



Tokyo Tech

中性子ビームを用いた 中性子捕獲反応の測定

片渕竜也

東京工業大学

ゼロカーボンエネルギー研究所

研究会「中性子捕獲反応で迫る宇宙の元素合成」 2023年2月10日

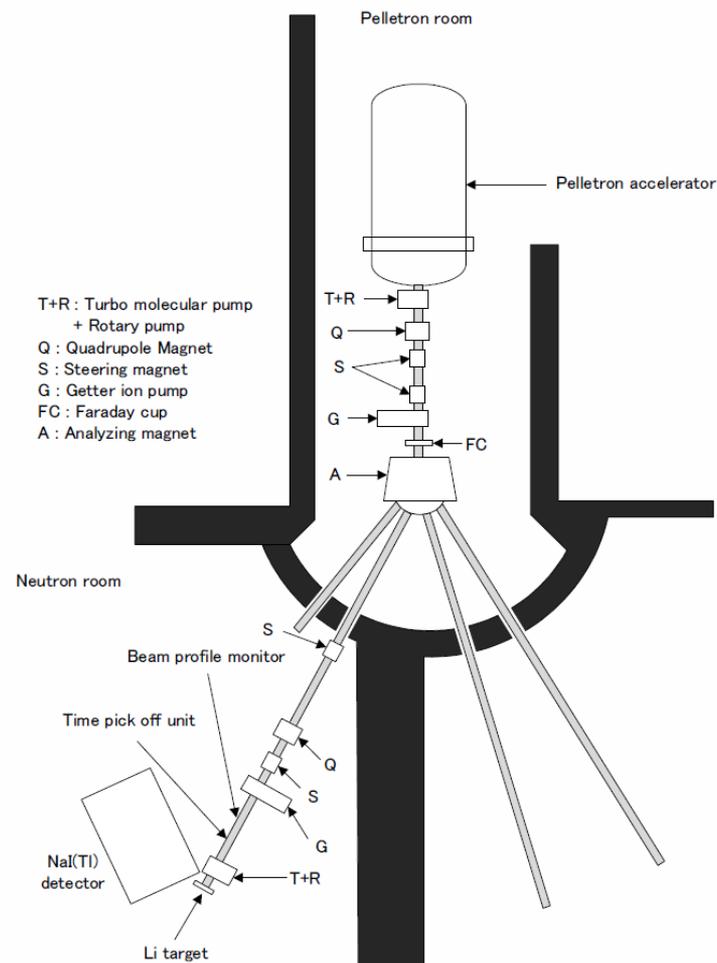
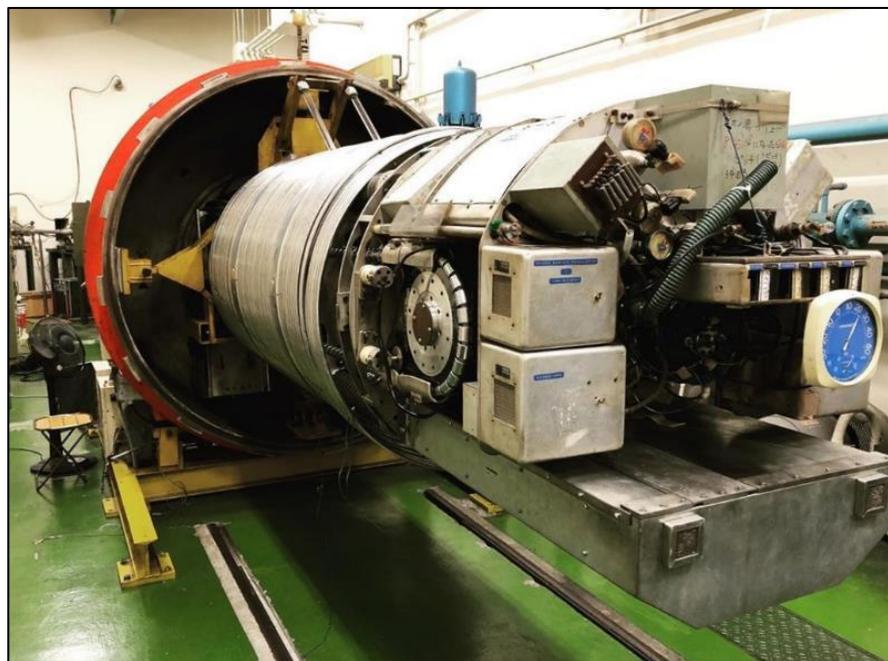


1. 東工大ペレットロン加速器
2. 飛行時間法と放射化法
3. ^{138}Ba の中性子捕獲断面積測定
4. ^{185}Re の中性子捕獲断面積測定
5. J-PARCでの実験

東工大ペレットロン加速器

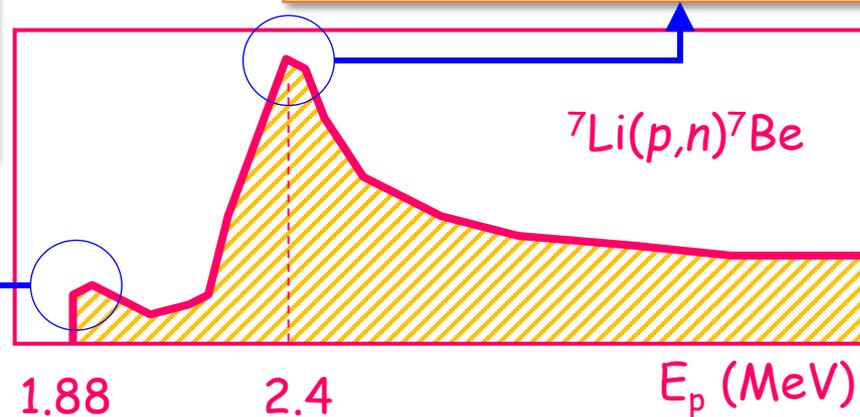
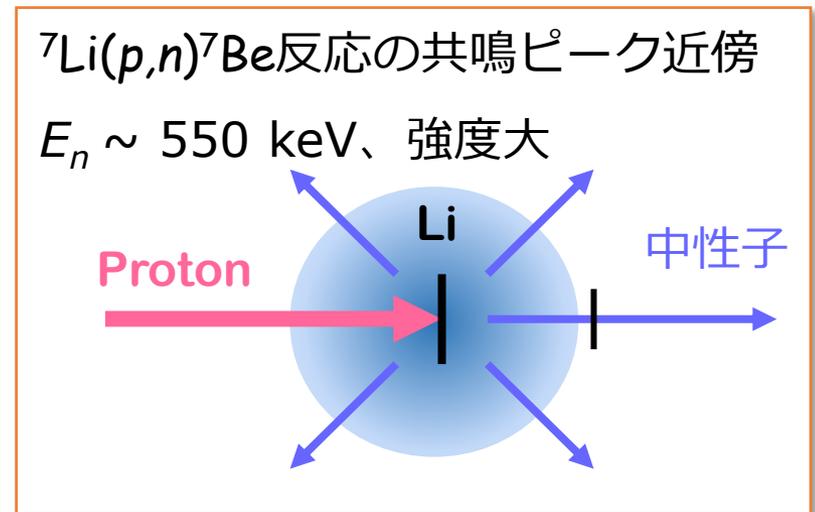
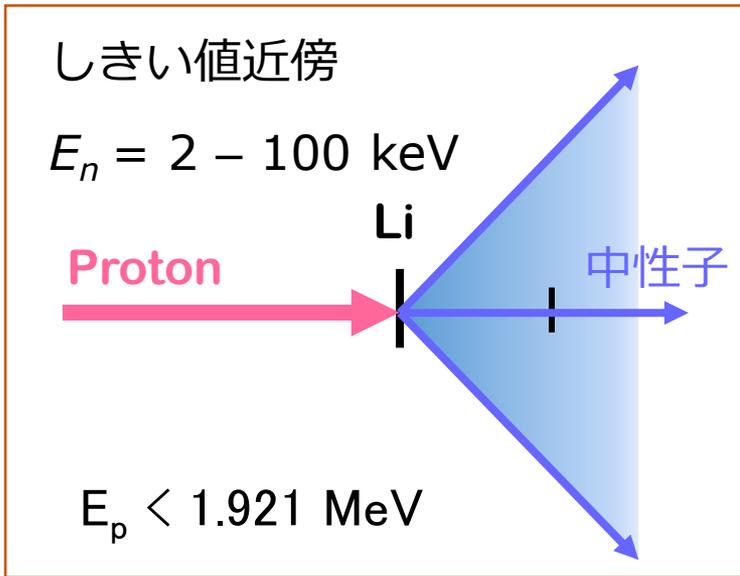
性能・仕様：

- Pelletron 3UH-HC (米国nec社製)
- 加速電圧：最大 3 MV
- 加速可能イオン種：p, d, α
- 最大ビーム電流：40 μ A (パルス)
- 最大ビームエネルギー：pで3 MeV
- パルスビーム (幅1.5ns, 4MHz)



中性子源

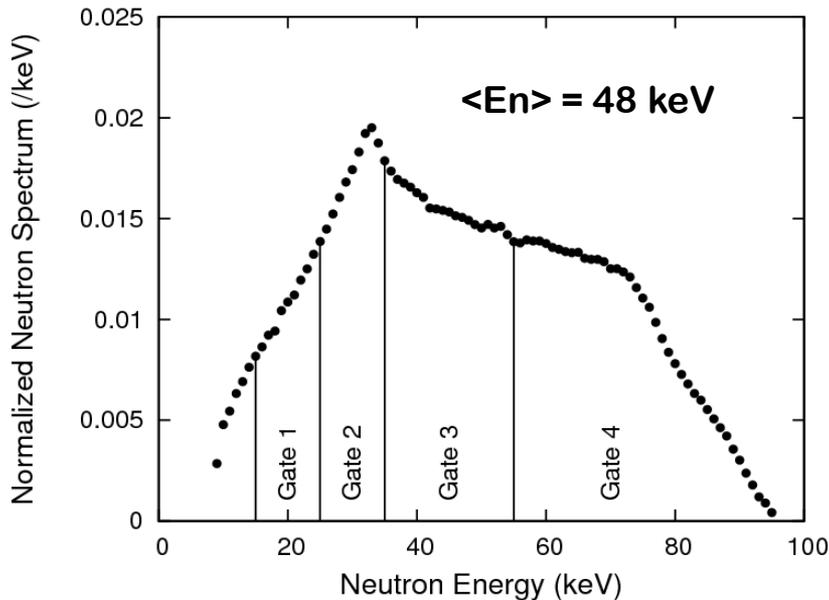
- ${}^7\text{Li}(p,n){}^7\text{Be}$ 反応により中性子発生、 $Q=-1.644\text{ MeV}$, $E_{\text{th}}=1.880\text{ MeV}$
- keVエネルギーの中性子を発生
- 中性子飛行時間法を利用



中性子エネルギースペクトル

しきい値近傍

$$E_n = 2 - 100 \text{ keV}$$



全中性子数:

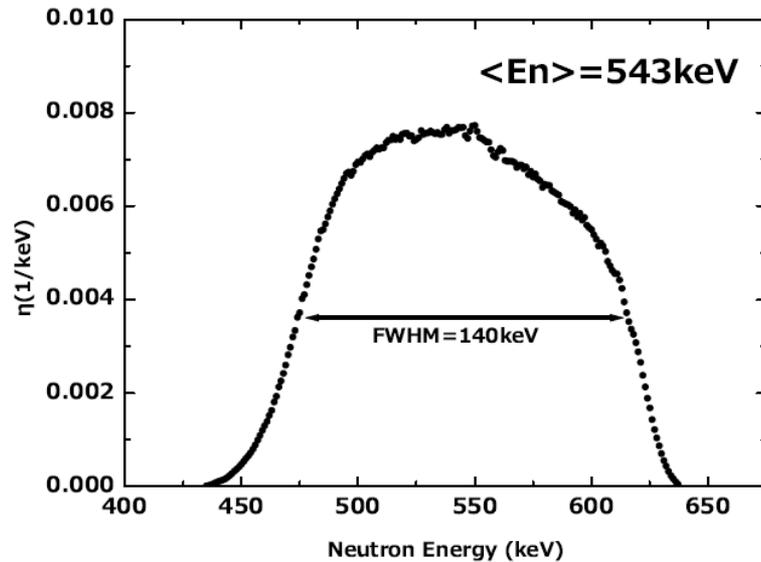
$$8 \times 10^6 \text{ n/sec}$$

中性子フラックス@12 cm:

$$3 \times 10^5 \text{ n/cm}^2\text{/sec}$$

${}^7\text{Li}(p,n){}^7\text{Be}$ 反応の共鳴ピーク近傍

$$E_n \sim 550 \text{ keV、強度大}$$



全中性子数:

$$5 \times 10^9 \text{ n/sec}$$

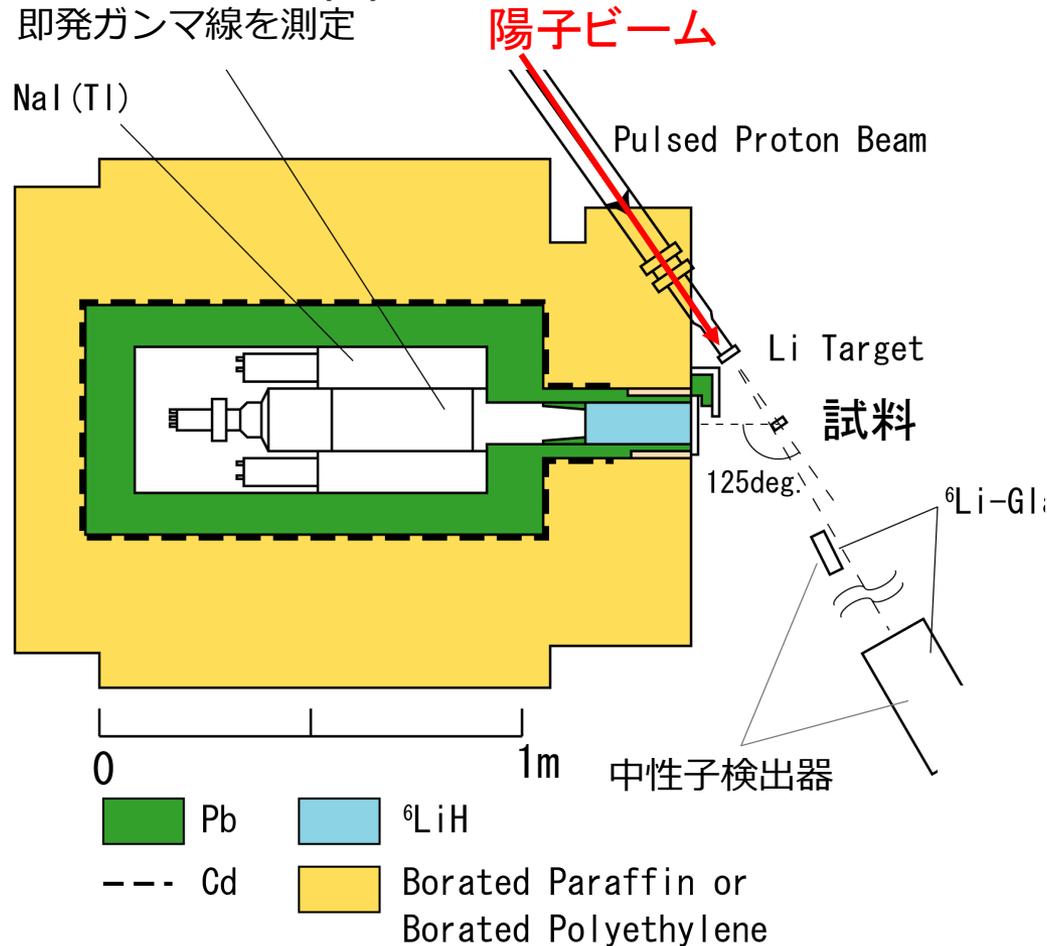
中性子フラックス@20 cm:

