Dissociative Recombination of Cold $\text{H}_3^+$
(and its interstellar implications)

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- A. Paal, F. Österdahl (Manne Siegbahn Laboratory)
- A. Al-Khalili, A. Ehlerding, F. Hellberg, S. Kalhori, A. Neau, R. Thomas, M. Larsson (Stockholm University)
Astronomer's Periodic Table
H$_3^+$: Cornerstone of Interstellar Chemistry
Observing Interstellar H$_3^+$

- Equilateral triangle
- “No” rotational spectrum
- “No” electronic spectrum
- Vibrational spectrum is only probe
- Absorption spectroscopy against background or embedded star
Interstellar Cloud Classification

**Dense molecular clouds:**
- $H \rightarrow H_2$
- $C \rightarrow CO$
- $n(H_2) \sim 10^4$–$10^6$ cm$^{-3}$
- $T \sim 20$ K

**Diffuse clouds:**
- $H \leftrightarrow H_2$
- $C \rightarrow C^+$
- $n(H_2) \sim 10^1$–$10^3$ cm$^{-3}$
  - $[\sim 10^{-18}$ atm$]$
- $T \sim 50$ K

Diffuse atomic clouds
- $H_2 << 10\%$

Diffuse molecular clouds
- $H_2 > 10\%$ (self-shielded)


Photo: Jose Fernandez Garcia
$N(\text{H}_3^+) = 1 - 5 \times 10^{14} \text{ cm}^{-2}$

McCall, Geballe, Hinkle, & Oka
Dense Cloud $\text{H}_3^+$ Chemistry

**Formation**

\[ \text{H}_2 \xrightarrow{\text{cosmic ray}} \text{H}_2^+ + e^- \]
\[ \text{H}_2 + \text{H}_2^+ \rightarrow \text{H}_3^+ + \text{H} \quad \text{(fast)} \]

**Rate**

\[ \text{Rate} = \zeta [\text{H}_2] \]

**Destruction**

\[ \text{H}_3^+ + \text{CO} \rightarrow \text{HCO}^+ + \text{H}_2 \]

**Rate**

\[ \text{Rate} = k [\text{H}_3^+] [\text{CO}] \]

**Steady State**

\[ \frac{(3 \times 10^{-17} \text{ s}^{-1})}{(2 \times 10^{-9} \text{ cm}^3 \text{ s}^{-1})} \times (6700) = 10^{-4} \text{ cm}^{-3} \]

Density Independent!

H$_3^+$ as a Probe of Dense Clouds

- Given $n$(H$_3^+$) from model, and $N$(H$_3^+$) from infrared observations:
  - path length $L = N/n \sim 3 \times 10^{18}$ cm $\sim 1$ pc
  - density $\langle n$(H$_2)$\rangle = N$(H$_2$)$/L \sim 6 \times 10^4$ cm$^{-3}$
  - temperature $T \sim 30$ K

- Unique probe of clouds
- Consistent with expectations
  - confirms dense cloud chemistry
Diffuse Molecular Cloud $H_3^+$ Chemistry

Formation

$$H_2 \overset{\text{cosmic ray}}{\rightarrow} H_2^+ + e^- \quad \text{Rate} = \zeta [H_2]$$

$$H_2 + H_2^+ \rightarrow H_3^+ + H$$

Destruction

$$H_3^+ + e^- \rightarrow H + H_2 \text{ or } 3H \quad \text{Rate} = k_e [H_3^+] [e^-]$$

Steady State

$$[H_3^+] = \frac{\zeta [H_2]}{k_e [e^-]} = \frac{(3 \times 10^{-17} \text{ s}^{-1})}{(5 \times 10^{-7} \text{ cm}^3 \text{ s}^{-1})} \times (2400)$$

$$= 10^{-7} \text{ cm}^{-3}$$

Density Independent!

$10^3$ times smaller than dense clouds!
Lots of $\text{H}_3^+$ in Diffuse Clouds!

- $\text{N}(\text{H}_3^+) \sim$ dense clouds
- $n(\text{H}_3^+) \sim$ 1000 times less
- $\therefore L \sim$ 1000 times longer ?!!

