Application of Positron Annihilation for defects investigations in thin films

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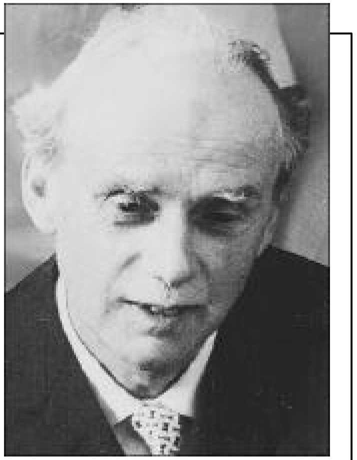
Outlook:

- Introduction to Positron Annihilation
- Methods
  - Positron lifetime spectroscopy
  - Doppler broadening spectroscopy
- Applications to thin films
  - Slow positron beam
  - Positron microscopy

Positron – the first discovered antiparticle



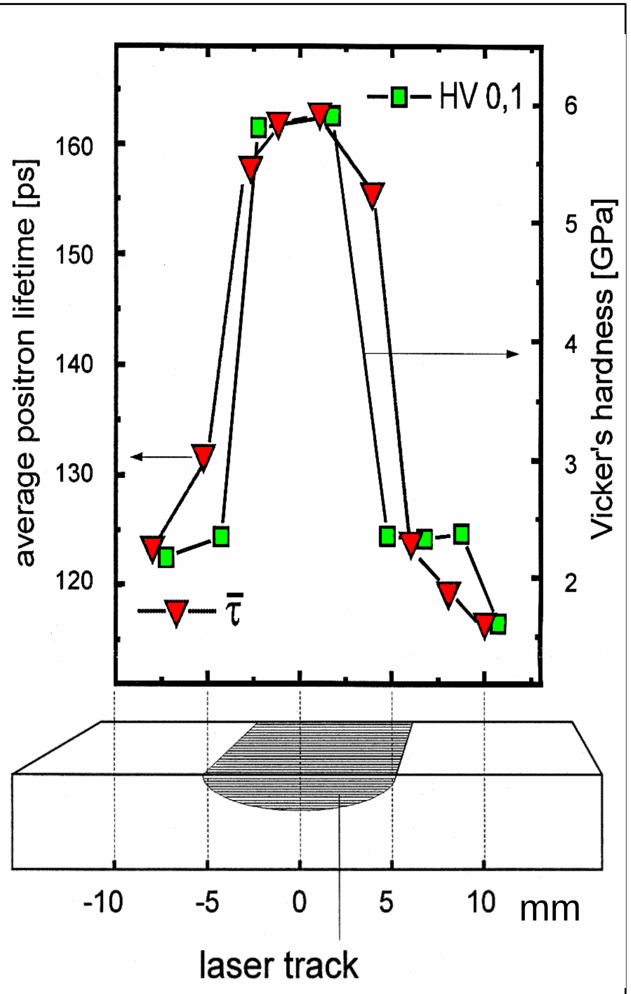
- D.A.M. Dirac
  - predicted the existence of a positron in 1928 as an explanation of negative energy solutions of his equation: $E = \pm\sqrt{p^2c^2 + m^2c^4}$

    Dirac D.A.M. (1928): Proc. Roy. Soc. 117, 610 **(Nobel prize 1933)**
- C.D. Anderson
  - **1932** discovers positrons in a cosmic ray event in a Wilson cloud-chamber

    Anderson C.D. (1932): Science 76, 238 **(Nobel prize 1936)**
  - **1933** evidence of e^+-e^- pair formation by registration of **annihilation** Gamma quanta

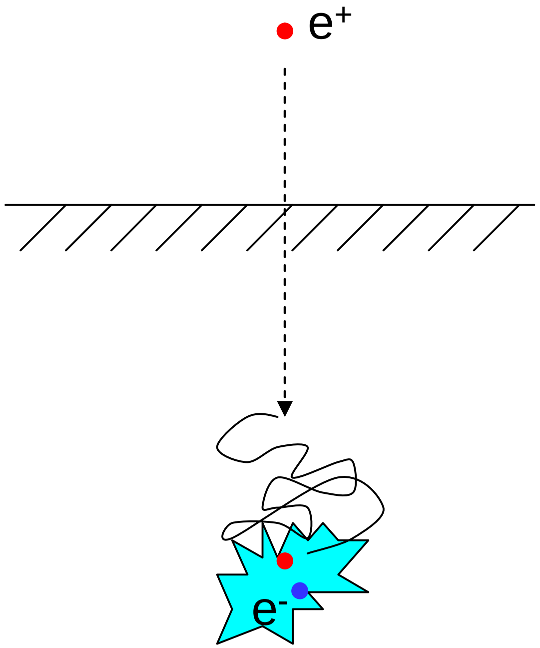
Application of Positron Annihilation

- Materials
  - Condensed matters
    (metals, semiconductors, polymers…)
  - Liquids
  - Gases
- Sensitivity
  - Vacancy-like defects and defect complexes
  - Concentration limits 10^{14}-10^{19} cm^{-3}
- Information
  - Type of vacancy-like defects
  - Chemical surrounding of a vacancy
  - Vacancy-like defects depth profiling
  - 3D-imaging using micro-beam

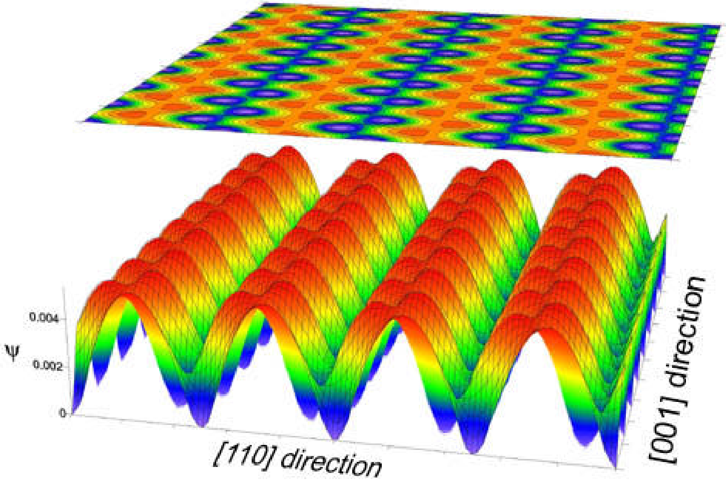


Ex: Laser hardening of Ck60-Steel

Positron in condensed matter



- Thermalization
  - energy loss through electron/phonon excitation
  - 1 - 3 ps
  - Penetration depth ≈ E/ρ
- Diffusion
  - $L_+ \approx 100$ nm
  - Positron wave function in [110] plane of GaAs



- Annihilation
  - mainly with emitting of two γ-quanta

$$3g \;/\; 2g \;= 0.27\%$$

2γ-annihilation

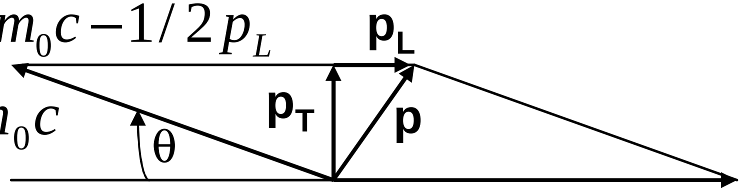
Sensitivity to electron momentum

energy and momentum conservation leads to

- angular correlation of annihilation radiation
- Doppler broadening of annihilation line

$$p_2 = m_0 c - 1/2\, p_L$$

$$\Theta \cong p_T / m_0 c$$



$$p_1 = m_0 c + 1/2\, p_L$$

p – momentum of e$^+$-e$^-$ pair

p$_1$, **p**$_2$ – γ-quanta's momentum

Sensitivity to electron density

- Positron Lifetime Spectroscopy (PALS)

positron diffusion: $L_+ = \sqrt{D_+ \tau_b}$ during τ_b – positron bulk lifetime

