# Positron annihilation studies at an electron accelerator: From thin films to bulk samples and 3-D imaging

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HZDR HELMHOLTZ ZENTRUM DRESDEN ROSSENDORF

# Credits

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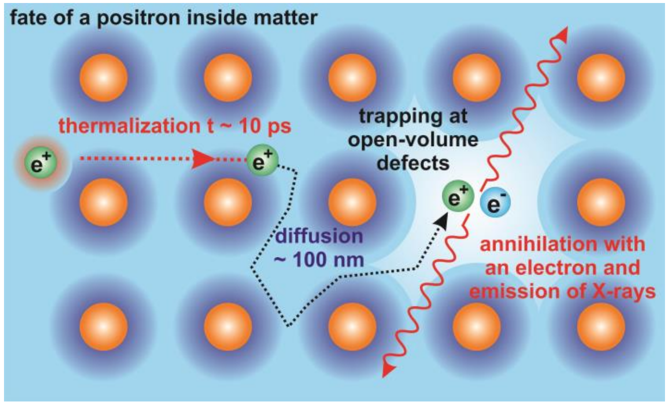
# Outline




- Motivation

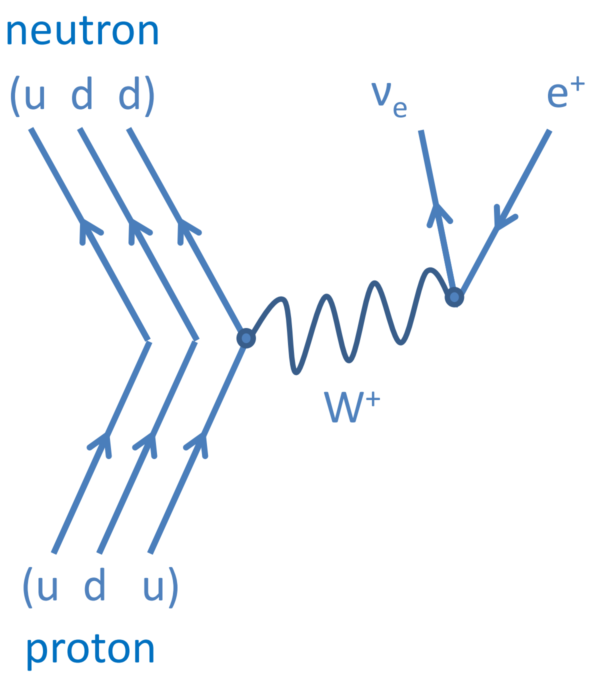
- Accelerator-based positron production and annihilation studies at a superconducting electron LINAC: What marks the difference to reactors and radio-isotope sources?

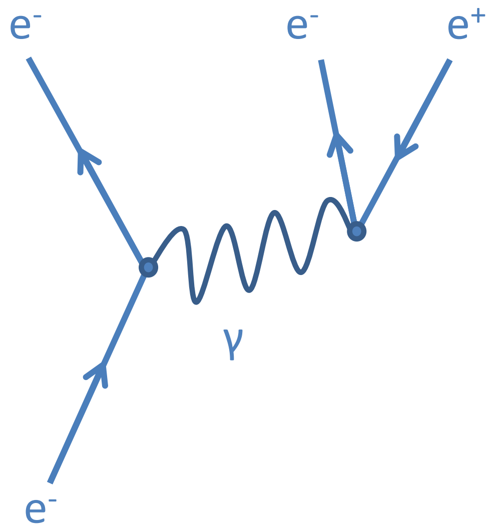
- Applying pulsed beams: positron annihilation lifetime spectroscopy at thin films, bulk materials, and fluids

- Development of a pixelated detection system for position-sensitive positron annihilation lifetime measurements and experiments with structured targets and tomographic image reconstruction

# Isotopes, reactors, accelerators

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



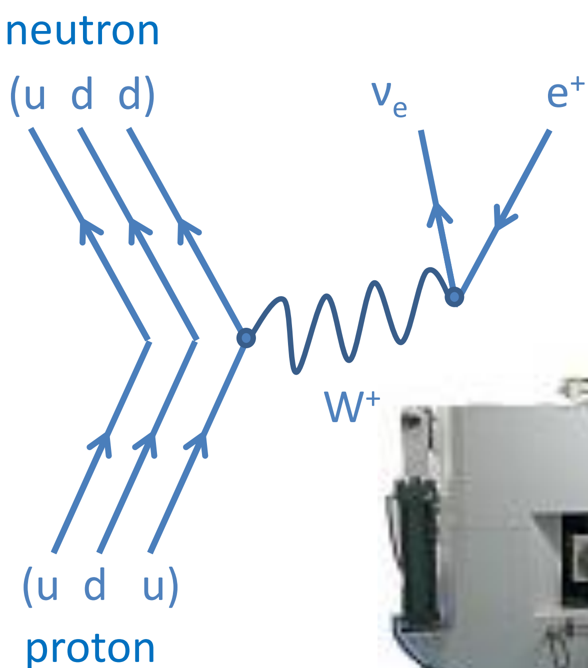




Free proton decay is forbidden by energy conservation

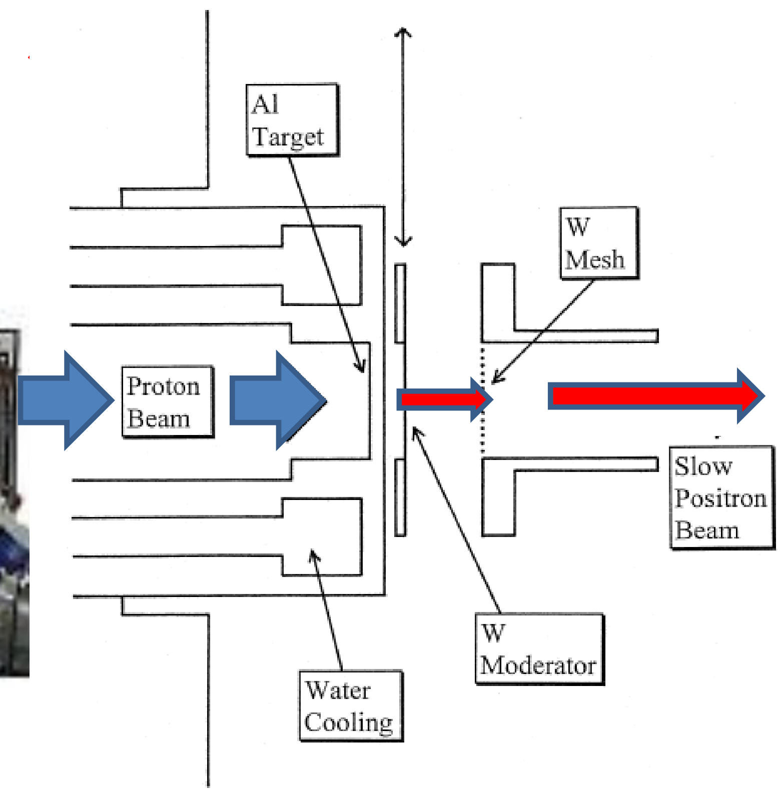
→ we need the proton inside a nucleus where it undergoes $\beta^+$-decay

# Isotopes, reactors, accelerators

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




$^{27}Al(p,n)^{27}Si(\beta^+\nu_e, 4.2\ s)\ ^{27}Al$




Sumitomi Heavy Industries Cyclotron
18 MeV protons, 50 µA beam current

# Isotopes, reactors, accelerators

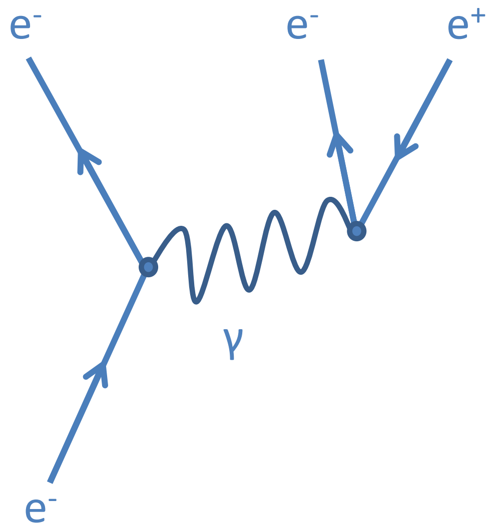
Production of positrons in weak (mediated by W's) or electromagnetic interactions (mediated by photons)

Use intense source of photons for pair production

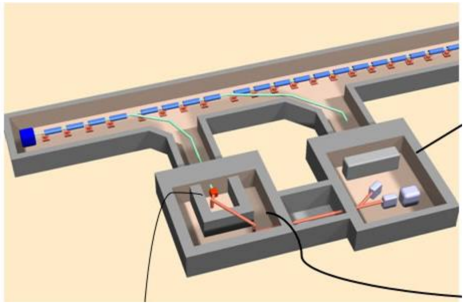
→ Capture-neutron gamma-rays from reactor

$^{113}Cd(n,\gamma)^{114}Cd$

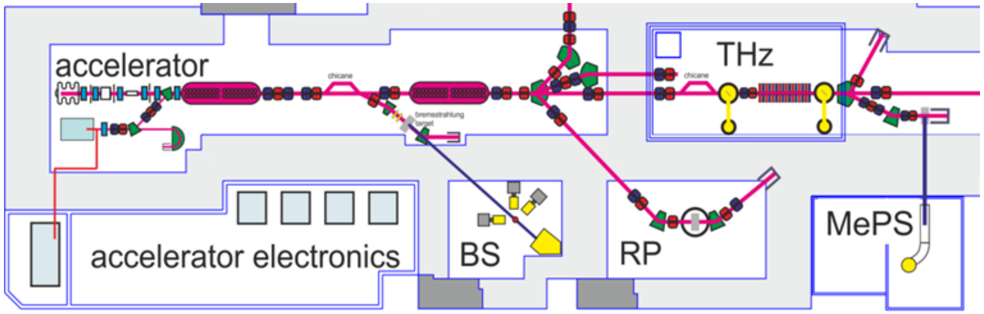
FRMII Munich




→ Bremsstrahlung from electron accelerators



AIST, Tsukuba, Japan




ELBE, Dresden

DRESDEN concept HZDR

# Positrons from accelerators

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

A) normal conducting LINAC (AIST)
$E \sim 50$ MeV
$I_{peak} \sim 100$ mA
$t_{bunch} \sim 1$ µs
$f_{rep} \sim 100$ Hz

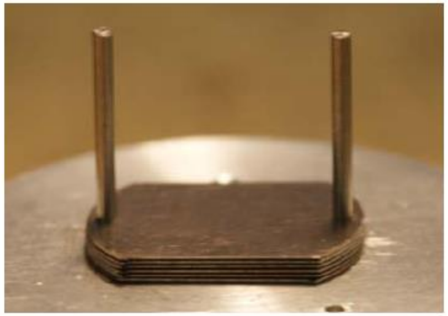
beam power 500 W

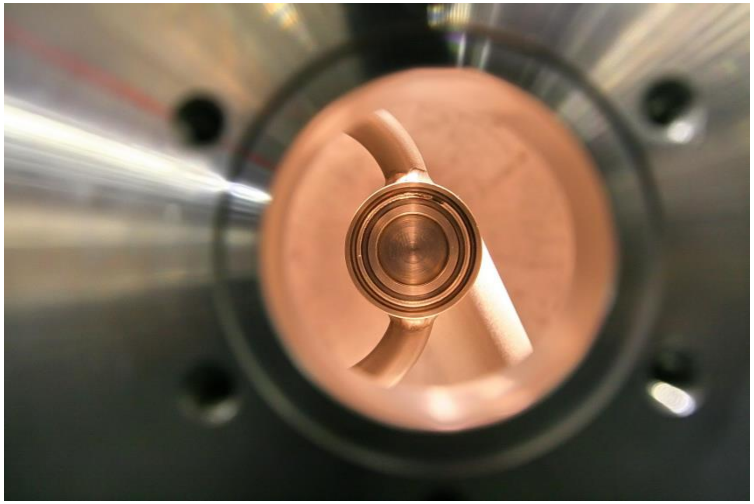
B) superconducting LINAC (HZDR)
$E \sim 50$ MeV
$I_{average} \sim 1$ mA
$f_{rep} \sim 10$ MHz

beam power 50 kW

sophisticated converter designs and heavy shielding needed

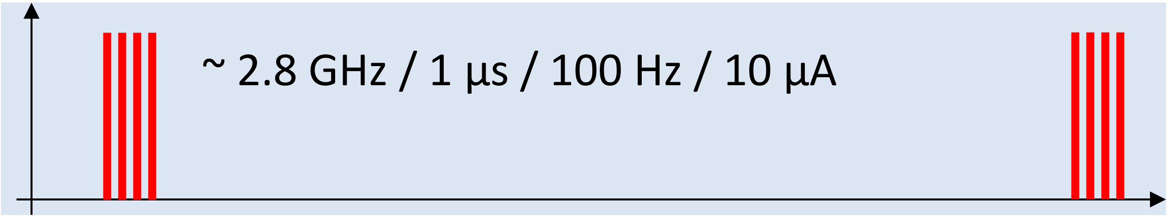
stack of 50
100 µm thick W foils



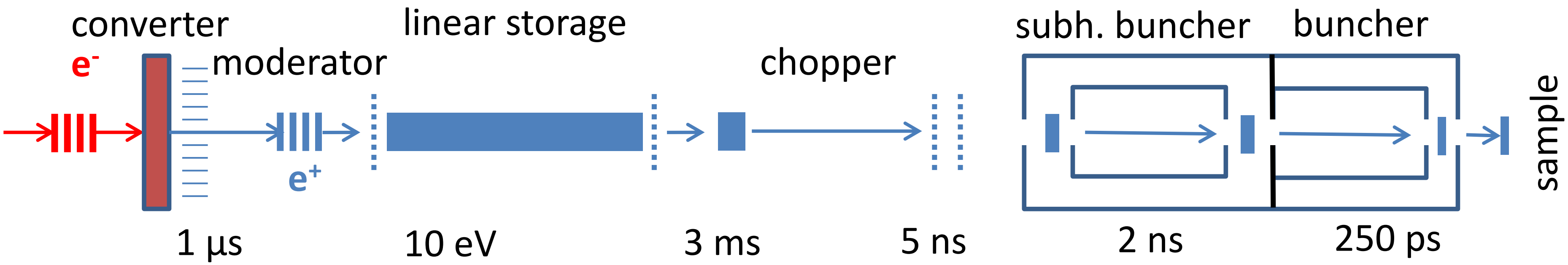


EPOS water-cooled converter

# Positrons from accelerators




**NC-LINAC** in bunched mode

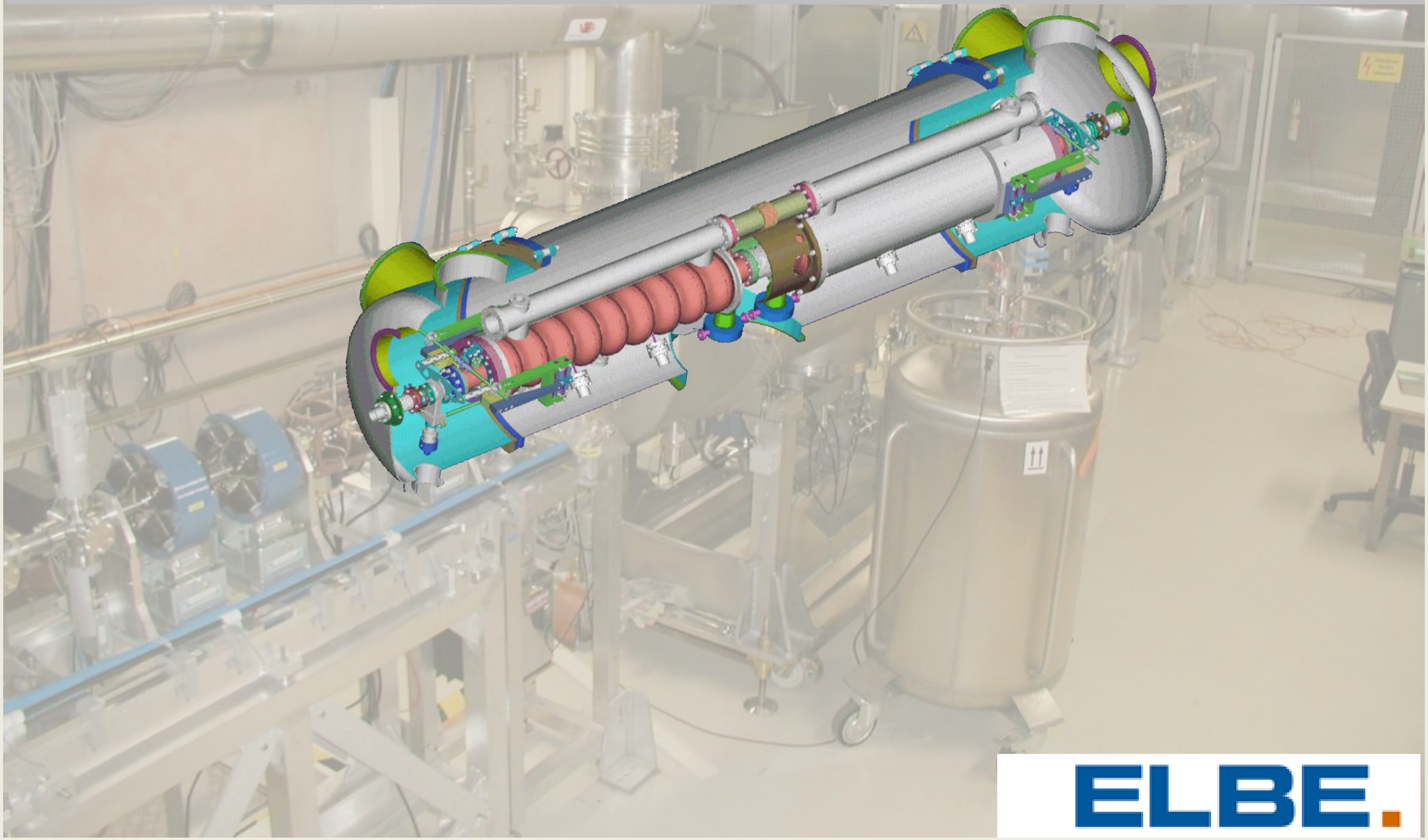


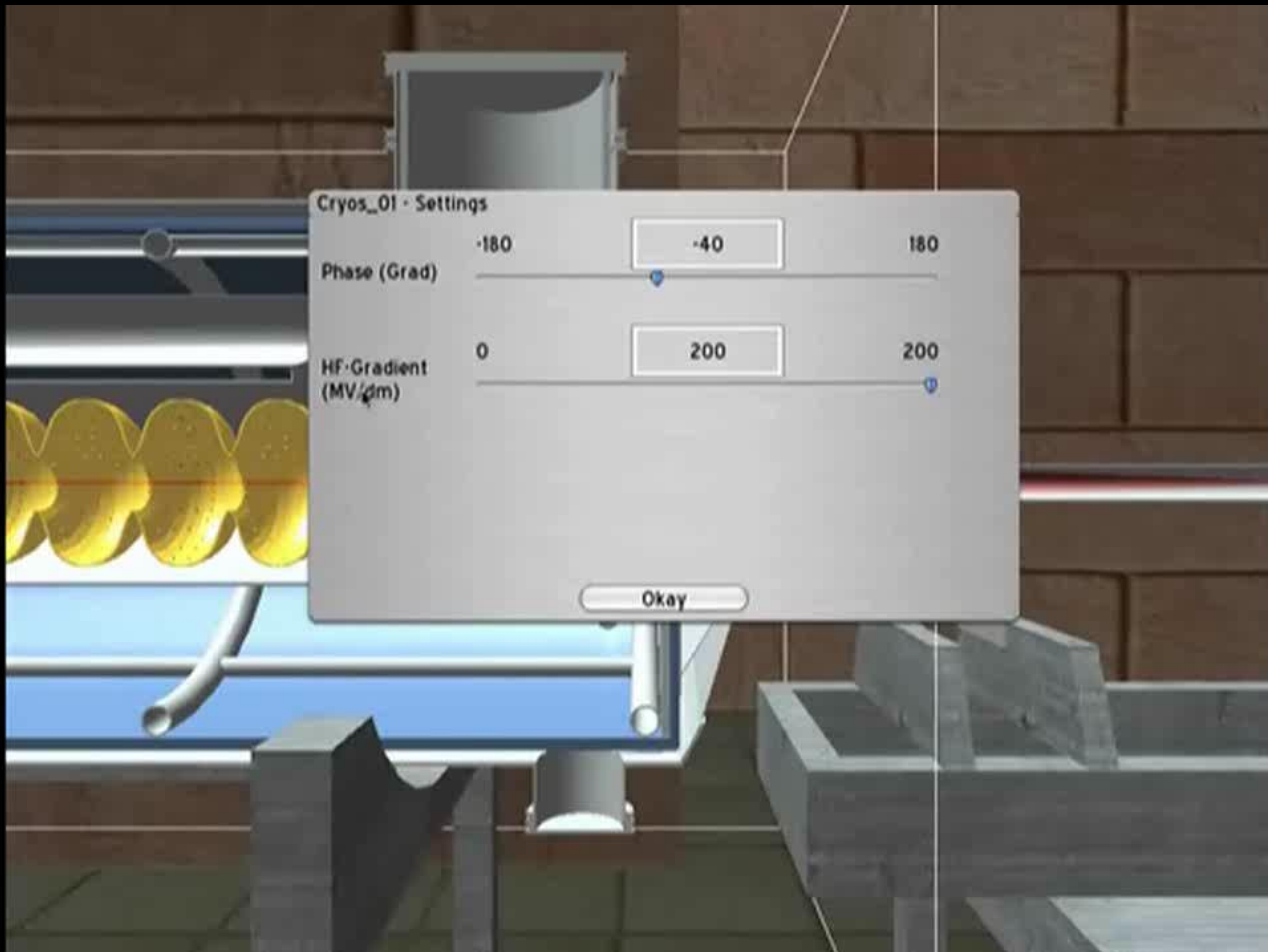




**SC-LINAC** in CW mode

# Superconducting Linear Accelerator with up to 40 MeV and 1.6 mA CW (= 64 kW power)
# SC-Structures by Cornell Univ. & Desy/TESLA

