Improving the Neutron and Gamma-Ray Response of $Cs_2LiLaBr_6:Ce^{3+}$

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$Cs_2LiLaBr_6:Ce^{3+}$ (CLLB:Ce^{3+})

Cerium doped $Cs_2LiLaBr_6:Ce^{3+}$ (CLLB:Ce^{3+}) elpasolite crystal has attracted interest as an excellent detector of gamma rays [1,2]. Almost as interesting is its ability to discriminate neutron detections from gamma-rays via pulse shape. Although pulse-shape discrimination (PSD) in CLLB is not as distinct as in fellow elpasolite, Cs_2LiYCl_6 (CLYC), CLLB is superior in light yield and energy resolution. Thus, interest exists in optimizing the gamma ray response and the PSD through changes in dopant concentration and improved PSD techniques.

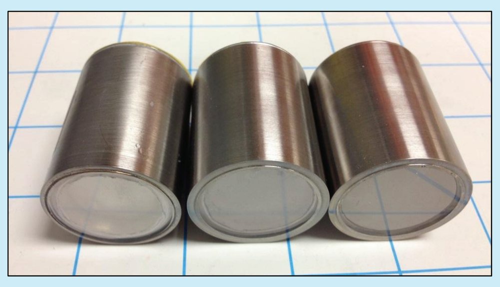

Fig. 1 Photo of packaged CLLB crystals (left to right: 2%, 0.5% and 3.5% Ce)

Table 1. Summary of CLLB samples

	Size
0.5% Ce	Ø1" x 1/4"
2% Ce	Ø1" x 1/2"
3.5% Ce	Ø1" x 1"

[1] J. Glodo, et al., IEEE TNS, 58:333-338, 2011.
[2] U. Shirwadkar, et al., NIMA, 652:268-270, 2011.

Emission Characteristics

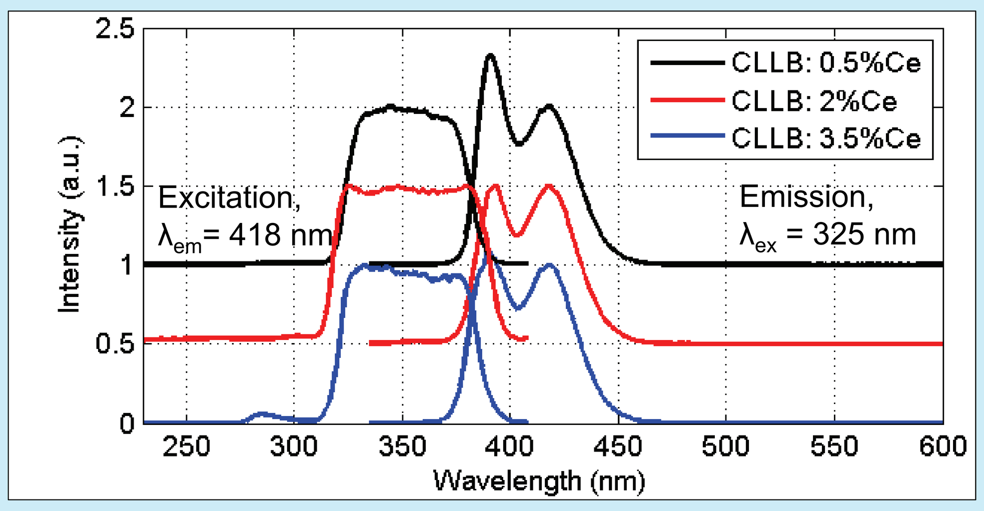

Fig. 2 Emission and excitation spectra of CLLB samples

Characteristic Ce 4f-5d transitions are observed. The overlap between excitation and emission spectra increases with increasing Ce concentration. The intensity of Ce emission peak at 391 nm also decreases. This indicates some level of self-absorption. The CLLB: 3.5% Ce shows an additional excitation peak at 285 nm. This most likely relates to additional defects introduced by high Ce concentration.

