# Materials Research with Positrons - The EPOS Project at FZD

**EPOS-Team & R. Krause-Rehberg**

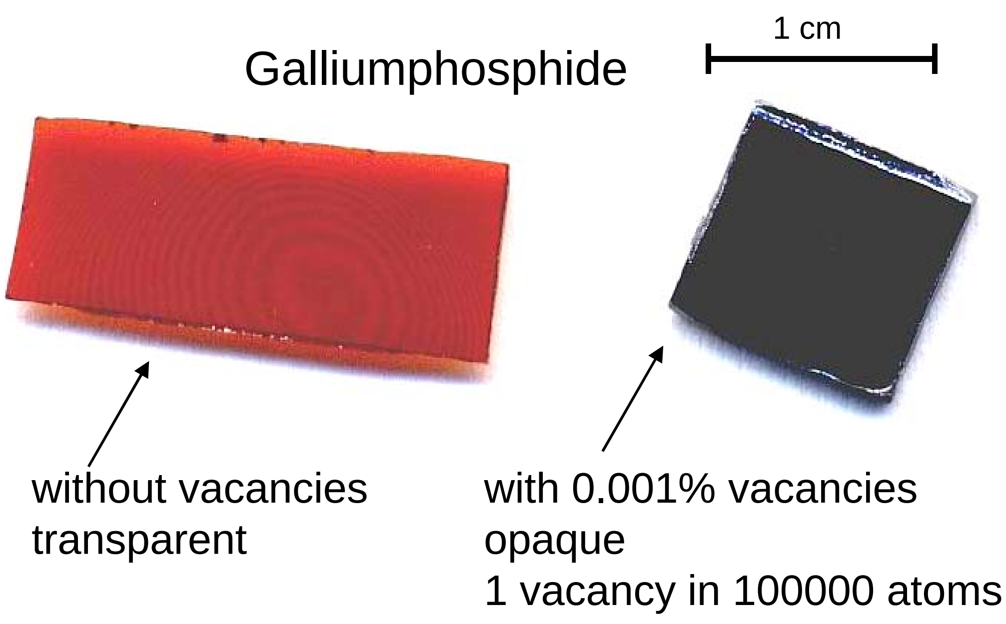
- Positron Annihilation for Materials Sciences
- Extended Concept of EPOS
- Progress of the mono-energetic Positron Beam (MePS)
- Gamma-induced Positron Spectroscopy (GiPS)
- Digital detector system

Martin-Luther-University Halle

Forschungszentrum Dresden-Rossendorf

# Point defects determine properties of materials

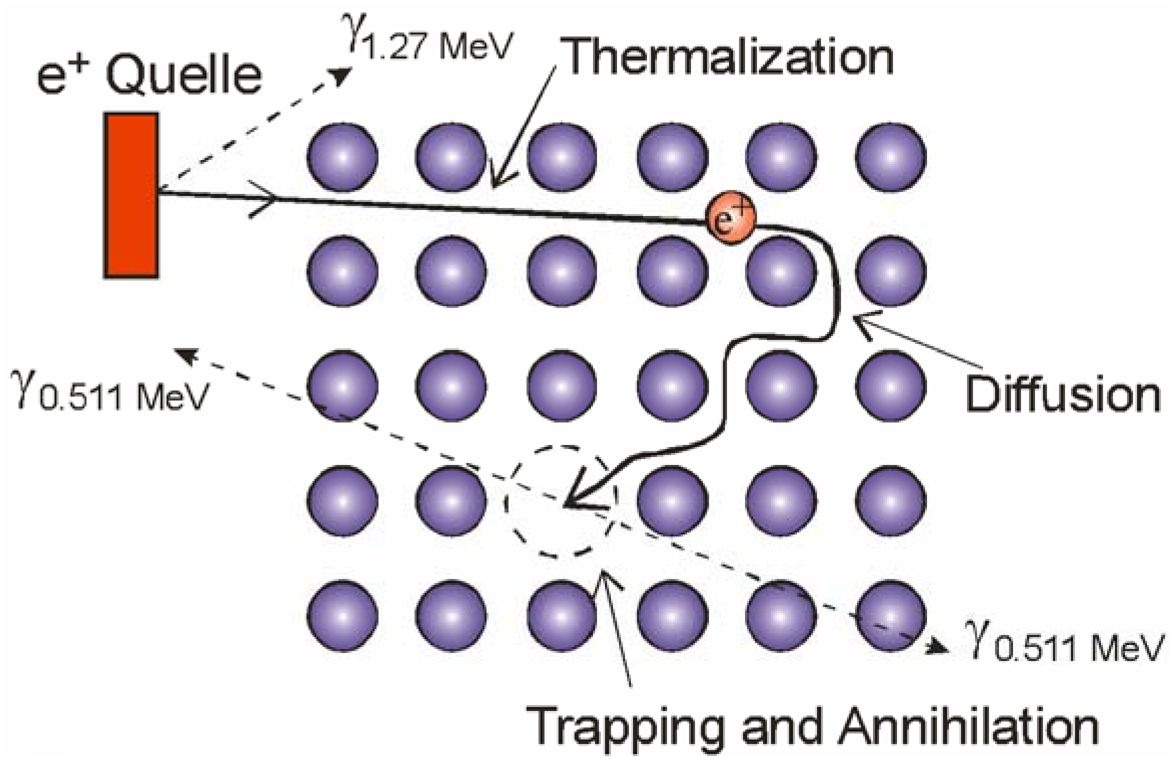
- Point defects determine electronic and optical properties
- electric conductivity strongly influenced
- Doping of semiconductors (n-, p-Si)

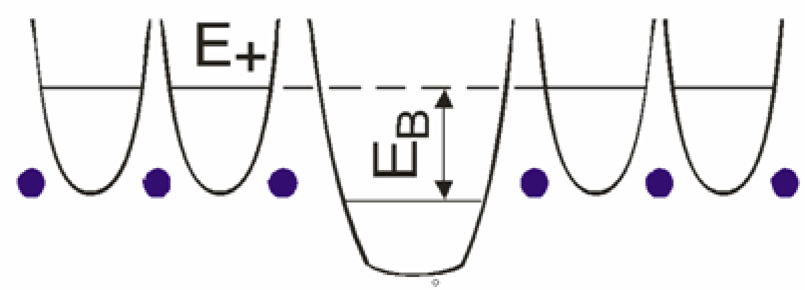



- Point defects are generated by irradiation (e.g. cosmic rays), by plastic deformation or by diffusion, …
- Metals in high radiation environment -> formation of voids -> embrittlement
- -> Properties of vacancies and other point defects must be known
- Analytical tools are needed to characterize point defects

# The positron lifetime spectroscopy

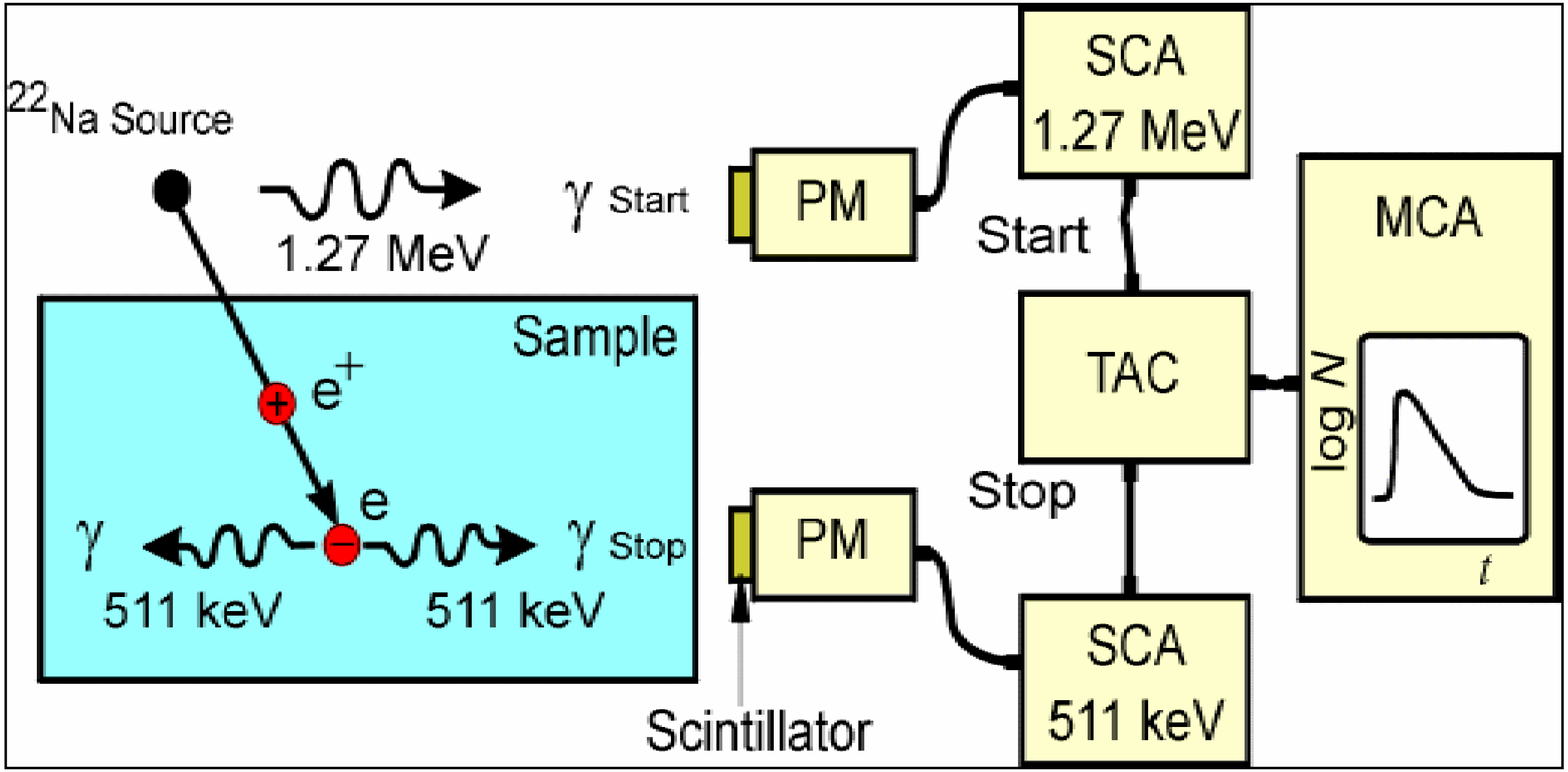
$^{22}Na$



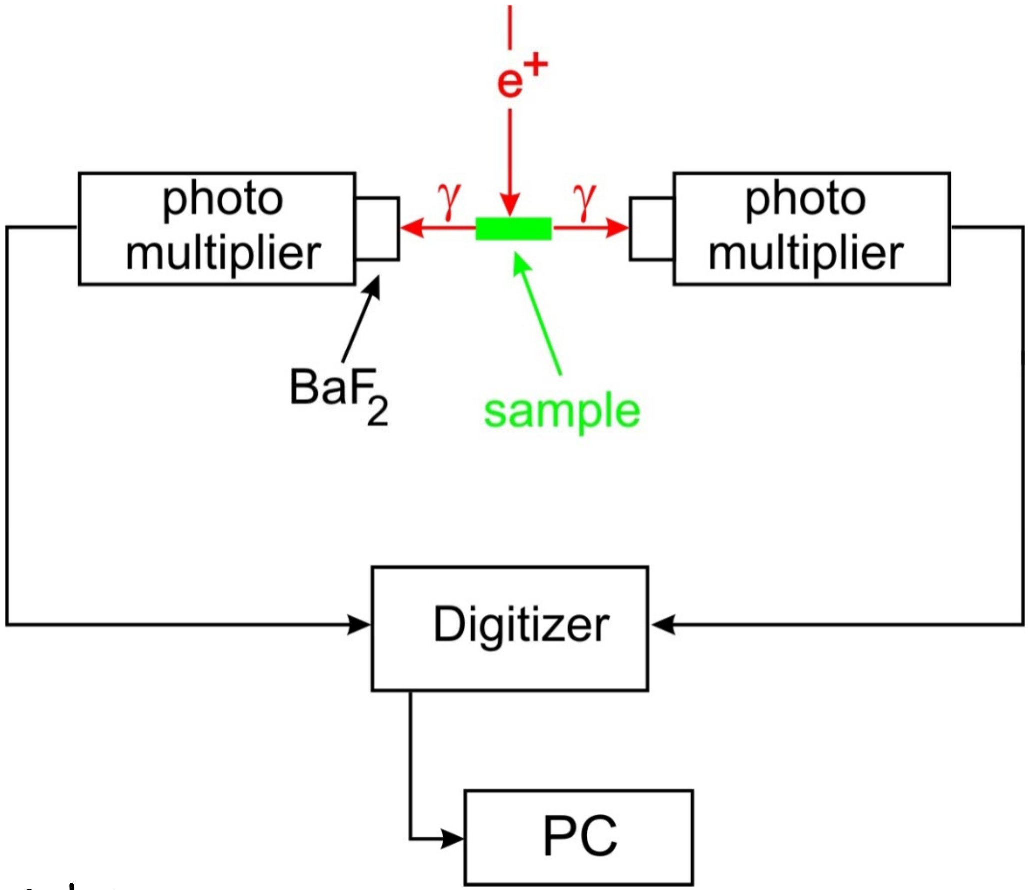

- positron wave-function can be localized in the attractive potential of a defect
- annihilation parameters change in the localized state
- e.g. positron lifetime increases in a vacancy
- lifetime is measured as time difference between appearance of 1.27 (start) and 0.51 MeV (stop) quanta
- defect identification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

# The positron lifetime spectroscopy



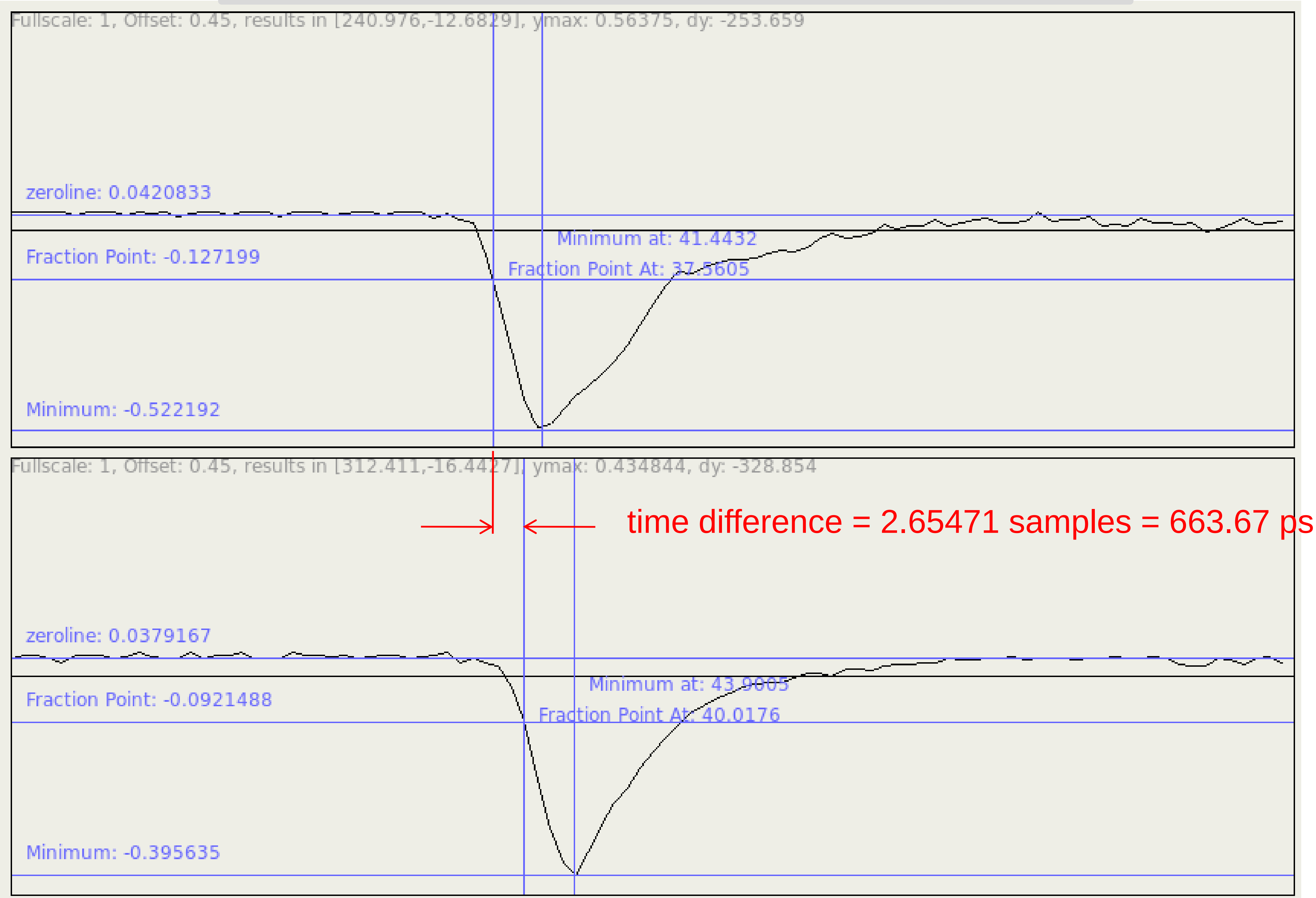

Positron lifetime: time between 1.27 MeV and 0.511 MeV quanta

# Digital lifetime measurement

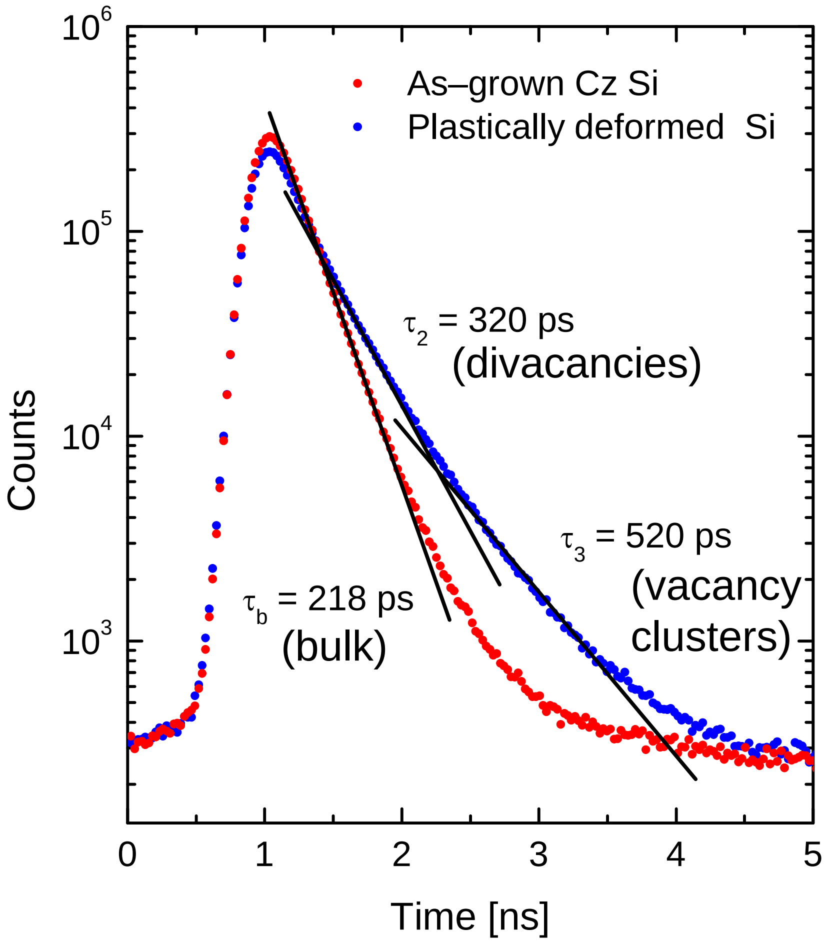



- much simpler setup
- timing very accurate
- pulse-shape discrimination (suppress "bad pulses")
- each detector for start & stop (double statistics)

# screenshot of two digitized anode pulses



# Positron lifetime spectroscopy




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

positron lifetime spectrum:

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

trapping coefficient

$$\kappa_d = \mu C_d = \frac{I_2}{I_1}\left(\frac{1}{\tau_b} - \frac{1}{\tau_d}\right)$$

