation indicated that if there were a 25% discrepancy between the two sets of apparent cross sections then the short-chamber apparent cross section would be in error by 5% or less. Since this is within the the reproducibility, we simply weight the short-chamber data very heavily, particularly at the lower energies, and admit to the possibility of a small underestimate of the cross sections at the lower energies.

The background from the interaction of the molecular-ion beam with the residual gas in the observation chamber is considerably greater than when the proton beam is used. At a distance of $v\tau_{3s}$ from the exit aperture, the fraction of the total signal which is due to this background is about three times that observed with a proton beam. This fact reduces the molecular-ion reproducibility relative to the proton work of Ref. 1. The molecular-ion cross-section reproducibility is between 10 and 15%.

IV. RESULTS AND DISCUSSION

Considerable analysis has been performed on the reaction $H_2^+ + H_2$. The cross section for the reaction $H_2^+ \rightarrow H + H^+$, termed σ_1, has been measured by Guidini[3] and by Sweetman[4]. The cross section for the charge-transfer reaction $H_2^+ + e \rightarrow H + H$, termed σ_3, has been determined by McClure.[2] A certain fraction of each of these two reactions will contribute to the total cross section for the production of H atoms in the 3*s* state.

In Fig. 2 we show not only a plot of our cross section for the production of H atoms in the 3*s* state by H_2^+ on H_2 but also σ_1 (Guidini) and σ_3, taken from Fig. 2 of Ref. 2, and σ_1 as measured by Sweetman. We noted that σ_3 is a rather rapidly varying function of energy, particularly at the higher energies, while σ_1 is a relatively slow function of energy. We made the assumption that the fraction of each reaction contributing to the 3*s* cross section is a slowly varying function of energy over a small energy range. Thus $Q(3s) = f_1\sigma_1 + f_3\sigma_3$, where f_1 and f_3 are the fractions associated with each reaction. Treating f_1 and f_3 as constants over relatively small energy intervals of 20 keV, we set up simultaneous equations for $Q(3s)$ and solved for f_1 and f_3 within each interval. Using this procedure we found that we could fit a constant f_1 and f_3 not only for small energy inter-

[2] G. W. McClure, Phys. Rev. **140**, A769 (1965).

[3] J. Guidini, Compt. Rend. **253**, 829 (1961).

[4] D. R. Sweetman (private communication). These cross sections are also displayed in Oak Ridge National Laboratory Report No. ORNL-3113, revised 1964 (unpublished).

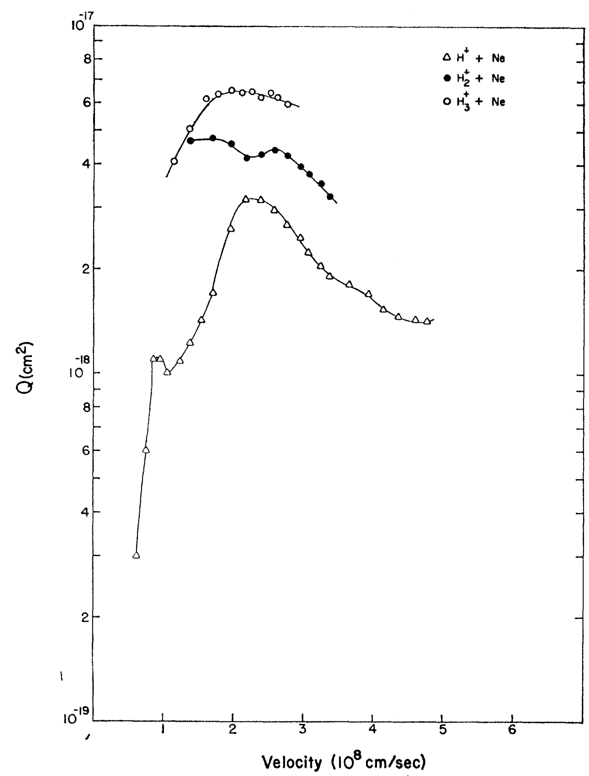

FIG. 7. Cross sections for the production of H(3*s*) atoms by H^+ (Ref. 1), H_2^+ and H_3^+ impact on Ne versus projectile velocity.

