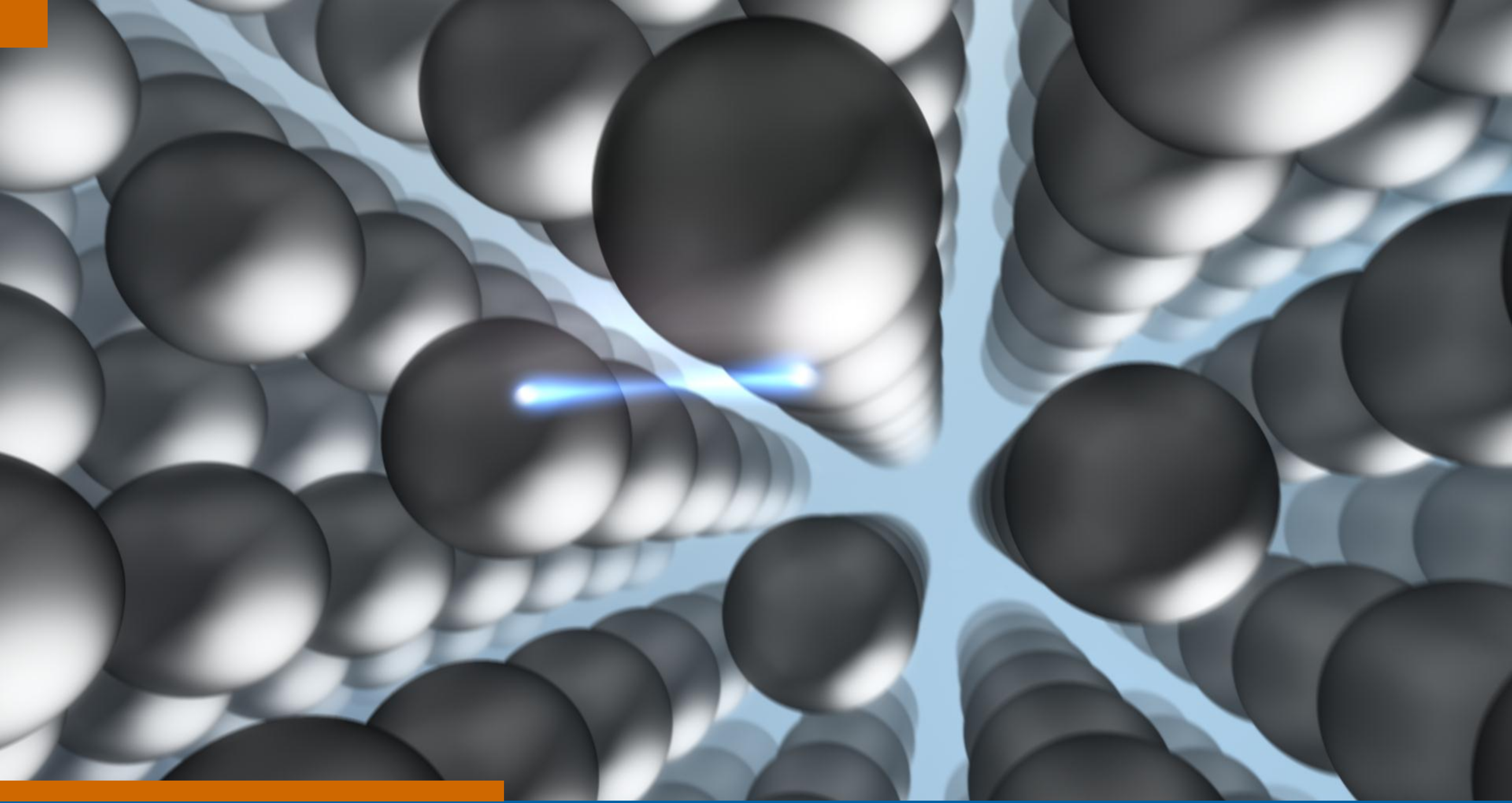


Positron Annihilation Spectroscopy
- A non-destructive method for material testing -

Maik Butterling

HELMHOLTZ ZENTRUM DRESDEN ROSSENDORF

Positron Annihilation Spectroscopy
- A non-destructive method for material testing -

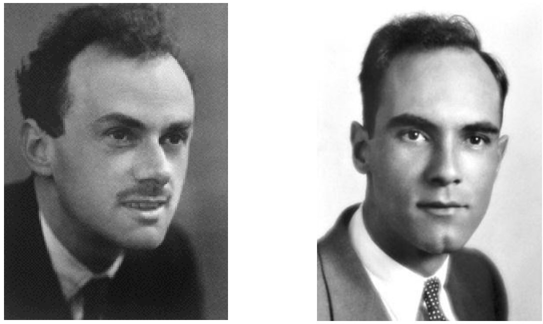
M. Butterling, W. Anwand, K. Potzger, A. Wagner

- Basics about Positron Annihilation Spectroscopy (PAS)
  - The Positron
  - Utilitiy of positrons for spectroscopy
- Measurements
  - Positron lifetime
  - Doppler broadening
  - Depth-resolved defect profiling
- Technical hints of PAS
- First measurements within the DETI.2 project

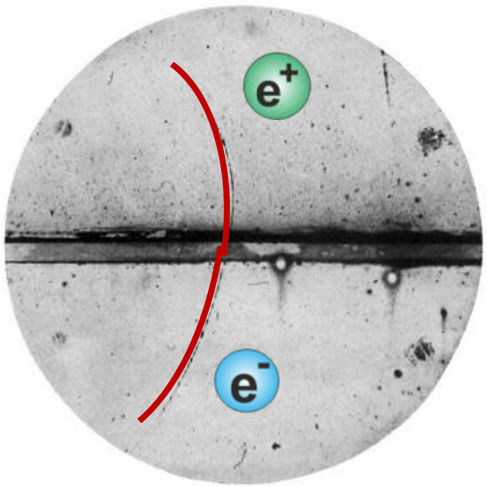
The Positron

antiparticle of the electron:

- same mass m_e
- spin ½
- opposite electric charge $+e$
- annihilation with an electron by emitting photons



P.A.M Dirac (1928) and C.D. Anderson (1932)



MARCH 15, 1933 PHYSICAL REVIEW VOLUME 43

The Positive Electron

CARL D. ANDERSON, *California Institute of Technology, Pasadena, California*

(Received February 28, 1933)

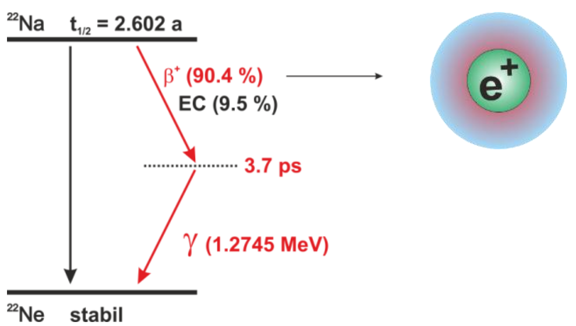
Out of a group of 1300 photographs of cosmic-ray tracks in a vertical Wilson chamber 15 tracks were of positive particles which could not have a mass as great as that of the proton. From an examination of the energy-loss and ionization produced it is concluded that the charge is less than twice, and is probably exactly equal to, that of the proton. If these particles carry unit positive charge the curvatures and ionizations produced require the mass to be less than twenty times the electron mass. These particles will be called positrons. Because they occur in groups associated with other tracks it is concluded that they must be secondary particles ejected from atomic nuclei.

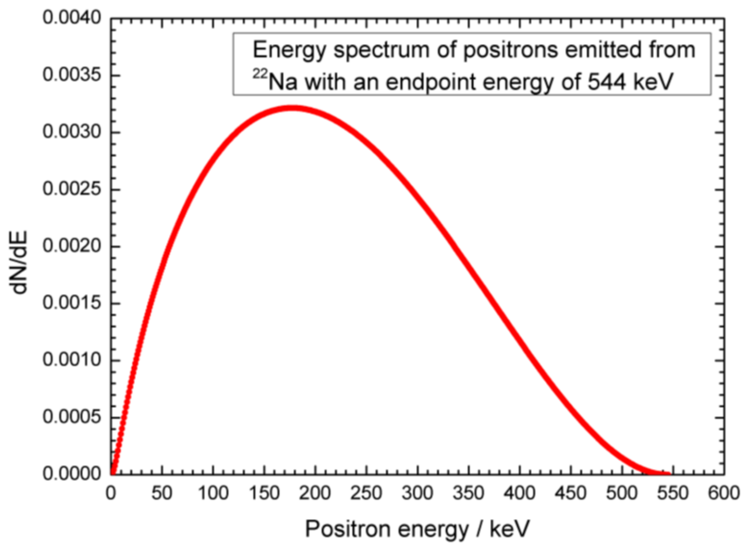
Editor

The Positron

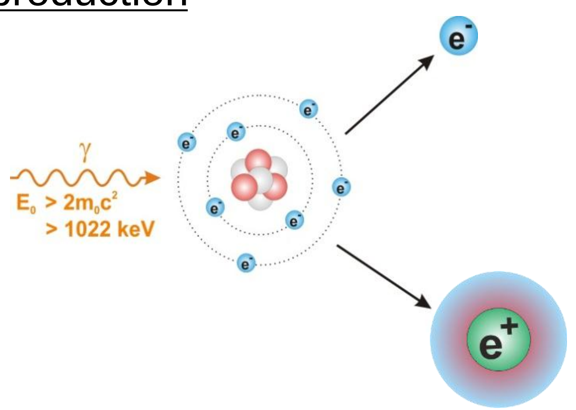
Generation methods

β^+ decay of ^{22}Na





pair production



$$E_{gamma} \longrightarrow (m_{elec}c^2 + E_{kin,positron}) + (m_{pos}c^2 + E_{kin,electron})$$

$$E_{gamma} \geq 2 \cdot m_0 c^2 \geq 2 \cdot 511\ \text{keV}$$

The Positron

Thermalization and Diffusion

Thermalization

- energy transfer to target atoms/molecules via inelastic scattering
- within a few ps
- leads to an energy dependent penetration depth profile

- in metals: excitation of conduction electrons
- in semiconductors: excitation of electron-hole pairs with E > bandgap width

Diffusion

- behaviour of charged particles
- repelled from the nuclei
- largest position probability in interstitial regions

Utility of positron for spectroscopy

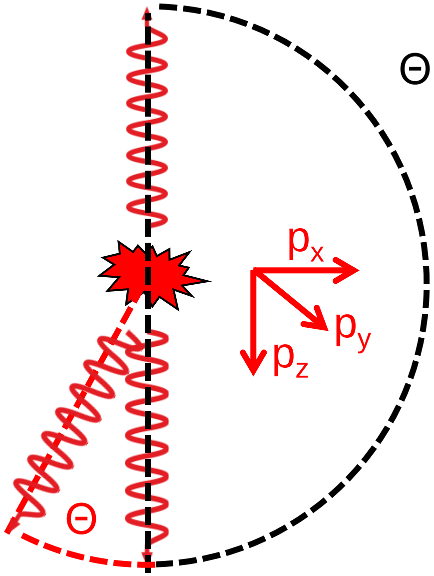
Annihilation of positrons – information from annihilation photons

$$\Delta E = \frac{(p_z^{el} + \cancel{p_z^{pos}}) \cdot c}{2}$$

Positron is thermalized:

$E_{pos} = k_B T$
~ 26 meV at 300 K
<< E_{el}

$E = m_0c^2 - \Delta E$



$\Theta = 180° + \Theta_{x,y}$

$E = m_0c^2 + \Delta E$

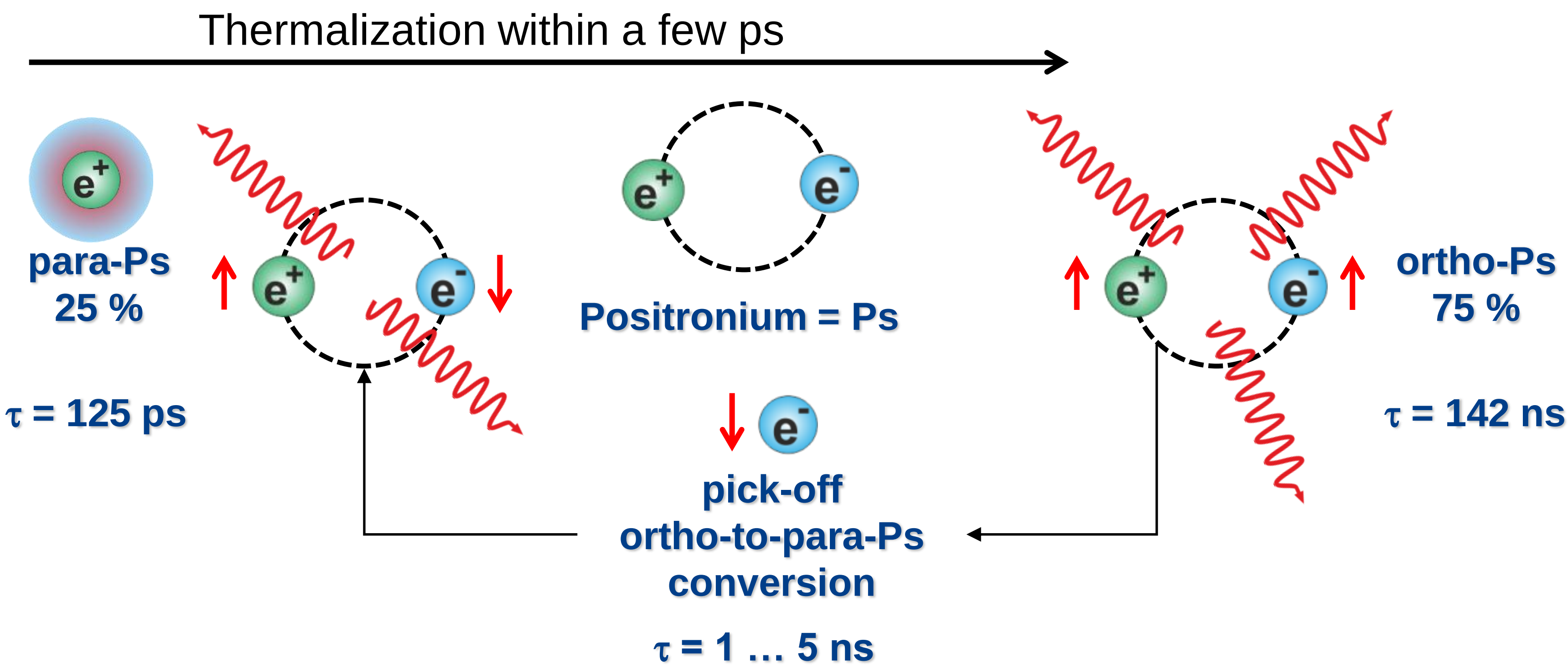
$$\theta_{x,y} = \frac{p_{x,y}^{el}}{m_0 c}$$

