

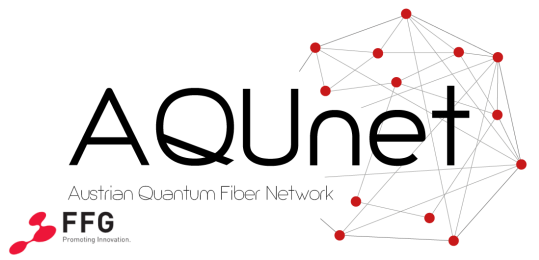


European Research Council
Established by the European Commission
Synergy project 856415



Th

Nuclear laser spectroscopy of ^{229}Th in CaF_2 crystals

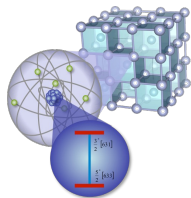


Funded by the Austria
Research Promotion Agency Infrastructure project 884678



Thorsten SCHUMM
TU Wien, Atominstitut

PSAS2026 18.05.2026



The „solid state approach“ to ^{229}Th nuclear spectroscopy

- Basic concept and implementation

Nuclear quadrupole structure

- Nuclear – solid coupling
- Separating nuclear properties and material parameters

JILA experiment

- Absolute Mössbauer spectroscopy
- Temperature dependence
- Towards a solid-state nuclear clock

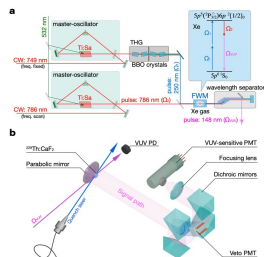
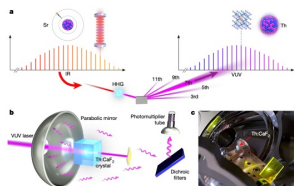
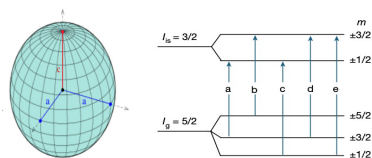
JILA

OKAYAMA experiment

- Multiple defect centers
- Site-dependent properties



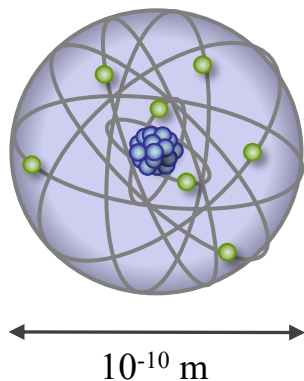
OKAYAMA UNIVERSITY



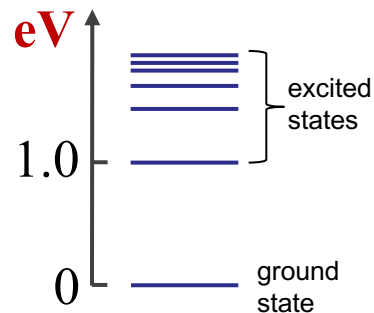


or contact:

thorsten.schumm@tuwien.ac.at

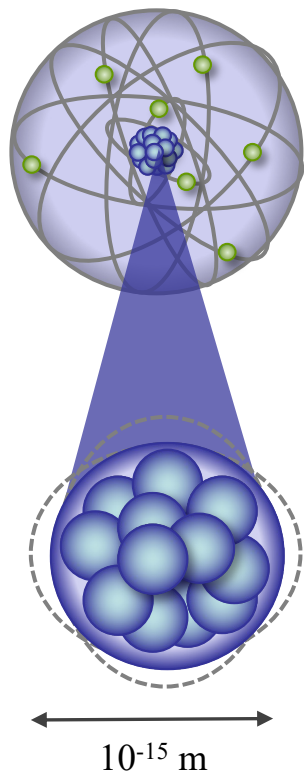


discrete energy levels

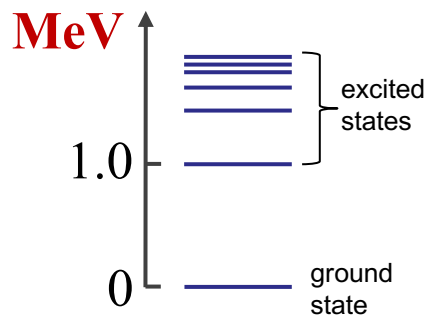
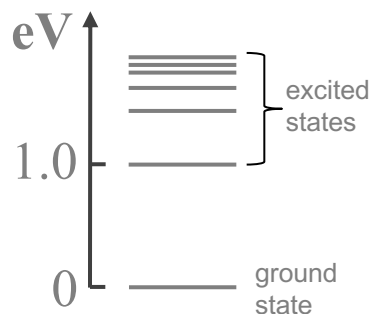


Atomic quantum state manipulation

- laser spectroscopy
- atomic clocks, quantum sensing
- quantum computation, communication
- many-body physics (BEC...)
- fundamental physics



discrete energy levels

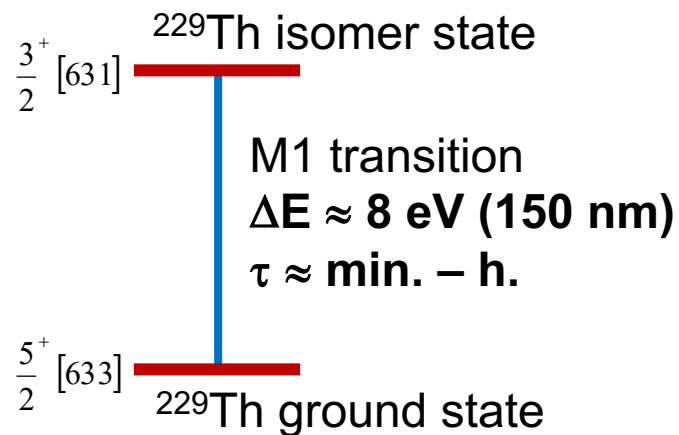
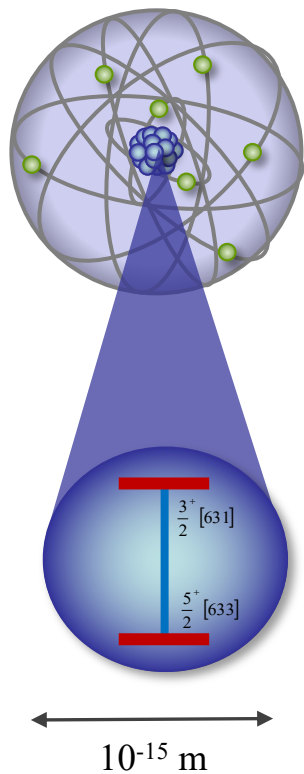


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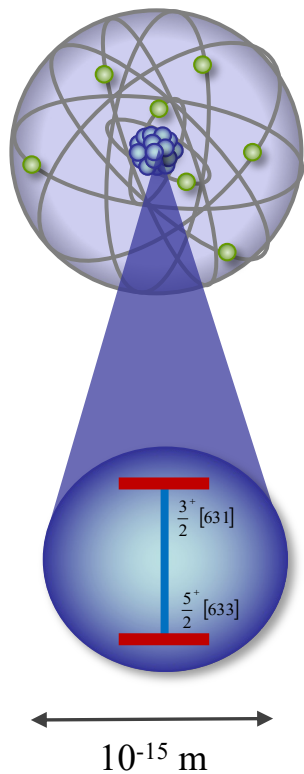
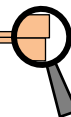
Nuclear quantum state manipulation?

- **laser spectroscopy???**
- material science
- fundamental physics
- ...

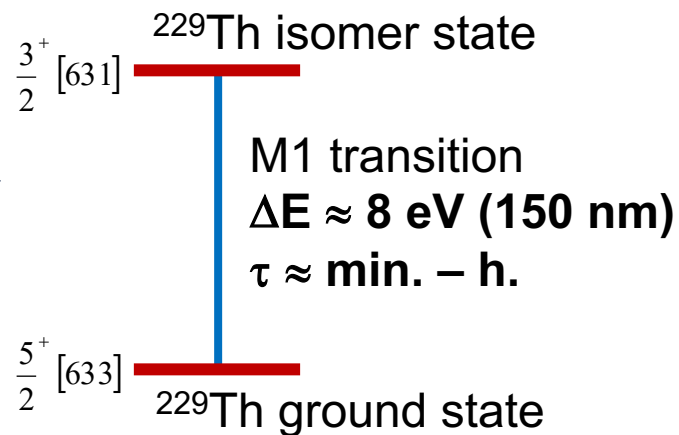




Concept: ^{229}Th nuclear laser excitation

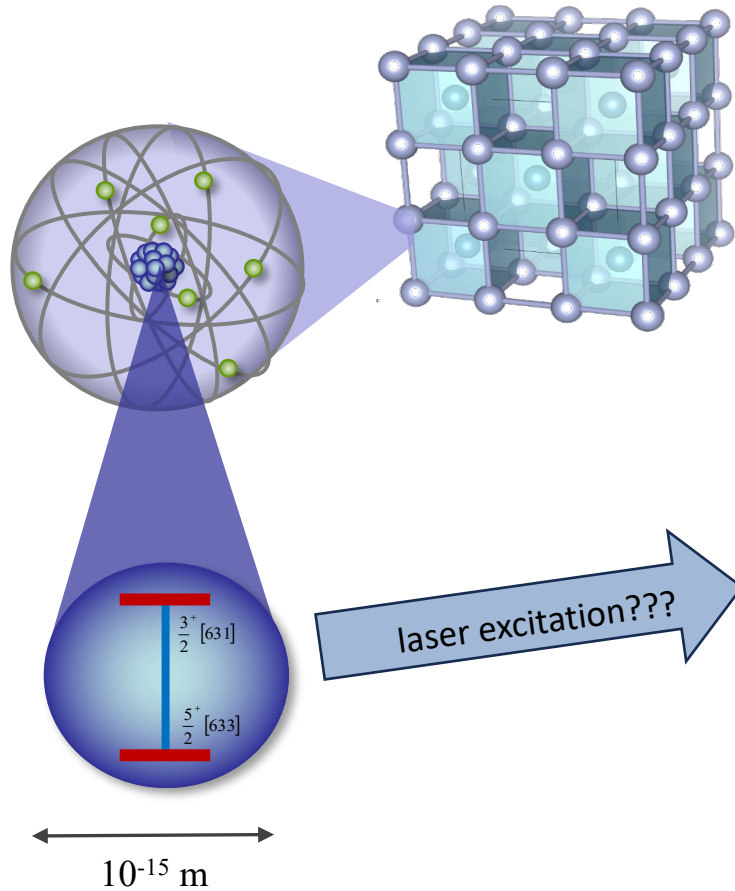


laser excitation???



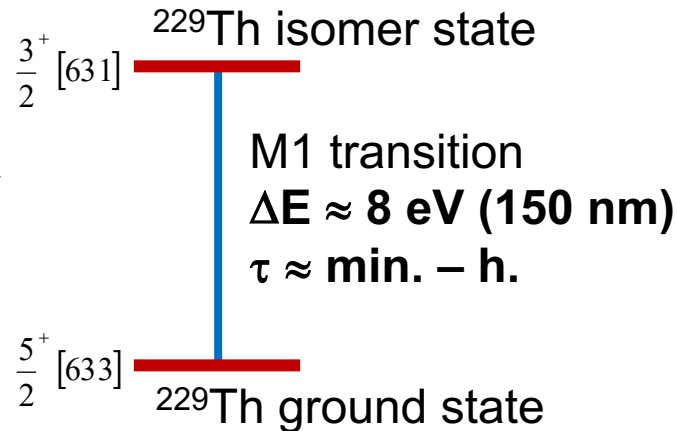


Concept: ^{229}Th nuclear laser excitation in CaF_2



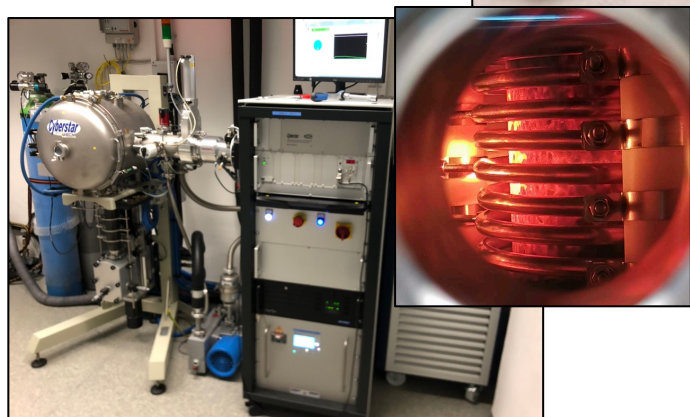
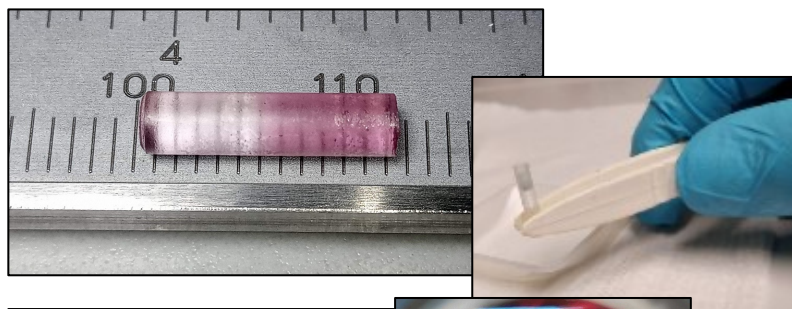
The „solid state approach“:

- Th nuclei inside VUV transparent material
- large band ($\sim 10 \text{ eV}$) gap \rightarrow fluoride crystals
- doping, implantation, diffusion...
- control of electronic environment: [Rn]
- **a solid-state optical nuclear clock?**





> 10 years of painful method development...



