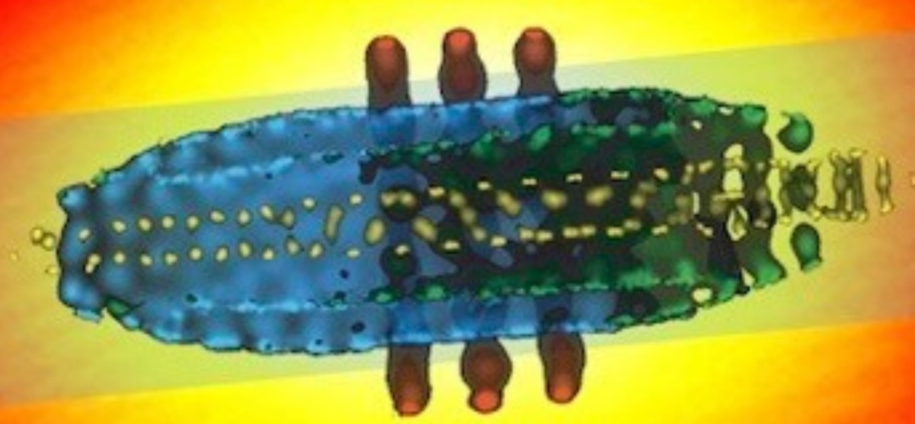
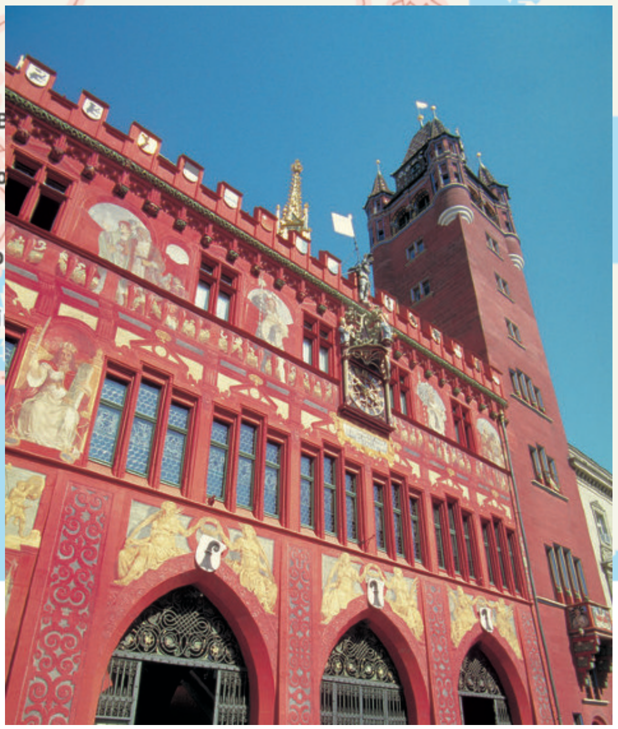
Cold molecular ions in traps: methods and applications
Lecture I



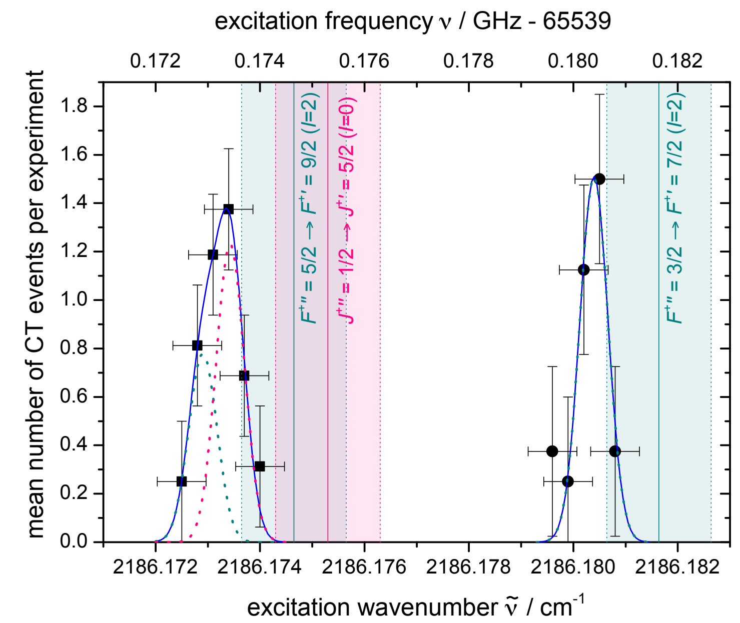
TIFR-ICTS Online School and Discussion Meeting on Trapped Atoms, Molecules and Ions
May 10-22, 2021

Stefan Willitsch
Department of Chemistry
University of Basel, Switzerland

Basel, Switzerland

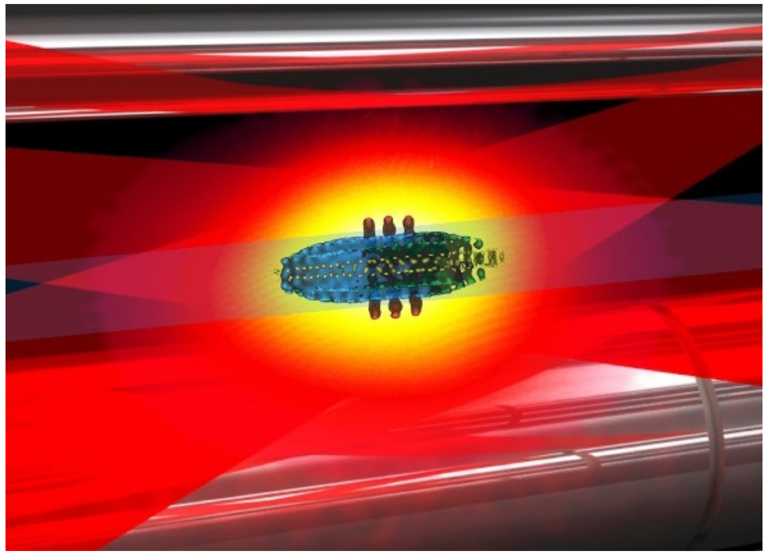


Applications of cold molecular ions in physics and chemistry



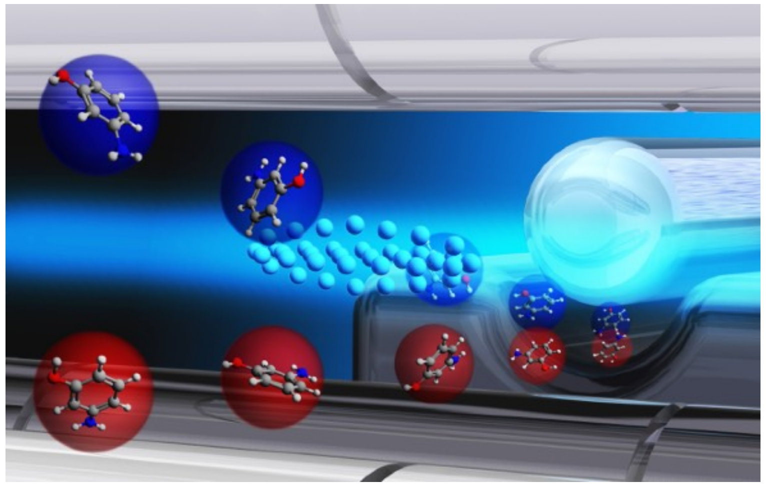
Quantum control over single trapped particles

- Quantum logic and quantum technologies
- Precision spectroscopy



Ion-neutral interactions, collisions and chemical reactions in a new physical regime

- Study “exotic” chemical processes
- Explore quantum character of collisions
- Probe fine details of intermolecular interactions



New methods for studying and controlling chemical reactions

- Accurate quantum-state AND collision-energy control
- Controlled chemistry of large molecules

Contents

Lecture I
Cold molecular ions: Basic techniques and spectroscopy

1. Cooling and trapping of molecular ions: basic techniques
2. Internal-state preparation of cold molecular ions
3. Precision spectroscopy of cold molecular ions
4. Molecular-ion quantum technologies and quantum-logic spectroscopy

Lecture II
Cold ion-neutral interactions

5. Ion-neutral interactions: theory
6. Ion-atom hybrid systems
7. Molecular ions in hybrid traps

Recommended books on cold molecules and cold ions

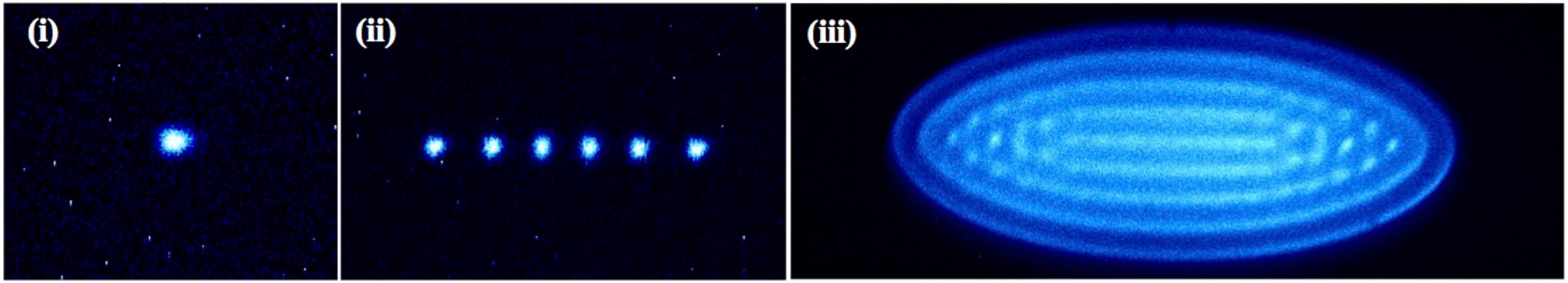
- R.V. Krems et al. (ed.), Cold Molecules, CRC Press 2009
- R.V. Krems, Molecules in Electromagnetic Fields, Wiley, 2019
- I.W.M Smith (ed)., Low Temperatures and Cold Molecules, Imperial College Press, 2008
- A. Osterwalder, O. Dulieu (ed.), Cold Chemistry, RSC Publishing, 2018
- F. G. Major et al., Charged Particle Traps Vol. I+II, Springer, 2005 + 2007
- M. Knoop et al. (ed.), Ion Traps For Tomorrow's Applications, Proc. Int. School Phys. E. Fermi Vol. 189, IOS Press, 2015
- M. Knoop et al. (ed.), Trapped Charged Particles, World Scientific, 2016

... plus many reviews and research articles referenced later in this lecture

1. Cooling and trapping of molecular ions: basic techniques

Cold ions in traps: ion Coulomb crystals

See also other lectures on ion trapping in this school



Fluorescence images of laser-cooled Ca^+ ions in an ion trap

Properties of Coulomb-crystallized ions:

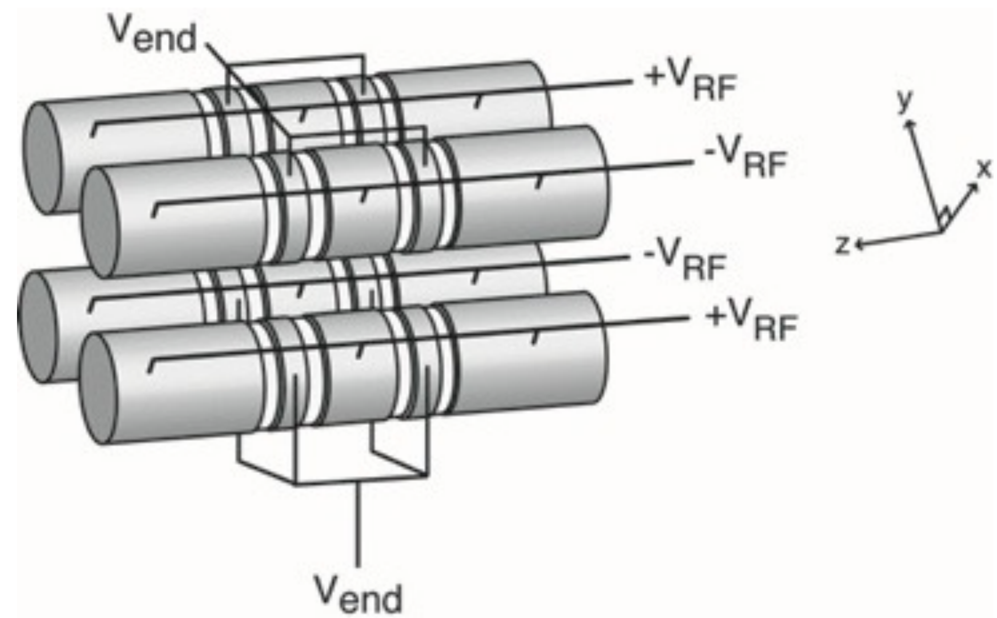
- Cold (μK-mK)
- Long trapping times (> hrs)
- Ordered structures of single, localized particles:
  observe, address and manipulate single ions on the quantum level

Lit.:
- D. Leibfried et al., Rev. Mod. Phys. 75 (2003), 281
- H. Häffner et al., Phys. Rep. 469 (2008), 155
- S. Willitsch, Int. Rev. Phys. Chem. 30 (2012), 175

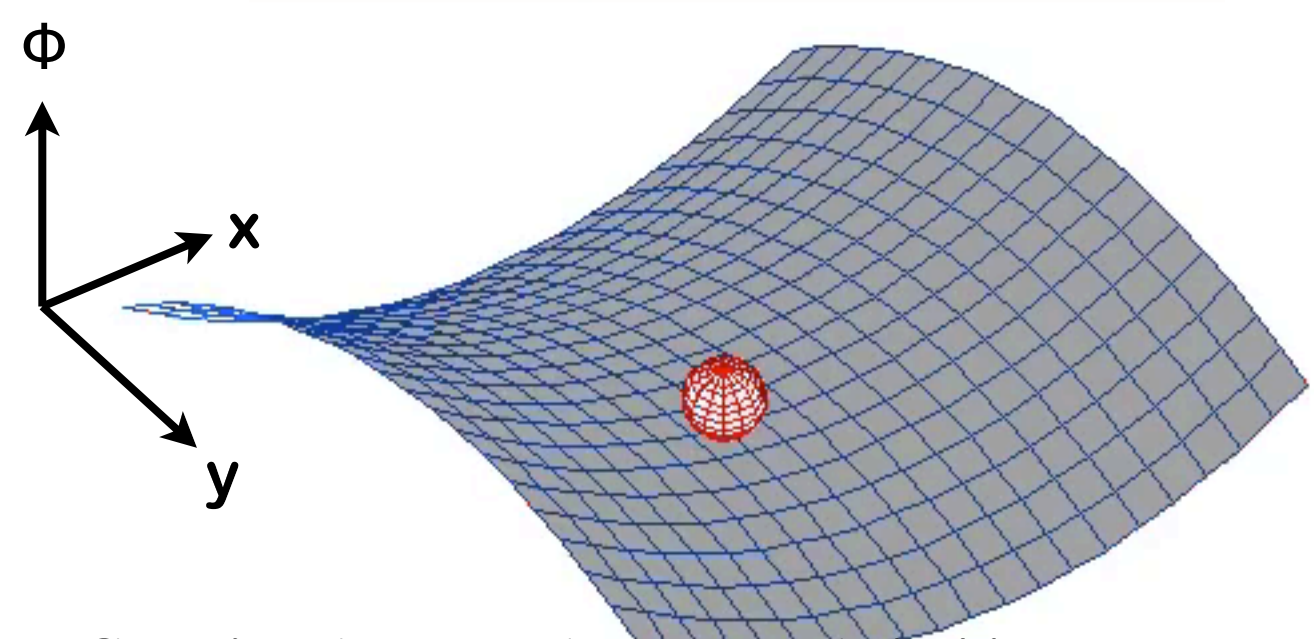
RF ion trapping

See also other lectures on ion trapping in this school

- Trapping = confine particles in a suitable potential minimum. For the trapping of ions electric potentials Φ are an obvious choice.
- The Laplace equation $$\left(\frac{\partial^2}{\partial x^2}+\frac{\partial^2}{\partial y^2}+\frac{\partial^2}{\partial z^2}\right)\Phi=0$$
  forbids the formation of a potential minimum with static electric fields in 3D space.



- Solution: radiofrequency ion traps
  Trap ions dynamically in the x,y plane using time-varying voltages V_{RF} applied to the electrodes: $V_{RF}=V_{RF,0}\cos\left(\Omega_{RF}t\right)$



- Linear Paul trap: four electrodes arranged in a quadrupolar configuration
- The RF fields create a rotating potential saddle point which dynamically confines the ions.
- Along the trap axis (=z axis) the ions are confined using static potentials V_{end} applied to the "endcap electrodes".

- The **trajectory** u(t), u ∈ {x,y,z}, of a trapped ion is approximately given by:

$$u(t) \approx u_0 \cos(\omega_u t + \varphi)\left[1 + \frac{1}{2} q_u \cos(\Omega_{\mathrm{RF}} t)\right]$$

- u_0: motional amplitude (reduced by laser cooling)
- φ: phase depending on initial conditions
- 1: **secular motion** (see below)
- $\frac{1}{2} q_u \cos(\Omega_{\mathrm{RF}} t)$: **micromotion** (fast oscillating motion driven by the RF fields)

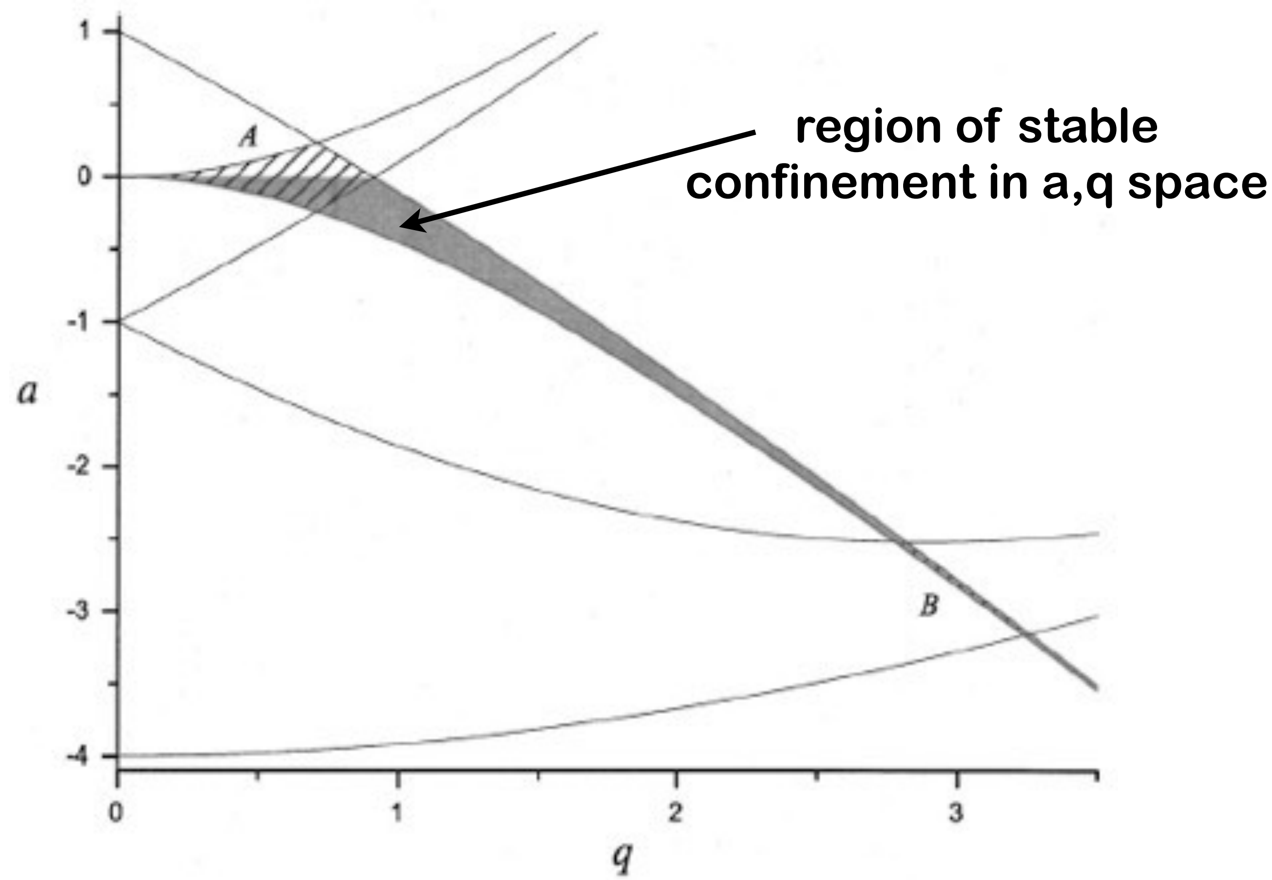
D. Berkeland et al., J. Appl. Phys. 83 (1998), 2025