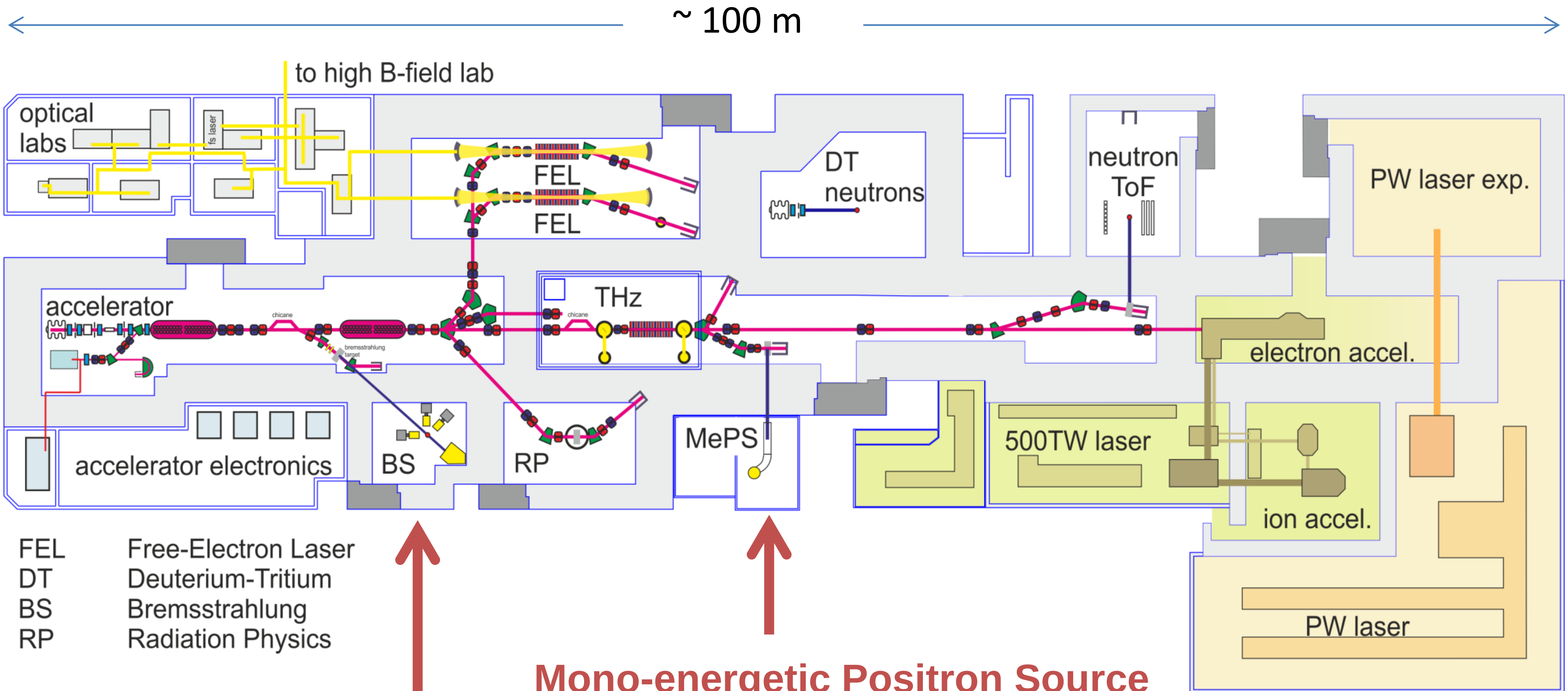



Cryos_01 - Settings
Phase (Grad)
-180
-40
180
HF-Gradient
(MV/dm)
0
200
200
Okay

ELBE.

# Facility layout

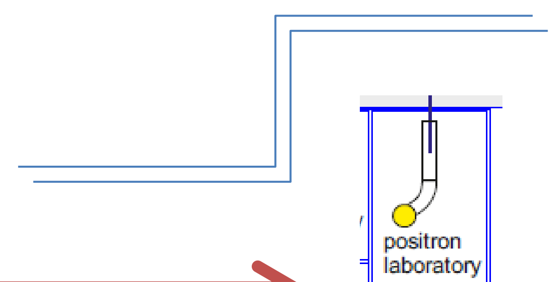
ELBE.




FEL Free-Electron Laser
DT Deuterium-Tritium
BS Bremsstrahlung
RP Radiation Physics

**Mono-energetic Positron Source**
**< 30 keV beam**

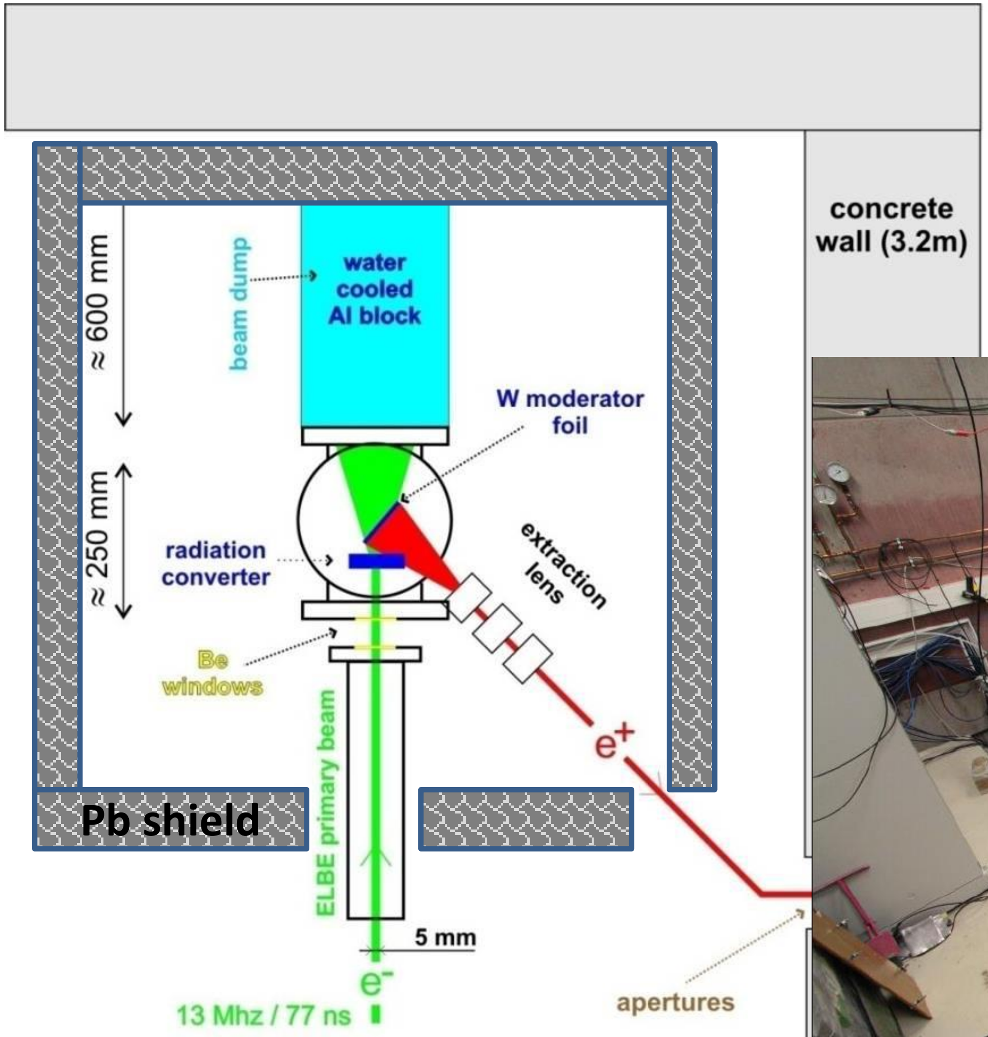
**Gamma-induced Positron Source**

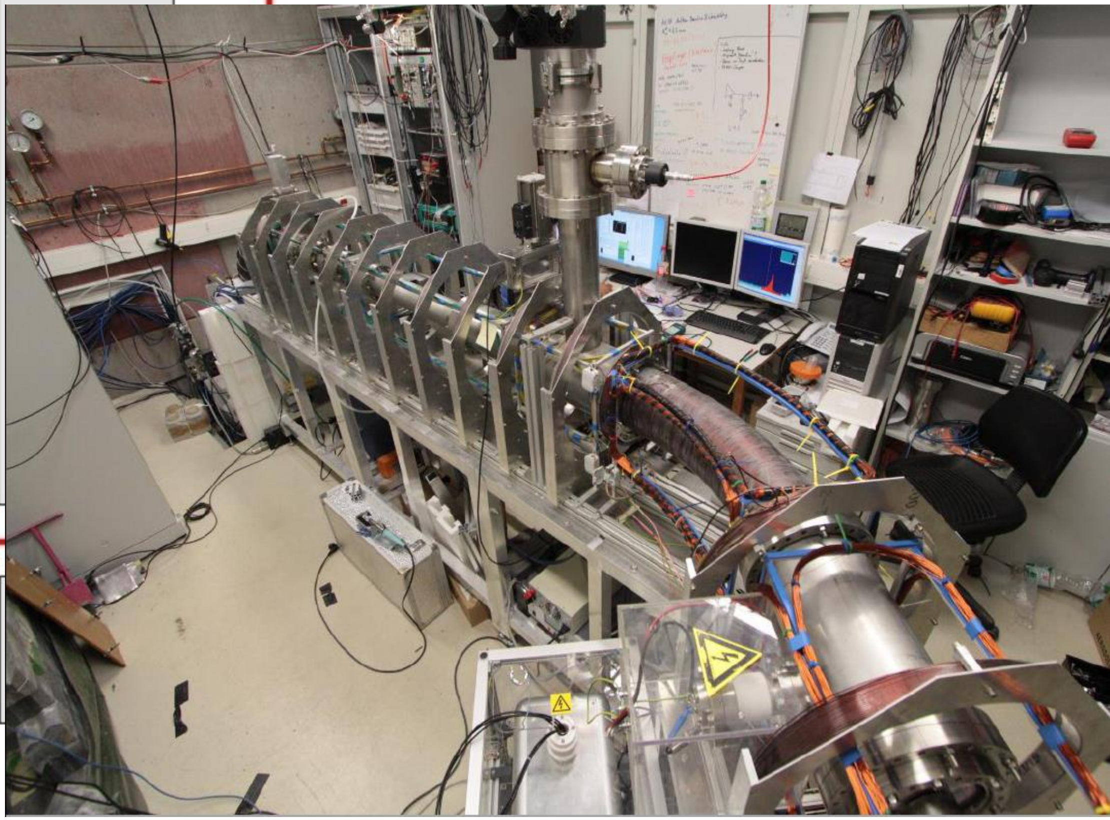



**Conventional Positron Source**
**$^{22}$Na $\beta^{+}$-decay**

# MePS setup

- variable positron energy   2 - 30 keV
- **annihilation lifetime spectroscopy**
- Doppler-broadening spectroscopy with a mono-energetic positron beam
- age-momentum correlation

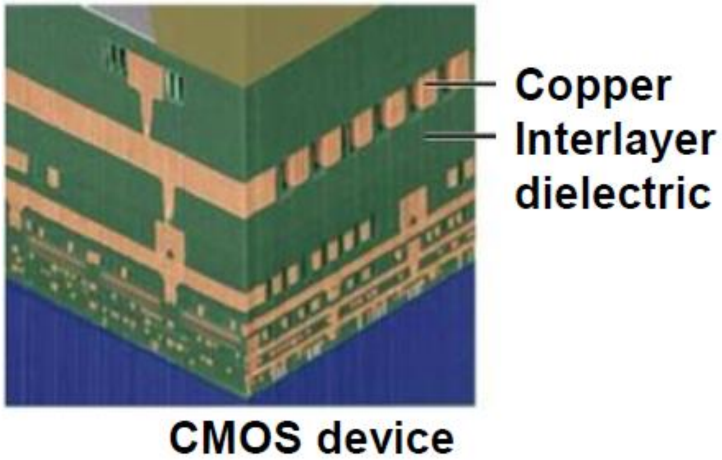


~ 100 kSv / s dose rate

# Physics with MePS

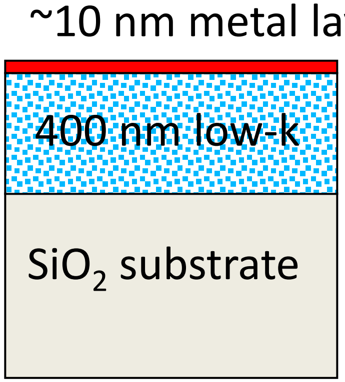
- Low permittivity materials for fast switching semiconductor devices (collab. with Fraunhofer ENAS / Chemnitz N. Ahner, R. Ecke)

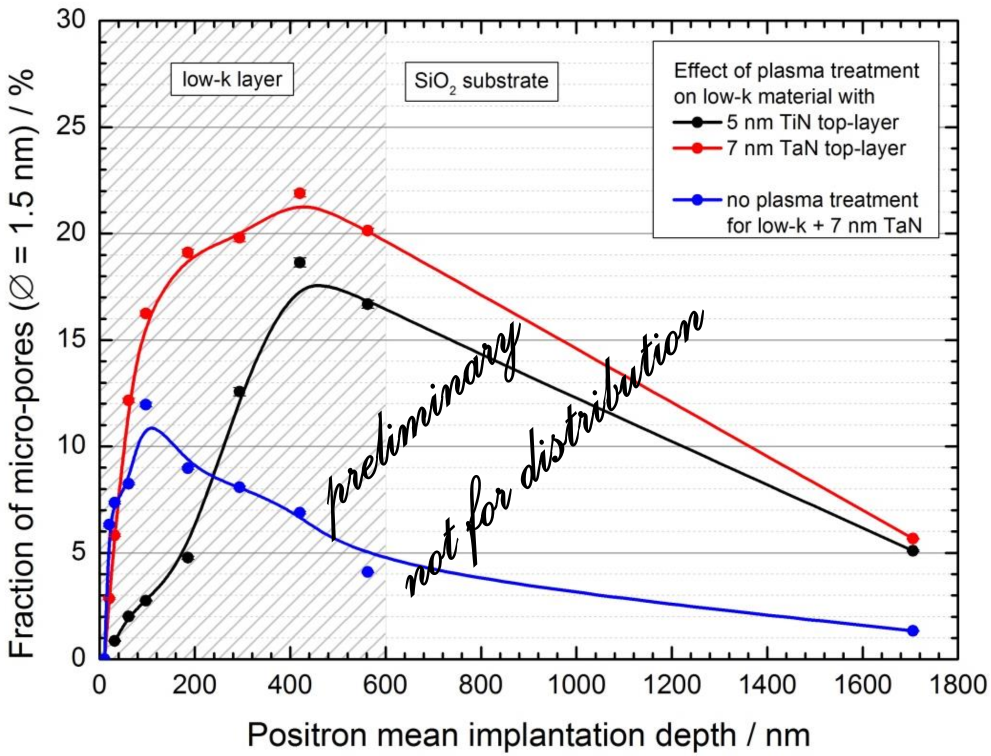



**low-k materials**

- scaling of microelectronic devices
  -> thinner insulating barriers
- materials with smaller permittivity than bulk $SiO_2$ ($\varepsilon = 3.9$) are needed
- reduce material density
  -> generate free volume

→ **microporous layers with nm-sized pores**

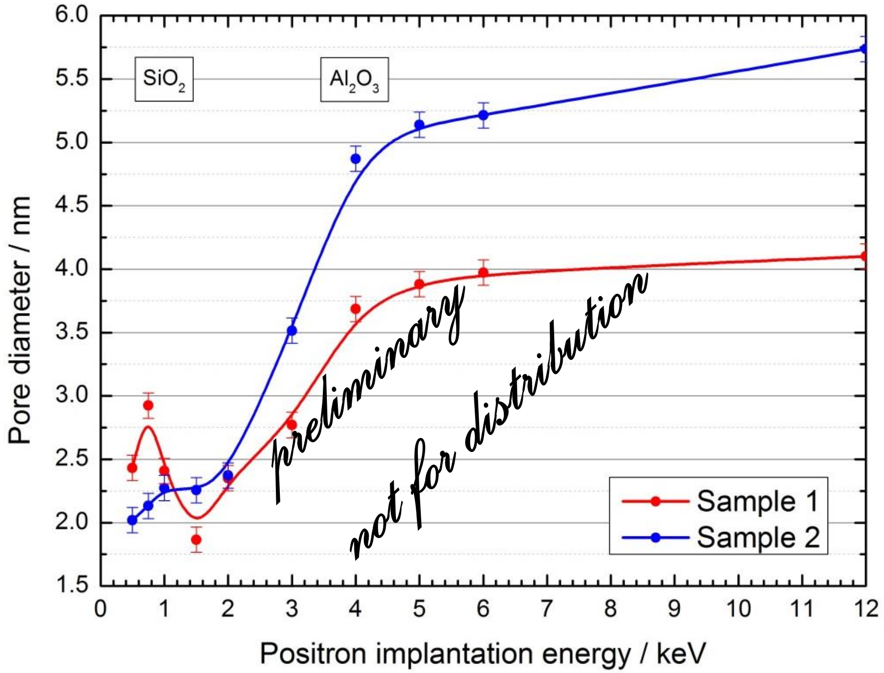


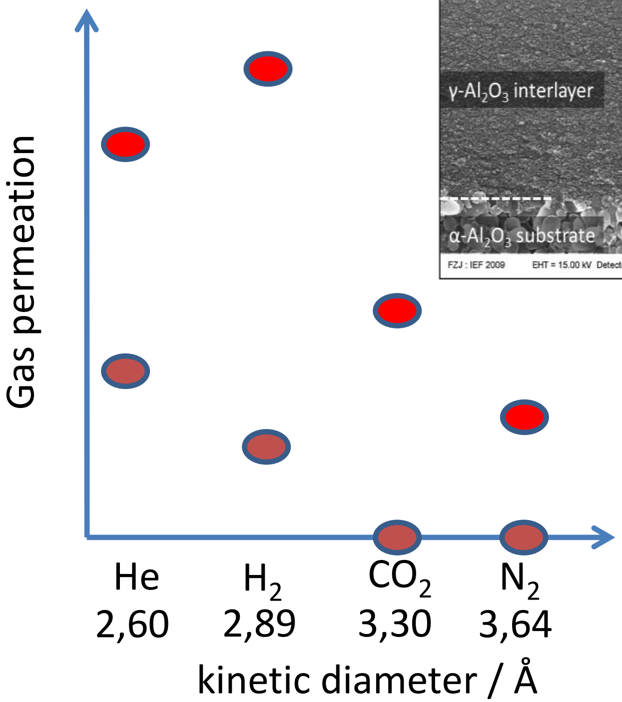



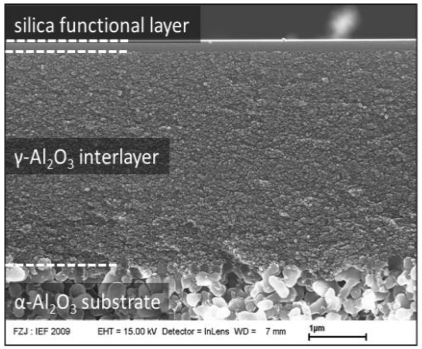
# Physics with MePS

- Microporous silica-based membranes for gas separation ($CO_2$) in future power plants w / FZ Jülich

