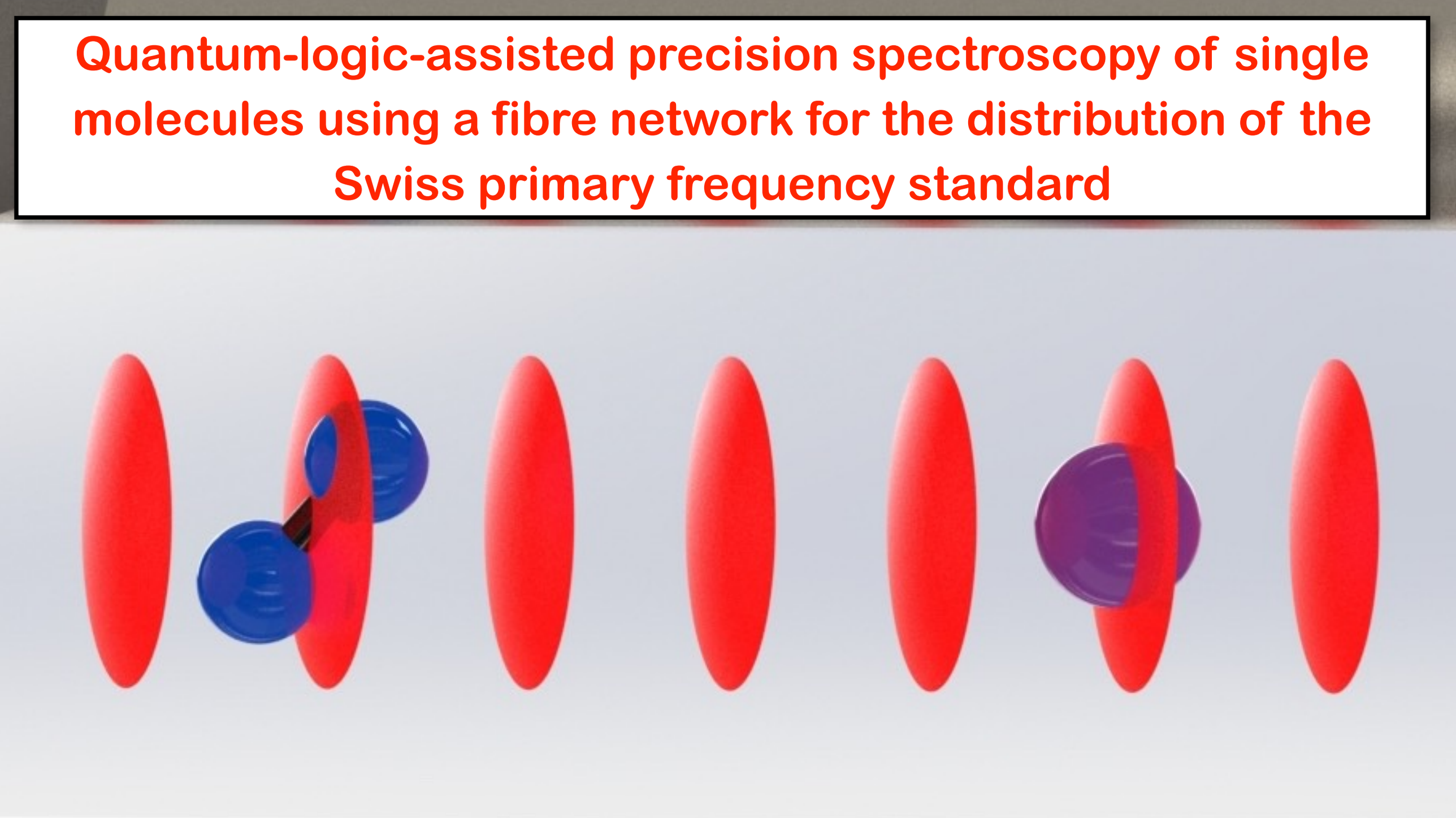
Quantum-logic-assisted precision spectroscopy of single molecules using a fibre network for the distribution of the Swiss primary frequency standard



Quantum Photonics Workshop
Basel, Oct. 28, 2021

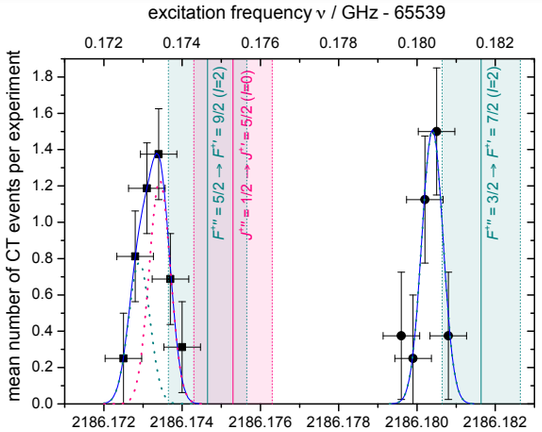
Stefan Willitsch
Department of Chemistry
University of Basel, Switzerland

University of Basel

Quantum-logic-assisted precision spectroscopy of single molecules

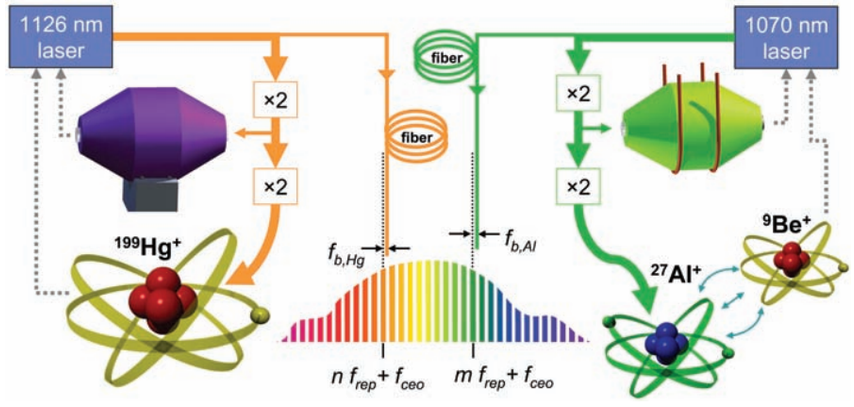
Motivation: Quantum control of single trapped molecules

Precision spectroscopy



- M. Germann et al., Nature Physics 10 (2014), 820
- S. Schiller, V. Korobov, PRA 98, 022511 (2018)
- P. Jansen et al., PRL 115, 133202 (2015)

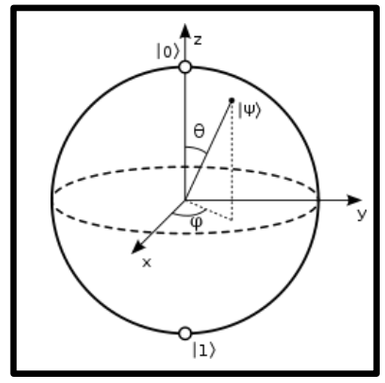
Molecular clocks



Science 319 (2008), 1808

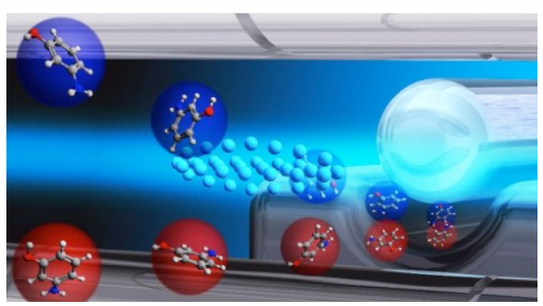
- M. Kajita, PRA 92, 043423 (2015)
- S. Schiller et. al, PRL 113, 023004 (2014)
- J.Ph. Karr, J. Mol. Spectr. 300 (2014), 37

