Neutron response of Gd-doped glass scintillators

Glass Scintillator Collaboration
闪烁玻璃合作组
Glass Scintillator Collaboration

Ruiqiang Song, Sen QIAN

qians@ihep.ac.cn; On Behalf of the GS R&D Group, The Institute of High Energy Physics, CAS

CALOR Meeting-TSUTUBA -2024. 05. 20th

Outline

- 1. Applications and Requirements of Neutron Detection
- 2. The R&D of Gd-doped GS
- 3. Neutron detection experiment
- 4. Summary

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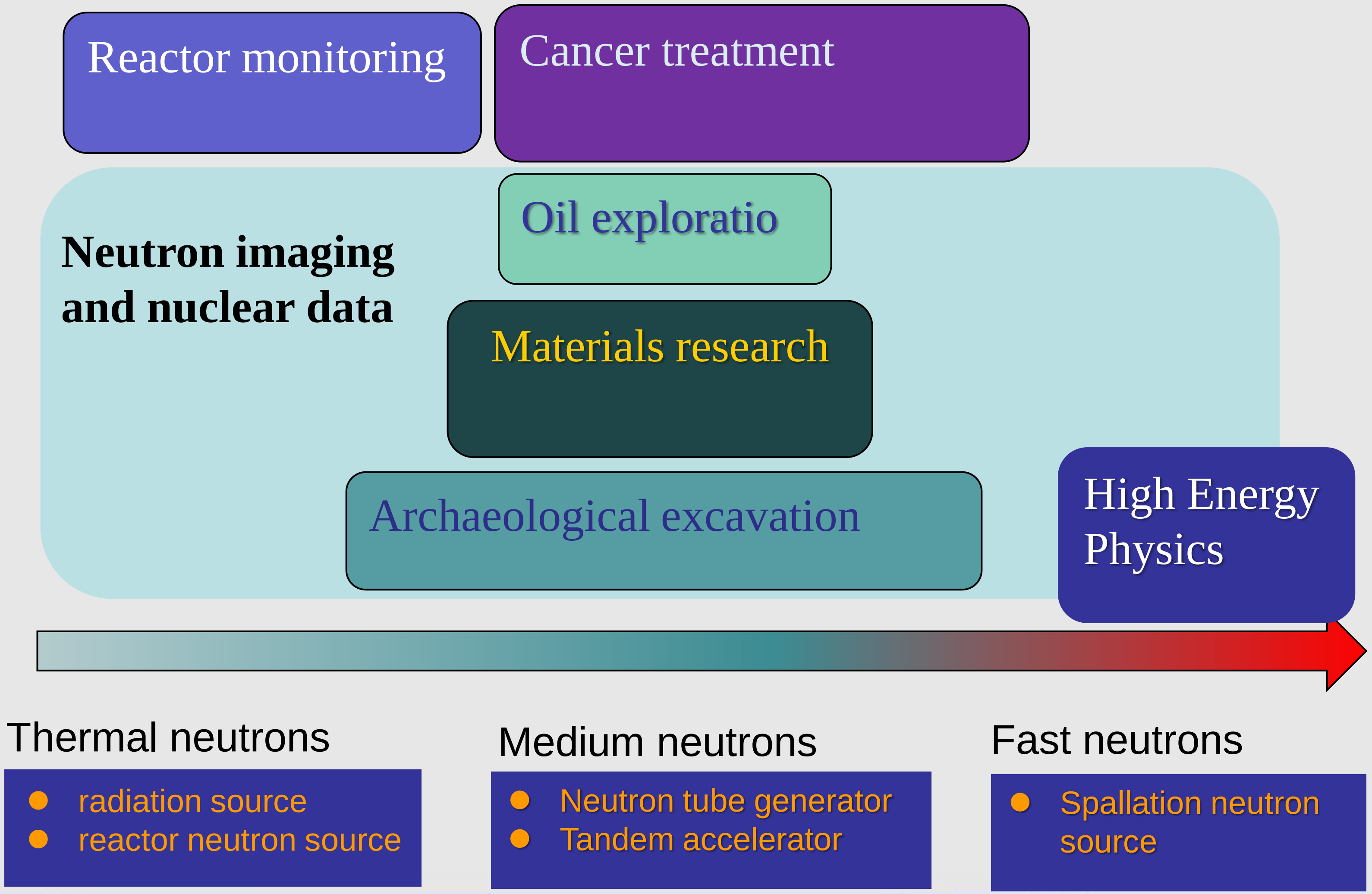
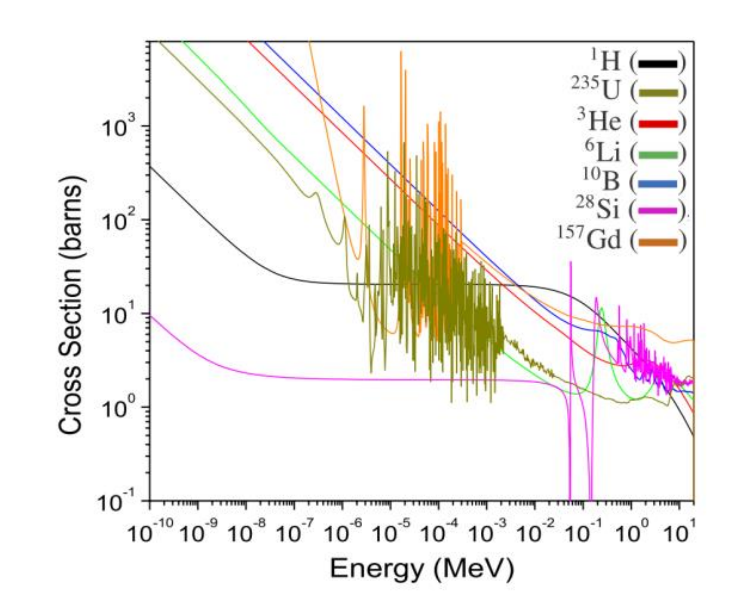
1.1 Applications of Neutron Detection

- **Neutron detection is a widely used non-destructive testing technique**
- Neutron scattering experiment
  - neutron diffraction
  - small angle scattering
  - Inelastic scattering
  - Reflectivity
- Neutron imaging
- Oil exploratio
- archaeological excavation
- Cancer treatment（BNCT）
- Semiconductor doping
- Neutron irradiation effect
- Reactor monitoring
- Defence and security



1.2 Neutron detection materials



Data from ENDF(https://www-nds.iaea.org)

Nuclides	Reaction products	Thermal neutron cross-section（b）
^{1}H	Recoil P	30
^{10}B	$\alpha + {}^{7}$Li	3840
^{3}He	p + ^{3}H	5330
^{6}Li	$\alpha + {}^{3}$H	940
^{157}Gd	γ + $^{-}$e	24000
^{113}Cd	γ（558.6 & 651.3keV）	20000

Materials Critical to Neutron Detection

- low energy neutron : High-efficiency detection is the mainstay, ^{3}He,^{10}B,^{6}Li,^{157}Gd, ^{235}U et al. . The main detection method is the nuclear reaction method.
- Fast and high-energy neutrons: In the case of high-energy neutrons, more attention is paid to irradiation resistance, time response. Diamond, SiC, Irradiation-resistant scintillators etc.

