

原子核時計実現に向けたトリウム²²⁹原子核の 極低エネルギーアイソマー状態探索



岡山大学 自然科学研究科 博士後期課程

岡山大学 異分野基礎科学研究所

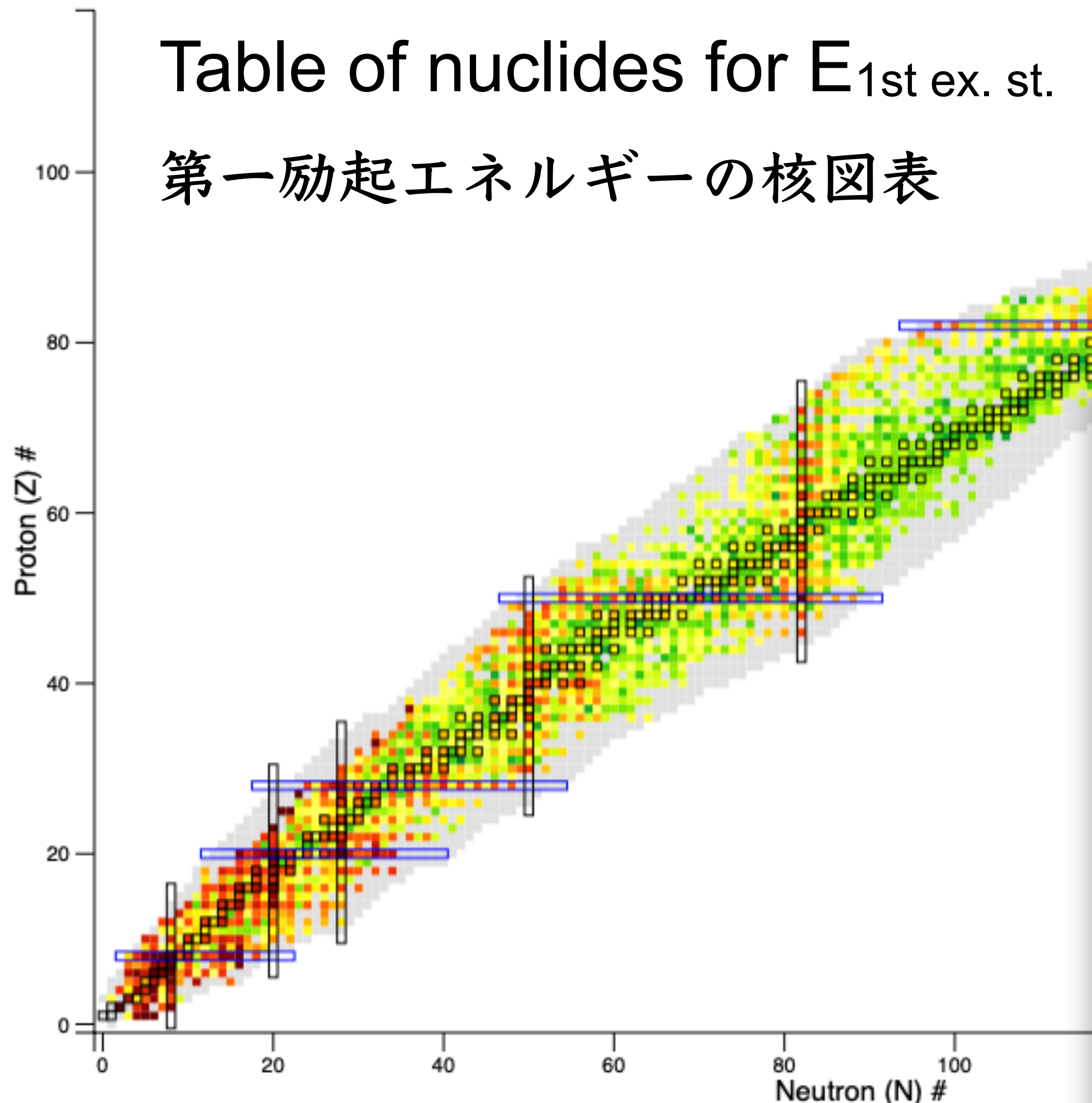
岡井晃一



What's Thorium 229?

Table of nuclides for $E_{1st\ ex. \ st.}$

第一励起エネルギーの核図表

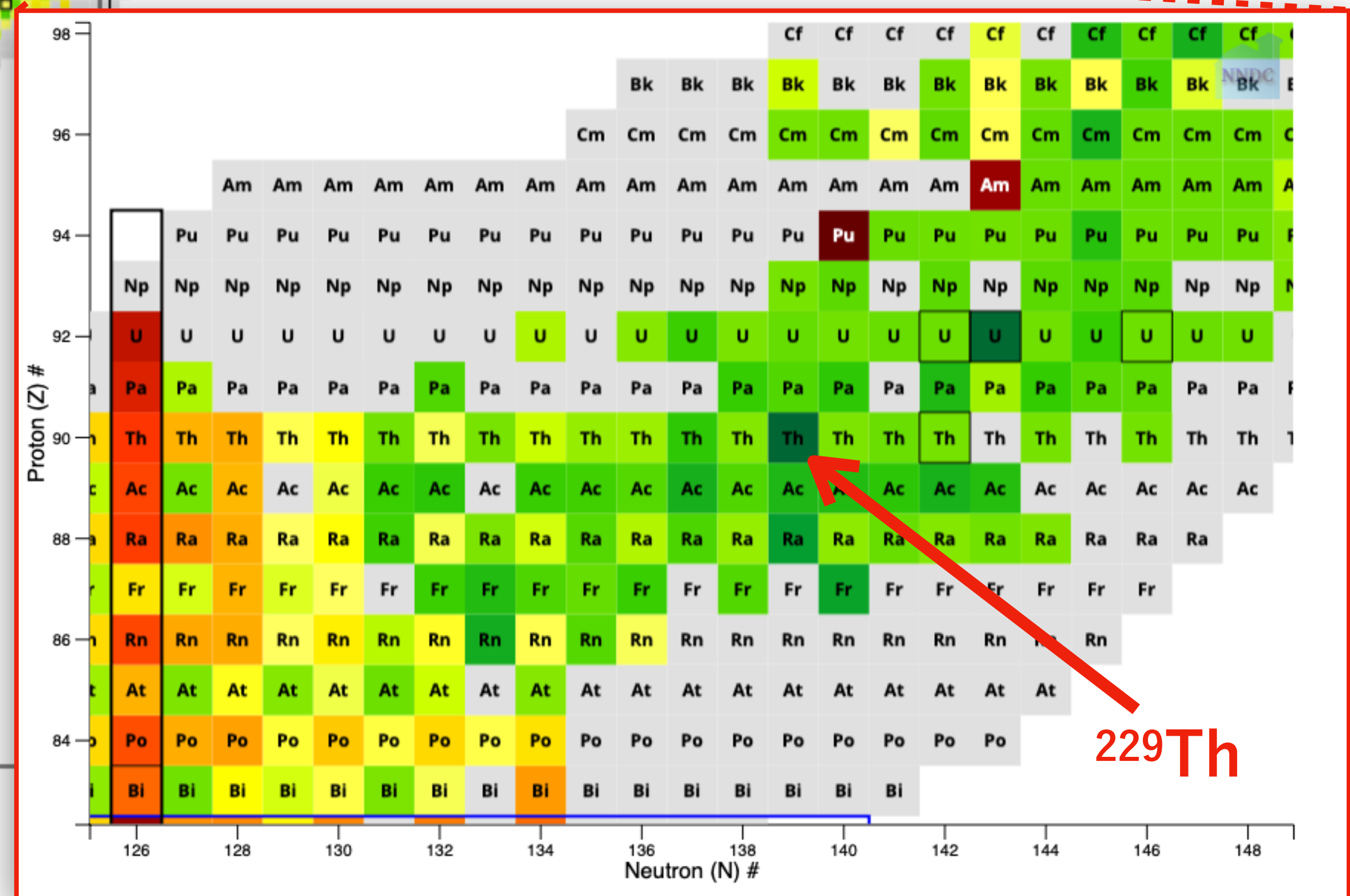


- **Lowest nuclear excited state**

- Typically keV ~ MeV scale

1st lowest: ^{229}Th ~8 eV

2nd lowest: ^{235}U ~76 eV



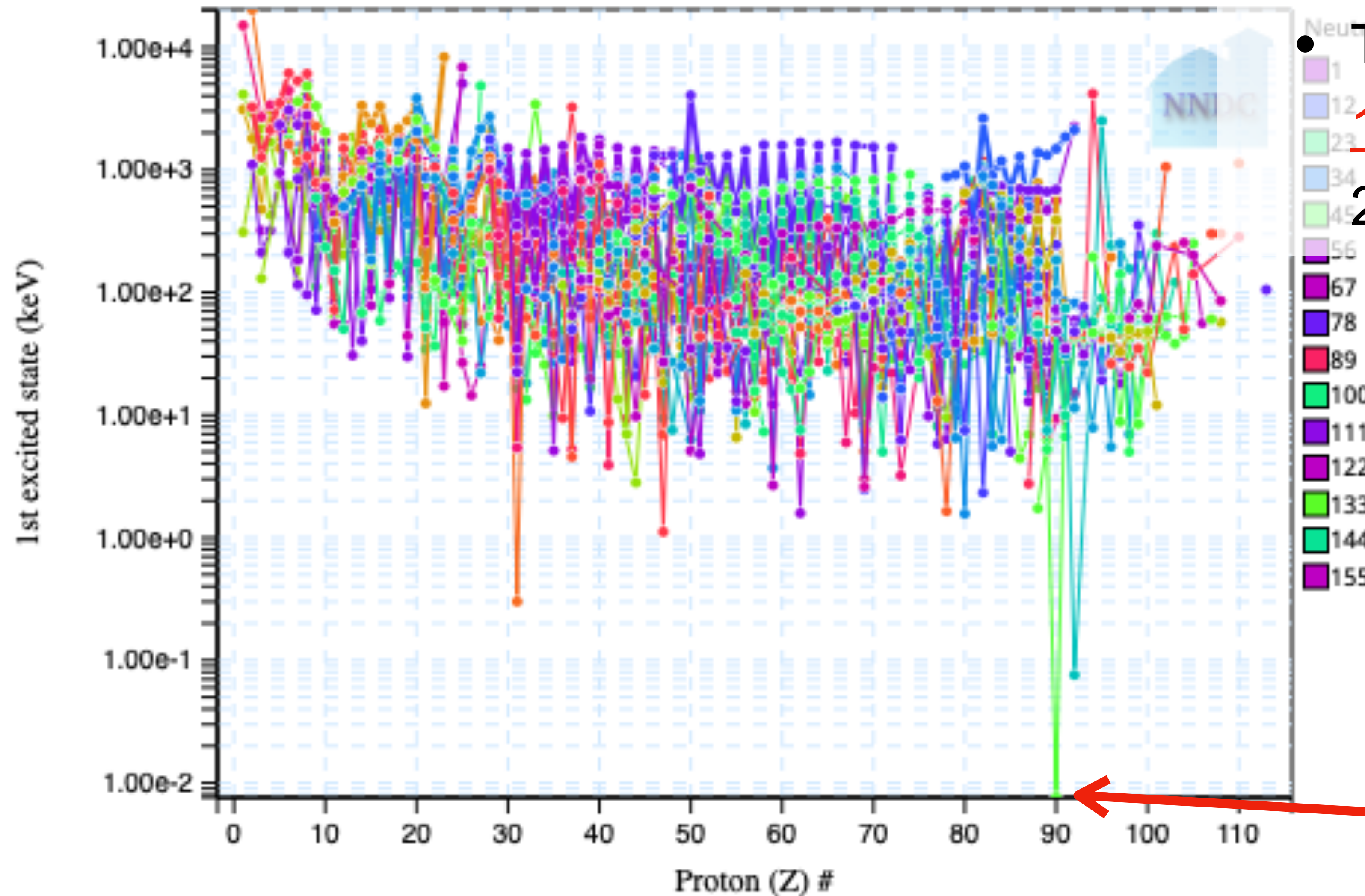
What's Thorium 229?

- **Lowest nuclear excited state**

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^{229}Th

What's Thorium 229?

- **Lowest nuclear excited state**

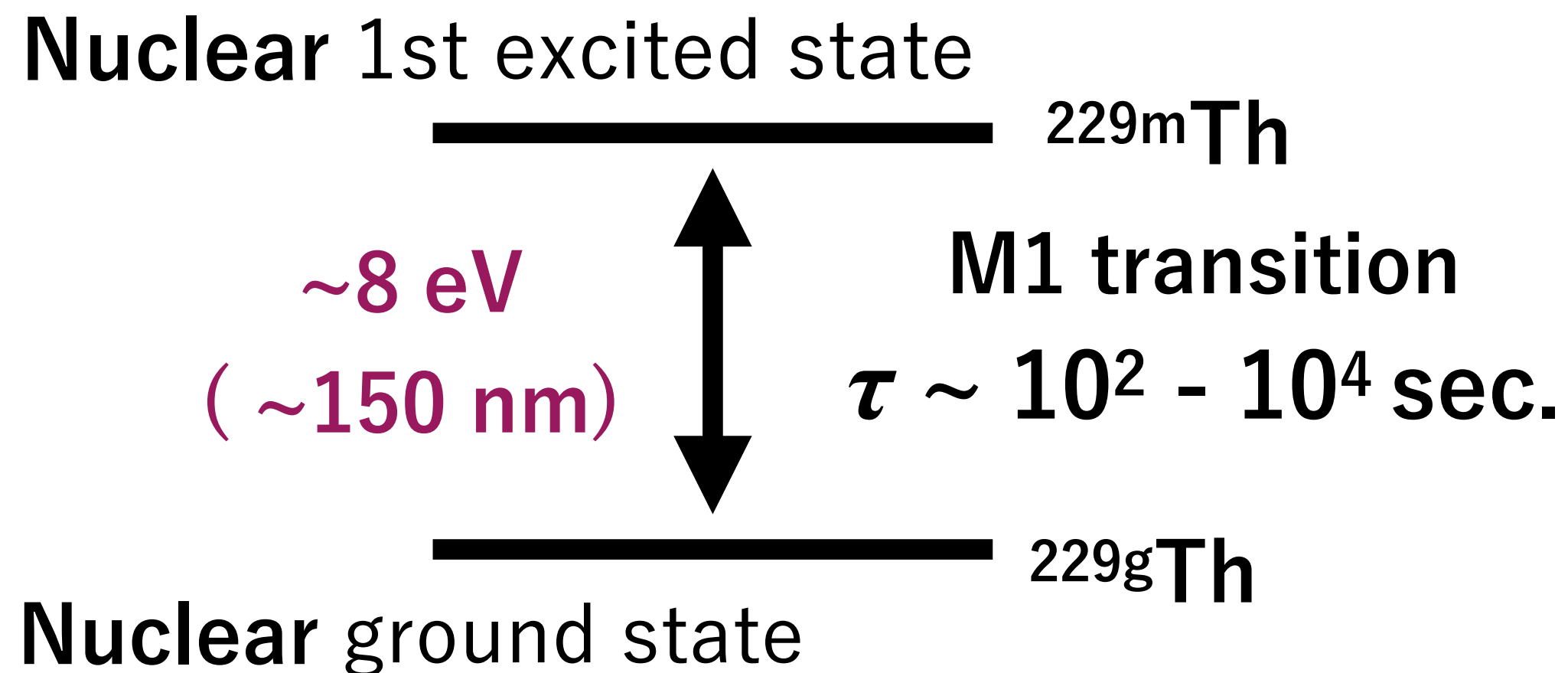
- Typically keV ~ MeV scale

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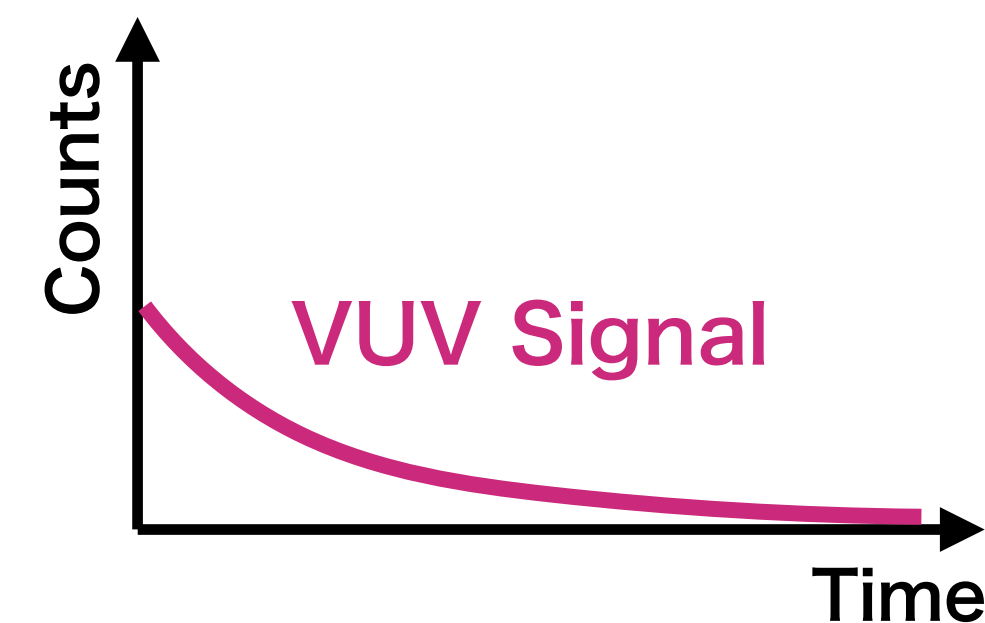
2nd lowest: ^{235}U ~76 eV

アイソマー状態
 ^{229}Th has the very low isomeric state $^{229\text{m}}\text{Th}$:
Direct nuclear laser excitation is possible
($\lambda \sim 150$ nm)

