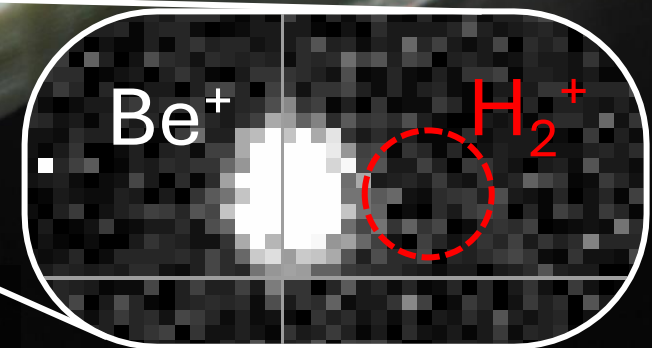


Quantum logic spectroscopy of the hydrogen molecular ion

F. Schmid, D. Holzapfel, Q. He, H. J. Kim, F. Vouzinas, N. Zischka,
N. Schwegler, M. Stadler, A. Ferk, J. P. Home, D. Kienzler

Trapped Ion Quantum Information Group



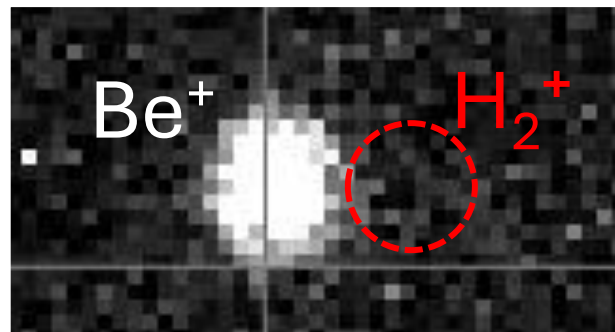
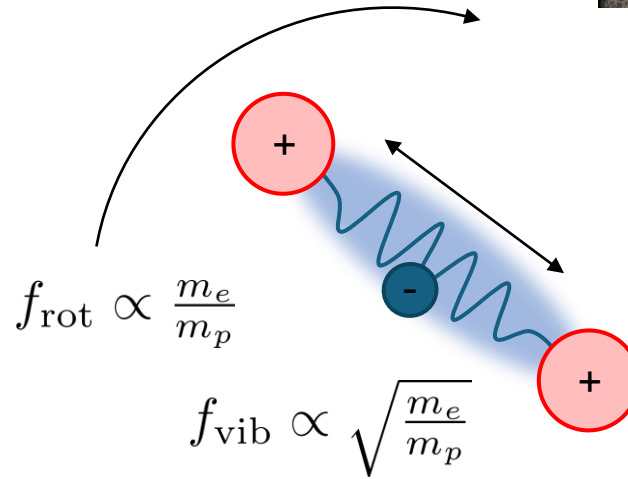
Hydrogen molecular ions

H_2^+ , HD^+ , D_2^+ , HT^+ , DT^+ , T_2^+

Ab-initio theory accurate to few-ppt (10^{-12}) level

V. I. Korobov *et al.*,
PRL **118**, 233001 (2017)

- Precise measurements of nucleon-electron mass ratios, p,d,t charge radii, Rydberg constant
- Test QED, probe physics beyond the standard model



Ensembles - Paul traps

Düsseldorf, Amsterdam,
Paris, Wuhan

Destructive state detection

Single-ion - Penning trap

MPIK Heidelberg +
Düsseldorf

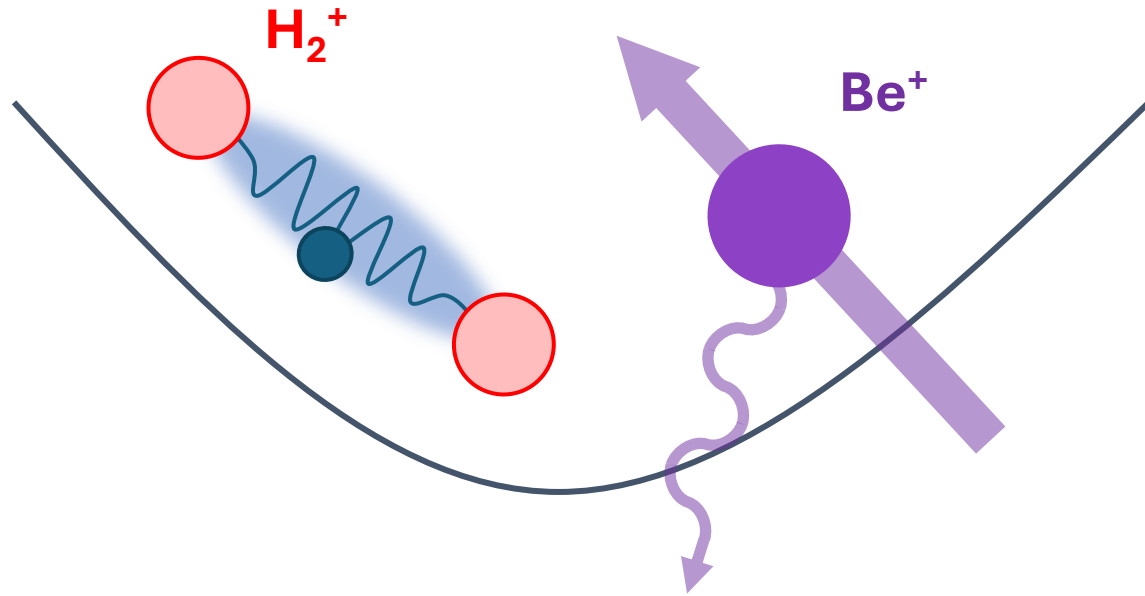
Spin orientation detection

Single-ion - Paul trap

ETH Zürich



QLS - Quantum logic spectroscopy



Single (Paul-)trapped-ion control

- Laser cooling
- State preparation (optical pumping)
- Non-destructive readout

H_2^+
cannot
scatter
photons

QLS - Quantum logic spectroscopy

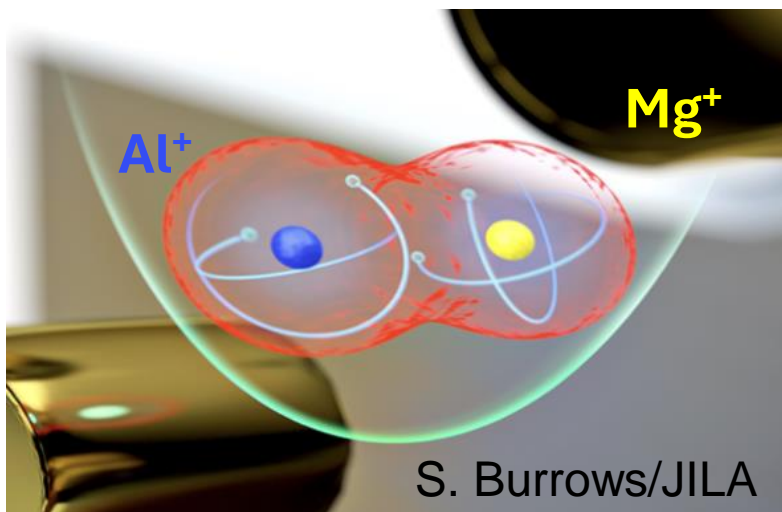
Al^+ : P. O. Schmidt *et al.*, Science **309**, 749 (2005)
 Al^+ : M. C. Marshall *et al.*, PRL **135**, 033201 (2025)
 Al^+ clock syst. uncertainty: 5.5×10^{-19}
Highly charged ions (PTB, MPIK)

QLS of Molecules

MgH^+ : F. Wolf *et al.*, Nature **530**, 457 (2016)
 CaH^+ : C.-W. Chou *et al.*, Nature **545**, 203 (2017)
 N_2^+ : M. Sinhal *et al.*, Science **367**, 6483 (2020)
 H_2^+ : D. Holzapfel *et al.*, PRX **15**, 031009 (2025)

H_2^+ projected uncertainty with QLS: 10^{-17}

S. Schiller *et al.*, PRL **113**, 023004 (2014)
J.-P. Karr *et al.*, J. Phys. Conf. Ser. **723**, 012048 (2016)



S. Burrows/JILA

Apparatus

Tabletop experiment (\sim few m^3)

Linear Paul trap

- particle motion: **harmonic oscillator** with frequency $O(1 \text{ MHz})$

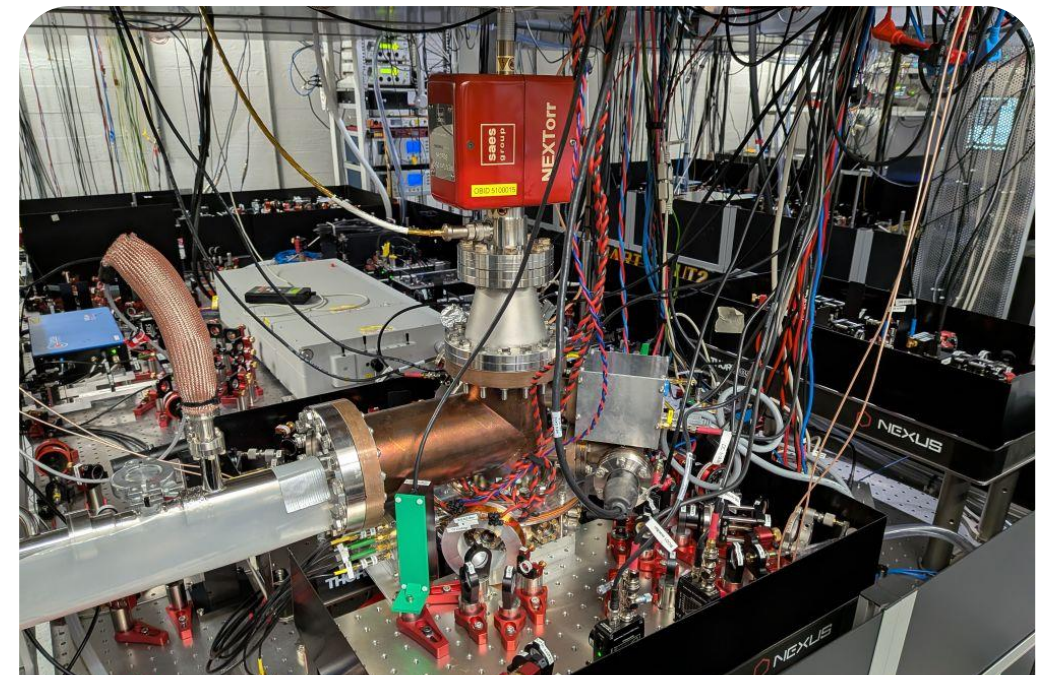
Loading procedure

- Trap single Be^+ (photo-ionization)
- Electron impact ionization or multiphoton ionization of background H_2

