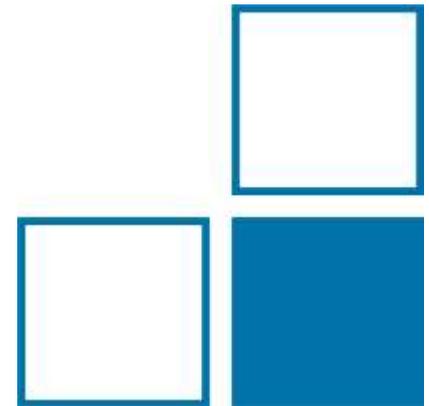
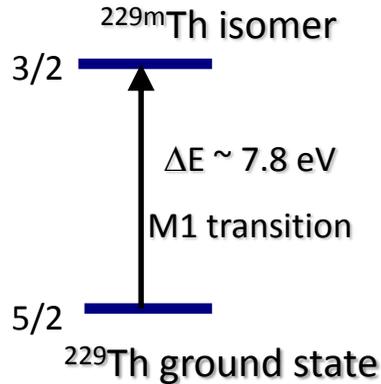


# Search for the low-energy optical transition in $^{229}\text{Th}$ .

M. Okhapkin, D.-M. Meier, J. Thielking,  
P. Glowacki, E. Peik





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

✓  $\gamma$  - spectroscopy of two decay cascades from the 71.82-keV-level.

Isomer energy: VUV – range –  $7.8 \pm 0.5 \text{ eV}$

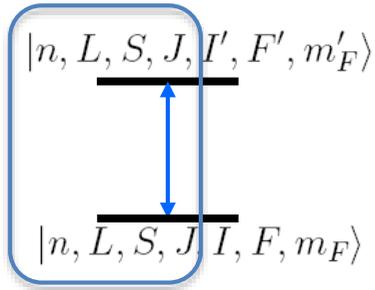
B. R. Beck et al. (LLNL), PRL 98,142501 (2007).

✓ Deexcitation of the isomer is observed in the recoil experiment (unfortunately the energy of the isomer is not derived). LMU: Direct detection of the  $^{229}\text{Th}$  nuclear clock transition. L. von der Wense et. al. Nature17669 (2016).

✓ Proposal for nuclear optical clocks E. Peik and Chr. Tamm, Europhys. Lett. **61**, 181 (2003)

- The nuclear transition frequency is independent of the shifts depending only on the electronic quantum numbers.
- More stable against external perturbations due to small dimensions of nucleus in comparison with atoms.

# Th 229: proposal for nuclear optical clocks



Nuclear and total-angular-momentum quantum numbers ( $I, F, m_F$ ) can change, purely electronic quantum numbers ( $n, L, S, J$ ) remain constant.

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

**Influence of the electron shell to the nuclear transition:** Frequency shifts that depend of ( $n, L, S, J$ ) are common for both levels. The nuclear transition frequency is independent of the shifts depending only on the electronic quantum numbers.

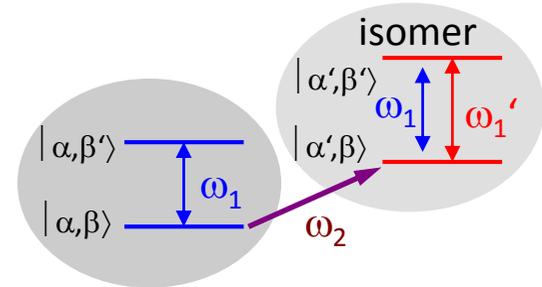
**Holds for:** Scalar quadratic Stark shift, including the effects of static electric fields, collisions, blackbody AC Stark shift

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

Hyperfine Stark shift: expected:  $\approx 10^{-19}$

Linear Zeeman shift: use component  $m_F = 0 - 0$  or a pair of stretched states (diff. shift of 4 kHz/mT)

Doppler shift: use ion trapping and laser cooling



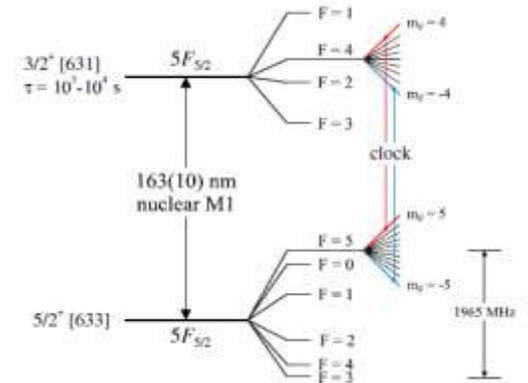
- Cryogenic ion trap ( $\text{Th}^{3+}$  is very reactive).
- Sympathetic cooling of  $\text{Th}^{3+}$

Frequency standard based on the pair of stretched hyperfine states:  $5F_{5/2}$ ,

$|I_g=5/2, F=5, m_F=\pm 5\rangle \leftrightarrow |5F_{5/2}, I_m=3/2, F=4, m_F=\pm 4\rangle$ .

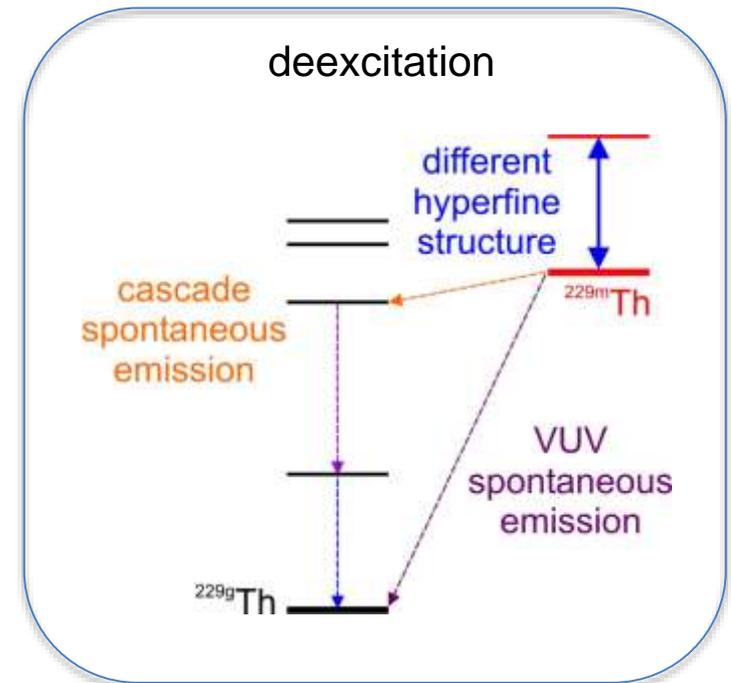
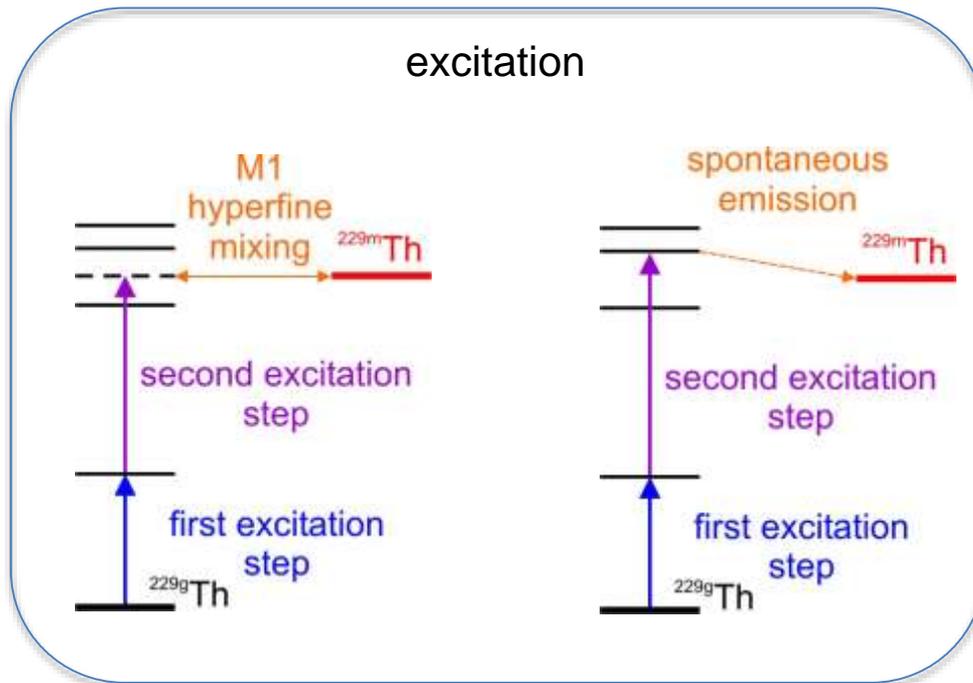
Systematic shift suppression allows clock performance with a total fractional inaccuracy approaching  $1 \times 10^{-19}$ .

C.J. Campbell et al. PRL 108, 120802 (2012).



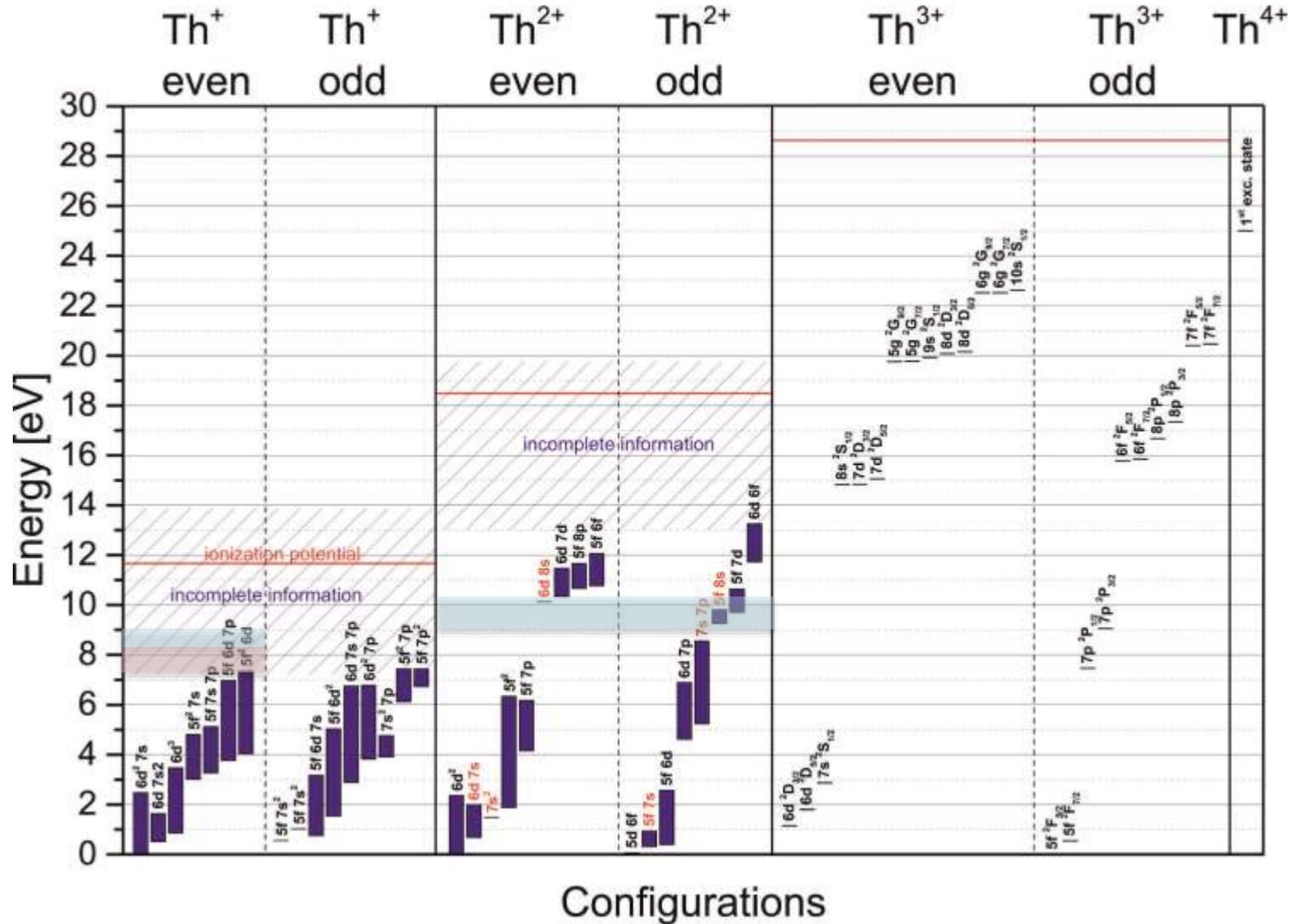
Our search method: Two-photon electronic bridge (or NEET) excitation

- uses the electron shell as an „antenna“ to enhance the nuclear excitation rate
- does not require the use of a widely tunable VUV laser



E.V. Tkalya, Usp. Fiz. Nauk 46, p. 315 (2003);  
S.G. Porsev, V.V. Flambaum, PRA, 042516 (2010);  
S.G. Porsev, V.V. Flambaum, E. Peik, Chr. Tamm, PRL, 105, 182501 (2010).