- The conditions for stable confinement only depend on the **Mathieu parameters a,q**:

$$a_x = a_y = -\frac{1}{2} a_z = -\eta \frac{4QV_{\mathrm{end}}}{m\Omega_{\mathrm{RF}}^2 z_0^2}$$

$$q_x = -q_y = \frac{4\,QV_{\mathrm{RF},0}}{m\Omega_{\mathrm{RF}}^2 r_0^2}, \qquad q_z = 0$$

η, z_0, r_0 ... geometrical parameters of the trap



M. Drewsen and A. Broner, Phys. Rev. A 62 (2000), 045401

- **Pseudopotential-well model:**
  the secular motion of the ion can be described in terms of an effective time-independent pseudopotential Φ^* after time-averaging over the fast oscillating micromotion:

$$\Phi^*(r,z) = \frac{1}{2} m\omega_r^2 r^2 + \frac{1}{2} m\omega_z^2 z^2$$

with

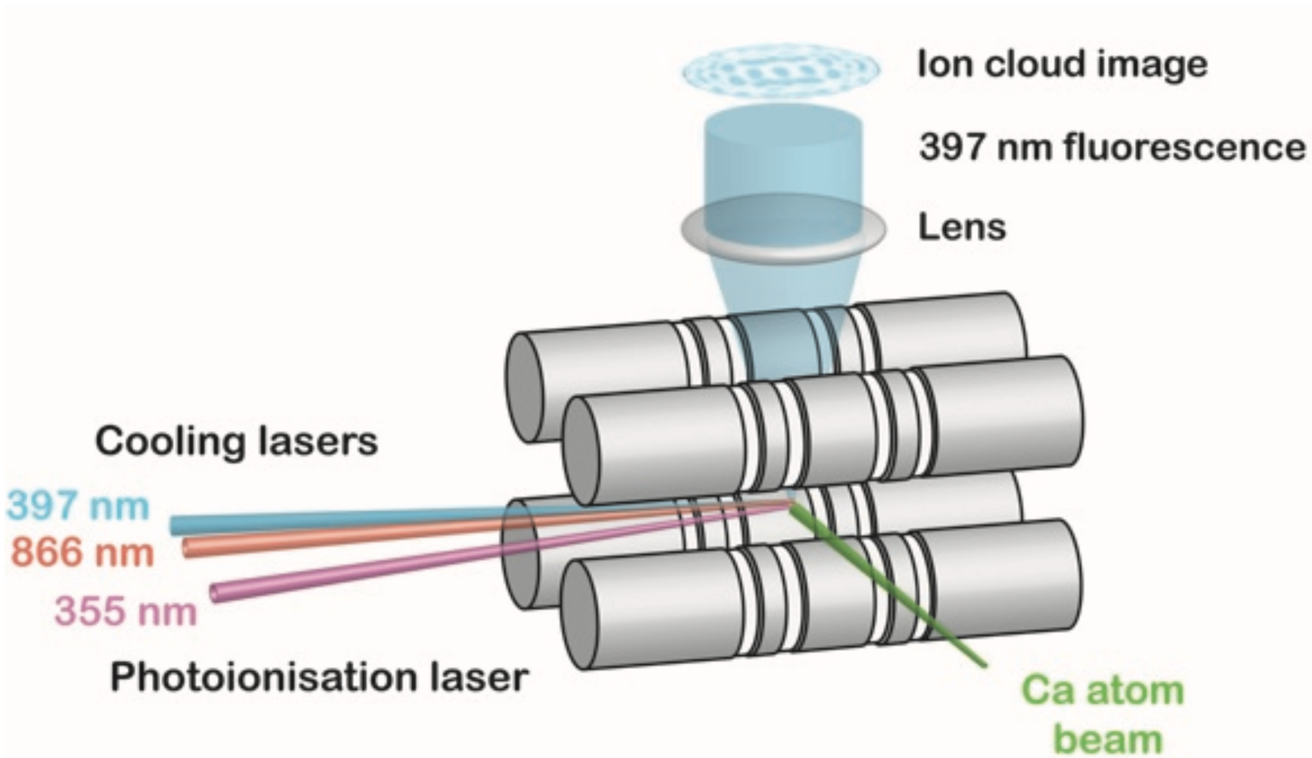
$$r = \sqrt{x^2+y^2}$$

$$\omega_i = \sqrt{a_i + (1/2)q_i^2}\,\frac{\Omega_{\mathrm{RF}}}{2}$$

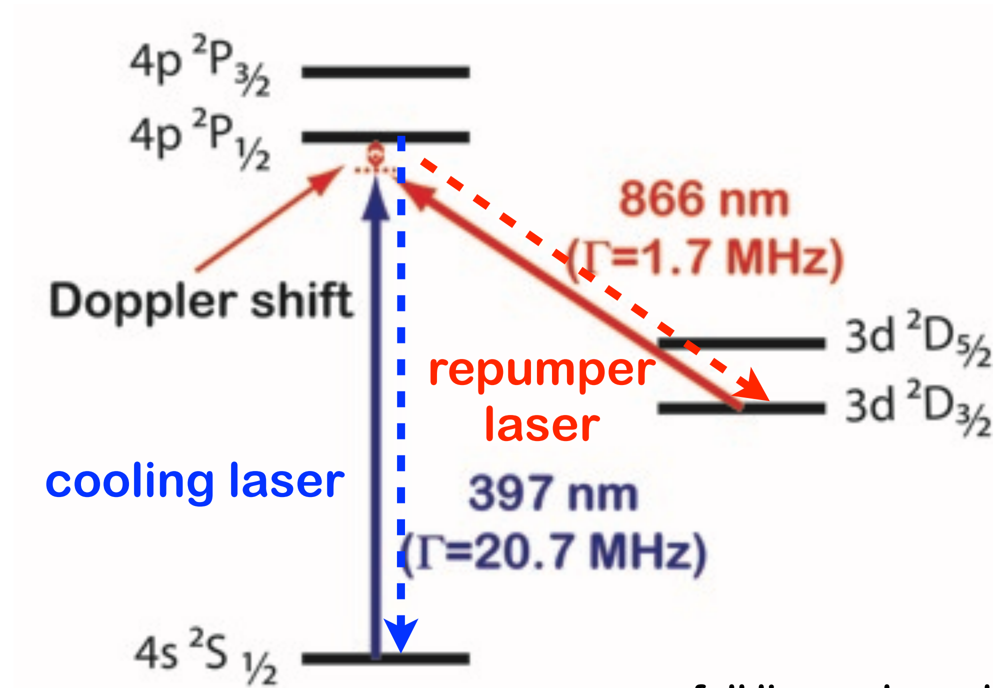
F. G. Major et al., Charged Particle Traps Vol. I+II, Springer, 2005 + 2007
D. Gerlich, Adv. Chem. Phys. 82 (1992), 1

Laser cooling of trapped ions

- For large ion ensembles, laser cooling of all translational degrees of freedom can be achieved by cooling only along one axis as the Coulomb interaction between the ions and trap anharmonicities couples all translational degrees of freedom.
- Typical ions used for laser cooling are alkaline earth ions (Be^+, Mg^+, Ca^+, Ba^+) often produced directly inside the trap by photoionisation of their neutral atoms.



- Example: laser cooling of $^{40}Ca^+$:



full lines = laser beams
dashed lines = spontaneous emission

Sympathetic cooling of molecular ions by laser-cooled atomic ions

PHYSICAL REVIEW A, VOLUME 62, 011401(R)

Formation of translationally cold MgH^+ and MgD^+ molecules in an ion trap

K. Mølhave and M. Drewsen*

Institute of Physics and Astronomy, University of Aarhus, 8000 Aarhus C, Denmark

(Received 1 March 2000; published 2 June 2000)

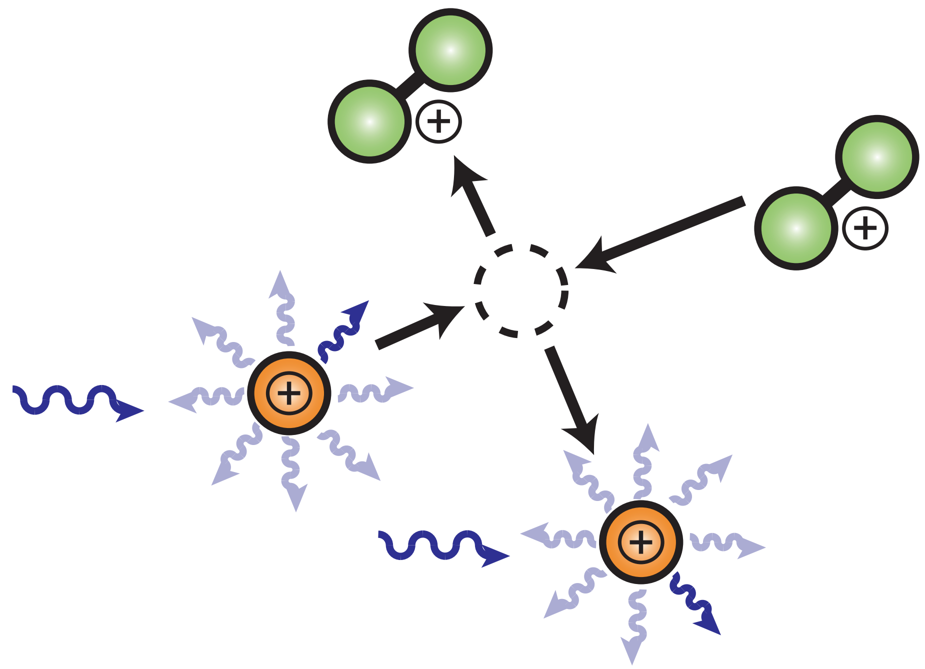
We have produced and cooled the molecular ions MgH^+ and MgD^+ in a linear Paul trap. These ions were generated by the photochemical reactions $Mg^+(3p\,^2P_{3/2})+H_2\ (D_2)\rightarrow MgH^+\ (MgD^+)+H\ (D)$, and identified by the radial separation in the trap of ions with different charge-to-mass ratios. The molecular translational motion was cooled sympathetically by Coulomb interaction with laser-cooled Mg^+ ions to a temperature estimated to be below 100 mK. Ordered structures (ion crystals) containing more than 1000 ions, with more than 95% being molecular ions, were obtained. Such translationally cold and well-localized samples of molecular ions could become very useful for molecular physics and chemistry.

PACS number(s): 32.80.Pj, 34.50.Rk, 82.30.Fi

Sympathetic cooling of molecular ions

Principle:

- Molecular ions are simultaneously trapped with laser-cooled atomic ions
- The molecular ions exchange kinetic energy in Coulomb-collisions with the atomic ions
- The energy is removed by laser cooling on the atomic ions

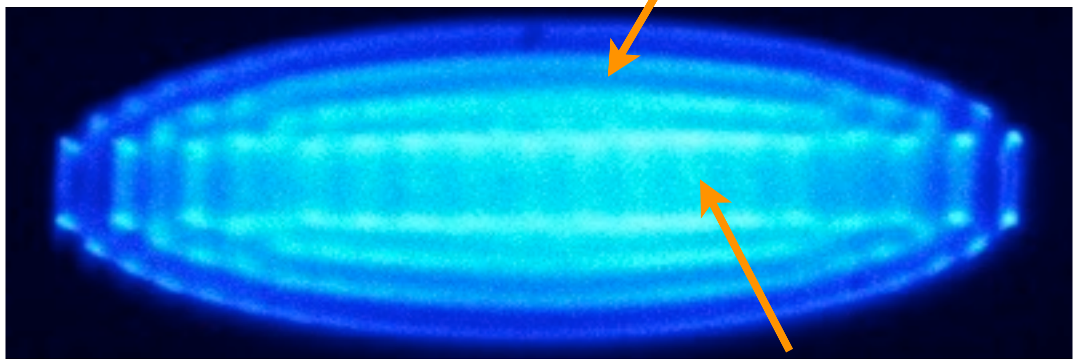


T. Baba & I. Waki , Jpn. J. Appl. Phys. 35 (1996), L1134
K. Molhave & M. Drewsen, Phys. Rev. A 62 (2000), 011401
S. Willitsch et al., Int. Rev. Phys. Chem. 31 (2012), 175

Molecular (or bi-component) Coulomb crystals:

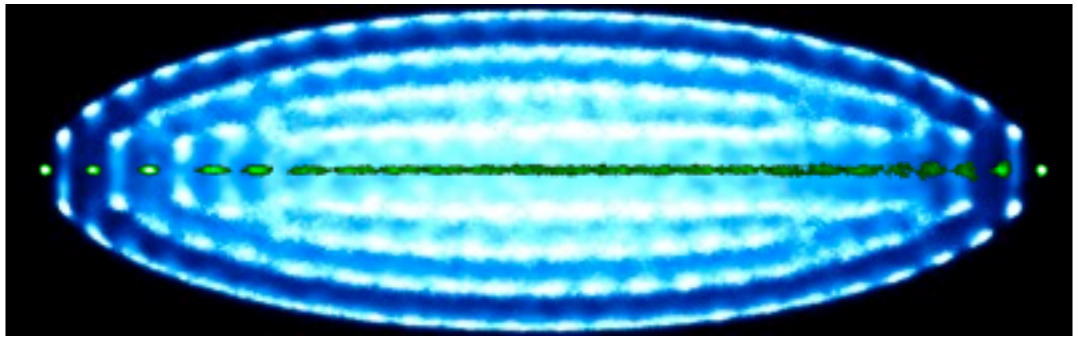
Experiment:

Laser-cooled Ca^+ ions



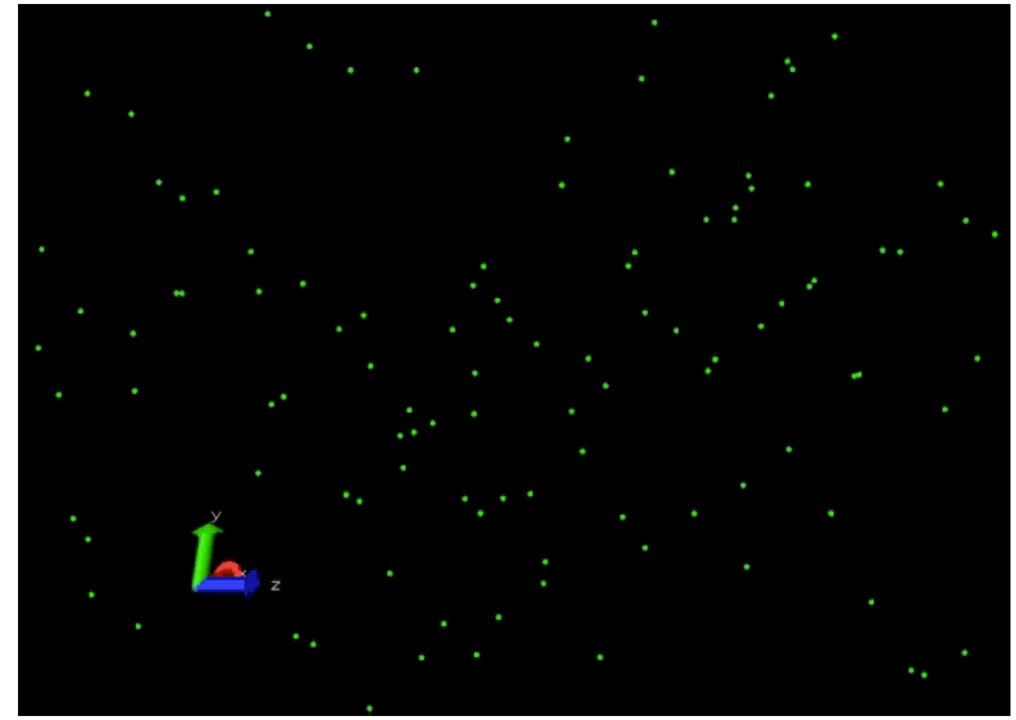
Sympathetically-cooled N_2^+ ions

Simulation:



Molecular dynamics simulations of Coulomb crystals

- M.T. Bell et al., Faraday Discuss. 142 (2009), 73
- I. Rouse and SW, PRA 92 (2015), 053420
- C.B. Zhang et al., PRA 76 (2007), 012719
- T. Matthey et al., PRL 91 (2003), 165001



Force model :

$$m_i\ddot{\mathbf{x}}_i = q_i\nabla\left[\frac{U_{\mathrm{RF}}}{2r_0^2}(x^2-y^2)\cos(\Omega_{\mathrm{RF}}t)+\frac{\eta U_{\mathrm{END}}}{z_0^2}\left(z^2-(1/2)\cdot(x^2+y^2)\right)\right]$$

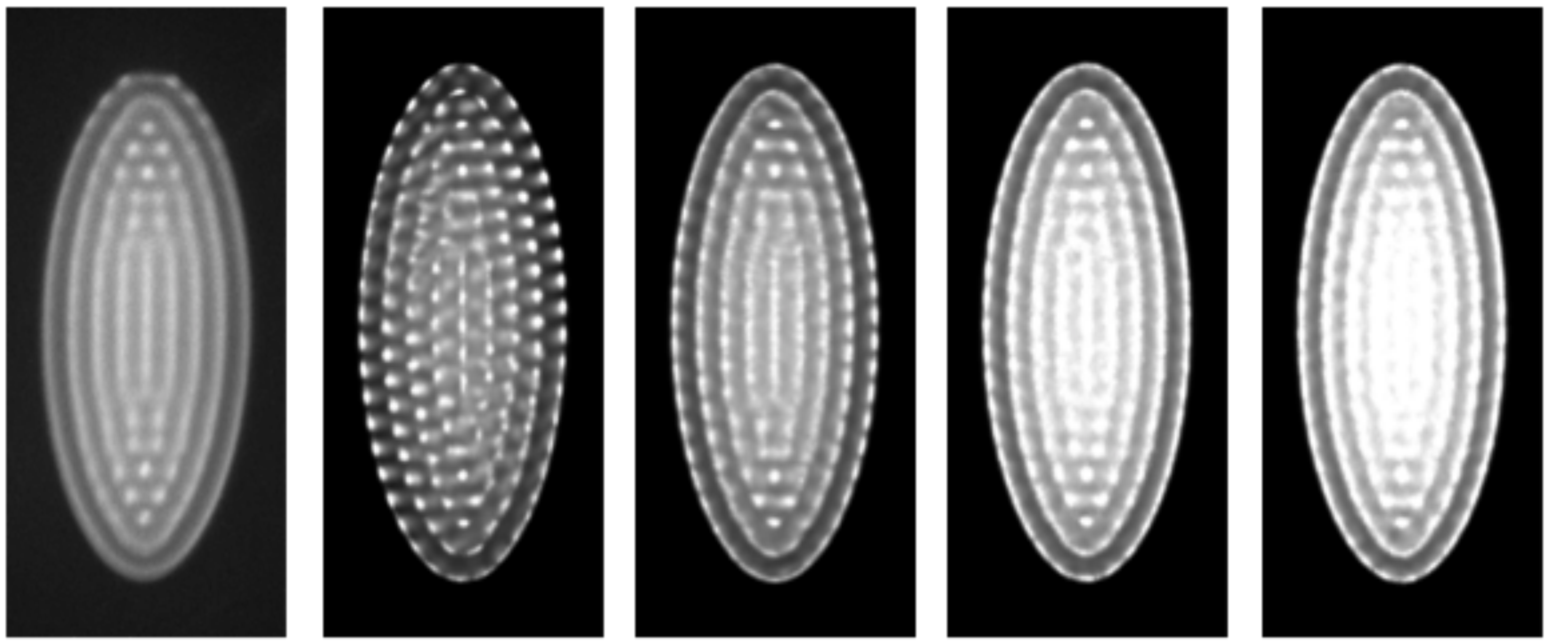
$$+\frac{q_i}{4\pi\epsilon_0}\nabla\left[\sum_j\frac{q_j}{r_{ij}}\right]$$

$$-\beta\dot{z}$$

$$+\mathbf{F}_{\mathrm{heating}}$$

- Harmonic/numerical time-dependent potentials for the ion trap
- Coulomb interaction between ions
- Laser-cooling force
- Effective force to model heating mechanisms

