

### Merged-beam studies of the dissociative recombination of $H_3^+$ ions with low internal energy

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Cross sections for the dissociative recombination of  $H_3^+$  ions with low internal energy have been obtained using the merged-beam technique. Dissociative-excitation measurements have confirmed that only the  $v=0$  and 1 states were present in the ion beam. The resulting cross sections can be expressed in the form  $(2.17 \times 10^{-16})E_{c.m.}^{-0.57}$  ( $cm^2$ ) for  $E_{c.m.}$  below 0.03 eV and  $(3.23 \times 10^{-17})E_{c.m.}^{-1.15}$  ( $cm^2$ ) above.

The dissociative recombination of  $H_3^+$  is a process of great importance to the physical and chemical state of ionized-hydrogen plasmas. Such plasmas are encountered in planetary ionospheres, interstellar clouds, ion sources, and fusion reactors. When molecular hydrogen is ionized the  $H_2^+$  ions rapidly react with neutral  $H_2$  gas to form  $H_3^+$  via the reaction



For  $H_2^+(v=0)$  this reaction is exothermic by 1.68 eV.

The ionization of  $H_2$  by energetic electrons or photons, however, yields  $H_2^+$  ions with all 19 available vibrational states populated.<sup>1</sup> Since  $H_2^+$  has no dipole moment, the radiative lifetime of these states is  $\sim 10^6$  s (Ref. 2) so that in practice the exothermicity of reaction 1 will be greater. Experimental studies by Leventhal and Friedman<sup>3</sup> have shown that the bulk of this exothermicity appears in the form of internal excitation of the  $H_3^+$  ions which are formed with, on average, about 2 eV of internal energy. A calculation of the initial populations of individual vibrational states of  $H_3^+$  formed via reaction 1 has been performed by Anicich and Futrell<sup>4</sup> using a statistical approach and the results are tabulated in Table I. Unlike  $H_2^+$  however,  $H_3^+$  ions are susceptible to collisional deexcitation<sup>5</sup> so the populations of excited vibrational

states will decrease with time, the rate depending upon the collision frequency.

An important question for the modeling of hydrogen plasmas concerns the fate of the  $H_3^+$  ions formed via reaction (1). It has always been assumed<sup>6</sup> that  $H_3^+$  decays via dissociative recombination with electrons, i.e.,



A number of measurements of this process have been made using microwave-afterglow,<sup>7,8</sup> inclined-beams,<sup>9</sup> merged-beams,<sup>10,11</sup> and electron-beam-trap techniques.<sup>12</sup> These measurements all agreed with each other to within a factor of 2 and they all pointed to the process having a large room-temperature rate coefficient of  $\sim 2 \times 10^{-7} cm^3 s^{-1}$ . Measurements of the individual cross sections for reactions (2a) and (2b) were made by Mitchell *et al.*<sup>13</sup> who showed that reaction (2b) dominated reaction (2a) by about a factor of 2.

Theoretical studies of  $H_3^+$  recombination, however, indicated that for ions in low vibrational states, the recombination rate should in fact be quite small.<sup>14,15</sup> The reason for this can be seen from Fig. 1. The direct dissociative recombination of  $H_3^+$  must proceed via an initial

TABLE I. Calculated population distribution of the vibrational energy of  $H_3^+$  ions formed from reaction 1 prior to deexcitation (Ref. 4).

$v$	Population	Energy (eV)
0	0.0341	0.0
1	0.0919	0.372
2	0.1601	0.744
3	0.2197	1.116
4	0.2303	1.488
5	0.1387	1.860
6	0.0796	2.232
7	0.0299	2.604
8	0.0109	2.976
9	0.0037	3.348
10	0.0010	3.720
11	0.0001	4.092

