Materials Research using Positron Annihilation

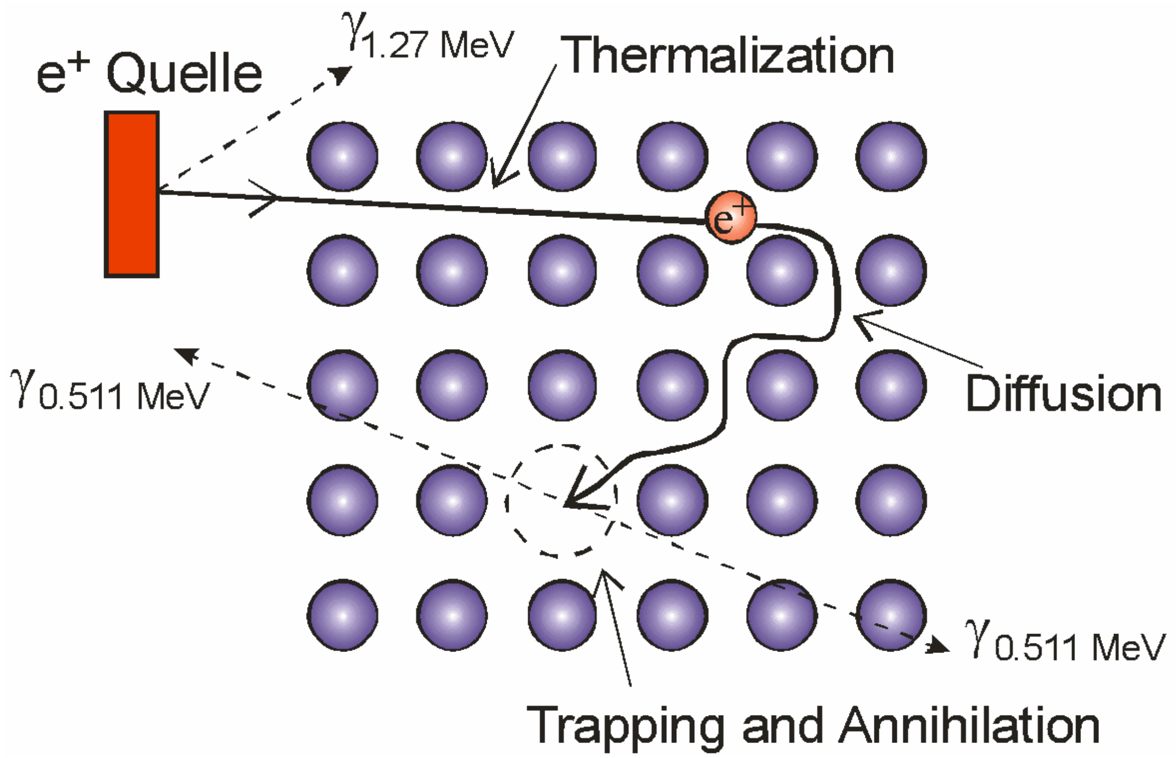
R. Krause-Rehberg

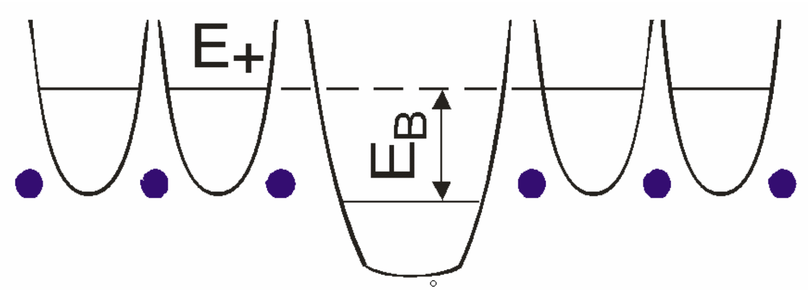
Martin-Luther-Universität Halle-Wittenberg
Germany

Martin-Luther-Universität
Halle-Wittenberg

- Introduction: Positrons detect lattice defects
- Examples:
    - irradiated Ge
    - study of defects in GaAs
    - new getter centers in Si after high-energy self-implantation (Rp/2 effect)
- Large Positron Facility Projects in Germany
- Conclusions

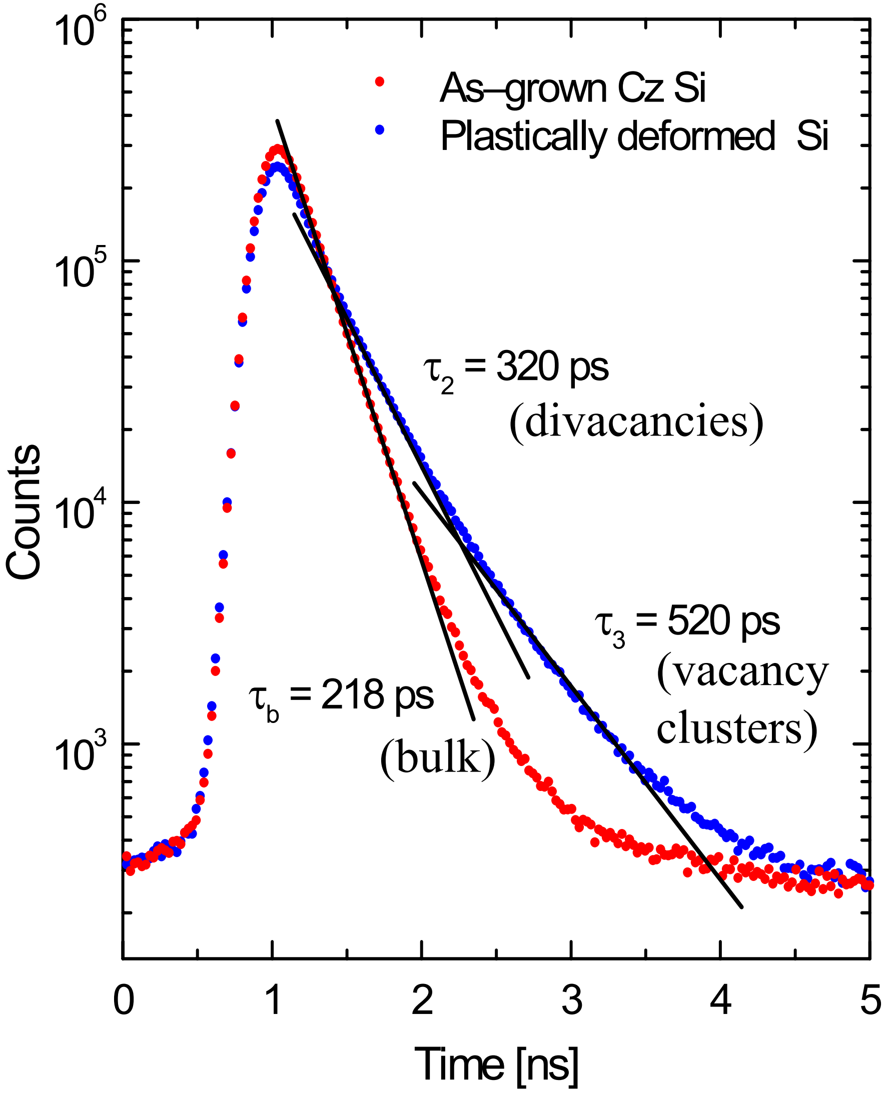
The positron lifetime spectroscopy





- 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 1.27 and 0.51 MeV quanta
- defect identification and quantification possible

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:

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

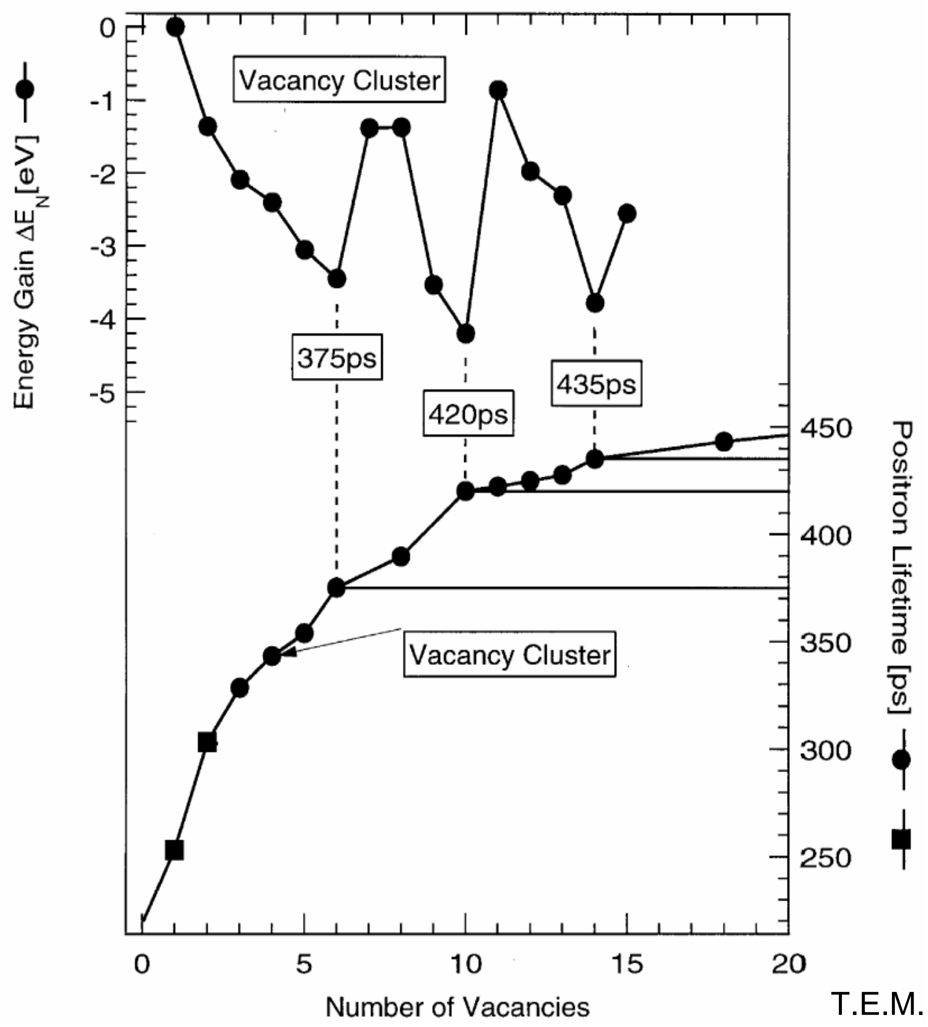
trapping coefficient

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

trapping rate

defect concentration

Theoretical calculation of vacancy clusters in Si

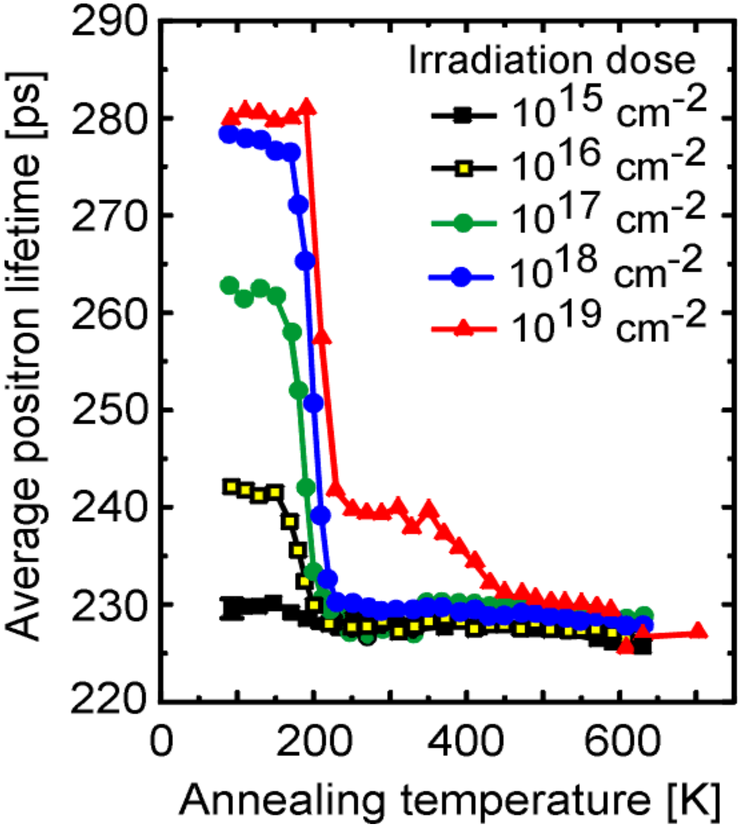


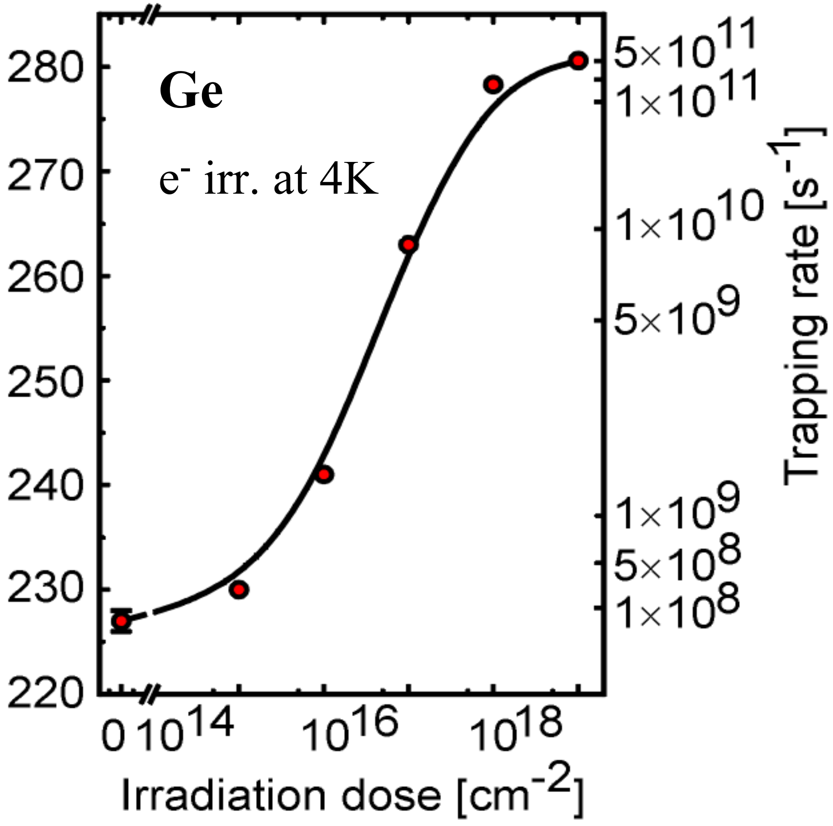
- there are cluster configurations with a large energy gain
- „Magic Numbers“ with 6, 10 und 14 vacancies
- positron lifetime increases distinctly with cluster size
- for n > 10 saturation effect, i.e. size cannot be determined

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

Defects in electron-irradiated Ge

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

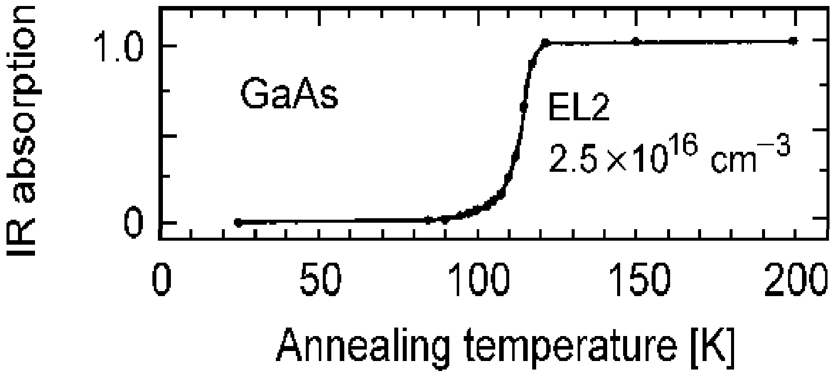


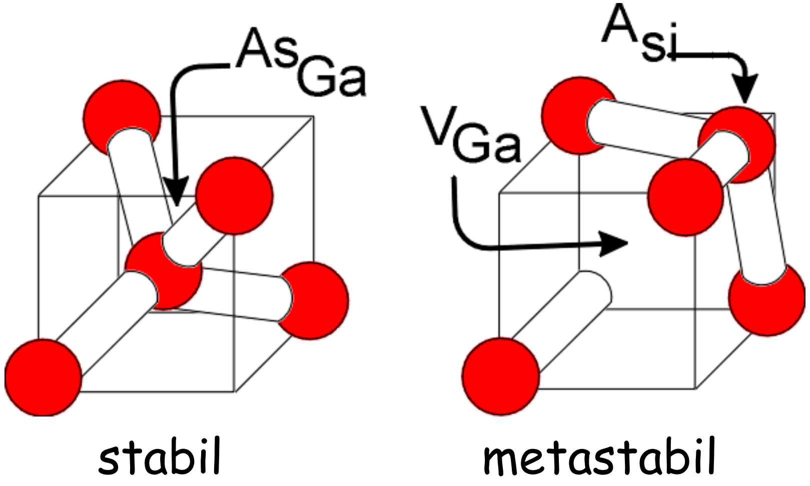


(Polity et al., 1997)

The Nature of the EL2-Defect in GaAs

- one of the most frequently studied crystal lattice defects at all
- responsible for semi-insulating properties of GaAs: large technological importance
- is deep donor, compensates shallow acceptors, e.g. C^- impurities
- defect shows metastable state after illumination at low temperatures
- IR-absorption of defect disappears during illumination at T < 100 K
- ground state recovers during annealing at about 110 K
- many structural models proposed
- Dabrowski, Scheffler and Chadi, Chang (1988): simple As_{Ga}-antisite defect responsible
- must show a metastable structural change

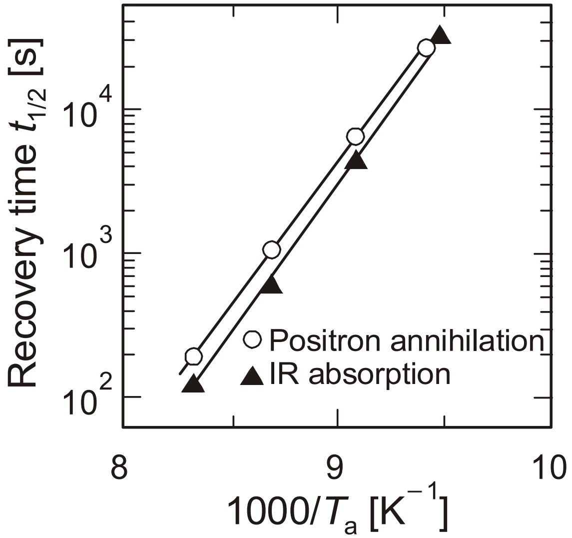


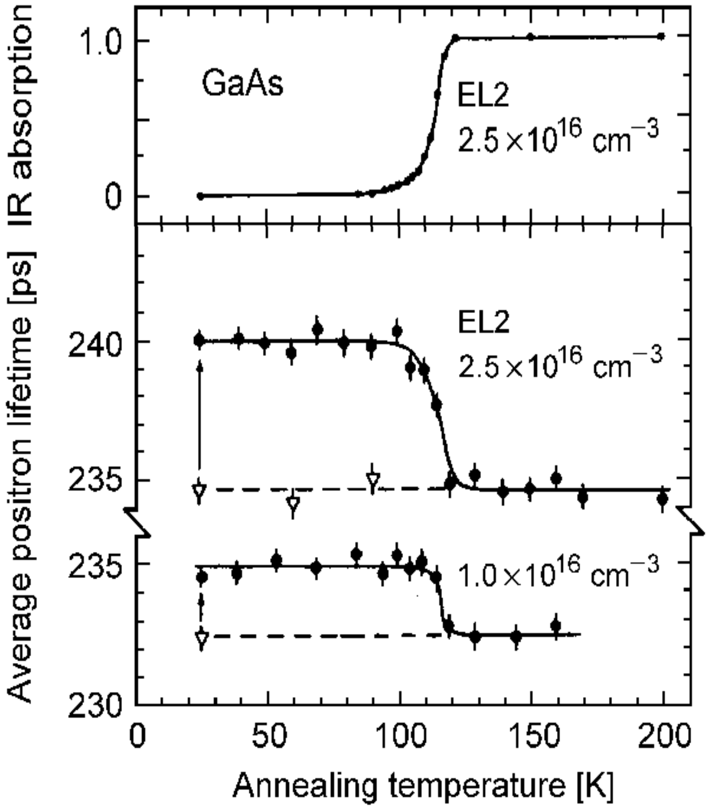


(Dabrowski 1988, Chadi 1988)

