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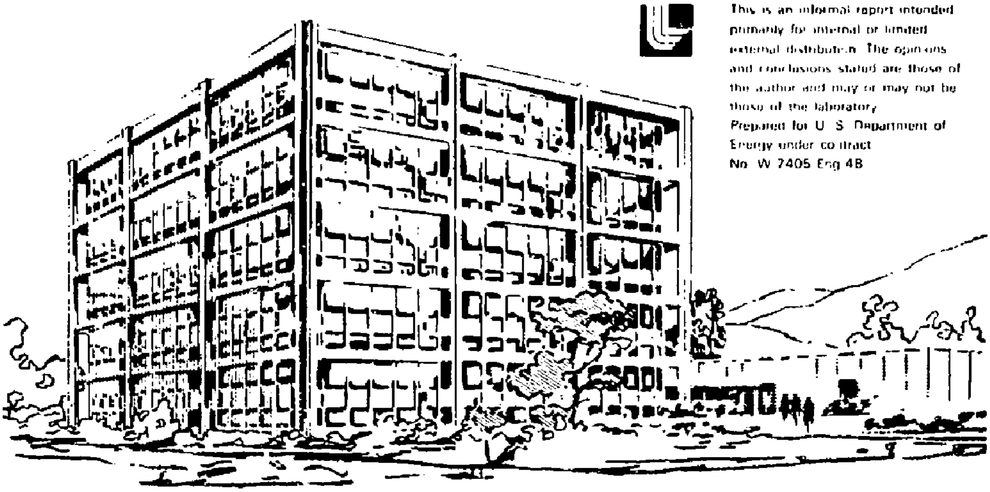

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ATOMIC REACTION RATES IN H_2 AND D_2 PLASMAS*

J. R. Hiskes, M. Bacal†, G. W. Hamilton

ABSTRACT

The processes leading to negative ion production in a hydrogen plasma are reviewed. The dissociative attachment of electrons to highly excited vibrational states of hydrogen molecules appears to be the most probable source of negative ions. The possibility of significant negative ion formation from dissociative recombination of H_3^+ with electrons cannot be excluded. The de-excitation probability of vibrationally excited hydrogen molecules in wall collisions may be a critical parameter in interpreting the data.

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I. INTRODUCTION

Measurements[1,2] of H^- and D^- in plasmas indicate that non-linear mechanisms involving intermediate states are required to explain the functional form and also the large magnitude of the rate of production of H^- and D^-. Therefore a review of the atomic reaction rates in the plasma is required to determine which mechanisms are responsible for negative ion production. The results should be useful in development of a negative ion plasma of the density and area required for neutral injection.

We know of only three atomic processes for H^- production with sufficiently large reaction rates to possibly explain the measurements. All three processes involve hydrogen molecules or ions produced by intermediate mechanisms. The three processes proposed[3] are the following:

a. Dissociative attachment of electrons to vibrationally excited states of hydrogen molecules:[4,5]

$$e + H_2(v*) \rightarrow H_2^-(^2\Sigma_u^+) \rightarrow H(1s) + H^-$$

b. Dissociative attachment of electrons to an electronically excited long-lived state of hydrogen molecules:

$$e + H_2(^3\pi_u) \rightarrow H_2^-(^2\pi_u) \rightarrow H(2p) + H^-$$

and c. Dissociative recombination of H_3^+:

$$e + H_3^+ \rightarrow H_3^* \rightarrow H_2^+ + H^-$$

We must also compute the rates of production and loss of the excited molecules and H_3^+ ions to compute the production rate of H^-. These species are derived from H_2^+ produced by ionization of the background gas. Therefore the complete problem consists of the computation of the production and loss rates of five species in the plasma: H_2^+, H_3^+, $H_2(v^*)$, $H_2(^3\pi_u)$, and H^-.

In order to provide a standard data base for the discussions to follow, we summarize the current experimental data in the following table.

TABLE I

	n	T
	cm^{-3}	eV
Negative Species		
e (thermal)	2×10^{10}	1
e (fast beam)	2×10^{7}	120
H^-	4×10^{9}	0.1*
Positive Ions		
n_+	2.4×10^{10}	0.1*
H_1^+	$0.03\ n_+$	
H_2^+	$0.05\ n_+$	
H_3^+	$0.92\ n_+$	
Gas		
$H_2(v = 0)$	4×10^{14}	

*Inferred values.

