Positron Annihilation Spectroscopy for Non-Destructive Testing

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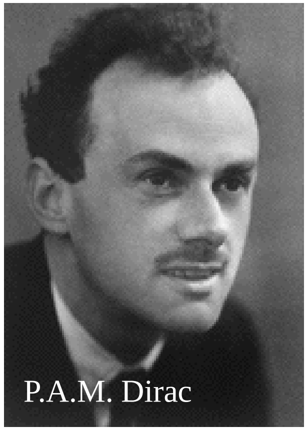
- Historical remarks about positrons
- Defect detection by positrons
- Non-destructive testing in Steel

13. October 2010

SKF

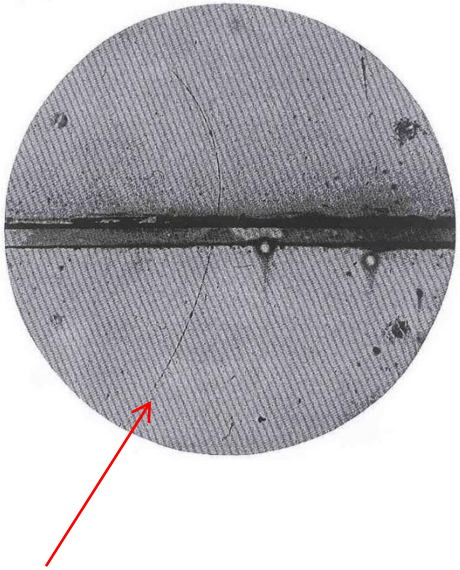
Discovery of the Positron

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


P.A.M. Dirac


C.D. Anderson

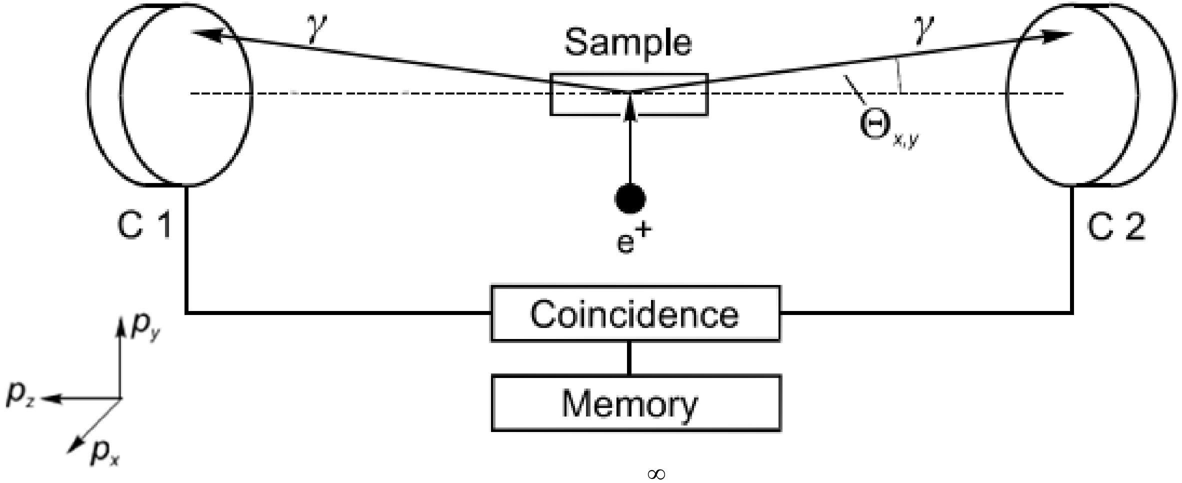
- Positronium as bound state of e^- and e^+ lightest atom was predicted (1934) and discovered (1951)
- Annihilation in matter was studied beginning in the 40th
- Positrons can be obtained by
  - pair production from gamma radiation ($E_\gamma > 1022$ keV)
  - β^+ decay from isotopes (mostly ^{22}Na)



- 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

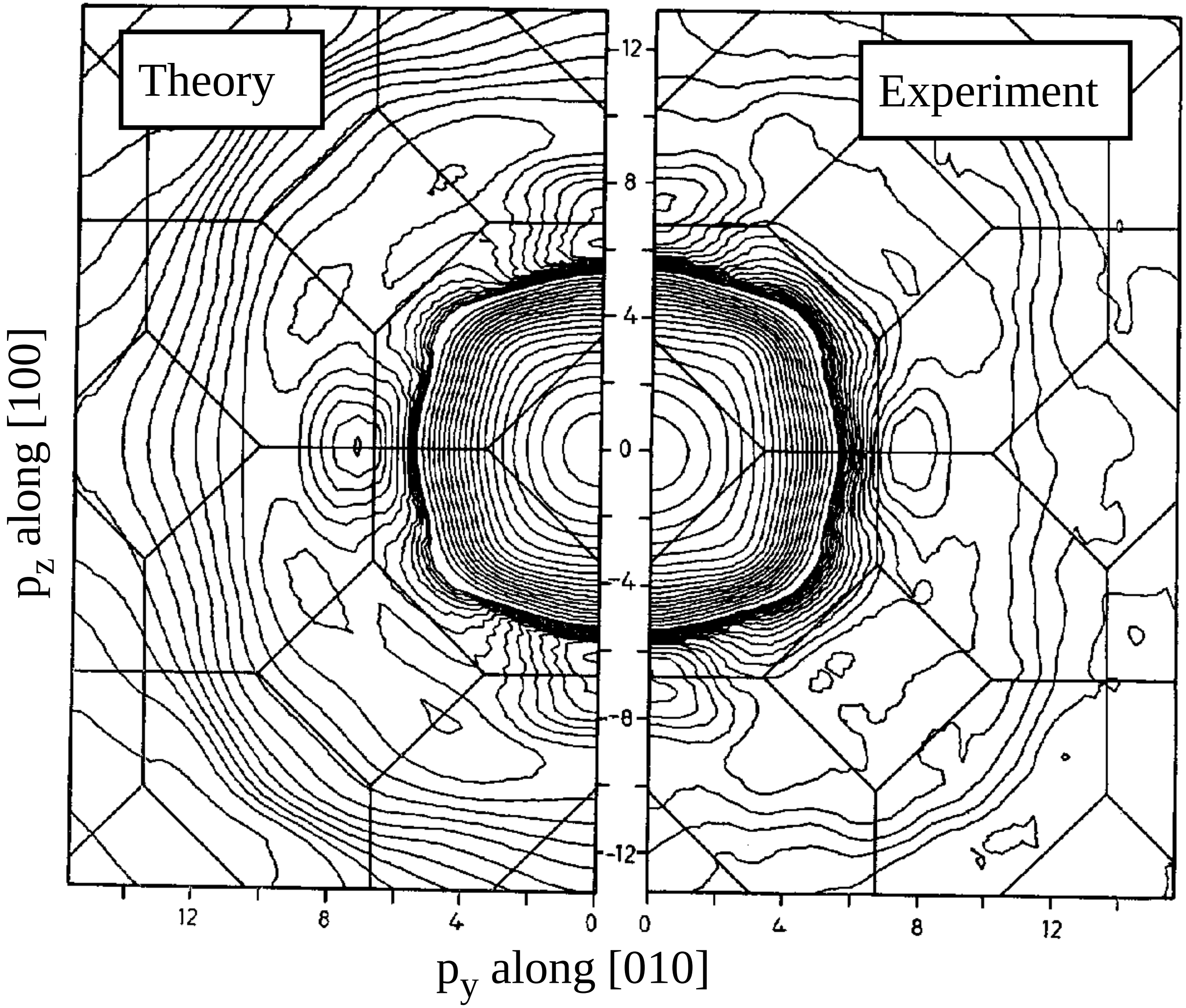
- during annihilation: conservation laws must be fulfilled (energy, momentum)
- electron momentum distribution can directly be measured
- 2D-ACAR: two-dimensional (position-sensitive) detectors
- measurement of single crystals in different directions:
- reconstruction of Fermi surface possible

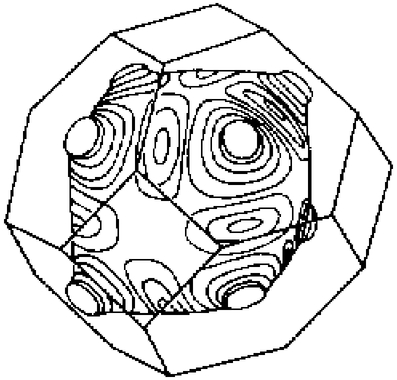


2D – ACAR (Angular Correlation of Annihilation Radiation)

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

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

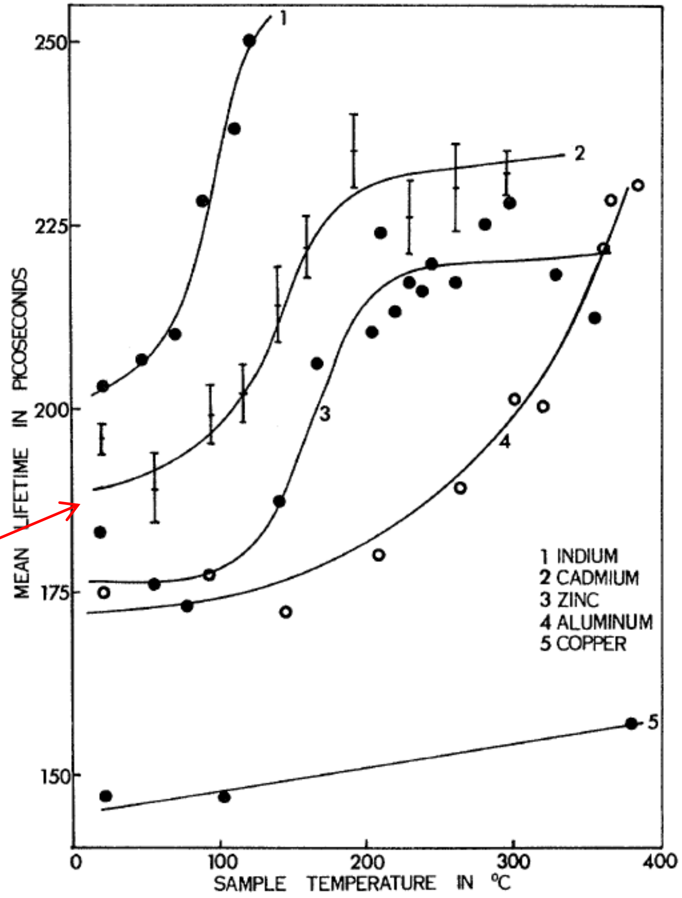
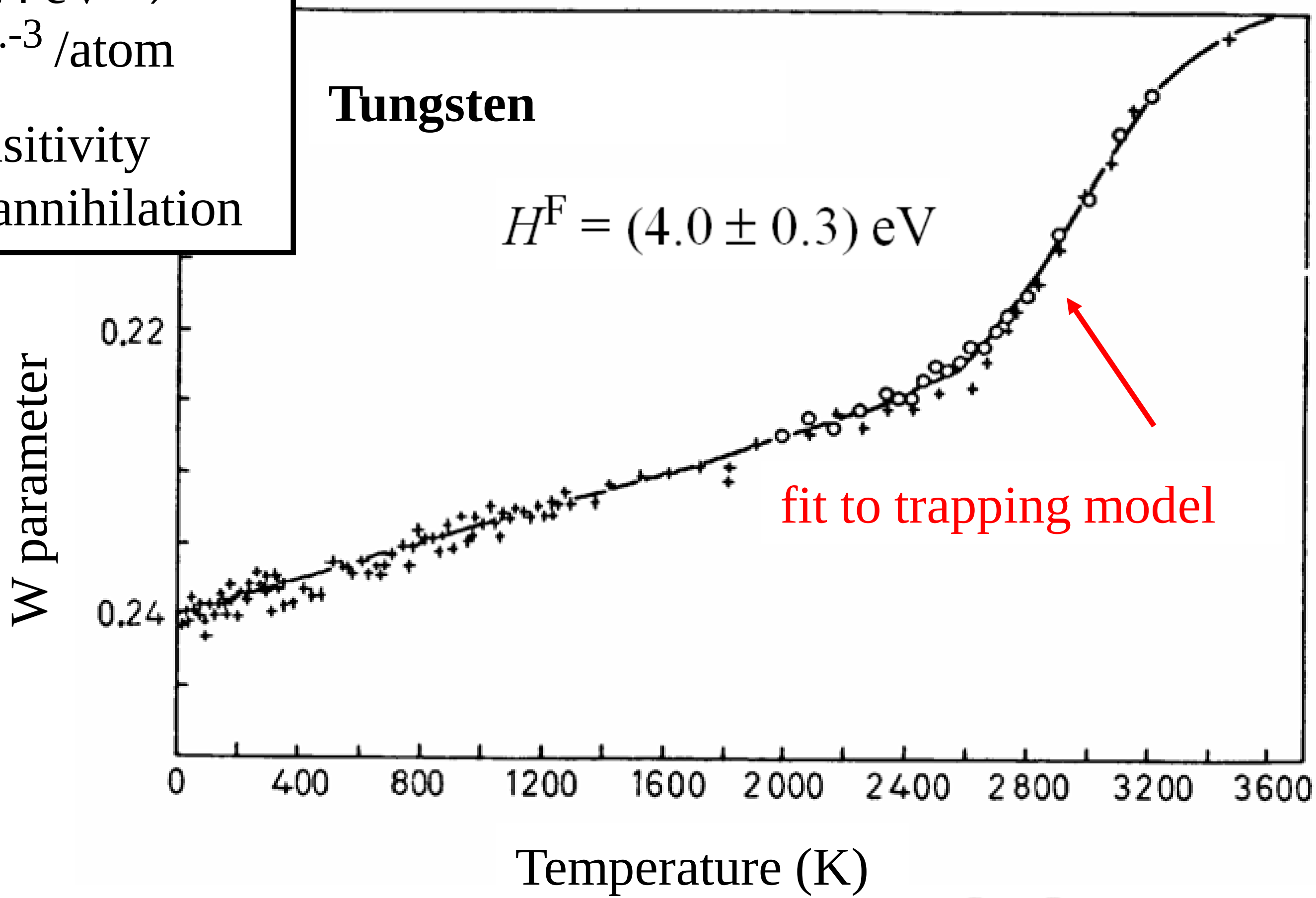


FIG. 1. Positron mean lifetimes in several metals as a function of temperature.

Vacancies in thermal Equilibrium

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

$$C_{1\mathrm{v}}(T) = \exp\left(\frac{S^{\mathrm{F}}_{1\mathrm{v}}}{k}\right)\exp\left(\frac{H^{\mathrm{F}}_{1\mathrm{v}}}{kT}\right)$$



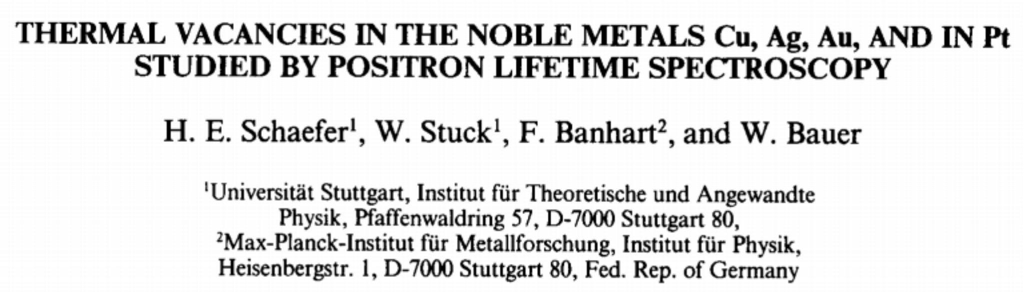
Tungsten

$H^{\mathrm{F}} = (4.0 \pm 0.3)$ eV

fit to trapping model

(Ziegler, 1979)

Determination of Vacancy Formation Enthalphy



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

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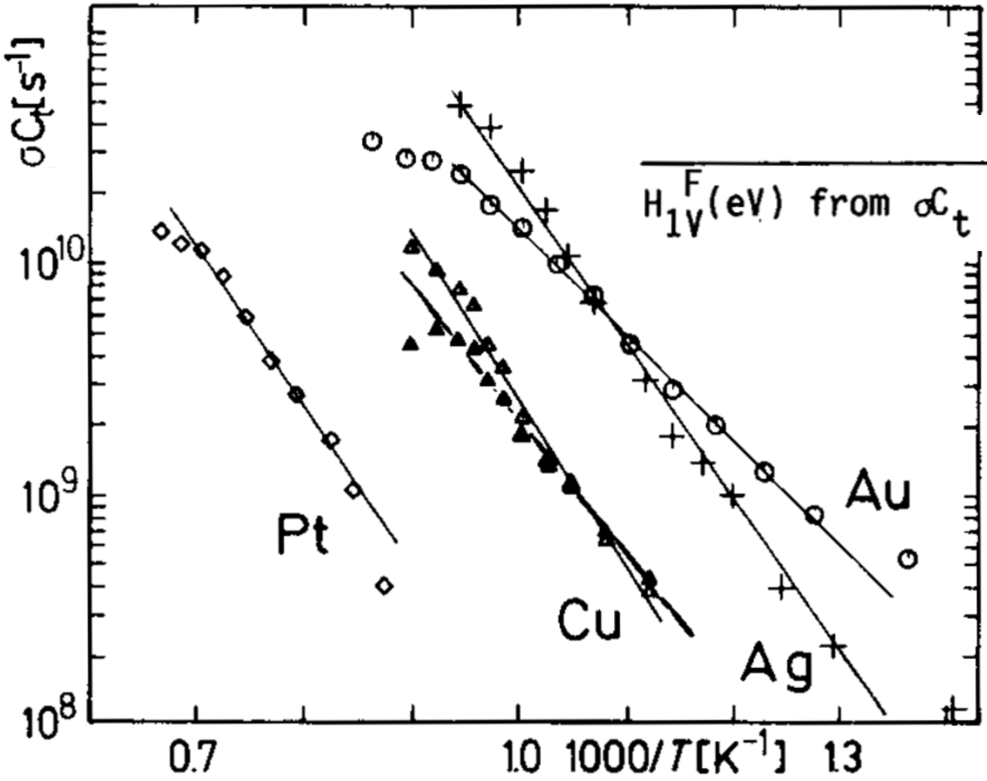
Heisenbergstr. 1, D-7000 Stuttgart 80, Fed. Rep. of Germany