- $^{229\text{m}}\text{Th}$: nuclear isomer



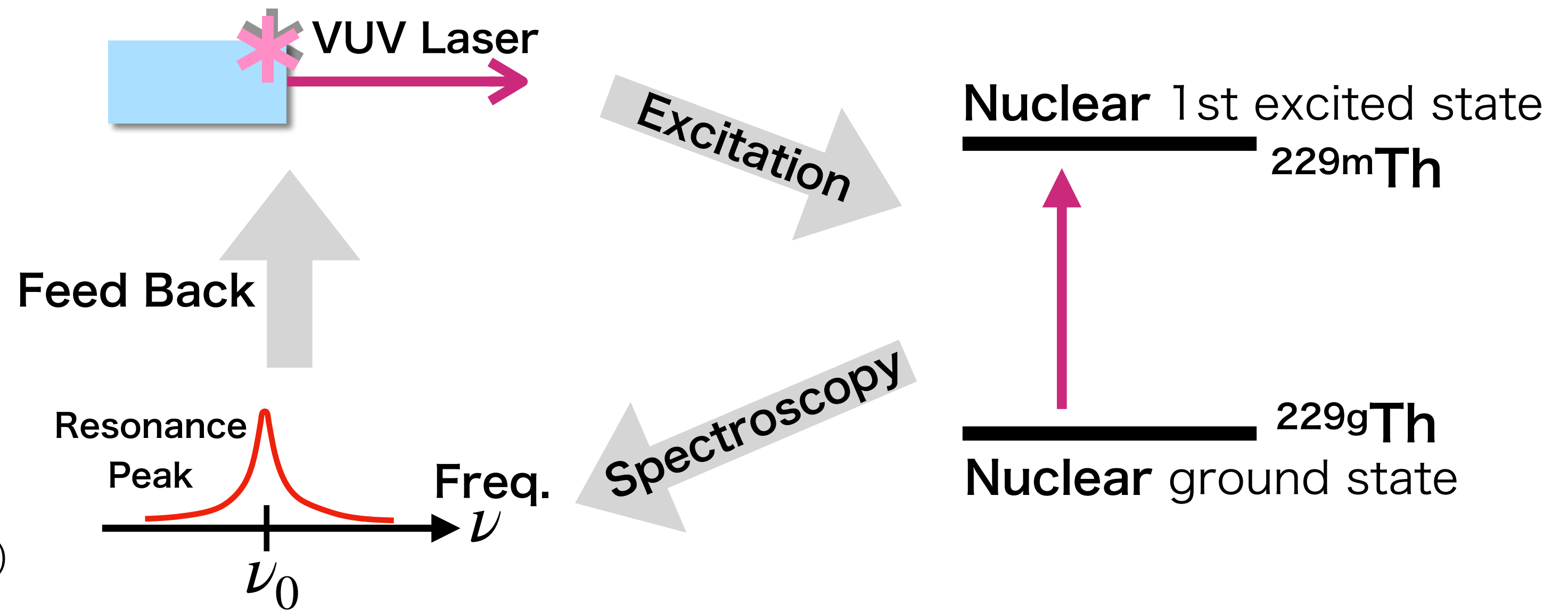
- **Lifetime** and **absolute energy** are unknown
- Optical transition hasn't been observed yet
- 150 nm light is **Vacuum Ultra Violet (VUV)**



- **Nuclear clock:**
 - use nuclear transitions as frequency standard

- Estimated uncertainty $\sim 10^{-19}$

C. J. Campbell et al., Phys. Rev. Lett. 108, 120802 (2012)
 E. Peik and M. Okhapkin, C. R. Physique 16, 516 (2015)



- Very sensitive to **time variation of fine structure constant (α)**

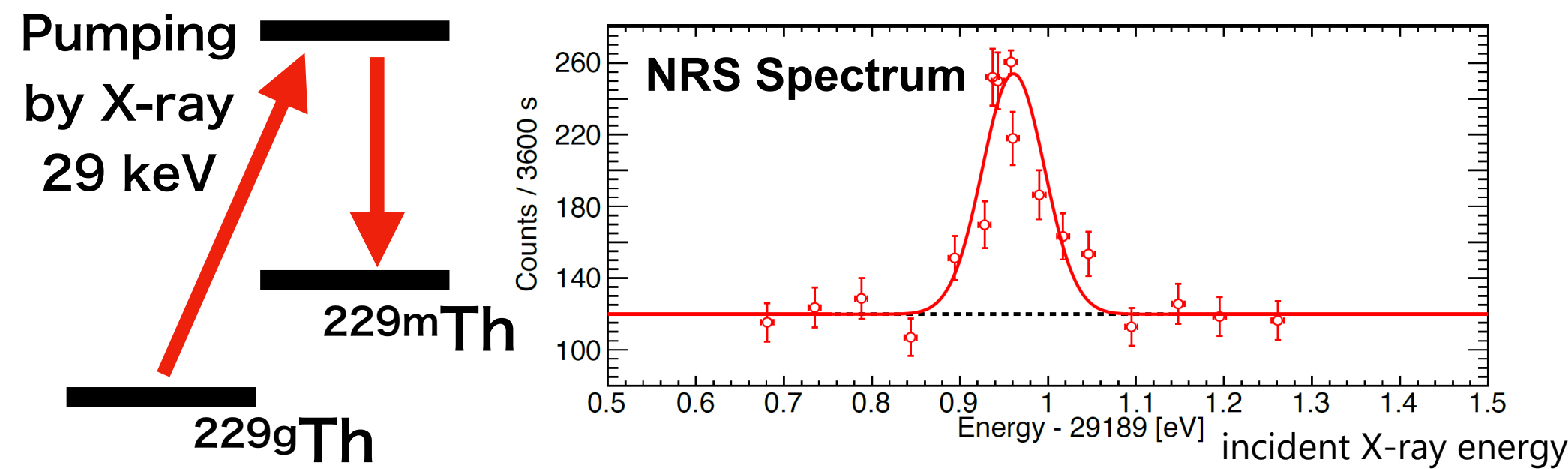
$$\frac{\delta f}{f} = \frac{\Delta E_{\text{Coulomb}}}{E_{\text{isomter}}} \frac{\delta \alpha}{\alpha}$$

P. Fadeev, et. al., Phys. Rev. A **102**, 052833 (2020)

Strategy to Detect VUV Signal

Step 1

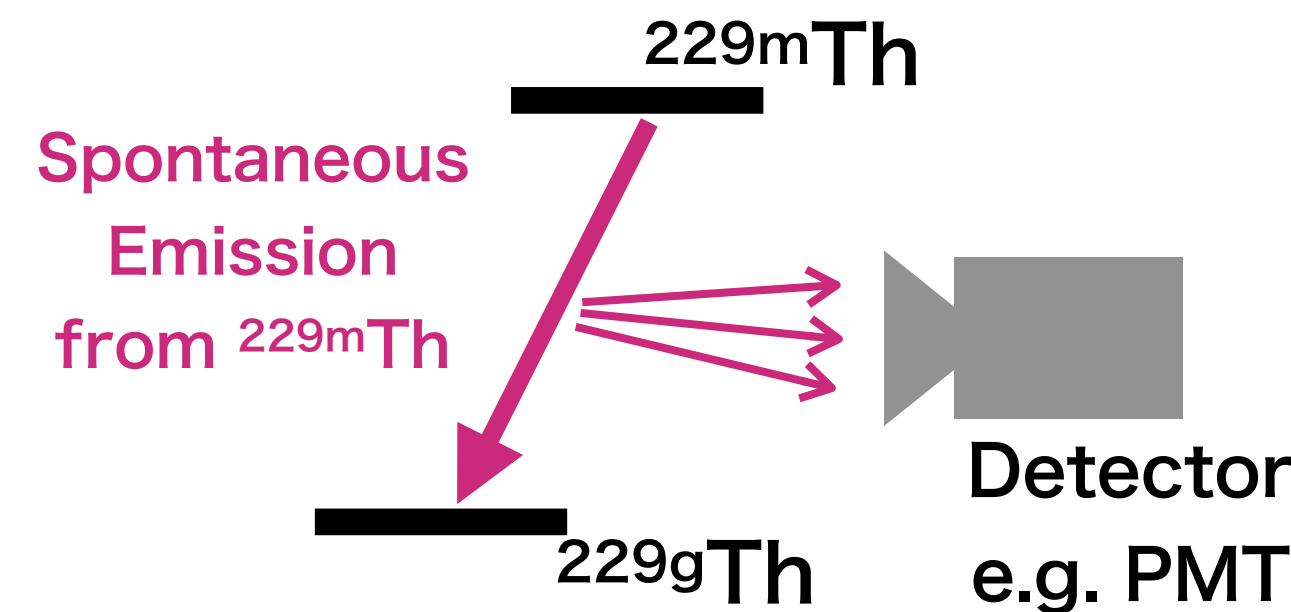
Artificial production of ^{229m}Th by NRS* method



Step 2

Our Current Target

Detect VUV light emitted from ^{229m}Th



Step 3

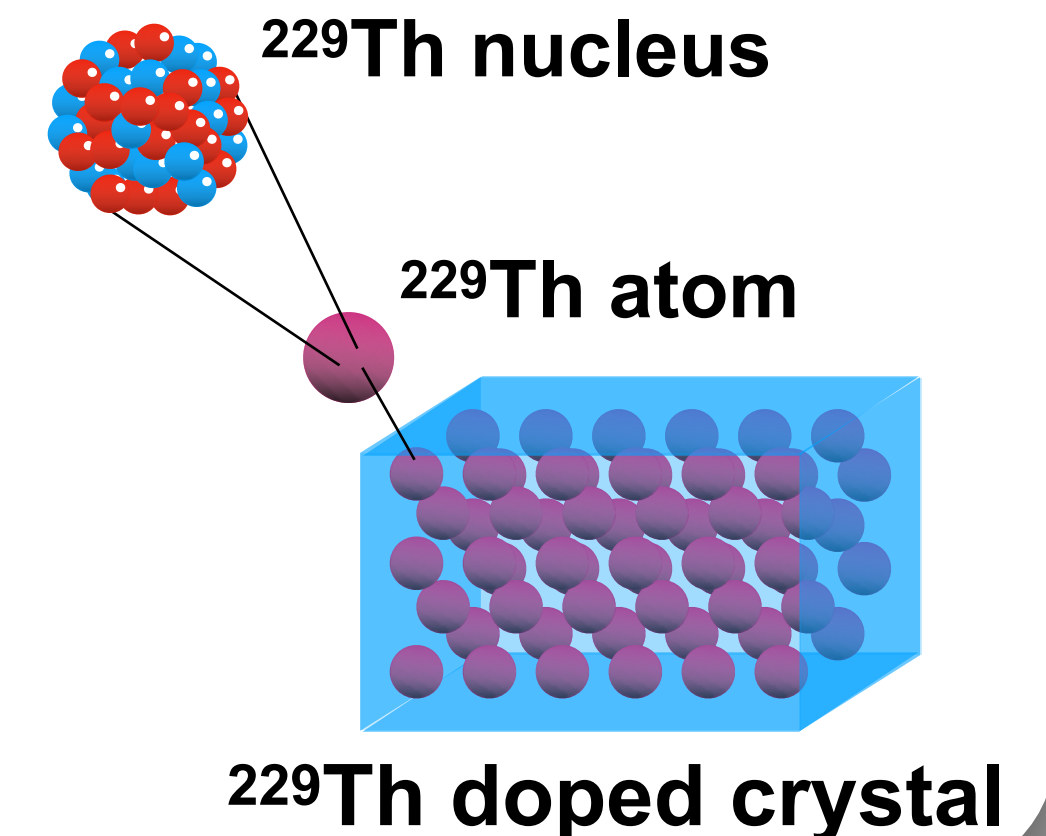
Spectroscopy of VUV Signal to determine ^{229m}Th energy

E.g. Developing detector which has large acceptance of VUV light

* Nuclear Resonance Scattering

- To precisely measure the energy of isomeric state ^{229m}Th , We set three steps
- Production of ^{229m}Th can be confirmed by measuring NRS signal
 - **Step 1 is already achieved at SPring-8!**
- ^{229}Th nuclei can be contained in the optical crystal
 - Nuclei aren't affected by crystal because valence electron work as shield

T. Masuda, et al., Nature, 573, 238(2019)

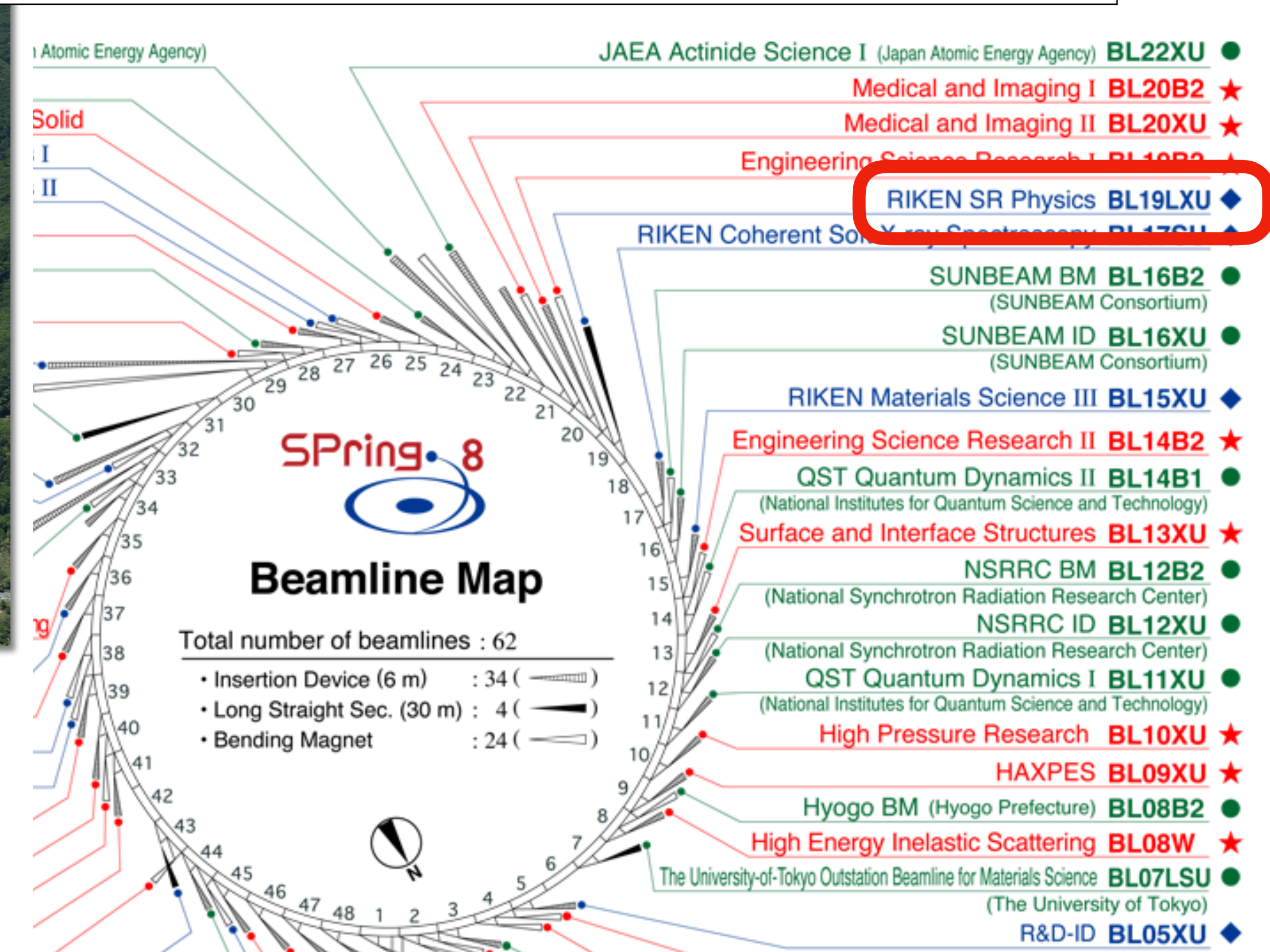


BL19LXU on SPring-8



“SPring-8 is a large synchrotron radiation facility which delivers the most powerful synchrotron radiation currently available.”
“The research conducted at SPring-8, located in Harima Science Park City, Hyogo Prefecture, Japan, includes nanotechnology, biotechnology and industrial applications”

http://www.spring8.or.jp/en/about_us/whats_sp8/

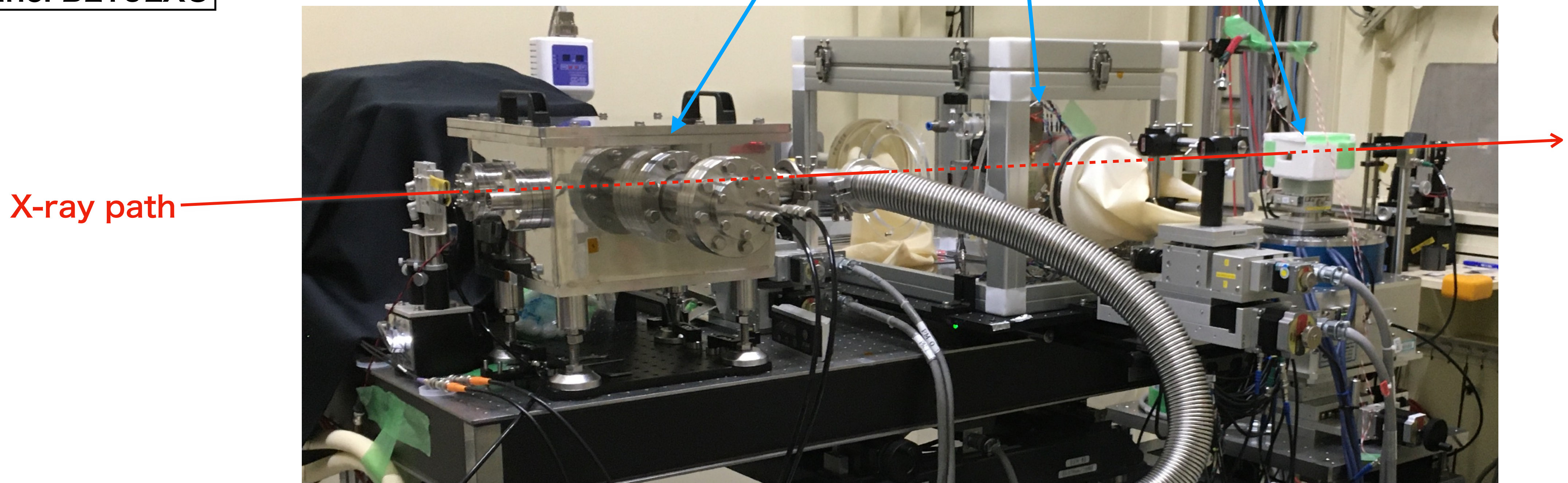
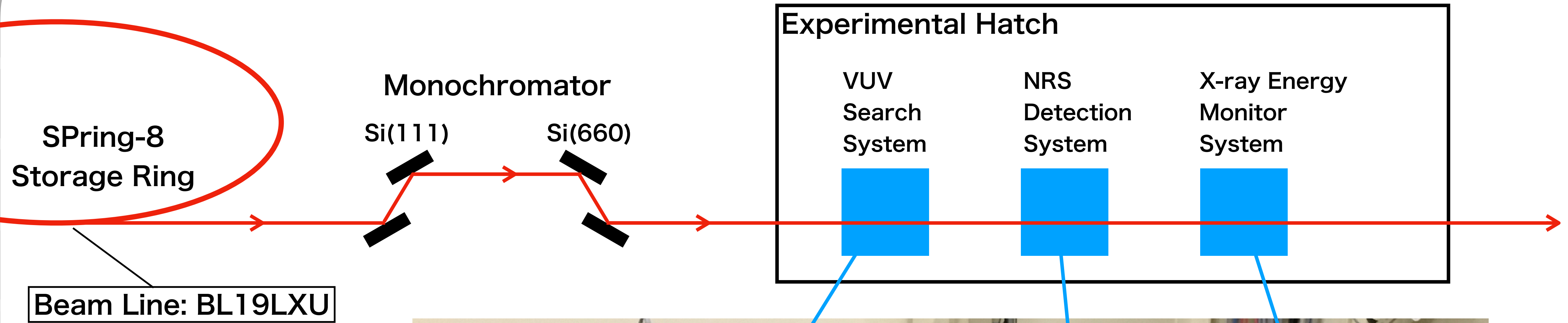


