Precision infrared molecular spectroscopy as an instrument for probing variations of fundamental constants

Aleksandr Shlykov

Searching for the New Physics
at the Quantum Technology Frontier

Ascona
04.07.2023

Cold and Controlled
Molecules and Ions Group

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The Standard Model… and beyond

- The Discovery of the Higgs boson at the LHC experimentally proved the last theoretical prediction of the Standard Model
- String theory and other multidimensional theories
- Grand Unification Theories (supersymmetry)
- Chameleon model


5% of SM matter [1]
(26% Dark matter, 69% Dark energy)

$$\alpha = \frac{1}{4\pi\epsilon_0}\frac{e^2}{\hbar c}$$

$$\mu = \frac{m_p}{m_e}$$

[1] Adam, R., et al. (Planck Collaboration), 2016, "Planck 2015 results," Astron. Astrophys. 594, A1.
[2] J.-P. Uzan. Varying constants, gravitation and cosmology. Living Reviews in Relativity, 14:2, 2011.

Picture: symmetrymagazine.org

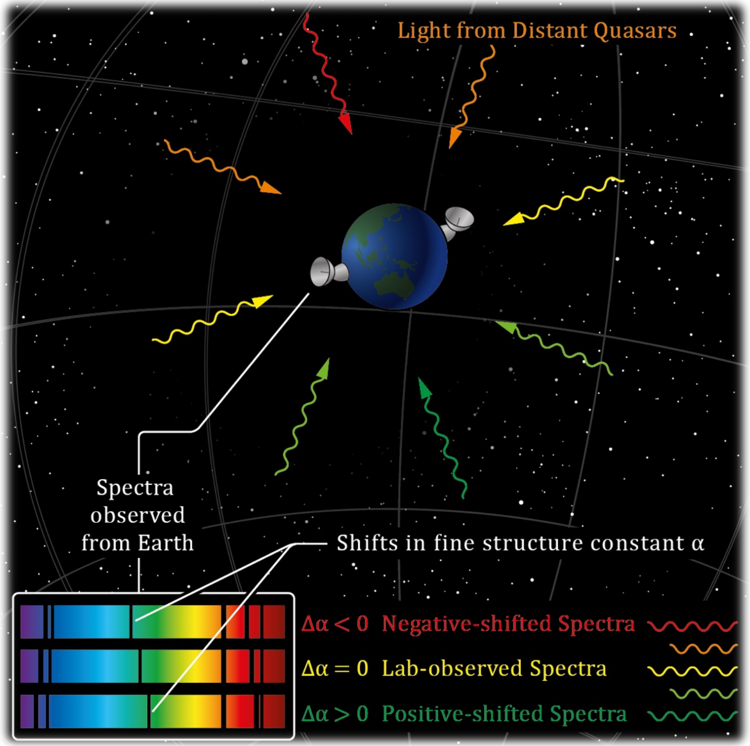
Quasar absorption spectra

- Results from quasar absorption spectra:

$\frac{\dot{\alpha}}{\alpha} = 0.01(0.15)\times 10^{-5}\ \mathrm{yr}^{-1}$ [1] (VLT/UVES Chile, 21 system in the southern hemisphere, $0.4<z<2.3$)

$\frac{\dot{\alpha}}{\alpha} = -0.57(0.11)\times 10^{-5}\ \mathrm{yr}^{-1}$[2] (Keck/HIRES Hawaii, 143 system in the northern hemisphere, $0.2<z<4.2$)

$\frac{\dot{\mu}}{\mu} = 0.26(0.3)\times 10^{-6}\ \mathrm{yr}^{-1}$[3] (VLT/UVES, 3 systems, $2.59<z<3.02$)



[1] Srianand, R., Chand, H., Petitjean, P. and Aracil, B., Phys. Rev. Lett., 99, 239002, 2007.
[2] M. T. Murphy, V. V. Flambaum, J. K. Webb, V. V. Dzuba, J. X. Prochaska, and A. M. Wolfe. Springer Berlin Heidelberg, Berlin, Heidelberg, 2004.
[3] King, J.A., Webb, J.K., Murphy, M.T. and Carswell, R.F., Phys. Rev. Lett., 101, 251304, 2008.

Optical and microwave atomic clocks

- $E \sim R_\infty A_{elec} F(\alpha)$ – optical clock
- $E_{hfs} \sim R_\infty A_{hfs} g_i \mu \alpha^2 F_{hfs}(\alpha)$ - microwave clock

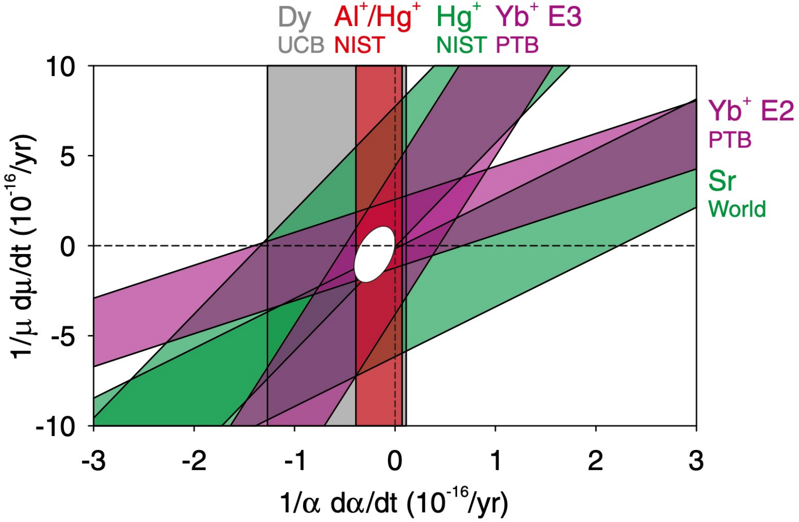
- Strongest constraint for the present day (PTB [2]):

$$\frac{\dot\alpha}{\alpha} = 1.0(1.1)\times10^{-18}\,\mathrm{yr}^{-1}$$ Yb⁺E3/E2

$$\frac{\dot\mu}{\mu} = -0.8(3.6)\times10^{-17}\,\mathrm{yr}^{-1}$$ Yb⁺E3/Cs

- The dependence of g_i on $\frac{m_q}{\Lambda_{QCD}}$ variation is model dependent[3]