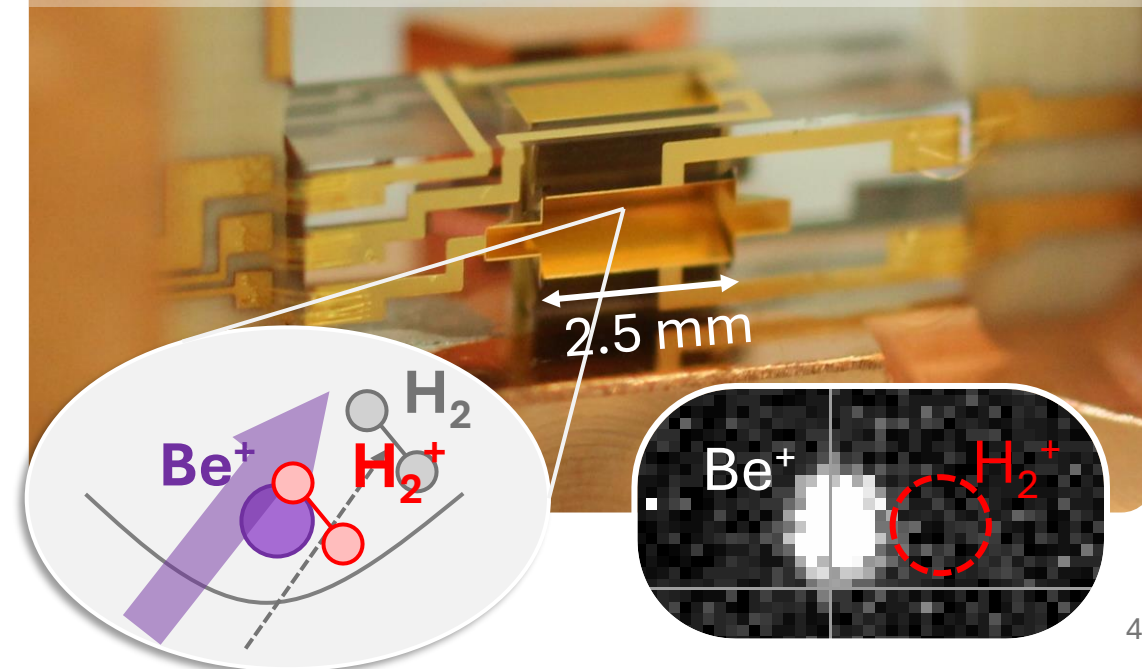
Cryostat + UHV:

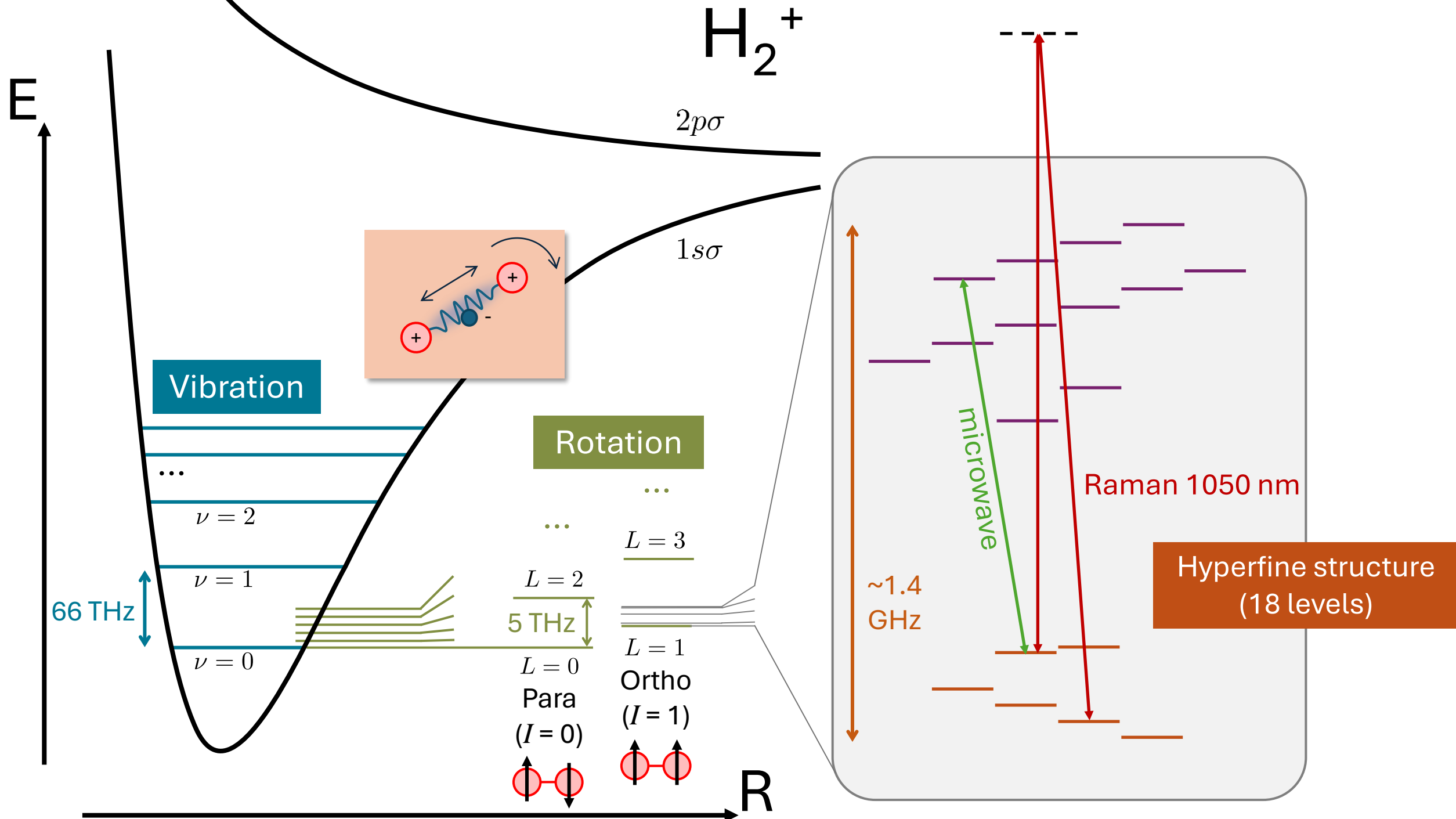
- Prevent H_2^+ reaction to H_3^+
- H_2 partial pressure $\sim 10^{-15}$ mbar

N. Schwegler *et al.*, PRL **131**, 133003 (2023)

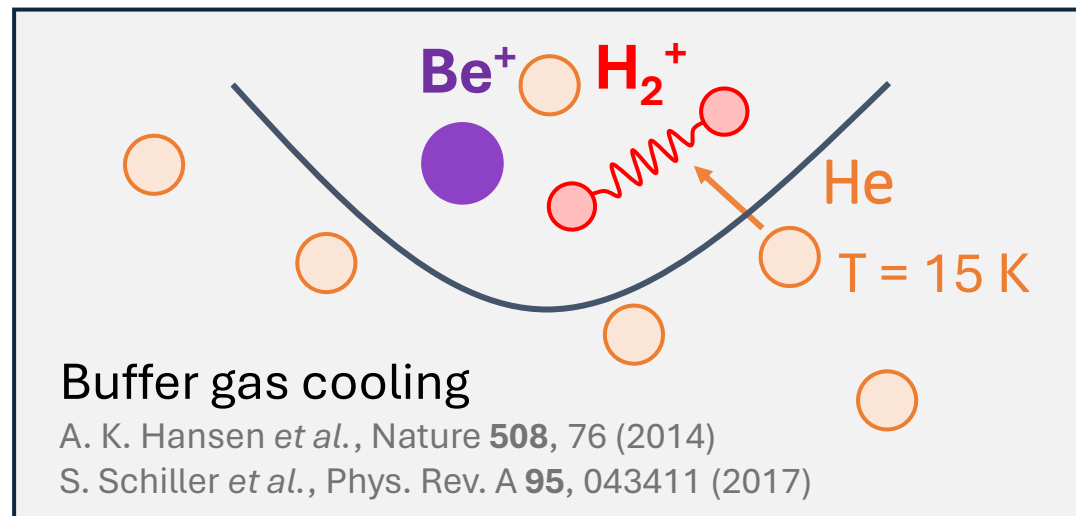
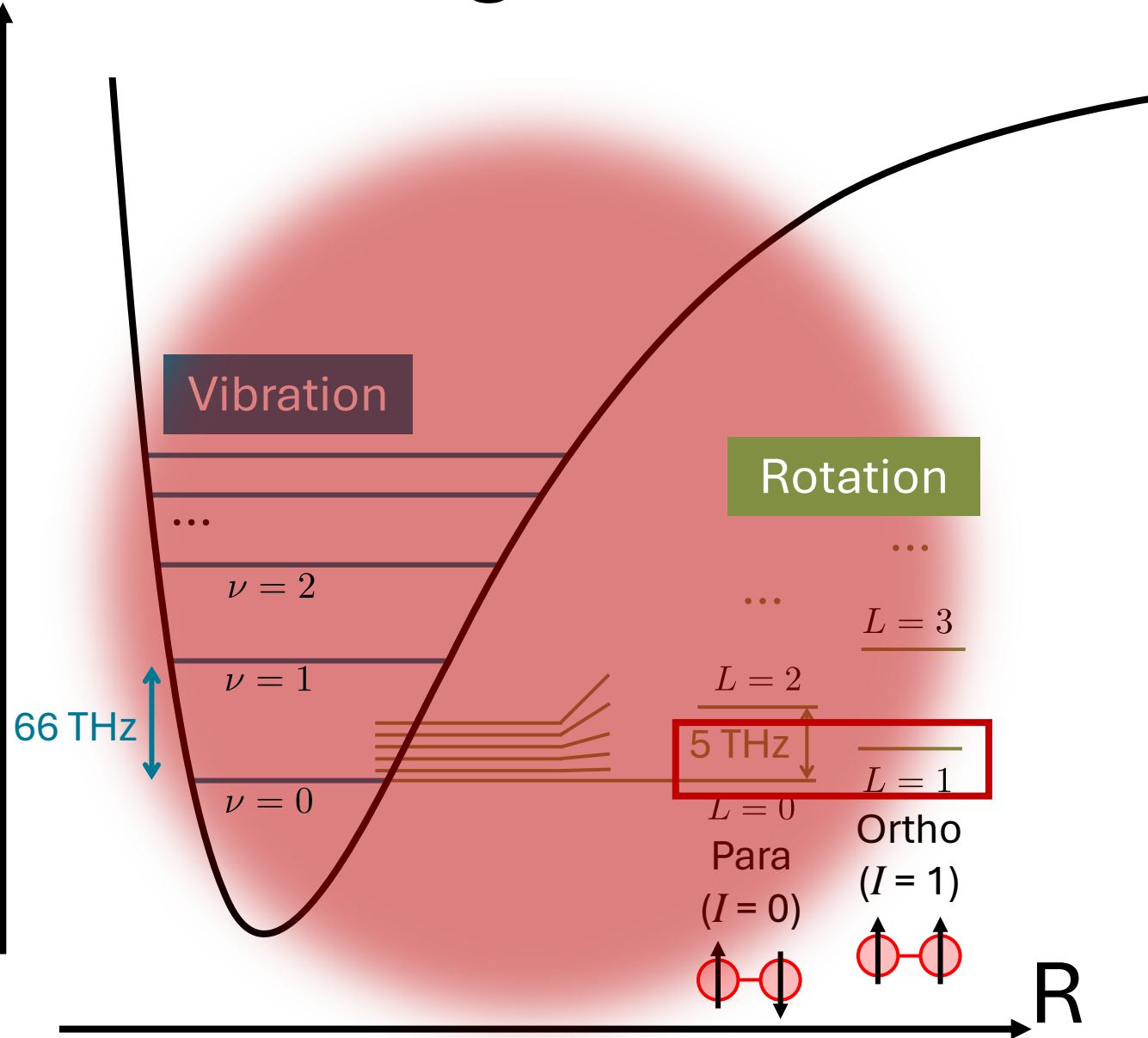


