Performance Engineering of NaI:Tl$^+$ with Co-doping

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SAINT-GOBAIN

Improving Gamma Performance with Ca^{2+} / Sr^{2+} co-doping

Discovered nearly 70 years ago, NaI:Tl$^+$ is still the most used crystal scintillator by volume. NaI:Tl$^+$ possesses many favorable properties including high light yield, moderately fast decay time, good mechanical strength, good temperature stability and most importantly, low cost. However, NaI:Tl's poor intrinsic energy resolution significantly limits its usefulness in gamma spectroscopy applications. Its moderate decay time also makes it difficult to use in high count rate applications such as Prompt Gamma Neutron Activation Analysis (PGNAA). Saint-Gobain Crystals has been actively working on scintillator performance engineering for the past several years. In this research, gamma performance of NaI:Tl$^+$ crystal is significantly improved by co-doping with Ca^{2+} and Sr^{2+} [1].

Energy Resolution and Decay Time Improvement

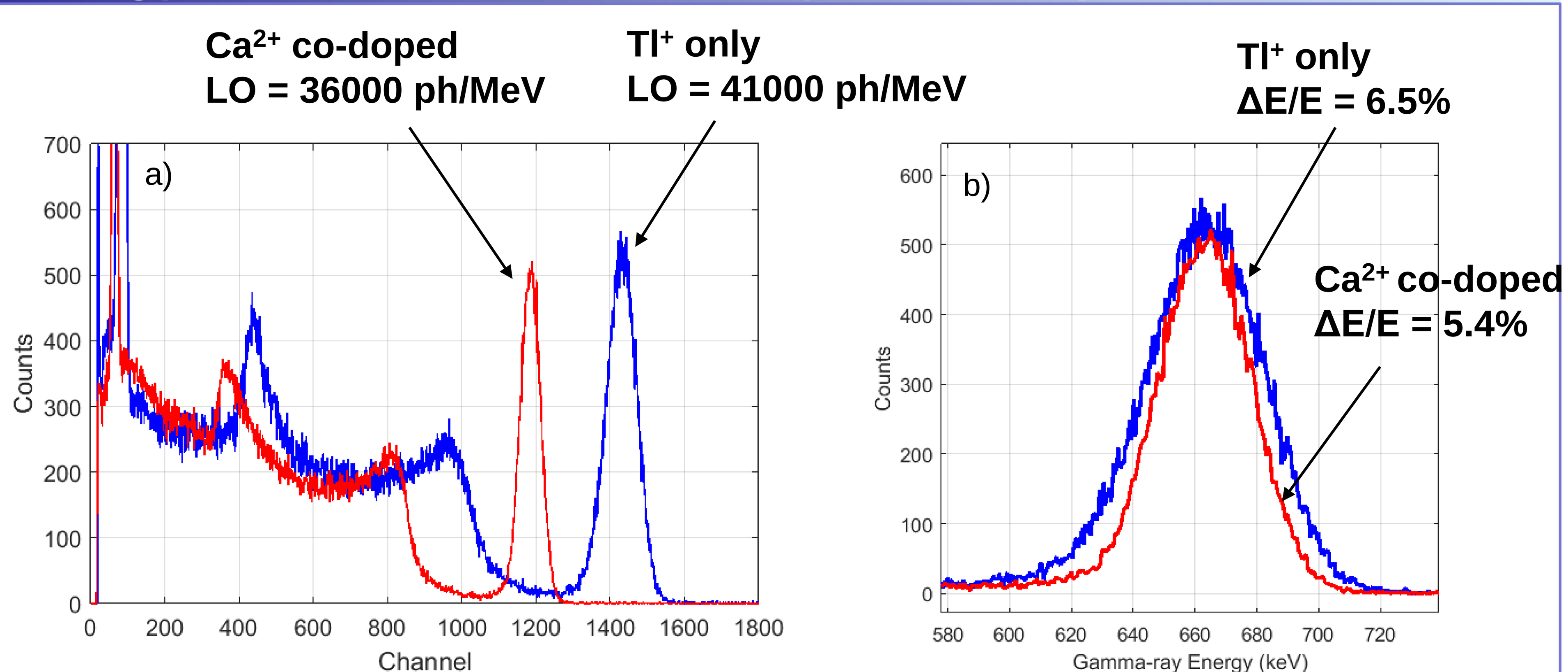


Fig. 1 a) Pulse height spectra of Tl$^+$ only and 0.3% Ca^{2+} co-doped NaI excited with ^{137}Cs source; b) Enlarged view of overlayed 662 keV photo peaks

