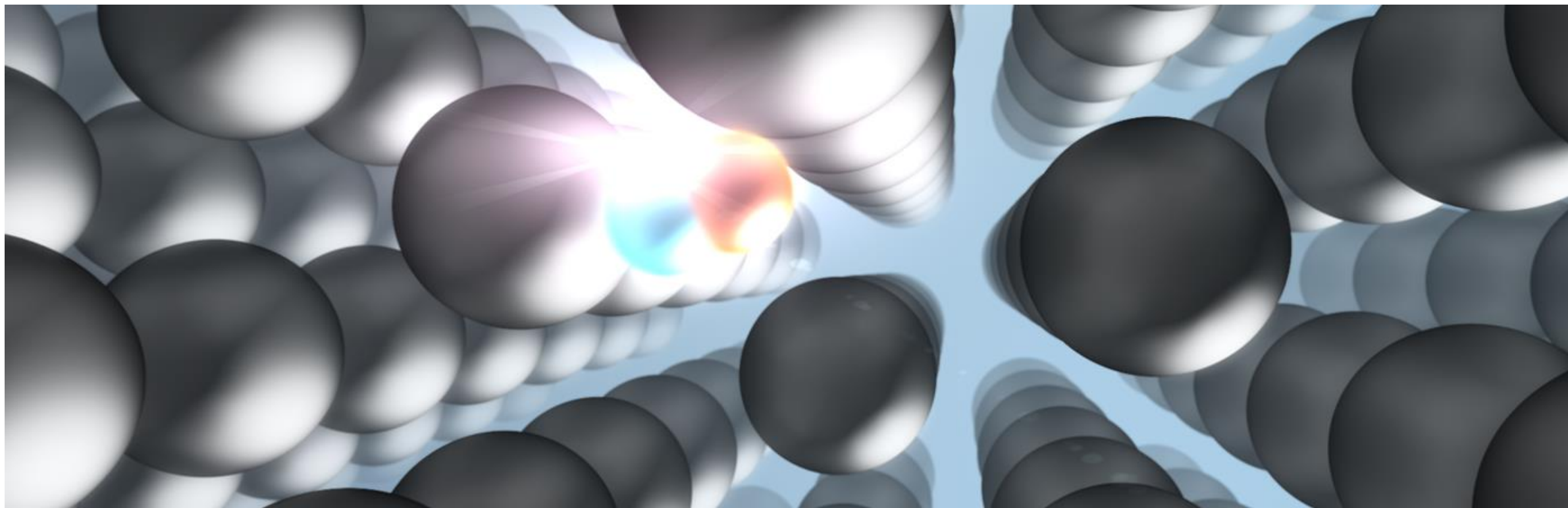




11th International Workshop on Thin Films and New Ideas for Pushing the Limits of RF Superconductivity - TFSRF2024



Positron annihilation spectroscopy: pursuing point defects in superconducting film

Institute of Radiation Physics · Department of Nuclear Physics · Sebastian Klug · s.klug@hzdr.de · www.hzdr.de



Introduction

M. Sc. Sebastian Klug

- Doctoral Student since May 2024
- HZDR, Institute of Radiation Physics
 - topics: Positron Annihilation Spectroscopy (PAS), SRF cavities, Magnetron Sputtering (DC-/HiPIMS)

Ph.D. project

- role of **defects & defect development** in superconducting thin films
 - impact on superconducting properties
- defects/voids/interfaces created **during** thin film creation
 - **in-situ investigations** of defect dynamics with **PAS**
 - materials: **Nb, NbN, NbTiN, Nb₃Sn**
 - study of **multilayer thin films** (SIS structure)

Introduction

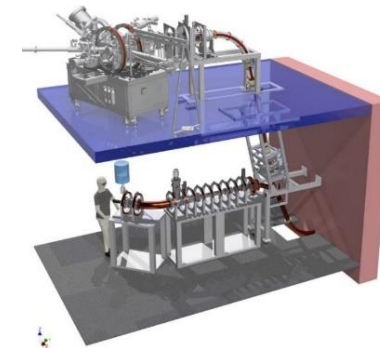
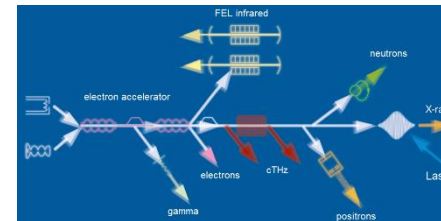
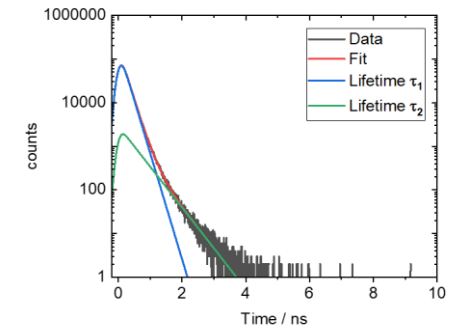
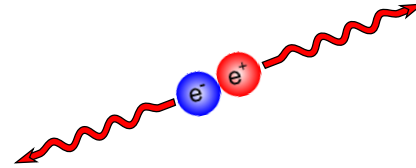
NOVALIS project (Novel accelerator technology for efficient light sources)

- German research project (Federal Ministry of Education and Research of Germany)
- Consortium of different German research institutions:
 - Technical University of Darmstadt (TUDA)
 - University of Hamburg (UHH)
 - University of Siegen (USI)
 - Helmholtz-Zentrum Berlin (HZB)
 - University of Wuppertal (BUW)
 - **Helmholtz-Zentrum Dresden-Rossendorf (HZDR)**
- **AIM: significant reducing of operational power losses of SRF cavities**
 - **Beyond Niobium** research, focus on thin films
 - task of HZDR: providing facilities for **positron annihilation spectroscopy** of thin film samples



Outline

- **Positron Annihilation Spectroscopy**
- **Recent research of point defects**
- **Positron facilities at HZDR**
- **Outlook: Magnetron Sputtering & in-situ PALS**
- **Conclusion**



Positron Annihilation Spectroscopy (PAS)

Positron / DBS / PALS

Positron Annihilation Spectroscopy (PAS)

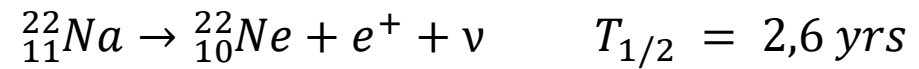
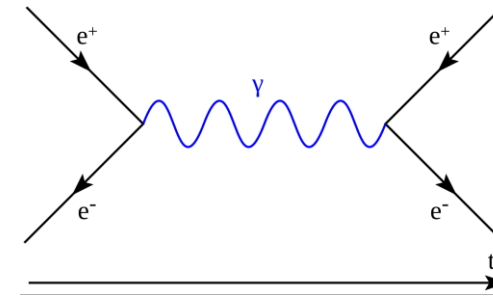
Positron

anti-particle of electron

- same mass, opposite charge
- annihilation with characteristic radiation (**511 keV**)

positron generation

- natural source → **β^+ decay**
- artificial creation → **pair production**



with photons or bremsstrahlung

Positron Annihilation Spectroscopy (PAS)

Positron

Interaction mechanism positron (e^+) \leftrightarrow matter

1) Generation & implantation of e^+

- dependence on implantation energy

2) Thermalization

- <10 ps
- losing of kinetic energy

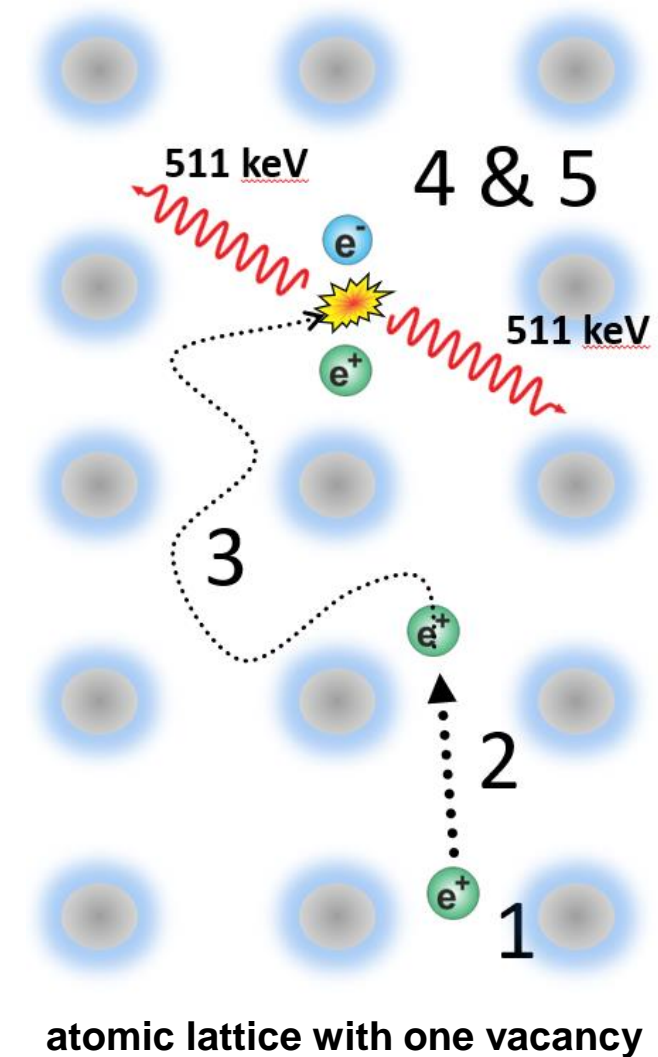
3) Diffusion through the lattice

- 100 ps, up to 100 nm

4) Trapping in defects

- different electron densities

5) Annihilation with electron & Emission of photons



Positron Annihilation Spectroscopy (PAS)