- **Determination of the secular (thermal) energies of the ions**



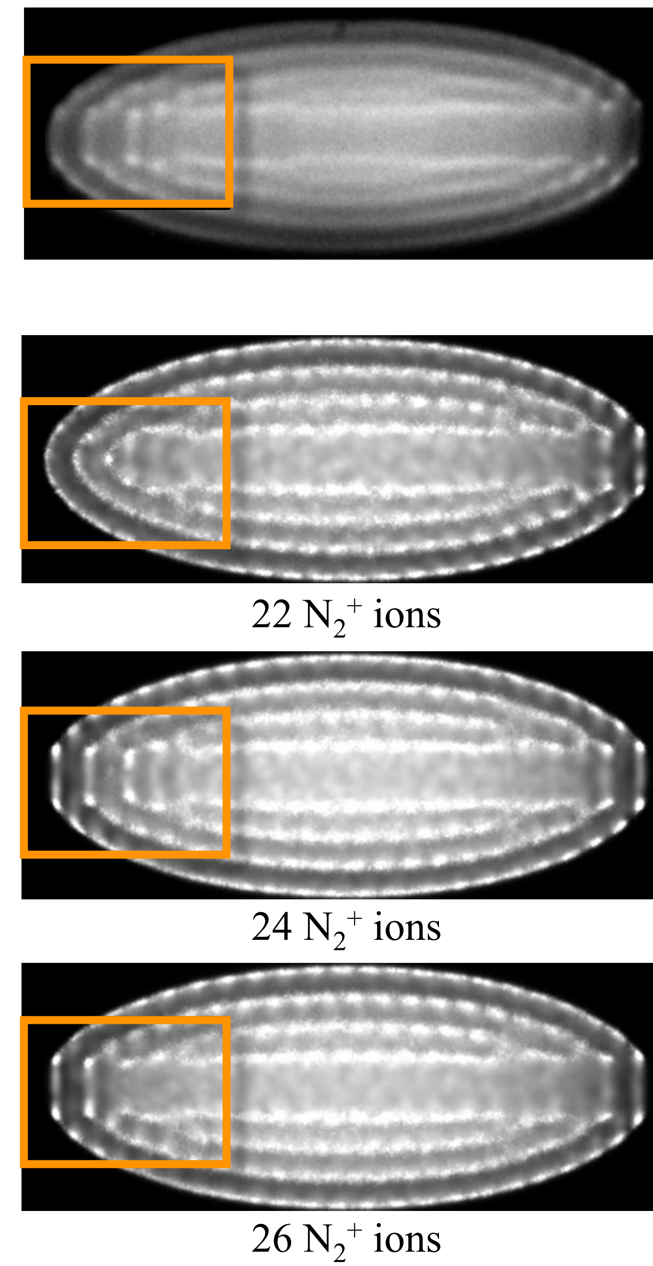
Expt. | 5 mK | 7 mK | 9 mK | 10 mK

- **Determination of ion numbers**

Bicomponent crystal after loading with N_2^+ ions

Expt.:

Sim.:

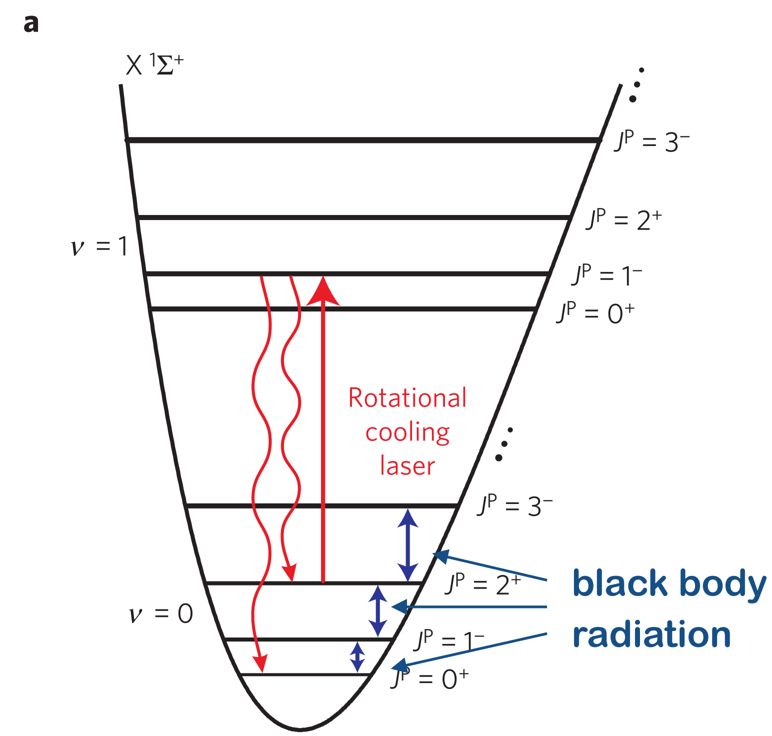


2. Internal state preparation of cold molecular ions

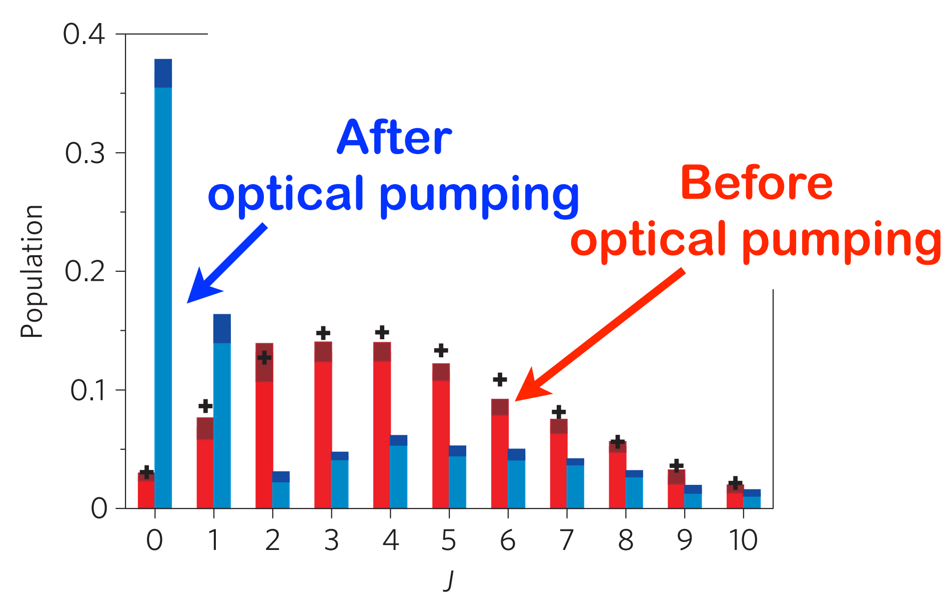
Rotational laser cooling

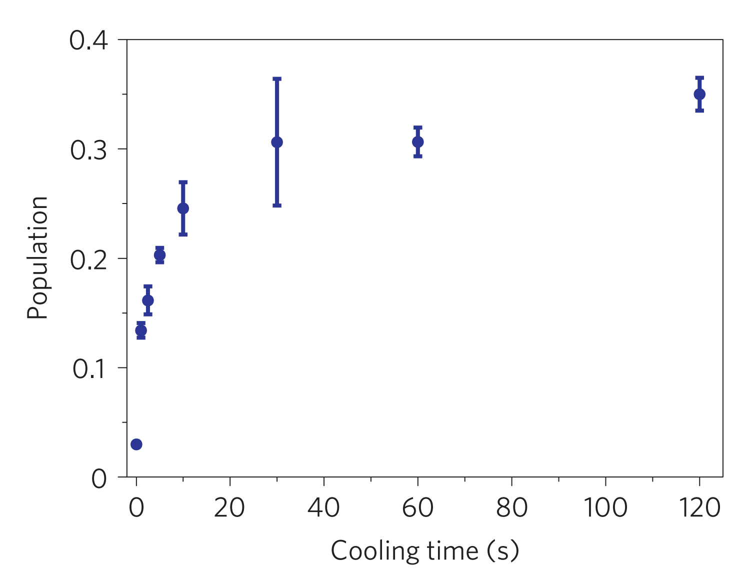
Staanum et al., Nature Phys. 6 (2010), 271
see also: Schneider et al., Nature Phys. 6 (2010), 275

- **Principle:** sympathetically cool the translational motion of molecular ions and then use optical pumping by laser fields to accumulate the population in a specific rotational-vibrational level
- **Example: MgH^+.** Use an IR laser to pump on the v=0,J=2 → v=1, J=1 transition. The v=1, J=1 level can only fluoresce down to the v=0, J=0 and J=2 levels (selection rules !).
- Transitions between rotational levels in the v=0 state induced by blackbody radiation constantly redistribute the population.
- In combination with the optical pumping, an accumulation of the population in the v=0, J=0 level results.



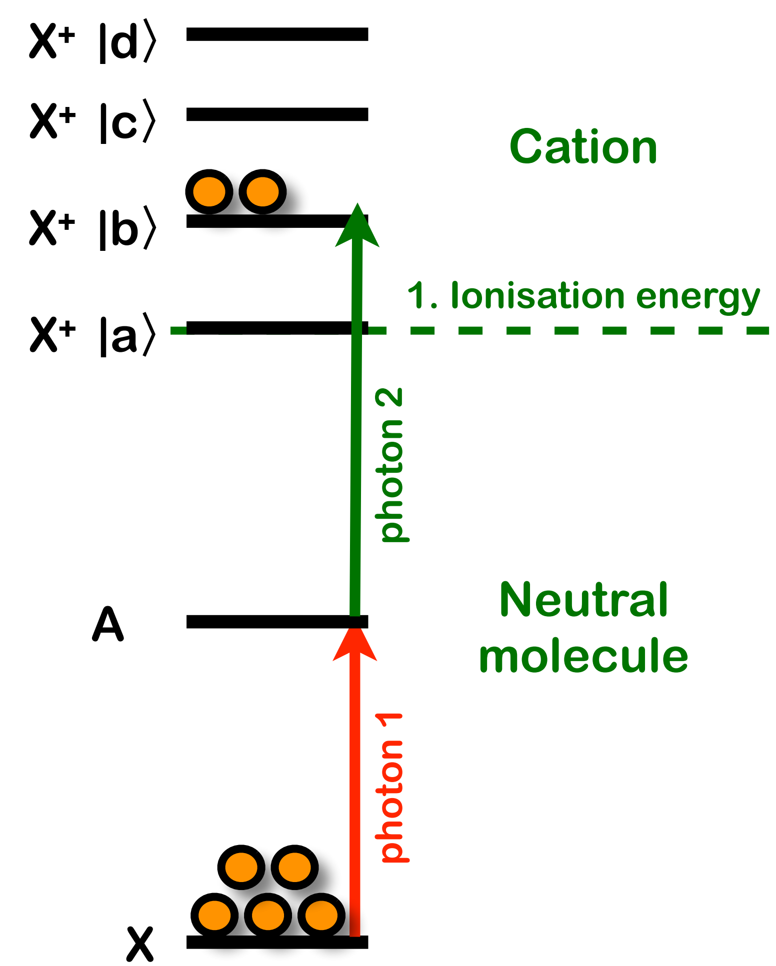
Ro-vibrational energy level scheme of MgH^+





Sympathetic cooling of state-prepared molecular ions

- **Principle:** Prepare the molecular ions right from the beginning in a specific rotational-vibrational state by threshold photoionization of their neutral precursor moelcules (i.e., photoionization just above a given ionization energy) and then sympathetically cool their translational motion.

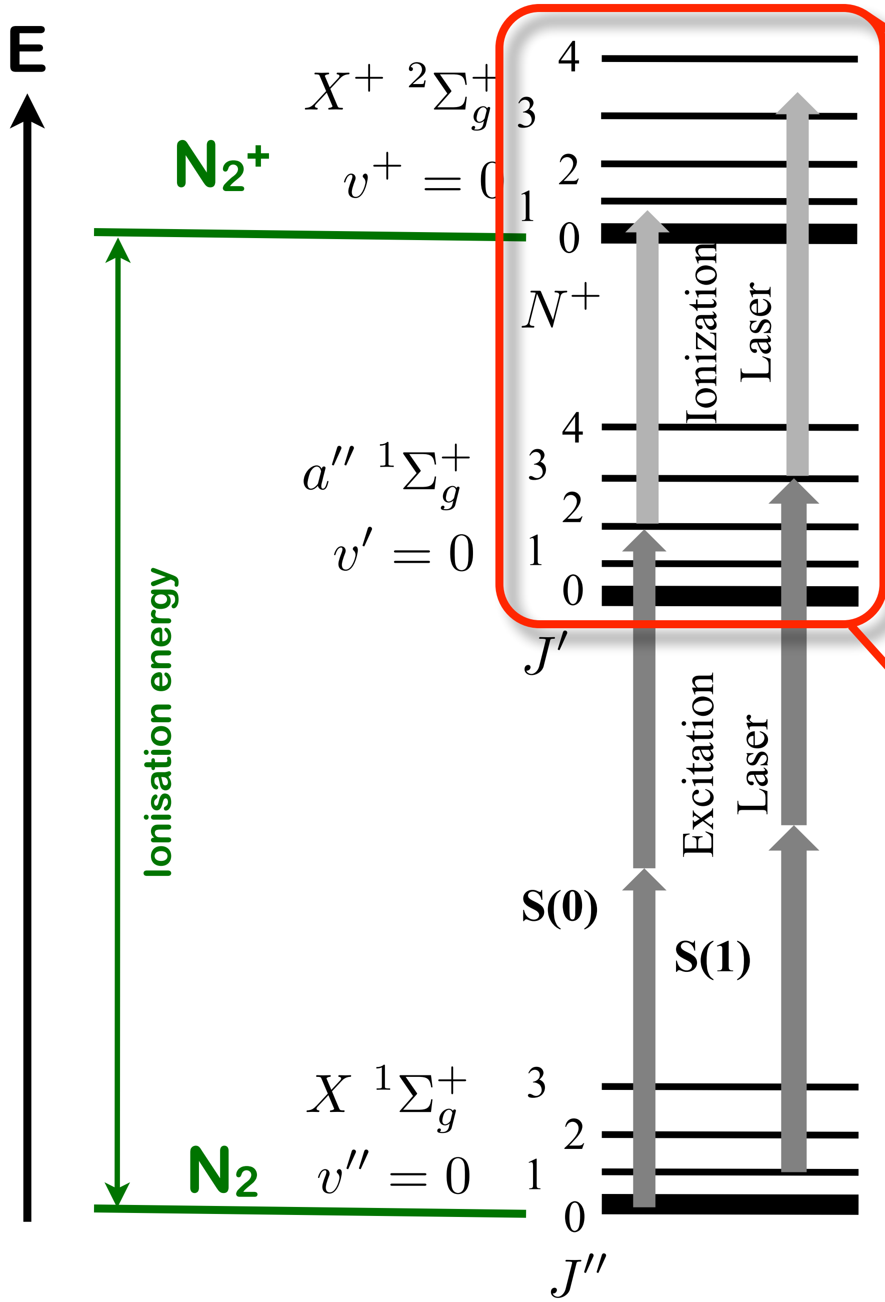


Threshold photoionization of molecules

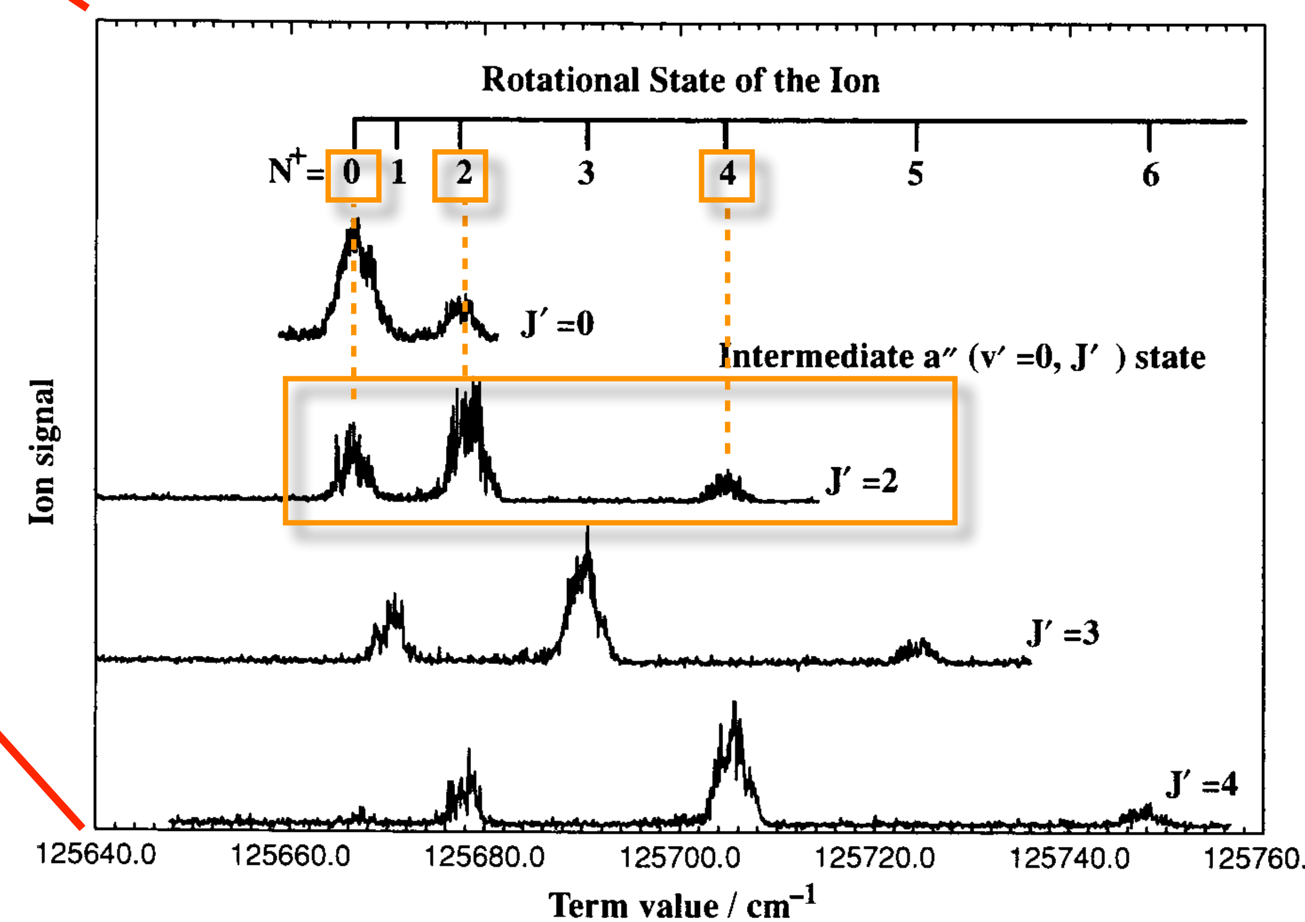
X. Tong, A. H. Winney and S. Willitsch, Phys. Rev. Lett. 105 (2010), 143001