The Nature of the EL2-Defect in GaAs

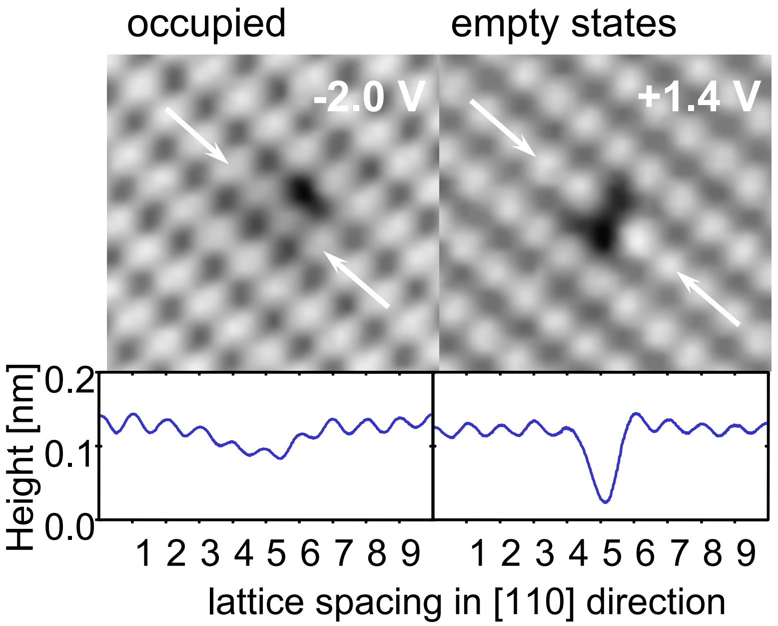
- in metastable state at low temperature: Ga vacancy
- should disappear during annealing at about 110 K
- confirmed by positron lifetime measurements
- kinetics of recovery of ground state is identical for IR- und positron experiment: $E_A = (0.37 \pm 0.02)$ eV
- evidence of the vacancy in metastable state confirms the proposed structural model

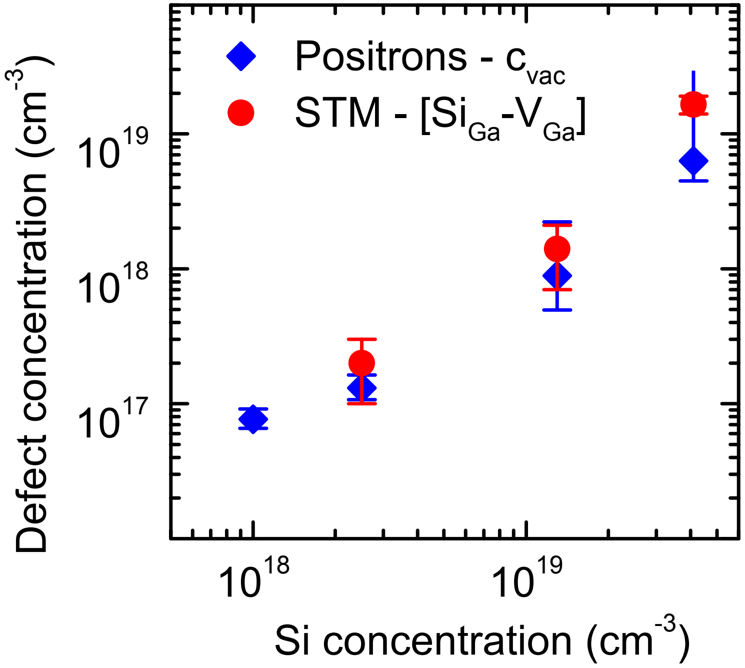




Krause et al., Phys. Rev. Lett. **65** (1990) 3329

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





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

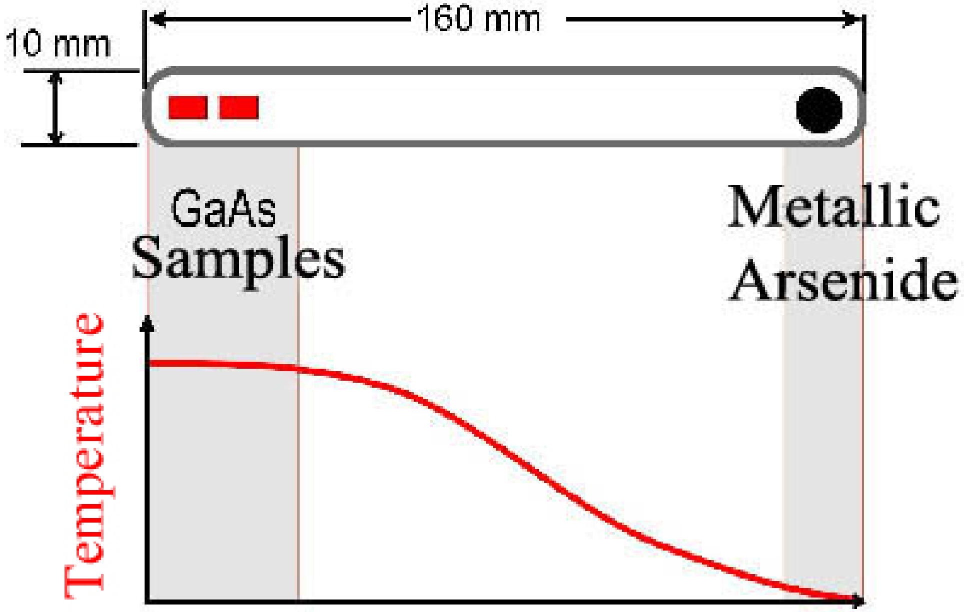
- Quantification → Agreement

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

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

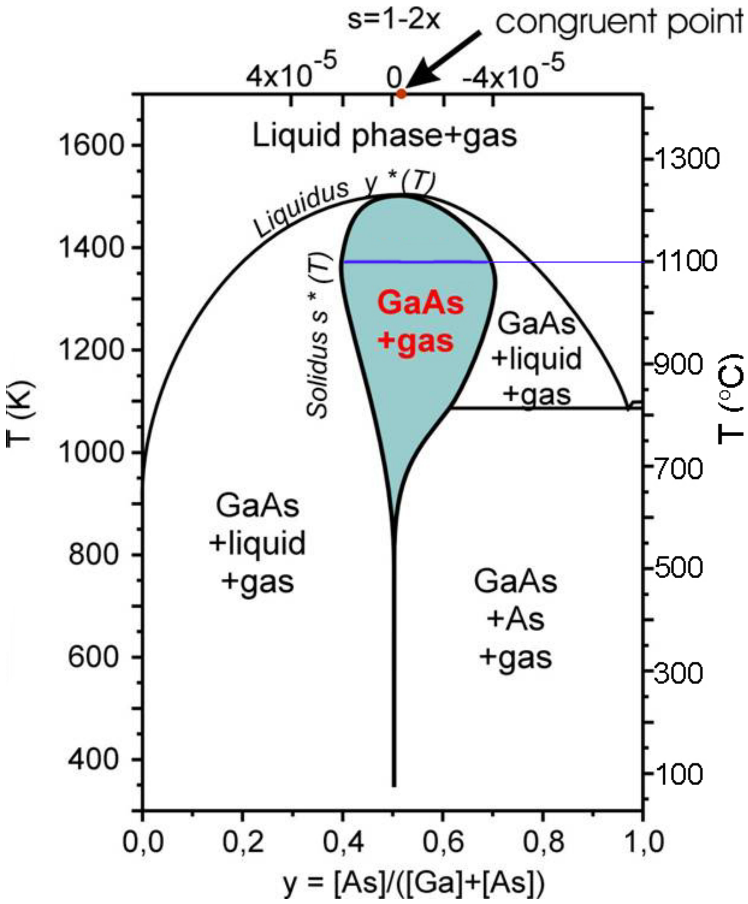
GaAs: annealing under defined As-partial pressure