- We use **BL19LXU**
 - Energy Range: 7.1~18 keV (1st), 22~51 keV (3rd)
 - $\sim 2 \times 10^{14}$ photons/s @14 keV

http://www.spring8.or.jp/wkg/BL19LXU/instrument/lang-en/INS-0000000361/instrument_summary_view

http://www.spring8.or.jp/en/about_us/whats_sp8/facilities/bl/map/

Experimental Setup at SPring-8



- **Procedure of VUV search**

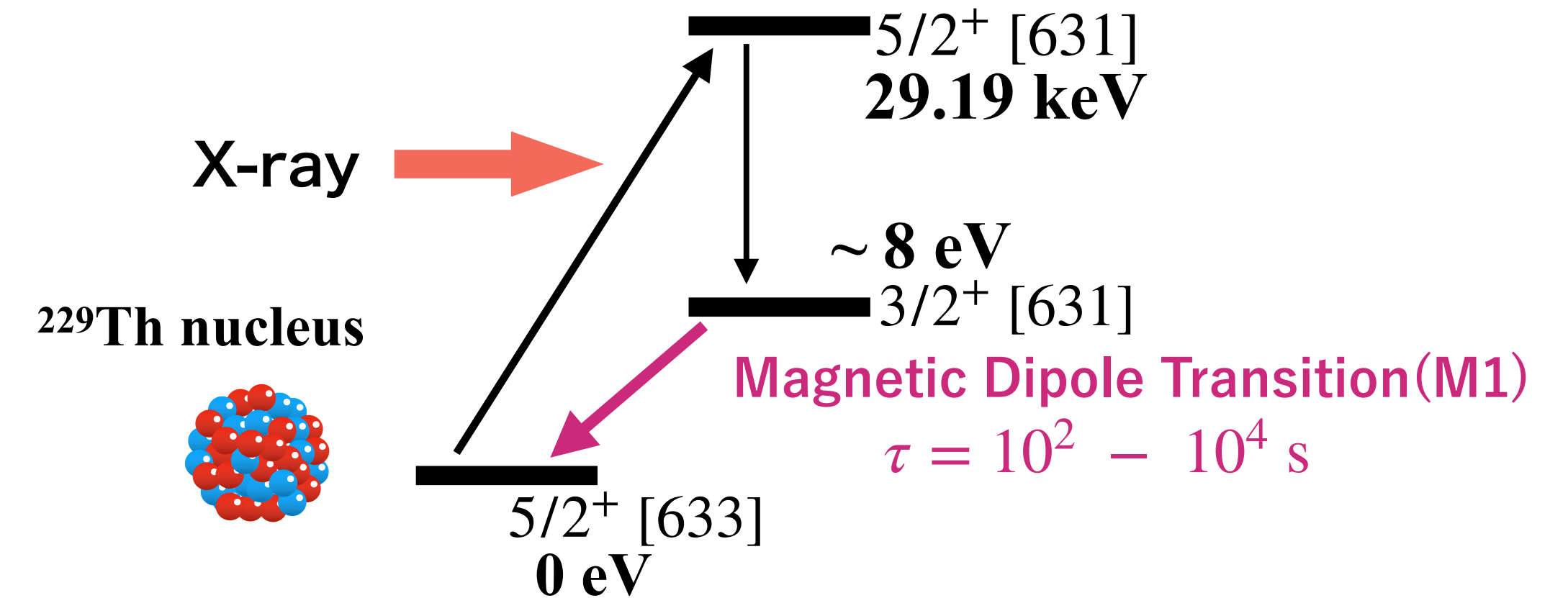
1. Produce $^{229\text{m}}\text{Th}$ by NRS
2. Detect the emitted photons from $^{229\text{m}}\text{Th}$

- **Target crystal**

- The target must have VUV transparency around 150 nm
 - Materials: Calcium fluoride (CaF_2), LiSAF (LiSrAlF_6)
- $^{229}\text{Th}:\text{CaF}_2$ crystals are developed by TU Wien group
 - ~50% transmittance at 150 nm
 - Thorium 229 density is $4 \times 10^{17} / \text{cm}^3$

S. Stellmer, et. al., Scientific Reports 5, 15580 (2015)

- $^{229}\text{Th}:\text{LiSAF}$ crystals are developed by UCLA group



size: 1x1x1 mm

VUV Signal and Backgrounds Estimation

- Estimation of **VUV signal rate**

- Time depend rate : $R_{\text{detection}}(t) = f R_{\text{isomer}} [1 - \exp(-T/\tau)] \exp(-t/\tau)$

- τ : Lifetime of $^{229\text{m}}\text{Th}$

- T : Irradiation time of X-ray beam from SPring-8

- R_{isomer} : Production rate of $^{229\text{m}}\text{Th}$ (known by NRS measurement)

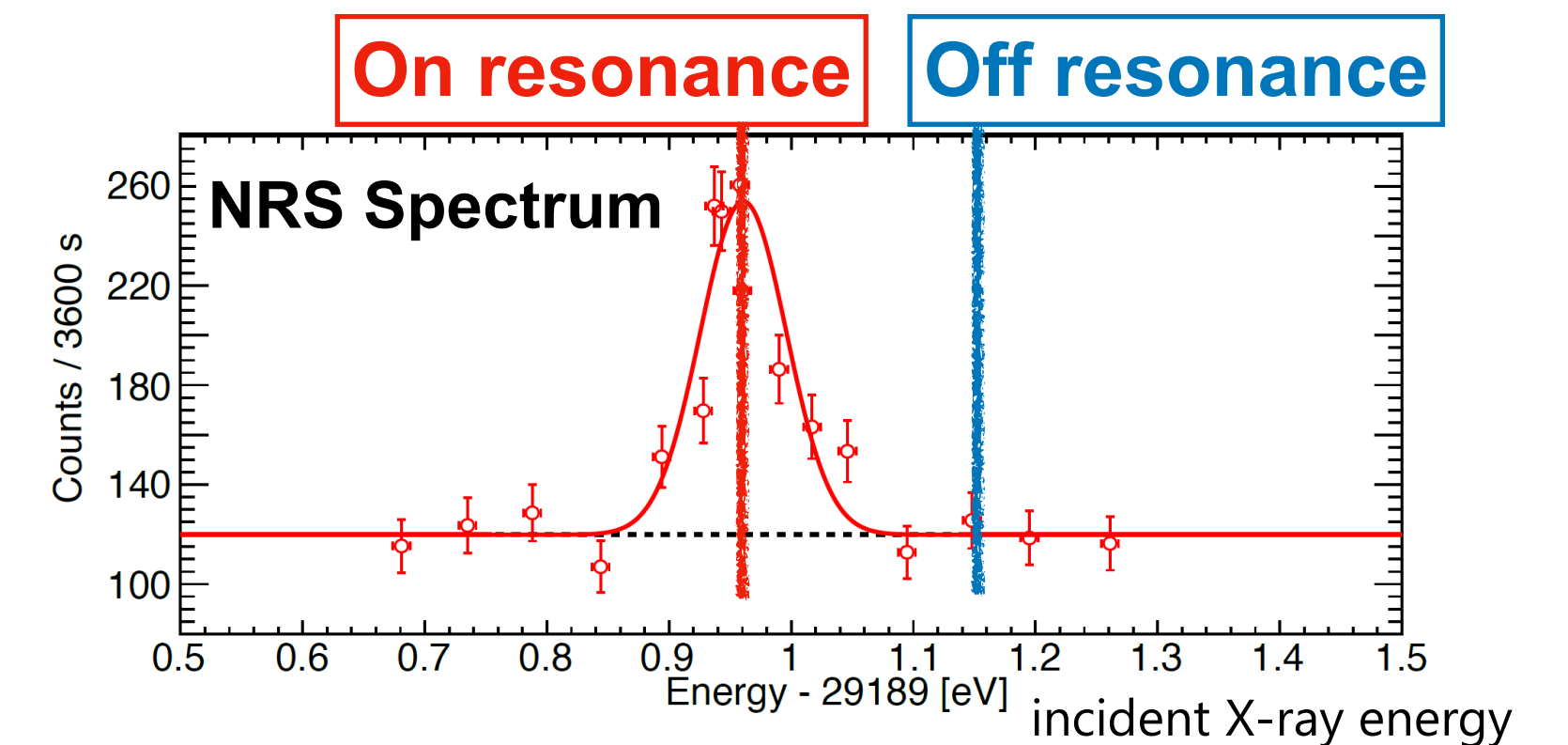
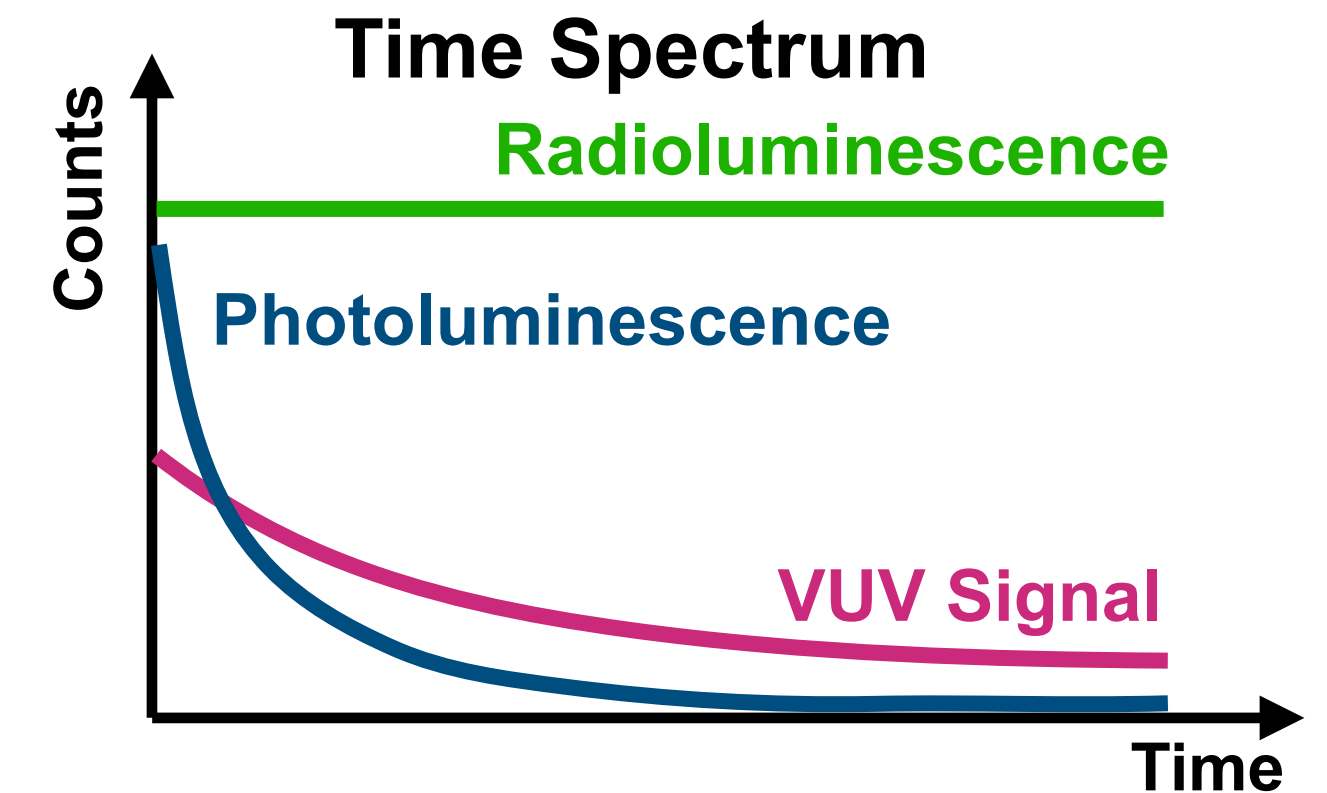
- Backgrounds

- **Radioluminescence**

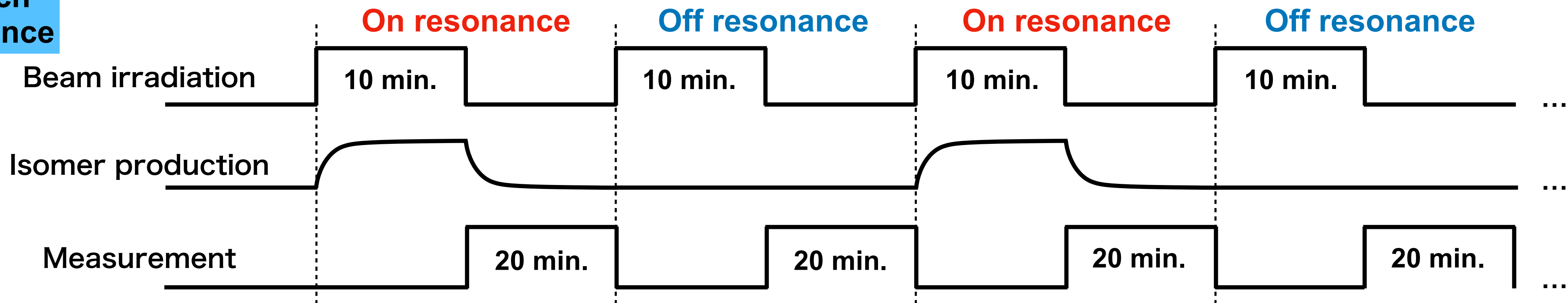
- It's caused by Alpha-decay and Beta-decay of ^{229}Th

- **Photoluminescence**

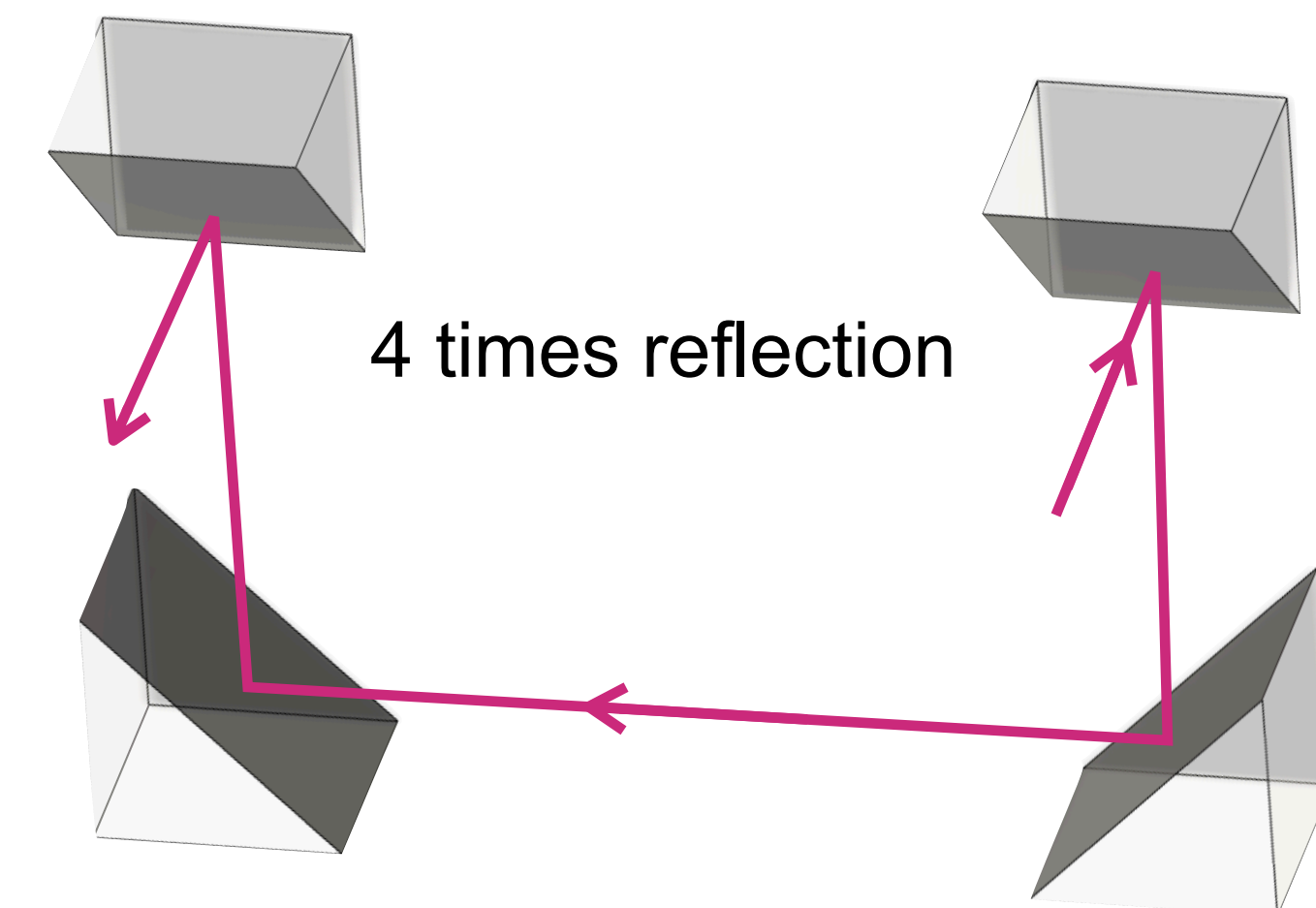
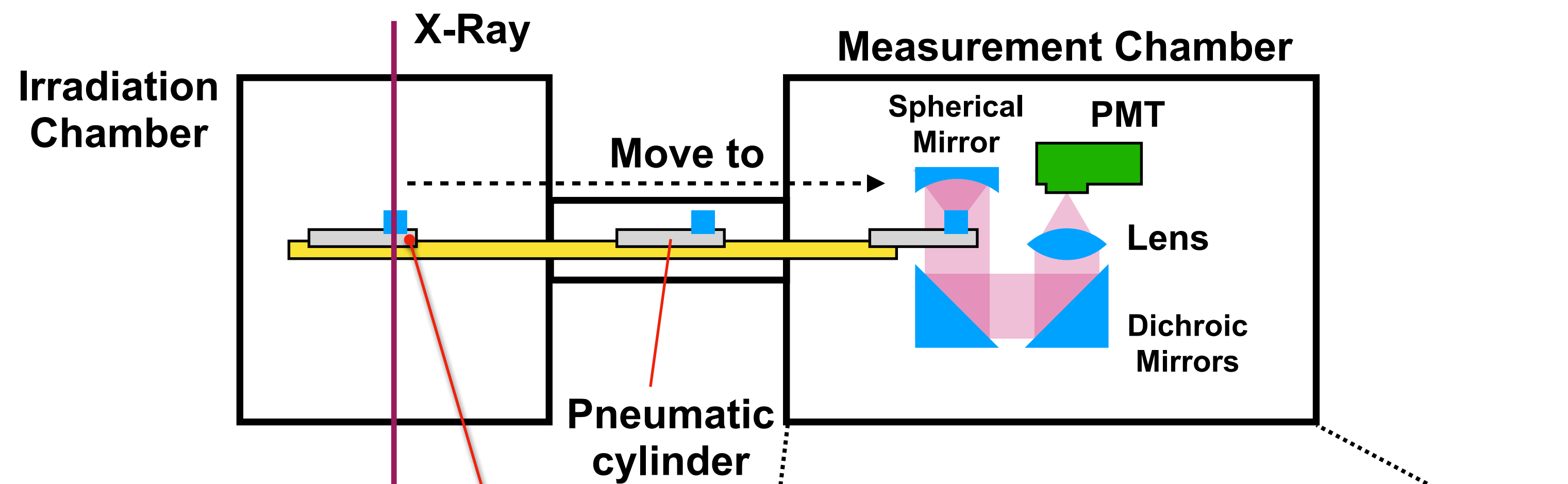
- It's caused by irradiation of X-ray to the target crystal



