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)

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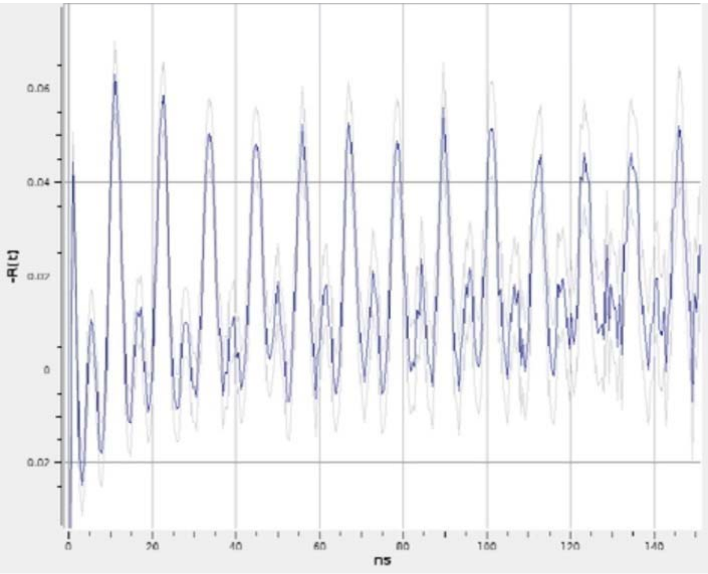
Motivation: gamma timing in nuclear materials sciences

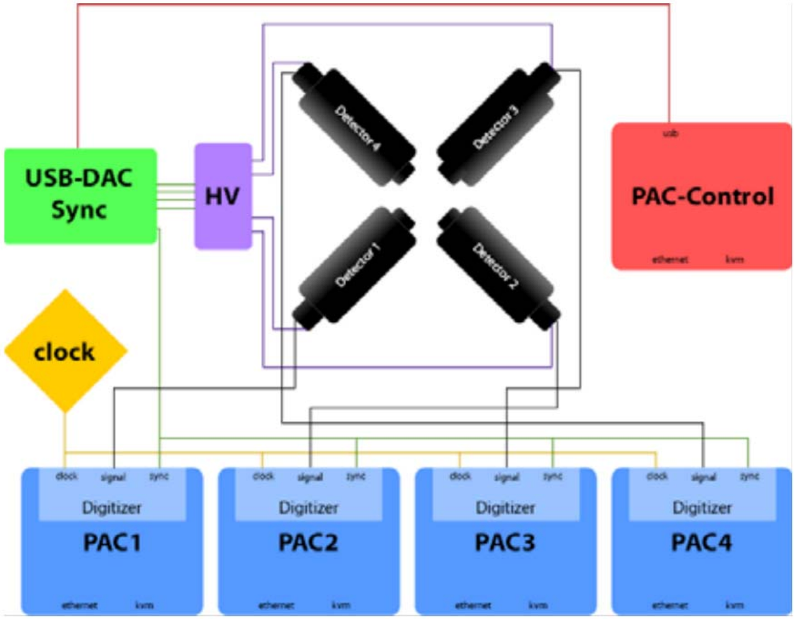
There are several powerful spectroscopies using nuclear techniques:

1. Example: Perturbed Angular Correlation (PAC):

- time-dependent change of γ-emission spectrum of radioactive probe atoms
- Time and energy of gamma events are recorded



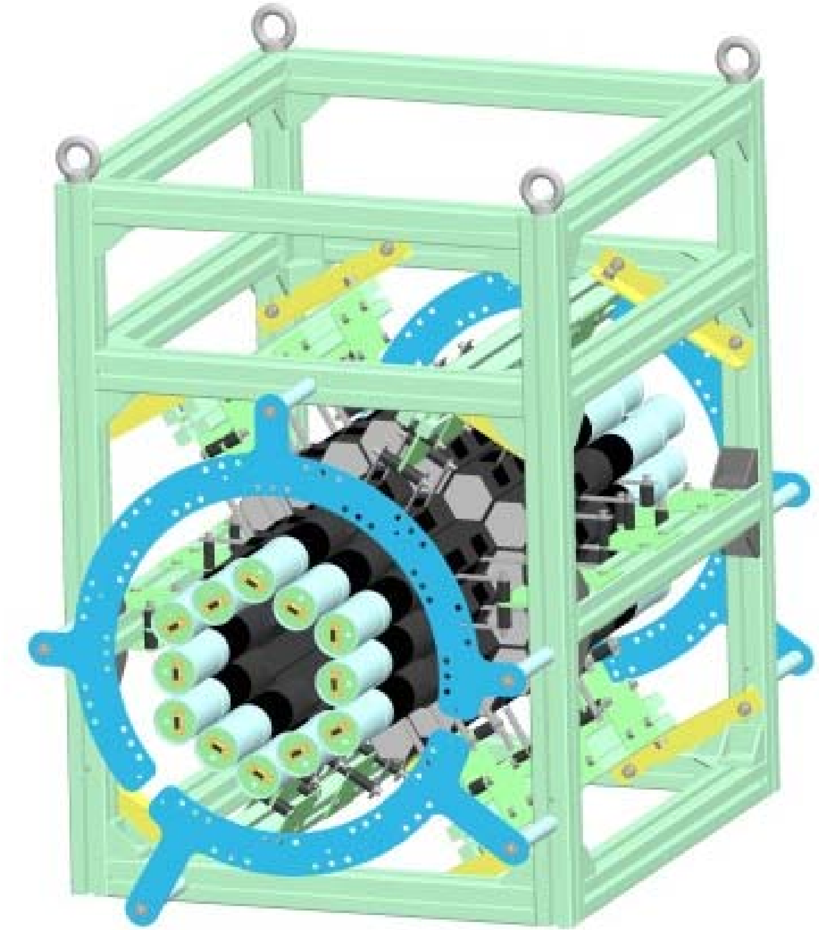
PAC spectrum



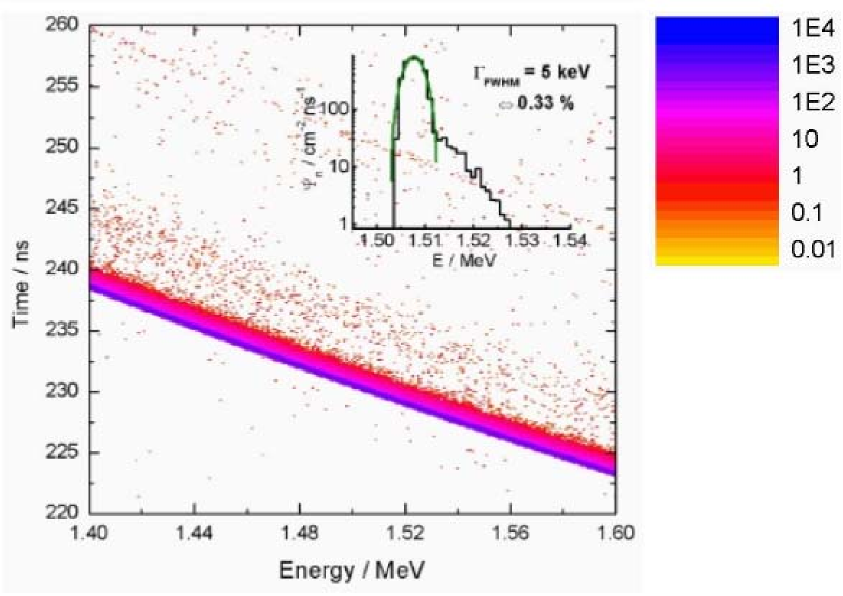
Digital PAC Spectrometer (Univ. Göttingen)

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*



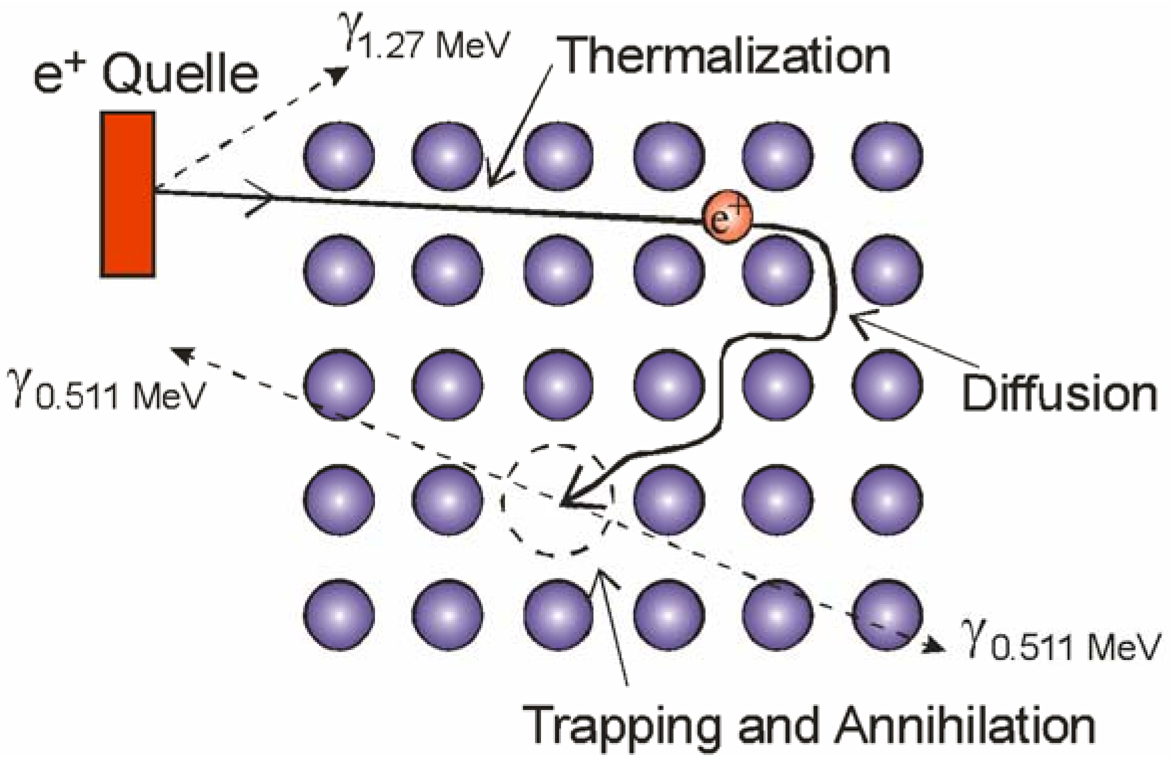
Neutron TOF Spectrometer using a multi-detector gamma spectrometer