	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

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Positron lifetime spectra

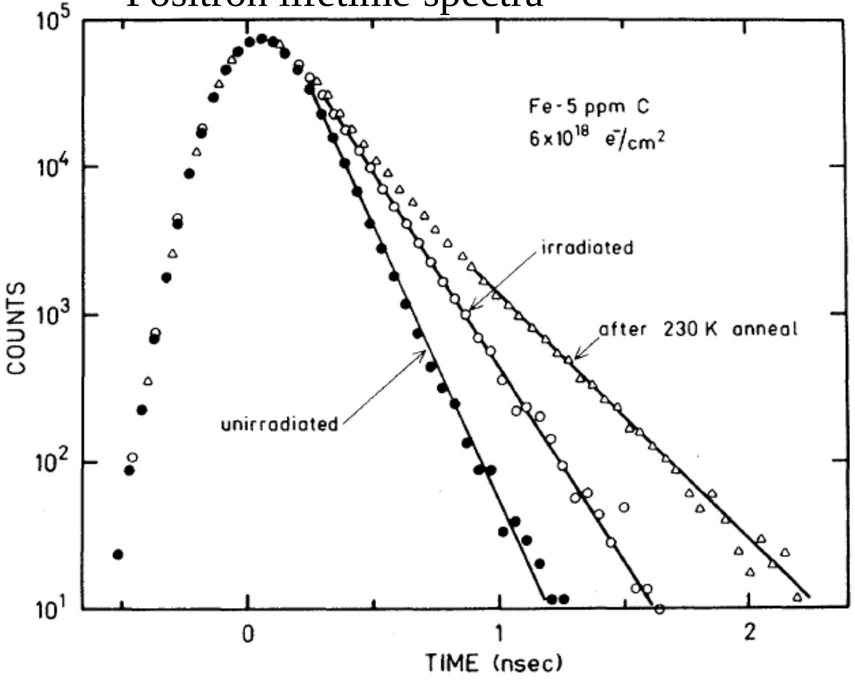
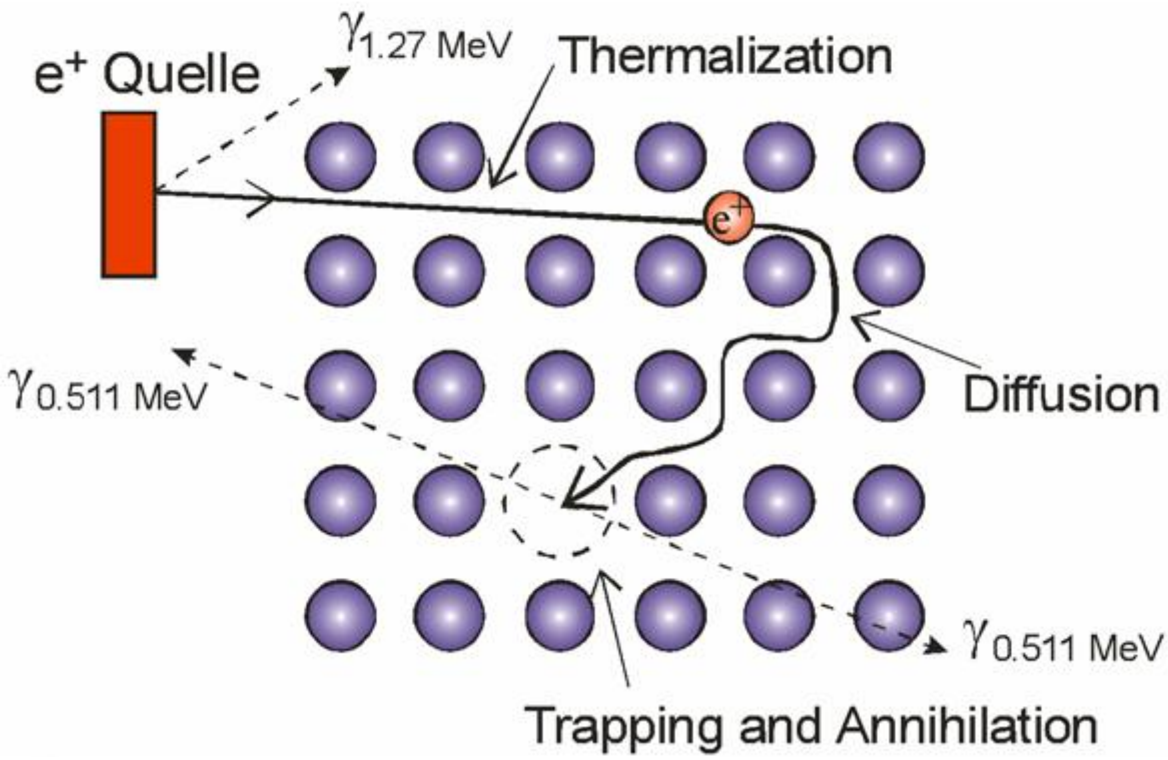


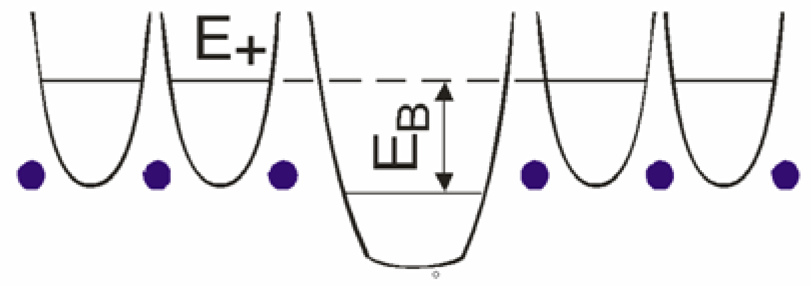
FIG. 1. Positron-lifetime spectra after source-background subtraction in electron-irradiated (6×10^{18} e^-/cm^2) high-purity iron at various stages of isochronal annealing. The dramatic occurrence of a long-lifetime component after 230 K annealing is clearly visible.

- 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

Defect Sensitivity

atomic open-volume defects

- vacancies ($\rho_v > 10^{-7}$)
- vacancy clusters (n=1…50)
- dislocations (> 10^8 cm^{-2})
- grain boundaries (only ultra-fine grained materials)
- surface

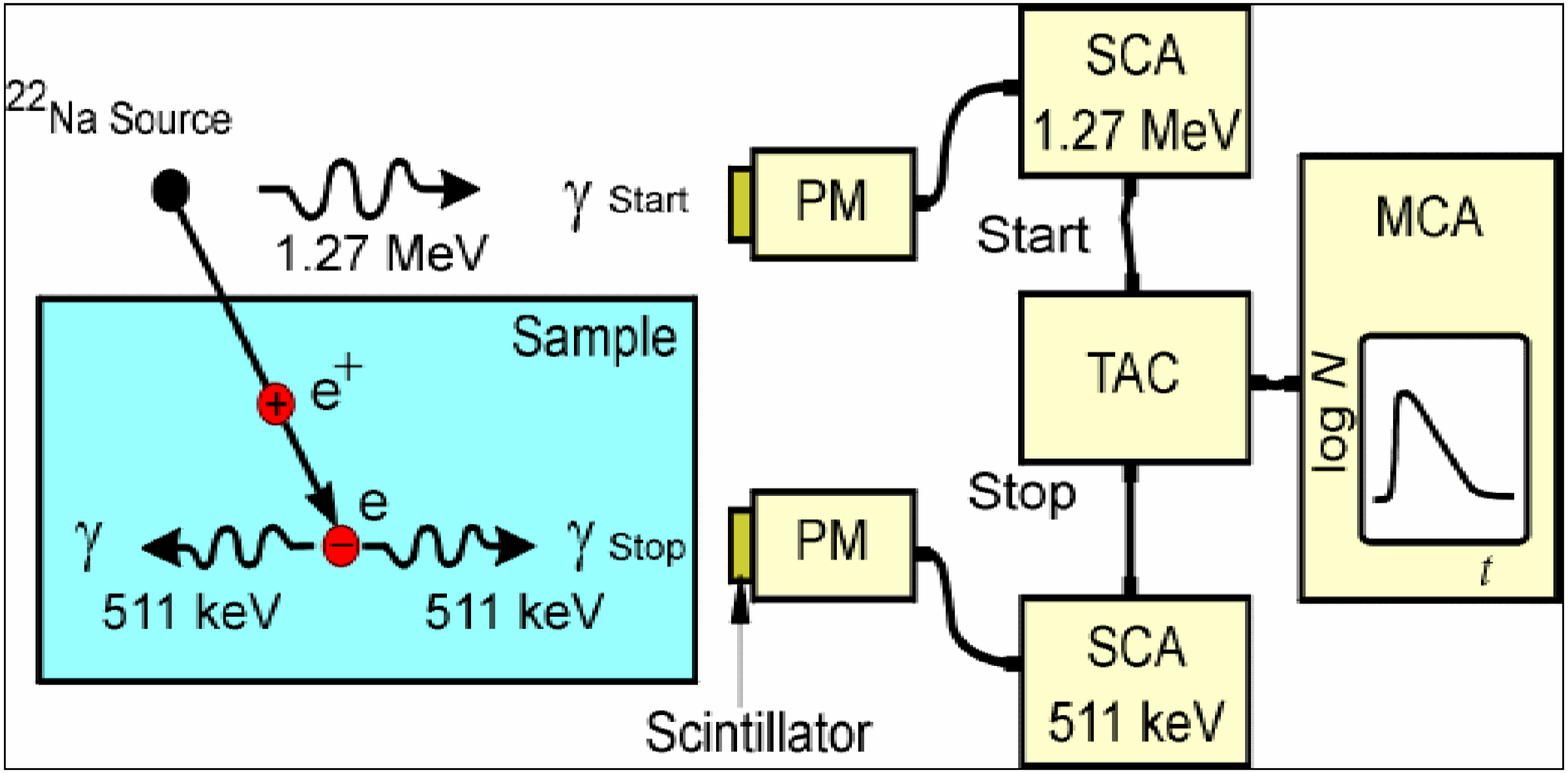
non-open volume defects

- coherent precipitates (e.g. GPZ in Al-Zn)
- negatively charged acceptors in semiconductors ("shallow traps")

large open volume 1…50 nm (Positronium formation)

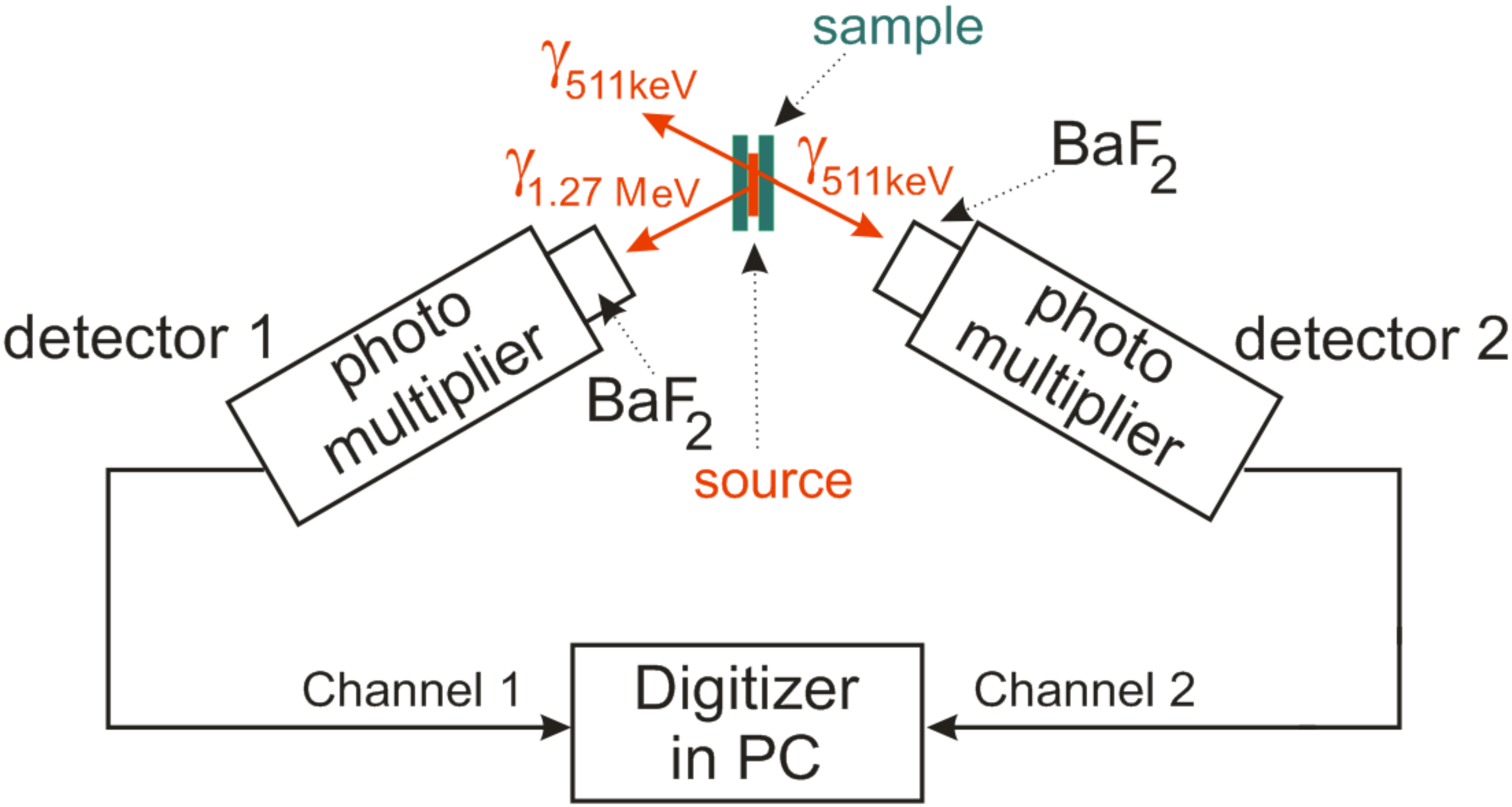
- open volume between molecular chains in polymers (> 100 Å^3)
- mesoporous dielectrica (1 nm < d_{pore} < 50 nm)

Positron lifetime spectroscopy



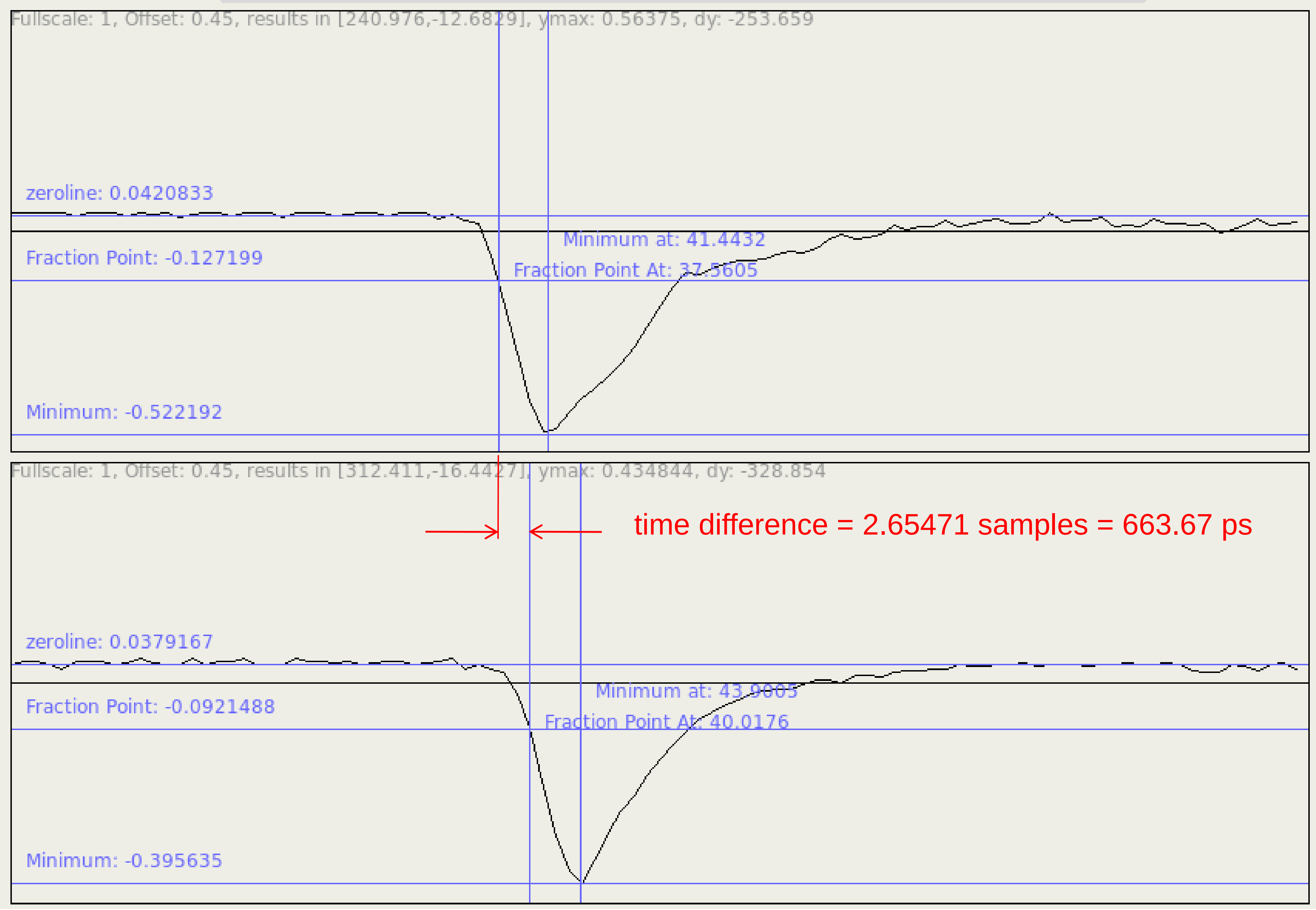
Positron lifetime: time difference between 1.27 MeV and 0.511 MeV quanta

