Current activities and future plans for nuclear data measurements at J-PARC

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原子力基礎工学研究センター
Nuclear Science and Engineering Center

Content

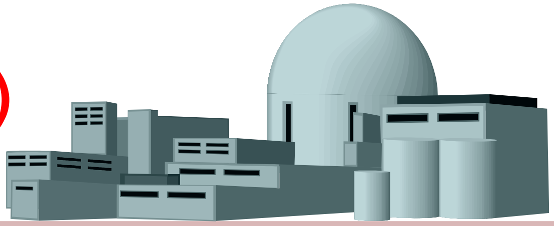
- Introduction
- Our Facility, “ANNRI”
- Example of Measurements
  - Experiment of $^{244\ \mathrm{and}\ 246}Cm$ (MAs)
  - Experiment of ^{112}Sn (stable isotope)
- Summary

Introduction(1)
~Motivation~

- To construct new types of reactors,
- To reduce High-Level radioactive Waste (Transmutation),
- To evaluate source terms from the Fukushima reactors,

Neutron Capture Cross Sections of
- **minor actinides (MAs) and**
- **long-lived fission products (LLFPs)**

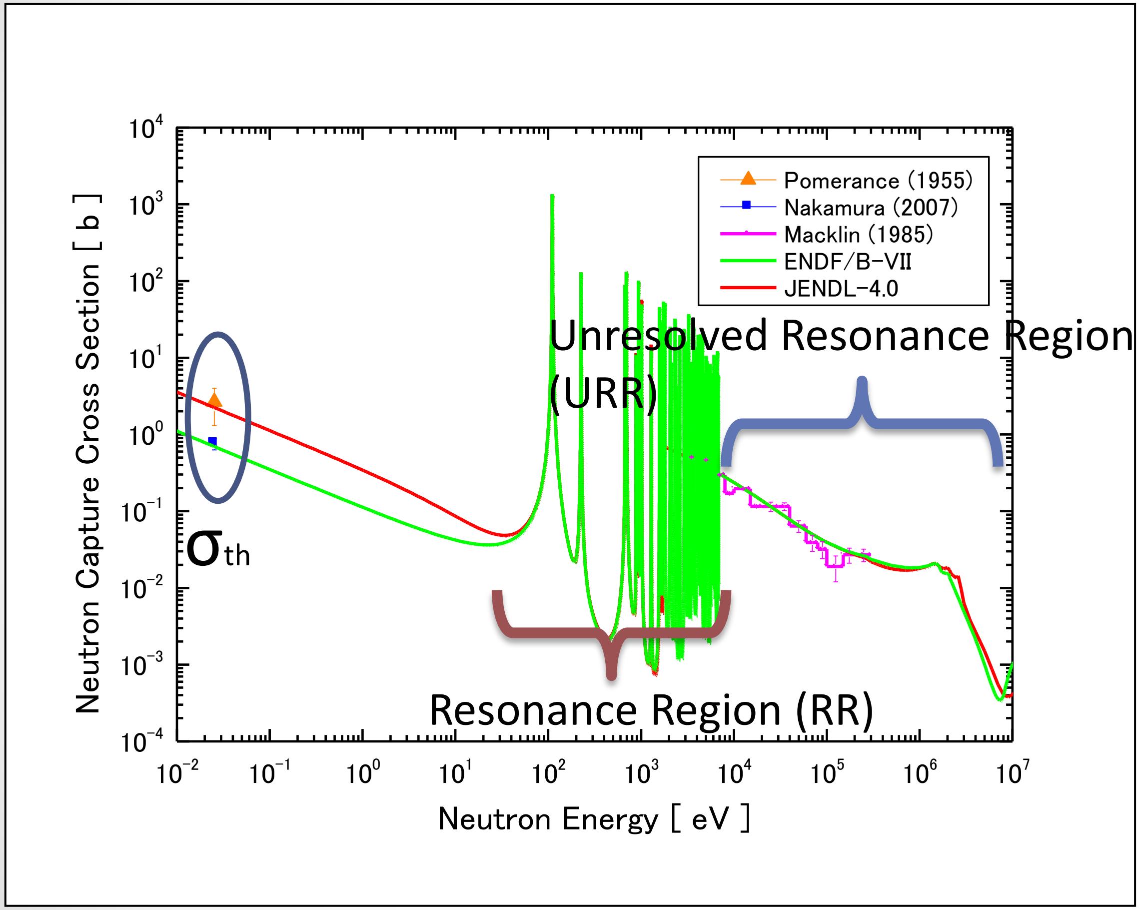
are very important.



However, Experimental data of MAs and LLFPs is **not sufficient** both in quality and in quantity.
For example…..

Introduction(2)

Present status of nuclear data (1)



Zr-93(LLFP)

σth
Num. of exp. data : 2
・Reactor Neutron (1955, 2007)

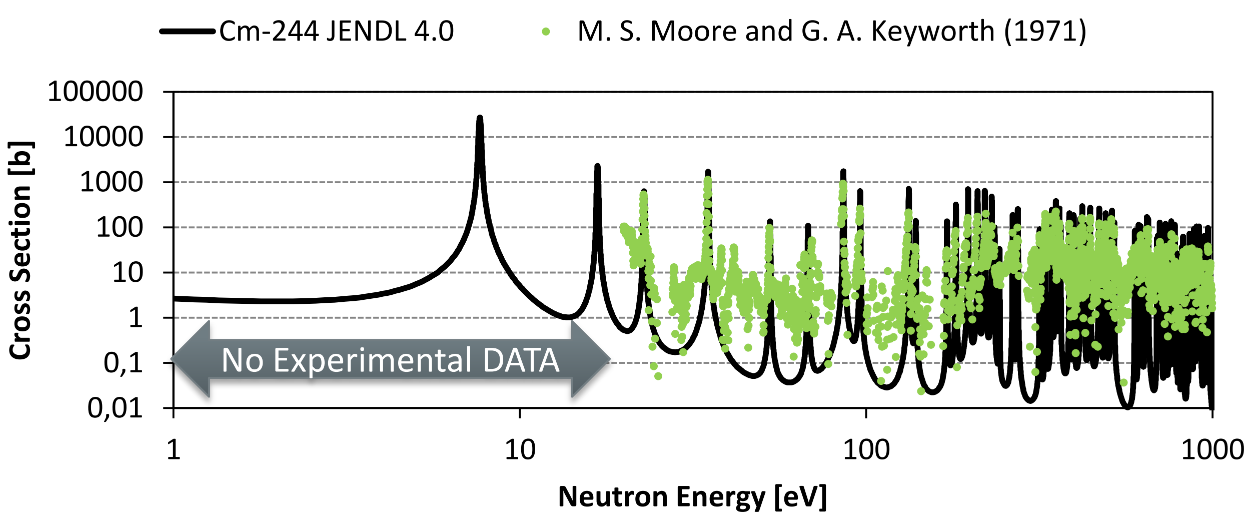
Unresolved Resonance Region
Num. of exp. data : 1
・TOF
ORELA (1985: C_6D_6)

Resonance Region
No experimental data

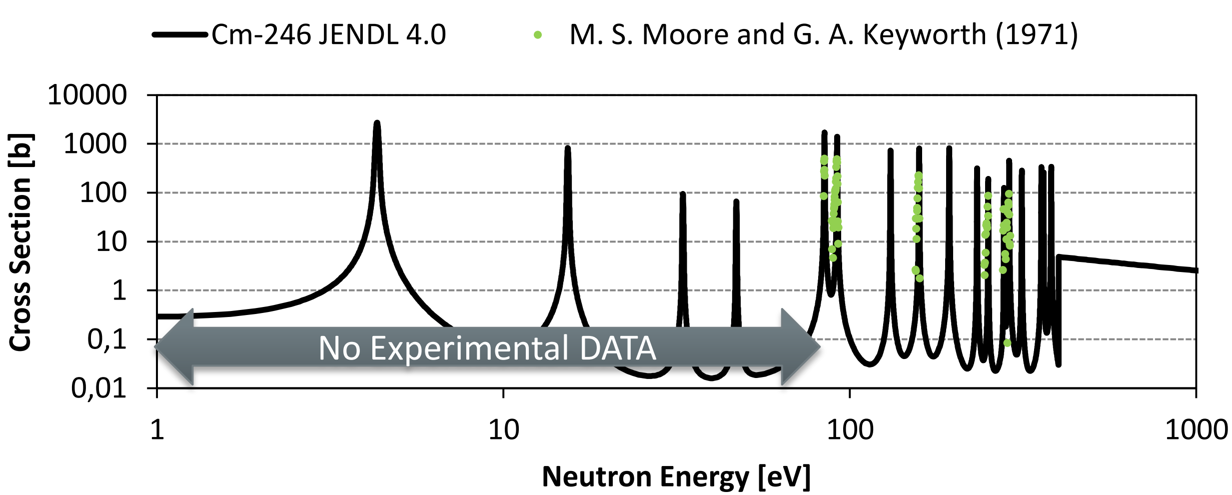
Resonance Region : Cross sections show sharp peaks and valleys due to resonances.
Unresolved Resonance Region : At higher energies, the resonances get so close together that they can no longer be resolved into separate lines.

Introduction(3)

Present status of nuclear data (2)



^{244}Cm (half-life: 18.1 years) and ^{246}Cm (4730 years) are among the most important MAs.

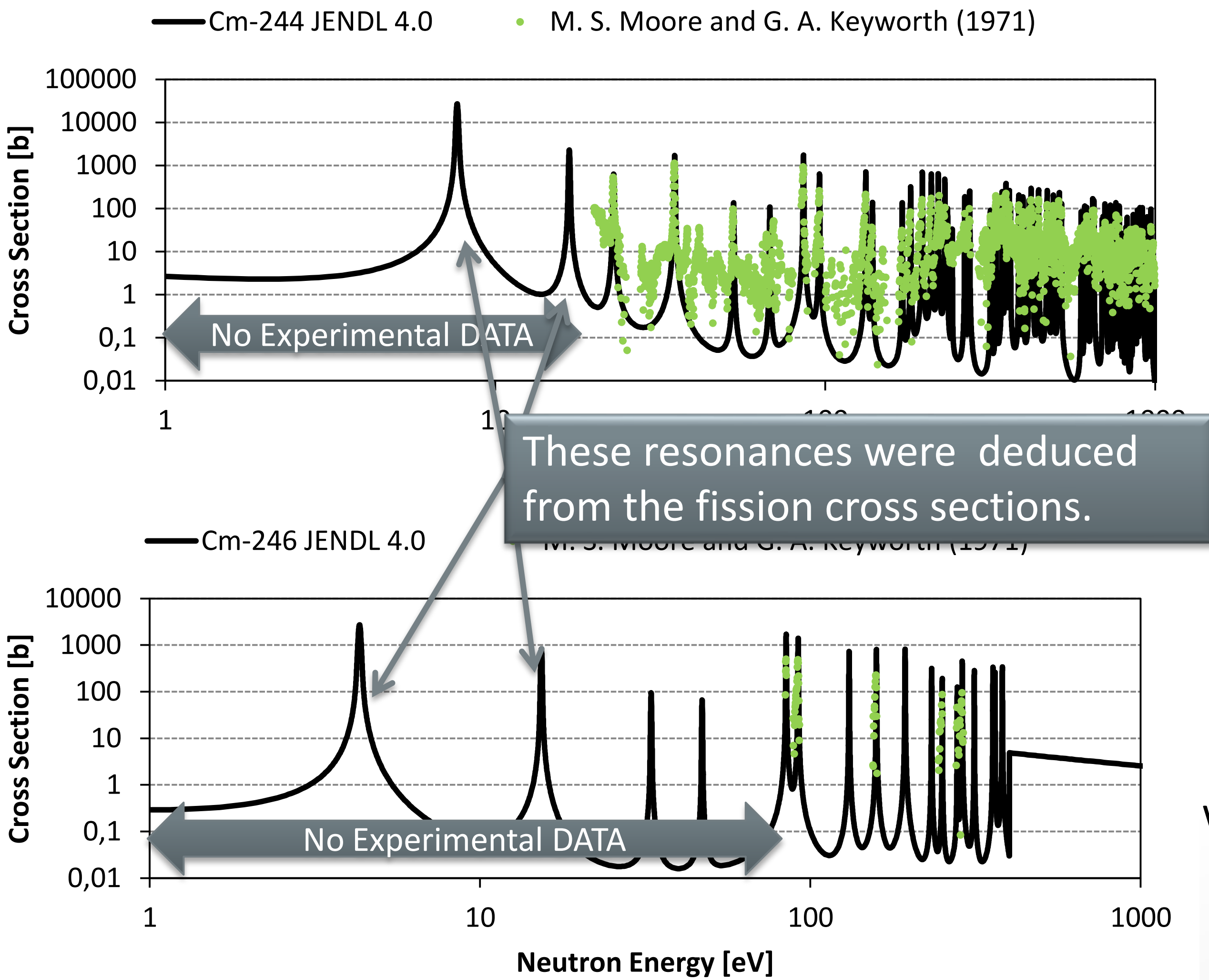


However, there is only one experimental data set.

This previous measurement was performed using a nuclear explosion in 1969.

Introduction(3)

Present status of nuclear data (2)



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However, there is only one experimental data set.

This previous measurement was performed using a nuclear explosion in 1969.

Introduction(4)

Present status of experimental data for MAs and LLFPs is not sufficient both in quality and in quantity.

This is because it is not easy to prepare enough amount of sample with a high purity.
Moreover, some MAs are high radio activities.

To overcome these problems

1) Intense neutron source
   → Small amount of sample can be used
   → Influence due to decay γ-rays can be reduced.
2) High energy resolution and high efficiency g-ray detector system was applied to the TOF measurement (For LLFPs)
   → Background due to impurities can be removed.

Introduction(5)

~Measurement Plan~

Sealed RI Samples

MA Samples

^{237}Np, ^{241}、^{243}Am,

^{244}Cm, ^{246}Cm

LLFP Samples

^{93}Zr, ^{99}Tc,

^{107}Pd, ^{129}I,

$^{135,137}Cs$

Stable Samples

$^{54, 56, 57}Fe$

$^{58, 60, 61, 62}Ni$

$^{74, 76, 77, 78, 80}Se$

$^{90, 91, 94, 96}Zr$

$^{104, 105, 108}Pd$

112, 115, 116, 117, 118, 119, 120, 122, ^{124}Sn, ^{127}I, ^{133}Cs, ^{142}Nd

$^{155,157}Gd$

Red: Already Published. Green: Published with preliminary results.

Blue: Already Measured. Black: Future Plan in few years (got Budget).

Introduction(6)

In this talk, as examples of our experiments, measurements for

- Cm-244, Cm-246, (MAs)
- Sn-112 (Stable)

will be demonstrated.

Cm-244,246

A. Kimura, et. al. , J,NST,49,(7),708 (2012)

Sn-112

A. Kimura, et al., Nuclear Data Sheets., 119, (2014) 150.

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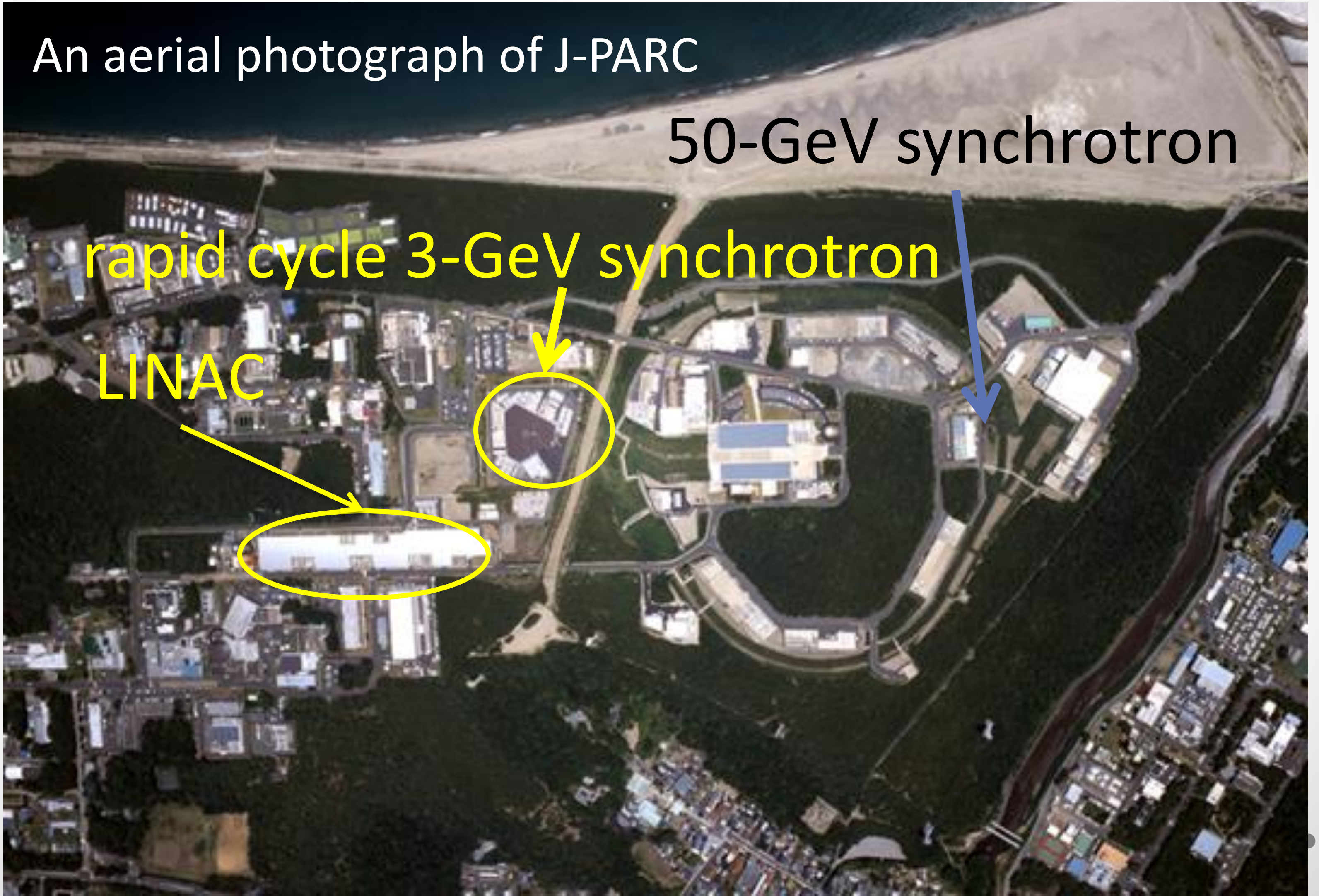
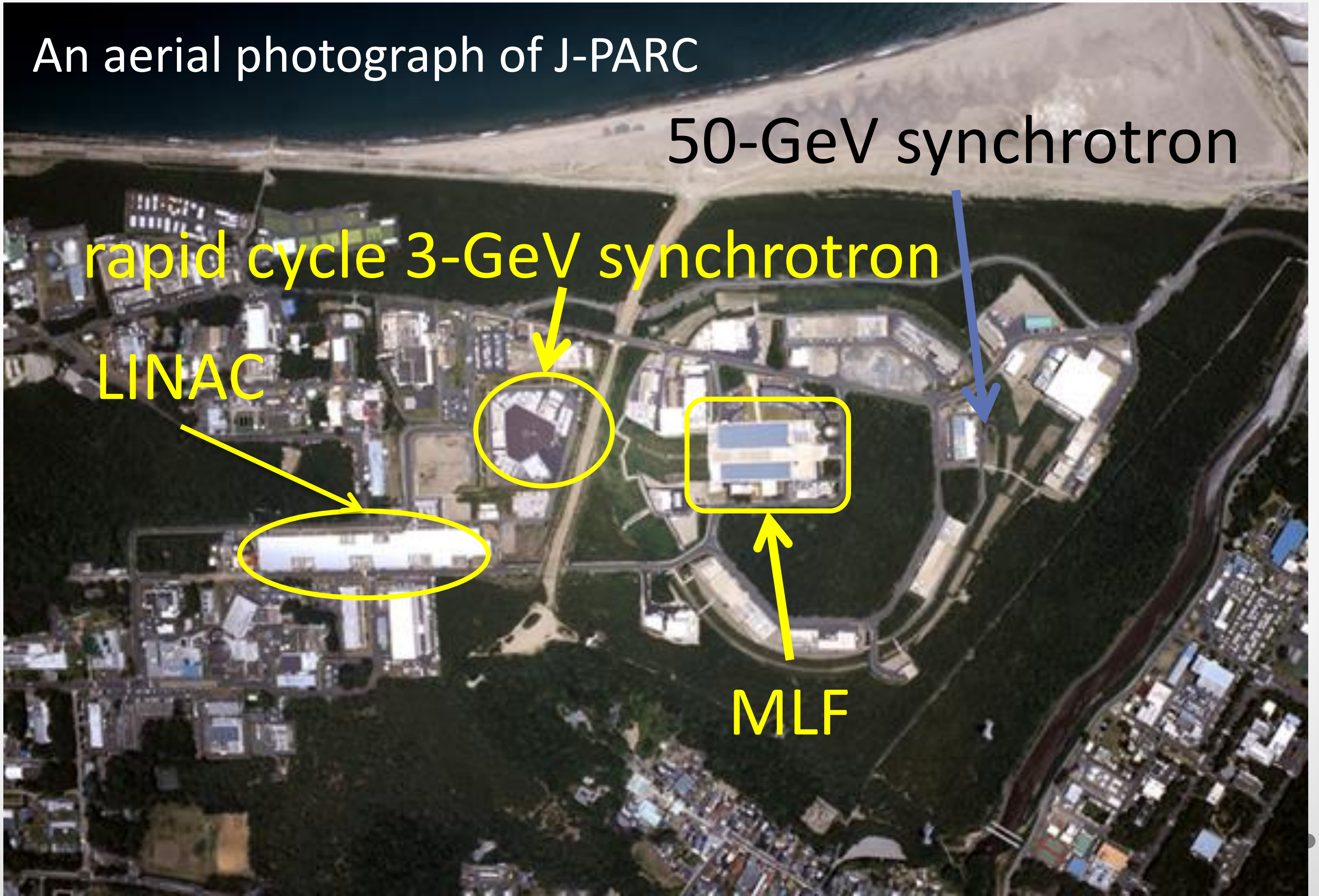
Our Facility, "ANNRI"
~ J-PARC~

