DISSOCIATION OF MOLECULAR HYDROGEN IONS IN COLLISIONS WITH GAS MOLECULES

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The cross sections for the dissociation of molecular hydrogen ions (H_2^+) in single collisions with H_2 and N_2 molecules and with He, Ar, and Kr atoms are measured at energies from 200 to 1200 kev.

1. INTRODUCTION

RECENT measurements of the cross sections for the dissociation of molecular hydrogen ions have been connected mainly with the problem of injecting hydrogen ions into thermonuclear devices and accelerators. Measurements have been performed by Damodaran,[1] Fedorenko et al.,[2] and Barnett[3] at energies up to 200 kev in hydrogen, nitrogen, helium, and argon. Barnett[3] has also investigated H_2^+ dissociation at 500 — 2200 kev in the same gases. Sweetman[4] very recently reported cross sections for H_2^+ dissociation in hydrogen at 100 — 800 kev. The cross sections for proton production given in the first three of the aforementioned papers do not agree; it is indicated in reference 2 that different geometries can account for the discrepancies.

There have been no measurements of the cross section for H_2^+ dissociation within the 200 — 500 kev range in any gas besides hydrogen. Because of the scientific and practical importance of such cross sections, we have investigated molecular hydrogen ion dissociation in different target gases from 200 to 1200 kev.

2. APPARATUS AND EXPERIMENTAL TECHNIQUE

We produced molecular hydrogen ions by means of an electrostatic accelerator, using an ion source with a cold cathode. We have described the accelerator in reference 5.

Figure 1 is a diagram of our apparatus. The hydrogen ion beam from the electrostatic accelerator passed through a collimating aperture 1 of diameter 4 mm into a magnetic mass monochromator (analyzer) 2, in which the ion beam was deflected 17°. The H_2^+ beam passed from the monochromator through a 2-mm diaphragm 4 and channel of diameter 6.5 mm and length 100 mm into the collision chamber 3, from which the ions

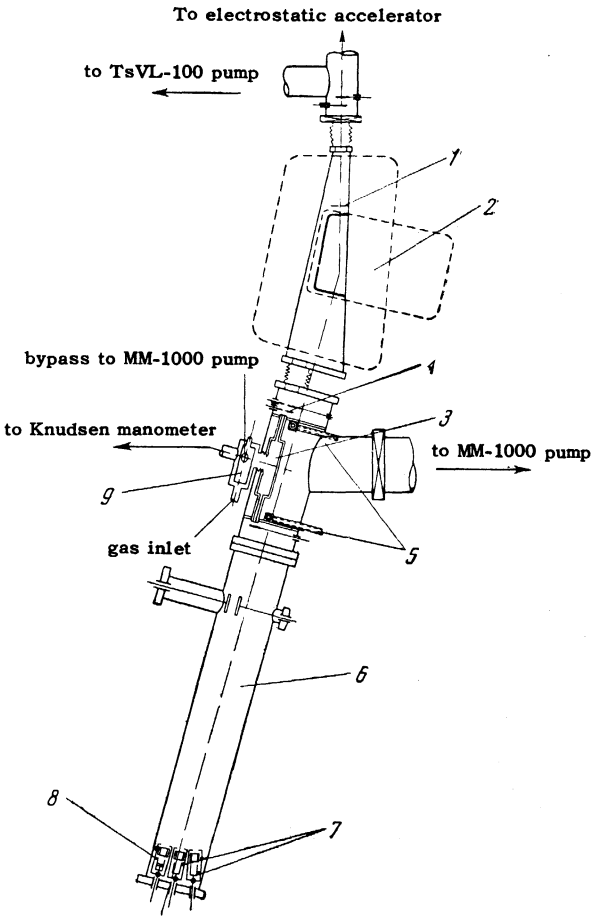

FIG. 1. Diagram of apparatus.

emerged through a channel of the same dimensions. The collision chamber, of effective length 310 mm, was fastened to two supports 5 which were regulated by means of vacuum-sealed screw rods for accurate positioning of the chamber with respect to the ion beam. After traversing the collision chamber the beam entered an electrostatic analyzer 6, which consisted of a copper tube 200 mm in diameter and 1000 mm long containing the plates of a flat condenser.

