Overview of ^{229m}Th based nuclear clock

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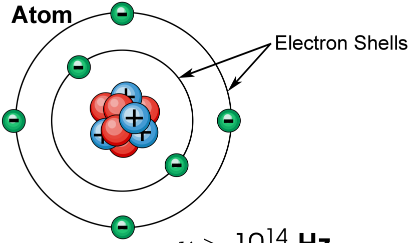
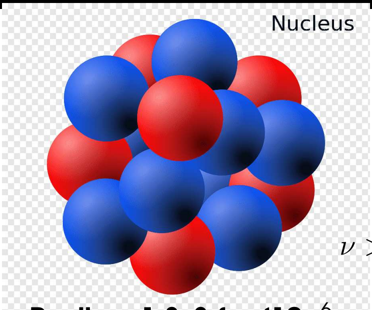
[1]INFN Genova

The Low Energy Frontier of Particle Physics,
Frascati, 12 February 2025

Atomic and nuclear scales



$\nu > 10^{14}$ **Hz**



$\nu > 10^{17}$ **Hz**

Radius: 0.5-2 Å (0.1 nm)
Excitations: eV - keV

- *highly monochromatic coherent* lasers (eV),
- visible-microwave comb ($\frac{\Delta\nu}{\nu} \sim 10^{-18}$),
- optical ion clocks: Yb^+, Al^+, Ca^+, Sr^+, Hg^+ etc,
- 1 s≡9.192.631.770 RF oscillations in Cs clock.

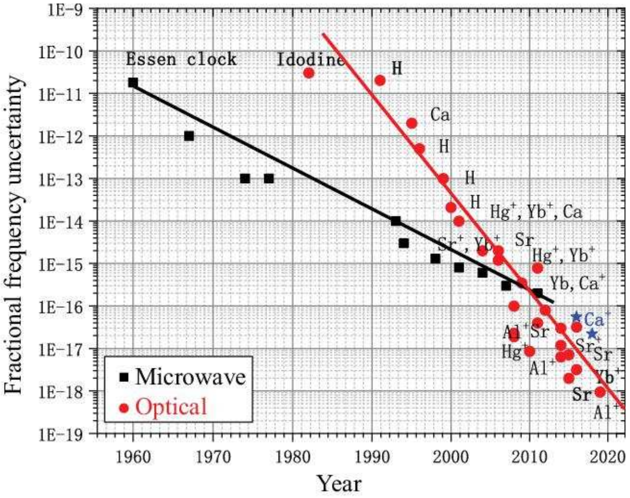
Radius: 1.3-8 fm (10^{-6} nm)
Excitations: keV - MeV

- incoherent γ-sources,
- broad linewidth FELs ($\frac{\Delta\nu}{\nu} \sim 10^{-3} \div 10^{-5}$),
- Mössbauer spectroscopy ($\frac{\Delta\nu}{\nu} > 10^{-22}$),
- no link to RF electronics.

Atomic clock evolution

1. Cs atomic fountain - J. Zacharias 1956,
2. GPS application -1978/1993,
3. optical ion clock - H.G. Dehmelt and W. Paul Nobel Prize 1989,
4. laser cooling - S. Chu, C. Cohen and W.D. Phillips Nobel Prize 1997,
5. visible-microwave comb - Th. Hänsch and J. Hall Nobel Prize 2005.

[1] K. Gao ,Nat.Sci.Rev.7 (2020) 1799



Atomic clock precision

1. **Black Body Radiation (BBR)**,
2. servo frequency steering,
3. 2d order Doppler shift,
4. electric quadruple shift,
5. Zeeman shift,
6. Stark shift.

[1] *K. Gao ,Nat.Sci.Rev.7 (2020) 1799*

The Ca^+-clock systematic-uncertainty budget table (unit in 10^{-18}).

Contribution	Fractional frequency shift	Fractional frequency uncertainty
BBR field evaluation (temperature)	863	19
BBR coefficient ($\Delta\alpha_0$)	0	0.3
Excess micromotion	0	0.4
Second-order Doppler (thermal)	−5.0	2.5
ac Stark shift	1.2	1.3
Residual quadrupole	0	2.3
Zeeman effect	0	1.5
Servo	0.0	3.0
Total	859	20

Nuclear clock precision

First idea from: E.V. Tkalya et al., Phys. Scr. 53, 296 (1996)., E. Peik and C. Tamm, Eur.Phys.Lett.61, 181 (2003),

- the same technique as in optical atomic clocks,
- small coupling of external fields to nucleus,
- atomic shell screening,
- **small black-body radiation uncertainty.**

Type of shift	Shift ($\times 10^{-20}$)	Uncertainty ($\times 10^{-20}$)
Excess micromotion	10	10
Gravitational	0	10
Cooling laser stark	0	5
Electric quadrupole	3	3
Secular motion	5	1
Linear Doppler	0	1
Linear Zeeman	0	1
Collisions	0	1
Blackbody radiation	0.013	0.013
Clock laser stark	0	$\ll 0.01$
Trapping field stark	0	$\ll 0.01$
Quadratic Zeeman	0	0
Total	18	15

[1] *C.J. Campbell et al.,Phys.Rev.Lett.108 (2012) 120802*

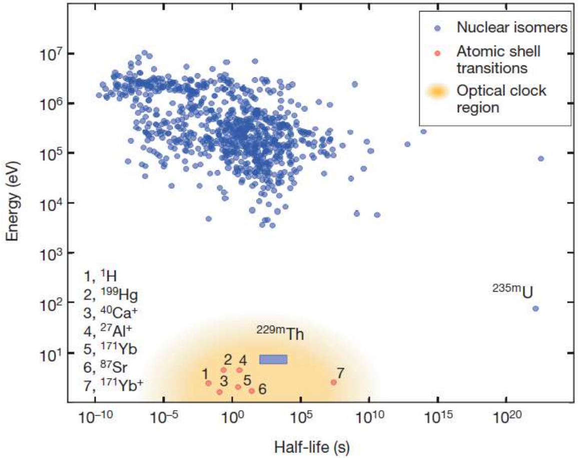
Known nuclear isomers

Need an isomer with long lifetime and large energy:

$$\frac{\Delta\nu}{\nu} = \frac{\Gamma_m}{E_m} = \ln(2)\frac{\hbar}{E_m t_{1/2}} = \frac{4.6\times 10^{-16}}{\frac{E_m}{eV}\frac{t_{1/2}}{s}} < 10^{-19}$$

1. ^{229m}Th at 8 eV with half life 7 μs (IC) or 1 hour (γ),
2. ^{235m}U at 76.8 eV with half life 25 min. (IC) or 10^{14} years (γ).

All other known nuclear isomers have energy $>$1 keV.



[1] *L. von der Wense et al.,Nature 533 (2016) 47*

^{229m}Th isomer - optical range nuclear level

- 43 years from the first indication based on level scheme to the final confirmation,
- resurgence of interest due to **nuclear clock**
- several multi M€ projects in EU:
  nuClock FET (2015-2019) 4 MEuro;
  ThoriumNuclearClock ERC (2020-2026) 14 MEuro.
- γ-decay (dominant IC) recently observed,
- photo-induced excitation of ^{229m}Th recently observed.

Year	Energy (eV)	Ref.
1976	<100	L.A. Kroger, C.W. Reich, Nucl.Phys.**A259**, 29
1990	1±4	C.W. Reich, R. Helmer, Phys.Rev.Lett.**64**, 271
1994	3.5±1	R. Helmer, C.W. Reich, Phys.Rev.**C49**, 1845
2007	7.6±0.5	B.R. Beck *et al.*, Phys.Rev.Lett.**109**, 142501
2019	8.3±0.2	B. Seiferle *et al.*, Nature **573**, 243
2023	8.34±0.02	S. Kraemer *et al.*, Nature **617**, 706

Non-spherical ^{229}Th rotational splitting

1. ^{229}Th deformed nucleus (axial),
2. Nilsson model[1] level scheme[2]:

$$E(\Omega^{\pi}[N, n_z, \Lambda]) \ ,$$

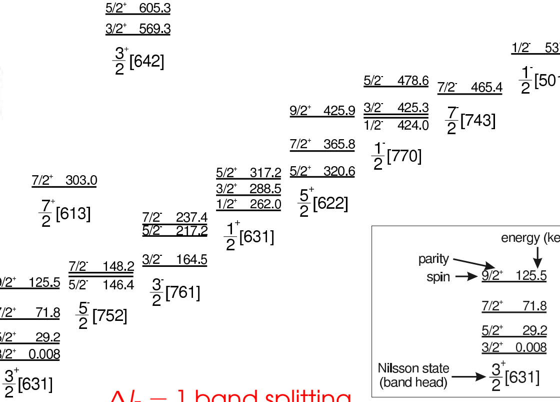
$$\Omega = j_z, \ [2j+1]$$
$$N = n_z + n_{\perp}$$
$$\Lambda = l_z, \ [2n_{\perp}+1]$$