$$1 \times 10^6 \text{ n/cm}^2\text{/sec}$$

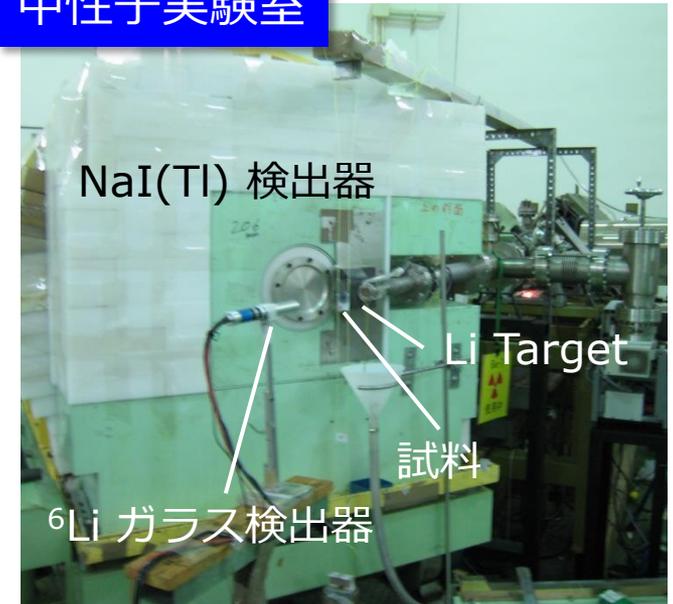
中性子ビームライン

- 飛行距離: 12 cm or 20 cm
- ガンマ線検出器: NaI(Tl) 150 mm Φ \times 300 mm t
- 中性子検出器: ^6Li ガラス検出器
- データ収集: 飛行時間 + 波高

ガンマ線検出器NaI(Tl)
即発ガンマ線を測定



中性子実験室



s-process 中性子魔法数近傍

- N=82

^{142}Nd , ^{141}Pr , ^{140}Ce , ^{139}La , ^{138}Ba

- N=50

^{88}Sr , ^{89}Y

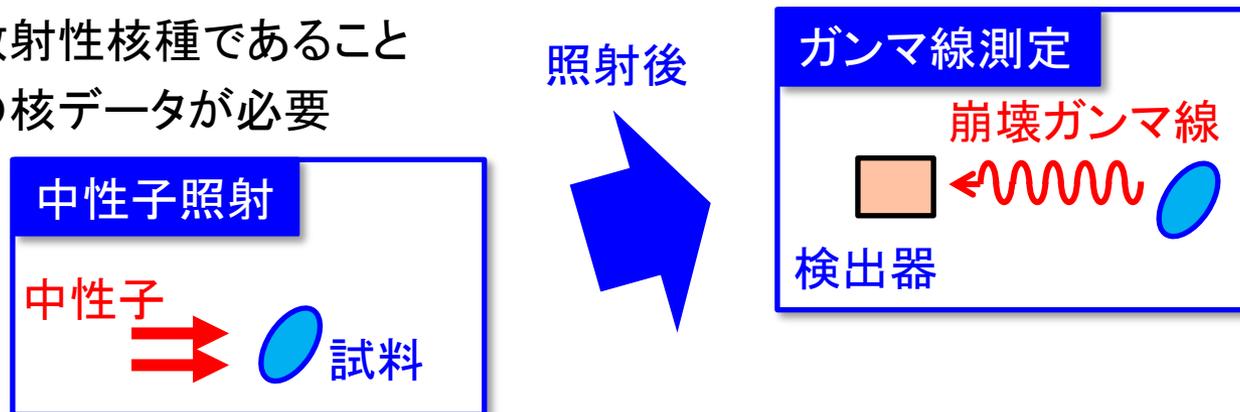
r-process関連、宇宙核時計

- ^{185}Re

中性子捕獲断面積の2つの測定方法

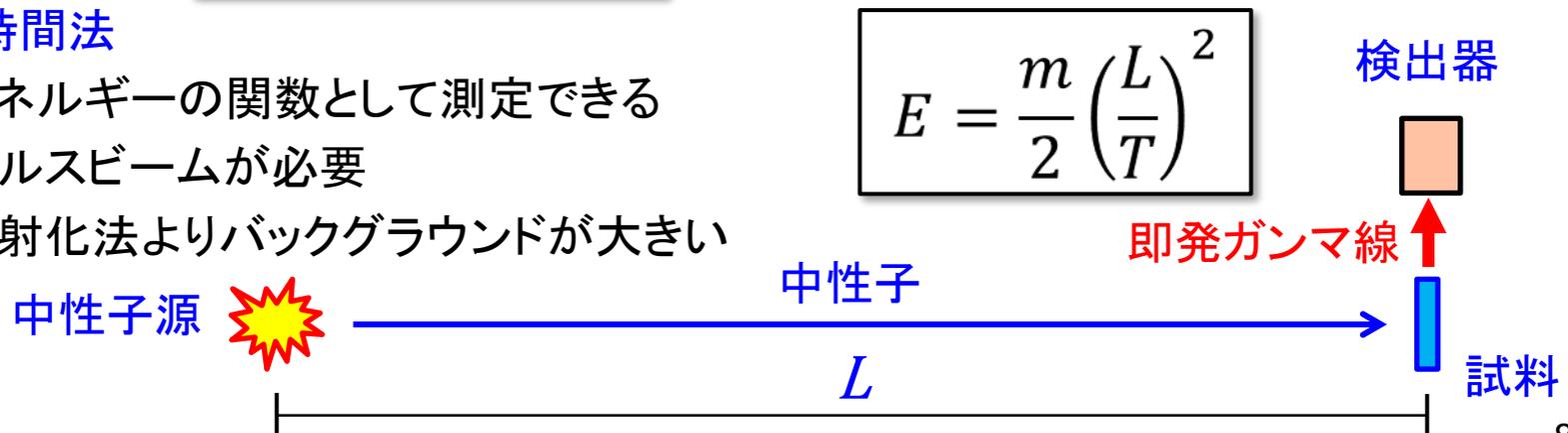
放射化法

- ◆ 飛行時間法に比べ低バックグラウンド。微量試料でも可能
- ◆ 特定のエネルギーのみ 25 meV(原子炉)、25 keV (Sb-Be中性子源)
 ${}^7\text{Li}(p,n){}^7\text{Be} \rightarrow$ 疑似マクスウェル分布 ($kT=25 \text{ keV}$)
- ◆ 残留核が放射性核種であること
- ◆ 分岐比等の核データが必要



飛行時間法

- ◆ エネルギーの関数として測定できる
- ◆ パルスビームが必要
- ◆ 放射化法よりバックグラウンドが大きい





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^{138}Ba の中性子捕獲断面積測定

- 飛行時間法による測定 -



Sample

Ba-138 sample

Chemical form	BaCO ₃
Physical form	Powder
Chemical purity	99.995 %
Net weight (Ba-138)	29.84 g
Diameter	56.2 mm
Thickness	12.7 mm

Heated at 150° C
for 6.5 h.

Encased in a graphite
container

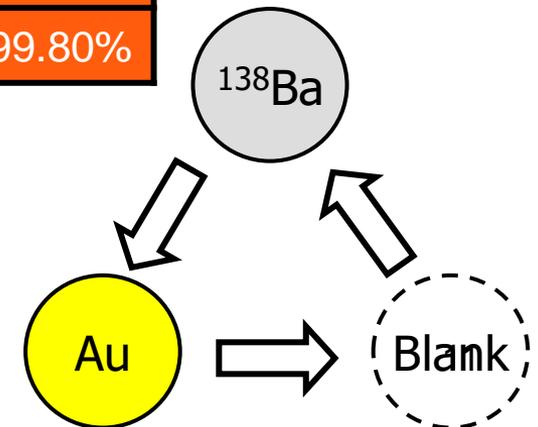


Isotopic composition

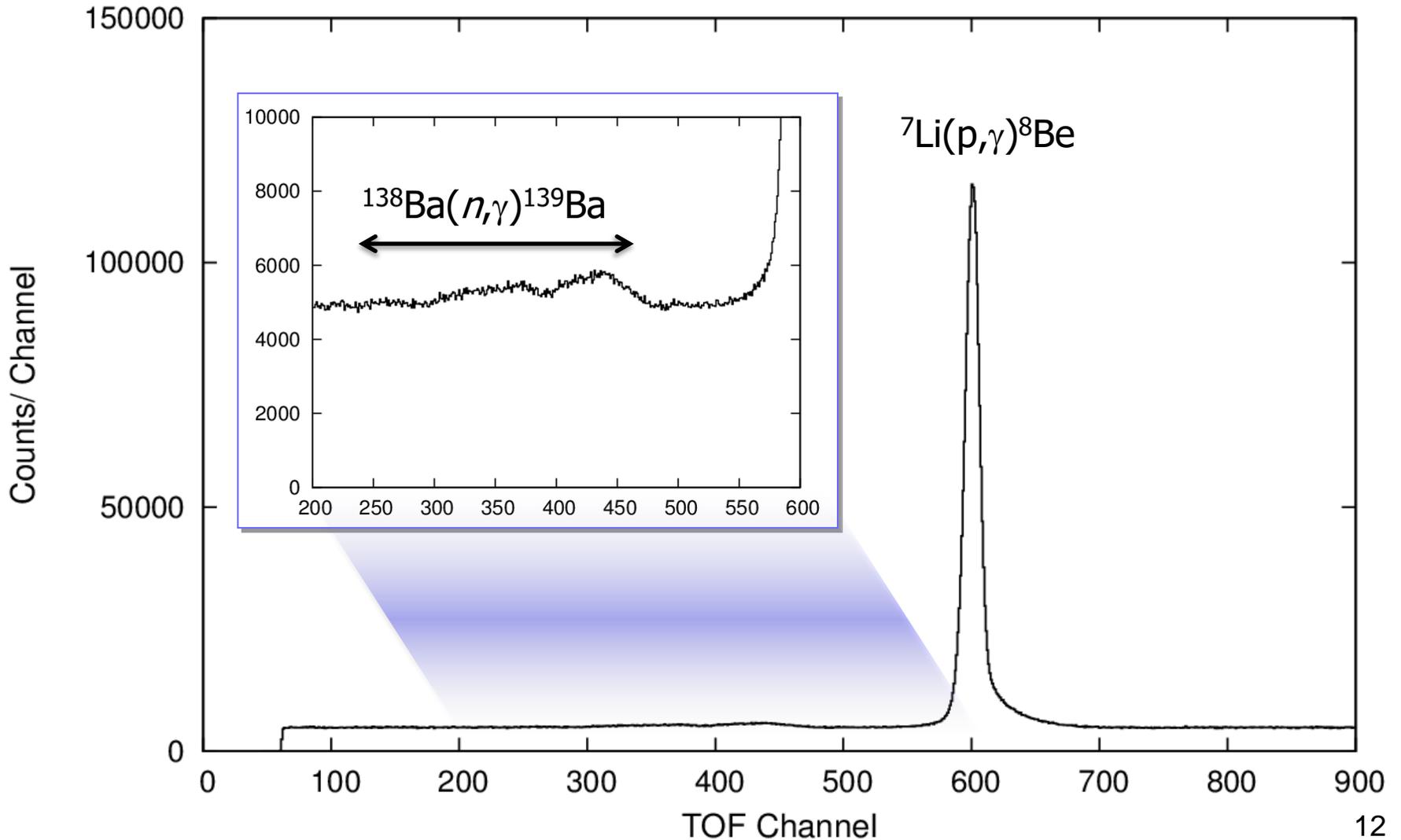
Ba-130	Ba-132	Ba-134	Ba-135	Ba-136	Ba-137	Ba-138
<0.01%	<0.01%	<0.01%	0.02%	0.02%	<0.16%	99.80%