Molecular qubits



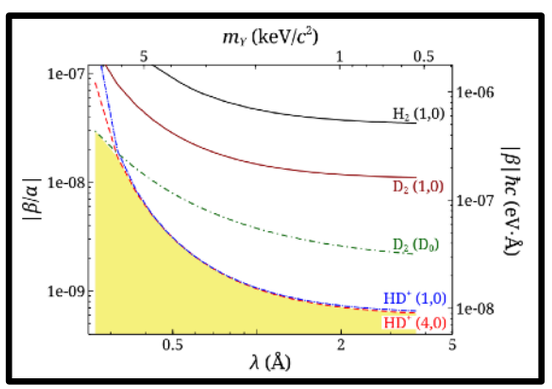
- J. Mur Petit et al., in "Advances in Atom and Molecule Machines", Springer 2012
- D. DeMille, PRL 88, 067901 (2002)

State- and energy controlled chemical reactions



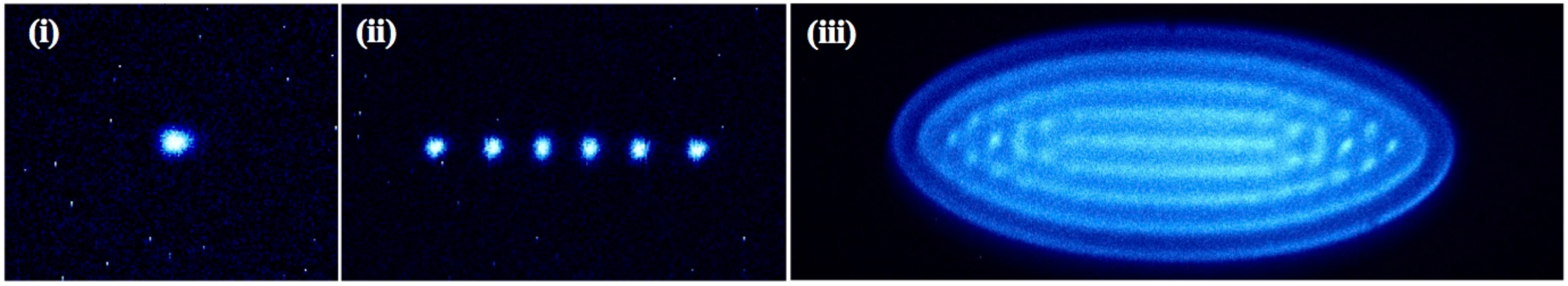
- A. Kilaj et al., Nat. Commun. 9, 2096 (2018)
- A.D. Dörfler et. al., Nat. Comm. 10, 5429 (2019)
- T. Sikorsky et. al., Nat. Comm. 9, 920 (2018)

Tests of fundamental physics



- E. Salumbides et. al., PRD 87, 112008 (2013)
- M. Safronova et. al., Rev. Mod. Phys. 90, 025008 (2018)
- H. Loh et al., Science 342, 1220 (2013)

Coulomb crystallisation of cold ions in traps



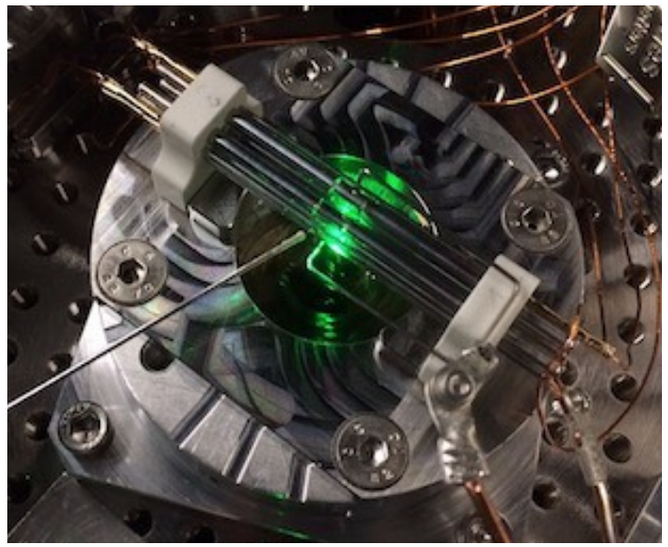
Fluorescence images of Coulomb crystals of laser-cooled Ca^+ ions in an ion trap

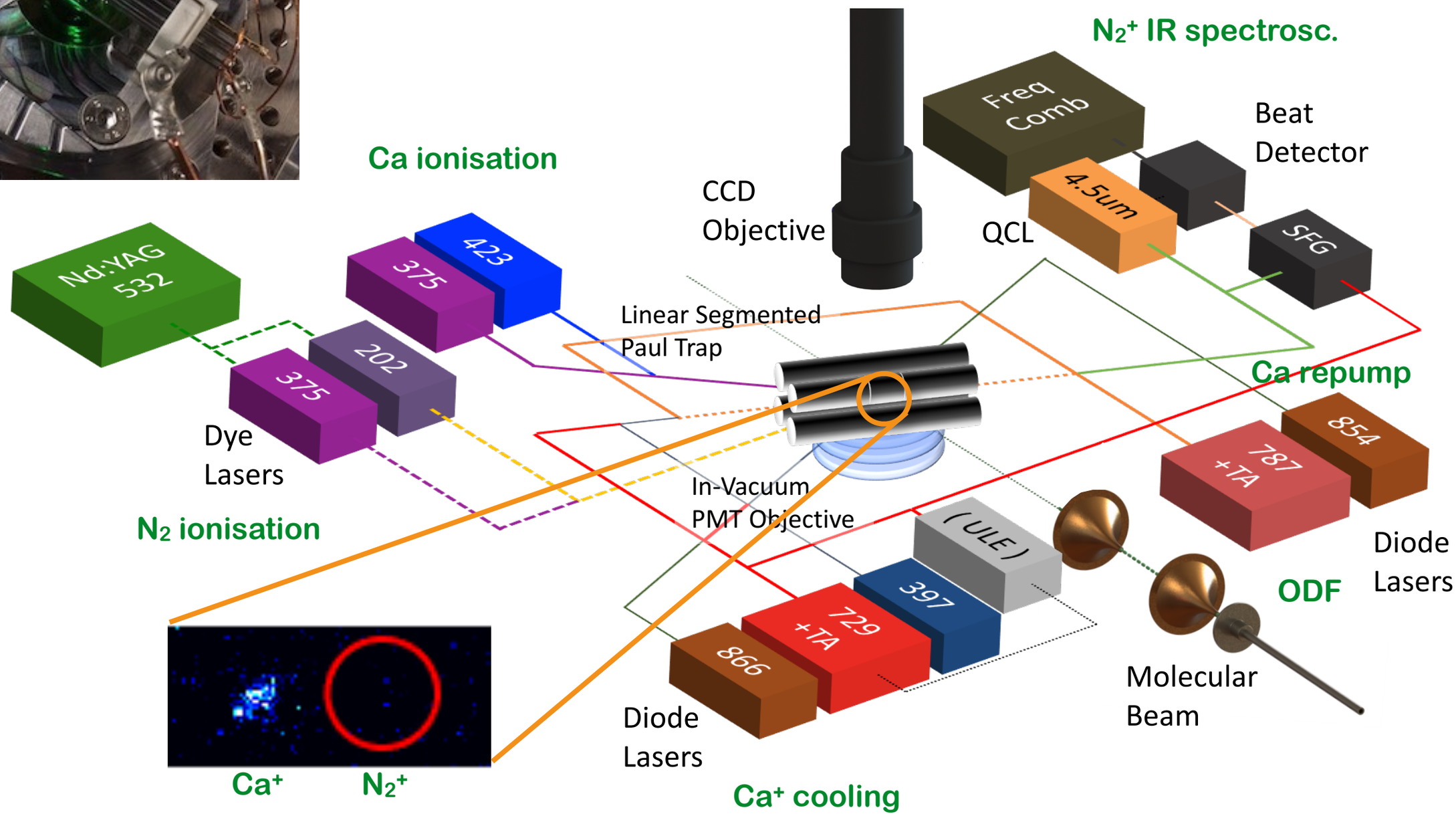
Properties of Coulomb-crystallised ions:

- Translationally cold (μK-mK)
- Long trapping times (> hrs)
- Extremely well controlled experimental environment
- Observe, address and manipulate single particles

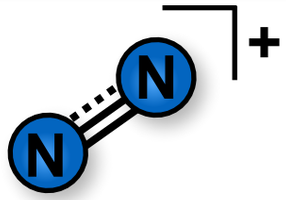
Lit.:
- D. Leibfried et al., Rev. Mod. Phys. 75 (2003), 281
- H. Häffner et al., Phys. Rep. 469 (2008), 155
- S. Willitsch, Int. Rev. Phys. Chem. 30 (2012), 175

Connecting two worlds: ion traps and molecular beams



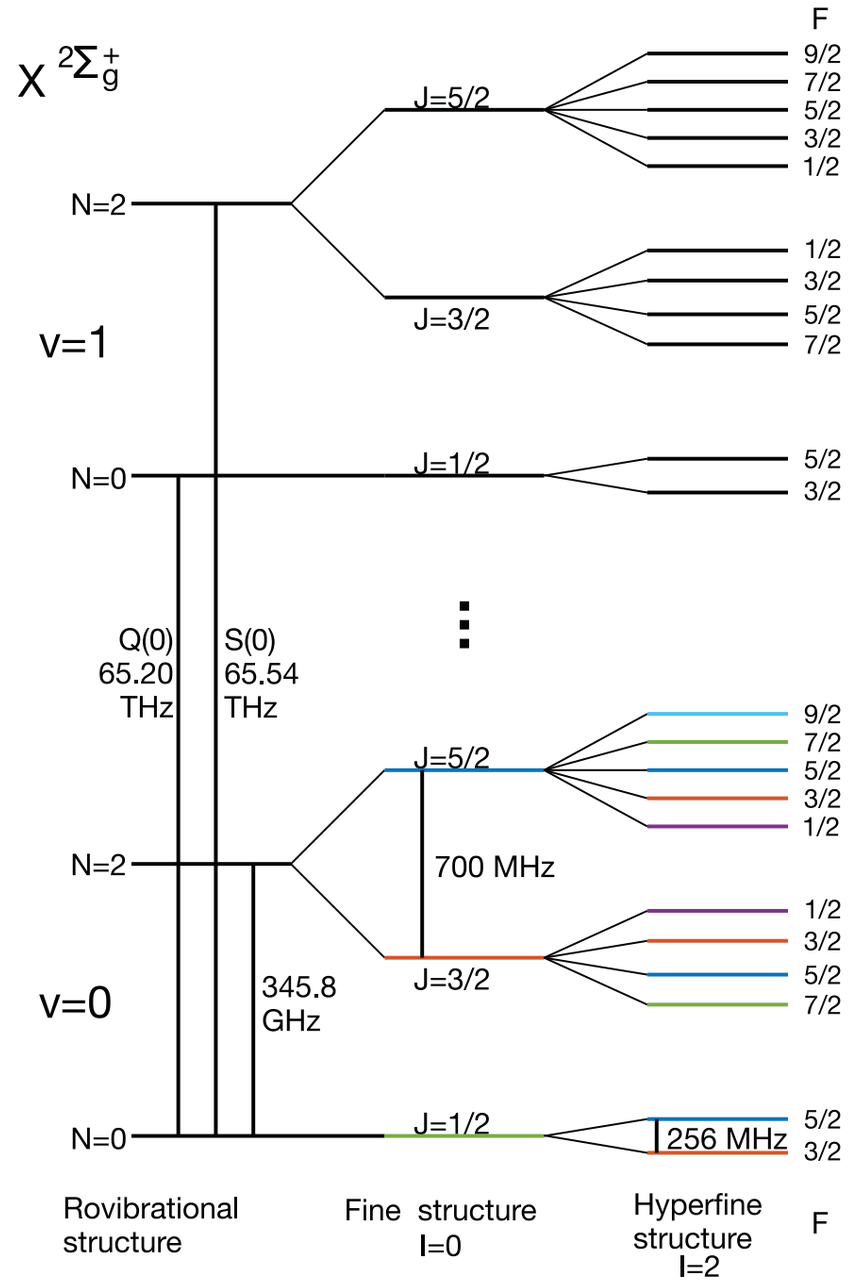


The N_2^+ molecular ion



Symmetric homonuclear molecule:

- No permanent dipole moment
- E1-forbidden rotational-vibrational transitions: narrow E2- and M1-allowed spectral lines
- Very small systematic shifts on rotational and vibrational spectroscopic transitions
- No redistribution of state populations due to black-body radiation
- Two nuclear spin isomers in the rotational ground state with I=0 and 2



K. Najafian et al., Phys. Chem. Chem. Phys. 22 (2020), 23083

A new approach to state detection and spectroscopy for single molecules: QND state readout using coherent motional excitation (CME) on a single molecular ion

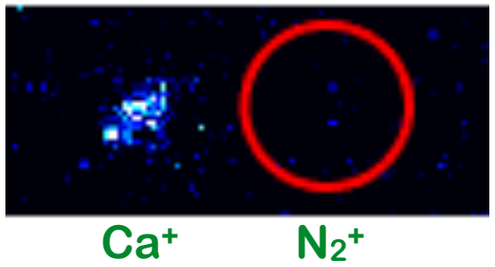
- Advantages:
  - Improvement of duty cycle by up to 5 orders of magnitude
  - Removal of ensemble averaging
- Inspired by previous work on atomic ions by

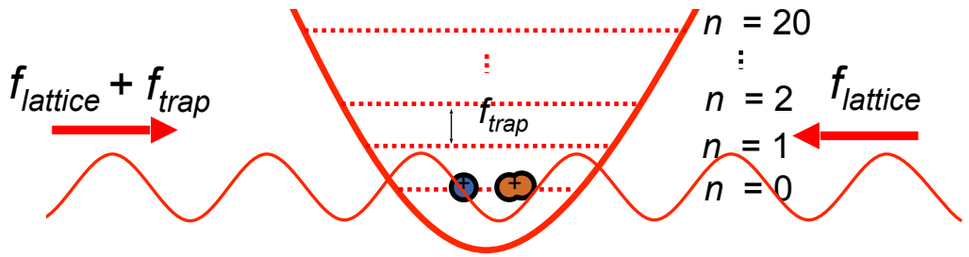
  D. Hume et al., Phys. Rev. Lett. 107 (2011), 243902
- See related work by:
  - F. Wolf et al., Nature 530 (2016), 457
  - C.-w. Chou et al., Nature 545 (2017), 203
  - E. Clausen et al., arXiv 2005.00529

