Title: Laser Excitation of the Th-229 Nucleus - Towards a Nuclear Clock

Speakers: Johannes Tiedau

Collection/Series: Particle Physics

Subject: Particle Physics

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Abstract:

The thorium-229 nucleus has a unique, low-lying isometric state allowing for laser spectroscopic investigations that are otherwise only accessible in electronic transitions. Here, we report on the first resonant laser excitation of the Th-229 nucleus. The fluorescence signal is observed from two Th-229 doped CaF2 crystals that enable us to determine the center frequency of 2020.409(7) THz corresponding to 148.3821(5) nm of the nuclear transition. The fluorescence lifetime in the crystal is 630(15) s, corresponding to an isomer half-life of 1740(50) s for a nucleus isolated in vacuum. These results pave the way towards high-resolution nuclear laser spectroscopy of Th-229 and an optical nuclear clock with high sensitivity in fundamental tests. This is work done in a cooperation of PTB and TU Wien: J. Tiedau et al., Phys. Rev. Lett. 132, 182501 (2024)

Physikalisch-Technische Bundesanstalt
Braunschweig and Berlin
National Metrology Institute

Laser excitation of the ^{229}Th Nucleus

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Braunschweig und Berlin

TU WIEN

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European Research Council
Established by the European Commission

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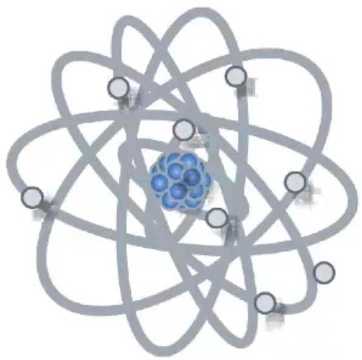
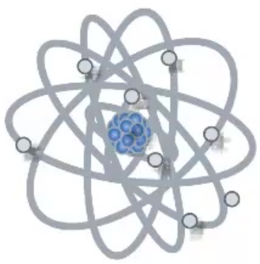
T. Schumm

Outlook

- Introduction Th-229
- First laser excitation of the Th-229 nucleus (+ Implications)
- Hyperfine Spectroscopy of Th-229
- Challenges towards a nuclear clock



How do atoms and nuclei interact with light?



Atoms

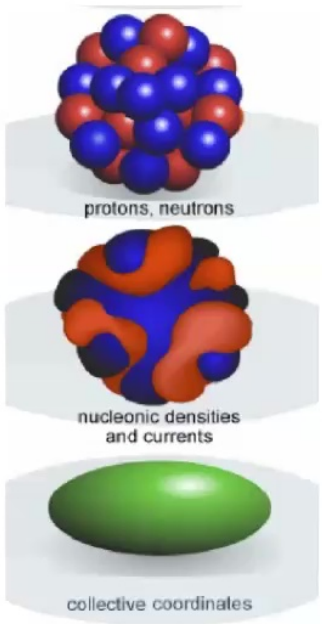
Two signs of charge: e, p

Bound by a central force
(symmetry, selection rules)

Nuclei

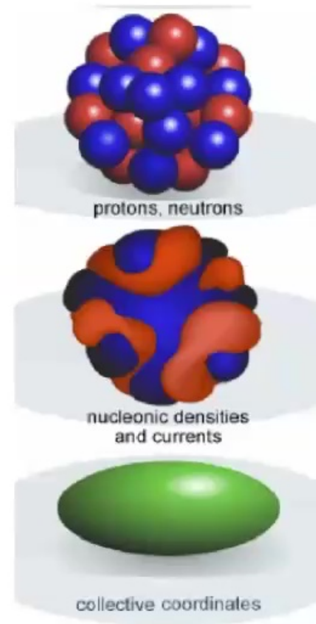
Only one sign of charge: p

Non-central forces
(deuteron already has electric quadrupole moment)

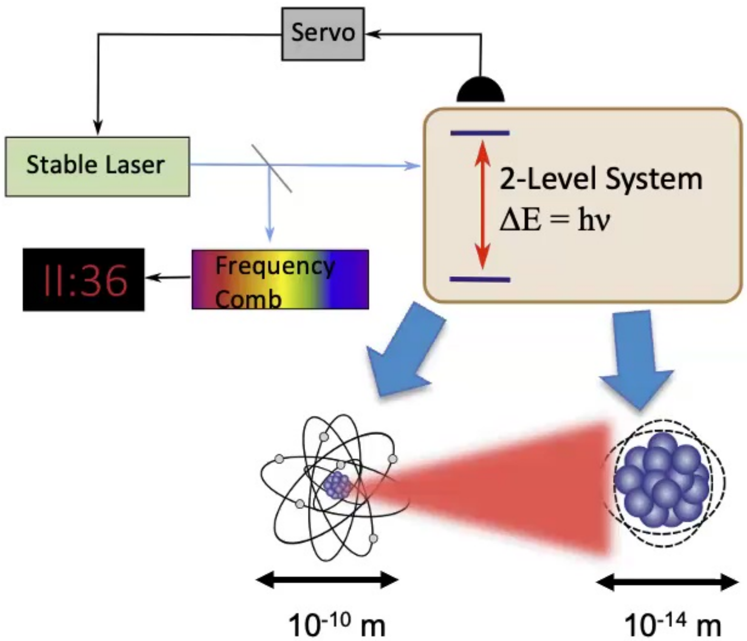


How do atoms and nuclei interact with light?

Atoms	Nuclei
Two signs of charge: e, p	Only one sign of charge: p
Bound by a central force (symmetry, selection rules)	Non-central forces (deuteron already has electric quadrupole moment)
Bohr magneton μ_B	Nuclear magneton $\mu_N = \mu_B/1836$
Small size: $r/\lambda \approx 10^{-4}$	Very small size $r/\lambda \approx 10^{-9}$

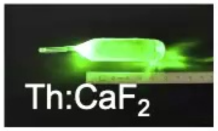


A nuclear clock



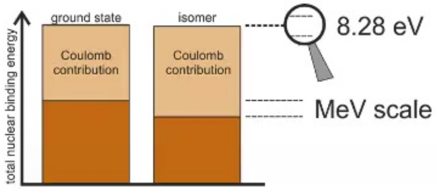
Several features of an optical clock based on a nuclear transition:

- High accuracy - Weak coupling to external fields. E. Peik, Chr. Tamm, Europhys. Lett. **61**, 181 (2003)
  C. J. Campbell et al., PRL **108**, 120802 (2012)
- High stability: Nuclear structure largely unaffected by chemical environment
  → ~ 10^{16} ions doped in crystal to achieve a very stable frequency standard.



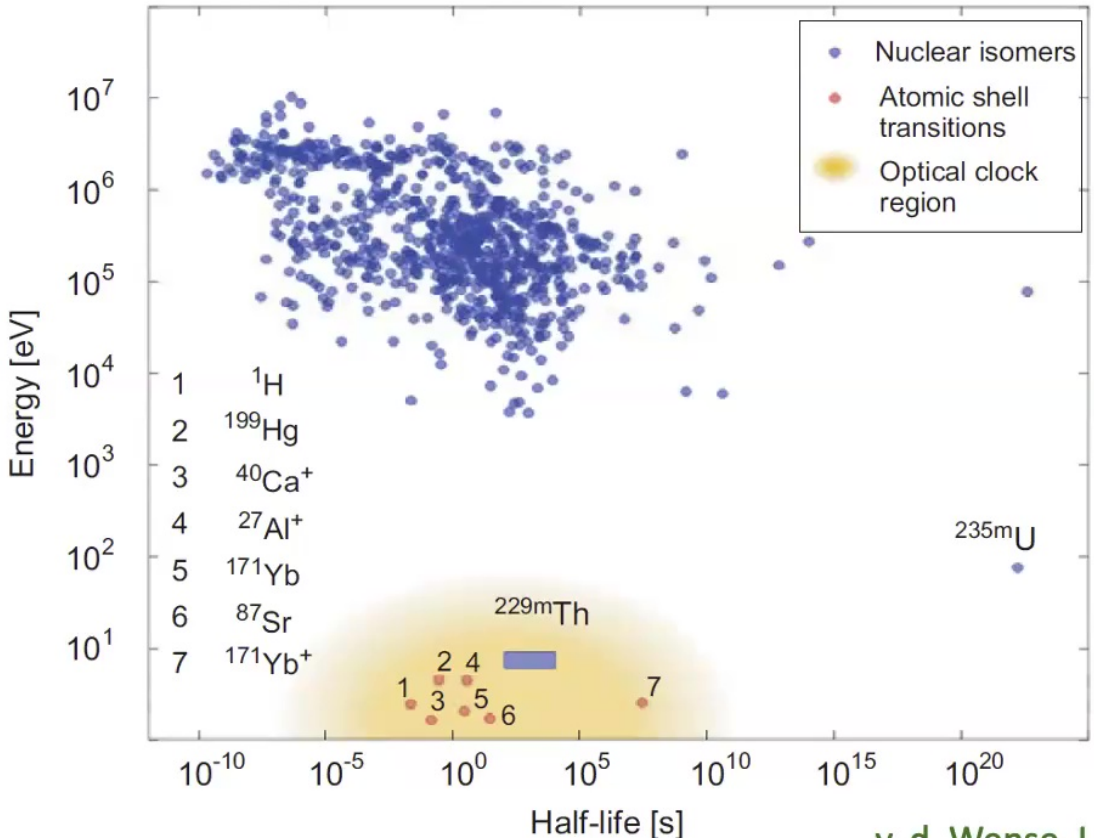
Proposed at PTB, UCLA, TU Wien

- High frequency (?)
- Transition depends on nuclear binding energy
  → high sensitivity to a possible temporal change of fundamental constants [4].