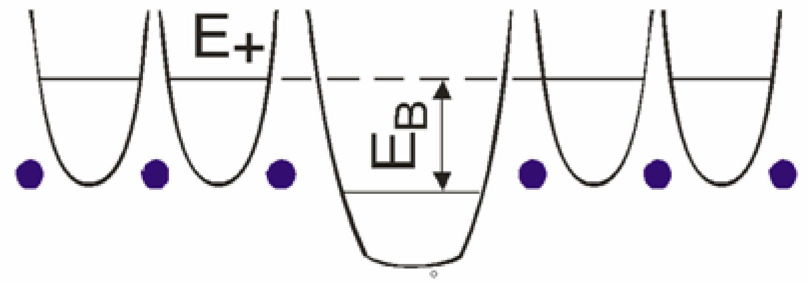


Neutron-TOF Spectrum

3. Example: Positron Lifetime Spectroscopy

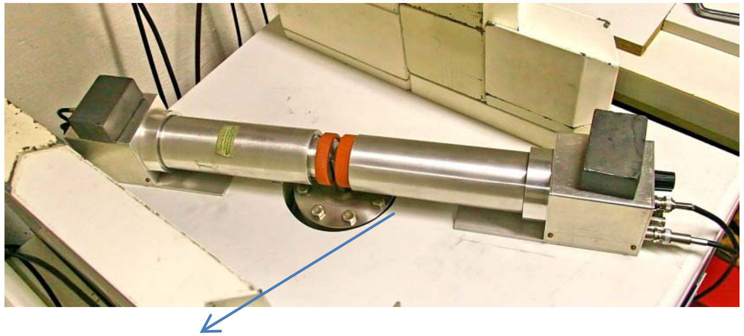
^{22}Na



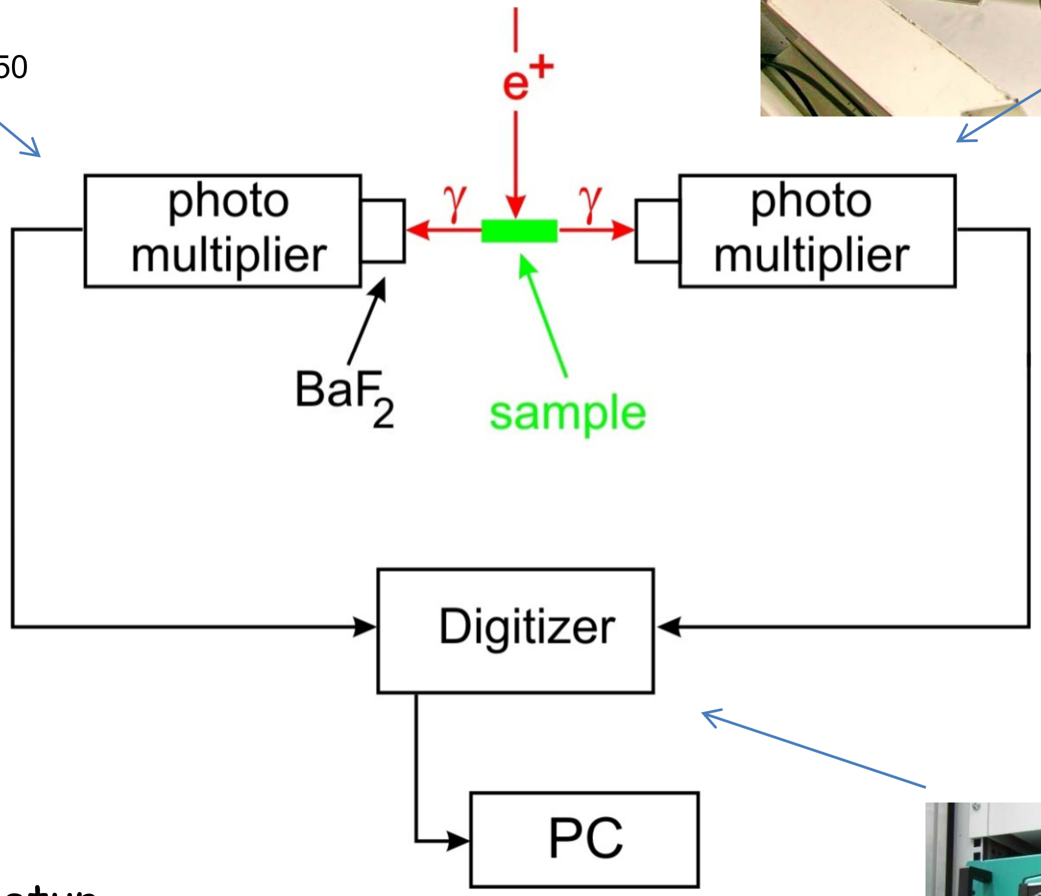


- positron wave-function can be localized in the attractive potential of a defect
- annihilation parameters change in the localized state
- e.g. positron lifetime increases in a vacancy
- lifetime is measured as time difference between appearance of 1.27 (start) and 0.51 MeV (stop) quanta
- defect identification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

Digital lifetime measurement



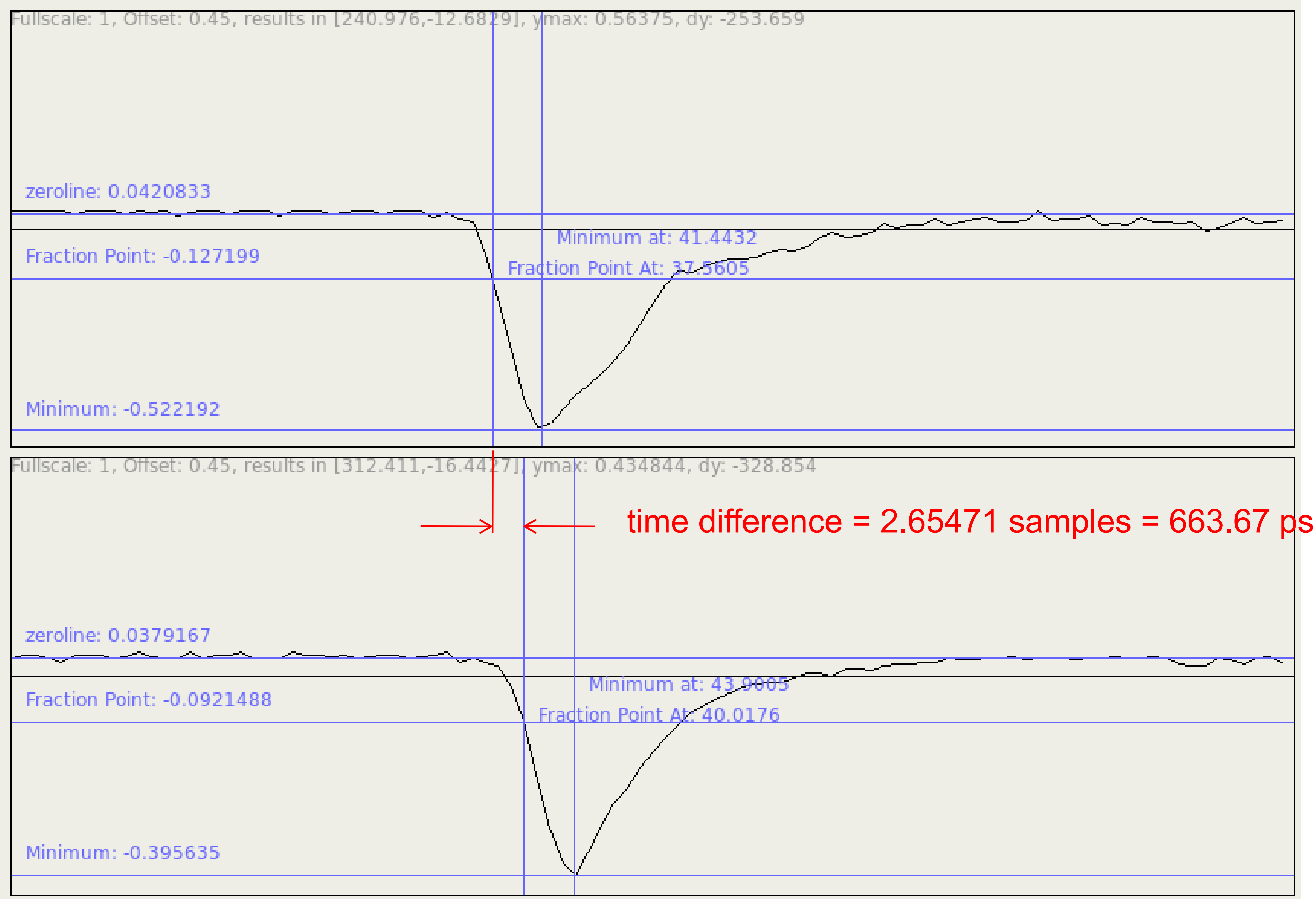
PMT: Hamamatsu H3378-50



Digitizer: Acqiris
8 GS/s & 3 GHz bandwidth

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

screenshot of two digitized anode pulses



Fullscale: 1, Offset: 0.45, results in [240.976,-12.6829], ymax: 0.56375, dy: -253.659