High-precision micro-fabricated ion trap





Electron gun ionization + buffer gas cooling

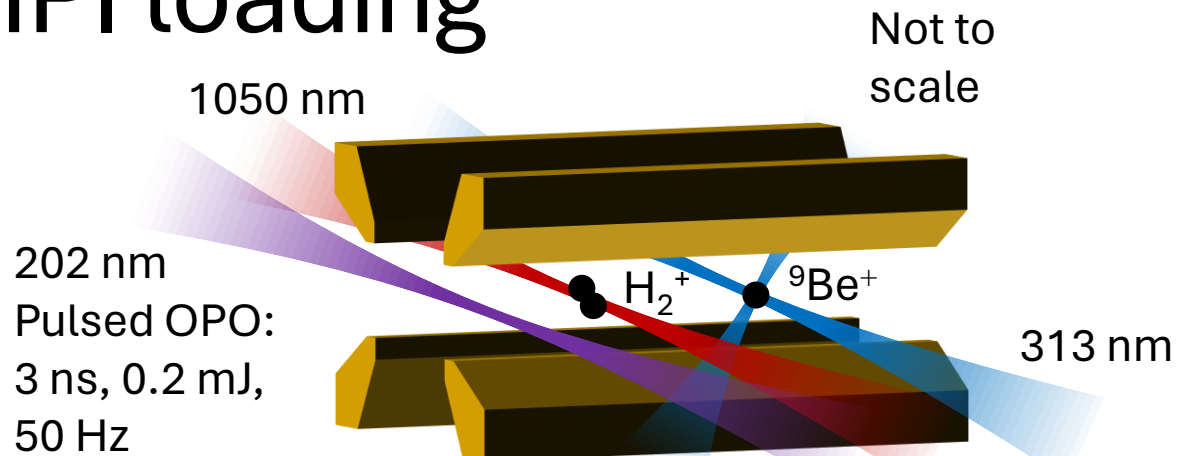
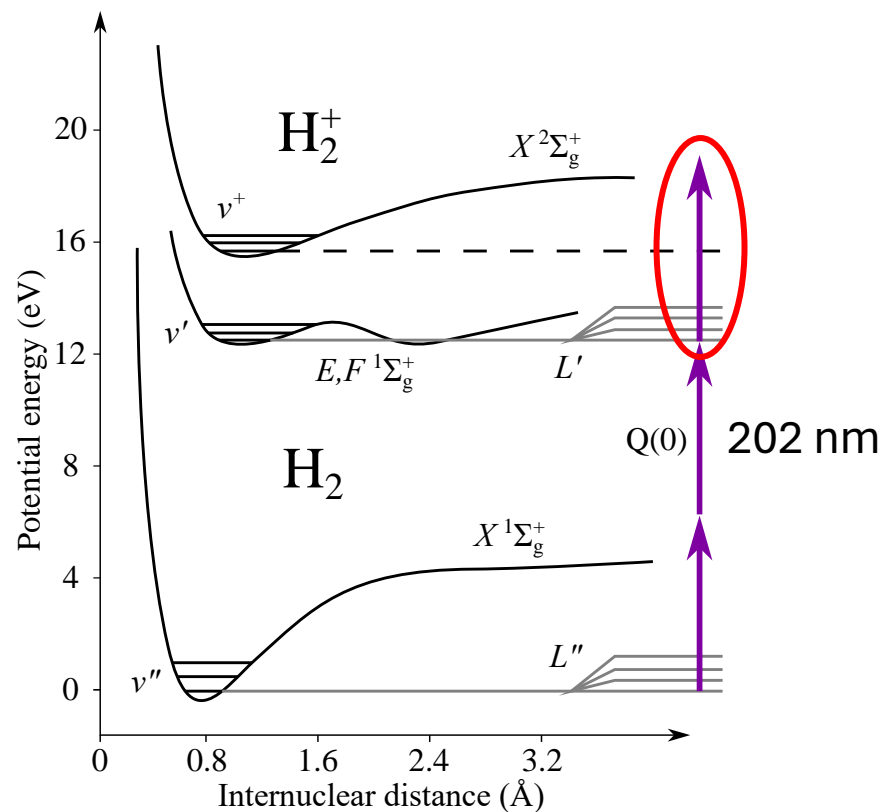


General tool, cheap.

Slow (~10 min)

Not ortho-para selective

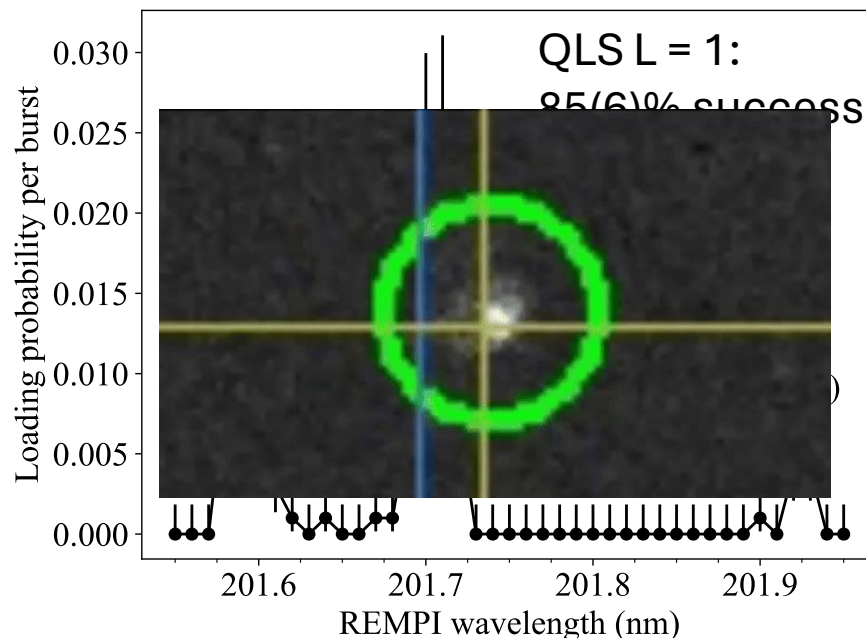
State selective REMPI loading



“Cold” H_2 ($L'' = 0$): raise cryostat temperature
 “Warm” H_2 ($L'' > 0$): leak valve

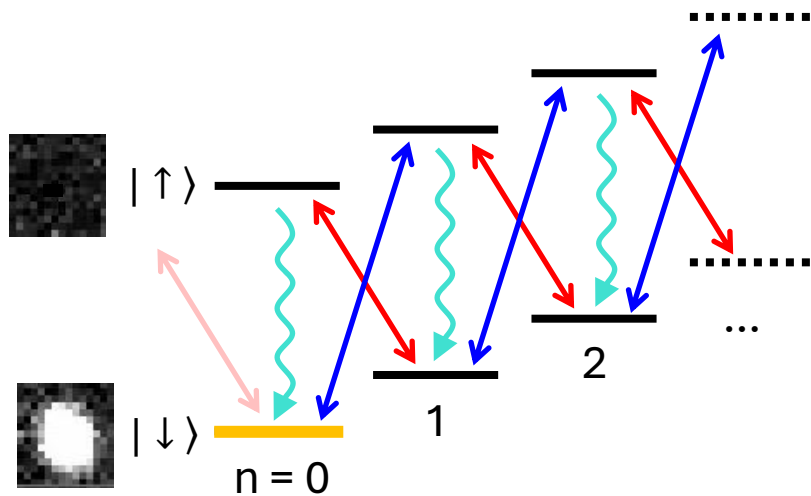
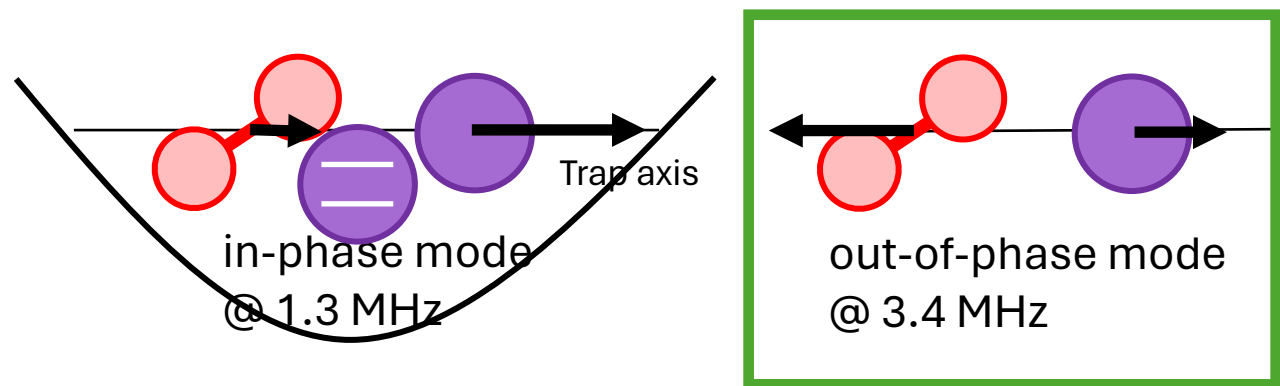
Loading protocol

1. Burst of 50 REMPI laser pulses
2. Check Be^+ ion position
3. Repeat



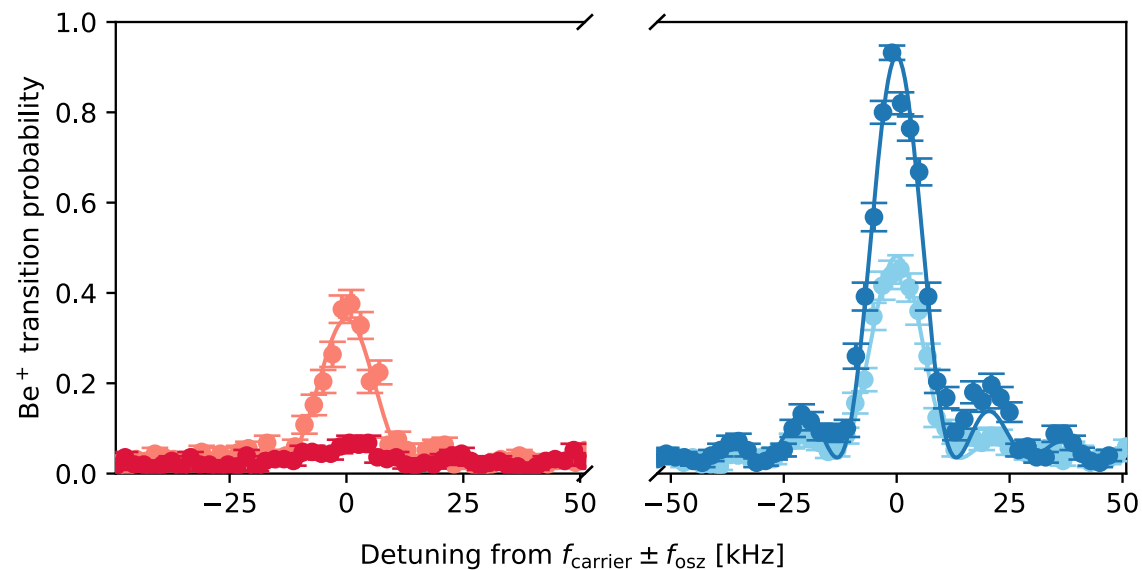
QLS $L = 2$:
 Lifetime > 1 day
 (no collisional quenching)