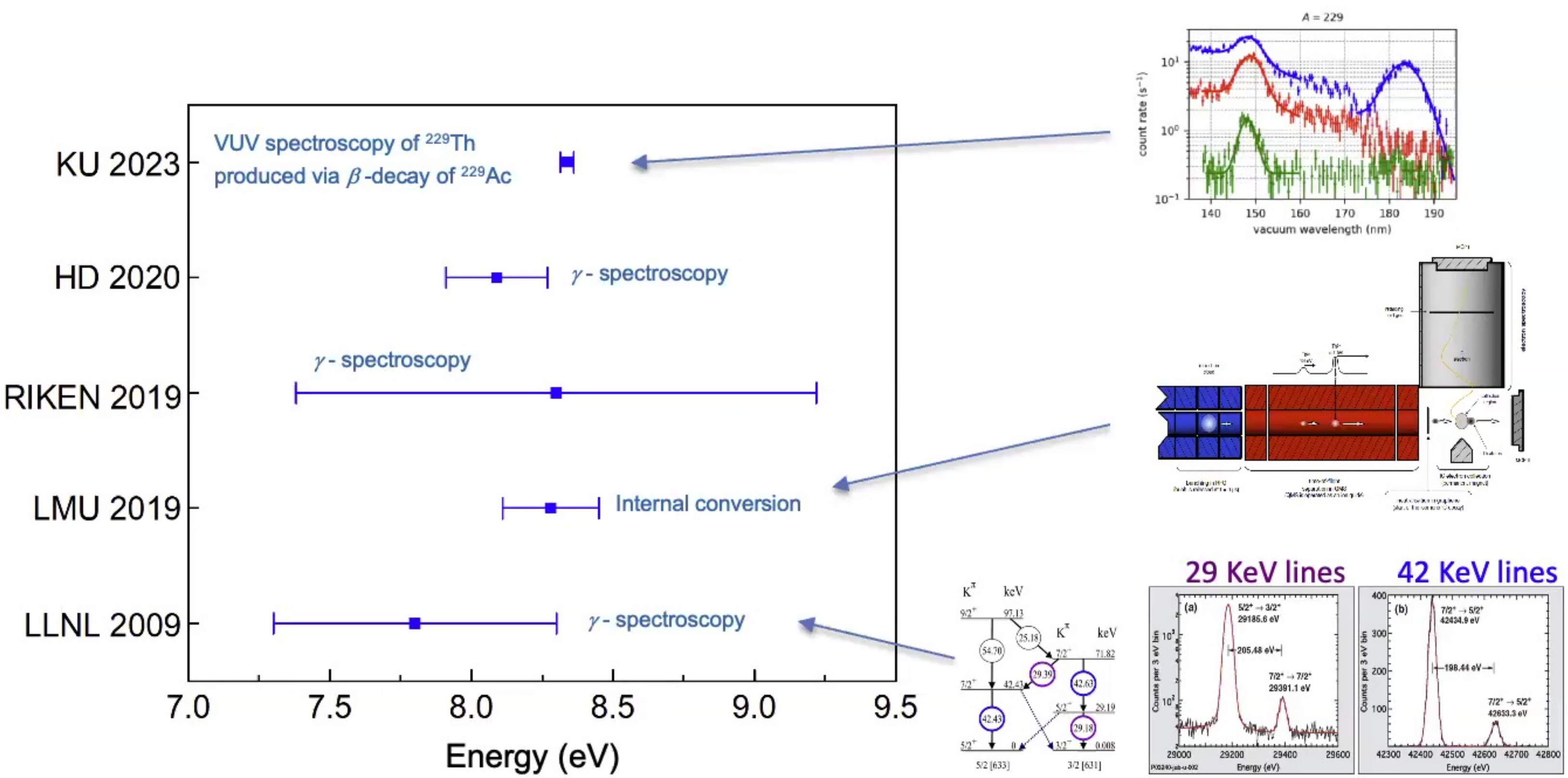
e.g. V.V. Flambaum et al. PRL **97**, (2006)

Thorium-229



v. d. Wense, L. et al., *Nature* **533**, 47-51 (2016).

History of Th isomer energy measurements



Outlook

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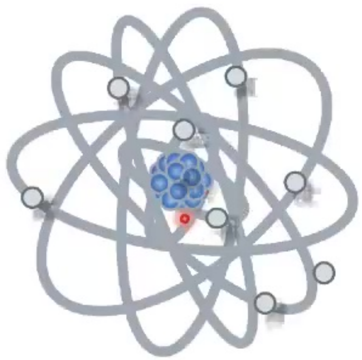
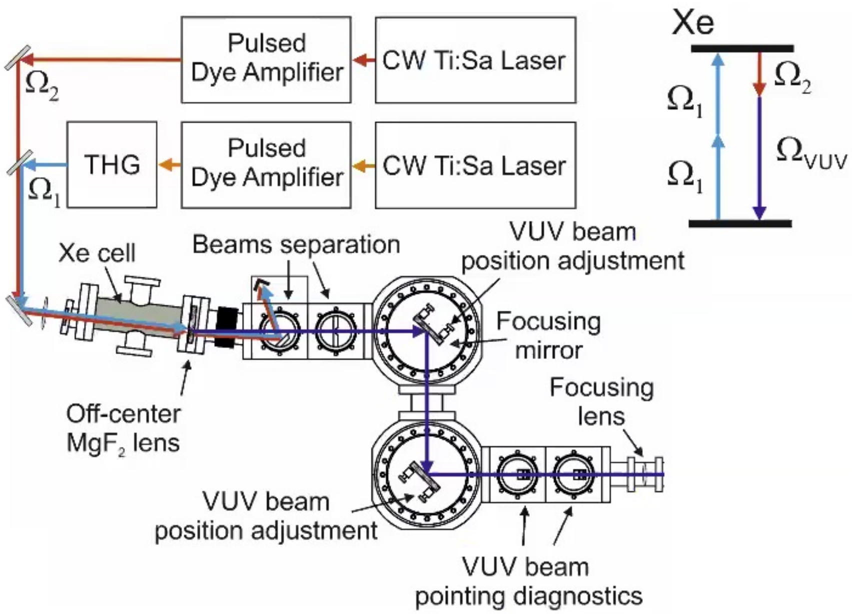
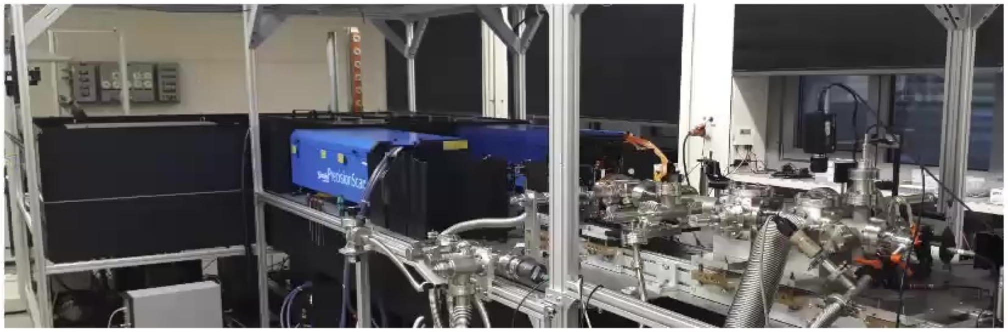
- **First laser excitation of the Th-229 nucleus (+ Implications)**
- Hyperfine Spectroscopy of Th-229
- Challenges towards a nuclear clock