- ❖ **Energy resolution improved to 5.4%**
- ❖ **80% light yield of standard NaI:Tl$^+$**

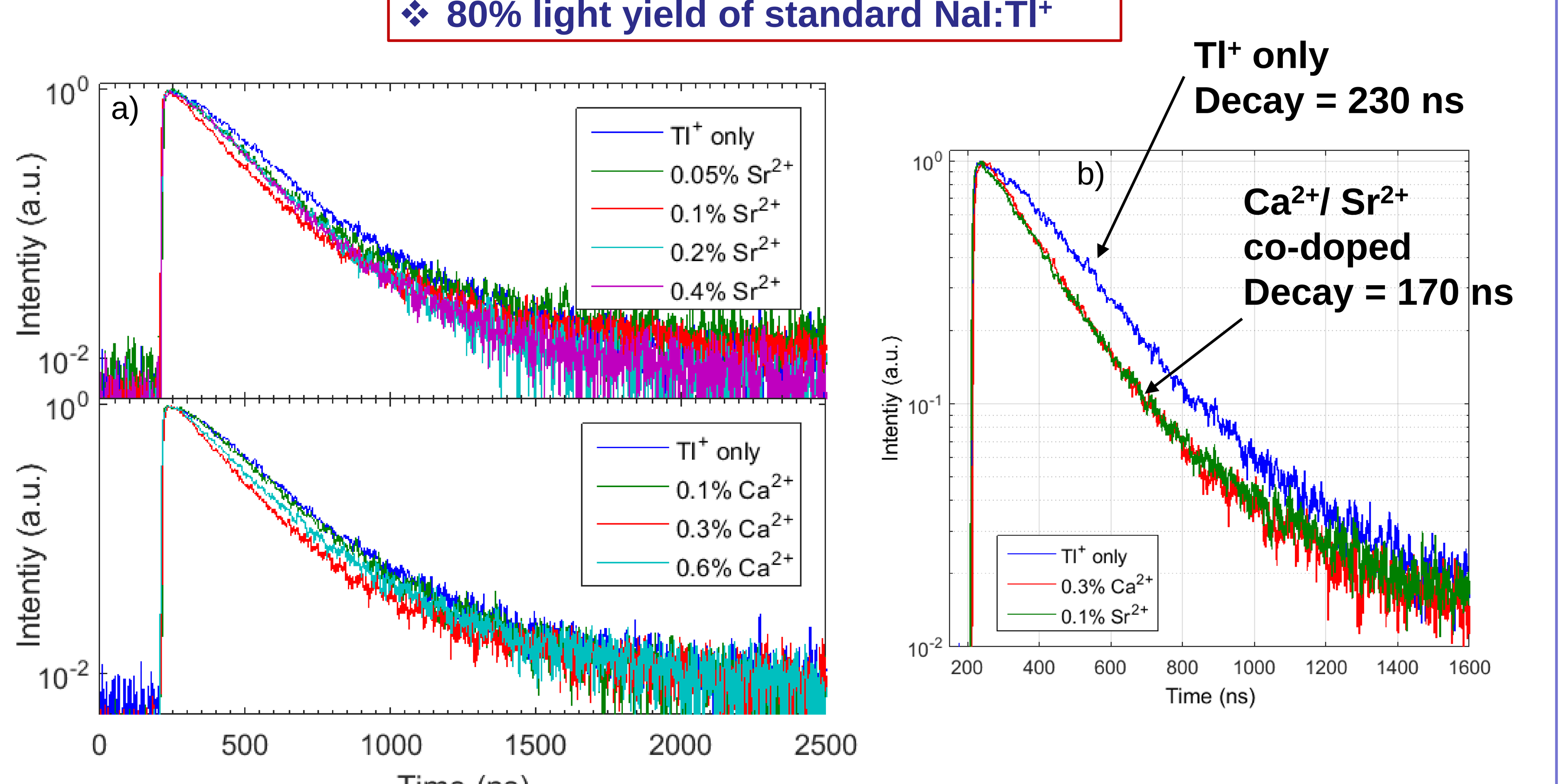


Fig. 2 a) Averaged PMT (XP20Y0) traces of Tl$^+$ only, Sr^{2+}, and Ca^{2+} co-doped NaI:Tl$^+$ b) Enlarged view of Tl$^+$ only, 0.1% Sr^{2+} and 0.3% Ca^{2+} averaged PMT traces

