Concepts and Prospects for a Thorium-229 Nuclear Clock

Ekkehard Peik

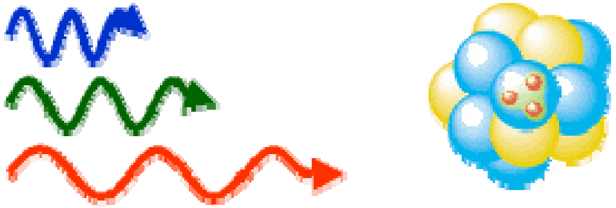
Physikalisch-Technische Bundesanstalt
Time and Frequency Department
Braunschweig, Germany

Outline

- The low-energy isomeric state of ^{229}Th
- ^{229}Th as a nuclear clock
- Expected accuracy of ^{229}Th nuclear clocks
- Search experiments at PTB with solutions and recoils
- Two-photon electronic bridge excitation of ^{229m}Th^{+}

Nuclear Clock:

Based on an oscillator that is frequency-stabilized to a nuclear (γ-ray) transition



Motivation:

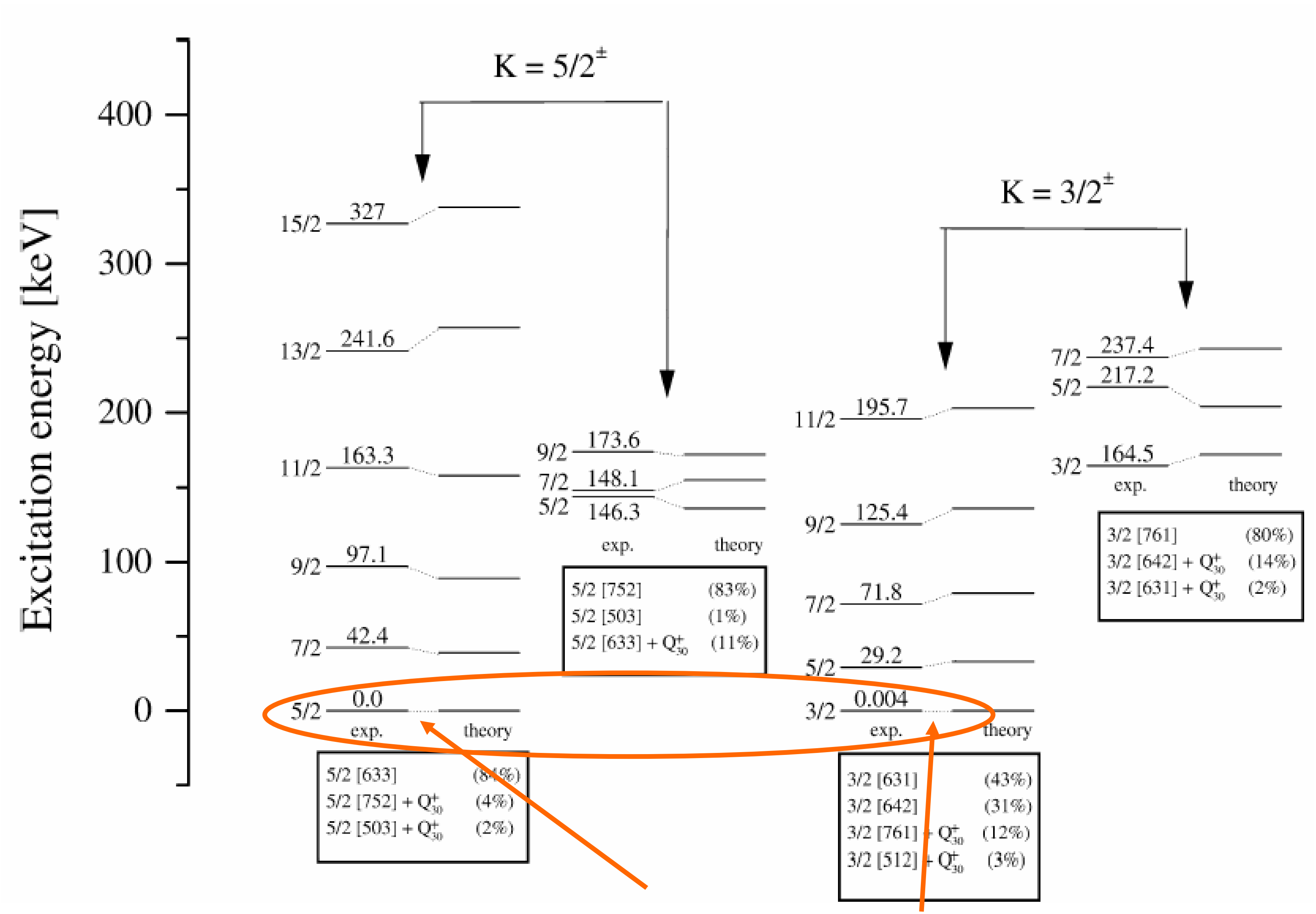
Higher precision: In most of the advanced optical clocks (trapped ion and optical lattice) field-induced shifts make a dominant contribution to the uncertainty budget (exception: Al^+ $J=0-0$). These can be reduced in a nuclear clock.

Higher stability: In a Mößbauer solid state nuclear clock, many absorbers may be interrogated ($>10^{10}$ instead of $\approx 10^0$ (ion) or $\approx 10^4$ (lattice)).

Higher frequency: → higher stability. EUV or even X-ray transitions may be used when suitable radiation sources become available.

K. Gulda et al. / Nuclear Physics A 703 (2002) 45–69

The nuclear structure of ^{229}Th



Two close-lying band-heads: ground state and isomer

Thorium-229: the nucleus with the lowest-lying excited state

The only known isomer with an excitation energy in the optical range and in the range of outer shell electronic transitions.

- Studied by C. W. Reich et al. at INL since the 1970s,
  established the low energy isomer,
  from γ-spectroscopy: 3.5 ± 1.0 eV, published in 1994

- Theoretical work by E. V. Tkalya, F. F. Karpeshin, and others
  isomer lifetime, coupling to electronic excitations, etc.

