Research on Time-Resolved Prompt Gamma Neutron Activation Analysis at Back-n facility

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Noninvasive Techniques

- **Noninvasive (Nondestructive) Analysis Techniques**: expose the target to an external effect (‘probe particles’) and detect its response (‘answer particles’) to investigate physical-chemical features of the target without damaging it.
- With many large facilities (accelerators, research reactors, and spallation sources) have been constructed, many high quality probe particles are available, and various **Noninvasive Techniques** are developed and applied in **archaeometry, research on extraterrestrial samples etc.**

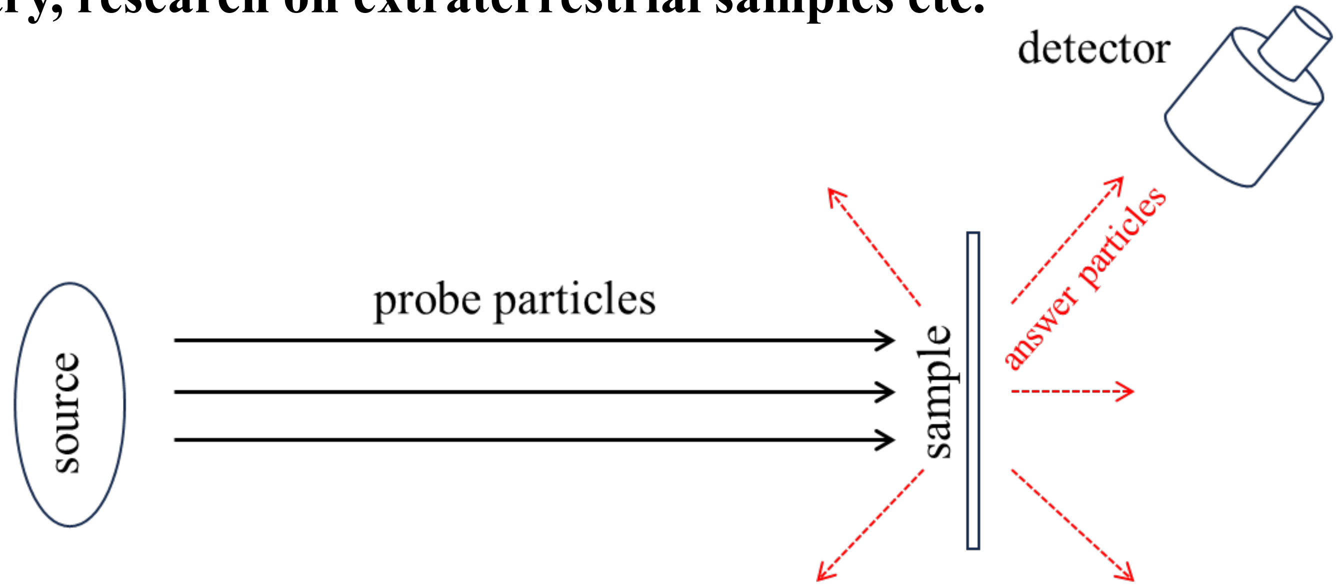

Figure: Basic principles for Noninvasive Techniques .

Noninvasive Techniques

Table: commonly used probe particles and of its noninvasive techniques:

Probe particle	Penetrability	Noninvasive Technique	Answer particle	Information
photon	~μm	RAMAN	photon	chemical bond
		XRD	X-ray	lattice structure
		XRF	X-ray fluorescence	elemental composition
		XAS	X-ray	chemical state
neutron	~cm	TOF-ND	neutron	lattice structure
		PGAA, T-PGAA	prompt γ-ray	elemental and isotopic composition
		NRA(NRCA, NRTA)	prompt γ-ray, neutron	elemental and isotopic composition
		NAA	decay γ-ray	elemental and isotopic composition
		NI	neutron	elemental and isotopic composition, geometry
muon	~cm	muon-fluorescence	X-ray fluorescence	elemental and isotopic composition
electron	~μm	EMPA	X-ray fluorescence	elemental composition
Ion	~μm	RBS	Ion	elemental composition lattice structure

Noninvasive Techniques with neutrons

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- Well developed and widely used non-destructive analysis methods using neutrons as probes: **neutron activation analysis(NAA), Prompt Gamma-ray Neutron Activation Analysis(PGAA).**
- **PGAA:** mainly used for **light elements** (Ca and below) and few **high-cross section elements** (Cd, Sm, Gd).
- **NAA:** 195/287 stable or long-lived isotopes exsit neutron activated delay gamma-rays.
- **Conclusion**: Noninvasive Techniques with neutrons may lack isotopic analysis capability for many isotopes.

Typical instrumental sensitivities in mg/kg (E + 4 denotes 10^4, etc.) for thermal NAA of elements in light element matrix (e.g., organic, silicate) biological and environmental samples