- ❖ **25% faster than standard NaI:Tl$^+$**
- ❖ **Slow rise component ("flat top") eliminated by Ca^{2+}/Sr^{2+} co-doping**

Table I Scintillation properties of Ca^{2+}/ Sr^{2+} co-doped NaI:Tl$^+$

Co-doping*	Light Yield (N_{ph}/MeV)	ΔE/E @ 662 keV (FWHM)	$\delta_{intrinsic}$ (FWHM)	Decay Constant (fast, ns)	Decay Constant (slow, ns)
Tl$^+$ only	41000 ± 2000	6.4%	5.7%	220 ± 10 (96%)	1500 ± 200 (4%)
0.05% Sr^{2+}	33000 ± 2000	6.8%	6.0%	201 ± 21 (94%)	860 ± 240 (6%)
0.1% Sr^{2+}	**26000 ± 3100**	**5.3%**	**3.7%**	**172 ± 10 (92%)**	**860 ± 160 (8%)**
0.2% Sr^{2+}	30000 ± 2600	6.0%	4.9%	195 ± 16 (96%)	690 ± 90 (4%)
0.4% Sr^{2+}	32000 ± 4000	6.8%	6.0%	195 ± 7 (96%)	1000 ± 300 (4%)
0.1% Ca^{2+}	32000 ± 3000	5.9%	5.0%	199 ± 10 (95%)	1030 ± 150 (5%)
0.3% Ca^{2+}	**34000 ± 1800**	**5.4%**	**4.4%**	**173 ± 12 (94%)**	**830 ± 230 (6%)**
0.6% Ca^{2+}	36000 ± 2700	5.6%	4.7%	186 ± 11 (94%)	870 ± 110 (6%)

*All Tl$^+$ doping is 0.1 at%, with respect to Na$^+$

- ❖ **Ca^{2+}/ Sr^{2+} co-doping significantly improves the light yield non-proportionality of NaI:Tl$^+$ thus improves its intrinsic energy resolution.**