- False detections of optical emission in the U-233 decay chain in 1997/98

- Proposal of nuclear laser spectroscopy and nuclear clock
  E. Peik and Chr. Tamm, published in 2003

- Unsuccessful search for optical nuclear excitation or decay

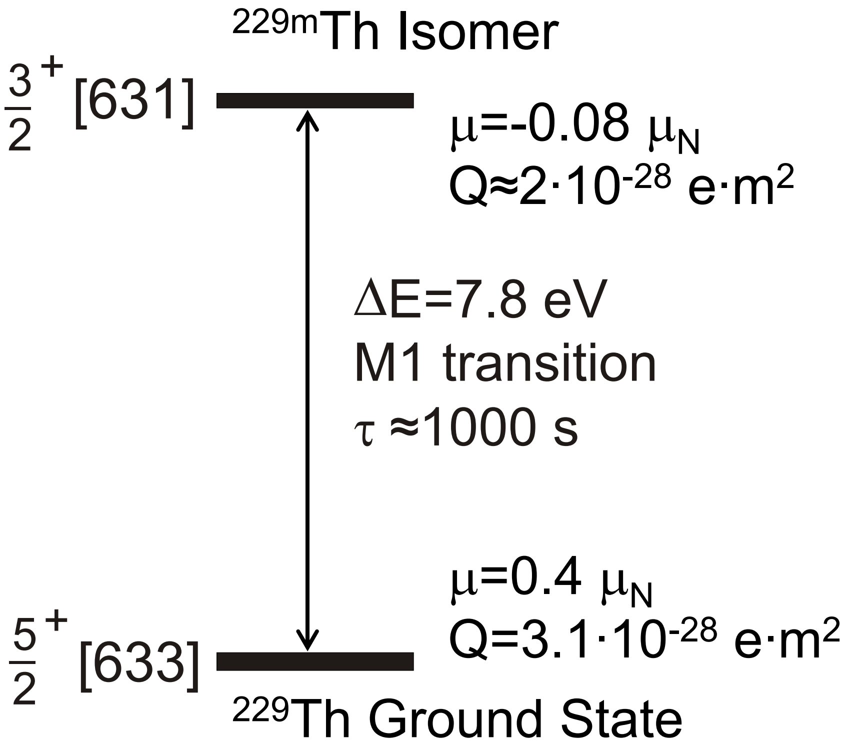
- More precise energy measurement from γ-spectroscopy at LLNL:
  7.6 ± 0.5 eV, published in 2007

- 2012: still no direct detection of the optical transition;
  experimental efforts in several groups worldwide

A high-precision nuclear clock

Nuclear moments are small.
Field induced systematic frequency shifts can be smaller than in an (electronic) atomic clock.

Consider hyperfine coupling, shielding and anti-shielding.
Select suitable electronic state for the nuclear excitation.



^{229m}Th Isomer

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

$\mu=-0.08\ \mu_N$
$Q\approx 2\cdot 10^{-28}\ e\cdot m^2$

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

$\mu=0.4\ \mu_N$
$Q=3.1\cdot 10^{-28}\ e\cdot m^2$

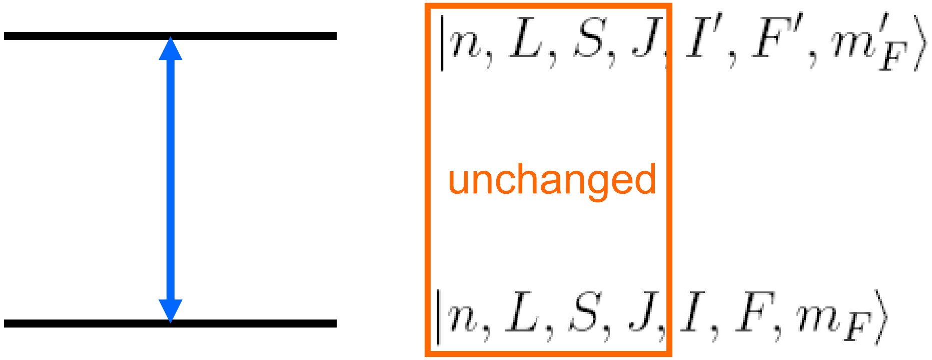
$\frac{5}{2}^{+}$ [633]

^{229}Th Ground State

Analyzed for the Th^{3+} system in:

E. Peik, Chr. Tamm,
Europhys. Lett. **61**, 181 (2003)

LS coupled eigenstates of the electronic + nuclear system



$|n, L, S, J, I', F', m'_F\rangle$

unchanged

$|n, L, S, J, I, F, m_F\rangle$

Frequency shifts that only depend on |n,L,S,J> are common in both levels and do not change the transition frequency.

Holds for: scalar quadratic Stark shift, including the effects
static electric fields, collisions, blackbody AC Stark shift

Tensor quadratic Stark and electric quadrupole shift:
vanish for J<1 or F<1

Hyperfine Stark shift: F-dependent, e.g. blackbody radiation shift
analogy with Cs for m_F=0-0: ≈10^{-19} at room temperature
analysis for stretched states $m_F=\pm(J+I)$: ≈10^{-22},
C.J. Campbell et al., PRL **108**, 120802 (2012)

Linear Zeeman shift: use component $m_F=0-0$

Quadratic Zeeman shift:

$$\Delta f_{2oZ} = \left(g_J - \frac{m_e}{m_p} g_I\right)^2 \frac{\mu_B^2 B^2}{A}$$

Effects are comparable to other atomic clocks

Doppler shift: use ion trapping and laser cooling

→ choice of electronic state:

Half integer nuclear spin (like Th-229): $^2S_{1/2}$ or $^2P_{1/2}$
Integer nuclear spin $^1S_0$ or $^3P_0$

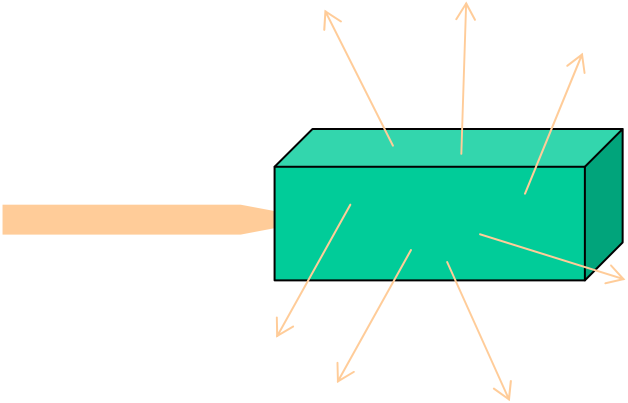
Options for the Th-229 trapped ion clock:

- Metastable 7s $^2S_{1/2}$ level in Th^{3+} (direct laser cooling, quantum jump detection) (our proposal 2003)
- Combined stretched states of $^2F_{5/2}$ ground state in Th^{3+} C. J. Campbell et al., PRL 108, 120802 (2012)
- Sympathetic cooling and quantum logic in higher Th charge states

Eliminates field induced shift to a level not achievable in (electronic) atomic clocks.

Optical Mössbauer Spectroscopy

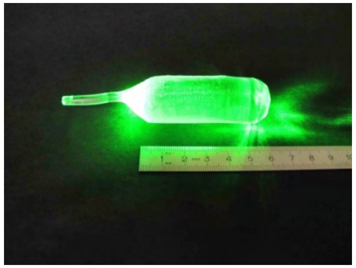
Laser excitation of Th-ions in a solid
→ compact optical frequency standard
of very high stability



Crystal doped with 1 nucleus per λ^3: 10^{11} / mm^3
Simple fluorescence detection is possible;
initial broadband excitation experiment with synchrotron light.

Host crystal should be: transparent, symmetric, diamagnetic.

Possible candidates: fluorides: CaF_2, LiCAF etc.



TU Vienna

Dominant crystal field shift: Electric quadrupole shift

Typical field gradient (e.g. ThB_4 (tetragonal)): $V_{zz}=5\cdot10^{21}$ V/m^2
Antishielding factor for Th: ≈ -100
Resulting Th-229 nuclear ground state quadrupole shift: ≈ 0.1 THz
→ Use suitable crystal symmetry (e.g. cubic).