E_{el} = 10 eV
→ ΔE = 1.6 keV
→ Φ = 6 mrad

- electron momentum influences energy and emission angle of annihilation photons

The Positron

Bound states

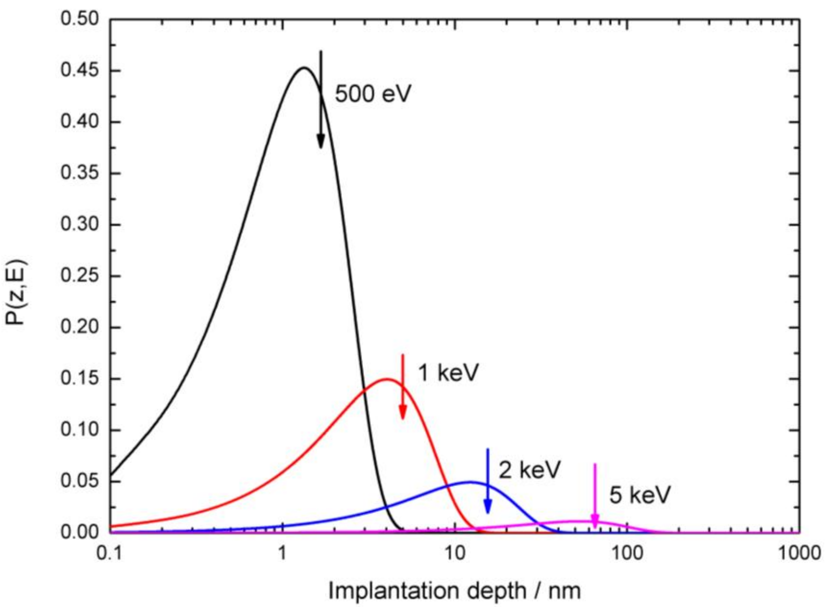


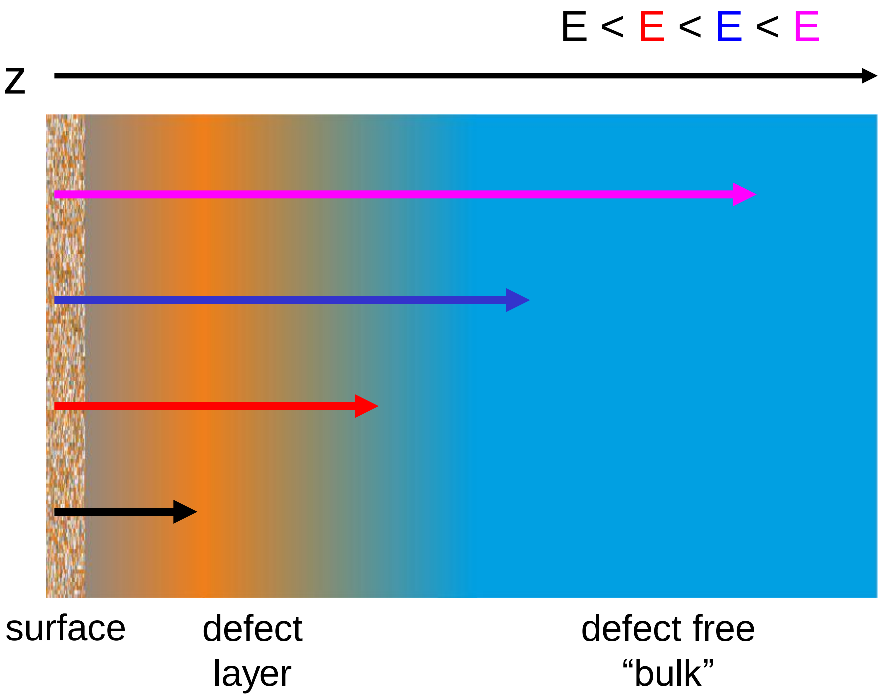
Ore (1949) „Positronium generation in solids if“:

$$\Delta E_{pos} = E_{max} - E_{min} = E_{excite} - (E_{ion} - 6.8\ \text{eV})$$

Utility of positron for spectroscopy

Implantation profiles for solids





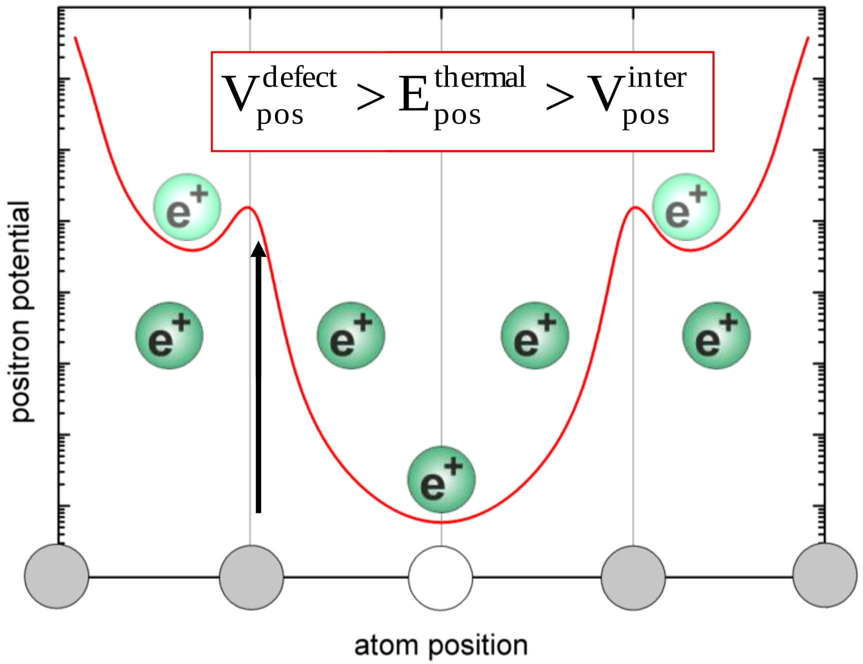
$$\bar{z} = \frac{A \cdot E^{r}}{\rho}$$

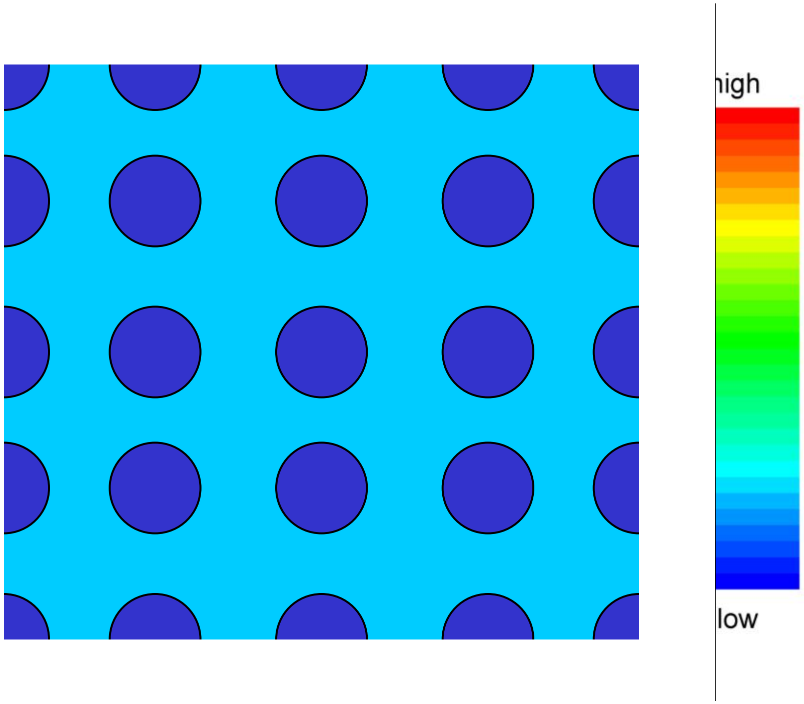
mean positron implantation depth (with empirical parameters A and r)

- Implantation profile (Makhovian profile) is result of the thermalization process
- smearing with increasing energy: limit in energy necessary

Utility of positron for spectroscopy

Trapping in negatively charged defects





- missing positive repelling charge
- reduction of the ground potential for positrons

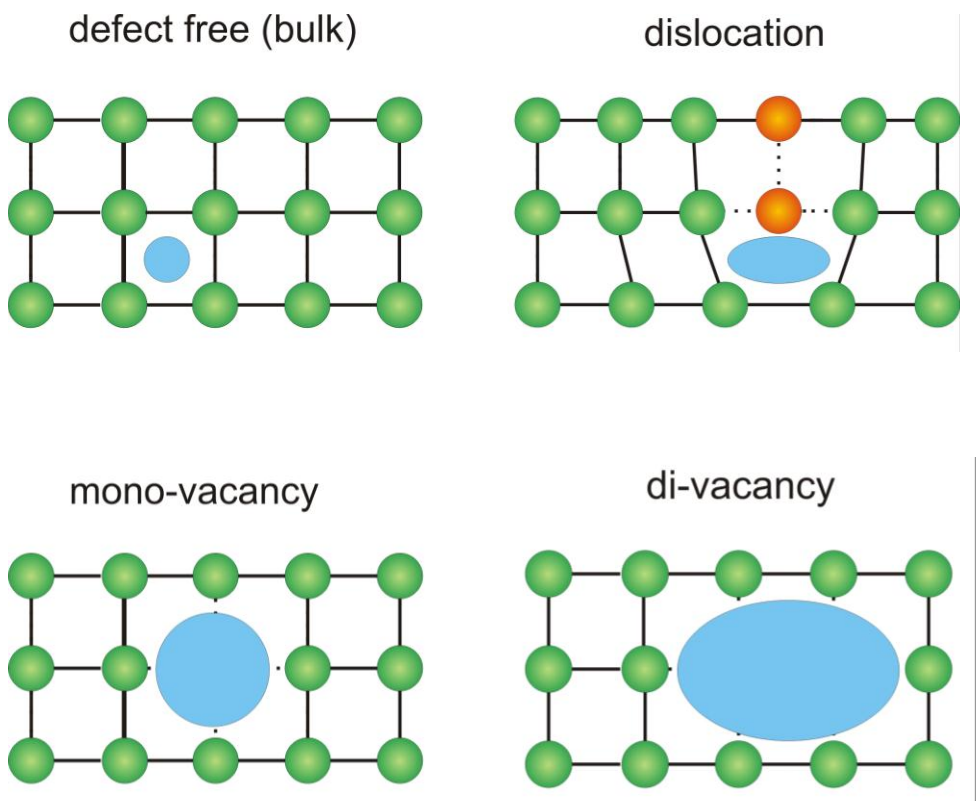
→ trap for positrons

→ positrons are suitable for detecting atomic defects

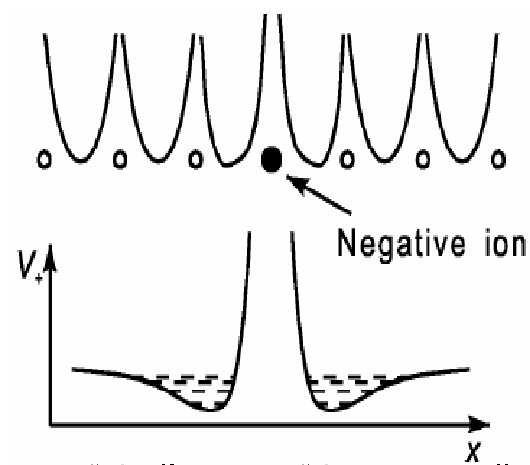
- positive vacancies repell positrons

Utility of positron for spectroscopy

Varity of defects



shallow trap

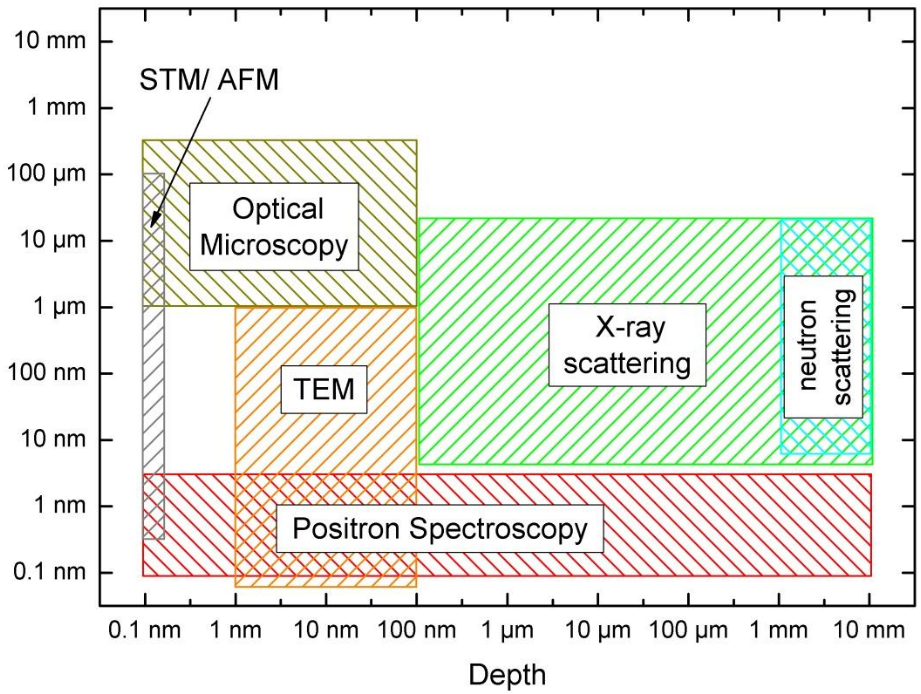


a “shallow trap“ has a small positron binding energy

- acceptor-type impurities
- dopants (p doped Si)
- negative antisite defects Si_{Ga}