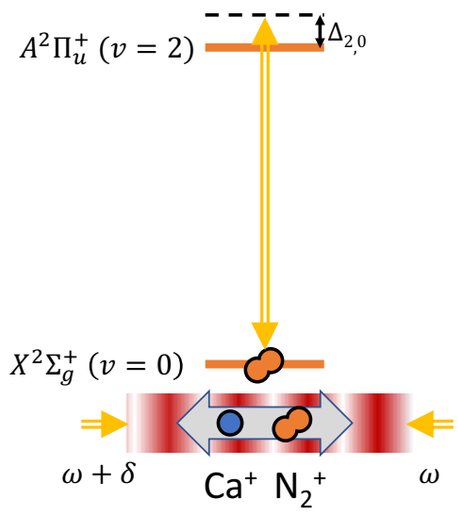
Z. Meir et al., Faraday Discuss. 217 (2019), 561
M. Sinhal et al., Science 367 (2020), 1213
K. Najafian et al., Nat. Commun. 11 (2020), 4470

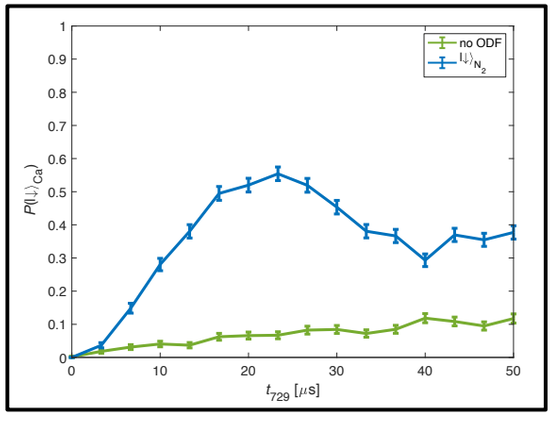
State-dependent coherent motional excitation