Our Facility, “ANNRI” ~ J-PARC~



Our Facility, "ANNRI"
~ J-PARC~



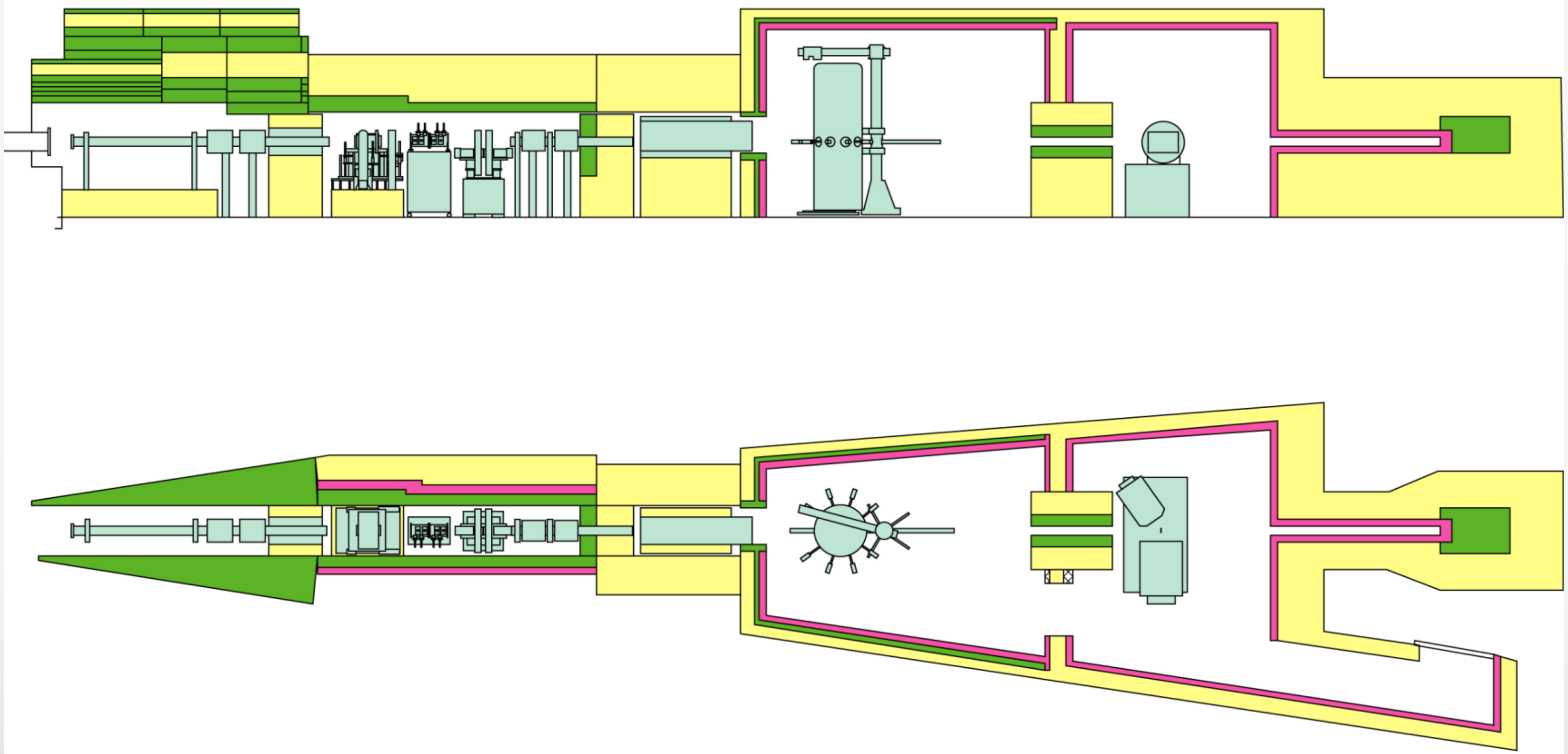
Our Facility, “ANNRI”
~ TOF facility ~

BL04: ANNRI

(Accurate Neutron-Nucleus Reaction measurement Instrument)

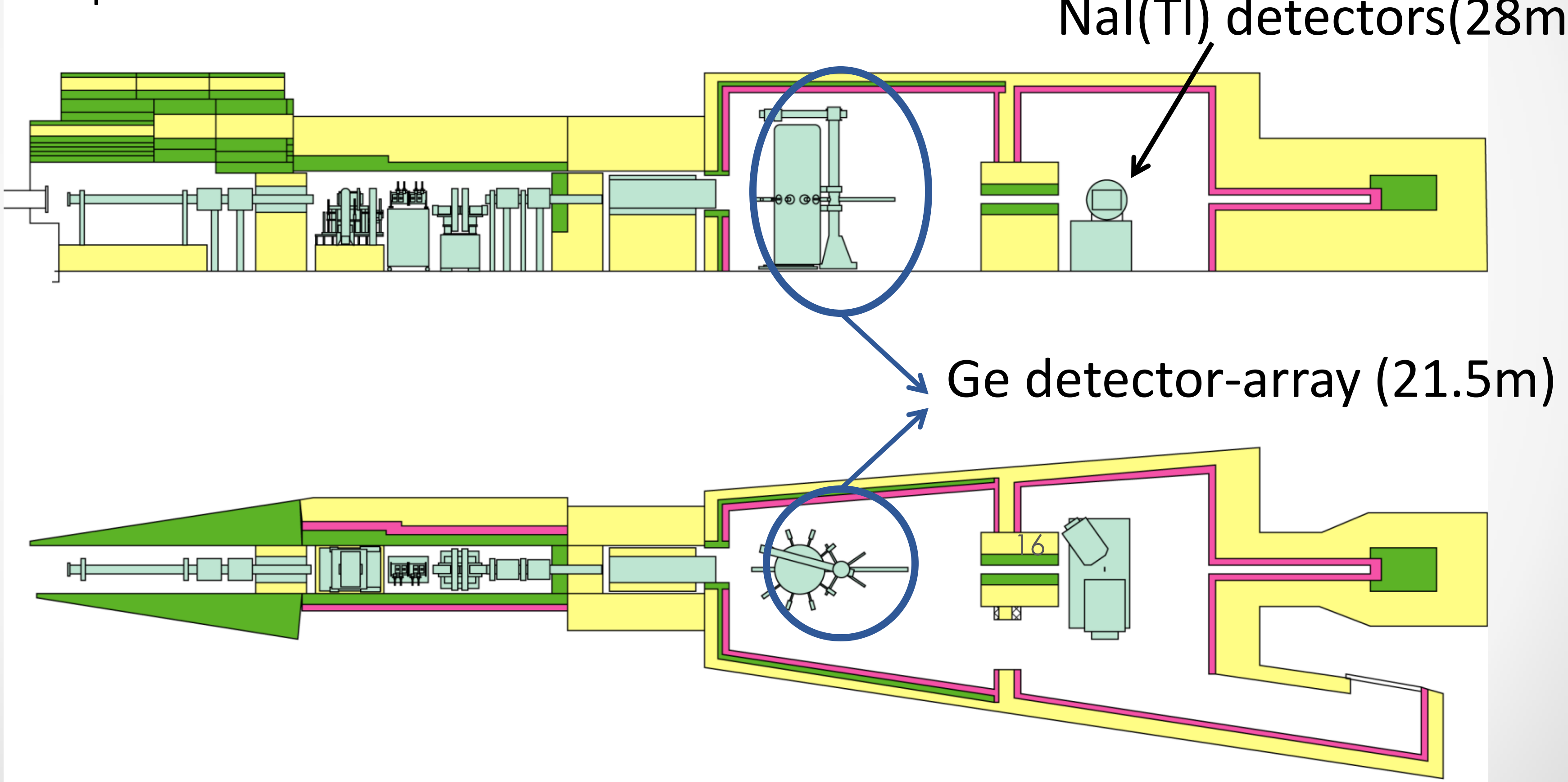
Our Facility, “ANNRI”
~ TOF facility ~

top view and side view of the ANNRI.



Our Facility, "ANNRI"
~ TOF facility ~

top view and side view of the ANNRI.



Our Facility, "ANNRI"
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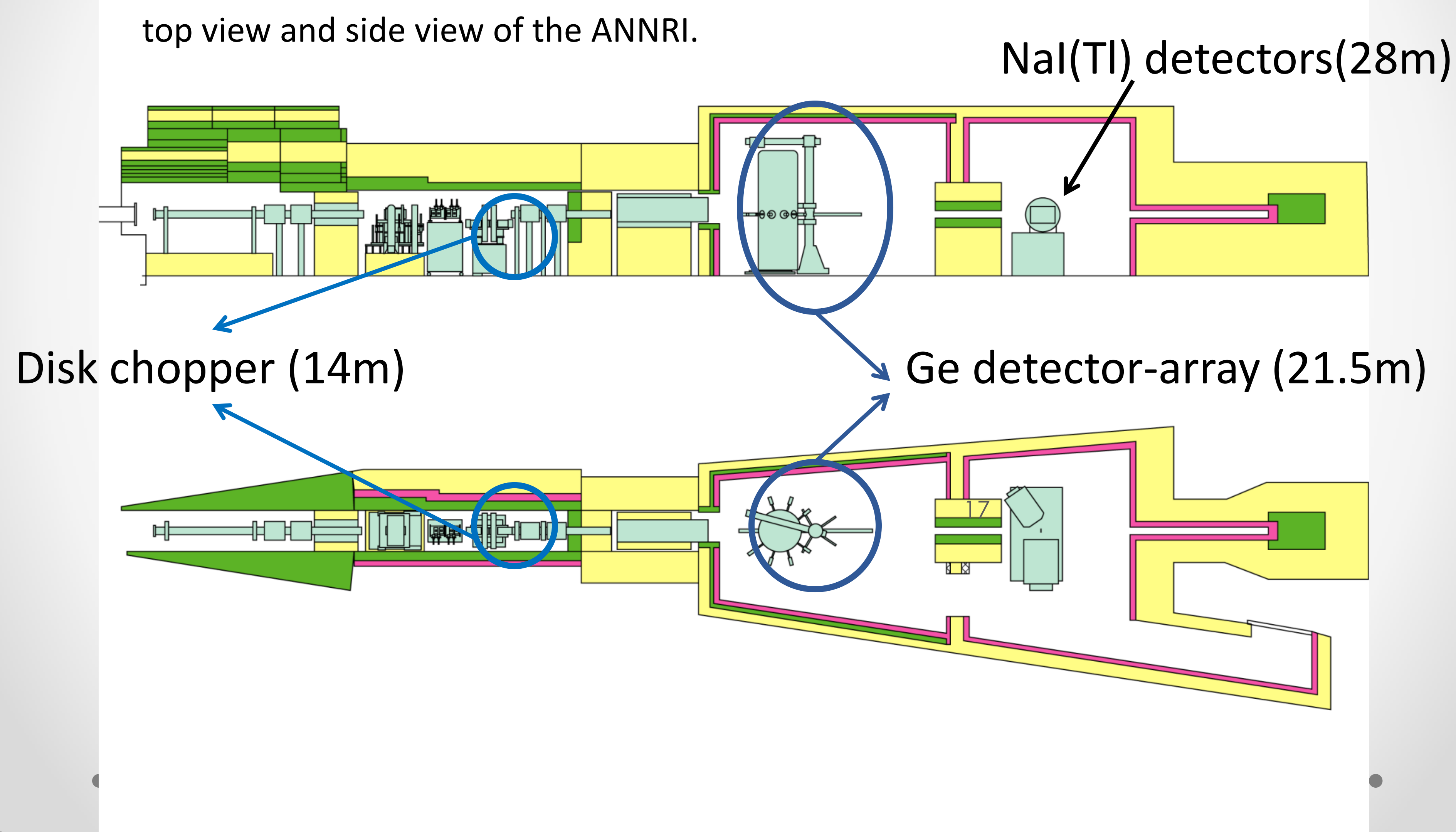

Our Facility, "ANNRI"
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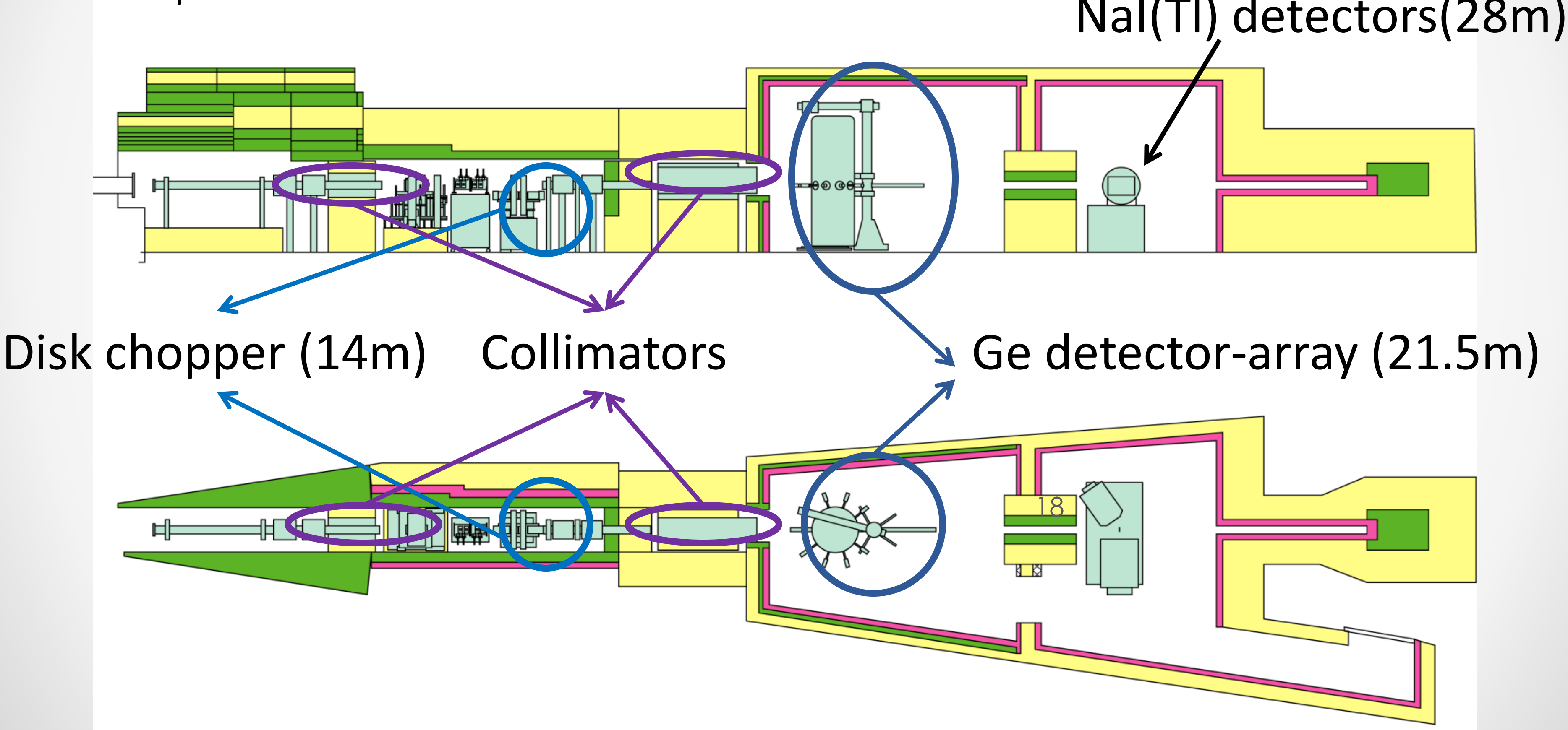
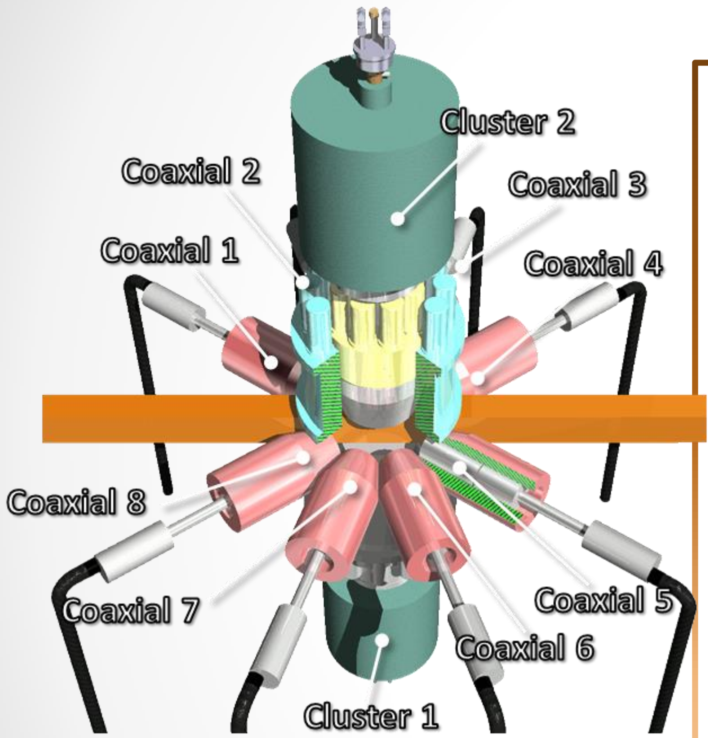


Our Facility, “ANNRI” ~Detector system~



Our spectrometer has

- 2 cluster-Ge detectors
  (7 Ge crystals are installed in the detector)
- 8 coaxial-Ge detectors
- Compton suppressing BGO detectors

⇒22 Ge Crystals.

