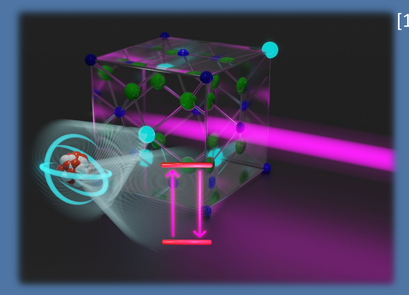
The Road to Laser Excitation of the Th-229 Nucleus in a $^{229}Th:CaF_2$ Crystal Environment


[1]

Fabian Schaden, T. Pronebner, I. Morawetz, L. Toscani De Col, A. Leitner, M. Bartokos, G. A. Kazakov, K. Beeks, T. Sikorsky, and T. Schumm

Vienna Center for Quantum Science and Technology, Atominstitut, TU Wien, Vienna, Austria

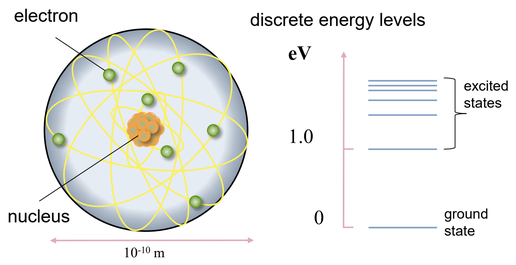
J. Tiedau, M. V. Okhapkin, K. Zhang, J. Thielking, G. Zitzer, E. Peik

Physikalisch-Technische Bundesanstalt, 38116 Braunschweig, Germany

Contact: fabian.schaden@tuwien.ac.at

Introduction

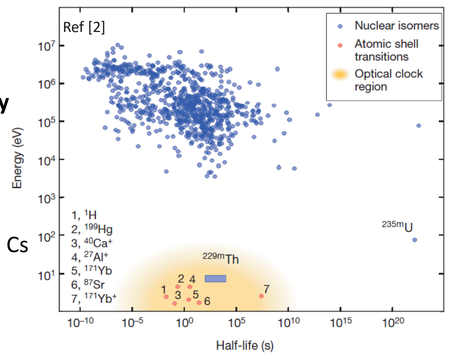
Atomic physics



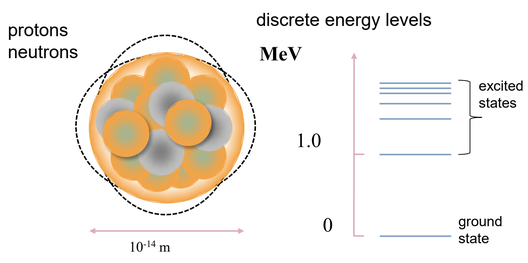
main tool of study: **laser spectroscopy**

Atomic spectroscopy

- Provides a frequency standard
- 1s ≡ 9.192.631.770 oscillations in Cs
- Crucial for fundamental research
- Applications: GPS, metrology
- Miniaturization possible



Nuclear physics



main tool of study: **particle accelerators**

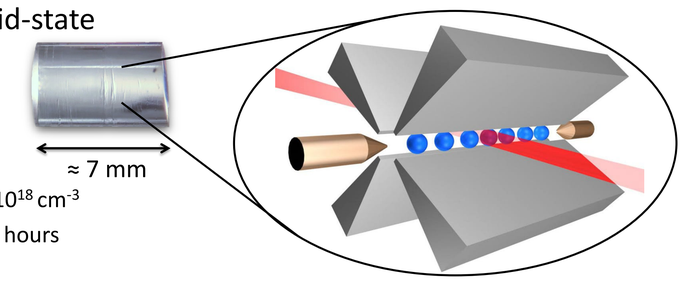
Nuclear spectroscopy

- No frequency standard
- Used in fundamental research
- Mössbauer spectroscopy
- No direct metrology applications
- No miniaturization?