1.3.1 Neutron detector (^3He)

- Neutron detection
- high thermal neutron cross-section(5333 barn)
- high detection efficiency (>70%@thermal neutron)
- high n/γ suppression ratio
- Enables large area detection(^3He tube arrays)

expensive

Current $ ~2750 per liter [*Energy 6134 2020 13-22*]

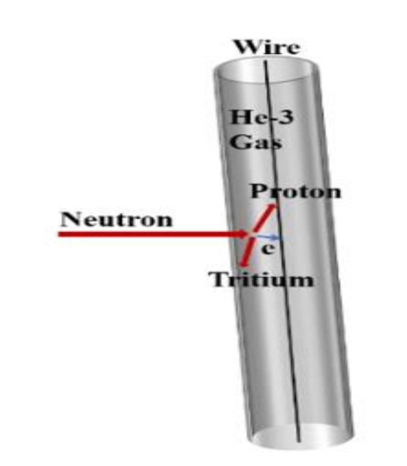
Allocated Helium-3 prices per liter (in dollar)

Customers	2009	2010	2011
Federal agencies and their grantees	450	365	600
Commercial and nonfederal agencies	450	365	1000
Medical users	600	485	720

[Data from GAO(U.S. Government Accountable Office)]

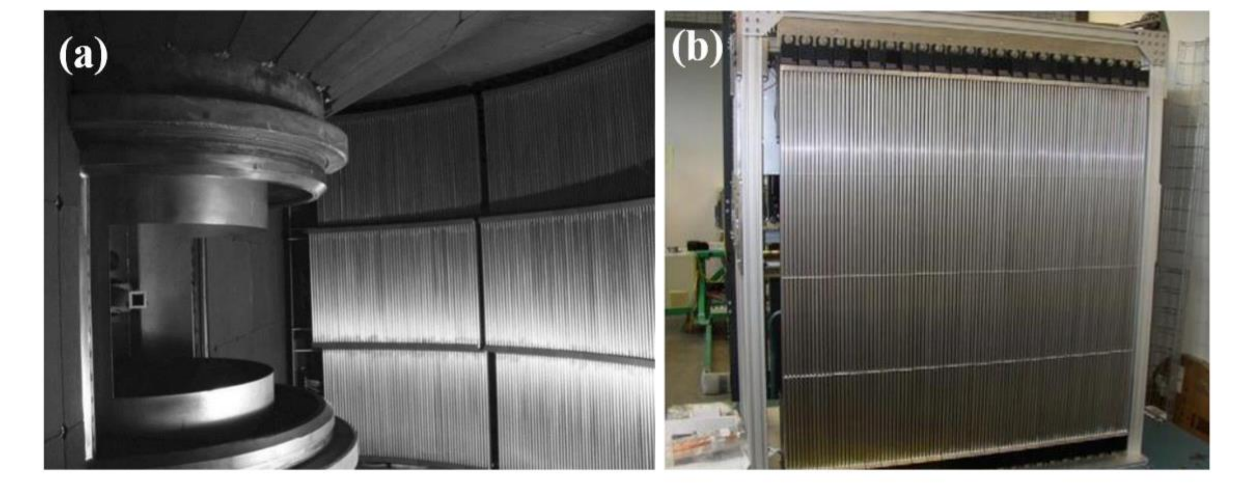
Prices

The supply of ^3He gas has become severely deficient because of the continuous depletion of ^3He sources and the increasing demand of neutron detectors



^3He & Neutrons

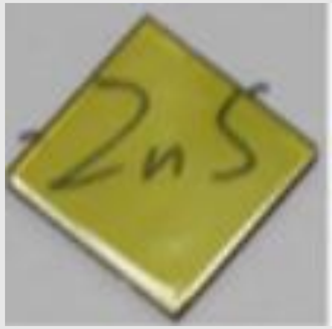
^3He tube



SNC ARCS spectrometer

1.3.2 Neutron detector (^{6}Li-based scintillator)

CAS

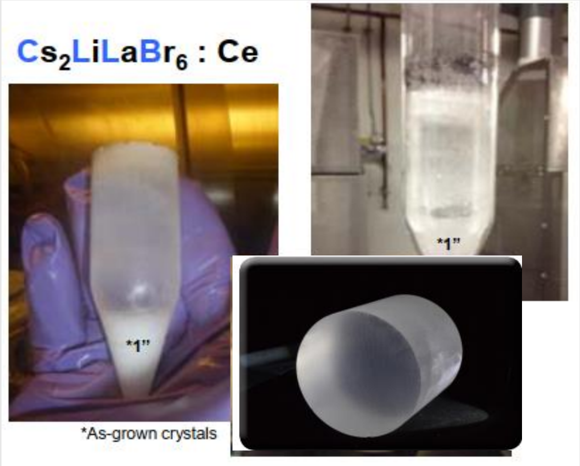


https://sic.cas.cn/

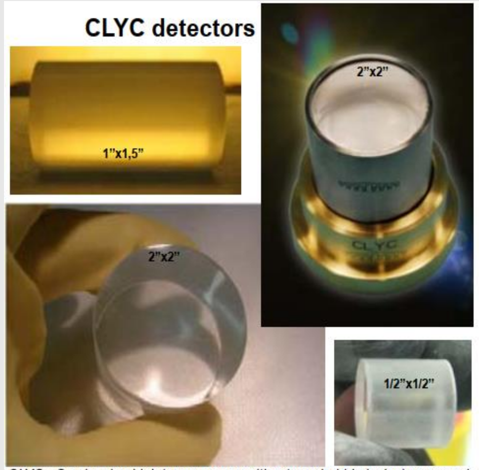
SCINTACOR

https://scintacor.com/

Saint-Gobain



RMD



CLYC : Ce showing high transparency without any bubble inclusion or crack

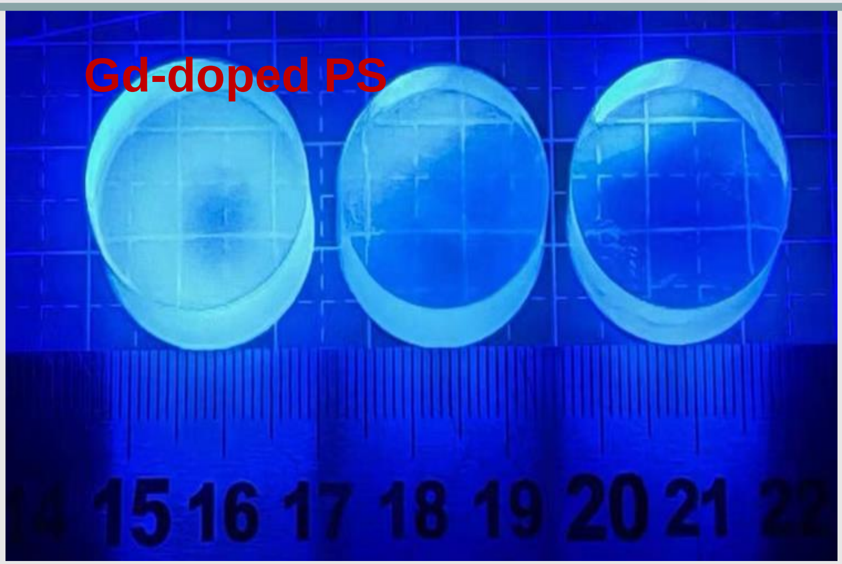
NIMA 729 (2013) 747

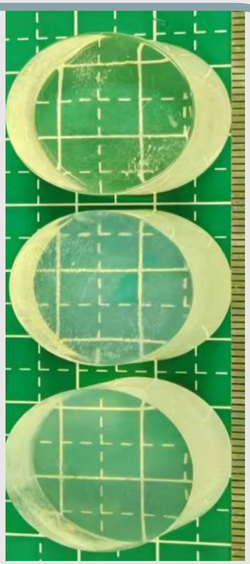
- **ZnS:Ag/^{6}LiF:** thermal neutron detection, high neutron light yield (~150,000 ph/n)[Neutron News, 17, 2006, 16-18], long afterglow, low energy resolution, opacity, highly malleable.
- **GS20 glass:** thermal neutron detection, relatively fast decay time(~70ns)[Neutron News, 17, 2006, 16-18] suitable for high count rate neutron detection, highly malleable.
- **Cs_2LiYCl_6(CLYC):** thermal and fast neutron detection, high light yield , n/γ discrimination
- **Cs_2LiYBr_6:Ce (CLLB):** thermal neutron detection, high light yield , n/γ discrimination
- **^{6}Li-doped plastic scintillator:** thermal and fast neutron detection, low light yield , fast decay time, n/γ discrimination

