Positron Annihilation in Metal Physics

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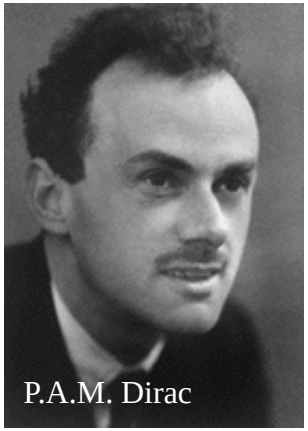
- History
- Means of Positron Annihilation
- Applications in Metal Physics

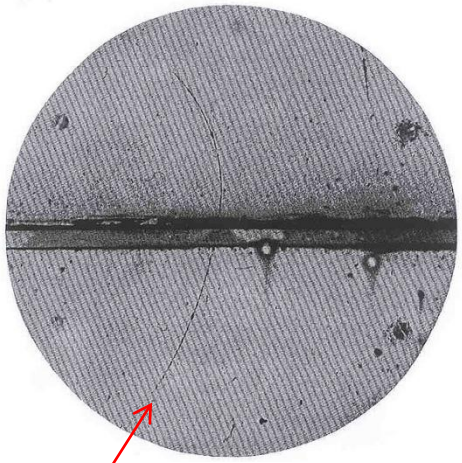
Discovery of the Positron


P.A.M. Dirac

- Positron was predicted in 1928 by Paul A.M. Dirac
- Discovery in 1932 in cloud chamber pictures by C.D. Anderson


C.D. Anderson

- Positronium as bound state of e^- and e^+ lightest atom was predicted (1934) and discovered (1951)
- Annihilation in matter was studied beginning in the 40s
- Positrons can be obtained by
  - pair production from gamma radiation (E_γ > 1022 keV)
  - β^+ decay from isotopes (mostly ^{22}Na)



- first Identification of a positron in a cloud chamber
- 5 mm lead plate
- photo taken by C.D. Anderson

Electron structure of solids can be discovered

- during annihilation: conservation laws must be fulfilled (energy, momentum)
- positron cools down to thermal energies ->
- energy of annihilating electron-positron pair = energy of electron
- electron momentum distribution can directly be measured

MARCH 1 AND 15, 1942 PHYSICAL REVIEW VOLUME 61

The Angular Distribution of Positron Annihilation Radiation

ROBERT BERINGER* AND C. G. MONTGOMERY
Sloane Physics Laboratory, Yale University, New Haven, Connecticut

(Received January 7, 1942)

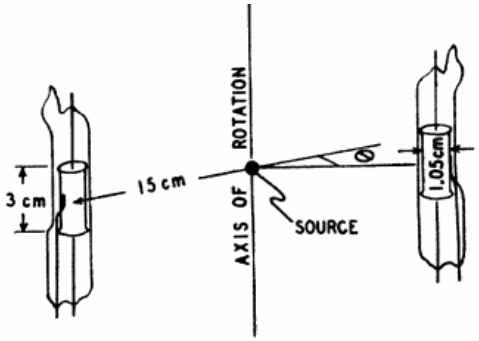
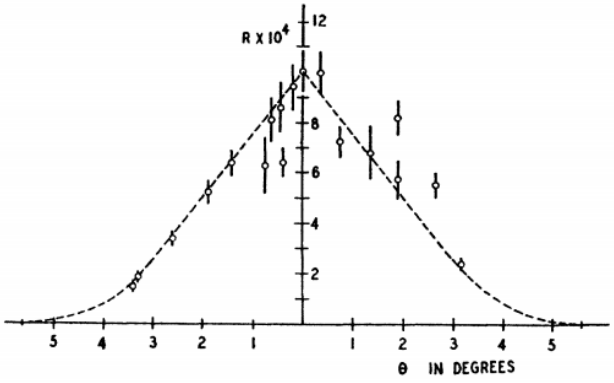
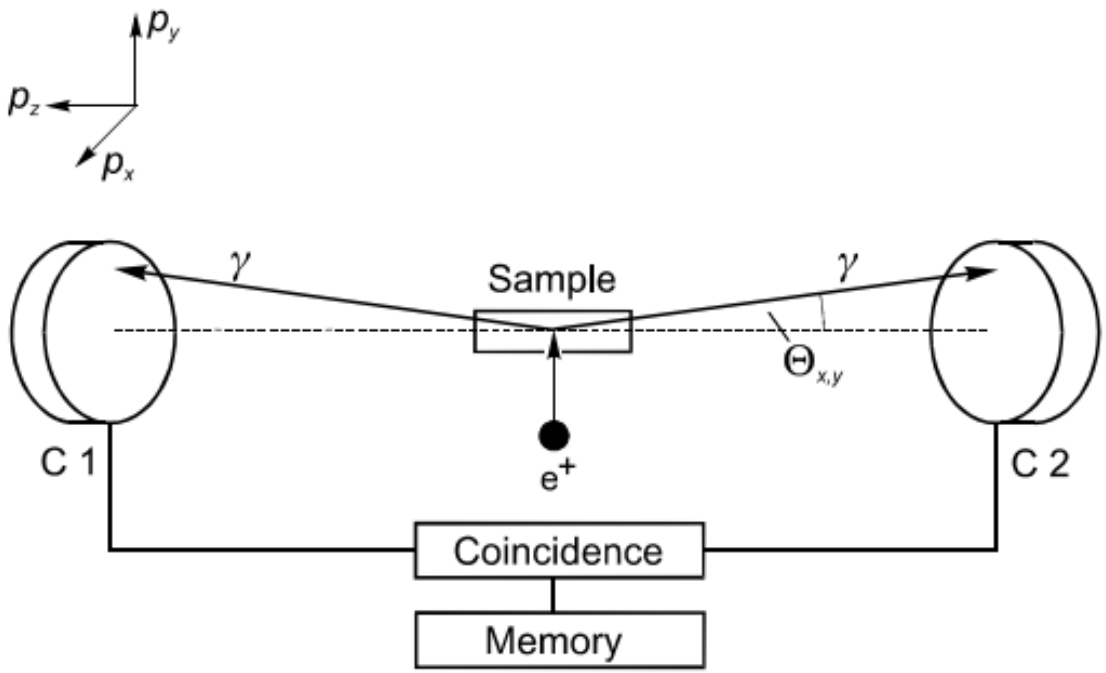


FIG. 1. Schematic arrangement of counters for observing coincidences from annihilation radiation.



2D – ACAR (Angular Correlation of Annihilation Radiation)

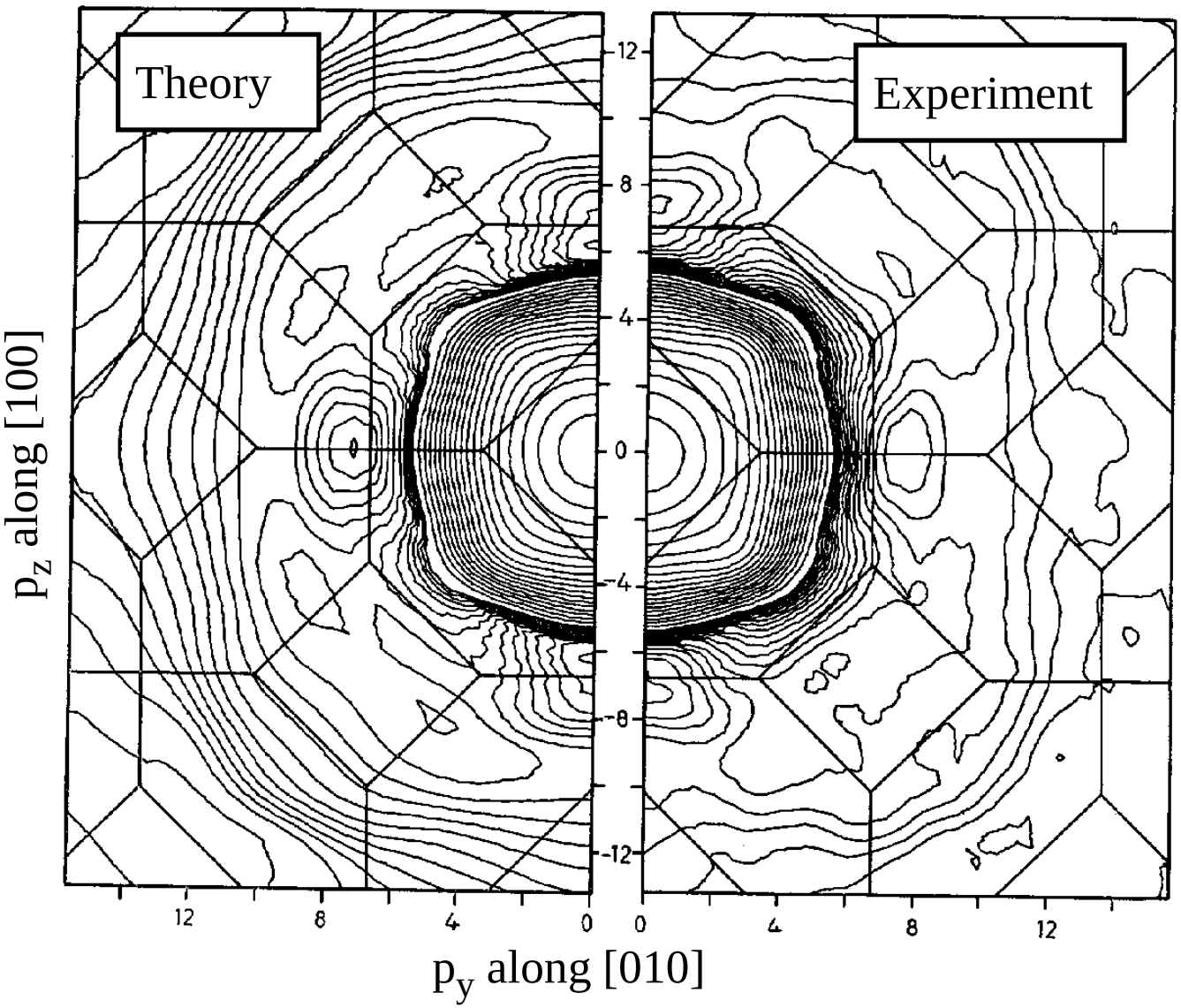
- now: two-dimensional (position-sensitive) detectors
- measurement of single crystals in different directions:
- reconstruction of Fermi surface possible

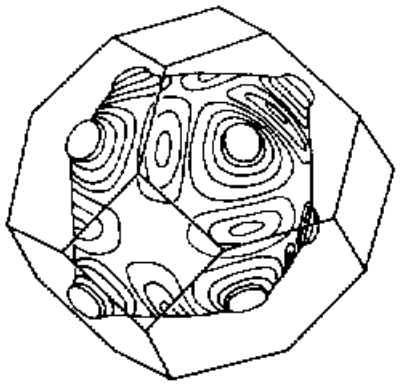


- sophisticated device available at Univ. Delft
- intense positron source at reactor
- slow (moderated) positrons allow study of near-surface layers

$$N_c(\Theta_x, \Theta_y) = A_c \int_{-\infty}^{\infty} \sigma(\Theta_x m_0 c, \Theta_y m_0 c, p_z) \mathrm{d}p_z$$

2D-ACAR of Copper





Fermi surface of copper

(Berko, 1979)

Positrons are sensitive for Crystal Lattice Defects

- 1950…1960: in addition to ACAR -> different experimental techniques were developed
- Positron lifetime spectroscopy and Doppler broadening spectroscopy
- end of 60s: lifetime is sensitive to lattice imperfections
  - Brandt et al. (1968): **vacancies in ionic crystals**
  - Dekhtyar et al. (1969): **plastically deformed semiconductors**
  - MacKenzie et al. (1967): **vacancies in thermal equilibrium** in metals
- Positrons are localized (trapped) by open-volume defects

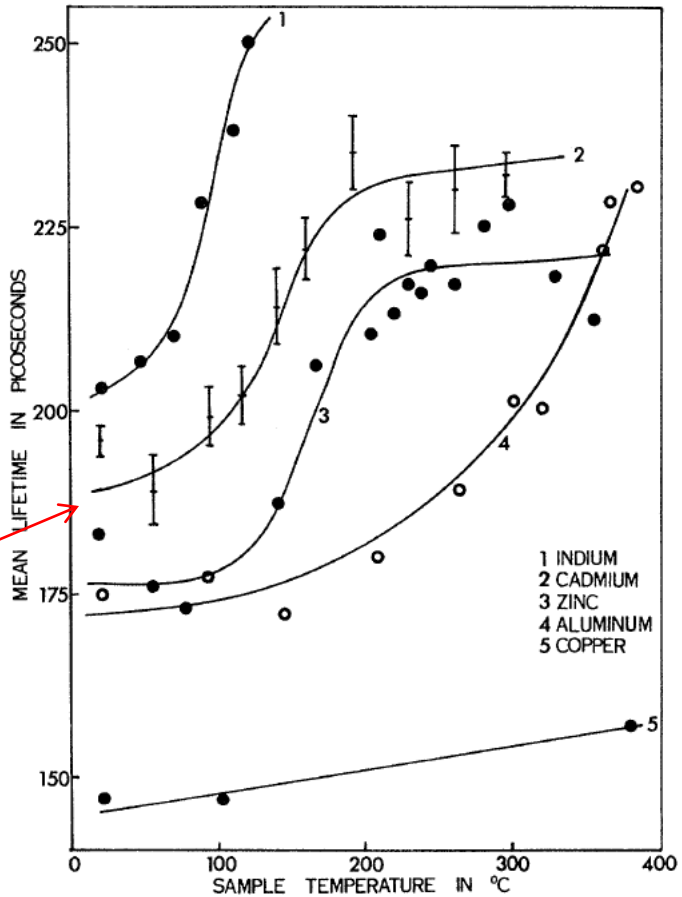
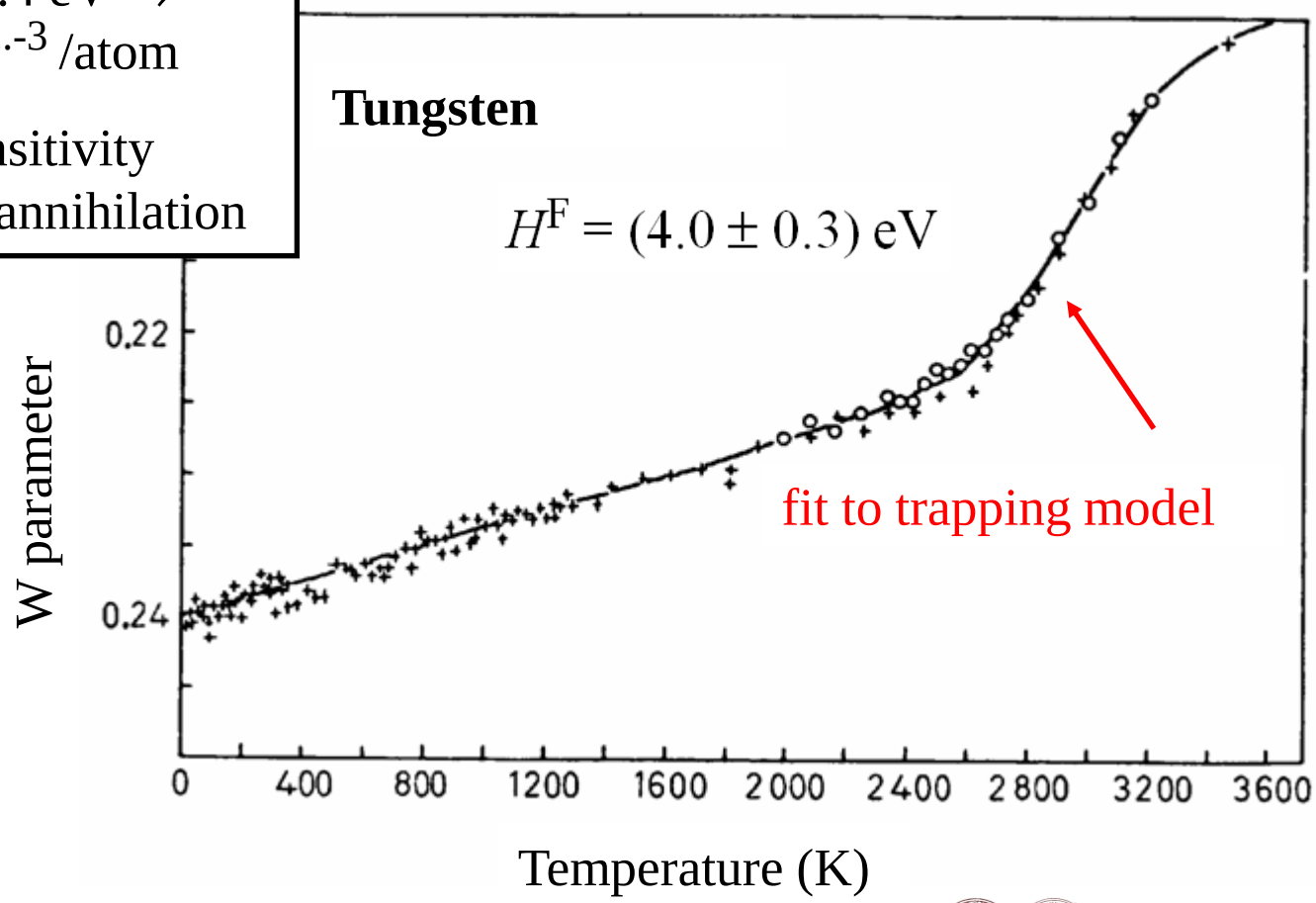


FIG. 1. Positron mean lifetimes in several metals as a function of temperature.

Vacancies in thermal Equilibrium

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

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



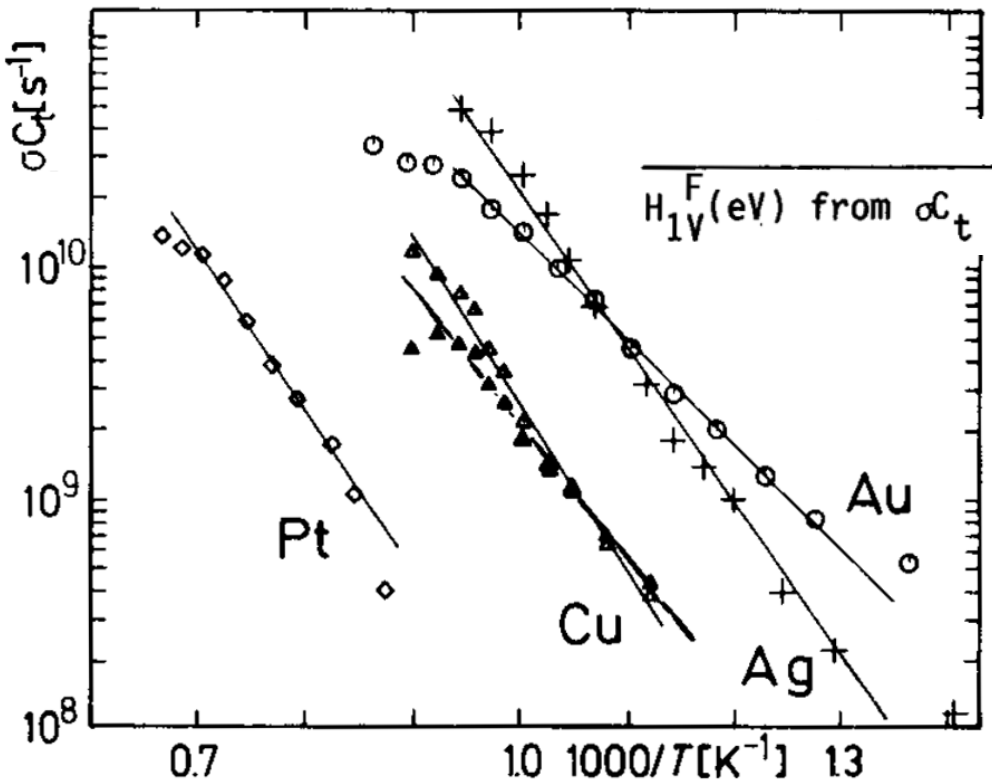
(Ziegler, 1979)

Determination of Vacancy Formation Enthalphy

THERMAL VACANCIES IN THE NOBLE METALS Cu, Ag, Au, AND IN Pt STUDIED BY POSITRON LIFETIME SPECTROSCOPY

H. E. Schaefer[1], W. Stuck[1], F. Banhart[2], and W. Bauer

[1]Universität Stuttgart, Institut für Theoretische und Angewandte Physik, Pfaffenwaldring 57, D-7000 Stuttgart 80,
[2]Max-Planck-Institut für Metallforschung, Institut für Physik, Heisenbergstr. 1, D-7000 Stuttgart 80, Fed. Rep. of Germany



	Cu	Ag	Au	Pt
H_{1V}^{F}(eV) from σC_t	1.13±.04*	1.31±.07	.89±.02	1.35±.09

- Arrhenius-Plot delivers H_{1V}
- was performed for many alloys

Study of non-equilibrium Defects

PHYSICAL REVIEW B VOLUME 25, NUMBER 2 15 JANUARY 1982

Vacancies and carbon impurities in α-iron: Electron irradiation

A. Vehanen, P. Hautojärvi, J. Johansson, and J. Yli-Kauppila
Laboratory of Physics, Helsinki University of Technology, SF-02150 Espoo 15, Finland
P. Moser
Section de Physique du Solide, Département de Recherche Fondamentale, Centre d'Etudes Nucléaires de Grenoble, 85 X, 38041 Grenoble Cédex, France