Energy resolution for 1.33MeV γ-rays:

5.8keV (for 200kevents/s),

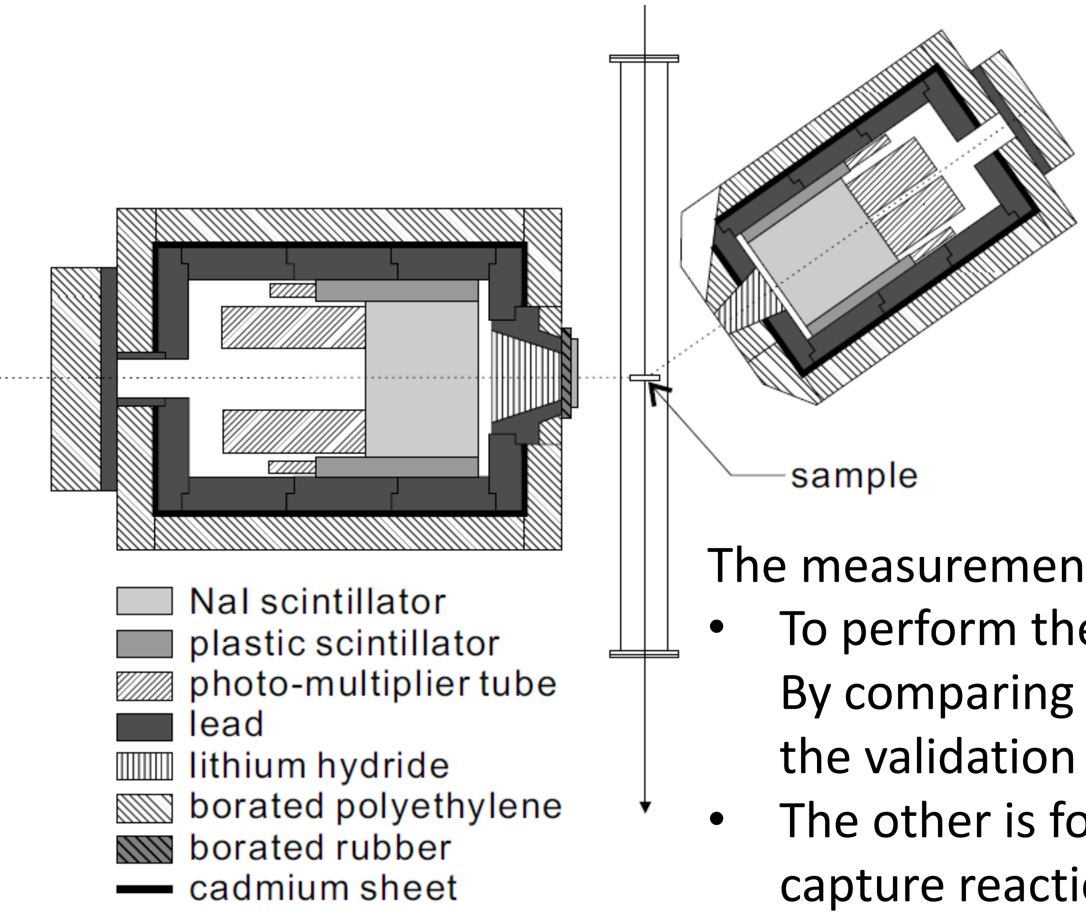
2.4keV (for 20kevents/s) [1]

Peak efficiency for 1.33MeV γ-rays:

3.64 ± 0.11 %

[1] T. Kin et. al., *the 2009 NSS-MIC Conf. Rec.* , N24-2, (2009).

~ NaI(Tl) detectors~



The NaI spectrometer consists of two NaI detectors.

Size: 330 mmϕ 203 mmL 90°

203 mmϕ 203 mmL 125°

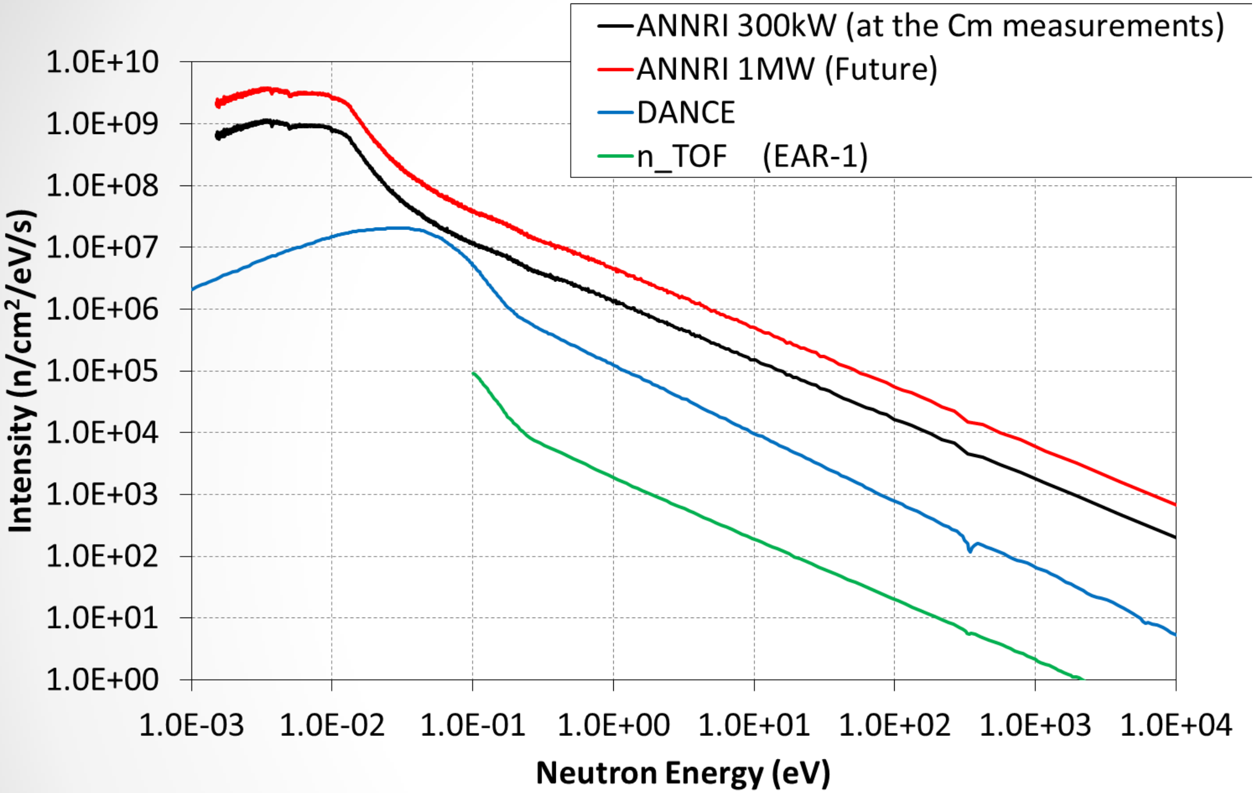
Surrounded by plastic scintillator for cosmic-ray and shields.

The measurement with the NaI(Tl) has two objectives.

- To perform the complementary measurement. By comparing the results for the same sample, the validation can be performed.
- The other is for measurements of the fast-neutron capture reaction. The upper limit was 100~keV

Our Facility, "ANNRI" ~ Beam Intensity ~

By Dr. Kino



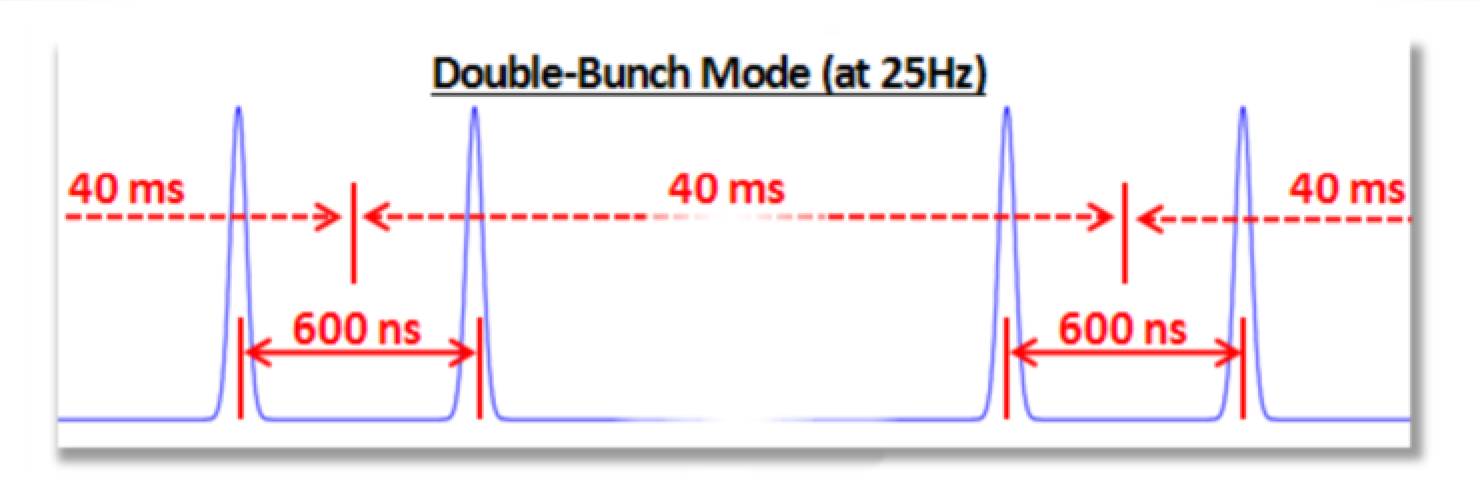
In the epithermal energy region, the neutron intensity is more than 10 times as high as the values of the other instruments.

Moreover, under the future 1-MW operation, these intensities are expected to increase.

K. Kino, *et. Al.*, NIM-A, **626**, 58 -66 (2011).

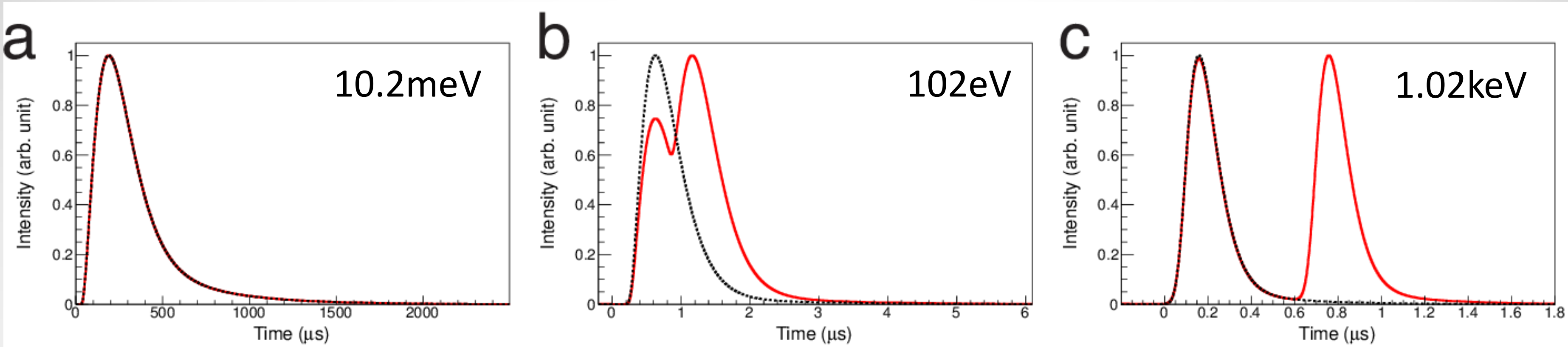
energy-integrated intensities	1MW (Future)	300kW
1.5-25 meV	3.7×10^7 n/s/cm^2	1.1×10^7 n/s/cm^2
0.9-1.1 keV	1.0×10^6 n/s/cm^2	2.9×10^5 n/s/cm^2

However, in J-PARC, pulsed protons usually consist of two bunches (called “double-bunch mode”), each with a width of 60 ns, at intervals of 600 ns

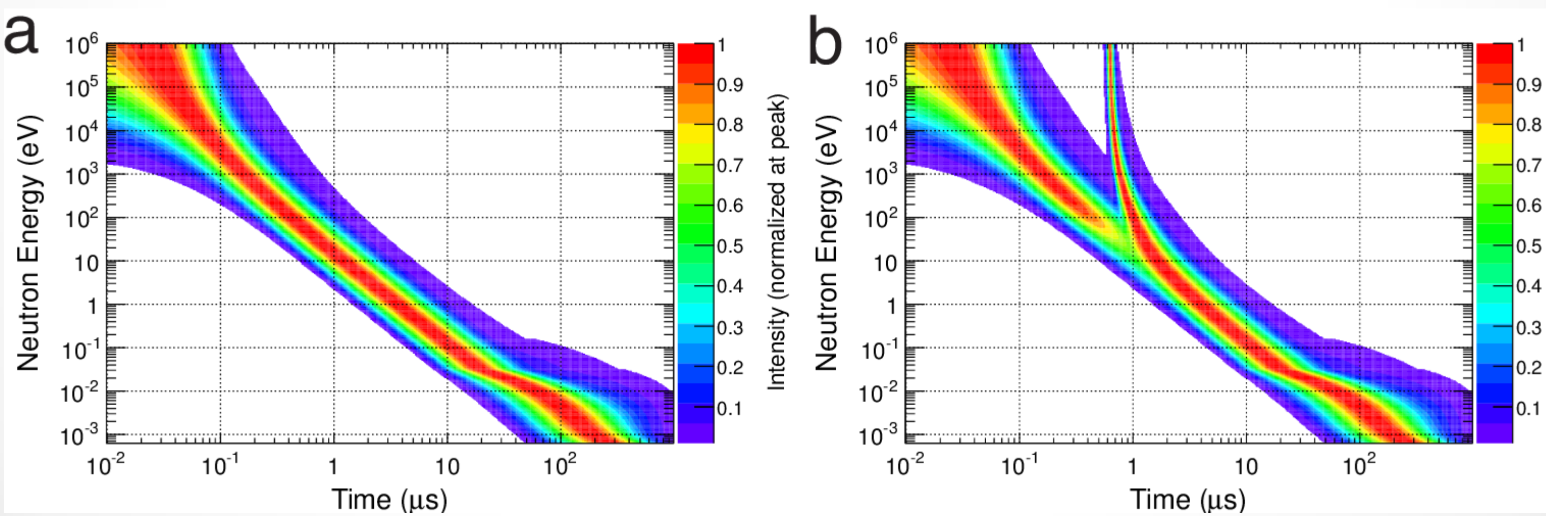


Most users in MLF are users of diffractometers, scattering spectrometers and reflectometers.
They require “neutron intensity”.

~ "Double Bunched" Resolution Function~



At the low neutron energy (a), the time structures of the single and double bunches are almost same. However, the time structure is affected by the double bunch as the neutron energy increases (b, c).



Two-dimensional plots as a function of the time and energy of neutrons at the moderator surface for single bunch (a) and double bunch (b) modes.

Regulations

Furthermore, because of the regulations of MLF,

- Un-sealed samples
  (Fission chambers are recognized as unsealed
  by radiation safety team.)
- Pu and U

can not be handled.

-> To do fission experiments are very difficult!!

(Is there a merit to do fission experiments in ANNRI?)

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Experiments of $^{244,246}Cm$

~Samples and Measurement conditions~

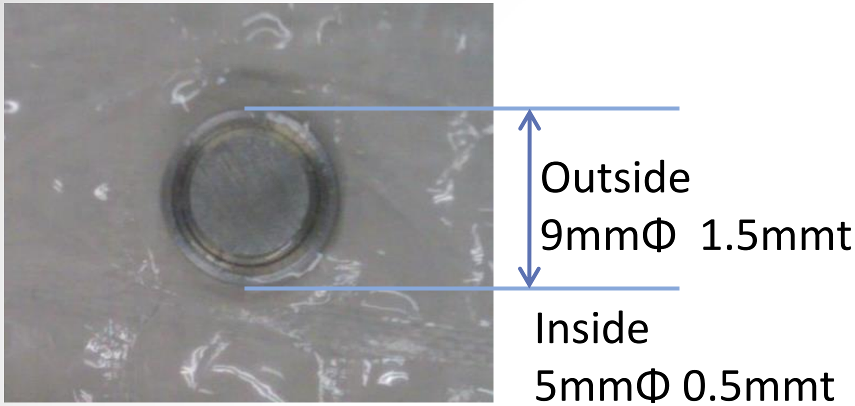


Table 1 The isotopic composition of the ^{244}Cm sample or the ^{246}Cm sample.[1]

	^{244}Cm sample	^{246}Cm sample
	TIMS (mole%)	TIMS (mole%)
^{244}Cm	90.1±1.7	27.5±0.5
^{245}Cm	2.71±0.34	1.06±0.28
^{246}Cm	7.22±0.34	59.4±1.3
^{247}Cm	N.D.	2.9±0.4
^{248}Cm	N.D.	9.10±0.24

Samples:

Cm-244 ($T_{1/2}$=18.1y: MA)

Net weight = 0.6 mg

Activity = 1.8 GBq

Measurement Periods: 64 hours

Cm-246 ($T_{1/2}$=4753y: MA)

Net weight = 2.1 mg

Activity = 12.1 MBq (^{244}Cm: 1.7GBq)

Measurement Periods: 94 hours

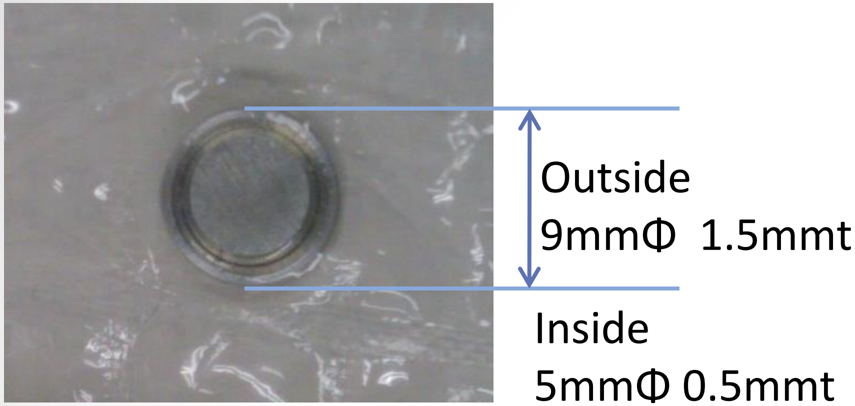
Both of the samples

Chemical form = CmO_2

Container = Al capsule

For the background estimation, a dummy case (Al 278mg) and a blank sample was measured for done for 48 and 44 hours.

