H_3^+ Destruction Rates Due to Ambipolar Diffusion in AC Positive Column Discharges

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Abstract: One of the major destruction paths of ions in laboratory plasmas is the electron recombination of ions at the walls of the discharge tube. In positive column discharges, ions drift toward the wall by ambipolar diffusion. In order to analyze the chemistry in plasmas quantitatively, direct measurements of destruction rates due to this process are needed. In this work, we studied variations in H_3^+ concentration in plasmas when small amounts of CH_4, N_2 and CO were added under various discharge conditions. The concentration was measured from intensities of the H_3^+ spectra observed in a liquid nitrogen cooled positive column discharge using a color center laser and the velocity modulation method. The destruction rate due to ambipolar diffusion was obtained using a steady state model and previously reported ion-neutral reaction rate constants.

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H_3^+ In H_2 Dominated Discharges

- Predominant ion
- Large role in formation of other ions

  ex: $H_3^+ + CH_4 \longrightarrow CH_5^+ + H_2$

- Understanding from chemical models

  ex: CH_5^+ vs. CH_4^+

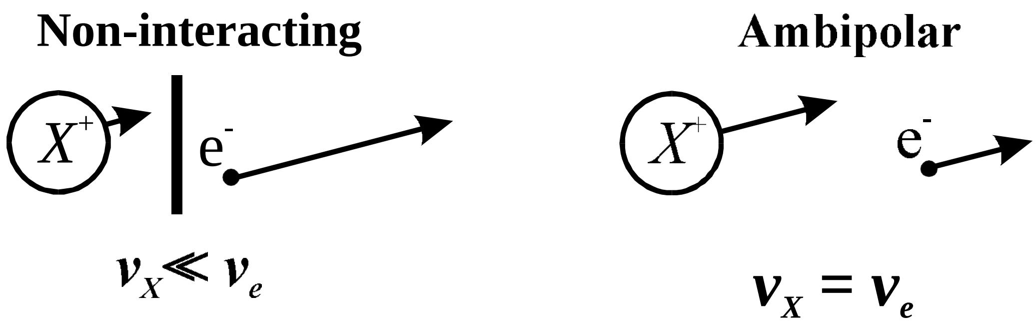
- Primary destruction: electron recombination at walls of cell

$$H_3^+ + e^- \xrightarrow{\text{Wall}} H + H + H$$

Rate poorly quantified

Ambipolar Diffusion

- Ions drift faster due to electron interaction



- Few studies in weakly ionized plasmas

- Direct measurement of ambipolar diffusion difficult

- Estimated by radial electric field measurements to be ~ 10^5 $\sec^{-1}$

Direct approach...

Study the primary destruction rate by supplying a secondary destruction mechanism of the same magnitude

Dominant Reactions

Formation:

$$H_2 + e^- \longrightarrow H_2^+ + 2e^- \qquad k_{EB} = 2.0 \times 10^{-9}\ \text{cm}^3/\text{sec}$$

$$H_2^+ + H_2 \longrightarrow H_3^+ + H \qquad k_{Form} = 2.0 \times 10^{-9}\ \text{cm}^3/\text{sec}$$

Destruction:

$$H_3^+ + X \longrightarrow XH^+ + H_2 \qquad k_{Imp} = \sim 2 \times 10^{-9}\ \text{cm}^3/\text{sec}$$

$$\{X = CH_4,\ N_2,\ \text{or } CO\}$$

$$H_3^+ + e^- \xrightarrow{\text{Wall}} H + H + H \qquad k_{AD} = ???\ \sim 10^4 - 10^6\ \text{sec}^{-1}$$

Steady State Model

$$\frac{d[\mathrm{H}_2^+]}{dt} = k_{EB}[\mathrm{e}^-][\mathrm{H}_2] - [\mathrm{H}_2^+]\{k_{Form}[\mathrm{H}_2]\} = 0 \quad (1)$$

$$\frac{d[\mathrm{H}_3^+]}{dt} = k_{Form}[\mathrm{H}_2^+][\mathrm{H}_2] - [\mathrm{H}_3^+]\{[X]k_{Imp} + k_{AD}\} = 0 \quad (2)$$