A Solid-State Approach to Ion Trapping

Storing the Thorium ions into solid-state

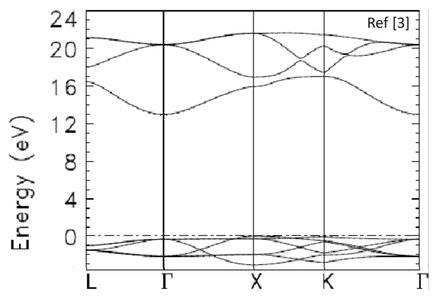
- Trapping frequency MHz->THz
- Recoil Energy 30kHz
- No micromotion
- Lamb Dicke regime
- Massive increase of density 10^{10} cm^{-3} → 10^{18} cm^{-3}
- Lifetime increased from few hours → 10^8 hours

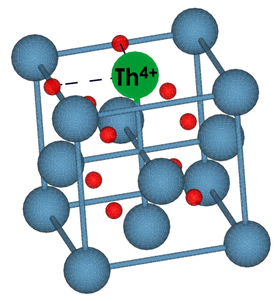


Choosing the right host material

- VUV transparent → high bandgap
- 1st Ionization energy>isomer energy

} CaF_2





Pure CaF_2

- F^- is surrounded by 4 Ca^{2+}
- Ca^{2+} is surrounded by 8 F^-
- Electronic bandgap ~12eV
- Optical bandgap ~10eV

Doping with Th

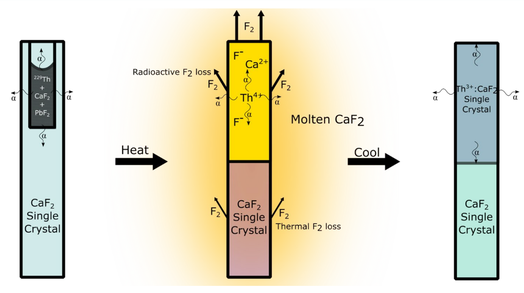
- Replaces a Ca atom
- Th has 4 valence electrons

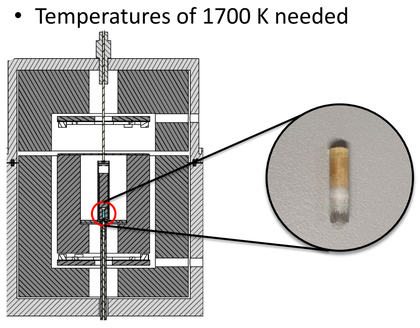
→ Needs 2 interstitial F^-

Growing Doped CaF_2 Crystals[4]

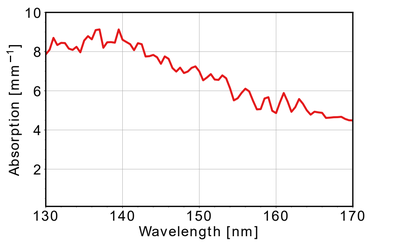
Vertical Gradient Freeze method

- No residue
- Shape determined by crucible
- Slow growth speed is possible (less defects)
- Down scalable (but difficult)
- Growth process takes days
- 1 mm/hr crystallization speed
- Temperatures of 1700 K needed

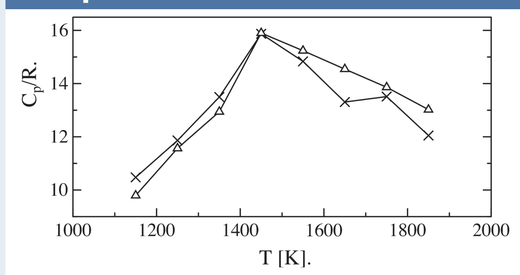




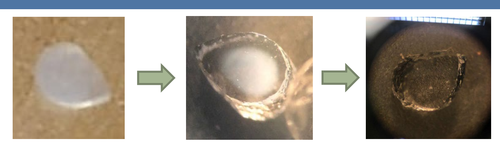
- High concentration (200 ppm)
- Turned orange (should be transparent)
- VUV opaque (even undoped part)



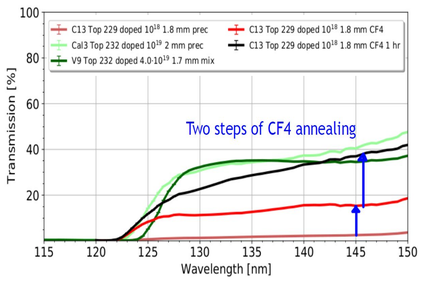
Superionic Fluoride Transfer[5]



