History of Scintillating Crystals


- **Fifties:** NaI:Tl and CsI:Tl
- **Seventies:** BGO, BaF$_2$
- **Nineties:** PWO, LSO:Ce
- **21 century:** LYSO:Ce, LaBr$_3$:Ce & LSO:Ce/Ca

**Invention of the photomultiplier tube**

- Cs$_2$LiYCl$_6$:Ce 2003
- LuI$_3$:Ce 2003
- K$_2$LaI$_5$:Ce 2002
- LaBr$_3$:Ce 2001
- LaCl$_3$:C 2000
- Lu$_2$O$_3$:Eu, Tb 2000
- Lu$_2$Si$_2$O$_7$:Ce 2000
- RbGd$_2$Br$_7$:Ce 1997
- $^6$Li$_6$Gd(BO$_3$)$_3$:Ce 1996

Fast UV response

Trigger

HPGe

Ge:Li

Fifties: NaI:Tl and CsI:Tl

April 28, 2011

Talk presented in the Time Resolution Workshop at Chicago by Ren-yuan Zhu, Caltech
# Crystals for HEP Calorimeters

<table>
<thead>
<tr>
<th>Crystal</th>
<th>NaI(Tl)</th>
<th>CsI(Tl)</th>
<th>CsI</th>
<th>BaF$_2$</th>
<th>BGO</th>
<th>LSO(Ce)</th>
<th>PWO</th>
<th>PbF$_2$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Density (g/cm$^3$)</td>
<td>3.67</td>
<td>4.51</td>
<td>4.51</td>
<td>4.89</td>
<td>7.13</td>
<td>7.40</td>
<td>8.3</td>
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<td>Melting Point (°C)</td>
<td>651</td>
<td>621</td>
<td>621</td>
<td>1280</td>
<td>1050</td>
<td>2050</td>
<td>1123</td>
<td>824</td>
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<tr>
<td>Radiation Length (cm)</td>
<td>2.59</td>
<td>1.86</td>
<td>1.86</td>
<td>2.03</td>
<td>1.12</td>
<td>1.14</td>
<td>0.89</td>
<td>0.93</td>
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<td>4.13</td>
<td>3.57</td>
<td>3.57</td>
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<td>2.23</td>
<td>2.07</td>
<td>2.00</td>
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<td>1.95</td>
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<td>2.15</td>
<td>1.82</td>
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<td>Slight</td>
<td>Slight</td>
<td>No</td>
<td>No</td>
<td>No</td>
<td>No</td>
<td>No</td>
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<tr>
<td>Luminescence $^b$ (nm) (at peak)</td>
<td>410</td>
<td>550</td>
<td>420</td>
<td>300</td>
<td>480</td>
<td>402</td>
<td>425</td>
<td>420</td>
</tr>
<tr>
<td>Decay Time $^b$ (ns)</td>
<td>245</td>
<td>1220</td>
<td>30</td>
<td>6</td>
<td>650</td>
<td>0.9</td>
<td>300</td>
<td>40</td>
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<tr>
<td>Light Yield $^{b,c}$ (%)</td>
<td>100</td>
<td>165</td>
<td>3.6</td>
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<td>21</td>
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<td>0.1</td>
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<tr>
<td>$d$(LY)/dT $^b$ (%/°C)</td>
<td>-0.2</td>
<td>0.4</td>
<td>-1.4</td>
<td>-1.9</td>
<td>-0.9</td>
<td>-0.2</td>
<td>-2.5</td>
<td>?</td>
</tr>
</tbody>
</table>

**Experiment**

- Crystal Ball
- BaBar
- BELLE
- BES III
- KTeV
- TAPS (L$^*$)
- L3
- BELLE
- Mu2e
- SuperB
- SLHC?
- CMS
- ALICE
- PANDA
- A4
- HHCAL?

