Towards high-precision spectroscopy of sympathetically cooled $H_2^+$

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**Motivations**
- High-resolution spectroscopy of rovibrational transitions in $H_2^+$ or HD⁺ allows for precise tests of molecular QED and determination of $m_i/m_n$ at the $-0.01$ ppb level and shed light on the proton radius puzzle.

\[ \nu = c E_m \left( \left( \nu_p + \nu_A + \nu_b \right) + \nu (\nu_a + \nu_b + \nu + \frac{1}{2}) \right)^2 \] and \[ E_m = \frac{1}{2} \mu \omega_0^2 \]

- Comparison of $H_2^+$ and HD⁺

<table>
<thead>
<tr>
<th>Line width</th>
<th>Transition</th>
<th>Strength</th>
</tr>
</thead>
<tbody>
<tr>
<td>$10^4$ Hz</td>
<td>$\nu_A$</td>
<td>Weak two-photon transition</td>
</tr>
<tr>
<td>$10^5$ Hz</td>
<td>$\nu_B$</td>
<td>Strong two-photon transition</td>
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**Systematic effects** Light shift $1 \times 10^{-5}$

**HD⁺-two-photon spectroscopy in the Lamb-Dicke regime**

**Contributions to uncertainty:**
- $(v=0, L=2) \rightarrow (v'=1, L'=2)$ in $H_2^+$

\[ \lambda = \frac{2 \pi}{\nu} \]

**Theory:** Transition frequencies from $(v=0, L=2) \rightarrow (v'=1, L'=2)$ in $H_2^+$

\[ \nu_{v',L'} = \frac{2 \pi}{\lambda} \]

**Experimental method**

- **Step 1:** State-selective production of cold $H_2^+$ molecular ions
  - Molecular beam injects $H_2^+$ molecules into ion trap
  - $(3+1)$ Resonant multiphoton ionization (REMPI) using a pulsed laser at 303 nm at the center of an ion trap
  - $H_2^+$ is sympathetically cooled by laser cooled $Be^+$ ions

- **Step 2:** REMPD spectroscopy
  - Drive 2-photon transition from $v=0, L=2 \rightarrow v=1, L=2$ with cavity-enhanced mid-IR spectroscopy laser (9.17 µm)
  - $H_2^+$ ions excited to the $v=1$ state are dissociated by 213 nm laser and lost from the trap
  - Count number of $H_2^+$ ions before and after spectroscopy and dissociation

**Experimental setups**

- **Hyperbolic trap features:**
  - Smaller hyperbolic trap size
  - Ion trap ready
  - Micromotion minimisation by fluorescence/RF correlations
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- **Linear trap features:**
  - In situ ionisation of $H_2^+$ by electron impact or REMPI
  - Sympathetic cooling with $Be^+$
  - Detection: imaging of mixed Coulomb crystals ($Be^+ + H_2^+$)

**Conclusions and outlook**
- Theory: $m_i/m_n$ can be obtained from hydrogen molecular ion spectroscopy with 15 ppt accuracy
- Ion trap ready
- Micromotion minimisation by fluorescence/RF correlations
- $H_2^+$ spectroscopy enhancement cavity implemented
- Addition of the $H_2^+$ source to the linear trap setup
- Find best method to measure a fractionnal loss of $H_2^+$
- State selected $H_2^+$ ion creation inside the laser cooled $Be^+$ ion cloud
- Search for $H_2^+$ two-photon signal

**First $Be^+$ ion clouds**
- With $H_2^+$ ions from background gas
- Beryllium ion loading
  - with e-gun or non-resonant photoionization with 213 nm laser
  - from oven (resistive heating) or ablation (532 nm pulsed laser)

**Comparison of ion crystal images with simulations**
- For given CCD image, what is the number of $Be^+$, $H_2^+$, $H_2^+$ ions?
  - Compare with molecular dynamics simulations
  - Tickle curve (fluorescence signal depends on $H_2^+$ number)
  - Count $Be^+$ ions (blobs) with image analysis, then identify $H_2^+$ dark ions ("empty sites") in CCD image:
    - by estimating the surface
    - with pattern recognition

- MD simulated CCD image $100 \text{ Be}^+ + 10 \text{ H}_2^+$
- Blob detection of 95 Be²⁺ ions of flat crystal (CCD image)

**Contributions to uncertainty:**

- $(v=0, L=2) \rightarrow (v'=1, L'=2)$ in $H_2^+$

**State-selective production of $H_2^+$ molecular ions**
- Experimental results from hyperbolic trap setup (see "Experimental setups")

**H₂ reservoir and ion signal (arb. u.)**

- $H_2$ reservoir
- Ion signal (arb. u.)
- rem. ion fract.