Physikalisch-Technische Bundesanstalt
Braunschweig and Berlin
National Metrology Institute

Laser Excitation of the Thorium-229 Nucleus – Towards a Nuclear Clock

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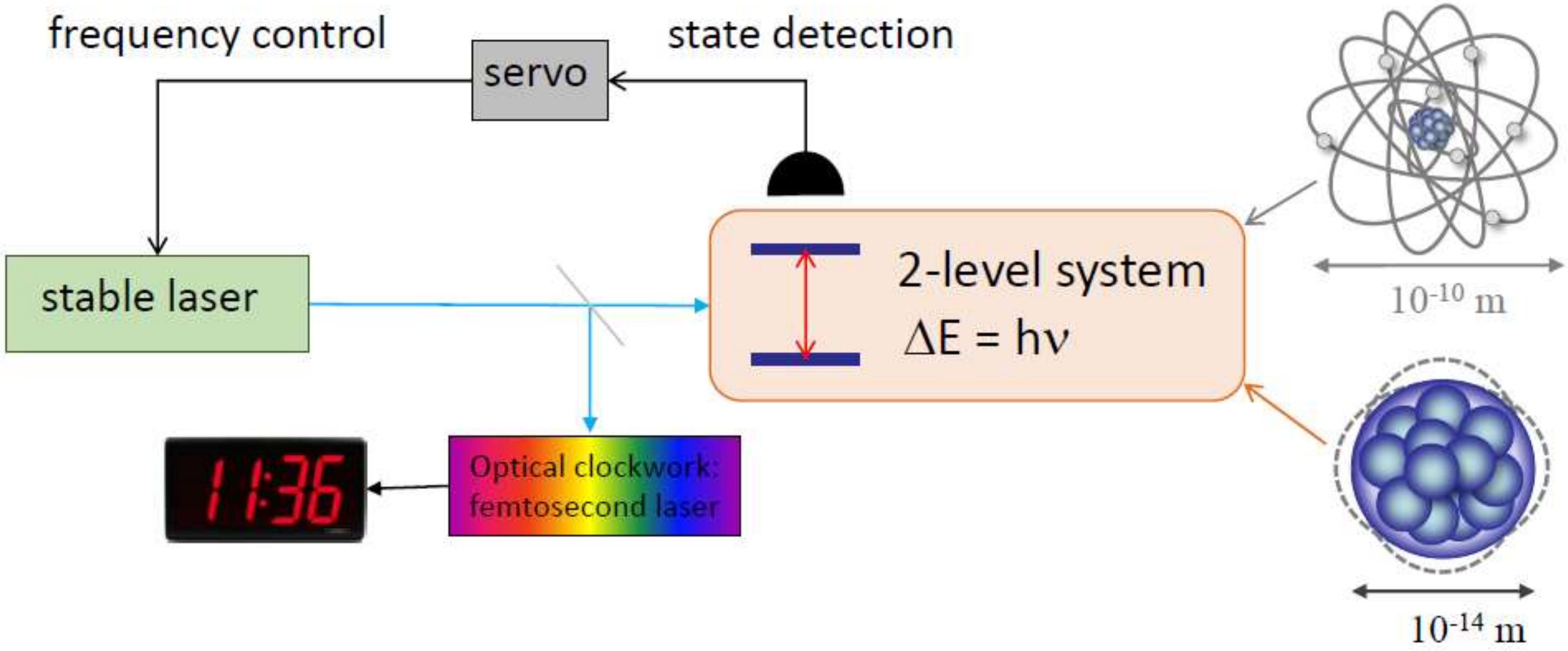
European Research Council
Established by the European Commission

841. Heraeus Seminar, Quantum Technologies, Steinbach, 1.- 4.9.2025

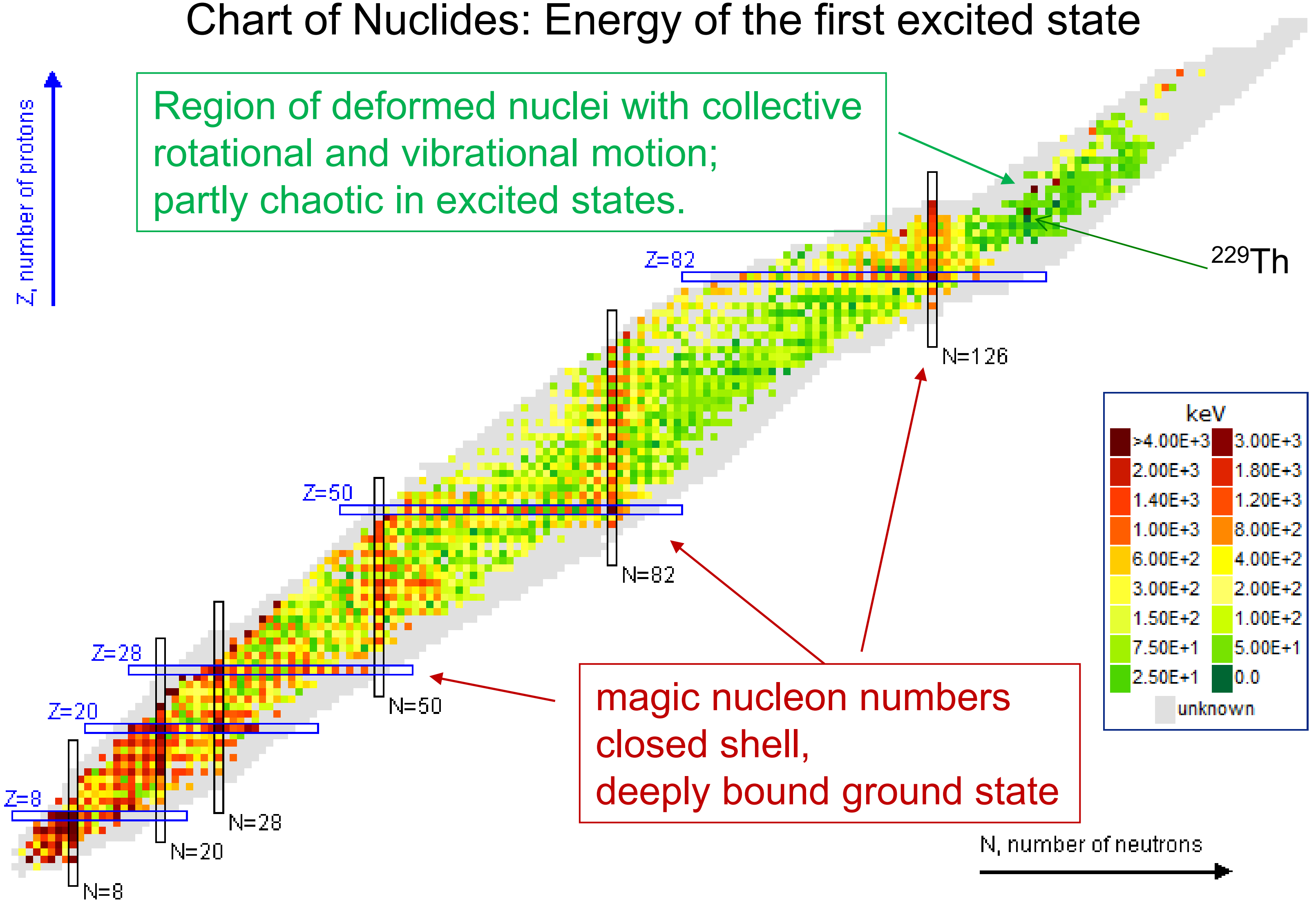
From the atomic to the nuclear clock



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)

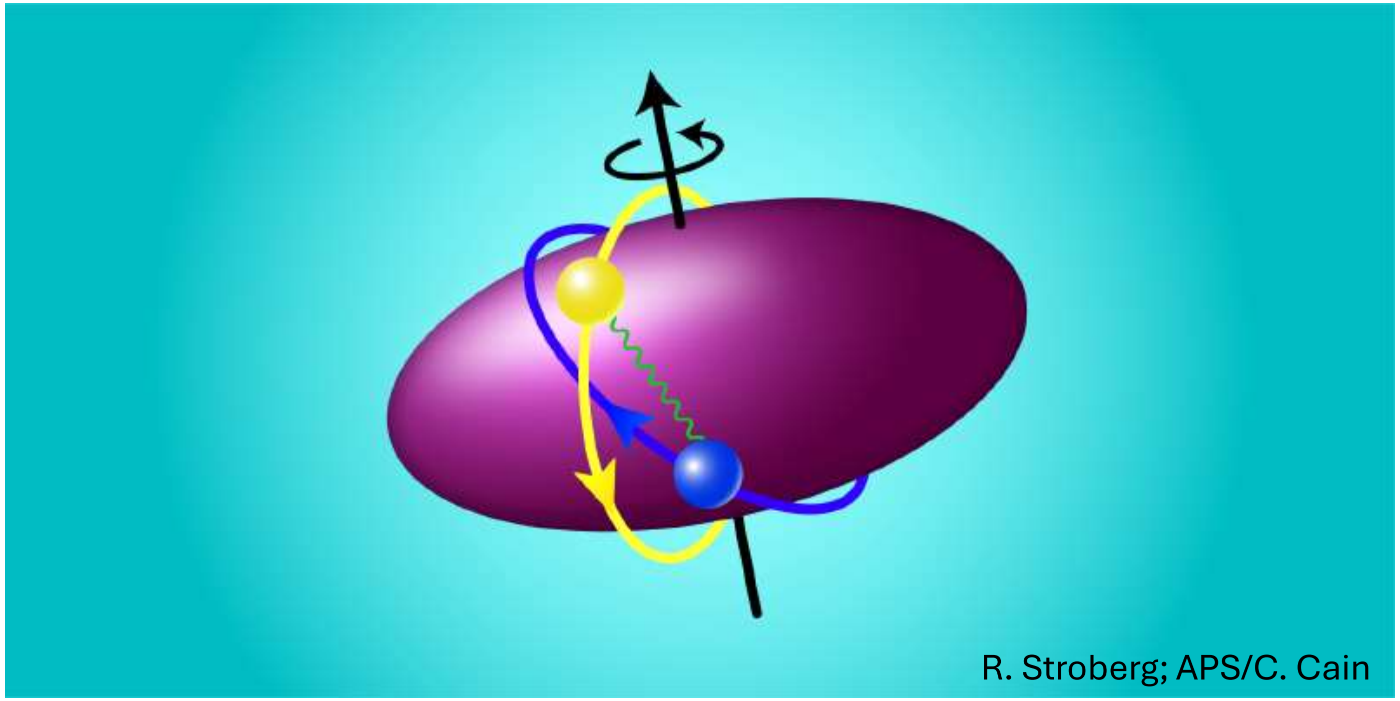
Chart of Nuclides: Energy of the first excited state



From: National Nuclear Data Center, Brookhaven National Laboratory

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)