Predictions for doped Th:CaF2

Electric field gradient from F^- interstitials: $V_{zz}=-5\cdot10^{22}$ V/m^2
Magnetic dipole interaction with ^{19}F nuclei: $\langle B^2\rangle \approx 5\cdot10^{-7}$ T^2
→ decoherence rate for Th nucleus ≈ 10^3/s
G. A. Kazakov et al., New J. Phys. **14**, 083019 (2012)

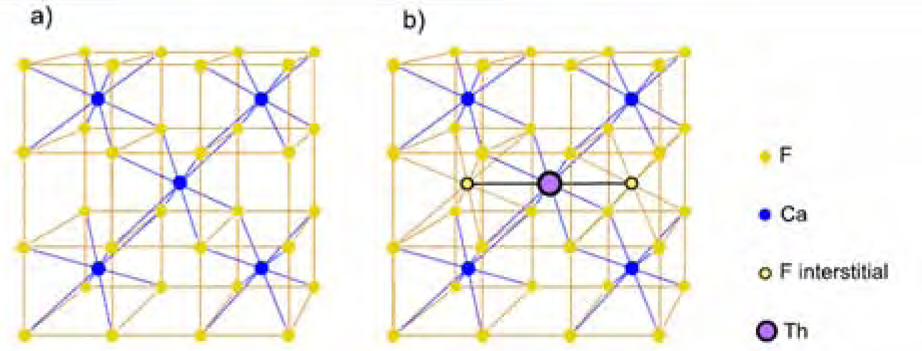


Figure 1. CaF$_2$ crystal lattice (a) and Thorium inclusion with charge compensation by two in-line Fluorine interstitials (b).

Crystal structure calculation for Th:LICAF:
R. A. Jackson et al., J. Phys.: Condens. Matter **21** (2009) 325403

Temperature dependence of linewidth and frequency shifts:

- line shape depends on phonon frequencies and correlation times
- relativistic Doppler shift: 10^{-15} / K
- electric crystal field shifts may be >> 10^{-15} / K

→ For high precision beyond 10^{-15},
work at cryogenic temperature to freeze out lattice fluctuations

E. Peik, K. Zimmermann, M. Okhapkin, Chr. Tamm
Proc. 7th Symp. on Frequency Standards and Metrology (arXiv:0812.3458)

W. Rellergert et al., Phys. Rev. Lett. **104**, 200802 (2010)

G. A. Kazakov et al., New J. Phys. **14**, 083019 (2012)

Possible realisations of Th-229 nuclear clocks:

- Laser-cooled Th^{3+} in an ion trap
- Th ions as dopant in a transparent crystal (like CaF_2, LiCAF etc.)

Experimental problem:
Transition energy known only to ≈ 10% uncertainty,
not a system for high resolution spectroscopy yet.

→ Several experimental projects worldwide,

gathered for the first time at this workshop !

Three search experiments at PTB without success **(2001-2008)**

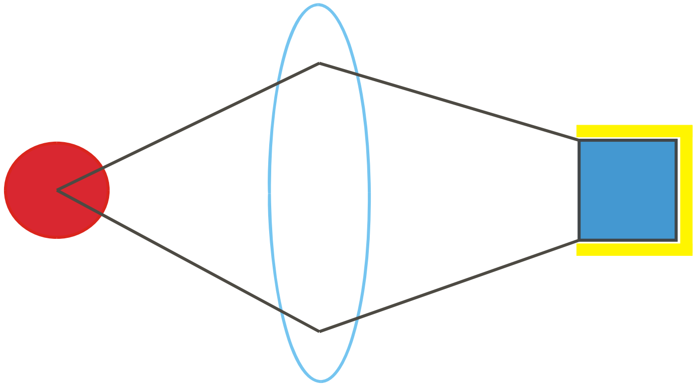
- Fluorescence detection after broadband excitation of the isomer in aqueous solution (search range: 2 - 6 eV)

- Detection in forward scattering of broadband light
(search range: 2 - 6 eV)

- „Freshly“ produced recoil isomers from U-233 alpha decay
(search range: 2 - 9.5 eV)

Fluorescence experiment

1.: Excitation

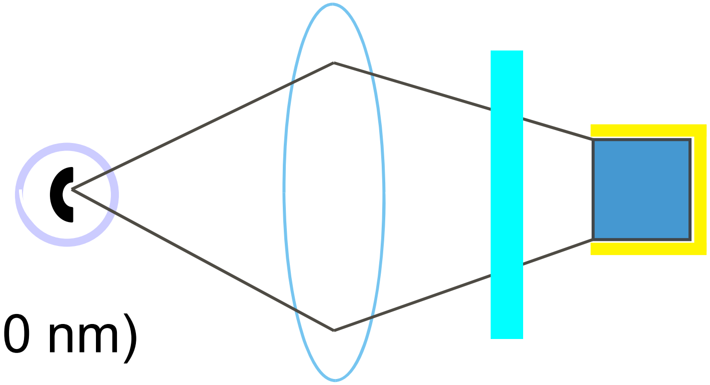


HgXe lamp

Th^{4+} in aqueous solution (10^{16} nuclei)

fused silica cell
with mirrors

2.: Detection



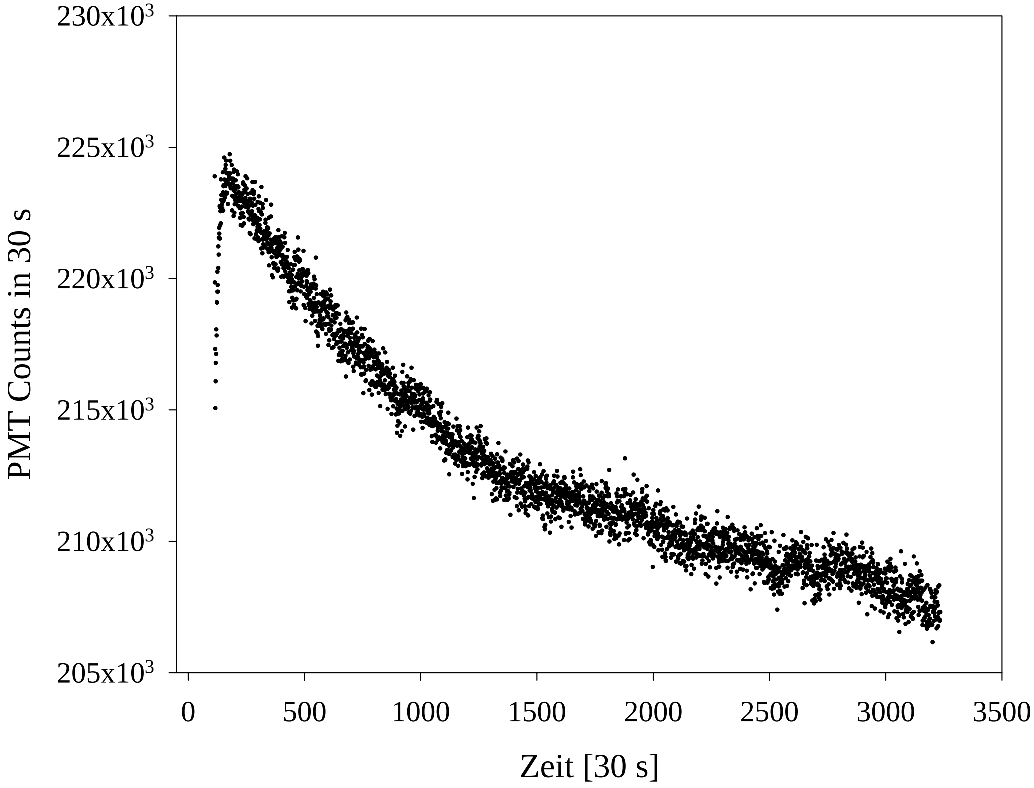
Photomultiplier

(Bialkali, 185 - 680 nm)

