Towards high-precision spectroscopy of sympathetically cooled H_2^+

Julian Schmidt[1], Thomas Louvradoux[1], Mohammad Haidar[1], Johannes Heinrich[1], Jean-Philippe Karr[1,2], Laurent Hilico[1,2]

[1]Laboratoire Kastler Brossel, Sorbonne Université, ENS, CNRS ; 4 place Jussieu, Case 74, 75005 Paris, France

[2]Département de Physique, Univ EVRY Université Paris Saclay, rue du père André Jarlan 91025 Evry, France

Laboratoire Kastler Brossel – Physique quantique et applications

Motivations

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

$$\nu = cR_\infty\left[E_{nr}(\mu_{pe}) + A_{QED}(\alpha) + A^{fs}\left(\frac{r_p}{a_0}\right)^2\right] \quad \text{and} \quad E_{nr} \propto 1/\sqrt{\mu_{pe}}$$

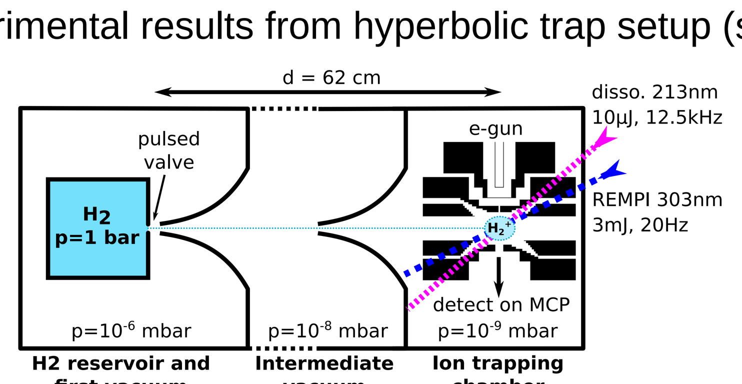
- **Comparison of H_2^+ and HD^+**

	H_2^+	HD^+
Line width	10^{-7} Hz	~ 10 Hz
Interesting for	m_p/m_e	μ/m_e ($\mu = m_d\, m_p / (m_d + m_p)$)
Transition	v=1 – v=0: Weak two-photon 2E1 transition	v=9 – v=4 – v=0: Strong two-photon transition / Rotational one-photon transition
Systematic effects	Light shift 5 x 10^{-16}	

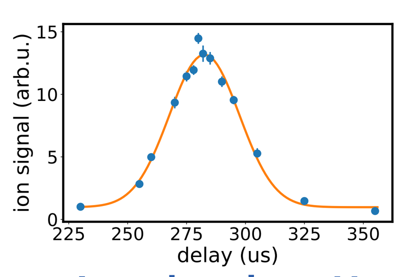
V. Tran et al., Phys. Rev. A **88**, 033421 (2013) — HD+ two-photon spectroscopy in the Lamb-Dicke regime
J.-Ph. Karr, J. Molec. Spectrosc. **300**, 37 (2014) — Hydrogen molecular ion clocks
S. Schiller et al., Phys. Rev. Lett. **113**, 023004 (2014) — Hydrogen molecular ion clocks
J.-Ph. Karr et al., Phys. Rev. A **94**, 050501 (2016) — Hydrogen molecular ions and fundamental constants
J. Biesheuvel et al., Nat. Comm. 7, 10385 (2016) — HD^+ v=0..v=8 transition frequency measurement
V.I. Korobov, L. Hilico, J.-Ph. Karr, Phys. Rev. Lett. (2017) — H_2^+ and HD^+ ionisation energies and trans. frequencies
S. Alighanbari et al., Nat. Phys. **14**, 555 (2018) — HD+ rotational spectroscopy in the Lamb-Dicke regime

State-selective production of H_2^+ molecular ions

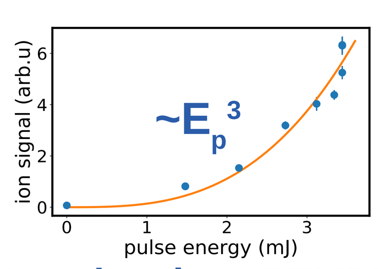
Experimental results from hyperbolic trap setup (see "Experimental setups")