INAA PGAA Both

H 10																	He
Li	Be											B 0.05	C E+4	N E+3	O	F 50	Ne
Na 0.1	Mg 100											Al 1	Si	P	S 200	Cl 10	Ar
K E+3	Ca E+3	Sc E–3	Ti 100	V 0.1	Cr 0.01	Mn 0.01	Fe 10	Co 0.01	Ni	Cu 10	Zn 1	Ga 0.1	Ge	As 0.01	Se 0.1	Br 1	Kr
Rb 1	Sr 100	Y 1	Zr 50	Nb	Mo 1	Tc	Ru	Rh	Pd	Ag 0.1	Cd 0.1	In E-4	Sn 100	Sb 0.01	Te	I 1	Xe
Cs 0.1	Ba 10	La E-3	Hf 0.1	Ta 0.2	W 0.1	Rc	Os	Ir	Pt 0.05	Au E-4	Hg 0.01	T1	Pd	Bi	Po	At	Rn
Fr	Ra	Ac	104	105	106	107	108	109									
			Ce 0.01	Pr	Nd 1	Pm	Sm 0.01	Eu E-3	Gd 0.1	Tb 0.1	Dy 0.01	Ho	Er	Tm	Yb 0.1	Lu 0.1	
			Th 0.5	Pa	U 0.1	Np	Pu	Am	Cm	Bk	Cf	Es	Fm	Md	No	Lr	

Time-resolved Prompt Gamma Neutron Activation Analysis(T-PGAA)

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- In 2014, Yosuke Toh *et al.*[1] firstly proposed a new analytical technique so called **Time-resolved Prompt Gamma-ray Neutron Activation Analysis(T-PGAA)**, which combines prompt gamma-ray analysis and time-of-flight (TOF) technique to **get better gamma-ray peak selectivity and signal-to-noise ratios** (SNR).

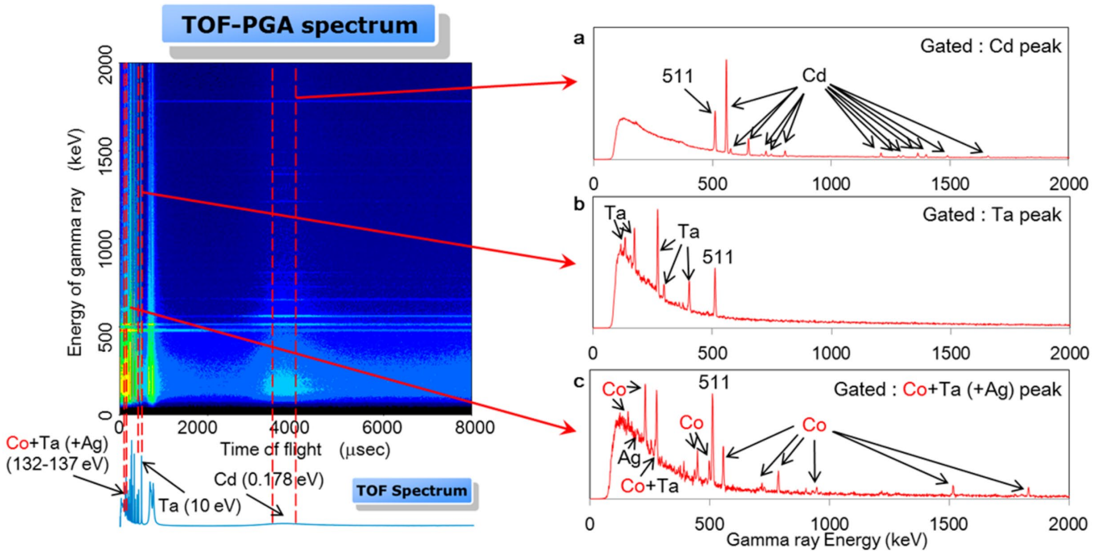

Figure: TOF-PGA spectrum analysis by gating on different gamma-ray peaks.[1]

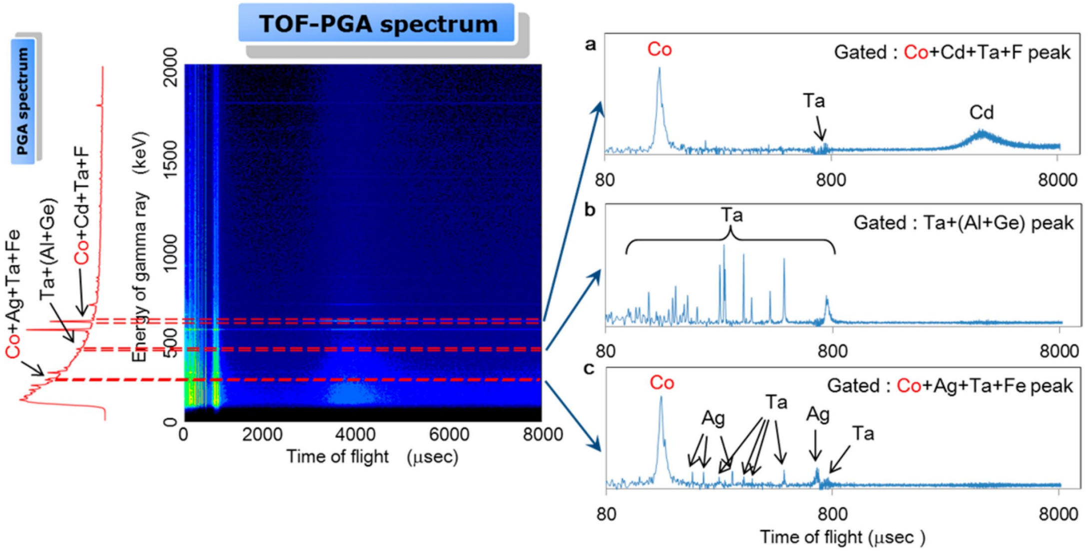

Figure: TOF-PGA spectrum analysis by gating on the TOF peaks.[1]

[1] Yosuke Toh, Mitsuru Ebihara, *et al.* Anal. Chem. 86, 24, 12030–12036 (2014).