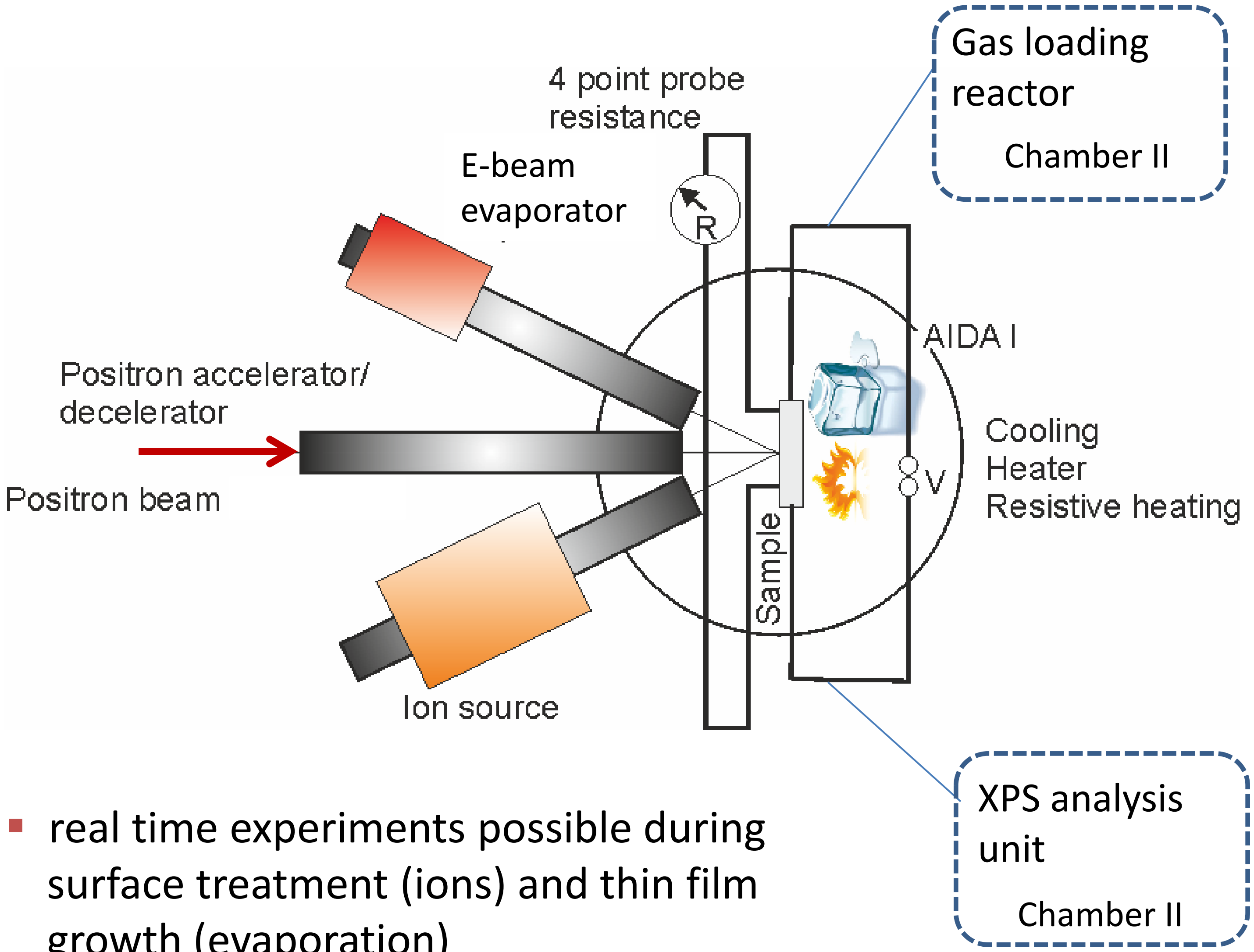

- how does the depth distribution of micropores (fabrication process) change the gas permission through the functional silica layer?



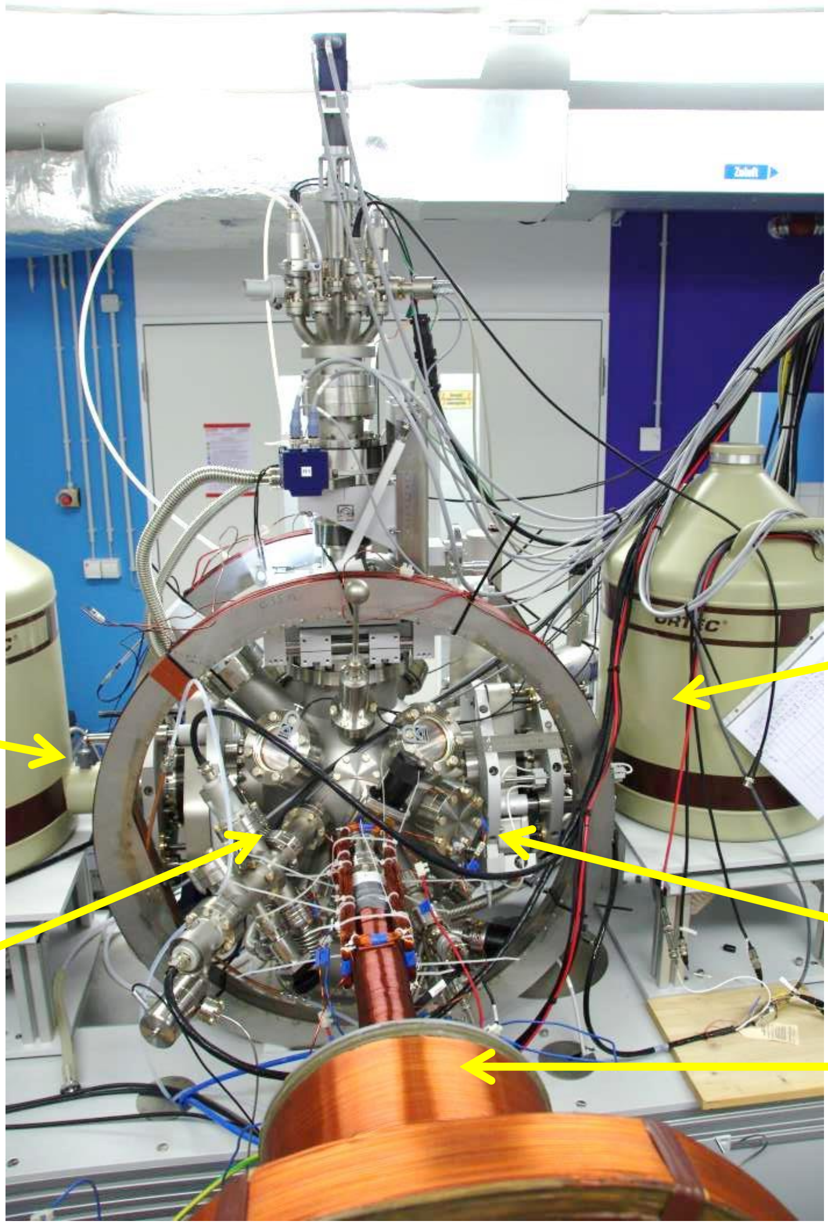
# Apparatus for In-situ Defect Analysis: AIDA




**Helmholtz Energy Materials Characterization Platform (2012 – 2015)**

- real time experiments possible during surface treatment (ions) and thin film growth (evaporation)
- adapts to MePS and CoPS
- Talks: W. Anwand, M. O. Liedke, O. Yildirim MI 2.8

# Apparatus for In-situ Defect Analysis: AIDA



Germanium detector

Germanium detector

sample chamber

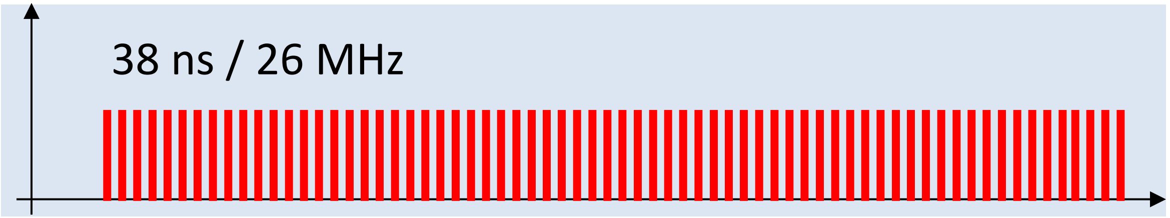
evaporator

ion source

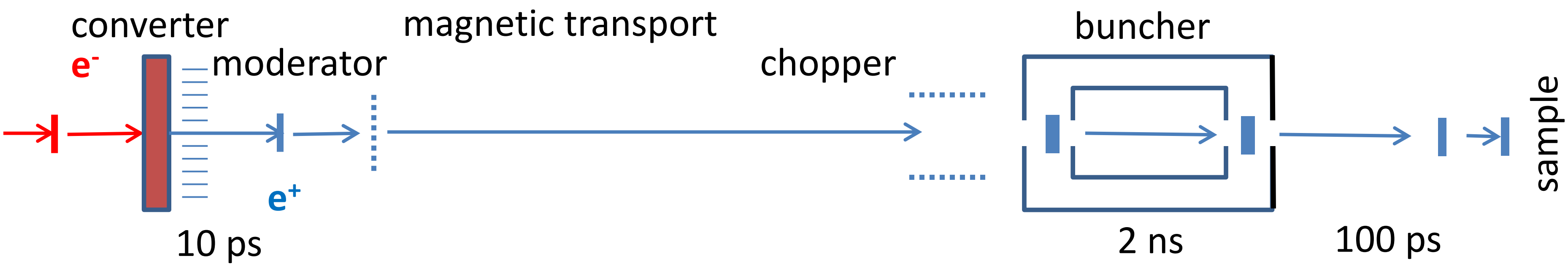
positron beam coils

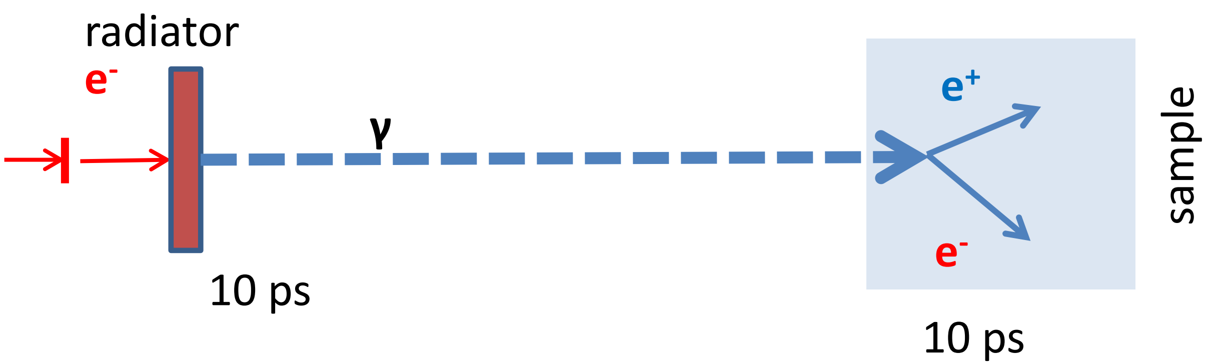
**Helmholtz Energy Materials Characterization Platform (2012 – 2015)**

# What about bulk materials, fluids, gases …?




**SC-LINAC** in CW mode



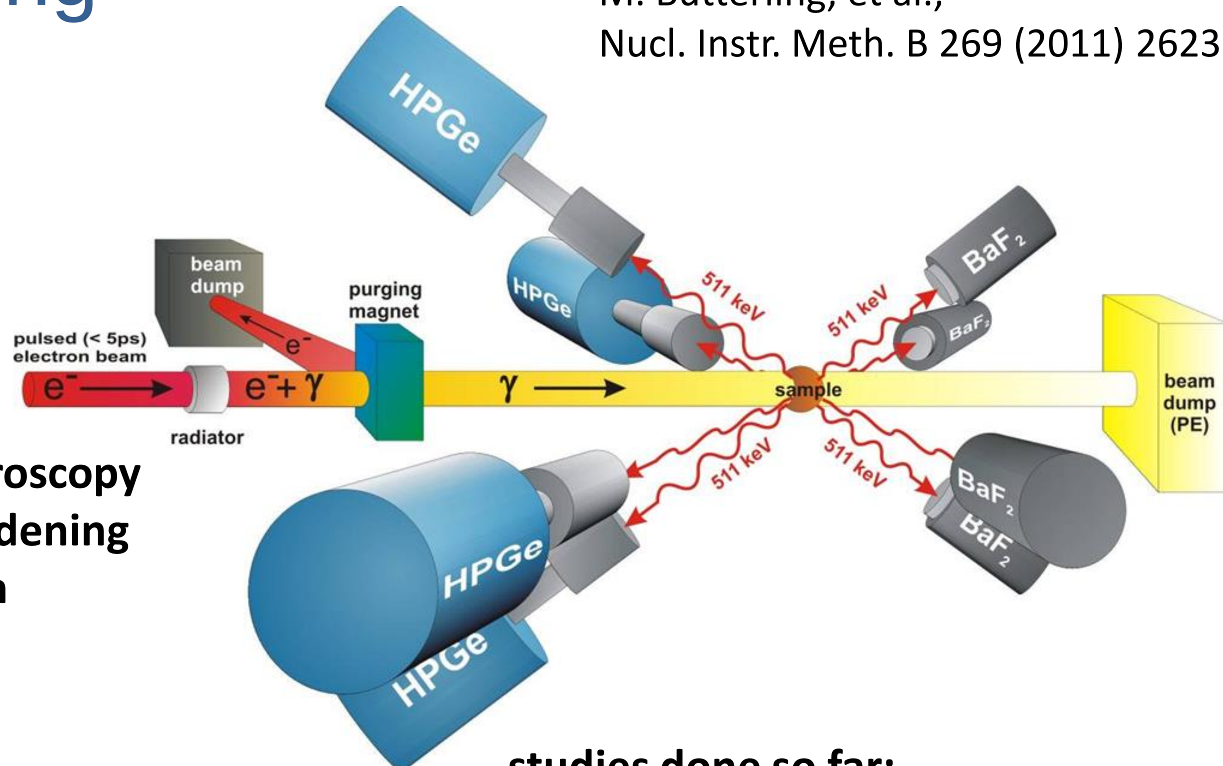

**GiPS**

The **G**amma-**i**nduced **P**ositron annihilation **S**pectroscopy

# Positron production using electron-bremsstrahlung

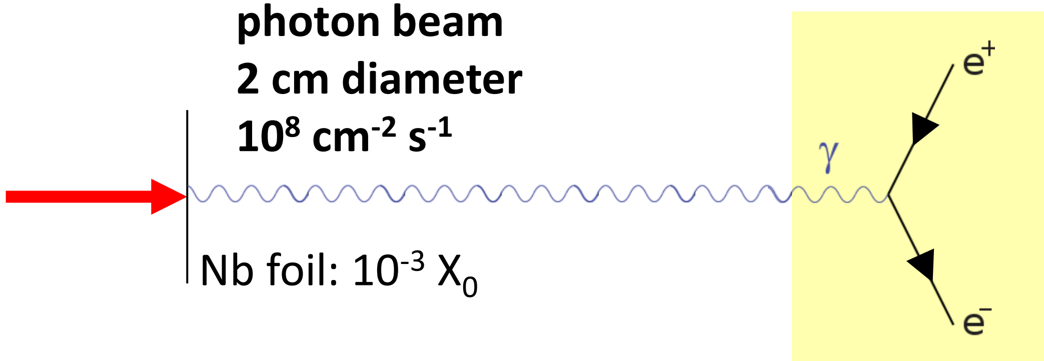
M. Butterling, et al.,
Nucl. Instr. Meth. B 269 (2011) 2623

$$E_e = 15\,\mathrm{MeV}$$
$$I_e = 900\,\mu\mathrm{A}$$
$$f = 26\,\mathrm{MHz}$$
$$\sigma_t < 10\,\mathrm{ps}$$




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

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




**studies done so far:**
- water, glycerol from 10°C to 100°C
- animal tissue
- metals and alloys
  (Talk: M. Reiner MI 2.8)
- **neutron-activated reactor materials**

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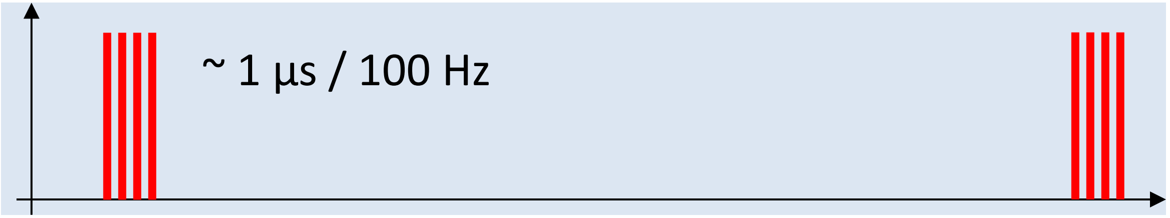
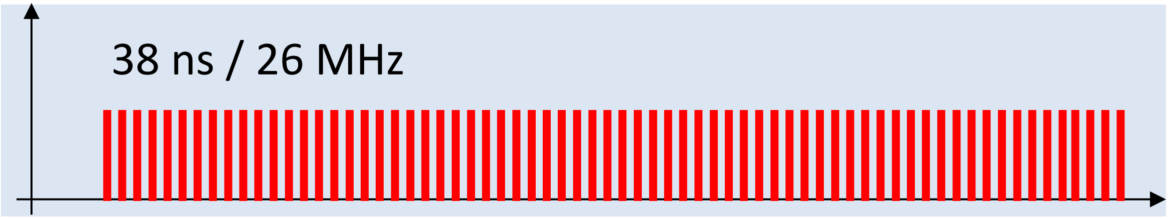
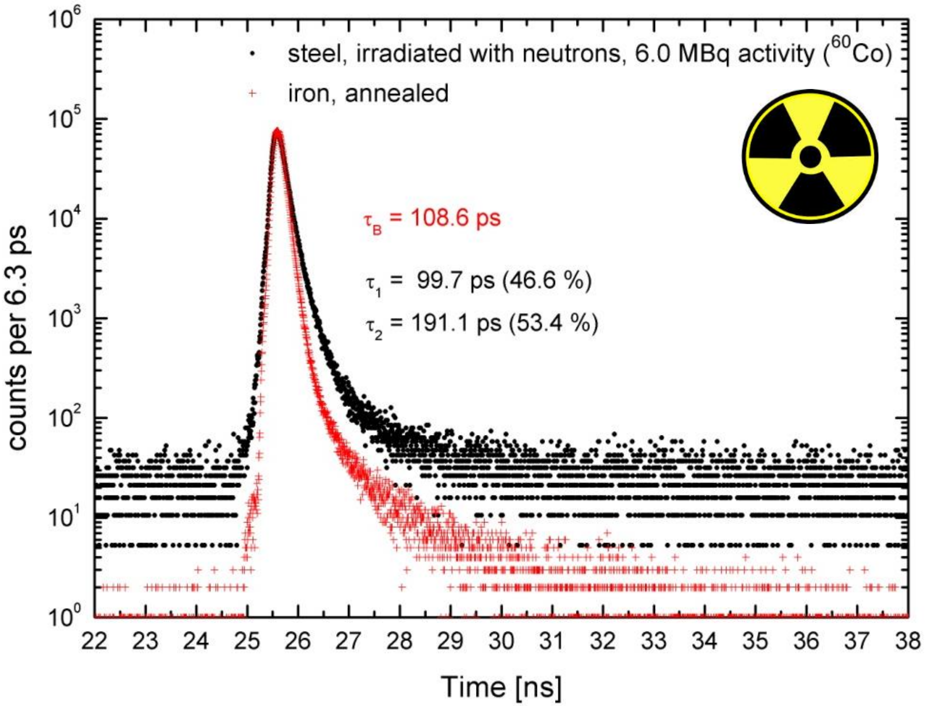
# Gamma-induced Positron Spectroscopy




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.