Sven Gösta Nilsson

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

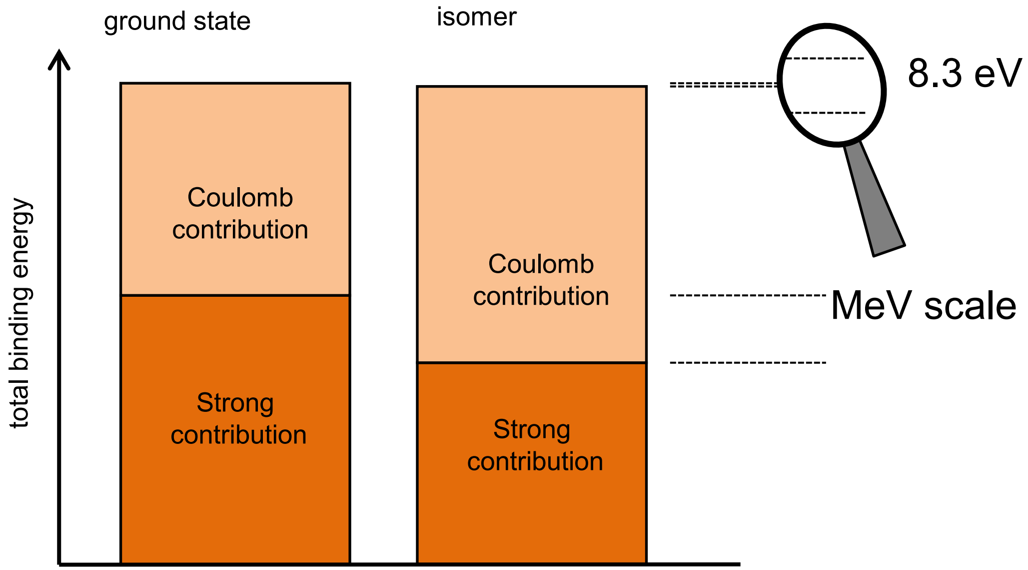
But we have measured a change in the rms charge radius:

$$\langle r^2_{229m} \rangle - \langle r^2_{229} \rangle = 0.0105(13)\ \mathrm{fm}^2$$

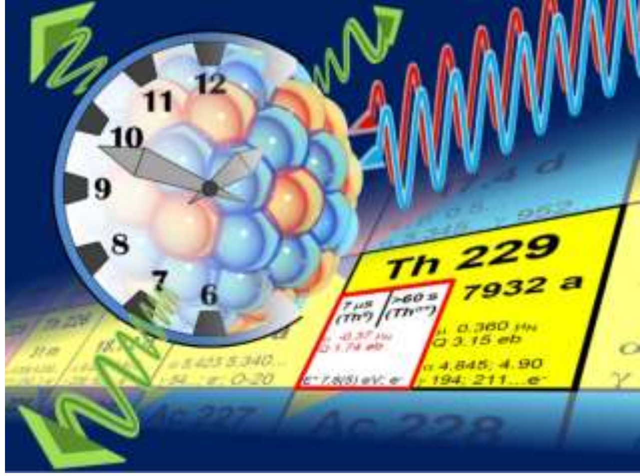
J. Thielking et al., Nature **556**, 321 (2018)

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:
Search for variations of fundamental constants
or other violations of the Einstein equivalence principle



Nuclear Clock:

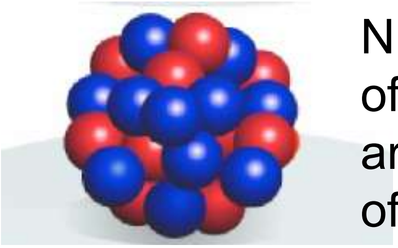
Oscillator that is frequency-stabilized
to a nuclear (γ-ray) transition

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:

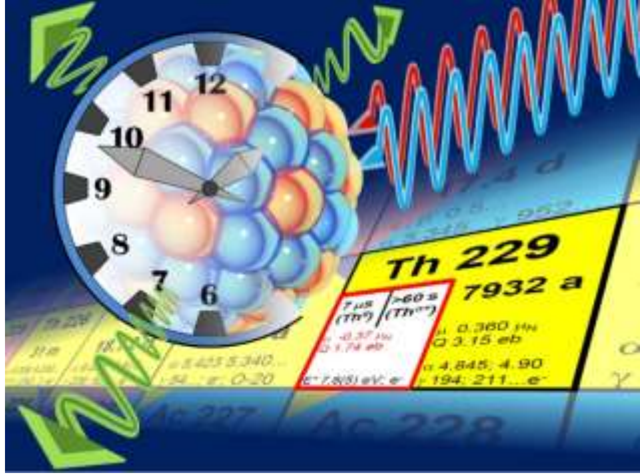
Electrons shield the nucleus
(from homogeneous, static fields)



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:

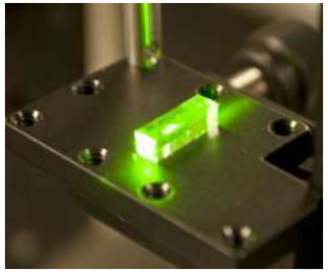


Oscillator that is frequency-stabilized
to a nuclear (γ-ray) transition

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



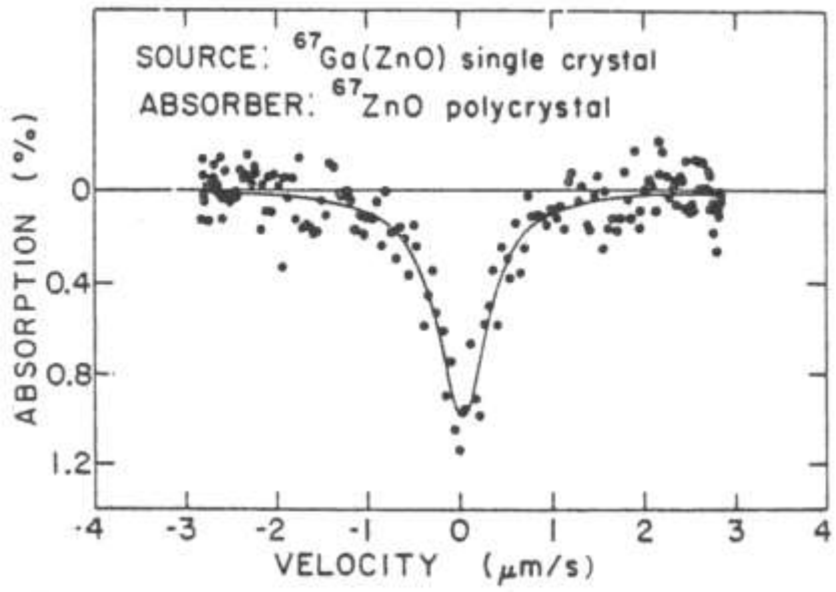
UCLA, LiCAF

TU Vienna, CaF_2

Nuclear gamma spectroscopy provides very high spectral resolution

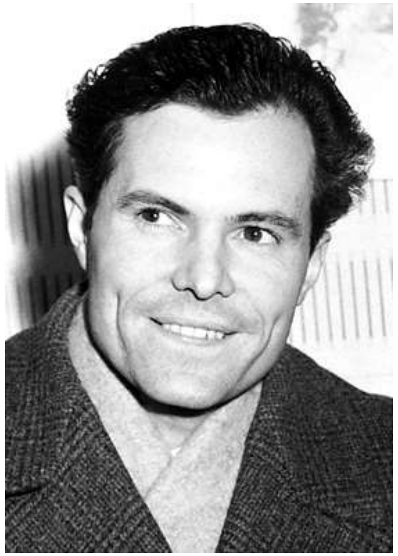
Mössbauer effect: Recoil-free resonance absorption of gamma radiation by nuclei in a crystal lattice

Mößbauer spectrum of the 93.3 keV resonance of Zn-67

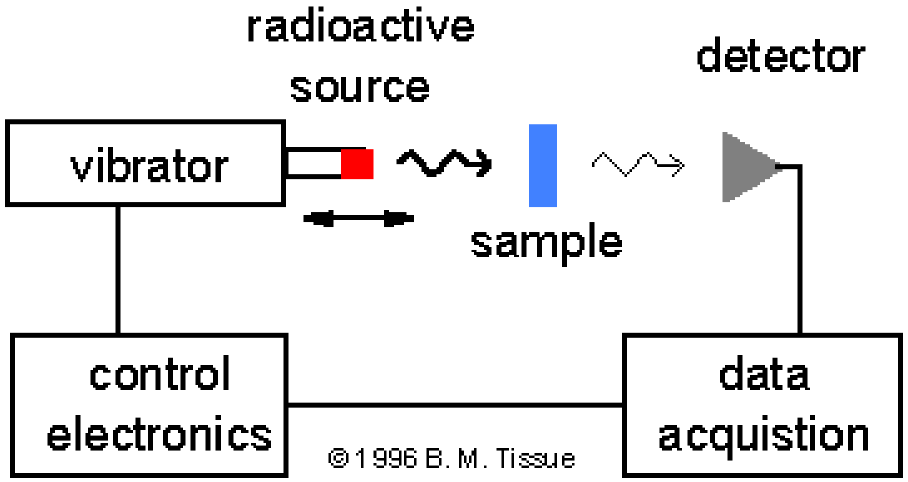


$\Delta\nu/\nu = 2.5 * 10^{-15}$

G. Perlow et al., 1974



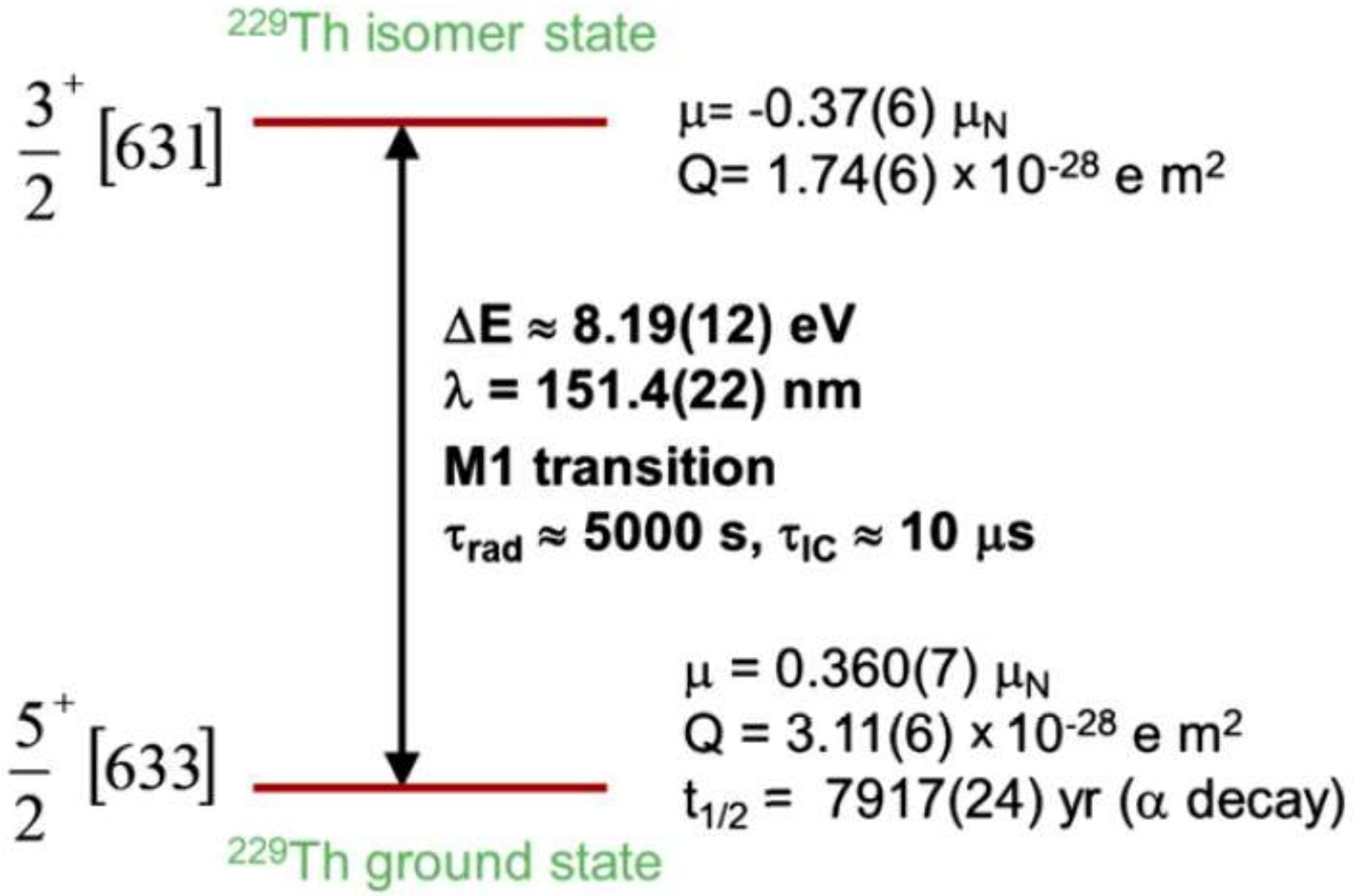
Rudolf Mößbauer, 1961
(Nobel Foundation)