The electric field of the condenser separated

the beam into a neutral component and H^+ and H_2^+ components. The currents of positive components were measured by vacuum-tube electrometers connected to Faraday cups 7. The intensity of the neutral beam formed from H_2^+ was measured by a thermocouple detector 8 similar to that described by Damodaran.[1] The thermocouple emf was measured by an M-21/4 mirror galvanometer. All currents were measured simultaneously in order to reduce the error resulting from fluctuations of the primary beam intensity. For the purpose of reducing the background and contaminations, the collision chamber was sealed by means of lead gaskets, and a liquid-nitrogen trap 9 was introduced to freeze condensing fractions. An auxiliary vacuum duct and valve accelerated the exhaustion of residual gas and gas liberated from the chamber walls as they were heated to 100 — 120° C.

The residual gas pressure in the collision chamber did not exceed $(4 - 5) \times 10^{-6}$ mm Hg. Gas was exhausted at the entrance and outlet of the collision chamber by an MM-1000 pump and liquid-nitrogen trap. The pressure in the space surrounding the collision chamber and in the analyzer did not exceed 3×10^{-6} mm. The accelerating tube and chamber of the mass monochromator were exhausted by a TsVL-100 pump and liquid-nitrogen trap. During the experiment the pressure at the accelerating-tube exit and in the mass monochromator varied from 7×10^{-6} to 1.2×10^{-5} mm. Gas pressure in the collision chamber was measured by a Knudsen manometer calibrated against a McLeod manometer.

Total cross sections for proton production (σ_{H^+}) and for the production of fast hydrogen atoms (σ_{H^0}) were measured mass-spectrometrically. The following formulas were used in the calculations:

$$\sigma_{H^+} = \left\{\frac{d}{d(nL)}\left[2N_{H^+} / (N_{H^+} + N_{H^0}) + 2N_{H_2^+}\right]\right\}_{nL\to 0}, \quad (1)$$

$$\sigma_{H^0} = \left\{\frac{d}{d(nL)}\left[2N_{H^0} / (N_{H^+} + N_{H^0}) + 2N_{H_2^+}\right]\right\}_{nL\to 0}. \quad (2)$$

Here N_{H^+}, N_{H^0}, and $N_{H_2^+}$ are the numbers of protons, hydrogen atoms, and molecular hydrogen ions, respectively, traversing the collision chamber after subtraction of the particles produced in the residual gas and at the edges of apertures; n is the concentration of target gas molecules, and L is the effective length of the collision chamber with adjacent segments of the beam path taken into account.

For the purpose of checking on the occurrence of single collisions we recorded the ratios of secondary to primary particles as functions of the gas pressure in the collision chamber. With these ratios extrapolated to zero pressure, σ_{H^+} and σ_{H^0} were determined from (1) and (2).

Since the cross sections were measured using thin targets, σ_{H^+} could be determined neglecting the effect of the $H_2^+ \to H_2^0$ process even at low energies. The result obtained for σ_{H^0} can be too high because H_2^0 molecules resulting from charge exchange of H_2^+ reach the detector along with H^0 atoms. However, the cross section for electron capture by molecular hydrogen ions is very small at 200 kev and falls off very rapidly with increasing primary ion energy. Sweetman's data[4] for energies above 250 — 300 kev show that the systematic error resulting from this process is small. For these reasons, and assuming also that the cross sections for processes leading to the production of negative hydrogen ions are negligibly small at the given energies, we find that the total dissociation cross section is determined practically by the following three processes:

$$\text{I. } H_2^+ \to H^+ + H^0,$$
$$\text{II. } H_2^+ \to H^+ + H^+,$$
$$\text{III. } H_2^+ \to H^0 + H^0.$$