zeroline: 0.0420833

Fraction Point: -0.127199

Minimum at: 41.4432

Fraction Point At: 37.5605

Minimum: -0.522192

Fullscale: 1, Offset: 0.45, results in [312.411,-16.4427], ymax: 0.434844, dy: -328.854

zeroline: 0.0379167

Fraction Point: -0.0921488

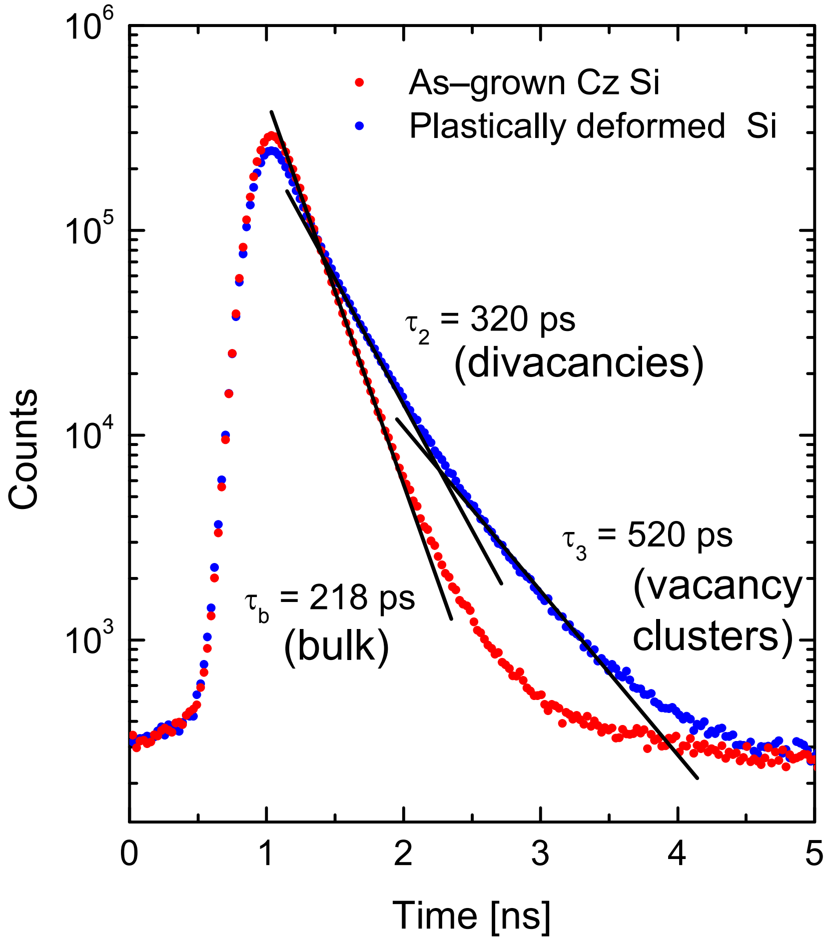
Minimum at: 43.9005

Fraction Point At: 40.0176

Minimum: -0.395635

time difference = 2.65471 samples = 663.67 ps

Positron lifetime spectroscopy



Time resolution is a Gaussian ≈ 230 ps (FWHM)

- 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 τ_i and intensities I_i

positron lifetime spectrum:

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

trapping coefficient

$$\boxed{\kappa_{\mathrm{d}} = \mu C_{\mathrm{d}}} = \frac{I_2}{I_1}\left(\frac{1}{\tau_{\mathrm{b}}} - \frac{1}{\tau_{\mathrm{d}}}\right)$$

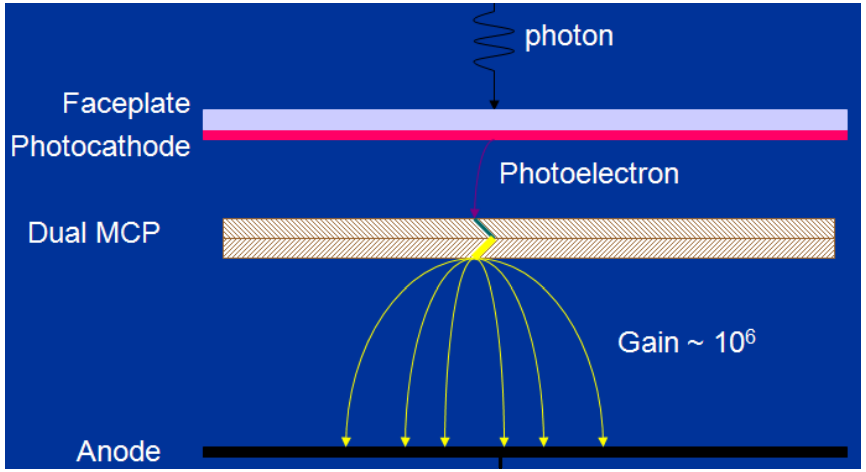
trapping rate

defect concentration

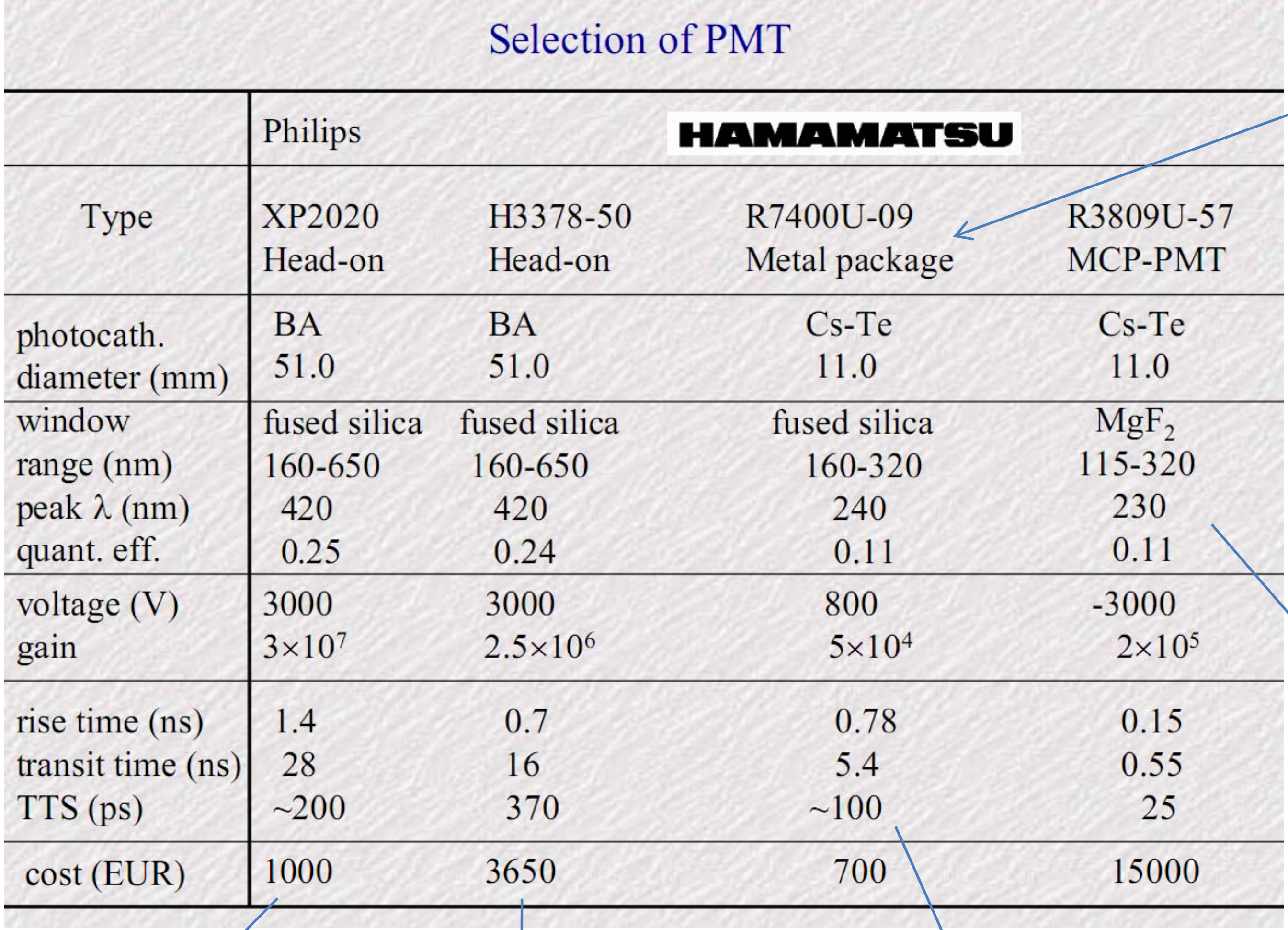
Improvement of time resolution

- 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)