cell blocks α,β and γ radiation while coupling 17% of the photons on the PMT.

filter for spectral
narrowing of fluorescence

Experimental result:



Background:

a) Cerenkov radiation (flat spectrum, constant intensity)

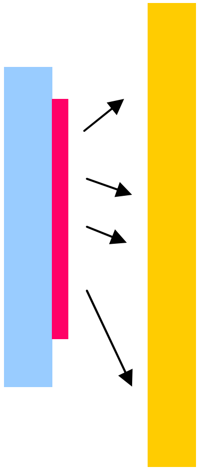
b) Photoluminescence from the fused silica cell

c) Thermoluminescence from the fused silica cell

b) and c): time correlated with excitation, non-exponential decay ($1/t^m$).

Search for the decay of the isomer in Th-229 recoil nuclei

Th-229 recoil nuclei from the alpha decay
of U-233 are emitted with about 80 keV kinetic energy…

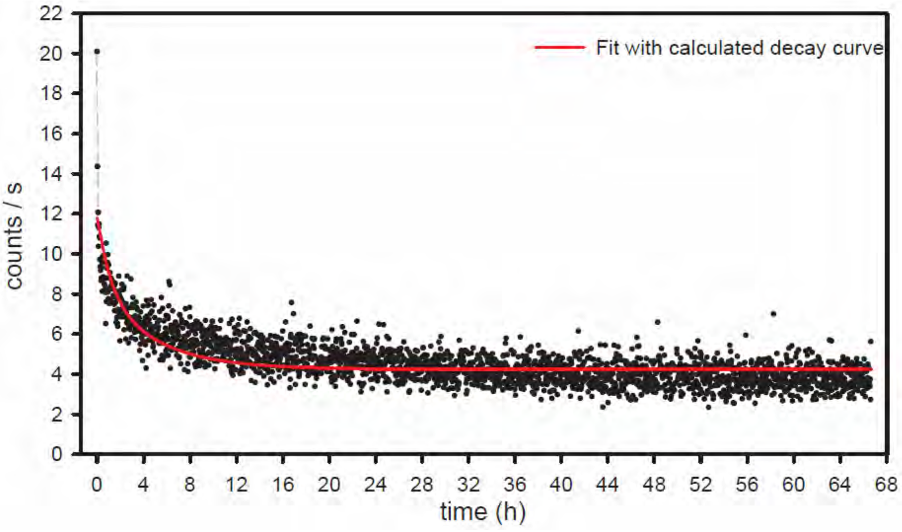


Thin layer of
U-233 (48 kBq),
Electrodeposited
on substrate

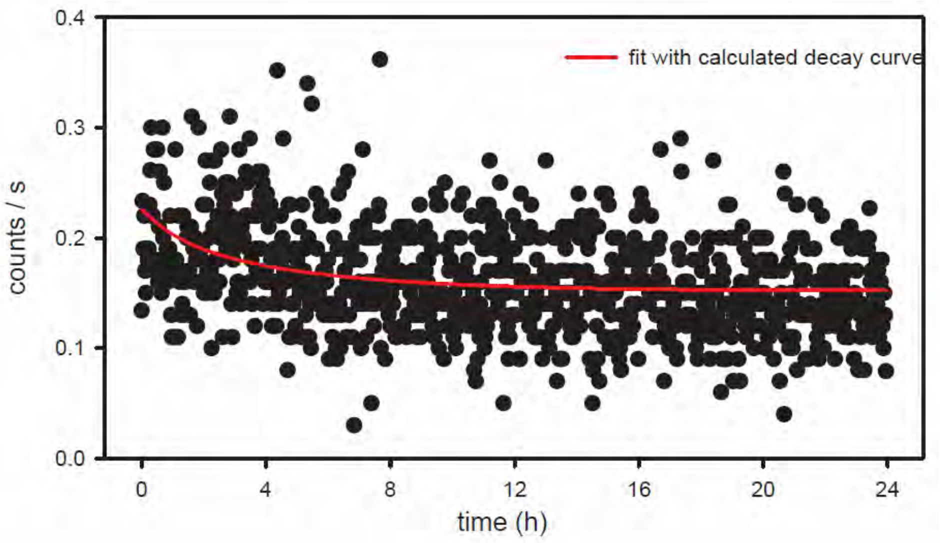
… and may be stopped in an absorber or a trap.
About 10^{-2} of these nuclei are in the isomeric state.

After ≈1 hour of activation, the absorber is placed in front of a photomultiplier (sensitive from 130 to 330 nm, N_2-purged) to look for the decay curve.

Observed decay curves of photons in the range 115 – 320 nm



2 mm CaF_2 absorber



50 μm PVDF absorber

Scaling of the signal with the absorber thickness indicates that we observe luminescence from the volume, most likely caused by β-decay of daughter recoil nuclei (red fit curves; age of the source: 3 yrs).

Total counts in PVDF: more than 10x less than expected from ^{229m}Th decay.

K. Zimmermann, PhD Thesis, Univ. Hannover, 2010

Possible reasons for the failure of this experiment:

- insufficient VUV transmission from absorber to detector
- less than 2% ^{229m}Th among the ^{229}Th recoils
- decay of the isomer in the solid is radiationless
- decay time < 30 s
- wavelength of emitted photons < 130 nm

Our „search“ strategy: Nuclear excitation via an „electronic bridge“

- „Electronic Bridge“ as an enhancement of γ-decay, theoretically discussed by V.A. Krutov and V.N. Fomenko, 1968

- „Inverse electronic bridge“ or NEET (Nuclear Excitation by Electron Transition): Transfer of excitation from the electron shell to the nucleus
  see reviews: S. Matinyan, Phys. Rep. **298**, 199 (1998),
  E. V. Tkalya, Phys. Uspekhi **46,** 315 (2003)

