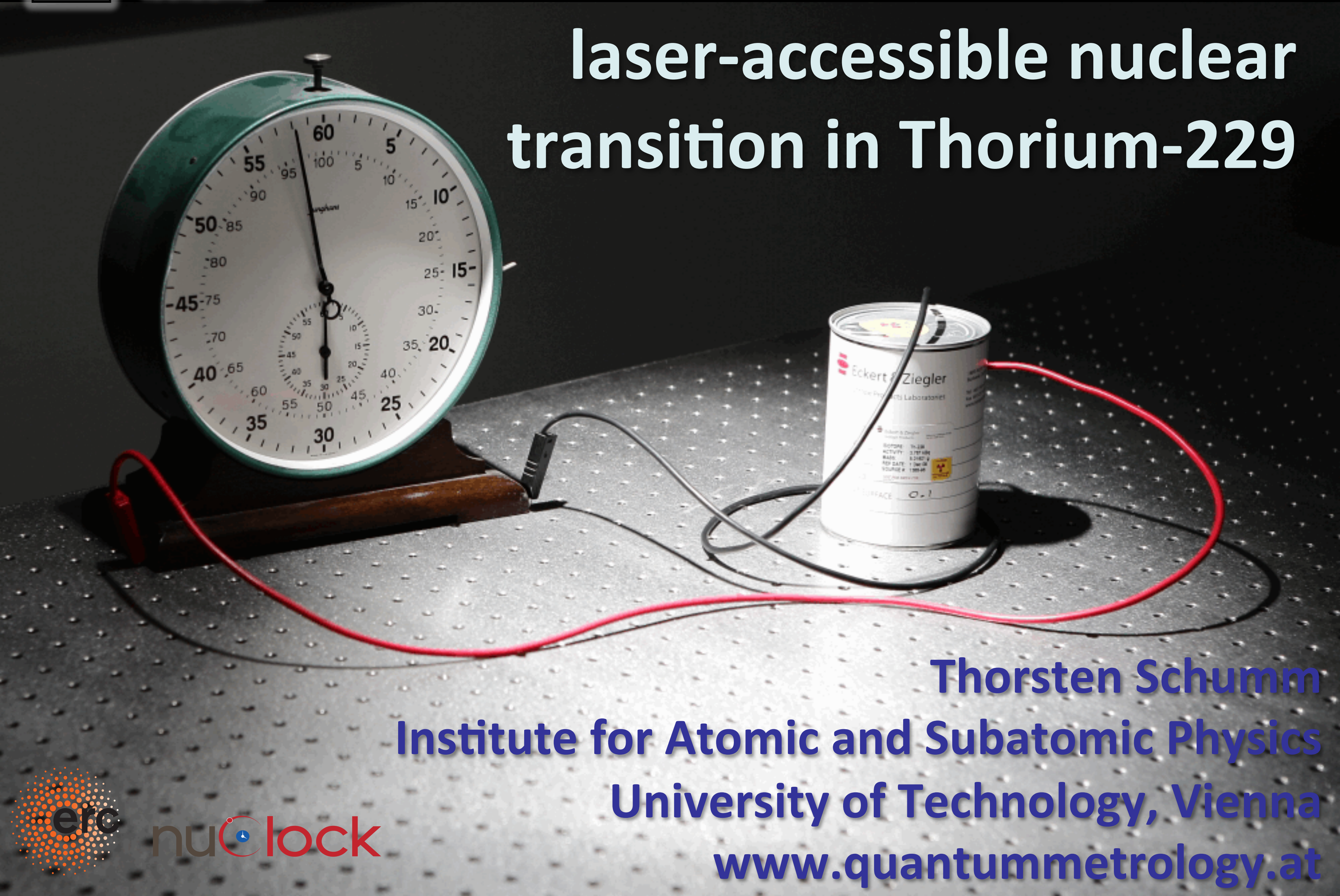
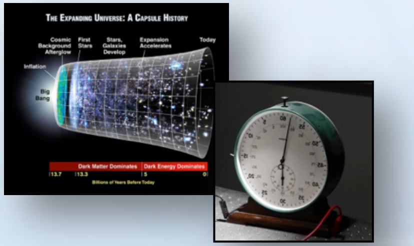
# New results on the laser-accessible nuclear transition in Thorium-229



**Thorsten Schumm**
**Institute for Atomic and Subatomic Physics**
**University of Technology, Vienna**
**www.quantummetrology.at**

# New results on $^{229}$Th
# Outline



## Introduction: nuclear physics with a laser?

- The low-energy nuclear transition in $^{229}$Thorium
- On nuclear clocks and fundamental constants

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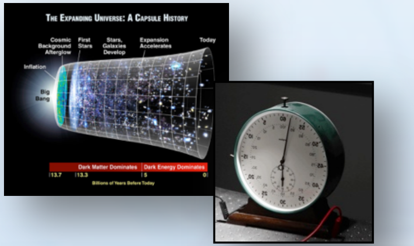

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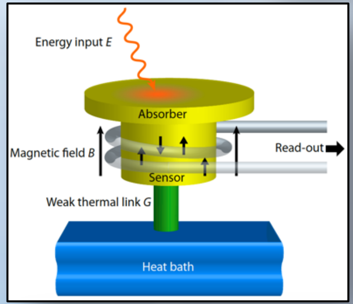
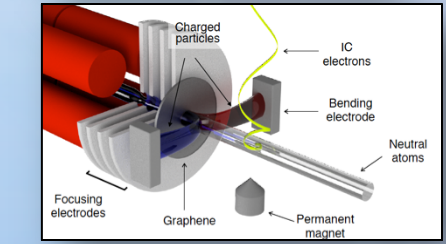


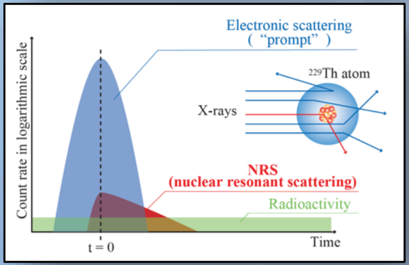
## Experiment 1: Gamma measurements

- A brief history of the $^{229}$Th isomer energy
- The 2007 Beck et al. gamma measurement
- The 2020 measurement: isomer energy + branching ratio



## Experiment 2: IC measurements

- The 2017 LMU measurement: proof of existence
- The 2018 PTB + LMU measurement: optical transitions
- The 2019 measurement: isomer energy

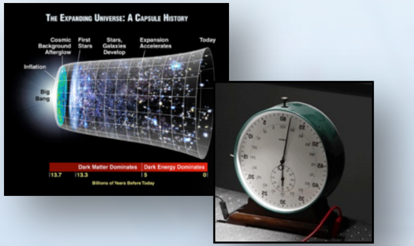


## Experiment 3: X-ray pumping

- The 2019 SPring-8 experiment: isomer pumping

+ isomer energy + branching ratio

# New (?) results on $^{229}$Th
# Outline



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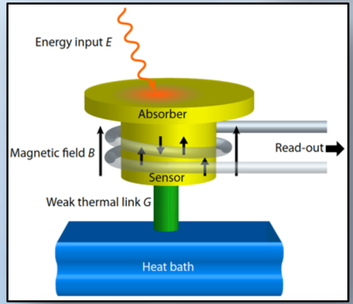

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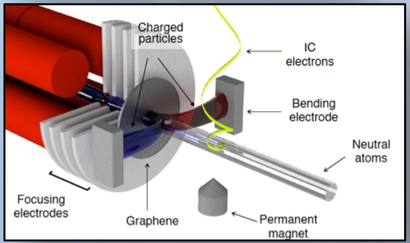
under review with PRL
arXiv:2005.13340



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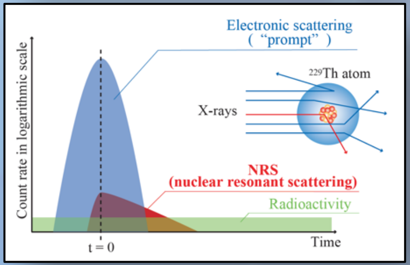
LETTER

https://doi.org/10.1038/s41586-019-1533-4

**Energy of the $^{229}$Th nuclear clock transition**

Benedict Seiferle[1*], Lars von der Wense[1], Pavlo V. Bilous[2], Ines Amersdorffer[1], Christoph Lemell[3], Florian Libisch[3], Simon Stellmer[4], Thorsten Schumm[5], Christoph E. Düllmann[6,7,8], Adriana Pálffy[2] & Peter G. Thirolf[1]



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- The 2019 SPri

+ isomer energy

LETTER

https://doi.org/10.1038/s415

**X-ray pumping of the $^{229}$Th nuclear clock isom**

Takahiko Masuda[1], Akihiro Yoshimi[1], Akira Fujieda[1], Hiroyuki Fujimoto[2], Hiromitsu Haba[3], Hideaki Hara[1], Takahi Hiroyuki Kaino[1], Yoshitaka Kasamatsu[4], Shinji Kitao[5], Kenji Konashi[6], Yuki Miyamoto[1], Koichi Okai[1], Sho Okubo[1] Noboru Sasao[1*], Makoto Seto[5], Thorsten Schumm[7], Yudai Shigekawa[4], Kenta Suzuki[1], Simon Stellmer[7,10], Kenji Ta Satoshi Uetake[1], Makoto Watanabe[6], Tsukasa Watanabe[2], Yuki Yasuda[4], Atsushi Yamaguchi[3], Yoshitaka Yoda[9], Ta Motohiko Yoshimura[1] & Koji Yoshimura[1*]

nature

THE INTERNATIONAL WEEKLY JOURNAL OF SCIENCE

A MATTER OF TIME

*Improved insight into thorium isomer brings nuclear clock closer to reality*

PAGES 202, 238 & 243

NATURE.COM
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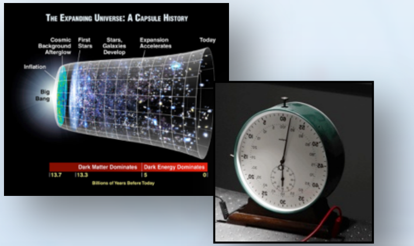
Vol. 573, No. 7773

# New results on $^{229}$Th

## Questions ???

Please ask anytime!

# New results on $^{229}$Th
# Outline



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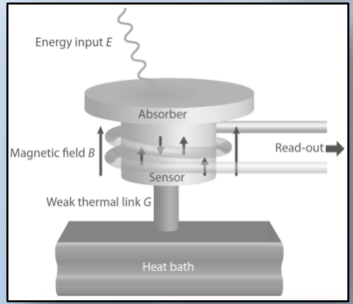

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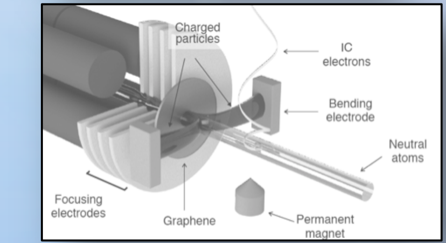

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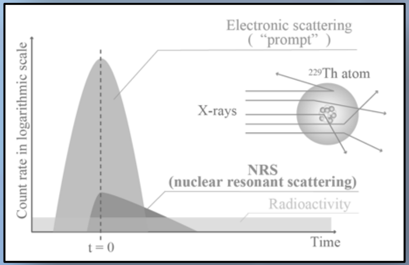

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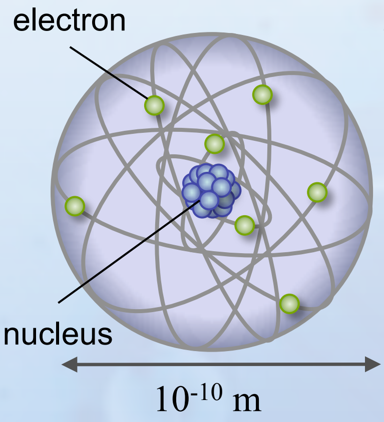
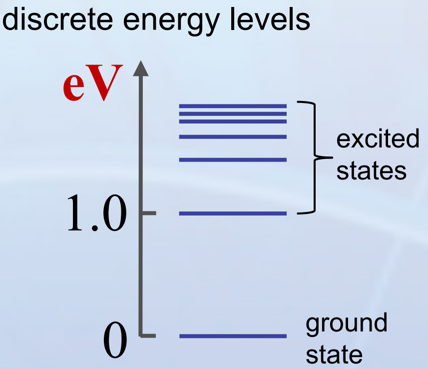
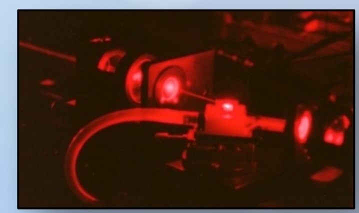
# Nuclear physics with a laser?
# The gap between atomic and nuclear physics

## Atomic physics







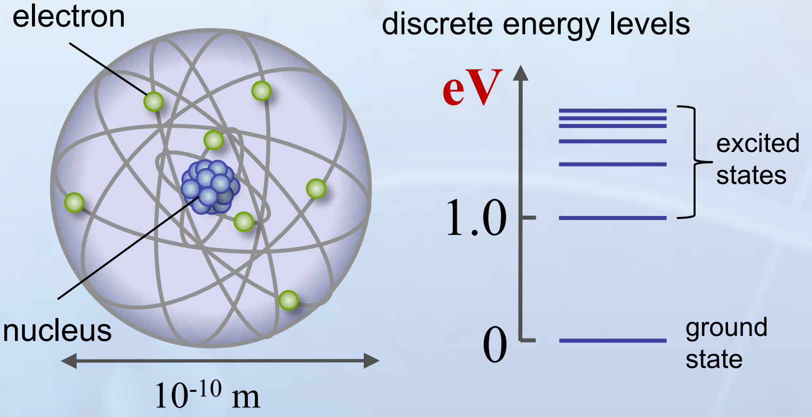
main tool
of study:
**laser**
**spectroscopy**

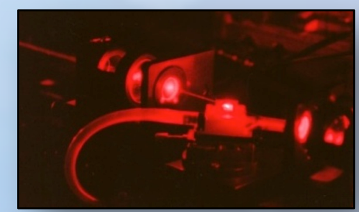
## Atomic spectroscopy for metrology

- provides a frequency standard
- 1s ≡ 9.192.631.770 oscillations in Cs
- crucial for fundamental research
- applications: GPS, communication
- miniaturization possible

# Nuclear physics with a laser?
# The gap between atomic and nuclear physics

## Atomic physics





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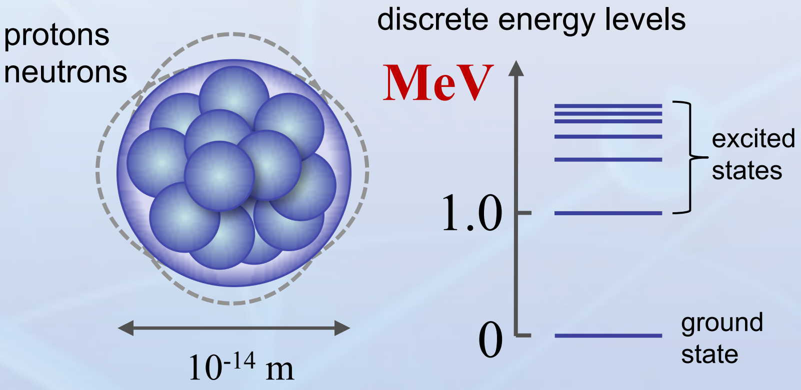
## Nuclear physics



main tool
of study:
**particle**
**accelerators**

## Atomic spectroscopy for metrology

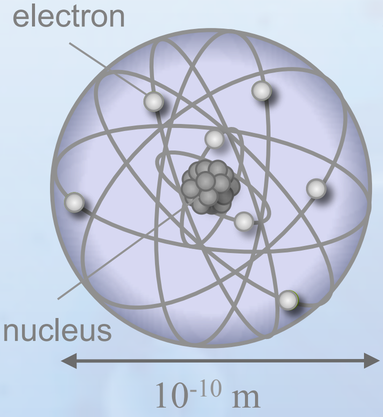
- provides a frequency standard
- 1s ≡ 9.192.631.770 oscillations in Cs
- crucial for fundamental research
- applications: GPS, communication
- miniaturization possible

## Nuclear spectroscopy for metrology

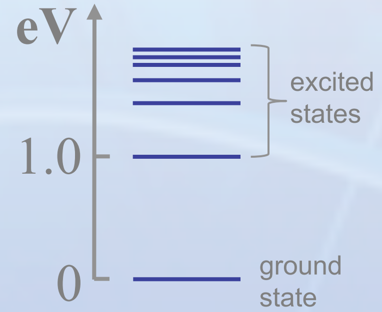
- no frequency standard (although suited!)
- used in fundamental research
- (Mössbauer spectroscopy, dating)
- no direct metrology applications
- no miniaturization

# Nuclear physics with a laser?
# The gap between atomic and nuclear physics

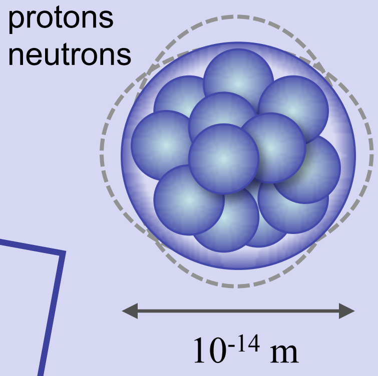
## Atomic physics



discrete energy levels



## Nuclear physics



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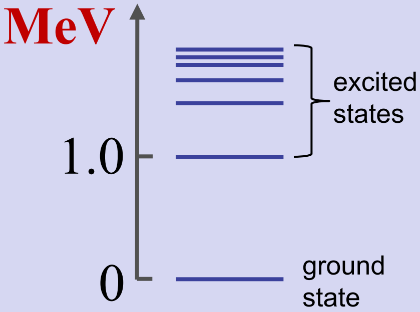
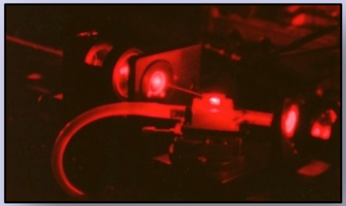




main tool
of study:
**laser**
**spectroscopy**

„nuclear physics with a laser“

main tool
of study:
**particle**
**accelerators**

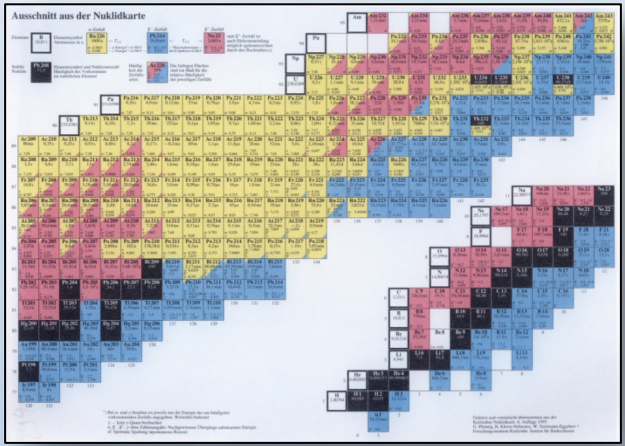
### Atomic spectroscopy for metrology

- provides a frequency standard
- 1s ≡ 9.192.631.770 oscillations in Cs
- crucial for fundamental research
- applications: GPS, communication
- miniaturization possible