3. deformation parameter:

$$\delta = 1 - \left(\frac{L_{\perp}}{L_z}\right)^2$$

4. ^{229}Th prolate[3,4,5]

$$\delta \sim 0.1, L_z \sim 1.05 L_{\perp}$$
$$M1\left(\tfrac{3}{2}^{+} \rightarrow \tfrac{5}{2}^{+}\right)$$



$\Delta l_z = 1$ band splitting

[1] S.Nilsson,Matematisk-fysiske Med-delelser,29(16)(1955
[2] G.Musiol et al.,Kern und El.physik.,Weinheim(1988
[3] E.Ruchowska et al.,Phys.Rev.C73,044326(2006).
[4] A.Hayes et al.,Phys.Rev.C78,024311(2008).
[5] E.Litvinova et al.,Phys.Rev.C79,064303(2009).

^{229m}Th isomer decays

1. magnetic dipole transition M1: $\frac{3}{2}^+ \to \frac{5}{2}^+$, with decay γ angular distribution (1+$\cos\theta_\gamma$),
2. calculated lifetime[1,2,3] (γ only):

$$\tau_\gamma \sim \frac{10.95h}{E_\gamma^3 B(M1)} \sim 1h, \quad B(M1) \sim 0.025\mu_N^2$$

3. ionization energy of 6.5 eV shortens the lifetime (IC) to 7 μs in neutral atom[4], ($\Delta\nu/\nu \sim 10^{-11}$ not suitable for nuclear clock, requiring $\Delta\nu/\nu < 10^{-16}$),
4. ionized or bound Th$^+$ ion suppresses IC-decay branch,
5. in MgF_2 (n=1.488) crystal measured lifetime of 967$\pm$147 s[5] in agreement with calculations including $1/n^3$-scaling[6]: 967 s$\times n^3\sim$1 h.

[1] *R.Helmer and C.Reich. Phys.Rev.C49,1845(1994).*
[2] *V.Strizhov and E.Tkalya.Sov.Phys.JETP,72,387(1991).*
[3] *E.Ruchowska et al.,Phys.Rev.C73,044326(2006).*
[4] *B.Seiferle et.al.Phys.Rev.Lett.118,042501(2017).*
[5] *S.Kraemer et al.Nature 617, 706 (2023).*
[6] *E.V. Tkalya et al., Phys.Rev.C61, 064308(2000).*

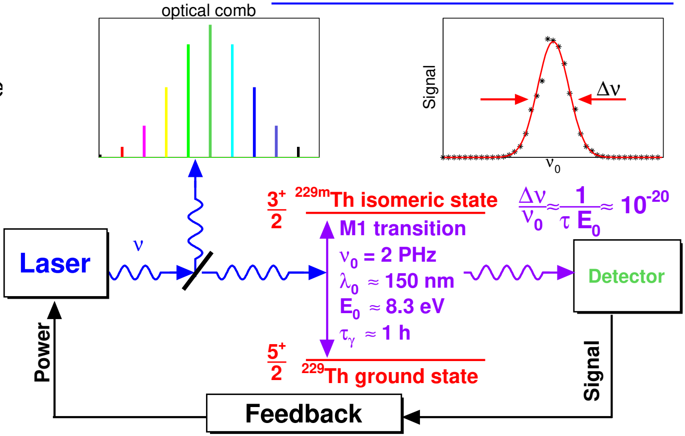
^{229}Th-based Nuclear Clock

1. Lowest nuclear exited state[1],
2. dominant IC decay[2] can be suppressed,
3. VUV-laser excitation,
4. γ-linewidth $\Delta\nu \sim 0.1$ mHz[3],
5. N=$10^5 \div 10^{12}$ oscillators[4,5],

$$FoM = \frac{\nu\sqrt{N}}{\Delta\nu},$$

6. $>10^1 \div 10^2$ FoM improvement wrt atomic clock.

N. Poli et al,Nuov.Cim.36 (2013) 555

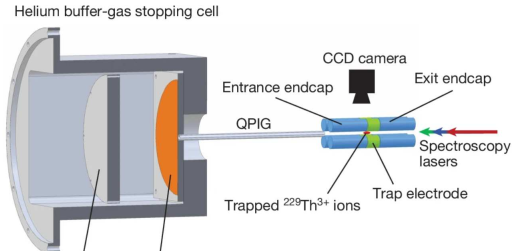


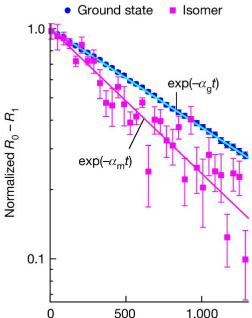
[1] L.Kroger and C.Reich, Nucl.Phys.A259, 29(1976).
[2] B.Seiferle et al., Nature 573, 243 (2019).
[3] V.Strizhov and E.Tkalya, Sov.Phys.JETP 72, 387 (1991).
[4] C.Campbell et al., Phys.Rev.Lett.102, 233004 (2009).
[5] R.Jackson et al.,J.Phys.Cond.Mat.21, 325403 (2009).

$^{229}Th^{3+}$ γ-decay

1. 2000 $^{229}Th^{3+}$ ions/s in linear Paul trap (40%,15 min.),
2. laser spectroscopy around $6d^2D_{5/2} \rightarrow 5f^2F_{7/2}$ (984 nm),
3. hyperfine constants of magnetic dipole and electric quadrupole of the $6d^2D_{5/2}$ state were determined,
4. magnetic dipole and electric quadrupole moments of ^{229m}Th were determined to be -0.378(8)μ_N and 8.84(10) *eb*, respectively,
5. $^{229m}Th^{3+}$ half-life τ_γ=1400^{+600}_{-300} s was measured.

A.Yamaguchi et al., Nature 629 (2024) 62,



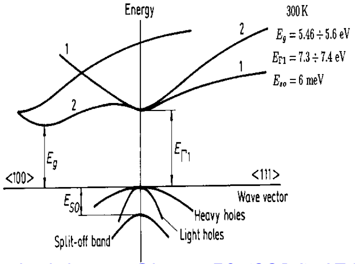


Chemical bonding IC suppression

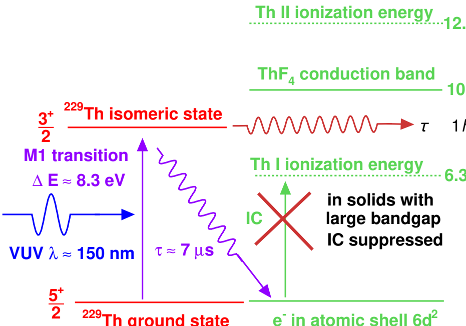
1. IC decay requires:
   $E_{decay} > E_{gap}$,
2. find Th-compound with
   $E_{gap} > 8.3$ eV,
3. theoretical calculations[1]
   suggested Na_2ThF_6 and ThF_4
   would have $E_{gap} > 8.9$ eV,
   while ThX_4 (X=Cl, Br, I)
   $E_{gap} < 6.5$ eV.

Comp.	Bandgap (eV)
CaF_2[†]	12
UO_2	2.2
UC,UN,UCl,UF	2-3
Other	?

[†]used in thoriumclock.eu project

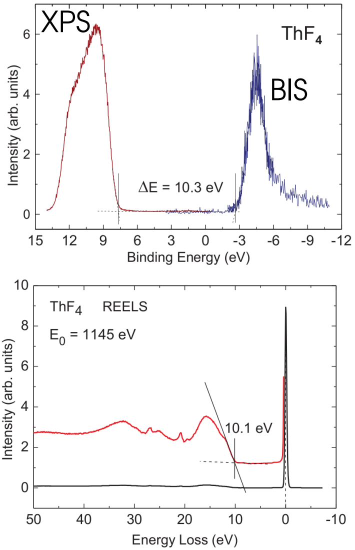


[1] J.K.Ellis et al.,Inorg.Chem.53 (2014) 6769



ThF_4 bandgap measurements (JRC, Karlsruhe)

1. 25 nm film of ThF_4 grown on Au substrate,
2. XPS measured valence band,
3. BIS measured conduction band,
4. REELS confirmed XPS-BIS results,
5. $E_g = 10.2 \pm 0.2$ eV,
6. > 8.3 eV, suitable for nuclear clock matrix.