Axial motion: ground state cooling

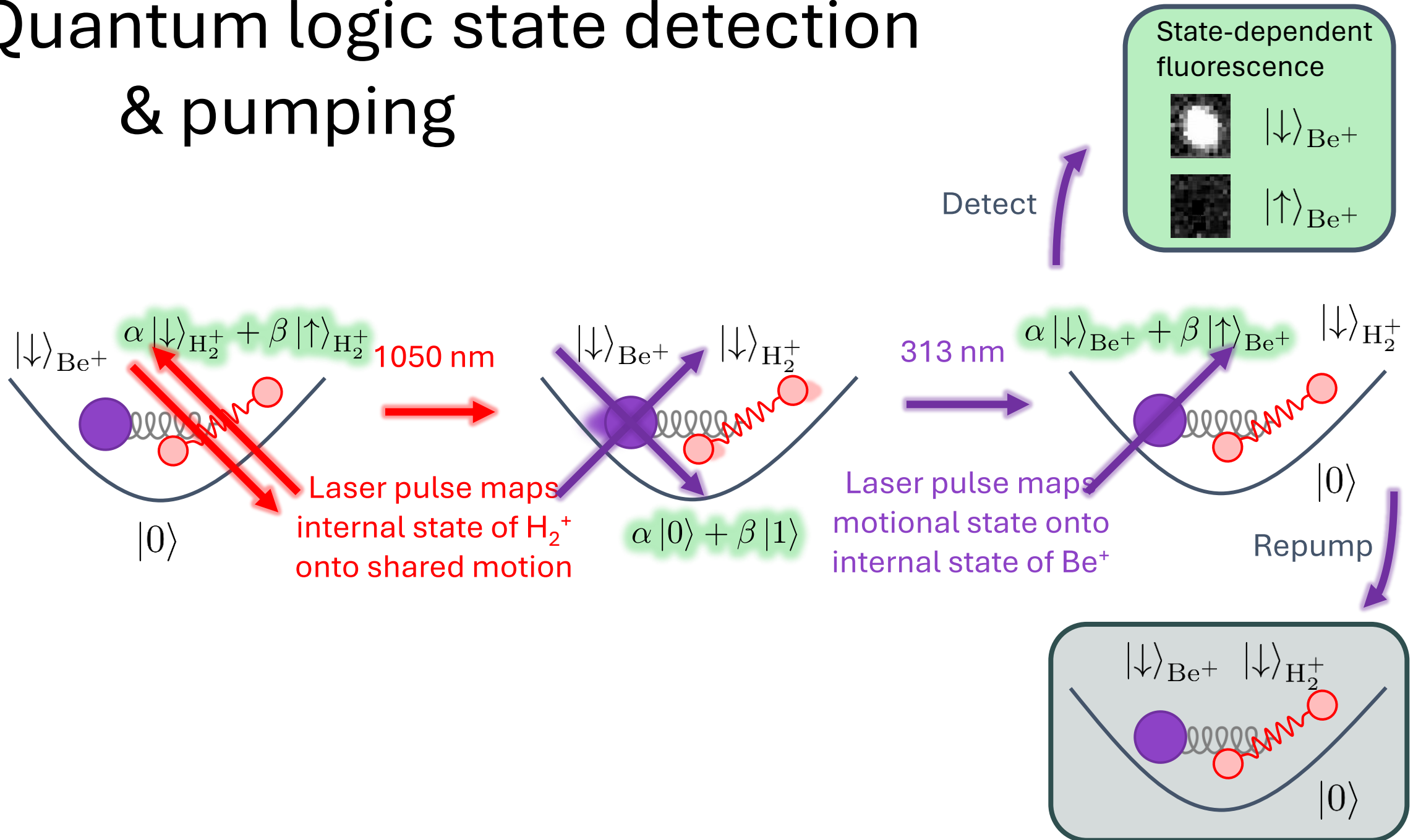


Be⁺ state-dependent fluorescence

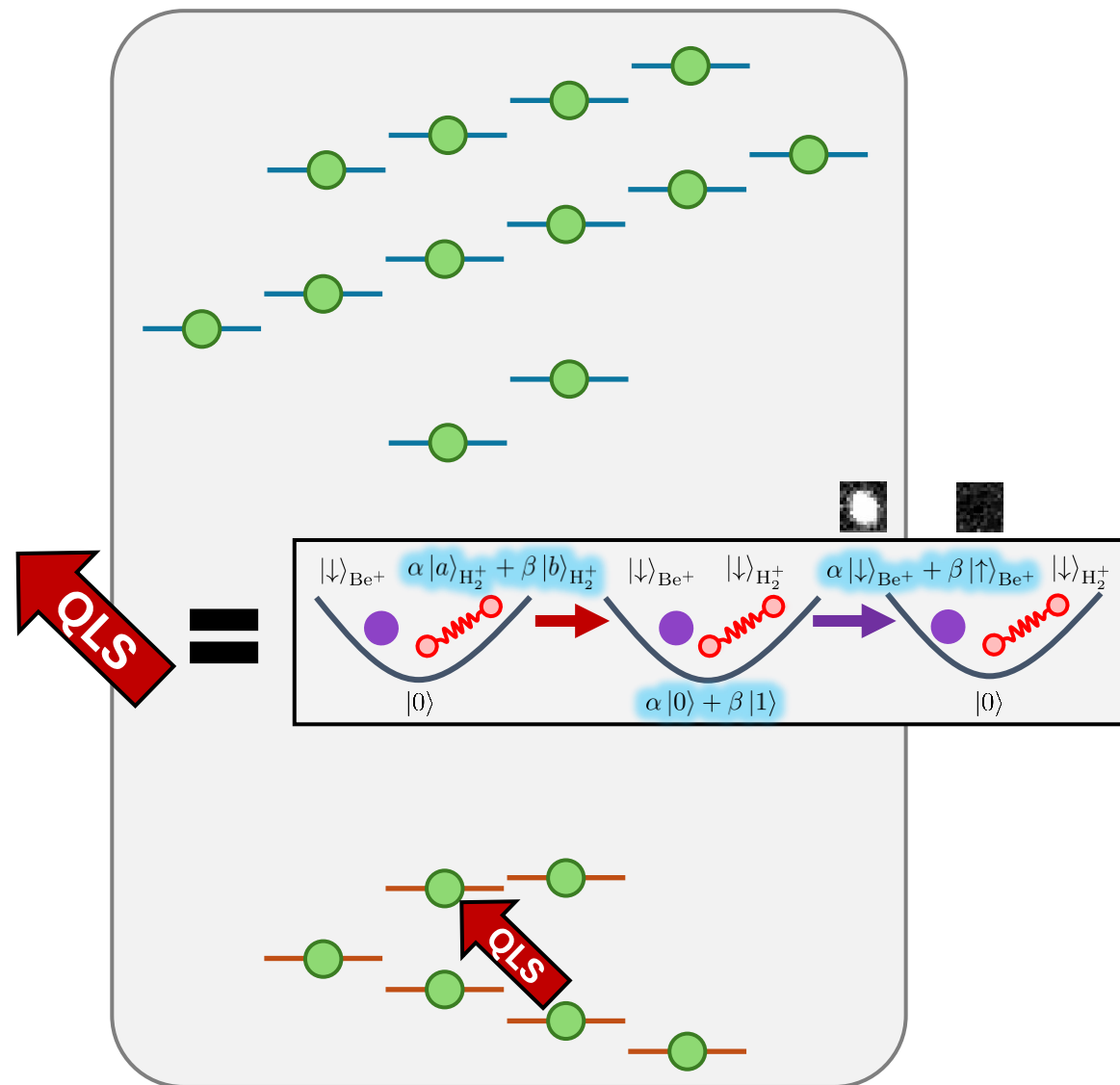
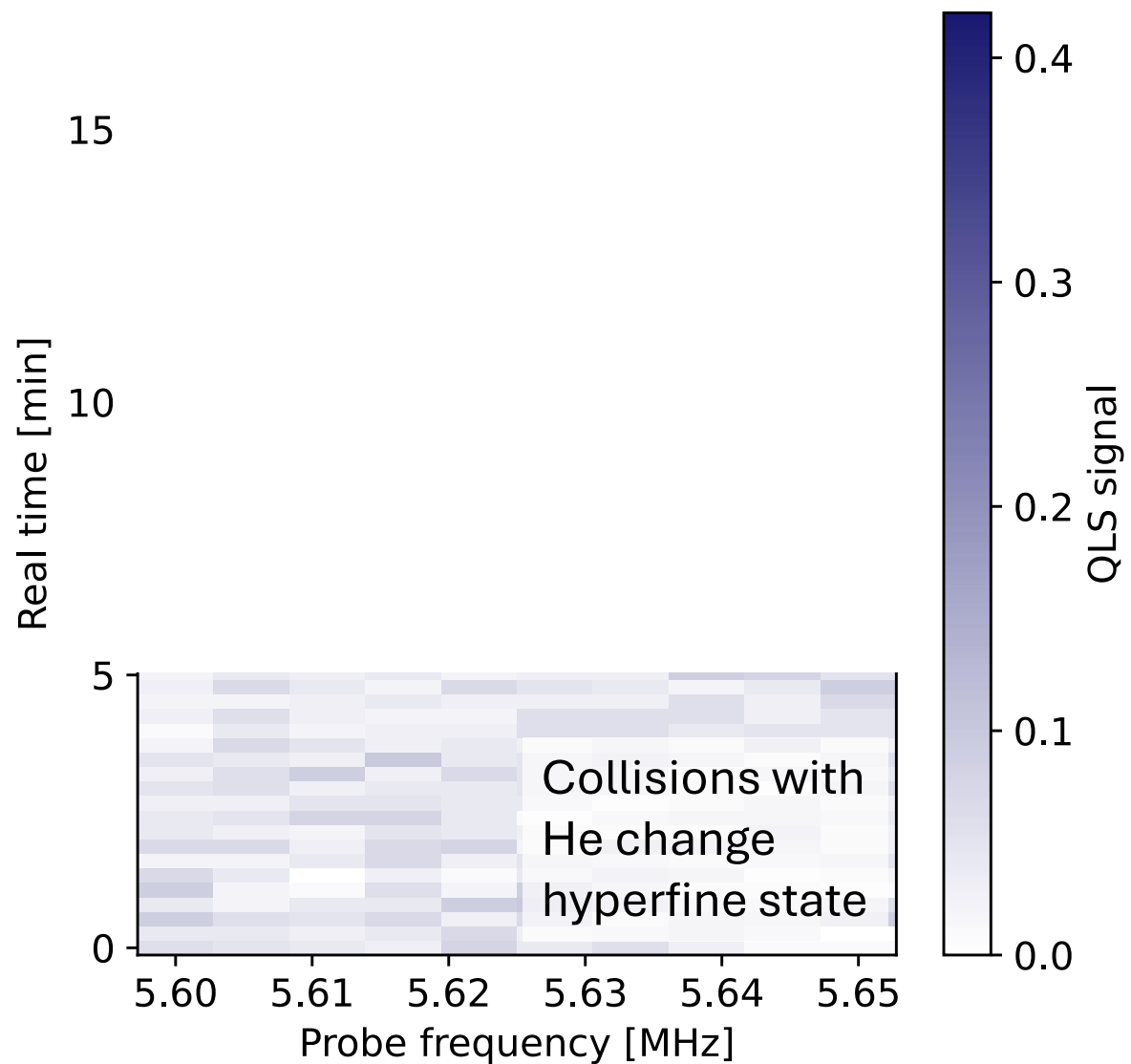
Red sideband: $|\downarrow, n\rangle \leftrightarrow |\uparrow, n-1\rangle$
 Blue sideband: $|\downarrow, n\rangle \leftrightarrow |\uparrow, n+1\rangle$