- dedicated lab for growth of radioactive dopants (only one in Europe)
- miniaturized crystal growth to increase doping concentration
- fluorine annealing to reduce defects
- in-house cutting + polishing
- characterization (optical, radiological)
- 1 crystal (attempt) takes 5 days

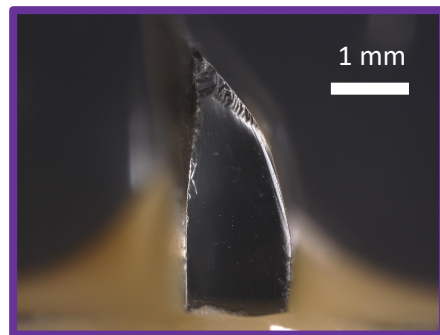
currently working on:

- pulling single crystal fibers
- growing bulk ThF₄ crystals

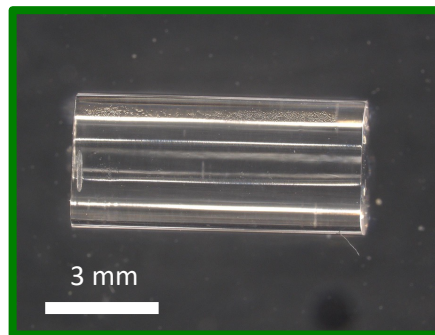


3 crystals from Vienna with different „history“ and concentration

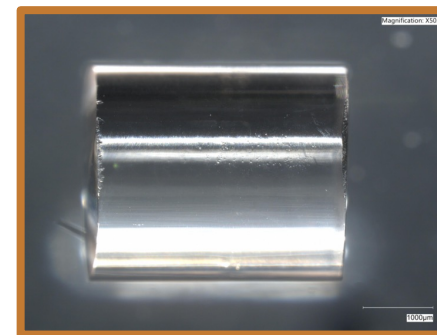
X2: $4 \times 10^{18} \text{ cm}^{-3}$
19 May 2021



C13: $0.8 \times 10^{18} \text{ cm}^{-3}$
7 Dec 2020

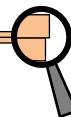


C10: $0.3 \times 10^{18} \text{ cm}^{-3}$
23 Nov 2020



...will be used in ALL experiments shown today

* apart from the UCLA experiment on slide 4

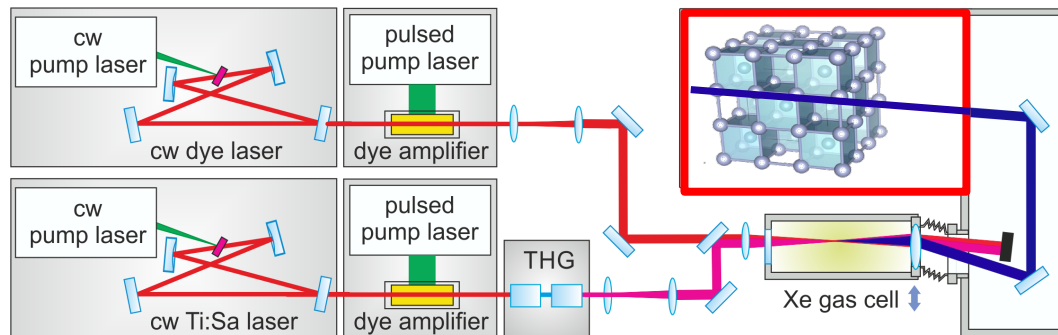


PTB Experiment





PTB Experiment

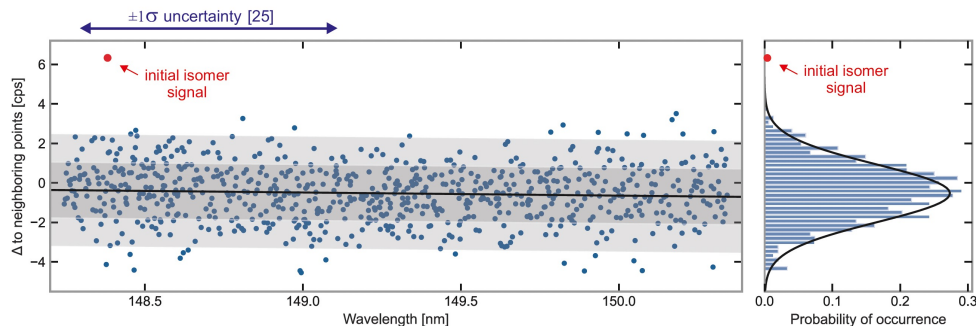
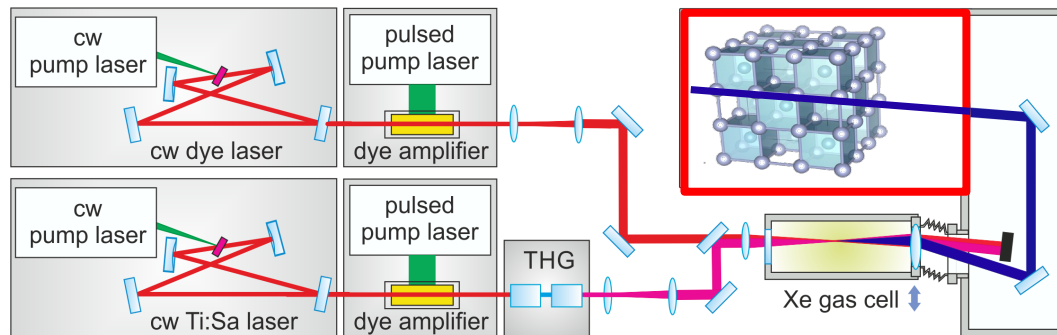




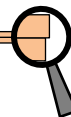
First laser excitation of ^{229}Th in 2024



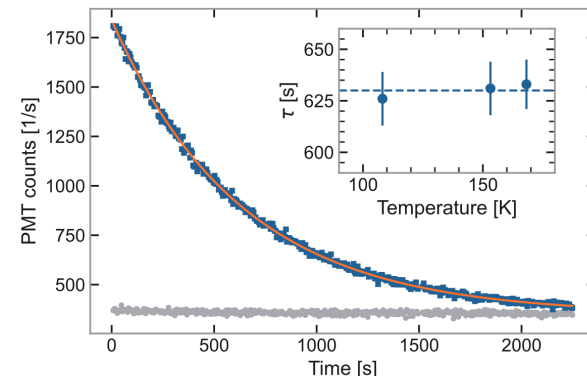
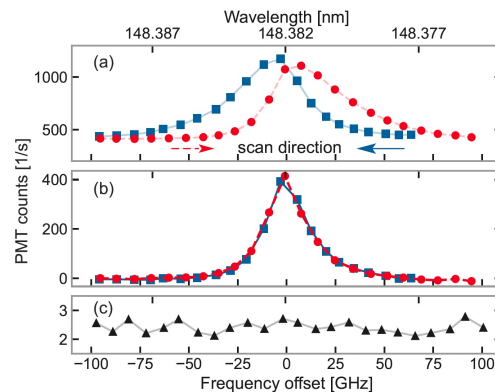
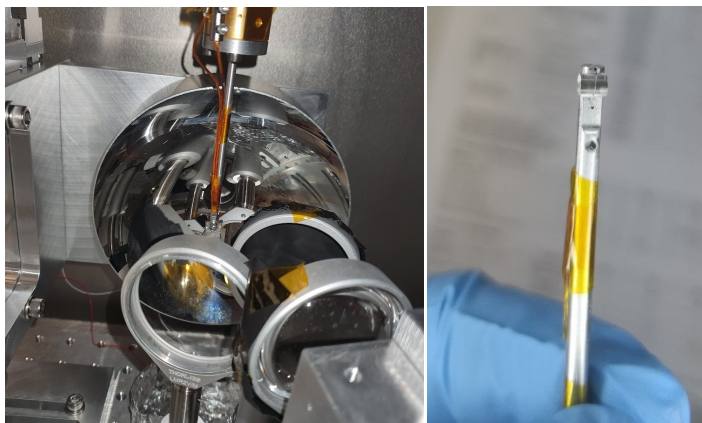
PTB Experiment



scanning for almost a year...



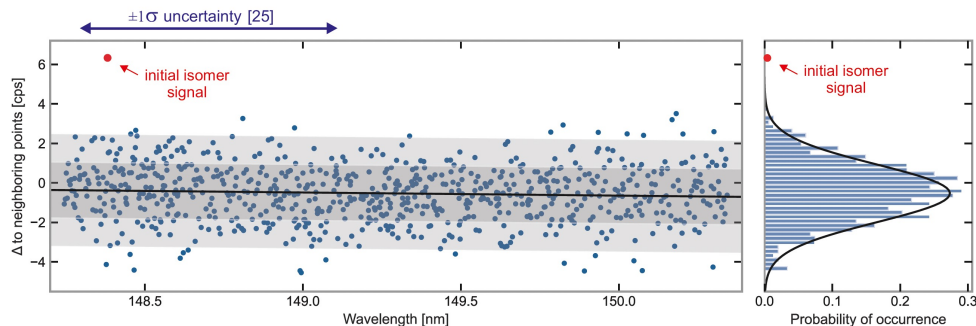
PTB Experiment

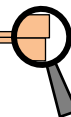


$$\nu_{\text{isomer}} = 2020.409(7) \text{ THz}$$

laser linewidth limited

$$\tau_{\frac{1}{2}} = 670(102) \text{ s (in crystal)}$$



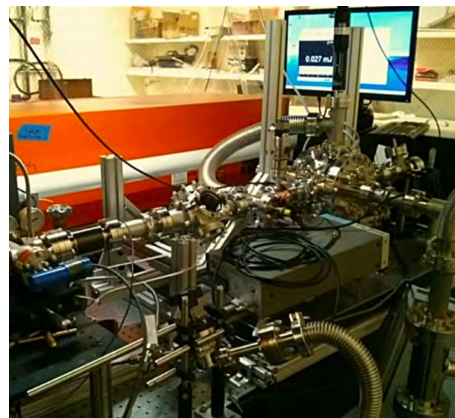


PTB Experiment

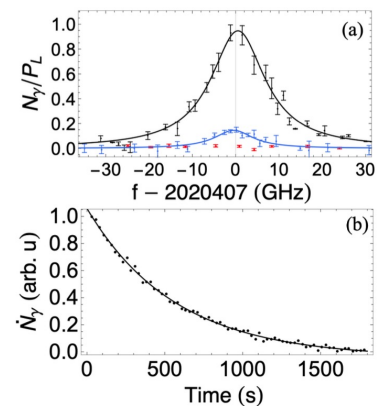


Host: CaF_2

UCLA Experiment

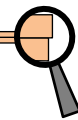


Host: LiSrAlF_6

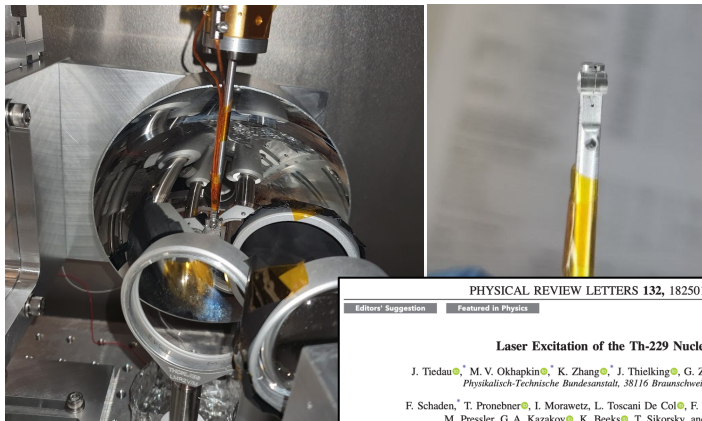




First laser excitation of ^{229}Th in 2024



PTB Experiment



PHYSICAL REVIEW LETTERS 132, 182501 (2024)