Example: threshold photoionization of N_2:

- [2+1']-photon resonance-enhanced threshold-photoionisation scheme for N_2

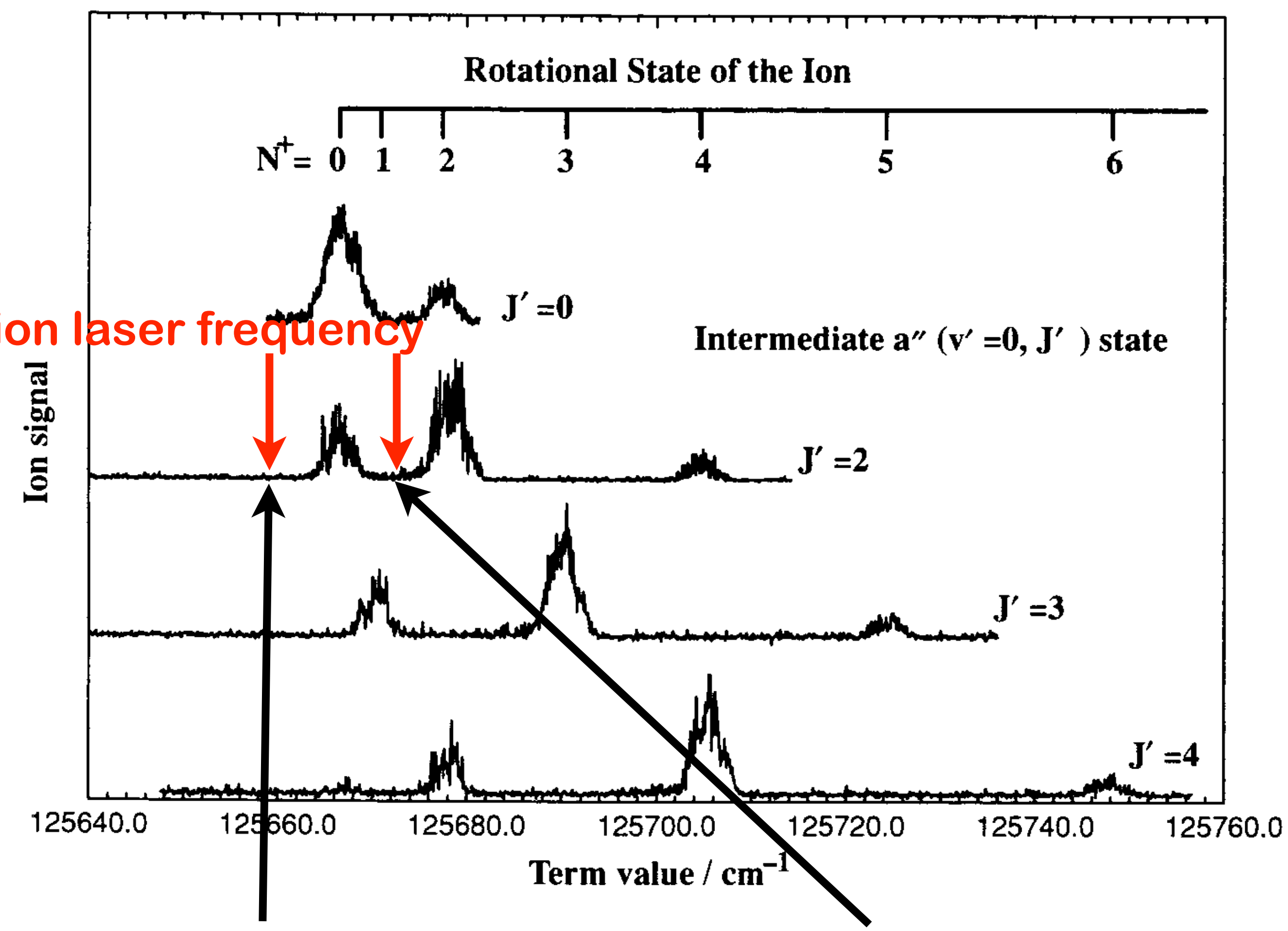


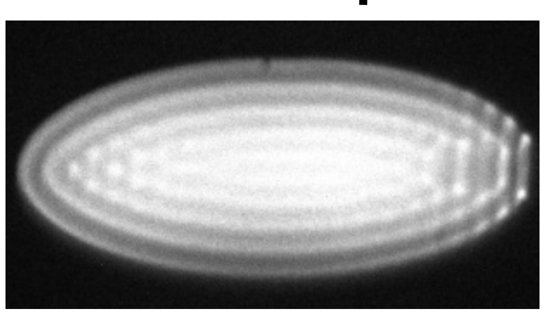
- Rotationally resolved photoelectron spectrum of N_2 showing the cationic rotational levels N^+ produced by threshold photoionisation out of well-defined intermediate rotational levels J'

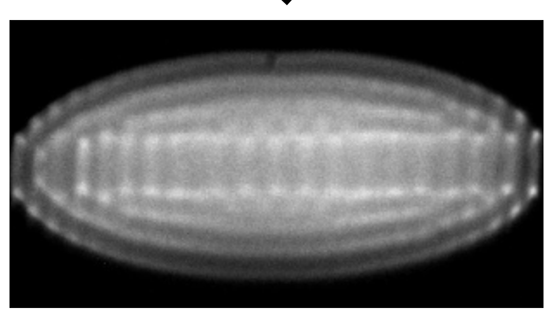


S.R. Mackenzie et al., Mol. Phys. 86 (1995), 1283

- [2+1'] resonance-enhanced threshold photoionisation via the a" $^1\Sigma^+_g$, J'=2 intermediate state of N_2:

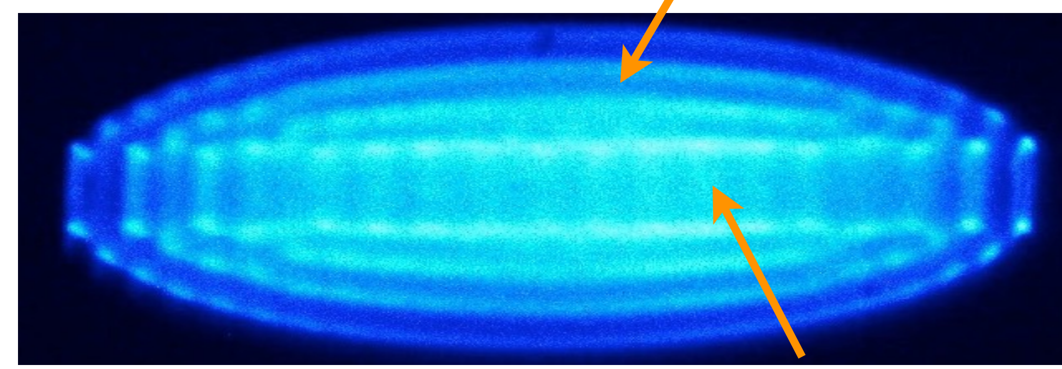






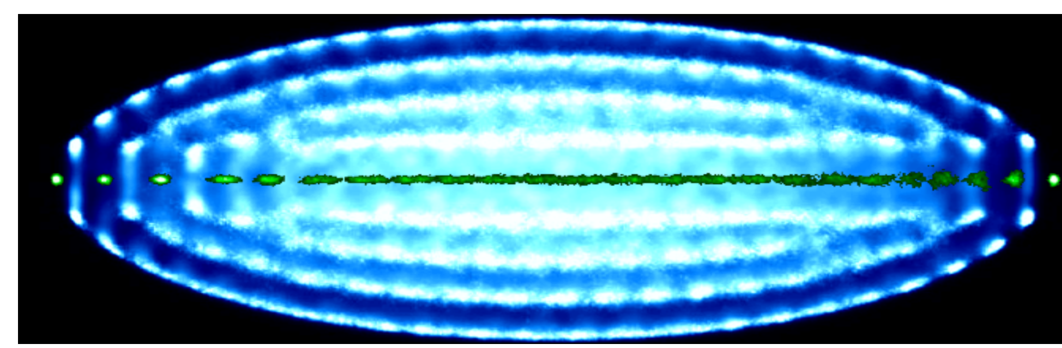
Experiment:

Laser-cooled Ca^+ ions



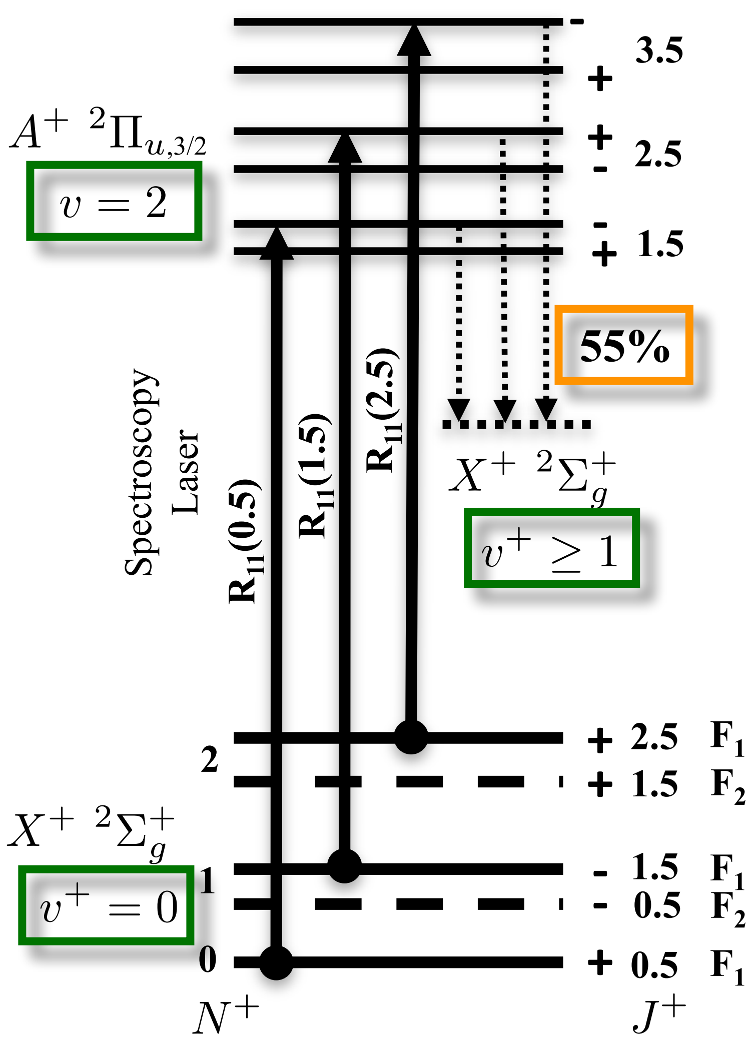
Sympathetically-cooled N_2^+ ions

Simulation:

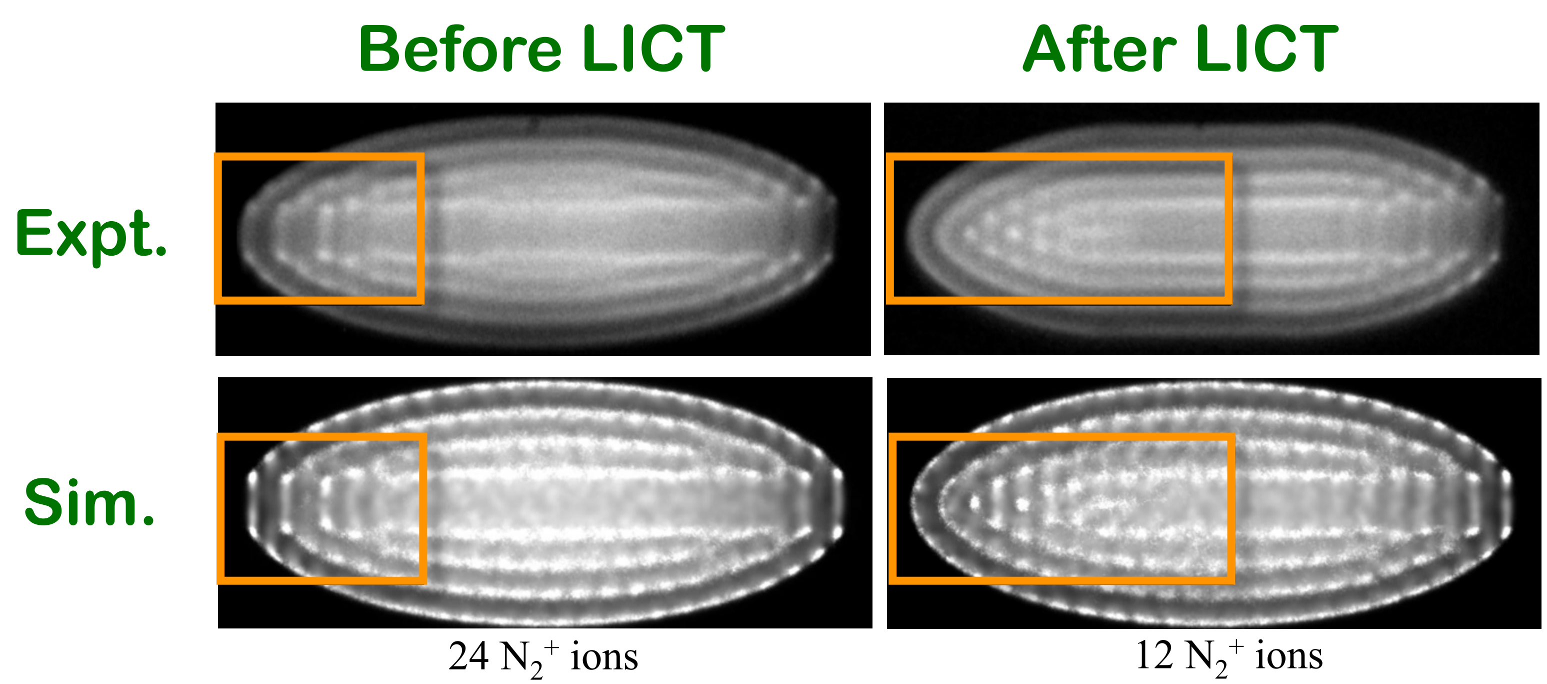


- **Population diagnostics: Laser-induced charge-transfer (LICT) spectroscopy**:
  - Optically pump the population out of selected rotational levels to vibrationally excited states of N_2^+.
  - Detect the vibrationally excited molecules by a vibrational-state-selective charge transfer process with Ar atoms:

$$N_2^+ + Ar \rightarrow N_2 + Ar^+ \quad (v^+ \geq 1)$$
$$N_2^+ + Ar \not\rightarrow \quad (v^+ = 0)$$



S. Schlemmer et al.,
Int. J. Mass Spectrom. 185 (1999), 589



LICT efficiency out of N^+=0: 50±8 % (max. 55 %)

Total population in N^+=0: 93±11% (averaged over 5 expts.)

Alternative state preparation / cooling schemes:

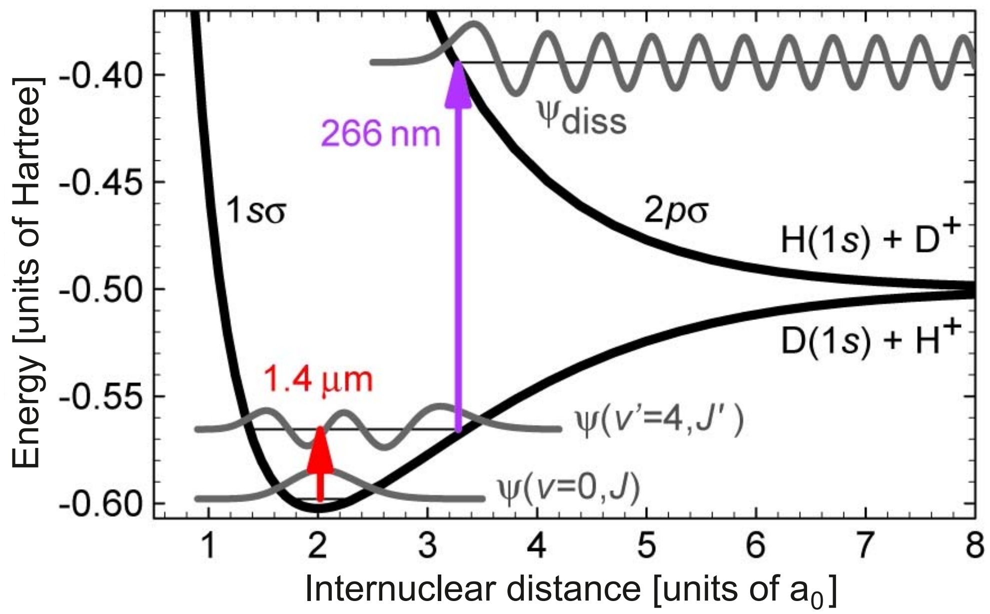
- **Combination of sympathetic and He buffer-gas cooling**
  A. K. Hansen et al., Nature 508 (2014), 76
- **Broadband rotational cooling with a fs laser**
  C.-Y. Lien et al., Nat. Commun. 5 (2014), 4783
- **Vibrational buffer-gas cooling with ultracold atoms**
  W.G. Rellegert et al., Nature 495 (2013), 490
- **Probabilistic state preparation**
  I.S. Vogelius et al., J. Phys. B 39 (2006), S1259; K. Najafian et al., Nat. Commun. 11 (2020), 4470

3. Precision spectroscopy of cold molecular ions

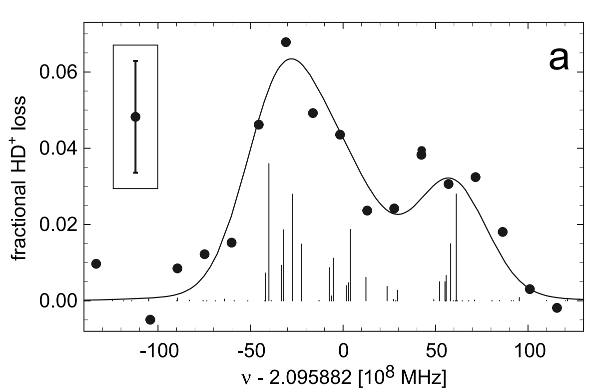
- New types of clocks
  PRL 113 (2014), 023004 ; J. Mol. Spectr. 300 (2014), 37
- Tests of validity of QED
  PRL 113 (2014), 023004; PRL 98 (2007), 173002; PRL 110 (2013), 193601; JCP 140 (2014), 104303
- Tests of a time variation of fundamental constants (m_p/m_e, ...)
  PRL 106 (2011), 100801; PRL 98 (2007), 173002; PRL 113 (2014), 210802; PRA 89 (2014), 032509
- Search for new physics (e.g., dipole moment of the electron, fifth forces, extra dimensions, ...)
  NJP 17 (2015), 033015; Science 343 (2014), 269; PRD 87 (2013), 112008; Nature Phys. 10 (2014), 933
- Probing the energy difference between enantiomeric molecules
  Angew. Chemie Int. Ed. 41 (2002), 4618
- .....