Quantum logic state detection & pumping

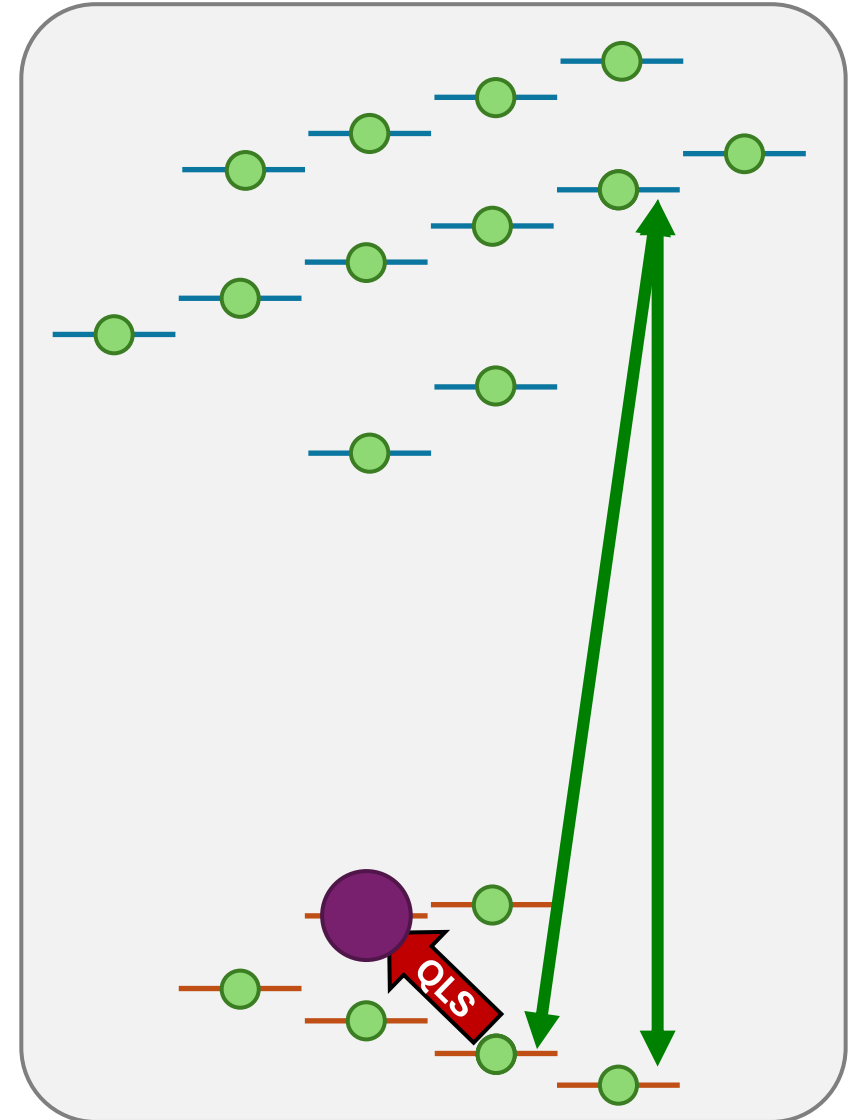


QLS detection



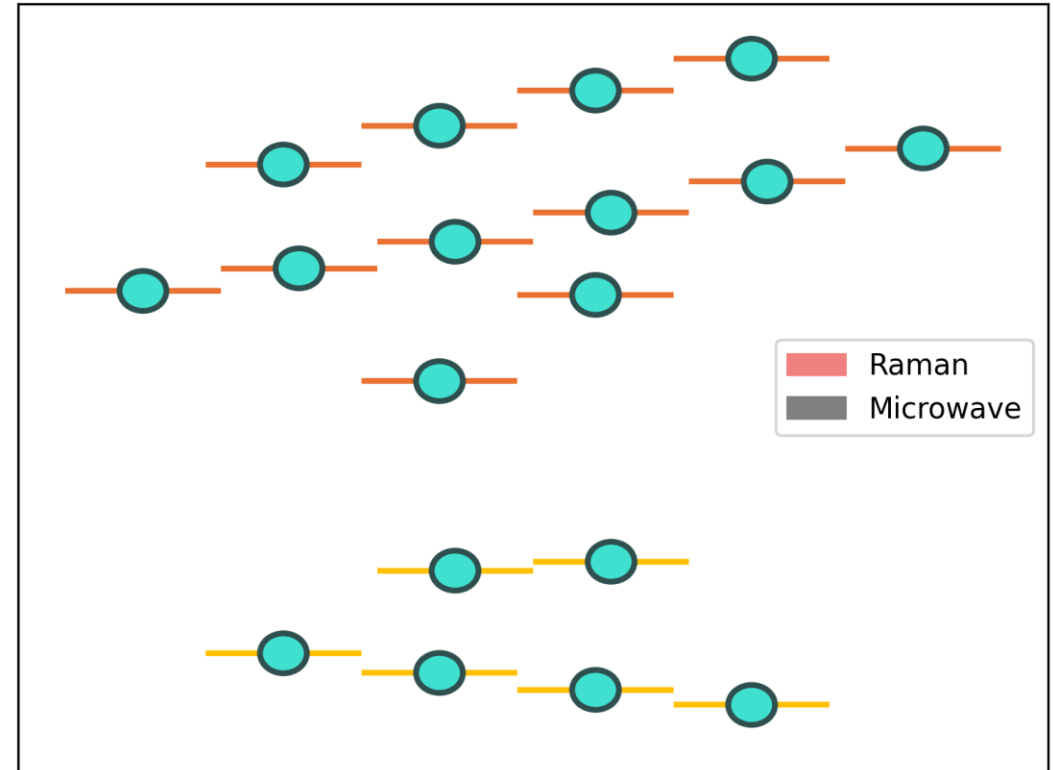
H_2^+ state preparation

- **Quantum logic pumping** with Raman to combine populations
- **Microwave** transitions to connect to all states
- Continue until state is prepared



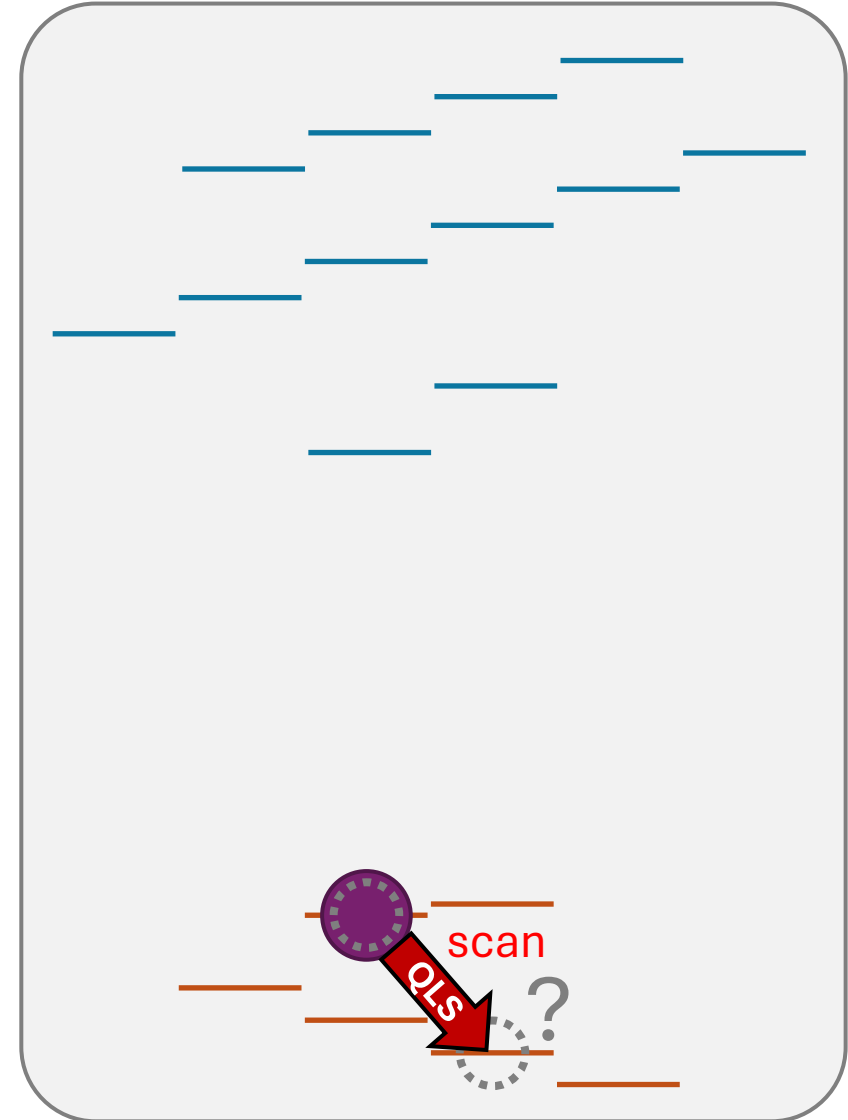
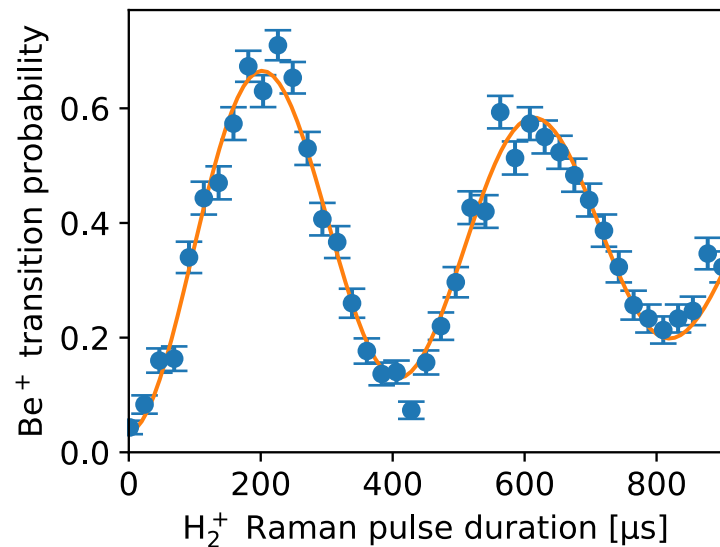
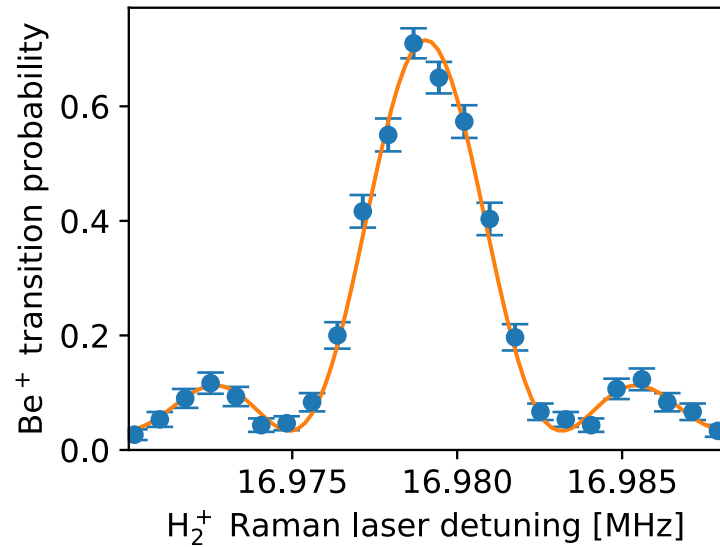
H₂⁺ state preparation