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



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

zeroline: 0.0420833

Fraction Point: -0.127199

Minimum at: 41.4432

Fraction Point At: 37.5605

Minimum: -0.522192

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

time difference = 2.65471 samples = 663.67 ps

zeroline: 0.0379167

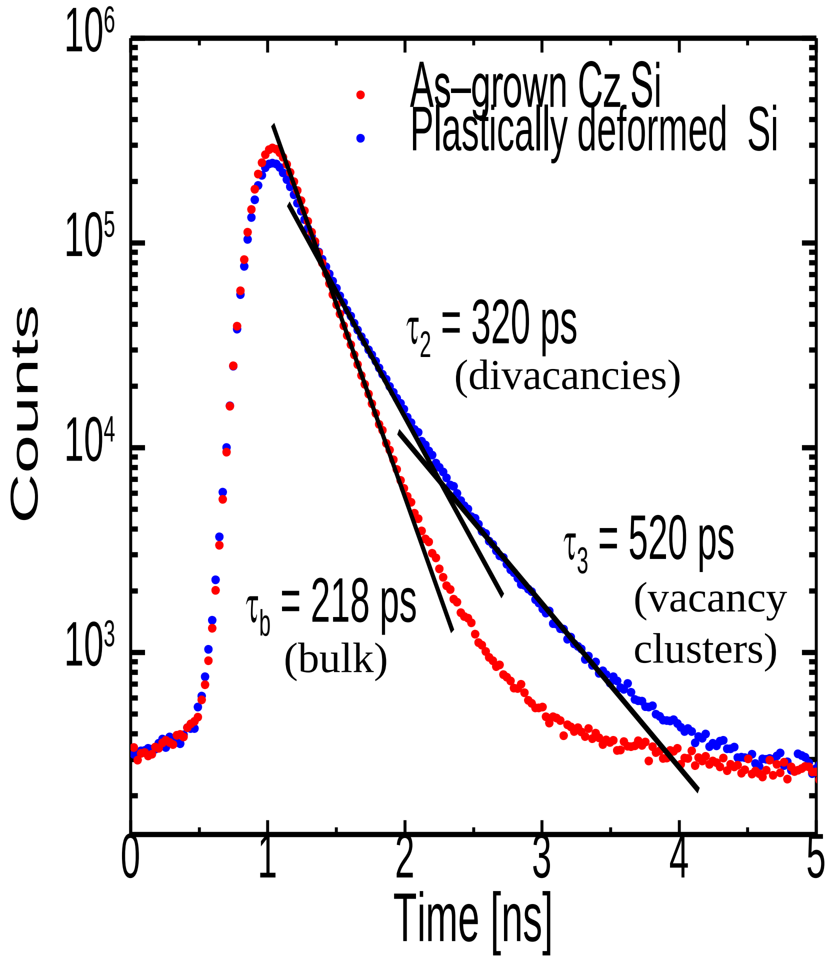
Fraction Point: -0.0921488

Minimum at: 43.9005

Fraction Point At: 40.0176

Minimum: -0.395635

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:

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

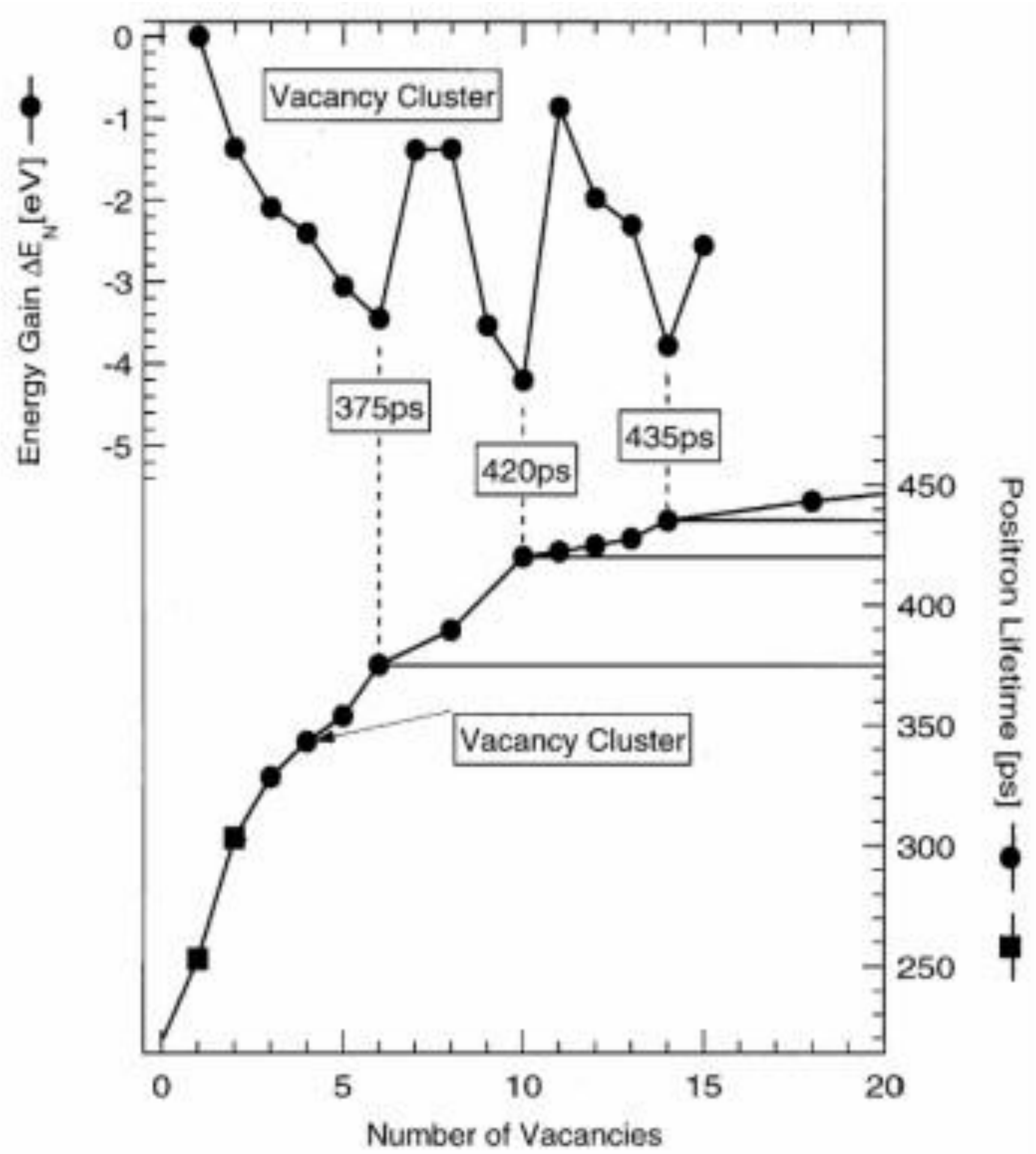
trapping coefficient

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

trapping rate

defect concentration

Theoretical calculation of vacancy clusters in Si

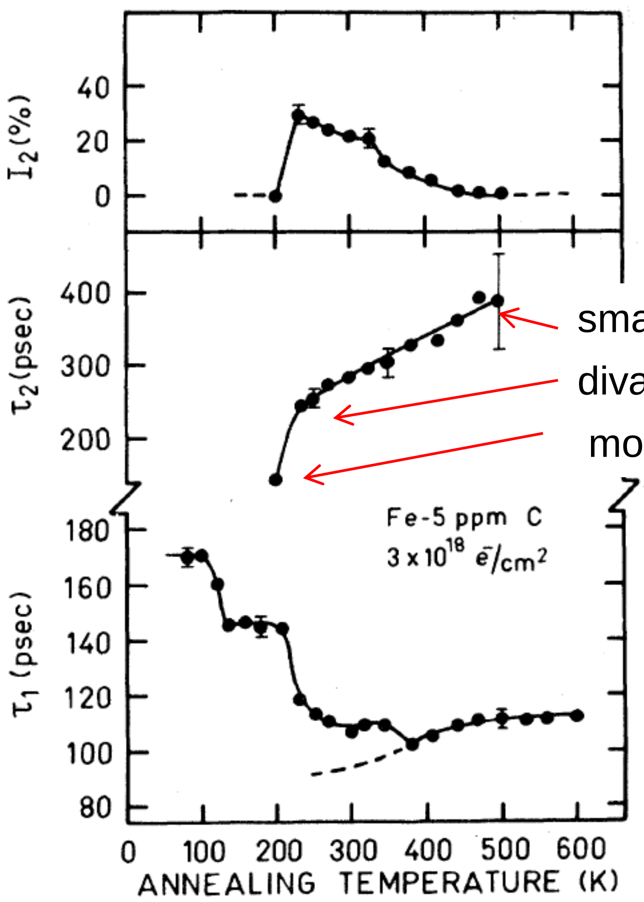


- there are cluster configurations with a large energy gain
- „Magic Numbers" with 6, 10 und 14 vacancies
- positron lifetime increases distinctly with cluster size
- for n > 10 saturation effect, i.e. size cannot be determined

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

Vacancy clustering during defect annealing

- electron irradiated 5N-Fe
- clustering in early stage can be observed
- very sensitive: formation of divacancies and small clusters (n < 10)



small vacancy clusters

divacancies

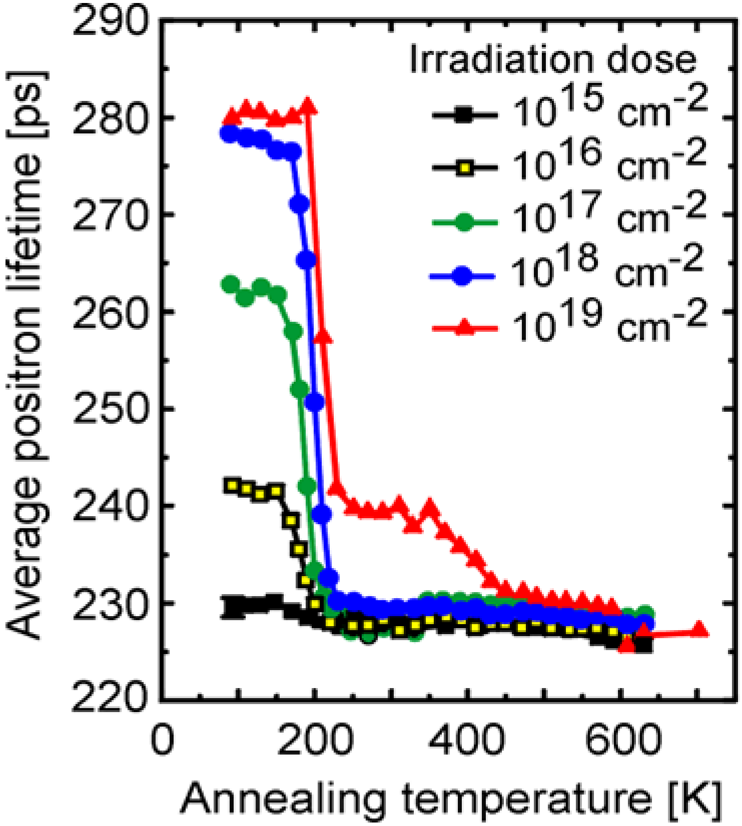
monovacancies

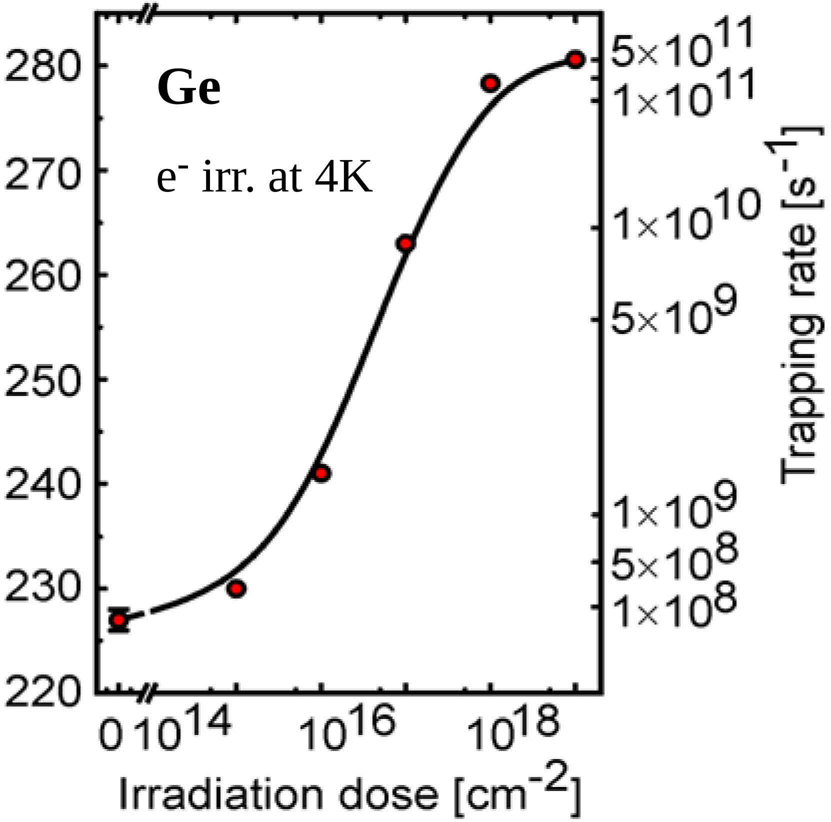
FIG. 2. Positron-lifetime parameters as a function of the isochronal annealing temperature in the low-dose electron-irradiated pure iron.

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

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)

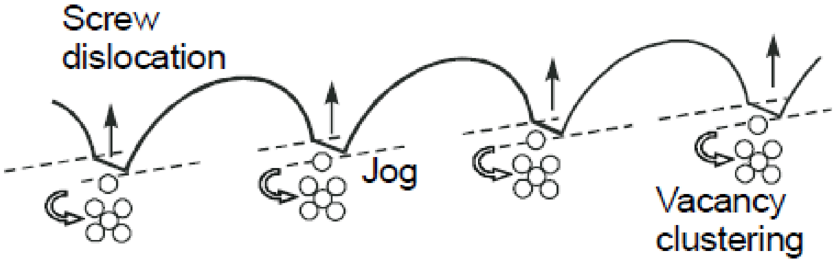


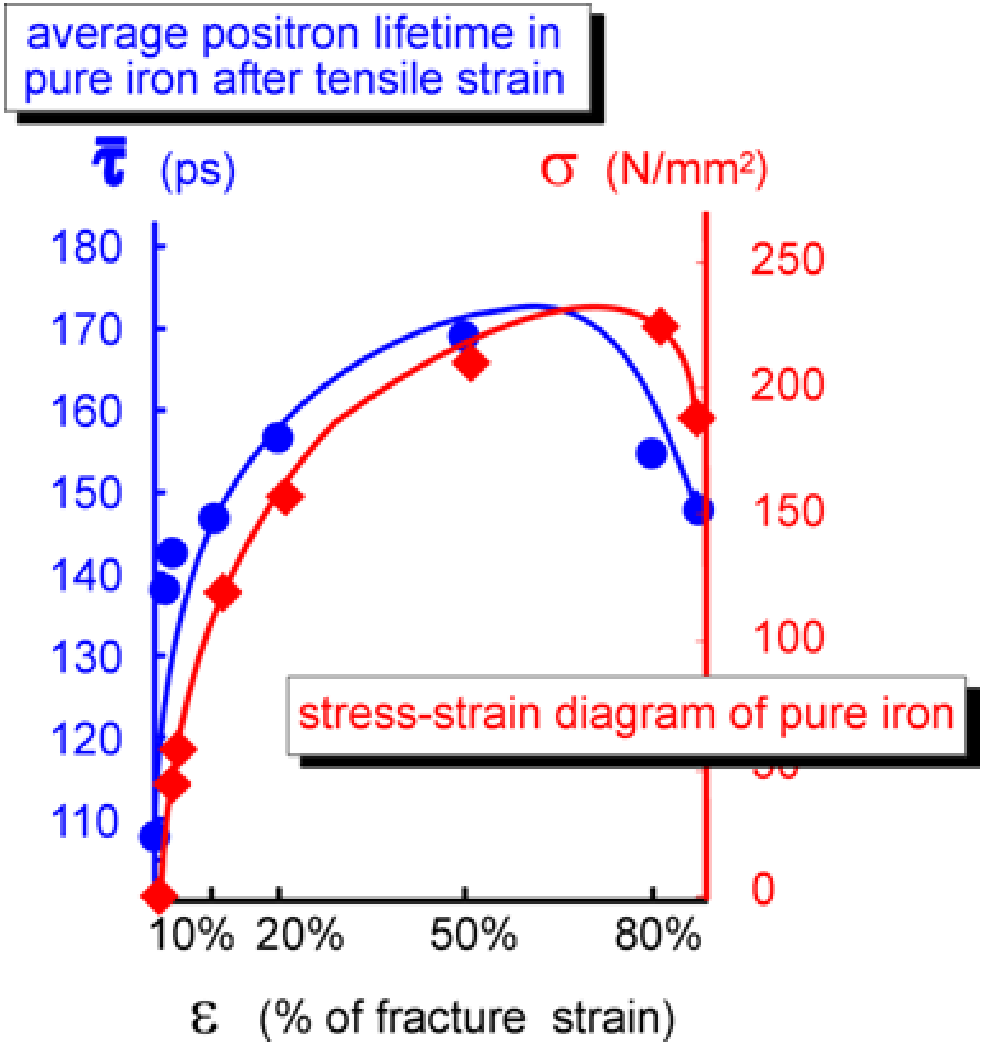


(Polity et al., 1997)

