Physikalisch-Technische Bundesanstalt
Braunschweig and Berlin
National Metrology Institute

Nuclear Laser Spectroscopy of Thorium-229

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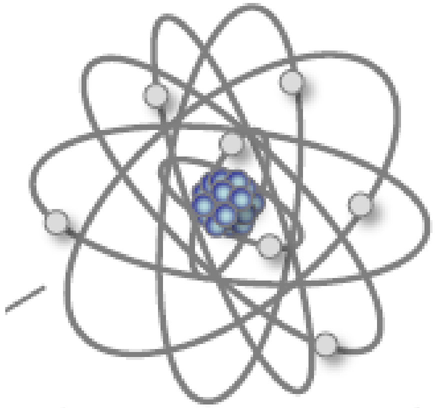
European Research Council
Established by the European Commission

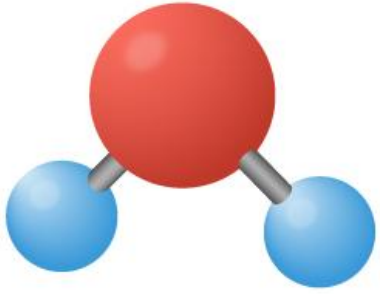
International Conference on Atomic Physics, ICAP29, Wuhan, China, 15.-19.06.2026

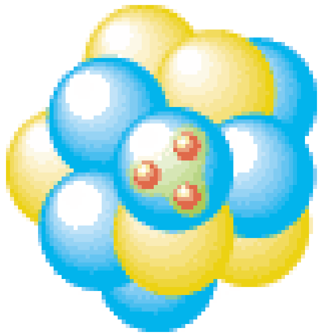
Atoms

Molecules

Nuclei





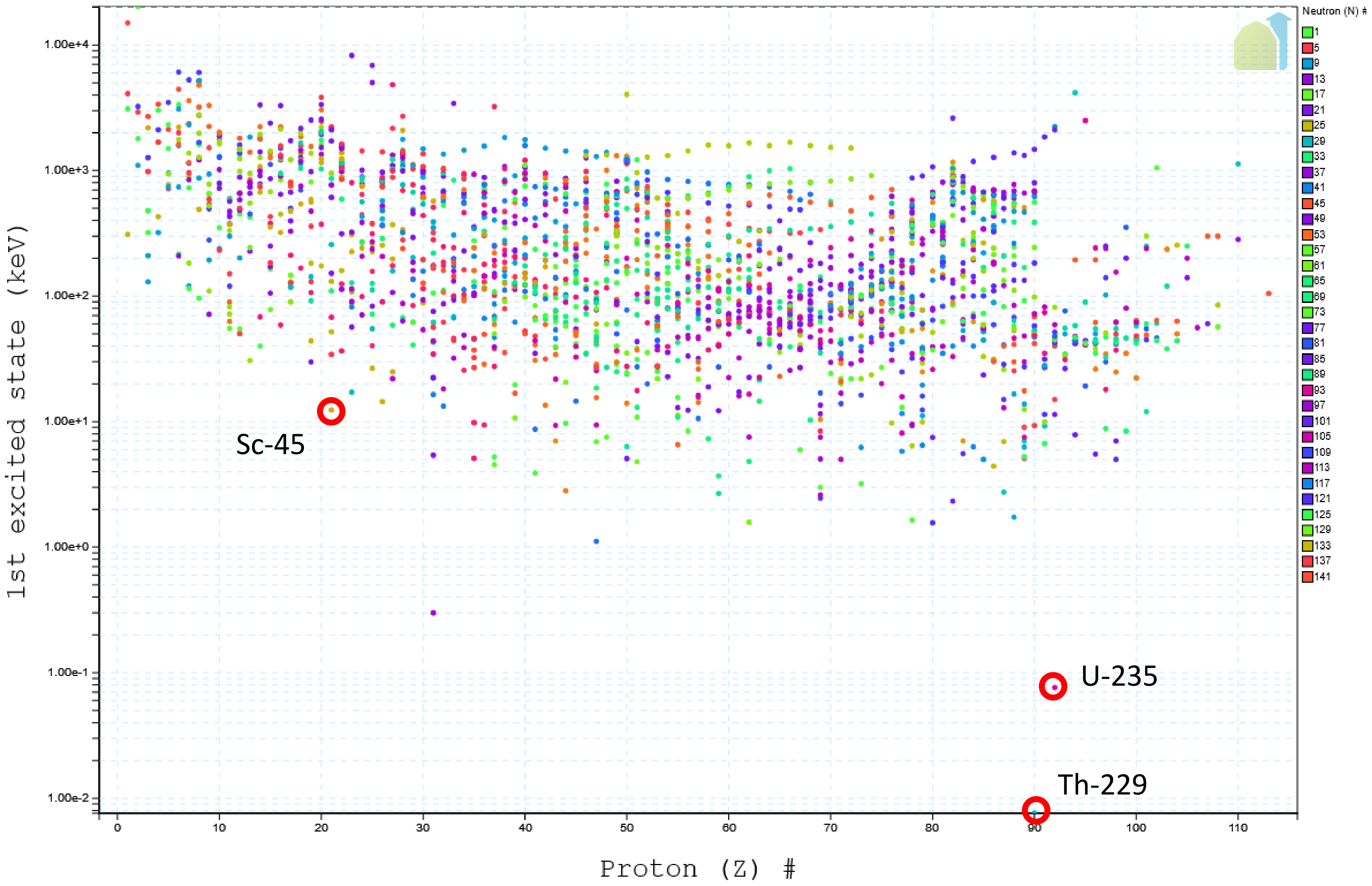


Precision Measurements
Coherent Control of Quantum States

Cooling and trapping
Laser Spectroscopy
Optical Frequency Measurements

Nuclear Magnetic Resonance
Laser Mössbauer Spectroscopy
Nuclear-electron Double Resonance

Low-energy nuclear physics: Energy of the first excited state



https://www.nndc.bnl.gov/nudat3/

Sc-45:
12389.59 eV, M2

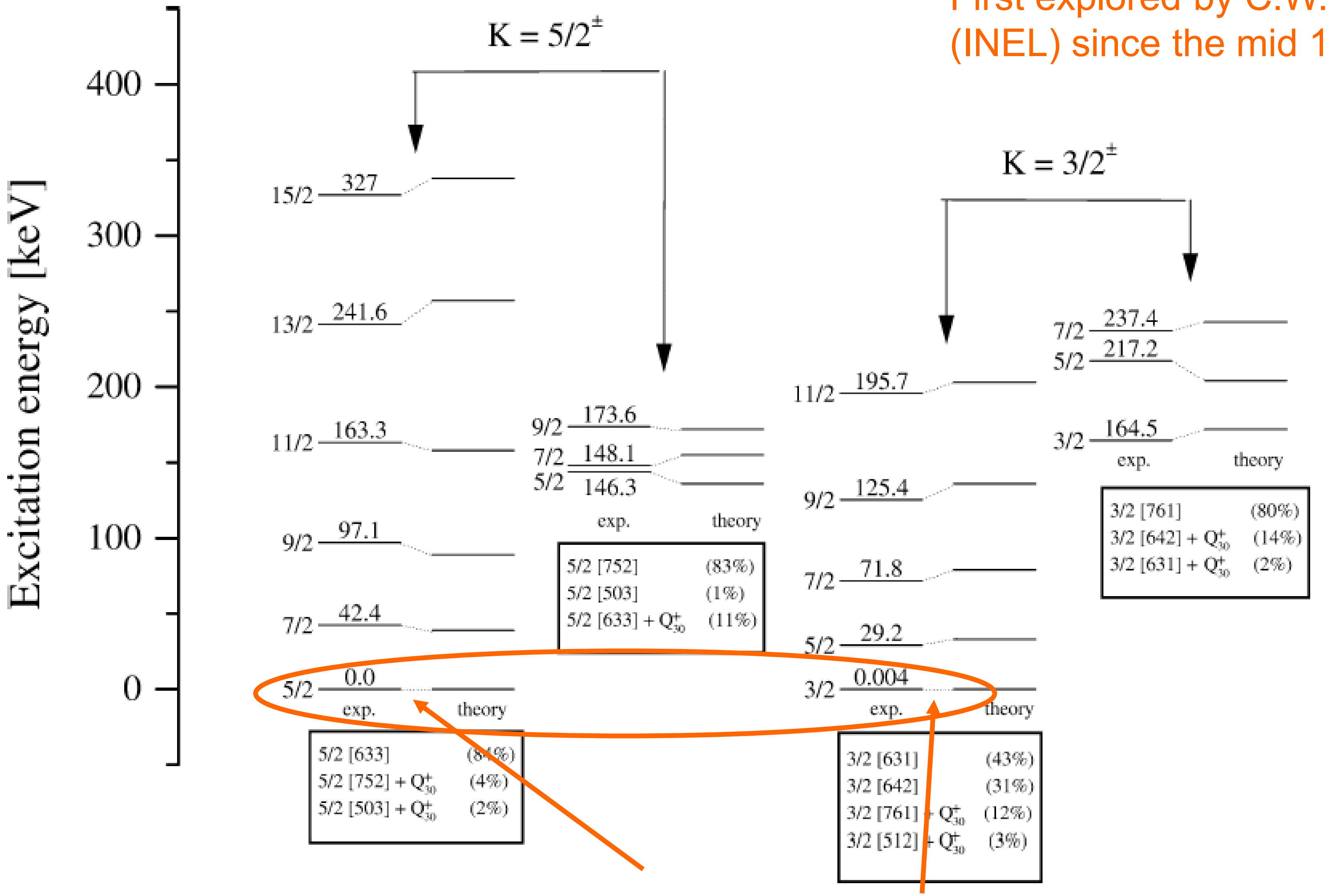
U-235:
76.737 eV, E3

Th-229:
8.4 eV, M1

K. Gulda et al. / Nuclear Physics A 703 (2002) 45–69

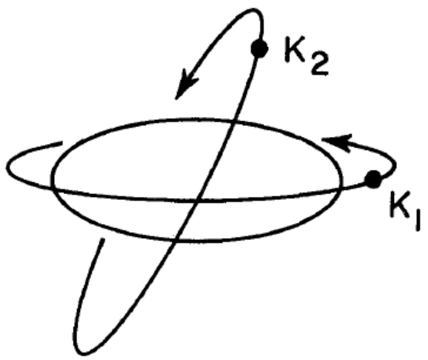
The nuclear structure of ^{229}Th

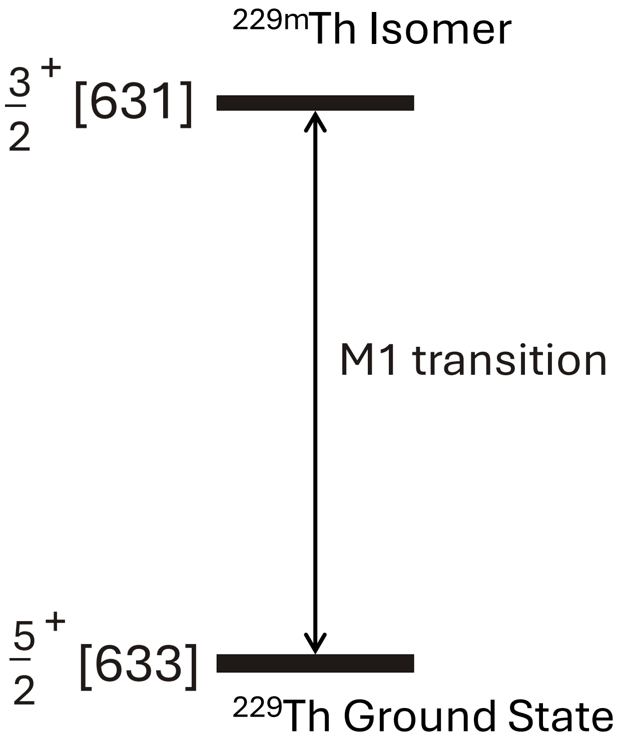
Very well classified into rotational bands.
First explored by C.W. Reich et al. (INEL) since the mid 1970ies.



Two close-lying band-heads: ground state and isomer

The Th-229 low-energy nuclear transition





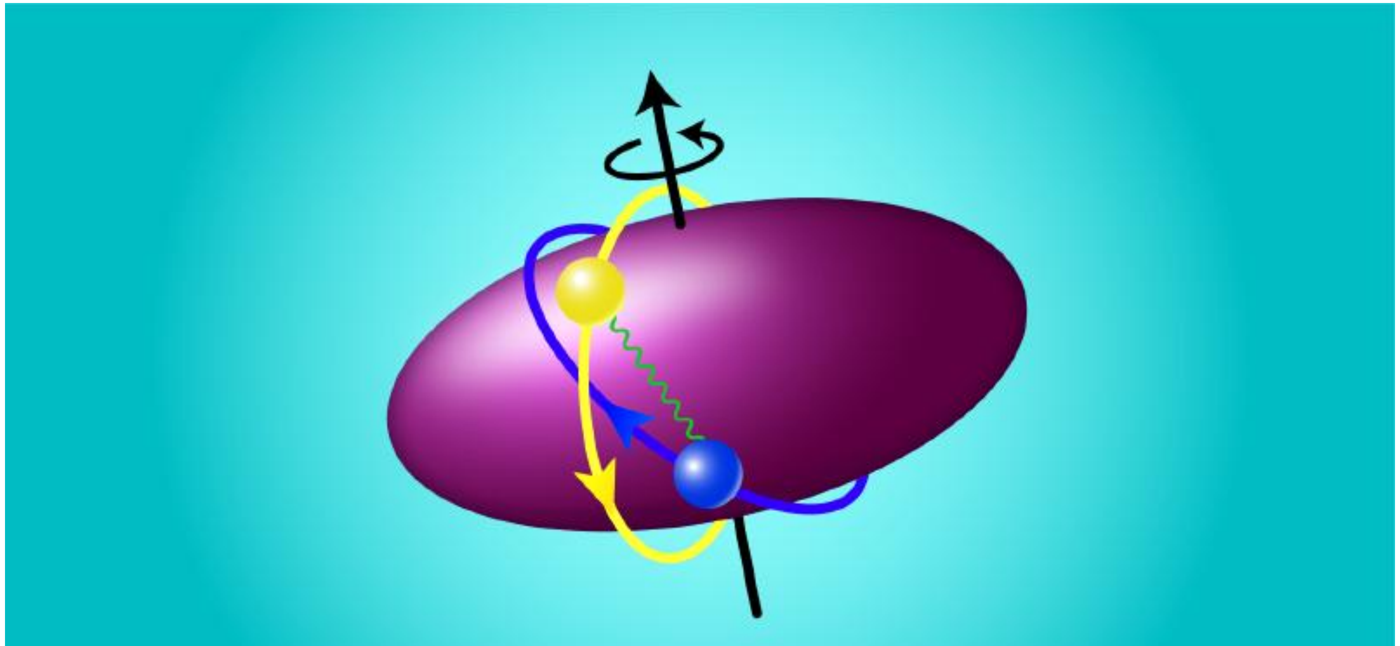
μ=-0.378 μ_N
Q=1.77×10^{-28} e·m^2
τ_{rad}=2500 s, τ_{IC}=7 μs

ΔE=8.355732 eV
λ=148.3822 nm
ν=2020.407 THz

μ=0.365 μ_N
Q=3.11 ×10^{-28} e·m^2
$t_{1/2}$=7920 yr (α-decay)

Nuclear structure:
The deformed shell model or Nilsson model

Collective motion of an elliptical core
combined with
single-particle motion of an
unpaired „valence“ nucleon
(much faster than motion of the core)



R. Stroberg; APS/C. Cain

Sven Gösta Nilsson

Not a complete description of the Th-229 transition:
The proton distribution in the core would not change.

