

# Prospects of direct Th229m energy measurement and search for its $\gamma$ decay

CSN3/TORIO\_229

M. Osipenko<sup>1</sup>

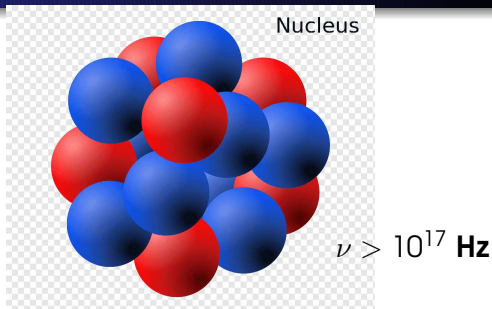
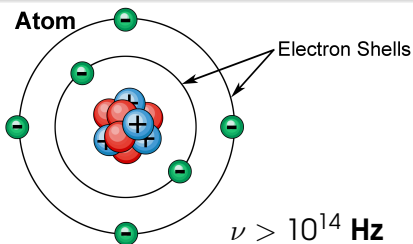
In collaboration with

M. Taiuti<sup>2</sup>, R. Caciuffo<sup>3</sup>, R. Malmbeck<sup>3</sup>, F. Gatti<sup>2</sup>,  
M. Giovannini<sup>2</sup>, M. Ripani<sup>1</sup>, L. Ferrari Barusso<sup>2</sup>, B. Siri<sup>1,2</sup>,  
M. Fedkevych<sup>2</sup>, M. Biasotti<sup>2</sup>, V. Ceriale<sup>2</sup>

<sup>1</sup>INFN Genova, <sup>2</sup>Università di Genova, <sup>3</sup>JRC Karlsruhe

Seminar, LNL, 15 April 2021

# Atomic and nuclear scales



**Radius: 0.5-2 Å**  
**Excitations: eV - keV**

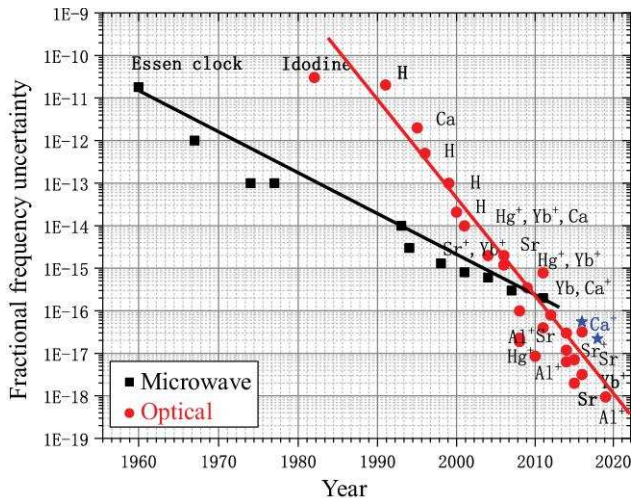
- $1 \text{ s} \equiv 9.192.631.770$  RF oscillations in Cs clock,
- *highly monochromatic coherent lasers (eV),*
- visible-microwave comb ( $\frac{\Delta\nu}{\nu} \sim 10^{-18}$ ),
- optical ion clocks:  $\text{Yb}^+$ ,  $\text{Al}^+$ ,  $\text{Ca}^+$ ,  $\text{Sr}^+$ ,  $\text{Hg}^+$  etc.

**Radius: 1.3-8 fm**  
**Excitations: keV - MeV**

- incoherent  $\gamma$ -sources,
- Mössbauer spectroscopy ( $\frac{\Delta\nu}{\nu} > 10^{-22}$ ),
- broad linewidth FELs ( $\frac{\Delta\nu}{\nu} \sim 10^{-3} \div 10^{-5}$ ),
- 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 1999 (Nobel Prize 2005).



<sup>1</sup> K.Gao ,Nat.Sci.Rev.7, 1799 (2020).

► K.Gao ,Nat.Sci.Rev.7, 1799 (2020).

# 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.

The  $\text{Ca}^+$ -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

<sup>1</sup> K.Gao ,*Nat.Sci.Rev.*7, 1799 (2020).

▶ K.Gao ,*Nat.Sci.Rev.*7, 1799 (2020).

# Nuclear clock precision

- 1 first idea from E. Peik and C. Tamm, Eur.Phys.Lett.61, 181 (2003),
- 2 small coupling of external fields to nucleus,
- 3 atomic shell screening,
- 4 small black-body radiation.

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

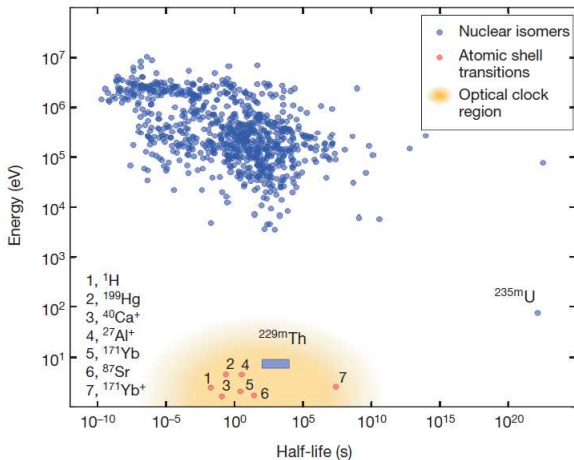
<sup>1</sup> C.J. Campbell et al., Phys.Rev.Lett. 108, 120802 (2012)

► C.J. Campbell et al., Phys.Rev.Lett. 108, 120802 (2012)

# Known nuclear isomers

- 1  $^{235m}\text{U}$  at 76.8 eV with half life 25 min. (IC) or  $10^{14}$  years ( $\gamma$ ),
- 2  $^{229m}\text{Th}$  at 8 eV with half life  $7 \mu\text{s}$  (IC) or 1 hour ( $\gamma$ ),

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



<sup>1</sup>L. von der Wense et al., *Nature* 533, 47 (2016)

► L. von der Wense et al., *Nature* 533, 47 (2016)

- 43 years from the first indication to the final confirmation,
- $\gamma$  decay has not been observed so far,
- induced excitation of  $^{229m}\text{Th}$  not yet demonstrated.

