Hunting for mysterious ultra-low energy isomer of Thorium-229

- to realize ultimate "nuclear clock"-

Koji YOSHIMURA
RIIS, Okayama University

2018.5.22. RIKEN RIBF Nuclear Physics Seminar

Research Institute for Interdisciplinary Science
Quantum Universe Division

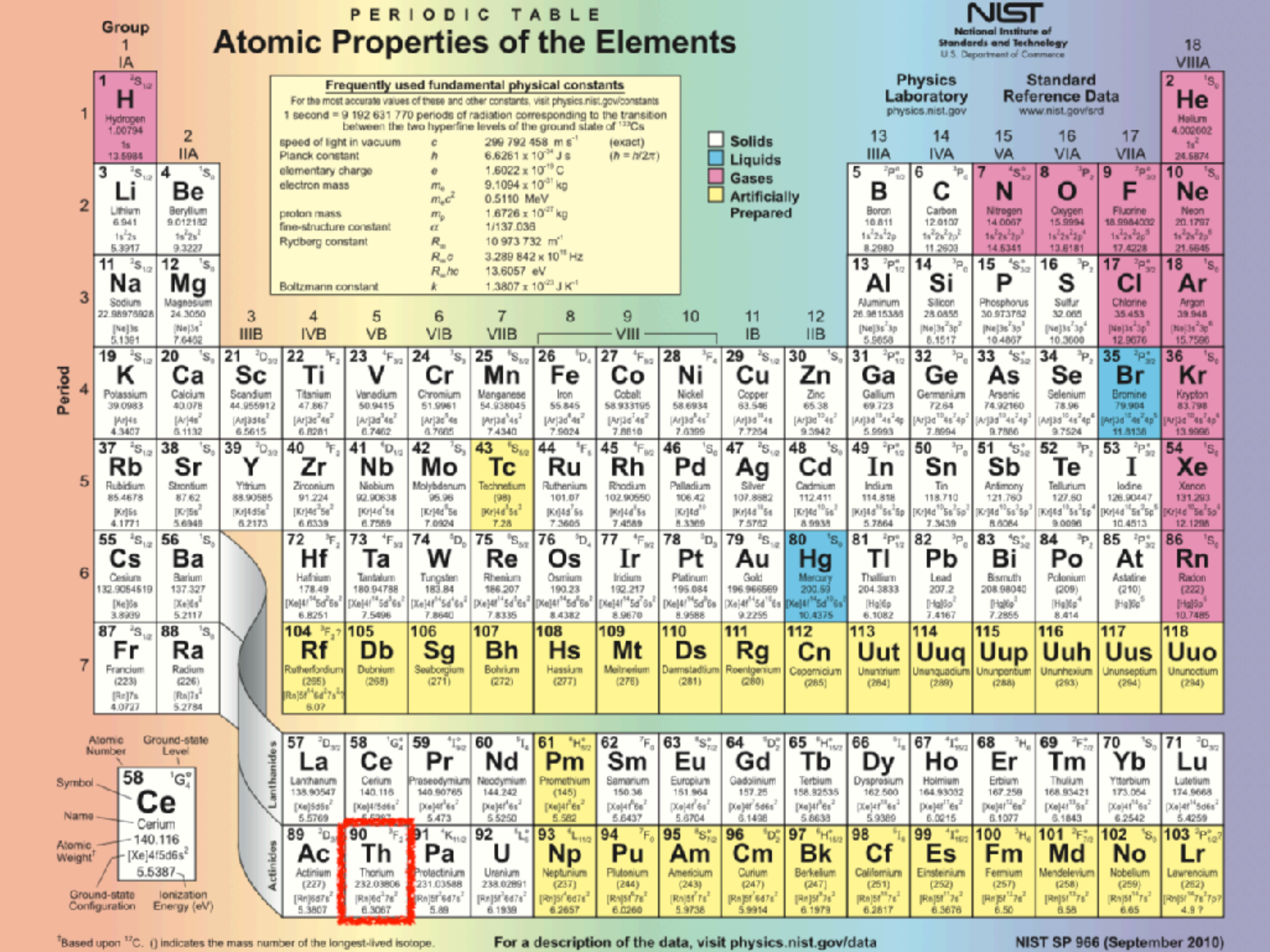
Particle Physics/ Cosmology(5) : M. Yoshimura, K. Yoshimura, N. Sasao, T. Masuda, T. Hiraki
Nuclear Physics(1) : H. Yoshimi
Quantum Electronis/ Atomic Physics(4) : S. Uetake, H. Hara, K. Imamura, Y. Imai
Quantum Chemistry(1): Y. Miyamoto
Students (10)

Pursue Fundamental Physics using various field of knowledge

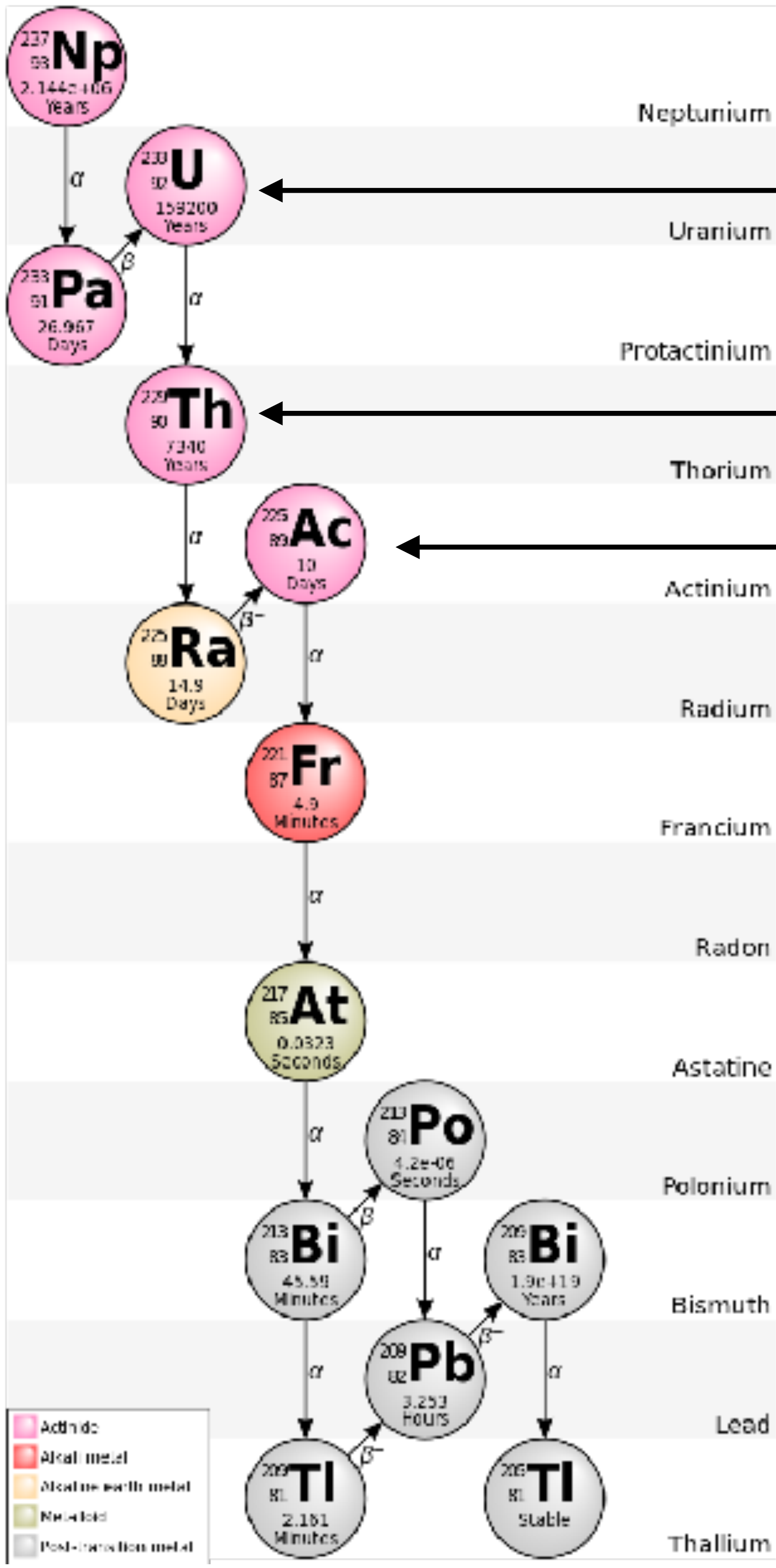
Outline

- Introduction
- History of hunting Th-229 Isomer
- A new method of Th-229 search (SPring-8 Experiment)
- Recent result
- Future prospect

Introduction


PERIODIC TABLE
Atomic Properties of the Elements
NIST
National Institute of Standards and Technology
U.S. Department of Commerce
Physics Laboratory
physics.nist.gov
Standard Reference Data
www.nist.gov/srd
Frequently used fundamental physical constants
For the most accurate values of these and other constants, visit physics.nist.gov/constants
1 second = 9 192 631 770 periods of radiation corresponding to the transition between the two hyperfine levels of the ground state of 133Cs
speed of light in vacuum c 299 792 458 m s-1 (exact)
Planck constant h 6.6261 x 10-34 J s (ħ = h/2π)
elementary charge e 1.6022 x 10-19 C
electron mass me 9.1094 x 10-31 kg
mec2 0.5110 MeV
proton mass mp 1.6726 x 10-27 kg
fine-structure constant α 1/137.036
Rydberg constant R∞ 10 973 732 m-1
R∞c 3.289 842 x 1015 Hz
R∞hc 13.6057 eV
Boltzmann constant k 1.3807 x 10-23 J K-1
Solids
Liquids
Gases
Artificially Prepared
Group
Period
Lanthanides
Actinides
H Hydrogen
He Helium
Li Lithium
Be Beryllium
B Boron
C Carbon
N Nitrogen
O Oxygen
F Fluorine
Ne Neon
Na Sodium
Mg Magnesium
Al Aluminum
Si Silicon
P Phosphorus
S Sulfur
Cl Chlorine
Ar Argon
K Potassium
Ca Calcium
Sc Scandium
Ti Titanium
V Vanadium
Cr Chromium
Mn Manganese
Fe Iron
Co Cobalt
Ni Nickel
Cu Copper
Zn Zinc
Ga Gallium
Ge Germanium
As Arsenic
Se Selenium
Br Bromine
Kr Krypton
Rb Rubidium
Sr Strontium
Y Yttrium
Zr Zirconium
Nb Niobium
Mo Molybdenum
Tc Technetium
Ru Ruthenium
Rh Rhodium
Pd Palladium
Ag Silver
Cd Cadmium
In Indium
Sn Tin
Sb Antimony
Te Tellurium
I Iodine
Xe Xenon
Cs Cesium
Ba Barium
Hf Hafnium
Ta Tantalum
W Tungsten
Re Rhenium
Os Osmium
Ir Iridium
Pt Platinum
Au Gold
Hg Mercury
Tl Thallium
Pb Lead
Bi Bismuth
Po Polonium
At Astatine
Rn Radon
Fr Francium
Ra Radium
Rf Rutherfordium
Db Dubnium
Sg Seaborgium
Bh Bohrium
Hs Hassium
Mt Meitnerium
Ds Darmstadtium
Rg Roentgenium
Cn Copernicium
Uut Ununtrium
Uuq Ununquadium
Uup Ununpentium
Uuh Ununhexium
Uus Ununseptium
Uuo Ununoctium
La Lanthanum
Ce Cerium
Pr Praseodymium
Nd Neodymium
Pm Promethium
Sm Samarium
Eu Europium
Gd Gadolinium
Tb Terbium
Dy Dysprosium
Ho Holmium
Er Erbium
Tm Thulium
Yb Ytterbium
Lu Lutetium
Ac Actinium
Th Thorium
Pa Protactinium
U Uranium
Np Neptunium
Pu Plutonium
Am Americium
Cm Curium
Bk Berkelium
Cf Californium
Es Einsteinium
Fm Fermium
Md Mendelevium
No Nobelium
Lr Lawrencium
Atomic Number
Ground-state Level
Symbol
Name
Atomic Weight†
Ground-state Configuration
Ionization Energy (eV)
58 Ce Cerium 140.116 [Xe]4f5d6s2 5.5387
†Based upon 12C. () indicates the mass number of the longest-lived isotope.
For a description of the data, visit physics.nist.gov/data
NIST SP 966 (September 2010)