The experiment has also shown that:

a. The density n(-) varies as $n^3(e)$, when $n(e) \leq 2 \times 10^{10}\ cm^{-3}$ with $n(-) = 5 \times 10^{-22}\ n^3(e)$.

b. There is no isotope effect for D^- production compared to H^-, to within an uncertainty of a factor of two.

In developing the discussion of negative ion production via vibrational excitation, process (a), we rely heavily on the recent theoretical work of Wadehra and Bardsley (WB)[5], and the experimental verification of this work up to v = 4 by Allan and Wong (AW).[4] In the discussion which follows we shall conclude that the higher vibrational levels, v = 6 → 9, provide the principal contribution to the dissociative attachment. This conclusion is due mainly to the fact that these levels are energetically accessible to the $H_2^-(^2\Sigma_u^+)$ electronic state for an electron temperature of one electron-volt, and the large magnitude of the WB cross sections for these high vibrational states. Endothermic transitions via the $v \geq 10$ levels through the $^2\Sigma_u^+$ state have not yet been calculated but may also make a significant contribution. Also, the importance of dissociative attachment at large internuclear separations and high vibrational excitation, as shown by WB, suggests that transitions through the first excited electronic state, $H_2^-(^2\Sigma_g)$, may be important. Taken together, these additional transitions may make a contribution comparable to the v = 6 → 9 yield.

The dissociative attachment to the ground vibrational state of the molecule, v = 0, is notorious for its strong isotope dependence[6]: the $H_2(v = 0)$ cross section exceeds that of $D_2(v = 0)$ by a factor of five hundred. Inspecting the published WB cross sections (Ref. 4) for vibrational excitations up to 1.75 eV, $H_2(v = 4)$, $D_2(v = 5)$, however, shows that the isotope dependence has narrowed to a factor of five. Unpublished $D_2(v = 9 \rightarrow 13)$ cross sections[7], corresponding to D_2 vibrational levels in the same excitation range as $H_2(v = 6 \rightarrow 9)$, are comparable to the $H_2(v = 6 \rightarrow 9)$ dissociative attachment cross sections. For the higher vibrational levels, i.e., comparable internal vibrational energy in either H_2 or D_2, the isotope dependence is rather weak, consistent with the experimental observations.

Estimates of the cross sections for the dissociative attachment to long-lived[8] electronically-excited $H_2(^3\pi_u)$ states have been made by Bottcher and Buckley.[9] These calculations also indicate a weak isotope dependence.

A calculation by Kulander[10] of the H_3 electronic resonance leading to the $H_2^+ + H^-$ dissociation channel, reaction (c), is in progress. Kulander notes that the threshold electron energy leading to this channel is at least 1.5 electron-volts, and the apparent threshold may be a few volts higher, depending upon the degree of vibrational excitation in the parent H_3^+ ion. The isotope dependence leading to the negative ion channel is unclear at present.

In the following sections we shall consider the reaction rates and equilibrium rate equations for the various ionic and neutral components of the hydrogen plasma. For the most part, reaction rates and cross sections used here are taken from the Oak Ridge Redbook.[11]

II. PRODUCTION AND LOSS OF H_2^+

A. Production Processes

(a) $e + H_2(v = 0) \rightarrow H_2^+(v^*) + 2e$

- (i) Fast electron energy 120 eV
- (ii) Fast electron density = 2×10^7 cm^{-3}
- (iii) $\overline{\sigma v}(2+) \equiv$ Ionization rate = 5.4×10^{-8} cm^3 sec^{-1}

B. Loss Processes

(a) Wall neutralization: $H_2^+(v^*) + \text{wall} \rightarrow H_2(v^{**})$

- (i) $v(2+)$ = drift velocity to walls; L = mean distance to wall; $v(2+)/L = 3 \times 10^4$ sec^{-1}

(b) $H_2^+(v^*) + H_2(v = 0) \rightarrow H_3^+(v^{**}) + H$

(i) H_2^+ energy = 0.1 eV (see comment on H_3^+ energy in Sec. III)

(ii) $\overline{\sigma v}(3+) = 1.2 \times 10^{-9}\ cm^3\ sec^{-1}$

(c) $H_2^+(v^*) + e \rightarrow$ neutrals

(i) Electron temperature = 1.0 eV

(ii) $\overline{\sigma v}(DR) = 3.3 \times 10^{-8}\ cm^3\ sec^{-1}$

C. Rate Equation for H_2^+ Density

$$\frac{dn(2+)}{dt} = n^f(e)\ n(v = 0)\ \overline{\sigma v}(2+) - n(2+)\ v(2+)/L - n(2+)\ n(v = 0)\ \overline{\sigma v}(3+) - n(2+)\ n(e)\ \overline{\sigma v}(DR) = 0$$