# Electronic levels of Th charged states



## Electronic bridge enhancement:

$$K \approx (E_{M1}^2 \Delta v_{el}) / (\Delta v_{ne}^2 \Delta v_{nu}) \sim 10^3 \dots 10^4$$

(if the isomer energy is located between electronic levels of Th<sup>+</sup>, or higher for small detunings)

Assumption:  $\Delta v_{ne} \gg \Delta v_{el} \gg \Delta v_{nu}$

$\Delta v_{el} \sim 10^7$  Hz – electronic transition width,

$\Delta v_{nu} \sim 10^{-3}$  Hz – width of the isolated nuclear transition,

$\Delta v_{ne} \sim 3 \times 10^{12}$  Hz – frequency difference between nuclear and electronic transitions,

$E_{M1} \sim 10^9$  Hz – magnetic dipole coupling energy between electron and nucleus (in Hz)

E.V. Tkalya, Phys. Uspekhi 46, 315 (2003)

## Excitation probability:

Laser parameters:  $f = 1$  kHz;  $P_p \sim 500$  W,  $\tau_p \sim 15$  ns,  $\Delta v \sim 1$  GHz;

Beam dia. 0.5 – 0.8 mm  $\rightarrow I_p \geq 10^9$  W/m<sup>2</sup>  $\rightarrow I_p(v) \sim 1$  W/(m<sup>2</sup>·Hz);

$\gamma \sim 10^{-3} \sim \Delta v_{nu}$

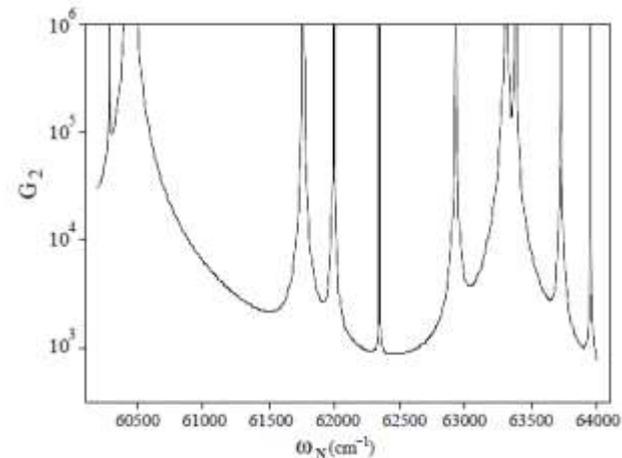
$$P \approx 2\pi^3 c^3 \gamma / (h\omega^3) \times \tau_p I_p / (c\Delta v) \sim 5 \times 10^{-7} [1/(\text{ion} \times \text{pulse})]$$

Ions in the beam  $\sim 2 \times 10^4$  (<sup>229</sup>Th)

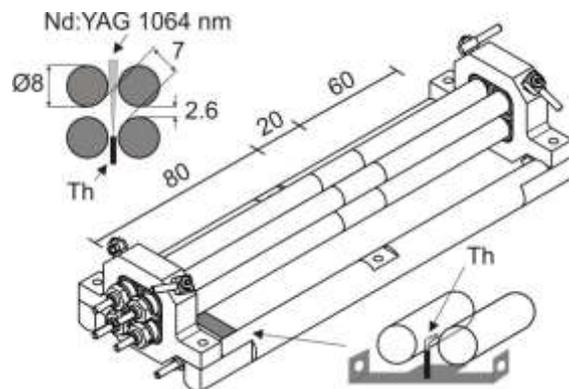
Enhancement factor (min)  $K \sim 10^3 \rightarrow (\gamma^* \sim \gamma \times 10^3)$ ;

Detection efficiency (PMT solid angle and quantum efficiency)  $\geq 3 \times 10^{-4}$ ;

$P \sim 5 \times 10^{-3}$  photons/pulse  $\geq 1000$  pulses average is required



S. G. Porsev, V.V. Flambaum, E. Peik,  
Chr. Tamm. PRL 105, 182501 (2010) based  
on *ab initio* calculations



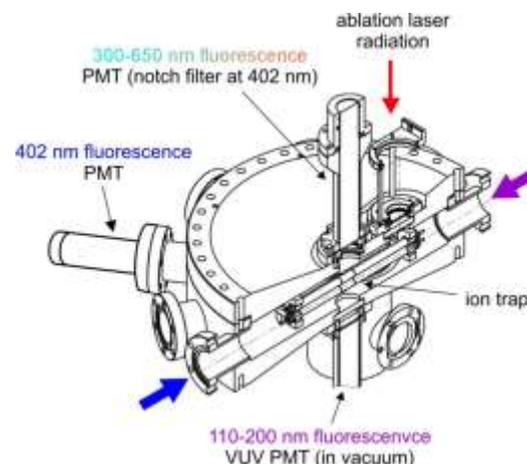
## \* Trap parameters

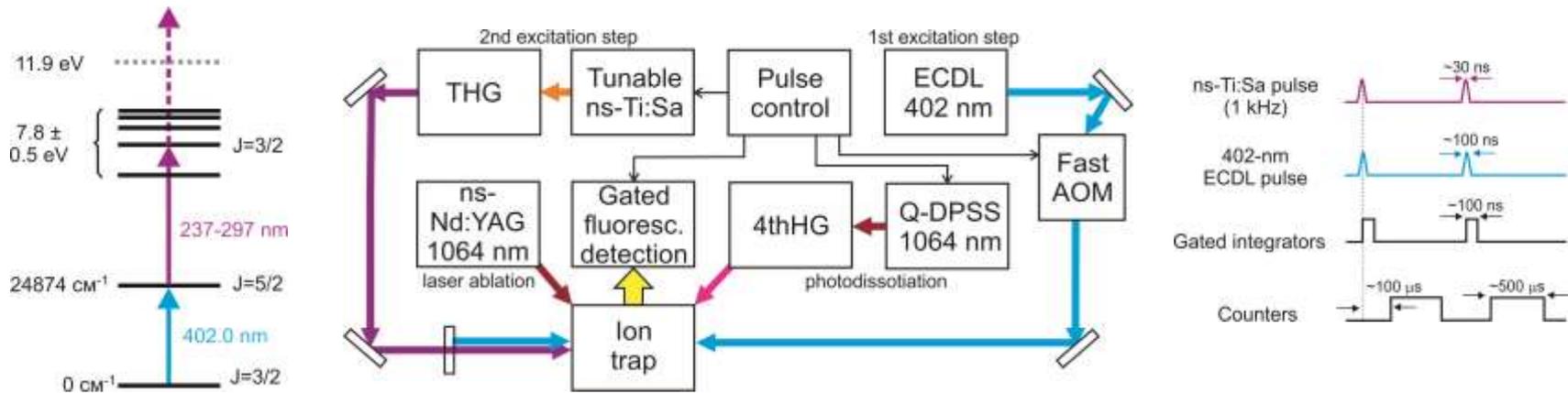
- \* RF voltage 500...900 V at frequency of  $\sim 2$  MHz;
- \* Transverse secular frequency of  $\sim 155...280$  kHz;
- \* Pseudopotential depth of 15...44 eV;
- \* Capacity to store up to  $10^6$  ions;
- \* Buffer gas cooling and quenching with Ar at 0.1 Pa pressure.

\* **Ablation:** Nd:YAG laser, single pulse mode, 10 ns, 1 mJ pulse energy;  
Loading efficiency for multiple loadings from  $\text{Th}(\text{NO}_3)_4$  solution is  $\sim 10^{-6}$  i.e. the ratio of the number of stored ions over multiple loadings to the number of Th atoms deposited on the substrate ( $\sim 10^{14}$  Th atoms on W substrate).

\* **Photodissociation:** 4th harmonics of Nd:YAG (266 nm), 1 ns pulse, pulse energy of 10 mJ, 1 kHz repetition rate.  
The storage time observed for  $\text{Th}^+$  is  $\sim 1000$  s, limited by the formation of  $\text{ThCH}_2^+$  molecules.

\* **Detection:** PMTs in visible and VUV ranges



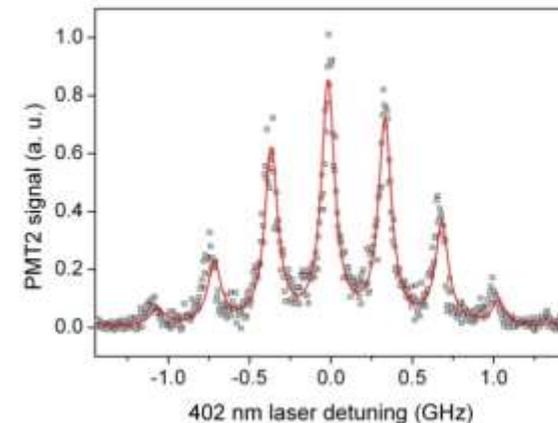


## ✓ Pulse mode:

- rep. rate 1 kHz
- pulse duration of ECDLs: ~ 70-100 ns.
- pulse duration of ns-TiSa laser: ~ 20-30 ns; THG pulse power: ~ 500 W (~ 10 mW CW).
- efficient population transfer during short pulse.

## ✓ Detection:

Gated fluorescence detection. The counters gates are shifted according to expected isomer lifetime to avoid signal of long-lived electronic decay channels and LIF of optical windows. Comparison of the detected signals for both  $^{229}\text{Th}$  and  $^{232}\text{Th}$  isotopes.

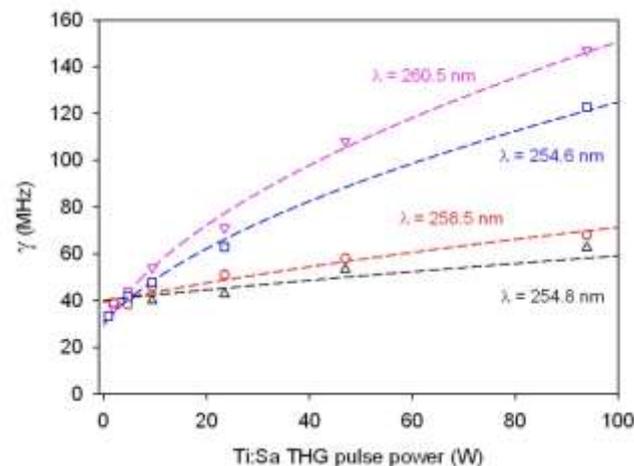
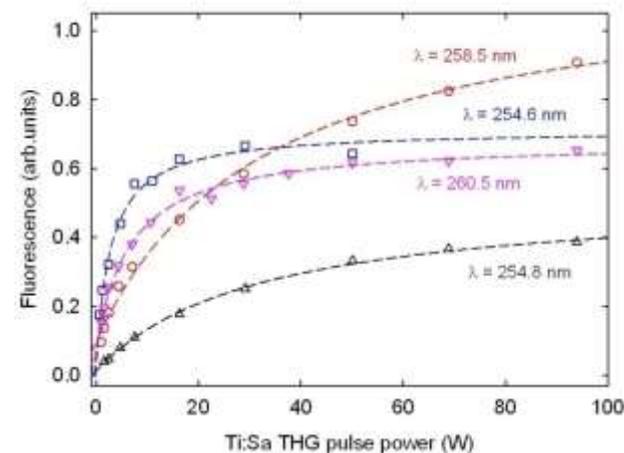


GI fluorescence signal from high-lying state

# Th<sup>+</sup> electronic levels in the isomer energy range

Levels of Th<sup>+</sup> for the isomer NEET excitation are observed with the <sup>232</sup>Th isotope.

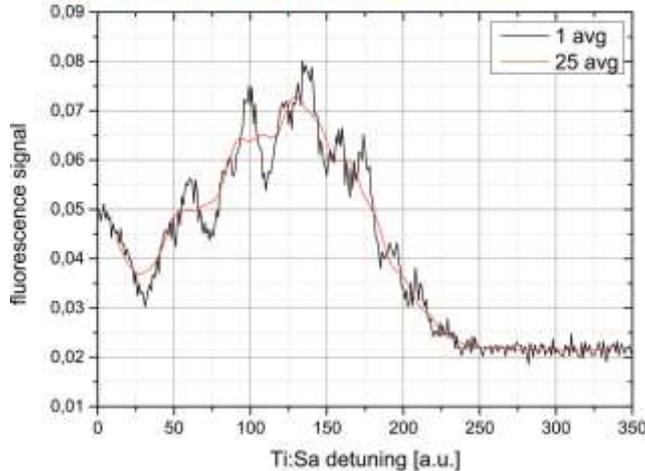
Level (cm <sup>-1</sup> )	J	Sf <sub>pulse</sub> (kW/cm <sup>2</sup> )	Level (cm <sup>-1</sup> )	J	Sf <sub>pulse</sub> (kW/cm <sup>2</sup> )
58875.5		197.9	64150.3	3/2	2.8
59387.1*		148.0	64560.4	3/2	6.95
59477.4	3/2	0.62	64813.7		0.32
59803.0		0.28	64860.4		> 460
60380.1	3/2	2.2	64920.1		20.8
60618.6	3/2	10.7	65037.7	3/2	6.5
60721.3		2.0	65144.4		1.1
61032.4	3/2	0.34	65191.1		>334
61388.0		0.1	65730.4	3/2	0.55
61428.6		0.54	65738.1		44.4
61726.3	3/2	4.0	65799.6	3/2	2.85
61963.6	3/2	0.36	65910.0		9.8
62307.2		5.6	65946.3		1.3
62373.8	3/2	63	66052.0		19.5
62477.0		8.9	66141.2		>113
62560.1		2.7	66333.7	3/2	15.9
62562.2	3/2	5.6	66558.0		0.3
62753.1		7.7	66609.0		4.2
63257.5		0.66	66702.9	3/2	64.1
63298.4		26.9	66831.1	3/2	0.28
63557.7		19.1	66855.6		14.9
64122.0		9.96	67066.2		>22