Neptunium Series



Uranium 233 Reactor fuel

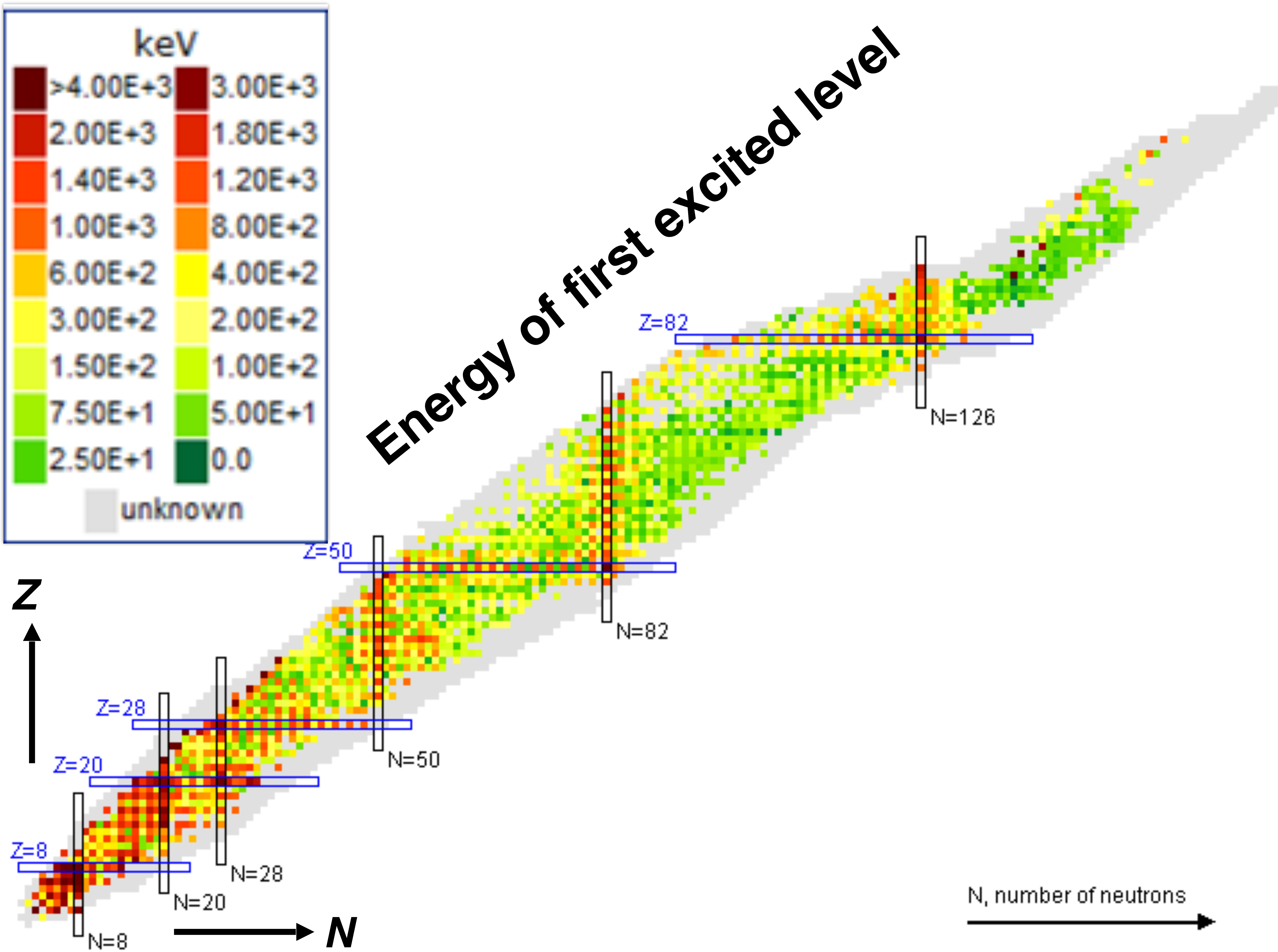
Thorium 229

Actinium 225 Unsealed radionuclide therapy

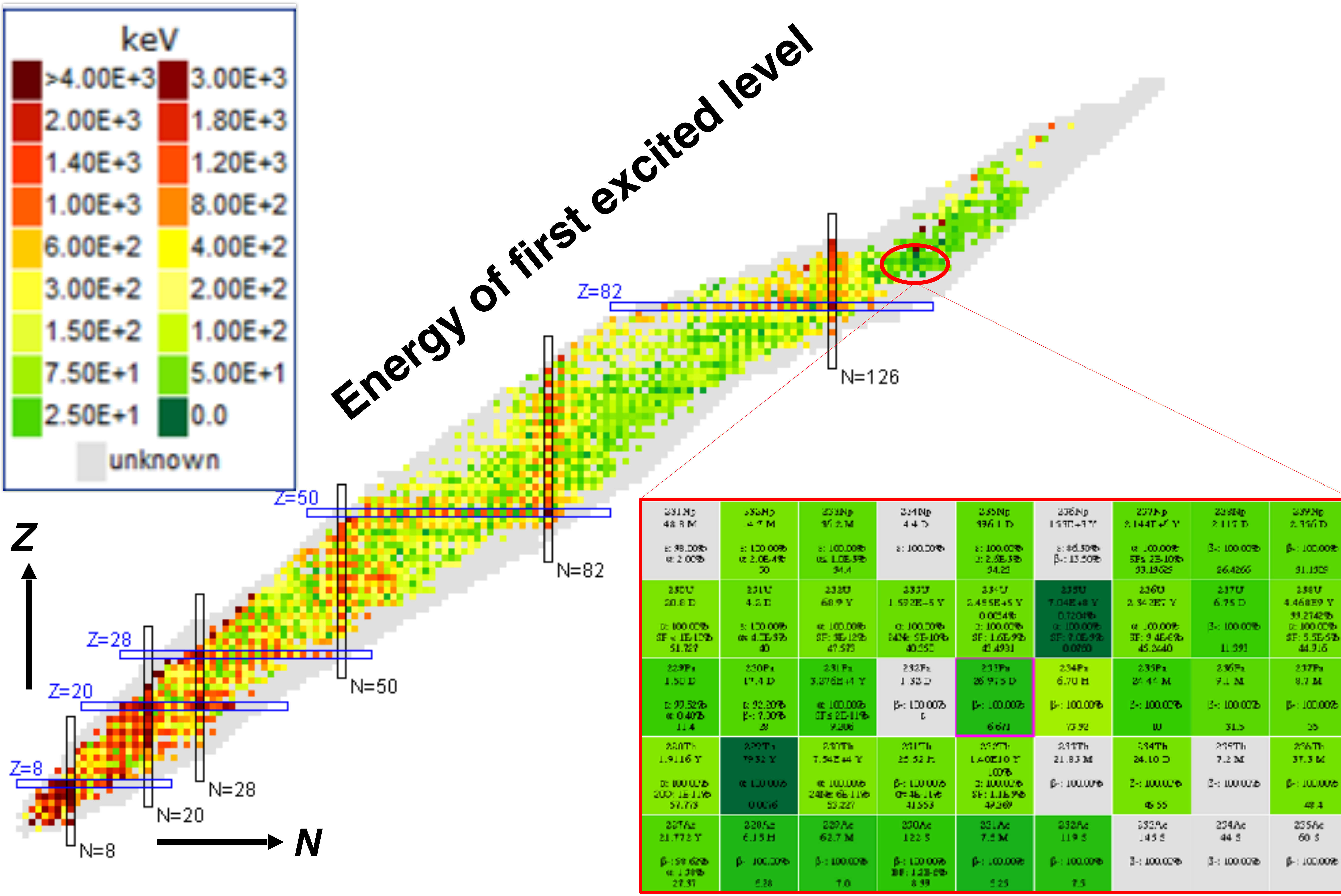
Cf. Natural Thorium-232 100%

$T_{1/2} = 1.4 \times 10^{10}$ y

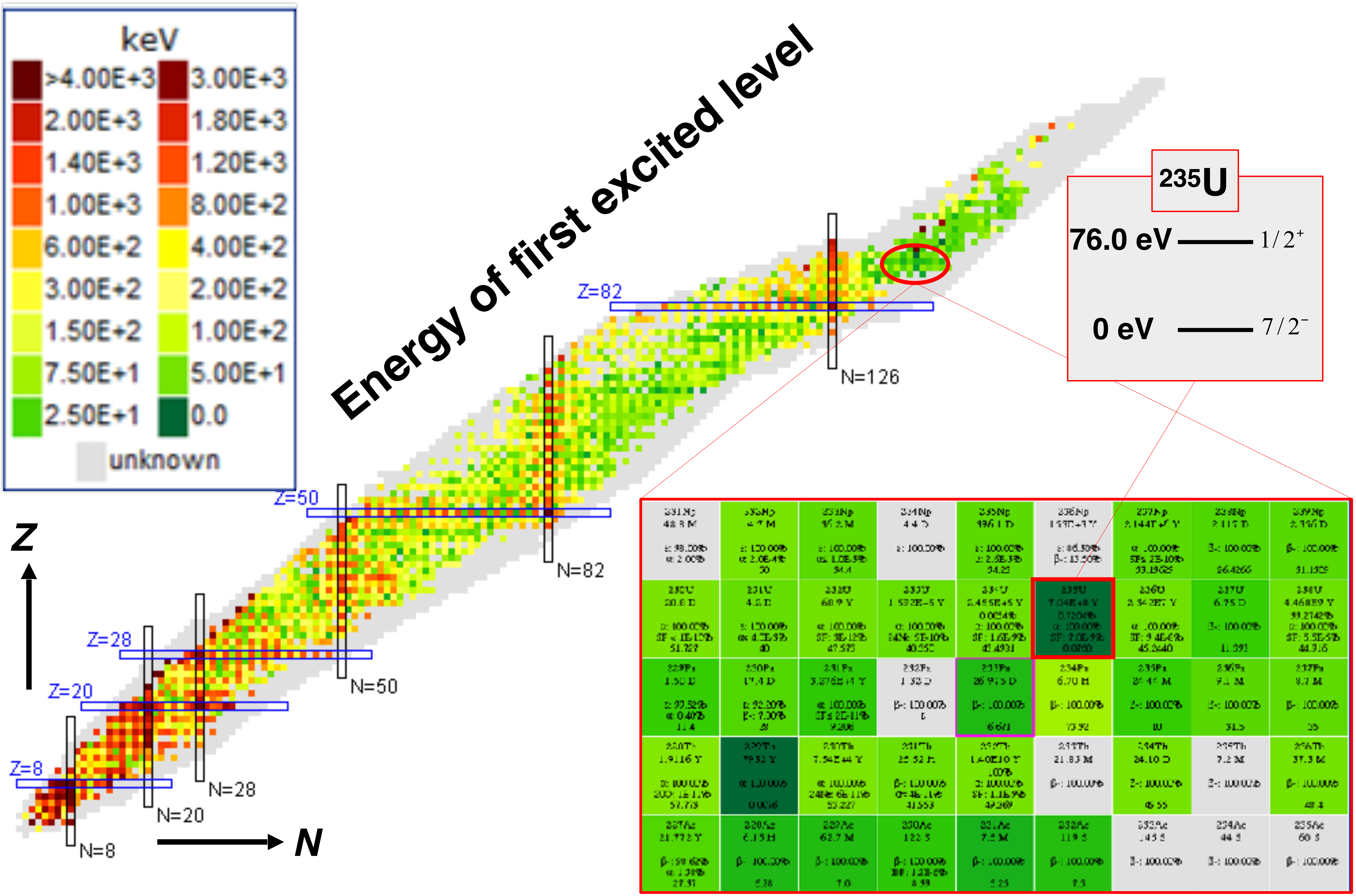
Energy of first excited level



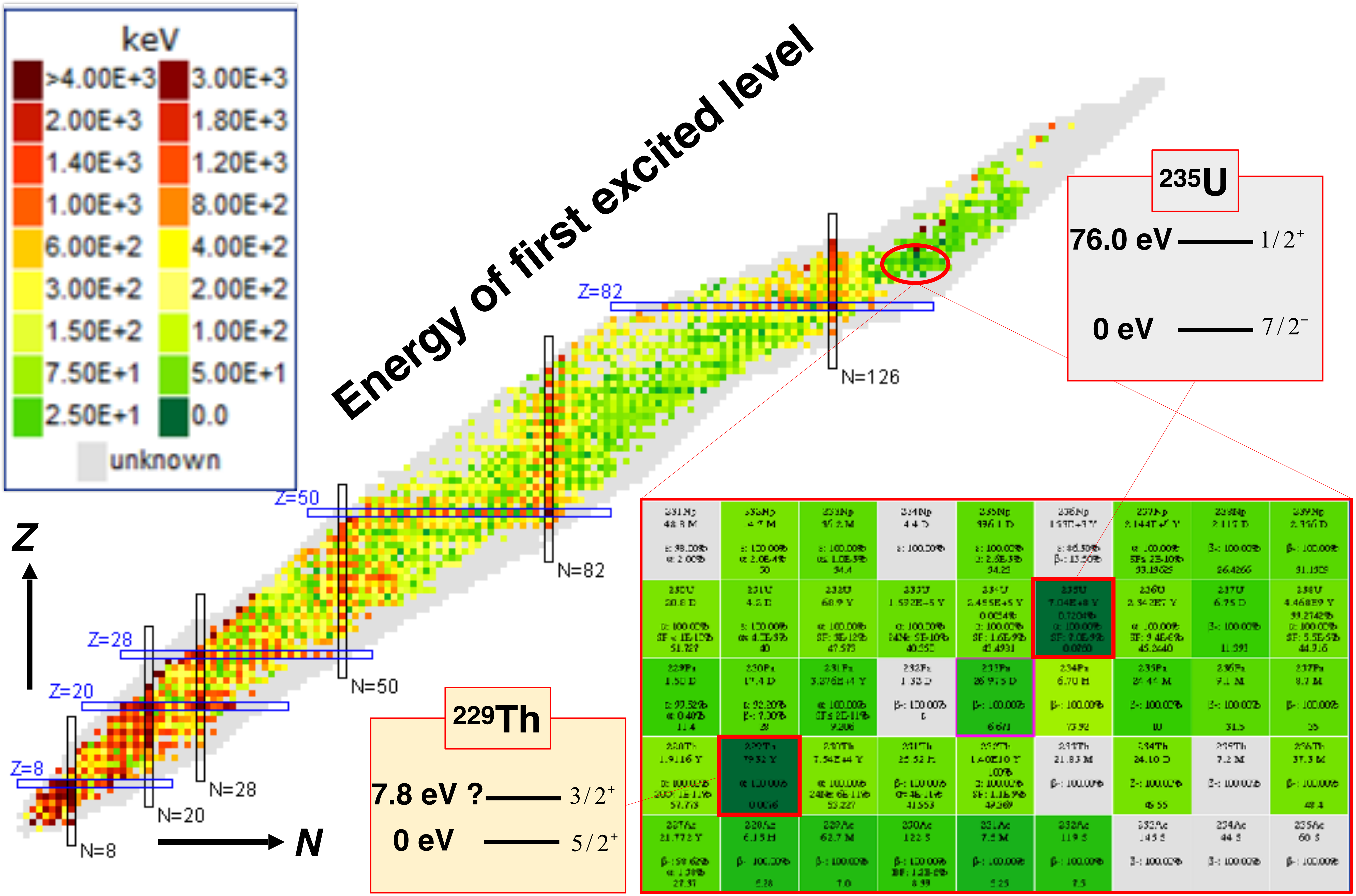
Energy of first excited level



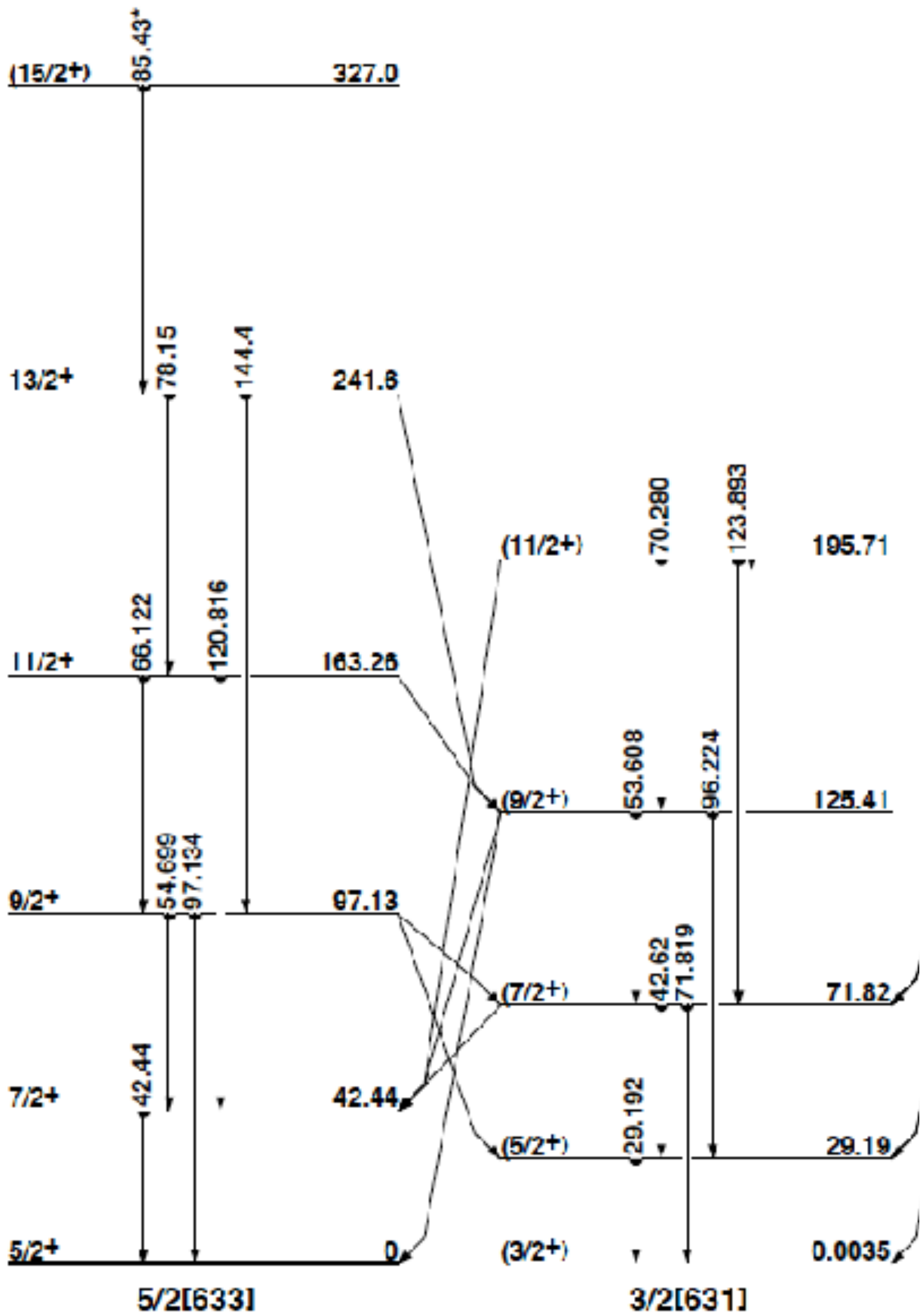
Energy of first excited level



Energy of first excited level

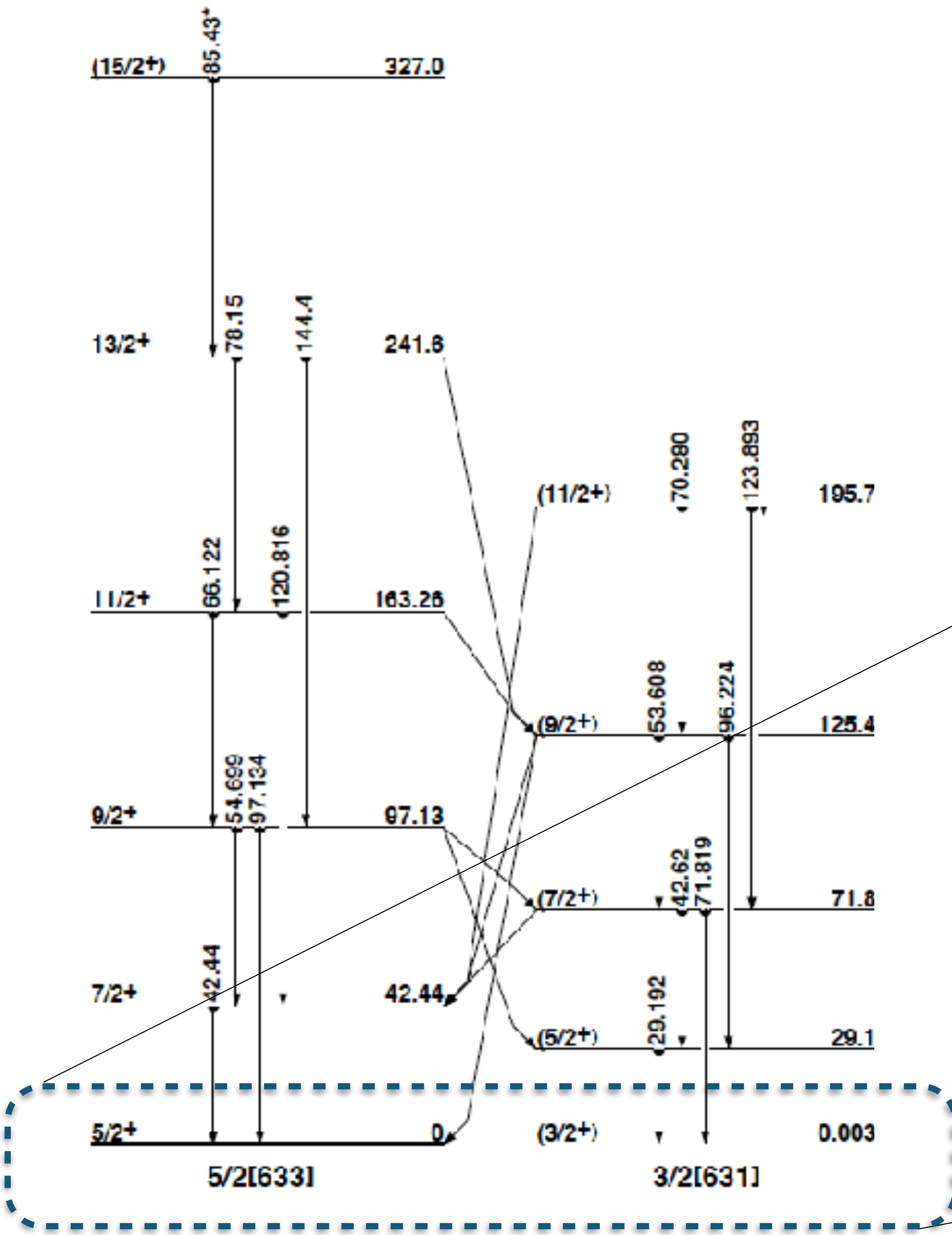


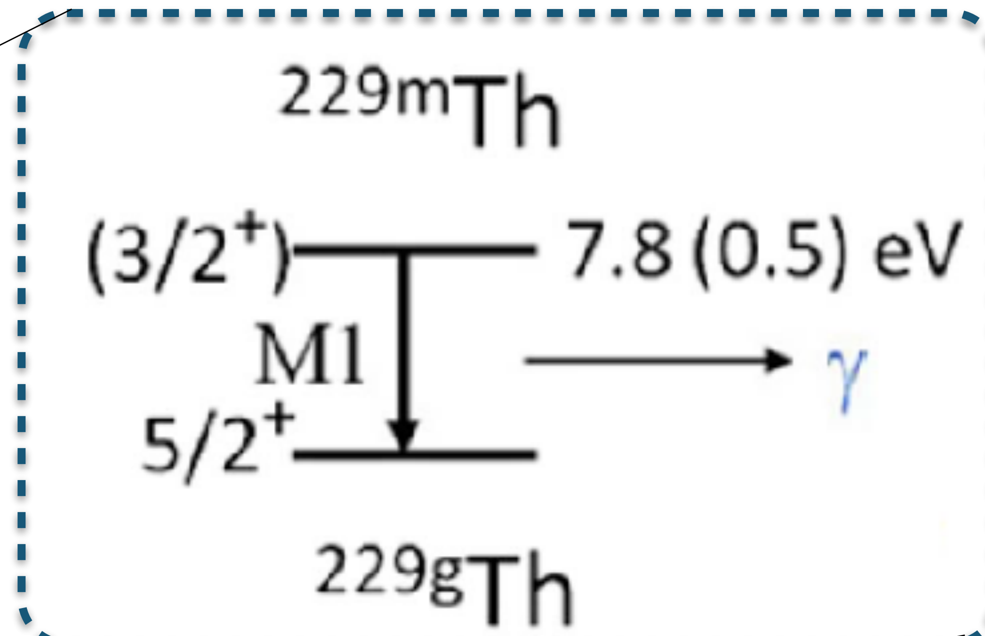
Th-229 level diagram



Ground State Isomer State

Th-229 level diagram





Excited State
7.8 eV
$T_{1/2}$: 1000s

Ground State
$T_{1/2}$: 7340 y

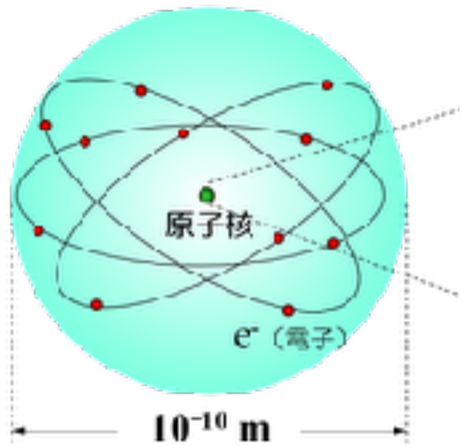
Ground State

Isomer State

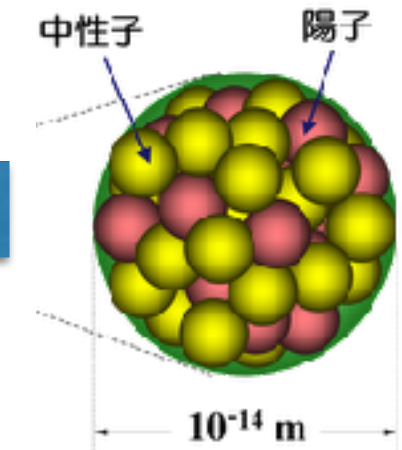
Application of Th-229m

Impact of eV-order excitation level

Atom



Nucleus



eV-older excitation level

keV, MeV excitation level

Experimental methods using MW, Laser,
Spectroscopy, Cooling, BEC, Trap

Accelerator based

Coherent

ex.: Atomic clock (Lattice, ion)
10^{-18}

Nuclear Clock
$<10^{-19}$

Shielded by electrons

Insensitive to external field

Laser excitation

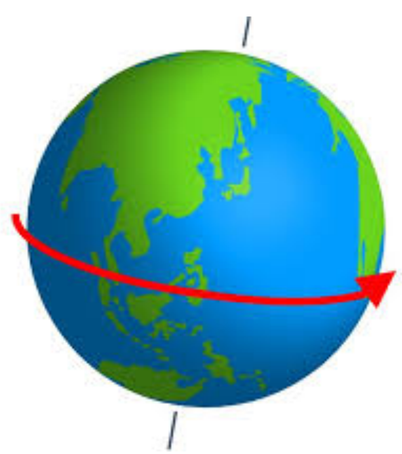
Three components of Clock

Counter

Instrument to measure counts (hand)

Oscillator

Object with periodic motion (pendulum)



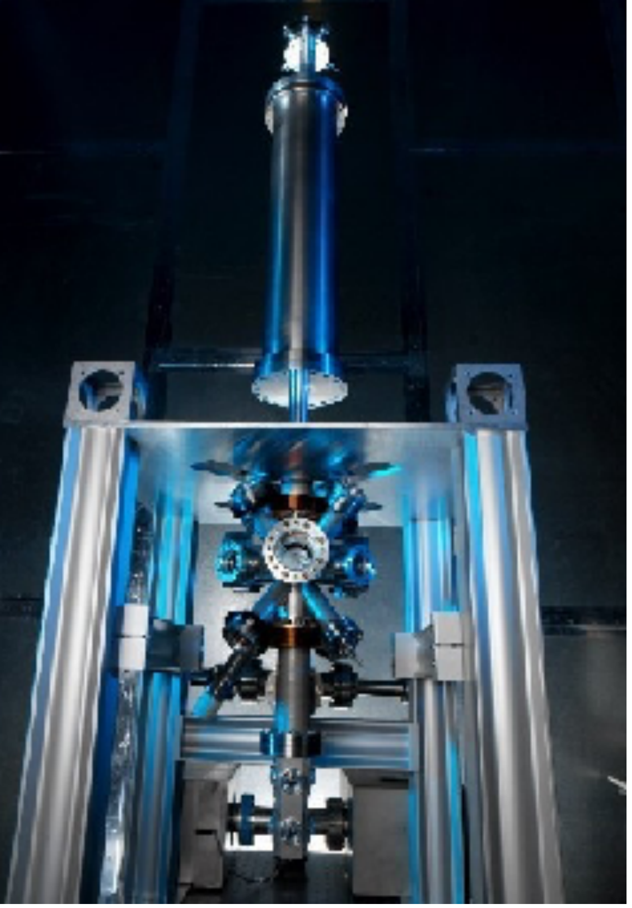
Reference

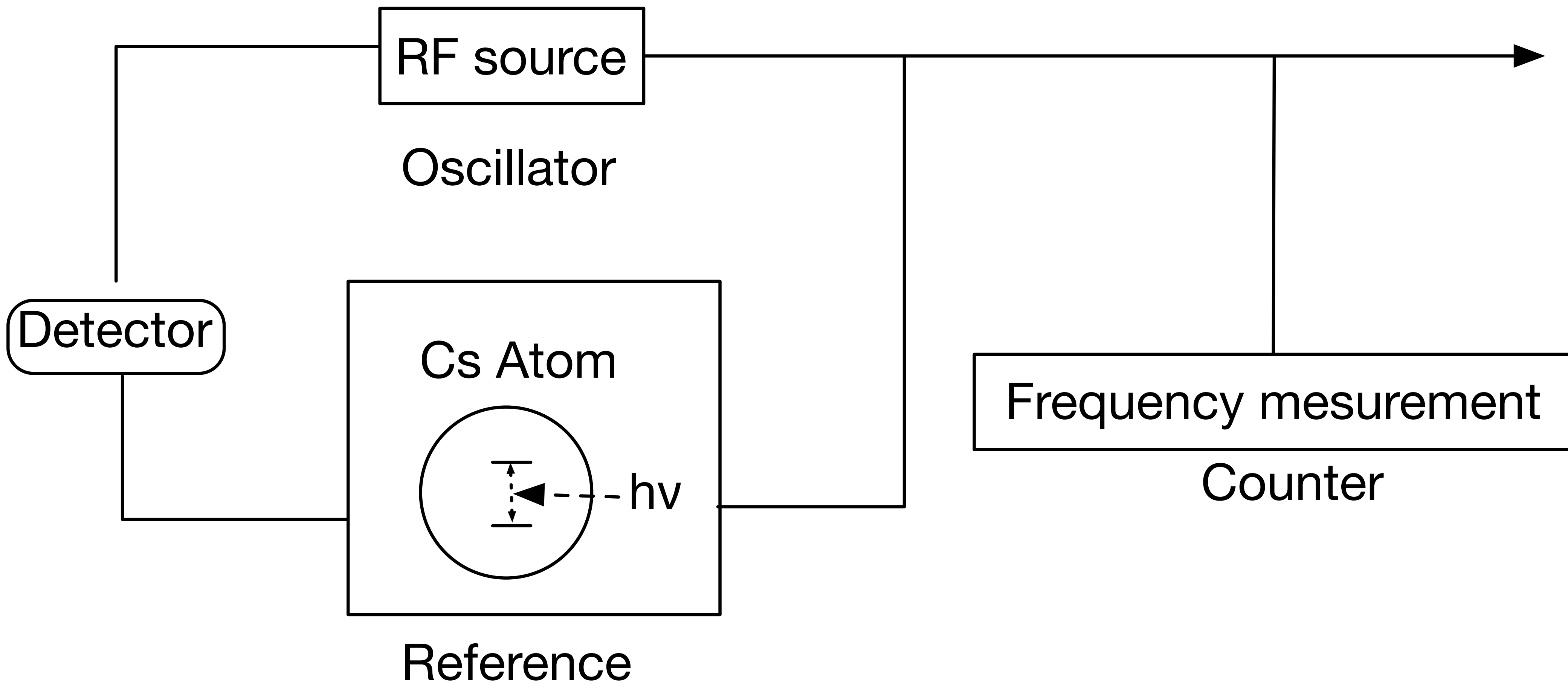
Reference (daily rotation)

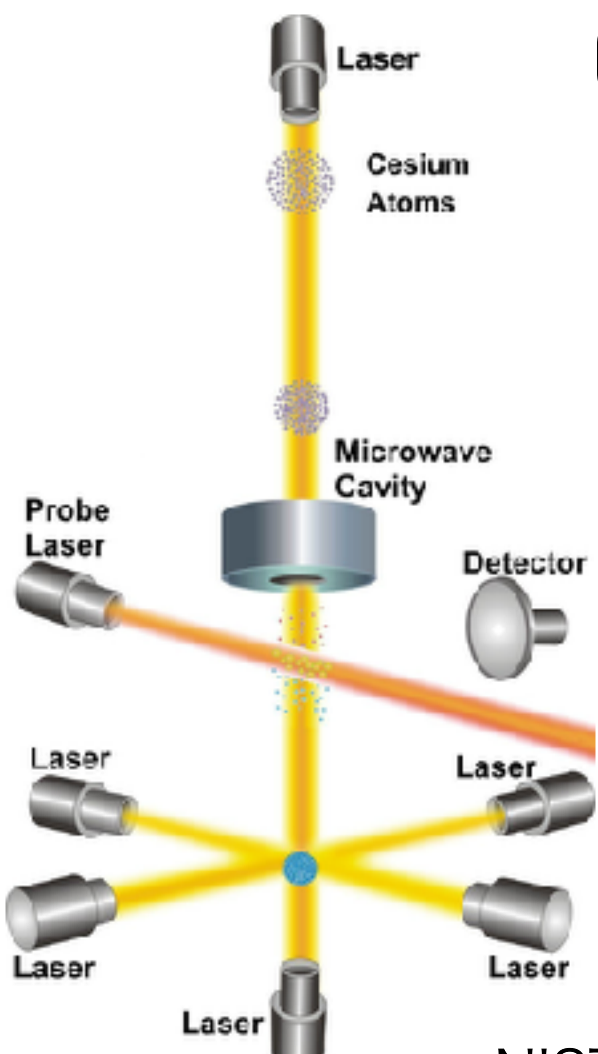
Shortt–Synchronome clock
1920 ~ 1930, precision 10^{-10}

Atomic clock—frequency standard

Cesium fountain clock







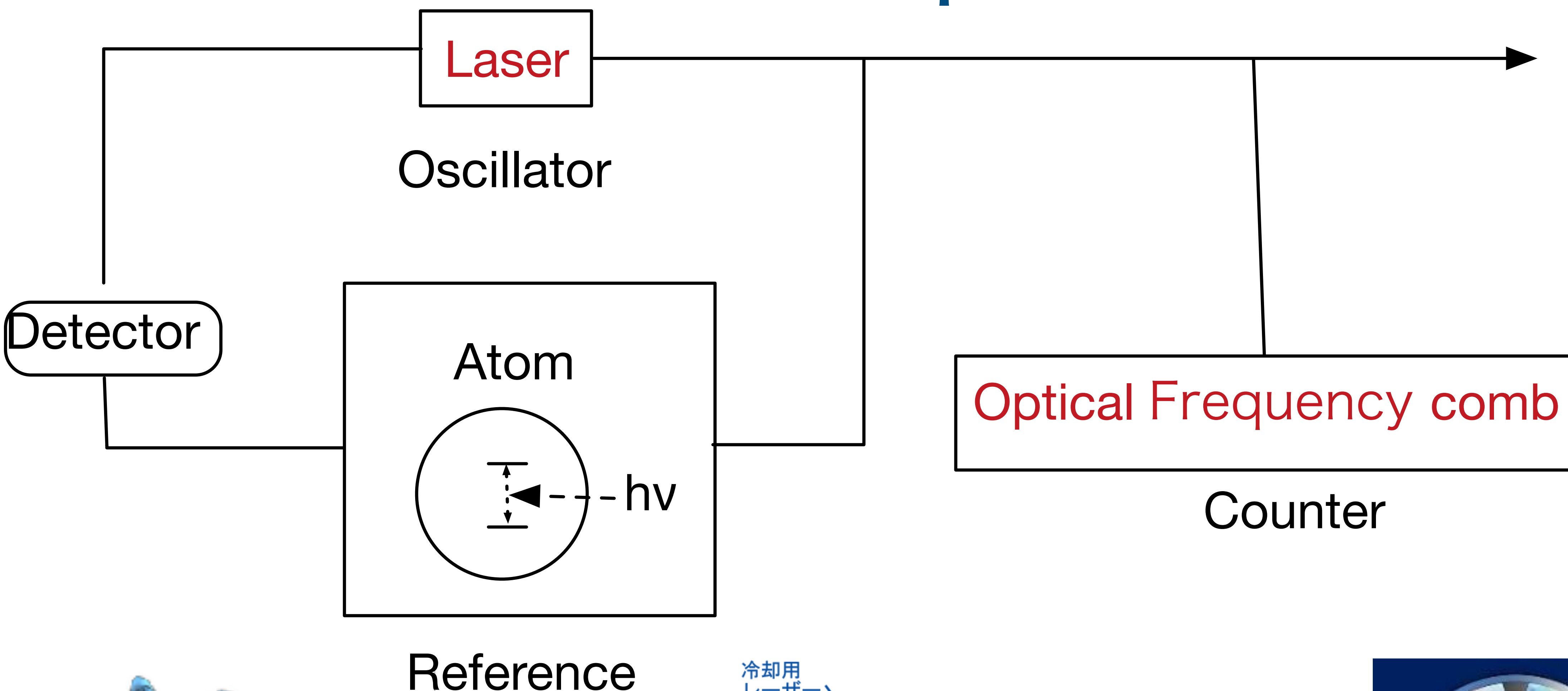
NIST

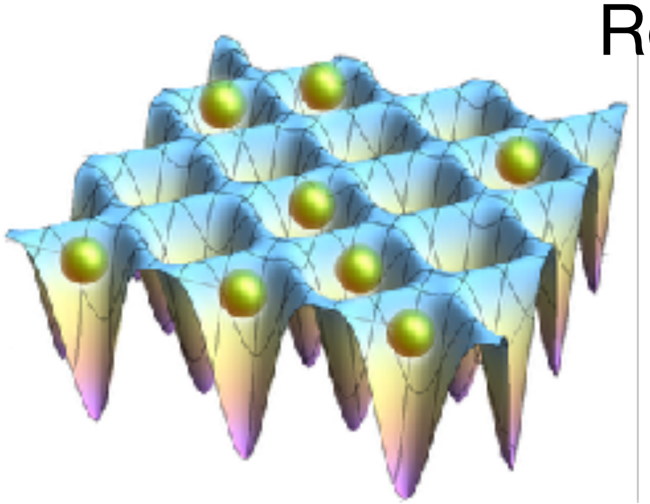
Cs137 9,192,631,770 Hz as a standard

Precision 10^{-16}

Development of next generation frequency Standard

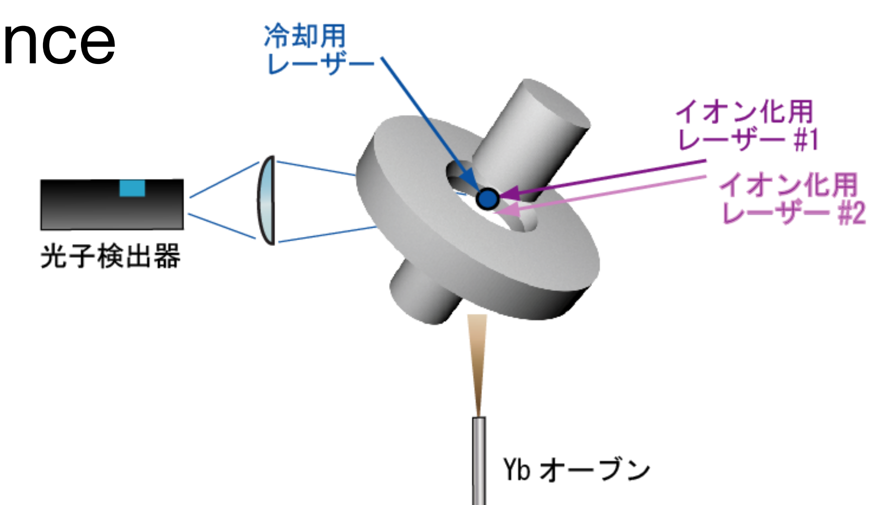
MW to Optical



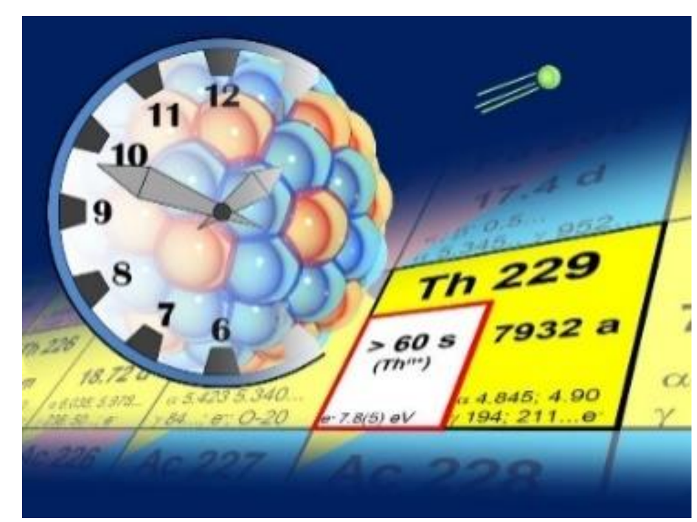


Optical lattice
Sr, Yb, Hg

400~500THz



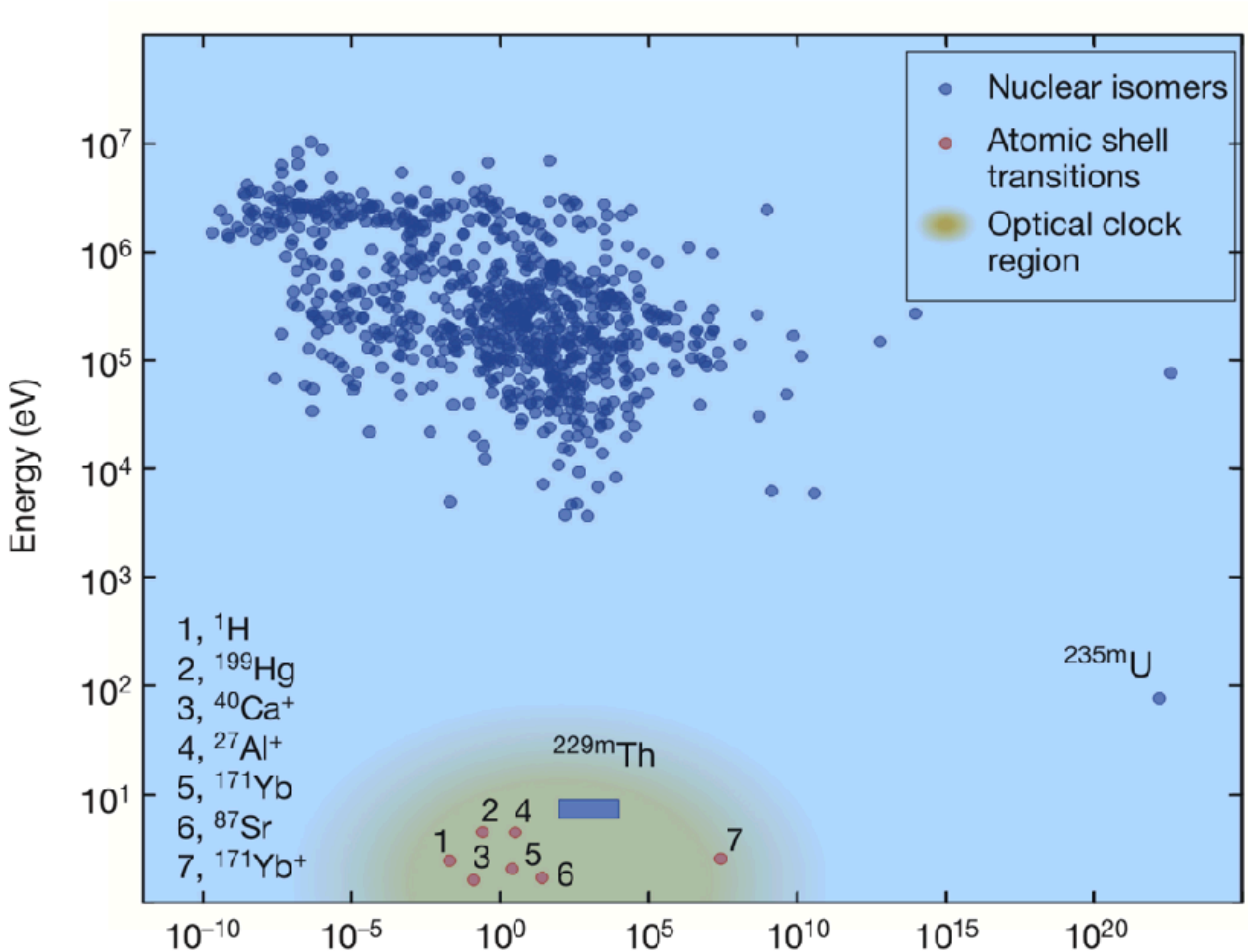
Ion trap
Yb



Ion trap, Solid state
Th

Precision 10^{-17}~10^{-18}

Precision 10^{-20}~10^{-21}



Lars von der Wense et al., Nature 2016

Why is precise clock needed?

- Communication speed
- Precision of GPS
  - In Futre, 1 μm precision could be achived.
- Gravitational sensor
  - General relativity 10^{-18} ~ 1cm resolution
  - Exploration of underground resources
- Fundamental Physics
  - Temporal variation of physical constants
  - Space probe（Darkmatter，Gravitational wave …)

Fundamental Physics with precision clock

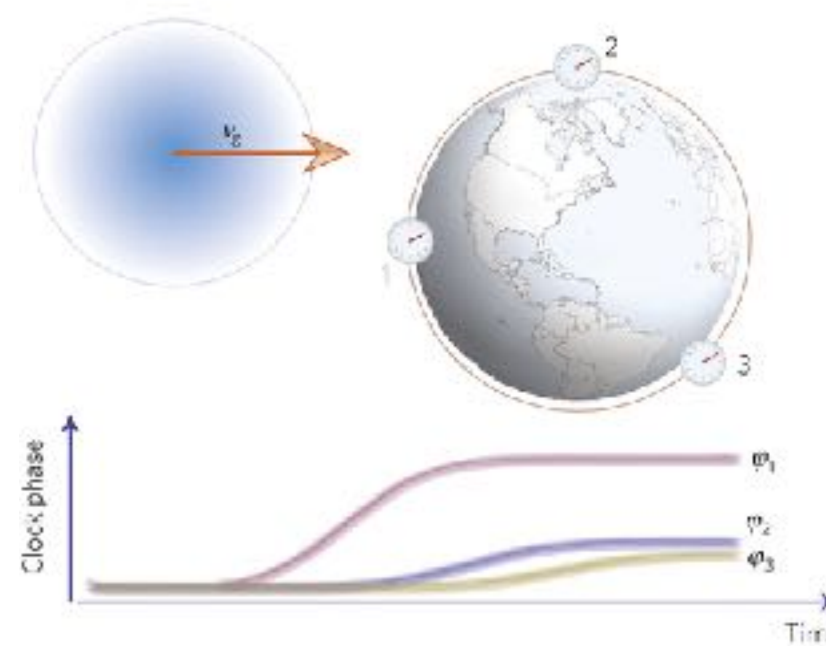
- Temporal variation of Physics constants
  - Some expanding universe model suggests possible variation of the fundamental
  - Yb atomic clock $$\frac{\dot{\alpha}}{\alpha} = (-2.0 \pm 2.0) \times 10^{-17}/\mathrm{yr}.$$

N. Huntemann et al., *Phys. Rev. Lett.* **113**, 210802 (2014).

- Sensitivity with Th-229 is improved by 10^5~10^6

V. V. Flambaum , *Phys. Rev. Lett.* **97**, 092502 (2006).

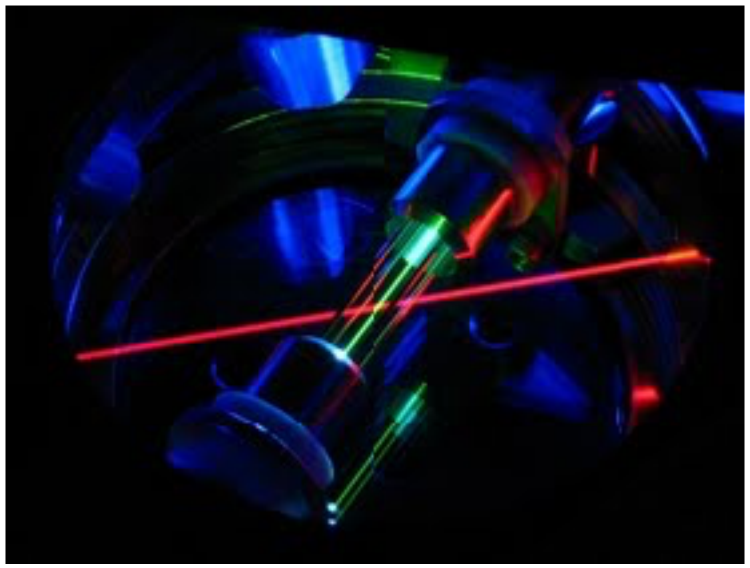
- Topological defect dark matter



A. Derevianko and M. Pospelov, Nat. Phys. **10**, 933 (2014)

Thorium clock

- **Ion trap (Kuzmich Group, Peak Group)**

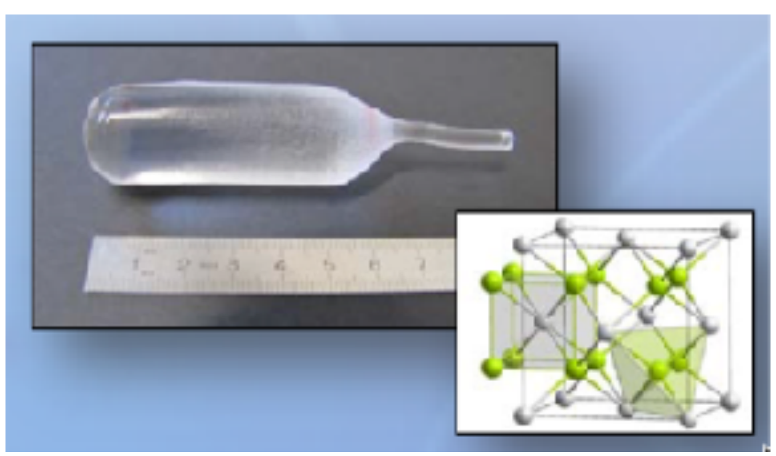


Trap Th^{3+}, Th^{+} in Paul trap

Single-ion can be laser cooled and detected

Electron bridge process

- **Crystal (Hudson Group, Scheme Group)**



Th doped UV transparent crystal

Th:LiCAF (Hudson Group, USA)

Th:CaF_2 (Schumm Group, AU)

Solid state nuclear clock

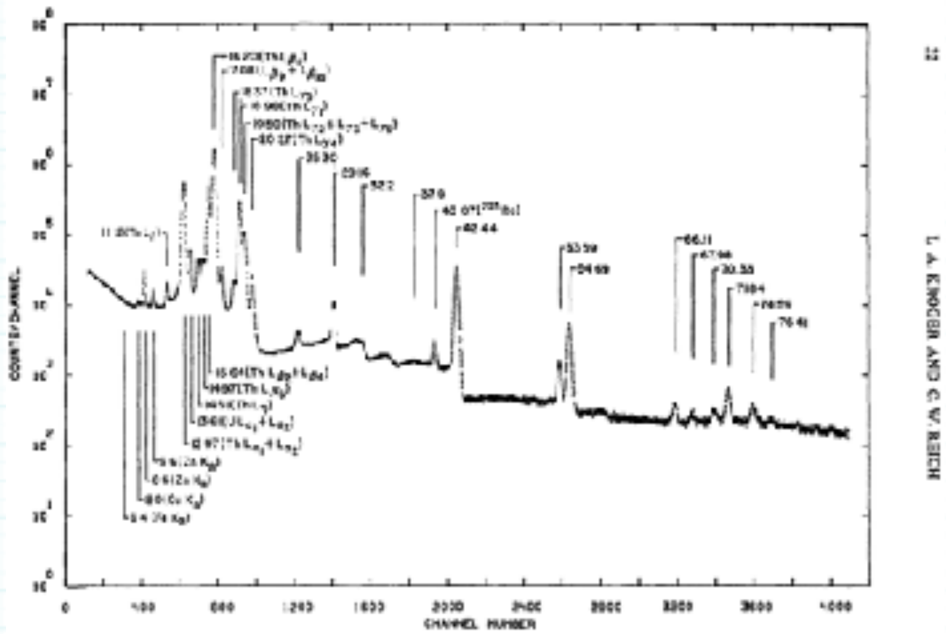
Crystal effect

J. Phys.:Condens. Mat. 21, 325403 (2009)

J. Phys.: Condens. Matter 26 (2014) 105402 (9pp)

History of hunting Th-229m isomer

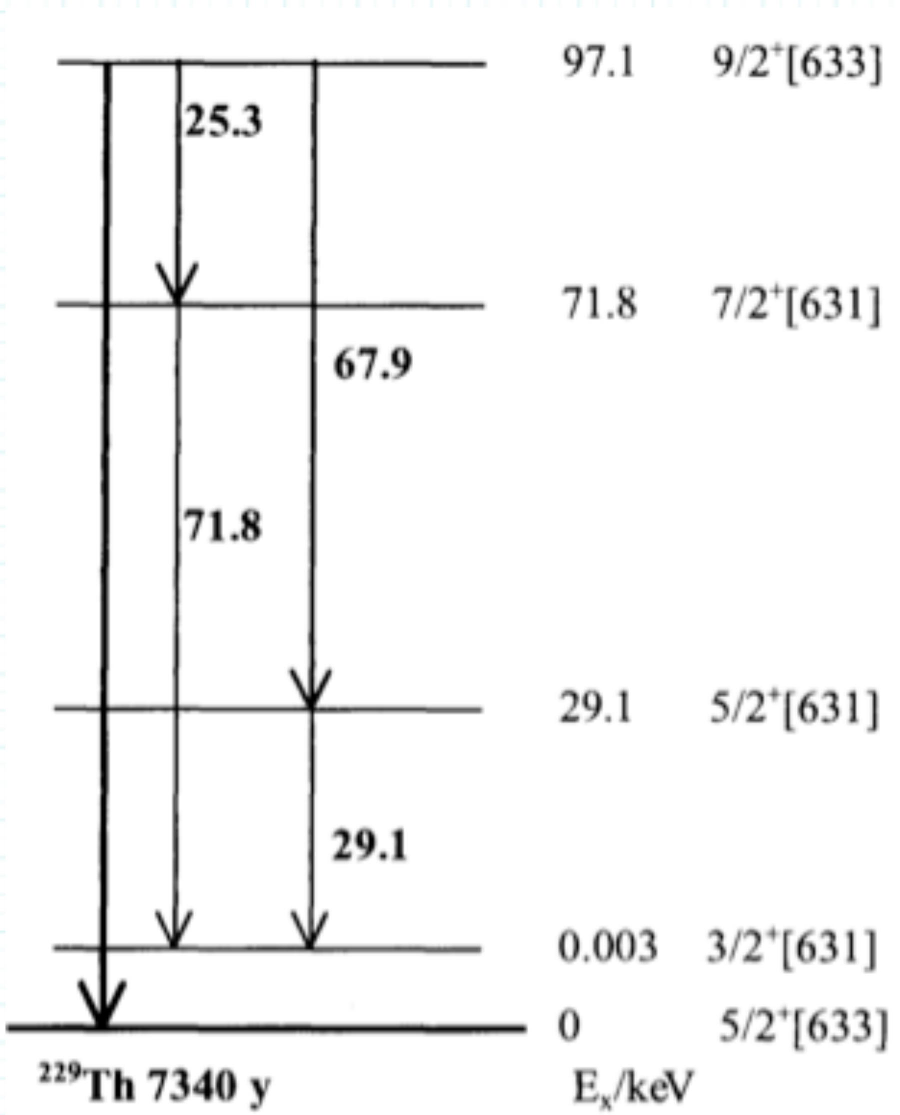
Indirect Method



Energy difference was obtained by combining γ spectrum with γ spectroscopy.