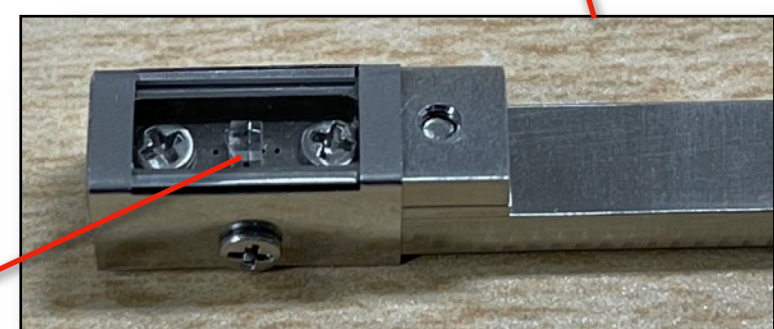
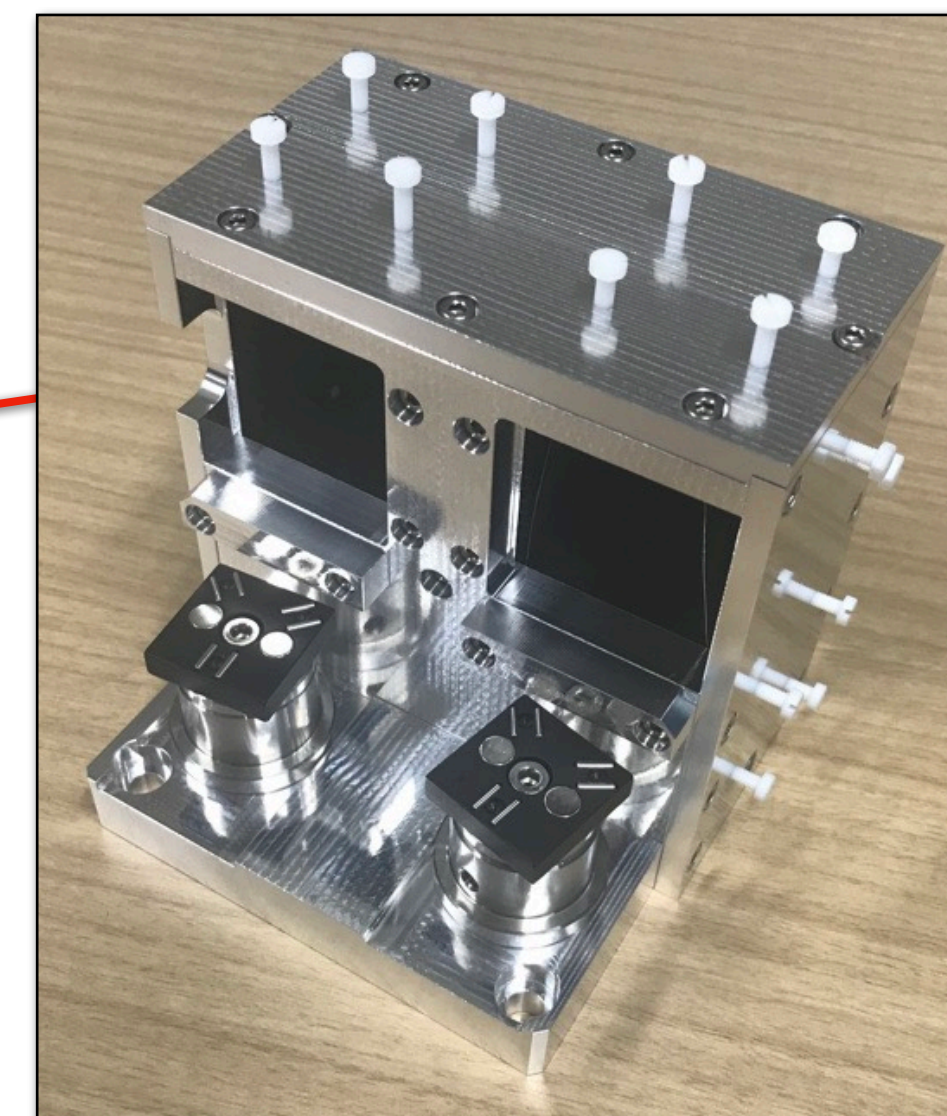
VUV Search Time sequence



