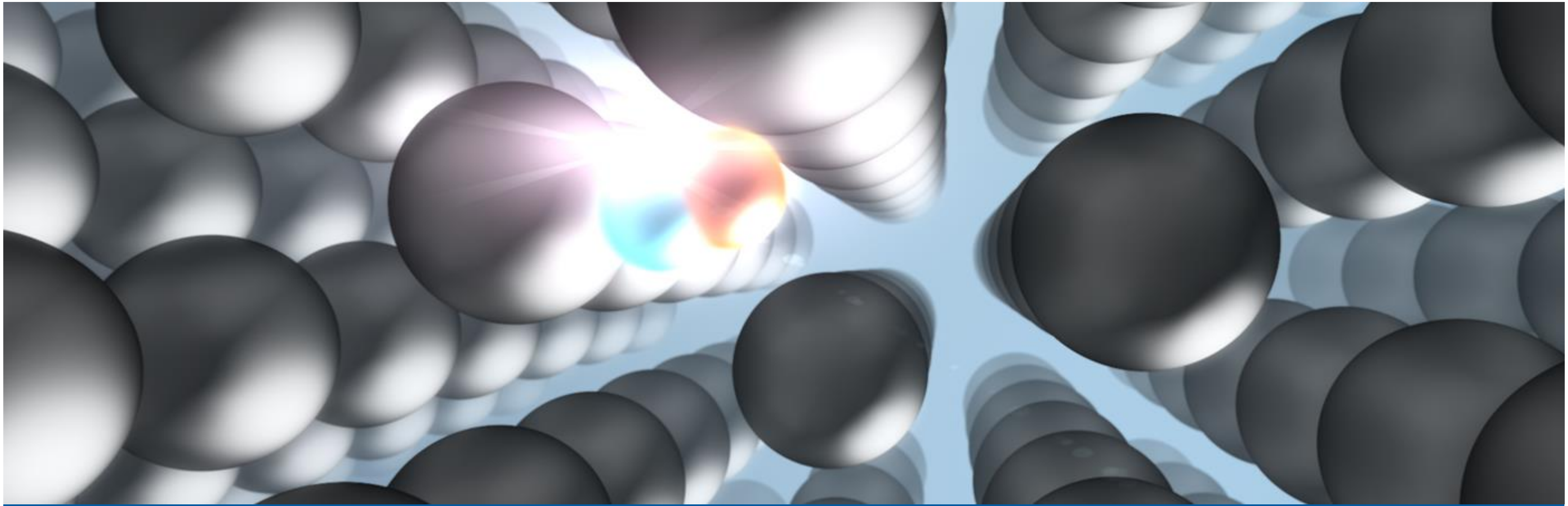
11th International Workshop on Thin Films and New Ideas for Pushing the Limits of RF Superconductivity - TFSRF2024

UNIVERSITÄT SIEGEN

HZDR
HELMHOLTZ ZENTRUM DRESDEN ROSSENDORF



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

DRESDEN concept

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, Nb_3Sn**
- study of **multilayer thin films** (SIS structure)

Introduction

NOVALIS project (Novel accelerator technology for efficient light sources)

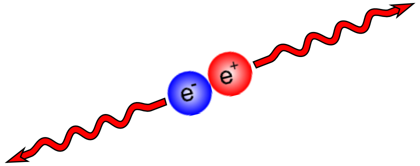
- German research project (Federal Ministry of Education and Research of Germany)
- Consortium of different German research institutions:
  - Technical University of Darmstadt (TUDA)
  - University of Hamburg (UHH)
  - University of Siegen (USI)
  - Helmholtz-Zentrum Berlin (HZB)
  - University of Wuppertal (BUW)
  - **Helmholtz-Zentrum Dresden-Rossendorf (HZDR)**

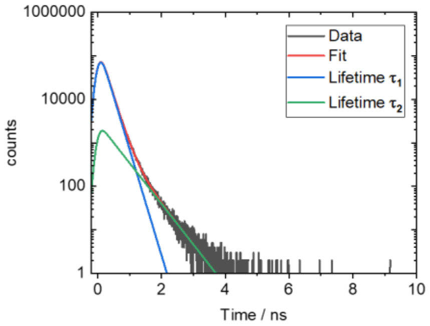
Federal Ministry of Education and Research

- **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

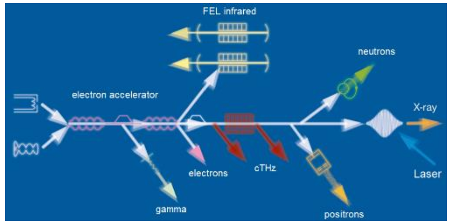
Outline

- **Positron Annihilation Spectroscopy**

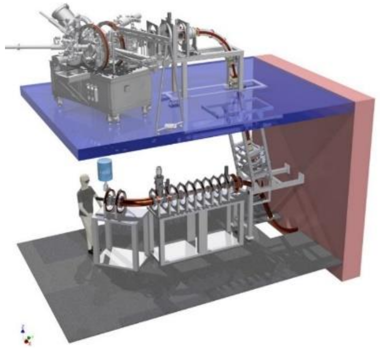




- **Recent research of point defects**



- **Positron facilities at HZDR**



- **Outlook: Magnetron Sputtering & in-situ PALS**

- **Conclusion**

Positron Annihilation Spectroscopy (PAS)

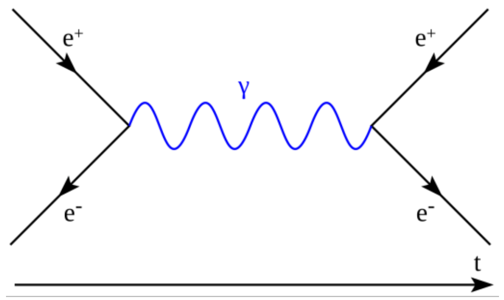
Positron / DBS / PALS

Positron Annihilation Spectroscopy (PAS)

Positron

anti-particle of electron

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



positron generation

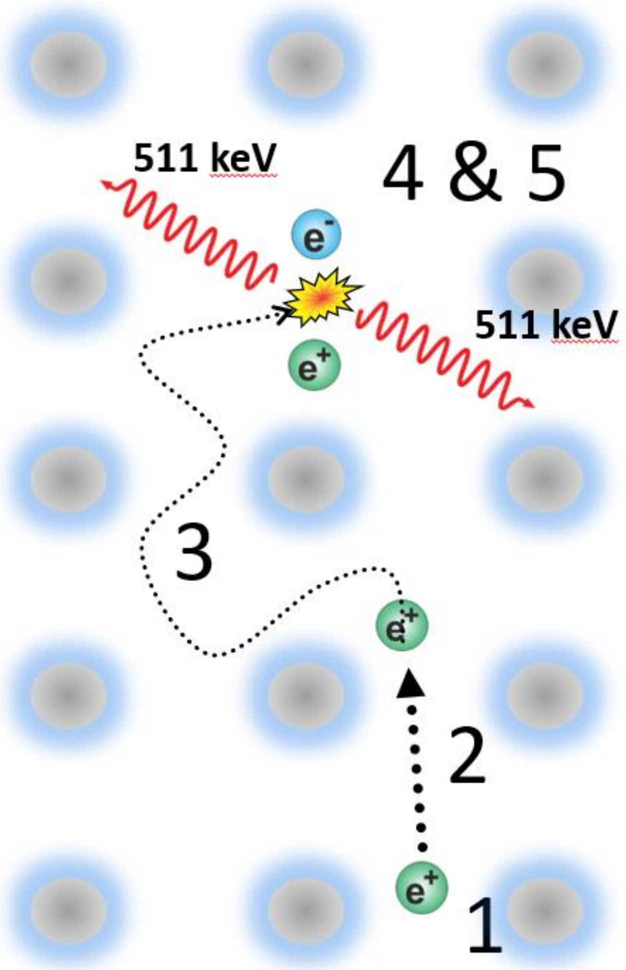
- natural source → **β⁺ decay** $\ {}^{22}_{11}Na \rightarrow {}^{22}_{10}Ne + e^{+} + \nu \qquad T_{1/2} = 2{,}6\ yrs$
- artificial creation → **pair production** with photons or bremsstrahlung

Positron Annihilation Spectroscopy (PAS)

Positron

Interaction mechanism positron (e^+) ←→ 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



atomic lattice with one vacancy

Positron Annihilation Spectroscopy (PAS)