1.3.3 Neutron detector (Gd-based scintillator)

NIMA 105 (2022) 109964

CAS https://sic.cas.cn/



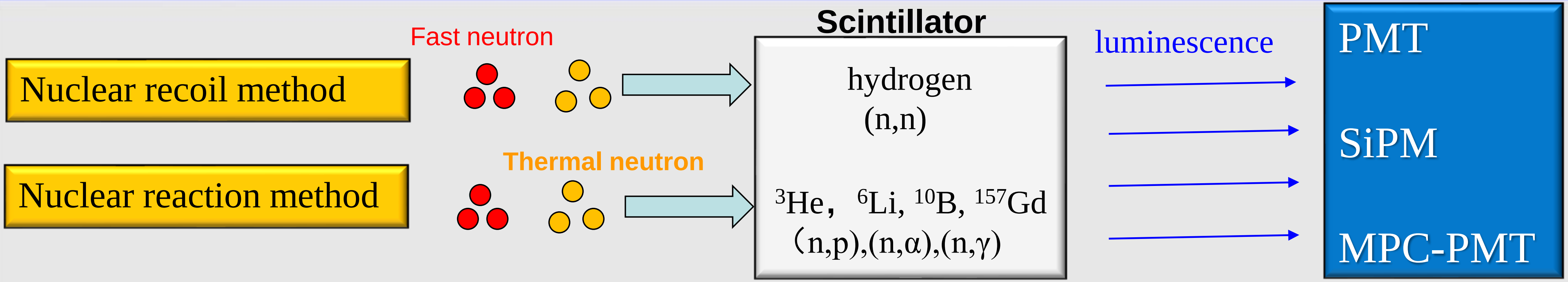


Yangzhou University (China)

- **GAGG:** thermal neutron detection, high light output， fast decay time(~100ns) [SDAE 840 2015 186]
- **GOS:** thermal neutron detection, high light output, high density and short afterglow
- **Gd-doped PS:** thermal and fast neutron detection Large-size, High detection efficiency， low cost

Gd(n, γ) has high captures neutron efficiency, Gd-containing scintillators open promising avenues for neutron detection in contemporary instruments

1.4 Summary of scintillator neutron detectors



Scintillator	sensitive nuclide	Density (g/cm³)	Light yield (ph/MeV)	Neutron yield (ph/n)	Resolution (@662keV)	deliquesce	Large-size production	Price
ZnS:Ag/^{6}LiF	^{6}Li	3.1	-	~150,000	-	No	★★	★★
GS20	^{6}Li	2.6	~4,000	~6,000	<25%	No	★★	★★
CLYC	^{6}Li, ^{35}Cl	4.2	~22,000	~70,000	~5%	Yes	★	★★★
CLLB	^{6}Li,^{35}Cl	4.2	~60,000	~88,000	~4%	Yes	★	★★★
Plastic scintillator	^{1}H、^{6}Li、^{157}Gd、^{10}B	1.0	~7,600	-	-	No	★★★	★
GAGG	^{157}Gd	6.3	~45,000	-	~6%	No	★★	★★
GOS	^{157}Gd	7.4	~28,000	-	~10%	No	★★	★★

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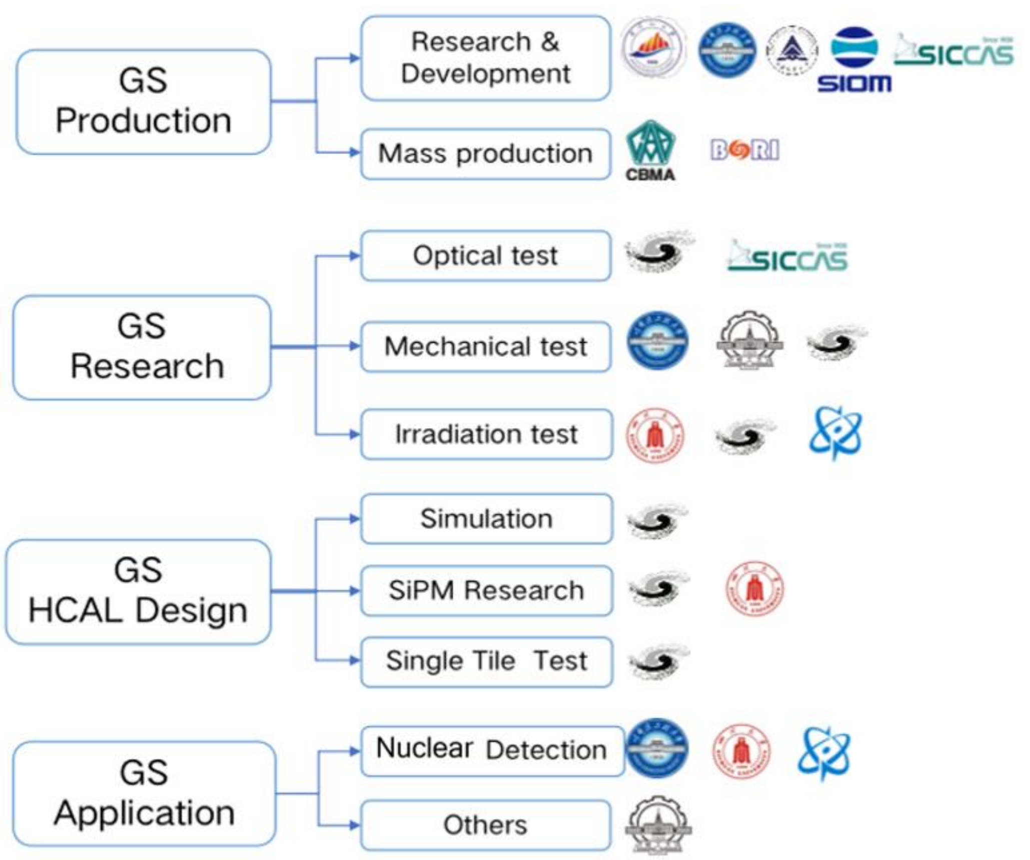
2.1 Large Area Glass Scintillator Collaboration