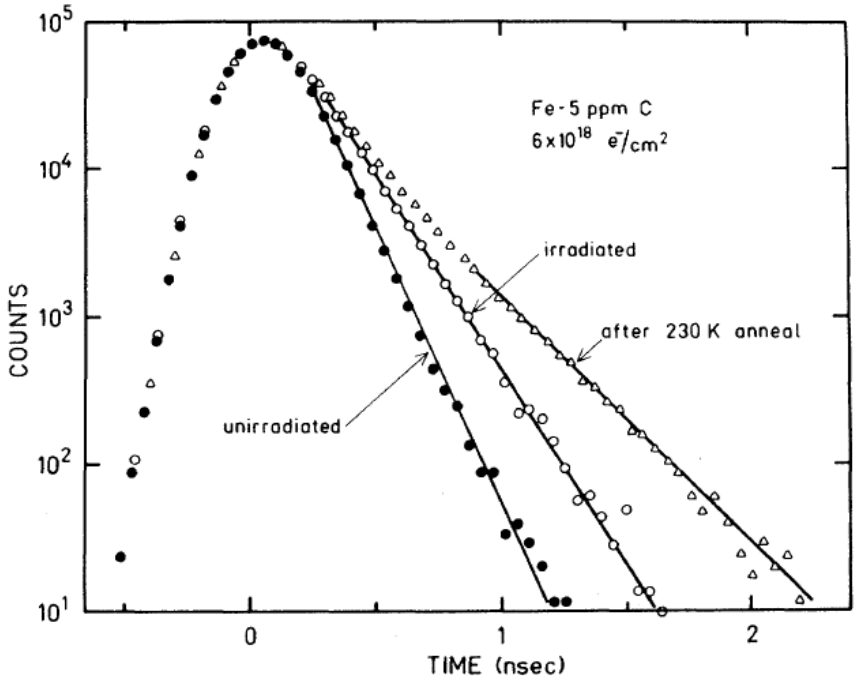
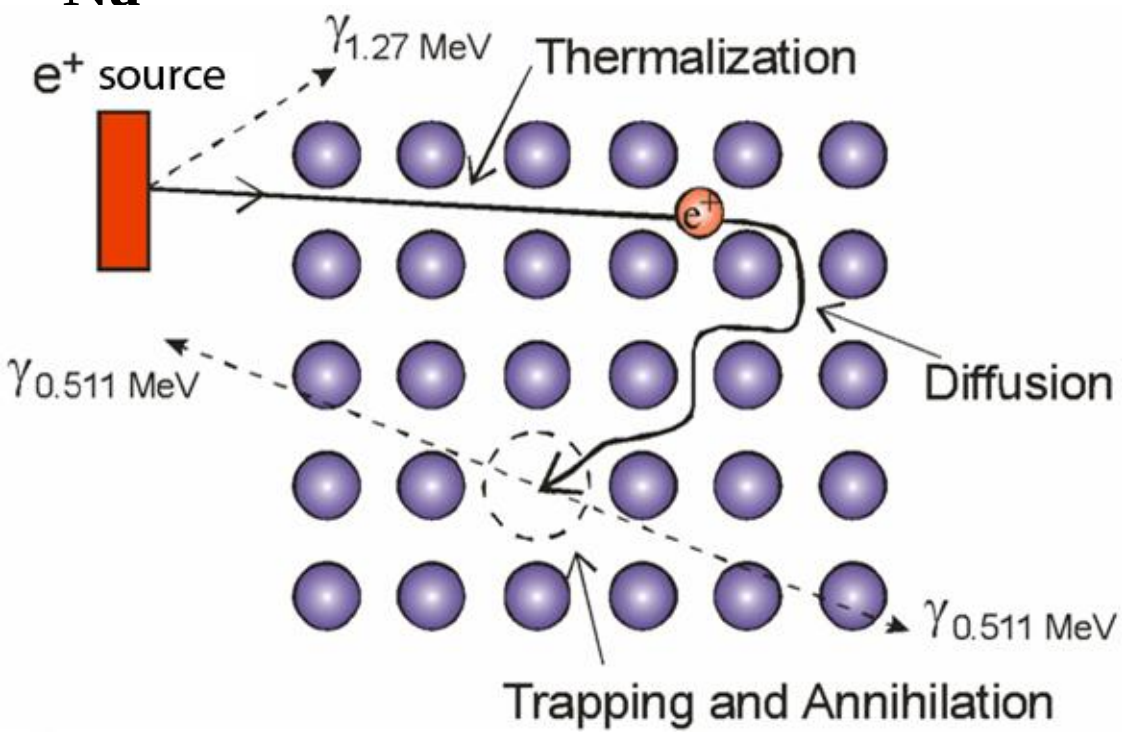


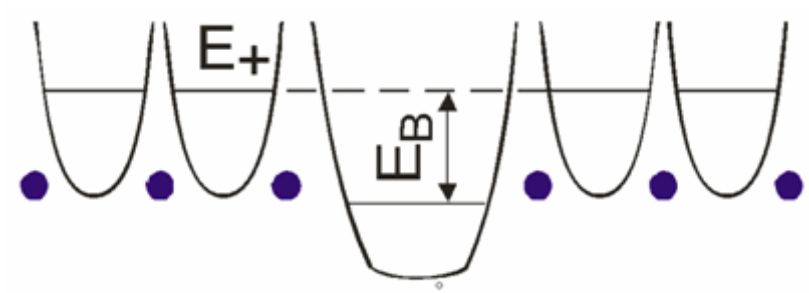
FIG. 1. Positron-lifetime spectra after source-background subtraction in electron-irradiated (6×10^{18} e^-/cm^2) high-purity iron at various stages of isochronal annealing. The dramatic occurrence of a long-lifetime component after 230 K annealing is clearly visible.

- positron lifetime is very sensitive for vacancy-type defects
- here: lifetime increases after irradiation
- and further increase after first annealing: vacancy clustering

The positron lifetime spectroscopy

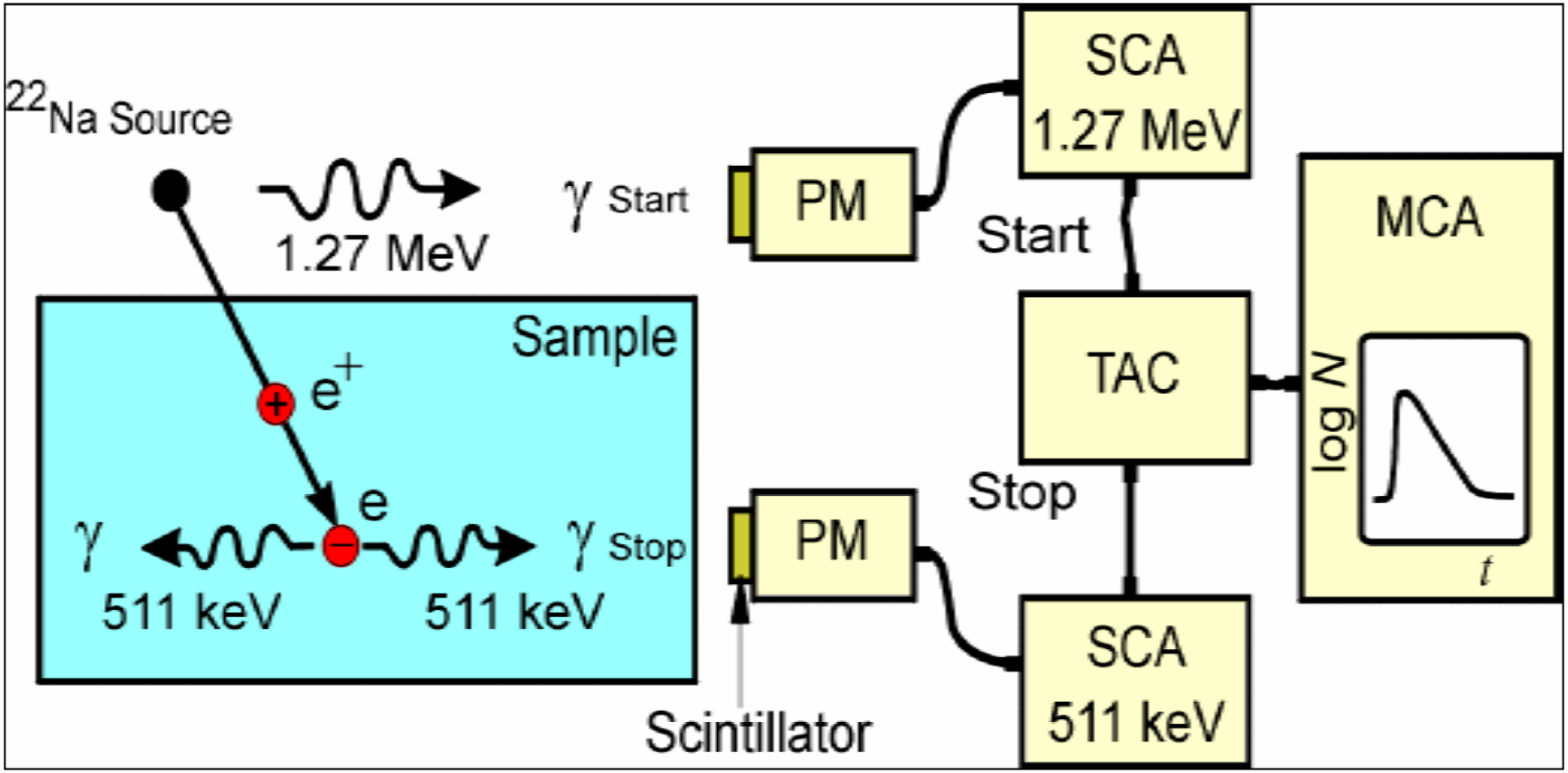
^{22}Na





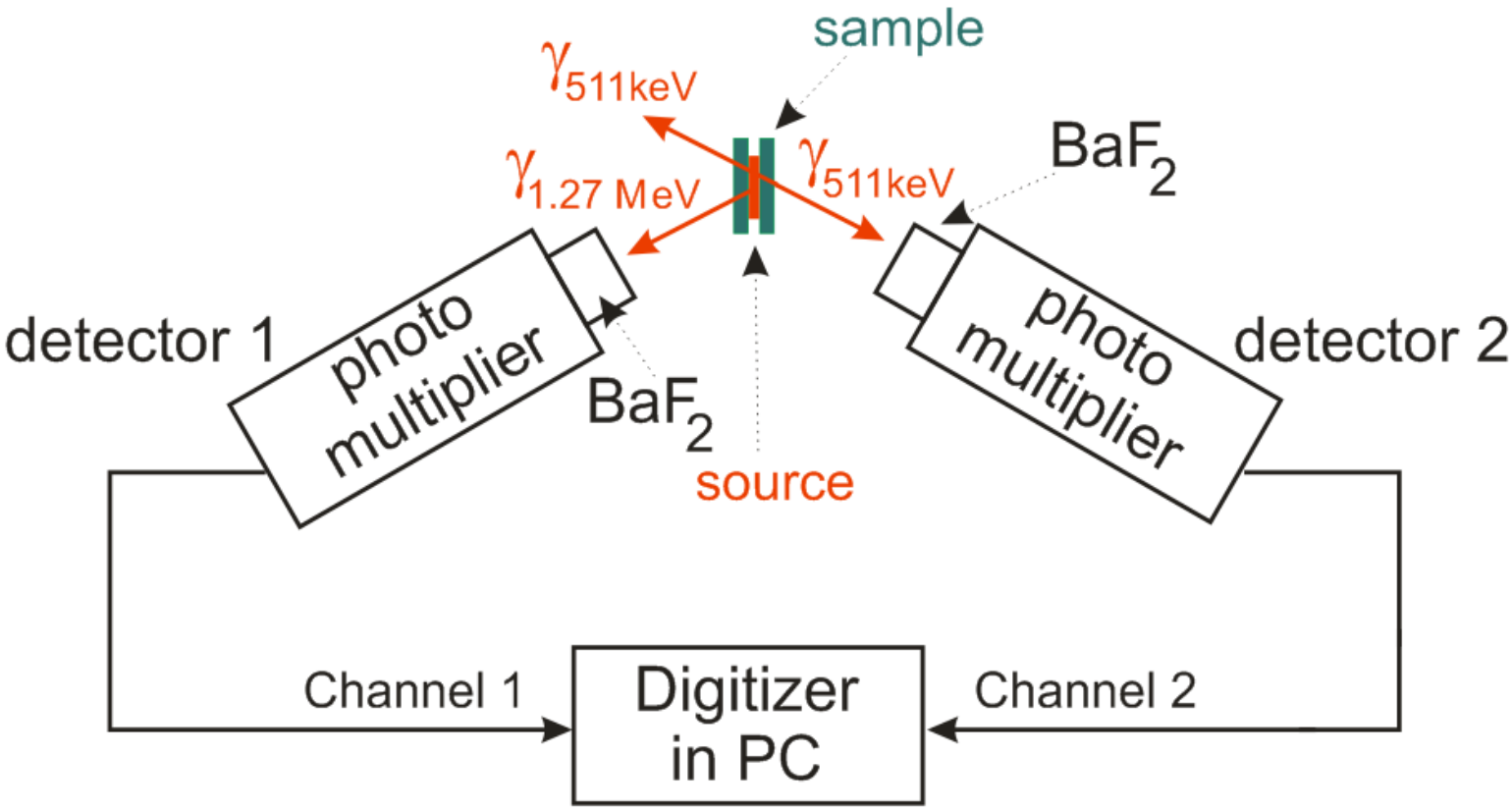
- positron wave-function can be localized in the attractive potential of a defect
- annihilation parameters change in the localized state
- e.g. positron lifetime increases in a vacancy
- lifetime is measured as time difference between appearance of 1.27 (start) and 0.51 MeV (stop) quanta
- defect identification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

Positron lifetime spectroscopy



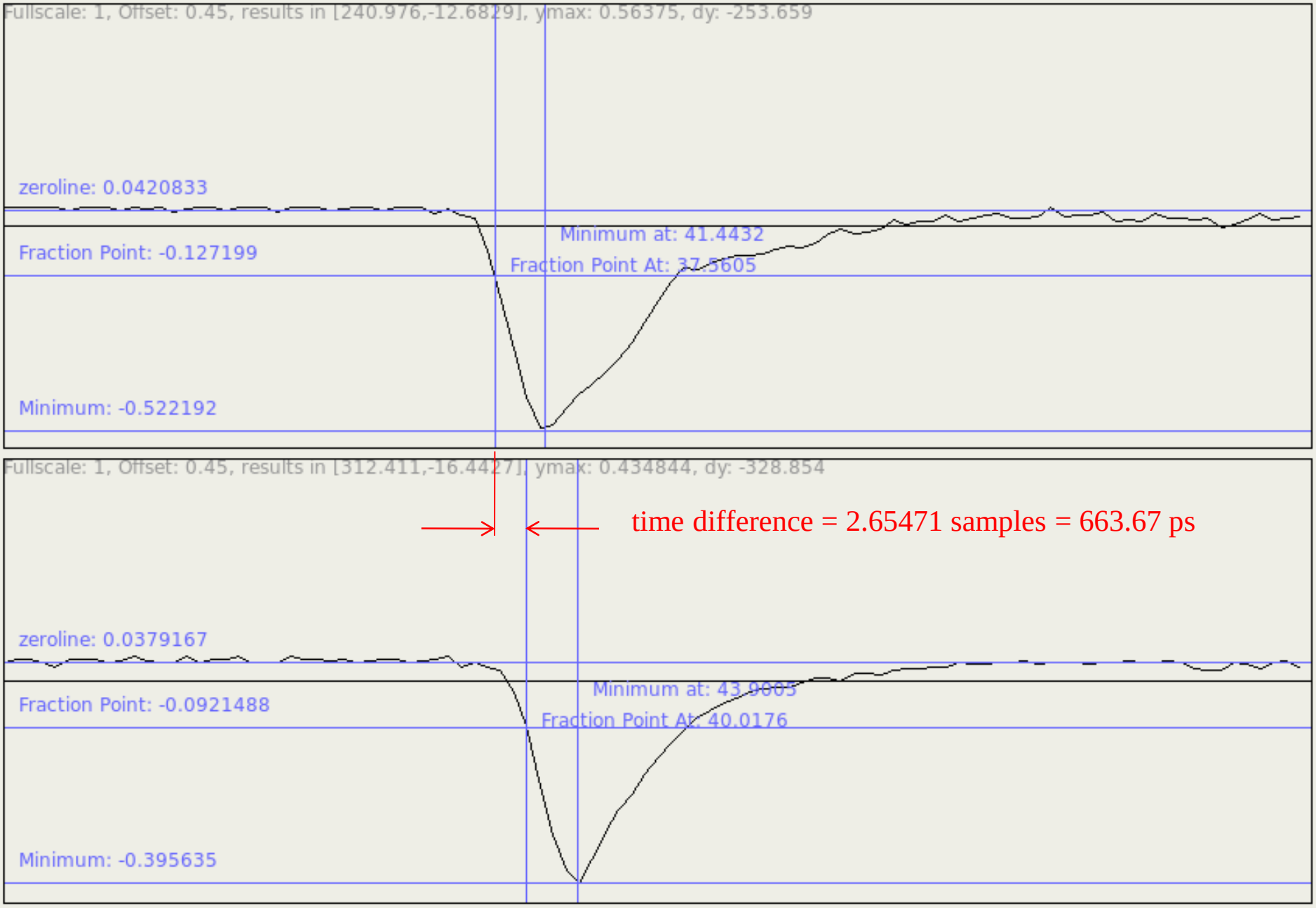
Positron lifetime: time between 1.27 MeV and 0.511 MeV quanta

Digital lifetime measurement

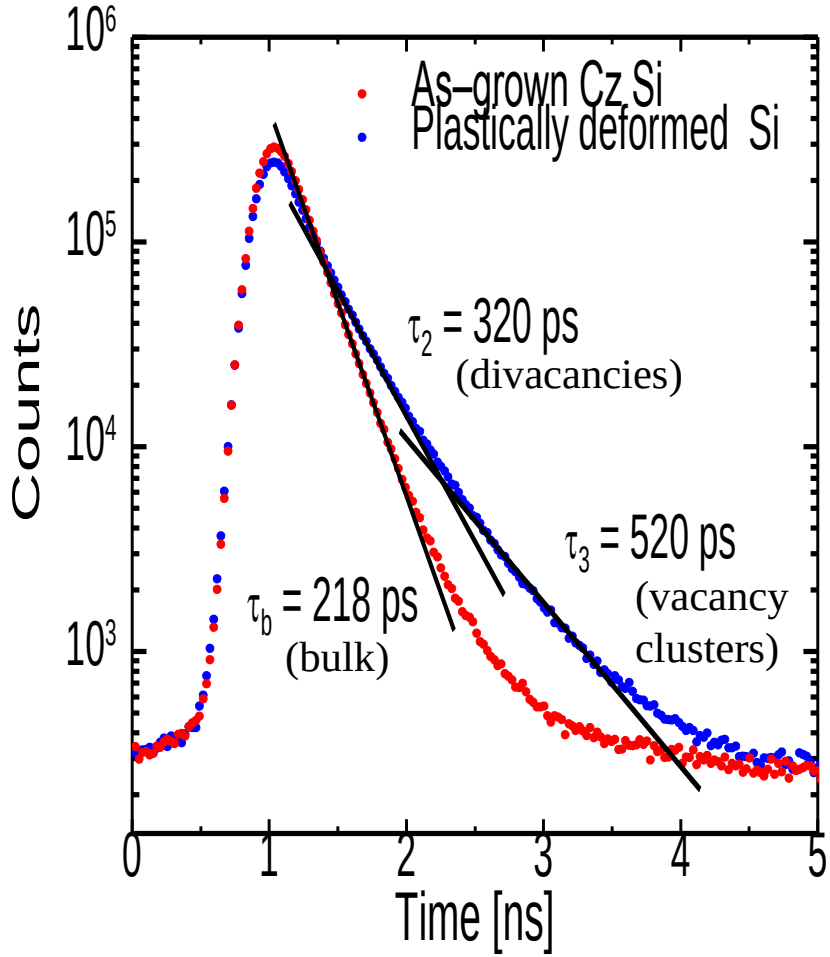


- much simpler setup
- timing very accurate
- pulse-shape discrimination (suppress "bad pulses")
- each detector for start & stop (double statistics)

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 τ_i and intensities I_i

positron lifetime spectrum:

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

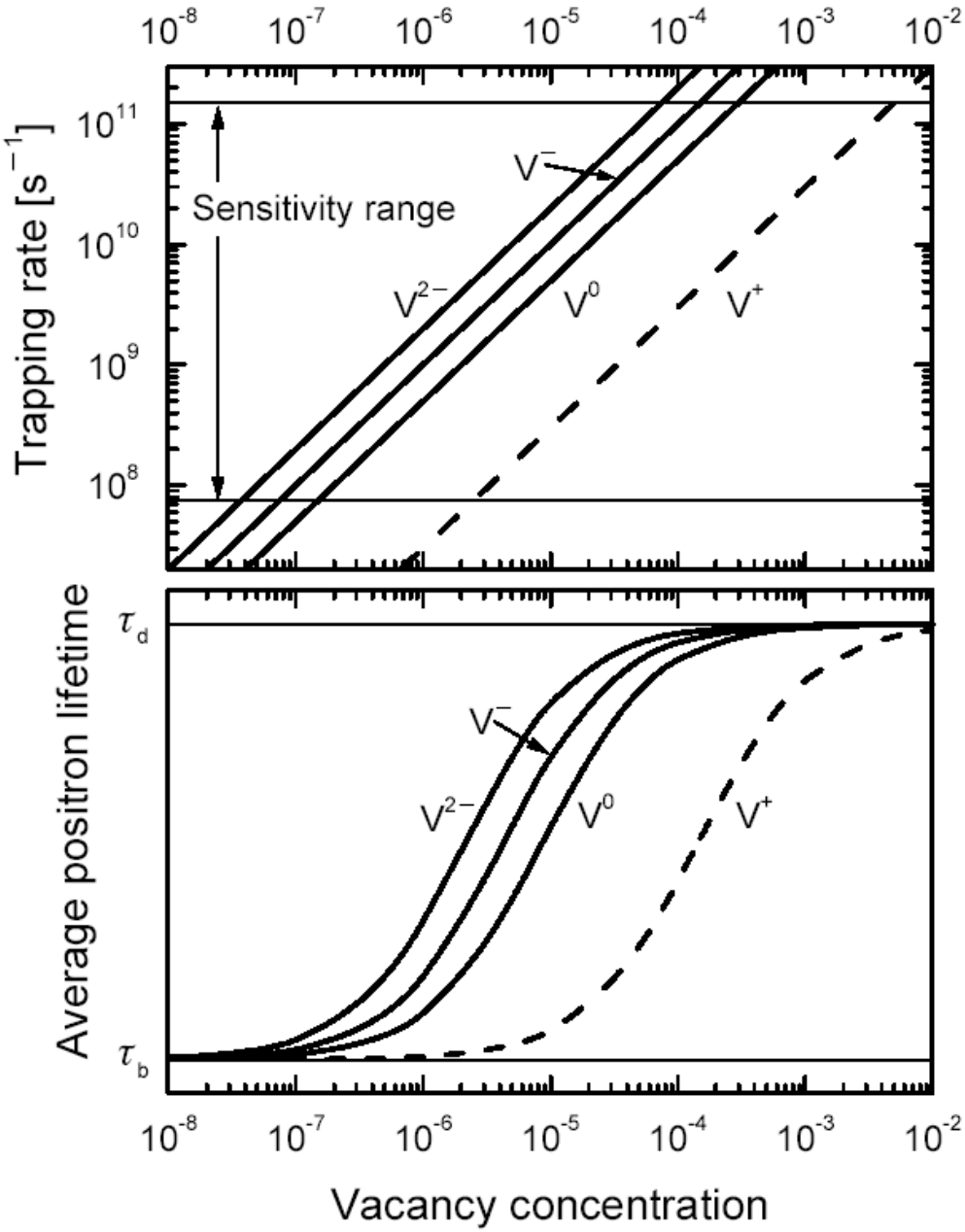
trapping coefficient

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

trapping rate

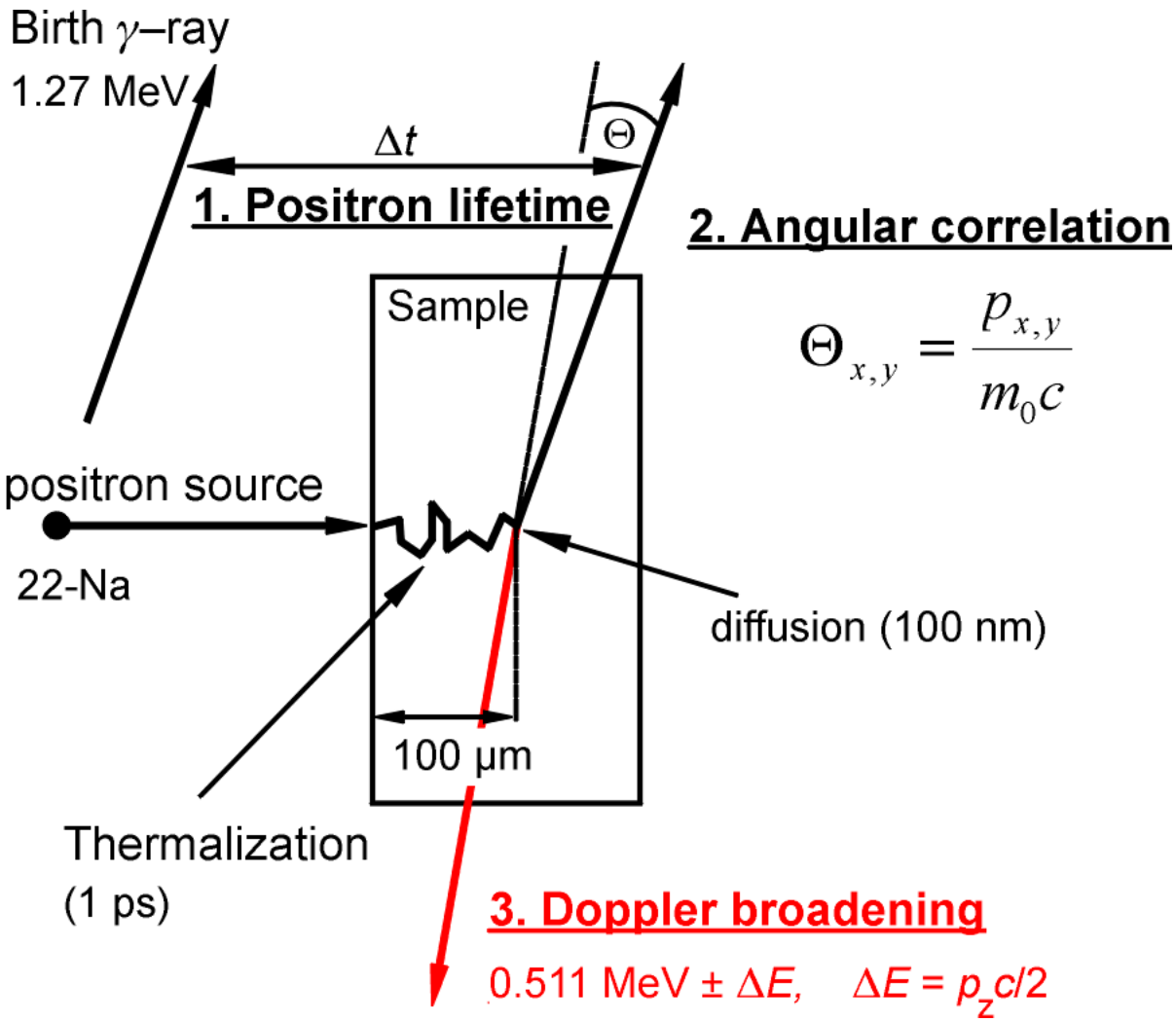
defect concentration

Sensitivity limits of PAS for vacancy detection



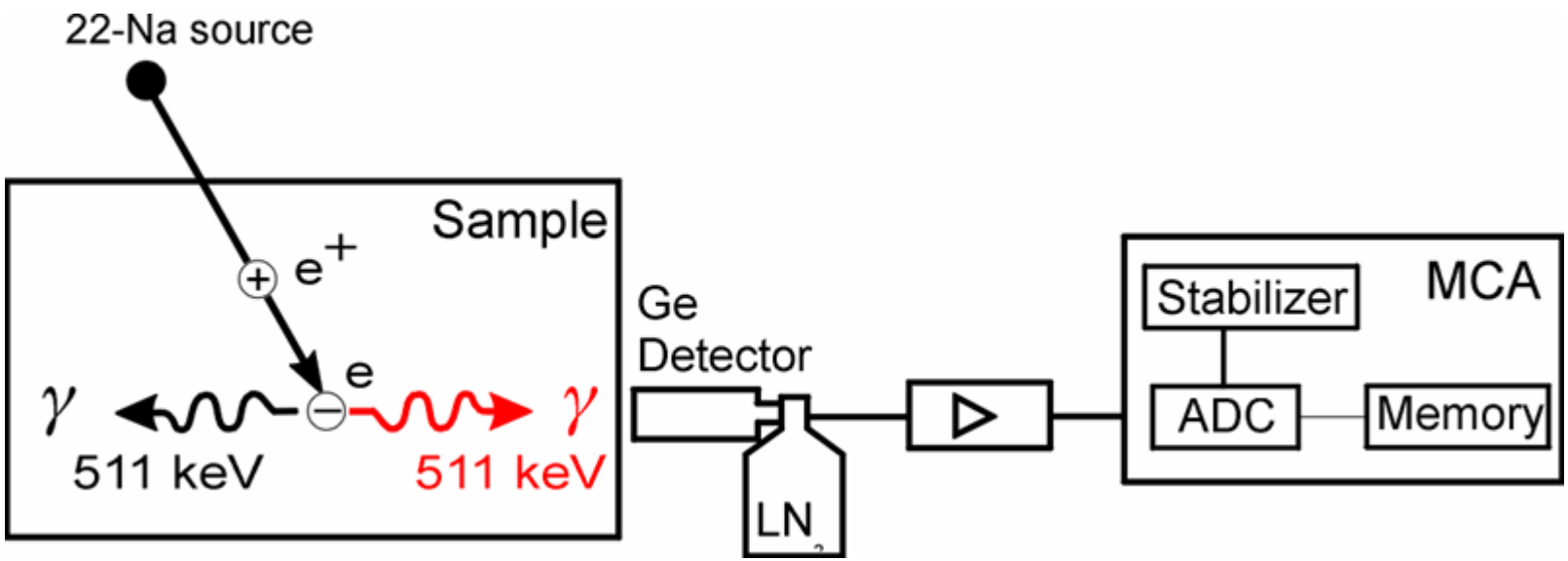
- **lower sensitivity limit** e.g. for negatively charged divacancies in Si starts at about 10^{15} cm^{-3}
- **upper limit**: saturated positron trapping
- defect identification still possible
- Then: only lower limit for defect density can be given

Doppler Broadening Spectroscopy



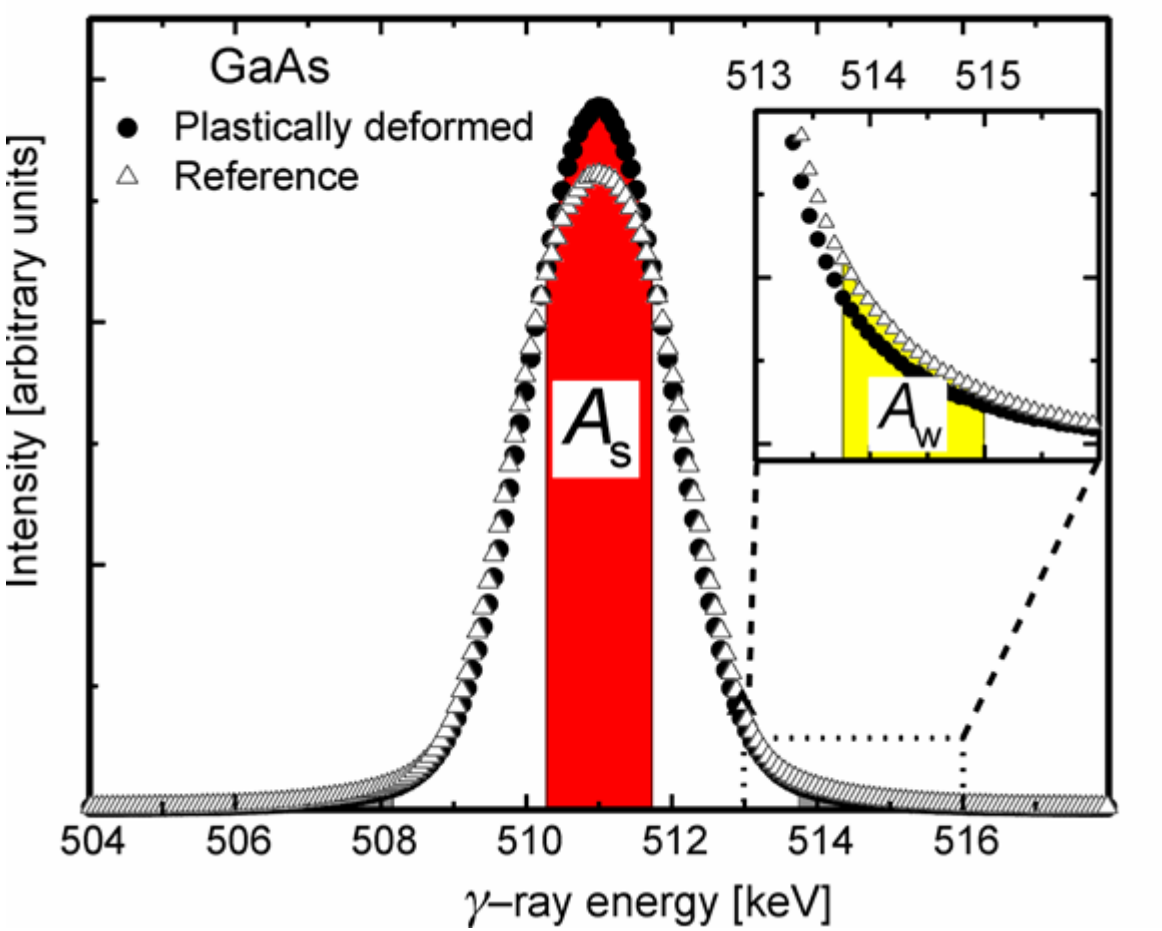
$$\Theta_{x,y} = \frac{p_{x,y}}{m_0 c}$$

Measurement of Doppler Broadening



- electron momentum in propagation direction of 511 keV γ-ray leads to Doppler broadening of annihilation line
- can be detected by conventional energy-dispersive Ge detectors and standard electronics

Line Shape Parameters



S parameter:

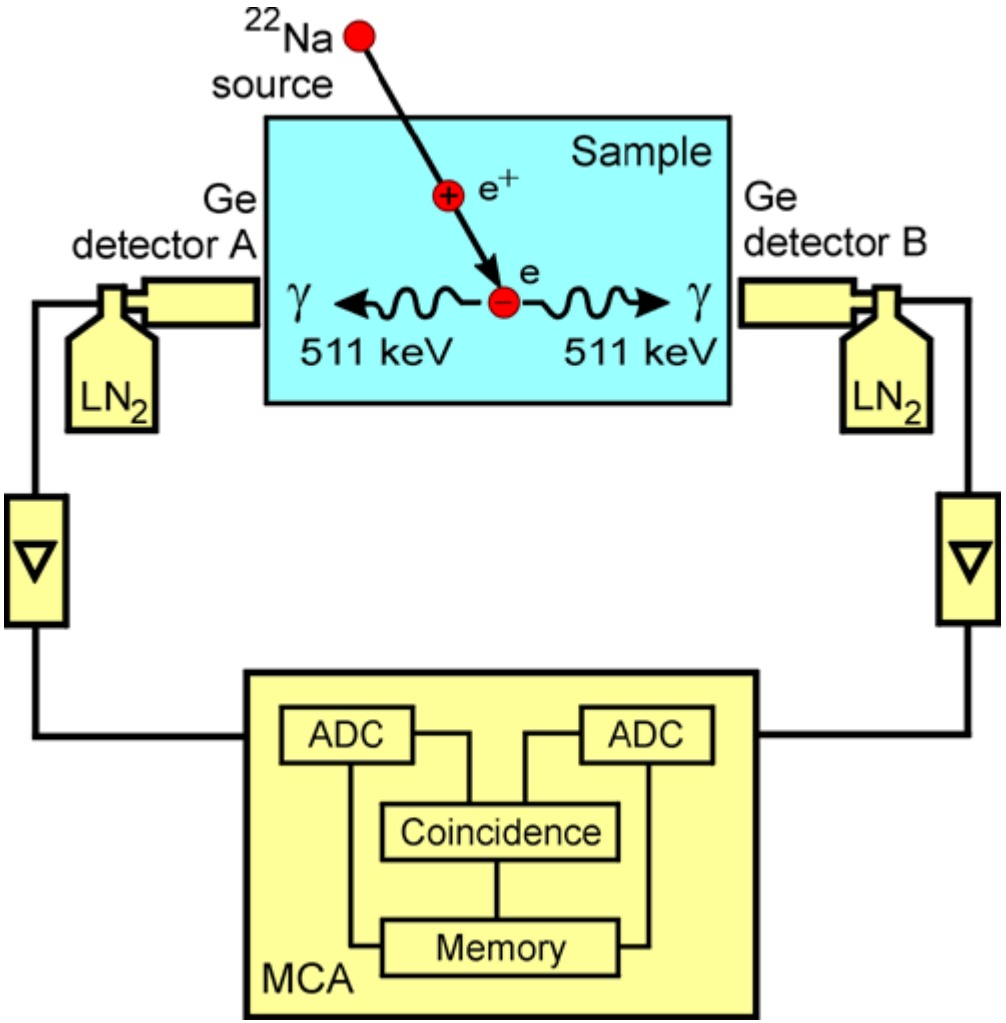
$S = A_S/A_0$

W parameter:

$W = A_W/A_0$

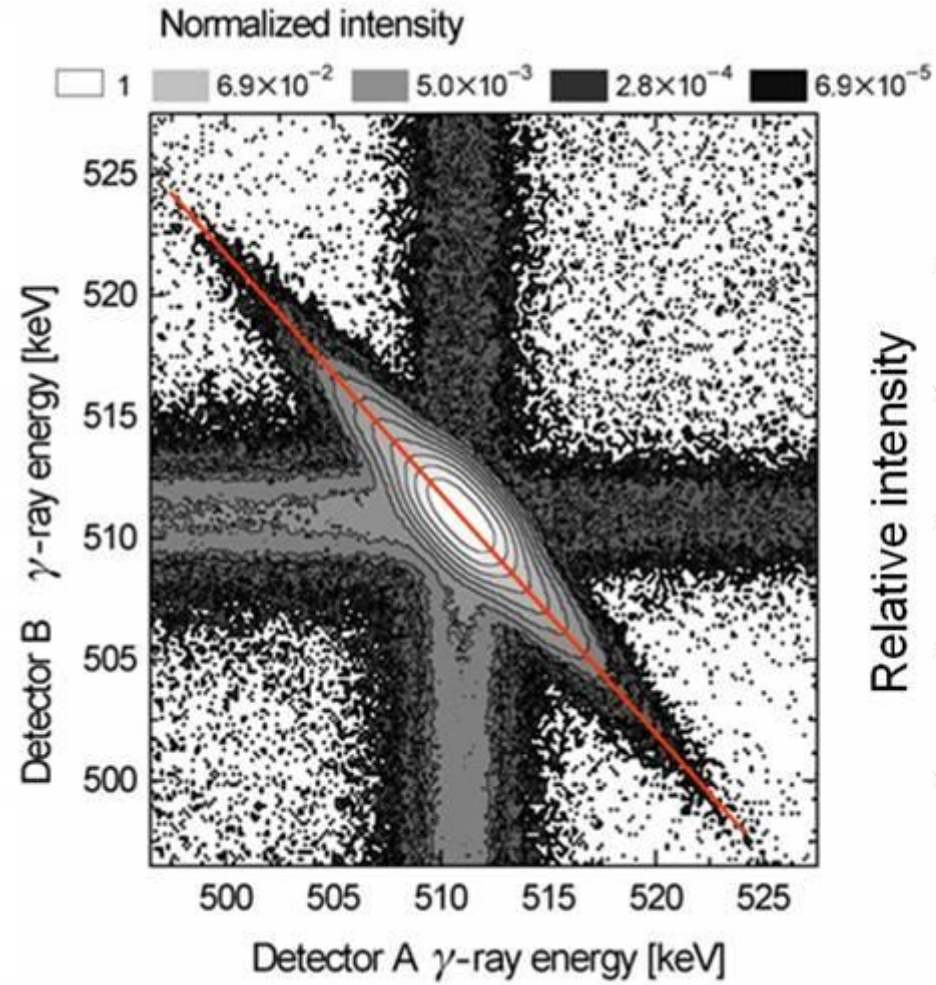
W parameter mainly determined by annihilations of core electrons (chemical information)