\* - observed by R. Zalubas, C.H. Corliss. J. Res. Nat. Bur. St. A78, 163 (1974)

fluorescence gated integrator signal

delay: 10 ns, width: 75 ns, 200 - 650 nm w/o 402 nm



## Scan in progress

### Problems:

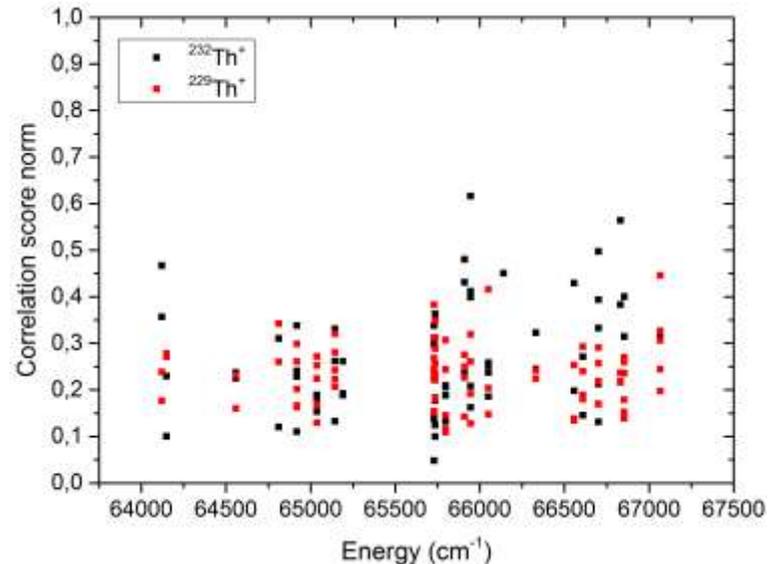
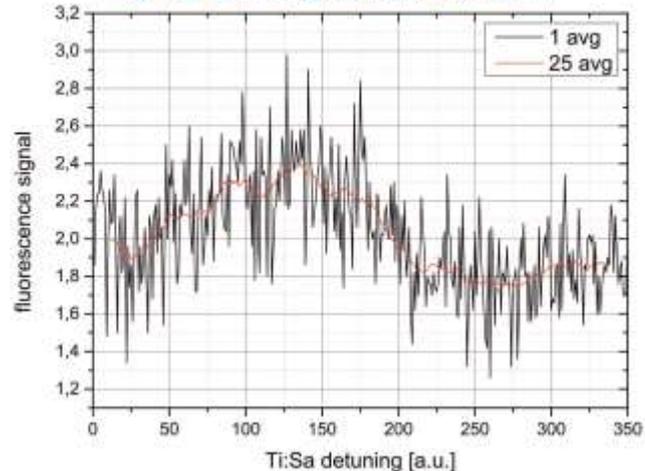
- LIF due to high power UV pulse limits detection sensitivity;
- Th<sup>+</sup> - second ionization potential lies in the 11.9...12.3 eV range. Resonant 3-photon ionization interferes with 2-photon excitation of the electronic levels.
- Long average time (~ 10<sup>2</sup> s for one data point)

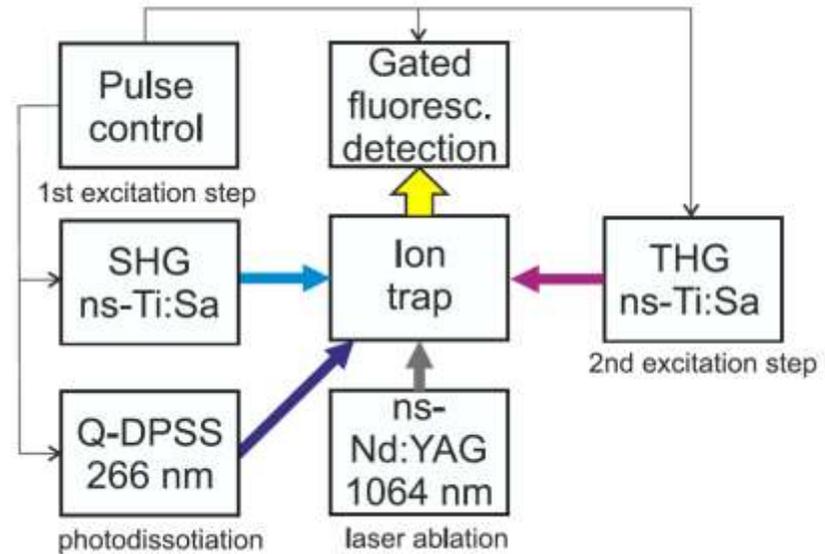
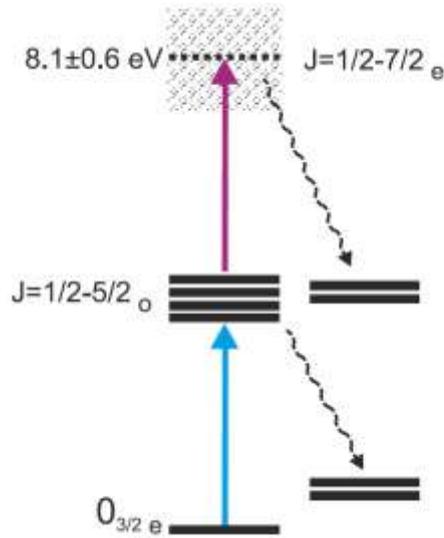
### Data analysis:

- Correlation analysis between the electronic decay signal of the gated integrator and counters signals from PMTs.
- The counters gates are delayed by 100 μs (to avoid LIF and decays from electronic states, the isomer lifetime estimated to be ≥ 1 ms).
- Comparison of the signals for <sup>229</sup>Th<sup>+</sup> and <sup>232</sup>Th<sup>+</sup>.

fluorescence counter signal

delay: 1 μs, width: 870 μs, 200 - 650 nm w/o 402 nm





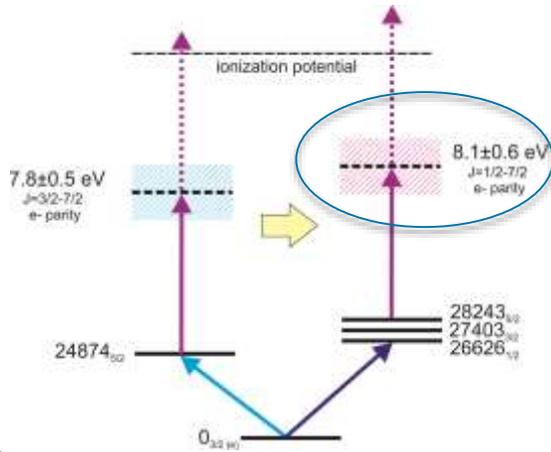
IS	$J$	Config.
25027	1/2	$7s7p+6d7s7p$
26626	1/2	$6d7s(^3D)7p^4D, ^2P$
26965	3/2	$5f(^3F)6d^2^4S, ^2P$
27403	3/2	$5f(^1D)6d^2^2D+5f(^3P)6d^2^4D$
26424	5/2	$5f(^3P)6d^2^4F, ^4D$
28244	5/2	$6d7s(^3D)7p^4F+6d^2(^3F)7p^4G$

Levels of Th<sup>+</sup> for the isomer NEET excitation are observed with the <sup>232</sup>Th isotope.

Energy [cm <sup>-1</sup> ]	Observed from intermediate state [cm <sup>-1</sup> ]							
	25027,038 26626,485		26586,273 26965,205		27403,18		24873,982 26424,481 28243,812	
	1	2	3	4	5	6	5	6
	1/2	1/2	3/2	3/2	3/2	5/2	5/2	5/2
62873,130		X		X	X			
63268,900	X	X		X	X			
63680,285	X	X		X	X		X	X
64107,508				X	X		X	X
64368,240				X	X		X	X
64442,103	X	X		X	X			
64887,795	X	X		X	X			
65753,450	X	X		X	X			
66324,519	X	X		X	X			
66388,834	X	X		X	X			
66429,640				X	X			X
66666,961	X	X			X			
67154,047	X			X	X		X	X
67177,763							X	X
67378,611							X	X
67508,637		X		X	X		X	X
67577,711				X				X
67657,301		X		X	X			
67737,621				X	X		X	X
67803,247				X				X
67843,316			X	X	X			X
67866,095					X		X	X
68033,328							X	X
68088,027	X			X	X			X
68278,648								X
68497,879					X			X
68564,189				X	X			X
68598,833				X	X			X
68752,065	X			X	X			X
68812,644				X	X			X
68898,666				X	X			X
68913,767					X			
68921,301					X			X
69381,631								X
69523,570								X
69582,982								X
69588,677								X
70036,553								X

Level	<i>J</i>	IS	Level	<i>J</i>	IS
62873.1(2)	1/2	2-4	67803.2(2)	5/2 (3/2)	3;6
63268.9(2)	1/2	1-4	67843.3(2)	5/2 (3/2)	3;4;6
63680.3(2)	3/2	1-6	67866.1(2)	5/2 (3/2)	4-6
64107.5(2)	5/2	3-6	68033.3(3)	7/2	5;6
64368.2(2)	5/2	3-6	68088.0(2)	3/2	2-4;6
64442.1(2)	1/2	1-4	68278.6(3)	7/2	6
64887.8(2)	1/2	1-4	68497.9(2)	5/2 (3/2)	4;6
65753.5(2)	1/2	1-4	68564.2(2)	5/2 (3/2)	3;4;6
66324.5(2)	1/2	1-4	68598.8(2)	5/2 (3/2)	3;4;6
66388.8(2)	1/2	1-4	68752.1(2)	3/2	2-4;6
66429.6(2)	5/2	3;4;6	68812.6(2)	5/2 (3/2)	3;4;6
66667.0(2)	1/2	1;2;4	68898.7(2)	3/2-5/2	3;4;6
67154.0(2)	3/2	1;3-6	68913.8(3)	1/2-5/2	4
67177.8(2)	7/2	5;6	68921.3(2)	3/2-5/2	4;6
67378.6(3)	7/2	5;6	69381.6(3)	3/2-7/2	6
67509.6(2)	3/2	2-6	69523.6(3)	3/2-7/2	6
67577.7(3)	5/2 (3/2)	3;6	69583.0(3)	3/2-7/2	6
67657.3(2)	1/2	2-4	69588.7(3)	3/2-7/2	6
67737.6(2)	5/2 (3/2)	3-6	70036.6(3)	3/2-7/2	6

## Th<sup>+</sup>



### ✓ Excitation:

The first step is provided by SHG radiation of Ti:Sa; the second excitation step: THG of Ti:Sa.

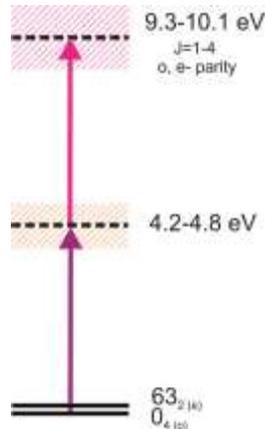
### ✓ Advantages:

High density of the levels.

### ✓ Disadvantages:

High ionization rate due to the resonant 3-photon ionization (limits the excitation power).

## Th<sup>2+</sup>



### ✓ Excitation:

THG radiation of 2 Ti:Sa lasers.

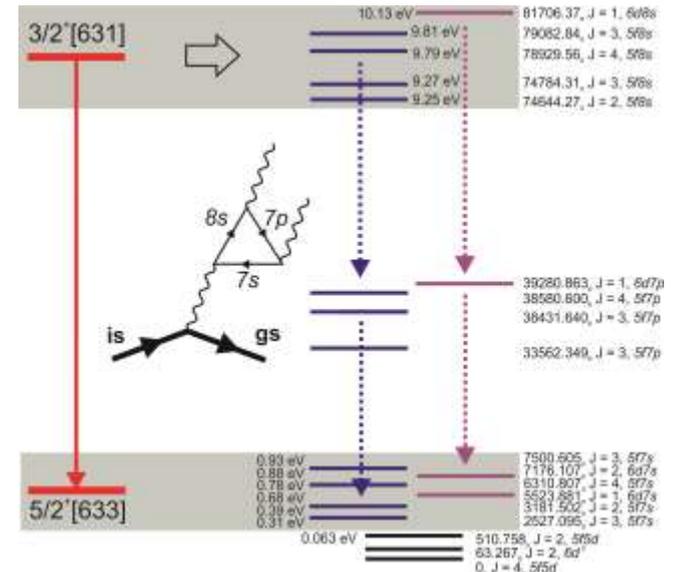
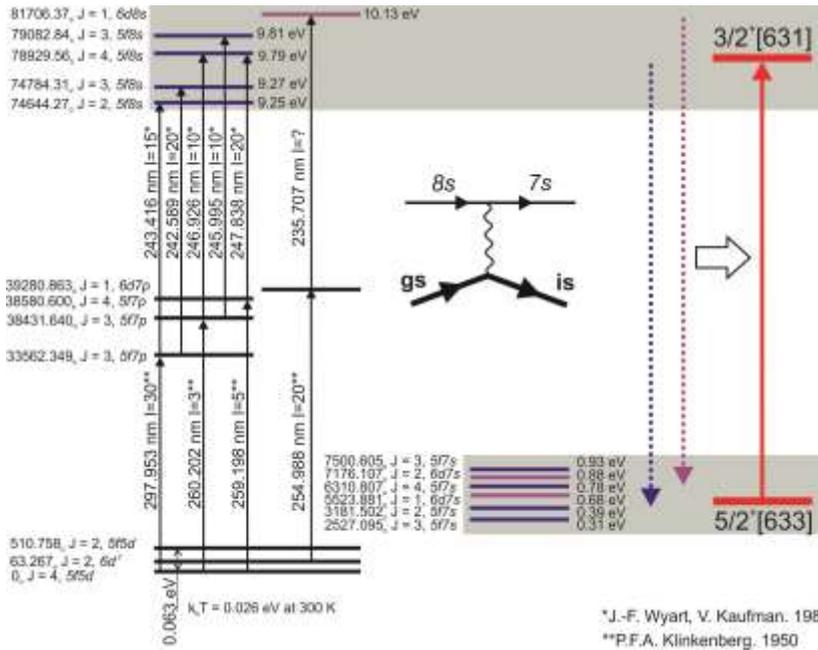
### ✓ Advantages:

Th<sup>2+</sup> ionization potential is  $\geq 18.3$  eV (avoids 3-photon ionization).