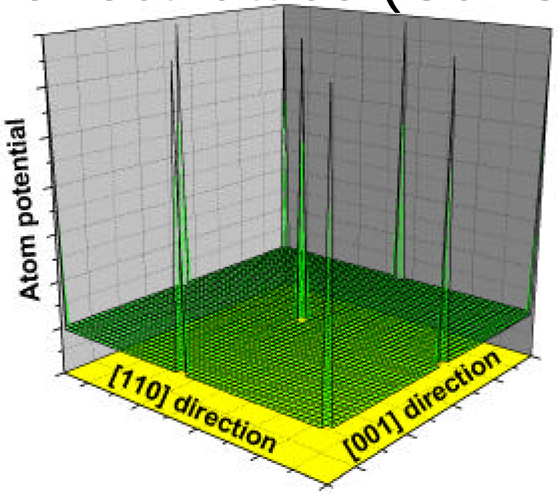
annihilation rate:

$$\lambda = 1/\tau_b = \pi \cdot r_0 \cdot c \int \psi_+(\mathbf{r})\, \psi_-(\mathbf{r})\, \gamma \, d\mathbf{r}$$

the lower the electron density is, the higher is the positron lifetime

Positron trapping

- Perfect lattice (GaAs plane [110])

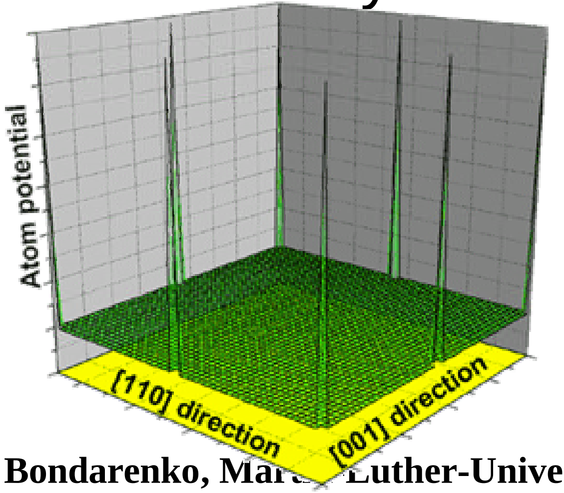


- Positrons are repelled by positive atom cores

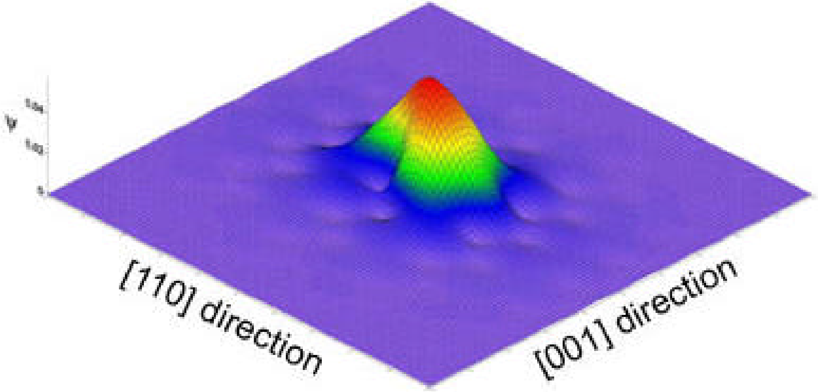
- Vacancy represents a positron trap due to the missing nuclei (potential well for a positron)

Positron Annihilation is sensitive to vacancy-like defects

- Mono-vacancy



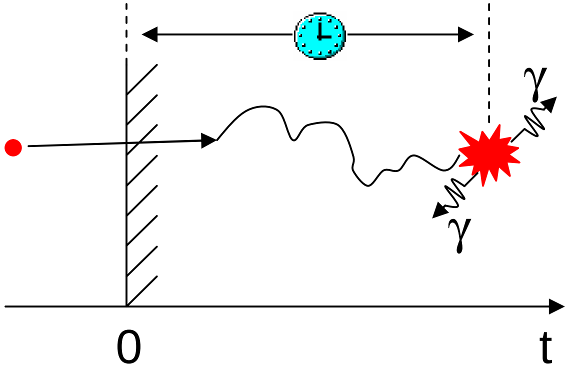
- Because of reduced electron density positrons live longer in vacancies



Positron Annihilation Lifetime Spectroscopy (PALS)

Technique

- γ-detection: scintillator + photomultiplier
- Time between positron penetration and it’s annihilation in a sample is measured
- 3-6×10^6 are accumulated in a spectrum



Mathematics

- probability **n(t)** that e^+ is alive at time **t**:

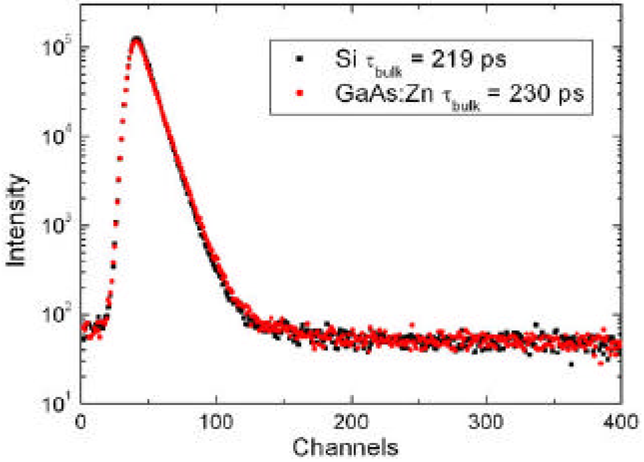
  λ - positron annihilation rate
- Positron lifetime spectrum in bulk:

$$\boxed{\frac{dn(t)}{dt} = -\lambda\, n(t)} \qquad n(0) = 1$$

$$n(t) = e^{-\lambda_{bulk} t} \longrightarrow$$

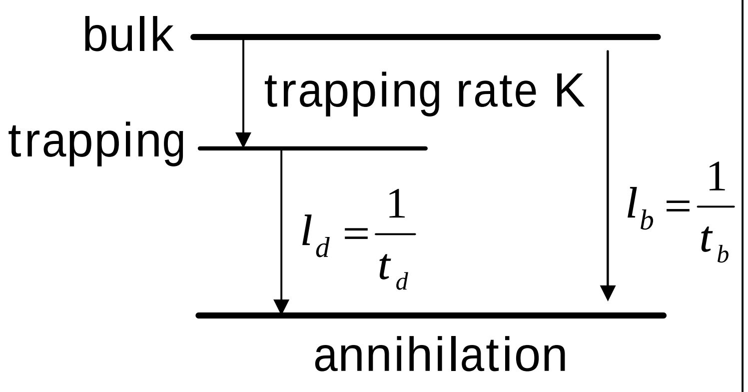
slope of the exponential decay

$$\lambda_{bulk} = \frac{1}{\tau_{bulk}}$$



Positron Annihilation Lifetime Spectroscopy

Physics



one-defect trapping model

- annihilation from bulk with $\lambda_b=1/\tau_b$ s^{-1}
- trapping to vacancy-defect with **K** s^{-1}
- annihilation from the defect with $\lambda_d=1/\tau_d$
- two-component lifetime spectrum

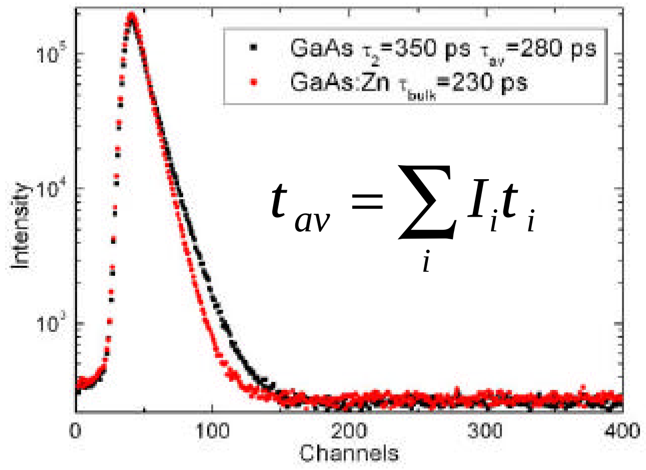
$$N(t) = I_1/\tau_1 \exp(-t/\tau_1) + I_2/\tau_2 \exp(-t/\tau_2)$$