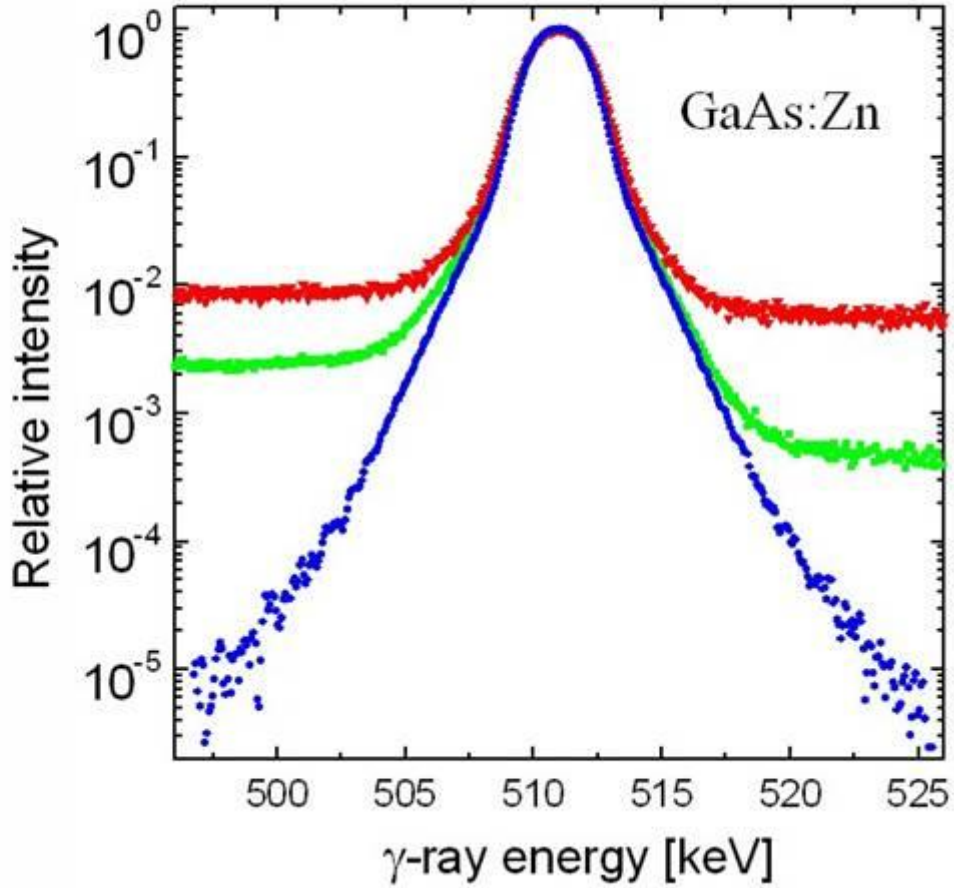
Doppler Coincidence Spectroscopy



- coincident detection of second annihilation γ reduces background
- use of a second Ge detector improves energy resolution of system

Doppler Coincidence Spectra

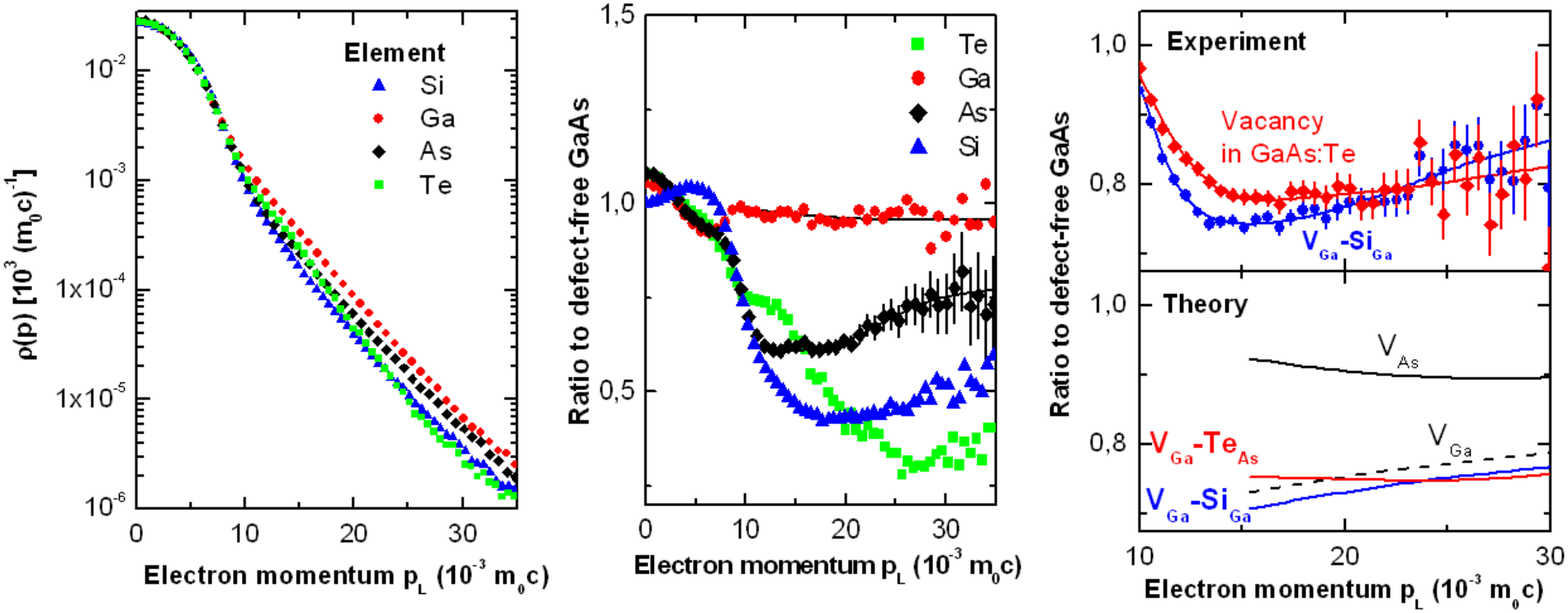




$$E_1+E_2= 2\ m_0\ c^2 =1022\ \text{keV}$$

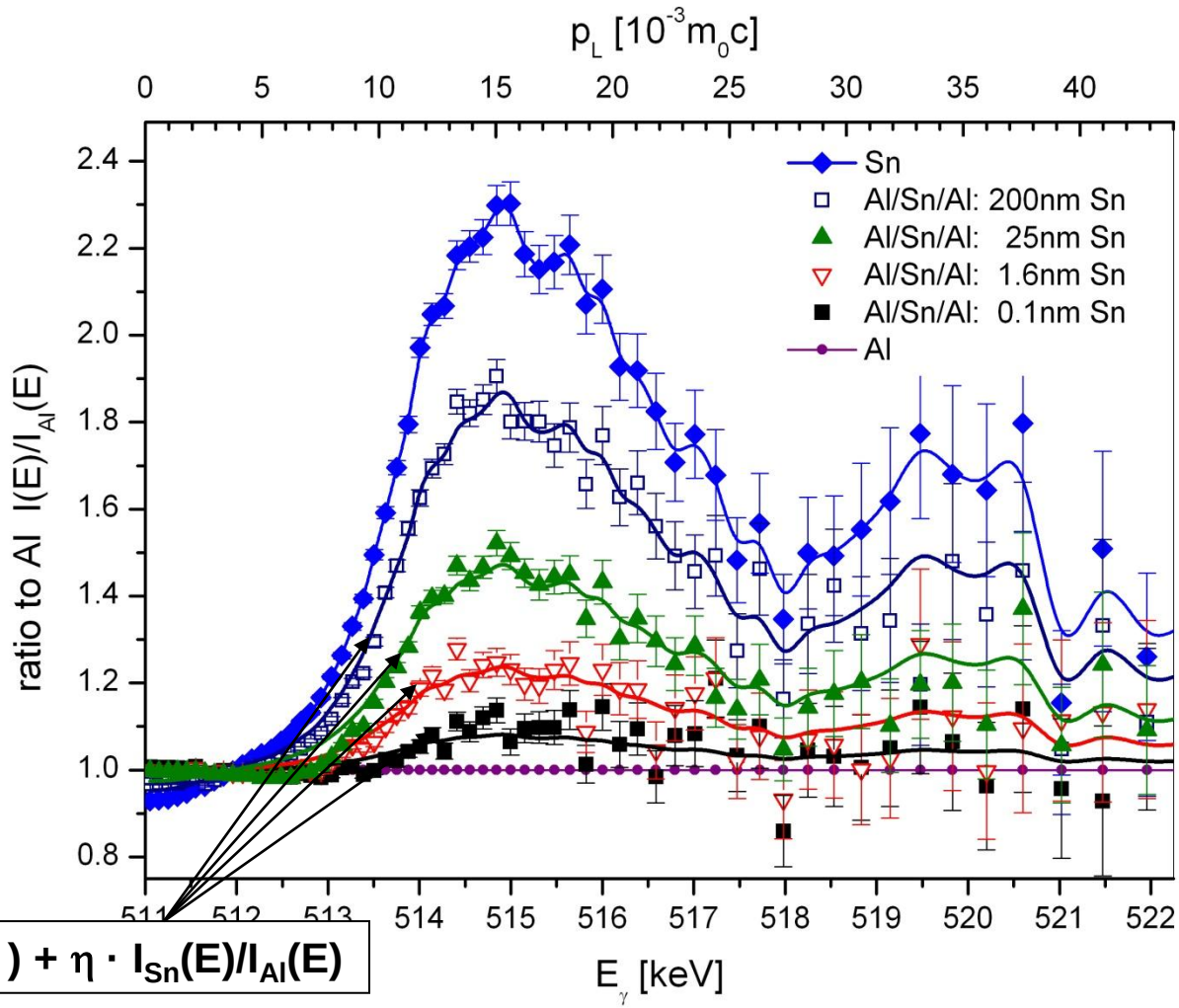
Doppler-Coincidence-Spectroscopy in GaAs

- Chemical sensitivity due to electrons at high momentum (core electrons)
- a single impurity atom aside a vacancy is detectable
- examples: V_{Ga}-Te_{As} in GaAs:Te



J. Gebauer et al., Phys. Rev. B **60** (1999) 1464

High Sensitivity of CDBS for thin Layers



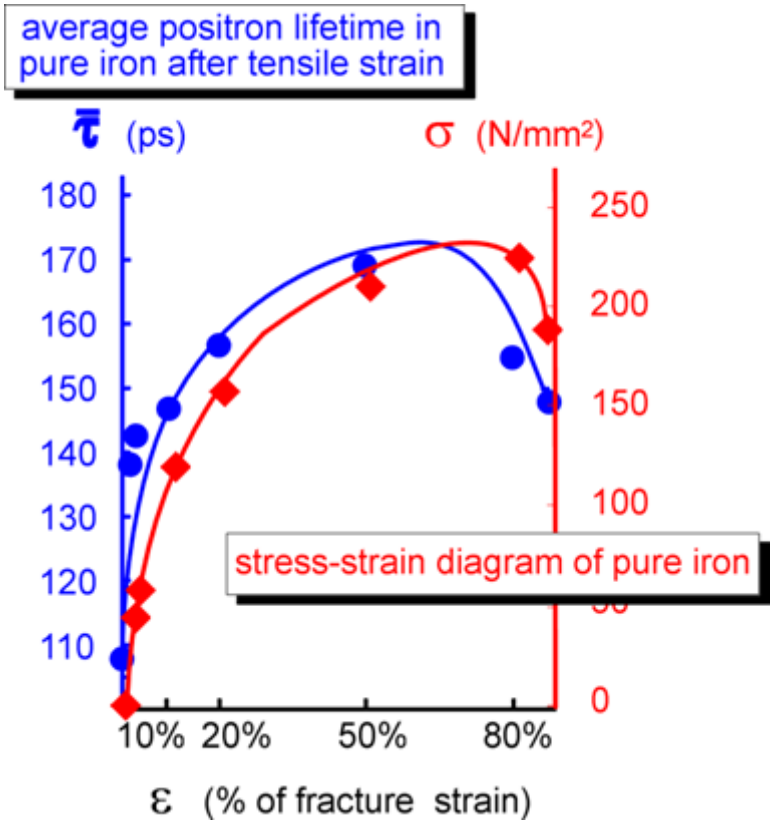
$$I_{ratio}(E) = (1 - \eta) + \eta \cdot I_{Sn}(E)/I_{Al}(E)$$

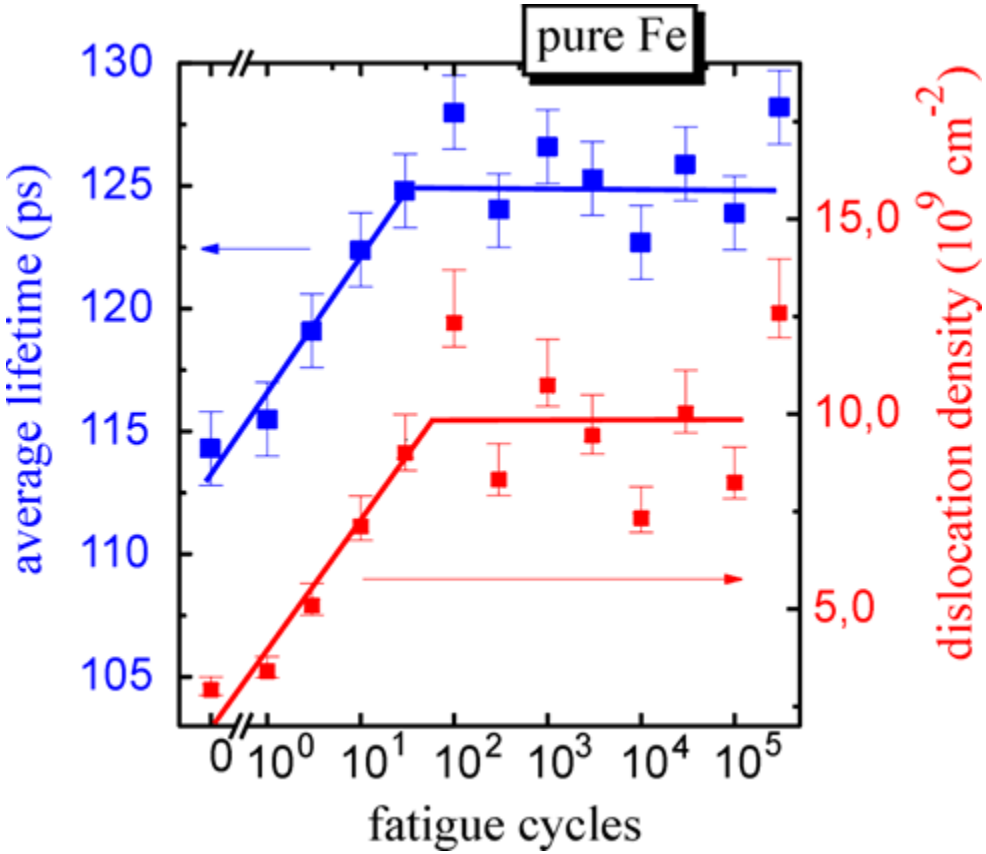
- Sn layers of different thickness under 200 nm Al
- even very thin layers are visible

C. Hugenschmidt et al. PRB 77 (2008) 092105

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

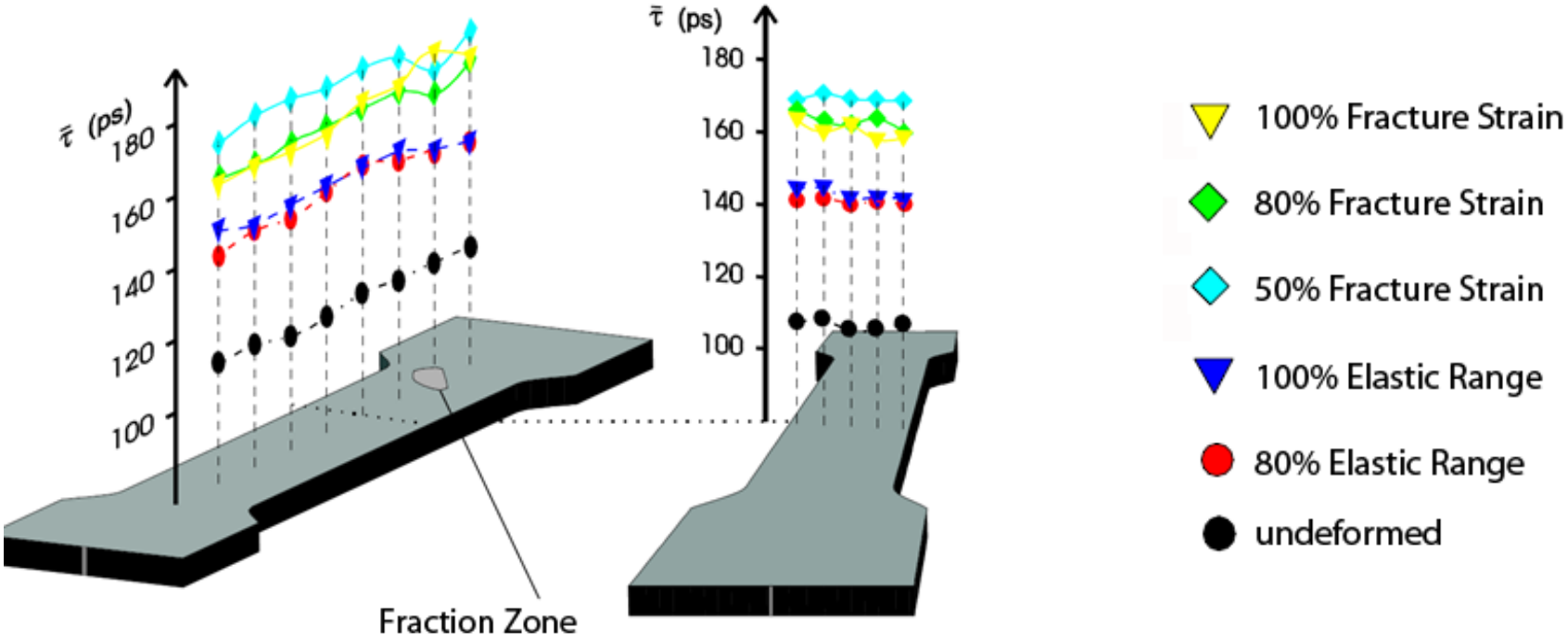




Somieski et al., J. Physique IV 5, C1/127-134 (1995)

Laterally resolved measurement across Test sample

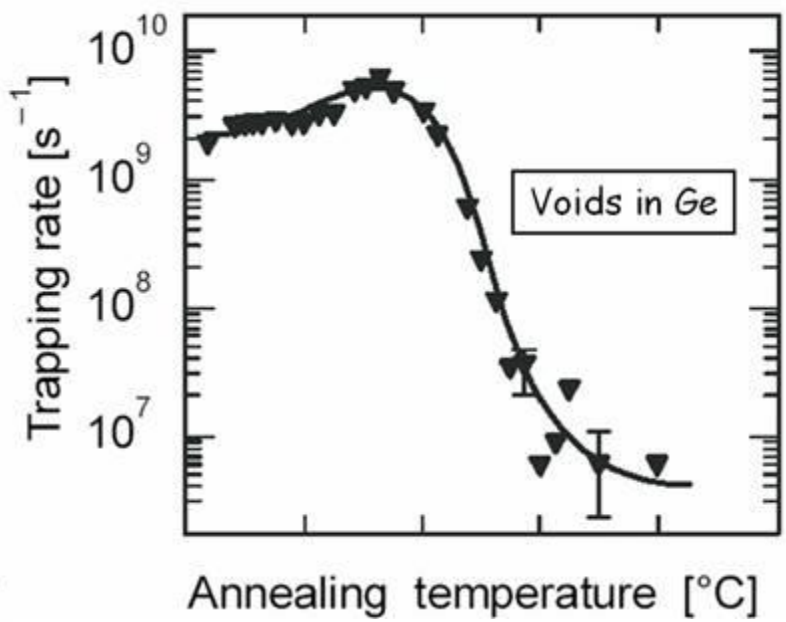
- Pure Fe sample
- strong damage already for strain < 100% of Hooke's range -> technically most interesting range
- fraction zone can be predicted from positron measurements



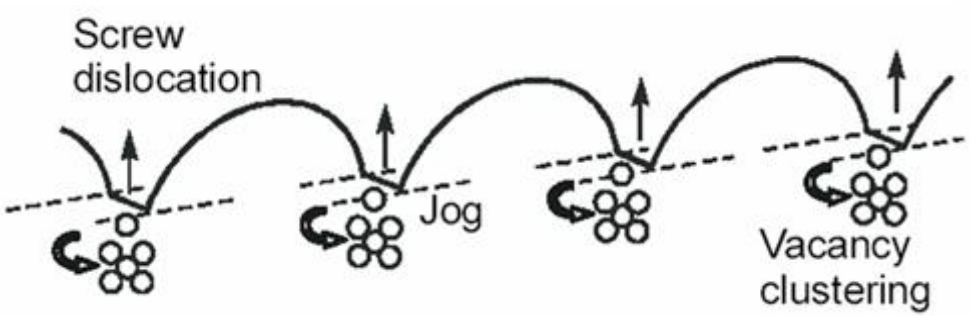
Somieski et al., J. Physique IV 5, C1/127-134 (1995)