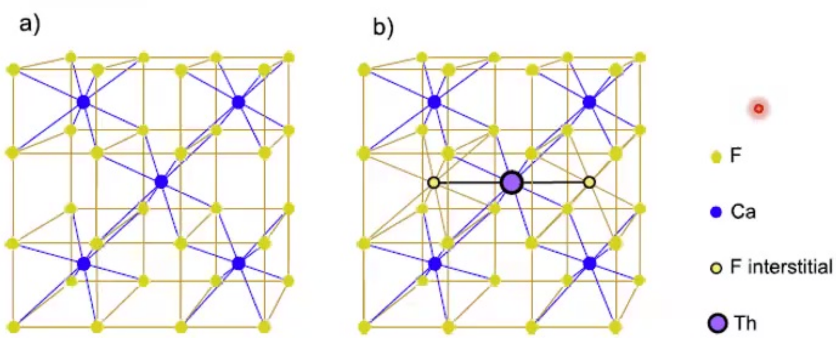
Vacuum-ultraviolet (VUV) generation



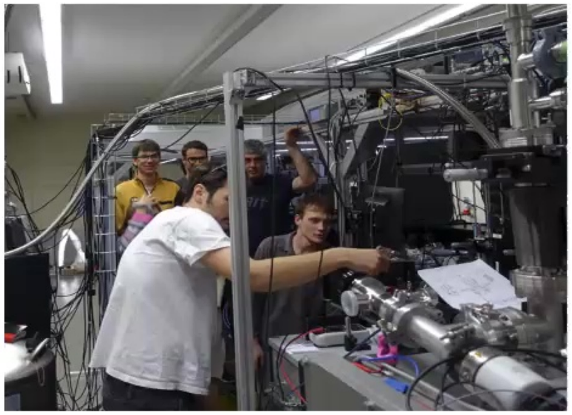
- Two cw ring lasers as seed;
- Pulsed dye amplifiers ~ 50 mJ/pulse, ~10 ns, 30 Hz repetition rate, THG~ 4 mJ/pulse;
- VUV pulse energy > 10 μJ (2×10^4 photons/(s Hz)), linewidth 6-10 GHz (measured by laser spectroscopy of Th ions);



Th-doped CaF_2 crystals



Th^{4+} replaces Ca^{2+};
charge compensation from two F^- interstitials



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

Key parameters of Th-doped crystals

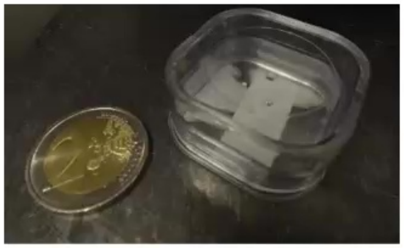


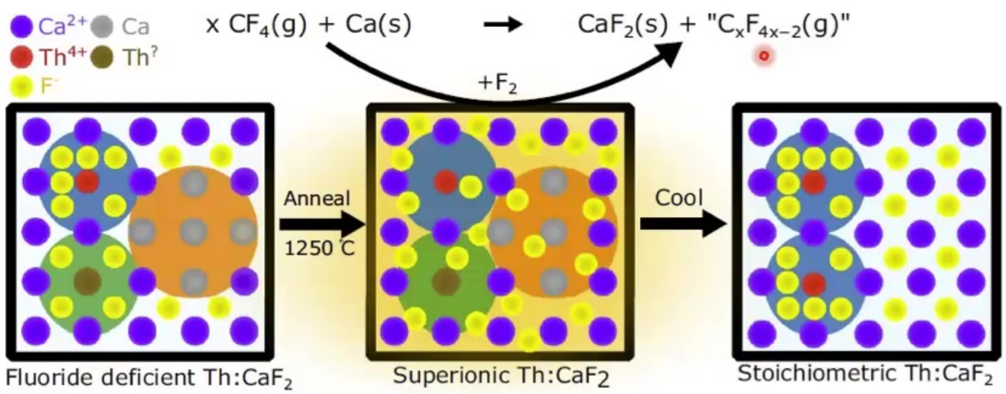
FIGURE 3.18: Four 1 mm^3 and one 4 mm^3 ^{229}Th:CaF_2 crystals cut and polished using above techniques.

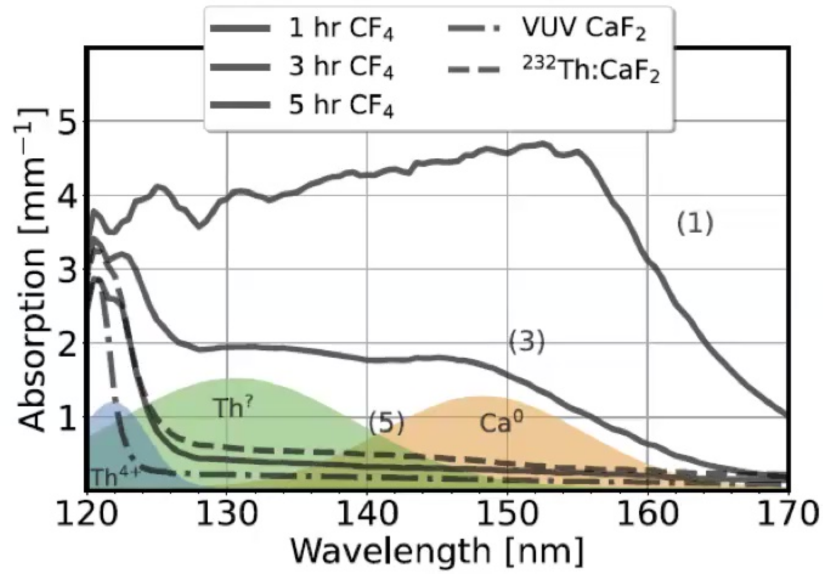
Crystal Code	X2	C10	V12
Dopant isotope	Th-229	Th-229	Th-232
Crystal ID	2021/04	2020/06	2020/05
Th activity [kBq]	66	22	0
Concentration [cm^{-3}]	5×10^{18}	3×10^{17}	1×10^{19}
Column density [mm^{-2}]	8×10^{15}	1×10^{15}	4×10^{16}
Transmission@150 nm	40 %	30 %	40 %

The resulting crystals are verified for chemical purity using optical and gamma spectroscopy.

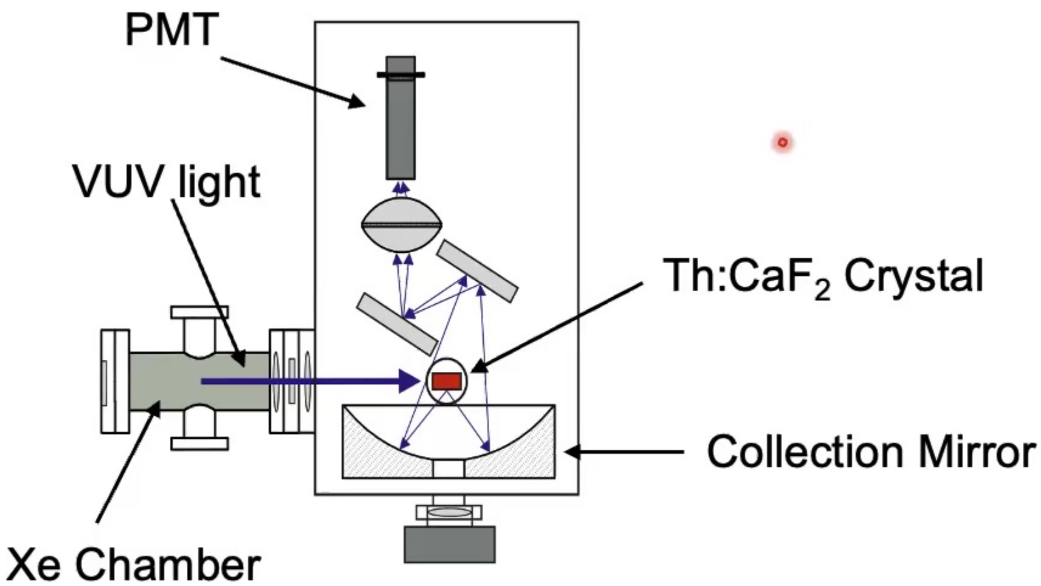
Radiolysis leads to a fluoride-deficiency of the resulting crystal: drastically reduces the VUV transmission.
Annealing under 10^5 Pa of CF_4 gas is used to replenish the fluoride in the grown crystals, recovering VUV transmission and increasing radiation hardness.

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

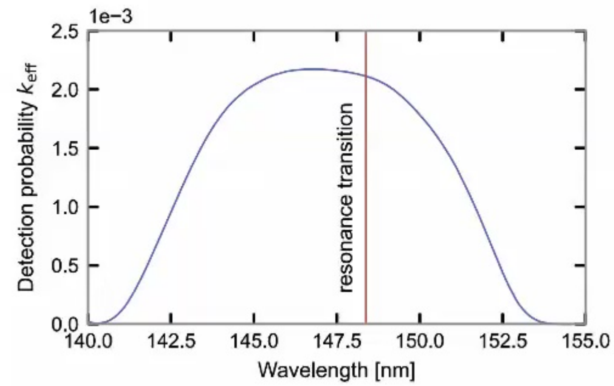




Fluoride ions are mobilized by VUV and ionizing radiation, remaining Ca forms broadly absorbing clusters
Transmission can be recovered by annealing under CF_4
VUV-induced damage can be prevented by cryogenic cooling (LN2)