Positron

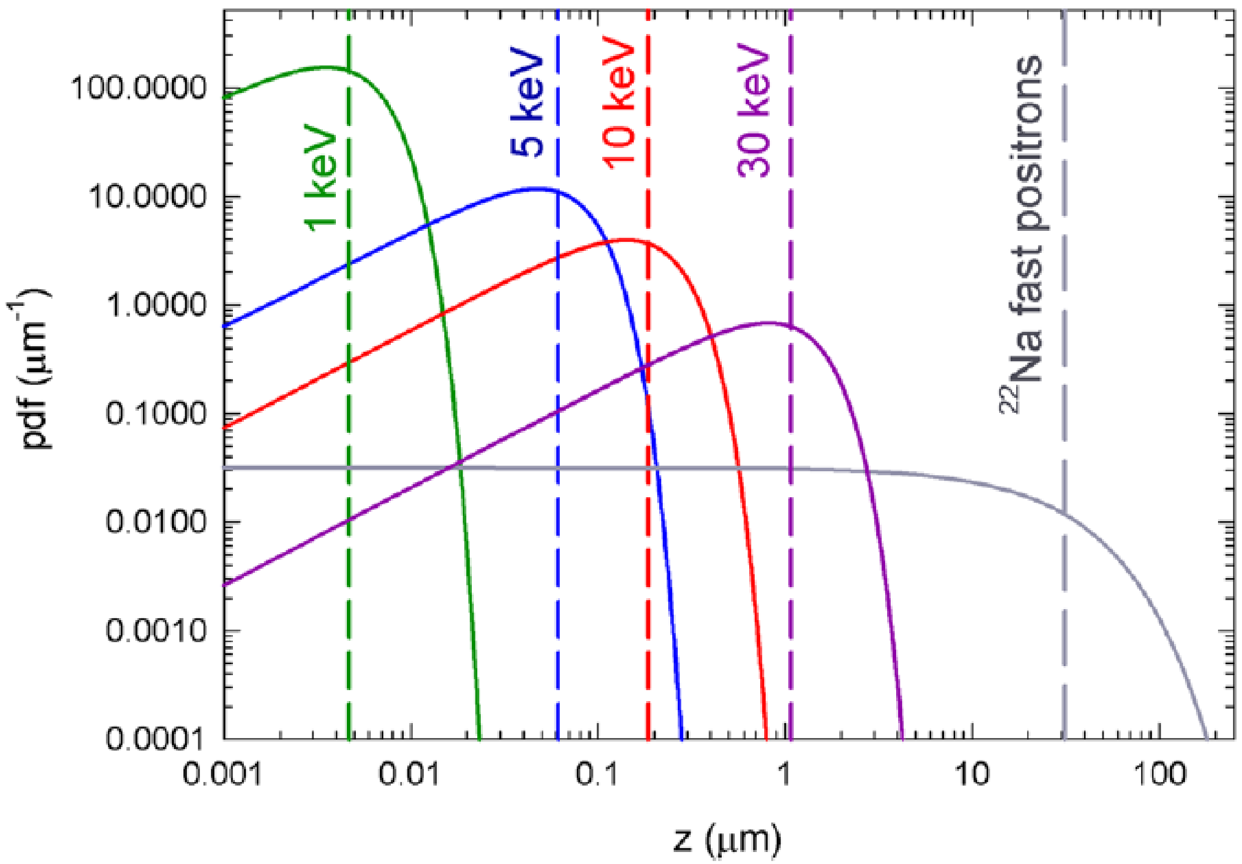
Implantation (Makhov) profile

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

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

- dependent on $\boldsymbol{E_{\mathrm{p}}}$ 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**



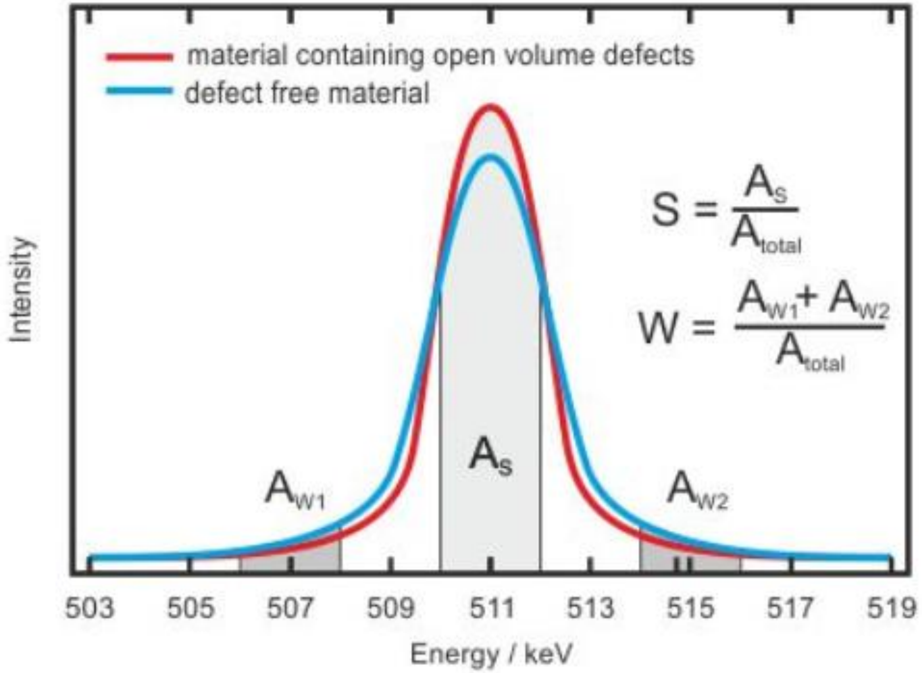
Makhov profile for Nb
Wenskat, M. et al. Scientific Reports 10(2020), 8300

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)

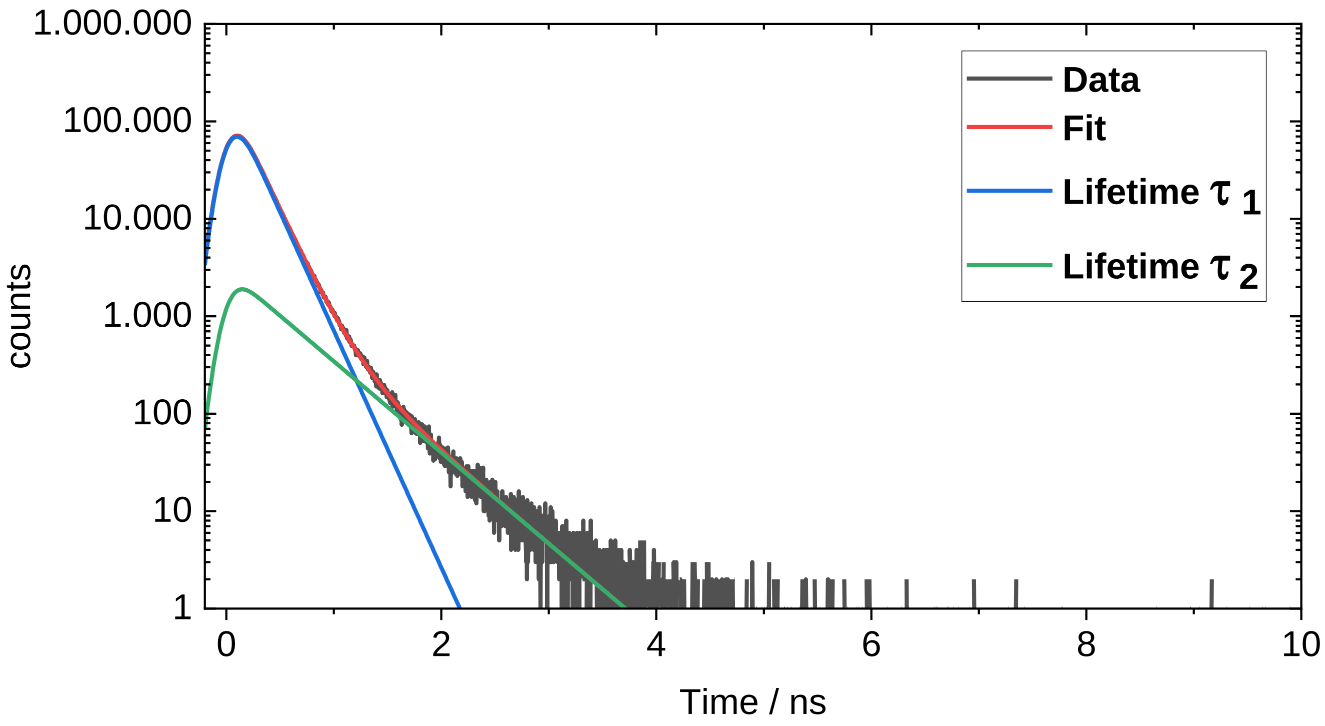
Lifetime of positrons

- 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 $\boldsymbol{\tau}_\mathbf{i}$** (defect size)
  - **intensity $\boldsymbol{I}_\mathbf{i}$** (defect density)