Overview of VUV Setup



Dichroic Mirror Assembly

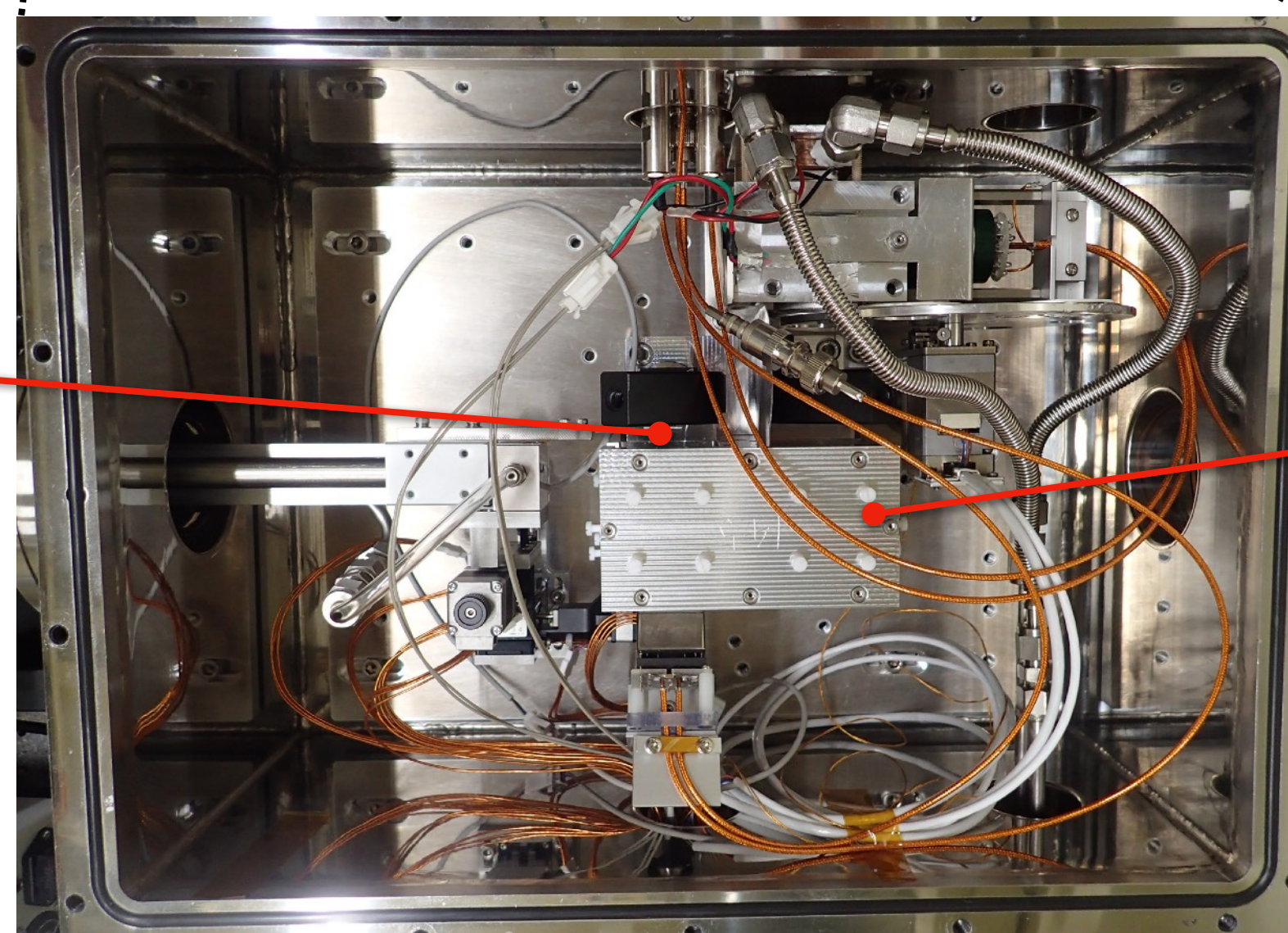


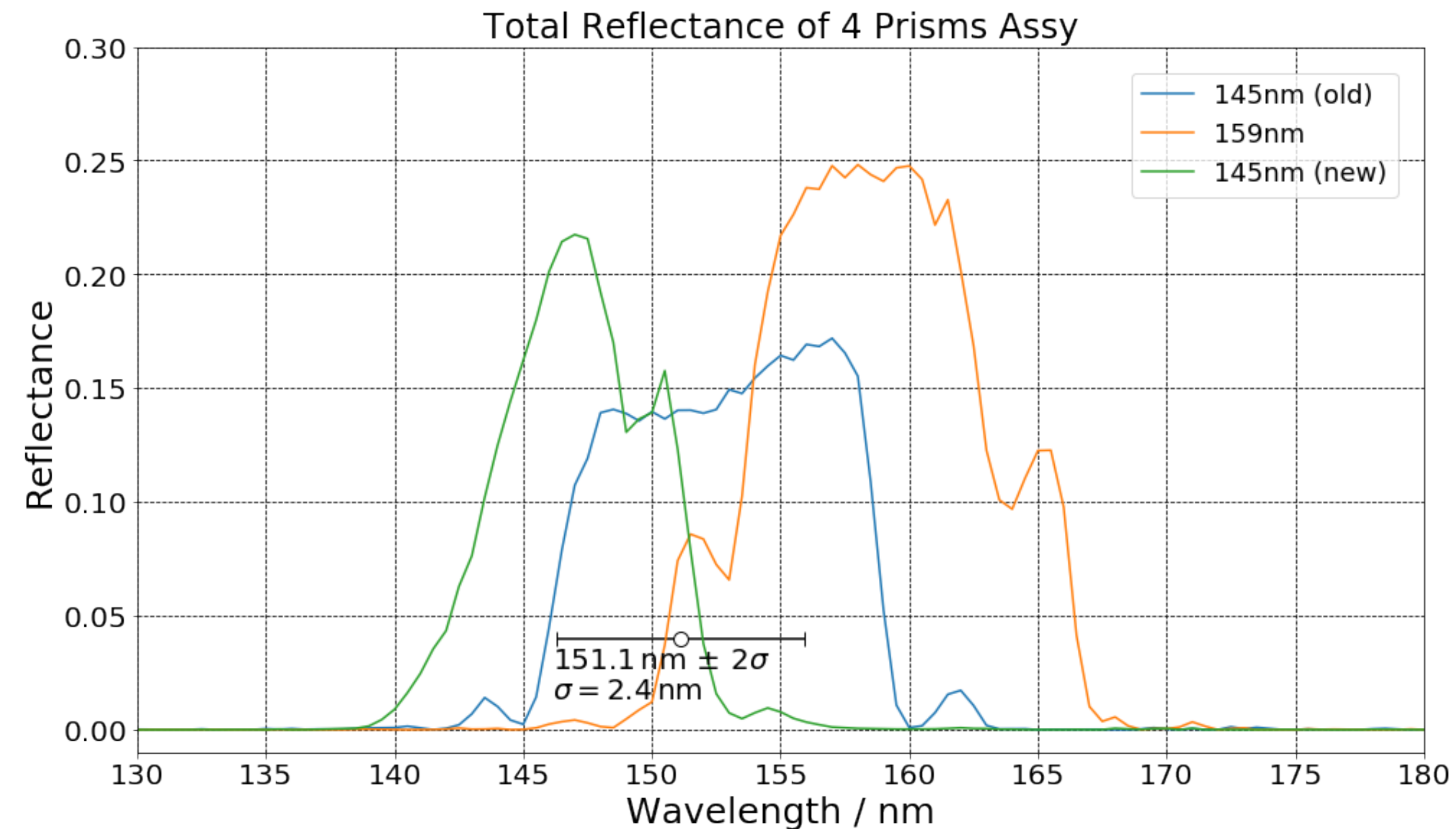
Crystal holder

$^{229}\text{Th}:\text{CaF}_2$
Crystal
(~ 1 mm³)
made by

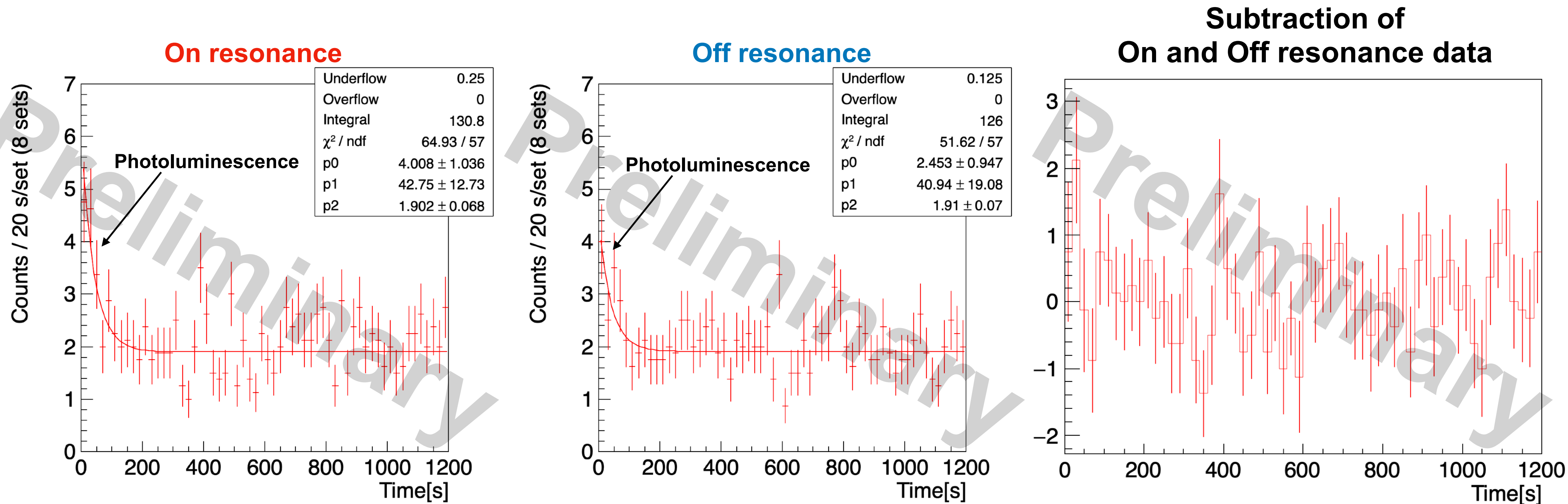


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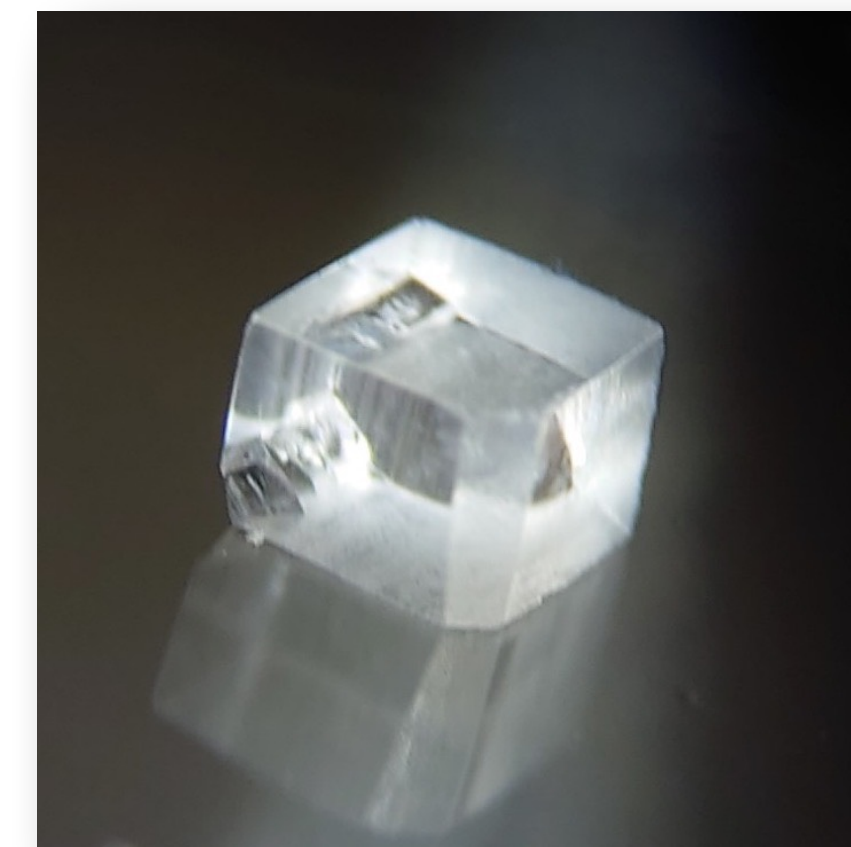
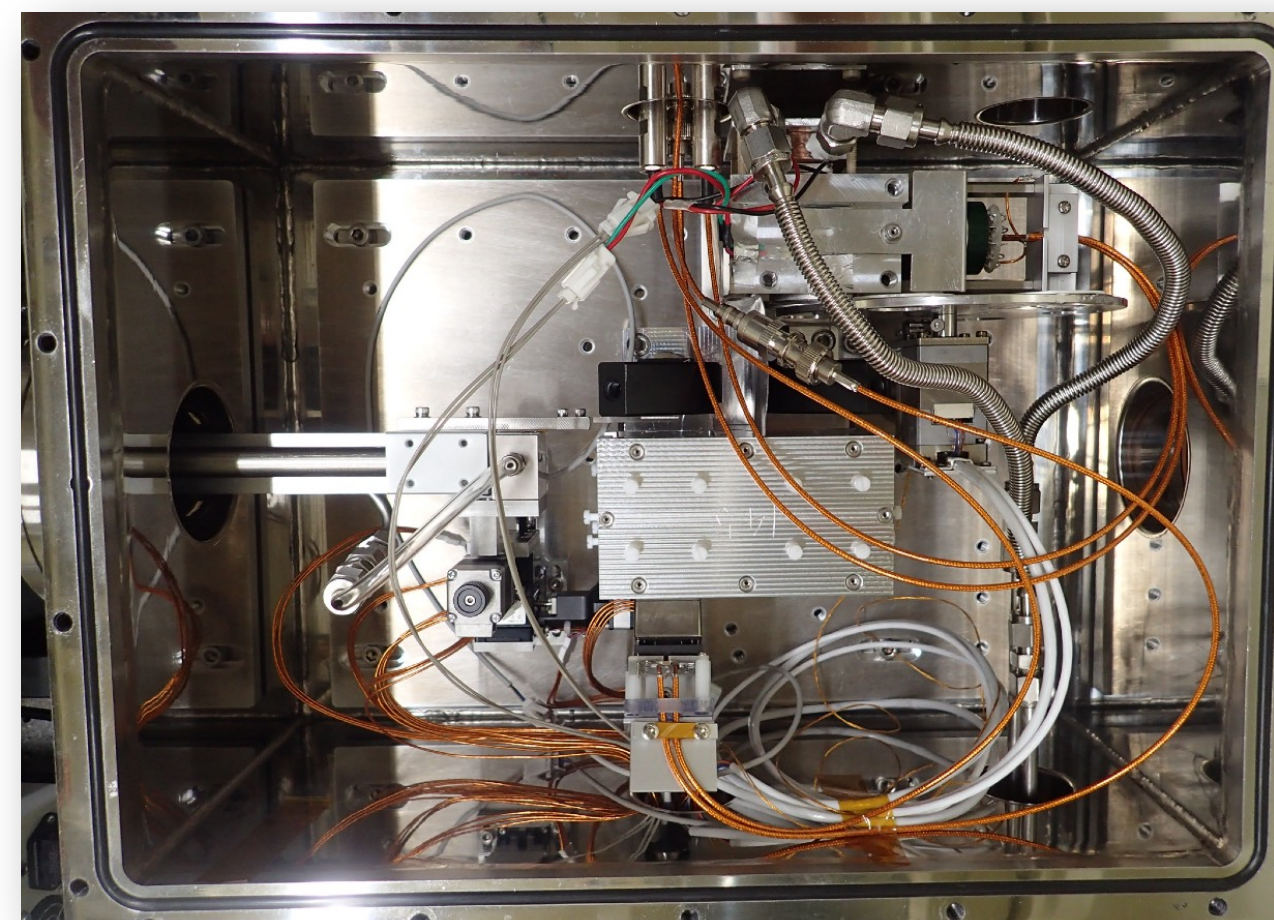
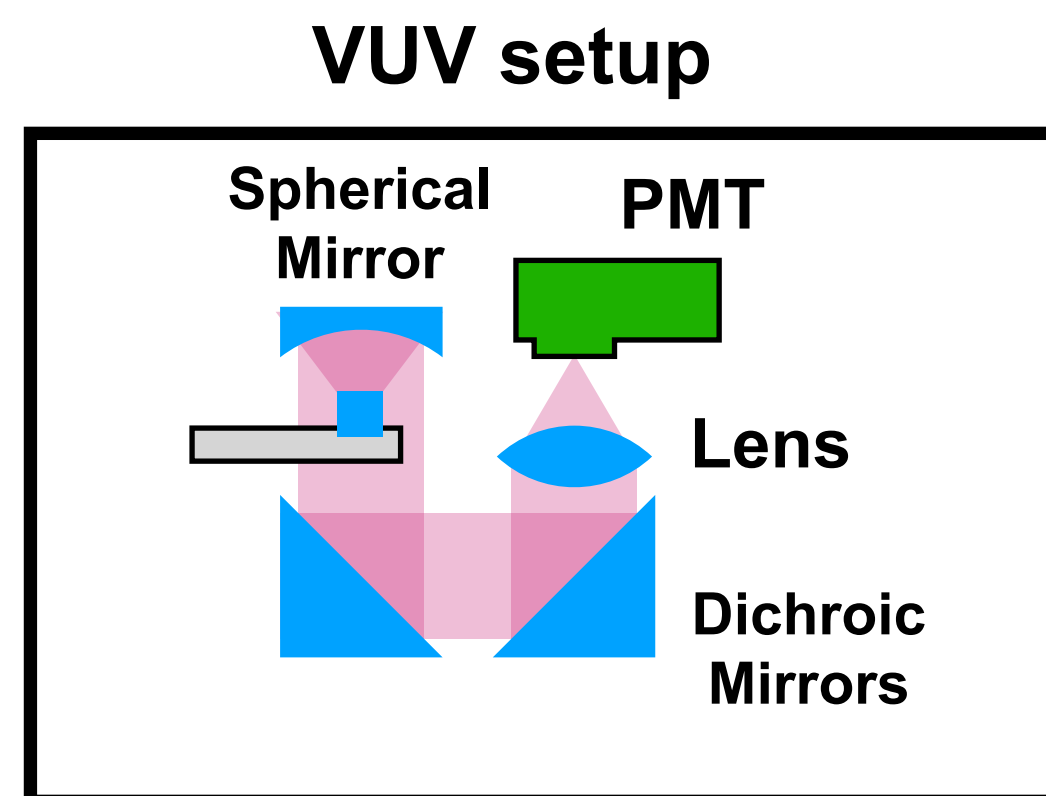




- Signal wavelength is determined by several variety of the prism assembly which has **four dichroic mirrors**.
- Reflectances of Dichroic mirrors had been measured by our own measurement system which will be introduced later.



- Time sequence
 - (10 min. irradiation + 20 min. measurement) x (On, Off res.) x 8 sets
- Most B.G. event such as radioluminescence was cut by veto
- No indication of the VUV signal so far
 - **Specification of optical setup is still unknown**



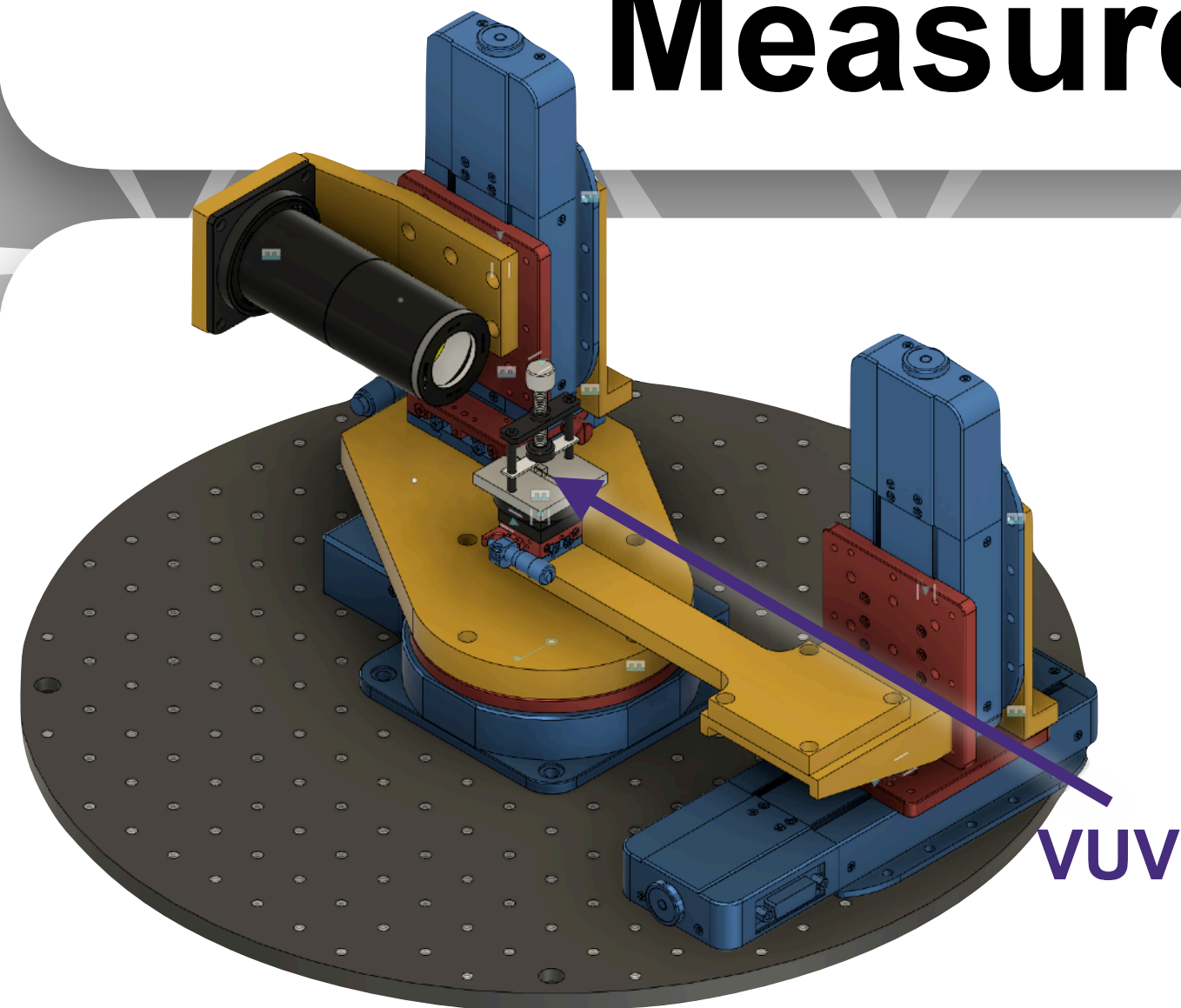
- Net efficiency : $\varepsilon = \prod_i \varepsilon_i$
 - Transmittance of the CaF_2 crystal (0.5?)
 - Reflectance of spherical mirror (0.8?)
 - Reflectance of dichroic mirrors
(0.6 / 1 mirror @150nm)
 - Transmittance of MgF_2 lens (0.8?)
 - Geometrical efficiency (0.0855)
 - Quantum efficiency of VUV PMT (0.23)

- Surface roughness varies with crystal
- custom-made radioactive crystal

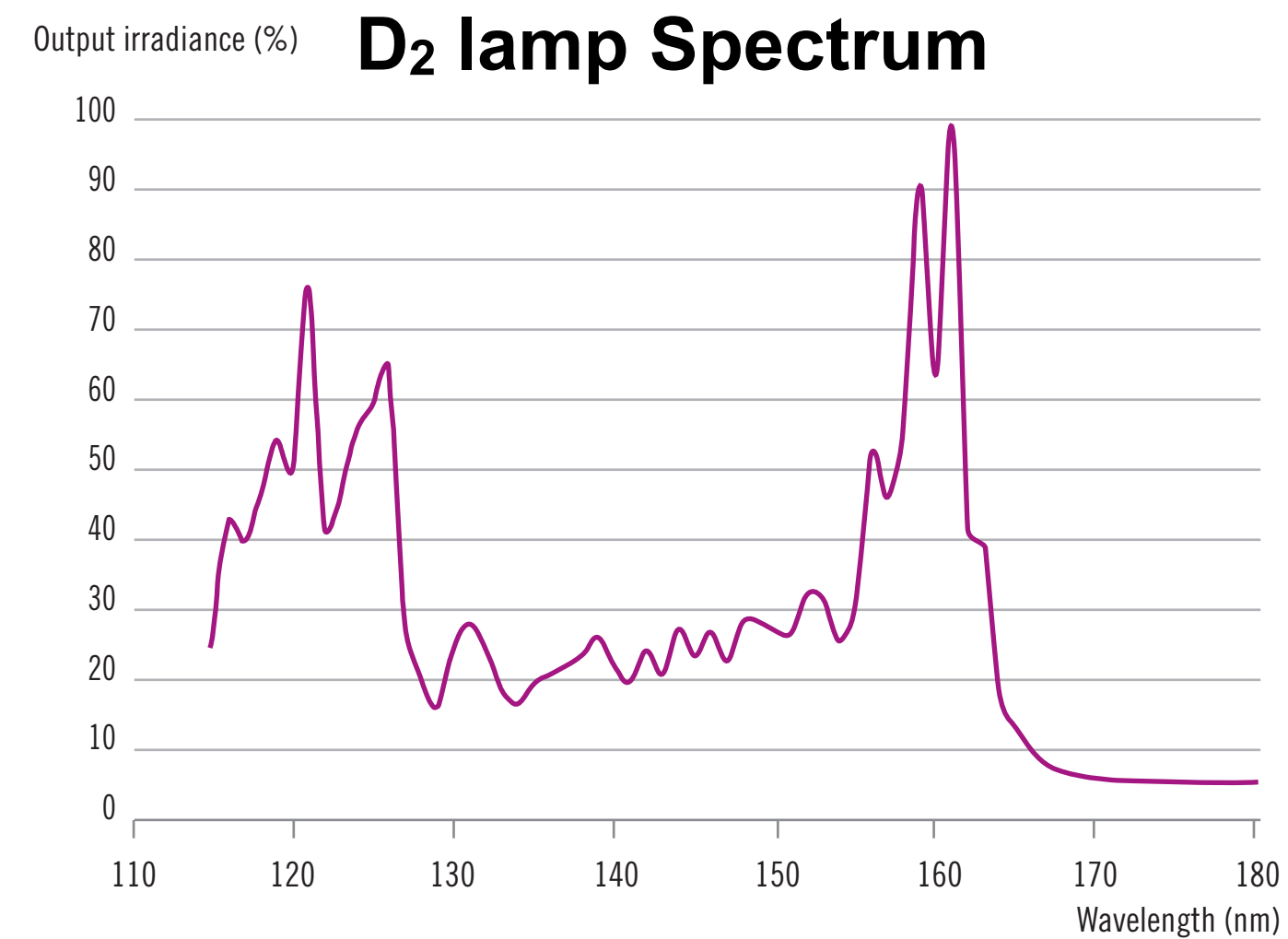
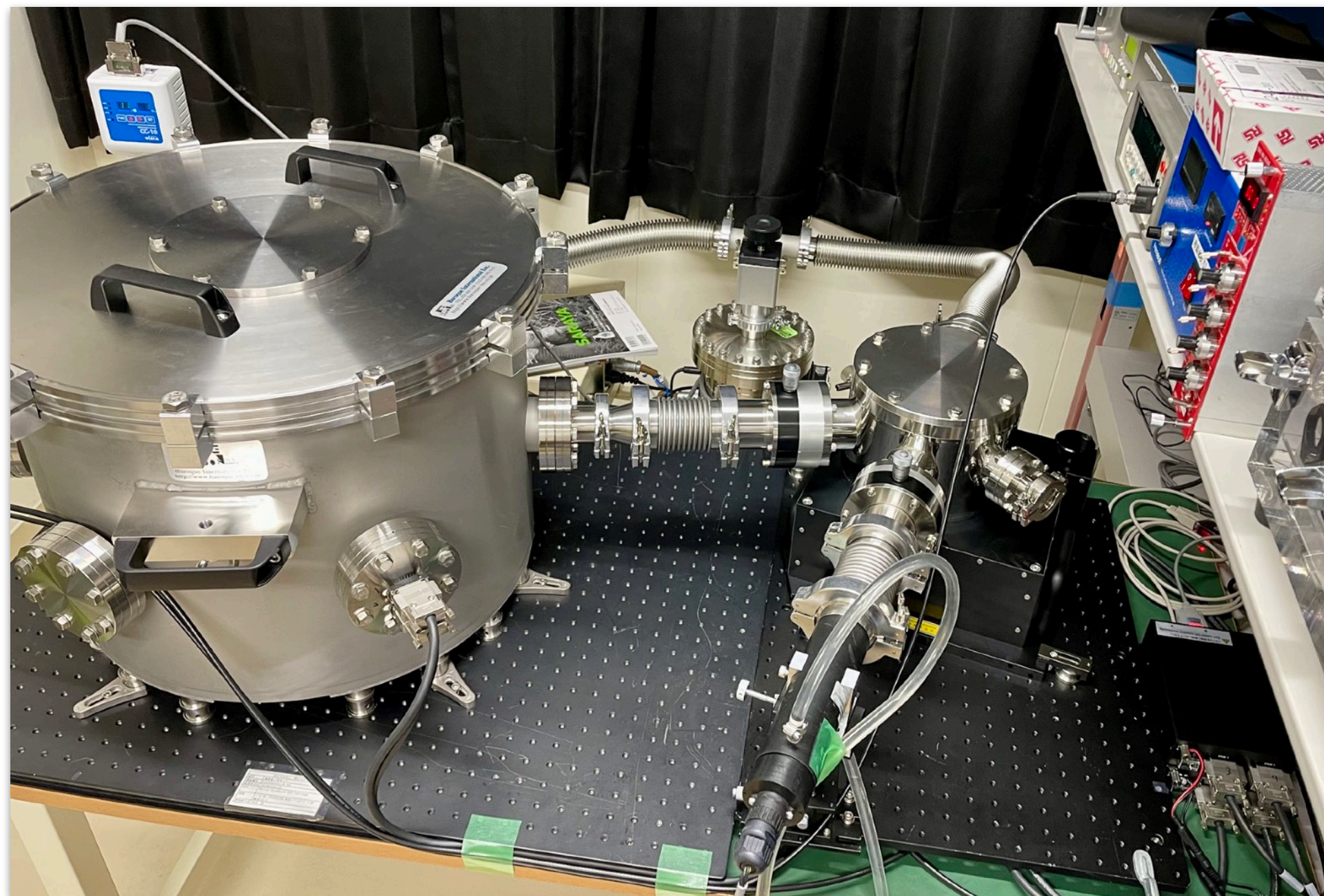
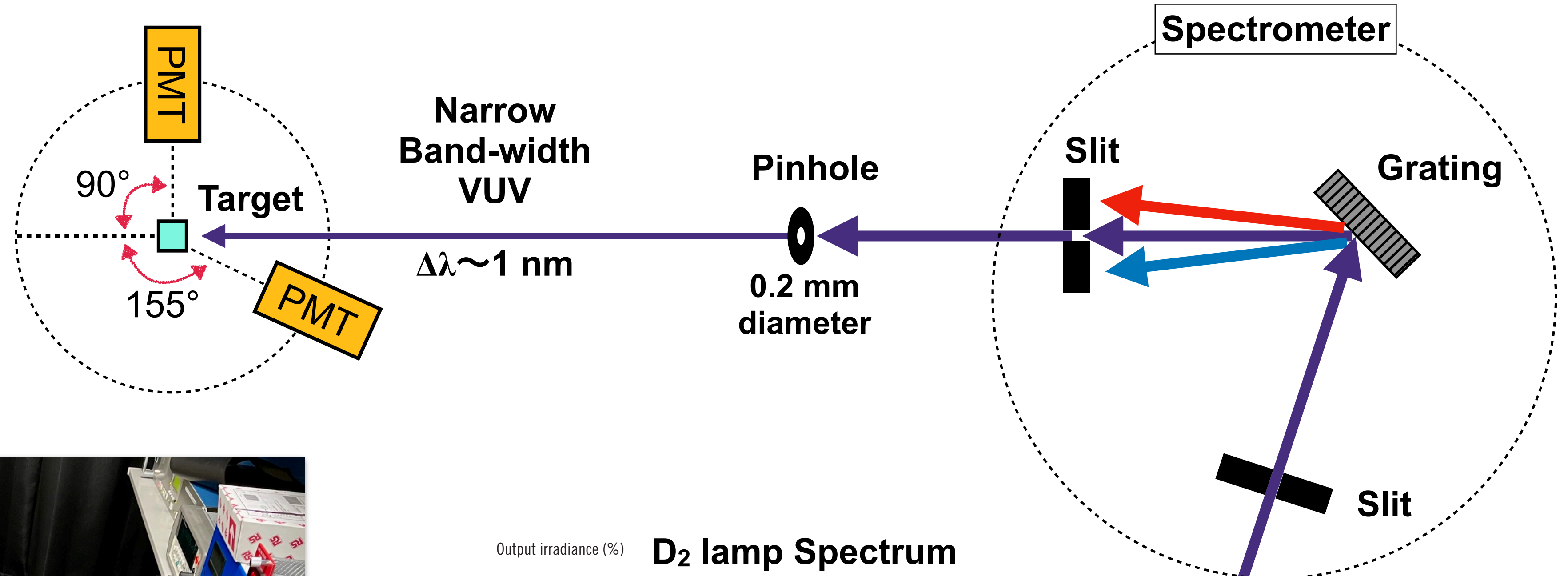
These are necessary to evaluate the transition rate