Year	Energy (eV)	Ref.
1976	<100	L.A. Kroger, C.W. Reich, Nucl.Phys. <b>A259</b> , 29
1990	$1\pm 4$	C.W. Reich, R. Helmer, Phys.Rev.Lett. <b>64</b> , 271
1994	$3.5\pm 1$	R. Helmer, C.W. Reich, Phys.Rev. <b>C49</b> , 1845
2007	$7.6\pm 0.5$	B.R. Beck <i>et al.</i> , Phys.Rev.Lett. <b>109</b> , 142501
2019	$8.3\pm 0.2$	B. Seiferle <i>et al.</i> , Nature <b>573</b> , 243

# Non-spherical $^{229}\text{Th}$ rotational splitting

- $^{229}\text{Th}$  deformed nucleus (axial),
- Nilsson model<sup>1</sup> level scheme<sup>2</sup>:

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

$$\Omega = j_z, [2j + 1]$$

$$N = n_z + n_\perp$$

$$\Lambda = l_z, [2n_\perp + 1]$$

- deformation parameter:

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

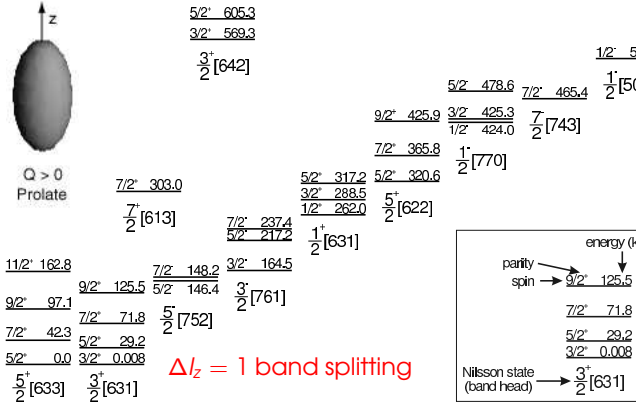
- $^{229}\text{Th}$  prolate<sup>3,4,5</sup>

$$\delta \sim 0.1, L_z \sim 1.05L_\perp$$

$$M1\left(\frac{3^+}{2} \rightarrow \frac{5^+}{2}\right)$$



$Q > 0$   
Prolate



$\Delta I_z = 1$  band splitting

<sup>1</sup> S. Nilsson, *Matematisk-fysiske Meddelelser*, 29(16) (1955).

<sup>2</sup> G. Musiol et al., *Kern und El. physik.*, Weinheim (1988).

<sup>3</sup> E. Ruchowska et al., *Phys. Rev. C* 73, 044326 (2006).

<sup>4</sup> A. Hayes et al., *Phys. Rev. C* 78, 024311 (2008).

<sup>5</sup> E. Litvinova et al., *Phys. Rev. C* 79, 064303 (2009).



- 1 magnetic dipole transition M1:  $\frac{3}{2}^+ \rightarrow \frac{5}{2}^+$ ,
- 2 lifetime<sup>1,2,3</sup> ( $\gamma$  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  $10^{-4}$  s in metal<sup>4</sup> (overestimates  $^{235m}\text{U}$  lifetime by 3 orders of magnitude),
- 4 energy scaling from the same transition  $\frac{3}{2}^+ \rightarrow \frac{5}{2}^+$  of  $^{233}\text{U}$  (312 keV) level yields lifetime of 4 h<sup>1</sup>,
- 5 scaling from another M1  $\frac{9}{2}^+ \rightarrow \frac{7}{2}^+$  transition yields lifetime of 0.3 h<sup>5</sup>,
- 6 electronic bridge (EB) process may be important in presence of resonant atomic levels.

<sup>1</sup>R. Helmer and C. Reich. *Phys.Rev.C*49,1845(1994).

<sup>2</sup>V.Strizhov and E.Tkalya. *Sov.Phys.JETP*,72,387(1991).

<sup>3</sup>E.Ruchowska et al., *Phys.Rev.C*73,044326(2006).

<sup>4</sup>E.Tkalya. *JETP Lett.* 70,371(1999).

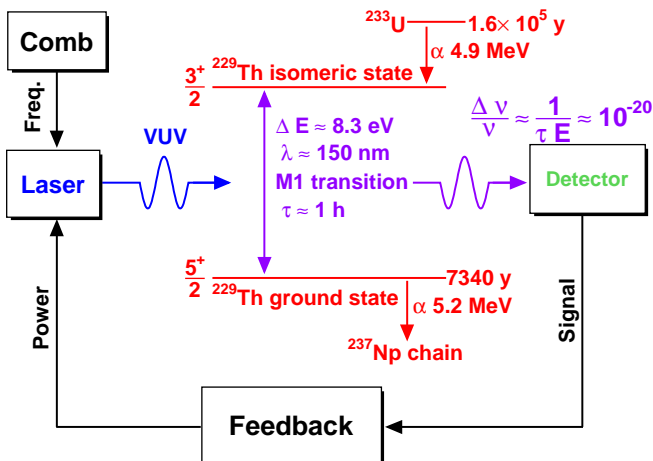
<sup>5</sup>J.Jeet et al. *Phys.Rev.Lett.* 114,253001(2015).

# $^{229}\text{Th}$ -based Nuclear Clock

- 1 Lowest nuclear excited state<sup>1</sup>,
- 2 dominant IC decay<sup>2</sup> can be suppressed,
- 3 VUV-laser excitation,
- 4 very narrow  $\gamma$ -linewidth<sup>3</sup>,
- 5  $N=10^5 \div 10^{12}$  oscillators<sup>4,5</sup>,

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

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



<sup>1</sup> L.Kroger and C.Reich, Nucl.Phys.A259, 29(1976).

<sup>2</sup> B.Seiferle et al., Nature 573, 243 (2019).

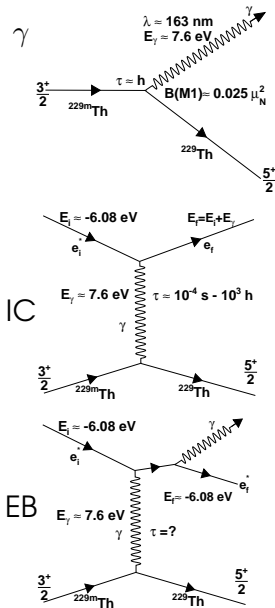
<sup>3</sup> V.Strizhov and E.Tkalya, Sov.Phys.JETP 72, 387 (1991).

<sup>4</sup> C.Campbell et al., Phys.Rev.Lett. 102, 233004 (2009).

<sup>5</sup> R.Jackson et al., J.Phys.Cond.Mat.21, 325403 (2009).

