

Realization of the active pumping of the Thorium-229 isomeric state

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Takahiko Masuda on behalf of the thorium group



Research Institute for Interdisciplinary Science,
Okayama Univ. (JAPAN)



X-ray pumping of the ²²⁹Th nuclear clock isomer

Takahiko Masuda¹, Akihiro Yoshimi¹, Akira Fujieda¹, Hiroyuki Fujimoto¹, Hideaki Hara¹, Takahiro Hiraki¹, Hiroyuki Kaino¹, Yoshitaka Kasamatsu¹, Kenji Konashi⁶, Yuki Miyamoto¹, Koichi Okai¹, Sho Okubo¹, Noboru Sasakawa¹, Thorsten Schumm⁷, Yudai Shigekawa⁴, Kenta Suzuki¹, Simon Stellmer^{7†}, Satoshi Uetake¹, Makoto Watanabe⁶, Tsukasa Watanabe², Yuki Yasuda⁴, Yoshitaka Yoda⁹, Takuya Yokokita³, Motohiko Yoshimura¹, Koji

¹Research Institute for Interdisciplinary Science, Okayama University, Okayama 700-8530, Japan
²National Institute of Advanced Industrial Science and Technology (AIST), Tsukuba, Ibaraki 305-8563, Japan
³RIKEN, 2-1 Hirosawa, Wako, Saitama 351-0198
⁴Graduate School of Science, Osaka University, Toyonaka, Osaka 560-0043, Japan
⁵Institute for Integrated Radiation and Nuclear Science, Kyoto University, Sennan-gun, Osaka 590-0494, Japan
⁶Institute for Materials Research, Tohoku University, Higashiibaraki-gun, Ibaraki 981-8585, Japan
⁷Institute for Atomic and Subatomic Physics, TU Wien, 1020 Vienna, Austria
⁸RIKEN SPring-8 Center, 1-1-1 Kouto, Sayo-cho, Sayo-gun, Hyogo, 679-5198, Japan
⁹Japan Synchrotron Radiation Research Institute, 1-1-1 Kouto, Sayo-gun, Hyogo, 679-5198, Japan

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Abstract

Thorium-229 is a unique case in nuclear physics: it presents a metastable first excited electronvolts above the nuclear ground state. This so-called isomer is accessible by VUV lasers and the amazing precision of atomic laser spectroscopy to nuclear physics. Being able to manipulate the isomer at will opens up a multitude of prospects, from studies of the fundamental interactions in the nucleus to a compact and robust nuclear clock. However, direct optical excitation of the isomer or its relaxation to the ground state has not yet been observed, and a series of key nuclear structure parameters such as the half-lives of the low-lying nuclear levels of ²²⁹Th are yet unknown. Here we present the first active pumping of the ^{229m}Th isomer. Our scheme employs narrow-band 29 keV synchrotron radiation to resonantly excite the isomer, which then predominantly decays into the isomer. We determine the resonance energy with 0.1 eV, the half-life of 82.2 ps, an excitation linewidth of 1.70 neV, and extract the branching ratio of the isomer to the ground and isomeric state respectively. These measurements allow us to re-evaluate gamma-ray data that have been collected over 40 years.

1 Introduction

The first excited nuclear state of ²²⁹Th is known to be an isomeric state ^{229m}Th (metastable excited state). It is the lowest nuclear excitation energy known in nature, and it is the lowest nuclear excitation energy known in nature.

it is the lowest nuclear excitation energy known in nature. Although the optical excitation of the isomer to ^{229m}Th is yet to be established, investigations for exploiting it as a new nuclear clock are expanding.

トピックス

放射光で探るレーザー光による超精密原子核制御の可能性：²²⁹Th 極低核励起準位

吉見彰洋
岡山大学 異分野基礎科学研究所 〒700-8530 岡山市北区津島中 3-1-1
笠松良崇
大阪大学 大学院理学研究科 〒560-0043 大阪府豊中市侍兼山1-1
北尾真司
京都大学 複合原子力科学研究所 〒590-0494 大阪府泉南郡熊野町1-1
瀬戸 誠
京都大学 複合原子力科学研究所 〒590-0494 大阪府泉南郡熊野町1-1
増田孝彦
岡山大学 異分野基礎科学研究所 〒700-8530 岡山市北区津島中 3-1-1
山口敦史
国立研究開発法人理化学研究所 〒351-0198 埼玉県和光市広沢
依田芳卓
公益財団法人高輝度光科学研究センター 〒679-5198 兵庫県佐用町1-1
吉村浩司
岡山大学 異分野基礎科学研究所 〒700-8530 岡山市北区津島中 3-1-1



要旨 原子核としては異常に低いエネルギー（eV オーダー）の励起準位を持ち、レーザーによる核状態操作が可能で超精密原子核時計等への応用が期待されているアイソトープ ²²⁹Th に近年注目が集まっている。我々は、この研究を展開するのに欠かせない低エネルギー準位のエネルギー・寿命の決定のために、高輝度放射光 X 線と高性能検出器・標的システムを組み合わせた核共鳴散乱実験を行っている。高度化した核共鳴測定システムの開発、それを用いた短寿命準位（²⁰¹Hg の 26 keV 準位）の高精度核共鳴実験の結果、および ²²⁹Th の核共鳴実験の現状について紹介する。

1. はじめに

原子核は陽子と中性子（総称して核子）が強い力で束縛された系であり、個々の核子はそれぞれがお互いに作る核力ポテンシャルの中で形成される軌道を運動している。また個々の核子の運動だけでなく、原子核全体の回転や振動という集団運動に起因するスペクトルが観測されるという大きな特徴がある。原子物理における軌道電子とは違い、原子核を励起する、つまり核子をより高いエネルギー軌道に上げたり回転・振動モードを励起するには通常 keV から MeV 程度のエネルギーが必要である。このため、量子エレクトロニクス分野で見られるようなレーザー光を利用した高度な量子操作は、原子核に対しては一般的に不可能である。一方で、原子核の量子状態は原子のそれに比べて極めて壊れにくい、外界から隔離された良い量子力学系で

あるという魅力がある。レーザー光を用いた原子核に対する量子エレクトロニクスの実現は新たな科学分野の開拓につながるかと期待されている。このような理由で近年大きく興味を持たれている原子核が ²²⁹Th（自然存在比 0%, 半減期 7880 年）である。²²⁹Th はその第一励起状態が原子核としては異常に低く、レーザー光でアクセス可能な eV 領域にあることが分かっていた。このことは 1970 年代から指摘されていたが¹⁾、このあまりにも低い励起状態を特定することは困難を極め、それがはっきりしてきたのは 2000 年代に入ってからである。2007 年に複数の γ 線測定エネルギーを足し引きすることで、間接的にこの状態の励起エネルギーが 7.8 ± 0.5 eV であるという報告²⁾ がなされ、最近ではその励起エネルギーは確かに 6.3~18.3 eV の間に存在することが実験的に確かめられた³⁾（Fig. 1）。しかしこのエネルギーを直接的に



Division of Quantum Universe, RIIS, Okayama University

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



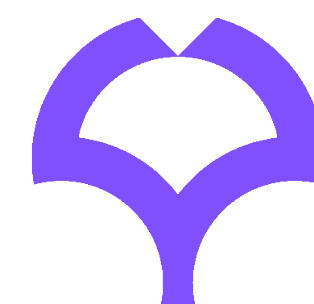
SPRING-8, JASRI
Y. Yoda



IMR, Tohoku Univ.
K. Konashi, M. Watanabe



IIRNS, Kyoto Univ.
S. Kitao, M. Seto,



Lab. of Radio Chemistry, Osaka Univ.
Y. Kasamatsu, Y. Shigekawa, Y. Yasuda



Quantum Metrology Lab., RIKEN
A. Yamaguchi
Nishina center, RIKEN
H. Haba, T. Yokokita
SPRING-8 center, RIKEN
K. Tamasaku