Vacancy clusters in semiconductors

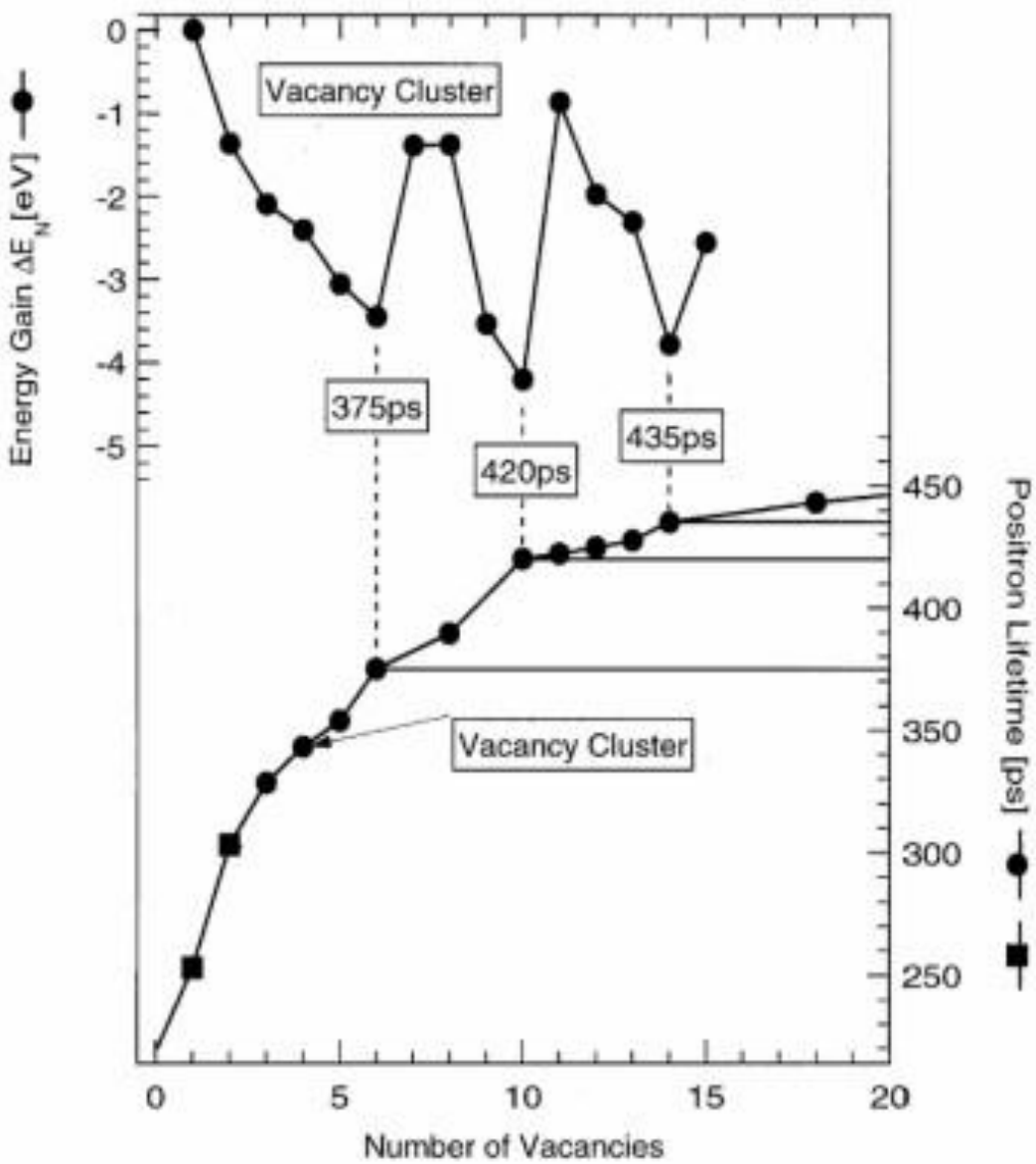
- vacancy clusters were observed after neutron irradiation, ion implantation and plastic deformation
- due to large open volume (low electron density) -> positron lifetime increases distinctly
- example: plastically deformed *Ge*
- lifetime: τ = 525 ps
- reason for void formation: jog dragging mechanism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



Krause-Rehberg et al., 1993



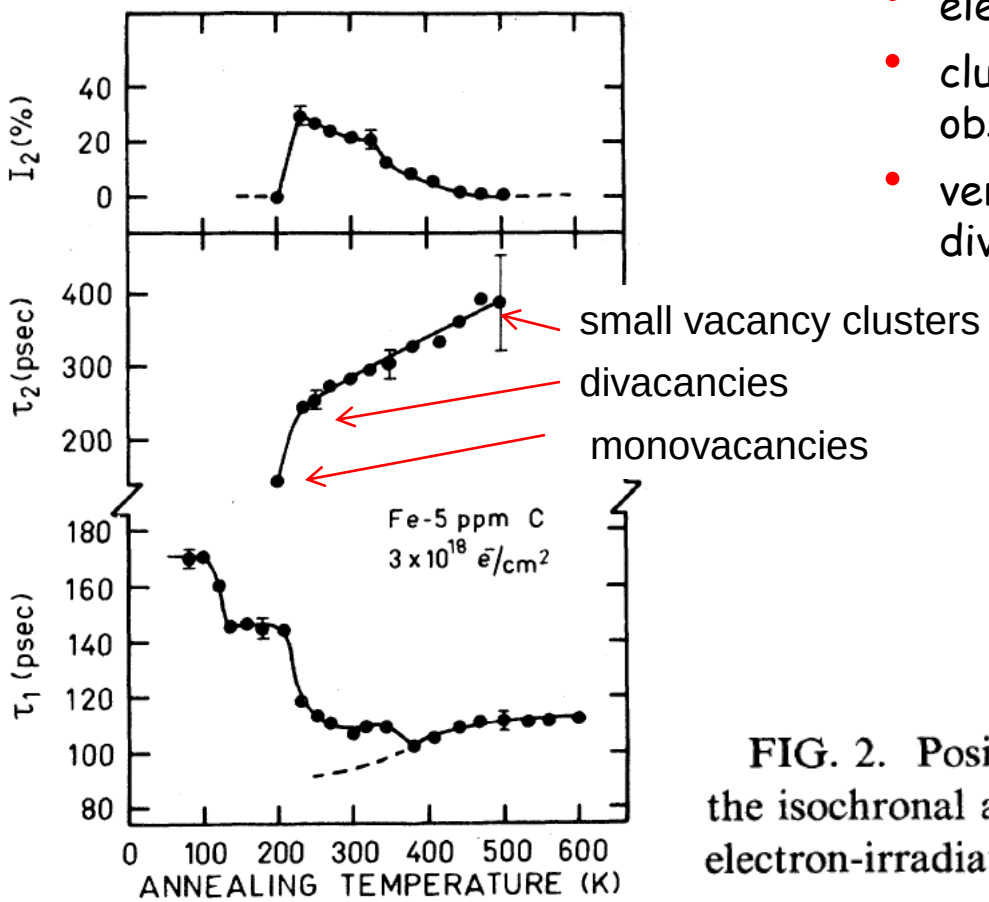
Theoretical calculation of vacancy clusters in Si



- 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

Vacancy clustering during defect annealing

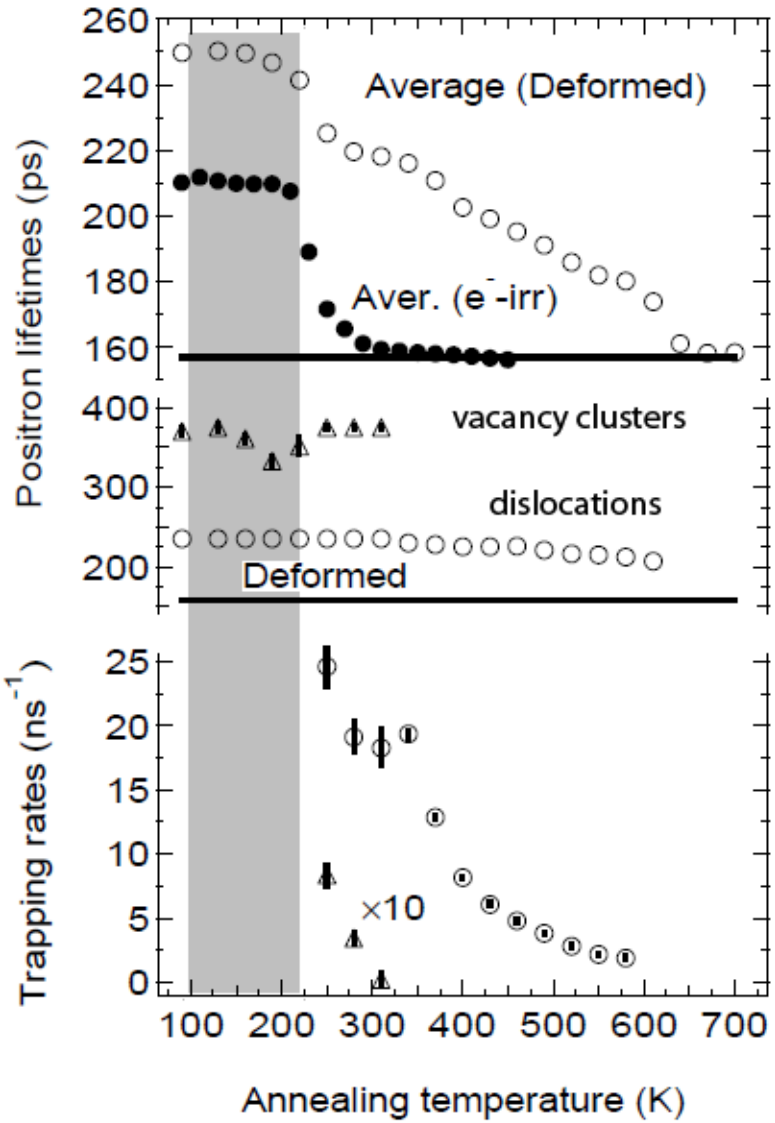


- electron irradiated Fe
- clustering in early stage can be observed
- very sensitive: formation of divacancies and small clusters (n < 10)

FIG. 2. Positron-lifetime parameters as a function of the isochronal annealing temperature in the low-dose electron-irradiated pure iron.

Vehanen et al., Phys. Rev. B 25, 762–780

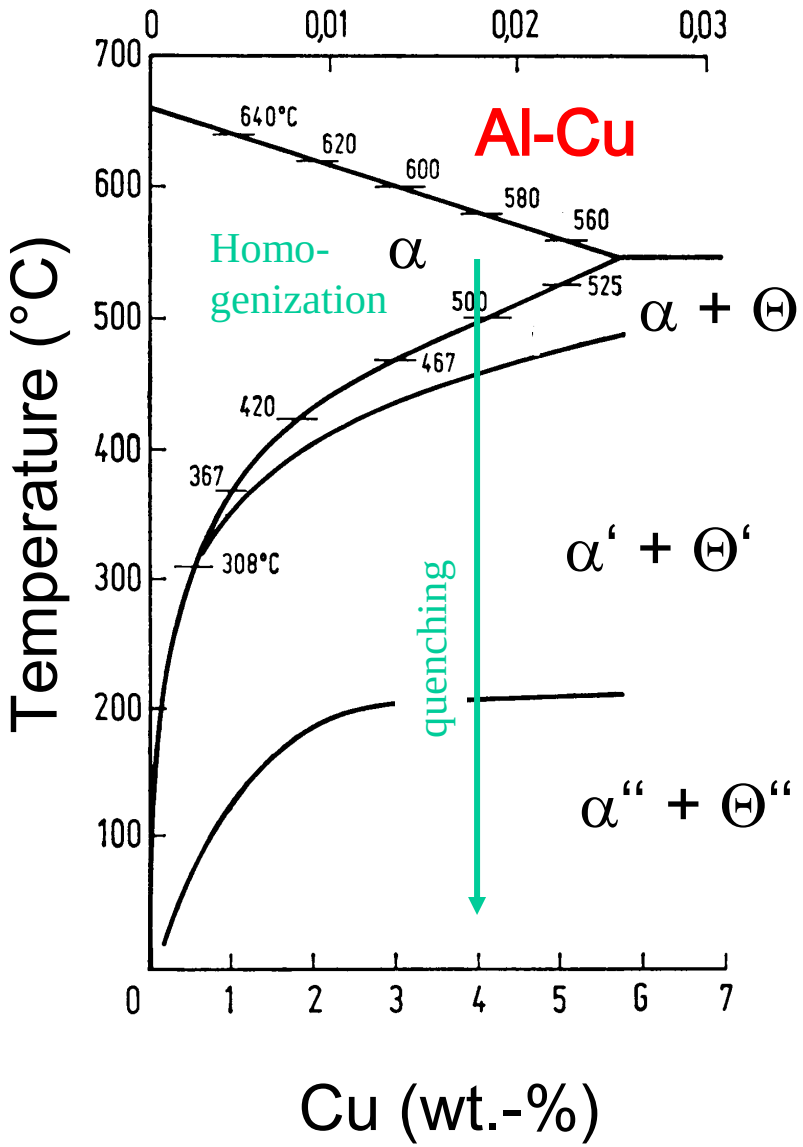
Plastic deformation of Al at 77K



- plastic deformation in LN2
- only gradual annealing (opposite to electron irradiation)
- two defects:
  - small vacancy clusters (n < 10) – annealing at RT
  - dislocations anneal at 600 K

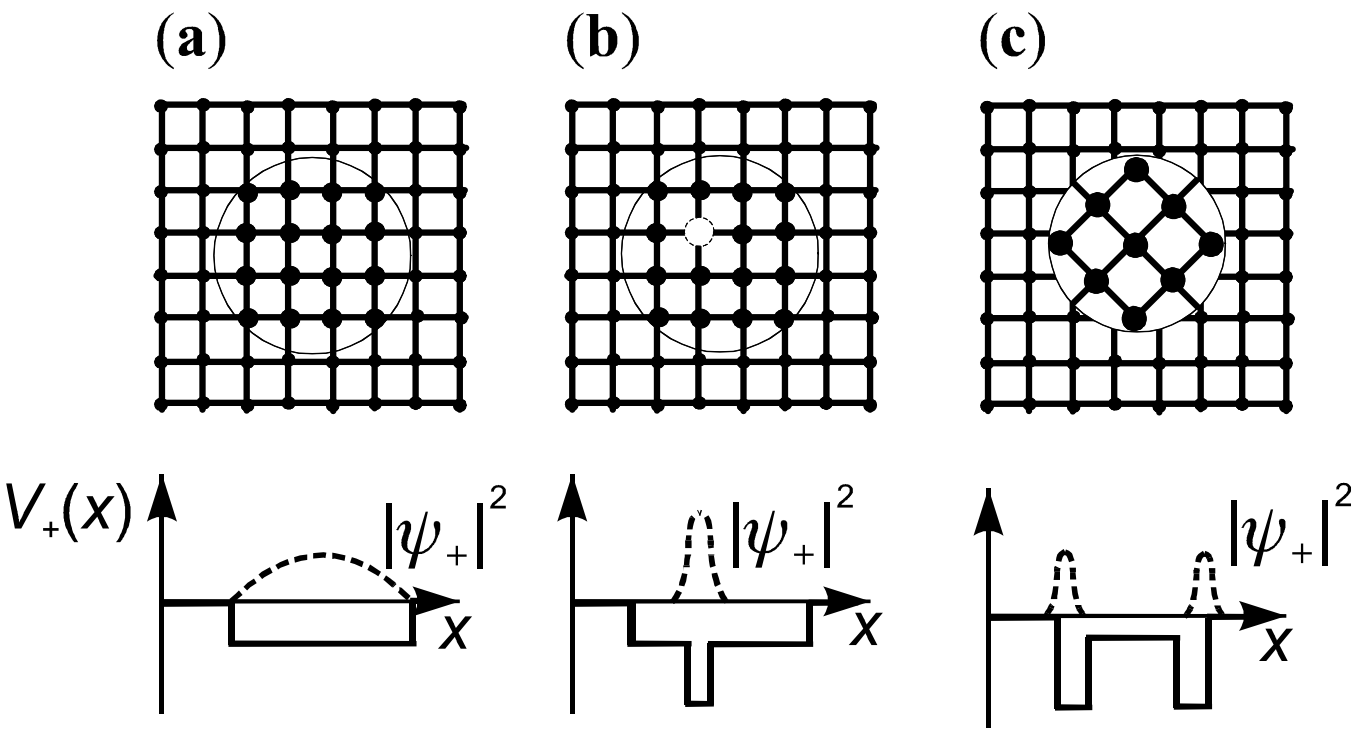
Chr. Hübner, Ph.D Thesis, Uni Halle, 1998

Precipitation phenomena in Al alloys



- Homogenization at 550°C dissolves Cu in Al
- Quenching to RT: oversaturation of Cu
- Equilibrium cannot be obtained at RT (Θ-Phase: $CuAl_2$)
- metastable particles are formed: Θ'' (Guinier-Preston-Zones) and Θ'
- are fully coherent in the beginning
- have extended strain field
- obstacles for dislocation motion: hardness increases
- transition to semi- or incoherent particles: stress field collapses
- hardness decreases

Positron capture at Precipitates



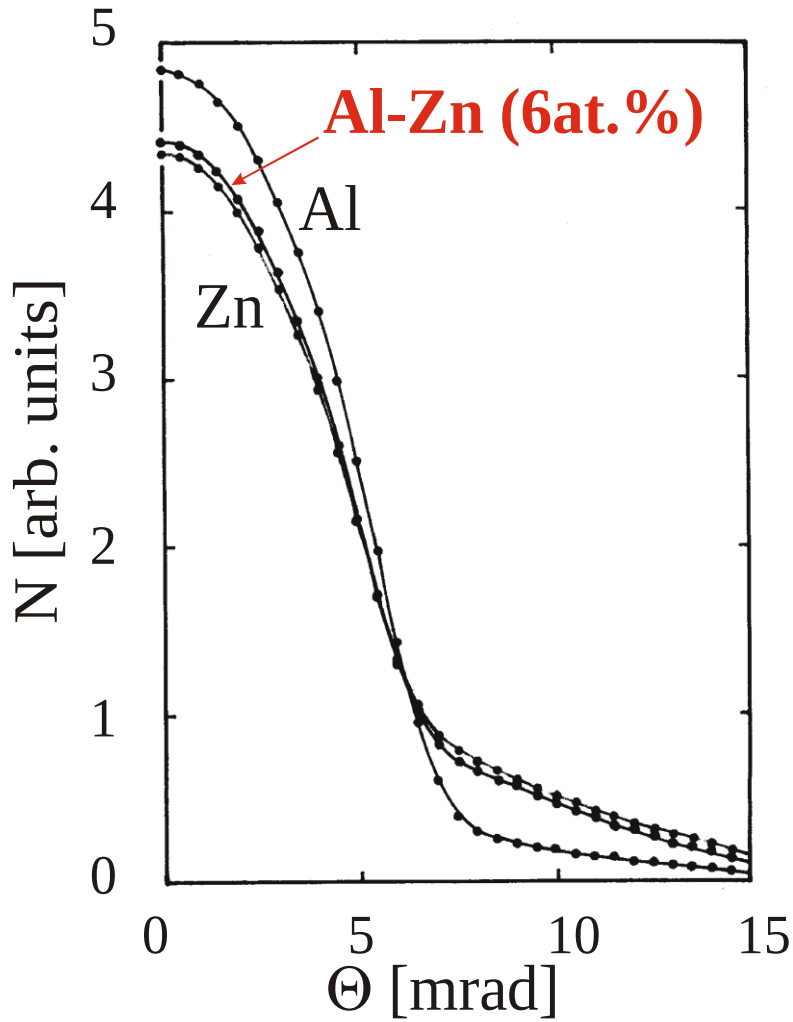
fully coherent	defect inside	semi- or incoherent
GPZ in Al Zn	Mg-V-Paar in Al ZnMg	Al Si

Positron capture in GPZ in Al-Zn (6 at%)

- Quenching of Al-Zn (6at%) from α-Phase: formation of fully coherent GPZ
- are free of defects (no vacancies, no dislocations)
- 1-dimensional ACAR curves
- Zn-content η of GPZ:

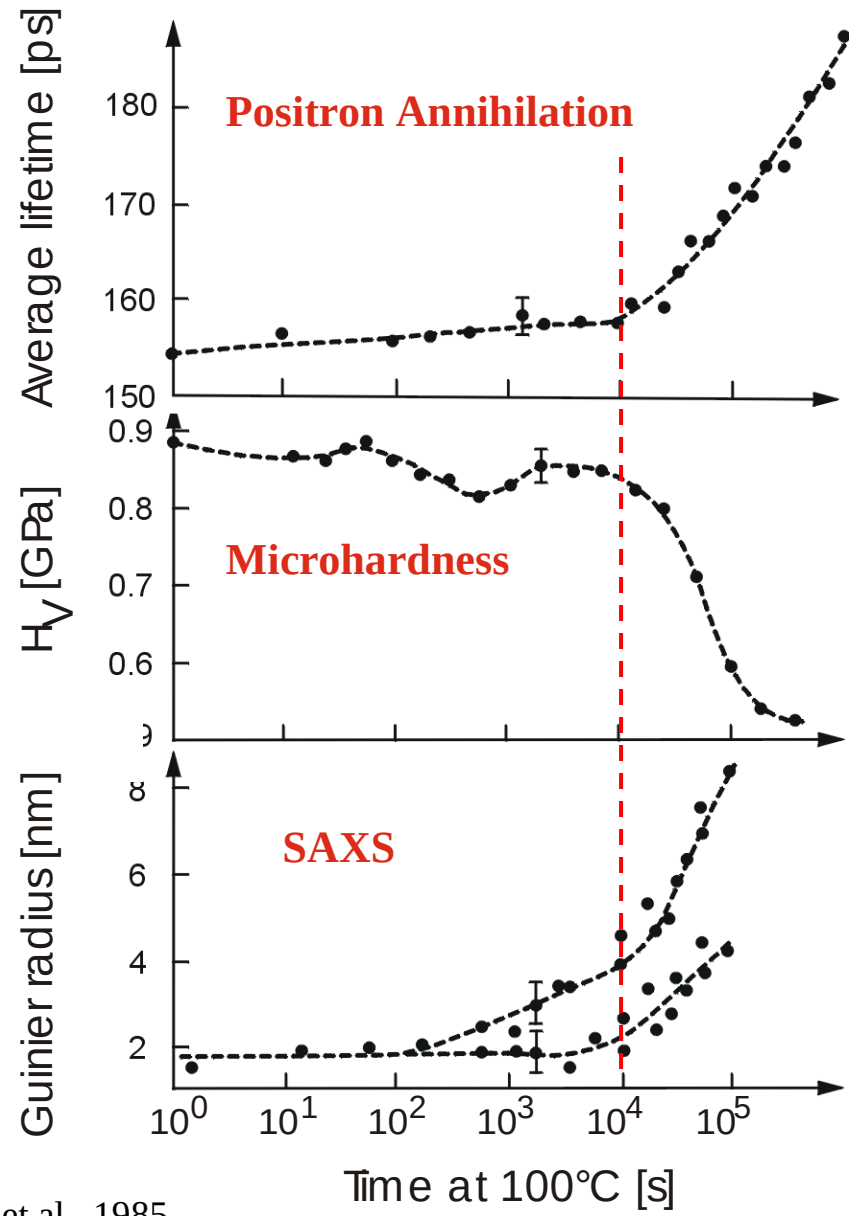
$$N_{AlZn} = (1-\eta)\, N_{Al} + \eta\, N_{Zn}$$