Constraints on temporal variations of α and μ from comparison of atomic clocks [1]



[1] N. Huntemann, B. Lipphardt, C. Tamm, V. Gerginov, S. Weyers, and E. Peik. Phys. Rev. Lett., 113:210802, 2014.
[2] R. Lange, N. Huntemann, J. M. Rahm, C. Sanner, H. Shao, B. Lipphardt, C. Tamm, S. Weyers, and E. Peik. Phys. Rev. Lett., 126(1):011102, 2021.
[3] F. Luo, K. A. Olive, and J.-P. Uzan. Phys. Rev. D, 84:096004, 2011.

Molecular clocks

- Rovibrational transitions in molecular clocks provide direct sensitivity to μ

- $E_{rovib} \sim \omega_e \left(v + \frac{1}{2}\right) - \omega_e x_e \left(v + \frac{1}{2}\right)^2 + B_e J(J+1)$

$$\omega_e \propto 1/\sqrt{\mu} \qquad \omega_e x_e \propto 1/\mu \qquad B_e \propto 1/\mu \qquad \omega_e \gg B_e$$

- Strongest constraint with molecules (KRb, Japan):

$$\frac{\dot{\mu}}{\mu} = -0.3(1.0)\times 10^{-14}\ \mathrm{yr}^{-1}$$ [1]

[1] J. Koboyashi, A. Ogino & S. Inouye, Nature Comm. 10, 3771, 2019.

	Absolut transition sensitivity	Properties	Group
KRb	-9.45 THz	**The current best limit,** near-degenerate pair of vibrational states provides enhanced sensitivity, microwave 644 MHz transition, sensitive to BBR	S. Inouye
O_2^+	-398 THz -323 THz (v=16<-v=0 E2)	Immunity to BBR, no hyperfine structure, small Stark and Zeeman for specific transitions, E2 can be driven with two-photon 767 nm	D. Hanneke P. Schmidt
Sr_2	-7.5 THz (Raman)	No hyperfine structure, low sensitivity to Zeeman shifts for some states, 25.1 THz Raman transition, AC-Stark shifts from the lattice limit uncertainty to 10^{-14}	T. Zelevinsky
H_2^+, HD^+	Precise m_p determination	Enables direct comparison to theory, HD+ sensitive to BBR	S. Schiller J. Koelemeij J-P. Karr L. Hilico D. Kienzler
N_2^+	**-32THz (v=1<-v=0 E2)**	**Immunity to BBR, small Stark and Zeeman shift for specific transitions, extremely narrow mid-IR E2 transition accessible with Quantum Cascade Laser**	**S. Willitsch** M. Keller

Based on: David Hanneke et all, Quantum Sci. Technol. 6, 014005, 2021.

Experiment with N_2^+

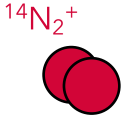
- Rovibrational transitions accessible with Quantum Cascade Lasers for direct interrogation (M1 and E2)

	Q(0) M1	S(0) E2
Blackbody shift (300 K)	4×10^{-18}	4×10^{-18}
DC Stark shift (cm/V^2)	8×10^{-20}	8×10^{-20}
Quadrupole shift (mm^2/V)	0	0 for specific HF states
Rel. Zeeman shift (G^{-1})	0	0 or 10^{-17} for Magic transitions
Probe laser AC Stark shift. (100 mW, 50 μm)	10^{-14}	10^{-14}

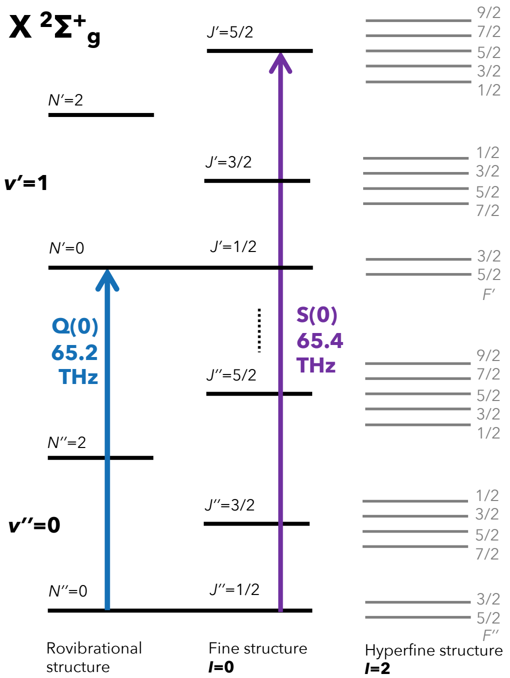
- Extremely narrow dipole-forbidden transitions in the N_2^+ ground state in mid-IR can be measured with 10^{-18} uncertainty

M. Kajita, G. Gopakumar, M. Abe, M. Hada, and M. Keller. Phys. Rev. A, 89:032509, 2014.

K. Najafian, Z. Meir and S. Willitsch, PCCP, 22, 23083, 2020.



N_2^+ ground state energy levels scheme



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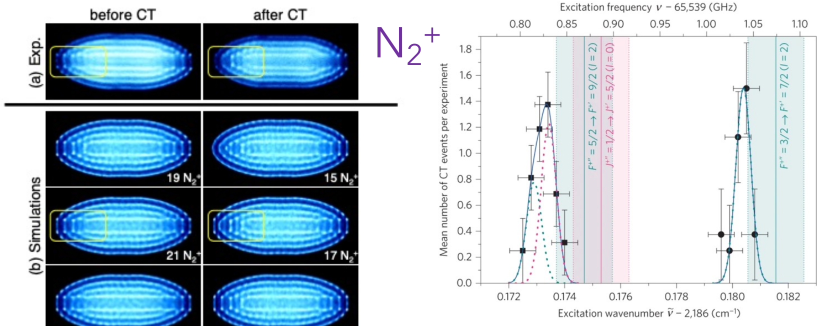
Problems with molecules