Information

- vacancy type (mono-, di-, vacancy cluster)

  τ_2 – reflects the electron density
- defect concentration C

$$K = \frac{I_2}{I_1}\left(\frac{1}{\tau_b} - \frac{1}{\tau_2}\right) \approx C$$

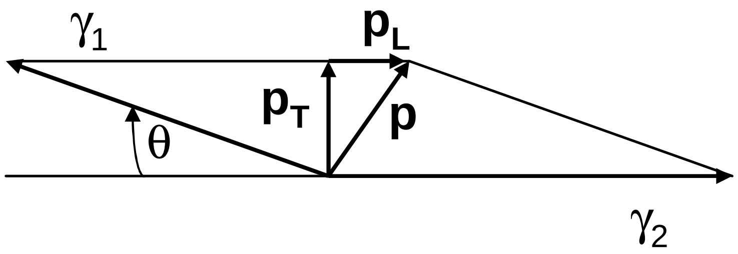


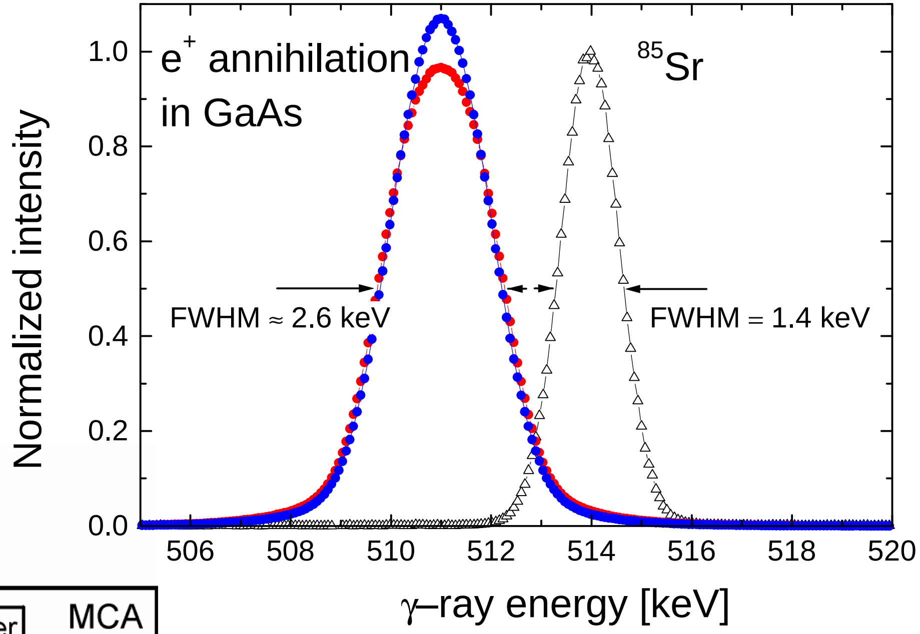
$$\tau_{av} = \sum_i I_i \tau_i$$

Annihilation-Line Doppler broadening spectroscopy

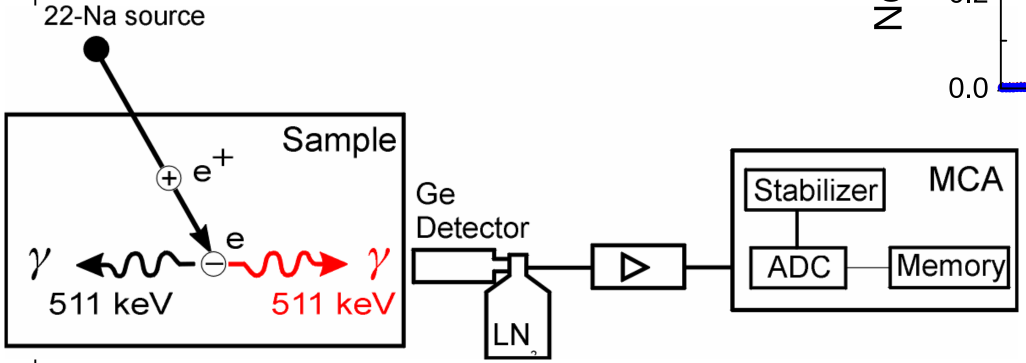
Doppler effect

- electron momentum in propagation direction of 511 keV γ-ray leads to Doppler broadening of annihilation line





Technique



$$E_1 - E_2 = p_L c$$

E_1, E_2 – energy of γ quanta

Annihilation-Line Doppler broadening spectroscopy

Data Treatment

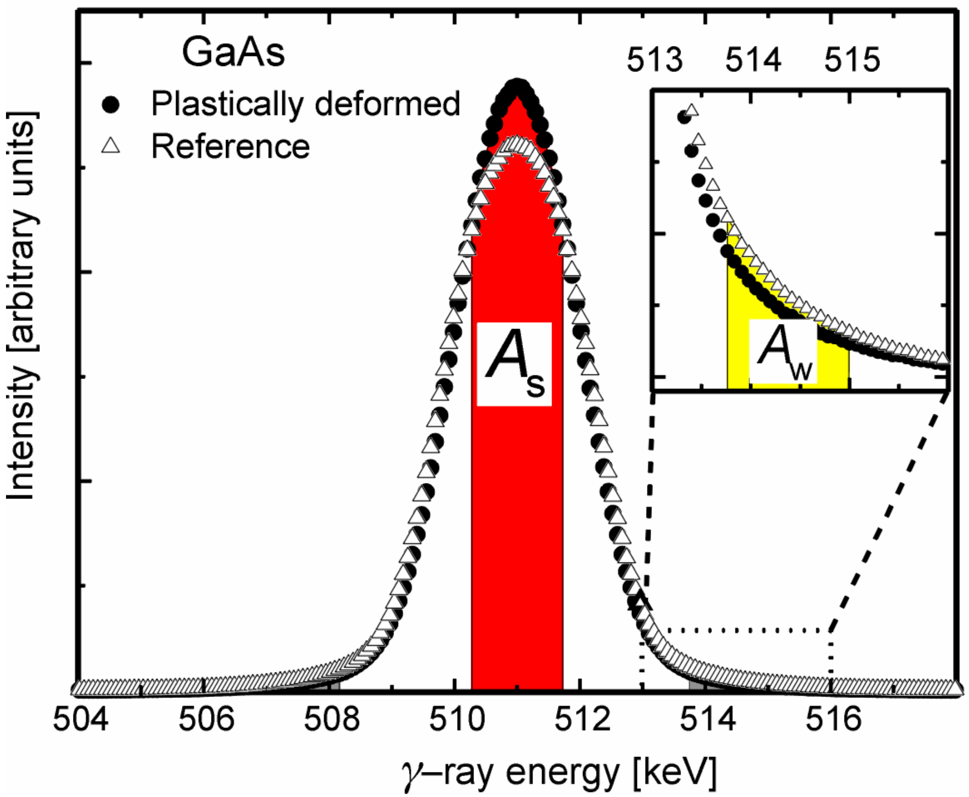
Line Parameters

- “Shape” parameter

$$S = \frac{A_s}{A_0}, \quad A_s = \int_{E_0-E_s}^{E_0+E_s} N_D dE$$

- “Wing” parameter

$$W = \frac{A_w}{A_0}, \quad A_w = \int_{E_1}^{E_2} N_D dE$$



Information

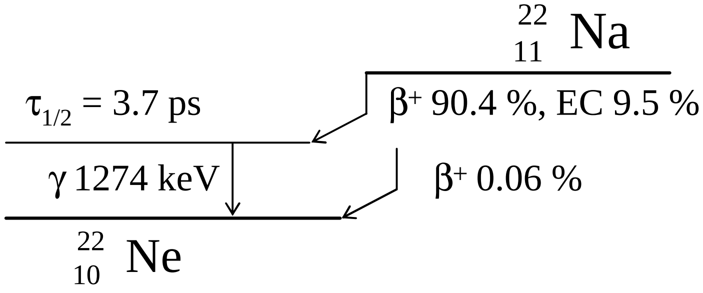
- Both S and W are sensitive to the concentration and defect type
- W is sensative to chemical surrounding of the annihilation site, due to high momentum of core electrons participating in annihilation