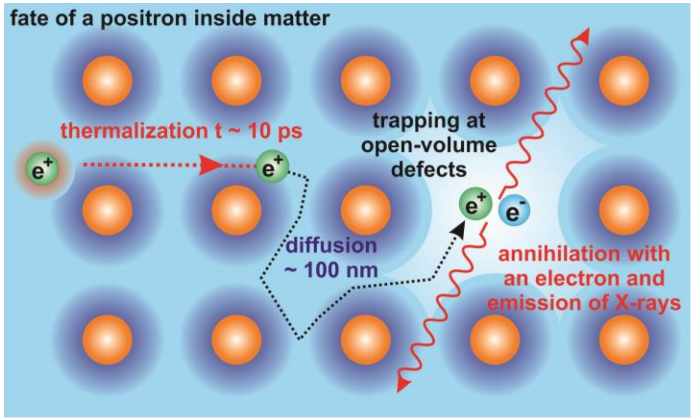
→ Long lifetimes reveal atomic defects caused by neutron-induced damage.
→ Can (and how) defects be removed by thermal annealing?

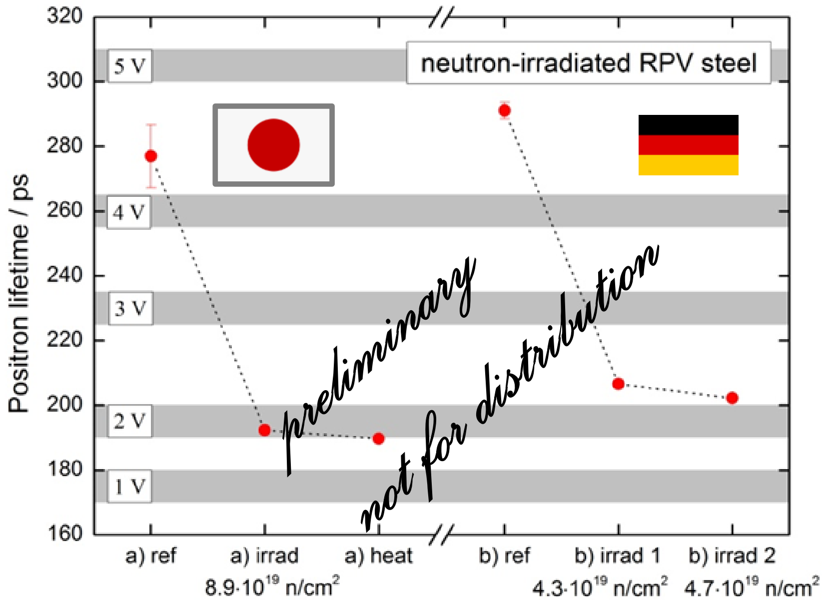
# Physics with GiPS: RPV steel

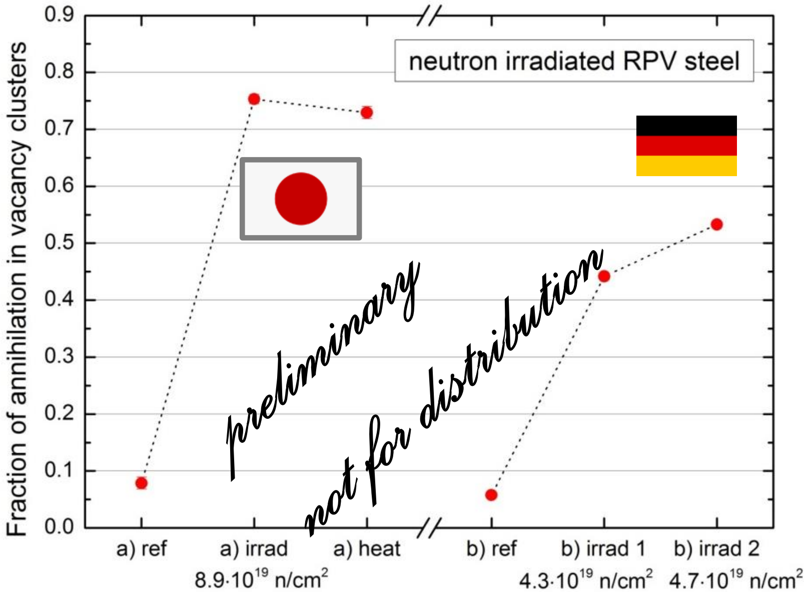
Reactor vessel steel becomes brittle due to neutron-induced defects like open-volume defects. The atomic defects act as seeds for cracks.

Collaboration with Institute for Ion Beam Physics and Materials Research.

Talk: M. Butterling MI 2.8




→ Preferential formation of double vacancies

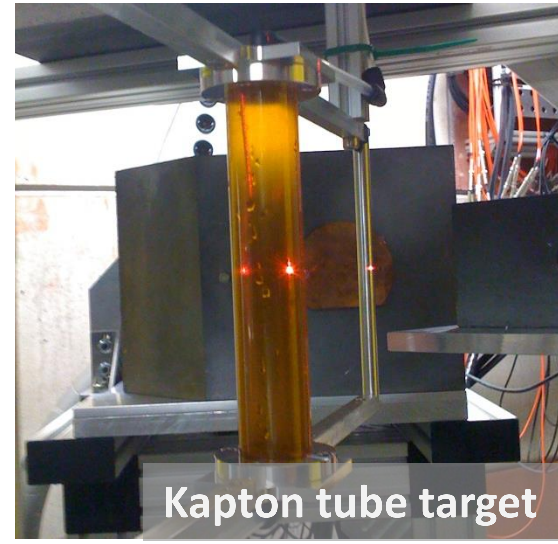
→ Thermal annealing (290°C) not sufficient to remove defects!

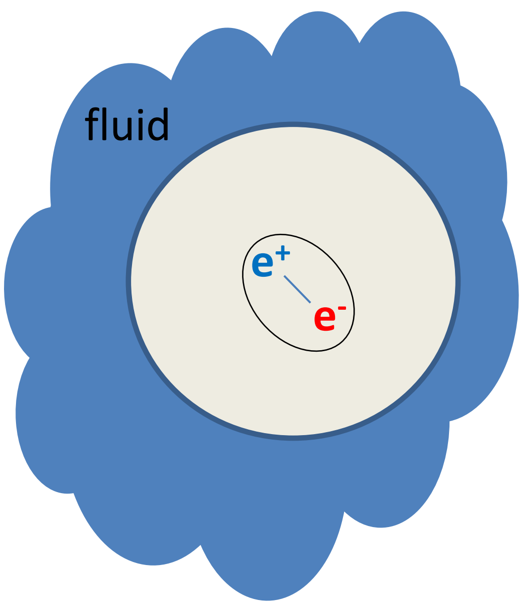
# Physics with GiPS: Fluids

Conventional lifetime measurements:
→ dissolve $^{22}$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.

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

o-Ps

142 ns

p-Ps

125 ps

R.A. Ferell, *Phys. Rev.*, 108,167, 1957
S.J. Tao, *J. Chem. Phys.*, 56,5499, 1972
M. Eldrup *et al., Chem. Phys.*, 63,51, 1981

# Physics with GiPS: Fluids

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

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

$$R(r) = R_0 j_l(kr)$$ Ansatz: spherical Bessel fct.

$$j_0(kr) = \frac{\sin kr}{kr}$$ 1st non-trivial solution

$$E_0 = \frac{h^2}{8m_{Ps}r_0^2} = \frac{\pi^2\hbar^2}{4m_e r_0^2}$$ zero-point energy

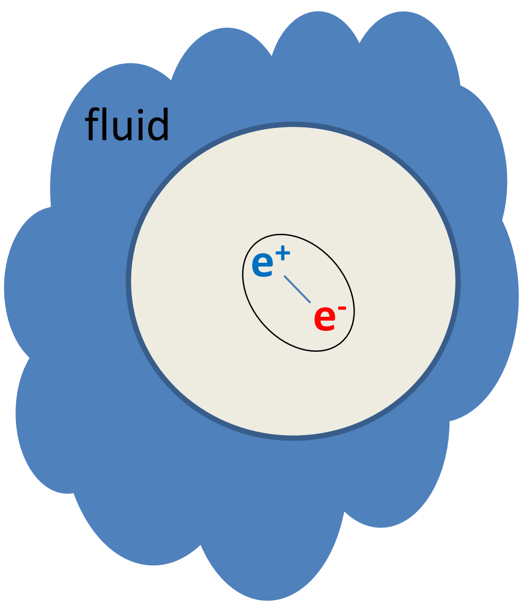
$$E_{surf} = 4\pi r_0^2\sigma$$

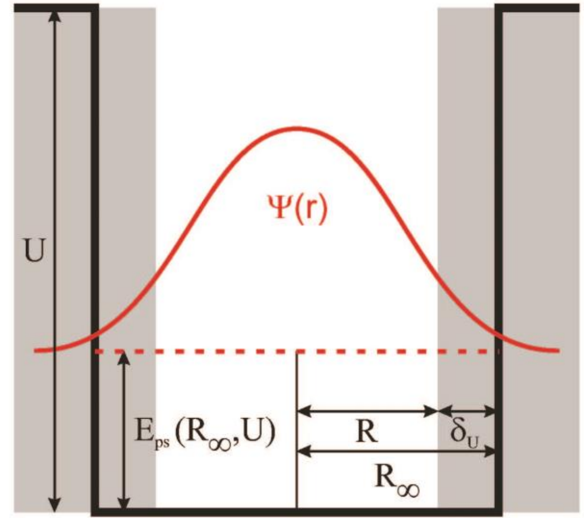
$$\frac{\partial}{\partial r_0}\left(E_0 + E_{surf}\right) = 0$$

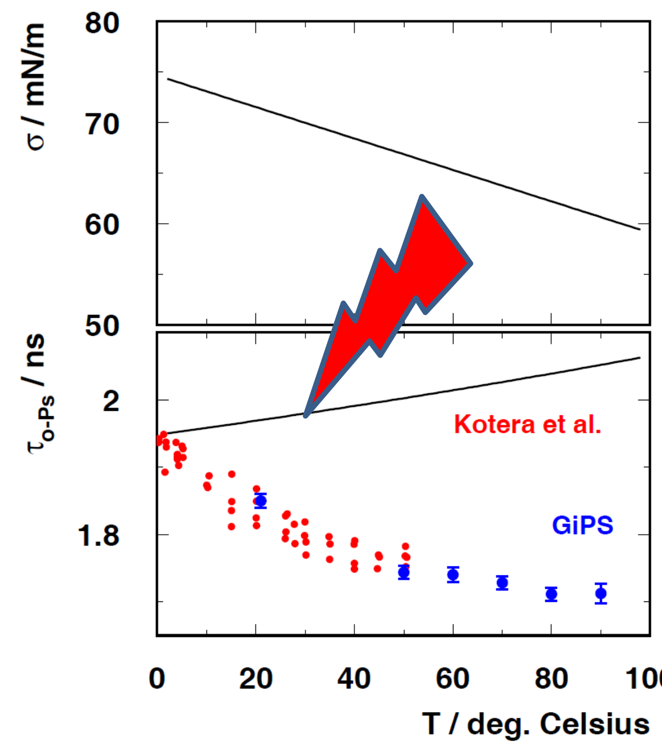
$$-\frac{\pi^2\hbar^2}{2m_e r_0^3} + 8\pi r_0\sigma = 0$$

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

$$a_0 = \frac{4\pi\varepsilon_0\hbar^2}{m_\mu e^2} = \frac{\hbar c}{\alpha\, m_\mu c^2} = 1.06\ \text{Å}$$

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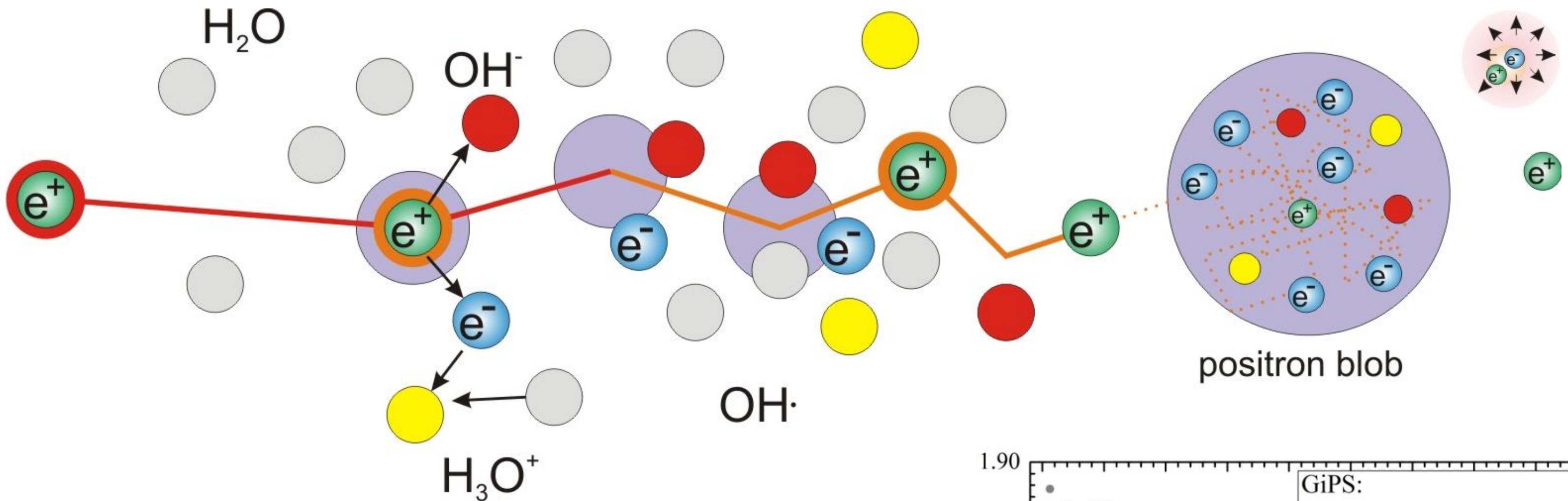
# Physics with GiPS: Positron Chemistry

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.

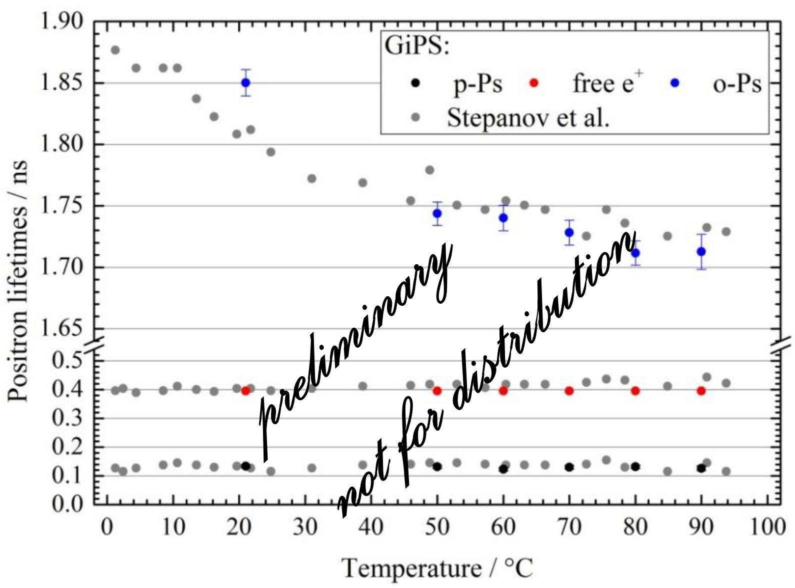



→ Radicals are positron scavengers which reduce annihilation lifetimes.

→ Extended bubble model including chemistry [S.V. Stepanov et al., Mat. Sci. Forum 607] describes data well.

→ Relevance for PET diagnostics since 2γ / 3γ ratio is affected.

→ Chemistry of radiolysis directly accessible since the probe creates the ionization itself



# Towards imaging of defects

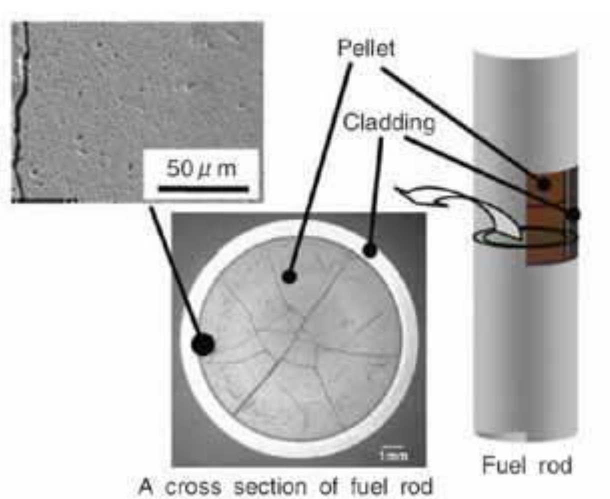
Material failures impose a significant threat to the integrity and the safety of technical systems. A thorough understanding of the microscopic origin and the development of defects requires advanced methods.

