

Dissociation of H_2^+ Ions in Collision with H Atoms: 3 to 115 keV*

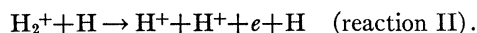
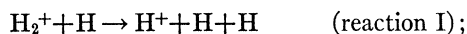
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The cross section for the production of fast protons in collisions between H_2^+ ions and H atoms has been measured in the energy range 3 to 115 keV. The cross section for the same process with H_2 as the target particle was also measured and results were compared with previous work as a test of the calibration method. The results show that the H-atom target is more effective in dissociating H_2^+ than is the larger H_2 molecule in the energy range 10 to 42 keV. This order of effectiveness is reversed below 10 keV and above 42 keV. Comparison of the H-target results with theoretical calculations based on the Born approximation shows that the collisional excitation of the lowest excited state of the H_2^+ ion—the $2p\sigma_u$ state—contributes a minor fraction of the total fast-proton production at 115 keV and a major fraction at 3 keV. The differential angular distributions of the fast protons were found to be the same for the H and H_2 targets at 10 keV.

COLLISIONS between fast H_2^+ ions and H atoms can cause dissociation of the former species to yield one or two protons in the following ways:



The present paper reports the results of measurements of the fast proton yield from reactions I and II, which we denote $\sigma_{H^+}(1)$, in the energy range 3 to 115 keV. The measured quantity equals $\sigma_I + 2\sigma_{II}$ where σ_I and σ_{II} are the cross sections for reactions I and II, respectively.

Reactions similar to I and II in several gases other than atomic hydrogen have been studied previously by a number of investigators.¹⁻⁷ The cross section for fast proton production in H_2 gas, measured perhaps more often¹⁻⁷ than that in other gases, will be denoted in the present paper by $\sigma_{H^+}(2)$. This cross section was redetermined in the present work as a check on the experimental procedure. The angular distributions of the H^+ and H dissociation fragments of the H_2^+ projectile ions in H_2 gas have been examined by McClure.⁸

The Born approximation to the cross section for reaction I via the collisional excitation of the H_2^+ projectile to the $2p\sigma_u$ electronic state has been calculated by Peek⁹ for all initial vibrational states of H_2^+ and all final states of the H target atom. Other modes of excitation of H_2^+ leading to proton production have not been theoretically examined. It is hoped that the

present experimental results will provide a stimulus toward further theoretical work.

The method of measurement is identical to that previously used for the study of dissociation of H_2^+ in H_2 gas³ except that the target chamber was replaced by a tungsten chamber exactly like that used in the study of proton-H-atom charge exchange.¹⁰ As in the latter experiment, measurements of the Coulomb scattering of 20-keV protons and the production of H^- ions by double electron capture by 20-keV protons were used for calibrating the H-atom density and residual H_2 density in the heated target chamber.¹¹ A mass spectrometer upstream from the collision chamber selected a pure beam of mono-energetic H_2^+ ions for the dissociation experiment. Separation of the H^+ and H_2^+ ion beams emergent from the collision chamber was accomplished by electrostatic deflection plates located at the exit aperture of the collision chamber. The proportional counter used in previous experiments measured the H^+ beam and a Faraday cup collected the H_2^+ beam providing both a primary beam monitor and a means of controlling the stepwise motion of the detector slit across the H^+ beam. The scanning technique has been described in previous reports.^{3,10}

The exit aperture of the collision chamber and the effective detector aperture were of sufficient size that all protons produced within an angle of 44.6 milliradians from the H_2^+ beam axis were included in the measurement. At the extremes of the solid angle covered by the detector the differential intensity of the H^+ ions was always less than 10% of the central intensity. The correction for particles not received, as estimated by extrapolation, is believed to be less than 10%. The maximum pressure of H_2 or H gas in the collision chamber was sufficiently low (0.5×10^{-3} Torr) to insure that single collisions prevailed in the collision chamber. The gas pressures in the ion drift spaces upstream and downstream from the collision chamber

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⁸ G. W. McClure, Phys. Rev. 140, A769 (1965).

⁹ J. M. Peek, Phys. Rev. 140, A11 (1965).

¹⁰ G. W. McClure, Phys. Rev. 148, 47 (1966). Furnace Design similar to that of G. J. Lockwood and E. Everhart, Phys. Rev. 125, 567 (1962).

¹¹ This method of calibration is due to G. J. Lockwood, H. F. Helbig and E. Everhart, J. Chem. Phys. 41, 3820 (1964).

were about 3 orders of magnitude smaller than the collision chamber pressure. Hence no corrections for drift-space collisions were required.