- 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^+



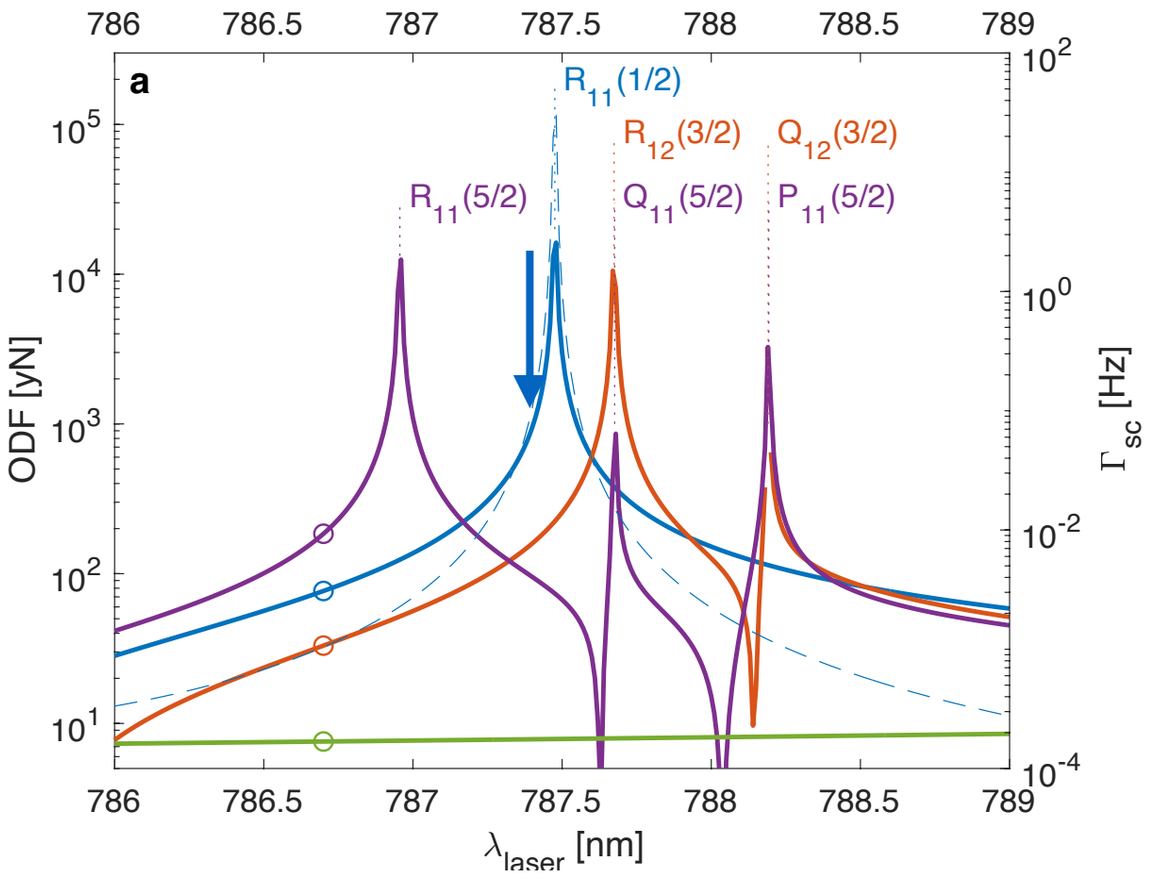


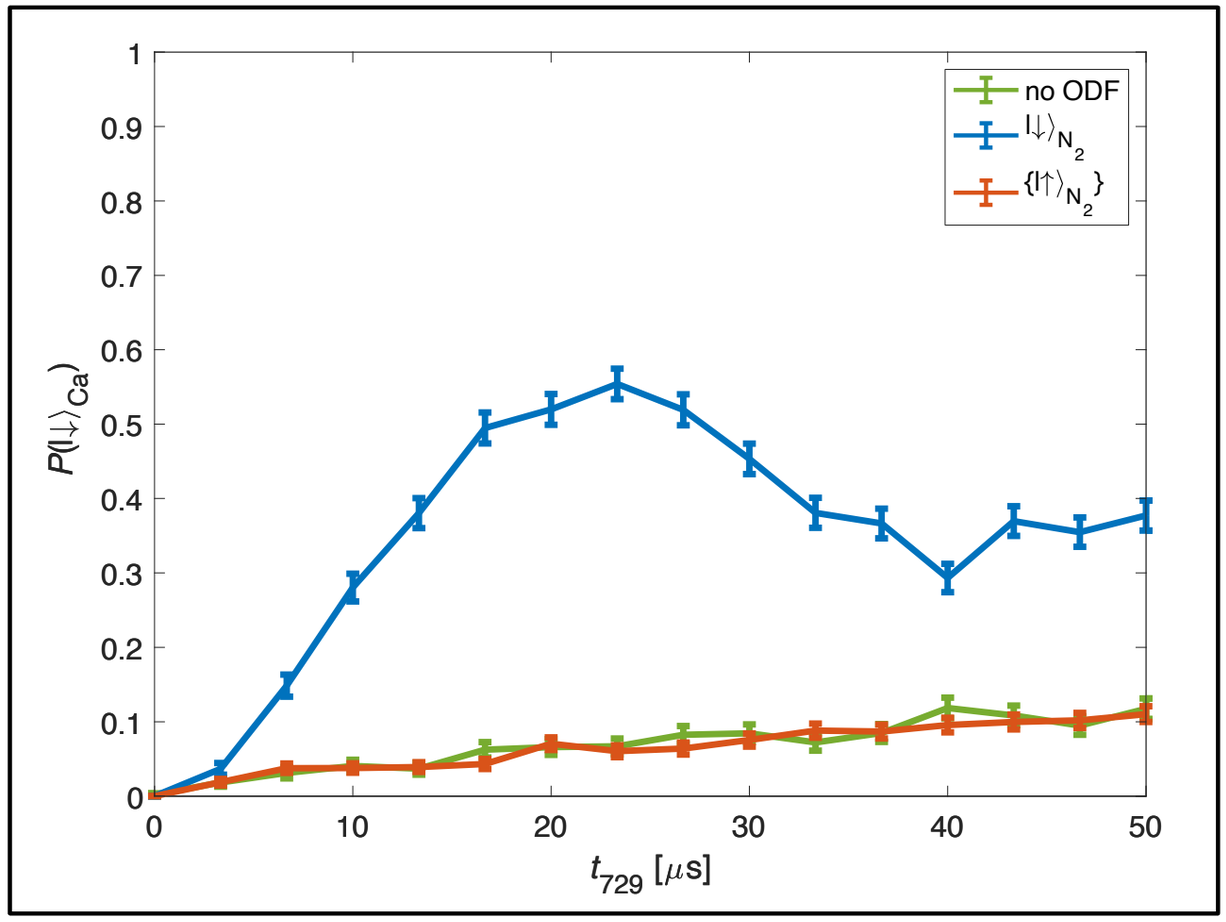


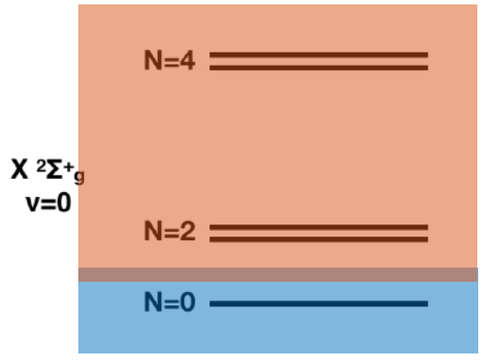


Z. Meir et al., Faraday Discuss. 217 (2019), 561
M. Sinhal et al., Science 367 (2020), 1213
K. Najafian et al., Nat. Commun. 11 (2020), 4470

Readout of CME using Rabi flops on a Ca^+ $^2S_{1/2} \leftarrow {}^2D_{5/2}$ sideband transition