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

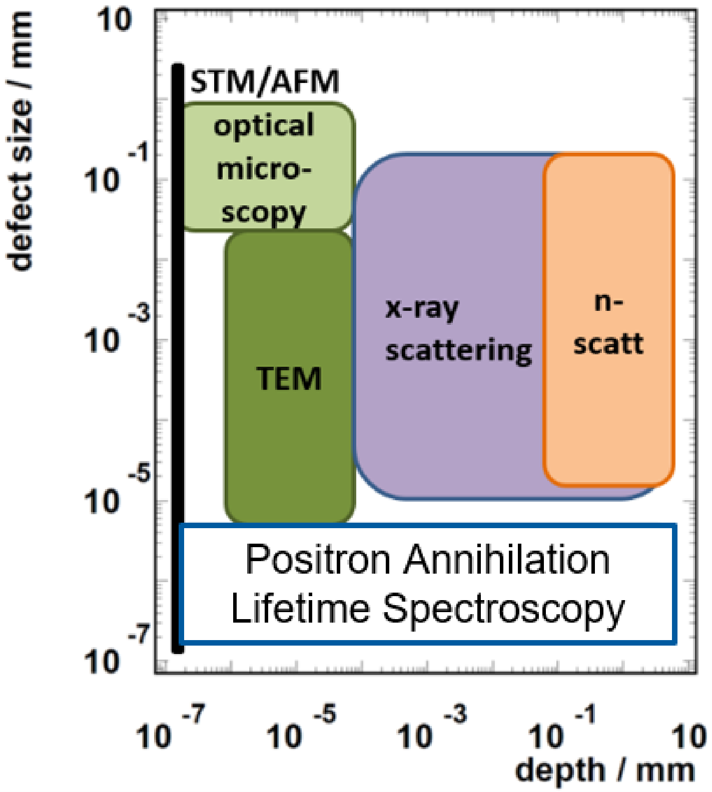


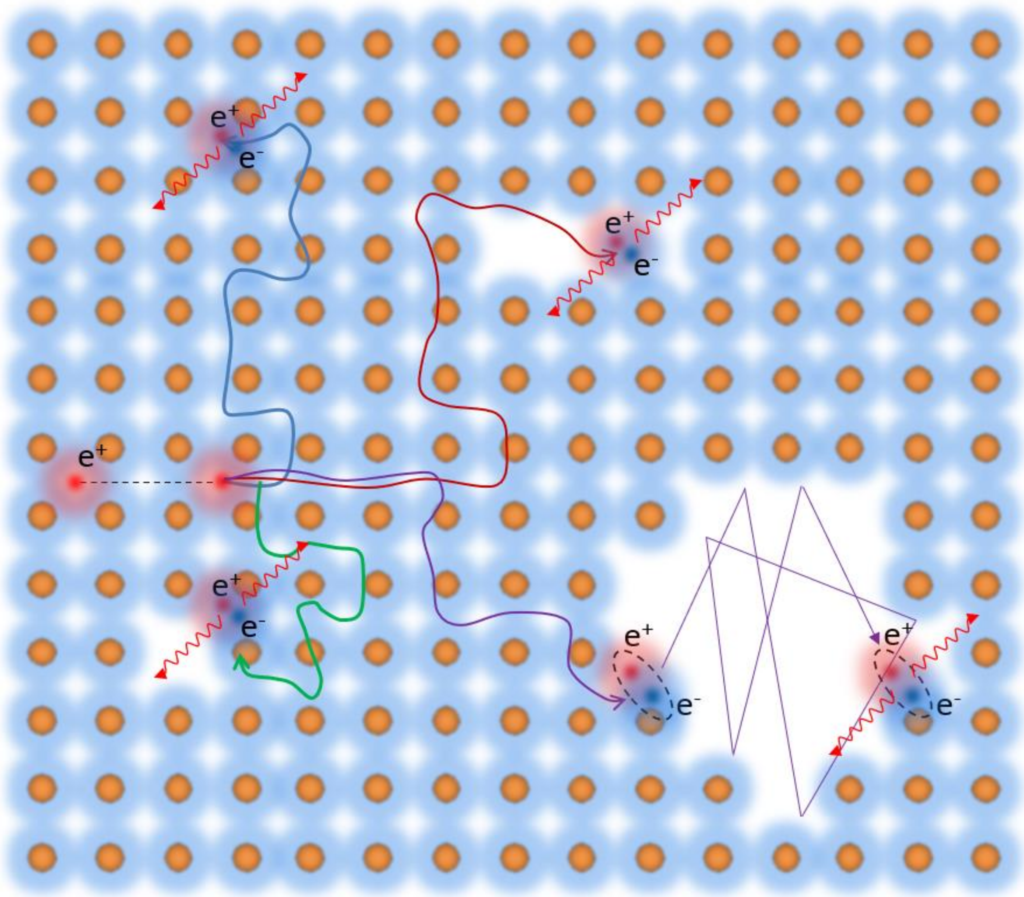
2 component fit for Nb sample (6 keV implantation energy)

Positron Annihilation Spectroscopy (PAS)

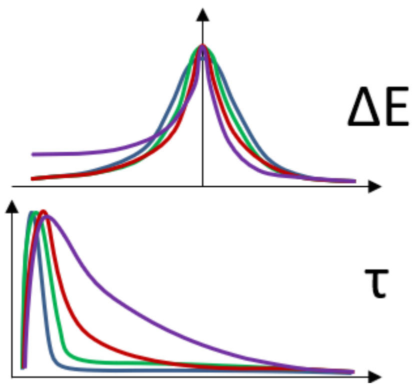
Positron Annihilation Lifetime Spectroscopy (PALS)

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





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



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Recent research of point defects

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

Vacancy kinetics in Nb

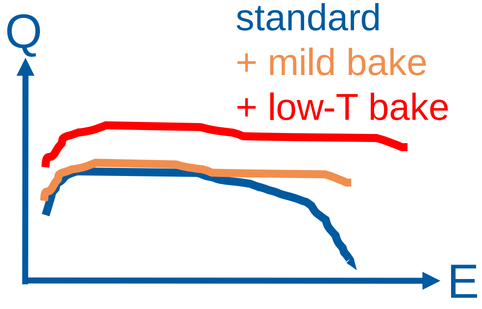
Collaboration with M. Wenskat (DESY, DE) & J. Čižek (Prague, CZ)

Origin of Q disease

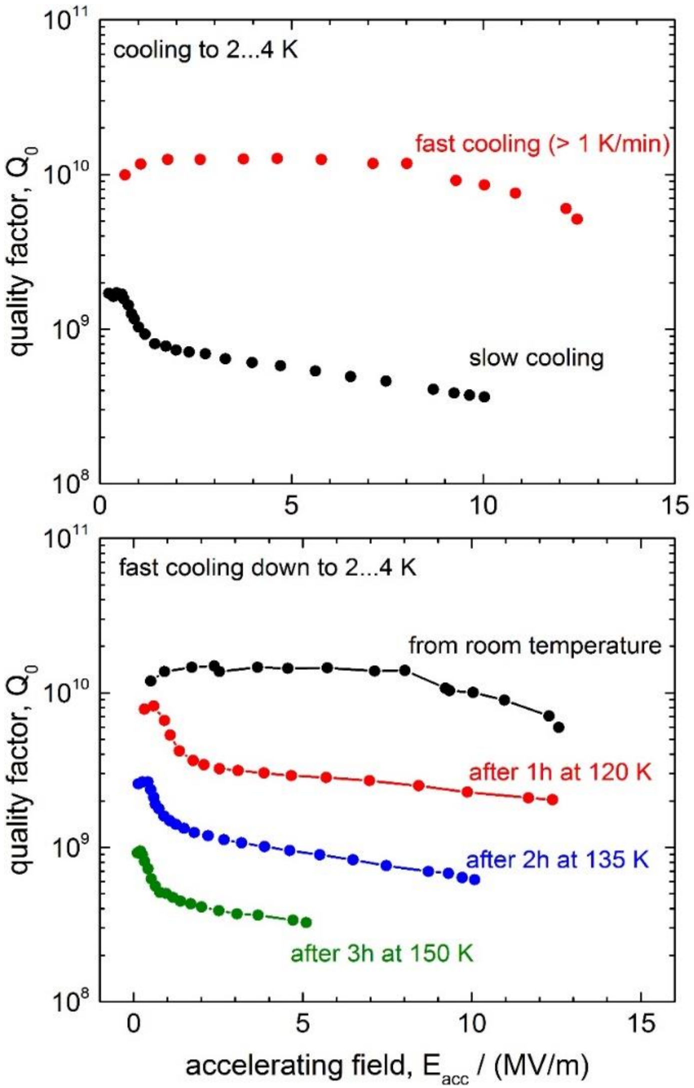
- formation of **niobium hydrides** in the rf surface layer during cooling down
- **open volume lattice defects** have high trapping potential for impurities, especially hydrogen: **vacancy-hydrogen complexes**