McCall, et al.
Big Problem with the Chemistry!

★ ~2 orders of magnitude!!

Steady State: \[ [H_3^+] = \frac{\zeta [H_2]}{k_e [e^-]} \]

To increase the value of \([H_3^+]\), we need:

• Smaller electron fraction \([e^-]/[H_2]\)
• Smaller recombination rate constant \(k_e\)
• Higher ionization rate \(\zeta\)
**H$_3^+$ toward ζ Persei**

---

**N(H$_2$) from Copernicus**

<table>
<thead>
<tr>
<th>HD</th>
<th>NAME</th>
<th>$\beta$</th>
<th>$\beta$</th>
<th>S. T.</th>
<th>$E$(B-V)</th>
<th>r [pc]</th>
<th>Log N(H$_2$) [cm$^{-2}$]</th>
<th>Log N(H$_2$) [cm$^{-2}$]</th>
<th>Log N(H$_2$) [cm$^{-2}$]</th>
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<td>162</td>
<td>-37</td>
<td>B1 Ib</td>
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</tr>
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<td>07.3 III IIn &amp; f</td>
<td>.33</td>
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<td>21.30</td>
</tr>
<tr>
<td>28497</td>
<td>η Per</td>
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<td>-37</td>
<td>B1.5 Ve</td>
<td>.01</td>
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<td>14.82</td>
<td>20.20</td>
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<tr>
<td>30514</td>
<td>α Cam</td>
<td>144</td>
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<td>B9.5 Ia</td>
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<td>1184</td>
<td>20.36</td>
<td>20.90</td>
<td>21.09</td>
</tr>
</tbody>
</table>

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**[e$^-$/[H$_2$]] not to blame**

**N(C$^+$) from HST**

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Big Problem with the Chemistry!

Steady State: \[ [H_3^+] = \frac{\zeta [H_2]}{k_e [e^-]} \]

To increase the value of \([H_3^+]\), we need:

- Smaller electron fraction \([e^-]/[H_2]\)
- Smaller recombination rate constant \(k_e\)
- Higher ionization rate \(\zeta\)
# H$_3^+$ Dissociative Recombination

Table II. Experimental rate constants of dissociative recombination of H$_3^+$

<table>
<thead>
<tr>
<th>$k_e$ (in $10^{-7}$cm$^3$ s$^{-1}$)</th>
<th>Method$^a$</th>
<th>Authors</th>
<th>Year</th>
</tr>
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<tbody>
<tr>
<td>25$^b$</td>
<td>MA</td>
<td>Biondi, Brown$^{19}$</td>
<td>1949</td>
</tr>
<tr>
<td>20$^b$</td>
<td>MA</td>
<td>Richardson, Holt$^{21}$</td>
<td>1951</td>
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<td>3.4$^b$</td>
<td>MA</td>
<td>Varnerin$^{22}$</td>
<td>1951</td>
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<td>&lt;0.3$^b$</td>
<td>MA</td>
<td>Persson, Brown$^{23}$</td>
<td>1955</td>
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<tr>
<td>2.3</td>
<td>MA/MS</td>
<td>Leu, Biondi, Johnsen$^{24}$</td>
<td>1973</td>
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<tr>
<td>2.5</td>
<td>IB</td>
<td>Peart, Dolder$^{25}$</td>
<td>1974</td>
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<td>2.1</td>
<td>MB</td>
<td>Auerbach et al.$^{26}$</td>
<td>1977</td>
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<td>1.5</td>
<td>IT</td>
<td>Mathur, Khan, Hasted$^{27}$</td>
<td>1978</td>
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<td>4.2</td>
<td>MB</td>
<td>McGowan et al.$^{28}$</td>
<td>1979</td>
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<td>1.6</td>
<td>FA</td>
<td>MacDonald, Biondi, Johnsen$^{29}$</td>
<td>1984</td>
</tr>
<tr>
<td>&lt;0.2</td>
<td>FALP</td>
<td>Adams, Smith, Alge$^{30}$</td>
<td>1984</td>
</tr>
<tr>
<td>&lt;0.0001</td>
<td>FALP$^c$</td>
<td>Smith, Adams$^{31}$</td>
<td>1987</td>
</tr>
<tr>
<td>0.2</td>
<td>MB</td>
<td>Hus et al.$^{32}$</td>
<td>1988</td>
</tr>
</tbody>
</table>
Low rate widely believed