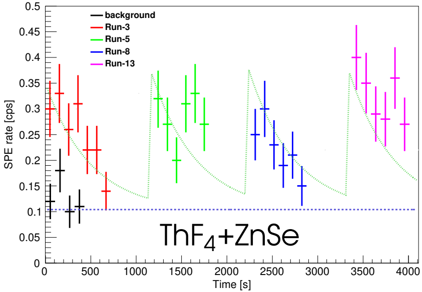


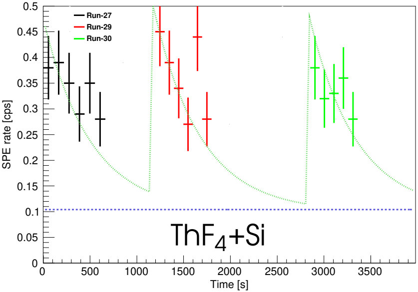
T.Gouder et al.,Phys.Rev.Research 1 (2019) 033005

UV photo-luminescence of ThF_4

- 0.2 Hz UV photo-lumi. observed on top of 0.1 Hz flat background (opposite side lumi. =bkg.)✓,
- UV photo-lumi. decay time of about 400 s ✗,
- light yield from ThF_4+Si sample was larger by factor 3/2 in agreement with ThF_4 layer thickness ratio ✓,
- does not depend on irradiation time (Run.30 after 15 min. irradiation should have 1.85 times higher lumi.) ✓,
- previous item demonstrates that the excitation process is different from the decay.

M.Osipenko et al.,NIM A 1068 (2024) 169744



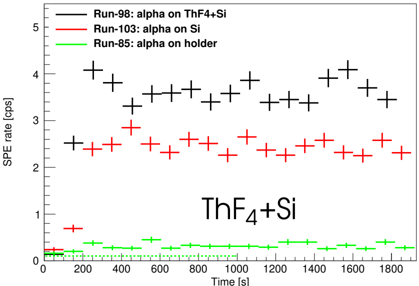


UV radio-luminescence of ThF_4

- 1.2 Hz UV radio-lumi. observed (opposite side lumi. subtracted) on top of 0.1 Hz background ✓,
- light yield from ThF_4+ZnSe sample =0.8 Hz was smaller by factor 3/2 in agreement with ThF_4 layer thickness ✓,
- 1.4 kBq ^{241}Am source placed 2 mm above the sample, provides 0.4 kHz of αs on ThF_4 film,
- mean α energy loss in 300 nm of ThF_4 film was 126 keV and PMT acceptance of about 0.1 gives: $1.7\times10^{-3}/EQ(\lambda)$ UV-photons(120-200 nm)/keVee ✓.

α-source above sample

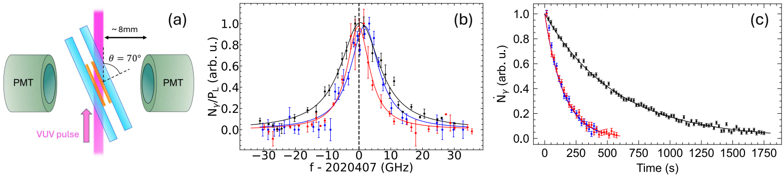
M.Osipenko et al.,NIM A 1068 (2024) 169744



^{229m}Th excitation in $^{229}ThF_4$ thin films

- 30 nm thin film of (21 kBq) $^{229}ThF_4$ was grown on MgF_2 and Al_2O_3 ✓,
- ^{229m}Th excitation was observed at $2020406.8(4)_{stat}(30)_{sys}$ GHz ✓,
- γ-decay of photo-excited ^{229m}Th in ThF_4 was observed ✓,
- ^{229m}Th lifetime in ThF_4 of $153(9)_{stat}(7)_{sys}$ s was measured ✓.

C.Zhang et al., Nature 636 (2024) 603



^{229m}Th lifetime medium dependence

- predicted ^{229m}Th lifetime dependence on the medium refractive index n^*:

$$\tau_{med} = \frac{\tau_{vac}}{n^3} ,$$

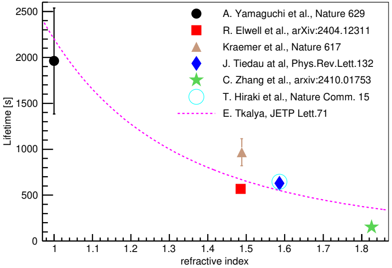
- measured lifetimes of ^{229m}Th are decreasing with n ✓,
- but lifetime in ThF_4 is 2.4 times smaller than expected,
- could be due to small thickness of the ThF_4 film,
- this may allow to manipulate ^{229m}Th lifetime in thin films.

M.Osipenko et al.,NIM A 1068 (2024) 169744

$$t_{ThF} \simeq 30\ nm << \lambda_0 \simeq 150\ nm ,$$

$$\tau_{ThF4} \simeq 153\ s$$

$$\tau_{ThF4-expected} \simeq 361\ s$$



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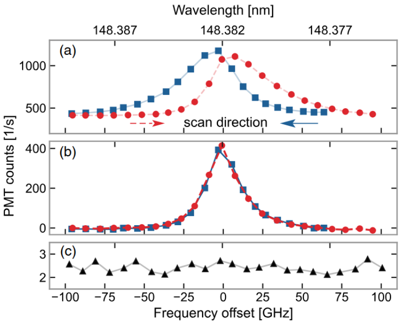
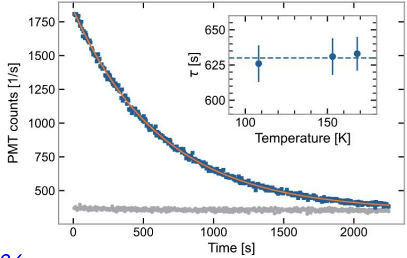
* E. Tkalya, JETP Lett. 71 (2000) 311

^{229m}Th excitation in CaF_2 bulk

- $\geq$1 mm^3 CaF_2 crystals with $10^{-5} \div 10^{-3}$ (w.r.t. Ca) concentration ^{229}Th doping were grown ✓,
- ^{229m}Th excitation was observed at 148 nm ✓,
- γ-decay of hadro[1]- and photo-excited ^{229m}Th was observed ✓,
- ^{229m}Th lifetime in ThF_4 of 630(15) s was measured ✓.



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



T. Hiraki et al., Nature Comm. 15 (2024) 5536

[1] S. Kraemer et al., Nature 617 (2023) 706

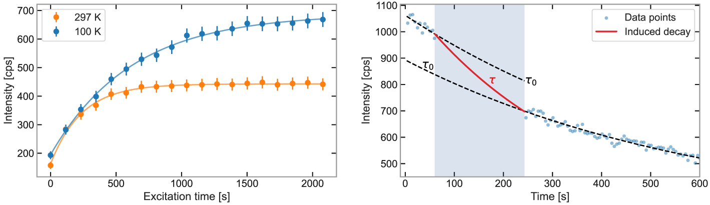
C. Zhang et al., Nature 633 (2024) 63

Quenching of ^{229m}Th decay in CaF_2

^{229m}Th lifetime quenching was observed varying crystal temperature and irradiating it with CW laser:

- 2.6 times quenching was observed at T=300 K w.r.t. T=100 K ✓,
- 3 times quenching was observed irradiating crystal by ≤420 nm CW laser beam of 20 mW ✓,
- quenching may allow to reduce nuclear clock cycle times,
- shorter cycle time improves frequency stability and important for new physics searches[1].

F. Schaden et al., arxiv:2412.12339 (2024)



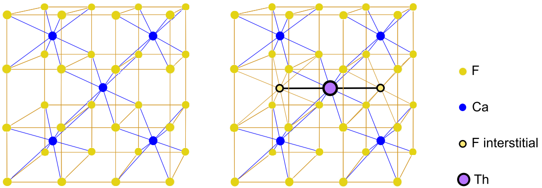
[1] M.S. Safronova et al., Rev.Mod.Phys.90 (2018) 025008

Solid State Nuclear Clock

- $^{229}Th^{4+}$ ion embedded in crystal,
- crystal bandgap $>$8.3 eV suppresses IC.