From (2)

$$[\mathrm{H}_3^+] = \frac{k_{Form}[\mathrm{H}_2][\mathrm{H}_2^+]}{(k_{Imp}[X] + k_{AD})}$$

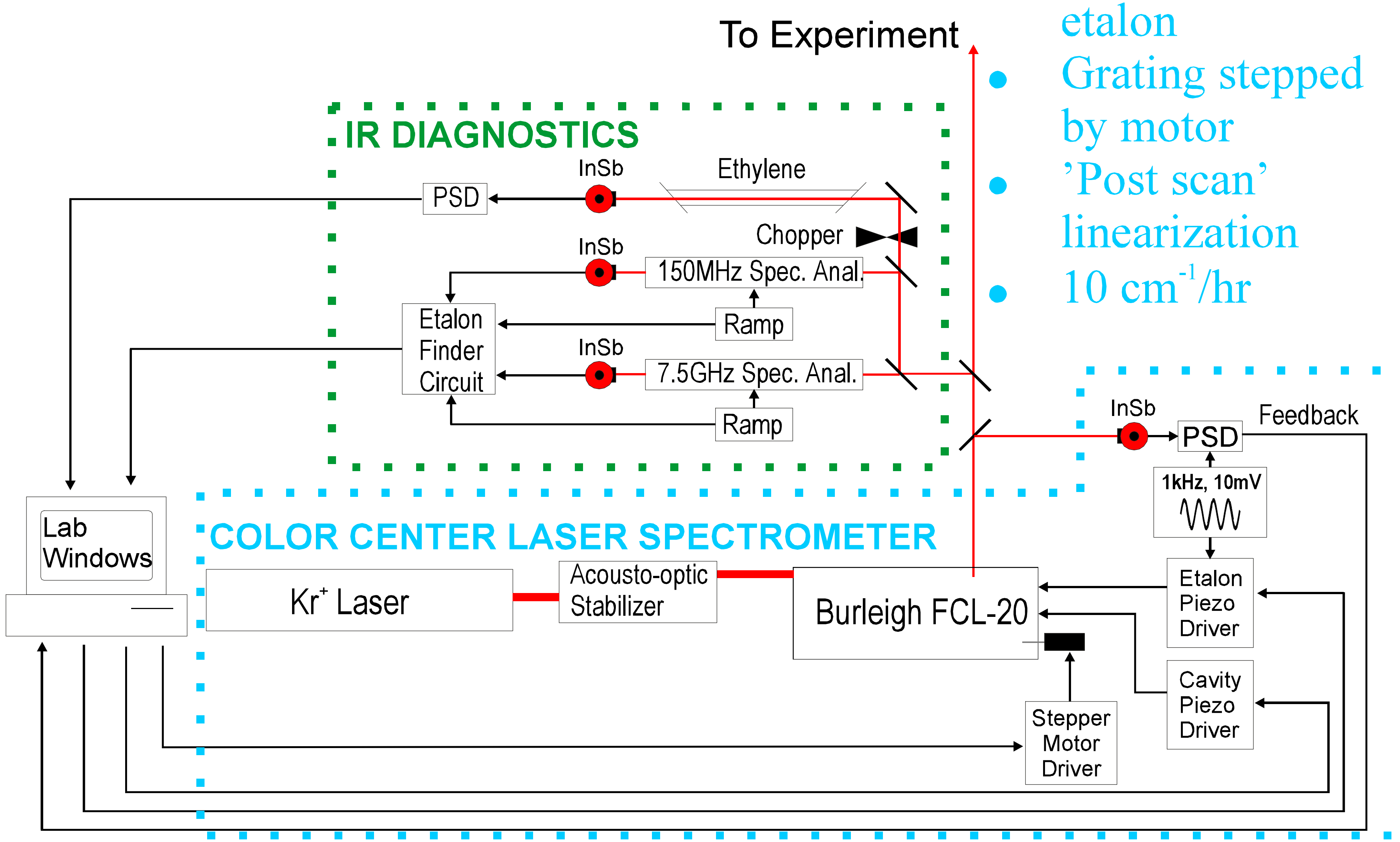
Rearrange and insert (1)

$$[\mathrm{H}_3^+] = \frac{k_{EB}[\mathrm{e}^-][\mathrm{H}_2]}{(k_{Imp}[X] + k_{AD})}$$