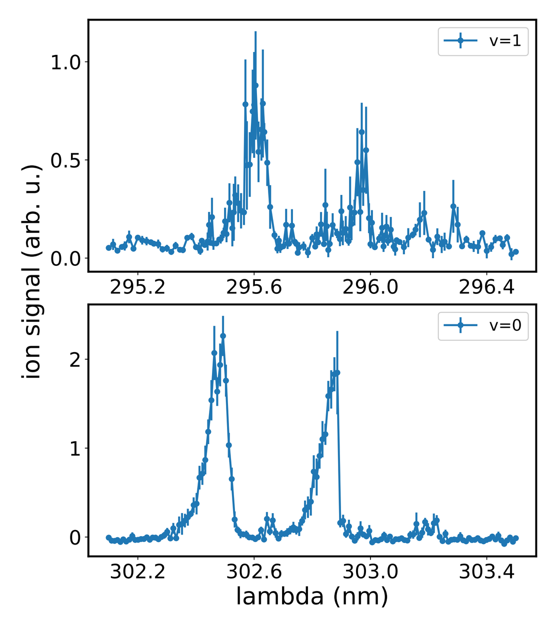
Schematic of experimental setup



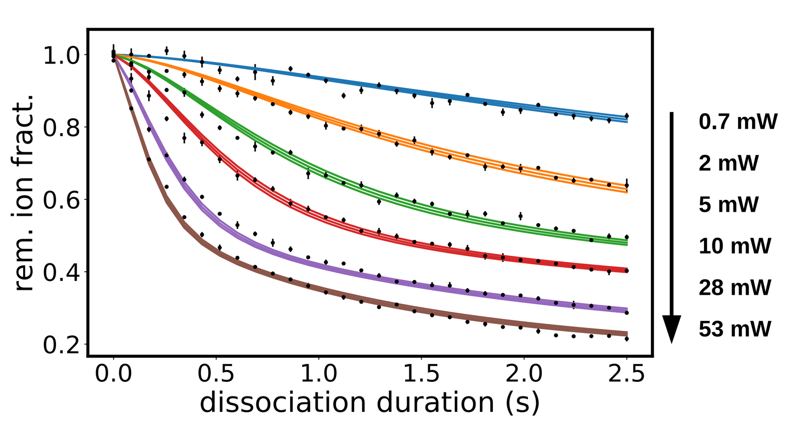
Ion signal vs. H_2 time of flight



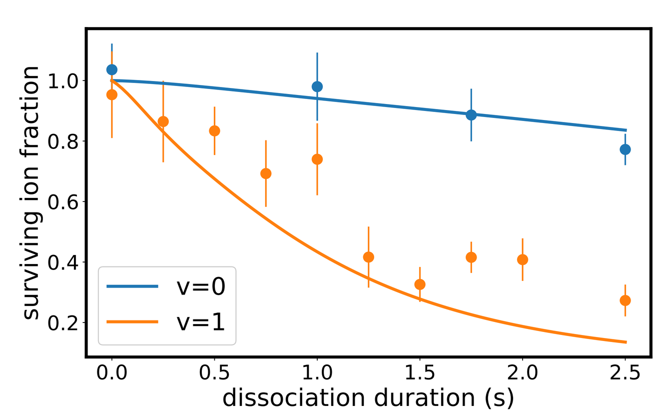
Ion signal vs. REMPI laser pulse energy



REMPI spectra for H_2^+ produced in *v=0* and *v=1*



Dissociation of H_2^+ produced by e-gun
(for increasing average laser powers from top to bottom)



Dissociation of H_2^+ produced by REMPI in *v=0* (blue) and *v=1* (orange) states
Curves: model with parameters from fit on the left (no free parameters)