But hyperfine structure measurements have shown changes in:

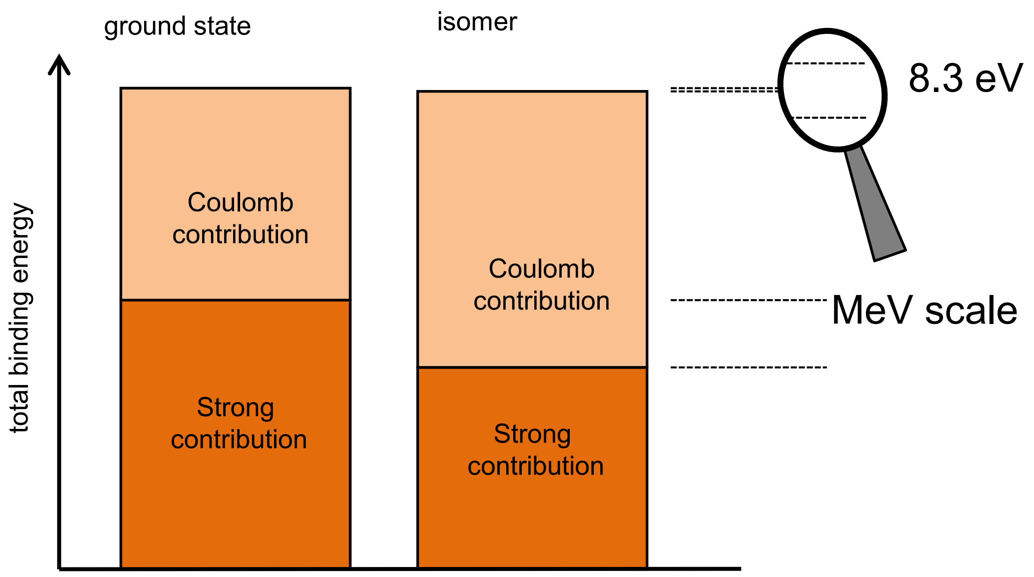
RMS charge radius:	0.016%
Intrinsic quadrupole moment:	1.791%

J. Thielking et al., Nature **556**, 321 (2018)
K. Beeks et al., Nature Comm. **16**, 9147 (2025)

How does the very low energy appear?

„Magic“ cancellation of contributions from Coulomb and strong forces

V.V. Flambaum, Phys. Rev. Lett. 97, 092502 (2006)



→ High sensitivity of a Th-229 nuclear clock in fundamental tests *(O(8 eV / 1 MeV)):*

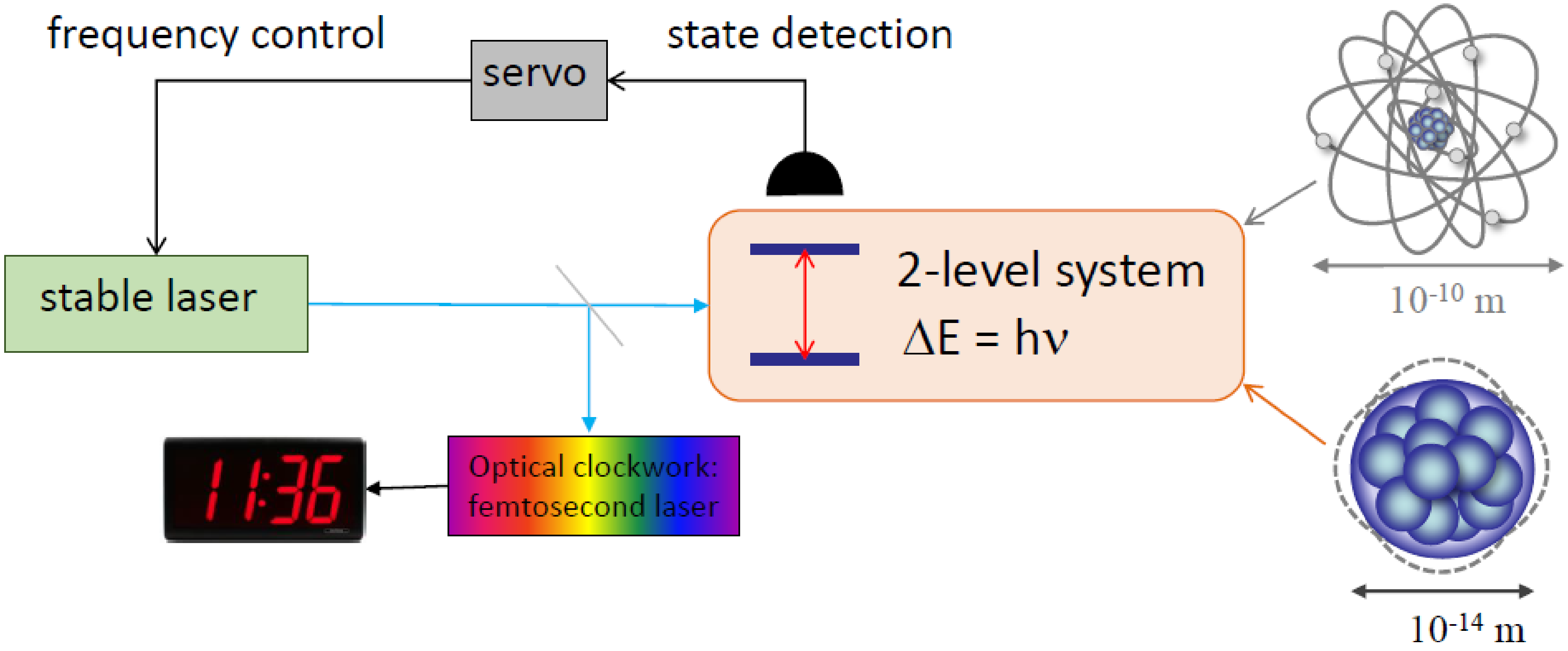
Search for variations of fundamental constants

or other violations of the Einstein equivalence principle

For a review see:

E. Peik, T. Schumm, M. Safronova, A. Pálffy, J. Weitenberg, P.G. Thirolf, Quant. Sci. Tech. 6, 034002 (2021)

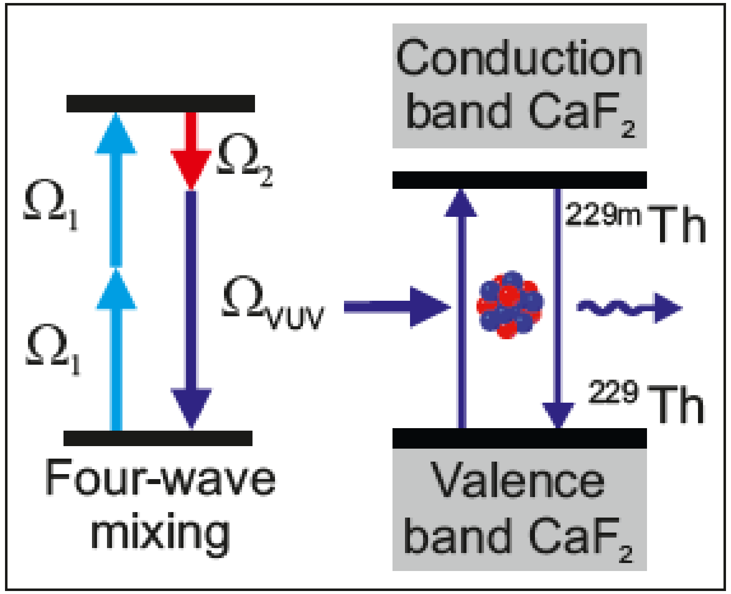
From the atomic to the nuclear clock

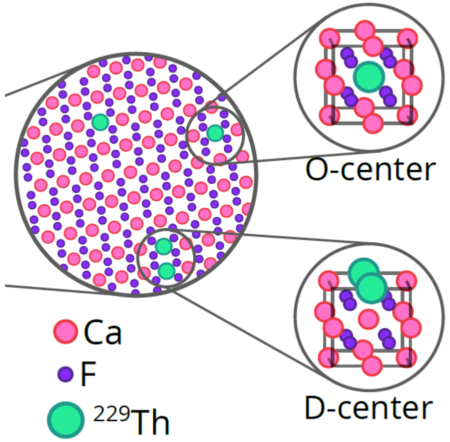


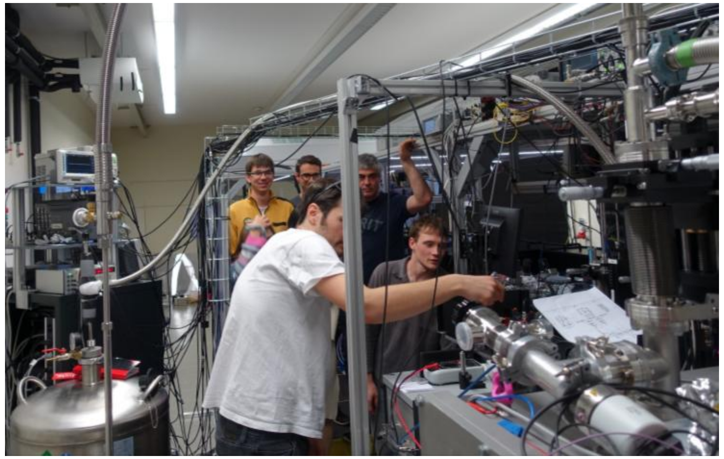
The nuclear clock promises:

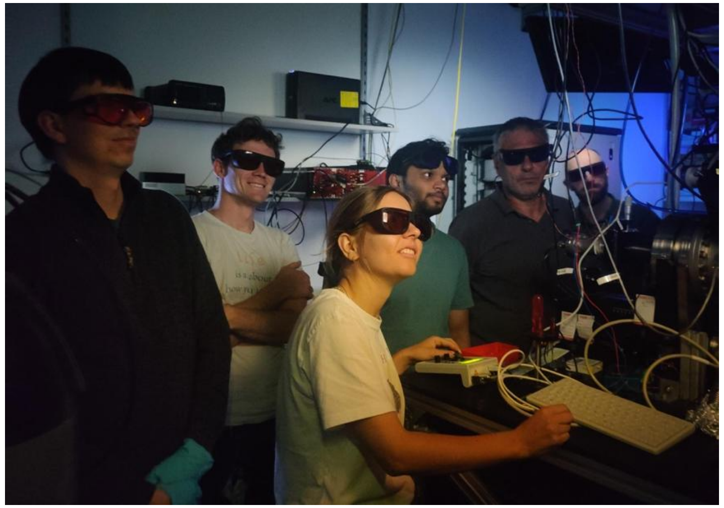
- High accuracy (with laser cooled trapped ions)
- High stability (in the solid state as a laser Mössbauer system)
- High sensitivity to new physics (also to strong interaction)

PTB - TU Wien cooperation on laser excitation of ^{229}Th-doped calcium fluoride crystals









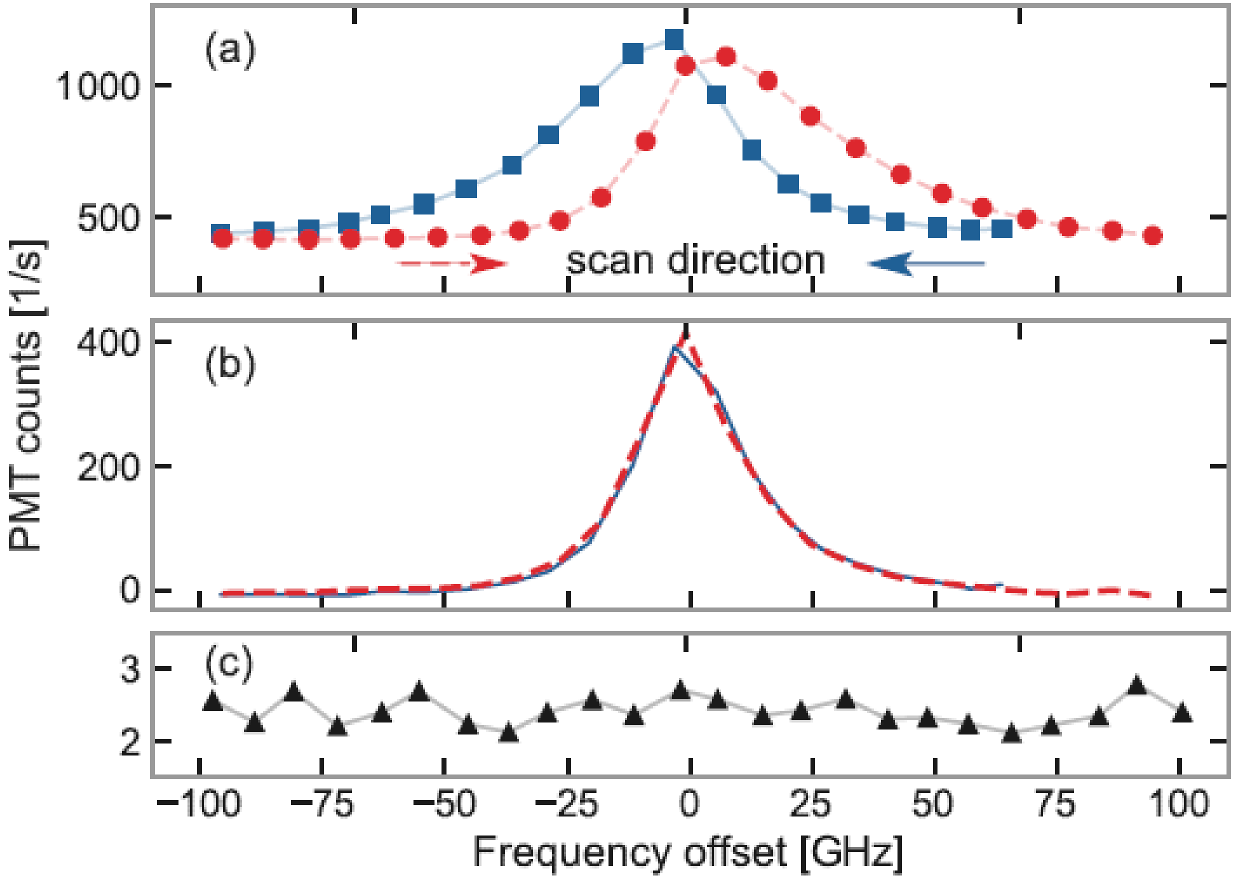
Joint experiments in Braunschweig and Vienna

European Research Council
Established by the European Commission

Crystal growth:
K. Beeks et al. (TU Wien),
Phys. Rev. B **109**, 094111 (2024)
DFT calculations:
M. Pimon et al. (TU Wien)

VUV lasers:
J. Thielking et al. (PTB),
New J. Phys. **25**, 083026 (2023)
V. Lal et al. (PTB, MBI),
Optica **12**, 1971 (2025)

Resonant laser excitation, detected in VUV fluorescence



Excitation spectra at 148 nm: Hitting a narrow line with a broad laser.
(each point: 120 s excitation, 150 s detection)

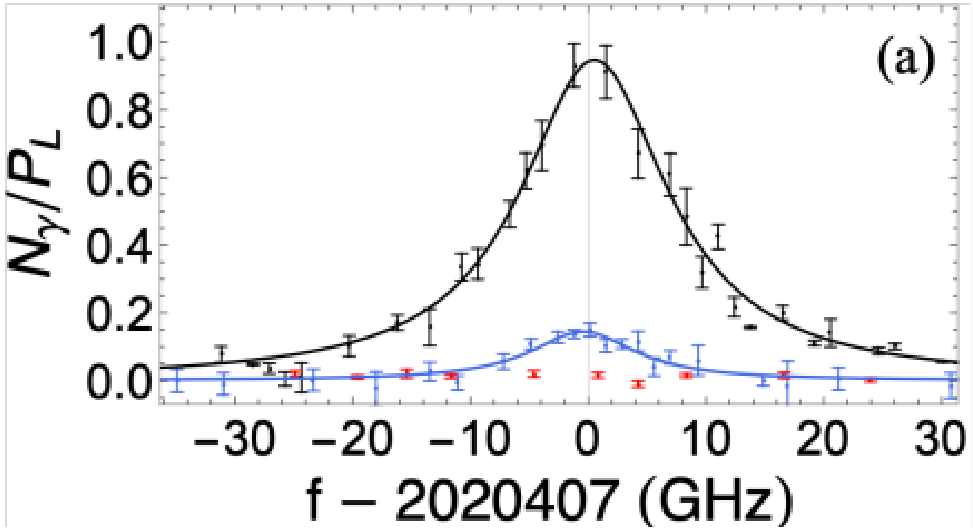
Line shapes after correction for the slow exponential fluorescence decay