Institute of High Energy Physics, CAS
中国科学院高能物理研究所

Jinggangshan University
井冈山大学

Beijing Glass Research Institute
北京玻璃研究院

China Building Materials Academy
中国建筑材料研究院

China Jiliang University
中国计量大学

Harbin Engineering University
哈尔滨工程大学

Harbin Institute of Technology
哈尔滨工业大学

Sichuan University
四川大学

Shanghai Institute of Ceramics, CAS
中国科学院上海硅酸盐研究所

Shanghai Institute of Optics and Fine Mechanics,
中国科学院上海光学精密机械研究所

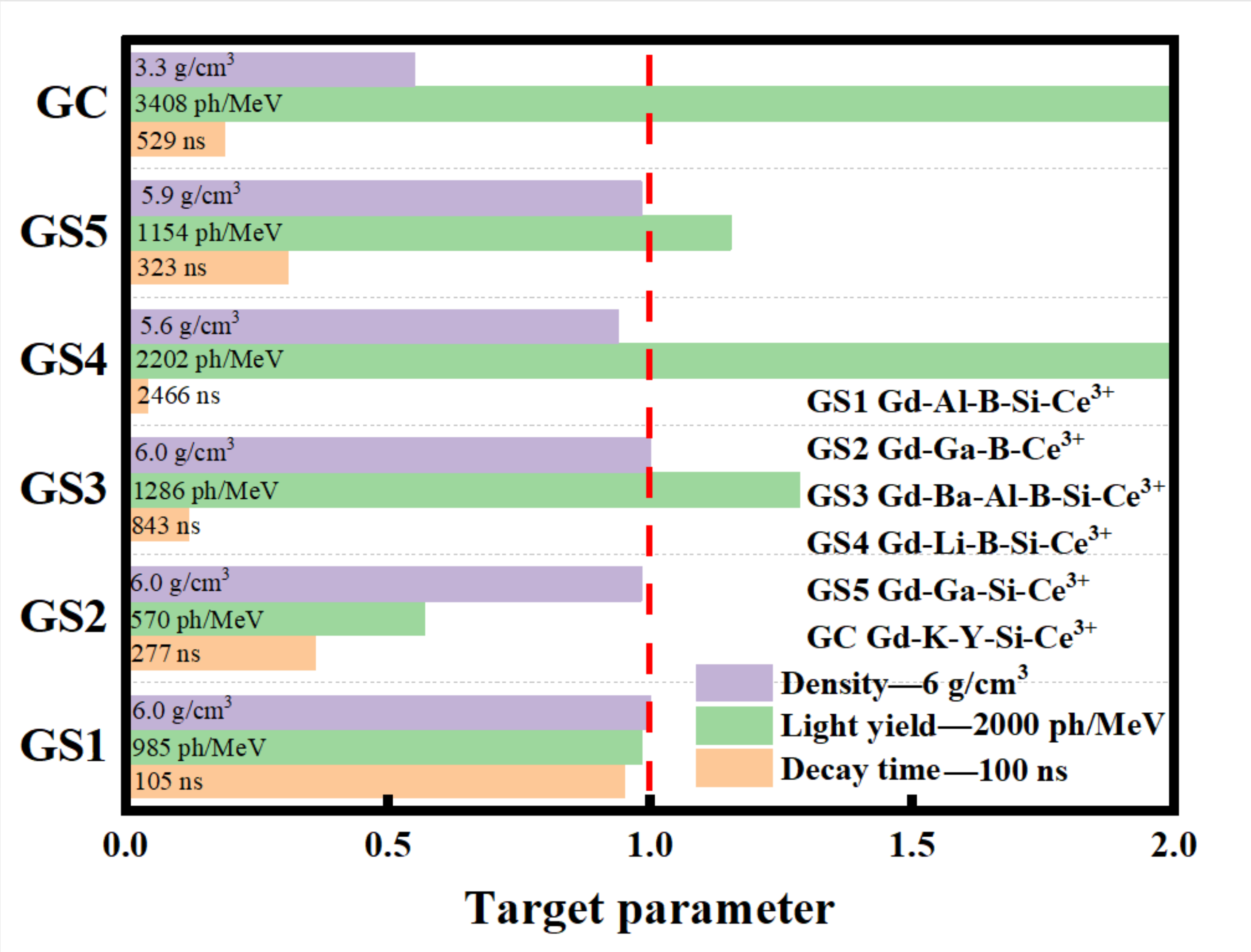
CNNC Beijing Nuclear Instrument Factory
中核（北京）核仪器有限责任公司

闪烁玻璃合作组
Glass Scintillator Collaboration

-- The Glass Scintillator Collaboration Group established in Oct.2021, only 5 groups join together；

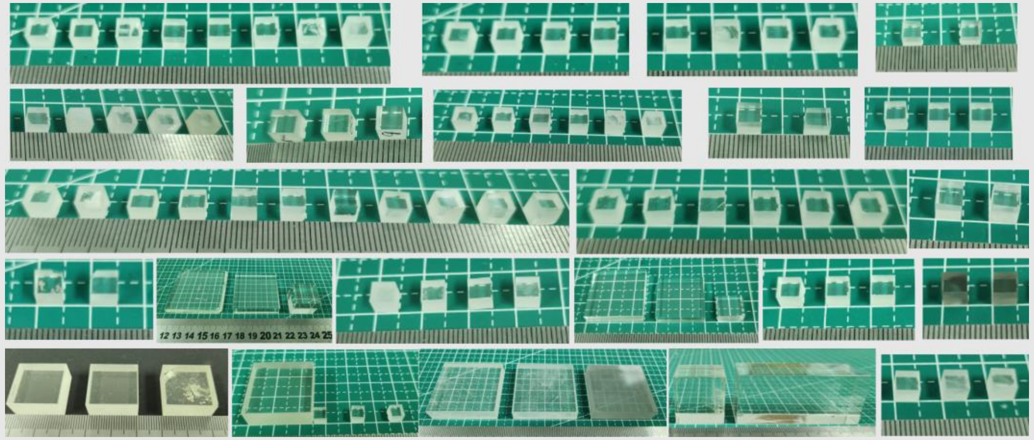
-- There are 3 Institutes of CAS, 5 Universities, 3 Factories join us for the R&D of GS;

2.2 Introduction to Gd-doped GS



Five types for Gd-doped GS

- GS1 Gd-Al-B-Si-Ce^{3+}
- GS2 Gd-Ga-B-Ce^{3+}
- GS3 Gd-Ba-Al-B-Si-Ce^{3+}
- GS4 Gd-Li-B-Si-Ce^{3+}
- GS5 Gd-Ga-Si-Si-Ce^{3+}



- The five GS types contain high concentrations of Gd
- Potential for neutron detection.

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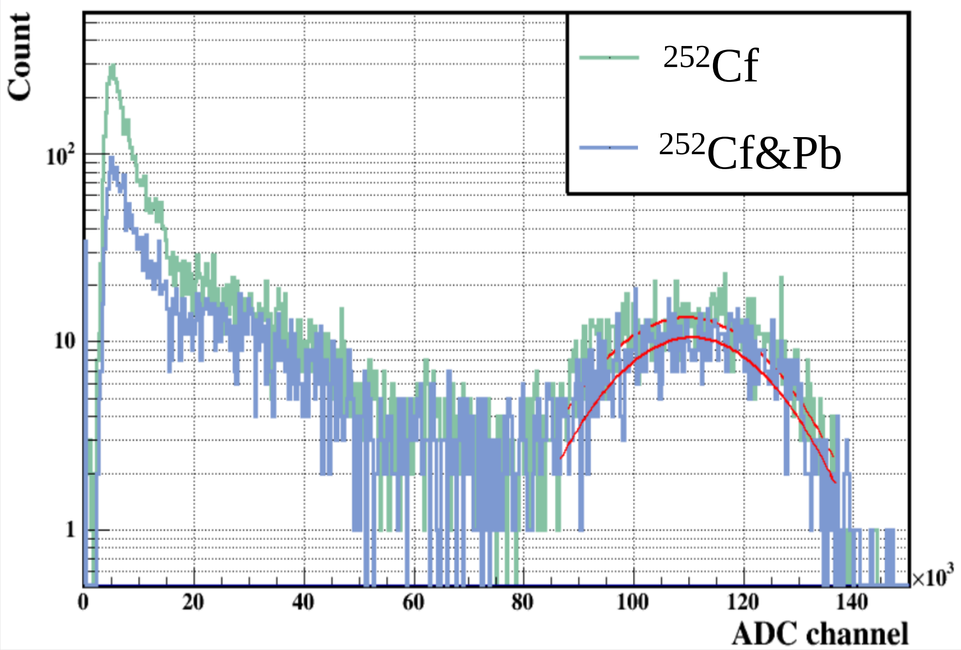
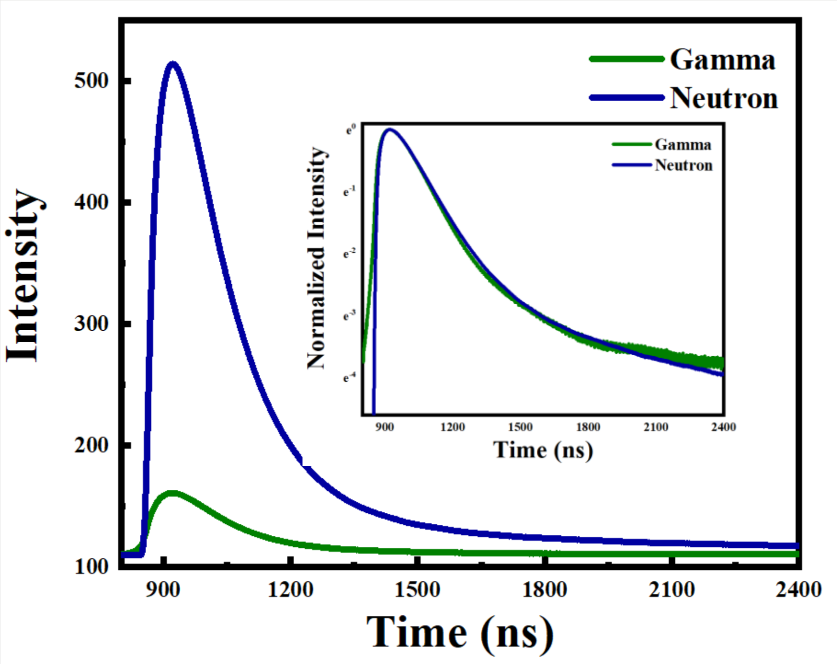
3.1 GS20 neutron detection