Editors' Suggestion Featured In Physics

Laser Excitation of the Th-229 Nucleus

J. Tiedau¹, M. V. Okhapkin², K. Zhang³, J. Thielking⁴, G. Zitzer⁵, and E. Peik¹
¹Physikalisch-Technische Bundesanstalt, 38116 Braunschweig, Germany

F. Schaden¹, T. Pronebner⁶, I. Morawetz, L. Toscani De Col⁶, F. Schneider⁶, A. Leitner, M. Pressler, G. A. Karakov⁶, K. Becks⁶, T. Sikorsky, and T. Schumm⁶
⁶Vienna Center for Quantum Science and Technology, Atominstiat, TU Wien, 1020 Vienna, Austria

(Received 5 February 2024; revised 12 March 2024; accepted 14 March 2024; published 29 April 2024)

The 8.4 eV nuclear isomer state in Th-229 is resonantly excited in Th-doped CaF_2 crystals using a tabletop laser system. A resonance fluorescence signal is observed in two crystals with different Th-229 dopant concentrations, while it is absent in a control experiment using Th-232. The nuclear resonance for the Th^{4+} ions in Th:CaF_2 is measured at the wavelength 148.3821(5) nm, frequency 2020.409(7) THz, and 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 toward Th-229 nuclear laser spectroscopy and realizing optical nuclear clocks.

DOI: 10.1103/PhysRevLett.132.182501

Host: CaF_2

Isomer energy:

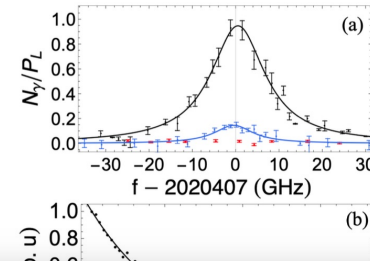
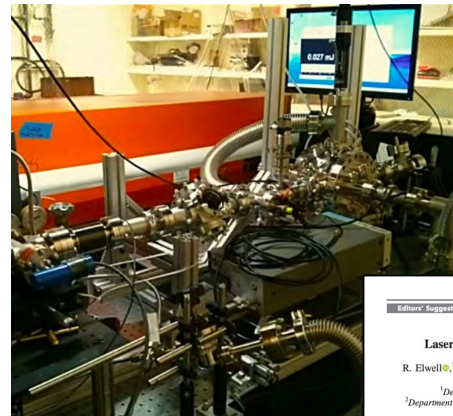
8.338(24) eV or 148.71(42) nm

Isomer lifetime:

670(102) s (in crystal)

April 2024

UCLA Experiment



PHYSICAL REVIEW LETTERS 133, 013201 (2024)

Editors' Suggestion Featured In Physics

Laser Excitation of the ^{229}Th Nuclear Isomeric Transition in a Solid-State Host

R. Elwell¹, Christian Schneider¹, Justin Jeet², J. E. S. Terhune³, H. W. T. Morgan⁴, A. N. Alexandrova^{5,6},
H. B. Tran Tan⁶, Andrei Derevianko⁶, and Eric R. Hudson⁶
¹Department of Physics and Astronomy, University of California, Los Angeles, California 90095, USA
²Department of Chemistry and Biochemistry, University of California, Los Angeles, Los Angeles, California 90095, USA
³Department of Physics, University of Nevada, Reno, Nevada 89557, USA
⁴Los Alamos National Laboratory, P.O. Box 1663, Los Alamos, New Mexico 87545, USA
⁵Challenge Institute for Quantum Computation, University of California Los Angeles, Los Angeles, California 90095, USA
⁶Center for Quantum Science and Engineering, University of California Los Angeles, Los Angeles, California 90095, USA

(Received 22 March 2024; revised 19 April 2024; accepted 17 May 2024; published 2 July 2024)

LiSrAlF_6 crystals doped with ^{229}Th are used in a laser-based search for the nuclear isomeric transition. Two spectroscopic features near the nuclear transition energy are observed. The first is a broad excitation feature that produces redshifted fluorescence that decays with a timescale of a few seconds. The second is a narrow, laser-line-width-limited spectral feature at 148.38219(4) nm [2020.409(7) THz], which decays with a lifetime of 568(13) s for a nucleus isolated in vacuum. This feature is assigned to the excitation of the ^{229}Th nuclear isomeric state, whose energy is found to be 8.35573(2) eV in $^{229}\text{Th:LiSrAlF}_6$.

DOI: 10.1103/PhysRevLett.133.013201

Host: LiSrAlF_6

Isomer energy:

148.38219(4)stat (20)sys nm

Isomer lifetime:

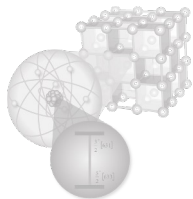
568(13)stat (20)sys s (in crystal)

July 2024



or contact:

thorsten.schumm@tuwien.ac.at

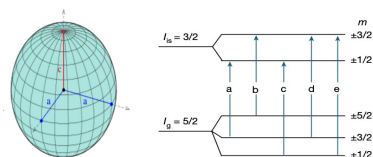


The „solid state approach“ to ^{229}Th nuclear spectroscopy

- Basic concept and implementation

Nuclear quadrupole structure

- Nuclear – solid coupling
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JILA experiment

- Absolute Mössbauer spectroscopy
- Temperature dependence
- Towards a solid-state nuclear clock

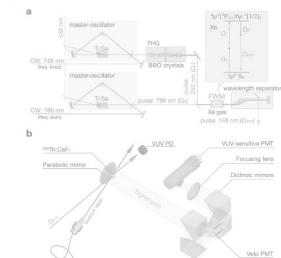
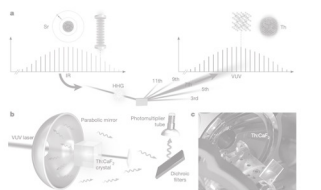
JILA

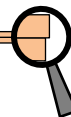
OKAYAMA experiment

- Multiple defect centers
- Site-dependent properties

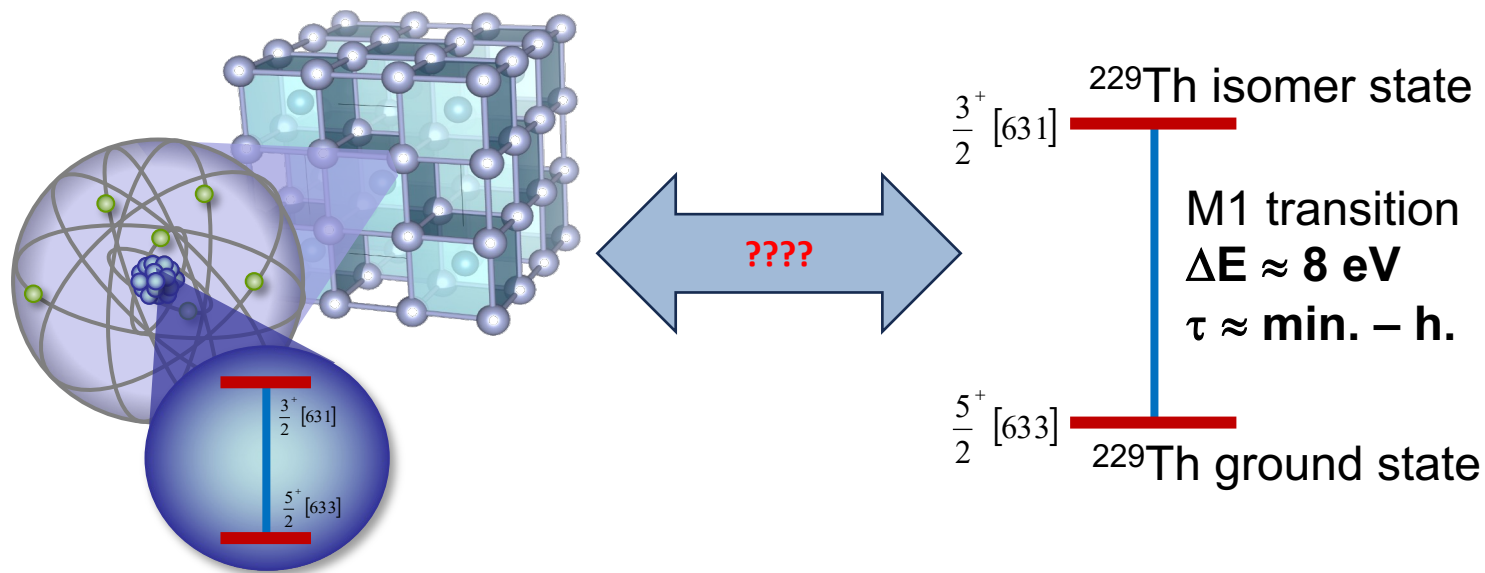


OKAYAMA UNIVERSITY



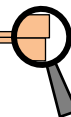


What about interactions between crystal and nuclear levels?

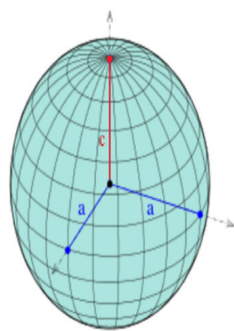




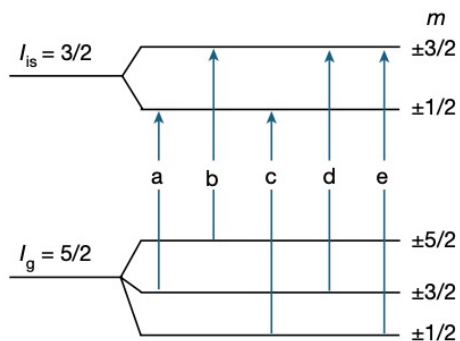
Nuclear quadrupole structure in $^{229}\text{Th}:\text{CaF}_2$



Quadrupole structure determined by **nuclear** and **material parameters**



Prolate



$$H = \frac{QV_{zz}}{4I(2I-1)} [3I_z^2 - \mathbf{I}^2 + \eta(I_x^2 - I_y^2)]$$

Q – (intrinsic) quadrupole moment

V_{zz} – electric field gradient (EFG)

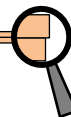
η – anisotropic parameter

„universal“

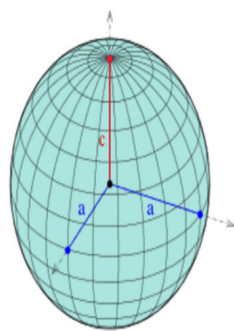
host-specific



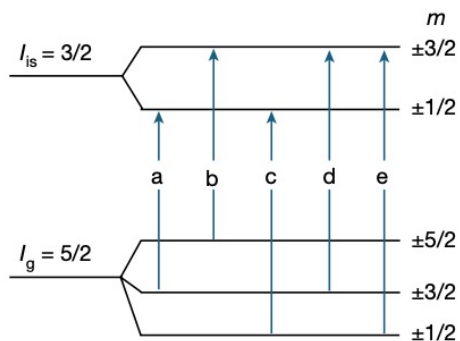
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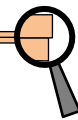
V_{zz} – electric field gradient (EFG)

