

# Positron-Annihilation Lifetime Spectroscopy using Electron Bremsstrahlung

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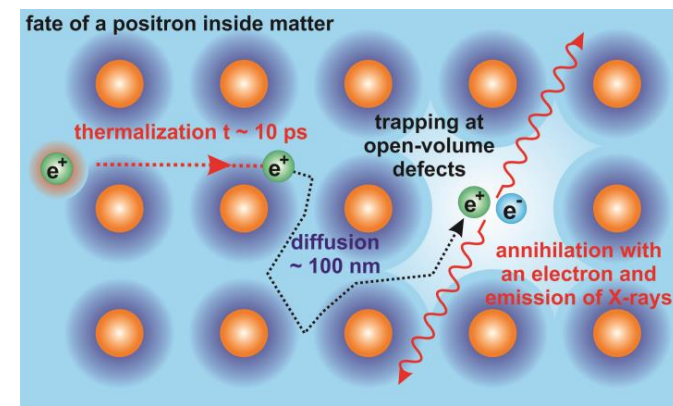
**hzdr**



HELMHOLTZ  
ZENTRUM DRESDEN  
ROSSENDORF

# Outline

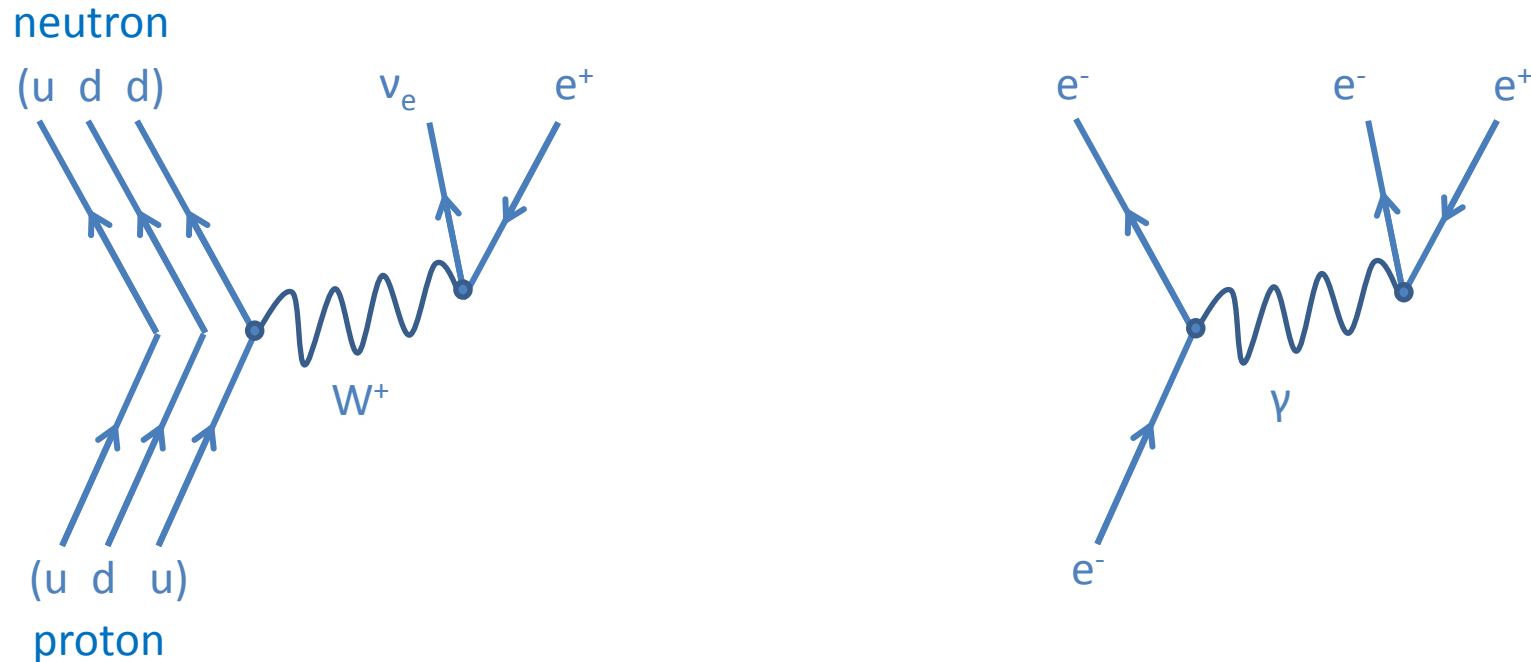
- Motivation
- Accelerator-based positron production and annihilation studies at a superconducting electron LINAC: What marks the difference to reactors and radio-isotope sources?
- Applying pulsed beams: positron annihilation lifetime spectroscopy at thin films, bulk materials, and fluids
- Development of a pixelated detection system for position-sensitive positron annihilation lifetime measurements and experiments with structured targets and tomographic image reconstruction



Courtesy: R. Krause-Rehberg / M. Butterling

# Isotopes, reactors, accelerators

Production of positrons in weak ( $W^+$ ) or electromagnetic interactions ( $\gamma$ )

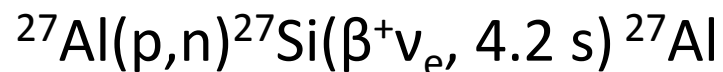
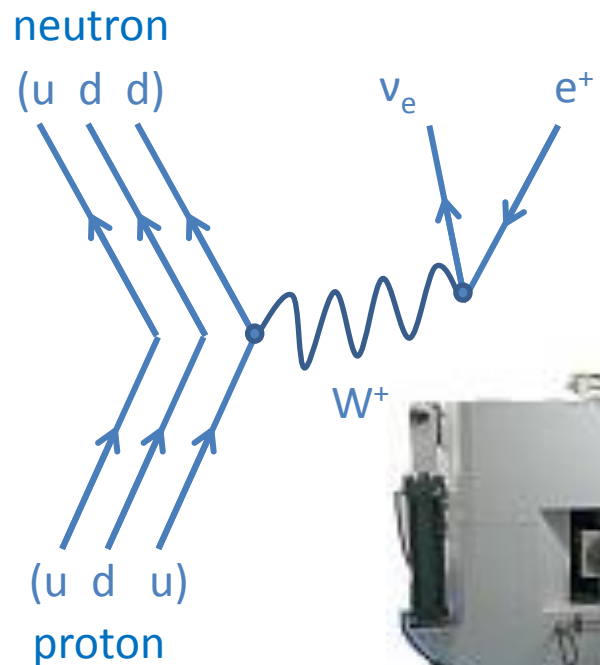


Free proton decay is forbidden by energy conservation

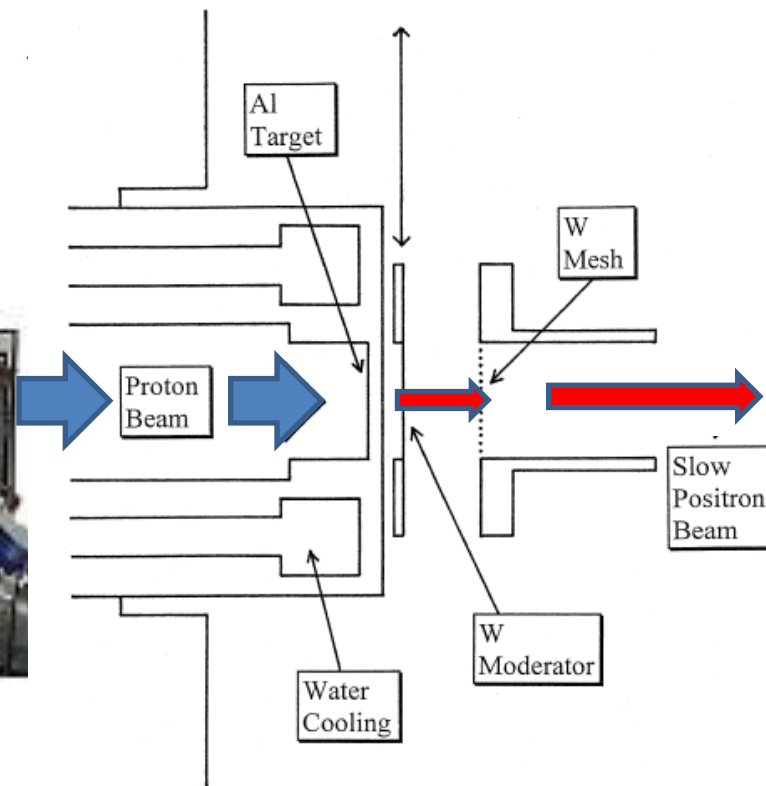
→ we need the proton inside a nucleus where it undergoes  $\beta^+$ -decay

# Isotopes, reactors, accelerators

Production of positrons in weak interactions (mediated by W's)



Sumitomi Heavy Industries Cyclotron  
18 MeV protons, 50  $\mu\text{A}$  beam current

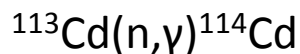


# Isotopes, reactors, accelerators

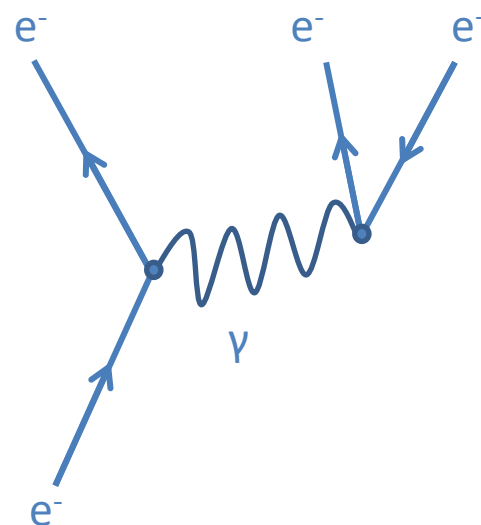
Production of positrons through electromagnetic interactions (photons)

Use intense source of photons for pair production

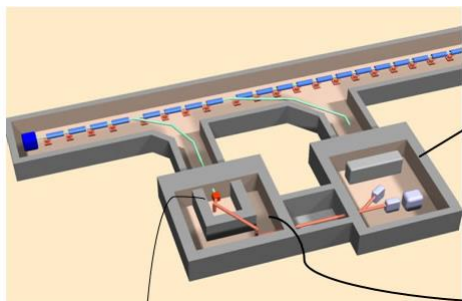
→ Capture-neutron gamma-rays from reactor



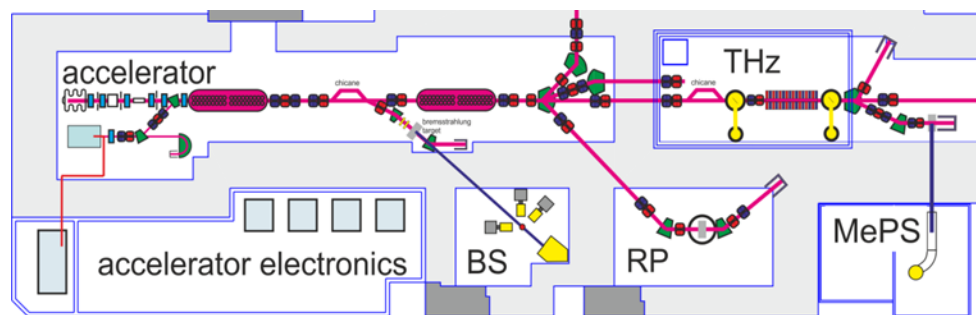
FRM II Munich



→ Bremsstrahlung from electron accelerators



AIST, Tsukuba, Japan



ELBE, Dresden

# Positrons from accelerators

Accelerators can produce intense and pulsed slow positron beams.  
LINear ACcelerators are favored due to their high beam power and time structure.

## A) normal conducting LINAC (AIST)

$E \sim 50 \text{ MeV}$

$I_{\text{peak}} \sim 100 \text{ mA}$

$t_{\text{bunch}} \sim 1 \mu\text{s}$

$f_{\text{rep}} \sim 100 \text{ Hz}$

beam power  
500 W

## B) superconducting LINAC (HZDR)

$E \sim 50 \text{ MeV}$

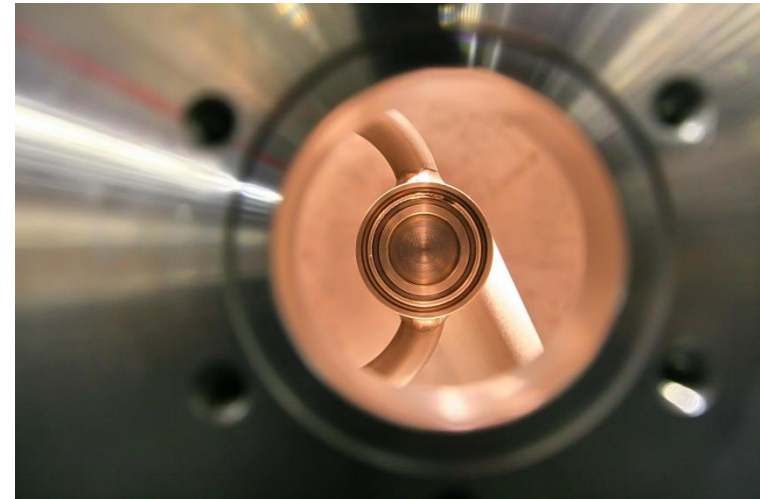
$I_{\text{average}} \sim 1 \text{ mA}$

$f_{\text{rep}} \sim 10 \text{ MHz}$

beam power  
50 kW

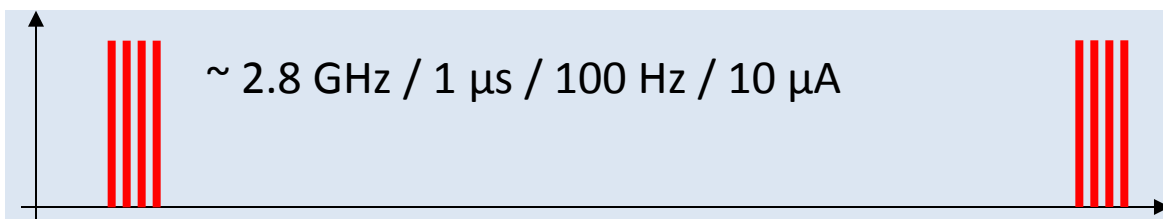
sophisticated converter designs  
and heavy shielding needed

stack of 50  
100  $\mu\text{m}$  thick W foils

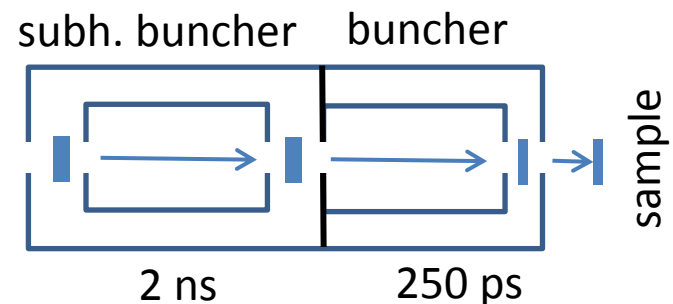
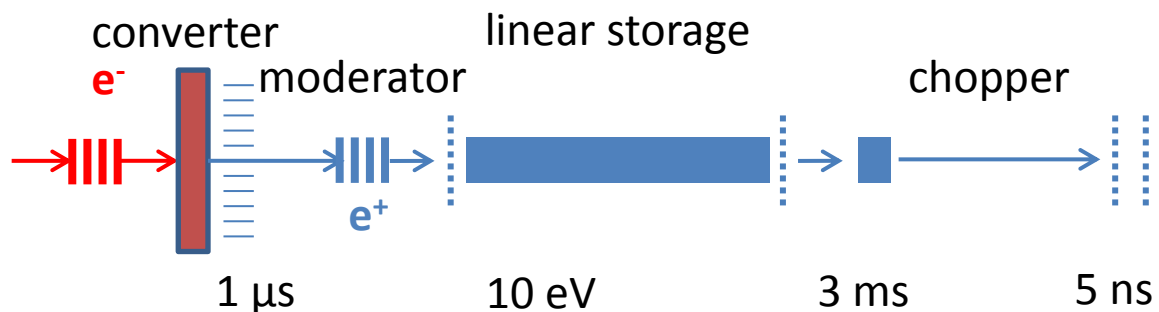


EPOS water-cooled  
converter

# Positrons from accelerators

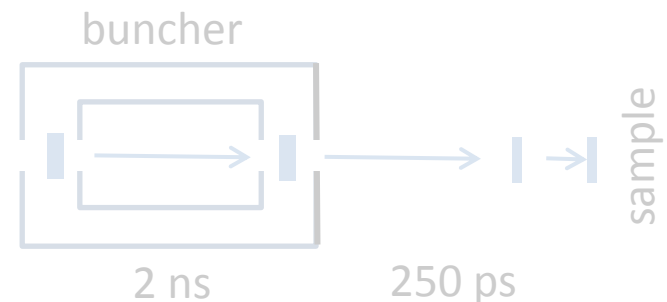
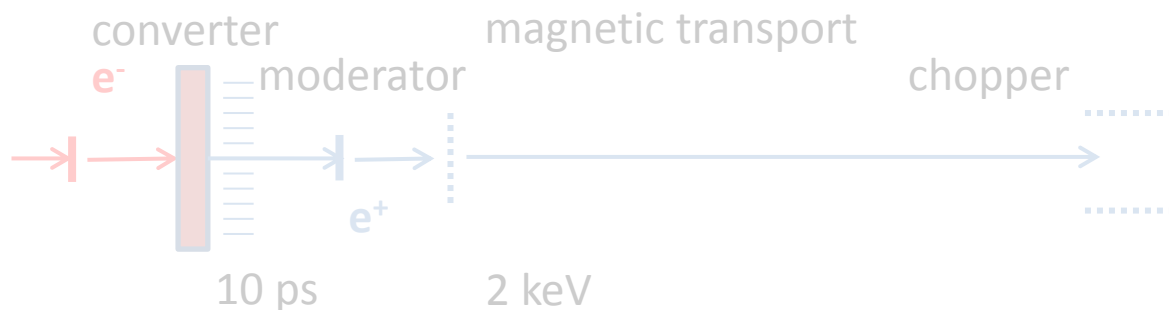