### Nuclear spectroscopy for metrology

- **NEW frequency standards ?**
- **new fundamental research possible !**
- (Mössbauer spectroscopy, dating)
- **miniaturization possible**
- **applications ahaid?**

# Nuclear physics with a laser?
# The low-energy nuclear transition in $^{229}$Th



## Isotopes with low-lying excited states:

| | | | | |
|---|---|---|---|---|
| Tc-99 | 2150 | eV | | |
| Hg-201 | 1561 | eV | | |
| W-183 | 544 | eV | | |
| U-235 | 76 | eV | ≈ | 17 nm |
| **Th-229** | **~8** | **eV** | **≈** | **150 nm** |

# Nuclear physics with a laser?
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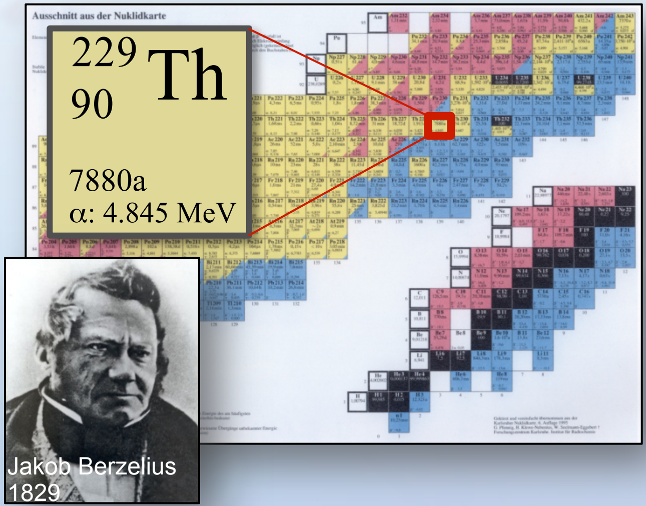

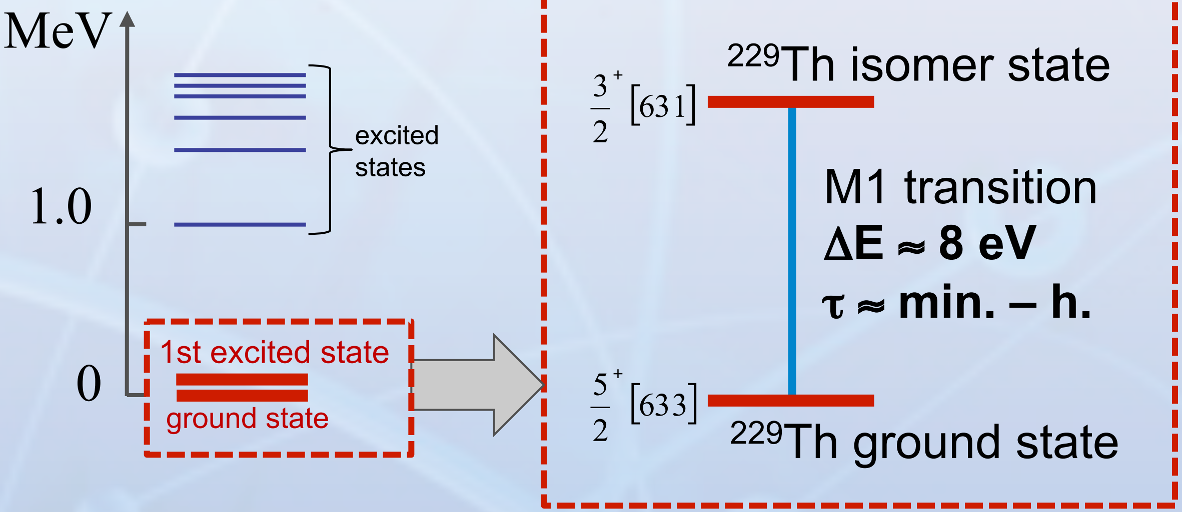

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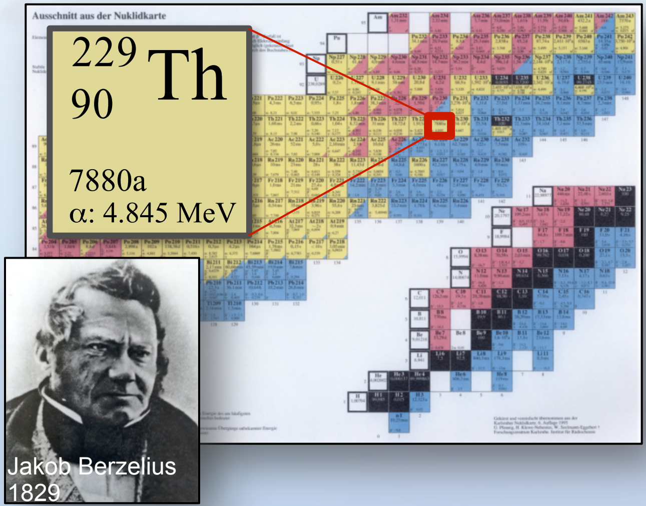
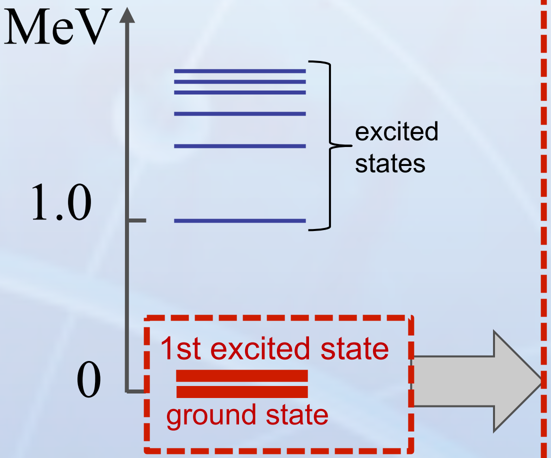



MeV

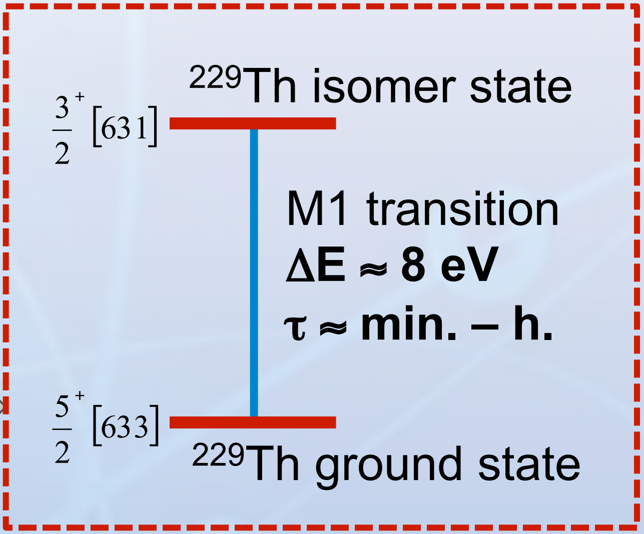
1.0

0

excited states

1st excited state

ground state


$^{229}$Th isomer state

$\frac{3}{2}^{+}$ [631]

M1 transition

**ΔE ≈ 8 eV**

**τ ≈ min. – h.**

$\frac{5}{2}^{+}$ [633]

$^{229}$Th ground state

## $^{229}$Thorium facts :

- ☹ radio isotope : half life 7880 years, **very rare, totally artificial substance**
- ☹ ionization energies: 6.1 eV, 11.5 eV, 20 eV → **need to work with the ion**
- ☹ low scattering cross section (estimated $\sigma \approx 10^{-21}$ cm$^2$) (a single Rb Atom has $10^{-9}$ cm$^2$)
- ☹ no easy (tunable) excitation source at 150 nm (VUV)

# Nuclear physics with a laser?
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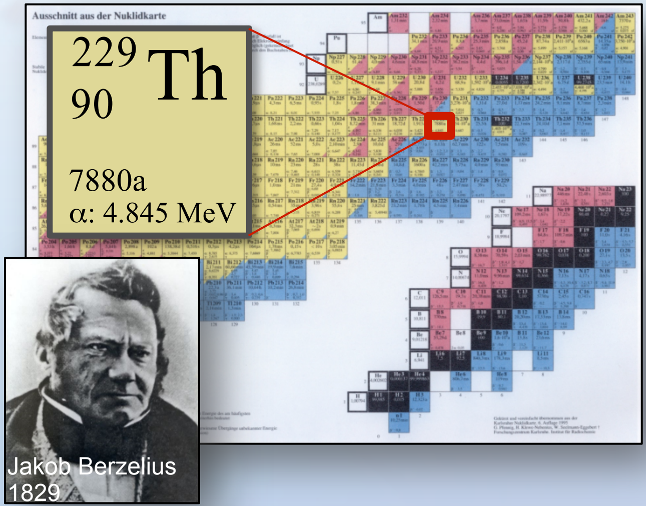
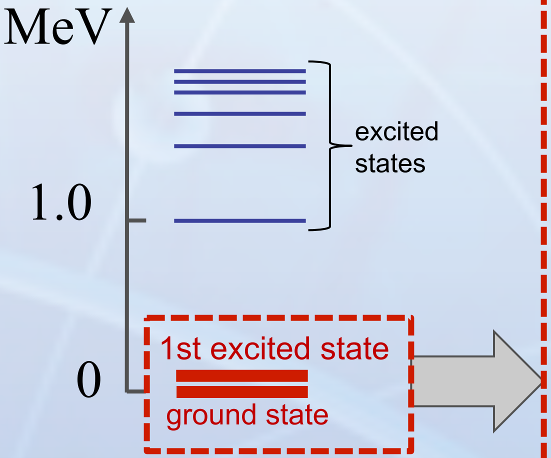


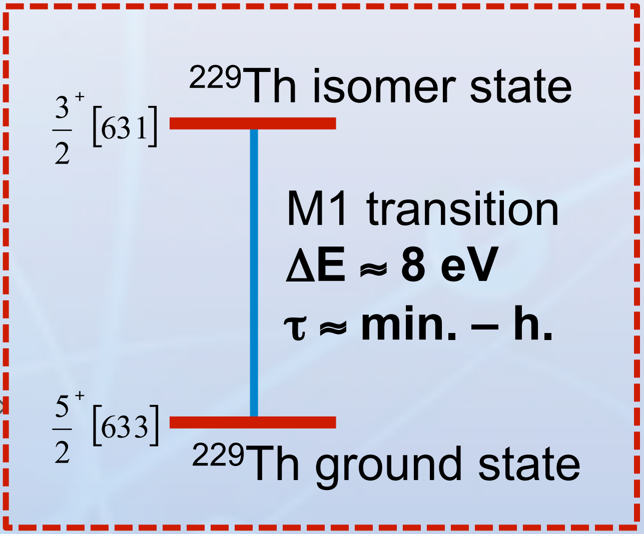
Jakob Berzelius
1829



excited states

1st excited state

ground state



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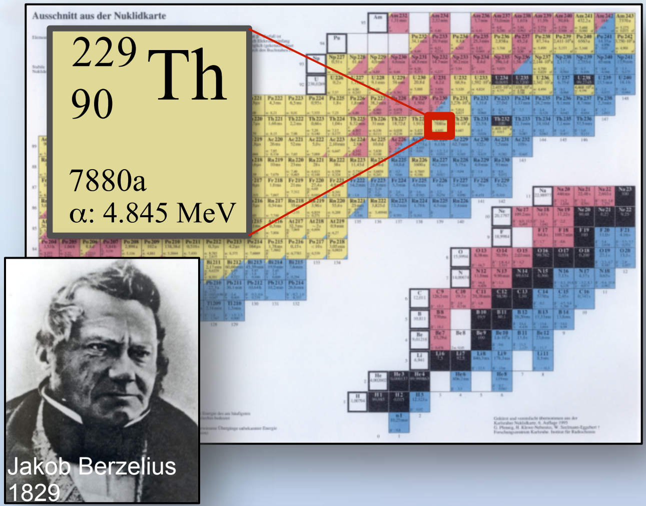
- ☺ robust nuclear transition
- ☺ can be embedded in a crystal

} **a solid-state „nuclear“ clock ?**

# Nuclear physics with a laser?
# The low-energy nuclear transition in $^{229}$Th



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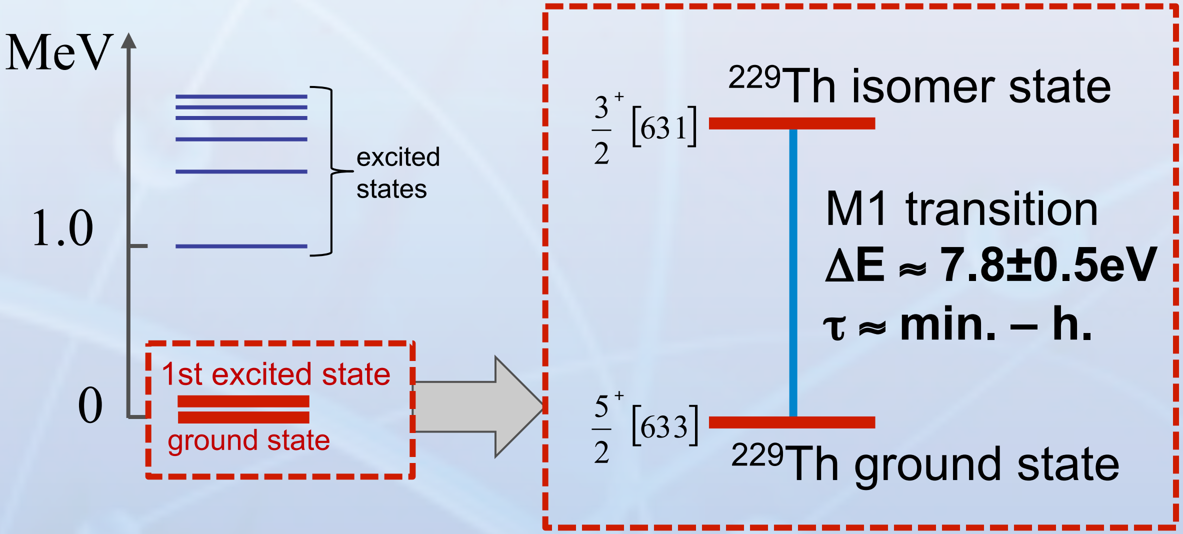

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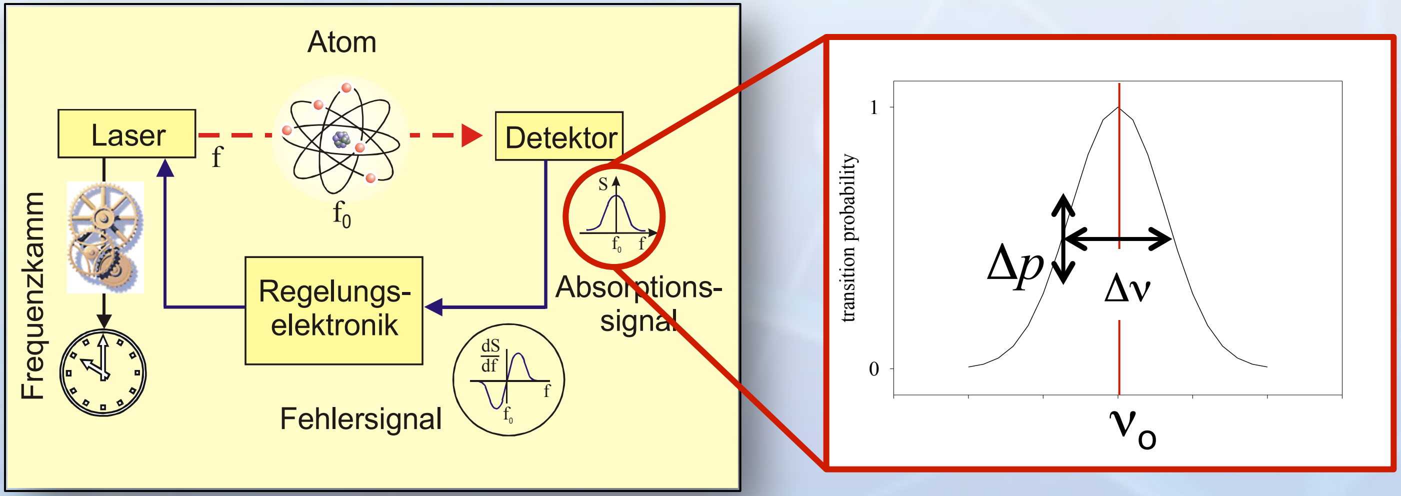
- ☺ narrow transition for a frequency standard
- ☺ robust nuclear transition
- ☺ can be embedded in a crystal
- ☺ very high sensitivity to the fine structure constant

**a solid-state „nuclear" clock ?**

**search for drifts in fundamental constants?**

# Nuclear physics with a laser?
# Why a “nuclear clock”? – figure of merit



Ideal atomic clock: unperturbed atomic transition frequency $\boldsymbol{\nu}_o$ (at $v = 0$, $T = 0$)

## what makes a good „clock transition“?

**$\Delta\nu$** : linewidth, ideally Fourier limited ~$1/T_{exc.}$, requires stable atomic levels

**$\Delta p$** : measurement noise, ideally projection noise/Fourier limited

**Quality factor** of a clock transition: $\boxed{Q = \frac{\nu_0}{\Delta\nu}}$

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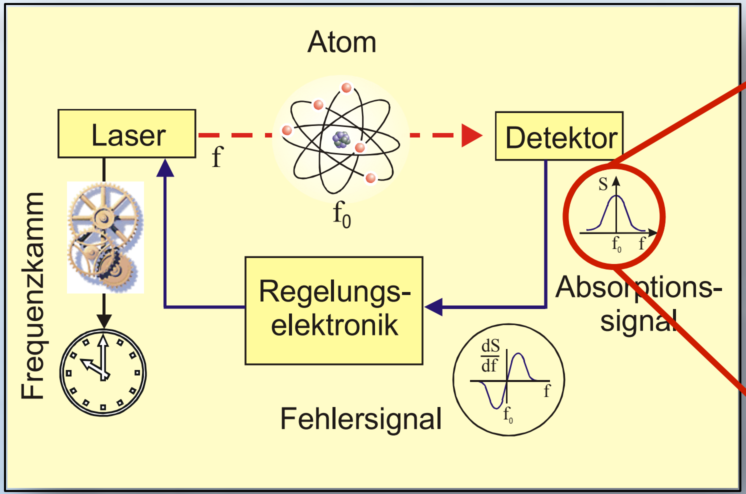

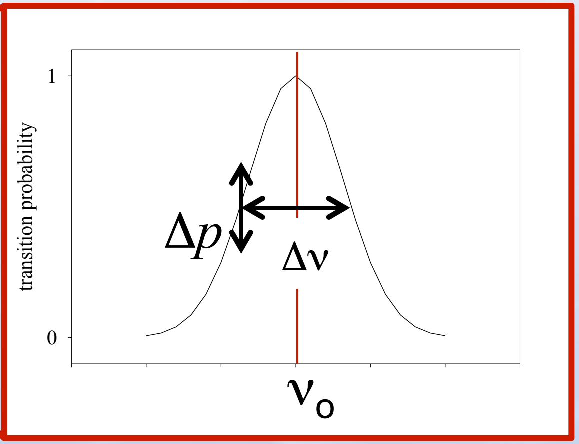

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**Quality factor** of a clock transition: $\boxed{Q = \frac{\nu_0}{\Delta\nu}}$

**Standard quantum limit**: $\boxed{\Delta\nu \approx 1/\sqrt{N T_{\text{int}} \tau_{av}}}$

for $T_{exc.} >> T_{\text{int}}, \tau_{av}$

to build a good clock...