Positron

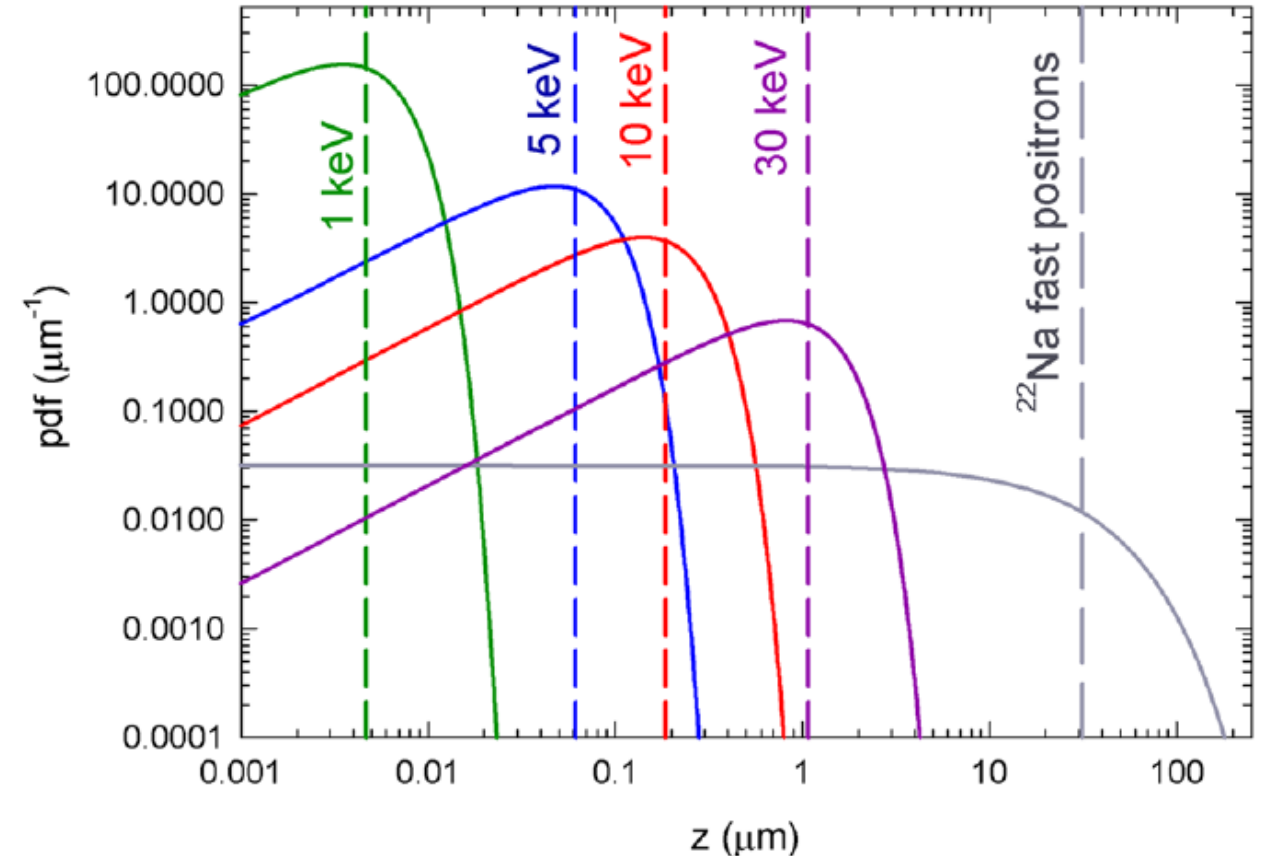
Implantation (Makhov) profile

- mean positron penetration depth $\langle z \rangle$

$$\langle z \rangle [\text{nm}] = \frac{36}{\rho [\text{gcm}^{-3}]} E_p^{1.62} [\text{keV}]$$

- dependent on E_p and ρ
- **low** implantation energy E_p
 - surface-near implantation
 - sharp profile
- **high** implantation energy E_p
 - deeper implantation
 - broadening of profile (positron scattering)

- Optimal thickness of examination: **50 - 300 nm**



Makhov profile for Nb

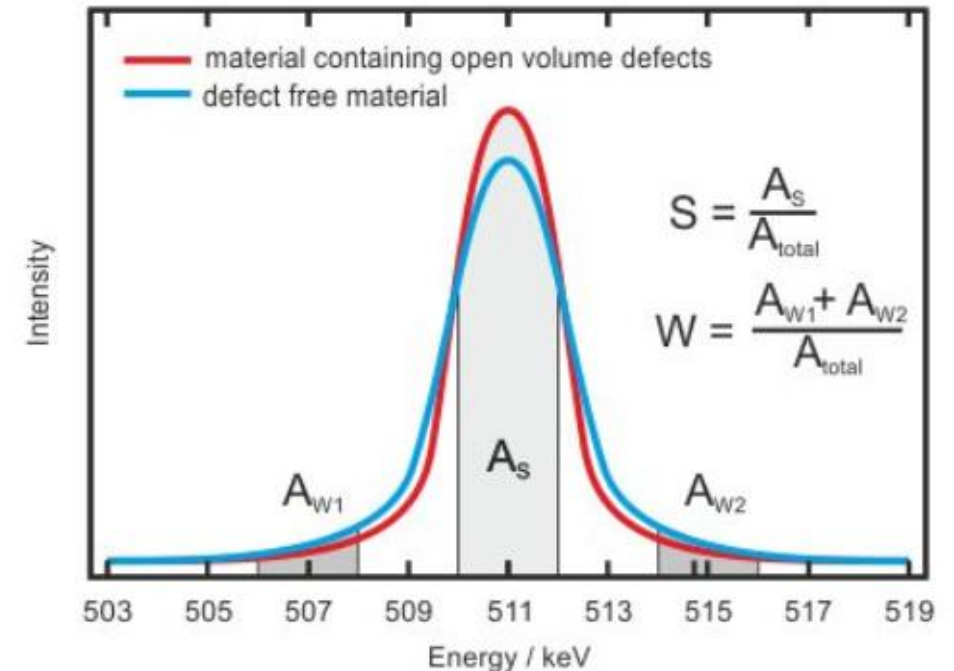
Wenskat, M. et al. Scientific Reports 10(2020), 8300

Positron Annihilation Spectroscopy (PAS)

Doppler-Broadening Spectroscopy (DBS)

Doppler broadening of annihilation line (511 keV)

- electron momentum → energy shift
- different fractions of electrons
- **low electron momentum** (valence electrons)
 - **S** (shape) parameter → **defect concentration**
- **high electron momentum** (core electrons)
 - **W** (wing) parameter → **atomic environment** of defects



annihilation line with **S** and **W** parameters

Positron Annihilation Spectroscopy (PAS)

Positron Annihilation Lifetime Spectroscopy (PALS)

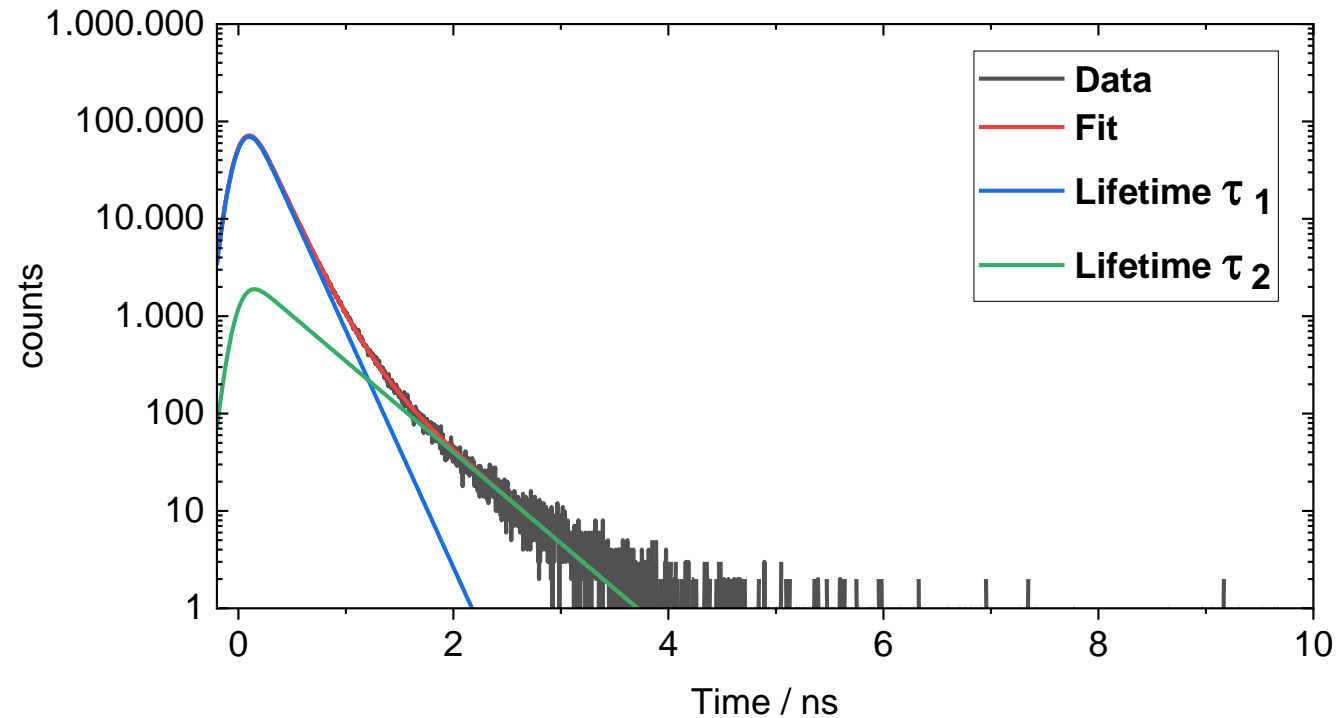
Lifetime of positrons

- time between generation and annihilation of positrons

Spectrum of positron lifetime

- **nonlinear** fitting process required
- convolution with **time resolution function**
- exponential decay terms ($i \in \mathbb{N}$)
 - **lifetime components** τ_i (defect size)
 - **intensity** I_i (defect density)