[1] G.Kazakov et al.,New J.Phys. 14 (2012) 083019

[2] W.Rellergert et al.,Phys.Rev.Lett. 104 (2010) 200802



Pros:

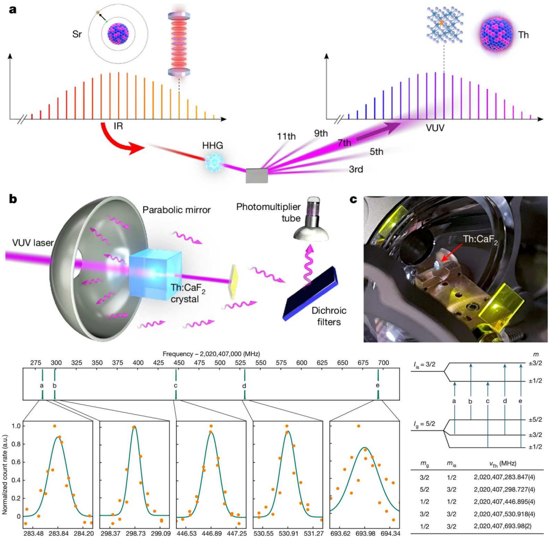
1. very high number of oscillators (10^{19} cm^{-3}, $>$10 orders of magnitude above ion traps) ✓,
2. stable, easy to handle solid state target ✓,
3. no first-order Doppler effect even at room temperature, recoil energy is far below the energy required for creating a phonon ✗.

Cons:

1. Zeeman interaction between ^{229}Th with nearby F nuclei and second-order Doppler effect ✗.

First Prototype

- ^{87}Sr atomic clock IR beam generated 7th harmonic for ^{229m}Th excitation ✓,
- ^{229m}Th quardupole splitting was observed ✓,
- 300 kHz FWHM (1.6×10^{-10}) linewidth of VUV-comb ✗,
- 2 kHz (10^{-12}) precision on frequency of ^{229m}Th achieved by a fit of five quadrupole lineshapes ✗.



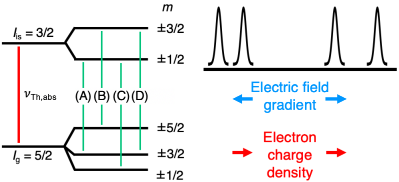
C. Zhang et al., Nature 633 (2024) 63

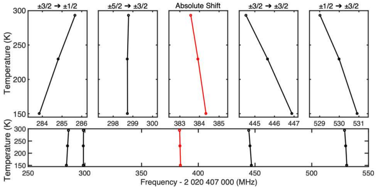
Temperature-dependent line shift

Absolute frequencies of the electric quadrupole splitting structure of ^{229m}Th$\rightarrow^{229g}$Th transition in CaF_2 crystal:

- observed in average MHz/K temperature drift ✗,
- only $m = \pm 5/2 \rightarrow \pm 3/2$ transition shows 0.4 kHz/K drift ✓,
- 5 μK temperature stability required to reach 10^{-18} precision.

J.S. Higgins et al., arxiv:2409.11590 (2024)

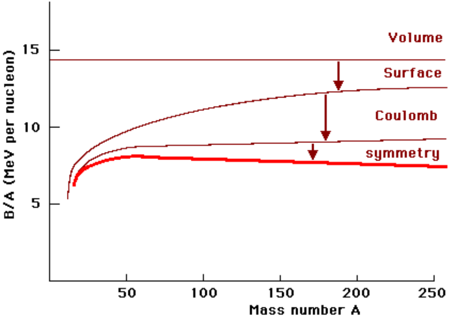




Variations of α_{EM} (with respect to α_S)

- Unification theories applied to cosmology suggest a possibility of variation of the fundamental constants in the expanding Universe , ‣ J.P. Uzan, Rev.Mod.Phys.75 (2003) 403
- searches temporal variation of α_{EM} in comparison of atomic transitions reached limit $\frac{1}{\alpha}\frac{d\alpha}{dt} < 10^{-16}/year$,
- increase sensitivity using large nuclear Coulomb energies (E_C ∼few MeV), ‣ V. Flambaum Phys.Rev.Lett.97 (2006) 092502

$$\frac{\delta\nu}{\nu} \simeq k\frac{\delta\alpha}{\alpha}, \quad with\ k = \frac{\Delta E_C}{\nu}$$



Variations of α_{EM} (with respect to α_S) cont.

- first estimates gave very optimistic results:

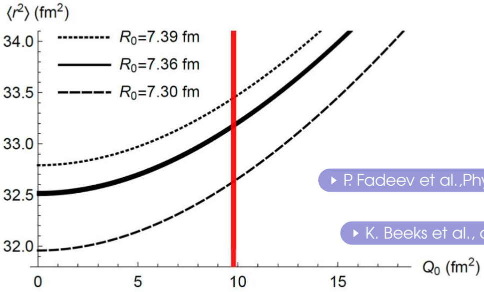
$$\frac{\delta\nu}{\nu} \simeq (k = 10^5) \times \left[4\frac{\delta\alpha}{\alpha} + \frac{\delta X_q}{X_q} - 10\frac{\delta X_s}{X_s} \right], \; X_{q,s} = \frac{m_{q,s}}{\Lambda_{QCD}}, \; k = \frac{\Delta E_C}{E_{iso}}$$

leading to sensitivity:

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

$$\frac{1}{\alpha}\frac{d\alpha}{dt} \simeq 10^{-20}/year << 10^{-17}/year(current) \; ,$$

- most recent estimate, based on ^{229}Th charge radii gave $k \simeq (5.9 \pm 2.3) \times 10^3$, leading to $\delta\nu < 100$ Hz/year.



▸ P. Fadeev et al.,Phys.Rev.A102 (2020) 052833

▸ K. Beeks et al., arXiv:2407.17300 (2024)

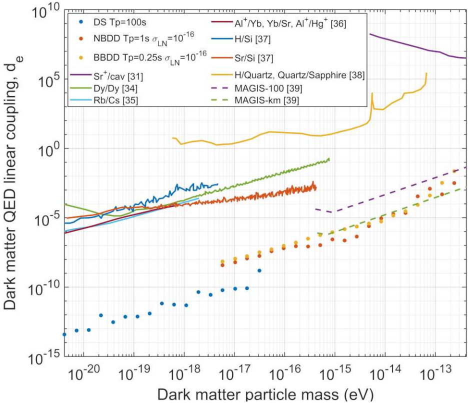
Ultra Light Dark Matter search

- coupling of ULDM to SM leads to oscillations of fundamental constants (e.g. α_{EM}), [1] arXiv:2203.14915 (2022).
- transient changes in fundamental constants may be induced by DM with large spatial extent[1],
- comparison of ^{229m}Th nuclear clock to optical lattice clock allows highest sensitivity.

Schemes:

1. Differential Spectroscopy (DS),
2. Narrowband Dynamical Decoupling (NBDD),
3. Broadband Dynamical Decoupling (BBDD).

M.H. Zaheer et al., arXiv:2302.12956 (2023)



Other ^{229m}Th applications

1. ^{229m}Th-based current-induced γ-source:
   E.V. Tkalya et al., Chin.Phys.C47 (2023) 024101
2. ^{229m}Th-based nuclear laser:
   Zeeman splitting in 100 T magnetic field;
   E.V. Tkalya, Phys.Rev.Lett.106 (2011) 162501
3. GPS (precision 3-5 m) main uncertainties:
   atmospheric disturbance, clock synchronization and stability, satellite position;
4. Chronometric geodesy: $\Delta\nu/\nu = \Delta U/c^2$
   10^{-19} clock geodesy at 1 mm, earthquake prediction, search for natural resources, tests of general relativity, clock-based gravitational wave detection.

For review see:
P.G. Thirolf et al.,Eur.Phys.J.Spec.Top.233 (2024) 1113
L. von der Wense and B. Seiferle, Eur.Phys.J. A56 (2020) 277

Summary

- ^{229m}Th is the lowest known nuclear isomer,
- its energy lies around 8.3 eV, corresponding to 150 nm wavelength, accessible with VUV-lasers ✓,
- neutral ^{229m}Th atom decays through IC in 7$\pm$1 μs ✗,
- ^{229m}Th IC decay is suppressed by ionization or chemical binding ✓,
- ^{229m}Th^{+} decays by γ-decay in about 2000 s ✓,
- ^{229m}Th^{+} γ-decay lifetime depends on the medium refractive index (1/n^{3}) and on crystal thickness,
- ^{229m}Th^{+} γ-decay linewidth is suitable for very precise nuclear clock (10^{-19} precision on a single atom in cooled trap, 10^{-17} precision on a single atom in crystal) ✓,
- first prototype of solid state nuclear clock has been tested ($\delta\nu/\nu \sim 10^{-10}$) ✓,
- more promising thin film ^{229}ThF$_4$ is under development.