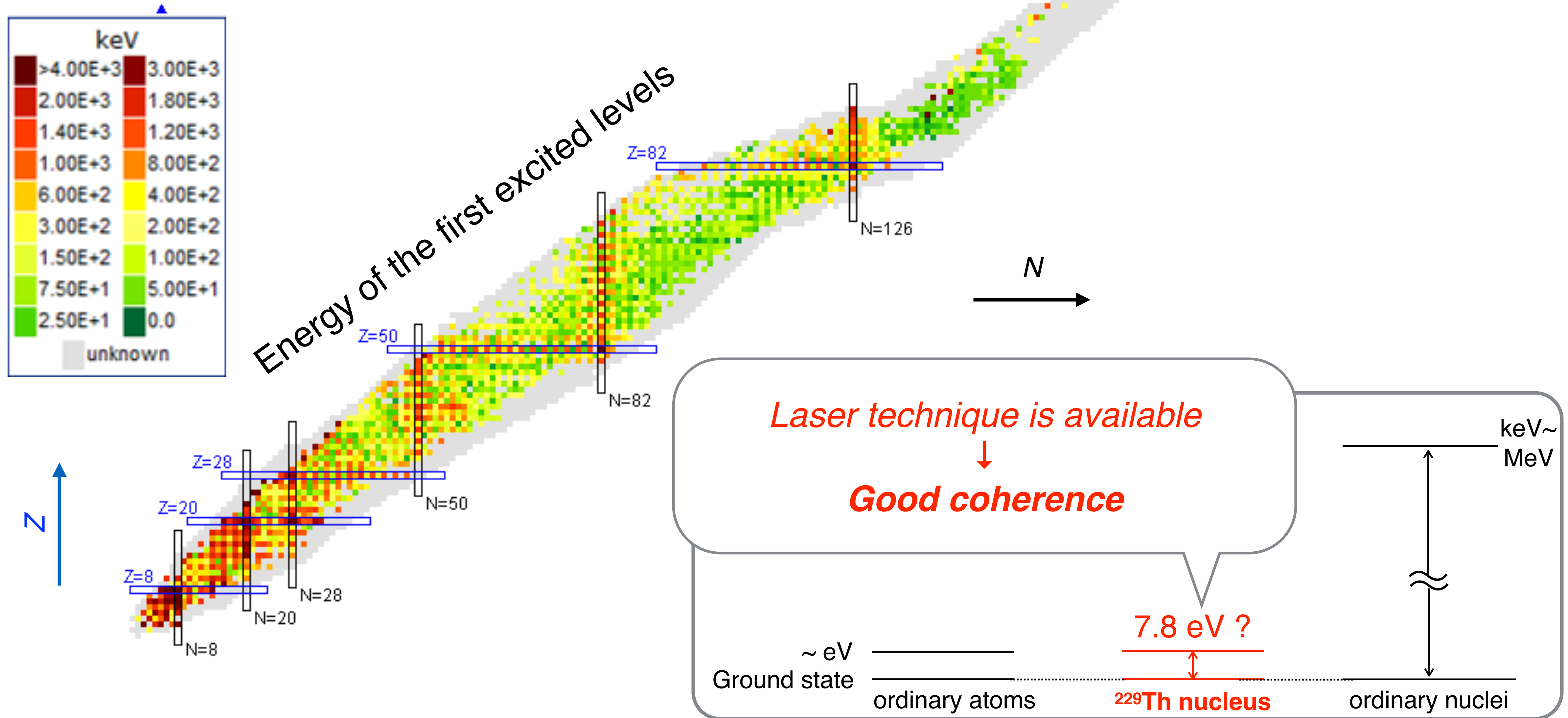
Dimensional standards group, AIST
H. Fujimoto, T. Watanabe



Institute for Atomic and Subatomic
Physics, TU WIEN
T. Schumm, S. Stellmer

8 institutes, ~ 30 collaborators

Extraordinary low-excitation energy among all known nuclei.



Frequency standard “Nuclear Clock”

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

- Current definition of second (based on Cs atomic clock) : Uncertainty $\sim 10^{-16}$
- ^{229}Th nuclear clock can improve the uncertainty to 10^{-19} even in a solid-state.
→ **Realize more accurate (& portable) clock**

→ Measurement of the temporal variation of the Fine Structure Constant

- Current limit (based on Yb atomic clock stability) : $\frac{d\alpha/dt}{\alpha} = (-2.0 \pm 2.0) \times 10^{-17} / \text{year}$
N. Hntemann et al., *Phys. Rev. Lett.* **113**, 210802 (2014).
- ^{229}Th nucleus is the most sensitive candidate. **Several orders! improvement is expected.**
V.V. Flambaum, *Phys. Rev. Lett.* **97**, 092502 (2006).
J.C. Berengut et al., *Phys. Rev. Lett.* **102**, 210801 (2009).

→ Dark matter search

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

- Topological defect dark matter

→ Heavy rare metal search

- Relativistic geodesy

***10⁻²²/year can be possible.
It can constrain parameters in BSMs
which relate “cosmic acceleration”***

History & situation of the ^{229}Th isomer ($^{229\text{m}}\text{Th}$)

year		result
1976	Kroger & Reich <i>Nucl. Phys. A</i> 259 (1976) 29. U-233 α -decay γ -spectroscopy	$< 100 \text{ eV}$
1994-	Helmer and Reich, Many searches...	$3.5 \pm 1.0 \text{ eV}$
2007	Beck et al. <i>Phys. Rev. Lett.</i> 98 (2007) 142501; LLNL-PROC-415170 (2009). γ -spectroscopy by microcalorimeter (indirect measurement)	$7.8 \pm 0.5 \text{ eV}$
2015	Jeet et al. <i>Phys. Rev. Lett.</i> 114 (2015) 253001. Direct VUV excitation & emission. $\rightarrow$ Null result	Reject : $7.3\text{-}8.8 \text{ eV}$ (τ : $1\text{-}5600 \text{ s}$)
2016	Wense et al. <i>Nature</i> 533 (2016) 47. Observation of IC electron from the isomer state (from ^{233}U α -decay)	$6.3\text{-}18.3 \text{ eV}$
2017	Seiferle et al. <i>Phys. Rev. Lett.</i> 118 (2017) 042501. Lifetime measurement of IC electrons (from ^{233}U α -decay)	$\tau_{\text{IC}} = 7 \pm 1 \text{ us}$ $\tau_{\gamma} \sim 10^4 \text{ s}$
2018	Thielking et al. <i>Nature</i> 556 (2018) 321. Laser spectroscopy of the isomer state (from ^{233}U α -decay)	HFS, M1, E2 of $^{229\text{m}}\text{Th}^{2+}$

× Precise energy is not known.

Gamma-ray spectroscopy	$7.8\pm0.5 \text{ eV}$
Direct excitation	$7.3 - 8.8 \text{ eV}$ excluded
Electron emission measurement	$6.3 - 18.3 \text{ eV}$

× Laser excitation & VUV photon emission have never been observed.

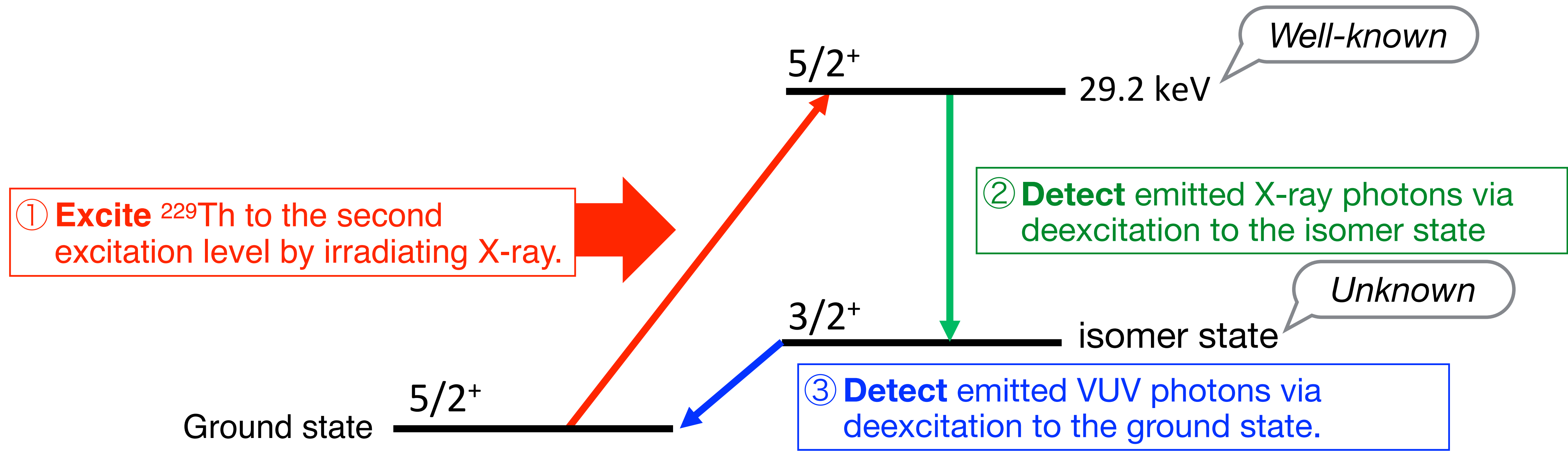
Introduction of our project

All the previous experiments used ^{233}U α -decay as a ^{229}Th isomer source.
But, α -decay is a violent process (huge background, > 80 keV recoil energy, ...),
and uncontrollable.

More **cleaner and controllable** isomer production method is preferred.

