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H_2 AND H PRODUCTION IN COLLISIONS OF 5- TO 30-keV H_2^+
IONS WITH XENON GAS†

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ABSTRACT

Cross sections for H_2 production (nondissociative charge transfer) from 5- to 30-keV H_2^+ collisions with xenon are reported. The measurements were made using a thin-window proportional counter as a particle detector. Using these and other available measurements, we determined the cross section for total dissociation and that for H atom production. We show that $H_2^+ \rightarrow H + H$ is the dominant dissociation process.

I. INTRODUCTION

The difficulty in separately detecting the molecular and atomic neutral products from collisions of molecular projectiles in a gas target can be overcome by using a proportional counter with a narrow entrance slit. Several investigators have used this technique to study neutral products from H_2^+ collisions in H_2 .¹⁻³ In this paper measurements of H_2 production from collisions of 5- to 30-keV H_2^+ ions with xenon gas are reported.

Neutral products from H_2^+ collisions in a gas target can arise from the following processes:

(The notation throughout will be that used by Sweetman⁴ and Morgan, et al.⁵)

The undissociated hydrogen molecules formed by process 4 have essentially the same energy as the primary ions and are practically all contained in a very narrow cone.³ The dissociation fragments have the original kinetic energy of the incident particle nearly equally divided between them. Therefore, any energy-sensitive detector can be used to distinguish between the dissociated and undissociated products, provided that the two atomic fragments from process 3 are not detected simultaneously, since they would deposit the same energy in

the detector as an undissociated molecule. The dissociation energy (≈ 4 eV) results in fragments from process 3 being more widely scattered than the hydrogen molecules formed in process 4. Hence the use of a narrow detector-entrance slit ensures they are not detected simultaneously. The molecular-beam intensity can then be obtained by sweeping this narrow slit across the beam.

These measurements together with those of Morgan, et al.⁵ (made in this laboratory with the same apparatus) contribute to the accumulation of a self-consistent set of cross sections for interactions of H_2^+ with xenon. With these results it was possible to calculate the dissociation cross section and H-atom-production cross section.

There are no experimental results available for direct comparison; however, measurements of H_2 production from H_2^+ collisions with H_2 in the present energy range have been reported in the literature.^{1,2} Therefore, at several energies, cross sections were obtained using a hydrogen target so that some comparison with previous measurements could be made.

II. EXPERIMENTAL APPROACH

A. Apparatus

H_2^+ ions, produced in the same source described by Morgan, et al.⁵ but operated in the simple electron-bombardment mode, were accelerated to the required energy and, after being focused, were mass analyzed in a 20° bending magnet before entering the experimental chamber (shown in Fig. 1) through a 0.5-cm-diam aperture. The H_2^+ currents were typically of the order of 10^{-15} A, ensuring a suitably small counting rate at the detector. The maximum counting rate for the total-beam measurements was about 25 000 counts/sec, while for H_2 this was typically about 5000 counts/sec; with the gas cell evacuated the H_2 and H backgrounds were approximately

300 and 200 counts/sec respectively.

The H_2^+ beam passed through a defining aperture of 0.25-cm diam before entering the target gas cell, which had an entrance aperture of 0.05-cm and an exit aperture of 0.3-cm diam. The xenon gas pressure in the target gas cell was measured using a Barocel capacitance manometer which had been checked with an oil manometer.⁵ The effective gas-cell length was calculated using a Monte Carlo code⁵ and this agreed with the geometric length of 9.37 cm to within 2%. The charged component of the beam leaving the target cell could be swept from the beam path electrostatically. The beam was detected using a thin-window proportional counter built by Barnett, et al.⁶ following a design by McClure and Allensworth.⁷ The beam entered the counter through a 2.5- x 0.0025-cm slit, which was covered with a Formvar window, the thickness of which was estimated to be $8 \mu\text{g}/\text{cm}^2$ using the techniques described by McClure and Allensworth.⁷ The slit could be moved horizontally at a rate of 6 mm per min (constant in both directions to within 6%) using a synchronous motor drive and had a distance of travel of 2.5 cm, corresponding to the width of the proportional counter entrance aperture. While this distance of travel was convenient in establishing the beam dimensions, moving the slit approximately ± 0.4 cm from the central position reduced the counting level to that of the detector background noise ($\sim$ few counts/sec).

The cathode of the proportional counter was biased at approximately +70 V with respect to ground, with the anode biased at approximately +760 V. The counter was operated at a methane pressure between 510 and 560 Pa. The amplification of the counter was found to vary with the pressure and electrode voltages; during measurements, therefore, the pressure was held constant to within 13 Pa and the electrode voltage to better than one volt, thereby holding the amplification constant to within 8%.