The loss processes (b) dominates.

$$n(2+) = \frac{n^f(e)\ \overline{\sigma v}(2+)}{\overline{\sigma v}(3+)} = 1 \times 10^9\ cm^{-3}$$

The H_2^+ density is calculated to be 5% of the total ion density and agrees with the mass spectrometer data listed in Table I.

III. PRODUCTION AND LOSS OF H_3^+

A. Production Process

(a) $H_2^+(v^*) + H_2(v = 0) \rightarrow H_3^+(v^{**}) + H$

(i) $\overline{\sigma v}(3+) = 1.2 \times 10^{-9}\ cm^3\ sec^{-1}$

B. Loss Processes

(a) Wall neutralization, $v(3+)/L = 2.5 \times 10^4\ sec^{-1}$. (The value for v/L was adjusted to give the observed H_3^+ density below.)

(b) $e + H_3^+ \rightarrow$ neutrals

(i) $\overline{\sigma v}(DR) = 5.4 \times 10^{-8}\ cm^3\ sec^{-1}$

C. Rate Equation for H_3^+ Density

$$\frac{dn(3+)}{dt} = n(2+)\ n(v = 0)\ \overline{\sigma v}(3+) - n(3+)\ v(3+)/L - n(3+)\ n(e)\ \overline{\sigma v}(DR) = 0$$

The wall loss process (a) dominates.

$$n(3+) = \frac{n(2+)\ n(v = 0)\ \overline{\sigma v}(3+)}{v(3+)/L} = 2 \times 10^{10}\ \mathrm{cm}^{-3}$$

The positive plasma potential is approximately 1 eV; the adjusted value for v(3+)/L indicates that the H_3^+ energy is 0.1 eV. This in turn is consistent with a plasma potential drop occurring near the edge of the plasma.

IV. PRODUCTION AND LOSS OF VIBRATIONALLY EXCITED $H_2(v^*)$

A. Production Processes

(a) Charge exchange: $H_2^+(v^*) + H_2(v = 0) \rightarrow H_2(v^*) + H_2^+(v^*)$. The $H_2^+(v^*)$ vibrational distribution[12] is peaked about $v = 2,3,4$ with approximately 15% of the total population in $v = 3$. The probability density of the $v = 3$ state is localized near the outer turning point near $R = 3a_0$. Charge exchange to H_2 from these higher vibrational levels of H_2^+ will lead to large Frank-Condon overlap integrals with the $v = 6 - 9$ states of $H_2(v^*)$. Consideration of the Frank-Condon factors and the H_2^+ population distribution suggests a reasonable estimate for the populations of H_2 vibrational states in the range $v = 6 - 9$ to be 10%. In the limit of a zero energy collision, however, not all vibrational levels are energetically accessible. The minimum energy for removing an electron from H_2 on the left is 15.4 electron volts; this would imply capture into $v \leq 5$ for H_2 on the right. The population of the higher vibrational states of H_2 may be very sensitive to the relative energy when the energy is low.

(i) H_2^+ energy = 0.1 eV

(ii) $\sigma v(CX) = 3 \times 10^{-10}$ cm^2 sec^{-1}

(iii) p(6 - 9) = probability of finding $H_2(v^*)$ in states v = 6 - 9

(b) Wall neutralization:[13] $H_2^+(v^*) + \text{wall} \rightarrow H_2(v^*)$

(i) $v(2+)/L = 3 \times 10^4$ sec^{-1}.
Here the Frank-Condon factors appear to be appropriate in a first approximation, and p(6 - 9) is taken to be 10%.

(c) $H_3^+ + e \rightarrow H_2(v^*) + H$

(i) $\sigma v(DR3+) = 5.4 \times 10^{-8}$ cm^3 sec^{-1}.
From the formation and structure of H_3^+, vibrational excitation of H_2 on the right would be expected to be substantial, but for lack of any quantitative data we take p(v*;3+) equal to 10%.

(d) $H_2(v) + e \rightarrow H_2(v^*) + e$. Principal excitations are $\Delta v = \pm 1$. Several sequential excitations are required to reach v = 6 - 9. These rates are small compared to those of the first three processes, but may become important at higher electron temperatures and densities.