- Result: $\eta = 70\%$
- formation and dissolution of precipitations can be studied



Age-hardening of Al-Zn(15) at 100°C

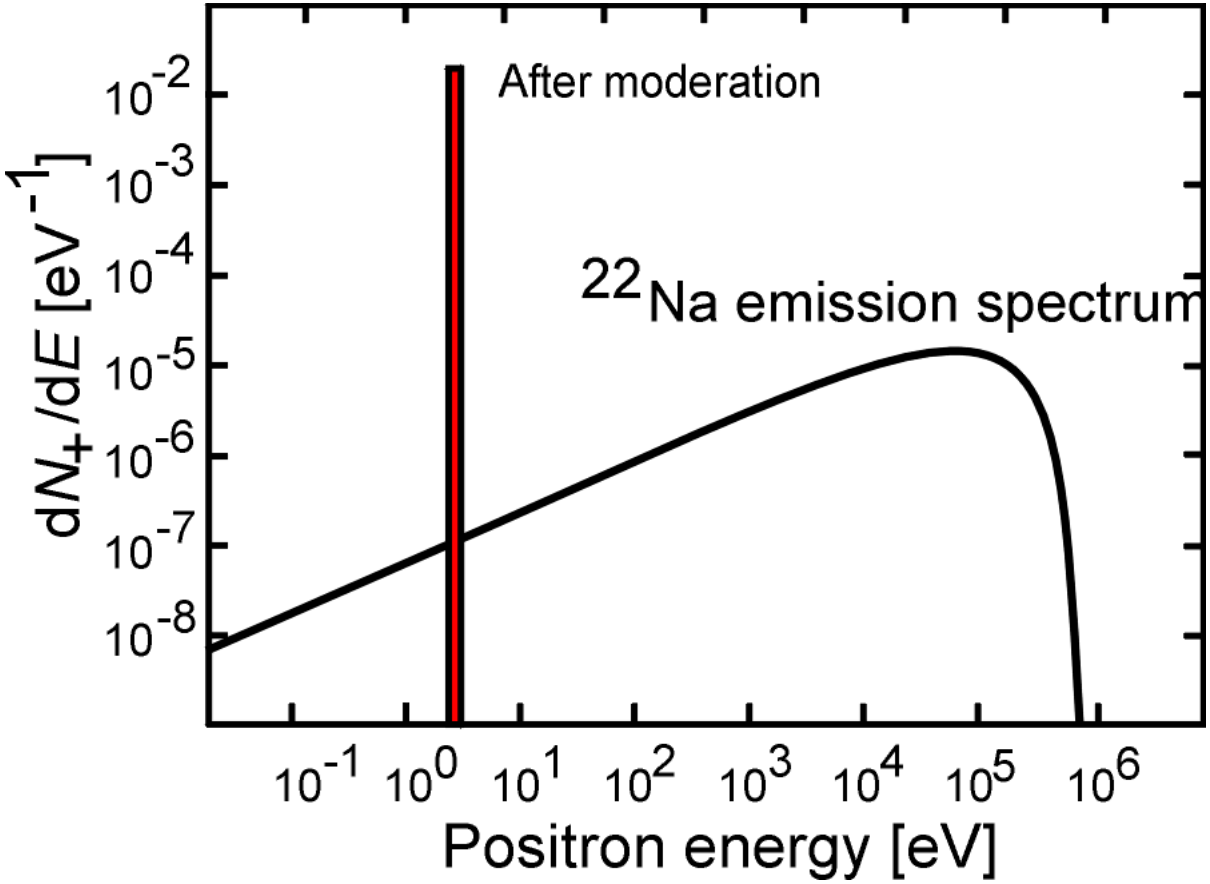
- Aging at 100°C: Growth of GPZ
- start to get ellipsoidal
- strong hardness reduction
- positrons detecting dislocation at GPZ
- transition of fully coherent GPZ to semi-coherent α'_R-particles
- positron detect directly the first misfit dislocation around the precipitate



R. Krause et al., 1985

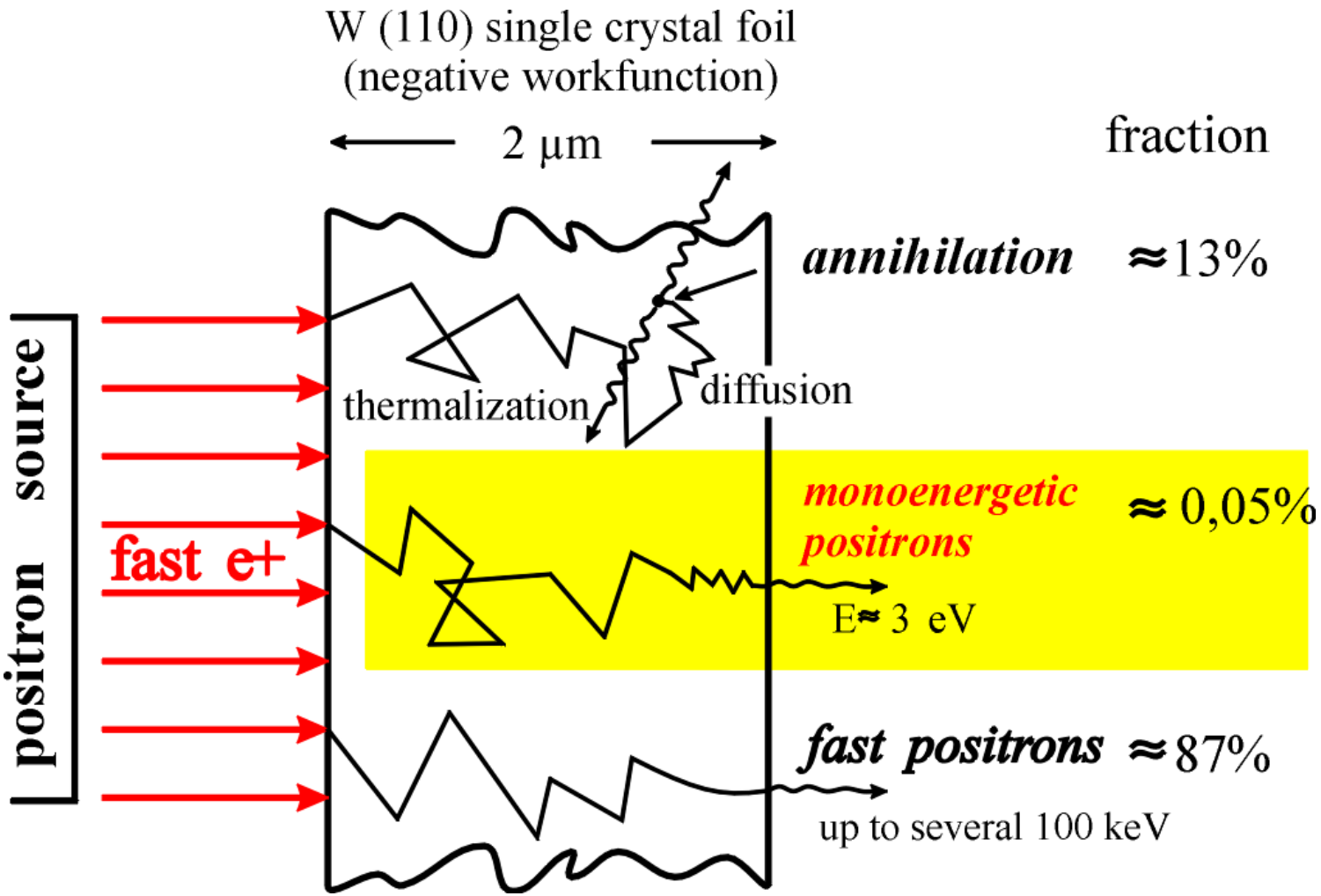
Moderation of Positrons

Mean implantation depth of un-moderated positrons from a isotope ^{22}Na source (1/e): Si: 100μm



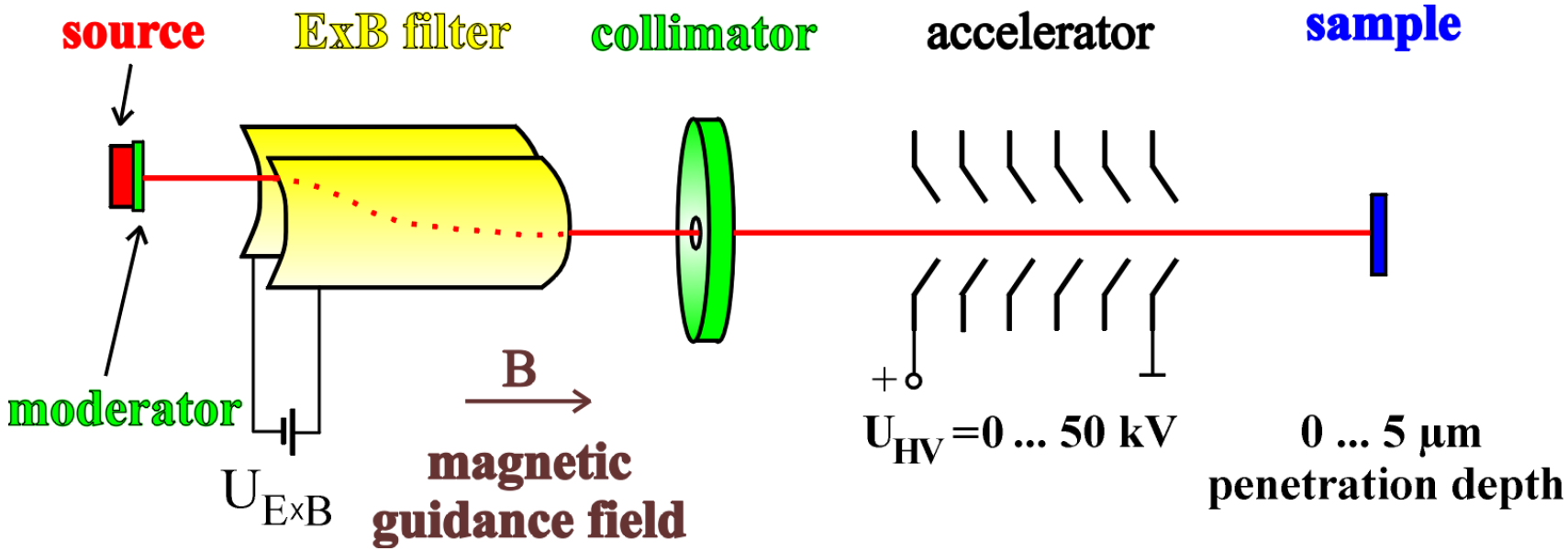
- broad β^+ positron emission spectrum
- deep implantation into solids
- not useful for study of defects in thin layers
- for defect depth profiling: moderation necessary
- monoenergetic positrons can be implanted to different depth

Moderation of Positrons



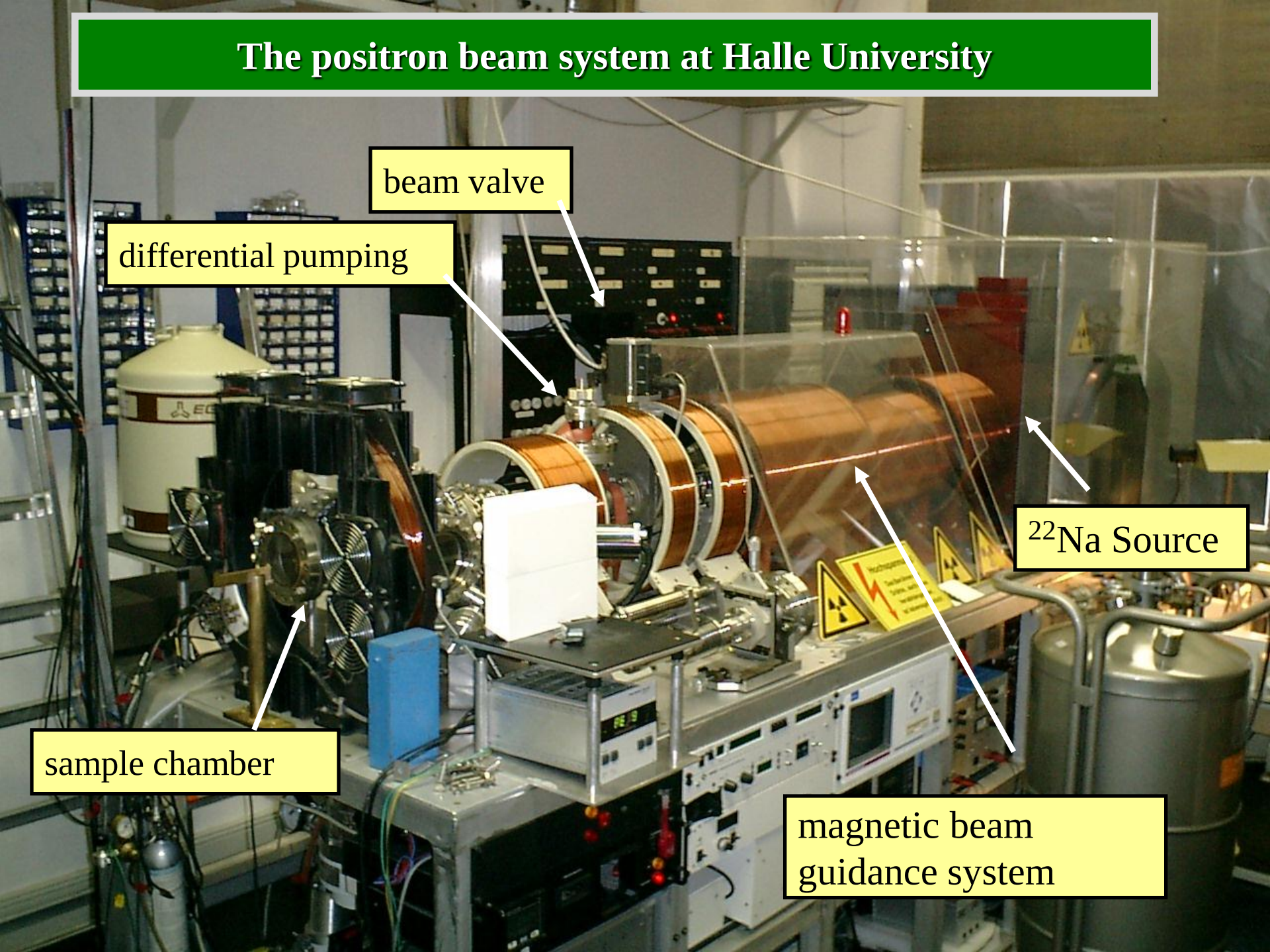
moderation efficiency: $\approx 10^{-4}$

The Positron Beam System at Halle University



- spot diameter: 5mm
- time per single Doppler measurement: 20 min
- time per depth scan: 8 hours

The positron beam system at Halle University



Study of Lubrication Defects

Tribology Letters Vol. 11, No. 1, 2001

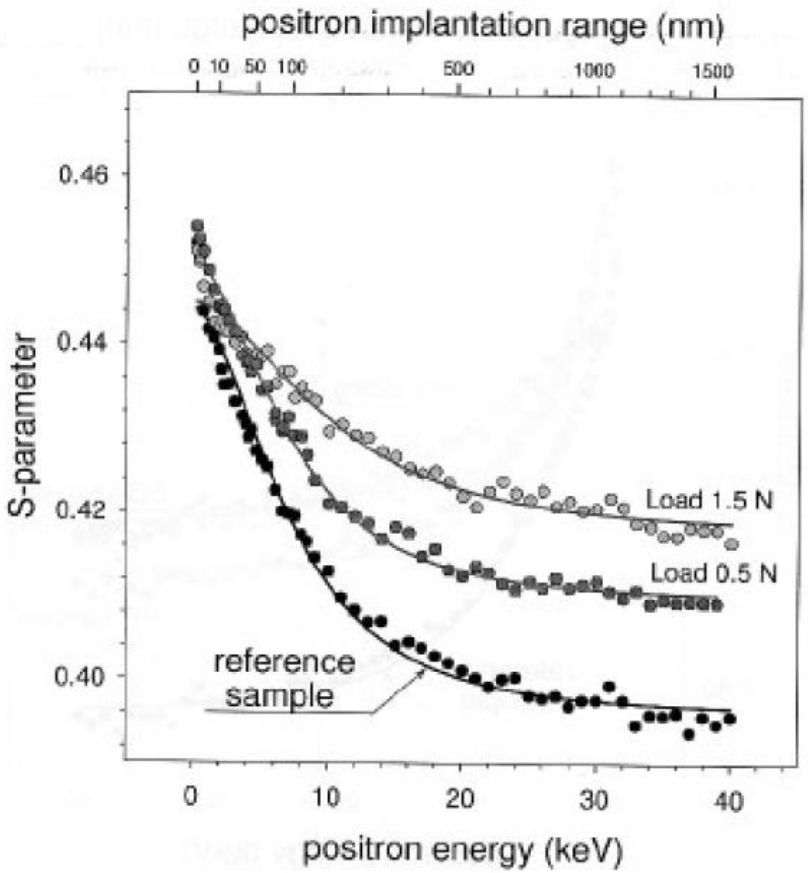
- Study of defects after lubrication treatment
- Steel ball on Cu surface
- effect of lubricant

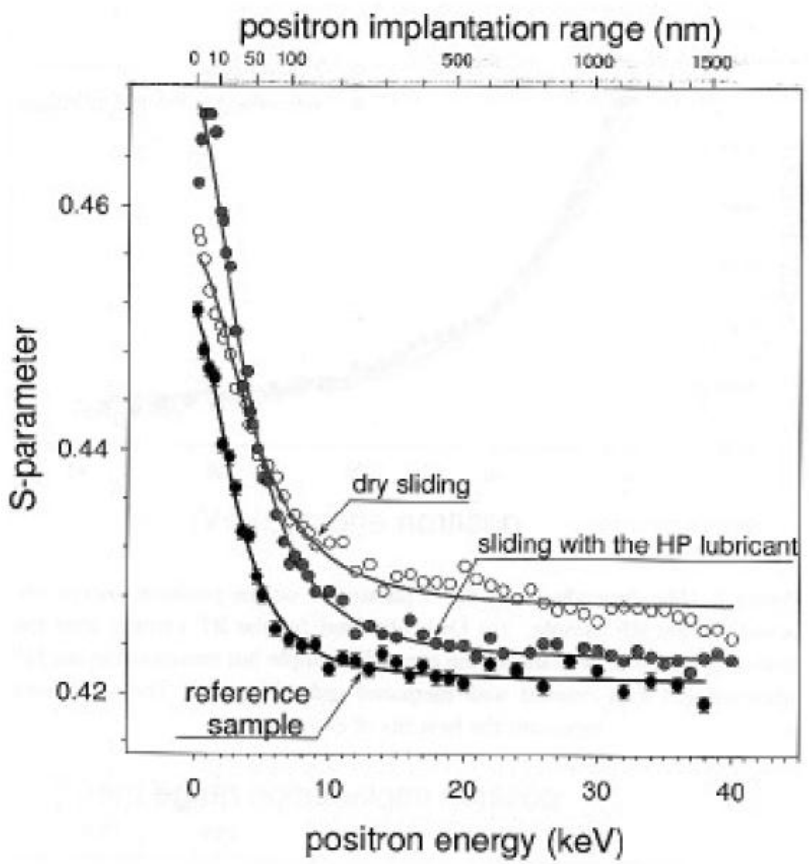
Subsurface zones created under lubrication conditions studied by positron annihilation