Selection of PMT

	Philips	HAMAMATSU		
Type	XP2020 Head-on	H3378-50 Head-on	R7400U-09 Metal package	R3809U-57 MCP-PMT
photocath.	BA	BA	Cs-Te	Cs-Te
diameter (mm)	51.0	51.0	11.0	11.0
window	fused silica	fused silica	fused silica	MgF$_2$
range (nm)	160-650	160-650	160-320	115-320
peak λ (nm)	420	420	240	230
quant. eff.	0.25	0.24	0.11	0.11
voltage (V)	3000	3000	800	-3000
gain	3×10^7	2.5×10^6	5×10^4	2×10^5
rise time (ns)	1.4	0.7	0.78	0.15
transit time (ns)	28	16	5.4	0.55
TTS (ps)	~200	370	~100	25
cost (EUR)	1000	3650	700	15000

very promising, extremely fast, not many experiences, very expensive

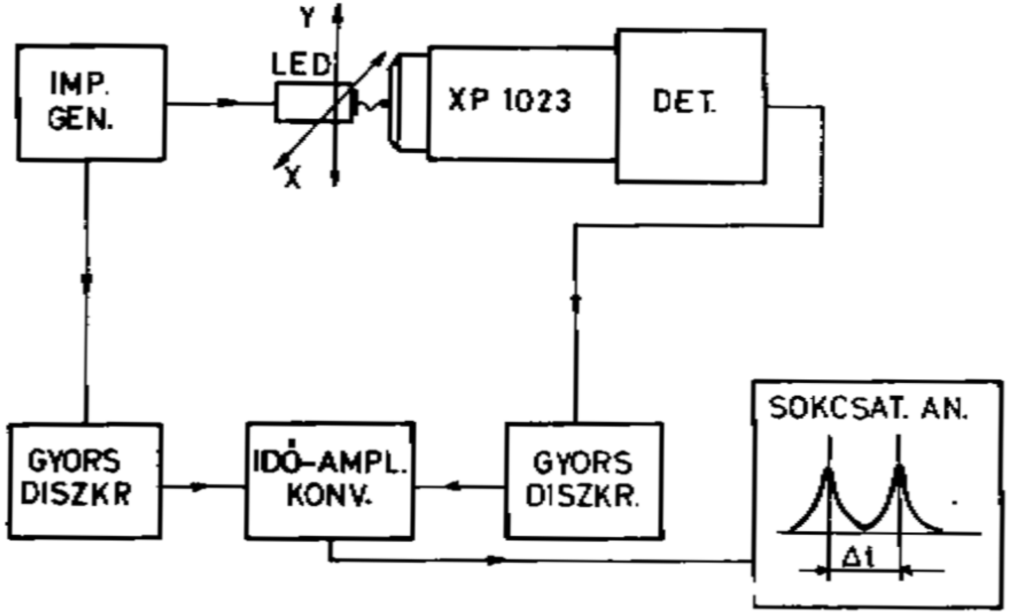
old tube, reliable, but rather slow

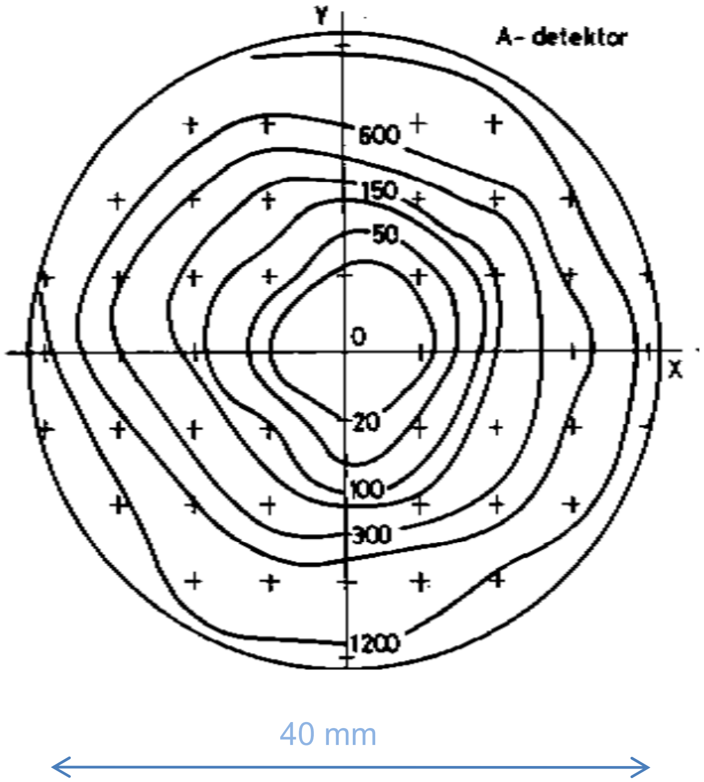
standard tube, large, efficient, pretty fast

very fast, but too small, not efficient enough

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)

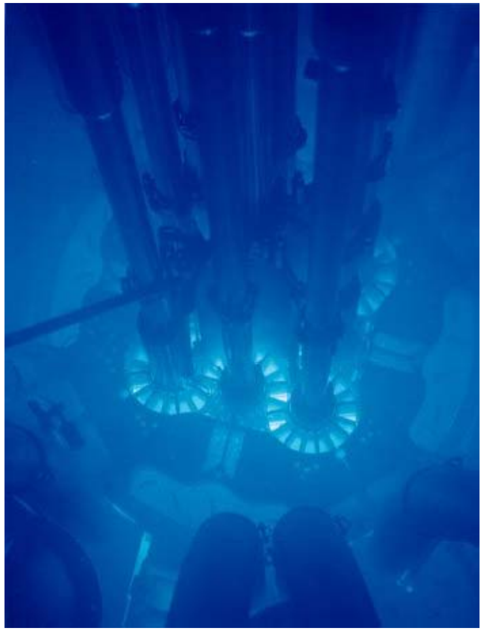




picture from: Sz. Kajsos, PhD Thesis, Budapest 1983

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

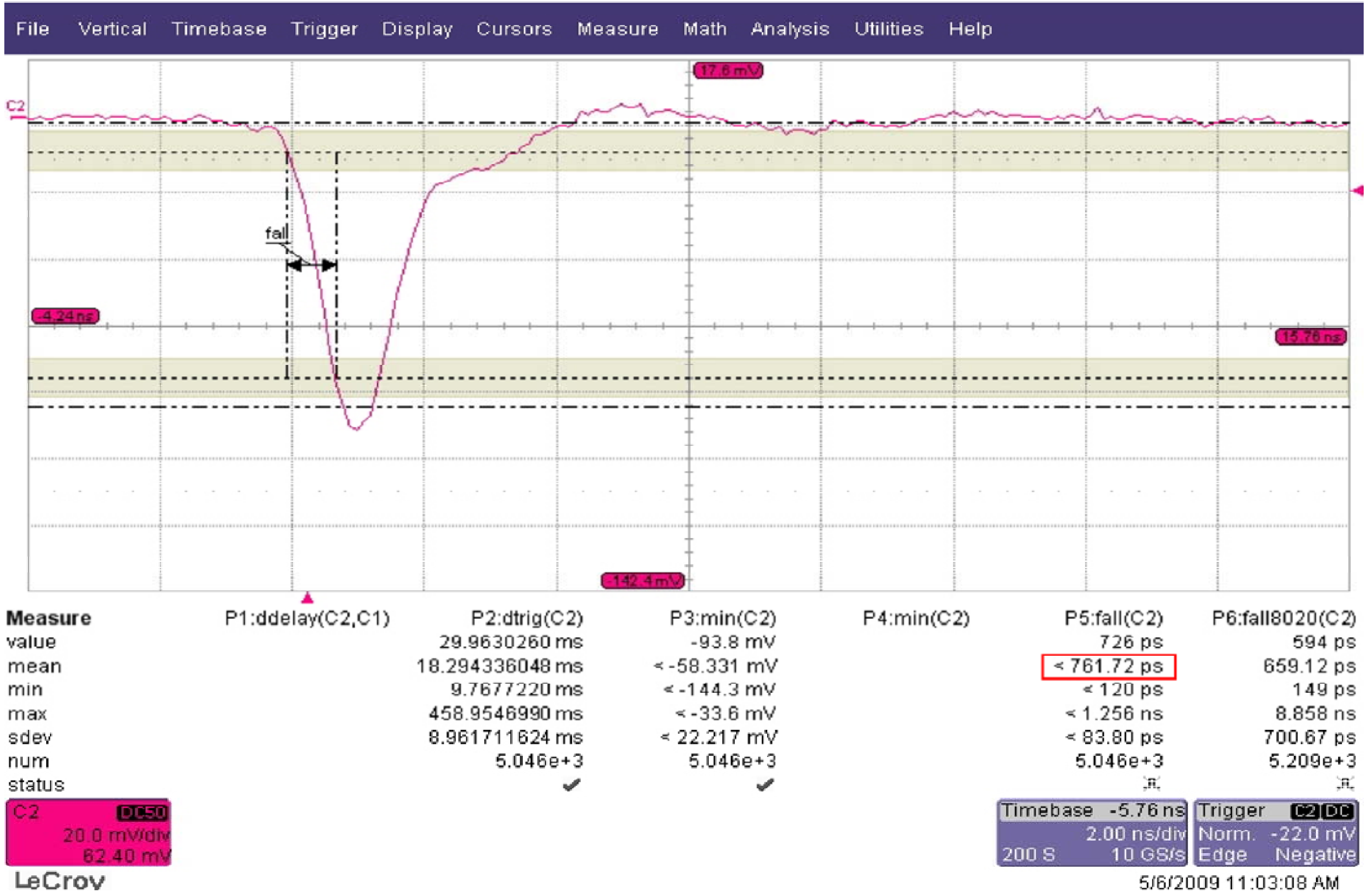


Cherenkov radiation emitted

from reactor core

- 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!