(e) $H^- + H_2^+ \rightarrow H + H_2(v^*)$

(i) $\sigma v(ii) = 2 \times 10^{-7}$ cm^3 sec^{-1} (E 0.1 eV).
The final vibrational distribution would be similar to that for charge exchange, discussed under (a). The rate here is approximately three orders of magnitude larger than for charge exchange but the density of H^- is five orders of magnitude less than the H_2 density. At higher ion densities or lower gas pressures this process may be important. This process is ignored in our equilibrium estimates.

(f) $H^- + H_3^+ \rightarrow H + H_3^* \rightarrow H + H_2(v^*) + H$

(i) The σv will be similar to that for (e); here however, the H_3^+ density is 20 times larger than the H_2^+ density. The contribution here appears to be comparable to process (c); here we shall include (c) but double the rate to include this process.

B. <u>Loss Processes</u>

(a) $H_2(v^*) + e \rightarrow H_2^-(v^*) \rightarrow H^- + H$

$\rightarrow H_2(v \neq v^*) + e$

(i) $\sigma v(DA;\ v = 6 - 9) = 2 \times 10^{-8}$ cm^3 sec^{-1} (from Wadehra and Bardsley, Ref. 5).

(ii) $\overline{\sigma v}$(decay). The lower channel indicated above is estimated to be comparable to the upper channel. Hence, the total $\overline{\sigma v}$ for reaction (a) is taken to be $\overline{\sigma v}(a) = 4 \times 10^{-8}\ cm^{-3}\ sec^{-1}$.

(b) Wall Collisions: $H_2(v^*) + \text{wall} \rightarrow H_2(v \neq v^*)$

(i) $b(v^*) \equiv$ *number of bounces off wall before* $v \neq v^*$.

(ii) $v/L \equiv$ arrival rate at wall $= 3 \times 10^4\ sec^{-1}$

(c) Gas Collisions: $H_2(v^*) + H_2(v = 0) \rightarrow H_2(v^{**}) + H_2(v)$. The lower vibrational levels move in a near harmonic potential with almost equidistant level spacings. At low energies transitions of the type

$$H_2(v^*) + H_2(v = 0) \rightarrow H_2(v^* - 1) + H_2(v = 1)$$

are near-resonant and may exhibit gas-kinetic cross sections. The higher level spacings are not equidistant, and for low energy collisions we would not expect these cross sections to be large; we are not aware of any data for this process when $v^* \geq 6$. For a cross section of $10^{-17}\ cm^2$, the mean *collision time would be about equal to the time for twenty-five* bounces.

C. Rate Equation for $H_2(v^*)$

$$\frac{dn(v^*)}{dt} = p(v^*)\ n(2+)\ n(v = 0)\ \overline{\sigma v}(CX)$$
$$+ p(v^*)\ n(2+)\ v_+/L$$
$$+ p(v^*;3+)\ n(3+)\ n(e)\ \overline{\sigma v}(DR3+)$$
$$- n(v^*)\ n(e)\ \overline{\sigma v}(a)$$
$$- n(v^*)\ \frac{1}{b(v^*)}\ \frac{v}{L}$$
$$= 0$$

The equilibrium density becomes

$$n(v{=}6\text{-}9) = \frac{p(v^*)n(2+)[n(v{=}0)\overline{\sigma v}(CX) + v_+/L] + 2p(v^*;3+)n(3+)n(e)\sigma v(DR3+)}{n(e)\overline{\sigma v}(a) + \frac{1}{b}\frac{v}{L}}$$

	With Charge Exchange	Without Charge Exchange
$b >> 1$:	$n(v = 6\text{-}9) = 2.4 \times 10^{10}\ cm^{-3}$;	$n(v = 6\text{-}9) = 0.9 \times 10^{10}\ cm^{-3}$
$b = 1$:	$n(v = 6\text{-}9) = 5 \times 10^{8}\ cm^{-3}$;	$n(v = 6\text{-}9) = 2.4 \times 10^{8}\ cm^{-3}$

The higher density requires at least 50 bounces.

V. THE PRODUCTION AND LOSS OF $H_2(^3\pi_u)$

A. Production Processes

(a) $H_2(v = 0) + e \rightarrow H_2(^3\pi_u;v^*) + e$. We do not know of a cross section value for this process in the literature.

