ELBE* – Center for High-Power Radiation Sources

Defects and Hydrogen in Nb films

*Electron Linear accelerator with high Brilliance and low Emittance

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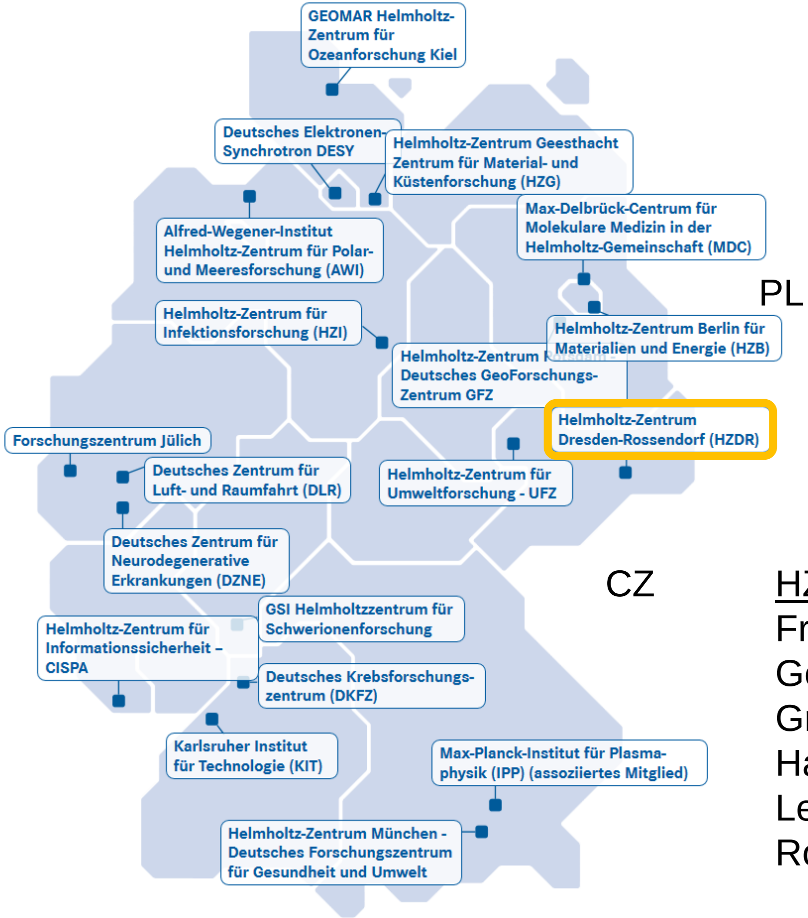
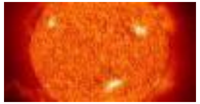
Helmholtz Center Dresden – Rossendorf





Energy

Key Technologies

Hermann von Helmholtz
1821 - 1894

Matter

Earth & Env.

Health

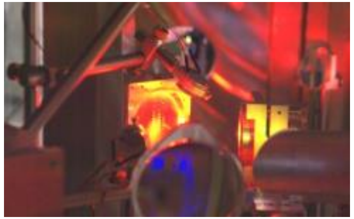
Space & Transport

HZDR outposts	
Freiberg	Resource technologies
Görlitz (CASUS)	Complex systems
Grenoble (ESRF)	ROBL beamline
Hamburg (European XFEL)	HiBEF
Leipzig	Radioisotopes
Rostock	High energy density physics

HZDR - large scale research infrastructures

ELBE.

SC electron CW-LINAC



PW-class lasers

Laser particle acceleration

HiBEF

IBC.

Ion beam center

HZDR INNOVATION

Tech transfer and industry services

HLD.