**NC-LINAC** in bunched mode



**SC-LINAC** in CW mode

EPOS facility



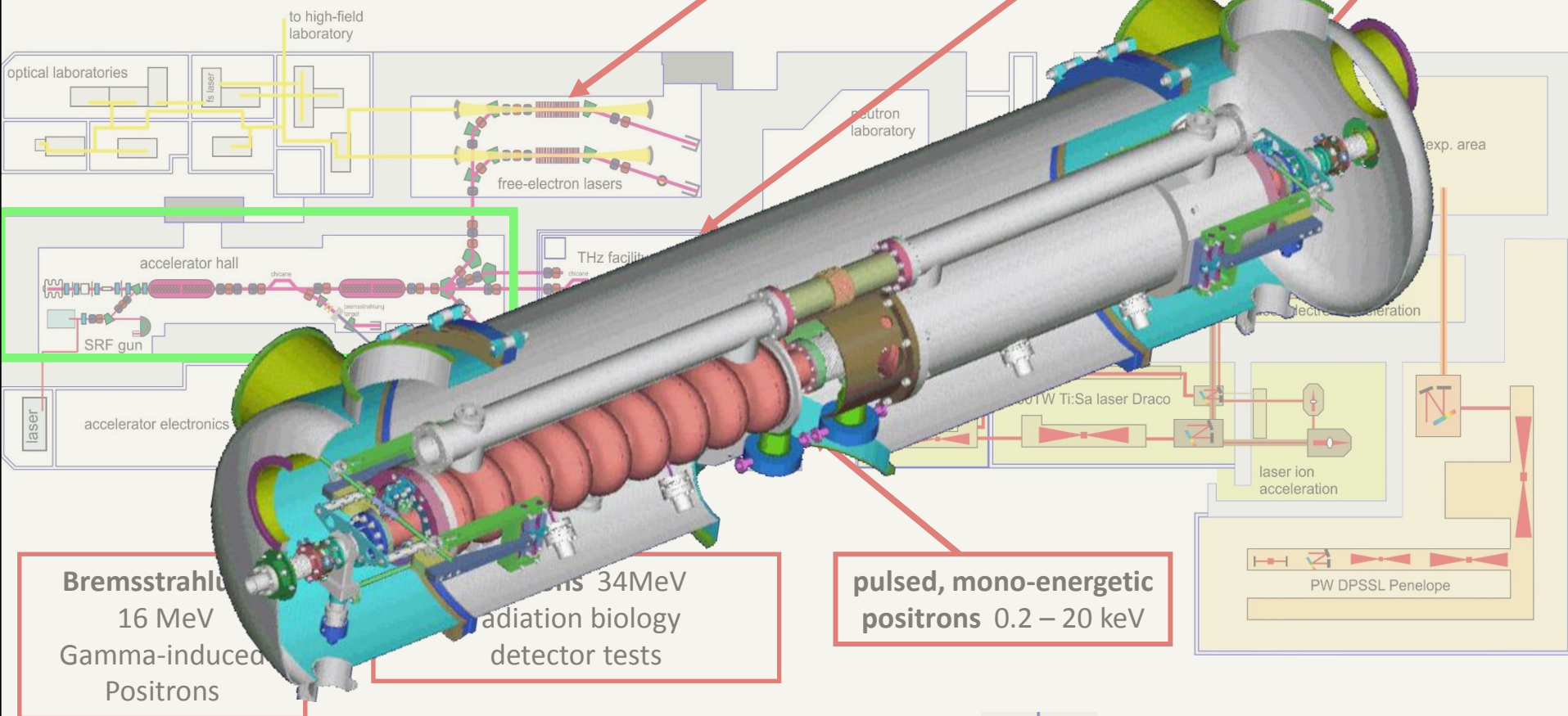
# Positrons from accelerators

1.6 mA, 40 MeV (64 kW)  
CW electron accelerator

coherent IR-radiation  
3 – 230  $\mu\text{m}$

THz radiation  
100  $\mu\text{m}$  – 3 mm

neutron time of flight  
 $E_n$  0 – 10 MeV

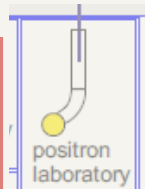


Bremsstrahlung  
16 MeV  
Gamma-induced  
Positrons

34 MeV  
radiation biology  
detector tests

pulsed, mono-energetic  
positrons 0.2 – 20 keV

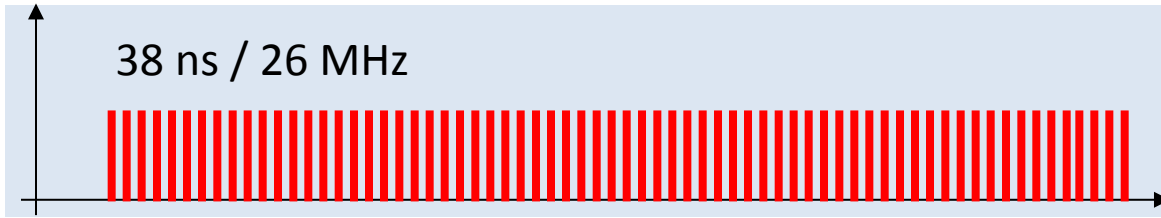
mono-energetic  
positrons 0.2 – 30 keV  
from  $^{22}\text{Na}$



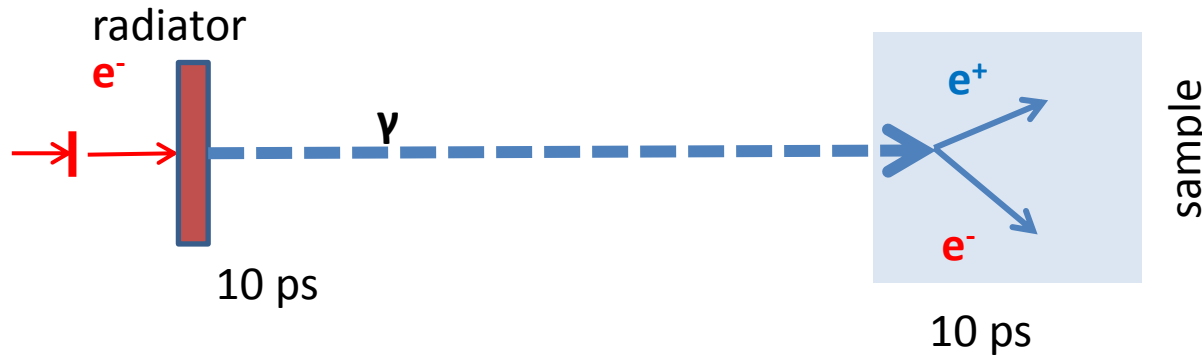
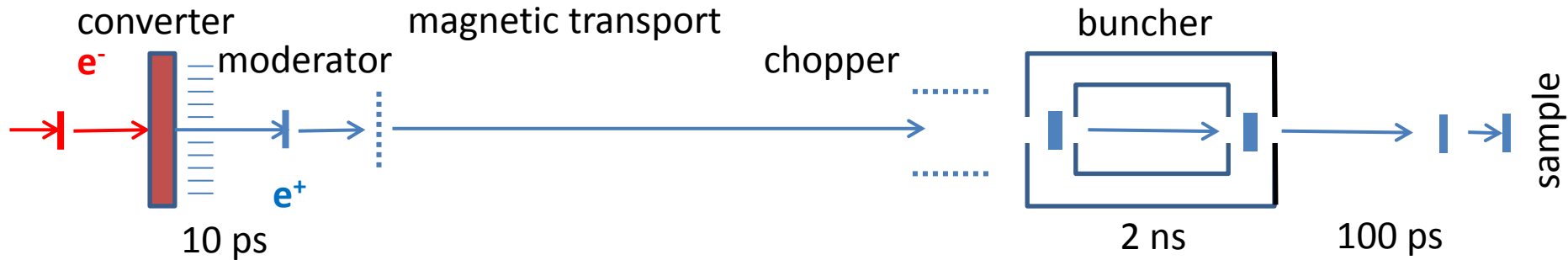
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# What about bulk materials, fluids, gases ...?



SC-LINAC in CW mode



**GiPS**

The **G**amma-induced  
**P**ositron annihilation  
**S**pectroscopy

# Positron production using electron-bremsstrahlung

M. Butterling, et al.,  
Nucl. Instr. Meth. B 269 (2011) 2623

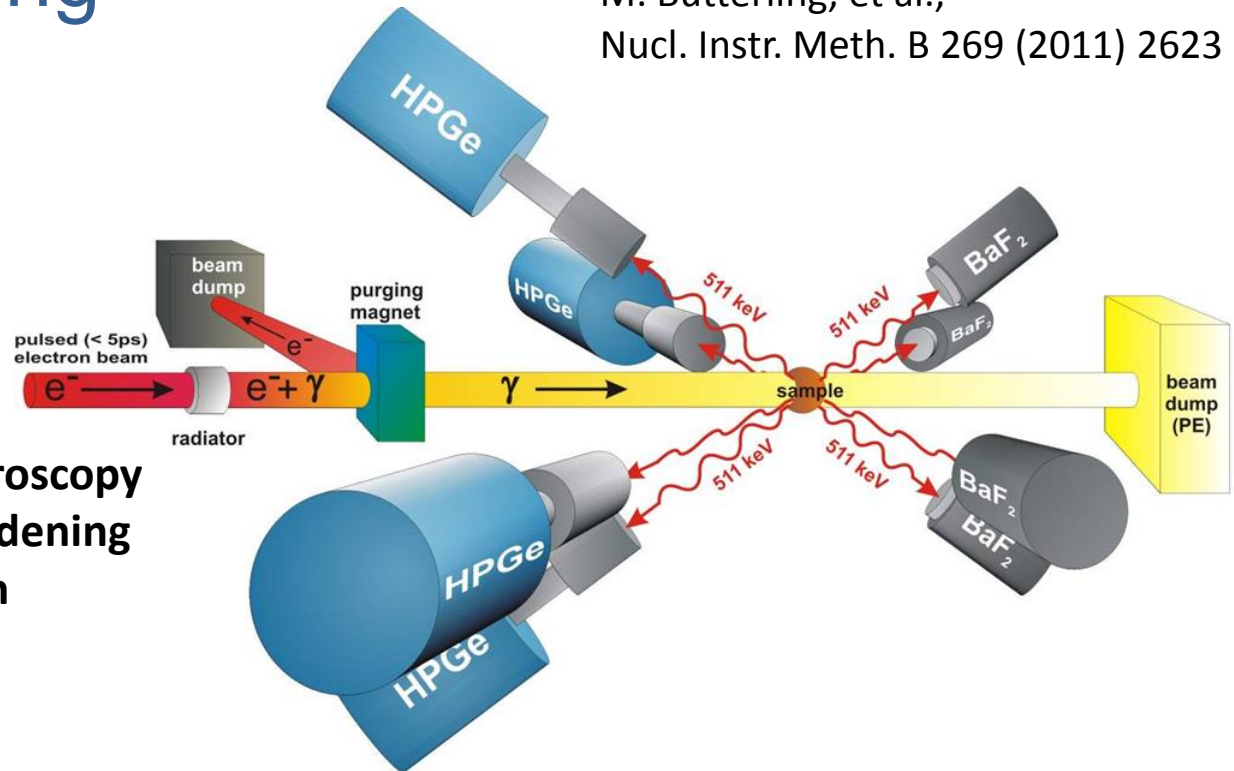
$$E_e = 16 \text{ MeV}$$

$$I_e = 900 \mu\text{A}$$

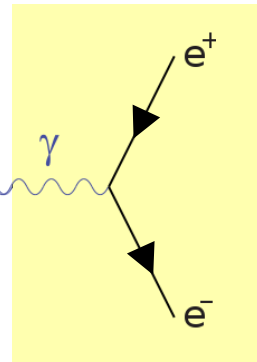
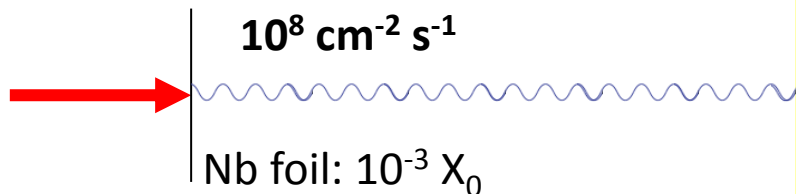
$$f = 26 \text{ MHz}$$

$$\sigma_t < 10 \text{ ps}$$

Annihilation Lifetime Spectroscopy  
(Coincidence) Doppler Broadening  
Age-momentum Correlation



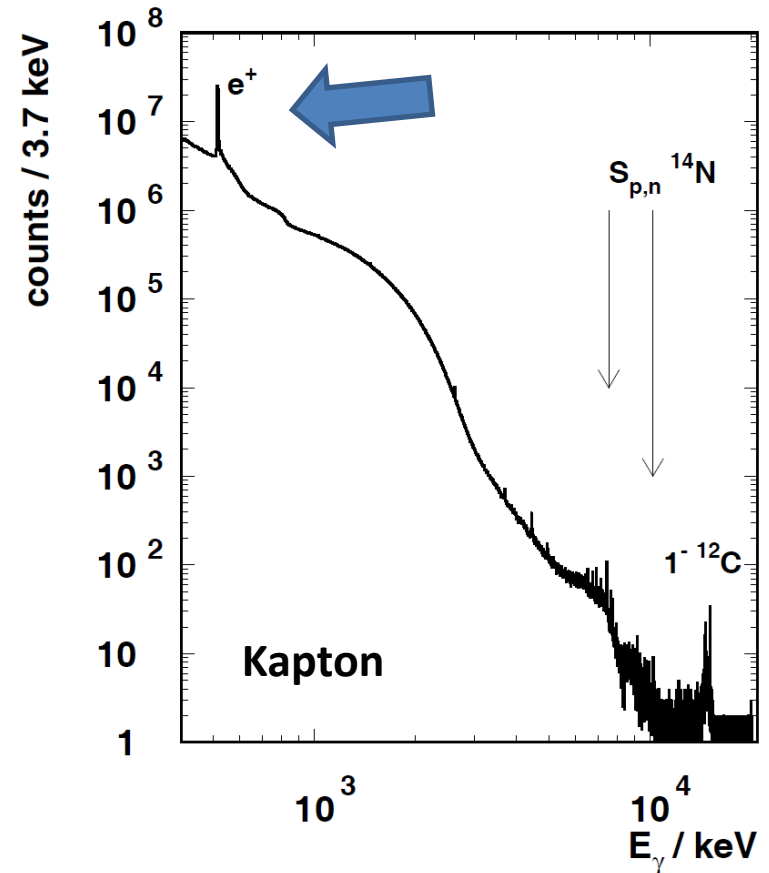
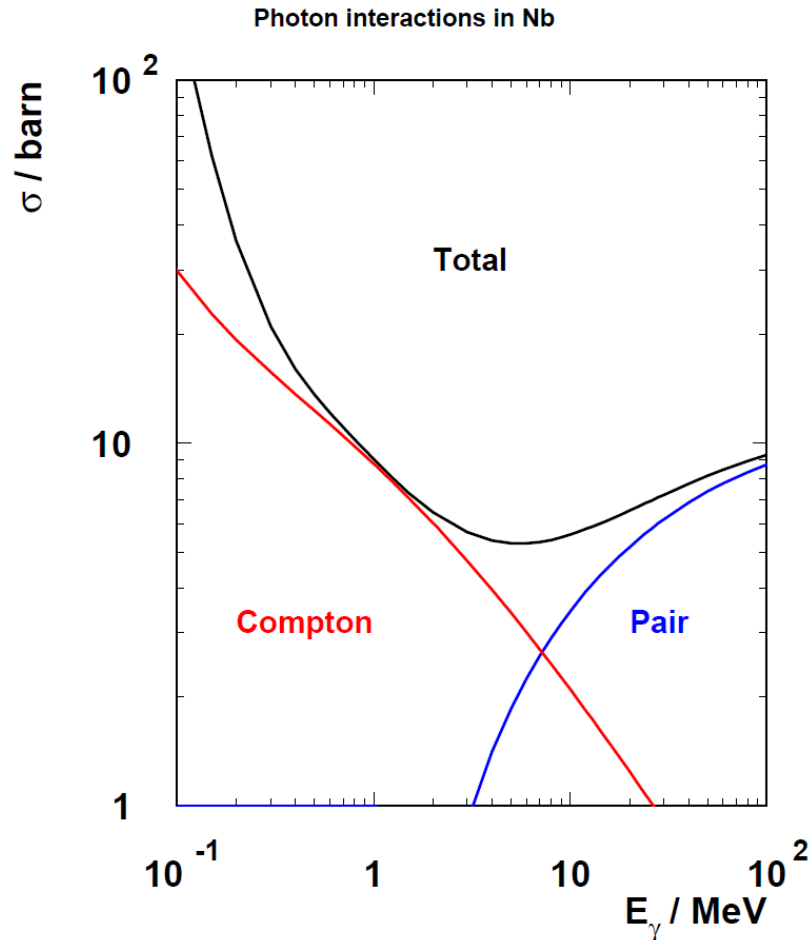
photon beam  
2 cm diameter  
 $10^8 \text{ cm}^{-2} \text{ s}^{-1}$



studies done so far:

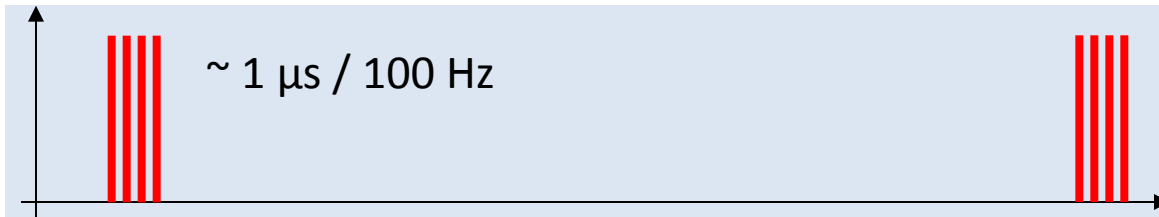
- water, glycerol from  $10^\circ\text{C}$  to  $100^\circ\text{C}$
- animal tissue
- metals and alloys
- **neutron-activated reactor materials**

# Positrons: backgnd for nuclear physics exp'ts

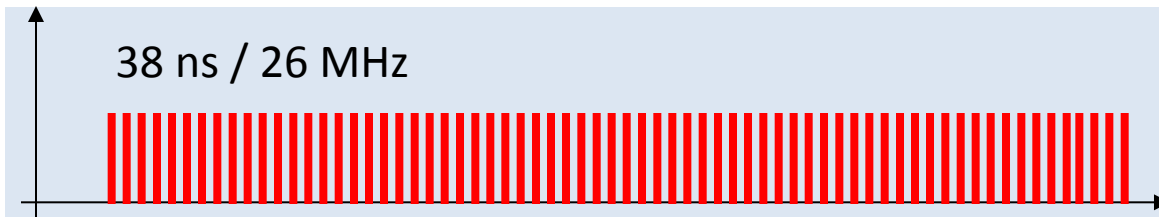


Hard bremsstrahlung produces a huge amount of positrons via pair production inside the target material. High-energy photons act as a **volume source of positrons throughout the entire volume.**