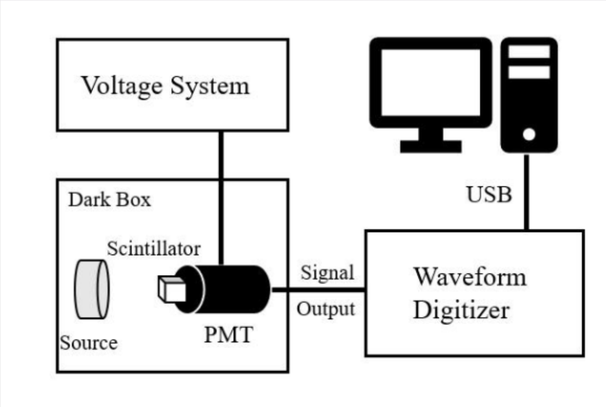


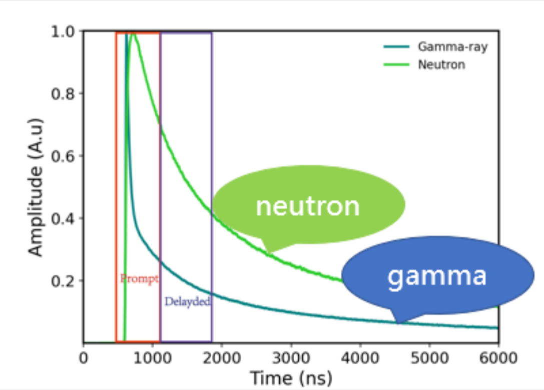


- GS20 is tested at the Cf source with a neutron yield of about 3144 ph/n;
- The decay time for the Gamma signal is 156.8 ns and for the Neutron signal is 165.8 ns;
- The difference between neutron pulse waveform and gamma pulse waveform is small, making it difficult to discriminate between neutrons and gamma rays;

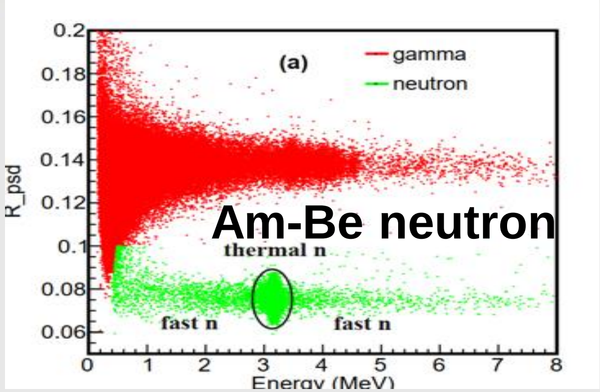
3.2 CLYC scintillator neutron detection

Cs_2LiYCl_6:Ce (CLYC)





n/γ pulses

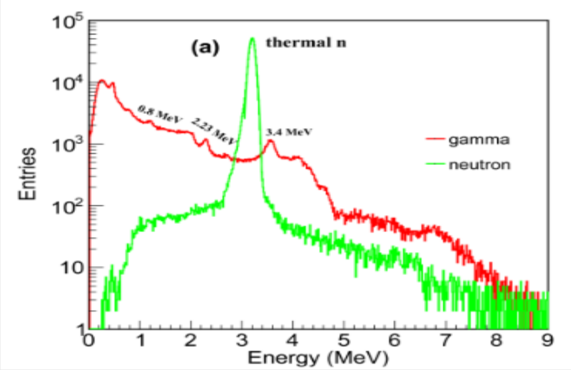


E vs PSD

- **Cs_2LiYCl_6:Ce (CLYC) crystal**
  - complex radiation field, safety
- **By using PSD method, realize**
  - thermal neutron detection, ^{6}Li(n,t)α
  - n/g PSD FOM>2,
  - gamma ER, ~5%@662keV

(ns)	T_Rise	T_fall_1	T_fall_2	T_fall_3
neutron	17.8	-	570	3193
gamma	7.35	10.2	43	918