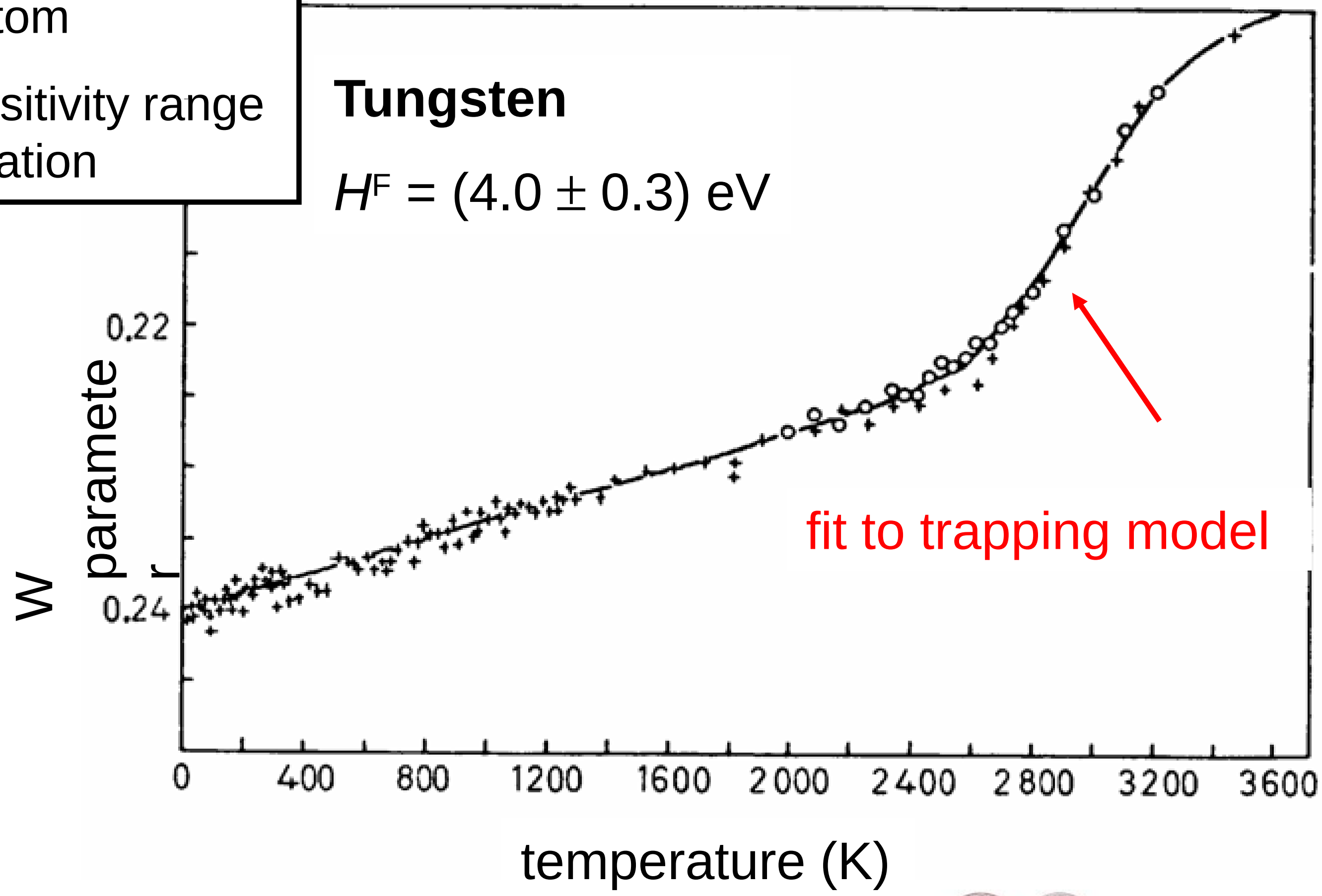
trapping rate

defect concentration

# Vacancies in thermal Equilibrium

- Vacancy concentration in thermal equilibrium:
- in metals $H^F \approx 1 \ldots 4$ eV $\Rightarrow$ at $T_m$ [1v] $\approx 10^{-4 \ldots -3}$ /atom
- fits well to the sensitivity range of positron annihilation

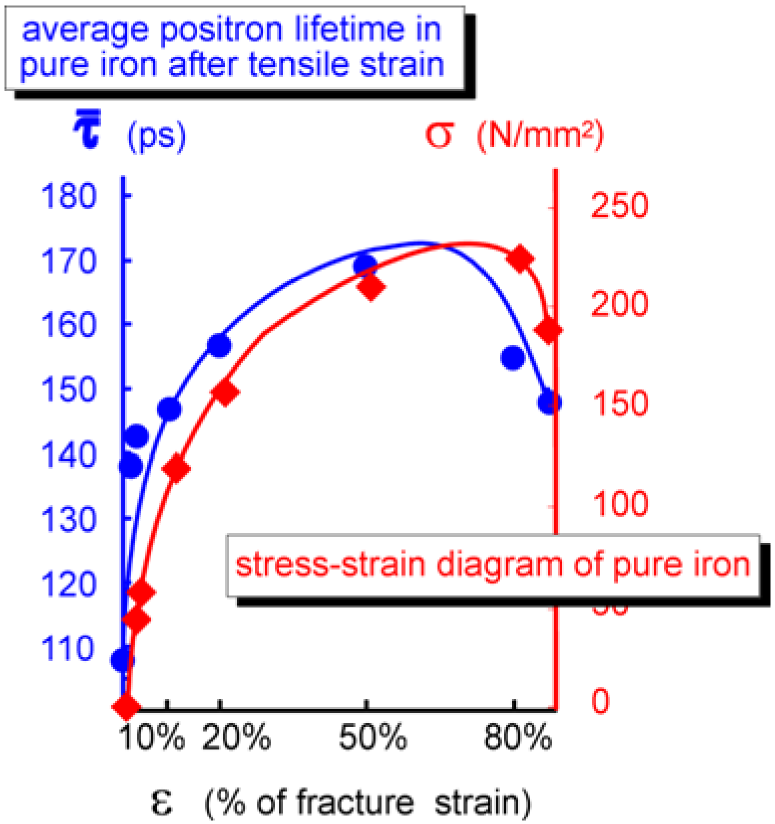
$$C_{1v}(T) = \exp\left(\frac{S_{1v}^{F}}{k}\right) \exp\left(\frac{H_{1v}^{F}}{kT}\right)$$

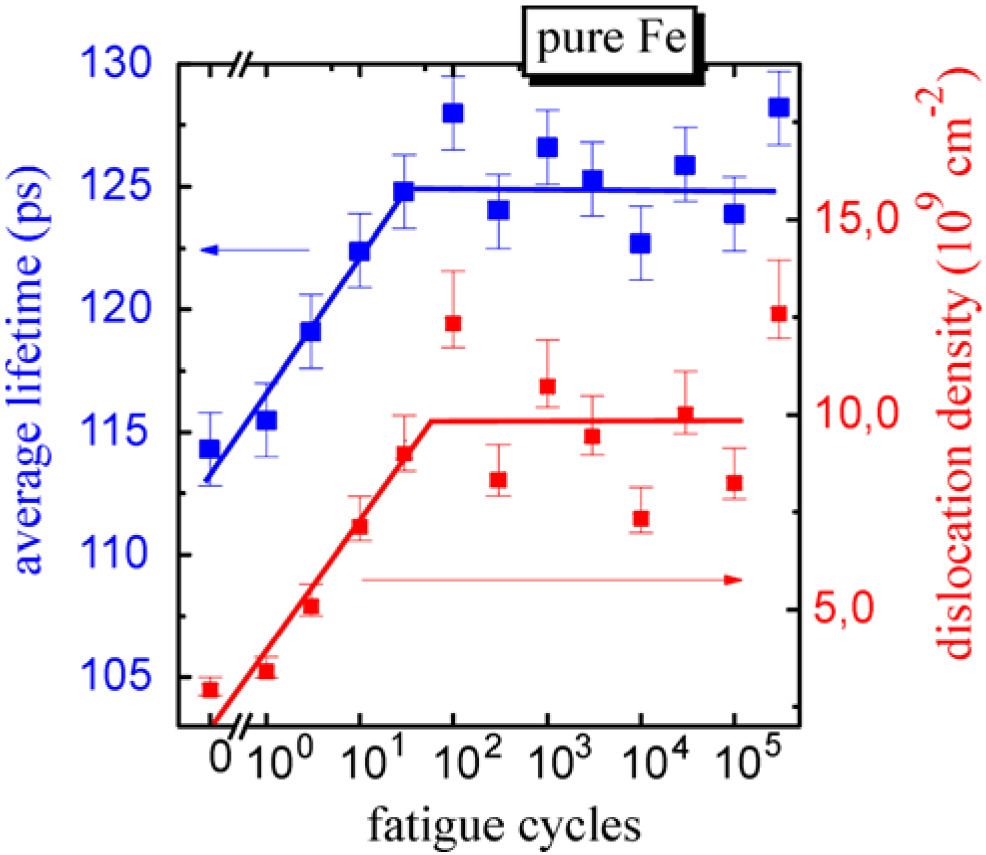



(Ziegler, 1979)

# Defects in Iron after tensile strength and fatigue treatment

- We performed an extensive study of defects in mechanically damaged iron and steel
- Positrons are very sensitive: detection of defects already in the Hooks range of the stress-strain experiment
- Vacancy cluster and dislocations are detectable in both cases

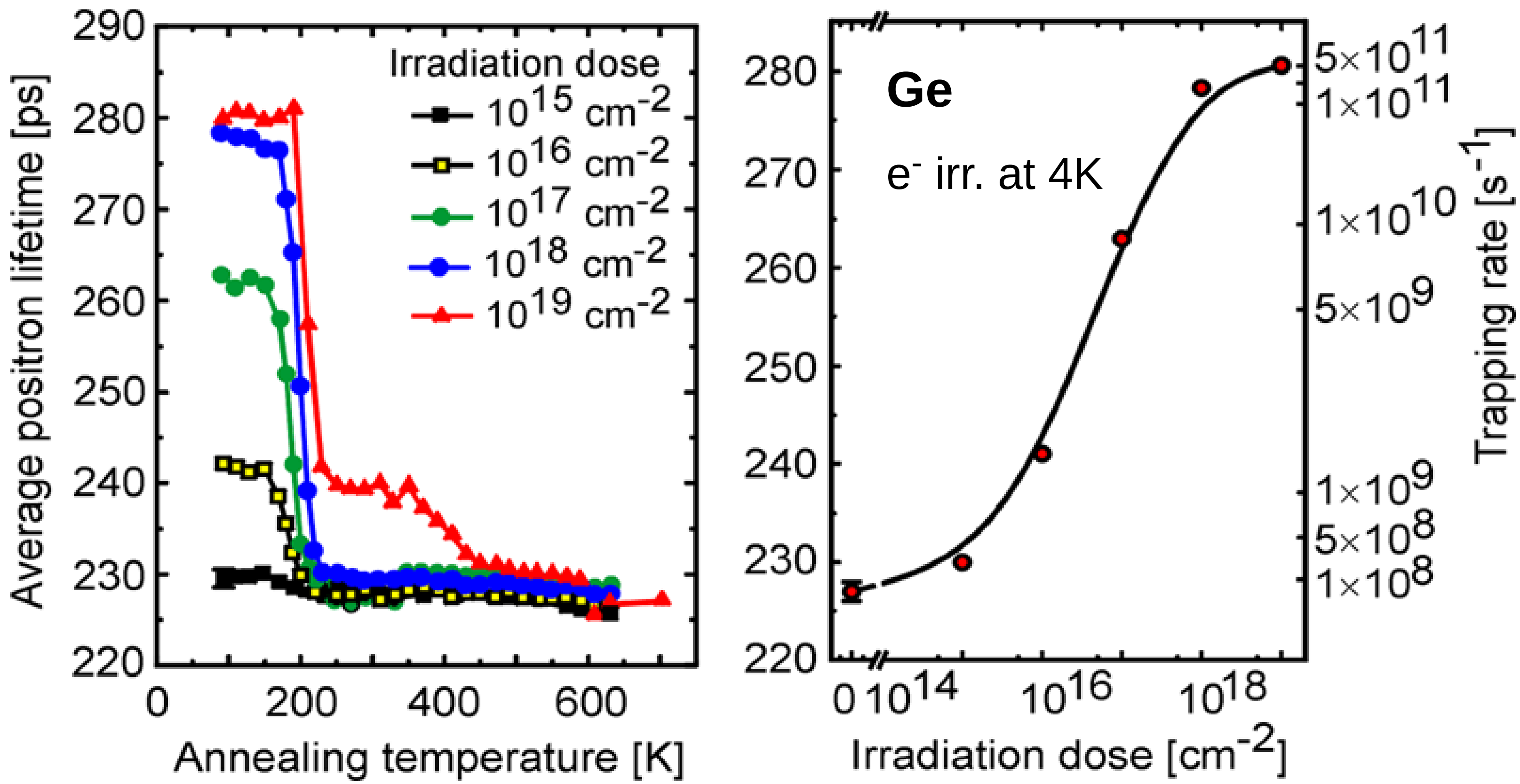



Somieski et al., J. Physique IV 5, C1/127-134, (1995)

# Defects in electron-irradiated Ge

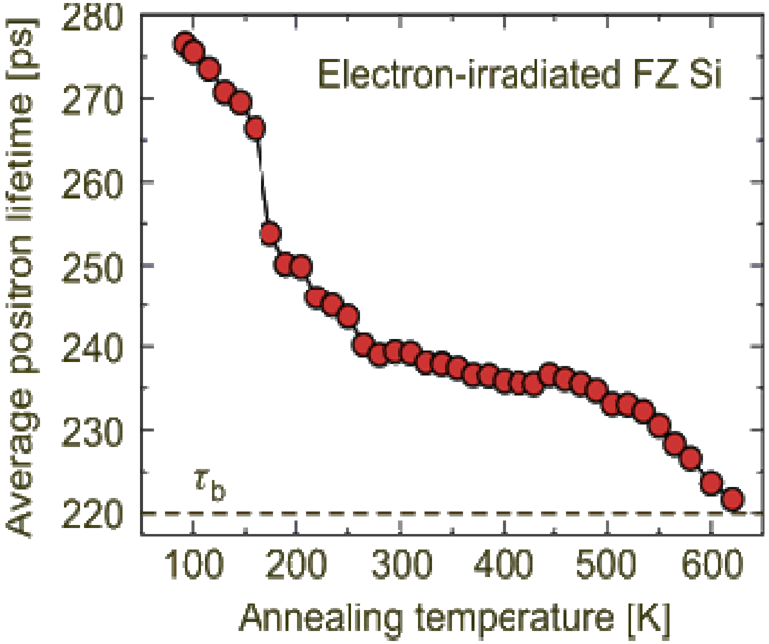
- Electron irradiation (2 MeV @ 4K) induces Frenkel pairs (vacancy - interstitial pairs)
- steep annealing stage at 200 K
- at high irradiation dose: divacancies are formed (thermally more stable)

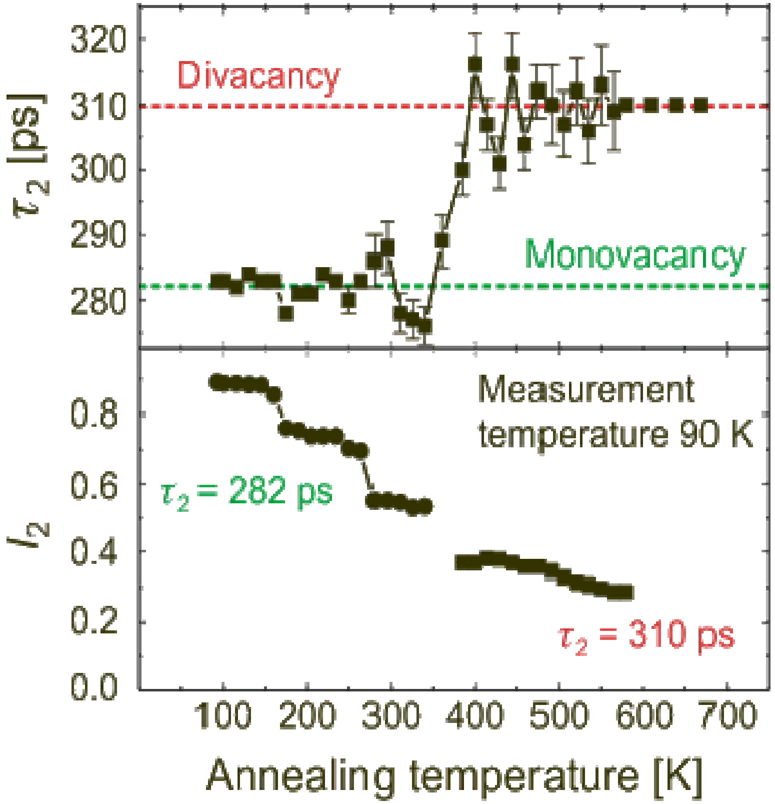



(Polity et al., 1997)

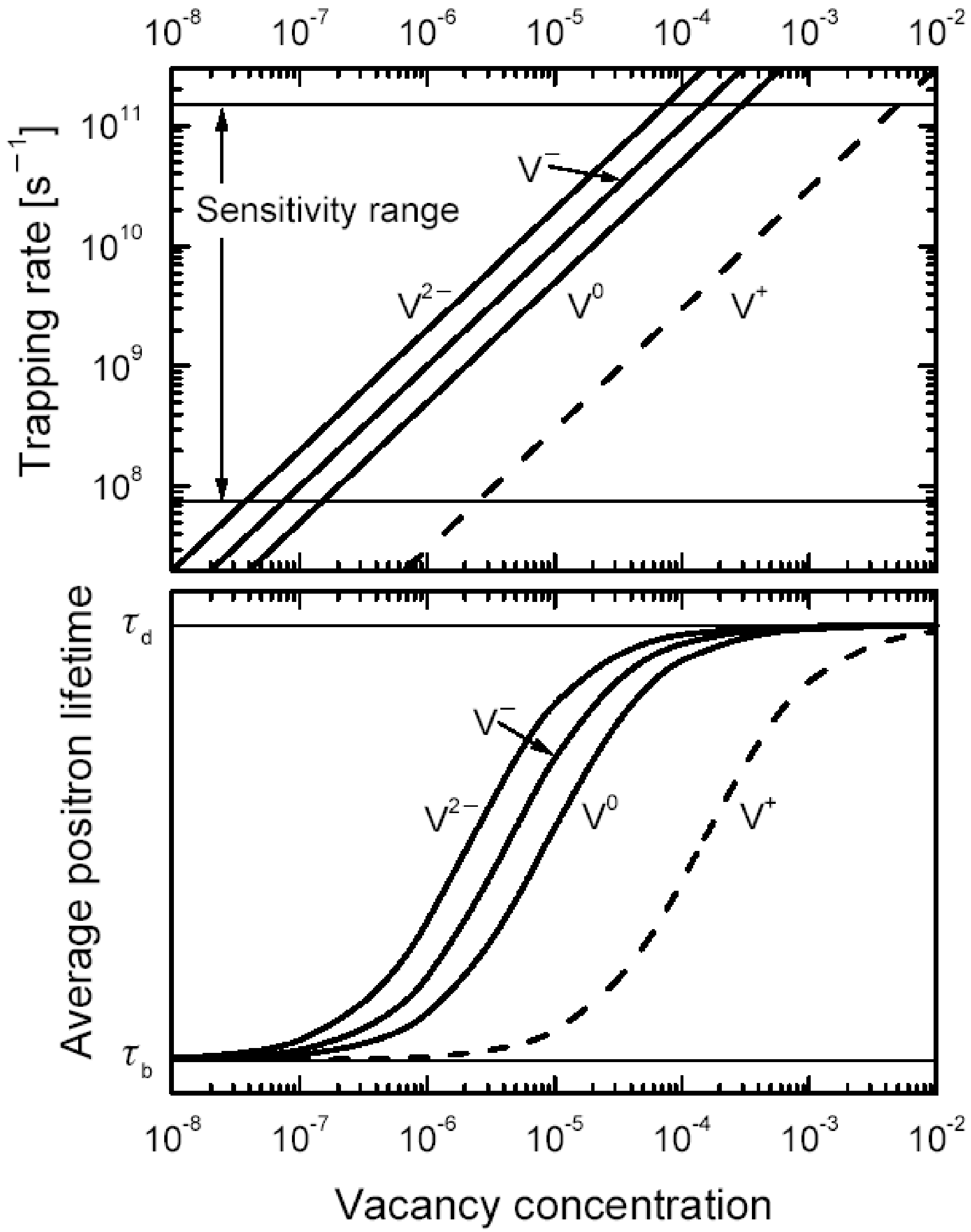
# Electron irradiation of Si

- low-temperature electron irradiation was performed at 4K ($E_{e-}$=2 MeV)
- annealing stage of monovacancies at about 170 K
- moving $V_{Si}$ partly form divacancies
- divacancies anneal at about 550…650 K




Polity et al., Phys. Rev. B **58** (1998) 10363

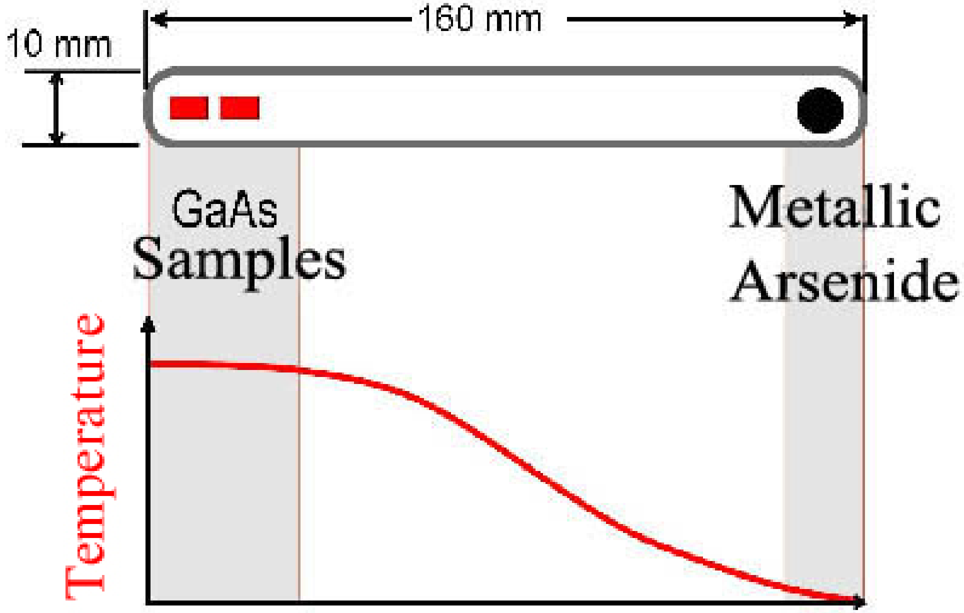



## Sensitivity limits of PAS for vacancy detection

- **lower sensitivity limit** e.g. for negatively charged divacancies in Si starts below $10^{15}$ $cm^{-3}$
- **upper limit**: saturated positron trapping
- defect identification still possible
- Then: only lower limit for defect density can be given
- With positron diffusion measurements using monoenergetic positron beams no upper sensitivity limit