Experiments of $^{244,246}Cm$
~Samples and Measurement conditions~



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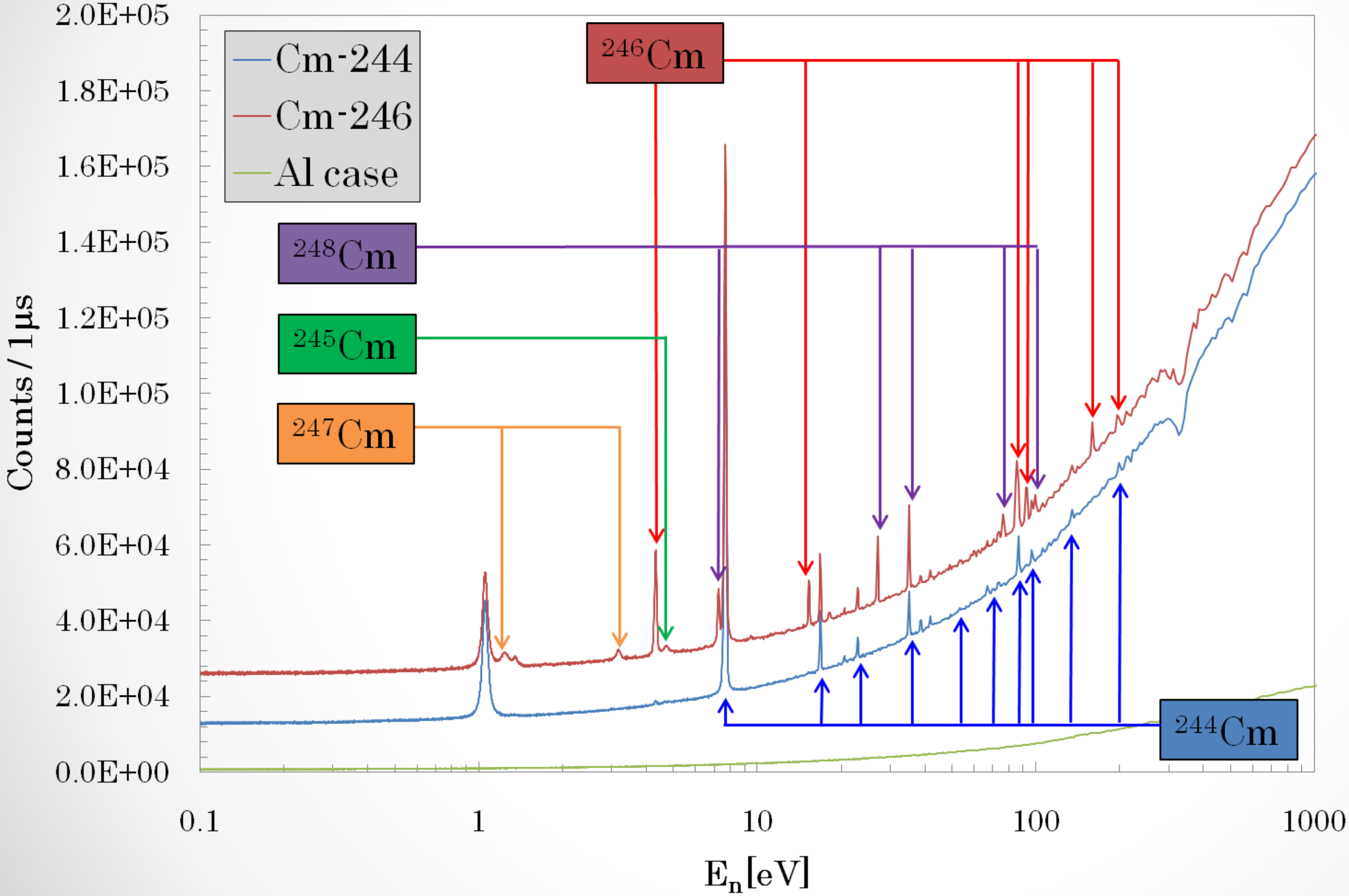
- To reduce air scattering, the air in the beam duct was replaced with helium.

Experiments of $^{244,246}Cm$

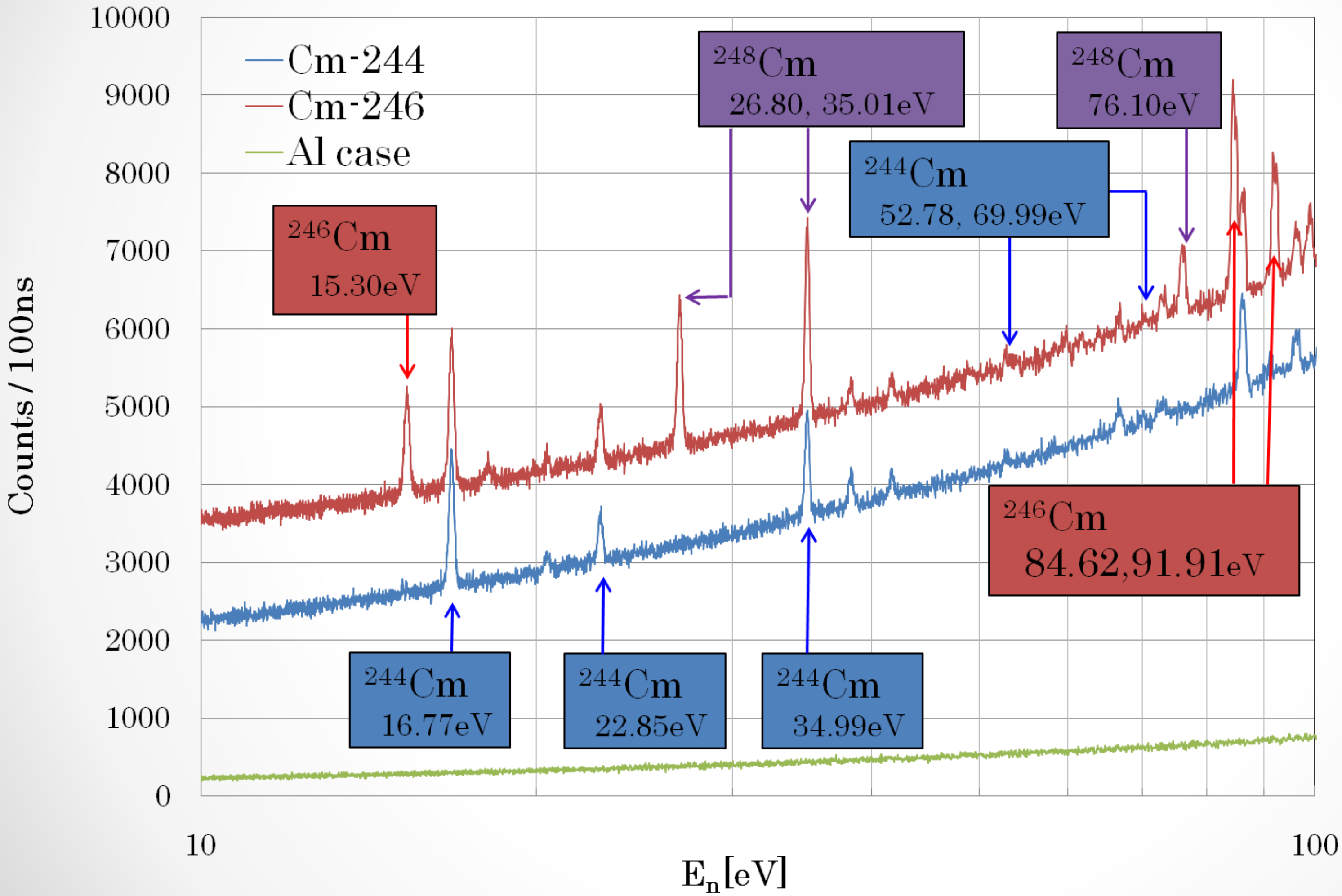
~TOF Spectrum ~



This graph shows TOF spectra of the ^{244}Cm, the ^{246}Cm sample, and the dummy case

Experiments of $^{244,246}Cm$

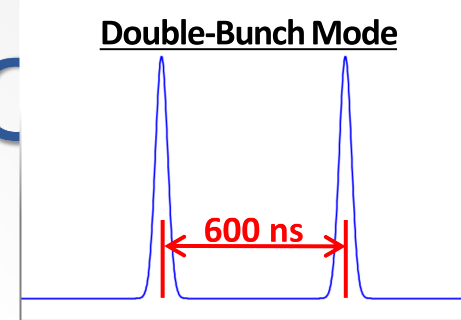
~TOF Spectrum ~

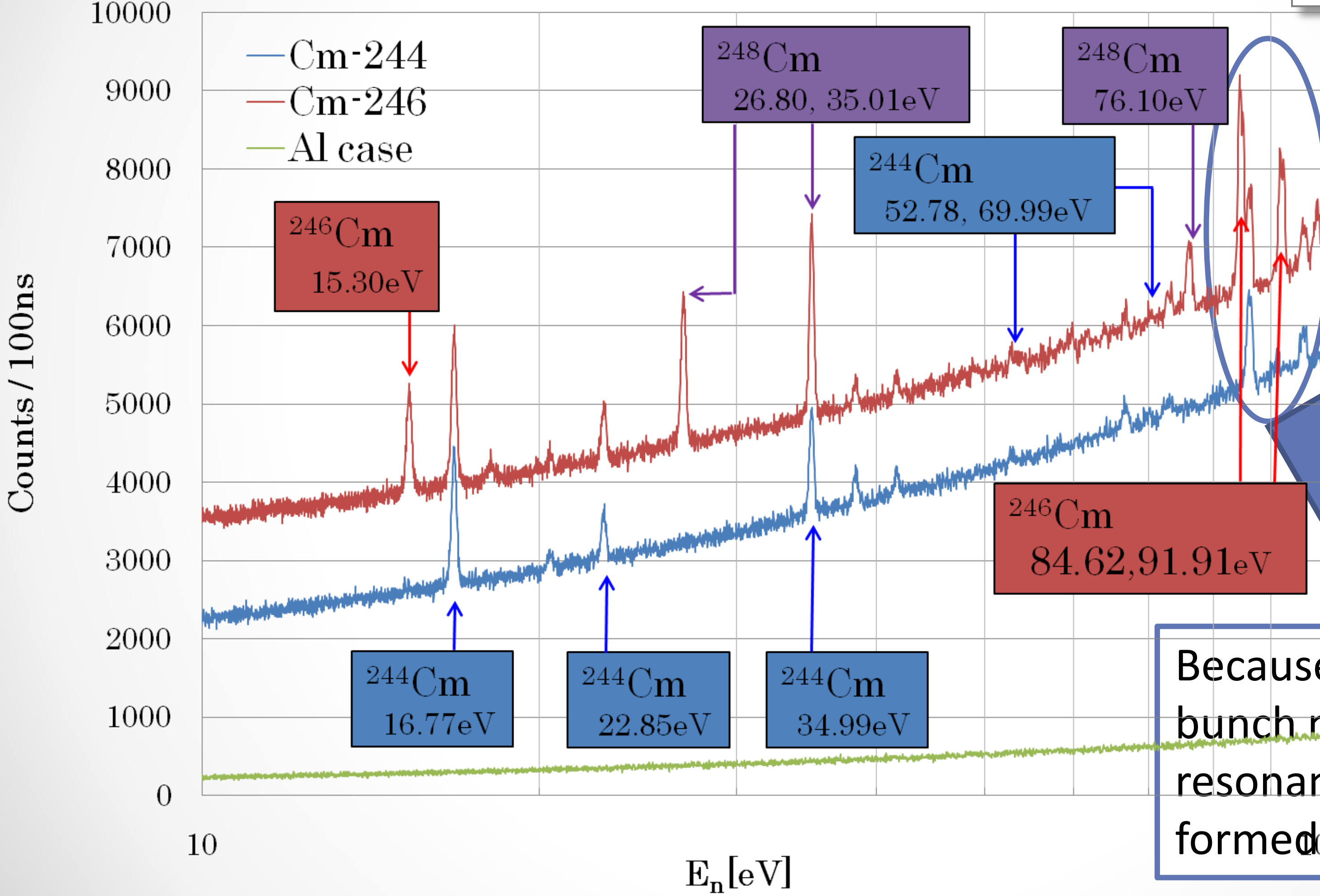


This graph shows TOF spectra of the ^{244}Cm, the ^{246}Cm sample, and the dummy case

Experiments of ^{244,246}C

~TOF Spectrum ~



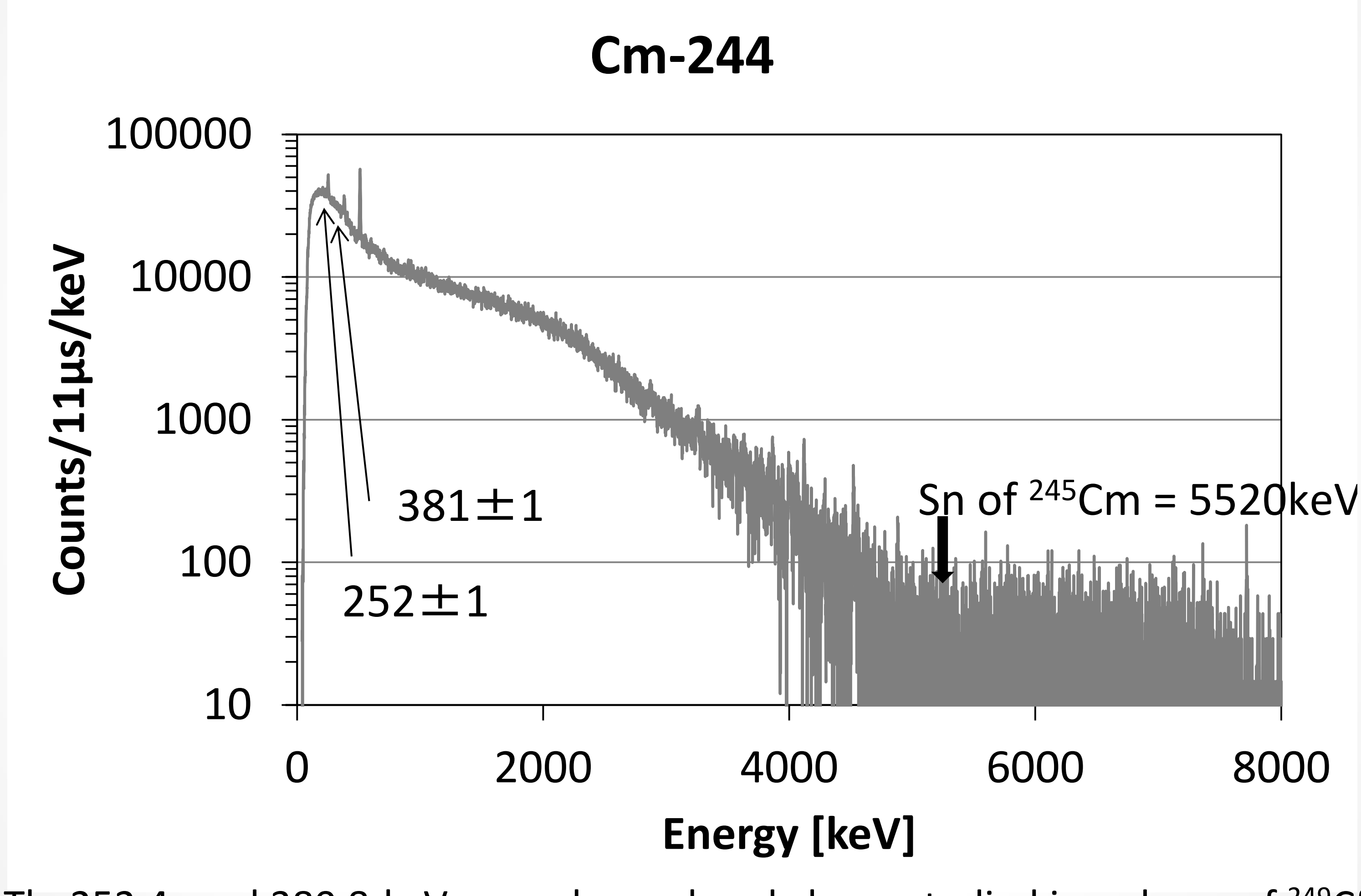


Because of the double-bunch mode, resonance peaks were formed double peaks.

This graph shows TOF spectra of the ^{244}Cm, the ^{246}Cm sample, and the dummy case

Experiments of $^{244,246}Cm$

~ γ-ray Spectrum at the 1st resonance of ^{244}Cm ~



The252.4- and 380.8-keV γ-rays have already been studied in α decay of ^{249}Cf, electron capture decay of ^{245}Bk, and β-decay of ^{245}Am.
The other γ-rays were previously unknown γ rays.