The experimental procedure consisted of determining $\sigma_{H^+}(2)$ with the target chamber cold so that the target gas was entirely molecular. Then the ratio $\sigma_{H^+}(1)/\sigma_{H^+}(2)$ was determined in a manner precisely analogous to that employed in previous work on proton- H -atom charge exchange.¹⁰ These two determinations then allowed $\sigma_{H^+}(1)$ to be calculated. Since $\sigma_{H^+}(2)$ was an absolute determination based on Coulomb scattering to determine the target thickness, the $\sigma_{H^+}(1)$ result is also absolute.

Results for $\sigma_{H^+}(1)$ and $\sigma_{H^+}(2)$ are given in Table I. The tabulated values of $\sigma_{H^+}(2)$ agree within 10% with previous measurements made in this laboratory³ when the latter are corrected as indicated in Ref. 8 for large angle H^+ dissociation fragments lost in the original detector geometry. Cross section $\sigma_{H^+}(1)$ is somewhat less than $\sigma_{H^+}(2)$ at energies higher than 42 keV and at energies lower than 10 keV, but in the intermediate energy range $\sigma_{H^+}(1)$ exceeds $\sigma_{H^+}(2)$. The excess reaches a maximum of 10% at 20 keV. This result shows that in regard to capacity to induce collisional dissociation of the H_2^+ projectile, the H_2 molecule is by no means the equivalent of two separate H atoms. The molecule is, in fact, not as efficient as a single H atom at 20 keV.

Unfortunately there does not exist a complete theoretical treatment of all of the excitation processes contributory to the dissociation of H_2^+ in collision with an H target. There is, however, a Born approximation calculation performed by Peek⁹ of H_2^+ excitation from the ground state ($1s\sigma_g$) to the lowest excited state ($2p\sigma_u$). This state leads to dissociation via reaction I and is the lowest in energy of a sequence of discrete electronic states, all of which contribute to reaction I. (No calculation exists for σ_{II} in which the electron of the projectile is excited to a continuum of unbound states.) The Born calculation of Peek⁹ summed over the vibrational population of Dunn¹² is presented in Table I. It is apparent that the experimental total cross section is considerably larger than $2p\sigma_u$ cross section at 115 keV. However, at 3 keV the theoretical cross section approaches the experimental result.

It may be seen by reference to Fig. 5 of Peek's article⁹ that the effective cross section for $2p\sigma_u$ excitation is rather strongly dependent on the vibrational population of the H_2^+ ions in the Born approximation. This effect is greatly enhanced at energies below 5 keV.

¹² G. H. Dunn, J. Chem. Phys. 44, 2592 (1966).

TABLE I. Cross sections $\sigma_{H^+}(1)$ and $\sigma_{H^+}(2)$ for fast proton production by H_2^+ collisions with H and H_2 , respectively, versus energy E of H_2^+ projectile. Quantity $\bar{\sigma}(2p\sigma_u, \Sigma)$ is the theoretical Born cross section for excitation of H_2^+ to the lowest dissociative state averaged over an assumed vibrational population and summed over all final states of the H target atom. Unit of E : keV; unit of cross section: 10^{-16} cm².

E	$\sigma_{H^+}(2)$	$\sigma_{H^+}(1)$	$\bar{\sigma}(2p\sigma_u, \Sigma)$
3.15	2.18	1.61	1.43
4.0	2.28	1.77	1.30
5.0	2.28	1.91	1.16
6.3	2.35	2.14	1.01
8.0	2.43	2.32	0.88
10.0	2.52	2.46	0.75
12.5	2.48	2.57	0.67
16.0	2.27	2.42	0.58
20.0	2.10	2.33	0.53
25.0	1.91	2.12	0.49
31.5	1.83	1.98	0.47
40	1.85	1.89	0.47
50	1.95	1.75	0.47
63	2.11	1.82	0.46
80	2.28	1.73	0.44
100	2.34	1.58	0.42
115	2.34	1.51	0.40

It is likely that dissociation caused by direct vibrational excitation may contribute to both $\sigma_{H^+}(1)$ and $\sigma_{H^+}(2)$ at low energies; however, classical arguments suggest that this effect is small throughout the present energy range.

A subsidiary experiment was performed by a method described previously⁸ to compare the differential angular distribution of fast protons produced in H_2^+ dissociation on H and H_2 targets. Within a 10% experimental uncertainty the two distributions were identical and each agreed with a result previously obtained for protons produced in the $H_2^+ + H_2$ interaction.⁸ Distributions induced by both H and H_2 targets have a shape very similar to that calculated in Ref. 8 by a method which assumed a pure $2p\sigma_u$ final state of H_2^+ and an H atom target. This result, coupled with the above mentioned evidence that the $2p\sigma_u$ final state of H_2^+ is not a predominant contributor to the total proton production, leads to the conclusion that the other contributing final states have somewhat similar angular distributions.

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