# Motivation

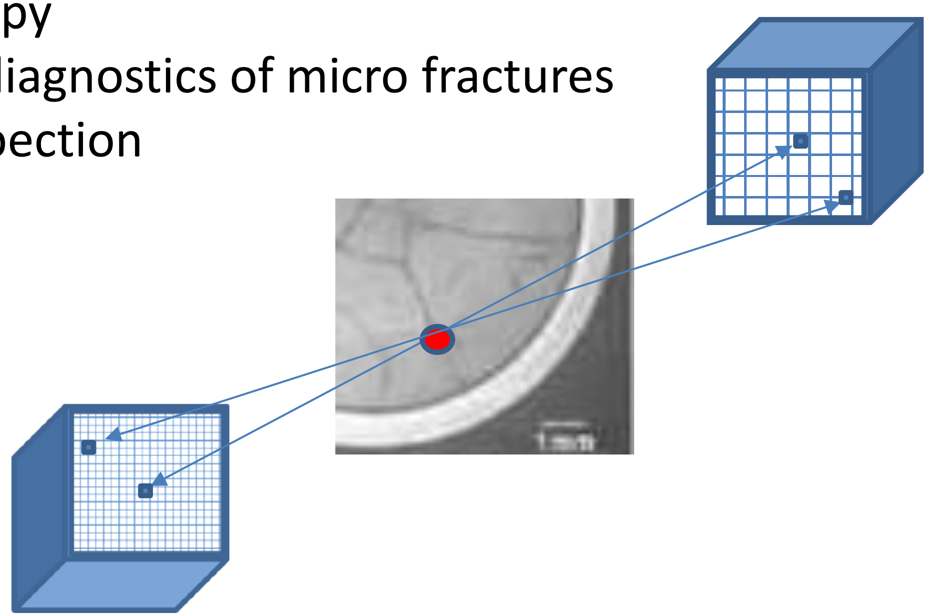
Establish a **non-destructive** and **non-intrusive** method which allows for **spatially resolved** positron-lifetime spectroscopy. Reconstruct PET-like images plus positron annihilation lifetime.

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

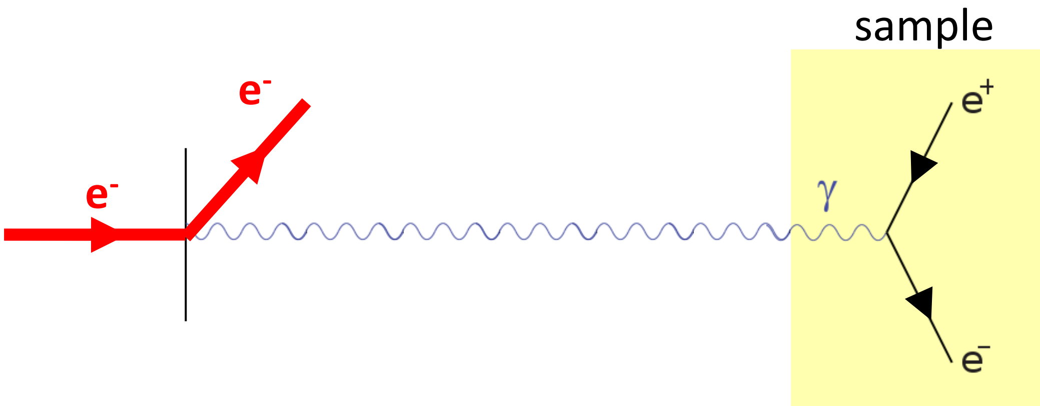



APS Physics & Society Newsletter 2011. R. Hargraves, R. Moir

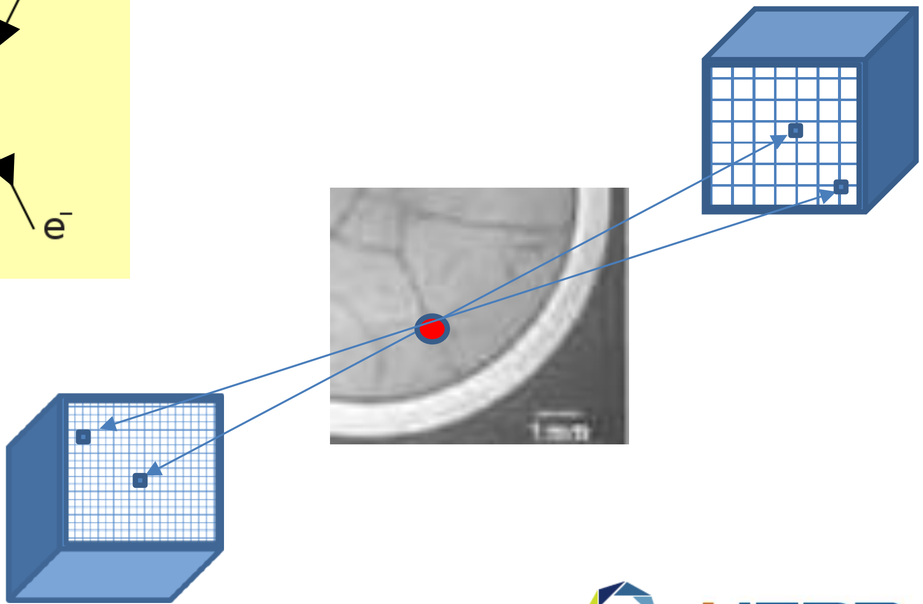


# Prerequisites

| | |
|---|---|
| • Intense source of positrons with deep penetration (cm) | ≈15 MeV X-rays |
| • Accurate time-stamping of positron creation (<10 ps) | CW LINAC |
| • Position-sensitive positron detectors (mm) | Siemens LSO PET |
| • Time-resolution for lifetime spectroscopy (~100 ps) | in-house (physics) |
| • Efficient data acquisition | in-house (physics) |
| • 3-D image reconstruction | in-house (medicine) |

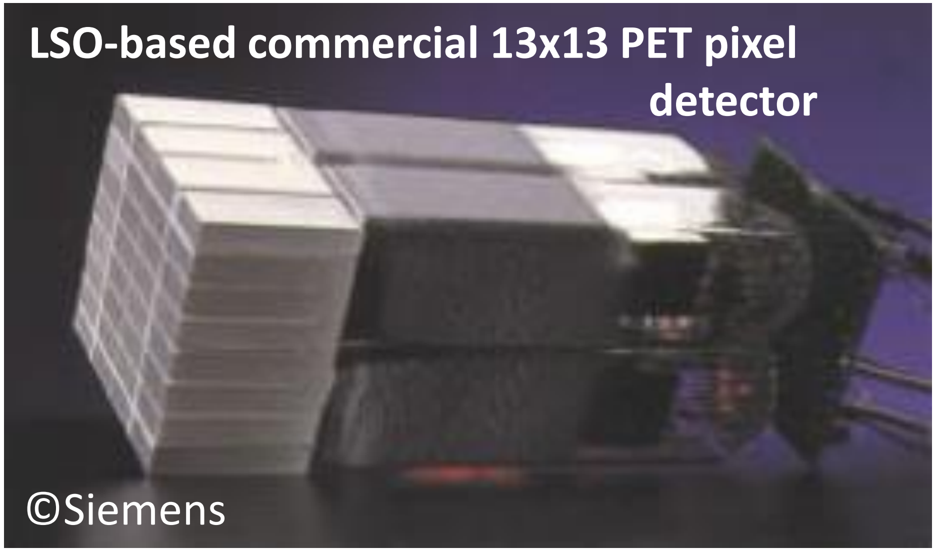


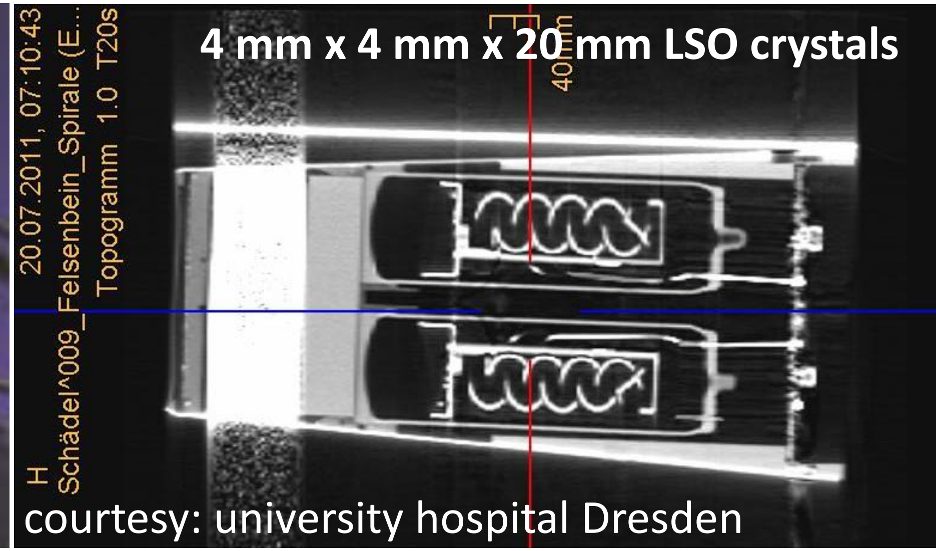

**G**amma-**i**nduced **P**ositron **S**pectroscopy



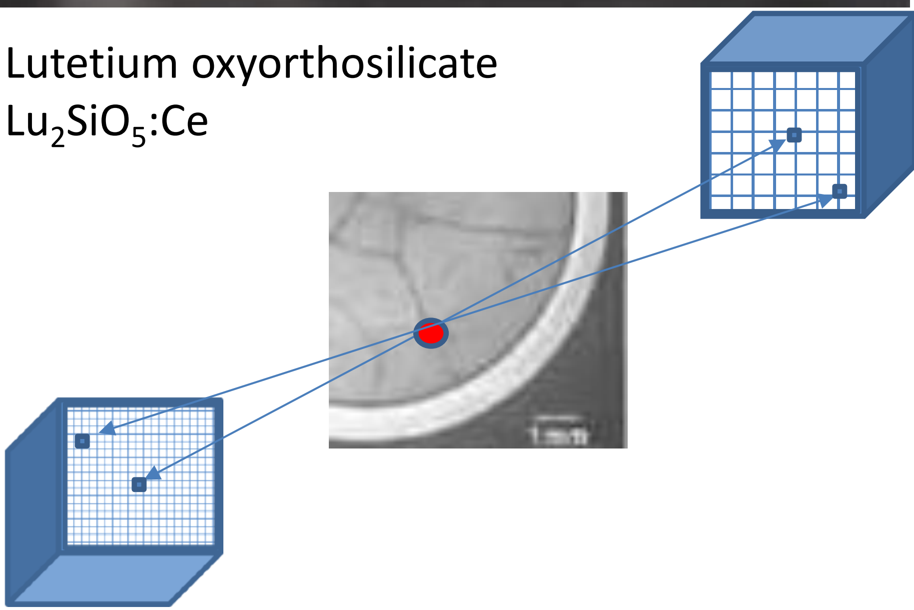
# Towards 2/3-D positron lifetime tomography

- Two position-sensitive photon detectors with 169 elements each



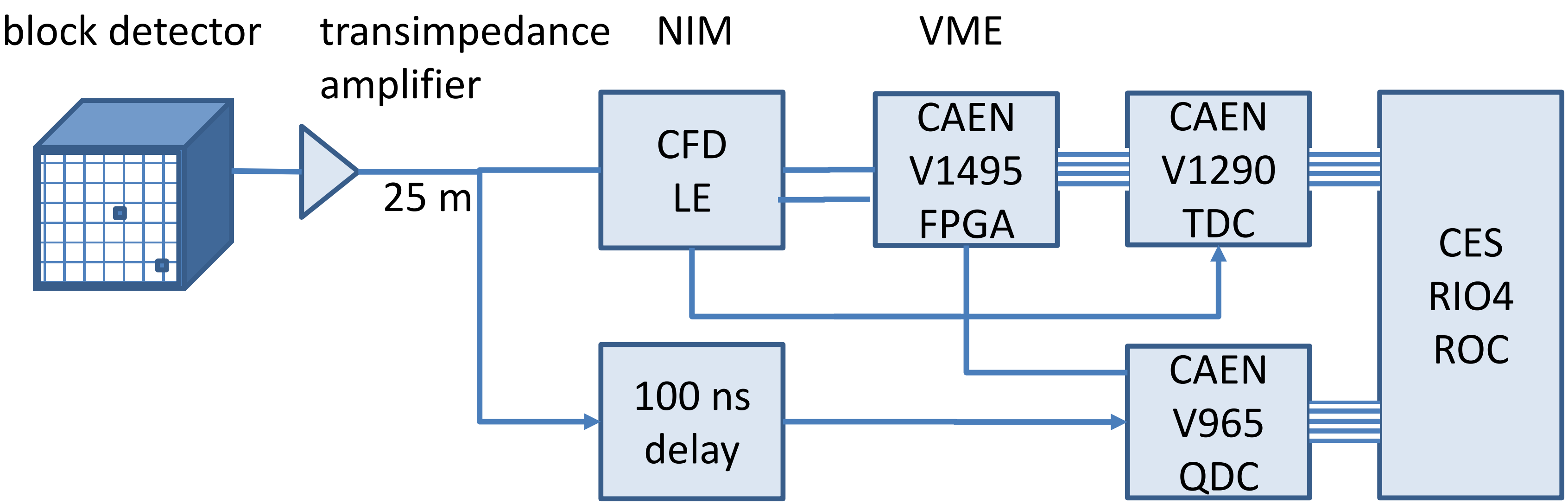



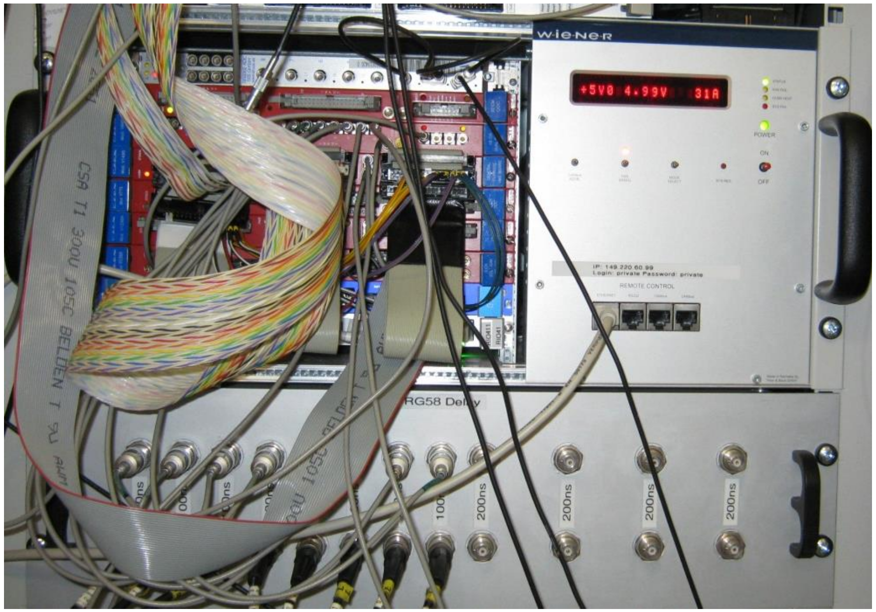

Lutetium oxyorthosilicate
$Lu_2SiO_5$:Ce




- 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

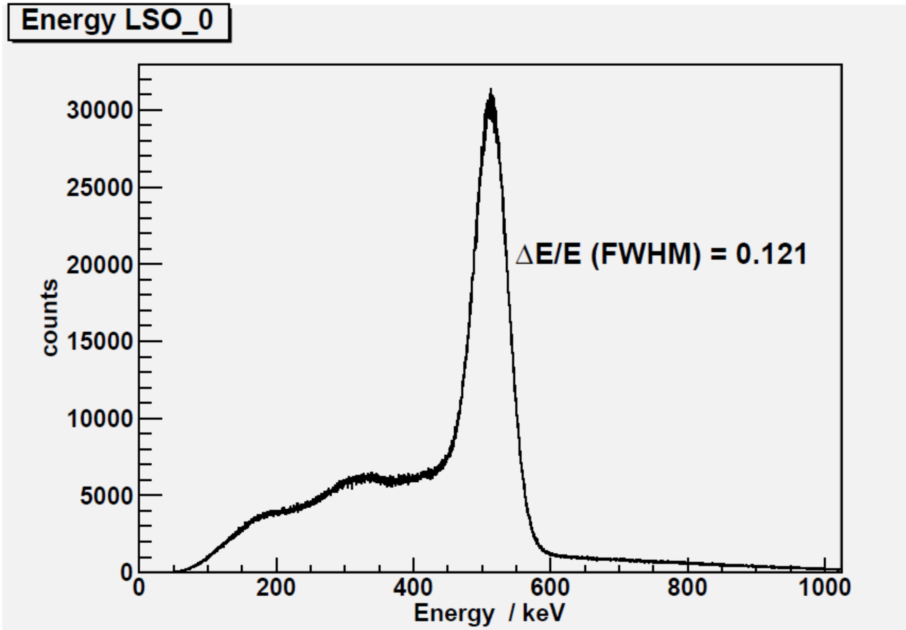
# Electronics (VME)

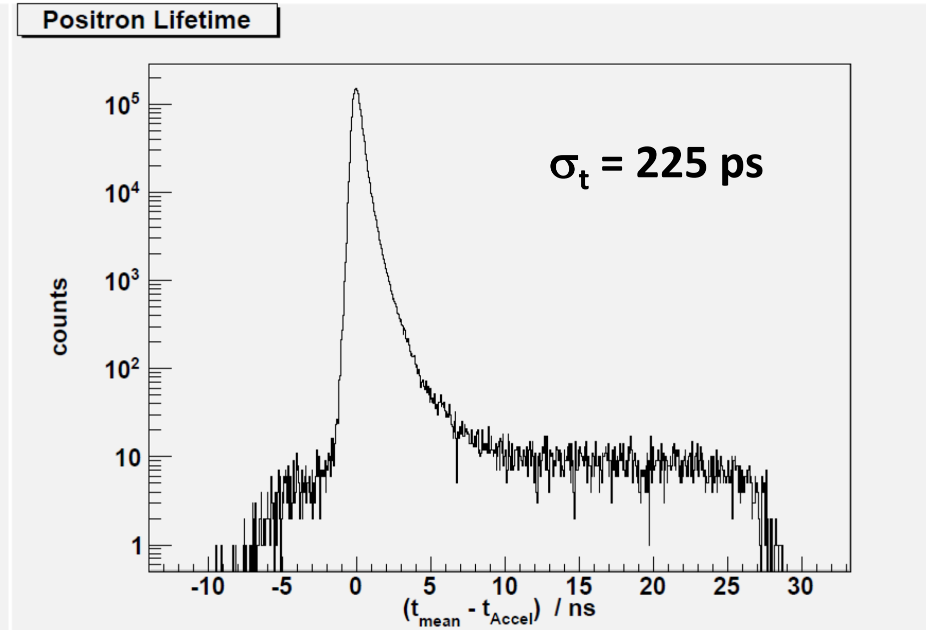


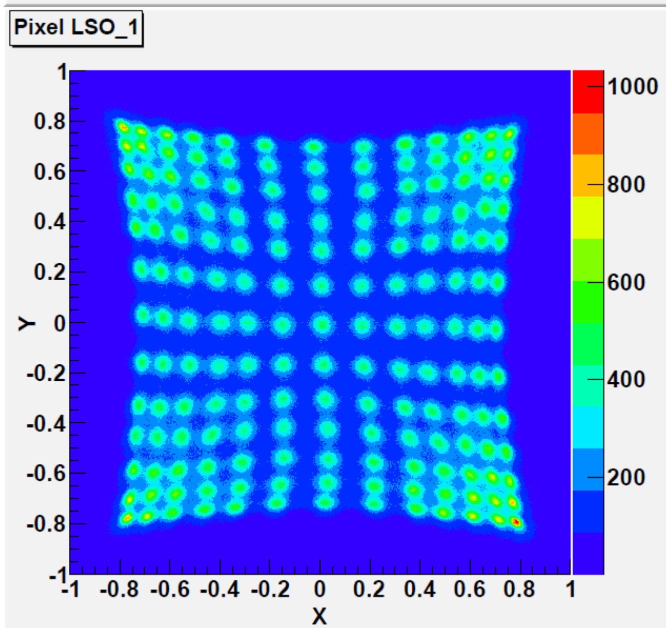



Multi-hit and multi-event buffered readout in VME block mode and readout with 10 µs dead time for 36 channels (QDC & TDC) per event.
Throughput is about 10 MB/s sustained.
Data acquisition and analysis framework using Multiple-Branch System MBS by Helmholtz-Center for Heavy Ion Research (GSI).