η – anisotropic parameter

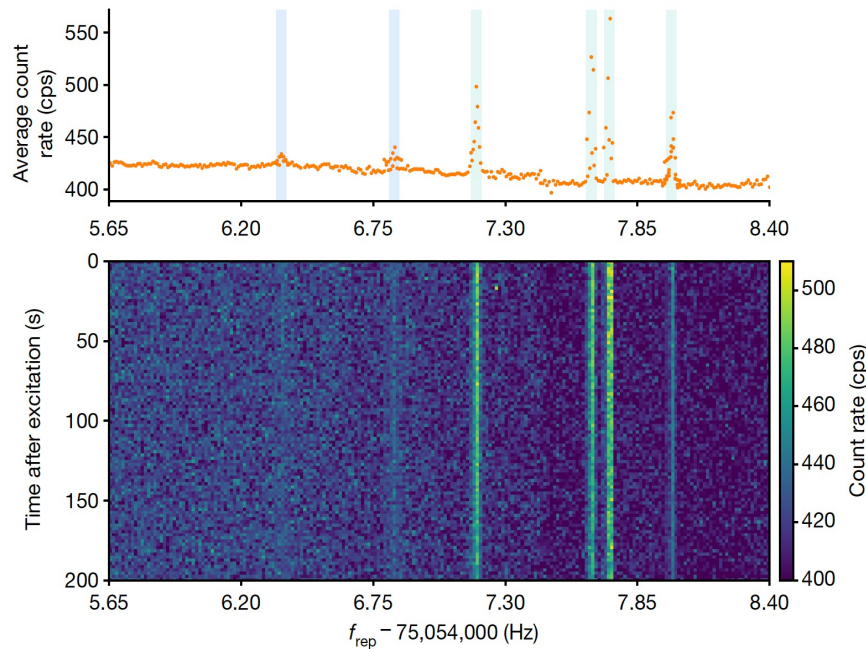
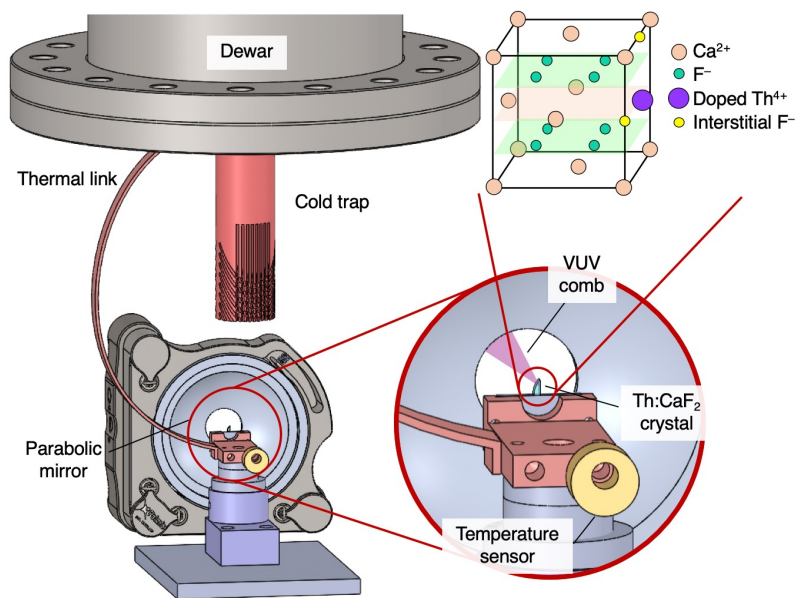
„universal“

host-specific

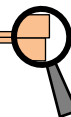
can we see these quadrupole levels...?



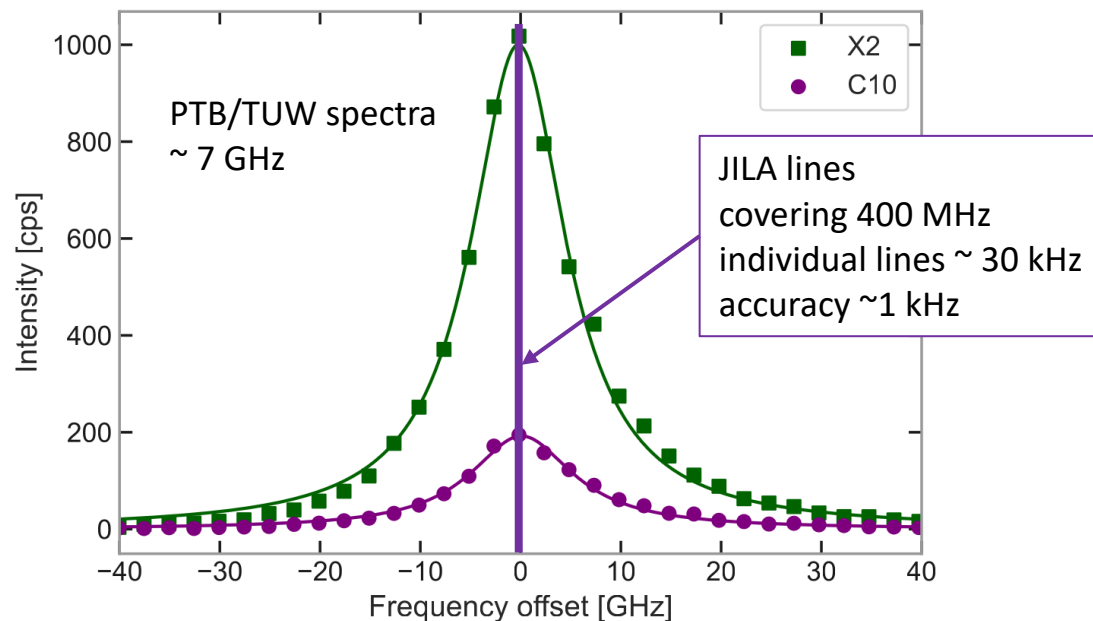
JILA experiment



- 4 strong, 2 weak lines observed
- absolute frequency determination with reference to Sr clock



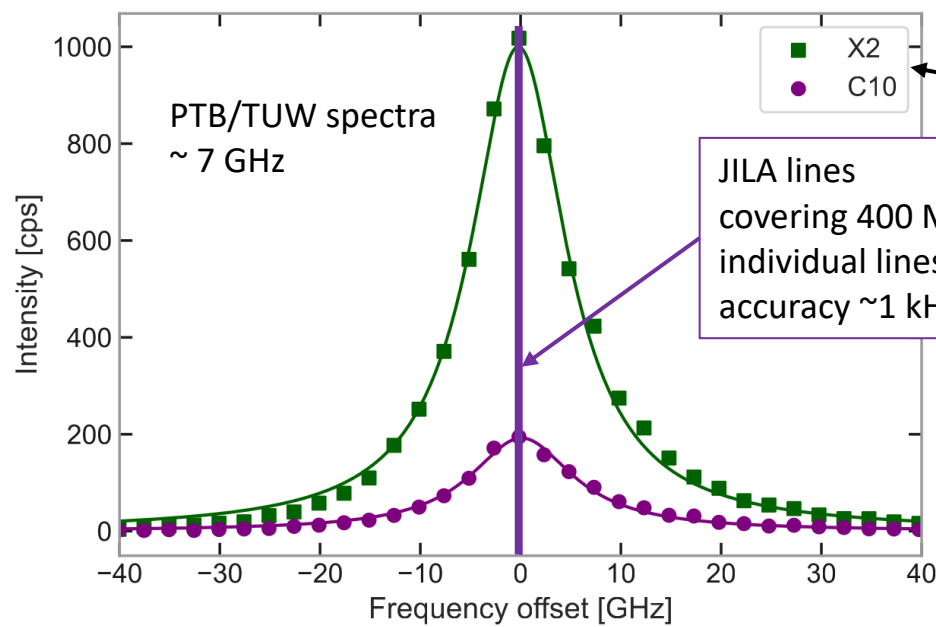
JILA experiment vs. PTB/UCLA



$$\nu_{\text{isomer}} = 2\,020\,407\,384\,335(2) \text{ kHz}$$



JILA experiment vs. PTB/UCLA



same crystals X2 and C10 used at PTB and JILA

Article

Frequency ratio of the ^{229m}Th nuclear isomeric transition and the ^{87}Sr atomic clock

<https://doi.org/10.1038/s41586-024-07839-6> Chuanbin Zhang^{1,2,3*}, Tian Qiu^{1,2,3}, Jacob S. Higgins^{1,2,3}, Jack F. Doyle^{1,2,3}, Lars von der Wense^{1,2,7}, Kjeld Beek^{1,4}, Adrian Leinert^{1,4}, Georgy A. Kazakov¹, Peng Li¹, Peter G. Thiroff¹, Thorsten Schumm¹ & Jun Ye^{1,2,3*}

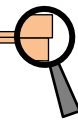
Received: 20 June 2024
Accepted: 17 July 2024
Published online: 4 September 2024

Check for updates

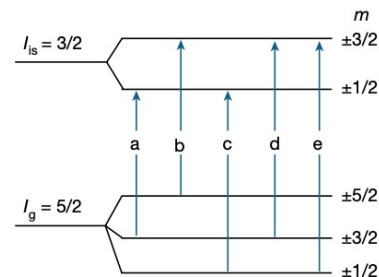
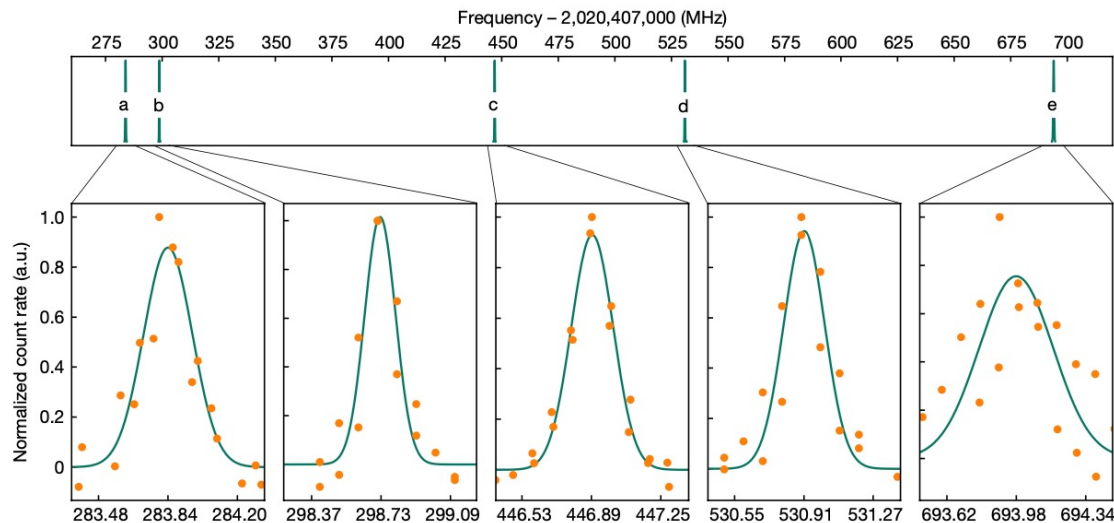
Optical atomic clocks^{1,2} use electronic energy levels to precisely keep track of time. A clock based on nuclear energy levels promises a next-generation platform for precision metrology and fundamental physics studies. Thorium-229 nuclei exhibit a uniquely low-energy nuclear transition within reach of state-of-the-art vacuum ultraviolet (VUV) laser light sources and have, therefore, been proposed for construction of a nuclear clock^{1,4}. However, quantum-state-resolved spectroscopy of the ^{229m}Th isomer to determine the underlying nuclear structure and establish a direct frequency connection with existing atomic clocks has yet to be performed. Here, we use a VUV frequency comb to directly excite the narrow ^{229m}Th nuclear clock transition in a solid-state CaF_2 host material and determine the absolute transition frequency. We stabilize the fundamental frequency comb to the JILA ^{87}Sr clock³ and coherently upconvert the fundamental to its seventh harmonic in the VUV range by using a femtosecond enhancement cavity. This VUV comb establishes a frequency link between nuclear and electronic energy levels and allows us to directly measure the frequency ratio of the ^{229m}Th nuclear clock transition and the ^{87}Sr atomic clock. We also precisely measure the nuclear quadrupole splittings and extract intrinsic properties of the isomer. These results mark the start of nuclear-based solid-state optical clocks and demonstrate the first comparison, to our knowledge, of nuclear and atomic clocks for fundamental physics studies. This work represents a confluence of precision metrology, ultrafast strong-field physics, nuclear physics and fundamental physics.