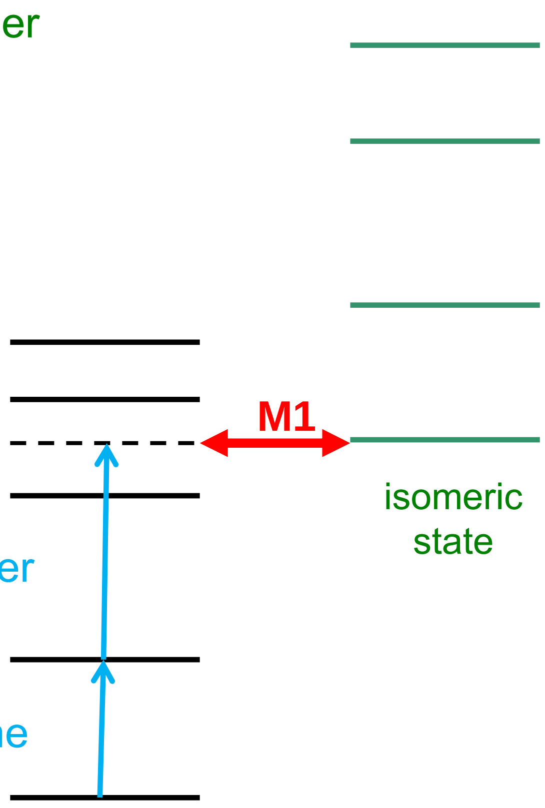
In our case:

- Excitation of the shell in a two-photon process,
  no tunable laser at ≈ 160 nm required

- Excitation rate may be strongly enhanced at resonance between electronic and nuclear transition frequency,
  This is very likely in the dense level structure of Th^+

Two-photon electronic bridge excitation (I)

Schematic of electronic energy levels
with nuclear ground state and isomer



M1

magnetic dipole
hyperfine mixing

isomeric
state

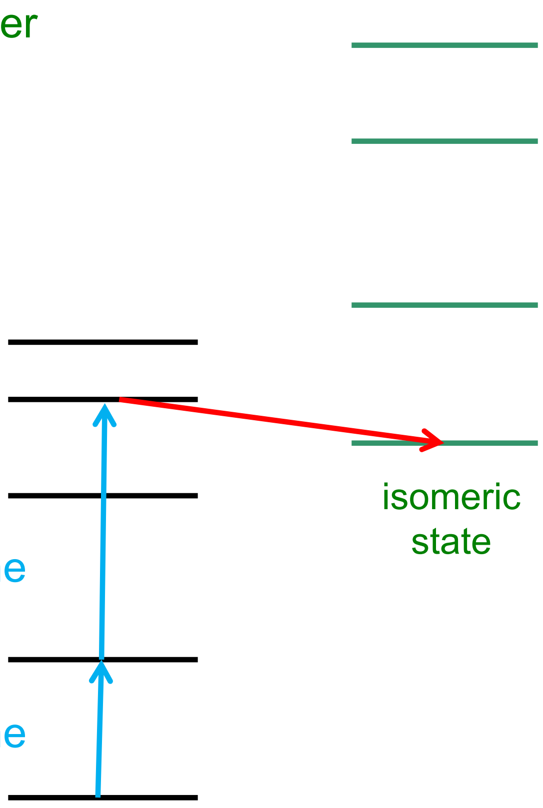
2. Photon from tunable laser

1. Photon on resonance line

nuclear
ground
state

Two-photon electronic bridge excitation (II)