# Calibrations



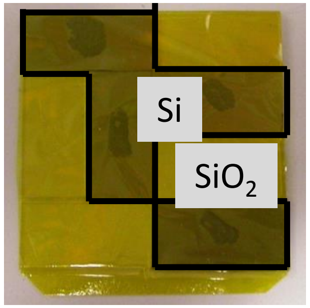

$$E = E_1 + E_2 + E_3 + E_4$$

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

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

Calibration done using 7 cm x 7 cm aqueous $^{18}$F source w/ 200 MBq ($T_{½} \approx 2$ h) produced in-house.

# Sample cases

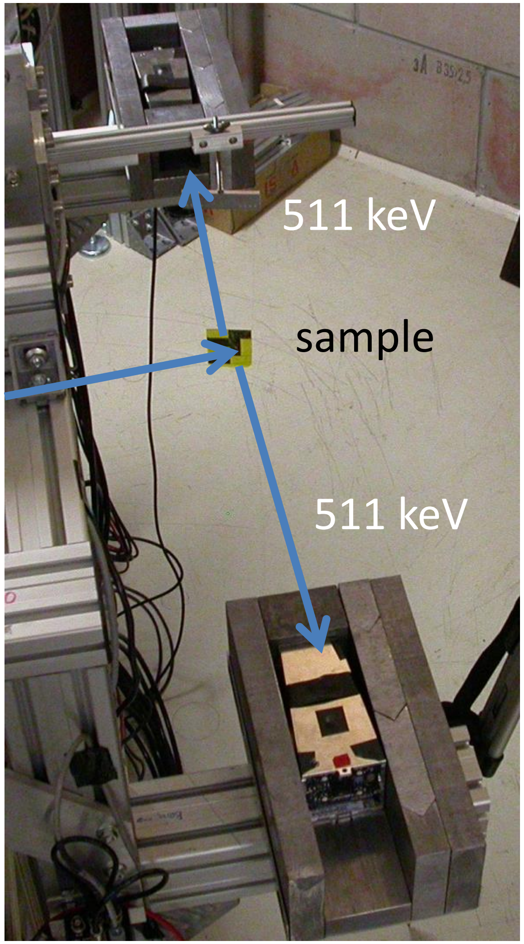


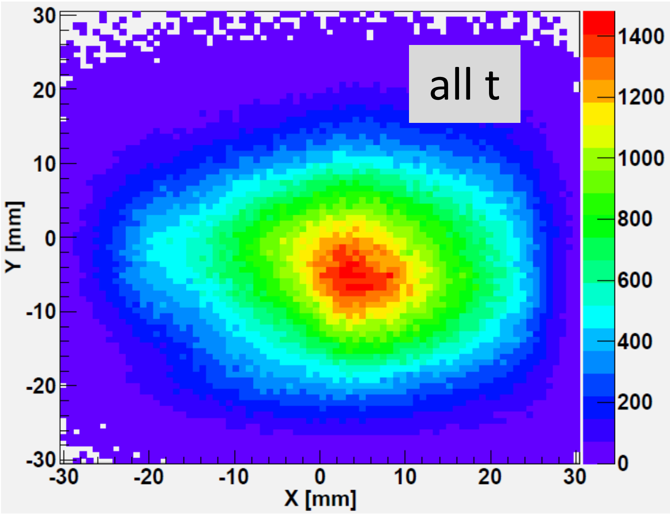

Proof of principle, first test
Simple 2D target
→ proof of principle
→ simple back-projection method

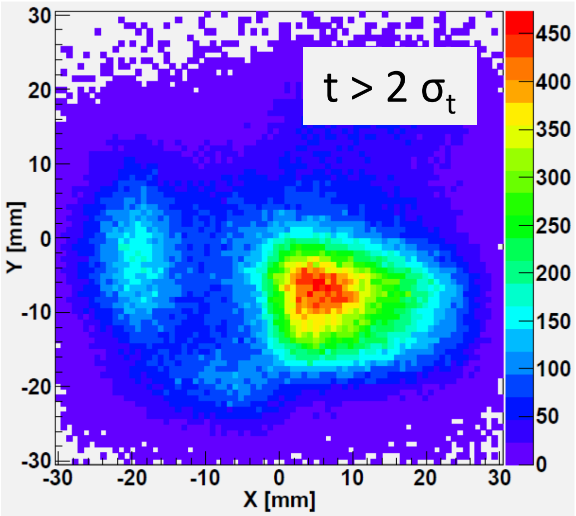
3D target
→ Reconstruction of data as a function of life time

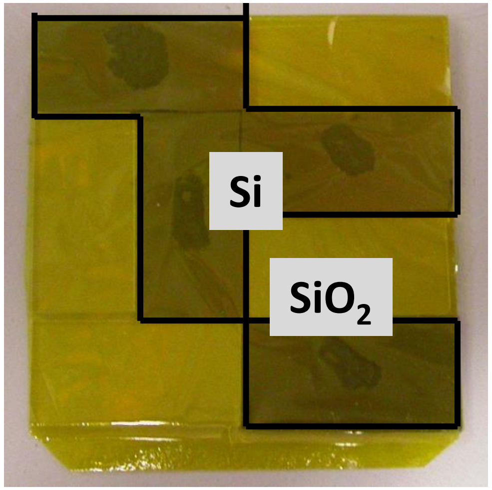
Real world sample (cutout from 94 T magnet coil)
→ What we can learn from our method

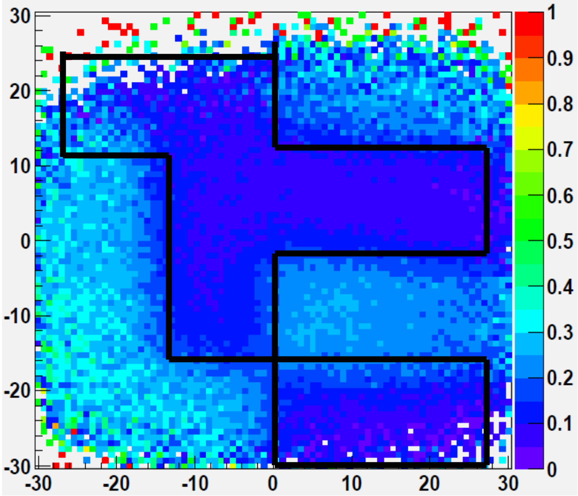
# Setup and results: 2D image reconstruction



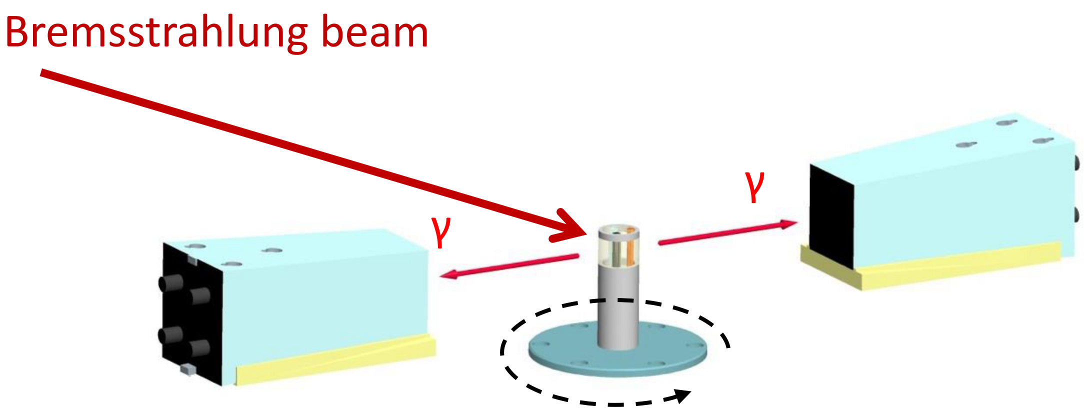



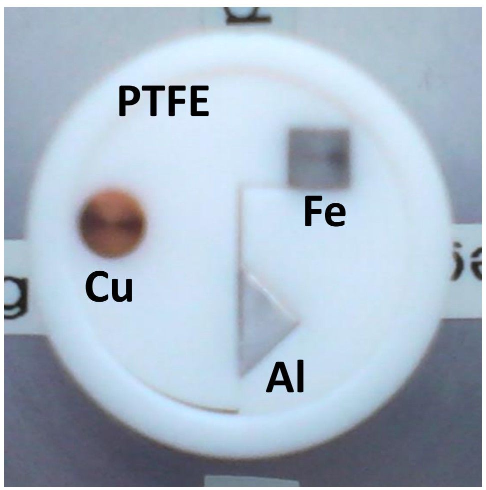
Sample selected to give balanced positron yield.

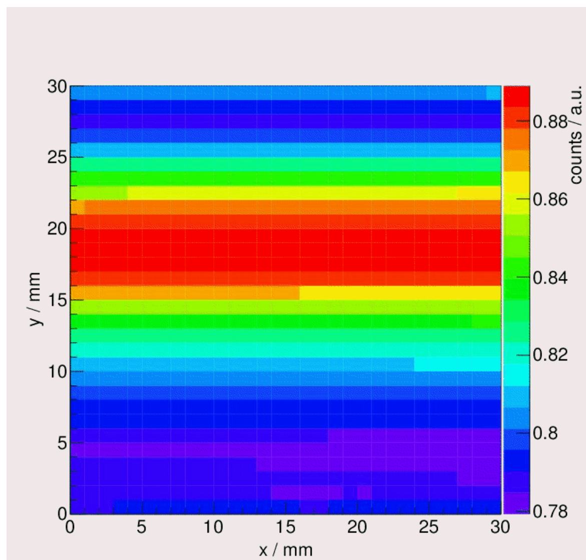
Lifetime-gated 2D reconstructed image by back-projection.

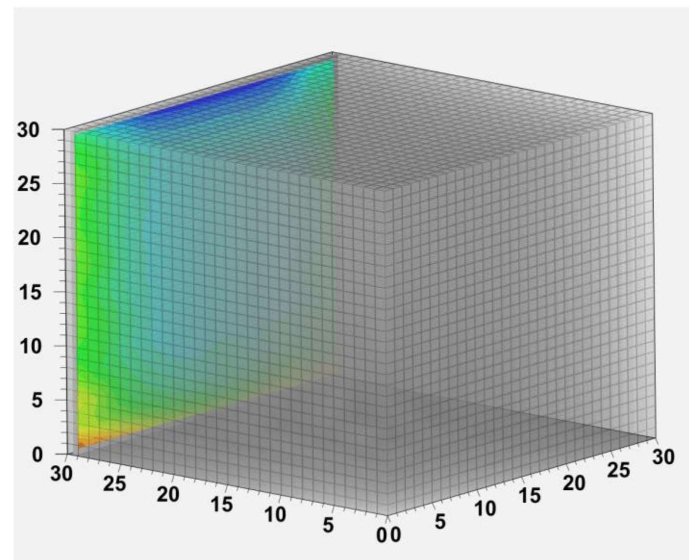
# 3D reconstruction




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)$^3$ voxel space and back-projection algorithm.



# Maximum Likelihood Expectation Maximization


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

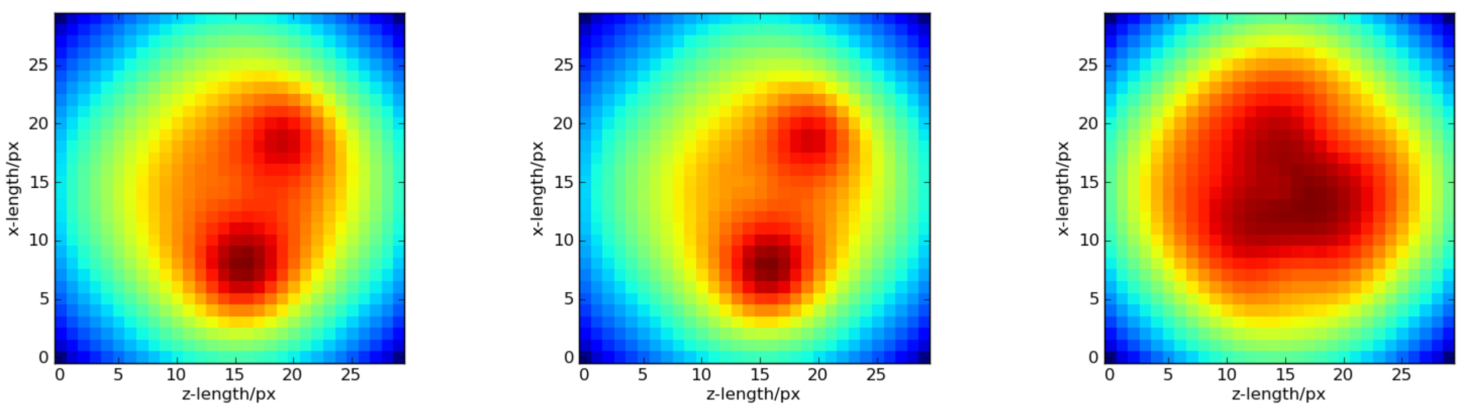
Solves the inversion problem numerically where one has a **system matrix** $M$, an a-priori unknown **source distribution** $s$ and a **measured distribution** $r$.

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

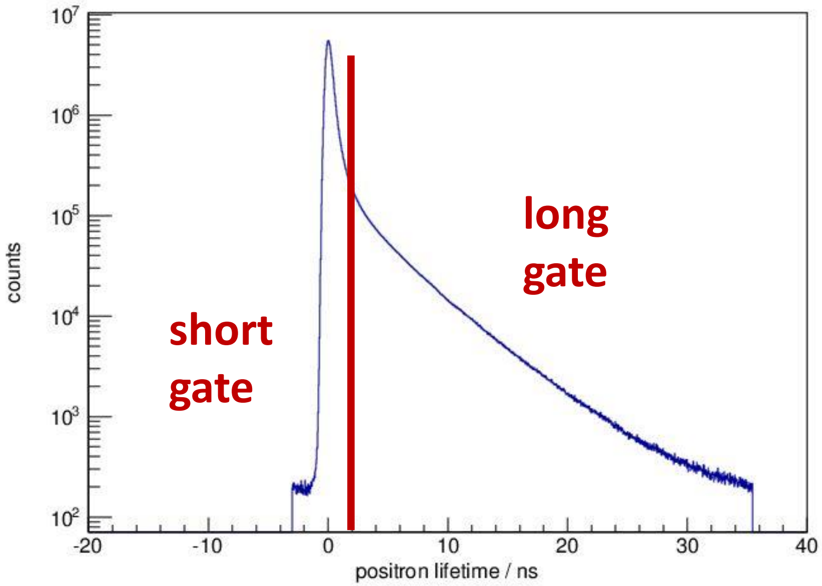
The system matrix has a size of $13^2$ x $13^2$ x 180 x $30^3$ = 138 x $10^9$.