Nature I

- L. von der Wense et al., "*Direct detection of the ^{229}Th nuclear clock transition*", Nature 533, 47 (2016),
- J. Thielking et al., "*Laser spectroscopic characterization of the nuclear-clock isomer ^{229m}Th*", Nature 556, 321 (2018),
- B. Seiferle et al., "*Energy of the ^{229}Th nuclear clock transition*", Nature 573, 243 (2019),
- T. Masuda et al., "*X-ray pumping of the ^{229}Th nuclear clock isomer*", Nature 573, 238 (2019),
- Kjeld Beeks et al., "*The thorium-229 low-energy isomer and the nuclear clock*", Nature Reviews Physics 3, 238 (2021),

Nature II

- S. Kraemer et al., "*Observation of the radiative decay of the ^{229}Th nuclear clock isomer*", Nature 617, 706 (2023),
- Adriana Pálffy, "*Photon lights a path towards a nuclear clock*", Nature 617, 706 (2023),
- Atsushi Yamaguchi et al., "*Laser spectroscopy of triply charged ^{229}Th isomer for a nuclear clock*", Nature 629, 62 (2024),
- Takahiro Hiraki et al., "*Controlling ^{229}Th isomeric state population in a VUV transparent crystal*", Nature Communications 15, 5536 (2024),
- C. Zhang et al., "*Frequency ratio of the ^{229m}Th nuclear isomeric transition and the ^{87}Sr atomic clock*", Nature 633, 63 (2024),

Nature III

- C. Zhang et al., "*$^{229}ThF_4$ thin films for solid-state nuclear clocks*", Nature 636, 603 (2024).

Phys. Rev. Lett. I

- B.R. Beck et al., "*Energy splitting of the ground-state doublet in the nucleus ^{229}Th*", Phys. Rev. Lett. 98, 142501 (2007).
- W. G. Rellergert et al., "*Constraining the evolution of the fundamental constants with a solid-state optical frequency reference based on the ^{229}Th nucleus*", Phys. Rev. Lett. 104, 200802 (2010).
- W.T. Liao et al., "*Coherence-enhanced optical determination of the ^{229}Th isomeric transition*", Phys. Rev. Lett. 109, 262502 (2012).
- X. Zhao et al., "Observation of the Deexcitation of the ^{229m}Th Nuclear Isomer", Phys. Rev. Lett. 109, 160801 (2012).

Phys. Rev. Lett. II

- E. Peik and K. Zimmermann, "*Comment on Observation of the Deexcitation of the ^{229m}Th Nuclear Isomer*", Phys. Rev. Lett. 111, 018901 (2013).
- J. Jeet et al., "*Results of a Direct Search Using Synchrotron Radiation for the Low-Energy ^{229}Th Nuclear Isomeric Transition*", Phys. Rev. Lett. 114, 253001 (2015).
- A. Yamaguchi et al., "*Energy of the ^{229}Th nuclear clock isomer determined by absolute γ-ray energy difference*", Phys. Rev. Lett. 123, 222501 (2019).
- T. Sikorsky et al., "*Measurement of the ^{229}Th isomer energy with a magnetic microcalorimeter*", Phys. Rev. Lett. 125, 142503 (2020).
- J. Tiedau et al., "*Laser excitation of the Th-229 nucleus*", Phys. Rev. Lett. 132, 182501 (2024).

Phys. Rev. Lett. III

R. Elwell et al., "*Laser excitation of the ^{229}Th nuclear isomeric transition in a solid-state host*", Phys. Rev. Lett. 133, 013201 (2024).

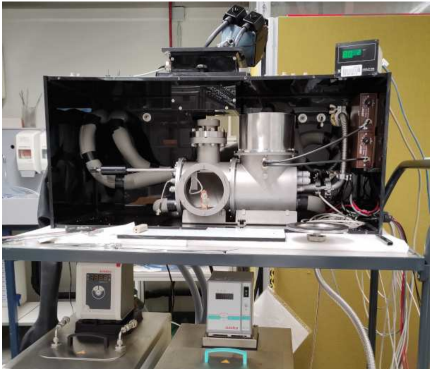
TORIO_229/CSN3 I

- T. Gouder et al., "Measurements of the band gap of ThF_4 by electron spectroscopy techniques", Phys. Rev. Research 1, 033005 (2019),
- M. Fedkevych et al., "Direct Search for Low Energy Nuclear Isomeric Transition of Th-229m With TES Detector", IEEE T. Appl. Supercon., Vol. 31, No. 5, 2100904 (2021),
- M. Fedkevych et al., "An Examination of Thermal Coupling of an Ir/Au TES for TORIO-229 Experiment", J. Low Temp. Phys. 209, 473 (2022),
- M. Osipenko et al., "Measurement of photo- and radio-luminescence of thin ThF_4 films", Nucl. Instrum. Methods A 1068, 169744 (2024).

Backup slides

GRaDeTh229 setup at JRC (Karlsruhe)

1. Hamamatsu VUV-lamp L11798 (120-170 nm),
2. solar-blind PMT R6835 (115-200 nm) + visible veto PMT R1450 (300-650 nm), CAEN DT5533 power supply,
3. charge sensitive amplifiers Ortec 113 and digitizer CAEN DT5730 with PHA firmware,
4. vacuum chamber for 10^{-4} mbar,
5. chiller capable to cool to -40 C^{o},
6. 2 commercial (amorphous) ThF_4 samples from II-VI Inc.

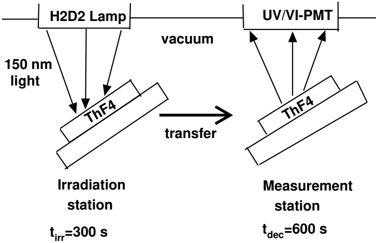


Method of photo-luminescence measurements

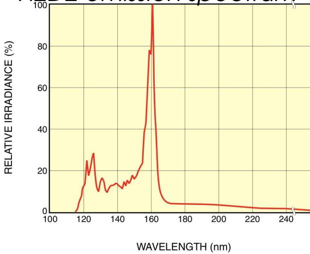
1. H2D2 VUV-lamp yields: $\sim 10\ \mu W/nm$ at 150 nm,
2. it will excite defects/impurities in ThF_4 crystal layer;
3. after transfer we observe:

$$Sig = N_{dec} \times Acc \times Eff$$

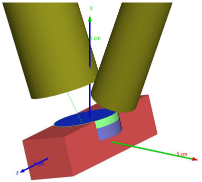
4. acceptance $Acc \sim 0.03$, efficiency $Eff \sim 0.1 \div 0.2$.

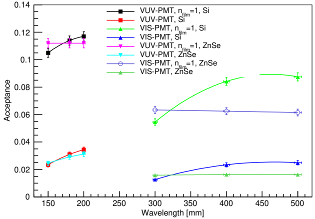


H2D2 emission spectrum



Geant4 simulation

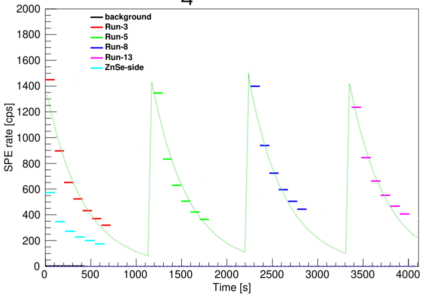




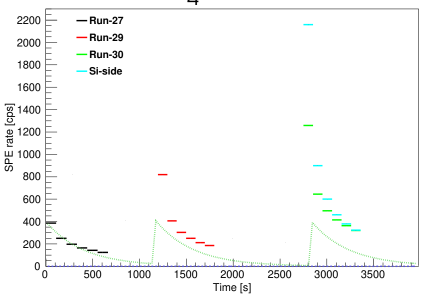
Visible photo-luminescence of ThF_4

- 0.8 kHz vis. photo-lumi. observed on top of 2.5 Hz noise and 0.6 kHz opposite side lumi.,
- Visible photo-lumi. decay time of 400 s ✗,
- projected accidental rate in 100 ns: $<10^{-5}$ Hz ✓,
- on Si sample lumi. build-up was observed over time: humidity entered during sample change?
- first run on Si sample showed lower luminescence: 0.4 kHz vs 0.8 ± 0.2 kHz ✗.