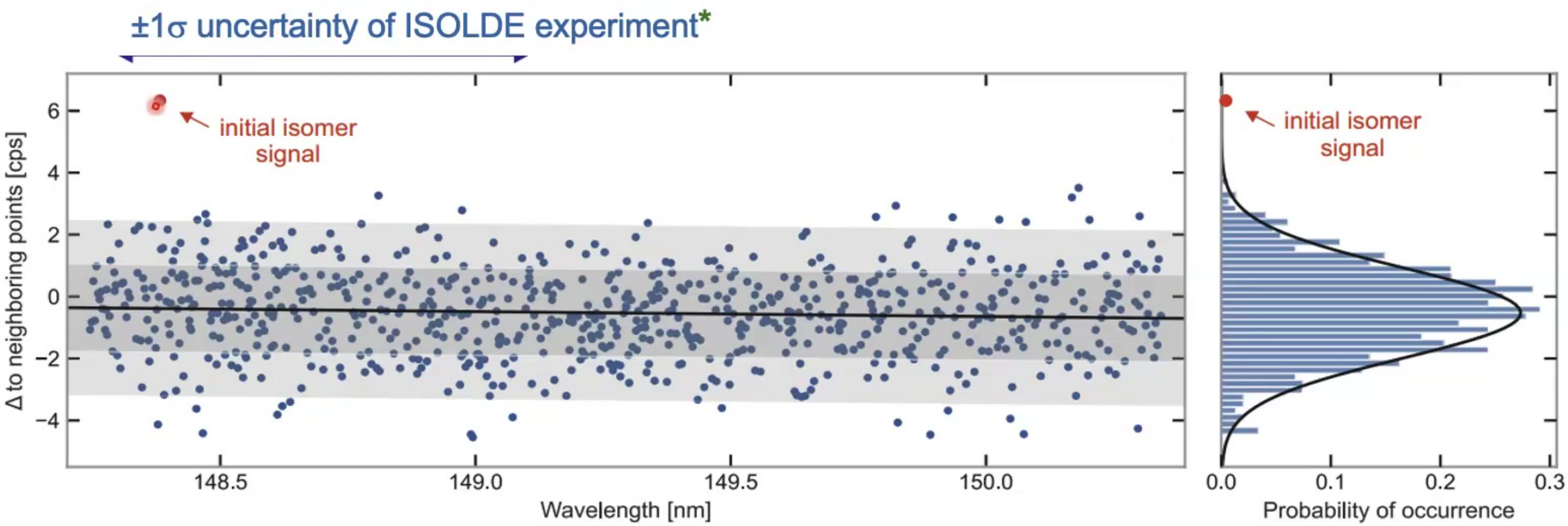


- Detection scheme: narrowband to cut out Cherenkov radiation and radioluminescence;



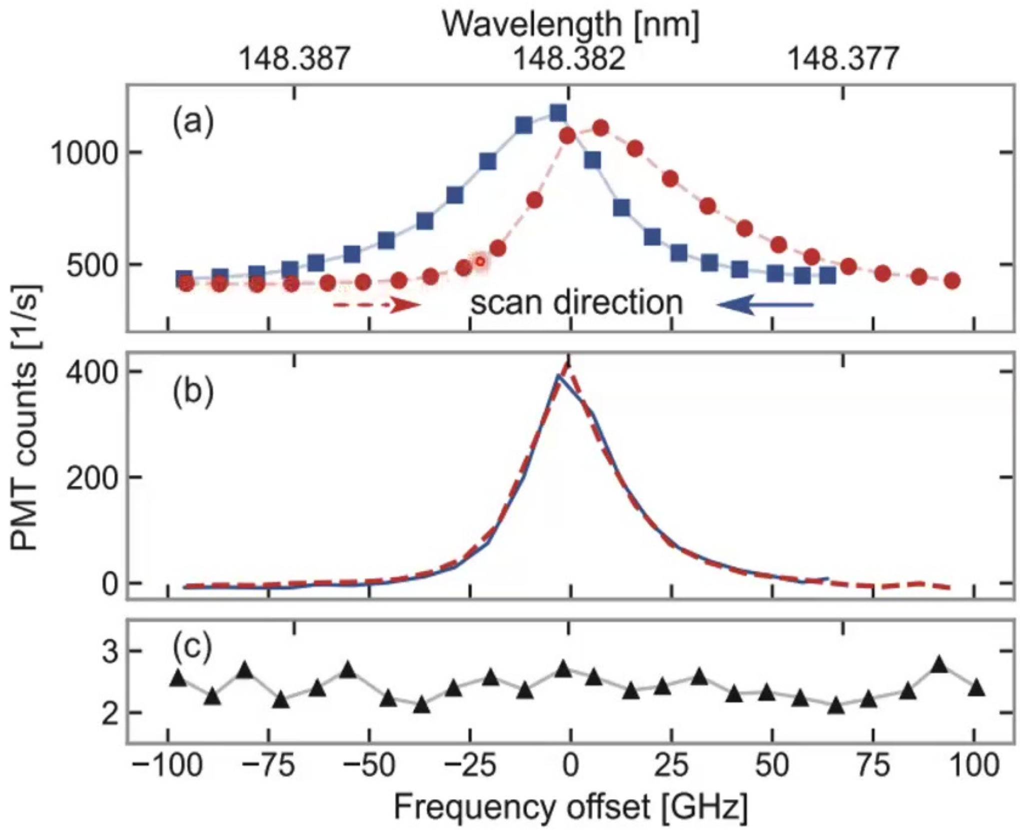
- Th-doped crystal is mounted on a cold finger. Operation temperature ~ 150 K.

Initial isomer signal



*S. Kraemer et al., Nature 617, 706 (2023)

Resonant laser excitation, detected in VUV fluorescence



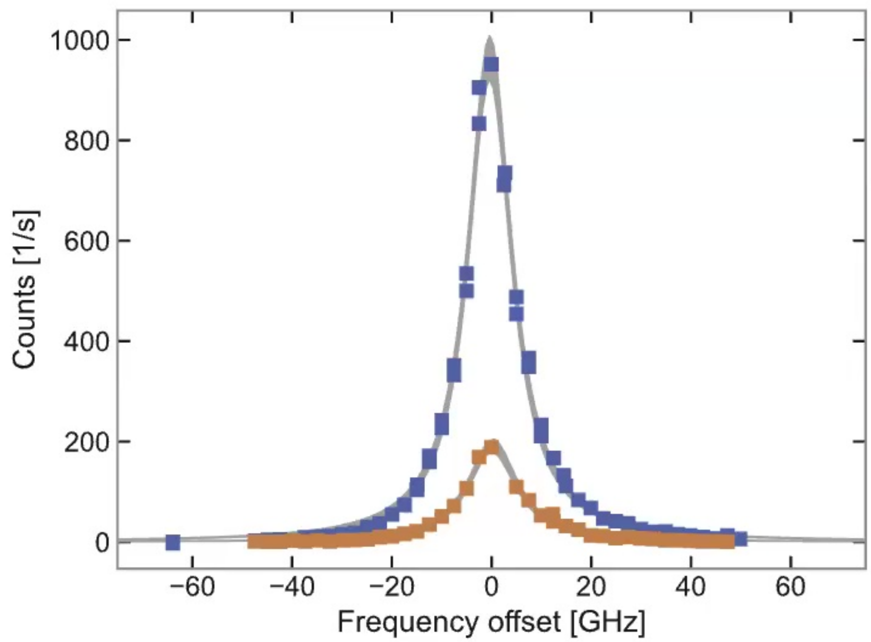
Excitation spectra at 148 nm: Hitting a narrow line with a broad laser.
(each point: 120 s excitation, 150 s detection)

Line shapes after correction for the slow exponential fluorescence decay

Control experiment with Th-232: no signal

J. Tiedau et al. PRL **132**, 182501 (2024)

Fluorescence signals (T=150 K)



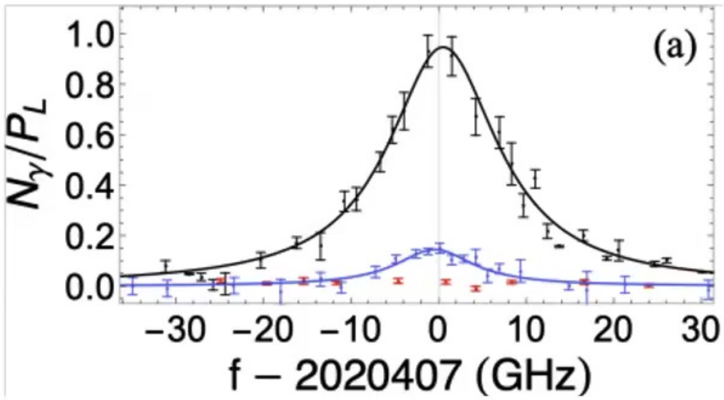
Crystal Code	X2	C10
Dopant isotope	Th-229	Th-229
Th activity [kBq]	66	22
Concentration [cm^{-3}]	5×10^{18}	3×10^{17}
Column density [mm^{-2}]	8×10^{15}	1×10^{15}