Measurement system has been developed

Measurement System of Optical Components



Stages in the cylindrical chamber



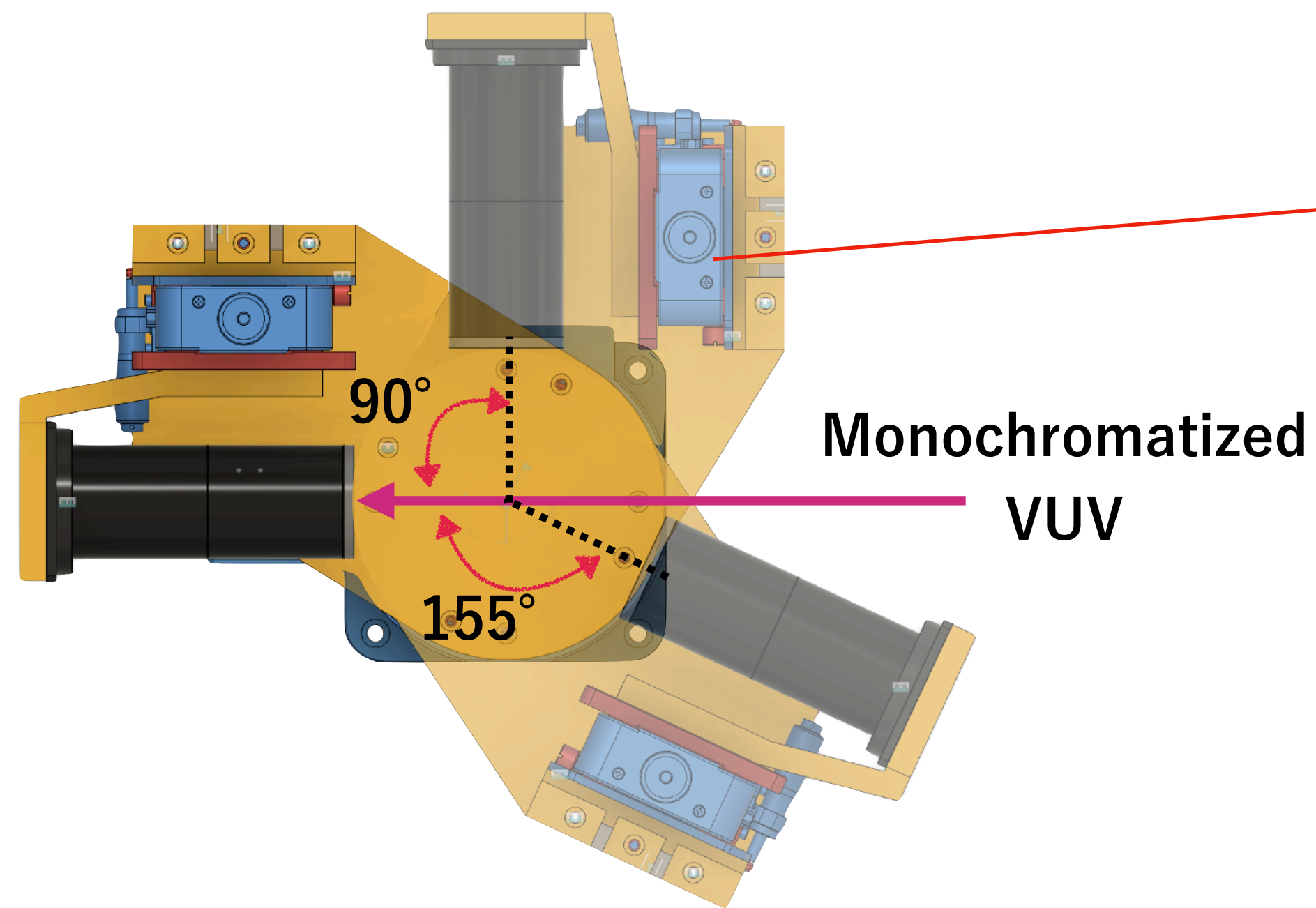
D200F-HV, Heraeus

D₂ lamp
Effective Region:
115 nm - 300 nm

Motorized Stages

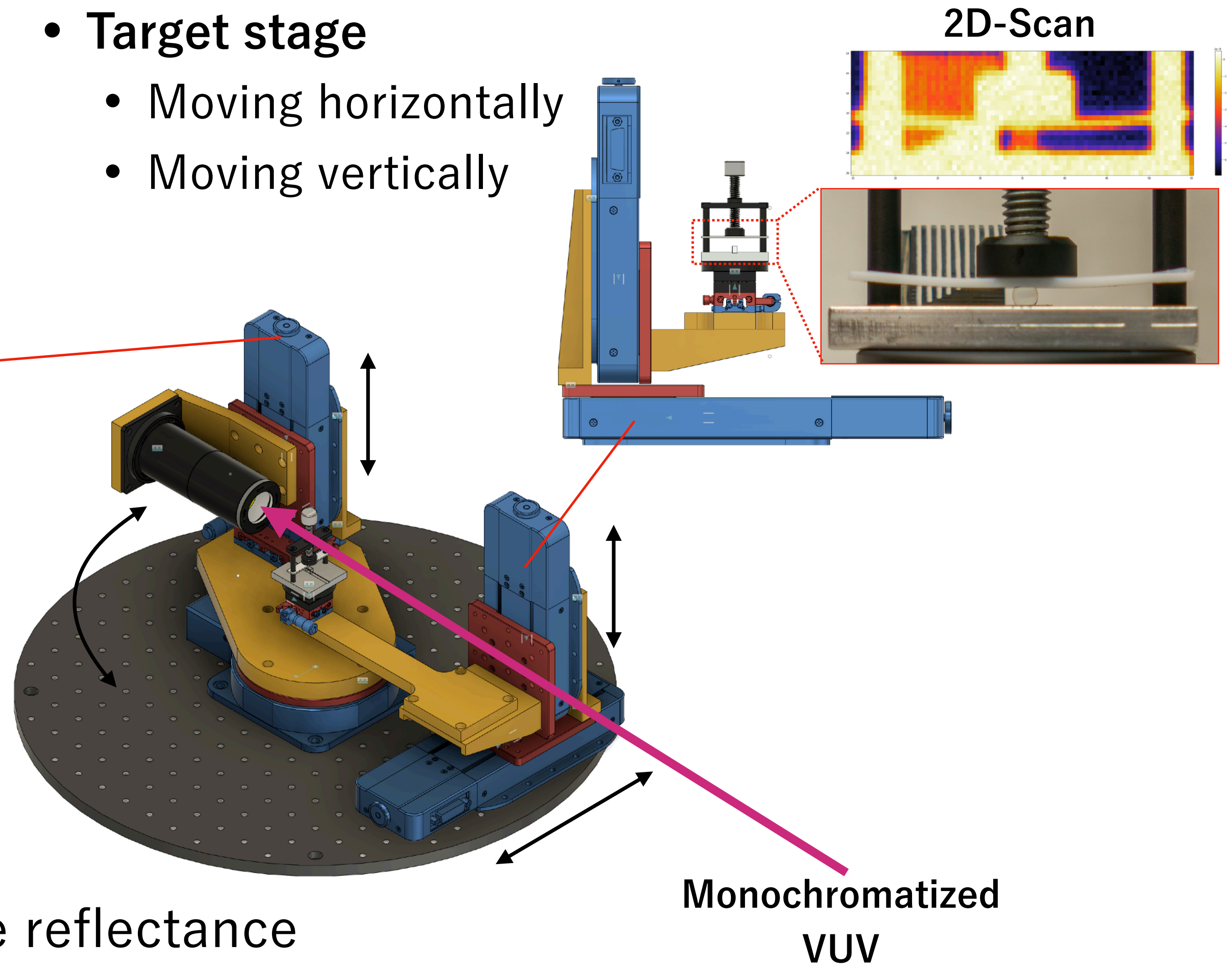
- **PMT stage**

- Rotating around center
- Moving vertically



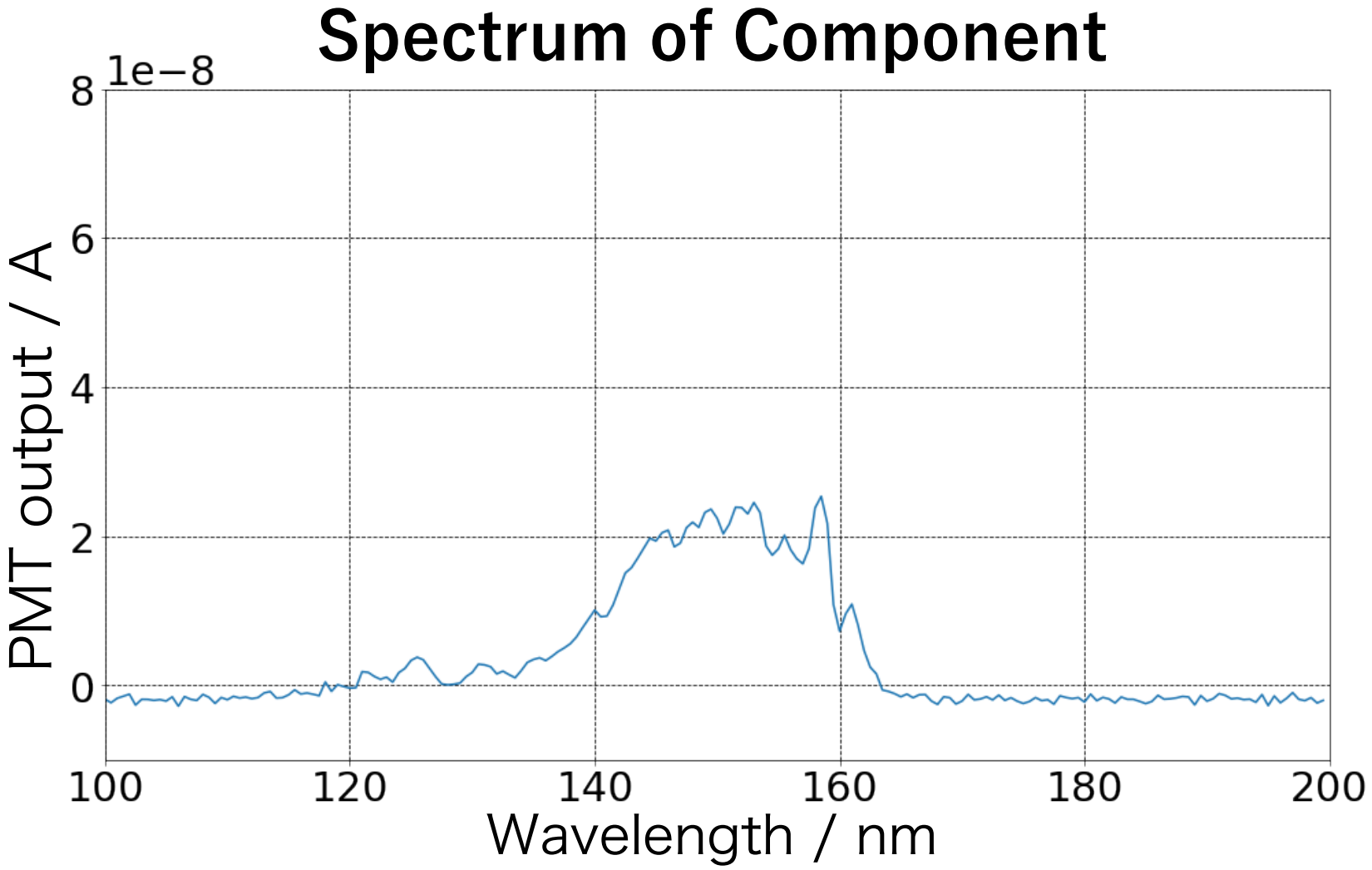
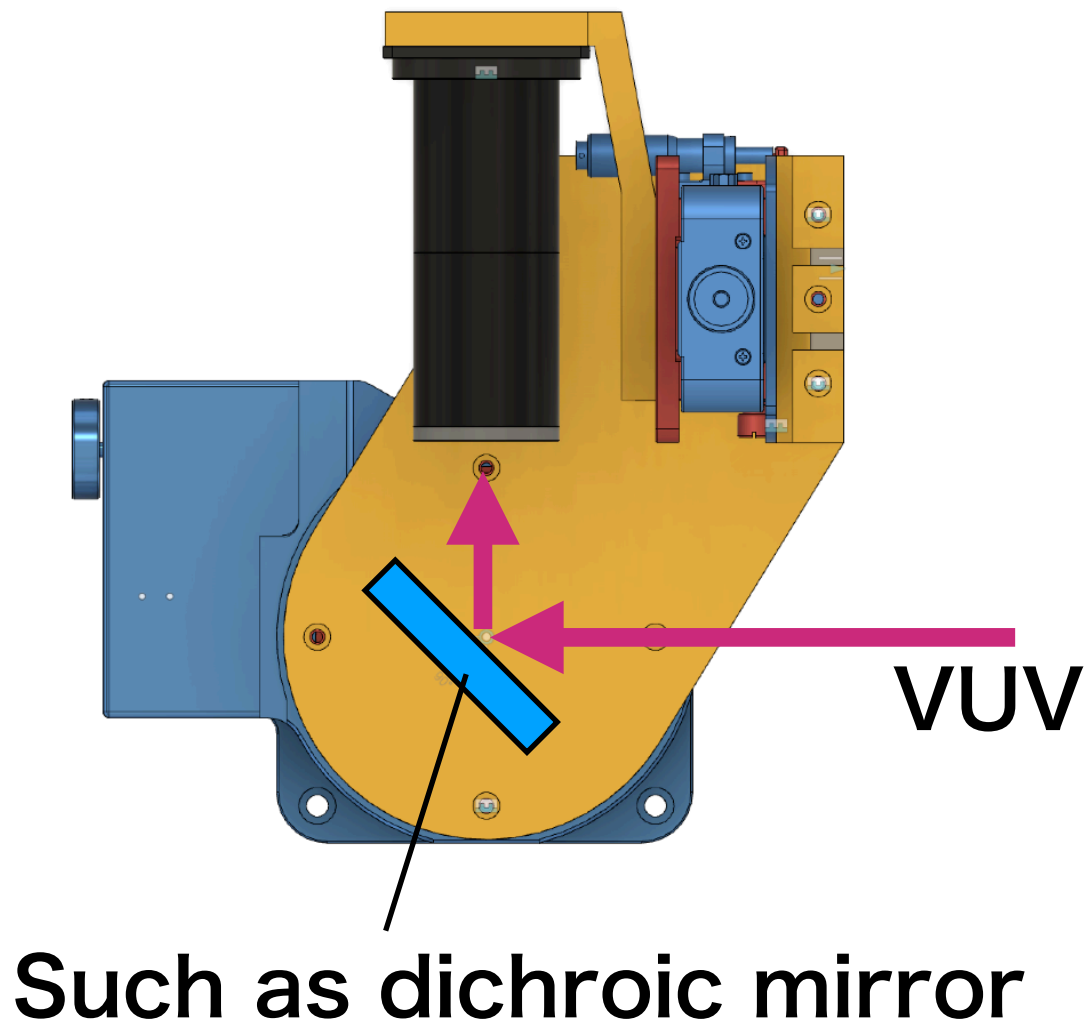
- **Target stage**