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%

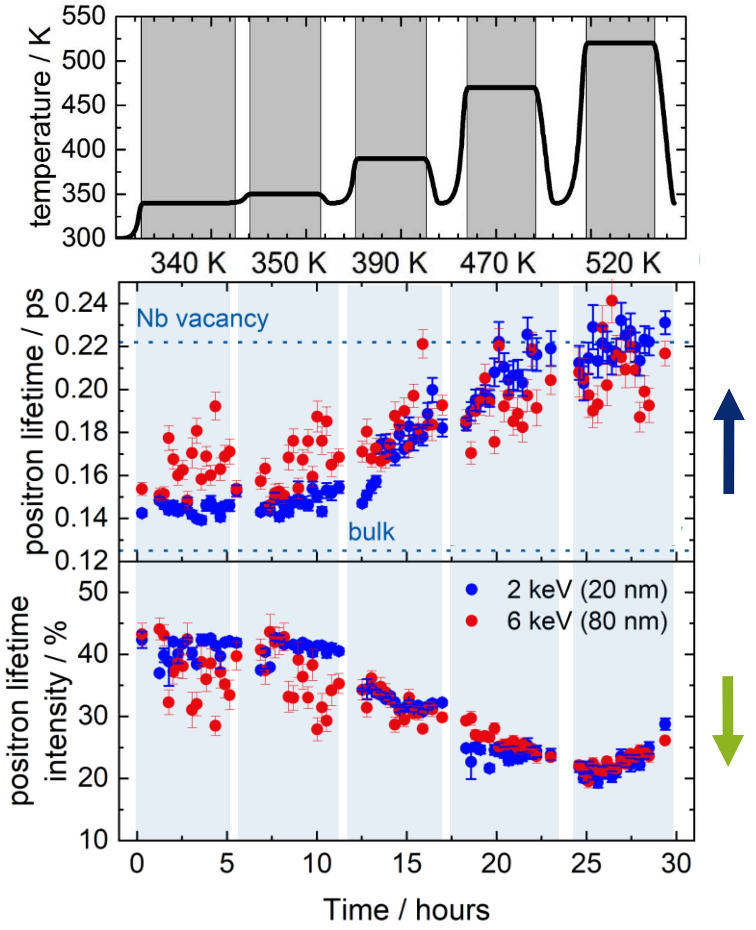


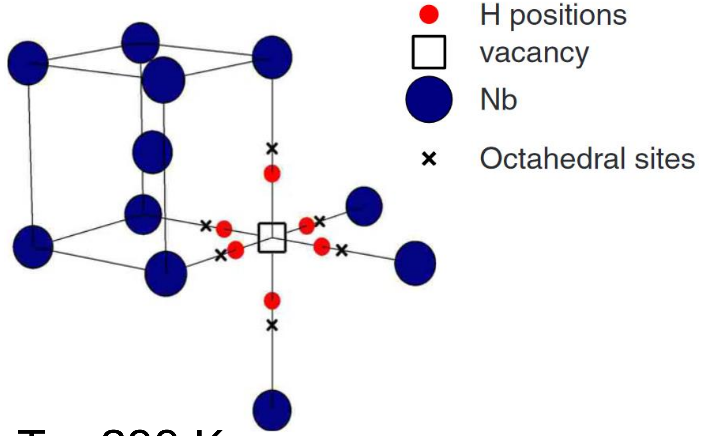
Q disease

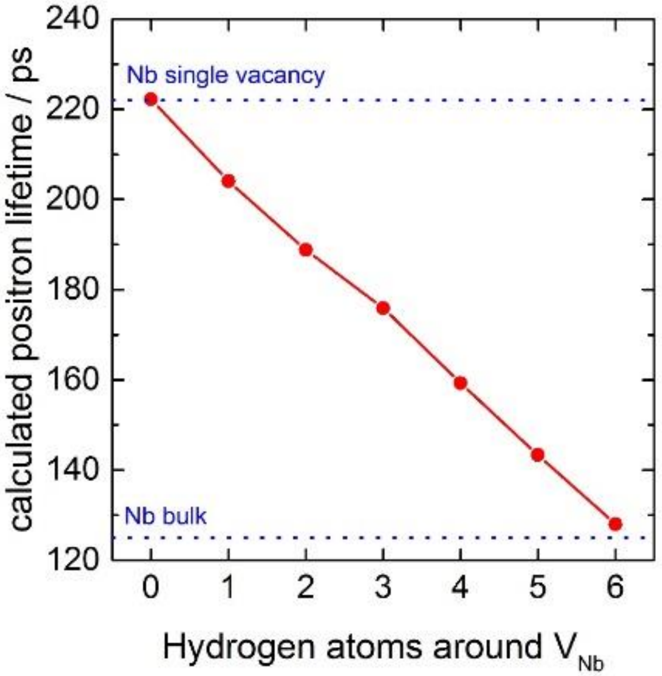


Vacancy kinetics in Nb

Collaboration with M. Wenskat (DESY, DE) & J. Čižek (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

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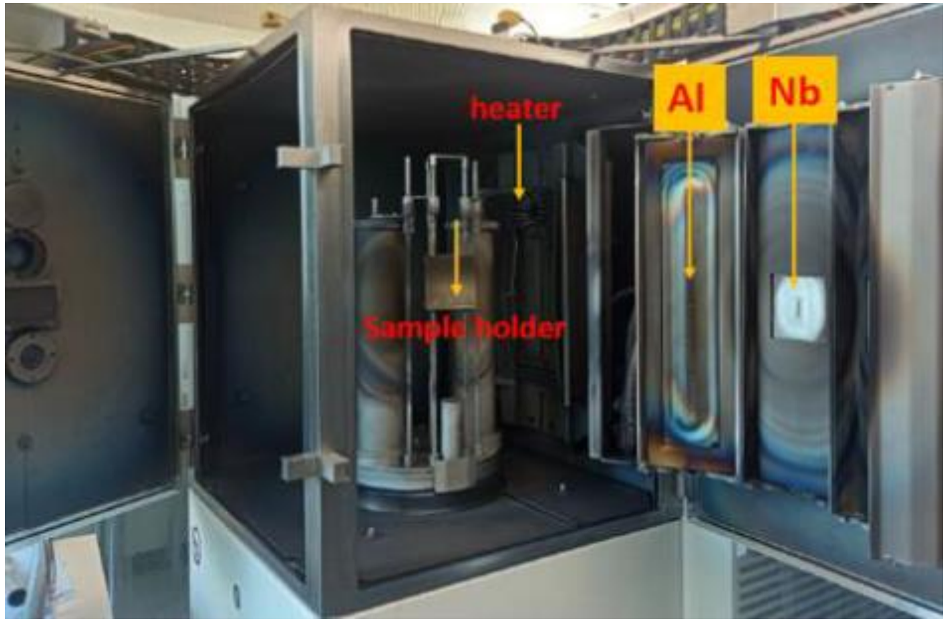
Sample characterization @HZDR, Germany

→ XRD (θ - 2θ scan)

→ DBS & PALS

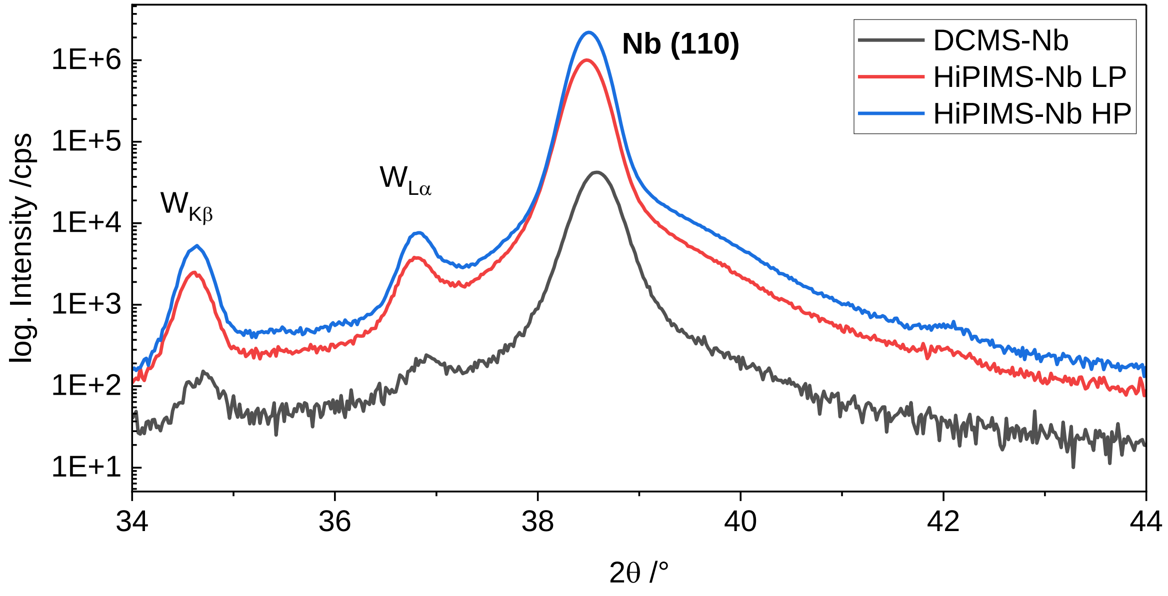


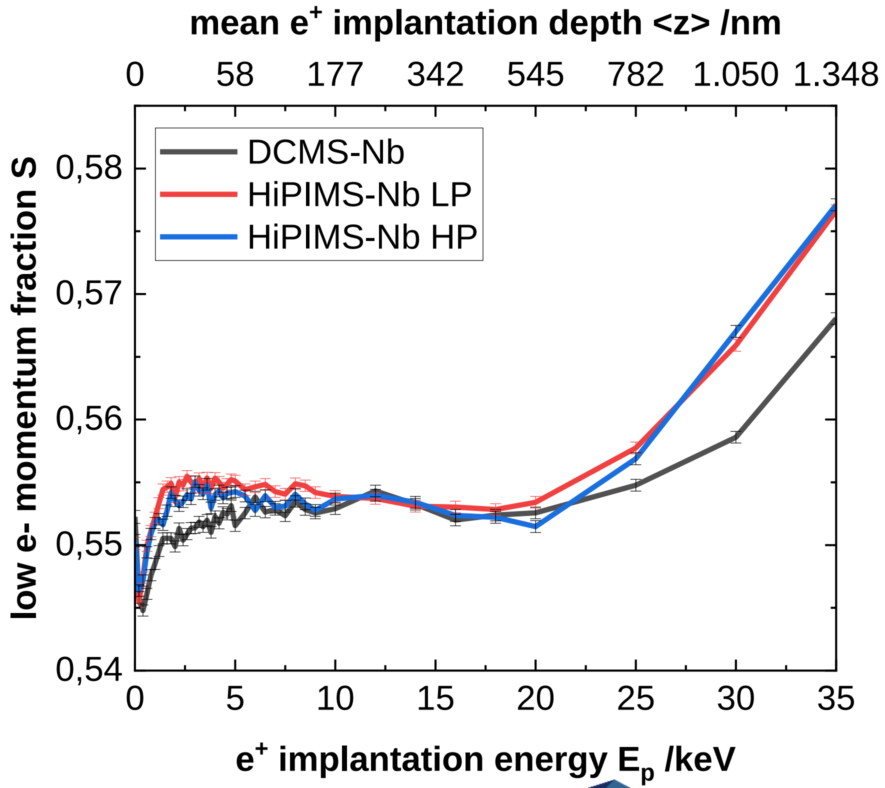
CemeCon CC800 at University of Siegen

Point defects of Nb-based thin films

Nb films on Si(100)