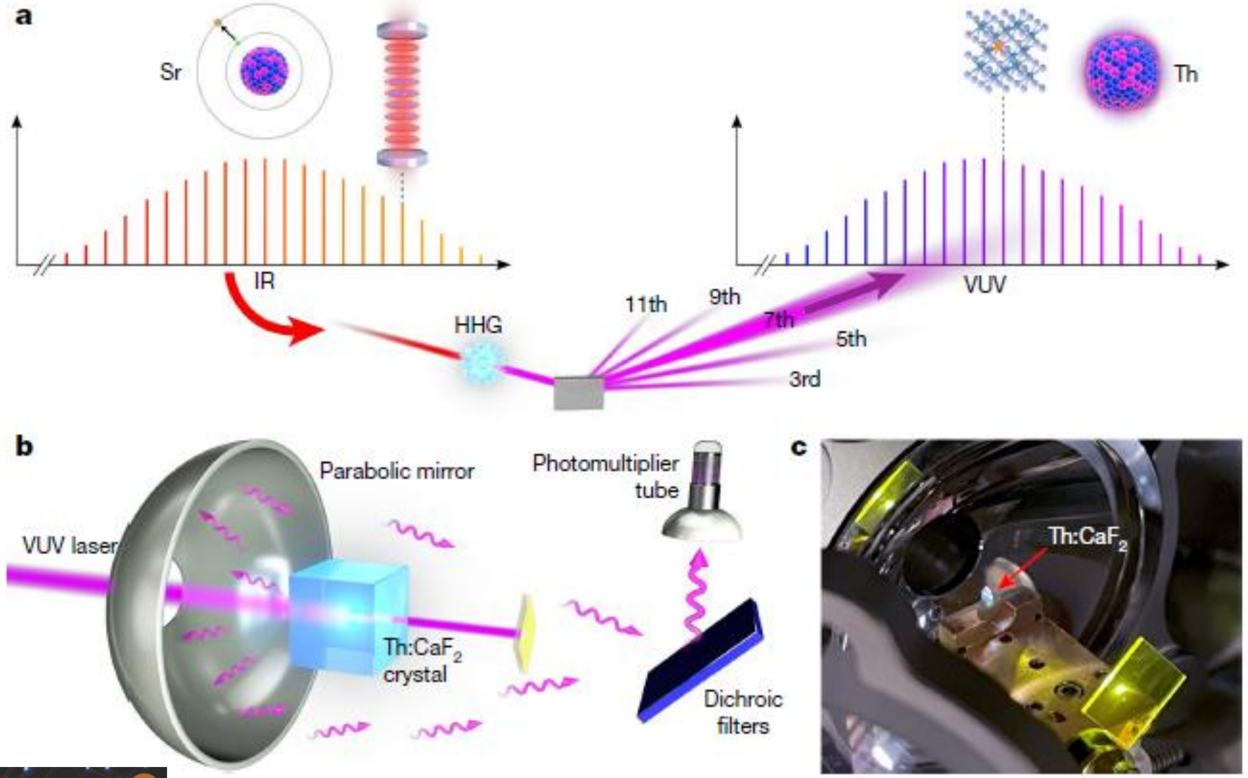
Control experiment with Th-232: no signal

J. Tiedau et al. (PTB – TU Wien cooperation), Phys. Rev. Lett. **132**, 182501 (2024)

Th-229 excitation in Th:LiSAF at UCLA

Th-229 excitation with the 7th harmonic of a fs-laser



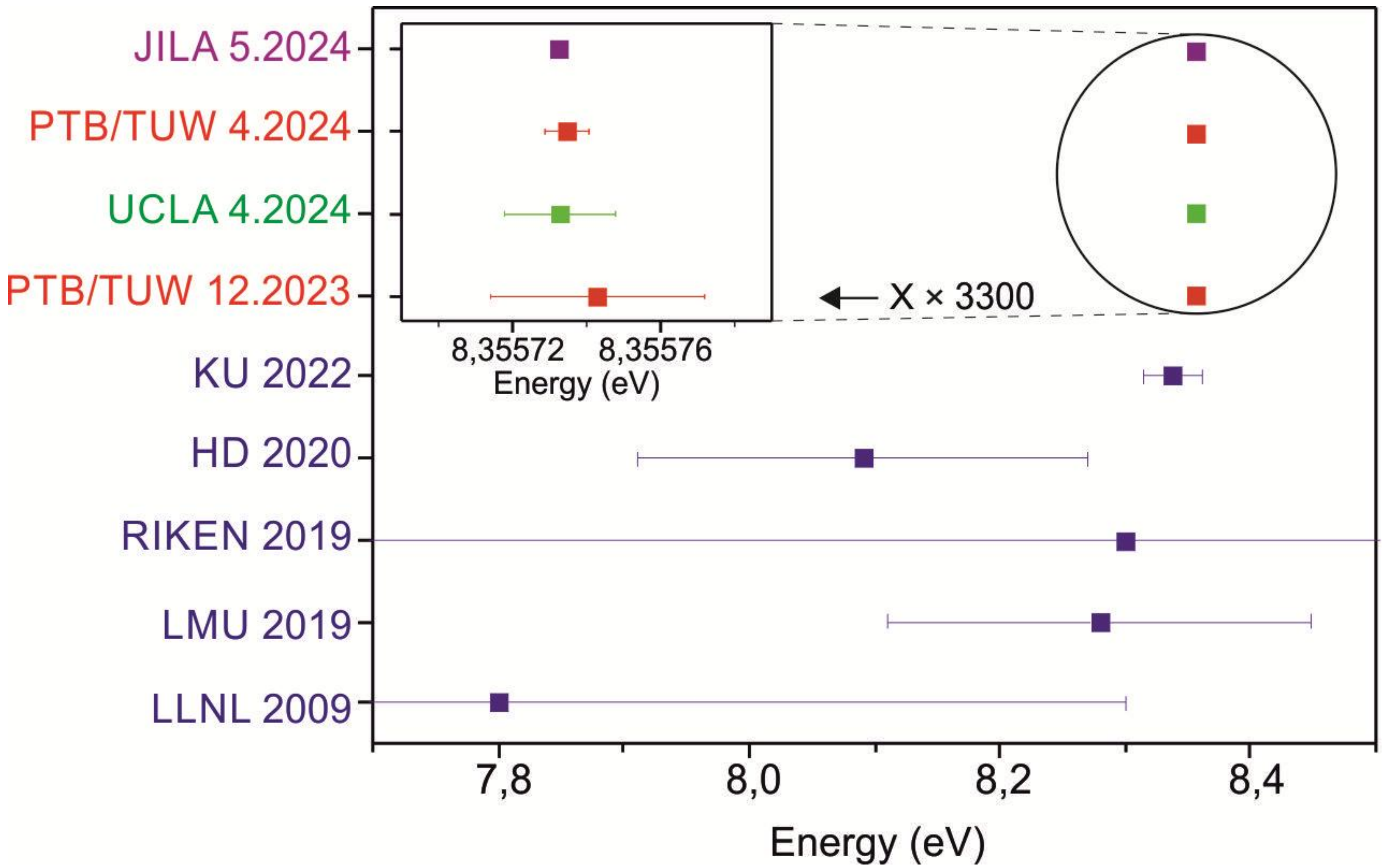


R. Elwell et al., Phys. Rev. Lett. **133**, 013201 (2024)

Ch. Zhang et al. (JILA and TU Wien), Nature **633**, 63 (2024)

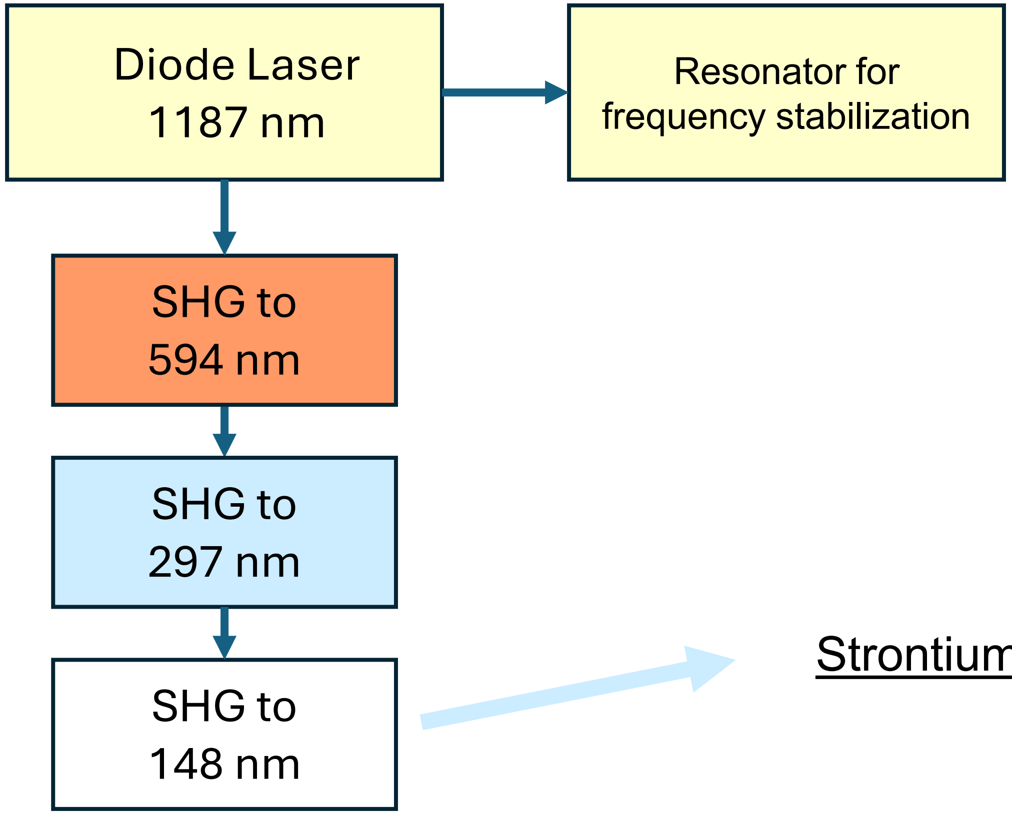
$\nu_{CaF2} = 2020.407384335(2)$ THz

C. Zhang et al.,
Nature 633, 63 (2024)



A narrow-linewidth solid-state CW laser source for 148 nm

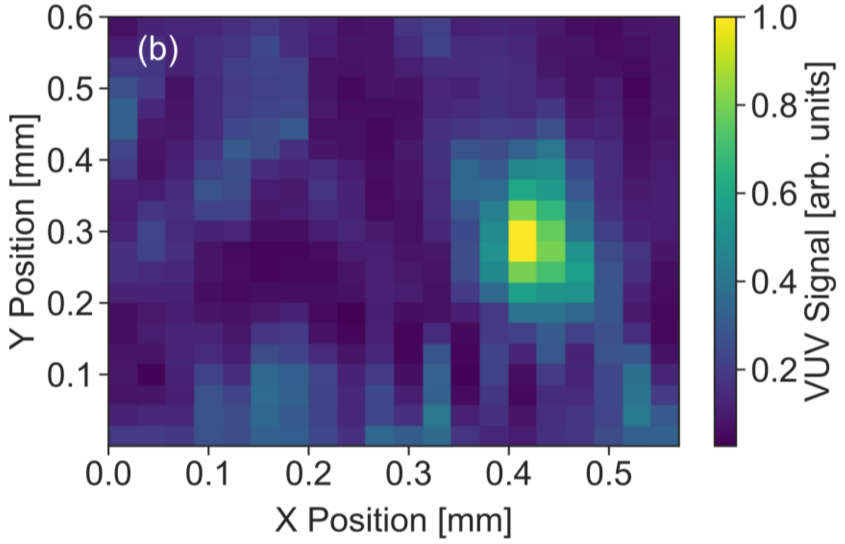
- Infrared diode laser at 1187 nm, stabilized to a reference resonator (e.g. cryogenic silicon cavity)
  → <<1 Hz linewidth demonstrated
- VUV generation by 3× successive frequency doubling in nonlinear optical crystals



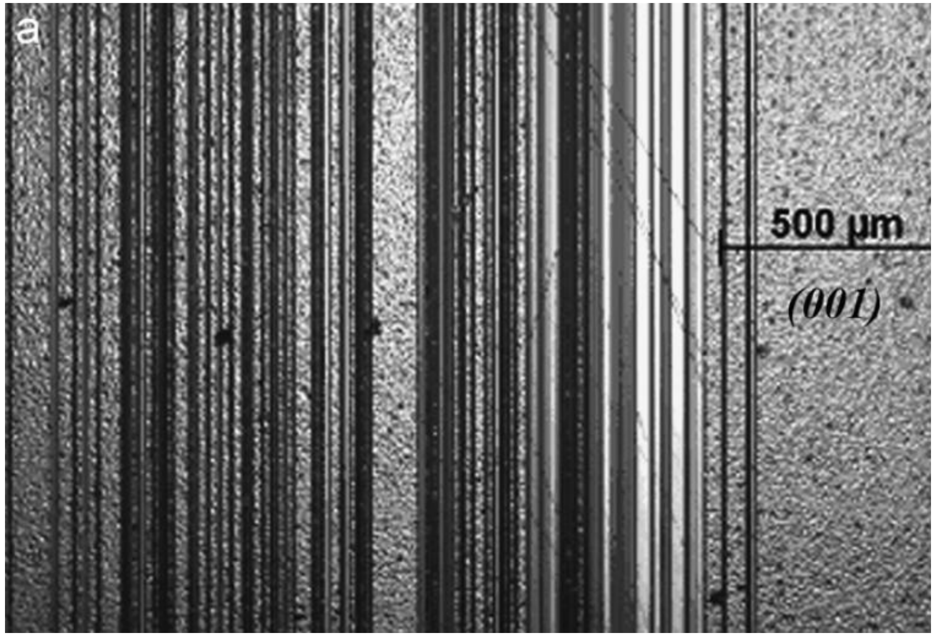
<u>Strontium tetraborate (SBO): SrB_4O_7</u>

Nonlinear optical crystal: Strontium tetraborate (SBO): SrB_4O_7

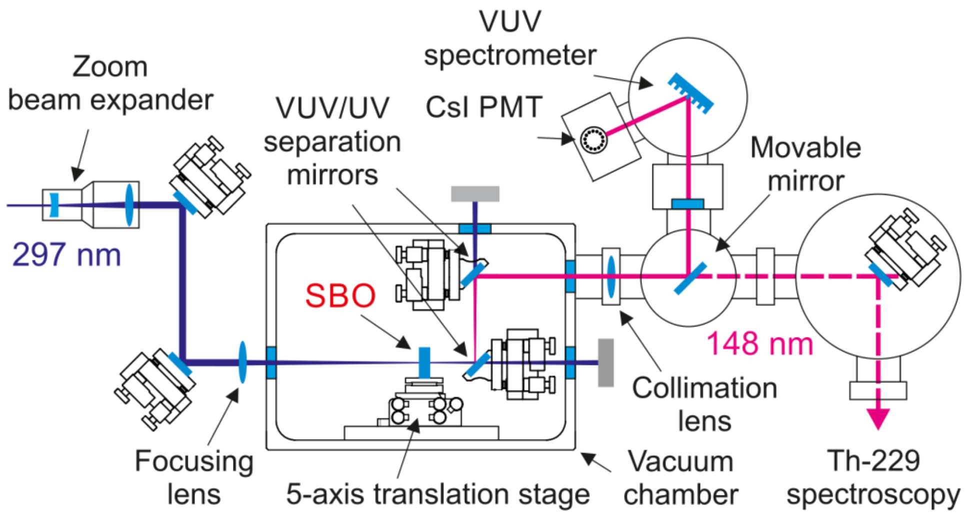
- Transparent down to 120 nm
- Nonlinear coefficient: d_{33} = 1.5 – 3.5 pm/V: highest of all known VUV crystals
- Spontaneously poled domains during growth with opposite orientation for **random quasi-phase-matching**
- SHG of fs-pulses demonstrated down to 121 nm:
  P. Trabs,…V. Petrov, (Max-Born-Inst., Berlin)
  Opt. Lett. **41**, 618 (2016)

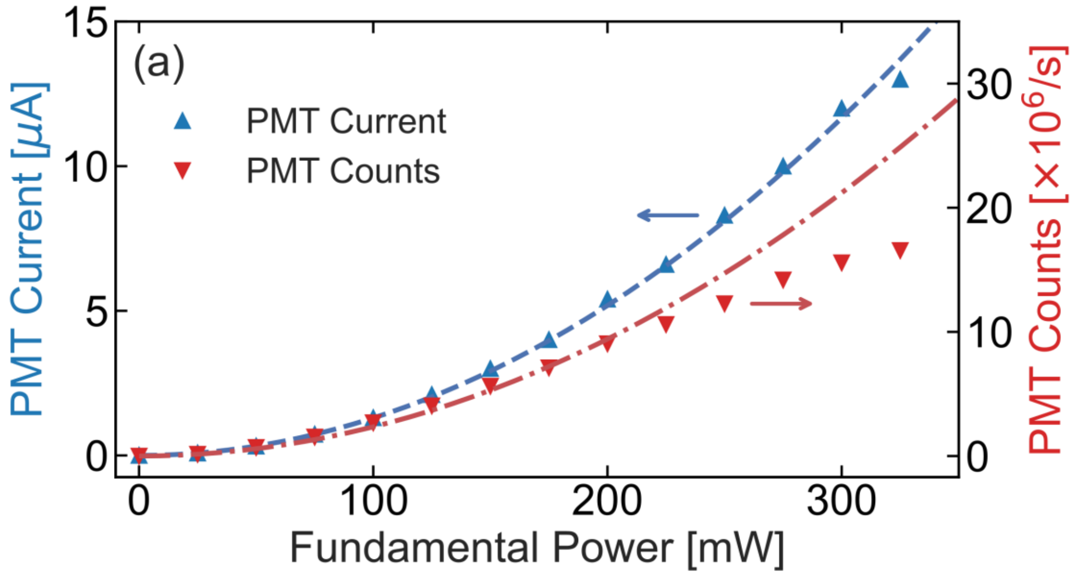


Map of SHG efficiency as function of beam position in the crystal.