Defects in Iron after tensile Strength in Stress-Strain Experiment

- extensive study of defects in mechanically damaged iron and steel
- sensitive: detection of defects already in the elastic Hooke's range
- Vacancy cluster and dislocations are detectable in both cases
- small vacancy clusters are generated by jog dragging process

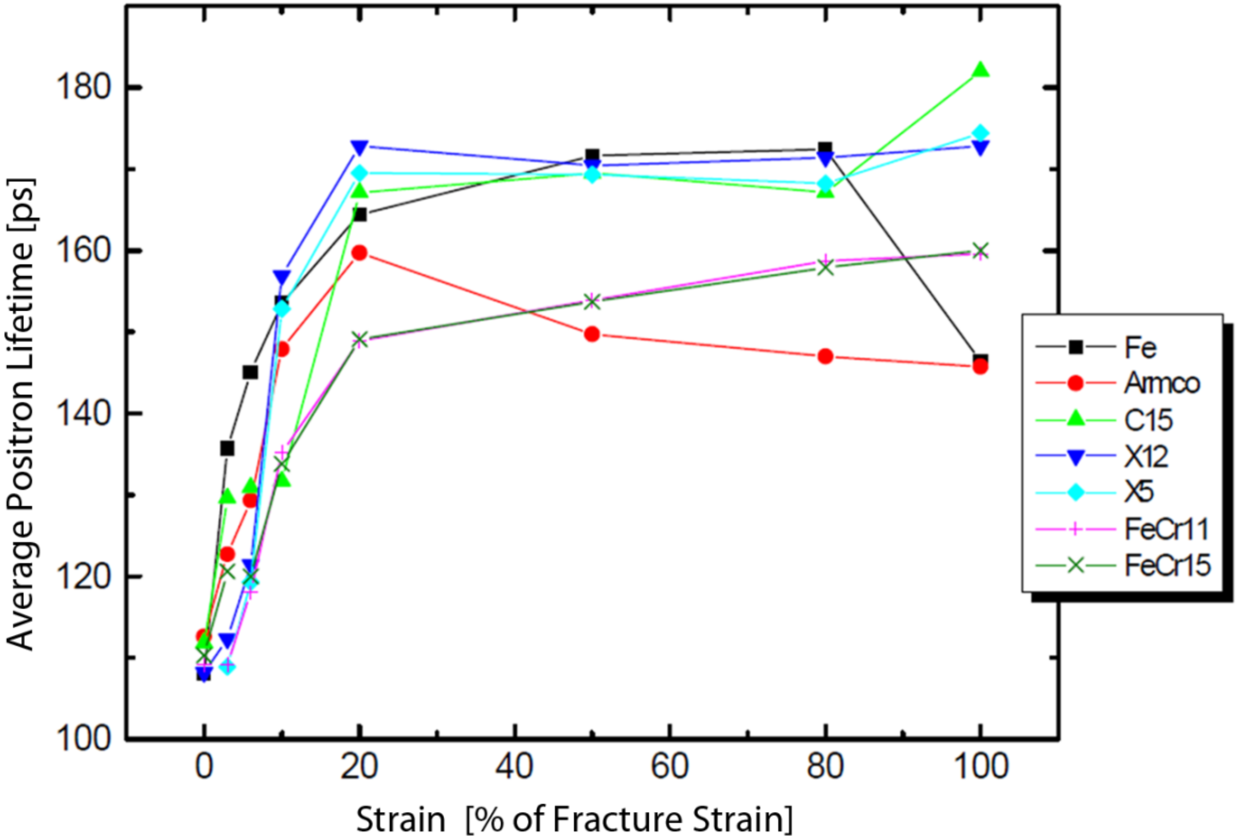




Somieski et al., J. Physique IV 5, C1/127-134 (1995)

Comparison of Iron and different Steels

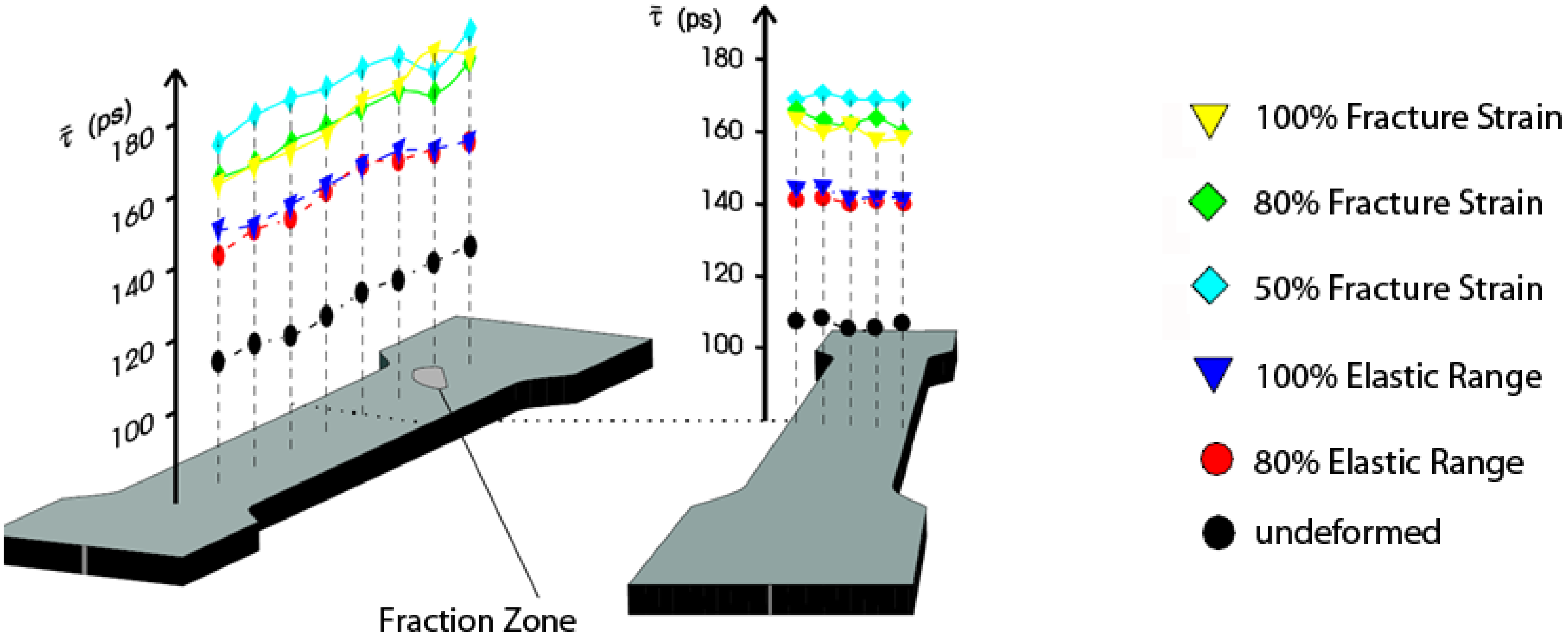
- samples were annealed before the stress-strain experiments
- strain normalized to fracture strain
- strongest effect for small strain



B. Somieski Dissertation 1996

Laterally resolved measurement across Test sample

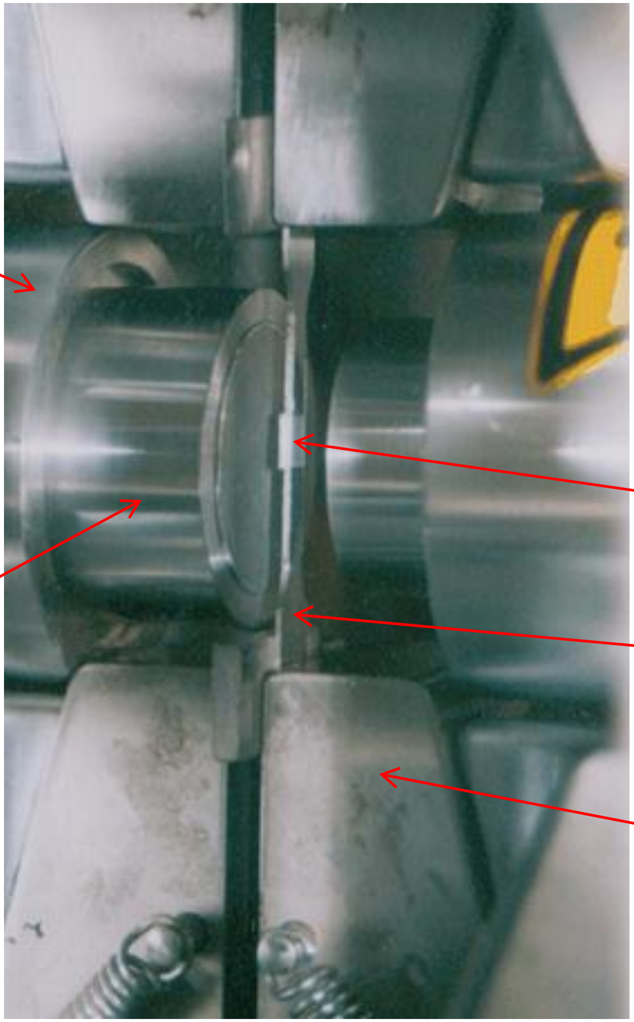
- Pure Fe sample
- strong damage already for strain < 100% of Hooke’s range -> technically most interesting range
- fraction zone can be predicted from positron measurements



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

Measurements live at testing machine

- measurements were partly performed during stress-strain and fatigue experiments directly at testing machine



PMT tube

Scintillator

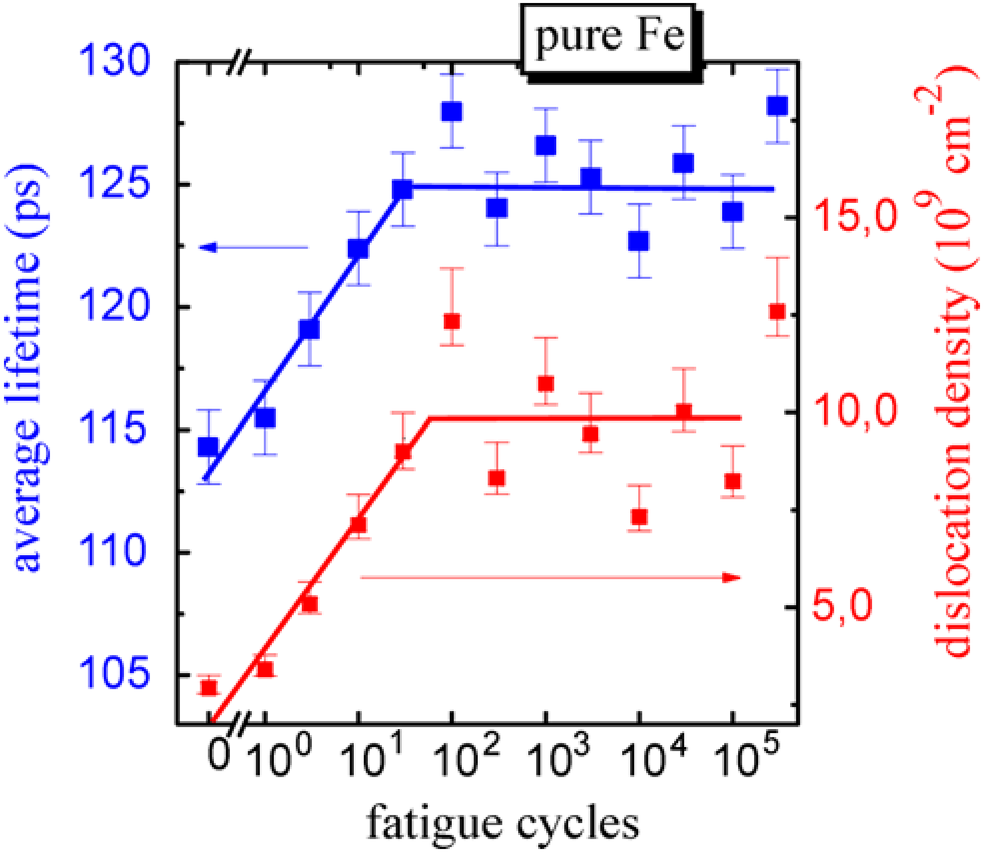
Source

Sample

Clamping jaws
of testing machine

Fatigue experiments in Fe and steels

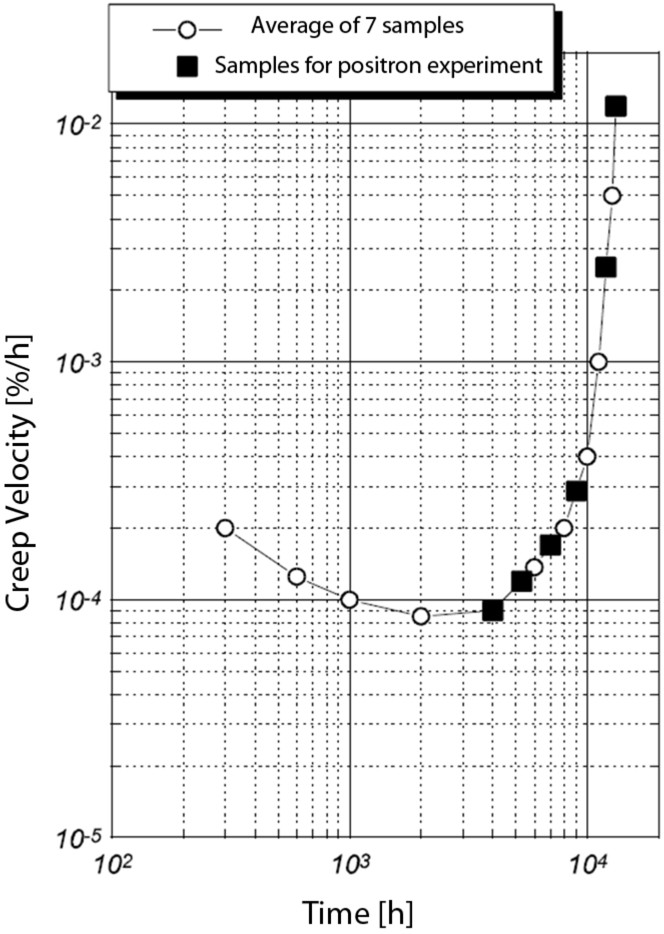
- measurement during fatigue experiment: up to 10^7 fatigue cycles in 11 days
- example: pure iron; σ_{max}= 94 MPa (80% of Hooke's range)
- sample remains in state of endurance (no fracture) -> and lifetime stays constant

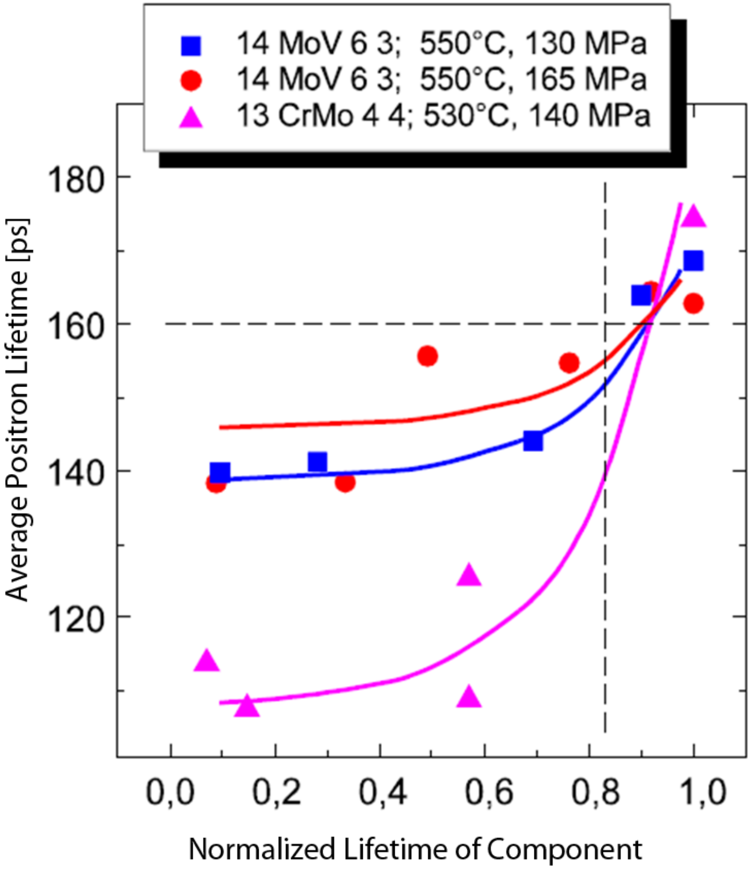


Somieski et al., J. Physique IV 5, C1/127-134 (1995)

High-temperature Creep Experiments

- set of samples was available for positron measurements (from IzfP Saarbrücken)
- component lifetime prediction seems to be possible