September 2024

$\nu_{\text{isomer}} = 2\,020\,407\,384\,335(2) \text{ kHz}$



JILA experiment



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)

- 4 strong + 1 additional line observed → assignment to nuclear quadrupole structure
- absolute frequency determination

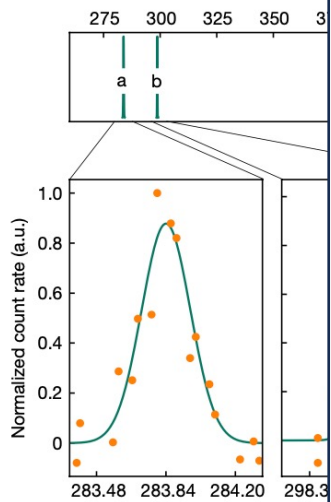
(let's not totally forget about the 2 weak lines)



Nuclear quadrupole structure in $^{229}\text{Th}:\text{CaF}_2$



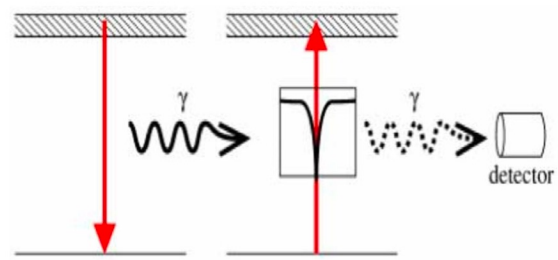
JILA experiment



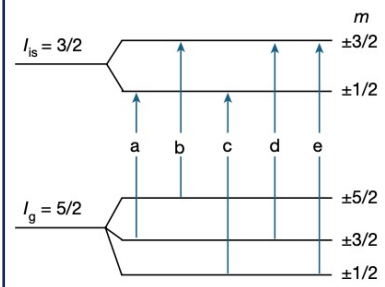
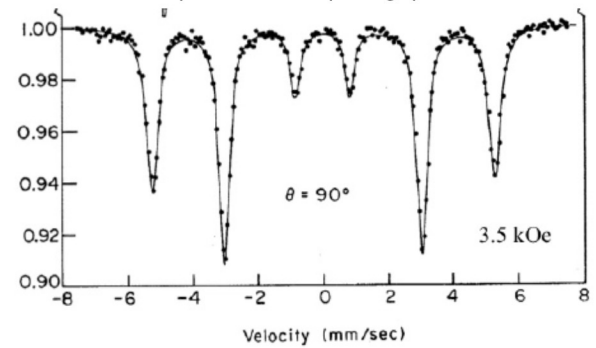
- 4 strong + 1 a
- absolute frequ

(let's not totally f

„traditional“ Mössbauer spectroscopy



Example of zeeman splitting spectrum



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)

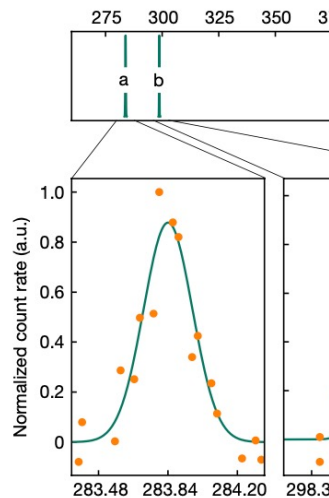
near quadrupole structure



Nuclear quadrupole structure in $^{229}\text{Th}:\text{CaF}_2$



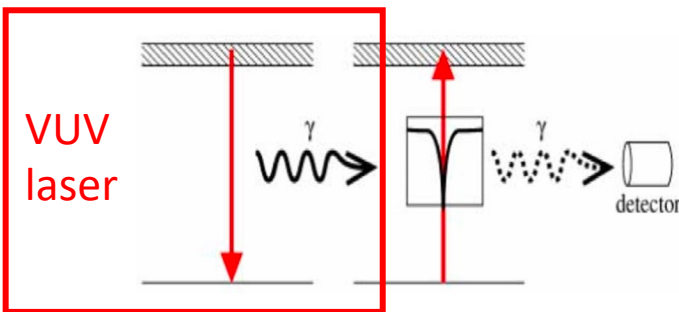
JILA experiment



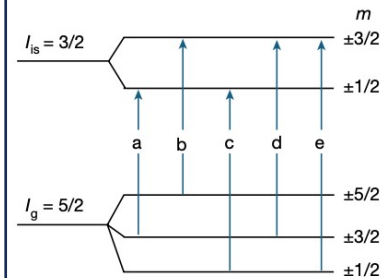
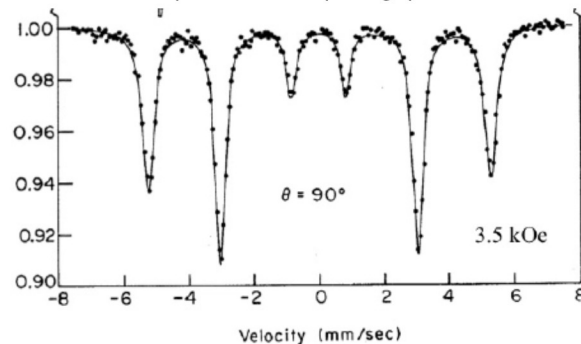
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(let's not totally f

„traditional“ Mössbauer spectroscopy



Example of zeeman splitting spectrum

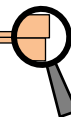


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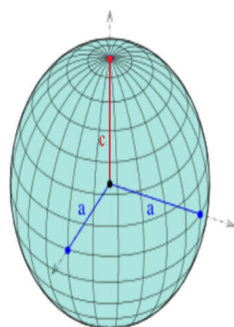
near quadrupole structure



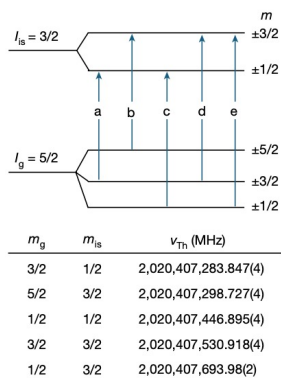
Nuclear quadrupole structure in $^{229}\text{Th}:\text{CaF}_2$



Quadrupole structure determined by **nuclear** and **material parameters**



Prolate



$$H = \frac{QV_{zz}}{4I(2I-1)} [3I_z^2 - \mathbf{I}^2 + \eta(I_x^2 - I_y^2)]$$

Q – (intrinsic) quadrupole moment

V_{zz} – electric field gradient (EFG)

η – anisotropic parameter

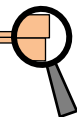


„universal“

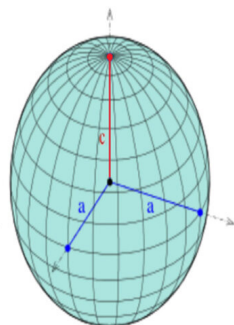
host-specific

change in quadrupole moment...

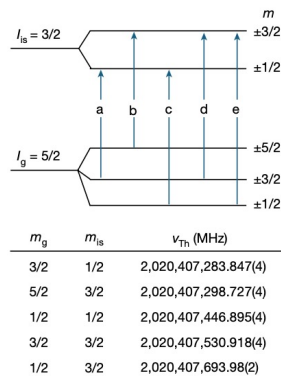
$$\Delta Q/Q = 1.791(2)\%$$



Quadrupole structure determined by nuclear and material parameters



Prolate



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η – anisotropic parameter



„universal“

host-specific

change in quadrupole moment...

nature communications



Article

<https://doi.org/10.1038/s41467-025-64191-7>

Fine-structure constant sensitivity of the Th-229 nuclear clock transition

Received: 17 December 2024

Kjeld Beeks^{1,2}, Georgy A. Kazakov¹, Fabian Schaden¹, Ira Morawetz¹,

Luca Toscani De Col¹, Thomas Riebner¹, Michael Bartokos¹,

Accepted: 8 September 2025

Tomas Sikorsky^{1,3}, Thorsten Schumm¹, Chuankun Zhang⁴, Tian Ooi⁴,

Published online: 15 October 2025

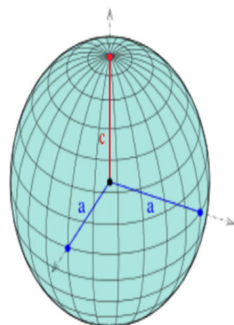
Jacob S. Higgins¹, Jack F. Doyle⁴, Jun Ye⁴ & Marianna S. Safronova⁵



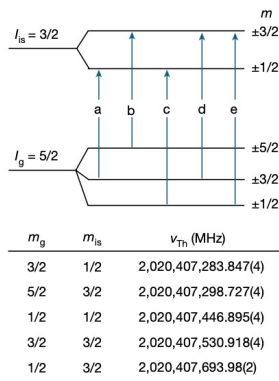
Nuclear quadrupole structure in $^{229}\text{Th}:\text{CaF}_2$



Quadrupole structure determined by **nuclear** and **material parameters**



Prolate



$$H = \frac{QV_{zz}}{4I(2I-1)} [3I_z^2 - \mathbf{I}^2 + \eta(I_x^2 - I_y^2)]$$

Q – (intrinsic) quadrupole moment

V_{zz} – electric field gradient (EFG)

η – anisotropy parameter

„universal“

host-specific

Extracted electrical field gradient (EFG)

$$V_{zz} = 109.1(7) \text{ V/\AA}^2$$

$$\eta = 0.59163(5)$$

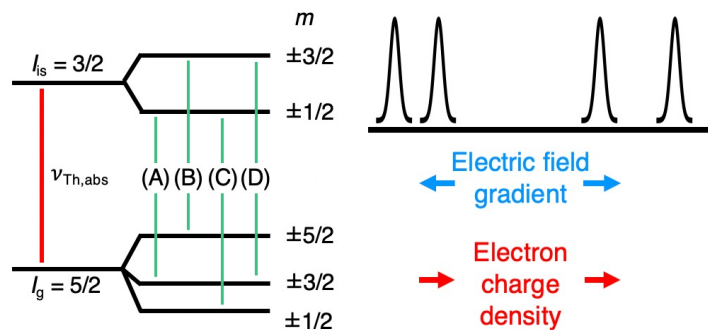
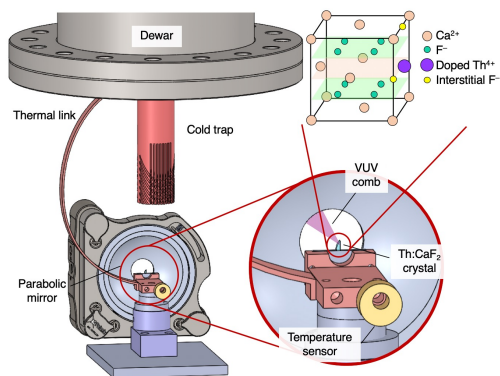
extremely well reproduced over 3 different crystals

change in quadrupole moment...

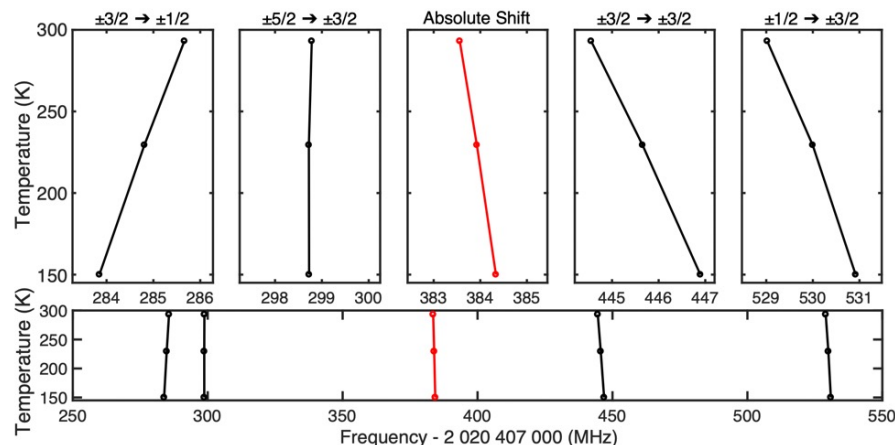
$$\Delta Q/Q = 1.791(2)\%$$



JILA experiment



tracing 4 strongest lines over temperature

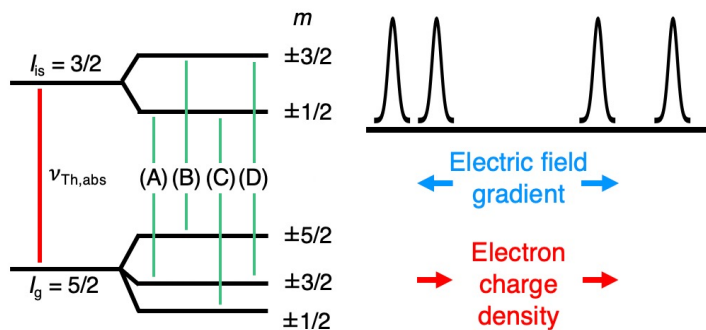
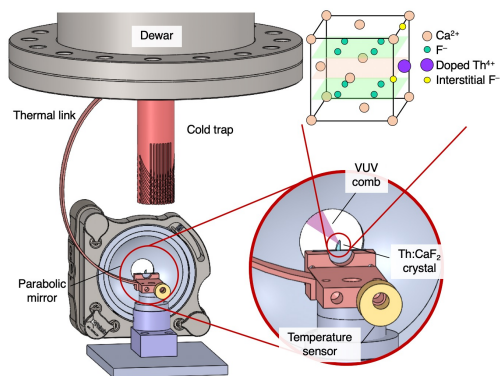