**step 1**

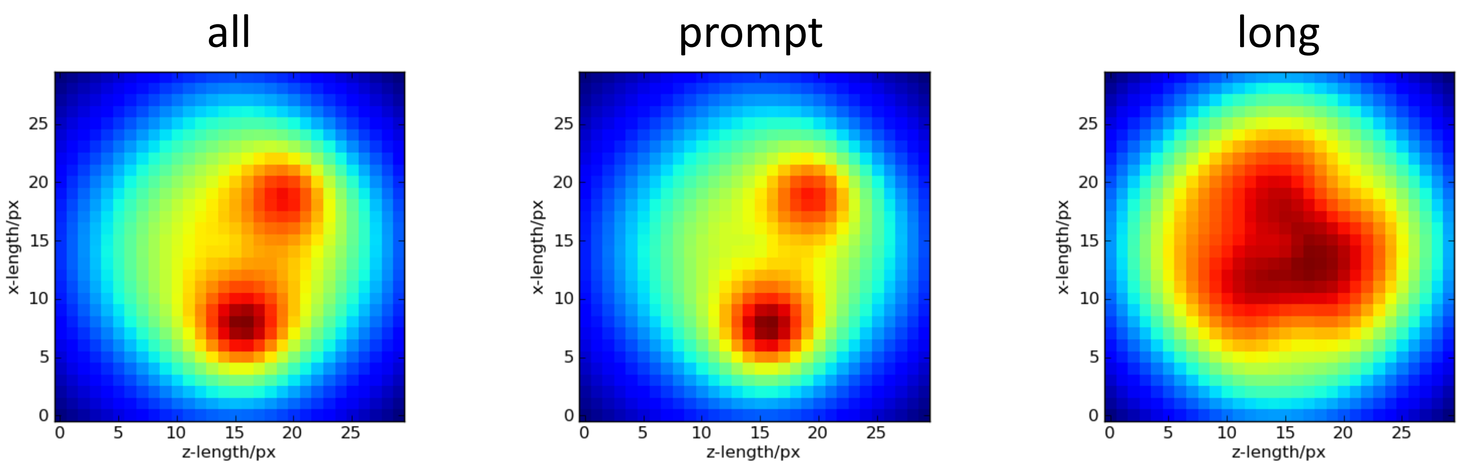
all    prompt    long



# MLEM

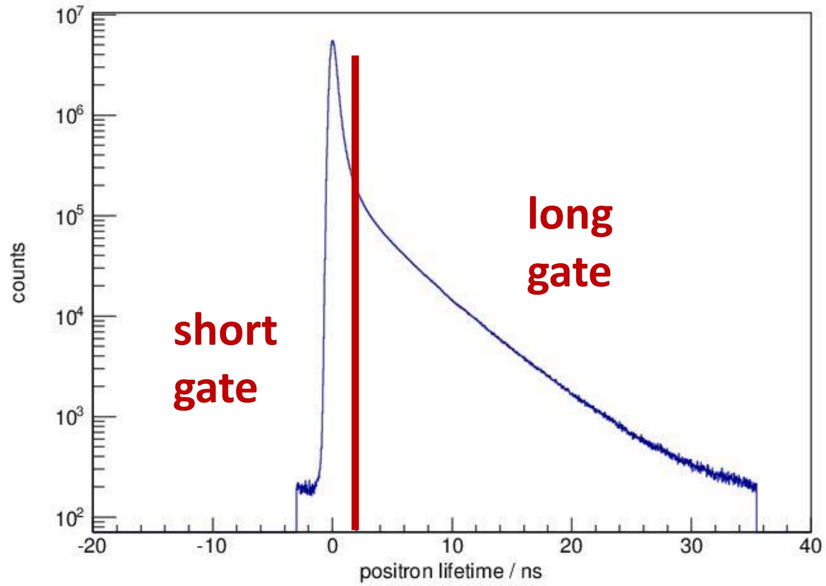




**step 2**

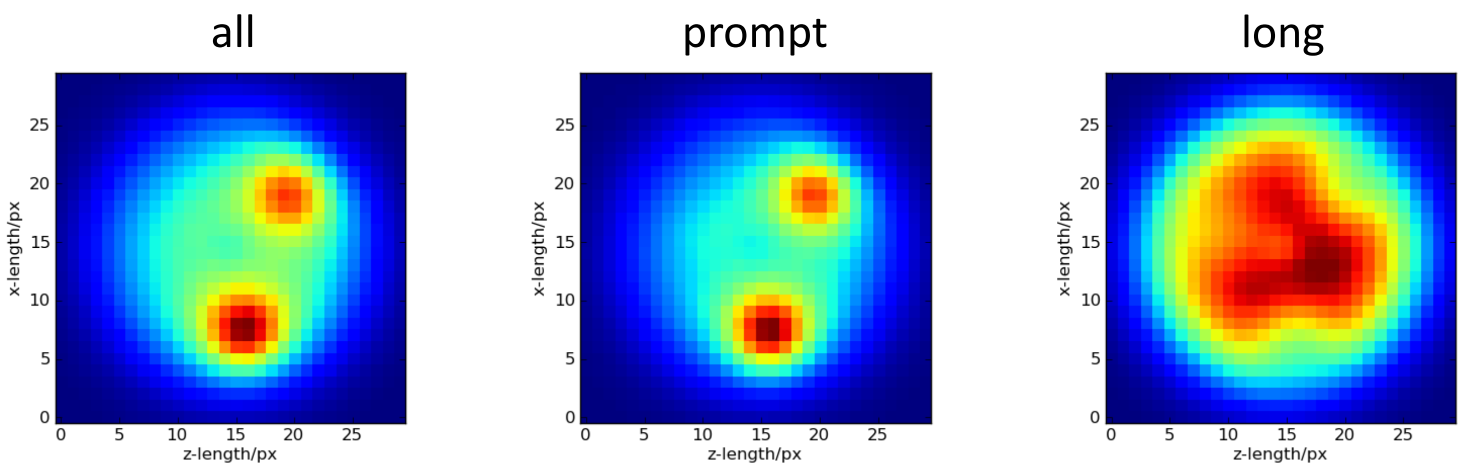



2nd Iteration

# MLEM

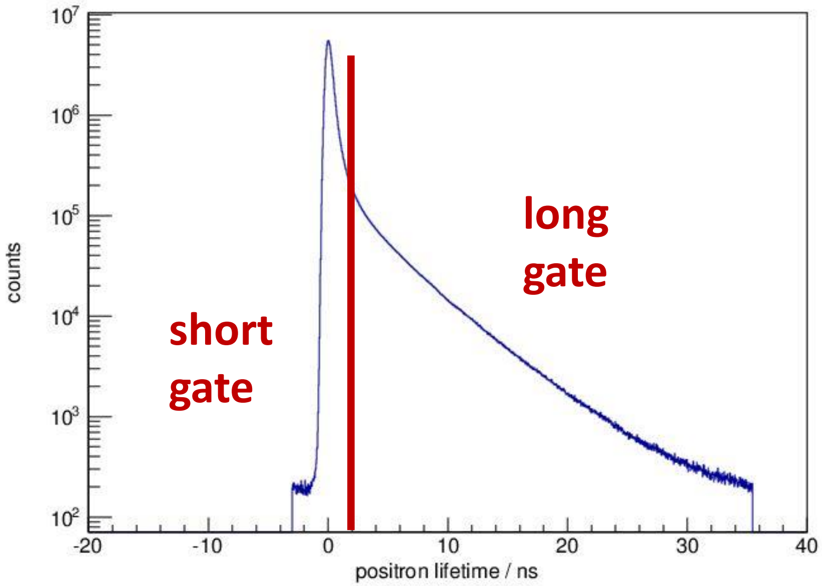




**step 5**

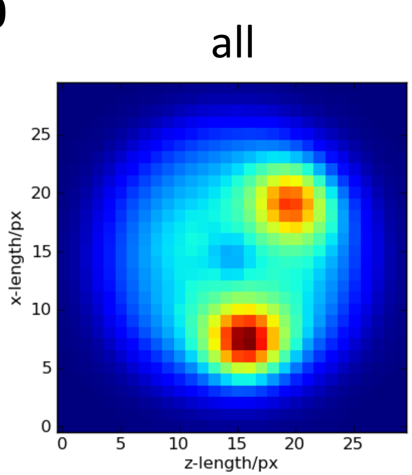


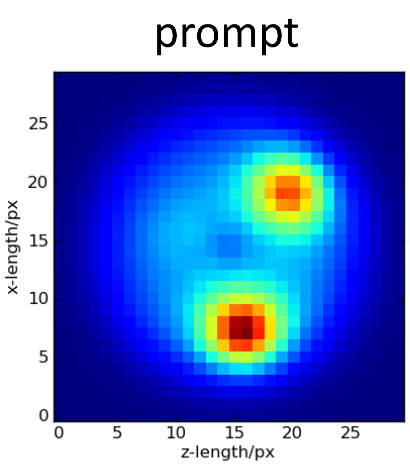

5th Iteration

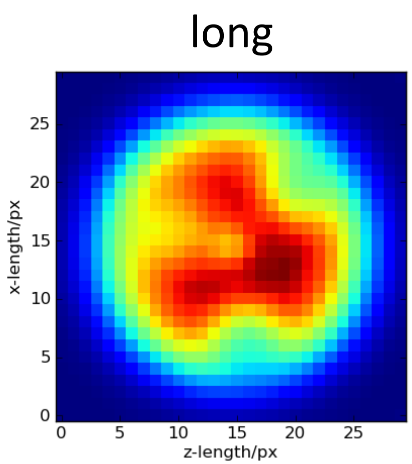
# MLEM


**step 10**

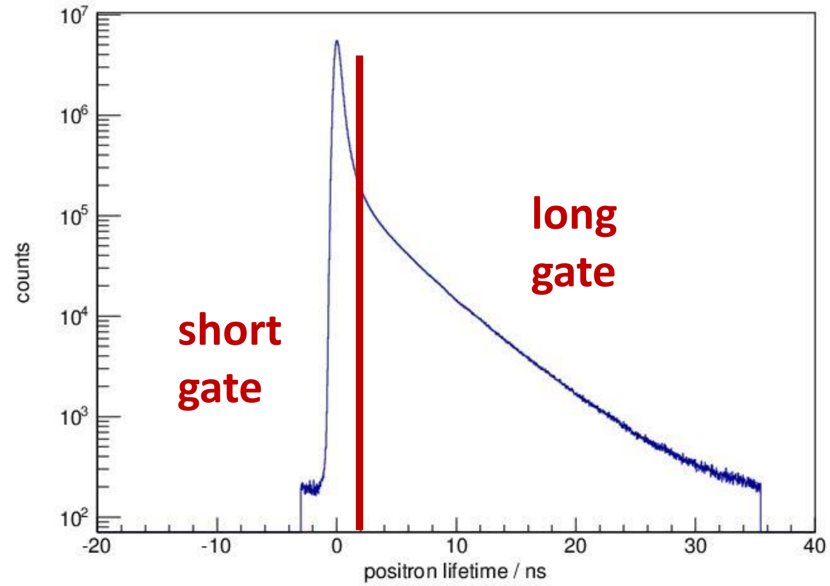


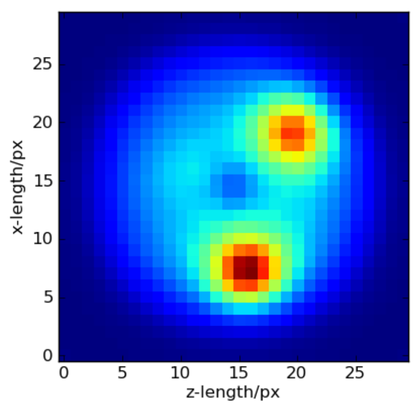

10th Iteration

# MLEM

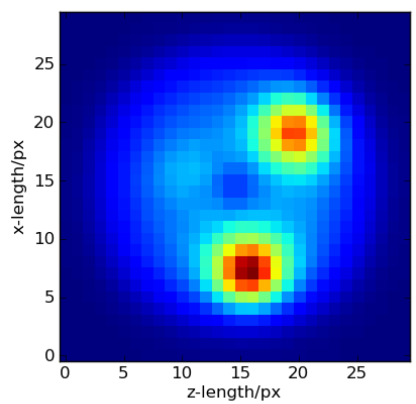




**step 20**

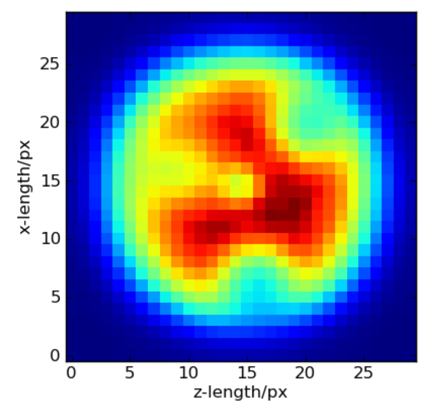
all




prompt



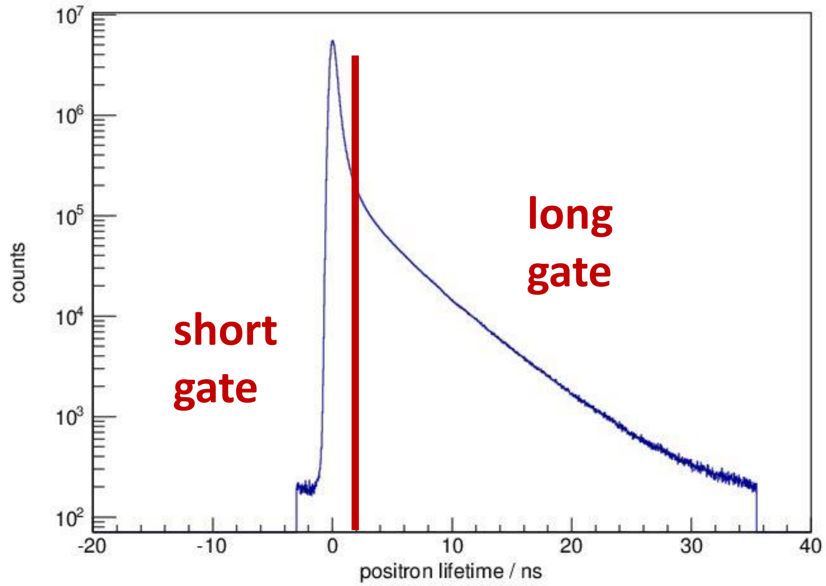

long



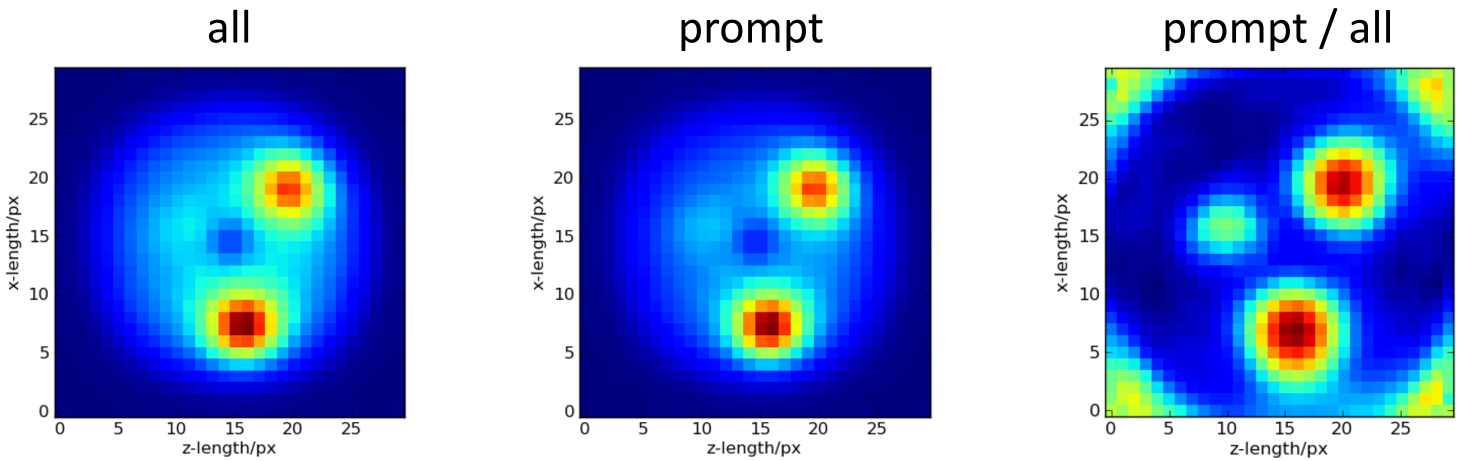

20th Iteration

# MLEM

Aluminium
Steel
Copper

counts
positron lifetime / ns
short gate
long gate

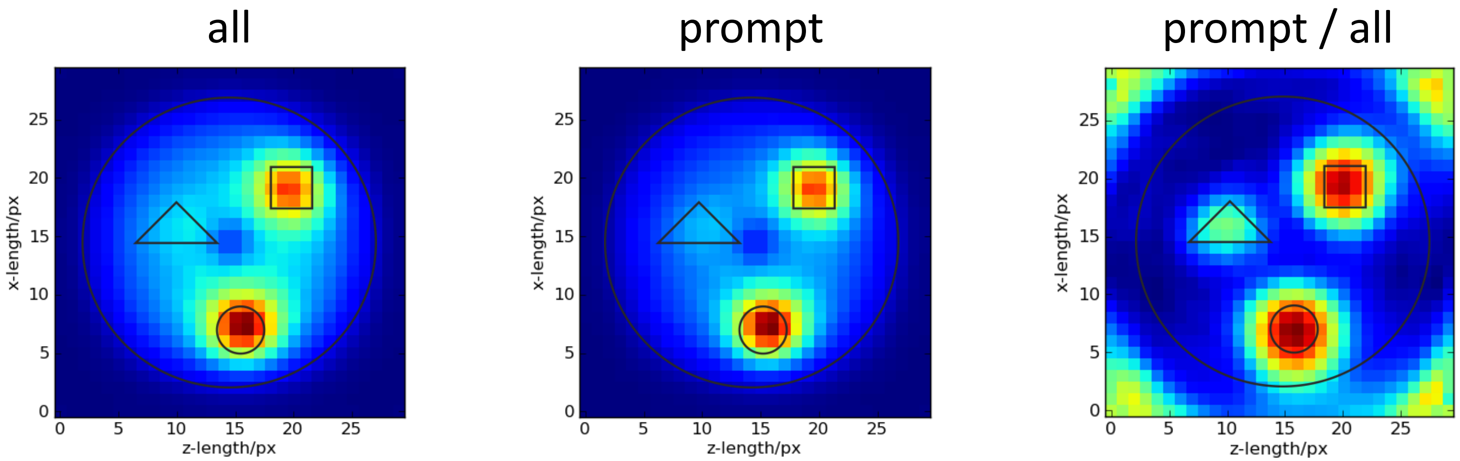



all
prompt
prompt / all
x-length/px
z-length/px

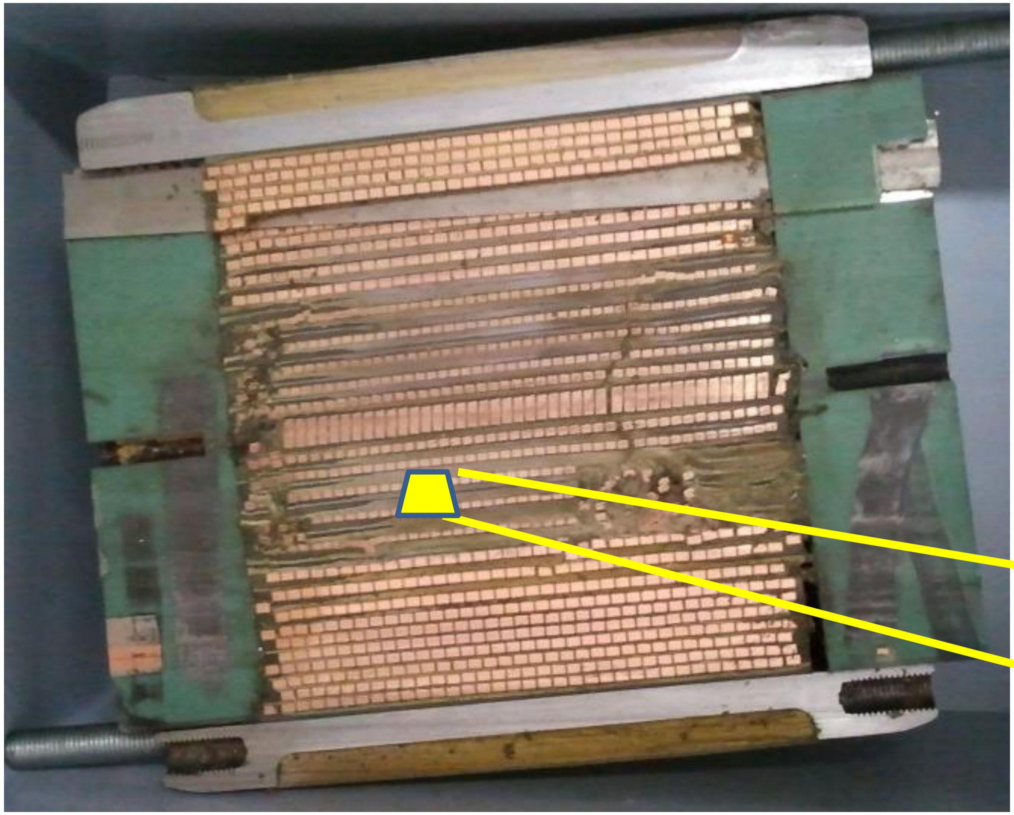
# MLEM

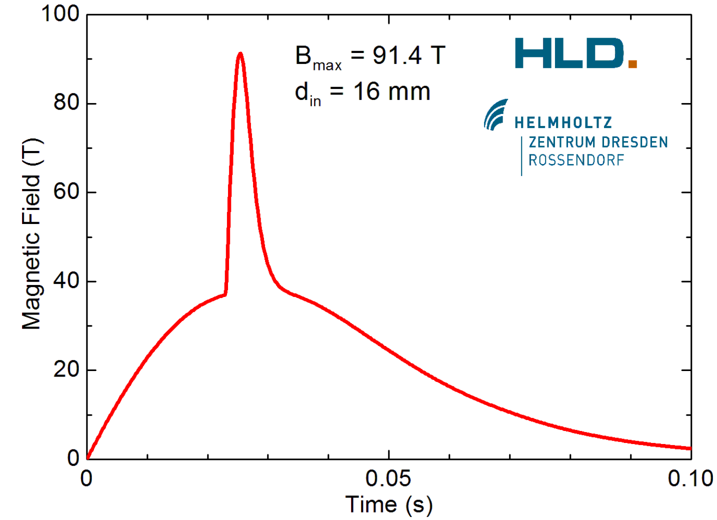

Gating on positron lifetimes with 225 ps timing resolution.

Now the Al is clearly discriminated against the surrounding Teflon.