> 90 T magnets

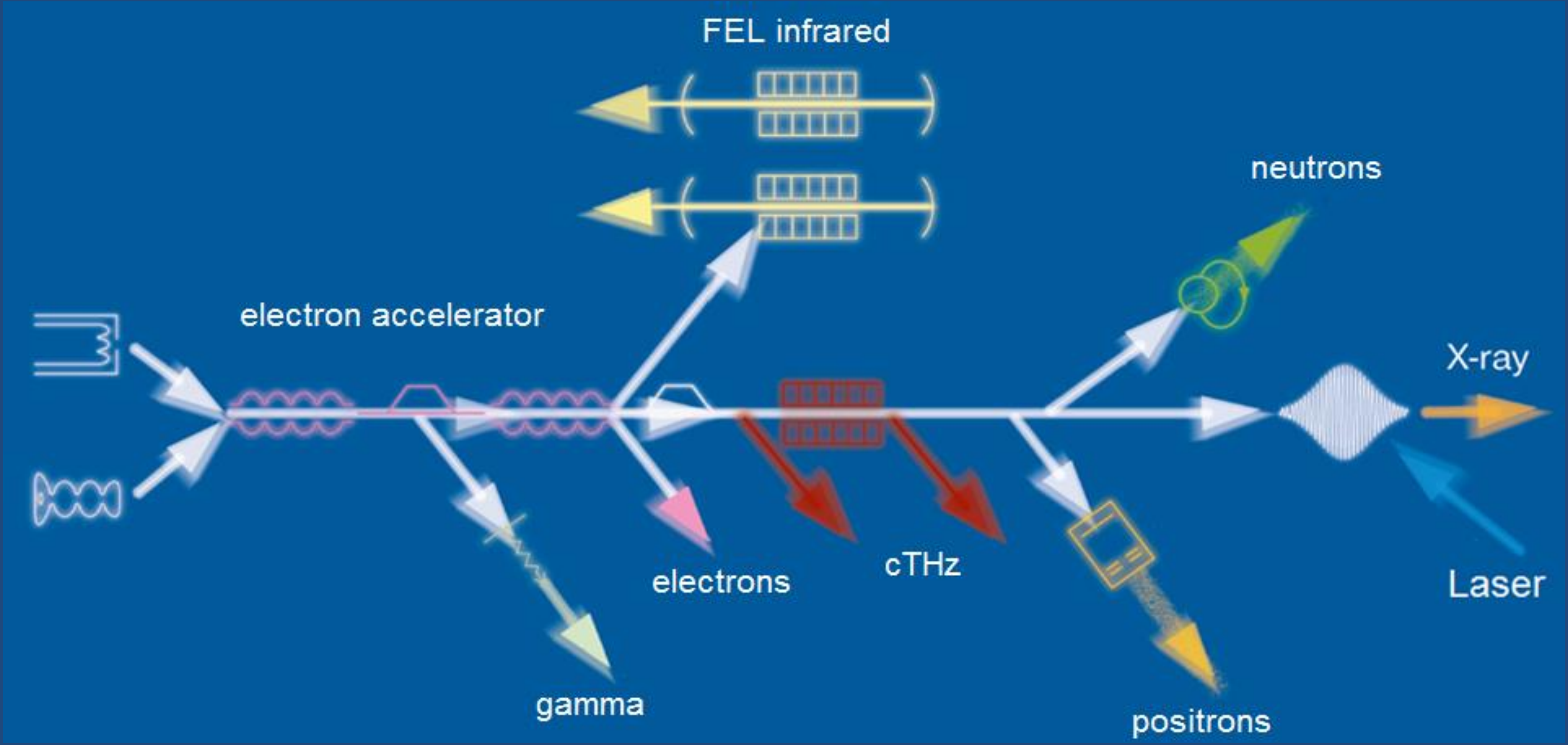
Proton tumor therapy and PET Center

OncoRay

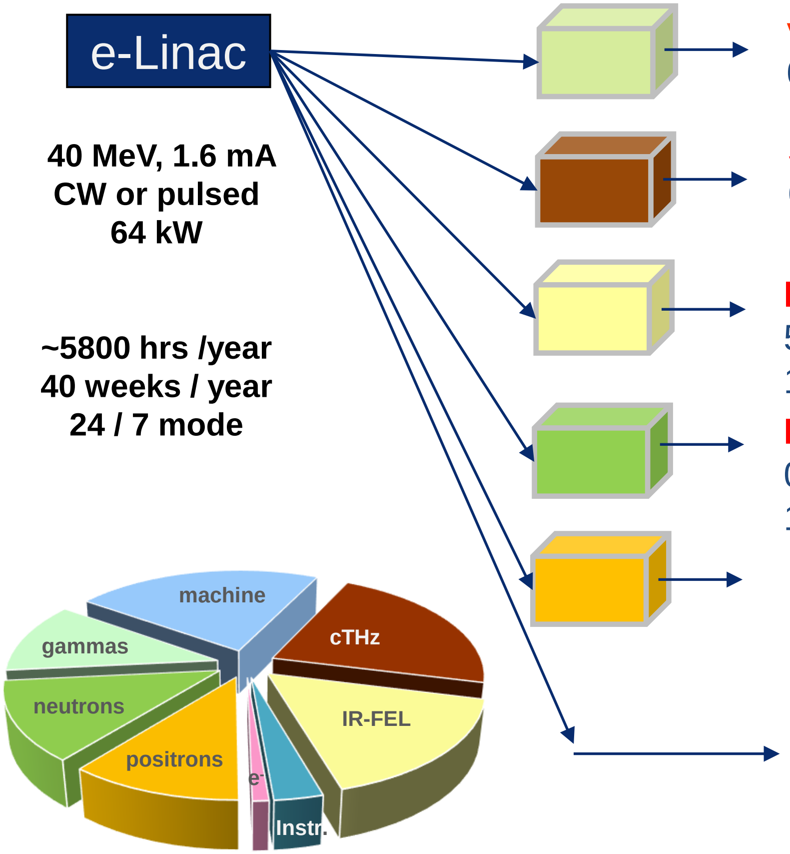
250 MeV protons

ELBE Center for High-Power radiation Sources

(Electron Linear accelerator w/ high Brilliance and low Emittance)