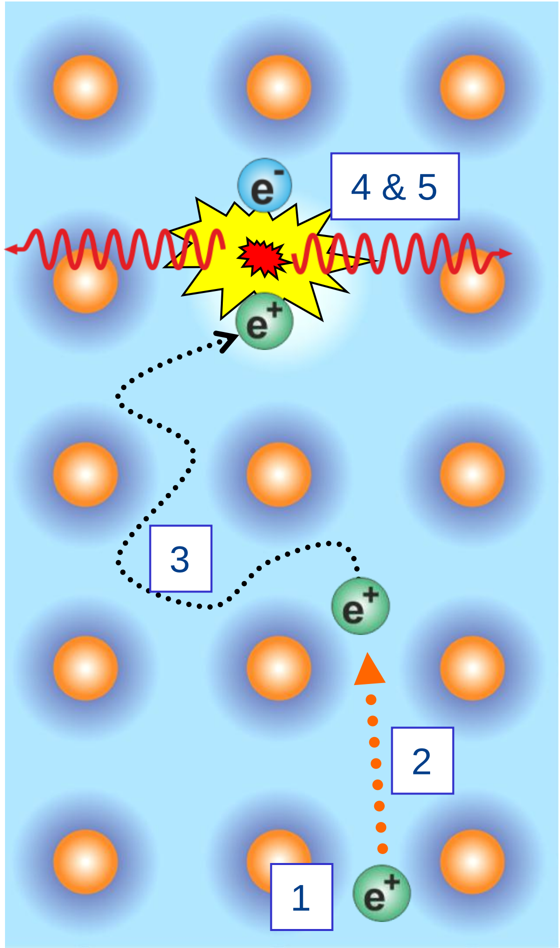
Krause-Rehberg, Positron annihilation in semiconductors,Springer Verlag 1999

Utility of positron for spectroscopy



- non destructive method
- sensitive to atomic defects (even single dislocations or mono-vacancies are detectable)
- lowest concentrations detectable: 1 vacancy per 10^6 … 10^3 atoms
- elemental sensitivity
- depth profiling possible

Summary – Fate of positrons in solid matter



lattice of a solid with a single vacancy

1) Positron generation & implantation in the solid
- β^+ decay of ^{22}Na
- pair production

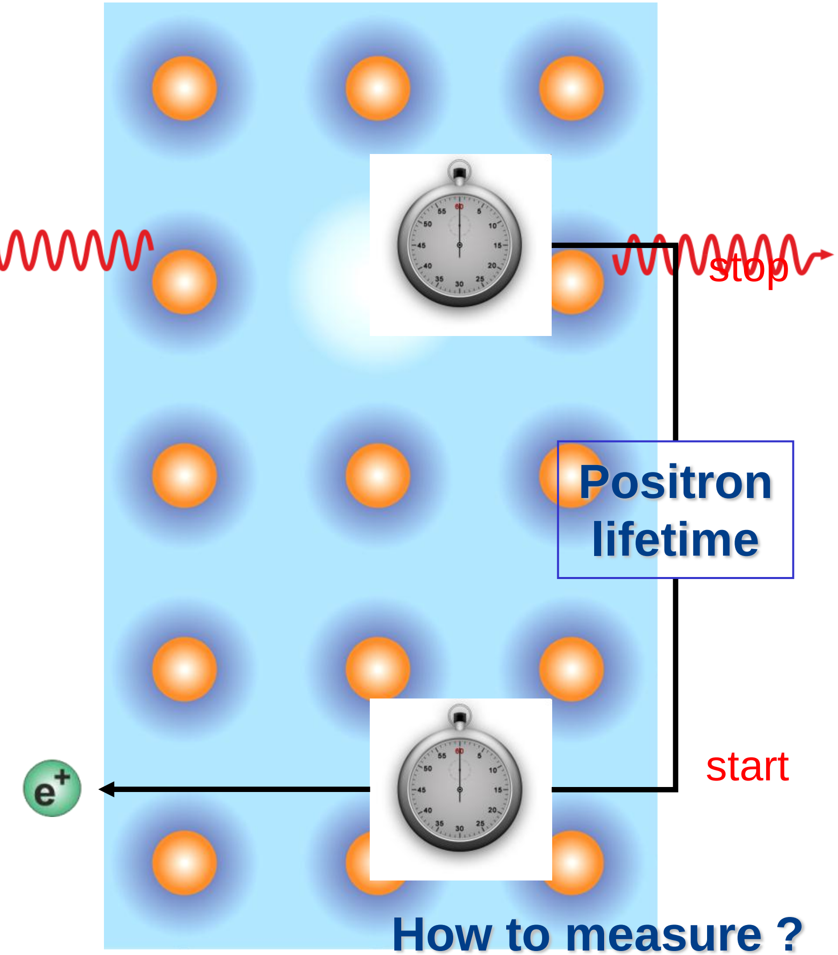
2) Thermalization – reducing energy
- ~ 10 ps

3) Diffusion through the lattice
- ~ 100 nm

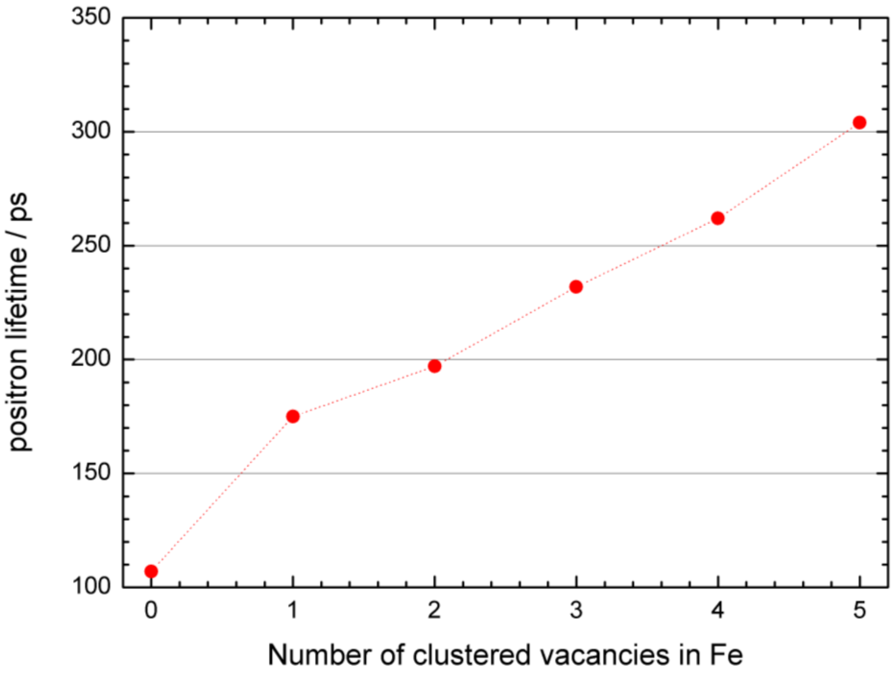
4) Trapping in defects

5) Annihilation with an electron
- emission of two photons in metals/ semiconductors
- angle and energy depend on momentum of electron

Measurement – positron lifetime



How to measure ?

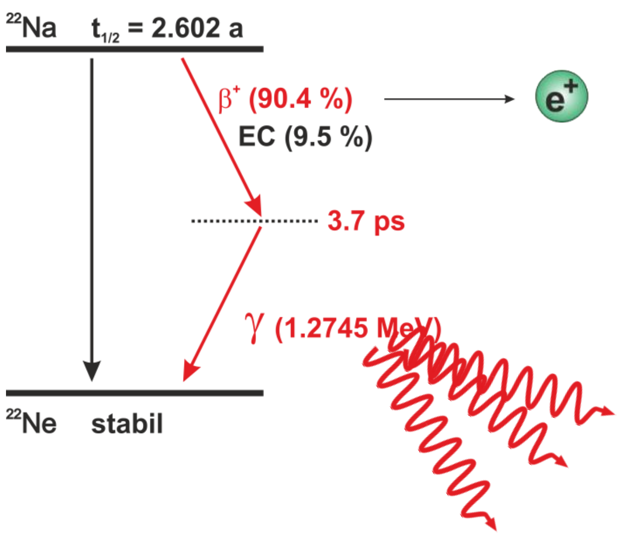


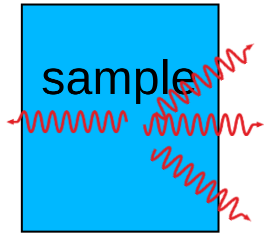
identification and concentration of open-volume defects

DRESDEN concept HZDR

Measurement – positron lifetime

Start signal – with radio-isotope ^{22}Na



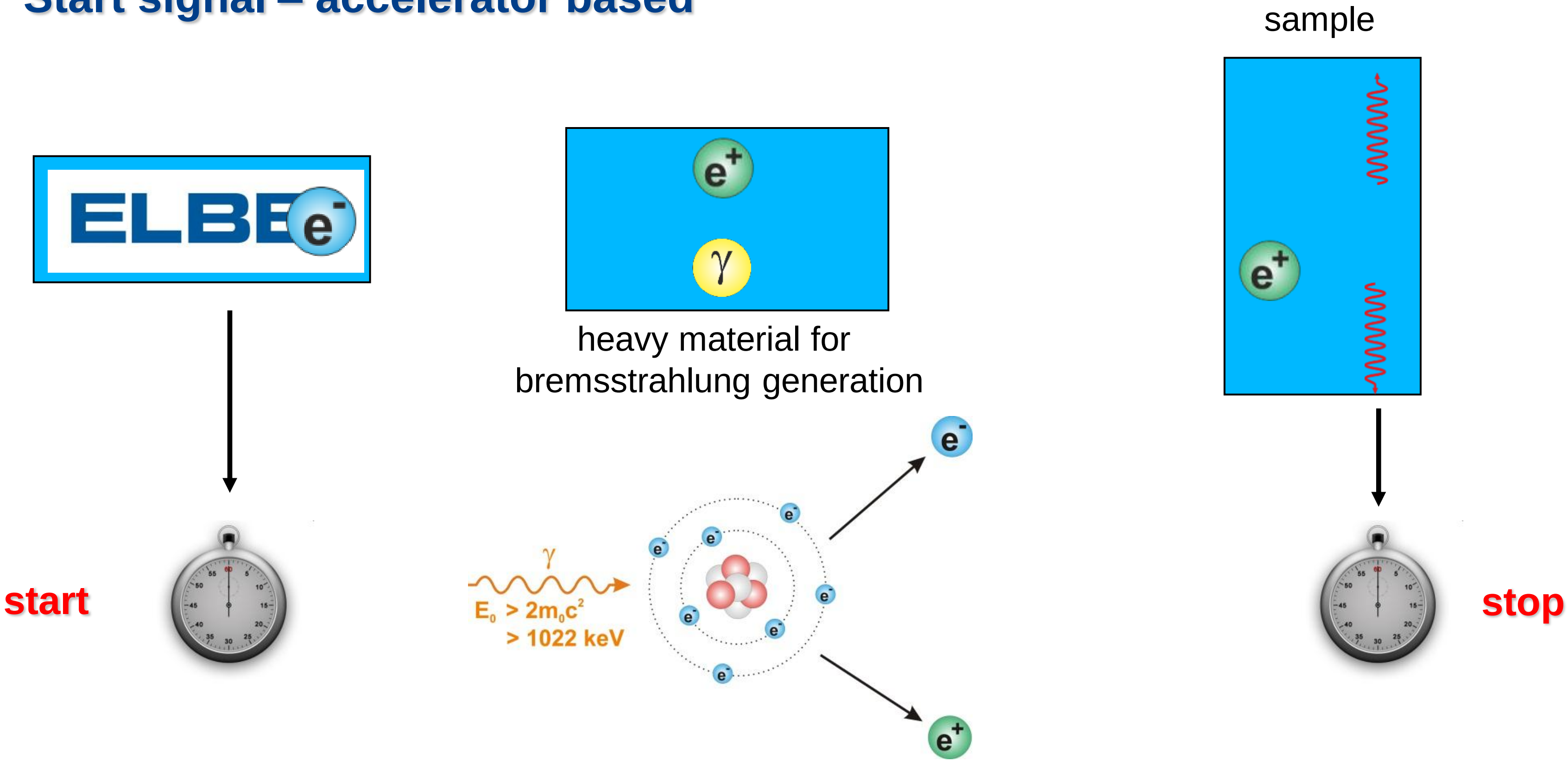


Problem: low probability of detecting both rays

Solution: increase efficiency of start signal

Measurement – positron lifetime

Start signal – accelerator based



condition: electron pulse short in time → sharp time signal for the start

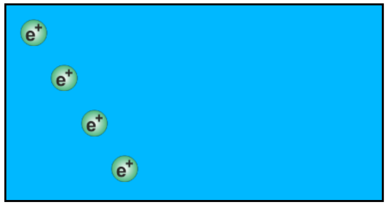
at ELBE: ~ 5 ps width → possible to use

Measurement – positron lifetime

Positron generation methods and consequences

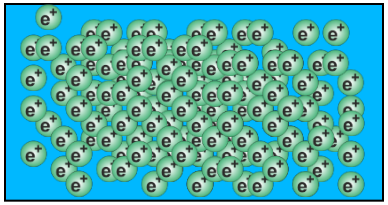
Implantation of positrons

- \+ adjustable energy → adjustable implantation depth
- – limited implantation depth of a few µm