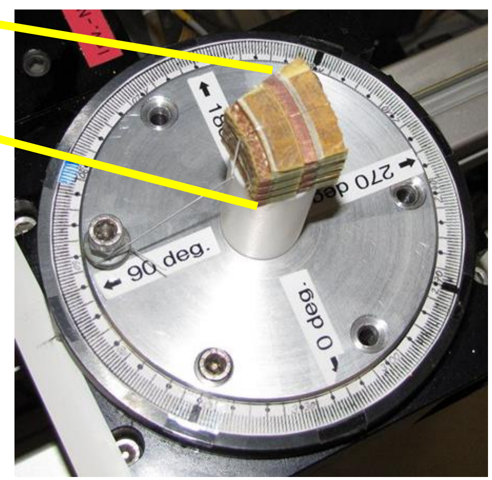


# Lifetime-sensitive analysis
# B-field coil



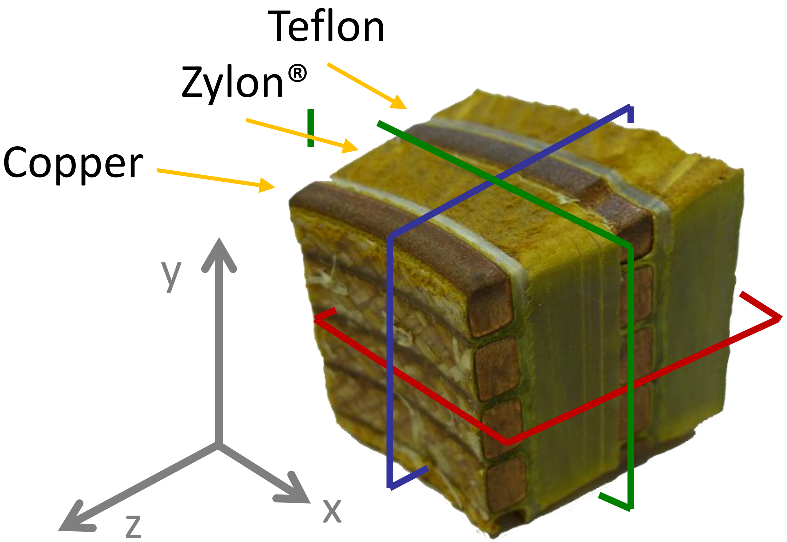


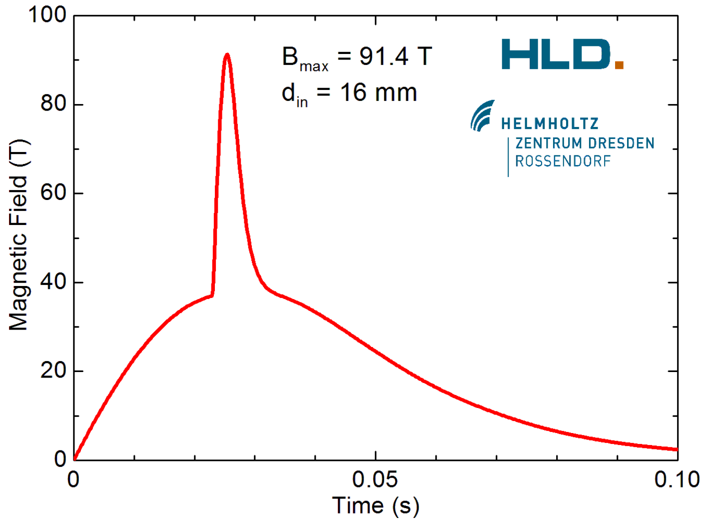

Courtesy: Jochen Wosnitza

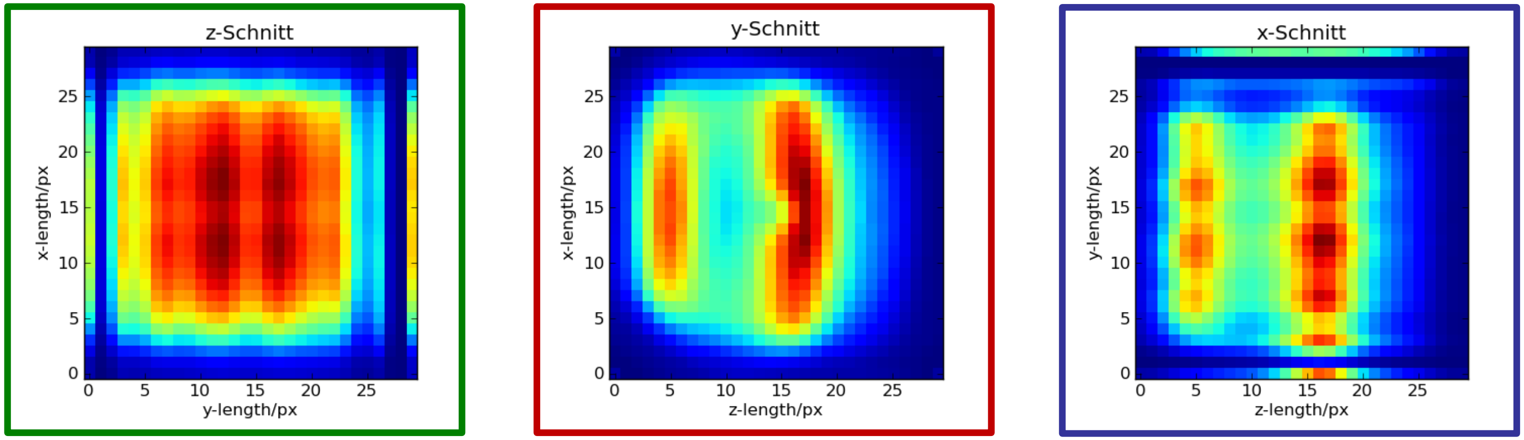



Cut through the record coil which reached 91.4 T peak field. Coil is fed by the world's largest capacitor bank w/ 50 MJ stored energy.

# Tomography: B-field coil



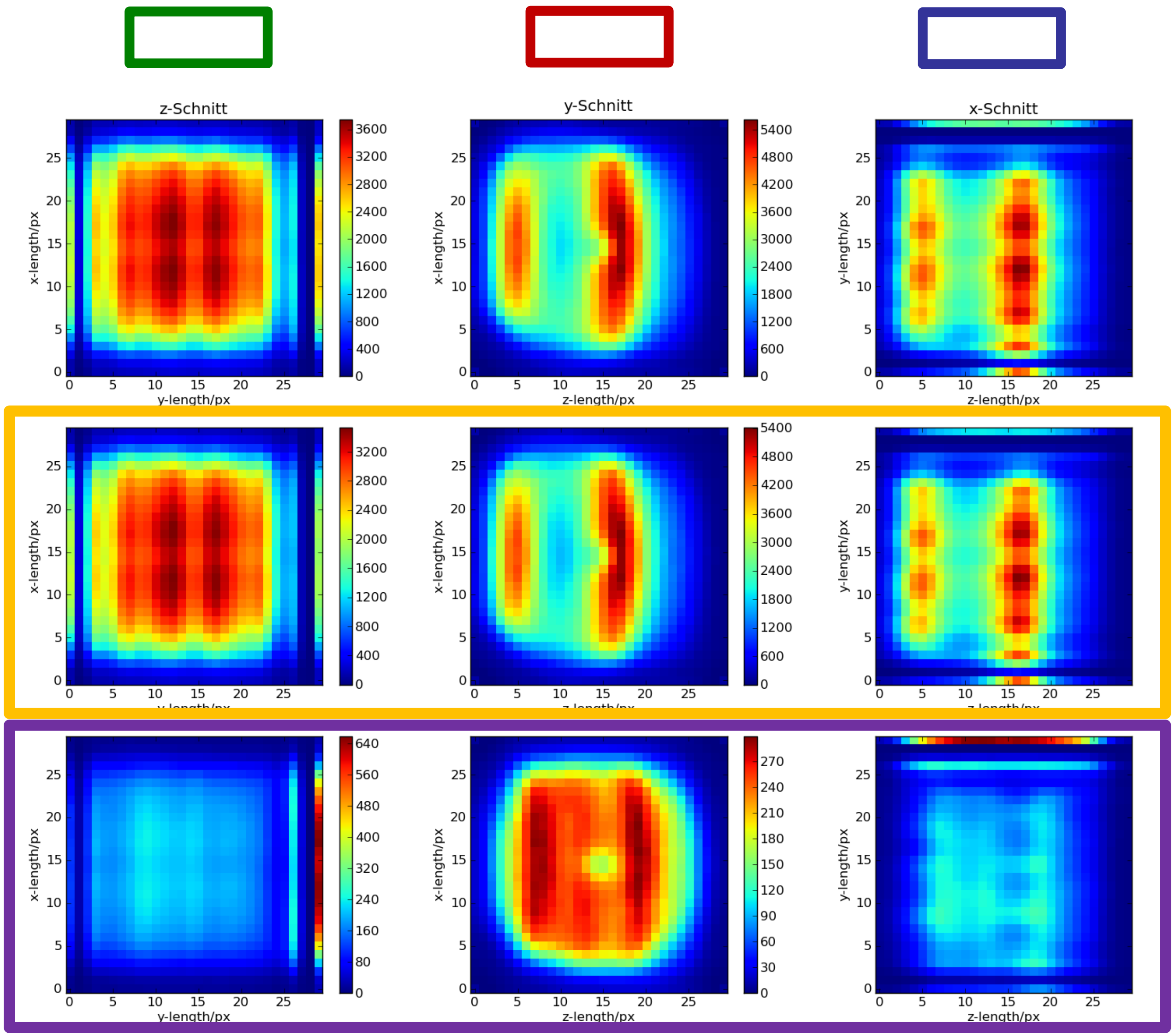



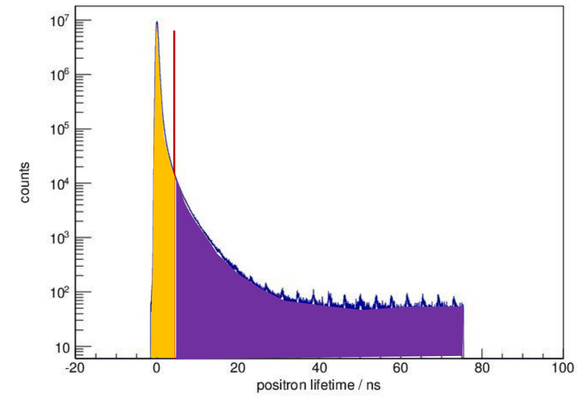



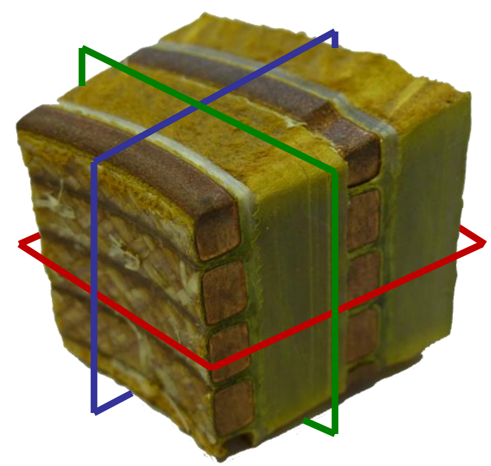

48 h measurement time, 316 GB, 1.6 G events

324 M filtered coincidences

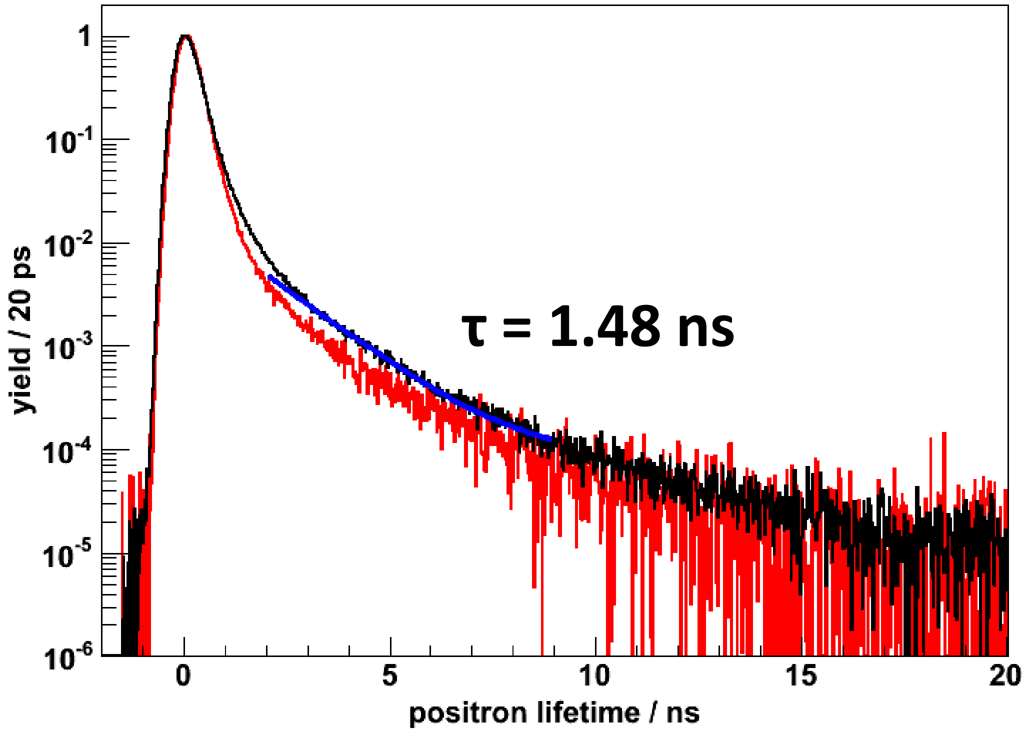
# Lifetime-sensitive analysis: B-field coil

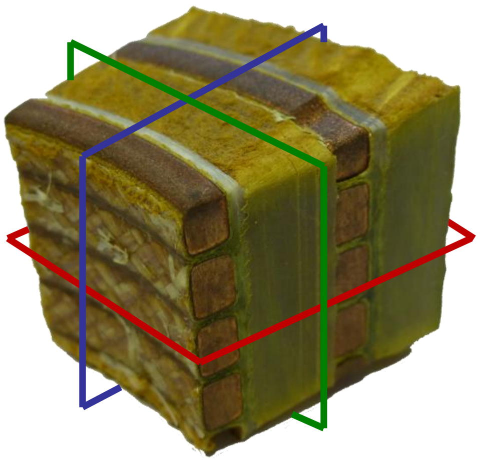


# Lifetime-sensitive analysis: B-field coil



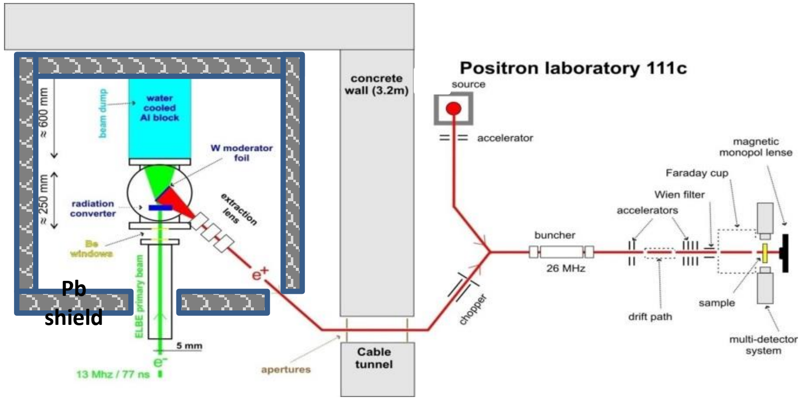



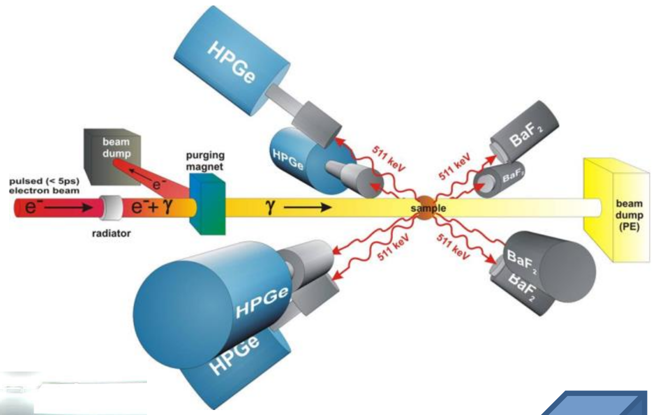
Now, we select specific voxels and determine the annihilation lifetimes for spatially separated regions. Since the voxel is identified as an ensemble over all possible lines-of-response between two detector crystals, the lifetime distribution is a convolution as well. Some real physics questions needed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

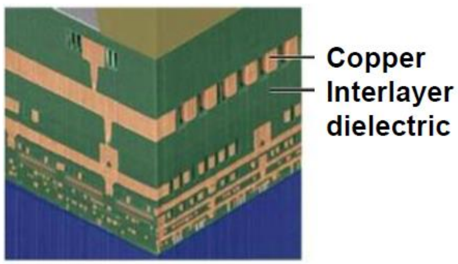
# Summary

Summary:

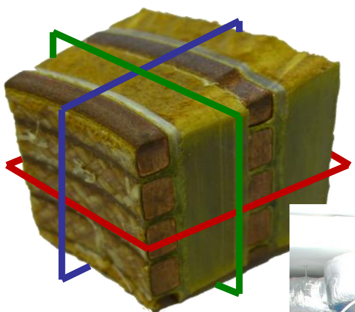
- Accelerator-driven positron production
- Annihilation lifetime spectroscopy for semiconductors, membranes, fluids, reactor materials…
- First results for 3D tomography

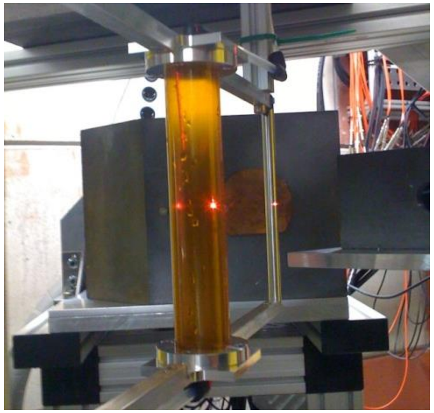


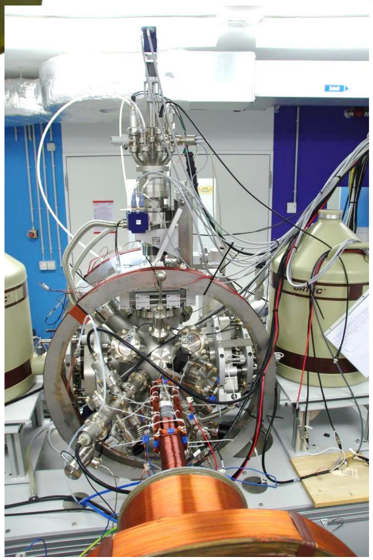



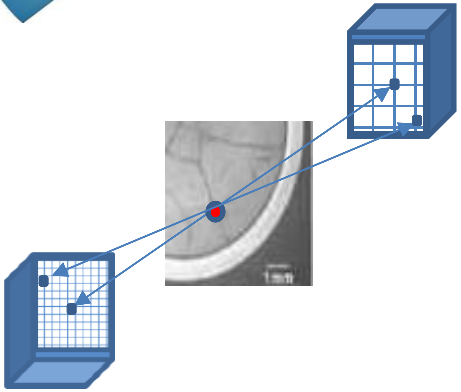



CMOS device









# Collaborations

**Application deadline for 2014 experiments: May 5th**
**http://www.hzdr.de/elbe/**

<u>Helmholtz-Zentrum Dresden-Rossendorf</u>

Thomas E. Cowan*
Wolfgang Anwand
Maik Butterling
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Mathias Kempe *
Maciej Oskar Liedke
Fine Fiedler
Fabian Fritz *
Kay Potzger
Thu Trang Trinh

<u>Martin-Luther-Univ. Halle-Wittenberg</u>

Reinhard Krause-Rehberg
Nicki Hinsche
Marco Jungmann
Arnold Krille
Andreas Müller


Institute of Theoretical and Experimental Physics Moscow
Technische Universität Bergakademie Freiberg
Bhaba Atomic Research Centre Mumbai
Universität Leipzig
Fraunhofer IZFP
Fraunhofer ENAS
אוניברסיטת בן-גוריון בנגב
Ben-Gurion University of the Negev
جامعة بن غوريون في النقب
Lomonosov Moscow State University
TH Wildau
Charles University Prague
MARTIN-LUTHER-UNIVERSITÄT HALLE-WITTENBERG
HELMHOLTZ ASSOCIATION

# More e+ talks and posters

| | Topic | Date |
|---|---|---|
| T. Staab | Positron µ-beam | Monday, 10:00, MER02 |
| U. Ackermann | 4D-AMOC | Monday, 10:45, MER02 |
| M. Dickmann | Scanning positron microscopy | Monday, 11:00, MER02 |
| H. Ceeh | 2D-ACAR w/ Ni | Monday, 11:15, MER02 |
| B. Löwe | CDBS & ACAR | Monday, 11:45, MER02 |
| M.O. Liedke MePS | In-situ defect analysis | Monday, 12:00, MER02 |
| W. Anwand SPONSOR | Ion damage in $ZrO_2$ | Monday, 12:15, MER02 |
| M. Butterling GiPS | RPV steel | Monday, 12:30, MER02 |
| M. Reiner GiPS | Open vol. defects in MnSi | Monday, 12:45, MER02 |
| F. Schumann | Positron-electron pair emission | Monday, 18:15, GER38 |
| T. Gigl | Positron µ-beam | Wednesday, 17:00, P4 |
| J.A. Weber | 2D-ACAR algorithm | Wednesday, 17:00, P4 |
| T. Koschine | Carbon nanotubes | Thursday, 15:00, P2 |
| O. Yildirim SPONSOR | $TiO_2$:Co | Friday, 11:30, POT251 |
| S. Zimnik | Pt & Fe/Pt surfaces | Friday, 12:45, GER38 |