

## Charge Exchange and Dissociation Cross Sections for $H_1^+$ , $H_2^+$ , and $H_3^+$ Ions of 2- to 50-keV Energy Incident Upon Hydrogen and the Inert Gases

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Within the energy range 2 to 50 keV, the cross sections for the single-electron capture by protons, for dissociation of  $H_2^+$  ions into  $H_1^+$  and  $H_1^-$  ions, and for dissociation of  $H_3^+$  ions into  $H_2^+$ ,  $H_1^+$ , and  $H_1^-$  ions, have been measured in hydrogen, nitrogen, and inert-gas targets. The dissociation cross sections were found to decrease by as much as 30% for changes in the operating conditions of the electrodeless discharge-type ion source. The dissociation cross sections were independent of changes in the oscillator frequency, the rf input power, and the dc ion extraction voltage. Changes in both the source gas pressure and transverse magnetic field cause variations in the mean energy and the energy spread of the positive ions extracted from the source; these variations have been related to the population of the vibrationally excited states of the primary molecular ions. The measured cross-section values are accurate relative to one another within 5%. It is shown that the measurements are not invalidated by effects such as competing capture and loss processes outside the collision cell, beam size, and dissociation-product scattering.

### 1. INTRODUCTION

FAST protons may be produced from the dissociation of fast  $H_2^+$  ions in single collisions with hydrogen molecules in which the  $H_2^+$  ion either loses an electron to form  $2H_1^+$ , or captures an electron to form  $H_2^0$  with an improbable dissociation to  $H_1^+ + H_1^-$ , or simply dissociates without electron transfer into  $H_1^0 + H_1^+$ . Early measurements of the total proton-production cross section<sup>1-8</sup> in the energy range of 5 to 300 keV differ by as much as a factor of 3. Subsequent studies<sup>9-16</sup> have sought an explanation of this difference mainly in (a) the state of excitation of the  $H_2^+$  ion prior to collision, and (b) instrumental effects such as the interception of some fast collision products by beam-defining apertures and the widening of the product ion beam after collision caused by the conversion of internal energy of

the  $H_2^+$  ion into kinetic energy of the dissociation products. There are two significant results. McGowan and Kerwin<sup>14</sup> have shown, using an electron velocity-selector-type ion source to control the populations of the excited states of the  $H_2^+$  ion, that each vibrational level not only has a different cross section for the dissociation collision, but that the total measured cross section corresponds to the sum of the cross sections for each state multiplied by the relative populations of that state. McClure<sup>17</sup> has measured the differential angular distributions of the  $H_1^+$  dissociation fragments and obtains good agreement with predicted values, based upon the Born-approximation calculations of Peek<sup>18</sup> for 10-keV  $H_2^+$  ions incident upon H atoms, when consideration is given to the internuclear spacings and orientations of the molecular ions. These results indicate that previous studies,<sup>12,16</sup> in which sufficient detail is given to enable a comparison to be made, would not have detected all of the dissociation fragments at low energies.

In the present experiments an apparatus has been set up and tested with primary proton and  $H_2^+$  beams and then used to investigate the effects of the operating conditions of an electrodeless discharge ion source and instrumental sources of error, upon the dissociation cross section of  $H_2^+$  ions for total proton production. Changes in the gas pressure, excitation frequency, input power, extraction voltage and magnetic field of the ion source were made over a range of ion energies from 2 to 50 keV to obtain an indication of their effect on the energy of the emergent  $H_2^+$  ions.

Subsequent measurements of the cross sections  $\sigma(H_2^+)$  and  $\sigma(H_1^+)$  from both  $H_3^+$  and  $H_2^+$  primary ions, for inert-gas targets of He, Ne, Ar, Kr, and Xe, have been made using those ion-source operating con-

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<sup>3</sup> N. V. Fedorenko, V. V. Afrosimov, R. N. Ilin, and D. M. Kaminker, *Zh. Eksperim. i Teor. Fiz.* **36**, 385 (1959) [English transl.: *Soviet Phys.—JETP* **9**, 267 (1959)].

<sup>4</sup> J. Guidini, *Compt. Rend.* **253**, 829 (1961).

<sup>5</sup> C. F. Barnett, *Proceedings of the Second United Nations International Conference on Peaceful Uses of Atomic Energy* (United Nations, Geneva, 1958), Vol. 32, p. 398.

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<sup>8</sup> K. K. Damodaran, *Proc. Roy. Soc. (London)* **239**, 382 (1957).

<sup>9</sup> A. P. Irsa and L. Friedman, *J. Chem. Phys.* **34**, 330 (1961).

<sup>10</sup> A. C. Riviere and D. R. Sweetman, *Proc. Phys. Soc. (London)* **78**, 1215 (1961).

<sup>11</sup> S. Kaplan, G. A. Paulikas, and R. V. Pyle, *Phys. Rev. Letters* **7**, 96 (1961); *Phys. Rev.* **131**, 2574 (1963).

<sup>12</sup> G. W. McClure, *Phys. Rev.* **130**, 1852 (1963).

<sup>13</sup> C. F. Barnett and J. A. Ray, in *Proceedings of the Third International Conference on the Physics of Electronic and Atomic Collisions, London, 1963*, edited by M. R. C. McDowell (North-Holland Publishing Company, Amsterdam, 1964), p. 743.

<sup>14</sup> J. W. McGowan and L. Kerwin, *Can. J. Phys.* **42**, 972 (1964).

<sup>15</sup> R. Caudano, J. M. Delfosse and J. Steyart, *Ann. Soc. Sci. Bruxelles* **76**, 127 (1963).

<sup>16</sup> S. E. Kupriyanov, N. N. Tunitskij and A. A. Perov, *Zh. Tekhn. Fiz.* **33**, 1252 (1963) [English transl.: *Soviet Phys.—Tech. Phys.* **8**, 932 (1964)].

<sup>17</sup> G. W. McClure, *Phys. Rev.* **140**, A769 (1965).

<sup>18</sup> J. M. Peek, *Phys. Rev.* **140**, A11 (1965).

ditions which were determined from the above experiments with  $H_2^+$  ions to give the largest values of  $\sigma(H_1^+)$ .