➡ *Optical active pumping to the isomer state*

Principle of the pumping

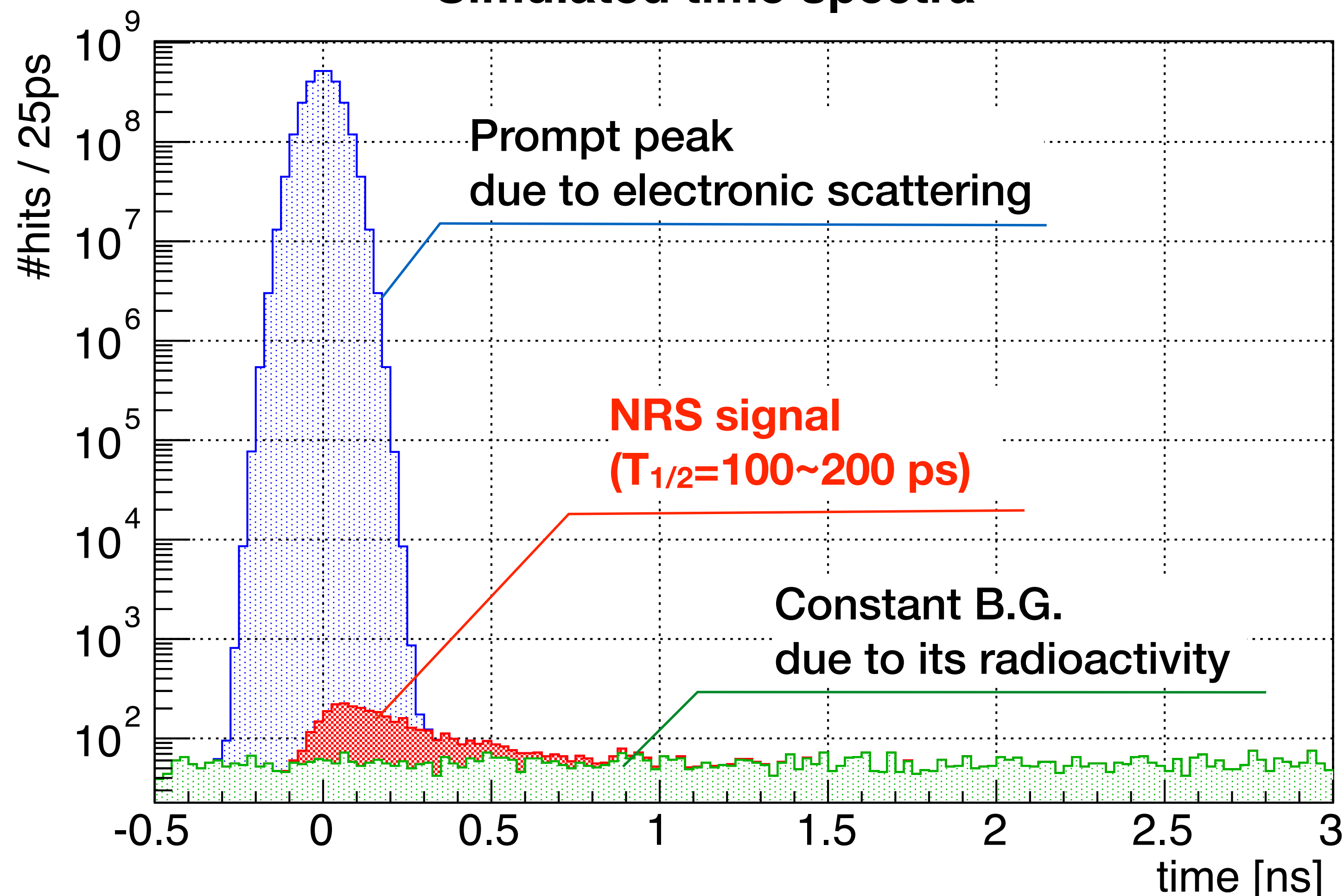


NRS (Nuclear resonant scattering)

By detecting **X-ray photon emission** in ②, we can confirm the isomer production.
Then, we simply wait for **the VUV photon emission** in ③ to measure its wavelength.

Key points for Th-229 NRS

Simulated time spectra



Difficulties *Poor SNR*

Short lifetime	$T_{1/2} = 100 - 200$ ps
Low S/N ratio	$10^{-6} - 10^{-7}$
Low Signal rate	< 1 cps
Constant B.G.	Radioactivity

Requirements to apparatus

- Short tail in the time response
- Fast time resolution
- Low dark counts
- High rate tolerance
- Simultaneous energy & timing meas.

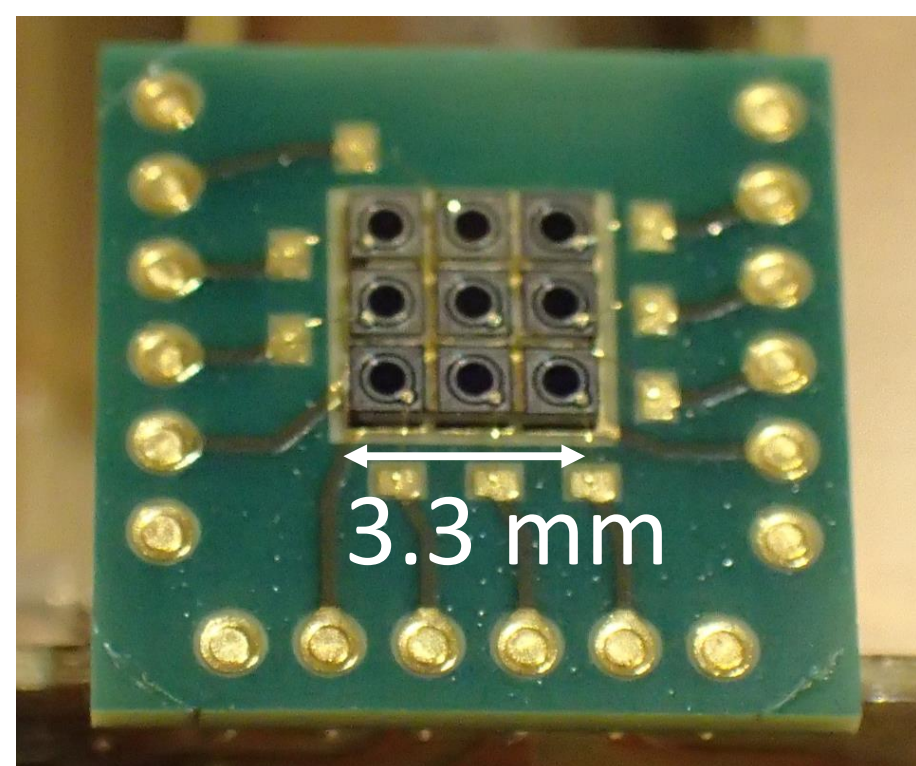
X-ray detector system

Si-APD: Hamamatsu Photonics S12053-05

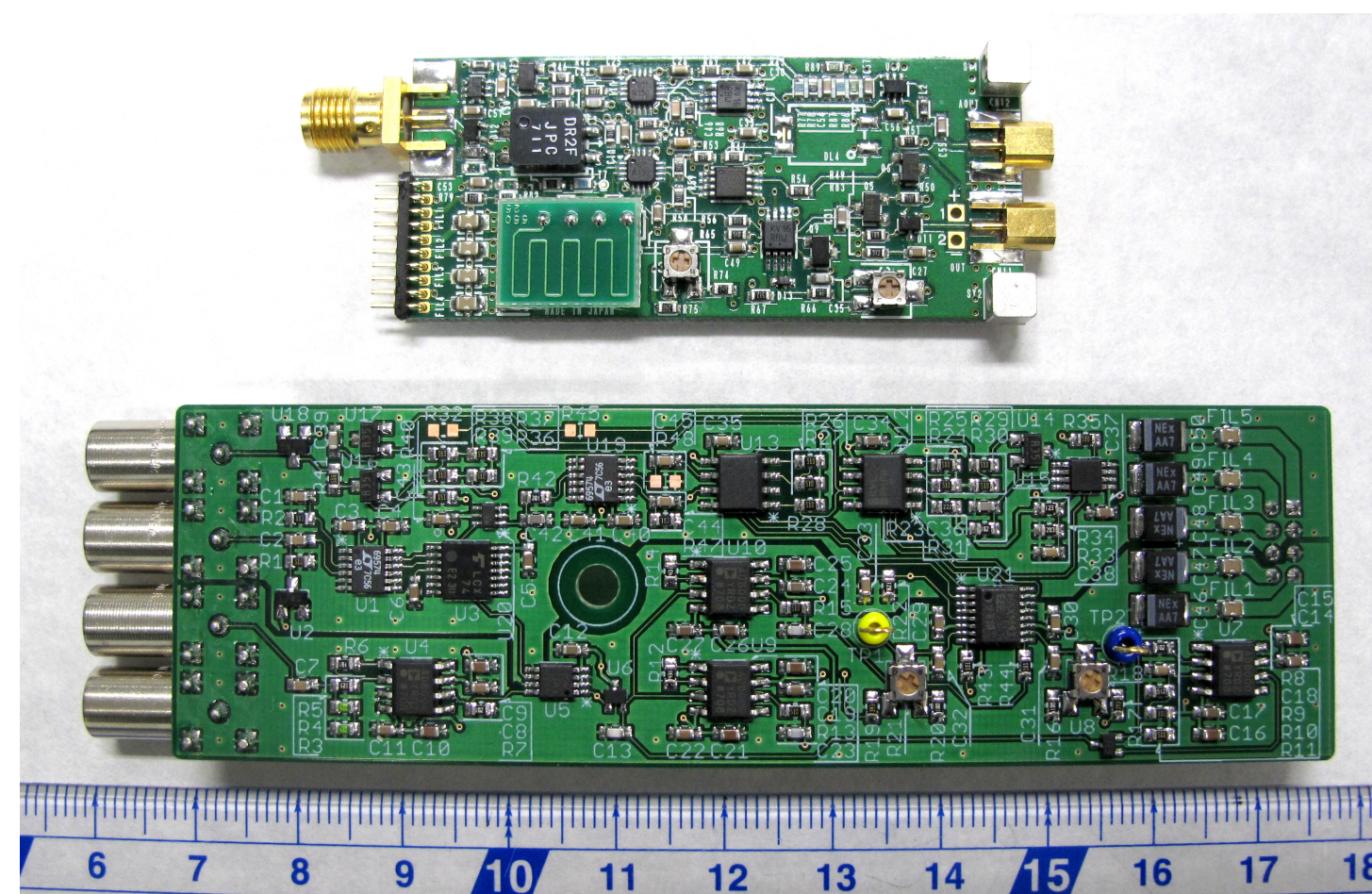
- Small diameter | $0.5\text{mm}\phi$
- Thin depletion layer | $\sim 10\text{ }\mu\text{m}$