- EFG and eta parameter „well behaved“
- compatible with thermal expansion
- interesting cancellations in (B) transition (clock?)
- strong eta indicates breaking of lattice symmetry

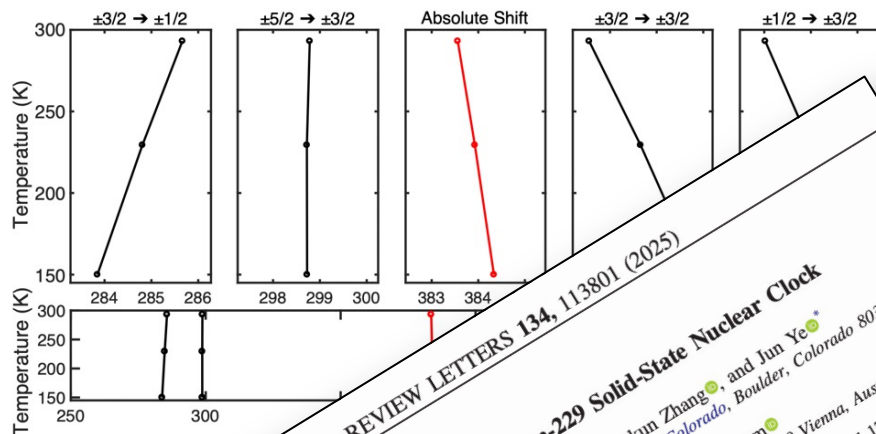
transitions now clearly depend on host material and temperature (CaF₂, LiSaF₆...)



JILA experiment



tracing 4 strongest lines over temperature



• EFG

• **PHYSICAL REVIEW LETTERS 134, 113801 (2025)**

• **Temperature Sensitivity of a Thorium-229 Solid-State Nuclear Clock**

JILA, NIST and University of Colorado, Department of Physics, University of Colorado, and Jun Ye*
 Jacob S. Higgins, Tian Ooi, Jack F. Doyle, Chuankun Zhang, and Jun Ye*
 Kjeld Beeks, Tomas Sikorsky, and Thorsten Schumm
 Vienna Center for Quantum Science and Technology, Atominstiut, TU Wien, 1020 Vienna, Austria
 (Received 17 September 2024; revised 28 October 2024; accepted 17 January 2025; published 17 March 2025)

• **Transition (clock?)**

• **of lattice symmetry**

• **depend on**

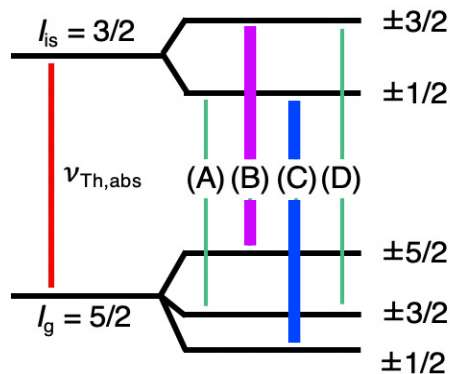
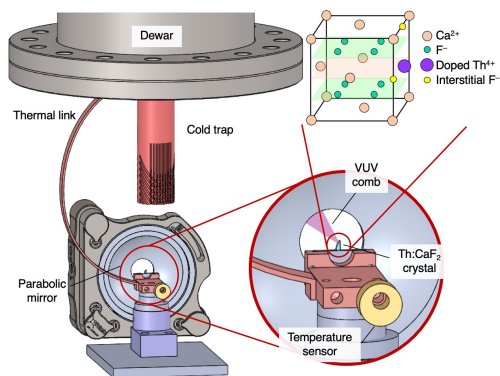
• **and temperature**



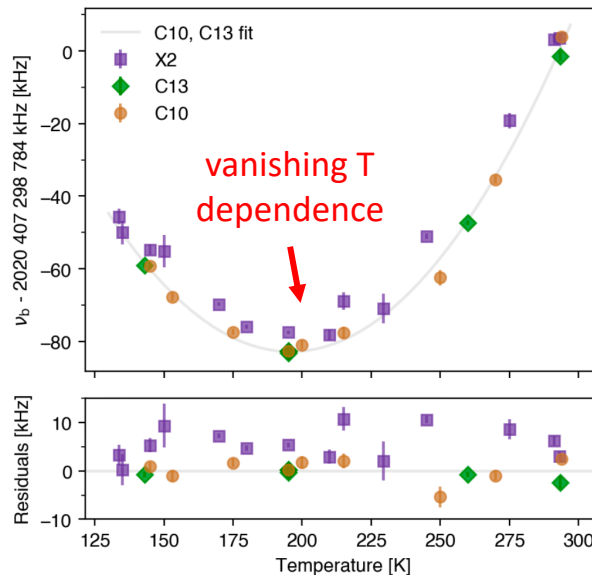
Towards a $^{229}\text{Th}:\text{CaF}_2$ solid-state nuclear clock



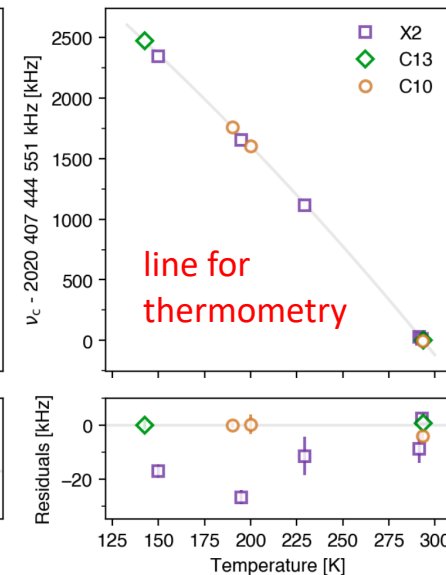
JILA experiment



(B) transition



(C) transition



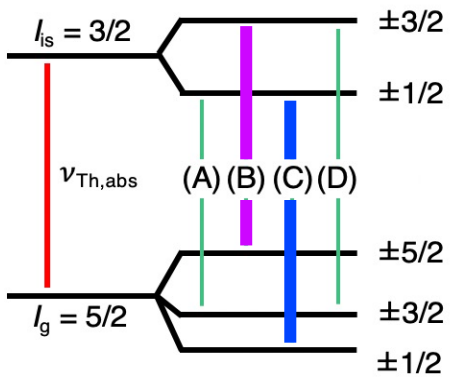
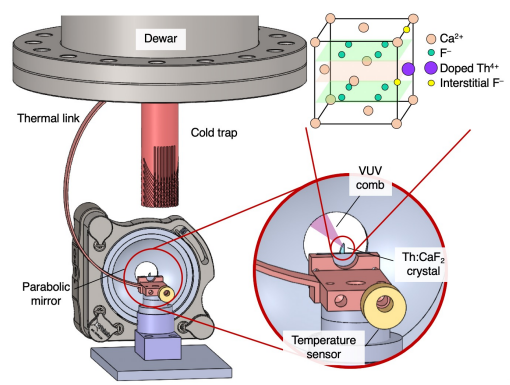
- transition frequencies consistent within ~kHz over different crystals
- linewidth 10-100 kHz, scaling with concentration („micro-strain“)
- Continuous interrogation at fixed T (over 1 year) within 1 kHz



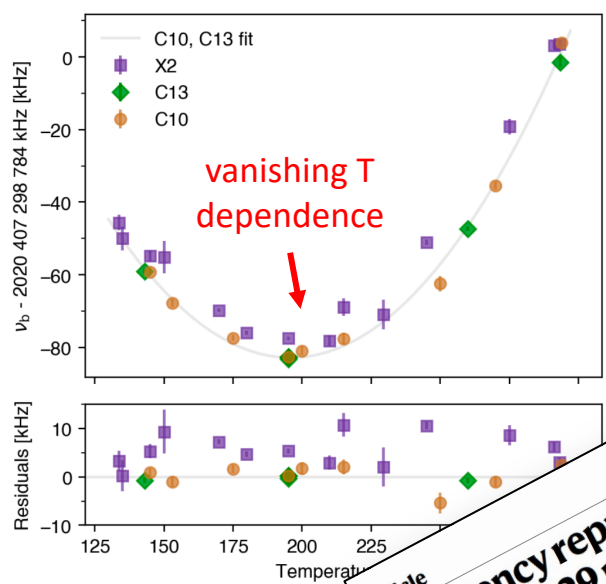
Towards a $^{229}\text{Th}:\text{CaF}_2$ solid-state nuclear clock



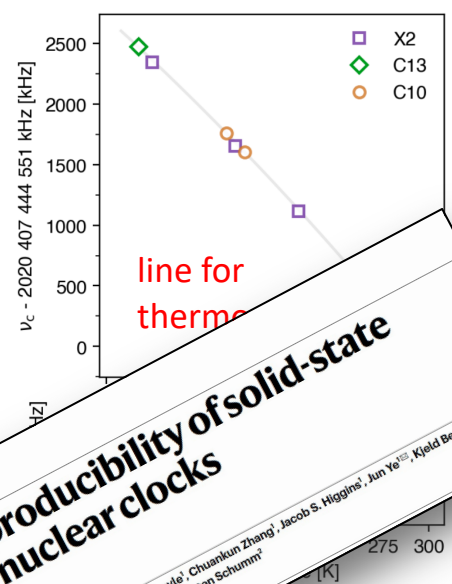
JILA experiment



(B) transition



(C) transition



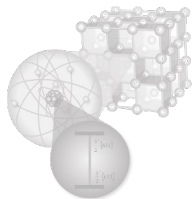
Article
Frequency reproducibility of solid-state thorium-229 nuclear clocks
Tian Ooi¹, Jack F. Doyle¹, Chuankun Zhang¹, Jacob S. Higgins¹, Jun Ye^{1,2}, Kjeld Beekes², Tomas Sikorsky² & Thorsten Schumm¹
<https://doi.org/10.1038/s41586-025-09999-5>
Received: 19 June 2025
Accepted: 3 December 2025

- transition frequencies reproducible over different crystals
- linewidth 10-100 kHz, concentration („micro-strain“)
- Continuous interrogation (over 1 year) within 1 kHz



or contact:

thorsten.schumm@tuwien.ac.at



The „solid state approach“ to a Th-229 nuclear clock

- Basic concept and implementation

Nuclear quadrupole structure

- Nuclear – solid coupling
- Separating nuclear properties and material parameters

JILA experiment

- Absolute Mössbauer spectroscopy
- Temperature dependence
- Towards a solid-state nuclear clock

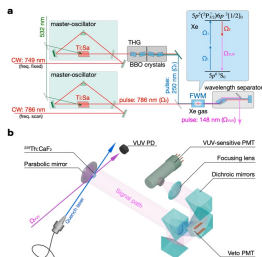
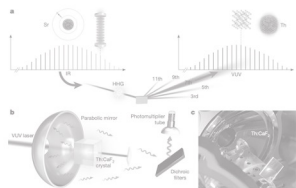
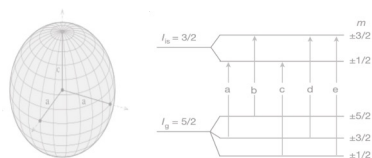
JILA