Further the cross section for total  $H_1^-$  ion production from the dissociation of fast  $H_2^+$  and  $H_3^+$  ions has been measured simultaneously with the total proton-production cross sections.

## 2. EXPERIMENTAL METHOD

The positive-ion beam is extracted from an electrodeless discharge ion source (described elsewhere<sup>19</sup>), accelerated and focused, and momentum analyzed by an inflection-type magnet<sup>20</sup> of 30-cm radius and  $\frac{1}{2}\pi$  deflection, giving an energy resolution of 1/108. The beam then entered the experimental region by passing through aperture  $A_1$  as shown in Fig. 1. The emergent beam from the collision cell was momentum analyzed by a conventional  $\frac{1}{2}\pi$  deflection, 10-cm-radius magnet. The positively and negatively charged particles of the same mo-

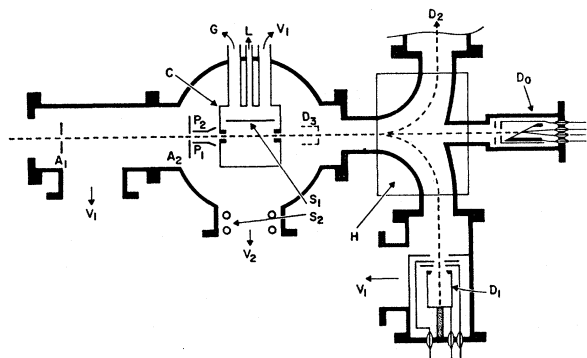


FIG. 1. Apparatus diagram. C, collision cell. G, ionization gauge.  $V_1$  and  $V_2$ , 250-liter/sec, 4-in. baffled diffusion pumps.  $A_1$  and  $A_2$ , beam-defining apertures.  $S_1$  and  $S_2$ , liquid-nitrogen-cooled surfaces.  $P_1$  and  $P_2$ , electrostatic deflection plates.  $D_0$ ,  $D_1$ , and  $D_2$ , secondary electron-type detector and Faraday cups. L, variable gas leak. H, beam-analyzing magnetic field with a mean beam radius of 10 cm.

mentum could be collected simultaneously in Faraday cups while the uncharged particles were detected by a secondary electron-emitting-type detector, similar to that used by Gardon.<sup>21</sup> The detector currents were measured with electrometers covering the range from  $10^{-6}$  to  $2 \times 10^{-15}$  A.

It is readily shown<sup>22,23</sup> that the growth of the number  $N_f$  of atoms of charge state  $f$  within a beam, composed initially of  $N_i$  particles of charge state  $i$  as it passes through a target gas, is given by

$$\Delta N_f [N_i \Delta(nl)]^{-1} = \sigma_{if} + a_{if}(nl) + b_{if}(nl)^2 + \text{higher powers of } (nl), \quad (1)$$

<sup>19</sup> J. F. Williams, Rev. Sci. Instr. **37**, 1205 (1966).  
<sup>20</sup> L. Kerwin, Rev. Sci. Instr. **20**, 36 (1949).  
<sup>21</sup> R. Gardon, Rev. Sci. Instr. **24**, 366 (1953).  
<sup>22</sup> Y. M. Fogel, Usp. Fiz. Nauk **71**, 243 (1960) [English transl.: Soviet Phys.—Usp. **3**, 90 (1960)].  
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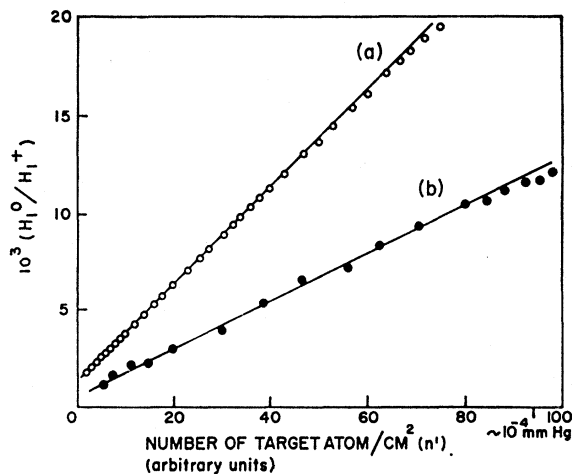


FIG. 2. Typical curves showing the growth of  $H_1^0$  from (a) a 10-keV  $H_1^+$  beam in hydrogen and (b) a 2-keV  $H_1^+$  beam in argon as a function of the relative target gas number density  $n'$ .

where  $(nl)$  is the equivalent number of gas atoms in the collision cell per  $cm^2$  of target area normal to the primary beam direction,  $\sigma_{if}$  is the cross section for the collision in which a particle of charge  $ie$  is changed to a particle of charge  $fe$ ,  $a_{if}$  and  $b_{if}$  are functions of all the possible  $\sigma_{if}$  for the colliding atoms. The method of measuring the cross section  $\sigma_{if}$  which is used in the present experiments, readily follows from Eq. (1). In principle one studies the dependence of the growth of  $N_f$  from a constant  $N_i$  on the quantity  $(nl)$ . This dependence is seen to be linear, that is the changes of charge of the primary atoms result only from single collisions, when  $a_{if}(nl)/\sigma_{if} \ll 1$ . Since this condition of linearity of  $\Delta N_f/\Delta(nl)$  is dependent upon the nature of the primary and target atoms and their relative velocity, every individual cross section is to be measured by plotting  $\Delta N_f/N_i$  against  $\Delta(nl)$  to ensure the separation of the linear and parabolic components.

The  $N_i$  and  $N_f$  are readily measured absolutely by charge integration methods with electrometers. The absolute measurement of  $(nl)$  is difficult. Because molecular-flow conditions are well satisfied in the collision chamber  $(nl) = cn'$  where  $n'$  is the target gas number density as measured by an ionization gauge and  $c$  is a calibration constant which has a fixed value for a given target gas and a given statistical distribution of primary particle paths through the collision cell. It is assumed that the distribution of beam paths is independent of the type and energy of the primary beam used in the present experiments. Equation (1) can now be written in the form

$$\sigma_{if} = \Delta N_f |N_i \Delta(nl)|^{-1} \quad \text{to a first approximation} \\ = \Delta N_f |N_i c (\Delta n')|^{-1} \quad (2)$$

from which the cross section can be determined.