User facilities and science cases



e-Linac

40 MeV, 1.6 mA
CW or pulsed
64 kW

~5800 hrs /year
40 weeks / year
24 / 7 mode

γ - rays
0 – 20 MeV

- Nuclear physics – e-m strength in heavy nuclei
- Nuclear astrophysics – synthesis of elements
- In-sample positron generation

Superradiant THz
0.1 – 2.5 THz, 300 kV/cm
120 µm – 3 mm, 10-250 kHz

- Ultra-fast dynamics at fs – time scales
- Non-linear dynamics, spin-wave coupling
- Materials for high-bandwidth data transmission

IR-FELs
5 – 250 µm, 2 µJ
1.2 – 60 THz, 13 MHz

- Near-field microscopy
- Semiconductor spectroscopy, quantum dots
- Small bandwidth

Neutrons
0.1 – 10 MeV
10^4 n/(s cm^2), 100 kHz

- Materials for fusion reactors
- Nuclear transmutation
- Neutron time-of-flight

Positrons
0.5 – 20 keV,
$5 \cdot 10^6$ e^+/s

- Solid state physics – spectroscopy of defects
- Thin film semiconductors
- Porosimetry

Direct electrons
10 – 40 MeV

- Rad. biology – high local dose rates
- Technological developments
- Isotope production

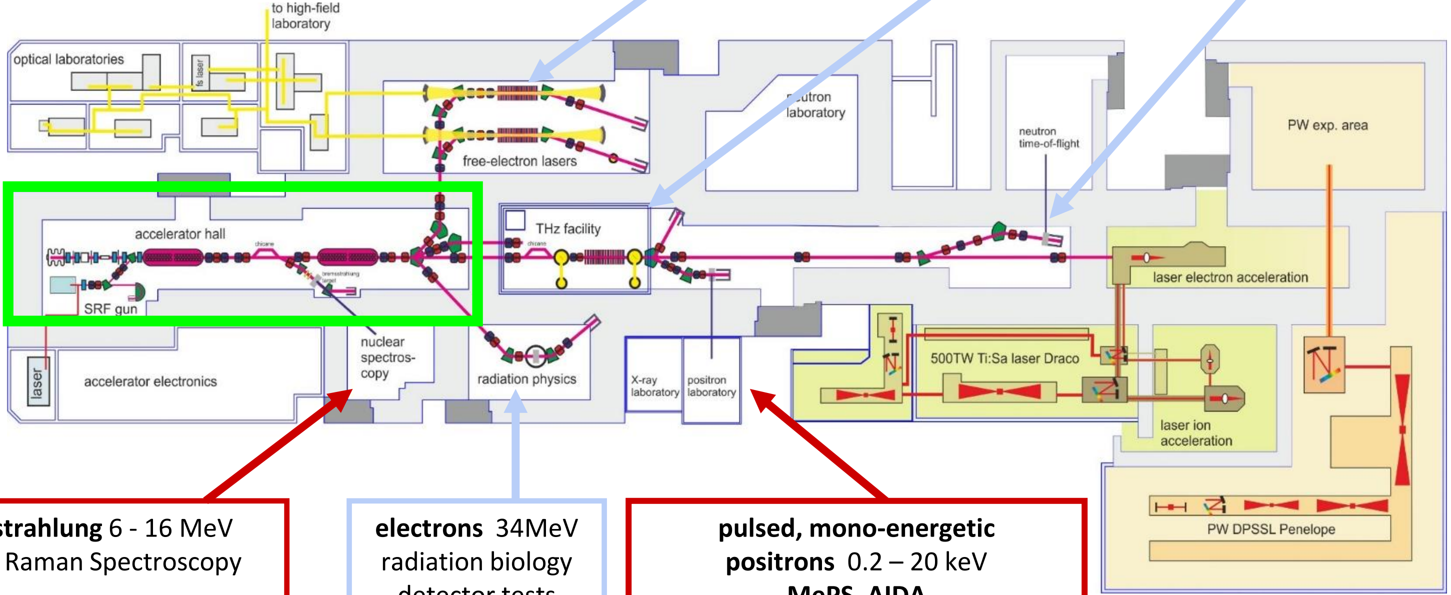
ELBE Center for High-Power radiation Sources