In 1994, 4 pairs of energy difference were avaraged.

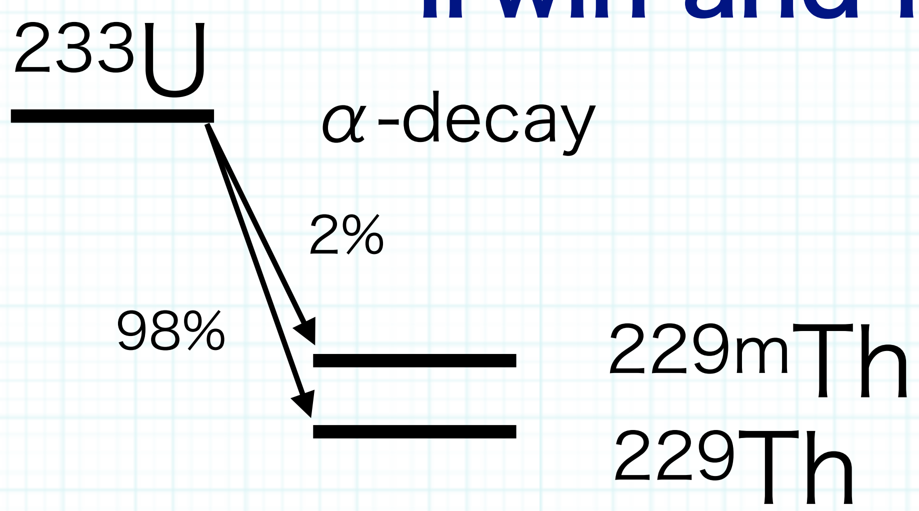
Ge detetor was used to report 3.5 eV ~ a few eh pair



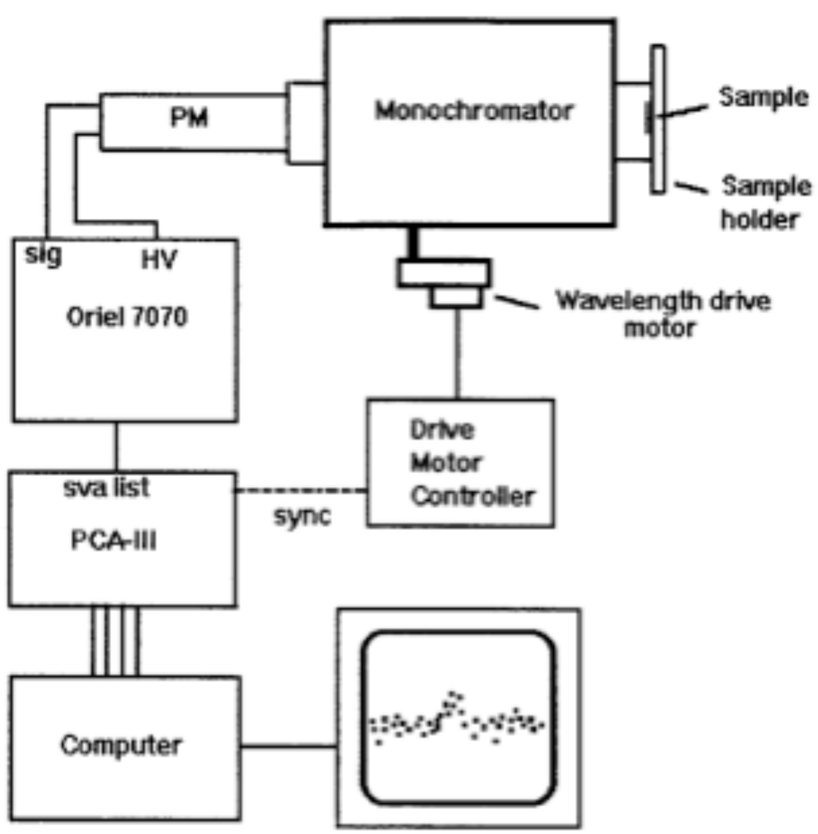
1976	Kroger & Reich	γ spectroscopy	<100 eV
1990	Reich & Helmer	γ spectroscopy	-1(4) eV
1994	Burke & Garret	Angular distribution from $^{230}Th(d,t)^{229}Th$	< 5 eV
1994	Helmer & Reich	γ spectroscopy	3.5(10) eV
2005	Guimaraes-Filho &Helene	reanalyze	5.5(10) ev
2007	Beck et al.	γ spectroscopy	7.6(5) eV 7.8(5) eV

Direct search (U-233)

Irwin and Kim, PRL79, 990 (1997)

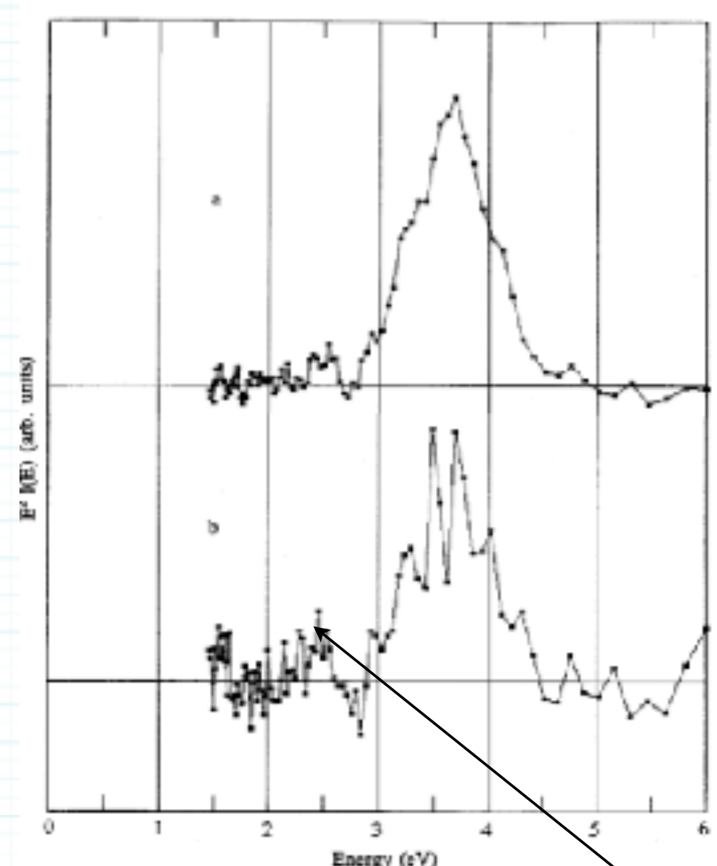


$$^{233}\mathrm{U} \rightarrow {}^{229m}\mathrm{Th} \rightarrow {}^{229}\mathrm{Th}$$

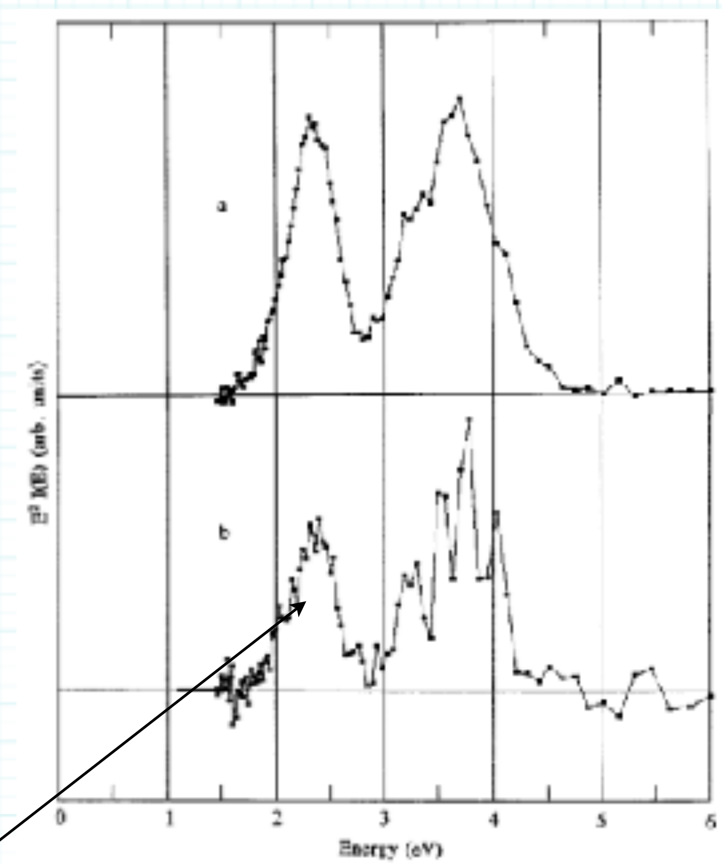


Richardson et al. PRL80, 3206 (1998)
Same peak

Sample1
100uCi U-233 on Kapton



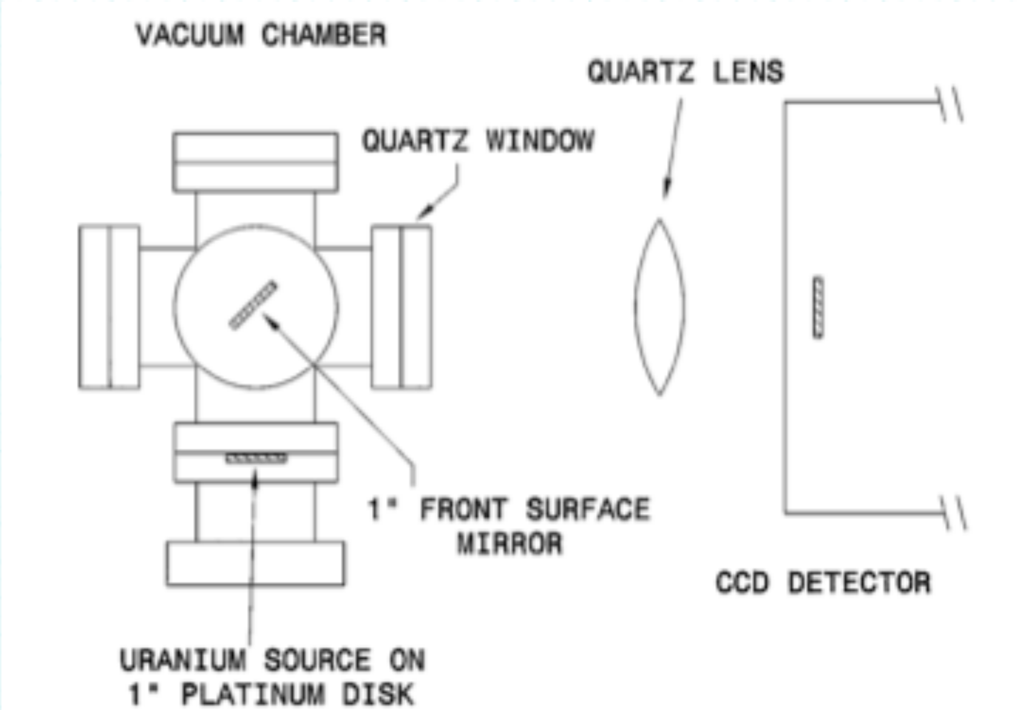
Sample2
300uCi U-233 on filter paper

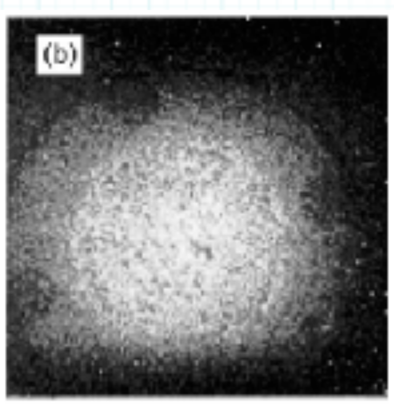


inelastic electron bridge?

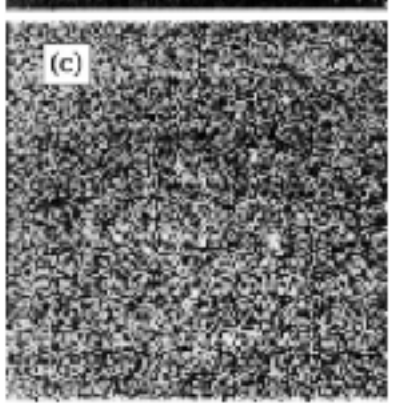
Signal Disappear in Vacuum

Utter et al., PRL 82, 505 (1999)
Shaw et al, PRL82, 1109 (1999)





Atmosphere



Vacuum（5X)

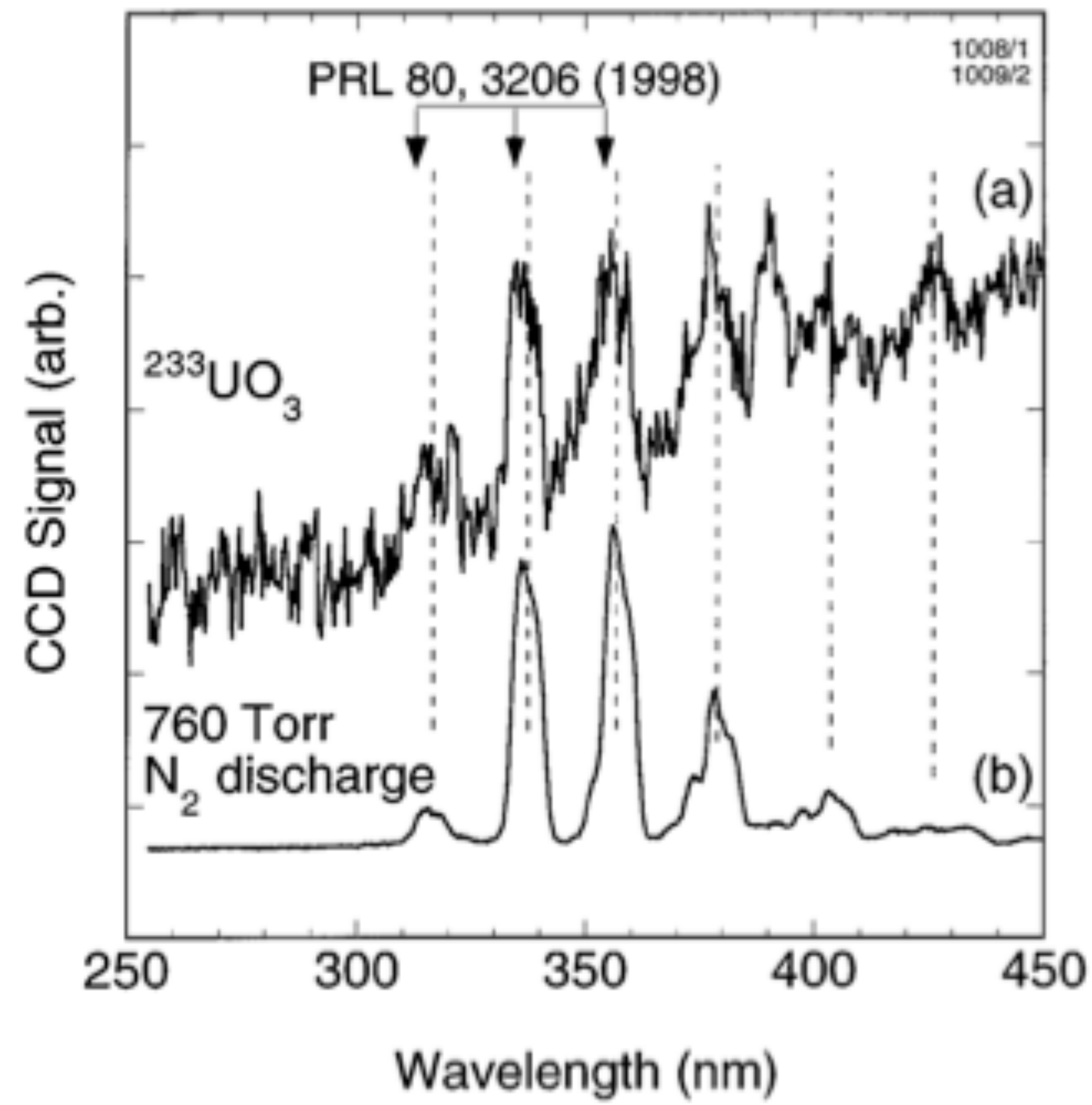


FIG. 1. (a) Spontaneous luminescence spectrum from a $^{233}UO_3$ powder sample (46 h integration, 0.5 mm slit). (b) Emission spectrum of an atmospheric pressure N_2 discharge, under the conditions of (a), except for a 1 s integration time. The traces are offset vertically for clarity. The arrows mark the line positions reported in Ref. [2].

“Nuclear clock” revived Th-229

EUROPHYSICS LETTERS 15 January 2003

Europhys. Lett., **61** (2), pp. 181–186 (2003)

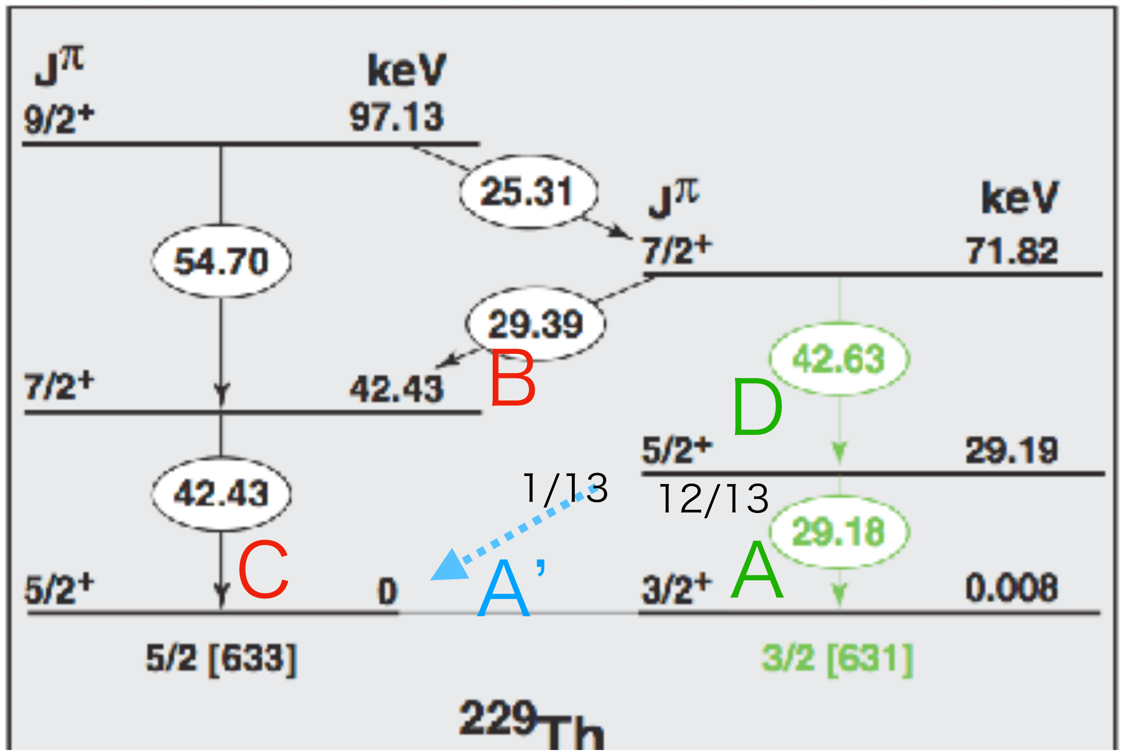
Nuclear laser spectroscopy of the 3.5 eV transition in Th-229

E. PEIK(*) and CHR. TAMM

Physikalisch-Technische Bundesanstalt - Bundesallee 100
38116 Braunschweig, Germany

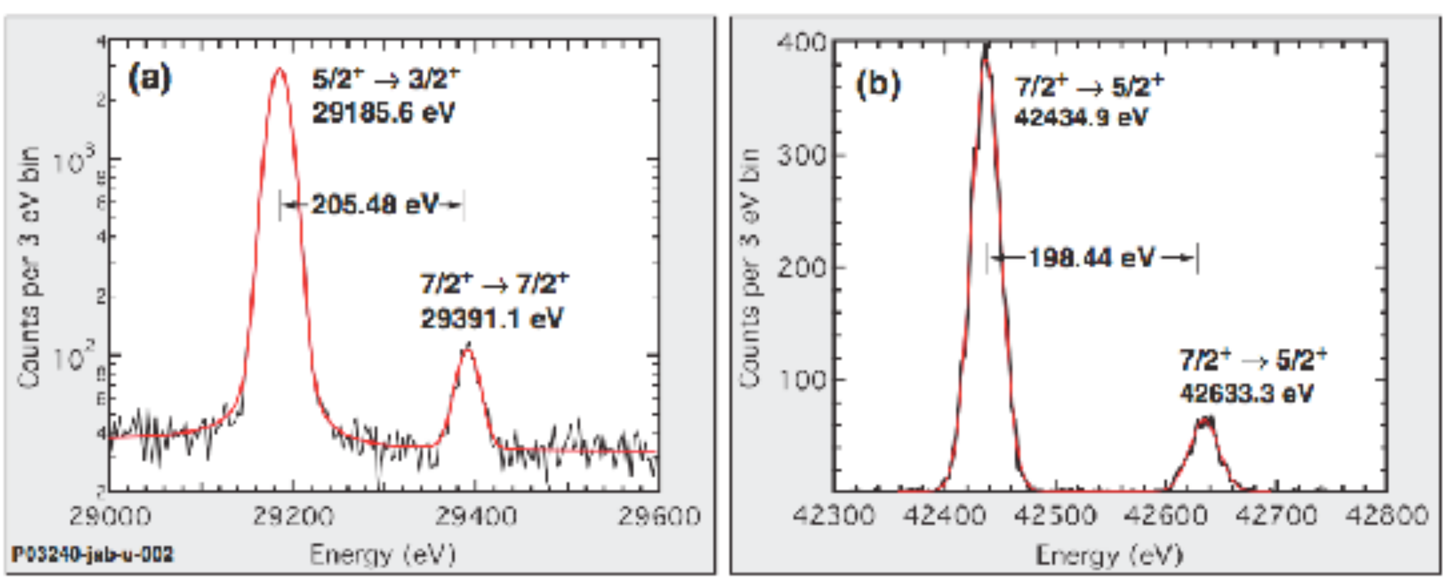
(received 17 June 2002; accepted in final form 11 November 2002)

Indirect method



- Utilize two pair of energy doublet.
- XRS micro calorimeter (ΔE ~ 200 eV)

$$E = (B + C) - (A + D)$$
$$= (B - A) - (D - C)$$
$$= 205.48 - 198.44 = 7.04$$



A B C D

Beck et al. PRL 98, 142501 (2007)

1976	Kroger & Reich	α-decay	<100 eV
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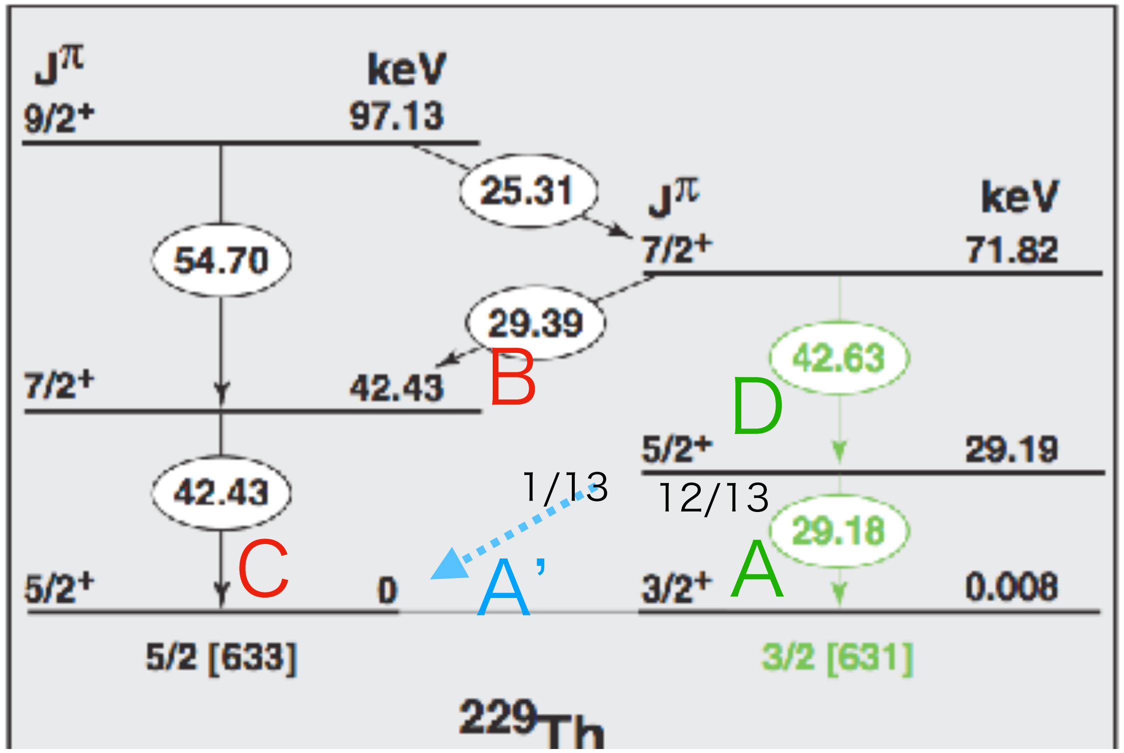
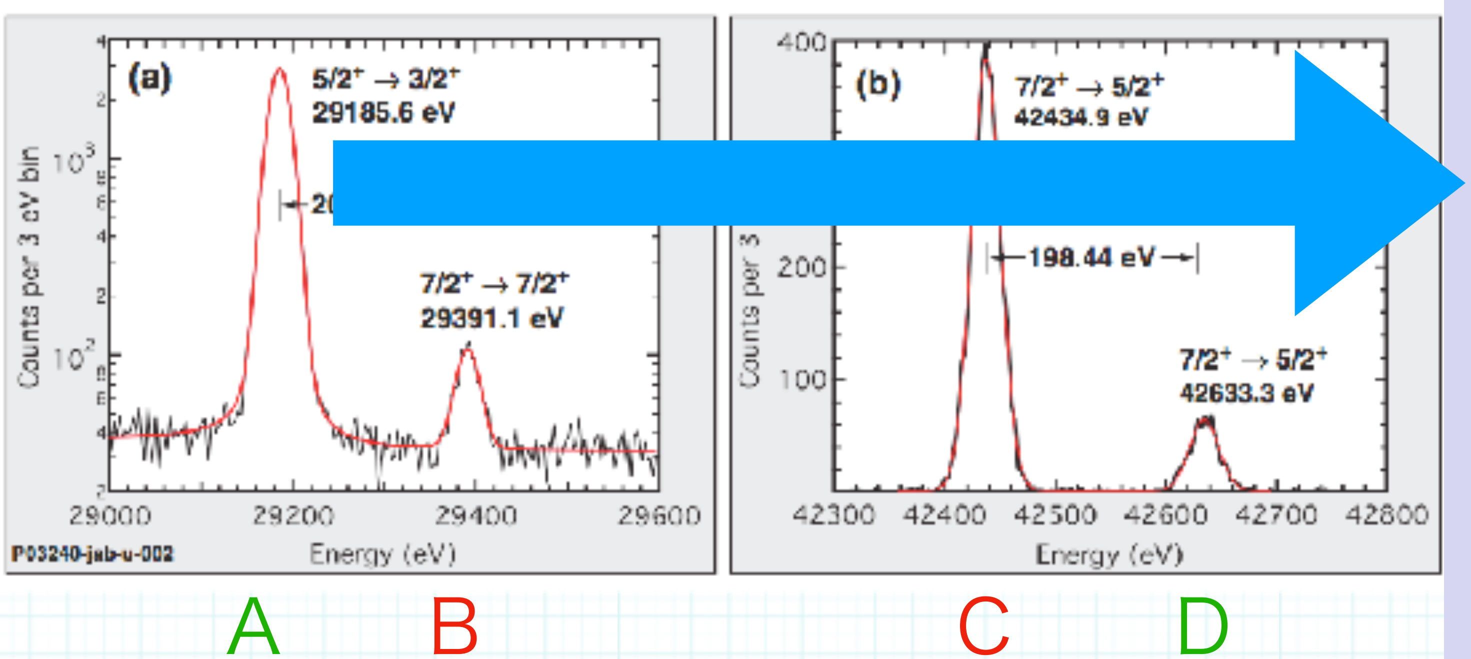


- Utiliz
- XRS r

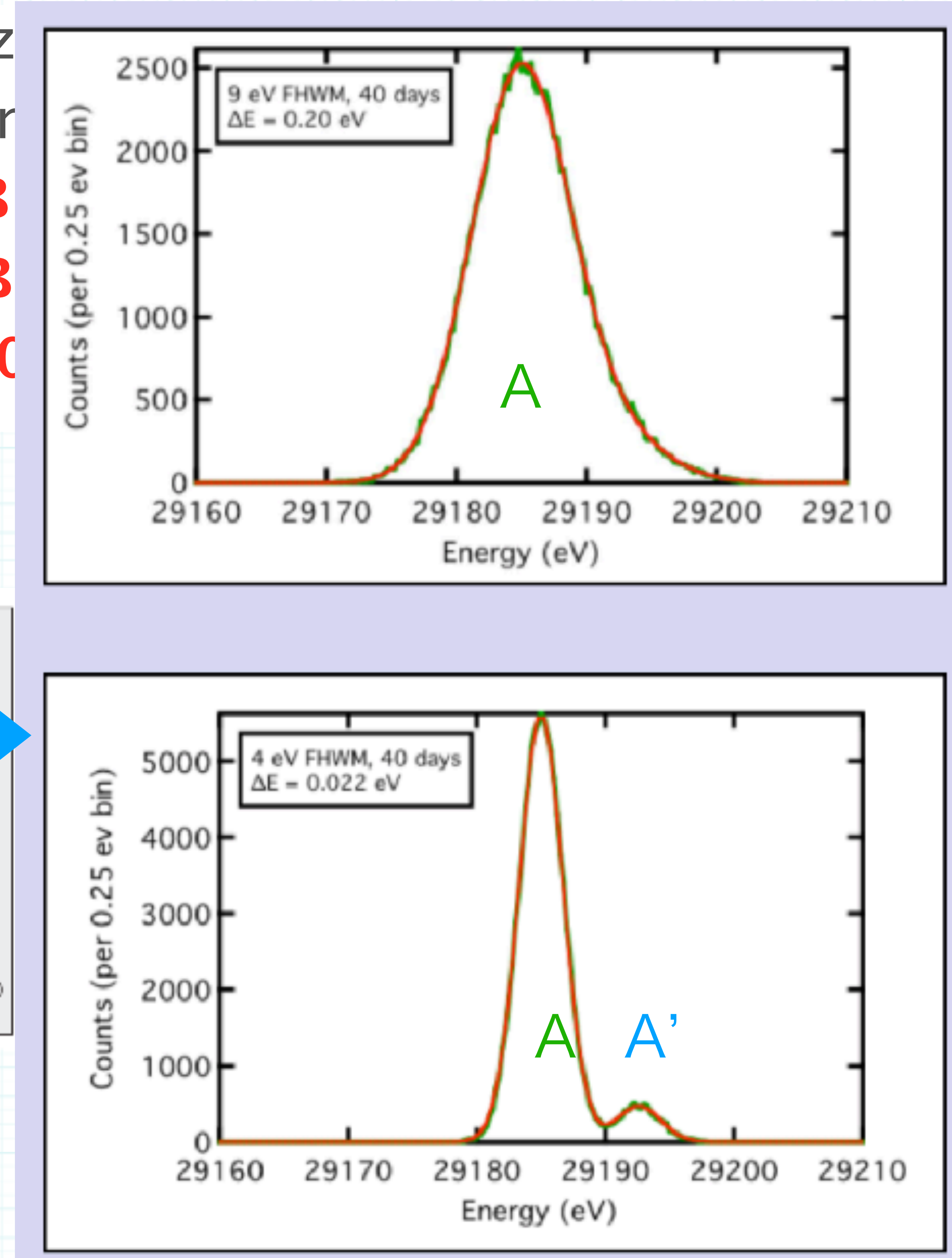
E = (B

= (B

= 20

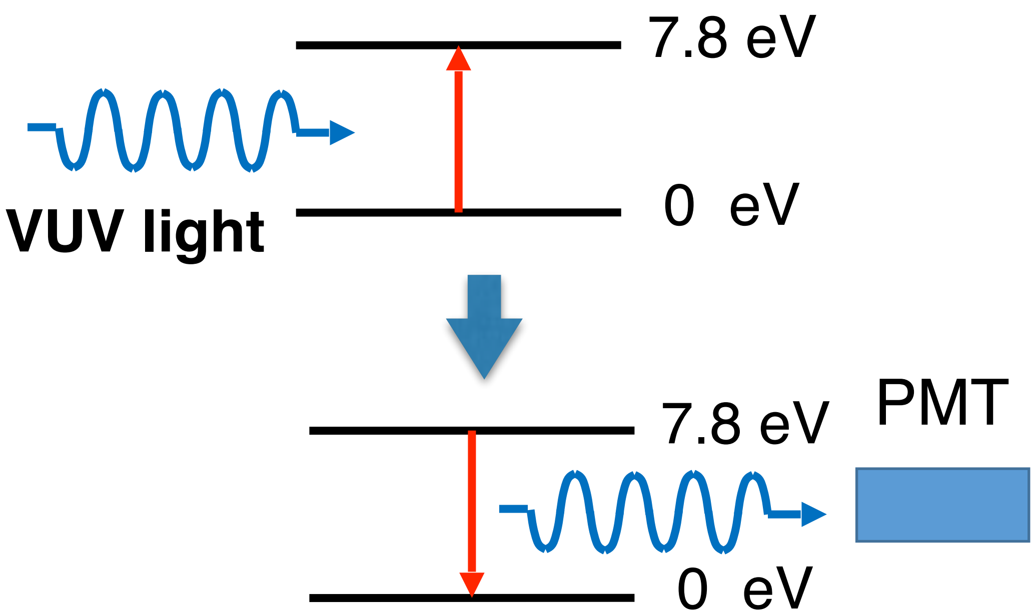


Beck et al. PRL 98, 142501 (2007)