Features for T-PGAA Technique

- Features of T-PGAA (compares to NAA and PGAA):
  - ① T-PGAA requires pulse epithermal neutron source (ToF technique);
  - ② T-PGAA utilizes epithermal neutron, in this region, similar to resonance absorption in atomic spectrum, neutron capture cross section appears many narrow and high resonance peak structures.

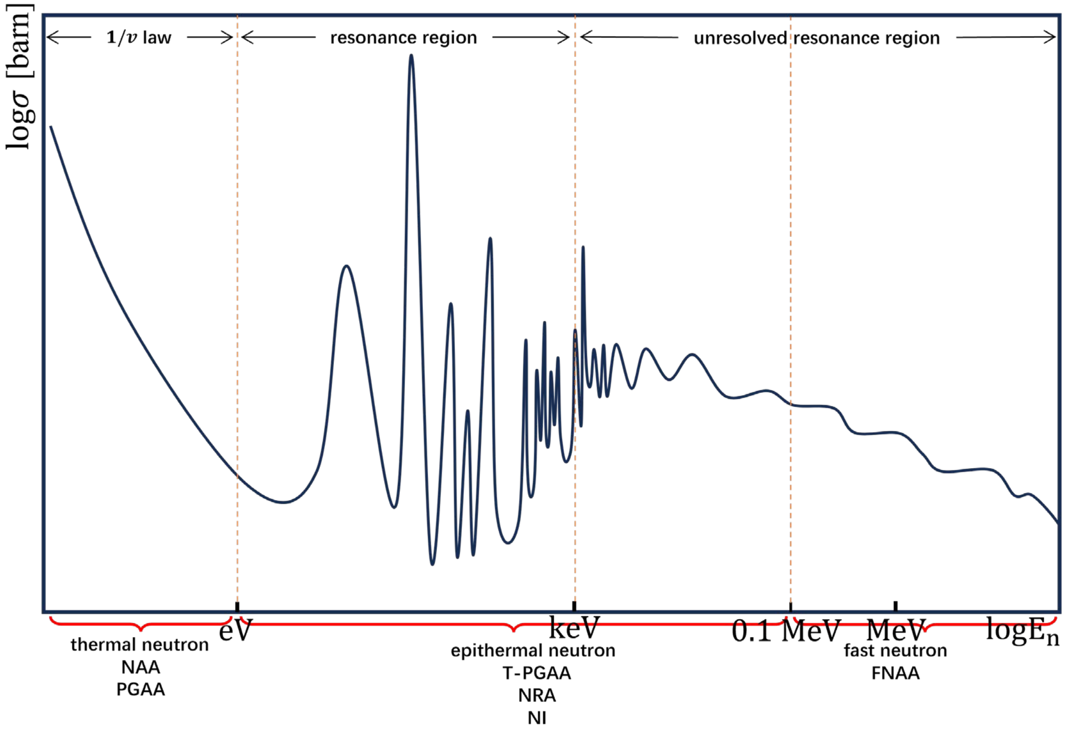

Figure: neutron reaction features for different Noninvasive Techniques.

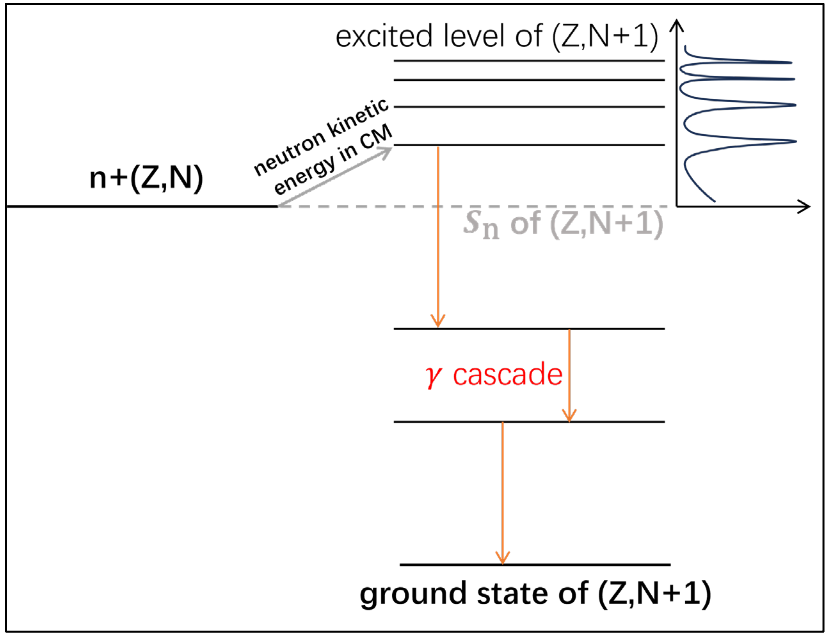

Figure: resonance absorption for neutron

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Back-n facility

Back-n white neutron source(@CSNS) provide us:

- Paused neutron for energy from 0.3 eV to 300 MeV, which covers whole epithermal neutron energy range;
- In #ES2 of Back-n, the neutron flight path ~ 78 m, provide a good energy resolution for ToF technique;
- The neutron flux at #ES2 can reach 1.12×10^7 neutrons/cm^2/s (measured in 2024.3 @160kW), high neutron flux means a better sensitivity and lower detect limit for T-PGAA;