Scintillation Properties

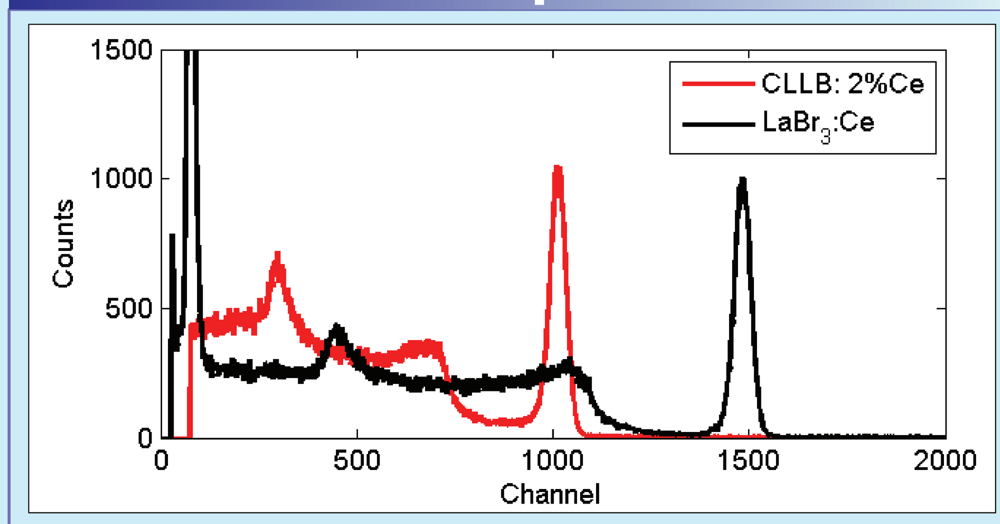

Fig. 3 Pulse height spectra of CLLB:2%Ce and $LaBr_3$:Ce

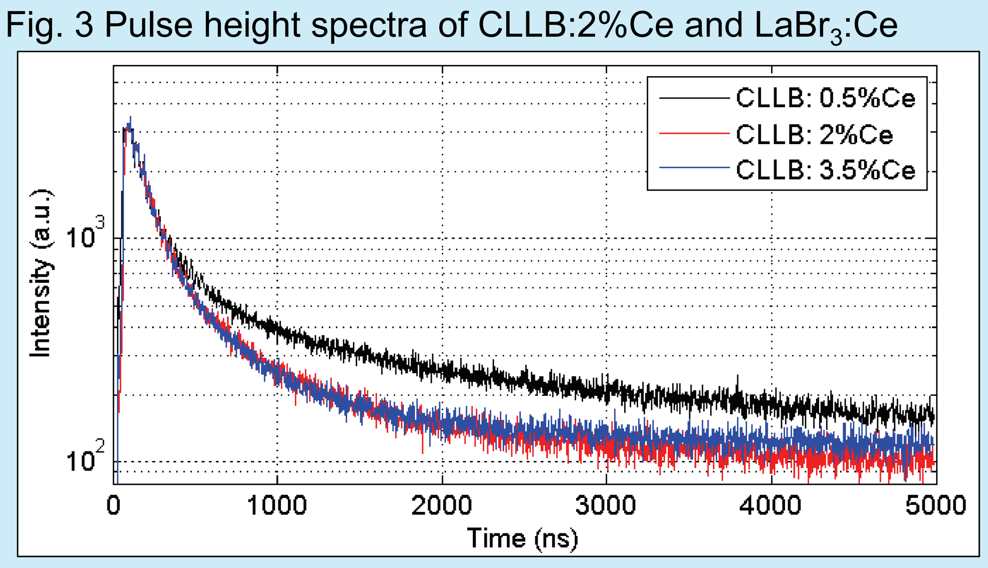

Fig. 4 Scintillation time profiles of CLLB crystals

Table 2. Summary of scintillation properties of CLLB

	Light Output (% of $LaBr_3$:Ce)	ΔE/E	Decay Constant	
			τ1 (ns)	τ2 (ns)
0.5% Ce	56%	7.3%	116 (53%)	975 (47%)
2% Ce	68%	4.6%	122 (61%)	661 (39%)
3.5% Ce	39%	8.6%	125 (64%)	590 (36%)

The CLLB:2%Ce shows the highest light output and the best energy resolution among three crystals. The CLLB:3.5%Ce crystal has cloudy area near its core, which reduces its light output and worsens its energy resolution. We are in the process of improving crystal quality of CLLB (**See NPO1-197**).