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



T_{sample}: 1100° C

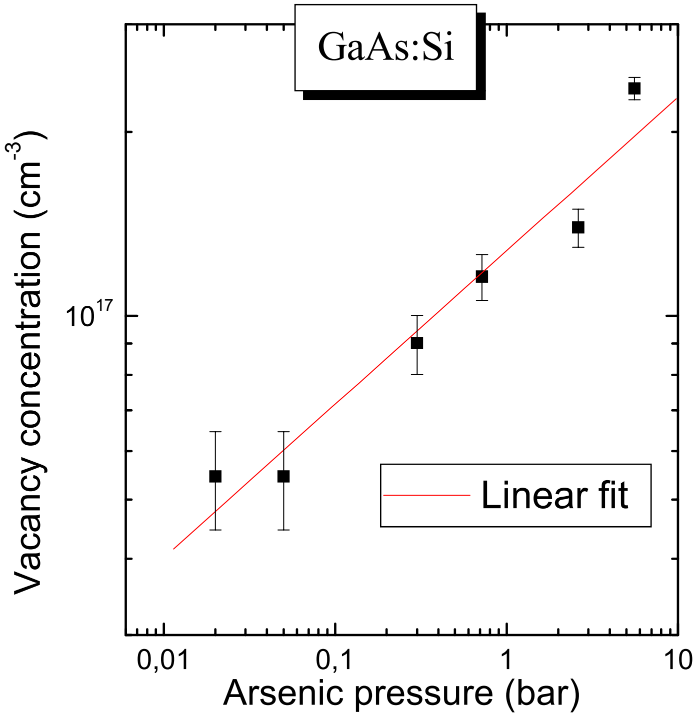
T_{As}: determines As-partial pressure



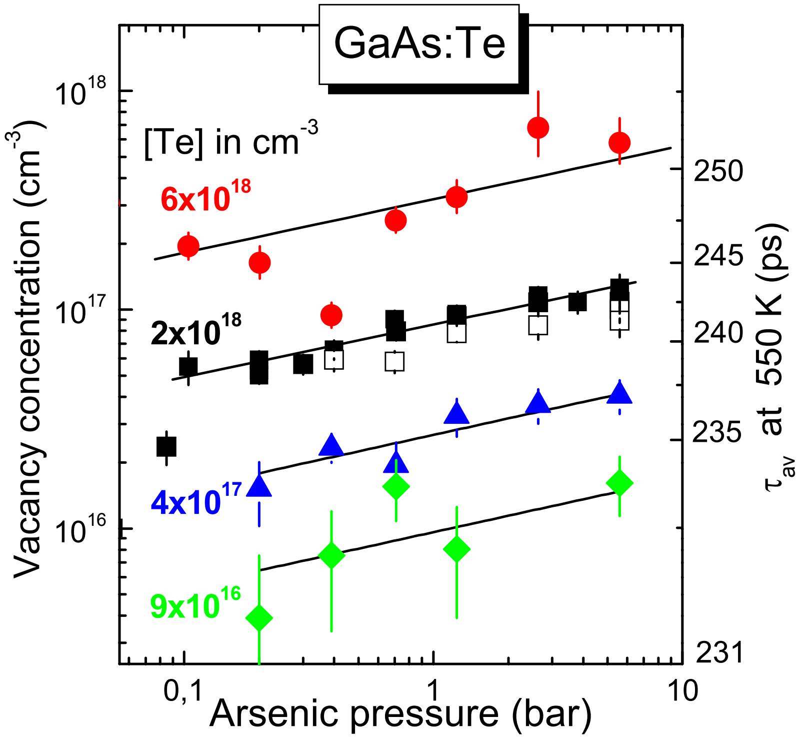
H. Wenzl et al., J. Cryst. Growth **109**, 191 (1991).

Experiments in n-GaAs

Si_{Ga}-V_{Ga}



Te_{As}-V_{Ga}



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

Thermodynamic reaction:

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

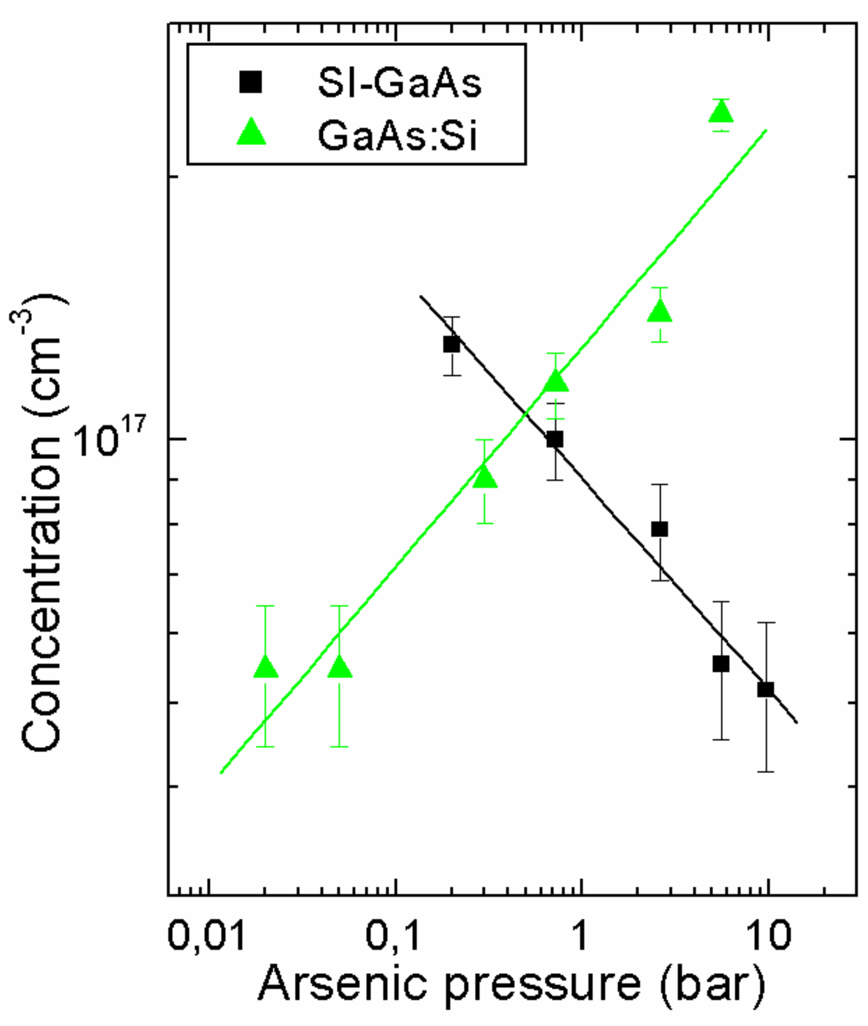
Mass action law:

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

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

$\rightarrow n = 1/4$

Comparison of doped and undoped GaAs



Thermodynamic reaction:

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

Mass action law:

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

Fit: [V-complex] ~ p_{As}^{n}

$\rightarrow n = -1/4$

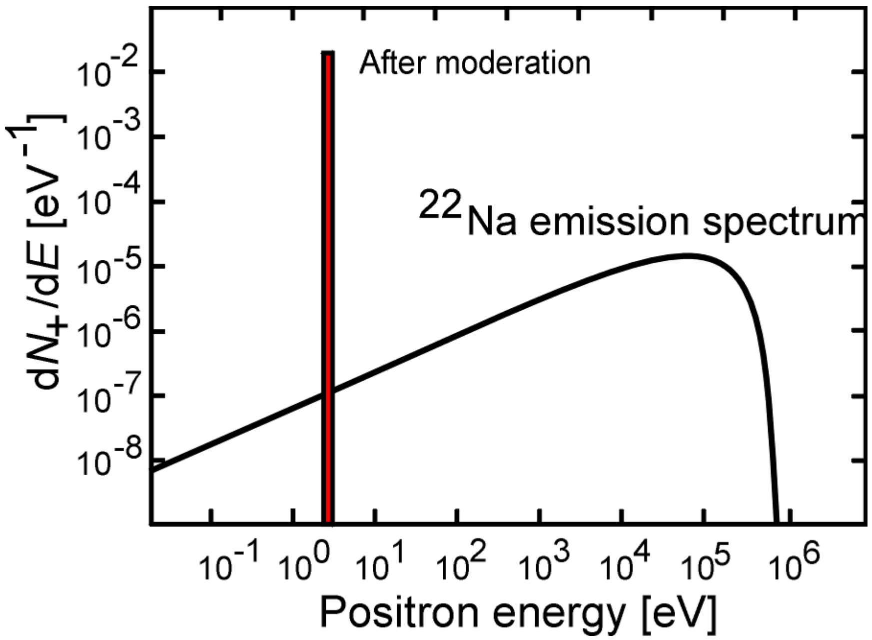
↓

As vacancy

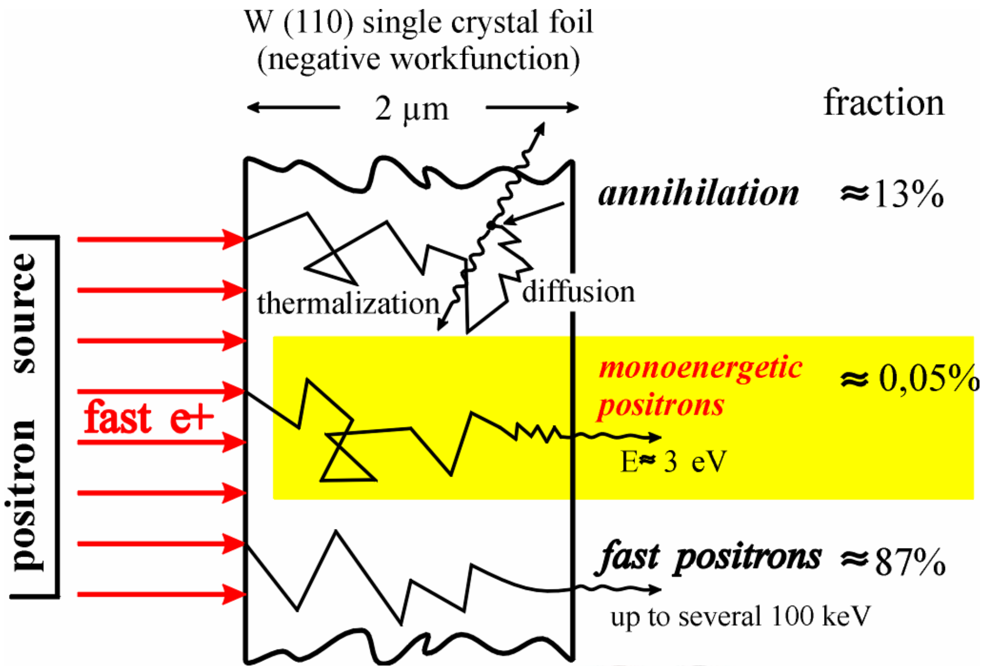
Monoenergetic positrons obtained by moderation

- semiconductor technology: thin layers (epitaxy, ion implantation)
- broad energy distribution due to β^+ decay
- some surfaces: negative workfunction ⇒ moderation (but rather inefficient)

Energy distribution after β^+ decay



Effect of moderation



W (110) single crystal foil
(negative workfunction)