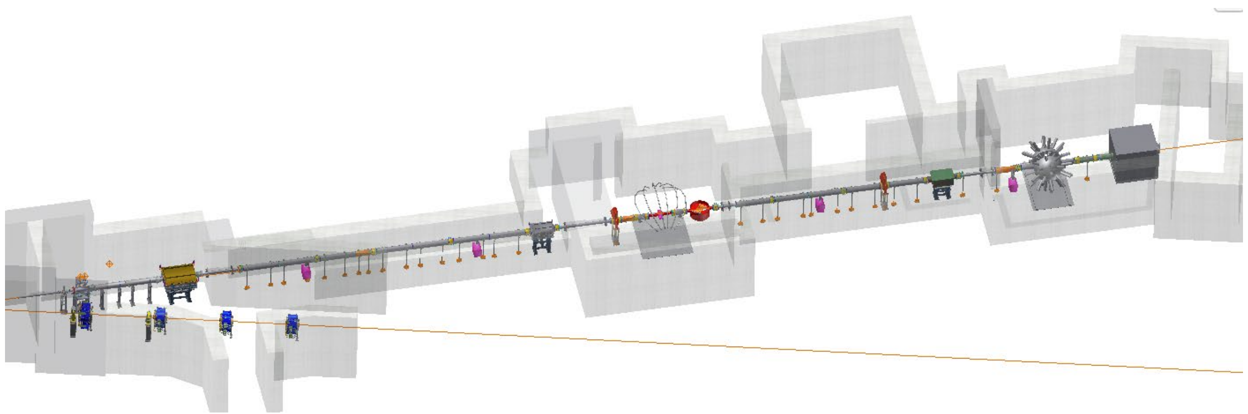


Figure: layout of Back-n facility.

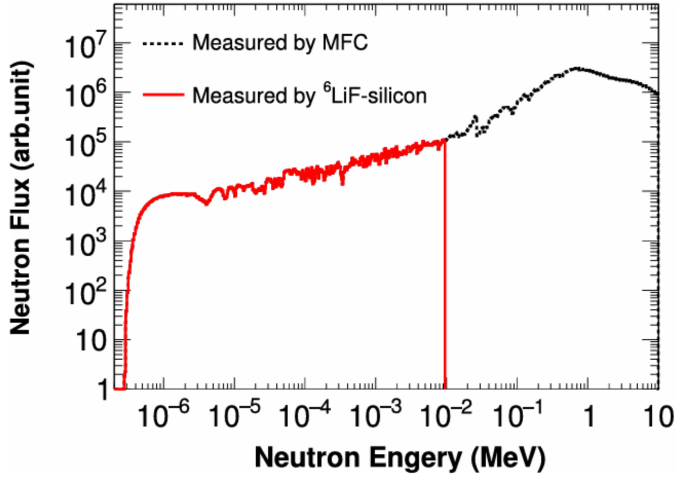


Figure: neutron flux spectrum for Back-n #ES2.

Lu target HPGe experiment

In 2023, Ahmadov Gadir *et al.*(JINR) perform a in-beam experiment, measure Lu target with HPGe detector:

- We share the data, but for different analysis purpose;
- We analysis the data to confirm the beam conditions and detector system of Back-n may be suitable for the requires of T-PGAA research.

Table: experimental conditions for Lu target HPGe experiment.

target	size(mm)	Beam power (kW)	Beam size (mm)	time(h)
^{nat}Lu	$\Phi 40 \times 1$	160	ϕ50-50-58	~168
NaCl	$\sim 50g$			~8
empty	——			~9

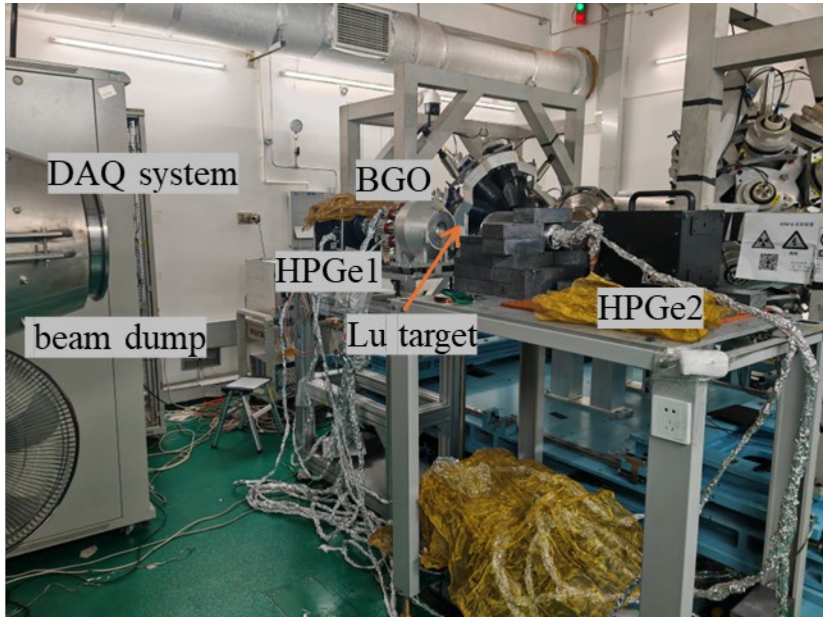


Figure: experimental setup for Lu target.

^{nat}Lu: 97.41% ^{175}Lu
2.59% ^{176}Lu

HPGe detector calibration

HPGe detector calibration:

- Radiation source ^{60}Co, ^{152}Eu is adapted for low-energy gamma-ray energy and absolute efficiency calibration;
- Reaction of $^{35}Cl(n,g)$ is adapted for high-energy gamma-ray and relativly efficiency calibration.