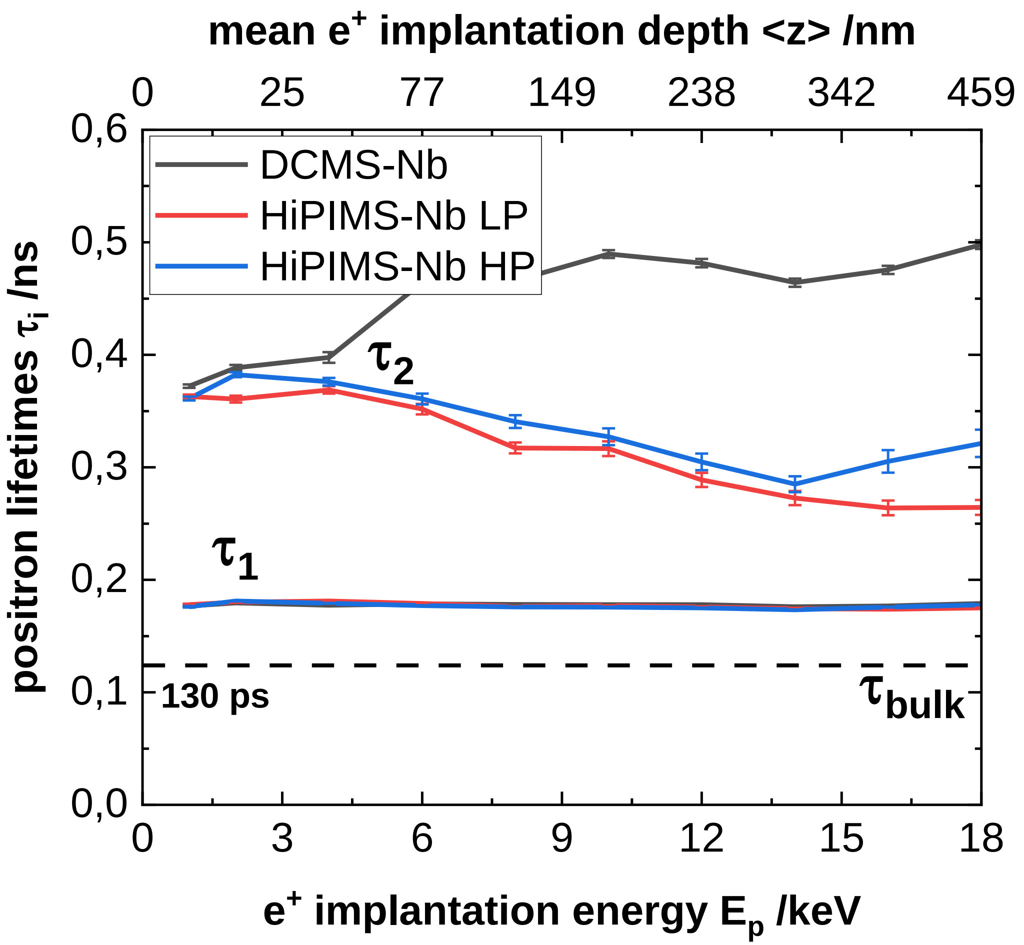
sample	cathode type	Power (W/cm^2)	p_gas (mbar)	Ar flow (sccm)	N2 flow (sccm)
DCMS-Nb	DC	4,54	***0,00801***	***410,0***	*0,0*
HiPIMS-Nb LP	HiPIMS	4,54	***0,00801***	***500,0***	*0,0*
HiPIMS-Nb HP	HiPIMS	6,82	***0,02001***	***700,0***	*0,0*

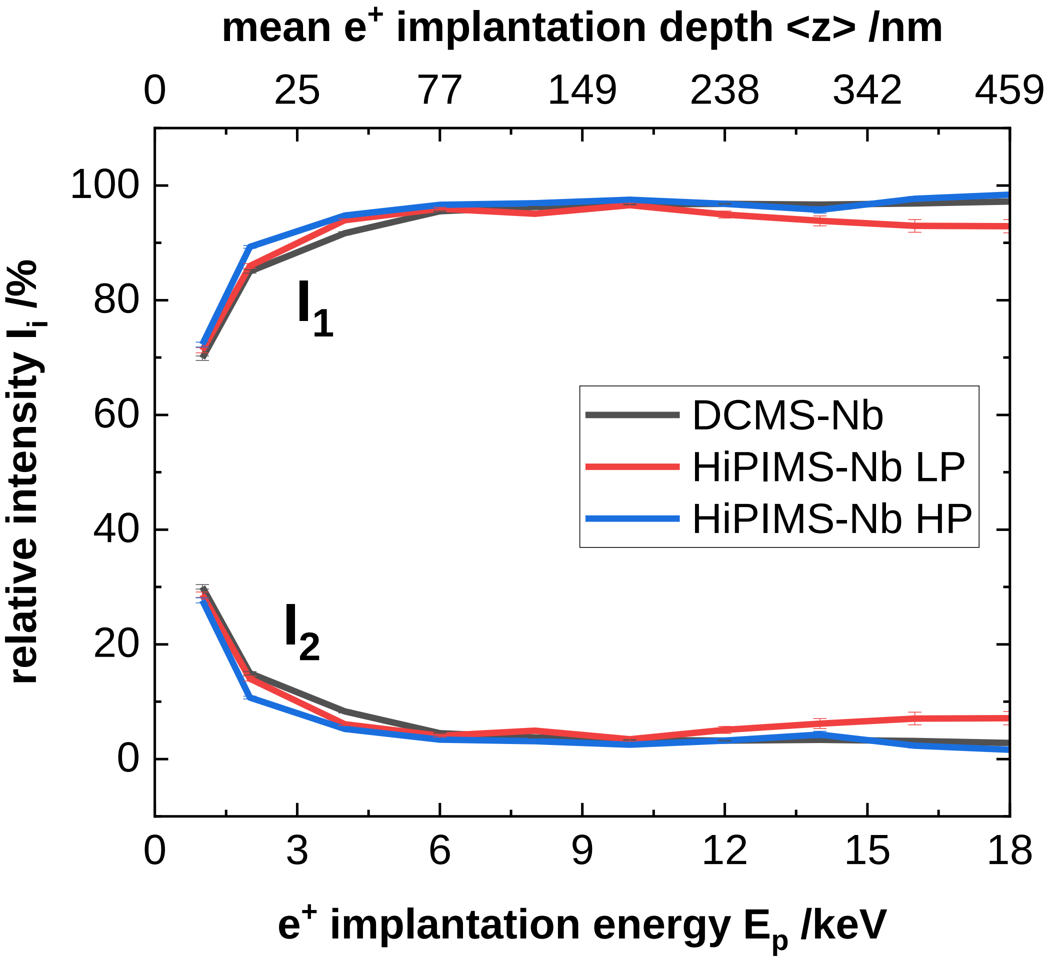




Point defects of Nb-based thin films

Nb films on Si(100)