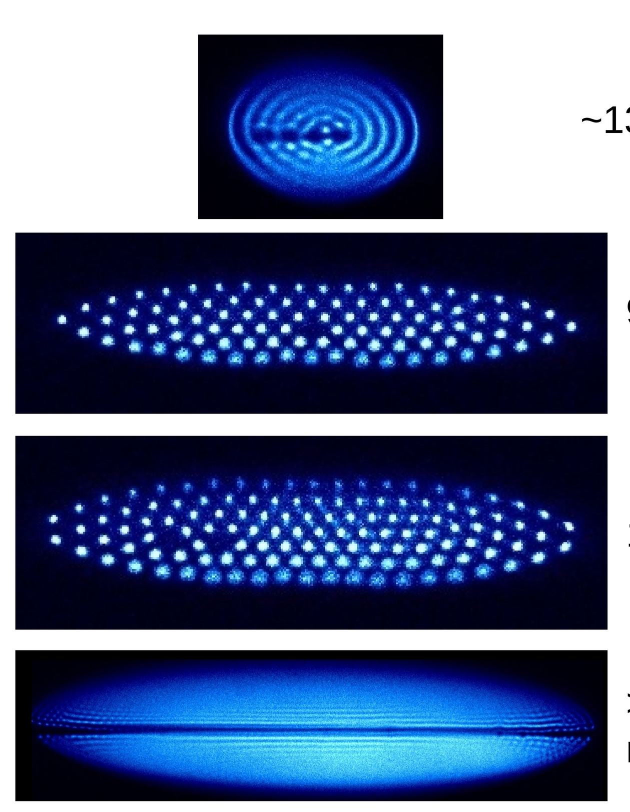
>> state-selective production and detection via dissociation method is working!

First Be^+ ion clouds

With H_2^+ ions from background gas

Beryllium ion loading

- with e-gun or non-resonant photo-ionization with 213 nm laser
- from oven (resistive heating) or ablation (532 nm pulsed laser)

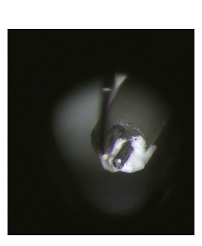


~130 Be^+ and 4 H_2^+

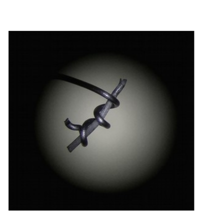
95 Be^+ and 1 H_2^+

124 Be^+ and 2 H_2^+

>1000 Be^+ and many H_2^+ and H_3^+



Be oven



Ablation target

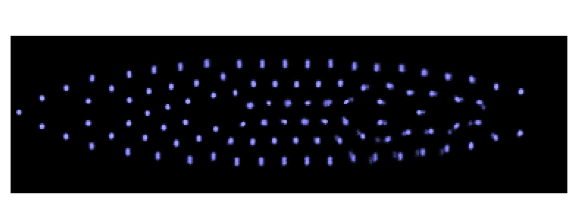
Comparison of ion crystal images with simulations

For given CCD image, what is the number of Be^+, H_2^+, H_3^+ ions?

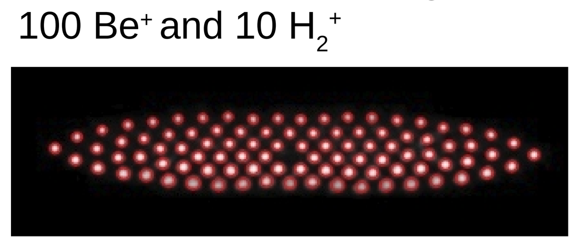
> Compare with molecular dynamics simulations

> 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 Be^+ and 10 H_2^+



Blob detection of 95 Be^+ ions of flat crystal (CCD image)