Etching pattern of a SBO sample.
A. I. Zaitsev et al.,
J. Cryst. Growth 310, 1 (2008)





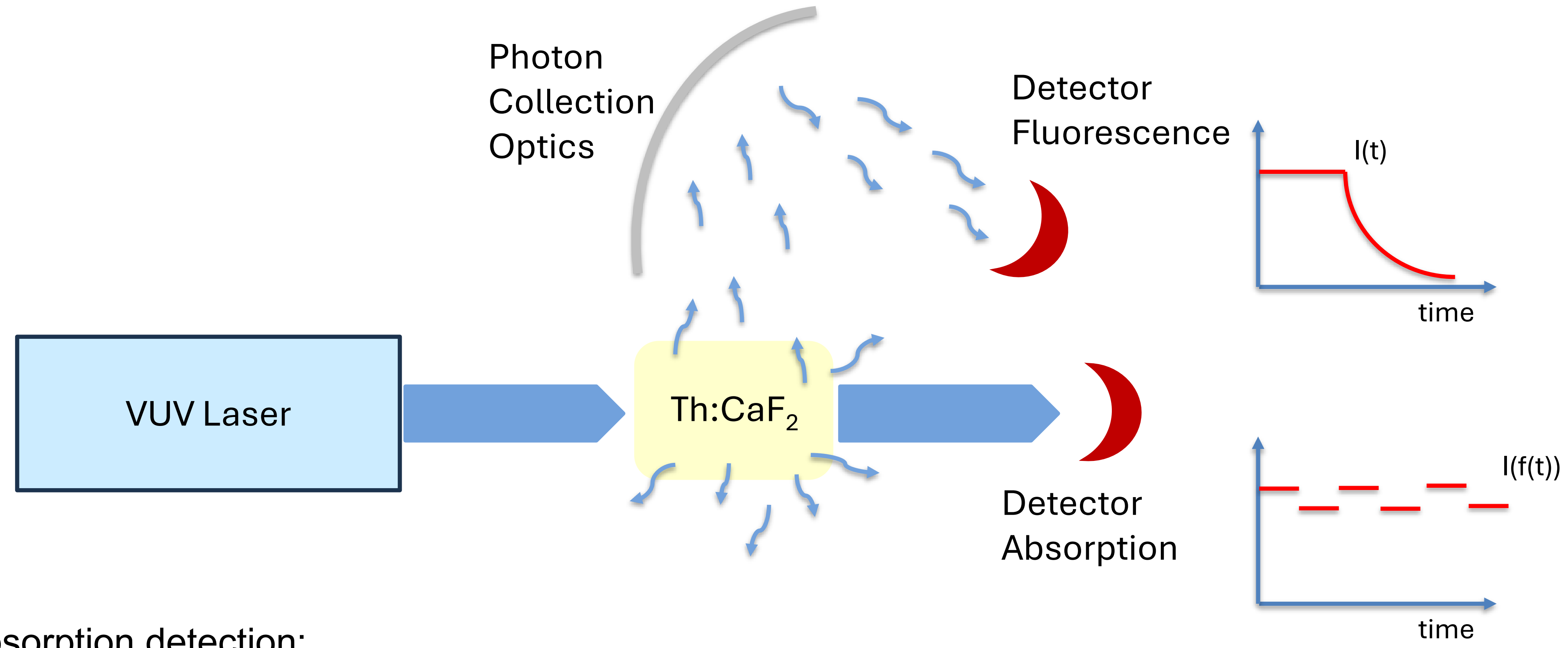
CW radiation at 297 nm from frequency-quadrupled diode laser (Toptica TA-FHG pro)

SHG detection on a solar-blind CsI photomultiplier, suppression of fundamental radiation with two dielectric separation mirrors and a VUV spectrometer.

The generated VUV power is $1.3^{+0.7}_{-0.5}$ nW at an incident UV power of 325 mW.

This work: V. Lal, M. V. Okhapkin, J. Tiedau, N. Irwin, V. Petrov, E. Peik, Optica **12**, 1971 (2025)
See also: Qi Xiao et al., CW four-wave-mixing in cadmium vapor, Nature **650**, 852 (2026)

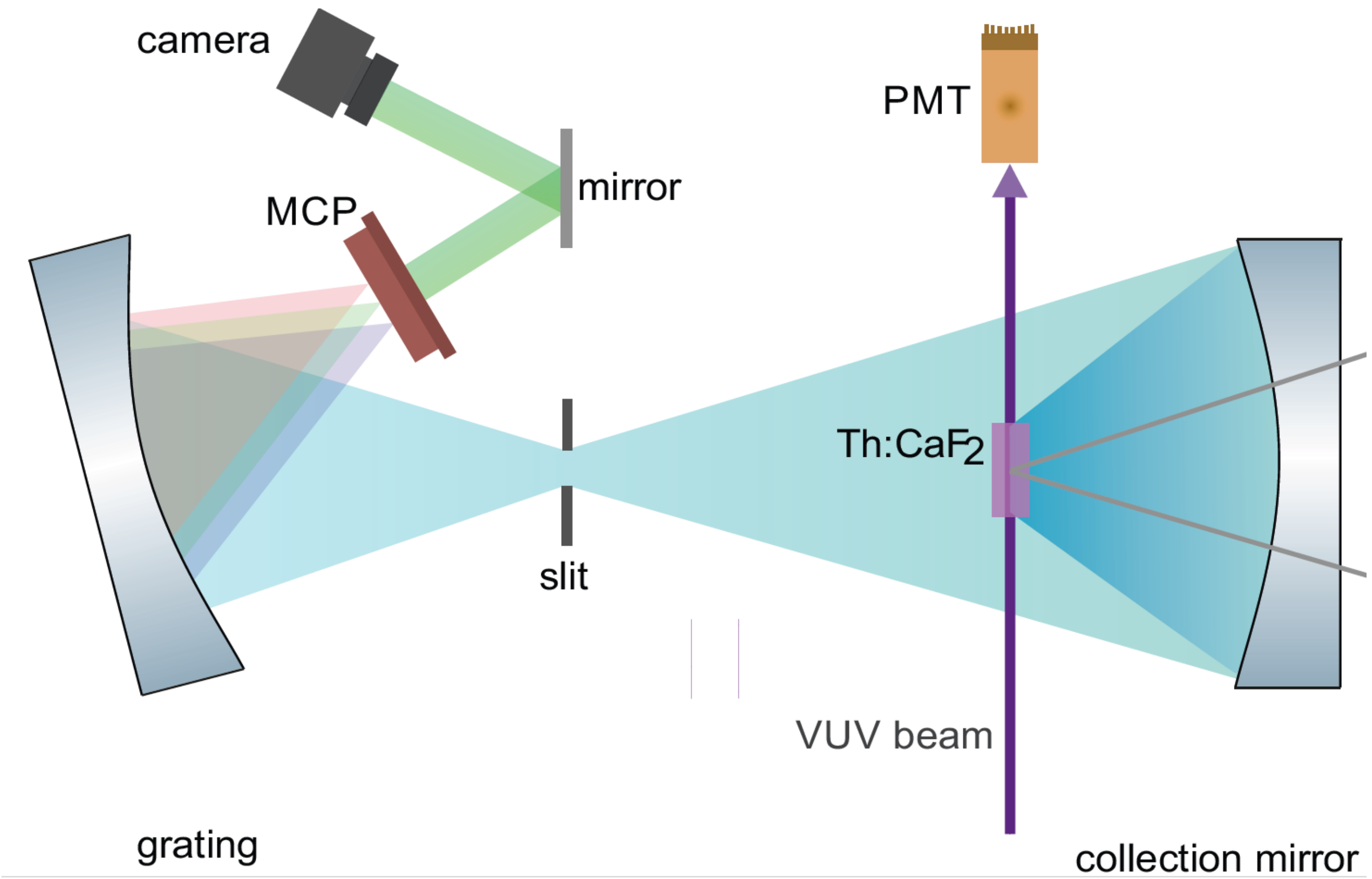
Fluorescence versus absorption detection of the nuclear excitation



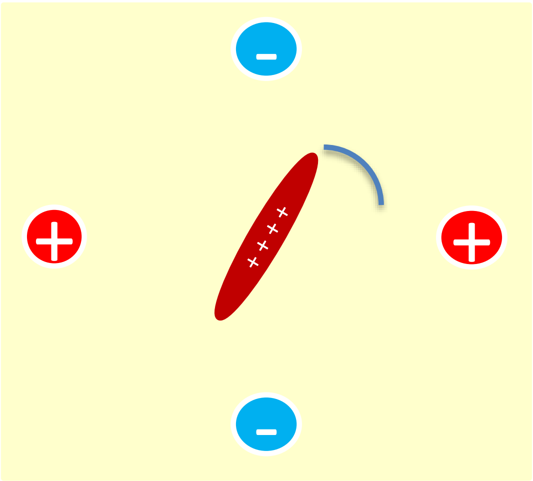
Absorption detection:

- Fast response, not limited by excited state lifetime (610 s)
- Compact setup, no collection optics required
- Easy to suppress the background from radioluminescence and Cherenkov radiation

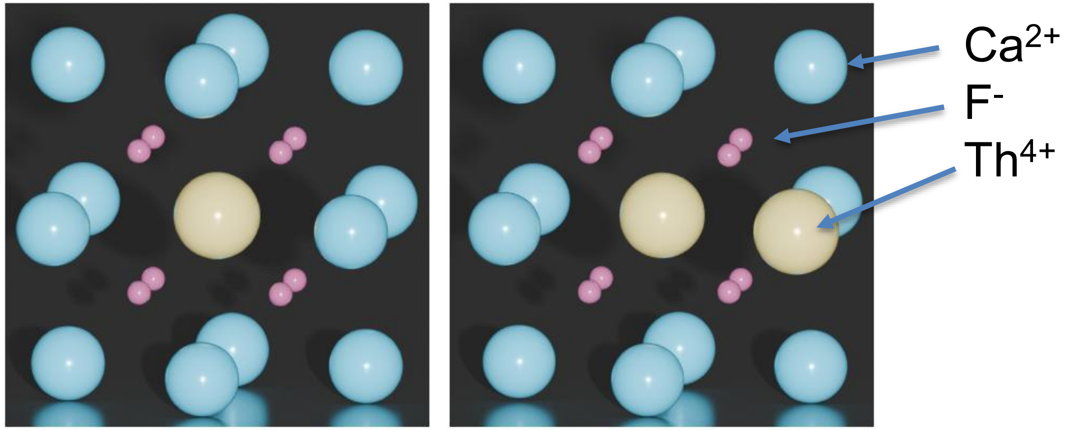
Experimental setup for fluorescence and absorption spectroscopy



Th-229 in CaF_2: structure and interaction with the crystal field



Prolate nuclear charge distribution interacting with an electric field gradient.



O - center

D - center

cubic O_h symmetry

dihedral D_{2h} symmetry

Earlier work on the D – center:
Ch. Zhang et al. (JILA and TU Wien), Nature **633**, 63 (2024)
J.S. Higgins et al., Phys. Rev. Lett. **134**, 113801 (2025)
T. Ooi et al., Nature **650**, 72 (2026)

And on the O – center:
T. Hiraki et al. (U. Okayama and TU Wien), arXiv:2509.00041

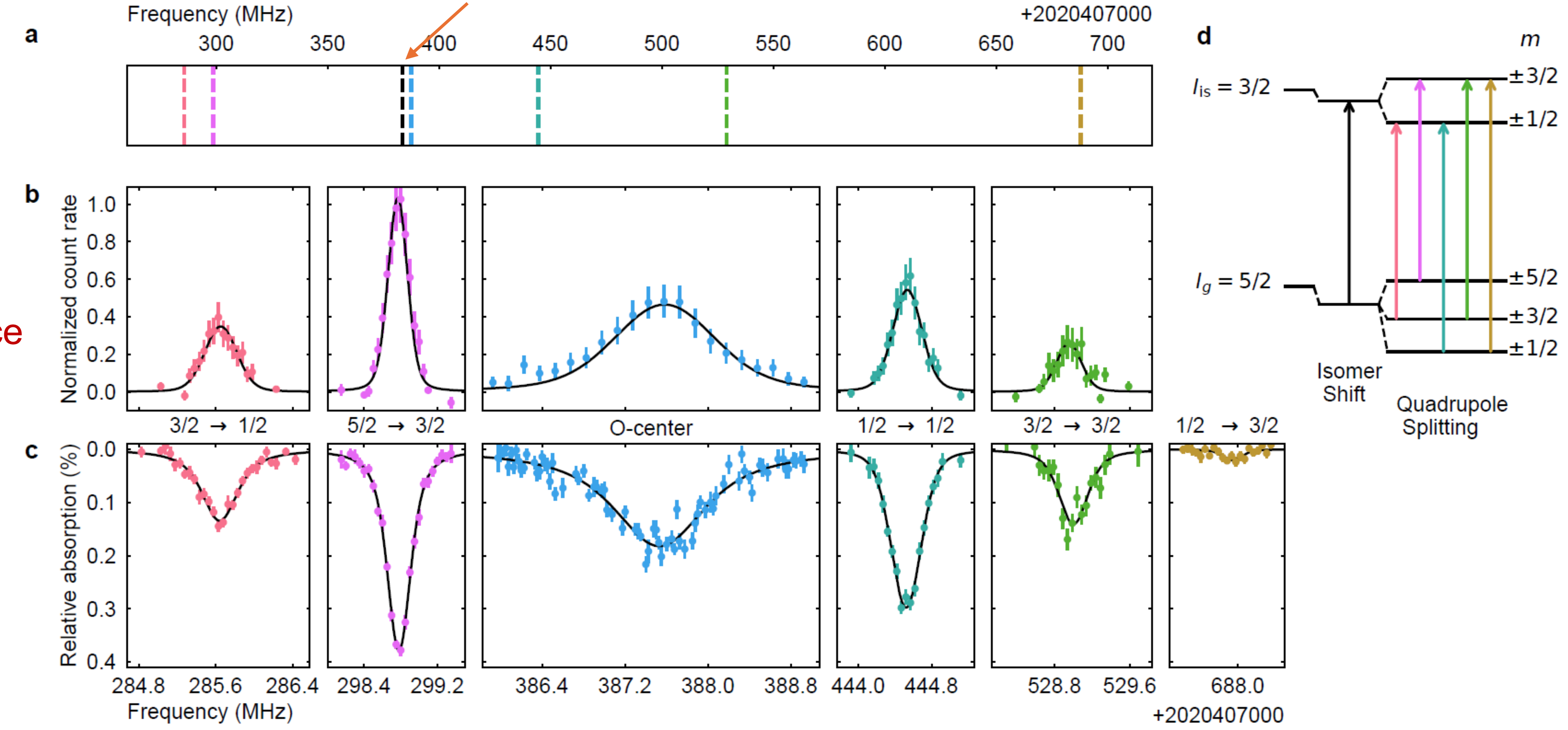
Fluorescence and absorption spectroscopy of the Th-229 nuclear transition

Signals from two Th-centers, O (cubic O_h symmetry)
and D (Th-dimer, dihedral D_{2h} symmetry, resolved quadrupole splitting)