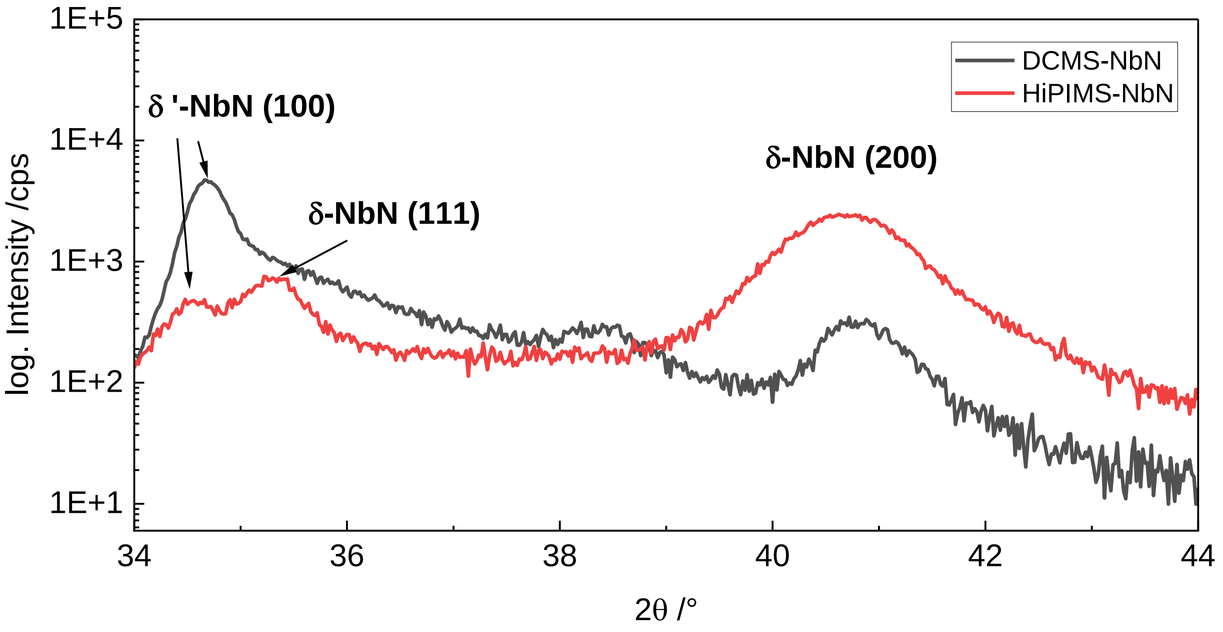


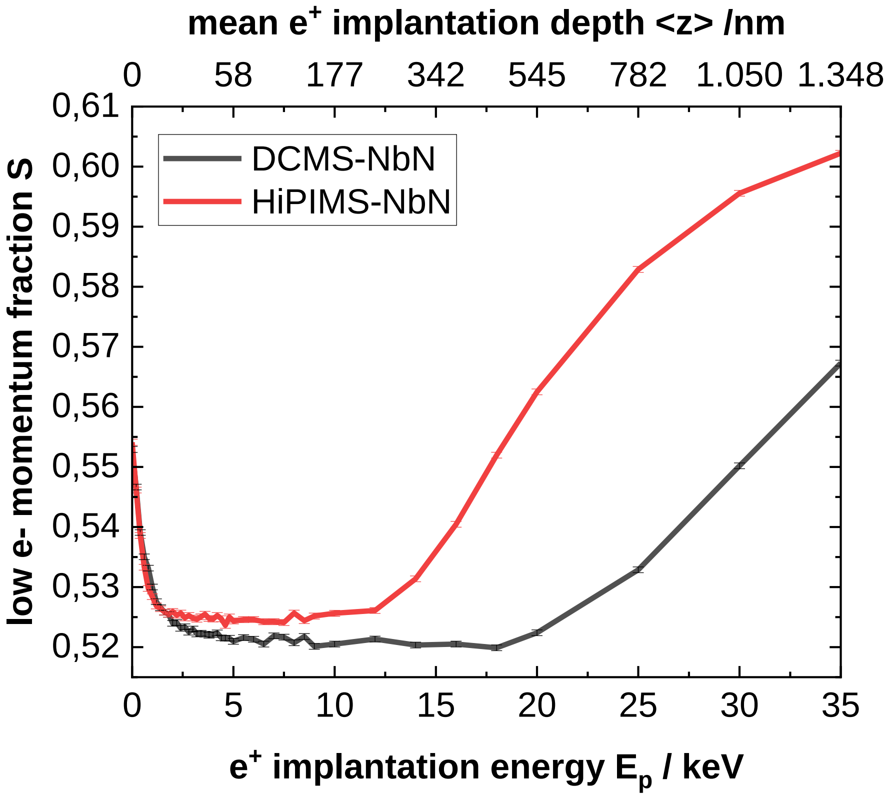


Point defects of Nb-based thin films

NbN films on Si(100)

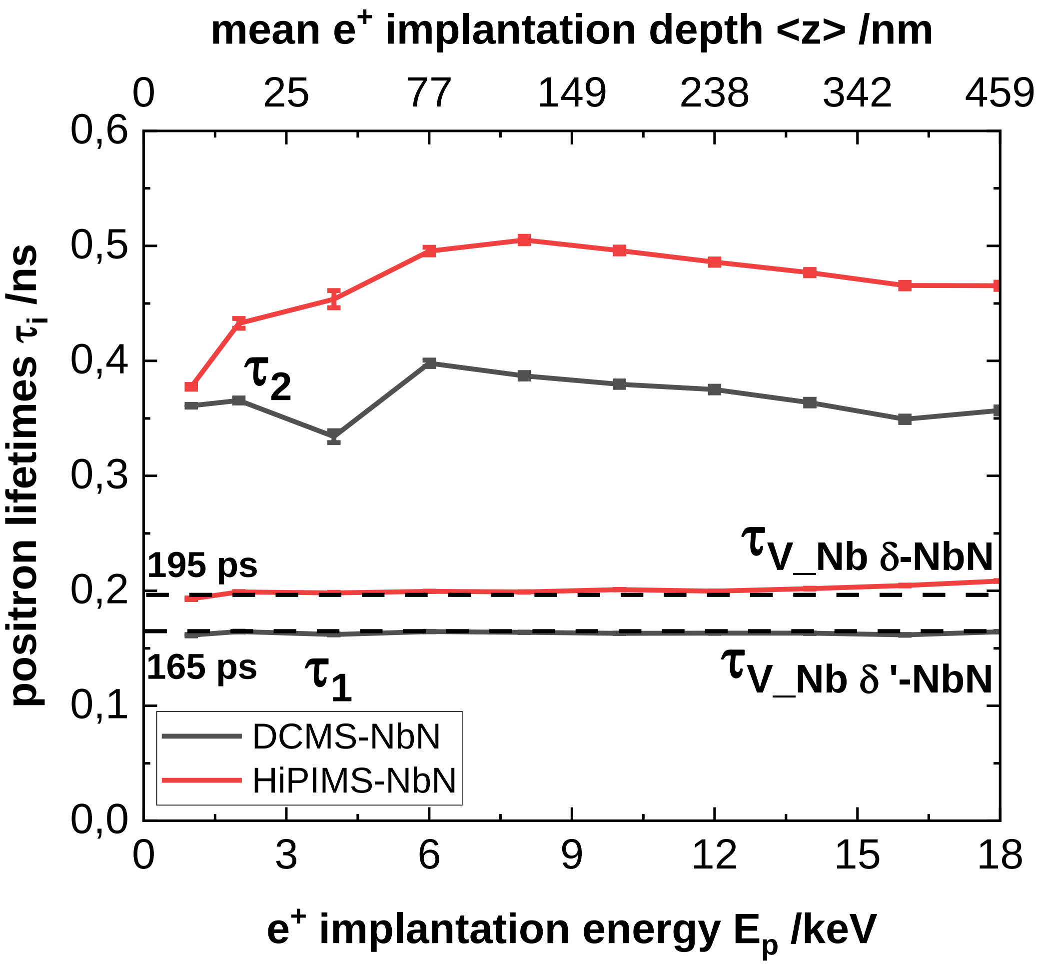
sample	cathode type	Power (W/cm^2)	p_gas (mbar)	Ar flow (sccm)	N2 flow (sccm)
DCMS-NbN	DC	5,68	***0,01315***	***522,5***	***45,5***
HiPIMS-NbN	HiPIMS	4,54	***0,02291***	***641,8***	***71,3***

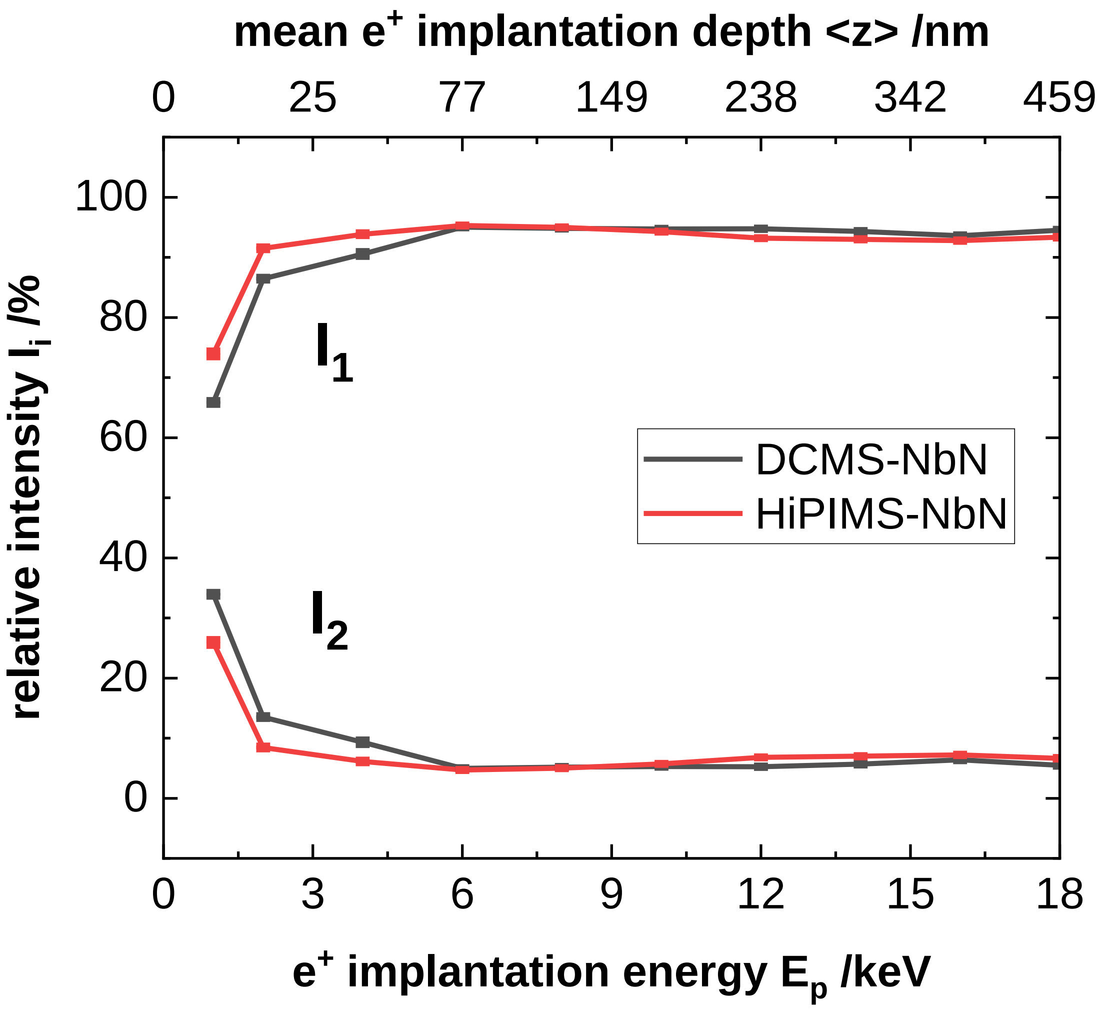




Point defects of Nb-based thin films

NbN films on Si(100)