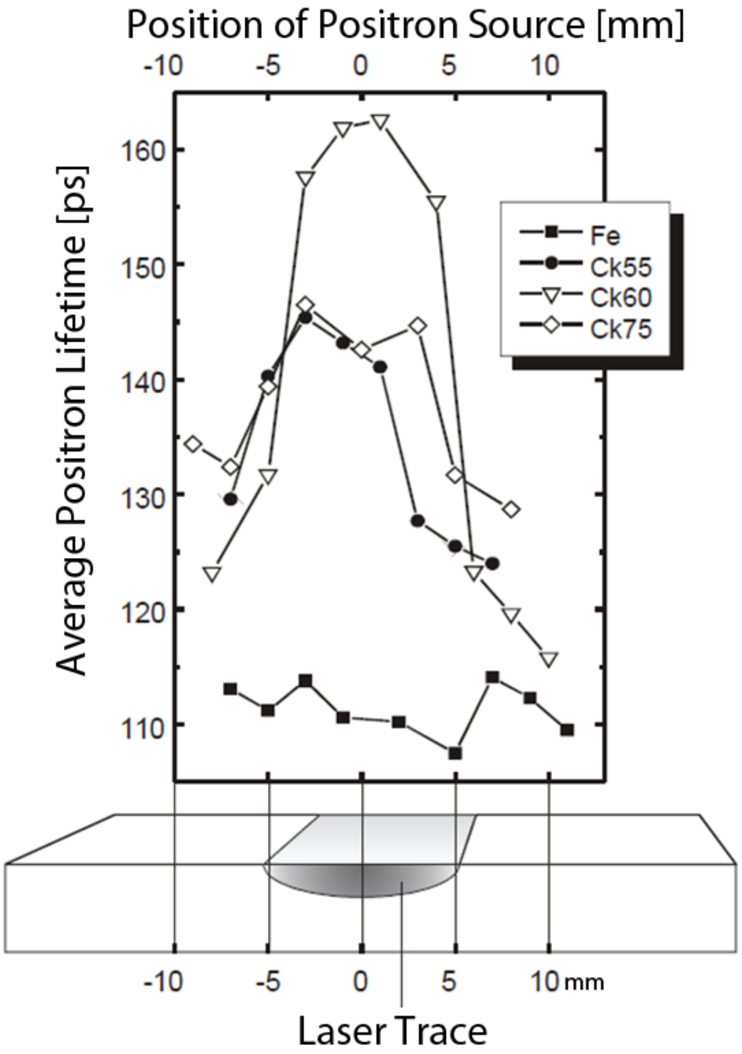




B. Somieski, Dissertation 1996

Defect detection after Surface Laser Hardening

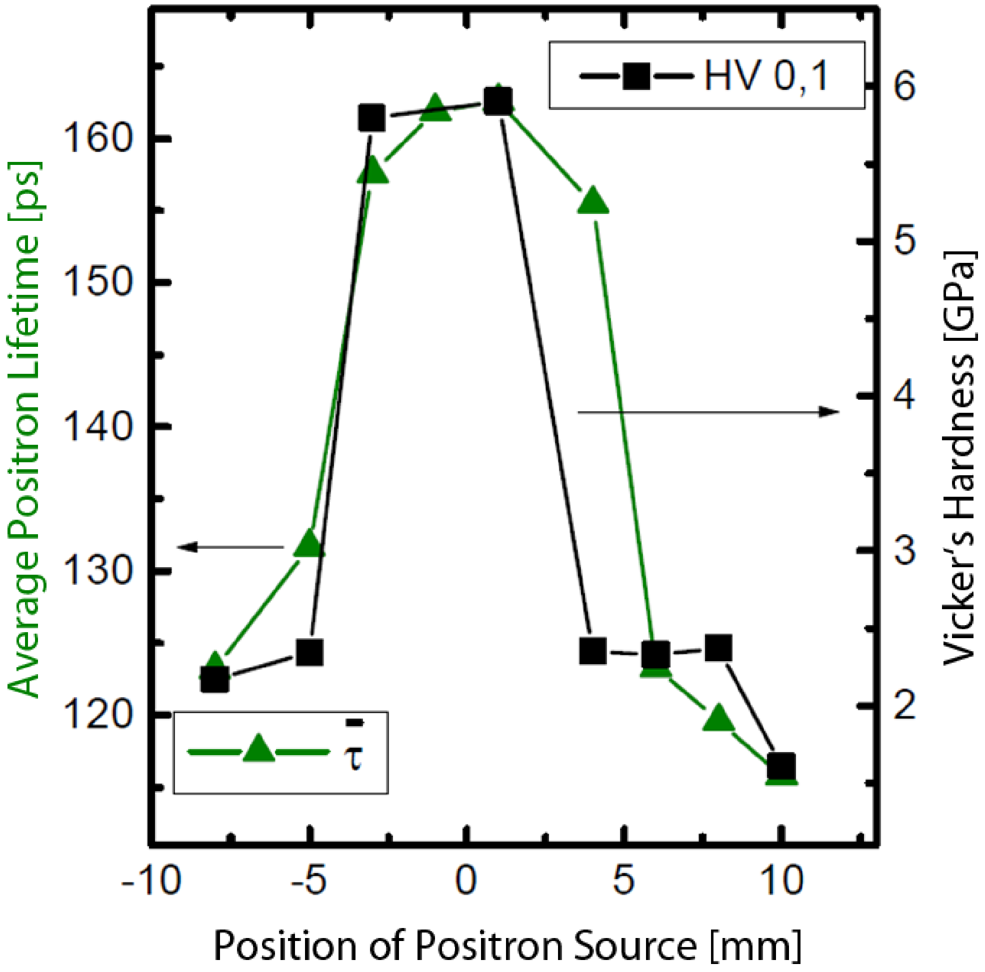
- Fe and different steels were treated by a 1.3 kW Laser in vacuum
- Laser spot 1 cm; velocity of Laser spot 0.3…1 m/min
- energy density: 10…30 MJ/m^2
- strong increase of positron lifetime in all steels due to defect generation during fast cooling
- Ck… Fe-C steel without any further alloying element
- Ck60 means Fe$_{99.4}$-C$_{0.6}$
- interesting: Fe does not show any defect generation - obviously no generation of dislocations



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

Comparison of Positron Lifetime with Vicker's Hardness

- measurement of Vickers's hardness and positron lifetime across a laser trace at Ck60 steel
- generation of dislocations leads to increase of hardness (desired effect)
- positron lifetime increases strongly
- positrons detect dislocations and small vacancy clusters



Somieski et al., Mat. Sci. Forum **255-257**, pp584 (1997)

Non-destructive testing outside the lab

- problem: only one "sample" instead of "sandwich geometry"
- positron source is covered by standard back-sample (e.g. pure iron or Si)
- this part of spectrum must be subtracted before data evaluation
- is standard procedure when only one sample exist

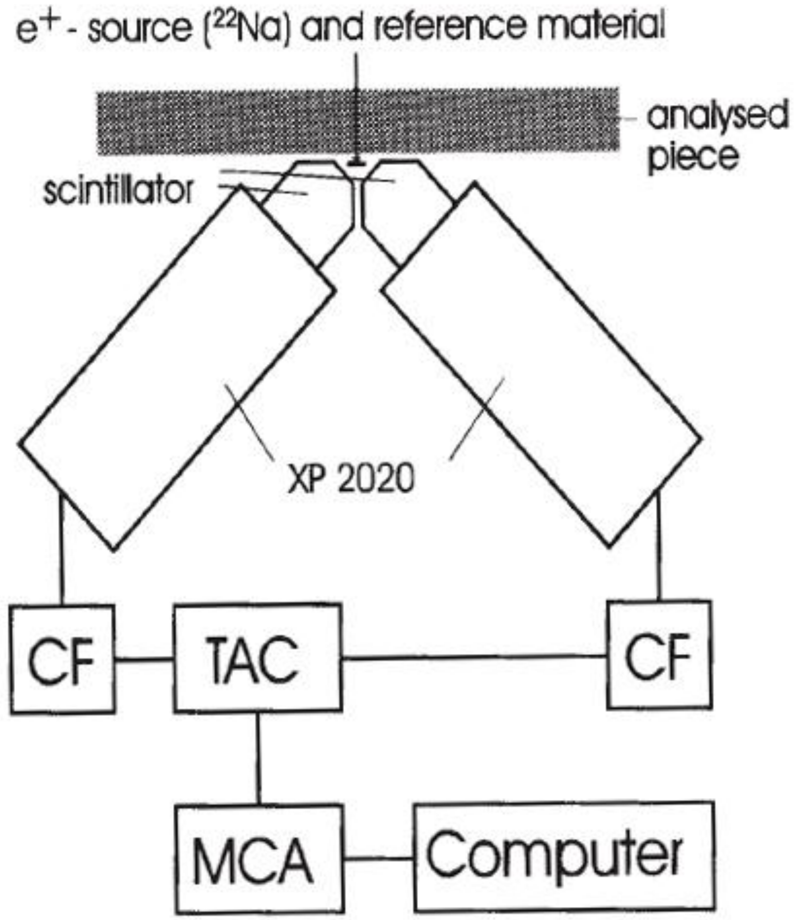
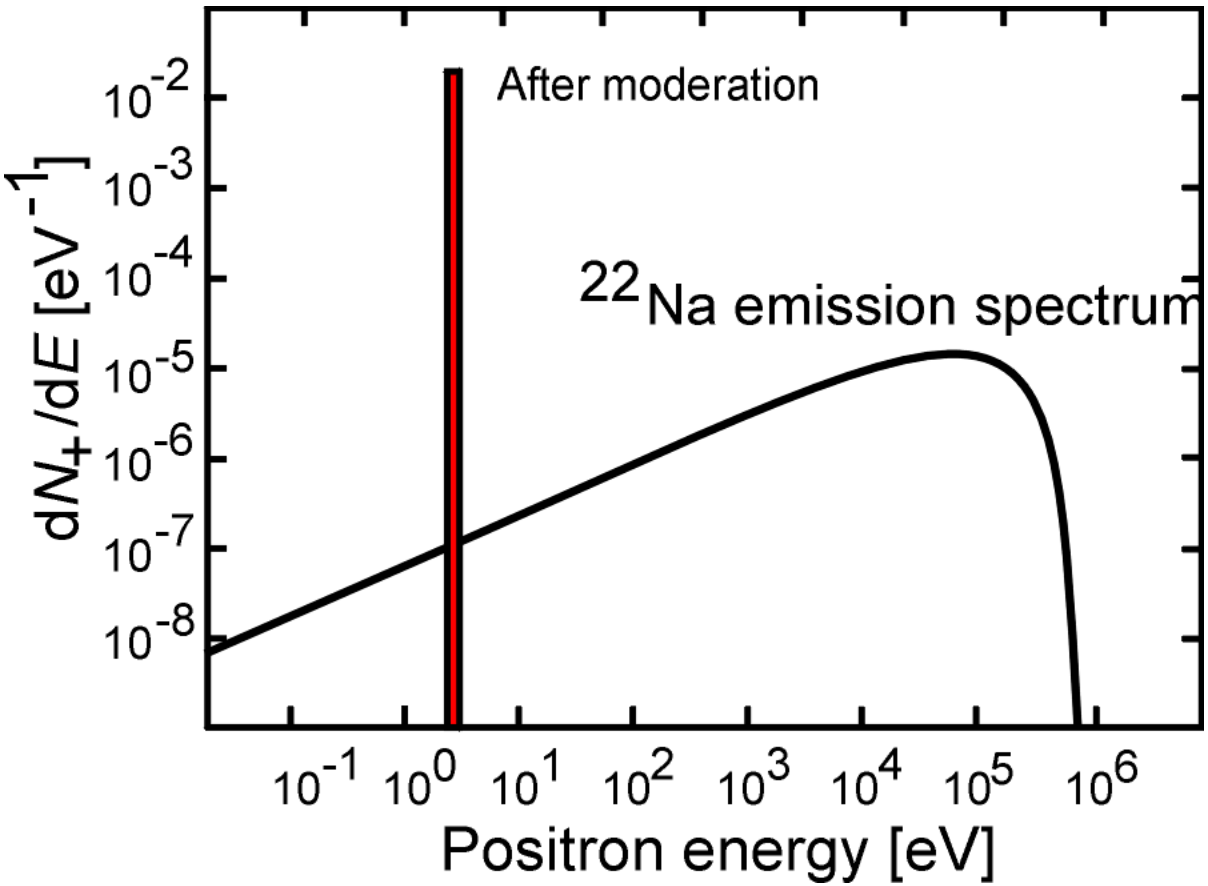


Figure 1 Modified positron lifetime spectrometer for in-the-field mapping.

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

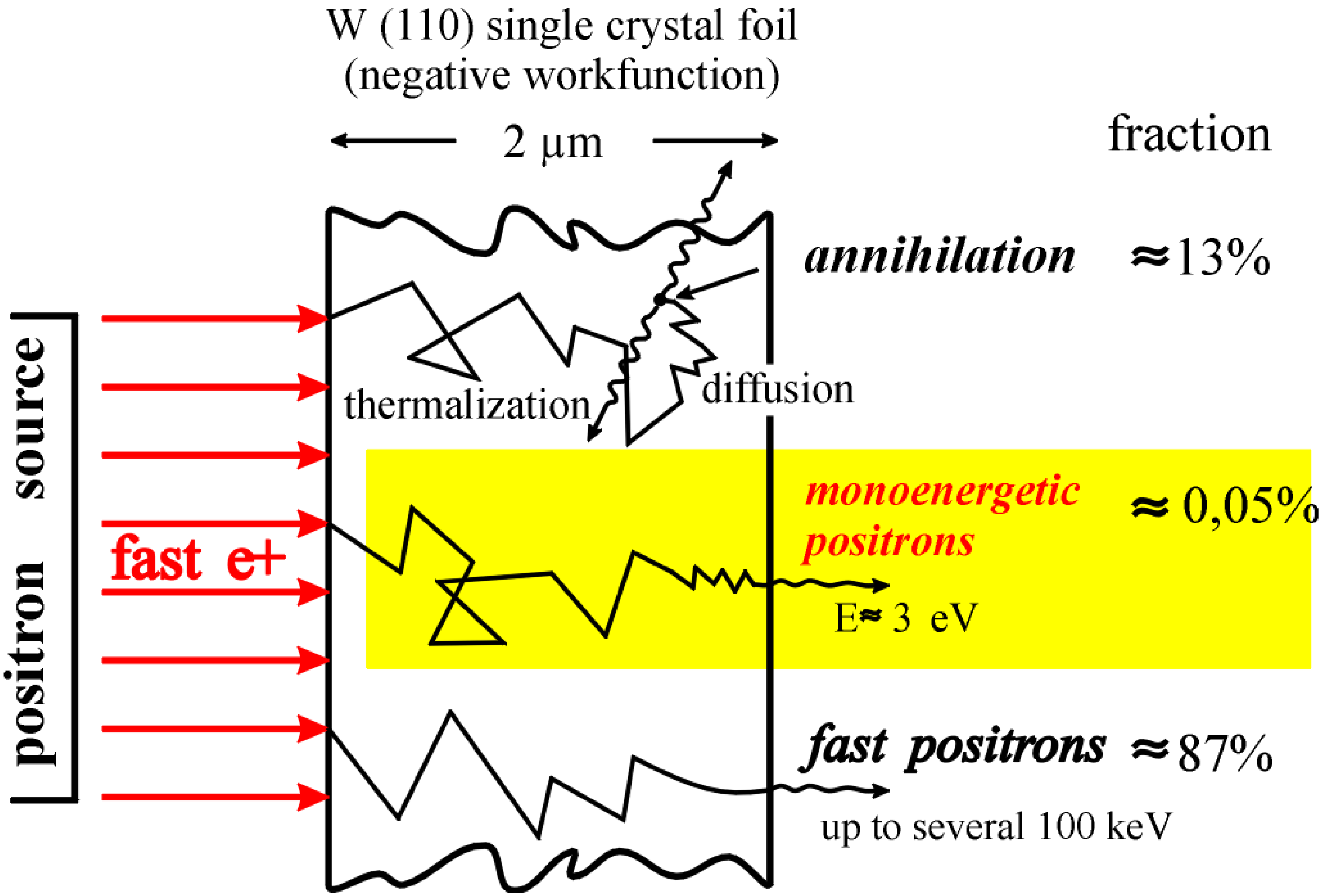
Moderation of Positrons

Mean implantation depth of un-moderated positrons from a ^{22}Na isotope source (1/*e*) for Si: 50μm



- 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



W (110) single crystal foil
(negative workfunction)