8s-electron configurations available for the NEET excitation.

### ✓ Disadvantages:

Relatively lower density of the electronic states in the range of search.

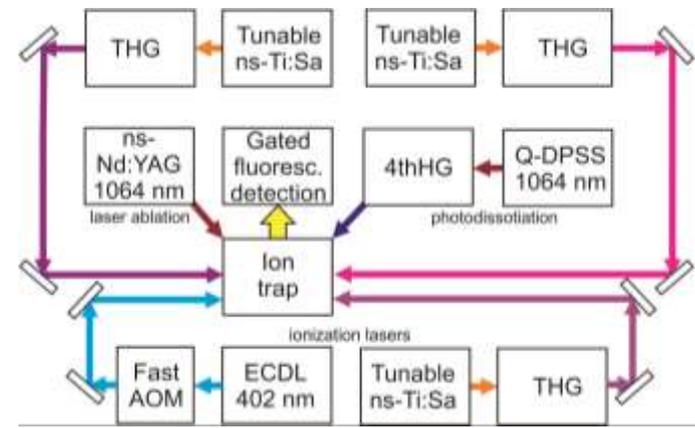


## Excitation:

THG of ns- Ti:Sa lasers;  
 rep. rate 1 kHz;  
 pulse duration 15 -20 ns;  
 pulse power  $\sim 10^2 - 10^3$  W.

## Trap loading:

Resonant 3-photon ionization of  $\text{Th}^+$ .

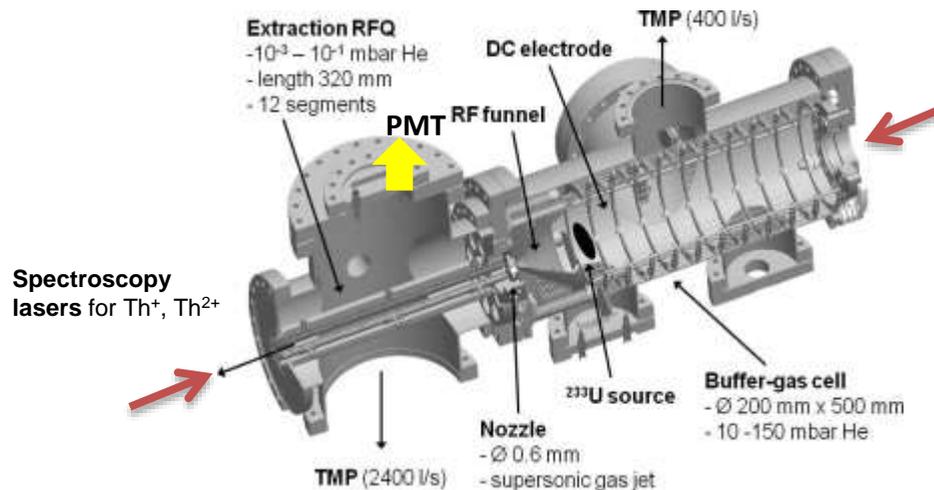


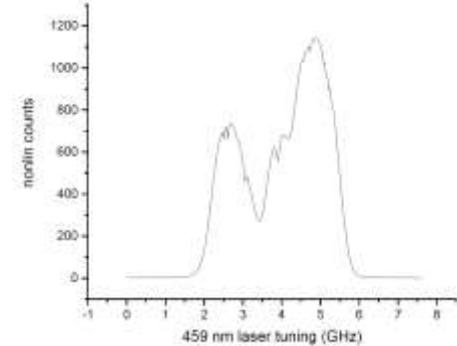
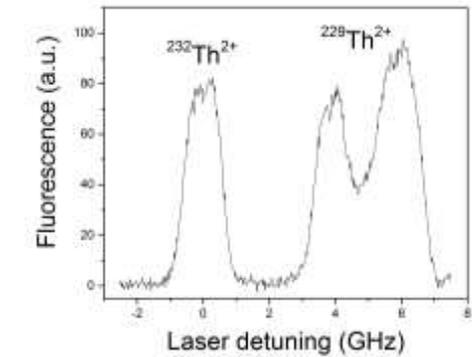
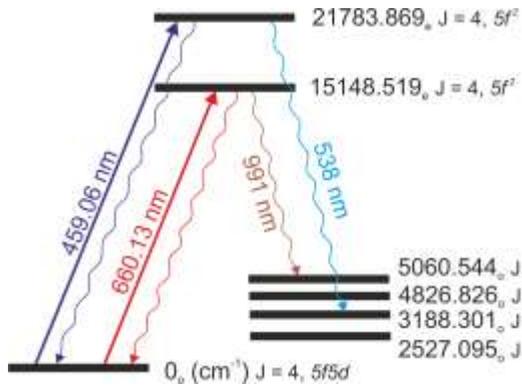
# Search of HFS of $^{229m}\text{Th}$ isomer

- Searching of the isomer energy
- Optical excitation of the isomer
- Development of lasers for the nuclear optical clock
- Development of optical clocks based on the ion trap technique and Th doped crystals

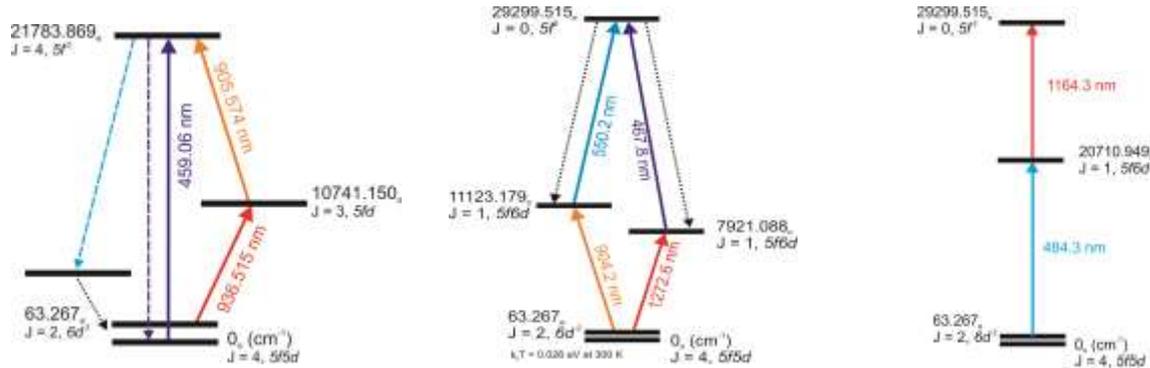
## PTB/LMU experiment:

Searching of the hyperfine structure of the isomer in different charge states of Th.





Palmer Engleman 1983 Atlas of the thorium spectrum



- ✓ Expected FWHM of the resonances is about 70 MHz;
- ✓ Background-free detection;

Requires signal selection in the case when the splitting of the intermediate state HFS components is  $\sim kv$

# Coming soon

- Tests of the NEET excitation in  $\text{Th}^{2+}$ ;
- Extension of the scanning range in  $\text{Th}^+$ .

## Thank you for your attention!



Group leader: E. Peik

Ion trap experiment / nuClock

Scientist: M. Okhapkin

Ph.d. students: D. Meier

J. Thielking

Experiment with crystals / nuClock

Scientist: P. Glowacky

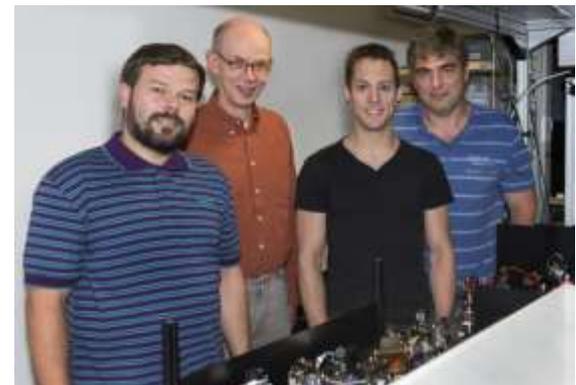


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[www.ptb.de](http://www.ptb.de)



## ➤ Th<sup>+</sup>:

- Small extraction efficiency from <sup>233</sup>U source, hence unknown isomer lifetime\*.
- + Laser system is already existing including Doppler-free two photon excitation.
- + HFS has been investigated and splitting factors are known.

## ➤ Th<sup>2+</sup>:

- + Extraction efficiency factor from <sup>233</sup>U source is 20 times higher compared to Th<sup>+</sup>.
- + Isomer lifetime > 60 s\*\*.
- + Suitable transitions in the visible range are available. Single photon excitation is developed.

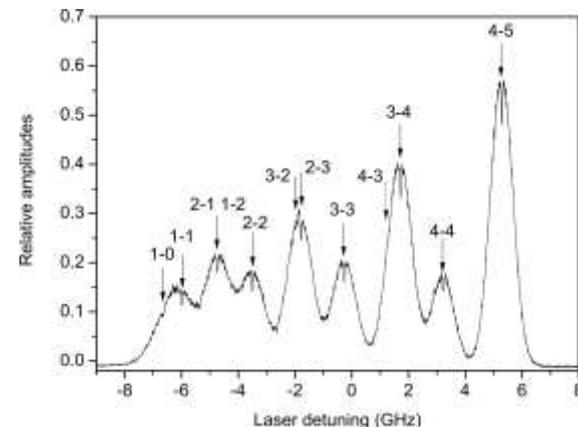
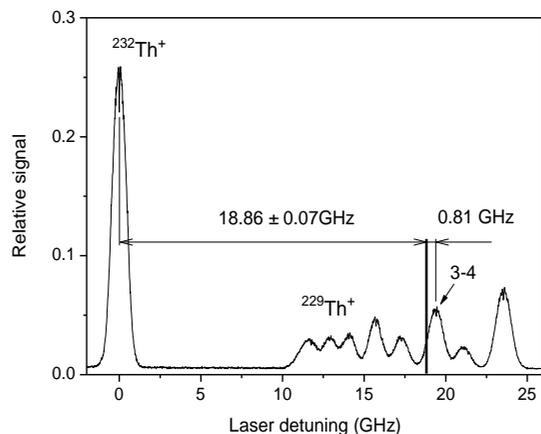
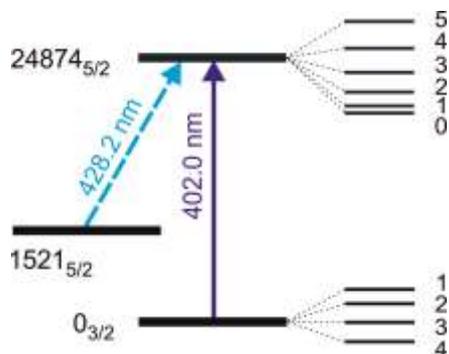
## ➤ Th<sup>3+</sup>:

- + Highest initial amount from <sup>233</sup>U source.
- Transitions in the IR and UV range.
- Production and storage in PTB experiments are not tested.

\* L. v. d. Wense et al., *Eur. Phys. J. A* **51**, 28 (2015).

\*\* L. v. d. Wense et al., *Nature* 533.7601 (2016): 47-51.

# Hyperfine structure of 402 nm $^{229}\text{Th}^+$ line



Level [cm <sup>-1</sup> ]	A factor [MHz]	B factor [MHz]	Ref.
0	-444.2(1.9) -444.2(3.4)	303(6) 308.4(12.7)	* **
24874	488.9(3.7) 489.2(3.7)	-412.8(15.5) -408.8(17.9)	**/* **



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

$^{229\text{m}}\text{Th}$  Isomer

$3/2^+ [631]$

$\Delta E = 7.8 \text{ eV}$

M1 transition  
 $\tau \approx 1000 \text{ s}$

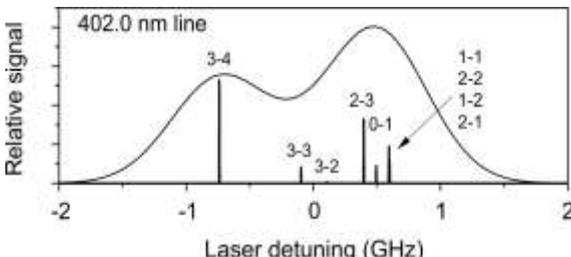
$5/2^+ [633]$

$^{229}\text{Th}$  Ground State

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

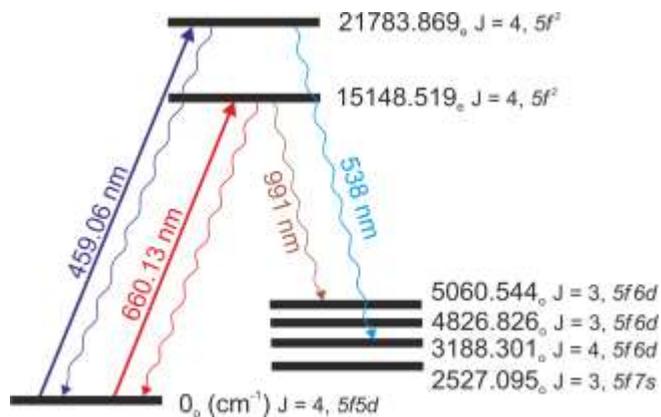
Expected HFS of the isomer

$A_{is}/A_{gs} \sim \mu_{is}/\mu_{gs}$ ,  $B_{is}/B_{gs} \sim Q_{is}/Q_{gs}$



\* W. Kälber et al., Z. Phys. A - Atomic Nuclei **334**, 103 (1989).