- use high transition frequency $\nu_0$
- use narrow transition (small $\Delta\nu$)
- have many oscillators (large $N$)
- have long interrogation time $T_{\text{int}}$

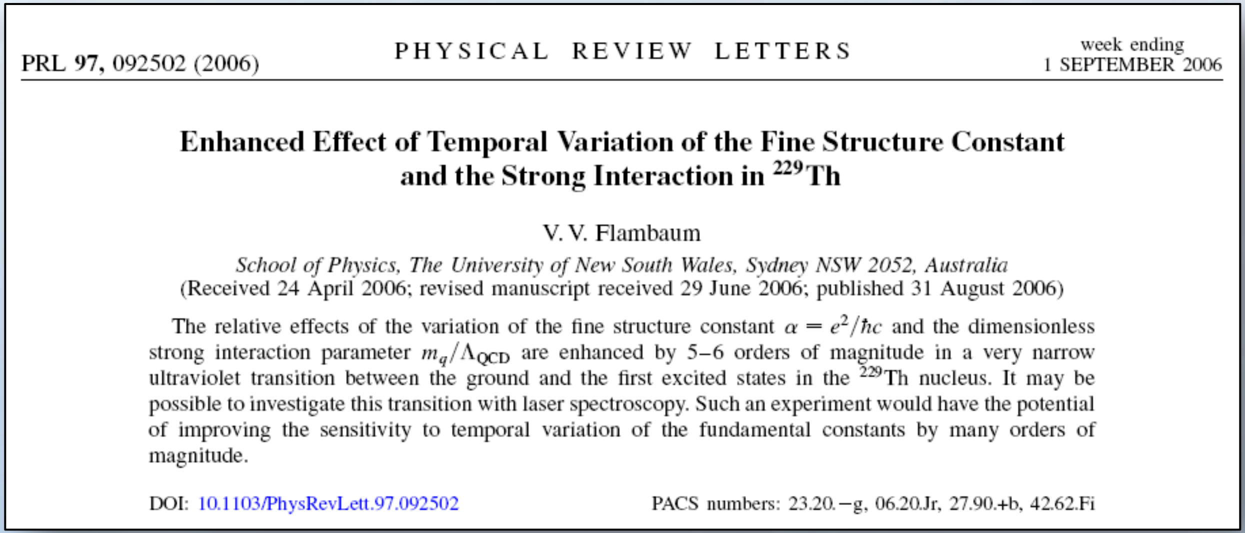
**ideal: solid-state nuclear**

**Figure of merit*:**

$\boxed{F = Q\sqrt{N}}$

assuming $\Delta\nu \approx 1 / (T_{\text{int}}\tau_{av})^{-1/2}$ independent of $N$

# Nuclear physics with a laser?
# What about “fundamental constants”?



PRL 97, 092502 (2006)  PHYSICAL REVIEW LETTERS  week ending 1 SEPTEMBER 2006

**Enhanced Effect of Temporal Variation of the Fine Structure Constant and the Strong Interaction in $^{229}$Th**

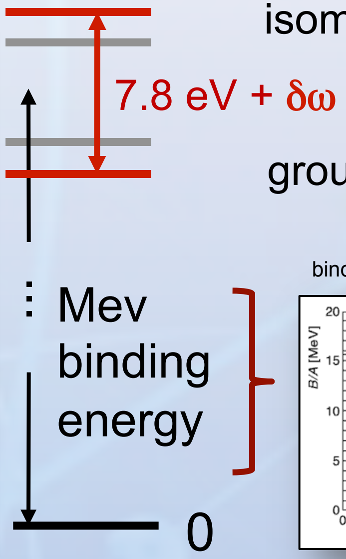
V. V. Flambaum

*School of Physics, The University of New South Wales, Sydney NSW 2052, Australia*

(Received 24 April 2006; revised manuscript received 29 June 2006; published 31 August 2006)

The relative effects of the variation of the fine structure constant $\alpha = e^2/\hbar c$ and the dimensionless strong interaction parameter $m_q/\Lambda_{QCD}$ are enhanced by 5–6 orders of magnitude in a very narrow ultraviolet transition between the ground and the first excited states in the $^{229}$Th nucleus. It may be possible to investigate this transition with laser spectroscopy. Such an experiment would have the potential of improving the sensitivity to temporal variation of the fundamental constants by many orders of magnitude.

DOI: 10.1103/PhysRevLett.97.092502  PACS numbers: 23.20.−g, 06.20.Jr, 27.90.+b, 42.62.Fi



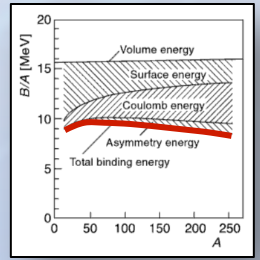
isomeric state

7.8 eV + δω

ground state

Mev binding energy

0



binding energy/nucleon

$$\boxed{\frac{\delta\omega}{\omega} \approx 10^{5}\left(4\frac{\delta\alpha}{\alpha}+\frac{\delta X_q}{X_q}-10\frac{\delta X_s}{X_s}\right)}$$

with $X_q = \dfrac{m_q}{\Lambda_{QCD}}$ $X_s = \dfrac{m_s}{\Lambda_{QCD}}$

$m_q$, $m_s$ light/strange quark mass (5/120 MeV)
$\Lambda_{QCD}$ quantum chromodynamic scale

$10^3$-$10^5$ enhancement in sensitivity!
(exact value under vivid discussion!)

comparing the $^{229}$Th nuclear transition frequency to standard atomic clocks may allow to look for variations of $\alpha$ at $10^{-18}$ - $10^{-20}$ per year

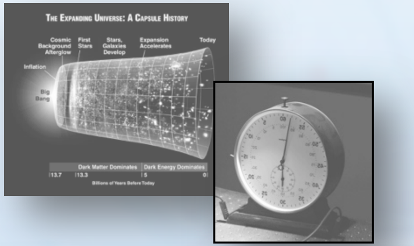
**fundamental physics in a tabletop experiment?**

# New results on $^{229}$Th

## Questions ???

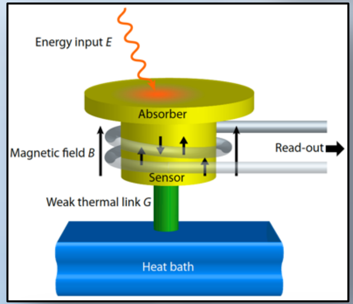
Maybe now?

# New results on $^{229}$Th
# Outline



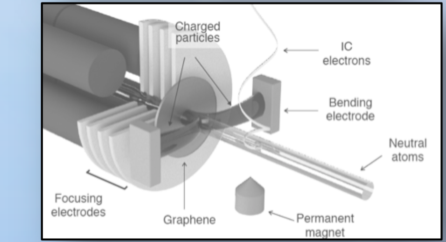
## Introduction: nuclear physics with a laser?

- The low-energy nuclear transition in $^{229}$Thorium
- On nuclear clocks and fundamental constants



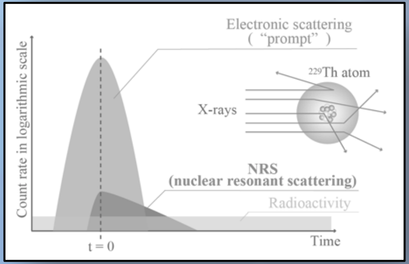
## Experiment 1: Gamma measurements

- A brief history of the $^{229}$Th isomer energy
- The 2007 Beck et al. gamma measurement
- The 2020 measurement: isomer energy + branching ratio



## Experiment 2: IC measurements

- The 2017 LMU measurement: proof of existence
- The 2018 PTB + LMU measurement: optical transitions
- The 2019 measurement: isomer energy



## Experiment 3: X-ray pumping

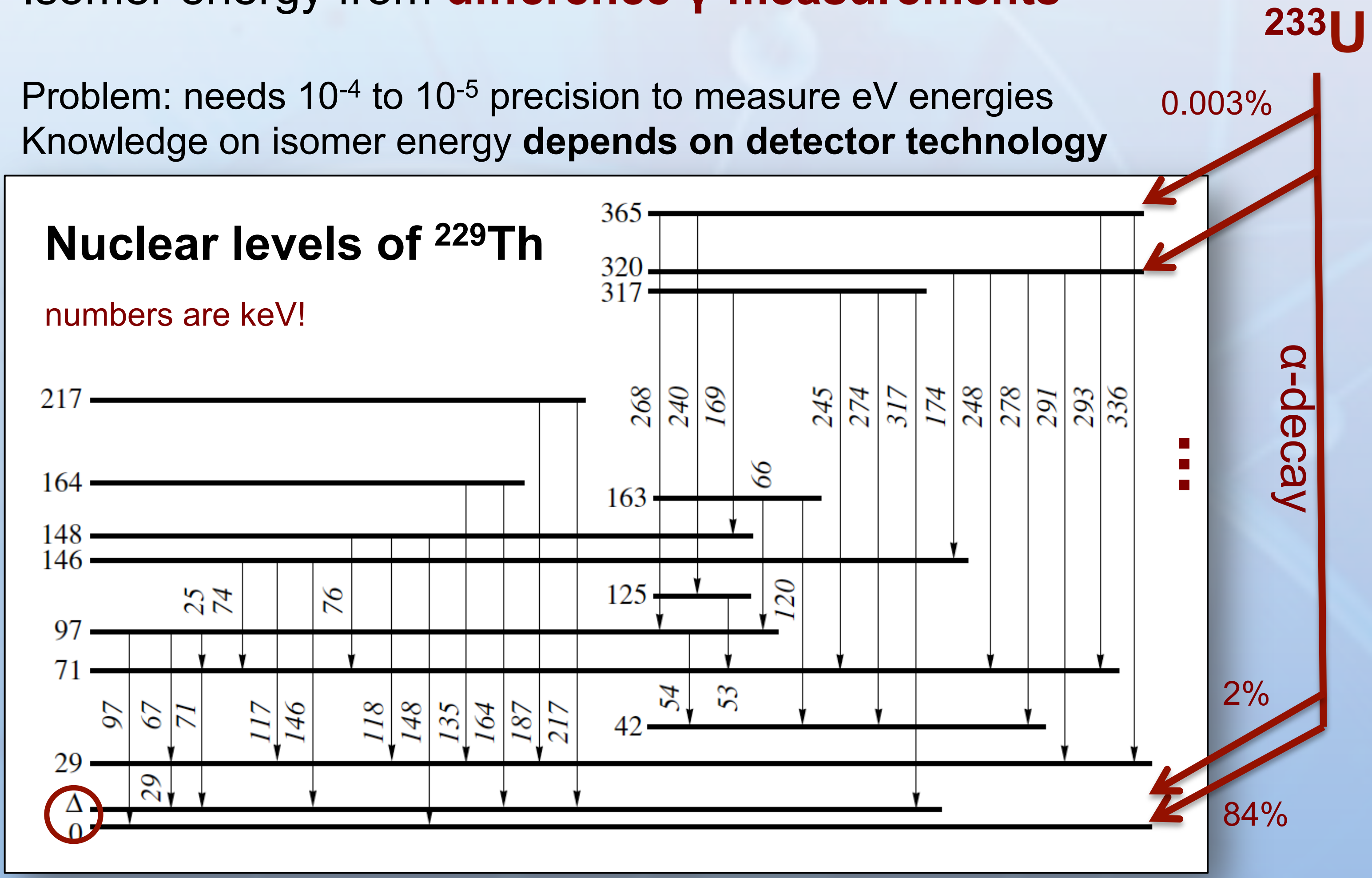
- The 2019 SPring-8 experiment: isomer pumping

+ isomer energy + branching ratio

# Gamma measurements
# A brief history of the $^{229}$Th isomer energy

## Isomer energy from **difference γ-measurements**

Problem: needs $10^{-4}$ to $10^{-5}$ precision to measure eV energies
Knowledge on isomer energy **depends on detector technology**



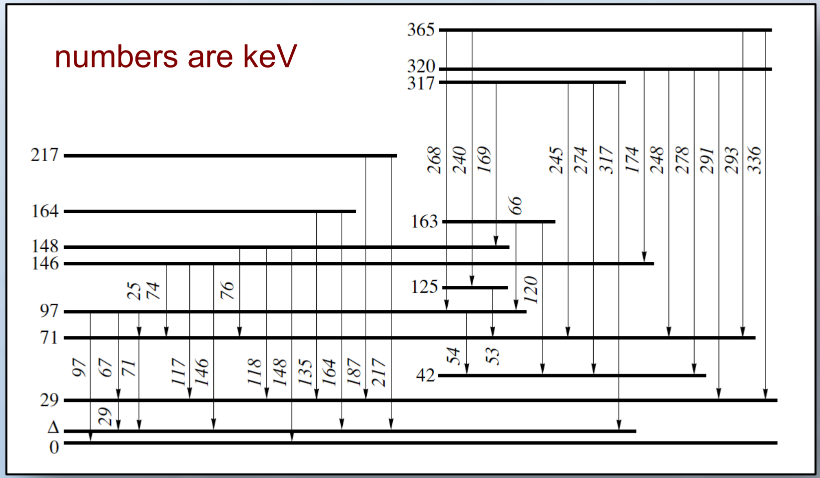
**Nuclear levels of $^{229}$Th**

numbers are keV!

S. L. Sakharov, Physics of at. Nuclei 73, 1-8 (2010)

# Gamma measurements
# A brief history of the $^{229}$Th isomer energy

The $^{229}$Th isomer energy until 2007:



S. L. Sakharov, Physics of at. Nuclei 73, 1-8 (2010)

30 years of improving detectors...

| ΔE [eV] | year | method |
|---|---|---|
| < 100 | 1976 | γ-spectro |
| 1 (4) | 1990 | γ-spectro |
| < 5 | 1990 | d-scatt. |
| 3.5 (1.0) | 1994 | γ-spectro |
| 3.4 (1.8) | 2003 | γ-spectro |

R. G. Helmer and C.W. Reich, Phys. Rev. C **49**, 1845 (1994)
V. Barci, G. Ardisson, G. Barci-Funel, et al., Phys. Rev. C **68**, 034329 (2003)

**lots of (unsuccessfull) experiments performed based on this value...**

# Gamma measurements
# The 2007 measurement by Beck et al.

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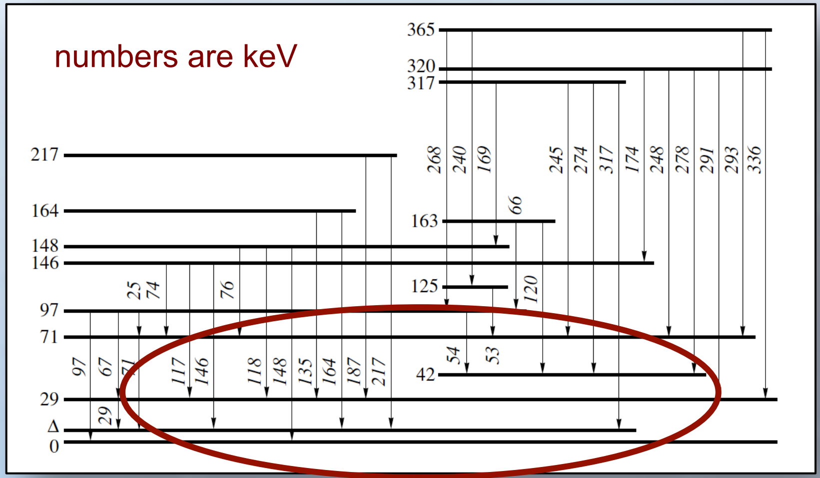

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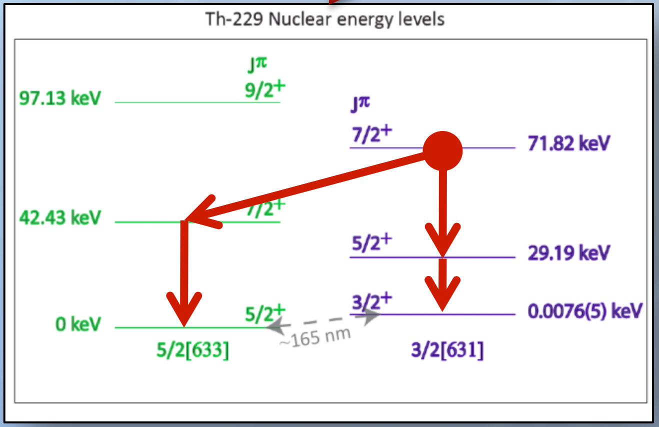
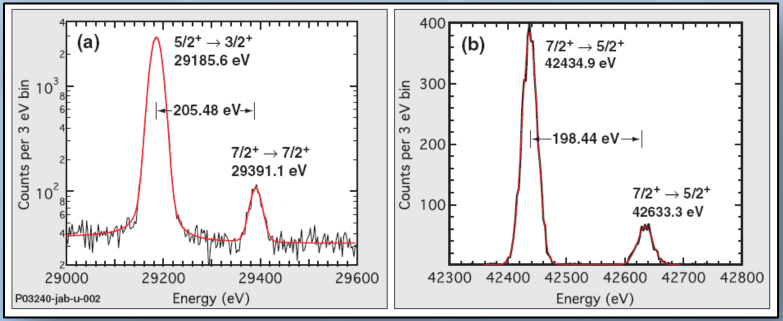