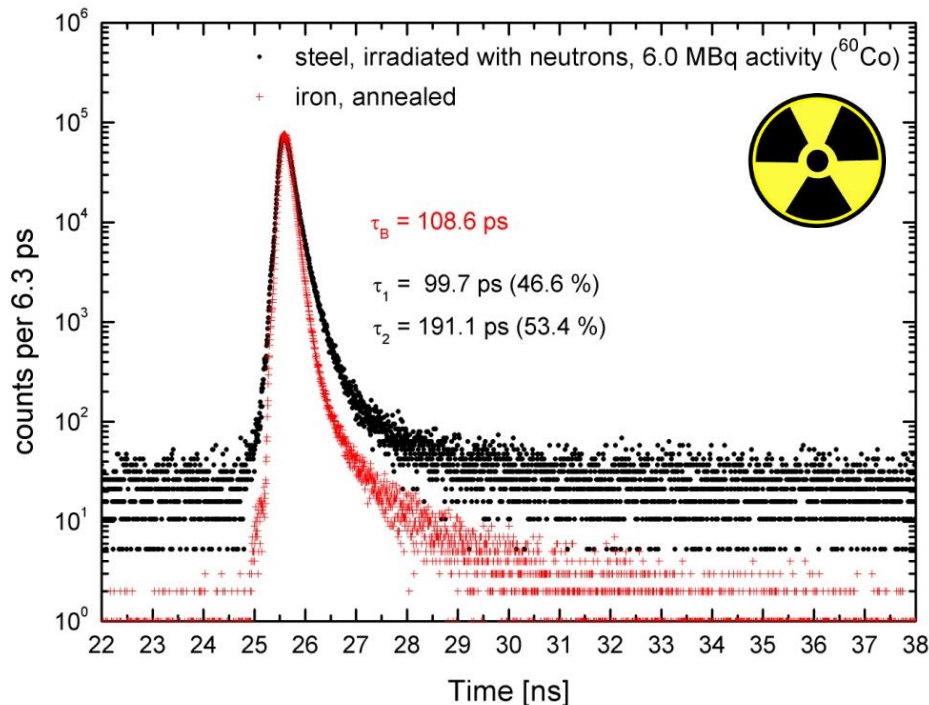
# Gamma-induced Positron Spectroscopy



conventional LINAC mode  
pulsed RF, highest energy  
typically pile-up problems  
F.A. Selim, D.P. Wells, J.F. Harmon, et al.  
Nucl. Instr. Meth. A 495 (2002) 154



SC-LINAC in CW mode  
highest average power –  
high yield and low pile-up



High resolution lifetime spectrum with  
signal to noise ratios of better than  $10^5:1$   
using gamma-gamma coincidence  
techniques for background reduction.  
Lifetime spectra are free from artefacts.

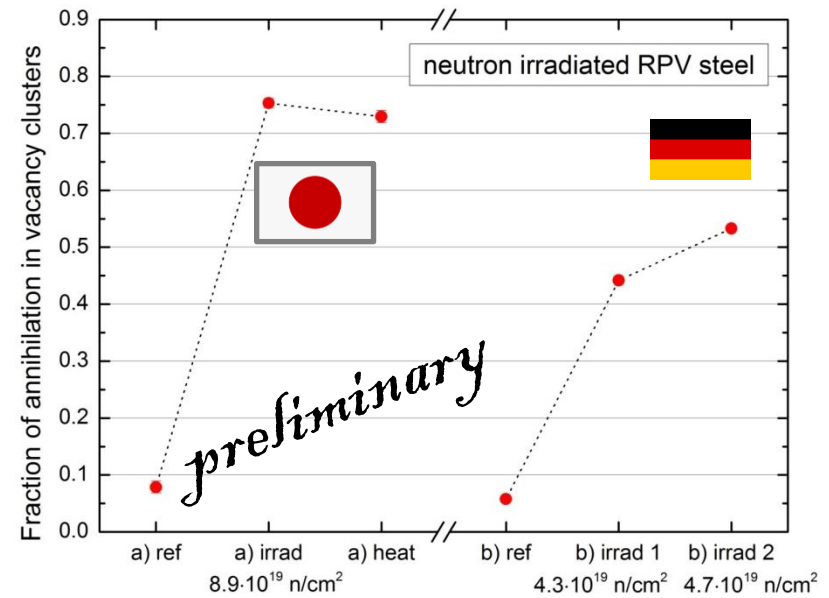
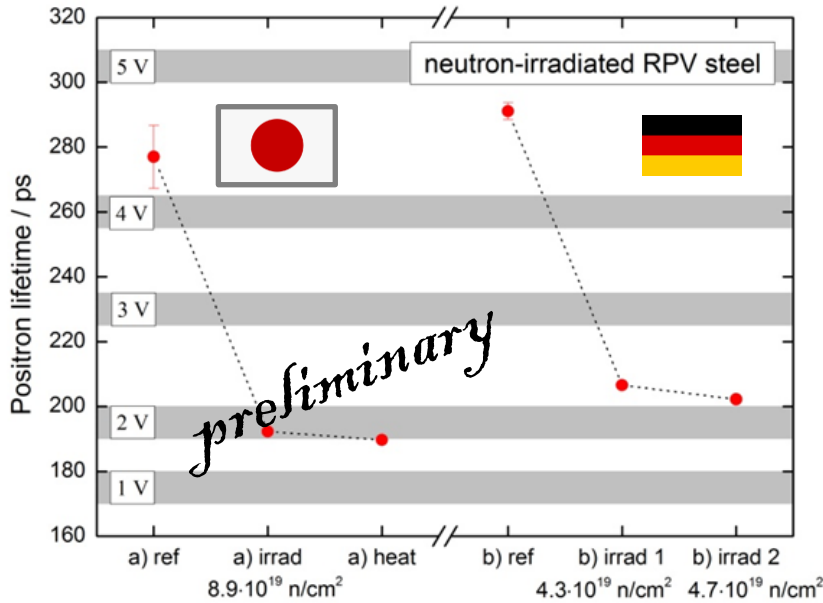
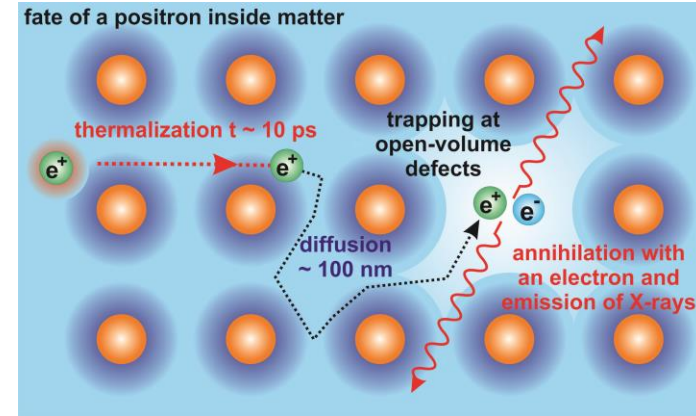
→ Long lifetimes reveal atomic defects  
caused by neutron-induced damage.  
→ Can (and how) defects be removed by  
thermal annealing?

# Physics with GiPS: RPV steel

Reactor vessel steel becomes brittle due to neutron-induced defects like open-volume defects. The atomic defects act as seeds for cracks.



Collaboration with Reactor Safety Division.

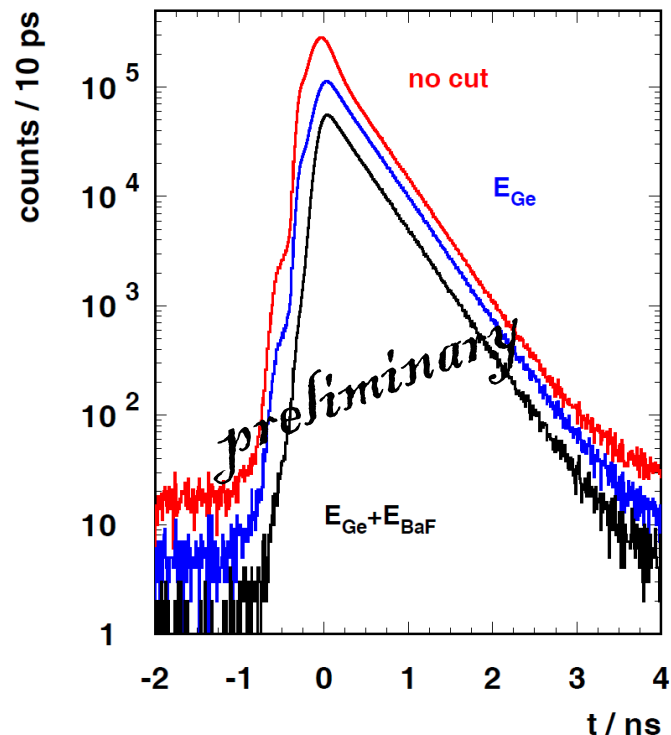
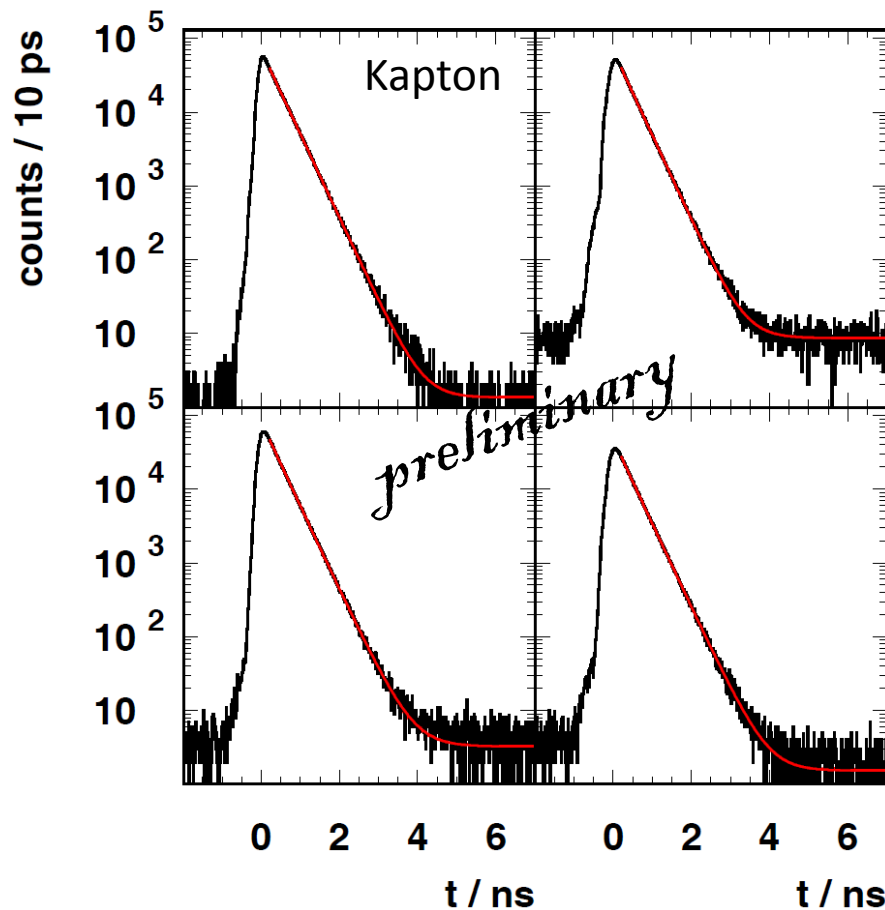


→ Preferential formation of double vacancies

→ Thermal annealing ( $290^\circ\text{C}$ ) not sufficient to remove defects!

# Physics with GiPS: Kapton

Annihilation lifetime in Kapton has been under debate for quite some time. Here, we try to get a measurement without source correction.



applied cuts on Germanium and BaF<sub>2</sub> detector energy signal reduce background from interactions outside the sample

→ consistent single positron lifetime of  $(381 \pm 1)$  ps  
two components show larger  $\chi^2$

# Physics with GiPS: Fluids

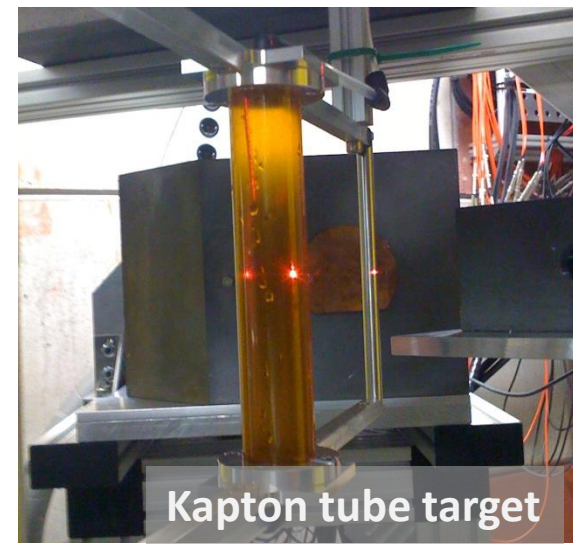
Conventional lifetime measurements:

→ dissolve  $^{22}\text{Na}$  and dispose it afterwards

Positrons from bremsstrahlung

→ homogeneously distributed, sharp time stamp

Target is temperature-stabilized, continuously circulated, degassed, dry-nitrogen flushed.



## Positron Physics

Ortho-Positronium (o-Ps) in a fluid forms a bubble given by its zero-point energy and the surface tension.

o-Ps



142 ns

We know estimate the change of the o-Ps pick-off annihilation lifetime with temperature in a bubble created by the o-Ps itself....

p-Ps



125 ps

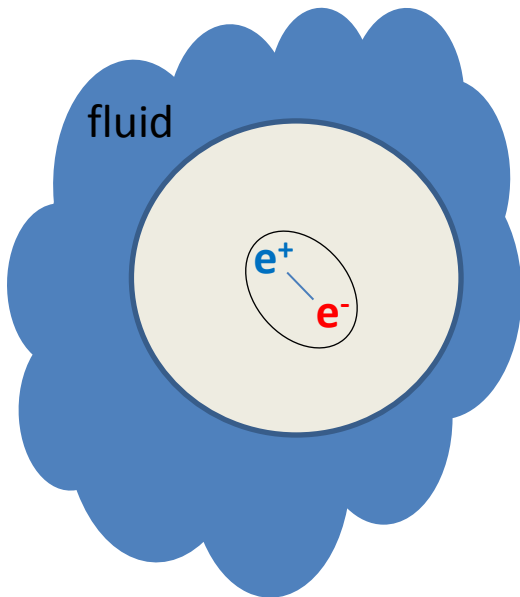
R.A. Ferrell, *Phys. Rev.*, 108,167, 1957

S.J. Tao, *J. Chem. Phys.*, 56,5499, 1972

M. Eldrup *et al.*, *Chem. Phys.*, 63,51, 1981



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# Physics with GiPS: Fluids

$$-\frac{\hbar^2}{2m_{Ps}} \Delta\Psi + U(r)\Psi = E\Psi \quad \text{stationary Schrödinger eqn.}$$

$$\Psi = R(r) \cdot \Theta(\vartheta) \cdot \Phi(\varphi)$$

$$R(r) = R_0 j_l(kr)$$

Ansatz: spherical Bessel fct.

$$j_0(kr) = \frac{\sin kr}{kr}$$

1<sup>st</sup> non-trivial solution

$$E_0 = \frac{\hbar^2}{8m_{Ps}r_0^2} = \frac{\pi^2\hbar^2}{4m_e r_0^2}$$

zero-point energy

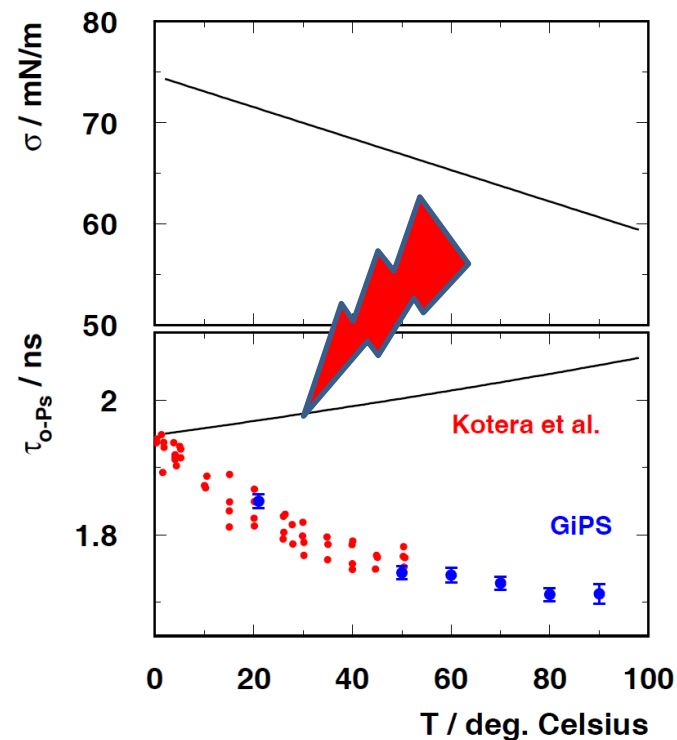
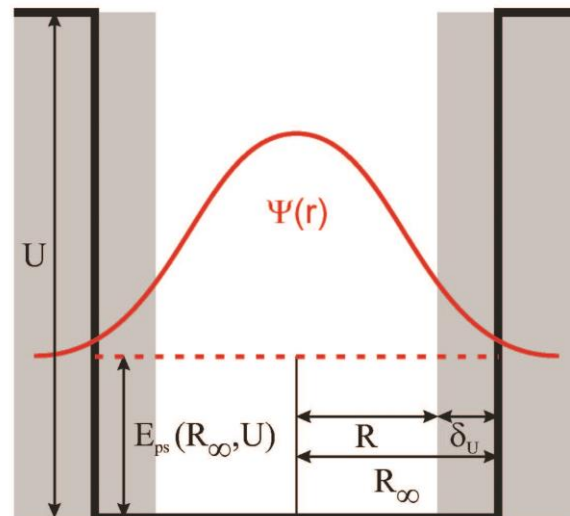
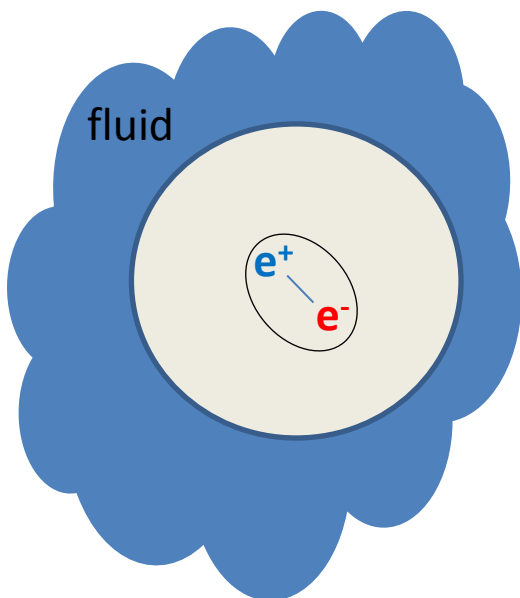
$$E_{surf} = 4\pi r_0^2 \sigma$$