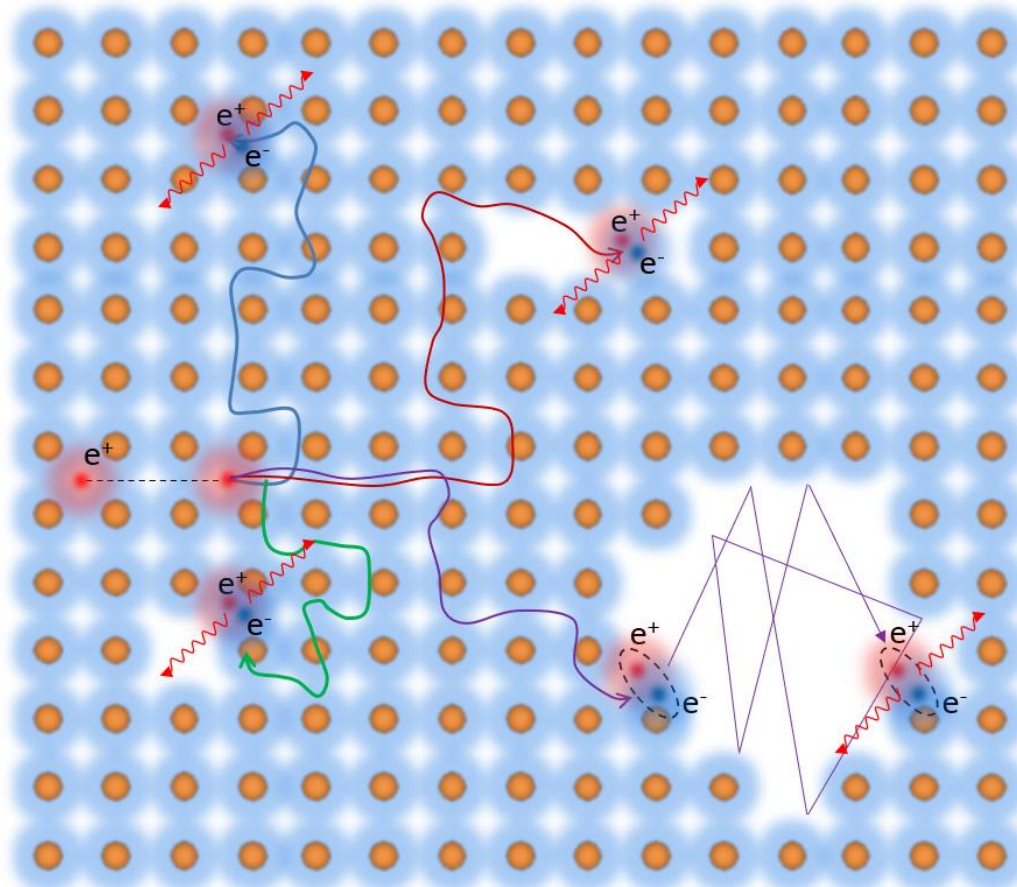
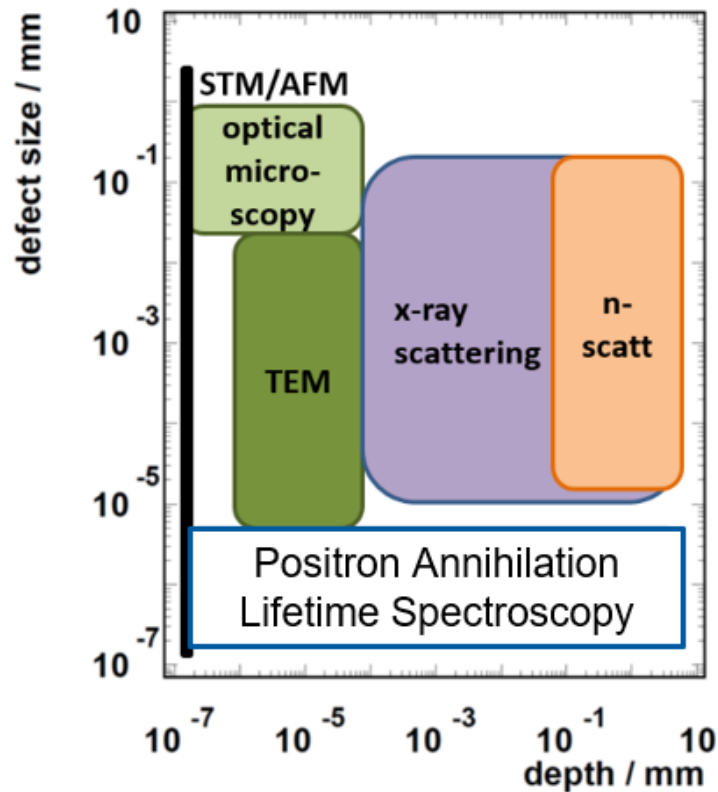
$$N(t) = \sum_{i=1}^{k+1} \frac{I_i}{\tau_i} \exp\left(-\frac{t}{\tau_i}\right)$$



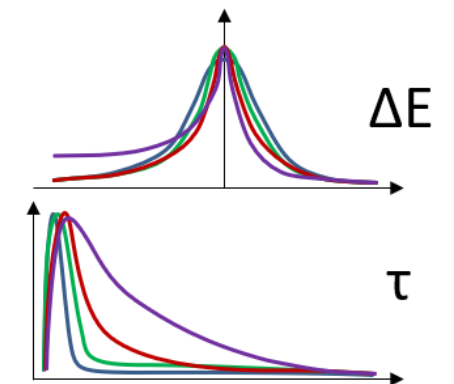
Positron Annihilation Spectroscopy (PAS)

Positron Annihilation Lifetime Spectroscopy (PALS)

Non-destructive and high-sensitivity to small defects (atomic scale)



- bulk
- mono-vacancy
- vacancy agglomeration
- void / pore



Recent research of point defects

Vacancy kinetics in Nb / Point defects of Nb-based thin films

Vacancy kinetics in Nb

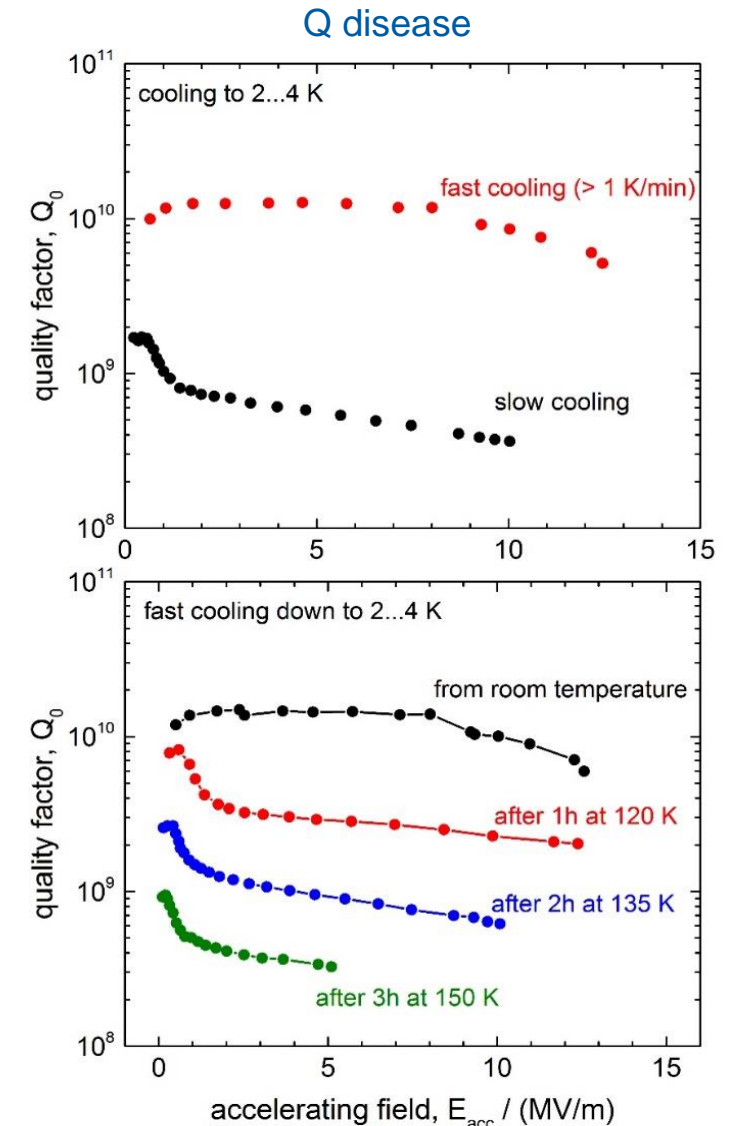
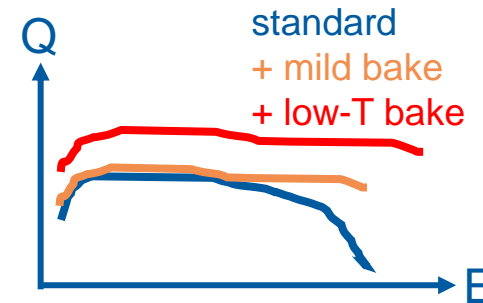
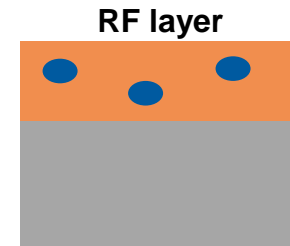
Collaboration with M. Wenskat (DESY, DE) & J. Čížek (Prague, CZ)

Origin of Q disease

- formation of **niobium hydrides** in the rf surface layer during cooling down
- **open volume lattice defects** have high trapping potential for impurities, especially hydrogen: **vacancy-hydrogen complexes**