Positron source

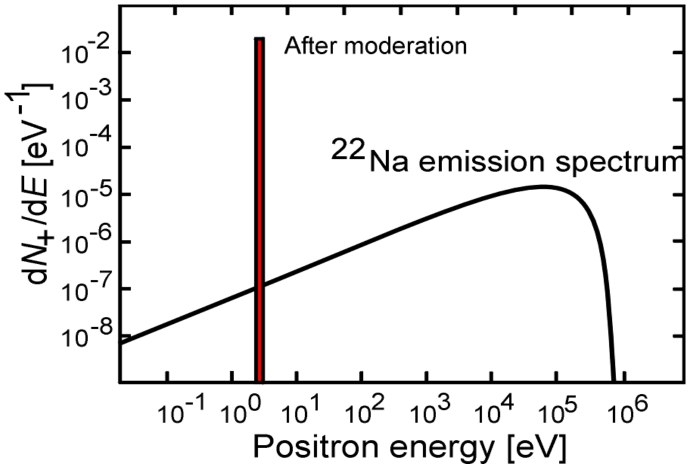
- β-decay of radioactive isotopes

Radionuclide	half-life	Maximum energy	γ-rays intensity
^{22}Na	2.6 years	545 keV	100 %
^{58}Co	71 days	470 keV	99 %
^{64}Cu	12.8 hours	1340 keV	0.5 %

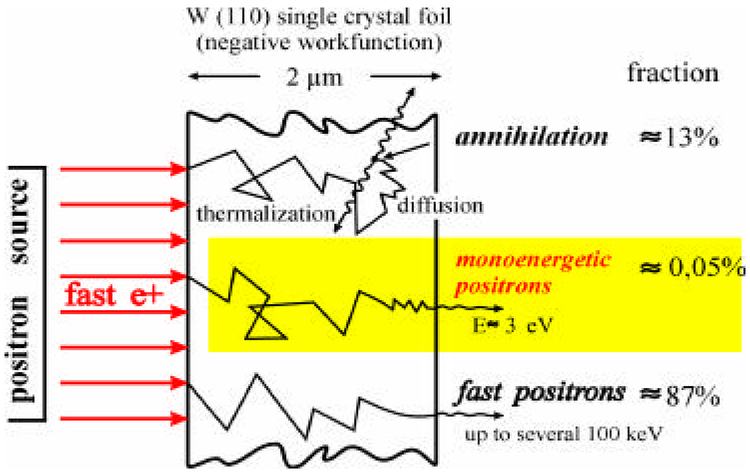
$$^{22}_{11}\mathrm{Na} \to {}^{22}_{10}\mathrm{Ne} + \mathrm{e}^{+} + u$$



- Energy distribution after β+-decay

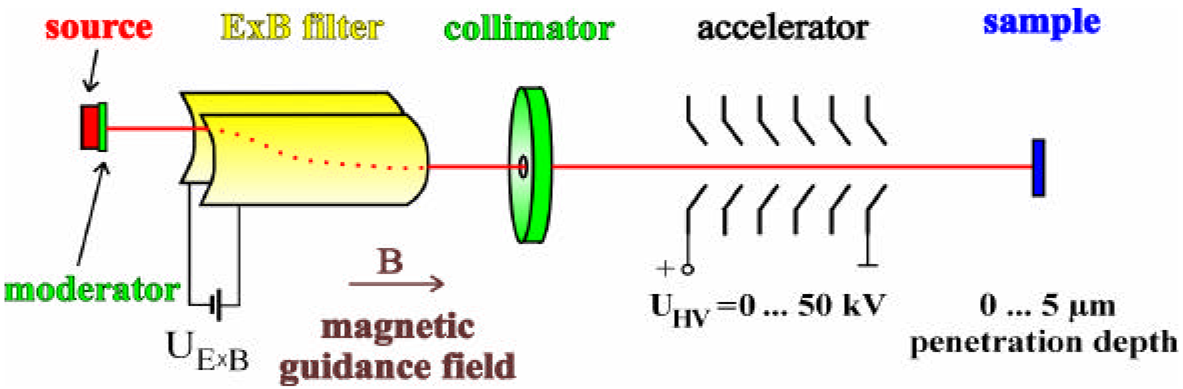


- Moderation



Conventional positron beam technique

- Monoenergetic positrons are used
- Magnetically guided



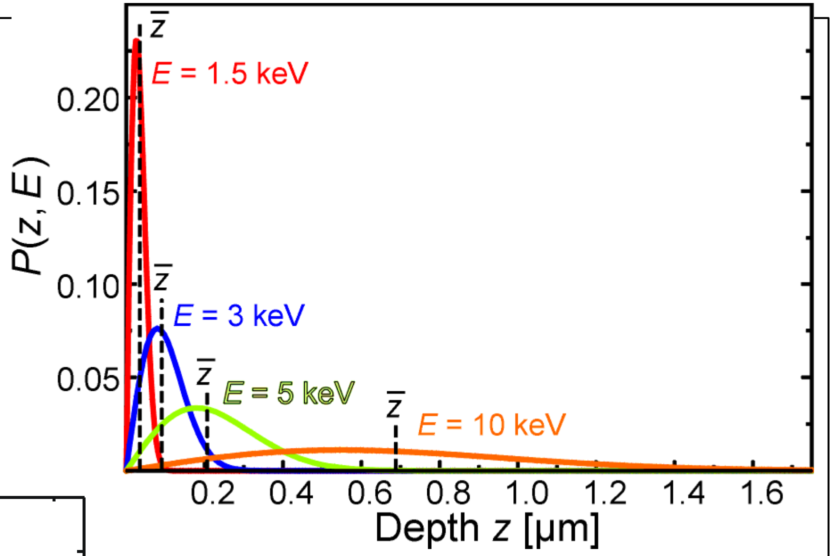
- Disadvantages
  - no simple lifetime measurements and bad lateral resolution (0.5-1 mm)
  - defect studies by Doppler-broadening spectroscopy
  - characterization of defects only by line-shape parameters or positron diffusion length