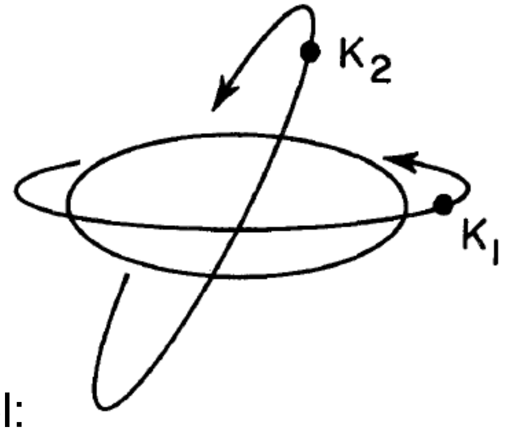


Nuclear Clock:
Replace the incoherent radioactive source with a tunable laser

The Th-229 low-energy isomer: State of knowledge in 2020

PTB





Nilsson Model:
Deformed core plus unpaired neutron

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

ERC Synergy Grant
Thorium Nuclear Clock
2020-2026

erc
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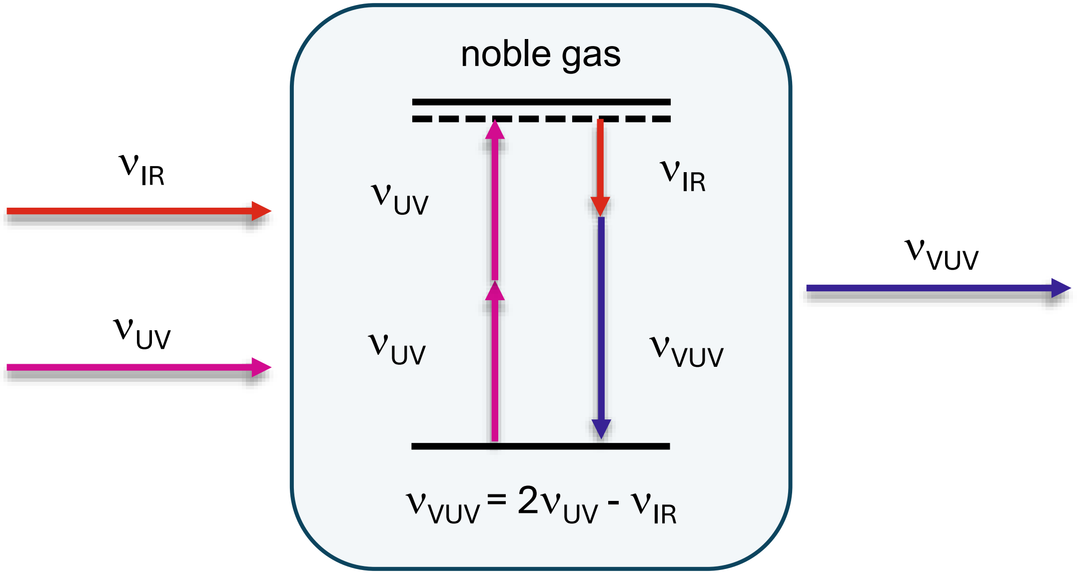
8-eV tunable VUV generation: Four wave mixing

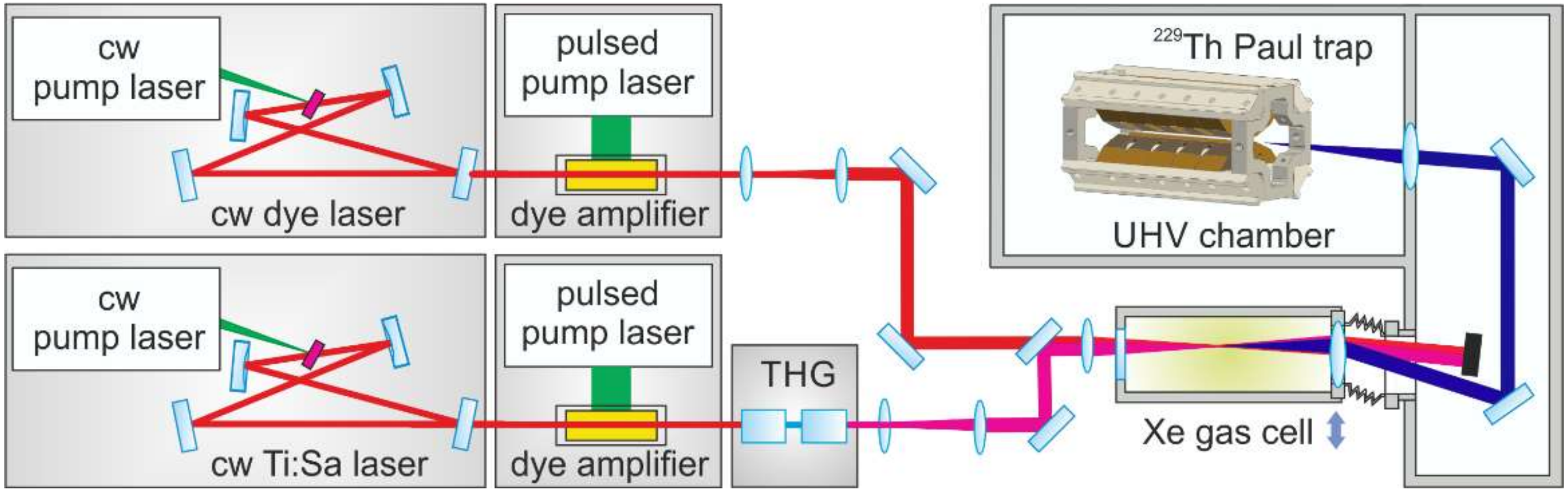
- Near-resonantly driving of 2-photon transition in noble gas.
- Supplying one photon with ν_{IR} yields fourth photon with $\nu_{VUV} = 2\nu_{UV} - \nu_{IR}$.
- Two-photon transition in Xe at 2 x 250 nm is suitable for VUV tunability from 167 nm to 148 nm, i.e. 7.42 eV to 8.38 eV.

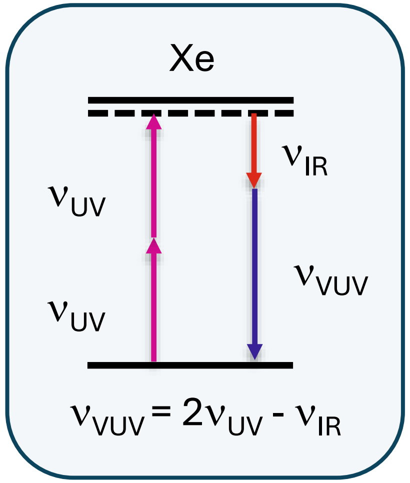


See e.g.: S. J. Hanna et al., Int. J. Mass. Spectrom. **279**,134 (2009)

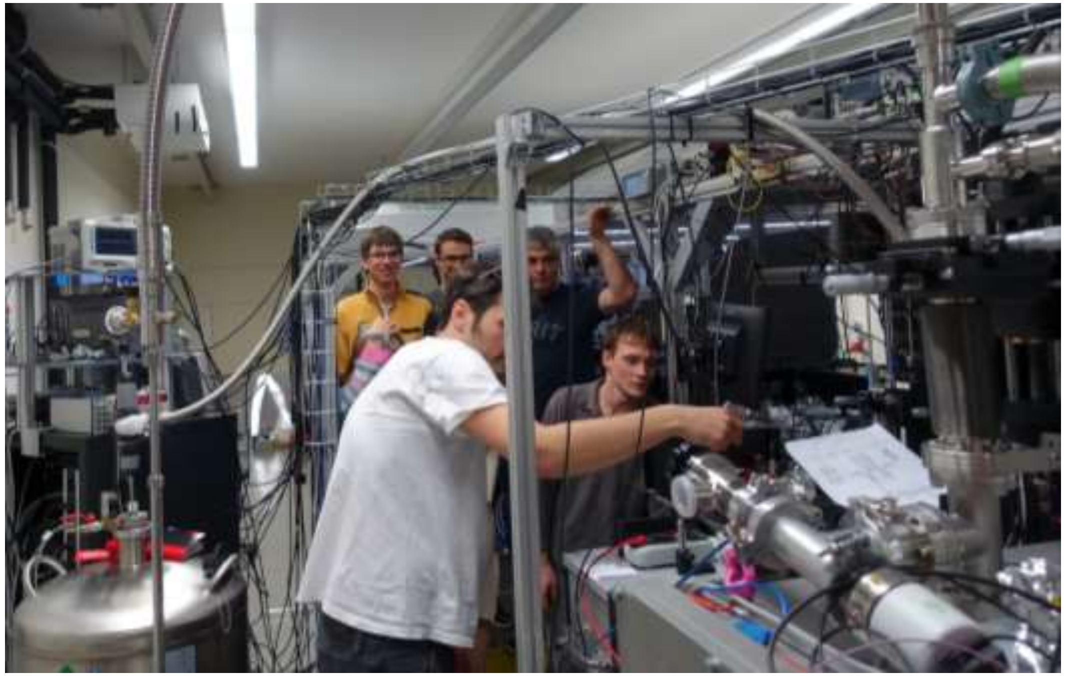
8-eV tunable VUV generation: Four wave mixing

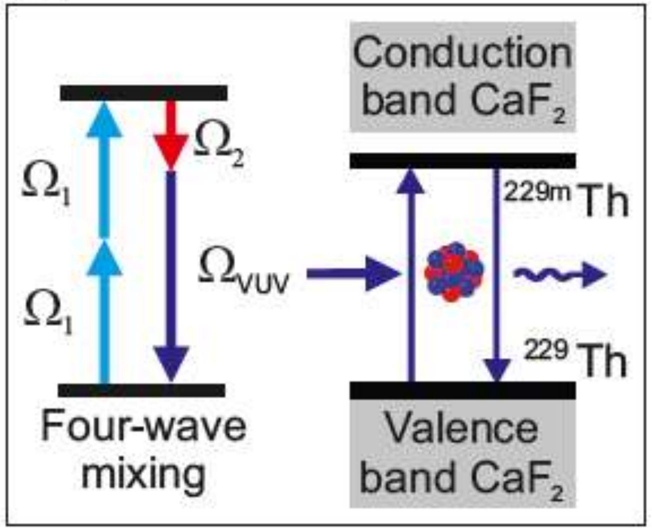
- Generating 148 nm VUV requires laser beams at 250 nm and 790 nm.
- Third order process needs high intensity to achieve suitable efficiency.
- Pulsed lasers (~10 ns, 30 Hz repetition rate) best compromise between VUV pulse energy ($>10^{13}$ photons/pulse) and linewidth (<10 GHz).
- Our setup:
  - Two cw Ti:Sa ring lasers as seed.
  - Pulsed dye amplifiers (~60 mJ/pulse, 30 Hz repetition rate).