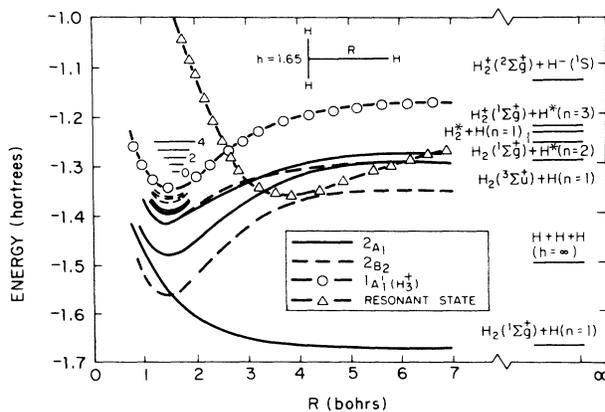


FIG. 1. Potential-energy curves for  $H_3^+$  and  $H_3$  in  $C_{2v}$  symmetry (reproduced with permission from Ref. 37).

capture into the lowest available  ${}^2A_1$  resonance state of  $H_3$  which is repulsive for low internuclear separations. In this way the potential energy of the system is rapidly converted to kinetic energy leading to a stabilization of the recombination. Although the asymptotic limit of the  ${}^2A_1$  state is actually  $H_2^+ + H^-$ , it has been shown<sup>16</sup> that, in fact, these products represent only a small fraction of the total recombinations. Curve-crossing transitions between this state and singly excited neutral states of  $H_3$  lead to the formation of neutral products.

It can be seen from Fig. 1 that the  ${}^2A_1$  state intersects the ground  ${}^1A_1'$  state of  $H_3^+$  at about 1 eV above the  $v=0$  level. It must be expected, therefore, that direct dissociative recombination of  $H_3^+$  in low vibrational states should be very slow and Michels and Hobbs (14) have argued that indirect dissociative recombination, involving initial capture into a vibrationally excited rydberg state followed by transition to the  ${}^2A_1$  resonance state and subsequent dissociation, is also unlikely because of the poor overlap.

This prediction was confirmed recently by studies of  $H_3^+$  recombination in a flowing afterglow apparatus.<sup>17</sup> It was found that  $H_3^+$  ions exhibited an initial rapid decay rate followed by a much slower decay, after about 0.5 ms. The reason for this rate change was interpreted as being due to the collisional deexcitation of higher vibrational states of  $H_3^+$  in the high-pressure (0.5–1.0 torr) environment of the afterglow. The  $H_3^+$  ions initially formed with high internal energy exhibit a large recombination rate but once deexcited, the rate diminishes dramatically. Adams *et al.* gave an upper limit to the recombination rate of  $2 \times 10^{-8} \text{ cm}^3 \text{ s}^{-1}$  at 95 K. More recently, however, this has been revised down to the very low figure of  $2 \times 10^{-11} \text{ cm}^3 \text{ s}^{-1}$ .<sup>18</sup> These results have stirred considerable controversy not only because they disagree with previous measurements but also because they require a dramatic reexamination of models of physical systems such as the Jovian atmosphere,<sup>19</sup> and interstellar chemistry.<sup>20,21</sup>

Previous merged-beam measurements<sup>10,11,13</sup> made in this laboratory were performed with ions formed in a conventional rf ion source. The mean residence time of ions in the source has been estimated to be  $\sim 30 \mu\text{s}$  and under these conditions it is estimated that about 60% of the ions were vibrationally excited. It is not surprising therefore that these ions displayed large recombination cross sections.

Recently a new ion source has been installed in the Van de Graaff accelerator which is used as the injector into the merged beam experiment. This source uses the method of ion trapping to confine ions for times sufficient to cause collisional deexcitation. The present paper describes measurements of  $H_3^+$  electron recombination using ions which have been demonstrated to be in vibrational states  $v=0$  and 1 only.

## EXPERIMENTAL

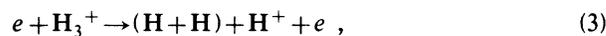
The ion source used in this study has been described elsewhere<sup>22</sup> so only a brief description will be given here. Ions formed by electron impact are confined by a radio

frequency field to move within a U-shaped channel. At the end of this channel they exit the source through an octupole lens.

An earthed aperture sits in front of this lens and the body of the source is maintained at a positive potential to assist in the extraction. Previous studies of collision-induced dissociation in  $H_3^+ - \text{He}$  collisions<sup>23</sup> showed that this source was indeed capable of producing  $H_3^+$  ions with low internal energy.