$$\frac{\partial}{\partial r_0} (E_0 + E_{surf}) = 0$$

$$-\frac{\pi^2\hbar^2}{2m_e r_0^3} + 8\pi r_0 \sigma = 0$$

$$r_0 = \sqrt[4]{\frac{\pi\hbar^2}{16m_e \sigma}} = 4.3 \text{ \AA}$$

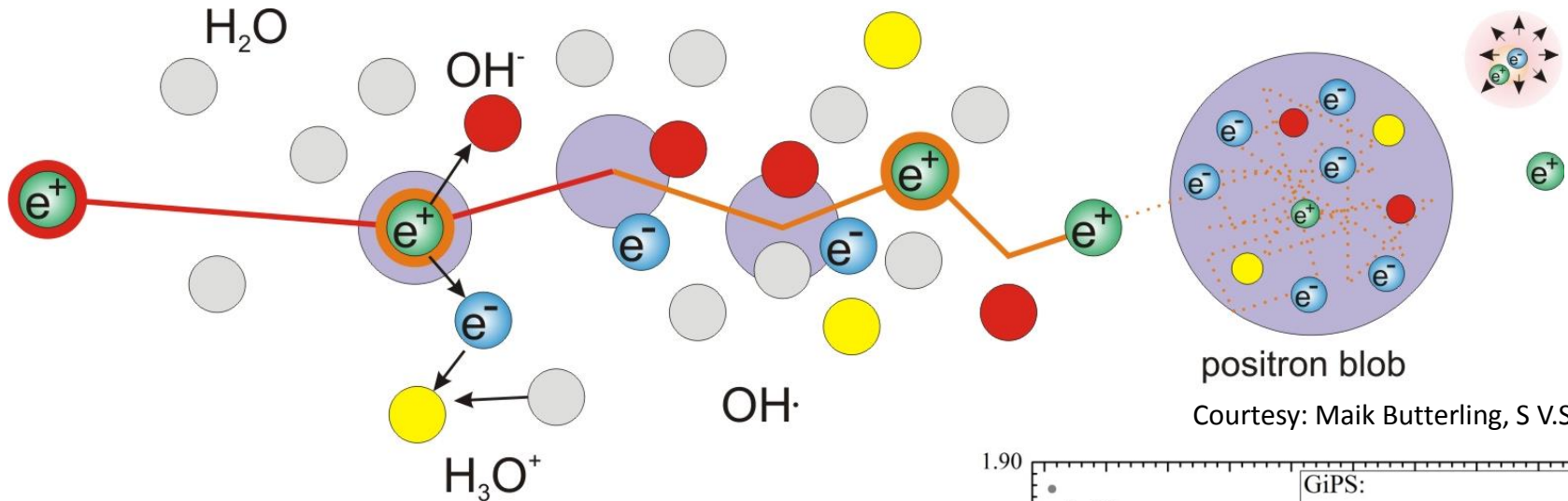
$$a_0 = \frac{4\pi\epsilon_0\hbar^2}{m_\mu e^2} = \frac{\hbar c}{\alpha m_\mu c^2} = 1.06 \text{ \AA}$$





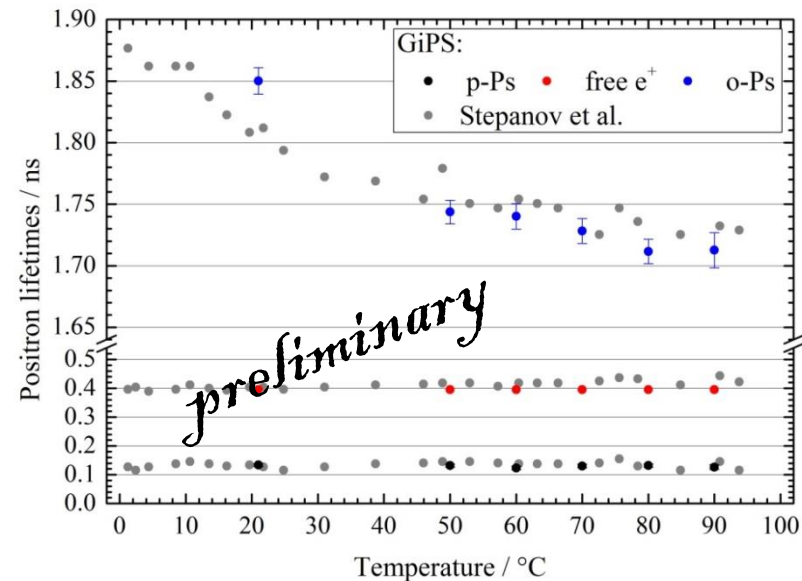
# Physics with GiPS: Positron Chemistry

Experiments with water are **in variance** with a simple bubble-type model. Extension: chemical reactions between radiolysis products of the slowing-down of the positron → Ps chemistry.



Courtesy: Maik Butterling, S.V.Stepanov

- Radicals are positron scavengers which reduce annihilation lifetimes.
- Extended bubble model including chemistry [S.V. Stepanov et al., Mat. Sci. Forum 607] describes data well.
- Relevance for PET diagnostics since  $2\gamma / 3\gamma$  ratio is affected.
- Chemistry of radiolysis directly accessible since the probe creates the ionization itself



# Towards imaging of defects

Material failures impose a significant threat to the integrity and the safety of technical systems. A thorough understanding of the microscopic origin and the development of defects requires advanced methods.



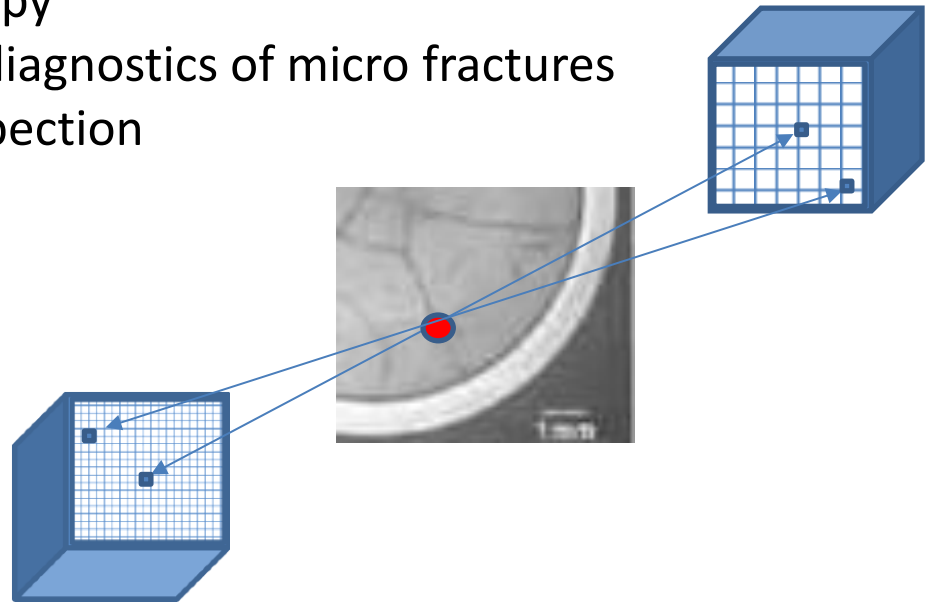
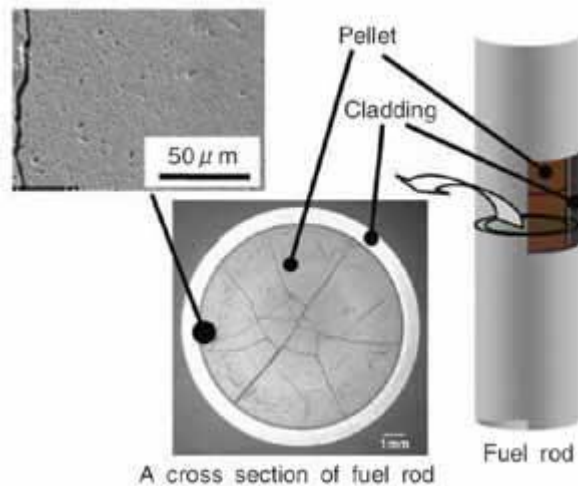
**... and the quests of today**

# Motivation

Establish a **non-destructive** and **non-intrusive** method which allows for **spatially resolved** positron-lifetime spectroscopy. Reconstruct PET-like images plus positron annihilation lifetime.

Possible Applications (list not complete):

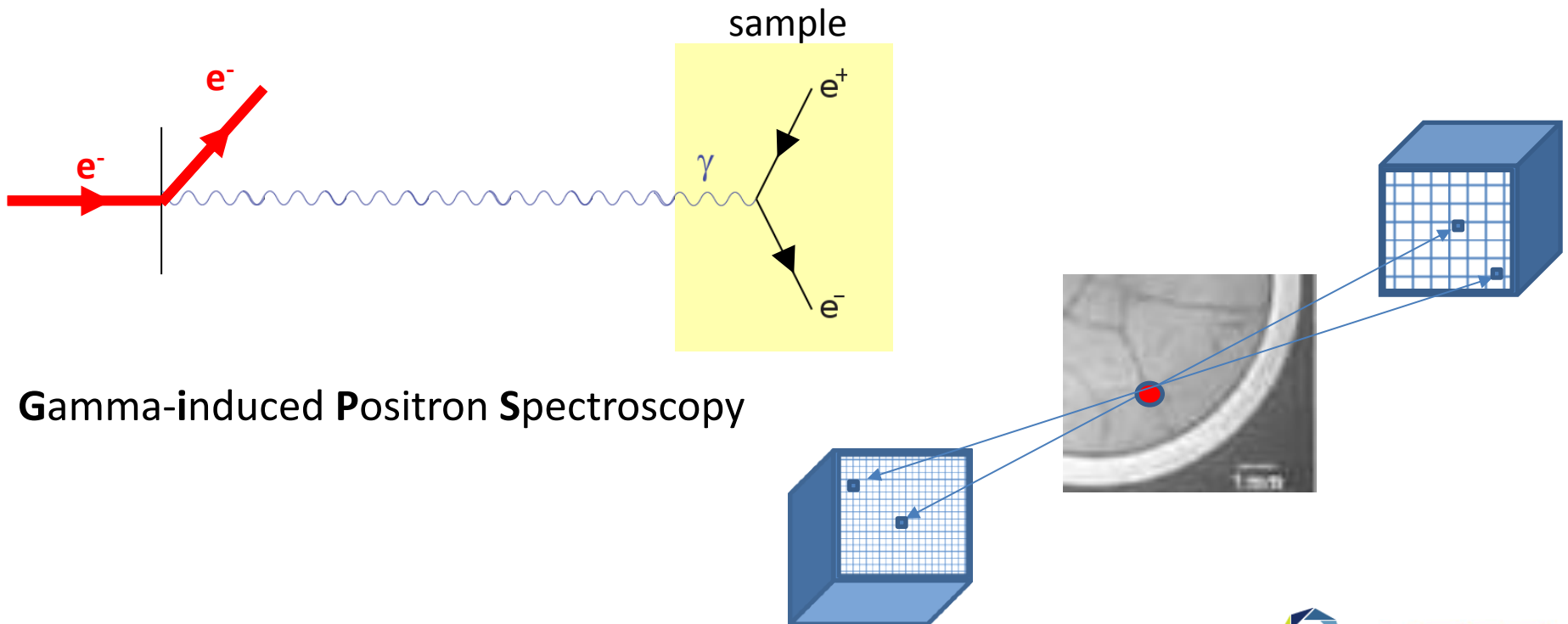
- Porosimetry
- Medicine in-beam positron lifetime spectroscopy during hard x-ray tumor therapy
- Engineering pre-failure diagnostics of micro fractures  
fuel rod inspection



# Prerequisites

- Intense source of positrons with deep penetration (cm)
- Accurate time-stamping of positron creation (<10 ps)
- Position-sensitive positron detectors (mm)
- Time-resolution for lifetime spectroscopy (~100 ps)
- Efficient data acquisition
- 3-D image reconstruction

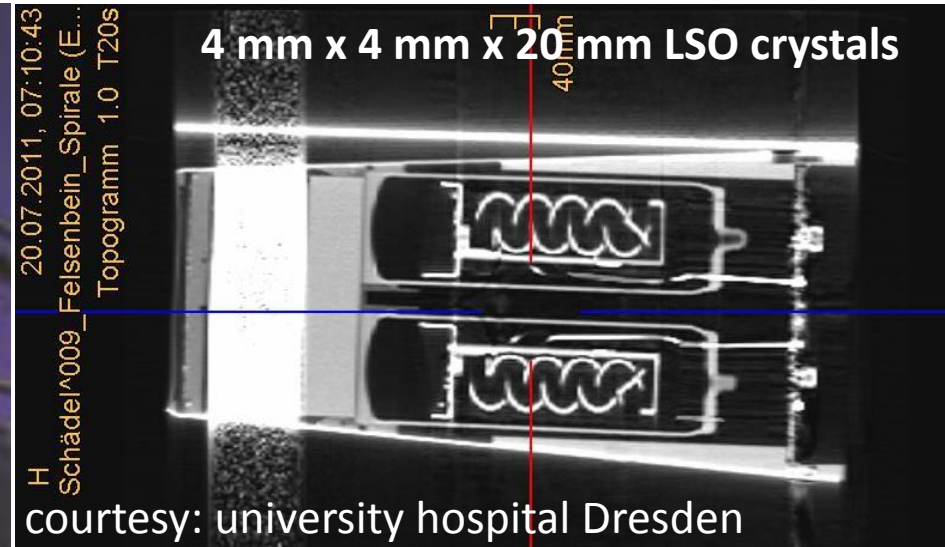
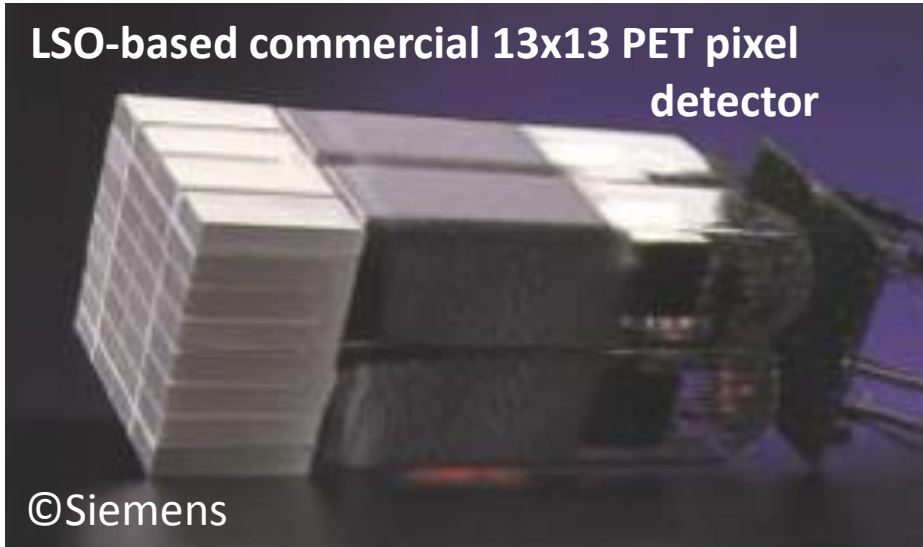
≈15 MeV X-rays  
CW LINAC  
Siemens LSO PET  
in-house (physics)  
in-house (physics)  
in-house (medicine)