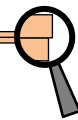
OKAYAMA experiment

- Multiple defect centers
- Site-dependent properties

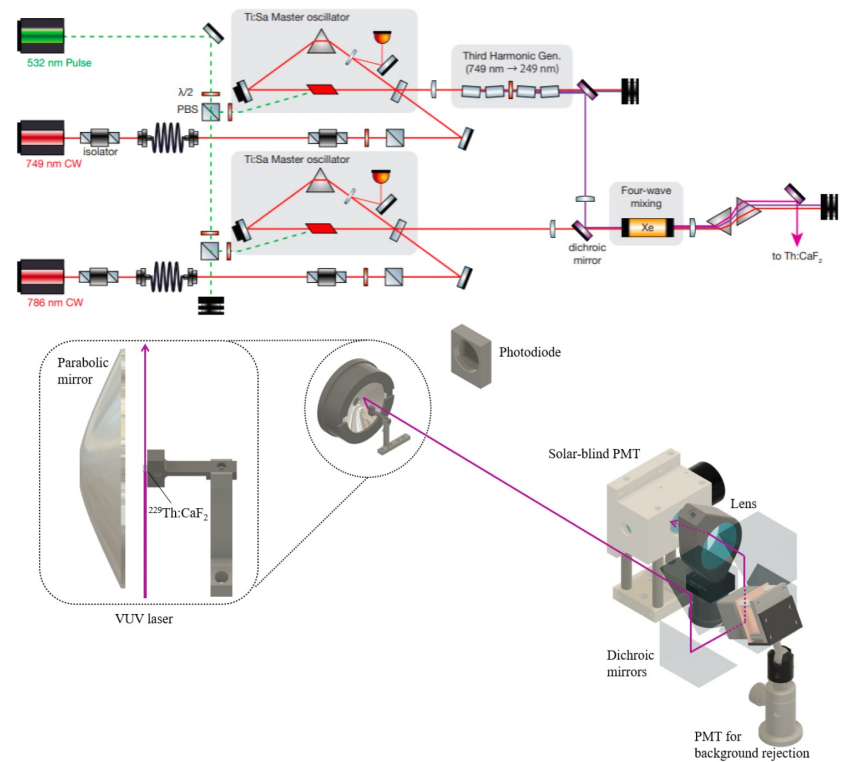


OKAYAMA UNIVERSITY





Okayama experiment



- 4-wave mixing laser similar to PTB/UCLA
- **linewidth 30 MHz** → resolve Q-structure
- factor 50 higher excitation rate than JILA

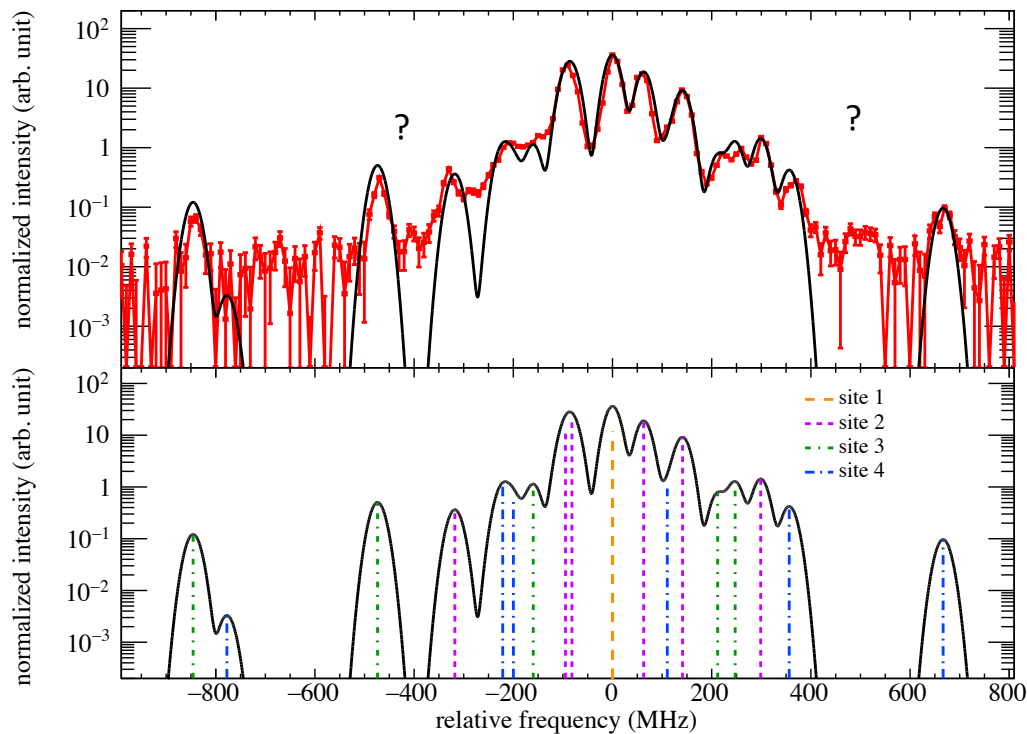
- detection system taken from SPring-8
- 3 crystals from Vienna (always the same)

Table 1: Parameters of $^{229}\text{Th}:\text{CaF}_2$ crystals.

crystal name	C10	C13	X2
concentration (/mm ³)	4×10^{14}	8×10^{14}	5×10^{15}
^{229}Th activity (kBq)	1.1	2.7	17.2

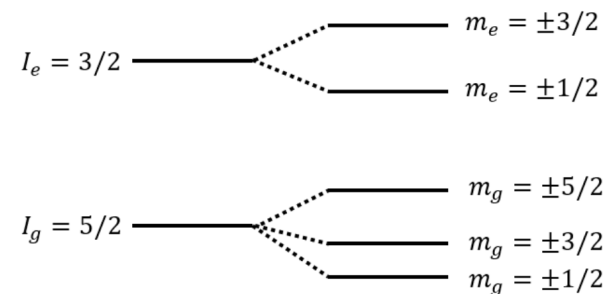


Okayama experiment

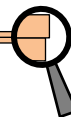


no splitting

with crystal
electric field



- at least 4 different Th sites identified in all 3 crystals
- one of them is „JILA site“ (site 2)
- missing 6th line of „JILA site“ observed
- Relative occurrences of sites changes with concentration



Okayama experiment

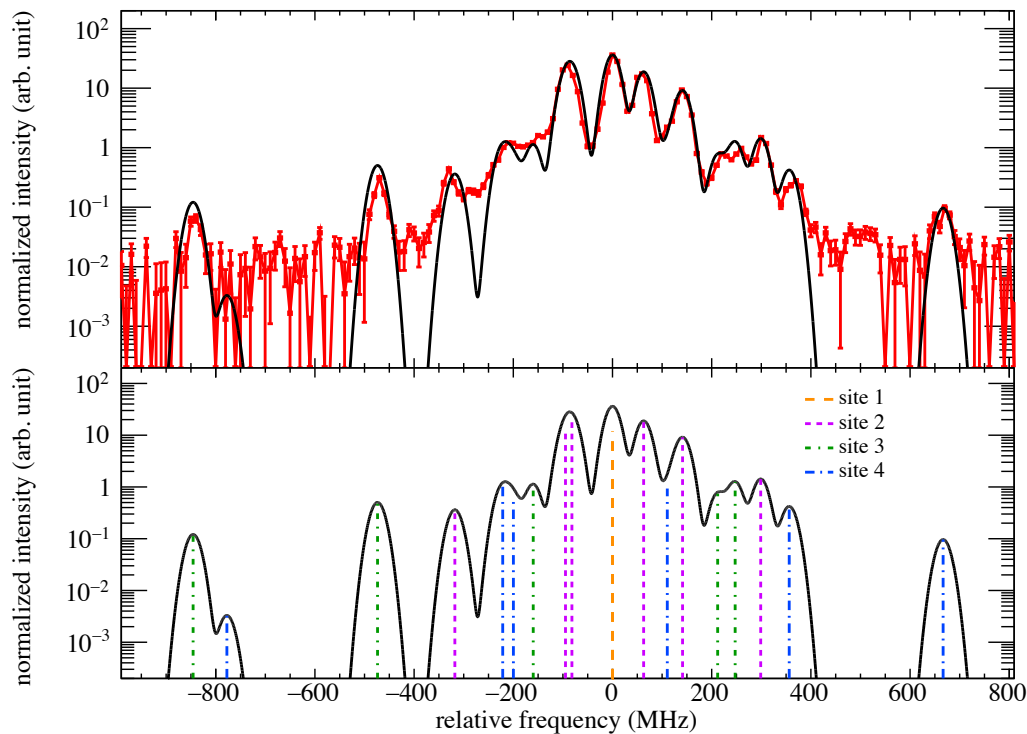
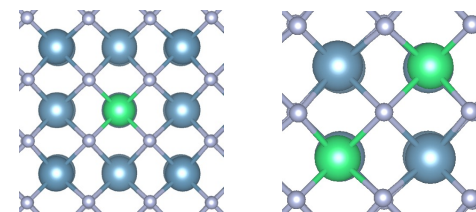


Table 1: V_{zz} ($\text{V}/\text{\AA}^2$) and η obtained by spectrum fitting. Values in parentheses are differences of fitting results of the two spectra.

target	EFG	0 EFG!	JILA site	site 3	site 4
		site 1	site 2		
C10	V_{zz}	1(1)	106(1)	-319(3)	261(1)
	η	-	0.59(1)	0.22(16)	0.00(0)
C13	V_{zz}	0(0)	107(1)	-319(3)	259(1)
	η	-	0.61(3)	0.05(9)	0.00(0)
X2	V_{zz}	2(5)	106(2)	-320(3)	258(3)
	η	-	0.60(1)	0.18(5)	0.21(6)



tentative assignment: single Th and 2Th cluster **without local charge compensation**



Okayama experiment

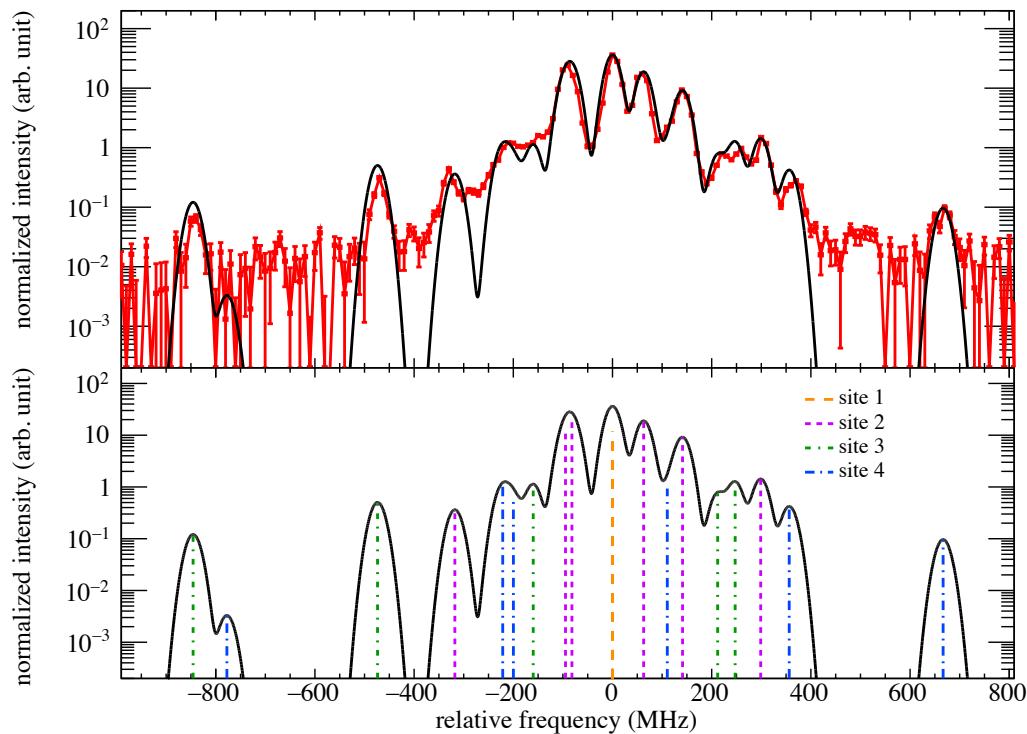


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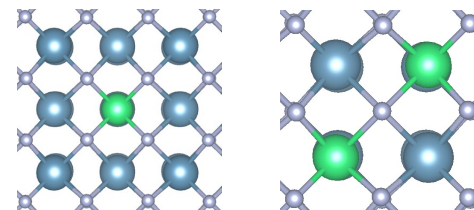
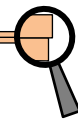


Table 2: **Relative contribution of microscopic sites to spectroscopy signal (%)**. Values in parentheses are differences of fitting results of the two spectra.

target	site 1	site 2	site 3	site 4
C10	72.6(2)	26.4(5)	0.4(3)	0.6(1)
C13	73.8(6)	24.0(7)	1.0(1)	1.2(1)
X2	34.8(3)	58.6(12)	3.6(4)	3.0(12)