\*\* M.V. Okhapkin et al., Phys. Rev. A **92**, 020503 (2015)

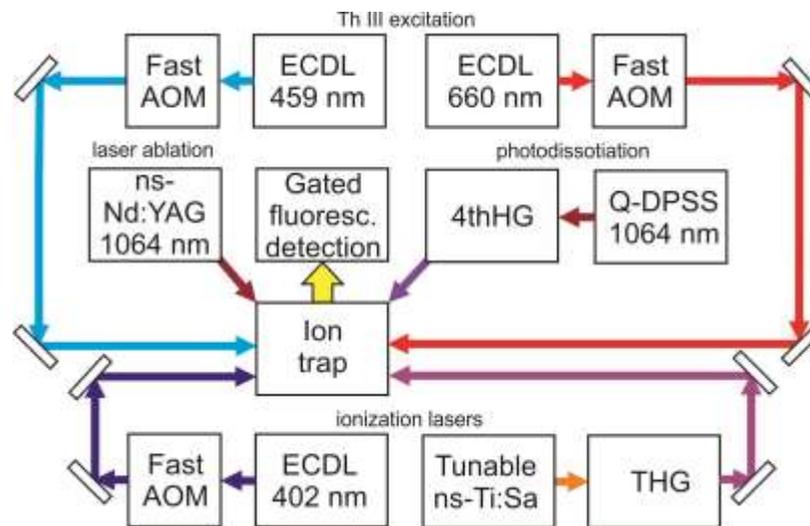
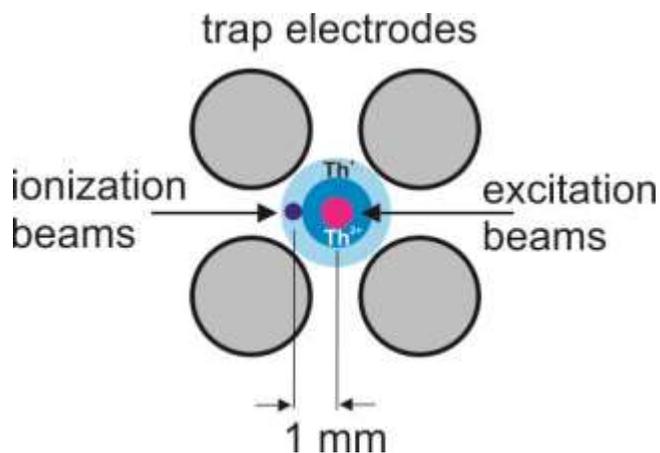


Palmer Engleman 1983 Atlas of the thorium spectrum

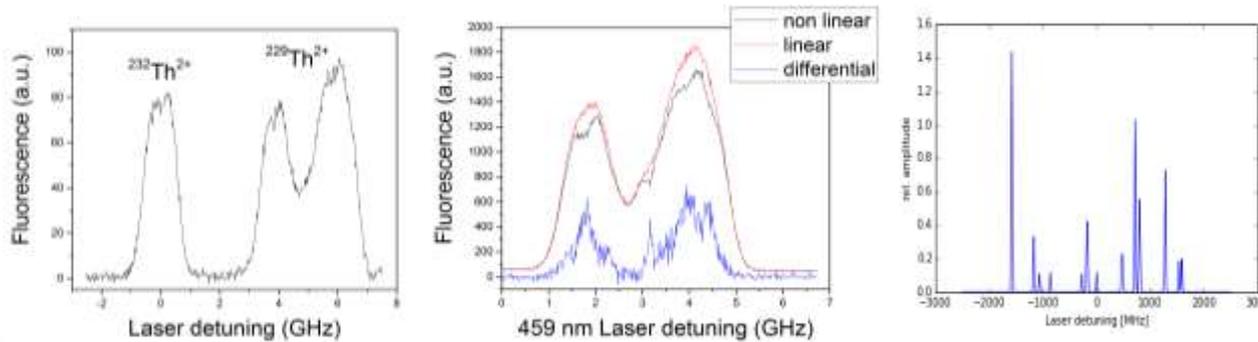
Th<sup>2+</sup> storage time is  $\tau \sim 10^3$  s (loss rate of  $\sim 1$  ion/s due to chemical reactions).

Steady state amount of Th<sup>2+</sup> ions in the trap is  $\sim 10^3$  (ionization rate = ion loss rate).

The trap operation time with one loading of <sup>229</sup>Th<sup>+</sup> is  $> 10^5$  s.



## $\text{Th}^{2+}$ 459 nm line. Detection at 538 nm.

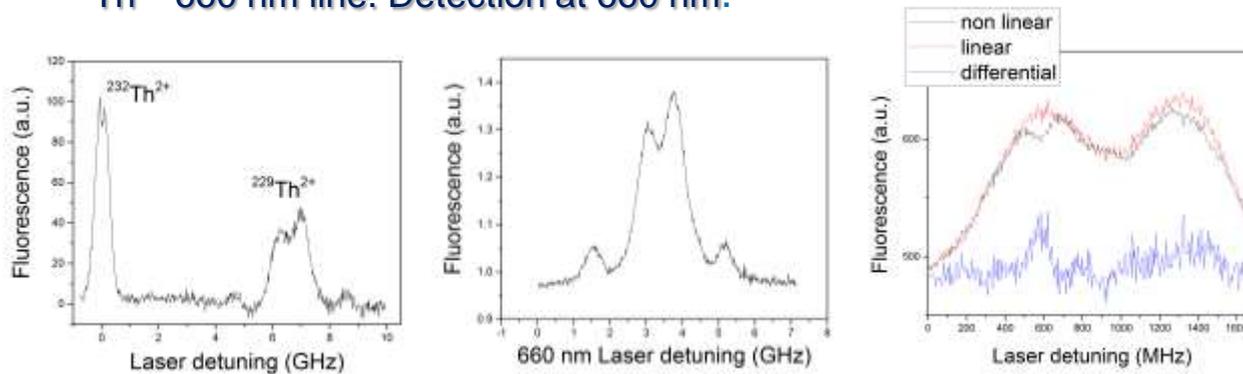


Positive isotope shift of  $^{229}\text{Th}^{2+}$  relatively to  $^{232}\text{Th}^{2+}$  is  $5.3 \pm 0.2$  GHz.

High signal-to-noise ratio due to background-free detection at 538 nm.

Hyperfine structures of  $^{229\text{m}}\text{Th}$  and  $^{229}\text{Th}$  expected to be superimposed.

## $\text{Th}^{2+}$ 660 nm line. Detection at 660 nm.

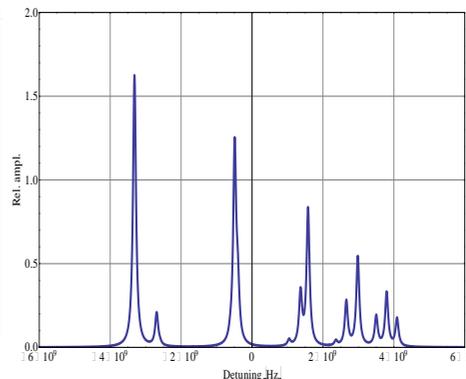
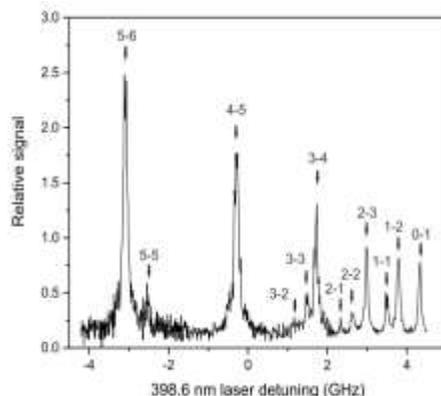
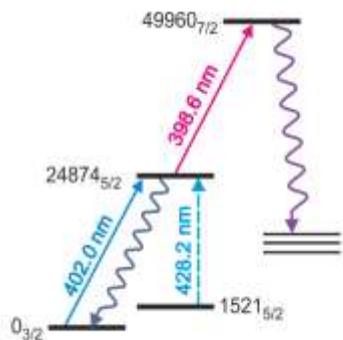


Positive isotope shift of  $^{229}\text{Th}^{2+}$  relatively to  $^{232}\text{Th}^{2+}$  is  $6.7 \pm 0.3$  GHz.

Disadvantage: Background-free detection is in the IR range. Detection at 660 nm is influenced by laser stray light.

## $\text{Th}^+$

- ✓ FWHM of the resonances is about 40 MHz;
- ✓ Background-free detection in the range of 300 nm;



Estimated HFS factors:

$$A_{24873} = 489 \times 10^6 \text{ *};$$

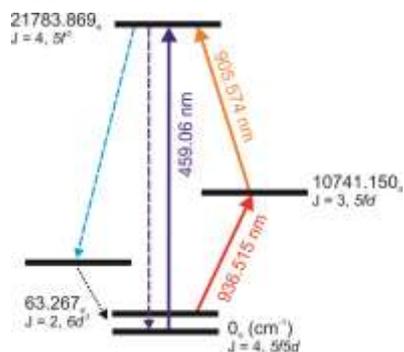
$$B_{24873} = -409 \times 10^6 \text{ *};$$

$$A_{49960} = -10 \times 10^6;$$

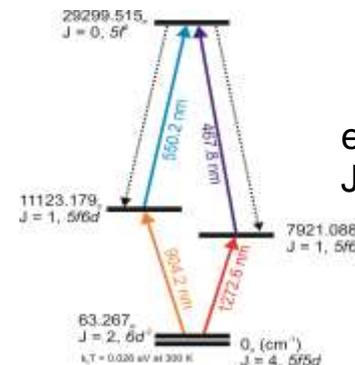
$$B_{49960} = -1100 \times 10^6$$

\* - fixed according to the measurements of the 402 nm line

## $\text{Th}^{2+}$



- ✓ Expected FWHM of the resonances is about 100 MHz;
- ✓ Background-free detection at 459 or 540 nm;
- ✓ The common upper level for the single photon excitation at 459 nm and for the two photon excitation.



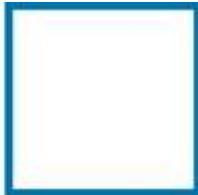
excitation of  $J=0$  state

Requires signal selection in the case when the splitting of the intermediate state HFS components is  $\sim kv$

# Coming soon

- First tests with the LMU system;
- Tests of the NEET excitation in  $\text{Th}^{2+}$ ;
- Extension of the scanning range in  $\text{Th}^+$ .

## Thank you for your attention!



Group leader: E. Peik  
Ion trap experiment / nuClock  
Scientist: M. Okhapkin  
Ph.d. students: D. Meier



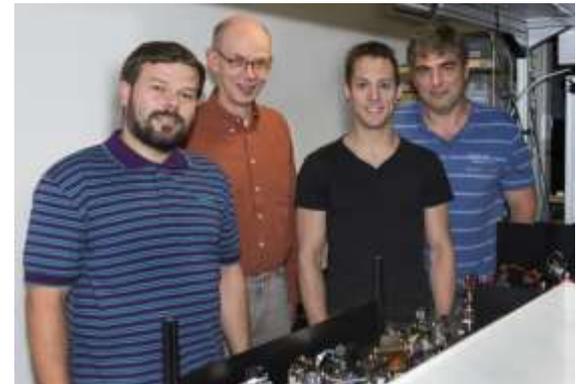
J. Thielking  
Experiment with crystals / nuClock  
Scientist: P. Glowacky



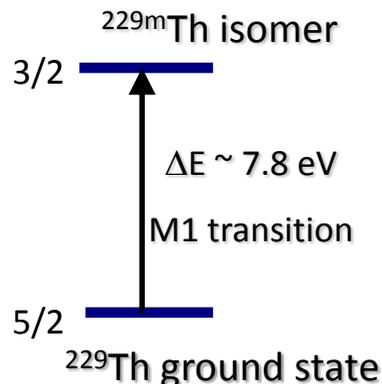
**Physikalisch-Technische Bundesanstalt  
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E-Mail: [maksim.okhapkin@ptb.de](mailto:maksim.okhapkin@ptb.de)

[www.ptb.de](http://www.ptb.de)



- ✓ Nuclear optical clocks and optical excitation of the isomer.
- ✓ Experiments for the optical excitation of the isomer in PTB:
  - Experimental setup for the excitation of the isomer in thorium.
  - Search of the isomer energy with trapped  $\text{Th}^+$  ions.
  - Extended range search of the isomer excitation with  $\text{Th}^{2+}$  ions.
- ✓ NuClock project: investigation of the hyperfine structure of thorium and search of the isomeric state HFS in the experiment with recoil nuclei (in collaboration with LMU).
  - Investigation of the HFS in  $\text{Th}^+$  and  $\text{Th}^{2+}$ .
  - Experimental setup for the observation of the isomer HFS.



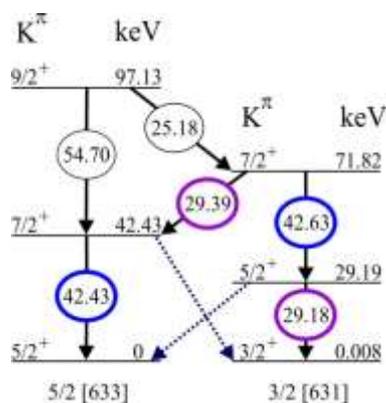
The only known isomer with an excitation energy in the optical range and in the range of outer shell electronic transitions.

- C.W. Reich, R.G. Helmer. PRL, 64, p.271 (1990) –  $3.5 \pm 1,0 \text{ eV}$

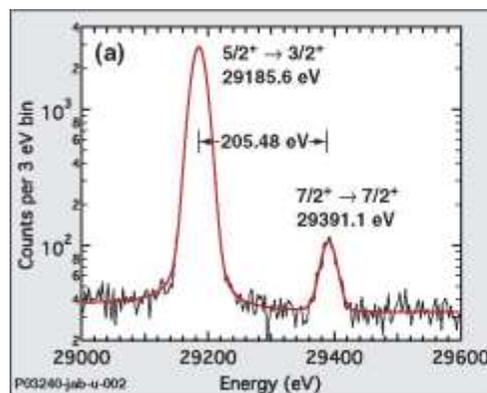
- B. R. Beck et al. (LLNL), PRL 98,142501 (2007).