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

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



# $\mu$ calorimeter with embedded source (2016)

Cryogenic  $\mu$ calorimeter heat capacity budget:

- 1 3.8 ng  $^{233}\text{U}$  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:

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

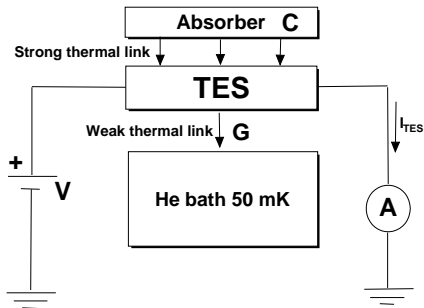
Energy dynamic range 160 eV:

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

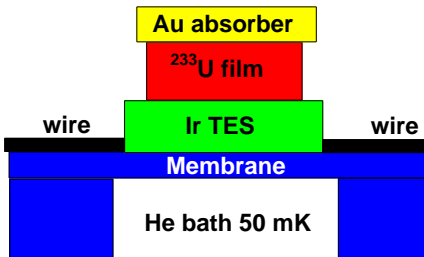
Target activity: 3.8 ng  $^{233}\text{U}$ ,  
1.3 Bq  $\alpha$  activity ( $A=0.35 \times M_U[\text{ng}]$ ),  
or 100  $^{229\text{m}}\text{Th}/\text{h}$ .

Signal decay time:

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

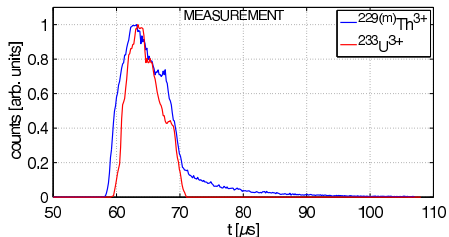
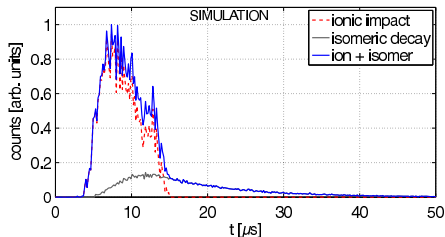


<sup>1</sup>F.Ponce, PhD thesis, LLNL (2017).



# LMU $^{229m}\text{Th}$ lifetime measurement (2017)

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



<sup>1</sup>L.von der Wense et.al.Nature 533,47(2016),

<sup>2</sup>B.Seiferle et.al.Phys.Rev.Lett. 118,042501(2017).

# $\mu$ calorimeter pile-up

Updated decay rate estimate:

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

Signal decay time:

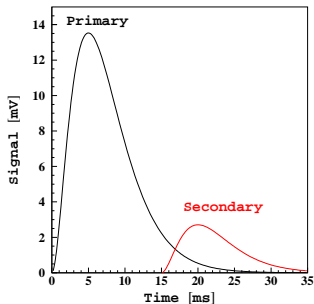
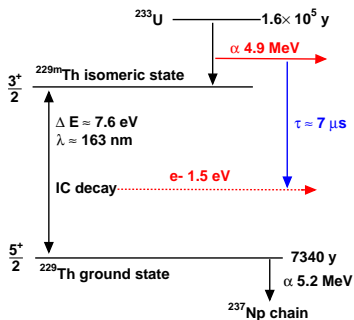
$$\tau_D \sim \frac{C}{G} \sim 2.5 \text{ms}.$$

Separation of two decays:

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

Probability of separated decay:

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

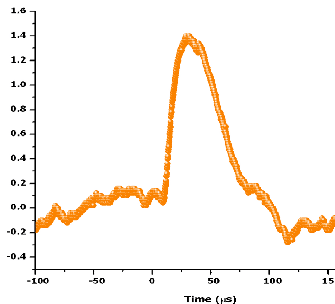
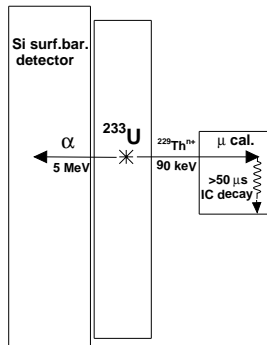


# TORIO-229 setup (CSN3 meeting 22/06/2017)

- Fast cryogenic  $\mu$ calorimeter with signal full decay time  $\Delta t < 50 \mu\text{s}^1$ ,
- $\alpha$ - $^{229}\text{Th}^+$  coincidence,
- $^{233}\text{U}$  deposited on Si detector,
- long. range of 90 keV  $^{229}\text{Th}^+$  in U is 6.7 nm (path 10.7 nm), in  $\text{UO}_2$  is 8.3 nm (path 16.1 nm),
- 10 nm thick  $^{233}\text{U}$  film with 1 nm Mg layer or 20 nm thick  $^{233}\text{UO}_2$  film, activity  $A_U < 5 \text{ kBq}$ ,
- expected rate of time-separated events (assumes  $t_{IC} = 7 \mu\text{s}$ ):

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

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



<sup>1</sup>D. Bagliani et al., *J. Low. Temp. Phys.* 151, 234 (2008).

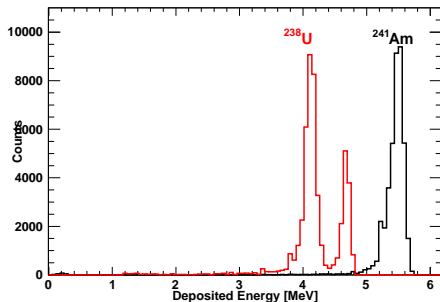
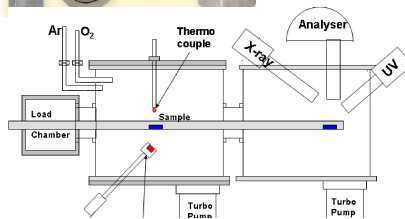
# Primary sources (R. Caciuffo, JRC, Karlsruhe)

First source was deposited by sputtering on active surface of Si detector ( $300 \text{ mm}^2$ ), in form of  $\text{UO}_2$  dielectric film with diameter 10mm and thickness 10 nm:

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

Source requirements:

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





# $\mu$ calorimeter principles

$\mu$ calorimeter 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.