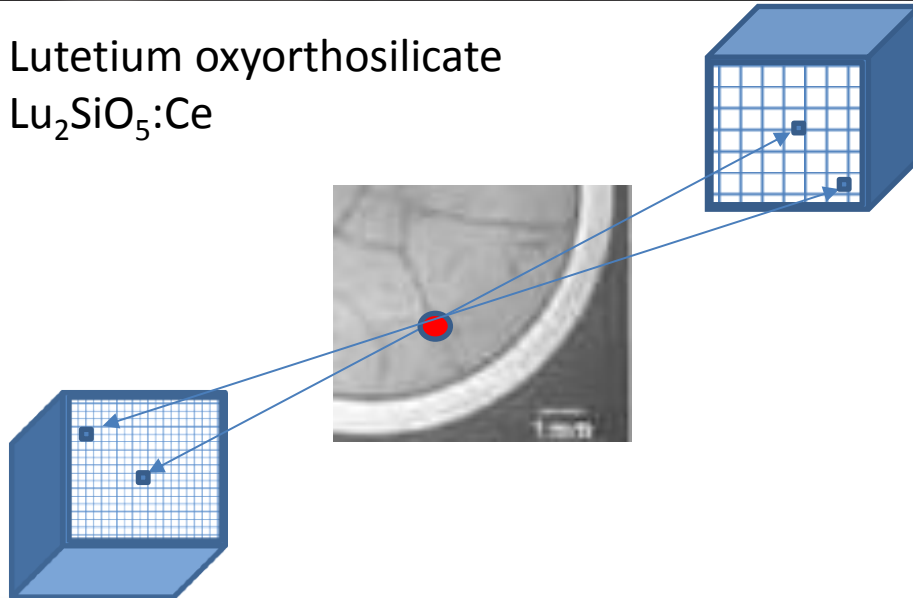
Gamma-induced Positron Spectroscopy

# Towards 2/3-D positron lifetime tomography

- Two position-sensitive photon detectors with 169 elements each

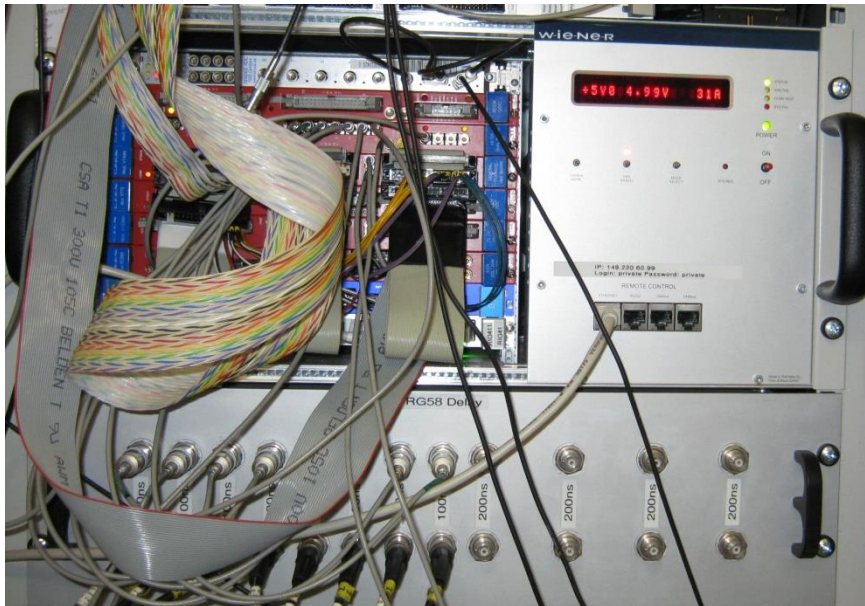
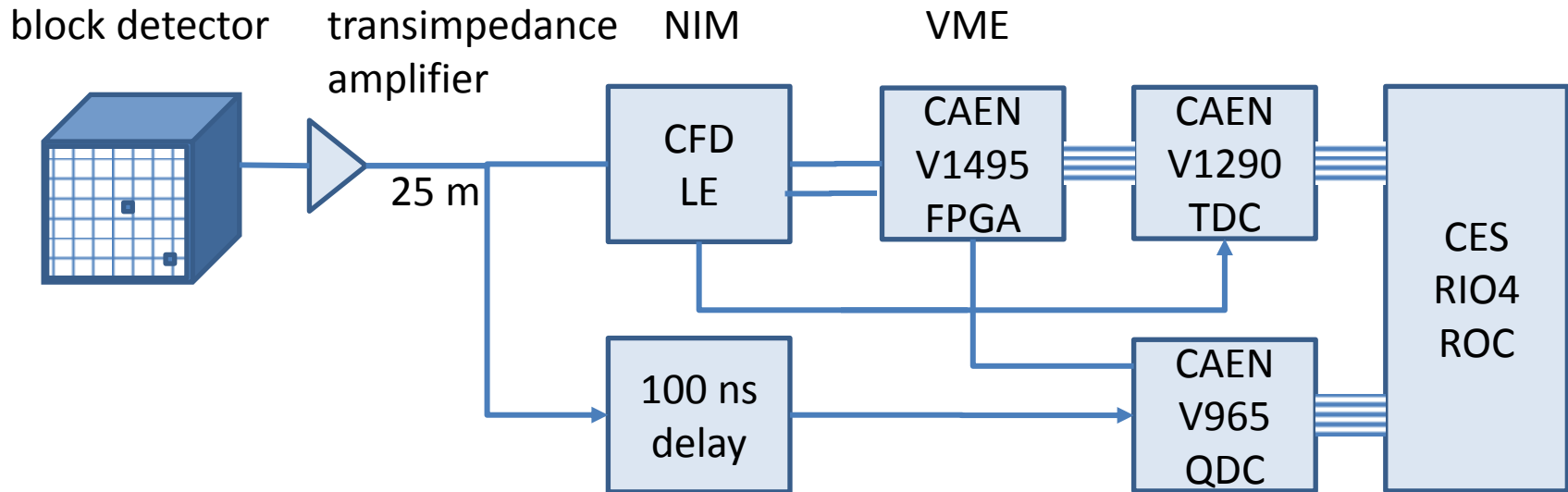


Lutetium oxyorthosilicate  
 $\text{Lu}_2\text{SiO}_5:\text{Ce}$



- Each crystal array read out using 4 PMT
- Summed PMT signal -> gamma energy
- Correlation of individual PMT signals -> position
- Positron annihilation time given by sum over all 8 PMT involved

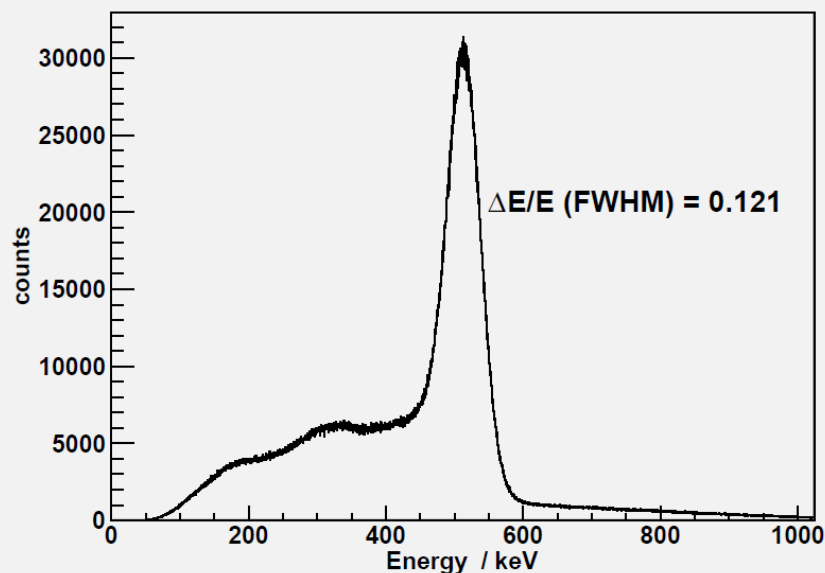
# Electronics (VME)



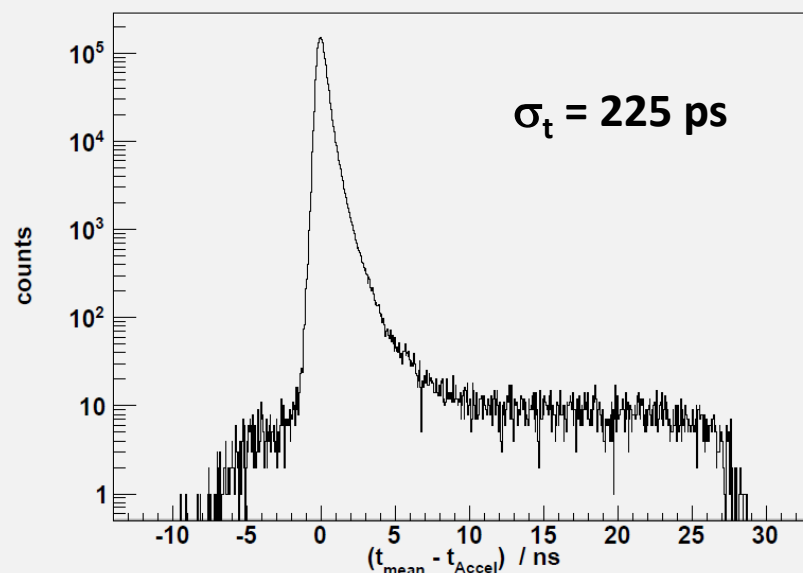
Multi-hit and multi-event buffered readout in VME block mode and readout with  $10 \mu\text{s}$  dead time for 36 channels (QDC & TDC) per event. Throughput is about 10 MB/s sustained. Data acquisition and analysis framework using Multiple-Branch System MBS by Helmholtz-Center for Heavy Ion Research (GSI).

# Calibrations

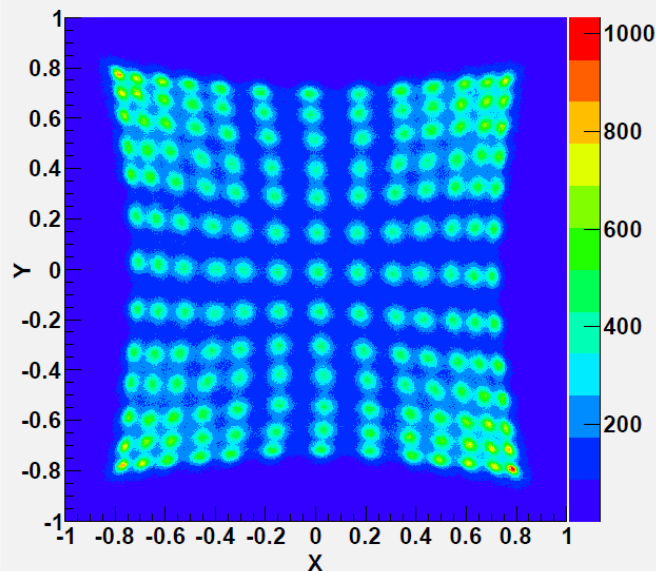
Energy LSO\_0



Positron Lifetime



Pixel LSO\_1



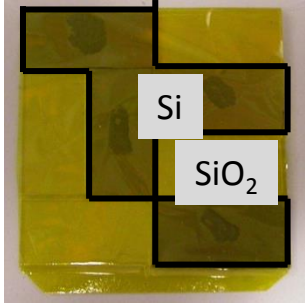
$$E = E_1 + E_2 + E_3 + E_4$$

$$t = \frac{1}{4}(t_1 + t_2 + t_3 + t_4) - t_{\text{accel}}$$

$$x = \frac{(E_1 + E_2) - (E_3 + E_4)}{E_1 + E_2 + E_3 + E_4}; \quad y = \frac{(E_1 + E_3) - (E_2 + E_4)}{E_1 + E_2 + E_3 + E_4}$$

Calibration done using 7 cm x 7 cm aqueous  $^{18}\text{F}$  source w/ 200 MBq ( $T_{1/2} \approx 2$  h) produced in-house.

# Sample cases



Proof of principle, first test  
Simple 2D target  
→ proof of principle  
→ simple back-projection method



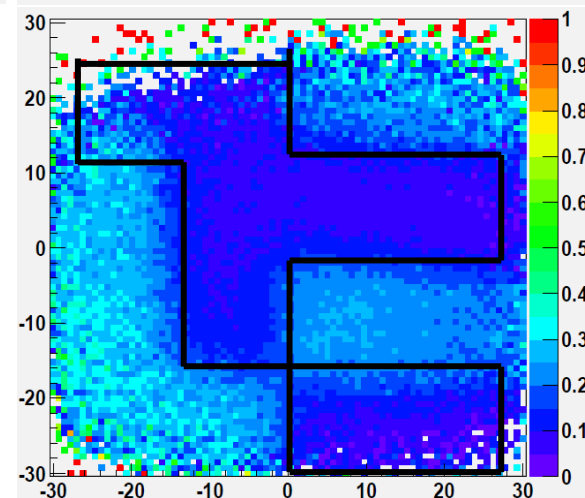
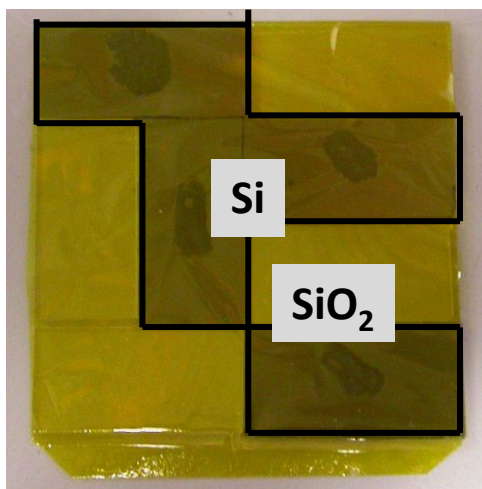
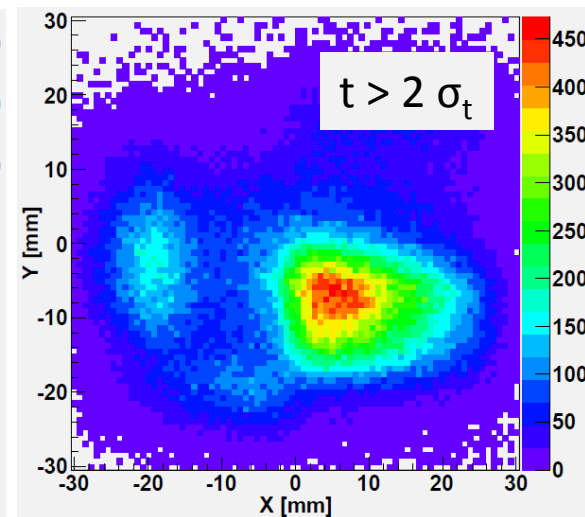
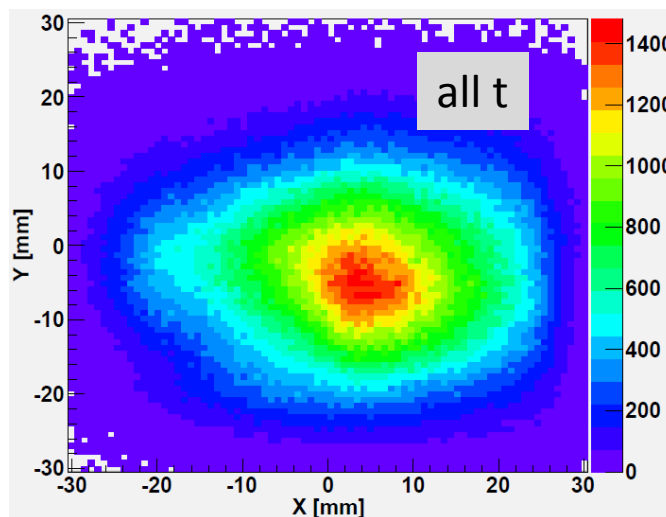
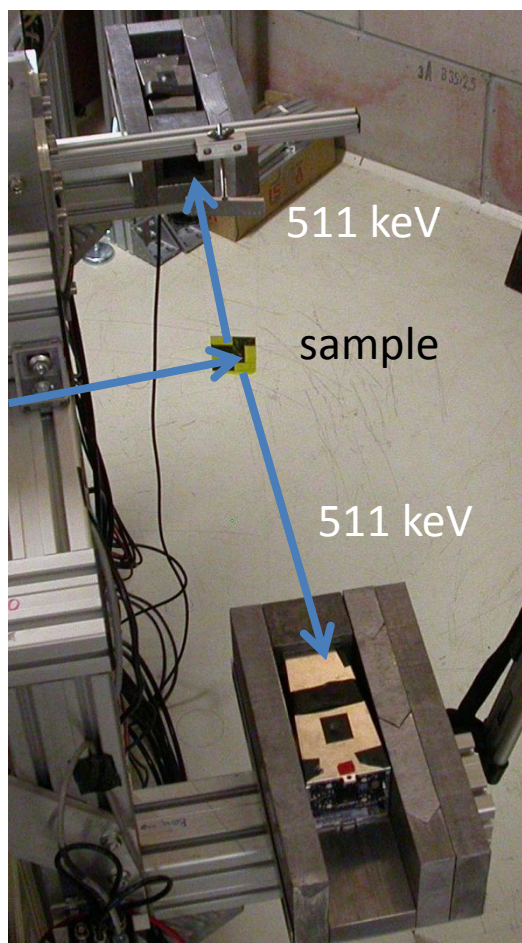
3D target  
→ Reconstruction of data as a function of life time



Real world sample (cutout from 91.4 T magnet coil)  
→ What we can learn from our method



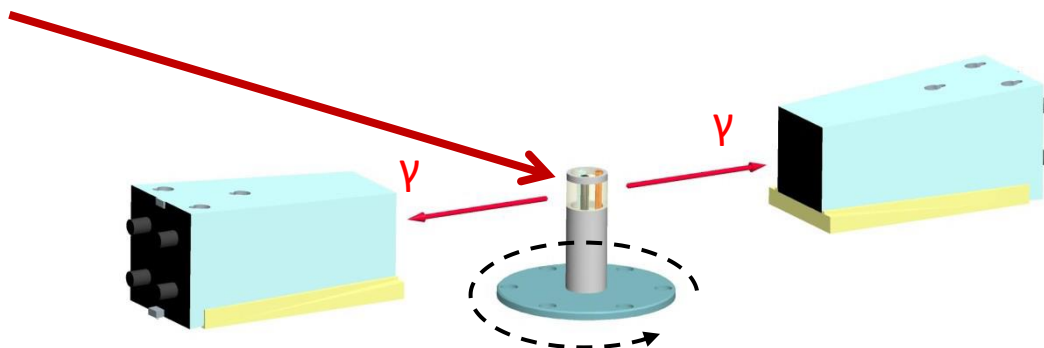
# Setup and results: 2D image reconstruction



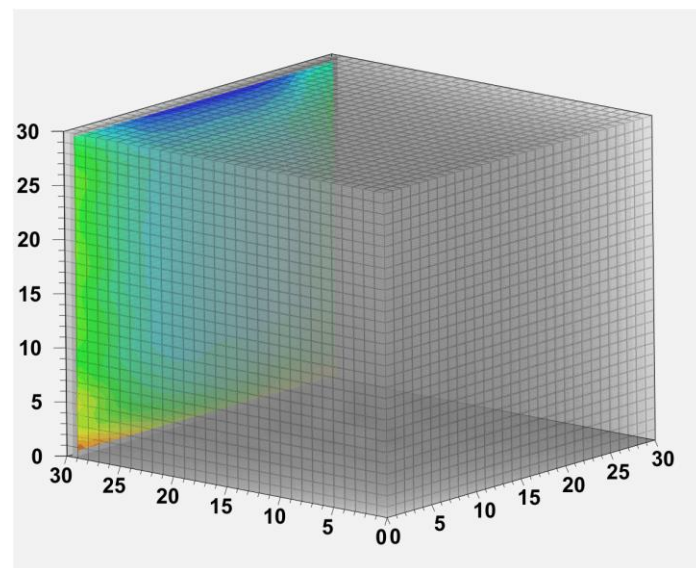
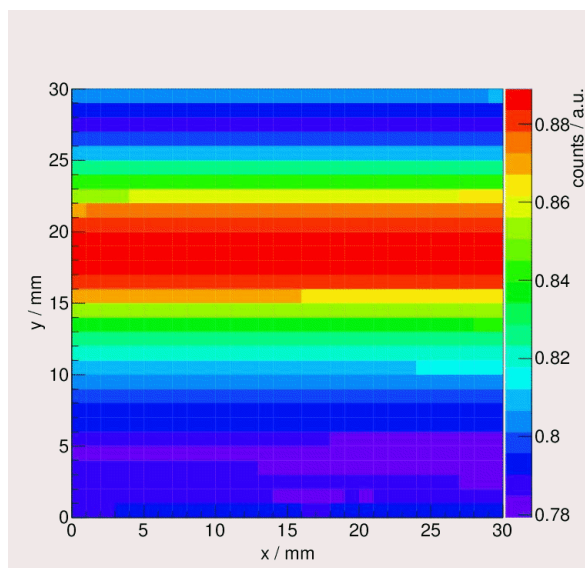
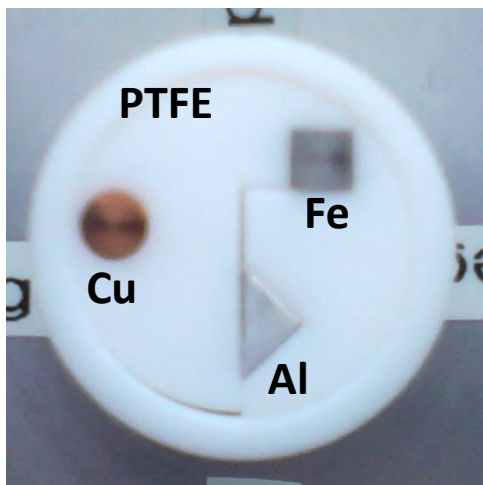
Sample selected to give balanced positron yield.  
Lifetime-gated 2D reconstructed image by back-projection.