Measurement time

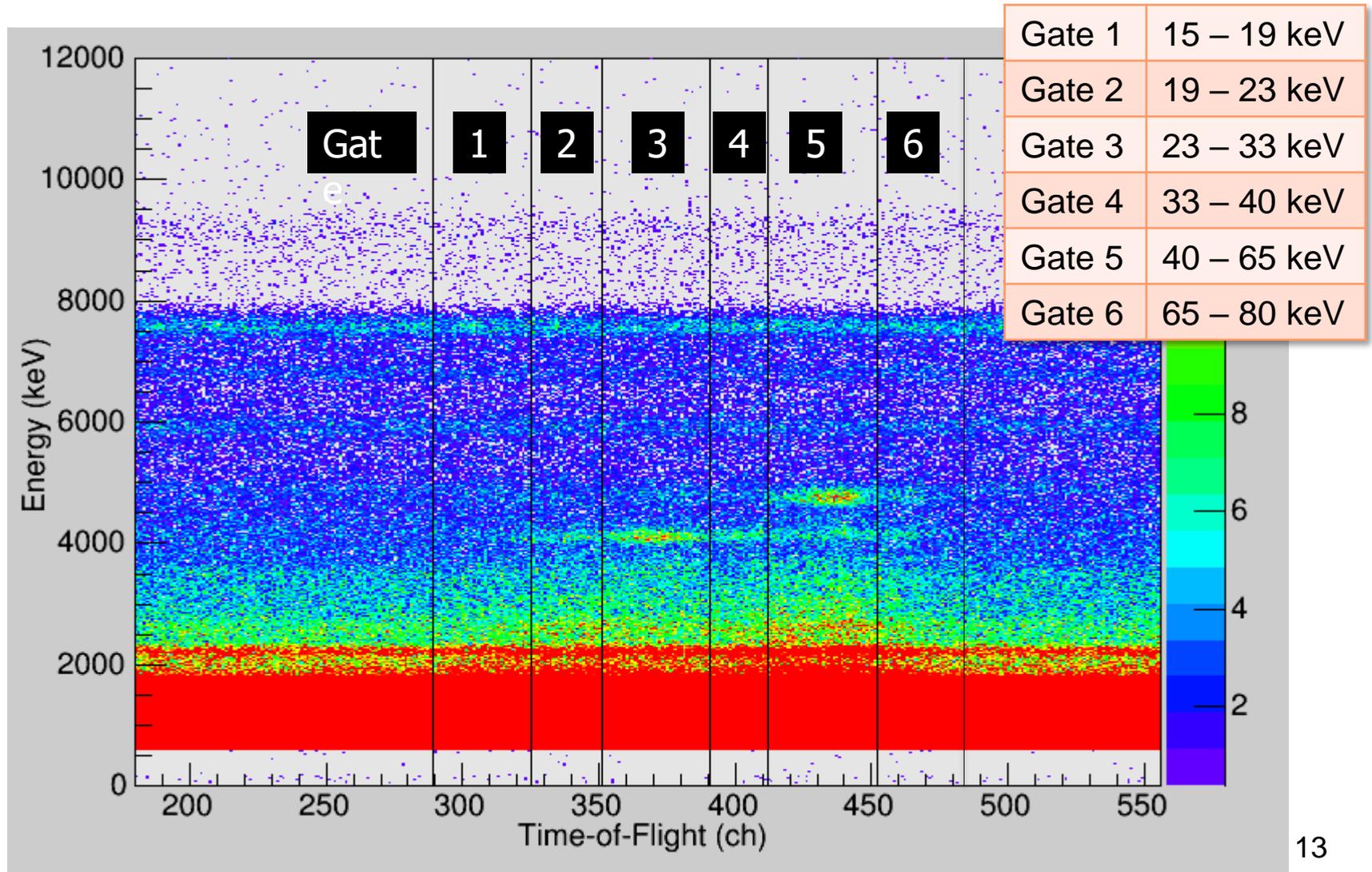
Ba-138	Au	Blank
63 h	2 h	6 h



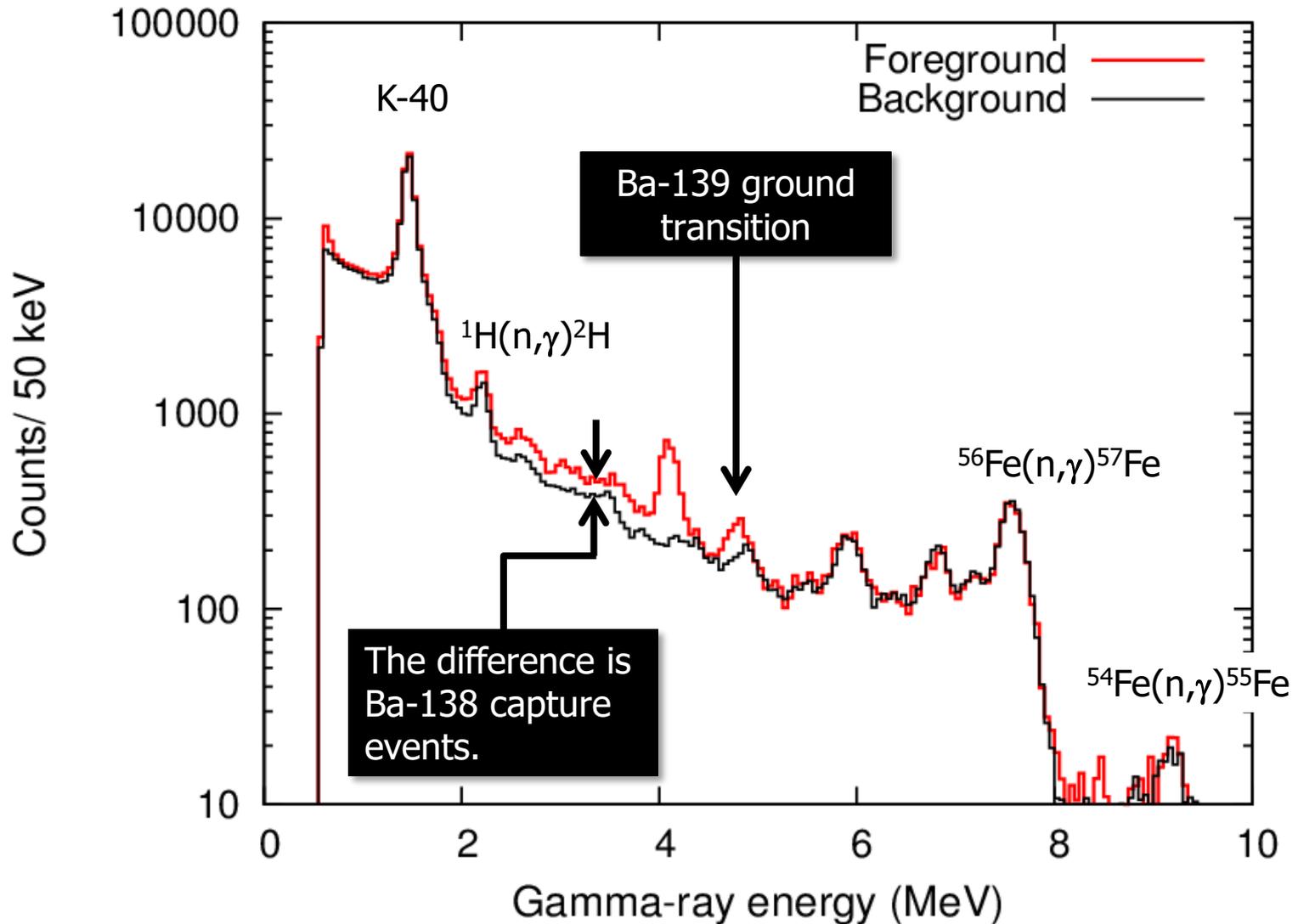
Gamma-Ray TOF Spectrum



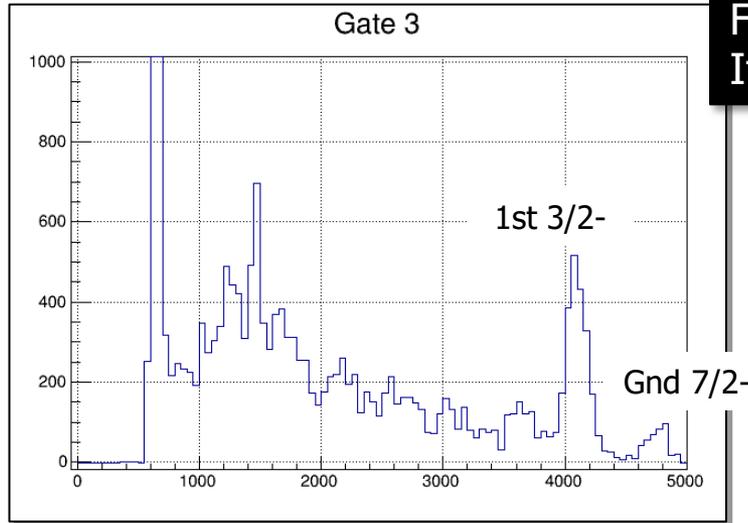
2D spectrum of TOF vs. PH



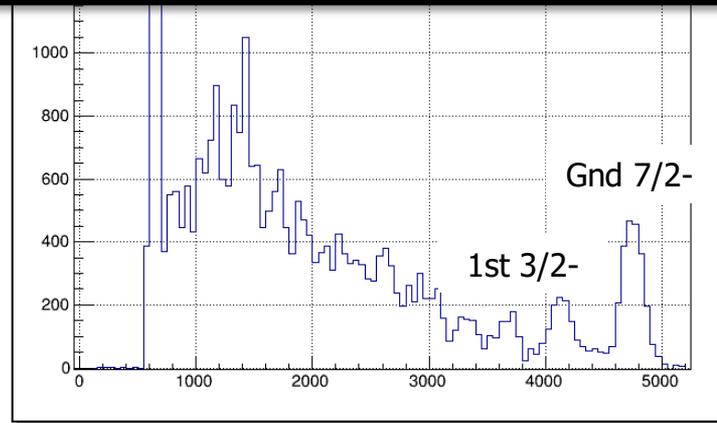
Background subtraction (23 - 33 keV)



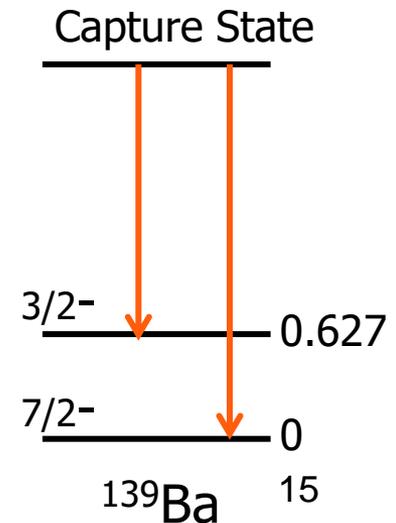
Net pulse-height spectra



From $3/2^-$ to $7/2^-$, E2 transition is required.
If it is E1 ($5/2^+ \rightarrow 7/2^-$), D-wave resonances exist.

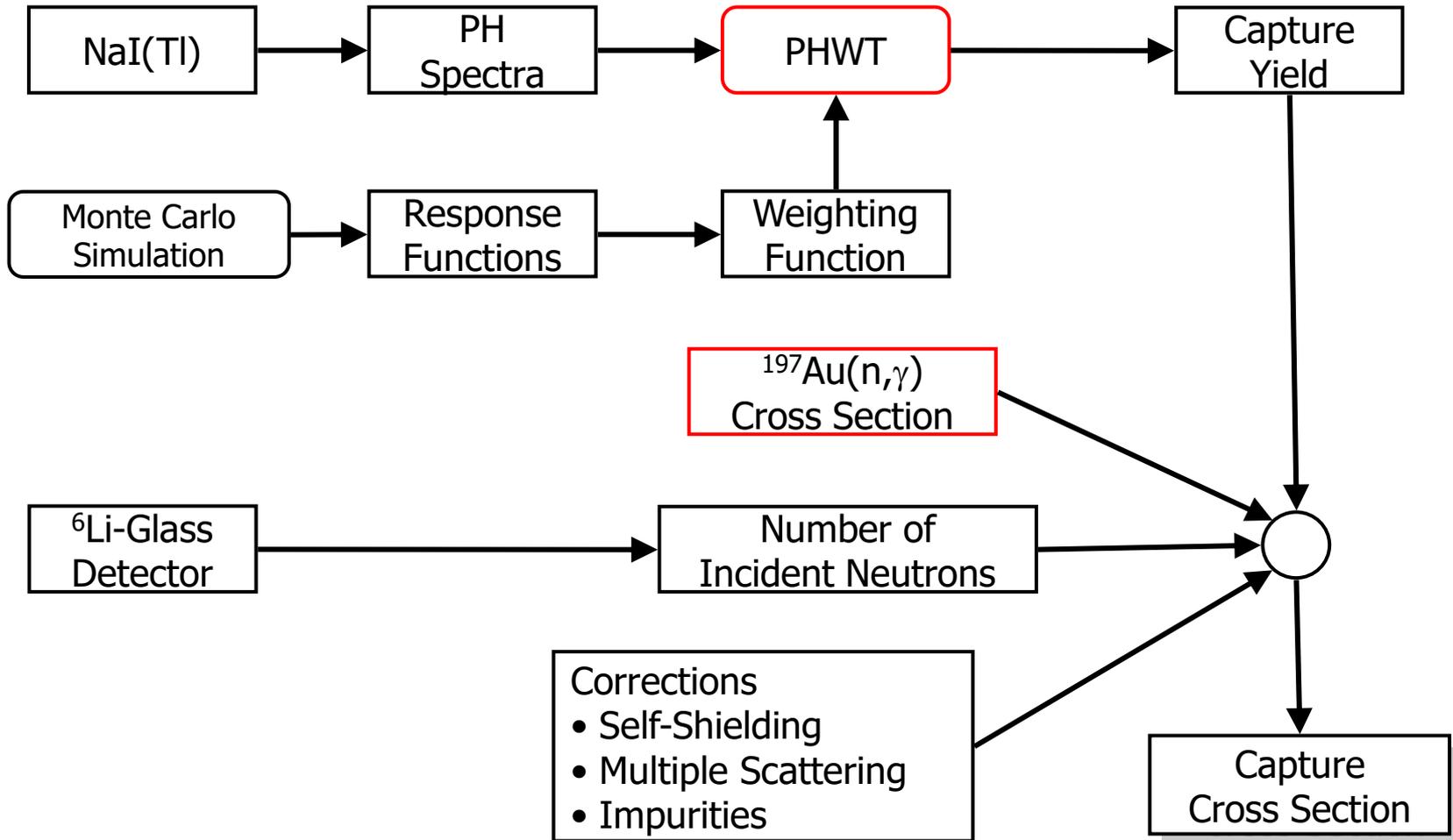


Gate	E_n (keV)	ENDF/B-VII.1			JENDL-4.0			Mughabghab (2006)			
		L	1	?	0	1	?	0	1	?	
	J	$1/2^+$	$1/2^-$	$3/2^-$	$1/2^+$	$1/2^-$	$3/2^-$	$1/2^+$	$1/2^-$	$3/2^-$?
1	15 - 19	0	0	0	0	1	0	0	0	0	0
2	19 - 23	0	0	0	0	1	1	0	0	0	2
3	23 - 33	1	1	1	1	2	3	1	1	1	6
4	33 - 40	0	0	0	0	2	2	0	0	0	1
5	40 - 65	2	1	5	6	0	6	2	1	5	9
6	65 - 80	1	1	1	3	0	3	1	1	1	6