2 μm

fraction

annihilation ≈13%

thermalization

diffusion

positron source

fast e+

monoenergetic positrons ≈ 0,05%

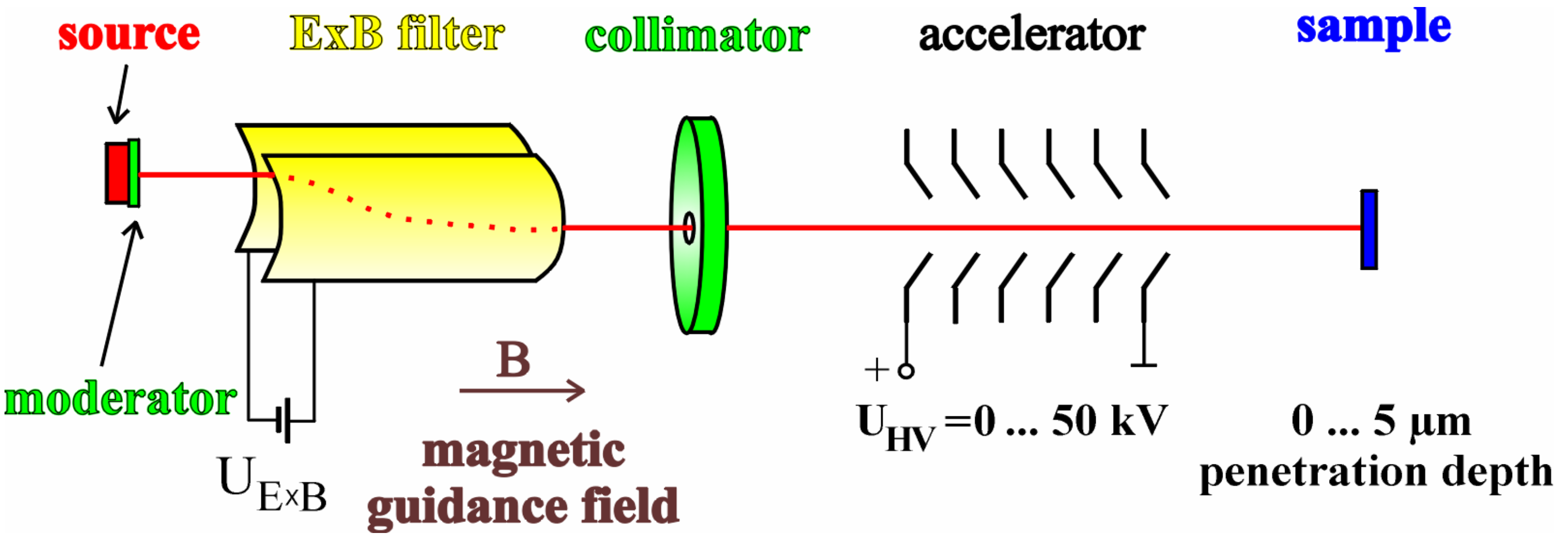
E≈ 3 eV

fast positrons ≈87%

up to several 100 keV

Conventional positron beam technique

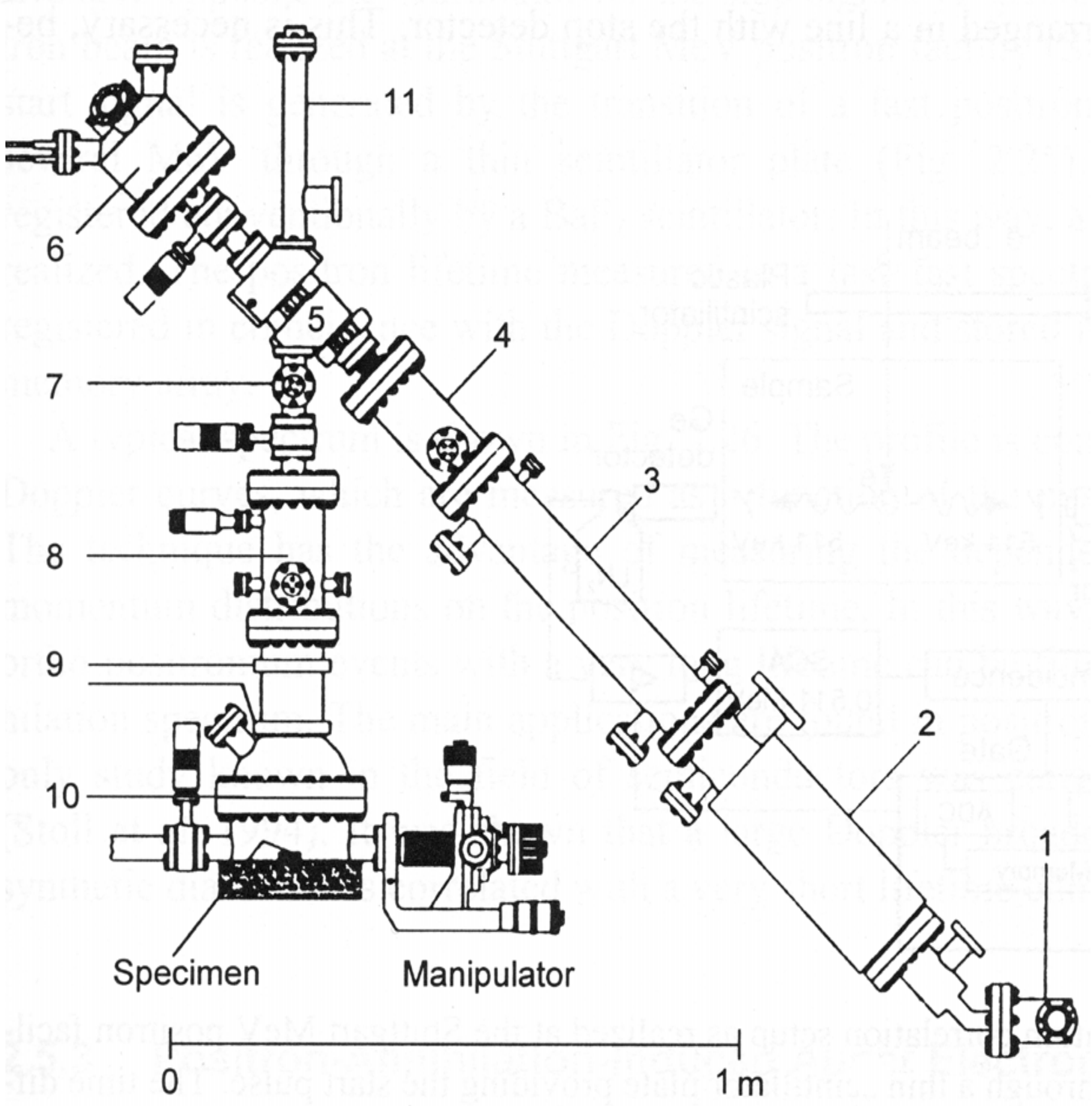
- positron beam can be formed using mono-energetic positrons
- often: magnetically guided for simplicity



- defect studies by Doppler-broadening spectroscopy
- characterization of defects only by line-shape parameters or positron diffusion length
- for positron lifetime spectroscopy: beam can be bunched