vals but over a much larger energy range. Using McClure's σ_3 values and Guidini's σ_1 values we found that $f_1 \approx 0.037$ and $f_3 \approx 0.004$ would fit well from 40 to 120 keV. Below 40 keV, the f_1 and f_3 values varied. However, using Sweetman's σ_1 and modifying McClure's σ_3 values, which he had deduced by using Guidini's values of σ_1, we found that $f_1 = 0.028 \pm 0.001$ and $f_3 = 0.002$ fit well over the range 30–120 keV. Extrapolating Sweetman's σ_1 to 20 keV indicated that these values of f_1 and f_3 would fit over the entire energy range of 20–120 keV. There is no reason *a priori* that such constant fractions should be maintained, but if they are maintained, then our 3*s* curve is more consistent with the shape of Sweetman's σ_1 curve than with the shape of Guidini's curve at lower energies. At any rate, this crude analysis would seem to indicate that charge transfer is not particularly efficient in producing atoms in the 3*s* state. It would follow, then, that at energies above 100 keV where the total charge transfer cross section is dropping rapidly, the dominant reaction is the one which produces an atom in the 3*s* state simultaneously with a proton.

Figure 3 shows the production of atoms in the 3*s* state by H_2^+ and H_3^+ impact on Ar and on H_2. Figure 4 shows the production of atoms in the 3*s* state by H_2^+ and H_3^+ impact on Ne and on He.

For the purposes of comparison we plot the production of atoms in the 3*s* state by H^+ (Ref. 1), H_2^+, and

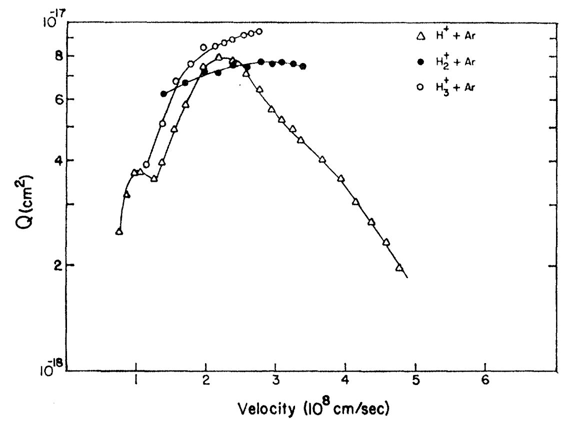

FIG. 8. Cross sections for the production of H(3*s*) atoms by H^+ (Ref. 1), H_2^+, and H_3^+ impact on Ar versus projectile velocity.

H_3^+ impact as a function of velocity for impact on He, H_2, Ne, and Ar in Figs. 5–8, respectively. A notable feature of these plots is the fact that the maximum cross sections for these three projectiles are all fairly comparable. The H^+ curves have the rapid decline on either side of the maximum which is characteristic of charge transfer. One also notices that the H_3^+ cross section exhibits a similar rapid decline on the low-energy side of the maximum for H_3^+ impact on H_2, Ne, and Ar, which may be indicative that charge transfer is an important process in these reactions at the lower energies.

We were curious to see what the ratio of our 3*s* cross section to σ_1 for high-energy H_2^+ impact would be for several gases. This would be especially meaningful if charge transfer is not contributing an appreciable amount to the 3*s* cross section. Since both Guidini and Sweetman have measured σ_1 for impact on N_2, we made a 3*s* cross-section measurement for H_2^+ impact on N_2. This cross section is included in Table I, where

TABLE I. Cross sections (in 10^{-18} cm^2) and cross-section ratios for 100-keV H_2^+ impact on gases.

	H_2	Ar	N_2	He	Ne
σ_1 (Guidini)	80	220	210	61	
σ_1 (Sweetman)	120	320	280	57	
$\bar{\sigma}_1$ (average)	100	270	250	59	
$Q(3s)$	3.6	7.7	8.8	1.8	3.8
$100\,Q(3s)/\bar{\sigma}_1$	3.6	2.9	3.5	3.1	

we also tabulate our interpretation of σ_1 from Guidini's and Sweetman's graphs.

Within experimental accuracy it would appear that the ratio of the 3*s* cross section to σ_1 for 100-keV H_2^+ impact is independent of the gas and has a value of about 3.3%. On this basis we would predict σ_1 for 100-keV H_2^+ on Ne to be about 1.3×10^{-16} cm^2.