Dedicated fast peripheral circuits

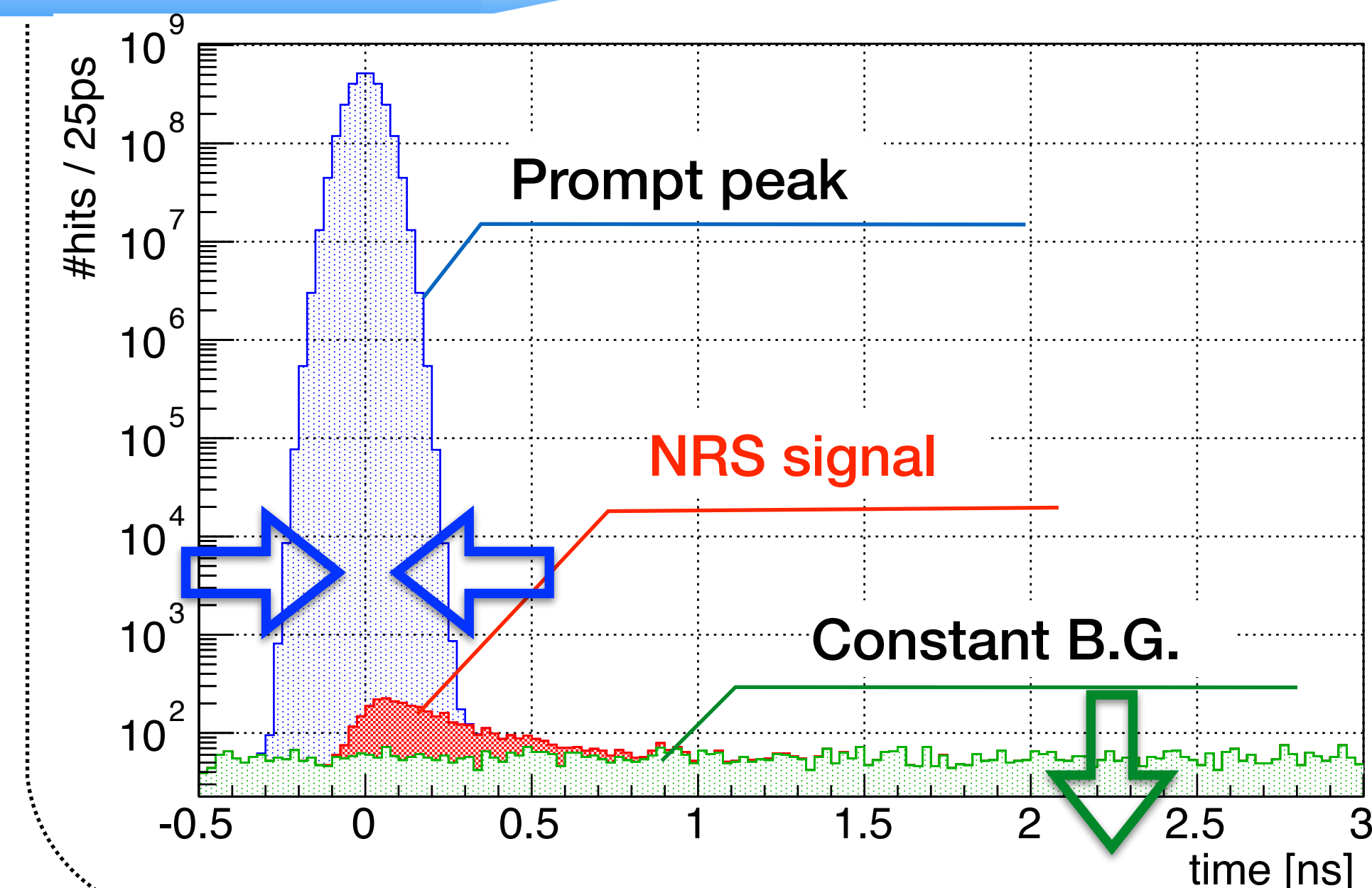
- High rate measurement | 10^6 cps / ch
- Simultaneous energy & timing measurement



customized 9-ch array



Constant fraction discriminator
Amplitude-to-time convertor
Trailing edge discriminator



Multihit TDC
MCS6 (FAST ComTec)

T. Masuda et al., *Rev. Sci. Instrum.* **88**, 063105 (2017).
T. Masuda et al., *Nucl. Instrum. Meth. A.* **913**, 72 (2019).
T. Masuda et. al., *Submitted to JPS: Conf. Ser.*