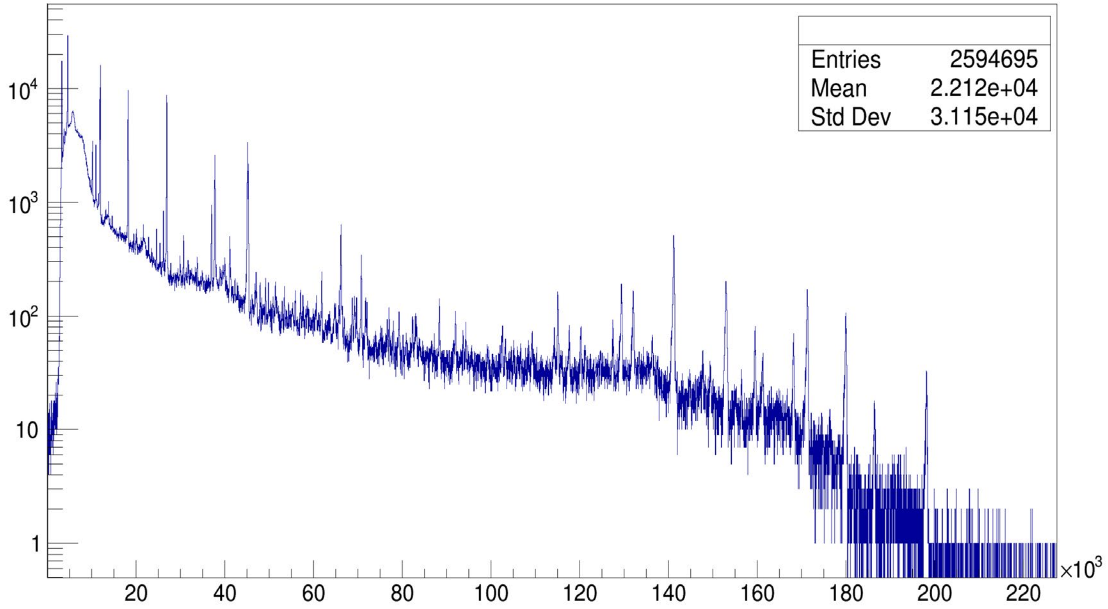


Figure: NaCl gamma-ray spectrum.

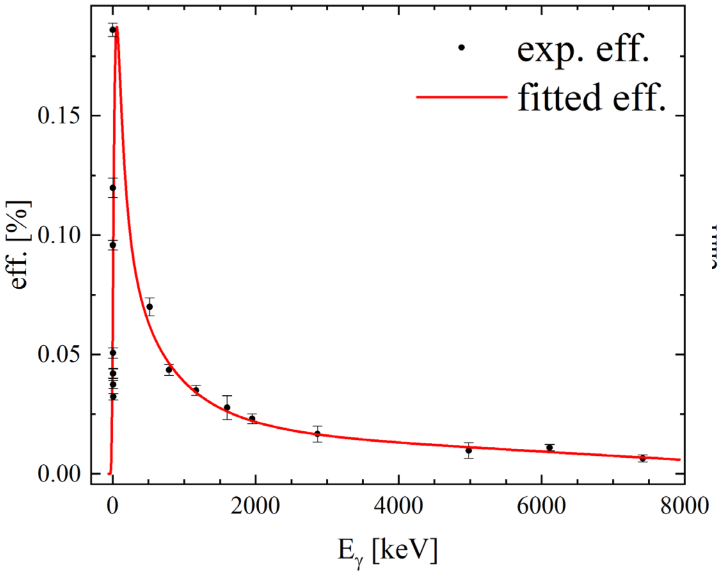


Figure: HPGe efficiency calibration.

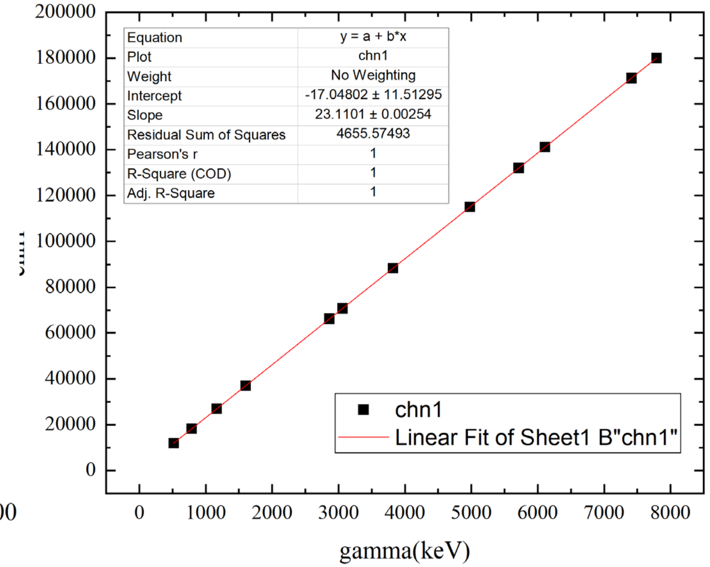


Figure: HPGe energy calibration.