The ion source is mounted in the terminal of a 400-keV Van de Graaff accelerator. After acceleration the ions are magnetically mass analyzed and injected into the merged electron ion beam apparatus which has been described in detail elsewhere.<sup>10</sup> Inside this apparatus, which is maintained at a pressure of  $10^{-9}$  torr, the beam is deflected to eliminate neutrals, formed via charge exchange in the beam line, and then passed through the interaction region. The electron beam is formed from an indirectly heated barium oxide cathode and accelerated to a velocity close to that of the ions and in a direction initially parallel to that of the ion beam. For these measurements an ion energy of 400 keV and electron energies in the range 25–75 eV were used. The electrons are merged with the ions using a trochoidal analyzer which employs an axial magnetic field and a transverse electric field to shift the electrons from their initial path to a parallel but offset path. The latter is arranged to coincide with the ion beam. The beams interact over a distance of 8.6 cm and the overlap of the two beams is measured using three equispaced scanners.<sup>24</sup>

Following the interaction, the electron beam is deflected into a Faraday cup using a second trochoidal analyzer and its intensity is measured. The ion beam is electrostatically deflected into a second Faraday cup. The undeflected neutral products formed in the interaction region are detected using an energy-sensitive surface barrier detector. Products from reaction 2 arrive almost simultaneously at the detector and hence appear as a single full beam-energy particle. At higher collision energies, the reactions



yield  $H_2$  molecules, or H-atom pairs and individual H atoms which appear with  $\frac{2}{3}$  and  $\frac{1}{3}$  of the full beam energy, respectively. Single-channel analyzers are used to select the signals arising in the three channels so that they can be counted separately. Hence reactions 2, 3, and 4 can be studied individually. Interaction of the ion beam with the background gas in the interaction region also leads to neutral products in each of the three channels. These signals are distinguished from true signals arising from electron-ion collisions by modulating the electron beam and counting in and out of phase with the modulation using pairs of gated scalars. The difference in the scalar counts gives the true signal.

The collision cross section is determined using the expression

$$\sigma = \frac{C_j e^2}{I_i I_e L} \left| \frac{v_i v_e}{v_i - v_e} \right| F, \quad (5)$$

where  $C_j$  is the neutral count rate for channel  $j$ ,  $I_i, I_e, v_i, v_e$  are the ion and electron currents and velocities, respectively,  $L$  is the length of the interaction region,  $e$ , the electronic charge, and  $F$  is the effective collision area.<sup>24</sup>

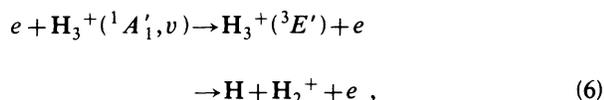
The center-of-mass collision energy is given by

$$E_{c.m.} = (E_e^{1/2} - E_+^{1/2})^2,$$

where  $E_+ = E_i m_e / m_i$  and  $E_i, E_e$  are the ion and electron energies. In practice the ion energy is held fixed and  $E_{c.m.}$  is varied by varying  $E_e$ . In related studies<sup>25</sup> of the recombination of  $H_2^+$  ions, sharp resonances have been seen and the width of these indicate that the energy resolution of the apparatus is better than 20 meV at low energies.

### DISSOCIATIVE EXCITATION MEASUREMENTS

Measurements of the threshold for the dissociative excitation process,



can be used to determine the internal energy of the  $H_3^+$  ions.<sup>26</sup> The  $^1A'_1 \rightarrow ^3E'$  transition is illustrated in Fig. 2 which shows potential-energy curves calculated by Kawaoka and Borkman.<sup>27</sup> These agree closely with earlier calculations by Schaad and Hicks.<sup>28</sup> It can be seen that for  $H_3^+(v=0)$ , this process is endothermic by about 14.9 eV. As the internal energy of the  $H_3^+$  ions is raised, the excitation threshold moves to lower energies. Since the  $^1A'_1 \rightarrow ^3E'$  transition is optically forbidden, it proceeds via electron exchange and so only displays a sizeable cross section close to threshold, falling off rapidly at higher energies.