B. R. Beck et.al. Phys. Rev. Lett 98, 142501 (2007)

## Using a NASA x-ray microcalorimeter...



**Gives the „latest“ value 7.8 (±0.5) eV**

# Gamma measurements
# The 2007 measurement by Beck et al.

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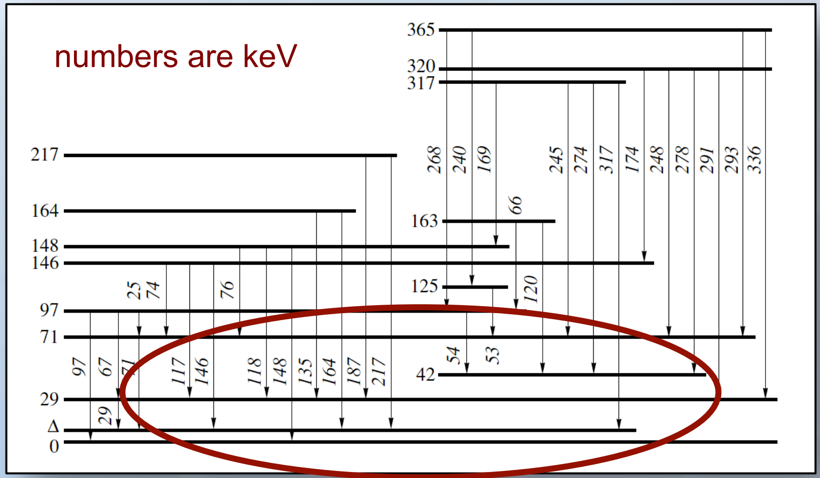

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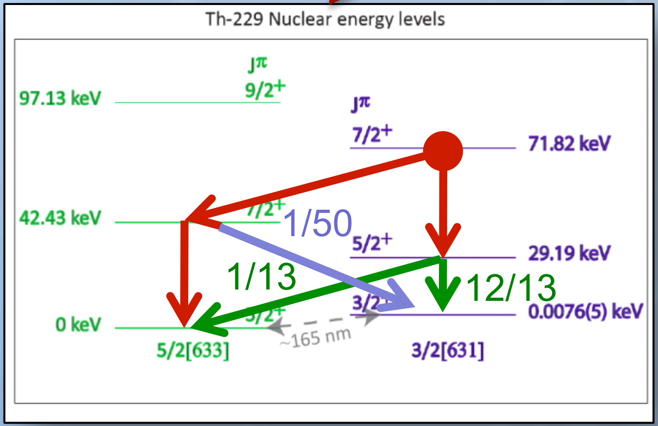

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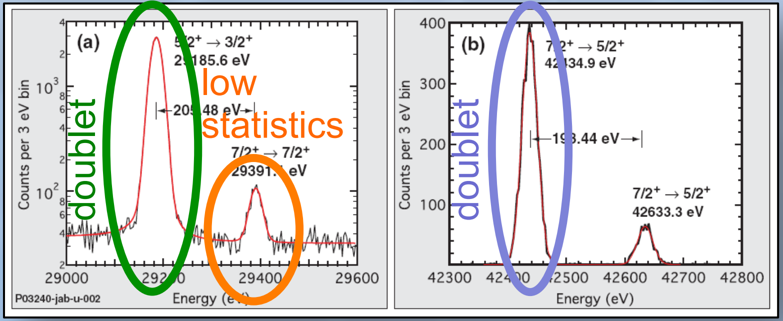


**Gives the „latest“ value 7.8 (±0.5) eV**

# Gamma measurements
# The 2007 measurement by Beck et al.

**What we learned from 2007 Beck et al.:**

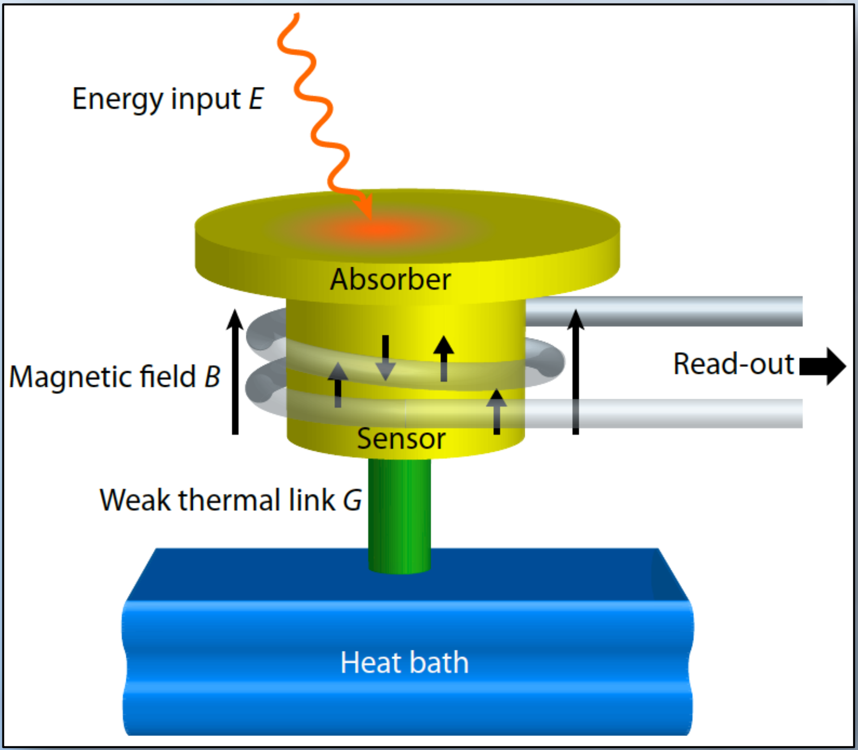
- transition shifted from visible into VUV (killing MANY previous attempts)
- still within the range of lasers (but MUCH harder!)
- now, excitation energy ~ ionization energy → electronic states matter
- indirect measurement, NO proof of existence
- further improving the detectors would directly reduce the error...

  (error limited by statisitics in the 29.39 keV line + abs. energy callibration + branching ratios)

# Gamma measurements
# The 2020 measurement

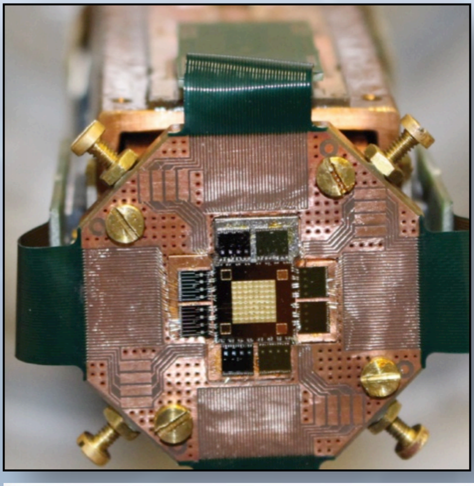
Building the worlds best gamma spectrometer (for this energy range):

Magnetic microcalorimeter

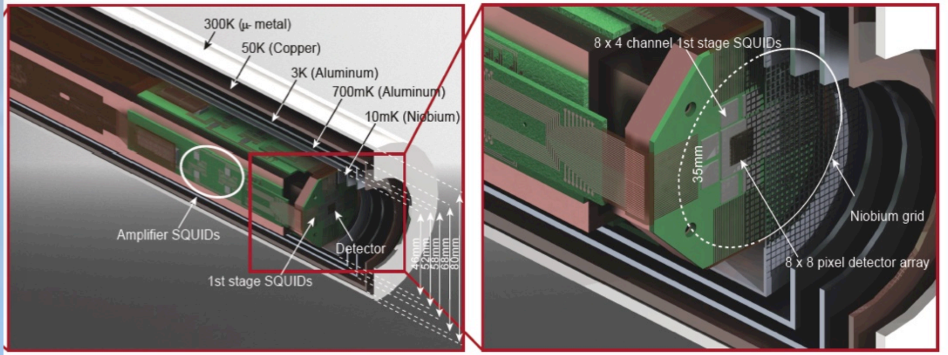


specifically designed for Th-229 optimal performance @30 keV

maXs-30 8x8 Array



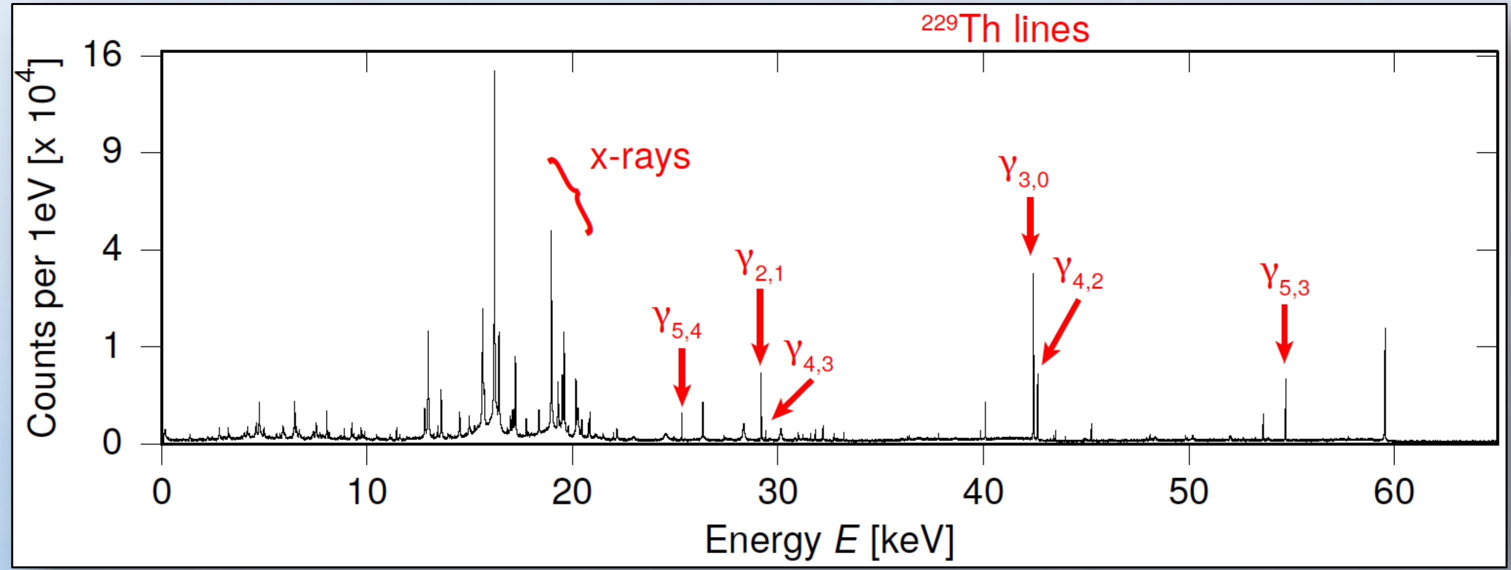
- 32 channels, 64 pixels
- 16 mm$^2$ detection area
- 4 temp. readouts
- expected energy resolution: 10 eV
- 20 weeks data collection



collaboration Univ. Heidelberg Group Enss, Fleischmann

# Gamma measurements
# The 2020 measurement

## Energy callibration of the magnetic microcalorimeter: $^{233}$U + $^{241}$Am



...identify the lines from literature...

# Gamma measurements
# The 2020 measurement

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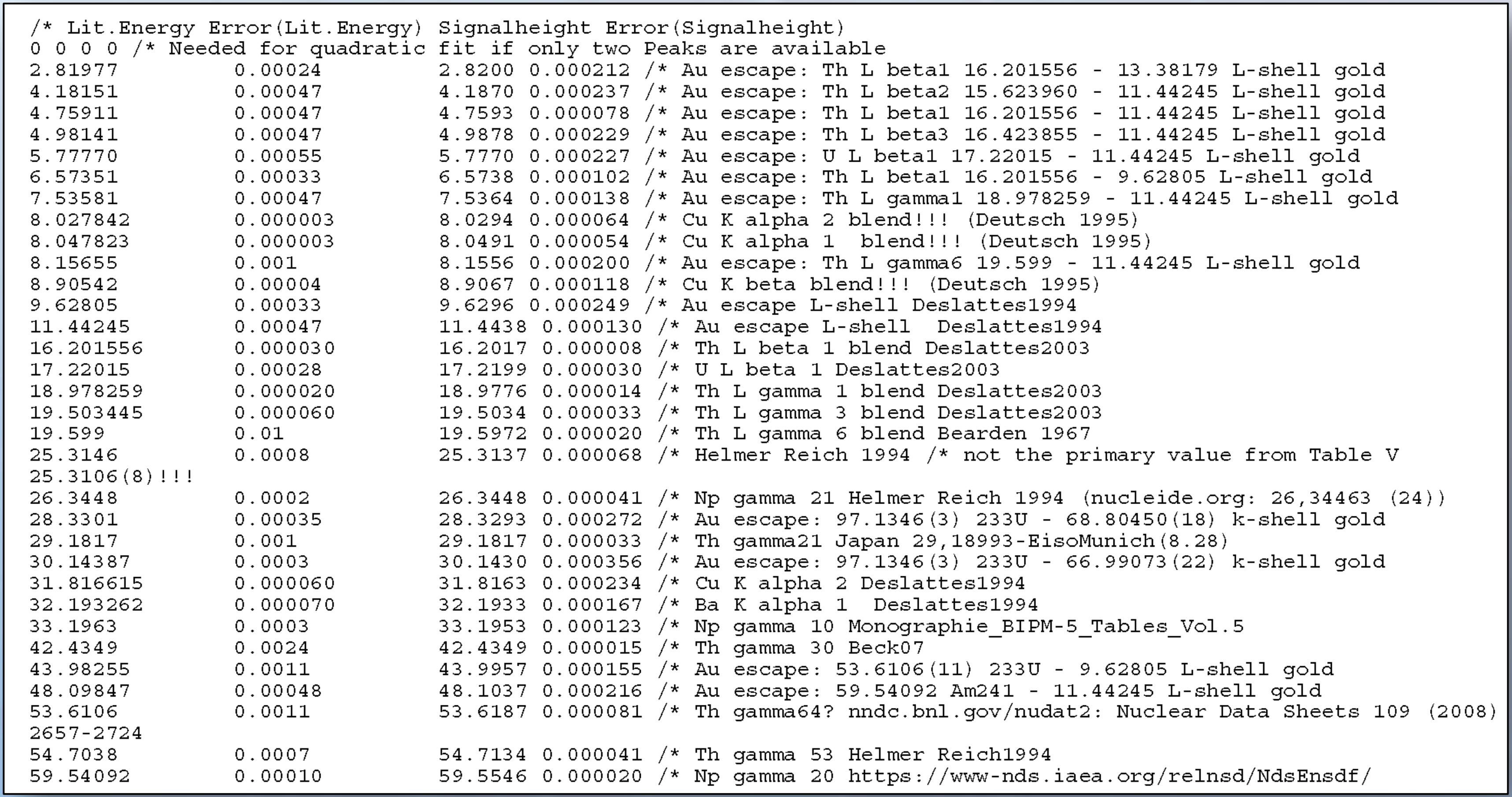
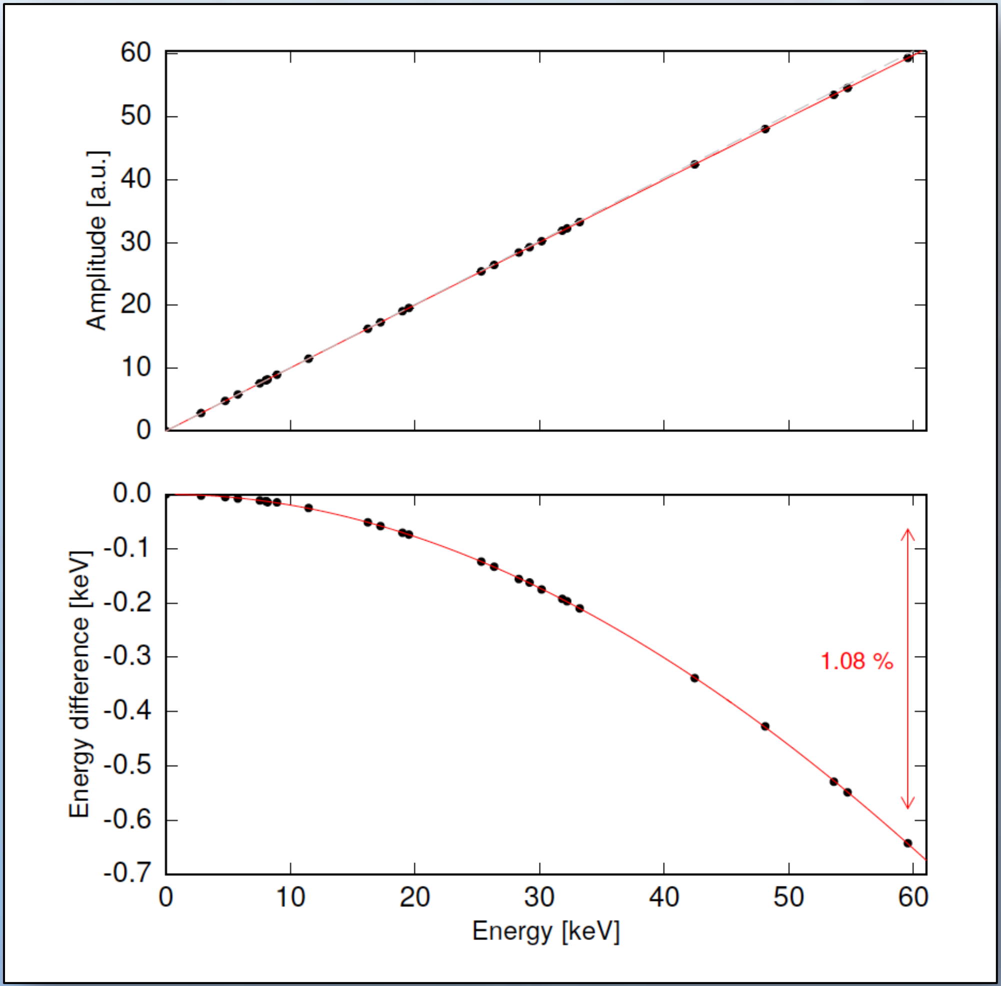


```
/* Lit.Energy Error(Lit.Energy) Signalheight Error(Signalheight)
0 0 0 0 /* Needed for quadratic fit if only two Peaks are available
2.81977         0.00024         2.8200 0.000212 /* Au escape: Th L beta1 16.201556 - 13.38179 L-shell gold
4.18151         0.00047         4.1870 0.000237 /* Au escape: Th L beta2 15.623960 - 11.44245 L-shell gold
4.75911         0.00047         4.7593 0.000078 /* Au escape: Th L beta1 16.201556 - 11.44245 L-shell gold
4.98141         0.00047         4.9878 0.000229 /* Au escape: Th L beta3 16.423855 - 11.44245 L-shell gold
5.77770         0.00055         5.7770 0.000227 /* Au escape: U L beta1 17.22015 - 11.44245 L-shell gold
6.57351         0.00033         6.5738 0.000102 /* Au escape: Th L beta1 16.201556 - 9.62805 L-shell gold
7.53581         0.00047         7.5364 0.000138 /* Au escape: Th L gamma1 18.978259 - 11.44245 L-shell gold
8.027842        0.000003        8.0294 0.000064 /* Cu K alpha 2 blend!!! (Deutsch 1995)
8.047823        0.000003        8.0491 0.000054 /* Cu K alpha 1  blend!!! (Deutsch 1995)
8.15655         0.001           8.1556 0.000200 /* Au escape: Th L gamma6 19.599 - 11.44245 L-shell gold
8.90542         0.00004         8.9067 0.000118 /* Cu K beta blend!!! (Deutsch 1995)
9.62805         0.00033         9.6296 0.000249 /* Au escape L-shell Deslattes1994
11.44245        0.00047         11.4438 0.000130 /* Au escape L-shell  Deslattes1994
16.201556       0.000030        16.2017 0.000008 /* Th L beta 1 blend Deslattes2003
17.22015        0.00028         17.2199 0.000030 /* U L beta 1 Deslattes2003
18.978259       0.000020        18.9776 0.000014 /* Th L gamma 1 blend Deslattes2003
19.503445       0.000060        19.5034 0.000033 /* Th L gamma 3 blend Deslattes2003
19.599          0.01            19.5972 0.000020 /* Th L gamma 6 blend Bearden 1967
25.3146         0.0008          25.3137 0.000068 /* Helmer Reich 1994 /* not the primary value from Table V
25.3106(8)!!!
26.3448         0.0002          26.3448 0.000041 /* Np gamma 21 Helmer Reich 1994 (nucleide.org: 26,34463 (24))
28.3301         0.00035         28.3293 0.000272 /* Au escape: 97.1346(3) 233U - 68.80450(18) k-shell gold
29.1817         0.001           29.1817 0.000033 /* Th gamma21 Japan 29,18993-EisoMunich(8.28)
30.14387        0.0003          30.1430 0.000356 /* Au escape: 97.1346(3) 233U - 66.99073(22) k-shell gold
31.816615       0.000060        31.8163 0.000234 /* Cu K alpha 2 Deslattes1994
32.193262       0.000070        32.1933 0.000167 /* Ba K alpha 1  Deslattes1994
33.1963         0.0003          33.1953 0.000123 /* Np gamma 10 Monographie_BIPM-5_Tables_Vol.5
42.4349         0.0024          42.4349 0.000015 /* Th gamma 30 Beck07
43.98255        0.0011          43.9957 0.000155 /* Au escape: 53.6106(11) 233U - 9.62805 L-shell gold
48.09847        0.00048         48.1037 0.000216 /* Au escape: 59.54092 Am241 - 11.44245 L-shell gold
53.6106         0.0011          53.6187 0.000081 /* Th gamma64? nndc.bnl.gov/nudat2: Nuclear Data Sheets 109 (2008)
2657-2724
54.7038         0.0007          54.7134 0.000041 /* Th gamma 53 Helmer Reich1994
59.54092        0.00010         59.5546 0.000020 /* Np gamma 20 https://www-nds.iaea.org/relnsd/NdsEnsdf/
```