1.6 mA, 40 MeV (64 kW)
CW electron accelerator

coherent IR-radiation
3 – 230 µm

coh. THz radiation
100 µm – 3 mm

neutron time of flight
E_n 0 – 10 MeV



Bremsstrahlung 6 - 16 MeV
Nuclear Raman Spectroscopy

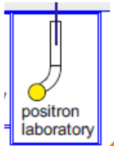
Gamma-induced Positron Source
GiPS

electrons 34MeV
radiation biology
detector tests

pulsed, mono-energetic
positrons 0.2 – 20 keV
MePS, AIDA

PW-class laser systems
Laser plasma acceleration
High-density plasma physics

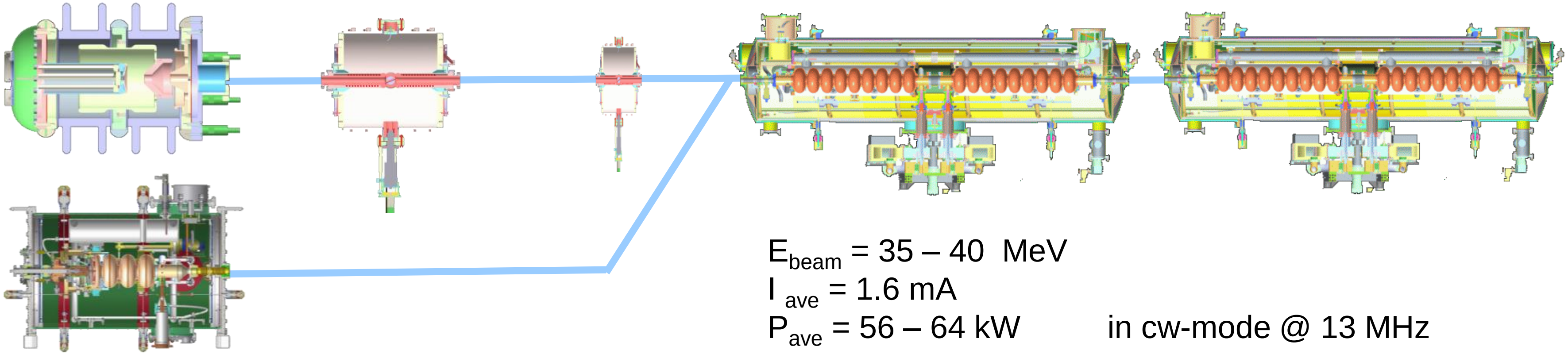
mono-energetic positrons
0.2 – 30 keV from ^{22}Na
SPONSOR



ELBE Center for High-Power radiation Sources

Superconducting Accelerator

SC resonant cavities developed by TESLA Technology Collaboration. Employed in CEBAF, FLASH, **ELBE**, EU-XFEL, LCLS-II, Lighthouse,…



Thermo-ionic
DC injector 250 keV
120 pC

250 MHz
buncher

1.3 GHz
buncher

Accelerator module
2 x 9-cell cavities

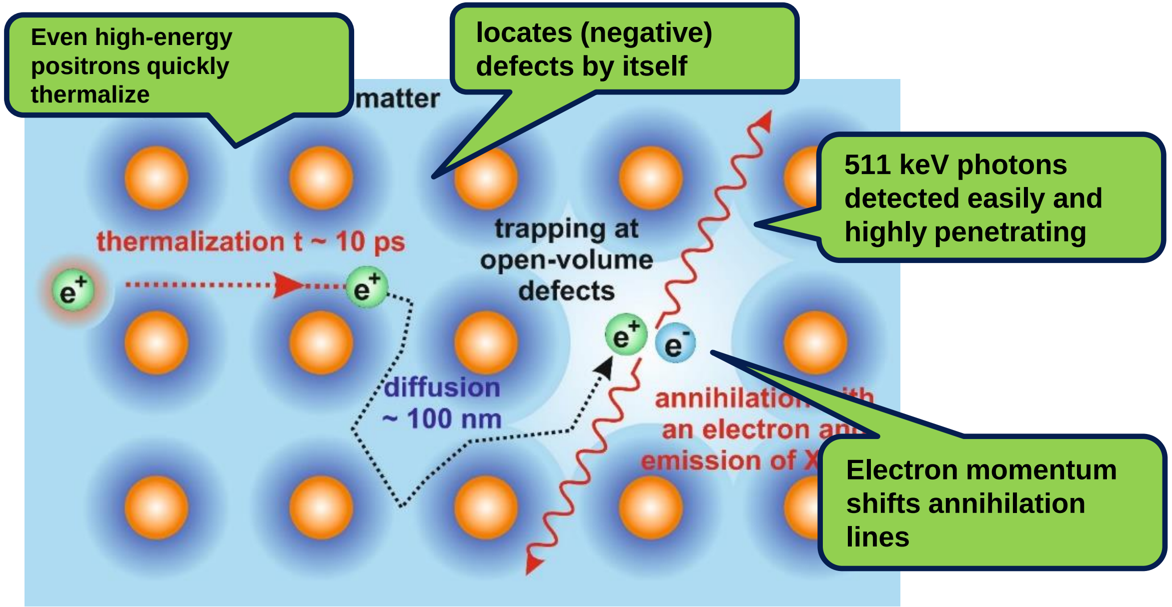
Accelerator module
2 x 9-cell cavities

E_{beam} = 35 – 40 MeV
I_{ave} = 1.6 mA
P_{ave} = 56 – 64 kW in cw-mode @ 13 MHz

SRF CW gun with
CsTe photocathodes
7.5 MV / m
250 pC (1 nC WIP)

SRF gun in routine operation for high-charge modes since 2016
Mainly serves cTHz and photo-neutron source at 10 – 250 kHz