Measurements of reaction 4 were performed for  $H_3^+$  ions formed under a variety of source conditions and the results of these measurements are shown in Figs. 3(a)–3(d). It can be seen from Fig. 3(a) that ions formed in a conventional rf ion source have states higher than  $v=4$  populated, as expected. By using an rf trap source and by varying the source pressure and extraction voltage it is possible to cool the ions in a controlled fashion.

In the absence of a Franck-Condon analysis for transitions involving states in both  $D_{3h}$  (symmetric stretch) and  $C_{2v}$  (asymmetric stretch), the indicated thresholds for the excited vibrational states must be considered approximate. The results indicate that the ion trap source operating at a low pressure (10 mtorr) produces  $H_3^+$  ions with  $v=0, 1, 2$ , and 3 [Fig. 3(b)] populated and increasing the pressure to 30 mtorr results in the quenching of the  $v=3$  state [Fig. 3(c)]. By reducing the extraction voltage to a low value and thus allowing the ions to remain for a longer period in the trap it is possible to quench the  $v=2$

state [Fig. 3(d)]. It should be noticed that the threshold in the latter case appears below the  $v=0$  threshold but above that ascribed to  $v=1$  using the potential curves shown in Fig. 2. A double-peaked structure is evident though and this would imply that excitation from the  $v=1$  level is occurring. Clarification of this point awaits the Franck-Condon analysis mentioned above.

Assuming that  $v=1$  is present and that the excitation cross section varies little for the  $v=0$  and 1 levels, it would appear that these states are approximately evenly populated.

### DISSOCIATIVE RECOMBINATION MEASUREMENTS

Cross sections for the dissociative recombination of  $H_3^+$  ions are illustrated in Fig. 4. The curves marked *a*, *b*, *c*, and *d* correspond to measurements made using ions formed under varying source conditions leading to the excitation functions as shown in Figs. 3(a)–3(d). It can be seen that for the case of  $H_3^+(v=0,1)$  the cross section is reduced by about a factor of 13 from that measured for ions obtained from a conventional rf source. Above 0.03 eV this cross section can be expressed in the form  $\sigma = 3.23 \times 10^{-17} E_{c.m.}^{-1.15} \text{ (cm}^2\text{)}$ .

Below 0.03 eV the energy dependence becomes less steep with the cross section being expressed in the form  $\sigma = 2.17 \times 10^{-16} E_{c.m.}^{-0.57} \text{ (cm}^2\text{)}$ . A similar change in the energy dependence can also be seen for curves *a*, *b*, and *c*,

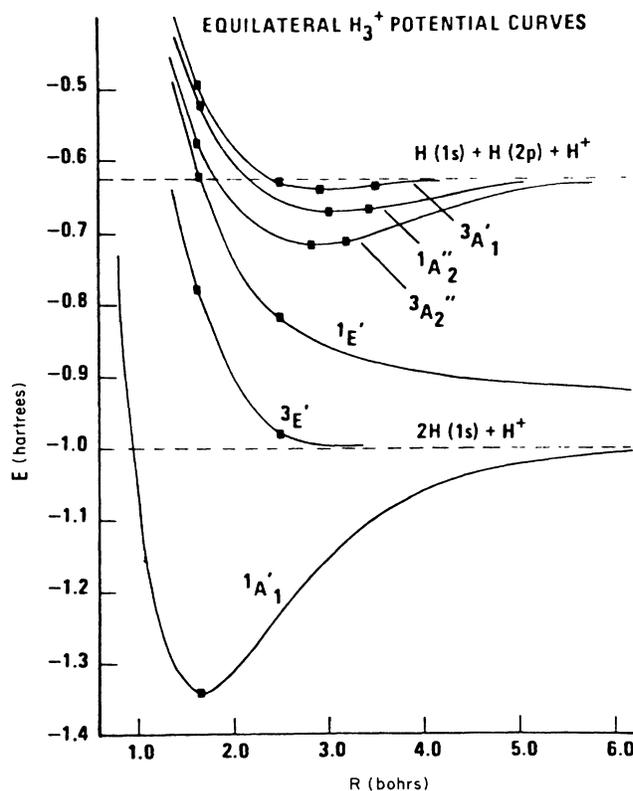


FIG. 2. Potential-energy curves for the ground and excited states of  $H_3^+$  in  $D_{3h}$  symmetry (reproduced with permission from Ref. 27).

