Scintillation materials for ultra-fast gamma timing

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- Motivation: gamma timing in nuclear materials sciences (3 examples)
- Positron Annihilation Spectroscopy
- Timing using scintillators on PMT or MCP-PMT
- Alternative: Cherenkov radiation instead of scintillation
- Gamma-induced Positron Spectroscopy (GiPS)
Motivation: gamma timing in nuclear materials sciences

There are several powerful spectroscopies using nuclear techniques:

1. **Example: Perturbed Angular Correlation (PAC):**
   - time-dependent change of $\gamma$-emission spectrum of radioactive probe atoms
   - Time and energy of gamma events are recorded

PAC spectrum

Digital PAC Spectrometer (Univ. Göttingen)
2. Example: Neutron Time of Flight Spectrometer at ELBE (Research Center Dresden-Rossendorf)

- monoenergetic pulsed neutron beam
- energy is measured as time difference in TOF spectrometer by fast PMT

Neutron TOF Spectrometer using a multi-detector gamma spectrometer

Neutron-TOF Spectrum
3. Example: Positron Lifetime Spectroscopy

- positron wave-function can be localized in the attractive potential of a defect
- annihilation parameters change in the localized state
- e.g. positron lifetime increases in a vacancy
- lifetime is measured as time difference between appearance of 1.27 (start) and 0.51 MeV (stop) quanta
- defect identification and quantification possible
Digital lifetime measurement

PMT: Hamamatsu H3378-50

- very simple setup
- timing very accurate ($<10^{-6}$)
- pulse-shape discrimination (suppress "bad pulses")
- each detector for start & stop (double statistics)

Digitizer: Acqiris
8 GS/s & 3 GHz bandwidth
screenshot of two digitized anode pulses

time difference = 2.65471 samples = 663.67 ps
Positron lifetime spectroscopy

- positron lifetime spectra consist of exponential decay components
- positron trapping in open-volume defects leads to long-lived components
- longer lifetime due to lower electron density
- analysis by non-linear fitting: lifetimes $\tau_i$ and intensities $I_i$

$$N(t) = \sum_{i=1}^{k+1} \frac{I_i}{\tau_i} \exp\left(-\frac{t}{\tau_i}\right)$$

- positron lifetime spectrum:
  - trapping coefficient
  - trapping rate
  - defect concentration

Time resolution is a Gaussian $\approx 230$ ps (FWHM)
Improvement of time resolution

- Standard time resolution for positron lifetime spectroscopy is nowadays 180...250 ps (positron lifetime in Fe: 108 ps; in Si: 218 ps)
- Our aim: to reach time resolution (FWHM) < 100 ps

- Time resolution is due to combination of PMT (Photomultiplier) and Scintillator
- Best PMT's have rise time of < 1 ns and
- electron transition time spread (TTS) = 100...300 ps (most important)
- in future: Microchannel plate PMTs (MCP PMTs) have TTS = 25 ps!
### Selection of Photo-Multiplier Tubes (PMTs)

<table>
<thead>
<tr>
<th>Type</th>
<th>Philips</th>
<th>HAMAMATSU</th>
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<tbody>
<tr>
<td></td>
<td>XP2020 Head-on</td>
<td>H3378-50 Head-on</td>
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<tr>
<td></td>
<td>BA 51.0</td>
<td>BA 51.0</td>
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<tr>
<td></td>
<td>fused silica</td>
<td>fused silica</td>
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<tr>
<td></td>
<td>160-650</td>
<td>160-650</td>
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<tr>
<td></td>
<td>420</td>
<td>420</td>
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<tr>
<td></td>
<td>0.25</td>
<td>0.24</td>
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<tr>
<td></td>
<td>3000</td>
<td>3000</td>
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<tr>
<td></td>
<td>3×10⁷</td>
<td>2.5×10⁶</td>
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<tr>
<td></td>
<td>1.4</td>
<td>0.7</td>
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<tr>
<td></td>
<td>28</td>
<td>16</td>
</tr>
<tr>
<td></td>
<td>~200</td>
<td>370</td>
</tr>
<tr>
<td></td>
<td>1000</td>
<td>3650</td>
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<tr>
<td></td>
<td>R7400U-09 Metal package</td>
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<tr>
<td></td>
<td>Cs-Te</td>
<td>MgF₂</td>
</tr>
<tr>
<td></td>
<td>160-320</td>
<td>115-320</td>
</tr>
<tr>
<td></td>
<td>240</td>
<td>230</td>
</tr>
<tr>
<td></td>
<td>0.11</td>
<td>0.11</td>
</tr>
<tr>
<td></td>
<td>5×10⁴</td>
<td>2×10⁴⁵</td>
</tr>
<tr>
<td></td>
<td>0.78</td>
<td>0.15</td>
</tr>
<tr>
<td></td>
<td>5.4</td>
<td>0.55</td>
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<tr>
<td></td>
<td>~100</td>
<td>25</td>
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<tr>
<td></td>
<td>700</td>
<td>15000</td>
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</tbody>
</table>