Figure 2 shows typical examples of the growth of  $N_f$  with  $n'$ . The large number of experimental points such as shown in curve (a) was taken only during the early

measurements of a given  $\sigma_{if}$  in each target gas. In all  $\sigma_{if}$  determinations, the ratio  $N_f/N_i$  rarely exceeded 1% and  $n'$  was varied by at least a factor of 50. The error in determining  $\Delta N_f/\Delta(n')$  from Fig. 2 is approximately 2%, composed of (a) an error not greater than 0.7% in measuring  $N_i$ ,  $N_f$  and  $n'$ , and (b) an error of not greater than 1% in fitting a straight line to the experimental data.

The constant  $C$  in Eq. (2) was determined by measuring the conversion of  $H_1^+$  to  $H_1^0$  by single-electron capture in each target gas used and assuming the cross section value for that process,  $\sigma_{10}$ , at a given energy.<sup>12,24</sup> The energy of 10 keV was selected for this standardization procedure because there are a large number of experimental measurements published which appear to have their best agreement at that energy. Further, as most of the accuracy tests, described in Sec. 4, were performed at 10 keV, the results at that energy are the most reliable of the present series. The value of  $8.2 \times 10^{-16}$  cm<sup>2</sup>/molecule has been used for  $\sigma_{10}$  at 10 keV in hydrogen gas.

### 3. EXPERIMENTAL ACCURACY AND VALIDITY

The accuracy and validity of the cross section measurements have been carefully investigated with particular attention being given to the considerations raised by Fogel,<sup>22</sup> Allison,<sup>23</sup> and Stier and Barnett.<sup>25</sup>

It was shown by tests at beam energies of 2, 10 and 50 keV that:

(a) Each of the detectors  $D_0$ ,  $D_1$ , and  $D_2$  gave identical responses to the same beam. Interchange of detectors had no detectable effect on the beam profiles.

(b) Normal variations, in the gas pressure, of either the target chamber or the ion source had no effect upon the response of the detectors. Nor did fluctuations in the analyzing magnetic field.

(c) Any scattering of the primary beam by collision with residual gas molecules along the beam path was shown to have a negligible broadening effect on the beam.

Initial tests and measurements have been conducted with a proton beam so that the results could be checked against well-known data.

The primary  $H_1^+$  beam will always contain some  $H_1^0$  and  $H_1^-$  particles which may be produced from the primary beam by collision with the edges of the beam defining apertures  $A_1$  and  $A_2$ , the walls of the entry and exit canals, or the residual gas molecules along the beam path between the aperture  $A_1$  (before which the beam follows a curved path) and the entry canal of the collision cell. The influence of such effects was minimized by using knife-edged apertures and a beam diameter considerably less than most aperture diameters. All apertures were aligned optically on assembly. Fortu-

<sup>24</sup> W. L. Fite, R. F. Stebbing, D. G. Hummer, and R. T. Brackman, *Phys. Rev.* **119**, 663 (1960).

<sup>25</sup> P. M. Stier and C. F. Barnett, *Phys. Rev.* **103**, 896 (1956).

nately, the numbers of  $H_1^0$  and  $H_1^-$  so formed are independent of the collision cell gas pressure. Effectively all of the  $H_1^0$  and  $H_1^-$  will pass through the collision cell because, in the present "single-collision" experiments, not more than 1% of the primary beam undergoes a charge-changing collision in the collision cell. Since the method of determination of a cross section is to measure the linear rate of growth of collision products with pressure, the number of  $H_1^0$  and  $H_1^-$ , which are produced in the above manner and which are pressure independent, will appear as a constant term which does not influence the slope of the collision product versus pressure graph. For the present measurements this constant term did not exceed 10% of the total collision products and was generally much smaller.

These  $H_1^0$  and  $H_1^-$  particles in their traversal through the collision cell may collide with the target gas atoms to give rise to linearly pressure-dependent collision products which are indistinguishable from those formed from the true primary beam. These spurious contributions have been analyzed as follows:

(a) The errors arising from the neutral atoms, which are formed in the residual gas or on the edges of the beam defining apertures, but not those from the walls of the entry canal, are simply determined by electrostatic deflection of all charged particles from the beam prior to their entry to the gas cell. Thus the remaining "primary" beam is now a neutral-atom  $H_1^0$  beam from which the collision products  $H_1^+$  and  $H_1^-$  may be observed as a function of the gas pressure. The fraction of such  $H_1^-$  impurities as compared with those  $H_1^-$  ions produced from the  $H_1^+$  will thus depend upon (i)  $\sigma_{10}$  for  $H_1^+$  in the residual gas of the vacuum system and (ii) the relative values of  $\sigma_{1,-1}$  and  $\sigma_{0,-1}$  in a given target gas. This type of investigation has been made for every cross section measurement. Table I shows typical values of the detected currents for the case of 10-keV  $H_1^+$  in hydrogen gas, for which the neutral-atom impurities discussed above give rise to a 0.5% error in  $\sigma_{1,-1}$ .

(b) The negative ions, formed in the primary beam before the electrostatic deflector plates, may lose electrons to form  $H_1^0$  or  $H_1^+$ . This error cannot be determined in the manner of paragraph (a) because the the electric field will deflect the  $H_1^-$ . The fraction of  $H_1^-$  present in the beam is so small, however ( $\sim 2 \times 10^{-6}$

TABLE I. Composition of charged and neutral ion beams product by a 10-keV  $H_1^+$  beam in hydrogen at typical target gas pressures.

Primary	Target gas pressure in mm Hg	Detector current in A		
		$H_1^+$	$H_1^0$	$H_1^-$
Charged plus neutral	$0.25 \times 10^{-5}$	$9.0 \times 10^{-7}$	$1.9 \times 10^{-10}$	$1.70 \times 10^{-12}$
	$8.2 \times 10^{-5}$	$8.93 \times 10^{-7}$	$6.1 \times 10^{-9}$	$3.13 \times 10^{-11}$
	$0.25 \times 10^{-5}$	$2.9 \times 10^{-14}$	$6.5 \times 10^{-11}$	$2.0 \times 10^{-15}$
Neutral only	$8.2 \times 10^{-5}$	$5.6 \times 10^{-13}$	$6.5 \times 10^{-11}$	$1.6 \times 10^{-13}$

of the primary  $H_1^+$  beam), that an increase in the fractions of  $H_1^+$  or  $H_1^0$  would not be detectable.