The total dissociation cross section σ_d is therefore given by

$$\sigma_d = (\sigma_{H^+} + \sigma_{H^0}) / 2. \quad (3)$$

The foregoing considerations regarding the smallness of the cross section for $H_2^+ \to H_2^0$ can also be applied to process III at energies beginning with 400 — 500 kev. In the range 500 — 1200 kev we can therefore assume without large error that the total dissociation cross section is the sum of the cross sections for processes I and II; the corresponding partial cross sections σ_I and σ_{II} can be evaluated accordingly. The partial cross section σ_I is then given directly by σ_{H^0}, while σ_{II} is determined from

$$\sigma_{II} = (\sigma_{H^+} - \sigma_{H^0}) / 2. \quad (4)$$

Diaphragm openings of different diameters at the collision-chamber exit were used to check for unequal scattering of primary and secondary particles. With openings of diameters greater than 4 mm the ratio of secondary to primary particles remained constant within the limits of error. Since the exit channel of the collision chamber had a 6.5-mm diameter, only a small systematic error resulting from unequal scattering is thus indicated. We estimated random errors of ± 12% for σ_{H^+} and ± 15% for σ_{H^0}. The energy of molecular hydrogen ions was determined from the accelerating voltage of the electrostatic

generator, measured by a rotary voltmeter that had been calibrated by means of γ-ray resonances. The error of primary ion energies does not exceed ± 2%.

3. RESULTS AND DISCUSSION

The target gases were hydrogen passed through a palladium filter, 99.97% pure nitrogen, and helium, argon, and krypton with at most 0.1% impurities.

The curves in Figs. 2, 3, and 4 represent the cross sections σ_{H^+} for proton production, and σ_d for hydrogen dissociation as functions of H_2^+ energy. Values of σ_{H^+} and σ_d given by other investigators are also shown. Figs. 2 — 4 show that over the entire investigated energy range σ_{H^+} and σ_d decrease steadily with increasing energy; σ_d decreases more rapidly than σ_{H^+}. The cross sections are enhanced with increased atomic number of the target gas, in agreement with the observations of other investigators. Besides the higher absolute values of σ_{H^+} and σ_d, in heavy gases the cross sections fall off less rapidly with increasing H_2^+ energy.

It is interesting to compare our results with those obtained by other workers. For hydrogen our data agrees satisfactorily with those of Sweetman,[4] are close to those of Barnett at relatively low energies,[3] and join only poorly at 200 kev with those of Damodaran[1] and Fedorenko et al.[2]

Our values for σ_{H^+} and σ_d in the cases of nitrogen and argon differ by 15 — 60% from the cross sections measured by Damodaran and by Barnett. Also, our curves for σ_d in the range 500 — 1200 kev fall off more rapidly with increasing energy than those of Barnett.

Considerably greater discrepancies between our data and those of other investigators were observed in the case of helium, for which our values of σ_{H^+} and σ_d were about 1.5 — 2 times smaller than the results given in references 1 — 3. The cause for the disagreement could lie in different degrees of purity of the helium in the collision chamber, since small admixtures of heavier gases can obviously result in highly exaggerated measurements of H_2^+ dissociation cross sections in helium.

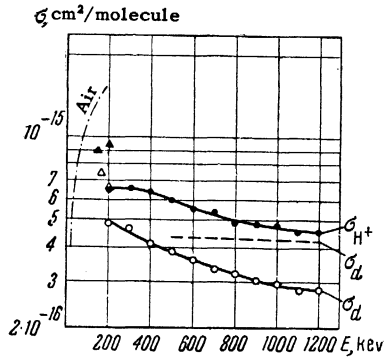

FIG. 3. Cross sections for H_2^+ dissociation and proton production in nitrogen. Solid curves – present work; Δ–σ_d, ▲–σ_{H^+} – reference 1; dot-dash curve – reference 2; dashed curve – reference 3.