# 3D reconstruction

Bremsstrahlung beam



3D tomography applied for the **first time** using bulk volume positron production. Target is rotated in 2 deg. steps and the image is reconstructed using a cubical (30 mm)<sup>3</sup> voxel space and back-projection algorithm.



# Maximum Likelihood Expectation Maximization



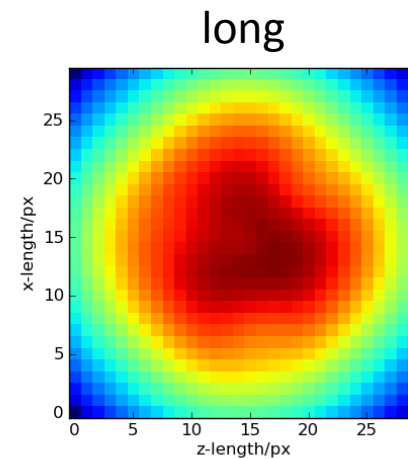
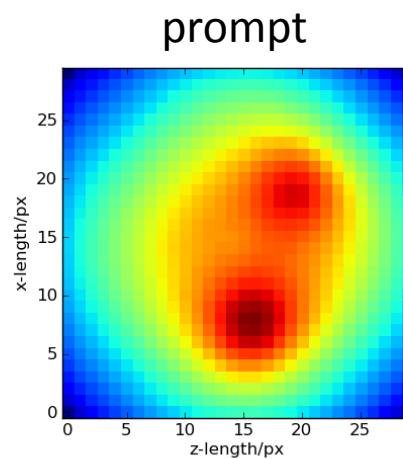
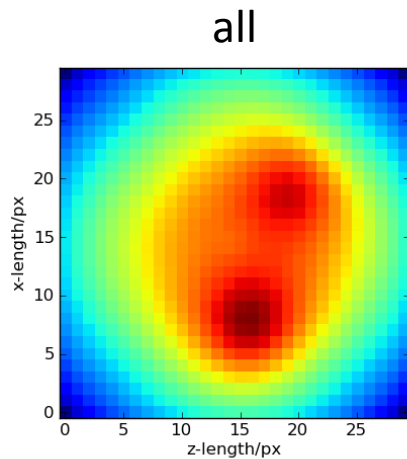
Iterative method for image reconstruction based on an algorithm developed in PET [L.A. Shepp, Y. Vardi, IEEE-MI 2 (1982) 113].

Solves the inversion problem numerically where one has a **system matrix**  $M$ , an a-priori unknown **source distribution**  $S$  and a **measured distribution**  $r$ .

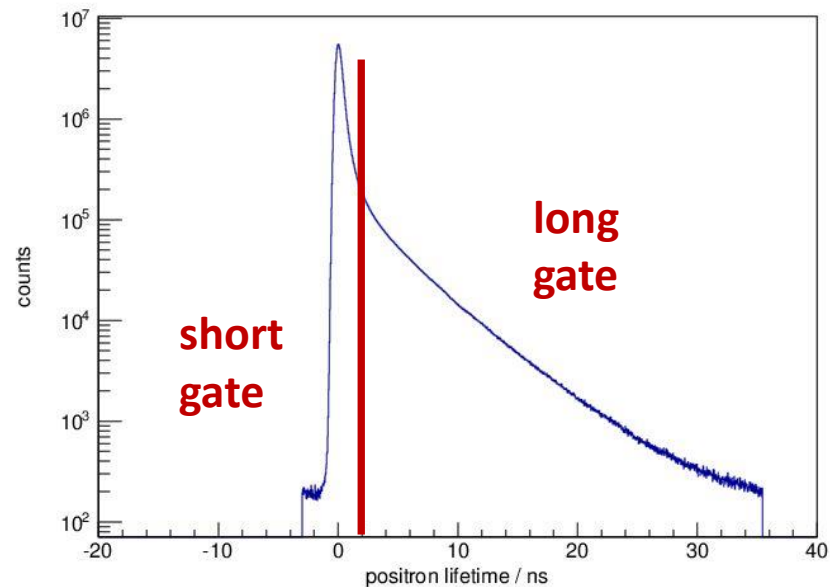
$$\hat{M} \cdot S = r$$

The system matrix has a size of  $13^2 \times 13^2 \times 180 \times 30^3 = 138 \times 10^9$ .

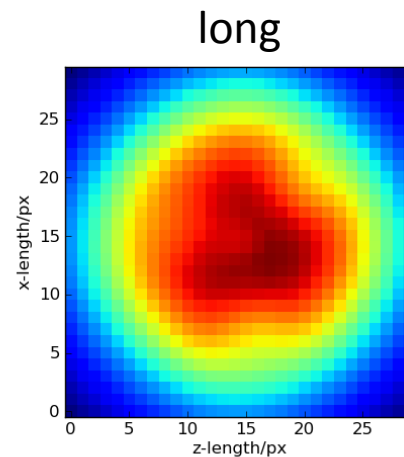
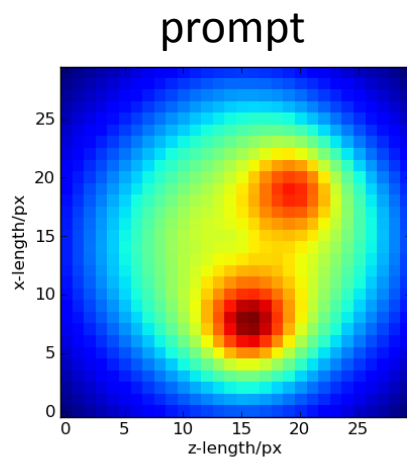
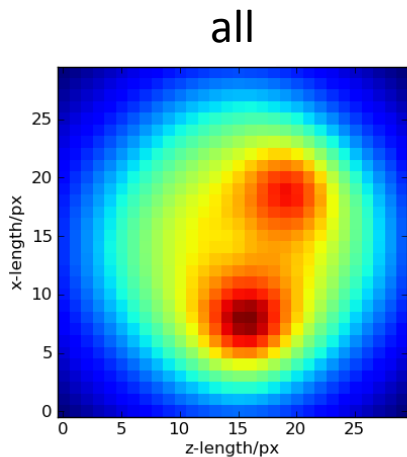
step 1



# MLEM

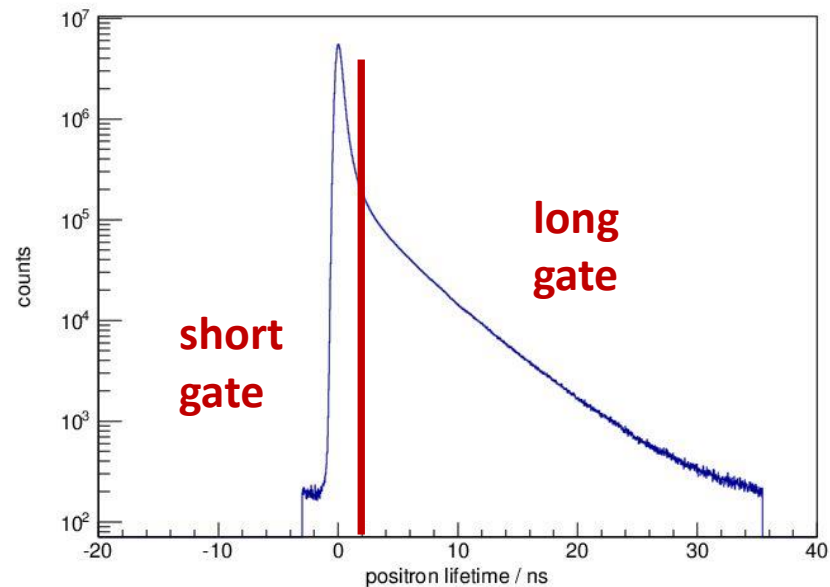


step 2

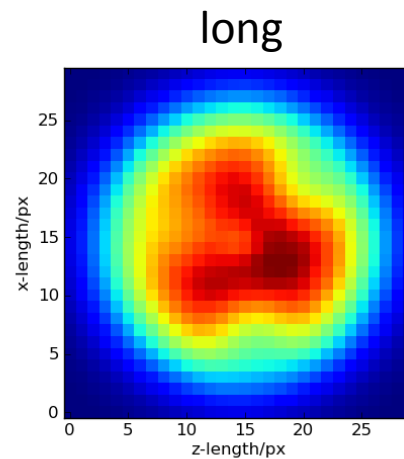
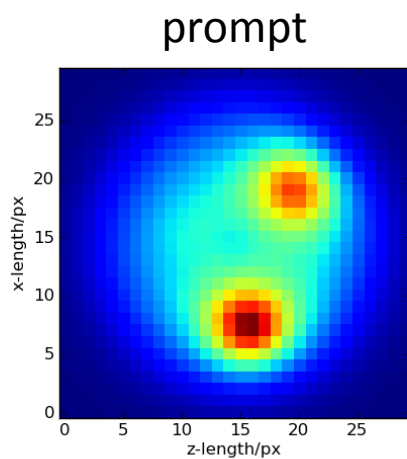
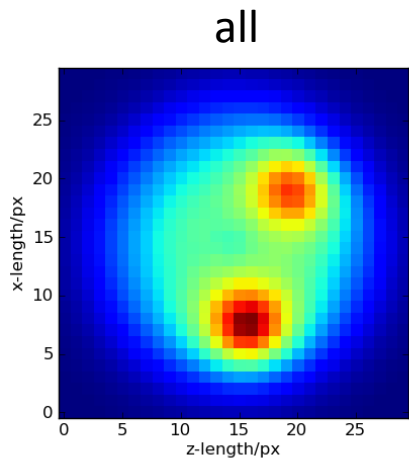


2<sup>nd</sup> Iteration

# MLEM

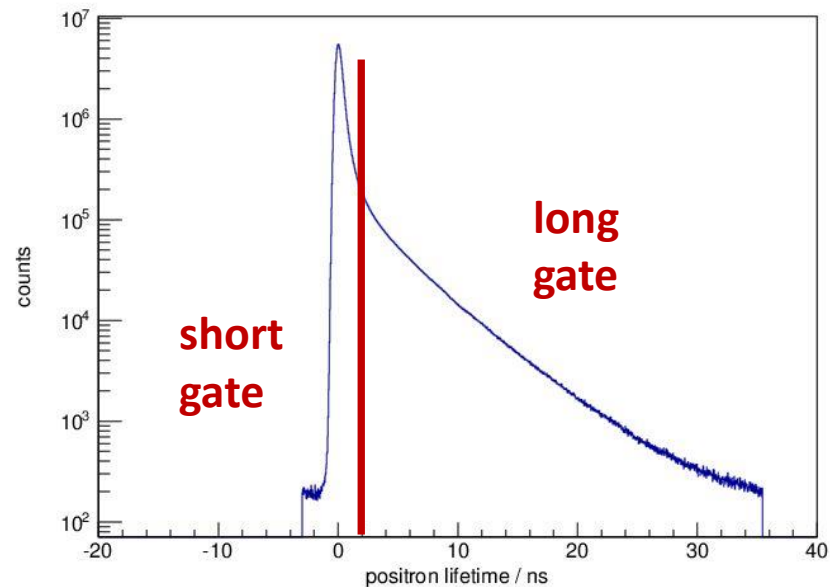


step 5

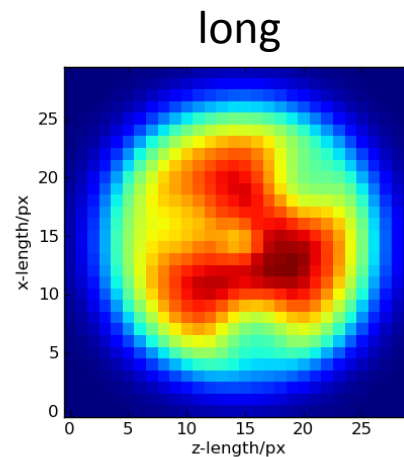
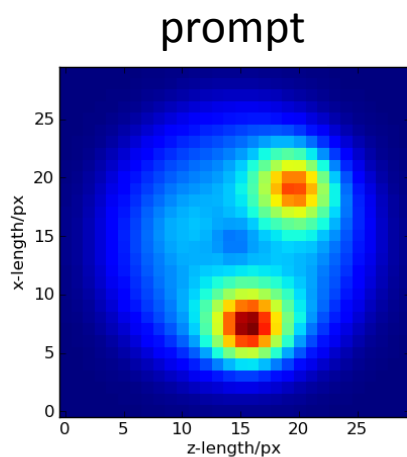
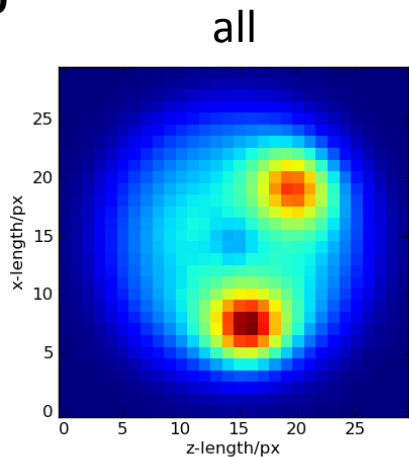


5<sup>th</sup> Iteration

# MLEM

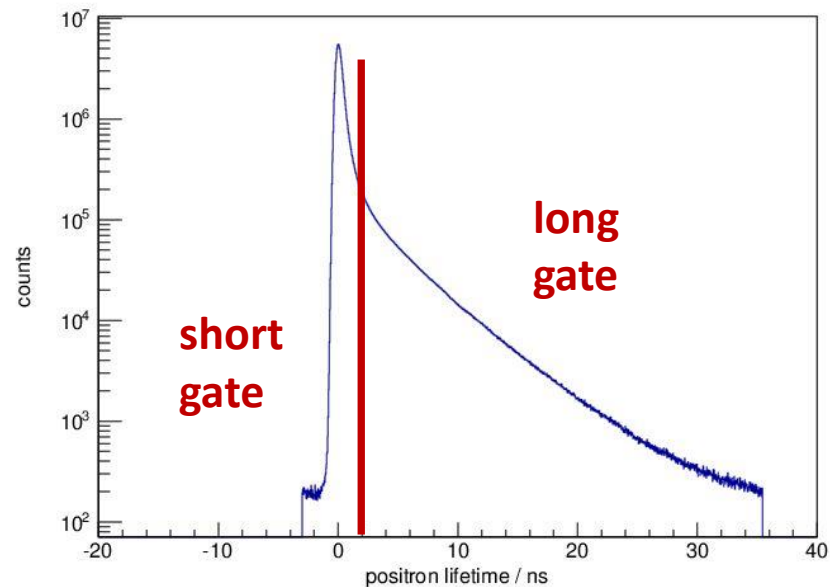


step 10

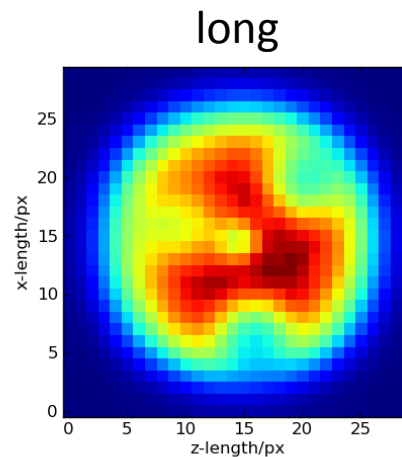
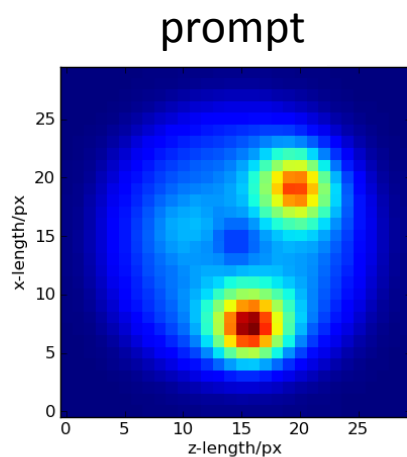
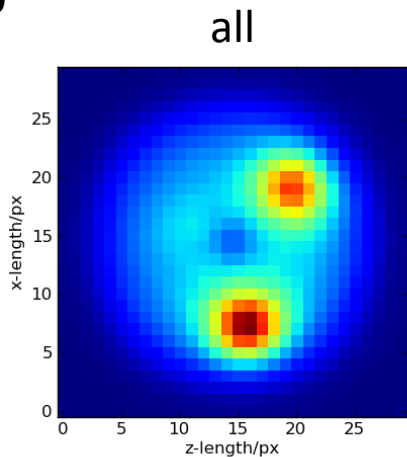