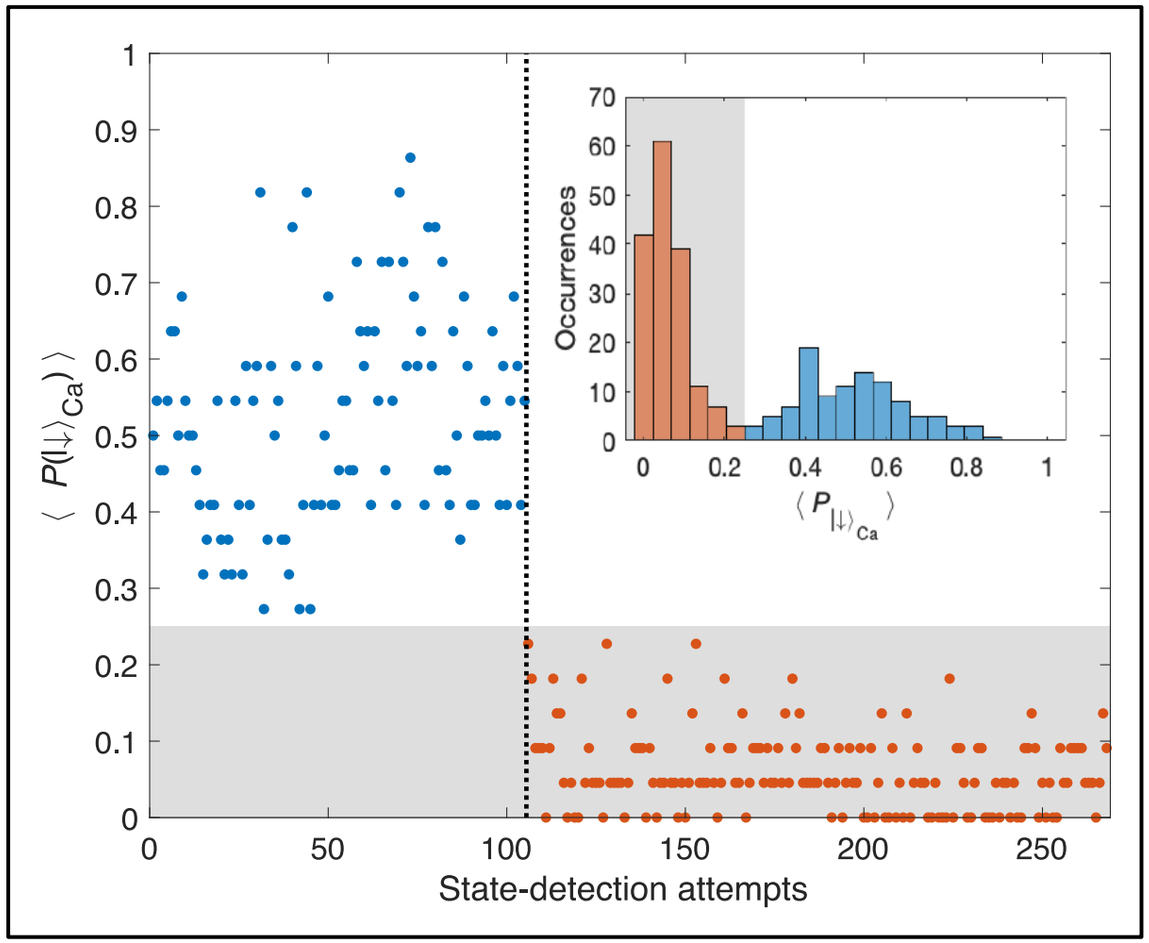




M. Sinhal et al., Science 367 (2020), 1213

Quantum-logic-assisted precision spectroscopy of single molecules

State-detection fidelity

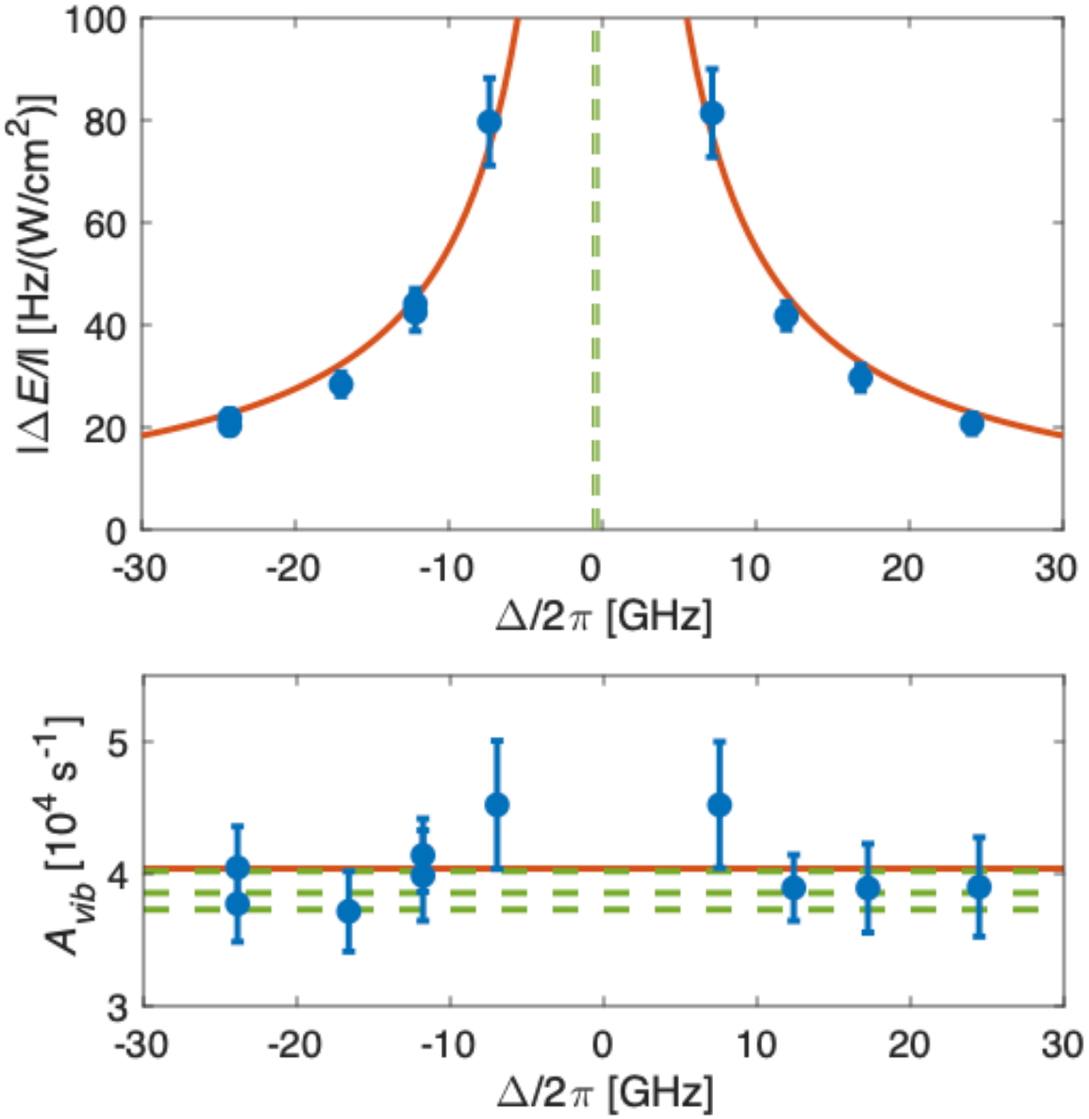


99.1(9)% **99.4(6)%**

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

Force spectroscopy on a single trapped molecule

- Mapping out the AC Stark shift around the $R_{11}(J=1/2)$ transition in X (v=0) -> A (v=2) band of N_2^+:



- Measurement of electronic transition:
  f = 380.7011(2) THz
  f = 380.7007(3) THz (literature)

- Einstein A coefficients:
  A = 3.98(11) x 10^4 s^{-1}
  A = 3.87(14) x 10^4 s^{-1} (literature)

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

Precision spectroscopy using a network for the distribution of the Swiss primary frequency standard

The Swiss primary frequency and time standard: continuous Cs fountain FoCS-2 at METAS (Berne)

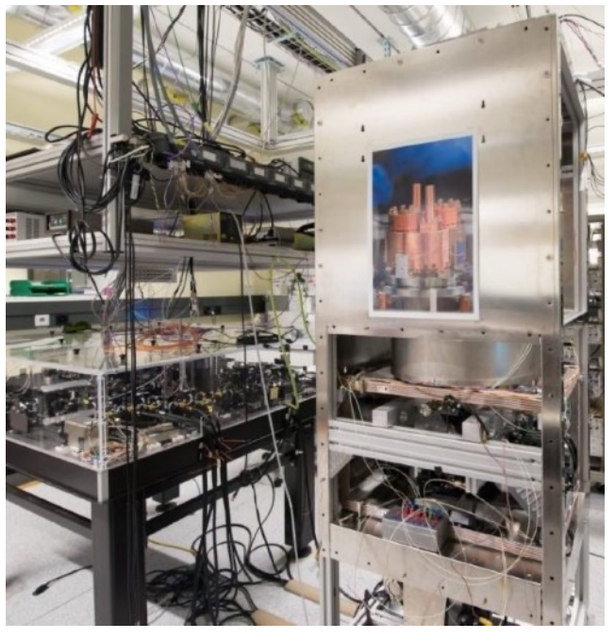
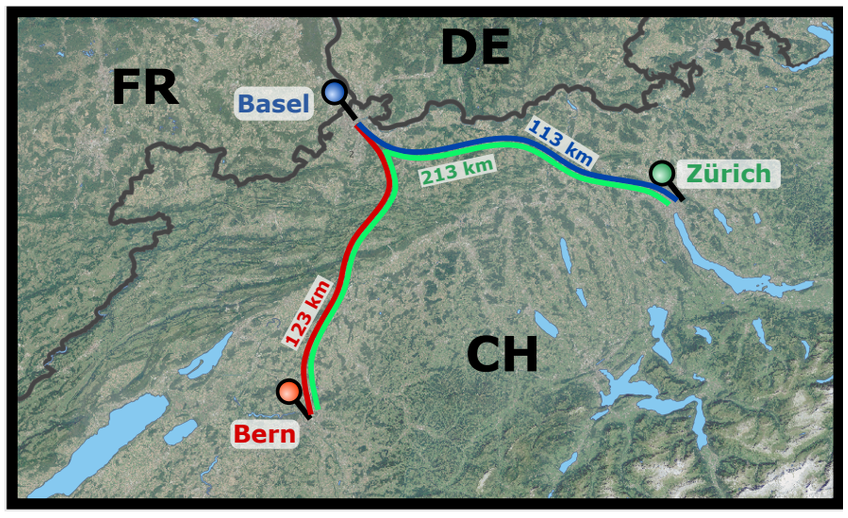


Image © METAS

- Frequency uncertainty: 2×10^{-15}
- Contributor to TAI

A. Jallageas et al., Metrologia 55 (2018), 366

Current extent of network:



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Stefan Willitsch

ETH Zürich

Frédéric Merkt

Jérôme Faist

METAS

Jacques Morel

SWITCH

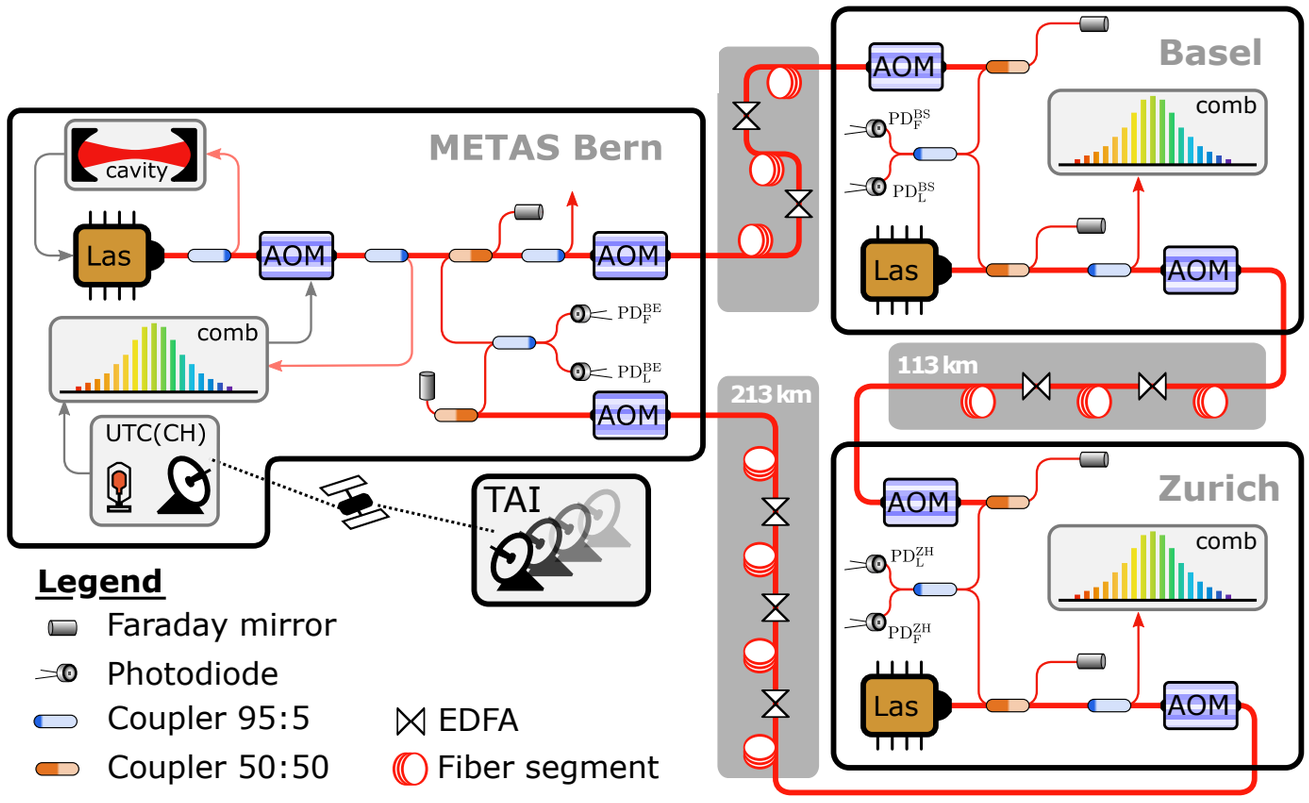
Ernst Heiri

Fabian Mauchle

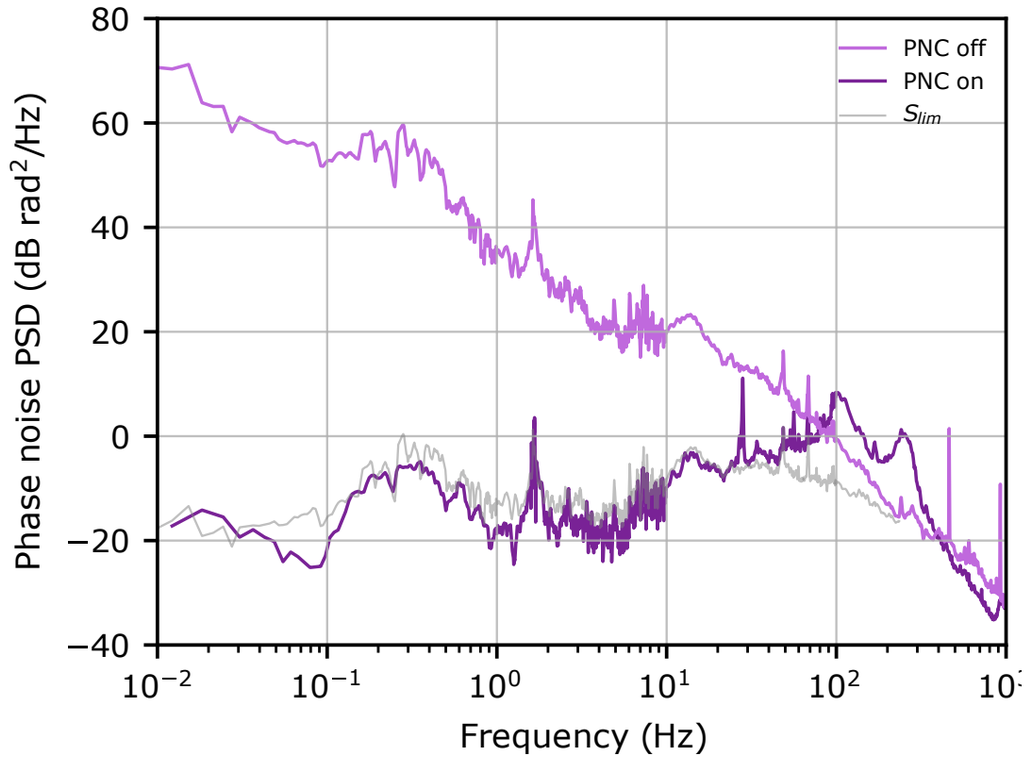
University of Basel

Quantum-logic-assisted precision spectroscopy of single molecules

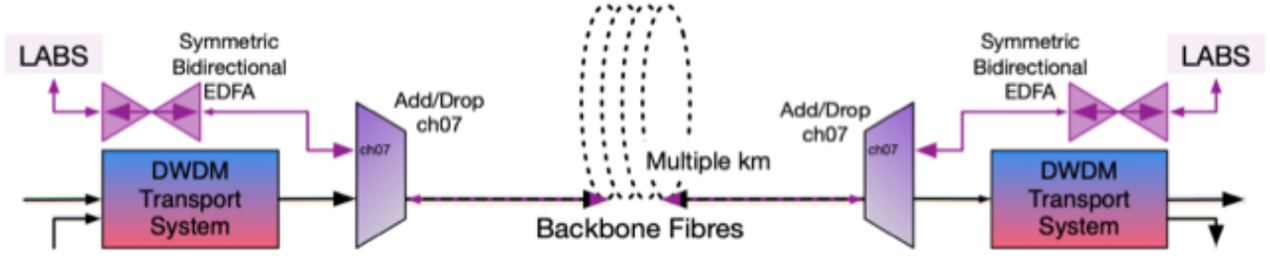
Optical layout of the frequency transfer



Phase-noise cancellation (PNC) of the transferred signal at 1572 nm



Using a dark channel on the SWITCH optical-fibre network in the telecom L-band at 1572 nm for the frequency transfer