Positron Annihilation: basics

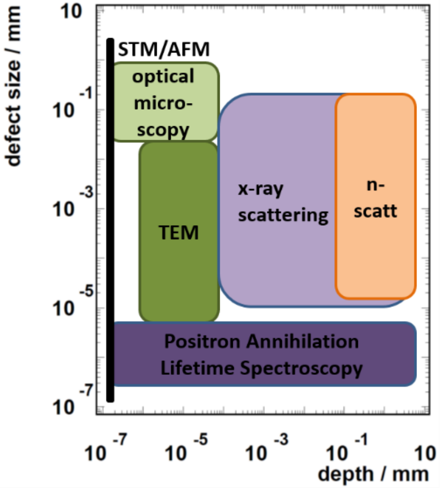


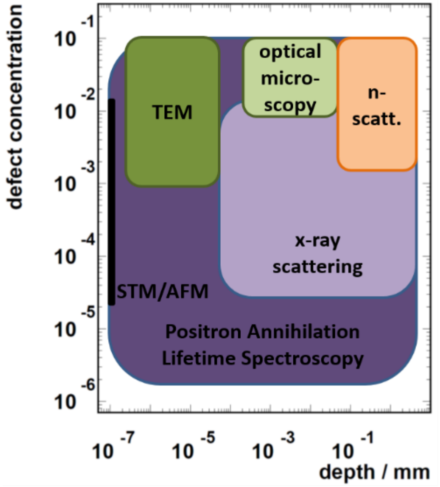
Even high-energy positrons quickly thermalize

locates (negative) defects by itself

511 keV photons detected easily and highly penetrating

Electron momentum shifts annihilation lines

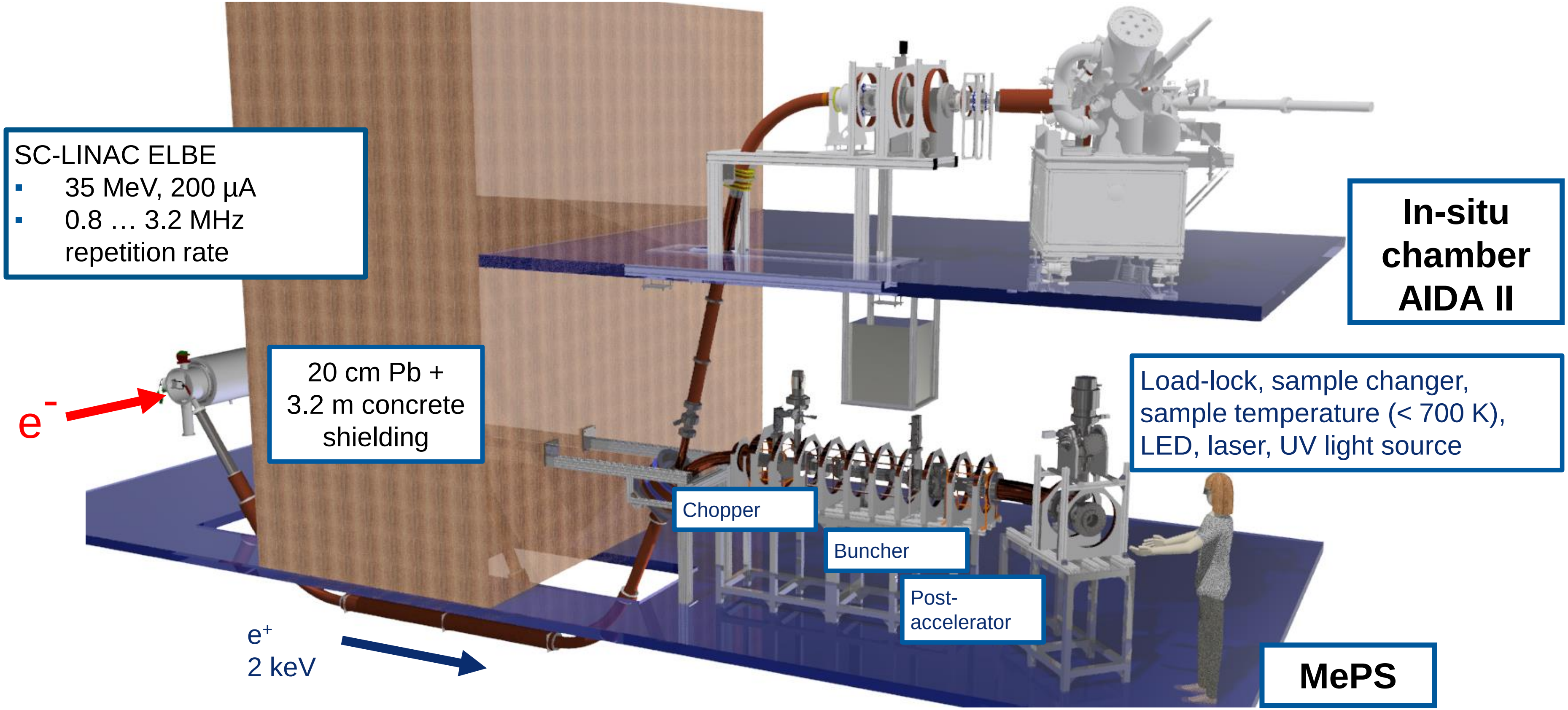




The e$^+$ probes…

- **local electron density** and electron momentum
- **defect sizes and types** by annihilation lifetime
- **defect concentrations** by component's intensity
- **large volumes** due to large diffusion lengths