- Non-polar molecules can’t be cooled directly
- No fluorescence detection as in atoms
- Many previous experiments employed destructive state detection techniques

laser-induced charge transfer

photodissociation of ions

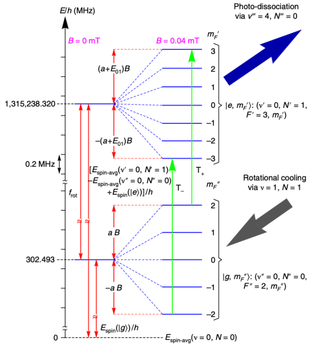
ionisation of neutrals



N_2^+

f=65540.039(12) GHz

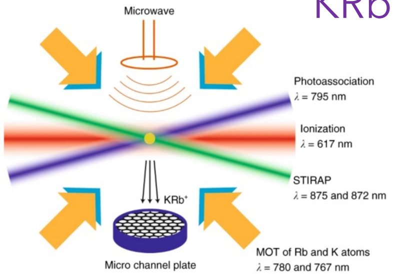
M. Germann, X. Tong and S. Willitsch, Nature Phys, 10, 8203, 2014.



HD^+

S. Alighanbari et. al., Nature Phys. 581, 152, 2020.

KRb



J. Koboyashi et. al., Nature Comm. 10, 3771, 2019.

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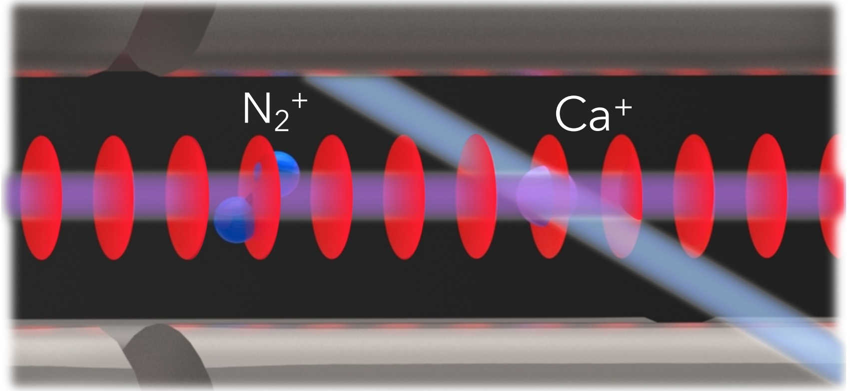
Quantum logic spectroscopy

- We trap a single N_2^+ molecular ion together with a Ca^+ ion
- Ca^+ serves for N_2^+ cooling and state detection
- Coherent optical dipole force maps the internal quantum state of N_2^+ to the common motional mode of the ions
- Motional excitation is detected on Ca^+ without destroying the N_2^+ state



[1] P. O. Schmidt, T. Rosenband, C. Langer, W. M. Itano, J. C. Bergquist, and D. J. Wineland. Science, 309:749, 2005. ($^{27}Al^+/^9Be^+$)

[2] D. Hume, C. W. Chou, **D. R. Leibrandt**, M. J. Thorpe, D. J. Wineland, and T. Rosenband. Phys. Rev. Lett., 107:243902, 2011. ($^{27}Al+/^{25}Mg+$, CME)

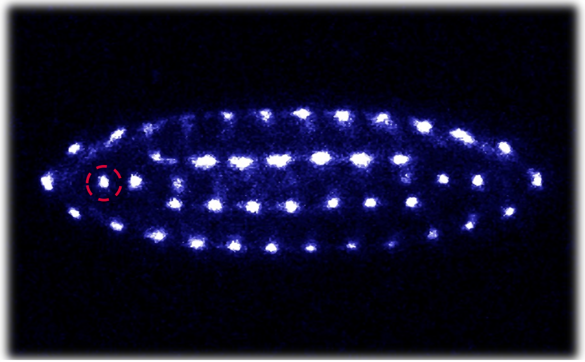
[3] F. Wolf, Y. Wan, J. C. Heip, F. Gebert, C. Shi, and P. O. Schmidt. Nature, 530:457, 2016. ($^{24}MgH^+/^{25}Mg^+$)

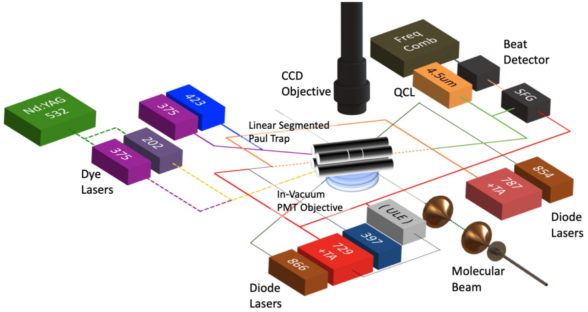
[4] C. W. Chou, C. Kurz, D. B. Hume, P. N. Plessow, **D. R. Leibrandt**, and D. Leibfried. Nature, 545:203, 2017 ($^{40}CaH^+/^{40}Ca^+$)

[5] M. Sinhal, Z. Meir, K. Najafian, G. Hegi, and S. Willitsch. Science, 367:1213, 2020. ($^{14}N_2^+/^{40}Ca^+$)

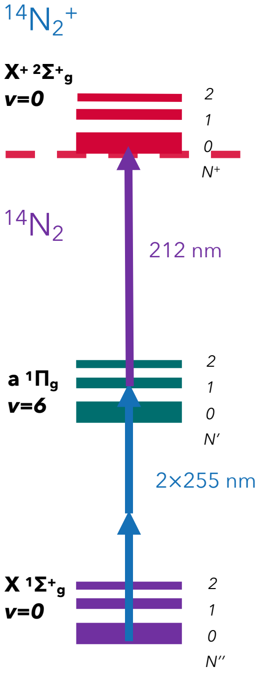
Ions state preparation and Doppler cooling