# Gamma measurements
# The 2020 HD measurement

## Energy calibration of the magnetic microcalorimeter: $^{233}$U + $^{241}$Am



the magnetic microcalorimeter shows excellent linearity

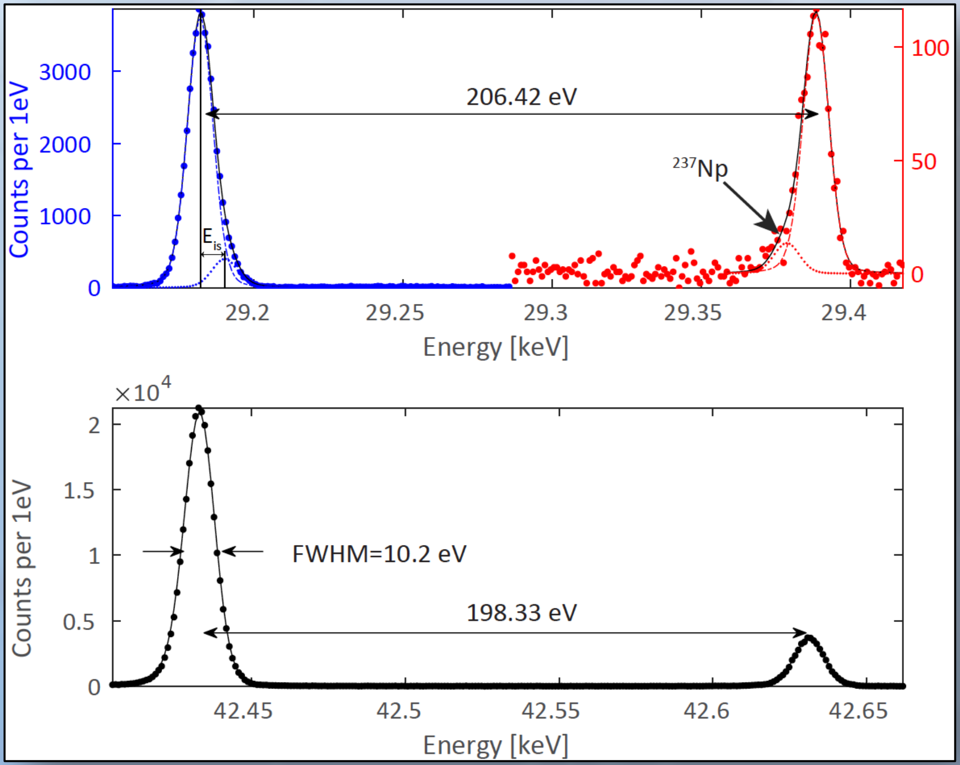
(in strong contrast to the „ohmic“ microcalorimeter used by Beck et al.)

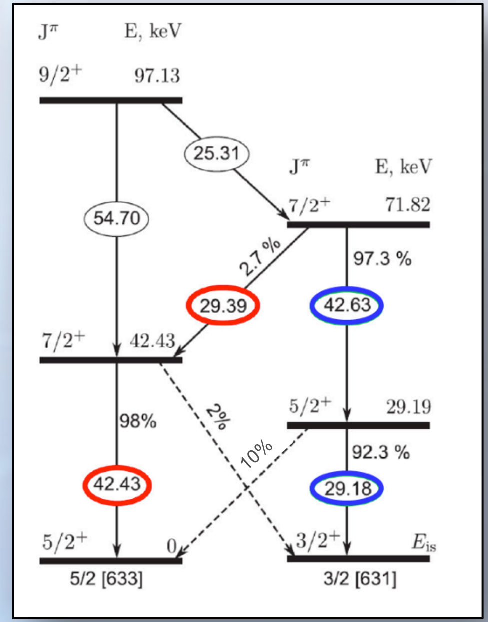
residual „calibration error“: 0.76 eV

# Gamma measurements
# The 2020 measurement

## Differencing scheme (as in Beck et al.)

$$\begin{aligned} E_{iso} &= (E_{29.39\,keV} + E_{42.43\,keV}) - (E_{42.63\,keV} + E_{29.18\,keV}) \\ &= (E_{29.39\,keV} - E_{29.18\,keV}) - (E_{42.63\,keV} - E_{42.43\,keV}) \\ &= \Delta E_{29\,keV} - \Delta E_{42\,keV} \end{aligned}$$





**Isomer energy:**

$E_{iso} = (8.10 \pm 0.17)$ eV

**Branching ratios:**

$b_{29} = 9.3(6)$ %

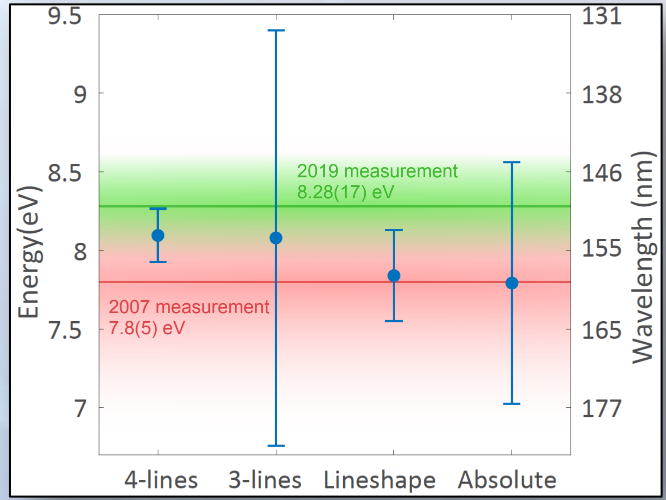
$b_{42} = 0.3(3)$ %

- callibration error drops out (close-by lines)
- exact value of branching ratio not critical
- absolute energy callibration not critical

# Gamma measurements
# The 2020 measurement

## 4 different ways to extract the isomer energy from this data

- 4-line difference (as 2007)
- different decay paths
- doublett fitting
- combining with Spring-8 result...



## What can we learn from Sikorsky et al. 2020

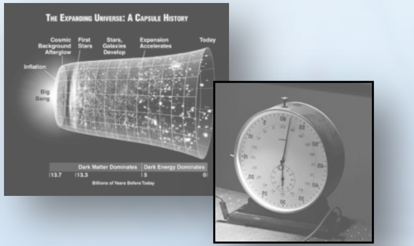
- isomer energy around 8.1 eV (153 nm)
- still within the range of VUV lasers (hard!)
- within the bandgap of VUV materials (fluoride crystals)
- further improving the detectors will further reduce the error...

# New results on $^{229}$Th

## Questions ???

Maybe now?

# New results on $^{229}$Th
# Outline



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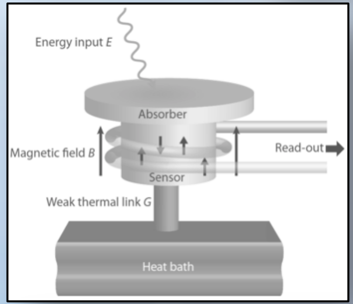

## Experiment 1: Gamma measurements

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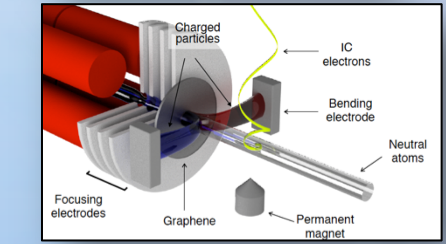
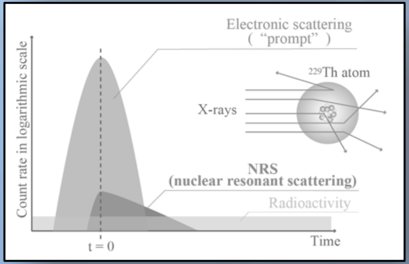


## Experiment 2: IC measurements

- The 2017 LMU measurement: proof of existence
- The 2018 PTB + LMU measurement: optical transitions
- The 2019 measurement: isomer energy



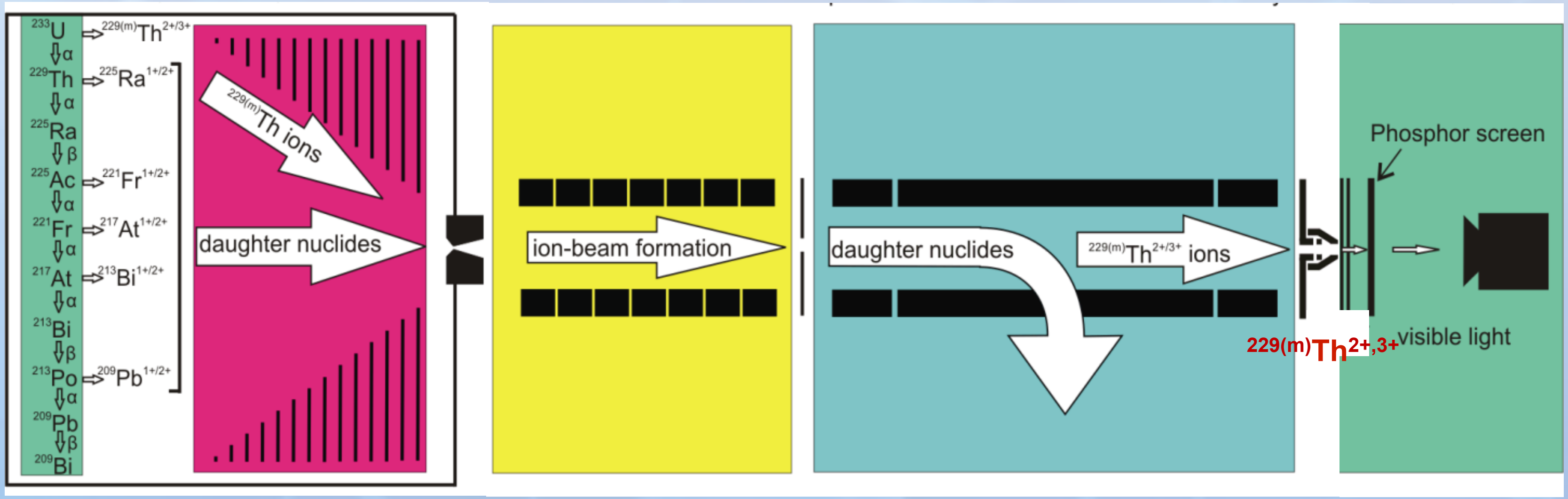
## Experiment 3: X-ray pumping

- The 2019 SPring-8 experiment: isomer pumping

+ isomer energy + branching ratio

# IC measurements
# The 2017 LMU measurement

**Exp. concept:**
- populate the isomeric state via 2% decay branch in $\alpha$ decay of $^{233}$U
- spatially decouple $^{229(m)}$Th recoils from the $^{233}$U source
- detect the subsequently occurring isomeric decay



**Buffer gas stopping cell**

**Laval nozzle**

**aperture electrode**

**triodic extraction**

**detection system**

**$^{233}$U source**

**RF + DC funnel**

**RF quadrupole ion guide/ ion trap**

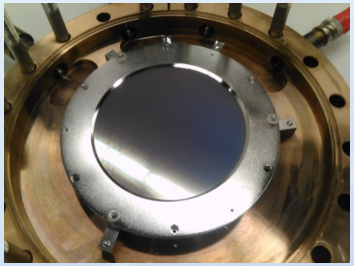
**Quadrupole mass separator**

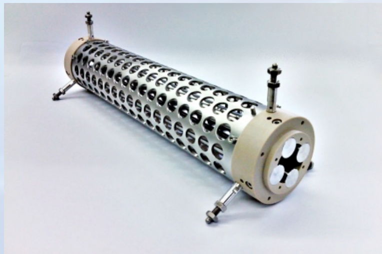
**MCP**

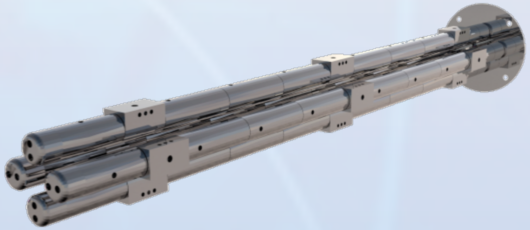
**CCD**

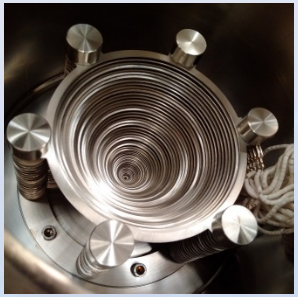
# IC measurements
# The 2017 LMU measurement

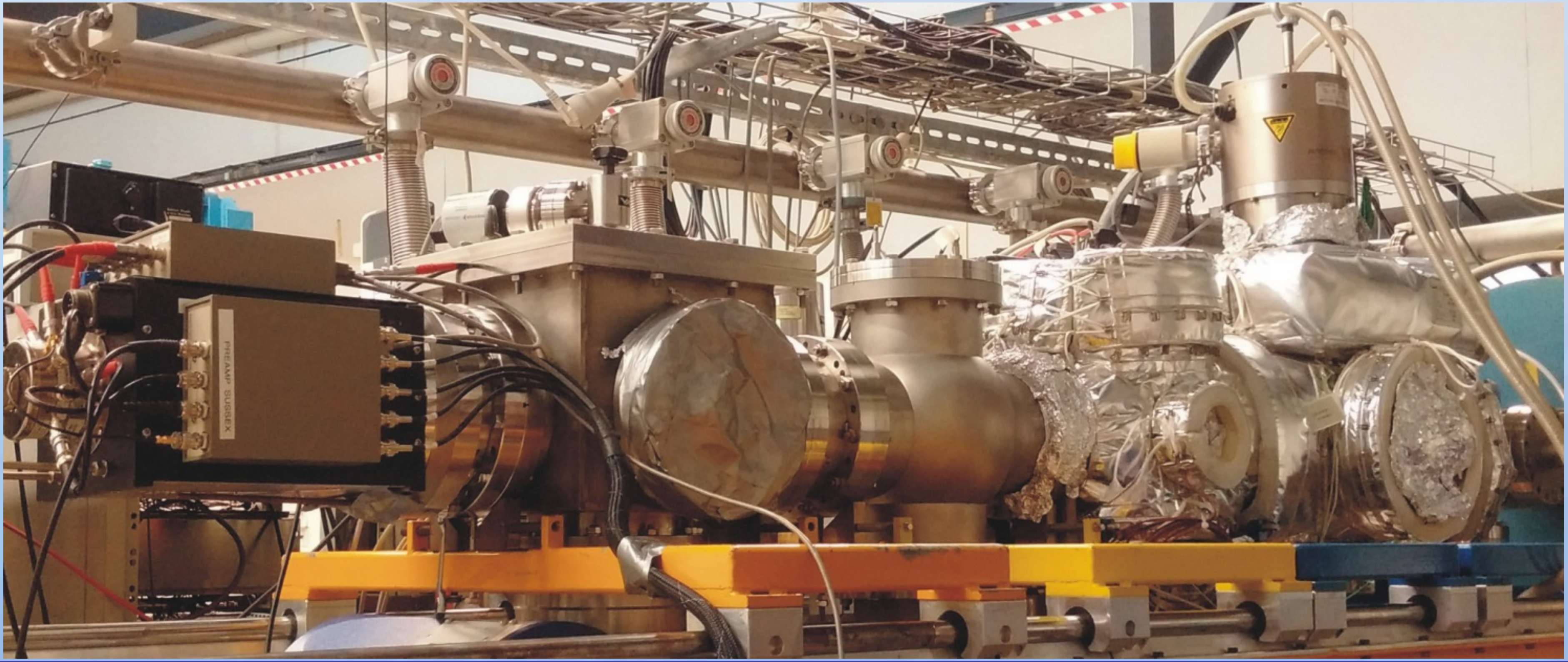
located at Maier-Leibnitz Laboratory, Garching:





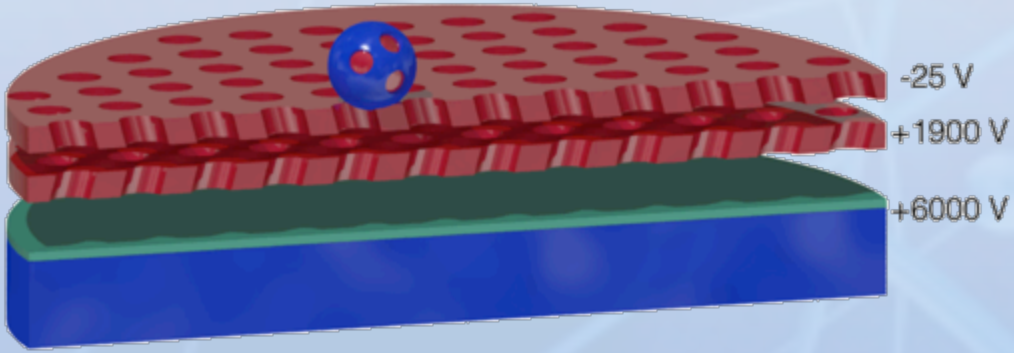






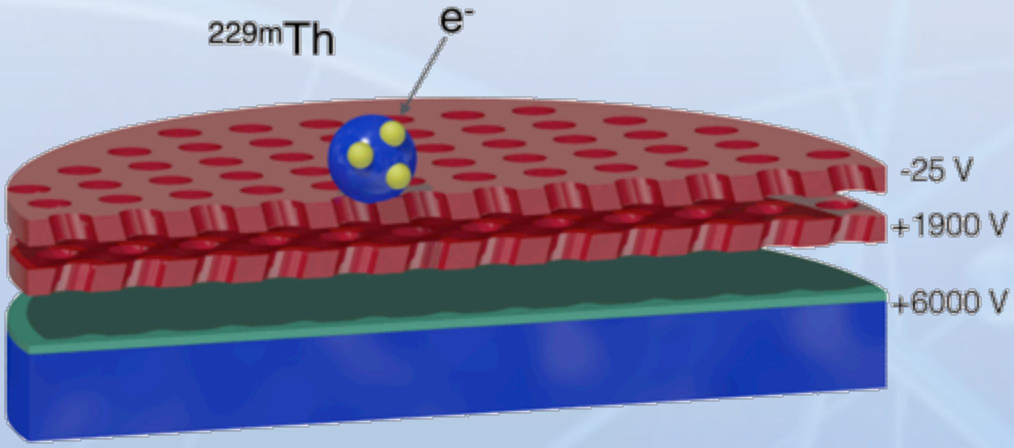
# IC measurements
# The 2017 LMU measurement

**extracted $^{229(m)}Th^{3+}$ ions: ‘soft landing’ on MCP surface**



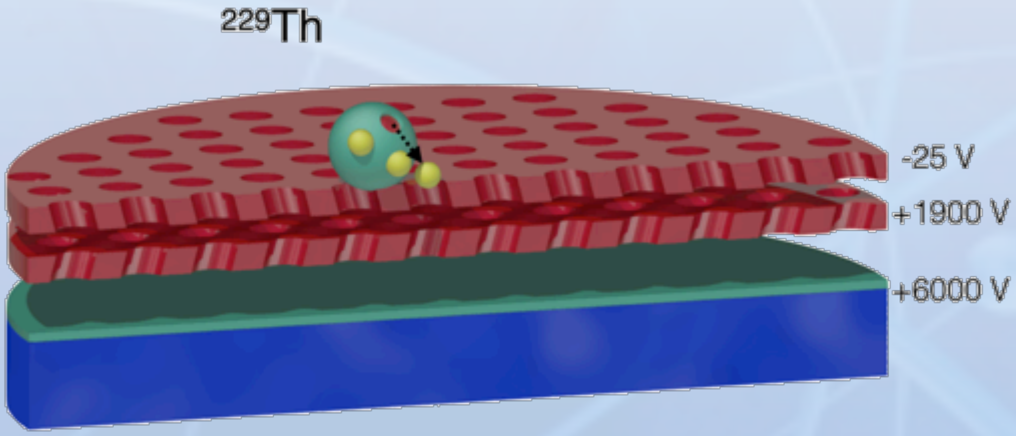
# IC measurements
# The 2017 LMU measurement

**electron capture on MCP surface:** **neutralization of Th ions**



# IC measurements
# The 2017 LMU measurement

## isomer decay by Internal Conversion: **electron emission**



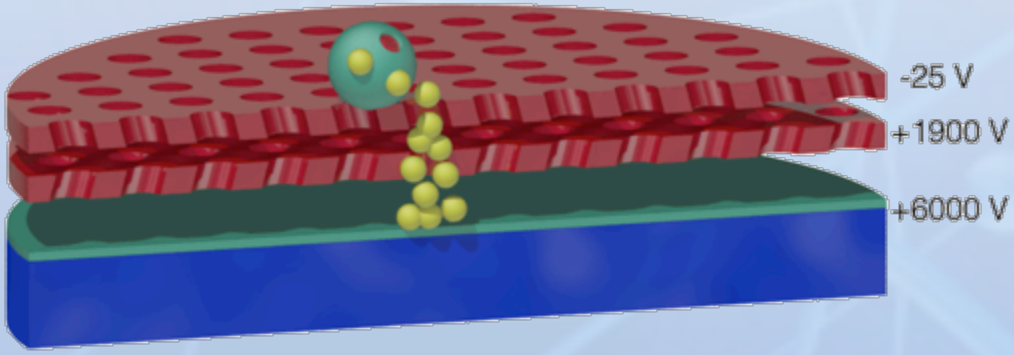
- internal conversion (IC) energetically allowed for neutral thorium:

  $I(Th^+, 6.31\ eV) < E^*(^{229m}Th, \sim 7.8\ eV)$

- isomer lifetime expected to be reduced by ca. $10^{-9}$ (from $\sim 10^4$ s → $\sim 10\ \mu s$)
- Th ions: IC is energetically forbidden, radiative decay branch may dominate

# IC measurements
# The 2017 LMU measurement

**amplification of IC electron:** **electron cascade generated, accelerated towards phosphor screen**



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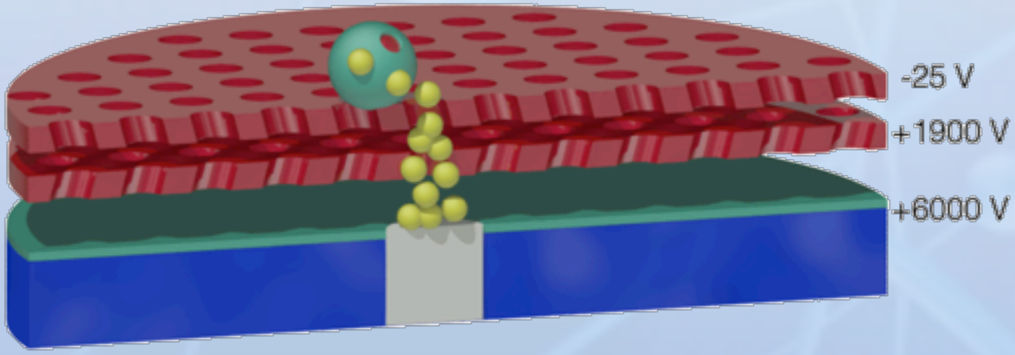
- Th ions: IC is energetically forbidden, radiative decay branch may dominate

# IC measurements
# The 2017 LMU measurement

**electrons hit phosphor screen:** **visible light imaged by CCD camera**



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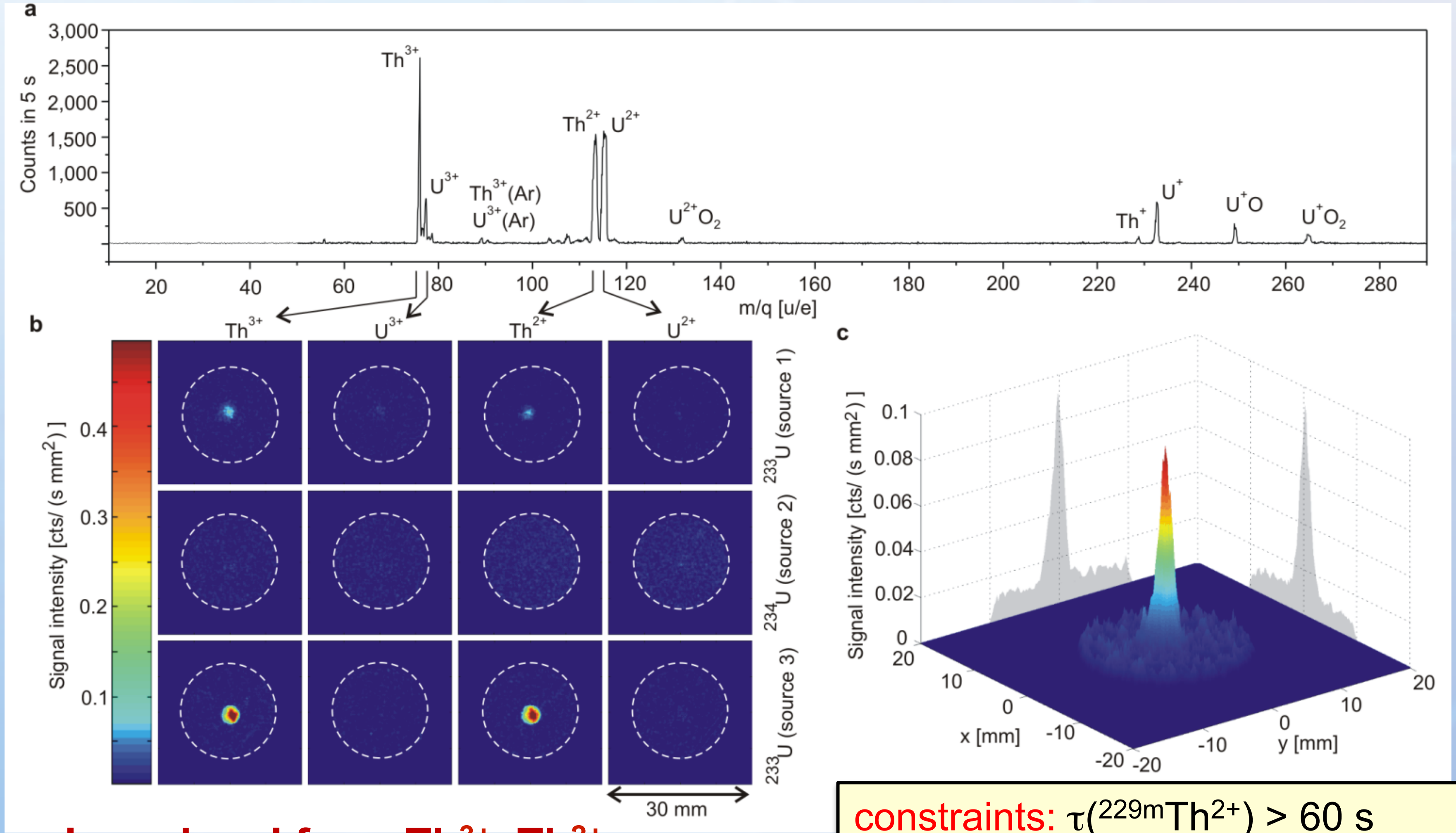
# IC measurements
# The 2017 LMU measurement



**clear signal from $Th^{3+}$, $Th^{2+}$**
**no signal from $U^{3+}$, $U^{2+}$**

constraints: $\tau(^{229m}Th^{2+}) > 60$ s
$6.3\ eV \leq E_{iso} \leq 18.3\ eV$

**L. v.d. Wense, B. Seiferle, P.G. Thirolf et al., Nature 533, 47-51 (2016)**

# IC measurements
# The 2017 LMU measurement

## What we learned from 2017 LMU experiment:

- „direct“ proof of existence of isomer state
- internal conversion THE dominating process for neutrals
- lifetime of $^{229}Th^{m}$ in neutrals: 7 µs (follow-up publication)
- isomer energy BELOW 18.3 eV

  (2nd ionization energy, $Th^{2+}$ lives „forever“)
- isomer energy is ABOVE 6.3 eV

  (1st ionization energy, otherwise IC would not work)

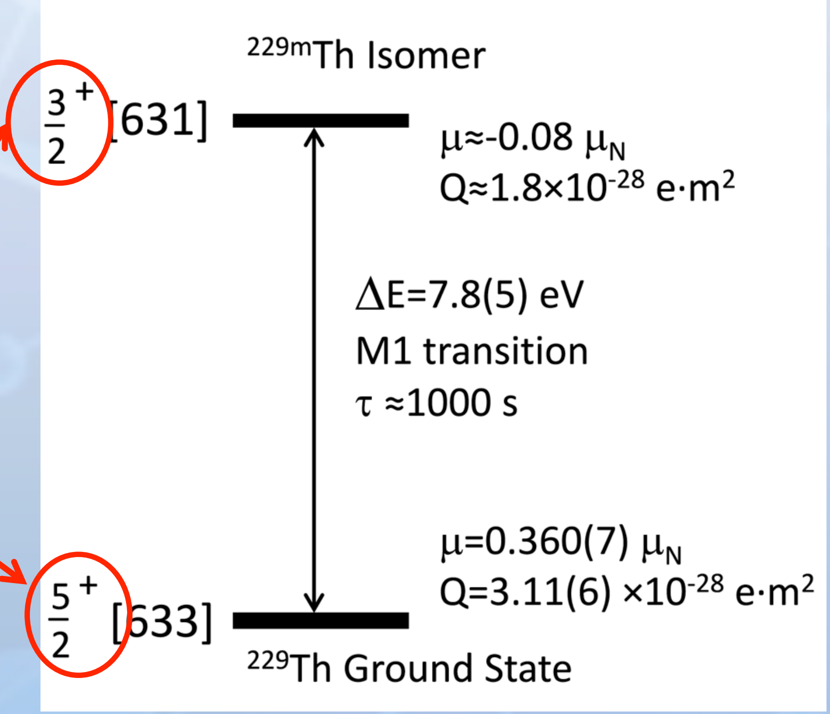
# IC measurements
# The 2018 PTB/LMU laser spectroscopy

## What we (believe to) know about the $^{229}$Th isomer

nuclear ground and isomer state have different magnetic moments! (quadrupole component)

→ different electronic hyperfine structures for ground and isomer state

→ should be able to see that in „standard“ laser spectroscopy

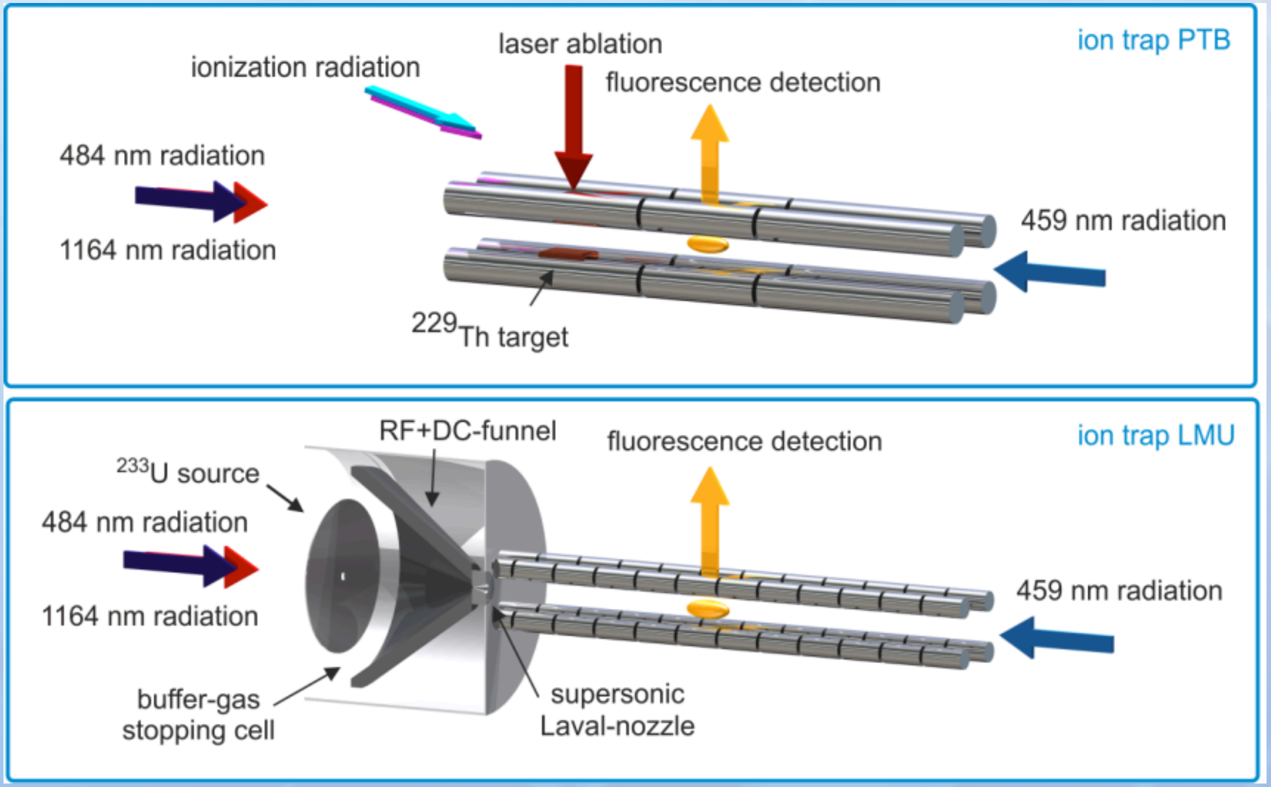


**E. Peik, M. Okhapkin, *Comptes Rendus Physique*, Volume 16, Issue 5, Pages 516-523 (2015)**

# IC measurements (not really!)
# The 2018 PTB/LMU laser spectroscopy

Joint experiments of LMU and PTB, combining:

- Production and storage of $^{229m}$Th ions
- High-resolution laser spectroscopy of Th$^{2+}$



Development of the laser spectroscopy scheme; reference spectra with $^{229g}$Th$^{2+}$ and $^{232}$Th$^{2+}$

ground state

Spectra of $^{229m}$Th$^{2+}$ and $^{229g}$Th$^{2+}$

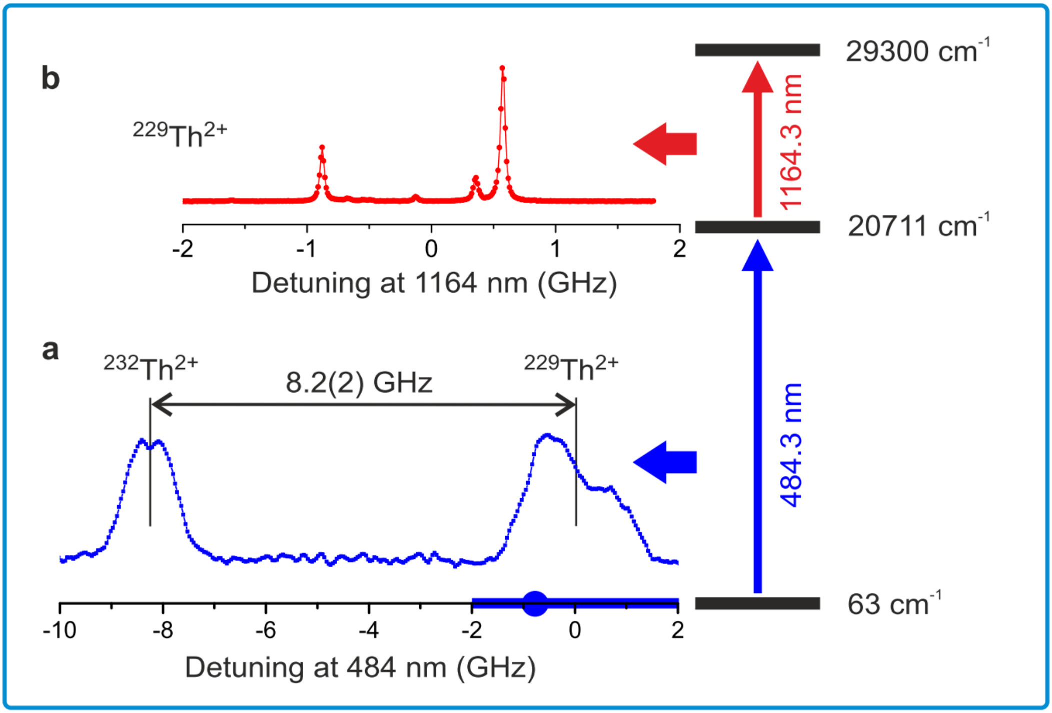
ground + isomer state

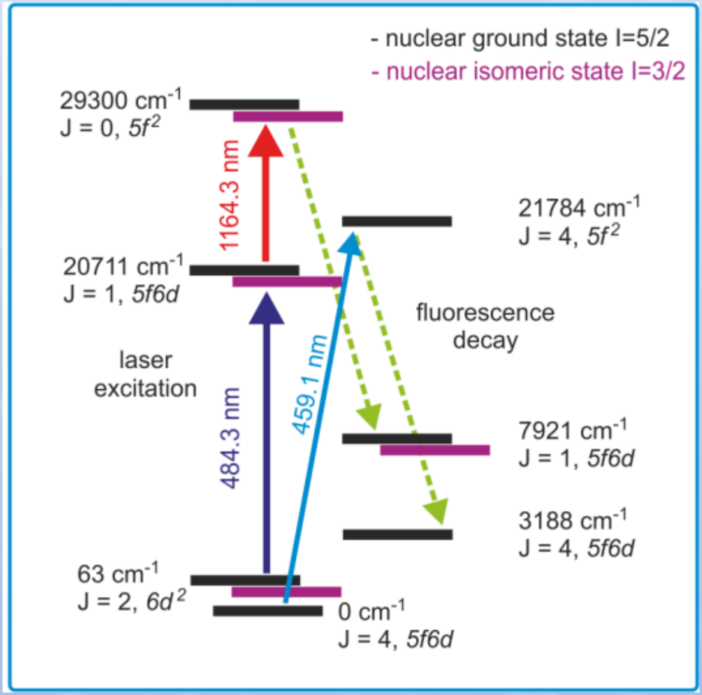
**J. Thielking, M. V. Okhapkin, P. Glowacki, D. M. Meier, L. v.d. Wense, B. Seiferle, C. E. Düllmann, P. G. Thirolf, E. Peik: Laser spectroscopic characterization of the nuclear clock isomer $^{229m}$Th**

# IC measurements (not really!)
# The 2018 PTB/LMU laser spectroscopy

Two-step laser excitation eliminates Doppler-broadening:

- 1st step: excitation of a narrow velocity class out of the 300 K thermal distribution
- 2nd step: high-resolution spectroscopy, free from Doppler-broadening
- Spectra of step 2 for various detunings of laser 1 provide the resolved HFS of both transitions

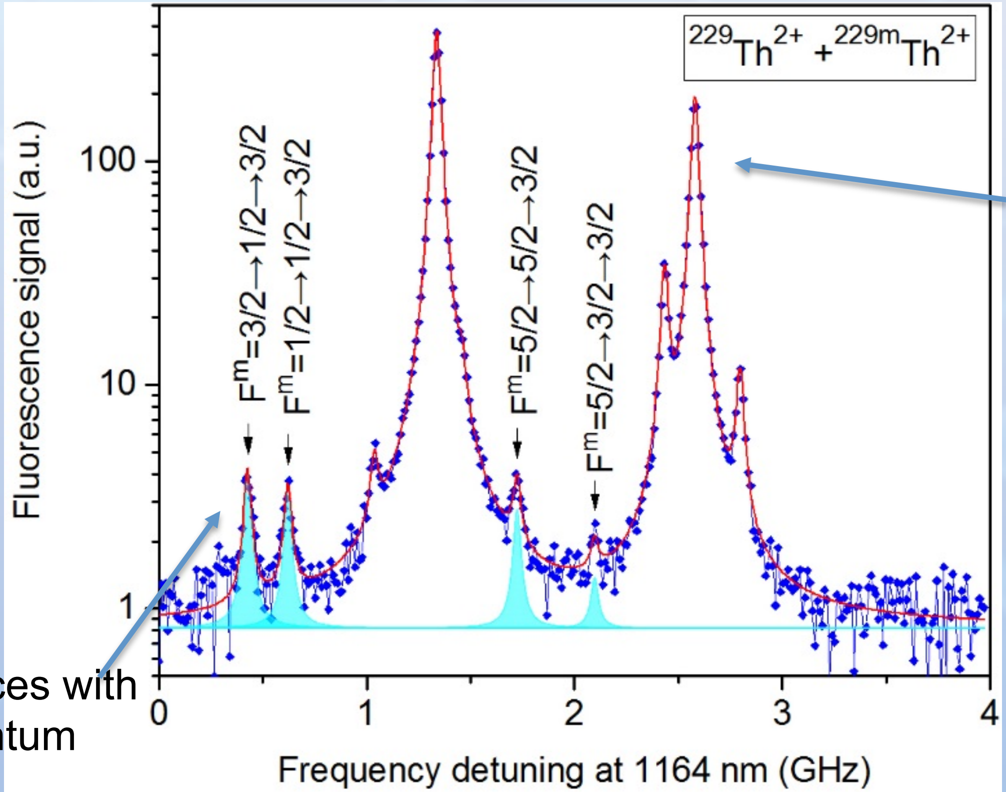




- Fluorescence detection free from laser straylight
- Levels with J=2─1─0 simplify the analysis

# IC measurements (not really!)
# The 2018 PTB/LMU laser spectroscopy

Typical HFS resonances of nuclear isomeric and ground states in scan of the 2nd-step laser.



Nuclear ground state resonances

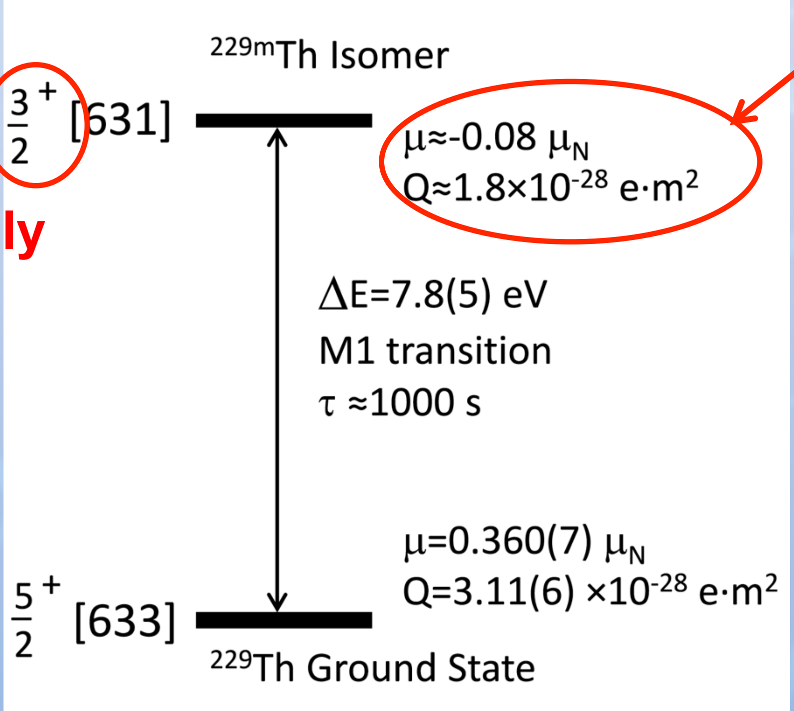
Cyan: isomer resonances with
F: total angular momentum

Total data set: 70 spectra for different detunings of the 1st-step laser and in co- and counter-propagation beam configuations.

# IC measurements (not really!)
# The 2018 PTB/LMU laser spectroscopy

## What we learned from 2018 PTB/LMU experiment:

- even more „direct“ proof of existence of isomer state
- laser spectroscopy can easily distinguish between ground/isomer
- a series a nuclear parameters measured...



$^{229m}$Th Isomer

$\frac{3}{2}^{+}$ [631]

$\mu \approx -0.08\ \mu_N$
$Q \approx 1.8 \times 10^{-28}$ e·m$^2$

$\Delta E = 7.8(5)$ eV
M1 transition
$\tau \approx 1000$ s

$\mu = 0.360(7)\ \mu_N$
$Q = 3.11(6) \times 10^{-28}$ e·m$^2$

$\frac{5}{2}^{+}$ [633]

$^{229}$Th Ground State

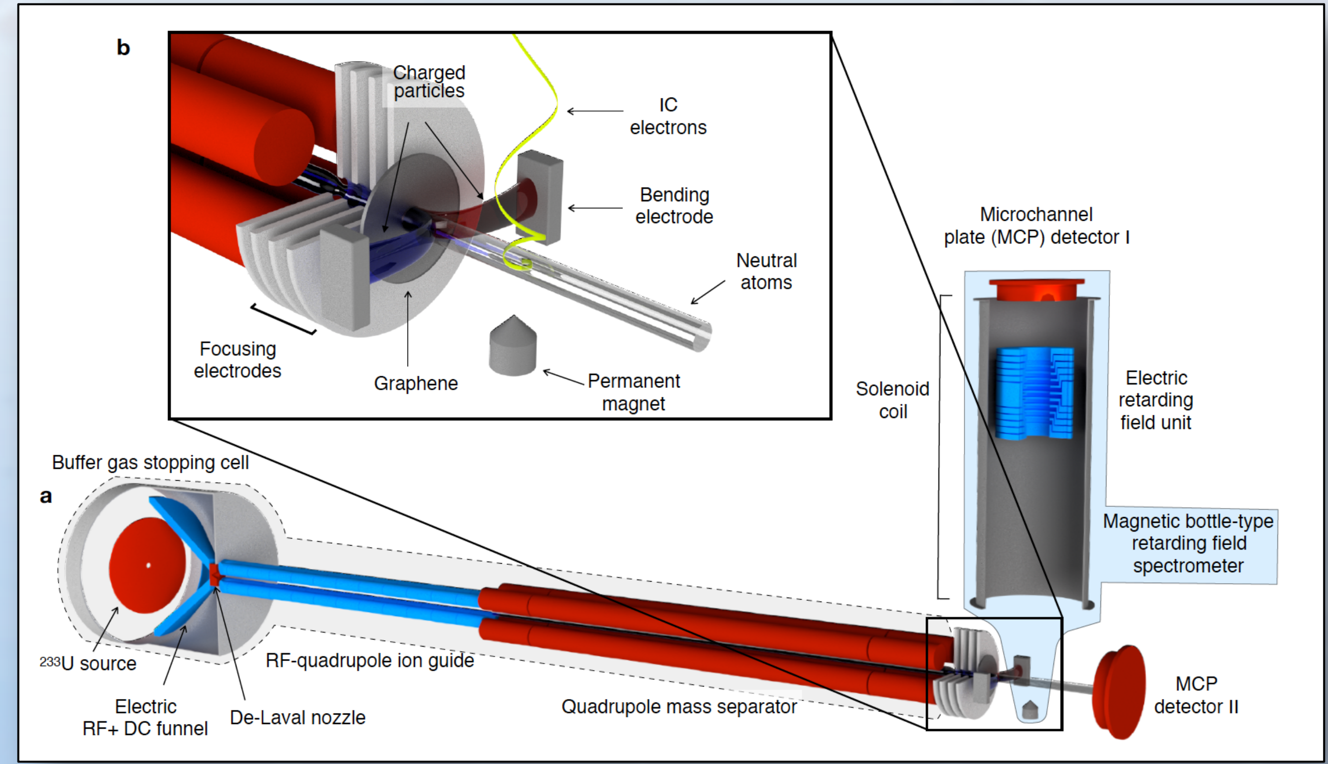
**unambiguously confirmed**

**measured to**
**$\mu = -0.37(6)\mu_N$**
**$Q = 1.74(6) \times 10^{-28}$ e·m$^2$**

**+ 2% branching into $^{229m}$Th in $^{233}$U $\alpha$-decay confirmed**

**+ mean square charge radius change**
**$\langle r^2 \rangle^{229m} - \langle r^2 \rangle^{229} = 0.012(2)$ fm$^2$**

# IC measurements (now again!)
# The 2019 measurement



- neutralization of $Th^{3+}$ ($Th^{2+}$) by passing through graphene foil
- in-flight internal conversion free of surface effects (subtleties here!)
- kinetic energy measurement of IC electron

$$E_{kin} = E_{iso} - IP + E_i - E_f$$

# IC measurements (now again!)
# The 2019 measurement

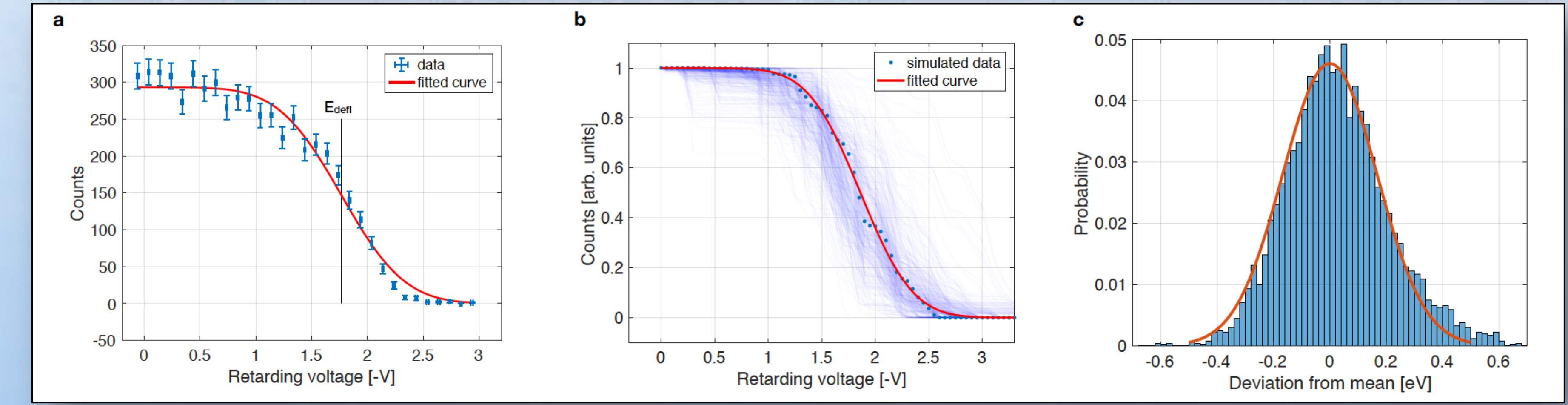
$$\boxed{E_{kin} = E_{iso} - IP + E_i - E_f}$$

**IP:** ionization potential of neutral ground-state Thorium: 6.31 eV

**$E_i$:** (possible) excitation energy of ion undergoing IC

**$E_f$:** final electronic state of the remaining Th1+ ion

} needs theory input!