$\gamma$  - spectroscopy of two decay cascades from the 71.82-keV-level

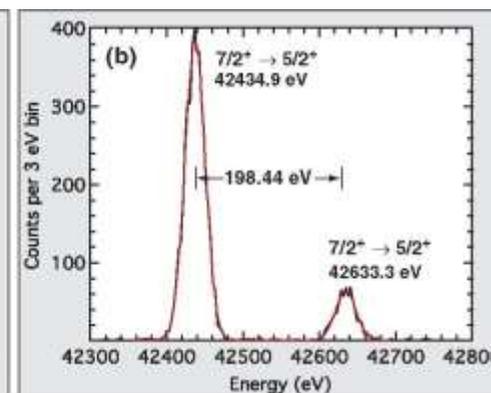
VUV – range –  $7.8 \pm 0.5 \text{ eV}$



29 KeV lines



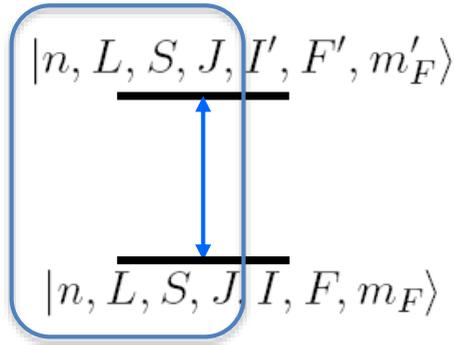
42 KeV lines



✓ Deexcitation of the isomer is observed in the recoil experiment (unfortunately the energy of the isomer is not derived).

Direct detection of the  $^{229}\text{Th}$  nuclear clock transition. L. von der Wense et. al. Nature 17669 (2016).

# Th 229: proposal for nuclear optical clocks



Nuclear and total-angular-momentum quantum numbers ( $I, F, m_F$ ) can change, purely electronic quantum numbers ( $n, L, S, J$ ) remain constant.

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

**Influence of the electron shell to the nuclear transition:** Frequency shifts that depend of ( $n, L, S, J$ ) are common for both levels. The nuclear transition frequency is independent of the shifts depending only on the electronic quantum numbers.

**Holds for:** Scalar quadratic Stark shift, including the effects of 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: expected:  $\approx 10^{-19}$  blackbody shift at room temperature

Linear Zeeman shift: use component  $m_F = 0 - 0$  or a pair of stretched states (diff. shift of 4 kHz/mT)

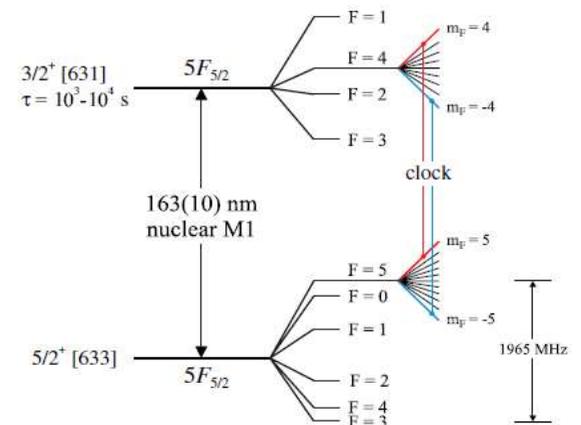
Doppler shift: use ion trapping and laser cooling

Frequency standard based on the pair of stretched hyperfine states:

$$|5F_{5/2}, I_g=5/2, F=5, m_F=\pm 5\rangle \leftrightarrow |5F_{5/2}, I_m=3/2, F=4, m_F=\pm 4\rangle.$$

Systematic shift suppression allows clock performance with a total fractional inaccuracy approaching  $1 \times 10^{-19}$ .

C.J. Campbell et al. PRL 108, 120802 (2012).



- The nuclear transition frequency is independent of the shifts depending only on the electronic quantum numbers.

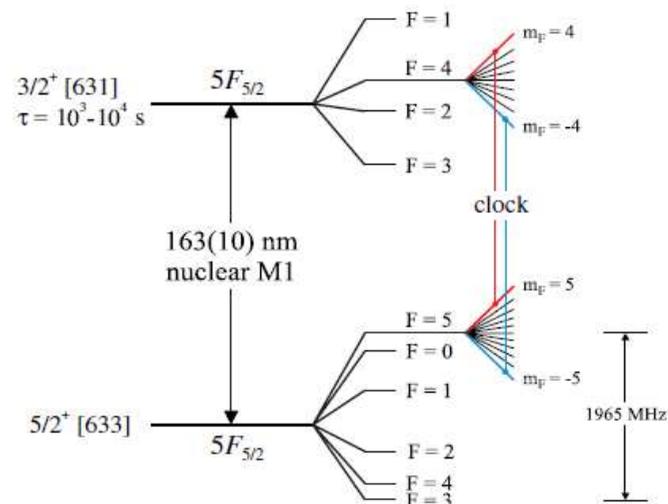
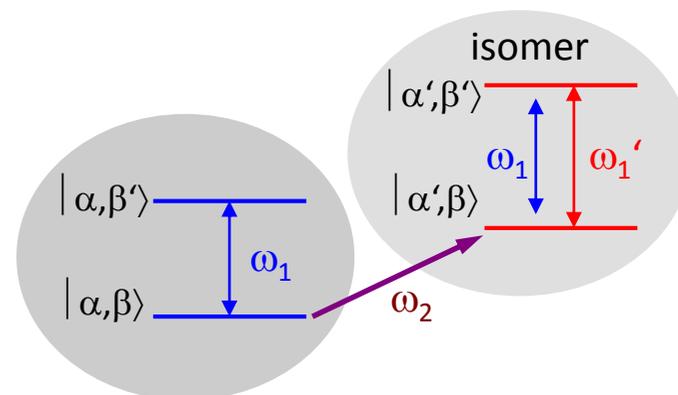
E. Peik and Chr. Tamm, Europhys. Lett. **61**, 181 (2003)

More stable against external perturbations due to small dimensions of nucleus in comparison with atom.

Nuclear and total-angular-momentum quantum numbers ( $I, F, m_F$ ) can change, purely electronic quantum numbers ( $n, L, S, J$ ) remain constant. Frequency shifts that depend of ( $n, L, S, J$ ) are common for both levels.

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# Test of fundamental constants

---

Th-229: the most sensitive probe in a search for variations of the fundamental coupling constants

Scaling of the  $^{229}\text{Th}$  transition frequency  $\omega$  in terms of  $\alpha$  and quark masses:  
V. Flambaum: Phys. Rev. Lett. **97**, 092502 (2006)

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

where  $X_q = m_q/\Lambda_{\text{QCD}}$  and  $X_s = m_s/\Lambda_{\text{QCD}}$

$10^5$  enhancement in sensitivity results from the near perfect cancellation of two  $\sim 1.4$  MeV contributions to the nuclear level energies.

>10 theory papers  
2006-2009

See for example:

A. C. Hayes, J. L. Friar, P. Möller, Phys. Rev. C **78**, 024311 (2008) ( $|A| \sim 10^3$ )

E. Litvinova et al., Phys. Rev. C **79**, 064303 (2009) ( $|A| \sim 4 \times 10^4$ )

# Test of fundamental constants

V.V. Flambaum. PRL 97, 092502 (2006).

## Ground state 5/2+[633]

(deformed oscillator quantum numbers  $N = 6$ ,  $n_z = 3$ , projection of valence neutron orbital angular momentum  $\Lambda = 3$ , spin projection  $\Sigma = -1/2$ , total angular momentum  $J = \Lambda + \Sigma = 5/2$ ).

## Excited state 3/2+[631]

$\Lambda = 1$ ,  $\Sigma = -1/2$ ,  $J = 3/2$ .

Energy of states:  $E_{e,g} = E_0 + C \Lambda \Sigma + D \Lambda^2$ ,  $\omega = E_e - E_g = 2C - 8D$ ,  $2C \approx 8D \approx -1.4 \text{ MeV}$ ,  $\omega \approx 3.5 \text{ eV}$ ,

$$\frac{\delta\omega}{\omega} = \frac{\delta(2C) - \delta(8D)}{\omega} \approx 4 \times 10^5 \left( \frac{\delta D}{D} - \frac{\delta C}{C} \right) \approx 10^5 \left( 4 \frac{\delta\alpha}{\alpha} + \frac{\delta X_q}{X_q} - 10 \frac{\delta X_s}{X_s} \right)$$

$$X_q = m_q / \Lambda_{QCD}, \quad X_s = m_s / \Lambda_{QCD}.$$

Comparing the Th nuclear frequency to present atomic clocks will allow to look for temporal variations at the level  $10^{-21}$  per year

# Photodissociation of Th<sup>+</sup>-containing molecular ions

Th<sup>+</sup> ions are highly reactive with O<sub>2</sub>, H<sub>2</sub>O, NO, CO<sub>2</sub>, CH<sub>4</sub>...

ThO<sup>+</sup> (double bond of 9.1 eV dissociation energy)

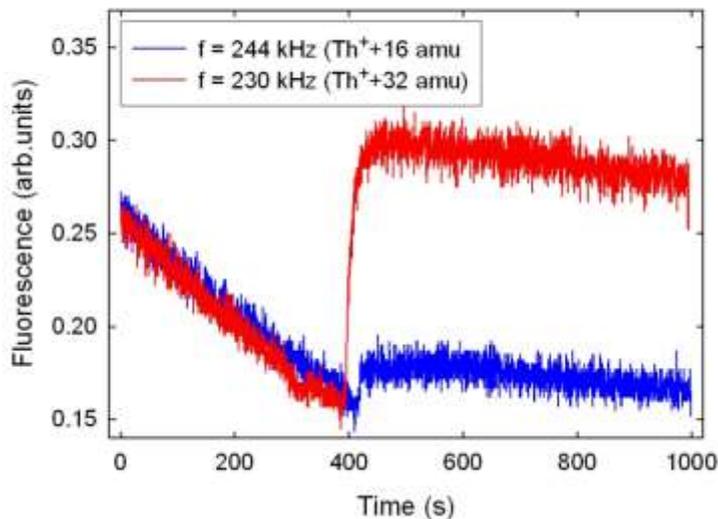
ThOH<sup>+</sup>, ThO<sub>2</sub><sup>+</sup>

ThCH<sub>2</sub><sup>+</sup> (dissociation energy of 4.8 eV)

Typical storage time in the trap with buffer gas:  
a few hundred sec.

With THG radiation of Ti:Sa ( $I_{\text{pulse}} \geq 50 \text{ kW/cm}^2$ ,  
wavelength range 237 – 289 nm)  $\tau \geq 30000 \text{ s}$

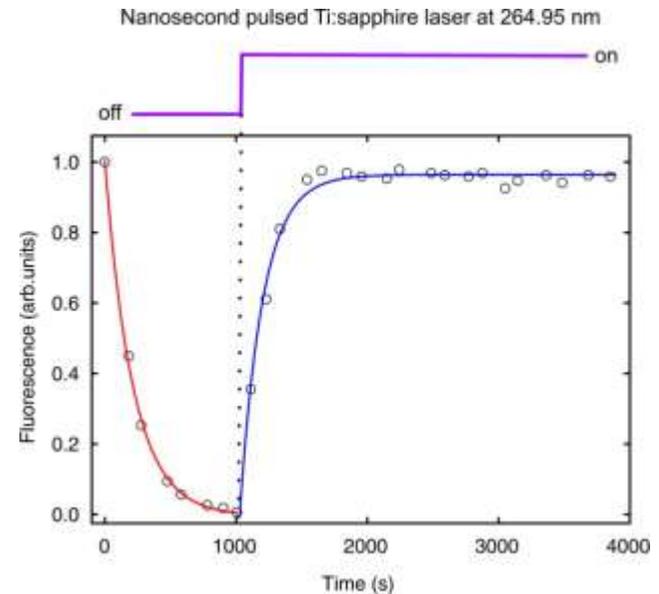
At low laser intensity observed resonant structure



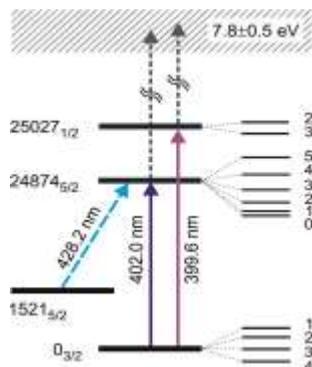
Ejection of ions with secular frequency, applied to DC electrodes of middle section.