- Chemical models (van Dishoeck & Black)
- Grain neutralization models

4.2. Observations of $H_3^+$

The detection of $H_3^+$ in diffuse gas has had some ironic twists. Chemical models employed in the original discussion of grain neutralization (Lepp & Dalgarno 1988a; Lepp et al. 1988) were poisoned by inclusion of the then-fashionable assumption of a very small low-temperature gas-phase recombination rate for $H_3^+$. This led the authors to a gross overprediction of $N(H_3^+)$, which they suggested would be observable; their suggestion seems not to have been acted upon in a timely manner, thus depriving the world of a seeming corroboration of the incorrect recombination rate. Instead, the recombination rate was corrected (Amano 1988; Larsson et al. 1993; Sundstrom et al. 1994), lowering expectations for the presence of $H_3^+$, and, by the time it was widely detected, this was considered surprising. With substantial fluctuations, $N(H_3^+)/E_{B-V} \approx$
IS THE DISSOCIATIVE RECOMBINATION OF $\text{H}_3^+$ REALLY SLOW? A NEW SPECTROSCOPIC MEASUREMENT OF THE RATE CONSTANT

T. AMANO
Herzberg Institute of Astrophysics, National Research Council
Received 1987 December 14; accepted 1988 March 14

ABSTRACT

The decay of an infrared absorption signal of $\text{H}_3^+$ was measured as a function of time. The decay curve was analyzed and found to fit very well to the form expected for a recombination decay. The signal decay is attributed to the dissociative recombination with electrons and the rate constant was determined to be $(1.8 \pm 0.2) \times 10^{-7}$ cm$^3$ s$^{-1}$, which disagrees with the recent value ($\leq 2 \times 10^{-8}$ cm$^3$ s$^{-1}$) obtained with the flowing afterglow/Langmuir probe (FALP) technique.

Subject headings: interstellar; molecules — molecular processes

Adams, Smith, and Algel (1984), on the other hand, obtained a much smaller value for the rate constant ($\leq 2 \times 10^{-8}$ cm$^3$ s$^{-1}$) by using the flowing afterglow/Langmuir probe (FALP) technique, and later they reached the conclusion that the rate is immeasurably small ($\sim 10^{-11}$ cm$^3$ s$^{-1}$) (Adams and Smith 1987). A theoretical calculation also suggested a small rate constant (Michels and Hobbs 1984). Also very recently Hus et al. (1988) repeated the merged beam experiments and obtained the cross section which is about an order of magnitude smaller than the previous value (Auerbach et al. 1977). These authors attributed the faster rate constants previously obtained to vibrationally excited $\text{H}_3^+$. Considering the astrophysical impact of these low values, we have carried out direct measurements of the decay of the infrared absorption signals of $\text{H}_3^+$, which can monitor the ion abundance in a particular vibration-rotation state without ambiguity.
Amano's Results

**Fig. 2.**—The transient absorption signal of the \( R_3(3) \) line of \( H_3^+ \) (top trace) and the discharge current (bottom trace). The hydrogen pressure was 500 mtorr.