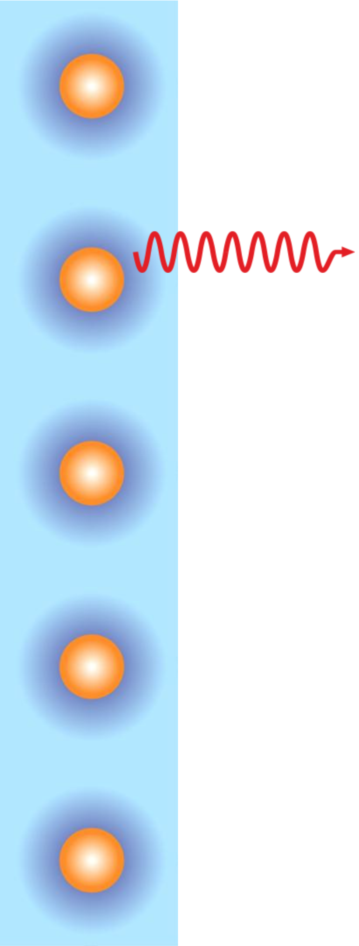


Pair production inside sample

- \+ information from the entire sample volume
- – no depth information

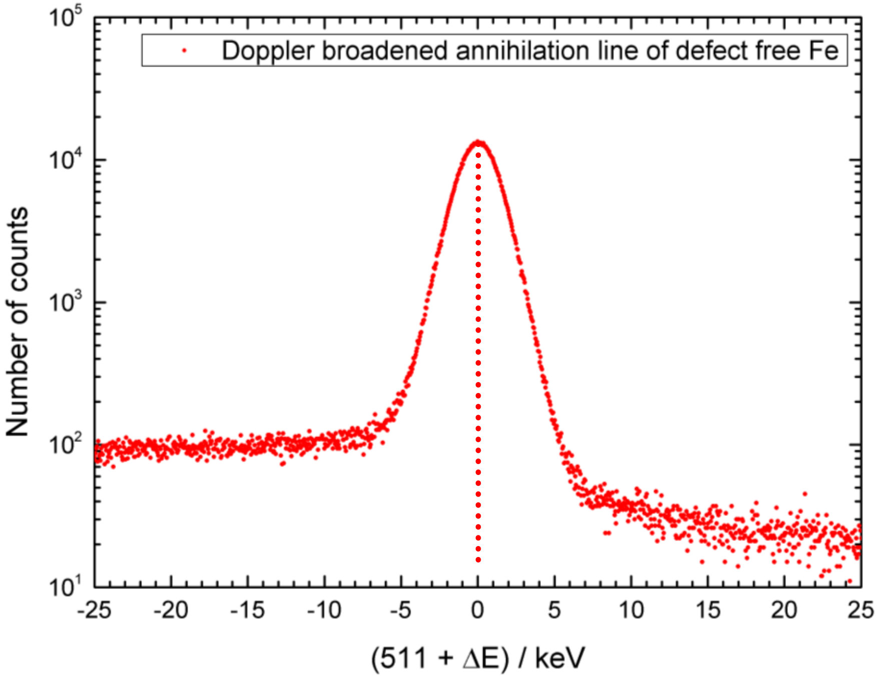


Measurement – Doppler broadening



Doppler broadening

energy deviation from 511 keV



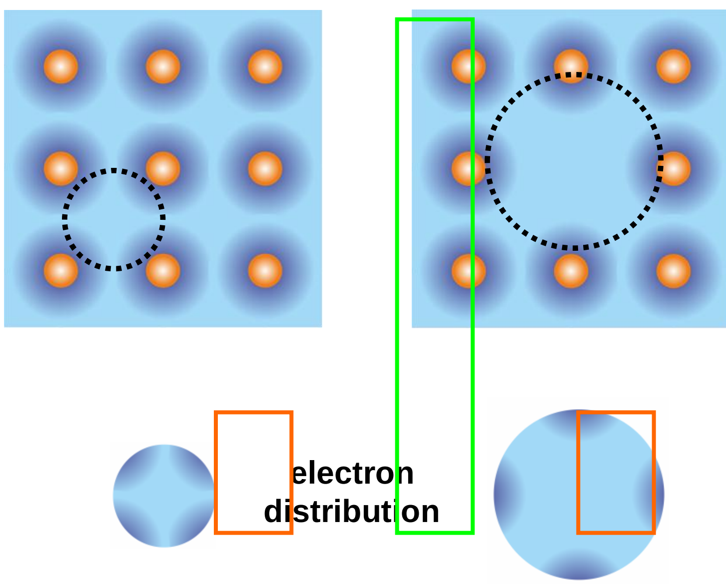
Doppler broadening of the 511 keV line due to the kinetic energy of the annihilated electron (positron is in the ground state)

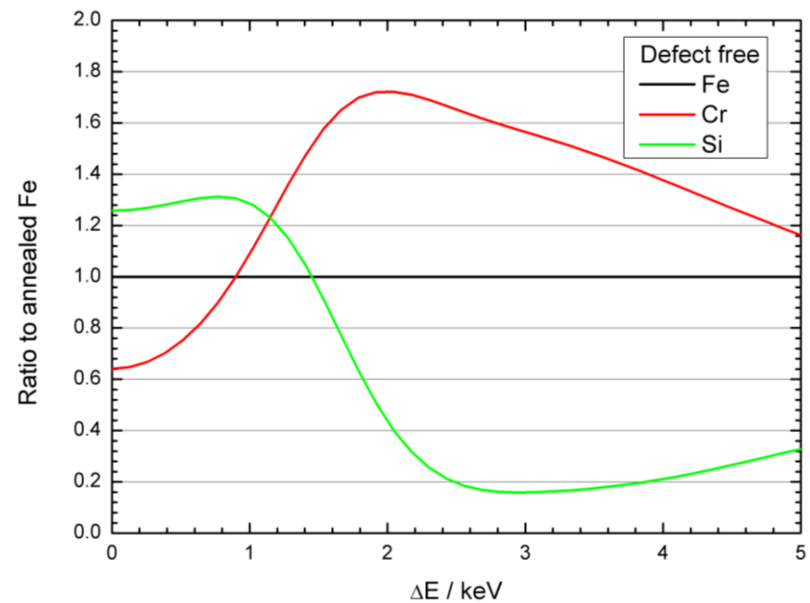
Example: E_{kin} = 10 eV ⟶ ΔE = 1.6 keV

DRESDEN concept HZDR

Measurement – Doppler broadening

From electronic structure to the defect situation





core | valence

$$S = \frac{A_{Peak}}{A_{All}}$$

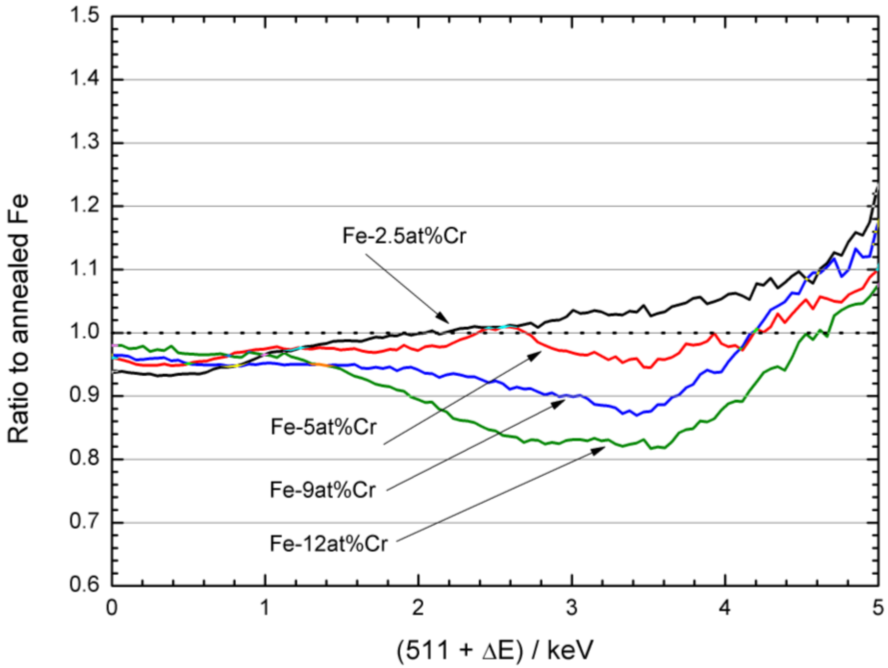
sensitive to size and concentration of open-volume defects

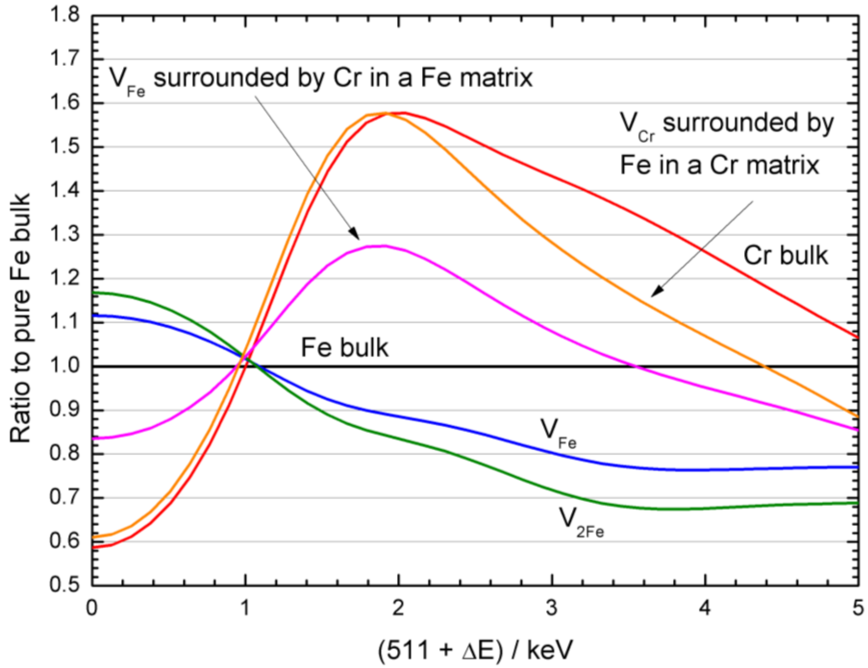
$$W = \frac{W_1 + W_2}{W_{All}}$$

sensitive to the chemical surrounding (elements) of the annihilation site

Measurement – Doppler broadening

Chemical surrounding investigated by positrons

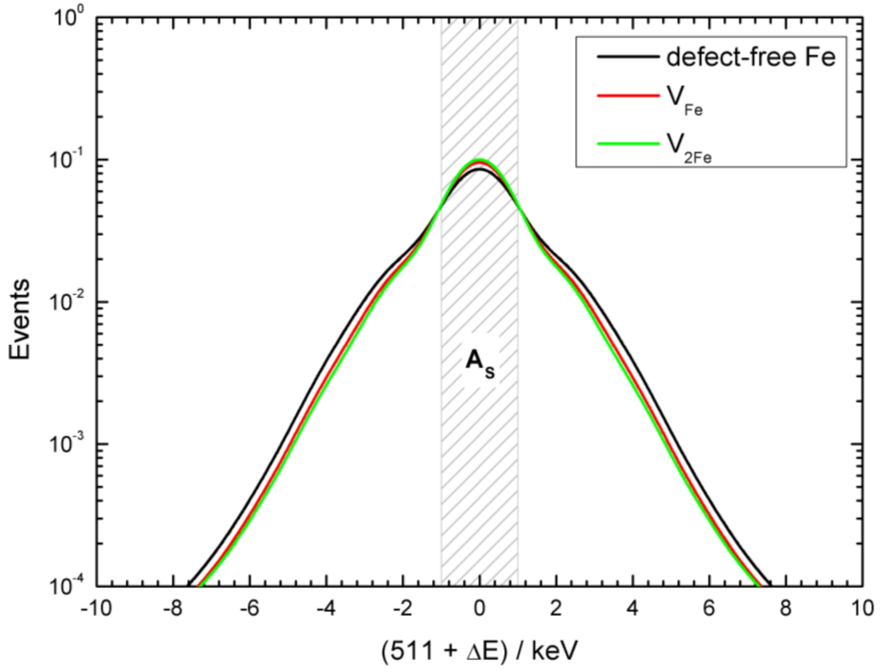




- α` phase with Cr rich precipitates for more than 9% Cr
- Cr precipitates repel vacancies
- Cr content < 9%: V_{Fe} and V_{Fe} - Cr defects
- Cr content > 9%: α` phase → V_{Fe} and Cr precipitates (invisible)