$f = 244 \pm 2 \text{ kHz}$  – ejection of Th<sup>+</sup>X with 246-259 amu

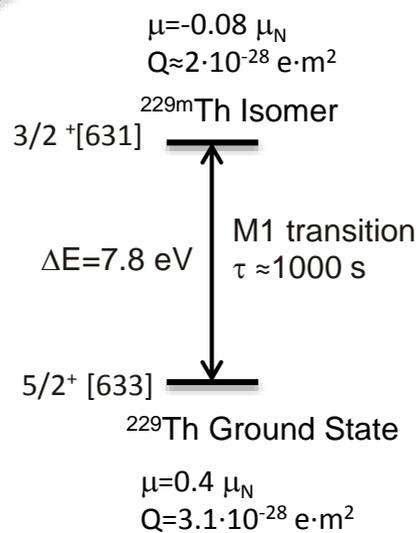
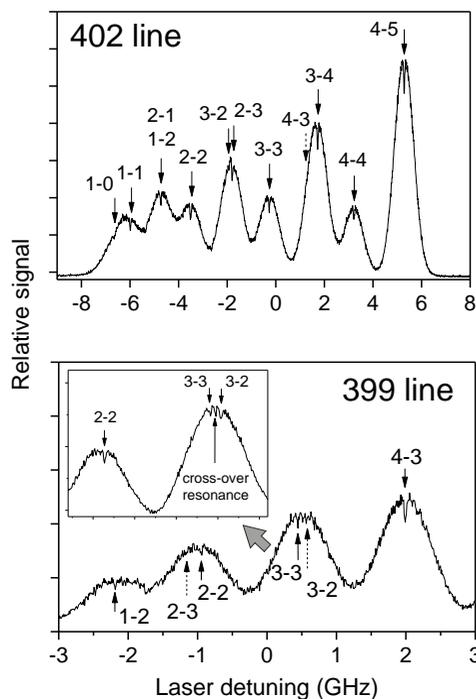
$f = 230 \pm 2 \text{ kHz}$  – ejection of Th<sup>+</sup>X with  $\approx 265 \text{ amu}$



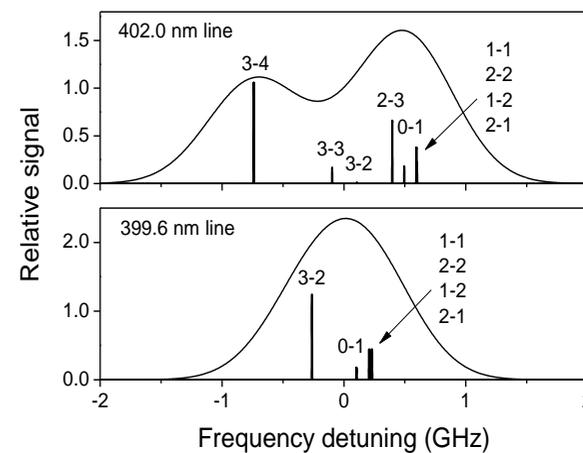
# Hyperfine structure of $^{229}\text{Th}^+$ lines



Level [cm <sup>-1</sup> ]	A factor [MHz]	B factor [MHz]	ref
0	-444.2(3.4) -444.2(1.9)	308.4(12.7) 303(6)	* Kälber et al.
24874	489.2(3.7) 488.9(3.7)	-408.8(17.9) -412.8(15.5)	Based on *
25027	-45(19) -42(16)		Based on *



## Expected HFS of the isomer



Assuming  $A_{is}/A_{gs} \sim \mu_{is}/\mu_{gs}$ ,  $B_{is}/B_{gs} \sim Q_{is}/Q_{gs}$

# Isotope shift of $^{229}\text{Th}^+$ lines

## 402 nm line:

Positive isotopic effect (blue shift of  $^{229}\text{Th}^+$  line)  $0.61 \text{ cm}^{-1}$  (18.3 GHz).

E.A. Vernyi, V.N. Egorov, Opt. Spectr. 9 (1960).

Ground state: leading components:  $6d^27s$  and  $6d7s^2$ ;

24874 state: leading components:  $6s7s7p$  and  $5f6d^2$

R. Zalubas, C.H. Corliss. J. Res. Nat. Bur. St. A78, 163 (1974)

Very small probability to observe negative isotopic effect in Th (since the s-electrons of the ground state (volume effect) make the greatest contribution to the isotopic effect).

A few hundred  $^{229}\text{Th}^+$  and  $^{230}\text{Th}^+$  lines were investigated - positive isotopic effect observed. G.L. Stukenbroeker, J.R. McNally, Jr. J. Opt. Soc. Am. 43, 1 (1953)

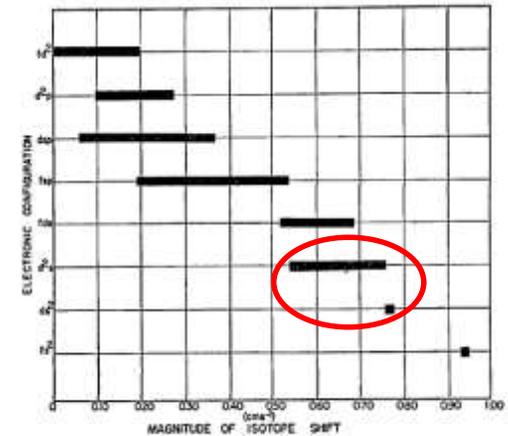
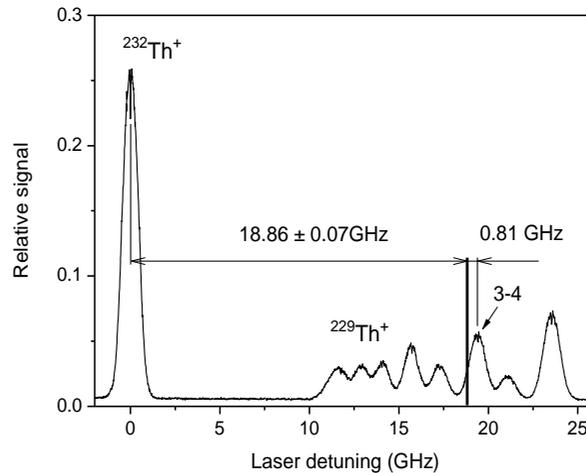


FIG. 3. Approximate range of isotope shifts as a function of electronic configurations.

## 399 nm line:

24874 state: leading component:  $5f6d^2$

R. Zalubas, C.H. Corliss. J. Res. Nat. Bur. St. A78, 163 (1974)

We observe negative isotopic effect  $f s^2$  configurations only appear at low energies  $s^2 p$  configuration? Missed on the graph.

A few levels with  $s^2 p$  configurations were identified in this energy range.

Ab initio calculations: S. Porsev, M. Safronova - leading configuration  $7s^2 7p!$

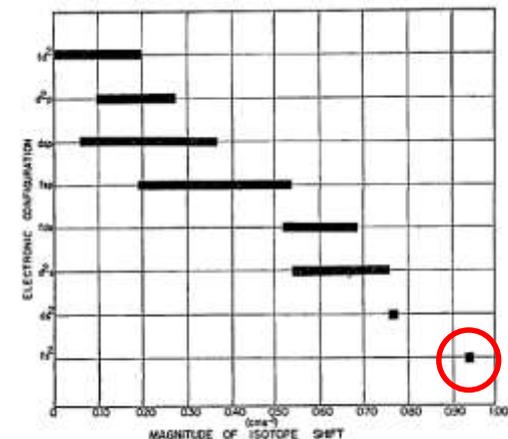
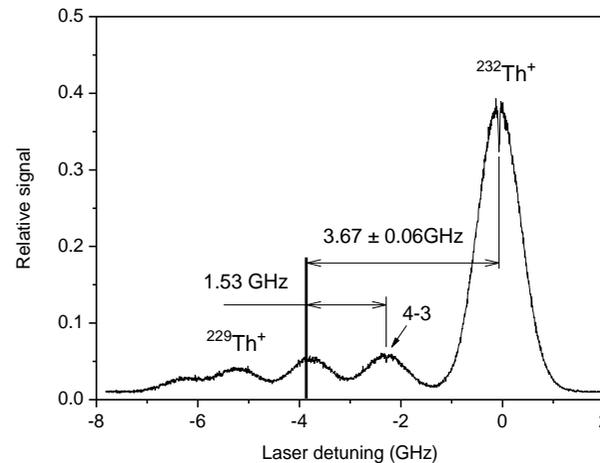


FIG. 3. Approximate range of isotope shifts as a function of electronic configurations.

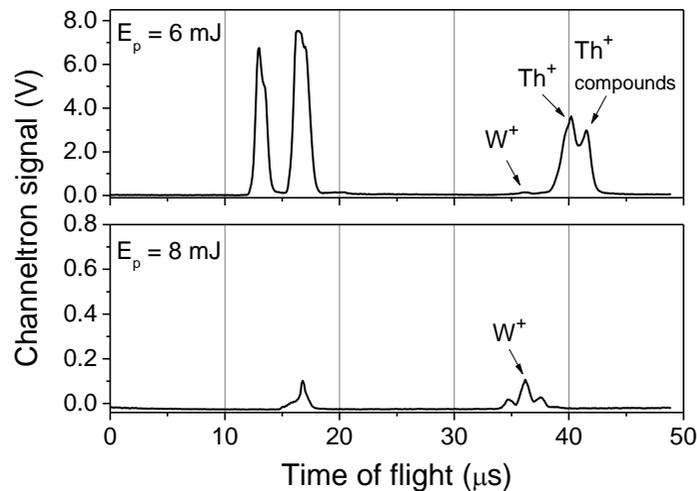
# Trap loading with $^{229}\text{Th}^+$

## Tests with TOF spectrometer

Different targets with  $^{232}\text{Th}(\text{NO}_3)_4$

Ablation laser: pulsed Nd:YAG (1064, 532, 355 nm); pulse duration  $\sim 10$  ns, pulse energy  $< 10$  mJ

Ablation of Th from Al and W substrates at 1064 nm demonstrates the most promising result.



## Trap loading tests

Amount of  $^{232}\text{Th}$  on substrate:  $\sim 4 \times 10^{14}$  atoms, 140 ng  $\rightarrow$  1 kBq activity of  $^{229}\text{Th}$

**Loading from W substrate:**

Executed  $> 500$  loadings with the ion number  $> 30\%$  of the maximum trap capacity

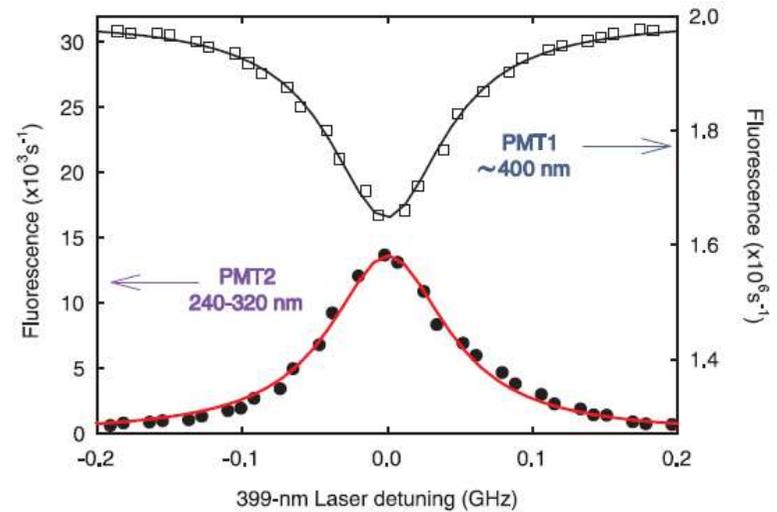
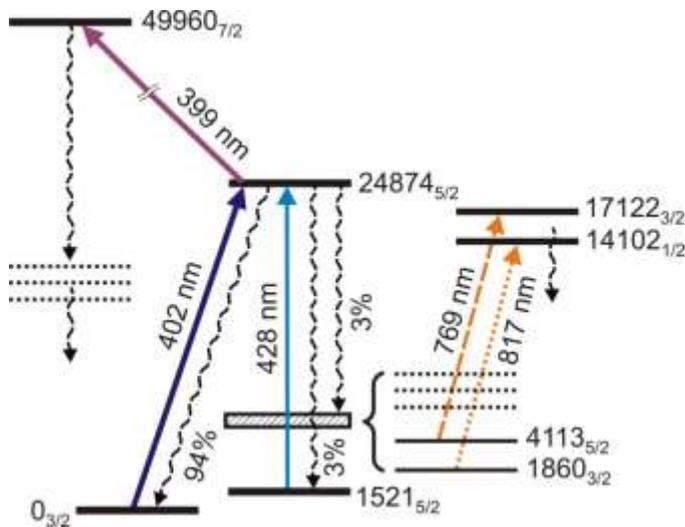
Loading efficiency  $\geq 10^{-6}$  (one order of magnitude higher than achieved with evaporated method\*).

**Loading from Al substrate:** not efficient. Influence of a big number of Al ions in ablation plume which affect Th trajectories.