Basic techniques: Photodissociation spectroscopy of vibrational overtones in sympathetically cooled HD^+ ions

Excitation scheme

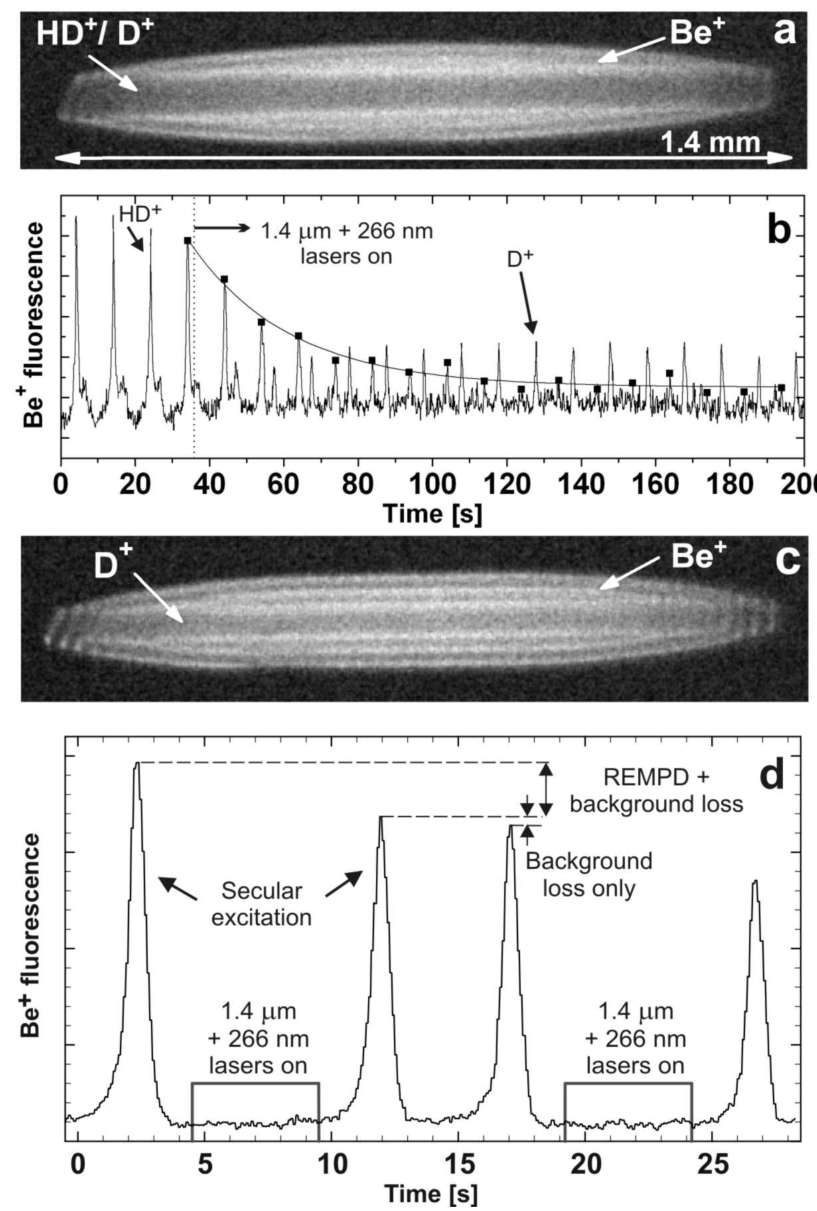


Example spectrum with unresolved hyperfine structure



(v'=4, J'=1) ← (v=0, J=2)

Detection scheme



Initial crystal

Detection of sympathetically cooled ions by repeated cycles of resonant excitation of secular ion motions under laser irradiation (see also d)

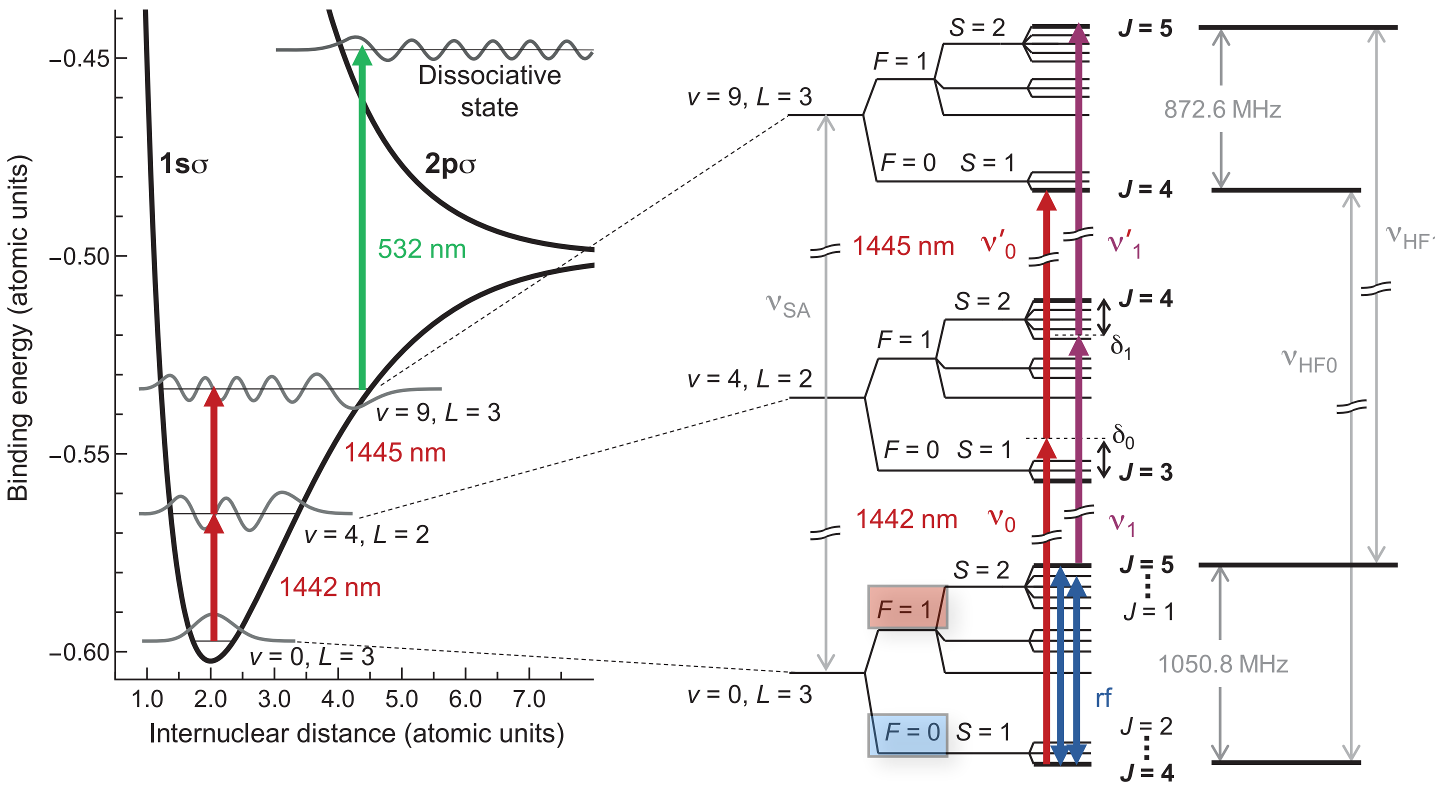
Crystal after photodissociation of all HD^+ ions

Magnification of b: single measurement cycles of laser irradiation and secular excitation

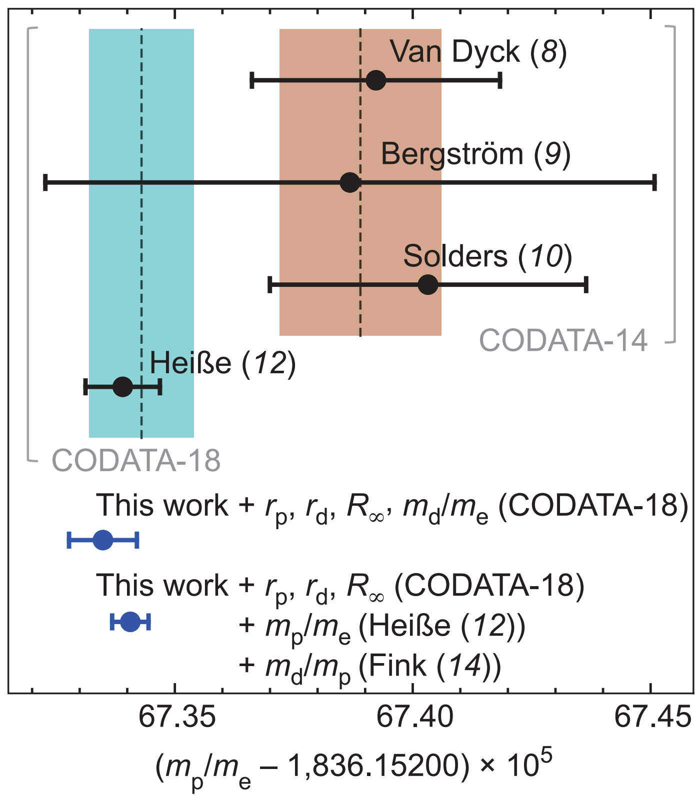
B. Roth et al., Phys. Rev. A 74 (2006), 040501

Hyperfine-resolved Doppler-free two-photon spectroscopy of HD^+ : precise determination of the proton-electron mass ratio m_p/m_e at the ppt level

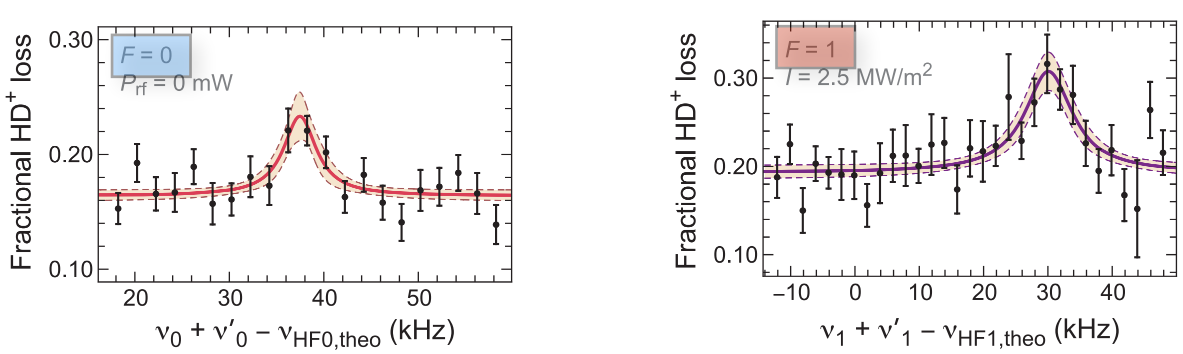
- **Energy-level and excitation scheme for two-photon spectroscopy**



- **Comparison of determinations of m_p/m_e**



- **Spectroscopic lines originating from F=0 and F=1**



S. Patra et al., Science 369 (2020), 1238

Spectroscopy of “forbidden” infrared transitions in cold molecular ions

- **Motivation:** precision spectroscopic measurements on single isolated molecules

PRL **113,** 023004 (2014) PHYSICAL REVIEW LETTERS

Simplest Molecules as Candidates for Precise Optical Clocks

S. Schiller,[1] D. Bakalov,[2] and V. I. Korobov[3]

PHYSICAL REVIEW A **89**, 032509 (2014)

Test of m_p/m_e changes using vibrational transitions in N_2^+

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Geetha Gopakumar, Minori Abe, and Masahiko Hada
Department of Chemistry, Tokyo Metropolitan University, Hachioji, Tokyo, Japan

Matthias Keller
Department of Physics and Astronomy, University of Sussex, Brighton, United Kingdom
(Received 12 February 2014; published 13 March 2014)

PHYSICAL REVIEW A **85**, 022308 (2012)

Temperature-independent quantum logic for molecular spectroscopy

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Jesús Pérez-Ríos, José Campos-Martínez, and Marta I. Hernández
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Journal of Molecular Spectroscopy 300 (2014) 37–43

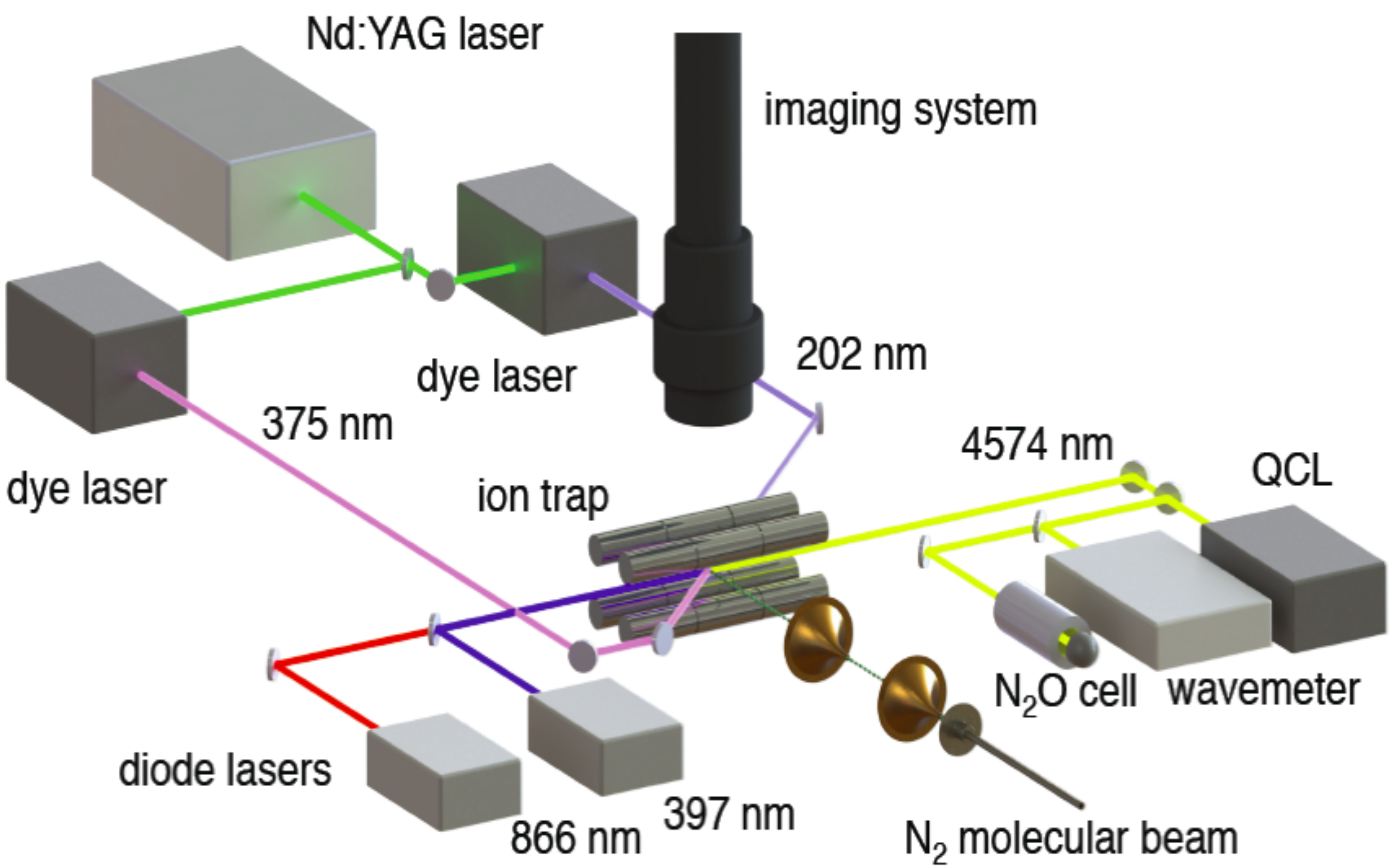
H_2^+ and HD^+: Candidates for a molecular clock

J.-Ph. Karr *

- **“forbidden” transition:** forbidden within the electric dipole approximation
- **Infrared spectra of homonuclear diatomics (H_2, N_2, …):** dipole forbidden, but allowed within the electric-quadrupole (E2) approximation
- **Quadrupole spectra:** extremely weak, extremely narrow spectral lines (ca. factor 10^{10} compared to dipole-allowed infrared spectra)

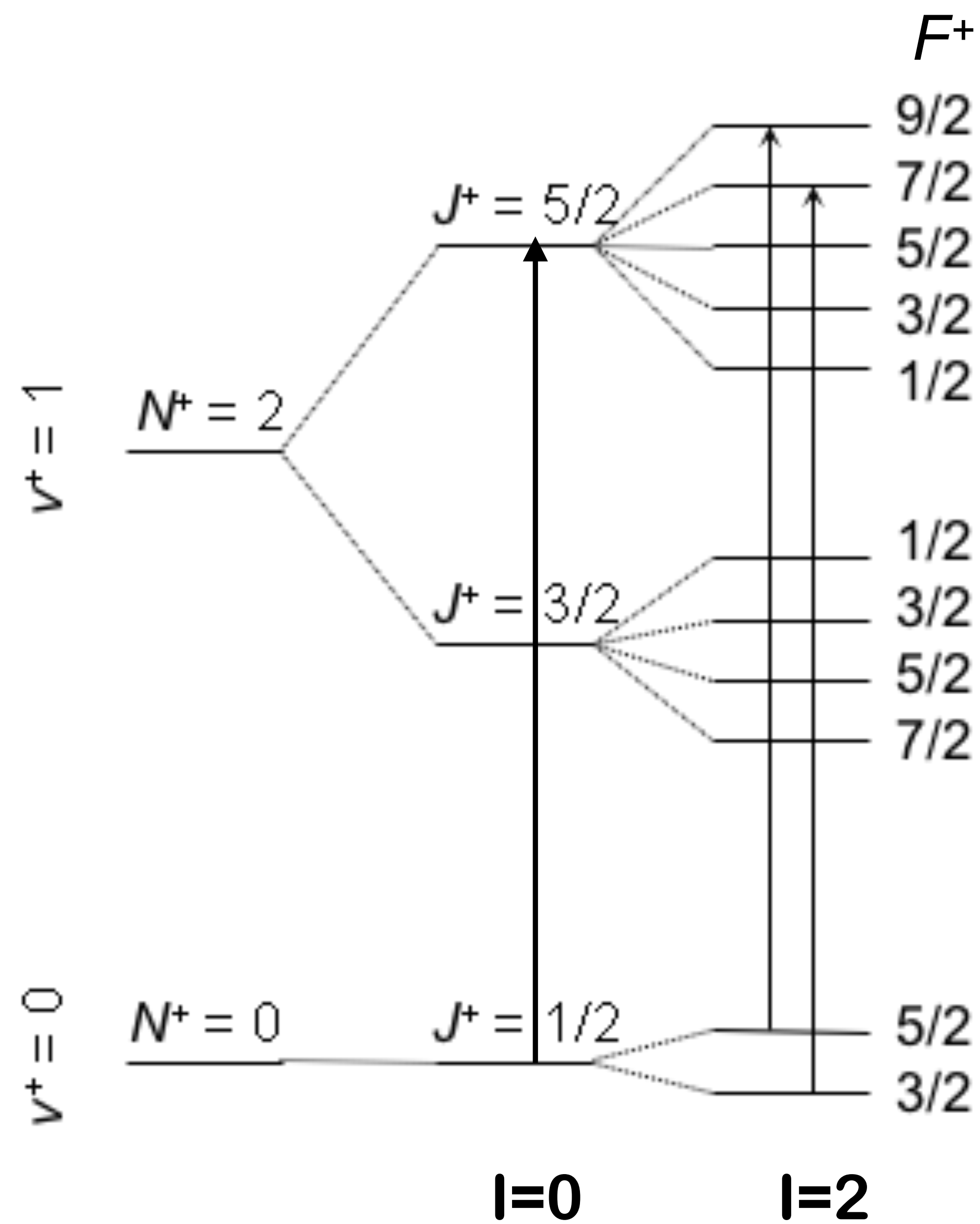
N_2^+ quadrupole infrared spectroscopy: experiment