MCP–PMT −2.2 kV, Co–60 shooting on pexiglas crystal



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Measure   P1:ddelay(C2,C1)  P2:dtrig(C2)      P3:min(C2)    P4:min(C2)  P5:fall(C2)   P6:fall8020(C2)
value                       29.9630260 ms     -93.8 mV                  726 ps        594 ps
mean                        18.294336048 ms   < -58.331 mV              < 761.72 ps   659.12 ps
min                         9.7677220 ms      < -144.3 mV               < 120 ps      149 ps
max                         458.9546990 ms    < -33.6 mV                < 1.256 ns    8.858 ns
sdev                        8.961711624 ms    < 22.217 mV               < 83.80 ps    700.67 ps
num                         5.046e+3          5.046e+3                  5.046e+3      5.209e+3
status                      ✓                 ✓

C2         DC50                                              Timebase  -5.76 ns   Trigger  C2 DC
     20.0 mV/div                                                     2.00 ns/div  Norm.   -22.0 mV
       62.40 mV                                              200 S      10 GS/s   Edge    Negative
                                                             5/6/2009 11:03:08 AM
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Overview: scintillator materials for fast timing

Scintillator	**Wavelength [nm]**	**Density [g/ccm]**	**Luminosity 10^3 photons/MeV**	**Energy Resolution % FWHM @ 662keV**	**Rise time (10 - 90%) [ns]**		**Measured Timing Resolution [ps]**
					literature	measured	
BaF2	220 310	4.88	2 10	11.4 7.9	0.6 – 1 300 – 600 decay time	0.9	160
LSO	420	7.4	27 – 33		40		275
LaBr3(Ce)	356 – 387	5.1	60 – 75	2.7 – 3.2	30	5,5	346
ZnO:Ga	385	5.7			0.36; 0.82		
ZnO						0.75	370
Plastic (BC-422)	380	1.032			0.35	2	325
NaI (for reference)	415	3.67	44	5.6 – 7.1	50 ns		

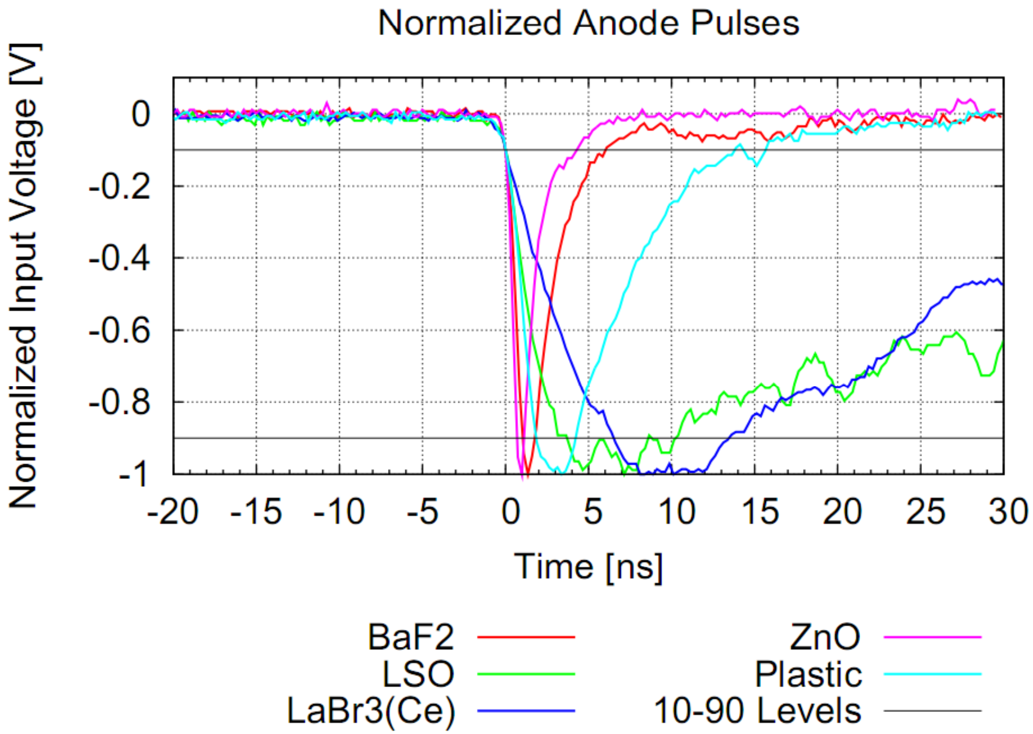
[1] scintillator.lbl.gov

[2] www.detectors.saint-gobain.com

Rise times are measured using Hamamatsu photomultipliers H3378-50

Timing of Scintillators

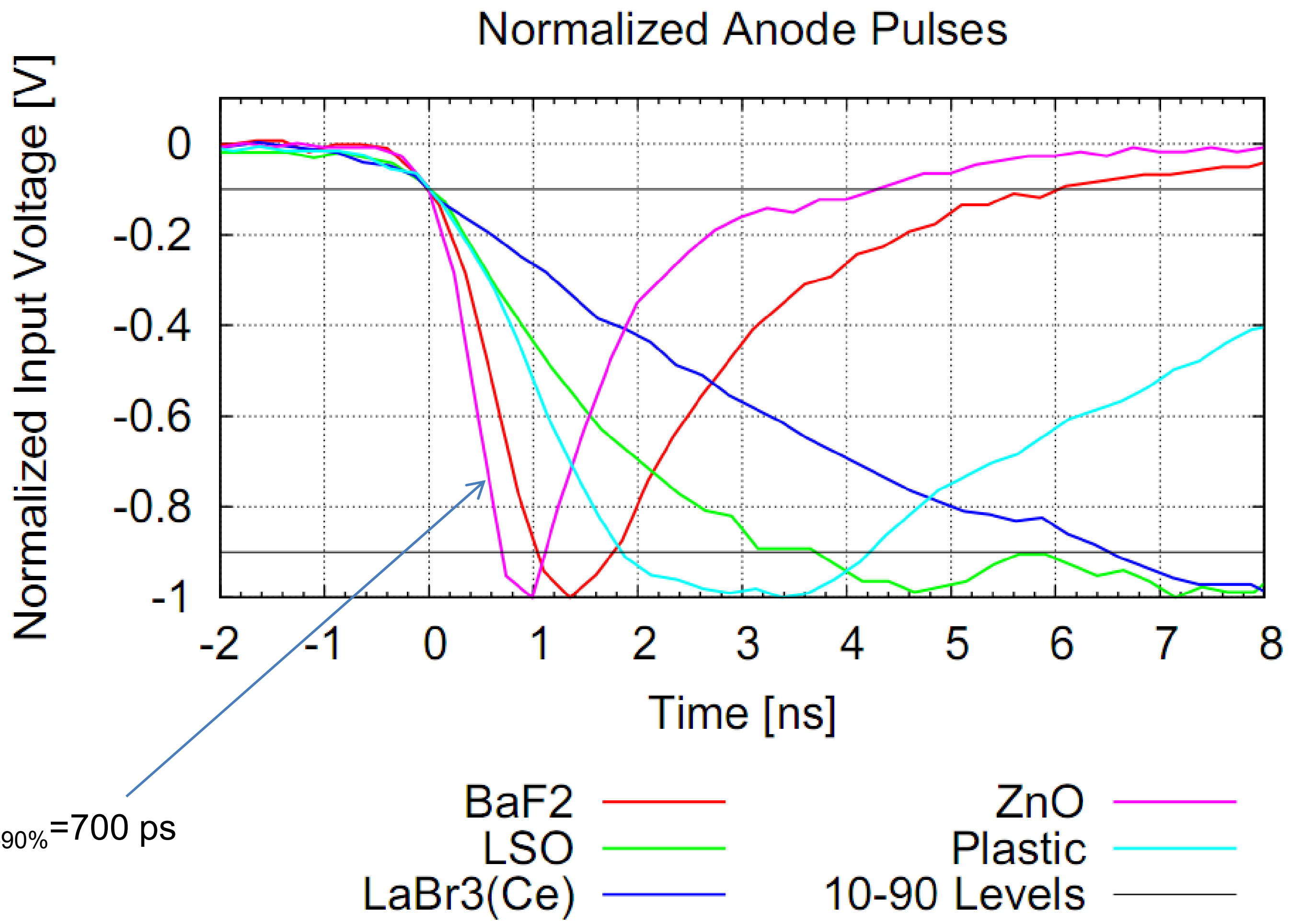
- demand: high-z for high efficiency and short rise time
- **LaBr3(Ce)** is very efficient but very slow
- **LSO (Lu_2SiO_5)**: rather heavy; very slow decay of scintillation light; not hygroscopic
- organic **plastic**: relatively fast; no gamma backscattering; too inefficient
- **BaF_2**: very fast rise and decay; slow component 310 nm – 600ns; hygroscopic
- **ZnO**: extremely fast rise time and decay – most promising