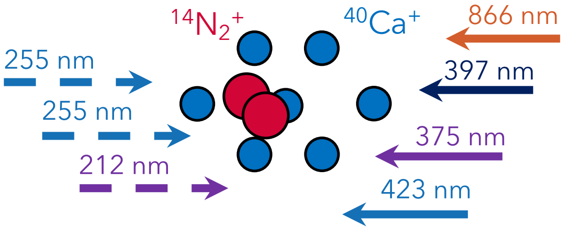
Coulomb crystal of Ca^+ ions and single N_2^+





Photoionisation scheme

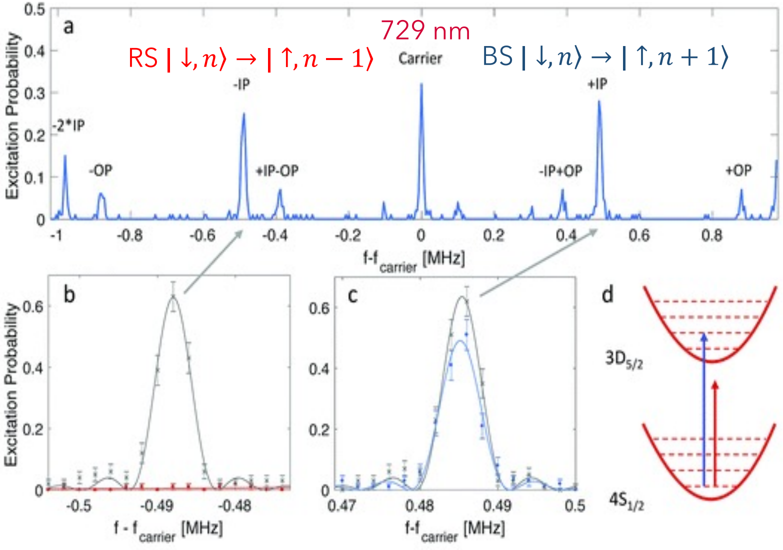




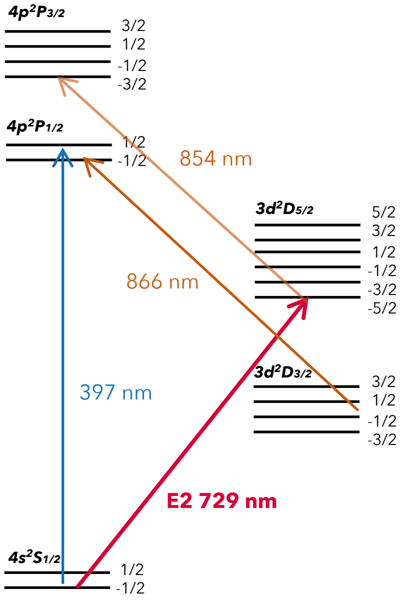
A. Gardner, T. Softley, and M. Keller. Sci Rep, 9:506, 2019.

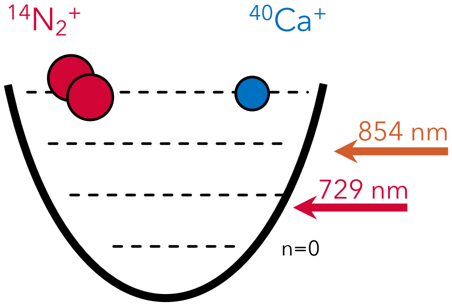
Motional ground-state cooling

Motional sidebands of the 729 nm clock transition in Ca^+-N_2^+ ion string



Ca^+ energy levels scheme

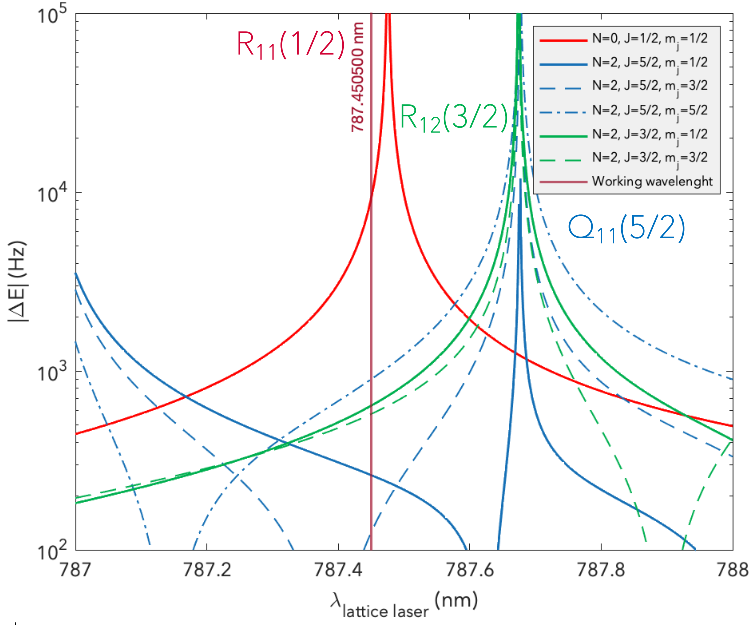




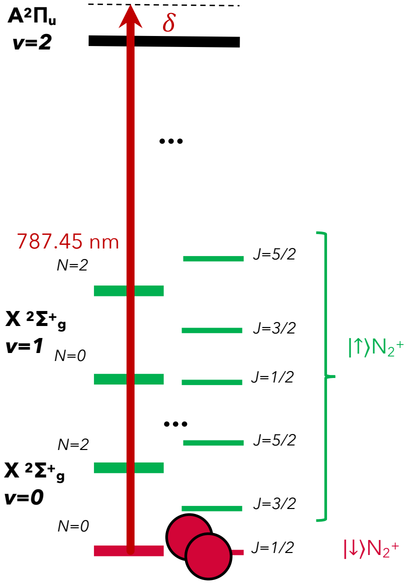
Z. Meir, G. Hegi, K. Najafian, M. Sinhal, and S. Willitsch. Faraday Discuss., 217:561, 2019.