MePS – The Mono-energetic Positron Source @ ELBE



SC-LINAC ELBE

- 35 MeV, 200 µA
- 0.8 … 3.2 MHz repetition rate

20 cm Pb +
3.2 m concrete
shielding

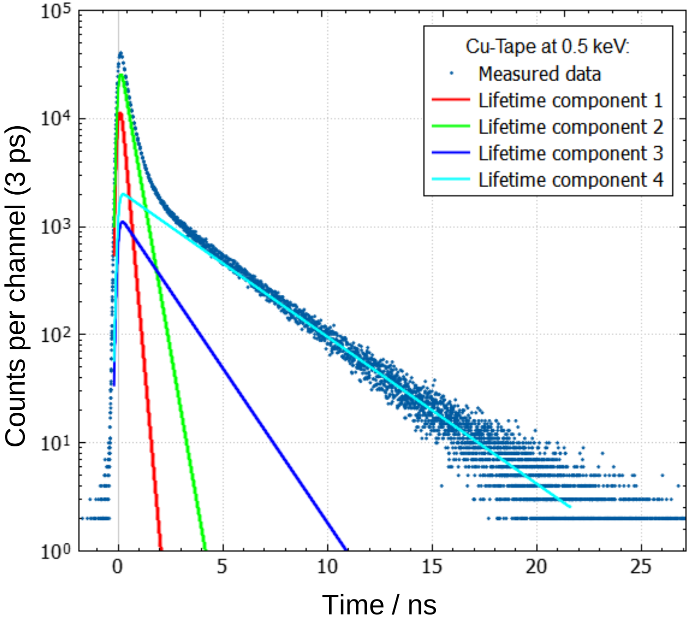
In-situ chamber AIDA II

Load-lock, sample changer, sample temperature (< 700 K), LED, laser, UV light source

MePS

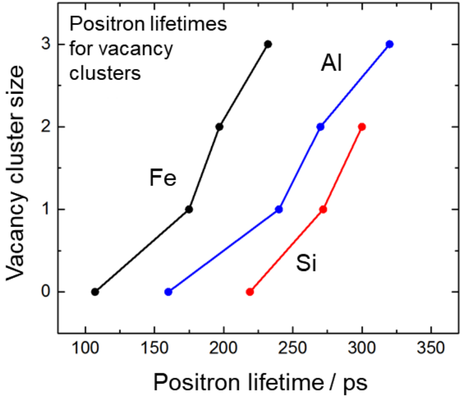
Positron Annihilation: basics

- Decomposing positron annihilation lifetime distribution
  - Lifetime correlates with size of cluster vacancies



$$f(t) = \sum_{j=1}^{k_0} (a_j * R)(t) \; + \; B$$

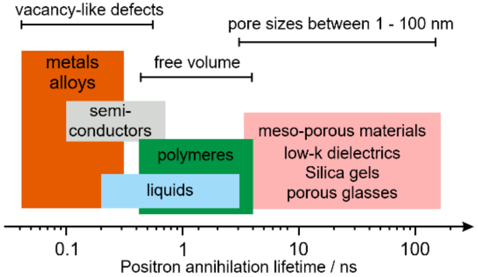
$$a_j(t) = \begin{cases} A_j \exp(-t/\tau_j), & t > 0 \\ 0, & t < 0 \end{cases}$$