Improved value of the isomer energy measured in both crystals

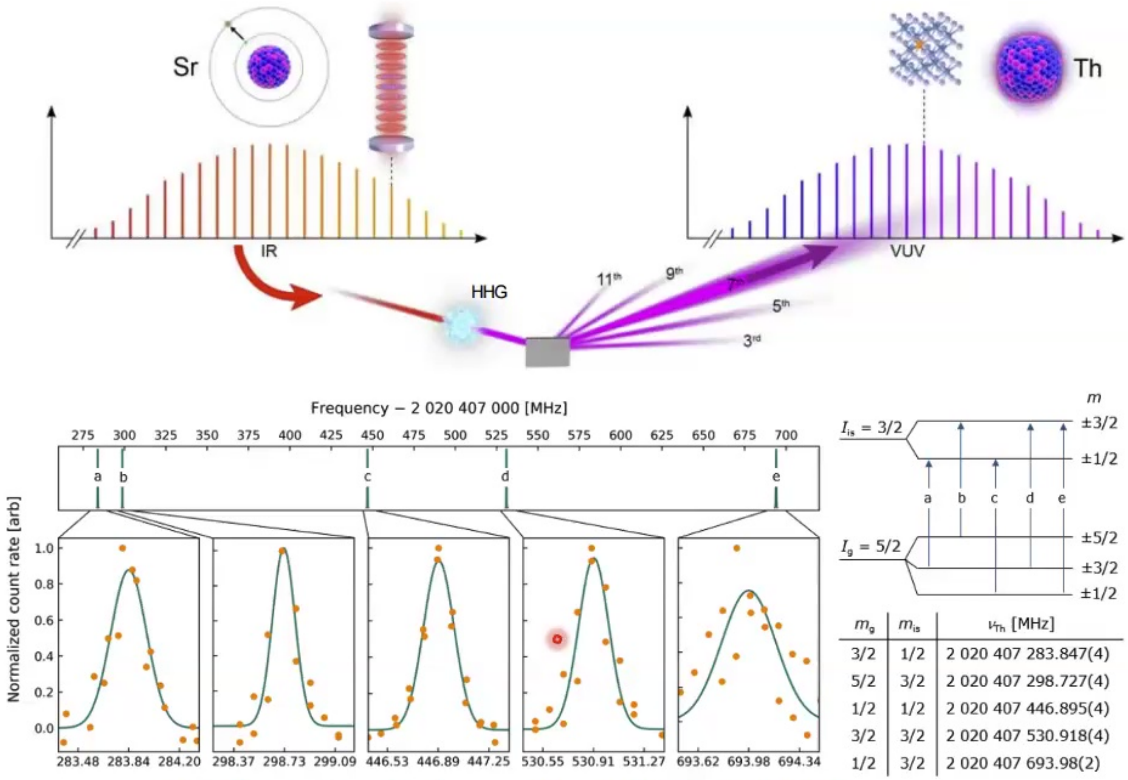
λ = 148.38125(33) nm
ν = 2020.4078(35) THz
ΔE = 8.355734(14) eV

Lineshape (FWHM: 12(1) GHz) and
resonance frequency (2020.4079(35) THz)
are consistent for two differently doped crystals.

UCLA / JILA



R. Elwell et al., PRL 133, 013201 (2024)

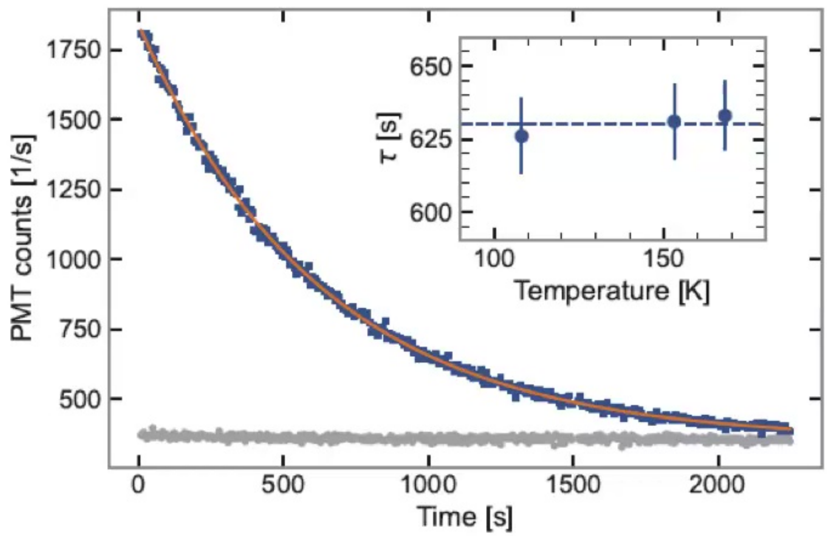


m_g	m_{is}	ν_{Th} [MHz]
3/2	1/2	2 020 407 283.847(4)
5/2	3/2	2 020 407 298.727(4)
1/2	1/2	2 020 407 446.895(4)
3/2	3/2	2 020 407 530.918(4)
1/2	3/2	2 020 407 693.98(2)

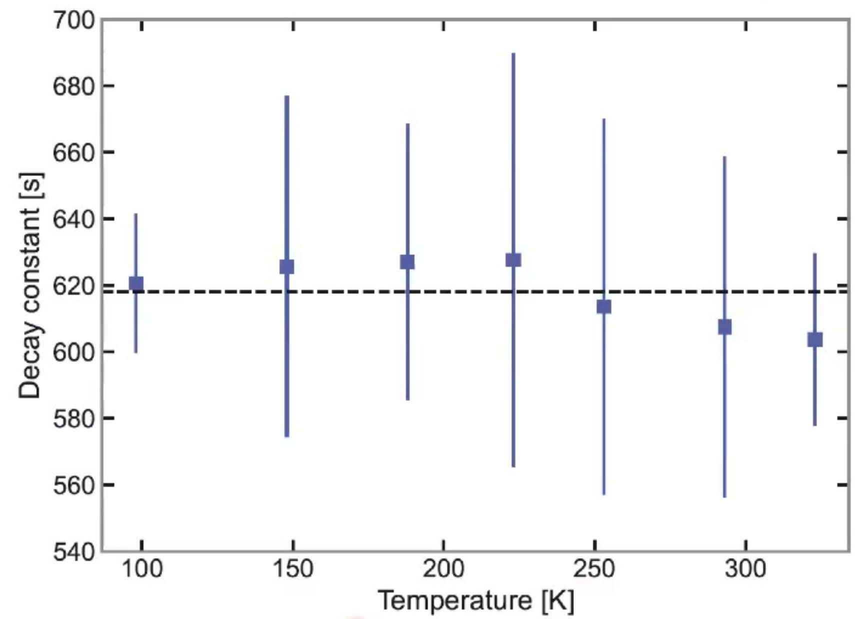
C. Zhang et al., arXiv 2406.18719

Fluorescence decay curves

X2 crystal: radiative lifetime: 630(15) s



C10 crystal: wider temperature range



Decay time constant:

- 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

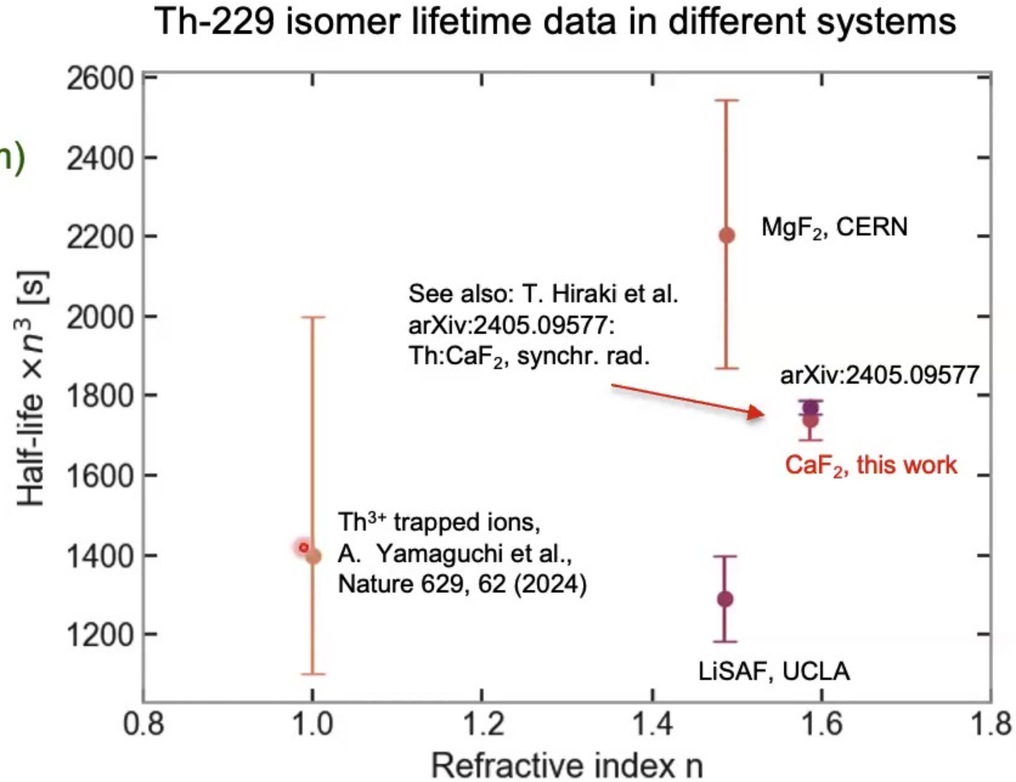
Lifetime comparison

The electromagnetic mode density in the dielectric medium is enhanced relative to vacuum, leading to a n^3 enhancement of the M1 spontaneous transition rate:
G. Nienhuis, C. Th. J. Alkemade, Physica (Amsterdam) 81, 181 (1976).