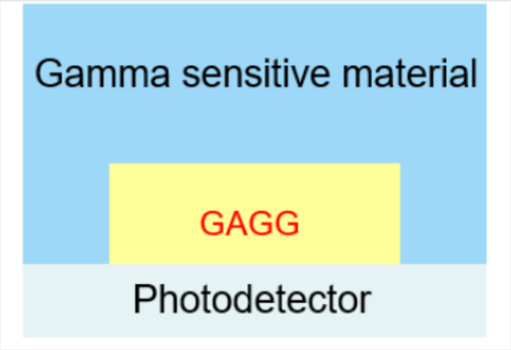
Rise/Fall time



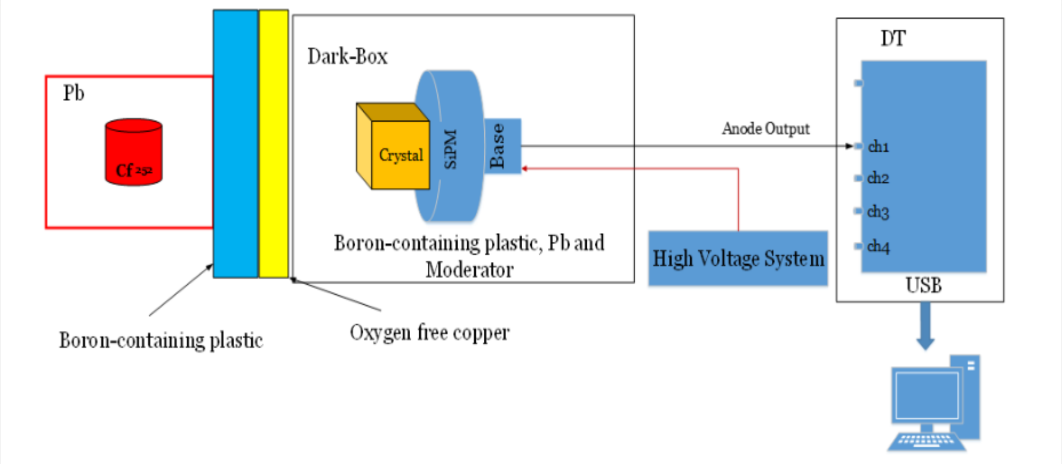
Energy spectrum

IEEE T Nucl Sci, 2023, 70, 2148

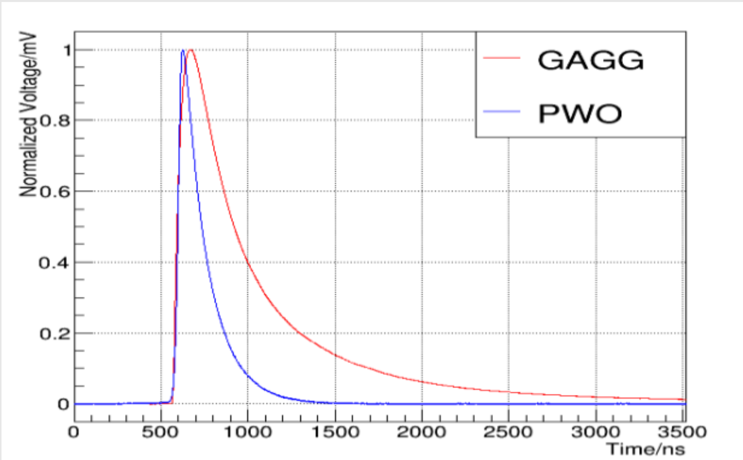
3.3 GAGG scintillator neutron detection



GAGG($Gd_3Al_2Ga_3O_{12}$)

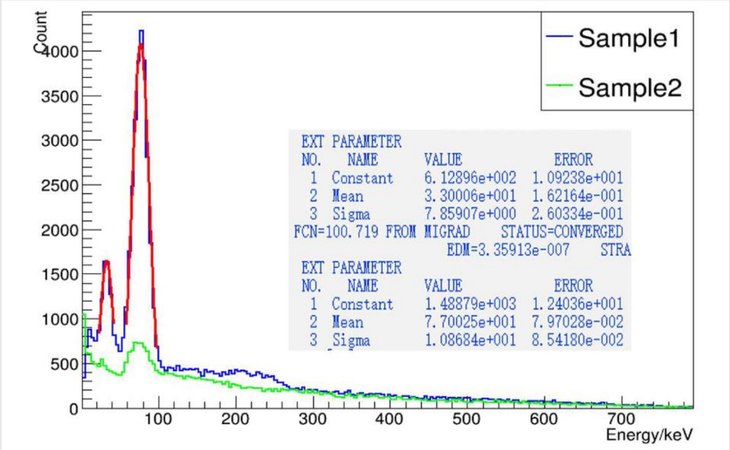


Experimental setup

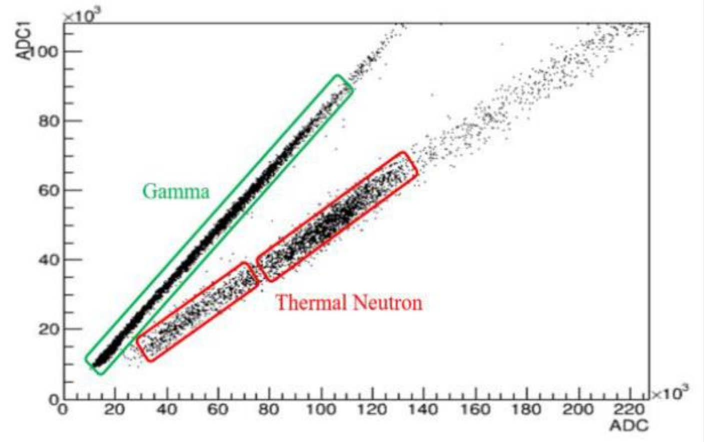


Normalized waveforms of GAGG and PbWO4

- **GAGG crystal coupled $PbWO_4$**
  - Thickness 0.5 mm GAGG: thermal neutron detection, Gd(n,γ)
  - Thickness 30 mm $PbWO_4$: γ rays detection
  - The peak of ~40 keV : the gadolinium K shell X-ray escape and internal conversion electron energy;
  - The peak of ~80 keV : internal conversion electrons(ICEs) and γ rays transition from the first excited states of ^{156}Gd and ^{158}Gd



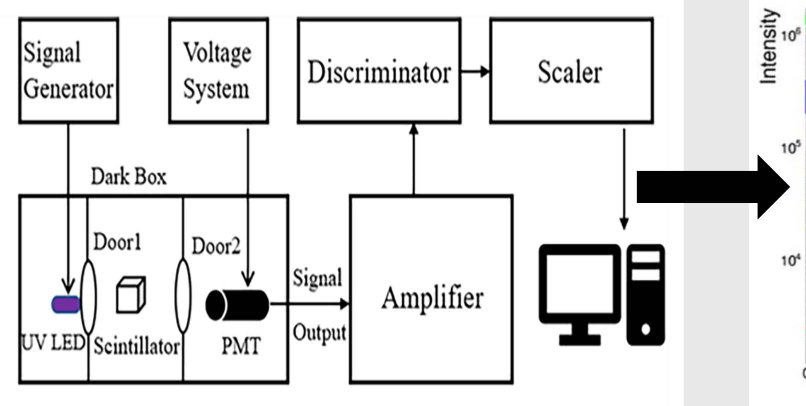
The neutron energy spectrum with GAGG



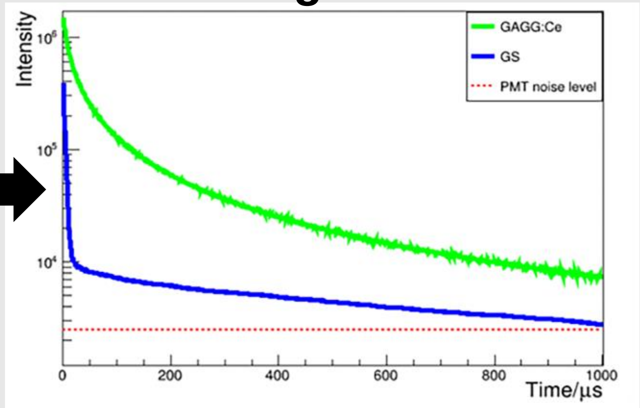
E vs PSD

JINST 2374 (2022) 012133

3.4.1 Optical test (Gd-doped GS)

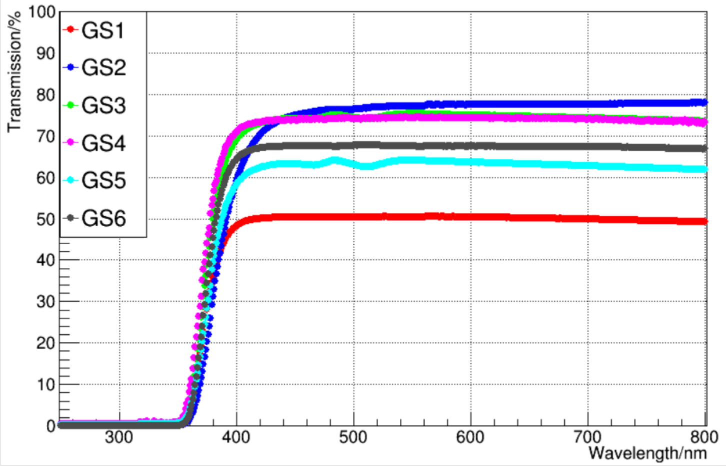


Afterglow

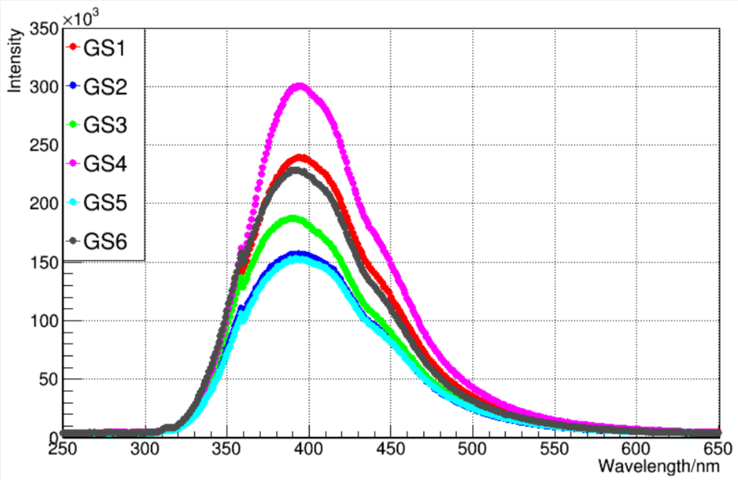


- Fast decay of afterglow
  - the afterglow of GS glass scintillator decreased faster. It indicates that most of the electrons and holes are captured by the traps in the glass, which prevents the photons from escaping from the glass.