(c) An estimate of any errors arising from those neutral atoms which may be formed on the entry canal was made by increasing the canal diameter from 1.5 to 2.0, then 3.0 mm (for a beam diameter of 1.0 mm). The minimum gas pressure within the cell remained unchanged since the cell had a separate pumping line. This increase of entry canal diameter had no detectable effect upon the results presented in Table I. Therefore an upper limit may be placed upon the number of neutrals formed on the entry canal walls of 2% of the number of  $H_1^0$  formed by collision of the primary  $H_1^+$  with the residual gas along the beam path. Less than 1% of these surface-formed  $H_1^0$  atoms will suffer a collision with the target gas so that the fraction of  $H_1^-$  ions formed from these  $H_1^0$  will be negligible compared with that formed from the  $H_1^+$  primaries in collision with the target gas at its minimum pressure.

A study of the detector response for a given beam as a function of the analyzing magnetic field indicated that the possibility of collisions with the walls of the exit canal was very small for those primary particles which had changed their charge by collision with the target gas. This possibility was further investigated by increasing the exit canal diameter from 2.0 to 2.5, 3.5, and 10 mm. For a constant minimum gas pressure in the collision cell, these changes in the exit-canal diameter had no detectable (i.e., less than 0.5%) effect on the collected currents of all charge states.

Also when the value of ( $nl$ ) of the target gas increases within the collision cell, there will be a further very small increase in ( $nl$ ) which originates from the efflux of the target gas through the entry and exit canals. This increase in ( $nl$ ) will add to that within the cell but causes no error in the relative values of the present cross sections.

For a 10-keV  $H_1^+$  beam emerging from the collision cell (in which the gas pressure was about  $10^{-4}$  mm Hg) identical beam currents were collected by a Faraday cup placed alternately at position  $D_3$ , which was 4 cm after the exit canal of the collision cell, and at position  $D_0$ . There is therefore a negligible loss of primary beam particles between the exit canal and the detectors. This result may naturally be extended to the beams of collision products which emerge from the collision cell. The gas pressure in the region between the collision cell and the detectors was measured to be the same as that pressure near the beam path before the gas cell. For the case of 10-keV  $H_1^+$  traversing the background gas, there is a approximately a 0.01% loss of protons from the primary beam.

When the primary beam is made up of  $H_2^+$  or  $H_3^+$  ions further considerations become necessary in view of the additional internal motions possible.

For molecular-ion beams the possibility arises that the internal energy of the molecules may change in a collision to give rise to a transverse velocity component

to its constituent atoms and hence broaden the beam. The broadening effect of any given transverse velocity on an ion beam will naturally be largest when the ion is travelling at its slowest forward velocity which occurs for the 2-keV  $H_2^+$  beam in this present work. Therefore the currents of collision products  $H_1^+$  and  $H_1^-$  from a 2-keV  $H_2^+$  beam traversing xenon were examined as a function of the exit-canal diameter. Canal diameters of 2.0, 2.5, 3.0, 5.0 and 10.0 mm were used. The collected  $H_1^+$  and  $H_1^-$  beam currents were independent of canal diameters which were equal to or larger than 3.0 mm. Therefore an exit-canal diameter of 3.0 mm has been used for all  $H_2^+$  charge changing cross section measurements. With this additional divergence of the beam, a detector aperture diameter of 12 mm was found to be adequate<sup>26</sup> to collect all the collision products of both  $H_1^+$  and  $H_1^-$ .

Measurements of the  $H_1^+$ ,  $H_1^-$ , and neutral-particle products for (i) maximum and minimum target gas pressures and for (ii) a large voltage between the deflector plates  $P_1$  and  $P_2$  of Fig. 1 with a 10-keV  $H_2^+$  primary beam incident on hydrogen gas, show that not more than 0.01% of the primary  $H_2^+$  ions were neutralized or dissociated on the edges of the beam defining apertures  $A_1$  and  $A_2$  (or Fig. 1). Such a percentage would have a negligible effect upon the measured cross sections.

Those error sources which depend upon cross-section values will naturally change when the projectile changes from protons to the  $H_2^+$  molecular ion and the number of possible beam components increases from 3 to 5. Experiment shows that such errors in  $\sigma(H_1^+)$  are negligible and in  $\sigma(H_1^-)$  amount to about 2.4%. This error in  $\sigma(H_1^-)$ , which is due to impurity atoms in the primary  $H_2^+$  beam, has been estimated and allowed for in all  $\sigma(H_1^-)$  measurements since it is a simple procedure to apply the deflection voltage  $V$  between plates  $P_1$  and  $P_2$ .

Considerations similar to those above for primary  $H_2^+$  ions were made for primary  $H_3^+$  ions. The  $H_2^+$  and  $H_1^+$  dissociation fragments were not detected simultaneously but collected alternately in the same Faraday cup. This procedure was shown to introduce no inaccuracy into the measurements when a collision-cell exit-canal diameter of 3.0 mm and a detector-entrance diameter of 22 mm were used.

## 4. RESULTS

### (a) Primary $H_1^+$

The cross sections  $\sigma_{10}$  for 2–50 keV  $H_1^+$  incident upon hydrogen and the inert gases are shown in Fig. 3. For

<sup>26</sup> This result indicates an angular distribution of the  $H^+$  dissociation fragments that is narrower than that observed by McClure (Ref. 17), who has shown (private communication) that his data indicate that the present aperture should be too small to collect more than about 62% of the  $H^+$  dissociation fragments at 5-keV  $H_2^+$  energy. However, the data of Irsa and Friedman (Ref. 9) for  $H_1^+$  and  $D^+$  dissociation fragments from 4-keV  $HD^+$  ions, indicate nearly an order-of-magnitude smaller angular spread than that observed by McClure.