10<sup>th</sup> Iteration

# MLEM

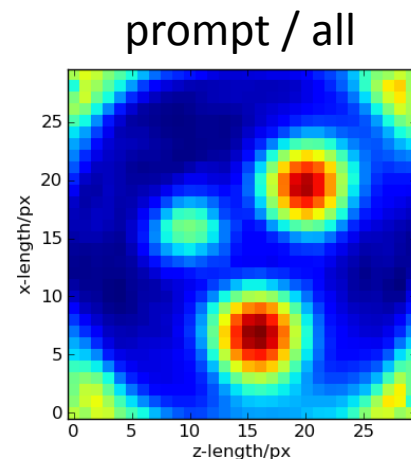
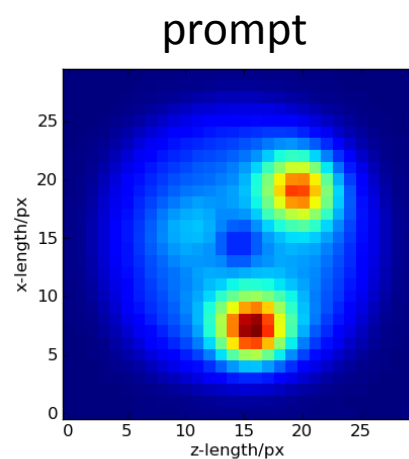
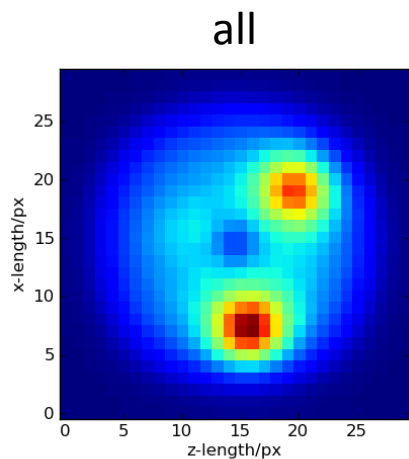
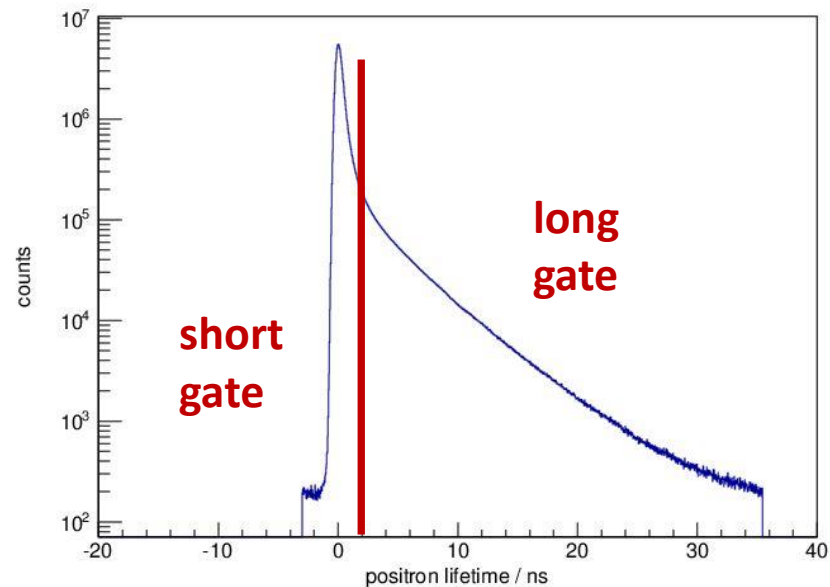


step 20



20<sup>th</sup> Iteration

# MLEM



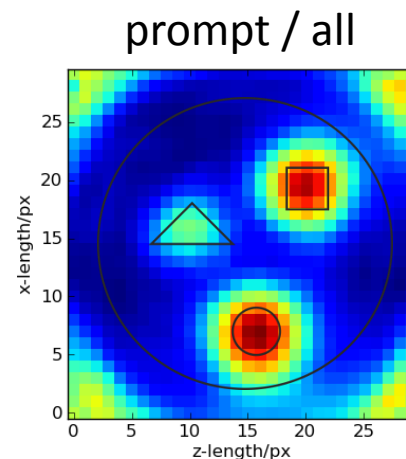
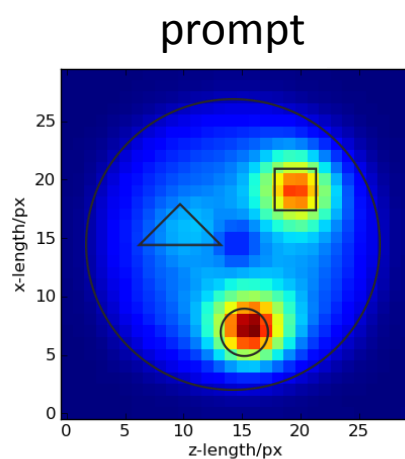
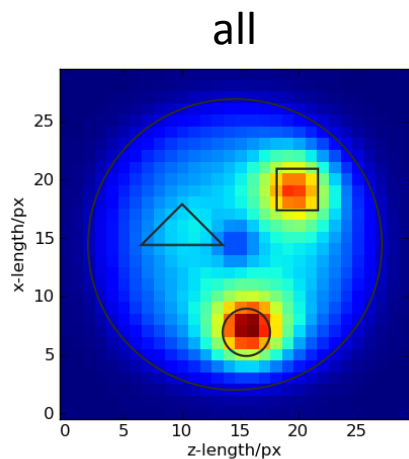


# MLEM

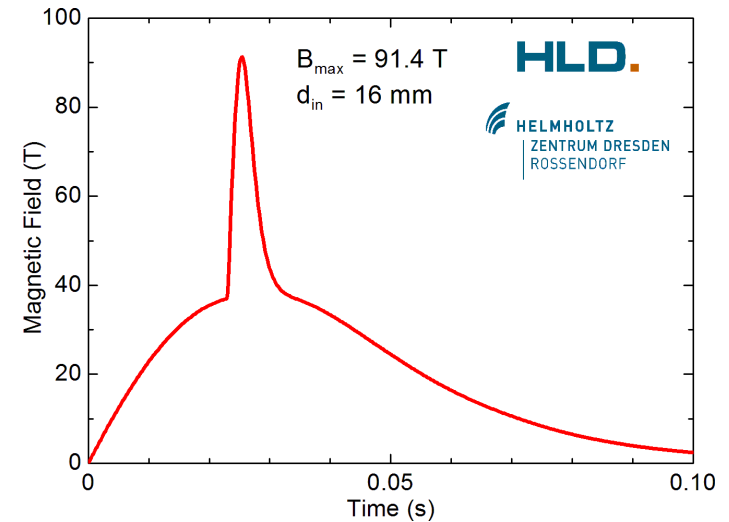
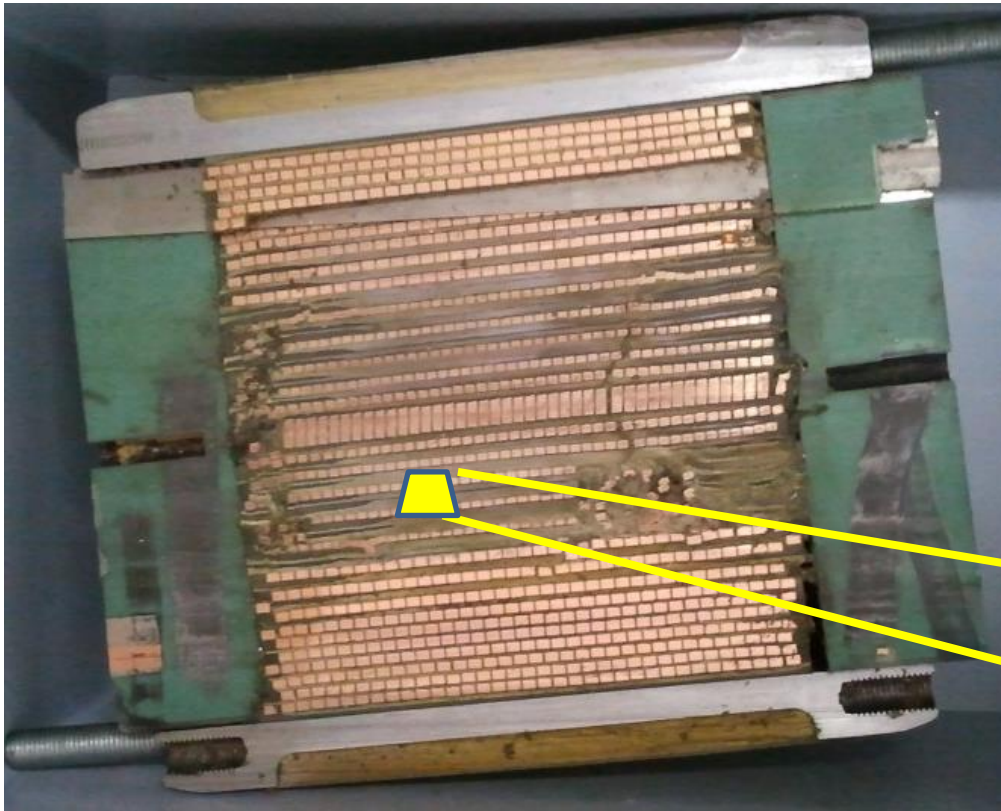


Gating on positron lifetimes with 225 ps timing resolution.

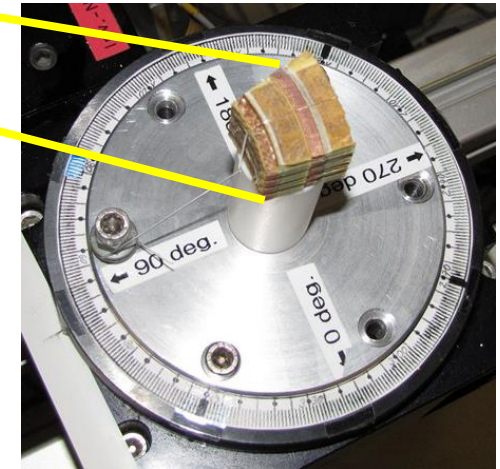
Now the Al is clearly discriminated against the surrounding Teflon.



# Lifetime-sensitive analysis B-field coil

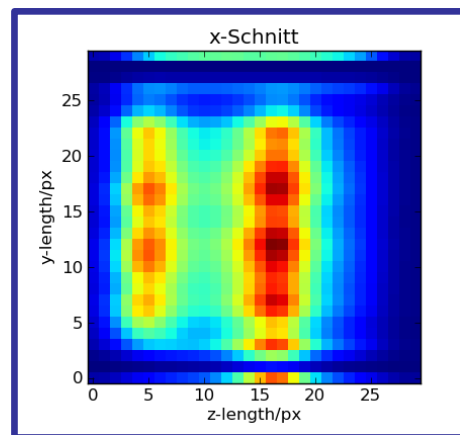
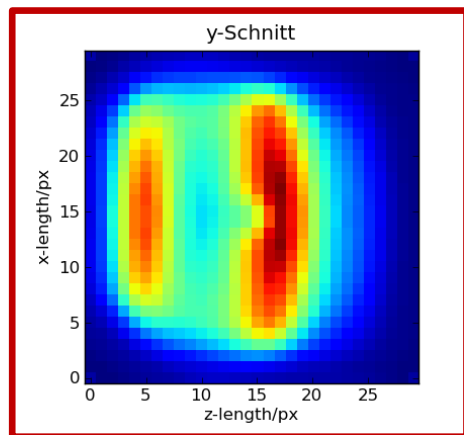
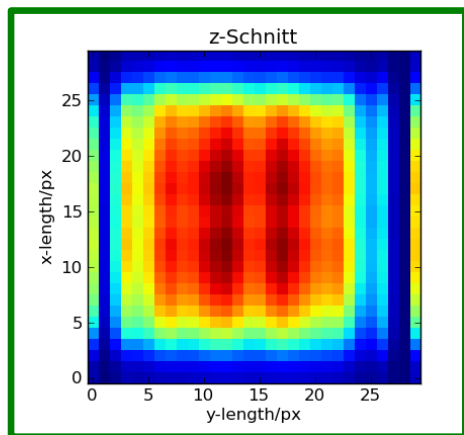
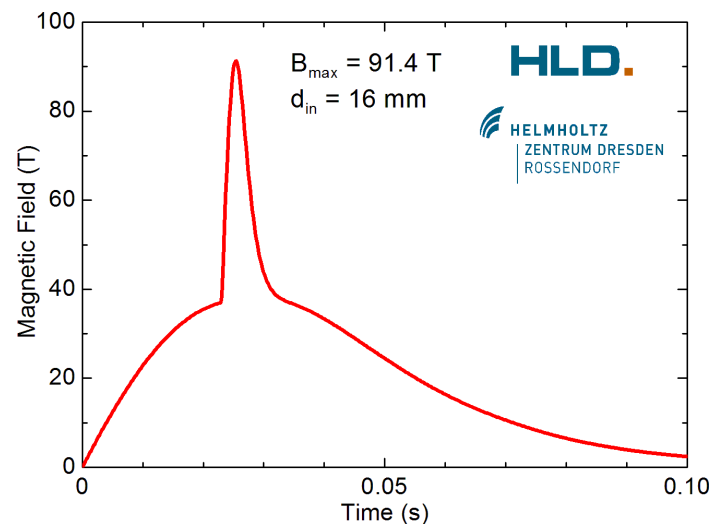
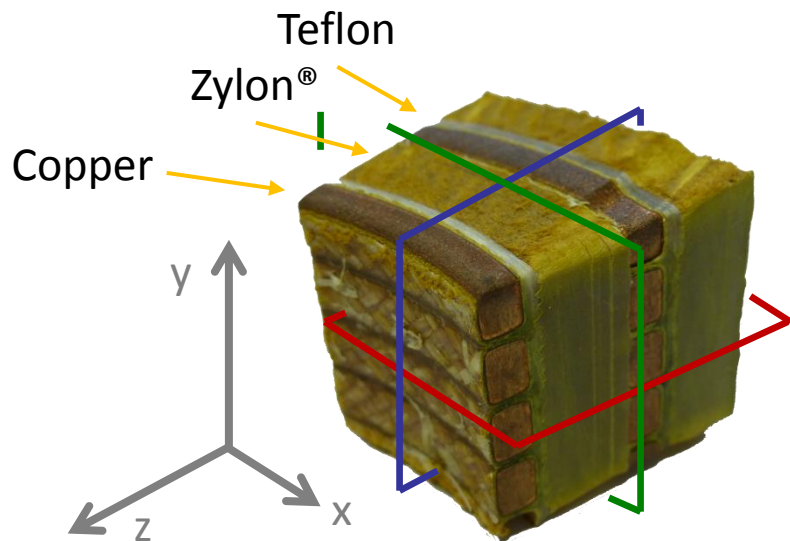


Courtesy: Jochen Wosnitza



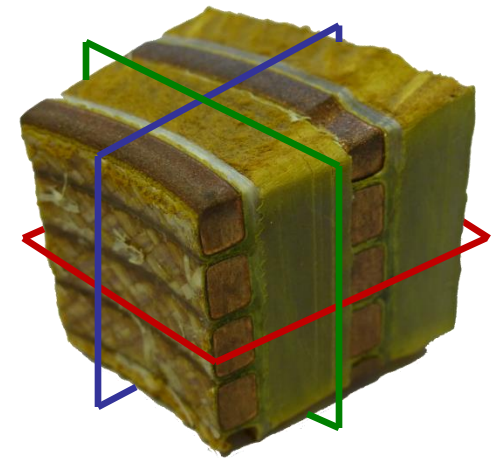
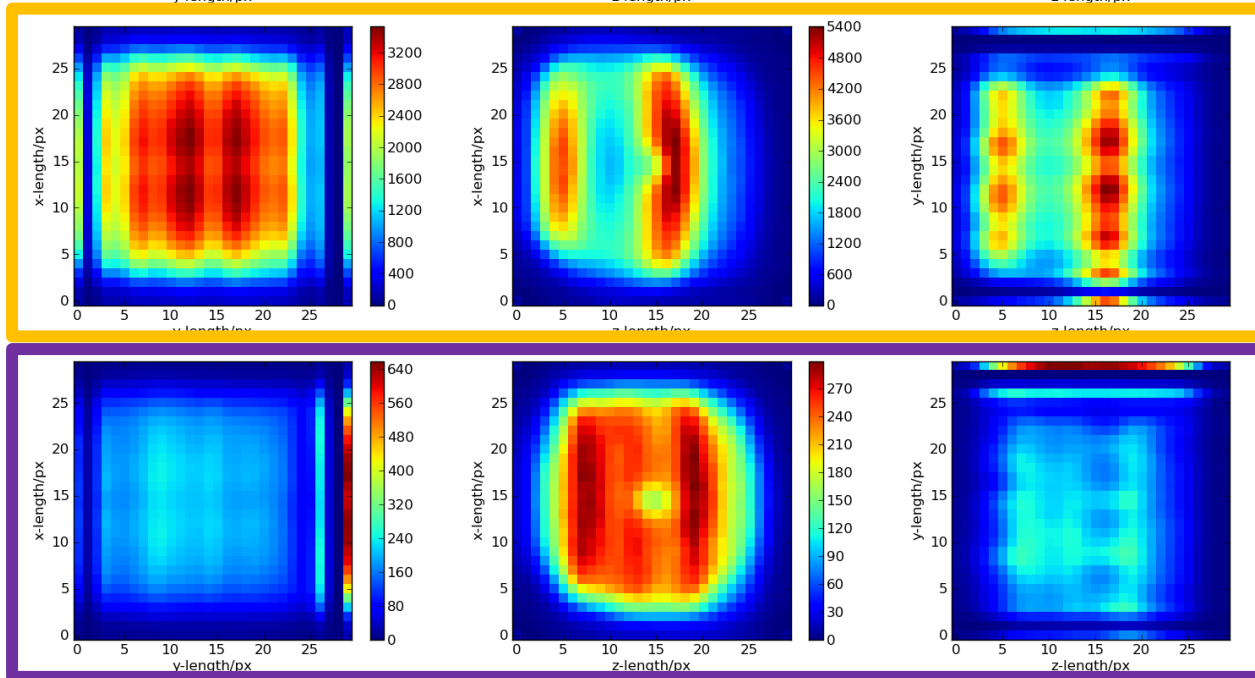
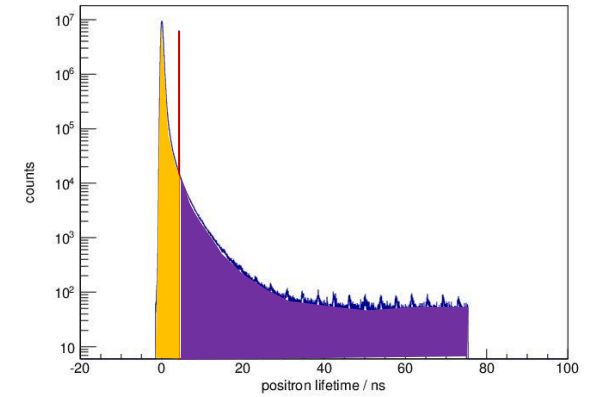
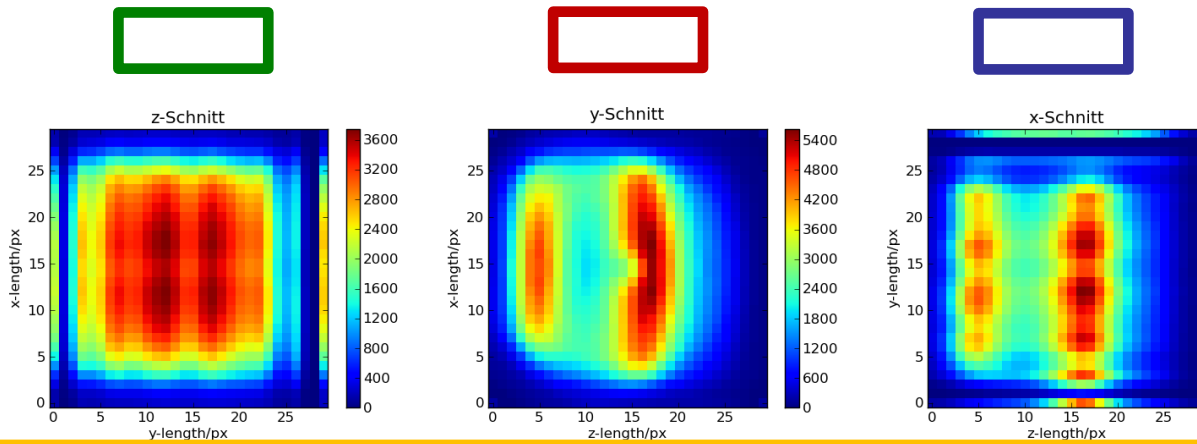
Cut through the record coil which reached 91.4 T peak field. Coil is fed by the world's largest capacitor bank w/ 50 MJ stored energy.