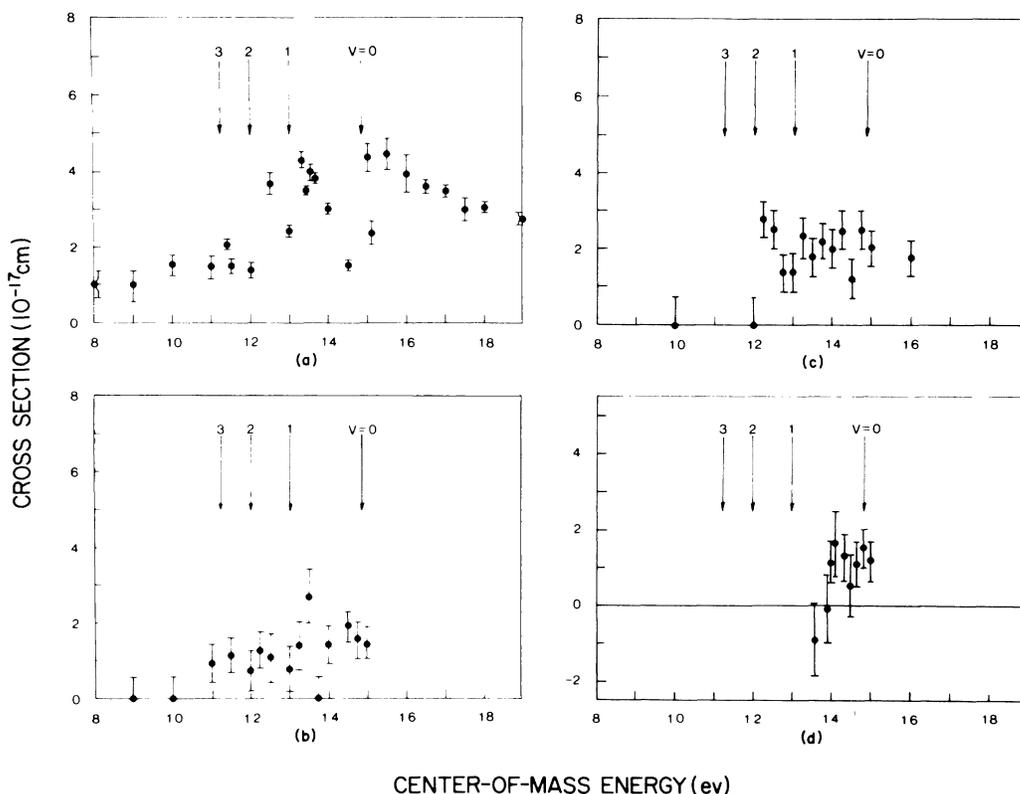


FIG. 3. Cross sections for the dissociative excitation process:  $E + \text{H}_3^+ \rightarrow \text{H}_3^+(^3E') + e \rightarrow \text{H}_2 + \text{H}^+ + e$ . (a) Ions produced in conventional rf source at 100 mtorr; pure  $\text{H}_2$ . (b) Ions produced in trap ion source at 10 mtorr; pure  $\text{H}_2$ . (c) Trap ion source 30 mtorr; pure  $\text{H}_2$ . (d) Trap ion source, 70 mtorr, low extraction 10:3:1,  $\text{H}_2$ :Ar:He mixture.