Depth-resolved defect profiling

Calculation of S for series

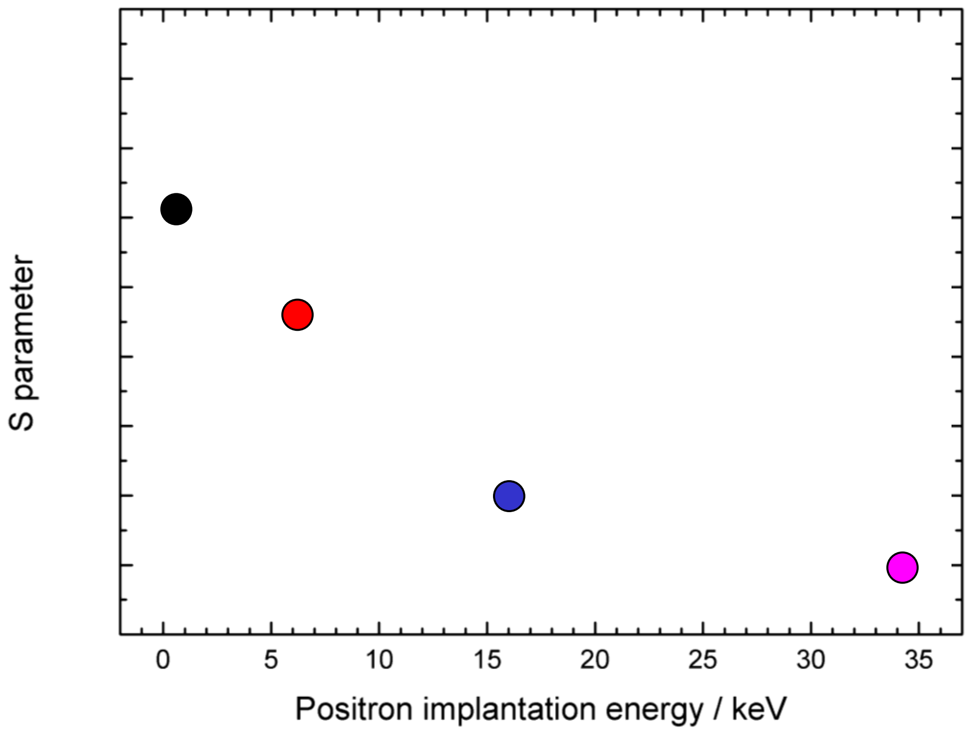


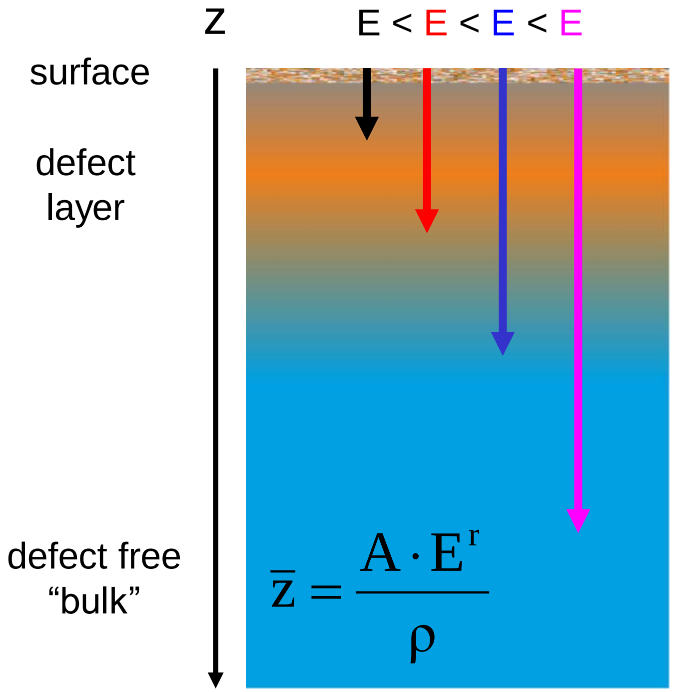
$$S = \frac{A_S}{A_{All}}$$

- S ~ 0.5 for the reference
- same limits then for each following annihilation line
- (set reference parameter as 1)
- relative changes in S visible for different implantation positron energies/ implantation depths