Experiment results

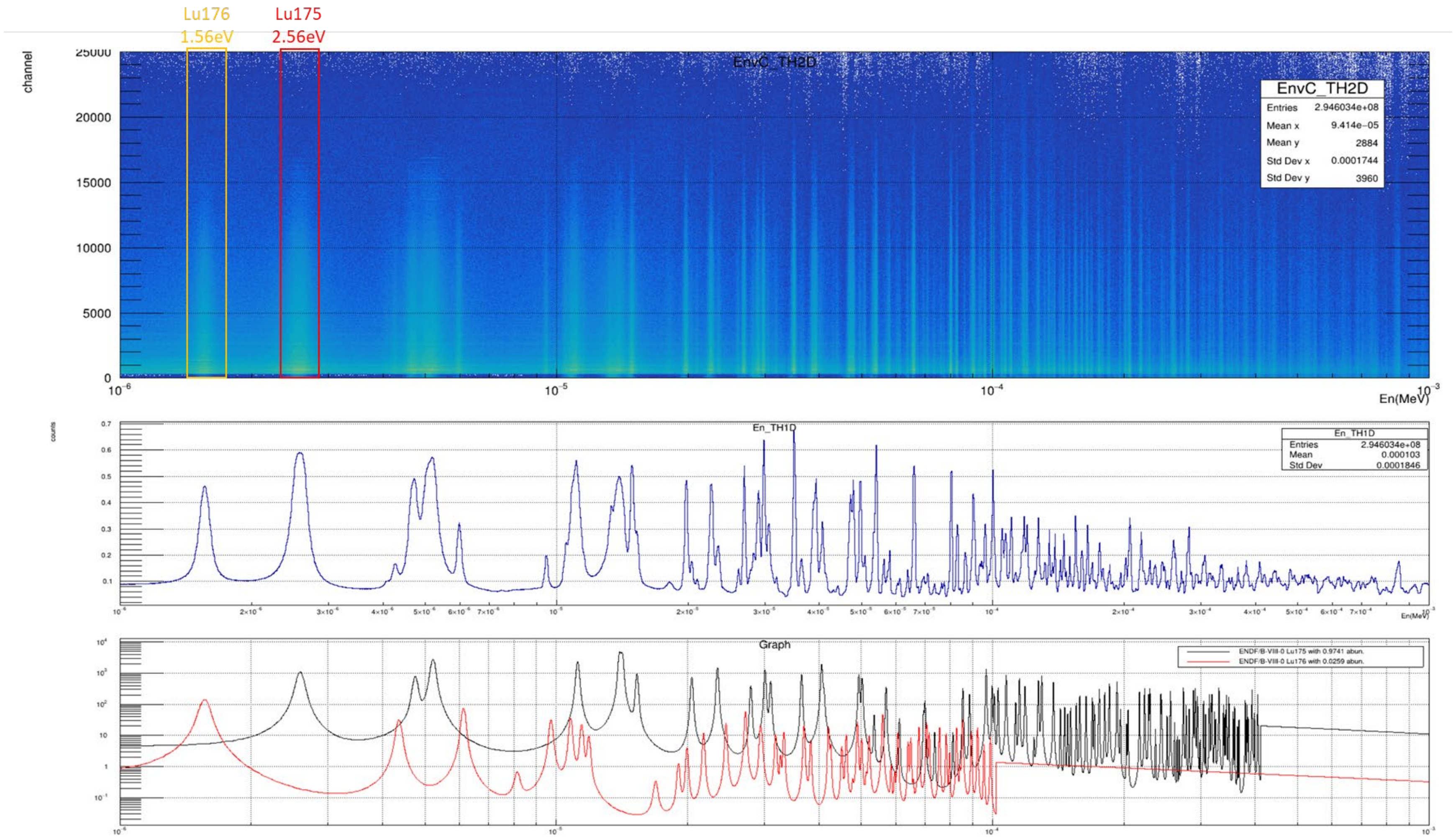


Figure: Lu target results: (top) ToF neutron energy vs gamma-ray energy 2-D spectrum; (middle) ToF neutron energy spectrum; (bottom)Lu ENDF/B-VIII cross section spectrum.

ToF Gated gamma-ray spectrum

Strong resonance structues of ^{175}Lu and ^{176}Lu can be obviously observed in ToF energy spectrum and gated:

- **Much better** gamma-ray peak selectivity and signal-to-noise ratios can be reached by ToF gating;
- Back-n beam condition and HPGe detector is **suitable** for T-PGAA research.