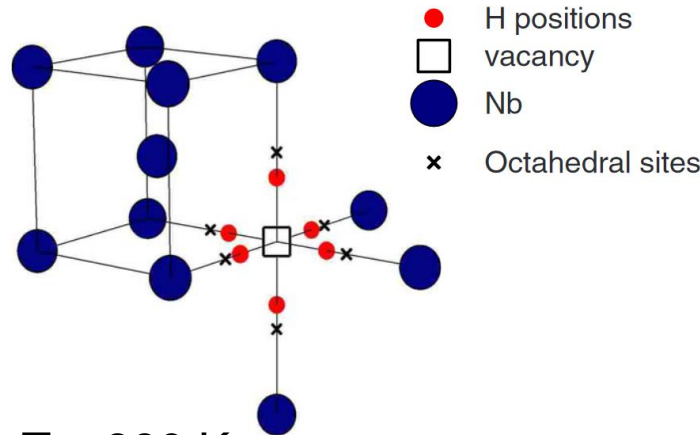
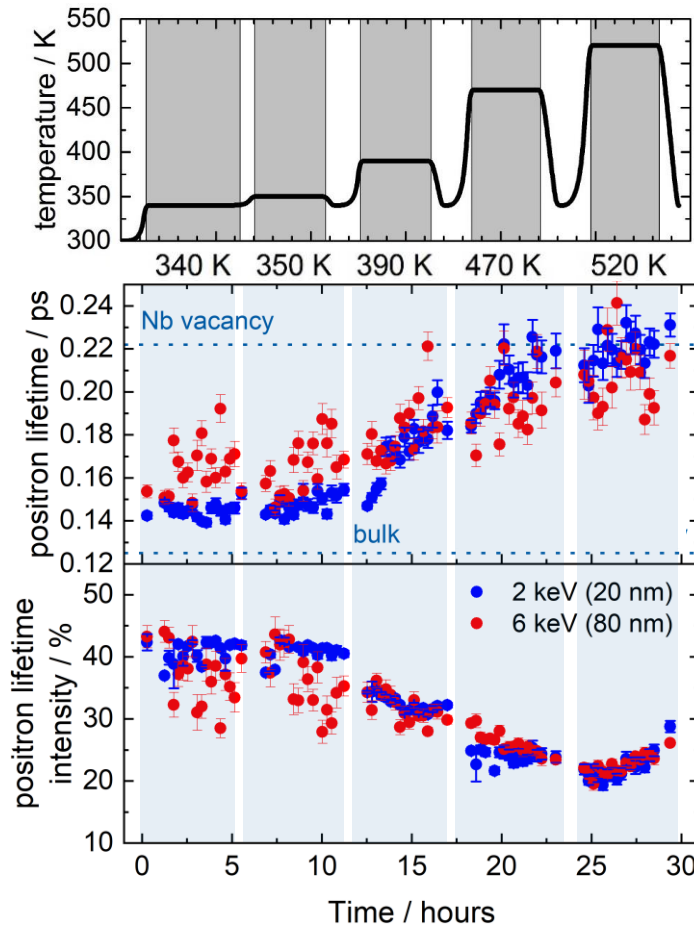
Curing Q disease

- **mild bake**: at 390 K for 48 h ($< 1e-6$ mbar) to avoid high-field Q slope (accidentally found!)
- **low-T bake**: only 350 K for first 2 h
Q losses reduced by factor 2
increased achievable accelerating field by $>10\%$

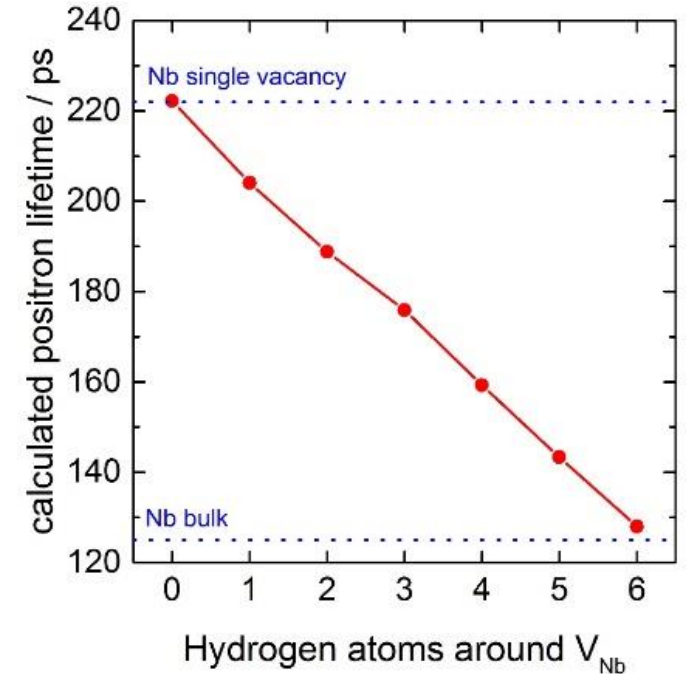


Vacancy kinetics in Nb

Collaboration with M. Wenskat (DESY, DE) & J. Čížek (Prague, CZ)



- T = 390 K:
shows most significant changes
(already after short times)
- T > 390 K:
hydrogen released from complexes (lifetime increases)
v+nH concentration decreases (intensities decrease)



Wenskat, M. et al. Phys. Rev. B 106(2022), 094516
Wenskat, M. et al. Scientific Reports 10(2020), 8300

Point defects of Nb-based thin films

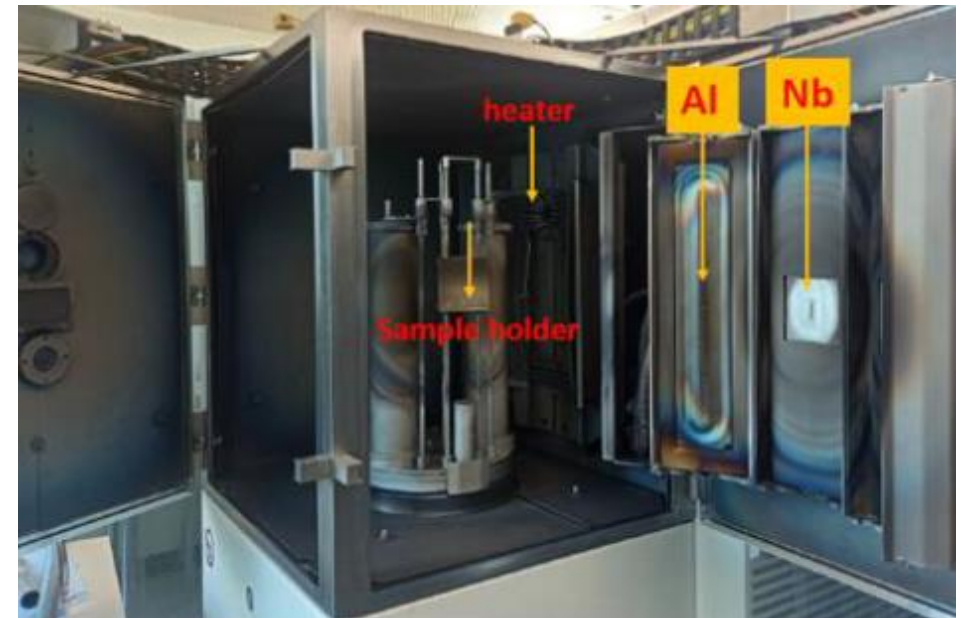
Magnetron sputtering

Thin film creation

- cooperation with **University of Siegen, Germany**
 - Prof. Dr. rer. nat. habil. Xin Jiang & Dr. Aleksandr Zubtsovskii
 - NOVALIS project
- **knowledge transfer** for new sputtering chamber @HZDR
- **reference samples** for future sample series

Sample characterization @HZDR, Germany

- XRD (θ - 2θ scan)
- DBS & PALS

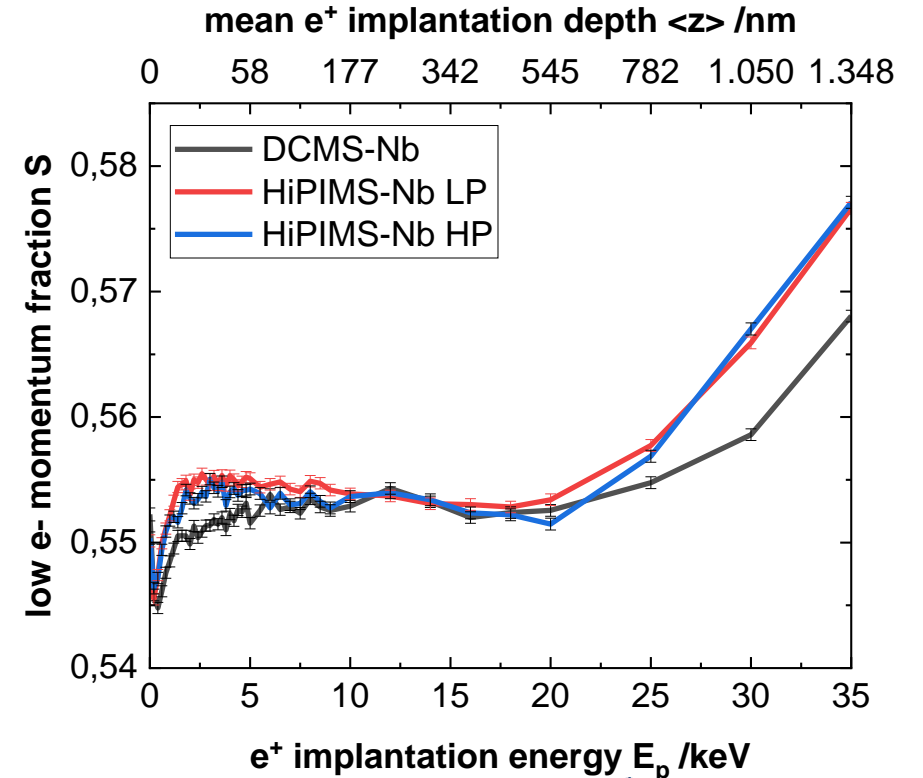
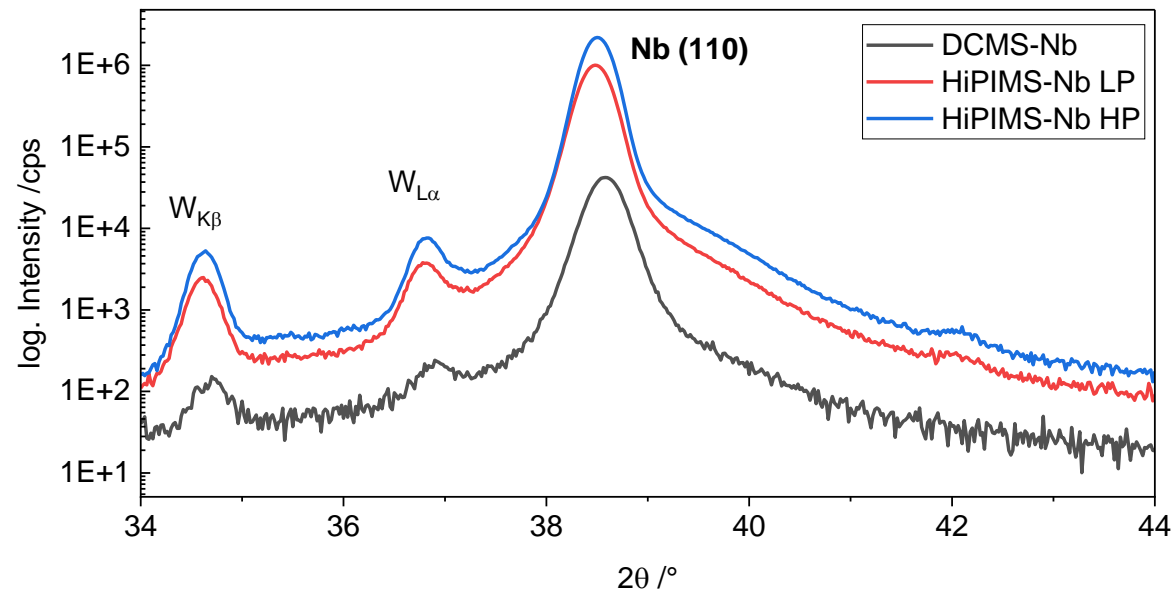