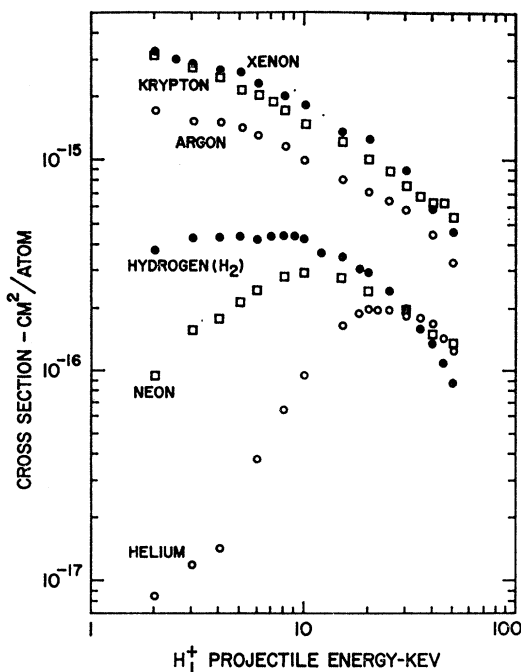


FIG. 3. Cross section  $\sigma_{10}$  for  $H_1^+$  ions incident on  $H_2$ , He, Ne, Ar, Kr, and Xe. In each target gas the values of  $\sigma_{10}$  are normalized at 10 keV to those values given by Stier and Barnett (Ref. 25).

each target gas a similar standardization procedure was used, the standard values selected being those given by Stier and Barnett<sup>25</sup> at an incident proton energy of 10 keV. There is excellent agreement between their values and the present values in all cases except in krypton when, for proton energies less than 8 keV, the present values become larger than Stier and Barnett's values until at 2 keV there is a 20% difference. However, there are few measurements of  $\sigma_{10}$  in krypton and xenon, and the cross section values cannot be ascertained with any certainty.<sup>27</sup>

#### (b) $H_2^+$ Ions on Hydrogen

The dependence of the cross section for the production of protons, negative hydrogen ions, and neutral hydrogen atoms from  $H_2^+$  upon the ion-source operating conditions of gas pressure, transverse magnetic field, rf oscillator input power, and frequency and upon ion-

extraction voltage has been examined at energies for the  $H_2^+$  ions of 6, 20, and 50 keV. The ranges of value of rf oscillator frequency  $\omega$  (15 to 30 Mc/sec), the input power  $X_0$  (100 to 160 W), and the ion-extraction voltage  $V_s$  (500 to 3500 V), were limited by the equipment in use and the need to maintain a reasonable beam current. Within the available ranges, changes in  $\omega$ ,  $X_0$ , and  $V_s$  affected the cross sections observed by less than 5%. The changes in the cross sections with gas pressure and magnetic field are shown in Table II from which it is apparent that the cross sections increase as either the gas pressure or the transverse magnetic field decreased. At 6 keV the maximum variation in  $\sigma(H_1^+)$  was 30% decreasing to about 5% at 50 keV. The variations in  $\sigma(H_1^0)$  and  $\sigma(H_1^-)$  were comparable with those for  $\sigma(H_1^+)$  for all tests.

A detailed investigation of the electrodeless discharge ion source characteristics is reported elsewhere.<sup>19</sup> Beam deceleration techniques were used to measure the mean beam energy  $\bar{E}$  and the beam energy spread  $\Delta E$  of the total positive-ion beam extracted from the ion source as functions of the source gas pressure  $\rho$  and the steady transverse magnetic field strength,  $B$ . As either  $B$  or  $\rho$  decreases, then  $\bar{E}$  increases and  $\Delta E$  decreases. In spite of the fact that the total positive-ion beam extracted from the source contained ions of different  $e/m$  ratios (predominantly  $H_1^+$ ,  $H_2^+$ , and  $H_3^+$ ) whose relative numbers were found to vary considerably during the present experiments and that each ion species of a given  $e/m$  can be expected<sup>28</sup> to possess a value of  $\bar{E}$  different from that of the total ion beam, it is not unreasonable to assume that the observed behavior of  $\bar{E}$  and  $\Delta E$  of the total ion beam for changes in the ion source operating conditions is also characteristic of the behavior of  $\bar{E}$  and  $\Delta E$  of the electrons within the source discharge. Then changes in the mean electron energy within the source may be related to changes in the relative populations of the vibrationally excited states of the  $H_2^+$  ion as shown by McGowan and Kerwin.<sup>14</sup> From this sequence one can associate the observed changes in the measured cross sections with changes in the relative populations of the various vibrationally excited states of the primary  $H_2^+$  ions.

In the present experiments, the time interval between the formation of  $H_2^+$  and its collision with a target molecule was of the order of  $10^{-5}$  to  $10^{-6}$  sec, governed

TABLE II. Dependence of  $H_2^+$  dissociation cross sections on ion source pressure and magnetic field.  $\omega = 22.5$  Mc/sec,  $X_0 = 160$  W,  $V_s = 3$  kV.

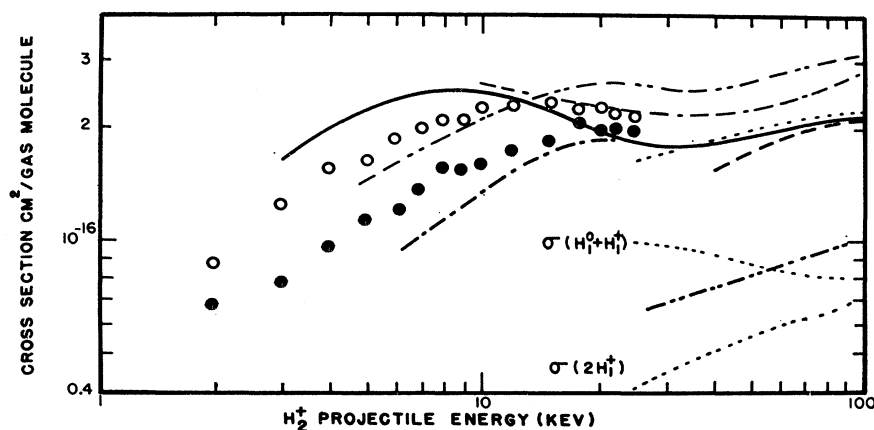
Energy of $H_2^+$ ions	6 keV						20 keV						50 keV					
	0		5.3		7.5		0		5.3		7.5		0		5.3		7.5	
Magnetic field in gauss	20	4	20	4	20	4	20	4	20	4	20	4	20	4	20	4	20	4
Pressure in $\mu$ Hg	1.46	1.88	1.32	1.68	1.20	1.40	2.13	2.33	2.03	2.27	1.92	2.12	2.08	2.24	2.08	2.20	2.06	2.12
$H_1^+$ , $10^{-16}$ cm <sup>2</sup> /molecule	2.16	1.92	1.01	1.56	1.16	1.53	7.8	9.7	8.3	9.4	4.2	6.9	8.4	9.5	9.0	7.9	7.6	8.2
$H_1^-$ , $10^{-19}$ cm <sup>2</sup> /molecule	4.48	5.22	4.22	4.82	4.02	4.45	7.2	8.4	6.9	8.0	6.65	7.6	6.1	6.3	6.1	6.28	6.0	6.28