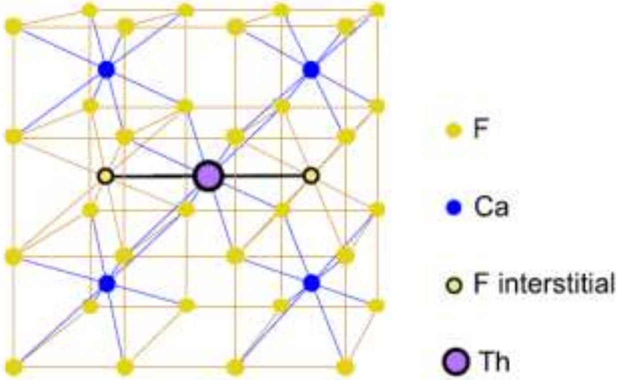




PTB - TU Wien cooperation:
Laser excitation of ^{229}Th-doped calciumfluoride crystals







European Research Council
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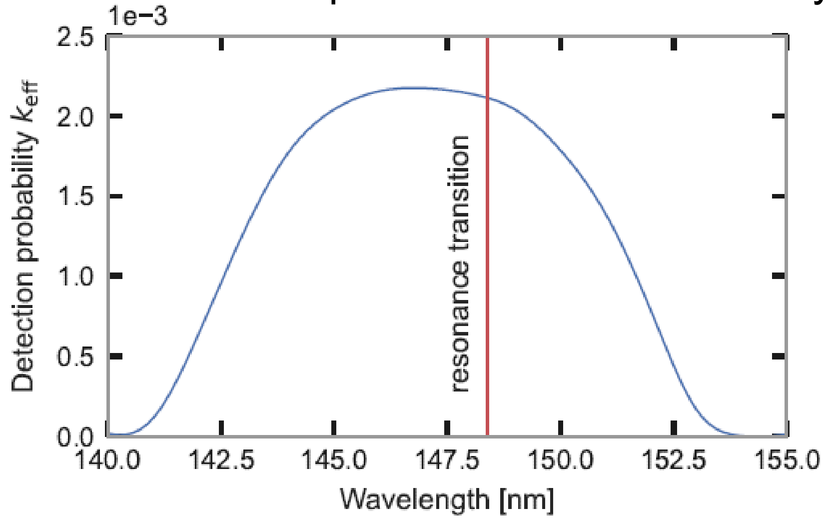
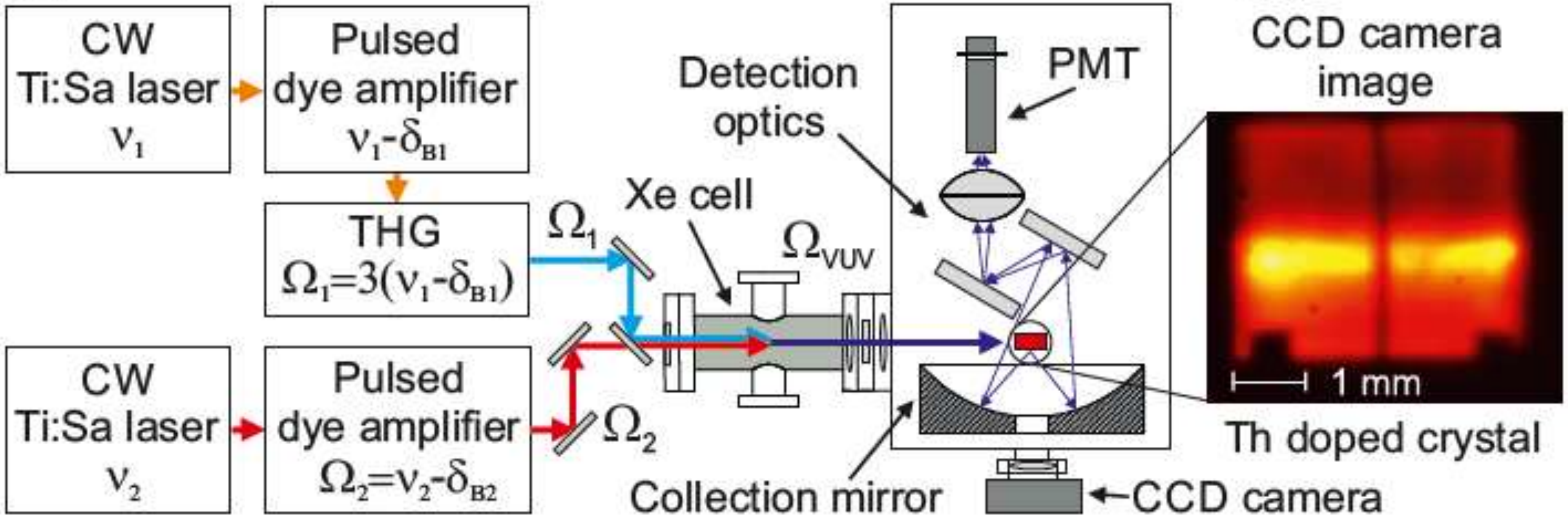
K. Beeks et al. (TU Wien), Phys. Rev. B **109**, 094111 (2024)