E_0 (keV)	J	l	$g\Gamma_n$ (eV)	Γ_γ (meV)	$g\Gamma_n^0$ (meV)	$g\Gamma_n^1$ (meV)	$g\Gamma_n\Gamma_\gamma/\Gamma$ (meV)
-0.1775	1/2	0		(55)	555.6		
0.648		1	0.0050 ± 0.0002			129	4.91 ± 0.14
1.948	[3/2]	1	0.101 ± 0.008	29 ± 10		500	37 ± 1
2		1					0.8 ± 0.1
4.704	[1/2]	1	0.81 ± 0.06	32 ± 3		1080	31 ± 1
7.876	1/2	0	6.172 ± 0.053	67 ± 5	69.6 ± 0.6		66 ± 2
9.918	[1/2]	1	0.36 ± 0.04	39 ± 2		160	35 ± 2
14.01	[3/2]	1	0.57 ± 0.06	33 ± 2		150	59 ± 3
19.65							45 ± 2
19.86							62 ± 3
23.39							73 ± 3
23.54							13 ± 2
24.19	[3/2]	1	6.71 ± 0.34	24 ± 3		802	48 ± 6
26.17							39 ± 3
29.19							60 ± 4
29.81							7 ± 2
30.78	1/2	0	207.2 ± 2.1	54 ± 20	1180 ± 12		54 ± 20
31.21	[1/2]	1	3.55 ± 0.54	25 ± 3		290	25 ± 3
32.58							4 ± 1
34.49							3 ± 1
40.18	3/2	1	89.6 ± 3.4	35 ± 12		5180	69 ± 24
42.89							33 ± 4
43.74							32 ± 40
47.42							9 ± 3
47.53							64 ± 6
48.66							34 ± 4
49.49	[3/2]	1	6.1 ± 1.3	30 ± 3		263	59 ± 6
50.34	1/2	0	270.2 ± 4.2	28 ± 14	1204 ± 19		28 ± 14
50.35							73 ± 10
53.25	[3/2]	1	12 ± 2	24 ± 3		470	47 ± 5
54.94							20 ± 5
55.19	[1/2]	1	42.8 ± 3.2	21 ± 5		1590	21 ± 5
58.92							218 ± 15
59.34	1/2	0	73.2 ± 3.6	53 ± 18	300 ± 15		53 ± 18
60.67	[3/2]	1	11.0 ± 2.1	30 ± 4		360	59 ± 7
61.4							82 ± 8
62.77	3/2	1	114.4 ± 8.4	31 ± 14		3550	61 ± 28
66.66							10 ± 4

Analysis for capture cross section

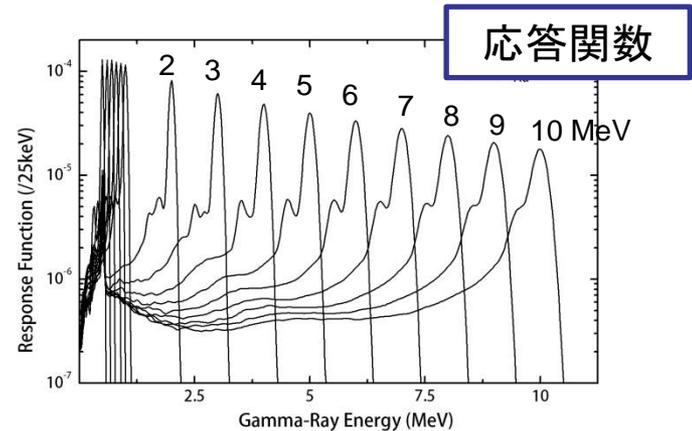


波高重み法

捕獲反応でカスケードで放出される即発ガンマ線のエネルギーの総和は、中性子束縛エネルギー B_n と入射中性子エネルギー E_n の和に等しいという関係を用いる。分岐比はいらない。

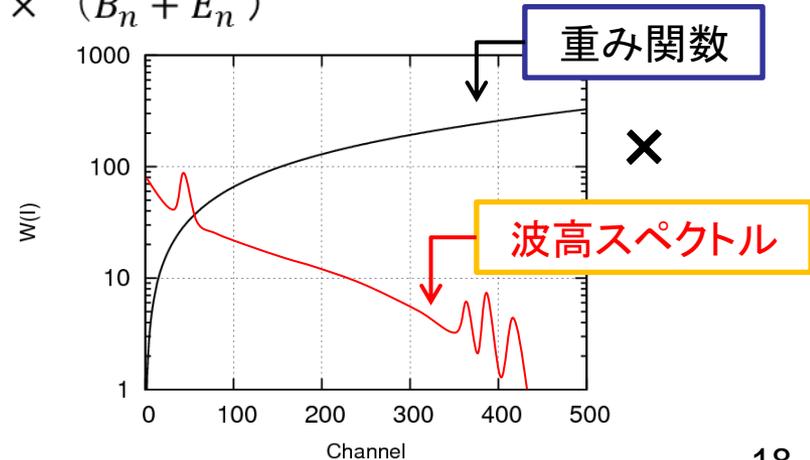
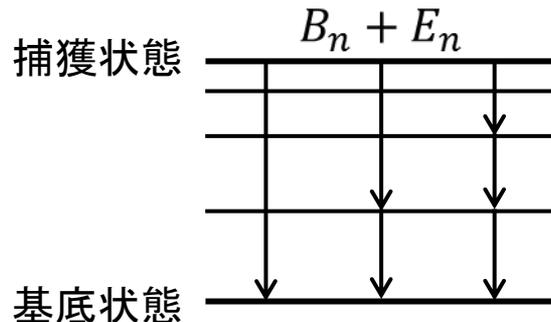
• 検出器の応答関数

- モンテカルロシミュレーション
- 検出器、遮蔽体、サンプルの模擬
- エネルギーごとの応答関数



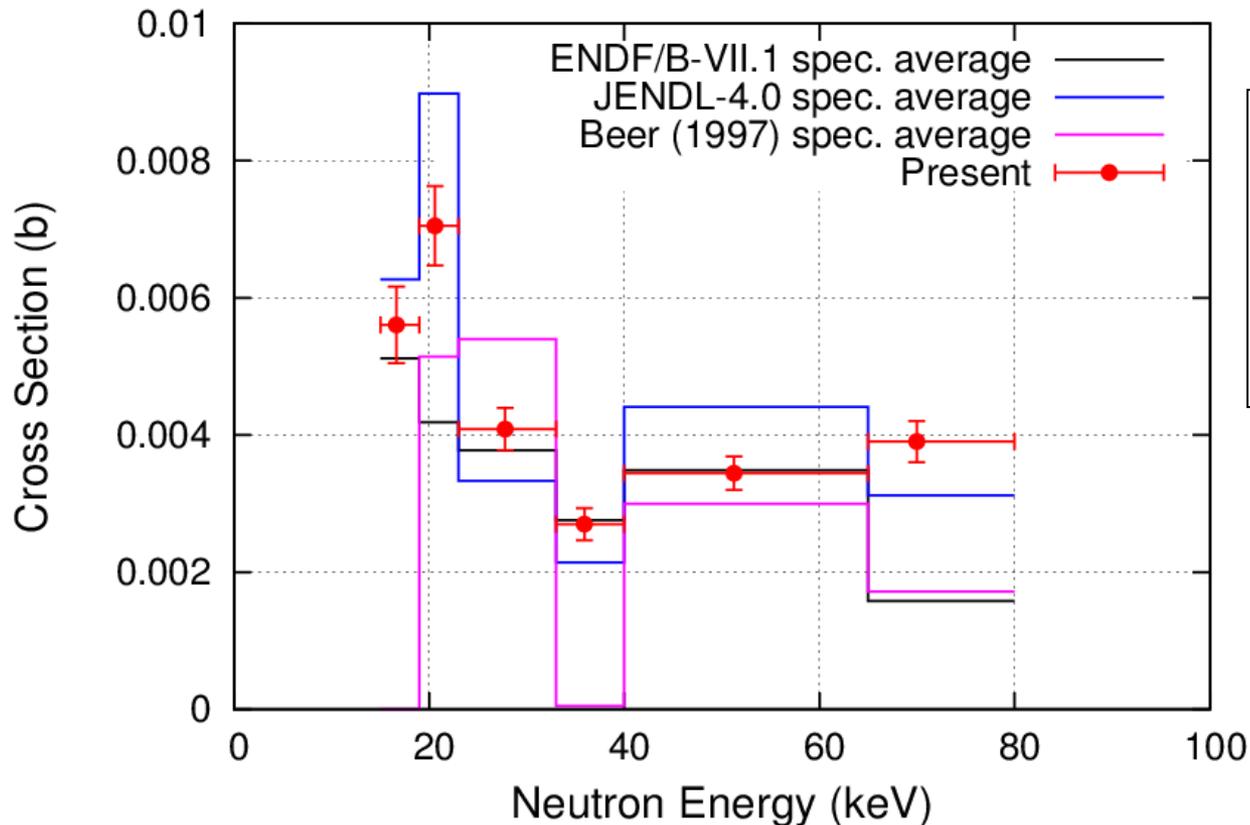
• 重み関数 $W(I)$

- 定義 $\sum_I W(I) \times (\text{応答関数}) = (\text{ガンマ線のエネルギー})$
- $\sum_I W(I) \times (\text{波高スペクトル}) = (\text{捕獲反応数}) \times (B_n + E_n)$

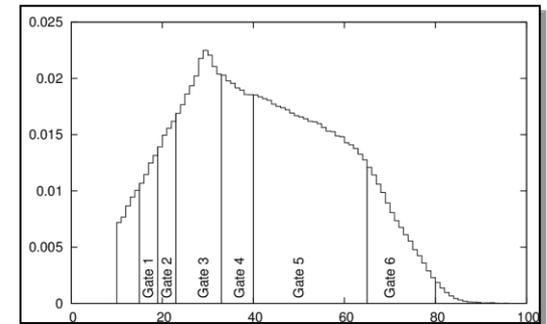


Comparison

- Spectrum averaged cross sections were calculated for ENDF/B-VII.1, JENDL-4.0.
- The Beer's experiments give only capture kernels. Average cross sections were calculated from resonance capture areas.

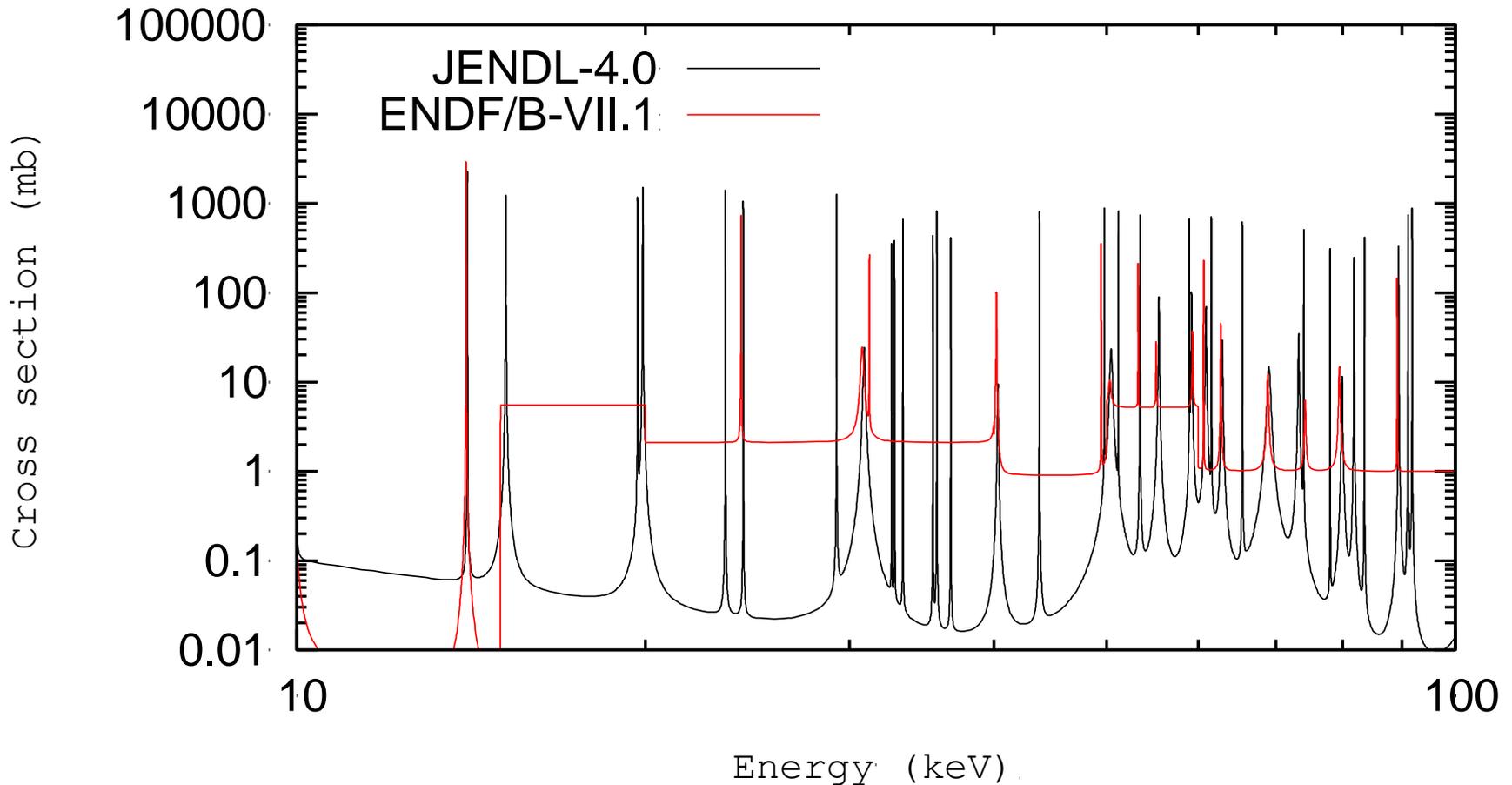


Neutron spectrum



Evaluation cross sections

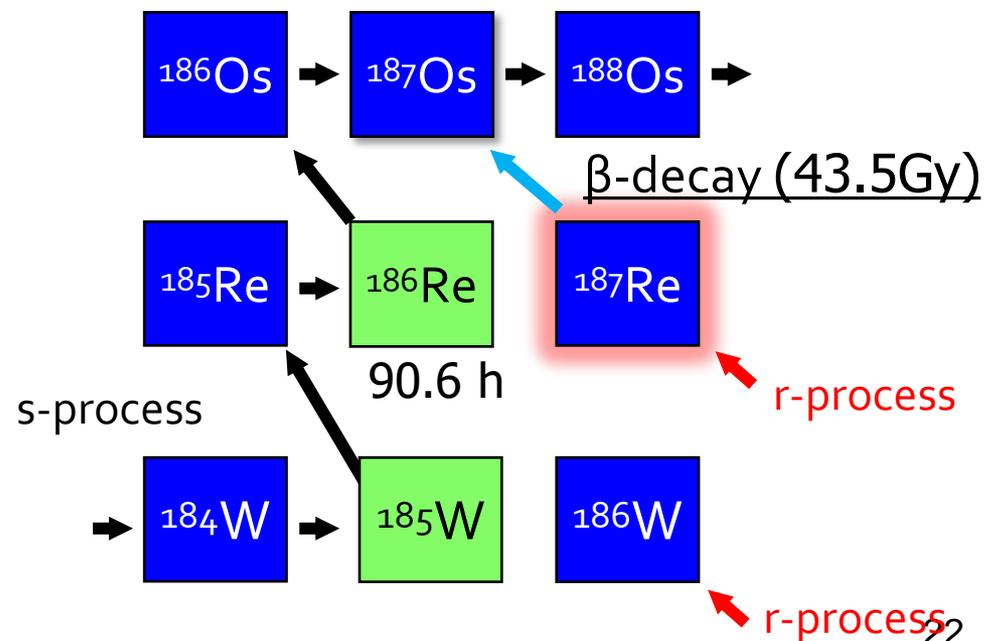
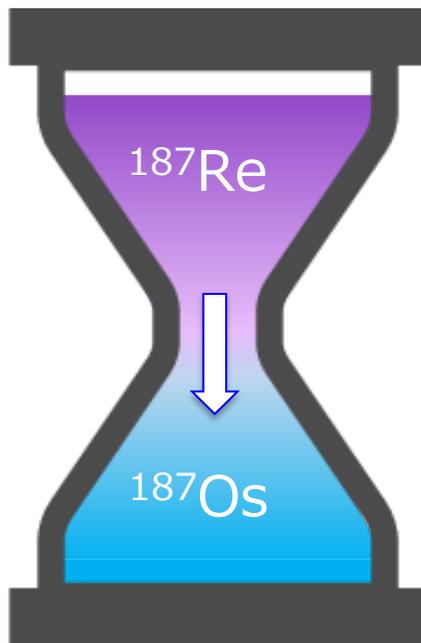
Instead of adopting resonance parameters, ENDF/B-VII.1 used background cross sections to reproduce the previous activation measurements.