<sup>27</sup> S. K. Allison and M. Garcia-Munoz, in *Atomic and Molecular Processes* edited by D. R. Bates (Academic Press Inc., New York, 1962), p. 751.

<sup>28</sup> P. C. Thoneman, Rept. Progr. Phys. 3, 92 (1953).

FIG. 4. The cross section for total proton production,  $\sigma(H_1^+)$ , from the passage of  $H_2^+$  ions through hydrogen for two ion-source pressures of  $4 \times 10^{-3}$  mm Hg ( $\circ \circ \circ$ ) and  $20 \times 10^{-3}$  mm Hg ( $\bullet \bullet \bullet$ ).

— McClure (Ref. 12);  
 - - - Schmid (Ref. 7);  
 - · - · Fedorenko (Ref. 3);  
 - · - · Fedorenko (Ref. 2);  
 - - - Barnett (Ref. 13);  
 - · - · Barnett (Ref. 5);  
 · · · Guidini (Ref. 4).



mainly by the time spent within the ion source. Taking into account the fact that there are no dipole transitions in  $H_2^+$ , while the intensity of quadrupole transitions is very low,<sup>29</sup> it can be assumed that in these experiments the initial vibrational excitation of the  $H_2^+$  ions within the source is practically unchanged until their collision with the target gas.

The known electronically excited states of the  $H_2^+$  ion<sup>30</sup> ( $2p\pi_u$  and  $3d\sigma_g$ ) are very weakly bound (0.25 and 1.36 eV, respectively) and the minima of the molecular potentials for these excited states are greatly displaced from the minimum of the  $H_2^+$  ion ground state. If such excited-state  $H_2^+$  ions were formed within the source, application of the Franck-Condon principle indicates that they would immediately dissociate.

A study of the ion source showed that the changes in source operating conditions which produced the maximum variation in  $\sigma(H_1^+)$  at 6-keV  $H_2^+$  energy also produced the maximum variation in  $\sigma(H_1^+)$  at 20 and 50 keV. Using those source operating conditions the two curves in Fig. 4 were obtained. From the work of Guidini,<sup>4</sup> who has made the only measurements of the partial cross sections  $\sigma(2H_1^+)$  and  $\sigma(H_1^+ + H_1^0)$  in this energy range, we may attribute the total cross section  $\sigma(H_1^+)$  almost entirely to  $\sigma(H_1^+ + H_1^0)$ . For  $H_2^+$  energies less than 15 keV, an extrapolation of the results of Guidini shows that  $\sigma(2H_1^+)$  is less than 5% of  $\sigma(H_1^+)$  so that the observed 30% variation in  $\sigma(H_1^+)$  is predominantly a variation in  $\sigma(H_1^+ + H_1^0)$ . At higher energies  $\sigma(2H_1^+)$  gradually increases in value relative to  $\sigma(H_1^+ + H_1^0)$  until, at 40 keV,  $\sigma(H_1^+ + H_1^0)$  is equal to  $2\sigma(2H_1^+)$ ; then the observed 5% variation in  $\sigma(H_1^+)$  may have originated in either  $\sigma(2H_1^+)$  or  $\sigma(H_1^+ + H_1^0)$ .

The present results permit a correlation of the observed "ion-source effects" of several workers. First, the result of Barnett and Ray,<sup>13</sup> that there was no dependence of  $\sigma(H_1^+)$  upon their electrodeless discharge ion source parameters of gas pressure and rf input power

for  $H_2^+$  energies from 40 to 200 keV, is not inconsistent with the present results since their experimental accuracy was 10% in the region where the present results show the variation of  $\sigma(H_1^+)$  to be  $5 \pm 3\%$ . Secondly, McClure<sup>12</sup> found, for 10-keV  $H_2^+$  energy, that variations in each of his Penning source parameters of kinetic energy of the ions emergent from the source and of gas pressure produced variations up to 20% in  $\sigma(H_1^+)$ . These variations are comparable with the present figures.

### (c) $H_2^+$ Ions and Inert-Gas Targets

The cross section  $\sigma(H_1^+)$  has been measured in the inert gases using primary  $H_2^+$  ions which were obtained from the ion source operating under those conditions which give the largest values of  $\sigma(H_1^+)$  in hydrogen gas.

Figure 5 shows  $\sigma(H_1^+)$  for 2–50 keV primary- $H_2^+$  ions incident upon the inert gases and nitrogen. Previous measurements of  $\sigma(H_1^+)$  have been made in He, Ne, Ar, and  $N_2$  by Fedorenko,<sup>2,3</sup> Guidini,<sup>4</sup> and Sweetman.<sup>6</sup> There are no published values for Kr and Xe.

In Ne, Ar, and  $N_2$  the present values are larger, and increasingly diverge as the  $H_2^+$  energy decreases from Fedorenko's 1954 data.<sup>2</sup> This divergence, which is also apparent in other results reported in the same paper, suggests that some of the larger-angle dissociation products may have been undetected.

In Ne and Ar, Sweetman's data<sup>6</sup> would appear to have the best agreement with the present results although the two sets of values have only a very small energy overlap.

In  $N_2$  there is a large range of values and no general agreement.