- Moving horizontally
- Moving vertically

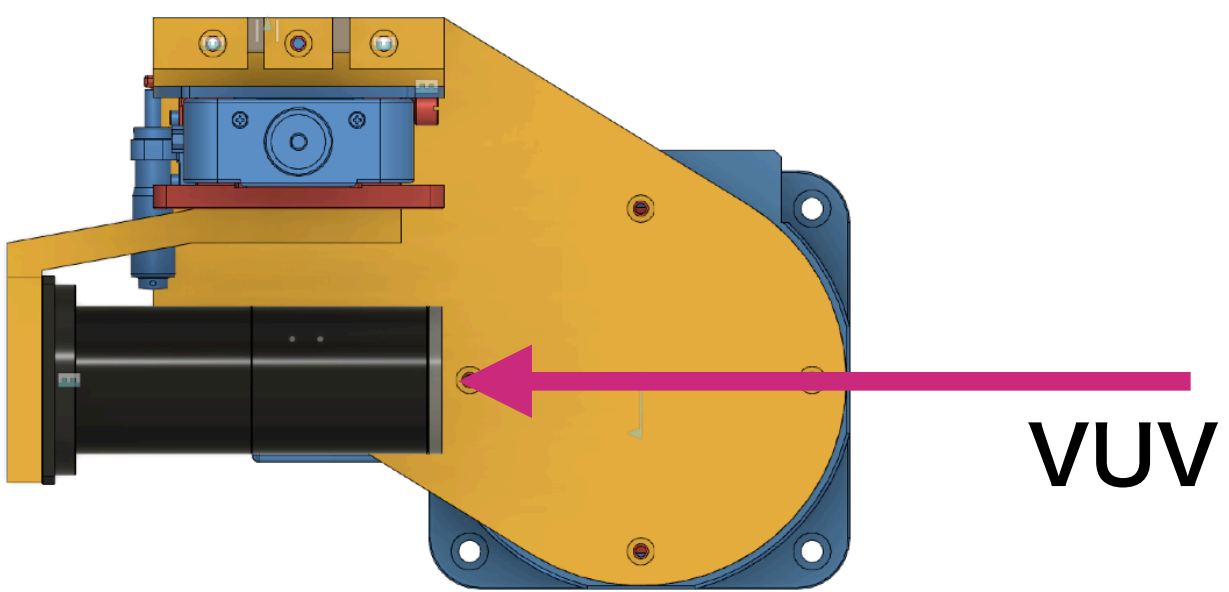
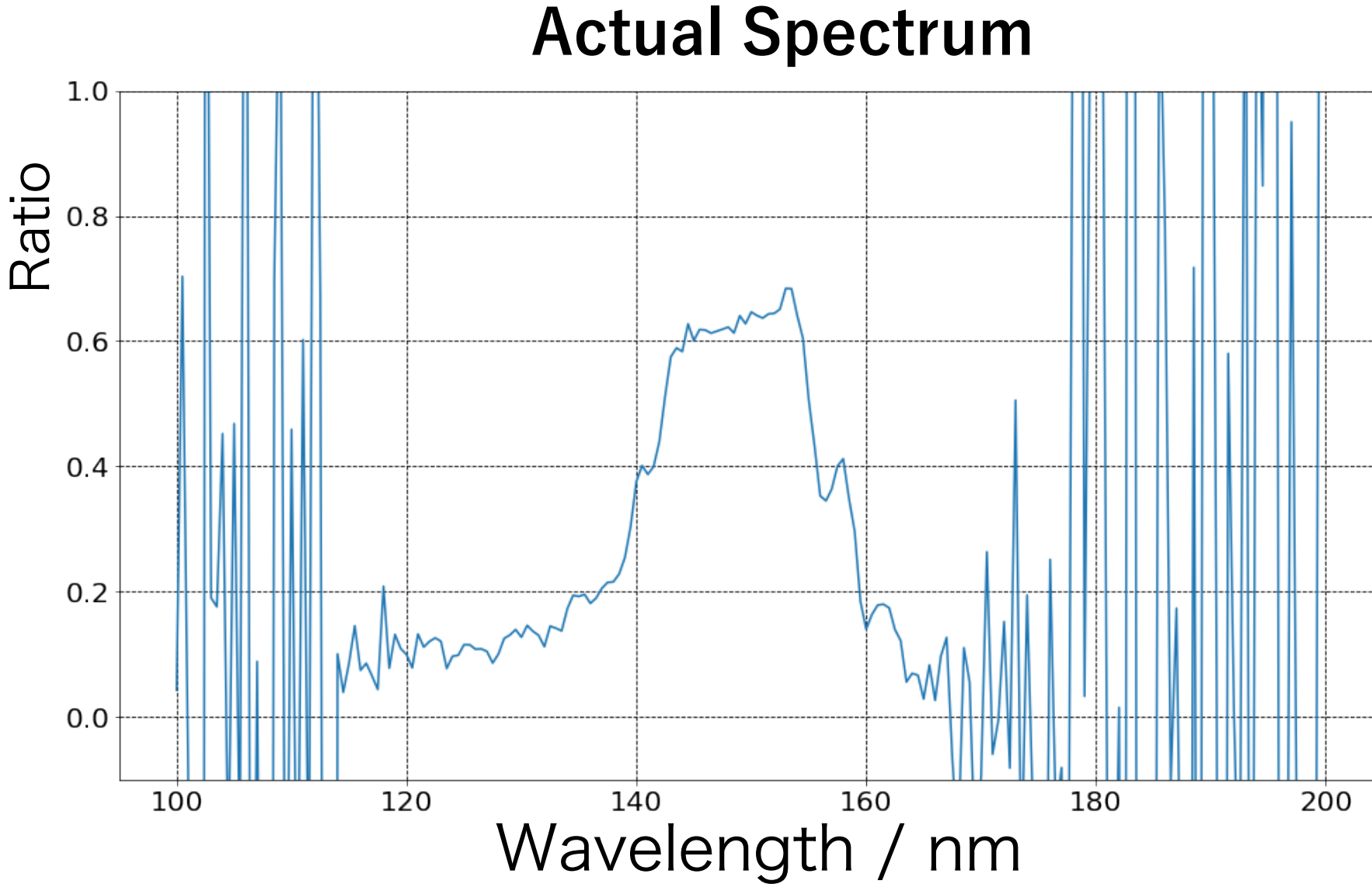
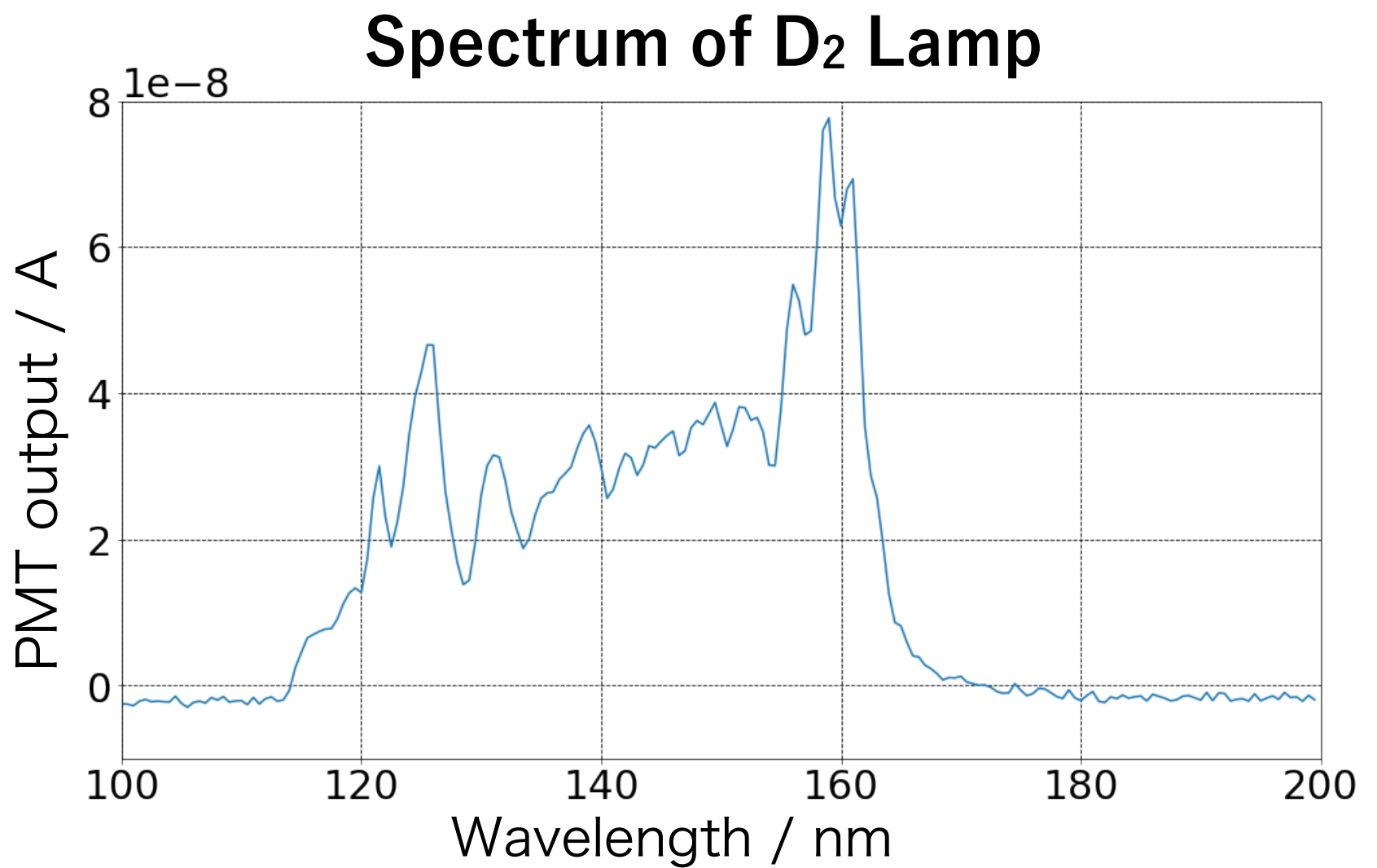


- Rotary stage makes us can measure reflectance and transmittance by rotating PMT around a target

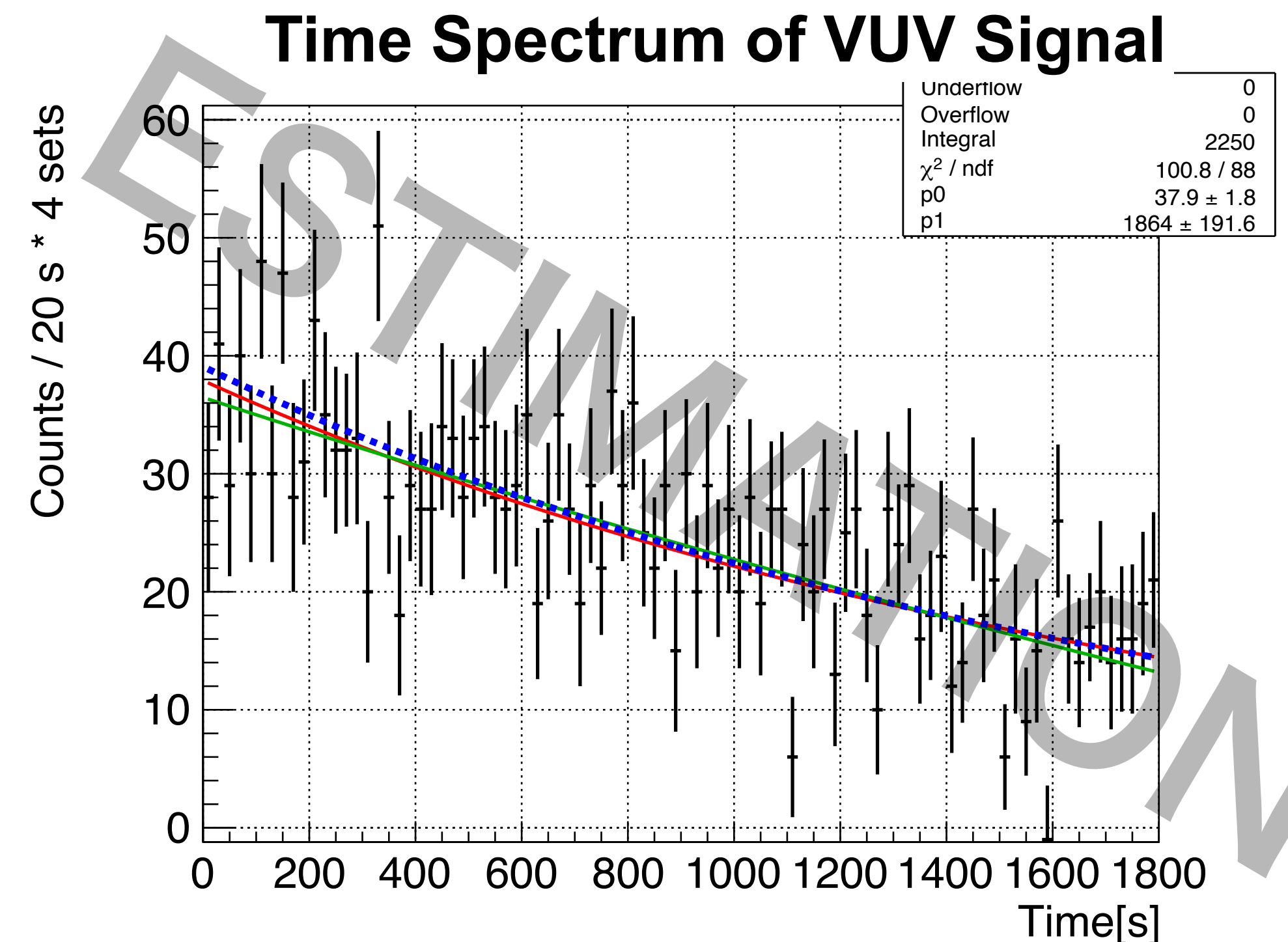
How to Get Spectra?



$$\div =$$



- Monte Carlo estimation of signal rate
 - Time depend rate : $R_{\text{detection}}(t) = f R_{\text{isomer}} [1 - \exp(-T/\tau)] \exp(-t/\tau)$
 - τ : Lifetime of $^{229\text{m}}\text{Th}$
 - T : Irradiation time of X-ray beam from SPring-8
 - R_{isomer} : Production rate of $^{229\text{m}}\text{Th}$ (known by NRS measurement)
- Net efficiency : $f = \prod_i f_i$
 - Transmittance of the CaF_2 crystal (0.5?)
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(0.8 / 1 mirror @150nm)
 - Transmittance of MgF_2 lens (0.8)
 - Geometrical efficiency (0.0855)
 - Quantum efficiency of VUV PMT (0.23)



Summary

- $^{229\text{m}}\text{Th}$ is already produced artificially by irradiation with X-ray beam.
- We are trying to detect VUV signal from $^{229\text{m}}\text{Th}$.
 - Beam time on 2022 Jan. has done
 - We minutely analyze the data which are taken on last beam time.
- We developed characterization system of optical components and measured properties such as reflectance and transmittance spectra.
 - We'll evaluate absolute efficiency of our VUV search setup and make upper limit clear which we searched on last beam time.

Collaborators

20



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A. Fujieda, H. Hara, T. Hiraki, H. Kaino, T. Masuda, Y. Miyamoto, K. Okai, S. Okubo, N. Sasao, K. Suzuki, S. Uetake, A. Yoshimi, **K. Yoshimura (PI)**, M. Yoshimura



Quantum Metrology Laboratory: A. Yamaguchi
RIKEN Nuclear Chemistry Research Team: H. Haba, Y. Shigekawa, T. Yokokita
SPring-8 center: K. Tamasaku



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Institute for Integrated Radiation and Nuclear Science, Kyoto University

S. Kitao, M. Seto



Japan Synchrotron Radiation Research Institute

Y. Yoda




National Institute of Advanced Industrial Science and Technology

T. Watanabe



Institute for Atomic and Subatomic Physics, TU Wien

K. Beeks, T. Schumm

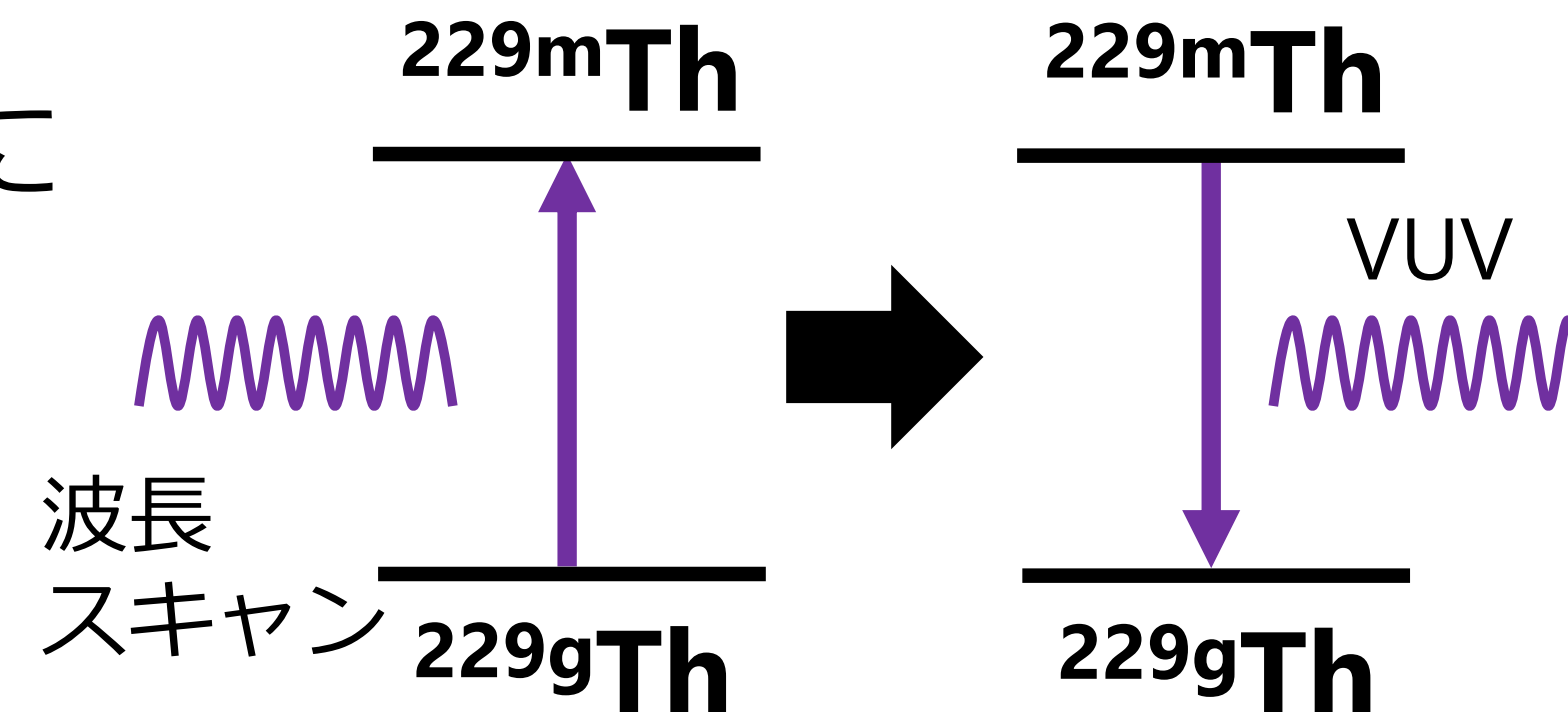


Back up

- 直接第一励起状態 ($^{229\text{m}}\text{Th}$) に励起する実験
- 脱励起光子は観測されず

J. Jeet *et al.*, Phys. Rev. Lett. **114**, 253001 (2015)

A. Yamaguchi *et al.*, New J. Phys. **17**, 053053 (2015)



- ^{233}U の α 崩壊生成される ^{229}Th 原子核励起状態の γ 線分光

$$E_{\text{isomer}} = 8.10 \pm 0.17 \text{ eV} \quad \text{T. Sikorsky *et al.*, Phys. Rev. Lett. **125**, 142503 (2020)}$$

- $^{229\text{m}}\text{Th}$ 内部転換過程 (IC) の測定

$$E_{\text{isomer}} = 8.28 \pm 0.17 \text{ eV}$$

$$T_{1/2} (\text{IC}) = 7 \pm 1 \mu\text{s}$$

$$T_{1/2} (^{229\text{m}}\text{Th}^{2+}) > \sim 60 \text{ s}$$

L.v.d. Wense *et al.*, Nature **533**, 47 (2016)