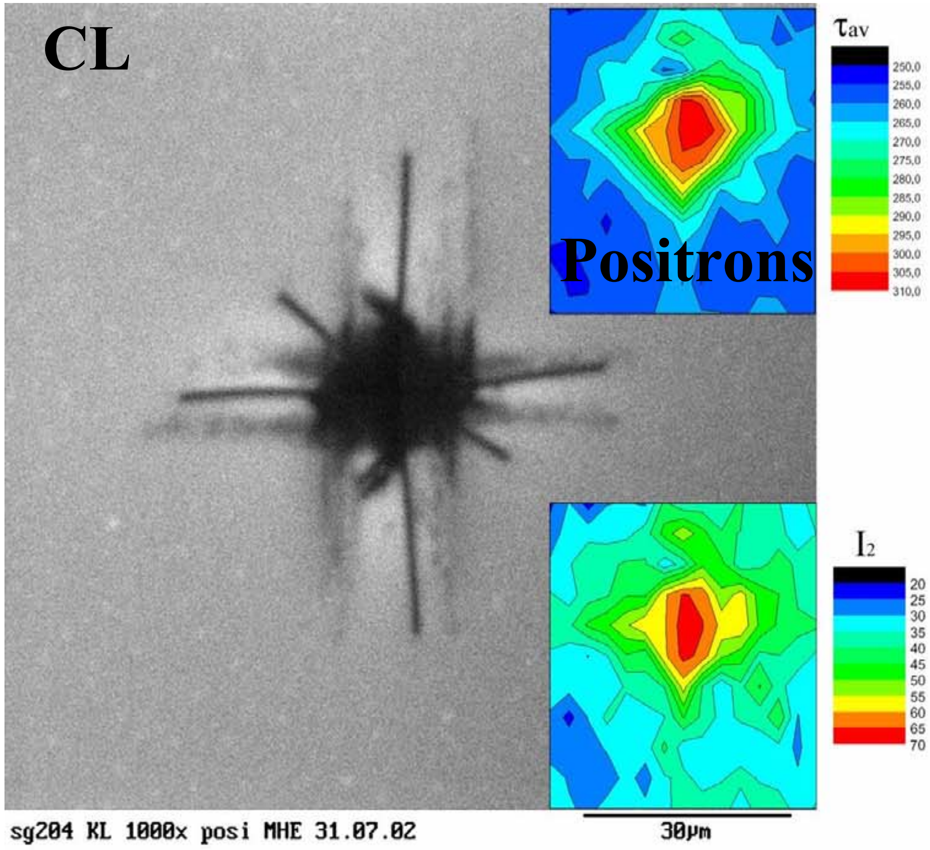
Scanning Positron Microscope in Munich

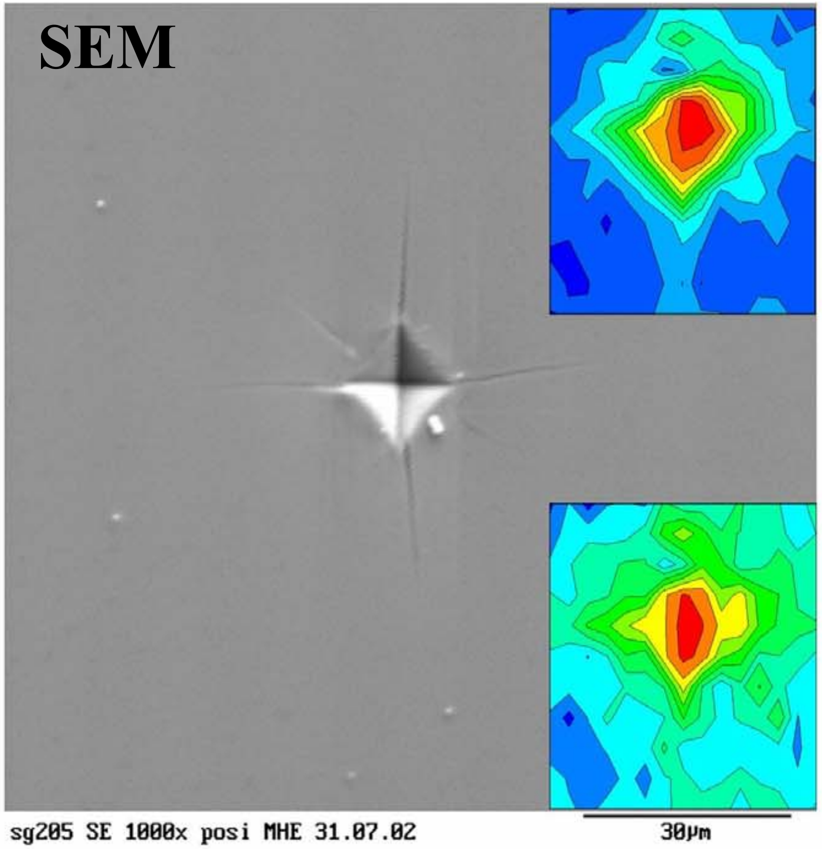
- Semiconductor devices nm-sized ⇒ Positron microprobes required
- images show directly distribution of positron traps, i.e. nanoscopic lattice defects
- However: positron diffusion length is fundamental limit for lateral resolution
- no sense to improve resolution much below 500 nm
- first instrument was realized at Univ. Bonn (20 μm; Doppler spectroscopy)
- first realization of scanning positron microscope for lifetime spectroscopy: in Munich



Example for use of Munich Microscope: Mikrohardness indentation in GaAs

- Comparison of SEM, cathodoluminescence (CL) and Munich Positron Scanning Microscope; problem here at the moment: intensity

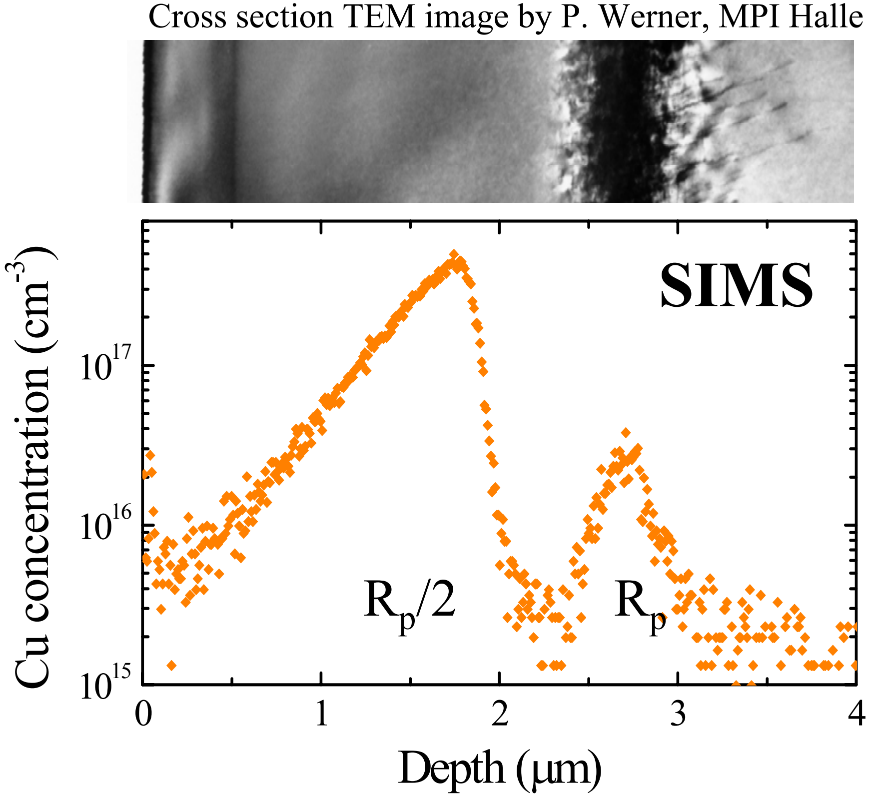




(Krause-Rehberg et al., 2002)

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 and $R_p/2$ (R_p = projected range of Si^+)
- visible by SIMS profiling after intentional Cu contamination


Cross section TEM image by P. Werner, MPI Halle

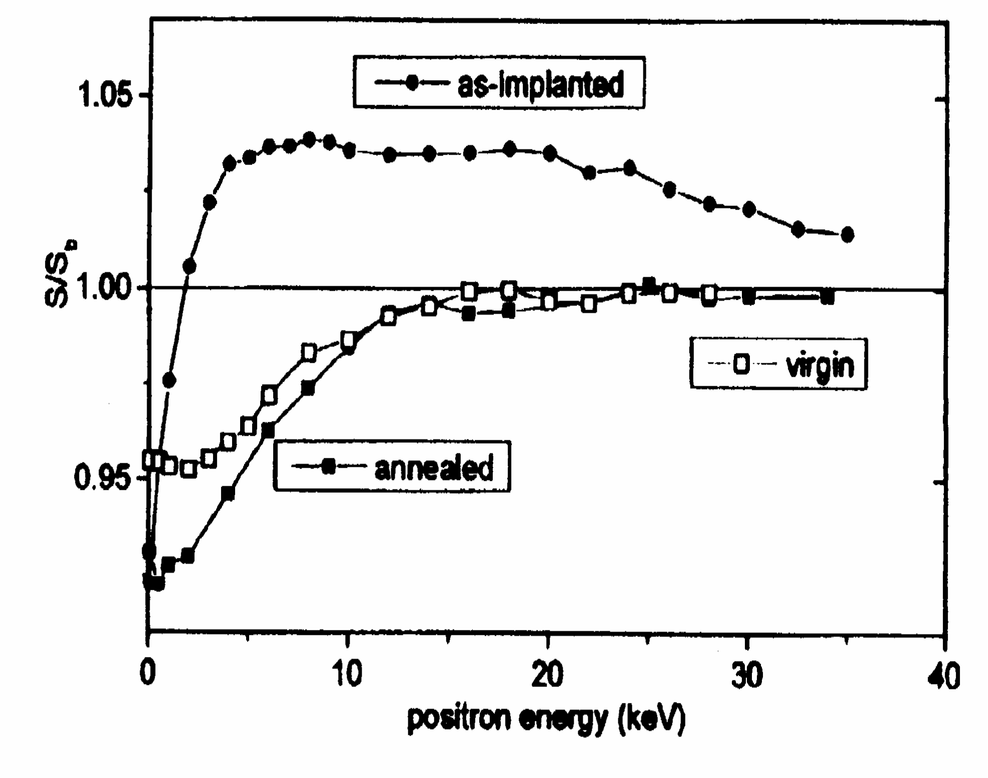
- at R_p: gettering by interstitial-type dislocation loops (formed by excess interstitials during RTA)
- no defects visible by TEM at $R_p/2$
- **What type are these defects?**

Interstitial type [3,4]

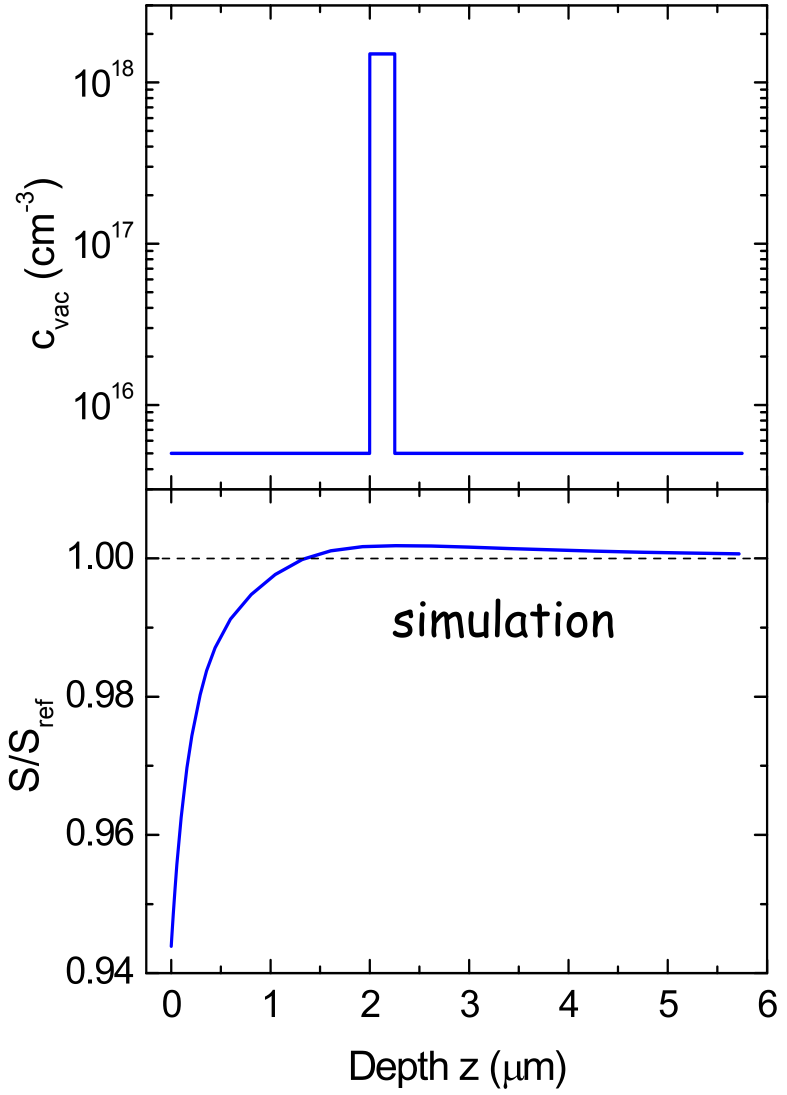
Vacancy type [1,2]