Transmittance Curves

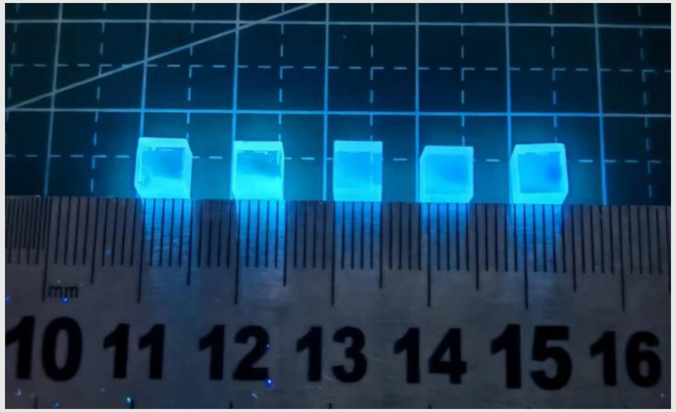


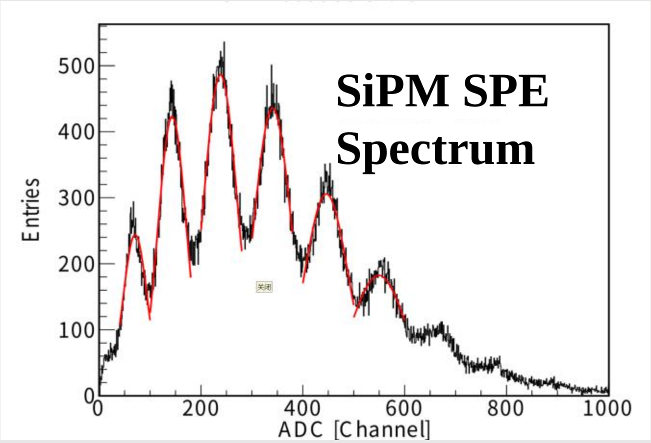
XEL spectra



- high transmittance
  - GS glass from the ultraviolet to near-infrared (250-800nm), transmittance above 500 nm >75% .
- Broad emission spectra
  - GS glass scintillators have broadband emissions in the range of 300-600 nm

3.4.2 Gamma rays test (Gd-doped GS)





The light yield (LY) can be expressed:

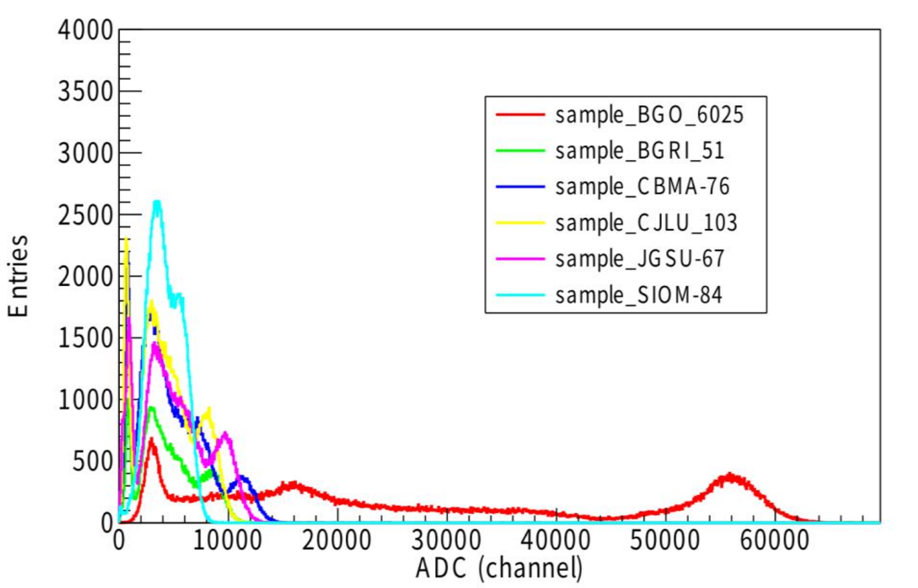
$$LY = \frac{M \times 1000\ keV}{S \times \varepsilon_{PDE} \times E\ keV}$$

M—channel number of full–energy peak in γ–ray energy spectra;

S—single photoelectron channel number of the SiPM;

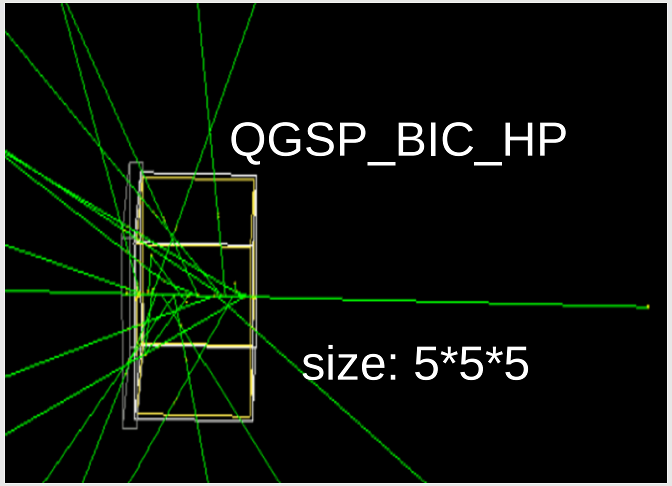
ε_{PDE}—weighted photon detection efficiency (PDE) of the SiPM;

E—energy corresponding to full–energy peak of γ source.



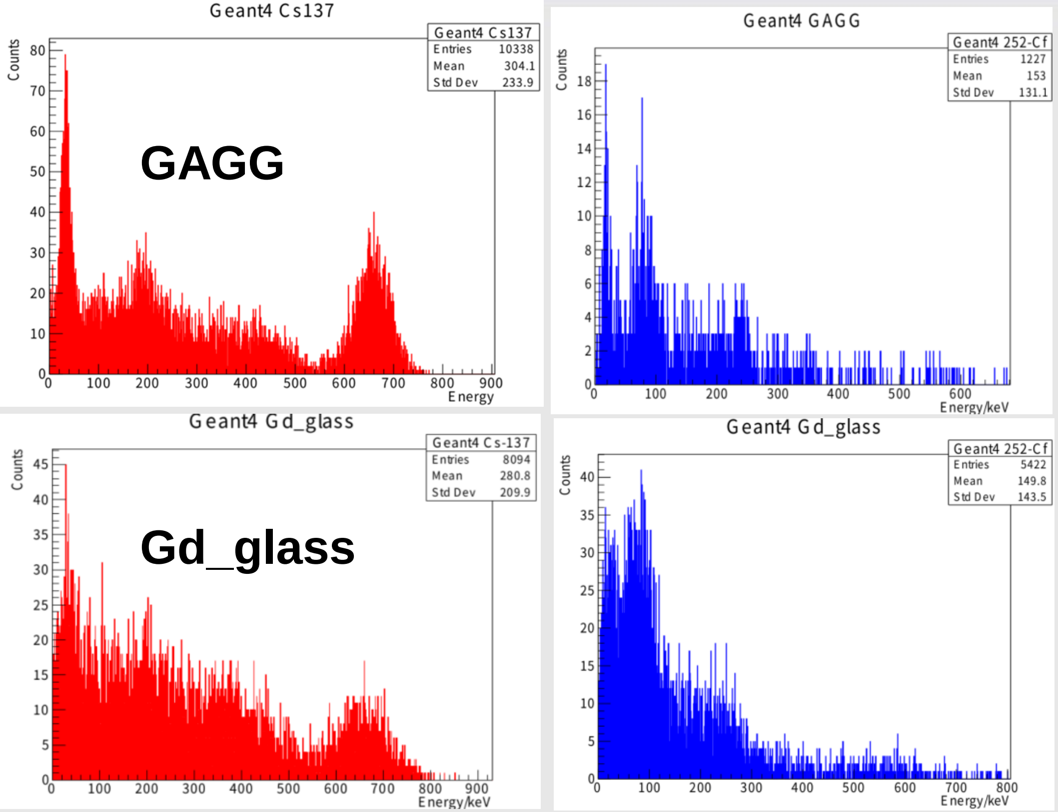
Sample	LY(Ph/MeV)	Energy resolution(662keV)
#1	1154	21.8%
#2	1146	25.4%
#3	1347	27.0%
#4	1128	29.6%
#5	1098	26.8%
#6	1056	28.5%
BGO	7025	14.3%