CemeCon CC800 at University of Siegen

Point defects of Nb-based thin films

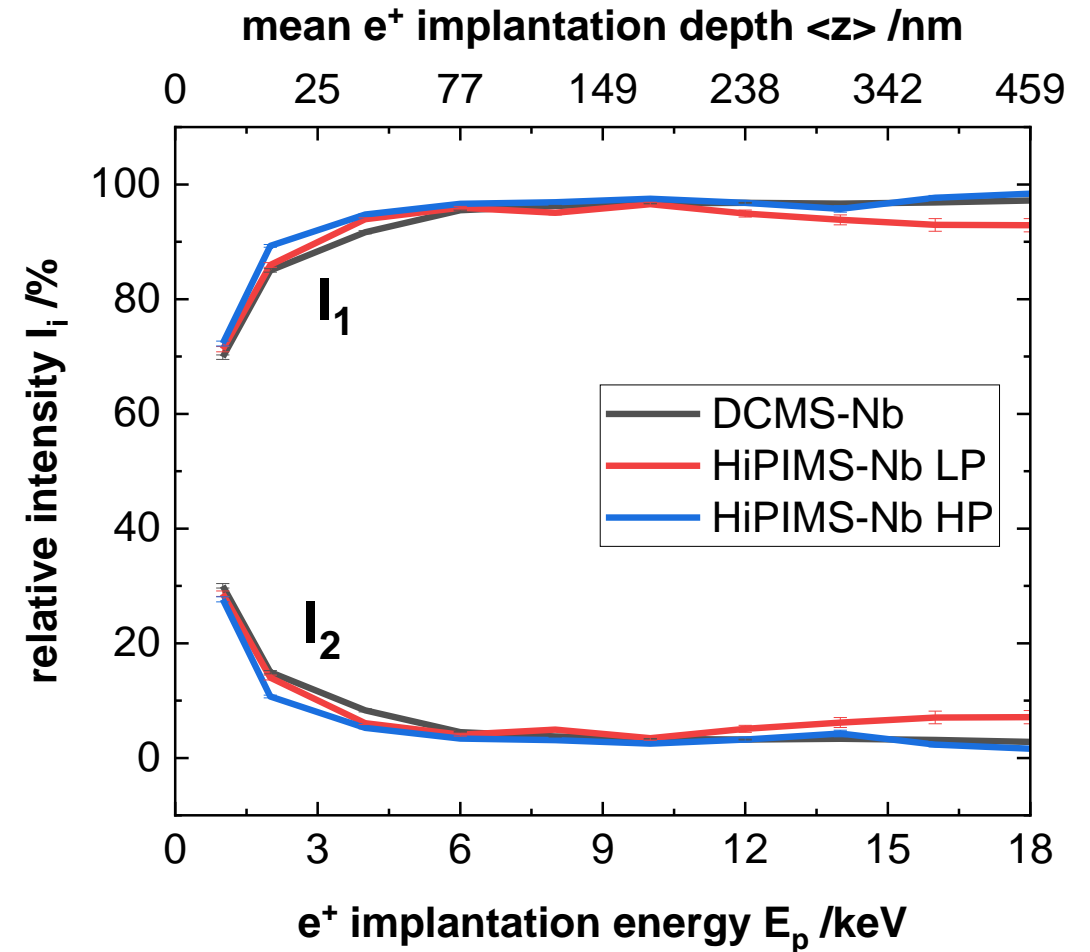
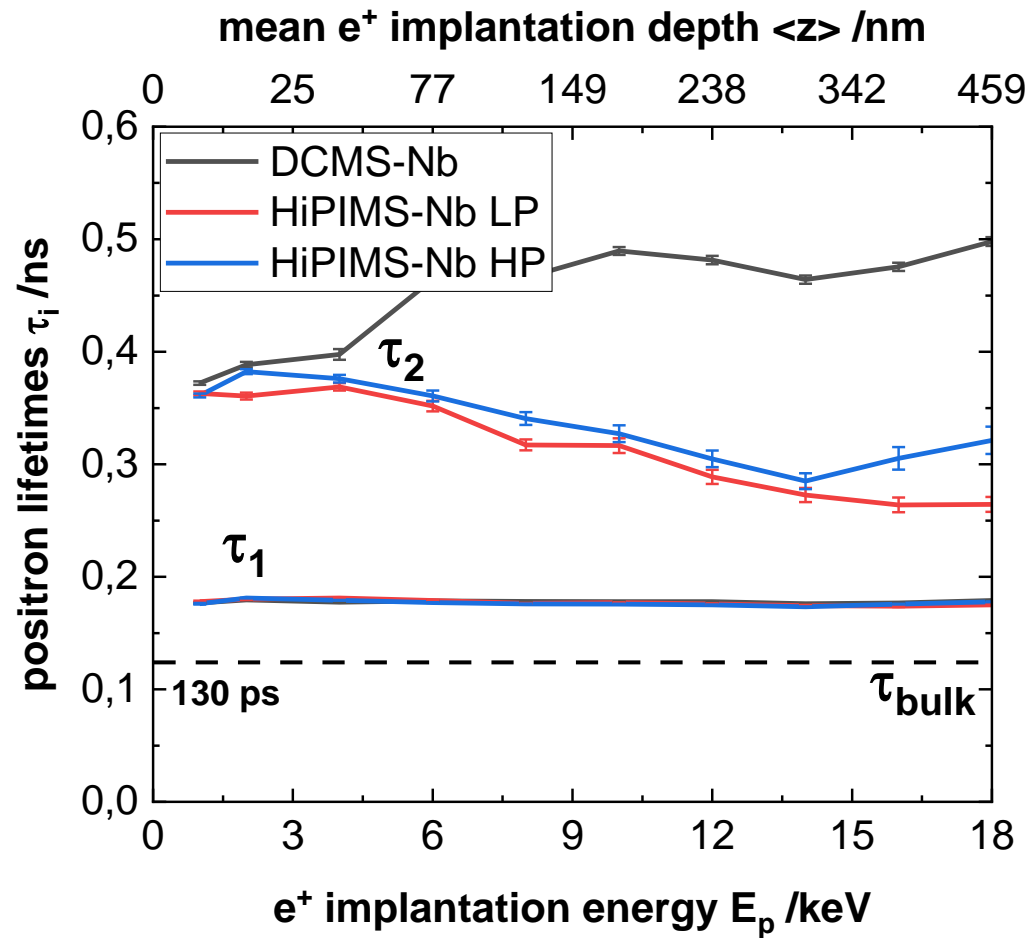
Nb films on Si(100)

sample	cathode type	Power (W/cm ²)	p_gas (mbar)	Ar flow (sccm)	N2 flow (sccm)
DCMS-Nb	DC	4,54	0,00801	410,0	0,0
HiPIMS-Nb LP	HiPIMS	4,54	0,00801	500,0	0,0
HiPIMS-Nb HP	HiPIMS	6,82	0,02001	700,0	0,0