Positron user facilities at the HZDR

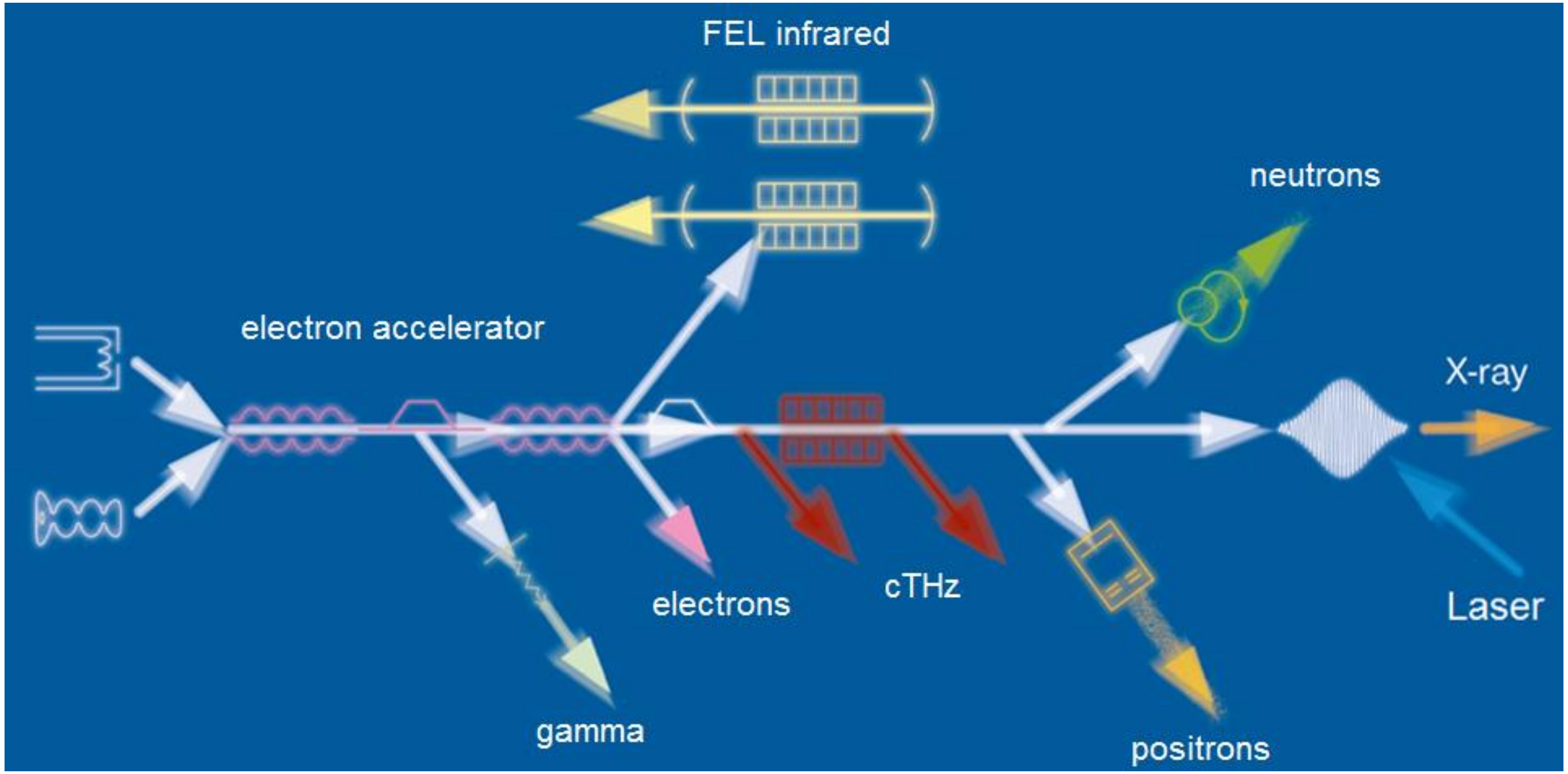
ELBE / SPONSOR / MePS / Access for user

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

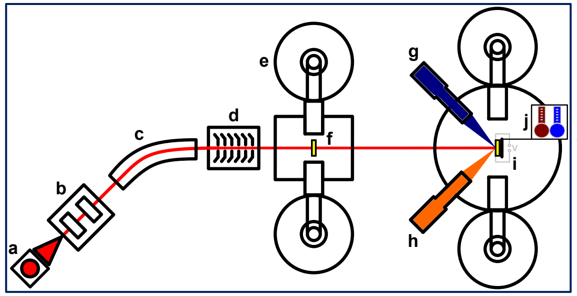
SPONSOR (The Slow-Positron System of Rossendorf)

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)

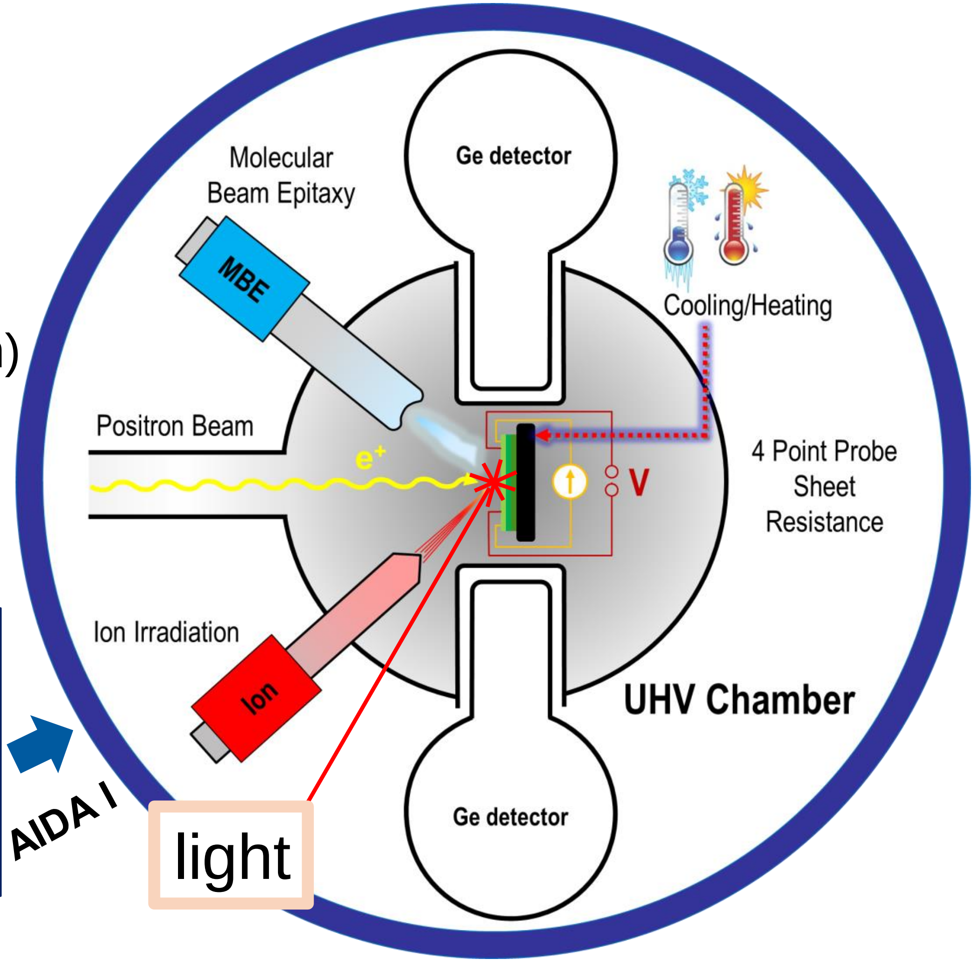
Apparatus for in-situ defect analysis (AIDA I)