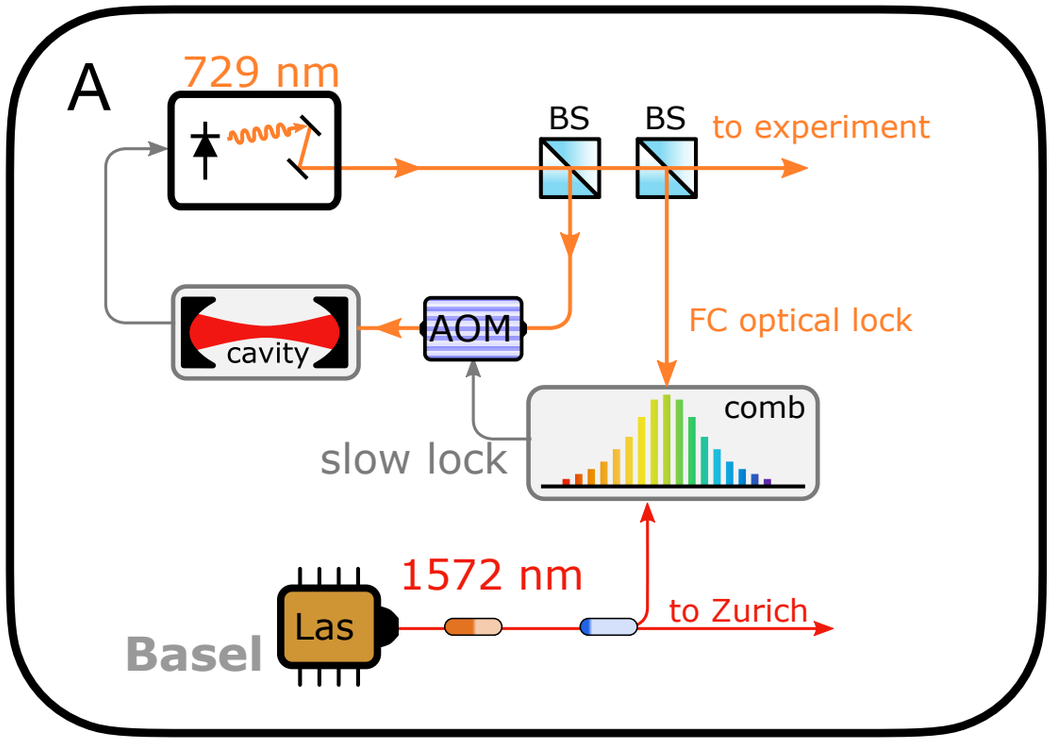


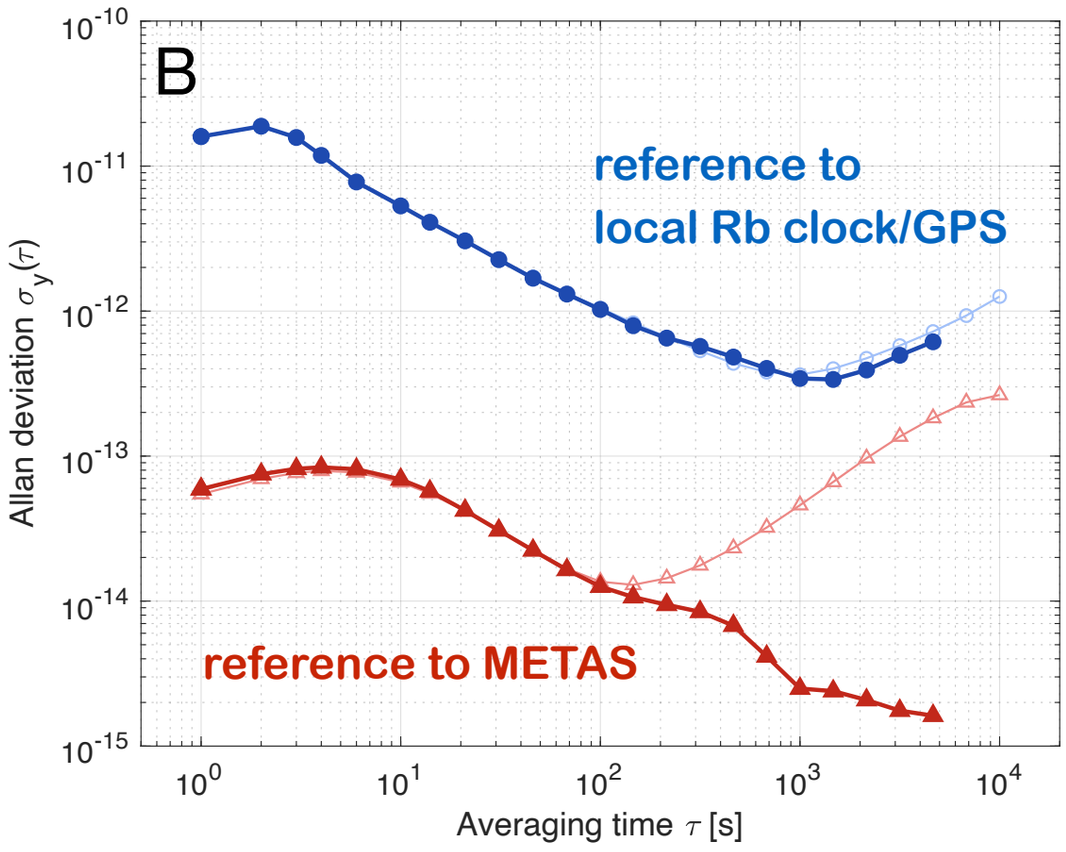
D. Husmann et al., Opt. Expr. 29 (2021), 24592

D. Husmann, METAS

Jacques Morel, METAS

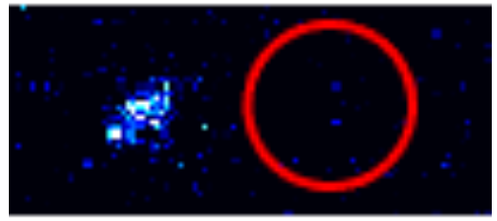
Stabilisation of the 729 nm master laser at Basel to the METAS standard



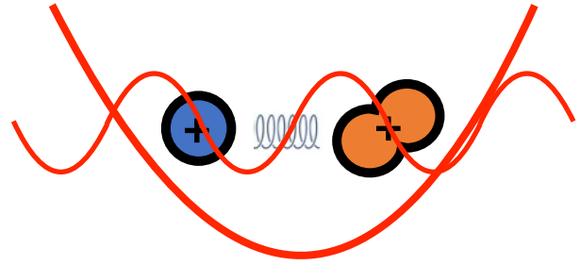


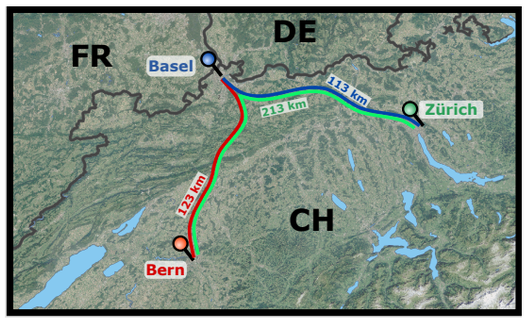
Summary and conclusions

- A new method for the non-destructive detection of molecular quantum states
- Quantum non-demolition and therefore highly sensitive
- A new approach to molecular-ion spectroscopy
- Applications in precision spectroscopy, state-to-state chemistry and quantum technologies



Ca^+ N_2^+





Acknowledgements

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Aleksandr Shlykov

Mudit Sinhal

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Jérôme Faist

Jacques Morel

D. Husmann

Ernst Heiri

Fabian Mauchle

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Schweizerischer Nationalfonds
Fondo nazionale svizzero
Swiss National Science Foundation

SNI Swiss Nanoscience Institute
Eine Initiative der Universität Basel und des Kantons Aargau

QSIT Quantum Science and Technology

QCI

Universität Basel