Dense target & beam

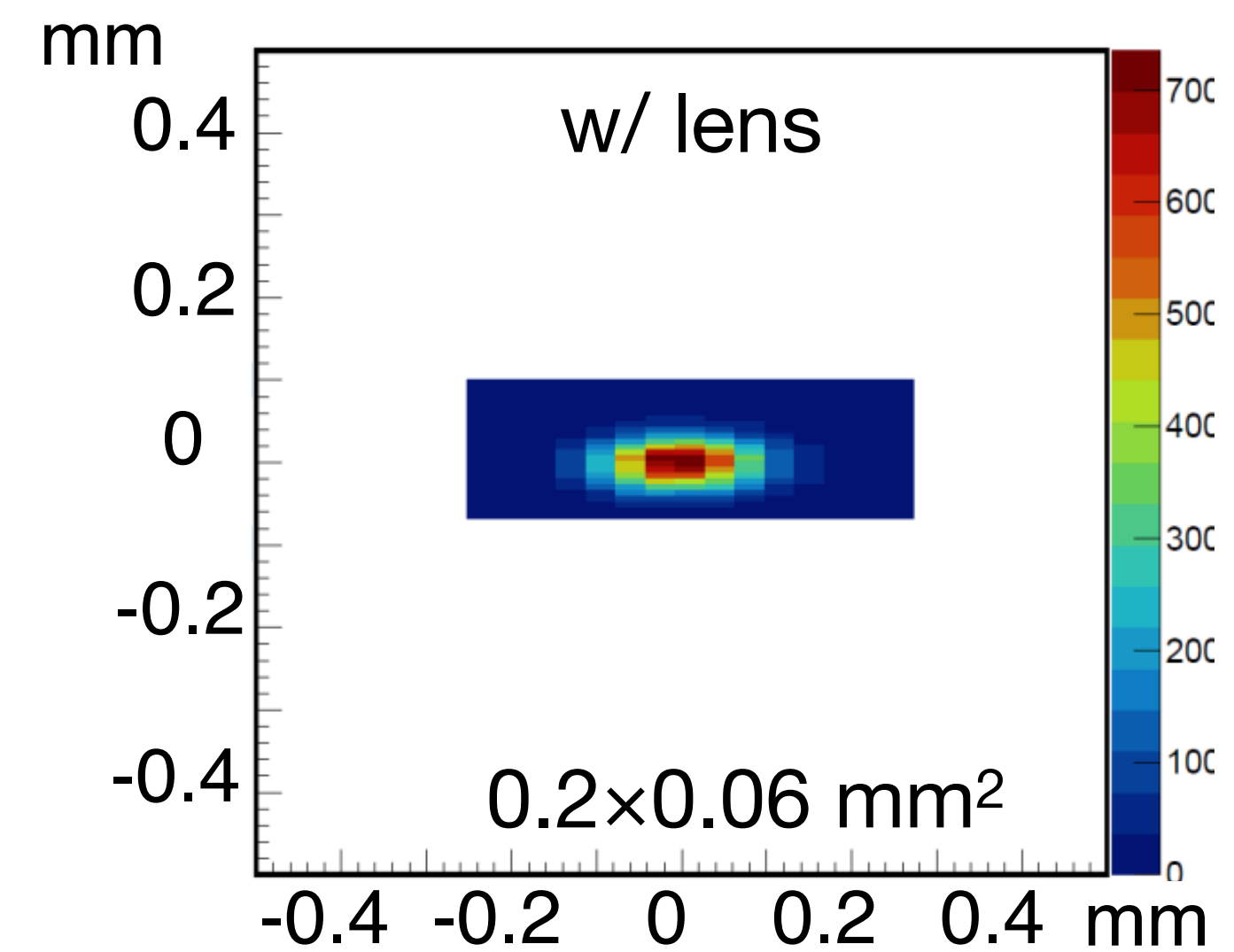
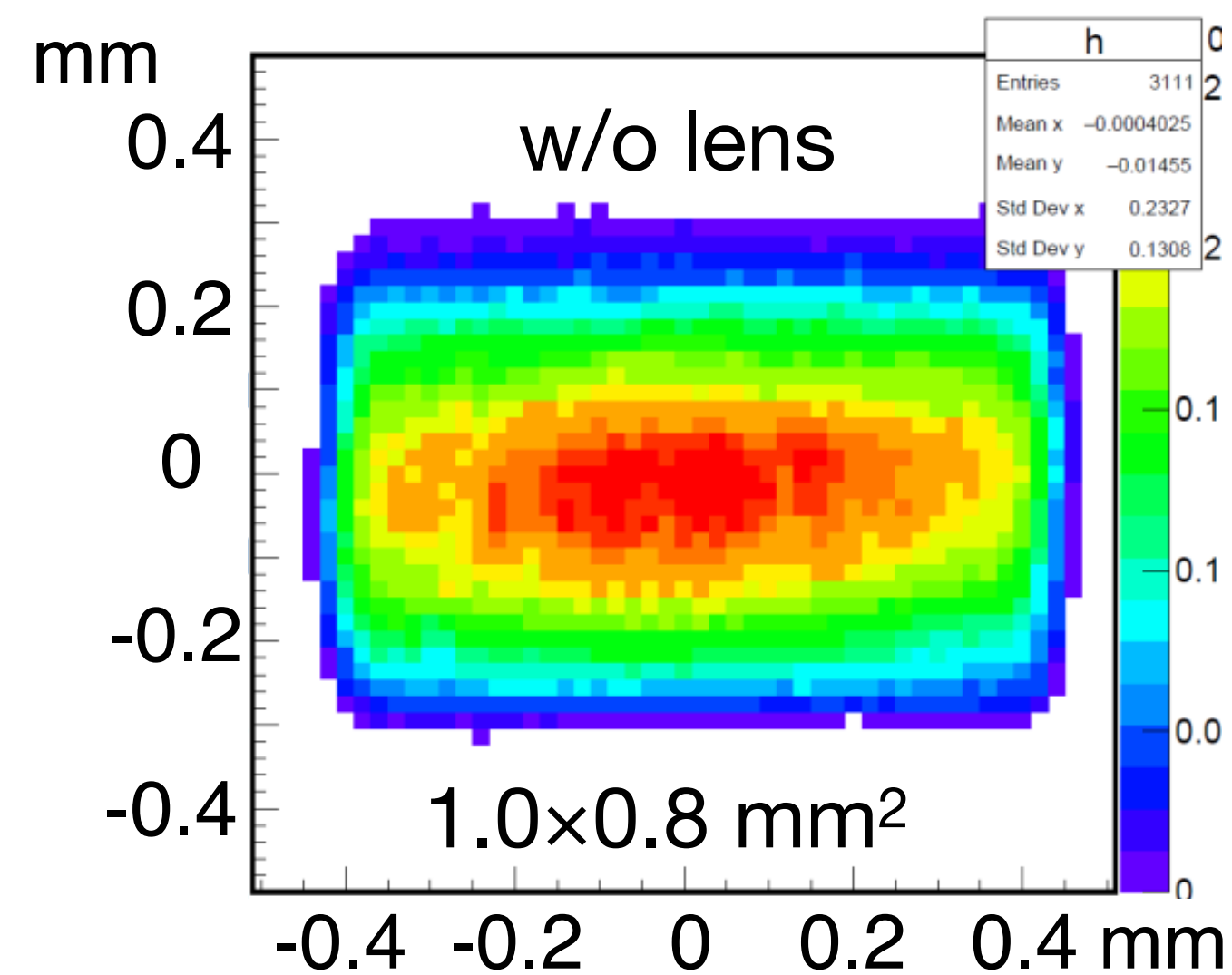
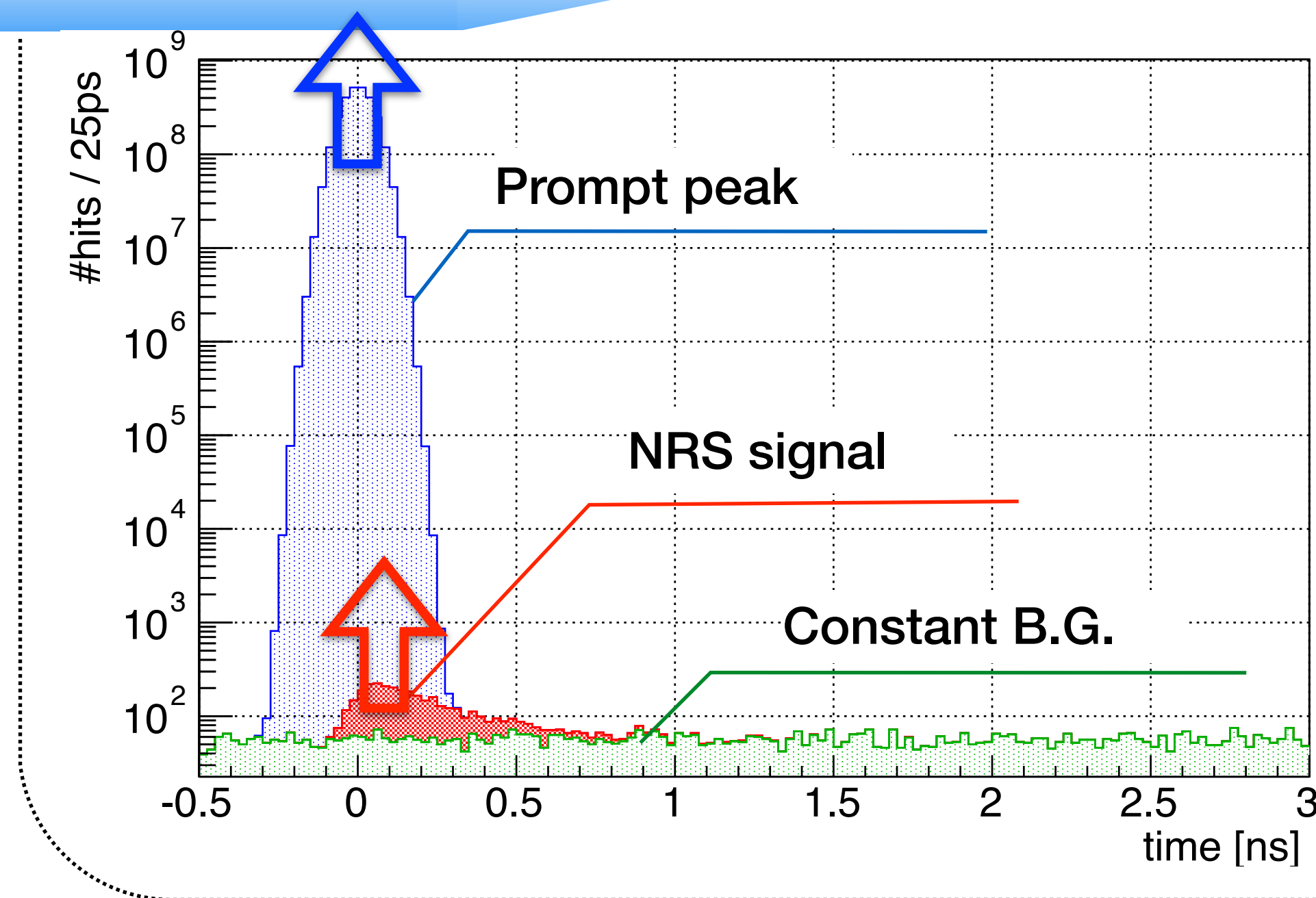
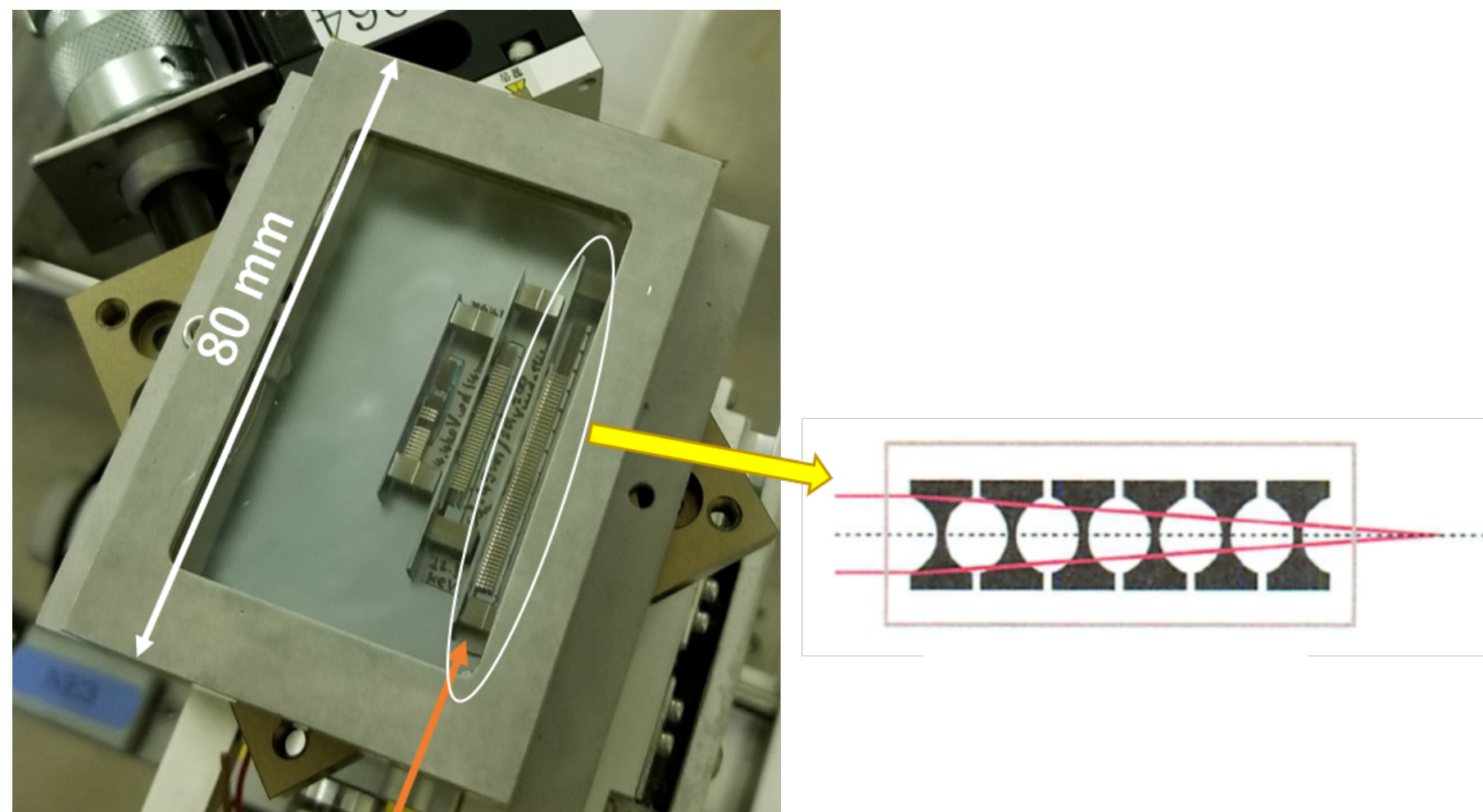
“Dense” Th target