- Sympathetically cool 20-25 state-selected N_2^+ ions
- Irradiate N_2^+ ions with 200 mW focused IR radiation at 4.5 μm for 2 minutes
- Detect excitation to $v^+=1$ by charge transfer with Ar

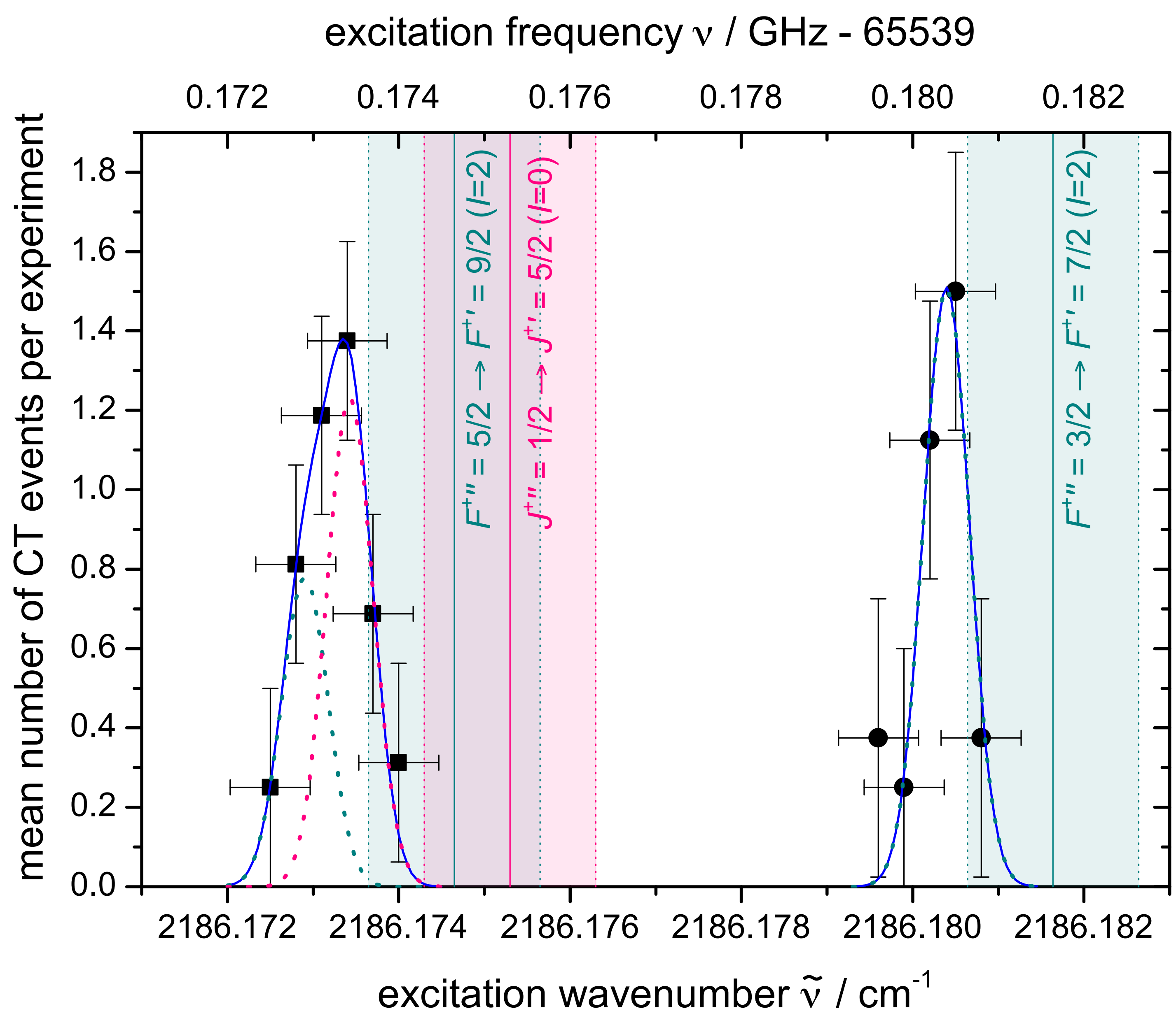


M. Germann, X. Tong, SW, Nature Phys. 10 (2014), 820

The v=0→1 quadrupole spectrum of N_2^+



Principal hyperfine transitions with ΔN=ΔJ=ΔF=2 in the S(0) line of the I^+=0,2 nuclear spin isomers



Spectrum of the principal HF transitions of he S(0) line

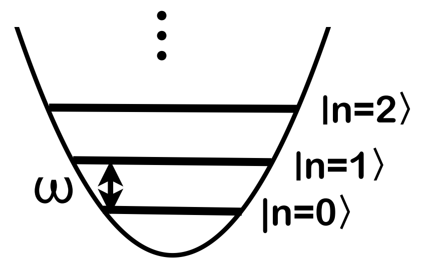
M. Germann, X. Tong, SW, Nature Phys. 10 (2014), 820

4. Molecular-ion quantum technologies and spectroscopy

Resolved sideband cooling of ions into the quantum regime

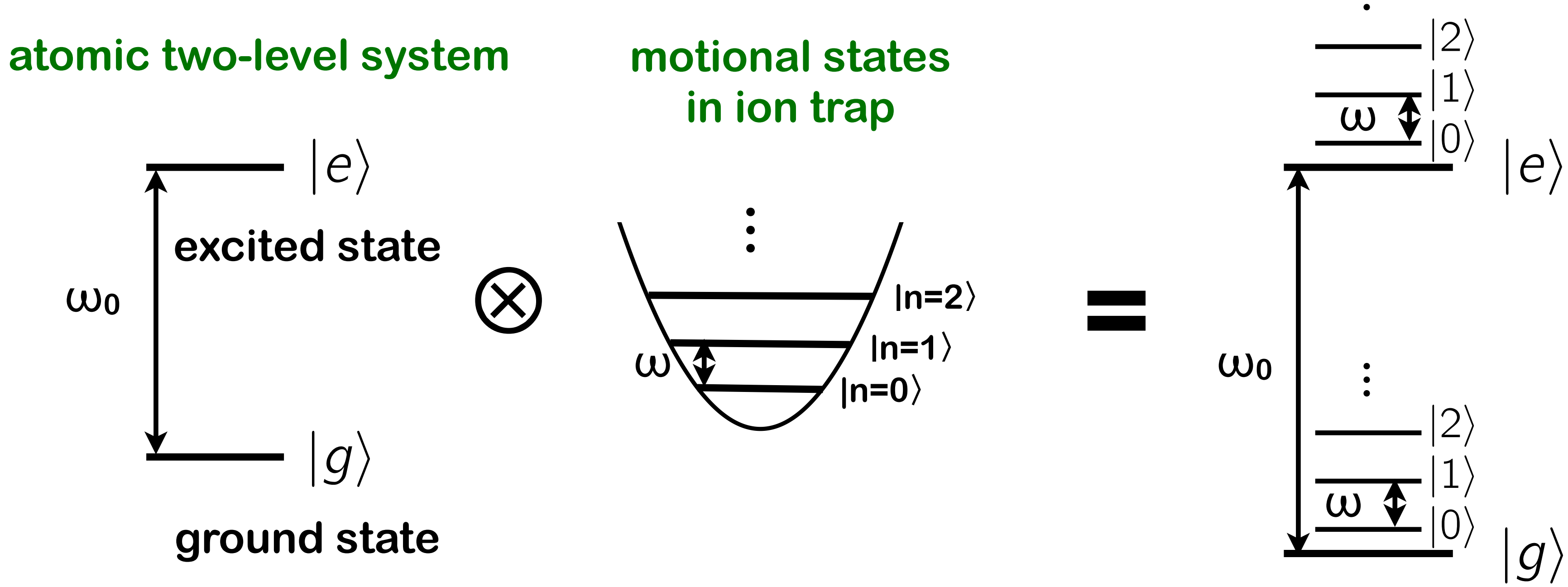
See other lectures on ion trapping in this school for details

- The motion of an ion in the harmonic pseudopotential of the ion trap corresponds to a QM harmonic oscillator problem:

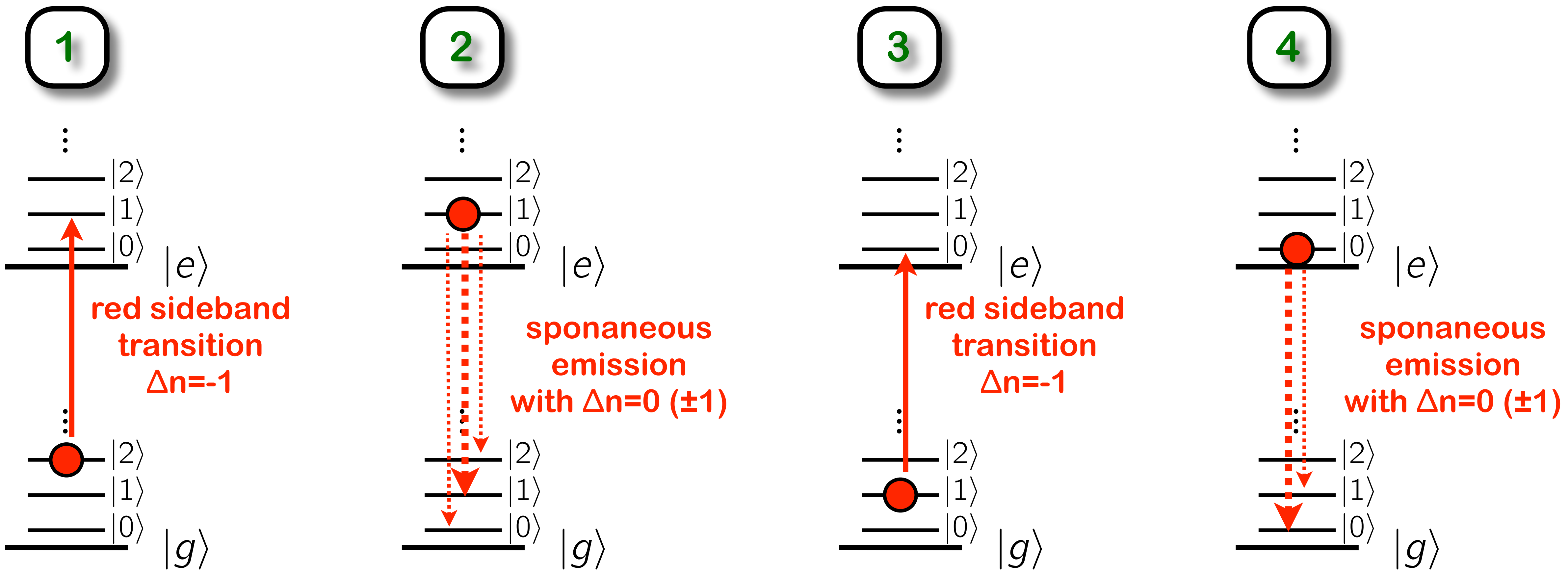


  - Motional states = the harmonic oscillator eigenstates $|n\rangle$, n = 0,1,2,..
  - Motional states form a ladder of equidistant energy levels $E_n = \left(n + \frac{1}{2}\right)\hbar\omega$

- **Resolved sideband cooling:** cooling to the motional ground state by addressing the individual motional states



Simple resolved sideband cooling scheme:

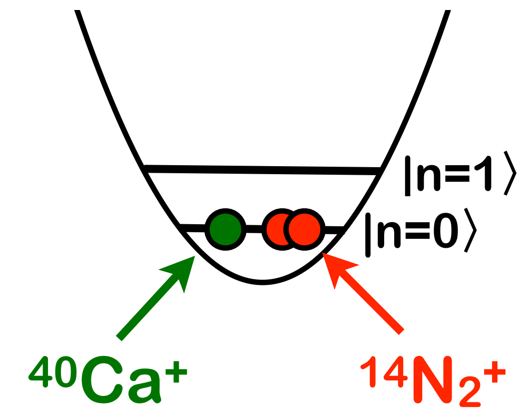


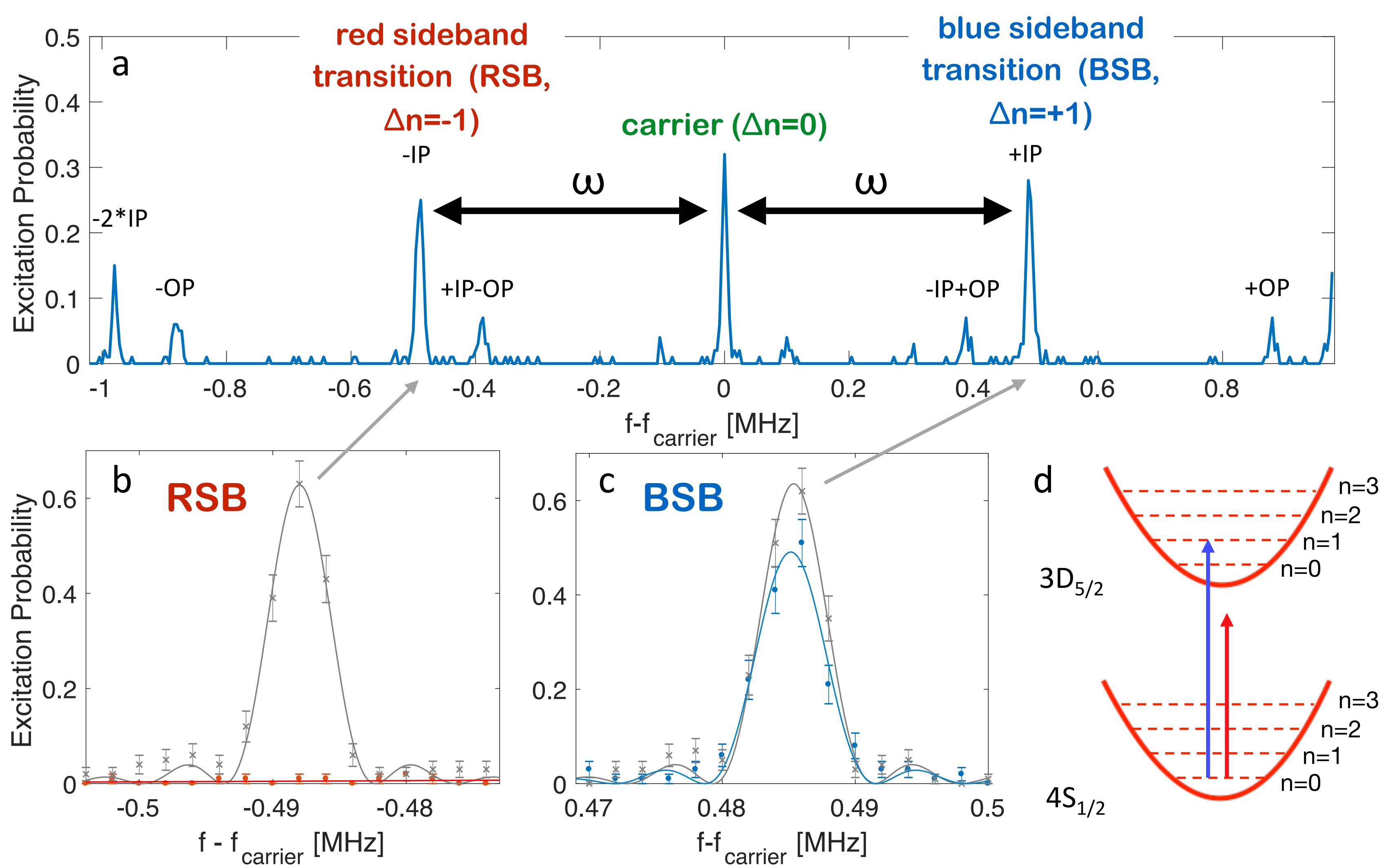
Conditions for resolved sideband cooling:

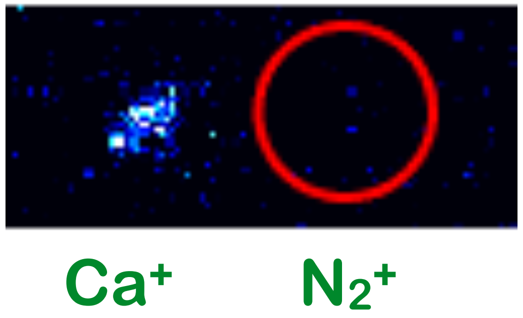
- Motional amplitude must be smaller than the wavelength of the cooling laser (Lamb-Dicke regime) ⇒ Doppler pre-cooling
- Bandwidth of cooling laser must be small enough to resolve sidebands

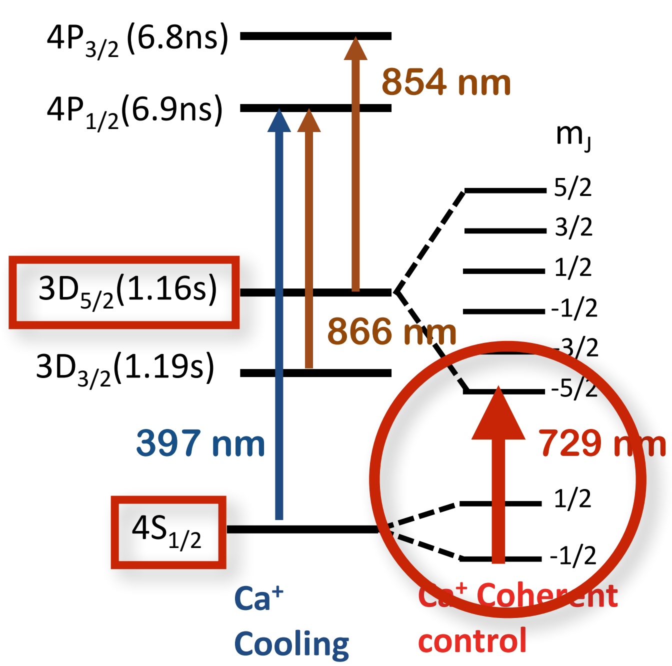
Lit.: D. Leibfried et al., Rev. Mod. Phys. 75 (2003), 281
H. Häffner et al., Phys. Rep. 469 (2008), 155

Example: Resolved-sideband cooling and spectroscopy of the Ca^+ 4p $^2S_{1/2} \rightarrow$ 3d $^2D_{5/2}$ transition at 729 nm in a Ca^+ - N_2^+ two-ion crystal





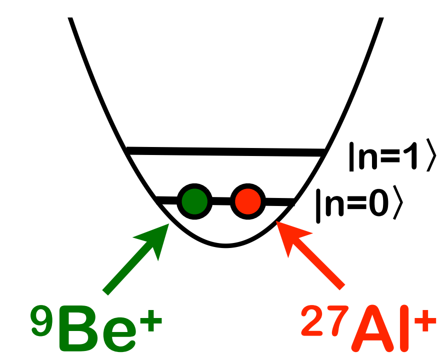




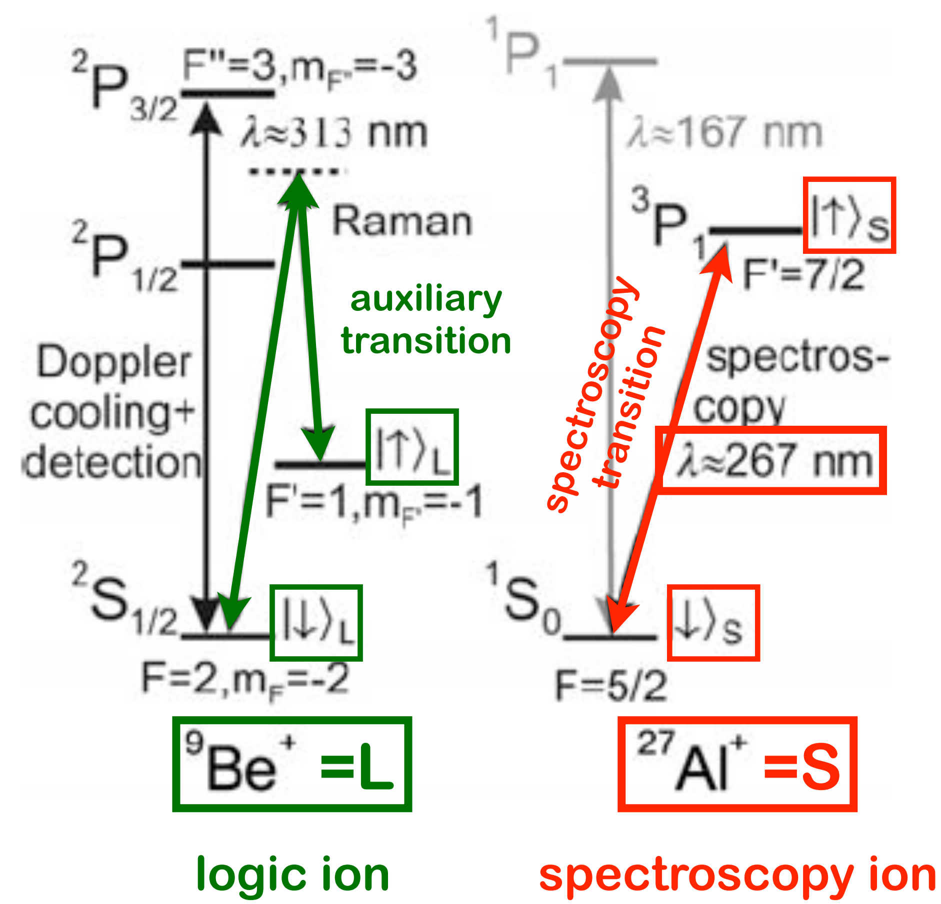
Ca+ spectroscopy

“Quantum-logic” spectroscopy of the $^1S_0 \rightarrow {}^3P_{1,0}$ transitions in $^{27}Al^+$

- The “spectroscopy ion” Al^+ does not have any suitable optical transitions for laser cooling. Hence it has to be sympathetically cooled by a simultaneously trapped Be^+ ion. Be^+ is cooled during the experiment.