- 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

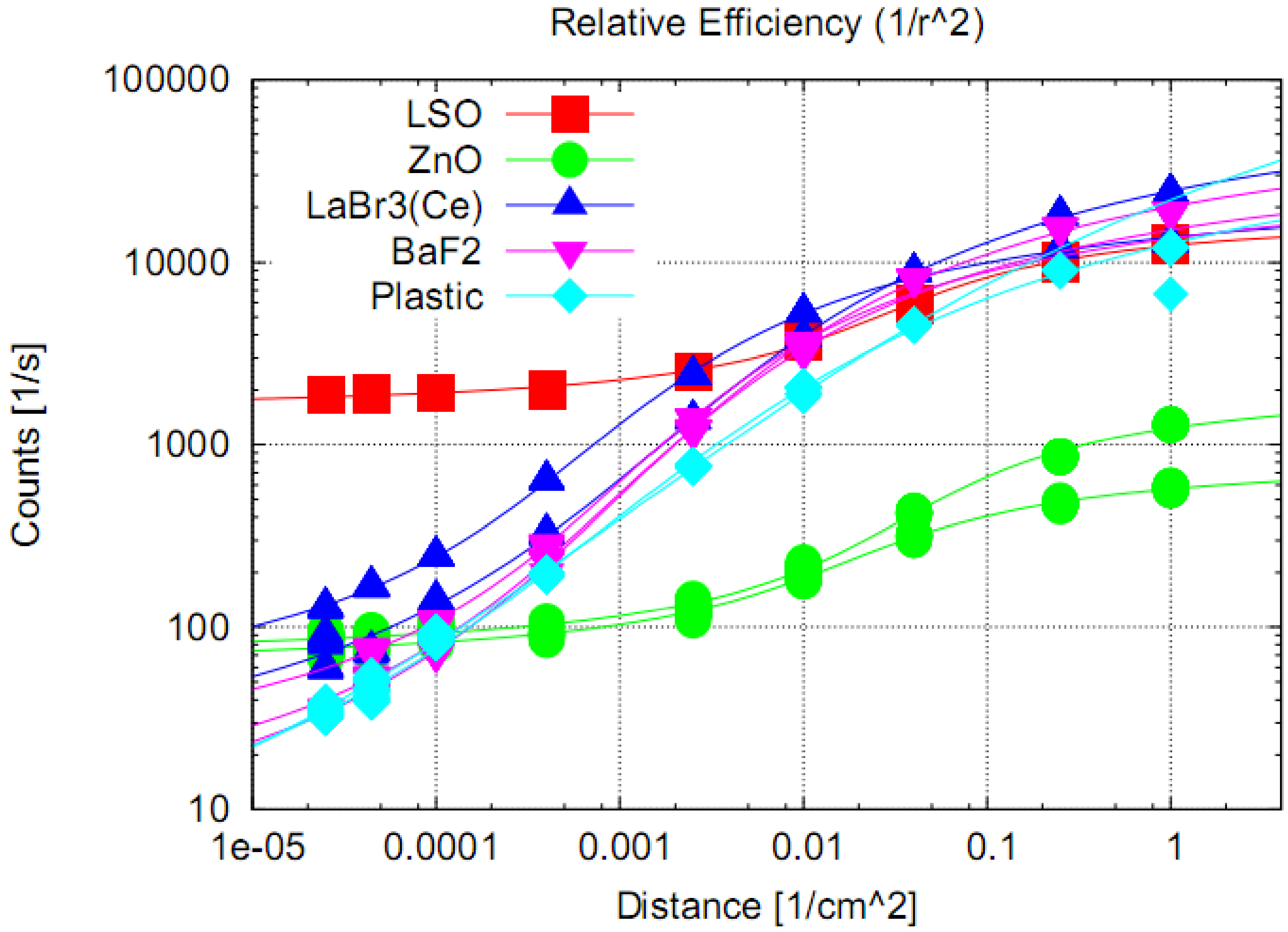
- ZnO clearly faster than BaF_2
- real rise and decay time not measurable with our setup
- problem up to now: no large single crystals available



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



ZnO as Scintillator

- 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

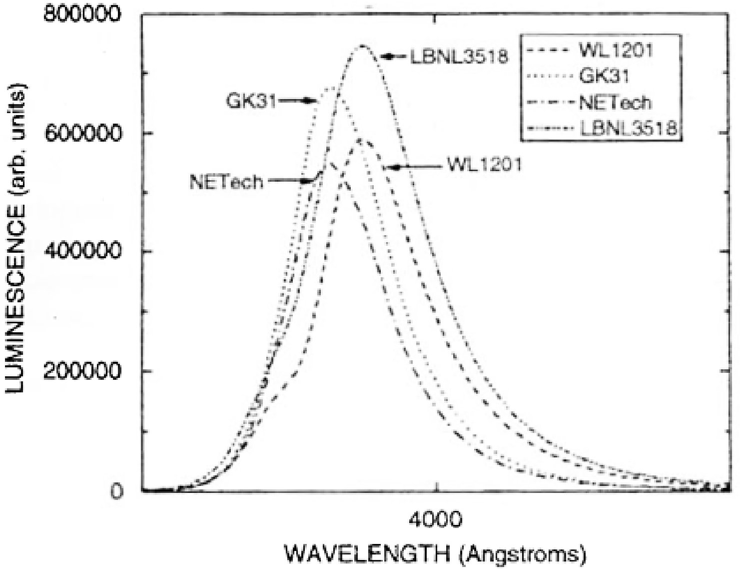


Fig. 4. Photoluminescence spectra for the four powder samples at 300 K. The spectra predominantly consist of fast, near-band-edge (NBE) emissions. (a) The vertical scale is logarithmic. (b) Enhanced view of the NBE region at 300 K. The PL peak positions of the GK31 and NETech powders are similar and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

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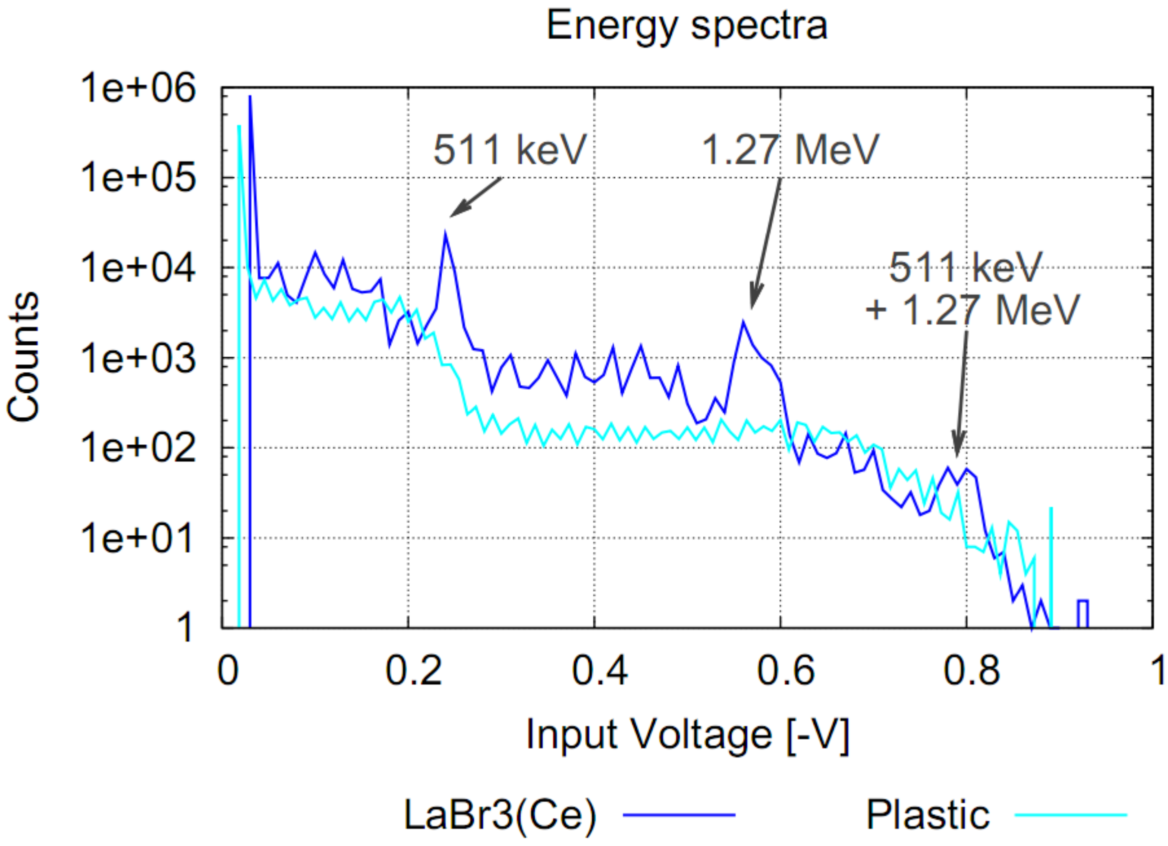
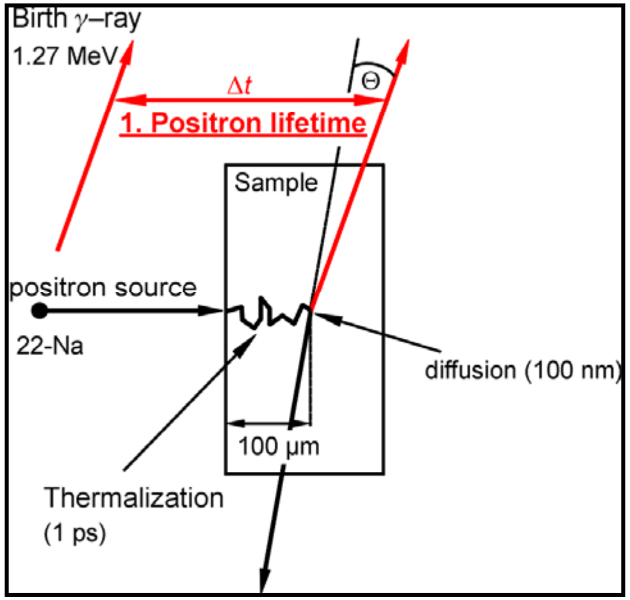
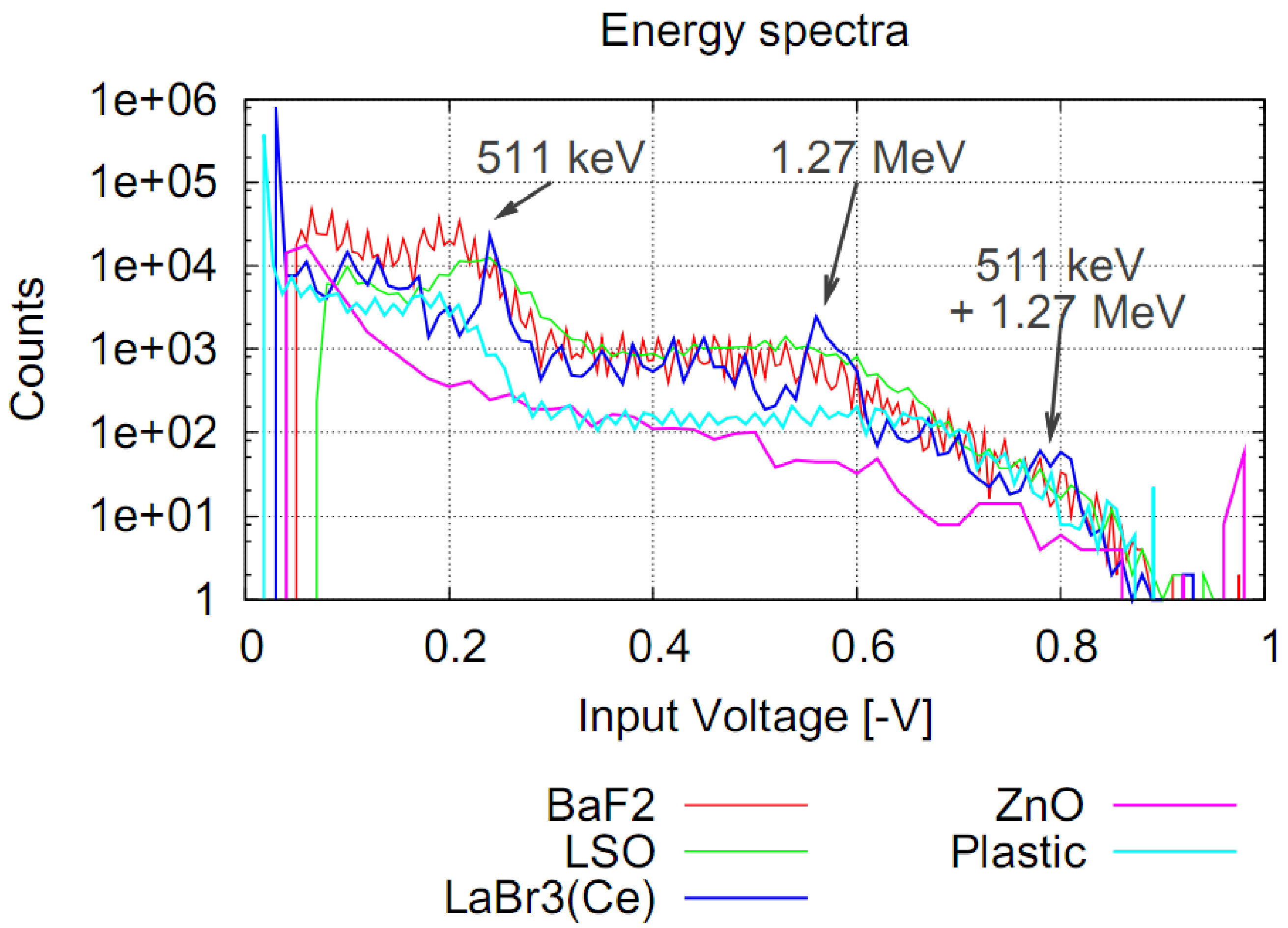
Energy resolution required