Ratio with and without impurity

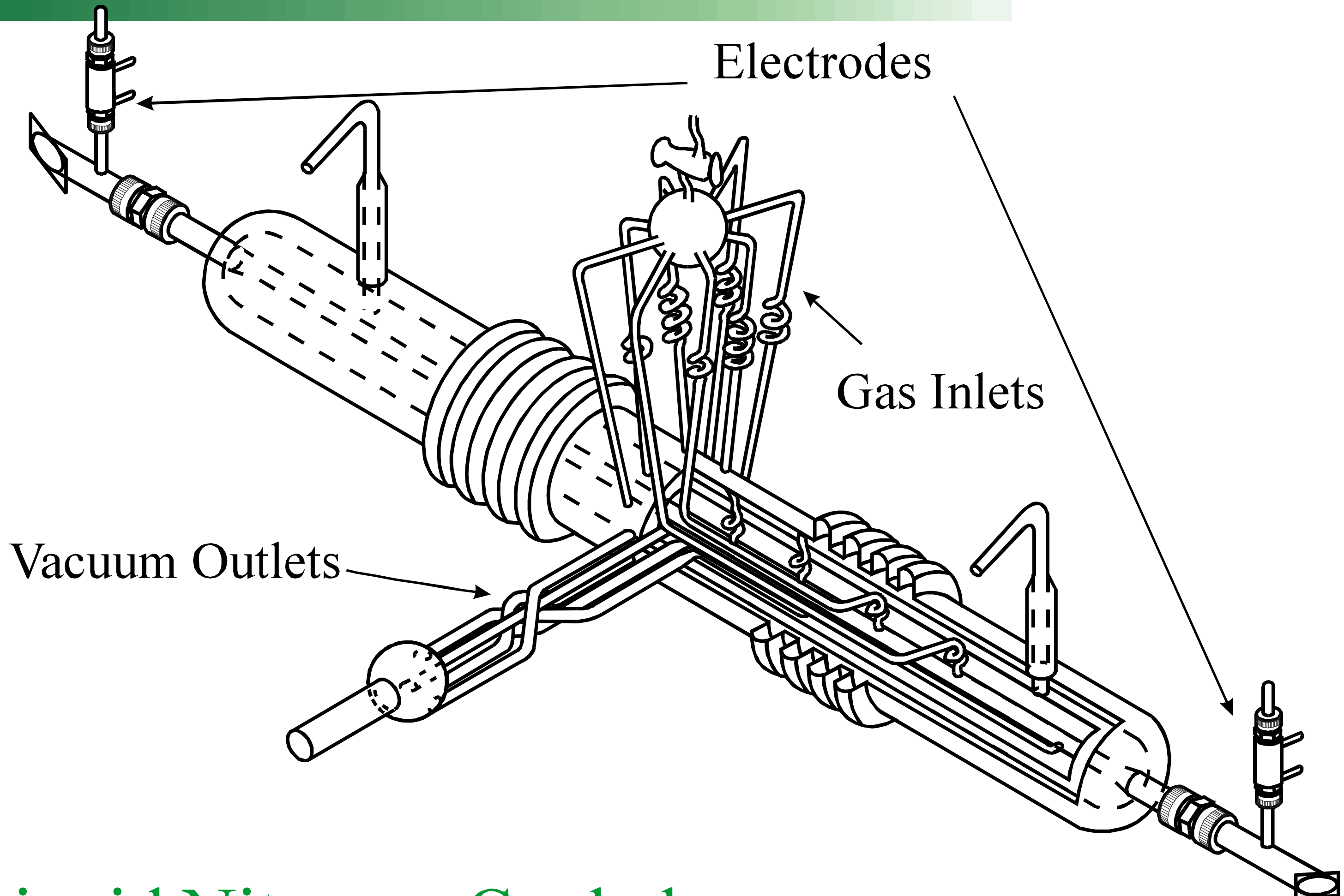
$$\frac{[\mathrm{H}_3^+]_{[X]=0}}{[\mathrm{H}_3^+]_{[X]}} = \frac{k_{Imp}}{k_{AD}}[X] + 1$$