The detector output voltage pulse was developed across a 10^6 -ohm resistor and coupled through a 0.001- μ F capacitor. After amplification the signal was displayed on a multichannel pulse-height analyzer and also routed through a single-channel analyzer to a scaler.

A typical pulse-height distribution for a 10-keV H_2^+ beam is shown in Fig. 2. The peaks of the "half-energy" H and H^+ and "full-energy" H_2 and H_2^+ beam components are clearly resolved. With the aid of the multichannel pulse-height analyzer we set the single-channel analyzer discriminator levels so that only the full-energy particles were counted. From the "half-energy" particle counting rate, we estimate that accidental coincidences would contribute less than 2% to the H_2 signal. While the entrance slit of the proportional counter was narrow it was 2.5 cm long in the vertical direction and so it was necessary to consider the possibility of two H atoms from process 3 entering the counter in that plane; to do this a simple calculation was performed. The two H atoms were assumed to have a velocity corresponding to dissociation with a minimum energy of 2.25 eV (the worst case). This velocity was superimposed on the velocity of the incident beam and the probability of the two particles entering the chamber simultaneously could be calculated using simple geometry. It was found that much less than 1% of the total signal would be due to these two H particles from process 3.

B. Procedure

Cross sections for H_2 production were obtained using the thin-target approximation

$$\sigma_4 = (S_{H_2}/S_{tot})/\pi \quad (1)$$

where σ_4 is the cross section for the production of the H_2 molecule in cm^2 /atom, S_{tot} is the H_2^+ -plus- H_2 signal, S_{H_2} is the signal for H_2 only, and π is the target thickness (atoms/ cm^2).

For each cross-section measurement a plot of the fraction S_{H_2}/S_{tot} versus pressure was made. Each fraction measurement was obtained by scanning the slit across the total primary beam and then repeating the scan with the charge component removed and recording the total number of counts in each case. Since only full-energy particles are counted, as described before, we have a measure of the total molecular-beam intensity and the H_2 -beam intensity. For each measurement the slit was swept in both directions and in all but a few cases the fractions agreed to within $\pm 10\%$; these fluctuations could be ascribed to variations in beam intensity and sweep rate. The gas-cell pressure was varied from 1.3×10^{-4} to 4×10^{-2} Pa.

Since the incident-beam intensity could be reduced by collisions involving fragmentation, the slope of the fraction-versus-pressure plot was obtained from a least-square fit of the data, which included an attenuation correction using the total-neutral-, proton-, and negative-production cross sections obtained by Morgan, et al.⁵ This attenuation correction at no time exceeded 5%. A typical plot of the fractional H_2 yield versus target thickness is shown in Fig. 3; also shown is the solution to the simple least-square fit to the data (solid line) and the result of the least-square fit with the attenuation correction (dashed line).

It was possible to observe scattering into angles of up to ± 0.75 deg, limited by the range of motion of the slit. Sweeps of the incident H_2^+ beam at 10 keV indicated a full width at half maximum of about 0.1 deg, while the H_2 beam from 10-keV- H_2^+ collisions in H_2 had a full width at half maximum of about 0.15 deg, which is consistent with the observations of McClure in H_2 .³ For 10-keV- H_2^+ collisions in Xe the H_2 beam

had a full width at half maximum of about 0.11 deg, which ensures that all the H_2 produced were detected as the slit was moved through its 1.5 deg range.

As a confirmation of these procedures, cross-section measurements for H_2 production in H_2^+ collisions with H_2 at 10 and 20 keV were made. These and the results of Schmid¹ and McClure² are shown in Table I. The absolute values agree to within the experimental uncertainties.

C. Accuracy of Measurements

The standard random uncertainty in the H_2 -production cross section, based on the reproducibility of measurements at 10 and 30 keV, is $\pm 8\%$, while the absolute magnitudes may have an additional systematic uncertainty of $\pm 5\%$ due to gas-cell-length determination, pressure measurement, extraneous coincidence counts, and possible beam loss in the analyzing region due to methane leakage through the detector window. This leakage increases the background pressure in the analysis region from 4×10^{-4} to 7×10^{-4} Pa. However, we estimate⁸ that this has not more than a 1% effect on the cross-section measurements. The good agreement with Schmid's¹ and McClure's² results (Table I) confirms that the effect is within the experimental uncertainties indicated. The acceleration voltage was measured with a calibrated high-impedance divider; we assign a maximum standard uncertainty of $\pm 4\%$ to our energy values.

III. RESULTS AND DISCUSSION

A. H_2 -Production Cross Sections

The cross sections σ_4 for the production of H_2 from 10- to 30-keV H_2^+ in Xe are presented in Table II and Fig. 4. The cross sections

below 10 keV were obtained using a D_2^+ primary beam; however, all cross sections are shown in Fig. 4 at the equivalent H_2^+ energy. As can be seen from Table II and Fig. 4, for the same relative impact velocity ($\sim 1.0 \times 10^8$ cm/sec), the results agree to within the estimated standard random uncertainty of $\pm 8\%$.