**Loading of  $^{229}\text{Th}$ :**  $\sim 50$  loadings are executed...

\* W Kälber et al. J. Mod. Opt. 39 335 (1992)

# Population trapping and two photon excitation in CW mode

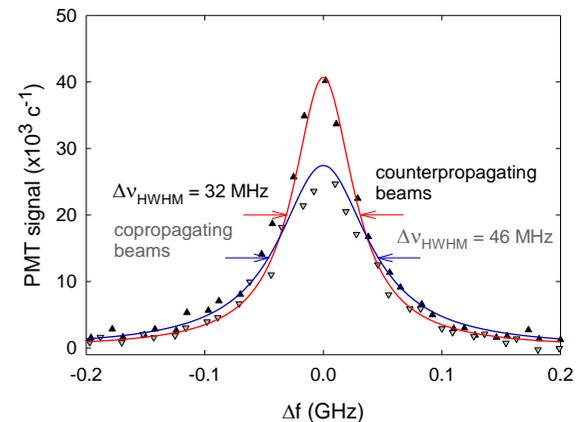


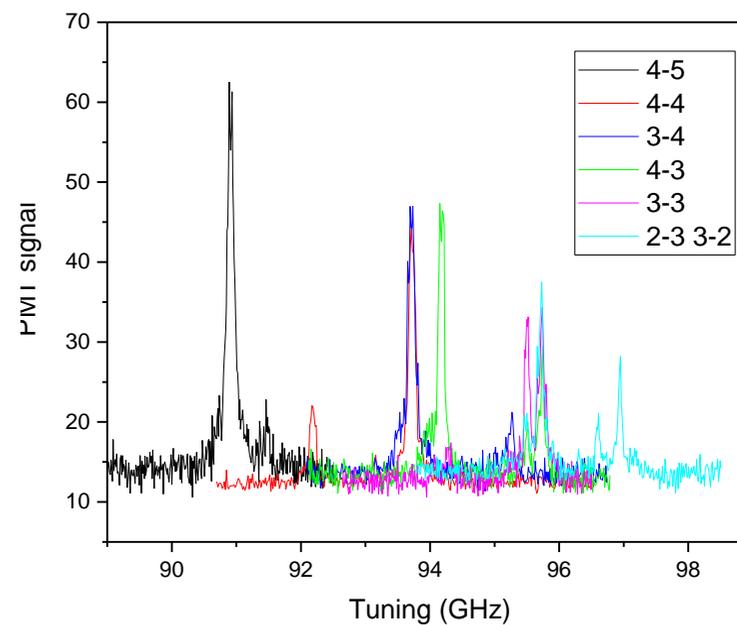
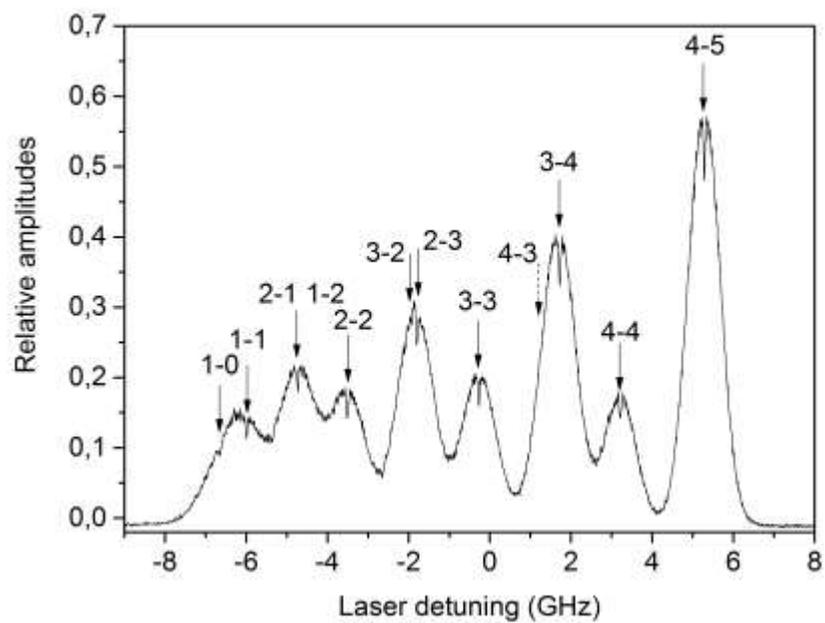
depletion of 24874  $\text{cm}^{-1}$  state and fluorescence from 49960  $\text{cm}^{-1}$  state

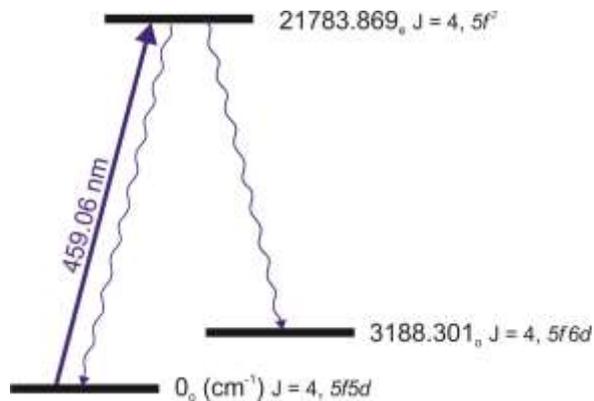
- CW mode – population trapping
- Collisional quenching (He, Ar,  $\text{N}_2$ )
- Repumper at 428 nm.

✓ Two photon excitation with 2 ECDLs: low fraction  $\sim 0.1\%$  is transferred to the 24874 state.

O.A. Herrera-Sancho, M.V. Okhapkin, K. Zimmermann, Chr. Tamm, E. Peik, A.V. Taichenachev, V.I. Yudin, P. Glowacki Phys.Rev. A 85, 033402 (2012).



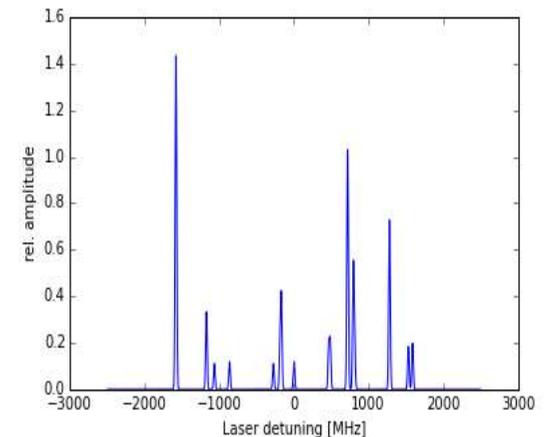
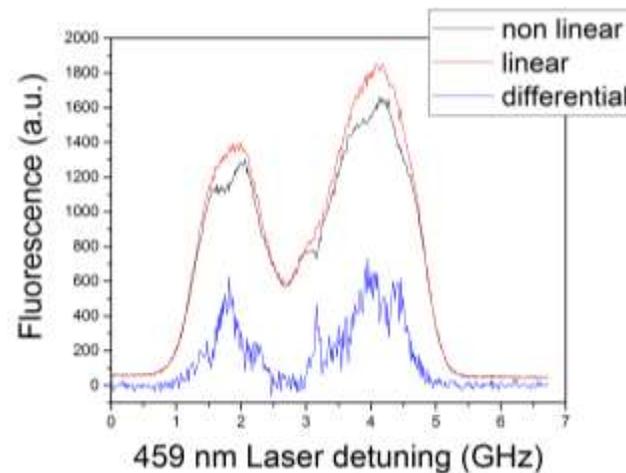
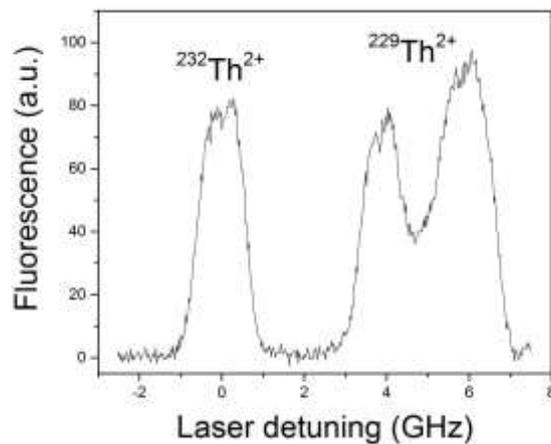


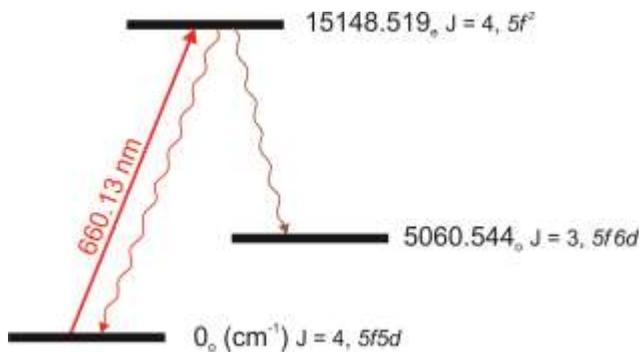


Positive isotope shift of  $^{229}\text{Th}^{2+}$  relatively to  $^{232}\text{Th}^{2+}$  is  $5.3 \pm 0.2 \text{ GHz}$ .

High signal-to-noise ratio due to background-free detection at 538 nm.

Hyperfine structures of  $^{229\text{m}}\text{Th}$  and  $^{229}\text{Th}$  expected to be superimposed.





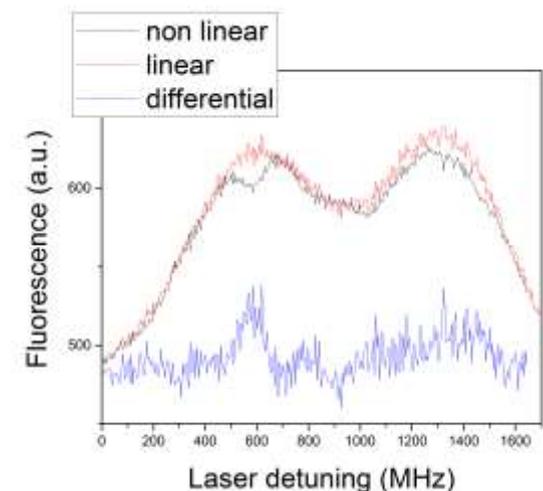
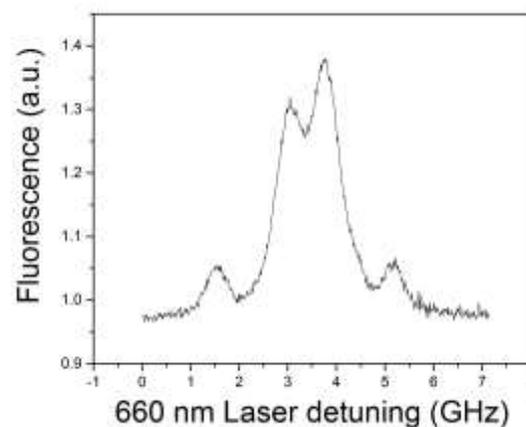
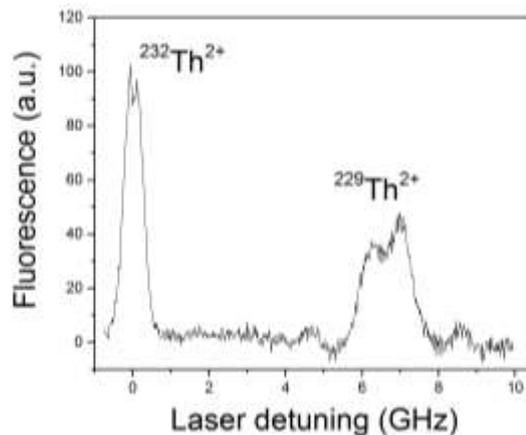
Positive isotope shift of  $^{229}\text{Th}^{2+}$  relatively to  $^{232}\text{Th}^{2+}$  is  $6.7 \pm 0.3 \text{ GHz}$ .

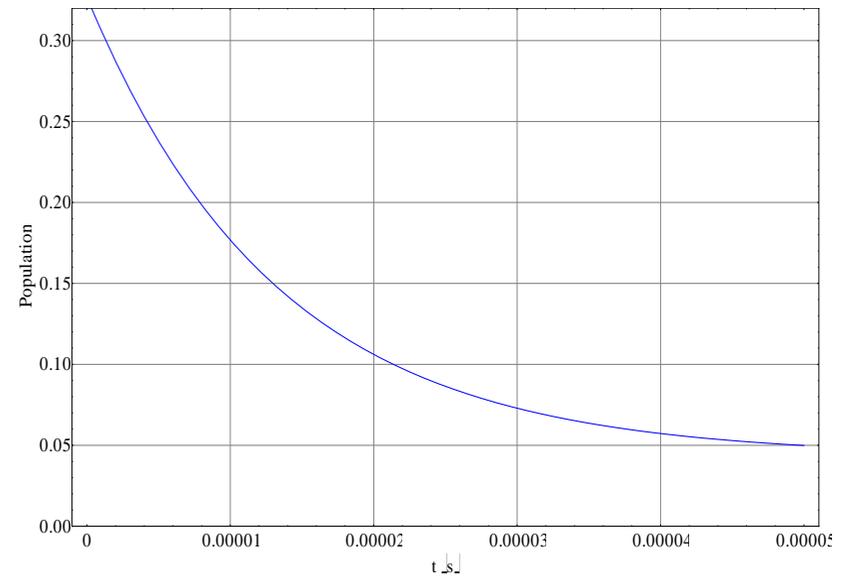
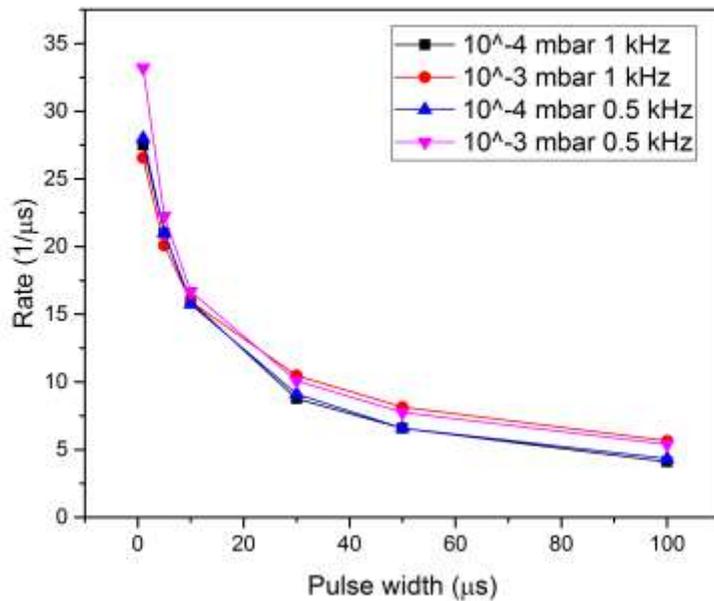
Disadvantage:

Background-free detection is in the IR range.

Detection at 660 nm is influenced by laser stray light.

Detection at 991 nm is in preparation.





$$\begin{aligned} \gamma_{21} &= 1 \cdot 10^8; \\ \gamma_{31} &= 1 \cdot 10^8; \\ \gamma_{23} &= 1 \cdot 10^8; \\ \gamma_{41} &= 1 \cdot 10^4; \\ \gamma_{24} &= 2 \cdot 10^5; \end{aligned}$$