^{185}Re の中性子捕獲断面積測定
- 放射化法による測定 -

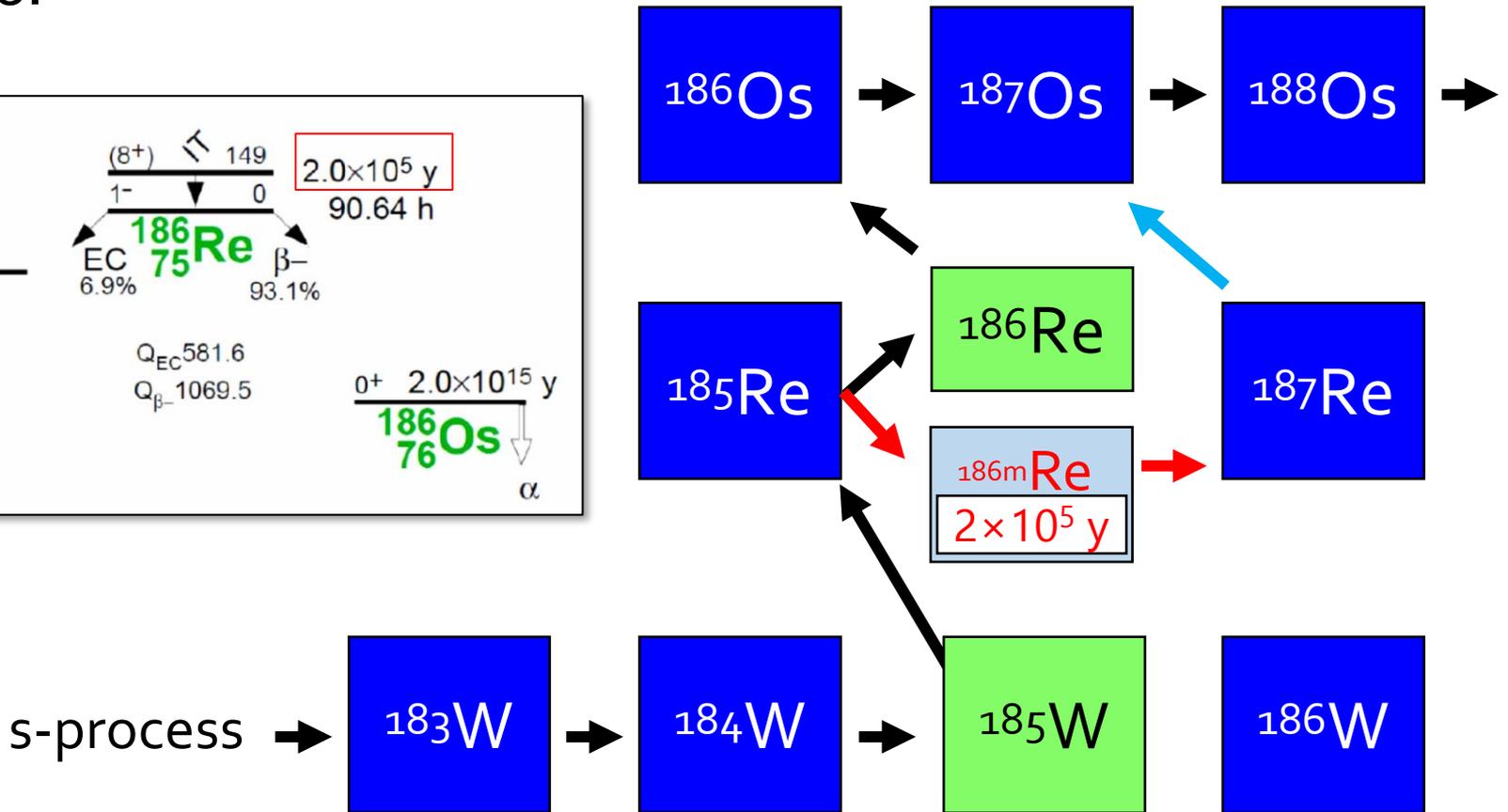
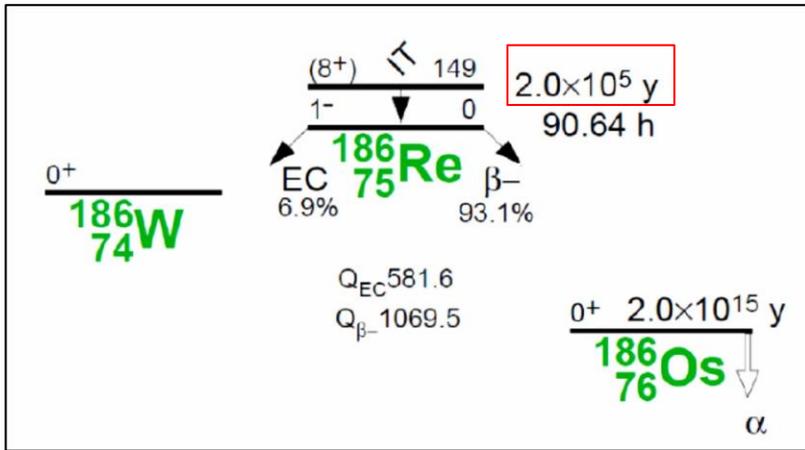
$^{187}\text{Re}/^{187}\text{Os}$ Nucleo-Cosmochronometer

- ◆ Re-187 is primarily considered to be created only in the *r*-process. Re-187 has a long half-life of 43.5 Gy.
- ◆ Decay of ^{187}Re to ^{187}Os can be used to determine when the *r*-process occurred (cosmochronometer).



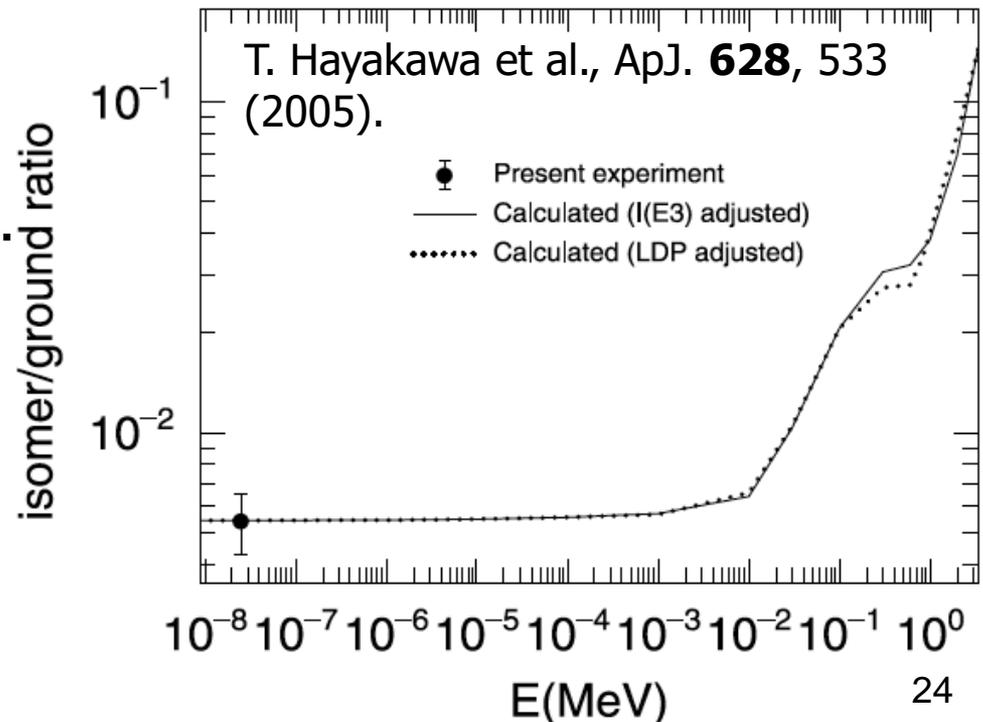
Isomer State of ^{186}Re

The s-process can reach ^{187}Re through an isomer state of ^{186}Re .



Isomer/Ground Ratio in $^{185}\text{Re}(n,\gamma)^{186}\text{Re}$

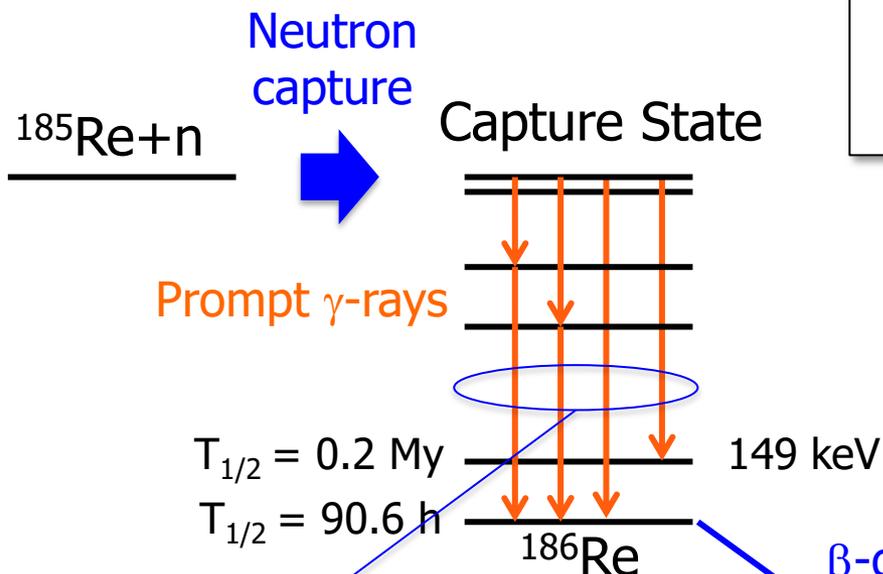
- ◆ To use ^{187}Re as a cosmochronometer, the isomer production ratio in neutron capture of ^{185}Re is necessary.
- ◆ The isomer/ground ratio is $0.54 \pm 0.11\%$ at the thermal energy (Hayakawa 2005).
- ◆ Theoretical calculations predict that the ratio increases with the incident neutron energy.
- ◆ Experimental data of the ratio in keV energy region is important.



Present Experiments - Goals

- ◆ To measure neutron capture cross section of ^{185}Re using both the TOF and activation methods.
- ◆ To compare results from the two different experimental methods.

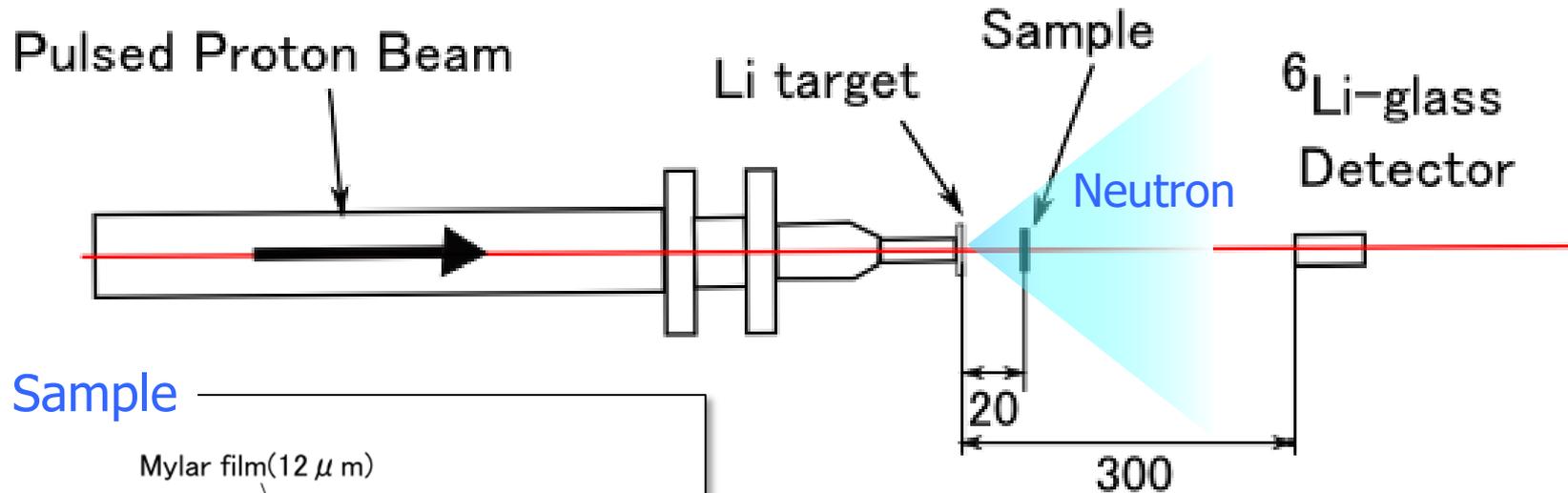
$$\sigma_m = \sigma_{\text{tot}} - \sigma_{\text{gnd}}$$