It is well known that the interpretation of experimental data for collisions involving fast H_2^+ ions is complicated since it is difficult to specify the degree of vibrational excitation of the primary H_2^+ beam. In previous cross-section measurements of H_2^+ incident on H_2 , McClure² observed changes in the H_2 -production cross sections and dissociation cross sections when the gas pressure in the ion source was varied by a factor of 5 (presumably due to changes in the population of vibrational states of the primary beam). The degree of vibrational excitation of our primary H_2^+ beam is not known, but we note that during the present measurements the gas pressure was varied by a factor of 8 in the electron-bombardment mode and the source was operated in a low-voltage-discharge mode; within the experimental uncertainties, no variation in the H_2 -production cross section was observed.

The apparatus used in these measurements of σ_4 has also been used by Morgan, et al.⁵ to make measurements of the cross sections for H^+ production (σ_+), H^- production (σ_-), and the total neutral-power production (σ_0) from H_2^+ collisions in Xe. The present measurements, along with those of Morgan, et al.⁵ allow several other cross sections to be calculated: the dissociation cross section, σ_d , and the cross section for the production of H atoms, σ_H .

B. Dissociation Cross Sections

Dissociation of H_2^+ in collisions with Xe can take place through the following processes:

The dissociation cross section σ_d is the sum of the cross sections for all these processes. The measurements of Morgan, et al.⁵ are

$$\sigma_+ = \sigma_1 + 2\sigma_2 + \sigma_5 \quad (2)$$

$$\sigma_- = \sigma_5 + \sigma_6 + 2\sigma_7 \quad (3)$$

and
$$\sigma_0 = 1/2 \sigma_1 + \sigma_3 + \sigma_4 + 1/2 \sigma_6. \quad (4)$$

It can be seen that the dissociation cross section can be calculated from these cross sections and the cross section for H_2 production (σ_4) reported in this paper, using the relationship

$$\sigma_d = \frac{\sigma_+}{2} + \frac{\sigma_-}{2} + \sigma_0 - \sigma_4. \quad (5)$$

The results of such a calculation are presented in Fig. 4 and Table II.

In Ref. 5 a thermal detector was used to measure the neutral particles produced in collisions with Xe, and it was not possible to distinguish between "full-energy" H_2 molecules and "half-energy" H atoms; hence a neutral-power-production cross section, σ_0 , was reported. Since we have measured the H_2 -production cross section, it is now possible to subtract this contribution from σ_0 to obtain the cross section for the production of H atoms:

$$\sigma_H = 2(\sigma_0 - \sigma_4). \quad (6)$$

The factor of 2 is necessary because two "half-energy" H atoms deliver the same power to a thermal detector as one "full-energy" H_2 molecule. In terms of the fundamental dissociation processes listed above, we see that

$$\sigma_H = \sigma_1 + 2\sigma_3 + \sigma_6. \quad (7)$$

This cross section is also listed in Table II.

We can determine which of the three processes (1, 3, or 6) is the dominant contributor to σ_H by considering the relative magnitudes of σ_H and σ_+ and σ_- reported in Ref. 5. The cross section σ_- is at least two orders of magnitude smaller than σ_H in our energy range, and, since process 6 is only one of the contributors to σ_- [Eq. (3)], we can conclude that the last term in Eq. (7) is negligible. The magnitude of σ_+ is less than 0.2 that of σ_H and from Eq. (2) we see that $\sigma_1 < \sigma_+$; therefore, the contribution of σ_1 to σ_H must be less than 0.2, and we can approximate Eq. (7) by

$$\sigma_3 \approx 1/2 \sigma_H. \quad (8)$$

We conclude that the production of H atoms in our energy range occurs mainly via dissociative charge transfer (process 3). We have plotted $1/2 \sigma_H$ in Fig. 4 to illustrate that process 3 accounts for most of the dissociation.

The absolute uncertainties in these calculated cross sections are shown in Fig. 4 and Table II and were obtained by combining the magnitude of the absolute uncertainties of each of the measured cross sections used in the calculation.⁹

C. Discussion

Stedeford and Hasted¹⁰ interpreted their charge-exchange cross sections in terms of Massey's adiabatic principle. They attributed the maximum in their H_2^+ -Xe cross section at about 1200 eV to a dissociative process. The present data show that the predominant process at the lower energies is nondissociative charge transfer, i.e.,

Dastidar and Barua¹¹ have used the sudden approximation to calculate the total vibrational-dissociation cross section at 10 keV for H_2^+ in Xe. They predict a value of $6.2 \times 10^{-18} \text{ cm}^2$ for this cross section. Our value for the total dissociation cross section at 10 keV is $9 \pm 2 \times 10^{-16} \text{ cm}^2$. This would seem to indicate that most of the dissociation is through electronic excitation at this energy.