With this correction, the half-life becomes: 1740(50) s

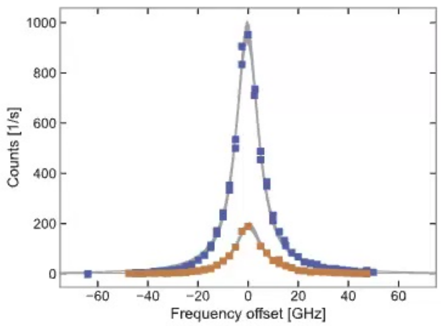
In Weisskopf units
(nuclear oscillator strength):
B(M1)=0.022 W.u.

Nuclear structure predictions
(E. Tkalya, N. Minkov, et al.):
B(M1)= 0.006 - 0.05 W.u.

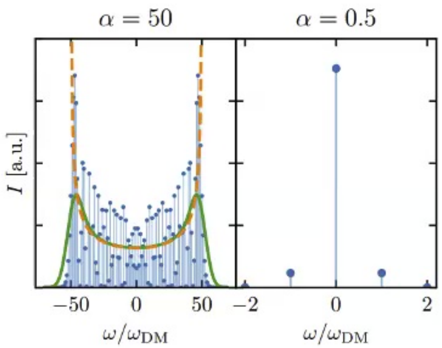


Search for dark matter – recent progress

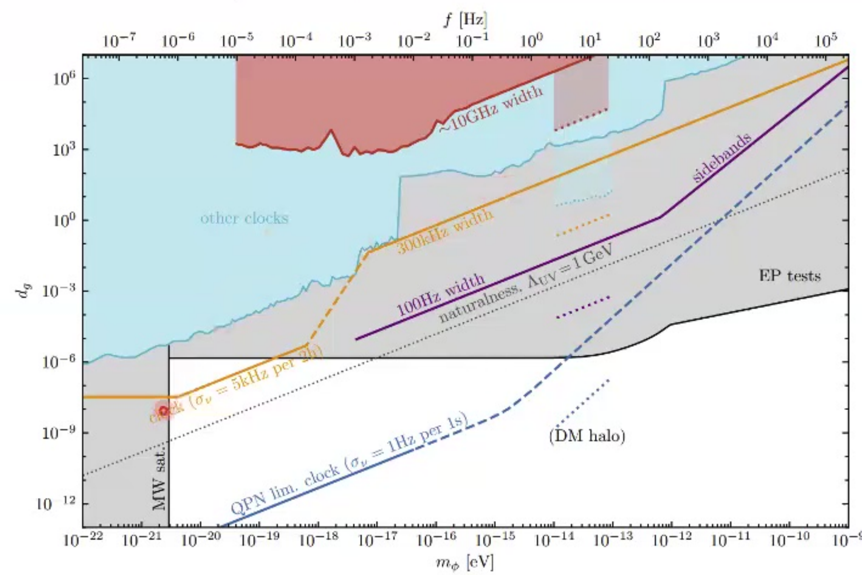
Lineshape Measurements



Calculations of dark matter induced sidebands



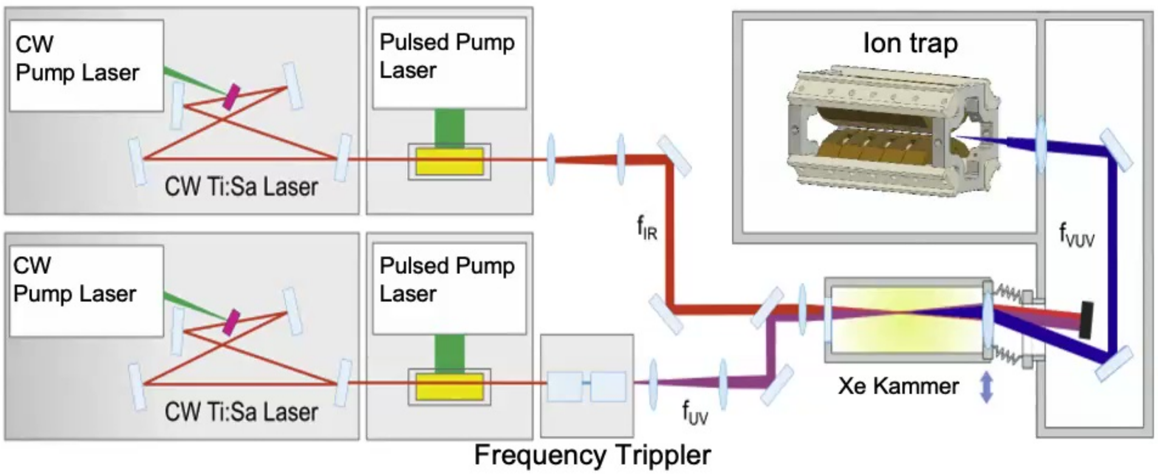
Updated exclusion limits

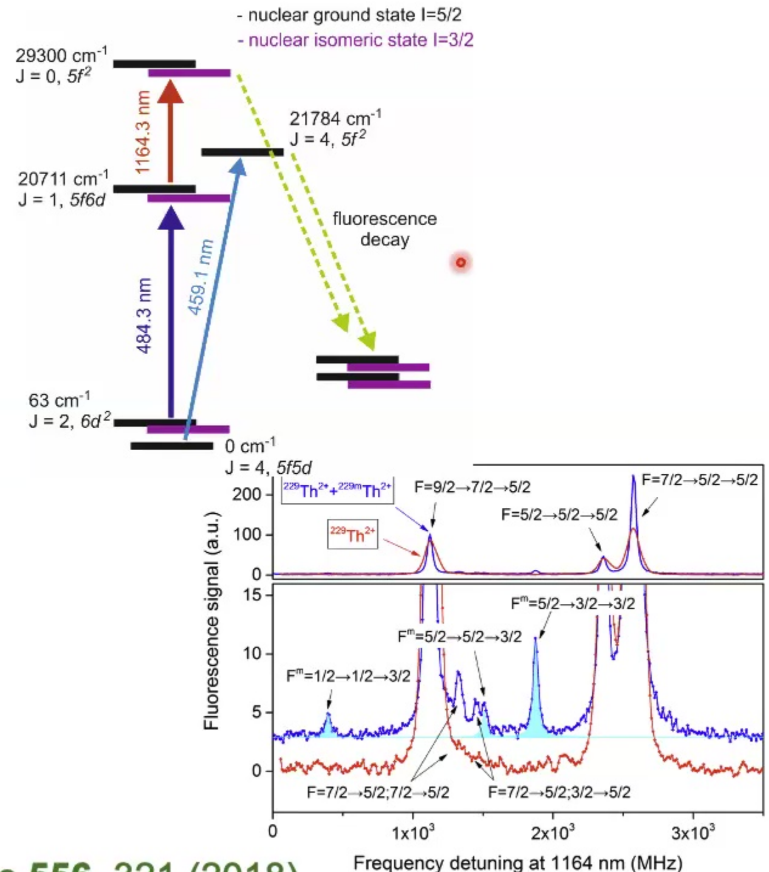


E. Fuchs et al. arXiv:2407.15924v1 (2024)

What‘s next?