- **Quantum logic pumping** with Raman to combine populations
- **Microwave** transitions to connect to all states
- Continue until state is prepared

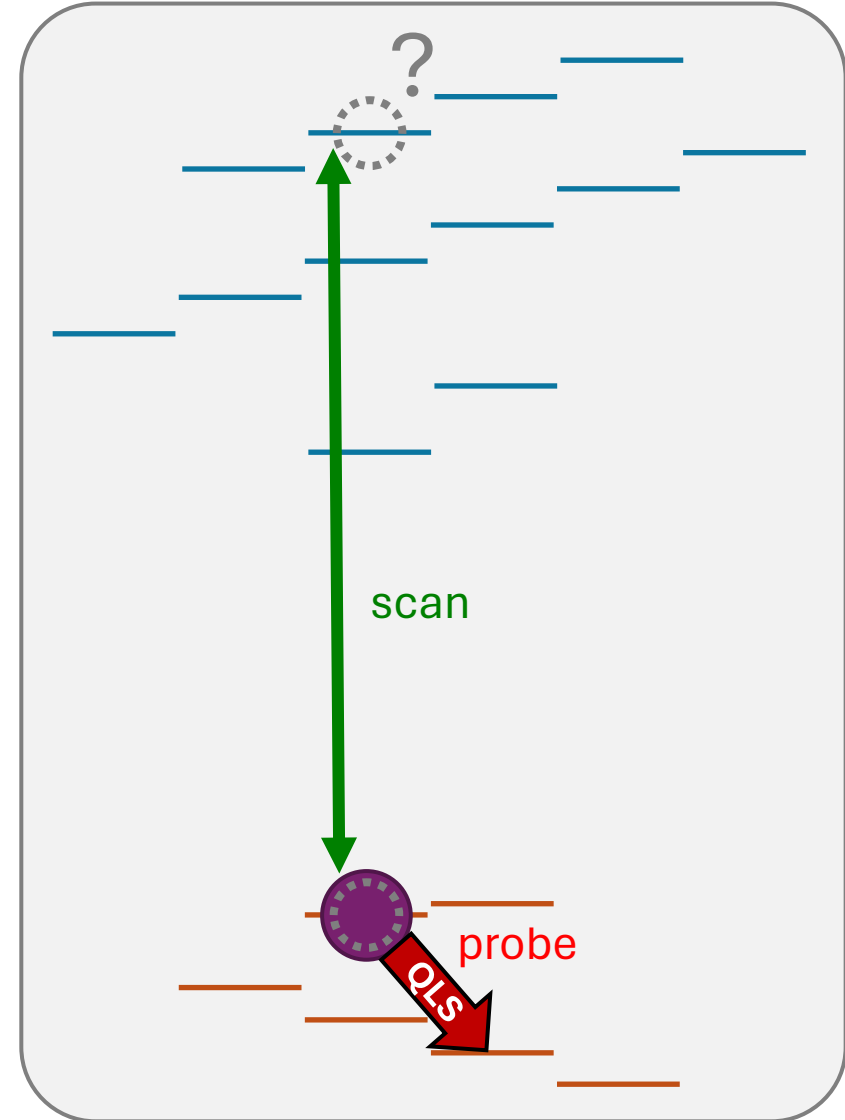
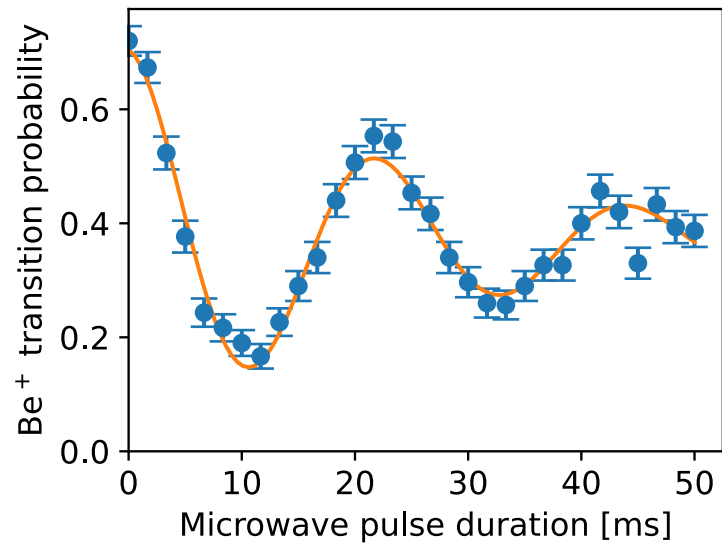
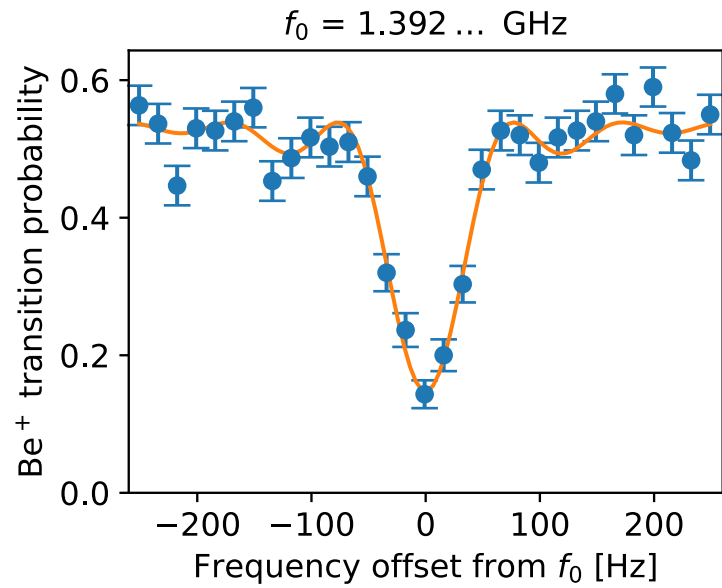


Quantum logic spectroscopy – Raman sideband

No post processing!

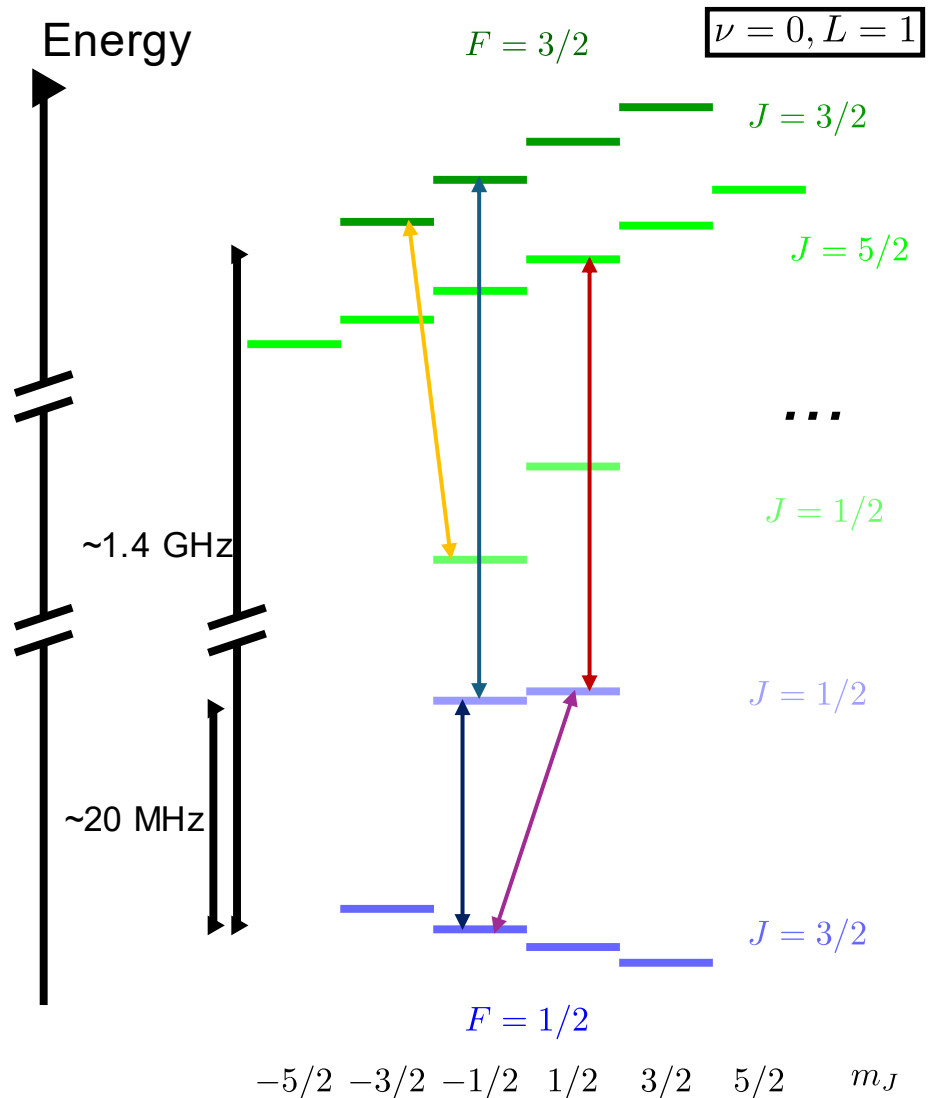


Quantum logic spectroscopy - microwave

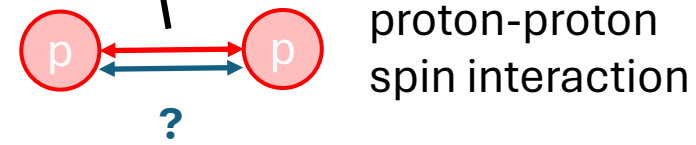


Hyperfine spectroscopy

Collaboration with J.-Ph. Karr (LKB, Paris)



$$H_{\text{eff}} = b_F(\mathbf{I} \cdot \mathbf{s}_e) + c_e(\mathbf{L} \cdot \mathbf{s}_e) + c_I(\mathbf{L} \cdot \mathbf{I}) + \frac{d_1}{(2L-1)(2L+3)} \times \left\{ \frac{2}{3} \mathbf{L}^2 (\mathbf{I} \cdot \mathbf{s}_e) - [(\mathbf{L} \cdot \mathbf{I})(\mathbf{L} \cdot \mathbf{s}_e) + (\mathbf{L} \cdot \mathbf{s}_e)(\mathbf{L} \cdot \mathbf{I})] \right\} + \frac{d_2}{(2L-1)(2L+3)} \left[\frac{1}{3} \mathbf{L}^2 \mathbf{I}^2 - \frac{1}{2} (\mathbf{L} \cdot \mathbf{I}) - (\mathbf{L} \cdot \mathbf{I})^2 \right].$$