Singlet and triplet excitations are expected to be comparable near the maximum of the cross sections. Excitation cross sections are comparable to ionization cross sections near the respective maxima. We shall take the product of $\overline{\sigma v}$ and the fast electron density equal to the values for H_2 ionization used in Sec. II.A.iii; i.e., $n(e)\overline{\sigma v} = 1.1$ sec^{-1}.

B. Loss Process

(a) $H_2(^3\pi_u;v^*) + e \rightarrow H_2(^3\Sigma_g;v^*) + e$. The $^3\Sigma_g$ state decays by a radiative transition in 10^{-8} sec[14]. These cross sections are expected to be very large, comparable to the 2s → 2p excitation cross section in hydrogen or to the $2^3s \rightarrow 2^3p$ excitations in He. These large cross sections are in part due to the small energy differences of the $^3\pi_u$ - $^3\Sigma_g$ states, typically 0.017 → 0.100 eV.[8] Seaton[15] has calculated the 2s-2p excitation rate in H to be 2.2×10^{-5} cm^3 sec^{-1} for an electron temperature near one electron-volt. Dividing this by a statistical factor of three for 2p → 2s excitation, we shall take as the upper limit the value $\overline{\sigma v} = 7 \times 10^{-6}$ cm^3 sec^{-1}. Moiseiwitsch[16] has calculated a maximum cross section of 2.7×10^{-14} cm^{-2} for He 2^3s - 2^3p excitation. Taking this value as a lower limit, for one electron volt electrons we have $\overline{\sigma v} = 1.5 \times 10^{-6}$ cm^3 sec^{-1}.

(b) Quenching of $H_2(^3\pi_u)$ by wall collisions

(i) Data for electronically excited atomic and molecular systems colliding with metal walls indicate virtually total quenching per collision. The wall loss rate is taken to be $v_u/L = 3 \times 10^4\ \text{sec}^{-1}$.

C. Rate Equation for $H_2(^3\pi_u)$ Density

$$\frac{dn(u)}{dt} = n(v = 0)\ [n^f(e)\ \overline{\sigma v}]$$
$$- n(u)\ n(e)\ \overline{\sigma v}(c)$$
$$- n(u)\ v_u/L$$
$$= 0$$

$$n(u) = \frac{n(v = 0)\ [n^f(e)\ \overline{\sigma v}]}{n(e)\ \overline{\sigma v}(c) + v_u/L}$$

$$n(u) = 2.6 \times 10^9\ \text{cm}^{-3};\ \text{lower limit}$$
$$= 7 \times 10^9\ \text{cm}^{-3};\ \text{upper limit}$$

And the ratio of the $H_2(^3\pi_u)$ density to the gas density is

$$\frac{n(u)}{n(v = 0)} = \begin{cases} 6 \times 10^{-6}; & \text{lower limit} \\ 2 \times 10^{-5}; & \text{upper limit} \end{cases}$$

VI. PRODUCTION AND LOSS OF H^-

A. Production Processes

(a) $H_2(v = 6 - 9) + e \rightarrow H_2^-(v^*) \rightarrow H^- + H$

(i) $\overline{\sigma v}(DA) = 2.2 \times 10^{-8}\ cm^3\ sec^{-1}$

(b) $H_2(^3\pi_u;v^*) + e \rightarrow H_2^-(^2\pi_u;v^*) \rightarrow H^- + H(2p)$.
Bottcher and Buckley[9] have estimated these dissociative attachment cross sections to be in the range $10^{-18}\ cm^2$ to $2 \times 10^{-17}\ cm^2$. Since the $H_2(^3\pi_u)$ potential curve has a similar shape to H_2^+, in first approximation the vibrational population distribution for $H_2(^3\pi_u)$ can be taken equal to that for H_2^+. The fraction of the total population of excited vibrational states which is effective at an electron temperature of one electron volt would appear to be approximately 50%. The reaction rate $\overline{\sigma v}(DA,u)$ is then in the range 2.7×10^{-11} to $5.4 \times 10^{-10}\ cm^3\ sec^{-1}$.

(c) $H_3^+(v^*) + e \rightarrow H_2^+ + H^-$. Kulander has pointed out that the threshold for this process is 1.5 eV for H_3^+ in the highest vibrational levels, and 6 eV for the ground vibrational level. The cross section is unknown. The rate for dissociative recombination for H_3^+ in the ground vibrational state is less than $4.3 \times 10^{-8}\ cm^3\ sec^{-1}$ for energies greater than 1.5 eV, with a value equal to 3×10^{-8} cm^3 sec^{-1} at 6 eV. We define R(2 + -) equal to the ratio of the ion-pair channel compared to the total dissociative recombination.