The cross sections in Fig. 4 also show that most of the dissociation in the present energy range takes place through the process

a feature also observed in H_2^+ collisions with H_2 at these energies.^{1,2}

IV. ACKNOWLEDGMENTS

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FOOTNOTES AND REFERENCES

[†]Work performed under the auspices of the U. S. Energy Research and Development Administration.

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8. We are not aware of any published cross sections for H_2^+ or H_2 collisions in methane. However, even if the total-loss cross sections for H_2^+ and H_2 in methane were as large as 10^{-15} cm^2 , the effect on our measured cross sections would be the order of 1%.
9. The absolute uncertainties in these derived cross sections were calculated by assuming all the uncertainties in the contributing measured cross sections were random. Therefore

$$\delta\sigma_d = (1/4 \delta\sigma_+^2 + 1/4 \delta\sigma_-^2 + \delta\sigma_0^2 + \delta\sigma_4^2)^{1/2}$$

and

$$\delta\sigma_H = 2(\delta\sigma_0^2 + \delta\sigma_4^2)^{1/2}.$$

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Table I. Cross sections (σ_4) for the production of H_2 from collisions of H_2^+ with H_2 gas. The present results are compared with those of Schmid¹ and McClure.² Our measurements have an estimated random uncertainty of $\pm 8\%$, and a possible systematic uncertainty of $\pm 5\%$, giving an absolute uncertainty of $\pm 9.5\%$.

H_2^+ Energy (keV)	σ_4 (10^{-16} cm ² /molecule)		
	Present ($\pm 9.5\%$)	Schmid ¹ ($\pm 25\%$)	McClure ² ($\pm 10\%$)
10	4.7	5.0	4.8
20	4.0	3.7	3.7

Table II. Experimental and deduced cross sections (in units of 10^{-15} cm^2) for collisions of H_2^+ in xenon. The cross sections for H_2 production (σ_4) have standard relative uncertainties of $\pm 8\%$ and possible systematic uncertainties of $\pm 5\%$. The dissociation cross sections, σ_d , and the H-atom-production cross sections, σ_H , were derived from σ_4 and the results reported in Ref. 5; the absolute uncertainties are indicated.⁹ Note that the cross sections for D_2^+ ions are tabulated at the H_2^+ energy (i.e., 10- to 20-keV D_2^+ ions were used).

	H_2^+ energy (keV)	σ_4 ($\pm 8\%$)	σ_d	σ_H
D_2^+	5	1.76	0.6 ± 0.3	1.0 ± 0.5
	6	1.36	0.8 ± 0.2	1.5 ± 0.4
	7.5	1.46	0.7 ± 0.2	1.2 ± 0.4
	10	1.15	0.9 ± 0.2	1.6 ± 0.3
H_2^+	10	1.07	0.9 ± 0.2	1.6 ± 0.3
	15	1.01	0.9 ± 0.2	1.5 ± 0.3
	20	0.84	0.8 ± 0.1	1.4 ± 0.3
	25	0.71	0.9 ± 0.1	1.6 ± 0.3
	30	0.67	---	---

FIGURE CAPTIONS

- FIG. 1. A schematic diagram of the experimental apparatus.
- FIG. 2. A typical pulse-height distribution (100 channels) of detector-output pulses for 10-keV primary H_2^+ ions, with the detector slit centered on the beam axis. The predominant peak is due to 10 keV H_2^+ and H_2 , and the smaller peak is due to 5 keV H^+ and H , the dissociation fragments.
- FIG. 3. Plot of fractional H_2 yield versus target thickness, π , for 15-keV H_2^+ in xenon. $\bullet$, data points; —, simple least-square fit; ---, least-square fit with attenuation correction.
- FIG. 4. Cross sections for collisions of H_2^+ with Xe gas. Present results: $\blacksquare$, $\square$, H_2 production, σ_4 (open squares indicate a D_2^+ incident ion); $\circ$, dissociation, σ_d ; $\triangle$, one-half the H-atom production, $1/2 \sigma_H$, which is approximately the cross section for $\text{H}_2^+ \rightarrow \text{H} + \text{H}$, σ_3 . Also shown: $\bullet$, total neutral-power production, σ_0 ; $\blacklozenge$, proton production, σ_+ (from Morgan, et al.⁵). Error bars on σ_4 indicate the relative uncertainties; those on σ_d and $1/2 \sigma_H$ indicate the absolute uncertainties. Note that the cross sections for D_2^+ ions are plotted at the equivalent H_2^+ energy.

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XBL 756-3294

Fig. 1

XBB 754-3137

Fig. 2

XBL 763-2365

Fig. 3

XBL763-2364

Fig. 4

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