**Fig. 3.**—The \( 1/N_+(t) \) vs. \( t \) plot of a decay signal of \( H_3^+ \). The origin of the time is taken at the point where the current falls to zero completely, as shown in Fig. 2. The ion concentration at \( t = 0 \) is measured to be \( 3 \times 10^{12} \) cm\(^{-3}\) in this example. The peak concentration is larger than that at \( t = 0 \) by about 30\%, as seen from Fig. 2.

\[
dN_+/dt = -kN_+^2
\]

\[
1/N_+(t) = 1/N_+(0) + kt
\]
Amano’s Analysis

IV. DISCUSSION

The rate constant was found to be unchanged in cathodes with different diameters, indicating that the ambipolar diffusion process is not a dominant depletion process. Our tech-

Since the dissociative recombination rate constant for H$_3^+$ obtained in the present experiment was very different from that obtained by Smith and his co-workers (Adams, Smith, and Alge 1984; Smith and Adams 1984; Adams and Smith 1987), we carefully checked whether any serious impurity (N$_2$ is the most serious one) might cause the decay of H$_3^+$ faster than the dissociative recombination with electrons. This possibility was ruled out for two reasons. First, by monitoring the signal of HN$_2^+$ in a “pure” hydrogen discharge, we concluded that the abundances of N$_2$ was $2 \times 10^{11}$ cm$^{-3}$ at most and was negligibly small. Second, the decay curves fit very well to the recombination decay curve given by equation (3). If constant leak or back diffusion causes the depletion of H$_3^+$, the decay should be exponential. Also if the hydrogen gas contains noncondensable impurities like CH$_4$ which react with H$_3^+$, the decay rate should show a linear dependence on the hydrogen pressure.

The dissociative recombination of H$_3^+$ is known to be rapid ($k_e = 3 \times 10^{-6}$ cm$^3$ s$^{-1}$) (Leu, Biondi, and Johnsen 1973). Small amount of H$_4^+$ may exist, although the spectroscopic detection of this species has never been successful under our experimental conditions. The dominant formation process of H$_5^+$ is

$$H_3^+ + H_2 + H_2 \rightarrow H_5^+ + H_2,$$

and the rate constant of this reaction was measured to be $9 \times 10^{-30}$ cm$^6$ s$^{-1}$ (Hiraoka and Kebler 1973), which gives the rate for the reaction (7) to be about $1 \times 10^6$ s$^{-1}$ at the hydrogen pressure of 1 torr. This rate is too slow to explain the decay observed in this investigation. Also if this process (7) is dominant, then the decay rate should have the hydrogen pressure dependence.

Smith and co-workers (Adams, Smith, and Alge 1984; Smith and Adams 1984) argued that the H$_3^+$ in the ground vibrational state does not react with electrons and the fast decay obtained previously might have been caused by vibrationally excited H$_3^+$. Our spectroscopic measurements are state specific. The absorption signal measured in the present experiment monitored the population decay of the $J = 3, K = 3$ rotational level in the ground vibrational state. In the pressure range of our experiments, the rotational equilibration is completed in a few microseconds after the discharge is terminated. Also the vibrational temperature is low and the population of the first excited vibrational state ($v_2 = 1$) is estimated not to exceed 1% and therefore the effect of the vibrational relaxation is negligible. We plan further measurements of the rate constant of H$_3^+$ and other ions at lower temperatures.
Amano 1990

TABLE I. The dissociative recombination rate coefficients of $\text{H}_3^+$ in the ground vibrational state (in units of $10^{-7}$ cm$^3$ s$^{-1}$).

<table>
<thead>
<tr>
<th>$J, K$</th>
<th>110 K</th>
<th>210 K</th>
<th>273 K</th>
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<tr>
<td>1, 0</td>
<td>4.1(2)*</td>
<td>2.5(1)</td>
<td>1.72(5)</td>
</tr>
<tr>
<td>1, 1</td>
<td>4.1(1)</td>
<td>2.7(2)</td>
<td>1.77(10)</td>
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<tr>
<td>2, 2</td>
<td>4.6(4)</td>
<td>2.4(2)</td>
<td>1.85(6)</td>
</tr>
<tr>
<td>3, 3</td>
<td>4.5(5)</td>
<td>2.6(2)</td>
<td>1.91(7)</td>
</tr>
<tr>
<td>4, 4</td>
<td>⋅⋅⋅</td>
<td>2.2(2)</td>
<td>1.9(4)</td>
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</tbody>
</table>

*Standard deviation in units of the last quoted digits

if any. Moreover, because the rotational relaxation is much faster than the recombination decay, only the rotationally averaged rate coefficient is possible to be observed.