Next steps: ion traps

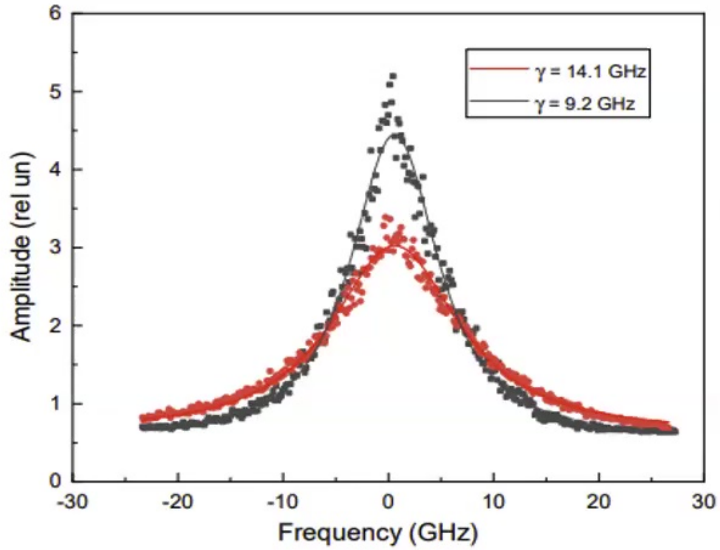




- Laser excitation of trapped Th^{2+} ions
  - Buffer gas cooled Th^{2+} ions.
  - Nuclear state detection via hyperfine spectroscopy

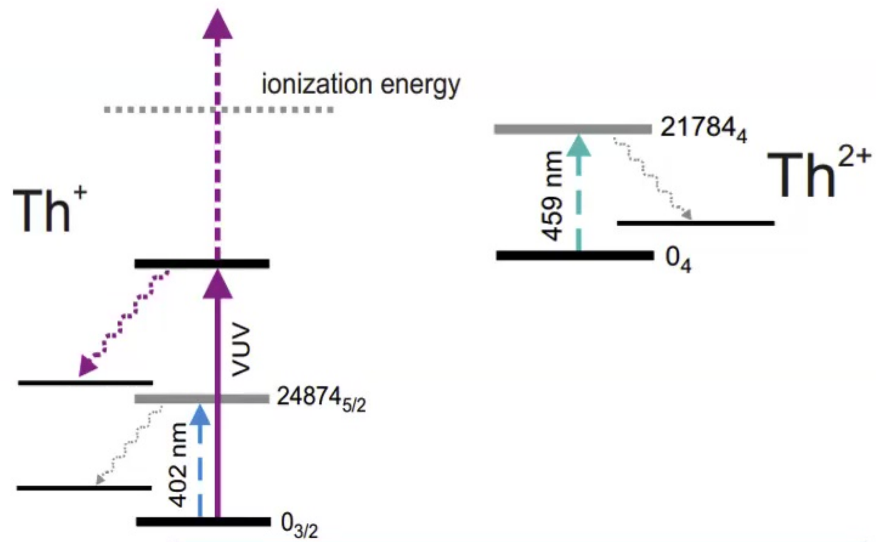
J. Thielking et al., Nature **556**, 321 (2018)

Laser spectroscopy of $^{232}Th^{+}$ in the ion trap



Black: optimum tripler efficiency, VUV pulse energy reduction with the pump laser delay
Red: VUV pulse energy reduction through changing the angle of half waveplate.
VUV pulse energy: 1.8 μJ.

J. Thielking, et al. NJP 25.8 (2023)



Three new **odd** atomic states of Th^{+} are found in the energy range of isomeric state

Excitation wavelength	Level
148.812 nm	67198.8(2) cm^{-1}
148.668 nm	67264.2(2) cm^{-1}
148.203 nm	67474.9(2) cm^{-1}

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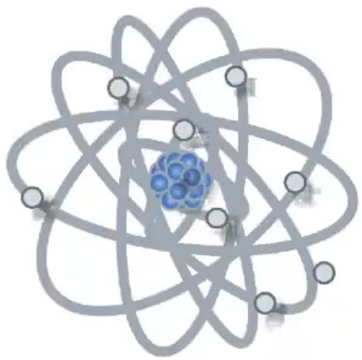
- Challenges towards a nuclear clock



Characterization of Th isomer

$\Delta\langle r^2\rangle^{229m,229} = 0.0105(13)\ \mathrm{fm}^2$ UD/PTB 2018
$\Delta\langle r^2\rangle^{229m,229} = 0.0097(26)\ \mathrm{fm}^2$ RIKEN 2023

← Recalculated from measurements of isomer / isotope shifts

^{229m}Th isomer

3/2$^+$ [631]

$\mu = -0.37(6)\ \mu_N$, $Q_0 = 8.7(3)\ eb$ PTB/LMU 2018
$\mu = -0.378(8)\ \mu_N$, $Q_0 = 8.84(10)\ eb$ RIKEN 2023

← HFS measurements

M1 transition

$$\mu_1 = \frac{A_1 I_1}{A_2 I_2}\mu_2$$

$$Q_{s1} = \frac{B_1}{B_2} Q_{s2}$$

$$Q_s = \frac{3K^2 - I(I+1)}{(I+1)(2I+3)} Q_0$$

$\mu = 0.46(4)\ \mu_N$, $Q_0 = 8.8(1)\ eb$ 1971/1988
$\mu = 0.360(7)\ \mu_N$, $Q_0 = 8.7(2)\ eb$ UD/GIT 2013
$\mu = 0.366(6)\ \mu_N$, $Q_0 = 8.71(6)\ eb$ UD 2021

← HFS measurements / *ab initio* calculations

5/2$^+$ [633]

^{229g}Th nuclear ground state

$\langle r^2\rangle^{229} = 33.1(2)\ \mathrm{fm}^2$

Temporal variation of fundamental constants

Th-229: the most sensitive probe in a search for variations of the fundamental coupling constants

V. Flambaum: Phys. Rev. Lett. **97**, 092502 (2006)
A. C. Hayes et al., Phys. Rev. C **78**, 024311 (2008)
B. E. Litvinova et al., Phys. Rev. C **79**, 064303 (2009)

Enhancement in sensitivity (10^3-10^5) results from the near perfect cancellation of two ~1.4 MeV contributions to the nuclear level energies.

Sensitivity factor K to the change of the fine-structure constant α:

$$K = \frac{\Delta E_C}{E_{is}} = \frac{1}{E_{is}} \cdot \left(-485 MeV \left(\frac{\Delta r^2}{r^2} \right) + 11.6 MeV \left(\frac{\Delta Q_0}{Q_0} \right) \right)$$

J. C. Berengut et al., Phys. Rev. Lett 102, 210801 (2009)

Atom, transition	K
$^{27}Al^+$, $^1S_0$-$^3P_0$	0.008
^{87}Sr, $^1S_0$-$^3P_0$	0.06
$^{171}Yb^+$, $^2S_{1/2}$-$^2D_{3/2}$	1.0
$^{171}Yb^+$, $^2S_{1/2}$-$^2F_{7/2}$	-6.0
$^{199}Hg^+$, $^2S_{1/2}$-$^2D_{5/2}$	-2.9
HCI	~ 100
^{229}Th-^{229m}Th	4(6)×10^4 (PTB) 4(13)×10^3 (RIKEN)

Fine strucutre constant variations

PTB

$$K = \frac{\Delta E_C}{E_{is}} = \frac{1}{E_{is}} \cdot \left(-485 MeV \left(\frac{\Delta \langle r^2 \rangle}{\langle r^2 \rangle} \right) + 11.6 MeV \left(\frac{\Delta Q_0}{Q_0} \right) \right)$$

J. C. Berengut et al., Phys. Rev. Lett 102, 210801 (2009)

Open Questions:

- $\Delta \langle r^2 \rangle$
- Octupole deformation?

$$\frac{\Delta Q_0}{Q_0} = 1.791(2)$$

K. Beeks et al. arXiv:2407.17300v1 (2024)

	K-value
Fadeev et al. PRA 102,052833 (2020)	-8200 (2500)
arXiv:2407.17300v1	5900 (2300)

$$E_C = \frac{3q_e^2 Z^2}{5R_0} \left(1 - \frac{1}{4\pi}\beta_2^2 - \frac{5}{14\pi}\beta_3^2 \right), \qquad (16)$$

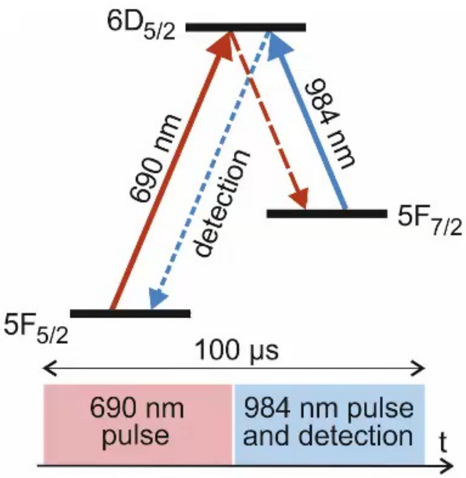
where β_2 and β_3 are the quadrupole and octupole deformations, respectively, and where we omit $O(\beta_n^3)$ terms.

