Rotational and vibrational cooling of $\text{H}_3^+$ in laboratory experiments

Holger Kreckel, Dirk Schwalm, Daniel Zajfman, Andreas Wolf
+ the TSR Group
Max-Planck-Institut für Kernphysik, Heidelberg, Germany

Jonathan Tennyson
Department of Physics and Astronomy
University College London, UK
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Outline

- Motivation,
- TSR Experiments,
- Relaxation model,
  - Vibrational decay,
  - Longlived rotational states,
  - Radiative heating,
- Conclusions.
Motivation: the $\text{H}_3^+$ DR dilemma

Dissociative Recombination (DR)

$\text{H}_3^+ + e^- \rightarrow \{ \text{H}_2 + \text{H}, \text{H} + \text{H} + \text{H} \}$

[Graph showing DR rate coefficient over time with upper limits and storage rings highlighted]
The TSR storage ring

Ultra-high vacuum system

H$_3^+$ formation:

H$_2^+$ + H$_2$ → H$_3^+$ + H + 1.7eV!
H$_3^+$ vibrational excitation

- **Bending mode 1**
- **Breathing mode**
- **Bending mode 2**

Phase:
- +90°
- -90°
H$_3^+$ vibrational levels
Coulomb Explosion Imaging Technique (CEI)

$v = 0.03c$

target foil
(thickness $< 10^{-8}$ m)

$\Delta t$

3D imaging detector

5 m

8 cm
Coulomb explosion principle
CEI Setup: Slow extraction
Coulomb explosion results $H_3^+$

The diagram shows the distribution of vibrational levels $P(v_a)$ as a function of $v_a$ (a.u.). The theoretical predictions for $(1,0^0)$, $(2,0^0)$, and $(0,0^0)$ are compared with experimental data for $T < 1\text{ms}$ and $T > 2\text{s}$. The data for $T > 2\text{s}$ is labeled as experimental data, while the data for $T < 1\text{ms}$ is not explicitly labeled but is included in the comparison.
Decay of the first breathing mode $(1,0^0)$ of $\text{H}_3^+$

exponential function
(breathing mode lifetime $T=1.18\,\text{s}$)
fitted to experiment
at $t > 500\,\text{ms}$
DR fragment imaging

three-body breakup

\[ \text{H}_3^+ + e^- \rightarrow \text{H} + \text{H} + \text{H} \]

reveals substantial rotational excitation

\[ T_{\text{rot}} \sim 2700 \text{K} \]

for storage times up to 60 s

D. Strasser et al., PRL 86, 779 (2001)

signature of excess energy up to 1 eV
The UCL line list for $\text{H}_3^+$

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<tr>
<th>$J_i$</th>
<th>$E_i$ (cm$^{-1}$)</th>
<th>$J_f$</th>
<th>$E_f$ (cm$^{-1}$)</th>
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Rovibrational relaxation model for $\text{H}_3^+$

Rovibrational relaxation model for $\text{H}_3^+$

- Initial temperature: $0.23\text{eV}$ (2700 K)
- Time points: $t=0\text{s}$, $t=0.1\text{s}$, $t=10\text{s}$, $t=60\text{s}$
- Diagram showing population changes over time for vibrations and rotations.
Decay of the first breathing mode \((1,0^0)\) of \(\text{H}_3^+\)

Exponential function (breathing mode lifetime \(T=1.18\text{ s}\)) fitted to experiment at \(t > 500\text{ ms}\)
Metastable rotational states (t=60 s)
Metastable rotational states (t=60 s)
ortho/para corrected

Population vs energy [eV]
Rotational levels

energy [cm$^{-1}$]

G quantum number

J - G
Selection rules

\( \Delta J = -1, 0, +1 \)
\( \Delta K = 2n + 1 \)
\( \Delta G = 3n \)

\( \Delta J = -1, 0, +1 \)
\( \Delta G = 3n, \; n \neq 0 \)
Radiative heating by 300 K blackbody radiation

initial distribution: 100 K Boltzmann
Conclusion $H_3^+$ DR:

2000

2005