Test new physics, 5th force:

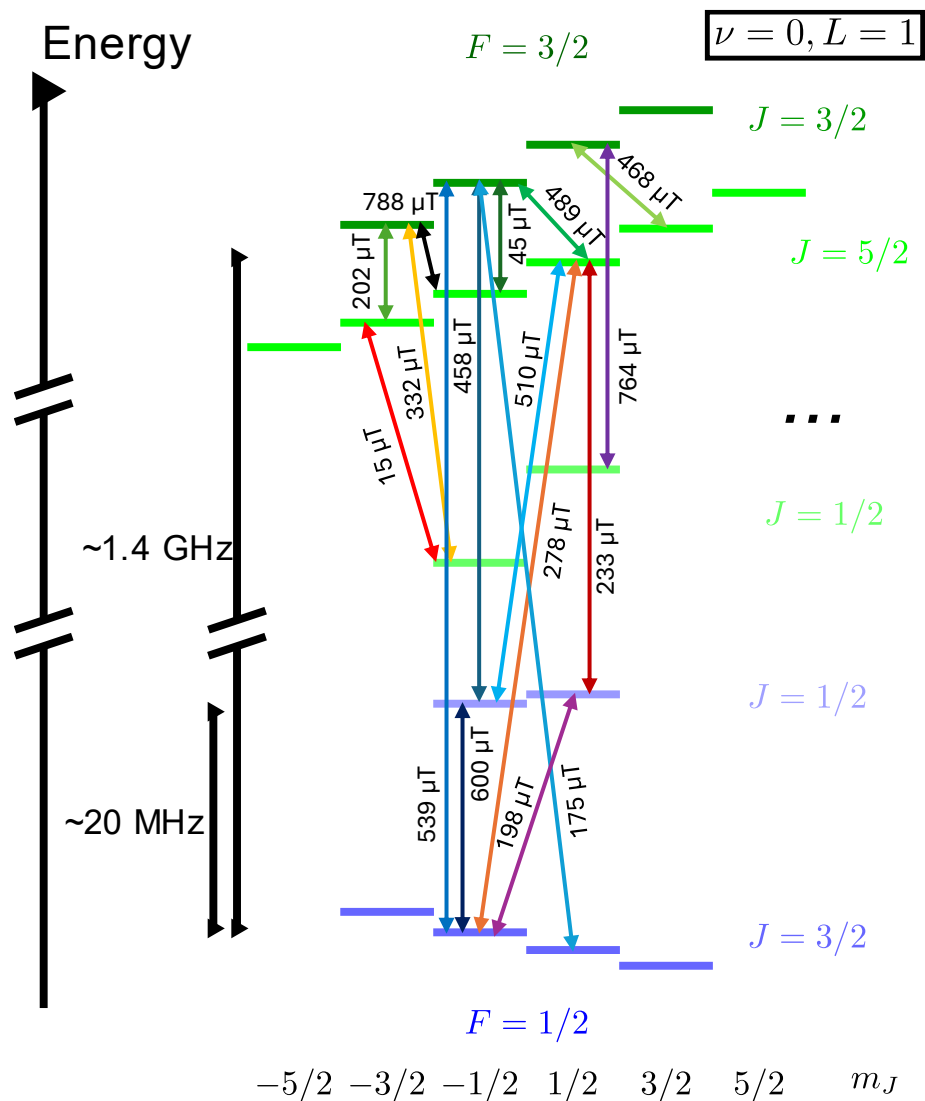
Additional spin-dependent proton-proton interaction

In H_2 : N. F. Ramsey, *Physica* **96A**, 285 (1979)

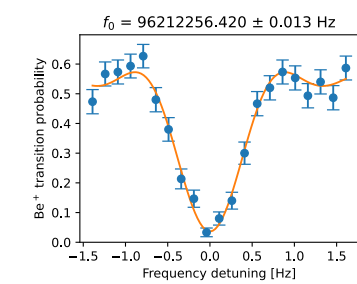
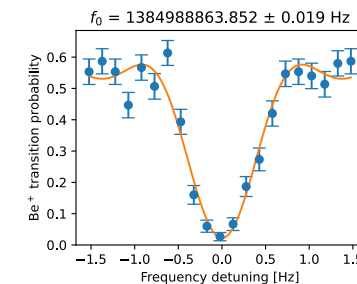
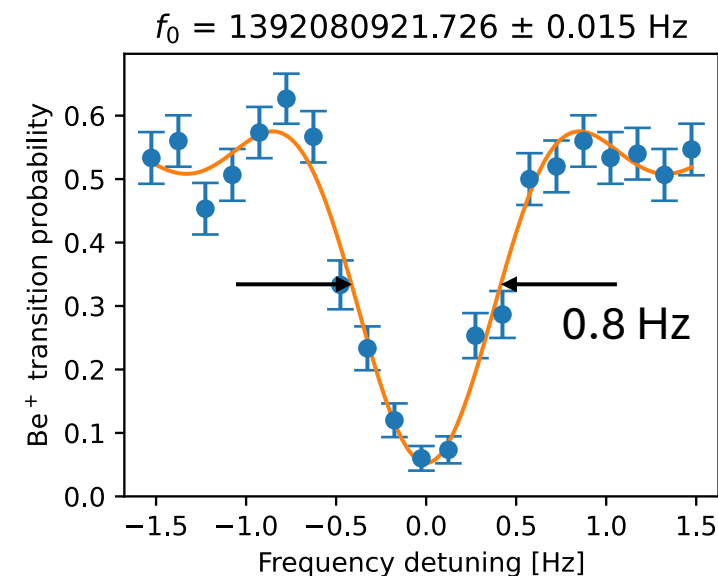
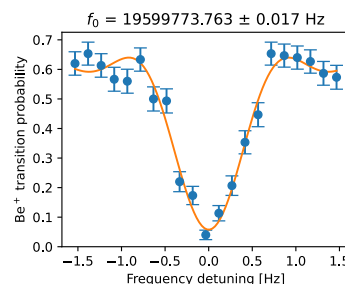
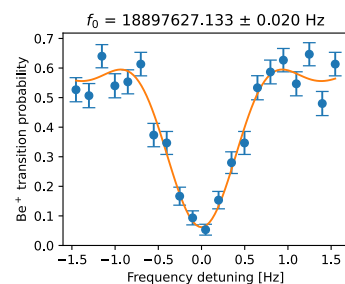
Review: L. Cong *et al.*, *RMP* **97**, 025005 (2025)

Hyperfine spectroscopy

Collaboration with J.-Ph. Karr (LKB, Paris)

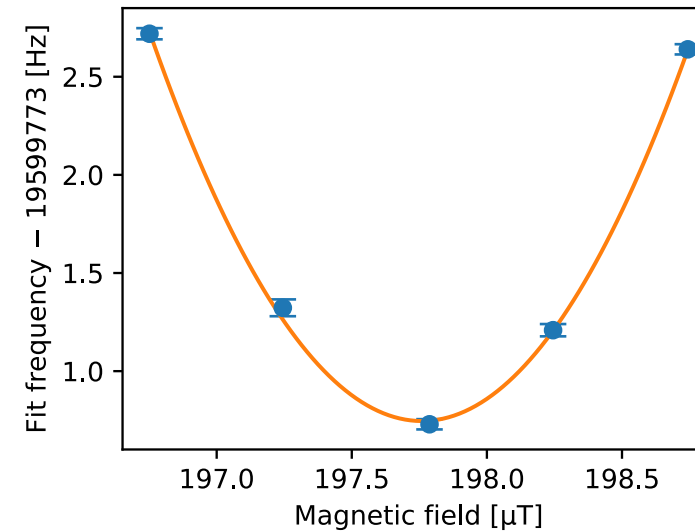
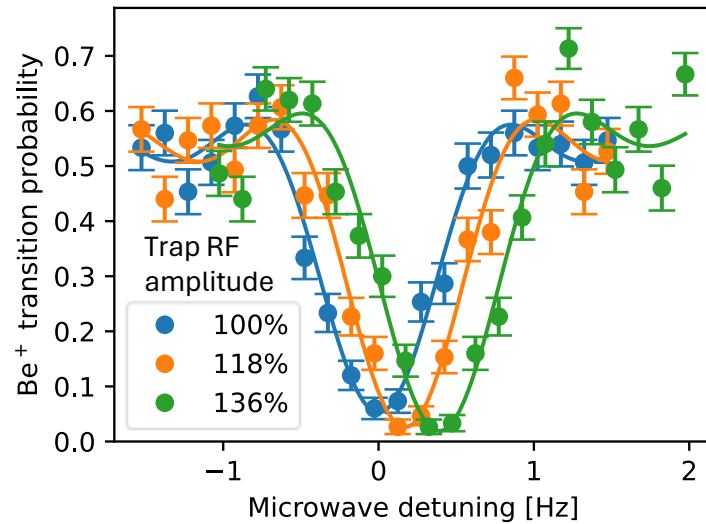


H_2^+ has large Zeeman shifts...
 Use magnetic field insensitive transitions (0 – 1 mT range)
 Be^+ as in-situ magnetic field sensor