Theory: Transition frequencies from (v = 0, L=2) → (v'=1, L'=2) in H_2^+

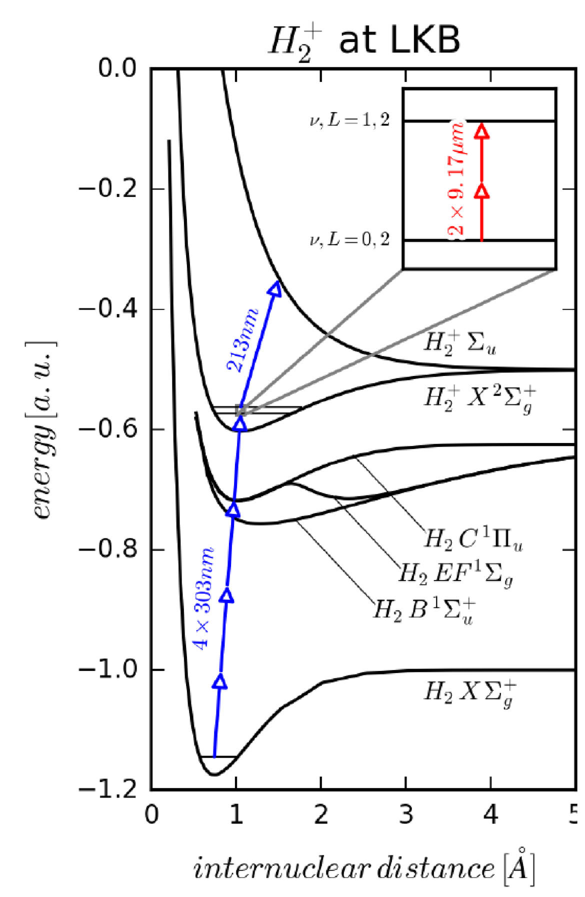
ν_{nr}	65 412 414 314.5
ν_{α^2}	1 077 263.8
ν_{α^3}	−274 145.4
ν_{α^4}	−1 935.5(1)
ν_{α^5}	120.8(1)
ν_{α^6}	−2.3(5)
ν_{tot}	65 413 215 616.1(5)

$7.6\ 10^{-12}$

Contributions to uncertainty:
(5) theory, (20) m_p/m_e, (11) r_p / Ry
CODATA 2018

- Hyperfine and QED corrections
- Combine H, D, H_2^+, HD^+ spectroscopy data to determine m_p/m_e, μ/m_e, Ry, r_p, r_d

Experimental method



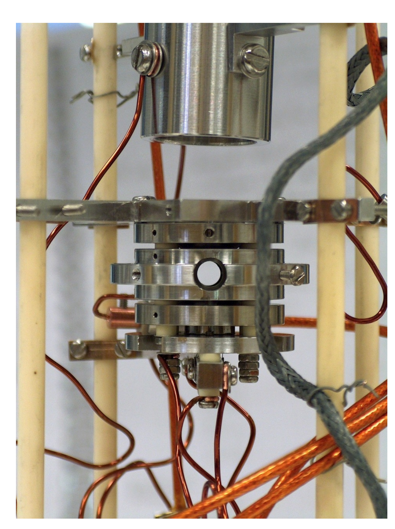
Step 1: State-selective production of cold H_2^+ molecular ions

- 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→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



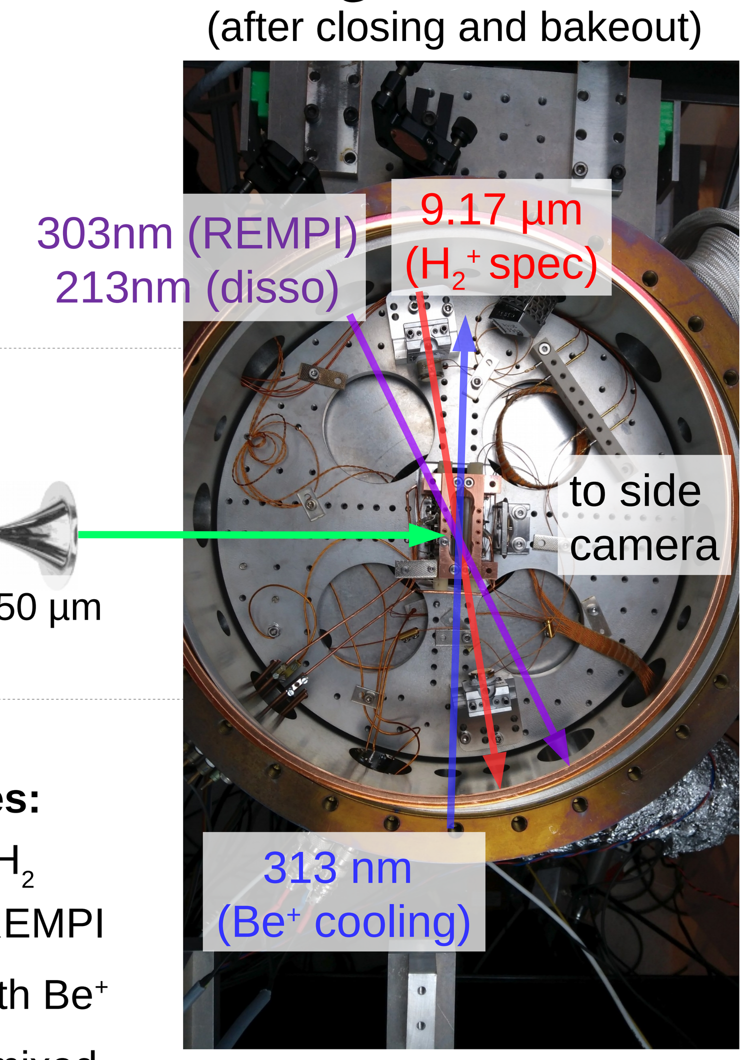
Hyperbolic trap features:

In situ ionisation of H_2 by electron impact or REMPI

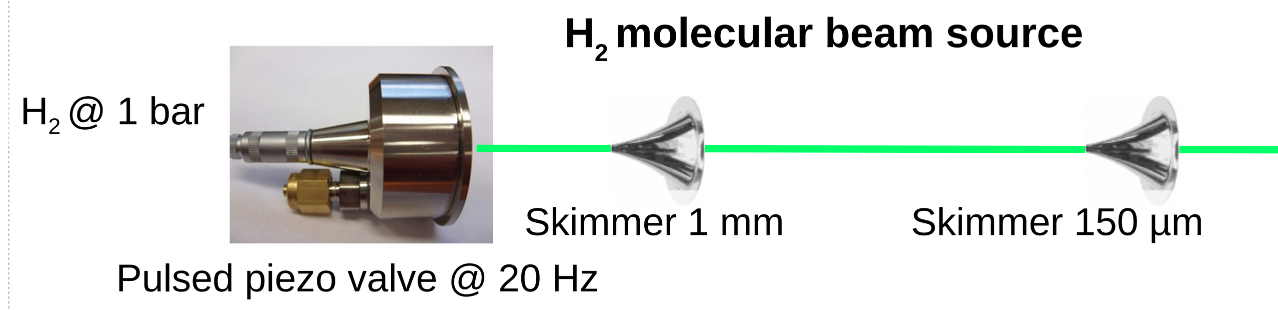
Detection: high voltage extraction to MCP

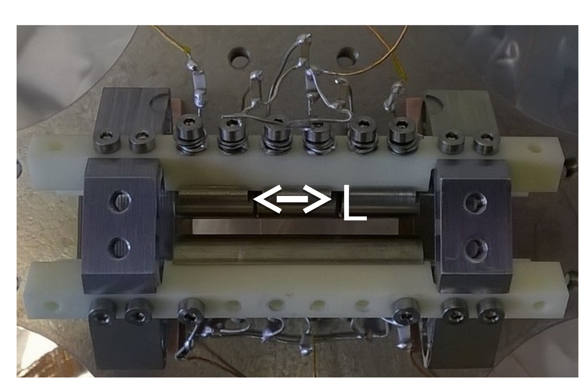
Trap properties:
R_0 = 4.2 mm, Z_0 = 3 mm
$\Omega = 2\pi$ x 14 MHz, U_{rf} = 150 V

UHV chamber with linear trap @ 10^{-10} mBar
(after closing and bakeout)



H_2 molecular beam source





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^+)

Trap properties:
R_0 = 3.5 mm, L=12 mm,
$\Omega = 2\pi$ x 13MHz, U_{rf} = 500V

Conclusions and outlook

- ✓ Theory: m_p/m_e 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 fractional loss of H_2^+
- ✓ State selected H_2^+ ion creation inside the laser cooled Be+ ion cloud
- ✓ Search for H_2^+ two-photon signal

Funding: nano-K, ANR, Johannes Gutenberg Universität Mainz, IUF, FIRST TF, SIRTEQ, VU Vrije Universiteit Amsterdam, GBAR
BESCOOL ANR13-ISO4-0002