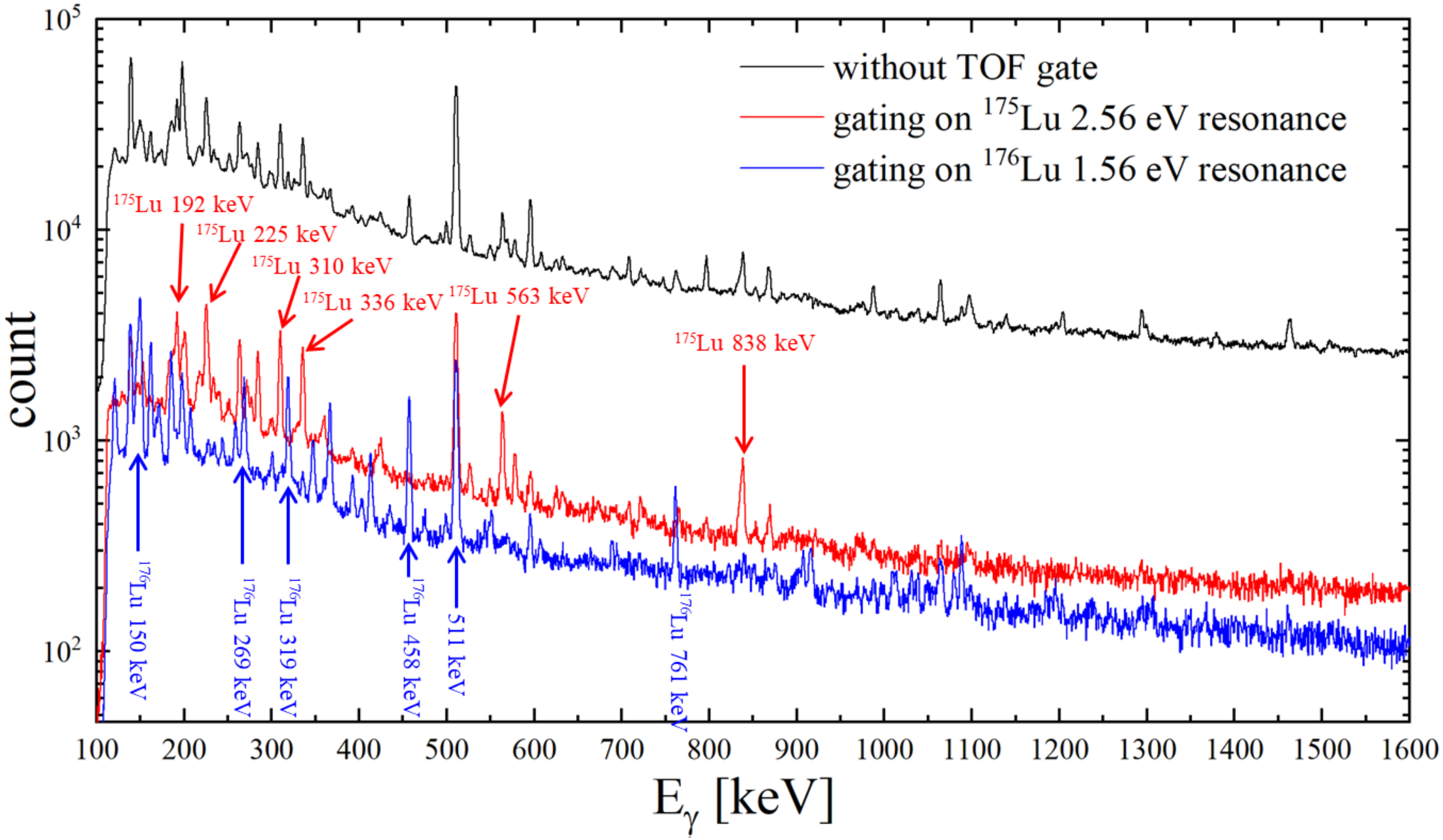


Figure: ToF Gated gamma-ray energy spectrum

T-PGAA quantitative analysis

quantitative analysis: net area of gamma-ray full-energy peak $N_{x,\mu,k}$ follows the relationship:

Neutron capture yield

$$N_{x,\mu,k} = \boxed{n_x F(E_\mu)\sigma_x(E_\mu)}\,\Omega\Phi(E_\mu)\varepsilon(E_k)\, f_{x,\mu,k}t,$$

$$F(E_\mu) = \frac{1 - e^{-\sum_j n_j \sigma_{j,\mathrm{tot}}(E_\mu)}}{\sum_j n_j \sigma_{j,\mathrm{tot}}(E_\mu)}$$

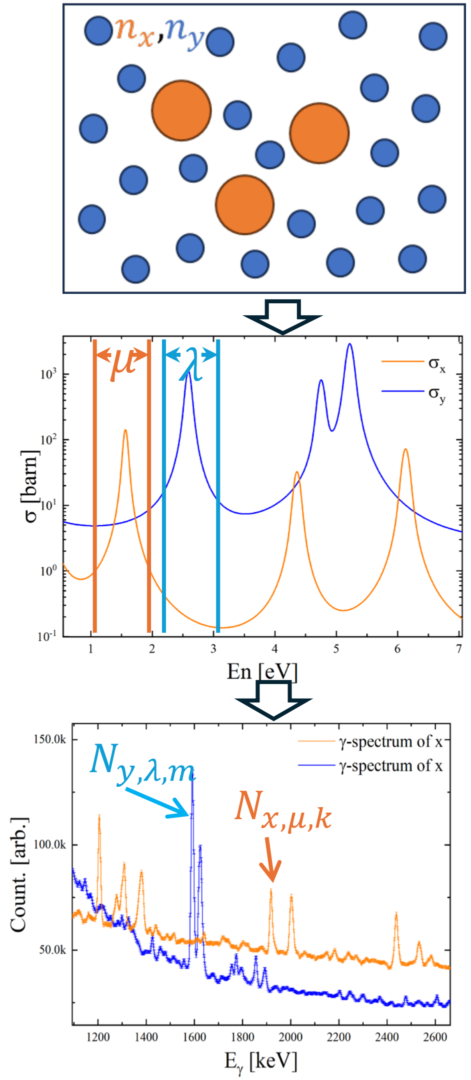
$\boldsymbol{F(E_\mu)}$: neutron self-shield factor $\boldsymbol{\Phi(E_\mu)}$: neutron flux

$\boldsymbol{\Omega}$: cross area of beam and target $\boldsymbol{\varepsilon(E_k)}$: detector efficiency