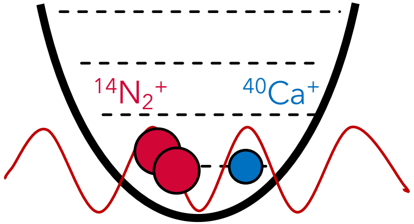
Optical dipole force

AC stark shift (ΔE) experienced by N_2^+ in different rotational states of the ground vibronic state



N_2^+ energy levels scheme





$$\Delta E_{Stark} = \frac{\hbar|\Omega_R^2|}{2\delta}$$

$$F_{ODF} = -\nabla \cdot \Delta E$$

M. Sinhal, Z. Meir, K. Najafian, G. Hegi, and S. Willitsch. Science, 367:1213, 2020.

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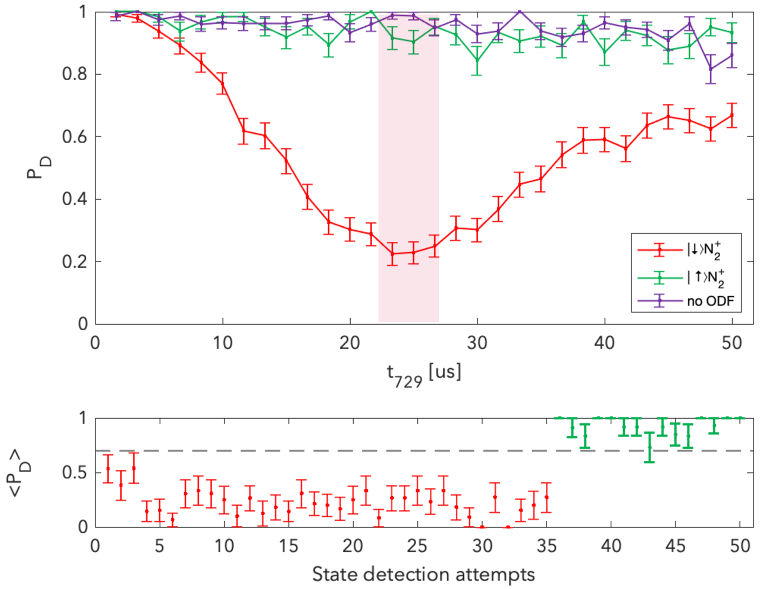
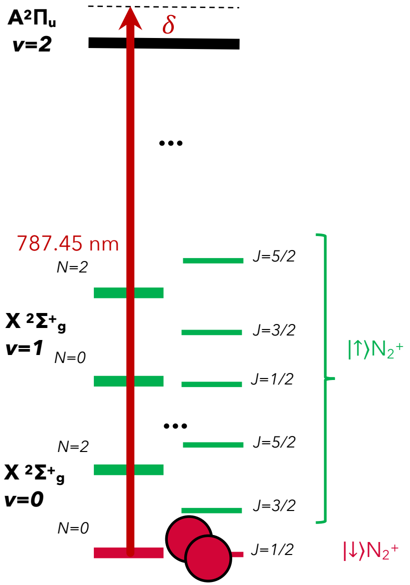
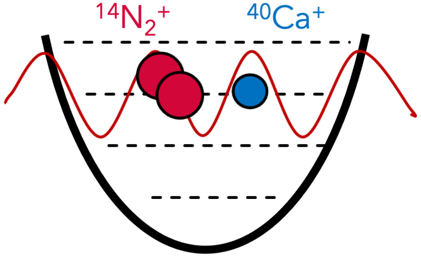
Non-demolition state detection

State-detection signal on the Ca$^+$ 729 nm clock transition's blue-motional sideband



N$_2^+$ energy levels scheme



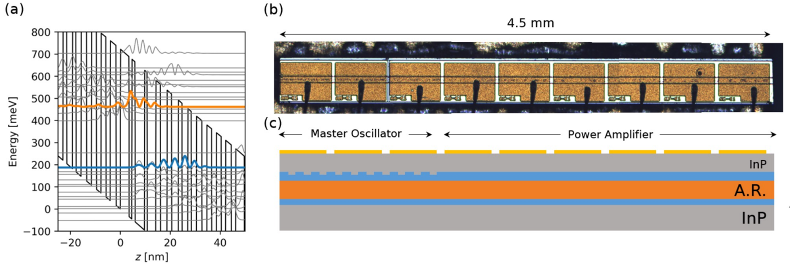


$$\Delta E_{Stark} = \frac{\hbar\left|\Omega_R^2\right|}{2\delta}$$

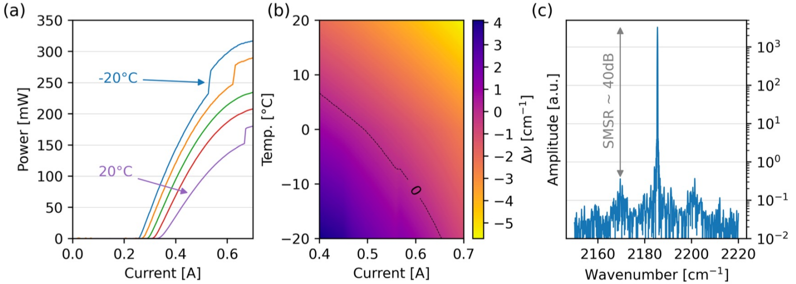
$$F_{ODF} = -\nabla \cdot \Delta E$$

Quantum Cascade Laser for mid-IR spectroscopy

One period of the QCL active region, top-view photograph and schematic of the device