Experimental setup

overall photon detection efficiency

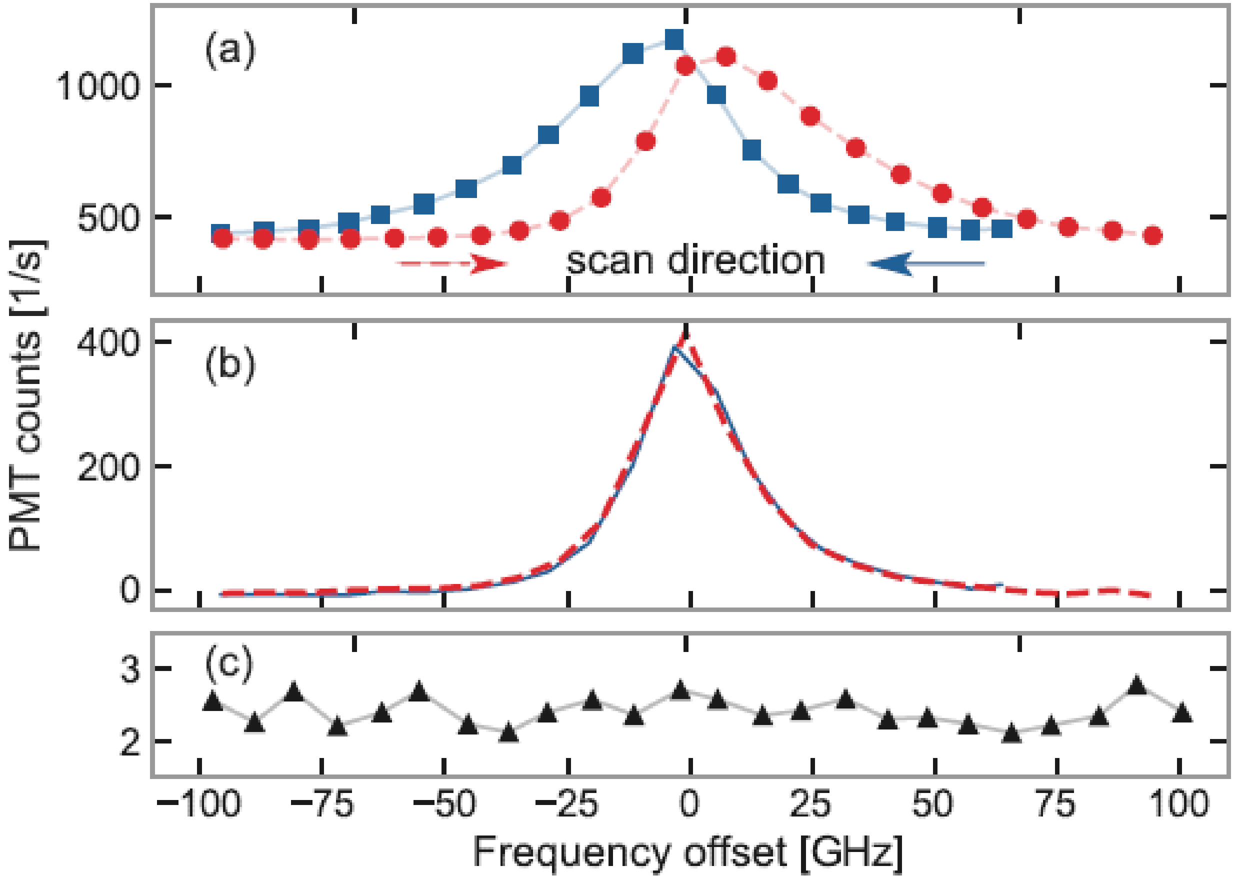


VUV-sensitive
Cs-I photomultiplier



Si-CCD detects
photoluminescence
in the vis. range

Resonant laser excitation, detected in VUV fluorescence



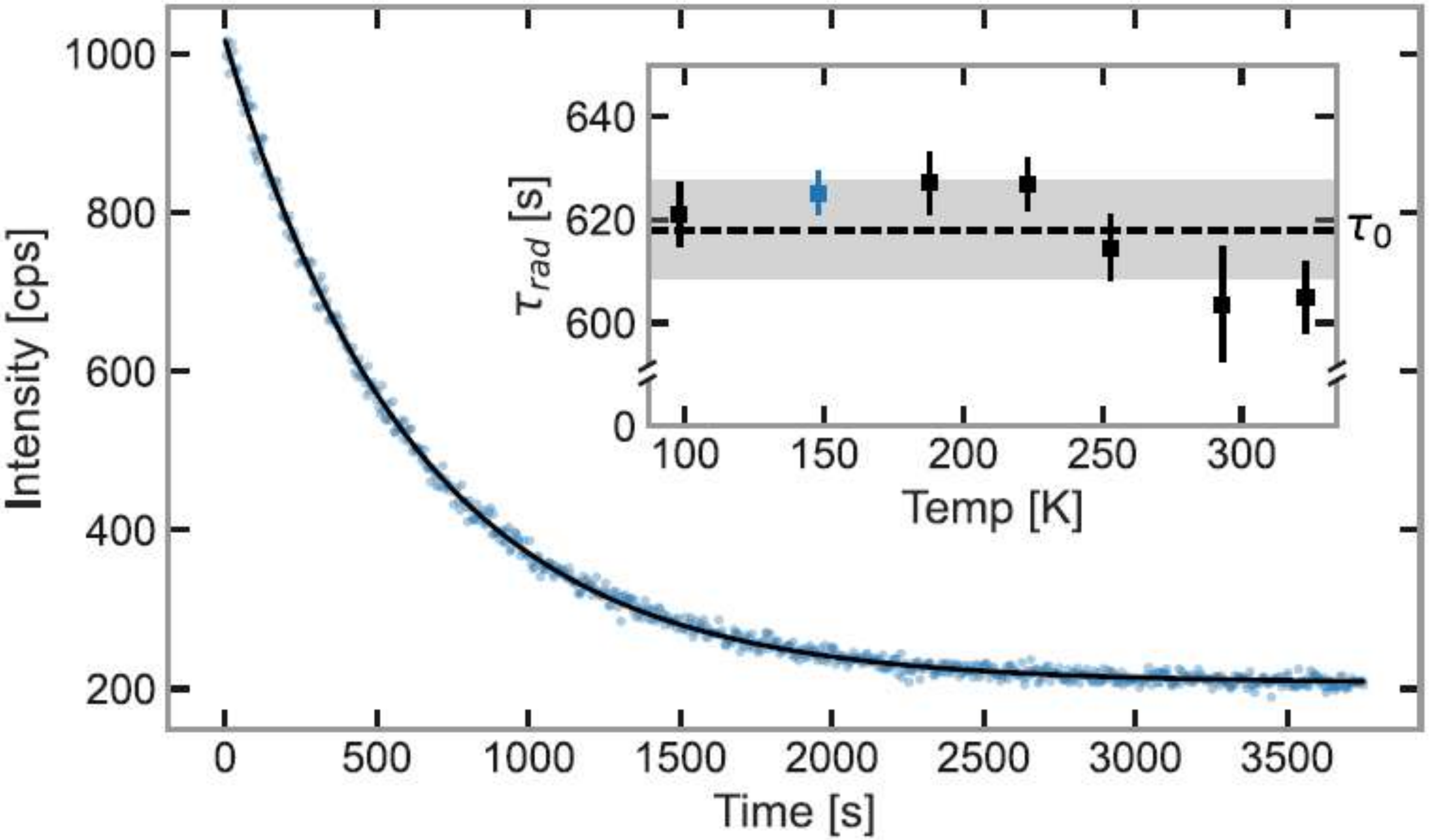
Excitation spectra at 148 nm: Hitting a narrow line with a broad laser.
(each point: 120 s excitation, 150 s detection)

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)

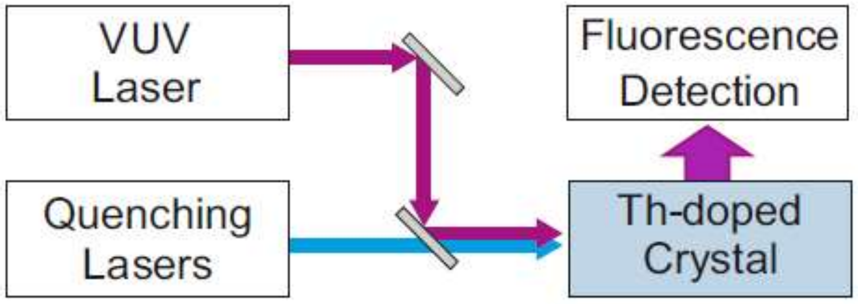
Fluorescence decay curves

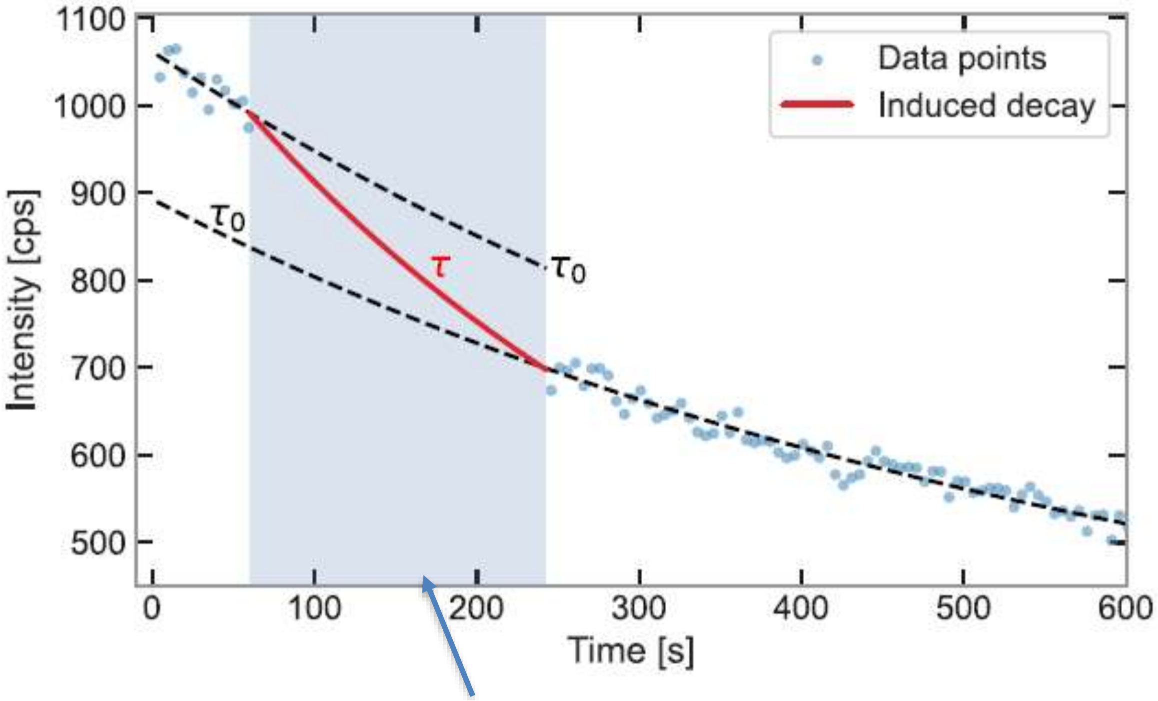


Decay time constant: 618(9) s:

- 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 $\propto n^3 \rightarrow$ isomer half-life 1740 s

Laser-Induced Quenching of the Th-229 Nuclear Clock Isomer in Calcium Fluoride

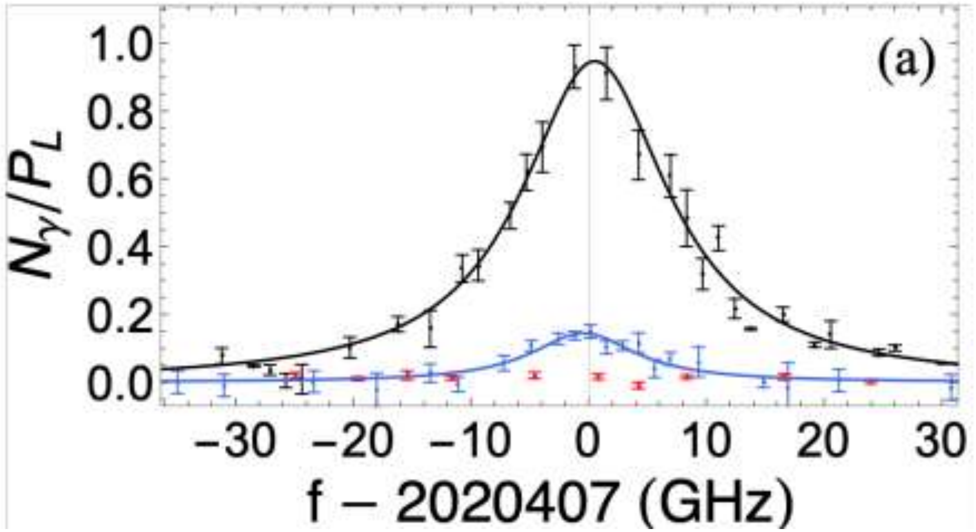




Off-resonant 148 nm laser radiation

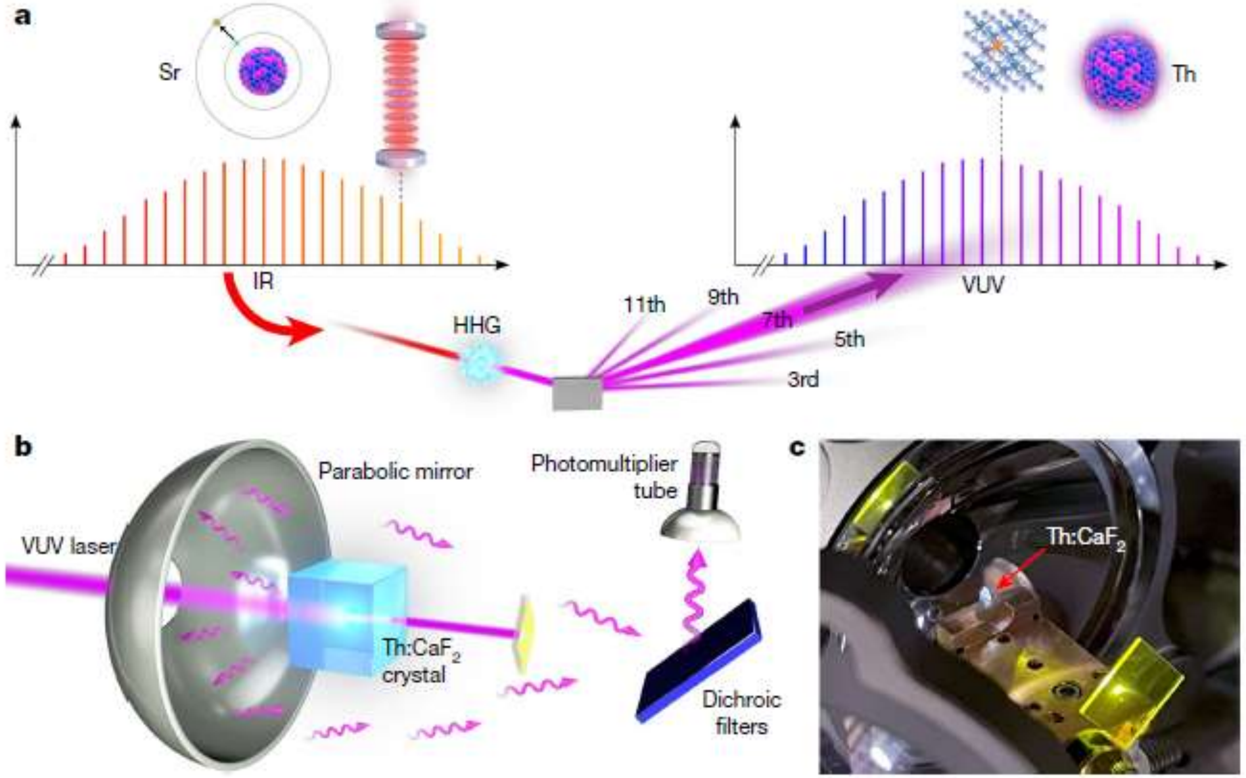
F. Schaden et al. (TU Wien – PTB), Phys. Rev. Res. **7**, L022036 (2025)