J. Dryzek [a], E. Dryzek [a], F. Börner [b] and R. Krause-Rehberg [b]

[a] *Institute of Nuclear Physics, ul. Radzikowskiego 152, 31-342 Kraków, Poland*
[b] *Universität Halle, Fachbereich Physik, D-06099 Halle/S, Germany*

Received 11 August 2000; accepted 22 February 2001



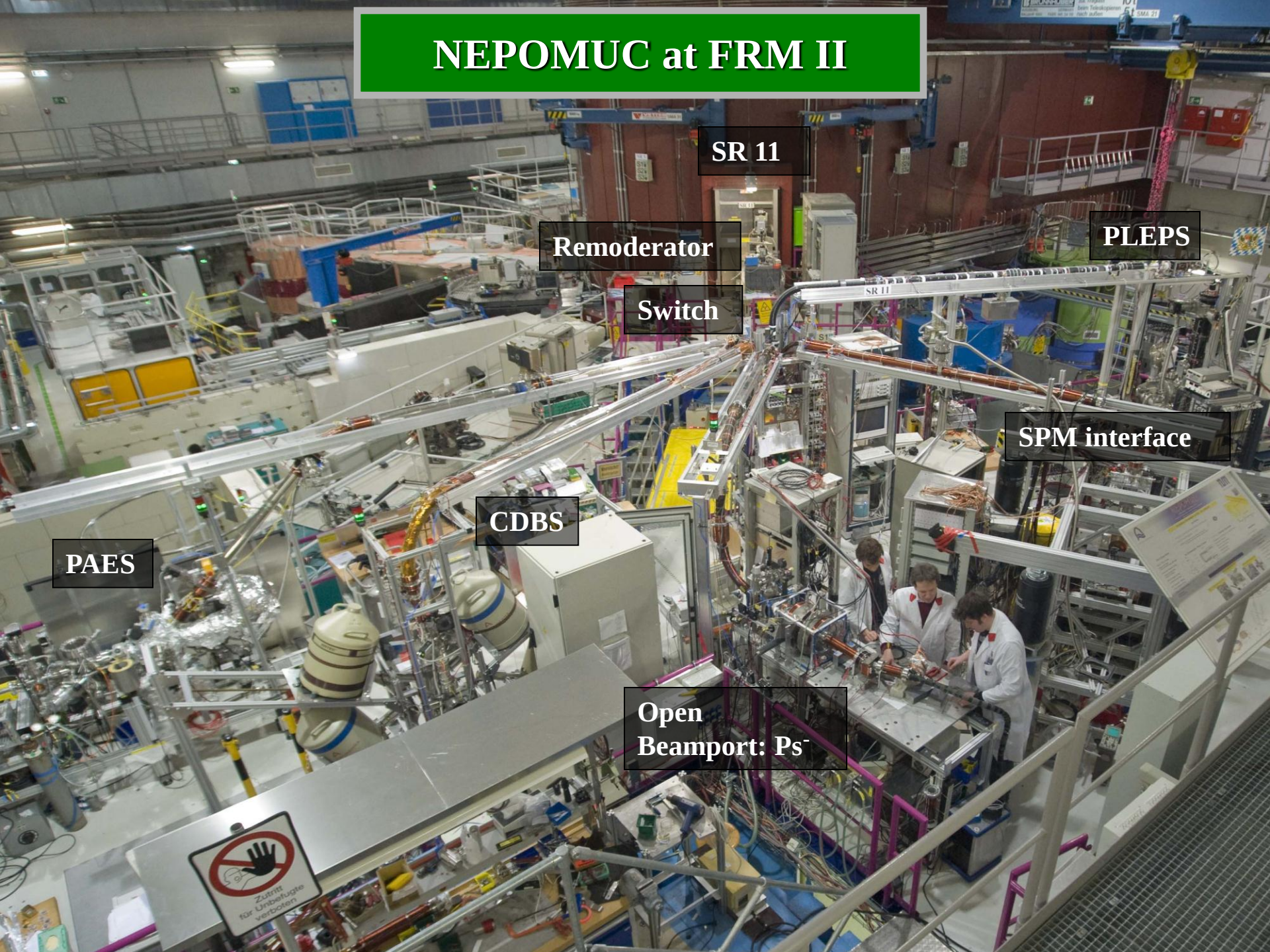


User-dedicated intense Positron Sources in Germany

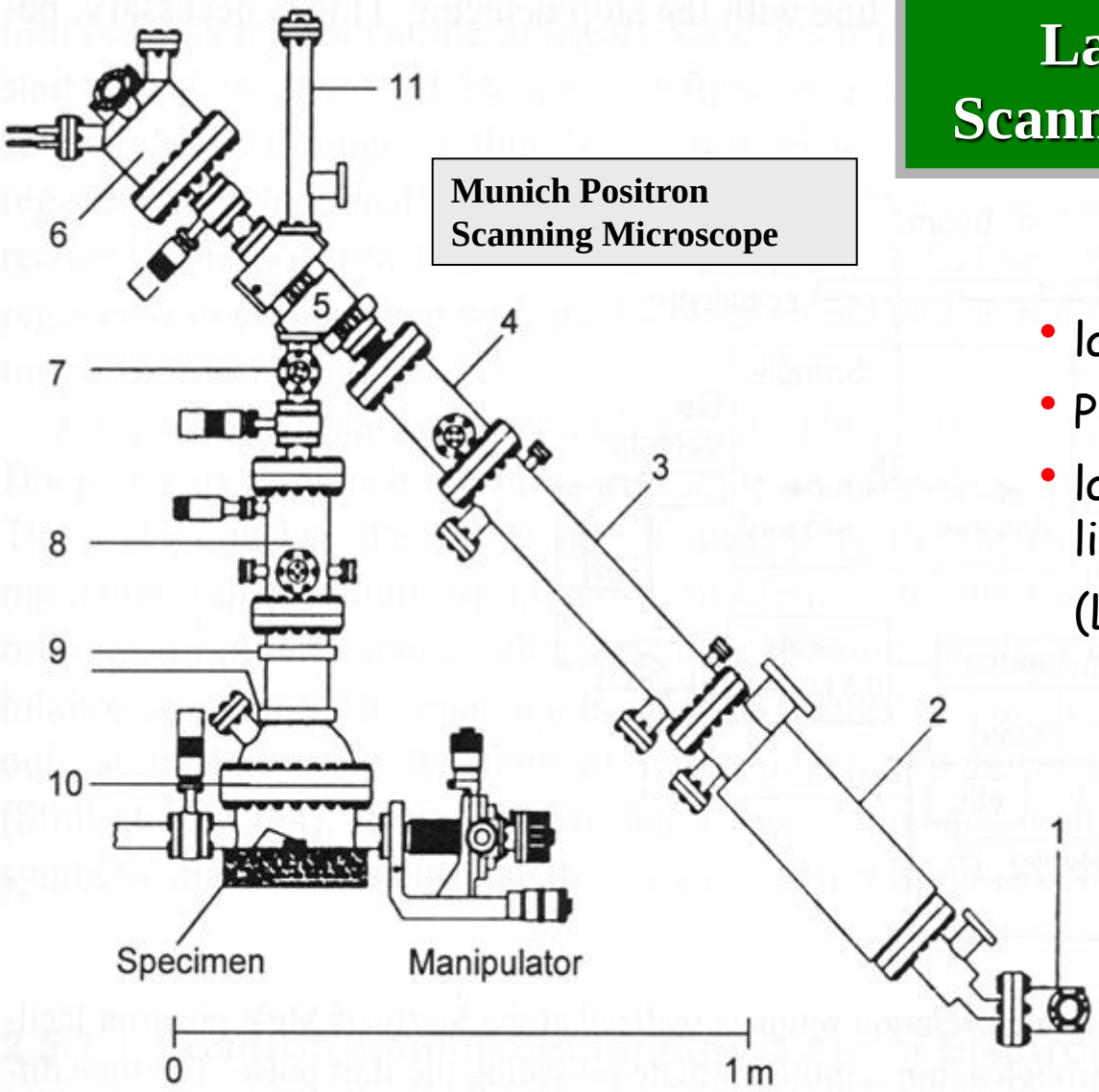
- Two intense positron sources available (positrons by pair production)
- **NEPOMUC** (NEutron induced POsitron Source MUniCh) at FRM-II
  - PLEPS (monoenergetic positron lifetime system)
  - PAES (Positron-induced Auger Electron Spectroscopy)
  - CDBS (Coincidence Doppler Broadening Spectroscopy)
  - SCM (Scanning Positron Microscope)
  - user beam line
- **EPOS** (ELBE Positron Source) at Research Center Dresden-Rossendorf
  - MePS (Mono-energetic Positron Spectroscopy)
  - GiPS (Gamma-induced Positron Spectroscopy)
  - CoPS (conventional setup using 22Na sources)

- at both sites: web-based application system for beam time

NEPOMUC at FRM II



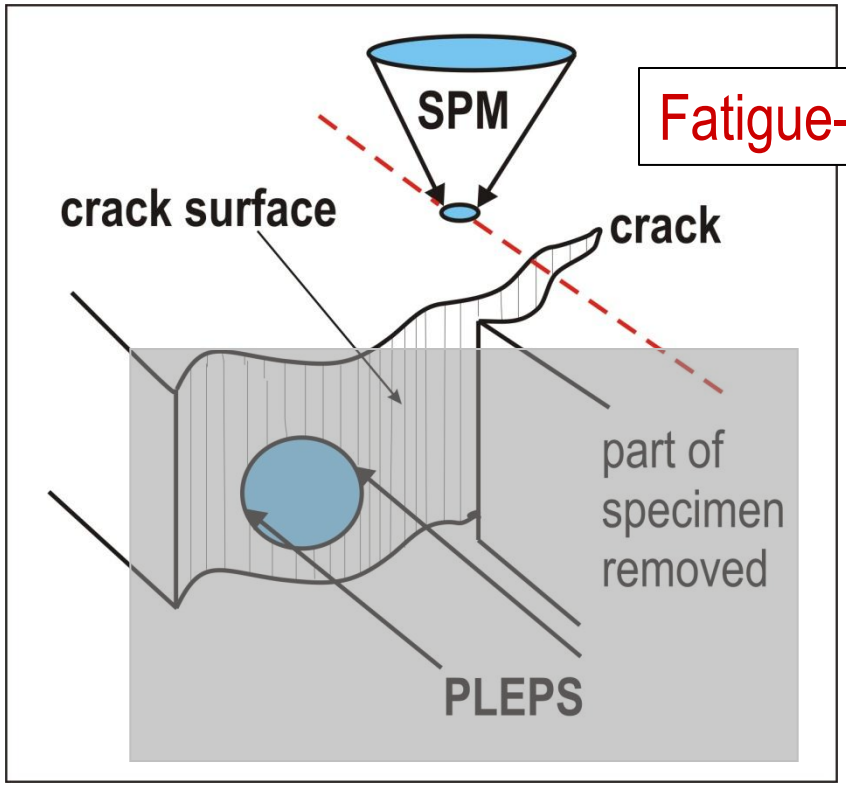
Lateral Resolution with Scanning Positron Microscope



- lateral resolution 1…2 μm
- Positron lifetime spectroscopy
- lateral resolution principally limited by positron diffusion ($L_+ \approx$ 100nm)

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

SPM on top of cracked sample

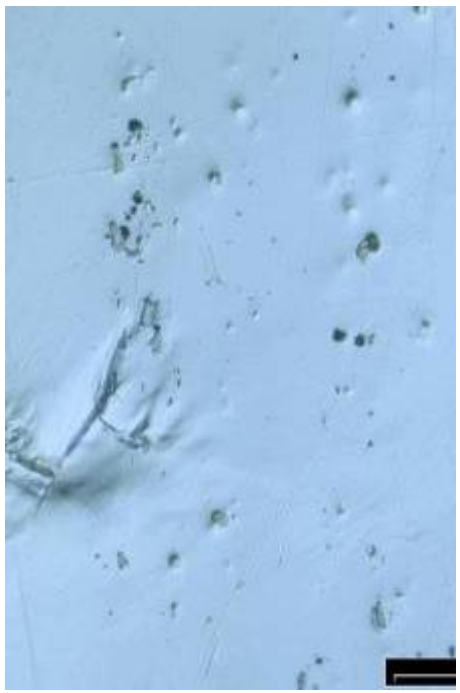


Fatigue-Crack in Al 6013

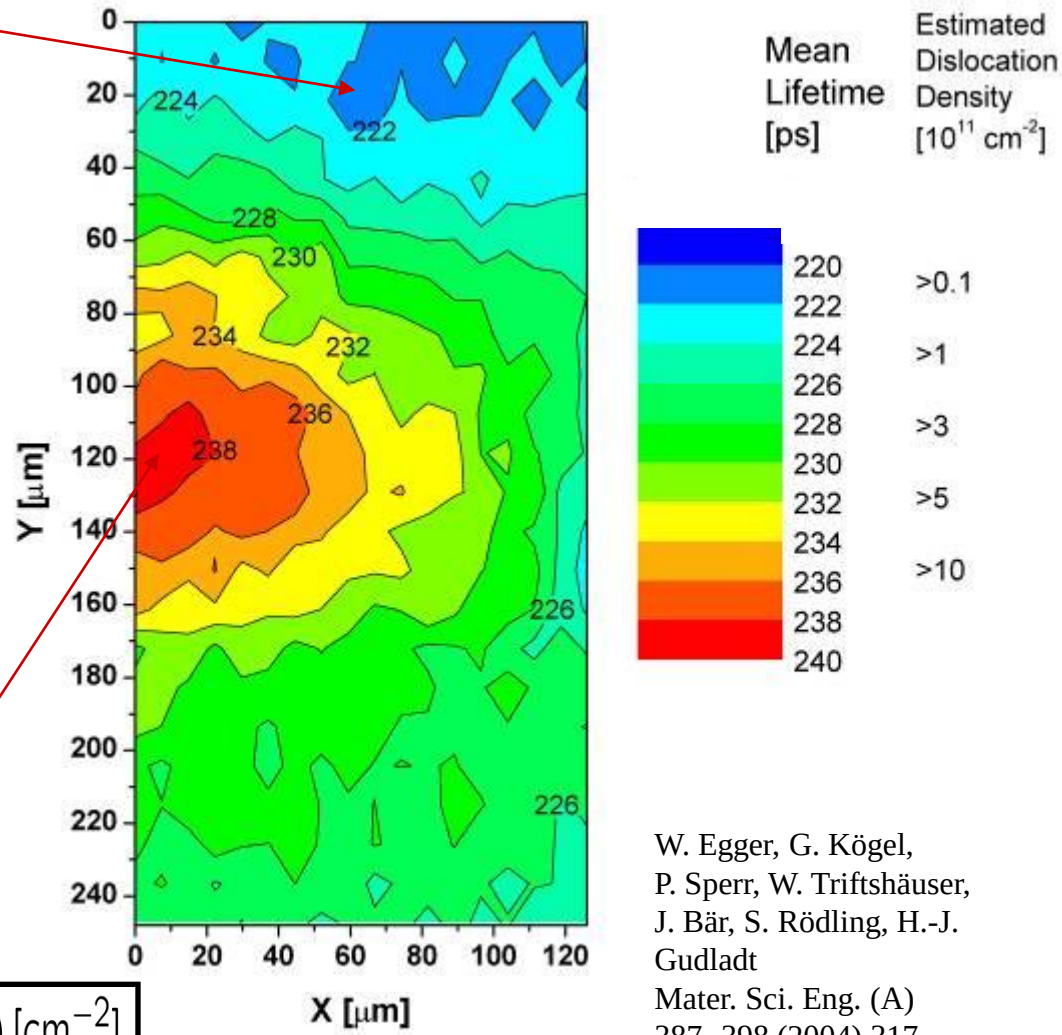
SPM: Positron Lifetime Image of Fatigue-Crack in Al 6013

resolution 5μm; step-size 10 μm

Trapping at Mg / Si-clusters!



Only dislocations close to crack-tip !



$$c_{disl} = 4 \cdot 10^{11} (\tau - 220\,\text{ps}) / (240\,\text{ps} - \tau)\ [\text{cm}^{-2}]$$

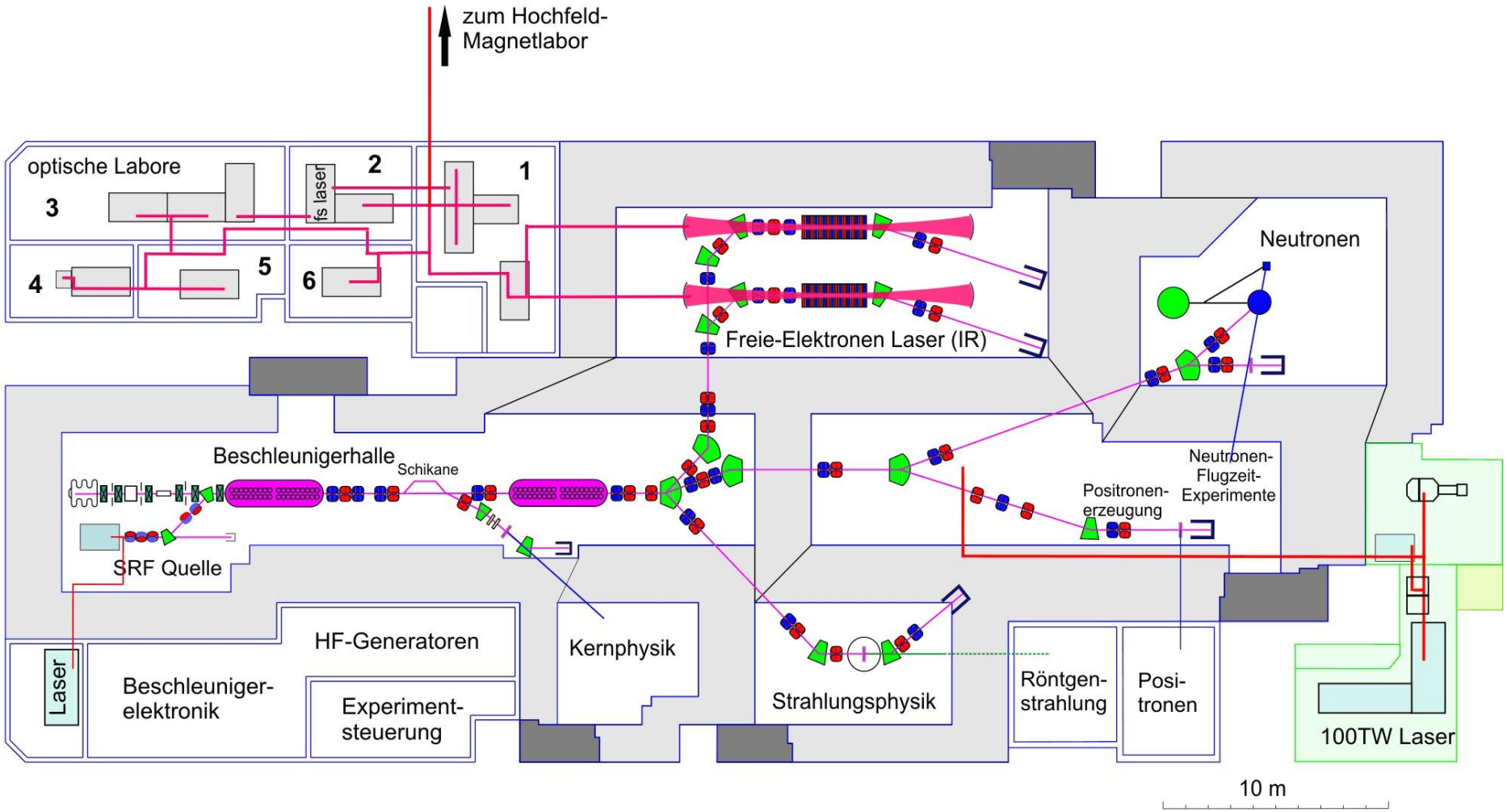
W. Egger, G. Kögel, P. Sperr, W. Triftshäuser, J. Bär, S. Rödling, H.-J. Gudladt
Mater. Sci. Eng. (A) 387- 398 (2004) 317