2 µm

	fraction
annihilation	≈13%
monoenergetic positrons	≈ 0,05%
fast positrons	≈87%

positron source

fast e+

thermalization

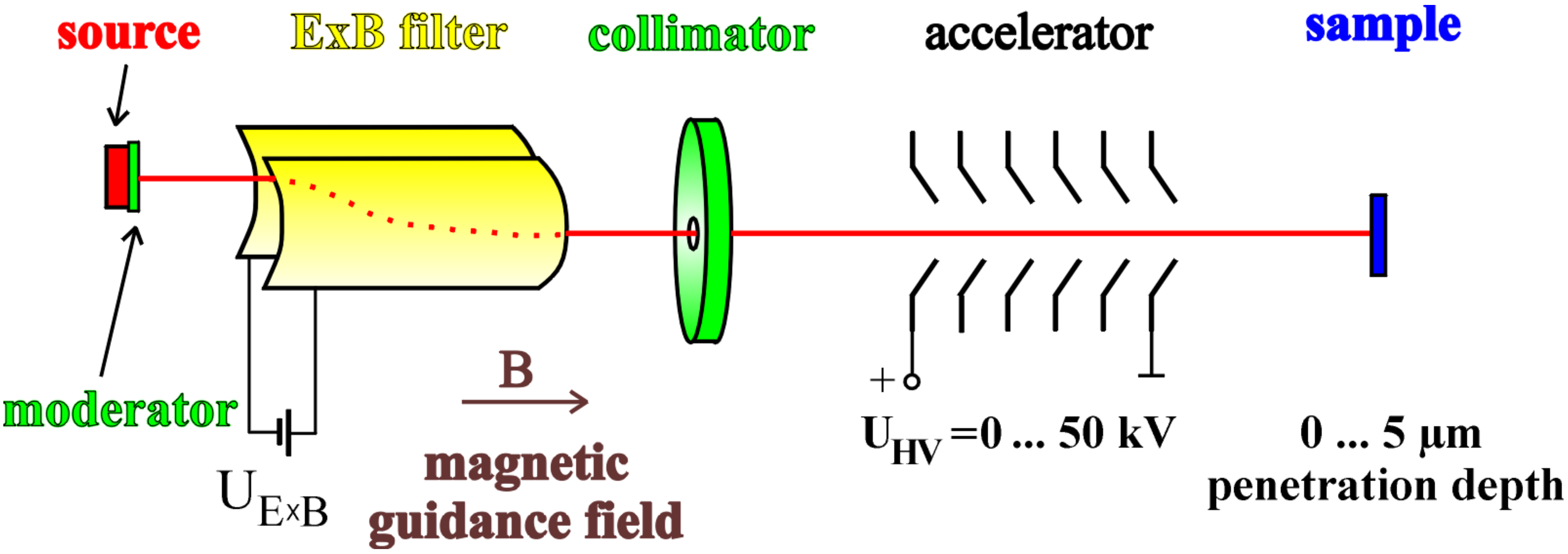
diffusion

E≈ 3 eV

up to several 100 keV

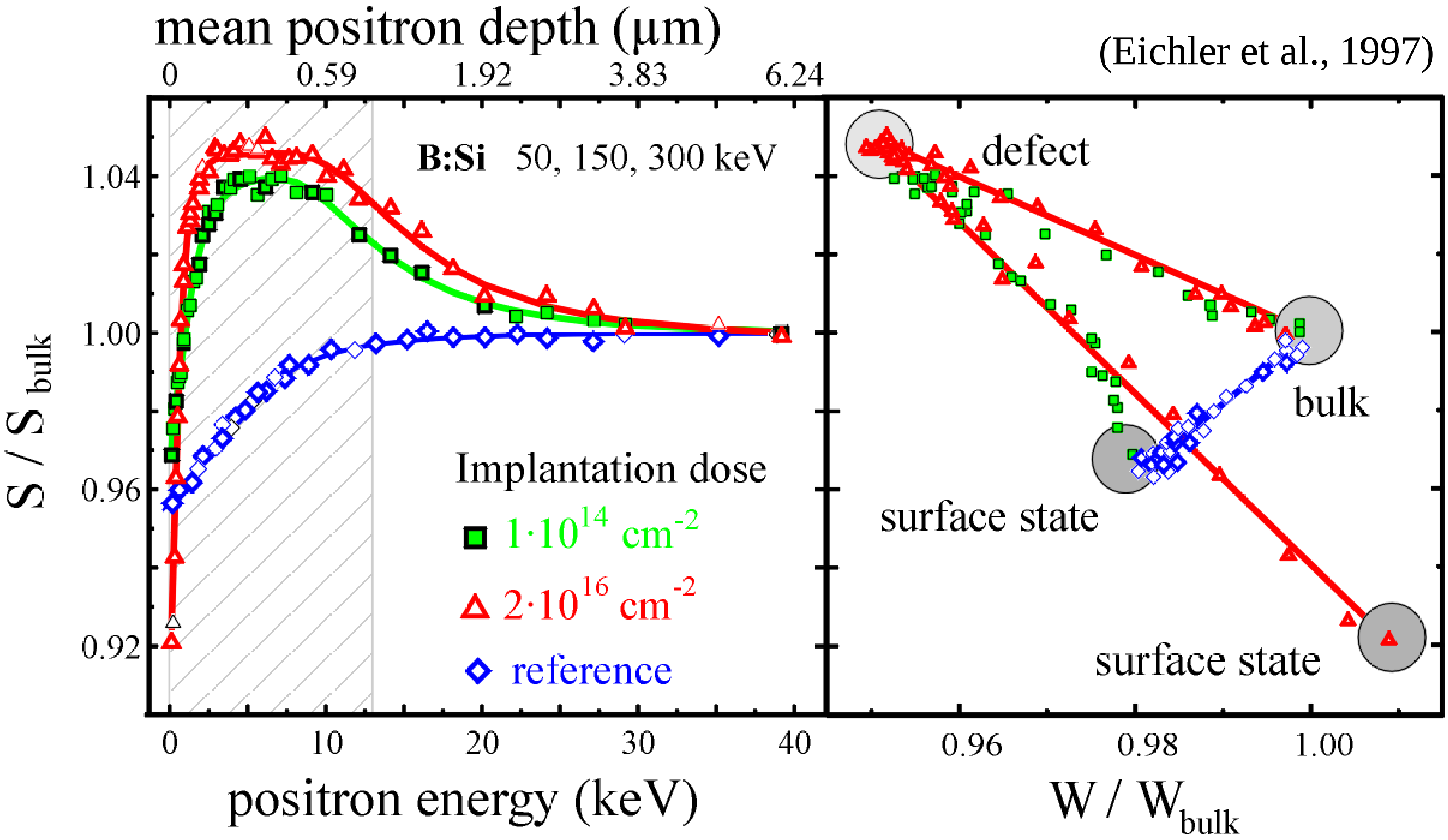
moderation efficiency: $\approx 10^{-4}$

The Positron Beam System at Halle University



Defects in Si induced by Ion Implantation

- ion implantation is most important doping technique in planar technology
- main problem: generation of defects ⇒ positron beam measurements



Study of Lubrication Defects

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

Tribology Letters Vol. 11, No. 1, 2001 29

Subsurface zones created under lubrication conditions studied by positron annihilation

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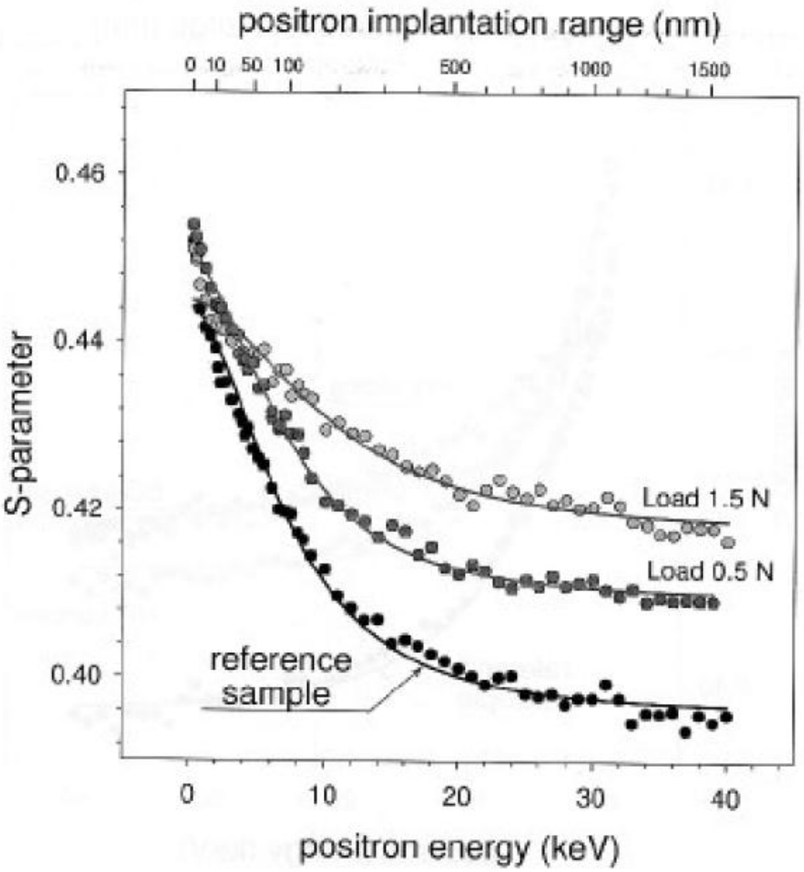
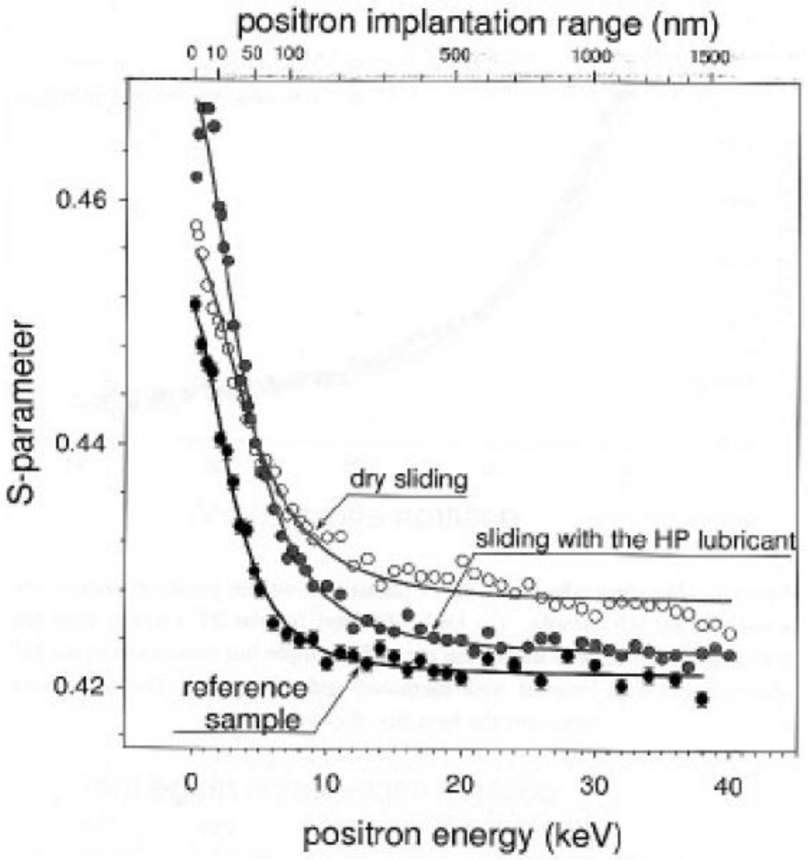
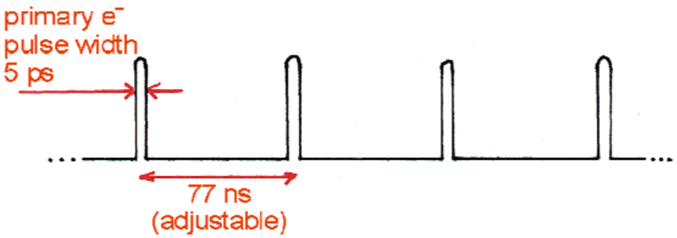
Received 11 August 2000; accepted 22 February 2001





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 ($E_+ = 0.5 \ldots 30$ 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



Concept of EPOS (ELBE Positron Source)

MePS

Mono**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

Conventional **P**ositron **S**pectroscopy

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

Information Depth:
10…200 µm

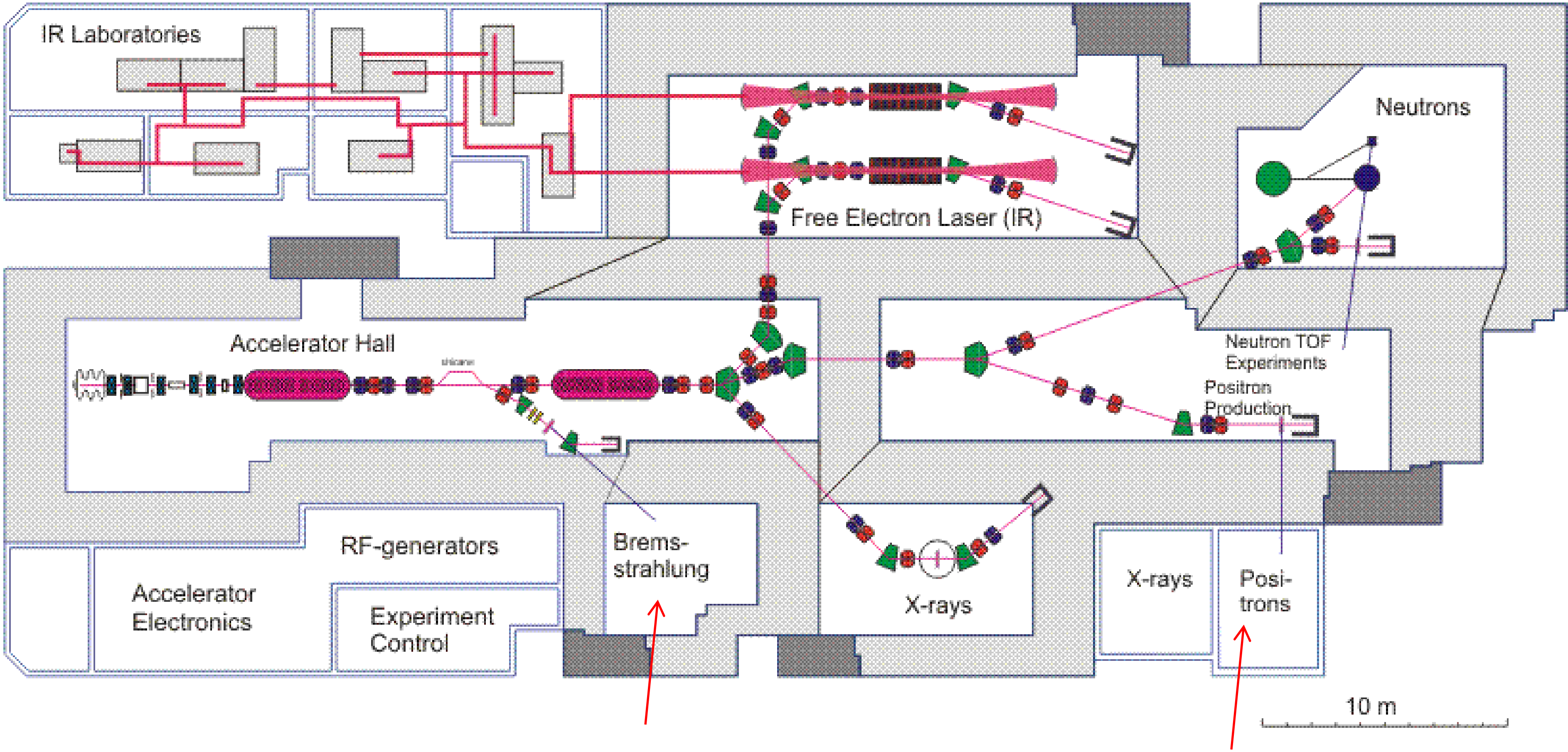
GiPS

Gamma-**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

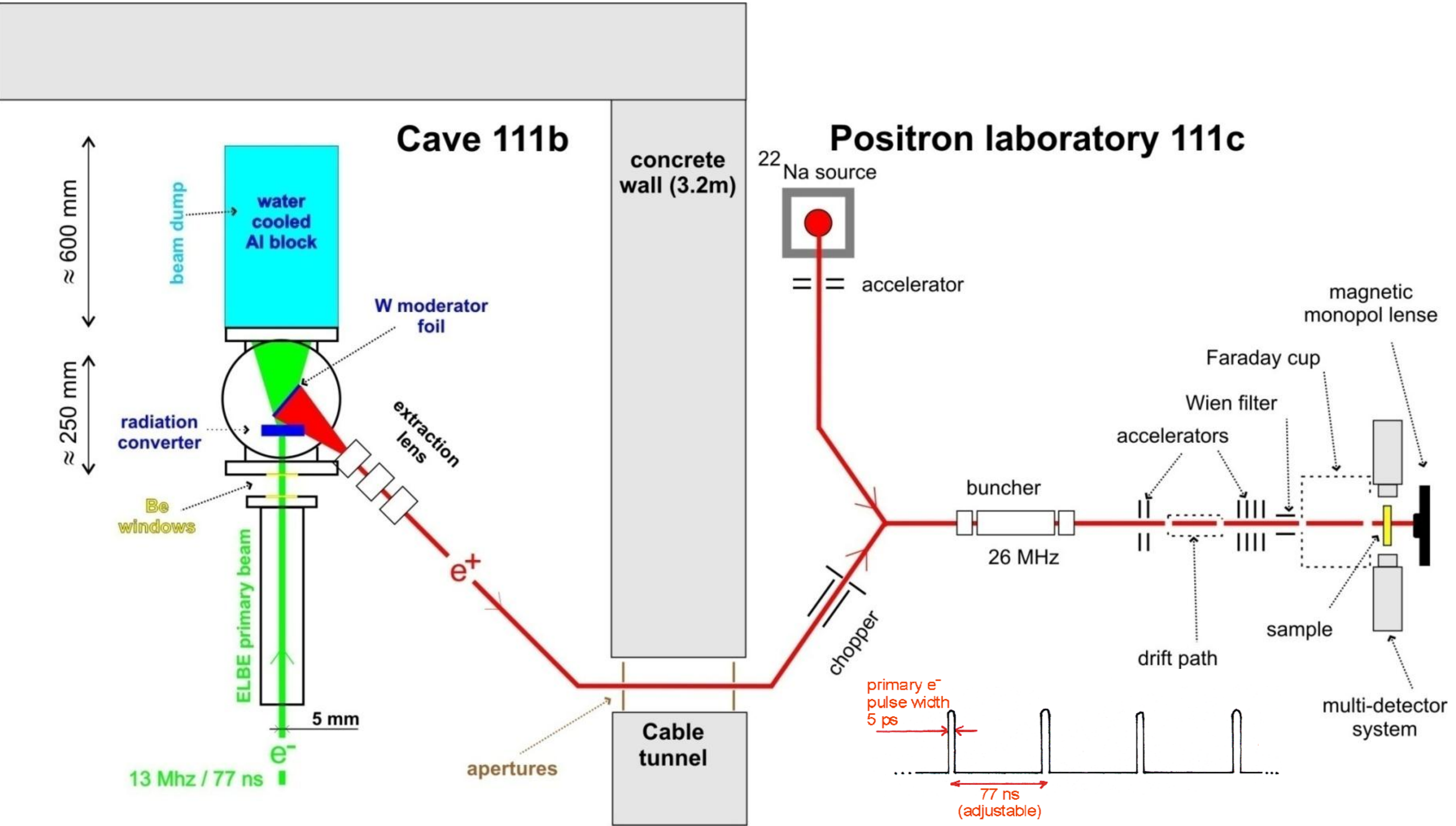


GiPS

MePS

Progress of Mono-energetic Positron Beam

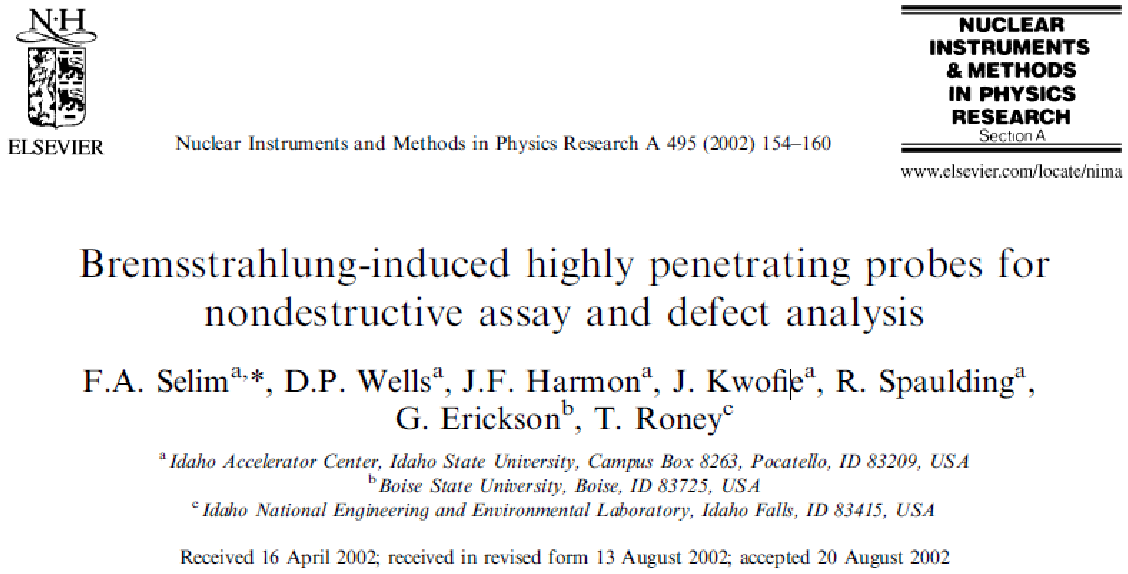
- 40 MeV, 1 mA, 26 MHz repetition time in cw mode; lifetime, CDBS and AMOC with slow e^+
- Retain original time structure for simplicity and best time resolution