Isomer shift: D -- O

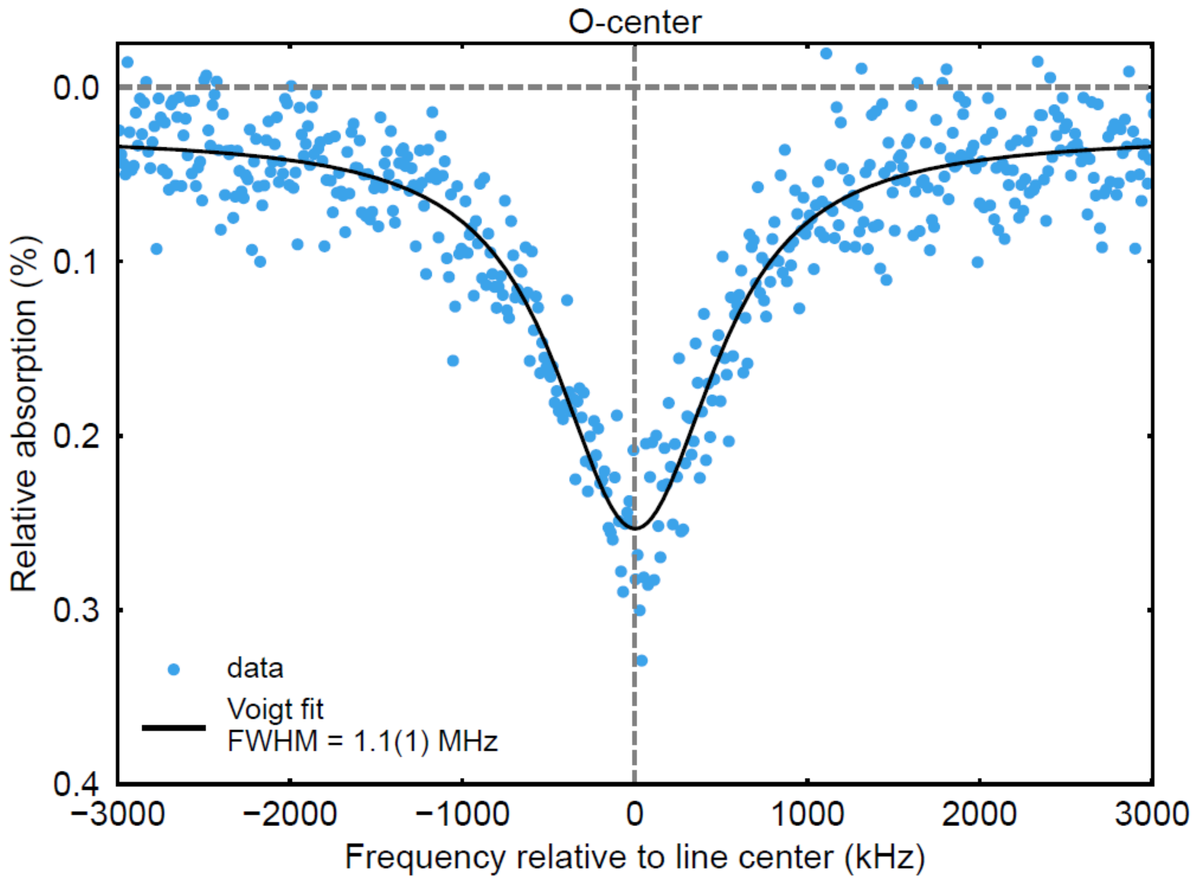
Fluorescence

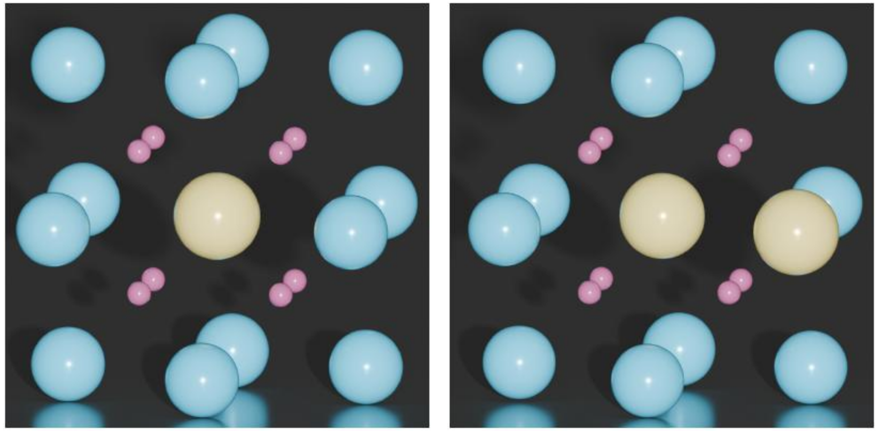
Absorption



I. Morawetz et al., arXiv: 2604.16640

Absorption spectrum of the O - center



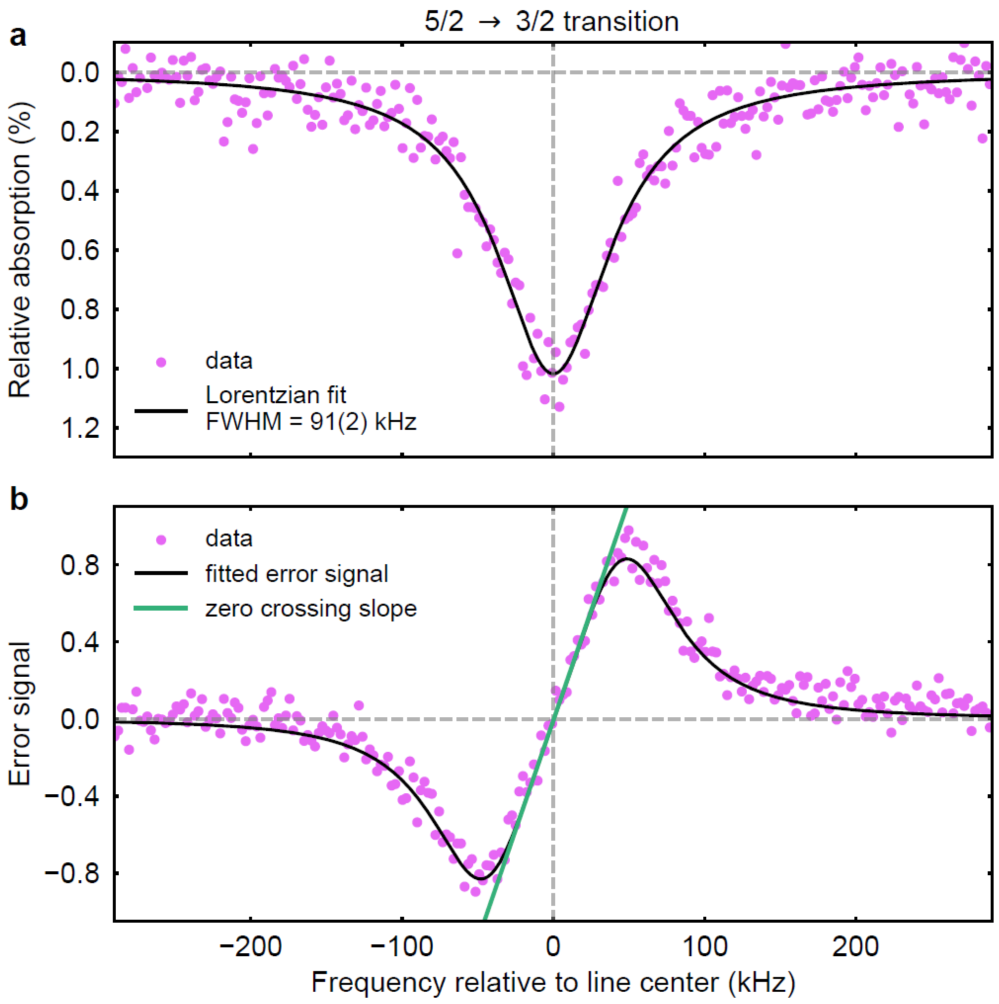


O - center D - center

Laser linewidth: < 100 kHz
Static electric crystal field gradient < 0.1 V/Å^2, (D- center: ≈ 100 V/Å^2)

Can the quadrupole components be resolved in a crystal with lower doping?
The O-center promises lines that are less sensitive to the lattice spacing.

Absorption signal of D-center 5/2 → 3/2 transition and error signal for laser lock

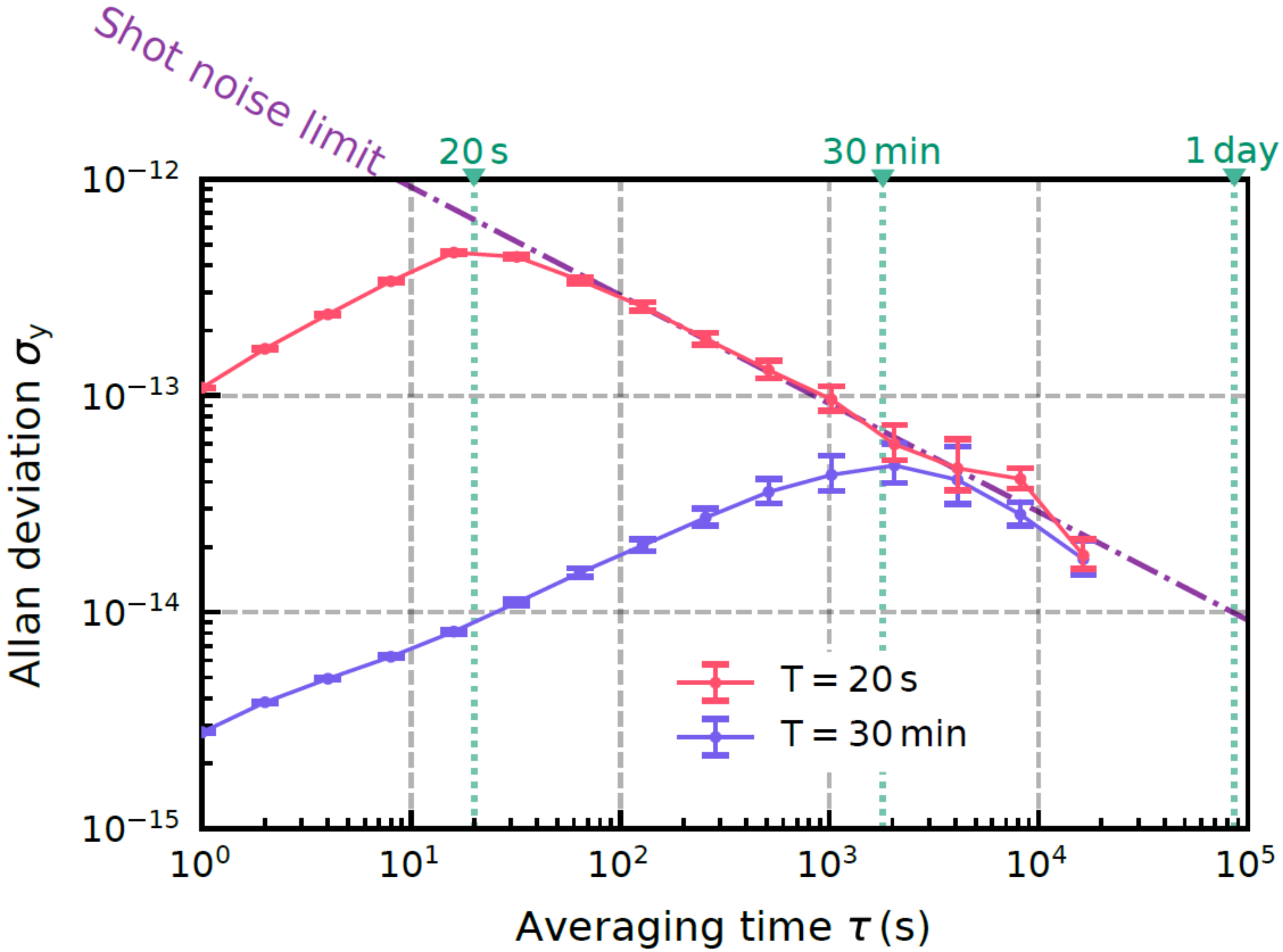


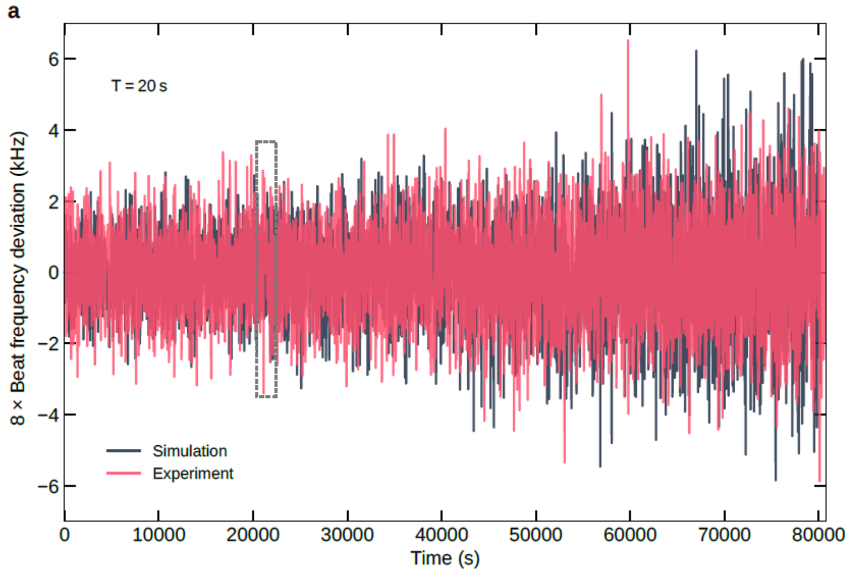
Narrowest linewidth observed in this crystal: 91 kHz.

Error signal for laser lock:
Frequency modulation at 10 Hz with 90 kHz amplitude,
Photon counting PMT signal.
(3 s per point).

I. Morawetz et al., arXiv: 2604.16640

Closing the feedback loop: 24 h unattended clock operation

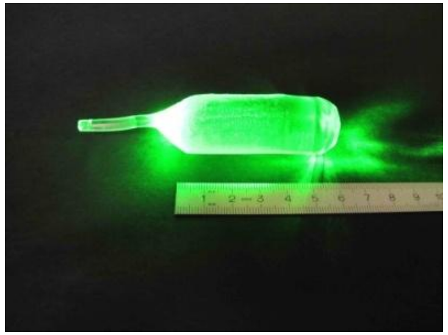




Data can be used to constrain models of ultralight dark matter.