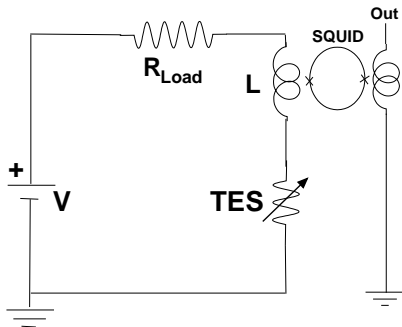
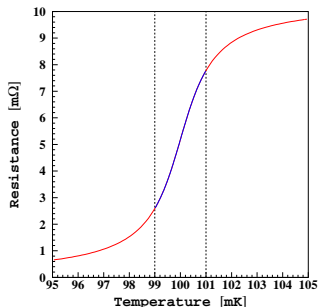
Signal:

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

Energy resolution:

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

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



# $\mu$ calorimeter optimization

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

$$\tau \sim \frac{C \sim 1 \text{ fJ/K}}{G \sim 10^5 \text{ fW/K}} \sim 10 \mu\text{s}.$$

- $T$ -dependence:

$$\frac{C \simeq \gamma VT}{G \sim G_{ep} \simeq 5\Sigma VT^4} = \frac{\gamma}{5\Sigma T^3}$$

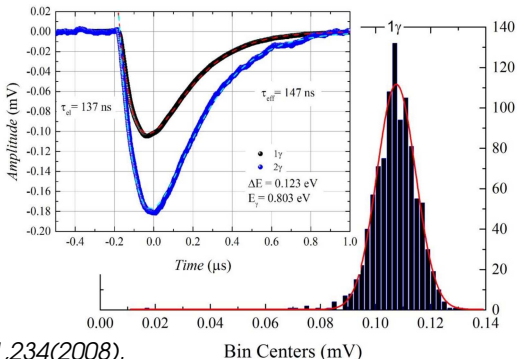
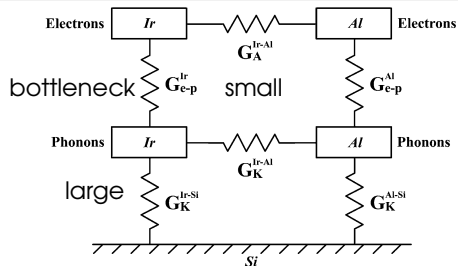
- $T = 100 \text{ mK} \rightarrow 300 \text{ mK}$ :

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

- noise power:

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

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

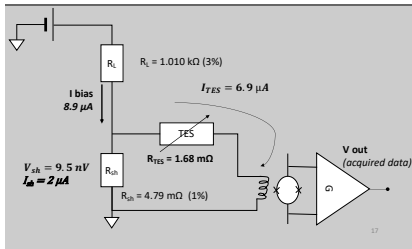
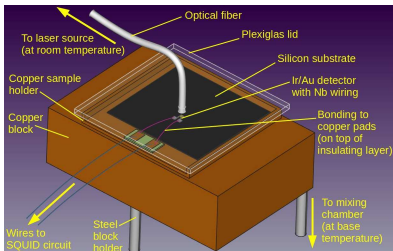
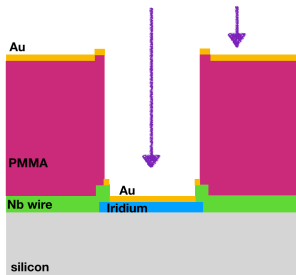


<sup>1</sup> D. Bagliani et al., *J. Low. Temp. Phys.* 151, 234 (2008).

<sup>2</sup> C. Portesi et al., *IEEE Trans. App. Supercond.* 25, 3 (2015).

# TES development details

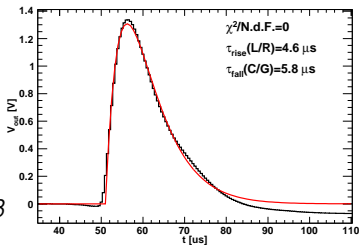
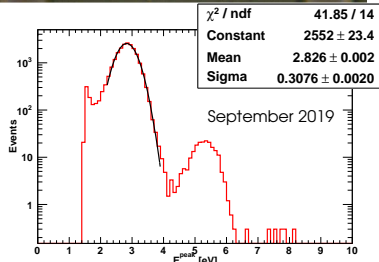
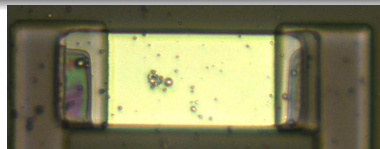
- 1 design lithographic mask,
- 2 tested Ti-Au and Ir TESs,
- 3 tested Nb and Al wiring,
- 4 PLD of Ir, sputtering of Nb,
- 5 PMMA+Au shadow mask<sup>1</sup>,
- 6 wire bonding to SQUID,
- 7 alignment of fiber,
- 8 SQUID calibrations.



<sup>1</sup> M. Fedkevych et al., 10.1109/TASC.2021.3063328

# Detector development and characterization

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

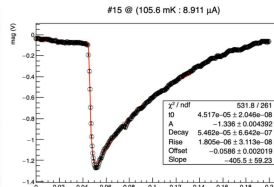
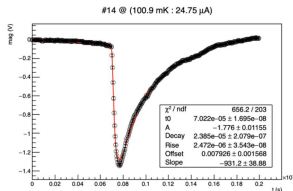
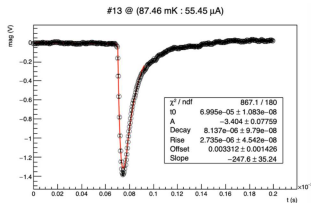
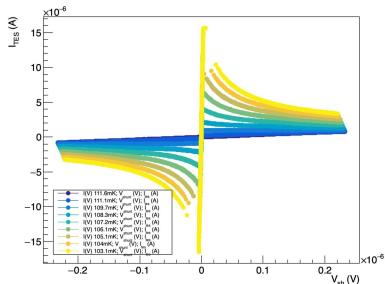


<sup>1</sup> M. Fedkevych et al, 10.1109/TASC.2021.3063328

# TES characterization details

- 1 high bias current  $60\mu\text{A}$ ,
- 2 operation at  $90\text{ mK} < T_C$ ,
- 3 large negative electrothermal feedback<sup>1</sup>:

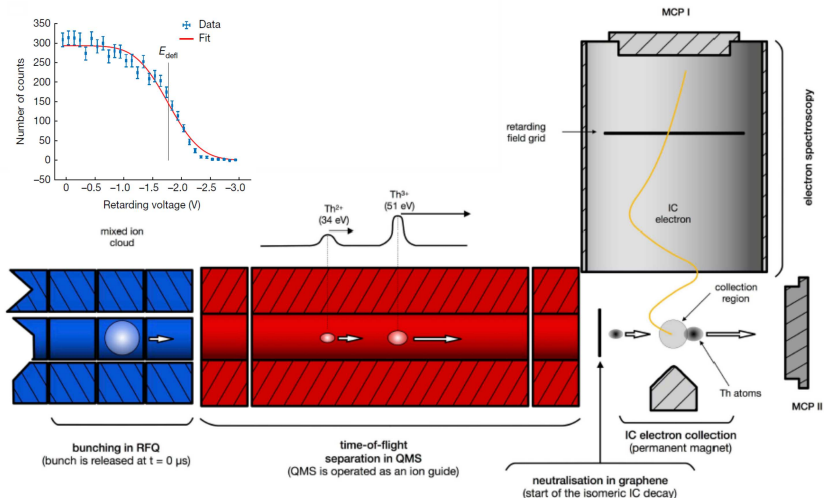
$$\tau_{\text{eff}} \simeq \frac{\tau_{\text{th}}}{1 + \alpha P / GT}$$



<sup>1</sup>M. Biasotti, internal report (2020),

# LMU $^{229m}\text{Th}$ energy measurement (2019)

measured  $1.77 \text{ eV} + 6.51 \text{ eV (binding)} = 8.28 \pm 0.17 \text{ eV}$



<sup>1</sup>B. Seiferle et al., Nature 573, 243 (2019).

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

- IC decay has broad linewidth:

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

- $\gamma$ -decay in neutral atom:

$$\frac{\Gamma_\gamma}{\Gamma_{IC}} \simeq \frac{1/1h}{1/7\mu\text{s}} \simeq 2 \times 10^{-9},$$

- 1  $\text{Th}^{n+}$  ion traps ( $10^6 \text{ Th/cm}^3$ ):

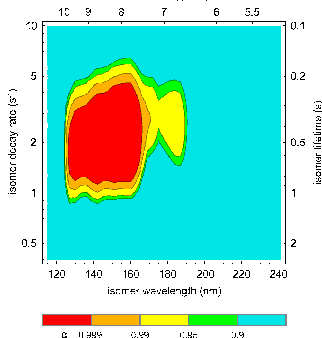
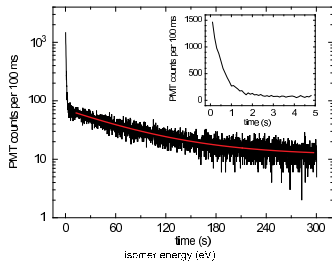
$$\sigma_{\gamma\text{abs}} \sim 10^{-11} \text{ cm}^2 \frac{3 \times 10^{-19} \text{ eV}}{\Delta\nu_{\text{source}}}$$

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

- 2 wide bandgap crystals with

$^{229}\text{Th}$ :  $\text{CaF}_2^1$ ,  $\text{MgF}_2$ ,  
 $\text{LiSrAlF}_6$ ,  $\text{LiCaAlF}_6$

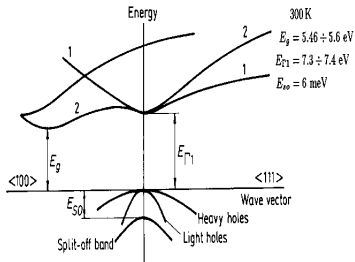
<sup>1</sup>S.Stellmer et al. Phys.Rev.A97 (2018) 062506.



# Chemical binding IC suppression

- IC decay requires:  
 $E_{decay} > E_{gap}$ ,
- find Th-compound with  
 $E_{gap} > 8.3$  eV,
- theoretical calculations<sup>1</sup> suggested  $\text{Na}_2\text{ThF}_6$  and  $\text{ThF}_4$  would have  
 $E_{gap} > 8.9$  eV, while  $\text{ThX}_4$  ( $\text{X}=\text{Cl}, \text{Br}, \text{I}$ )  $E_{gap} < 6.5$  eV.

Comp.	Bandgap (eV)
$\text{CaF}_2$	12
$\text{UO}_2$	1.3
$\text{UC}, \text{UN}, \text{UCI}, \text{UF}$	2-3
Other	?



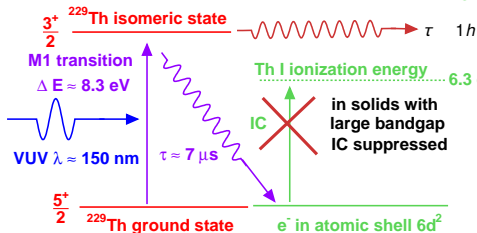
<sup>1</sup> J.K.Ellis et al., *Inorg.Chem.* 53 (2014) 6769.

▶ J.K.Ellis et al., *Inorg.Chem.* 53 (2014) 6769

Th II ionization energy...12.1 eV

ThF<sub>4</sub> conduction band 10.2 eV

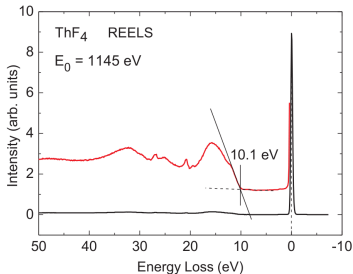
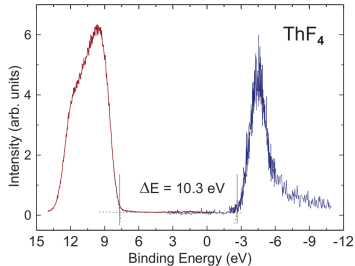
Th I ionization energy... 6.3 eV





# ThF<sub>4</sub> bandgap measurements (JRC, Karlsruhe)

- 1 25 nm film of ThF<sub>4</sub> 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

# ThF<sub>4</sub> $\gamma$ -decay search

1 H2D2-lamp yields:  
 $\sim 10 \mu W/nm$  at 150 nm,

2 it will excite  $^{229m}\text{Th}$ :

$$N_{\text{Th}229m} = n_{\text{Th}229} \frac{1}{3} \frac{\Gamma_{\gamma}}{2} \lambda_0^2 \frac{dN_{\gamma}}{dE},$$

3 1 kBq in 1 cm<sup>2</sup> gives:

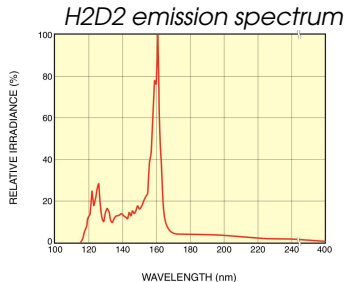
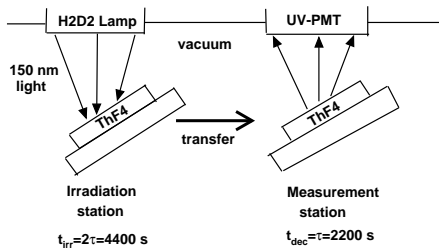
$$N_{\text{Th}229m} \simeq 6.7 \times 10^{-14} nm \frac{dN_{\gamma}}{d\lambda}$$

$$\frac{dN_{\text{Th}229m}}{dt} \simeq 0.5 \text{ Hz},$$

$$S = \frac{dN_{\text{Th}229m}}{dt} \tau (1 - e^{-\frac{t_{\text{irr}}}{\tau}}) (1 - e^{-\frac{t_{\text{dec}}}{\tau}})$$

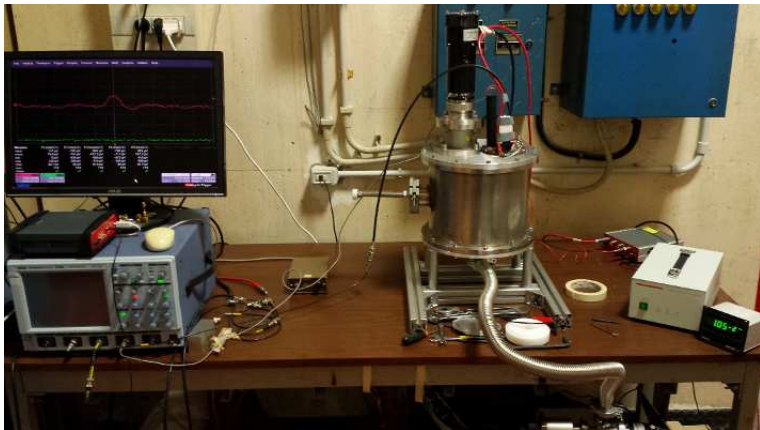
$$S \times \text{Acc} \times \text{Eff} / N \sim \frac{610 \times 0.1 \times 0.3}{R_{\text{bkg}} 2200 \text{ s}}$$

$> 4\sigma$  deviation in 10-100  
 measurements.



# ThF<sub>4</sub> setup in Genova

- 1 Hamamatsu VUV-lamp L11798 and solar blind PMT R10454, visible veto PMT R1450,
- 2 vacuum chamber for  $10^{-2}$  mbar,
- 3 commercial ThF<sub>4</sub> samples from II-VI Inc.,
- 4 parallel setup in glove box at JRC (Karlsruhe).



# Variations of $\alpha_{EM}$ (with respect to $\alpha_S$ )

- V. Flambaum in Phys.Rev.Lett.97, 092502 (2006):

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

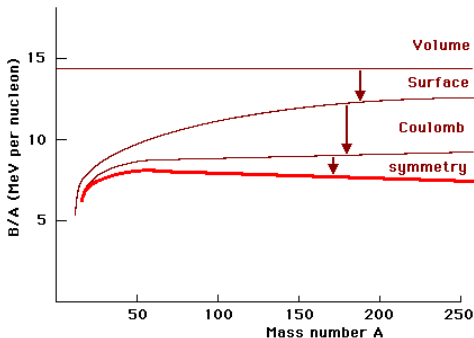
leading to sensitivity:

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

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

- in PRA102 052833 (2020)  $k \simeq -10^4$  was obtained, leading to  $\delta\nu \sim 200$  Hz/year.

► PFadeev et al., Phys.Rev.A102 (2020) 052833



# Other applications

- 1 topological and axionic dark matter detection:  
 $\alpha_{EM}$  and Schiff moment;
- 2  $^{229m}\text{Th}$ -based nuclear laser:  
Zeeman splitting in 100 T magnetic field;
- 3 GPS (precision 3-5 m):  
atmospheric disturbance, clock synchronization and stability, satellite position;
- 4 Chronometric geodesy:  
 $10^{-19}$  clock geodesy at 1 mm, earthquake prediction, search for natural resources, tests of general relativity, clock-based gravitational wave detection.

For review see: [▶ L. von der Wense and B. Seiferte, Eur. Phys. J. A56 \(2020\) 277.](#)

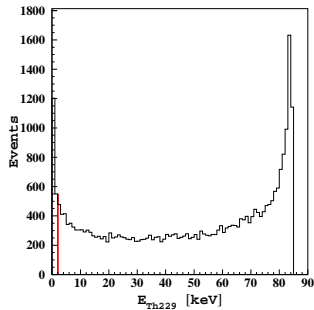
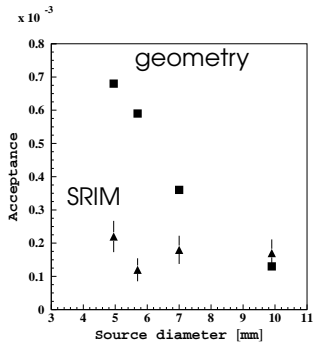
# Summary

- $^{229m}\text{Th}$  is the lowest known nuclear isomer,
- latest measurements give its energy around 8.3 eV, corresponding to 150 nm wavelength,
- neutral  $^{229m}\text{Th}$  atom decays through IC in 7  $\mu\text{s}$ ,
- if IC decay is suppressed (by ionization of chemical binding)  $\gamma$ -decay is expected to occur in 1 hour,
- $\gamma$ -decay-only linewidth is suitable for very precise nuclear clock ( $10^{-19}$  precision on a single atom in cooled trap,  $10^{-16}$  precision on a single atom in crystal),
- $\gamma$ -decay is not yet observed and our experiment is aiming to search for it in  $\text{ThF}_4$  crystal.