In those gases for which the partial cross section  $\sigma(H_1^+ + H_1^0)$  measurements of Guidini<sup>4</sup> are available, the present cross section  $\sigma(H_1^+)$  values would appear to be, at energies less than 20 keV, those for the simple "collision-induced" dissociation of the primary  $H_2^+$  ion into a proton and a hydrogen atom. At any given energy  $\sigma(H_1^+)$  increases with the size of the target atom.

<sup>29</sup> R. H. Hiskes, Phys. Rev. **122**, 1207 (1961).

<sup>30</sup> D. R. Bates, K. Ledsham, and A. L. Stewart, Phil. Trans. Roy. Soc. London **A246**, 215 (1953).

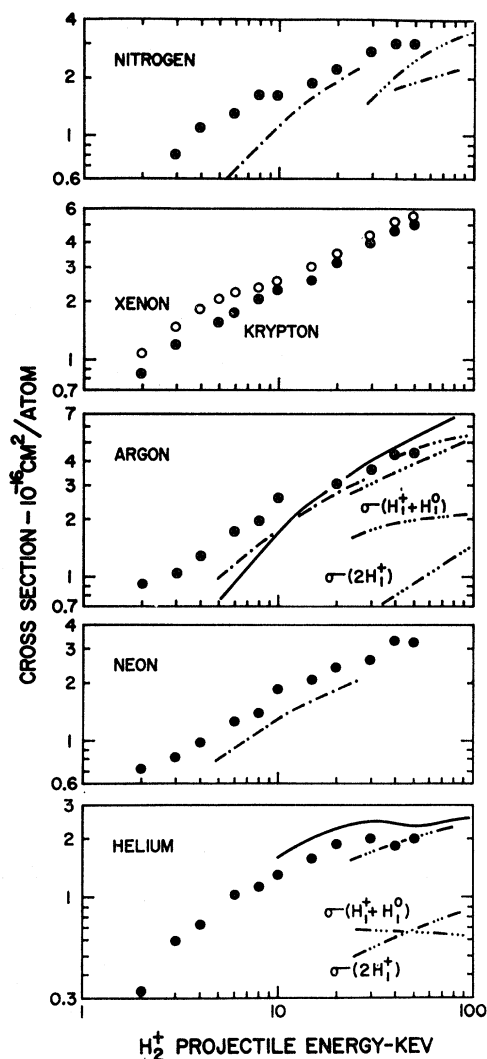


FIG. 5. The cross section for total proton production from the dissociation of fast  $H_2^+$  ions incident upon nitrogen and the inert gases as a function of the  $H_2^+$  ion energy.  $\bullet\bullet\bullet$  experimental values; — Fedorenko (Ref. 2); - - - Fedorenko (Ref. 3); - · - · - Sweetman (Ref. 6); - · - · - Guidini (Ref. 4). In helium and argon the partial cross sections,  $\sigma(2H_1^+)$  and  $\sigma(H_1^+ + H_1^0)$ , measured by Guidini, are given.

(d) Total  $H_1^-$ -Ion Production Cross Section,  $\sigma(H_1^-)$ , for  $H_2^+$  Ions

Figure 6 shows the total  $H_1^-$ -ion production cross section,  $\sigma(H_1^-)$ , for 2-50-keV  $H_2^+$  ions incident upon hydrogen gas for ion source gas pressures of 4 and 20  $\mu$

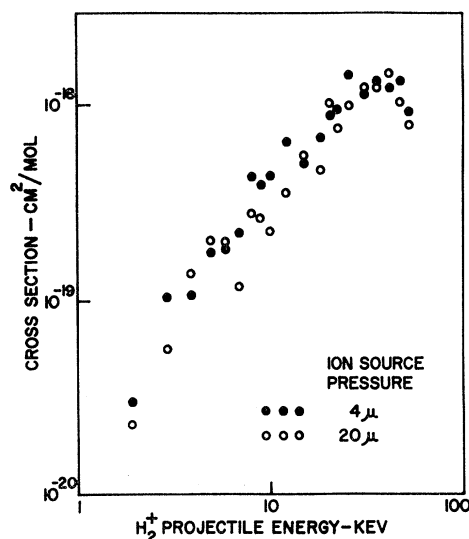
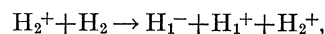


FIG. 6. Cross section,  $\sigma(H_1^-)$ , for the passage of  $H_2^+$  ions through hydrogen for ion source pressures of  $4 \times 10^{-3}$  mm Hg and  $20 \times 10^{-3}$  mm Hg.

Hg and other ion-source operating conditions identical to those used for obtaining the data of Fig. 4. In fact  $\sigma(H_1^-)$  was measured simultaneously with  $\sigma(H_1^+)$  for all ion source variations. Both cross sections exhibited a similar dependence upon the ion-source operating conditions.

The total  $H_1^-$  ion-production cross section is approximately the cross section for the collision in which the  $H_2^+$  ion captures one electron and dissociates into an ion pair, i.e.,



since other collisions involving the formation of fast  $H_1^-$  ions require the capture of more than one electron which is presumably a much less probable event. Changes in the relative populations of the vibrational levels of the  $H_2^+$  ions therefore influence the dissociation of  $H_2^+$  in an electron-capture collision as well as the simple "collision-induced" dissociation without electron capture as was shown from the total  $H_1^+$ -ion production cross section measurements.

(e) Primary  $H_3^+$  Ions

Table III shows the observed dependence of  $\sigma(H_2^+)$ ,  $\sigma(H_1^+)$ , and  $\sigma(H_1^-)$  upon ion source operating conditions

TABLE III. Dependence of  $H_3^+$  dissociation cross sections on ion source pressure and magnetic field.  $\omega = 16.7$  Mc/sec,  $X_0 = 160$  W,  $V_s = 3$  kV.