Electron-Positron Converter is finished in Cave 111b

MePS at EPOS still under Construction

Gamma-induced Positron Spectroscopy



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NUCLEAR INSTRUMENTS & METHODS IN PHYSICS RESEARCH
Section A

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

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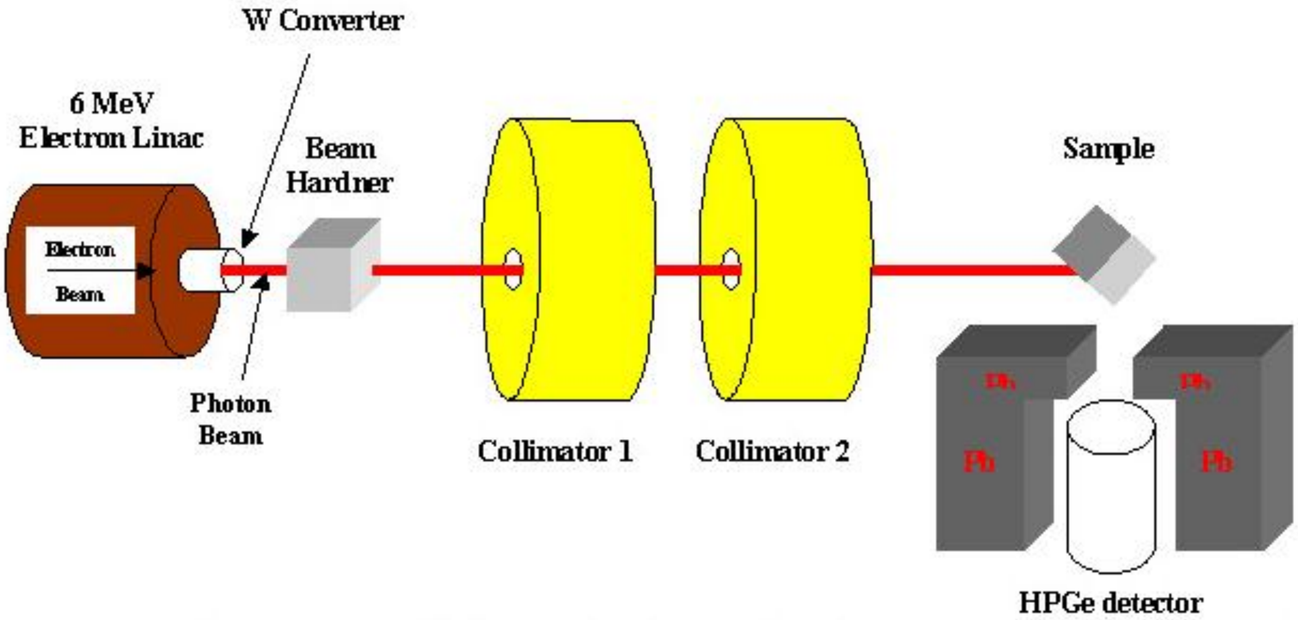
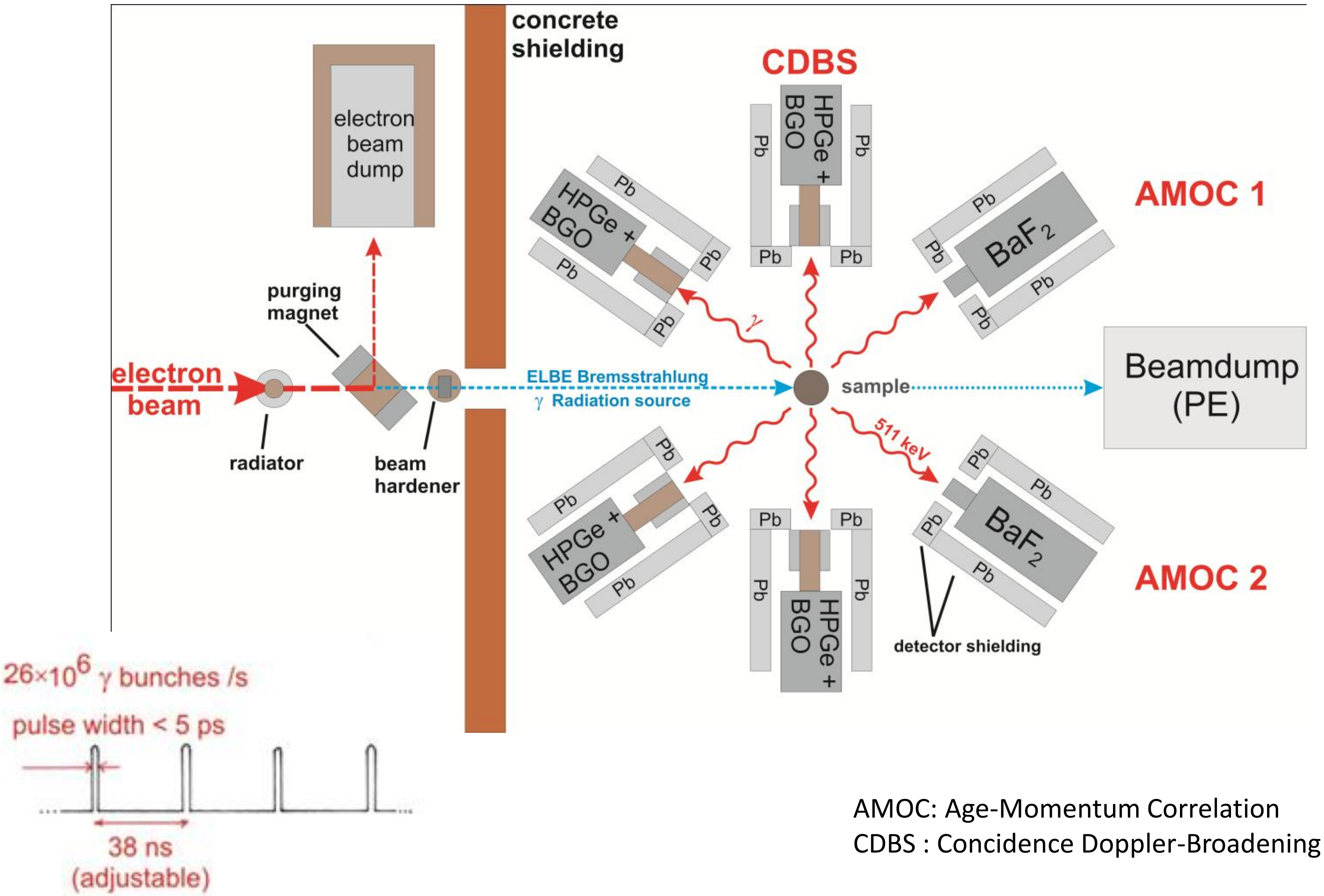
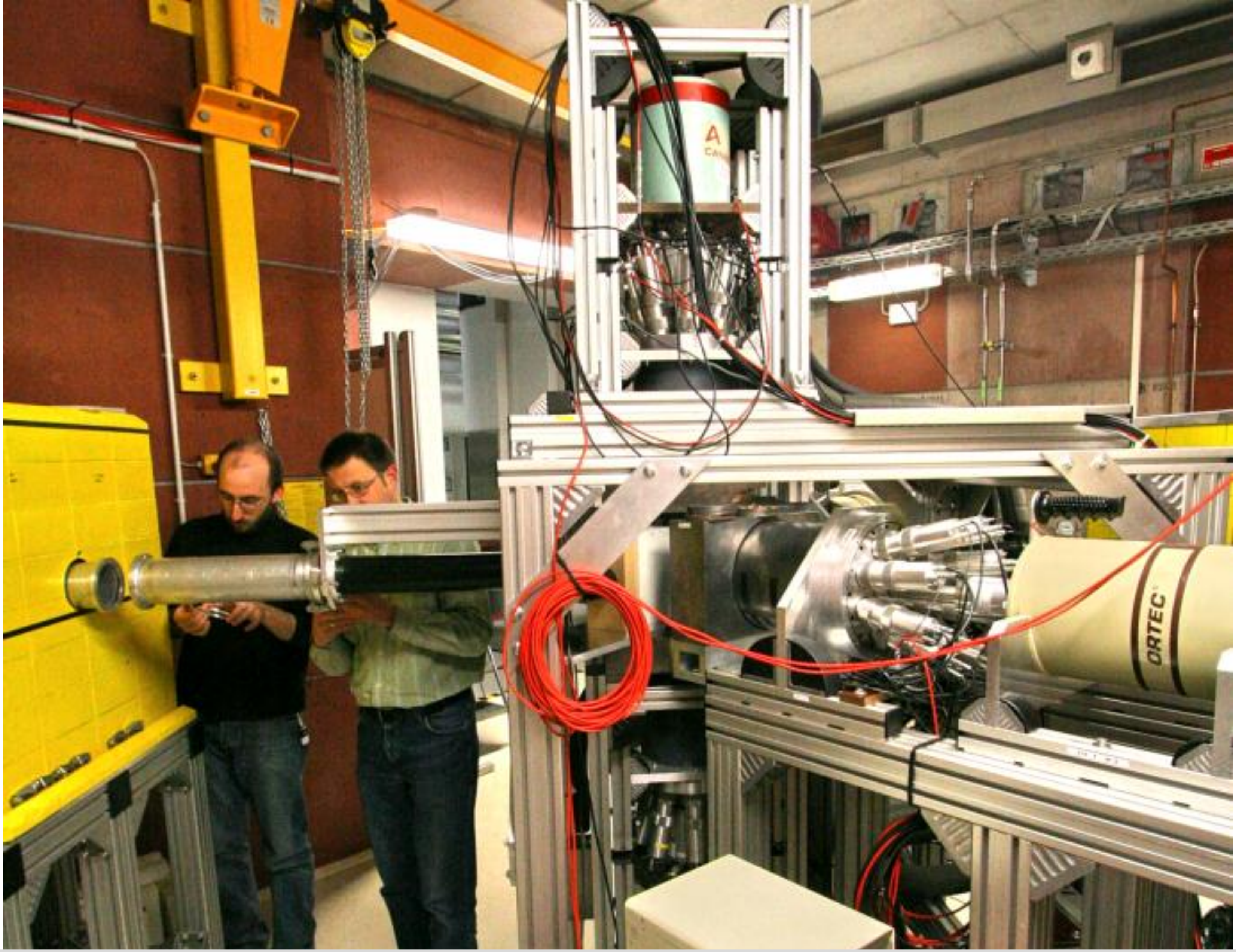


Setup extended by BaF_2 detectors for lifetime measurement



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

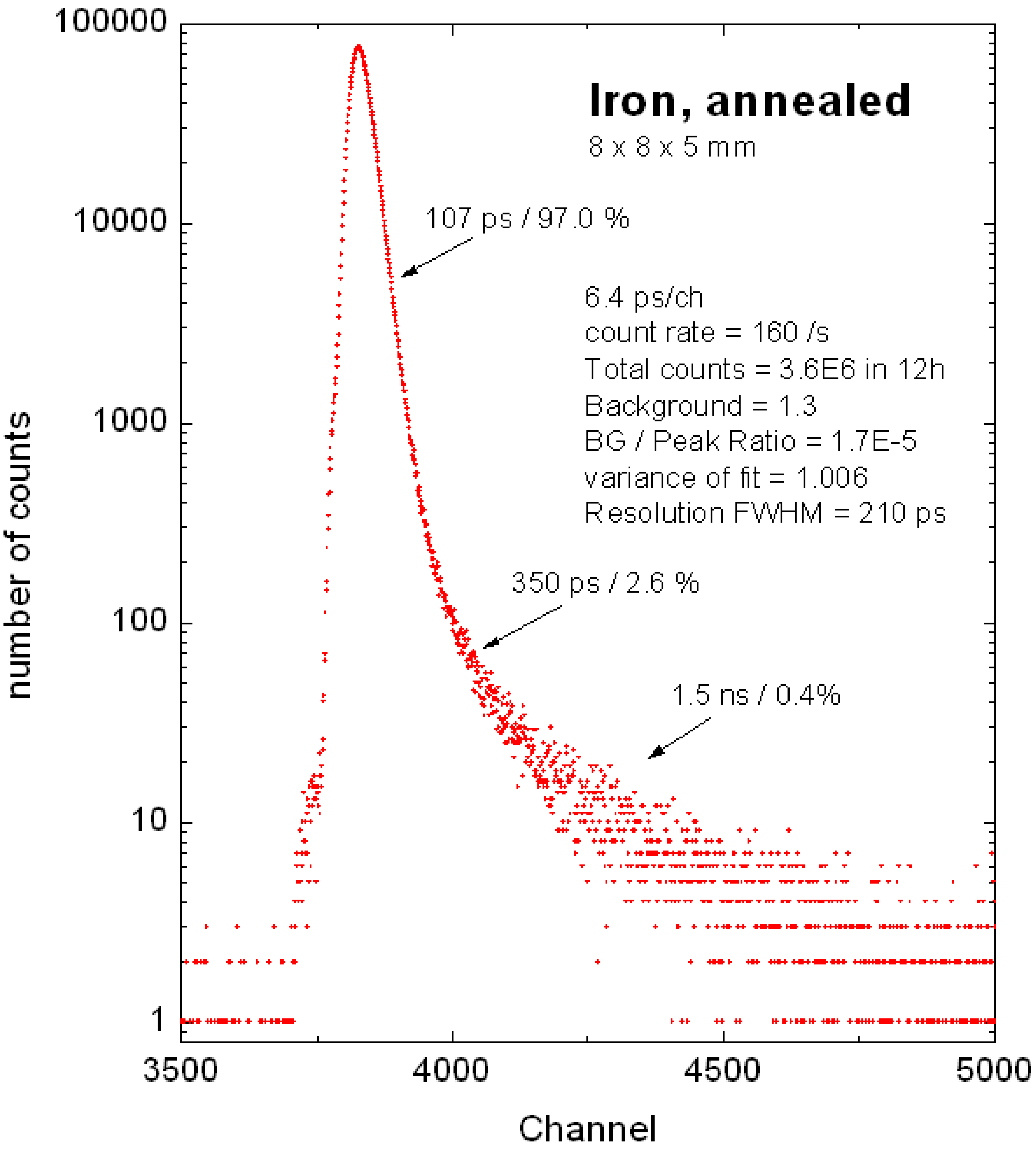
- 3 coincident setups were used: 2 AMOC and 1 CDBS spectrometer; only coincident detection ensures high spectra quality
- all scattered quanta appear within positron lifetime – time coincidence alone does not reduce background at all
- but distance helps: for 2 x 511 keV quanta in coincidence the distance 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)