- dry-up method
- $0.24\ \mu\text{g}$, $0.4\text{mm}\phi$



“Focused” X-ray beam

- Focused with a refractive lens array
- $1.0 \times 0.8\ \text{mm}^2 \rightarrow 0.2 \times 0.06\ \text{mm}^2$ (FWHM)

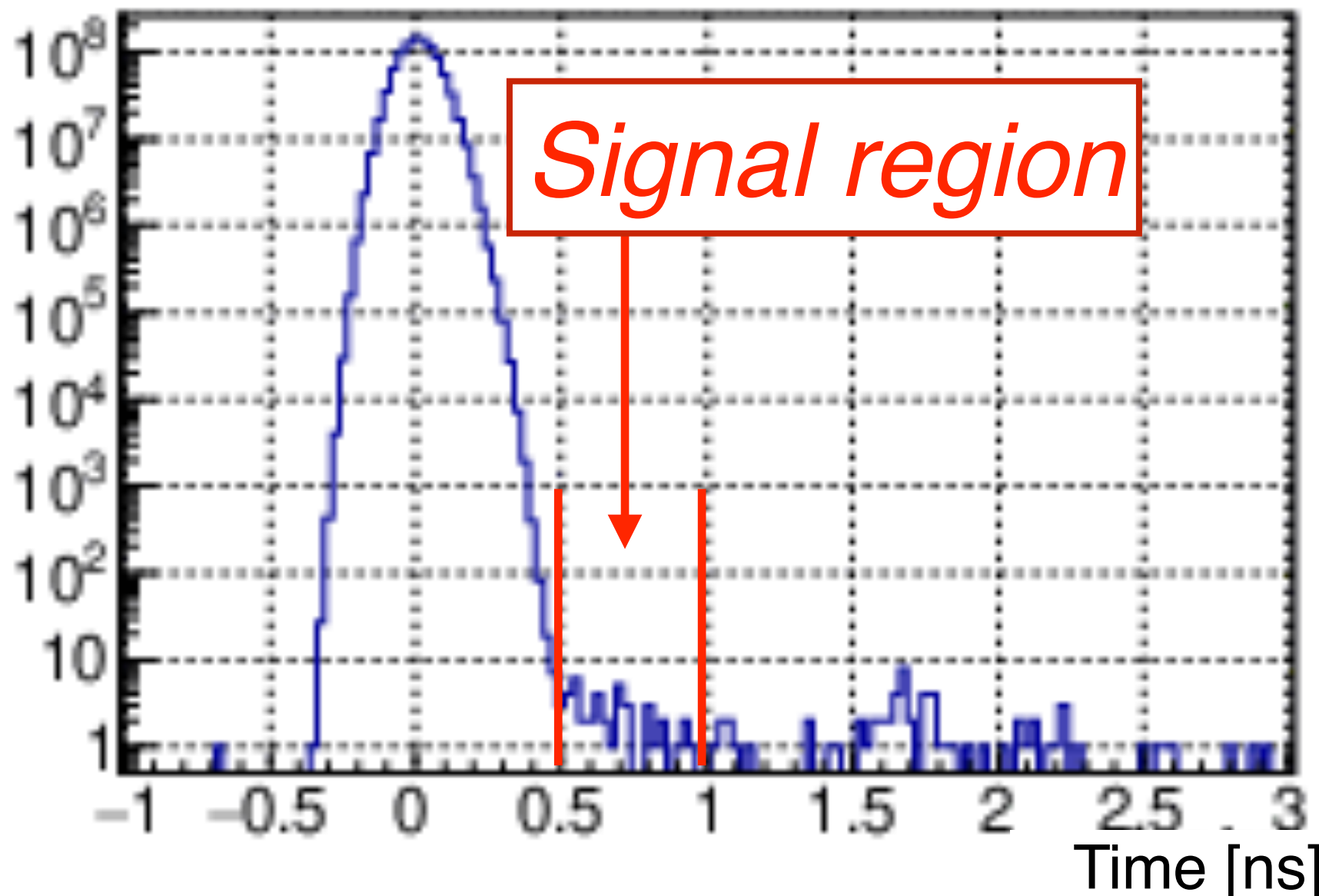


The most intense X-ray synchrotron radiation facility in Japan.