---

*a. at peak of emission; b. up/low row: slow/fast component; c. QE of readout device taken out.*
## Crystals for Homeland Security

<table>
<thead>
<tr>
<th>Crystal</th>
<th>NaI(Tl)</th>
<th>CsI(Tl)</th>
<th>CsI(Na)</th>
<th>LaCl₃(Ce)</th>
<th>SrI₂(Eu)</th>
<th>LaBr₃(Ce)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Density (g/cm³)</td>
<td>3.67</td>
<td>4.51</td>
<td>4.51</td>
<td>3.86</td>
<td>4.59</td>
<td>5.29</td>
</tr>
<tr>
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<td>621</td>
<td>621</td>
<td>859</td>
<td>538</td>
<td>788</td>
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<td>Radiation Length (cm)</td>
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<td>1.86</td>
<td>2.81</td>
<td>1.95</td>
<td>1.88</td>
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<td>3.57</td>
<td>3.71</td>
<td>3.40</td>
<td>2.85</td>
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<tr>
<td>Interaction Length (cm)</td>
<td>42.9</td>
<td>39.3</td>
<td>39.3</td>
<td>37.6</td>
<td>37.0</td>
<td>30.4</td>
</tr>
<tr>
<td>Refractive Index a</td>
<td>1.85</td>
<td>1.79</td>
<td>1.95</td>
<td>1.9</td>
<td>?</td>
<td>1.9</td>
</tr>
<tr>
<td>Hygroscopicity</td>
<td>Yes</td>
<td>Slight</td>
<td>Slight</td>
<td>Yes</td>
<td>Yes</td>
<td>Yes</td>
</tr>
<tr>
<td>Luminescence b (nm) (at peak)</td>
<td>410</td>
<td>550</td>
<td>420</td>
<td>335</td>
<td>435</td>
<td>356</td>
</tr>
<tr>
<td>Decay Time b (ns)</td>
<td>245</td>
<td>1220</td>
<td>690</td>
<td>570</td>
<td>1100</td>
<td>20</td>
</tr>
<tr>
<td>Light Yield b,c (%)</td>
<td>100</td>
<td>165</td>
<td>88</td>
<td>13</td>
<td>221</td>
<td>130</td>
</tr>
<tr>
<td>d(LY)/dT b (%)</td>
<td>-0.2</td>
<td>0.4</td>
<td>0.4</td>
<td>0.1</td>
<td>?</td>
<td>0.2</td>
</tr>
</tbody>
</table>

---

a. at peak of emission; b. up/low row: slow/fast component; c. QE of readout device taken out.
Crystal Density: Radiation Length

1.5 $X_0$ Cube Samples:
- Hygroscopic: Sealed
- Non-hygro: Polished

Full Size Crystals:
- BaBar CsI(Tl): 16 $X_0$
- L3 BGO: 22 $X_0$
- CMS PWO(Y): 25 $X_0$
**Excitation, Emission, Transmission**

\[
T_s = (1 - R)^2 + R^2(1 - R)^2 + \ldots = (1 - R)/(1 + R), \text{ with } R = \frac{(n_{\text{crystal}} - n_{\text{air}})^2}{(n_{\text{crystal}} + n_{\text{air}})^2}.
\]


Poor transmittance indicates scattering centers: LaBr\(_3\) and LaCl\(_3\).
Scintillation Light Decay Time

Recorded with an Agilent 6052A digital scope

Fast Scintillators

- $\tau = 30/6$ ns, CsI
- $\tau = 20$ ns, LaBr$_3$
- $\tau = 35$ ns, CeF$_3$
- $\tau = 30/10$ ns, PWO
- $\tau = 40$ ns, LSO
- $\tau = 40$ ns, LYSO

Slow Scintillators

- $\tau = 1250$ ns, CsI(Tl)
- $\tau = 630$ ns, CsI(Na)
- $\tau = 230$ ns, NaI(Tl)
- $\tau = 300$ ns, BGO
- $\tau = 600/25$ ns, LaCl$_3$(Ce)
- $\tau = 630/0.9$ ns, BaF$_2$
Rising Time for $1.5 \times X_0$ Samples