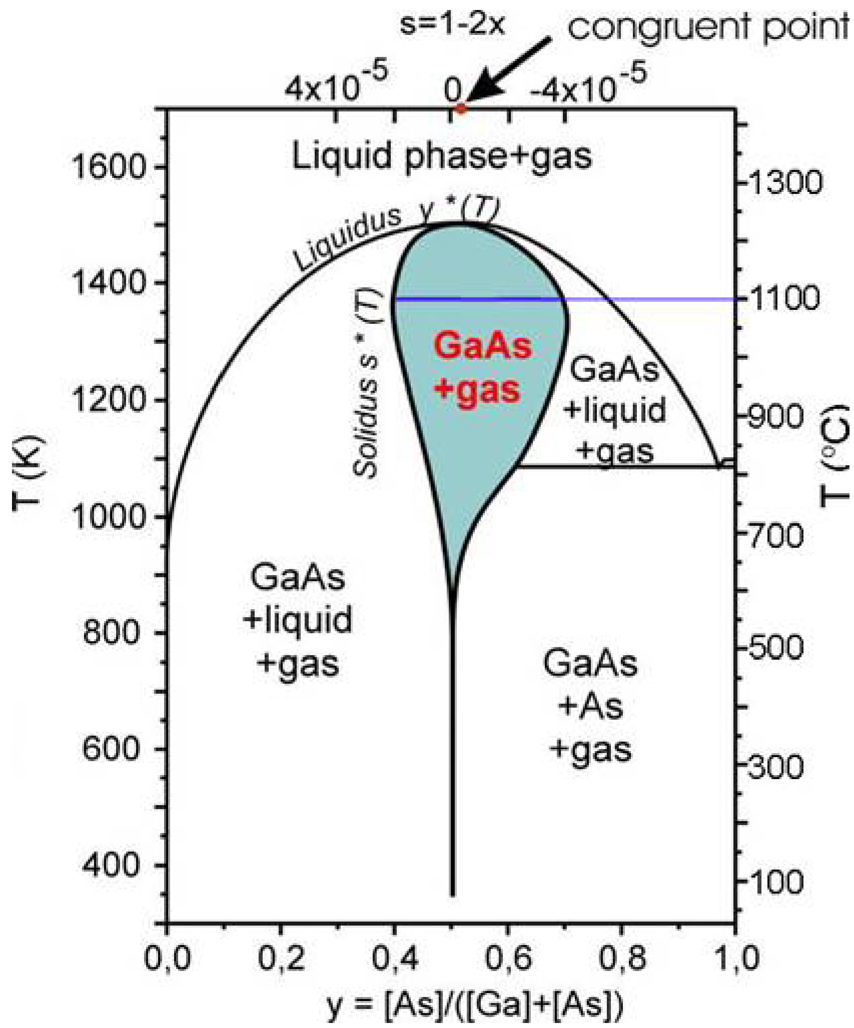
# GaAs: annealing under defined As-partial pressure

- two-zone-furnace: Control of sample temperature **and** As partial pressure allows to navigate freely in phase diagram (existence area of compound)




$T_{sample}$: 1100° C

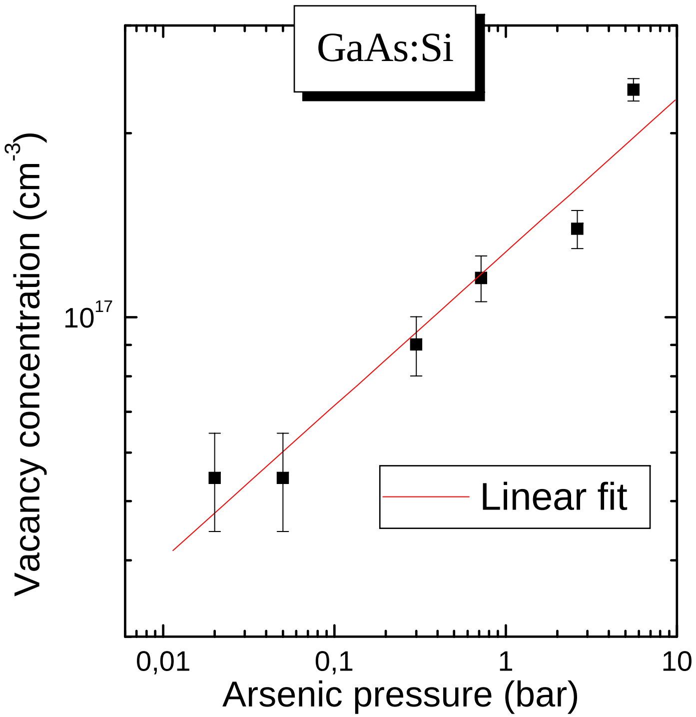
$T_{As}$: determines As-partial pressure



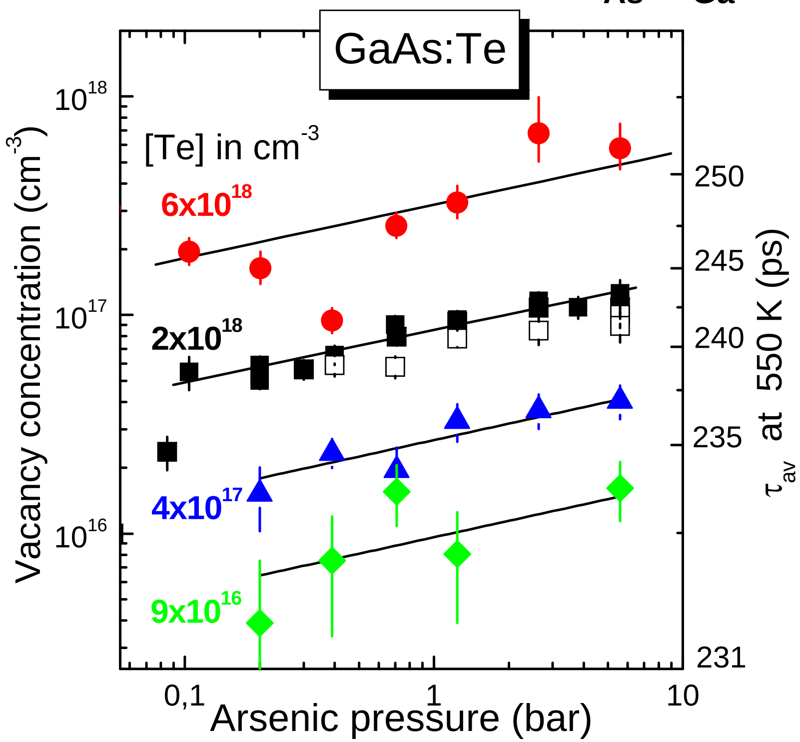

H. Wenzl et al., J. Cryst. Growth **109**, 191 (1991).

# GaAs: Annealing under defined As pressure

**$Si_{Ga}$-$V_{Ga}$**




**$Te_{As}$-$V_{Ga}$**




*J. Gebauer et al., Physica B 273-274, 705 (1999)*

**Thermodynamic reaction:**

$$1/4\ As_4^{gas} \leftrightarrow As_{As} + V_{Ga}$$

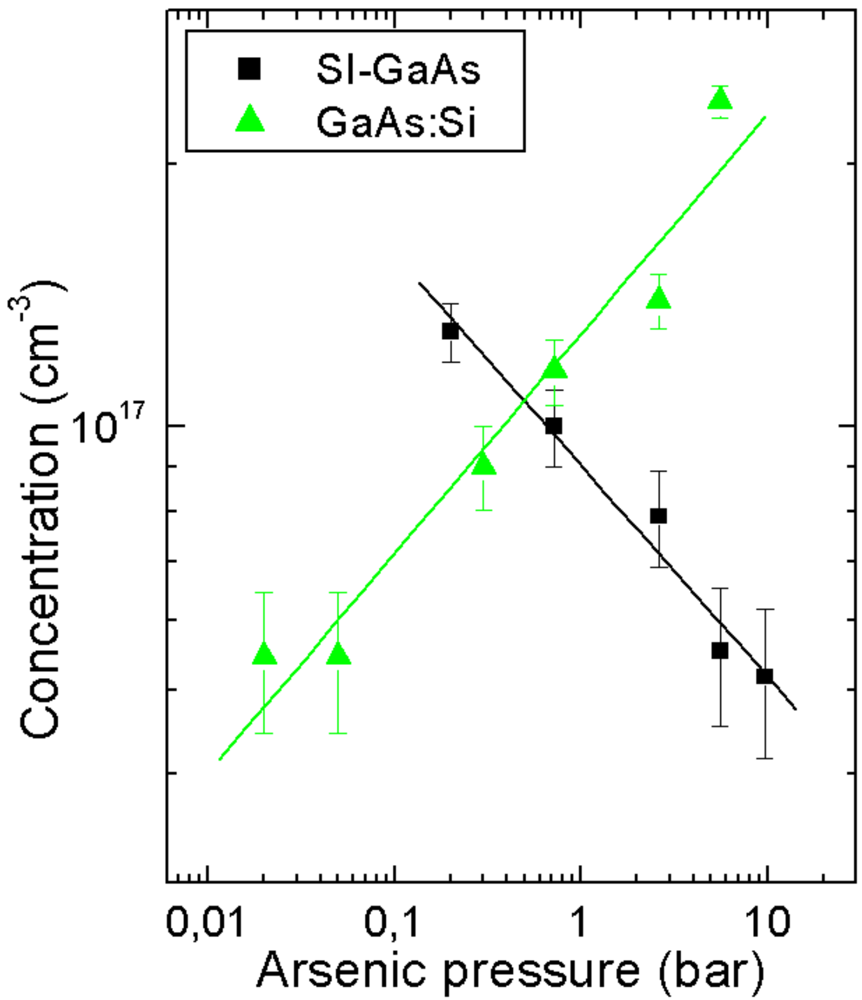
**Mass action law:**

$$[V_{Ga}] = K_{VG} \times p_{As}^{1/4}$$

Fit: $[V_{Ga}\text{-Dopant}] \sim p_{As}^n$

$\rightarrow n = 1/4$

# Comparison of doped and undoped GaAs




Thermodynamic reaction:

$As_{As} \leftrightarrow V_{As} + 1/4As_4^{gas}$

Mass action law:

$[V_{As}] = K_{VAs} \times p_{As}^{-1/4}$

Fit: $[\text{V-complex}] \sim p_{As}^{n}$

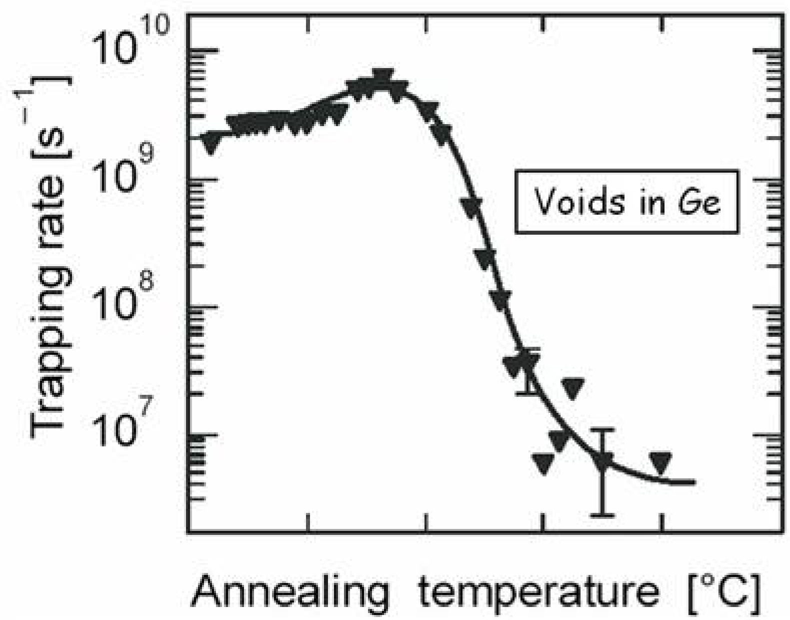
$\rightarrow n = -1/4$

undoped GaAs: As vacancy

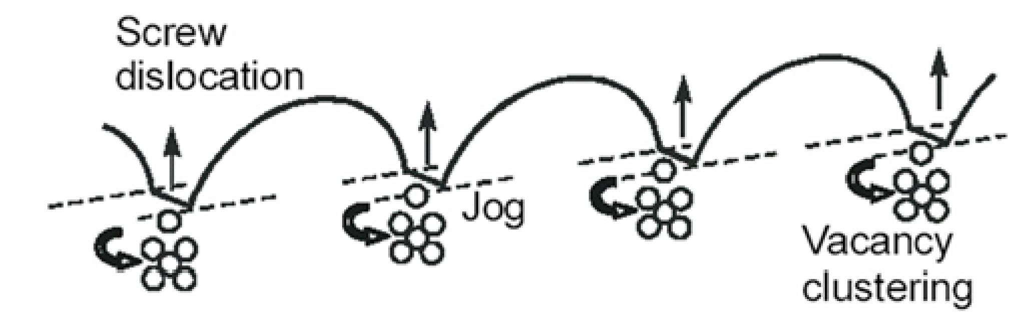
Bondarenko et al., 2003

# Vacancy clusters in semiconductors

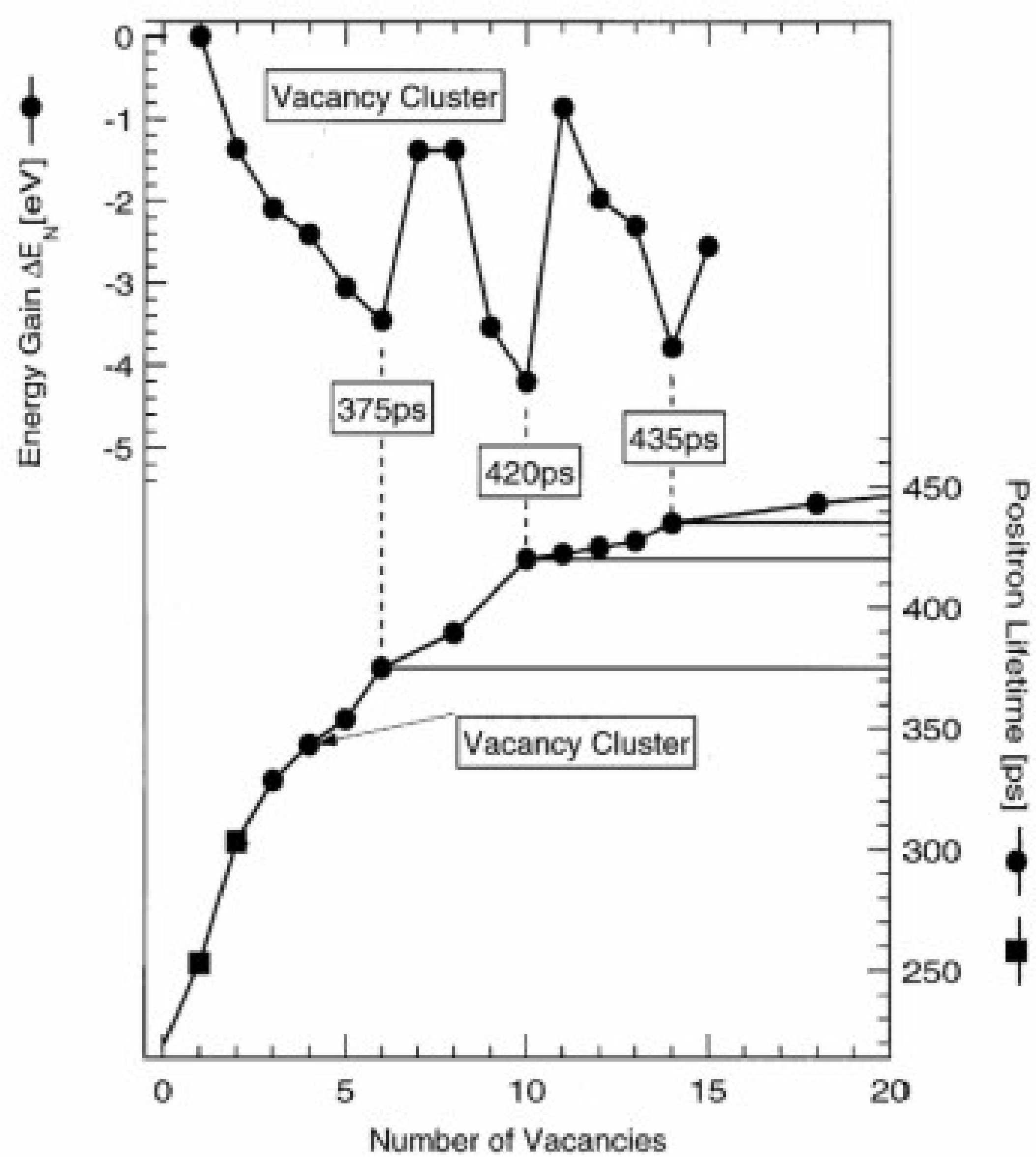
- vacancy clusters were observed after neutron irradiation, ion implantation and plastic deformation
- due to large open volume (low electron density) -> positron lifetime increases distinctly
- example: plastically deformed Ge
- lifetime: $\tau$ = 525 ps
- reason for void formation: jog dragging mechanism
- trapping rate of voids disappears during annealing experiment




Krause-Rehberg et al., 1993



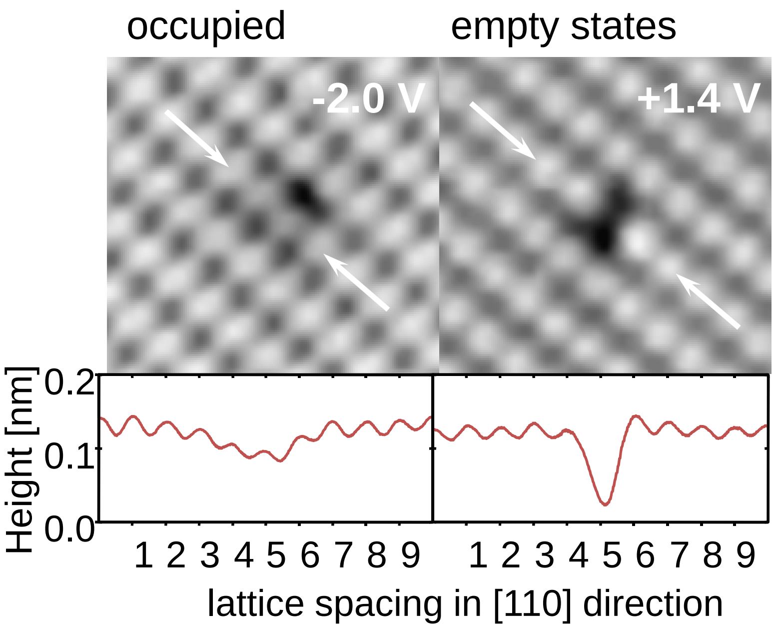
# Theoretical calculation of vacancy clusters in Si

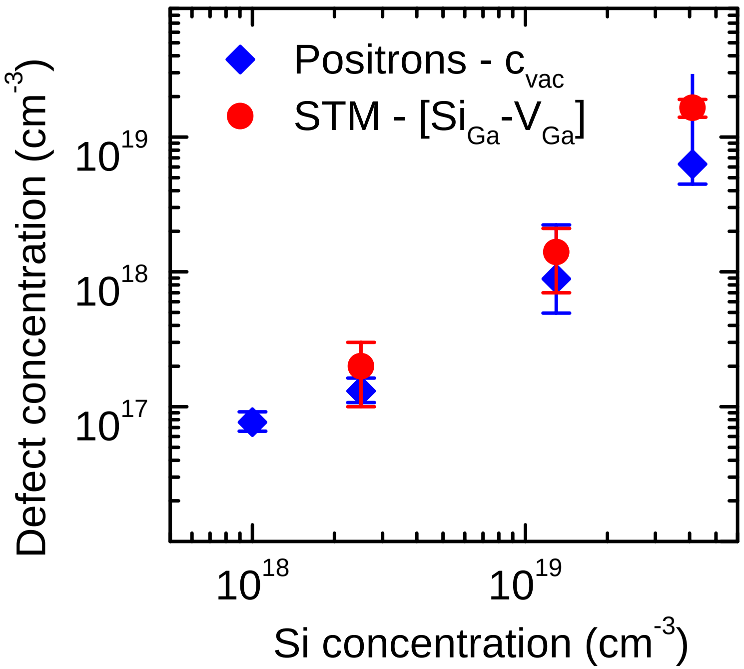



- there are cluster configurations with a large energy gain
- „Magic Numbers“ with 6, 10 und 14 vacancies
- positron lifetime increases distinctly with cluster size
- for n > 10 saturation effect, i.e. size cannot be determined