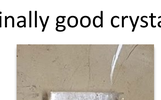
- Superionic phase of CaF_2 (> 1400 K)
- F^- becomes mobile and volatile
- Ca^{2+} remains at the lattice position
- Heated under vacuum → Significant F loss (reason for VUV opaqueness)
- Heated under CF_4 atmosphere → F can be replenished



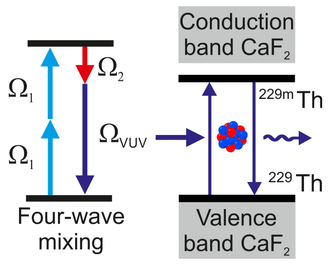
- Extreme fluoride deficient $^{232}Th:CaF_2$ (heated for 8 hrs at 1500 K)
- Annealing under CF_4 atmosphere for one hour at 1500 K
- Annealed it for one more hour
- Repeated same procedure for $^{229}Th:CaF_2$

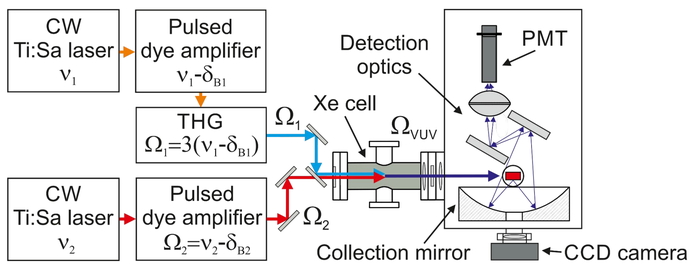


- Color disappeared
- VUV transmission: 40 % and more
- Finally good crystals!!

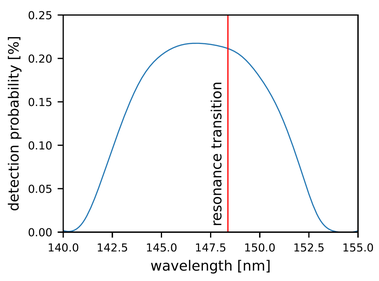


Laser Excitation of ^{229}Th[6]



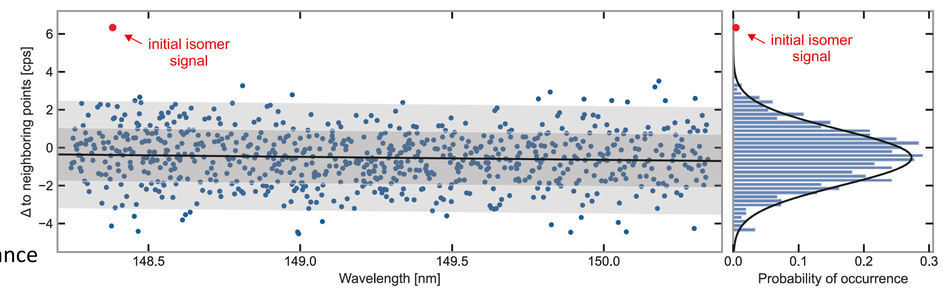


Laser	C10	X2
• E_{pulse} : 10 µJ	Ø3.2×3.7 mm	1.65×1.65×1.75 mm
• FWHM: 20 GHz	22 kBq	66 kBq
• ν_{rep}: 30 Hz	3×10^{17} cm^{-3}	5×10^{18} cm^{-3}
• A = 1 mm^2	30 %	40 %
• **Pumping rate:**	**≈ 10^6 s^{-1}**	**≈ 10^7 s^{-1}**