Information from Doppler broadening spectroscopy

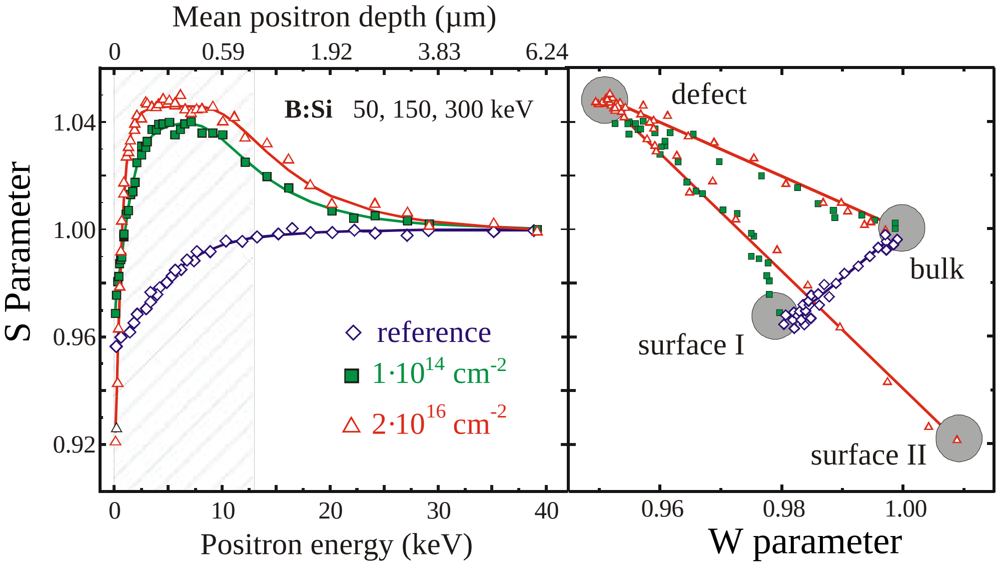
- Positron implantation profile

  Makhov function:

$$P(z,E) = \frac{mz^{m-1}}{z_0^m}\left[-\left(\frac{z}{z_0}\right)^m\right]$$



- S-E and S-W plots

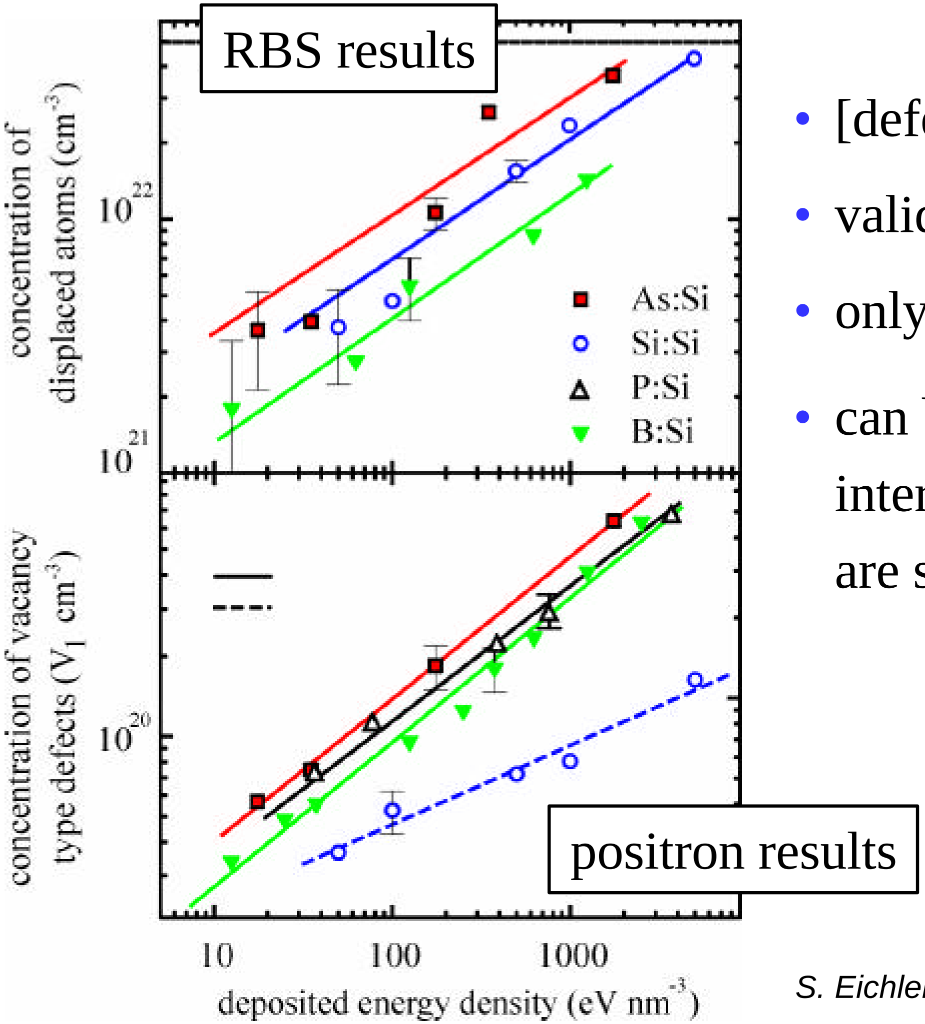


Ion implantation in Si

S. Eichler, PhD Thesis, 1997

- Positrons annihilation sites:
  - surface
  - bulk
  - vacancy defect

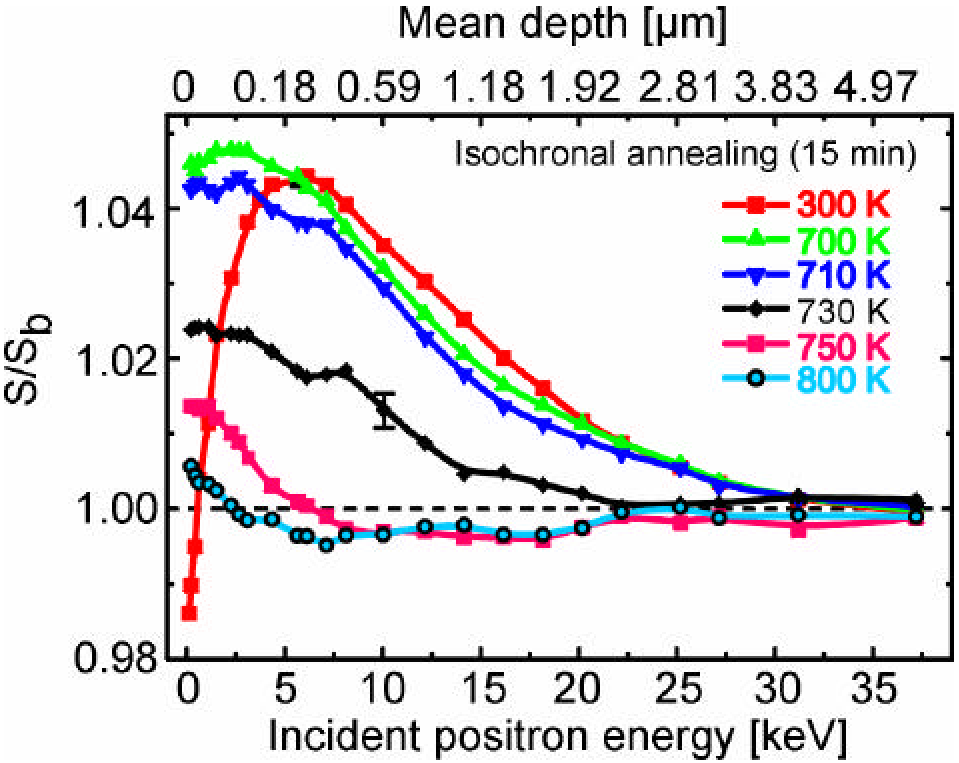
Defect density as a function of deposited ion energy



- [defect] ~ dose$^{0.5}$
- valid for RBS- and positron data
- only exception: Si self-implantation
- can be explained: extra Si atoms are interstitials and kill vacancies that are seen by positrons but not by RBS

S. Eichler, PhD Thesis, 1997

Annealing behavior of defects

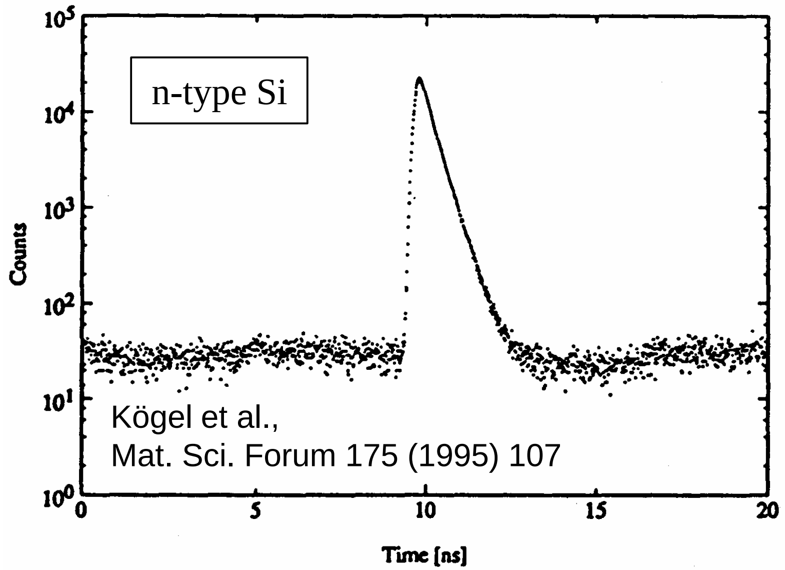


S. Eichler, PhD Thesis, 1997

- Annealing of defects in boron-implanted FZ-Si
- Main annealing stage at 730K
- but divacancies anneal at 550K
- larger clusters are the dominating defects