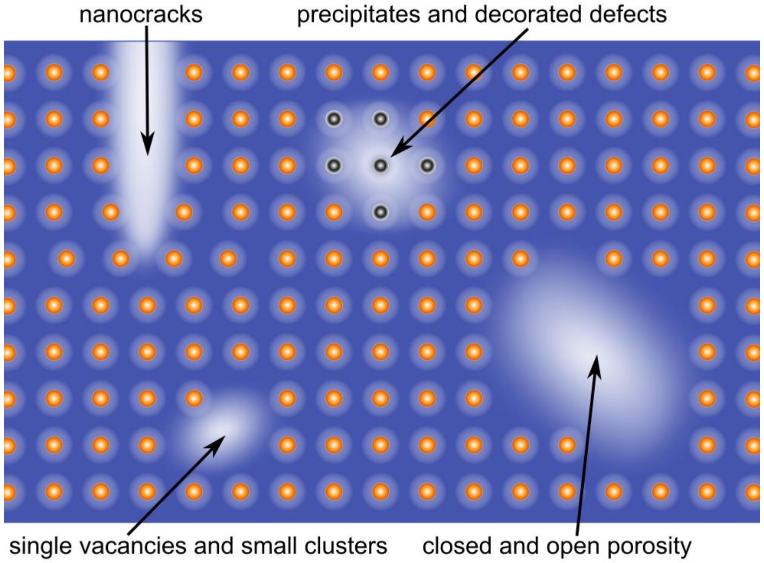


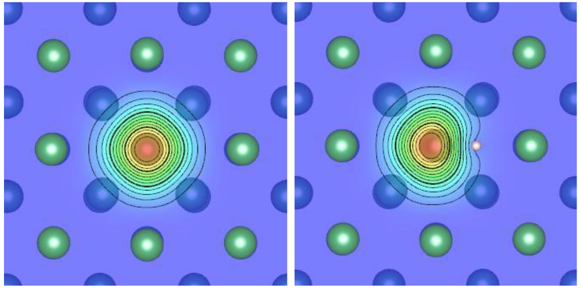
- well-suited for defect characterizations in Nb

Positron Annihilation: basics

Open-volume defects, vacancy clusters, precipitates and defect decorations

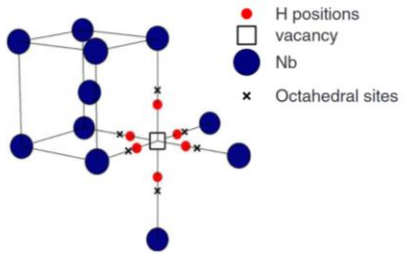


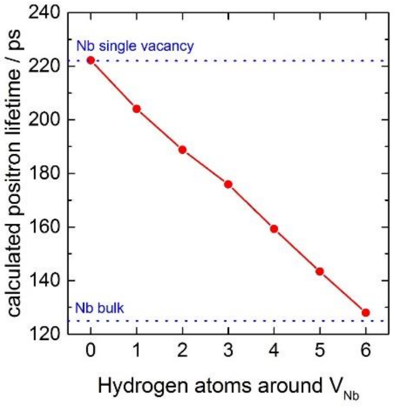




Positron wave functions in a Nb vacancy

+ relaxed ion configurations for v-H complex in the (001) plane
(DFT calculations by
J. Čižek, Charles-U Prague)



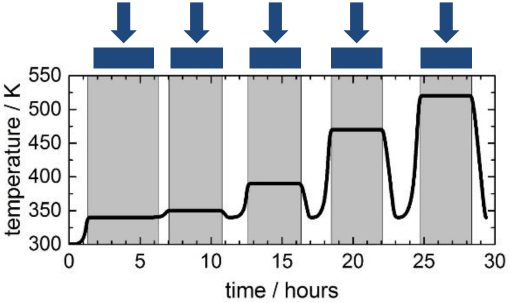


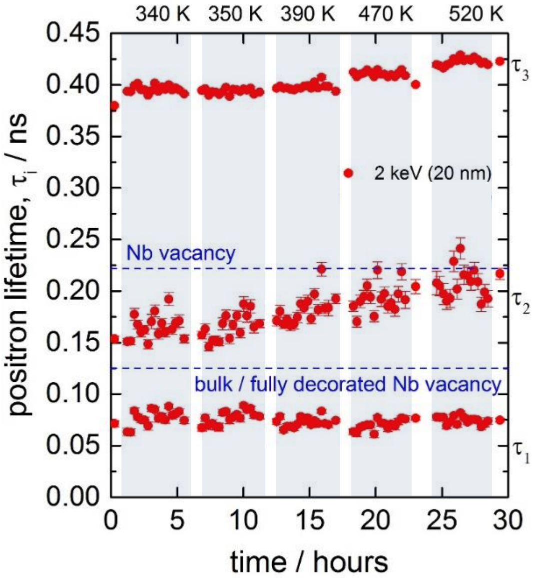
- well-suited for the study of decorated defects → vacancy-hydrogen complexes *v+nH in Nb*

J. Čižek et al., Phys Rev B79, 054108 (2009)
J. Čižek et al., Phys. Rev. B69, 224106 (2004)

Vacancy kinetics during in-situ low-T baking

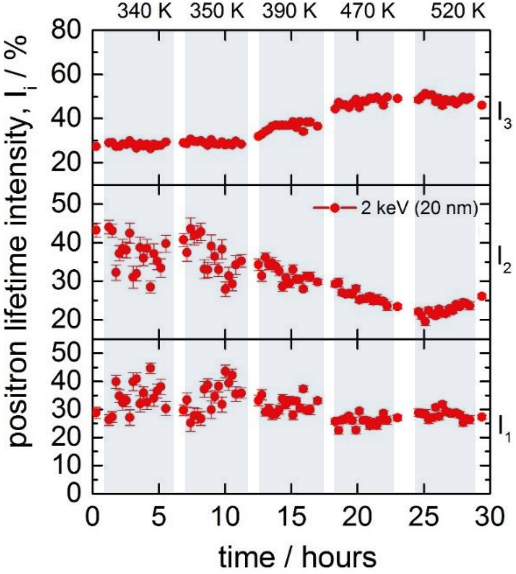
- ***v+nH* complex dynamics in surface near region**