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



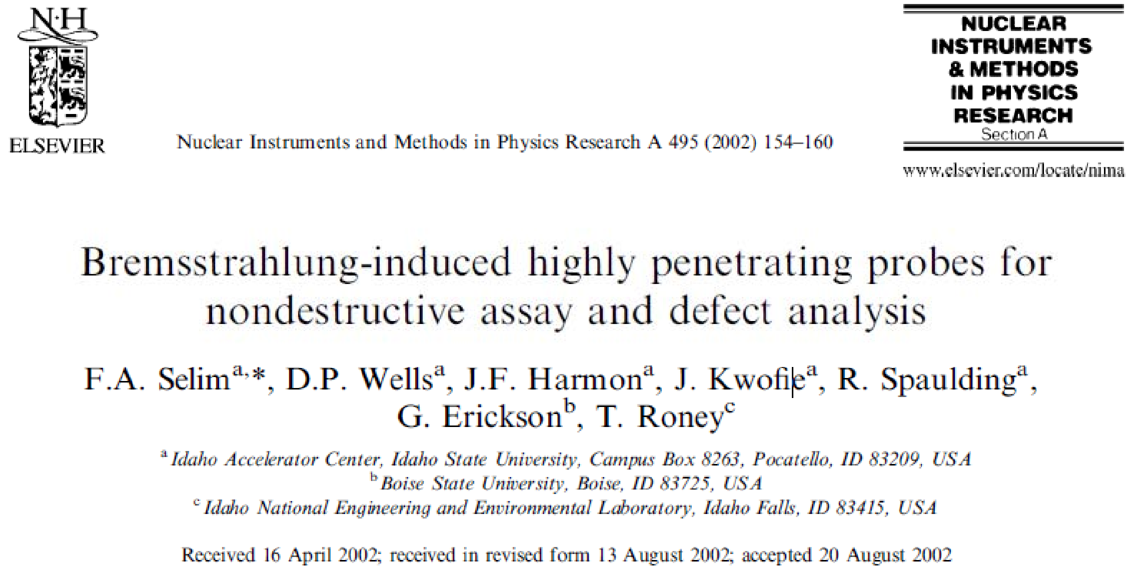


both materials are okay, although the photo peak is completely missing in plastics



- 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)?

Gamma-induced Positron Spectroscopy



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NUCLEAR INSTRUMENTS & METHODS IN PHYSICS RESEARCH
Section A

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Bremsstrahlung-induced highly penetrating probes for nondestructive assay and defect analysis

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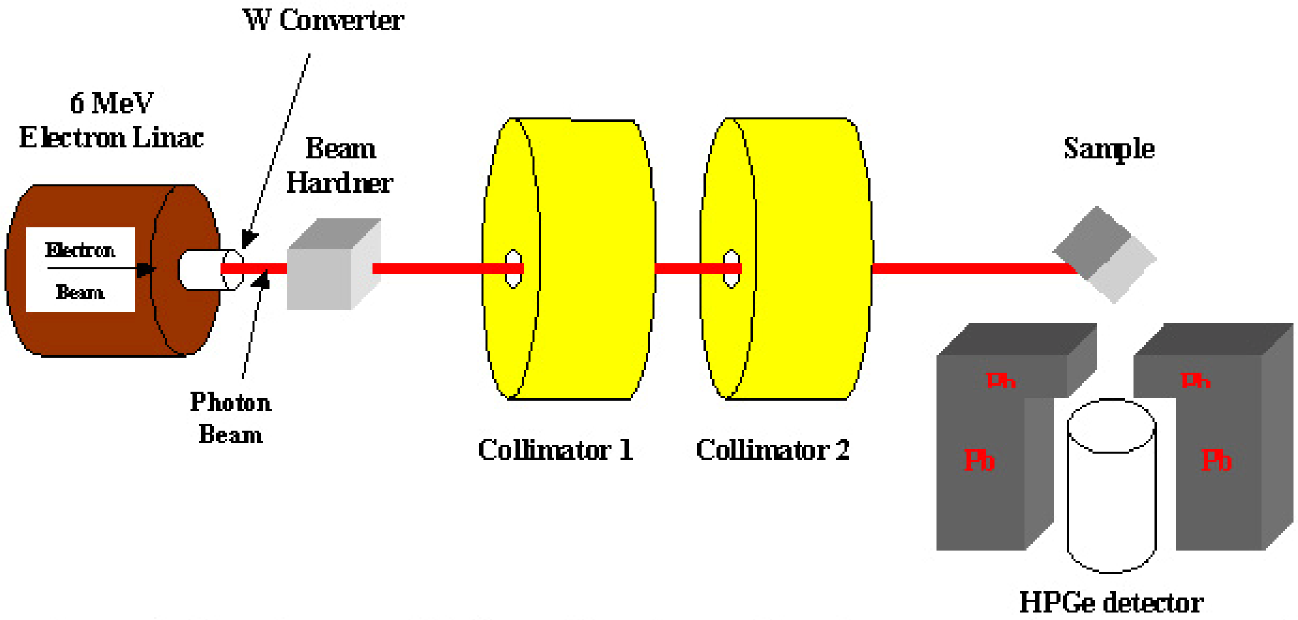
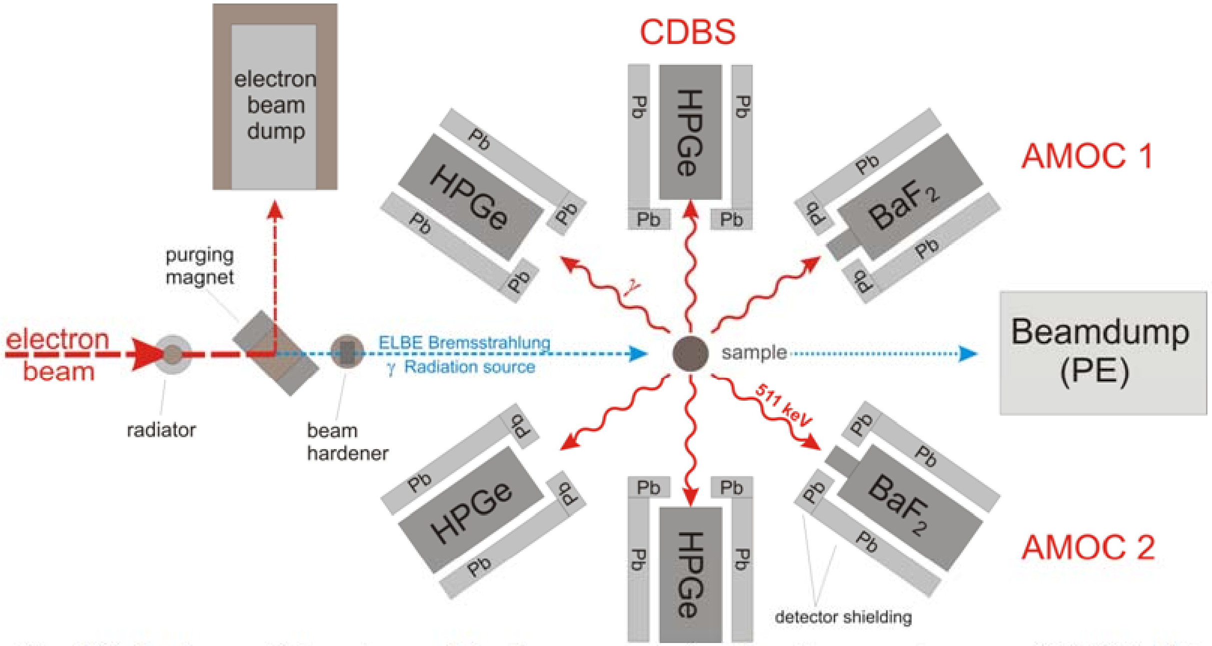
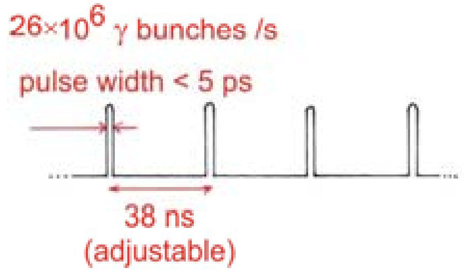