Depth-resolved defect profiling

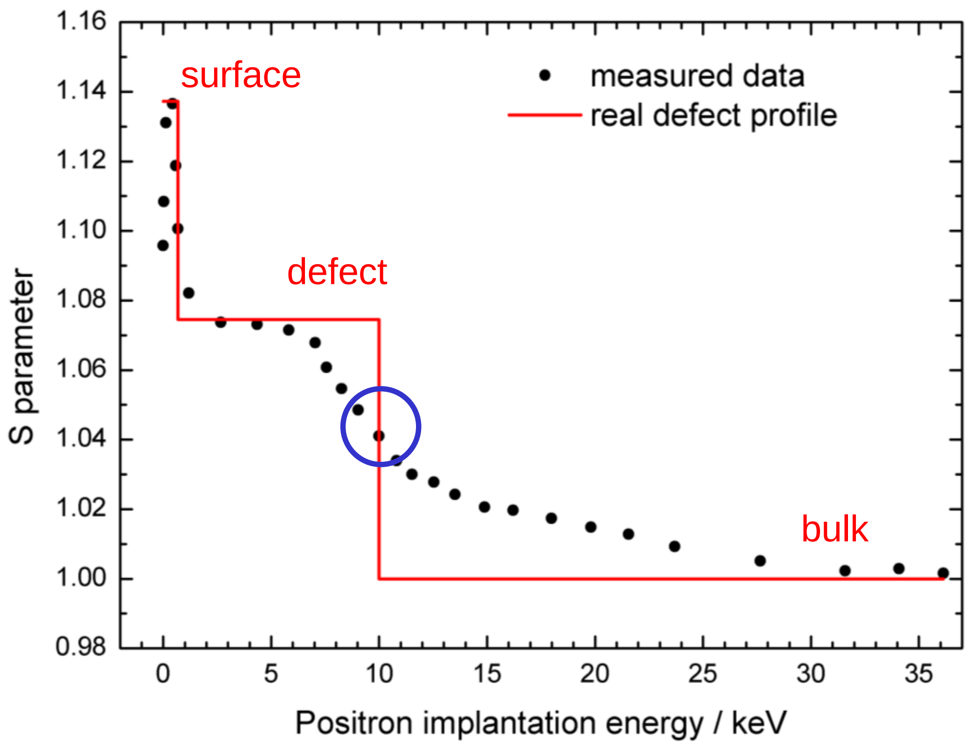
Depth-resolved S parameter

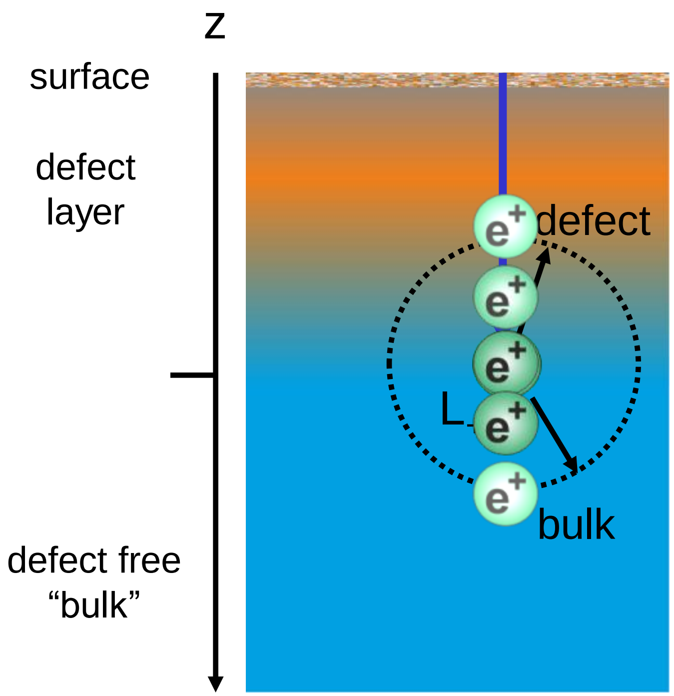




Depth-resolved defect profiling

Smearing of profile information – thermalization & diffusion



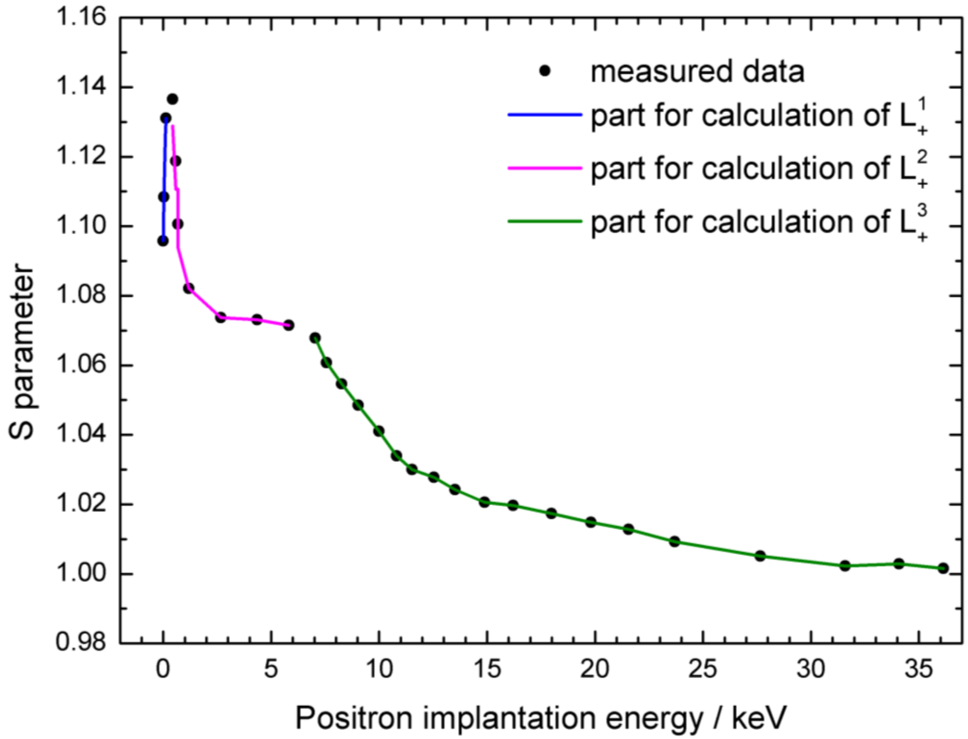


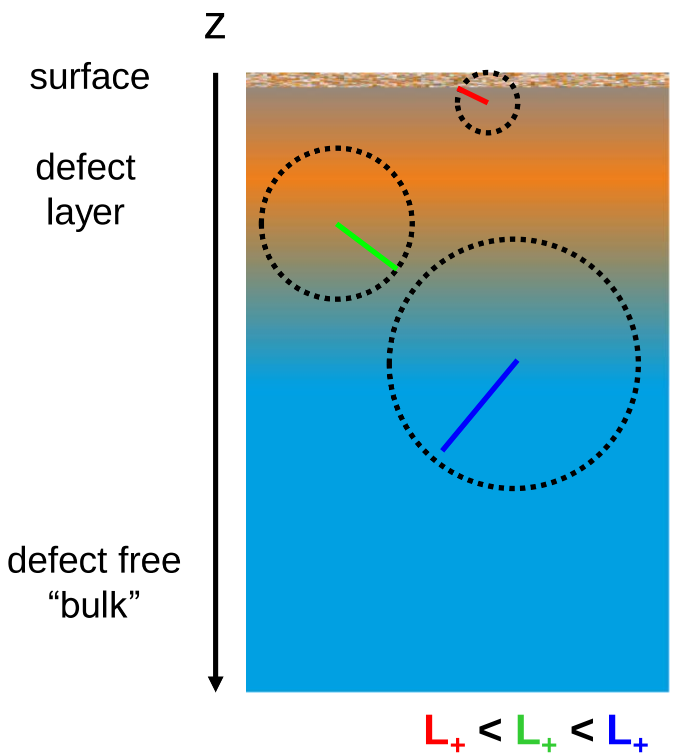
smearing due to
implantation profile

$$S = m \cdot S_{defect} + (1 - m) \cdot S_{bulk}$$

Depth-resolved defect profiling

Calculation of the diffusion length L_+

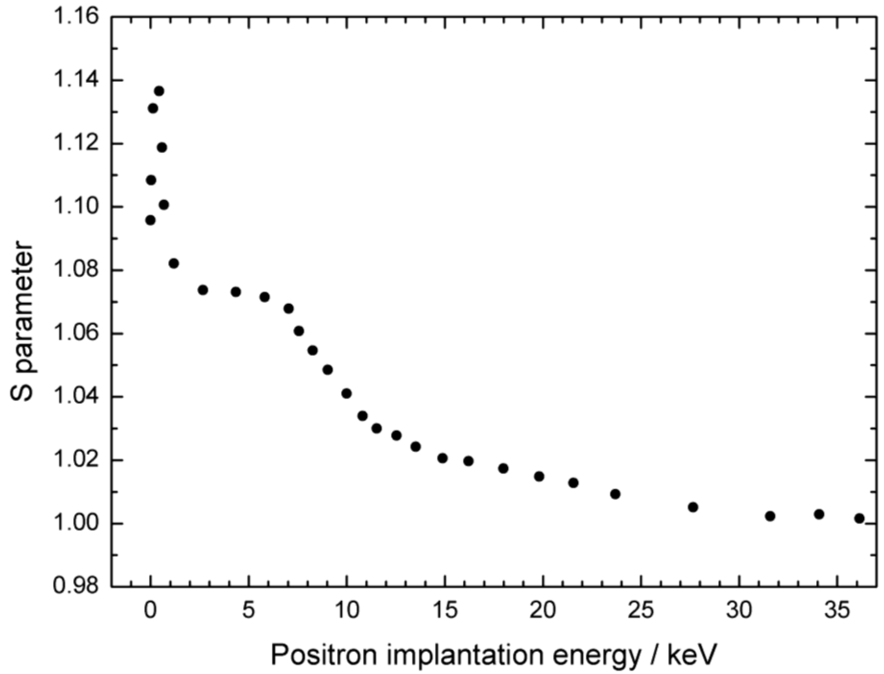


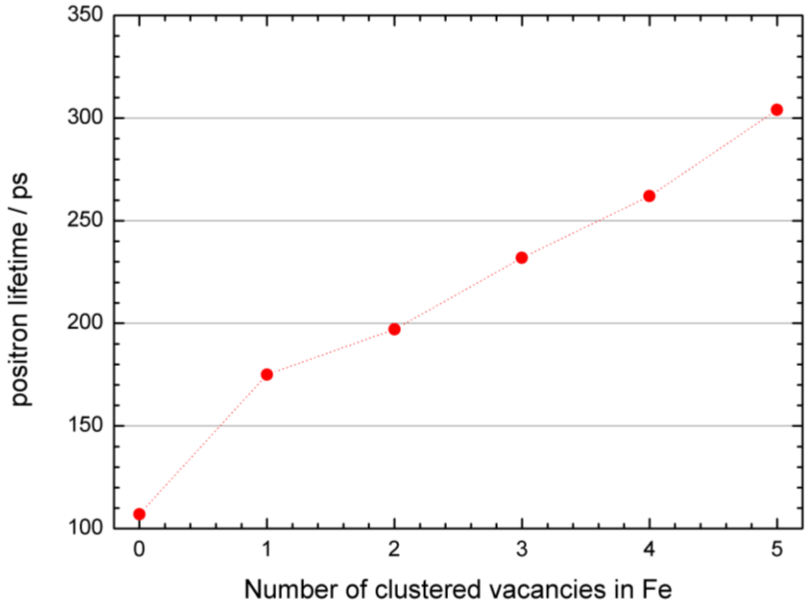


VEPFIT:
A. van Veen *et al.*, Analysis of positron profiling data by means of "VEPFIT", Positron beams for solids and surfaces, P.J. Schultz *et al.*, Amer. Inst. Phys., NY (1990) 171-196.

Depth-resolved defect profiling

Identification of defects – depth-resolved positron lifetime measurement

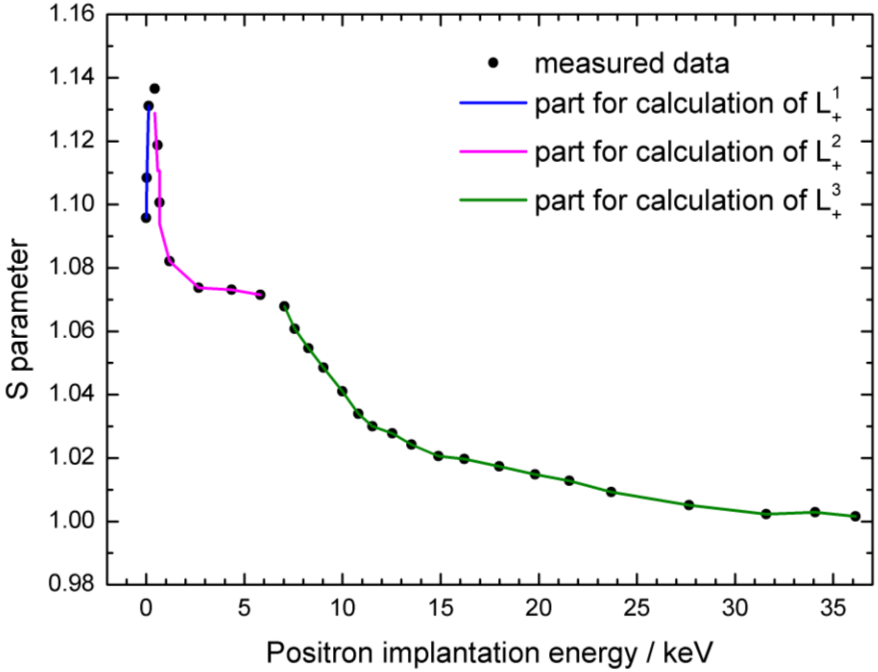


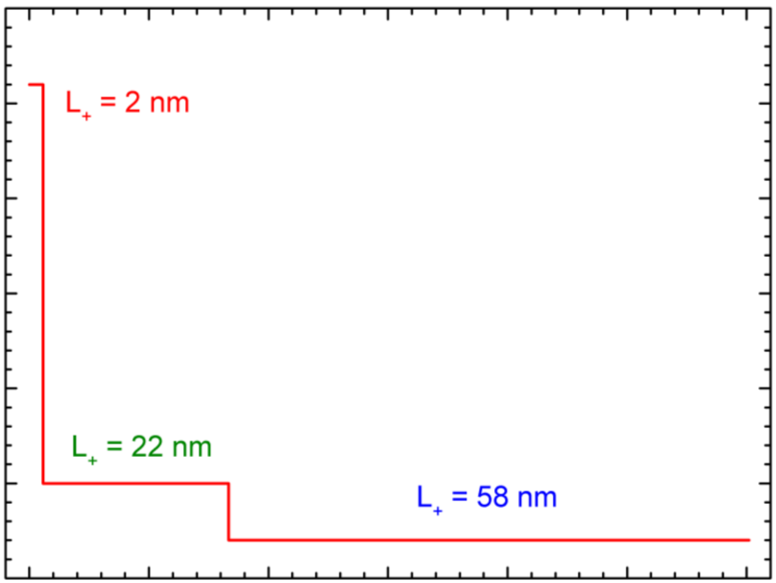


- identification of defect type via positron lifetime
- difficulty: availability of setups for depth-resolved positron lifetime

Depth-resolved defect profiling

S → L_+ → lifetime → defect profile



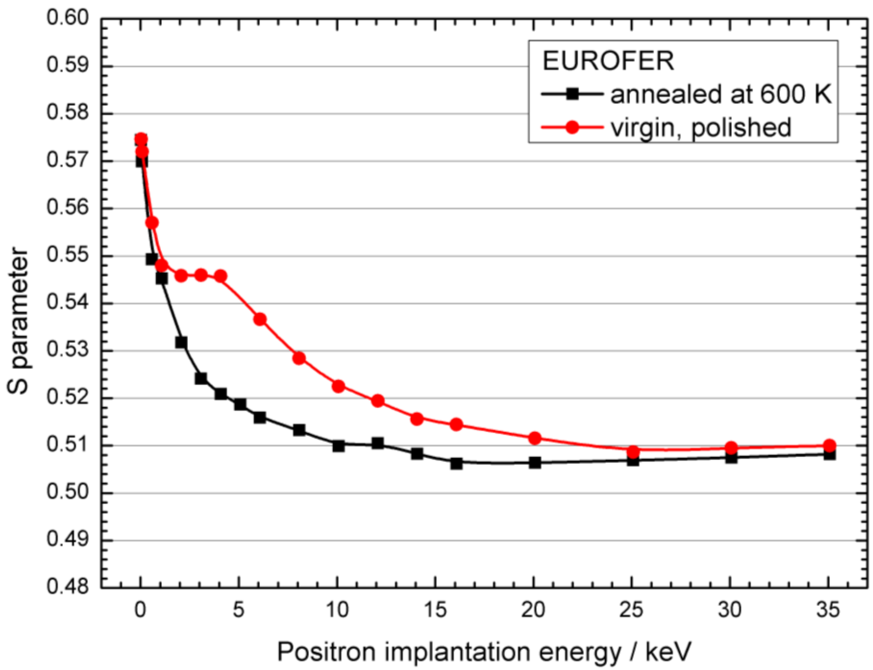


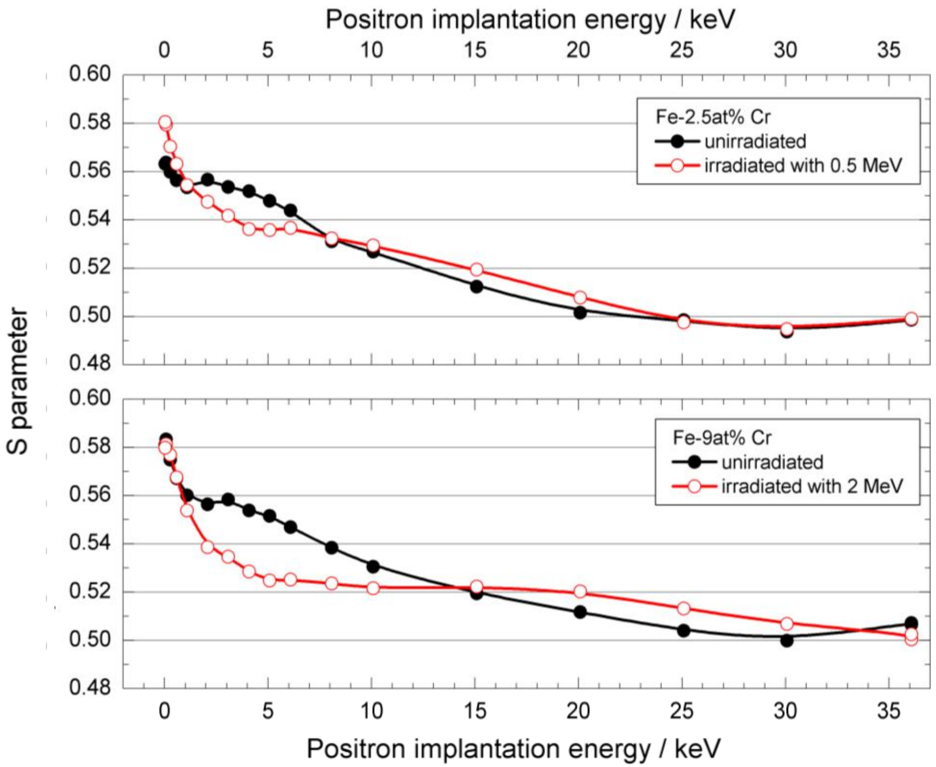
$$\bar{z} = \frac{A \cdot E^r}{\rho}$$

after Grynszpan *et al.*, Ann. Chim. Sc. Mat 32(4) (2007)

Depth-resolved defect profiling

Sensitivity of positrons





- positrons are sensitive to surface treatment (defects induced by polishing)
- temperature during ion implantation leads to annealing of defects

Technical hints of PAS

Source activity and positron lifetime measurements

positron source

e^+ e^+ e^+

correlation between not related events

sample

usage of ^{22}Na for metals:

$$A_{max} \sim \frac{1}{8 \cdot \tau_{max}^{estimated}} [Bq]$$

start

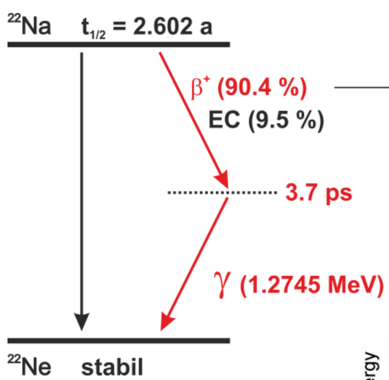
stop

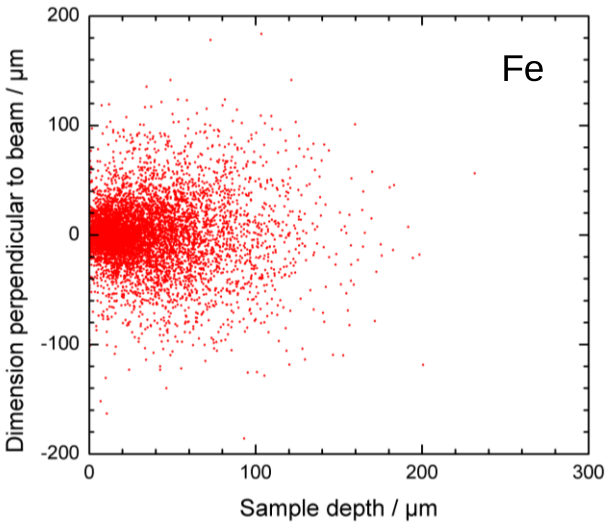
$\tau \sim 5$ ns → $A_{max} \sim 25$ MBq ~ 0.67 mCi