- **Selection of PMT**

- **Philips**
  - XP2020 Head-on
  - BA 51.0
  - fused silica 160-650
  - 3000 V
  - 3×10⁷ gain
  - 1.4 rise time (ns)
  - 28 transit time (ns)
  - ~200 TTS (ps)
  - 1000 cost (EUR)

- **HAMAMATSU**
  - H3378-50 Head-on
  - BA 51.0
  - fused silica 160-650
  - 3000 V
  - 2.5×10⁶ gain
  - 0.7 rise time (ns)
  - 16 transit time (ns)
  - 370 TTS (ps)
  - 3650 cost (EUR)

- **Comparison**
  - Old tube, reliable, but rather slow.
  - Standard tube, large, efficient, pretty fast.
  - Very fast, but too small, not efficient enough.
  - Very promising, extremely fast, not many experiences, very expensive.

**Martin-Luther-Universität Halle**
Photocathode of PMTs sensitive for increase of TTS

- Position of photon encounter on photocathode has large influence on transition time, and thus on time resolution
- x-y measurement with fast photodiode show time difference at anode of > 1 ns!
- tubes must be analyzed this way individually (we will do with ps-Laser)

Cherenkov radiation for extremely fast timing

- Cherenkov radiation is formed as visible light emitted when a charged particle or photon passes through an insulator at a speed greater than the speed of light in that medium.
- e.g. when gamma rays go through silica glass.
- Advantage: almost instantaneous generation of light, definitely faster than scintillation light.
- So test of PMT without influence of scintillator.
- And: very fast timing possible.
- Disadvantage: only a few photoelectrons, thus no energy information of incoming gamma.
- Limitation – low photoelectron yield requiring phototubes with best possible TTS.
- MCP-PMT a possible candidate.
• Cherenkov photon from Plexiglas hit by 1.17 and 1.33 MeV quanta of 60-Co on top of a MCP PMT
• rise time 750 ps - very short!
## Overview: scintillator materials for fast timing

<table>
<thead>
<tr>
<th>Scintillator</th>
<th>Wavelength [nm]</th>
<th>Density [g/ccm]</th>
<th>Luminosity (10^3) photons/MeV</th>
<th>Energy Resolution % FWHM @ 662keV</th>
<th>Rise time (10 - 90%) [ns]</th>
<th>Measured Timing Resolution [ps]</th>
</tr>
</thead>
<tbody>
<tr>
<td>BaF2</td>
<td>220</td>
<td>4.88</td>
<td>2</td>
<td>11.4</td>
<td>0.6 - 1</td>
<td>0.9</td>
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<tr>
<td></td>
<td>310</td>
<td></td>
<td>10</td>
<td></td>
<td></td>
<td></td>
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<tr>
<td>LSO</td>
<td>420</td>
<td>7.4</td>
<td>27 – 33</td>
<td>7.9</td>
<td>40</td>
<td>275</td>
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<tr>
<td>LaBr3(Ce)</td>
<td>356 387</td>
<td>5.1</td>
<td>60 75</td>
<td>2.7 3.2</td>
<td>30</td>
<td>5.5</td>
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<td>ZnO:Ga</td>
<td>385</td>
<td>5.7</td>
<td></td>
<td></td>
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<tr>
<td>ZnO</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>0.75</td>
</tr>
<tr>
<td>Plastic (BC-422)</td>
<td>380</td>
<td>1.032</td>
<td></td>
<td></td>
<td></td>
<td>0.35</td>
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<tr>
<td>NaI (for reference)</td>
<td>415</td>
<td>3.67</td>
<td>44</td>
<td>5.6 – 7.1</td>
<td>50 ns</td>
<td></td>
</tr>
</tbody>
</table>