Delayed coincidence method, originated by Bollinger and Thomas, was used to determine the scintillation time profiles [3]. All time profiles can be fitted with a double-exponential decay model.

[3] L. M. Bollinger, G. E. Thomas, Rev. Sci. Inst. vol.32, no.9, pp.1044-1050, 1961

Temperature Response - LO

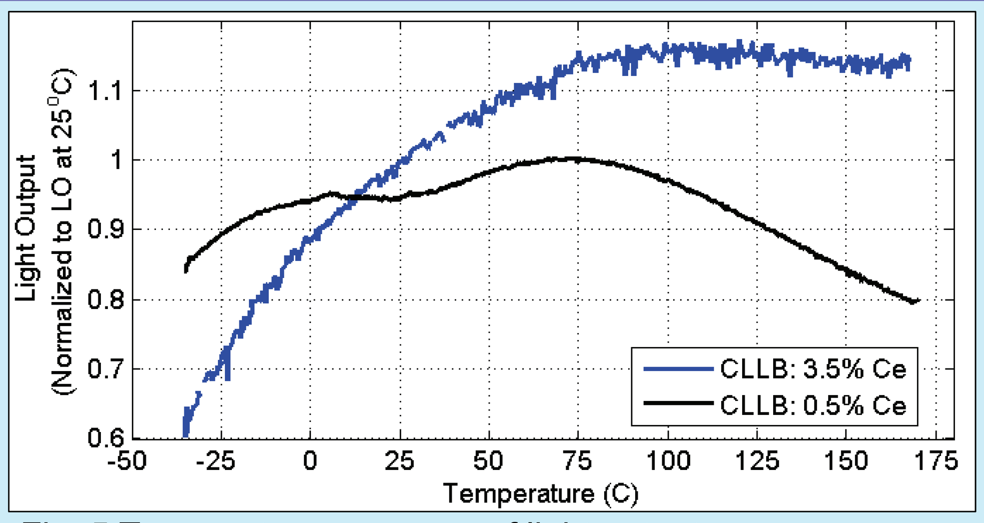

Fig. 5 Temperature response of light output

The light output temperature response is different between 0.5% Ce and 3.5% Ce doped CLLB.

CLLB:3.5%Ce gains ~15% of light when heated from 25 °C to ~170 °C.

CLLB:3.5%Ce shows nearly constant light output between 75 and 171°C. CLLB:0.5%Ce loses about 20% of its light when heated to 171 °C. CLLB's good performance at high temperature makes it interesting for high temperature gamma and neutron detection in applications including oil well logging.

Proportionality

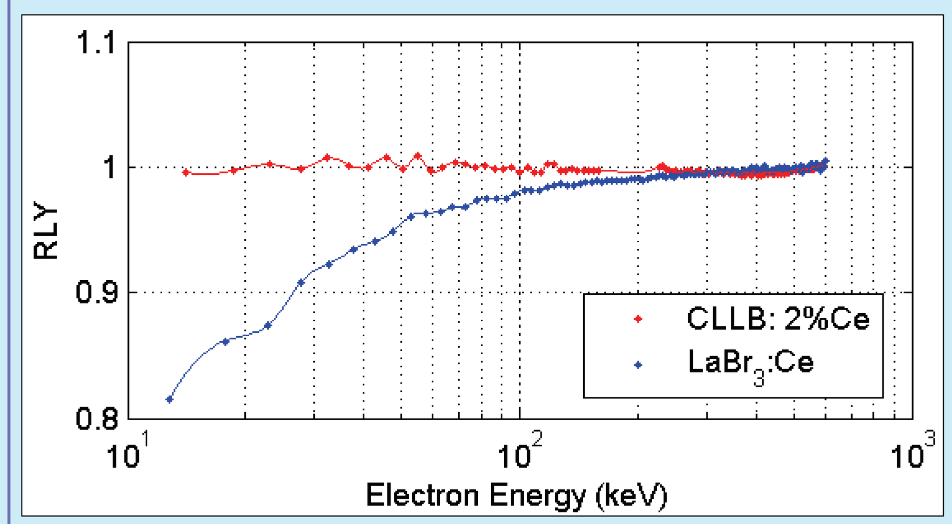

Fig. 6 Electron response of CLLB:2%Ce and $LaBr_3$:Ce

Electron response were measured by Close-Coupled Compton Coincidence method using a Zn-65 source [4]. CLLB shows one of the best proportionality responses among all known scintillators.