Experiments of $^{244,246}Cm$

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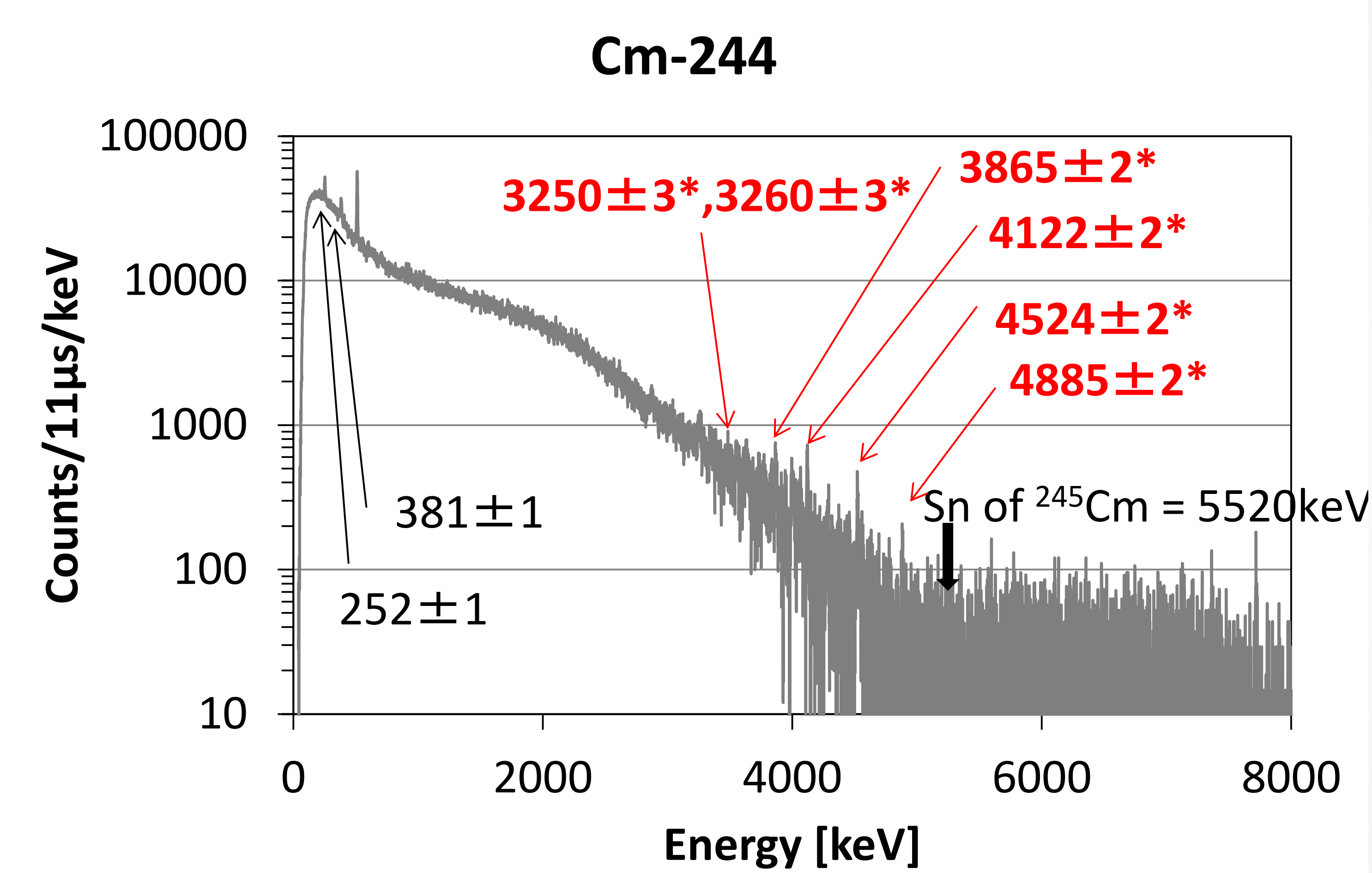

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Experiments of $^{244,246}Cm$ ~Analysis~

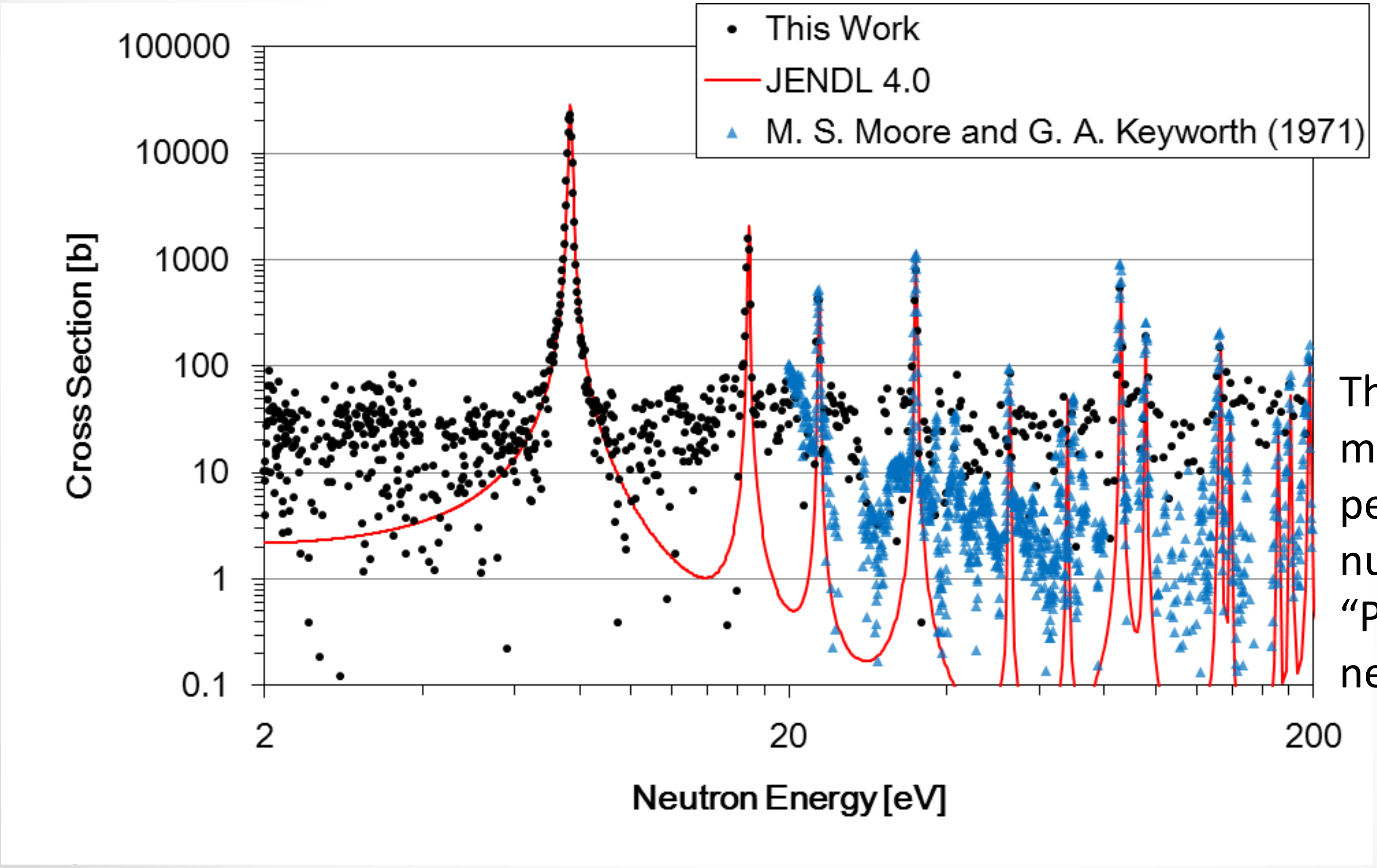
The data were analyzed with the procedure.

- **Dead-Time Correction**
- **Background Estimation and Subtraction**
- **Self-Shielding and Multiple-Scattering Correction**
- **Normalization (Using the 1st resonance of ^{240}Pu)**
- **Evaluation and Subtraction of Influence of Fission Events**
- **Evaluation and Subtraction of Influence of Impurities**

The obtained neutron-capture cross sections are….

Experiments of $^{244,246}Cm$
~Cross Section of ^{244}Cm~

Only one neutron-capture cross-section data of ^{244}Cm (n,g) was reported in 1969[1].



The previous measurement was performed using the nuclear explosion “Physics 8” as a pulsed neutron source in 1969.

[1]M. S. Moore et.al. , Physical Review C, 3, 1656 (1971).

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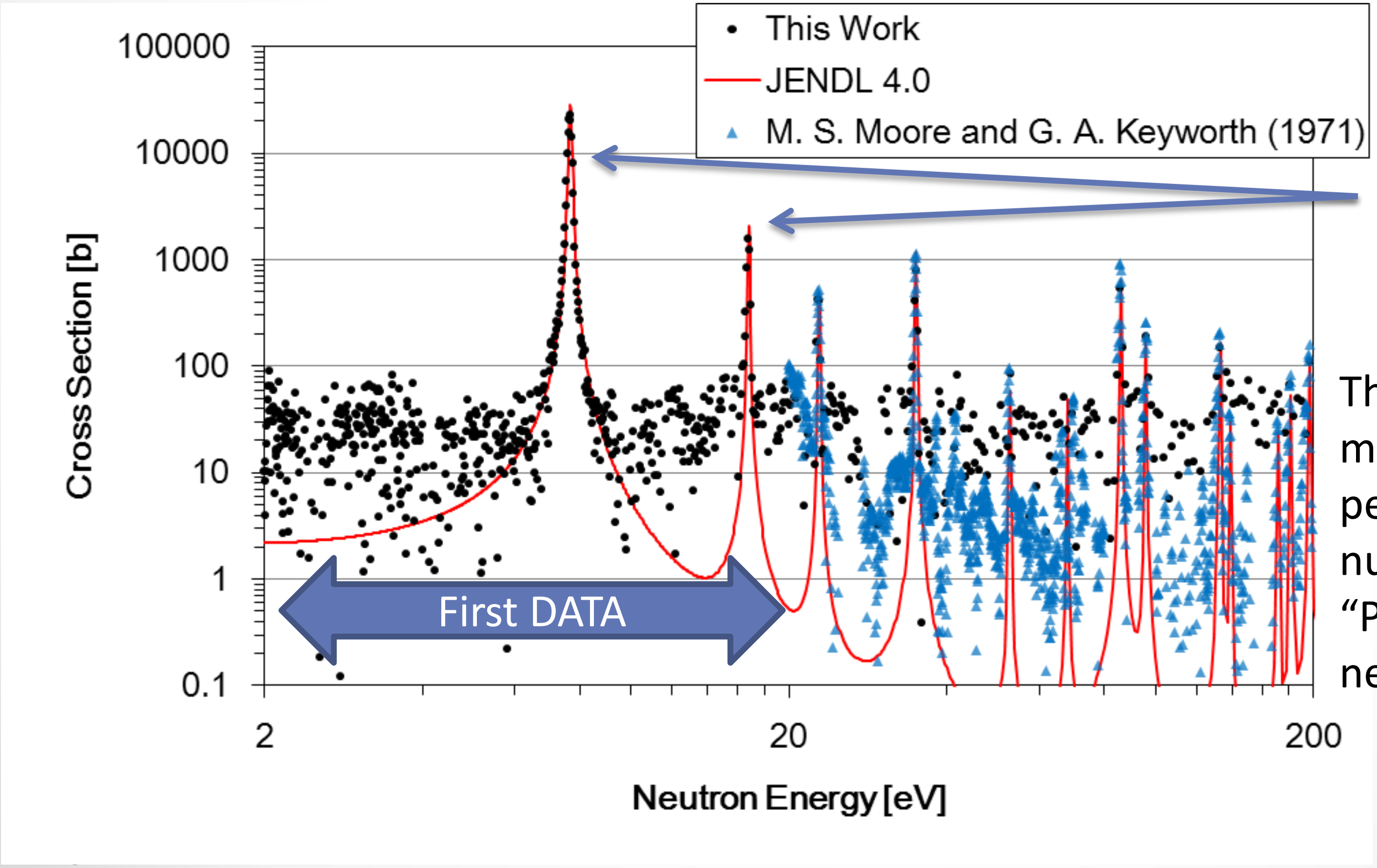

The results of the resonance peaks under 20-eV are also the first experimental results in the world.

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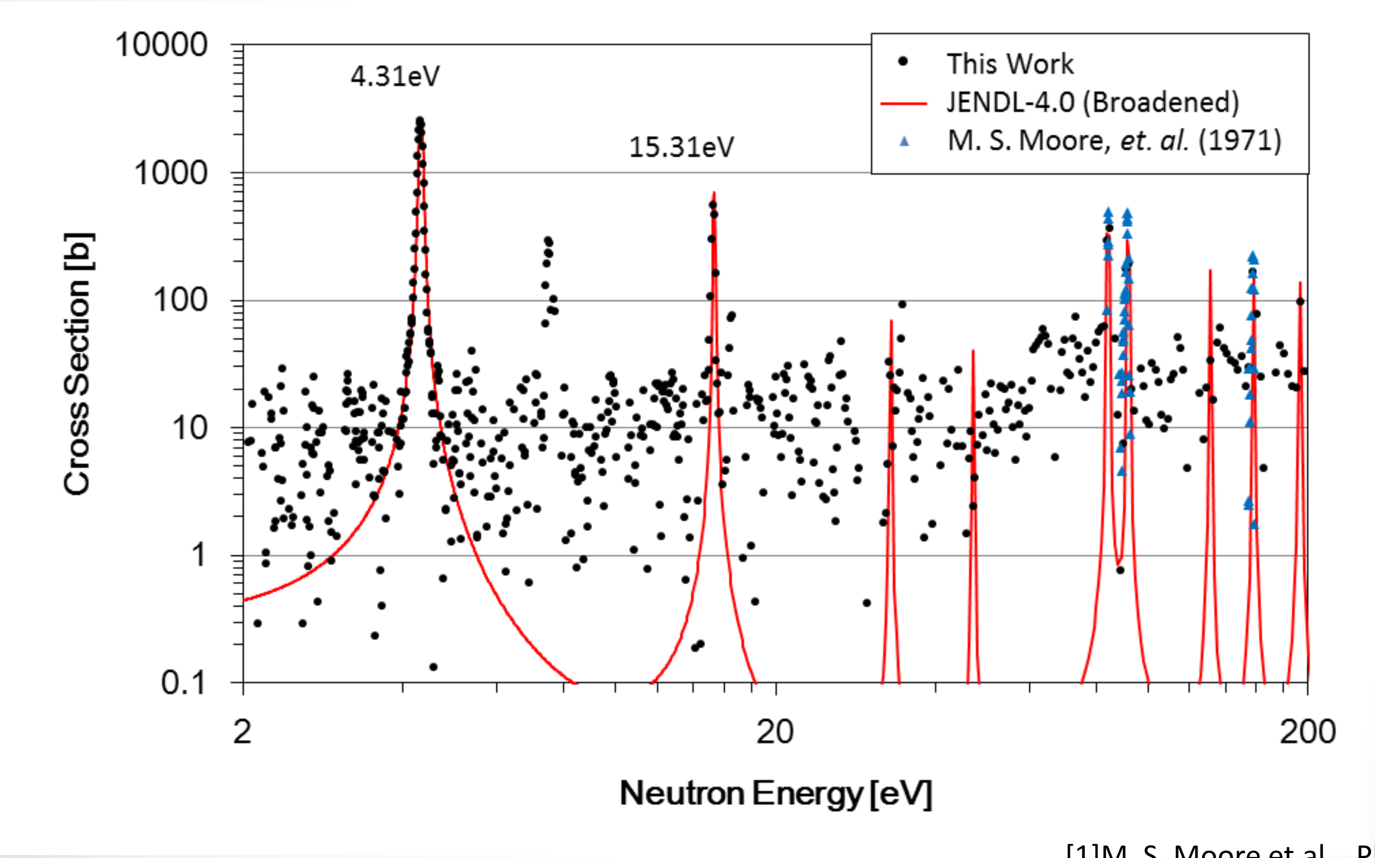
Experiments of $^{244,246}Cm$

~Cross Section of ^{246}Cm~

Only one neutron-capture cross-section data of ^{246}Cm (n,γ) was made in 1971[1].



[1]M. S. Moore et.al. , Physical Review C, 3, 1656 (1971).

Experiments of $^{244,246}Cm$

~Cross Section of ^{246}Cm~

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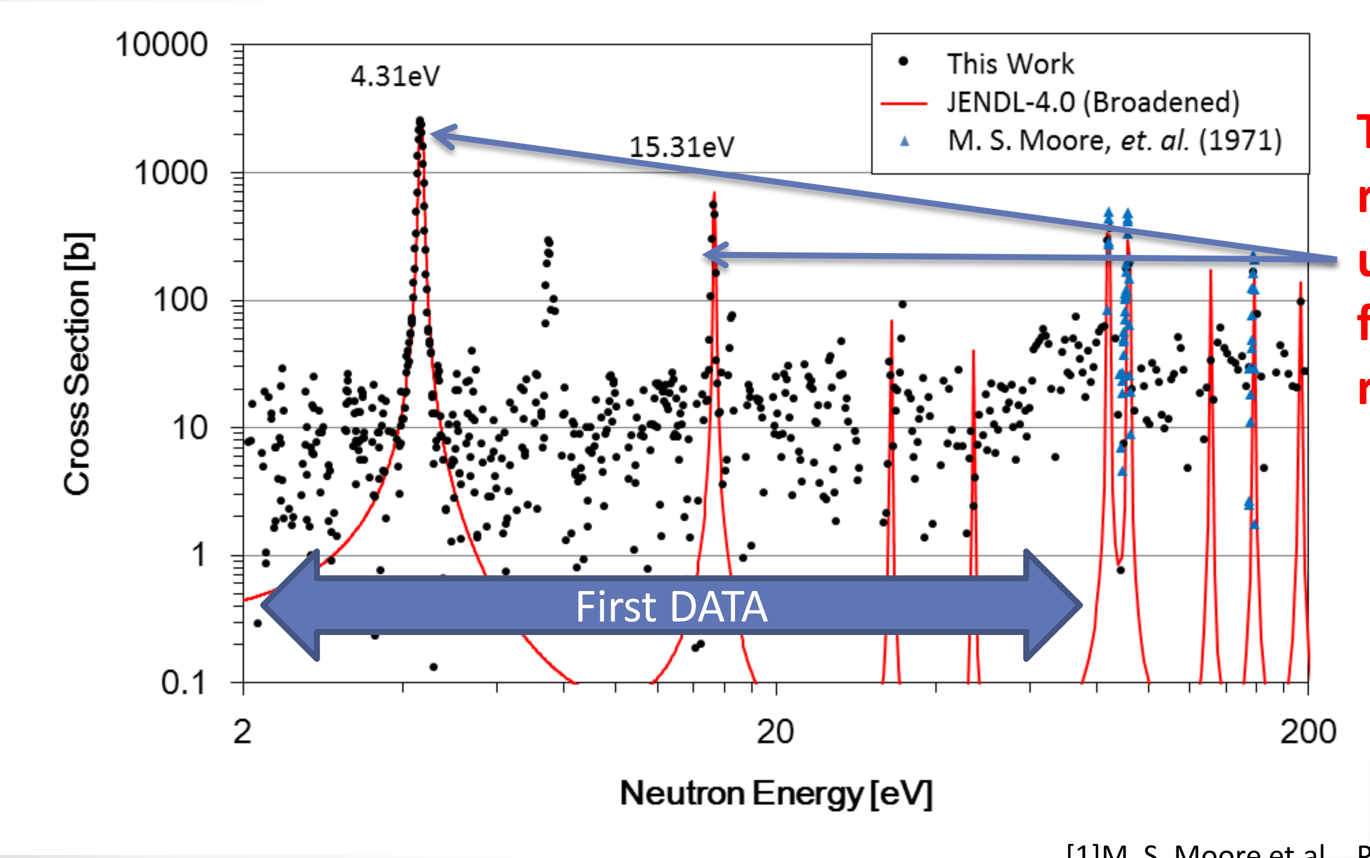

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~Experimental Set up For Sn-112~

	Chemical form	Diameter [mm]	Weight [mg]	Time [hr]
^{112}Sn	**Metal**	**5**	**97.3**	**37.6**
^{208}Pb	Metal	5	159.7	16.3
Blank	Holder and films only			22.7

Beam Condition

Beam Power = 210kW

Beam Size = 7mmϕ

Beam Duct = vacuum

Each sample was put in a FEP bag and attached to a PET sample holder.

- For the background estimation, a ^{208}Pb sample (99.57%) and a blank sample were measured.
- For dead-time correction, pulses from a random-pulse generator were input to the pre-amplifier of every Ge crystal.