ThF_4+ZnSe



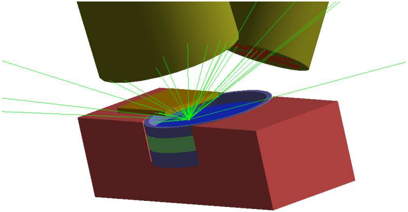
ThF_4+Si



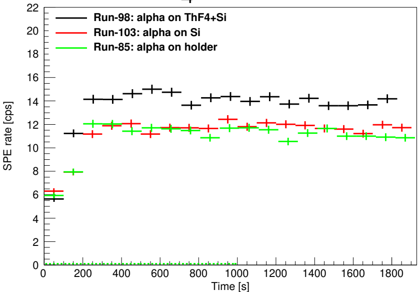
Visible radio-luminescence of ThF_4

- 2.3 Hz vis. radio-lumi. observed in ThF_4 on top of 2.5 Hz noise ✓,
- light yield from ThF_4+ZnSe sample was much larger since ZnSe is a high light yield scintillator, the subtraction of ZnSe-component was not very precise,
- comparison to Geant4 simulation shows that a similar event rate is obtained at ThF_4 light yield of: 0.1 photons/keV (0.7 ph/keVee assuming CaF_2 light quenching =0.15).

Geant4 simulation of radio-lumi.

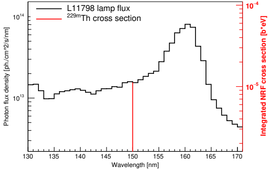


ThF_4+Si



Consequences for ^{229m}Th γ-decay search

1. H2D2-lamp at 150 nm yields: $\sim 10\ \mu W/nm$, measured $1.6\times10^{13}\ 1/\text{cm}^2\text{s nm}$,
2. it will excite ^{229m}Th:



$$\frac{dN_{Th229m}}{dt} = n_{Th229}\frac{1}{3}\frac{n^3_{ThF_4}\Gamma_\gamma}{2}\lambda_0^2\frac{dN_\gamma}{dE},$$

3. for n_{Th229}=1 kBq/1 cm^2 gives 6.5 Hz:

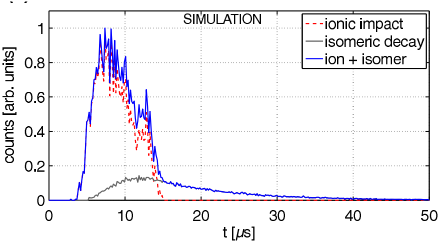
$$N_{decay} = \frac{dN_{Th229m}}{dt}\tau(1-e^{-\frac{t_{irr}}{\tau}})(1-e^{-\frac{t_{dec}}{\tau}})$$

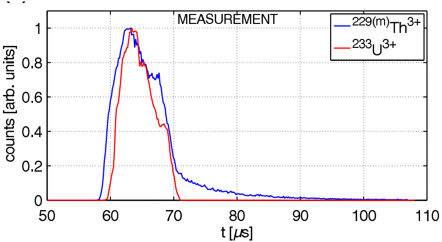
$$R = \frac{N_{decay}\times Acc\times Eff}{\tau} \sim \frac{1780\times 0.27\times 0.1}{500\ s} \sim 0.1\ Hz\,.$$

4. UV photo-lumi. of ThF_4:
   0.2 Hz/(300 nm×4.8 cm^2) for
   50 nm/1 cm^2=0.007 Hz ✓,
5. UV radio-lumi. of ThF_4:
   1.2 Hz/(300 nm×0.4 kBq) for
   25 nm×1 kBq=0.27 Hz (×5 for full chain) ✓.

LMU ^{229m}Th lifetime measurement (2017)

1. Extension of direct observation experiment[1],
2. long $^{229m}Th^{3+}$ ion transport (90 ms),
3. fast decay after impact onto MCP (neutralization),
4. neutral ^{229m}Th lifetime (IC)[2]: $7 \pm 1\ \mu s$,
5. 9 orders of magnitude lower than expected (γ): 1 h,
6. relative linewidth (IC): $\sim 10^{-11}$ (atomic clock $< 10^{-13}$),
7. neutral ^{229m}Th γ-decay branching: $\sim 10^{-9}$ (theor.).



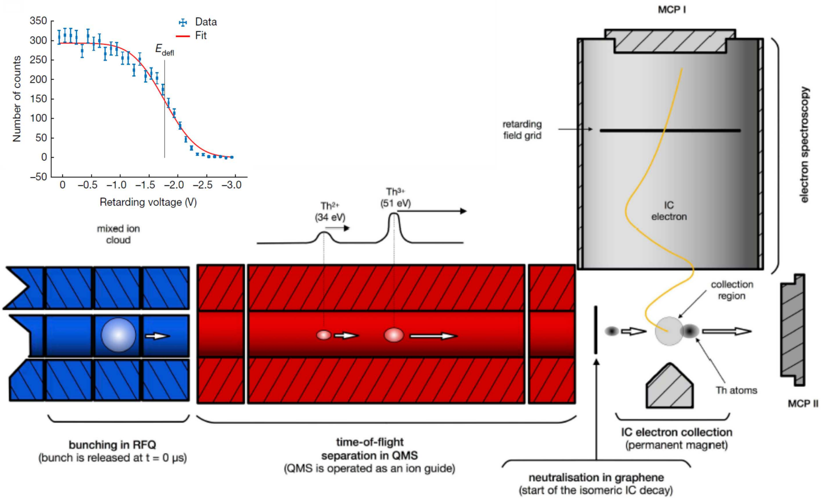


[1] *L.von der Wense et.al.Nature 533,47(2016),*

[2] *B.Seiferle et.al.Phys.Rev.Lett.118,042501(2017).* B.Seiferle et.al.Phys.Rev.Lett.118,042501(2017)

LMU ^{229m}Th energy measurement (2019)

measured 1.77 eV + 6.51 eV (binding) = 8.28 ± 0.17 eV



[1] *B. Seiferle et al., Nature 573, 243 (2019).*

▸ B. Seiferle et al., Nature 573, 243 (2019)

Previous ^{229m}Th γ-decay search

- IC decay has broad linewidth:

$$\frac{\Delta\nu}{\nu} \simeq \frac{1.4\times10^5 Hz}{2\times10^{15} Hz} \simeq 10^{-10}\,,$$

- γ-decay in neutral atom:

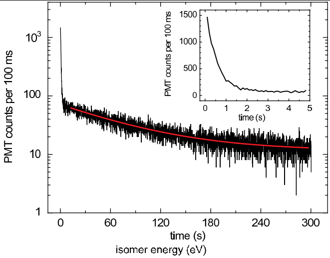
$$\frac{\Gamma_\gamma}{\Gamma_{IC}} \simeq \frac{1/1h}{1/7\mu s} \simeq 2\times10^{-9}\,,$$

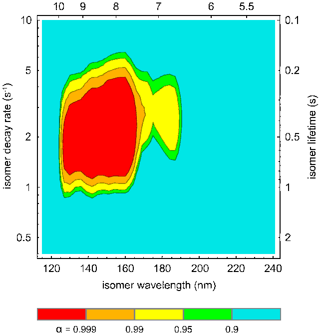
1. Th^{n+} ion traps (10^6 Th/cm^3):

$$\sigma_{\gamma_{abs}} \sim 10^{-11} cm^2 \frac{3\times10^{-19} eV}{\Delta\nu_{source}}$$

$$R_{\gamma_{dec}} \sim \rho x \sigma_\gamma \frac{n_\gamma}{\tau_\gamma\sim 1h} \sim \frac{10^2}{s}\,\frac{\Gamma_\gamma}{\Delta\nu_{source}}$$

2. wide bandgap crystals with ^{229}Th: CaF_2[1], MgF_2, $LiSrAlF_6$, $LiCaAlF_6$



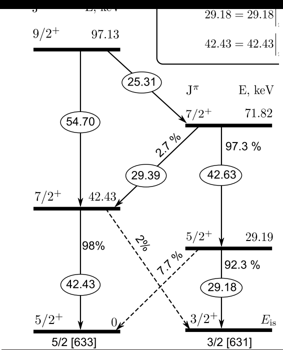


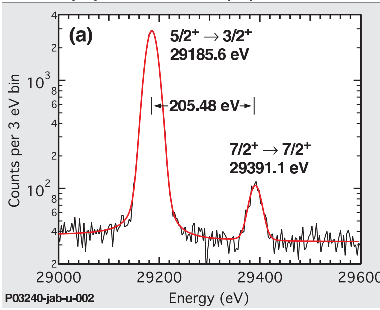
[1] S.Stellmer et.al.Phys.Rev.A97 (2018) 062506.