[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

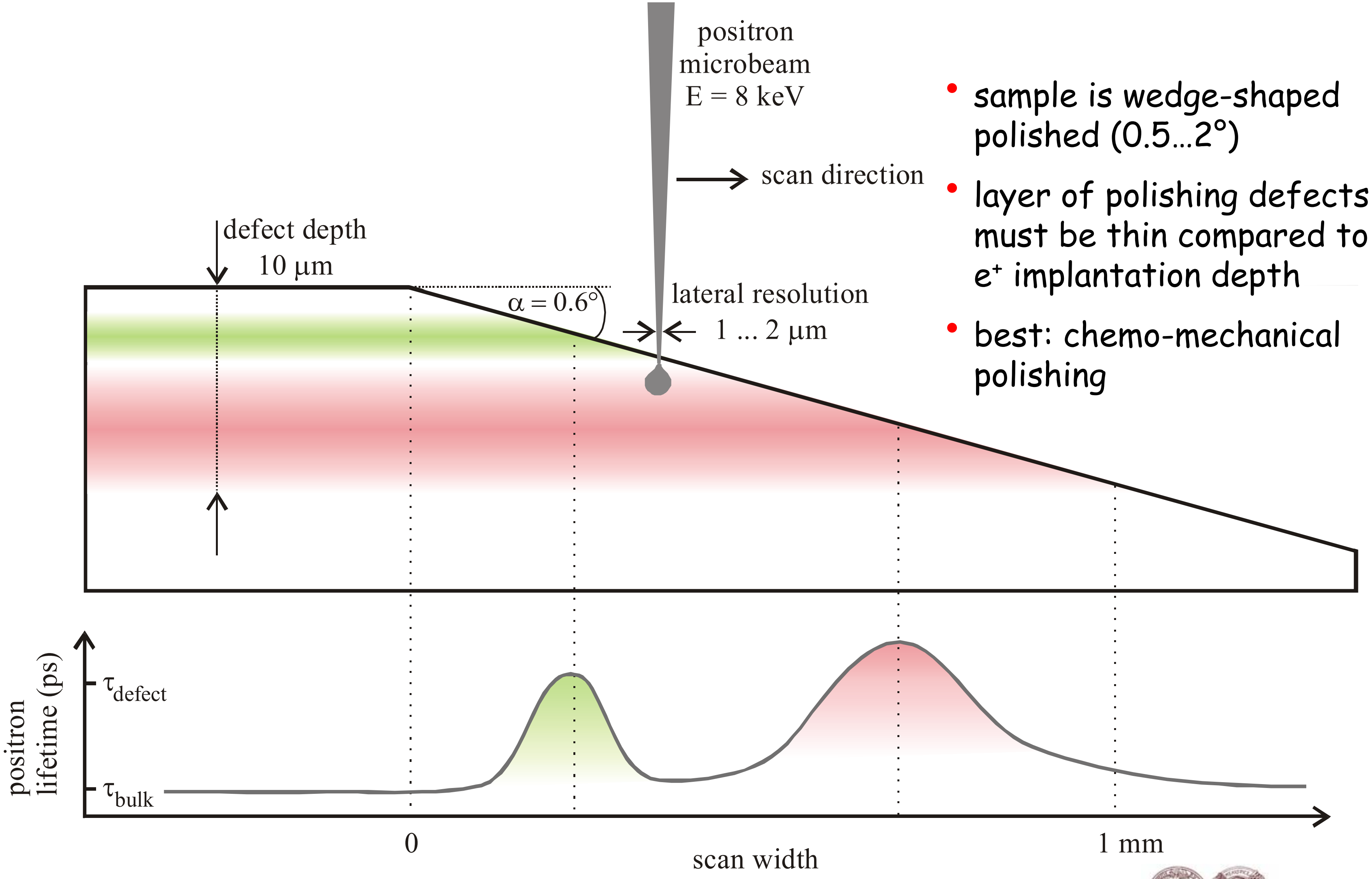
Determination of defect profiles – a model



Kögler et al. Appl. Phys. Lett. 75 (1999) 1279



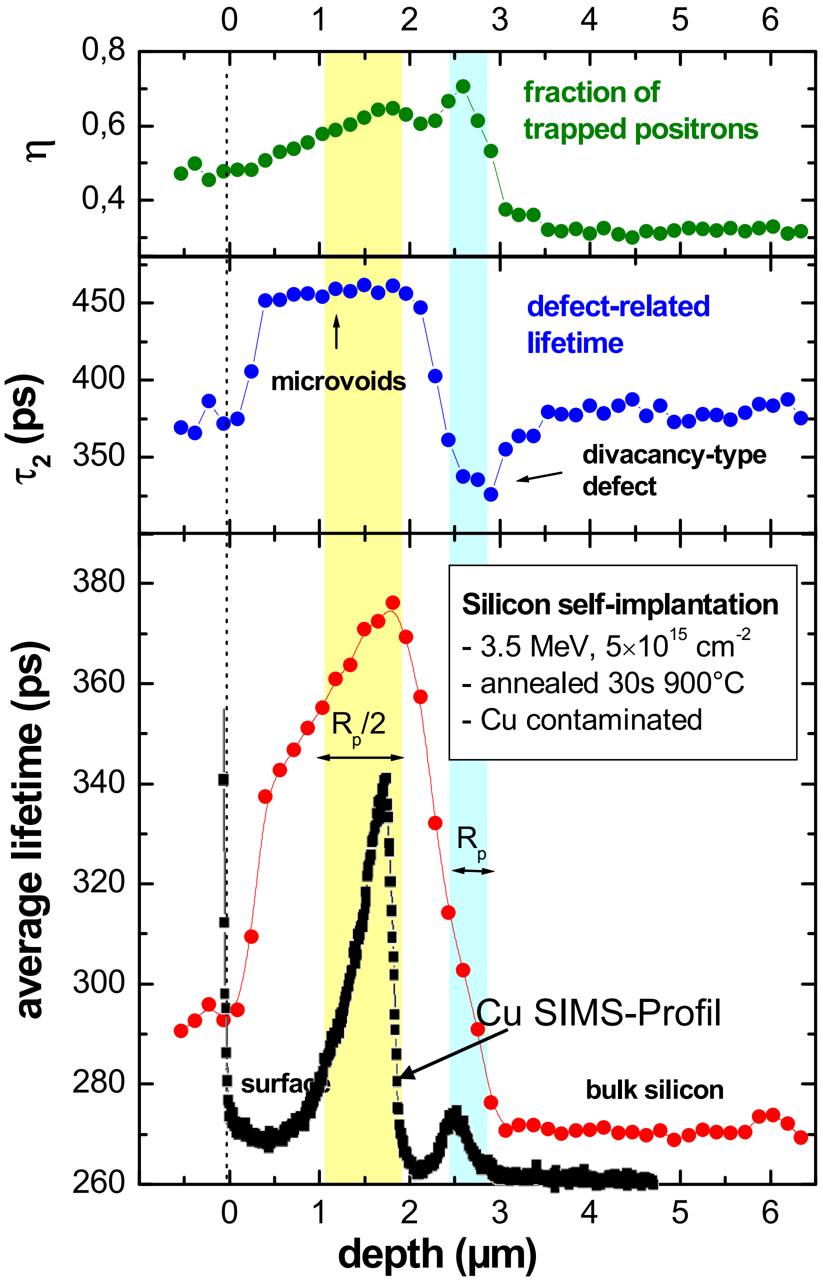
Enhanced depth resolution by using the Munich Scanning Positron Microscope



- sample is wedge-shaped polished (0.5…2°)
- layer of polishing defects must be thin compared to e^+ implantation depth
- best: chemo-mechanical polishing

Depth profile of Rp/2 smple measured using the Microscope

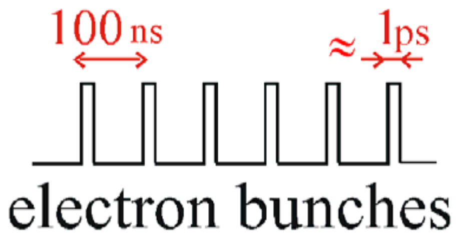
- 45 lifetime spectra: scan along wedge
- separation of 11 µm between two measurements corresponds to depth difference of 155 nm ($\alpha = 0.81°$)
- beam energy of 8 keV ⇒ mean penetration depth is about 400 nm; represents optimum depth resolution
- no further improvement possible due to positron diffusion: L_+(Si @ 300K) ≈ 230 nm
- both regions well visible:
    - vacancy clusters with increasing density down to 2 µm ($R_p/2$ region)
    - in R_p region: lifetime τ_2 = 330 ps; corresponds to open volume of a divacancy; must be stabilized or being part of interstitial-type dislocation loops
- Problem of microscope: Intensity



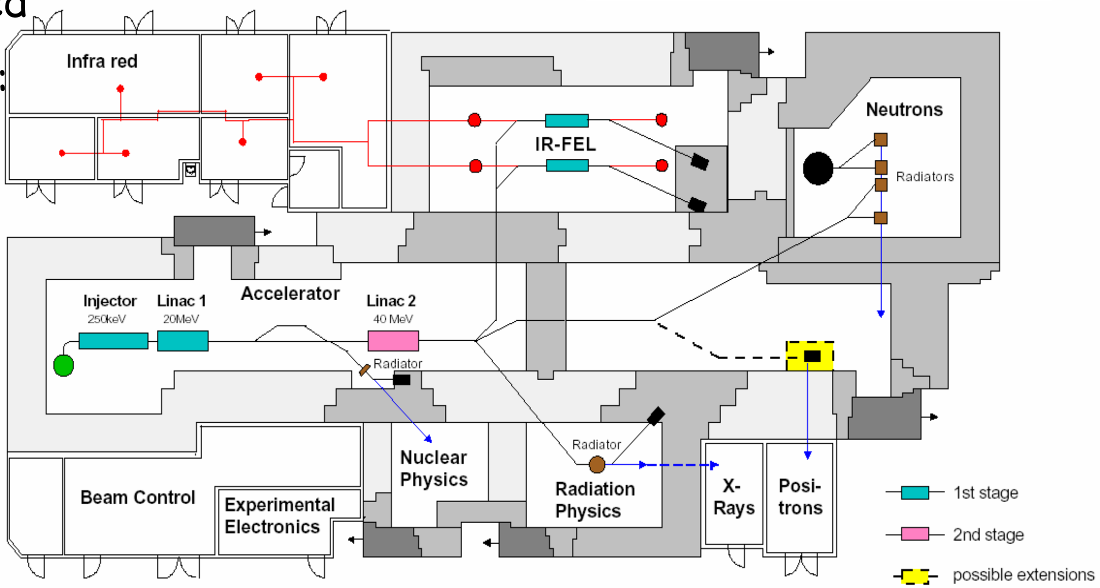
Large Positron Facility Projects in Germany