L. Toscani De Col et al., arXiv:2606.04997

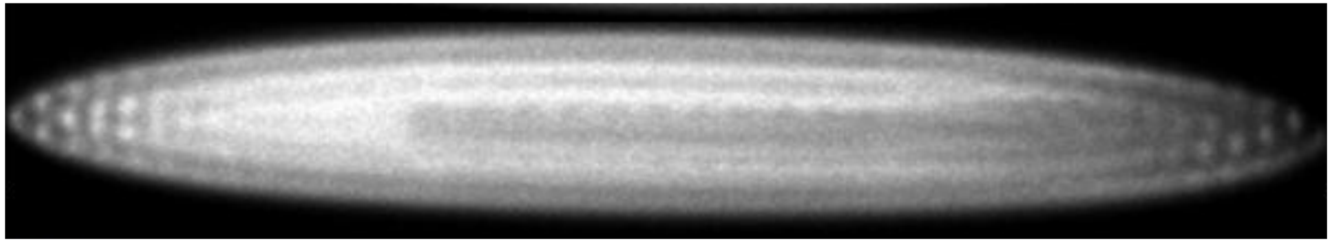
Th^{4+} dopant ions in solids



TU Wien, Th:CaF_2

T=100 – 300 K (so far)
n≈10^{15}
Electric crystal field gradients and magnetic spin-spin interaction (F-19)

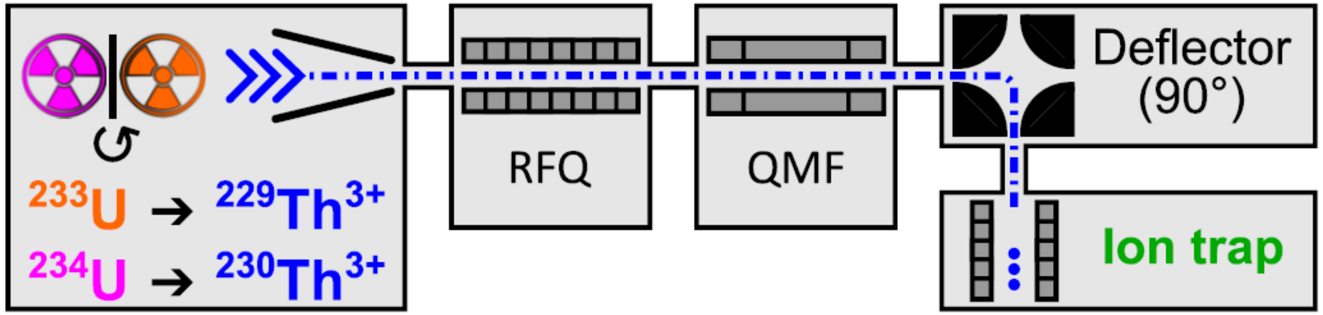
Laser-cooled Th^{3+} ions in a radiofrequency trap



PTB: $^{88}Sr^{+}$ (fluorescing) and $^{229}Th^{3+}$ (dark)

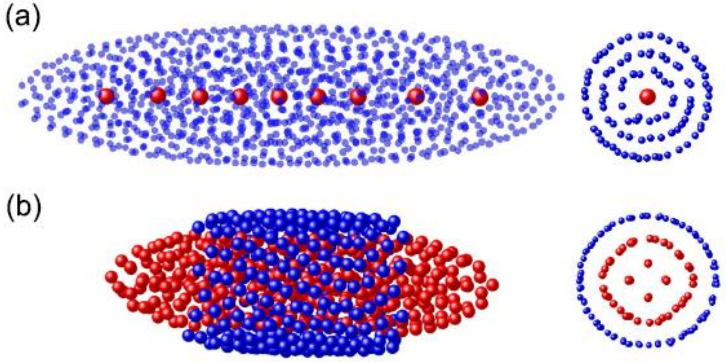
T ≈ mK
n≈100
Electric field of trap and neighboring ions

$^{229}Th^{3+}$ trapped recoil ions from ^{233}U, sympathetically cooled with $^{88}Sr^{+}$

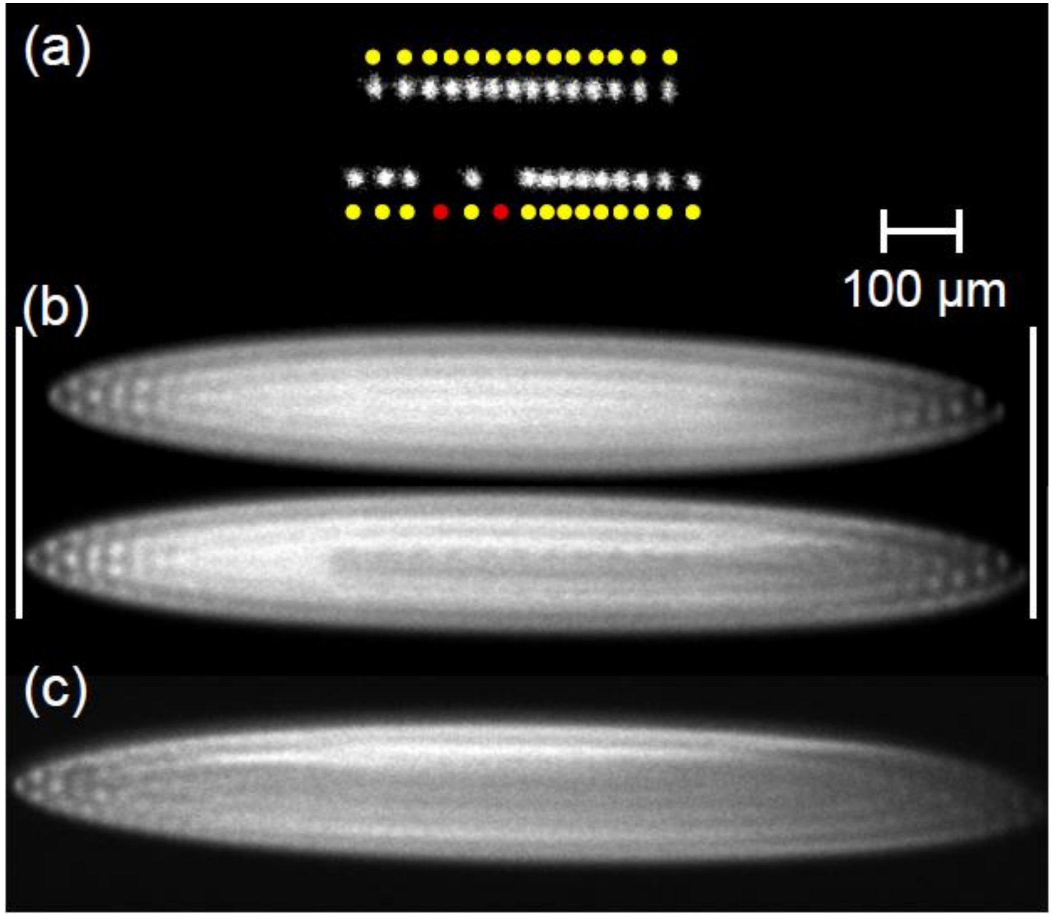


^{229}Th recoil ions include ≈2% isomers ^{229m}Th

^{230}Th: reference isotope with $I=0$



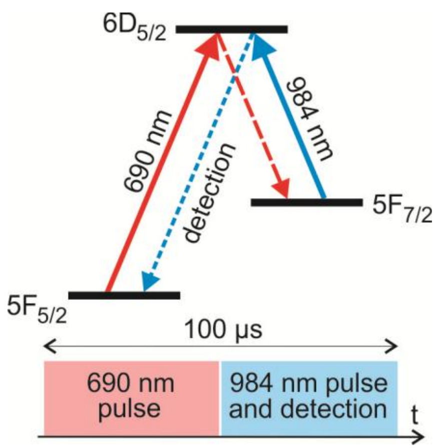
Simulated Coulomb crystals: blue Sr^{+}, red: Th^{3+}

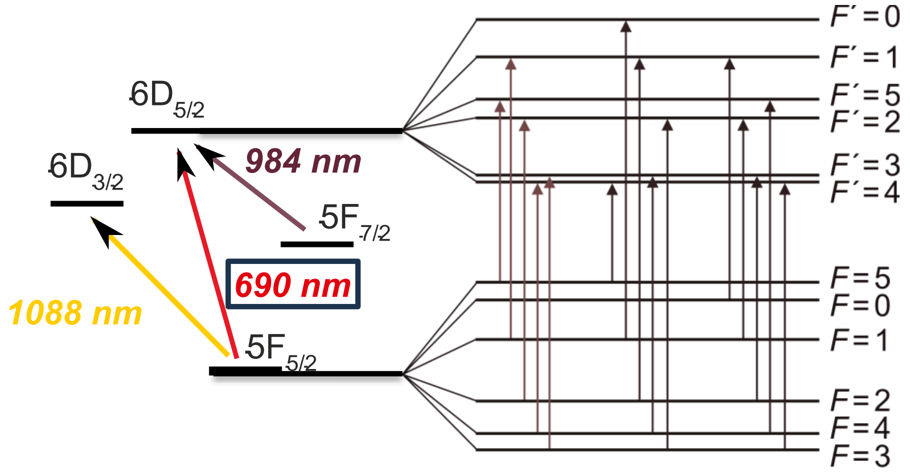


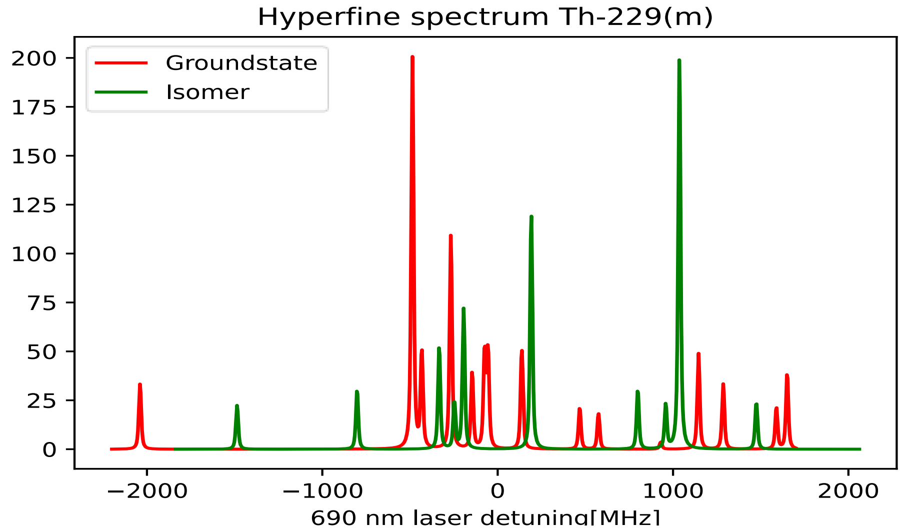
Fluorescence of Sr^{+}, Th^{3+} appear dark

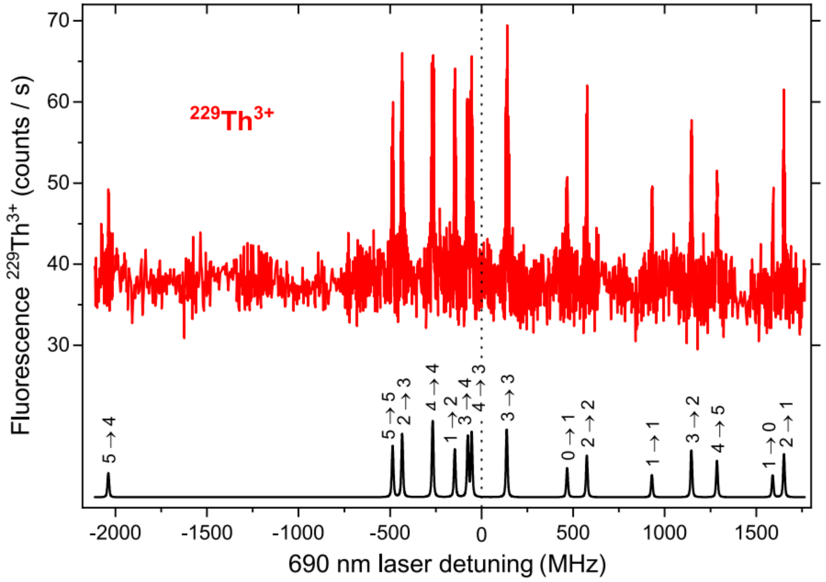
G. Zitzer, J. Tiedau , M. V. Okhapkin , K. Zhang, C. Mokry, J. Runke, Ch. E. Düllmann , E. Peik,
Phys. Rev. A **109**, 033116 (2024)

Hyperfine spectroscopy of $^{229}Th^{3+}$ sympathetically cooled with Sr^+

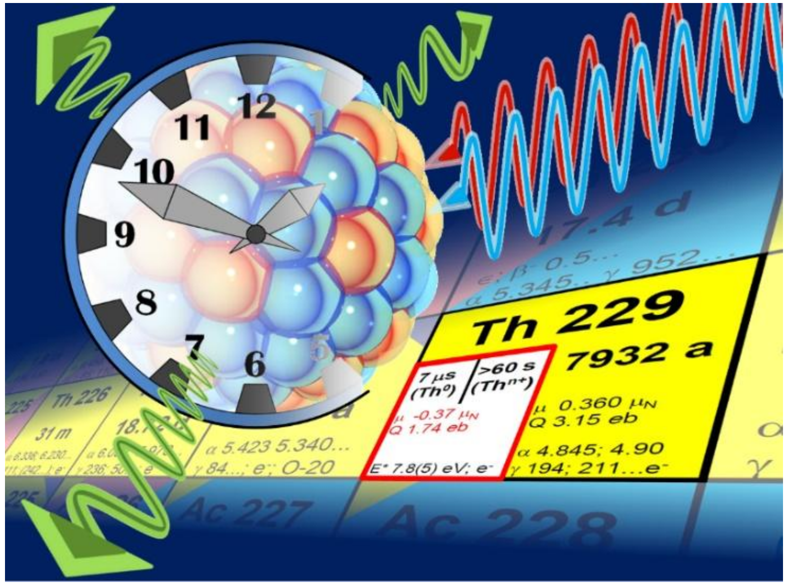








G. Zitzer, J. Tiedau, Ch. E. Düllmann, M. V. Okhapkin, E. Peik, Phys. Rev. A **111,** L050802 (2025)



What‘s next?

- Hz-level-linewidth continuous-wave VUV lasers
- Dynamics and lineshapes in Th-doped crystals
- New materials, different host crystals
- Exciting the nuclear transition in trapped Th ions
- Th-229 nuclear clocks in tests of fundamental physics

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V. Lal
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V. Petrov, MBI Berlin

European Research Council
Established by the European Commission

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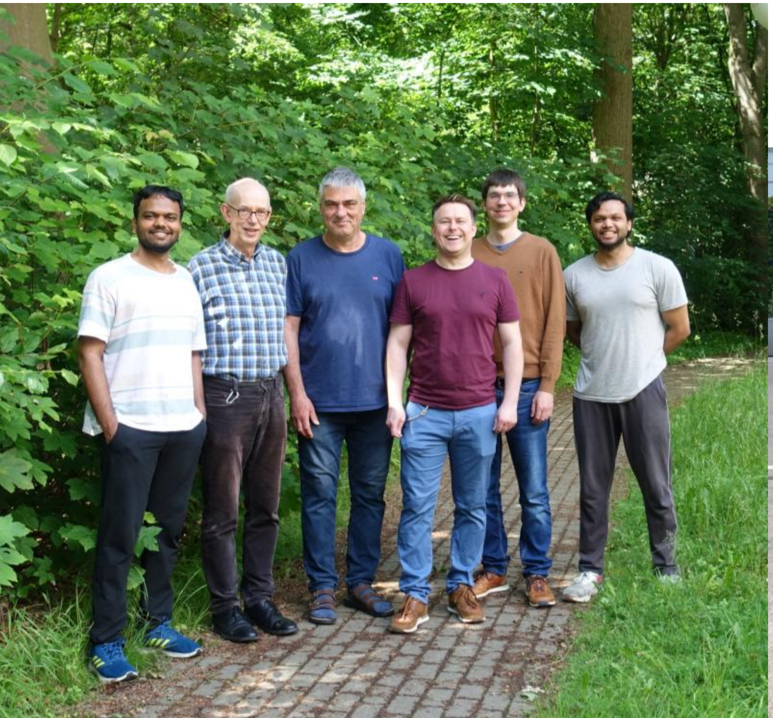
A. Grüneis
M. Pimon
T. Schumm



Th-229 nuclear moments (ground state) from HFS measurements

Ab initio atomic structure calculations:
* S. G. Porsev, M. S. Safronova, and M. G. Kozlov, PRL 127, 253001 (2021)

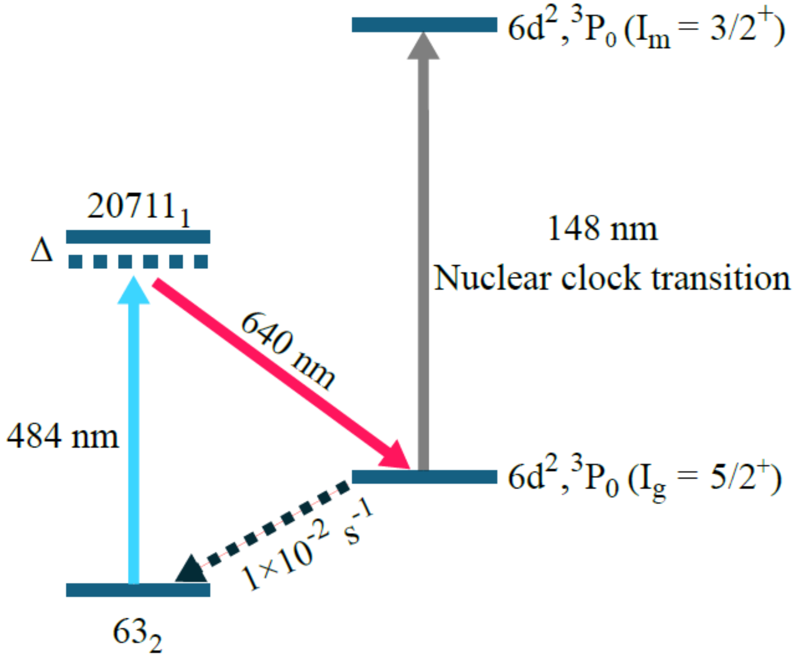
	C. J. Campbell et al, PRL. 106, 22301 (2011)		This work		From ab initio atomic structure calculations*	
State	A [MHz]	B [MHz]	A [MHz]	B [MHz]	μ/μ_N	Q/e [10^{-28} m^2]
$5F_{5/2}$	82.2(6)	2269(6)	82.0(2)	2270.3(1.8)	0.365(3)	3.11(5)
$6D_{5/2}$	-12.6(7)	2694(7)	-12.9(3)	2695.7(1.9)	0.31(7)	3.10(4)

The obtained magnetic moment $\mu = 0.365(3)\ \mu_N$ for the $5F_{5/2}$ level agrees well with the value $\mu = 0.366(6)\ \mu_N$ from *.