Infrared Spectrometer



- Cavity locked to etalon
- Grating stepped by motor
- ’Post scan’ linearization
- 10 cm^{-1}/hr

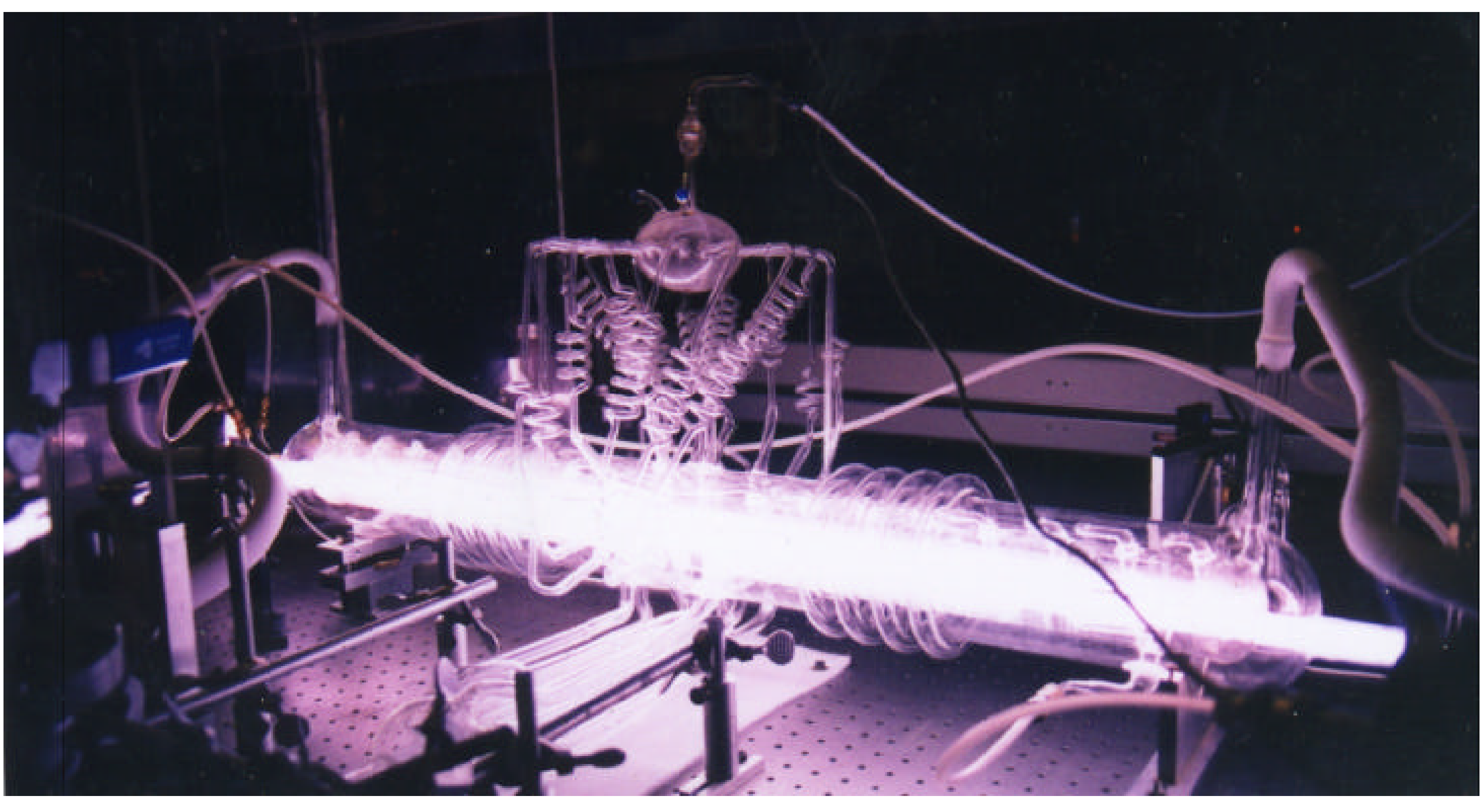