Backup slides

# Acceptance Simulations

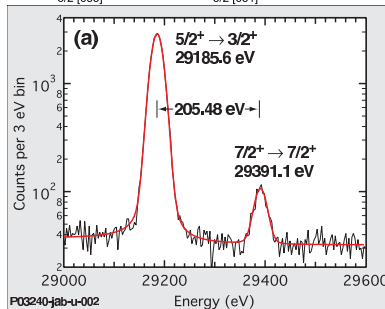
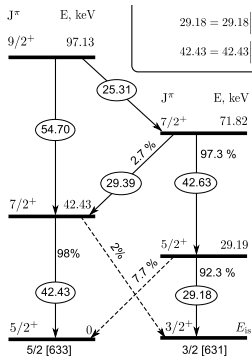
- 1  $\mu$ Cal. is small  $200 \times 200 \mu\text{m}^2$ ,
- 2 it can be installed at 1 mm from U-source,
- 3 source activity fixed  $< 5 \text{ kBq}$ ,
- 4 geometrical acceptance falls with source diameter,
- 5 SRIM acceptance is flat,
- 6 if  $\tau_D > 7 \mu\text{s}$  and noise  $\simeq$  signal only recoils with  $< 100 \text{ eV}$  have to be selected,
- 7 further reduction of rate by a factor of 10.





# Energy level of $^{229m}\text{Th}$ isomer

- 1 indirect measurement<sup>1</sup> by comparison of  $43+29$  keV  $\gamma$ s with 26 eV resolution (HgTe  $\mu$ calorimeter with 3 ms decay time),
- 2  $100 \mu\text{Ci } ^{233}\text{U}$  source covered by  $50 \mu\text{m}$  Ti foil at 3.5 cm distance,
- 3 direct search at ALS<sup>2</sup> with  $^{229}\text{Th}$ -doped  $\text{LiSrAlF}_6$  crystal excluded  $\gamma$ -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<sup>3</sup>, obtained upper limit  $< 18.3$  eV.



<sup>1</sup> B. Beck et al. Phys. Rev. Lett. 98, 142501 (2007).

<sup>2</sup> J. Jeet et al. Phys. Rev. Lett. 114, 253001 (2015).

<sup>3</sup> L. von der Wense et al. Nature 533, 47 (2016).

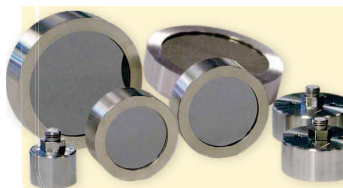
# Primary sources (Roberto Caciuffo, JRC, Karlsruhe)

Sources being to be deposited on active surface of Si detector (300 mm<sup>2</sup>), with film diameter 10mm:

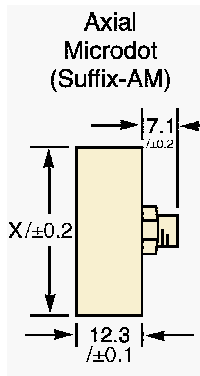
- 1 <sup>238</sup>U metal (10 nm): 0.2 Bq,
- 2 <sup>233</sup>U metal (10 nm): <5 kBq,
- 3 <sup>239</sup>Pu metal (10 nm): <5 kBq,

+1 nm Mg capping layer, or

- 1 <sup>238</sup>UO<sub>2</sub> dielectric (20 nm),
  - 2 <sup>233</sup>UO<sub>2</sub> dielectric (20 nm),
  - 3 <sup>239</sup>PuO<sub>2</sub> dielectric (20 nm),
- E-loss limits thickness  $\leq 20$  nm,
  - Si-rate limits activity <40 kHz,
  - <sup>239</sup>Pu smaller size: <2 mm,
  - <sup>238</sup>U rate too low (use <sup>234</sup>U?).

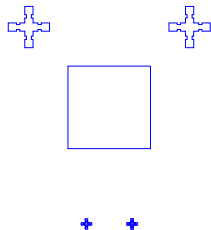
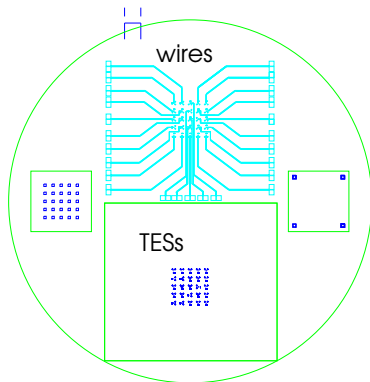


X=28.6 mm



# Lithographic mask design

- 1  $5 \times 5$  TESs of different size,
- 2 maximum size  $300 \mu\text{m}$ ,
- 3 minimum size  $10 \mu\text{m}$ ,
- 4  $30 \mu\text{m}$  arrays read in parallel,
- 5 Be mask to reduce  $\alpha$  heating.



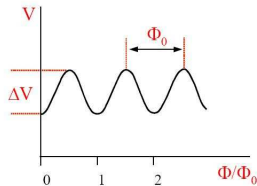
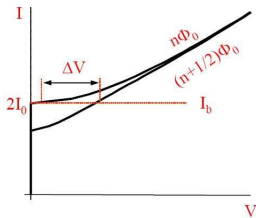
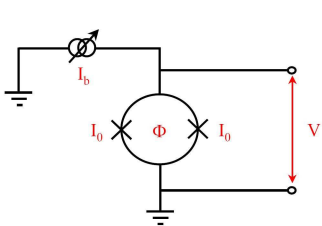
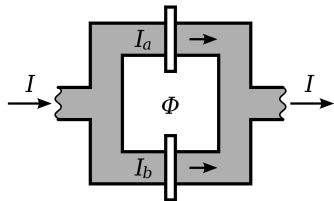
# DC SQUID principles

SQUID current:

$$I = I_0 \left\{ \sin \left( \delta_0 + \frac{e}{\hbar} \Phi \right) + \sin \left( \delta_0 - \frac{e}{\hbar} \Phi \right) \right\}$$

which can be rewritten as  
following:

$$I = 2I_0 \sin \delta_0 \cos \frac{\pi \Phi}{\Phi_0}$$



# SQUID calibrations

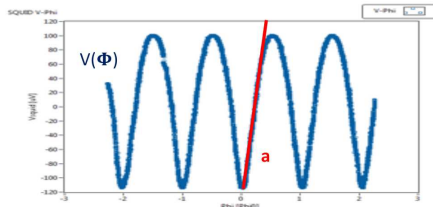
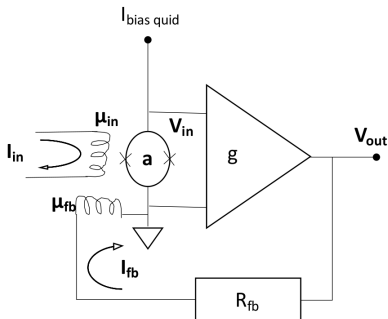
SQUID gain defined as  $V_{out} = G I_{in}$  is given by:

$$G = \frac{ga\mu_{in}}{1 + ga\mu_{fb}/R_{fb}} \rightarrow \lim_{ga \rightarrow \infty} G = R_{fb} \frac{\mu_{in}}{\mu_{fb}},$$

where  $g$  is the op-amp open loop gain,

$$V_{in} = a \left\{ \Phi_{in} - \Phi_{fb} \right\}, \quad \Phi_{in,fb} = \mu_{in,fb} I_{in,fb},$$

$$I_{fb} = \frac{V_{out}}{R_{fb}}, \quad V_{out} = g V_{in}.$$



# $\mu$ calorimeter calibrations

Laser based energy calibration<sup>1</sup>:

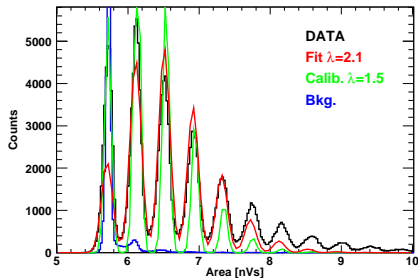
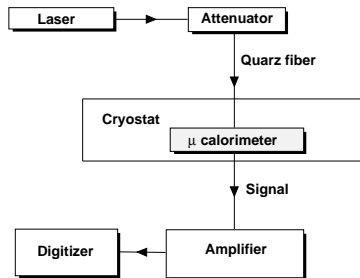
- 1 439 nm (2.824 eV) blue laser,
- 2 quartz optical fiber in cryostat,
- 3 1-7 photons on TES/pulse,
- 4 < 10 kHz pulse rate,
- 5 timing calibration,
- 6 pile-up test.

Convolutated Poissonian deposited energy spectrum:

$$\sum_n e^{-\frac{(E_{dep}/3eV-n)^2}{\sigma_E^2}} \frac{\mu^n}{n!} e^{-\mu}.$$

Test on known energy peaks:  $^{235}\text{mU}$   
and first atomic levels of U.

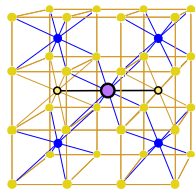
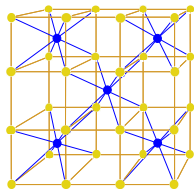
<sup>1</sup> A. Miller et al., *Appl. Phys. Lett.* 83, 791 (2003).



- 1 UV-transparent crystals (bandgaps 12 and 8 eV):  
CaF<sub>2</sub> (Vienna)<sup>1</sup> and LiCaAlF<sub>6</sub> (UCLA)<sup>2</sup>,
- 2 <sup>229</sup>Th<sup>4+</sup> ion replaces Ca,
- 3 high number of oscillators (> 10<sup>15</sup>),
- 4 < 10 photons/s spontaneous emission,
- 5 easy to handle solid state target.

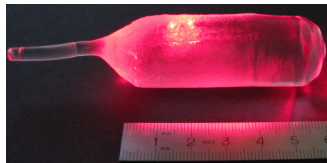
<sup>1</sup>G.Kazakov et al.,*New J.Phys.* 14,083019(2012).

<sup>2</sup>W.Rellergert et al.,*Phys.Rev.Lett.* 104,200802(2010).



- F
- Ca
- F interstitial
- Th

T.Schumm, [www.thorium.at](http://www.thorium.at)  
E.Hudson, [hudsongroup.physics.ucla.edu/229th-nuclear-clock](http://hudsongroup.physics.ucla.edu/229th-nuclear-clock)

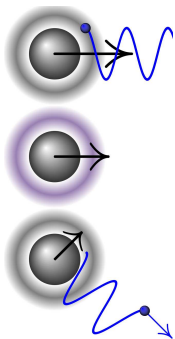


- 1 ion trap for  $^{229}\text{Th}^{3+}$ ,
- 2 laser cooling,
- 3 laser ablation source,
- 4 low number of oscillators ( $< 10^5$ ),
- 5 100 s trapping lifetime,
- 6 measurement of hyperfine structure of  $^{229}\text{Th}^{3+}$  atom.

<sup>1</sup>A.Radnaev et al.,*Phys.Rev.A*86,060501(R)(2012).

<sup>2</sup>N.Huntemann et al.,*Phys.Rev.Lett.* 116,063001(2016).

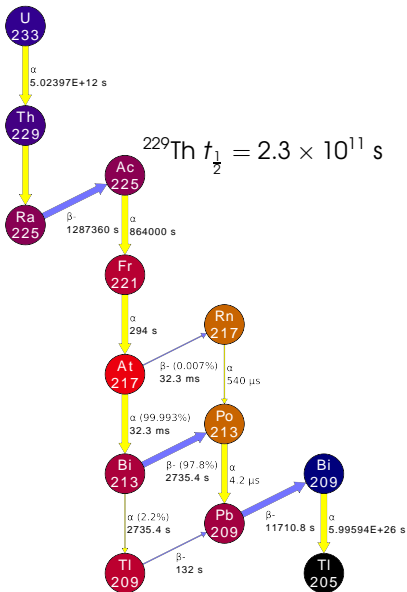
A.Kuzmich, [sites.lsa.umich.edu/kuzmich-lab/](http://sites.lsa.umich.edu/kuzmich-lab/)  
E.Peik, [www.ptb.de/cms/en/ptb/fachabteilungen/abt4/fb-44/ag-443.html](http://www.ptb.de/cms/en/ptb/fachabteilungen/abt4/fb-44/ag-443.html)



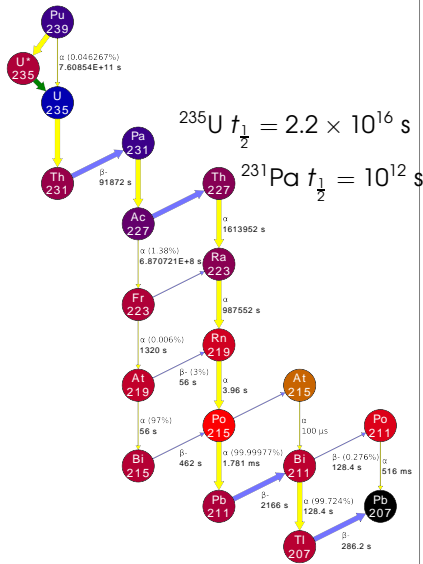


# U233 and Pu239 Decay Chains

ENDF/B-VIII.0 U233 decay path



ENDF/B-VIII.0 Pu239 decay path



# Thorium Decay Chain