Point defects of Nb-based thin films

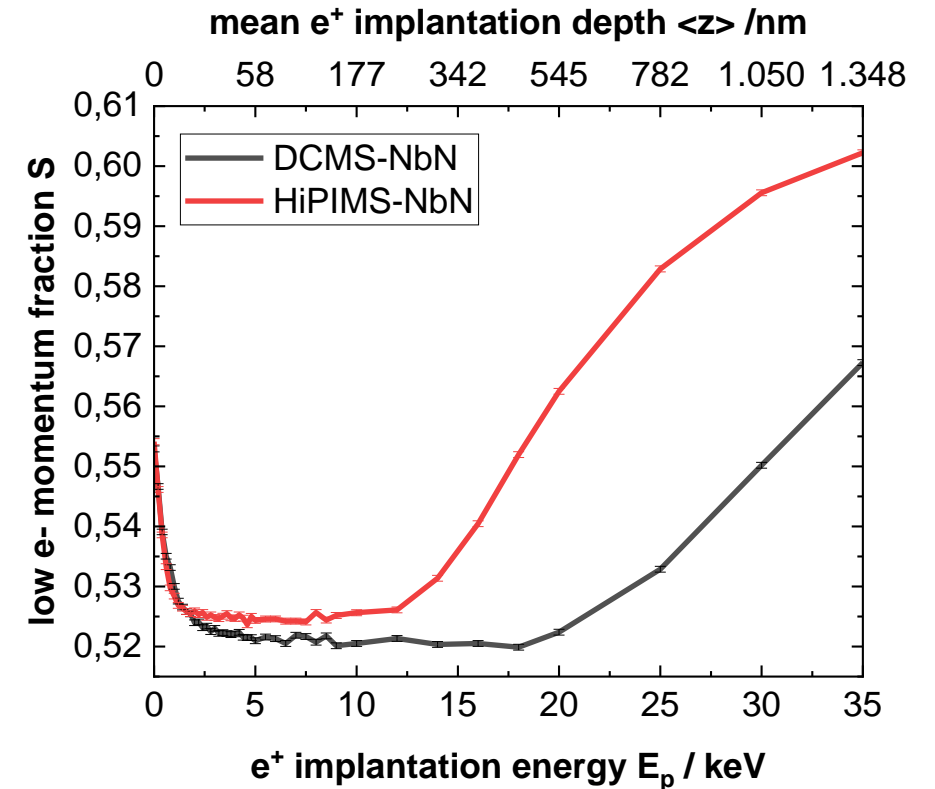
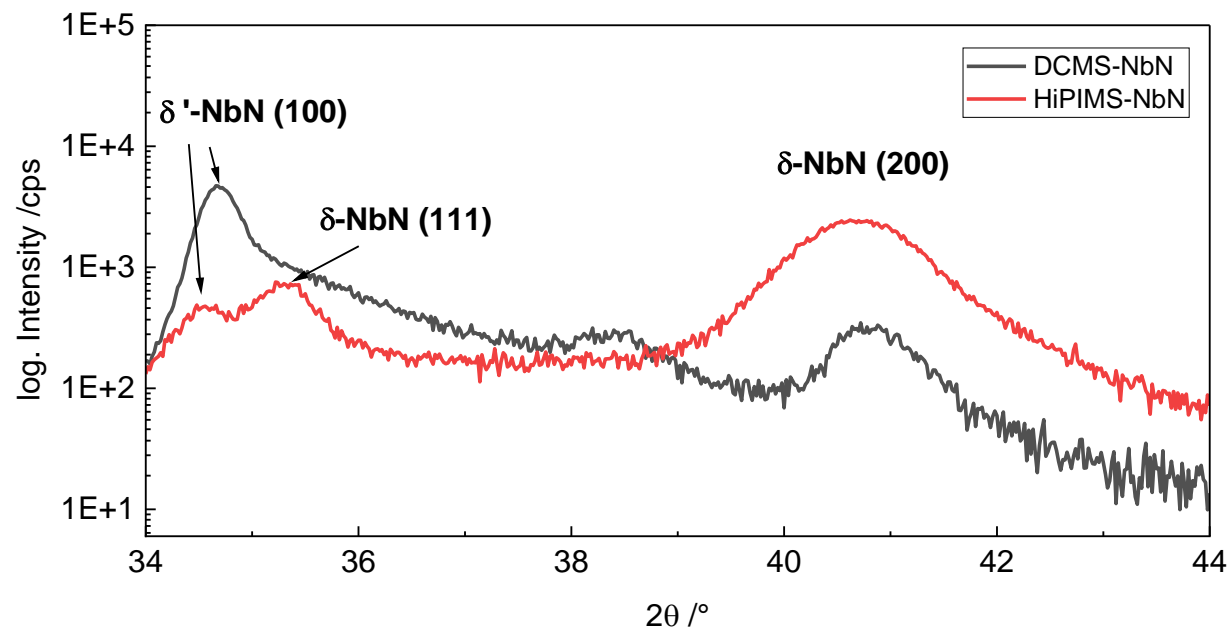
Nb films on Si(100)



Point defects of Nb-based thin films

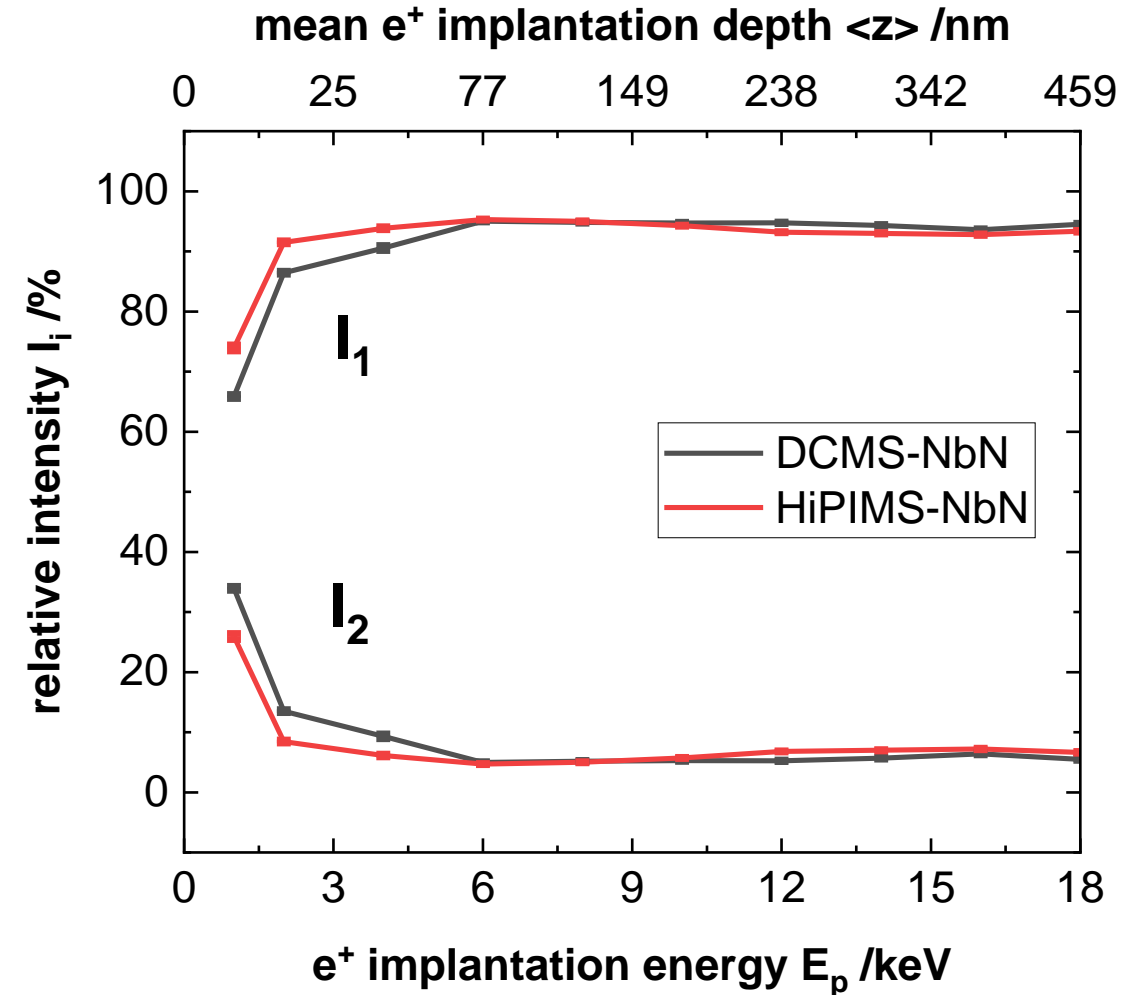
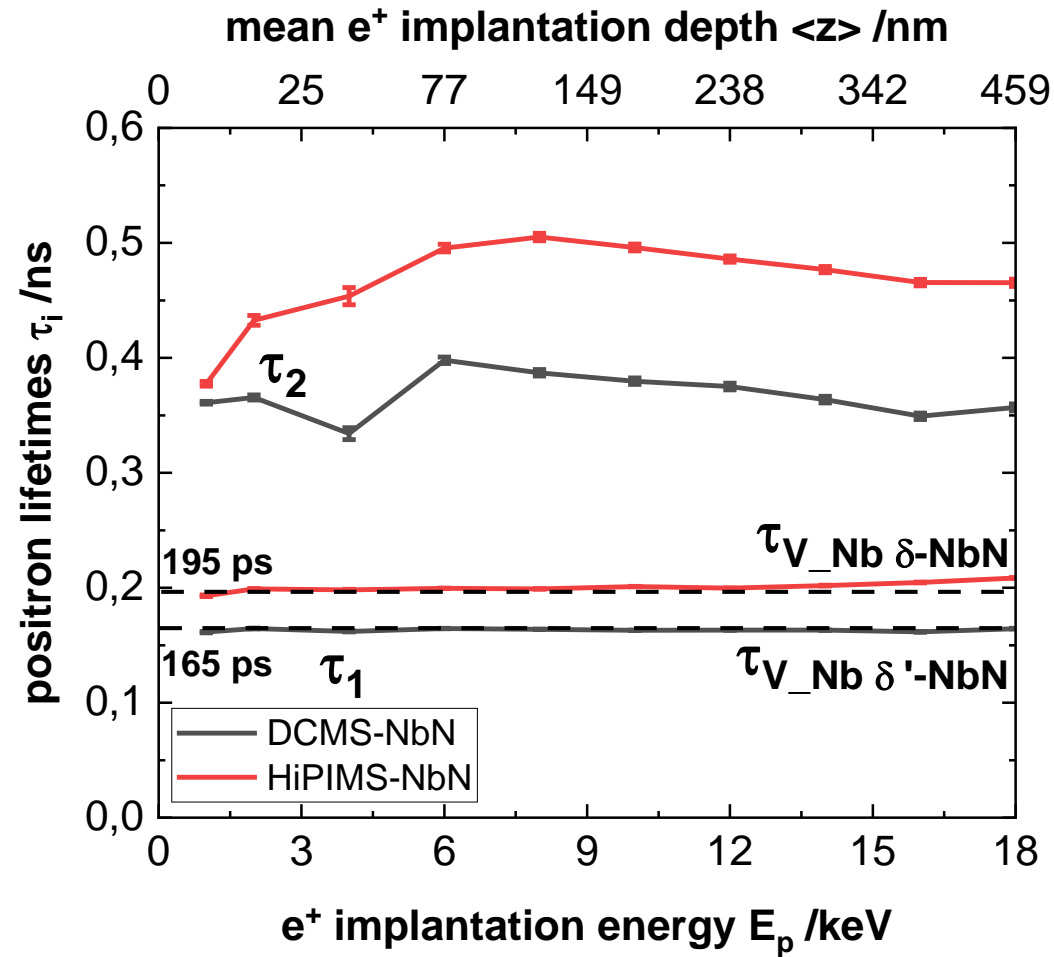
NbN films on Si(100)

sample	cathode type	Power (W/cm ²)	p_gas (mbar)	Ar flow (sccm)	N2 flow (sccm)
DCMS-NbN	DC	5,68	0,01315	522,5	45,5
HiPIMS-NbN	HiPIMS	4,54	0,02291	641,8	71,3



Point defects of Nb-based thin films

NbN films on Si(100)