Non-Proportionality Improvement

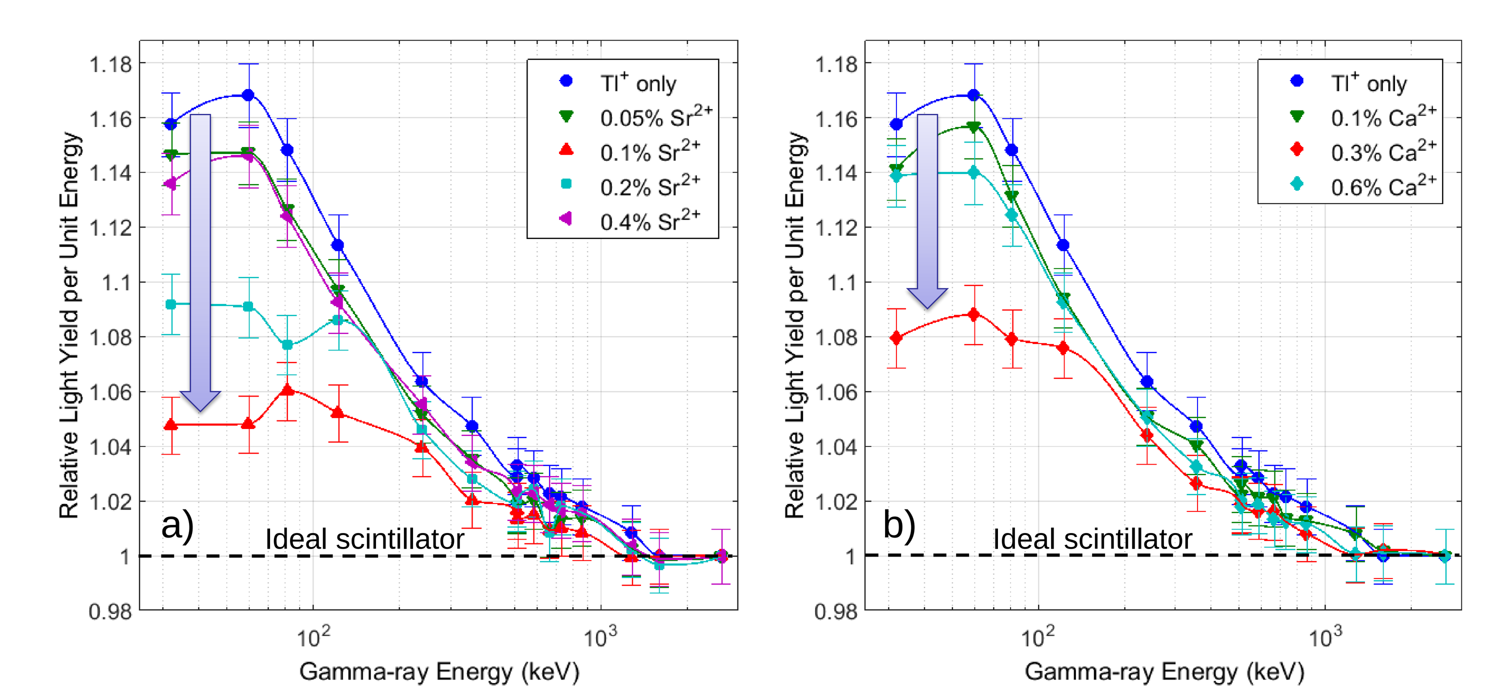


Fig. 3 Gamma-excited non-proportionality curves for a) Sr^{2+} and b) Ca^{2+} co-doped NaI:Tl$^+$

- ❖ **Significantly improved light yield non-proportionality**
- ❖ **Reduced "halide hump" at lower energies [2]**

Emission Characteristics

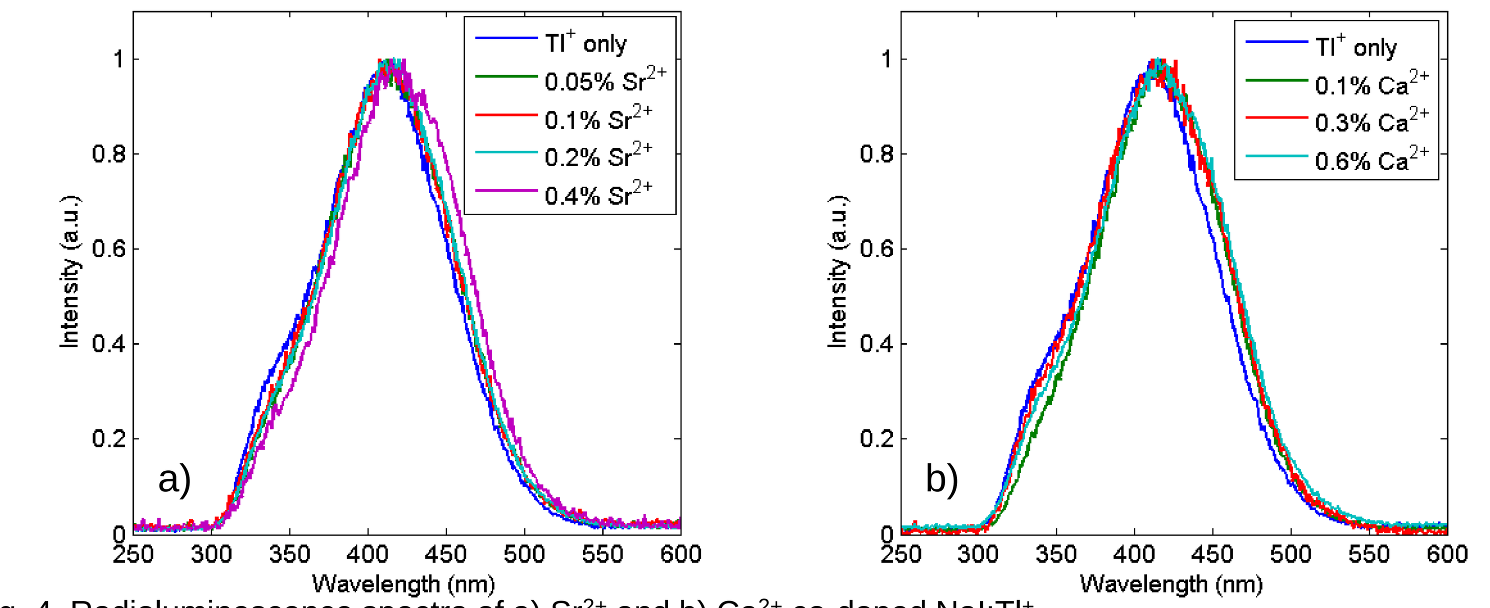
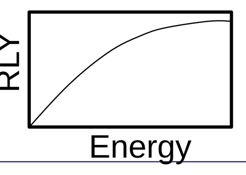
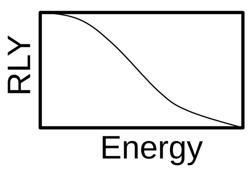
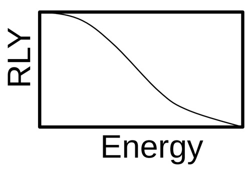