Positive Column Discharge Tube



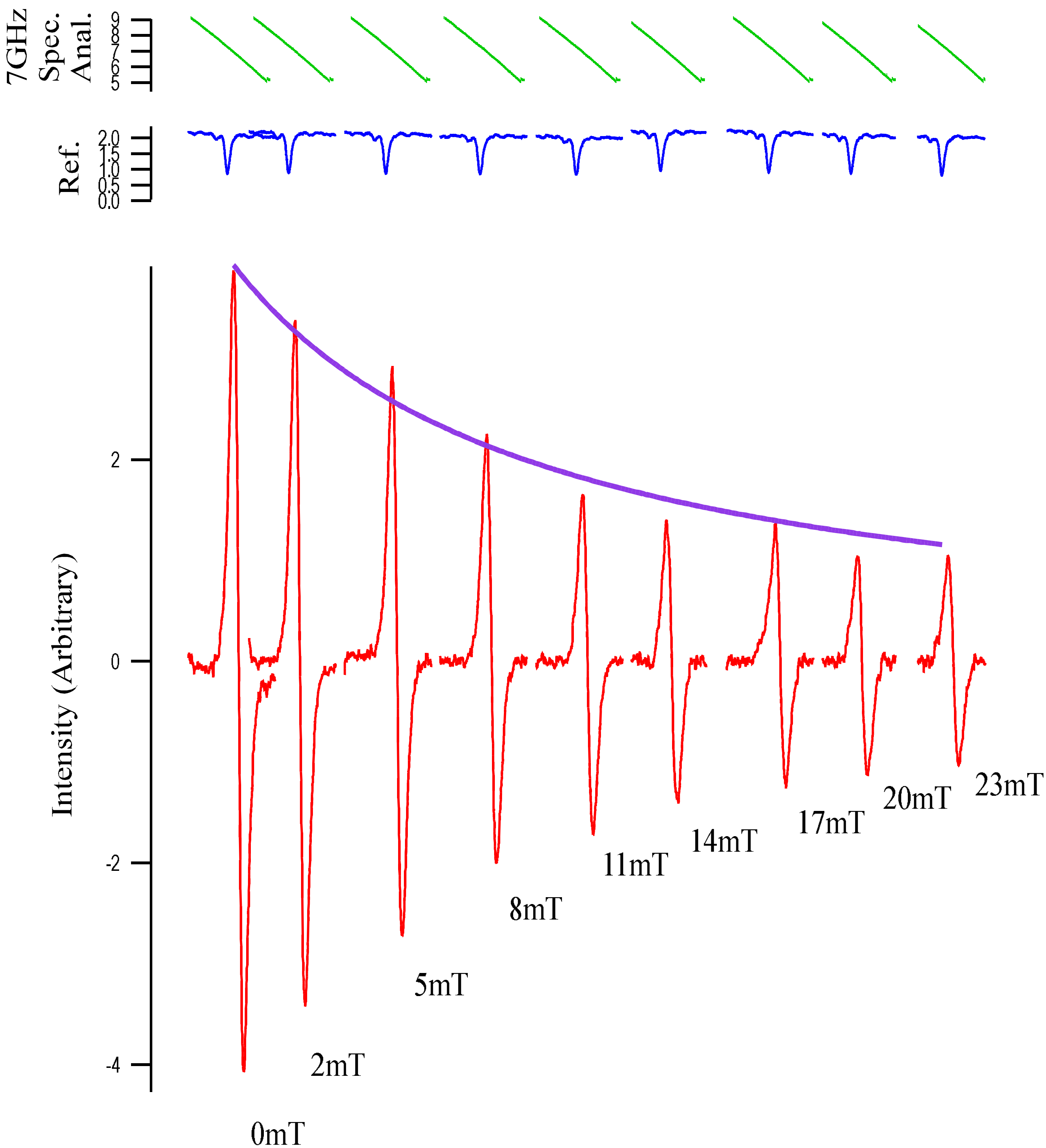
- Liquid Nitrogen Cooled
- 1 cm Diameter Inner Bore
- Discharge Column Length, ~125 cm
- 200 L/Min Flow Rate