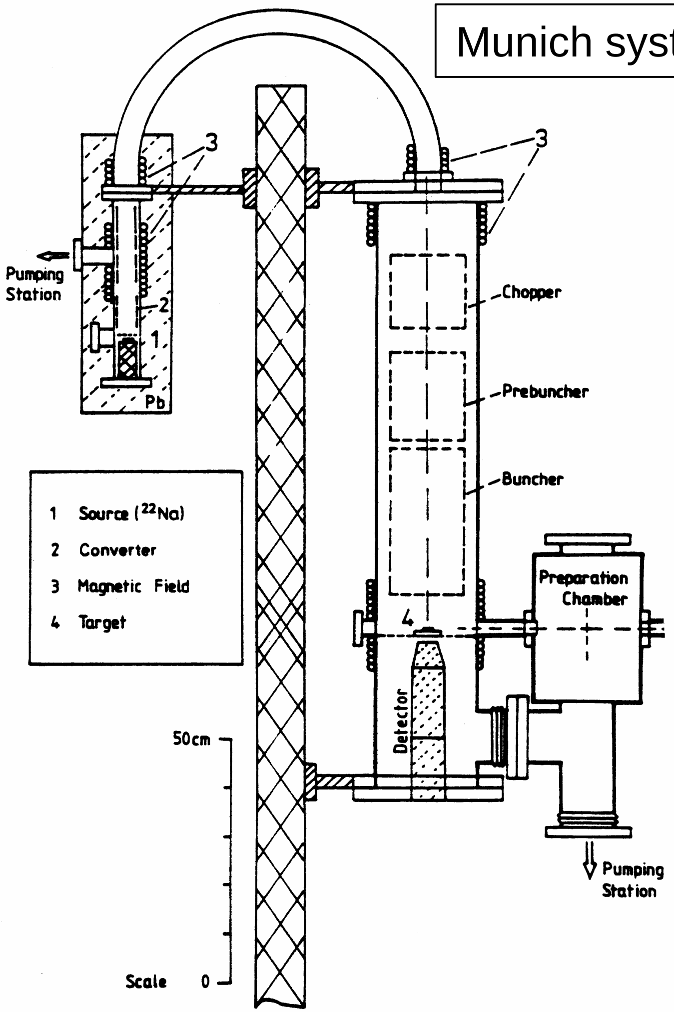
Positron lifetime beam

- lifetime measurements are more difficult
- a system of chopper and bunchers: short pulses of monoenergetic positrons
- two systems are available till now:
  - Munich (Germany)
  - Tsukuba (Japan)

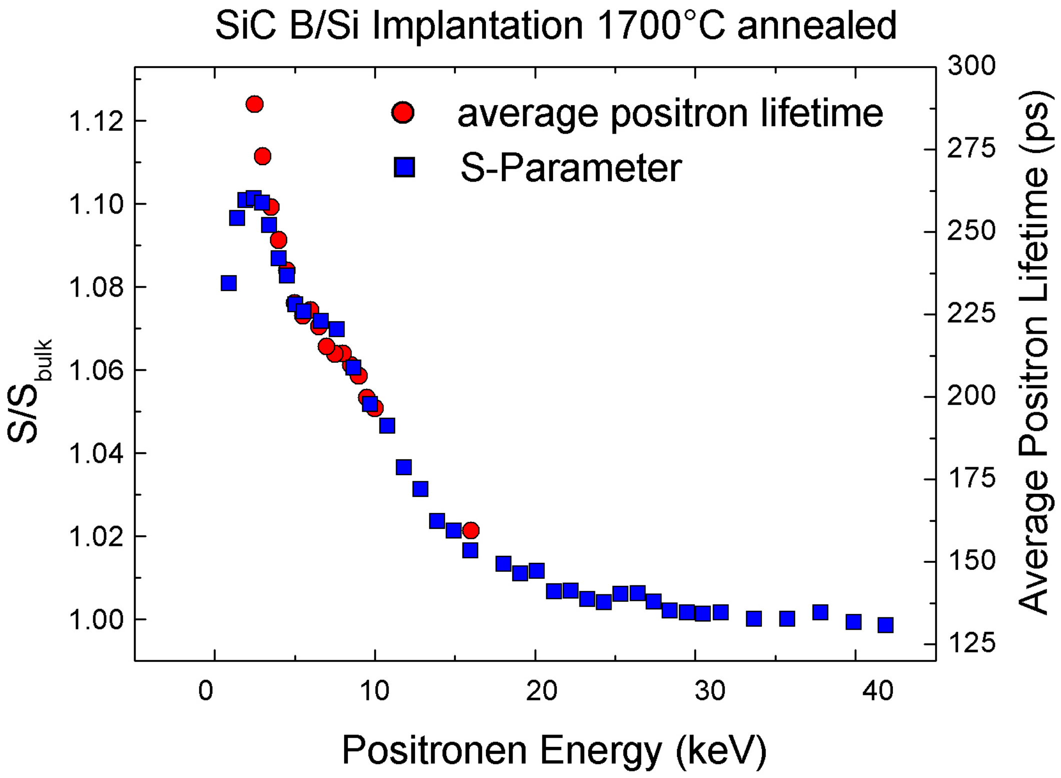


Kögel et al.,
Mat. Sci. Forum 175 (1995) 107

Munich system



Lifetime measurements in SiC layer



F. Redmann, PhD Thesis, 2003

- Si and B coimplantaion into SiC layers on Si
- Average positron lifetime behaves similar to S-parameter
- τ_2 = 300±6 ps → small vacancy cluster defects

Scanning positron microscope

- Variable energy micro-beam of monoenergetic positrons
- Lateral resolution of 2 µm is achieved
- Lifetime measurements at different beam energies are possible