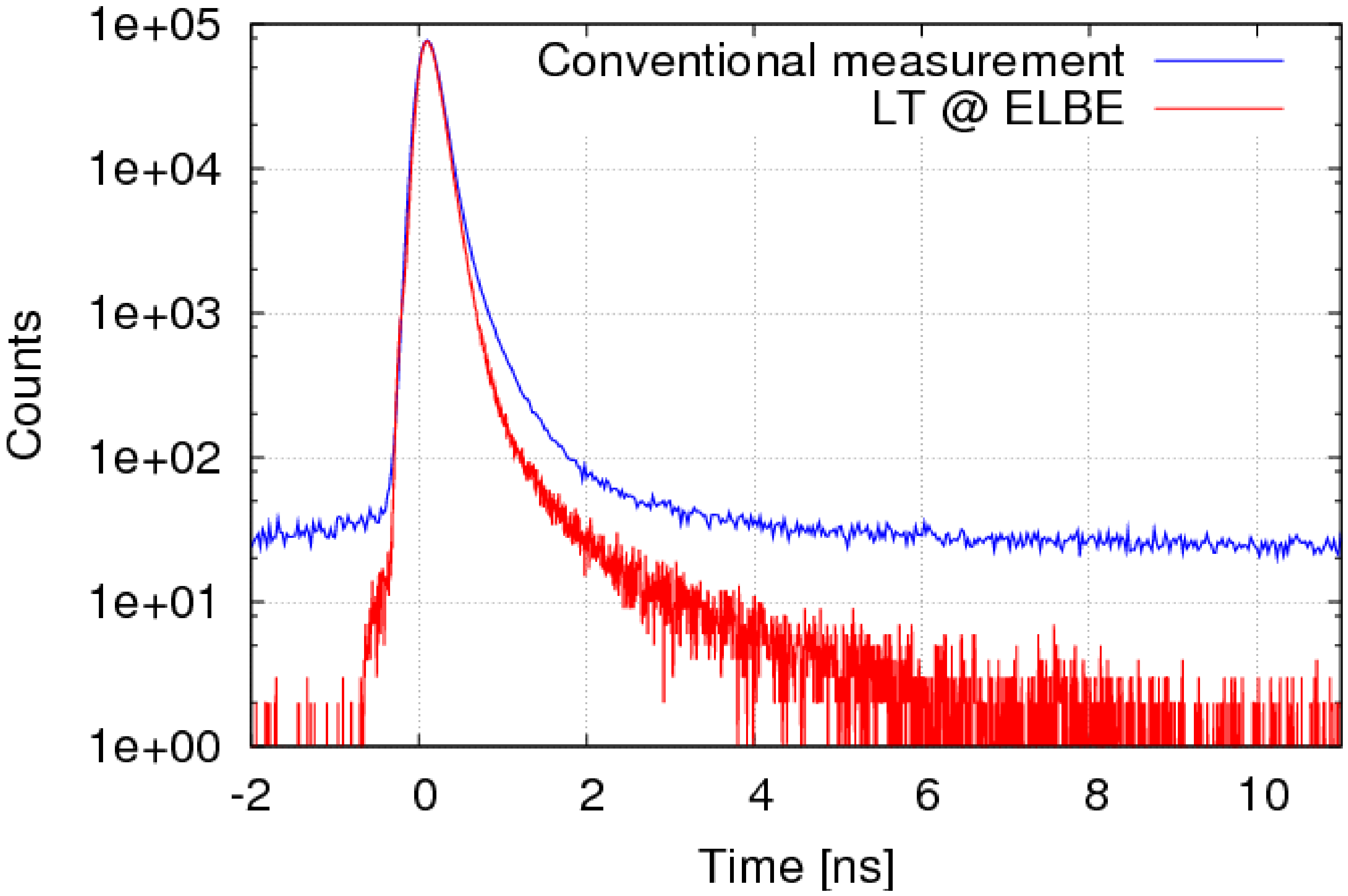
Coincident lifetime spectrum: annealed Fe

- 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)



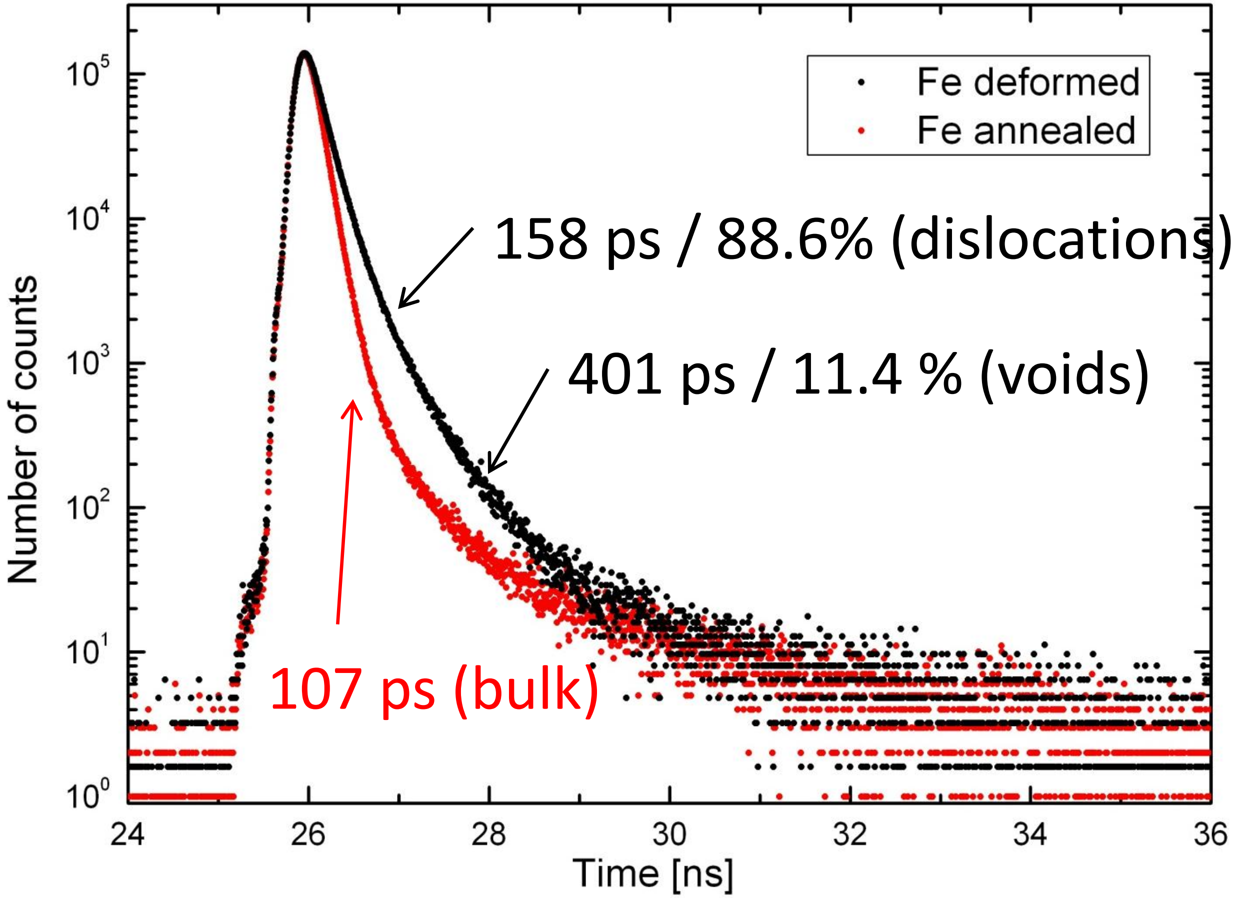
Comparison: GiPS spectrum with conventional measurement

- 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

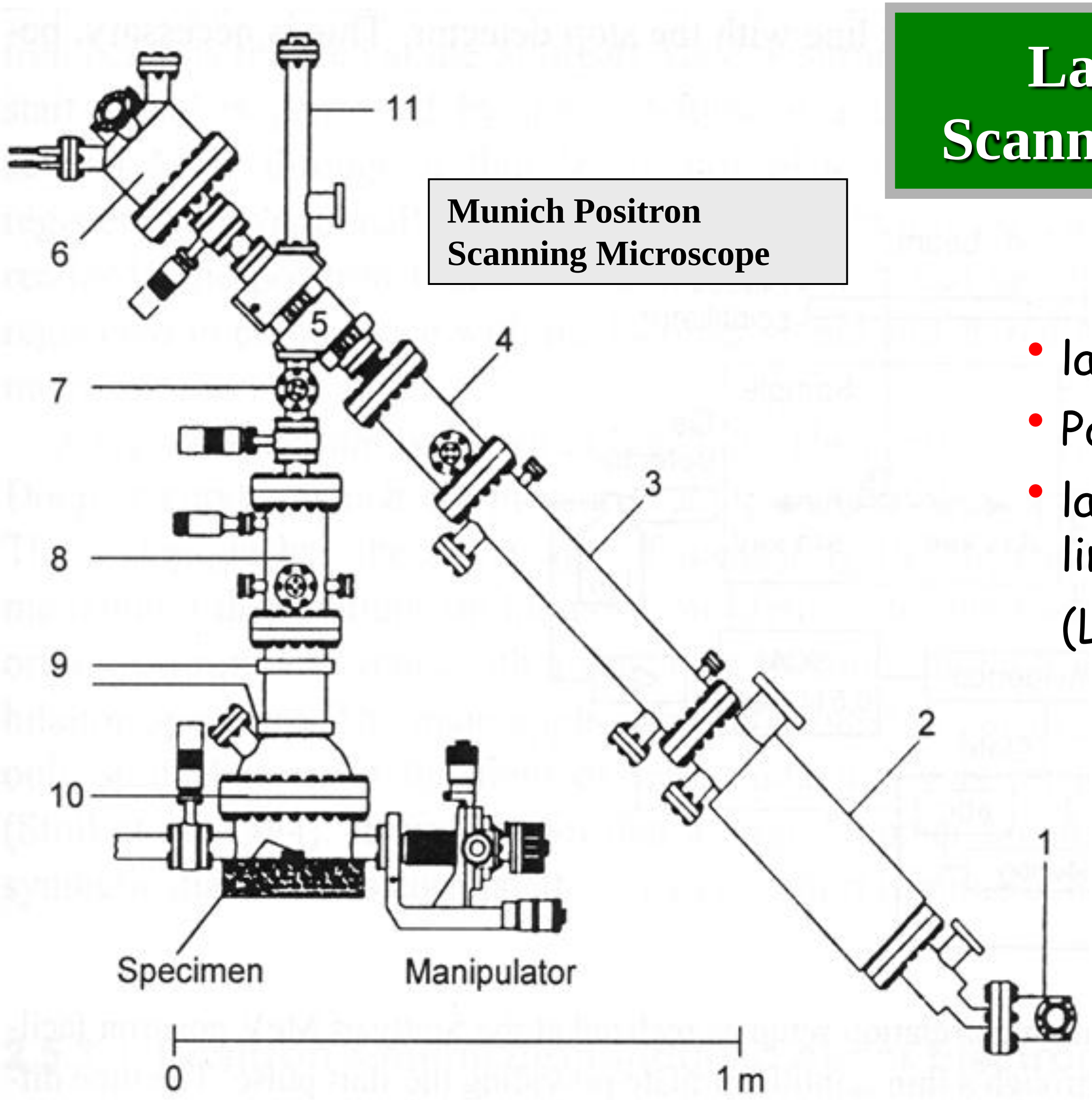
- two mechanically identical samples were prepared
- Fe annealed (1100°C; 2h in vacuum) and Fe (50% thickness reduction by cold rolling)
- spectra were easily decomposed
- expected results: annealed sample – one component 107 ps; deformed sample has 158 and 401 ps (dislocations and small vacancy clusters)



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

Lateral Resolution with Scanning Positron Microscope

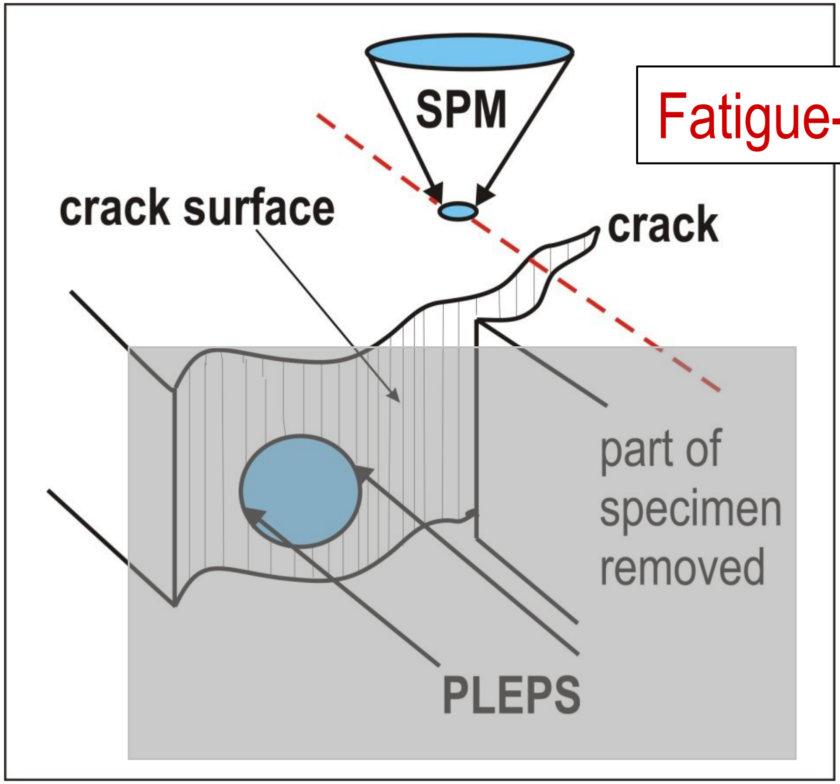


Munich Positron Scanning Microscope

- 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

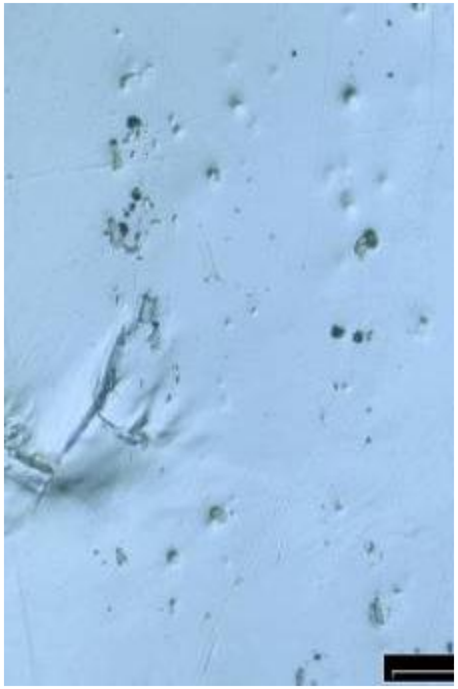


Fatigue-Crack in Al 6013

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

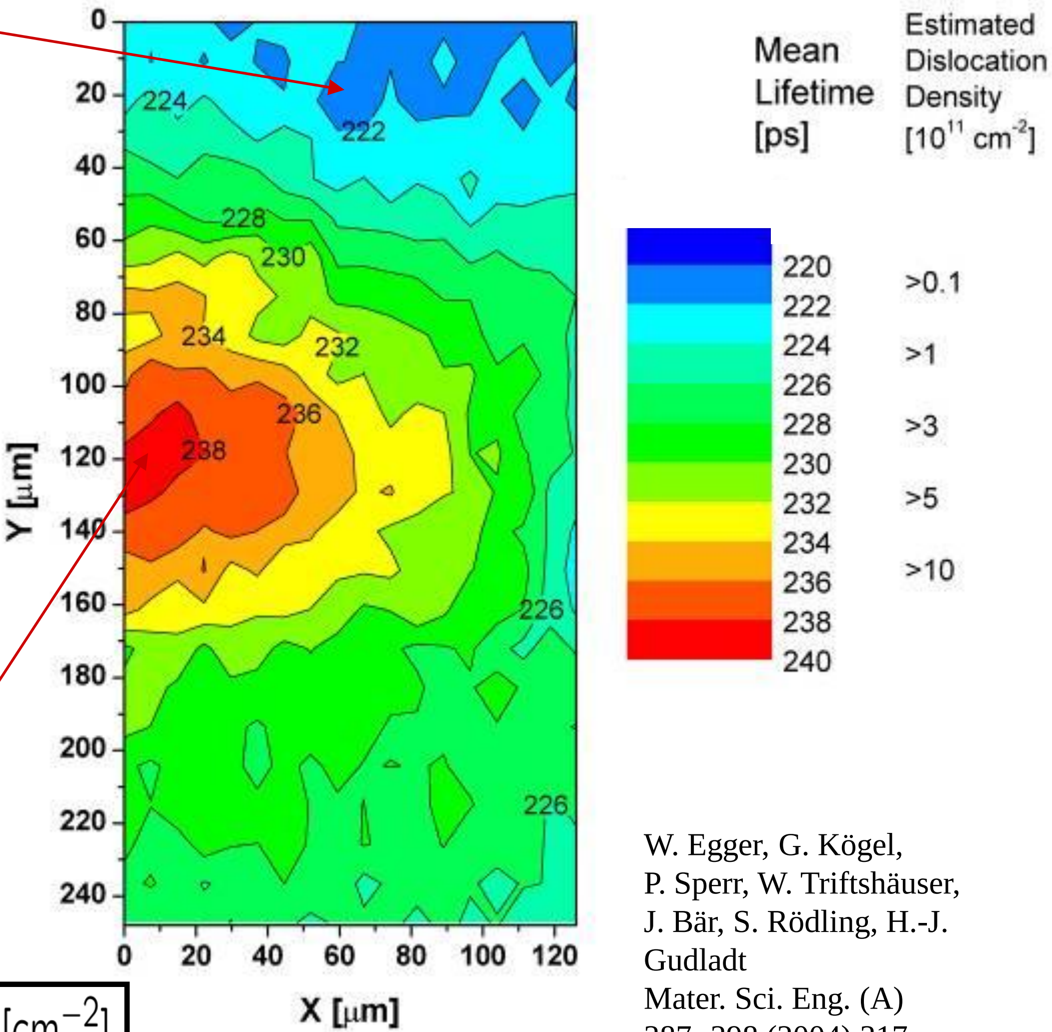
resolution 5µm; step-size 10 µm

Trapping at Mg / Si-clusters!



Only dislocations close to crack-tip !

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



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387- 398 (2004) 317

Positron Annihilation Spectroscopy: Applications

Variety of applications in all fields of materials science:

- bulk defects in semiconductors, ceramics and metals
- defect-depth profiles due to surface modifications (ion implantation; tribology)
- epitaxial layers (growth defects, misfit defects at interface, …)
- soft matter physics (open volume; interdiffusion; …)
- porosimetry 1…50 nm (e.g. low-k materials - highly porous dielectric layers)
- fast kinetics (e.g. precipitation processes in Al alloys; defect annealing; diffusion; …)
- radiation resistance (e.g. space materials)
- many more …