[1] scintillator.lbl.gov

Rise times are measured using Hamamatsu photomultipliers H3378-50
Timing of Scintillators

- demand: high-z for high efficiency and short rise time
- $\text{LaBr}_3(\text{Ce})$ is very efficient but very slow
- LSO ($\text{Lu}_2\text{SiO}_5$): rather heavy; very slow decay of scintillation light; not hygroscopic
- organic plastic: relatively fast; no gamma backscattering; too inefficient
- $\text{BaF}_2$: very fast rise and decay; slow component 310 nm - 600ns; hygroscopic
- $\text{ZnO}$: extremely fast rise time and decay - most promising

![Normalized Anode Pulses](image)

- Anode pulses are digitized by 8 GS/s and 3 GHz bandwidth
- measured with standard Hamamatsu H3378-50 PMT (rise time 0.7 ns)
ZnO is most promising

- ZnO clearly faster than BaF$_2$
- real rise and decay time not measurable with our setup
- problem up to now: no large single crystals available

ZnO: rise time$_{10-90\%}$=700 ps
Comparison of efficiency

- undoped ZnO exhibit very small efficiency
- LSO ends at high level: contains Lu isotope (2.6% ; 176-Lu); still okay for coincidence measurement
• big ZnO single crystals are now available (up to 2"")
• we will try to grow Ce/Ga-doped samples

ZnO single crystal ∅33 mm, grown at Inst. of Crystal Growth (IKZ), Berlin

Fig. 4. Photoluminescence spectra for the four powder samples at 300 K. The spectra predominantly consist of fast, near-band-edge (NBE) emissions. (a) The vertical scale is logarithmic. (b) Enhanced view of the NBE region at 300 K. The PL peak positions of the GK31 and NETech powders are similar and occur at slightly shorter wavelengths than the other two powders.

Energy resolution required

- Positron lifetime measurement is start-stop measurement
- Gamma energy must be detected to discriminate between 1.27 MeV and 0.511 MeV quanta

![Energy spectra graph](graph)

- Both materials are okay, although the photo peak is completely missing in plastics
• all materials deliver sufficient energy information, beside ZnO
• using ZnO 0.511 and 1.27 MeV cannot be discriminated
• Improvement by doping (Al, Ga, Ce, Fe)?
Bremsstrahlung-induced highly penetrating probes for nondestructive assay and defect analysis

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GiPS: Gamma-induced Positron Spectroscopy

- Background reduction due to coincident measurement
- Lifetime measurement requires only detection of annihilation radiation of 511 keV
- Thus, ZnO can be used here
- We obtained 180 ps FWHM time resolution: hope for < 100 ps
The GiPS setup includes 6 Detectors (4 Ge and 2 BaF$_2$)
Water spectrum: GiPS spectrum compared with conventional measurement


- advantage of periodic positron source is obvious: background distinctly reduced for GiPS spectrum (red)

- spectra quality distinctly improved (however resolution still $\approx 180$ ps)
Conclusions

- new combinations of Photo-Multiplier Tubes (PMT) and scintillating materials will give better time resolution
- Multi-Channel Plate PMTs exhibit by far the lowest TTS (electron transition time spread) of ≈ 25 ps (normal PMTs: 100…300 ps)
- they are very promising but very expensive too
- BaF₂ is still one of the best choices for fast scintillation; however, exhibit long light component (310 nm, 600ns); hygroscopic
- ZnO is still faster (not hygroscopic, no long-lifetime component, visible light)
- we couldn’t measure the rise time of ZnO because of limitation of PMT and digitizer used. It must be < 700 ps (BaF₂ < 1 ns)
- however: hardly any energy information in the pulse height spectrum
- no problem for our EPOS System
- There: combination of MCP PMT with ZnO might lead to time resolution of < 100 ps

Talk available at http://positron.physik.uni-halle.de
Research Center Dresden-Rossendorf, 28.-30. September 2009

Workshop on Digital Signal Processing in Nuclear Science

http://positron.physik.uni-halle.de/EPOS/

Open-source Project
http://positron.physik.uni-halle.de/EPOS/Software/