Experimental Conditions

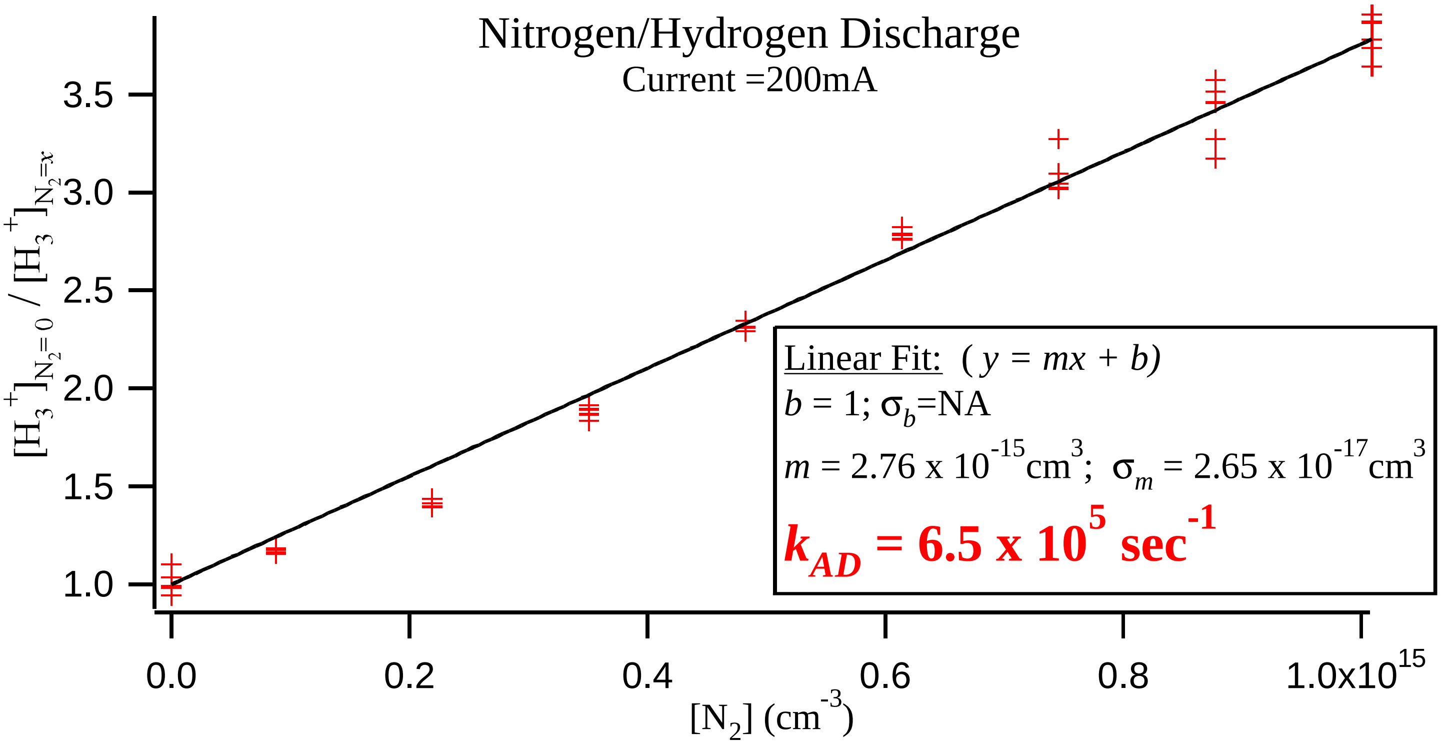
- H_3^+ R(4,3)$^-$ @ 3014.258 cm^{-1}
- Velocity modulation technique
- 0.5 Torr H_2
- ~5kV sine wave @ 6 kHz
- 150mA, 175mA, and 200 mA
- N_2 ,CH_4, and CO 0-23mTorr impurity