[4] P.B. Ugorowski et al., NIMA 615, 2, pp.182-187, 2010.

n-γ Pulse Shape Discrimination

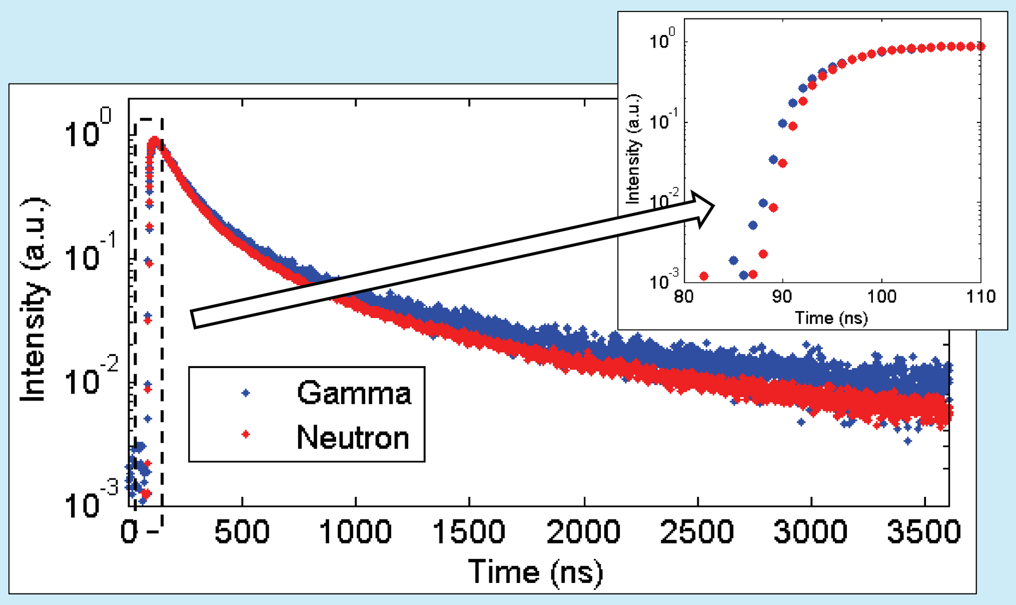

Fig. 7 Gamma and neutron excited pulses in CLLB

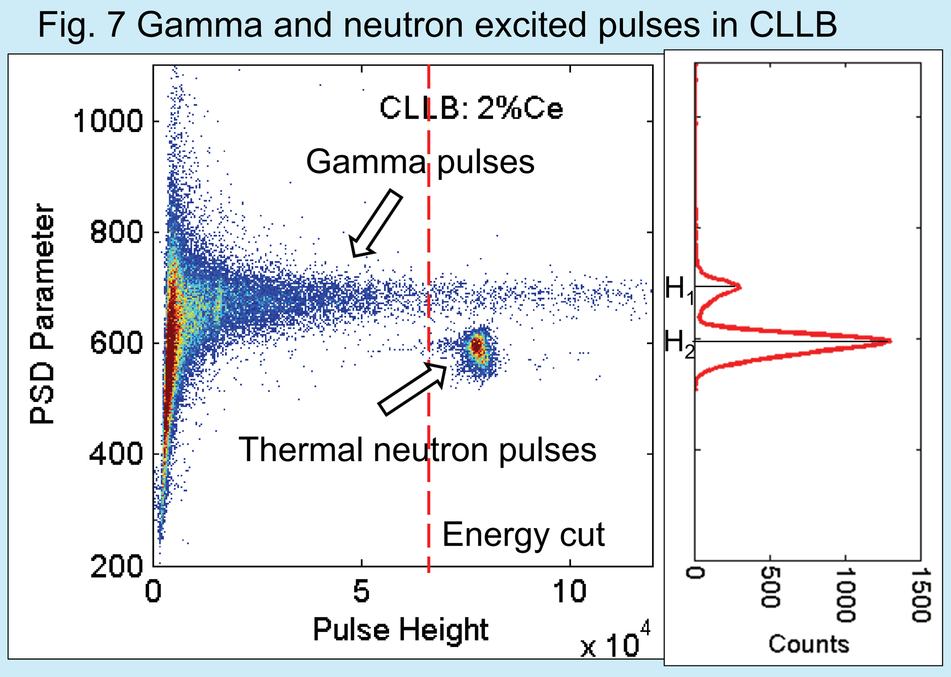

Fig. 8 PSD map of CLLB:2% Ce excited with Cf-252

Fig. 7 shows the averaged pulses for both gamma and neutron excited pulses in CLLB. Pulses are recorded by a CAEN DT5751 digitizer at 1GHz sampling rate. The coupled PMT is Photonis XP20Y0.

Unlike CLYC, **neutron excited pulses from CLLB are faster than gamma excited pulses in both rise and decay**. The relative difference in the rise part of the pulse is even more prominent than the difference in the decay part in terms of integrated intensity. In order to optimize the performance of PSD, **the sorting algorithm should allocate more weight to the rise part.** Table 2 compared the Figure of Merit (FOM) derived from several example PSD algorithms. Same data set recorded from CLLB:2%Ce is used. The 2-60 rise time algorithm is found to be the best*.

Table 3. Comparison of some example PSD algorithms for CLLB: 2%Ce

PSD Algorithm	FOM**
Charge Comparison	1.22
Fourier Transform	1.26
10-90% rise time	1.00
2-60% rise time	1.34

* Patent pending

$$** \; FOM = \frac{H_1 - H_2}{FWHM_1 + FWHM_2}$$

Table 4. PSD performance of different CLLB samples

Sample	GEE (MeV)	FOM
CLLB: 0.5%Ce	3.27	1.02
CLLB: 2%Ce	3.27	1.34
CLLB: 3.5%Ce	3.3 ± 0.3	/

Table 4. compares the PSD FOM of different CLLB sample using the 2-60% rise time algorithm. CLLB:2%Ce shows the best performance. The cloudy part in the 3.5% Ce sample appears to significantly degrade its PSD capability. PSD is possible but the PSD peak for neutrons cannot be clearly resolved for this particular sample. Better performance is expected with crystal quality improvement. The GEE for CLLB crystals are measured to be 3.27 MeV.