Th-229 excitation in Th:LiSAF at UCLA



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

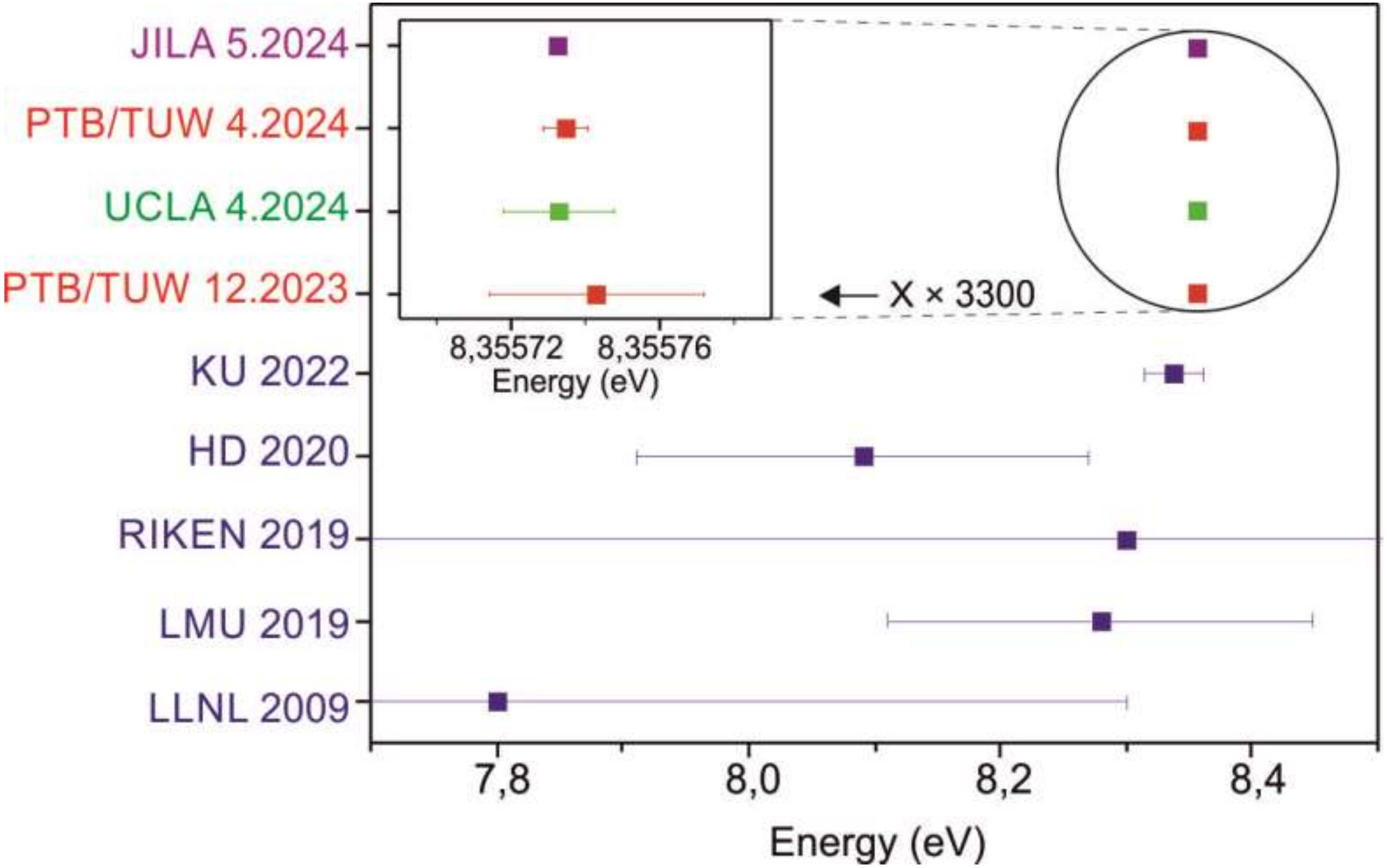
Th-229 excitation with the 7th harmonic of a fs-laser



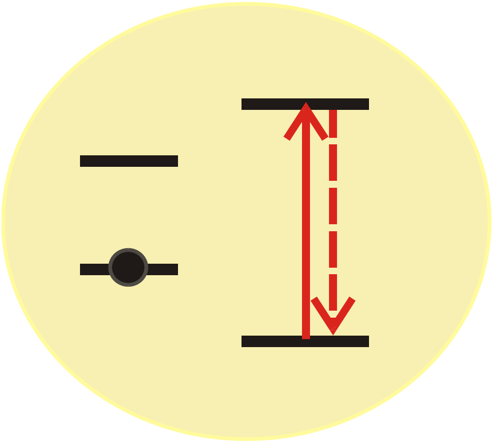
Ch. Zhang et al. (JILA and TU Wien), Nature **633**, 63 (2024)

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

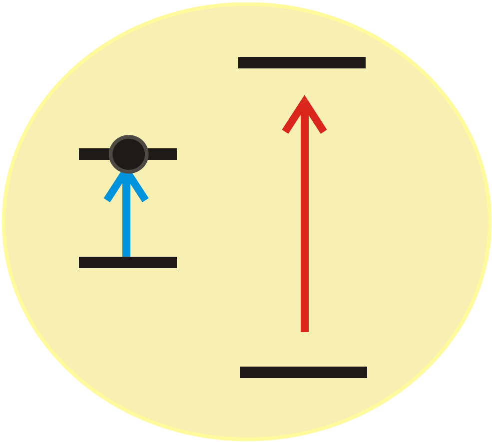
C. Zhang et al., Nature 633, 63 (2024)



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

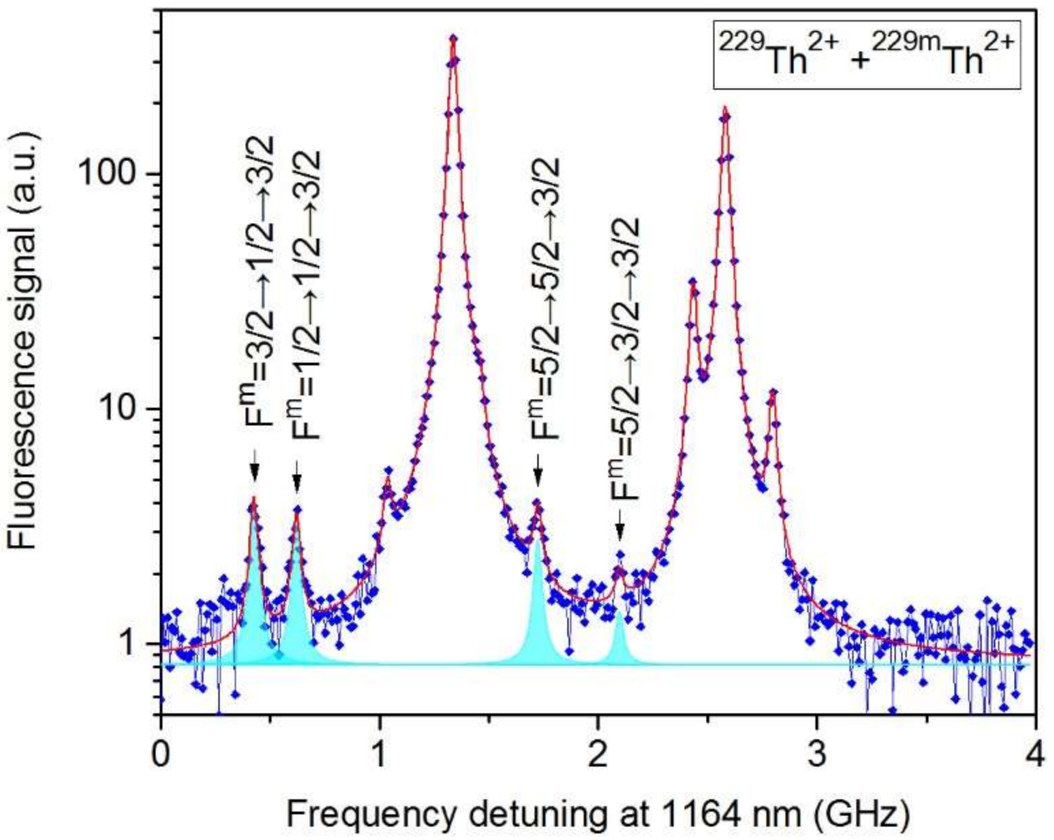


Nucleus in the ground state; laser-induced fluorescence from the shell.



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

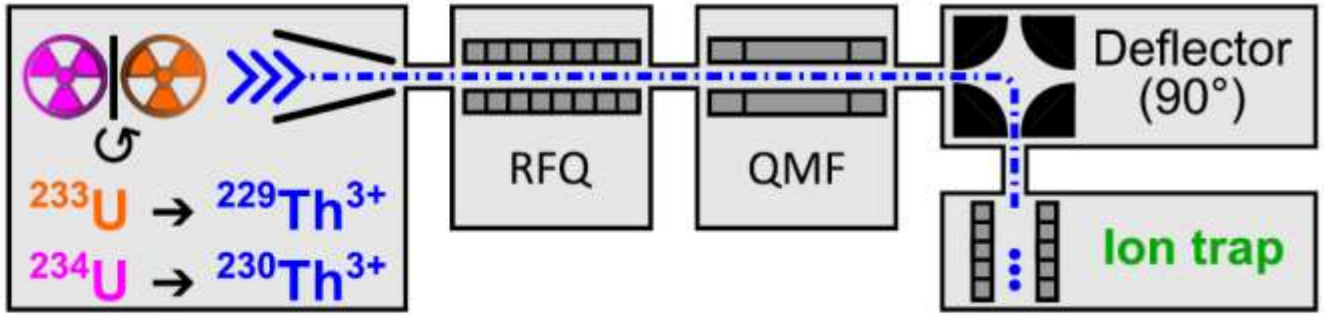


Analog of Dehmelt's „electron shelving"
Observation of „quantum jumps" in the single-ion fluorescence
Coupled degrees of freedom and long nuclear coherence time

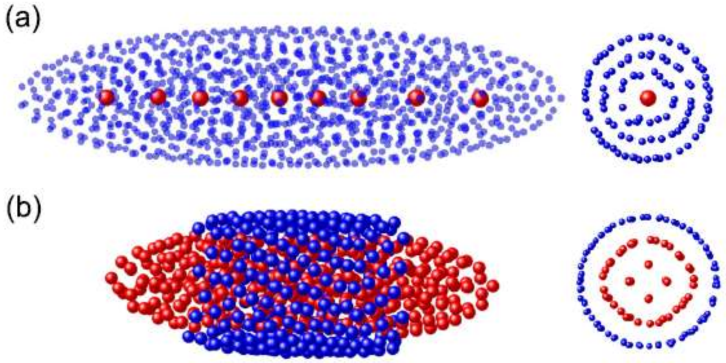
E. Peik, Chr. Tamm, Europhys. Lett. **61**, 181 (2003)