Agilent MSO9254A (2.5 GHz) DSO with 0.14 ns rise time
Hamamatsu R2059 PMT (2500 V) with rise time 1.3 ns

Measured rising time is dominated by photo-detector response, and is affected by light propagation in crystal.
Light Output & Decay Kinetics

Measured with Photonis XP2254B PMT (multi-alkali cathode)

p.e./MeV: LSO/LYSO is 6 & 230 times of BGO & PWO respectively

Fast Crystal Scintillators

- LaBr₃

Slow Crystal Scintillators

- LSO/LYSO
- BaF₂

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Emission Weighted QE

Taking out QE, L.O. of LSO/LYSO is 4/200 times BGO/PWO
Hamamatsu S8664-55 APD has QE 75% for LSO/LYSO

Hamamatsu PMT, R1306
- BGO: QE = 8.0 ± 0.4%
- LSO/LYSO: QE = 12.9 ± 0.6%
- CsI(Tl): QE = 5.0 ± 0.3%

Photonis PMT, XP2254B
- BGO: QE = 4.7 ± 0.2%
- LSO/LYSO: QE = 7.2 ± 0.4%
- CsI(Tl): QE = 3.5 ± 0.2%

Hamamatsu APD, S8664-55
- BGO: QE = 82 ± 4%
- LSO/LYSO: QE = 75 ± 4%
- CsI(Tl): QE = 84 ± 4%

Hamamatsu PD, S2744
- BGO: QE = 75 ± 4%
- LSO/LYSO: QE = 59 ± 3%
- CsI(Tl): QE = 80 ± 4%
$^{137}$Cs FWHM Energy Resolution

3% to 80% measured with Hamamatsu R1306 PMT with bi-alkali cathode

2% resolution and proportionality are important for $\gamma$-ray spectroscopy between 10 keV to 2 MeV
For TOF PET it is defined as the minimum time interval required to separate two subsequent photon events, it is the FWHM of the time difference distribution. For a detector with finite signal to noise ratio and transit time jitter it can be written as

\[ \sigma_{time}^2 \approx \left( \frac{\sigma_{noise}}{dV/dt} \right)^2 + \sigma_{TTS}^2 \]

where both the signal slope \((dV/dt)\) at the point of time pick-off and the jitter contribute. A fast and bright crystal scintillator combined with a photo-detector with high gain, fast response and small jitter are important for TOF PET.

TOF resolution in HEP is usually defined as the rms for single particles. Its numerical value is 3.3 times smaller than TOF PET.
The intrinsic rising time of fast crystals is 30 ps. Most measured time resolutions are dominated by instrumental effects, such as photo-detector responses and light propagations inside crystals.

Optimization of time resolution:
- Fast crystal scintillators: with fast rise time & maximum photo-electron numbers in the 1\textsuperscript{st} ns.
- Quantum efficiency of photo-detectors matches scintillation emission peak.
- Select appropriate pick off time to maximize dV/dt and minimize jitters.
A Measurement of Rising time

S. Derenzo et al., IEEE TNS 47 (2000) 860-864

Excitation: 60 ps FWHM x-ray pulse
Detector: MCP with 45 ps response

Effects of light propagation inside the samples are noticed: difference between small and large size samples and different wrappings.
### Crystal Scintillation Rising Time