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



SPONSOR

AIDA I

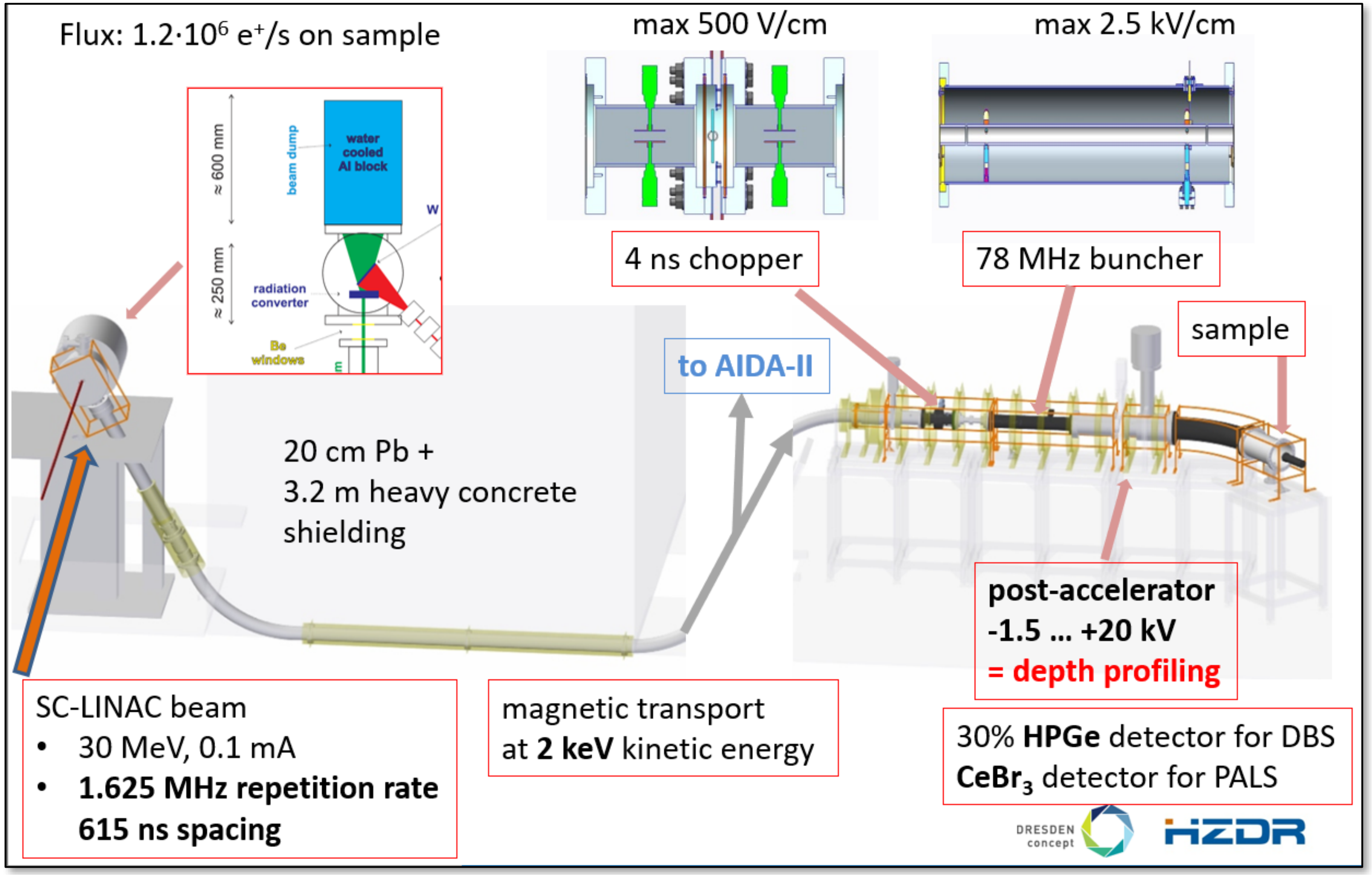


Positron user facilities at the HZDR

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

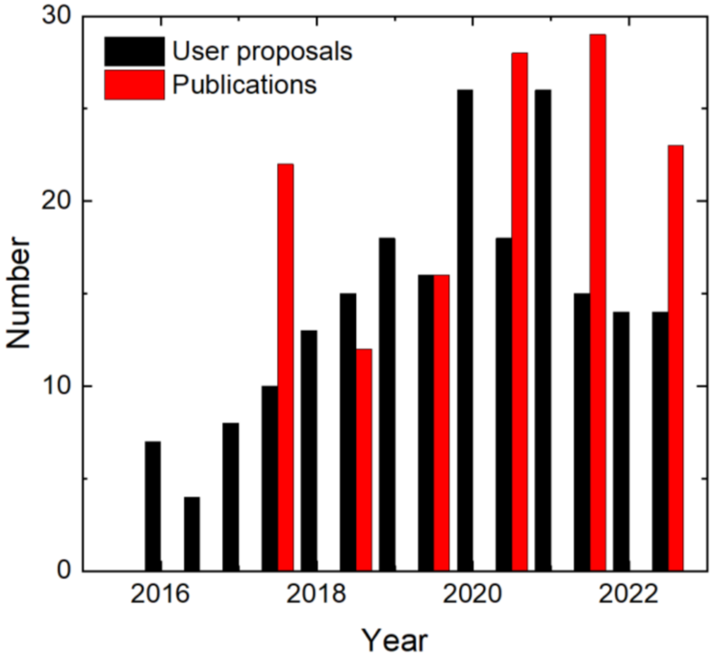
Application with scientific user proposal

- 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

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Outlook: Magnetron Sputtering & in-situ PALS

AIDA II / future plans

Outlook: Magnetron Sputtering & in-situ PALS

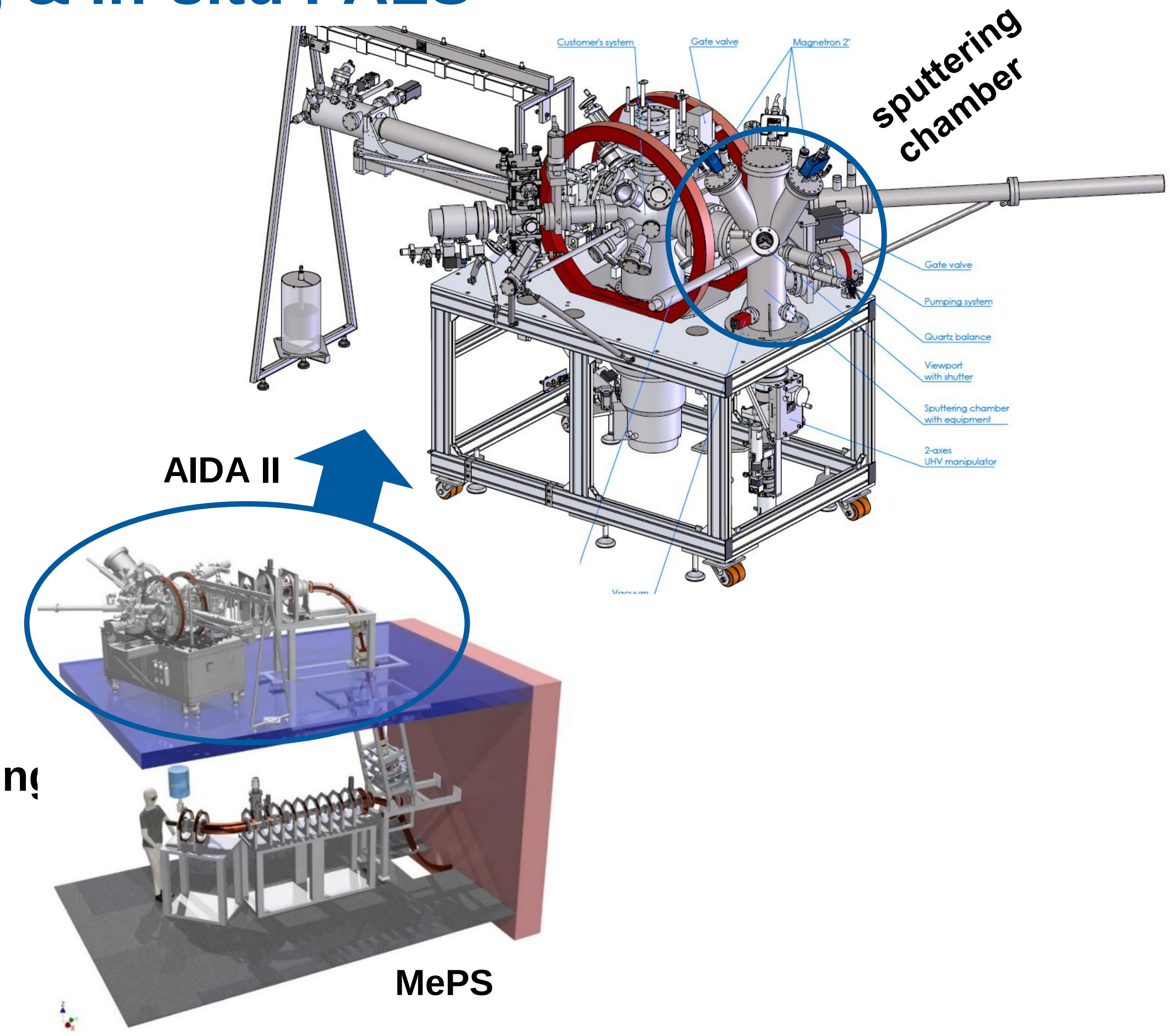
AIDA II

Magnetron sputter chamber

- 3 confocal magnetrons: **2x DC, 1x RF; 400 W**
- working gases: **Ar, N_2, O_2**
- **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, Nb_3Sn** 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

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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…

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