T.E.M. Staab et al.,
Physica B 273-274 (1999) 501-504

# Identification of $V_{Ga}$-$Si_{Ga}$-Complexes in GaAs:Si



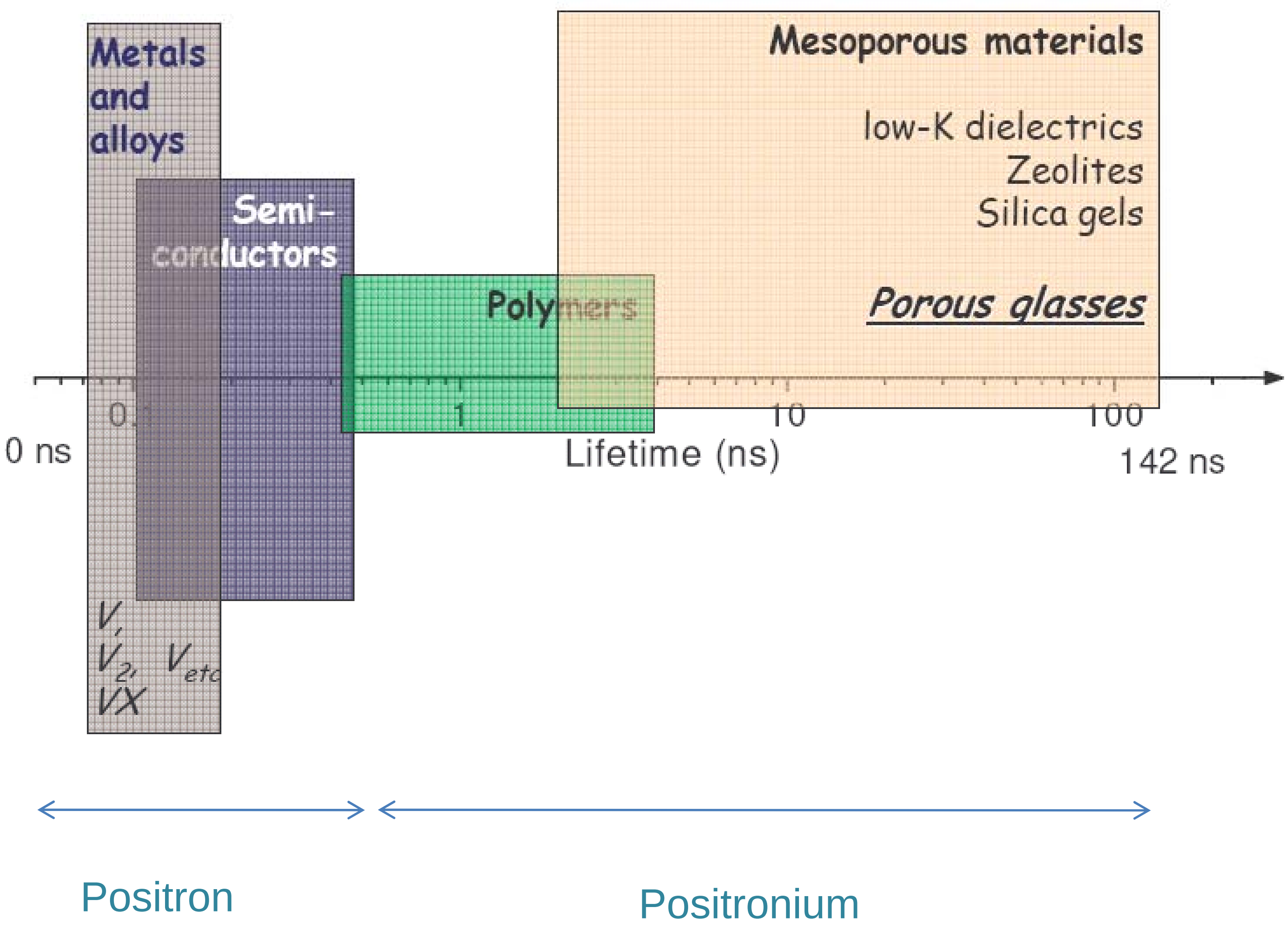

- Scanning tunneling microscopy at GaAs (110)-cleavages planes (by Ph. Ebert, Jülich)
- Defect complex identified as **$V_{Ga}$-$Si_{Ga}$**

- Quantification → Agreement

Mono-Vacancies in GaAs:Si are $V_{Ga}$- $Si_{Ga}$-complexes

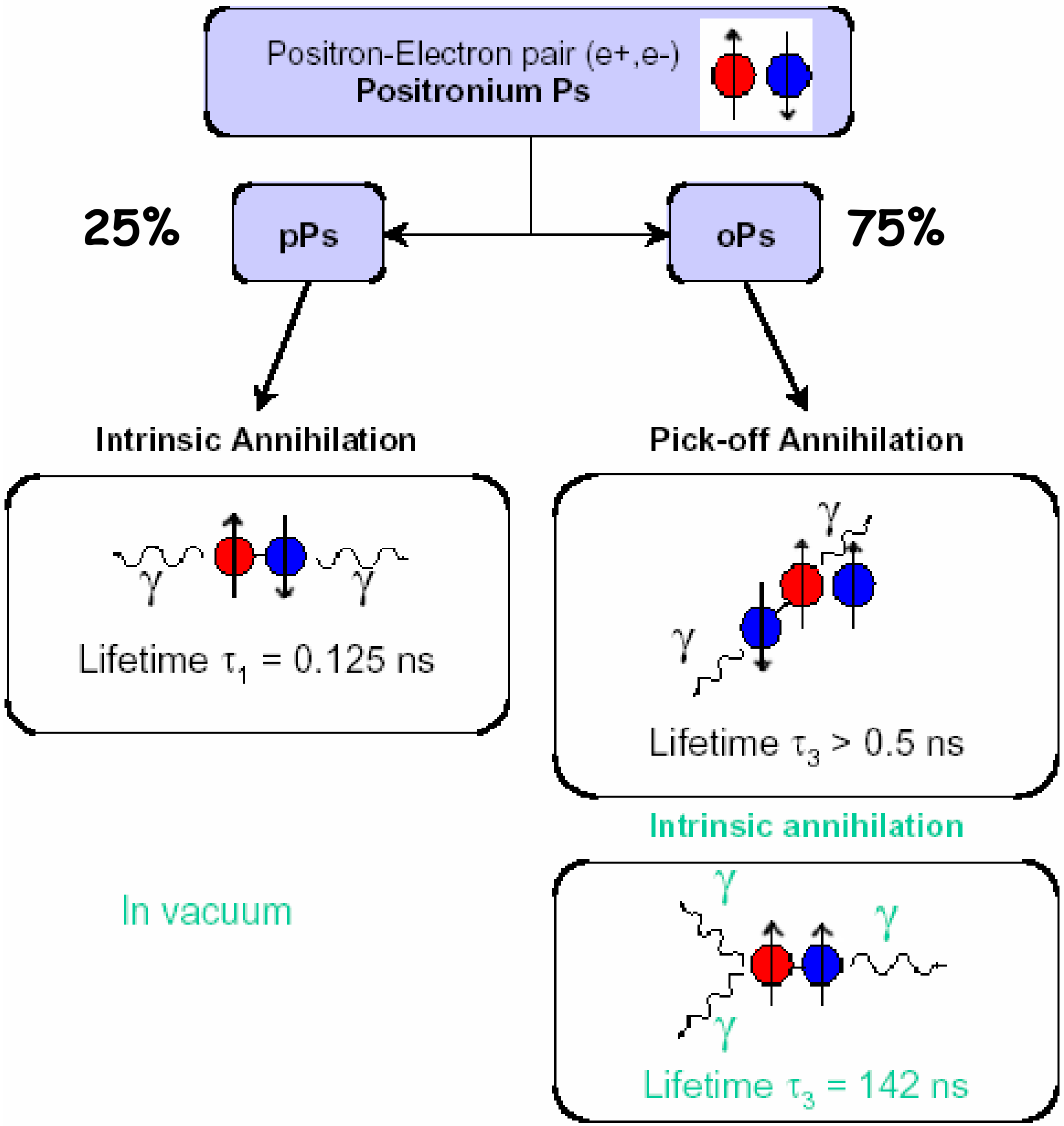
Gebauer et al., Phys. Rev. Lett. **78** (1997) 3334

# Typical Lifetimes



# Principles of PALS: ortho-Positronium

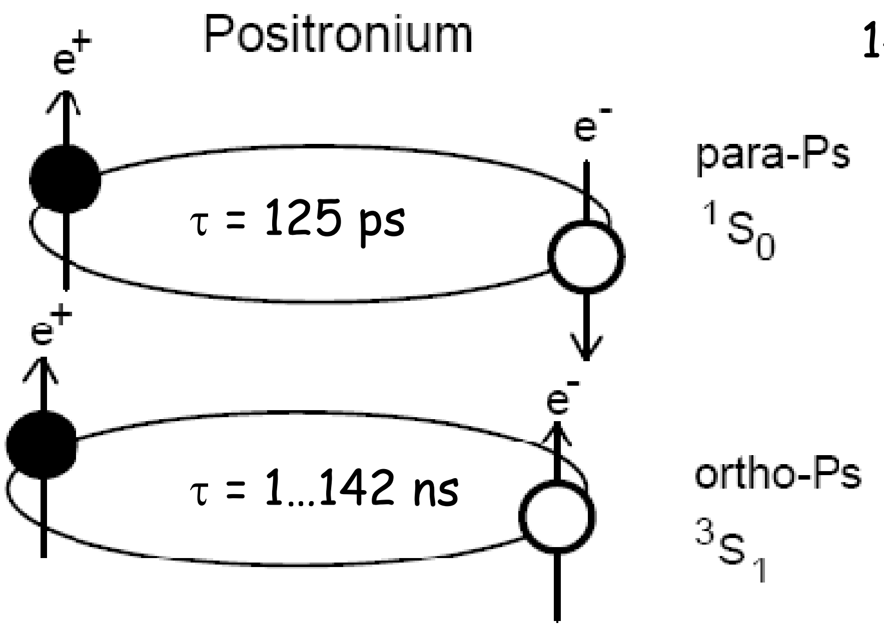
In materials without free electrons Positronium may be formed (Polymers, glass, liquids, gases).

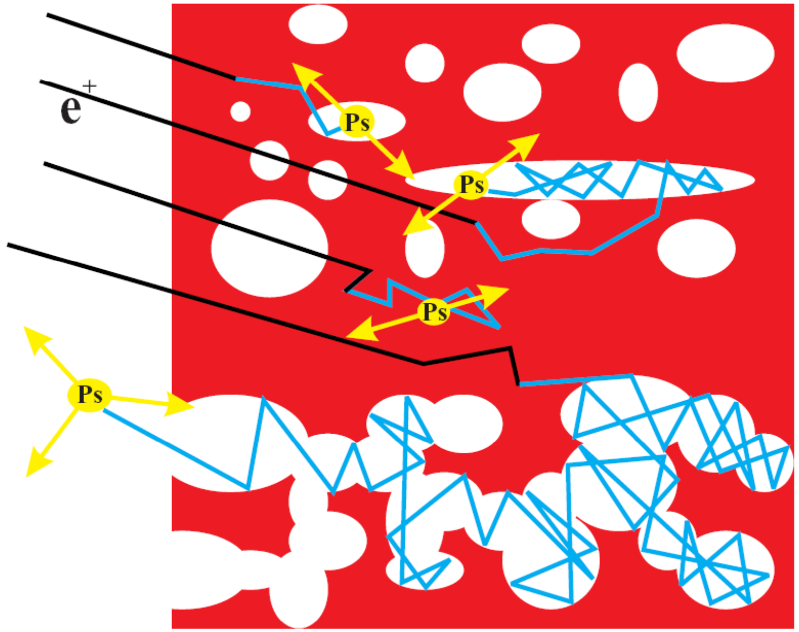


# Pick-off annihilation

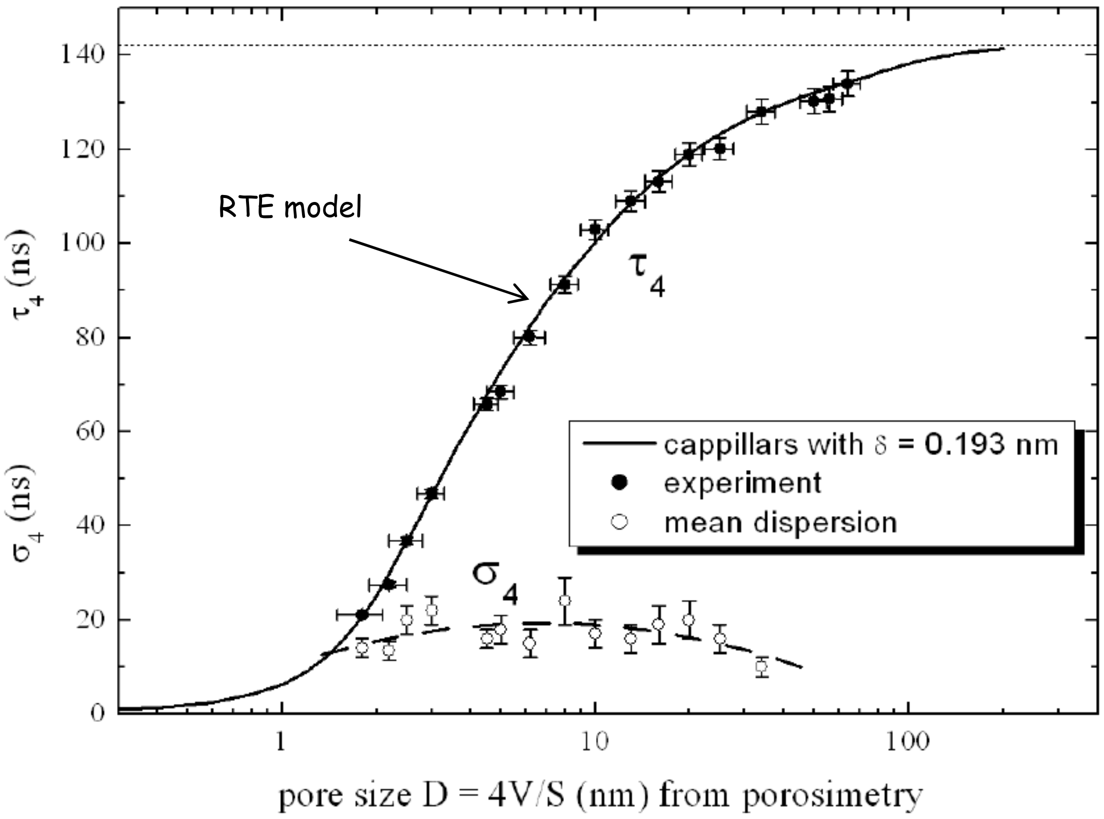
**pick-off annihilation:**

- o-Ps is converted to p-Ps by capturing an electron with anti-parallel spin
- happens during collisions at walls of pore
- lifetime decreases rapidly
- lifetime is function of pore size 1.5 ns to 142 ns



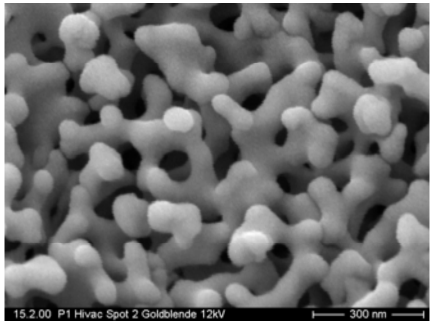



# o-Ps lifetime $\tau_4$ versus pore size



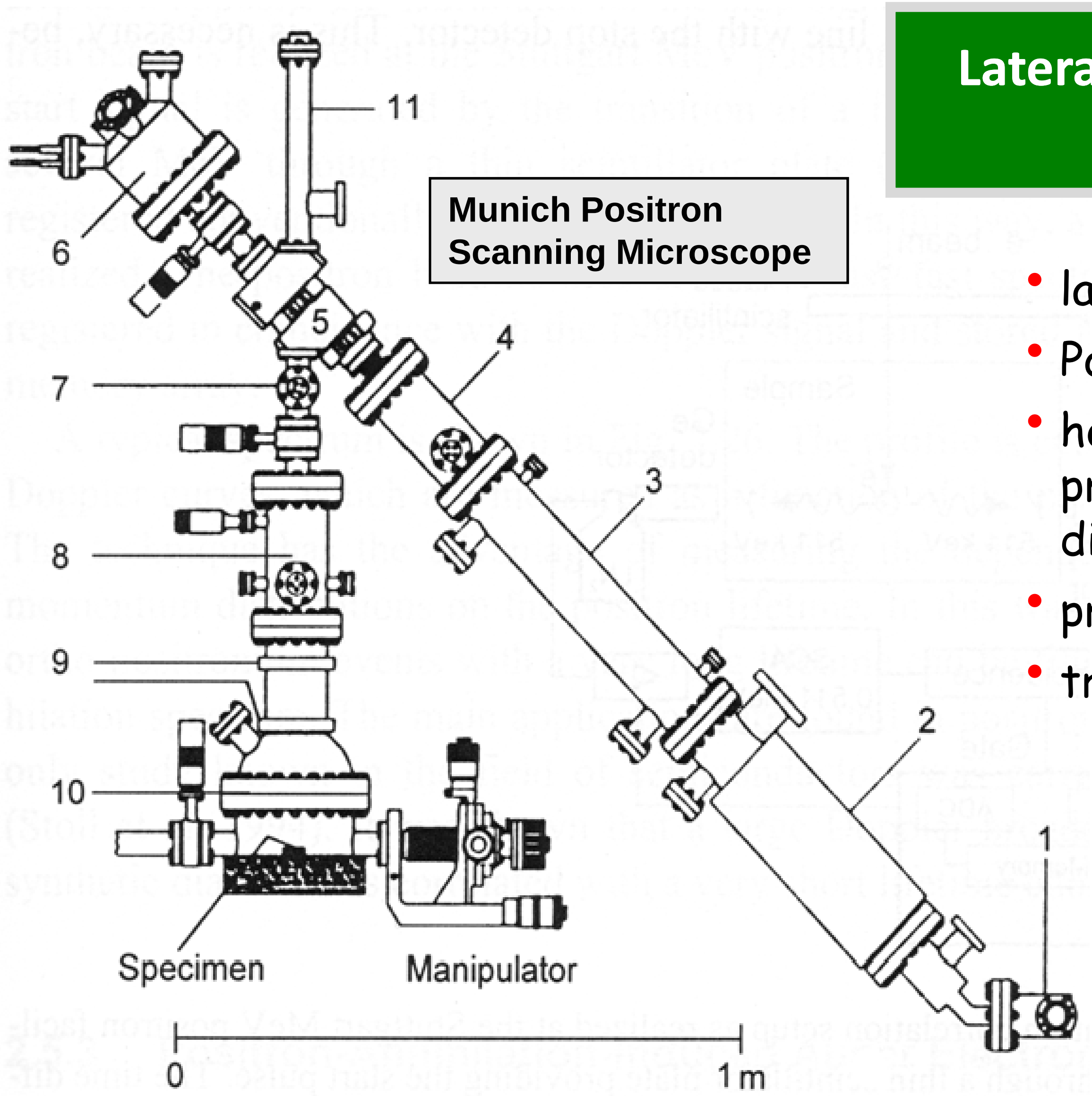

S.Thränert, Dissertation, MLU Halle 2008

- we measured porous CPG glass in a broad pore size range




- given pore size obtained by $N_2$-adsorption and/or mercury intrusion technique
- for T=300 K fair agreement to the RTE model for large pores