Fig. 4 Radioluminescence spectra of a) Sr^{2+} and b) Ca^{2+} co-doped NaI:Tl$^+$

- ❖ **No significant change other than a slight redshift (1-2 nm)**

Proposed Mechanism

Table II Scintillation processes in NaI:Tl$^+$

	Scintillation Processes	Lifetime Governed by	Influence on Non-Proportionality	Effect of Ca^{2+}/Sr^{2+} Co-doping
(i)	Prompt: Tl$^+$ + STE → (Tl$^+$)* or Tl$^+$ + h + e → (Tl$^+$)*	Lifetime of (Tl$^+$)* (150 ns) [3,4]	 large	Not affected
(ii)	Intermediate: 1. e$^-$ + Tl$^+$ → Tl^0 2. Tl^0 + STH → (Tl$^+$)*	Lifetime of (Tl$^+$)* + STH diffusion (50 ns delayed rise) [3]	 large	Suppressed
(iii)	Slow: 1. e$^-$ + Tl$^+$ → Tl^0 2. Tl$^+$ + STH → Tl^{2+} 3. Tl^0 → Tl$^+$ + e$^-$ 4. e$^-$ + Tl^{2+} → (Tl$^+$)*	Electron release from Tl^0 (1 μs) [5]	 small	Suppressed (possibly)

- ❖ **Process (i) and (ii) constitute the primary decay component (230 ns).**
- ❖ **Process (iii) contributes 5% to the total light yield.**
- ❖ **Ca^{2+} (possibly Sr^{2+}) forms (Tl_{Na} + Ca_{Na})$^-$ dimer center with Tl$^+$ [6].**
  - (Tl_{Na} + Ca_{Na})$^-$ is an electron trap [6];
  - (Tl_{Na} + Ca_{Na})$^-$ is likely to be deeper than a standalone Tl$^+$ [4,6,7];
  - (Tl_{Na} + Ca_{Na})$^-$ competes with Tl$^+$ in electron trapping and inhibits the formation of Tl^0.
- ❖ **Ca^{2+} and Sr^{2+} co-doping suppress process (ii) and (iii), thus:**
  - Expedites scintillation decay;
  - Partially decreases light yield;
  - Reduces the downward trend toward high E on the NP curve.