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

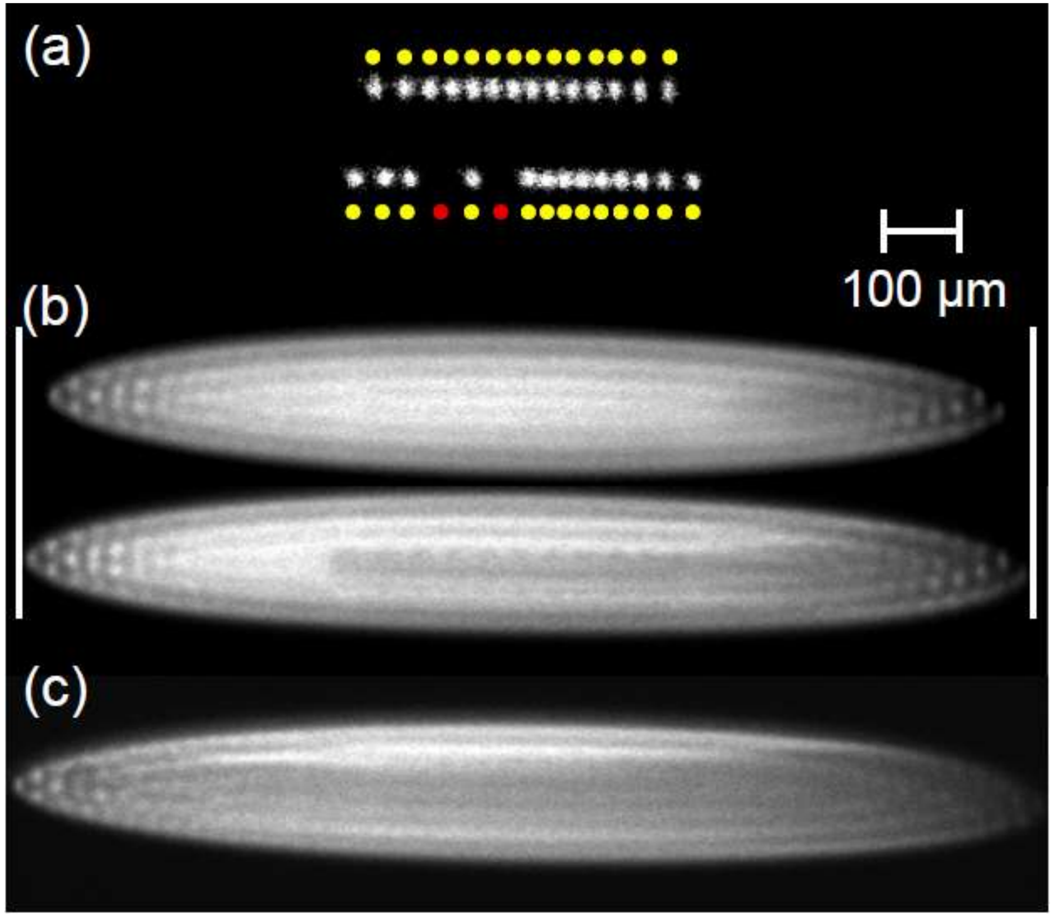
$^{229}Th^{3+}$ trapped recoil ions from ^{233}U, sympathetically cooled with $^{88}Sr^{+}$



^{229}Th recoil ions include
≈2% isomers ^{229m}Th
^{230}Th: reference isotope
with $I=0$



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

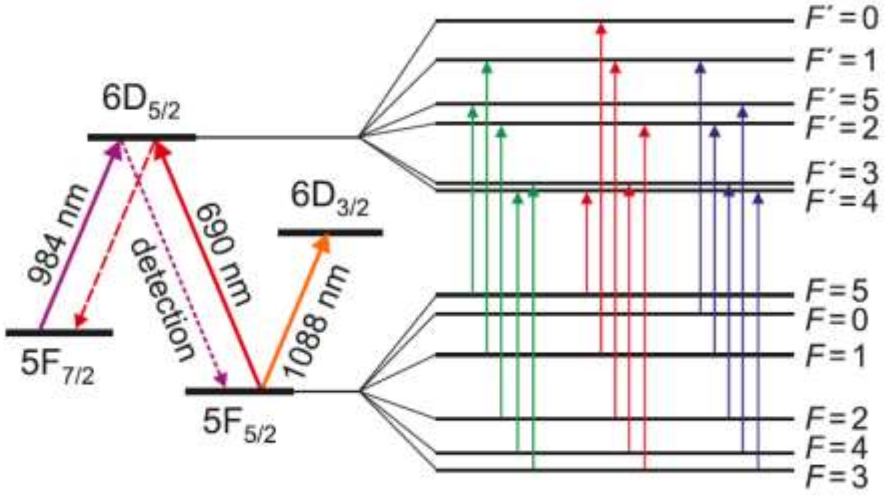


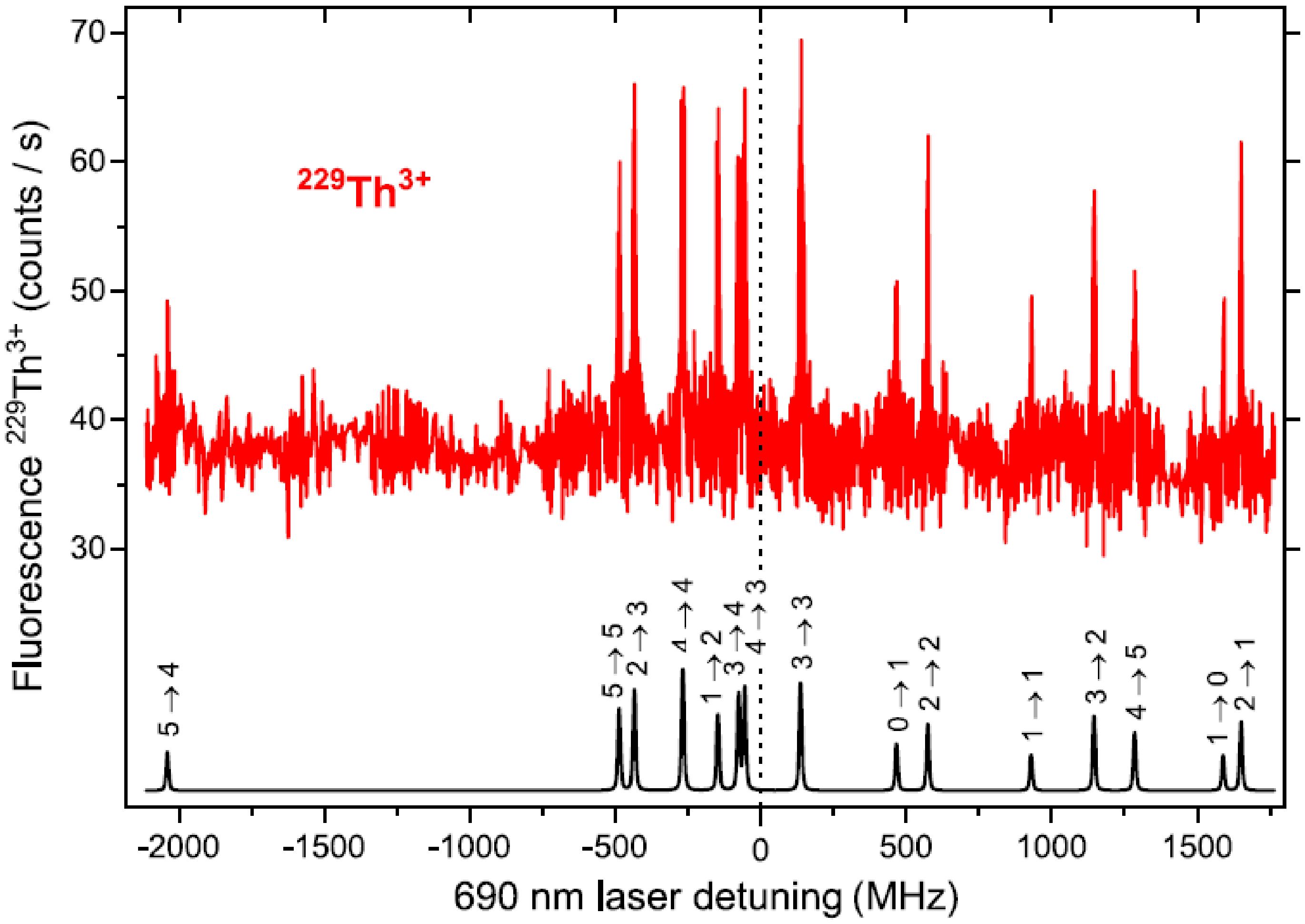
Fluorescence of Sr^{+}, Th^{3+} appear dark

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 structure of $^{229}Th^{3+}$ in the nuclear ground state

Electronic levels of $^{229}Th^{3+}$



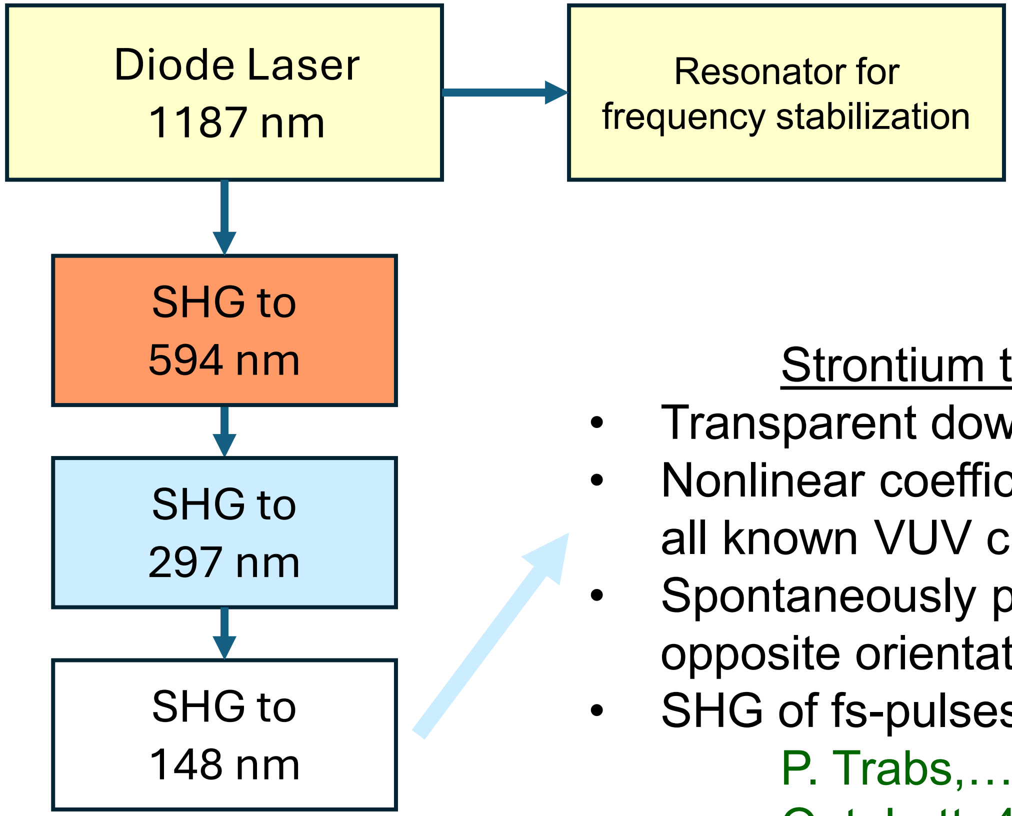


Next objective: improved hyperfine structure and nuclear moments of the isomer.
→ relevant info on nuclear charge distribution and nuclear transition matrix element.