# Tomography: B-field coil

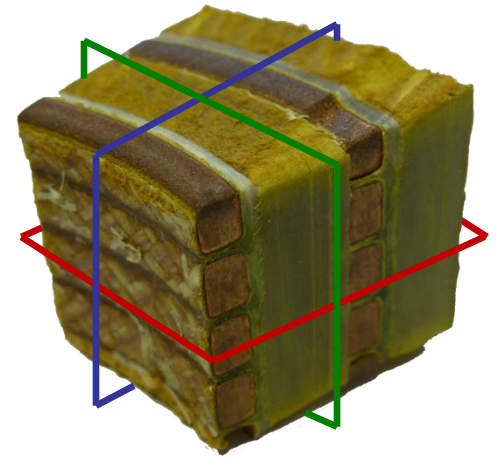
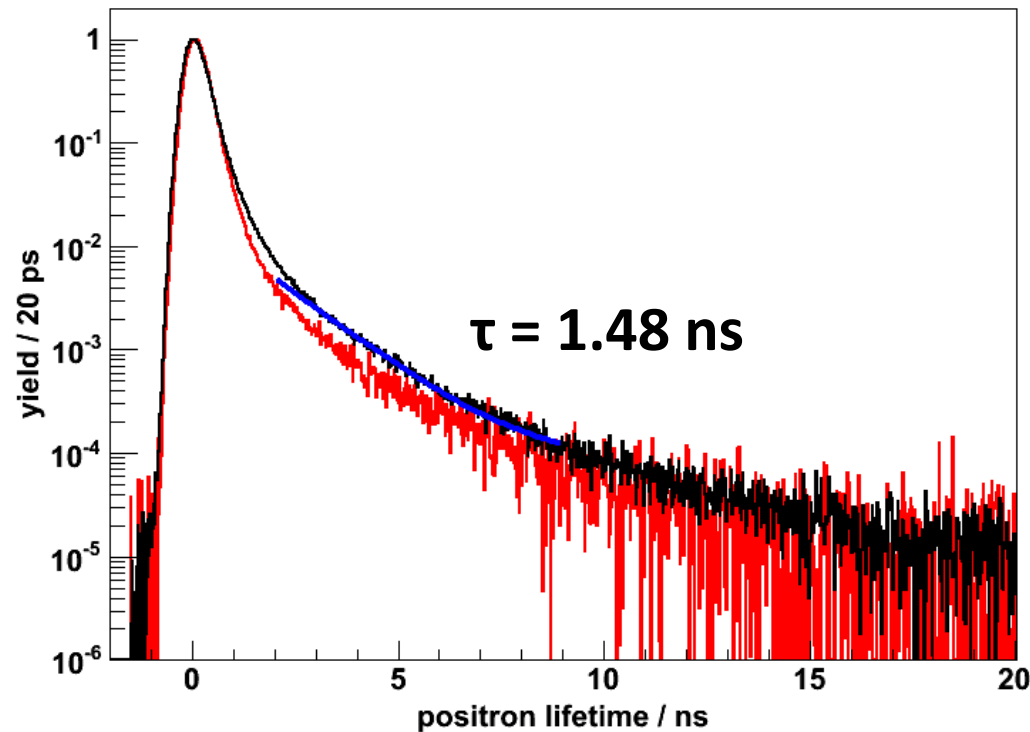


48 h measurement time, 316 GB, 1.6 G events  
324 M filtered coincidences

# Lifetime-sensitive analysis: B-field coil



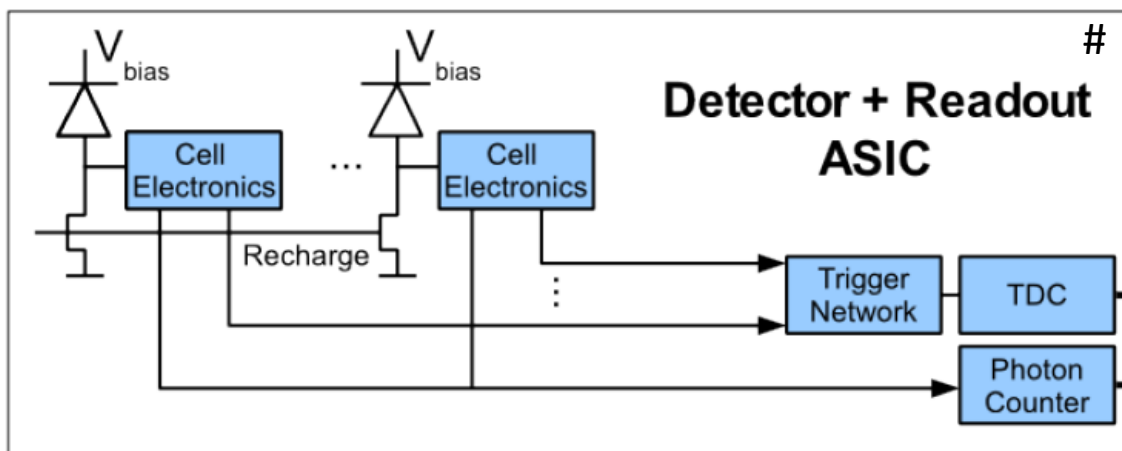
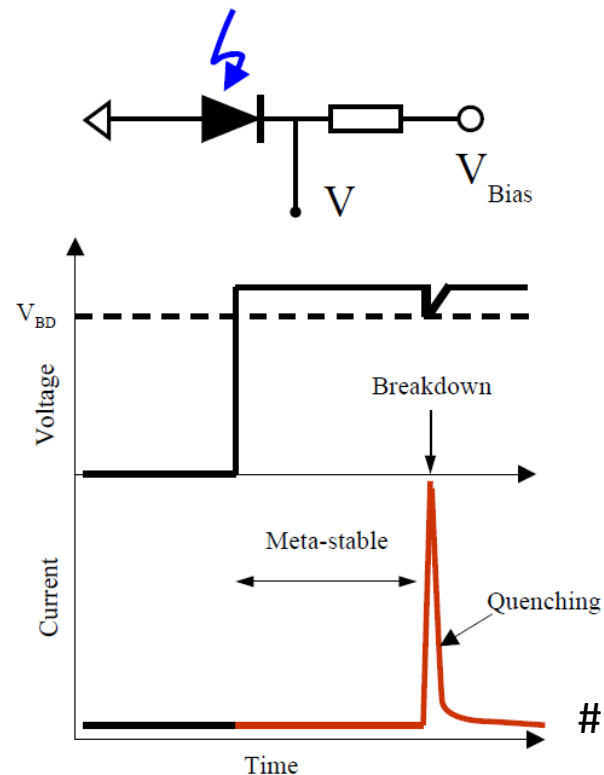
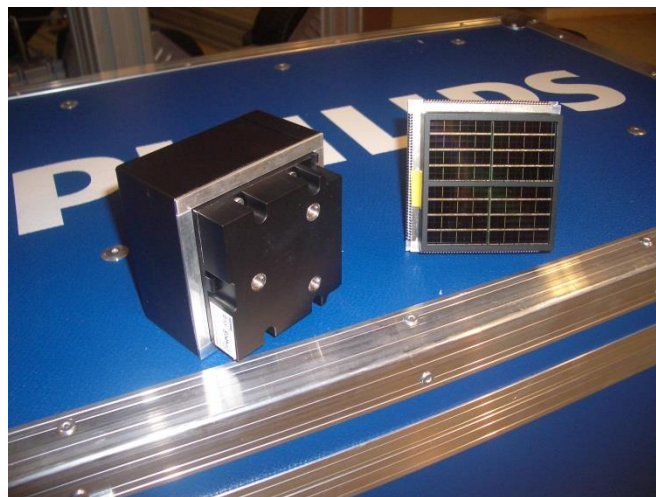
# Lifetime-sensitive analysis: B-field coil



Now, we select specific voxels and determine the annihilation lifetimes for spatially separated regions. Since the voxel is identified as an ensemble over all possible lines-of-response between two detector crystals, the lifetime distribution is a convolution as well. Some real physics questions needed ...

# Extensions

Digital Silicon Photomultiplier (dSiPM)  
Module  
DPC3200-22-44  
(819200 pixel each)



Digitally counting the number and the time of arrival of scintillation photons (here LYSO)

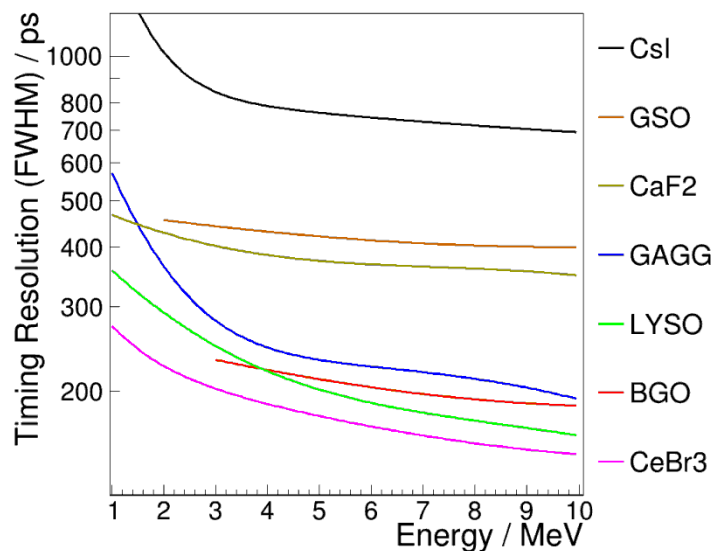
#Courtesy: Philips Digital Photon Counting

# Extensions

## digital Silicon Photomultiplier (dSiPM)

Employ the scaled accelerator radio frequency (13 MHz) via a phase-locked loop (PLL) as dSiPM system clock.

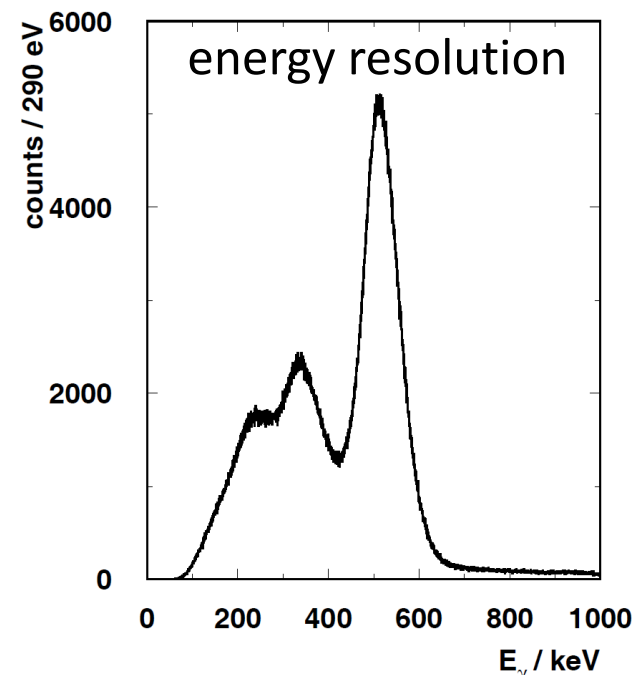
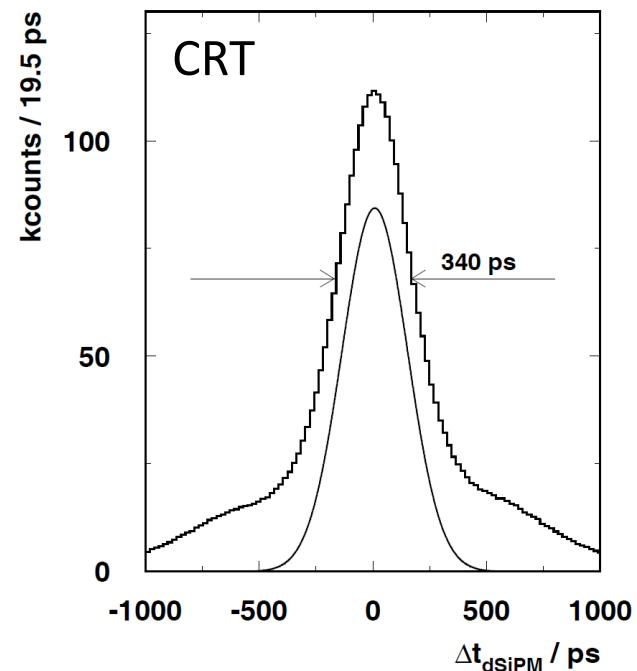
- > Intrinsic synchronization for optimal timing resolution.
- > 170 ps FWHM seem possible



## Scintillation materials

Collaborative effort within gamma-ray imaging group at particle-therapy center Oncoray.

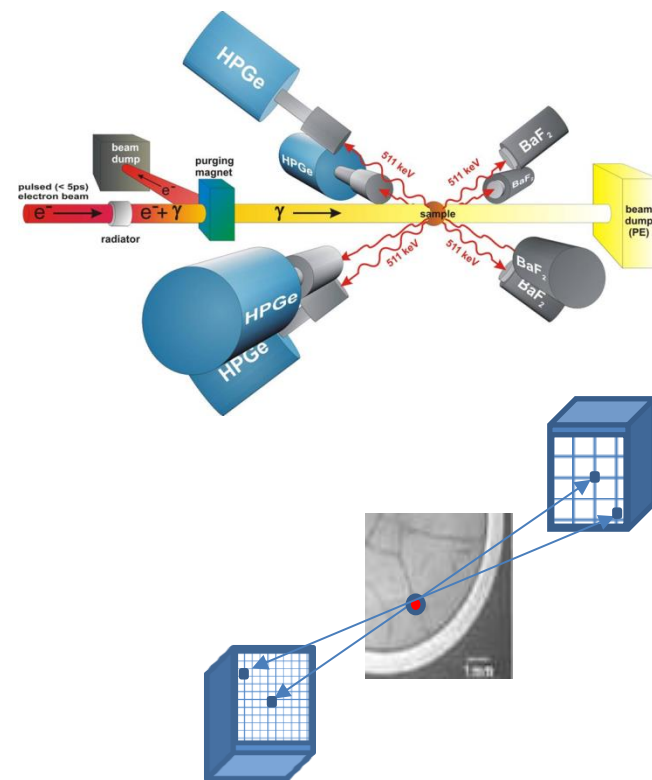
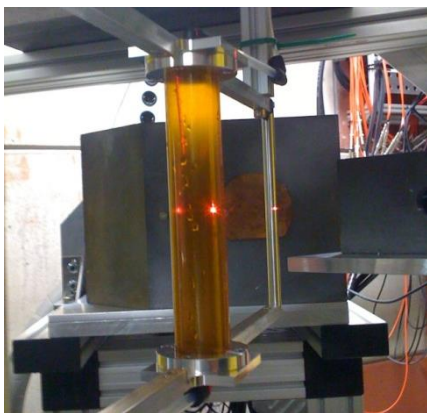
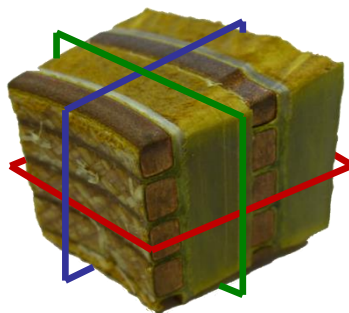
(Courtesy: J. Petzoldt, K. Römer, G. Pausch, et al.)



# Summary

Summary:

- Accelerator-driven positron production
- Annihilation lifetime spectroscopy for fluids, reactor materials...
- First results for 3D tomography





Apply for beam time: deadlines 1<sup>st</sup>  
weeks in May and November

# The team

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A.W.

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C. Kessler

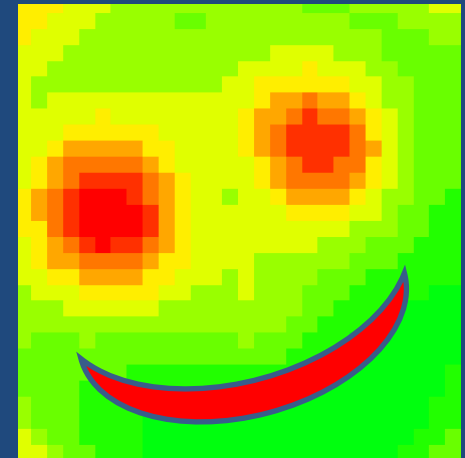
P. Fobe

and all the collaborators

A.Ulbricht, E. Altstadt (HZDR)

S.V. Stepanov, D.S.Zvezhinskiy (ITEP, MEPhI)

*Thank you for attention*



Member of the Helmholtz Association