# Lateral Resolution with Positron-Scanning-Microscope

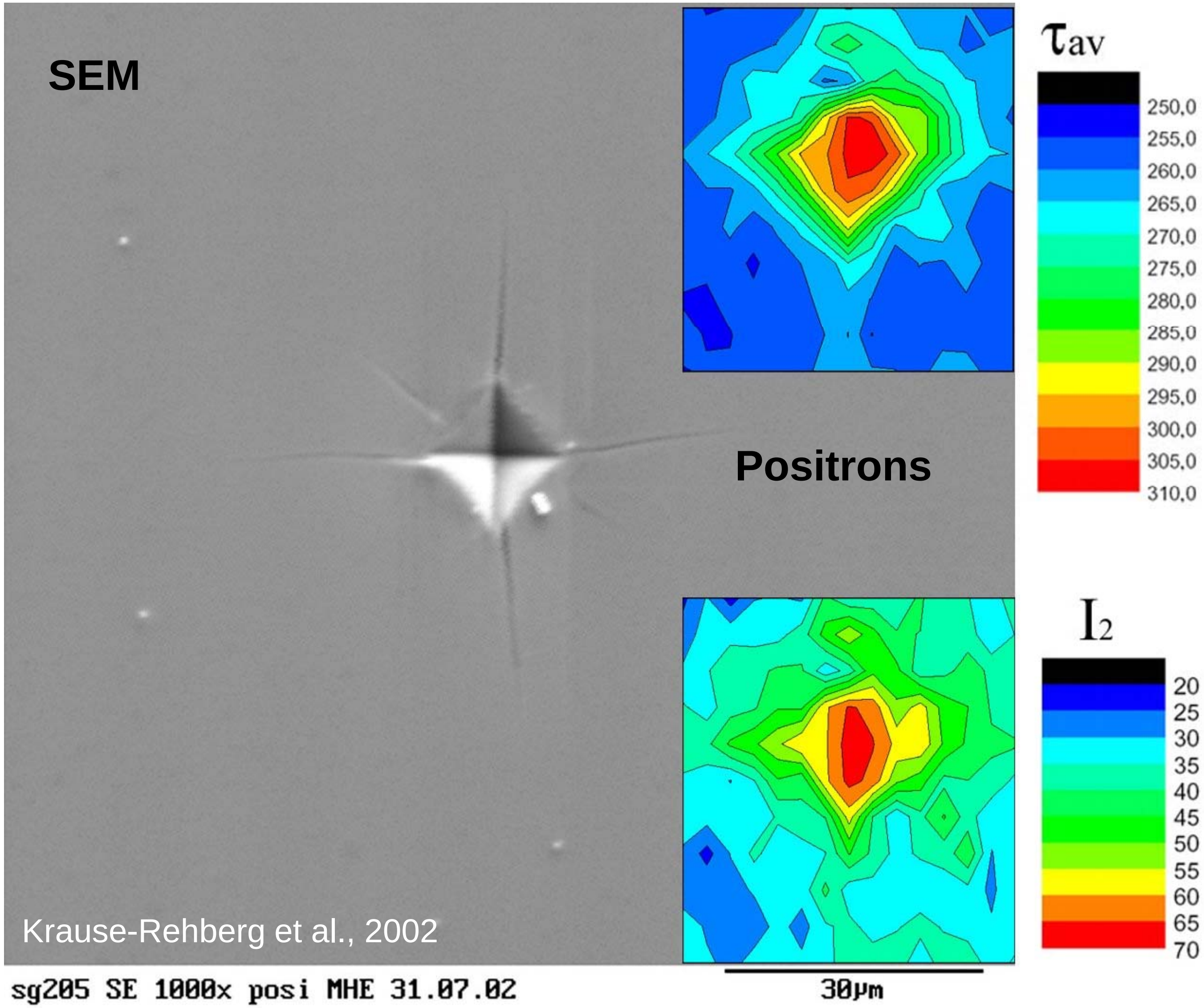



W. Triftshäuser et al., NIM B **130** (1997) 265

- lateral resolution 2 μm
- Positron lifetime spectroscopy
- however lateral resolution principally limited by positron diffusion ($L_+ \approx$ 100nm)
- problem: low count rate
- transfer to FRM-II

# Microhardness indentation in GaAs

- Comparison of SEM and Munich Positron Scanning Microscope
- problem here at the moment: intensity
- hope: strong positron source at FRM-II Garching or EPOS project in Rossendorf



## Extended Concept of EPOS (ELBE Positron Source)

### MePS

**M**ono**e**nergetic **P**ositron **S**pectroscopy

- Cave 111b / Lab 111d
- monoenergetic (slow) positrons
- pulsed system
- LT, CDBS, AMOC
- Still under construction

Information Depth: 0…5 µm

### CoPS

**Co**nventional **P**ositron **S**pectroscopy

- LT, CDBS, AMOC
- using $^{22}Na$ foil sources
- He-cryostat
- automated system
- digital detector system

Information Depth: 10…200 µm

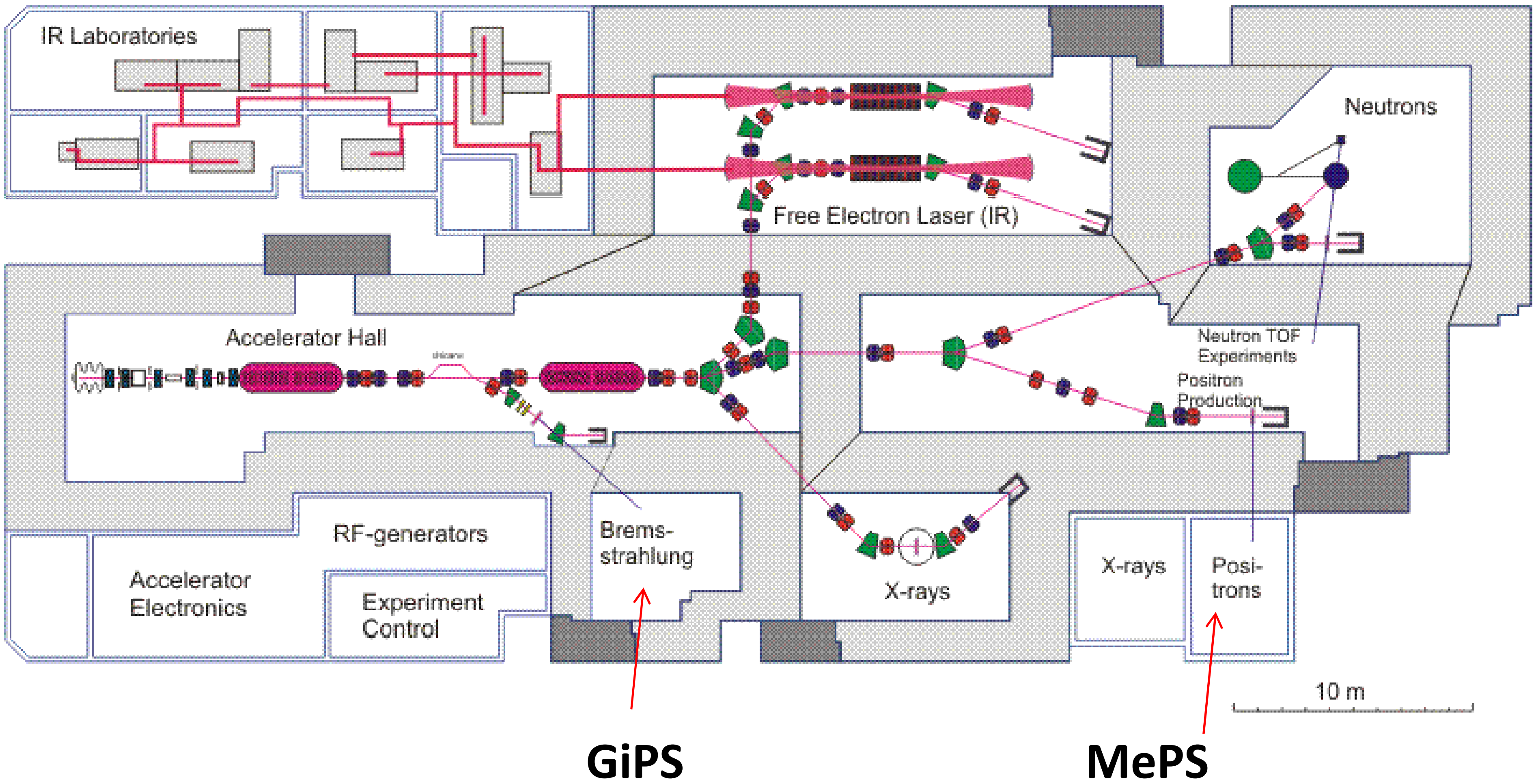
### GiPS

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

- Cave 109 (nuclear physics)
- Positron generation by Bremsstrahlung
- Information in complete bulky sample (up to 100 $cm^3$)
- all relevant positron techniques (LT, CDBS, AMOC)

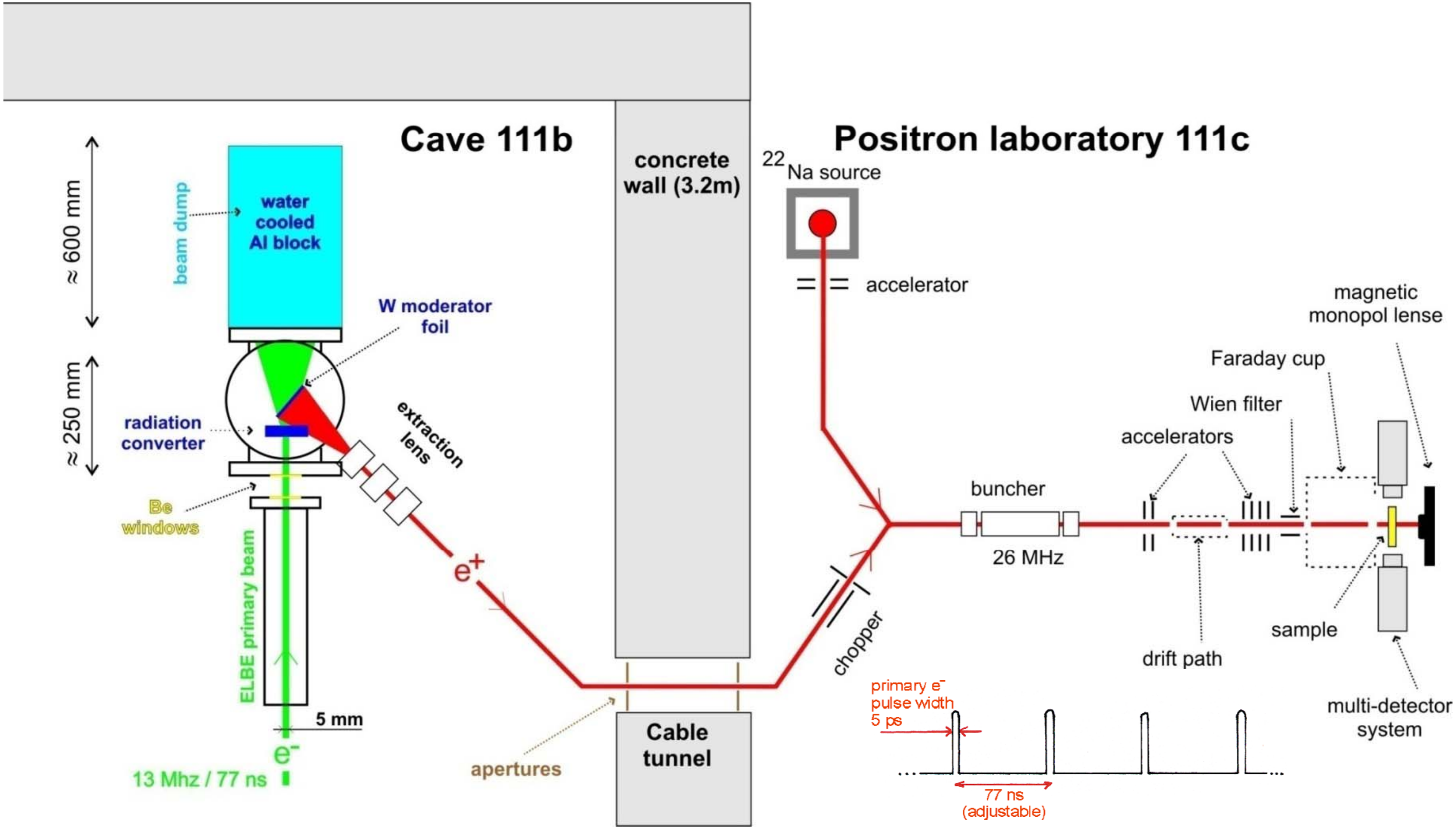
Information Depth: 0.1 mm …5 cm

# Ground plan of the ELBE hall

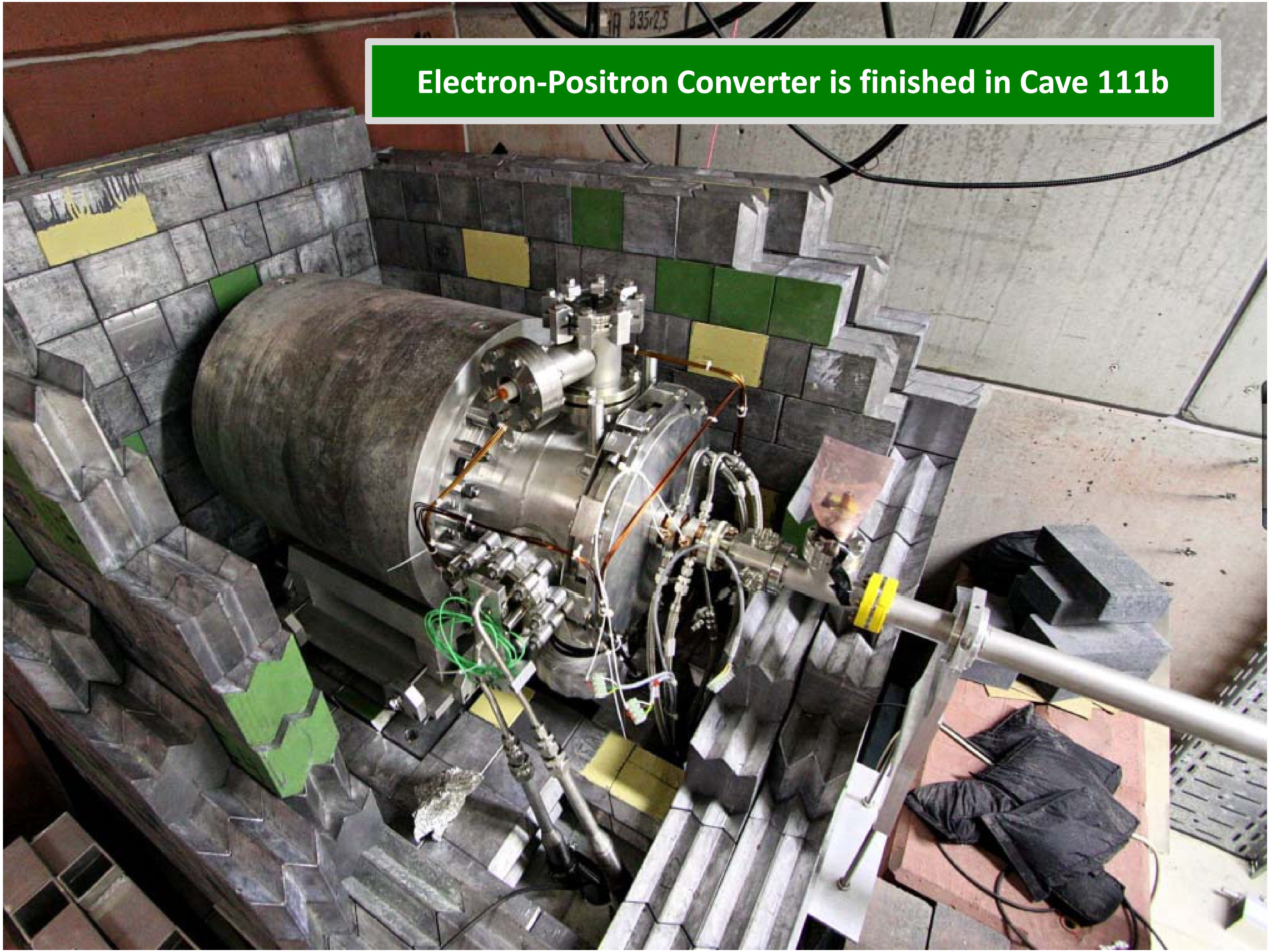


# Progress of Mono-energetic Positron Beam

- 40 MeV, 1 mA, 26 MHz repetition time in cw mode; lifetime, CDBS and AMOC with slow $e^+$
- Retain original time structure for simplicity and best time resolution

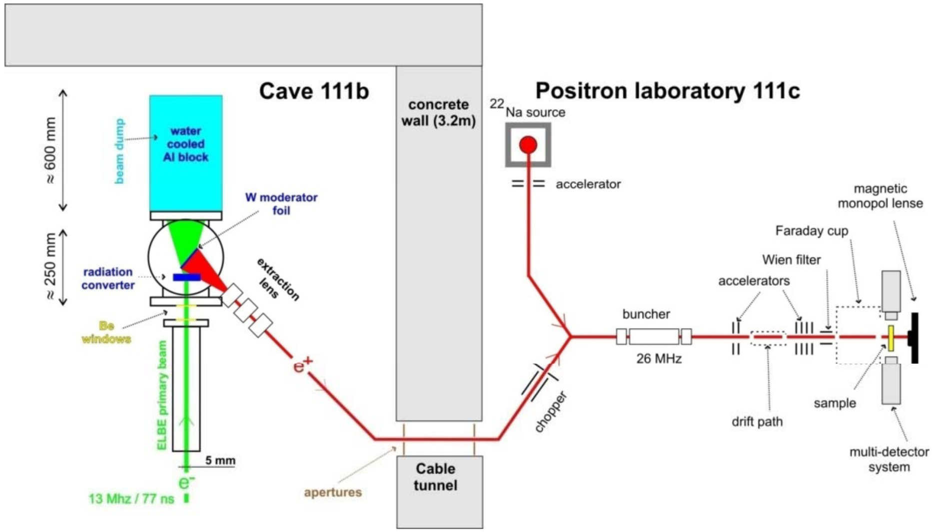


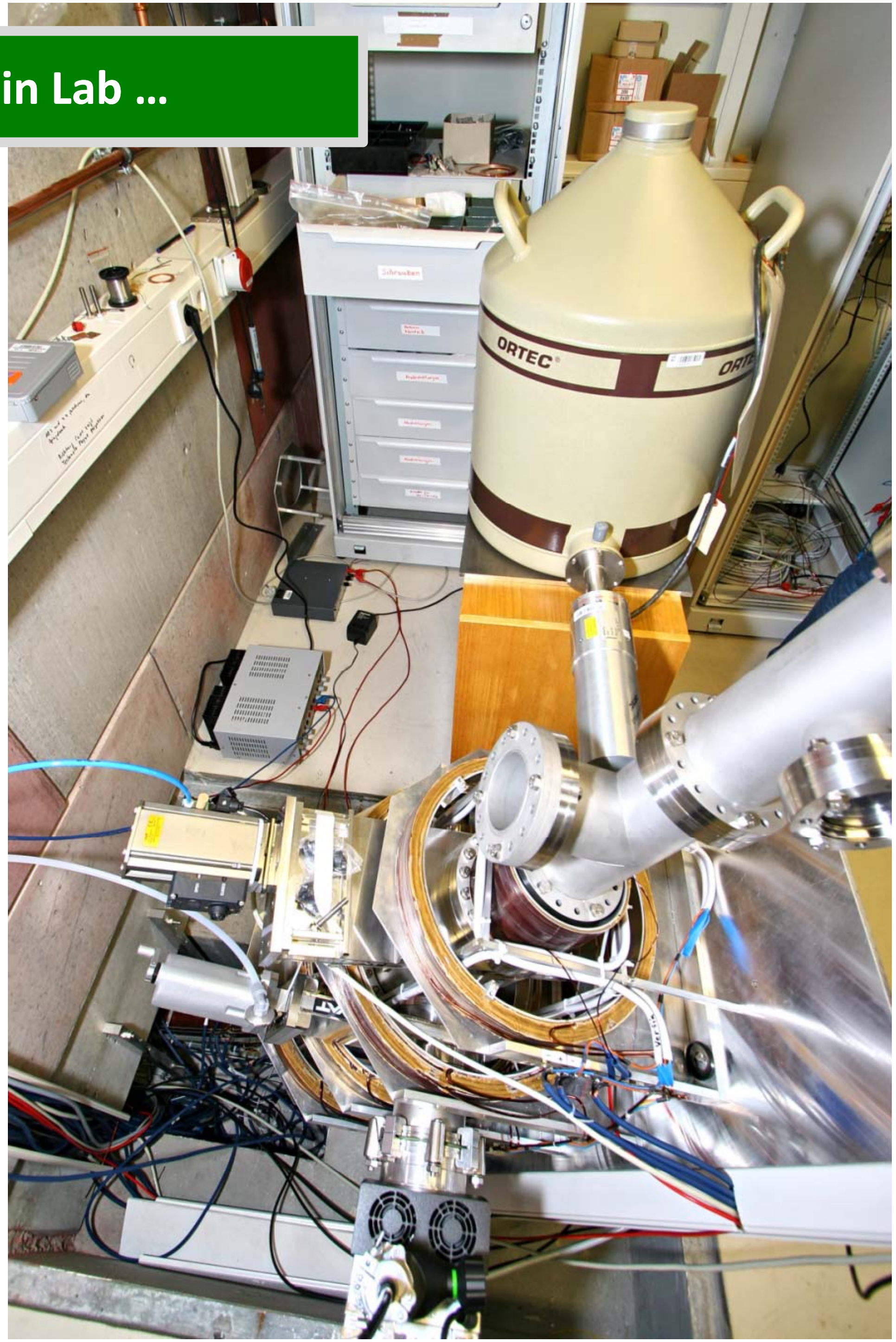
# Electron-Positron Converter is finished in Cave 111b