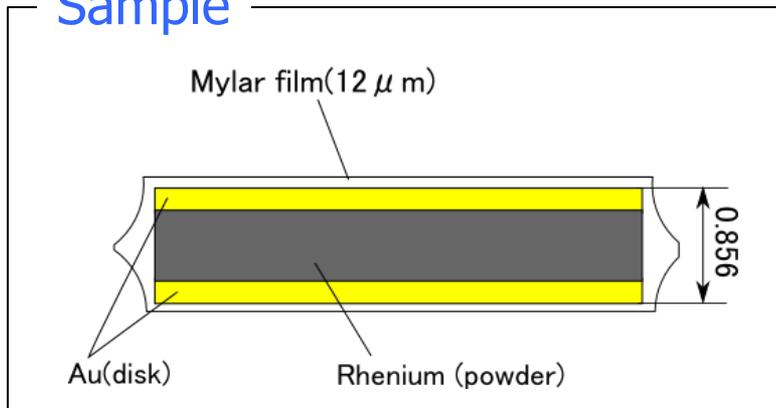
Activation experiments:
Decay γ -rays detected.
Partial cross section to
ground state of ^{186}Re σ_{gnd}

TOF experiments:
Prompt γ -rays detected.
Total capture cross section σ_{tot}

Activation Experimental Setup



Sample

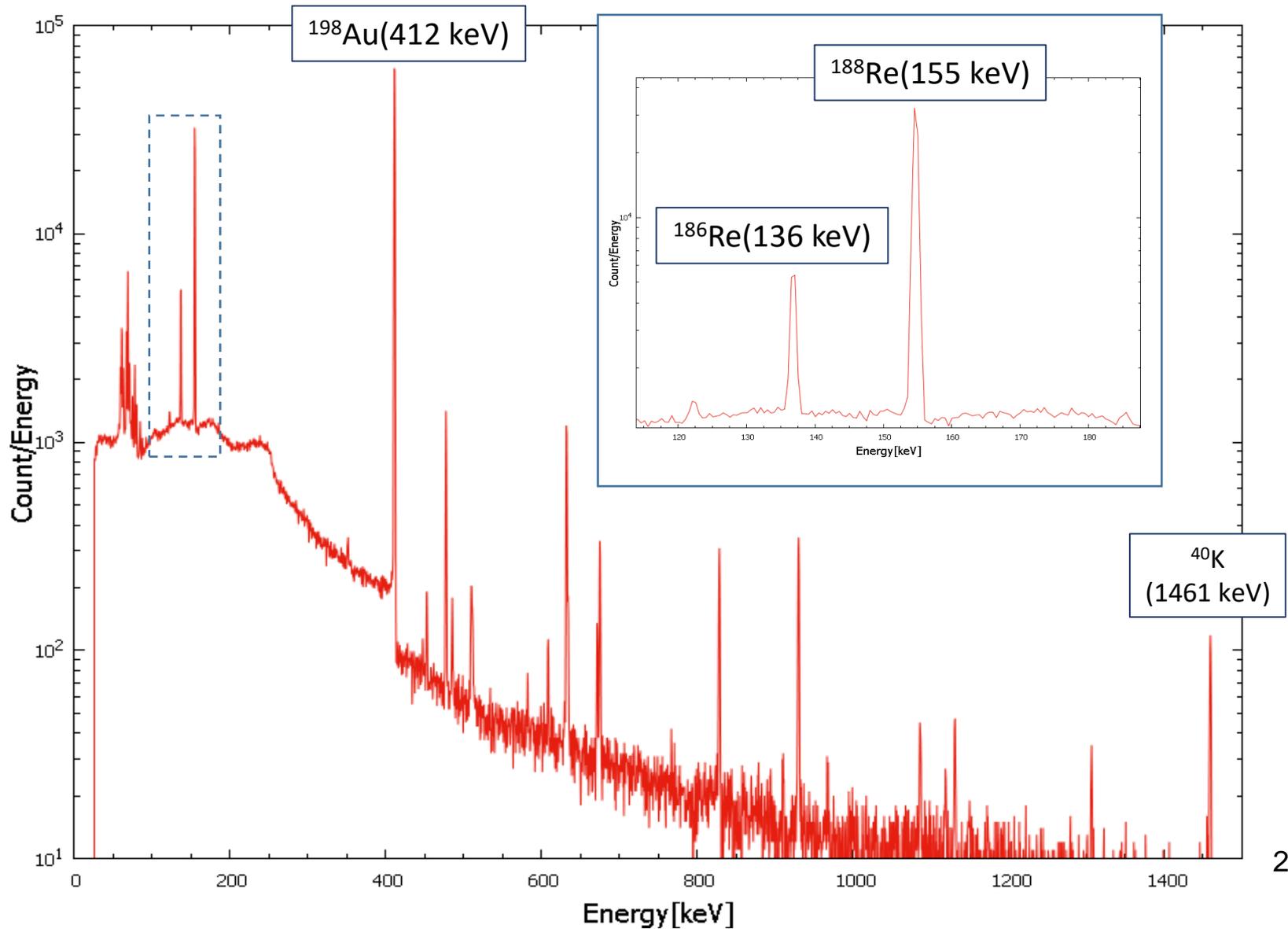


Irradiation : 24 h
Measurement : 1 week

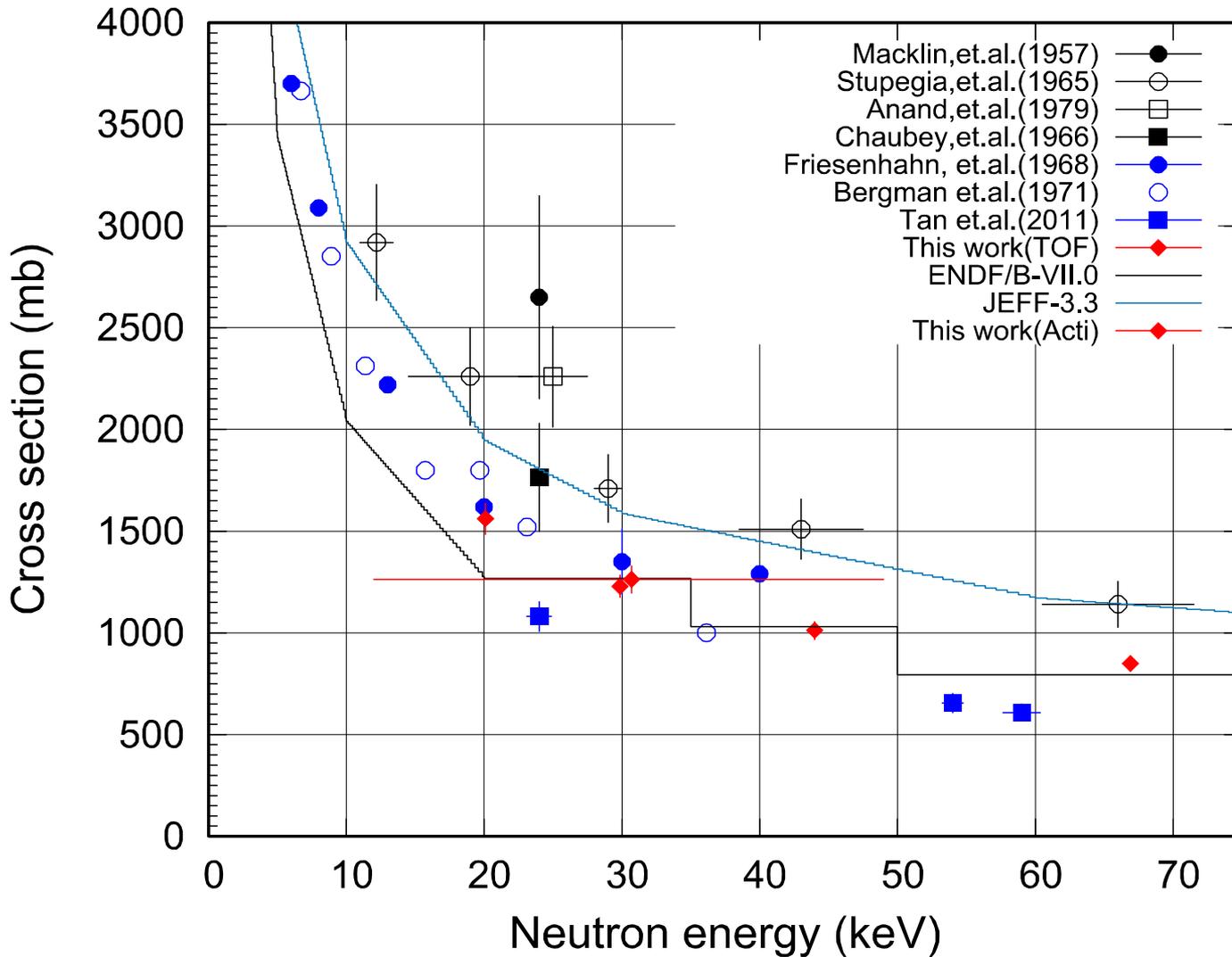
Measurement

Shielded HP Ge detector
Type: Coaxial
Diameter: 59.1 mm
Length: 59.8 mm
Energy resolution:
0.85 keV @ 122 keV
1.85 keV @ 1.33 MeV

Pulse Height spectrum with HP Ge

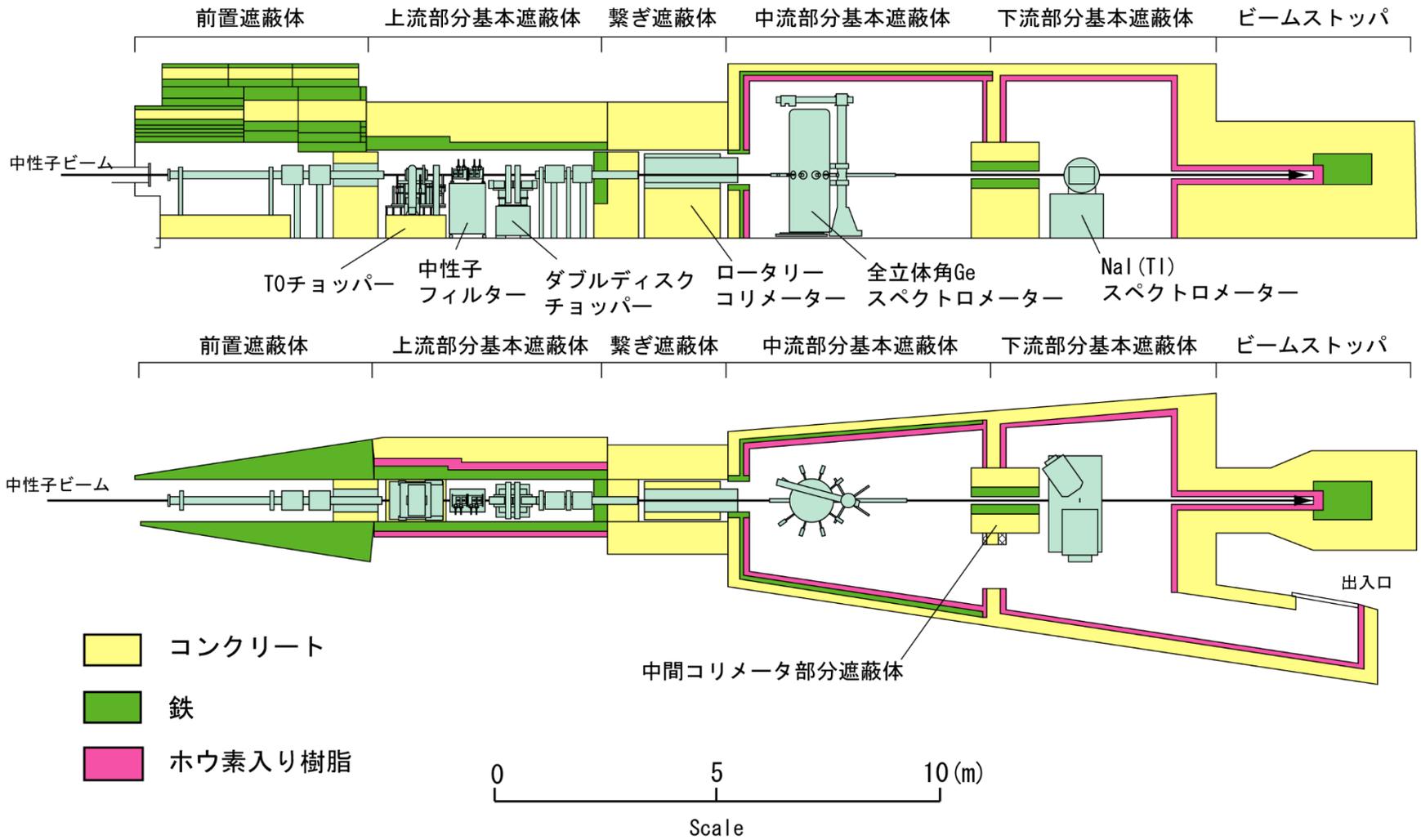


Capture Cross Section



J-PARCでの測定

実験施設 ANNRI @ J-PARC



Nd-142の測定

PHYSICAL REVIEW C 91, 037603 (2015)

Misassigned neutron resonances of ^{142}Nd and stellar neutron capture cross sections

Tatsuya Katabuchi,^{1,*} Taihei Matsubishi,^{1,†} Kazushi Terada,^{1,2} Masayuki Igashira,¹ Motoharu Mizumoto,¹ Kentaro Hirose,² Atsushi Kimura,² Nobuyuki Iwamoto,² Kaoru Y. Hara,^{2,3} Hideo Harada,² Jun-ichi Hori,⁴ Takashi Kamiyama,³ Koichi Kino,³ Fumito Kitatani,² Yoshiaki Kiyonagi,^{3,‡} Shoji Nakamura,² and Yosuke Toh²

¹*Research Laboratory for Nuclear Reactors, Tokyo Institute of Technology, Meguro-ku, Tokyo 152-8550, Japan*

²*Japan Atomic Energy Agency, 2-4 Shirakata Shirane, Tokai, Ibaraki 319-1195, Japan*

³*Faculty of Engineering, Hokkaido University, Kita 13 Nishi 8, Kita-ku, Sapporo 060-8628, Japan*

⁴*Research Reactor Institute, Kyoto University, 2-1010, Asashiro Nishi, Kumatori-cho, Sennan-gun, Osaka 590-0494, Japan*

(Received 26 February 2015; published 20 March 2015)