>90%



Okayama experiment

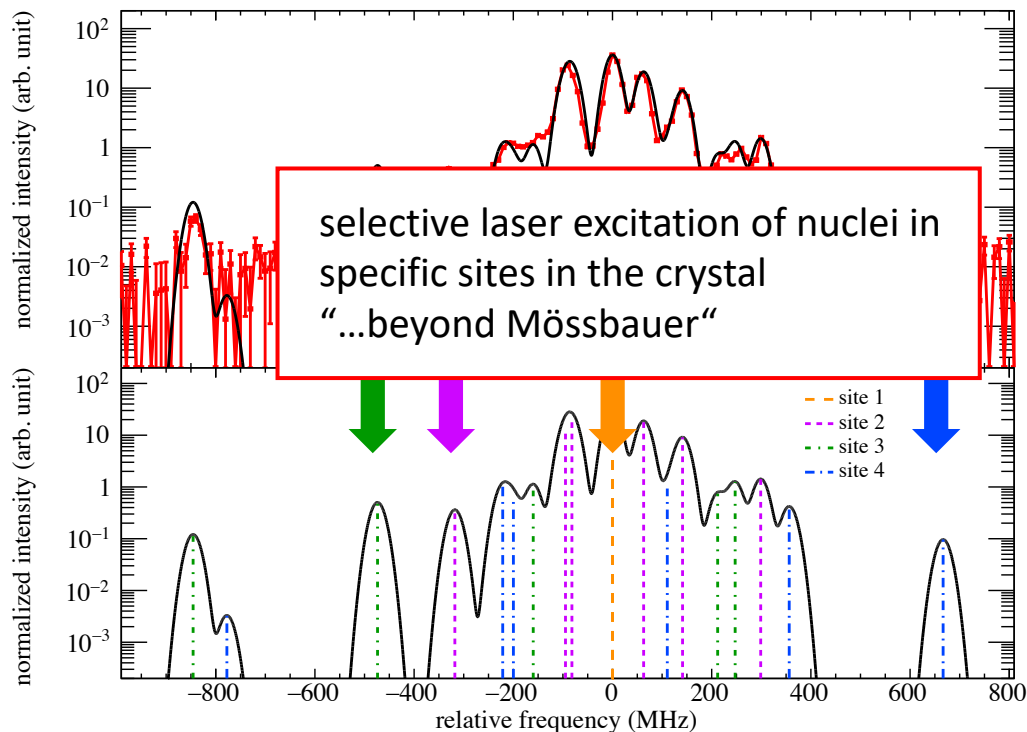


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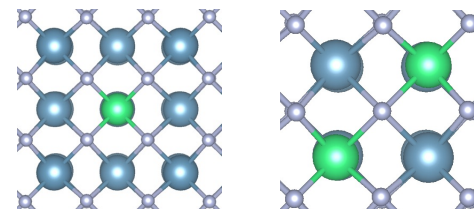
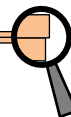


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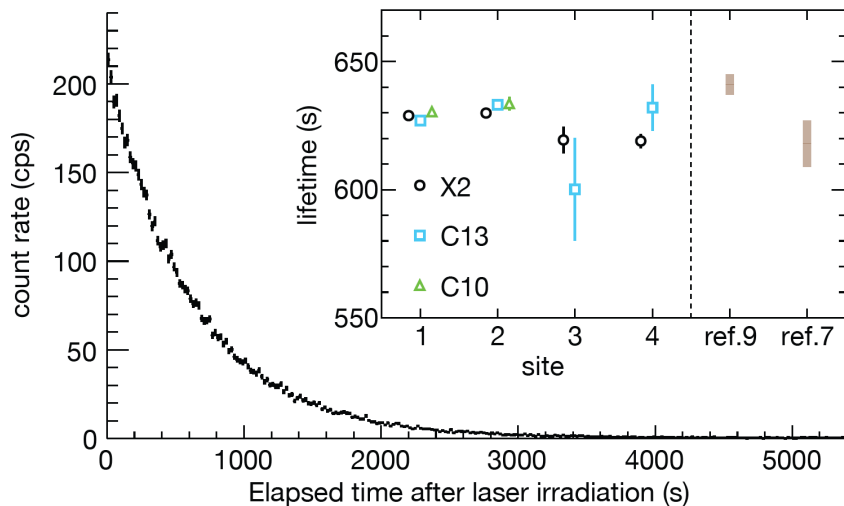
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Okayama experiment

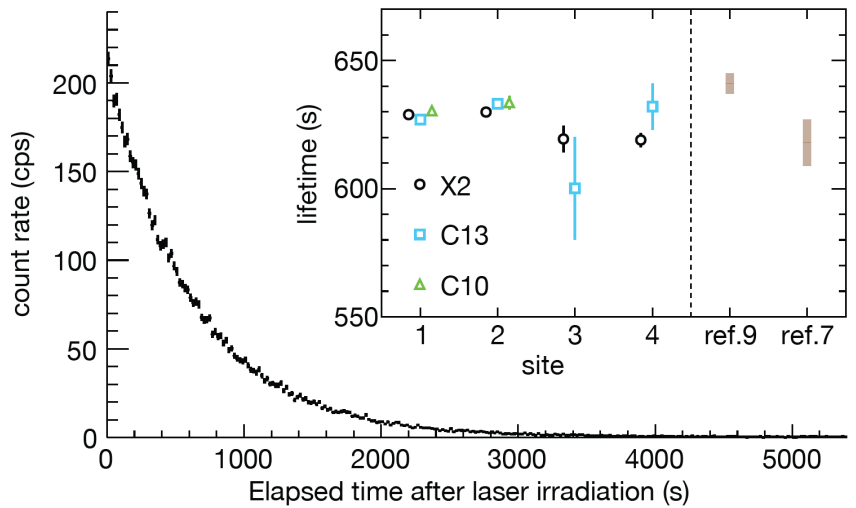
isomer lifetime **DOES NOT** seem to depend (much) on Th doping site
(all measurements at room temperature)



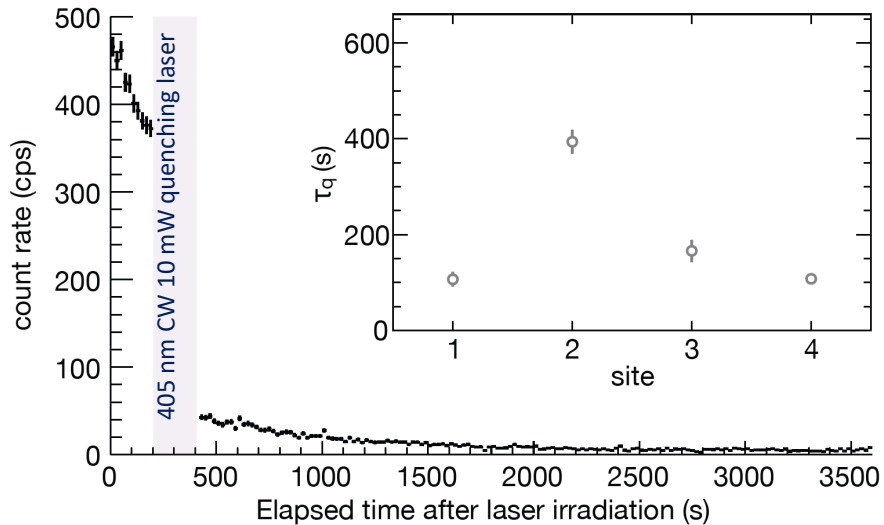


Okayama experiment

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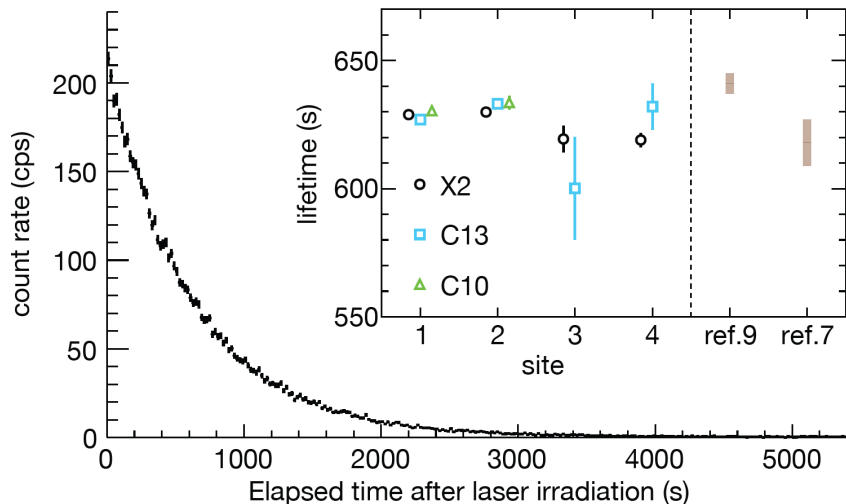
isomer laser quenching **DOES** depend on Th doping site (all measurements at room temperature)



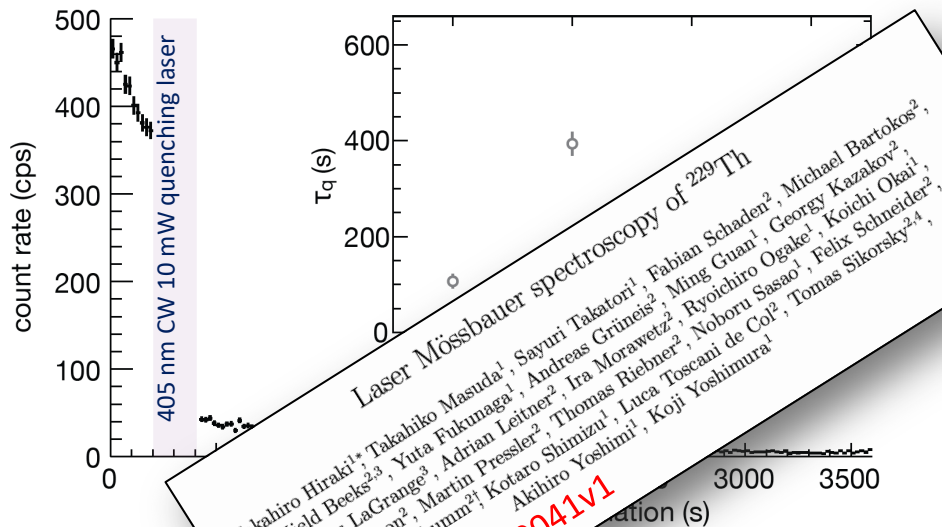


Okayama experiment

isomer lifetime **DOES NOT** seem to depend (much) on Th doping site (all measurements at room temperature)



isomer laser quench **DOES** depend on Th doping site (all measurements at room temperature)



Laser Mössbauer spectroscopy of ^{229}Th

Takahiro Hiraki^{1*}, Takahiko Masuda¹, Sayuri Takatori¹, Fabian Schaden², Michael Bartkos², Kjeld Beeks^{2,3}, Yuta Fukunaga¹, Andreas Grüneis², Ming Guan¹, Georgy Kazakov², Thomas LaGrange³, Adrian Pressler², Ira Morawetz², Ryoichiro Ogake¹, Koichi Okai¹, Martin Pimon², Martin Pressler², Thomas Riebner², Noboru Sasao¹, Felix Schneider², Thorsten Schumm^{2†}, Kotaro Shimizu¹, Luca Toscani de Col², Tomas Sikorsky^{2,4}, Akihiro Yoshimi¹, Koji Yoshimura¹

[arXiv:2509.00041v1](https://arxiv.org/abs/2509.00041v1)



or contact:

thorsten.schumm@tuwien.ac.at



ISOLDE-CERN measurements

S. Kraemer, J. Moens, S. Bara, P. Chhetri, A. Claessens, T. Cocolios,
S. Geldhof, L. Pareire, S. Sels, A. Vantomme, M. Verlinde, **P.v. Duppen**
M. Athanasakis-Kaklamanakis, K. Chrysalidis, R. Henke
K. Beeks, N. Hosseini, T. S.

KU Leuven

CERN
TU Wien

PTB measurements

J. Tidau, J. Thielking, G. Zitzer K. Zhang, M. Okhapkin, **E. Peik**
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SPRING-8/Okayama measurements

T. Masuda, A. Yoshimi, A. Fujieda, H. Hara, T. Hiraki, H. Kaino, Y. Miyamoto,
K. Okai, S. Okubo, N. Sasao, S. Uetake, M. Yoshimura, **K. Yoshimura**, K. Suzuki
H. Fujimoto, T. Watanabe
H. Haba, A. Yamaguchi, T. Yokokita, K. Tamasaku
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JILA measurements

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THE END



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