Schematic of electronic energy levels
with nuclear ground state and isomer



2. Photon on resonance line

1. Photon on resonance line

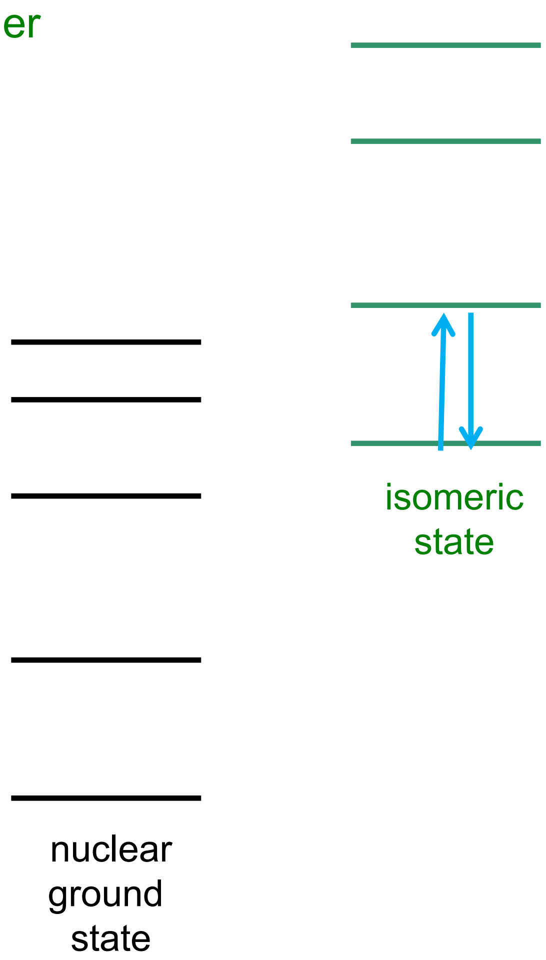
nuclear
ground
state

isomeric
state

spontaneous
emission

Detection of the nuclear excitation (I): long-lived isomer

Schematic of electronic energy levels
with nuclear ground state and isomer



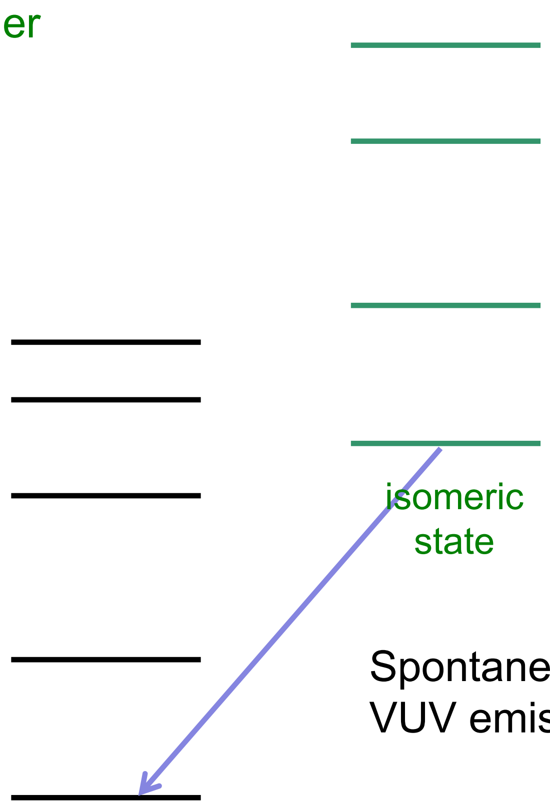
nuclear
ground
state

isomeric
state

Laser-induced fluorescence probes the change in hyperfine structure

Detection of the nuclear excitation (II): isomer decay

Schematic of electronic energy levels
with nuclear ground state and isomer



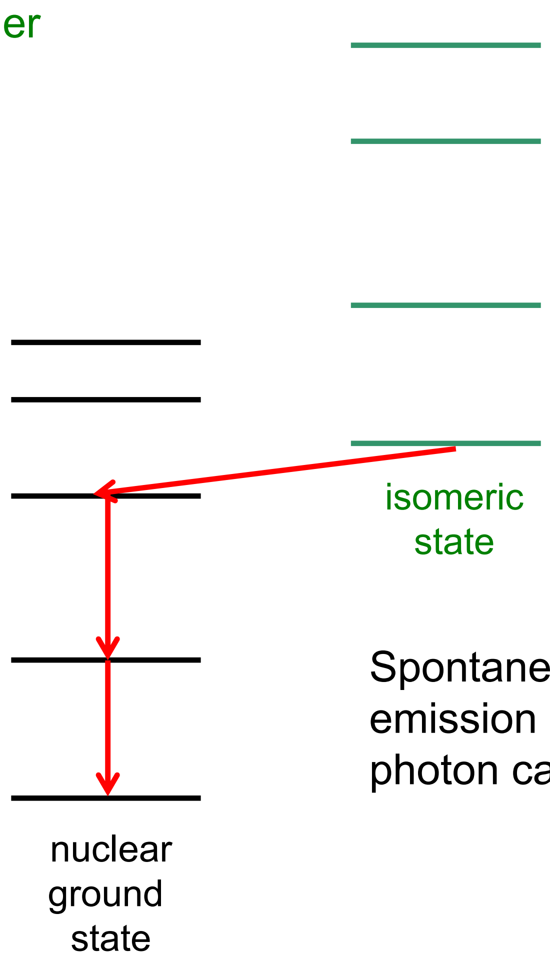
isomeric
state

Spontaneous
VUV emission

nuclear
ground
state

Detection of the nuclear excitation (III): decay via the electronic bridge

Schematic of electronic energy levels
with nuclear ground state and isomer

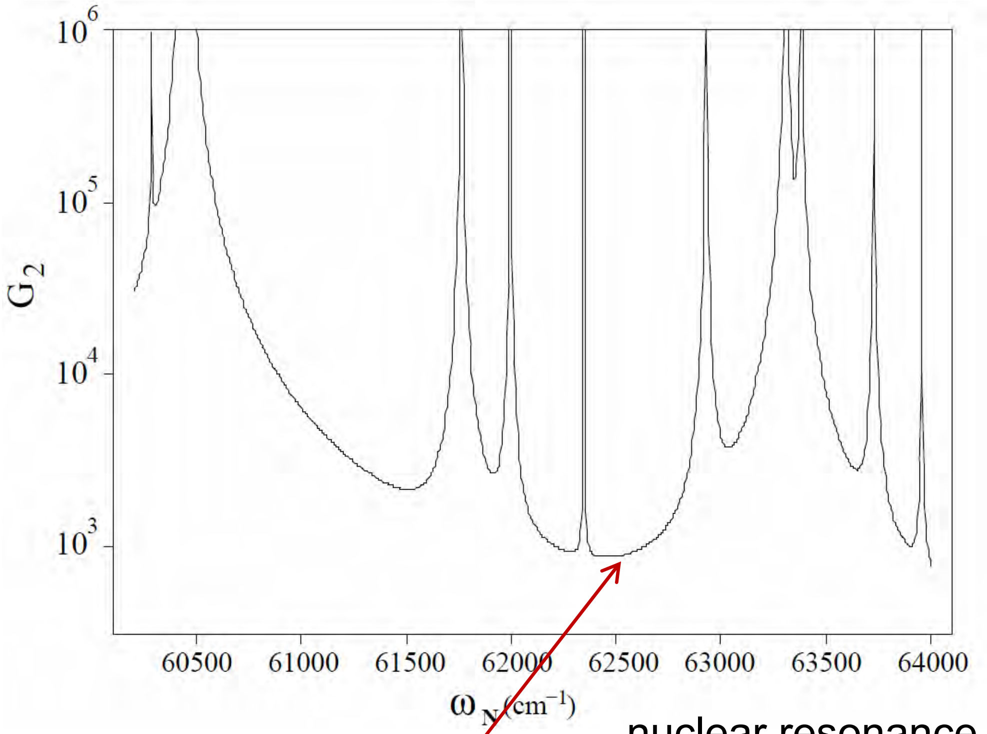


Spontaneous
emission of a characteristic
photon cascade

Two-photon electronic bridge excitation rate

S. G. Porsev, V. V. Flambaum, E. Peik, Chr. Tamm, Phys. Rev. Lett. **105**, 182501 (2010)

Based on ab-initio calculations: 10 relevant even-parity states with J=3/2 or 5/2