	Isotopic Distribution[%]									
	^{112}Sn	^{114}Sn	^{115}Sn	^{116}Sn	^{117}Sn	^{118}Sn	^{119}Sn	^{120}Sn	^{122}Sn	^{124}Sn
^{112}Sn	99.6	0.3	0.05	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01

Data processing and analysis

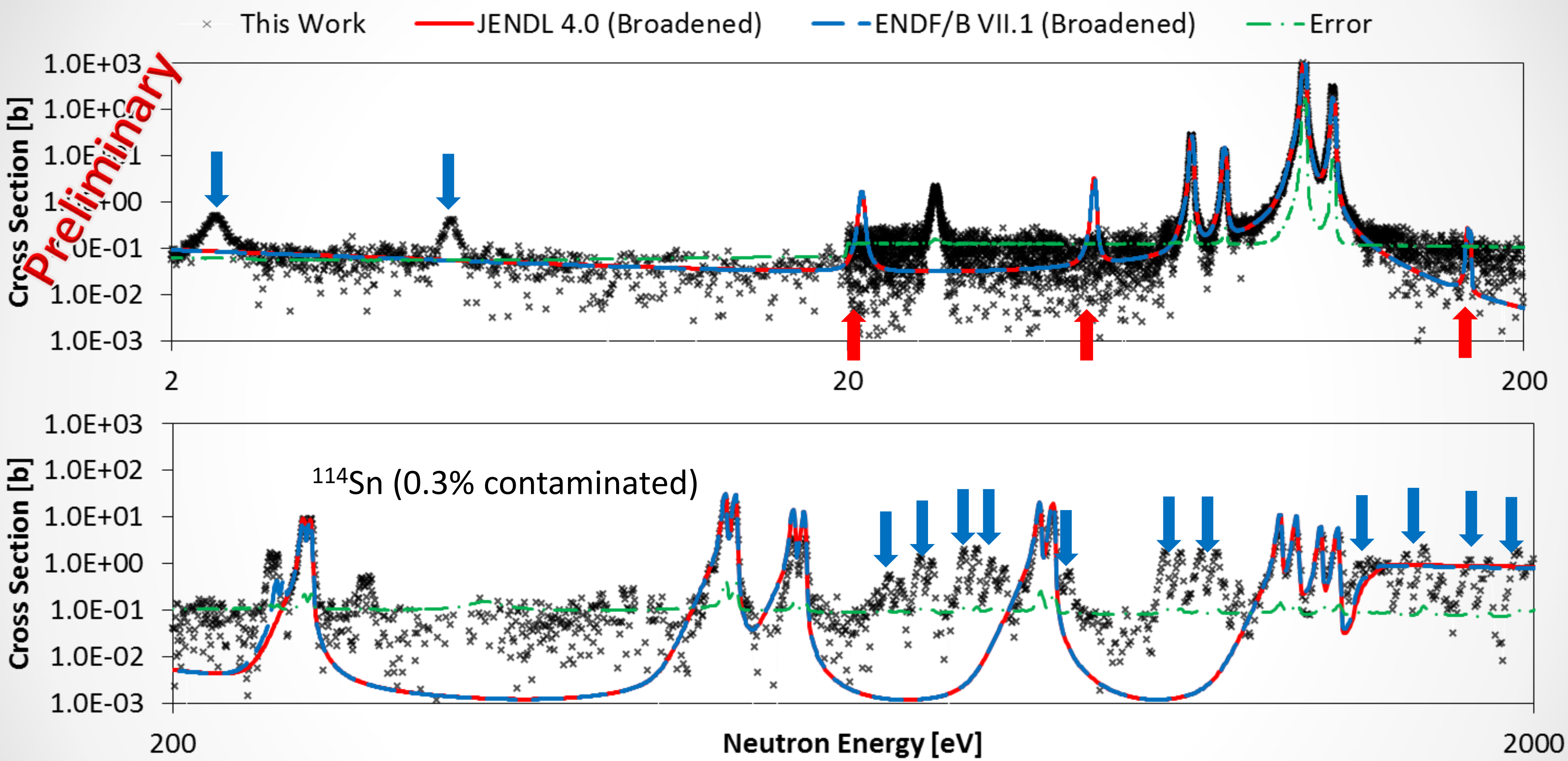
The data were analyzed with this procedure.

- **Dead-Time Correction**
- **Background Estimation and Subtraction**
- **Self-Shielding and Multiple-Scattering Correction**
- **Normalization**
  For ^{112}Sn : to thermal cross sections by Krane[1]

The obtained results of neutron-capture cross sections are….

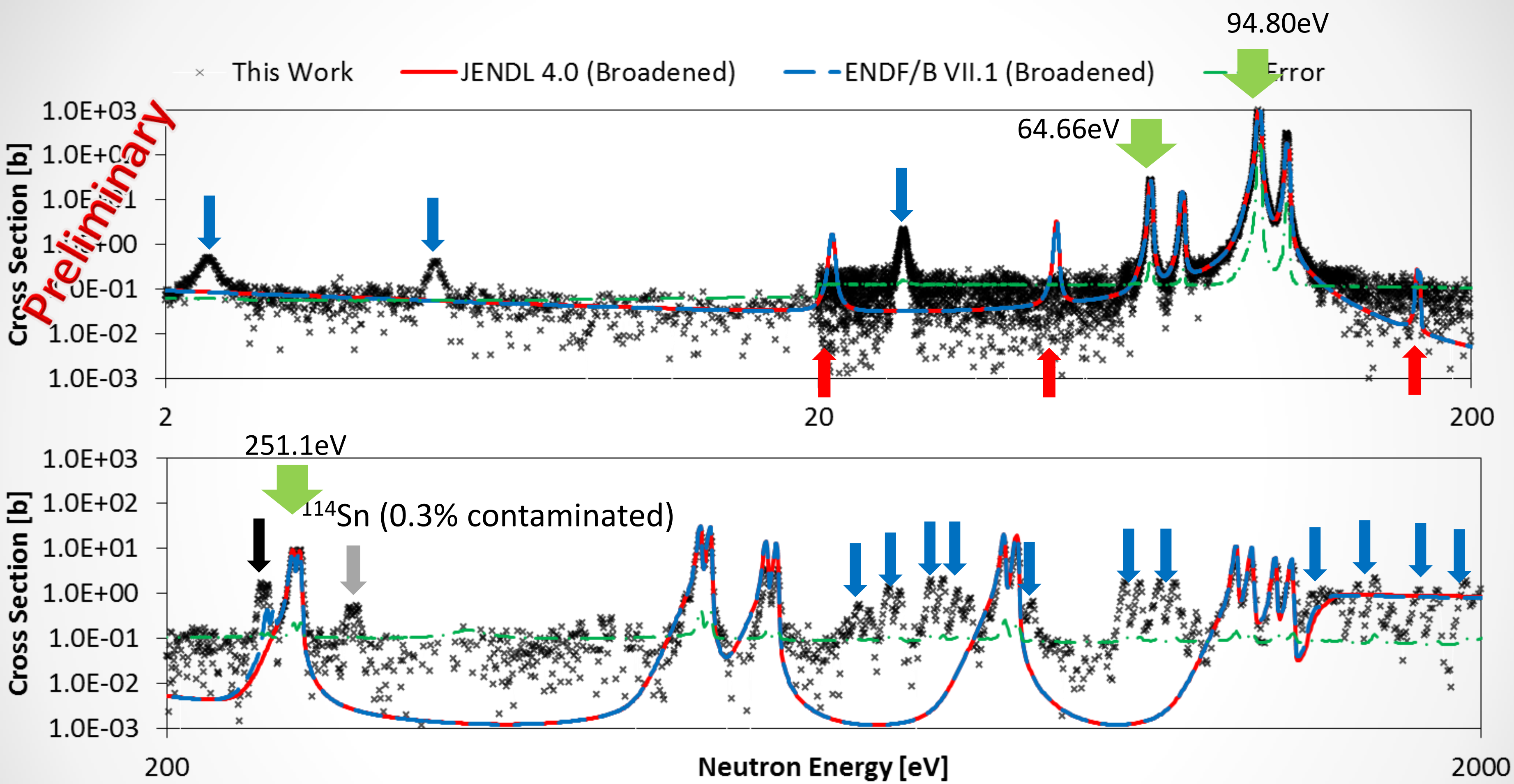
[1] K. S. Krane and J. Sylvester, PHYSICAL REVIEW C **73**, 054312 (2006)

~Cross Section of ^{112}Sn~



- 14 unknown resonances were observed
- Resonances at 21, 46, and 251 eV were not observed,
- The resonance at 240 eV is listed in ENDF B-VII but not listed in JEBDL 4.0.

~Cross Section of ^{112}Sn~



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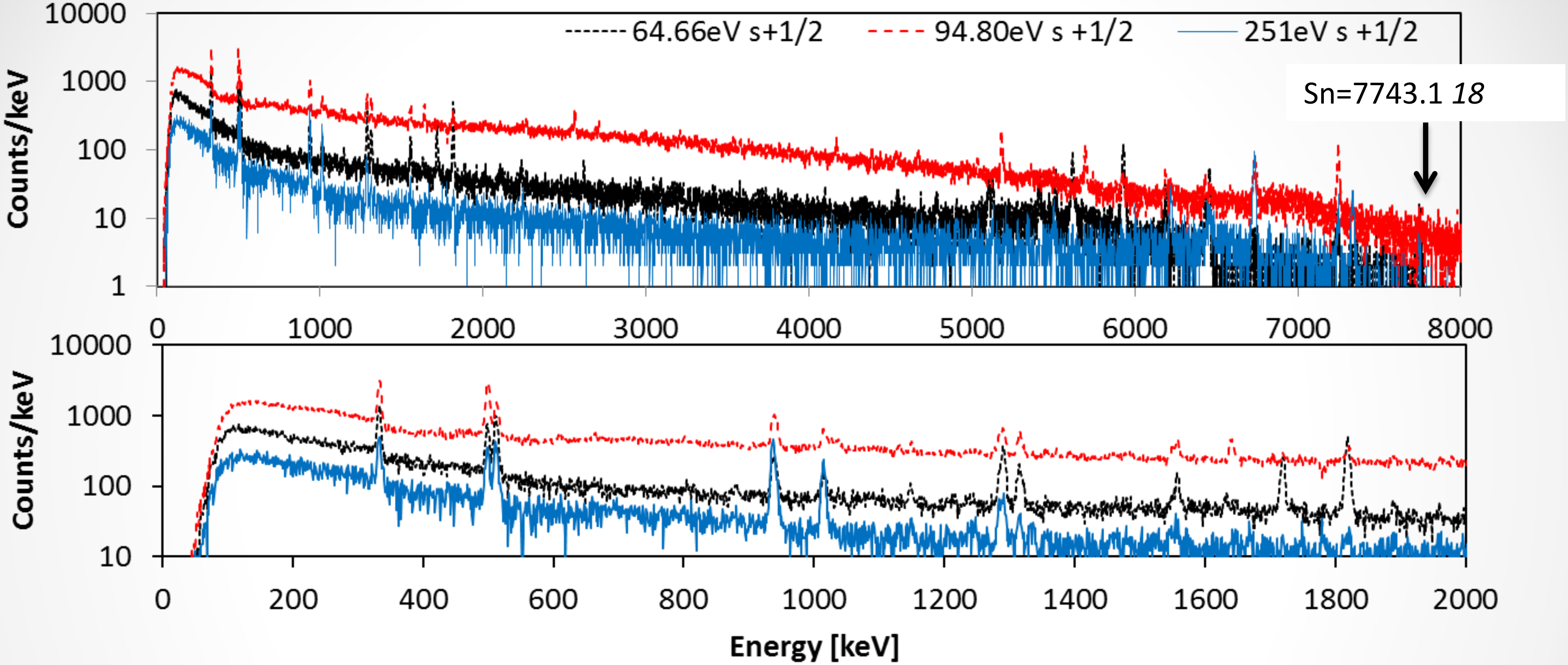
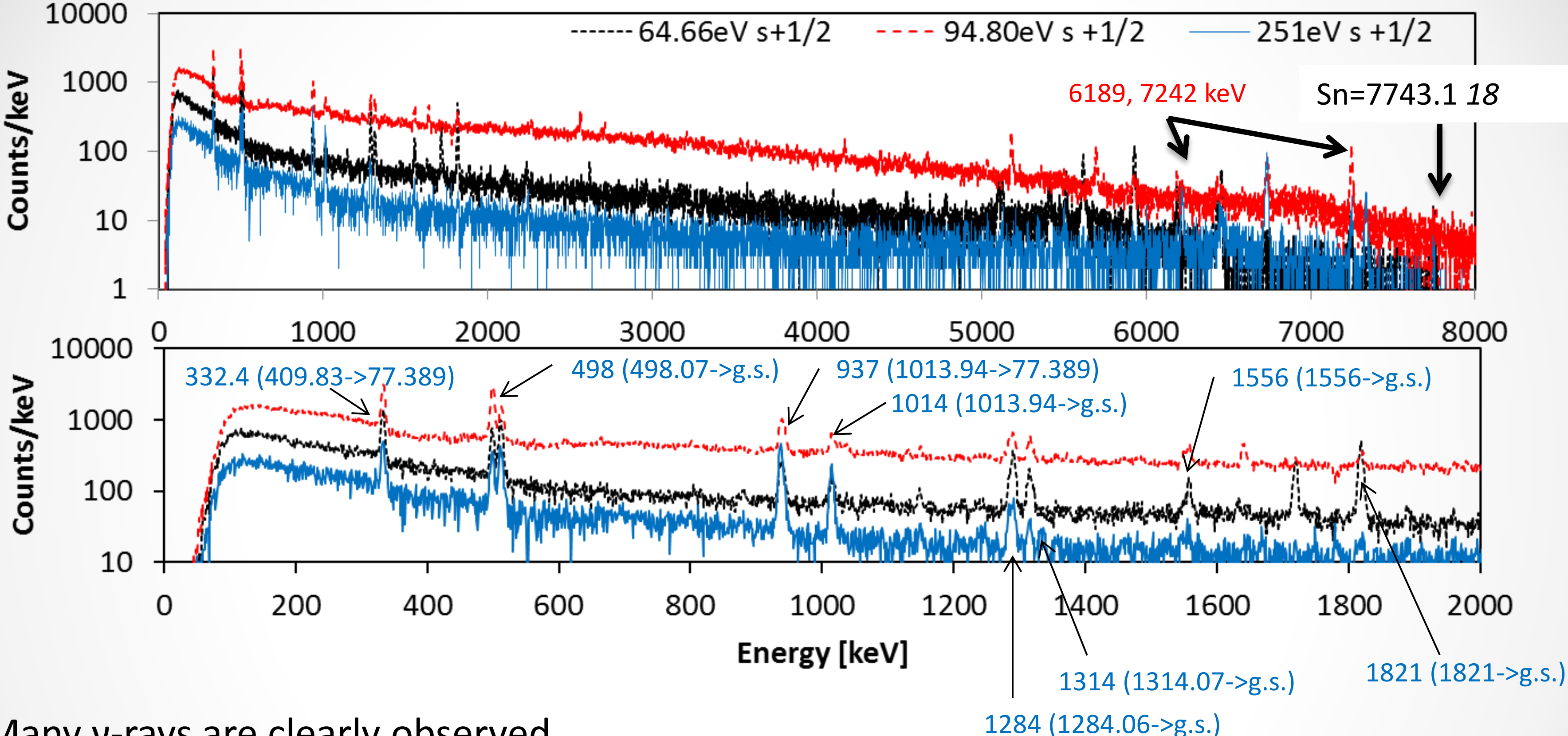
~γ-ray Distributions of each resonance~



Many γ-rays are clearly observed.

Are these spectra important??

~γ-ray Distributions of each resonance~

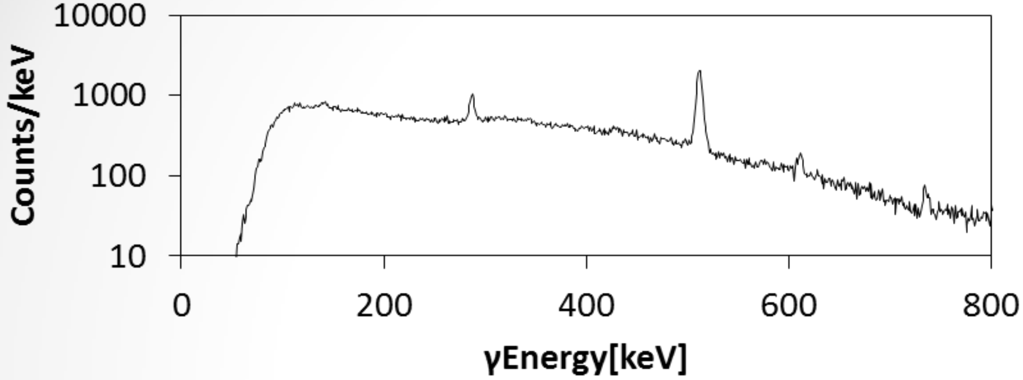


Many γ-rays are clearly observed.

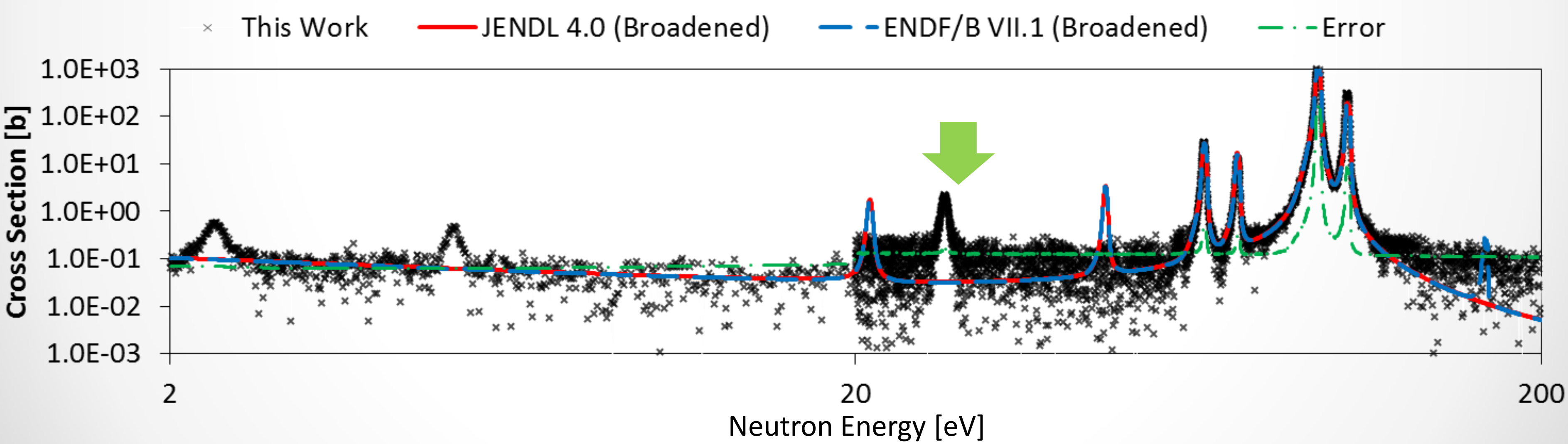
The 6189- and 7242-keV γ-rays have been reported by C. Samour.
8 γ-rays have already been studied in other reactions. ((p,nγ), (p,3nγ), (α,2n))
16 or more previously unknown γ-rays are observed.

Are these spectra important??