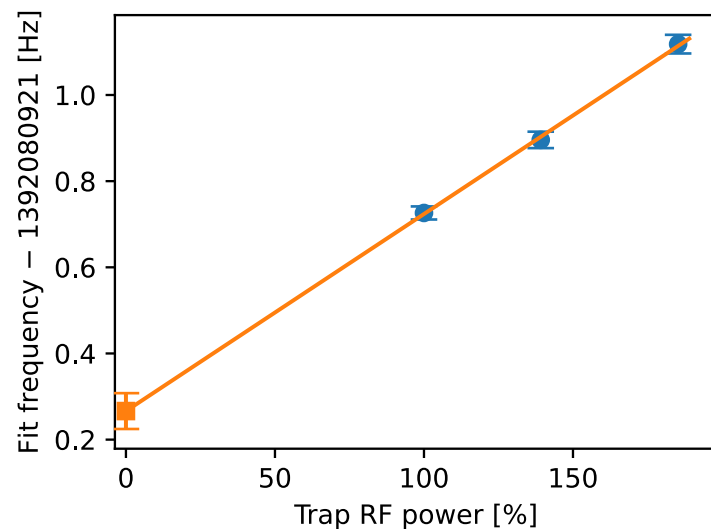


15 mHz precision in 90 min

Hyperfine spectroscopy - systematics



Trap RF-induced AC Zeeman shift

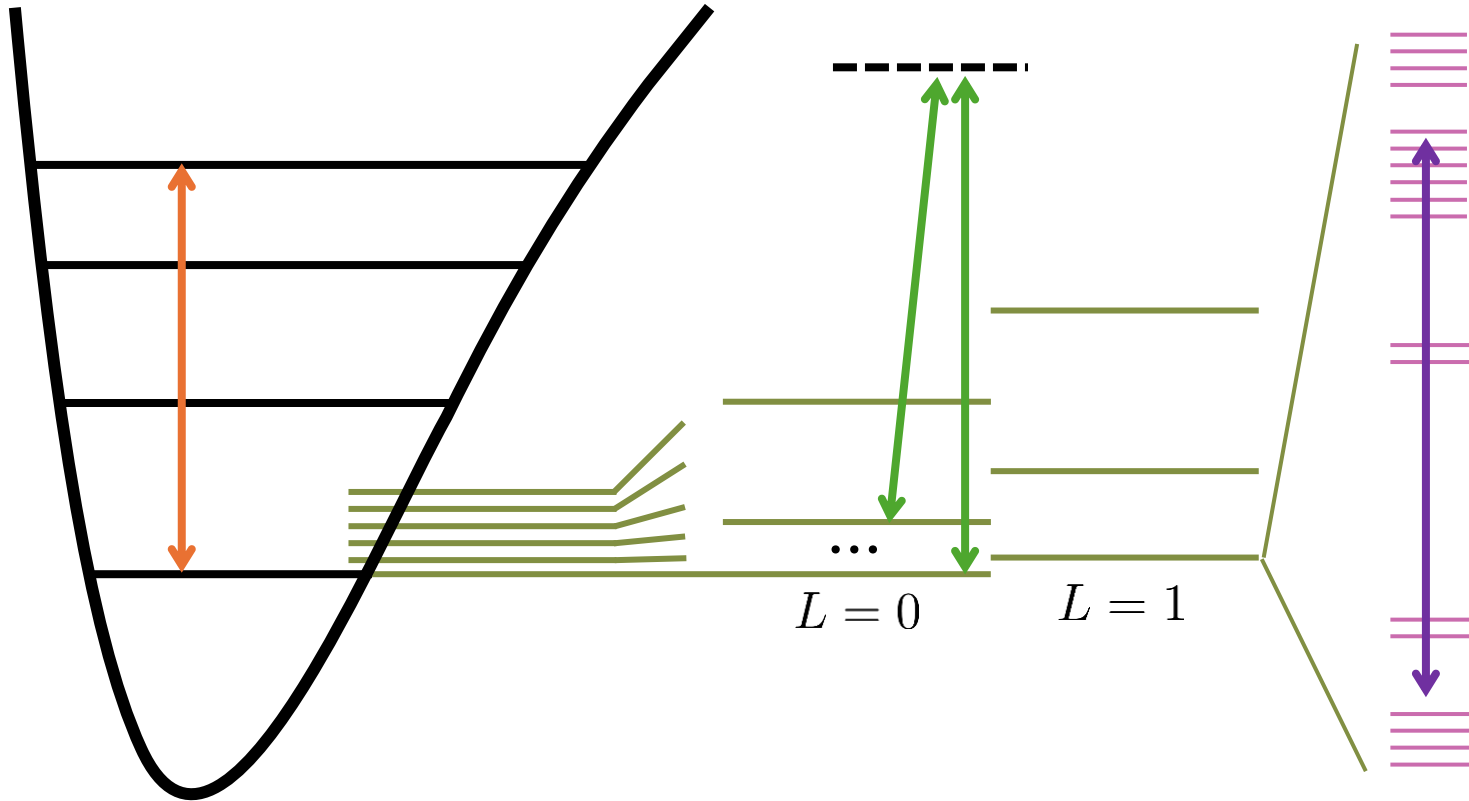


Preliminary uncertainty budget

Ro-vib. transition

	Shift	Uncertainty	Shift	Uncertainty
Trap RF AC Zeeman	(0 - 2) Hz	~50 mHz	≍	≍
Magnetic field fluctuations	0	< 10 mHz	≍	≍
Quadrupole shift	(0-0.3) Hz	?	≍	≍
Micromotion Stark shift	~ mHz	~ mHz	≍	≍
2 nd order Doppler shift	~ μHz	~ μHz	≍	≍
?	Probe laser AC Stark		Up to few Hz	< Hz

Outlook: H_2^+



Microwave hyperfine spectroscopy $L = 1$

Test of theory
Probe New Physics

Rovibrational single-photon transitions

- $\nu, L: 0, 0-3, 2$: 1576 nm quadrupole transition
J.-Ph. Karr et al., J. Phys.: Conf. Ser. 723, 012048 (2016)
- $\nu, L: 0, 0-3, 1$: 1600 nm ortho-para (dipole) transition
V. I. Korobov and D. Bakalov, PRA 107, 022812 (2023)

Rotational Raman transitions

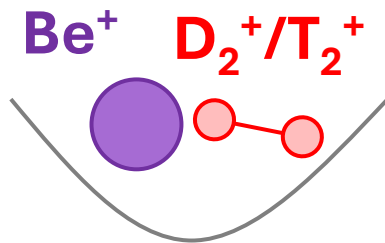
$L: 0-2, 1-3$ (5, 8 THz)

*C.-W. Chou et al., Science 367, 1458 (2020),
A. L. Collopy et al., PRL 130, 223201 (2023),
K. H. Leung et al., PRX 13, 011047 (2023)*

Fundamental constants, probe New Physics

Outlook: Other light molecules

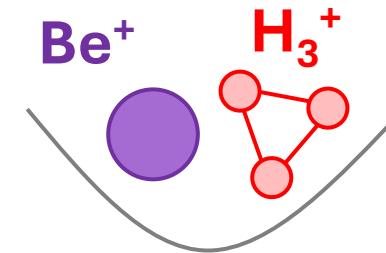
Deuteron quadrupole moment
Order-of-magnitude improvement of triton charge radius, check m_t/m_e
J.-Ph. Karr *et al.*,
PRA **112**, 022809 (2025)



F. Schmid

HeH^+
 He_2^+
...

Benchmark quantum chemistry calculations
Hyperfine structure theory:
G. Avila *et al.*,
PRL **135**, 043003 (2025)



D. Kienzler

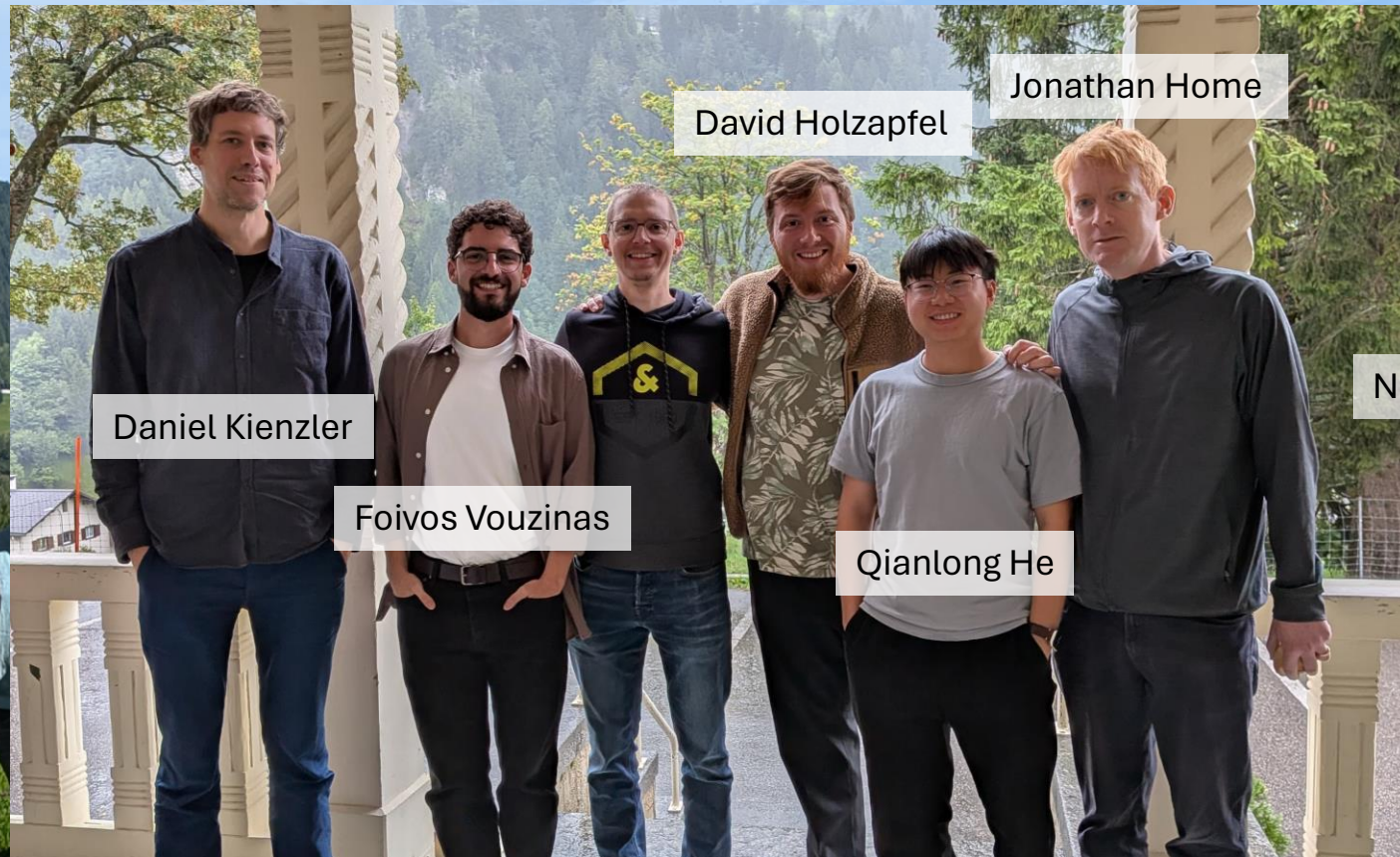


Tritium Laboratory Karlsruhe:
 10^{-2} mbar*l of T_2 (1 GBq) at VU Amsterdam

We load H_2^+ from 10^{-7} mbar*l...

PhD & PostDoc position available

daniel.kienzler@phys.ethz.ch
tiqi.ethz.ch



Daniel Kienzler

Foivos Vouzinas

David Holzapfel

Jonathan Home

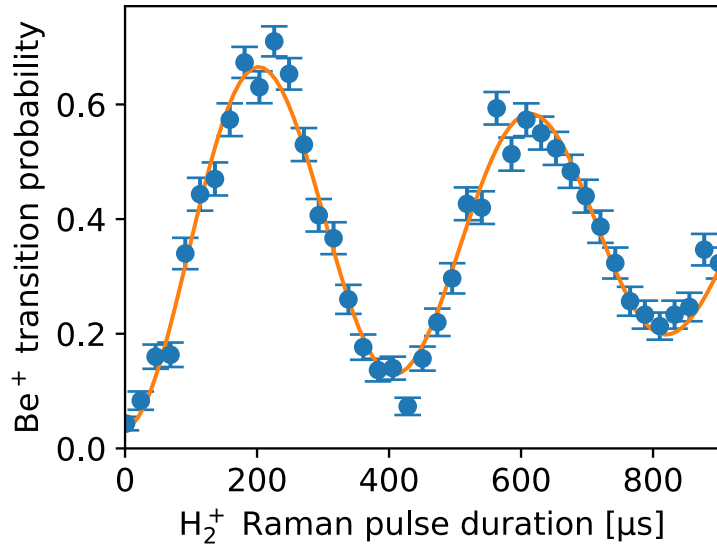
Qianlong He



Niklas Zischka



Thank you for your attention



N. Schwegler *et al.*, PRL 131, 133003 (2023)
D. Holzapfel *et al.*, PRX 15, 031009 (2025)
H. J. Kim *et al.*, PRA 113, 043119 (2026)