Temperature Response – PSD

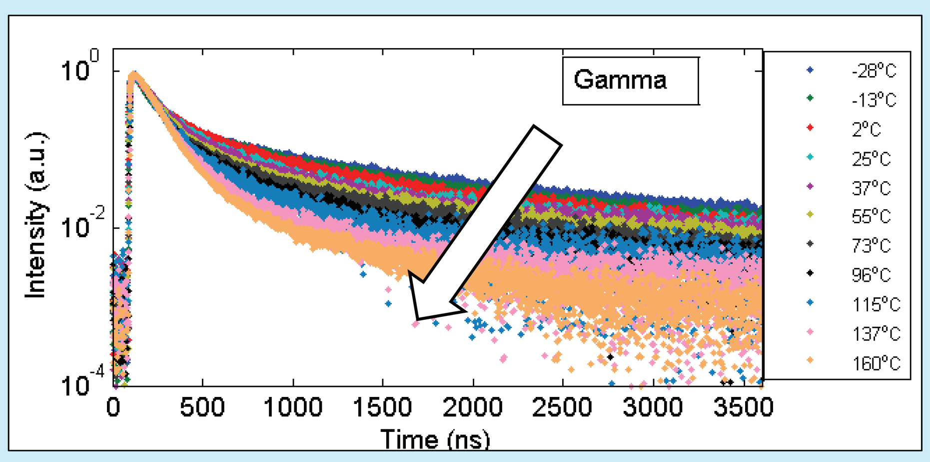

Fig. 9 Gamma pulse shapes at different temperatures

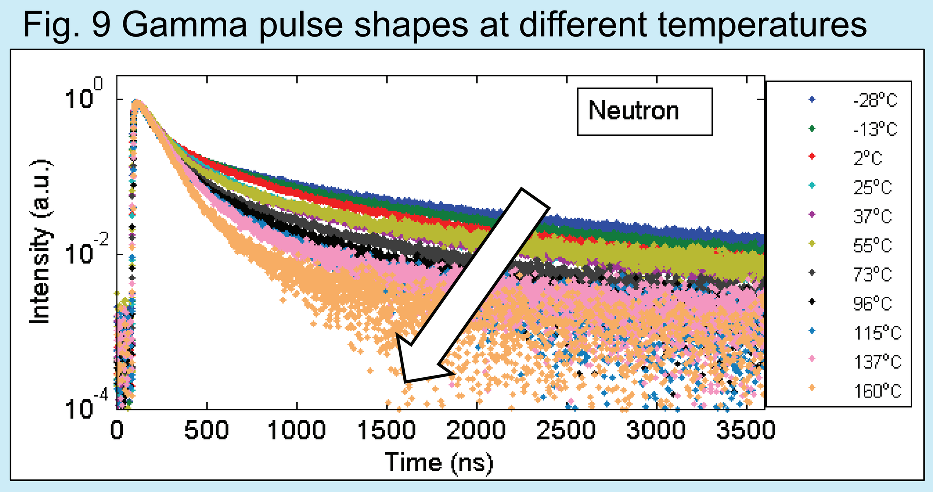

Fig. 10 Neutron pulse shapes at different temperatures

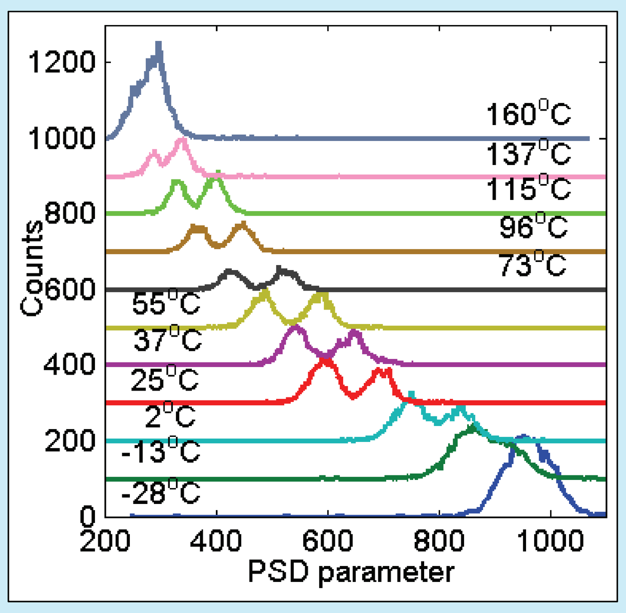

Fig. 12 PSD spectra vs. temperature

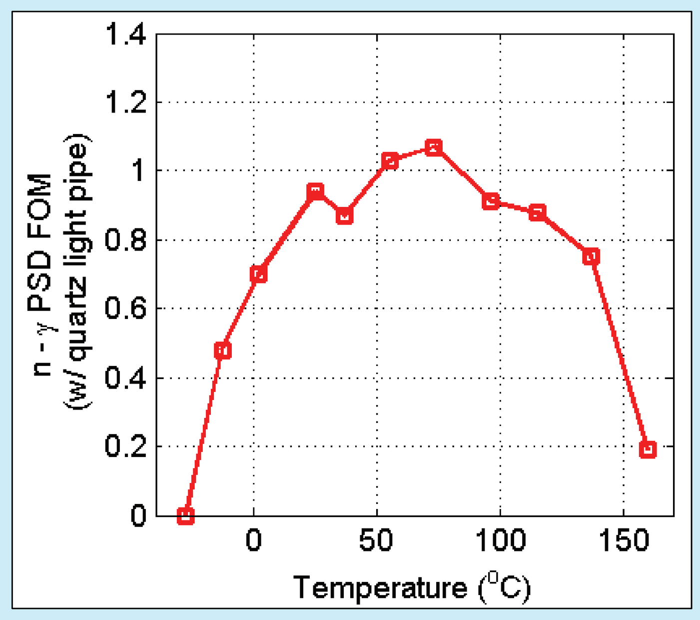

Fig. 13 n-γ PSD FOM vs. temperature

CLLB:2%Ce was used to study the crystal's PSD performance at different temperatures. Scintillation pulses were record with a Photonis XP20Y0 PMT through a quartz light pipe.

Both gamma and neutron pulses become significantly faster with increasing temperature. The pulse shape differences between gamma and neutron diminish when the crystal is cooled to -28°C. However, **PSD is preserved in a wide temperature range from