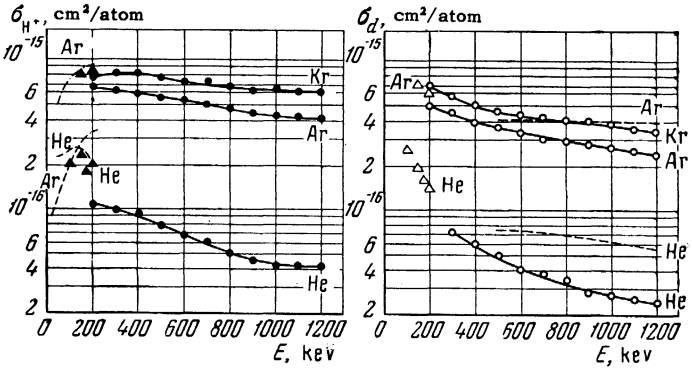

FIG. 4. Cross sections for H_2^+ dissociation and proton production in helium, argon, and krypton. Solid curves – present work; Δ, ▲ – reference 1; dot-dash curves – reference 2; dashed curves – reference 3.

The partial cross sections for processes I and II in the range 500 — 1200 kev were evaluated (see the table) from our data on the basis of the foregoing considerations regarding the smallness of the $H_2^+ \rightarrow 2H^0$ cross section at high energies. In all of the investigated gases σ_{II} decreases slowly and monotonically as the H_2^+ energy is increased. σ_I decreases considerably more rapidly in the same energy range.

Our calculated partial cross sections σ_I and σ_{II} can be compared with Sweetman's direct

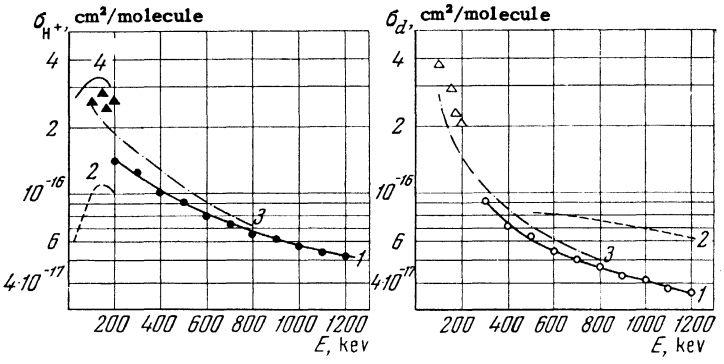

FIG. 2. Cross sections for H_2^+ dissociation and proton production in hydrogen: 1 – present work, 2 – reference 3, 3 – reference 4, 4 – reference 2, Δ, ▲ – reference 1.

Energy, kev	H_2		N_2		He		Ar		Kr	
	σ_I^*	σ_{II}	σ_I	σ_{II}	σ_I	σ_{II}	σ_I	σ_{II}	σ_I	σ_{II}
500	3.7	2.8	17	21	2.3	2.6	18	19,5	23	25
600	3,0	2.5	16	20	1.8	2.3	17	18	20	24
700	2.6	2.4	14	20	1.6	2.2	14	18	18	25
800	2.5	2.2	13	18	1.4	2.0	12	18	17	25
900	2.2	2.1	12	18	1.2	1.7	11	17	16	25
1000	2.2	1.9	11	17	1.1	1.6	10.5	16	15	25
1100	2	1.8	9	17	1.0	1.6	10	15	13	24
1200	1.9	1.7	9	16	0.9	1.55	9.5	15	12	24

*Cross sections are given in units of 10^{-17} cm²/molecule

measurements[4] only in the case of hydrogen. Our values are consistently lower than Sweetman's results by 15 — 20%, which is approximately the same discrepancy that occurred for the total cross sections σ_{H^+} and σ_d.

Salpeter[6] has performed the only theoretical calculation of the dissociation cross sections of fast molecular hydrogen ions in collisions with gas molecules, employing the Born approximation for high H_2^+ energies. Our cross sections for H_2^+ dissociation in hydrogen at 300 — 400 kev agree satisfactorily with Salpeter's calculations. We used energies that are too low to permit a comparison between the theoretical and experimental cross sections for H_2^+ dissociation in N_2 and Ar.

In conclusion we wish to thank Academician A. K. Val'ter of the Academy of Sciences, Ukrainian S.S.R., for his interest in this work, and Ya. M. Fogel' for a discussion of the results.

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