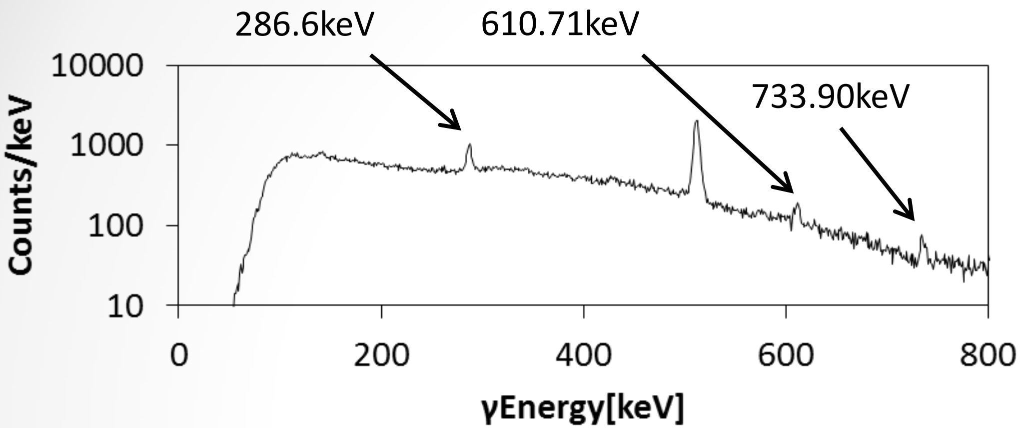
~Origin of the 27.1-eV resonance~



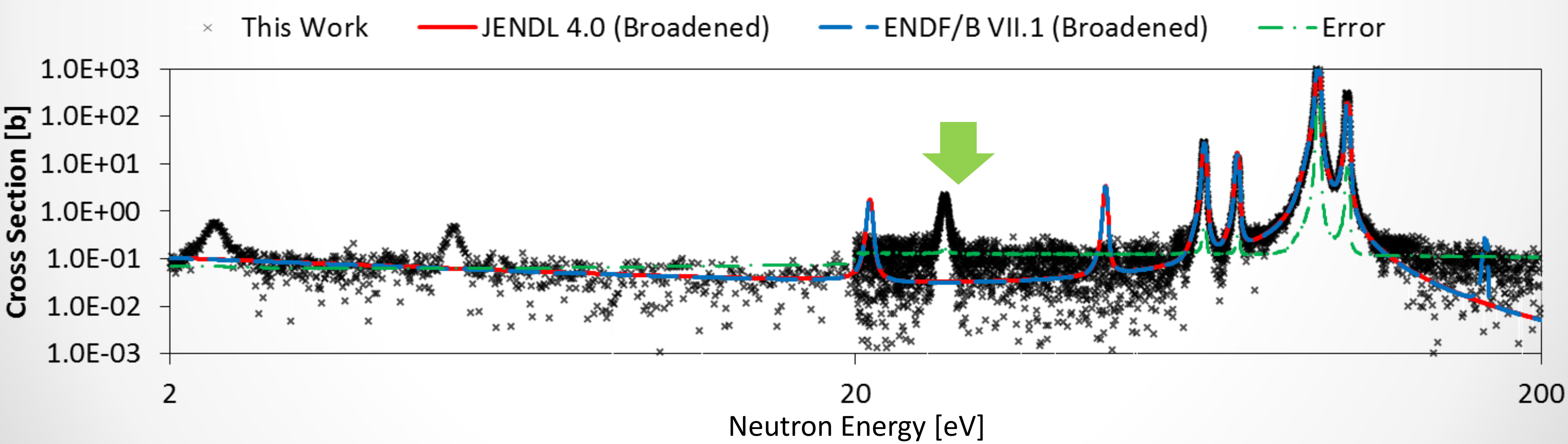
- Prompt γ-rays of ^{112}Sn are not observed. (332, 498 keV)



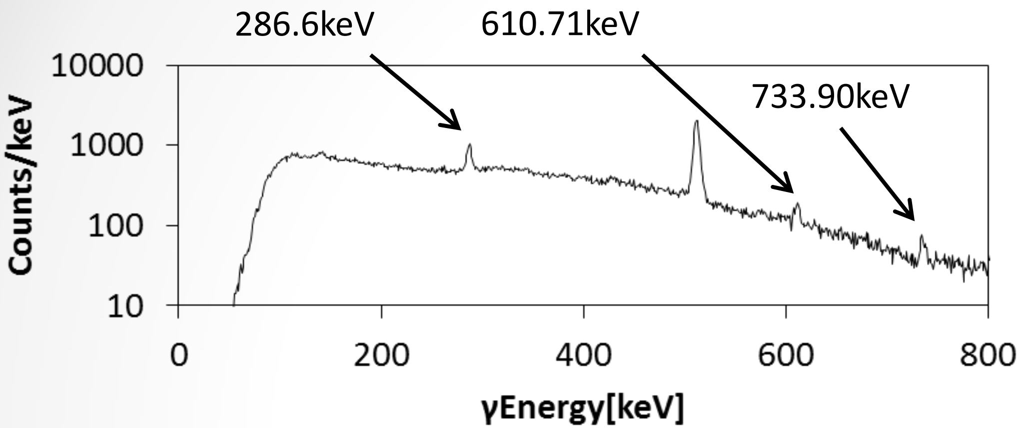
~Origin of the 27.1-eV resonance~



- Prompt γ-rays of ^{112}Sn are not observed. (332, 498 keV)
- Prompt g-rays of ^{74}Se (286.57, 610.71 and 733.90 keV) are clearly observed.

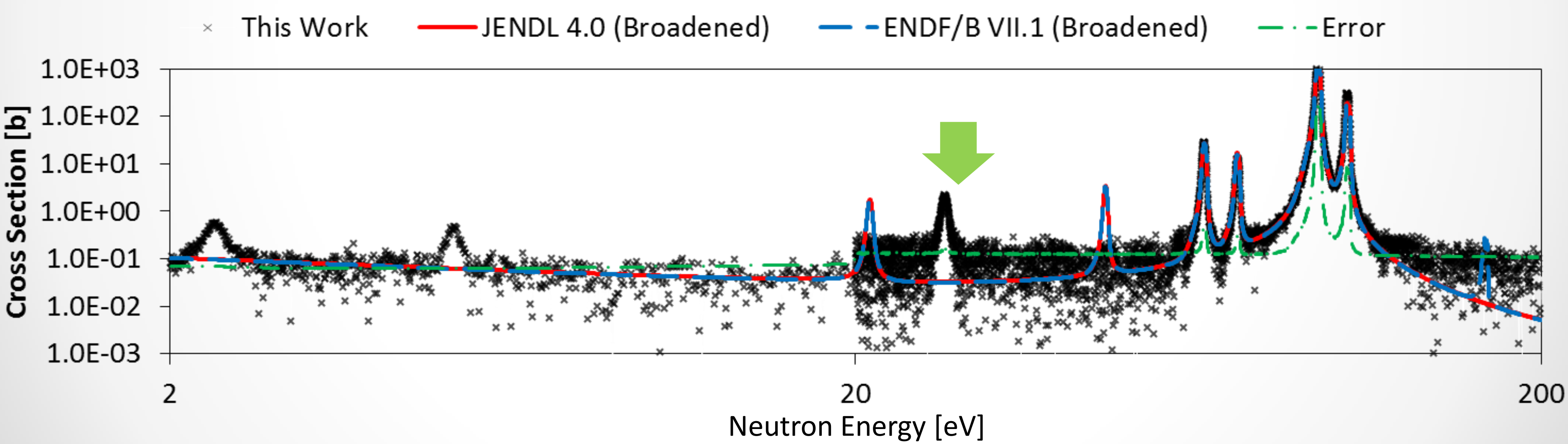


~Origin of the 27.1-eV resonance~

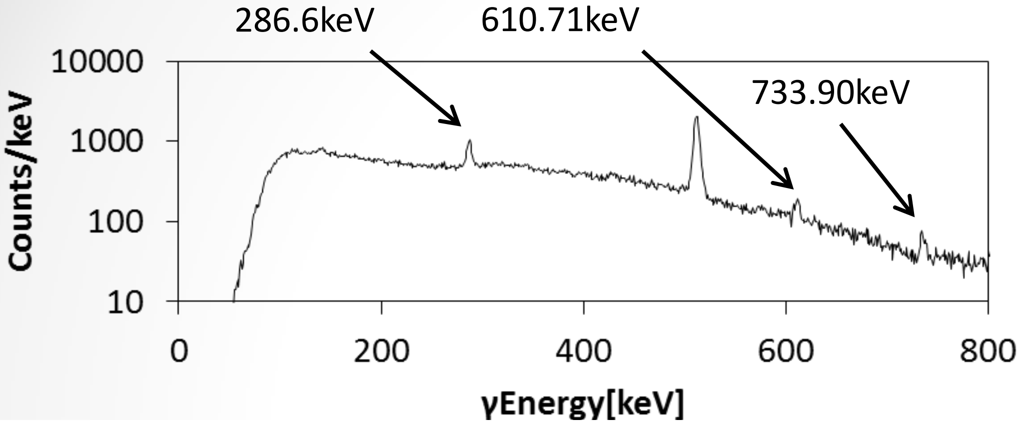


- Prompt γ-rays of ^{112}Sn are not observed. (332, 498 keV)
- Prompt g-rays of ^{74}Se (286.57, 610.71 and 733.90 keV) are clearly observed.

1st resonance of ^{74}Se (27.1eV)

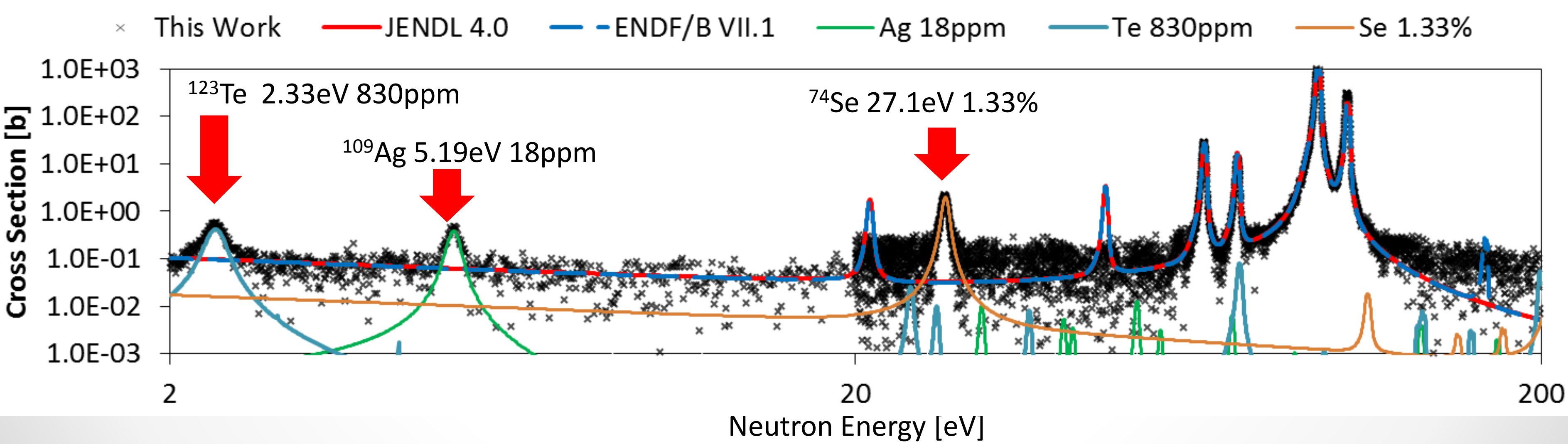


~Origin of the 27.1-eV resonance~



- Prompt γ-rays of ^{112}Sn are not observed. (332, 498 keV)
- Prompt g-rays of ^{74}Se (286.57, 610.71 and 733.90 keV) are clearly observed.

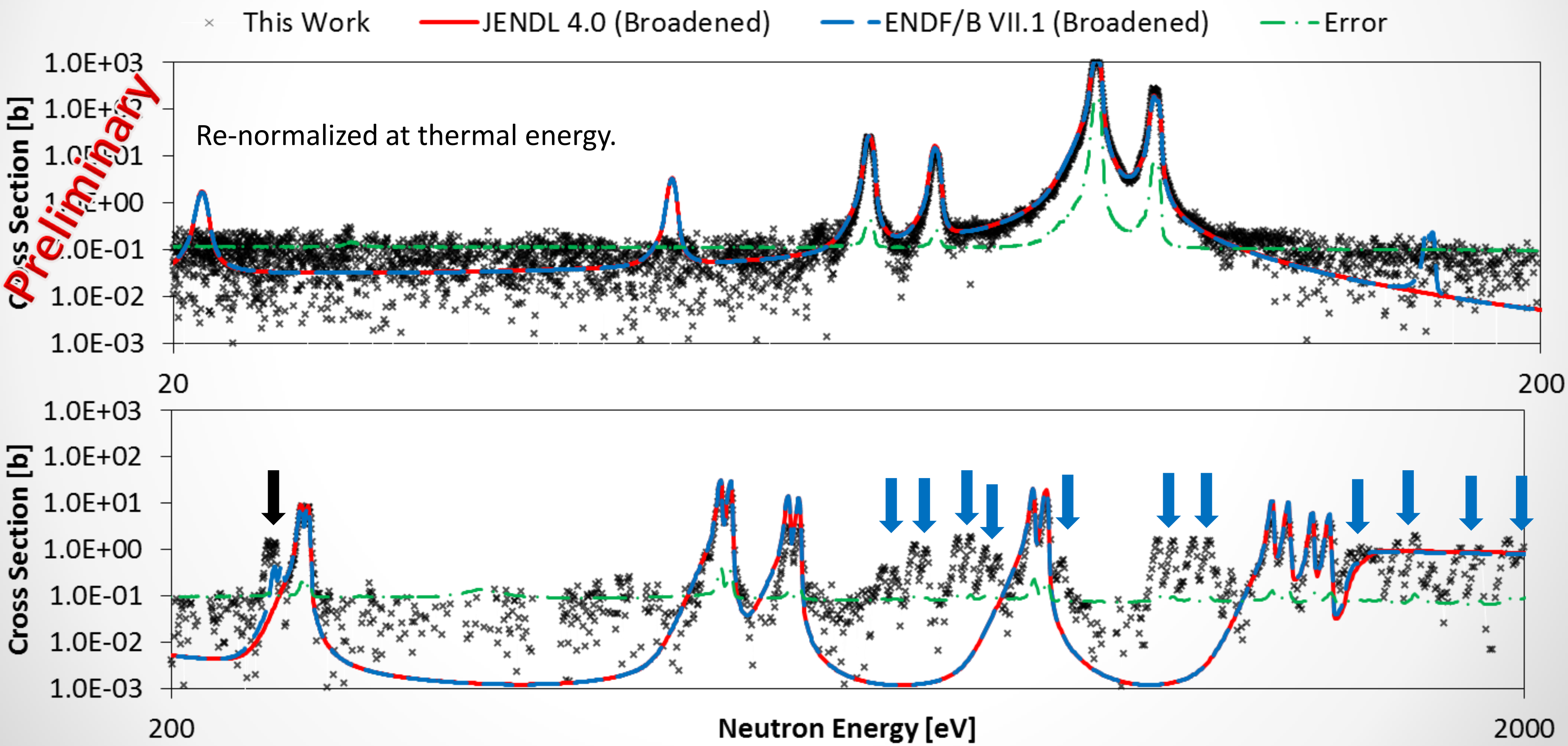
➡ **1st resonance of ^{74}Se (27.1eV)**



Our sample is contaminated with Se(1.33%), Te(830ppm), and Ag(18ppm). However, these impurities are not listed on the certification sheet.

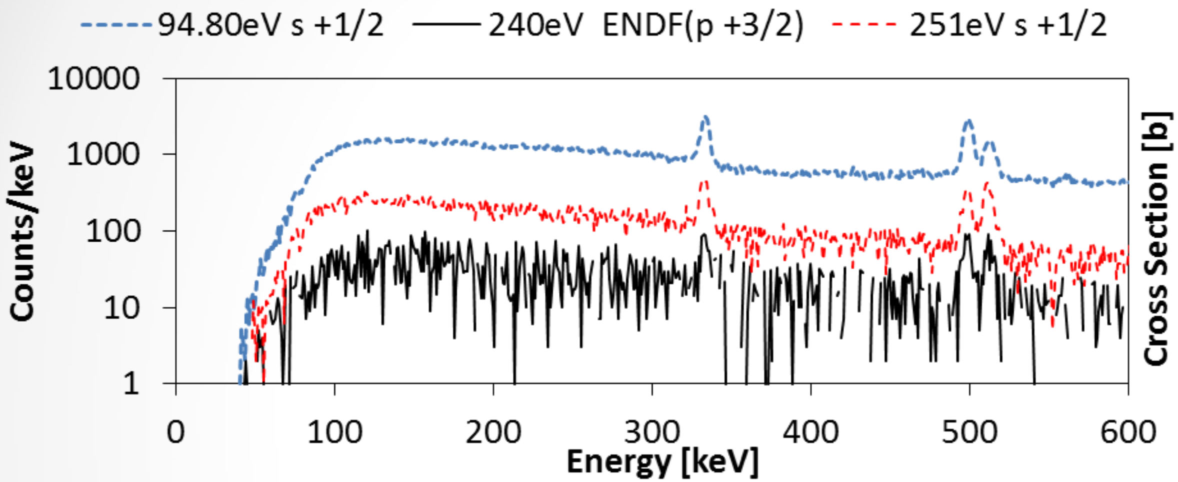
~Cross Section of ^{112}Sn~

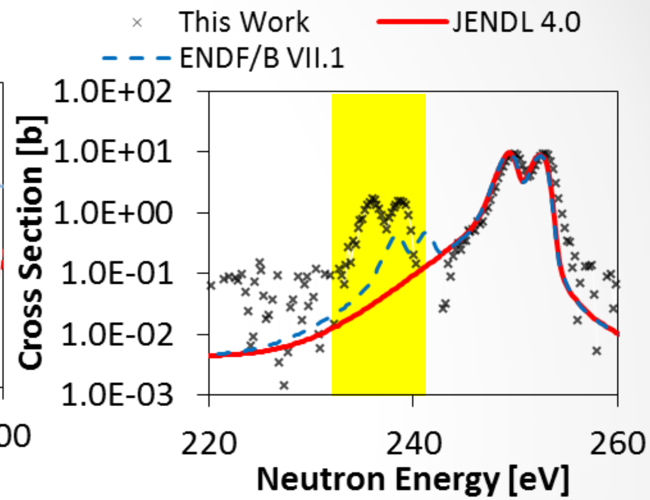
With subtracting the influences of the chemical and isotopic impurities, preliminary cross section for ^{112}Sn was obtained.



Confirm the 240-eV resonance…..