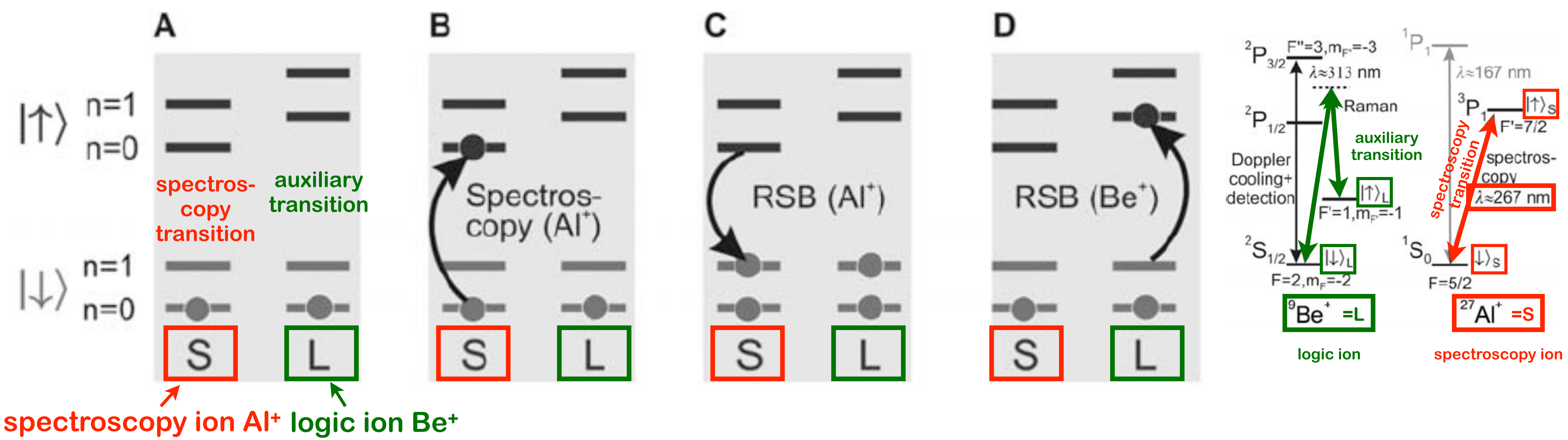


- The combined motion of the Be^+/Al^+ two-ion system is cooled to the motional ground state |n>=0 by sideband cooling on Be^+.
- The spectral transition of interest is the $^1S_0 \rightarrow {}^3P_1$ resonance in the “spectroscopy ion” Al^+. As Al^+ is not laser cooled itself, electron-shelving spectroscopy cannot be applied. Instead, the optical excitation in Al^+ is detected on the “logic ion” Be^+ using a quantum-logic protocol.



P.O. Schmidt et al., Science 309 (2005), 749

- **Quantum-logic protocol** for detecting an optical transition in Al^+:
  - A. Both ions are initialised in the **combined motional ground** state by resolved-sideband cooling on Be^+
  - B. The spectroscopy transition in Al^+ is excited ...
  - C. ... followed by a red-sideband transition (RSB) to n=1 in the $^1S_0$ state. Since the motion of the ions is coupled, BOTH ions are transferred into the n=1 motional state by the RSB.
  - D. The logic ion Be^+ is excited in another RSB from n=1 to n=0 in a metastable state. The optical cycle on Be^+ is disrupted and fluorescence stops.

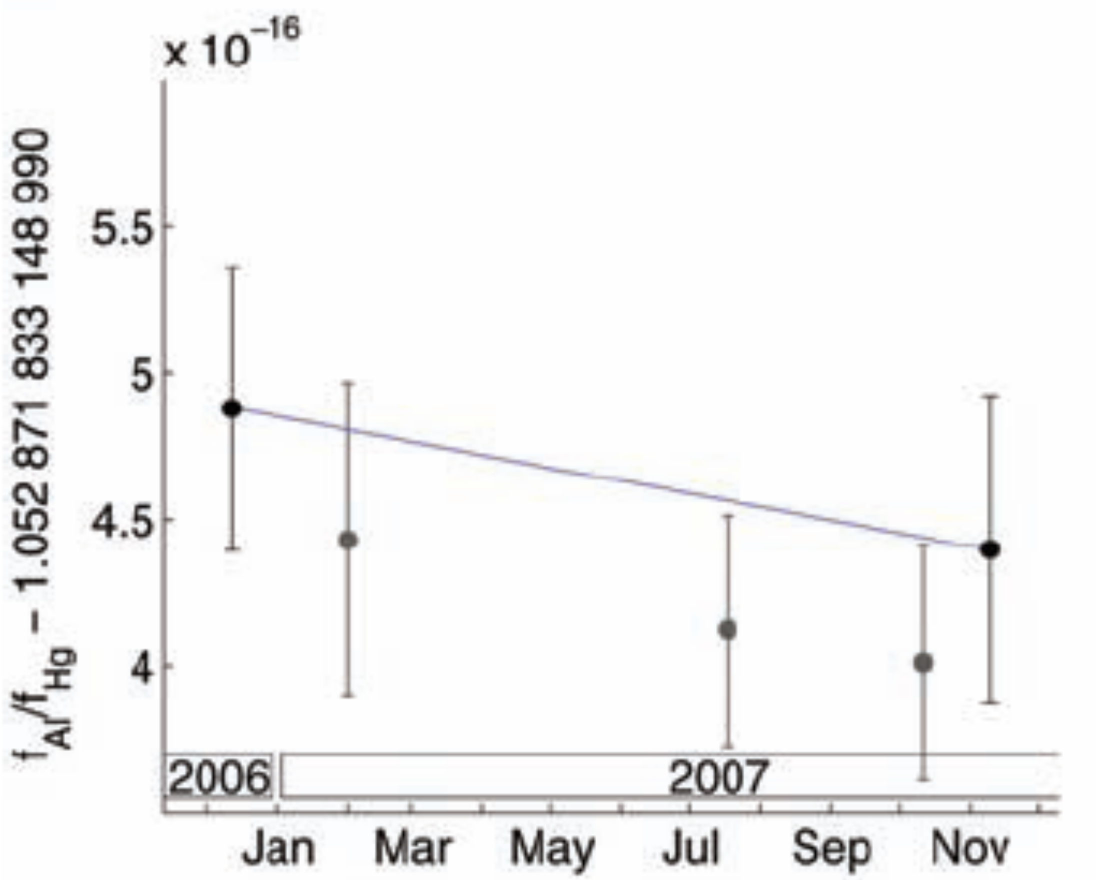


- The **fluorescence of Be^+ after step D ONLY breaks down after Al^+ has been excited beforehand in step B** → detection of a spectroscopic excitation in Al^+ by readout on Be^+.

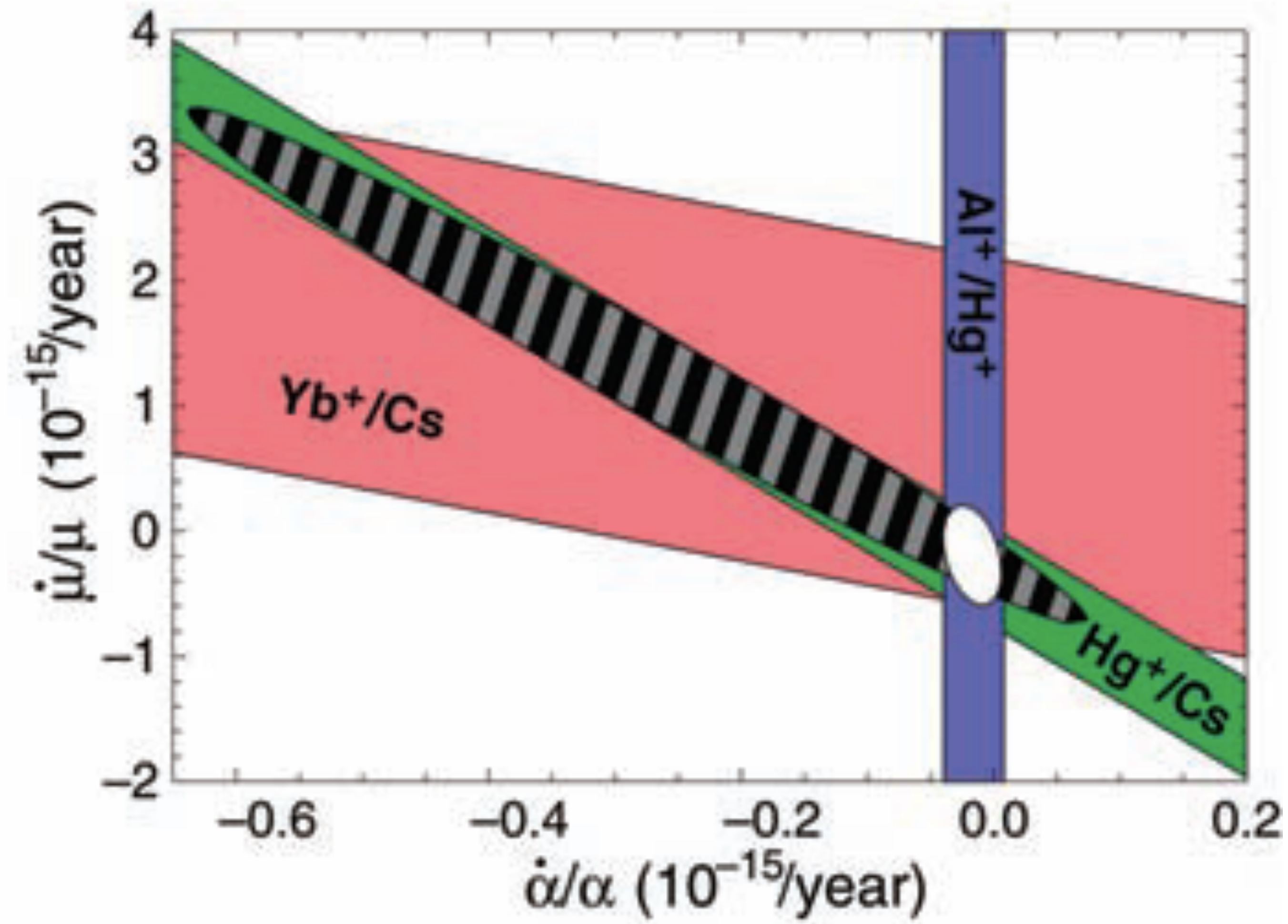
P.O. Schmidt et al., Science 309 (2005), 749

- **Application: measurement of the time constancy of the fine-structure constant α using the NIST Al^+/Hg^+ frequency standard.**

Variation of the frequency ratio f_{Al+}/f_{Hg+} as a function of time



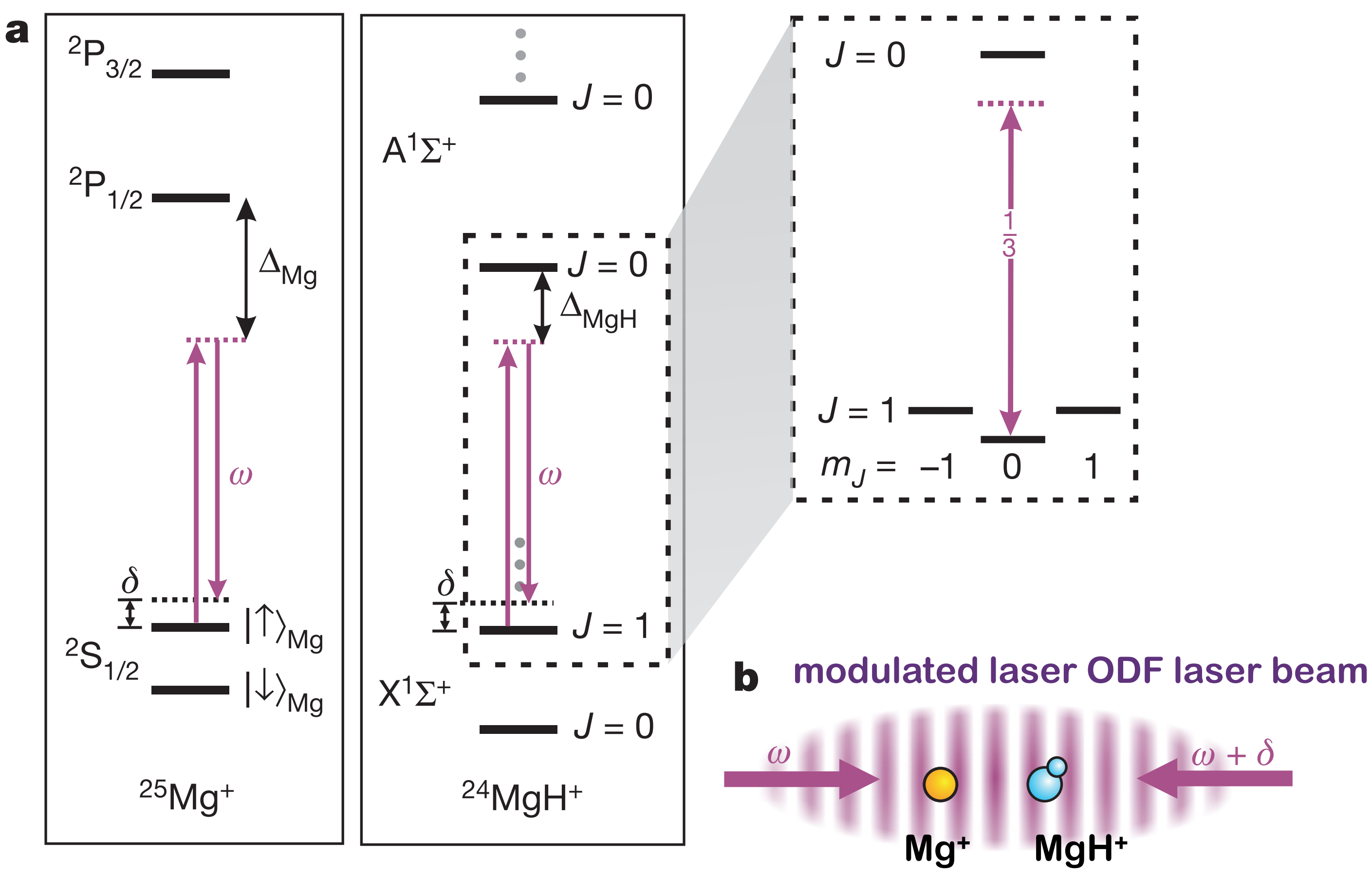
Corresponding variation of α and comparison with other frequency standards



T. Rosenband et al., Science 319 (2008), 1808

Quantum logic spectroscopy of cold molecular ions

- **Spectroscopy of a single MgH^+ ion by entanglement with and readout on a single Mg^+ ions using a state-dependent optical-dipole force (ODF) acting on a motional qubit of the two-ion string:**



(a) Energy level scheme of Mg^+ and MgH^+ and (b) schematic of the experiment

- **Implementation of a motional qubit:**

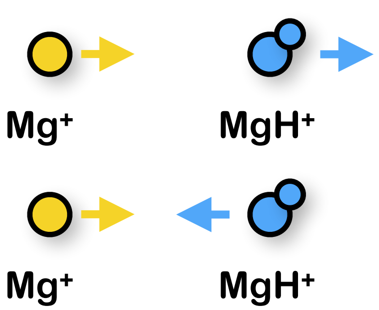
$$|\downarrow\rangle_m = |1\rangle_{ip}|0\rangle_{op}$$

$$|\uparrow\rangle_m = |0\rangle_{ip}|1\rangle_{op}$$

in-phase mode **out-of-phase mode**

in-phase mode:

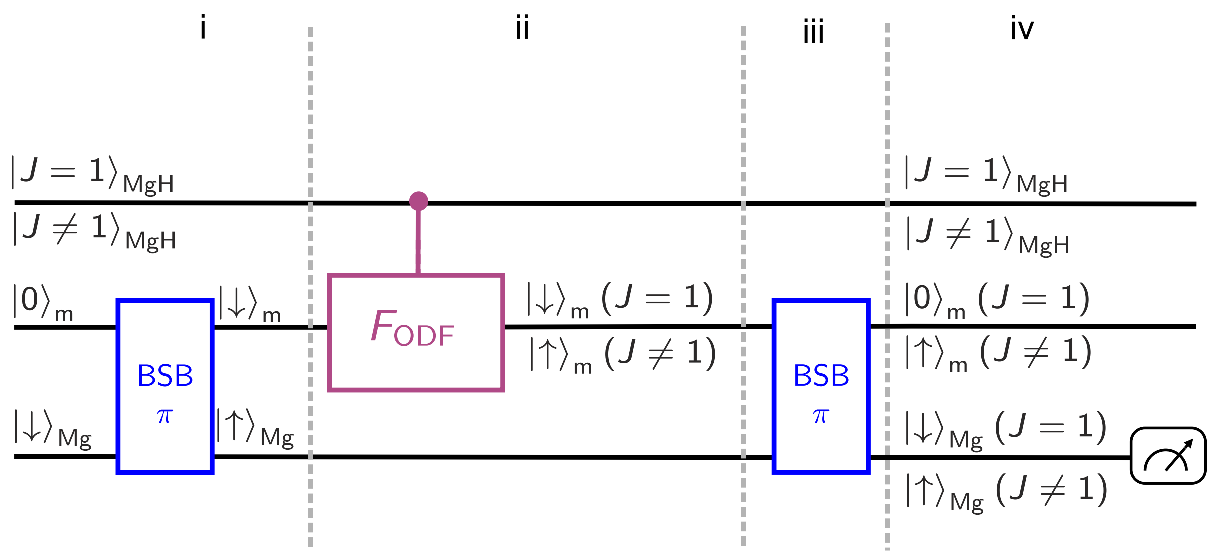
out-of-phase mode:

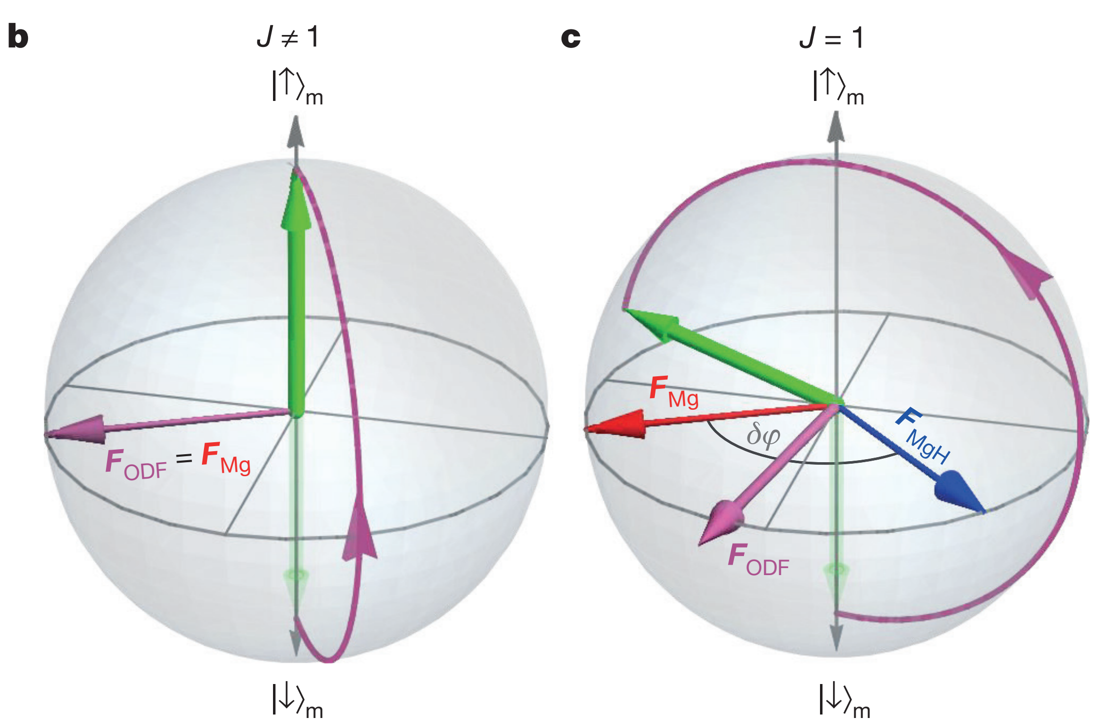


- **the ODF is modulated at $\delta = \omega_{op} - \omega_{ip}$ resonantly coupling the two motional qubit states**

F. Wolf, Y. Wan, J.C. Heip, F. Gebert, C. Shi and P.O. Schmidt, Nature 530 (2016), 457

Principle of experiment:

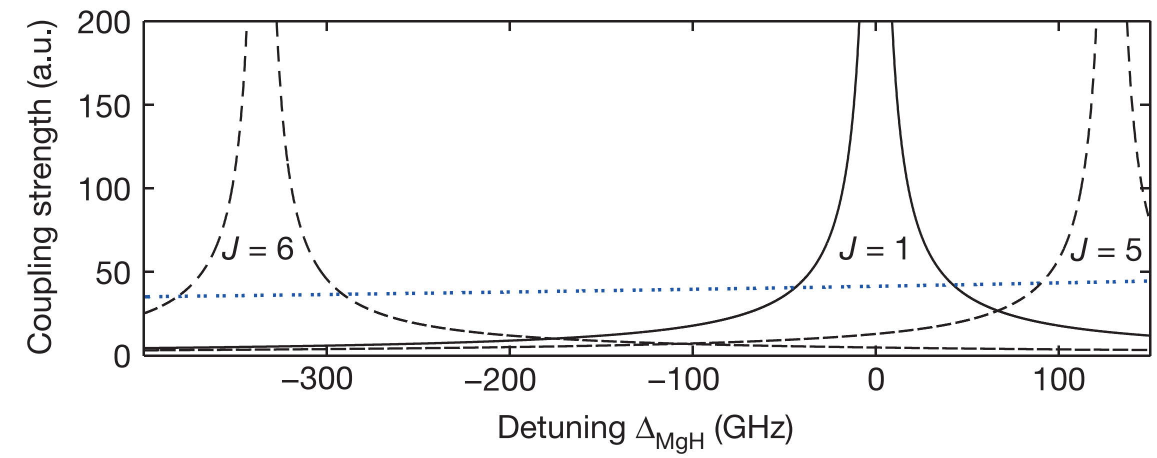




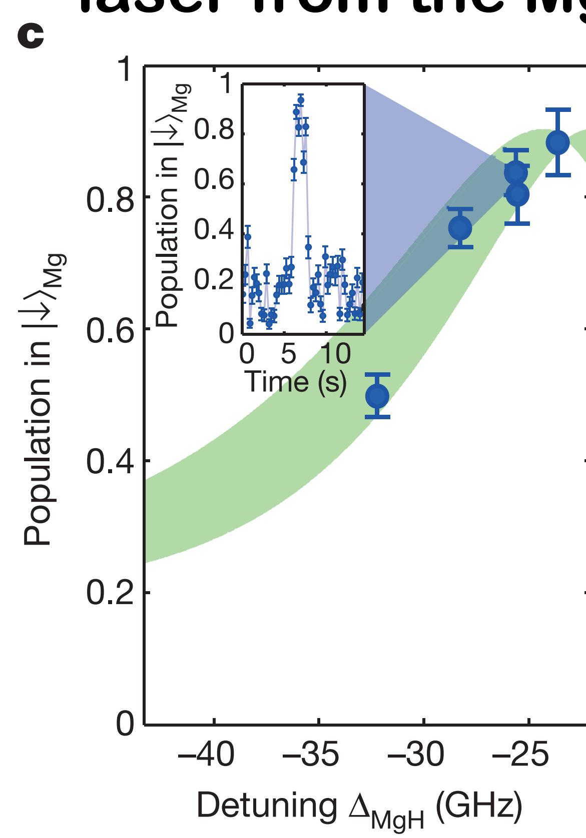
π-pulse on motional qubit w/o ODF on MgH^+ (ODF F_{ODF} acts only on Mg^+)

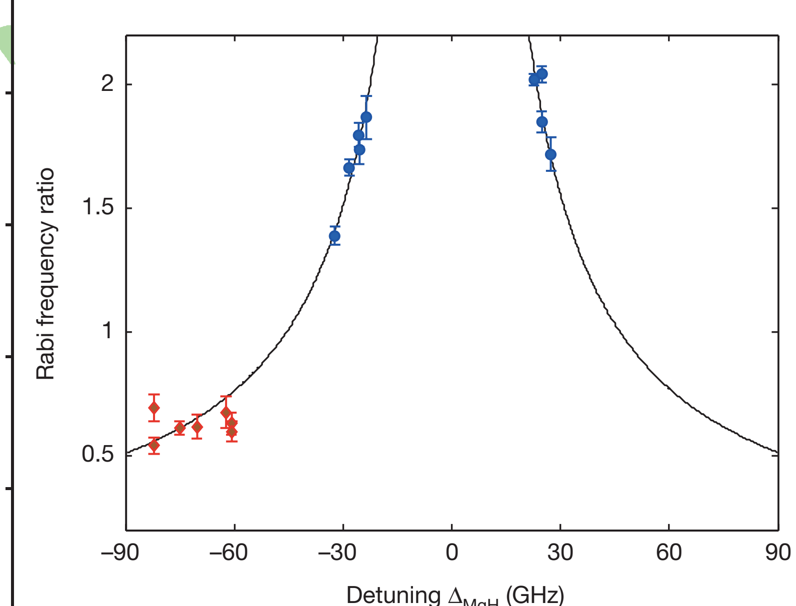
non-π-pulse on motional qubit with ODF on MgH^+ (ODF acts on both ions)

Coupling strength of the ODF as a function of the detuning from MgH^+ resonances:

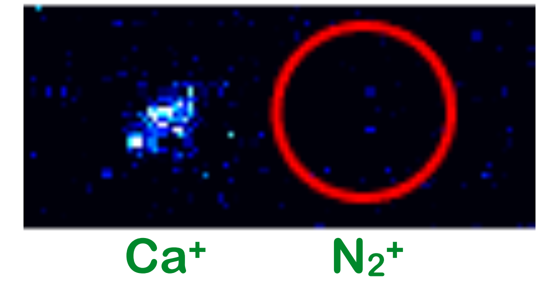


Population in lower motional qubit state as a function of the detuning of the ODF laser from the MgH^+ resonance:

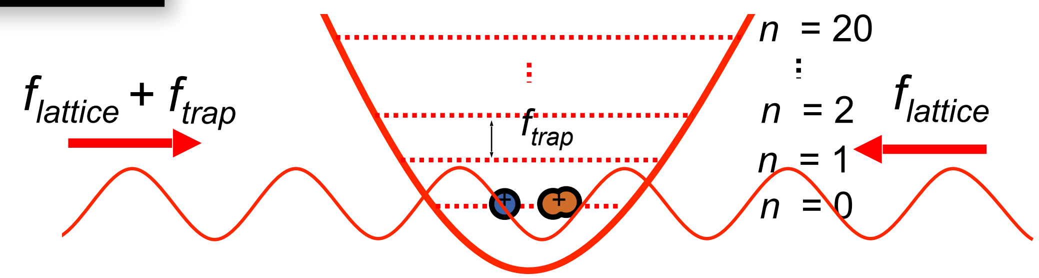




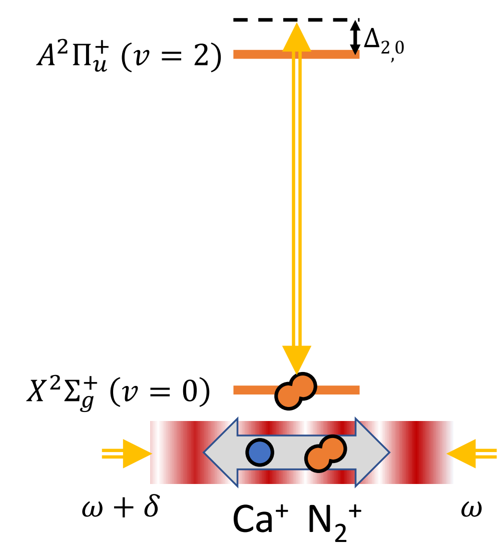
Quantum-non-demolition state detection of single molecular ions by coherent motional excitation of a two-ion crystal

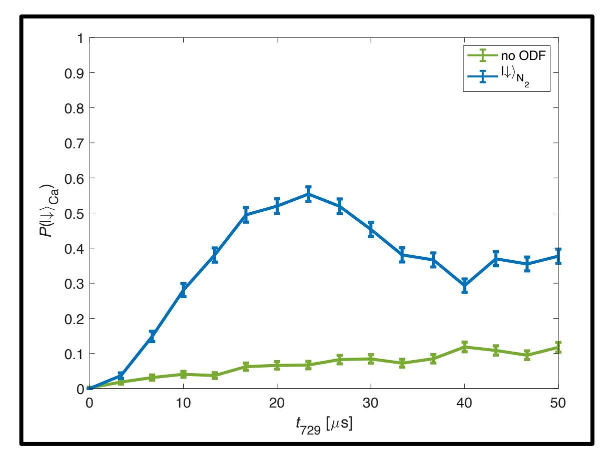


Non-destructive state-detection and spectroscopy of a single molecule



- Step 1: Preparation of a Ca^+ - N_2^+ two-ion string
- Step 2: Sympathetic cooling of the molecule to the QM ground state of the trap
- Step 3: Application of an 1D optical lattice near-resonant with a spectroscopic transition in the molecule to generate an optical dipole force
- Step 4: Modulation of the optical lattice at the frequency of vibration of the ions in the trap to excite their motion
- Step 5: Detection of the motional excitation of the ions by sideband Rabi thermometry on Ca^+

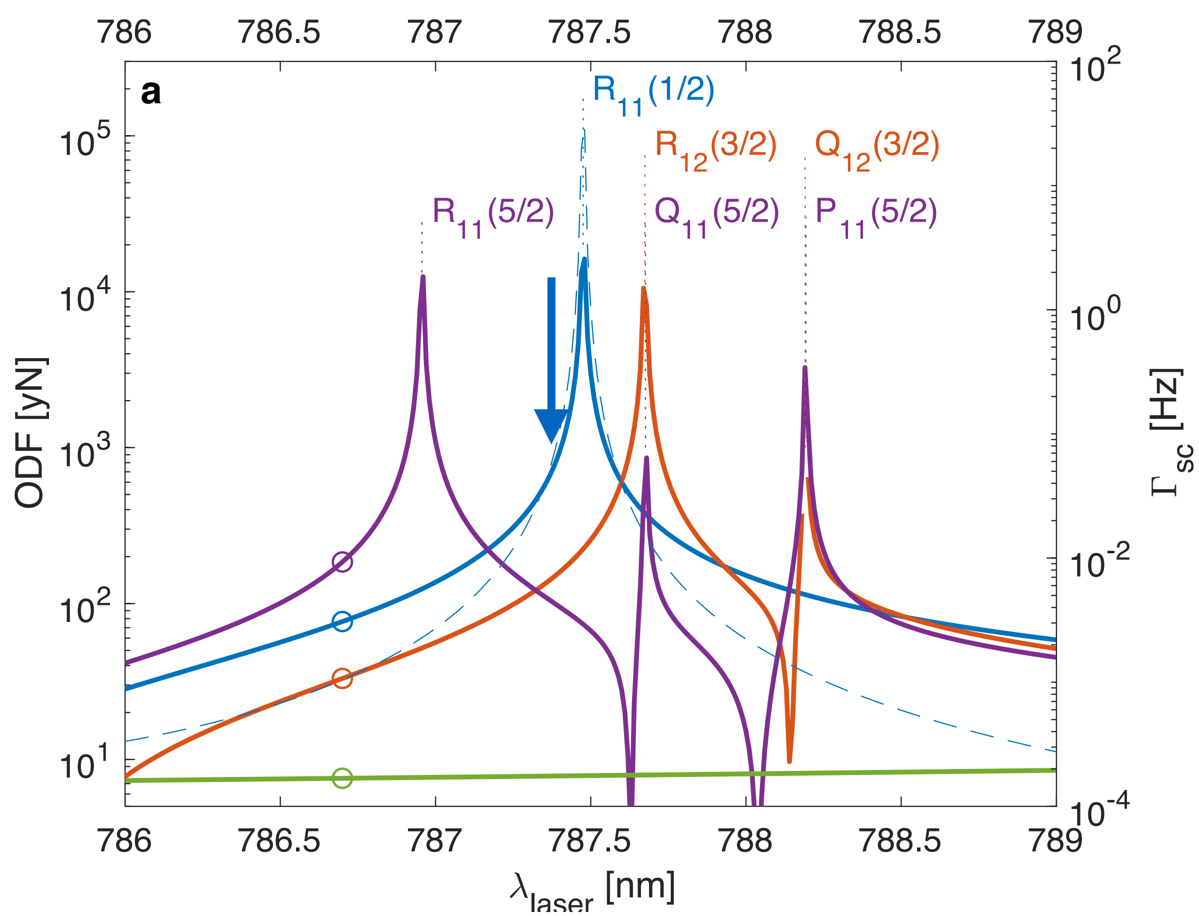




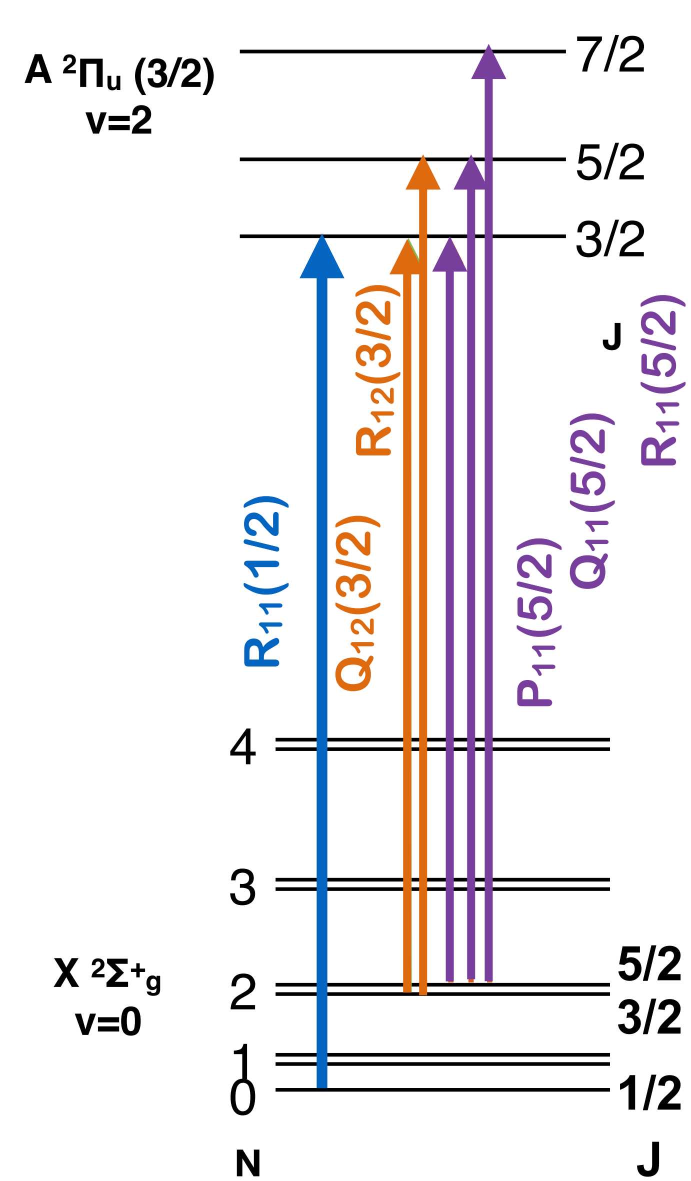
M. Sinhal et al., Science 367 (2020), 1213

ODF for N_2^+ in different spin-rotational states

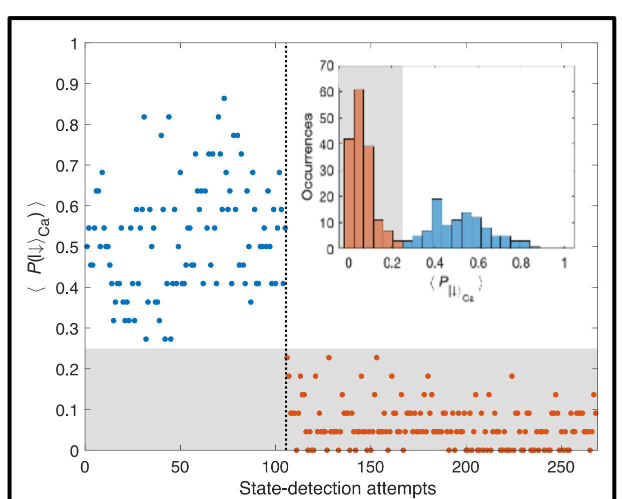
- ODF as a function of lattice-laser wavelength



- Excerpt of energy-level structure of N_2^+ with spectroscopic transitions giving rise to strong ODFs



- State-detection fidelity:



99.1(9)% 99.4(6)%

M. Sinhal et al., Science 367 (2020), 1213

Related experiments on molecular-ion quantum technologies

- Preparation and coherent manipulation of pure quantum states of a single molecular ion
  C.-w. Chou et al., Nature 545 (2017), 203
- Quantum entanglement between an atom and a molecule
  Y. Lin et al., Nature 581 (2020), 273
- Frequency-comb spectroscopy on pure quantum states of a single molecular ion
  C.-w. Chou et al., Science 367 (2020), 1458