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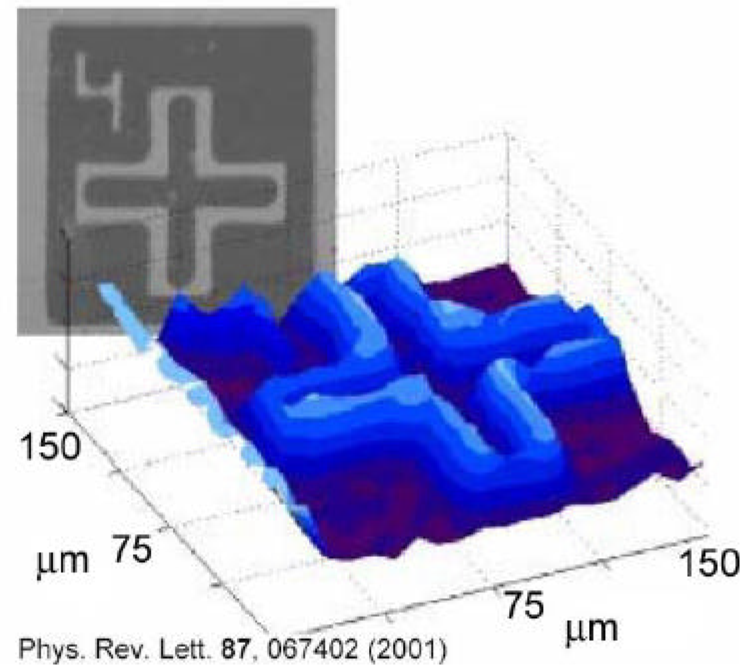
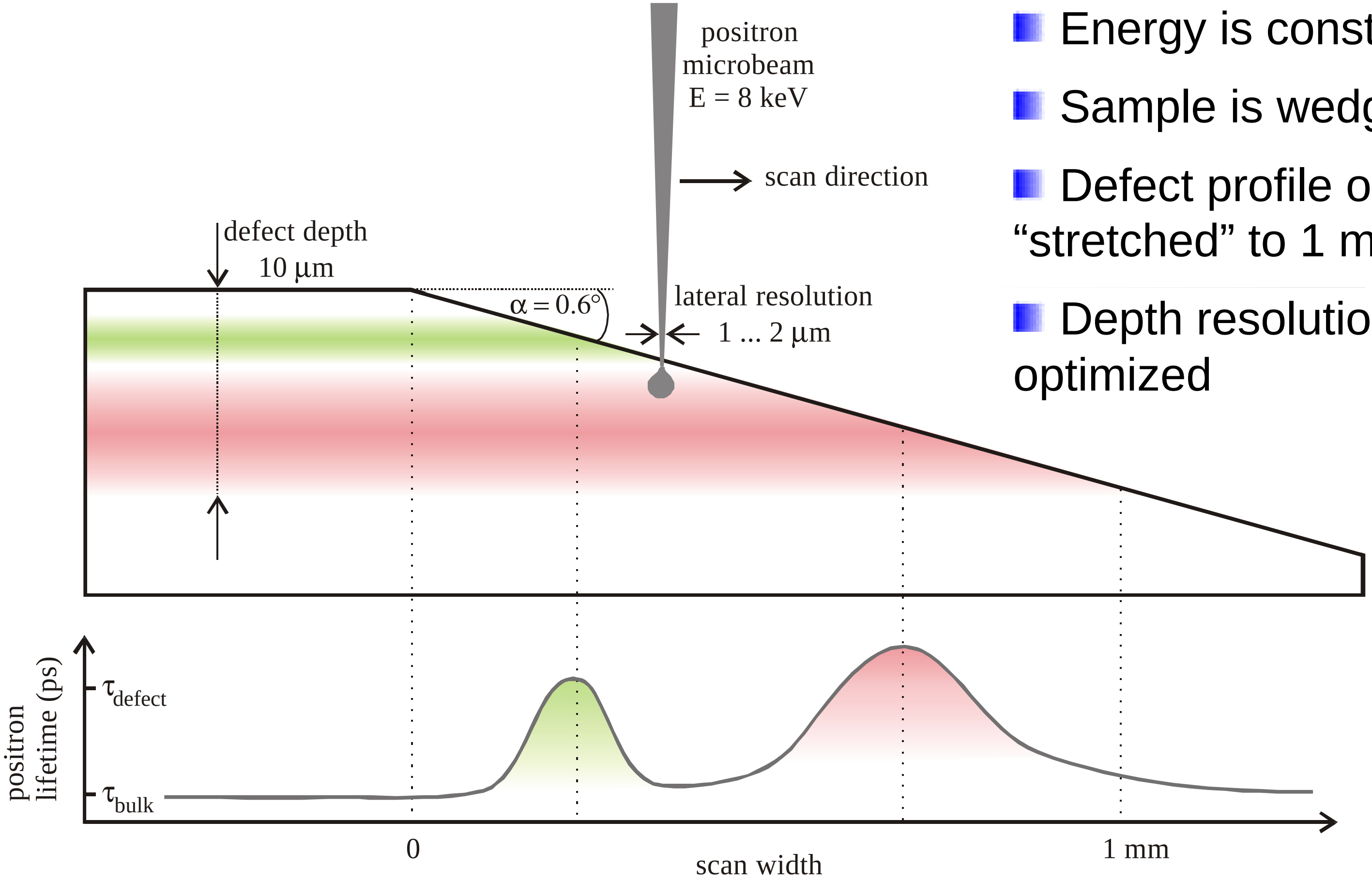
- Principle disadvantage: broad positron implantation profile at high energies



Phys. Rev. Lett. 87, 067402 (2001)

Electron and positron beam image of the surface of a test chip. Light area is SiO_2, dark area is platinum

Depth defect profiling with positron microbeam



- Energy is constant at 8 keV
- Sample is wedged at 0.6°
- Defect profile of 10 μm is "stretched" to 1 mm
- Depth resolution can be optimized

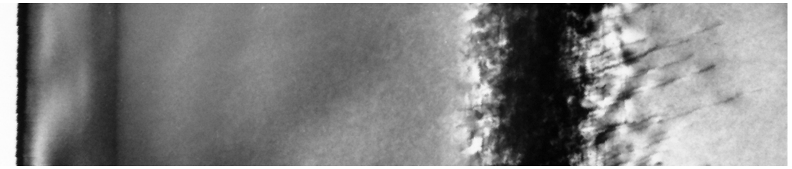
- First time used to study Rp/2 effect in Si after self-implantation

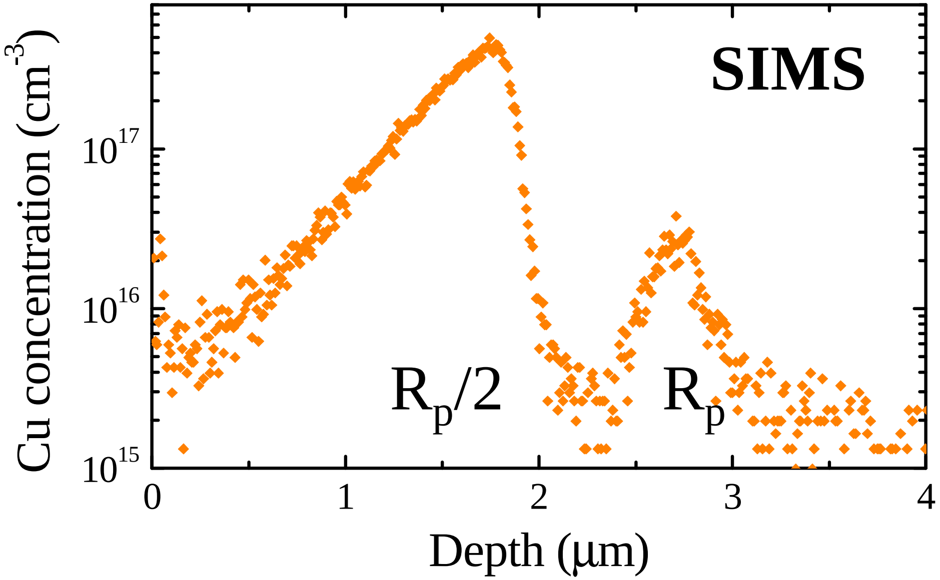
R. Krause-Rehberg et al., Appl. Phys. Lett. **77** (2000) 3932

Defects in high-energy self-implanted Si – The $R_p/2$ effect

- after high-energy (3.5 MeV) self-implantation of Si (5×10^{15} cm^{-2}) and RTA annealing (900°C, 30s): two new gettering zones appear at R_p an $R_p/2$ (R_p – projected range of Si^+)
- visible by SIMS profiling after intentional Cu contamination

TEM image by P. Werner, MPI Halle





- at R_p: gettering by interstitial-type dislocation loops (formed by excess interstitials during RTA)
- no defects visible by TEM at $R_p/2$
- **What type are these defects?**