The electric quadrupole moments for the $5F_{5/2}$ and the $6D_{5/2}$ levels agree well with $Q/e = 3.11(2) \cdot 10^{-28}\ m^2$ *, obtained from the weighted average of four states in $^{229}Th^{3+}$.

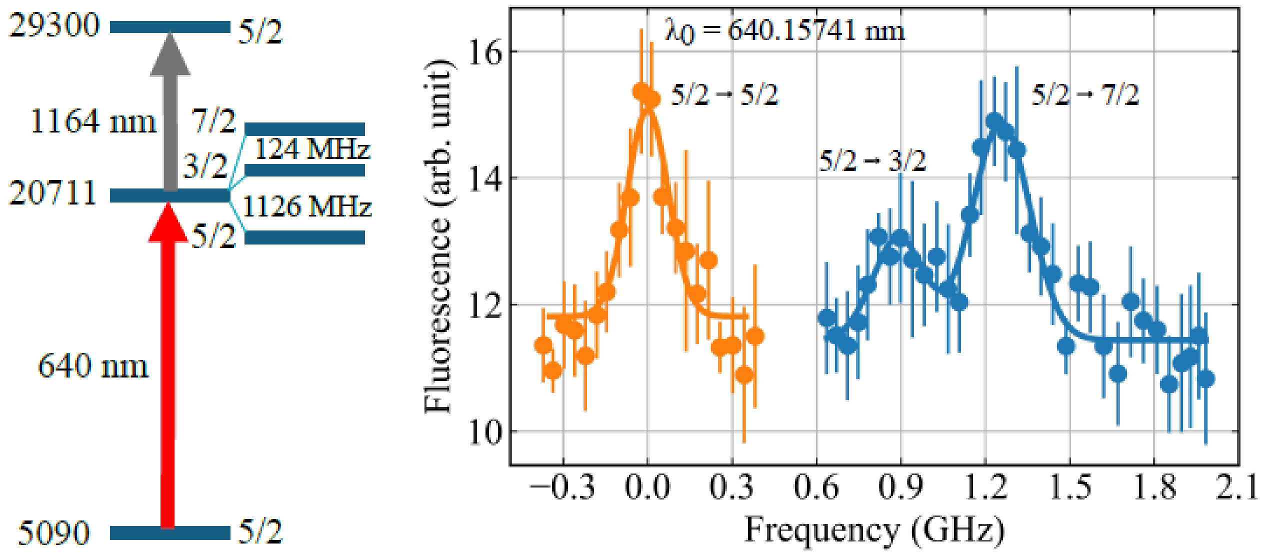
G. Zitzer, J. Tiedau, Ch. E. Düllmann, M. V. Okhapkin, E. Peik, Phys. Rev. A **111,** L050802 (2025)

J = 0 metastable state of Th^{2+} for a hyperfine-free nuclear clock



A metastable level (τ=96 s*) at 5090 cm^{-1} may be used to excite the nucleus independent from the leading hyperfine interactions.

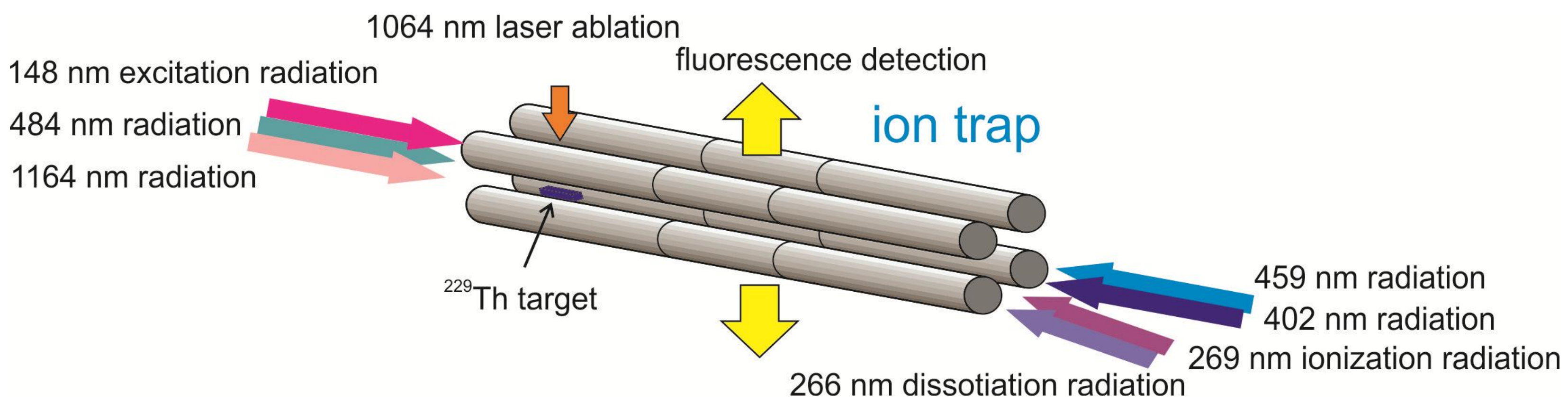
Fluorescence detection of $^{229}Th^{2+}$ in the *J=0* level



S. Sagar Maurya, V. Lal, J. Tiedau, M. V. Okhapkin, and E. Peik, arXiv:2605.07802

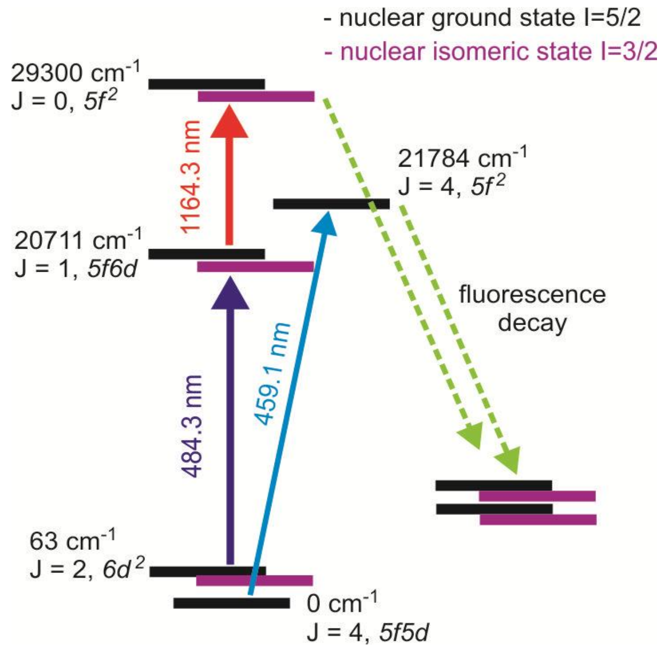
* M. S. Safronova, U. I. Safronova, C. W. Clark, Phys. Rev. A 90, 032512 (2014).

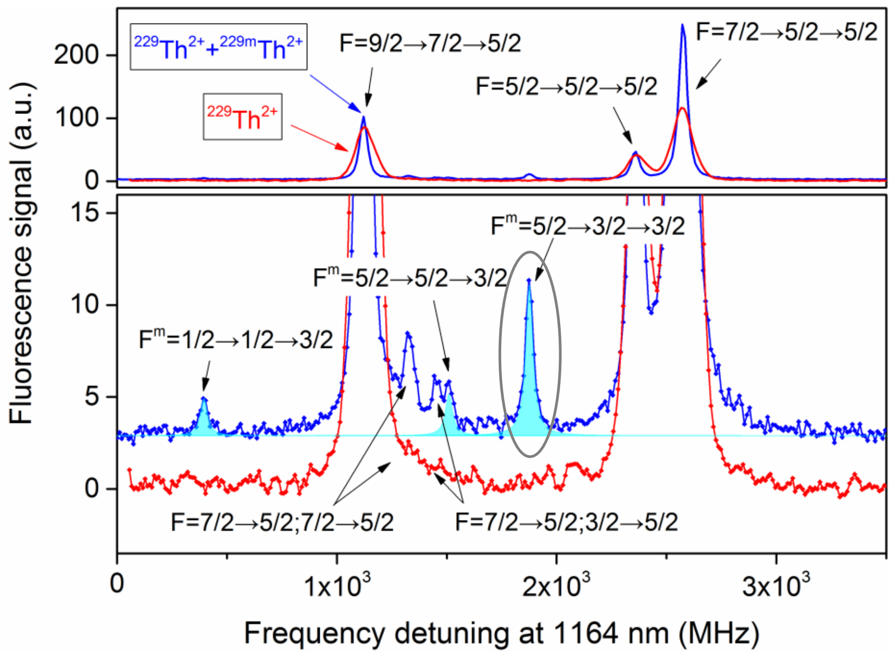
Search for nuclear laser excitation in trapped $^{229}Th^{2+}$ ions



Th^{+} ions produced by laser ablation, Th^{2+} by photoionization of Th^{+}.

Cooled with Ar buffer gas (300 K).

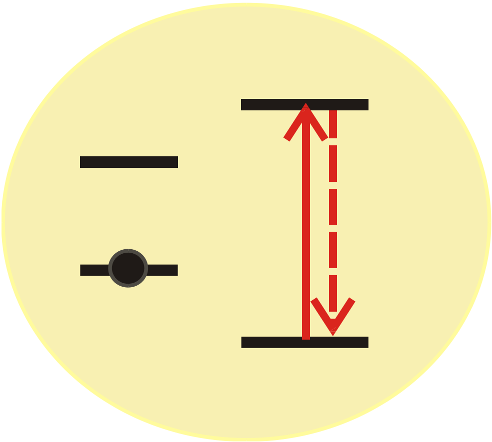




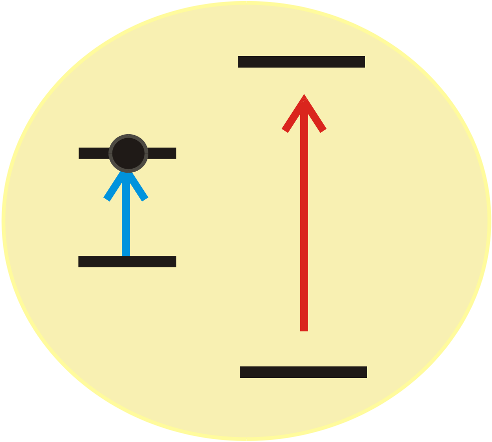
Detection:
Resonance of the isomer in HFS of electronic two-step excitation, free from Doppler broadening.

See: J. Thielking et al., Nature 556, 321 (2018)

Trapped ions: Detection of the Nuclear Excitation in Nuclear-Electronic Double-Resonance

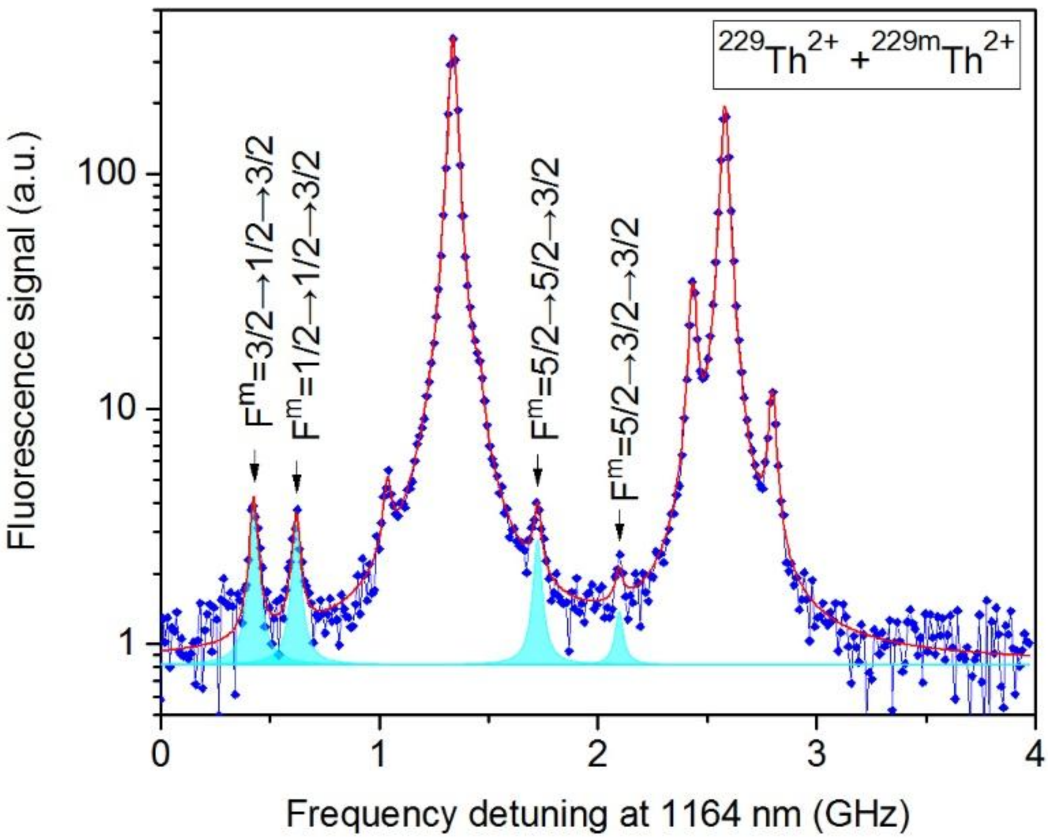


Nucleus in the ground state; laser-induced fluorescence from the shell.



Laser excitation of the nucleus; change of hyperfine structure detected in intensity or polarisation of fluorescence.

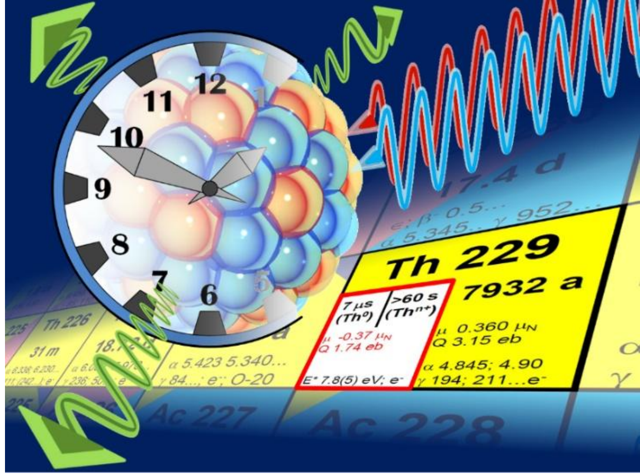
$^{229}Th^{2+}$: Detection of the isomer in HFS



Analog of Dehmelt‘s „electron shelving“
Observation of „quantum jumps“ in the single-ion fluorescence
Coupled degrees of freedom and long nuclear coherence time

E. Peik, Chr. Tamm, Europhys. Lett. **61**, 181 (2003)

J. Thielking et al. (PTB & LMU), Nature **556**, 321 (2018)



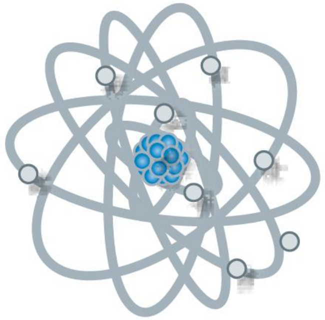
Nuclear Clock:

Oscillator that is frequency-stabilized
to a nuclear (γ-ray) transition

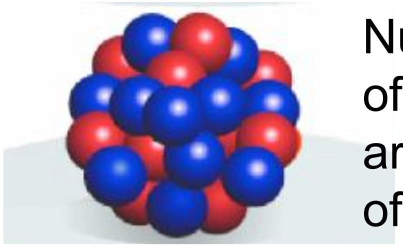
Higher accuracy:

The nuclear clock allows for a choice of a suitable electronic state for the interrogation of the nuclear resonance, for example in laser-cooled ions, Th^{3+}.