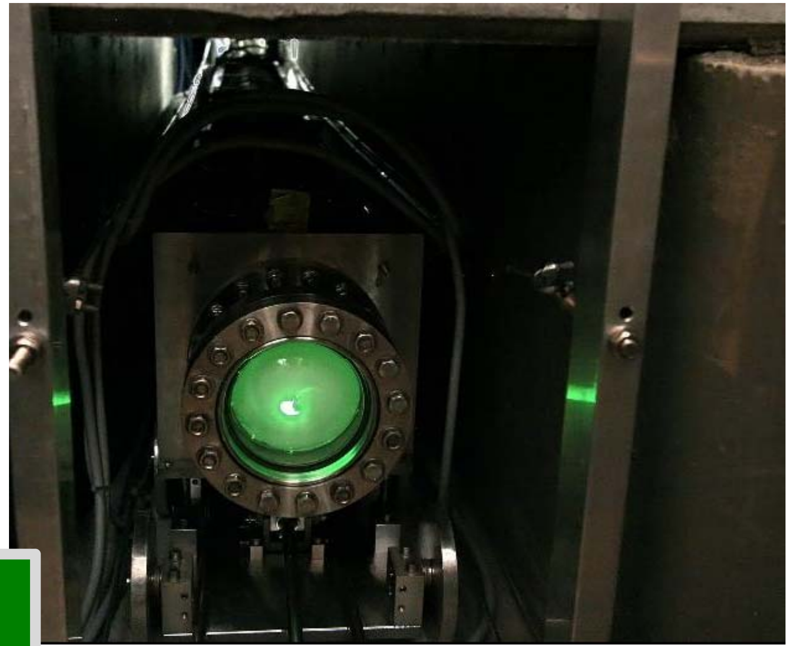


# Still waiting for $\gamma$ Quanta in Lab …

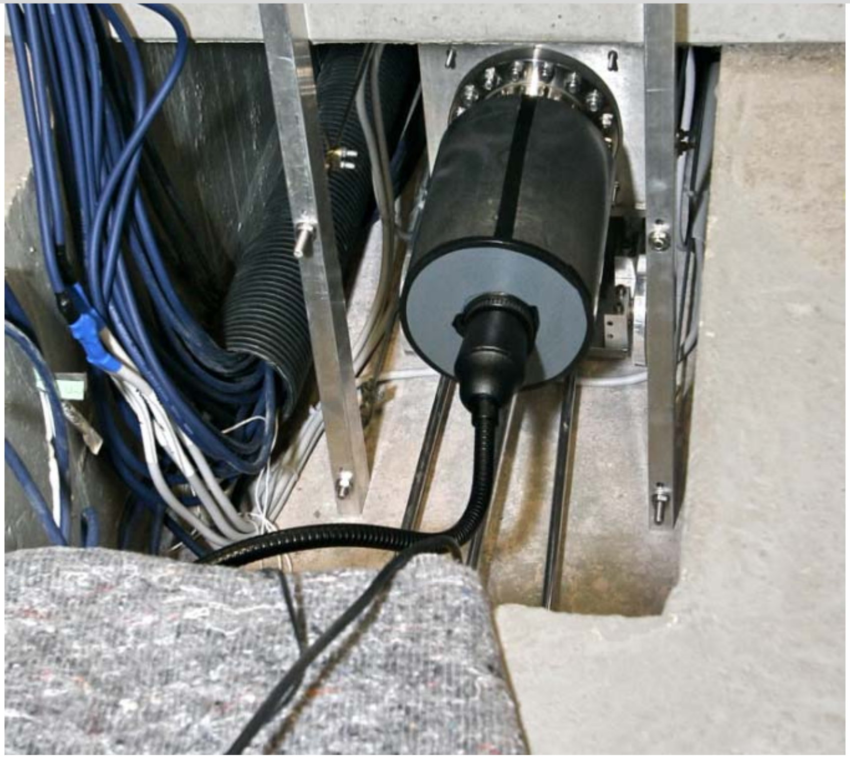
- Problem: 10 x 2 steering coils must be adjusted
- automatic LabView program is looking for annihilation gamma at end of beam line





# Test of Beam Guidance with an Electron Source



# Gamma-induced Positron Spectroscopy

ELSEVIER

Nuclear Instruments and Methods in Physics Research A 495 (2002) 154–160

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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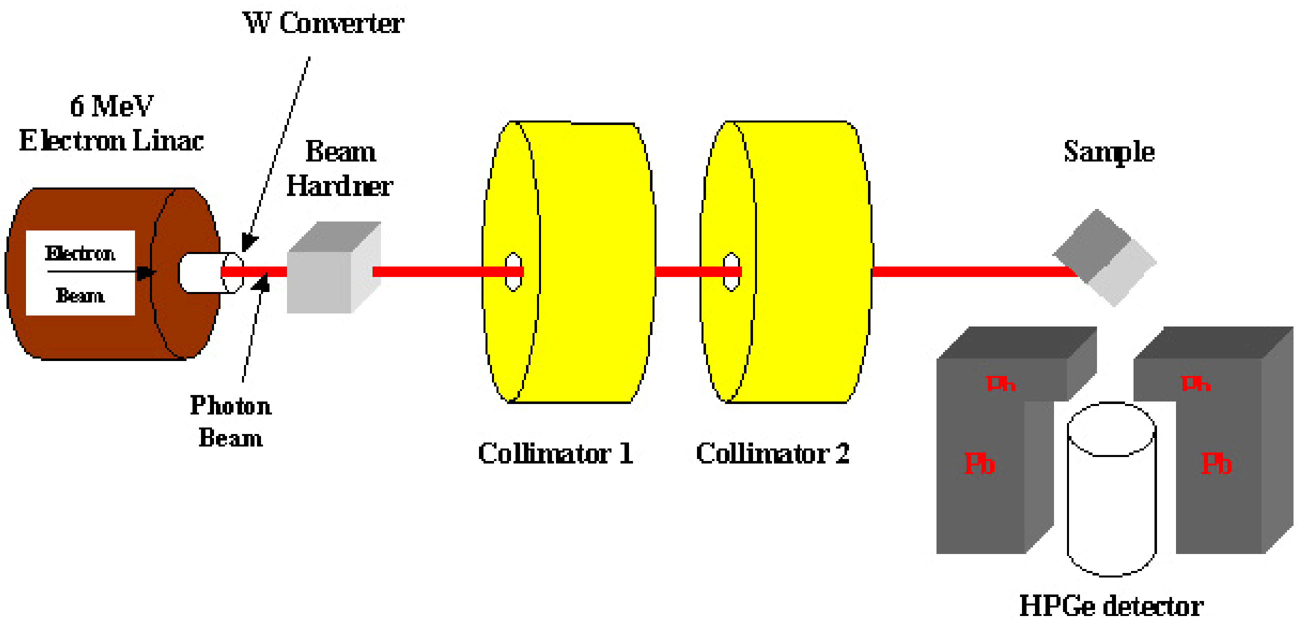


# First spectra

**Advantages**

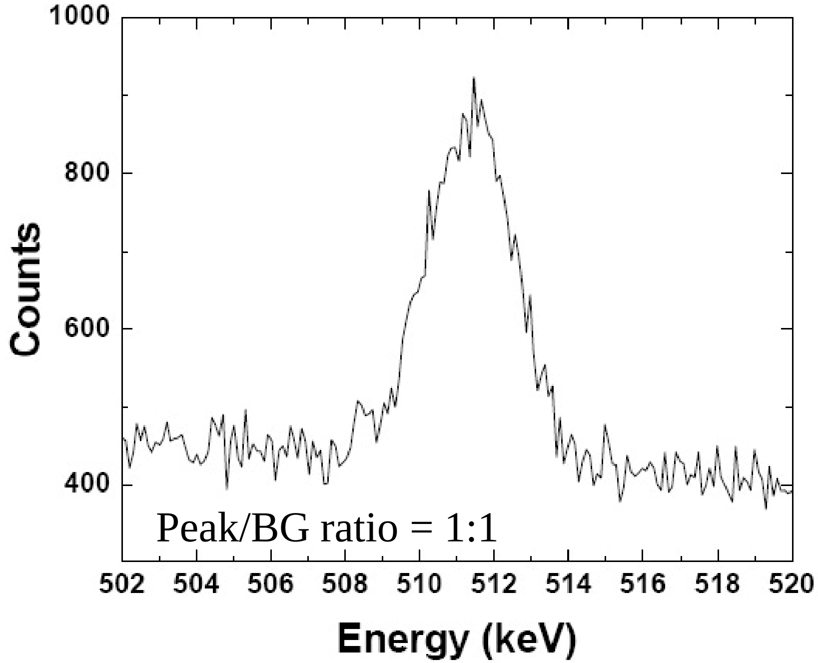
- information depth 0.1 … 5 cm; whole sample
- ideal for bulky samples (NDT), liquids, gases, biological objects, coarse powder, dispersions …

**Disadvantage of slow LINACs**

- Use of "normal" LINAC with 200 Hz has the problem of high gamma flux in only very few bunches
- Count rate very low, thus no coincidence techniques applicable such as CDBS or AMOC
- Peak / BG ratio bad (1:1)
- no lifetime spectroscopy possible
- No coincidence techniques applicable for improvement of peak-background ratio

All this **disadvantages can be overcome** by use of a superconducting LINAC with > 10 MHz

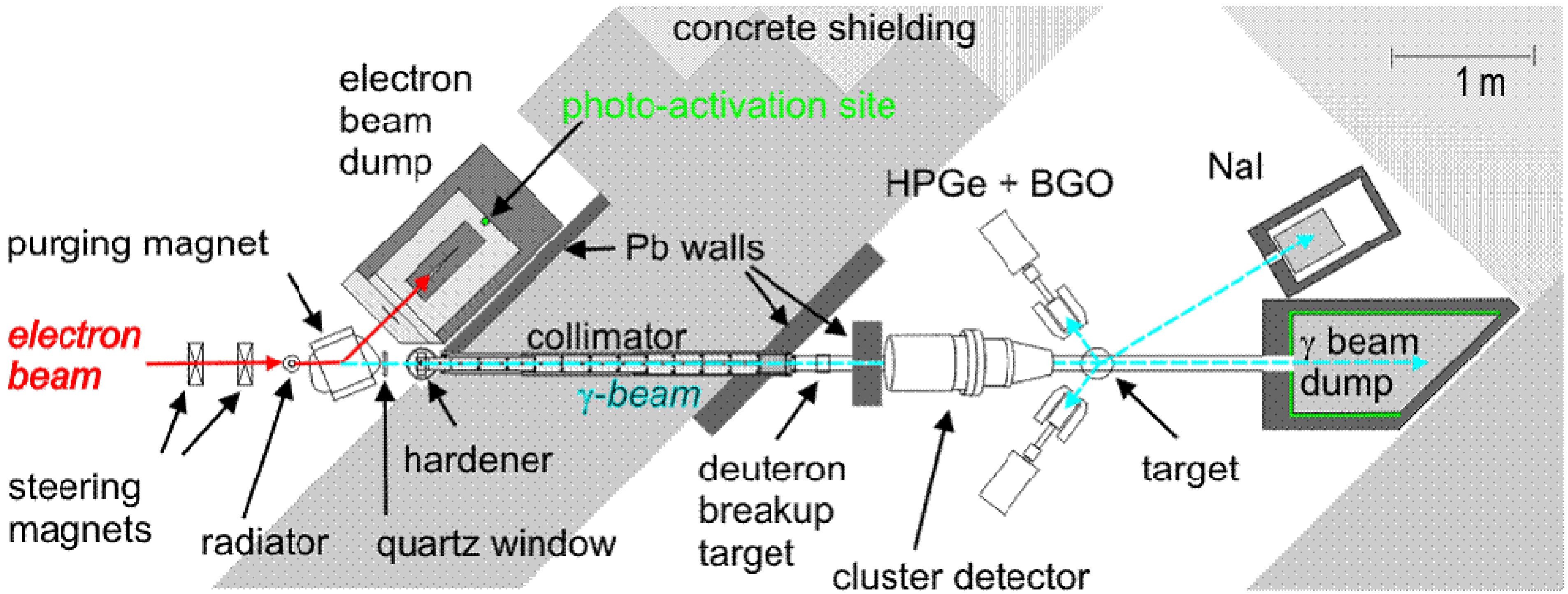
Annihilation line in steel



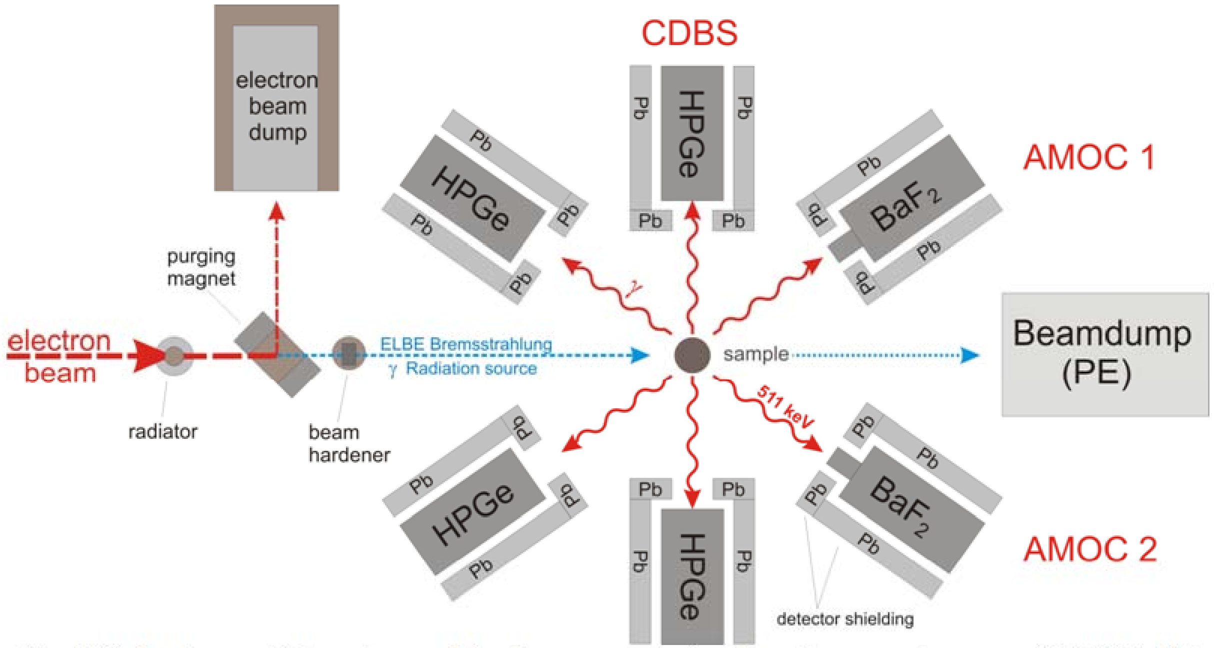

F.A. Selim et al., NIM B 192 (2002) 197

# Bremsstrahlung Gamma Source of ELBE (FZ Dresden-Rossendorf)

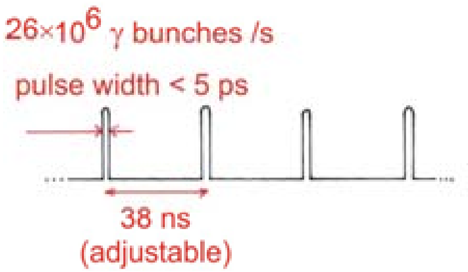
- Pulsed gamma source using superconductive Linac ELBE
  - repetition frequency 26 MHz (or smaller by factor $2^n$) in CW mode!
  - bunch length < 5 ps
  - up to 20 MeV (we used 16 MeV), no activation of samples by γ-n processes was found
  - average electron current 1 mA = 20 kW beam power; electron beam dump outside lab
  - thus gamma background at target position is very low (Ge detectors with 100% efficiency)
- **Ideal for GiPS ! Is now part of EPOS project – user dedicated positron source.**



# Setup extended by $BaF_2$ detectors for lifetime measurement

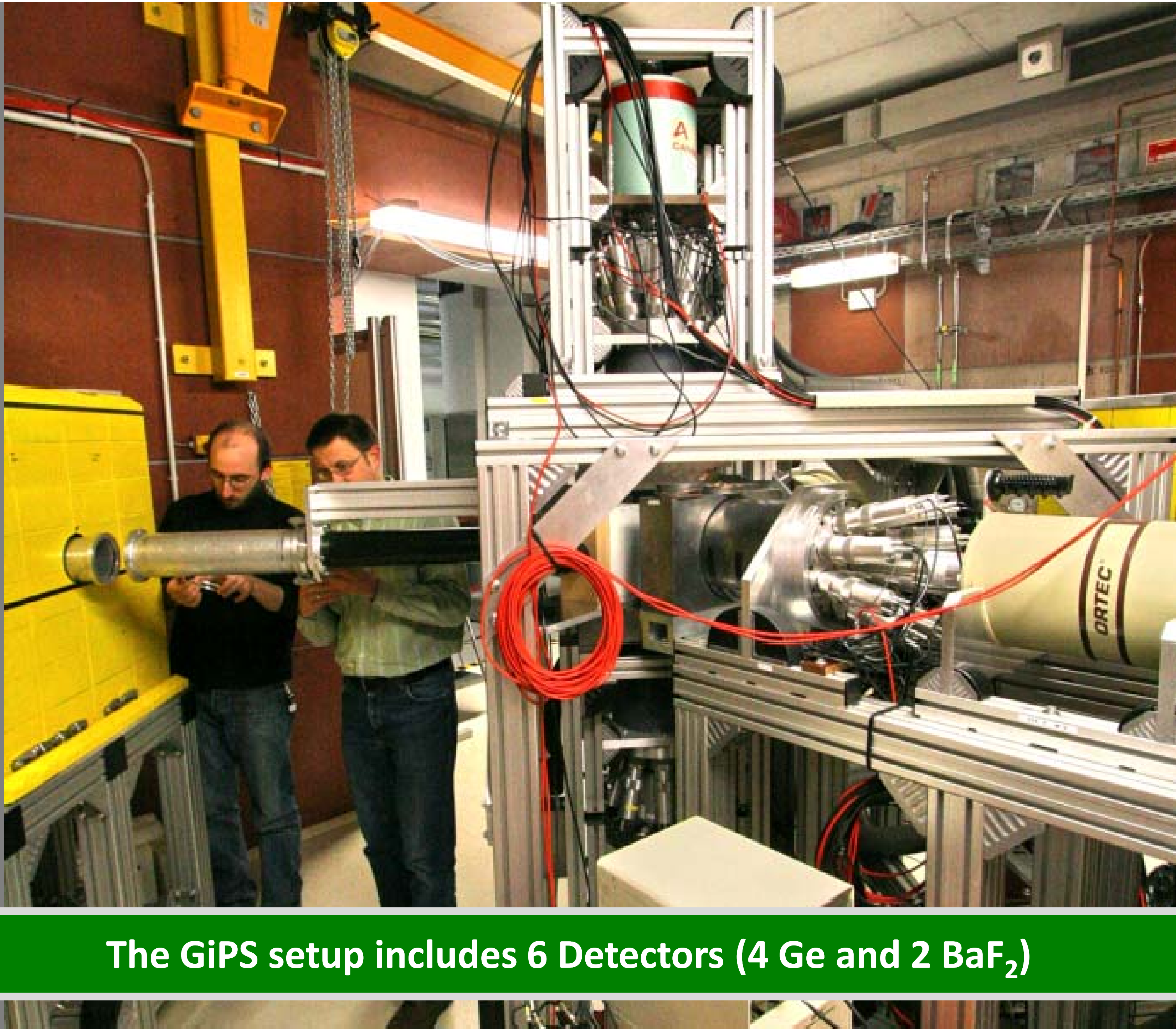



Simplified scheme of the setup used for the gamma induced positron spectroscopy (GiPS) facility at EPOS




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

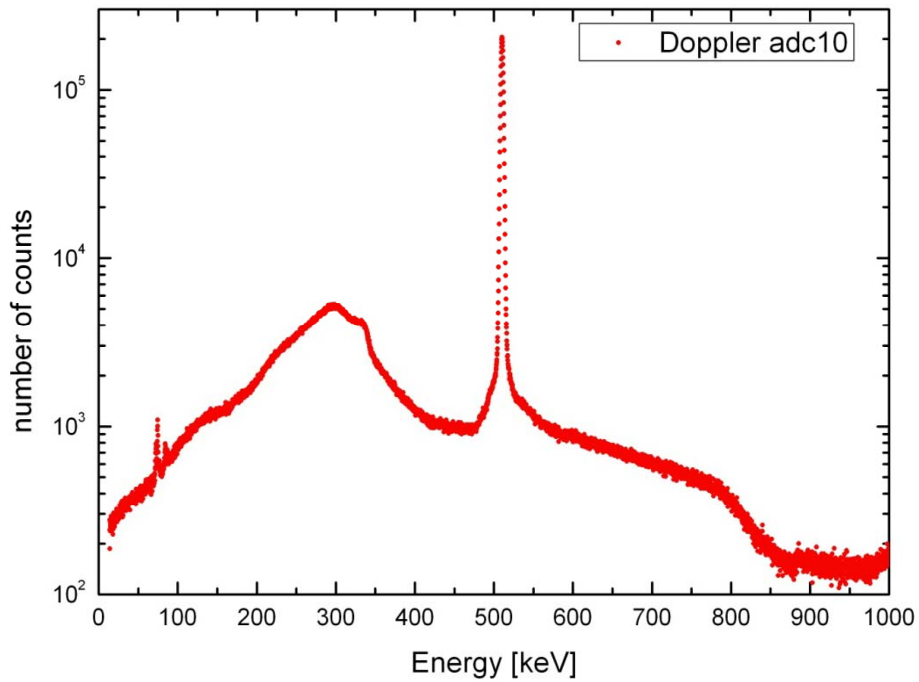
- 3 coincident setups were used: 2 AMOC and 1 CDBS spectrometer; only coincident detection ensures high spectra quality
- all scattered quanta appear within positron lifetime – time coincidence alone does not reduce background at all
- but distance helps: for 2 x 511 keV quanta in coincidence the distance dependence is proportional to $r^{-2}$
- for arbitrary scattered gamma it is $\propto r^{-4}$