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

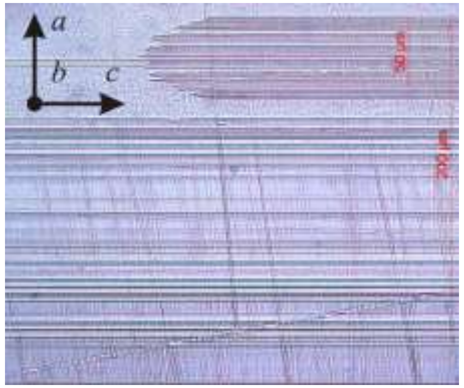
A narrow-linewidth CW laser source for 148 nm

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

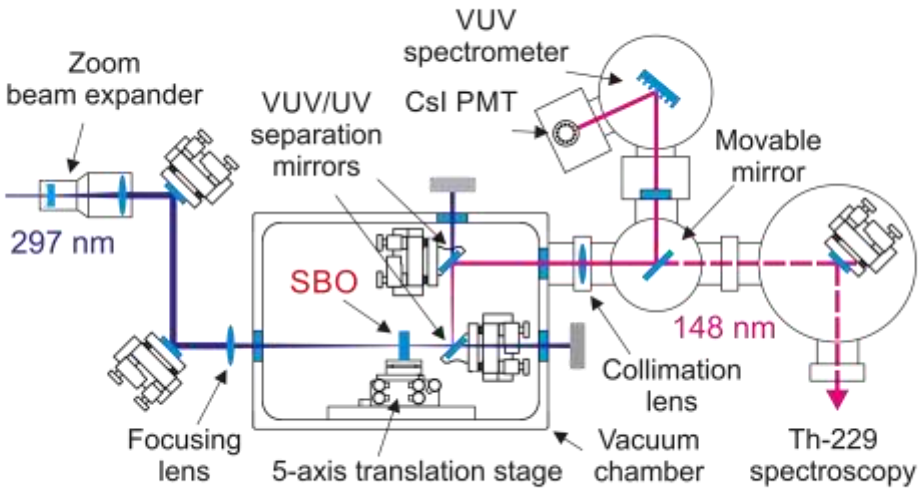


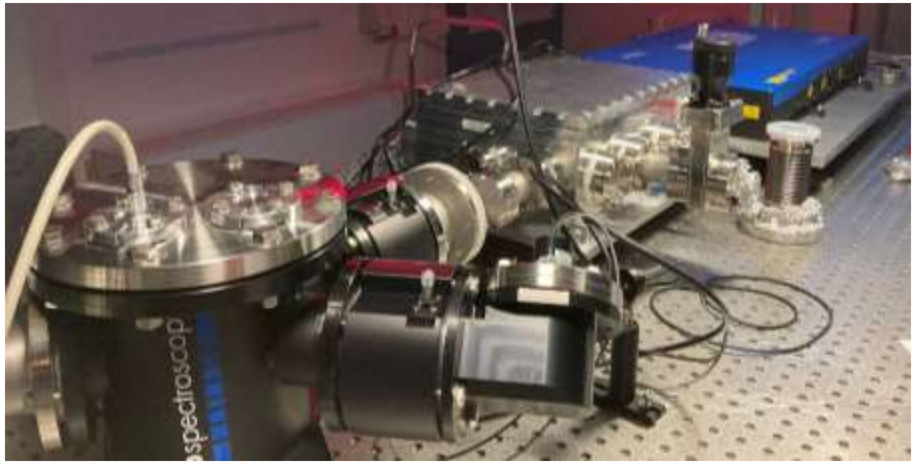
<u>Strontium tetraborate (SBO): SrB_4O_7</u>

- 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)



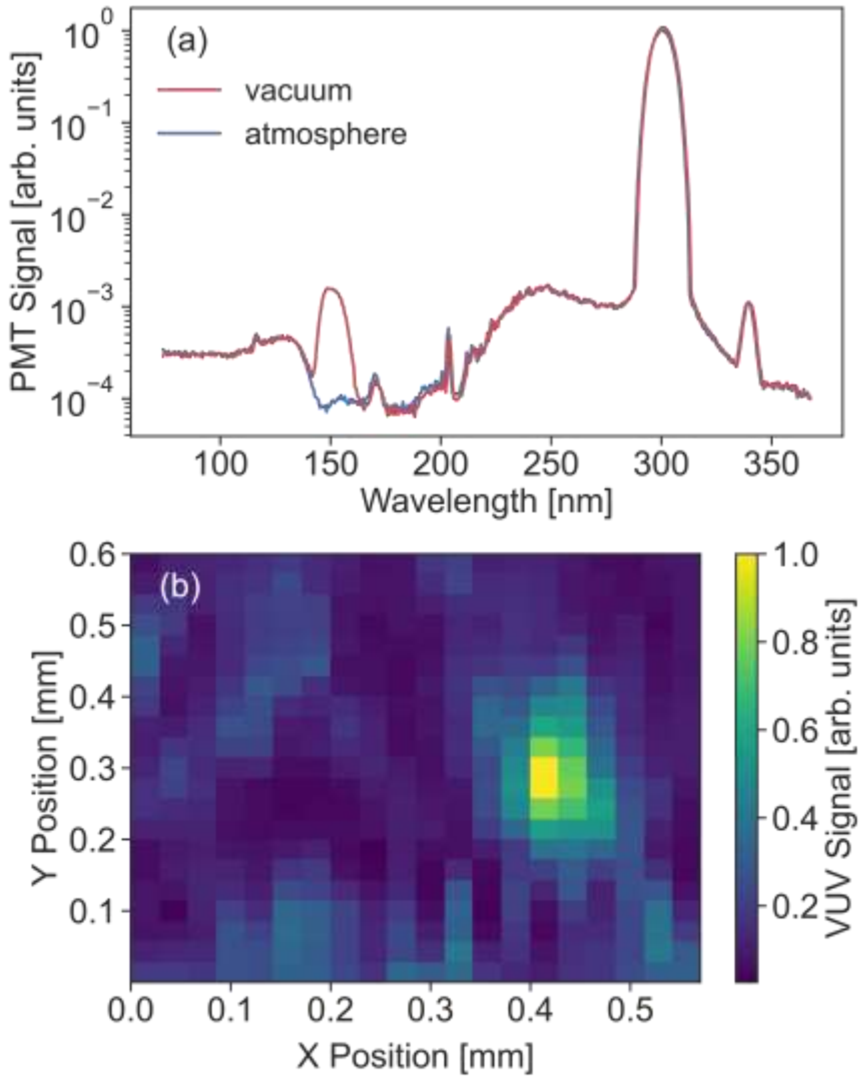
148 nm CW laser source





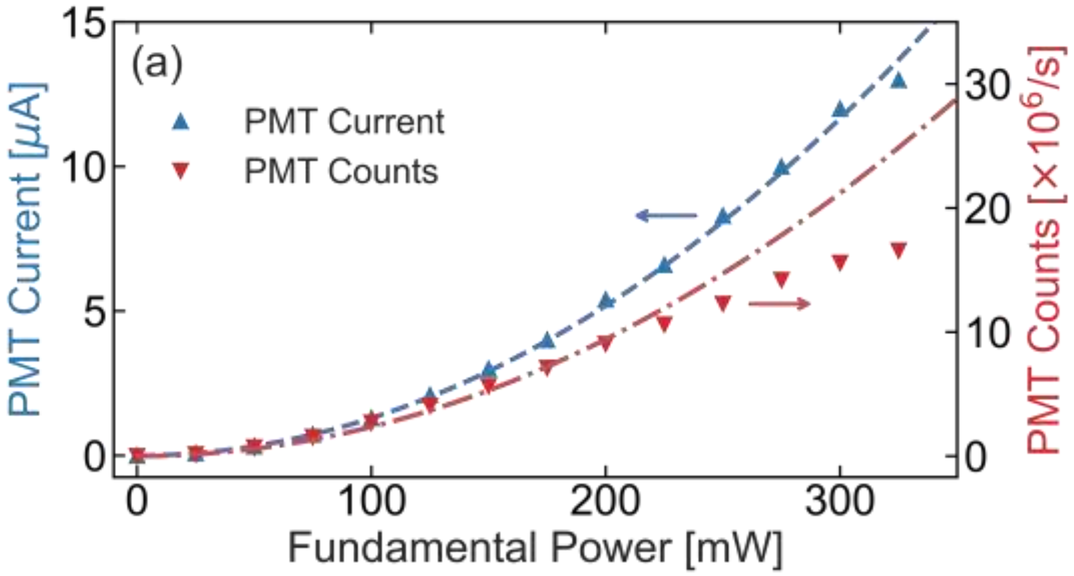
- CW radiation at 297 nm from frequency-quadrupled laser (Toptica TA-FHG pro)
- SBO is mounted on a 5-axis translation stage.
- The main challenge of SHG detection is to achieve sufficient suppression of fundamental radiation.
- To detect VUV signal in fW range from ~0.3 W fundamental radiation, a suppression factor of 10^{14} is required.
- Two dichroic mirrors in combination with a spectrometer grating, and solar-blind CsI PMT with low UV sensitivity.

SHG optimization



- The recorded PMT signal showing fundamental and SHG spectra.
- The SHG signal at 148 nm vanishes under atmospheric condition due to high absorption of the VUV light
- The irregular domain structure of SBO crystal is inhomogeneous across the sample.
- A beam radius of ~ 25 µm for the fundamental radiation was used.

VUV power measurements



- Signal detection: photon counting and PMT current measurements.
- Both PMT current and PMT count signals show quadratic dependence on the fundamental power below 200 mW.
- The generated VUV power is $1.3^{+0.7}_{-0.5}$ nW at an incident UV power of 325 mW.
- The large uncertainty arises from discrepancies between the two-detection method and the uncertainty in the detection efficiency of the setup.

V. Lal, M. V. Okhapkin, J. Tiedau, N. Irwin, V. Petrov, E. Peik, arXiv:2507.17719

See also: Qi Xiao et al., arXiv:2507.19449: CW four-wave-mixing in cadmium vapor

What‘s next?

- Building narrow-linewidth VUV lasers
- Dynamics and lineshapes in the Th-doped crystals
- New materials, different host crystals
- Exciting the nuclear transitions in trapped Th ions
- Th-229 nuclear clocks in tests of fundamental physics

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