Simulating a statistical average of the „most probable“ electronic configurations for a given $E_{iso}$ yields:

$$\boxed{E_{iso} = (8.28 \pm 0.03_{stat.} \pm 0.16_{syst.})\ \mathrm{eV}}$$

# IC measurements (now again!)
## The 2019 measurement

**What we learned from 2019 LMU experiment:**

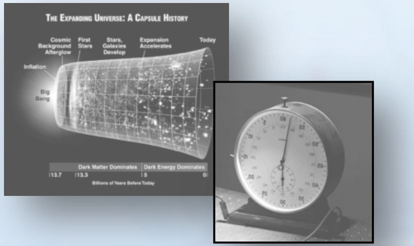
- isomer energy around 8.3 eV (150 nm)
- within the range of VUV lasers (hard!)
- within the bandgap of VUV materials (fluoride crystals)
- not much further improvement possible (systematic error)

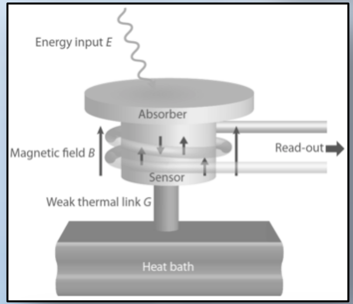
# New results on $^{229}$Th

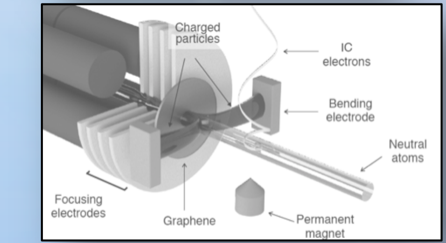
## Questions ???

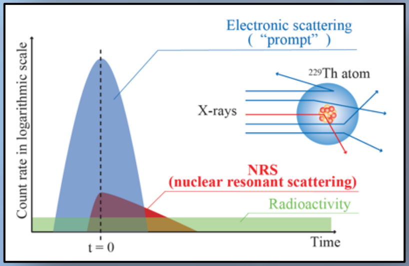
NOW!

# New results on $^{229}$Th
# Outline









## Introduction: nuclear physics with a laser?

- The low-energy nuclear transition in $^{229}$Thorium
- On nuclear clocks and fundamental constants

## Experiment 1: Gamma measurements

- A brief history of the $^{229}$Th isomer energy
- The 2007 Beck et al. gamma measurement
- The 2020 measurement: isomer energy + branching ratio

## Experiment 2: IC measurements

- The 2017 LMU measurement: proof of existence
- The 2018 PTB + LMU measurement: optical transitions
- The 2019 measurement: isomer energy

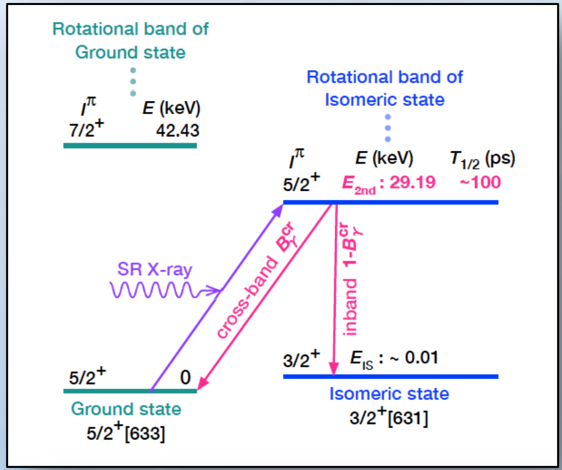
## Experiment 3: X-ray pumping

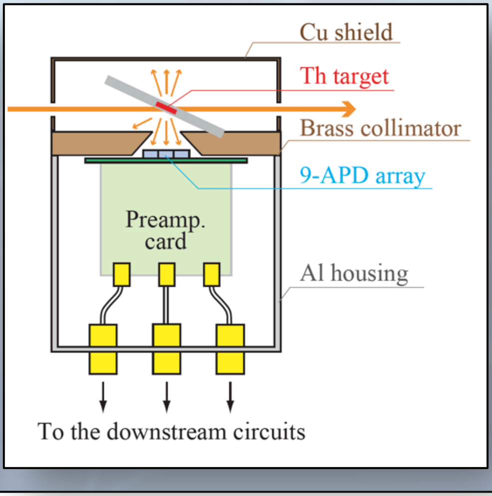
- The 2019 SPring-8 experiment: isomer pumping

+ isomer energy + branching ratio

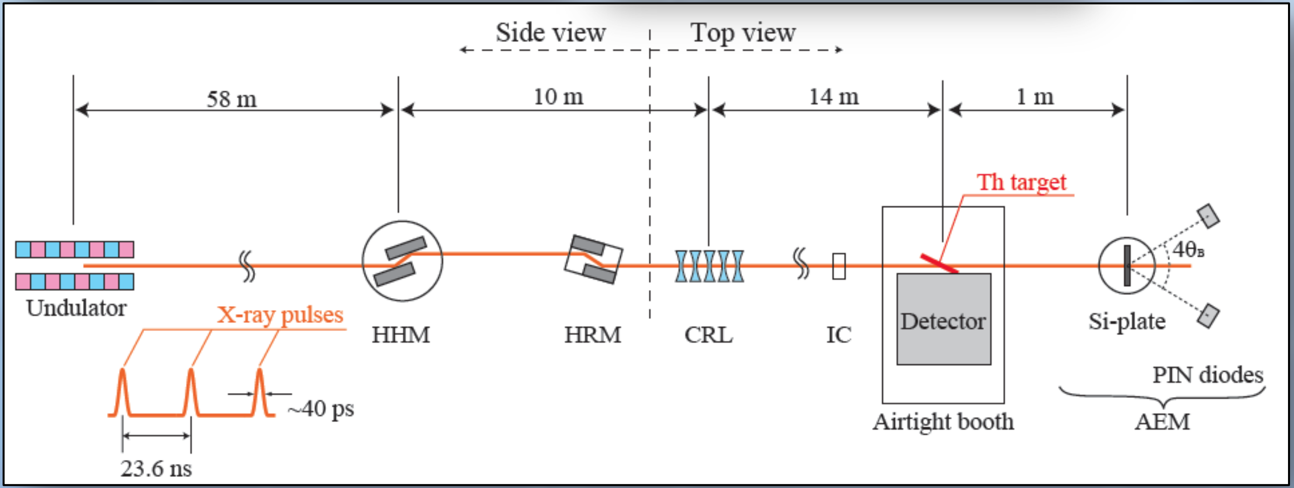
# X-ray pumping
# An introduction to NRS

## Concept: excite the 2nd $^{229}$Th nuclear state in SPring-8 synchrotron





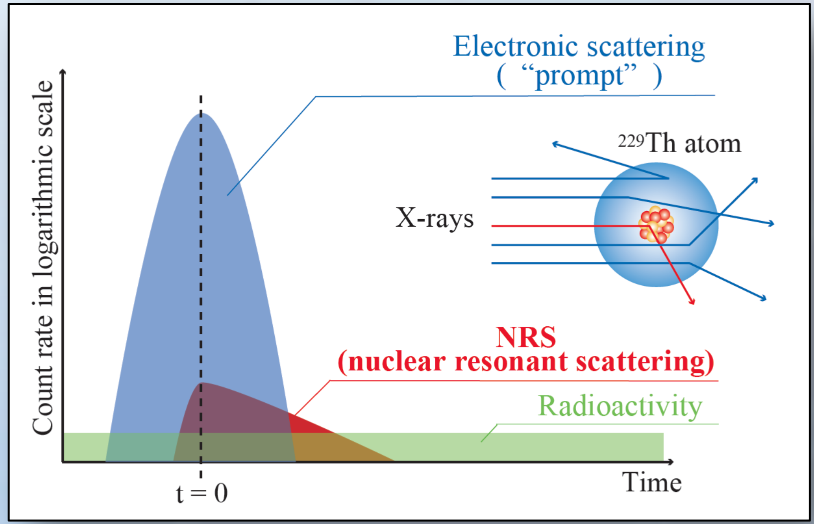
- 1.8 kBq solid $^{229}$Th sample (~$ThO_2$)
- 9 APD detectors (Hamamatsu)
  - energy resolution 21%
  - time resolution 60 ps



BL19LXU beam line, 4 x $10^{12}$ photons/s in 0.26 eV linewidth (later reduced to 0.1 eV)

# X-ray pumping
# An introduction to NRS

Concept: excite the 2nd $^{229}$Th nuclear state in SPring-8 synchrotron



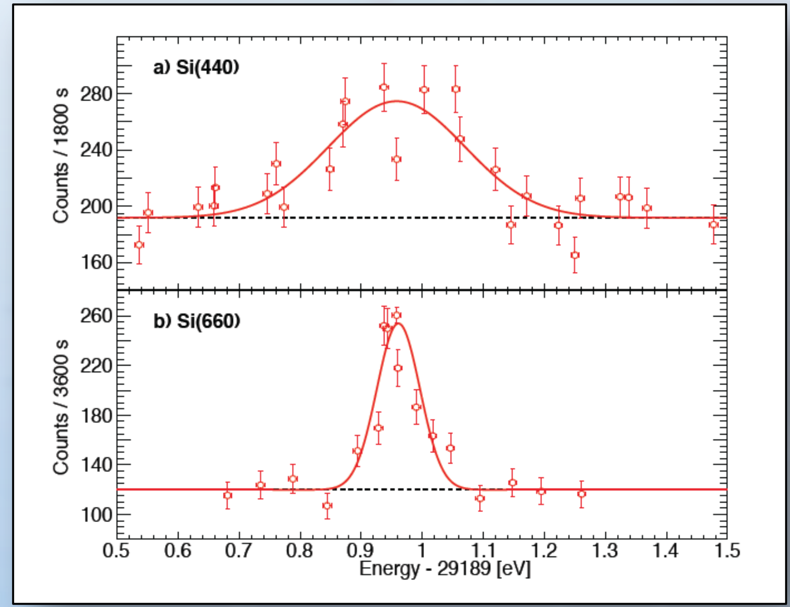
experimental challenges:

- separate NRS signal from prompt peak
- estimated lifetime 150 ps
- signal/background ratio $10^{-6}$
- photoelectric and NRS signature similar

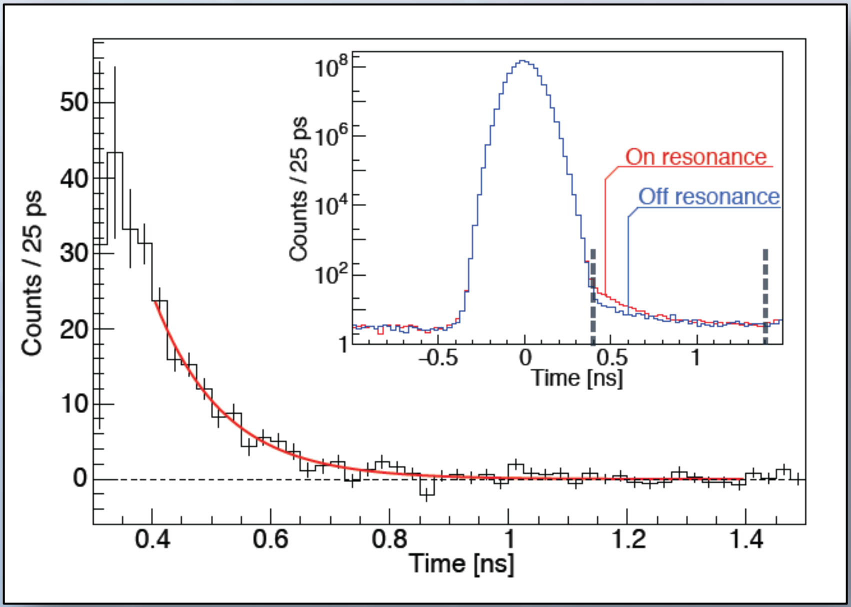
...5 beam times, 4 years of work...

# X-ray pumping
# The 2019 Spring-8 measurement

## NRS resonance curve



## Lifetime and excitation yield



$\boxed{E_{2nd} = (29189.93 \pm 0.07)\ \mathrm{eV}}$

$T_{½} = (82.2 \pm 4.0)\ \mathrm{ps}$

$\Gamma_{\gamma,cr} = (1.70 \pm 0.40)\ \mathrm{neV}$

transition linewidth,
quite some subleties here

combined with known

$\Gamma_{\gamma,in} = (14.3 \pm 1.40)\ \mathrm{neV}$

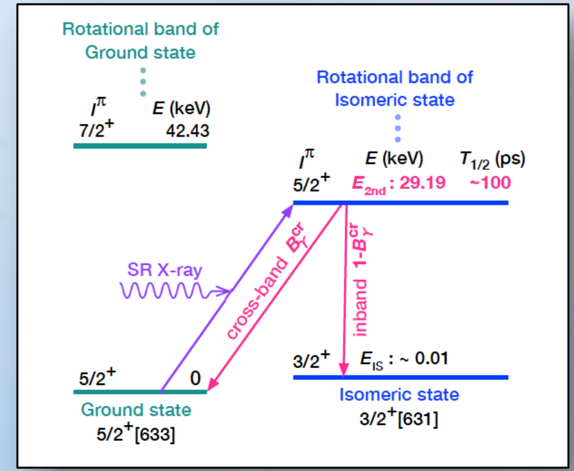
$\boxed{\rightarrow b_{\gamma,cr} = 1/(9.4 \pm 2.4)}$

IC branching ratio: $b_{IC,in} = 0.58 \pm 0.07$, isomer pumping rate 25 kHz

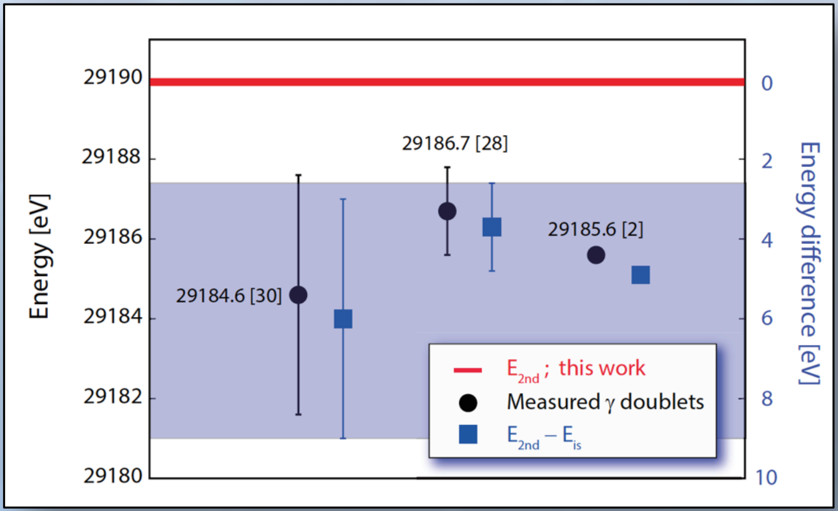
# X-ray pumping
# The 2019 Spring-8 measurement

## Extracting the isomer energy:

- SPring-8 measures ONLY the cross-band energy
- Gamma spectroscopy measures PREDOMINANTLY the intra-band energy (small correction for branching ratio required)



→ isomer energy can be extracted from „old“ gamma spectroscopy data...



- yields energy interval 2.5 eV < $E_{is}$ < 8.9 eV (bof!)
- approach suffers from poor ABSOLUTE calibration of detectors (absolute error not even quantified in Beck et al.)
- combined with 2020 HD measurement : $\boxed{E_{iso} = (7.8 \pm 0.8)\ \text{eV using } b = 1/9.4}$

# New results on $^{229}$Th
# Summary + Outlook

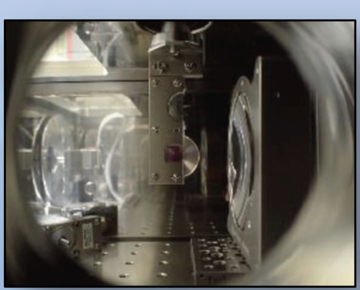
- isomer energy consistently measured around **8.2 eV**
  - clearly VUV (currently no CW laser)
  - clearly compatible with solid-state approaches (if IC suppressed)
- 2nd nuclear level measured with 0.07 eV error
- branching ratio confirmed to be ~ 90% - 10% (in-band – cross-band)

**STILL, no direct excitation of isomer
from ground state or radiative decay detected!**

→ new ERC Synergy

Whats next (from our subjective perspective):

- use X-ray pumping to populate isomer + detect VUV photon
  ...ongoing at SPring-8
- directly excite the isomer in $^{229}$Th-doped crystals
  ...ongoing at Vienna + MLS synchrotron Berlin (also UCLA)
- use beta-decay of $^{229}$Ac
  ...pioneered by Piet Van Duppen, Leuven...



Vienna crystals at Spring-8

ATOMINSTITUT

# We want YOU…
# …for Thorium research!

TU WIEN

thorium nuclear clock

erc
European Research Council
Established by the European Commission

new **ERC Synergy project** started Feb. 2020
laser spectroscopy of Th-229 (and much more)

**Ekkehard PEIK**
Physikalisch-Technische Bundesanstalt

**Peter THIROLF**
Ludwig-Maximilians-Universität München

**Marianna SAFRONOVA**
University of Delaware

**Thorsten SCHUMM**
**Technische Universität Wien**

## ...looking for PhDs and PostDocs!

contact **thorsten.schumm@tuwien.ac.at**

# New results on $^{229}$Th
# Who’s doing that?

## Gamma measurements

| | |
|---|---|
| J. Geist, S. Kempf, A. Fleischmann, L Gastaldo, Chr. Enss | Uni Heidelberg |
| C. Mokry, J. Runge, C. E. Düllmann | GSI, Uni Mainz, HIM |
| T. Sikorsky, J. H. Sterba, V. Rosecker, K. Beeks, G. Kazakov, T. Schumm | TU WIEN |

## IC measurements

| | |
|---|---|
| B. Seiferle, L. v.d. Wense, I. Amersdorffer, P. Thirolf | LMU |
| P. V. Bilous, A Pàlffy | Max-Planck Kernphysik |
| C. Lemell, F. Libisch, S. Stellmer, T. Schumm | TU Wien |
| C. E. Düllmann | GSI, Uni Mainz, HIM |

## SPring-8 measurments

| | |
|---|---|
| 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 | Okayama University |
| H. Fujimoto, T. Watanabe | AIST Tsukuba |
| H. Haba, A. Yamaguchi, T. Yokokita, K. Tamasaku | RIKEN |
| Y Kasamatsu, Y. Yasuda, Y. Shigekawa | Osaka University |
| S. Kitao, M. Seto | Kyoto University |
| K. Konashi, M. Watanabe | Institute f. Material res. Tohoku |
| S. Stellmer, T. Schumm | TU Wien |
| Y. Yoda | SPring-8 beamline |

# New results on $^{229}$Th

## Questions ???

**last chance!**