Direct method

VUV light source

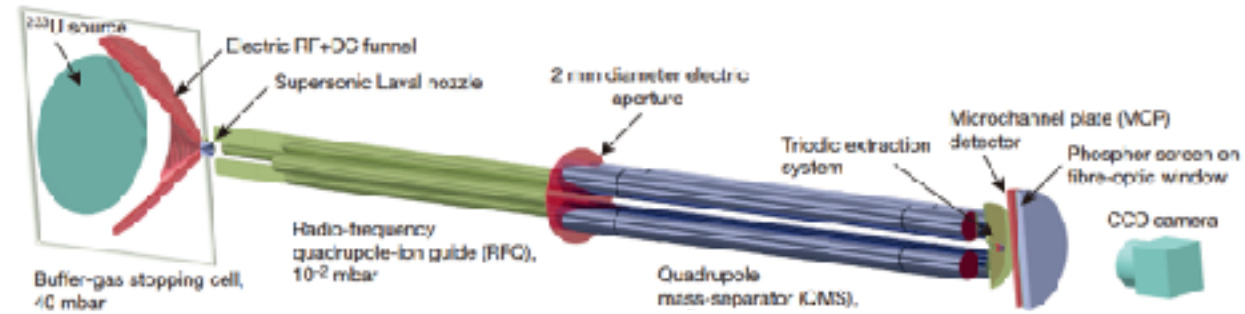


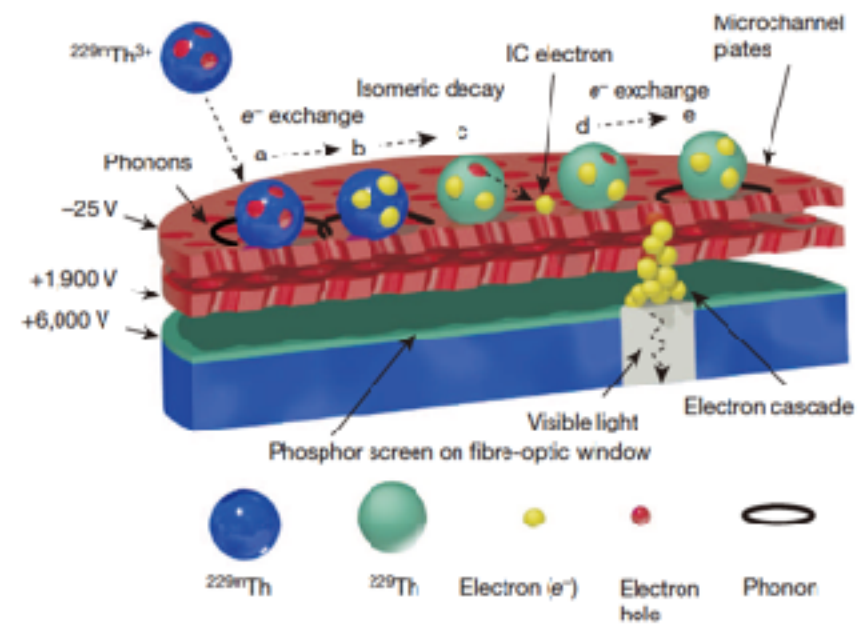
J. Jeet et al., PRL 114, 253001 (2015)
E=7.29 - 8.86 eV at ALS

A. Yamaguchi et al.,
New J. Phys. 17 (2015) 053053
E=3.54 - 9.54 eV at MLS

S. Stellmer et al., arXiv 1803.09294
E=7.5- 10 eV at MLS
Life ~ 1 sec

Th Ion beam





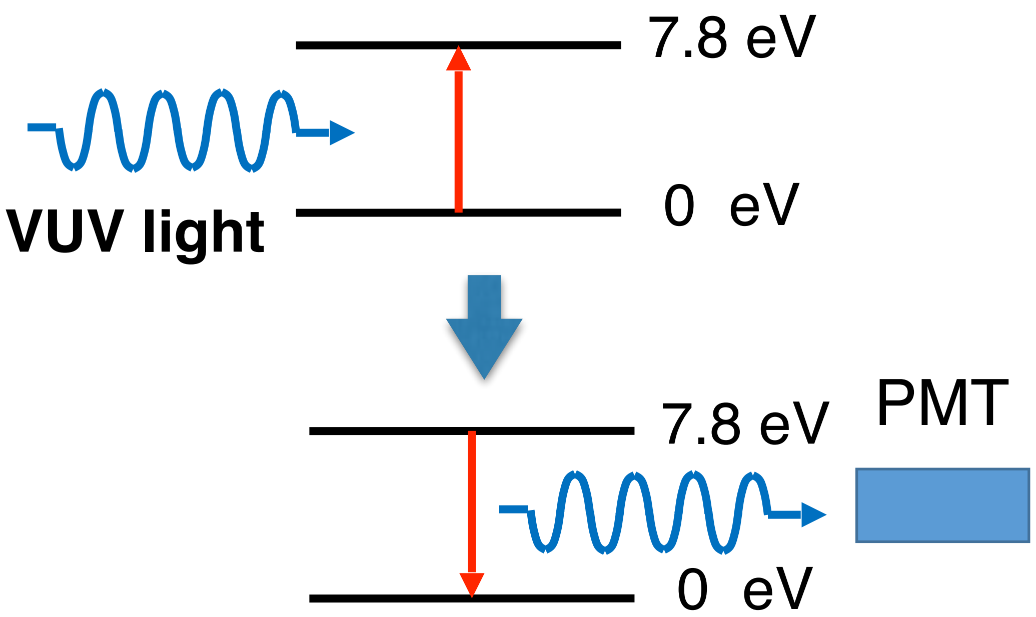
Lars von der Wense et al.,
Nature 533 (2016)

E=6.3~18.3 eV

Physics World 2016 Breakthrough of the Year
3rd Place

Direct method

VUV light source



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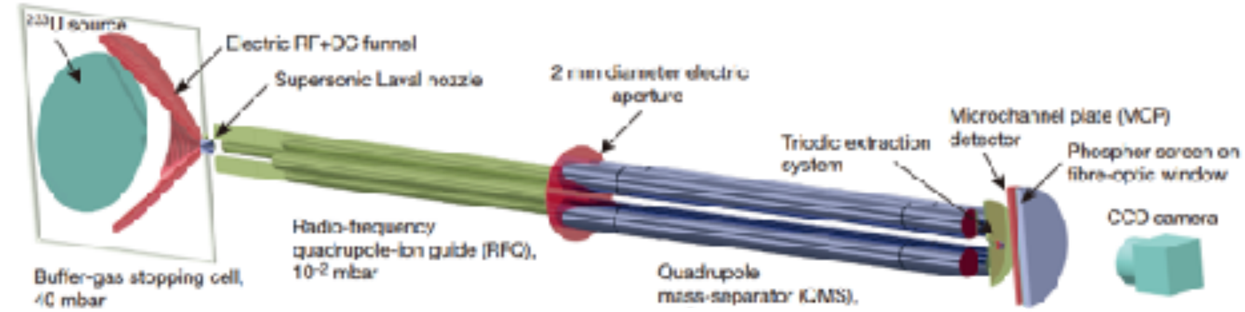
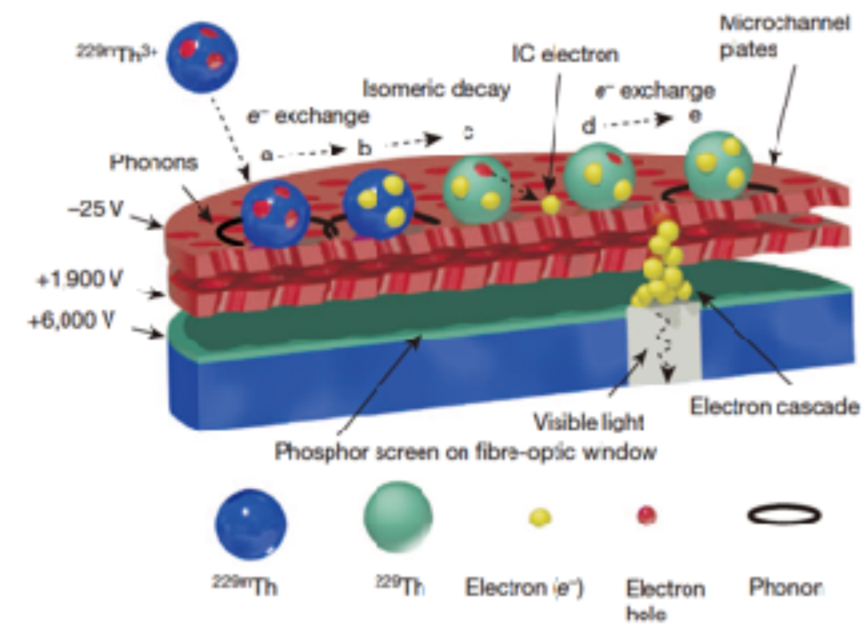




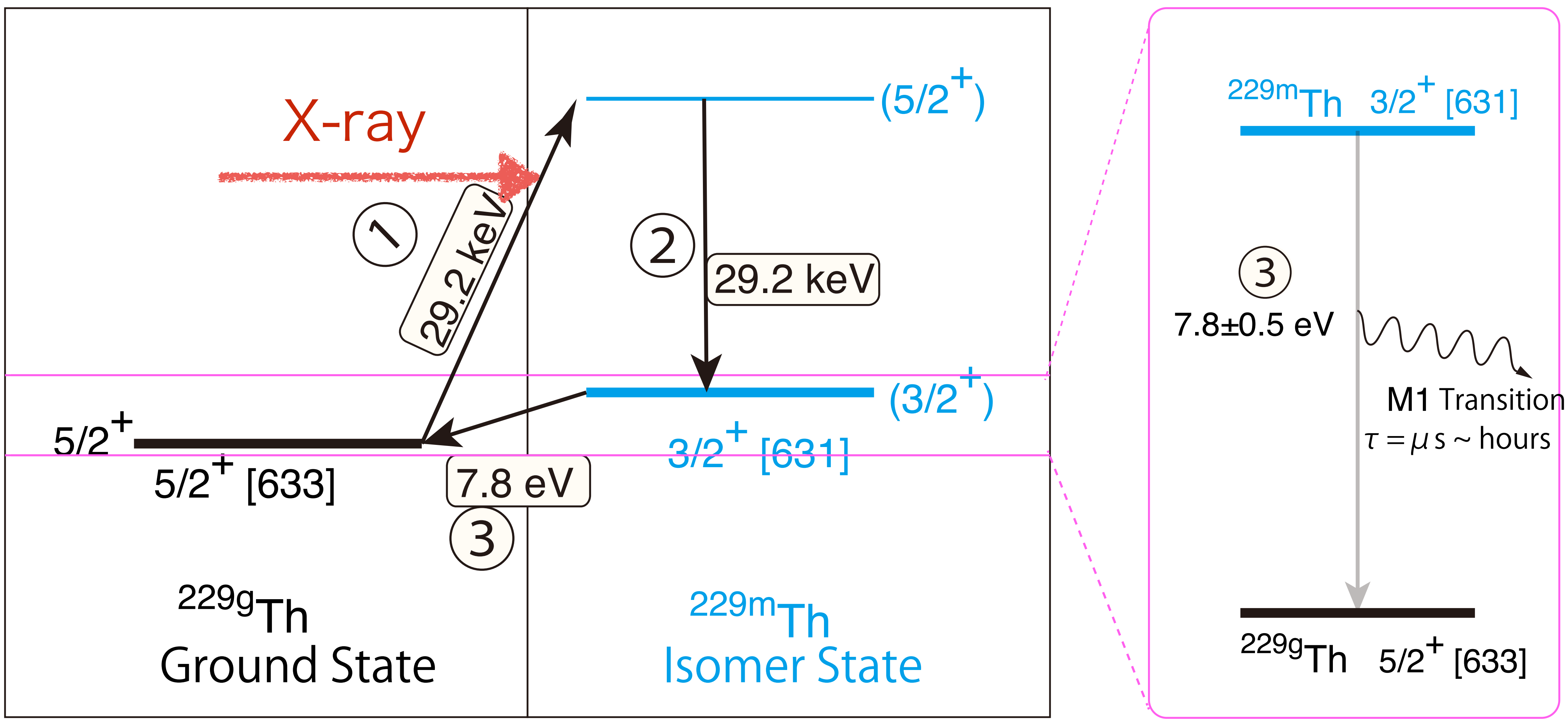
Lars von der Wense et al.,
Nature 533 (2016)

E=6.3~18.3 eV

Physics World 2016 Breakthrough of the Year
3rd Place

No VUV signal was observed yet!

A new method using intense X-ray source



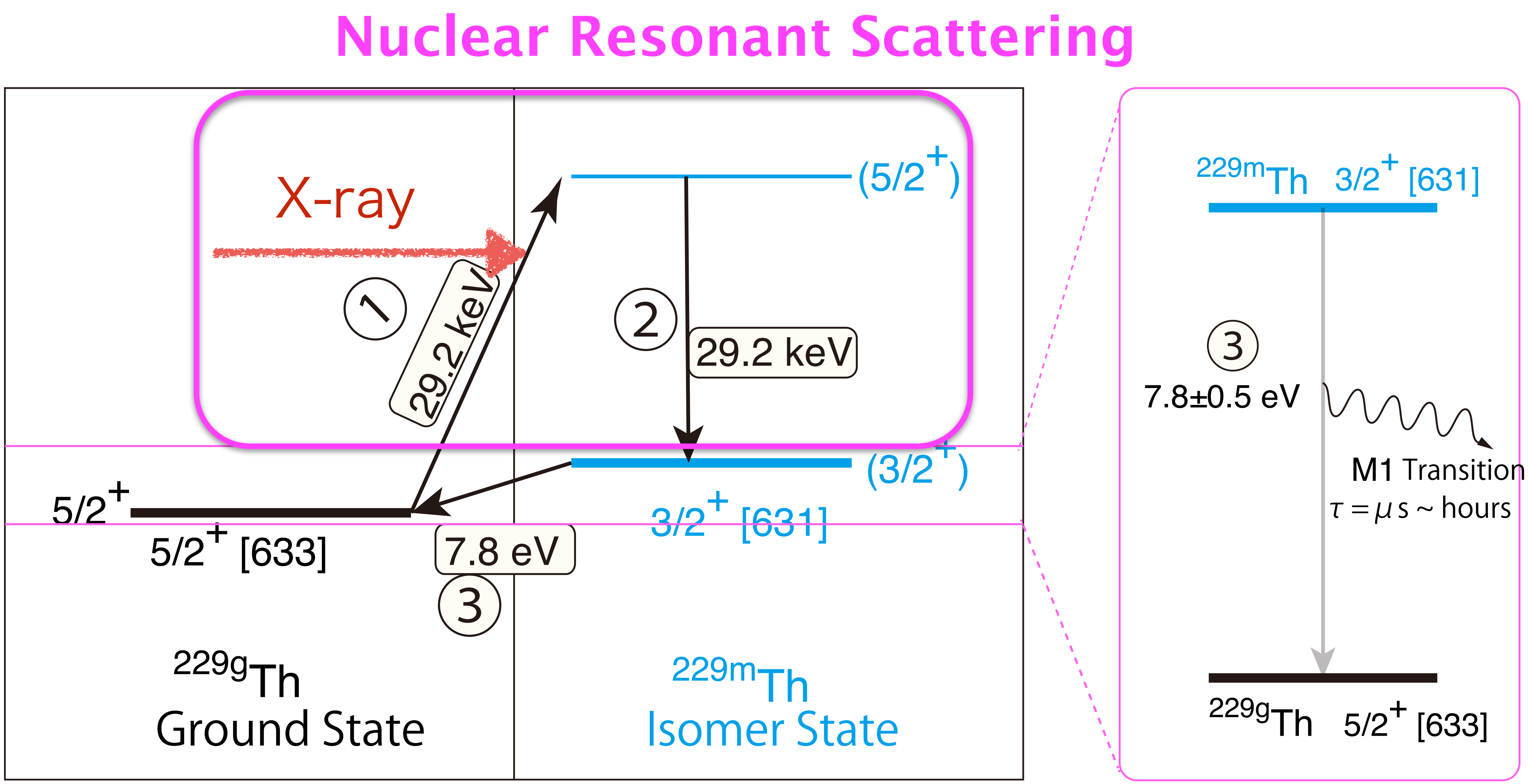
① Excite **Definitely using the known level**

② Confirmation **Using NRS signal**

③ Measure **VUV measurement**

Originally proposed in Tkalya et al., PRC 61, 064308, 2000

A new method using intense X-ray source



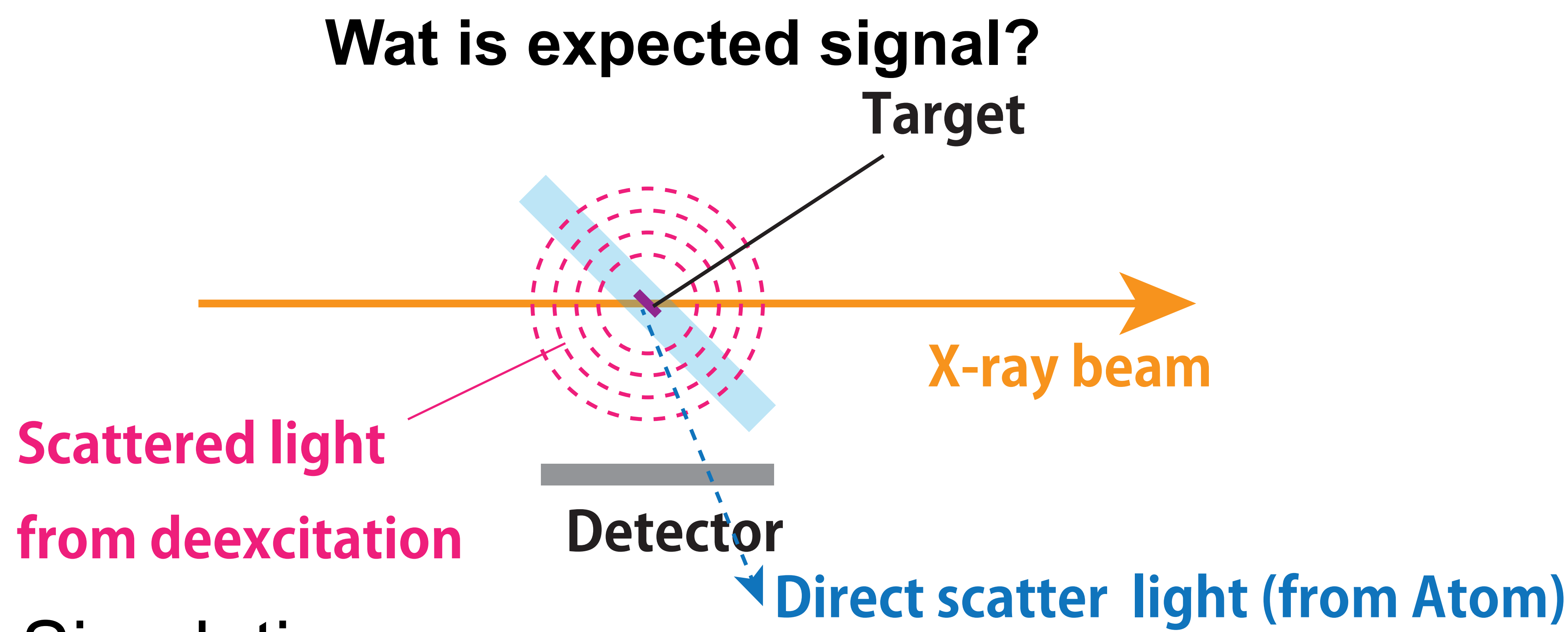
① Excite **Definitely using the known level**

② Confirmation **Using NRS signal**

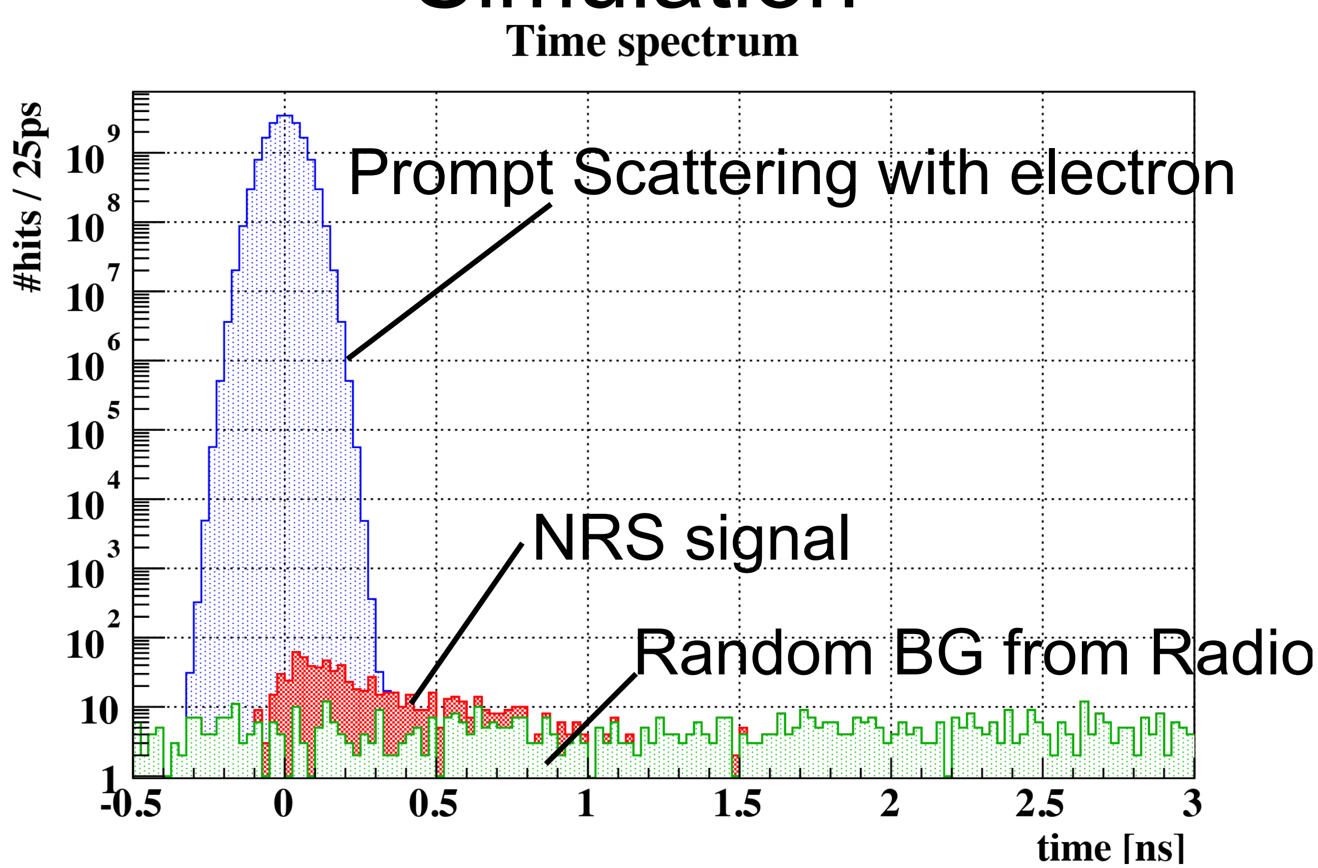
③ Measure **VUV measurement**

Originally proposed in Tkalya et al., PRC 61, 064308, 2000

Nuclear Resonant Scattering (NRS)



Simulation



Difficulties

- **Very short lifetime ($T_{1/2}$ 100~200p**
- **Rare event**
- **Random radiation background**

Baruch De Spinoza

Last part of “Ethica”

Sed omnia praeclara tam difficilia, quam rara sunt.

But all things excellent are as difficult as they are rare.

しかし，すべて高貴なものは稀であるとともに困難である

Inspired by T. Komatsubara (Rare K-decay)

Th collaboration

- **Okayama University**
  - **S.Okubo, H.Hara, T.Hiraki, T. Masuda, Y.Miyamoto, K.Okai, N.Sasao, S. Uetake, A.Yoshimi, K.Yoshimura, M.Yoshimura**
- **Riken**
  - **A.Yamaguchi, H. Haba, Yokokita**
- **Osaka University**
  - **Y.Kasamatsu, Y.Yasuda, Y.Shigekawa**
- **Tohoku University IMR**
  - **K.Konashi, M.Watanabe**
- **SPring-8**
  - **Y.Yoda, K. Tamasaku**
- **Kyoto University**
  - **M.Seto, K.Kitao, Y.Kobayashi, R.Masuda**
- **TU Wien**
  - **T. Schumm, S.Stellmer**

SPring-8 Experiment

SPring-8 Experiment

Monochro Meter → Experimental Hutch

Bunch mode	203 bunches
Bunch interval	23.6 nsec
Photon flux	4 x 10^{13} photon/s
Line width	4 eV
Pulse width	~35 PS

SPring-8 Experiment

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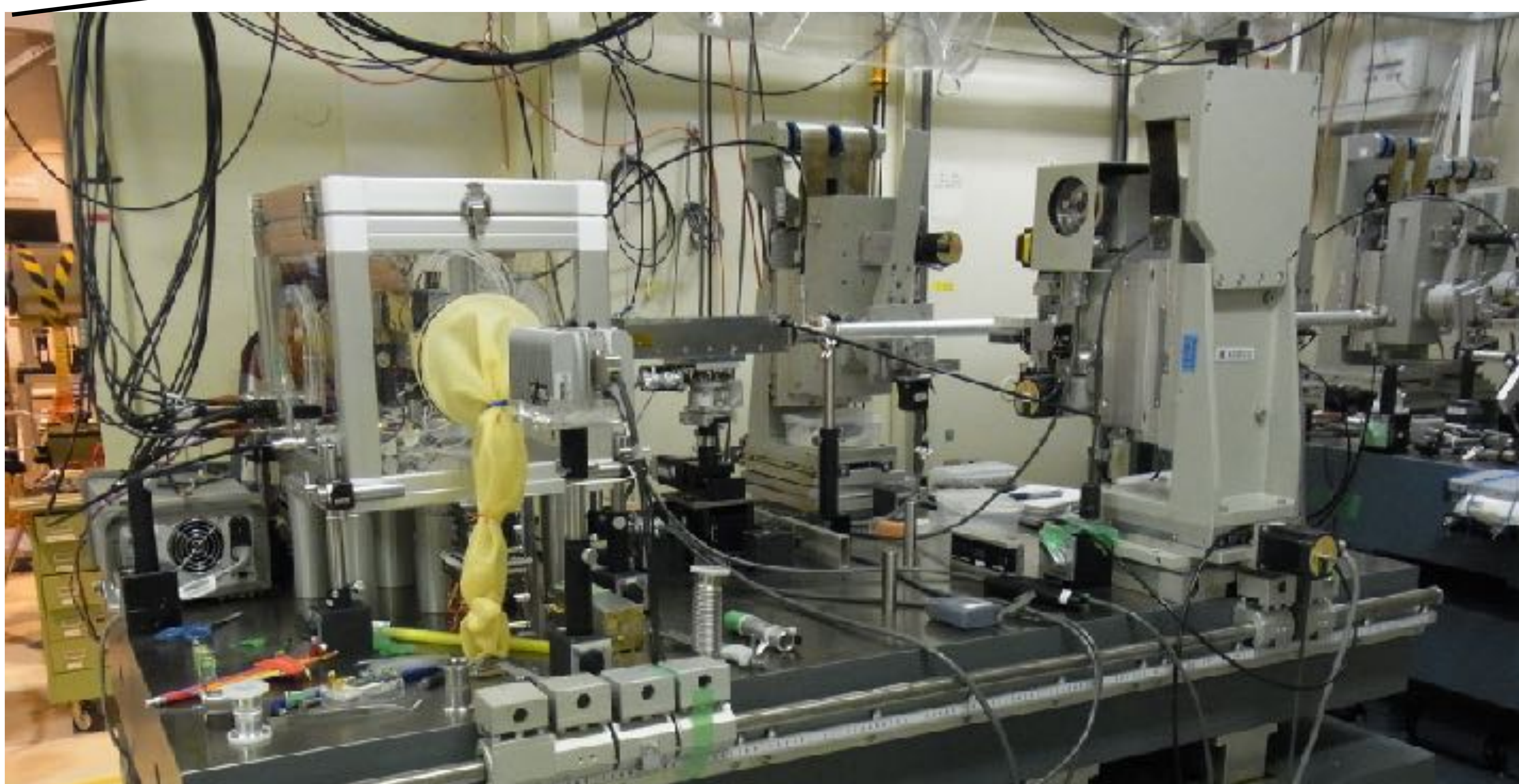
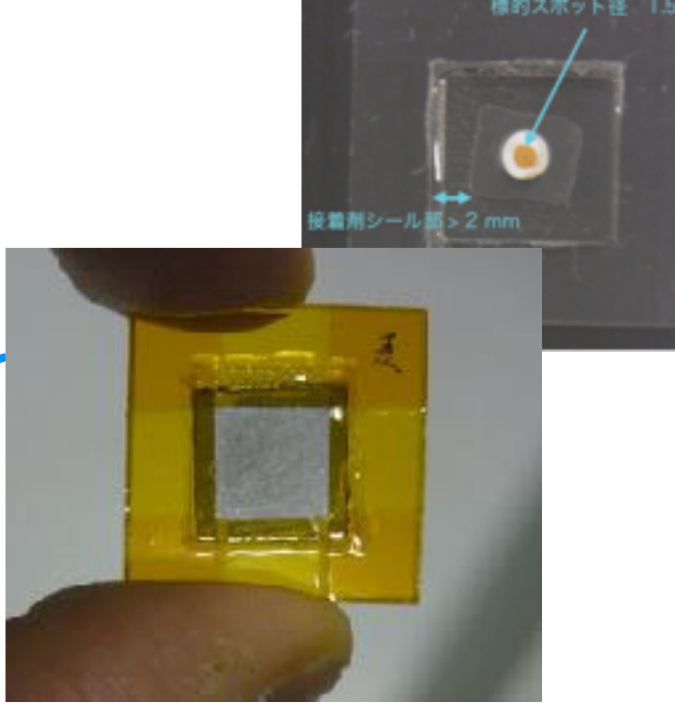
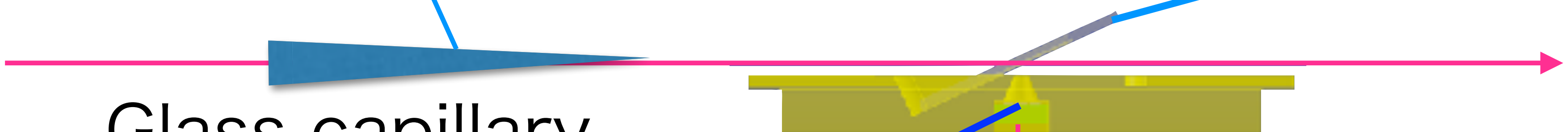


Experimental setup

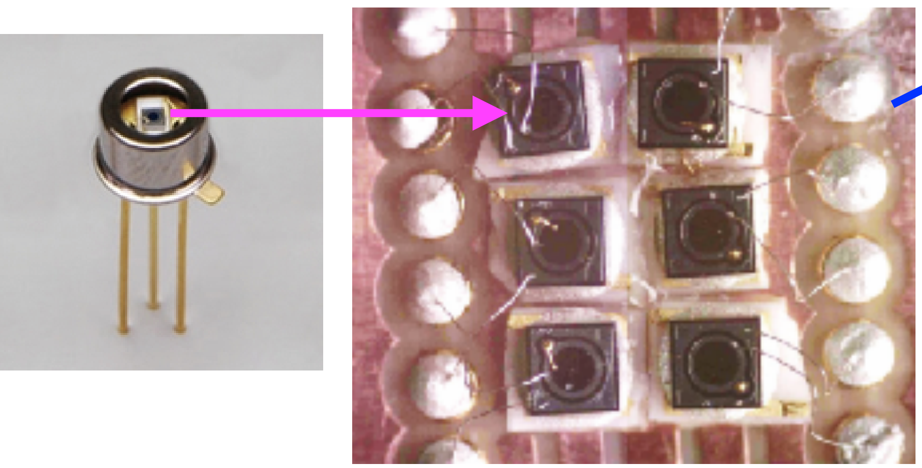




Glass capillary

Th-229 Target

ϕ 1.5

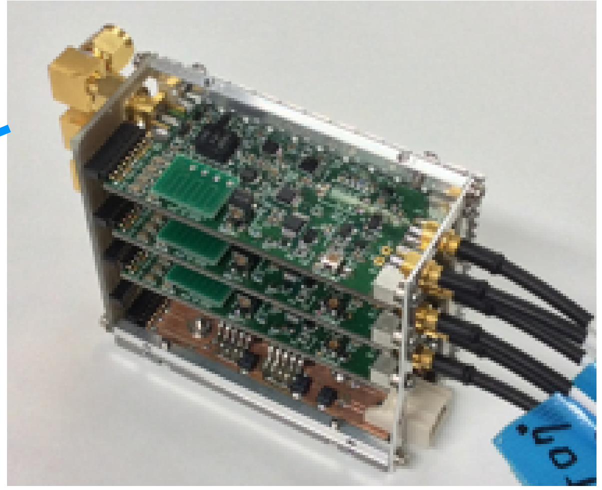


APD array

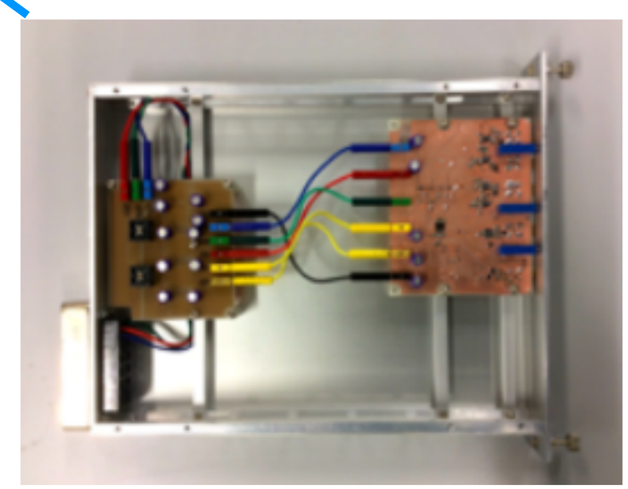
HPK S12053-05 (ϕ0.5)