Neutron-Gamma Dual Detection with Li$^+$ co-doped NaI:Tl$^+$

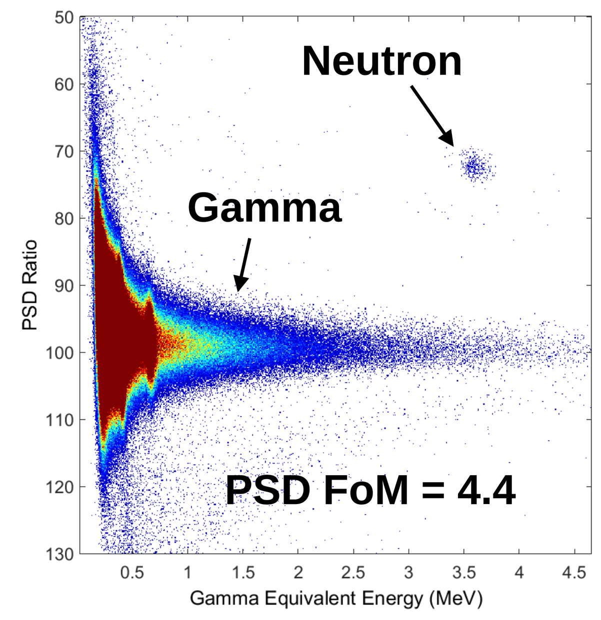


Fig. 5 PSD scatter plot of NaI:Tl crystals with 0.5% Li loading

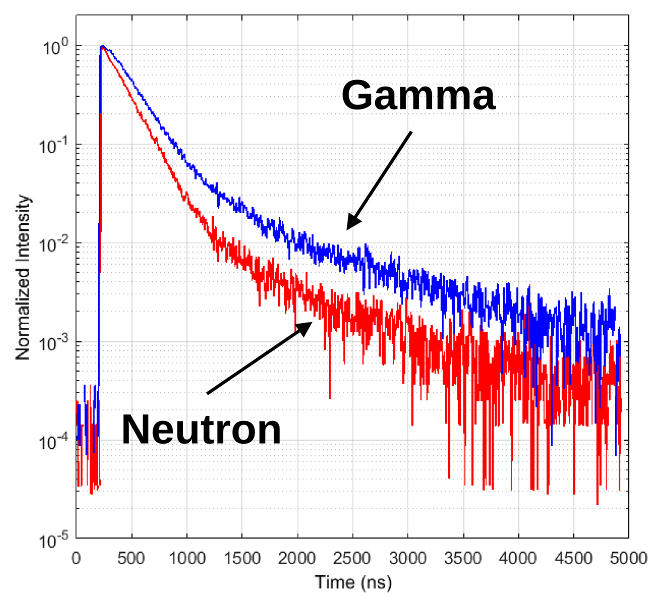


Fig. 6 averaged PMT traces for neutron and gamma pulse for NaI:Tl, Li crystal

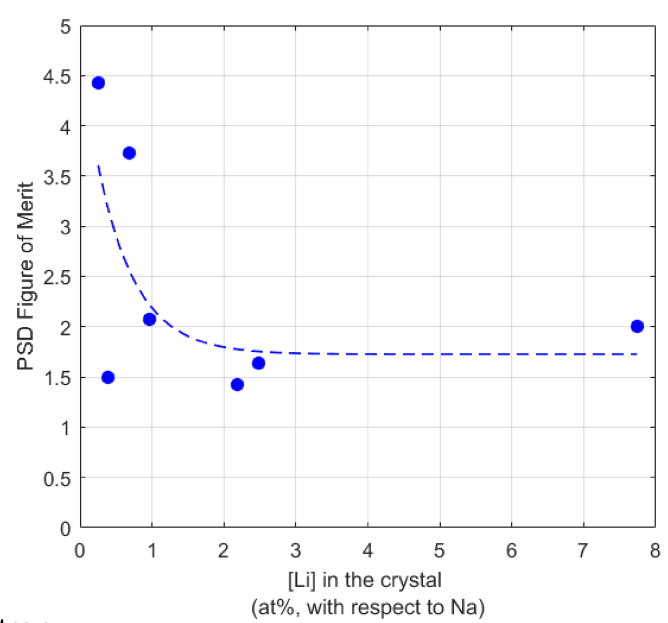


Fig. 7 n-γ PSD FoM vs [Li] in crystal

- ❖ **NaI:Tl, Li = large volume n-γ dual mode detector**
  - Low cost
  - Similar gamma performance as standard NaI:Tl$^+$
- ❖ **Exceptional pulse shape discrimination (PSD)**
  - PSD Figure of Merit = 1.5 – 4.4
  - Gamma rejection ratio <10^{-8}
- ❖ **High detection efficiency (neutrons + gammas)**
  - Li loading up to 8% confirmed
  - Large volume possible (>1000 cm^3)
  - Large area possible
  - Scaling up is already underway.
- ❖ **More details will be available at SPIE 2016 and IEEE NSS-MIC 2016.**

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