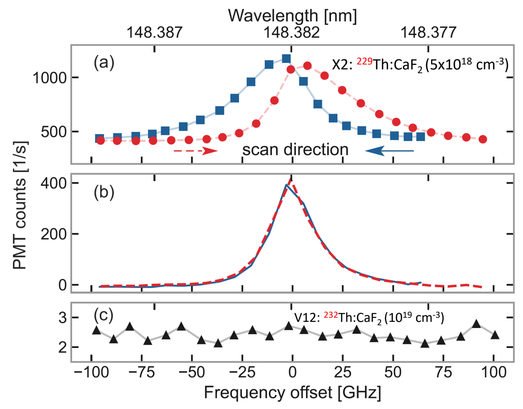


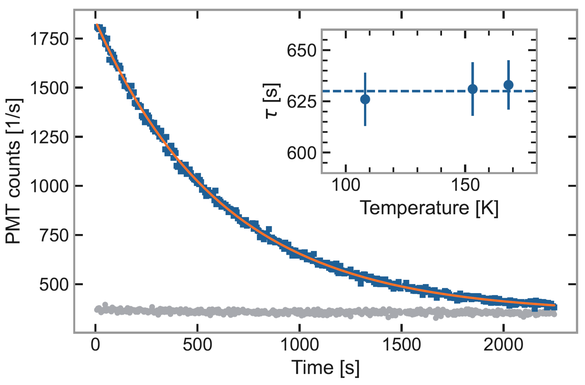
Scanning

- 120 s excitation
- 150 s detection
- 30 GHz triangular modulation
- Scanned ≈ 2 nm
- Initial isomer signal: 5σ significance



VUV ^{229m}Th Fluorescence[6]





Measurement

- ±5 GHz modulation
- Scanned from higher to lower frequency
- And from lower to higher frequency
- t_d between frequency < τ_{isomer} → Asymmetry

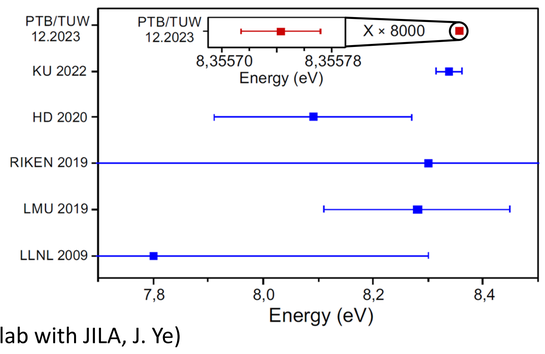
Results

- VUV signal in two different $^{229}Th:CaF_2$ crystals
- No signal in $^{232}Th:CaF_2$ crystal
- τ = 630(15) s
- λ = 148.3821(5) nm

Conclusion and Outlook

Conclusion

- Success in growing highly doped CaF_2 single crystals with varying dopants (Th, U, Np)
- Controlling charge state with fluorination
- Doped crystals used in many different experiments (Laser Spectroscopy, Uranium, SPring-8, Color Center characterization, NQR,..)
- Crystal enabled first laser spectroscopy of a ^{229}Th nucleus



Outlook:

- Resolve quadrupole structure with a frequency comb (collab with JILA, J. Ye)
- Finding NQR transition in a $^{229}Th:CaF_2$ crystal → Readout scheme for solid-state nuclear clocks (TU Wien)
- Characterizing defects that can quench the isomeric state (collab. with, Okayama University, K. Yoshimura)
- Finding a suitable material for a 148 nm CW laser (collab. with JGU Mainz, L. von der Wense)

References and Credits:

[1] Oliver Diekmann, TU Wien
[2] V. d. Wense, L. et al., Direct detection of the 229Th nuclear clock transition. Nature 533, 47–51, (2016).
[3] Bohara, B., Franklin L., et al, Diola. Ab-initio Calculations of Electronic and Transport Properties of Calcium Fluoride (CaF2), (2016).
[4] Beeks, K., Sikorsky T., et al. Growth and characterization of thorium-doped calcium fluoride single crystals. Sci Rep 13, 3897 (2023).
[5] Beeks, K., Sikorsky, T., et al. Optical transmission enhancement of ionic crystals via superionic fluoride transfer: Growing VUV-transparent radioactive crystals. Phys. Rev. B, 109, 094111. (2024).
[6] J. Tiedau*, M. V. Okhapkin*, K. Zhang*, et al. and F. Schaden*, T. Pronebner, I. Morawetz, et al Laser Excitation of the Th-229 Nucleus. Phys. Rev. Lett., 132, 182501. (2024).

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