PreAmp

CFD

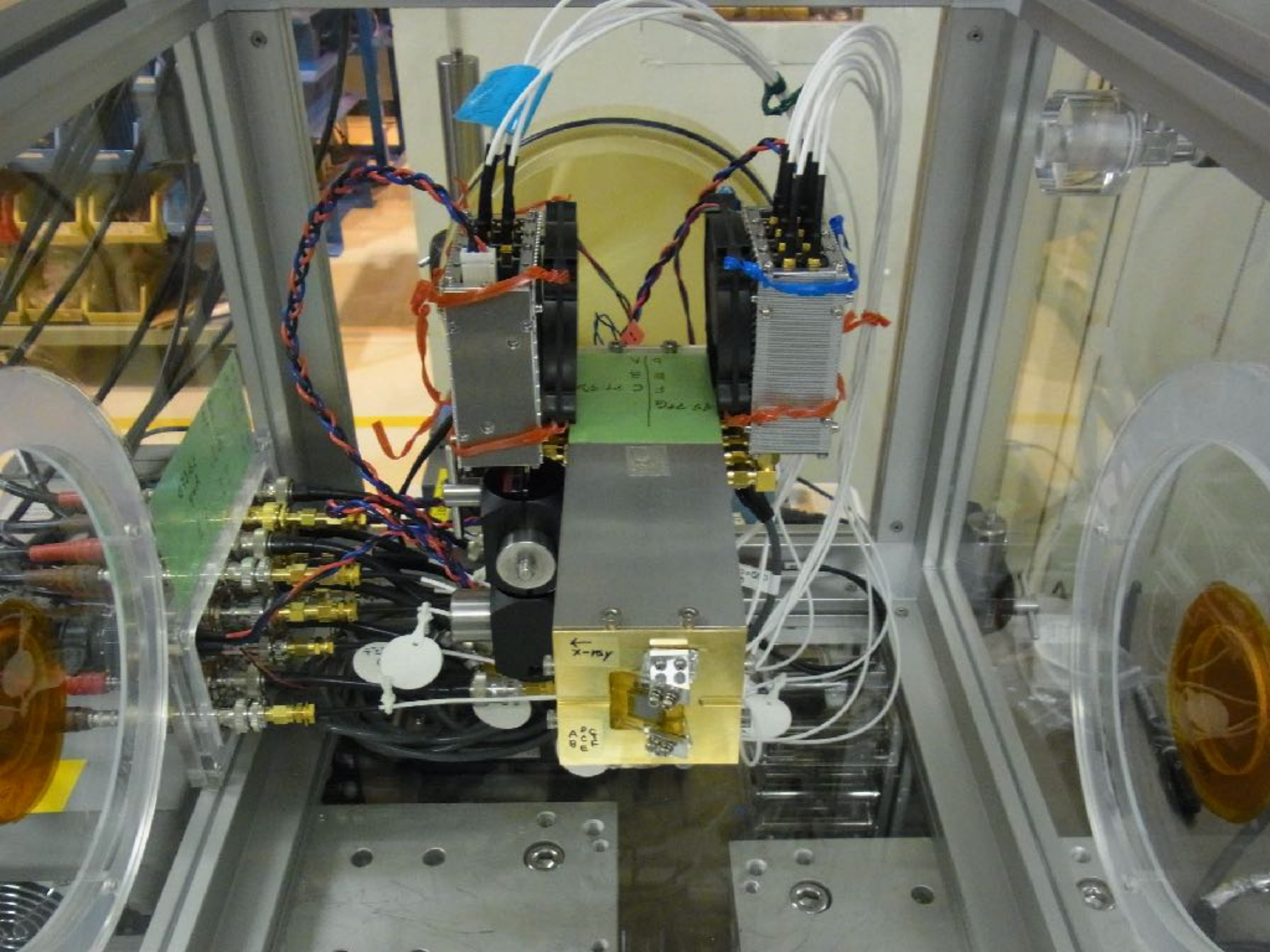


ATC



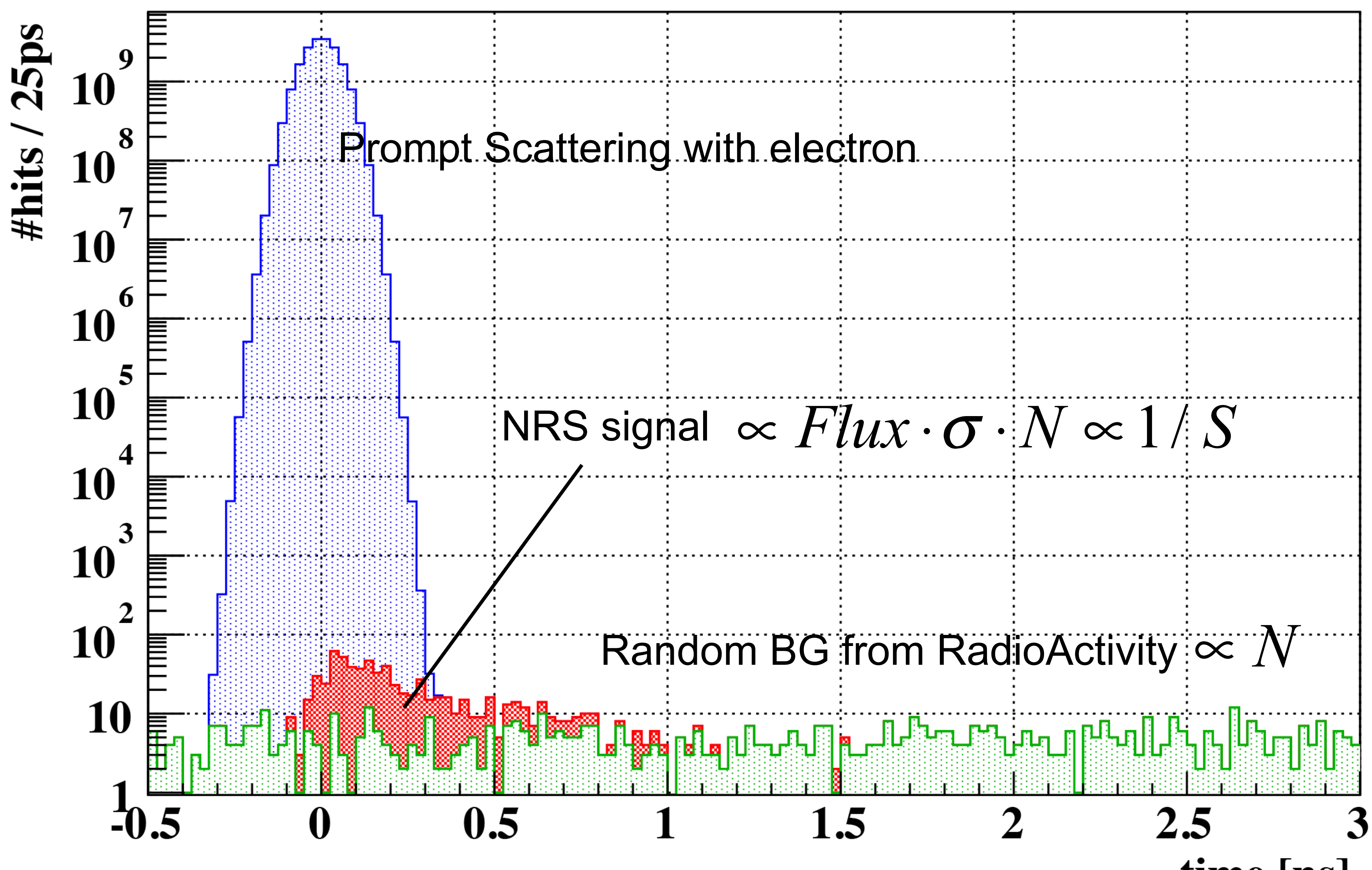
Multi-hit TDC

- MAX DAQ rate ~ 7 Mcps
- Acceptance 0.7 %



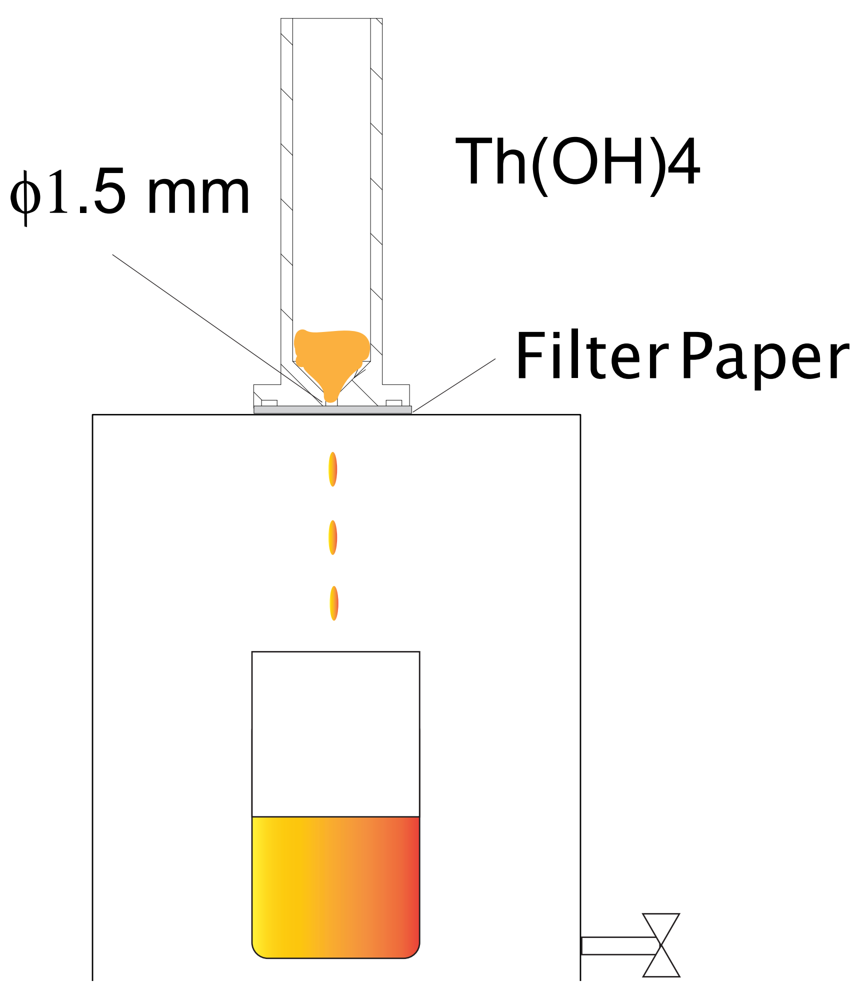
X-ray

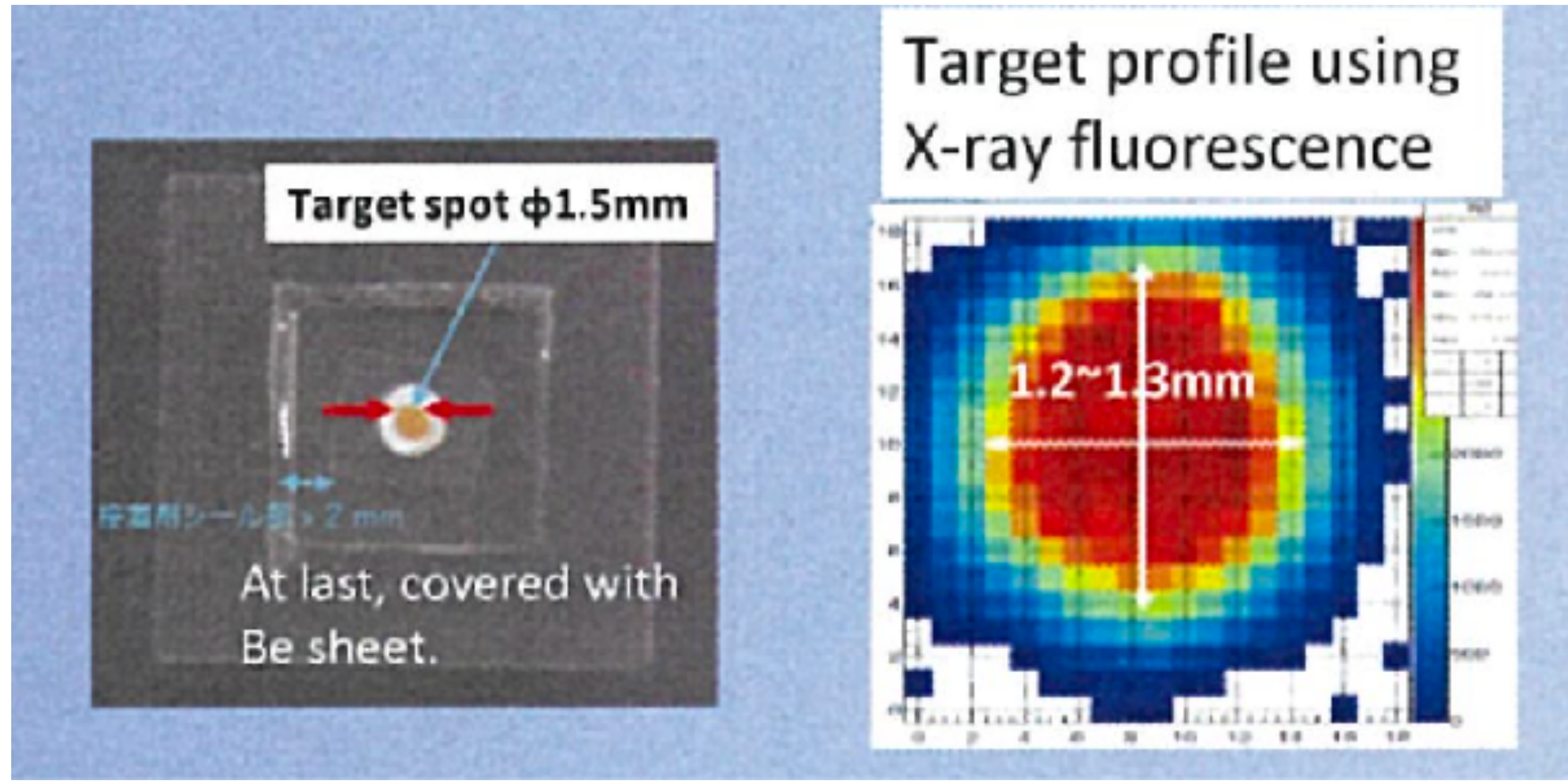
To detect NRS signal with good S/N

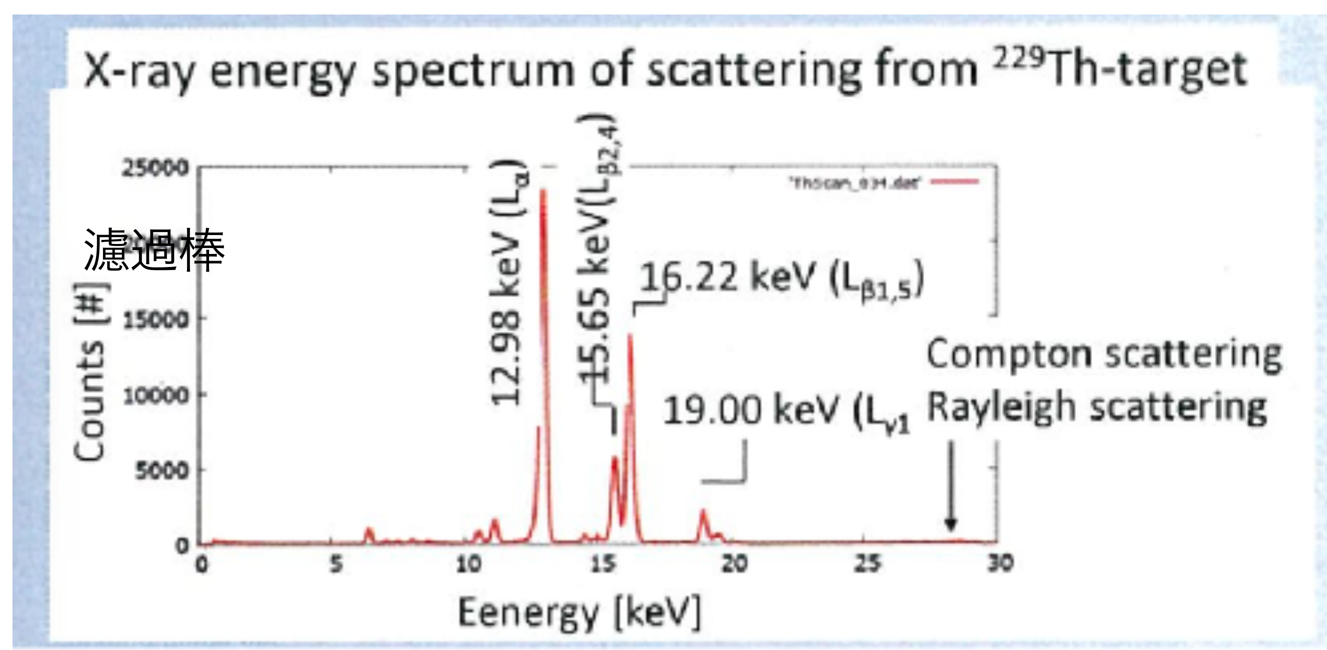


Thorium Target production

Precipitation method

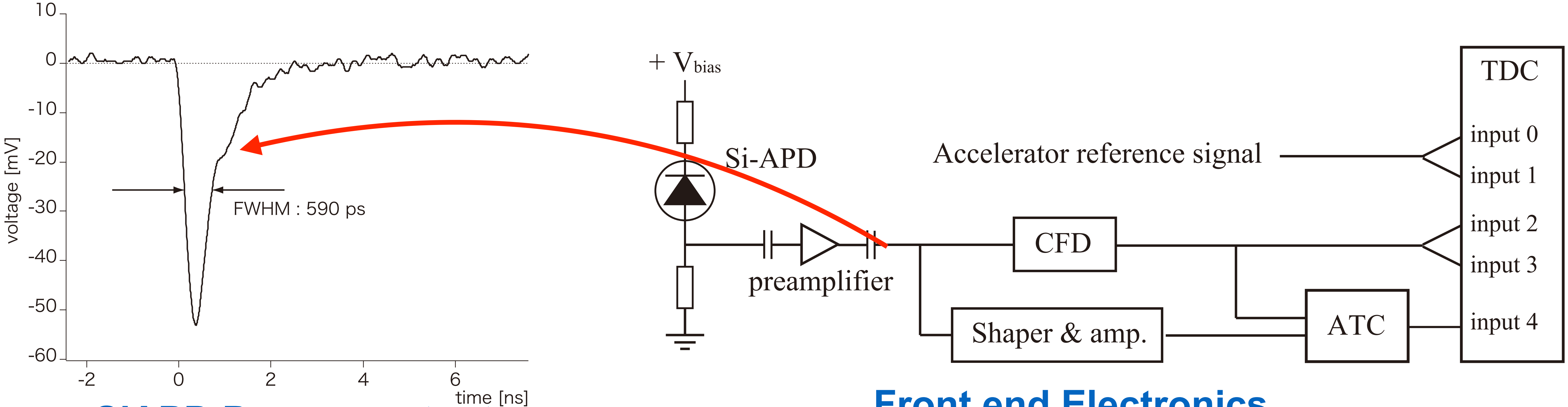






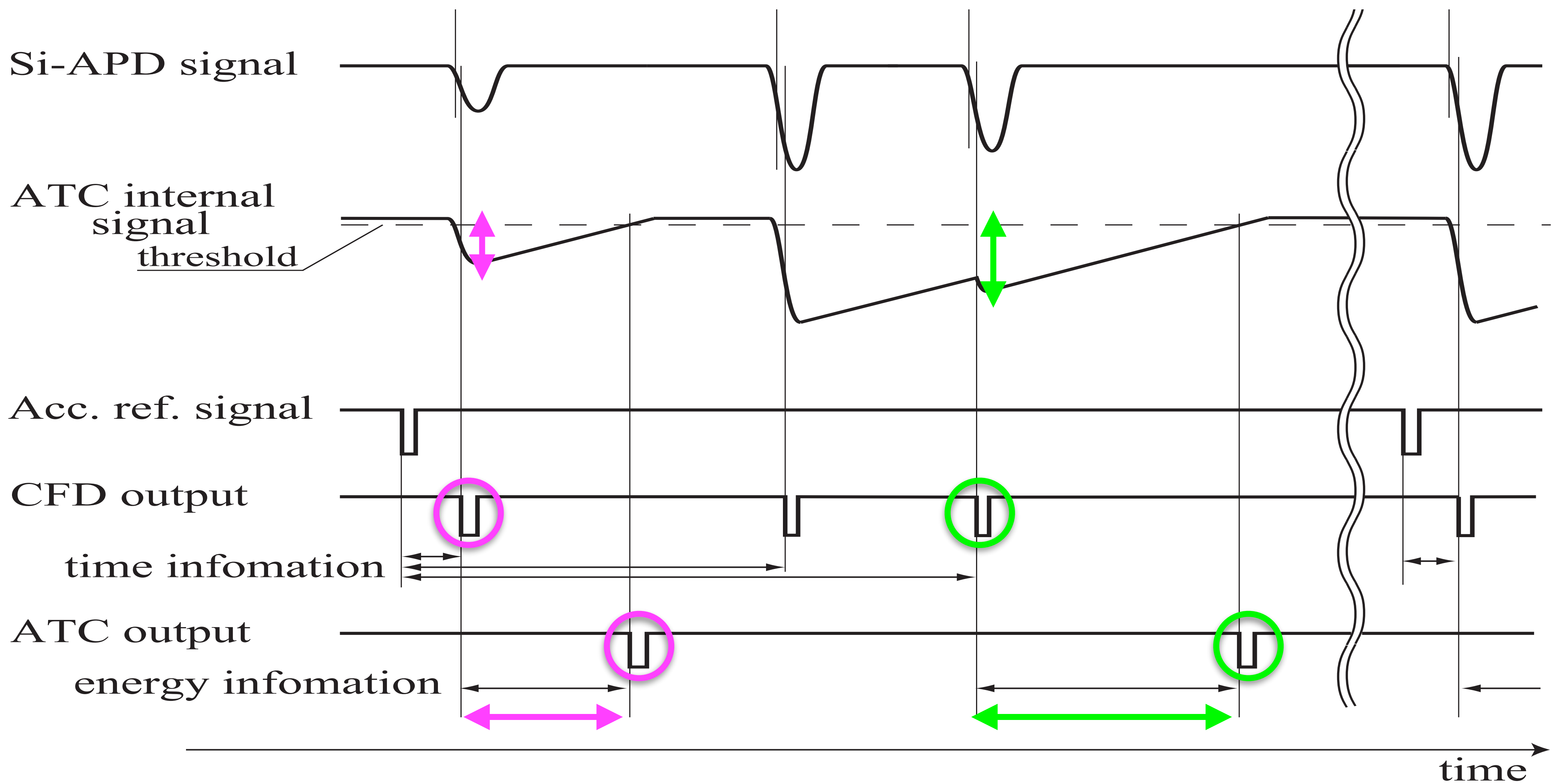
Osaka Univ /Tohoku Univ(Kasamatsu, Yasuda, Shigekawa, Konashi, Watanabe)

Fast X-ray detection system



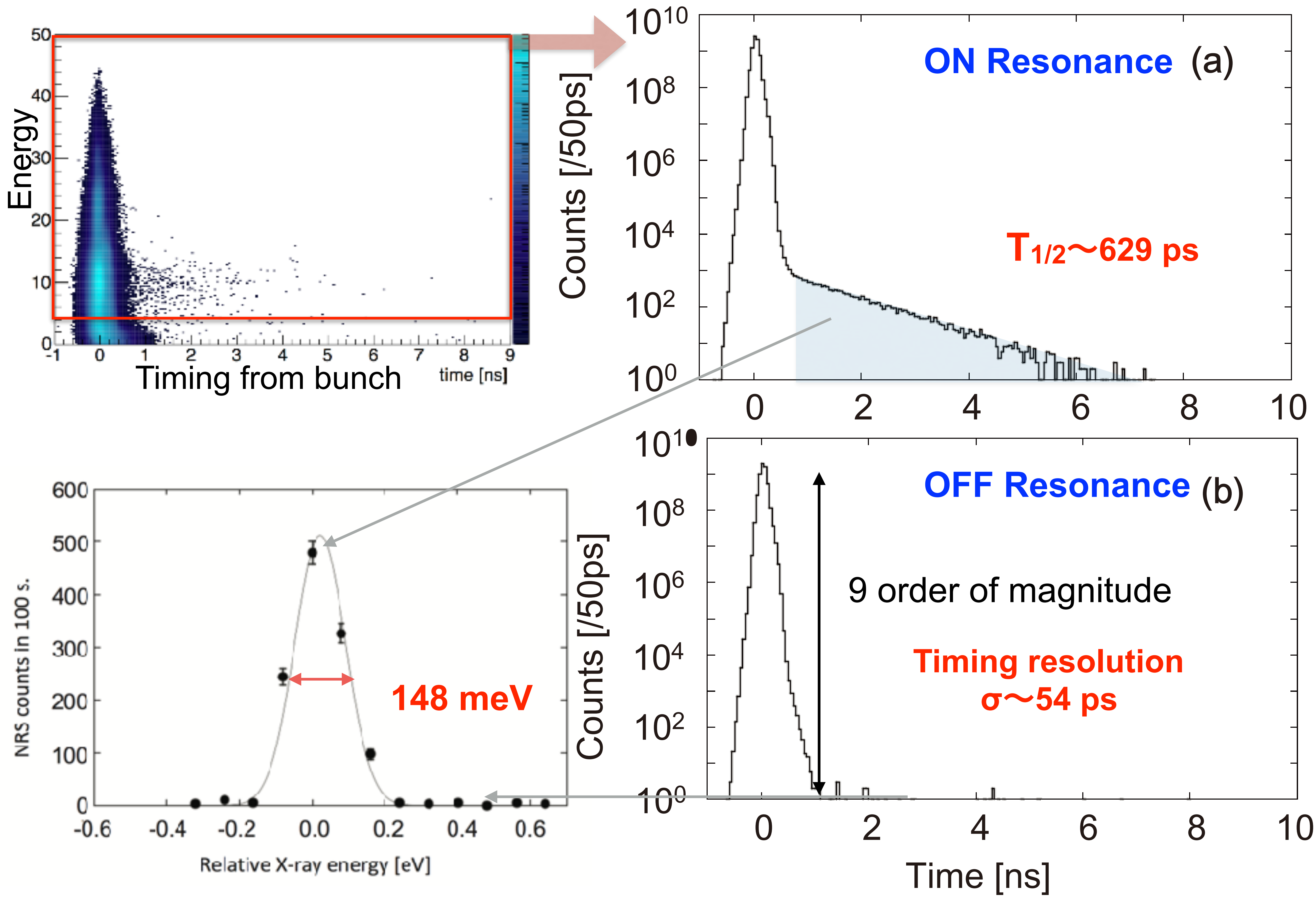
SiAPD Preamp output

Front end Electronics



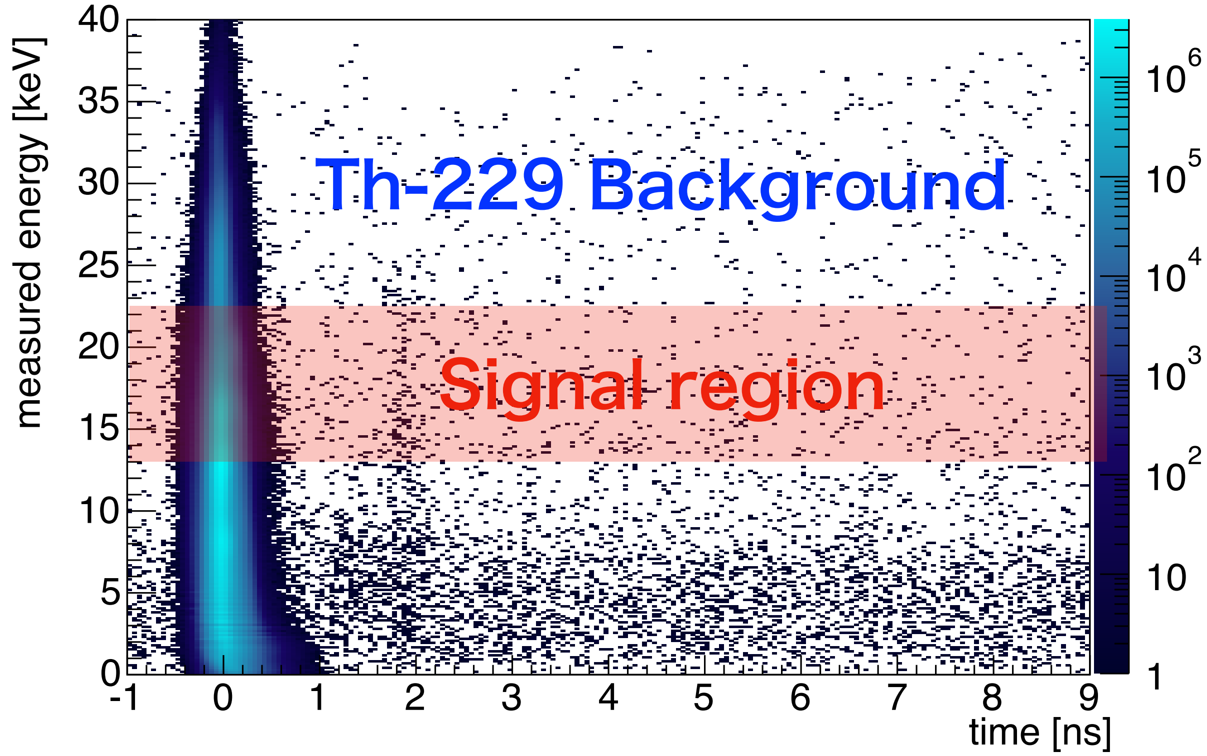
T. Masuda et al.,REVIEW OF SCIENTIFIC INSTRUMENTS 88, 063105 (2017)

NRS observation (Hg-201 26 keV)

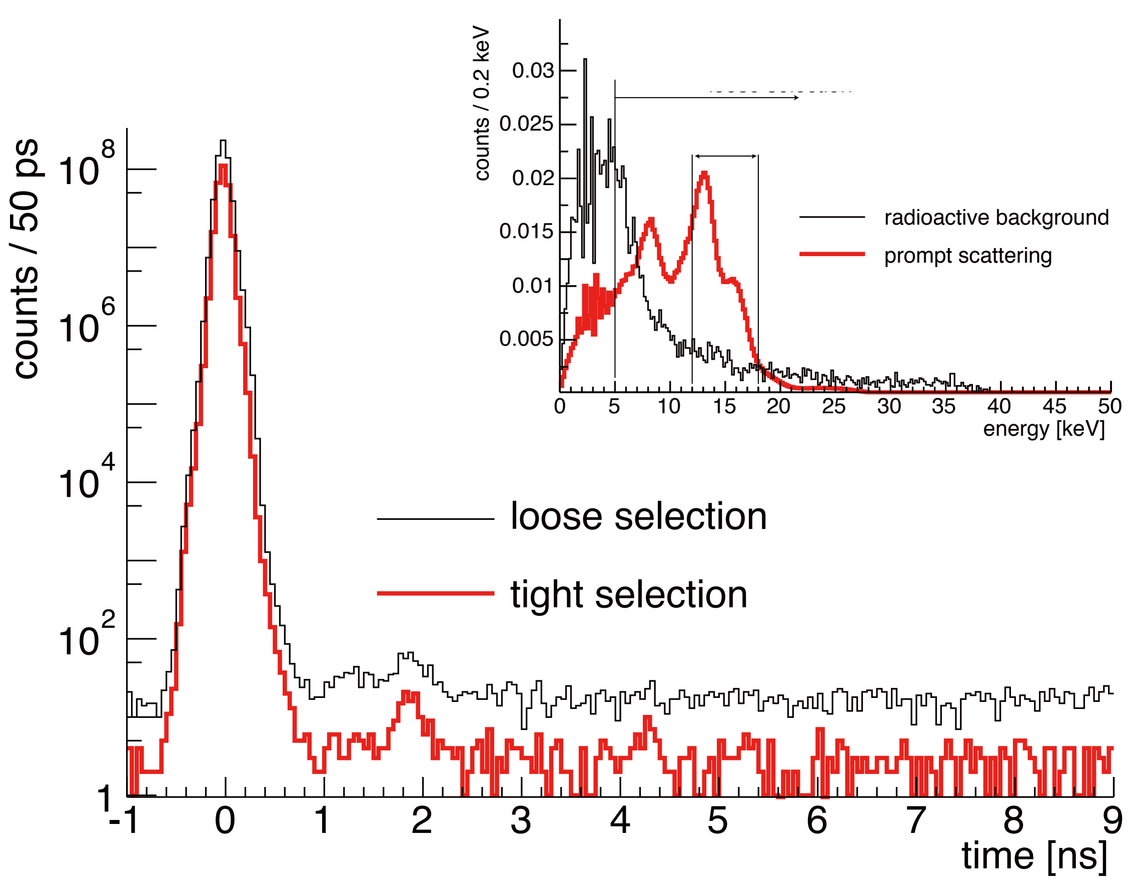


A. Yoshimi et al., submitted to PRC , arXiv:1705.07320v1

Preliminary search for Th NRS
2016.Dec



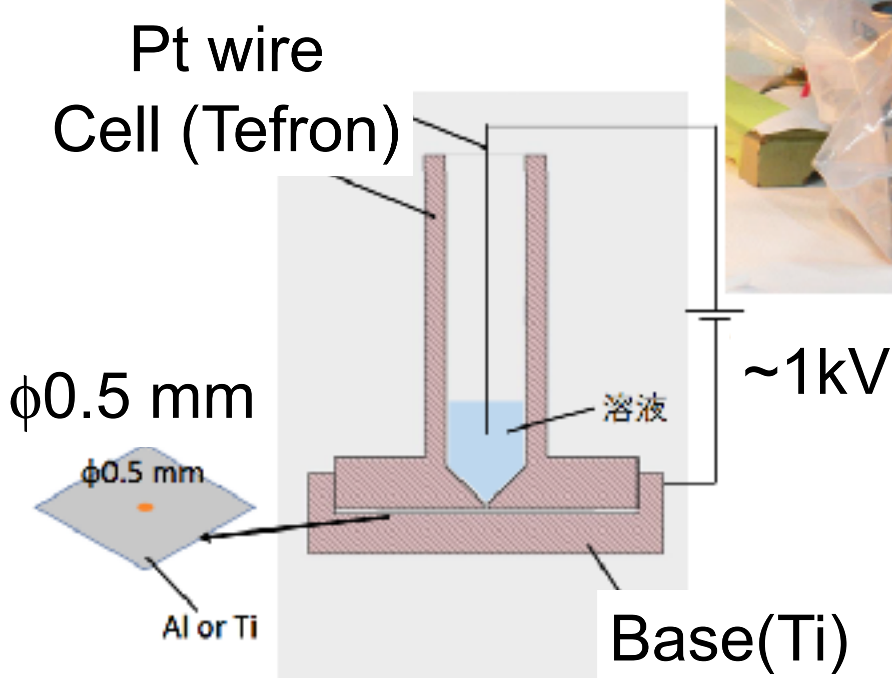
Energy selection to reduce radioactive background