▸ S.Stellmer et.al.Phys.Rev.A97 (2018) 062506

Energy level of ^{229m}Th isomer

1. indirect measurement[1] by comparison of 43+29 keV γs with 26 eV resolution (HgTe μcalorimeter with 3 ms decay time),
2. 100 μCi ^{233}U source covered by 50 μm Ti foil at 3.5 cm distance,
3. direct search at ALS[2] with ^{229}Th-doped $LiSrAlF_6$ crystal excluded γ-decay in the region: $1-2 < \tau < 2000-5600$ s for $7.3 < E_\gamma < 8.8$ eV at 90 CL,
4. MCP counting observation[3], obtained upper limit $<$18.3 eV.



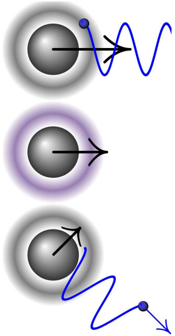


[1] B.Beck et.al.Phys.Rev.Lett.98,142501(2007).
[2] J.Jeet et.al.Phys.Rev.Lett.114,253001(2015).
[3] L.von der Wense et.al.Nature 533,47(2016).

University of Michigan/PTB

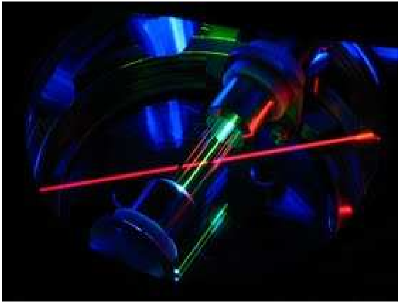
1. ion trap for $^{229}Th^{3+}$,
2. laser cooling,
3. laser ablation source,
4. low number of oscillators ($< 10^5$),
5. 100 s trapping lifetime,
6. measurement of hyperfine structure of $^{229}Th^{3+}$ atom.

[1] *A.Radnaev et al.,Phys.Rev.A86,060501(R)(2012).*
[2] *N.Huntemann et al.,Phys.Rev.Lett.116,063001(2016).*



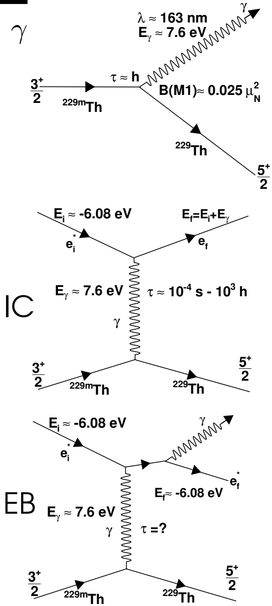
A.Kuzmich, sites.lsa.umich.edu/kuzmich-lab/

E.Peik, www.ptb.de/cms/en/ptb/fachabteilungen/abt4/fb-44/ag-443.html



TORIO-229 proposal (CSN3 meeting 22/06/2016)

1. Direct measurement of ^{229}Th lowest level energy and lifetime in the region 5-50 eV and 1 ms-1 h,
2. sum of all possible decay channels: γ, IC and EB,
3. cryogenic μcalorimeter with < 3 eV threshold and $<$0.5 eV resolution,
4. ^{233}U, ^{229}Th and ^{228}Th deposits,
5. background studies with ^{238}U and ^{232}Th samples,
6. signal studies with ^{235m}U (77 eV, 26 m, E3), ^{239}Pu deposit.



μcalorimeter with embedded source (2016)

Cryogenic μcalorimeter heat capacity budget:

1. 3.8 ng ^{233}U target 17.3 fJ/K,
2. Ir TES 8 fJ/K,
3. Au absorber 0 fJ/K,
4. Si membrane 0.013 fJ/K.

Energy resolution:

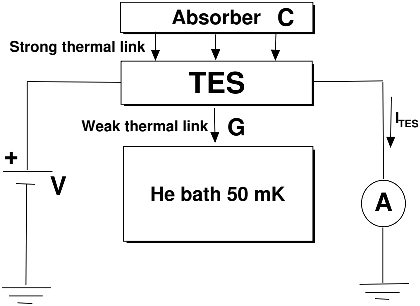
$$\sigma_E \sim \sqrt{kT^2C} \sim 0.42eV \ .$$

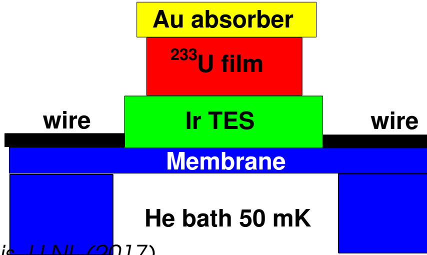
Energy dynamic range 160 eV:

$$\Delta T \sim \frac{E_{dep}}{C} < 1mK \ .$$

Target activity: 3.8 ng ^{233}U,
1.3 Bq α activity (A=0.35$\times M_U[ng]$),
or 100 ^{229m}Th/h.
Signal decay time:

$$\tau_D \sim \frac{C}{G} \sim 2.5ms \ .$$





[1] F.Ponce, PhD thesis, LLNL (2017).

μcalorimeter pile-up

Updated decay rate estimate:

1. 3.8 ng ^{233}U target (10^{13} atoms)
2. 1.3 Bq α activity ($A=0.35\times M_U[ng]$),
3. 100 ^{229m}Th/h,
4. 10^{-3} ^{229m}Th γ decay/y.

Signal decay time:

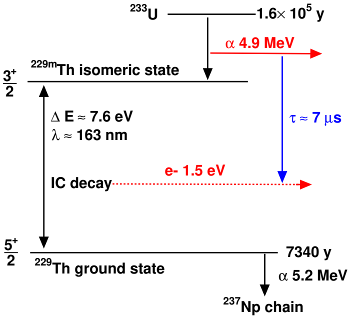
$$\tau_D \sim \frac{C}{G} \sim 2.5ms\ .$$

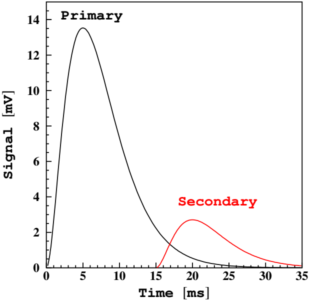
Separation of two decays:

$$\Delta t > \tau_D ln \frac{E_{primary}}{E_{secondary}}\ .$$

Probability of separated decay:

$$P \sim \exp\left[-\frac{2.5ms}{7\mu s}\right] \sim 10^{-155}\ .$$



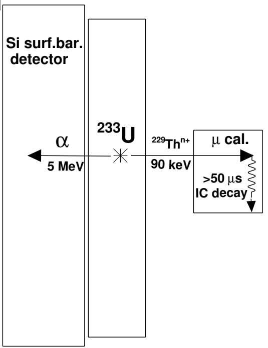


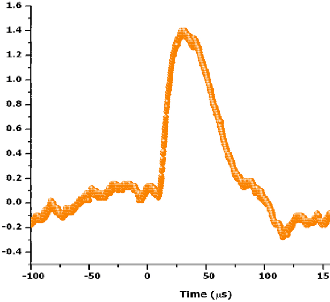
TORIO-229 setup (CSN3 meeting 22/06/2017)

- Fast cryogenic μcalorimeter with signal full decay time $\Delta t < 50\ \mu$s[1],
- α-^{229}Th^{+} coincidence,
- ^{233}U deposited on Si detector,
- long. range of 90 keV ^{229}Th^{+} in U is 6.7 nm (path 10.7 nm), in UO_2 is 8.3 nm (path 16.1 nm),
- 10 nm thick ^{233}U film with 1 nm Mg layer or 20 nm thick $^{233}UO_2$ film, activity A_U<5 kBq,
- expected rate of time-separated events (assumes $t_{IC} = 7\ \mu$s):

$$R_{IC}^{\mu cal} \sim A_U B_{229m} A_{\mu cal}^{geom} e^{-\Delta t/t_{IC}} \ ,$$

$$R_{IC}^{\mu cal} \sim 10^4 * 0.02 * 10^{-4} * 10^{-3} \sim \frac{1}{day} \ .$$





[1] D.Bagliani et al.,J.Low.Temp.Phys.151,234(2008).