**CONCLUSION**

Our spectroscopic measurements have clearly established that the dissociative recombination rate coefficient of $\text{H}_3^+$ in the ground state is not as small as advocated by Smith and co-workers, being in good agreement with the results obtained with the microwave afterglow and other techniques. Measurements have been extended to $\text{HN}_2^+$ and
H$_3^+$ Dissociative Recombination

Table II. Experimental rate constants of dissociative recombination of H$_3^+$

<table>
<thead>
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<th>Rate (cm$^3$ molecule$^{-1}$ s$^{-1}$)</th>
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<tr>
<td>0.2</td>
<td>MB</td>
<td>Hus et al.$^{32}$</td>
<td>1988</td>
</tr>
<tr>
<td>1.8</td>
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<td>1988</td>
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<td>$\leq0.0001$</td>
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<td>Adams, Smith$^{34}$</td>
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<td>Smith, Adams, Ferguson$^{35}$</td>
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<td>Féher, Rohrbacher, Maier$^{40}$</td>
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<td>$&lt;0.03$</td>
<td>ISA</td>
<td>Glosík$^{44}$</td>
<td>2001</td>
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</table>

- Theory unreliable (until recently)...
- Still not measuring H$_3^+$ in ground rotational states

Oka (2003)
Storage Ring Measurements

CRYRING

30 kV
900 keV
12.1 MeV

Very simple experiment
Complete vibrational relaxation
Control $H_3^+$ – e$^-$ impact energy
Rotationally hot ions produced
“No” rotational cooling in ring
Supersonic Expansion Ion Source

- Similar to sources for laboratory spectroscopy in many groups
- Pulsed nozzle design
- Supersonic expansion leads to rapid cooling
- Discharge from ring electrode downstream
- Spectroscopy used to characterize ions
- Skimmer employed to minimize arcing to ring
H$_3^+$ Energy Level Structure

probe of temperature

not detected
Spectroscopy of H$_3^+$ Source

Infrared Cavity Ringdown Laser Absorption Spectroscopy

- Confirmed that H$_3^+$ produced is rotationally cold, as in interstellar medium

CRYRING Results

- Considerable amount of structure (resonances) in the cross-section
- $k_e = 2.6 \times 10^{-7} \text{ cm}^3 \text{ s}^{-1}$
- Factor of two smaller

McCall, et al.  
Recent Theoretical Work

Theory of Dissociative Recombination of $D_{3h}$ Triatomic Ions Applied to $H_3^+$

Viatcheslav Kokouline and Chris H. Greene

Department of Physics and JILA, University of Colorado, Boulder, Colorado 80309-0440

(Received 3 December 2002; published 3 April 2003)
High-Resolution Dissociative Recombination of Cold H$_3^+$ and First Evidence for Nuclear Spin Effects


<table>
<thead>
<tr>
<th>CRYRING</th>
<th>TSR</th>
</tr>
</thead>
<tbody>
<tr>
<td>Supersonic expansion</td>
<td>RF 22-pole ion trap @ 13 K</td>
</tr>
</tbody>
</table>

| Electron target | kT$_\perp$ $\sim$ 2 meV | n$_e$ $\sim$ 6.3$\times$10$^6$ cm$^{-3}$ |
| Electron cooler | (same as target) | kT$_\perp$ $\sim$ 10.5 meV | n$_e$ $\sim$ 4.5$\times$10$^5$ cm$^{-3}$ |
| Beam energy    | 12.1 MeV            | 5.25 MeV            |
Back to the Interstellar Clouds!