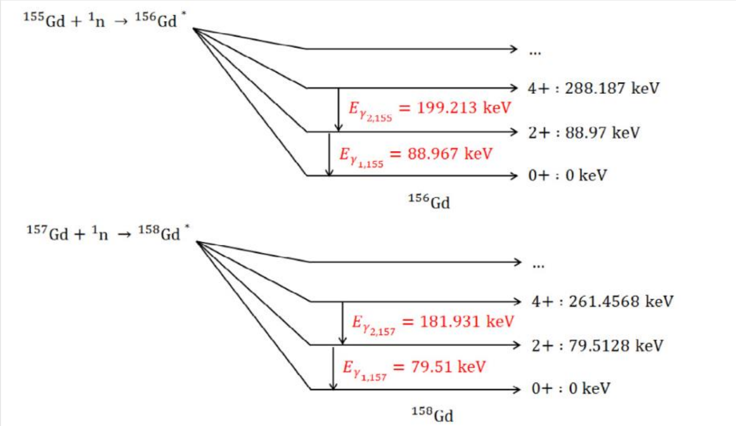
3.4.3 Neutron simulation (Gd-doped GS)



$$n + {}^{155}Gd \rightarrow {}^{156}Gd^{*} \rightarrow {}^{156}Gd + \gamma(8.5MeV) + e^{-}_{IC}(0.039-0.19MeV) + X$$

$$n + {}^{157}Gd \rightarrow {}^{158}Gd^{*} \rightarrow {}^{158}Gd + \gamma(7.9MeV) + e^{-}_{IC}(0.029-0.20MeV) + X$$



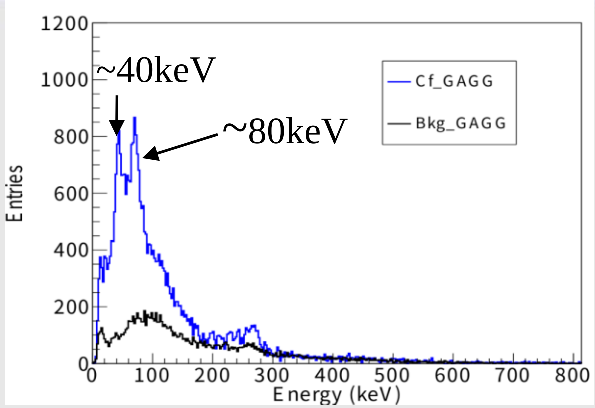


[Data from NIMA 882(2018) 53-668]

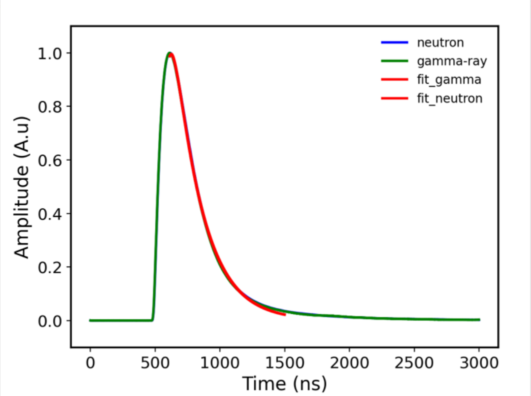
- First excited states of Gd-156 and Gd-158 nuclei and associated gamma transitions

- The neutron response of the Gd-doped glass scintillator and GAGG sample was simulated by using ^{252}Cf neutron source；
- The peak of ~40 keV : the gadolinium K shell X-ray escape and internal conversion electron energy;
- The peak of ~80 keV : the internal conversion electrons(ICEs) and γ rays transition from the first excited states of ^{156}Gd and ^{158}Gd

3.4.4 Neutron test of Gd-doped GS and GAGG

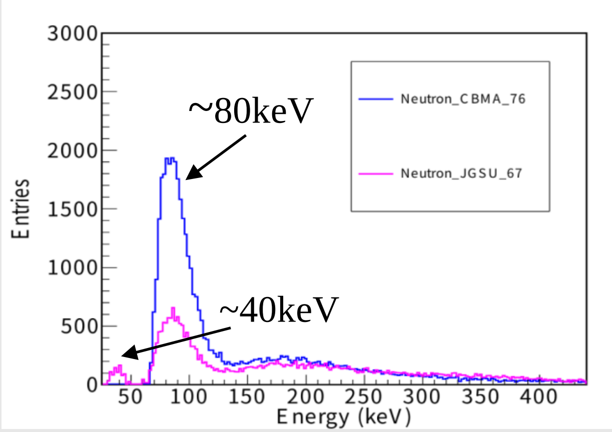


GAGG neutron energy spectrum

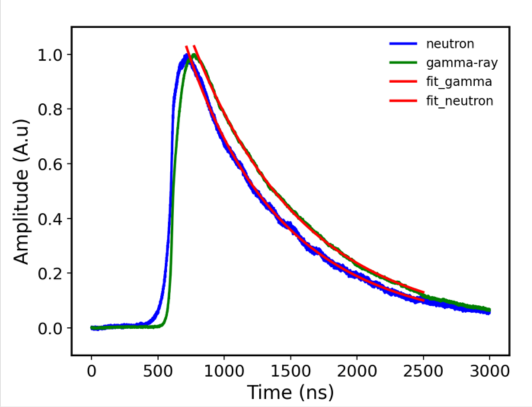


n/γ pulses

- The decay times for neutrons and gamma rays are similar at 226ns and 229ns respectively.



GS neutron energy spectrum



n/γ pulses

- Gamma: The decay time of fast and slow component are 49.2 ns (97.1%) and 861.9 ns (2.9%).
- neutron: The decay time of fast and slow component are 154.2 ns (36.9%) and 767.4 ns (63.1%).

- Neutron signal can be detected；

Prepare for publication

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Summary

- Gadolinium is a stable, naturally occurring element with the highest interaction probability with neutrons at thermal neutron, making it a material with high potential for neutron detection.

- Gd-doped GS exhibits a certain level of neutron response from low-energy (40–200 keV)

- Gd-doped GS exhibits certain waveform differences in neutron and gamma signals, raising hopes for neutron-gamma discrimination based on these waveform differences

- As a new material for neutron detection, Gd-doped GS has the advantages of high plasticity and low cost, and has the potential for large-area neutron detection.

See the unseen
change the unchanged

The Innovation

THANKS