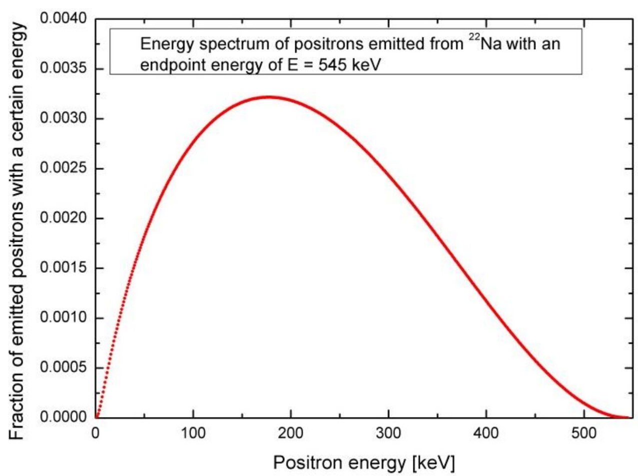
Valid event for positron lifetime

Technical hints of PAS

Usage of non-monoenergetic positrons





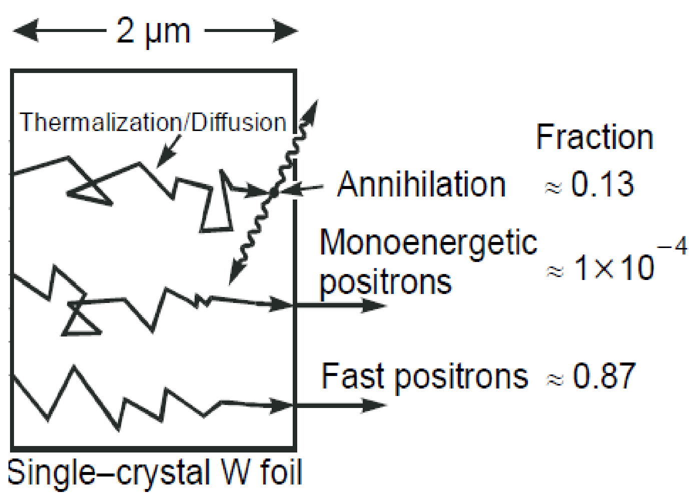


depth profiling becomes impossible without modification of energy !

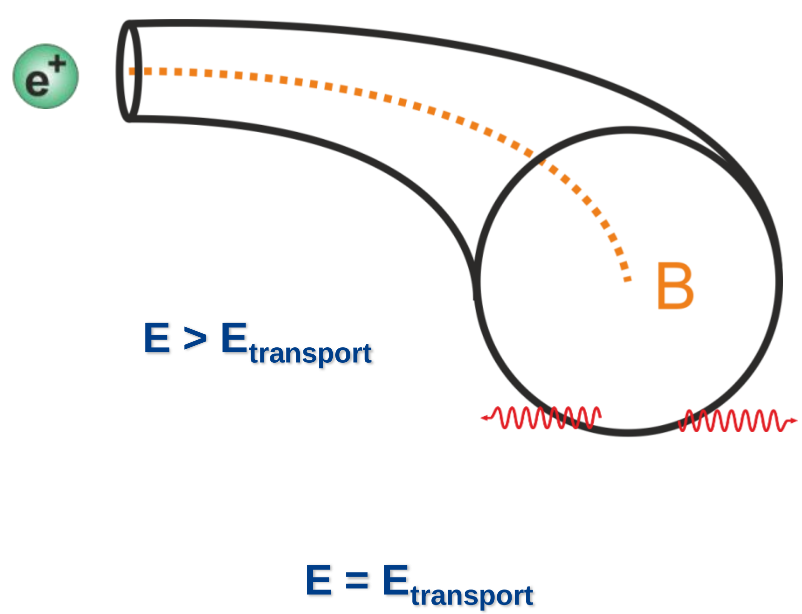
DRESDEN concept HZDR

Technical hints of PAS

Moderation of positrons and selection of correct energies



$$\vec{F} = q(\vec{v} \times \vec{B})$$



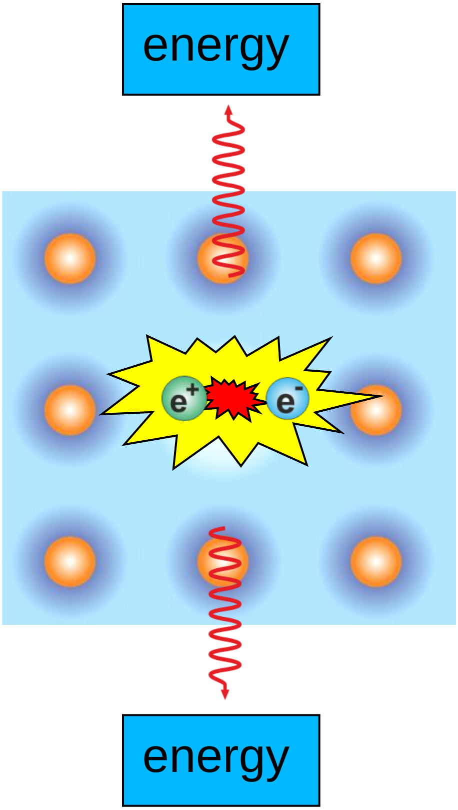
- energy of moderated positrons = 3 eV
- still a huge number of fast positrons

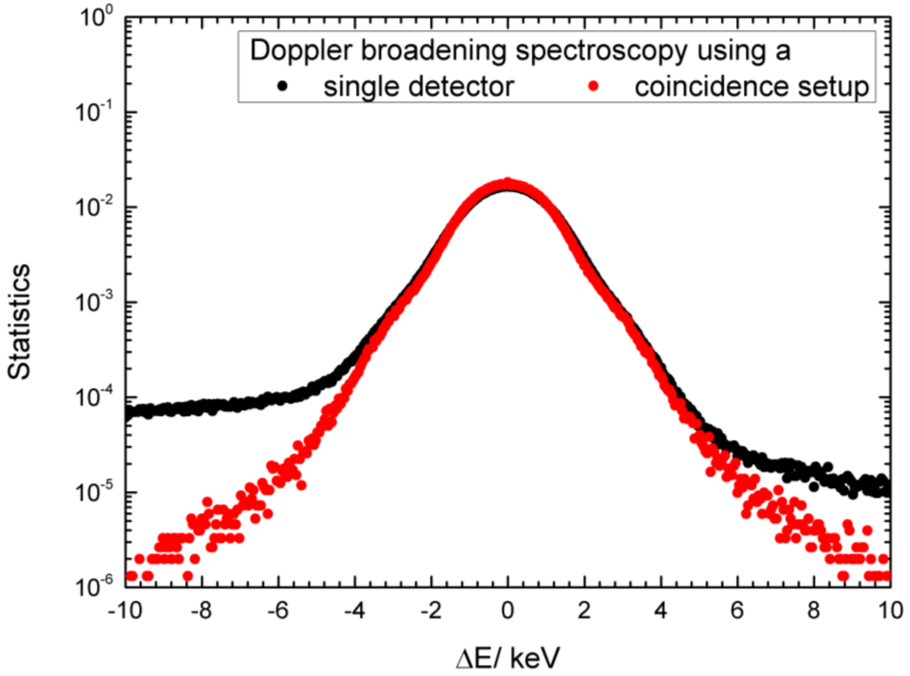
- bent tube to select positrons

Krause-Rehberg, Positron annihilation in semiconductors,Springer Verlag 1999

Technical hints of PAS

Coincidence Doppler broadening

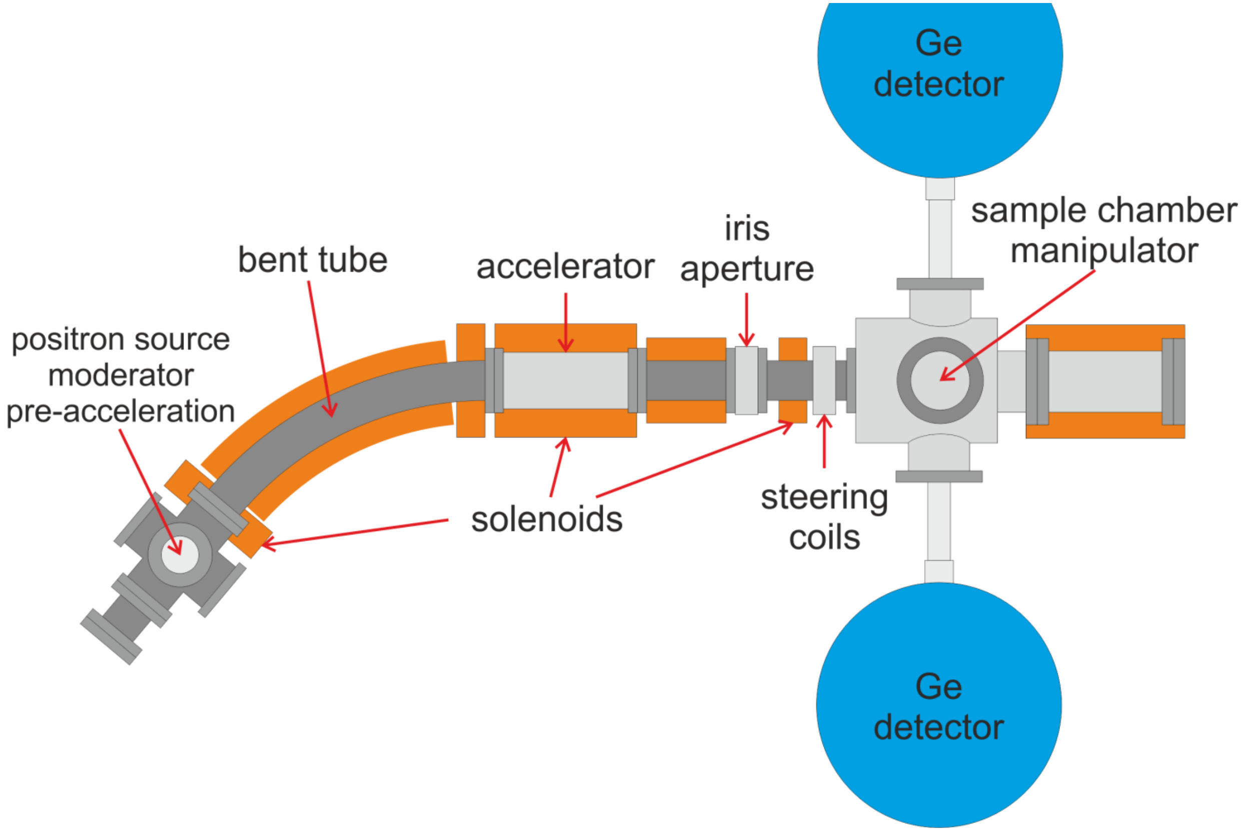




- better peak to background ratio
- important for chemical sensitivity

Realization of depth-resolved defect profiling

Slow POsitroN System Of Rossendorf - SPONSOR

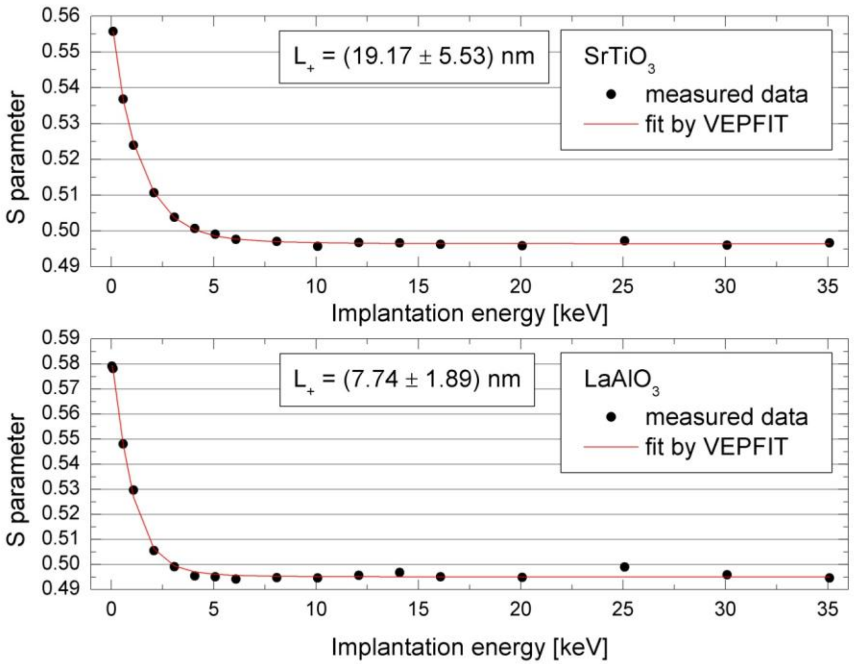


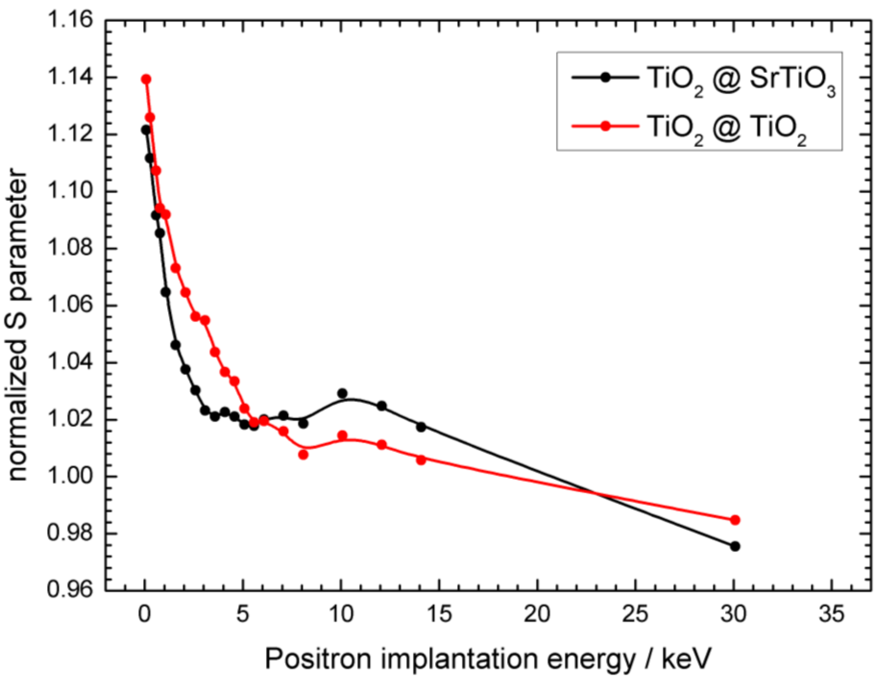
Doppler broadening spectroscopy

- positron energy: 27 eV … 36 keV
- energy resolution: (1.09 ± 0.01) keV at 511 keV