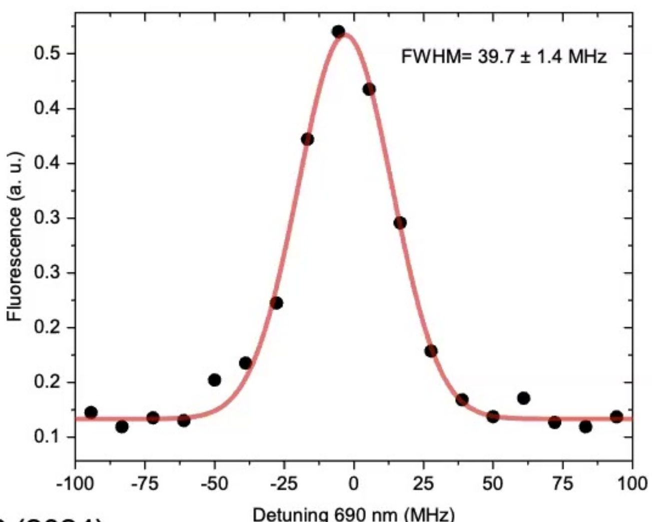
Th-230 spectroscopy

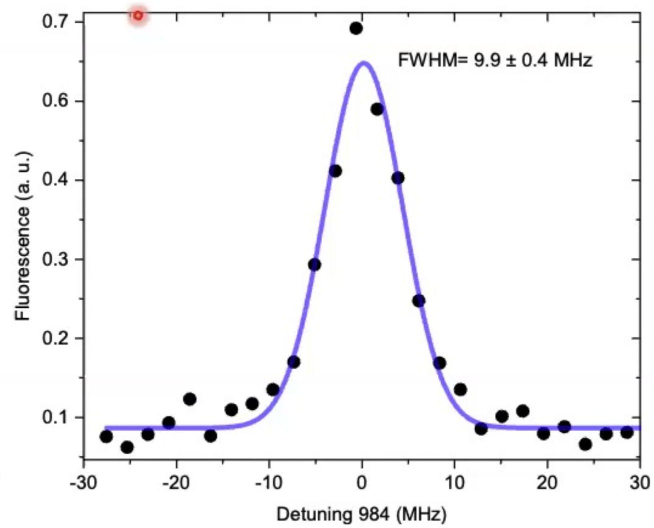
$^{230}Th^{3+}$ fluorescence signal

Detection: free from stray laser light

Th^{3+} storage time ~ 5000 s





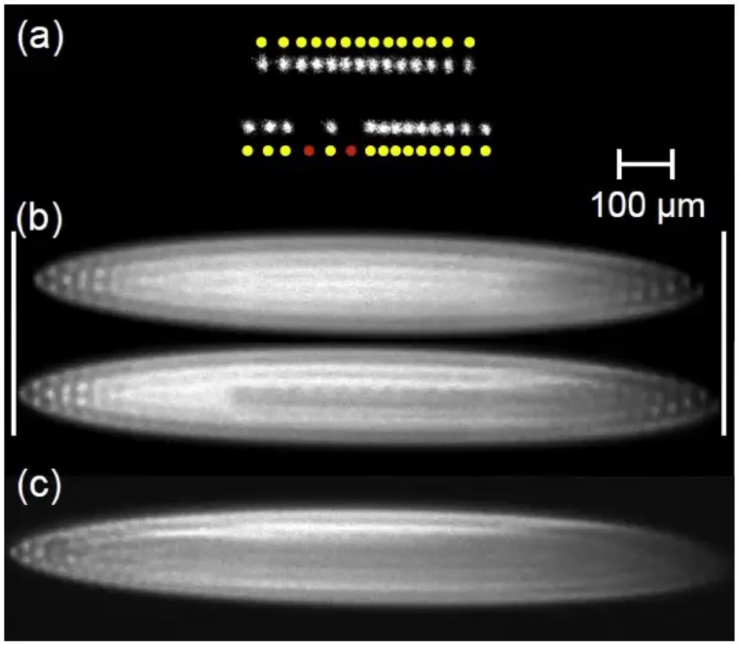


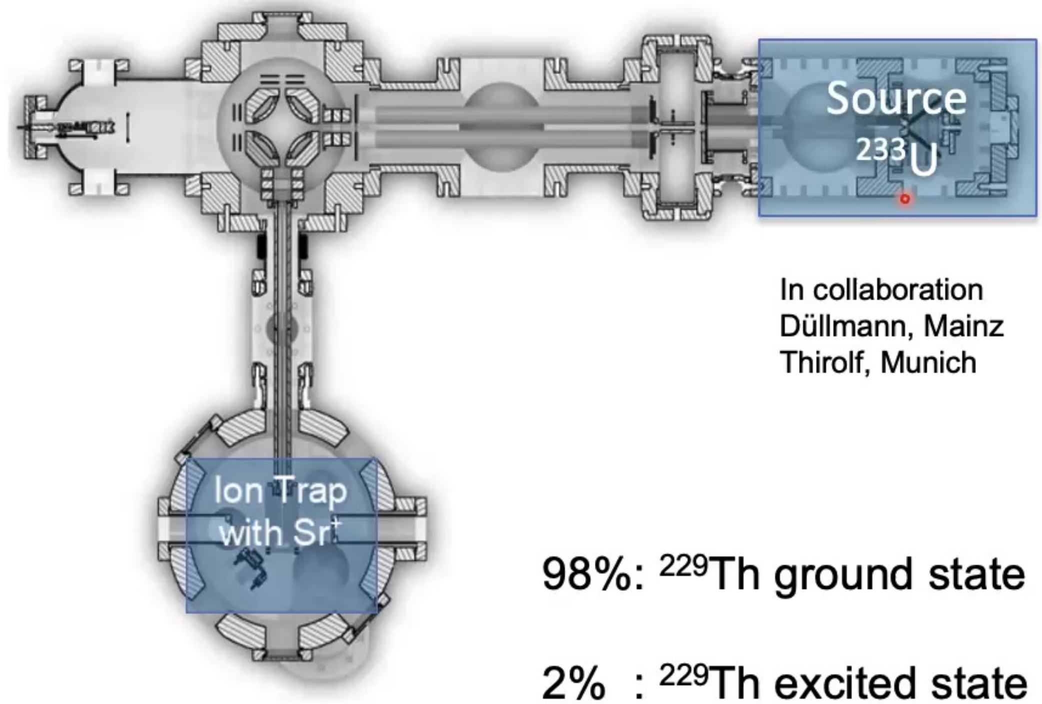
Scan of the 690 nm & 984 nm resonances with sympathetically cooled $^{230}Th^{3+}$ ions.

G. Zitzer et al., PRA 109, 033116 (2024)

Th-229 spectroscopy

$^{229}Th^{3+}$ sympathetically cooled with $^{88}Sr^{+}$





In collaboration
Düllmann, Mainz
Thirolf, Munich

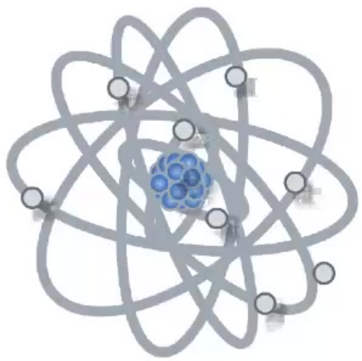
98%: ^{229}Th ground state

2% : ^{229}Th excited state

G. Zitzer et al., Phys. Rev. A **109**, 033116 (2024)

Outlook

- Introduction Th-229
- First laser excitation of the Th-229 nucleus (+ Implications)
- Hyperfine Spectroscopy of Th-229
- **Challenges towards a nuclear clock**



Towoards a nuclear clock: VUV Laser

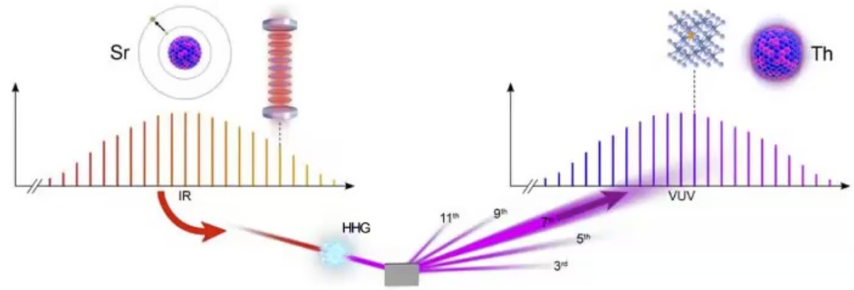
PTB

- Development of a narrowband VUV-laser

?

148.3821721174 nm

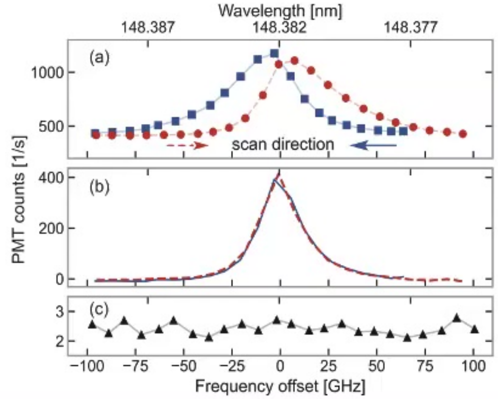
- High-Harmonic Generation

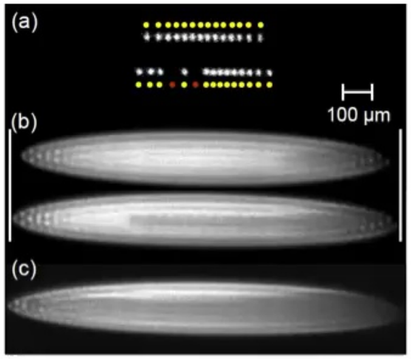


- (Solid-State) Second-Harmonic Generation
- Atomic vapors
- ...

Summary

- Laser excitation of the ^{229}Th nucleus
- Consistent results with UCLA / JILA
- Next: laser spectroscopy of Ions





European Research Council

THORIUM NUCLEAR CLOCK

DQmat

Thank you for your attention!

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