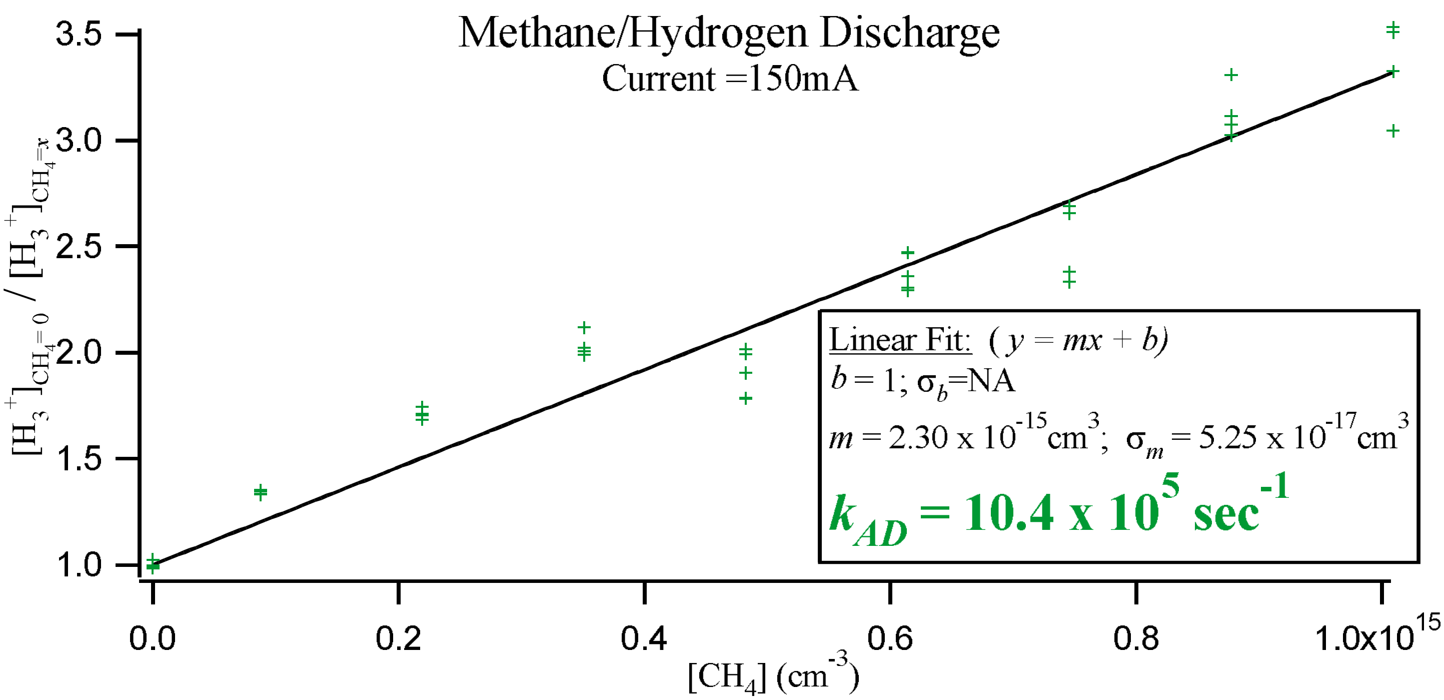


H_3^+ Signal Upon Addition of N_2



Destruction Rate Plots





$$\frac{[H_3^+]_{[X]=0}}{[H_3^+]_{[X]}} = \frac{k_{Imp}}{k_{AD}}[X] + 1$$

Summary

Impurity	Current	k_{AD}
N_2	200 mA	6.9×10^5 sec^{-1}
	175 mA	6.2×10^5 sec^{-1}
	150 mA	6.1×10^5 sec^{-1}
CO	200mA	8.5×10^5 sec^{-1}
	175 mA	8.5×10^5 sec^{-1}
	150 mA	9.0×10^5 sec^{-1}
CH_4	200 mA	10.3×10^5 sec^{-1}
	175 mA	10.3×10^5 sec^{-1}
	150 mA	10.4×10^5 sec^{-1}

mean = **7.8×10^5 sec^{-1}**

Uncertainties k_{Imp}? *

N_2 $k_{Imp} = 1.9 \times 10^{-9}$ cm^{3}sec^{-1}

CO $k_{Imp} = 1.8 \times 10^{-9}$ cm^{3}sec^{-1}

CH_4 $k_{Imp} = 2.4 \times 10^{-9}$ cm^{3}sec^{-1}

* *Claim ~20% Error. Taken from* V.G. Anicich and W.T. Huntress, *Astrophys. J. Supl. Ser* **62**,553 (1986)

Error in impurity number density?

Dissociation?
Polymerization?
Radial concentration gradient?