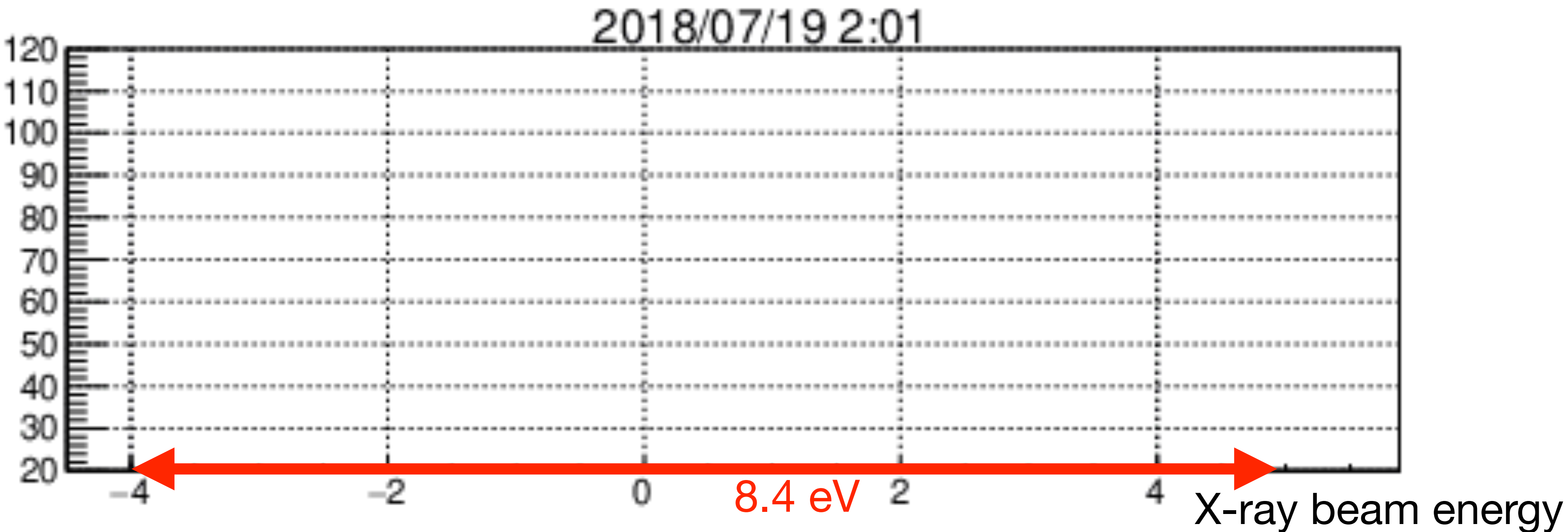


NRS search

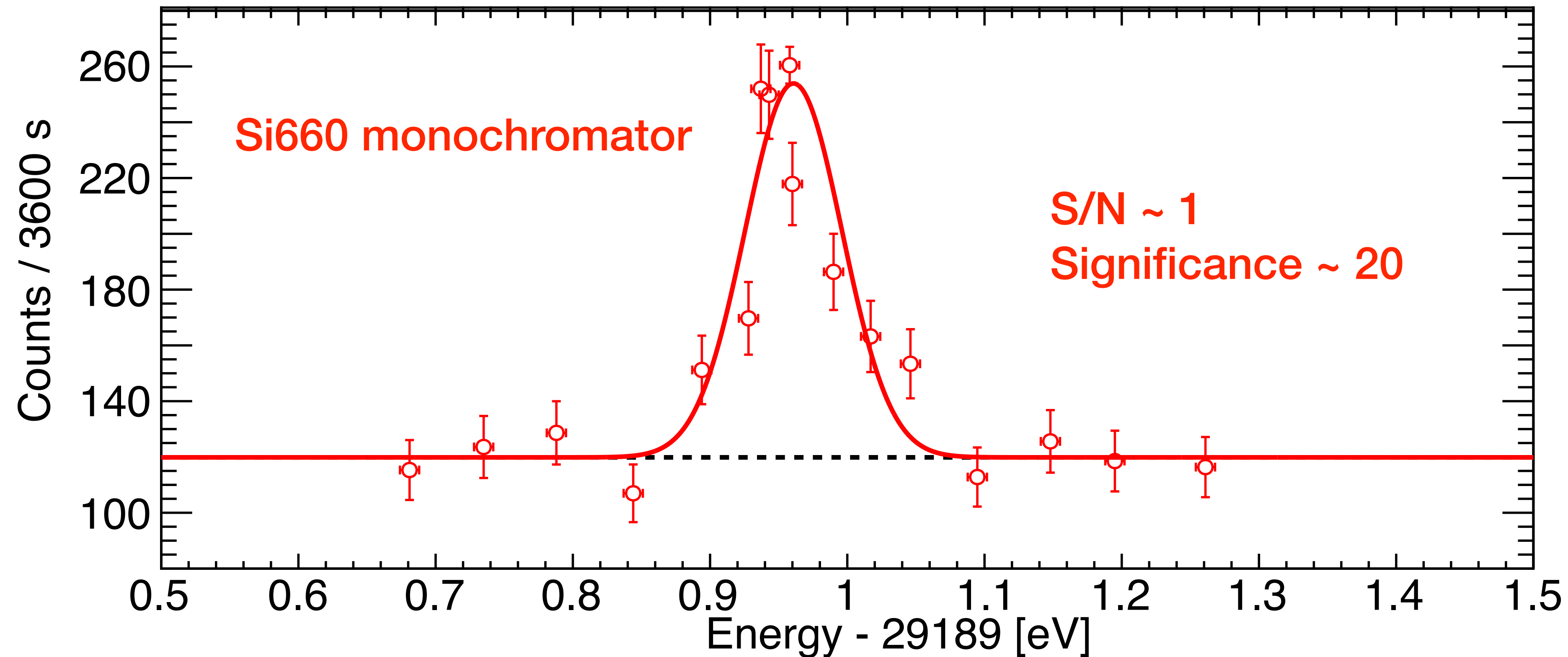
Beam time	2018/7/15 9:00- 7/23 9:00
Monochro.	Si440
Scan range	8.4 eV (0.1 eV step)
Scan points	113 points (1800s/point)
Scan time	67 hours (7/19 2:00 - 21 21:00)



Number of events
in the signal region



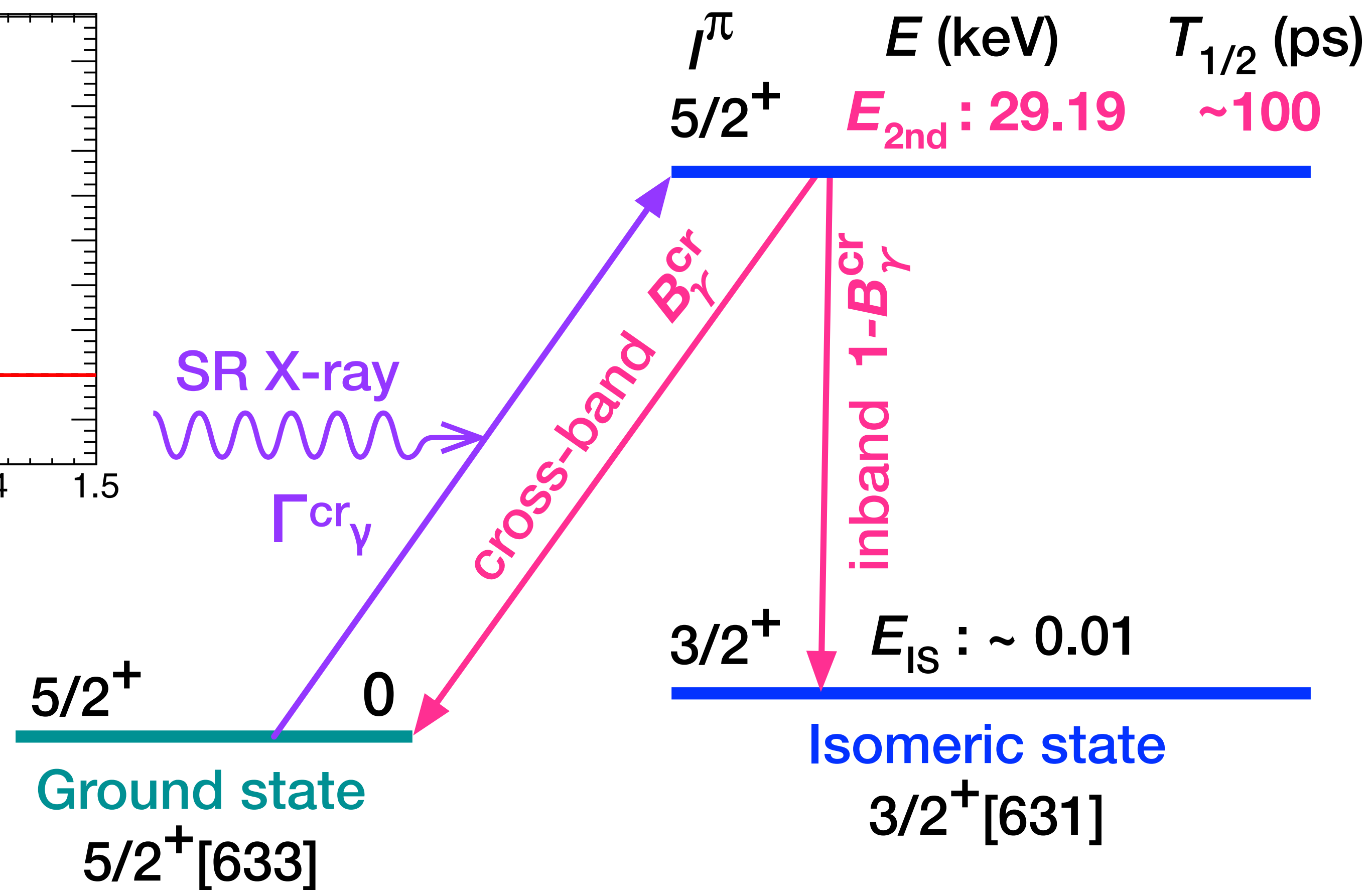
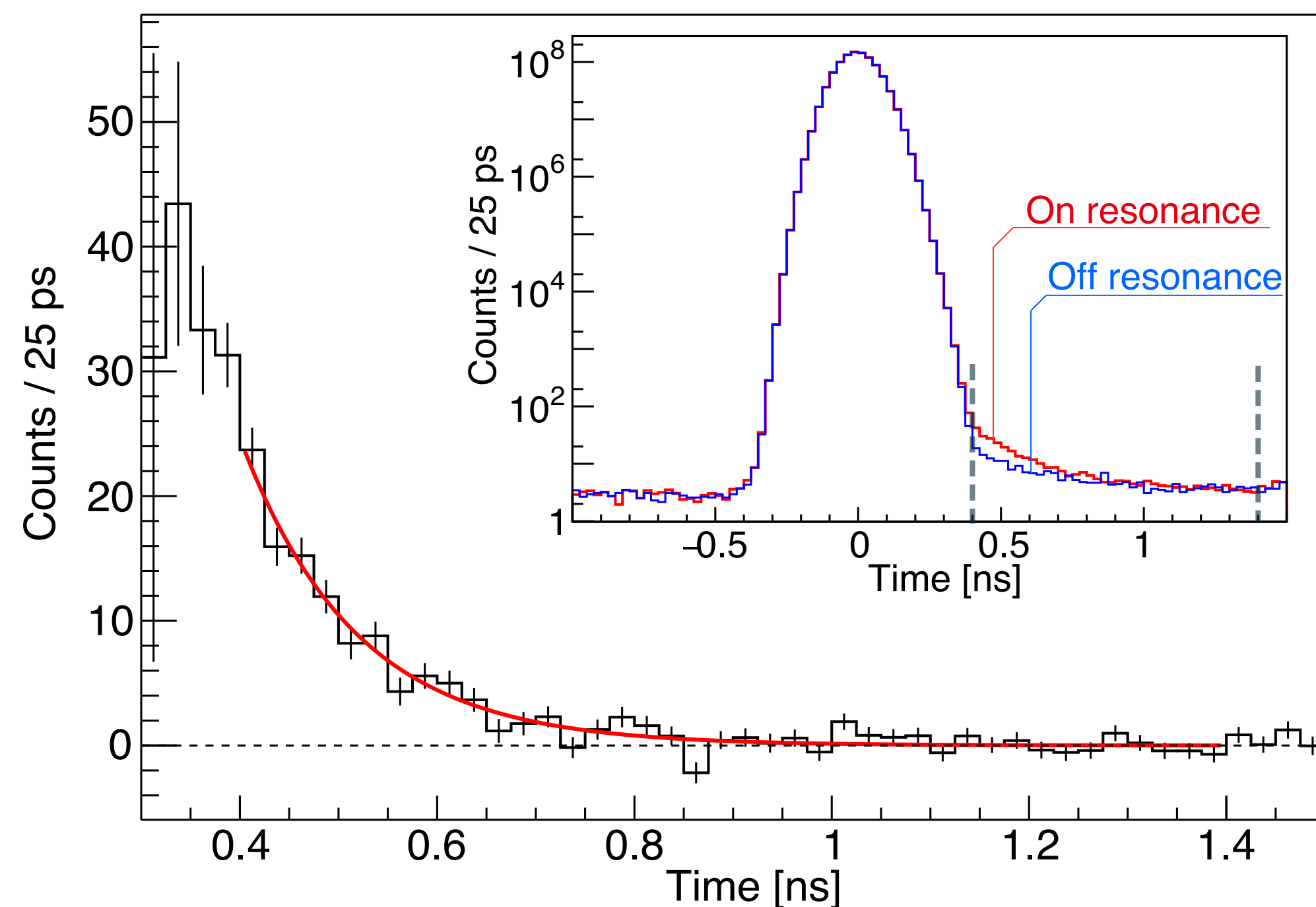
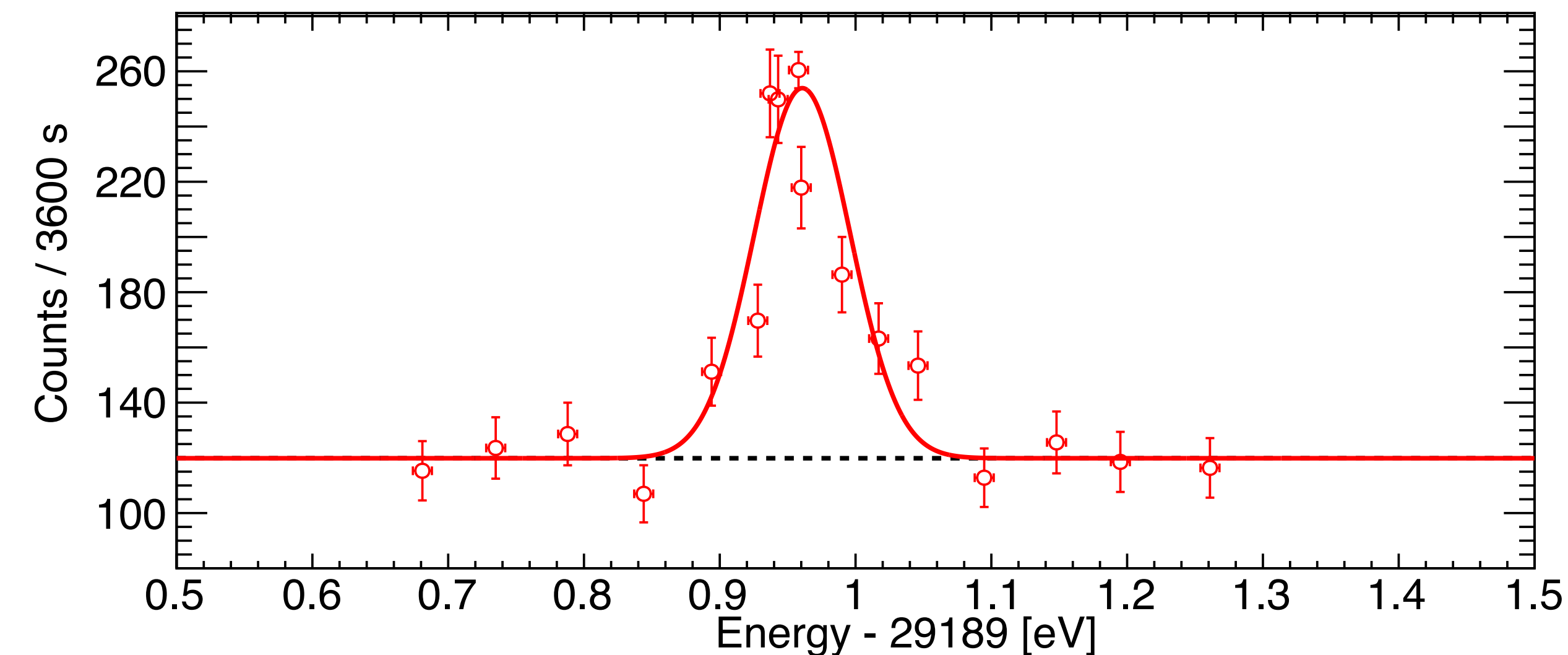
The NRS peak has been observed.