although the onset of the steeper dependences occurs at higher energies, i.e., 0.15, 0.08, and 0.05 eV, respectively. In an earlier publication,<sup>10</sup> the effect of a degradation of the energy resolution upon the measured cross section was analyzed and it was shown that this could give rise to a decreased measured energy dependence. This is not believed to be the reason for the effect shown in this paper. Resonant features have been found in recent studies<sup>25,29</sup> of  $\text{H}_2^+$  and  $\text{N}_2^+$  recombination which indicate that the energy resolution of the MEIBE apparatus is in fact very high ( $\Delta E_{\text{c.m.}} < 20$  meV) at low energies. Similar sudden slope changes have been seen in other measurements of recombination cross sections made using both the merged-beam<sup>30</sup> and ion-trap<sup>31</sup> techniques. In general, however, these are only seen with polyatomic ions while diatomic species yield continuous linear dependences over the entire energy range. This is very dramatically illustrated in the case of  $\text{OH}^+$  and  $\text{H}_2\text{O}^+$  recombination.<sup>30</sup> There are, however, intriguing exceptions to this rule.  $\text{CH}_2^+$  (Ref. 32) and  $\text{HD}_2^+$  (Ref. 11) display linear dependences while recent remeasurements of  $\text{N}_2^+$  recombination<sup>29</sup> display a more gradual energy dependence below 0.03 eV.

An explanation for these phenomena will require detailed theoretical studies of polyatomic ion recombination. Such calculations for  $\text{H}_2^+$  (Refs. 33 and 34) ions show that individual vibrational states do display sudden

changes in energy dependences which can be explained in terms of Franck-Condon overlap factors between the initial and final states involved in the recombination. The influence of neutral autoionizing states on the recombination process also leads to changes in the energy dependence.

It is possible to estimate a rate coefficient  $\alpha(T_e)$  for the recombination using the expression

$$\alpha(T_e) = \int_0^\infty \frac{8\pi m_e E_{\text{c.m.}}}{(2\pi m_e k T_e)^{3/2}} \times \sigma(E_{\text{c.m.}}) \exp(-E_{\text{c.m.}}/k T_e) dE_{\text{c.m.}}$$

Evaluation of this expression involves a linear extrapolation of our measured data to lower energies although this assumes that the form of the cross-section energy dependence does not change greatly outside our measured energy range. The change in energy dependence at 0.03 eV complicates this analysis. By assuming linear dependence above and below this, it is possible to obtain an approximate value for  $\alpha$ . The resulting rate coefficient for  $\text{H}_3^+$  ions with  $v=0$  and 1 populated varies from  $2 \times 10^{-8}$   $\text{cm}^3 \text{s}^{-1}$  at 100 K to  $1 \times 10^{-8}$  at 1000 K.

It can be seen that our findings reflect the earlier conclusion of Adams *et al.*, that the recombination rate is

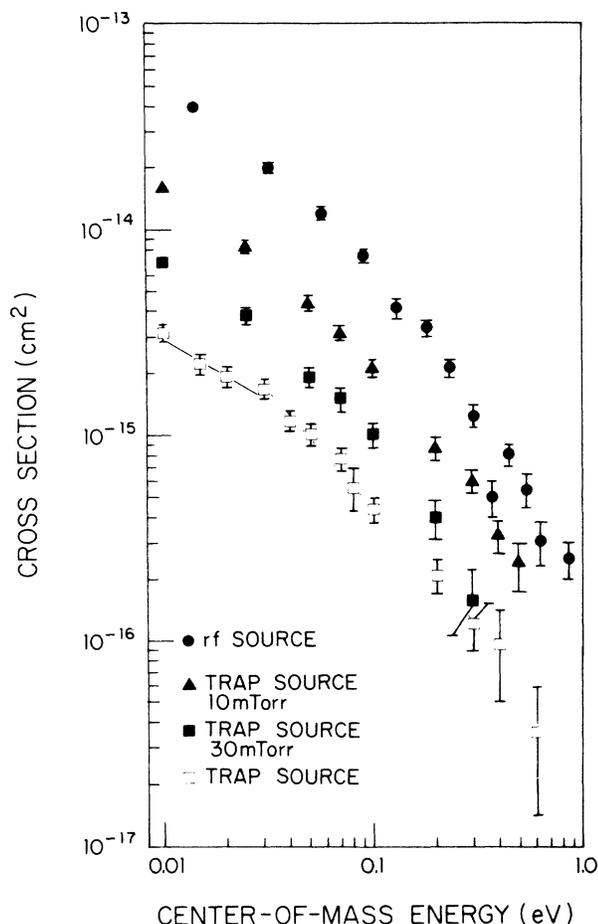


FIG. 4. Cross sections for  $e\text{-H}_3^+$  recombination. The four curves shown here were measured using ions formed under conditions which yielded the corresponding four excitation functions shown in Fig. 3.