- **FRM-II** (Positron source at Research Reactor-II in Garching near Munich)
    - intense continuous positron beam for different experiments, mainly for:
- **Scanning Positron Microscope** at “Universität der Bundeswehr”, Munich
    - system already working using isotope source (but too weak intensity)
    - positron lifetime measurement; lateral resolution about 2 µm
- **EPOS** – ELBE Positron Source (project at Research Center Rossendorf, near Dresden)
    - positron source for materials research at superconducting 40 MeV-FEL in Rossendorf
    - primary time structure suitable for positron lifetime spectroscopy

EPOS – ELBE Positron Source at Research Center Rossendorf

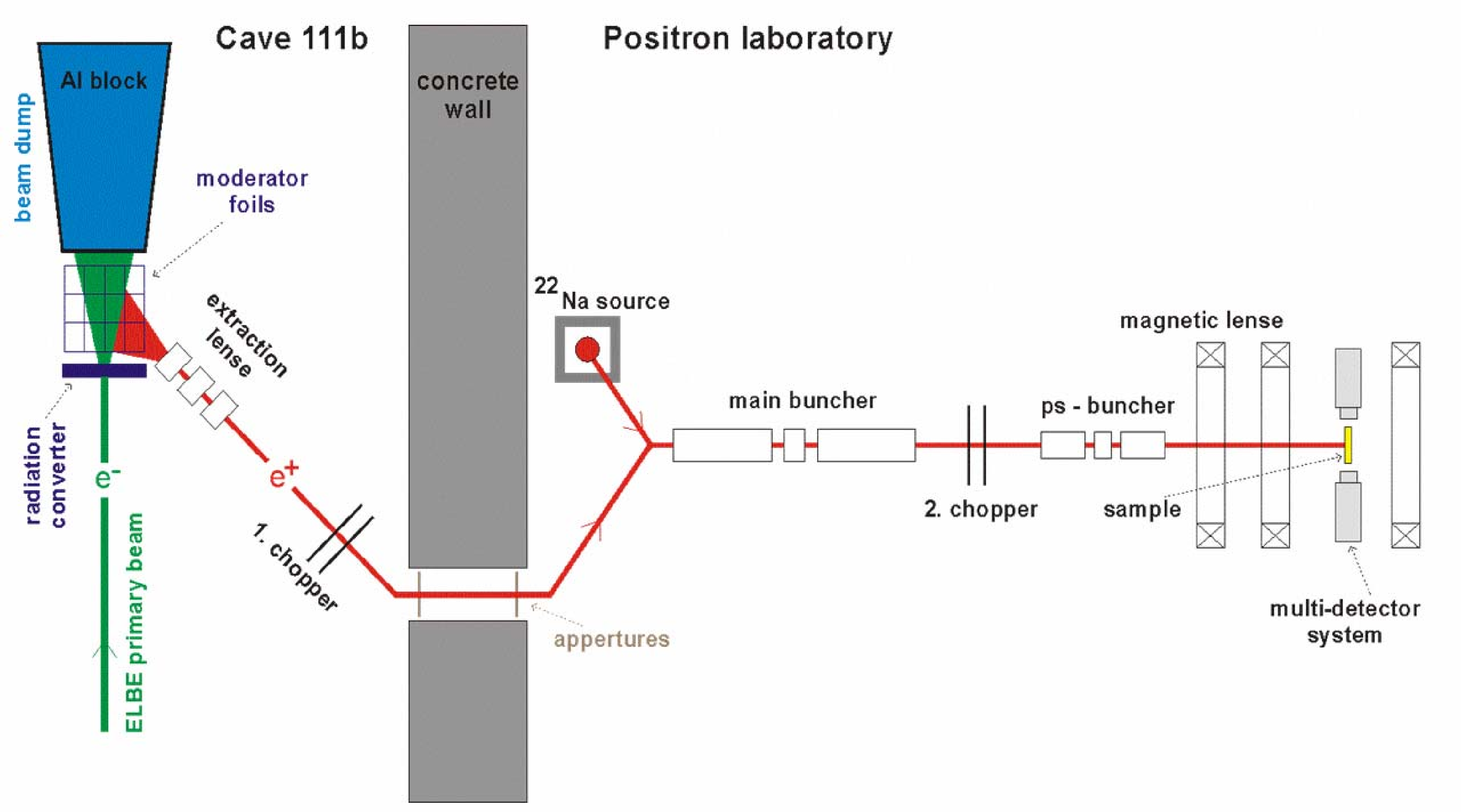


- electron beam at ELBE FEL is bunched
- bunch length: few ps, repetition time: ≈ 100 ns, cw-mode
- up to 10^8 e^-/bunch, 10^7 bunches/s
- beam energy: 40 MeV power: 40 kW
- FEL-system in Rossendorf under construction (ELBE)
- primary electron beam already available



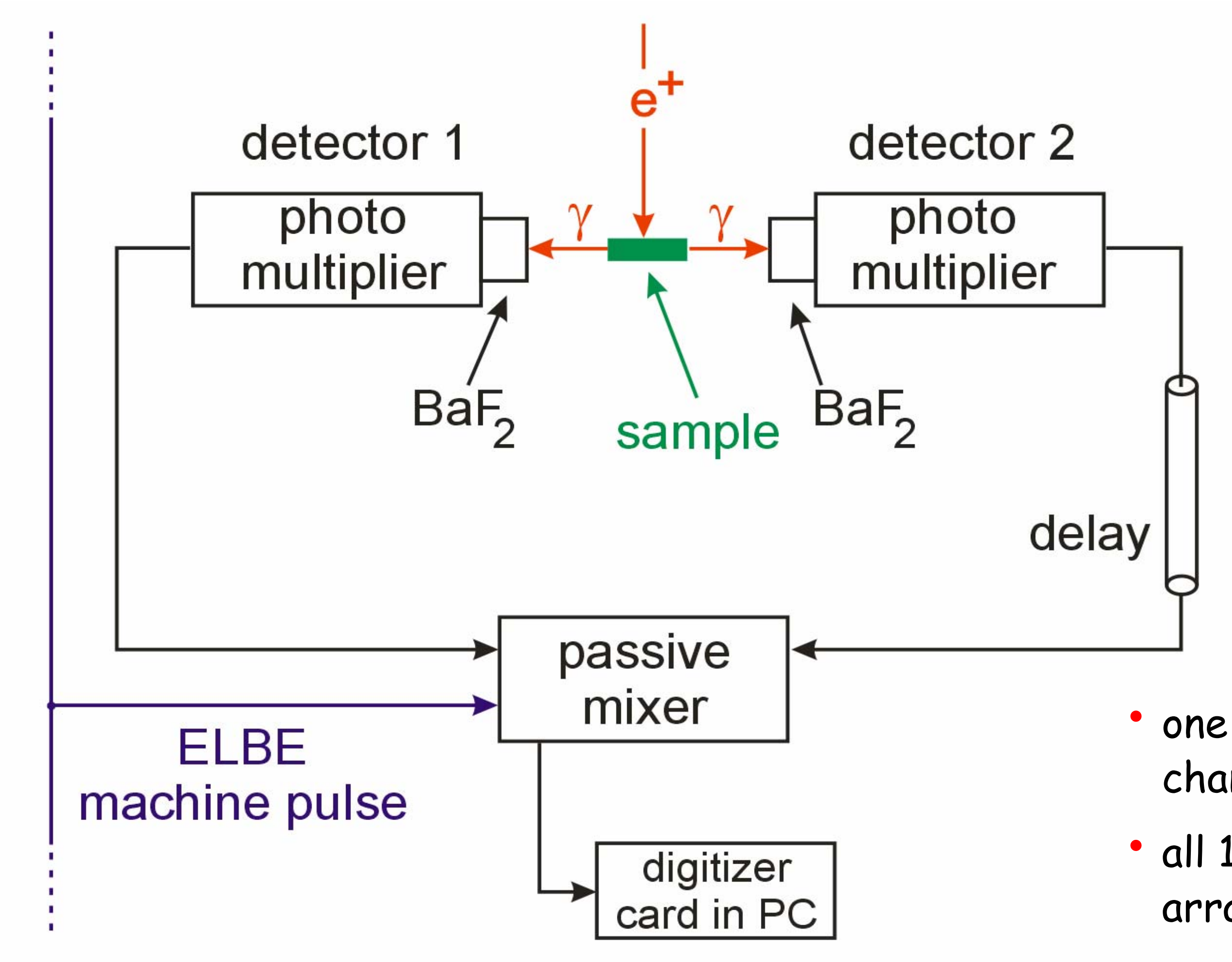
- direct positron lifetime measurement using time structure of e^- beam possible
- about 1 x 10^9 slow e^+/s; multi-detector system for high counting rate
- digital lifetime measurement
- combination with Doppler-coincidence spectroscopy and Age-momentum correlation (AMOC)

Schematic setup of EPOS



(top view, schematic drawing)

Digital coincidence lifetime measurement at EPOS



- one of 8 coincidence channels
- all 16 detectors are arranged in a circle
- very stable and simple setup

Conclusions

- many lattice defects can be detected in solids by means of positron annihilation (especially sensitive for vacancy-type defects)
- method very sensitive for early stage of vacancy agglomeration
- tools for thin layers (mono-energetic positron beams)
- scanning positron microbeams available
- intense positron sources under construction in Germany too

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

contact: mail@KrauseRehberg.de