Prop. excitation rate

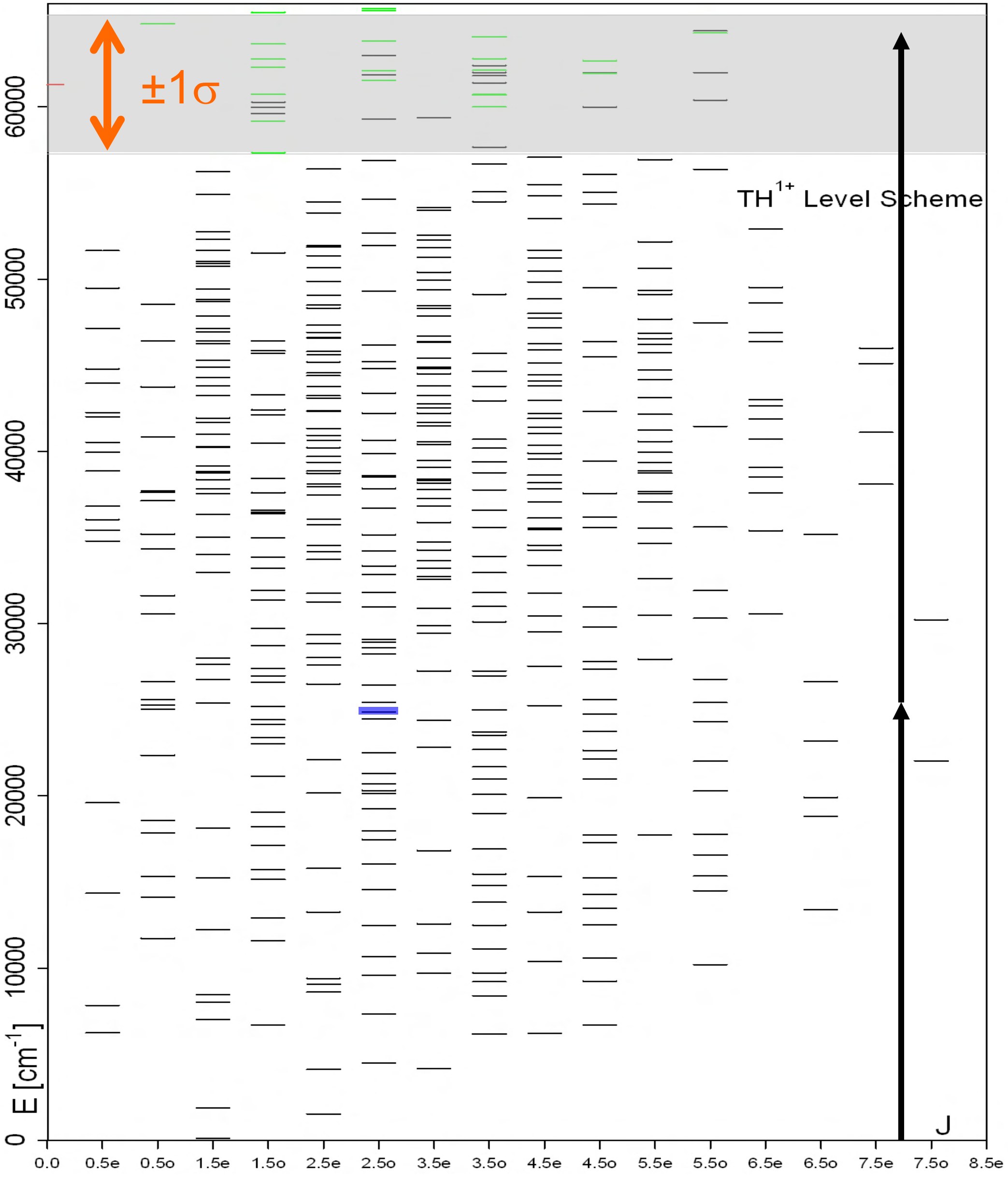
nuclear resonance frequency

→ predicted excitation rate in the range of 10 s^{-1} with a pulsed Ti:Sa laser at GHz resolution.

Experimental data on the Th^+ level structure in this range was not available.

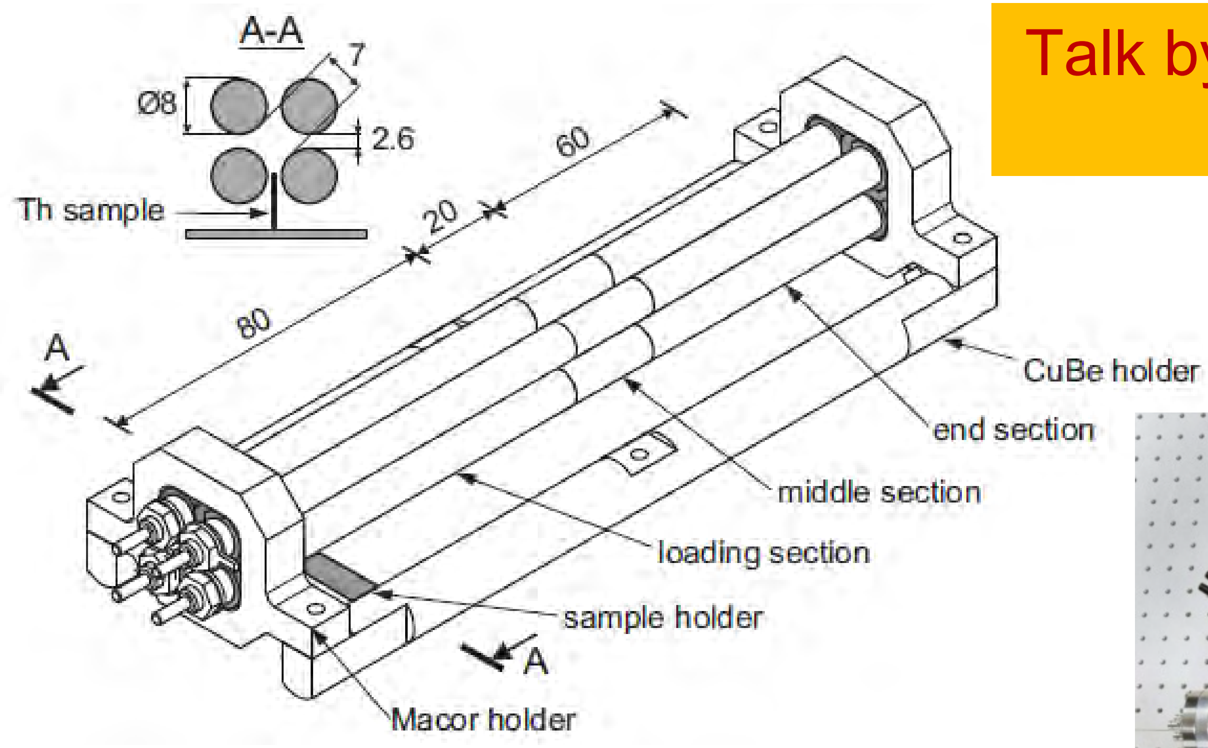
Electronic level structure of Th⁺ (as tabulated)

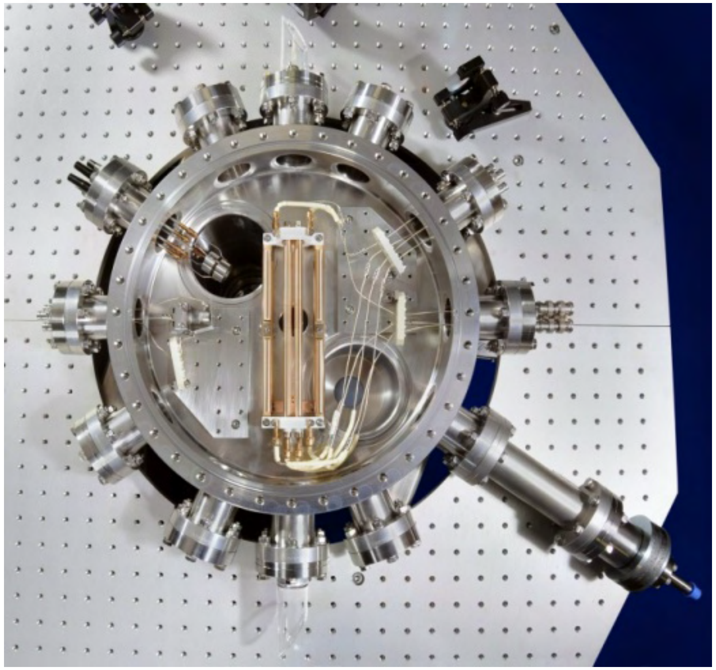
- Levels in the search range only incompletely known.
- Search range well covered by THG of Ti:Sa laser