Steady State: \[ [H_3^+] = \frac{\zeta}{k_e} \frac{[H_2]}{[e^-]} \]

To increase the value of \([H_3^+]\), we need:

- Smaller electron fraction \([e^-]/[H_2]\)
- Smaller recombination rate constant \(k_e\)
- Higher ionization rate \(\zeta\)
Implications for $\zeta$ Persei

$$\frac{N(H_3^+)}{L} = [H_3^+] = \frac{\zeta}{k_e} \frac{N(H_2)}{N(e^-)}$$

$$\zeta \, L = (2.6 \times 10^{-7} \text{ cm}^3 \text{ s}^{-1}) \frac{N(H_3^+)}{N(H_2)} \frac{N(e^-)}{(3.8 \times 10^{-4})}$$

$$\zeta \, L = 8000 \text{ cm s}^{-1} \text{ (solid)}$$

Adopt

$\zeta = 3 \times 10^{-17} \text{ s}^{-1}$

$L = 85 \text{ pc}$

$\langle n \rangle = 6 \text{ cm}^{-3}$

Adopt

$\zeta = 1.2 \times 10^{-15} \text{ s}^{-1}$

$L = 2.1 \text{ pc}$

(40x higher!)
What Does This Mean?

- Enhanced ionization rate in ζ Persei
- Widespread H₃⁺ in diffuse clouds
  - perhaps widespread ionization enhancement?
- Dense cloud H₃⁺ is "normal"
  - enhanced ionization rate only in diffuse clouds
  - low energy cosmic-ray flux?
  - cosmic-ray self-confinement?
  - no constraints, aside from chemistry!!
- New chemical models necessary
  - Harvey Liszt
  - Franck Le Petit
\( \text{H}_3^+ \) and other species in the diffuse cloud towards \( \zeta \) Persei: A new detailed model

F. Le Petit\(^1\)^, E. Roueff\(^1\), and E. Herbst\(^3\)

- Parameters: \( n=100 \text{ cm}^{-3} \), \( L=4.2 \text{ pc} \), \( T=60 \text{ K} \)
- Matches all observations within a factor of 3
- \( \zeta = 2.5 \times 10^{-16} \text{ s}^{-1} \)
  - 10\( \times \) canonical value
- OH not a problem
  - \( \text{H}^+ + \text{O} \rightarrow \text{O}^+ + \text{H} \)
    endothermic by 227 K
  - OH lowered: \( T \rightarrow 60 \text{ K} \)
- Still underpredicts \( \text{H}_3^+ \)
  - “Proof of concept”
CONFINEMENT-DRIVEN SPATIAL VARIATIONS IN THE COSMIC-RAY FLUX

PAOLO PADOAN¹ AND JOHN SCALO²

Received 2004 September 16; accepted 2005 March 30; published 2005 April 13

ABSTRACT

Low-energy cosmic rays (CRs) are confined by self-generated MHD waves in the mostly neutral interstellar medium. We show that the CR transport equation can be expressed as a continuity equation for the CR number density involving an effective convection velocity. Assuming a balance between wave growth and ion-neutral damping, this equation gives a steady state condition $n_{cr} \propto n_j^{1/2}$ up to a critical density for free streaming. This relation naturally accounts for the heretofore unexplained difference in CR ionization rates derived for dense diffuse clouds (McCall et al.) and dark clouds, and predicts large spatial variations in the CR heating rate and pressure.
Future Work

• More experiments!
  – Improved spectroscopy of ion source
    • Higher resolution & higher sensitivity
    • Better characterization of ro-vib distribution
  – Testing of new (piezo) ion source
  – Single quantum-state CRYRING measurements
    • produce pure para-H$_3^+$ using para-H$_2$

• More observational data!
  – Search for H$_3^+$ in more diffuse cloud sightlines
    • Confirm generality of result in classical diffuse clouds
  – Observations of H$_3^+$ in "translucent" sightlines
    • C$^+$ $\rightarrow$ C $\rightarrow$ CO
Rich Diffuse Cloud Chemistry

- From 1930s through the mid-1990s, only diatomic molecules thought to be abundant in diffuse clouds
- Recently, many polyatomics observed:
  - $\text{H}_3^+$ in infrared
  - $\text{HCO}^+$, $\text{C}_2\text{H}$, $\text{C}_3\text{H}_2$, etc. in radio (Lucas & Liszt)
  - $\text{C}_3$ in near-UV (Maier, et al.)
- Diffuse Interstellar Bands!
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