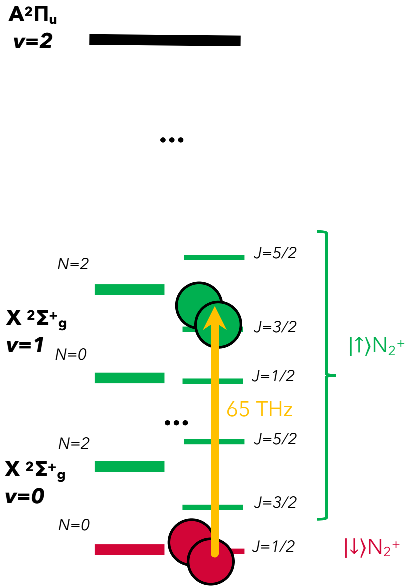


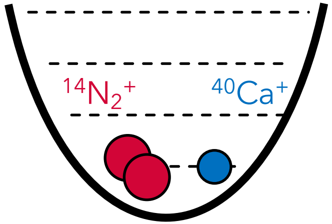
Power, frequency tunability and emission spectrum



M. Bertrand, A. Shlykov, M. Shahmohamadi, M. Beck, S. Willitsch, and J. Faist. Photonics, 9, 2022.

N_2^+ energy levels scheme





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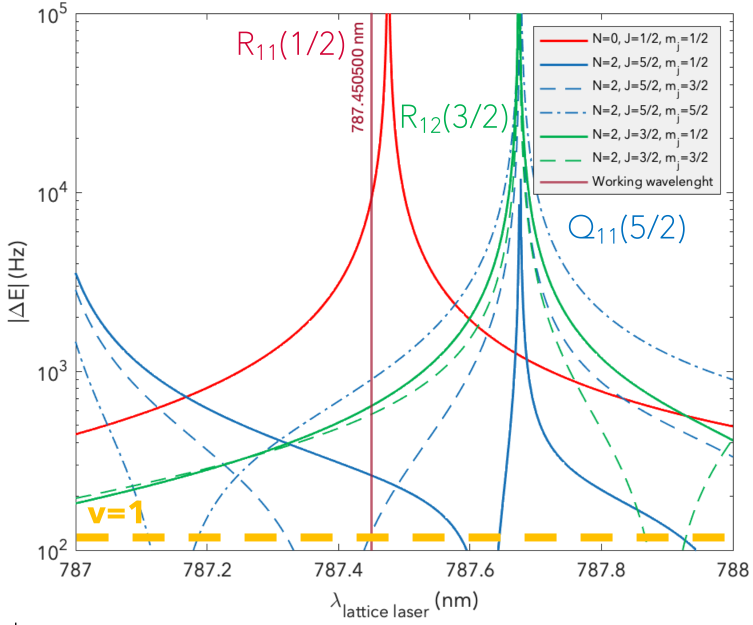
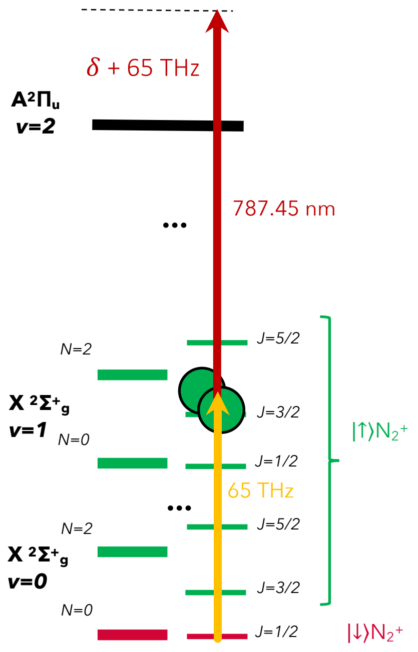
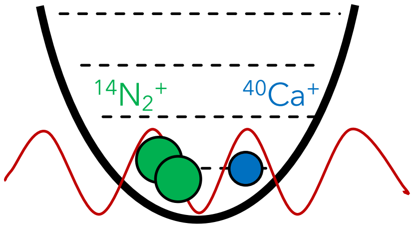
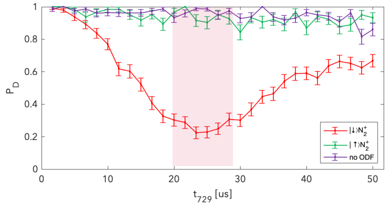
Quantum-logic spectroscopy of a single N_2^+

AC stark shift experienced by N_2^+ in different rotational and vibronic states









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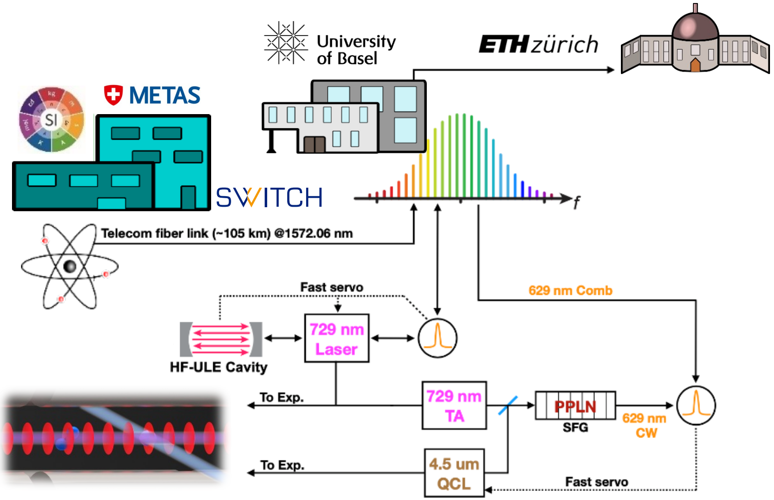
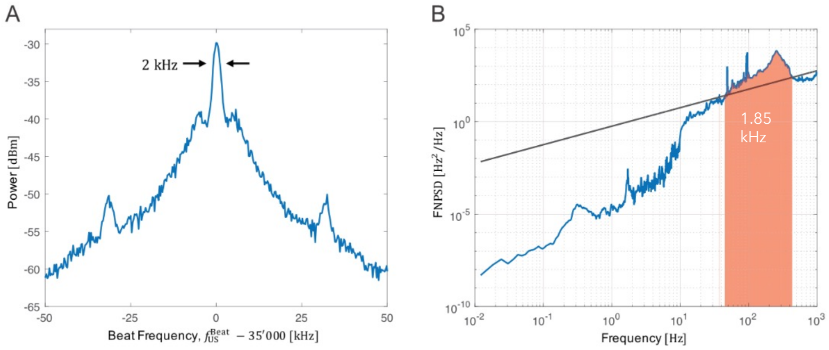
Frequency stabilisation of spectroscopy lasers