Example: Electric fields, Stark effect:



Electrons shield the nucleus (from homogeneous, static fields)

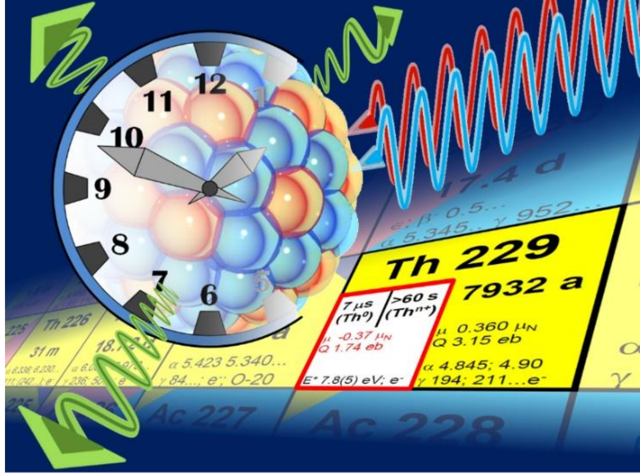


Nuclear polarizability is orders of magnitude smaller; electronic effects are common-mode for both states of the transition.

Analyses of best suited electronic configurations:

E. Peik, Chr. Tamm, Europhys. Lett. **61**, 181 (2003) → low values of electronic angular momenta J=0 or J=1/2

C. J. Campbell et al., PRL **108**, 120802 (2012) → stretched states F=J+I, aligned electronic and nuclear momenta



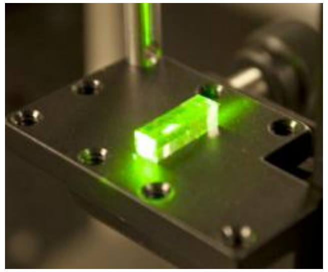
Nuclear Clock:

Oscillator that is frequency-stabilized
to a nuclear (γ-ray) transition

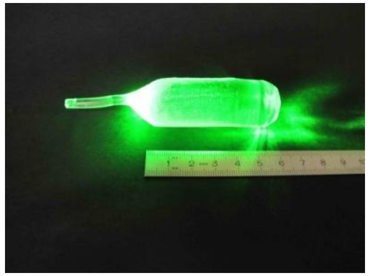
Higher stability:

In a Mössbauer solid state nuclear clock, many absorbers may be interrogated ($>10^{15}$ instead of $\approx 10^{0}$ (ion trap) or $\approx 10^{4}$ (optical lattice)). Systematics: Crystal field shifts.

Proposed at PTB, UCLA, TU Wien

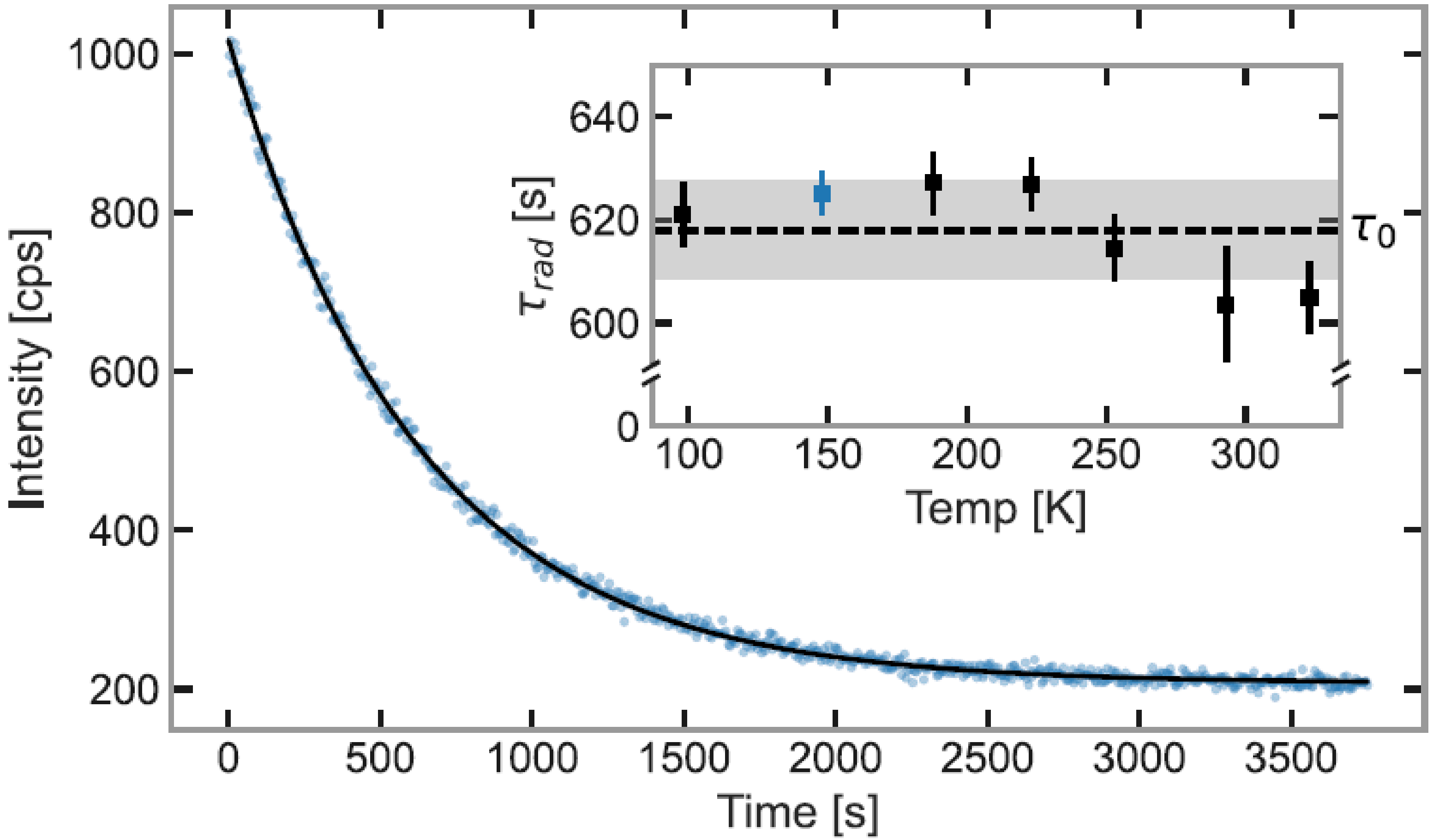


UCLA, LiCAF



TU Vienna, CaF_2

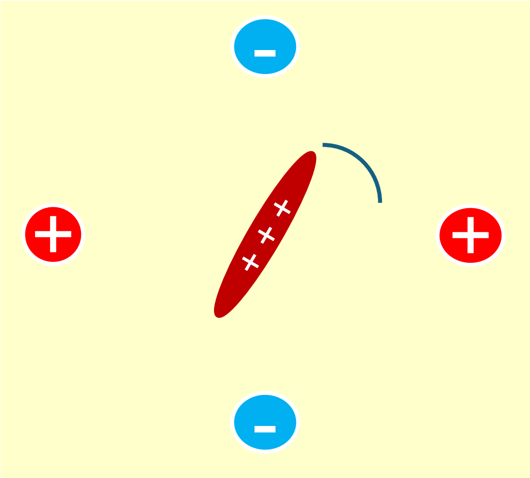
Fluorescence decay curves



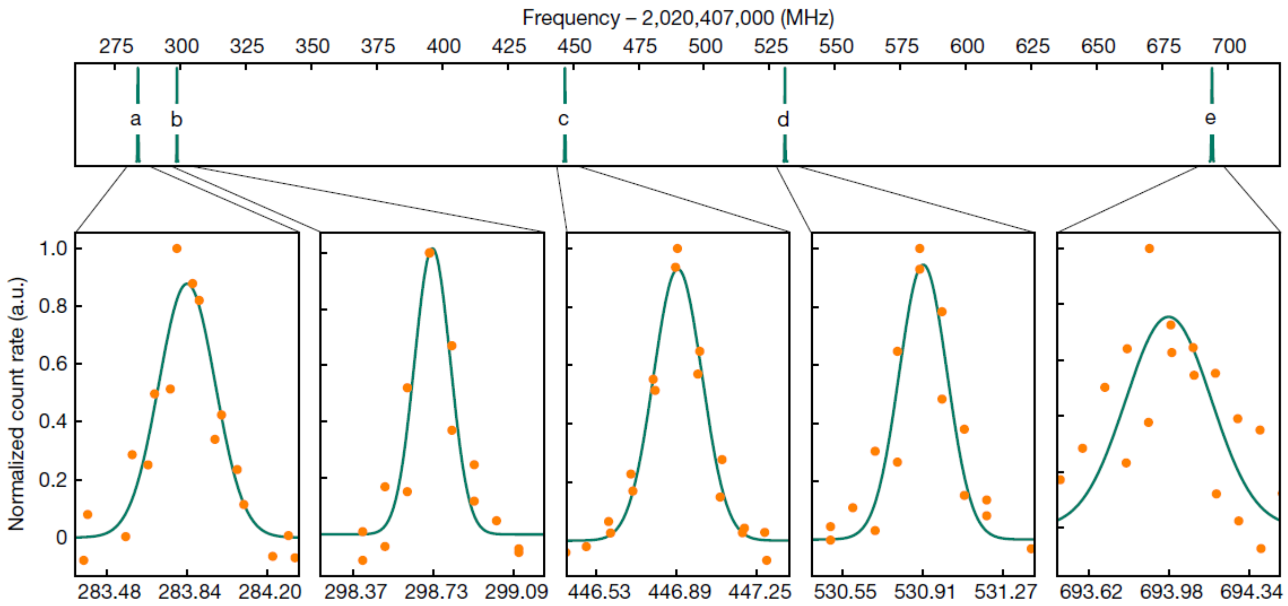
Decay time constant: 618(9) s:

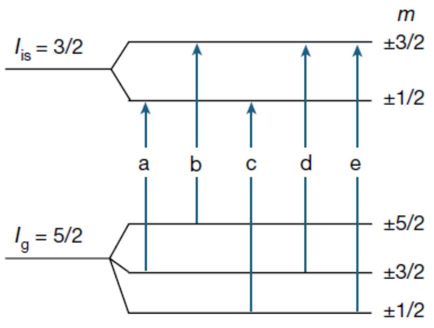
- Identical for differently doped X2 and C10 crystals
- Identical for C10 before and after refluorination with CF_4
- Independent of crystal temperature 100 – 320 K
- Expected to be enhanced by the mode density in the crystal $\propto n^3$ → isomer half-life 1740 s

Quadrupole splitting of the Th-229 nuclear resonance in CaF_2



Prolate nuclear charge distribution interacting with an electric field gradient.



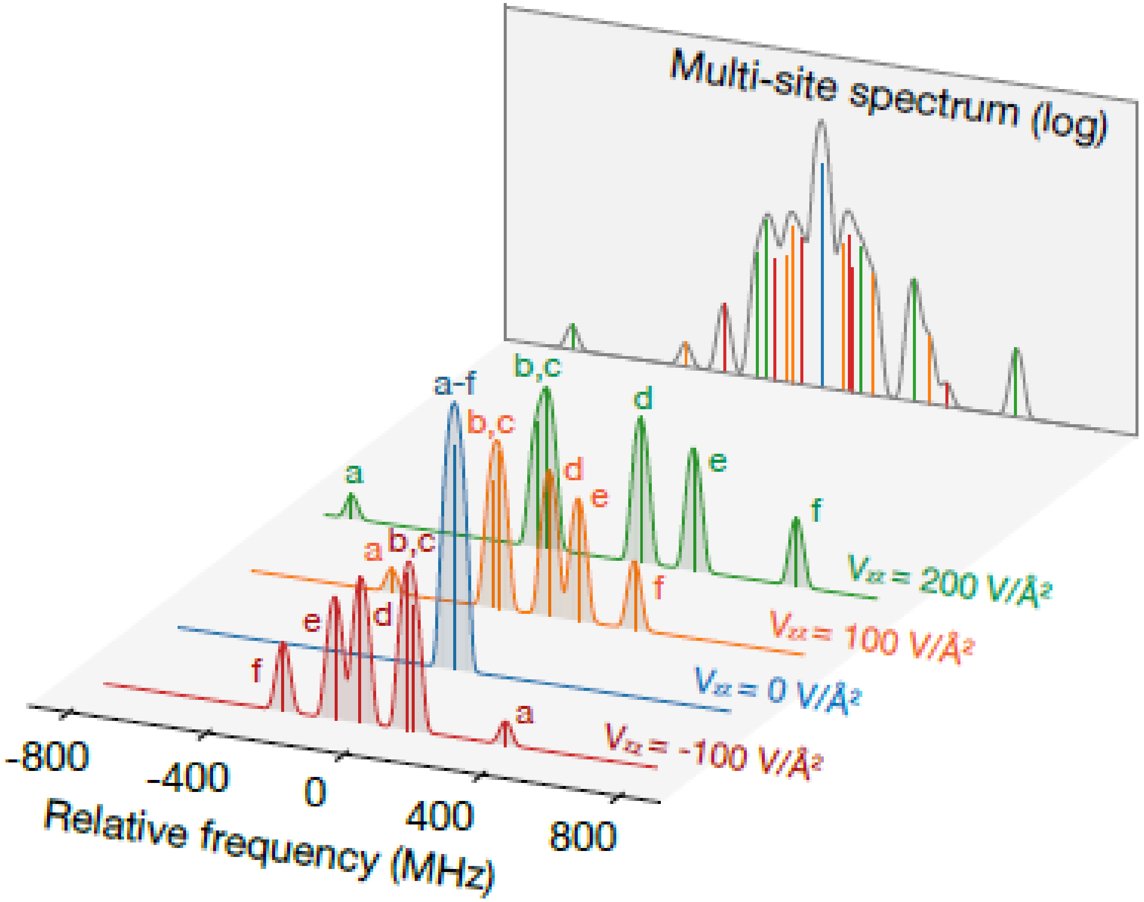


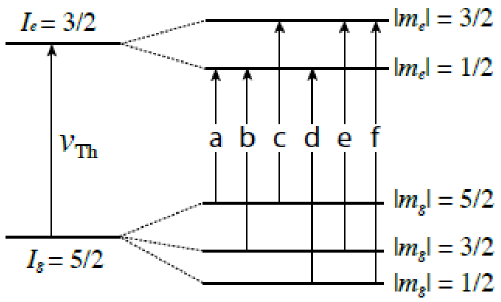
m_g	m_{is}	ν_{Th} (MHz)
3/2	1/2	2,020,407,283.847(4)
5/2	3/2	2,020,407,298.727(4)
1/2	1/2	2,020,407,446.895(4)
3/2	3/2	2,020,407,530.918(4)
1/2	3/2	2,020,407,693.98(2)

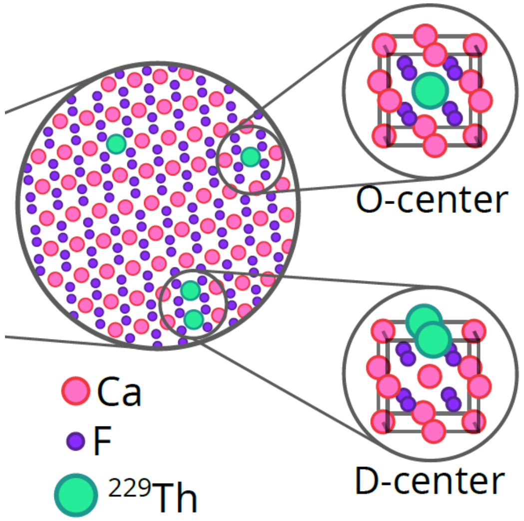
Measured with a VUV fs-frequency comb
Ch. Zhang et al. (JILA and TU Wien), Nature **633**, 63 (2024)

Different temperature shifts of the components:
J.S. Higgins et al. (JILA, TUW), Phys. Rev. Lett. 134, 113801 (2025)
T. Ooi et al., Nature 650, 72 (2026)

Quadrupole splitting of the Th-229 nuclear resonance for different Th-centers in CaF_2







Four distinct centers of Th in CaF_2 with different electric field gradients:
T. Hiraki et al. (Okayama, Wien), arXiv:2509.00041

DFT analysis: M. Pimon et al. (TU Wien)

Atomic / nuclear physics with Th-229 atomic ions in different charge states

Th^{+}	electronic bridge processes in excitation and decay (?)
Th^{2+}	two-photon excitation via coupling to an electronic level (?) J=0 metastable levels
Th^{3+}	direct laser cooling trapped-ion nuclear clock electron-nuclear double resonance detection
Th^{4+}	J=0 ground state quantum logic clock with sympathetic laser cooling (?) (dominant charge state of Th in solids)
Th^{35+}	magnetic dipole fine-structure-nuclear mixing
Th^{89+}	magnetic dipole HFS-nuclear mixing