substantially reduced due to the poor overlap with the dissociating state for vibrationally cool ions. More recently, Smith and Adams<sup>18</sup> have revised their estimate for the upper limit for the recombination rate to  $2 \times 10^{-11} \text{ cm}^3 \text{ s}^{-1}$  at 300 K but this result is difficult to reconcile with our measurements. Even if complete deexcitation of  $\text{H}_3^+$  is achieved in the FALP technique (and this is inferred, not measured), it is difficult to believe that the rate coefficient for  $\text{H}_3^+(v=0)$  is three orders of magnitude smaller than for  $\text{H}_3^+(v=1)$ .

Figure 5 shows the variation of the recombination cross section at 0.01 eV as a function of source pressure and extraction and it can be seen that a plateau is reached at the lowest value. At this point the cross section is independent of source conditions suggesting that either it has become rather insensitive to the vibrational state population or that further deexcitation cannot be achieved.

It has been suggested<sup>35</sup> that the method of measuring the rate coefficient might explain the discrepancies between the measurements. In the FALP technique, a Langmuir probe is used to measure the rate of change of electron concentration in the afterglow while in the

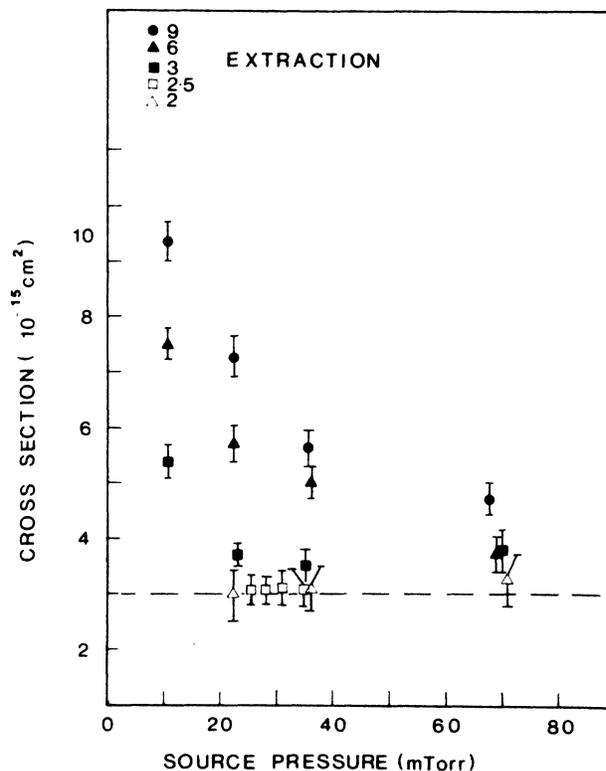


FIG. 5. Variation of the  $e\text{-H}_3^+$  recombination cross section measured at 0.01 eV as a function of source pressure and extraction voltage (arbitrary units).

merged-beam experiment the neutrals following the recombination are detected. It may be that the recombination of  $\text{H}_3^+(v=0)$  with electrons does not proceed via a dissociative path but rather through a process in which long-lived  $\text{H}_3$  rydberg molecules are formed. These may subsequently decay via radiation emission.<sup>36</sup> It is possible that such an entity might be reionized at the surface of a Langmuir probe due to potential electron ejection and thus increase the measured electron concentration.

In the merged-beam case, it takes approximately  $10^{-7}$  s for the neutral products to reach the detector and therefore such a rydberg molecule would have to be stable against autoionization for at least that time. In future studies the branching ratio for the various possible decay channels following the recombination will be measured in an effort to elucidate this problem.

*Note added:* A recent reanalysis of the measurements of  $\text{H}_3^+$  recombination by Biondi and coworkers indicates that contamination of the afterglow by  $\text{CH}_5^+$  ions may explain their high measured rate coefficients.<sup>35</sup>

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