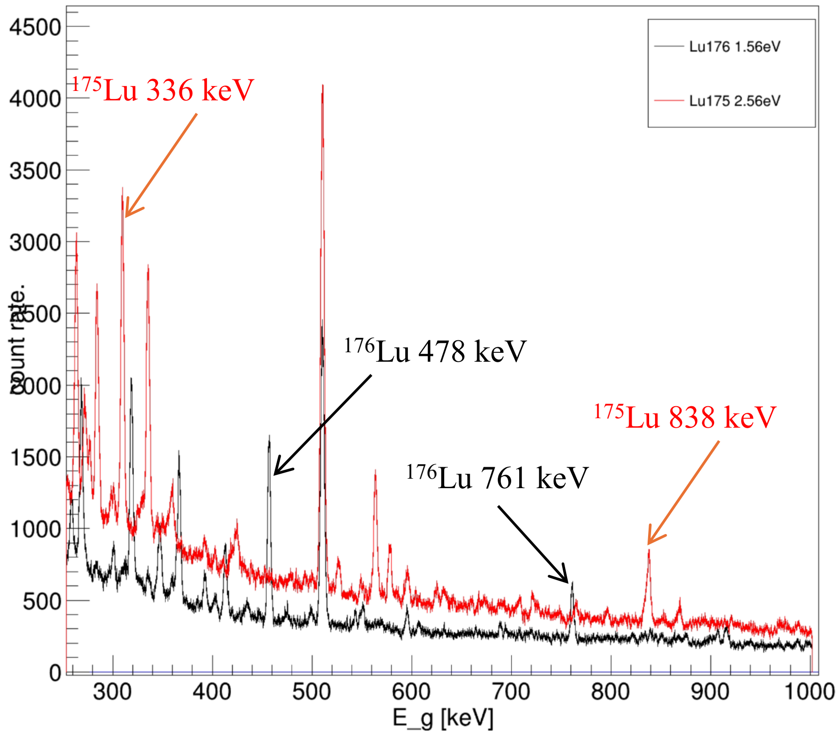
$\boldsymbol{f_{x,\mu,k}}$: branch ratio for gamma-ray(k) of nuclide (x) at the resonance peak (μ);

Relatively isotopic abundance ratio can be calculated by $N_{x,\mu,k}$ with equation:

$$\frac{n_x}{n_y} = \frac{N_{x,\mu,k} F(E_\lambda)\sigma_y(E_\lambda)\Phi(E_\lambda)\varepsilon(E_m)\, f_{y,\lambda,m}}{N_{y,\lambda,m} F(E_\mu)\sigma_x(E_\mu)\Phi(E_\mu)\varepsilon(E_k)\, f_{x,\mu,k}} \qquad (1)$$



quantitative analysis results



Relatively isotopic abundance ratio for ^{176}Lu/ ^{175}Lu:

- Quantitative results are near the theoretical ^{176}Lu/ ^{175}Lu value ~ 0.265.
- Fluctuation of results with different gamma-ray peaks are bigger than statistical error.

➢ For neutron resonance absorption, different resonance response to different level of compound nuclei, which means **different resonance have different gamma-ray decay branch ratio**. But, at present, we **lack the gamma-ray database in resonance region**.

<table>
<tr><th>^{175}Lu resonance</th><th>^{175}Lu gamma</th><th>^{176}Lu resonance</th><th>^{176}Lu gamma</th><th>^{176}Lu/^{175}Lu</th></tr>
<tr><td rowspan="4">2.56 eV</td><td rowspan="2">838 keV</td><td rowspan="4">1.56 eV</td><td>761 keV</td><td>0.0245</td></tr>
<tr><td>458 keV</td><td>0.0180</td></tr>
<tr><td rowspan="2">335 keV</td><td>761 keV</td><td>0.0265</td></tr>
<tr><td>458 keV</td><td>0.0195</td></tr>
</table>

Table : quantitative analysis results with different gamma-ray peaks

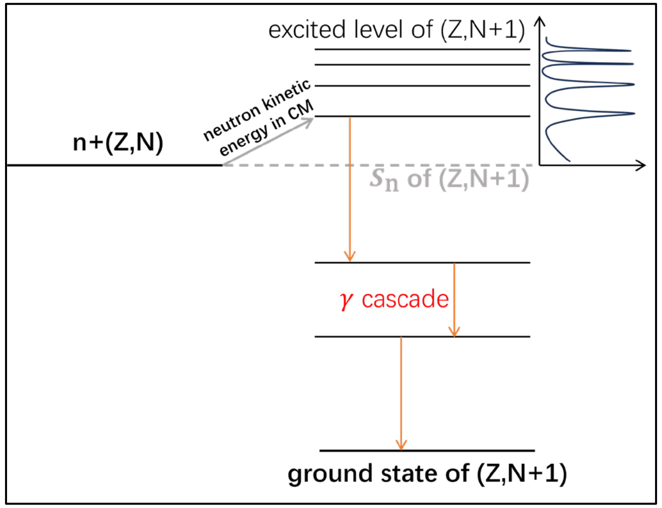


Figure: resonance absorption for neutron

conclusion

Summary:

- With Lu experimental data, we verified the Back-n beam condition and HPGe detector capability for T-PGAA research;
- T-PGAA can reach better gamma-ray peak selectivity and SNR;
- We can get quantitative results near the actual value, but exist big fluctuation caused by the lack of gamma-ray database in resonance region.

Future plan:

- Design and construct a shield system for HPGe detector for shielding neutron and gamma-ray background, to get better SNR and detect limits;
- Design and perform new experiments and develop new relative quantitative method to avoid use of resonance region gamma-ray database to get better quantitative results.

Thanks for your attention!