room temperature up to at least 140°C**, which is even higher than that of CLYC [5]. This enables the possibility for down-hole gamma and neutron measurements in oil well logging applications.

[5] P.R. Menge, et al., *NSS/MIC, 2011*, pp.1598,1601, 23-29 Oct. 2011

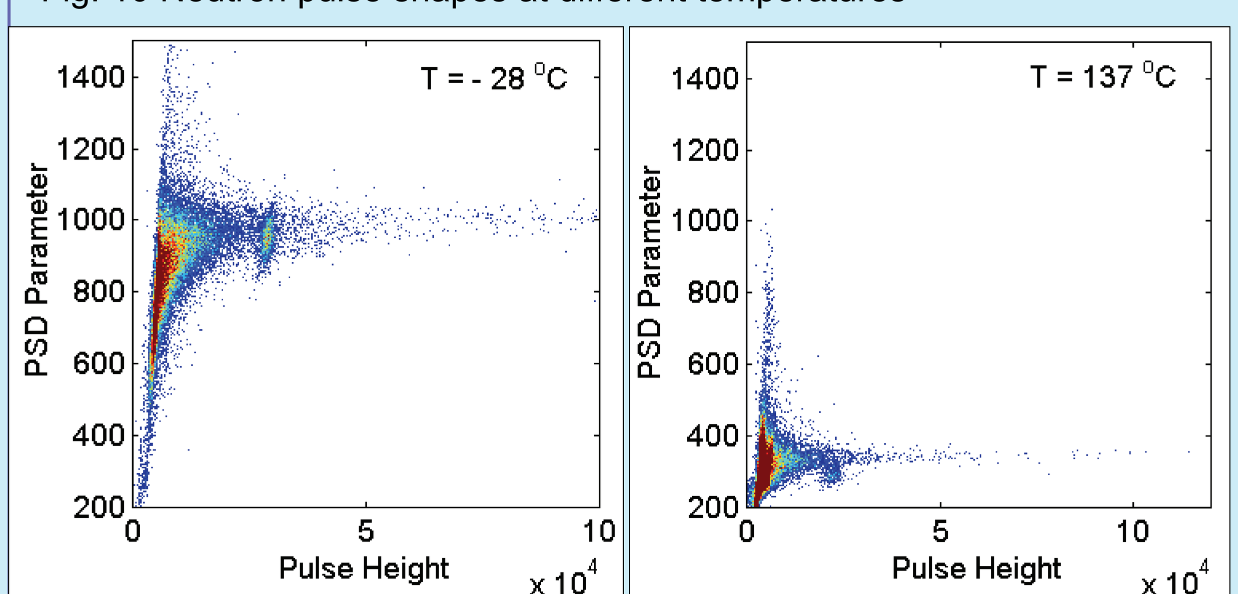

Fig. 11 Example PSD maps at T = -28°C and 137 °C

Summary

- Gamma and neutron responses of $Cs_2LiLaBr_6$ elpasolite crystals with 0.5, 2, and 3.5% Ce doping are presented.
- Pulse shape discrimination (PSD) between gamma and neutron is possible with CLLB;Ce crystal
- Neutron excited pulses are faster than gamma excited pulses in both rise and decay.
- n-γ PSD in CLLB is preserved in a wide temperature range from RT up to at least 140 °C

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