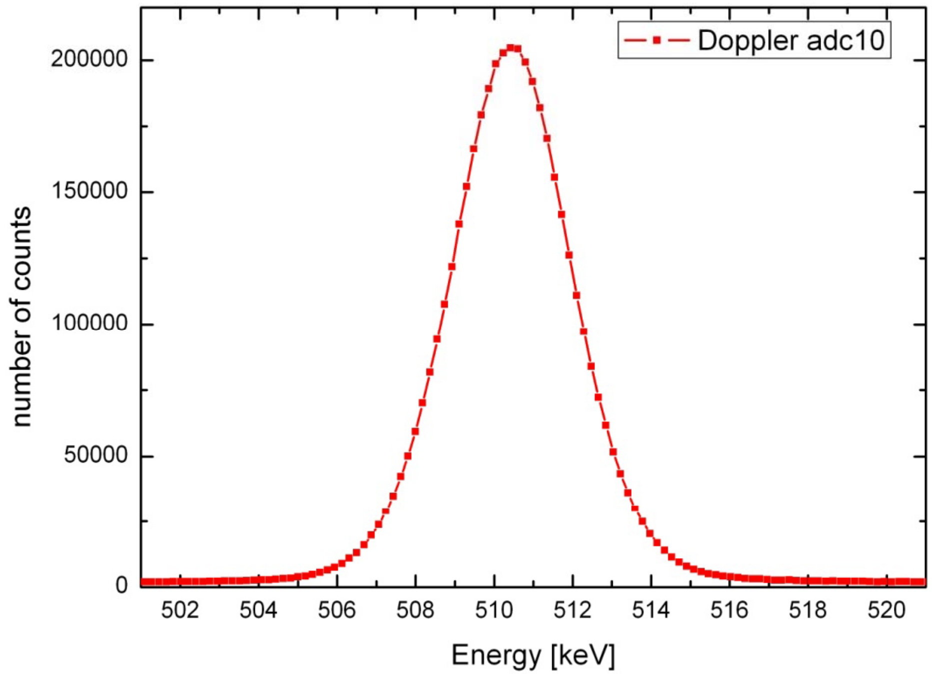



The GiPS setup includes 6 Detectors (4 Ge and 2 $BaF_2$)

# Single-channel Ge Spectrum of annealed Fe

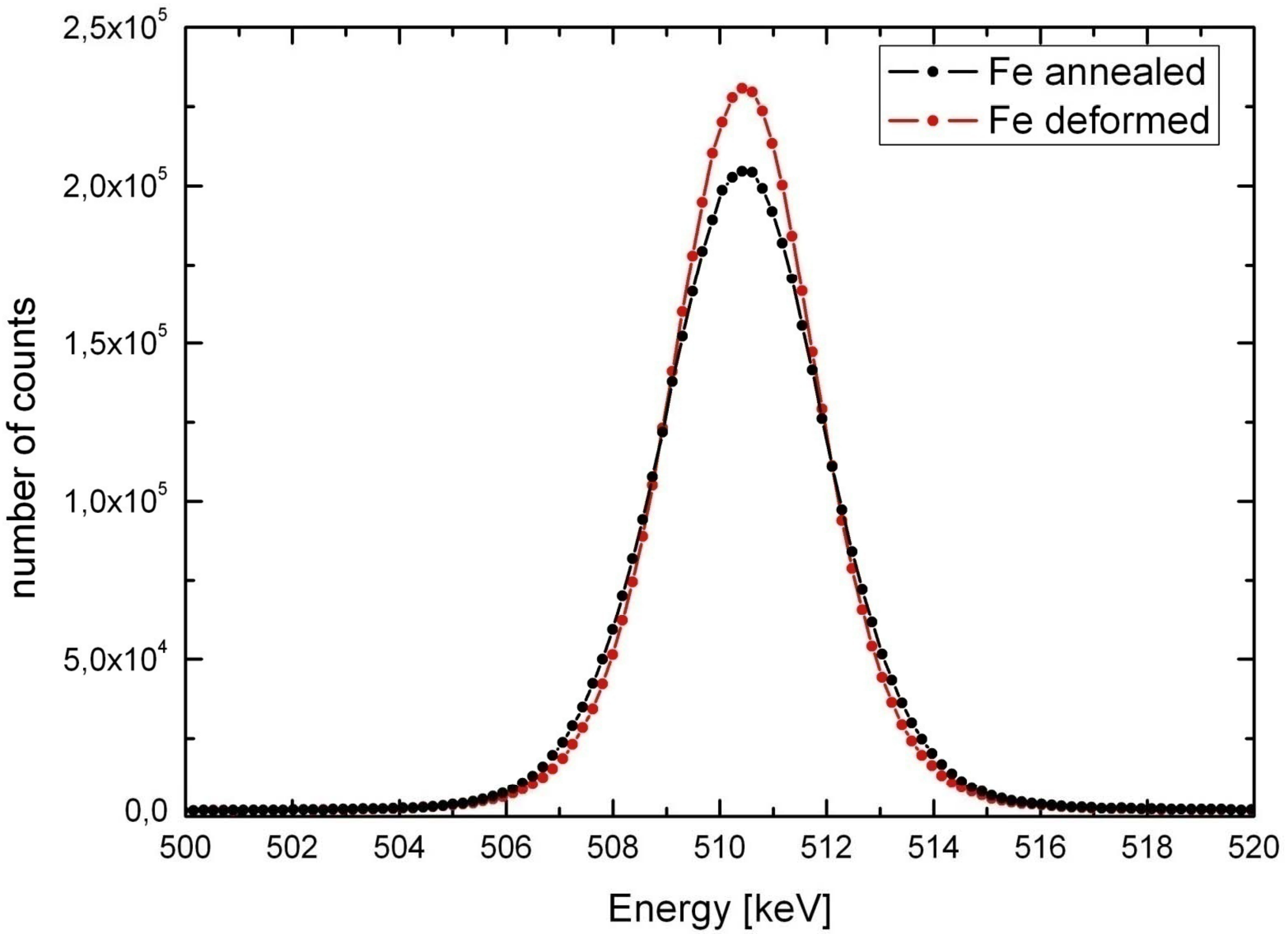
- count rate about 20 kHz (100 kHz would be theoretically possible); total counts in example: $8 \times 10^6$
- about 50% of intensity in 511 peak of annihilation line
- decrease below 350 keV due to 5 mm Cu absorber plates in front of Ge detectors
- detection with analog electronics



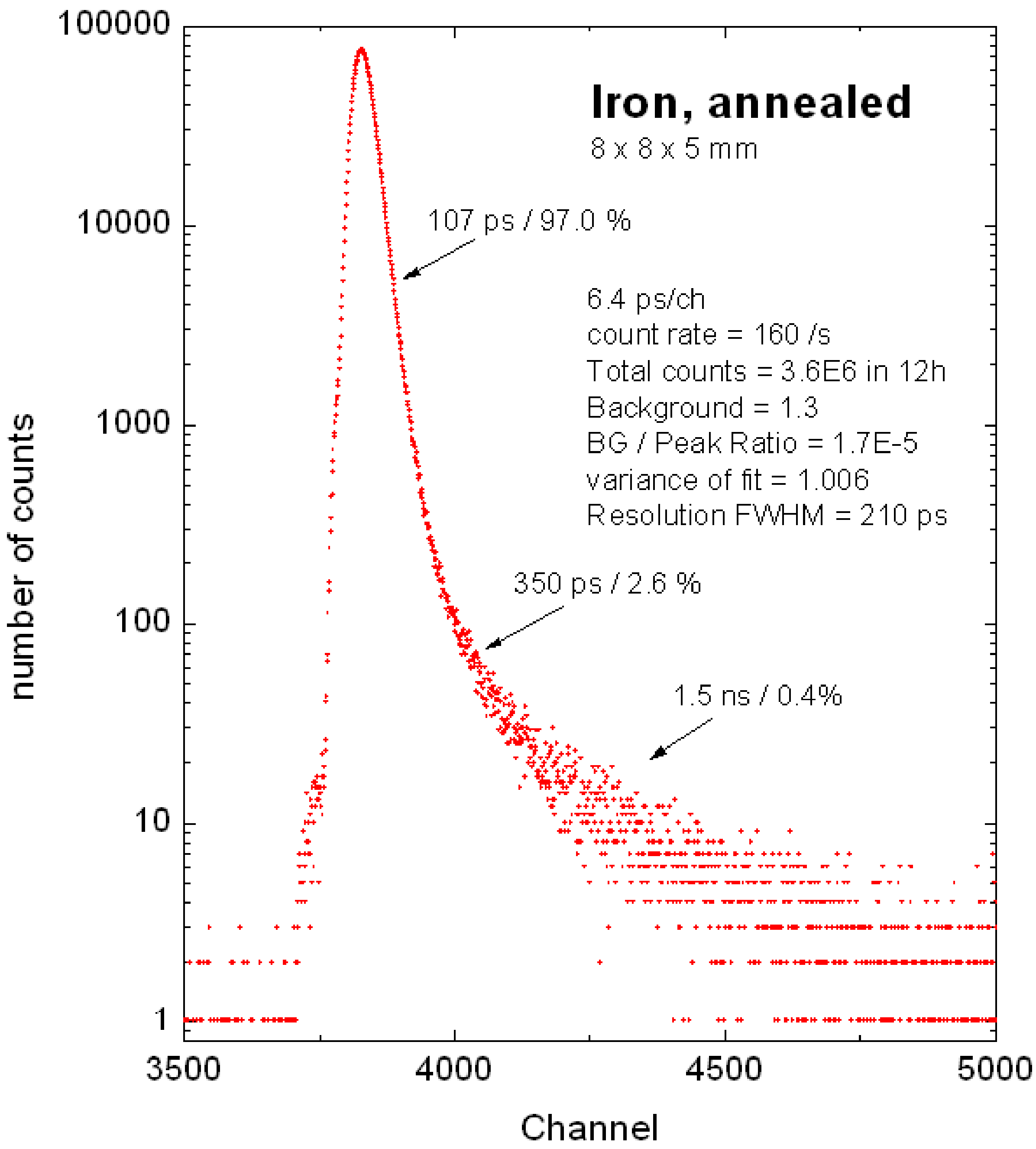
# Comparison annealed and deformed Fe

- expected behavior
- curve of deformed Fe is distinctly taller due to open-volume defects and thus increased fraction of annihilation with valence electrons (small energies – small Doppler shift)



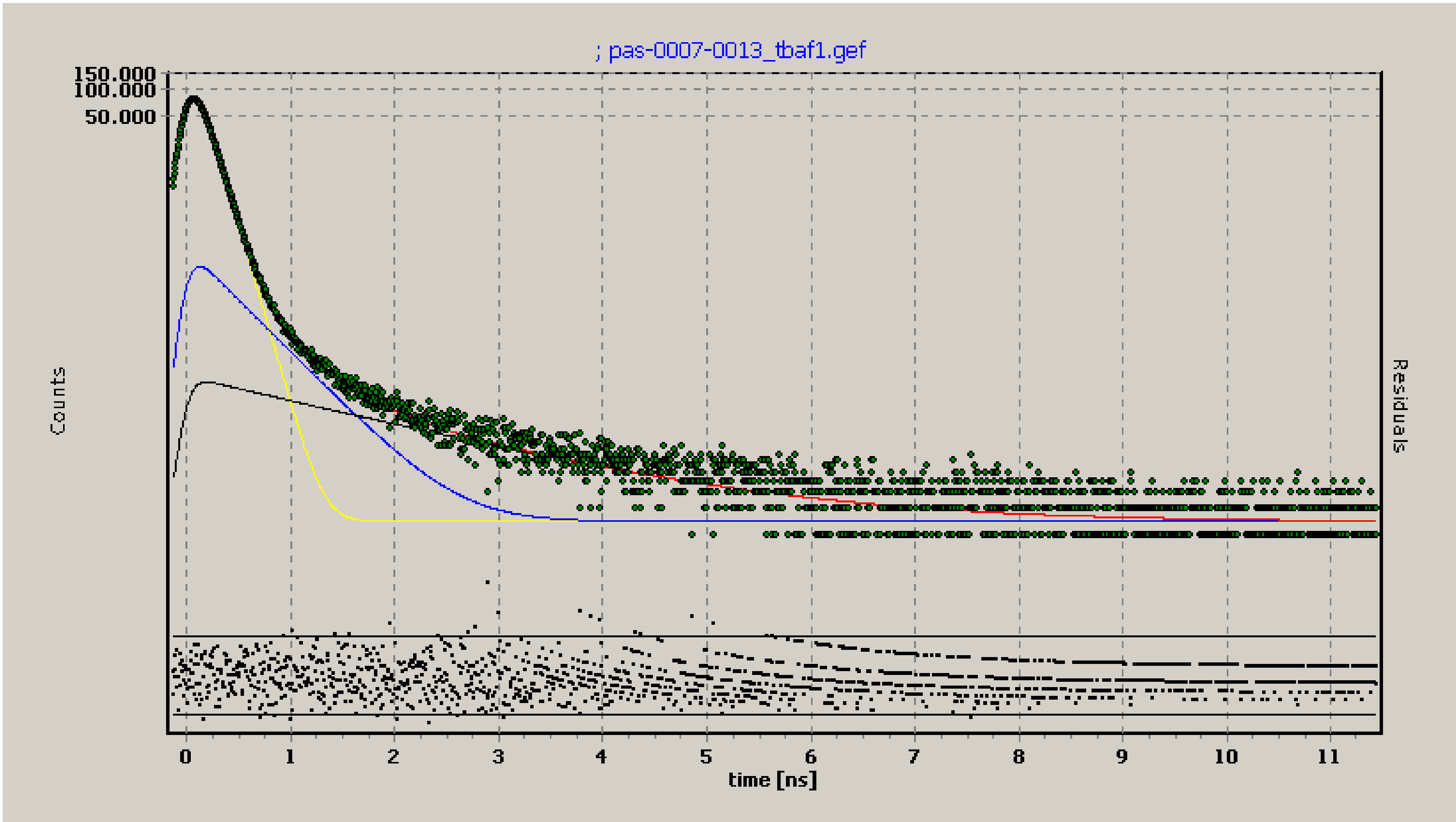
# Coincident lifetime spectrum: annealed Fe

- here coincidence with Ge detector
- spectrum is projection to the time scale of AMOC spectrum
- Count rate for AMOC spectrum = 320 /s
- One spectrum in 2h
- Time resolution = 210 ps
- BG/Peak = 1.7 x 10-5
- 350 ps & 1.5 ns: annihilation at vacuum tube (polyethylene)



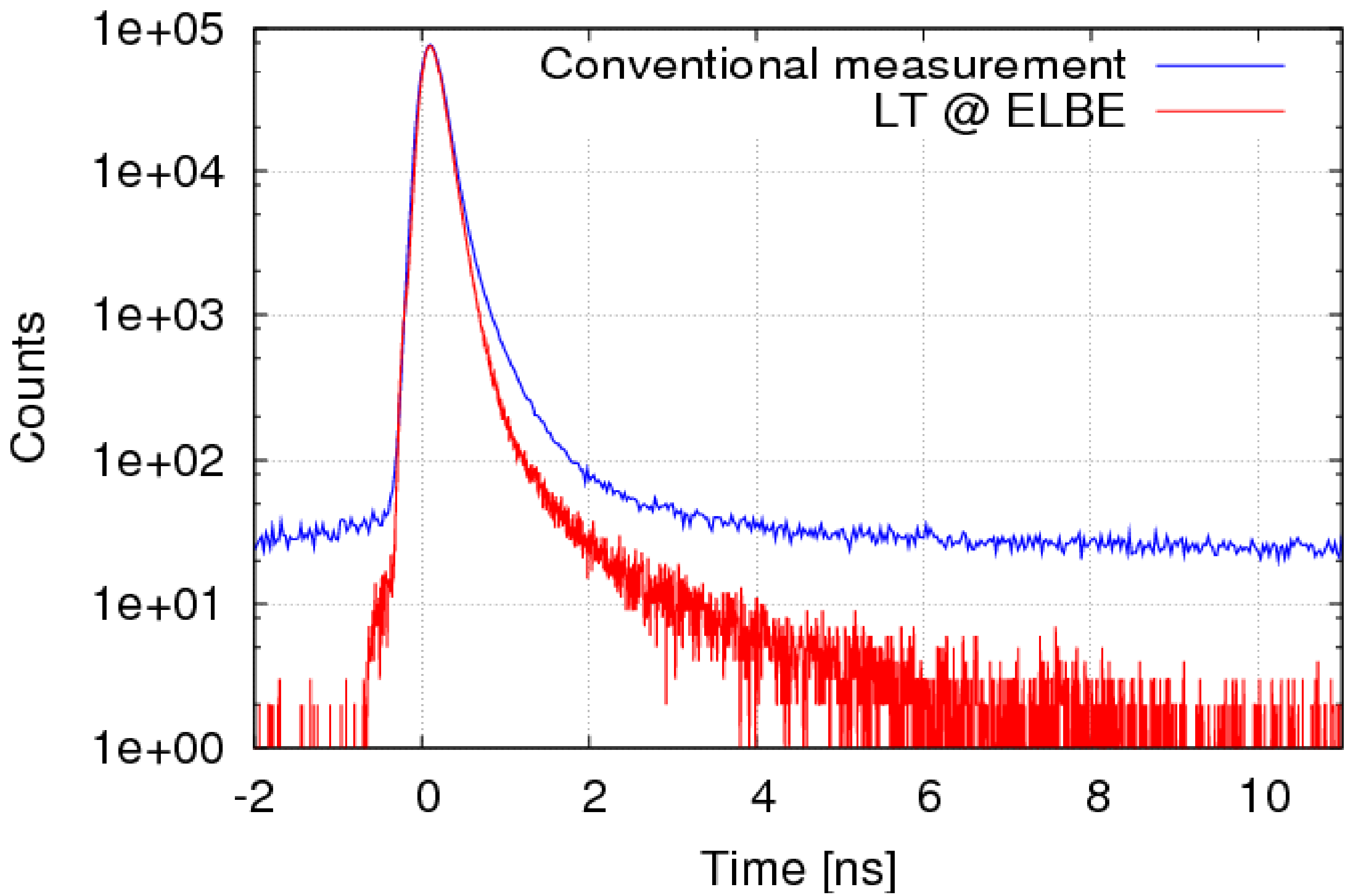
# Residuals of fit show perfect fit

- analysis by LT 9.0 (J. Kansy)



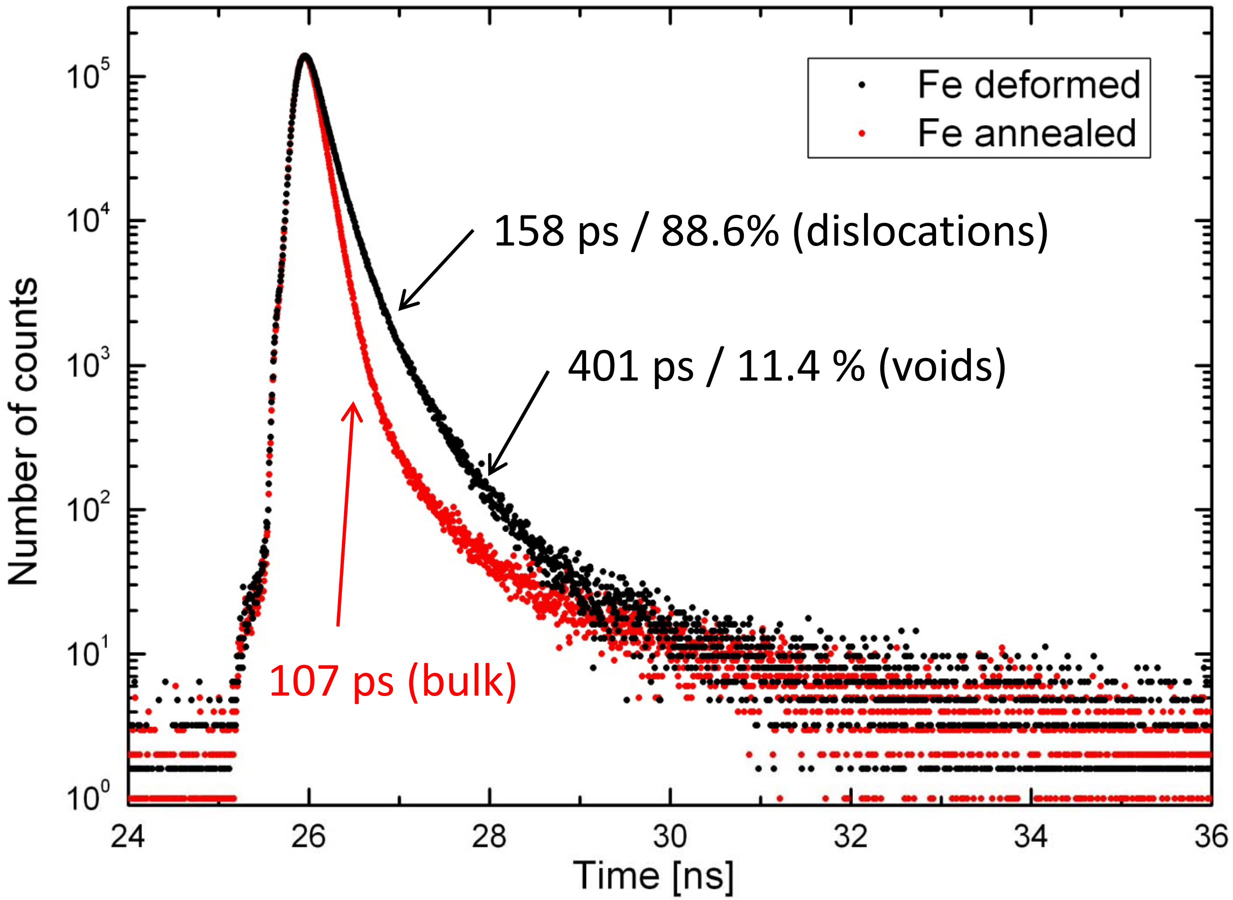
# Comparison: GiPS spectrum with conventional measurement