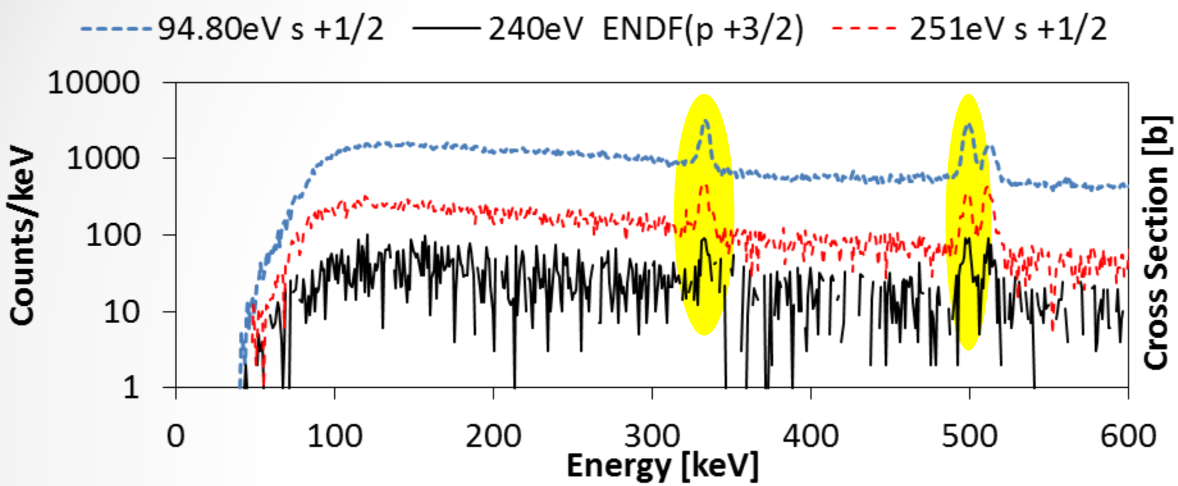
~Origin of the 236.6-eV resonance~

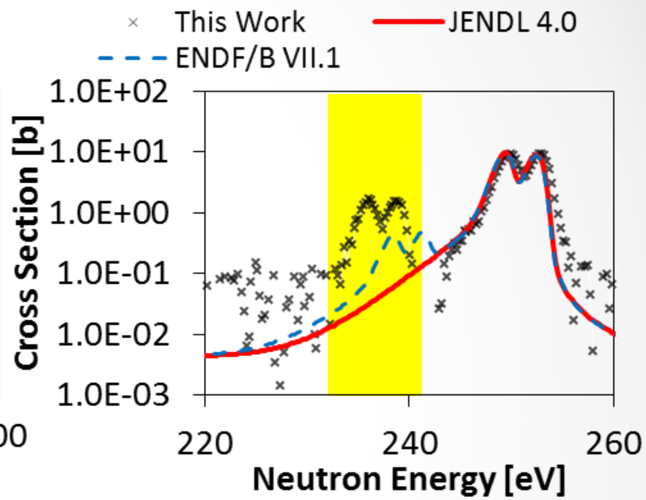




The resonance at 236.6 eV is listed in ENDF B-VII.1 but not listed in JENDL 4.0.

~Origin of the 236.6-eV resonance~



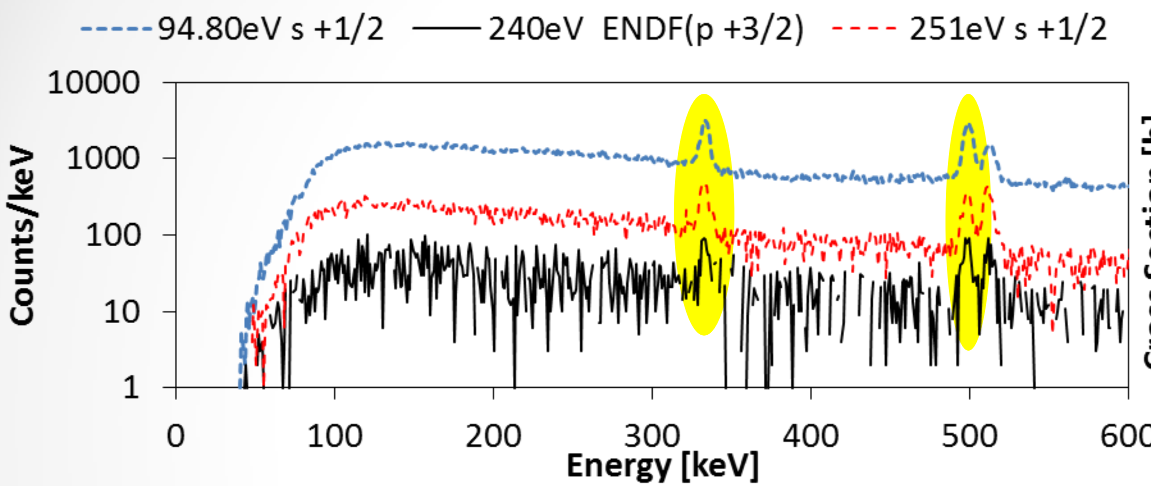


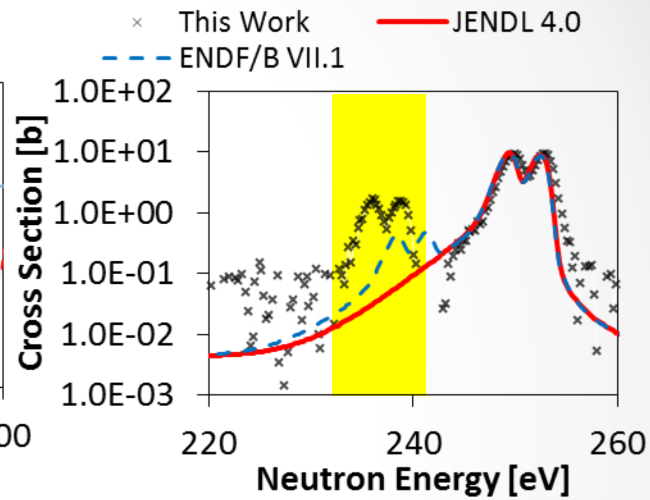
The resonance at 236.6 eV is listed in ENDF B-VII.1 but not listed in JENDL 4.0.

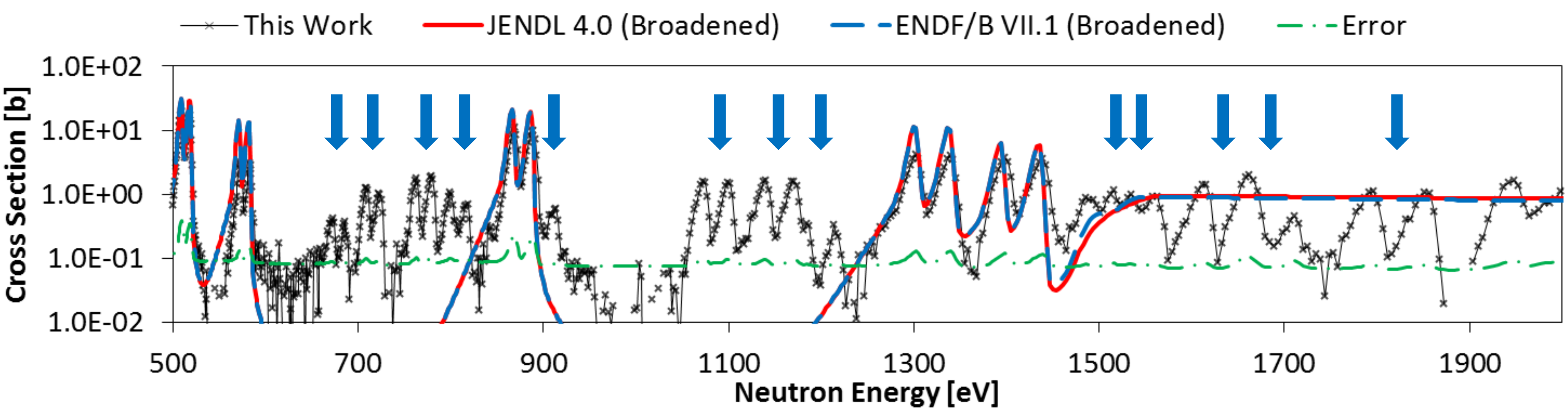
The 332-and 489-keV γ-rays are clearly observed in the 240-eV resonance.

The 236.6-eV resonance is one of the ^{112}Sn resonances.

~Origin of the 236.6-eV resonance~

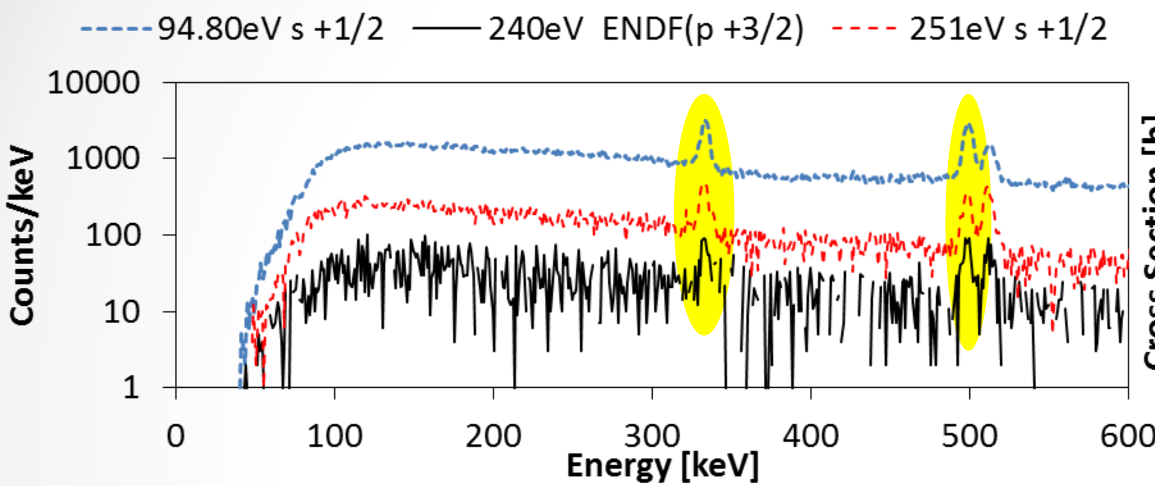


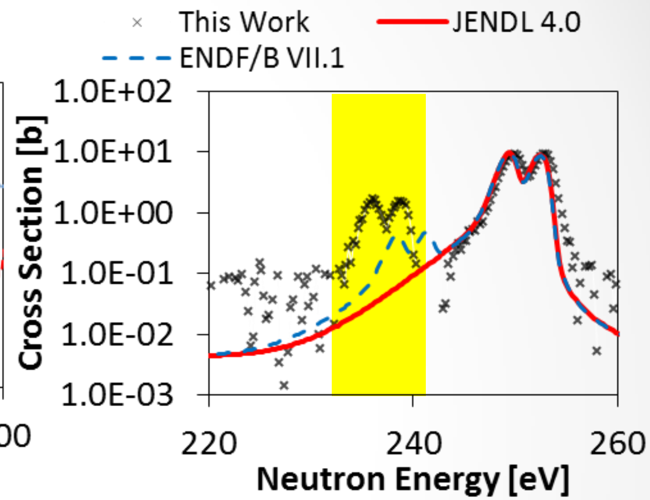


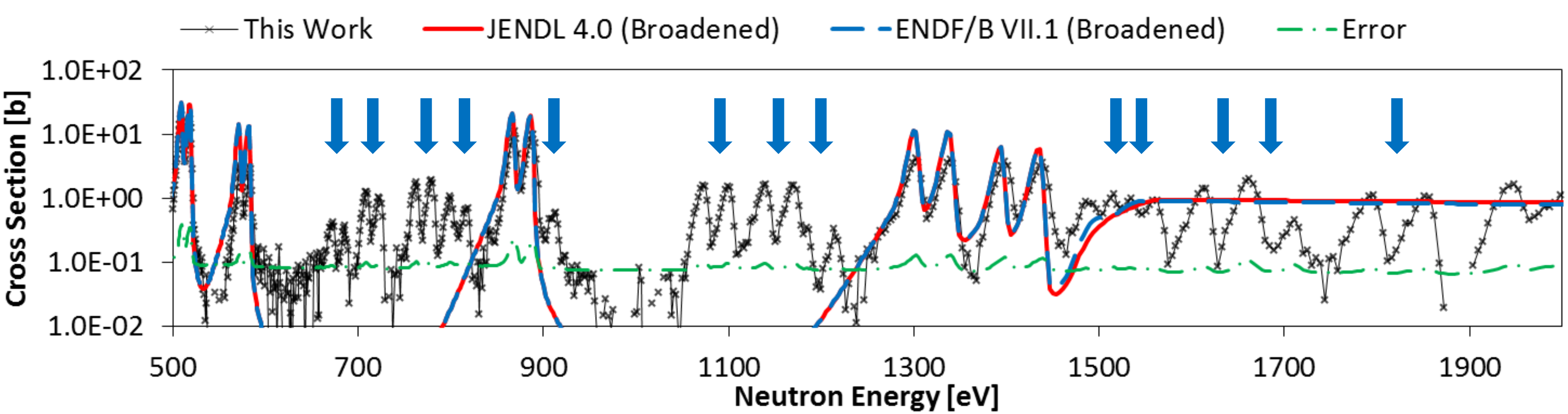


The other resonances were confirmed.

~Origin of the 236.6-eV resonance~







The other resonances were confirmed.

~Resonance energies for ^{112}Sn~

Table Resonance energies for ^{112}Sn comparison to evaluated values.

This work	JENDL 4.0	ENDF/B VII.1	Confirm*	This work	JENDL 4.0	ENDF/B VII.1	Confirm*
Not Observed.	**21.02**	**21.02**		**809.2±1.7**			○
Not Observed.	**40.38**	**40.38**		879.3±1.9	877	877	○
64.61±0.06	64.66	64.66	○	**903.3±2.0**			× **
72.24±0.08	72.26	72.26	○	**1086.0±2.5**			○
94.76±0.11	94.8	94.8	○	**1156.0±2.8**			○
104.65±0.12	104.7	104.7	○	1321±3	1321	1321	○
Not Observed.		**166**		1419±4	1416	1416	○
236.6±0.3		240	○	**1511±4**			△***
252.9±0.4	251.1	251.1	○	**1539±4**			△***
514.0±0.9	514	514	○	**1639±5**			○
577.3±1.1	577	577	○	**1687±5**			× **
679.7±1.3			× **	**1824±5**			○
715.6±1.4			○	**1982±6**			○
771.3±1.6			○				

*: 332-and 498-keV γ-rays
** : The numbers of the events are not enough.
***: Overlapped resonances. (cannot separate clearly).

~Resonance energies for ^{112}Sn~

We found 3 miss assigned resonances and 13 new resonances.

This work	JENDL 4.0	ENDF/B VII.1	Confirm*	This work	JENDL 4.0	ENDF/B VII.1	Confirm*
Not Observed.	**21.02**	**21.02**		**809.2±1.7**			○
Not Observed.	**40.38**	**40.38**		879.3±1.9	877	877	○
64.61±0.06	64.66	64.66	○	**903.3±2.0**			× **
72.24±0.08	72.26	72.26	○	**1086.0±2.5**			○
94.76±0.11	94.8	94.8	○	**1156.0±2.8**			○
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236.6±0.3		240	○	**1511±4**			△***
252.9±0.4	251.1	251.1	○	**1539±4**			△***
514.0±0.9	514	514	○	**1639±5**			○
577.3±1.1	577	577	○	**1687±5**			× **
679.7±1.3			× **	**1824±5**			○
715.6±1.4			○	**1982±6**			○
771.3±1.6			○				

*: 332-and 498-keV γ-rays
** : The numbers of the events are not enough.
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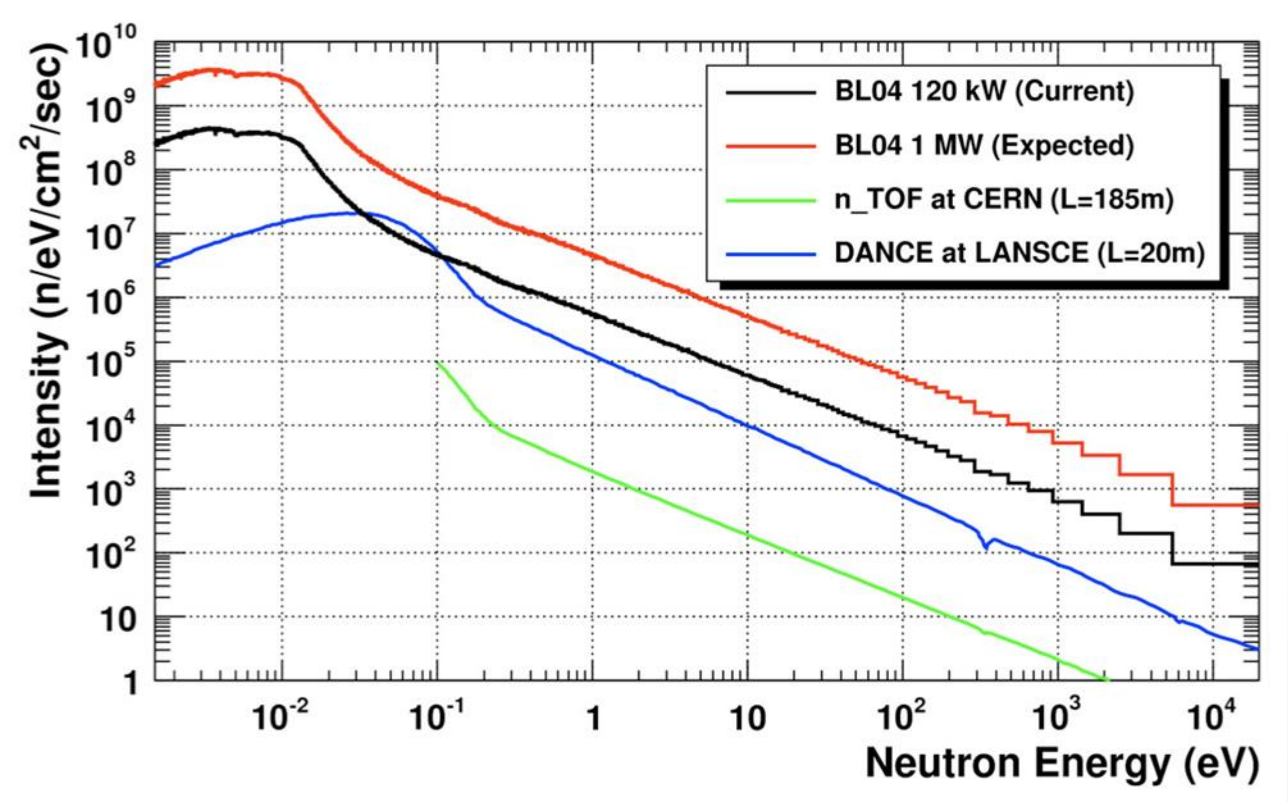
Content

- Introduction
- Our Facility, "ANNRI"
- Example of Measurements
  - Experiment of $^{244 \text{ and } 246}Cm$ (MAs)
  - Experiment of ^{112}Sn (stable isotope)
- Summary

SUMMARY

~Advantages of ANNRI~

- **High intensity pulsed neutron with High speed DAQ.**
  - ✓ A small amount (less than 1 mg) sample can be used.



SUMMARY

~Advantages of ANNRI~

- **High intensity pulsed neutron with High speed DAQ**.
  - ✓ A small amount (less than 1 mg) sample can be used.
- **High γ-ray energy resolution with Ge detectors.**
  - ✓ Prompt γ-ray spectra from resonances can be obtained.
    -> An origin of each resonance can be checked.
    (An impurity nuclide or the target nuclide)
    Spin assignments may be checked.
- **Sufficient beam time.**
  - ✓ In next 2013 JFY, ANNRI will be used in "user program" for 85 days.
    Everybody can apply this program.

SUMMARY

~Future plan~

- To measure neutron capture cross sections of $^{155,157}Gd$, ^{116}Sn, $^{133,135,137}Cs$ and so on.

In order to deduce resonance parameters precisely,

- To prepare neutron total cross section measurements.
- To develop an analysis method from "double-bunched" results using the REFIT-code and the "double-bunched" resolution function.

SUMMARY

We have built a new experimental instrument (ANNRI) , and obtained preliminary neutron-capture cross sections of MAs and LLFPs.

- **Neutron-capture cross sections are deduced using a small amount (less than 1 mg) of a high radioactive sample.**

- **Even a stable isotope, there are many miss-assigned resonances in the evaluated values.**
However, using the array of Ge detectors at the ANNRI and by checking prompt γ-ray spectra, γ rays from impurities can be discriminated.

Thank you
for your kind attention!!