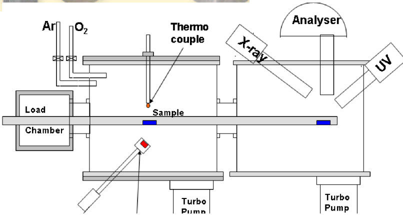
Primary sources (R. Caciuffo, JRC, Karlsruhe)

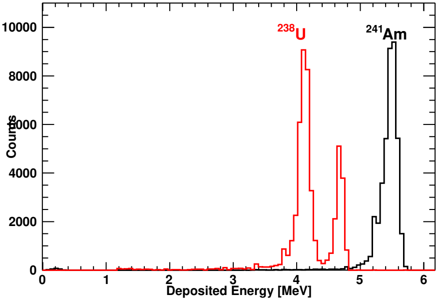
First source was deposited by sputtering on active surface of Si detector (300 mm^2), in form of UO_2 dielectric film with diameter 10mm and thickness 10 nm:

1. ^{238}U <0.2 Bq (+0.1 Bq ^{234}U), delivered for background measurements,
2. ^{233}U <5 kBq, authorization received, waiting for working TES prototype.

Source requirements:

- E-loss limits thickness $\leq$20 nm,
- Si-rate limits activity <40 kHz.





μcalorimeter principles

μcalorimeter operation:

1. constant current flows through TES without resistance,
2. deposited energy is transformed in heat,
3. heat increases TES resistance,
4. Resistance reduces flowing current,
5. current read by SQUID.

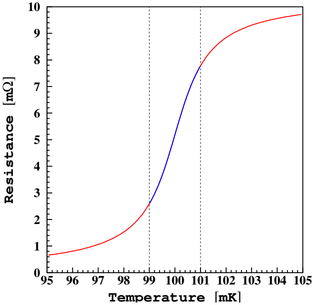
Signal:

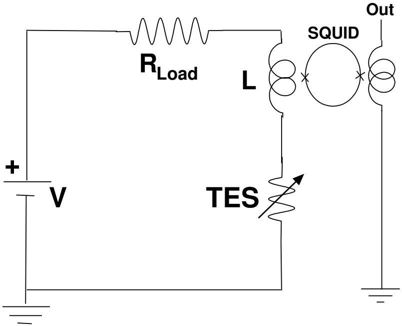
$$\Delta T = E_{dep}/C \sim 2fJ/K \ .$$

Energy resolution:

$$\sigma_E \sim \sqrt{kT^2C} \ .$$

SQUID noise: 1 pA/$\sqrt{Hz}$.





μcalorimeter optimization

- Signal decay time at $T = 100$ mK:

$$\tau \sim \frac{C_{bottleneck} \sim 1\,fJ/K}{G \sim 10^5\,fW/K} \sim 10\mu s: \text{small}$$

- T-dependence:

$$\frac{C \simeq \gamma V T}{G \sim G_{ep} \simeq 5 \Sigma V T^4} = \frac{\gamma}{5\Sigma T^3}: \text{large}$$

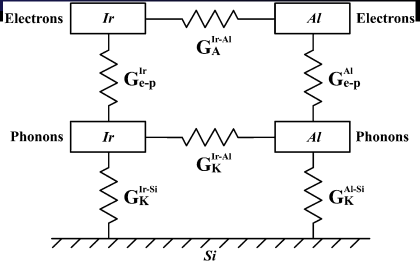
- $T = 100\text{mK} \rightarrow 300\text{mK}$:

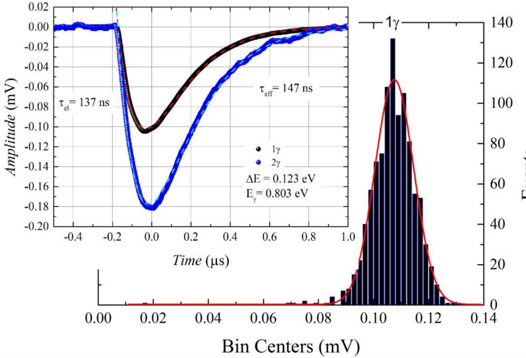
$$\tau(300mK) \sim \frac{\tau(100mK)}{30}$$

- noise power:

$$NEP \sim \sqrt{4 k_b T^2 G},$$

$$V_{noise} \sim [20 k_b \Sigma V]^{1/4} T^{3/2}$$

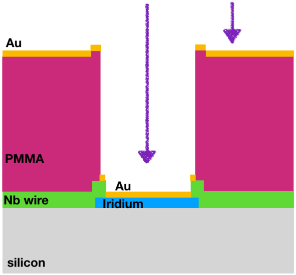


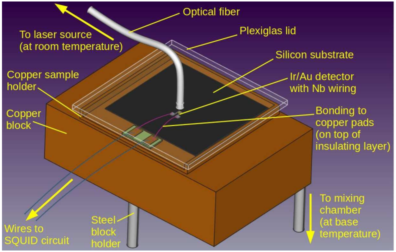


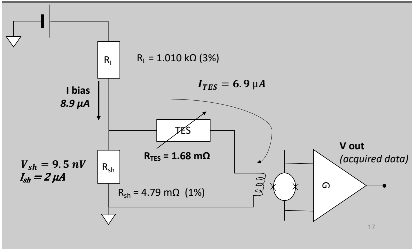
[1] D.Bagliani et al.,J.Low.Temp.Phys.151,234(2008).
[2] C.Portesi et al.,IEEE Trans.App.Supercond.25,3(2015).

TES development details

1. design lithographic mask,
2. tested Ti-Au and Ir TESs,
3. tested Nb and Al wiring,
4. PLD of Ir, sputtering of Nb,
5. PMMA+Au shadow mask[1],
6. wire bonding to SQUID,
7. alignment of fiber,
8. SQUID calibrations.





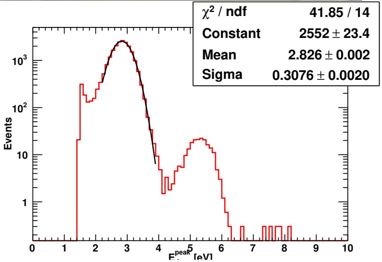


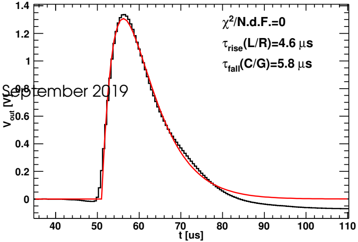
[1] M.Fedkevych et al., 10.1109/TASC.2021.3063328

▸ M.Fedkevych et al.,IEEE TASC (2021)

Detector development and characterization

1. developed Ir TES satisfying experimental requirements[1]: signal fall time $\tau < 10\ \mu s$, resolution $\Delta E < 1$ eV,
2. $15\times 26\ \mu m^2$, 150 nm thick Ir,
3. few μm PMMA + 50 nm Au shadow mask,
4. transition $T_c = 110.5$ mK,
5. laser calibrations and Poissonian fit,
6. analysis of data and extraction of TES parameters.



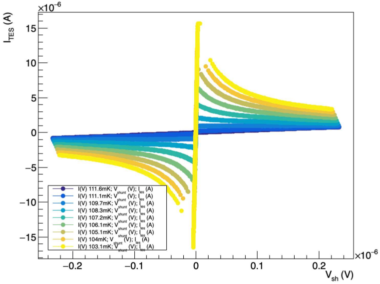


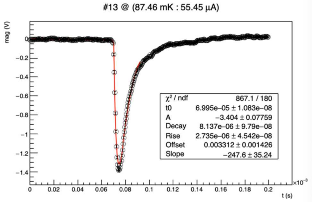
September 2019

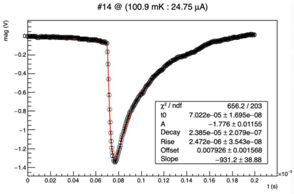
TES characterization details

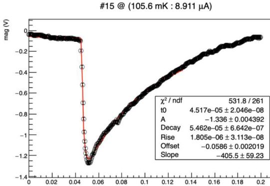
1. high bias current 60μA,
2. operation at 90 mK$< T_C$,
3. large negative electrothermal feedback[1]:

$$\tau_{eff} \simeq \frac{\tau_{th}}{1+\alpha P/GT} \ .$$









[1] *M.Biasotti, internal report (2020),*