- The frequency standard delivered via the academic fibre link provides long-term stability and SI definition of second tracing



(A) Beat note between stabilised OFC and ultrastable laser
(B) FNPSD of ultrastable signal dominated with fibre noise



M. Sinhal, A. Johnson, and S. Willitsch. Molecular Physics, 0:e2144519, 2022.

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Frequency stabilisation of spectroscopy lasers

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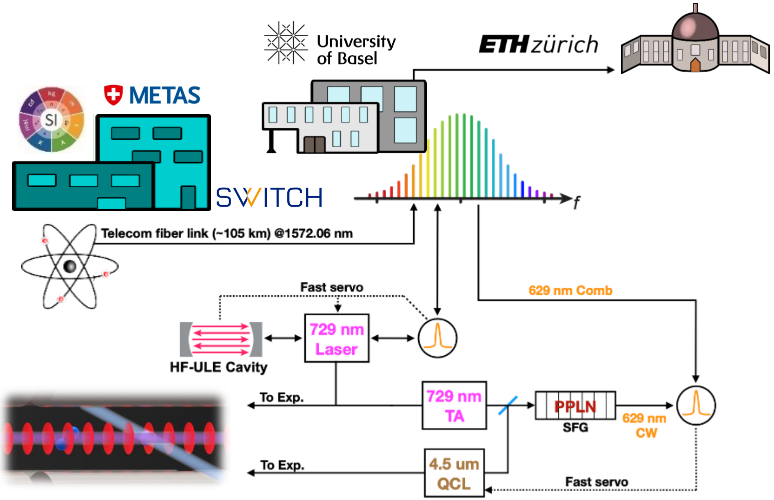
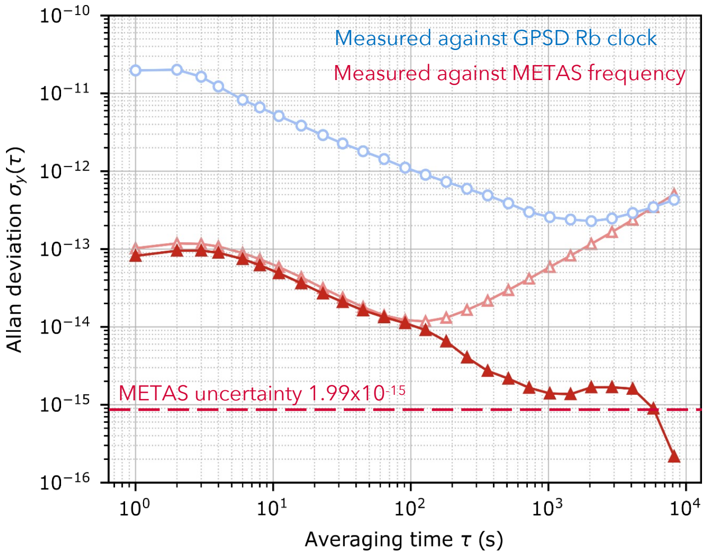


Alan deviation of the 729 nm laser referenced to Rb clock and ultra-stable laser in METAS



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

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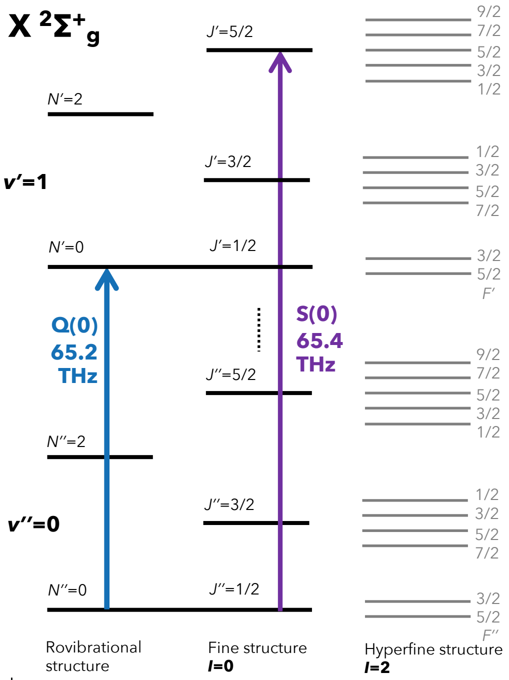
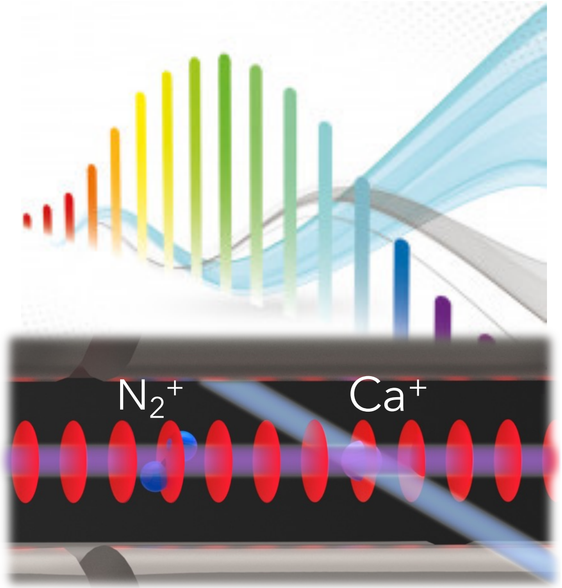
Frequency comparison