S. Derenzo et al., IEEE TNS 47 (2000) 860-864

<table>
<thead>
<tr>
<th>Sample</th>
<th>Form*</th>
<th>Supplier**</th>
<th>$\tau_r \text{ (ps)}$†</th>
<th>$\tau_d \text{ (ns)‡‡}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>BaF$_2$</td>
<td>2 x 10 x 10 mm</td>
<td>Harshaw</td>
<td>0 (calibration)</td>
<td>0.12(2%), 0.78(12%) + longer</td>
</tr>
<tr>
<td>BaF$_2$</td>
<td>powder</td>
<td>Aesar, Inc.</td>
<td>&lt; 30</td>
<td>0.18(2%), 0.88(8%) + longer</td>
</tr>
<tr>
<td>BC-422</td>
<td>2 x 10 x 20 mm</td>
<td>Bicron</td>
<td>&lt; 30</td>
<td>1.1(59%), 2.3(29%) + longer</td>
</tr>
<tr>
<td>Bi$_4$Ge$<em>3$O$</em>{12}$</td>
<td>3 x 3 x 30 mm</td>
<td>Harshaw</td>
<td>30 ±30</td>
<td>5.8(1%), 28(4%) + longer</td>
</tr>
<tr>
<td>CaF$_2$:Eu</td>
<td>2 x 30 x 30 mm</td>
<td>Bicron</td>
<td>40±30</td>
<td>slow decay components</td>
</tr>
<tr>
<td>CeF$_3$</td>
<td>2 x 10 x 10 mm</td>
<td>Optovac</td>
<td>30±30</td>
<td>several decay components</td>
</tr>
<tr>
<td>CdS:Te</td>
<td>2 x 5 x 10 mm</td>
<td>Peter Trower</td>
<td>80±30</td>
<td>several decay components</td>
</tr>
<tr>
<td>CdWO$_4$</td>
<td>2 x 10 x 10 mm</td>
<td>Harshaw</td>
<td>&lt; 30</td>
<td>13(1%) + longer</td>
</tr>
<tr>
<td>CsI</td>
<td>2 x 10 x 10 mm</td>
<td>Bicron</td>
<td>30±30</td>
<td>several decay components</td>
</tr>
<tr>
<td>CsI:Tl</td>
<td>10 x 10 x 10 mm</td>
<td>Optovac</td>
<td>9,500(66%), 41,000(34%)§</td>
<td>long decay</td>
</tr>
<tr>
<td>Lu$_2$SiO$_5$:Ce</td>
<td>3 x 3 x 30 mm</td>
<td>CTI</td>
<td>30±30 (88%), 350±70(12%)</td>
<td>7(1%), 38.8(99%)</td>
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<tr>
<td>PbWO$_4$</td>
<td>2 x 10 x 10 mm</td>
<td>FSU</td>
<td>60±30</td>
<td>several decay components</td>
</tr>
<tr>
<td>YAlO$_3$:Ce</td>
<td>10 x 10 x 10 mm</td>
<td>Peter Trower</td>
<td>240±50</td>
<td>26(90%), 67(10%),</td>
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<tr>
<td>ZnO:Ga</td>
<td>powder</td>
<td>Westinghouse</td>
<td>&lt; 30</td>
<td>0.36(35%), 0.82(65%)</td>
</tr>
</tbody>
</table>

*All samples (except powders) roughened and painted black on five sides

**Bicron Chemical, Solon, Ohio; FSU = Former Soviet Union; CTI, Inc. Knoxville, TN; Optovac, Inc., North Brookfield, MA.

†Best fit rise time using x-ray tube calibration of 60 ps fwhm shown in Figure 4.

‡‡Decay times may vary from sample to sample.

§Rise time is temperature dependent. See Ref. [9].

**Scintillation rising time is about 30 ps or less for BaF$_2$, BGO and CeF$_3$**
Crystals of high density, good UV transmittance and some scintillation light, not necessary bright and fast, are required. The volume needed is 70 to 100 m$^3$: cost-effective material. Following 2/19/08 workshop at SICCAS, 5 x 5 x 5 cm samples evaluated.
Cosmic Setup with Dual Readout

- UG11 and GG 400 used to select Cherenkov & scintillation
- Agilent 6052A (500 MHz) DSO with rise time 0.7 ns
- Hamamatsu R2059 PMT (2500 V) with rise time 1.3 ns
No Discrimination in Front Edge

Consistent timing and rise time for all Cherenkov and scintillation light pulses with crystals wrapped black.