GiPS: Gamma-induced Positron Spectroscopy

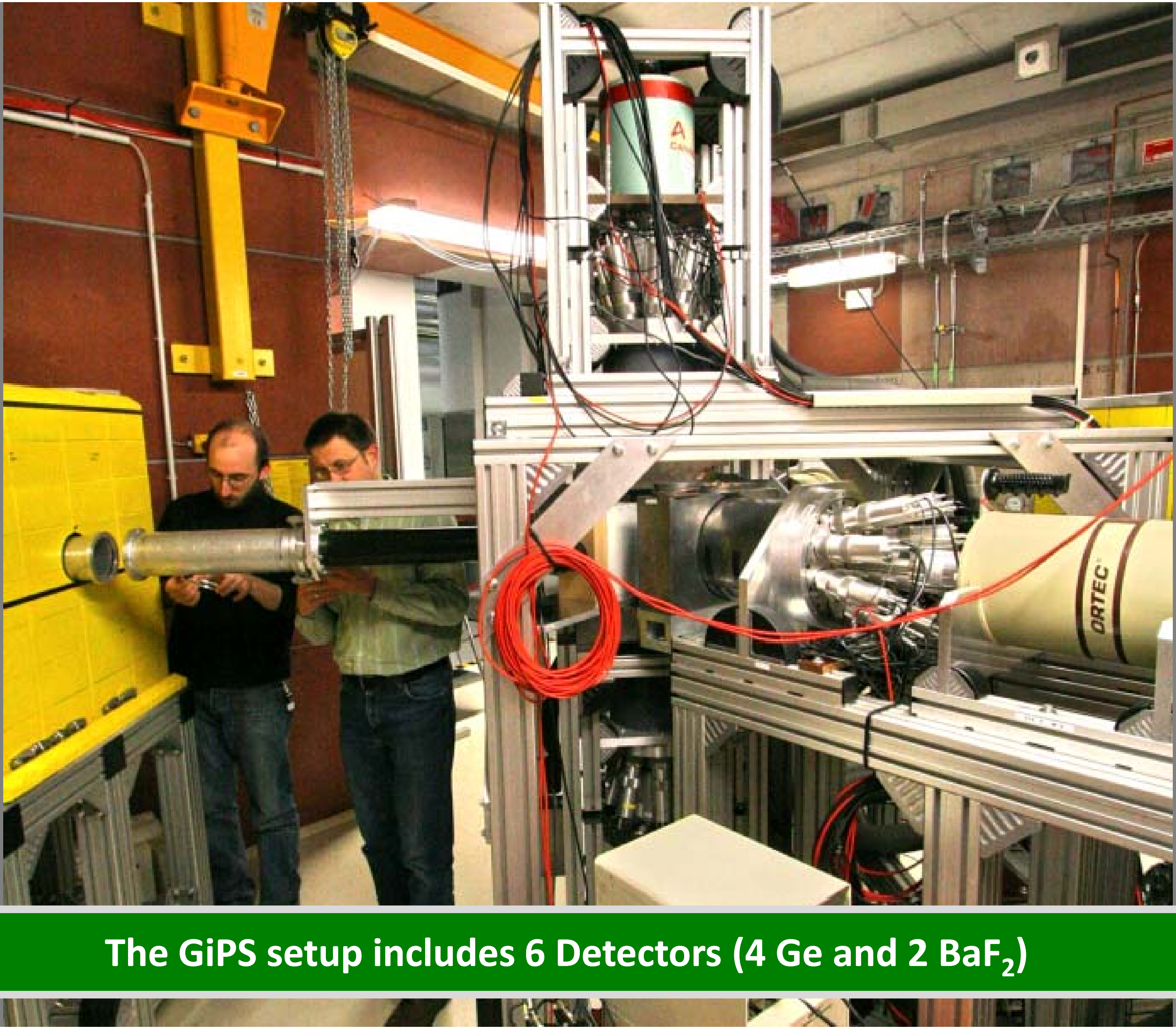


Simplified scheme of the setup used for the gamma induced positron spectroscopy (GiPS) facility at EPOS



AMOC: Age-Momentum Correlation
CDBS : Concidence Doppler-Broadening Spectroscopy

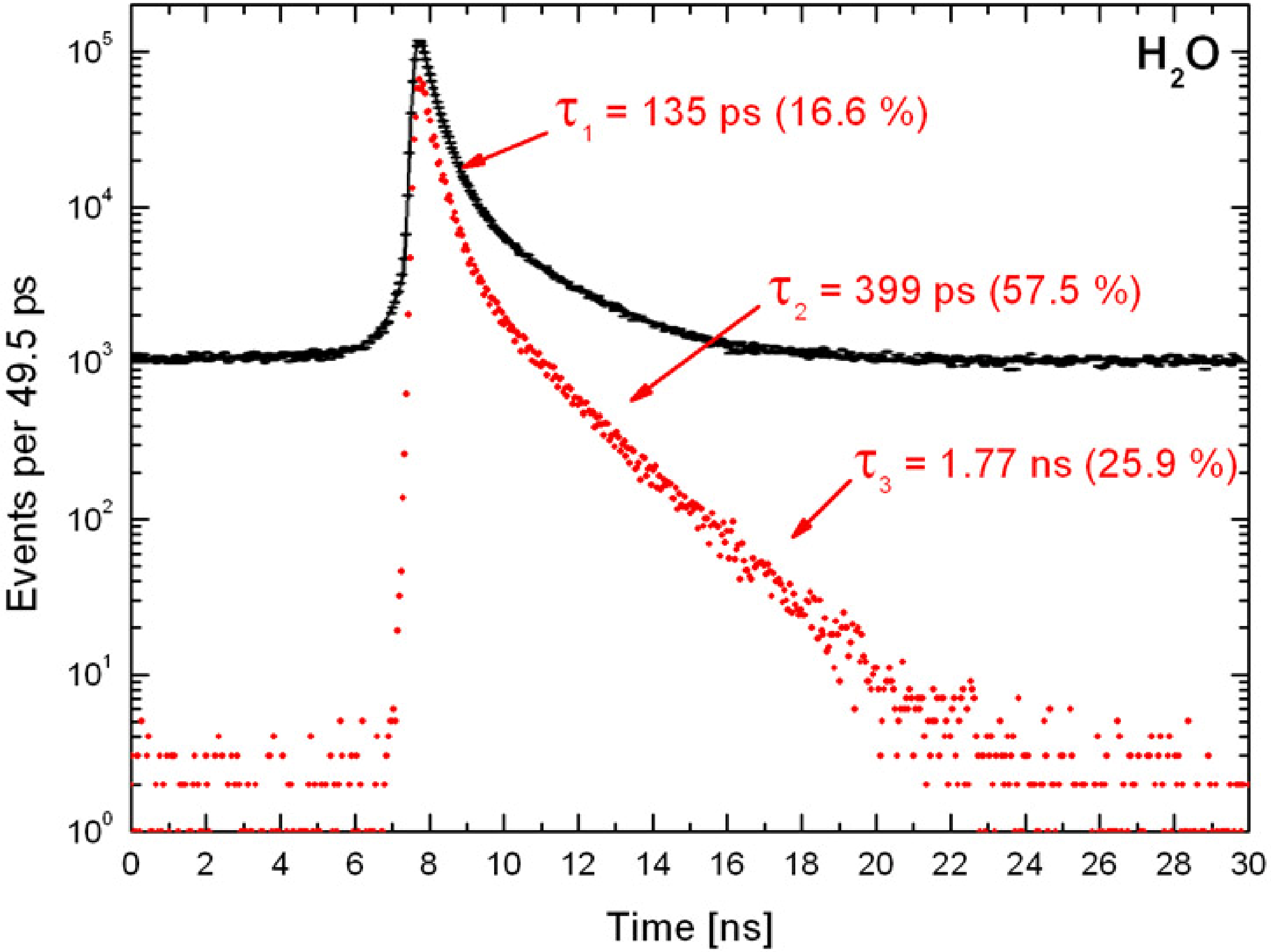
- background reduction due to coincident measurement
- lifetime measurement requires only detection of annihilation radiation of 511 keV
- thus, ZnO can be used here
- we obtained 180 ps FWHM time resolution: hope for < 100 ps



The GiPS setup includes 6 Detectors (4 Ge and 2 BaF_2)

Water spectrum: GiPS spectrum compared with conventional measurement

- in black: conventional measurement with ^{22}Na source (K. Kotera, T. Saito, T. Yamanaka, Phys. Lett. A 345, 184-190 (2005)
- advantage of periodic positron source is obvious: background distinctly reduced for GiPS spectrum (red)
- spectra quality distinctly improved (however resolution still ≈ 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
- BaF_2 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 (BaF_2 < 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

Forschungszentrum
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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/