Energy of $H_2^+$ ions	5 keV						12 keV						20 keV					
	0		4.2		6		0		4.2		6		0		4.2		6	
Magnetic field in gauss	4	20	4	20	4	20	4	20	4	20	4	20	4	20	4	20	4	20
Pressure in $\mu$ Hg	3.0	5.2	2.7	5.0	2.4	4.6	12.4	10.0	11.8	9.5	10.6	9.3	15.0	13.1	14.1	12.5	13.5	12.7
$H_2^+$ , $10^{-17}$ cm <sup>2</sup> /molecule	3.7	3.1	3.5	2.9	3.2	2.7	9.3	7.5	8.8	7.1	8.1	6.8	13.8	11.8	12.8	11.0	12.0	10.4
$H_1^+$ , $10^{-17}$ cm <sup>2</sup> /molecule	4.2	3.1	2.15	2.0	2.5	2.35	9.6	6.8	5.8	5.5	7.2	4.5	13.0	7.4	8.0	11.5	9.8	9.5

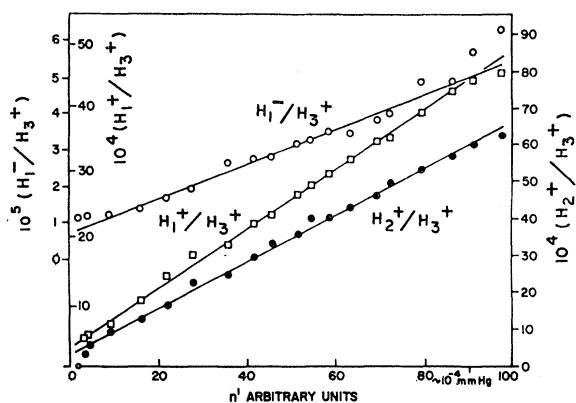


FIG. 7. Typical curves showing the growth of  $H_2^+$ ,  $H_1^+$ , and  $H_1^-$  ions from a 10-keV  $H_3^+$  ion beam in hydrogen as a function of the relative target-gas number density,  $n'$ .

for 5-, 12-, and 20-keV  $H_3^+$  ions incident upon hydrogen gas. From a similar argument to that given for primary  $H_2^+$  ions it is believed that the observed dependence of the  $H_3^+$  dissociation cross section upon the ion-source operating conditions is associated with a change in population of excited states of the  $H_3^+$  ions.

Figure 7 shows a typical set of curves for the growth of collision products with the target-gas number density for 10-keV  $H_3^+$  ions incident upon hydrogen gas, from which dissociation cross sections have been determined. The scatter of the experimental points for the  $H_1^-$  ions reduces the accuracy of  $\sigma(H_1^-)$  to about 30%.

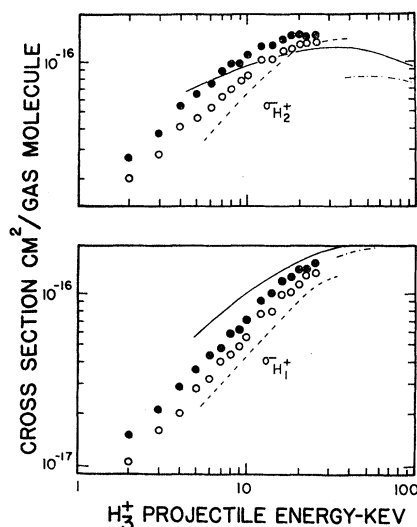


FIG. 8. The cross sections,  $\sigma(H_2^+)$  and  $\sigma(H_1^+)$ , for  $H_3^+$  ions incident upon hydrogen gas as a function of  $H_3^+$  ion energy for values of the ion source gas pressure of  $4 \times 10^{-3}$  mm Hg ( $\bullet \bullet \bullet$ ) and  $20 \times 10^{-3}$  mm Hg ( $\circ \circ \circ$ ). --- Fedorenko (Ref. 2); — McClure (Ref. 12); - - - - Barnett (Ref. 31).

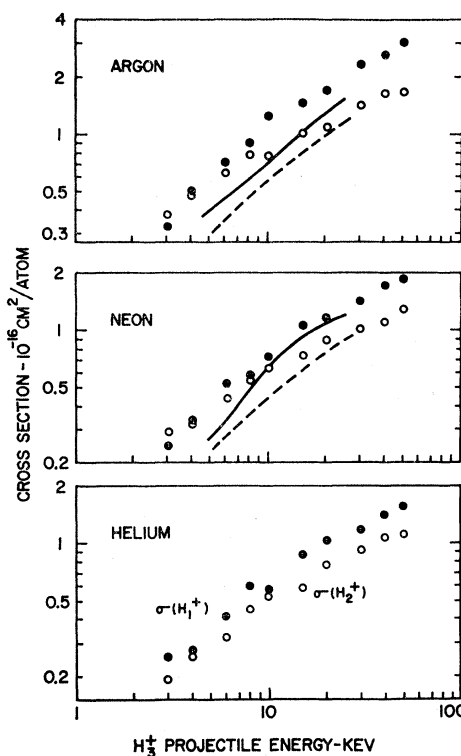


FIG. 9. The cross sections,  $\sigma(H_2^+)$  and  $\sigma(H_1^+)$  for  $H_3^+$  ions incident upon helium, neon and argon. The present experimental values are indicated by closed circles for  $\sigma(H_1^+)$  and by open circles for  $\sigma(H_2^+)$ , while Fedorenko's values (Ref. 2) are given by an unbroken line for  $\sigma(H_1^+)$  and a broken line for  $\sigma(H_2^+)$ .

The values of  $\sigma(H_2^+)$  and  $\sigma(H_1^+)$  for 2–50 keV  $H_3^+$  ions are shown in Fig. 8<sup>31</sup> for a hydrogen gas target and in Fig. 9 for helium, neon, and argon targets. Fedorenko's values are again generally lower than the present ones especially at lower energies suggesting, as before, that some of the dissociation products scattered through large angles may not have been collected. At high energies near 50-keV, where the present work indicates that the effect of vibrational excitation of the  $H_3^+$  upon its dissociation cross sections is small, there are differences of as much as a factor of two between the results of the various workers.

Little can be said about these cross sections because of their composite nature and the fact that there is meager information available about the structure and excited states of the  $H_3^+$  ion. Both  $\sigma(H_2^+)$  and  $\sigma(H_1^+)$  increase with the size of the target atom at all energies from 2 to 50 keV, and both increase monotonically with energy.

<sup>31</sup> C. F. Barnett, Oak Ridge National Laboratory Report No. 3113, 1962 (unpublished).