Positron user facilities at the HZDR

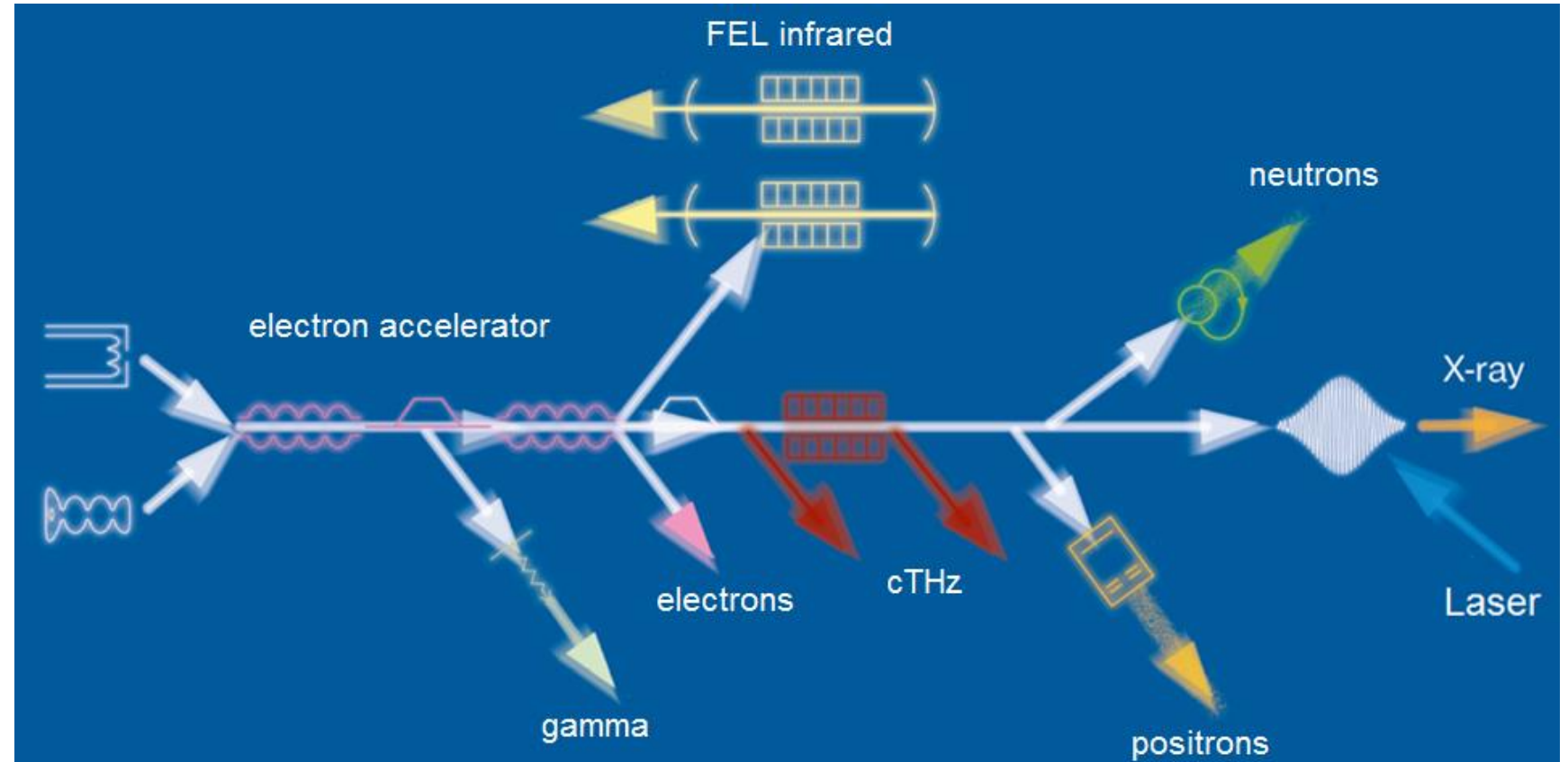
ELBE / SPONSOR / MePS / Access for user

Positron user facilities at the HZDR

ELBE (Electron Linear accelerator with high Brilliance and low Emittance)

ELBE as User Facility

- > 50 % of beamtime for **external user groups**



Positron user facilities at the HZDR

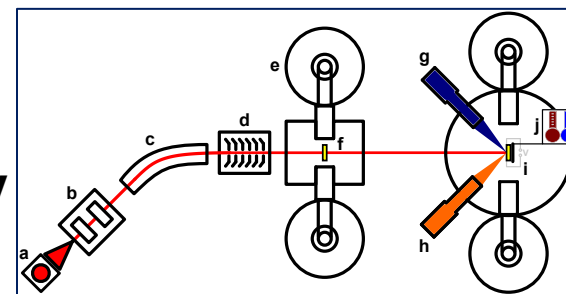
SPONSOR (The Slow-Positron System of Rossendorf)

Usage of ^{22}Na source

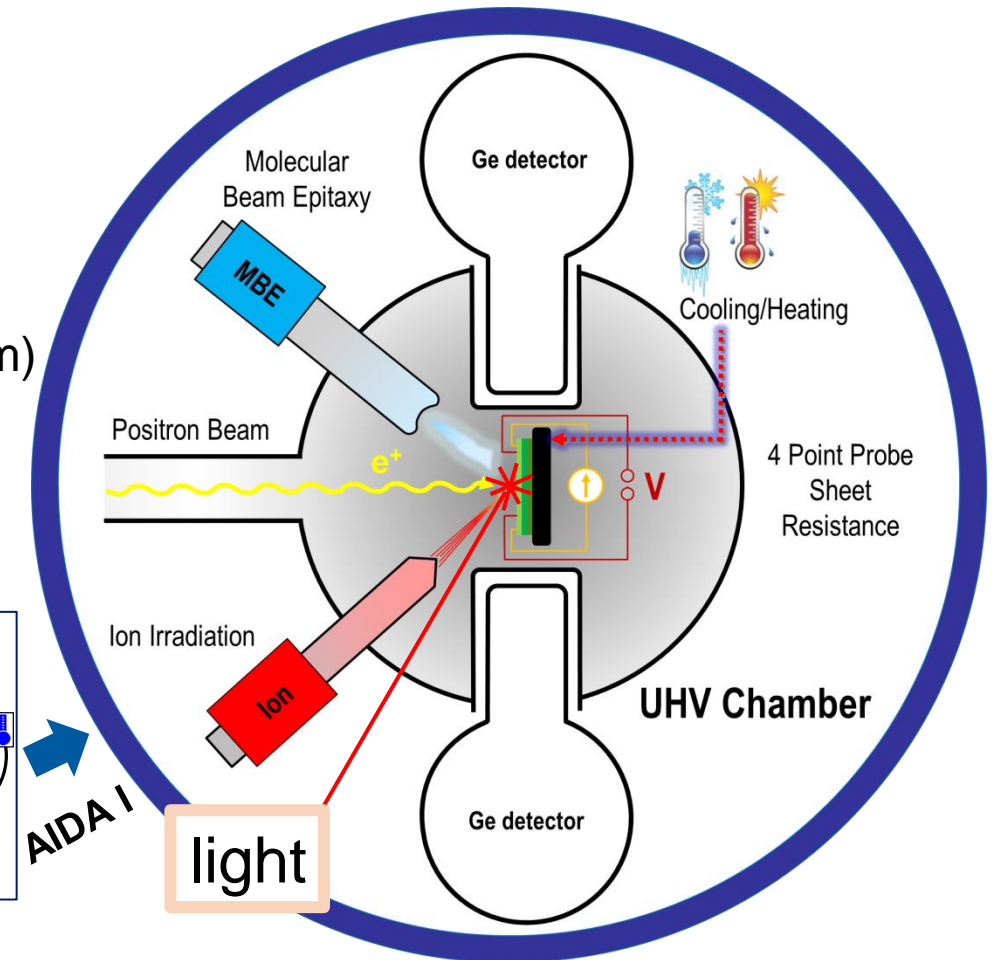
- **DBS** (defect size, density and chemistry)
- **depth resolved** measurements
- implantation energy (E_p) **0...35 keV** (depth ~20 nm to few μm)

Apparatus for in-situ defect analysis (AIDA I)

- base pressure: **$2\text{e-}9$ mbar**
- temperature: **100 - 1200 K**
- Molecular Beam Epitaxy (**MBE**)
- ion irradiation: $E_{\text{ion}} = 0,5...5,0$ keV
- sheet resistance (**4 point probe**)
- sample bias (**± 1000 V**)