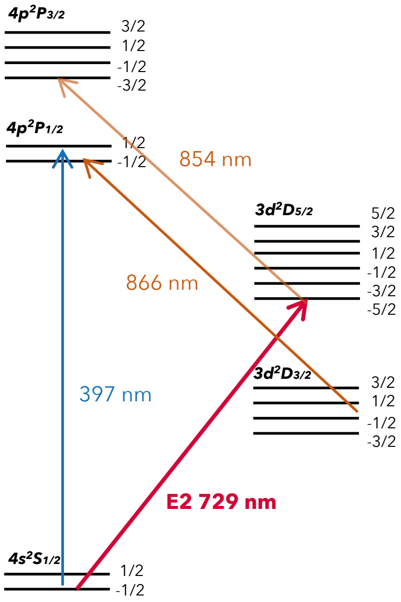
N_2^+ ground state energy levels scheme





Pictures: Mudit Sinhal, Menlo Systems

Ca^+ clock $3x10^{-18}$ [1]



[1] Y. Huang, B. Zhang, M. Zeng, Y. Hao, Z. Ma, H. Zhang, H. Guan, Z. Chen, M. Wang, and K. Gao. Physical Review Applied, 17:034041, 2022.

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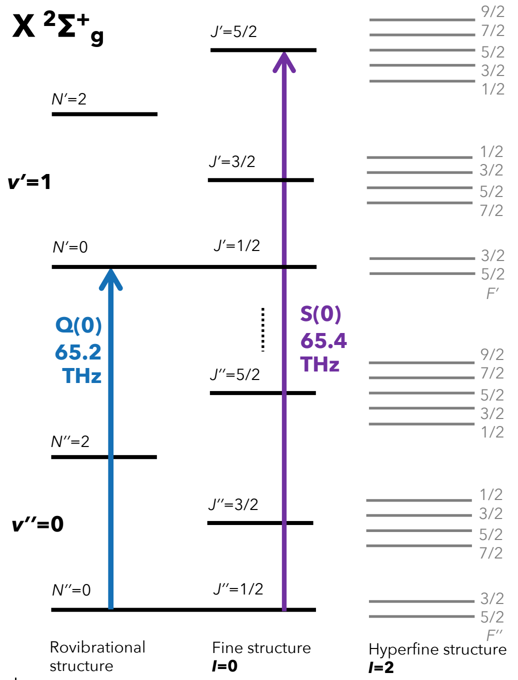
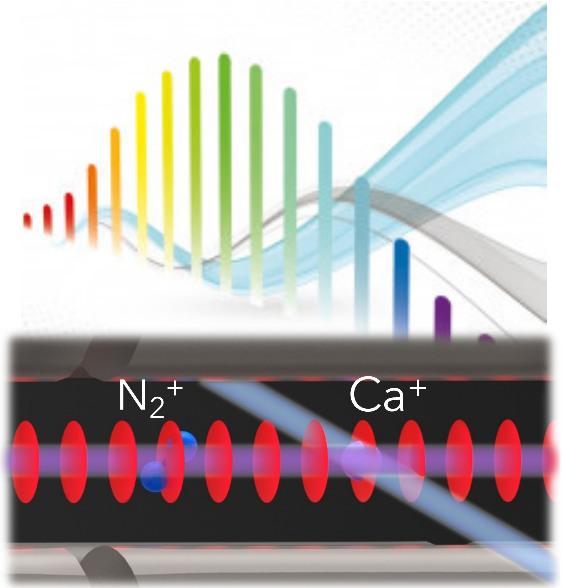
Frequency comparison

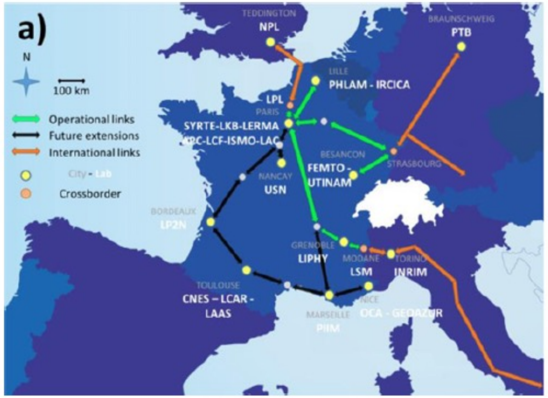
N_2^+ ground state energy levels scheme





Pictures: Mudit Sinhal, Menlo Systems

European ultra-stable frequency dissemination network



Cantin et al., New J. Phys. 23 (2021) 053027

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Summary

- Molecules provide direct sensitivity to the possible variation of the proton-to-electron masses ratio
- Quantum-logic allows to control individual molecules on the quantum level
- The mid-IR molecular nitrogen ion clock can become a new-generation device for probing BSM physics

Acknowledgments

- Quantum Technologies team:


Prof. S. Willitsch


A. Shlykov


M. Roguski


R. Karl


Dr. Z. Meir


Dr. M. Sinhal


Dr. K. Najafian


Dr. G. Hegi

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SWITCH
FNSNF
QSIT Quantum Science and Technology
National Centre of Competence in Research
QCQT PhD School

Thank you!

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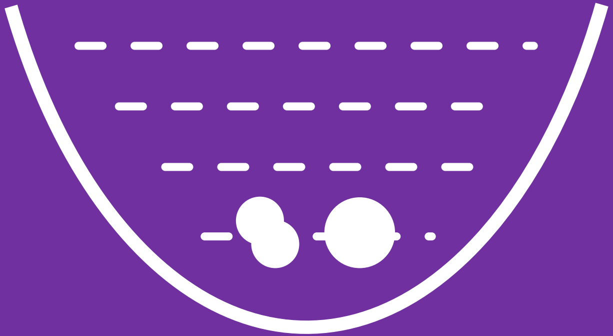
Molecules and Ions Group



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