PLEPS at NEPOMUC

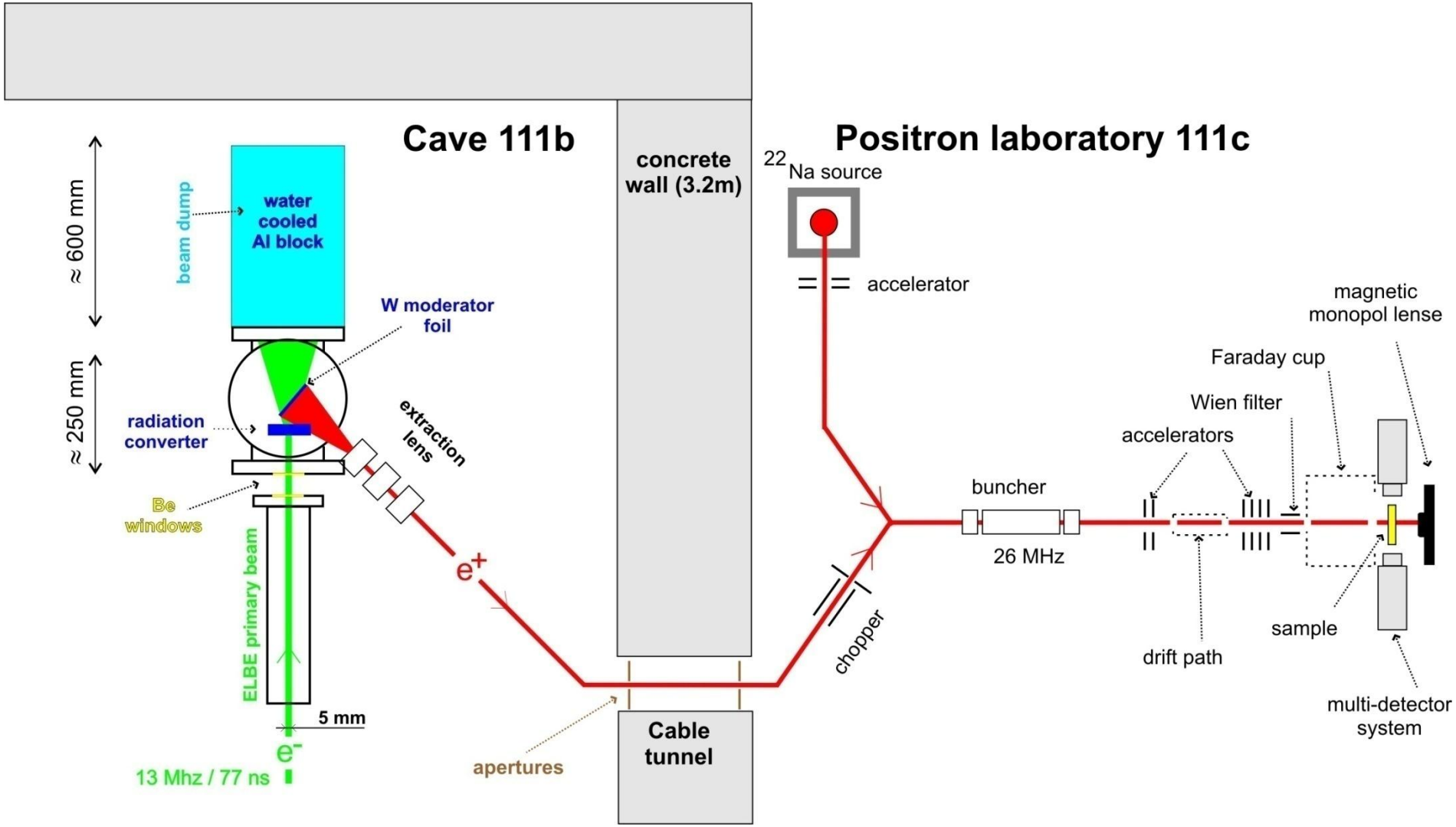
EPOS = ELBE Positron Source

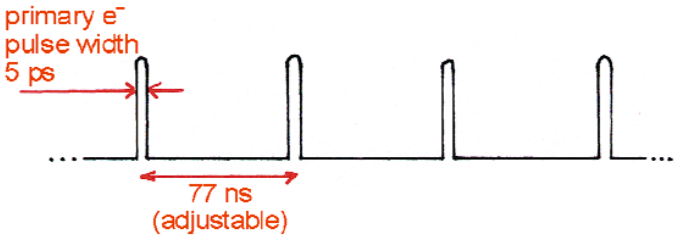
- ELBE -> electron LINAC (40 MeV and up to 40 kW) in Research Center Dresden-Rossendorf
- EPOS -> collaboration of Univ. Halle with FZD
- EPOS will be the combination of a positron lifetime spectrometer, Doppler coincidence, and AMOC
- User-dedicated facility
- main features:
  - high-intensity bunched positron beam (E_+ = 0.5…30 keV)
  - very good time resolution by using the unique primary time structure of ELBE
  - digital multi-detector array
  - fully remote control via internet by user

Ground plan of the ELBE hall



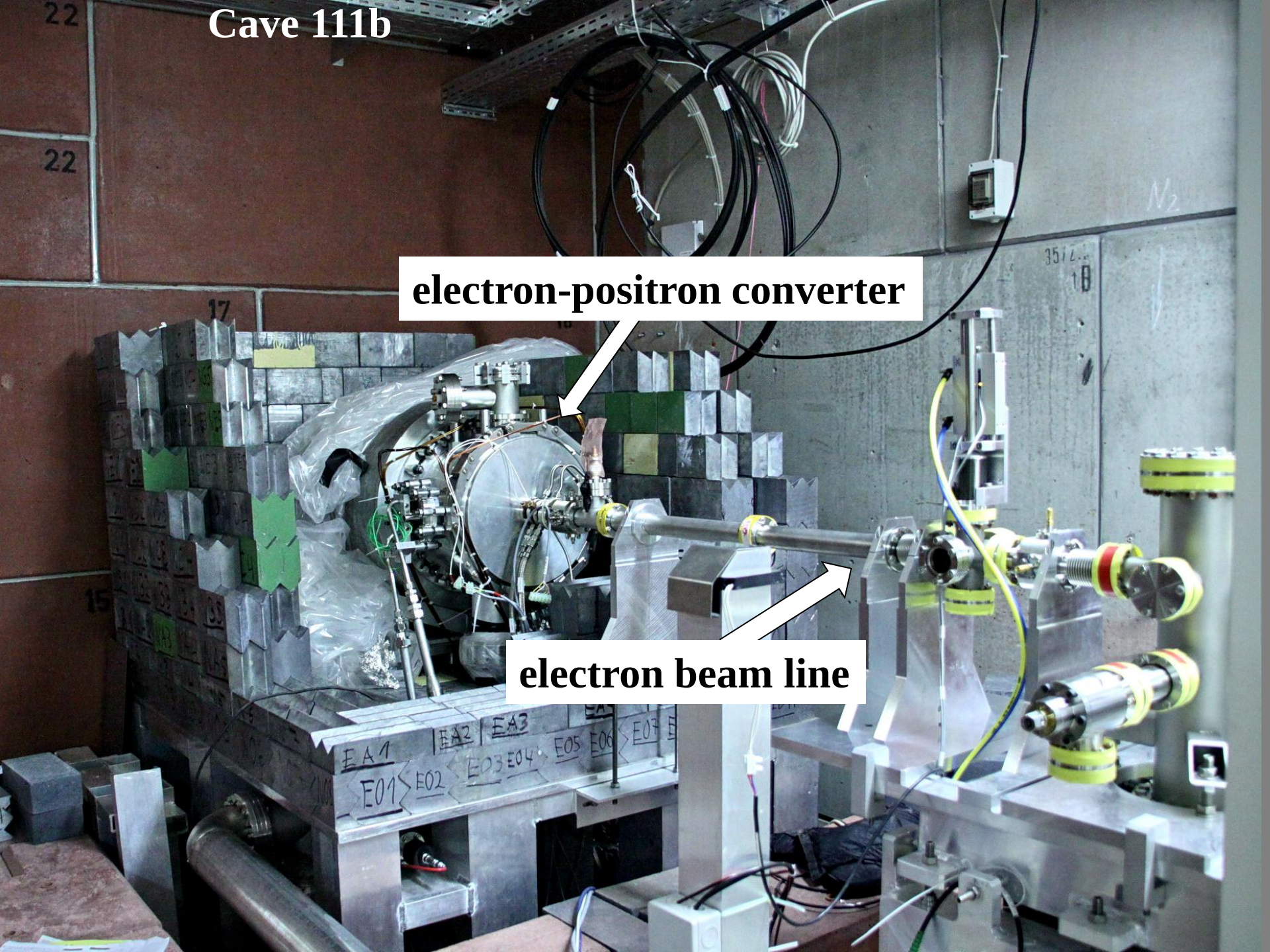
1: Diagnosestation, IR-Imaging und biologische IR Experimente
2: Femtosekundenlaser, THz-Spektroskopie, IR Pump-Probe Experimente
3: Zeitaufgelöste Halbleiter-Spektroskopie, THz-Spektroskopie
4: FTIR, biologische IR Experimente
5: Nahfeld und Pump-Probe IR Experimente
6: Radiochemie und Summenfrequenz-Erzeugung, photothermische Spektroskopie


Cave 111b
Positron laboratory 111c
concrete wall (3.2m)
22 Na source
accelerator
≈ 600 mm
≈ 250 mm
beam dump
water cooled Al block
W moderator foil
radiation converter
extraction lens
Be windows
ELBE primary beam
5 mm
e-
13 Mhz / 77 ns
e+
apertures
Cable tunnel
chopper
buncher
26 MHz
accelerators
Wien filter
Faraday cup
magnetic monopol lense
drift path
sample
multi-detector system

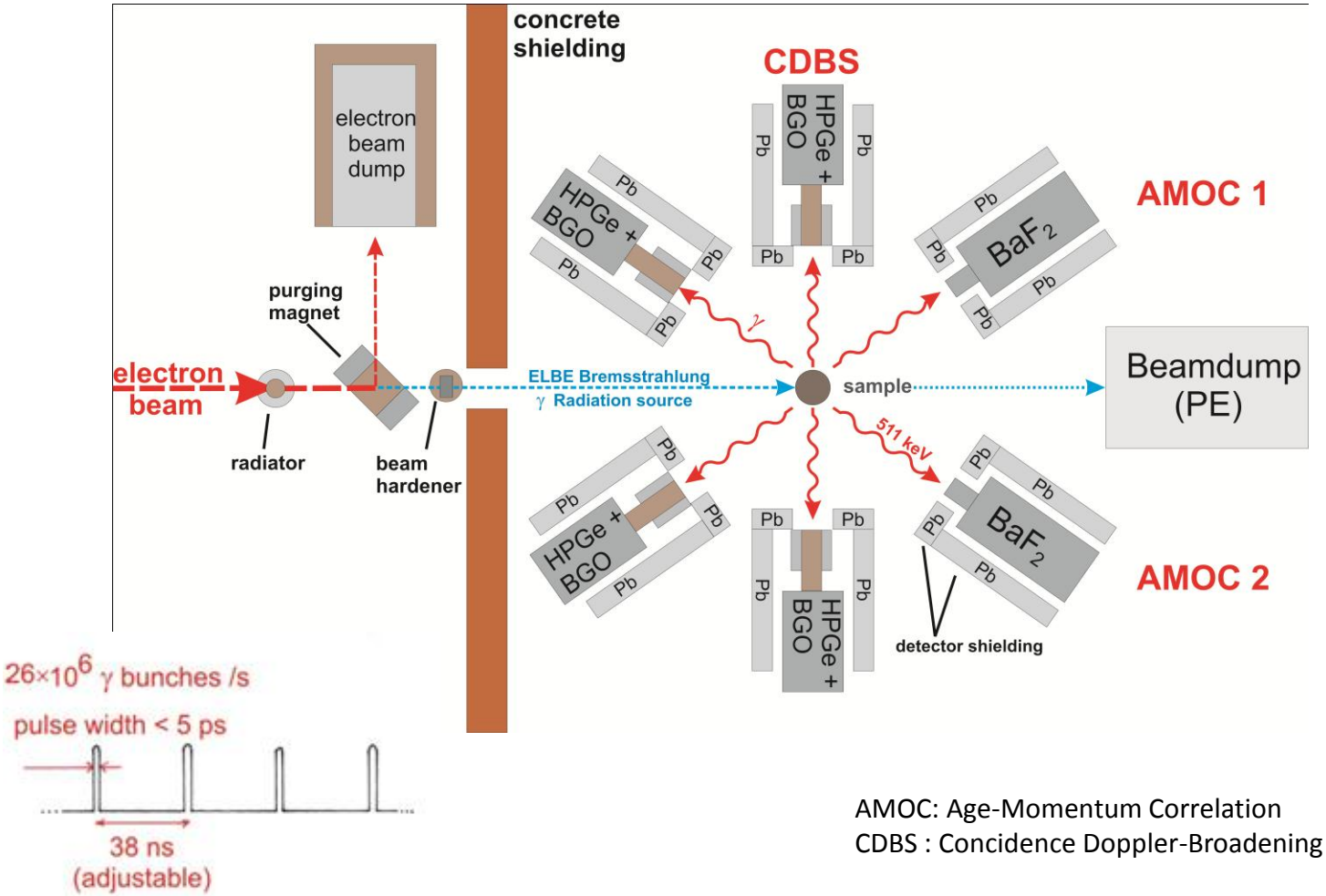

primary e-
pulse width
5 ps
77 ns
(adjustable)

EPOS scheme

Cave 111b

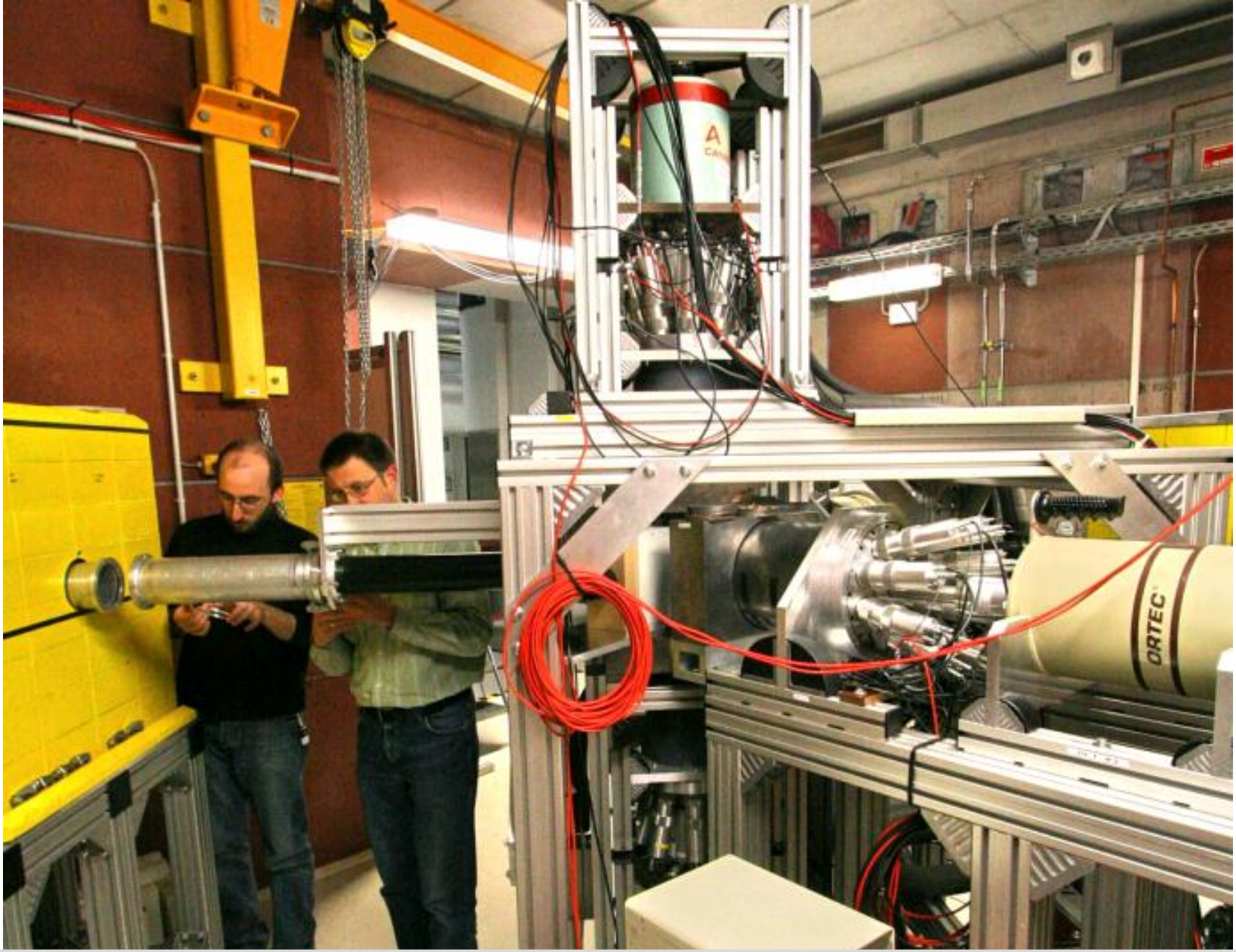


GiPS: Gamma-induced Positron Spectroscopy



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

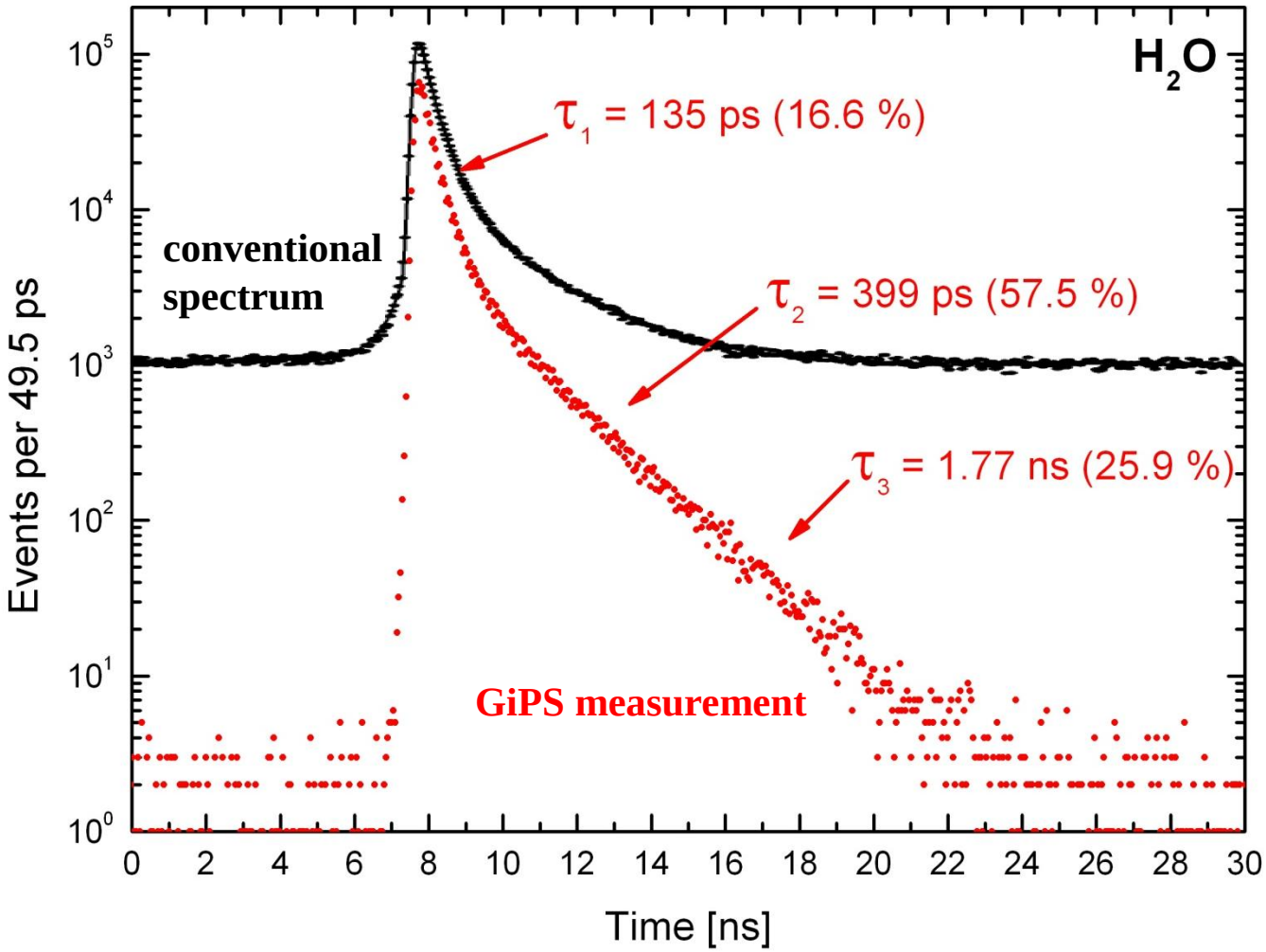
- 3 coincident setups were used: 2 AMOC and 1 CDBS spectrometer
- only coincident detection ensures high spectra quality



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

Example: Water at RT

- total count rate in spectrum: 12×10^6



- Black spectrum: conventional measurement by Kotera et al., Phys. Lett. A 345, (2005) 184

Applications of GiPS since begin of 2009

- neutron irradiated Fe-Cr alloys (highly activated up to 50 MBq ^{60}Co)
- Reactor pressure vessel steel samples from Greifswald nuclear power station
- Iron samples after mechanical damage (LCMTR-ISCSA-CNRS, Frankreich)
- set of Zircony alloys (Collaboration Mumbai/India)
- porous glass (Chem. Department/Univ. Leipzig)
- biological samples
- liquids

Positron Annihilation Spectroscopy: Applications

Variety of applications in all fields of materials science:

- defect-depth profiles due to surface modifications (ion implantation; tribology)
- soft matter physics (open volume; interdiffusion; …)
- porosimetry (e.g. low-k materials - highly porous dielectric layers)
- bulk defects in semiconductors, ceramics and metals
- epitaxial layers (growth defects, misfit defects at interface, …)
- fast kinetics (e.g. precipitation processes in Al alloys; defect annealing; diffusion; …)
- radiation resistance (e.g. space materials)
- many more …

This presentation can be found as pdf-file on our Website:
http://positron.physik.uni-halle.de