The world's first demonstration on
the active pumping to the isomer state



Obtained nuclear parameters

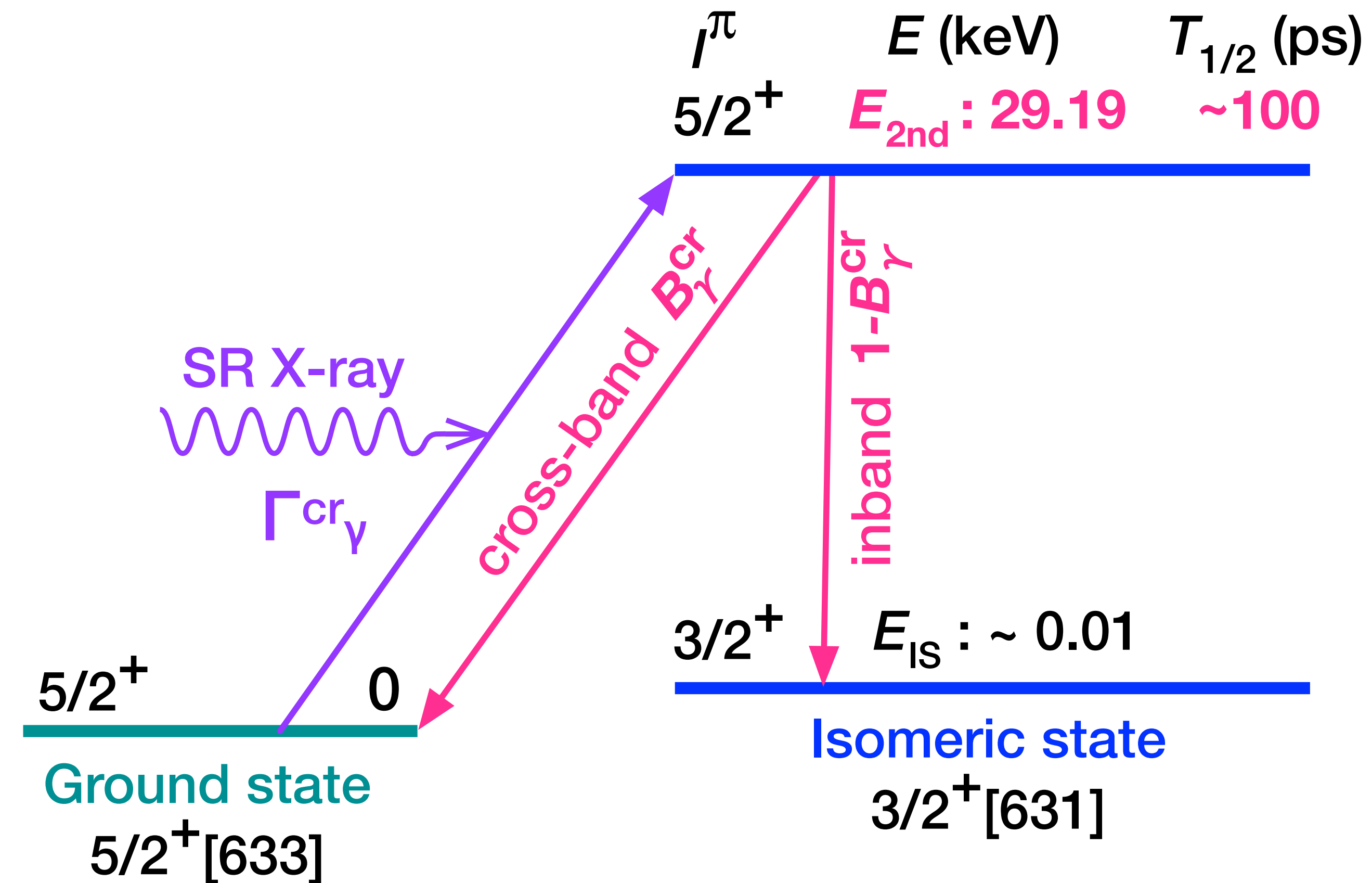


$$E_{2\text{nd}} = 29189.93 \pm 0.07 \text{ eV (2 ppm)}$$

$$T_{1/2} = 82.2 \pm 4.0 \text{ ps (Fastest NRS meas.)}$$

$$\Gamma_{\text{cr}\gamma} = 1.70 \pm 0.40 \text{ neV}$$

$$B_{\text{cr}\gamma} = 1/(9.4 \pm 2.4) \sim 0.11$$



Isomer production rate $\sim 2.5 \times 10^4$ cps

- Isomer state of Th-229 : the extremely-low energy ~ 8 eV
will building a new bridge between atomic & nuclear physics
- The world's first artificial pumping to the isomer state has been realized.
- The next step : VUV emission detection & spectroscopy