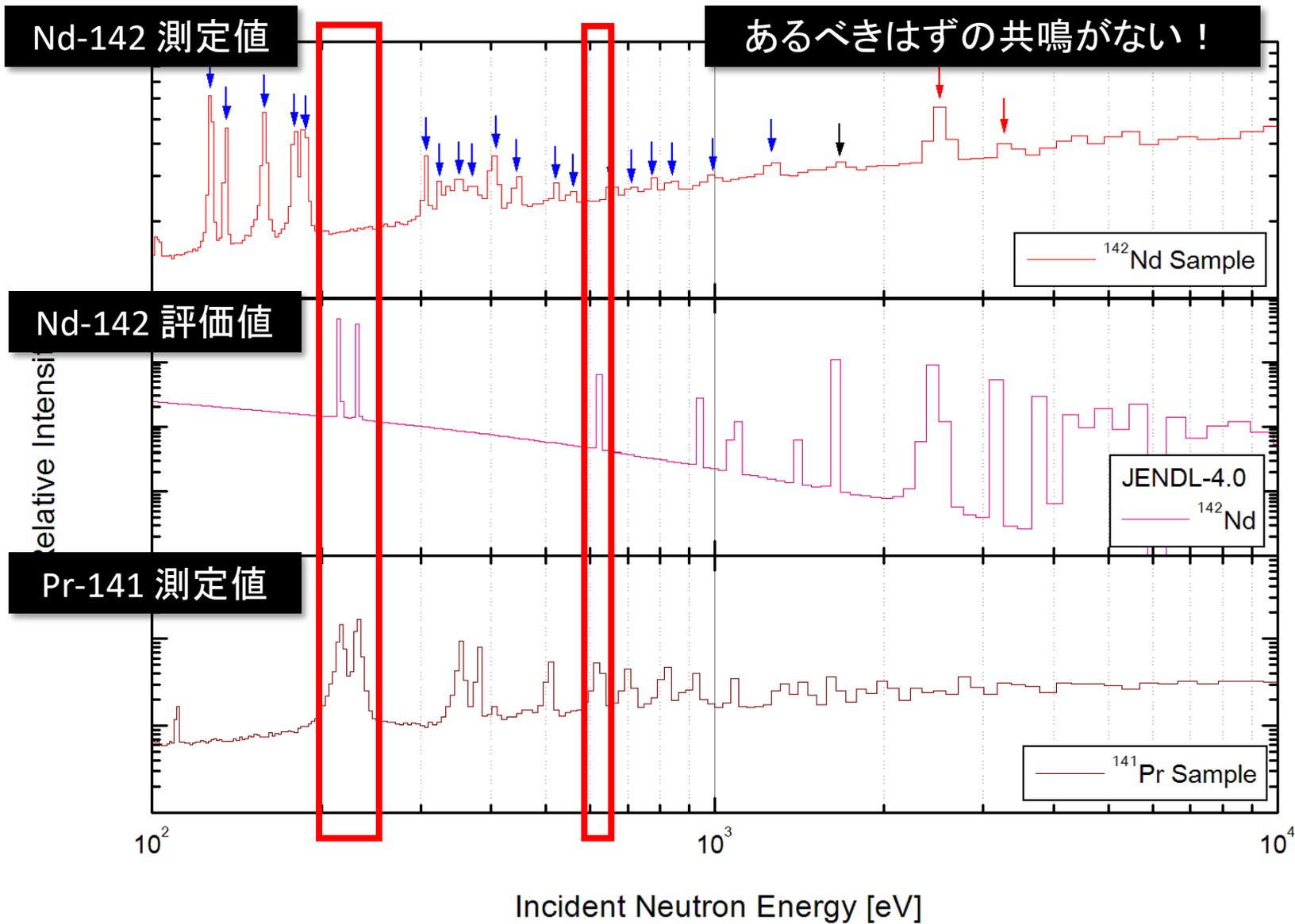
Time-of-flight spectra of the neutron capture events of ^{142}Nd were measured using a spallation neutron source at the Japan Proton Accelerator Research Complex. The first six resonances of ^{142}Nd reported in a previous work were not observed. The experimental results and cross-search of resonance energies in nuclear data libraries suggested that resonances of the impurity nuclide ^{141}Pr have been mistakenly assigned as ^{142}Nd in the previous experiment. To investigate the impact of the nonexistence of the resonances on the *s*-process nucleosynthesis model, the Maxwellian averaged neutron capture cross sections with and without the misassigned resonances were compared.

DOI: [10.1103/PhysRevC.91.037603](https://doi.org/10.1103/PhysRevC.91.037603)

PACS number(s): 25.40.Ny, 25.40.Lw, 28.20.Np, 26.20.Kn

T. Katabuchi *et al.*, Physical Review C 91, 037603 (2015)

TOFスペクトル



JENDL-4の共鳴パラメータ ^{142}Nd と ^{141}Pr

評価値にある1.5keVまでの6本の共鳴は、Pr-141である可能性が高い。過去の実験が、不純物のPr-141によるものをNd-142として同定してしまった。

Resonance energy of ^{142}Nd (eV)	This work	Resonance energy of ^{141}Pr (eV)
218.6±0.2	×	218.7±0.3
235.0±0.2	×	235.2±0.3
636.4±0.6	×	635.8±0.5
956.8±1.0	?	956.8±1.0
1119±1	?	1119.5±1.0
1485±2	?	1484.0±1.5
1687±2	?	
2431.0±2.4	?	
2528.5±2.5	○	

2 keV以下の共鳴パラメータ: 唯一の測定

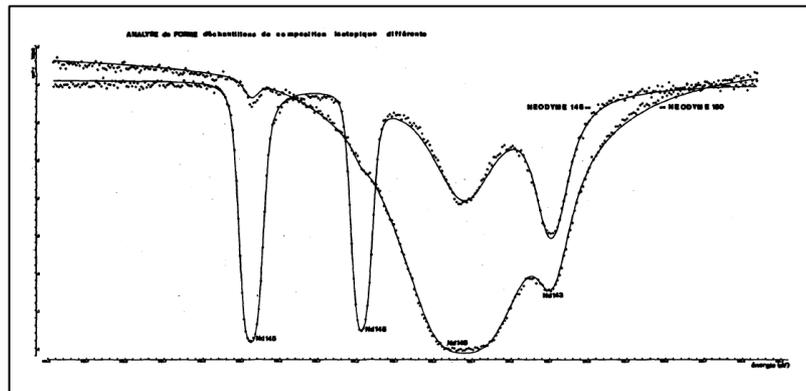
- H. Tellier, Report Note No. 1459, Centre d'Etudes Nucleaires, France, 1971

- 透過実験

60 MeV Saclay linac

- Ndの7核種の同位体濃縮試料を使用

- 化学的不純物については記載なし



- 共鳴解析

Echan- tillon	A	142 %	143 %	144 %	145 %	146 %	148 %	150 %	Quantité (g)
142 Nd		95,0	2,7	1,5	0,3	0,3	0,1	0,1	68,3
143 Nd		2,9	83,2	11,8	1,0	0,9	0,1	0,1	30,0
144 Nd		0,8	2,2	91,5	4,3	1,2	-	-	50,0
145 Nd		1,0	0,9	7,9	70,8	18,8	0,4	0,2	20,0
146 Nd		0,5	0,5	1,3	1,2	94,7	1,6	0,2	29,0
148 Nd		1,1	0,9	2,0	1,1	3,5	89,4	2,0	40,0
150 Nd		1,2	1,0	2,2	1,1	2,5	3,5	88,5	20,0

^{89}Y の測定

Eur. Phys. J. A (2021) 57:4
<https://doi.org/10.1140/epja/s10050-020-00320-8>

THE EUROPEAN
PHYSICAL JOURNAL A



Letter

Discovery of a new low energy neutron resonance of ^{89}Y

Tatsuya Katabuchi^{1,a}, Yosuke Toh², Motoharu Mizumoto¹, Tatsuhiro Saito¹, Kazushi Terada^{1,3}, Atsushi Kimura², Shoji Nakamura², Huang Minghui², Gerard Rovira², Masayuki Igashira¹

¹ Laboratory for Advanced Nuclear Energy, Institute of Innovative Research, Tokyo Institute of Technology, 2-12-1, Ookayama, Meguro-ku, Tokyo 152-8550, Japan

² Japan Atomic Energy Agency, 2-4 Shirakata Shirane, Tokai, Ibaraki 319-1195, Japan

³ Present address: Institute for Integrated Radiation and Nuclear Science, Kyoto University, 2-1010 Asashironishi, Kumatori, Sennan-gun, Osaka 590-0494, Japan

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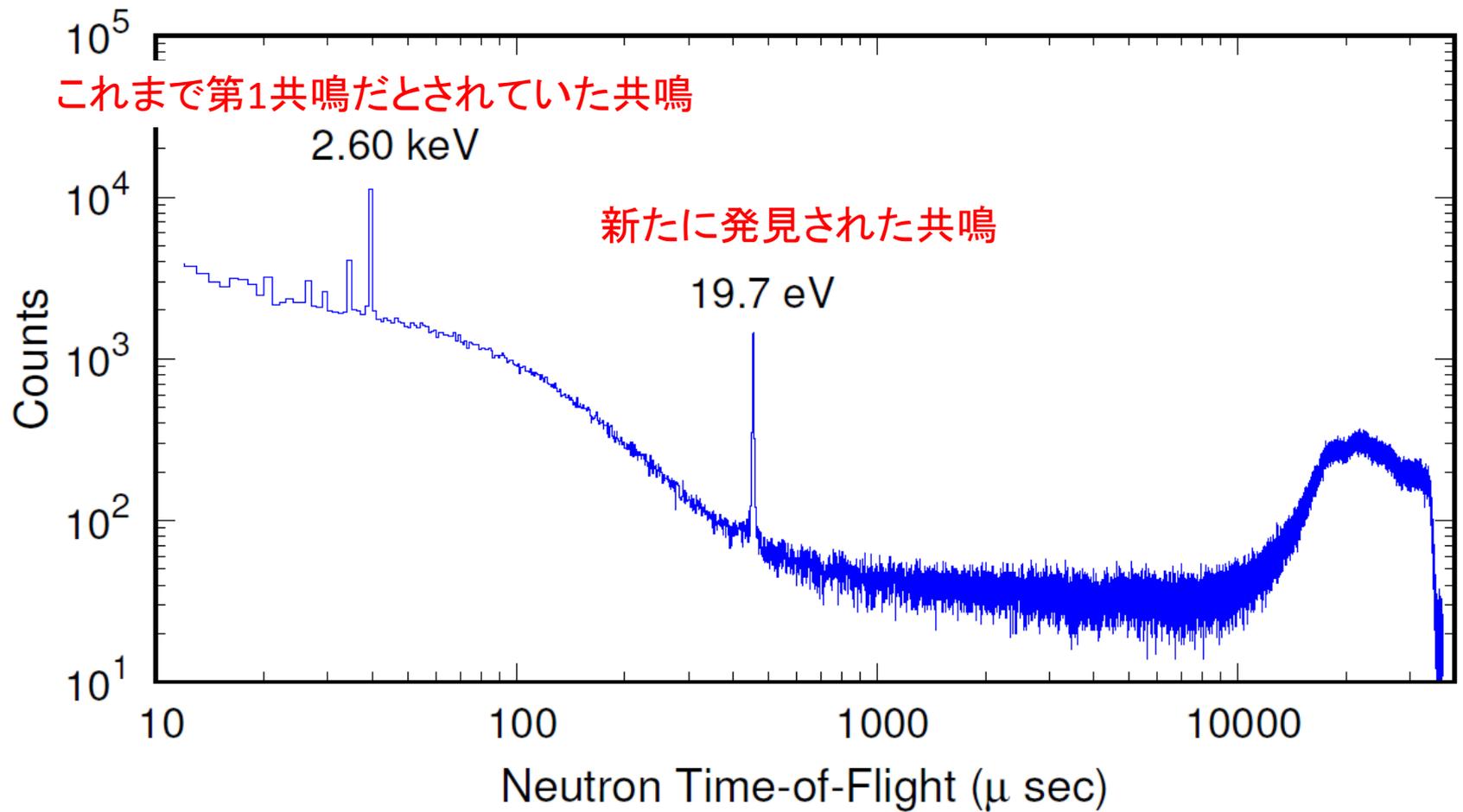
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Abstract A new neutron resonance of ^{89}Y was discovered by time-of-flight (TOF) neutron capture experiments using a pulsed neutron beam from a spallation neutron source of the Japan Proton Accelerator Research Complex. The observed resonance energy was 19.7 eV that is much lower than the lowest energy (2.60 keV) of the previous reported reso-

In recent years, demands of neutron resonance data increased in other research fields. For example, an advanced prompt γ -ray analysis (PGA) combined with neutron time-of-flight (TOF) technique (TOF-PGA) has been developed [6]. The technique relies on a resonance database to increase the sensitivity of analysis of trace elements. Thus, this appli-