B. Loss Processes

(a) H^- + positive ion → neutrals

(i) $\overline{\sigma v}(ii) = 2 \times 10^{-7}$ cm^3 sec^{-1} (E = 0.1 eV)

(b) $H^- + e \rightarrow H + 2e$

(i) $\overline{\sigma v}(c) = 10^{-8}$ cm^3 sec^{-1}. This rate is insignificant compared to (a).

(c) The effective loss rate to the walls, v(-)/L, is estimated by Bacal and Hamilton[1] to be in the range 1×10^3 sec^{-1}.

C. The Rate Equation for the H^- Density

$$
\begin{aligned}
\frac{dn(-)}{dt} &= n(v = 6 \rightarrow 9)\, n(e)\, \overline{\sigma v}(DA) \\
&+ n(u)\, n(e)\, \overline{\sigma v}(DA,u) \\
&+ n(3+)\, n(e)\, R(2 + -)\, \overline{\sigma v}(DR) \\
&- n(-)\, n(+)\, \overline{\sigma v}(ii) \\
&- n(-)\, v(-)/L \\
&= 0
\end{aligned}
$$

The equilibrium density becomes

$$n(-) = \frac{n(e)[n(v=6\rightarrow 9)\overline{\sigma v}(DA) + n(u)\overline{\sigma v}(DA,u) + n(3+)R(2+-)\overline{\sigma v}(DR)]}{n(+)\ \overline{\sigma v}(ii) + v(-)/L}$$

(a) b = 1; upper limits for $H_2(^3\pi_u)$; H_3^+ rate for 1.5 eV; with charge exchange formation of $H_2(v^*)$.

$$n(-) = 4 \times 10^6\ [11.0 + 3.8 + 860\ R(2 + -)] \quad .$$

The H_3^+ will make a significant contribution if R is of order 1%. The upper limit for the $H_2(^3\pi_u)$ contribution is about one-third the vibrational excitation contribution. If R = 0, $n(-) = 6 \times 10^7\ cm^{-3}$. The observed H^- density is approximately seventy times larger. If b = 1, R must be unity to achieve the observed density.

(b) b >> 1; upper limits for $H_2(^3\pi_u)$; H_3^+ rate for 1.5 eV; with charge exchange formation of $H_2(v^*)$.

$$n(-) = 4 \times 10^6\ [530.0 + 3.8 + 860.0\ R(2 + -)] \quad .$$

If R = 0, $n(-) = 2 \times 10^9\ cm^{-3}$. Note that if the vibrational levels above v = 9 together with transitions through the $H_2^-(^2\Sigma_g)$ state contribute an amount equal to the contributions included here, then vibrational excitation alone could explain the observed negative ion density of $4 \times 10^9\ cm^{-3}$. Without charge exchange the density will be less than half this value.

VII. CONCLUSIONS

A. A complete interpretation of the negative ion density observed in hydrogenic plasmas in terms of the rate constants and plasma species is not yet possible.

B. The most probable interpretation of the negative ion yield for the density range studied here is via dissociative attachment of electrons to vibrationally excited ($v > 6$) hydrogen molecules. The principal source of $H_2(v \geq 6)$ vibrational excitation is unclear. At very low relative energy charge exchange of H_2^+ and H_2 ($v = 0$) may not be a sufficient source of high vibrational excitation. Auger neutralization of H_2^+ in wall collisions will contribute to high vibrational excitation.

C. The role of the H_3^+ ion remains one of the principal mysteries. It may contribute significantly to H^- production in either of two ways: Dissociative recombination via the channel $H_2^+ + H^-$; dissociative recombination yielding $H_2(v^*)$ in highly excited vibrational states.

D. The electronically excited $H_2(^3\pi_u)$ molecular density appears to be limited to low values because of the large destruction cross sections.

E. The survival probability of $H_2(v^*)$ striking a wall, or alternatively, the mean number of bounces an $H_2(v^*)$ can undergo before changing its vibrational state, is a critical parameter in interpreting the data.

F. The present analysis can serve as a starting point for extrapolation; the dominant terms will change relatively as a function density, ion species, and temperature.

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