April 28, 2011
Talk presented in the Time Resolution Workshop at Chicago by Ren-yuan Zhu, Caltech
Effect of Light Propagation

Reflectors increase light output, but slow down rising time. Appropriate choice of time pick-off may avoid this effect.

Reflectors increase light output, but slow down rising time. Appropriate choice of time pick-off may avoid this effect.

- BGO (5 x 5 x 5 cm)
  - τ: 302 ns (black tape wrapping)
  - τ: 318 ns (tyvek wrapping)

- BGO (5 x 5 x 5 cm)
  - τ: 2.0 ns (Black tape wrapping)
  - τ: 20 ns (Tyvek wrapping)
Effect of Time Pick-Off

S. Ziegler et al., IEEE TNS 37 (1990) 574-579
Time resolution may be optimized by appropriate trigger threshold

200 ps FWHM, or 60 ps $\sigma_{\text{single}}$, measured with BaF$_2$ & Hamamatsu R3377 PMT readout: a factor of three better than commercial system
Figure of Merit for Crystals

FoM is calculated as the LY in 1st ns obtained by using light output and decay time data measured for 1.5 $X_0$ crystal samples.

<table>
<thead>
<tr>
<th>Crystal Scintillators</th>
<th>Relative LY (%)</th>
<th>$A_1$ (%)</th>
<th>$\tau_1$ (ns)</th>
<th>$A_2$ (%)</th>
<th>$\tau_2$ (ns)</th>
<th>Total LO (p.e./MeV, XP2254B)</th>
<th>LO in 1ns (p.e./MeV, XP2254B)</th>
<th>LO in 0.1ns (p.e./MeV, XP2254B)</th>
<th>LY in 0.1ns (photons/MeV)</th>
</tr>
</thead>
<tbody>
<tr>
<td>BaF$_2$</td>
<td>40.1</td>
<td>91</td>
<td>650</td>
<td>9</td>
<td>0.9</td>
<td>1149</td>
<td>71.0</td>
<td>11.0</td>
<td>136.6</td>
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<tr>
<td>LSO:Ca$_2$Ce</td>
<td>94</td>
<td>100</td>
<td>30</td>
<td></td>
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<td>2400</td>
<td>78.7</td>
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<td>LSO/LSO:Ce</td>
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<td>100</td>
<td>40</td>
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<td></td>
<td>2180</td>
<td>53.8</td>
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<td></td>
<td>208</td>
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<td></td>
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<td>0.1</td>
<td>2.5</td>
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<td>PWO</td>
<td>0.377</td>
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<td>10</td>
<td>9.2</td>
<td>0.42</td>
<td>0.04</td>
<td>0.4</td>
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<td>LaBr$_3$:Ce</td>
<td>130</td>
<td>100</td>
<td>20</td>
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<td>3810</td>
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<td>55</td>
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<td>570</td>
<td>76</td>
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<td>100</td>
<td>245</td>
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<td>2604</td>
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<td>14.5</td>
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<td>77</td>
<td>30</td>
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<td>131</td>
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<td>0.8</td>
<td>10.6</td>
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<td>CsI:Tl</td>
<td>165</td>
<td>100</td>
<td>1220</td>
<td></td>
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<td>2093</td>
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<td>4.8</td>
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<td>CsI:Na</td>
<td>88</td>
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<td>690</td>
<td></td>
<td></td>
<td>2274</td>
<td>3.3</td>
<td>0.3</td>
<td>4.5</td>
</tr>
</tbody>
</table>

The best crystal scintillator for time resolution is BaF$_2$ and LSO: Ce/Ca and LYSO. LaBr$_3$ is a material with high potential.
## TOF with LSO & LYSO Crystals

100 ps achieved with LYSO & LSO:Ce/Ca with MCP & G-APD readout.