新しい低エネルギー共鳴の観測



まとめ

東工大ペレトロン加速器

- 飛行時間法による測定

例： ^{138}Ba

- 放射化法による測定

例： ^{185}Re

J-PARC/ANNRI

- 飛行時間法による共鳴測定

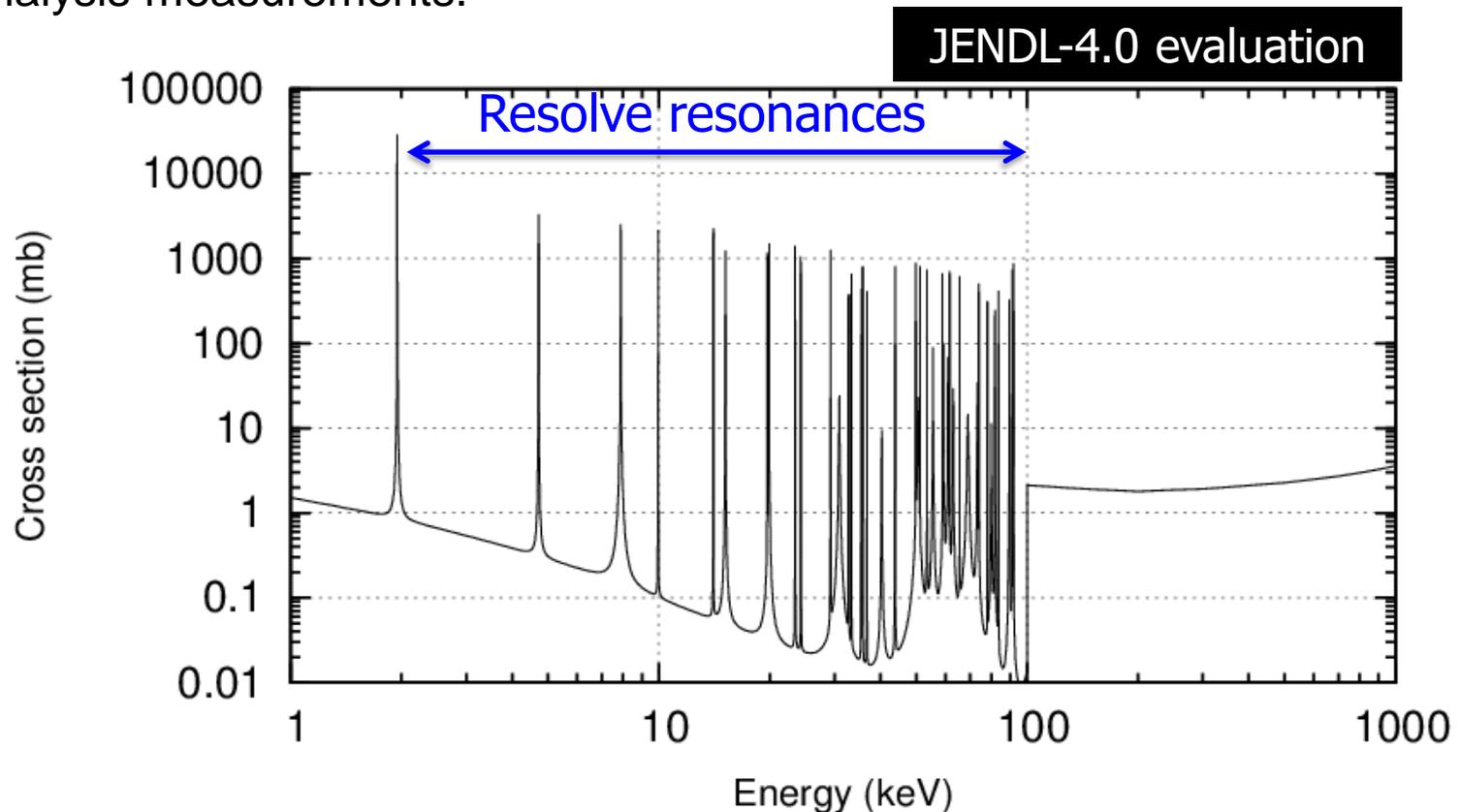
例： ^{142}Nd , ^{89}Y

最後に

東工大ペレトロン加速器の共同利用を進めています。興味のある方はぜひご連絡ください。

$^{138}\text{Ba}(n,\gamma)$ cross section in keV region

- $^{138}\text{Ba}(n,\gamma)$ reaction in the keV energy region is dominated by resolved resonances.
- Evaluated cross sections of ^{138}Ba in keV region are based on resonance analysis measurements.



Previous measurements in keV region

□ Resonance parameters

	Year	Facility	Type	Energy
Beer et al.	1997	Geel	Capture	0.65 – 200 keV
Brusegan et al.	1994	Geel	Transmission	0.65 – 195 keV
Mizumoto et al.	1988	JAEA	Transmission	0.65 – 63 keV
Musgrove et al.	1978	Oak Ridge	Capture	4.7 – 91 keV
Bilpuch et al.	1961	Duke	Transmission	31.7 – 192 keV

□ Average cross section

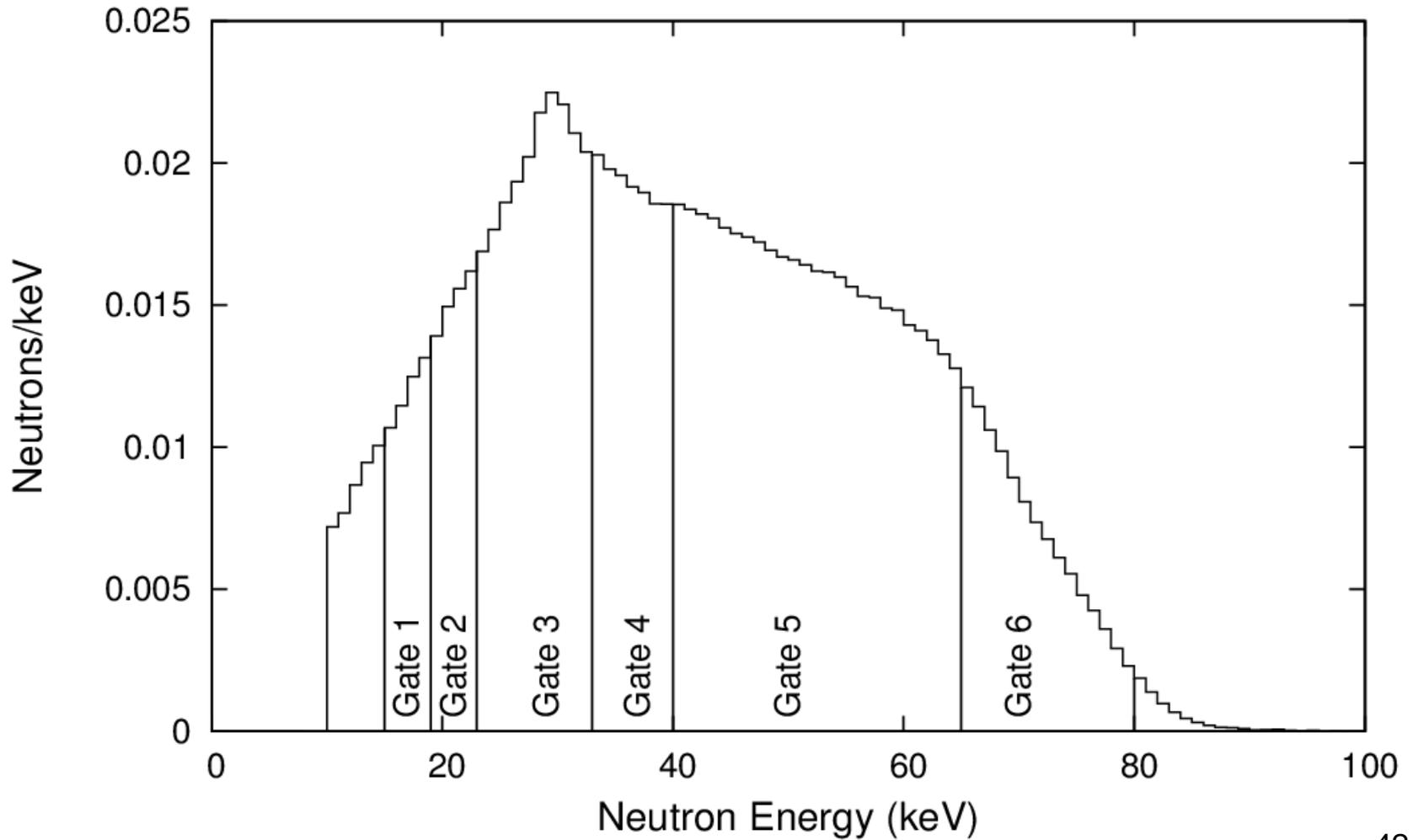
	Year	Facility	
Heil et al	2005	Karlsruhe	KT = 5 keV Maxwellian
Beer et al.	1980	Karlsruhe	KT = 23 keV Maxwellian

Present measurement

- The present method has less energy resolution but more sensitive to smaller cross section than resonance analysis.
- Resonance analysis cannot measure a contribution of the direct capture process.
- The gamma-ray spectra can be measured.

	Present	Beer (1997)	Musgrove (1978)
Source	Van de Graaf ${}^7\text{Li}(p,n){}^7\text{Be}$ reaction	Electron LINAC (γ,n) reaction	Electron LINAC (γ,n) reaction
Energy	15 – 80 keV	0.65 – 200 keV	0.65 – 91 keV
Flight length	12 cm	28.4 m, 58.6 m	40 m
Time resolution	3 ns	1 ns	2 ns
Energy resolution	12% @ 30 keV	0.02% @ 30 keV	0.02% @ 30 keV
Detector	NaI(Tl)	C_6D_6	C_6F_6
Quantity	Average cross section	Resonance area	Resonance area
γ -ray spectrum	○	×	×

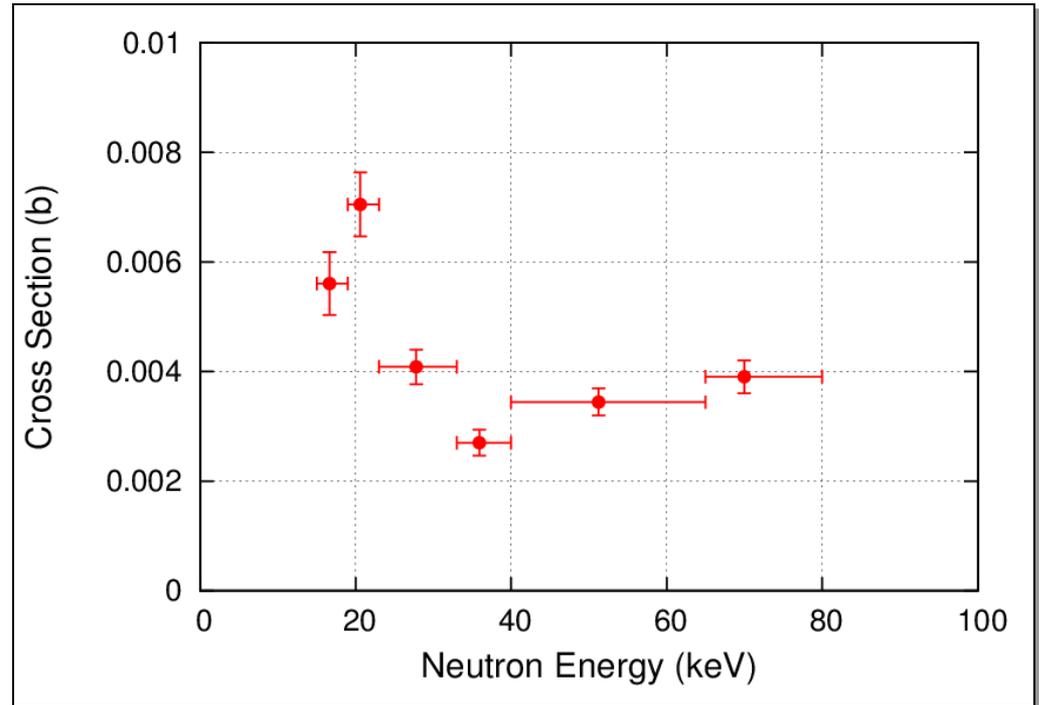
Incident Neutron Spectrum



Results

The $^{138}\text{Ba}(n,\gamma)^{139}\text{Ba}$ cross section was determined within uncertainties of 7 – 10%.

E_n (keV)	Cross section (mb)
15 - 19	5.61 ± 0.58 (10%)
19 - 23	7.05 ± 0.59 (8.3%)
23 - 33	4.09 ± 0.32 (7.7%)
33 - 40	2.70 ± 0.24 (8.7%)
40 - 65	3.45 ± 0.25 (7.2%)
65 - 80	3.91 ± 0.30 (7.7%)



飛行時間法による捕獲反応断面積の測定

● 結果と考察

- 誤差の主な要因は、標準断面積としていよ
うした核データの誤差、中性子輸送の補正
計数の誤差、統計誤差。
- 誤差データが与えられている過去の測定値
と比べ、誤差が最も小さい。
- TOF法による測定が行われていなかった高
いエネルギー領域も含まれる。

Table 2 各領域の断面積と誤差

Gate No.	Cross section [b]	Error [%]
1	1.561	5.38
2	1.229	4.85
3	1.012	4.58
4	0.849	4.71

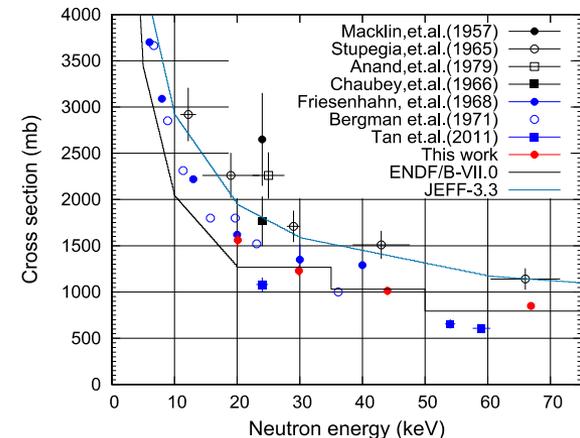


Fig 11 TOF測定の結果と過去の測定値

● 結果

- ^{185}Re の 捕獲断面積は、12-49keVの領域において、過去の測定値は今回の結果と一致している。
- 本研究のTOF測定の結果とも、誤差の範囲で一致。
- ^{187}Re の捕獲断面積は、過去の測定値と大きく異なる。
- 過去の測定のひとつは、本研究の結果に比べ、2倍程度の値を示している。
- 誤差の主な要因は、標準断面積の誤差、HPGe検出器の検出効率の誤差がある。

Table4 得られた断面積と誤差

生成核種	断面積	誤差
^{186}Re	1.263 b	5.27%
^{188}Re	0.560 b	5.26%

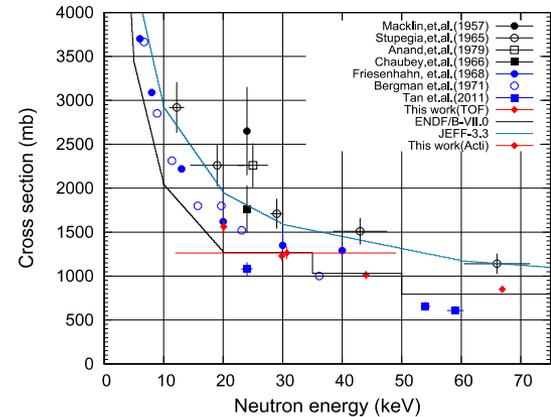


Fig.17 ^{185}Re の捕獲断面積

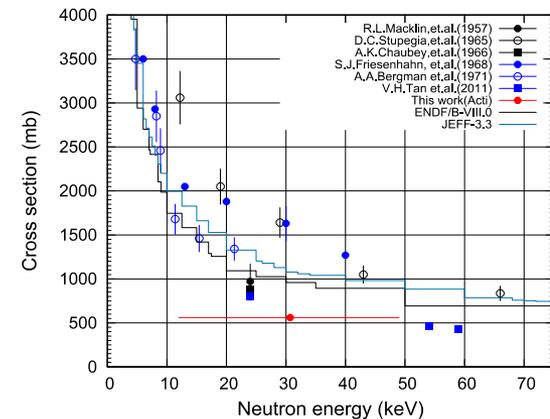


Fig.18 ^{187}Re の捕獲断面積