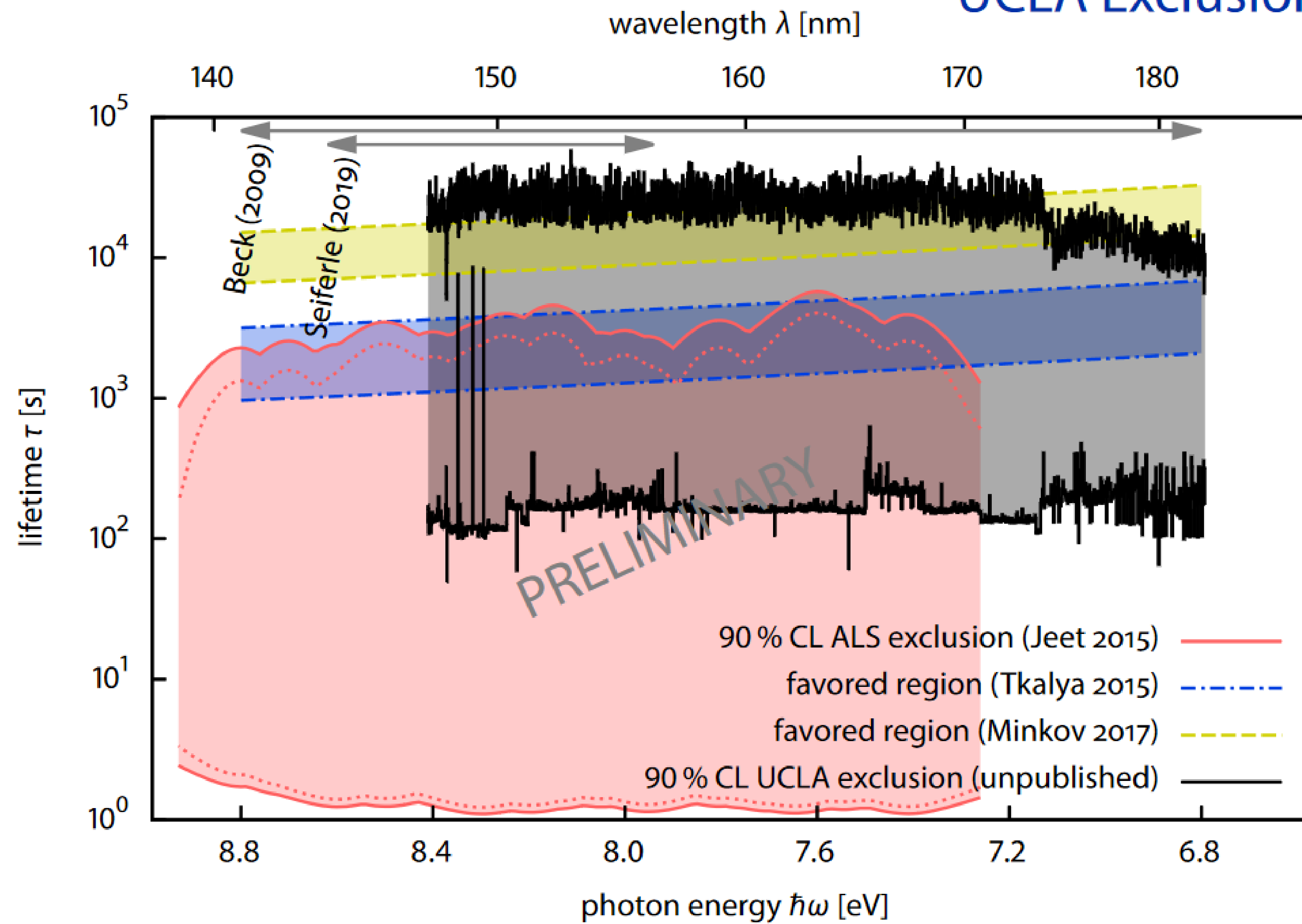
B. Seiferle, L.v.d. Wense and P.G. Thirolf, Phys. Rev. Lett. **118**, 042501 (2017)

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

- $^{229\text{m}}\text{Th}$ からの脱励起光の観測に成功した例はない
- $^{229\text{m}}\text{Th}$ の光学遷移寿命の測定例もない
 - 理論予想だと $O(10^2 - 10^4) \text{ s}$

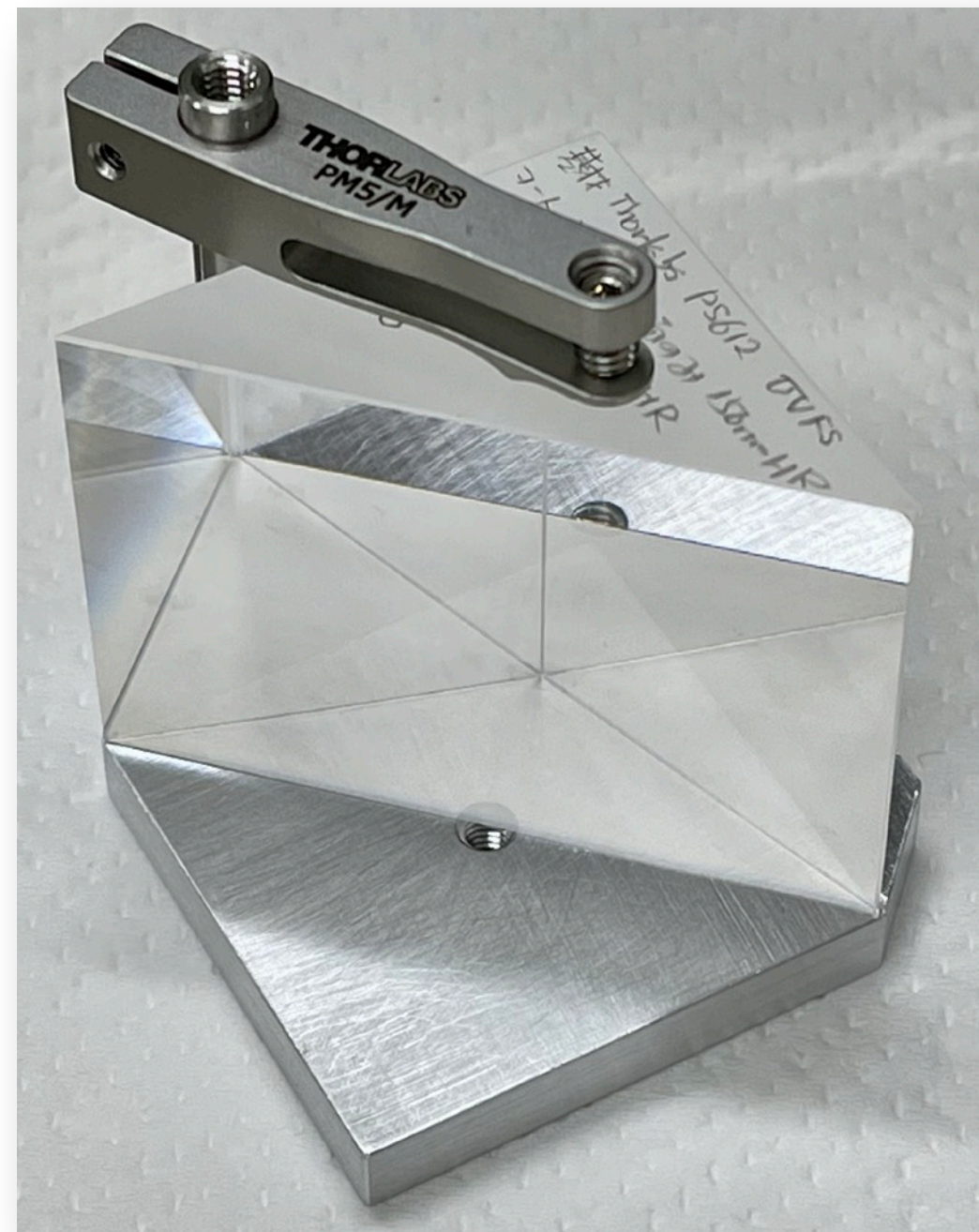
Excluded Region

UCLA Exclusion Region

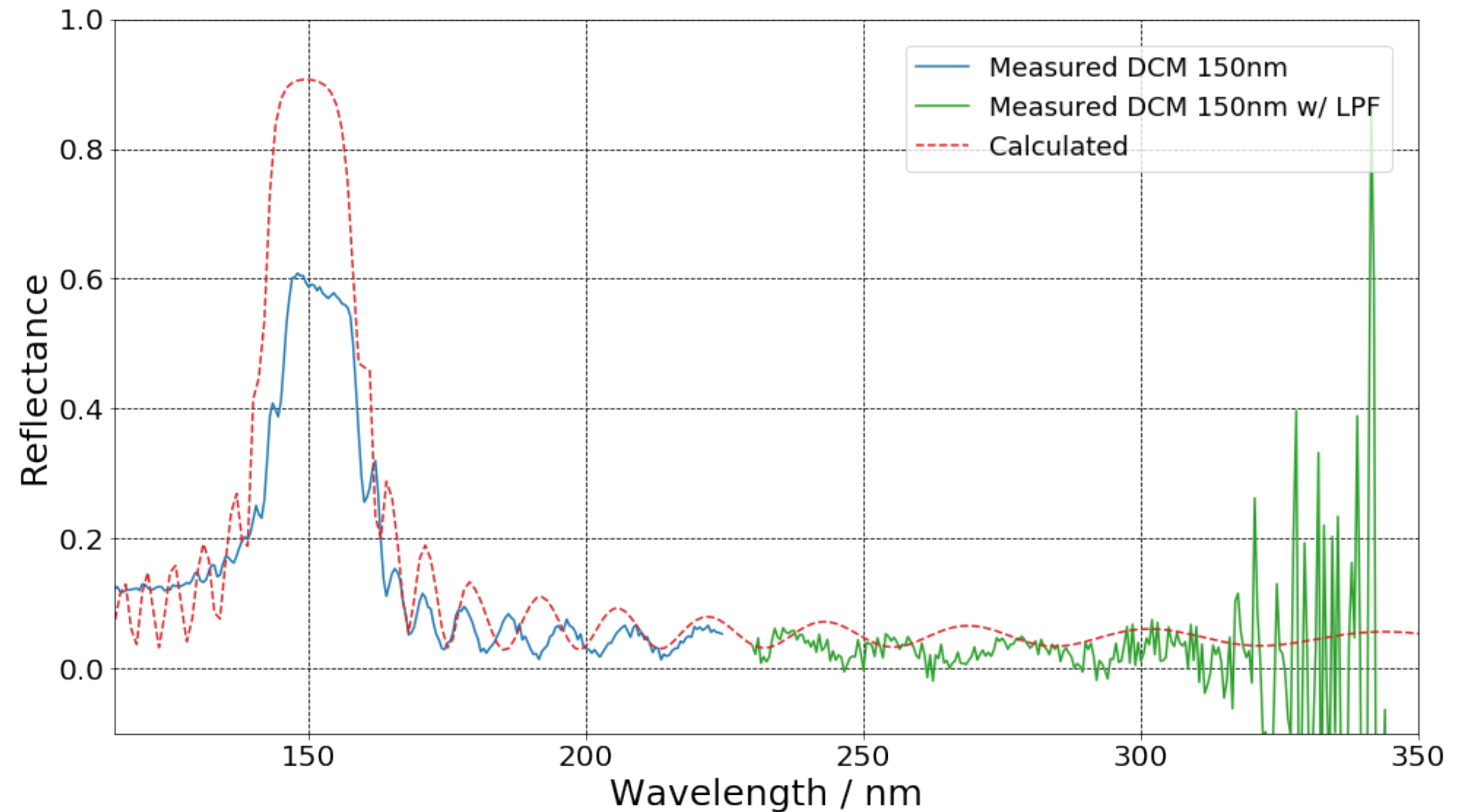


- **Optical setup must be put in the vacuum (< 0.1 Pa)**
 - VUV light can't pass through in the air because absorption by oxygen
 - Absorption coefficient is worse than 10 um^{-1} in the air
- **Transmittance and reflectance can't easily be estimated**
 - because it is deeply affected by surface condition and crystal purity
- **We have developed system to do that and actually measured it.**
 - We must make the estimate of net efficiency of our setup which aims to VUV search
 - It's necessary to directly measure all components which be included in our setup

Measurement Example

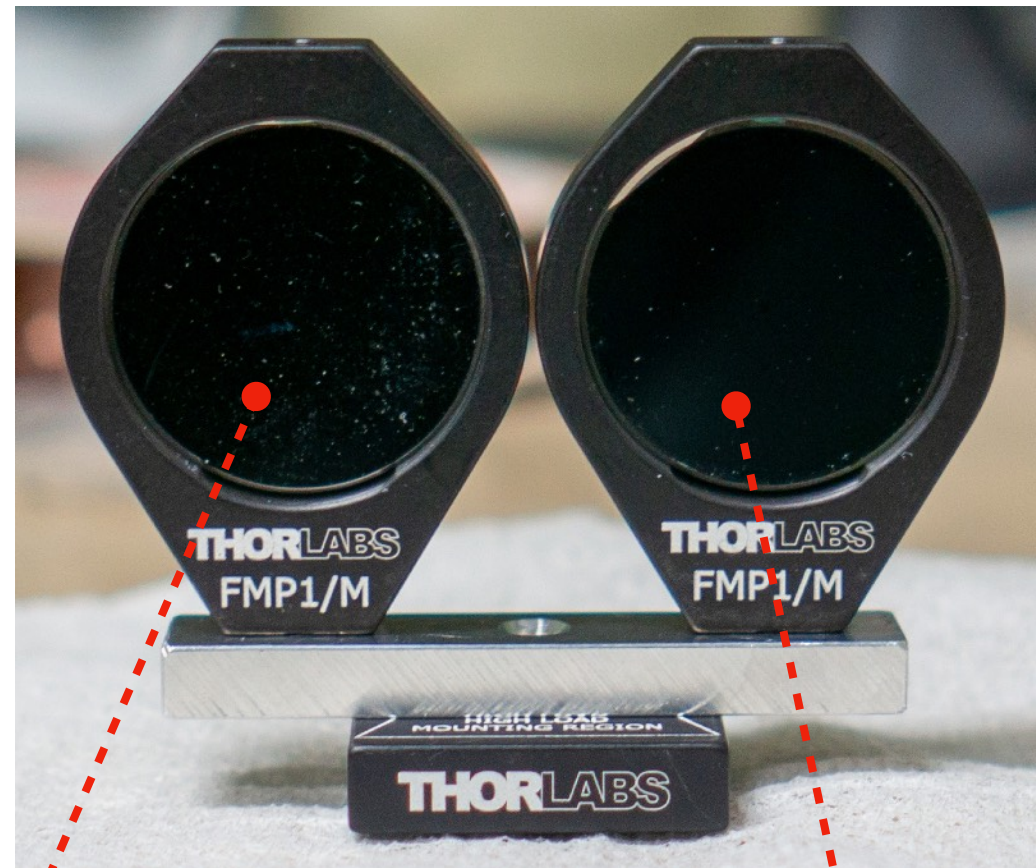


Custom-made mirror
(Dichroic 150 nm)



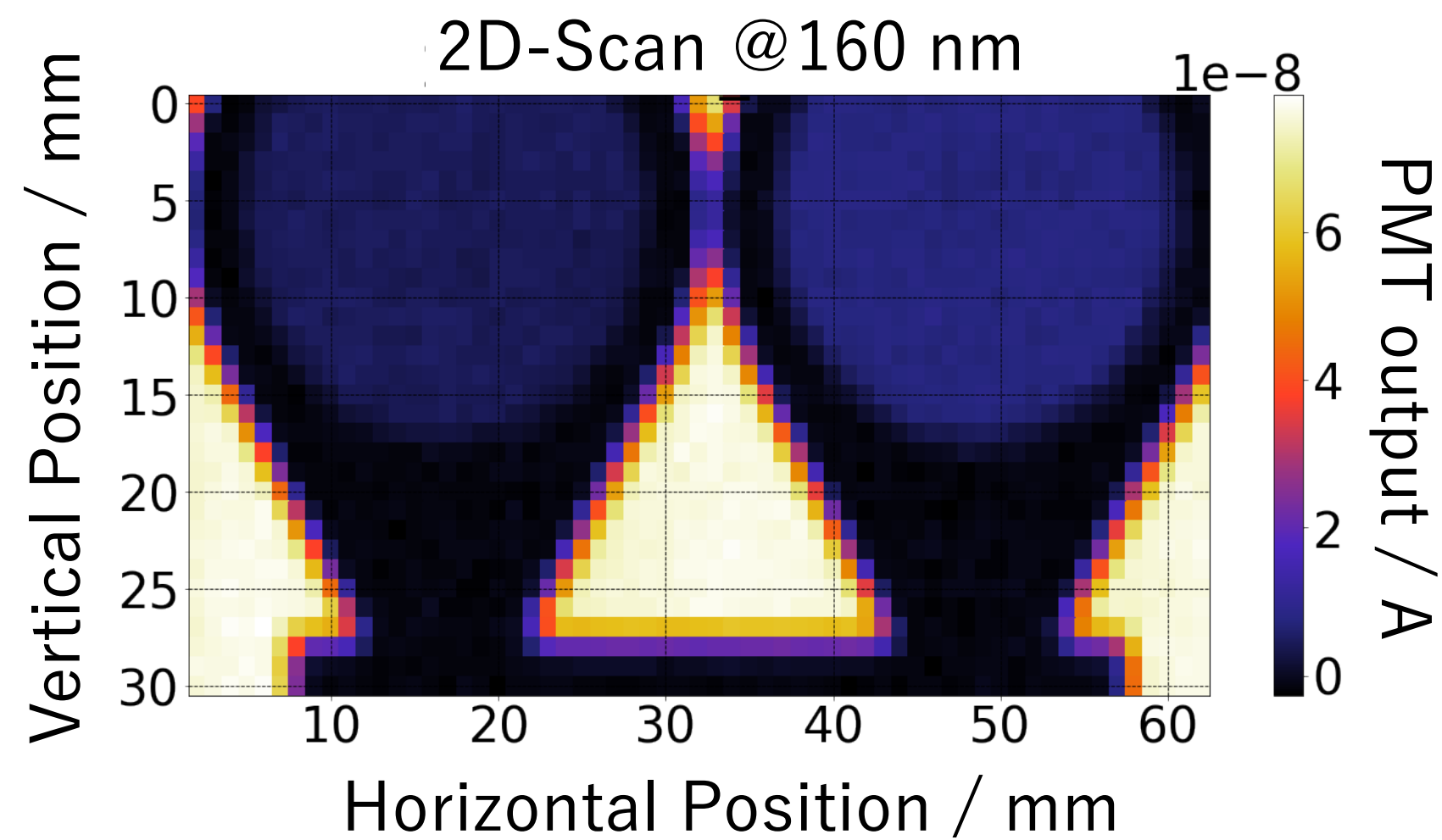
- There is discrepancy between calculated spectra and measured one.
- We can remove uncertainty of Optical components by using such system.

Band Pass Filters



147 nm

150 nm



Filter Transmittance Spectrum