SPONSOR

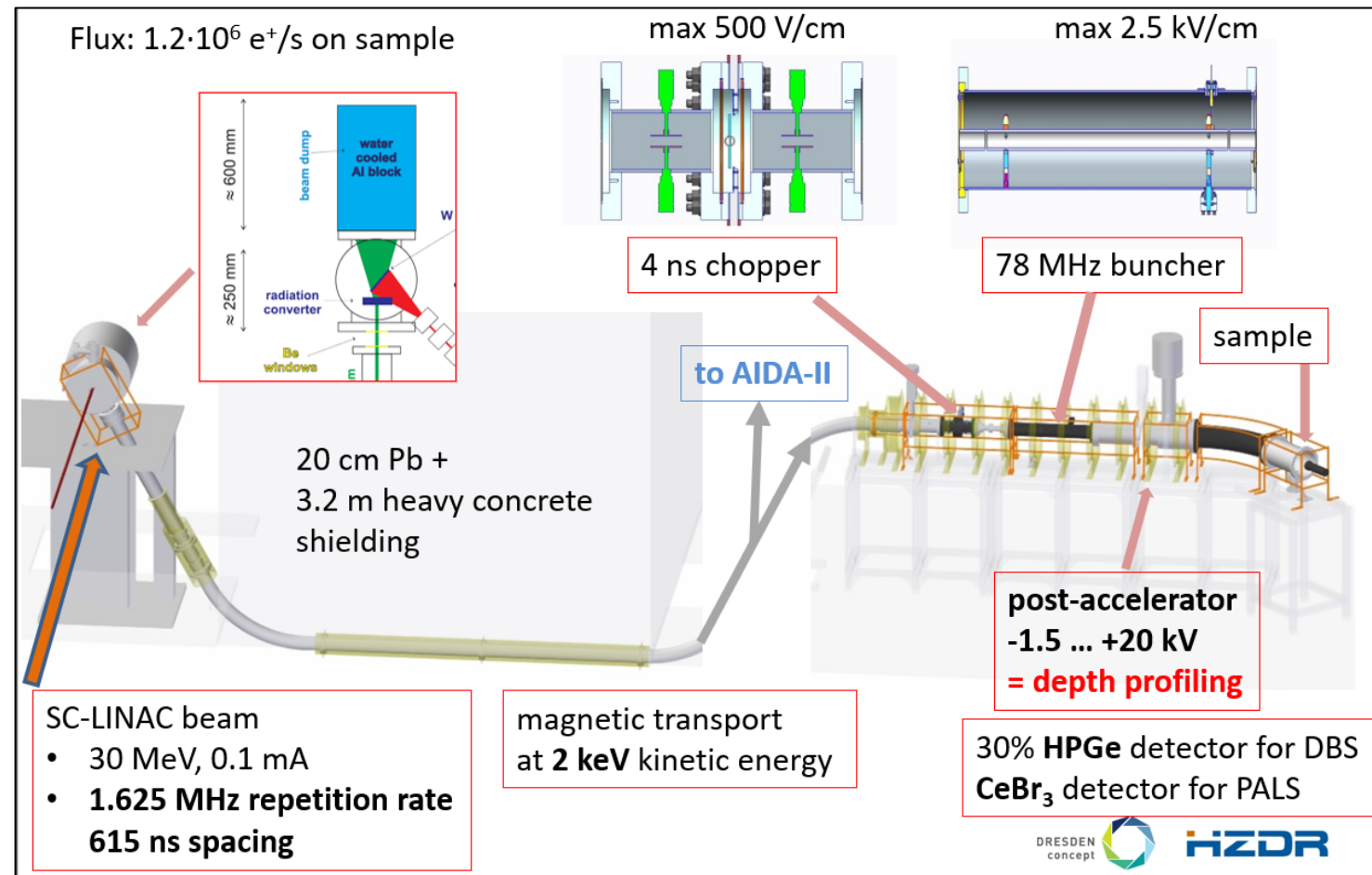


Positron user facilities at the HZDR

MePS (Mono-energetic Positron Spectroscopy)

Usage of Linac

- **PALS** (defect size and density)
- **depth resolved** measurements
- implantation energy (E_p) **0...18 keV**
- temperature **up to 800 K**
- planned implantation of cryostat (**20 K**)



Positron user facilities at the HZDR

Access for user

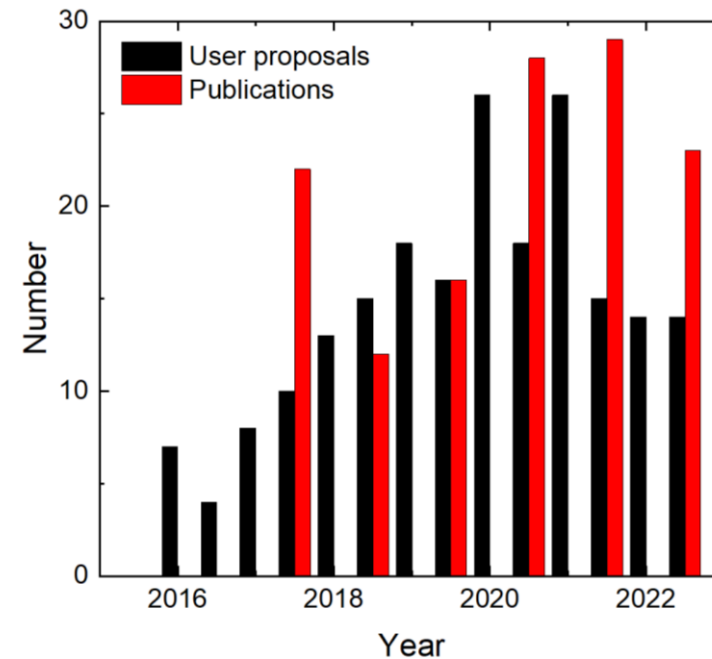
Application with scientific user proposal

- submitted through the user portal **GATE**
- evaluation by international and interdisciplinary Scientific Advisory Committee
- free of charge for all non-proprietary research

- Website <https://www.hzdr.de/db/Cms?pNid=1732>

Deadline

- twice a year
- next call for 1st half 2025:
 - **September 23rd, 2024** (already next Monday!)



~ 30 proposals / year

Outlook: Magnetron Sputtering & in-situ PALS

AIDA II / future plans

Outlook: Magnetron Sputtering & in-situ PALS

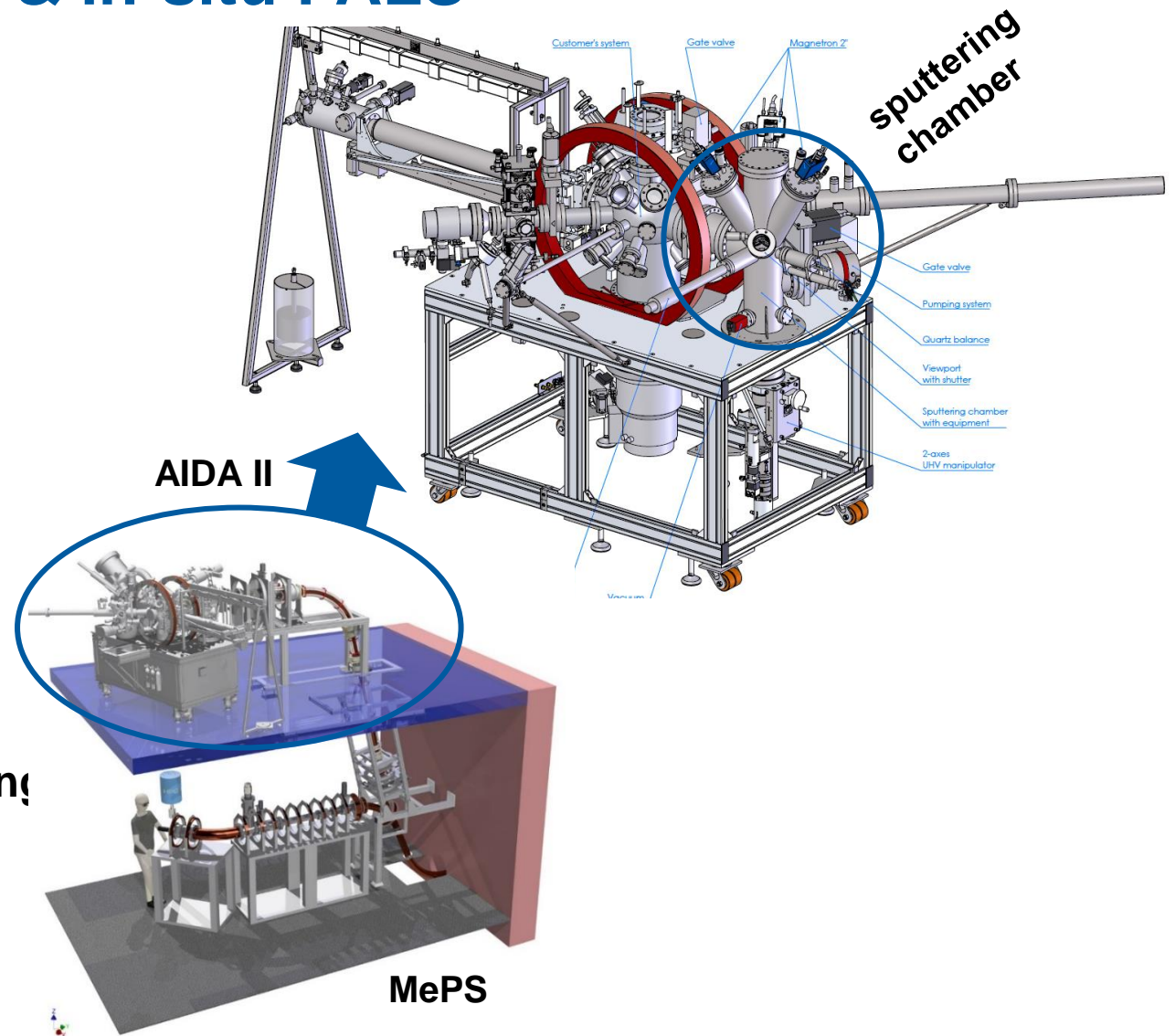
AIDA II

Magnetron sputter chamber

- 3 confocal magnetrons: 2x DC, 1x RF; 400 W
- working gases: Ar, N₂, O₂
- **2-inch** targets
- base pressure < 1e-9 mbar
- temperature range -180 to 800 °C

In-situ characterization (PALS)

- Apparatus for in-situ defect analysis (**AIDA II**)
- combination of **MePS** and magnetron sputtering
- novelty: **PALS during sputtering process**
 - tracking of development of point defects



Outlook: Magnetron Sputtering & in-situ PALS

future plans

next steps

- finishing work on **AIDA II**
- investigations of **influence of magnetron sputtering process parameters**
- in-situ PALS of **Nb** and **NbN** thin films
- ex-situ characterization with **XRD, SEM, AFM**
- characterization of **superconducting properties**

later

- Study of **NbTiN, Nb₃Sn** and final aim of **SIS multilayers**
- **Cu (interdiffusion studies)**

Conclusion

Positron Annihilation Spectroscopy

Conclusion

Positron Annihilation Spectroscopy

- **non-destructive** characterization of defects
- vacancy-like defects **down to atomic scale**
- **depth profiling**
- combination with **ATSUP** calculations

- Doppler-Broadening-Spectroscopy (**DBS**)
 - **defect concentration & atomic environment** of defects

- Positron Annihilation Lifetime Spectroscopy (**PALS**)
 - **defect typ, size & density**

Acknowledgement

Contact:
Sebastian Klug

+49 351 260 3127
s.klug@hzdr.de

***THANK YOU FOR
YOUR ATTENTION !***



ELBE as user facility!

Institute of Radiation Physics, HZDR, Germany

- Dr. Andreas Wagner
 - Dr. Maciej Oskar Liedke
 - Dr. Ahmed Gamal Attallah Elsherif
 - Dr. Maik Butterling
 - Dr. Eric Hirschmann
- and more...

Institute of Materials Engineering, University of Siegen, Germany

- Prof. Dr. rer. nat. habil. Xin Jiang
 - Dr. Aleksandr Zubtsovskii
 - M.Sc. Bharath Reddy Lakki Reddy Venkata
- and more...