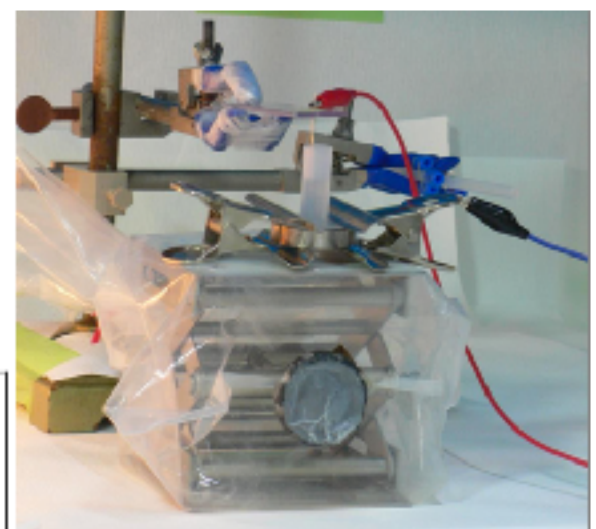


Recent Result

Higher density target

- $\phi 1.5 \rightarrow \phi 0.5$
  - x10 sensitivity
- Electrodeposition method (cf. Precipitation method)

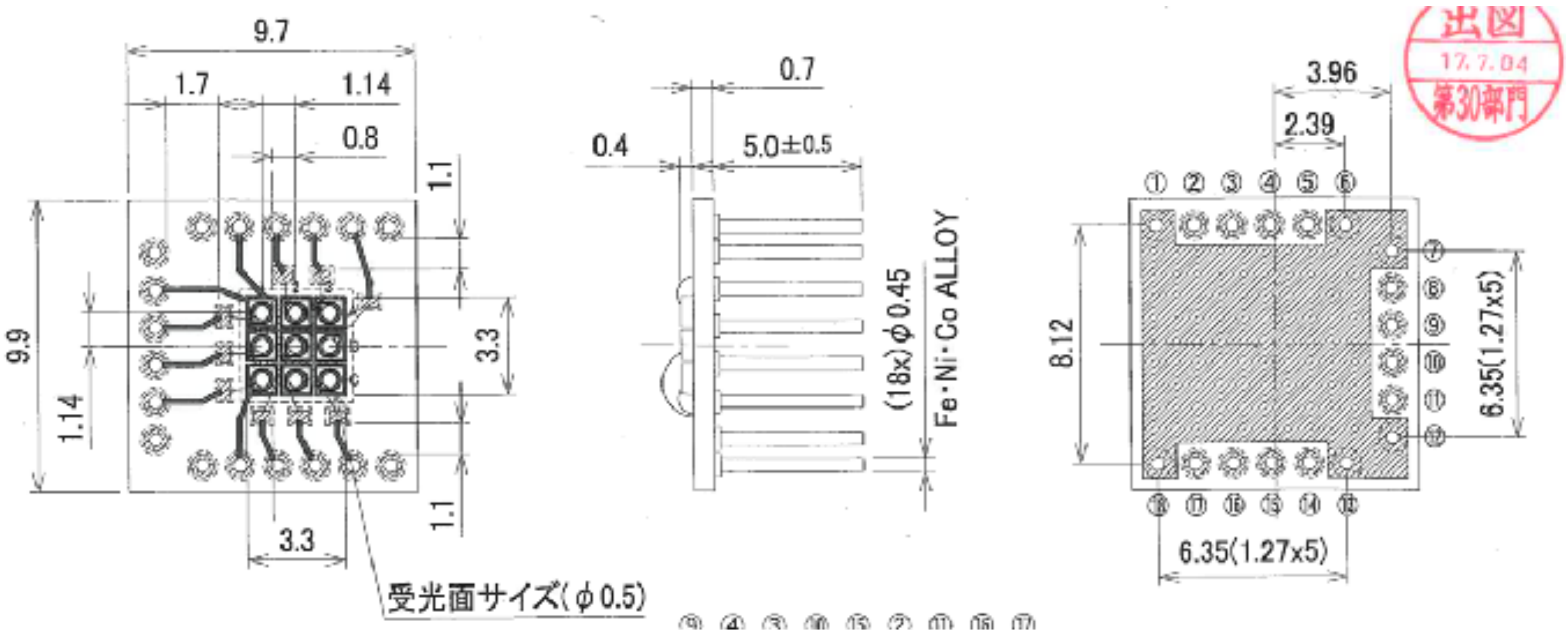


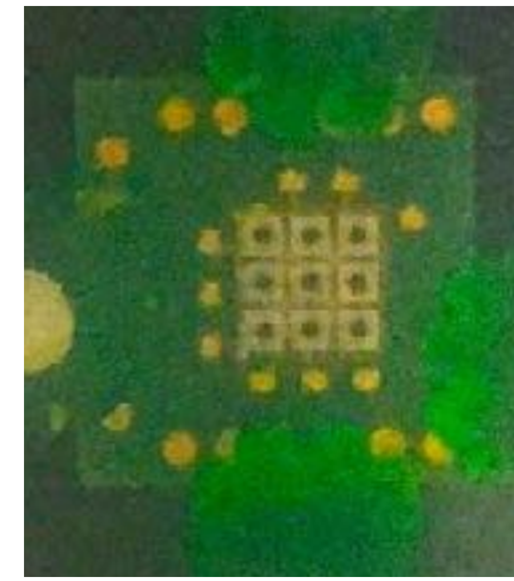


- Be 0.1 mm (Th-232 ~1ug)

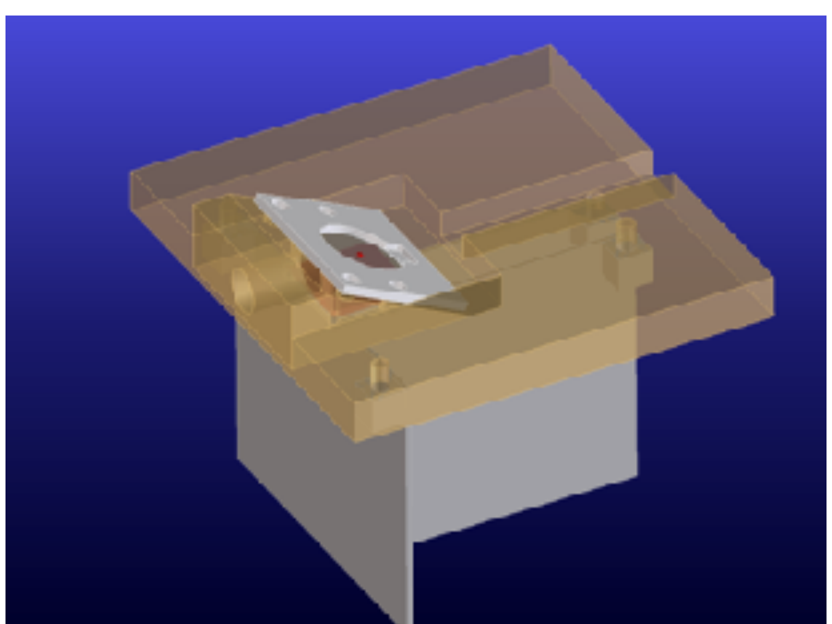
RIken (Haba, Yokokita, Yamaguchi), Osaka Univ (Kasamatsu, Yasuda, Shigekawa)

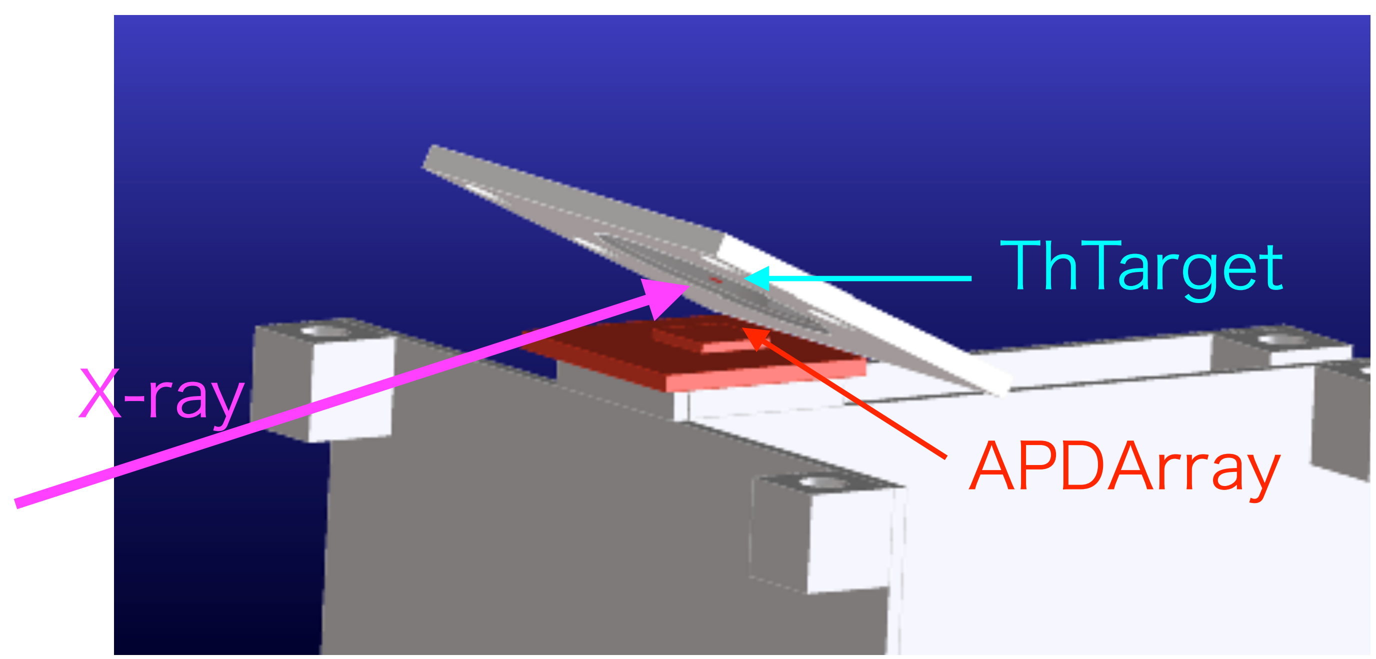
New 9ch APD array





9ch ϕ0.5 APD (10um)



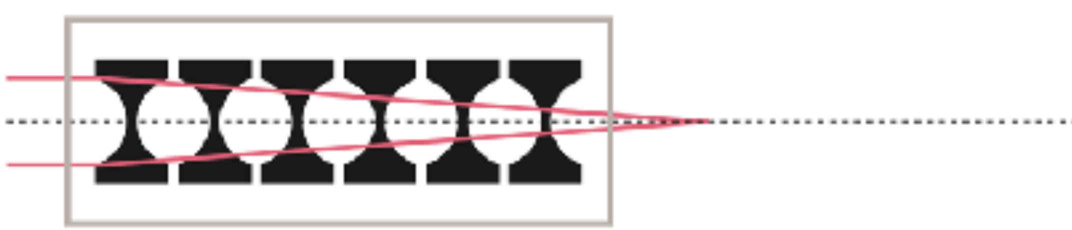


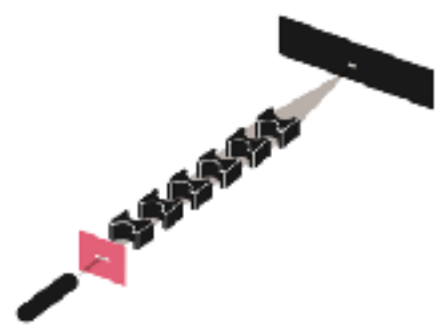
Distance between APD array and target
3.5 mm →2 mm

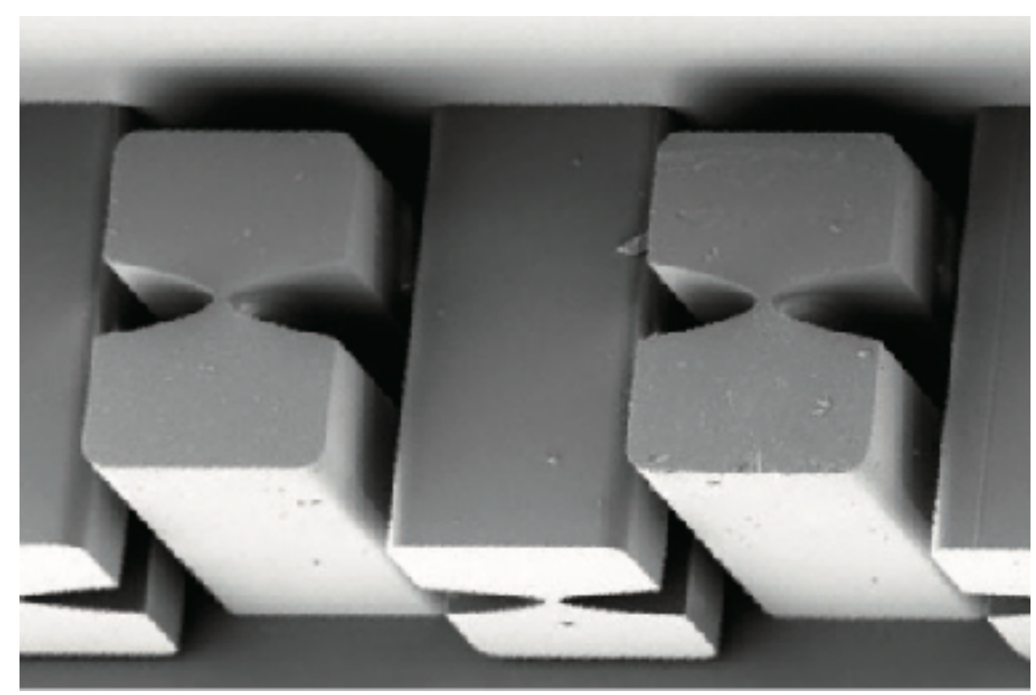
Acceptance x10

New X-ray focusing system
- compound refractive lens -

- Installed in BL09XU in Oct. 2017
- 2D focusing for 29.2 keV X-ray
  - Aperture 1.5 mm x 1.5 mm
  - Transmission efficiency 53%
  - Beam size at focal point ～50μm（V）×200μm（H）
  - Focal length 14 m



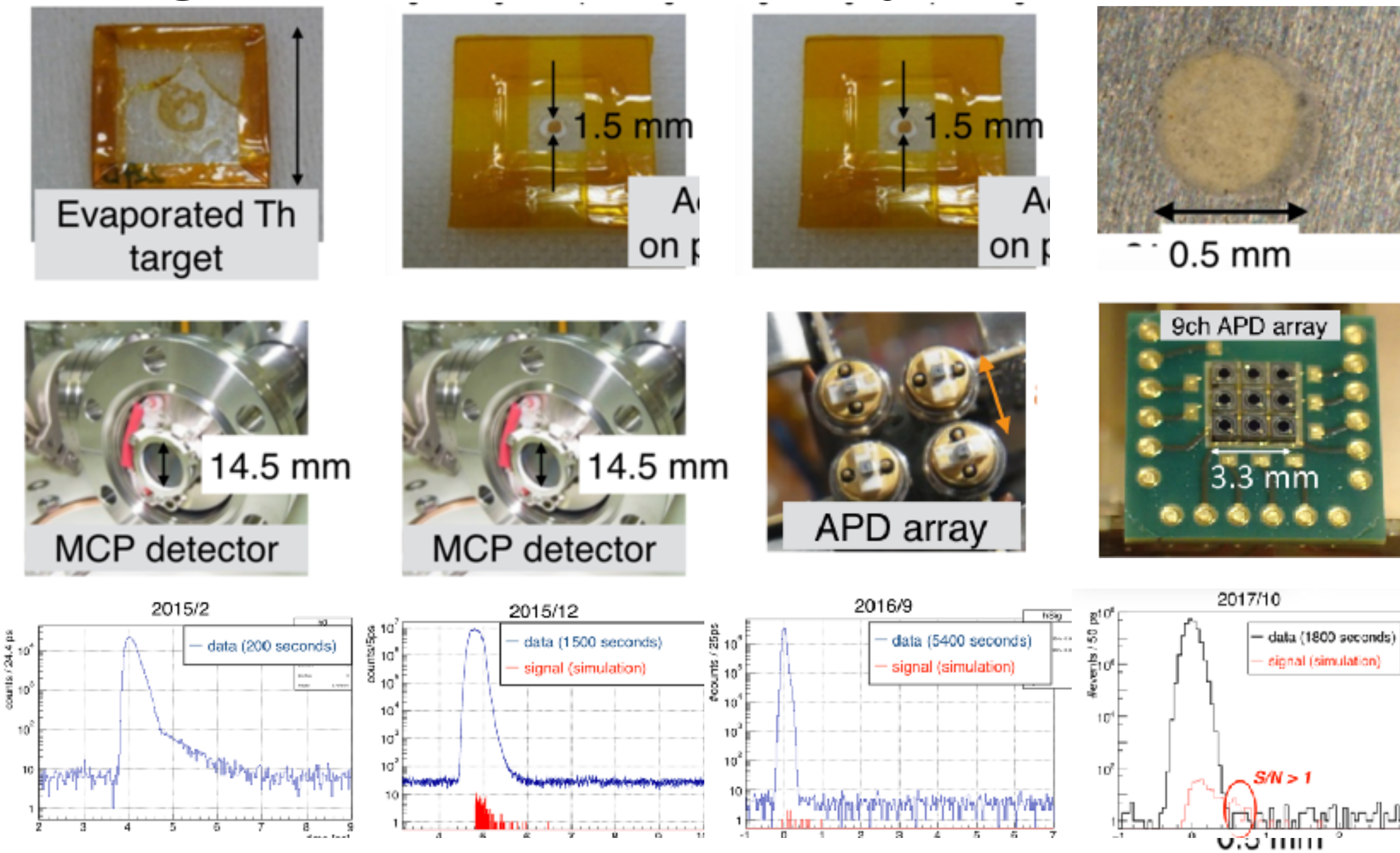




Karlsruhe Institute of Technology (KIT)
Institute of Microstructure Technology (IMT)

microworks GmbH

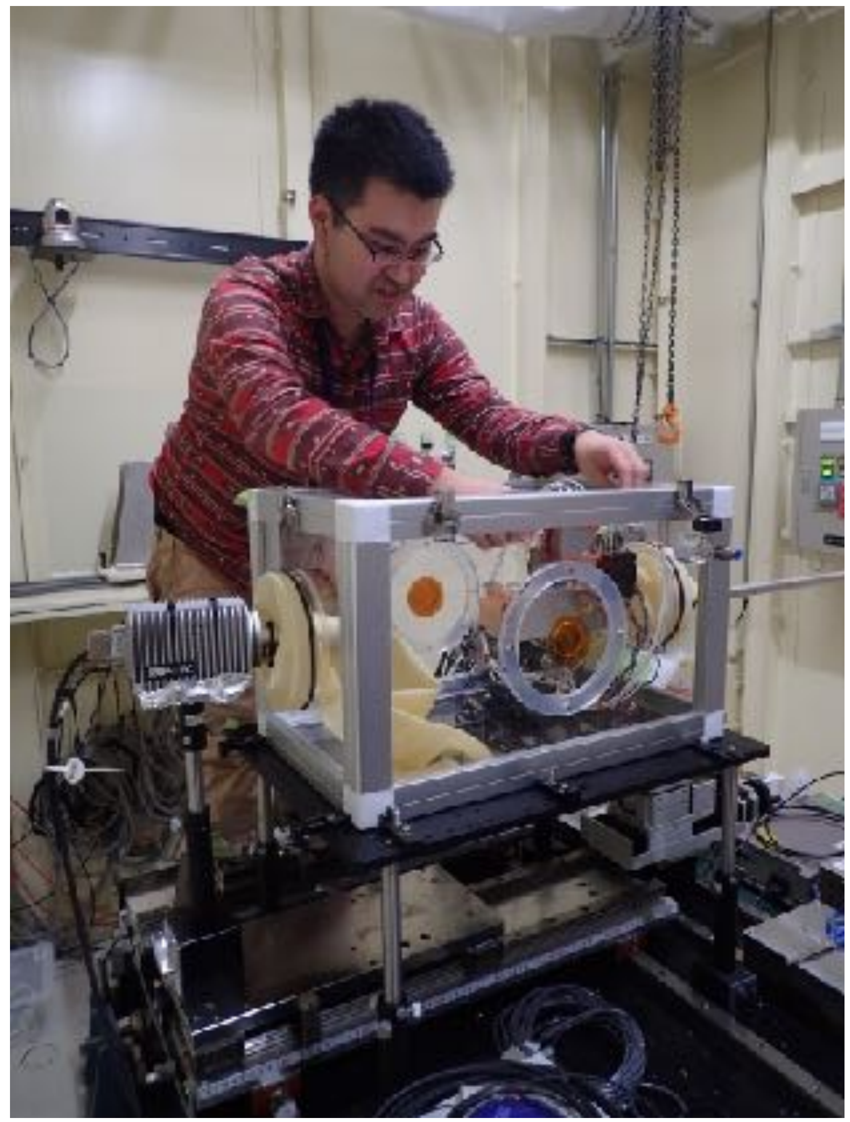
Progress over three years

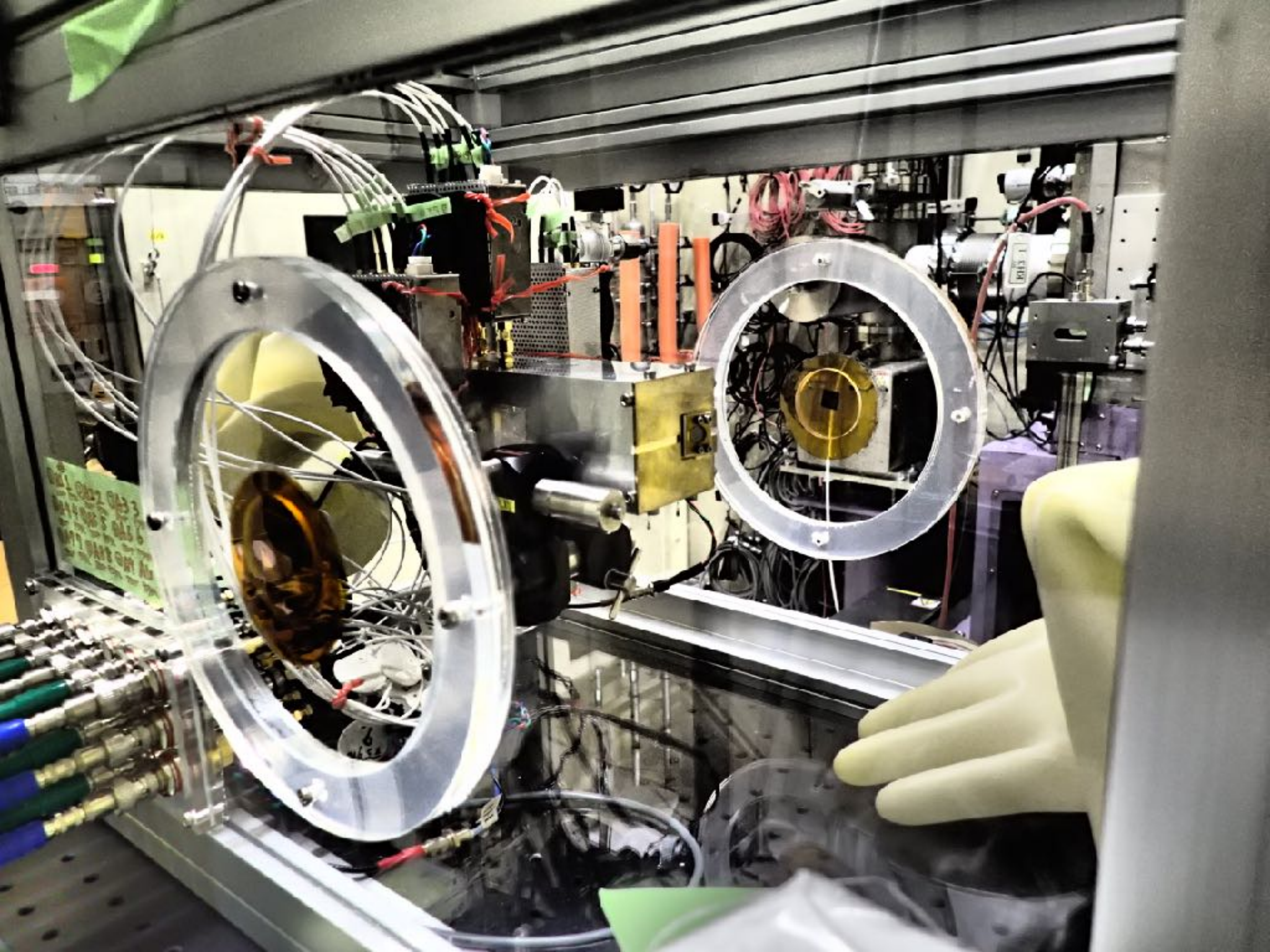


	2015/2	2015/12	2016/9	2017/10 (New)
Detector	14.5mmϕ MCP	14.5mmϕ MCP	0.5mmϕ **APD 4ch**	0.5mmϕ APD 9ch
Th target	0.06 μg/mm2 × 5mmϕ	**1.7 μg/mm2** × 1.5mmϕ	1.7 μg/mm2 × 1.5mmϕ	**3.0 μg/mm2** × 0.5mmϕ
Beam size (FWHM)	~1×1 mm2	1.0×0.8 mm2	**0.2×0.1 mm2**	**0.2×0.05 mm2**
count rate	1.3 kHz	300-400 kHz	22 kHz	310 kHz