Realization of depth-resolved defect profiling

DETI.2 – Qualitity of substrate materials

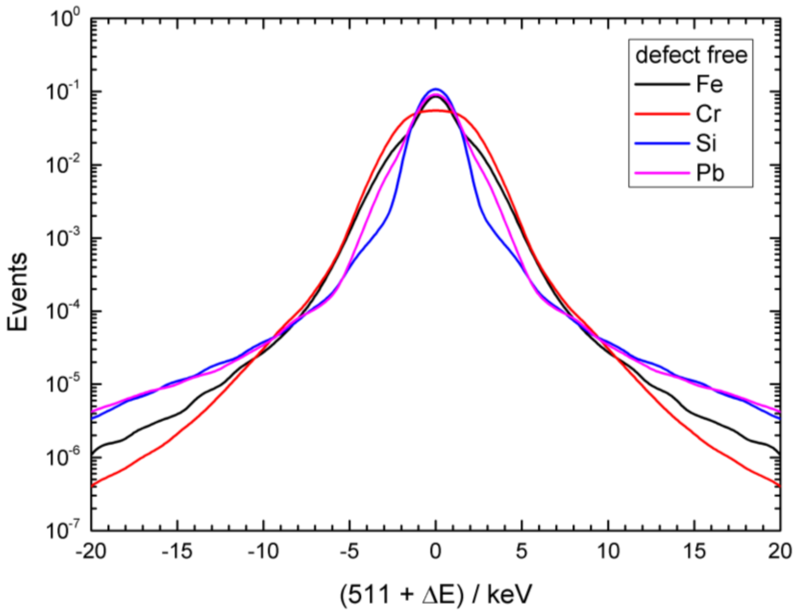


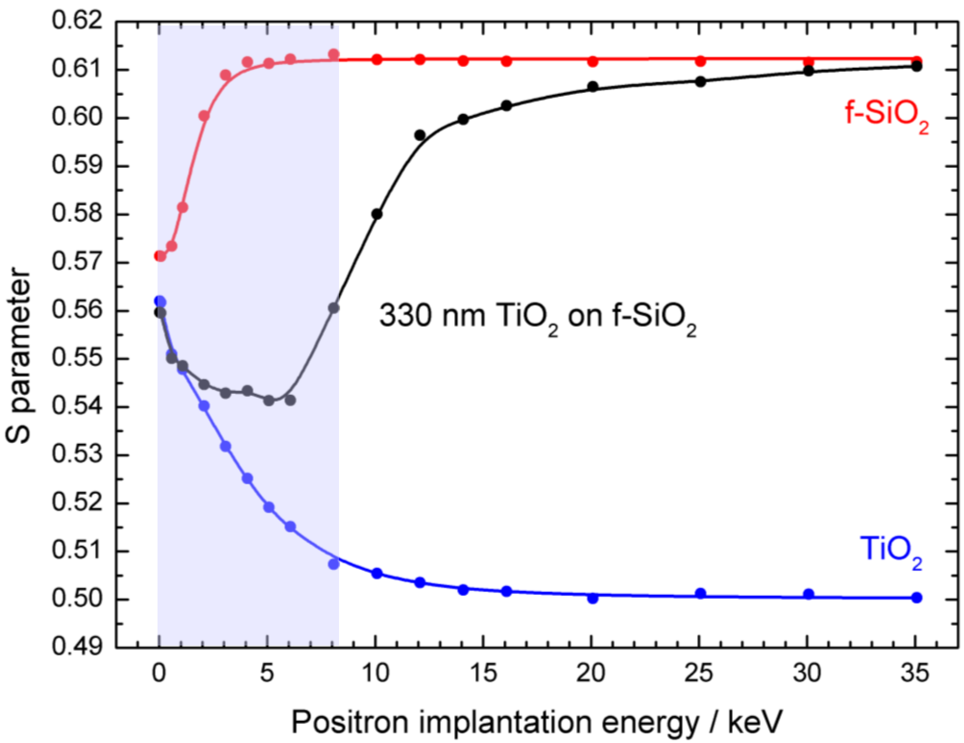


- Shorter diffusion length due to higher defect density/ larger defects
- $SrTiO_3$ is of better quality

DETI.2 – Components of the depth profile

Differences in S parameter – a question of reference

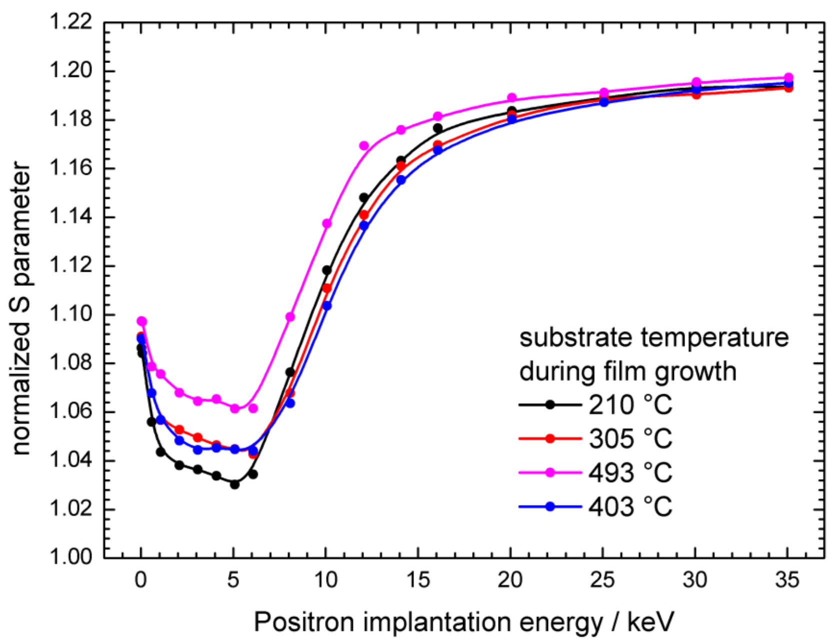


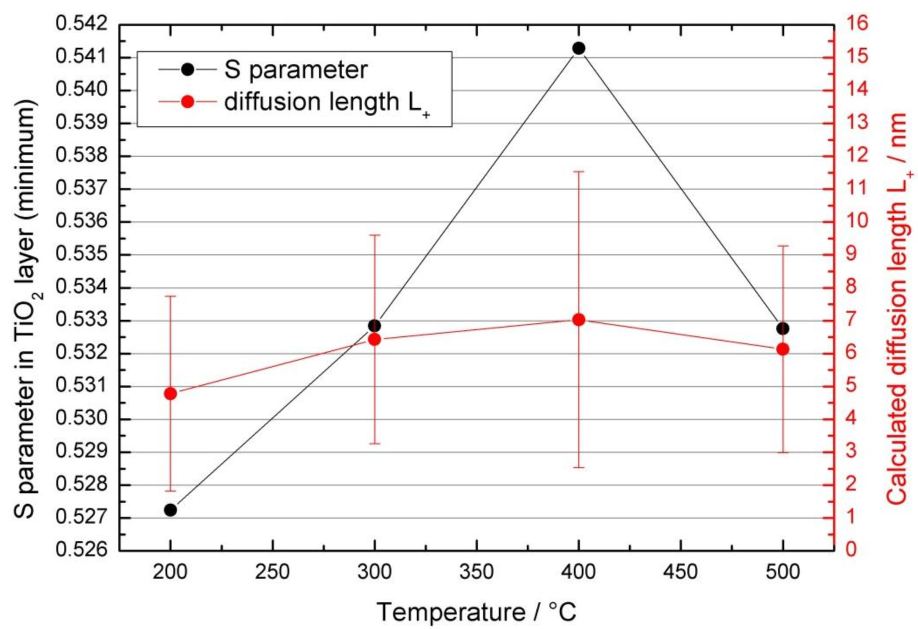


- S not only changes for different defect types/ concentrations
- also differences for different materials

DETI.2 – Influence of growth conditions

film growth temperature

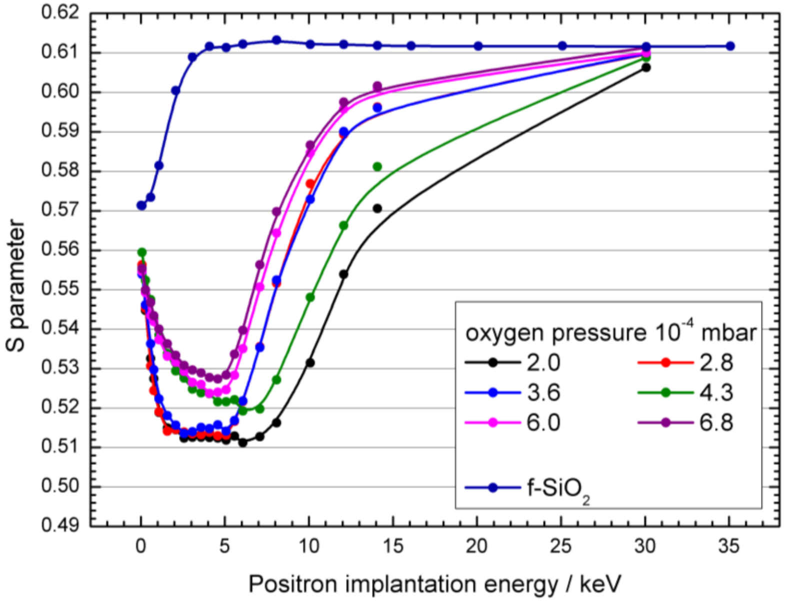


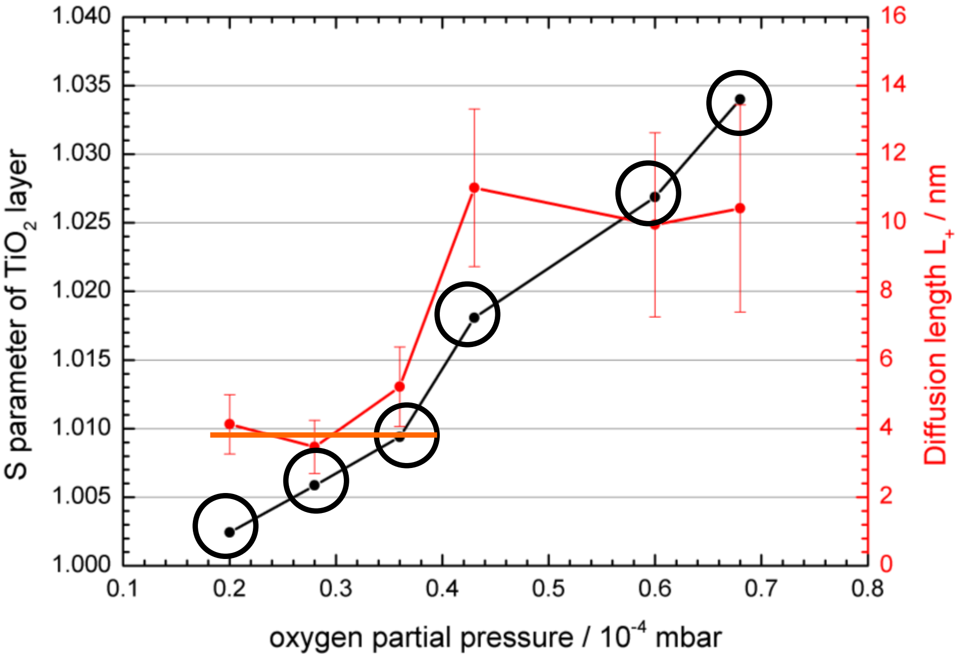


- Differences due to temperature
- unexpected jump in the S parameter

DETI.2 – Influence of growth conditions

oxygen partial pressure





- behaviour of L_+ and S:
  possible defect agglomeration

Many thanks for your attention !