Laser spectroscopy of trapped Th⁺ ions

Talk by O.A. Herrera-Sancho tomorrow



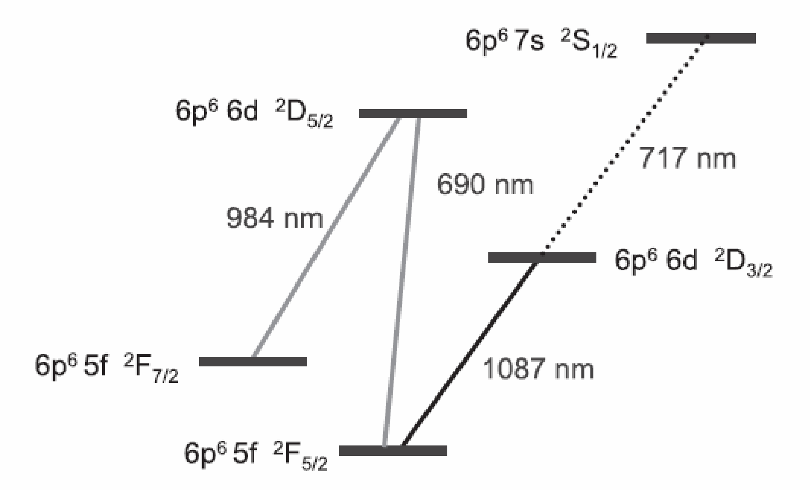


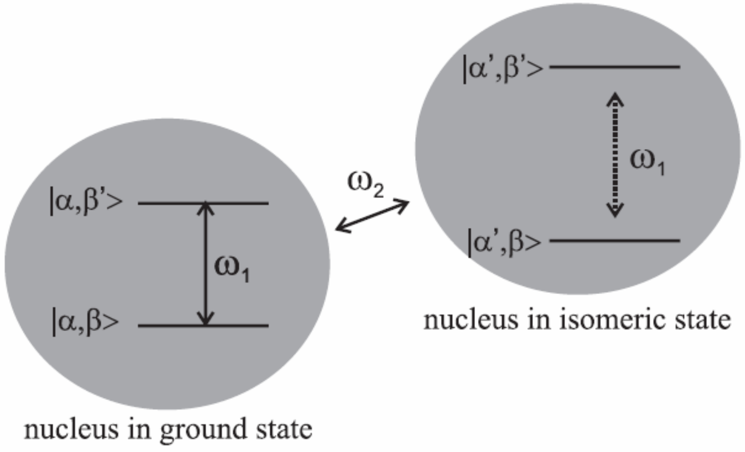
Linear Paul trap with laser ablation loading,
about 10^6 trapped Th^+ ions with buffer gas cooling

K. Zimmermann, M.V. Okhapkin, O.A. Herrera-Sancho, E. Peik
Appl. Phys. B, **107**, 883 (2012)

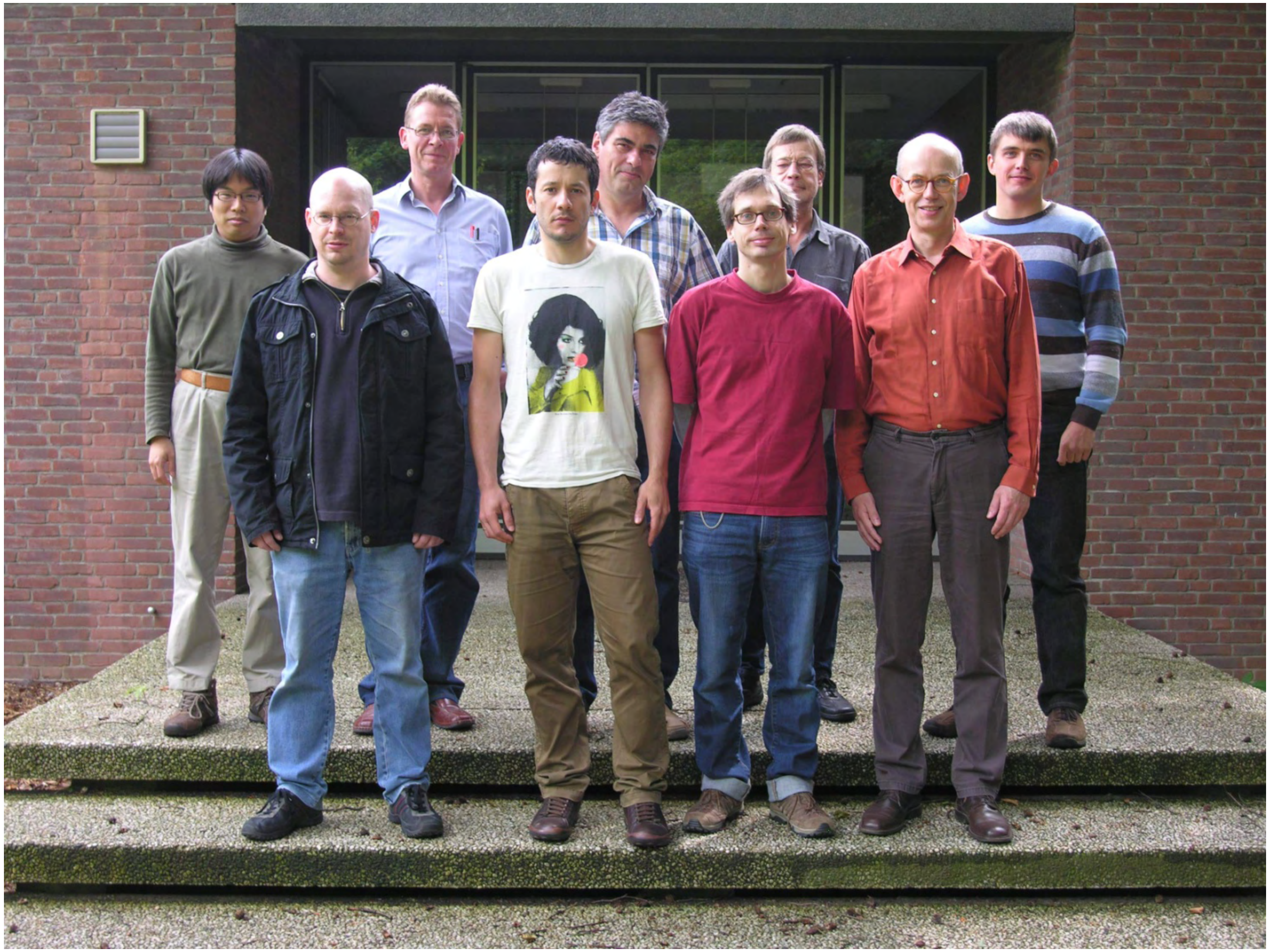
Highly precise optical nuclear clock with trapped $^{229}Th^{3+}$

- Direct laser cooling or sympathetic cooling with $^{232}Th^{3+}$ (demonstrated at Georgia Tech)
- Single-photon VUV laser excitation (e.g. 5th harmonic of a Ti:Sa laser)
- Electronic-nuclear double resonance for detection of the nuclear state





Working Group: Optical Clocks with Trapped Ions at PTB



A. Yamaguchi
A. Hoppmann
B. Lipphardt
O.A. Herrera-Sancho
M.V. Okhapkin
N. Nemitz
Chr. Tamm
E. Peik
N. Huntemann

Theory:
A.V. Taichenachev, Inst. Las. Phys. RAS Novosibirsk
V.I. Yudin, Inst. Las. Phys. RAS Novosibirsk
S.G. Porsev, Petersburg Nucl. Phys. Inst.
V.V. Flambaum, Univ. New South Wales

Funding:
DFG
QUEST
DAAD, TEC
Humboldt-Foundation

PTB