Last Spring-8 beam time

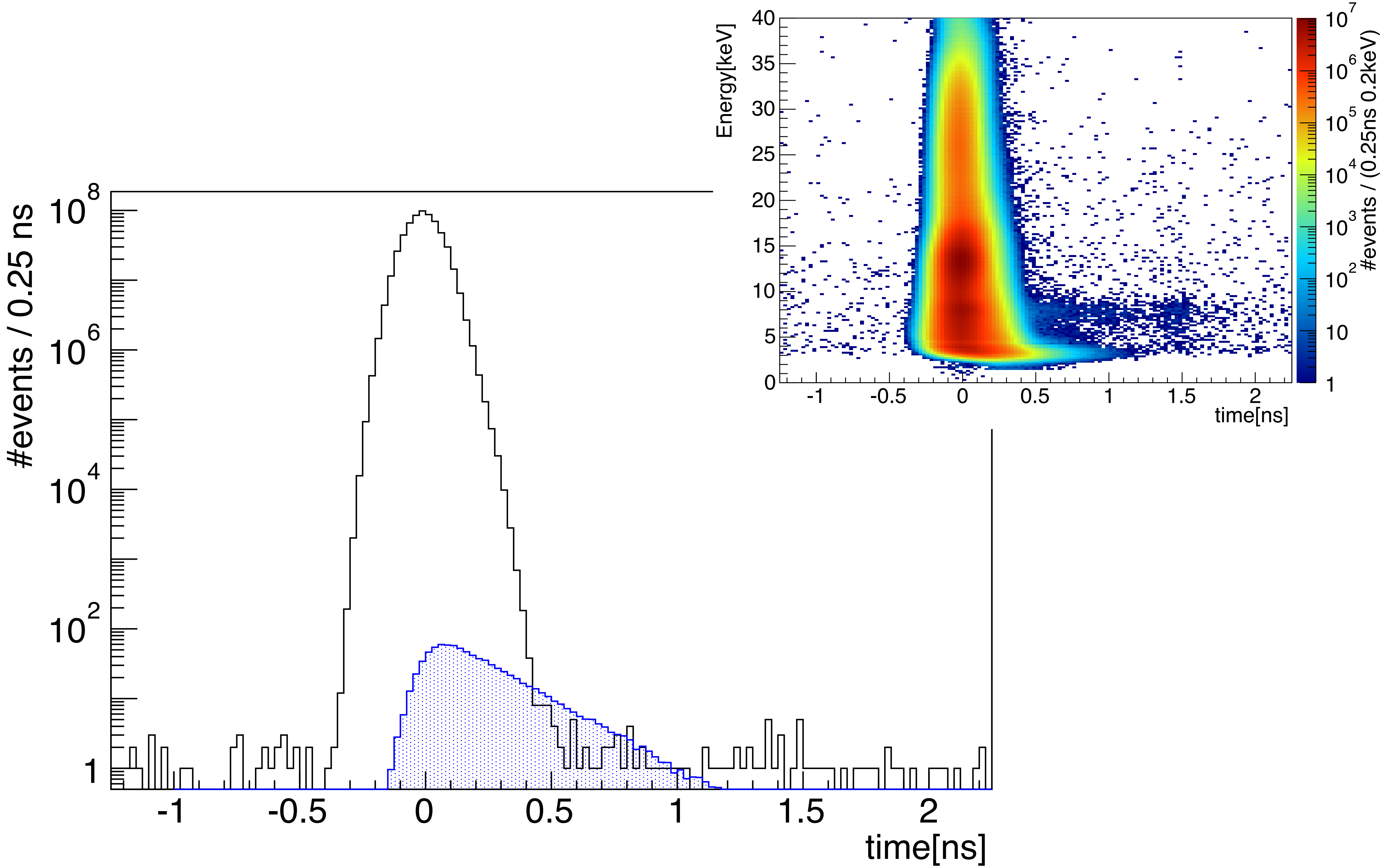
2018 April





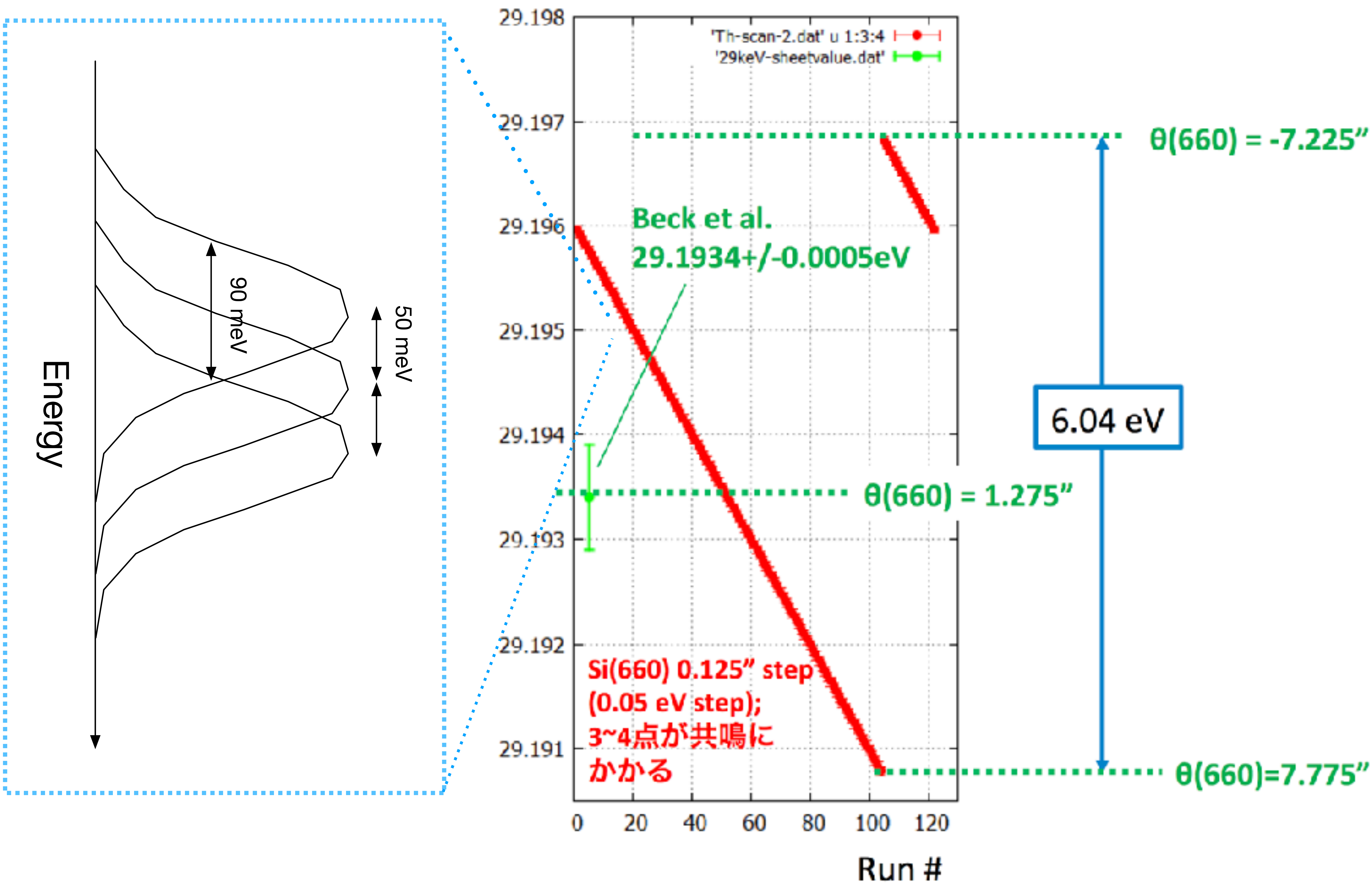
Recent result

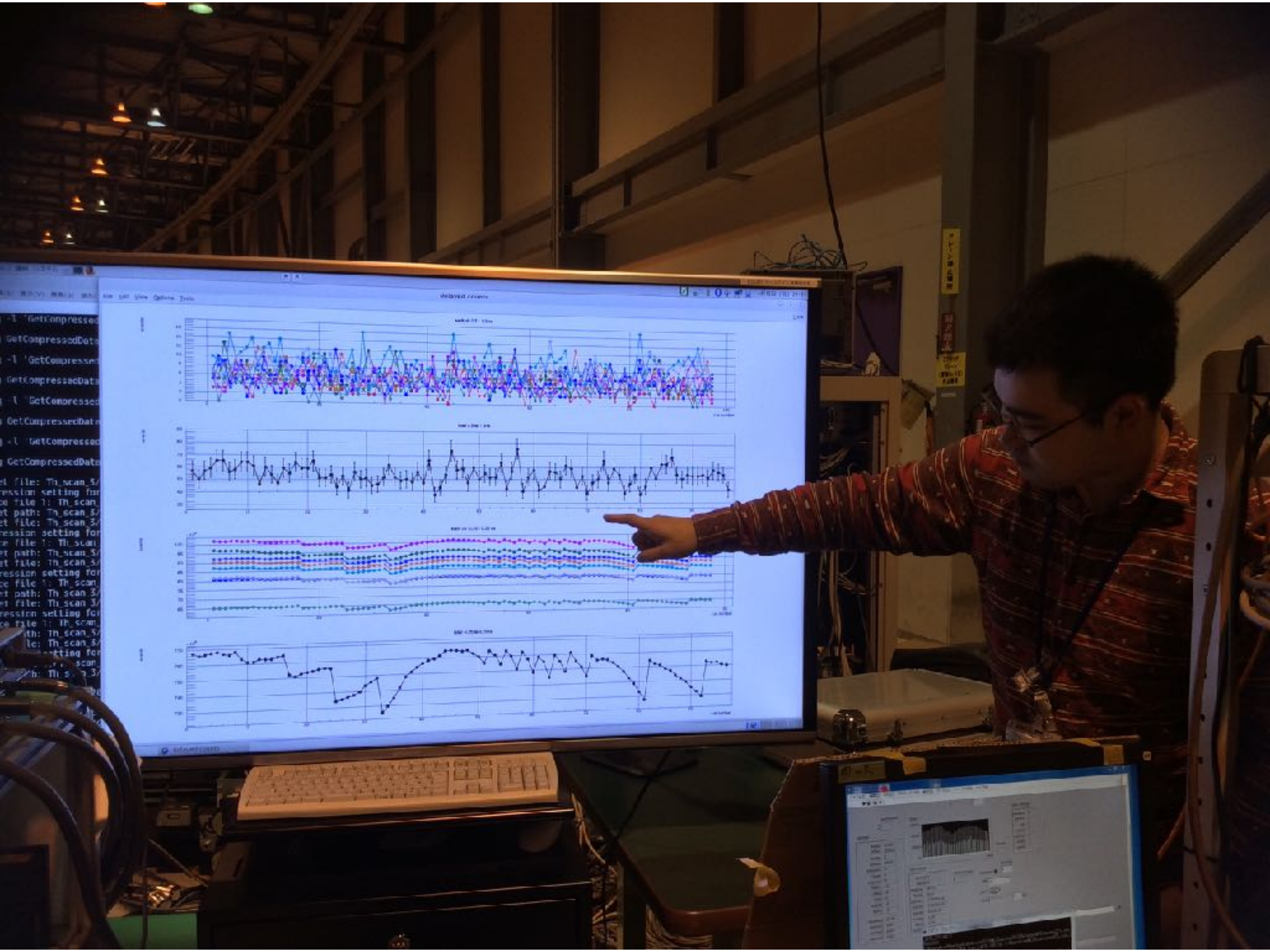
2018 April



Recent result

2018 April



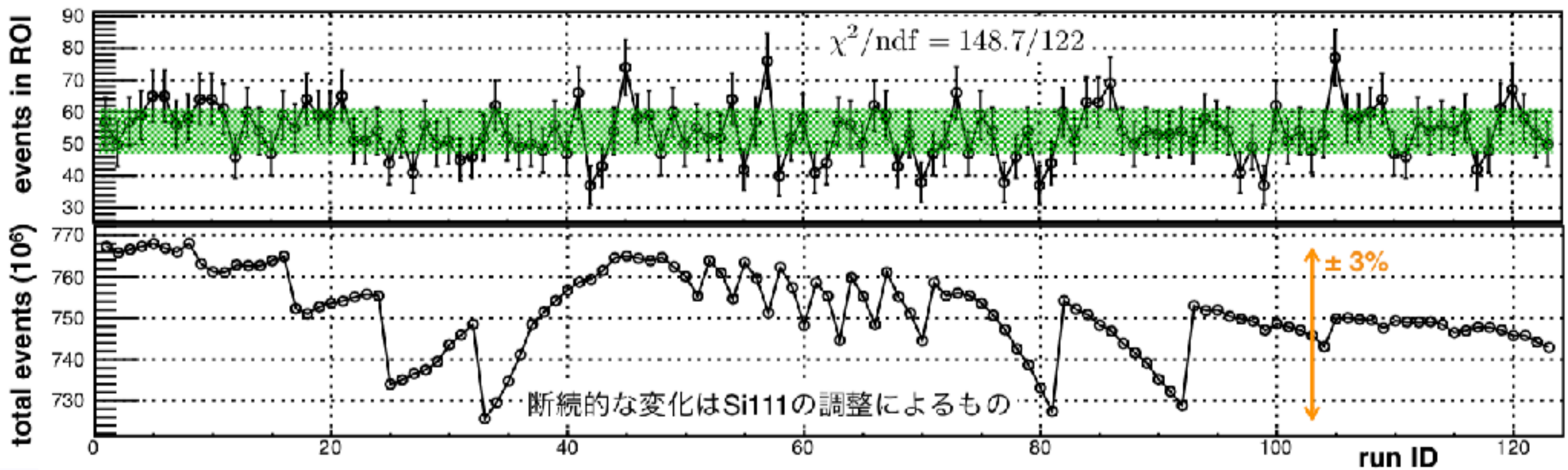


No signal at all

2018 April

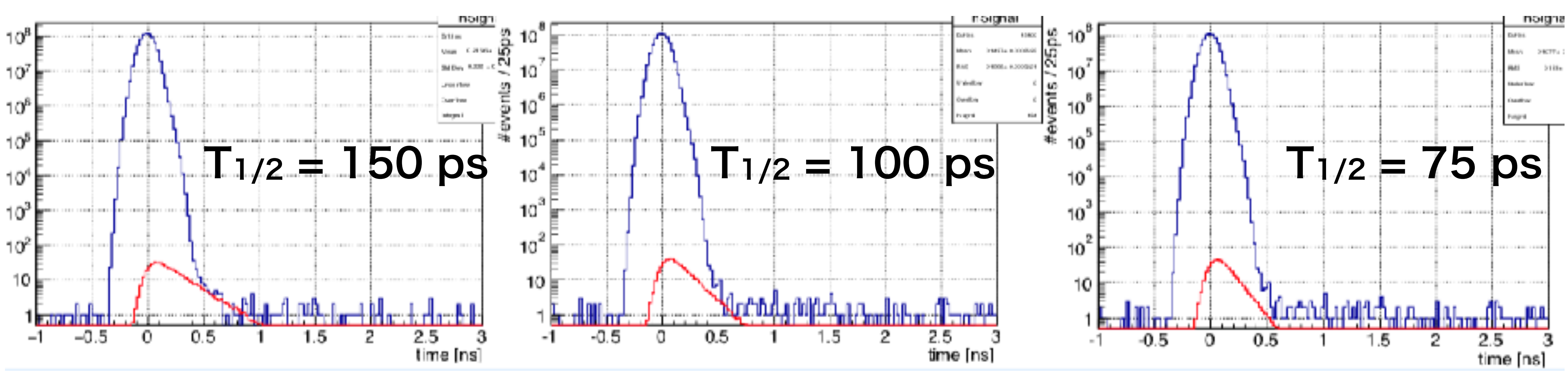
ROI 0.5 ns ~ 1.0 ns

2.5 days scan



Why not observed?

- Search energy range
    - NRS resonance would be outside the range?
- Energy precision
    - Energy of Monochrometer may be drifted during scan.
- Lifetime
    - Life time might be too short to detect.

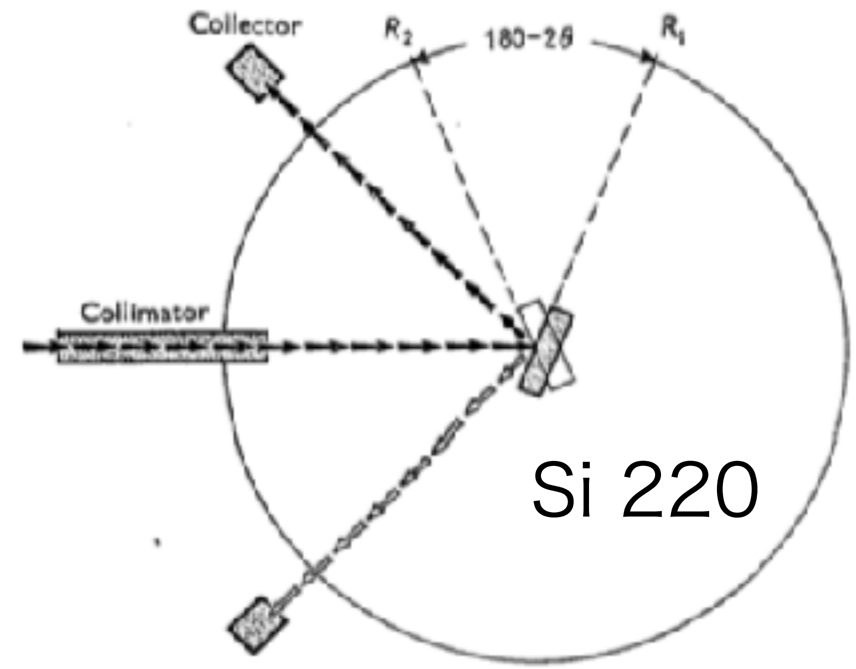


For next challenge

- In stu Energy calibration
  - Bond method

    W.L.Bond Acta Cryst. (1960) 13, 814
  - Angular measurement
    - $\Delta\theta$~0.1” if ΔE〜10 meV
    - Self calibrated rotary encoder (SelfA AIST)

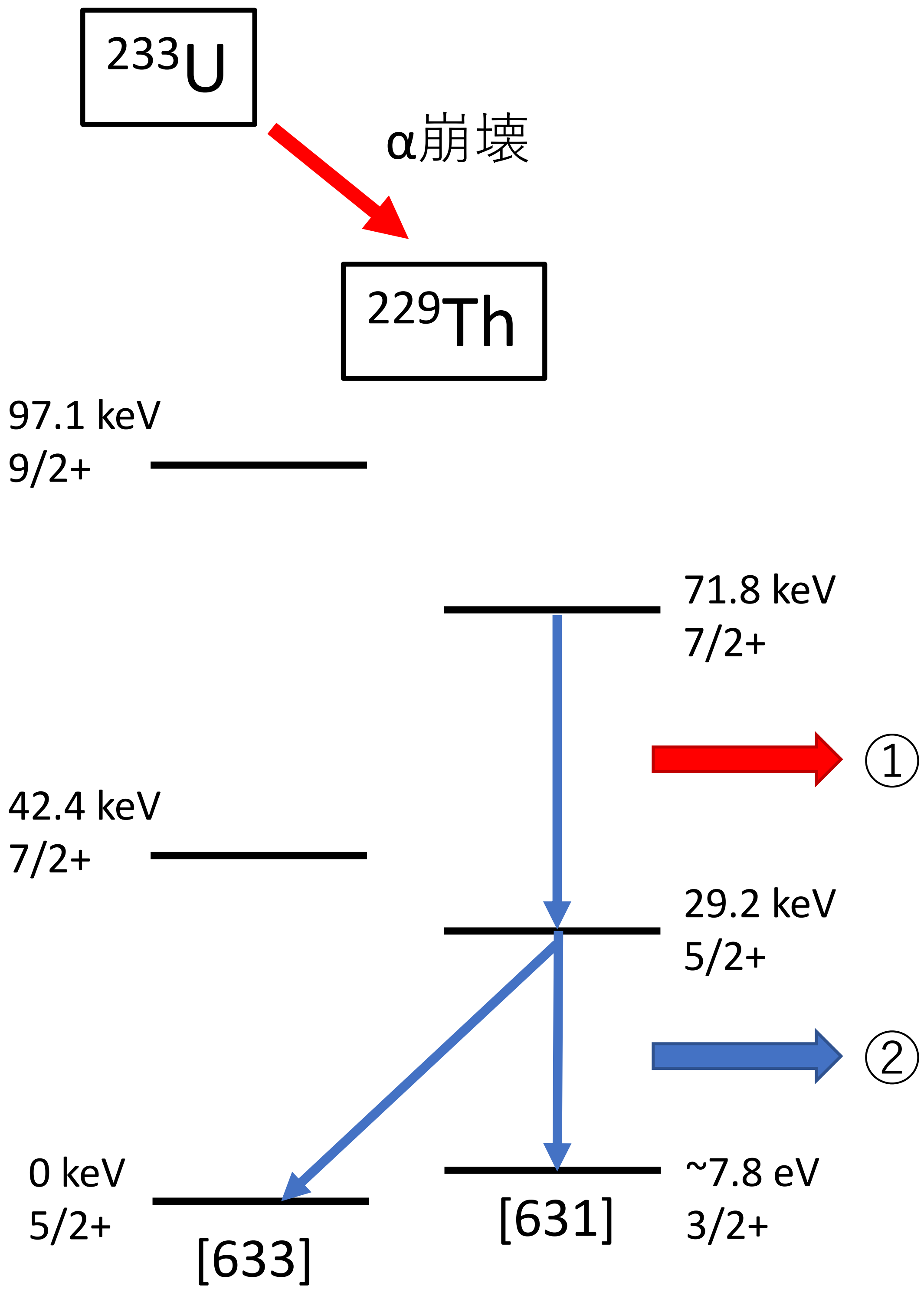
  T. Watanabe Journal of Japan Society for Precision Engineering Vol 82, No9, PP 792-796 2016



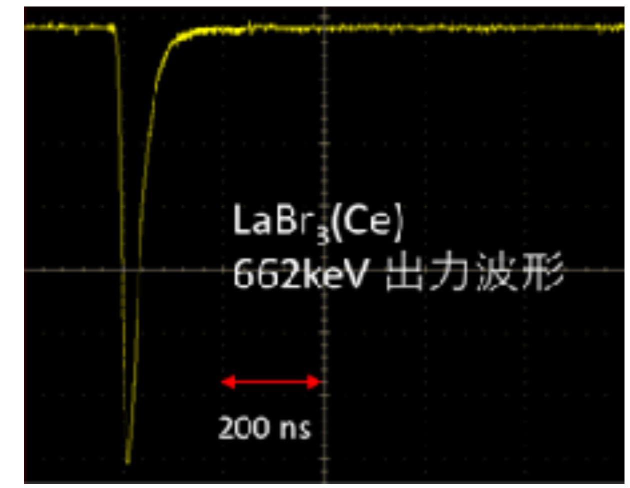
- Lifetime measurement
  - Lifetime of 2nd Excitation level
  - Critical parameters

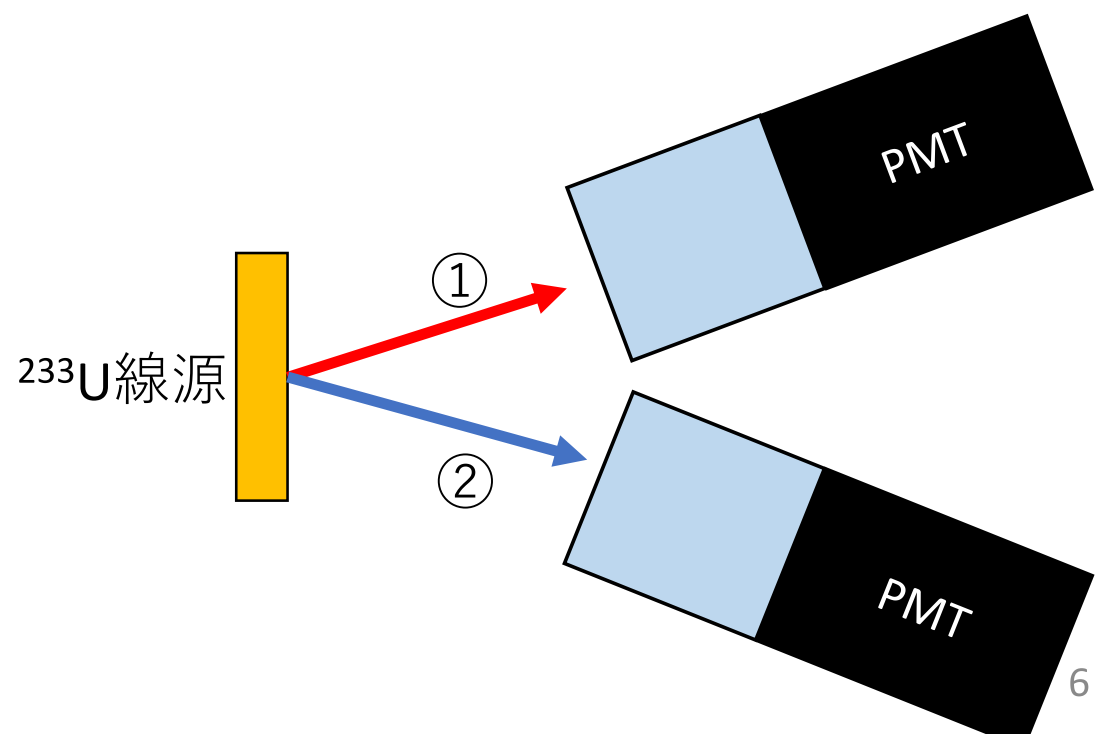
Lifetime measurement

2017 summer, Oarai center, Tohoku Univ



1x1 inch $LaBr_3(Ce)$

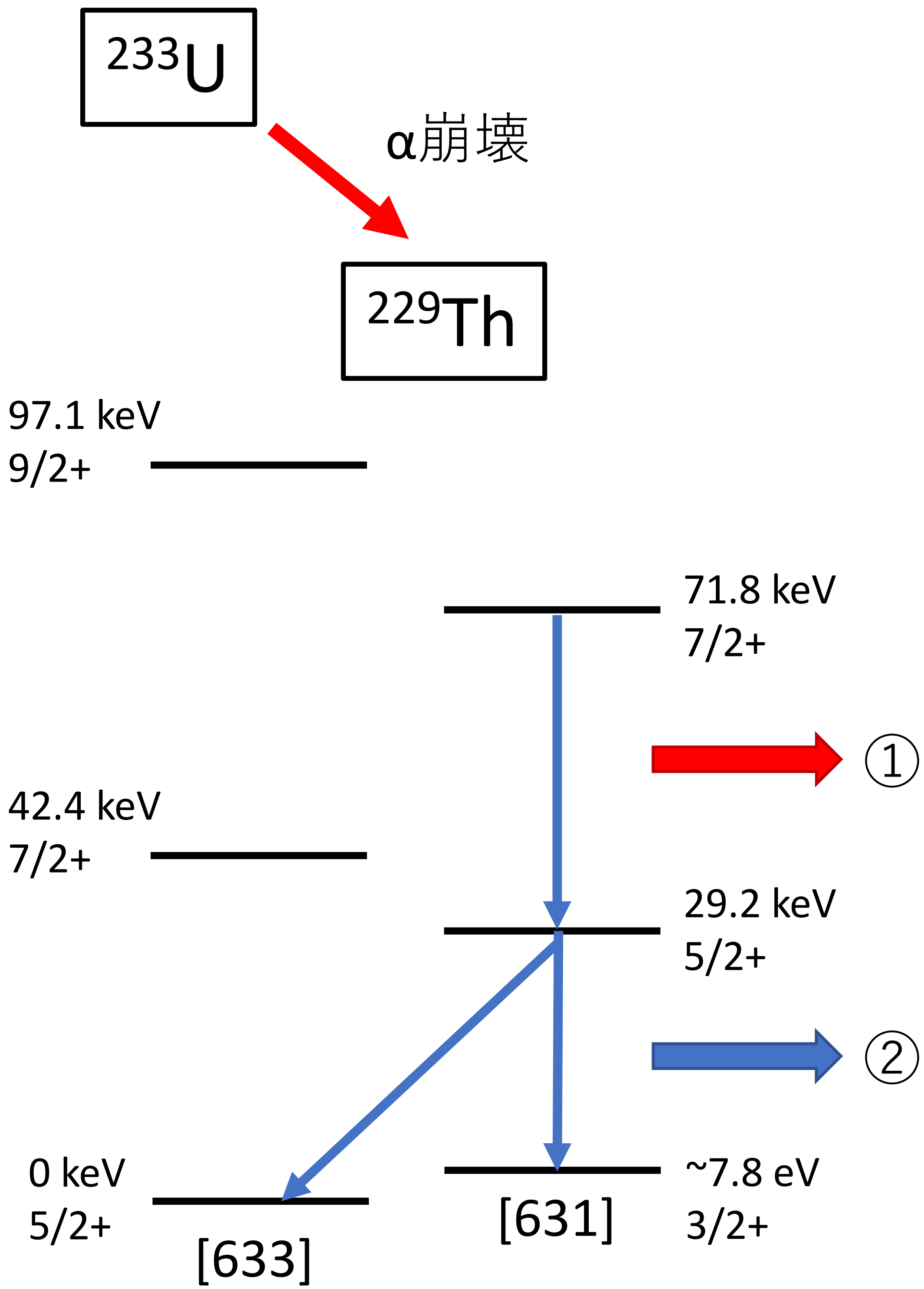


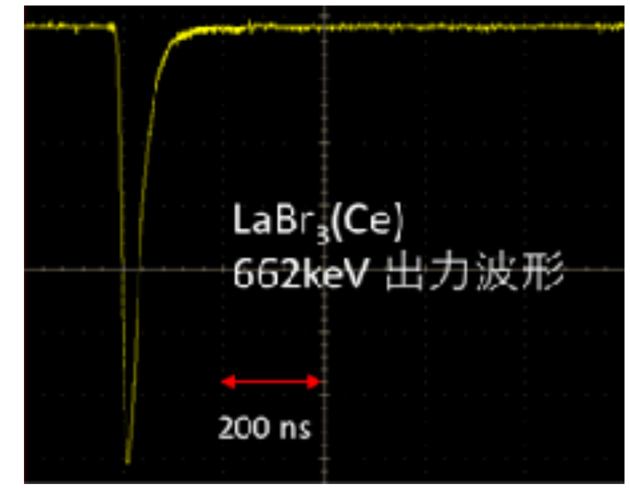


R13449-100-10(PhotoCathod:SBA)

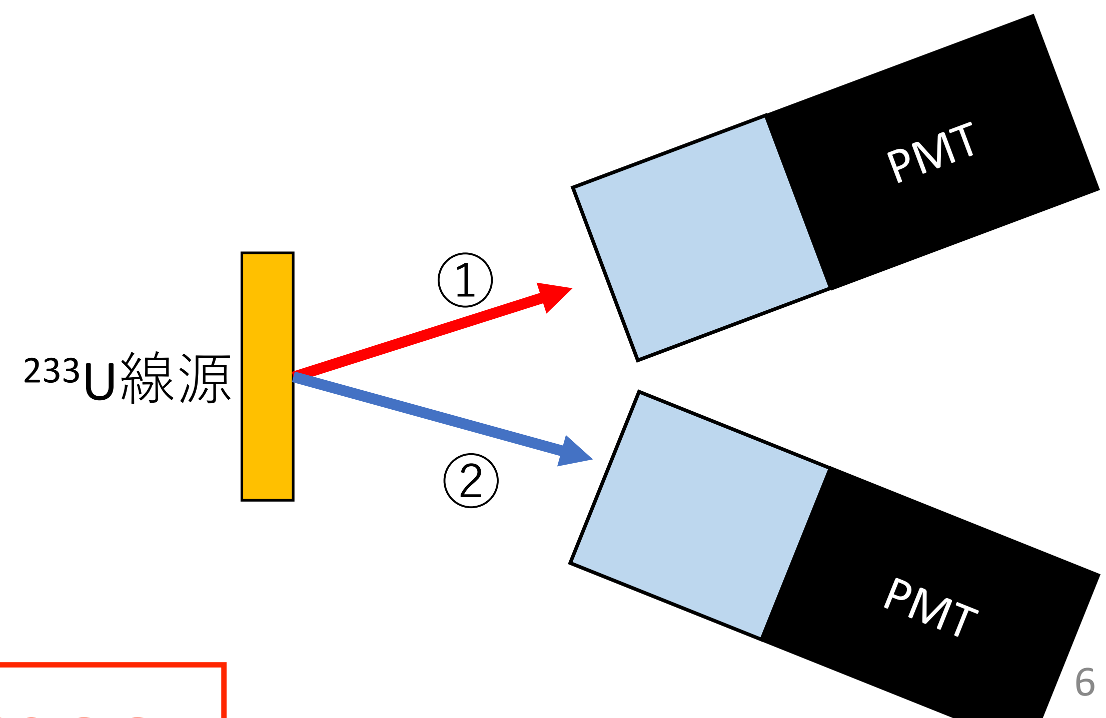
Lifetime measurement

2017 summer, Oarai center, Tohoku Univ





1x1 inch $LaBr_3(Ce)$



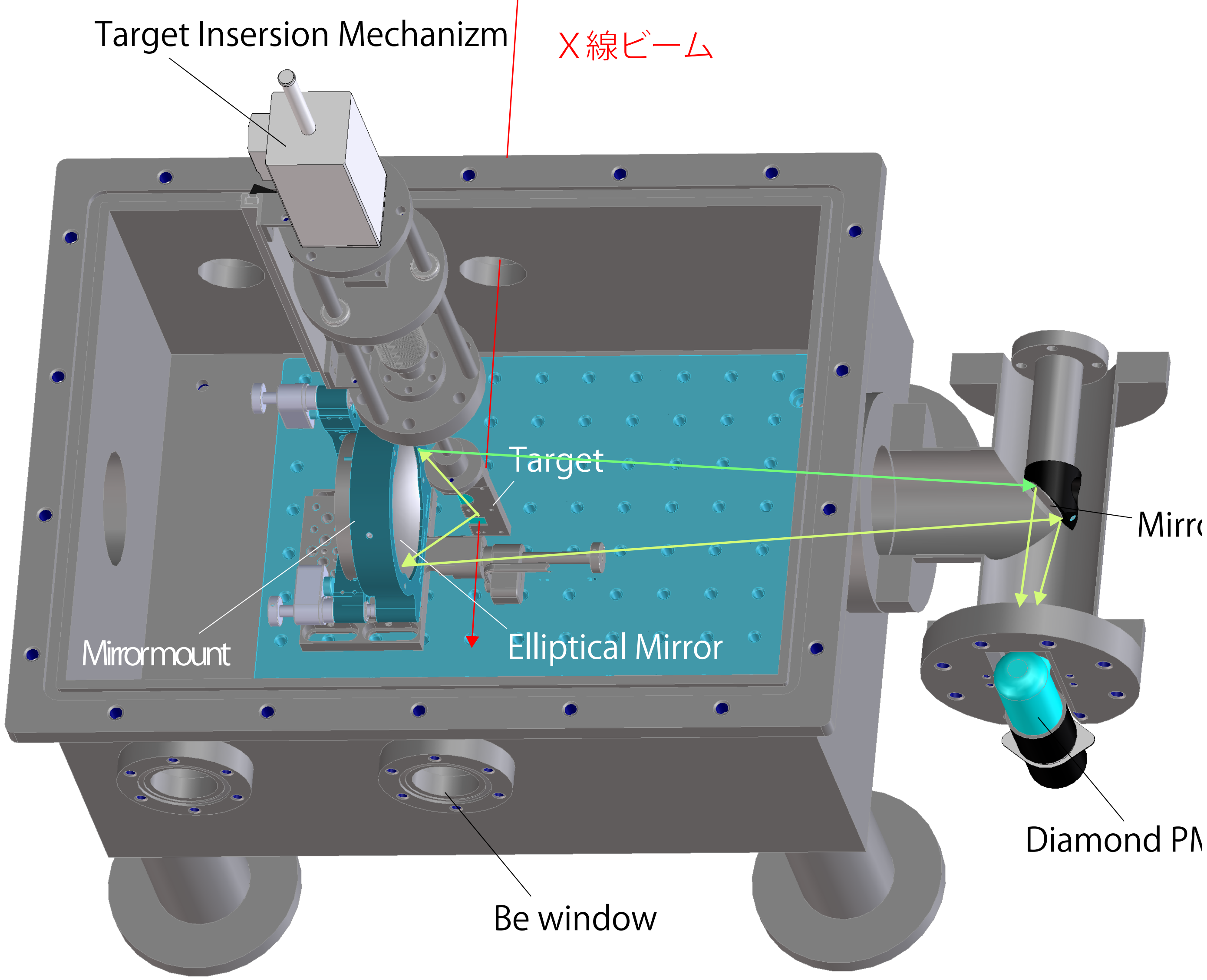
R13449-100-10(PhotoCathod:SBA)

Data analysis is still in progress.

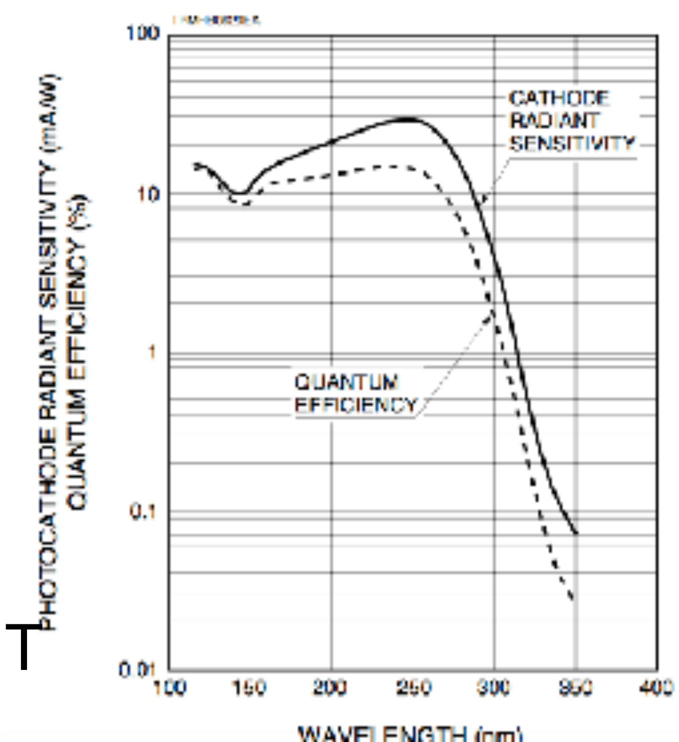
Future prospect

To observer VUV signal

- Ellipsoidal mirror system is developed
- Th:CaF2 crystal povided by TU Wien
- VUV emitted from the crystal is viewed by solar-blind PMT

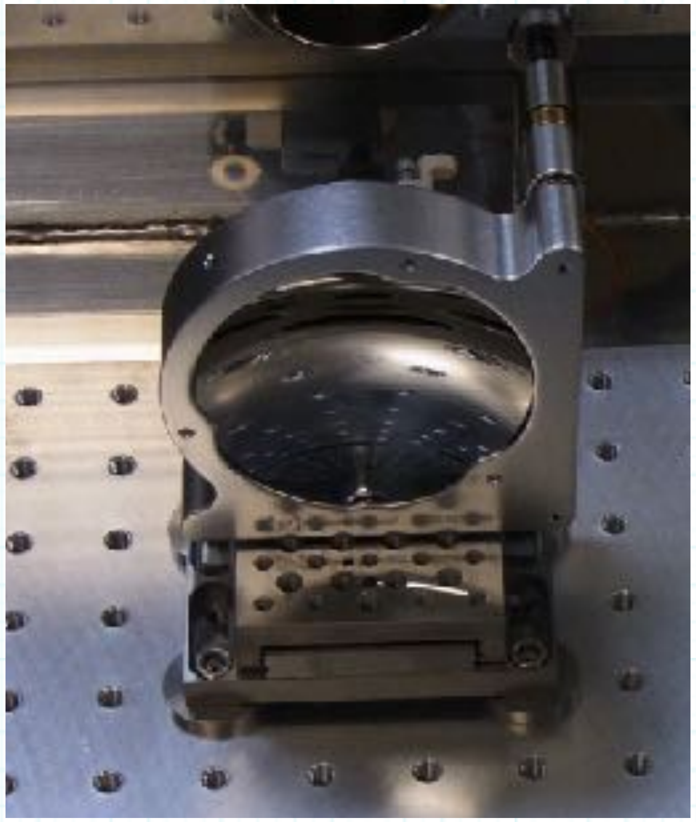


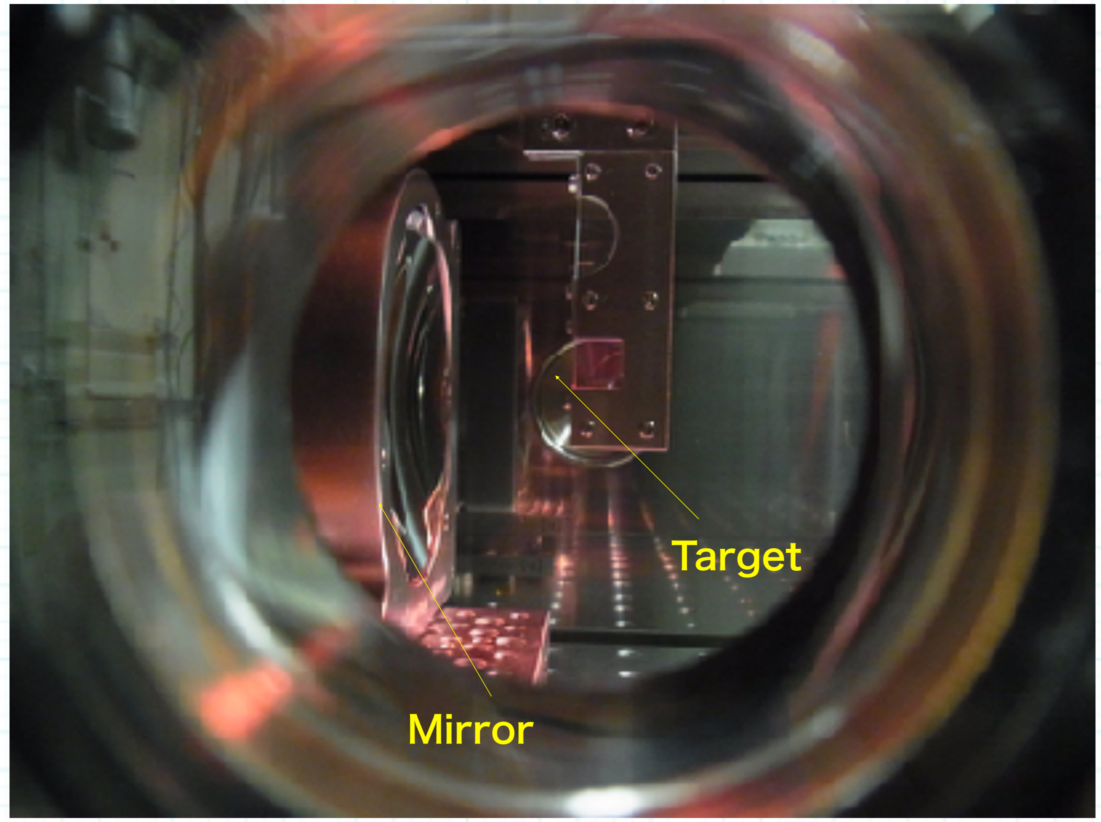
Th-doped CaF2 provided by Wien Tech.