- same sample material – almost same statistics, similar time resolution
- conventional measurement with $^{22}Na$ source  20 µCi (0.7 MBq) in sandwich geometry
- advantage of periodic positron source is obvious: background distinctly reduced
- result of spectra analysis is the same: 107 ps (bulk value for Fe; corresponds to literature)



# Comparison annealed and deformed Fe

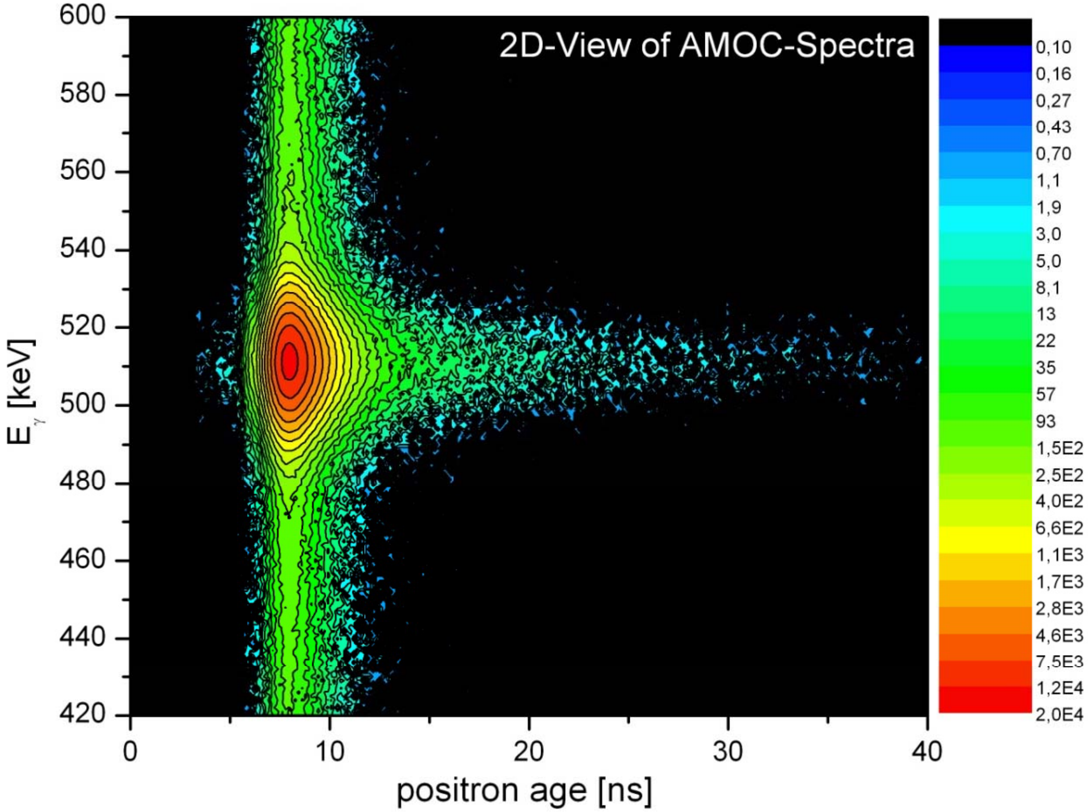
- two mechanically identical samples were prepared
- Fe annealed (1100°C; 2h in vacuum) and Fe (50% thickness reduction by cold rolling)
- spectra were easily decomposed
- expected results: annealed sample – one component 107 ps; deformed sample has 158 and 401 ps (dislocations and small vacancy clusters)



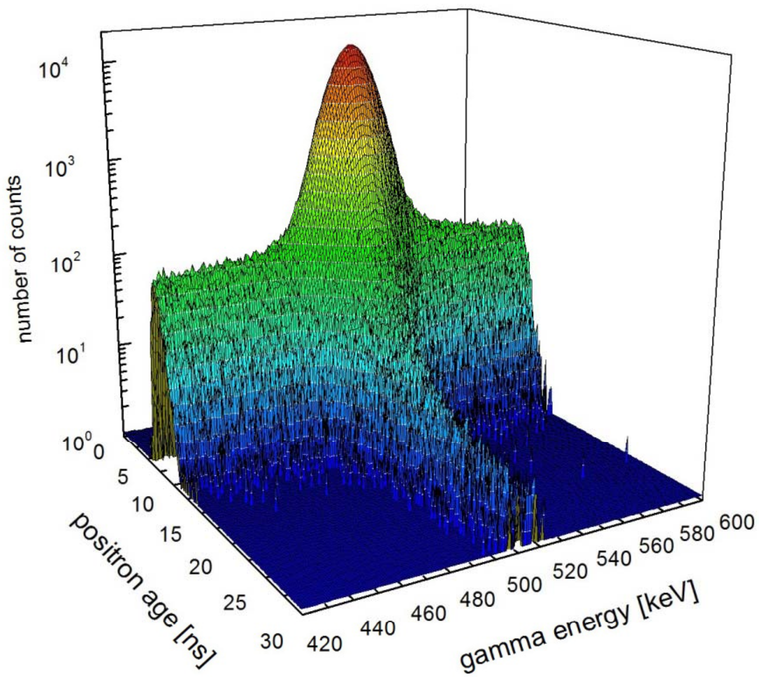
# AMOC spectrum of annealed Fe

- AMOC: measurement of momentum of annihilating electron as function of positron age
- AMOC detection is not an extra gimmick, but is required to maintain quality of spectra
- only by coincident measurement of 511 keV annihilation line: suppression of scattered gamma (can be concluded from lifetime spectra)

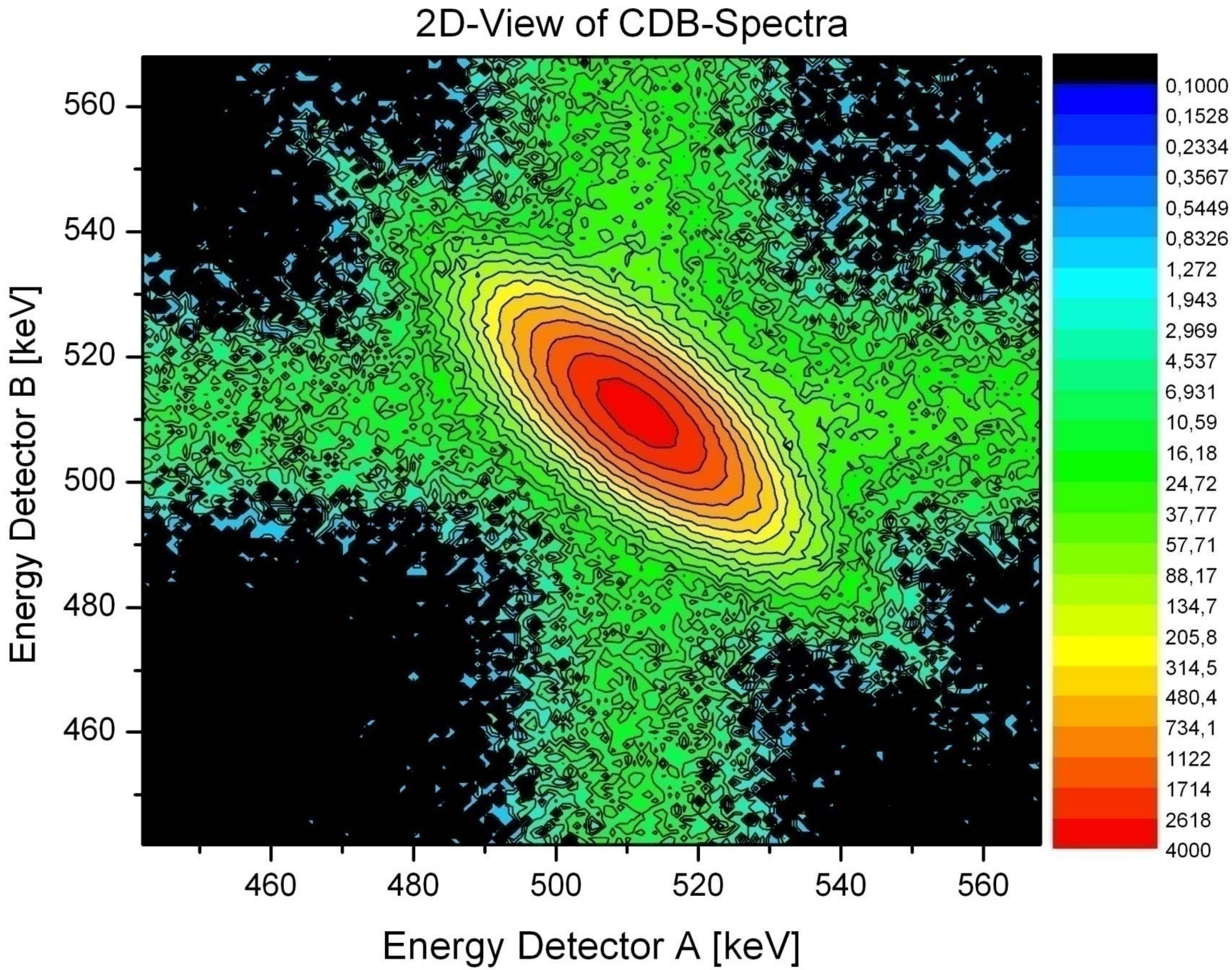
2D-Plot




3D-Plot

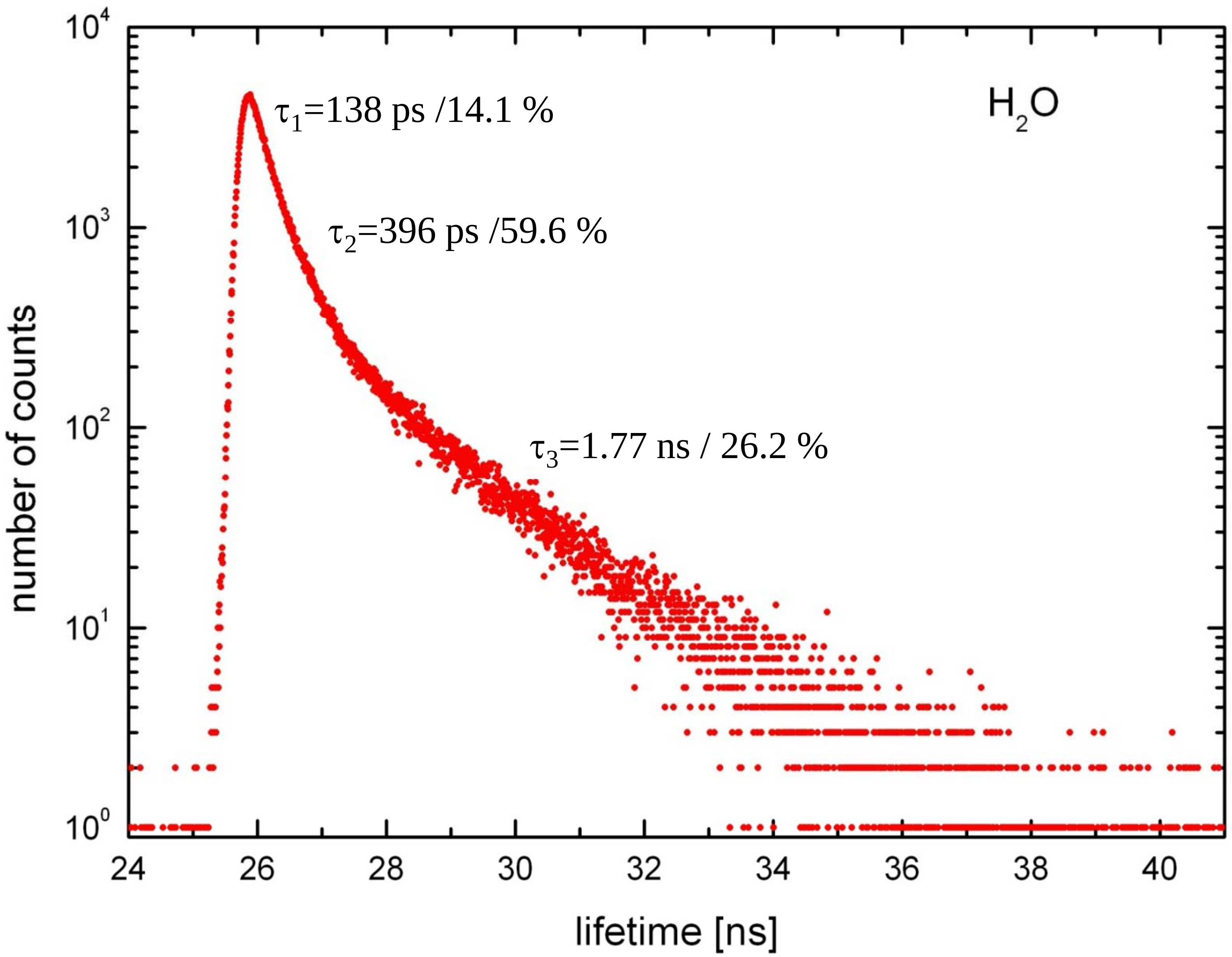


# Coincidence Doppler-Broadening Spectroscopy of Fe sample



# Water at RT

- total count rate: $5x10^5$
- no such visible deviations on $t < t_0$ like for Fe (due to much smaller gamma scattering compared to Fe)

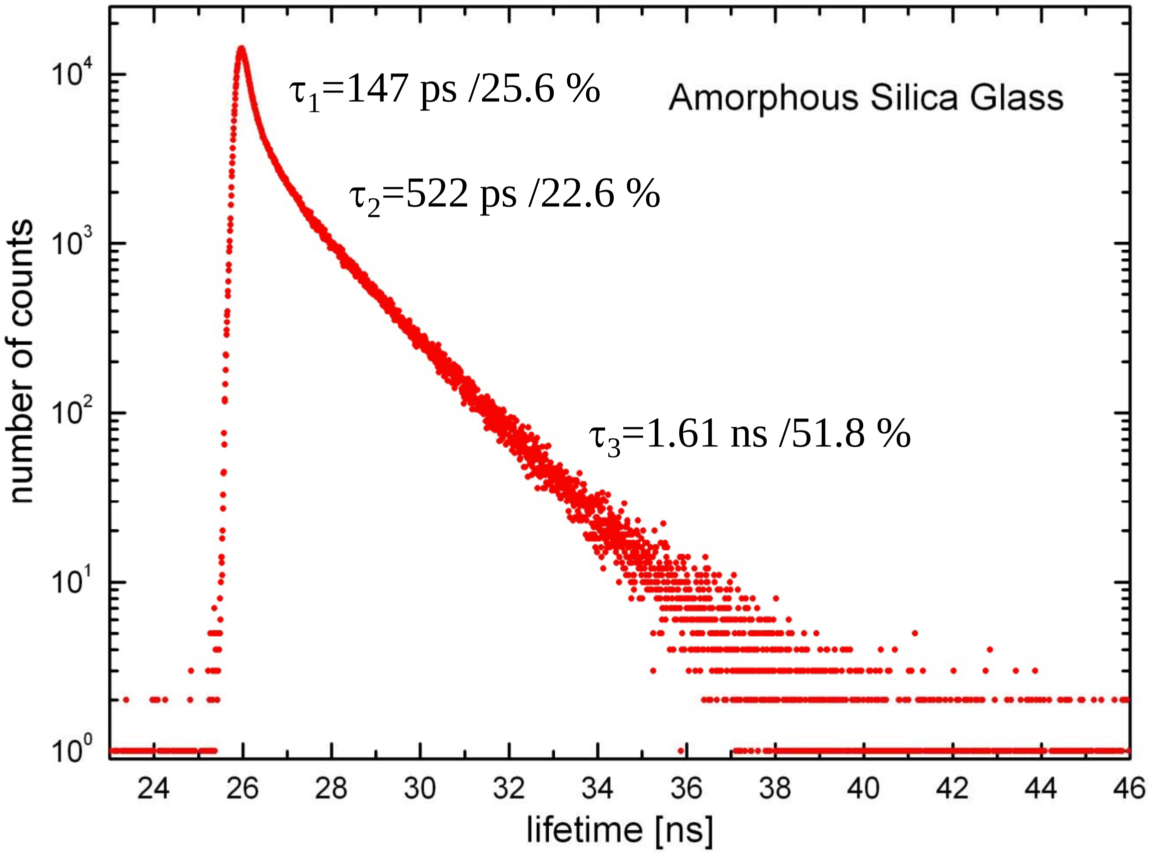


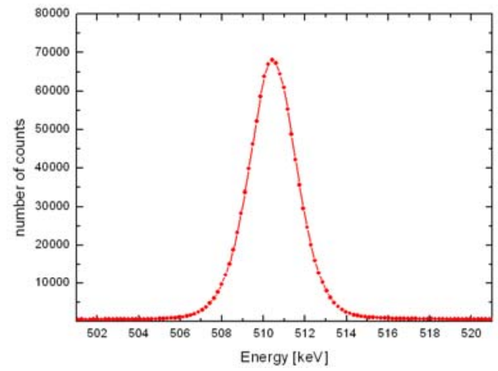
# Amorphous Silica Glass

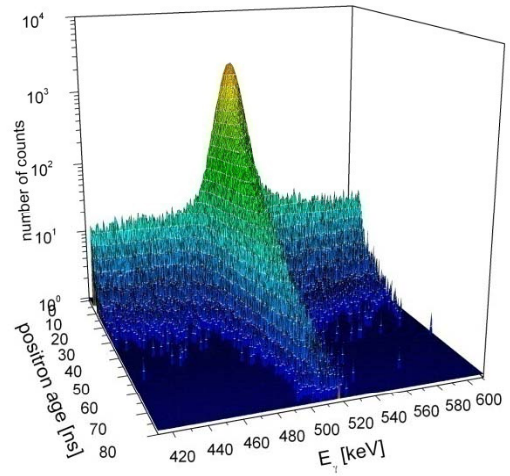
- round piece 1.5 cm thick, about 5 $cm^3$
- lifetime spectrum: total count rate: $2x10^6$
- same sample was measured conventionally in 1978 also in the same institute (former ZfK Rossendorf):

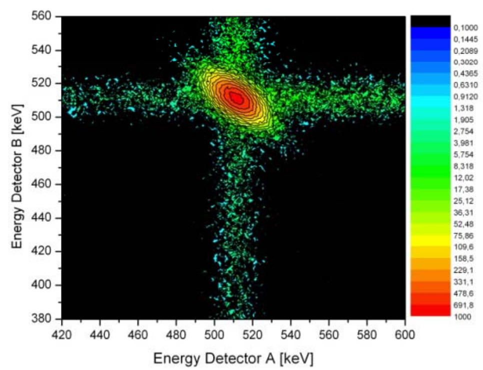
  151 ps - 523 ps - 1.57 ns (FWHM ≈350 ps)

  G. Brauer et al., Appl. Phys. 16 (1978) 231



# Conclusions

- new concept of EPOS project is now extended to use mono-energetic Positrons (MePS), Gamma-induced (GiPS) and conventional spectroscopy (CoPS)
- all spectrometers are equipped with LT, CDB, AMOC
- fully digital system (in the future)
- EPOS can cover sample thickness range from 10 nm to 10 cm (7 orders of magnitude)
- MePS still under construction
- GiPS has been tested successfully
  - GiPS only possible because of the unique properties of the ELBE Linac (cw mode of 26 MHz intense and extremely short electron bunches, < 5ps bunch length)
  - background suppression by coincident measurement of Lifetime and Doppler (AMOC)
  - surprisingly good spectra quality
  - coincidence between 2 $BaF_2$: resolution improves by 24% (FWHM = 160 ps)
  - problem: heating / cooling of sample because in holder positrons are also generated

Talk available at http://positron.physik.uni-halle.de

Research Center Dresden-Rossendorf, 28.-30. September 2009

# Workshop on Digital Signal Processing in Nuclear Science

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

**Open-source Project**

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