<table>
<thead>
<tr>
<th>Test</th>
<th>Crystal</th>
<th>Detector</th>
<th>Particle</th>
<th>$\sigma_{\text{single}}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fermilab</td>
<td>LYSO 3 x 3 x 7 mm</td>
<td>G-APD 3 mm$^2$</td>
<td>$^{60}\text{Co}: \gamma$'s</td>
<td>110 ps</td>
</tr>
<tr>
<td>PISA</td>
<td>LSO:Ce/Ca 3 x 3 x 10 mm</td>
<td>G-APD 3 mm$^2$</td>
<td>$^{22}\text{Na}: \gamma$'s</td>
<td>107 ps</td>
</tr>
<tr>
<td>SLAC</td>
<td>LYSO 17 mm Cube</td>
<td>MCP</td>
<td>CRT: $\mu$'s</td>
<td>109/159 ps</td>
</tr>
<tr>
<td>SLAC</td>
<td>Scintillator 17 mm Cube</td>
<td>G-APD</td>
<td>CRT: $\mu$'s</td>
<td>136 ps</td>
</tr>
<tr>
<td>SLAC</td>
<td>LYSO 17 mm Cube</td>
<td>G-APD</td>
<td>CRT: $\mu$'s</td>
<td>140 ps</td>
</tr>
<tr>
<td>SLAC</td>
<td>LYSO 2.5 x 2.5 x 20 cm</td>
<td>G-APD</td>
<td>CRT: $\mu$'s</td>
<td>220 ps</td>
</tr>
</tbody>
</table>

100 ps corresponds to 330 ps for TOF-PET
Time Resolution of LaBr$_3$

J. Glodo et al., IEEE TNS 52 (2005) 1805-1808

<table>
<thead>
<tr>
<th>Ce$^{3+}$ Contri. %</th>
<th>Light Output %</th>
<th>Decay / Rise Times (intensity) ns/ns (%</th>
<th>Effective Rise Time, ns</th>
<th>Timing Resolution</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.5</td>
<td>97</td>
<td>19/15 (56%), 15.2/2 (28%), 55 (16%)</td>
<td>9.4</td>
<td>390 361 83</td>
</tr>
<tr>
<td>5.0</td>
<td>100</td>
<td>15/0.38 (70%), 15/2.2 (27%), 55 (3%)</td>
<td>0.93</td>
<td>260 214 62</td>
</tr>
<tr>
<td>10.0</td>
<td>94</td>
<td>16.5/0.5 (89%), 4.5/0.5 (5%), 55 (6%)</td>
<td>0.5</td>
<td>182 106 67</td>
</tr>
<tr>
<td>20.0</td>
<td>92</td>
<td>17.5/0.16 (89%), 4.5/0.15 (5%), 55 (6%)</td>
<td>0.16</td>
<td>177 97 70</td>
</tr>
<tr>
<td>30.0</td>
<td>93</td>
<td>18/0.2 (91%), 2.5/0.2 (4%), 55 (6%)</td>
<td>0.20</td>
<td>165 73 70</td>
</tr>
</tbody>
</table>

Time resolution achievable: 100 ps for TOF-PET with MCP readout.
Summary

• Most fast crystals have a rising time at a level of 30 ps. Laboratory measurements of TR are often dominated by the response time of photo-detectors and readout electronics, and also affected by light propagation.

• Time resolution of a scintillator based system can be optimized by choosing (1) bright and fast scintillators, (2) fast photo-detector with high gain and low jitter and (3) appropriate time pick off or trigger threshold.

• Photo-electron # in the 1st ns can be seen as a figure of merit for time resolution of crystals with fast rise time.

• The achievable FWHM time resolution for TOF PET is about 100 ps by LaBr₃, and 200 and 300 ps respectively for non-hygroscopic crystals BaF₂ and LSO/LYSO.