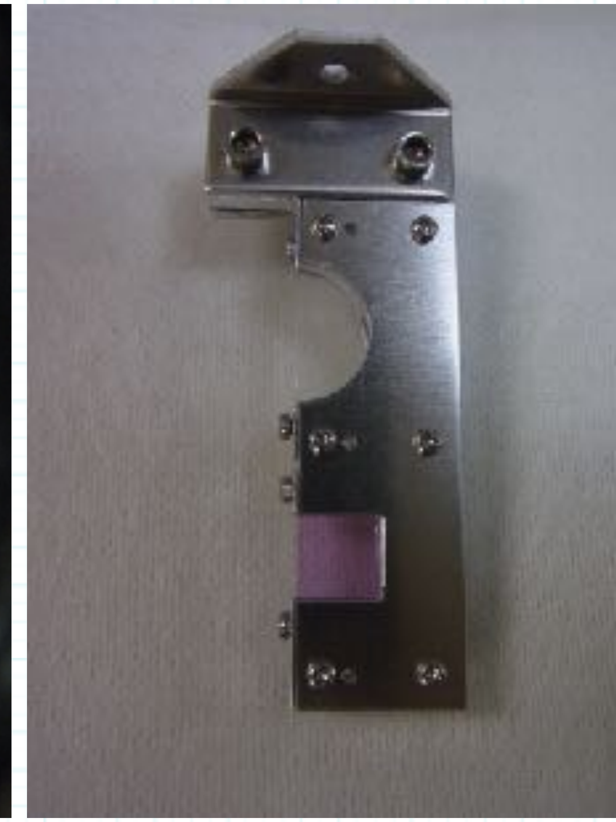


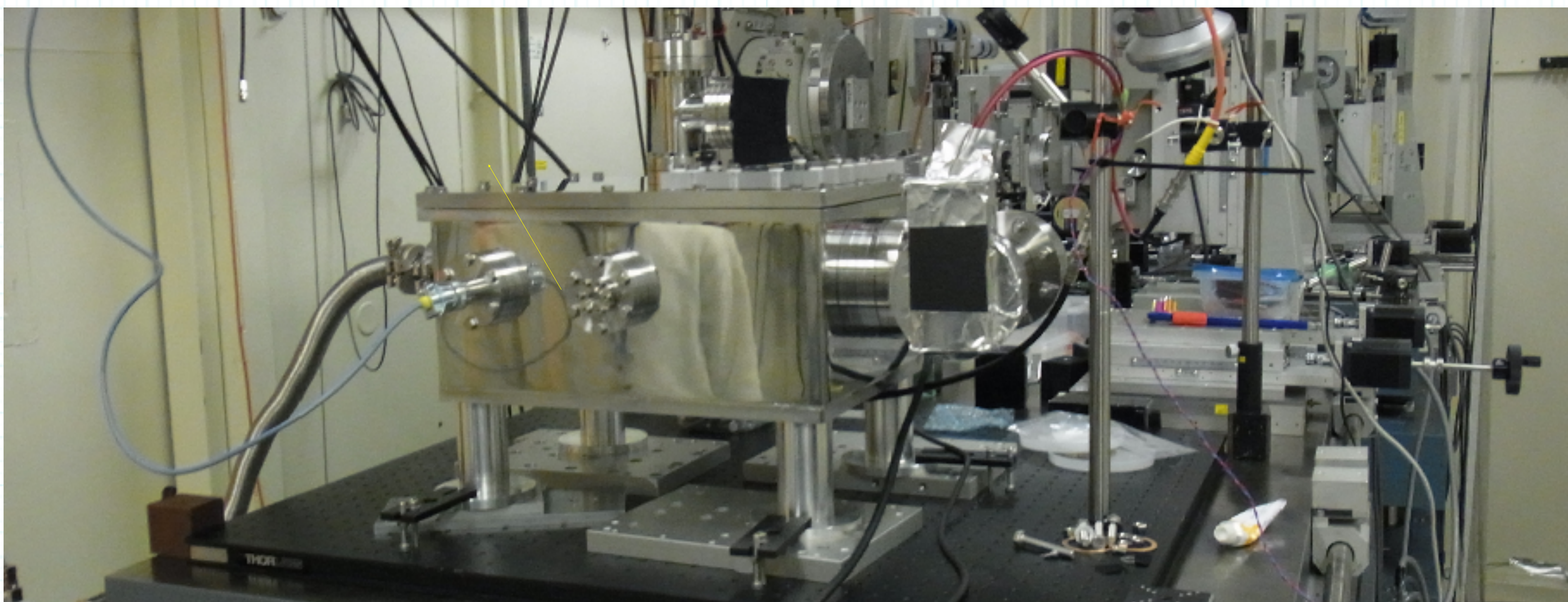
Solar Blind PMT

Optical system check (2015)



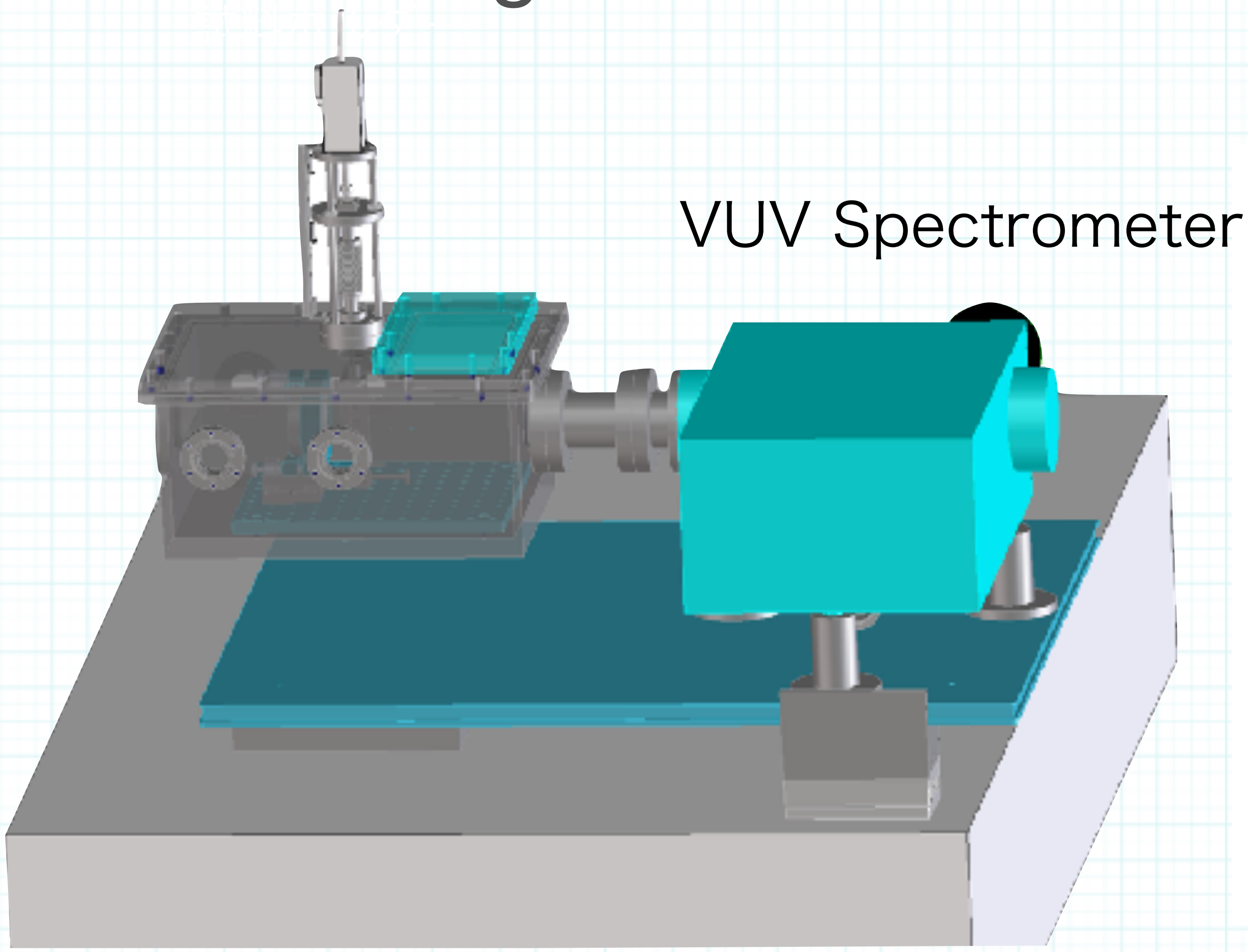


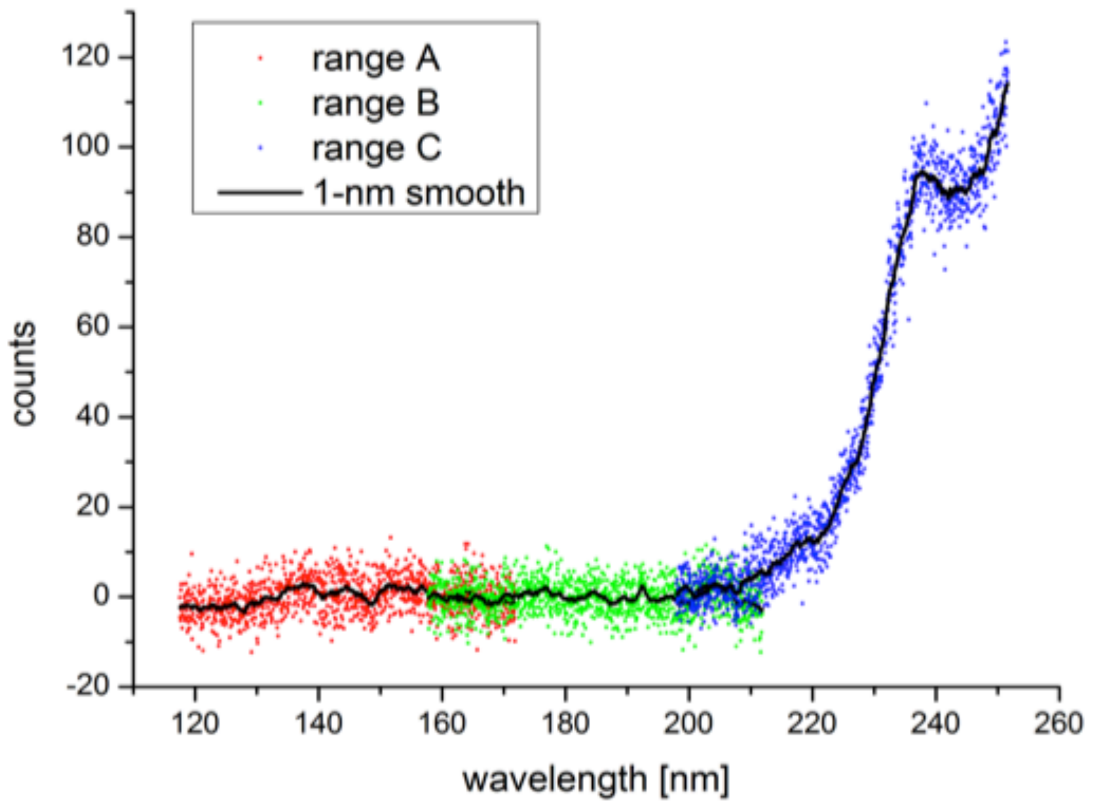




Measurement of VUV wavelength

- VUV spectrometer with cooled CCD camera.
- Wavelength could be determined by 0.2 nm (0.01 eV)



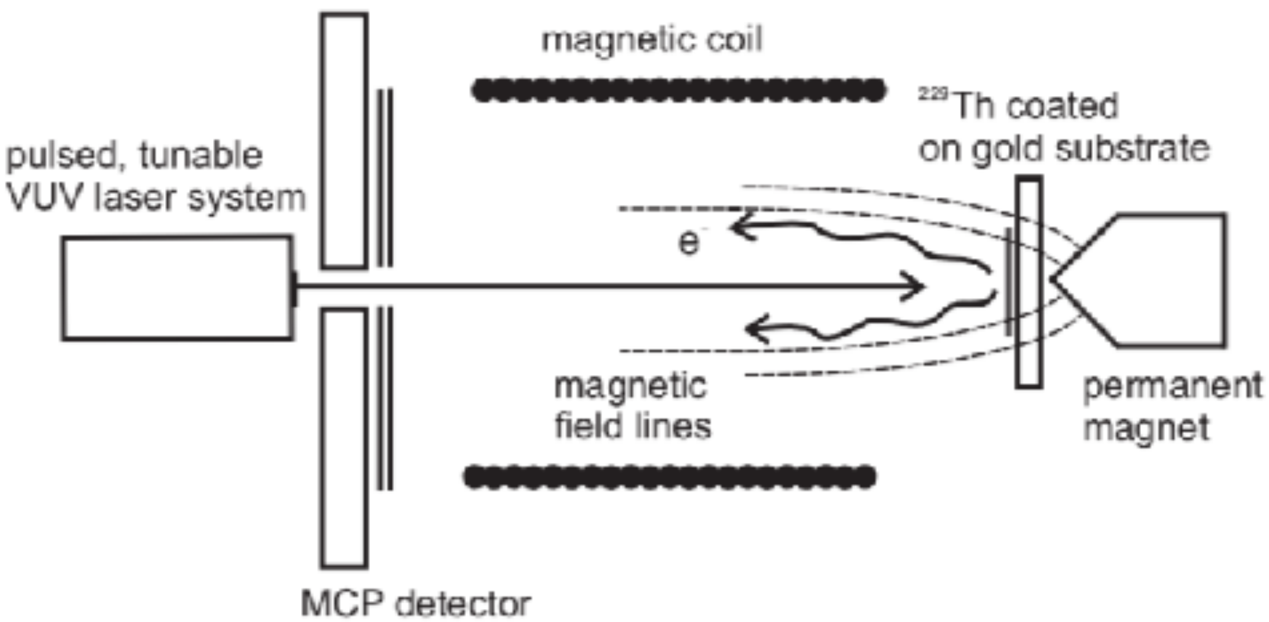


- No severe background in the signal range. (2014)

By Simon Stellmer

Excitation, Detection using IC electron

Laser excitation with magnetic bottle

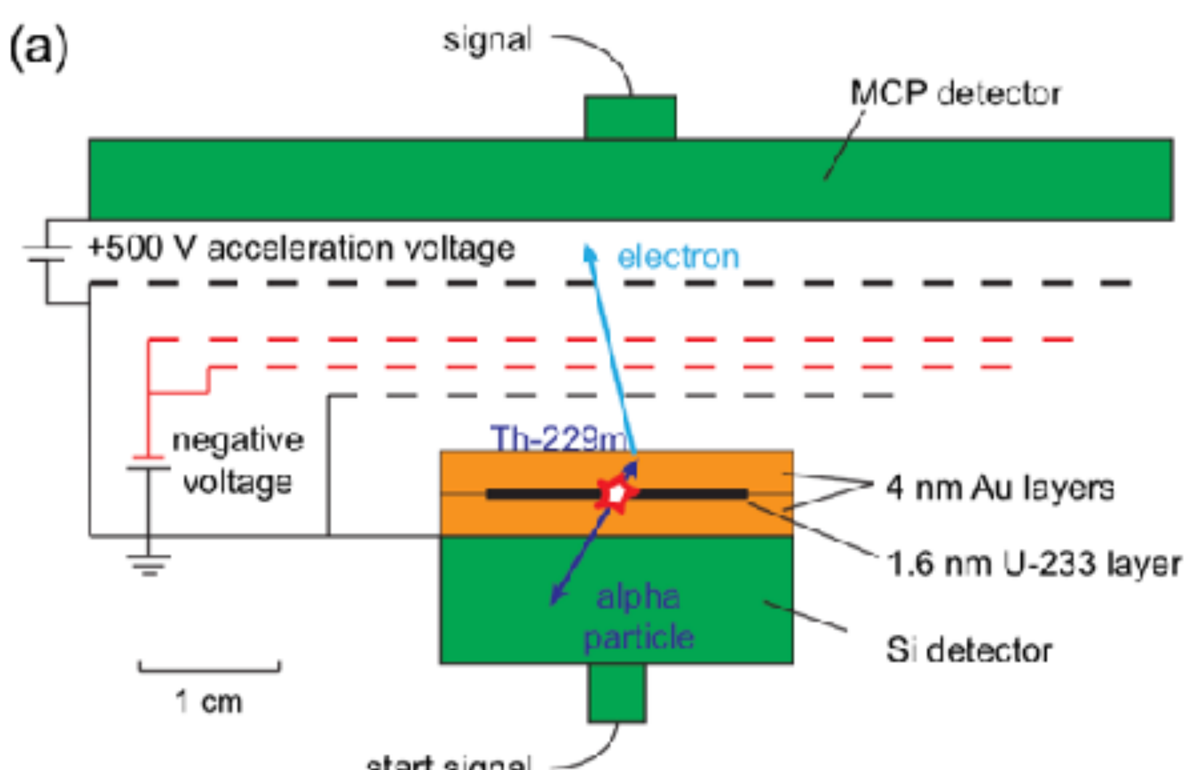


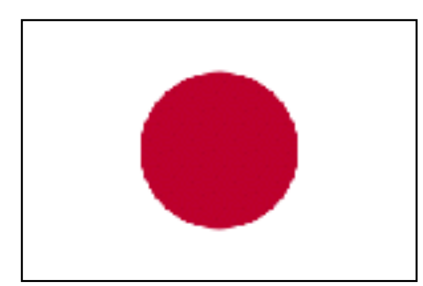
S/N ~ 10^4

<3 day scan

Lars von der Wense et al., PRL 119 132503 (2017)

Energy measurement of IC electron



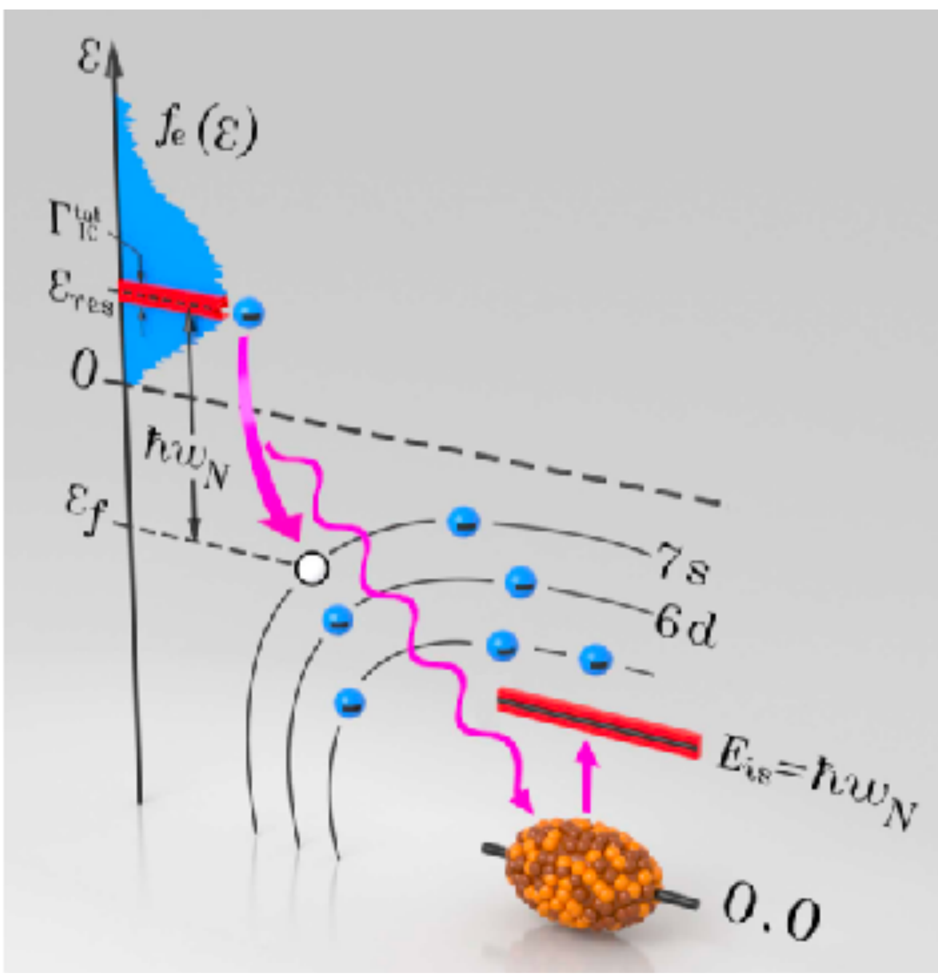


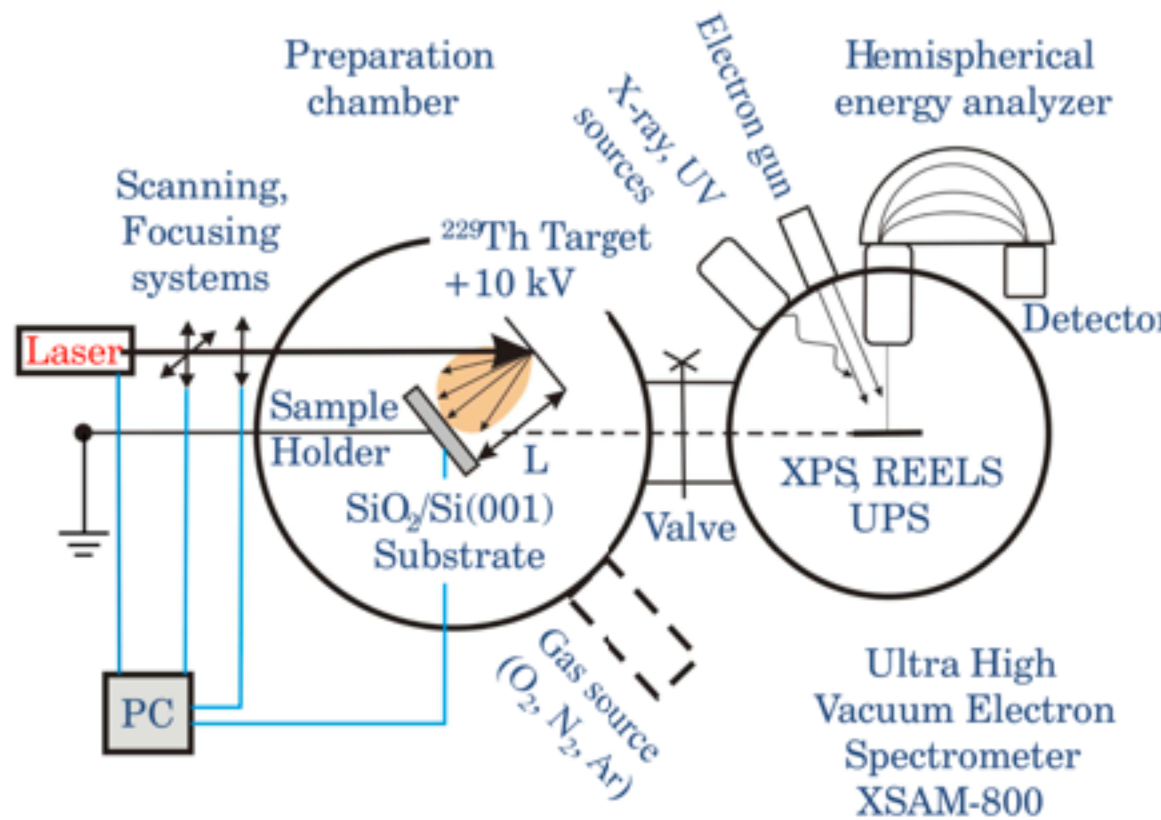
S. Stellmer et al., arXiv 1805.04929v1

Excitation in laser plasma

Inverse IC using Laser ablation

Inverse IC using Laser ablation



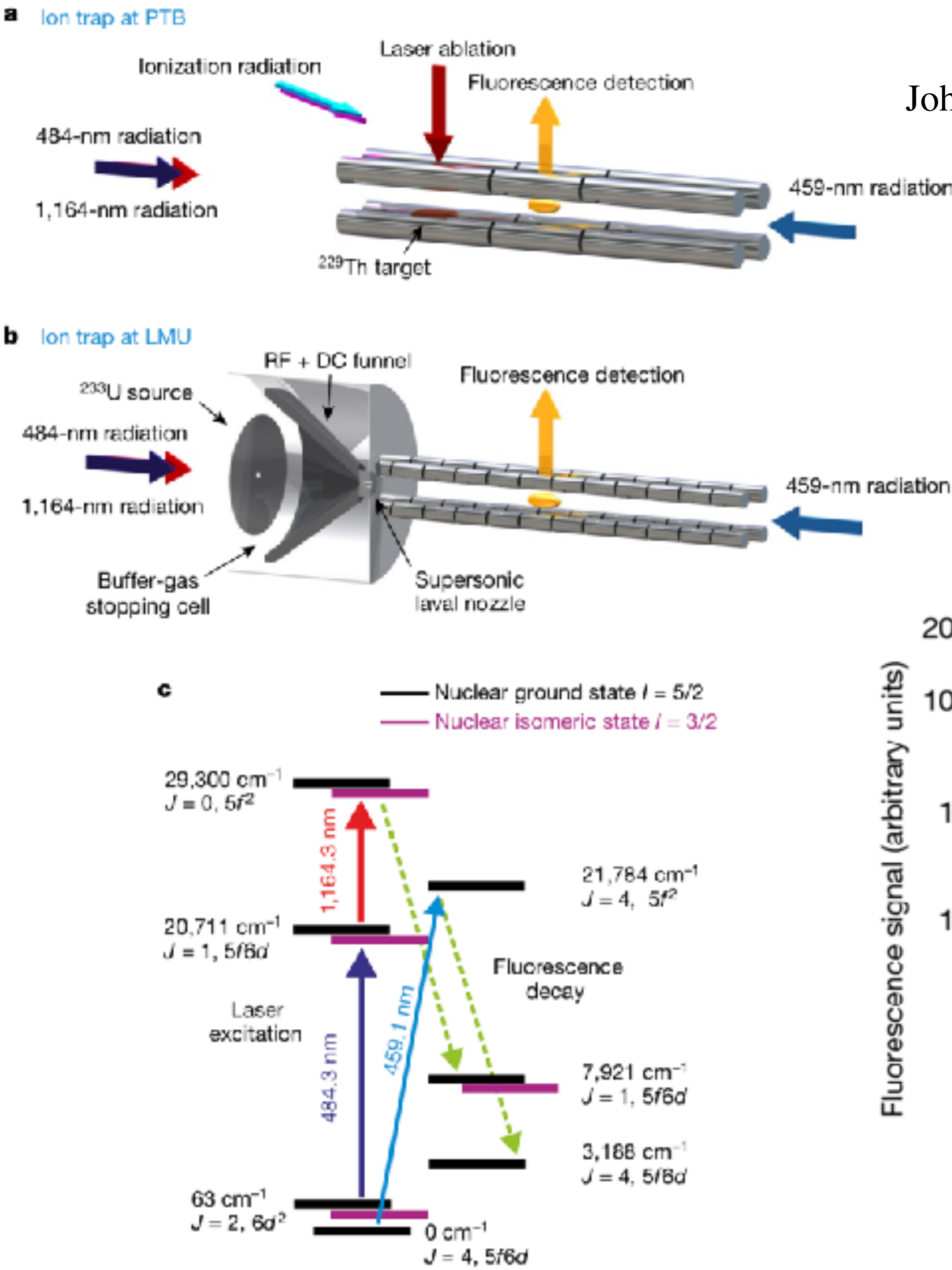


P.V.Borisyuk et al., arXiv 1804.00299v1

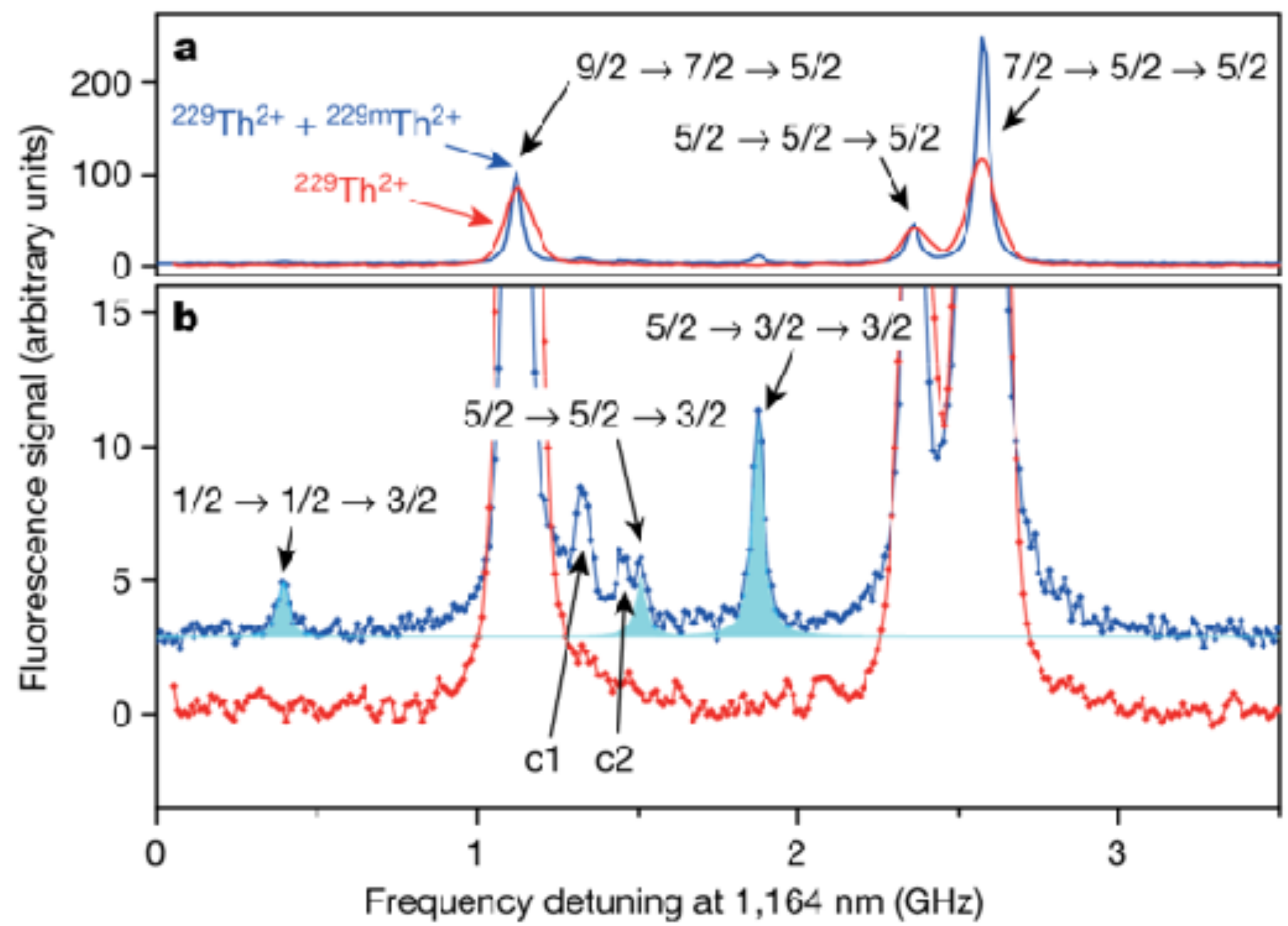
Eis = 7.1(+0.1-0,2) eV

T1/2 = 1880±170 s

Spectroscopy



Johannes thielking et al., Nature vol 556 (7701) pp. 321-325 (2018)



Summary

- New method using high intense x-ray source has been developed.
- We are now ready for full search of Th-229 29.2 keV state.
- If we could observe NRS signal of 29.2 keV, then we start search for VUV photon using Isomer state.