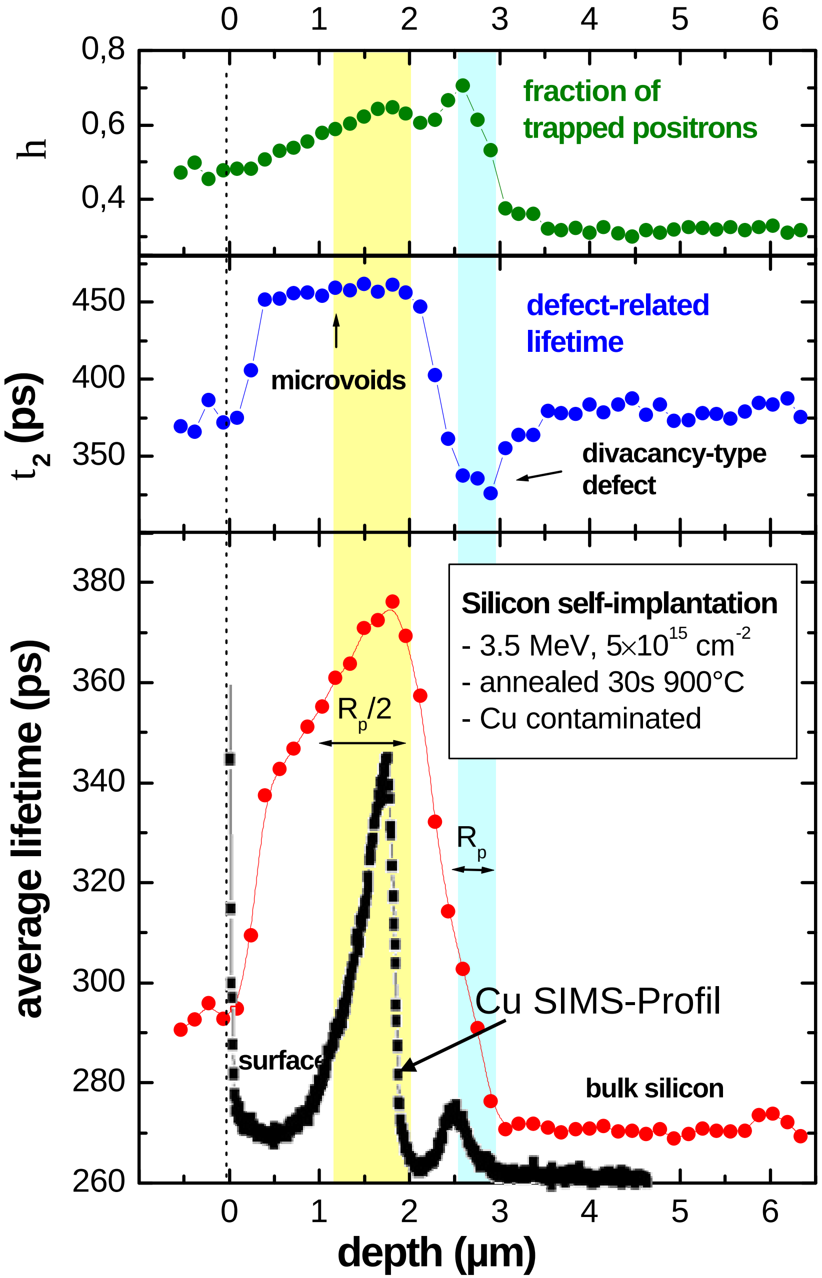
Interstitial type [3,4]

Vacancy type [1,2]

[1] R. A. Brown, et al., J. Appl. Phys. **84** (1998) 2459
[2] J. Xu, et al., Appl. Phys. Lett. **74** (1999) 997
[3] R. Kögler, et al., Appl. Phys. Lett. **75** (1999) 1279
[4] A. Peeva, et al., NIM B **161** (2000) 1090

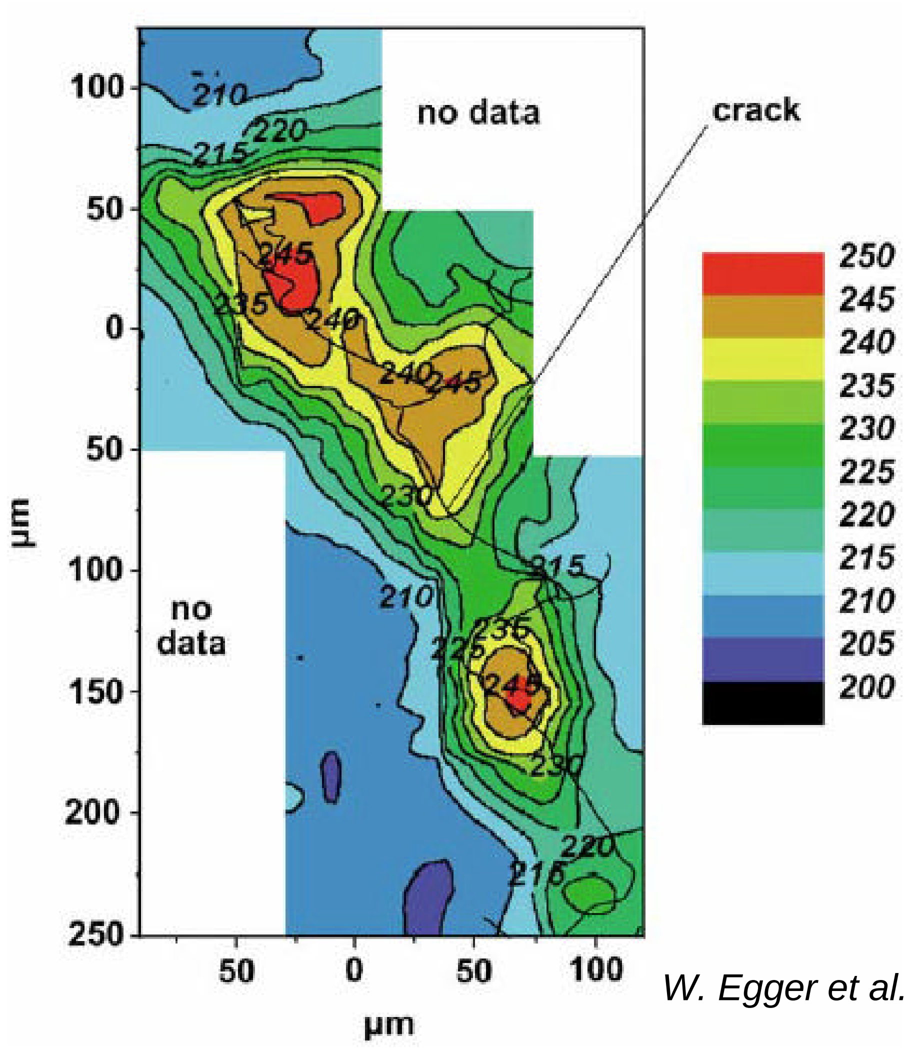
$R_p/2$ effect investigation

- Both defect regions are gut visible
  - vacancy clusters with increasing concentration up to 2 µm ($R_p/2$)
  - in R_p region: lifetime τ_2=320 ps; open volume corresponds to di-vacancy; defects are stabilized by dislocation loops
- very good agreement with the SIMS profile of in-diffused Cu



R. Krause-Rehberg et al., Appl. Phys. Lett. **77** (2000) 3932

Positron lifetime image of fatigue crack with SPM



- Lifetime measurements around a fatigue crack created in technical copper was measured
- e^+ Energy = 16 keV
- spatial resolution about 5 μm
- two lifetimes were observed:
- 190 ps – dislocations
- 360-420 ps – within 40 μm from the crack – vacancy clusters

↓

have been for the first time microscopically observed

W. Egger et al., Applied Surface Sci. 194 (2002)

Conclusion

- positron annihilation is a sensitive tool for investigation of vacancy-like defects in solids
- information on type and concentration of vacancies is received
- thin layers can be studied by mono-energetic positron beam
- improved defect depth profiling is possible by using positron microbeams
- microscopic observation of defects with scanning positron microscope is nowadays possible

This presentation can be found as a pdf-file on our Websites:

http://positron.physik.uni-halle.de
http://PositronAnnihilation.net