Cluster formation

H-release

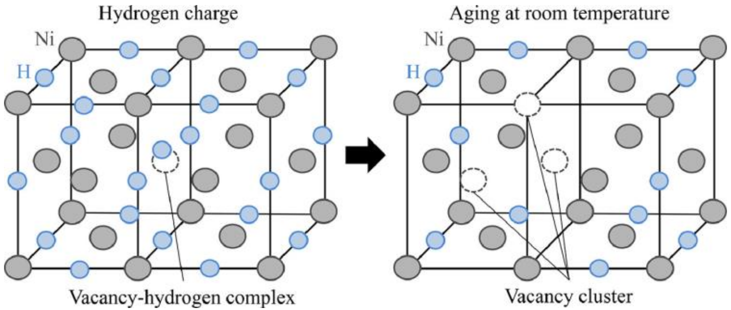


- T ≥ 390 K: shows most significant changes (already after short times)
  - ***v+nH* concentration (I_2) decreases**
  - **hydrogen release from the complexes (τ_2 increases)**
  - **defect type and surface states start to change (τ_3 increases)**

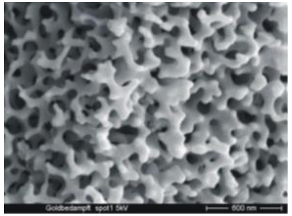
M. Wenskat, et al. "Vacancy-Hydrogen interaction in **niobium** during Low-temperature Baking." *Scientific reports 10.1 (2020) 8300*
M. Wenskat, et al. "Vacancy dynamics in **niobium** and its native oxides and their potential implications for quantum computing and superconducting accelerators"
Phys. Rev. B 106 (2022) 094516
L. Chiari, et al. "Formation and time dynamics of hydrogen-induced vacancies in **nickel**"
Acta Materialia 219 (2021) 117264

Positron Science Cases

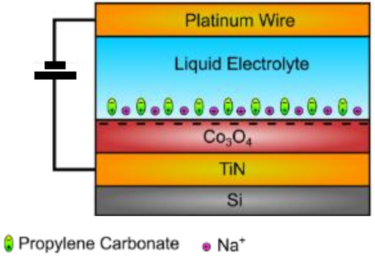
- Pulsed beams: **annihilation lifetime spectroscopy** for thin films, bulk materials, fluids, gases and (coincidence) **Doppler broadening spectroscopy**



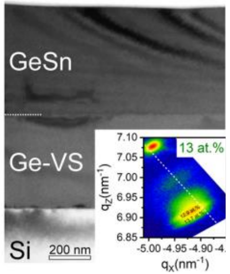
Hydrogen-induced vacancies in Ni



porous glasses, membranes, metal-organic frameworks

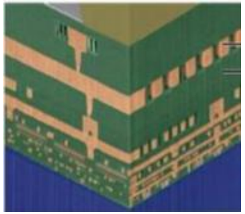


magneto-ionics

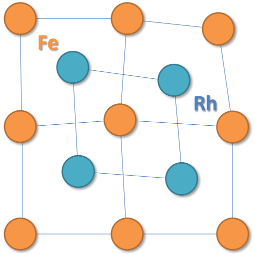


semiconductors for optoelectronics

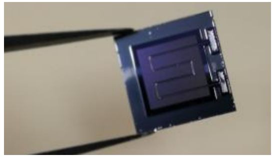
Niobium-hydrides & superconductivity



low-k dielectrics for CMOS devices



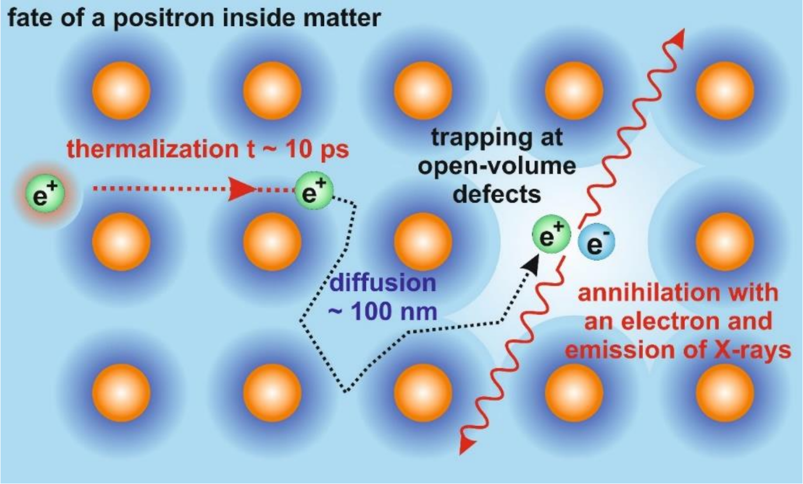
magnetic phase transitions through ion irradiation

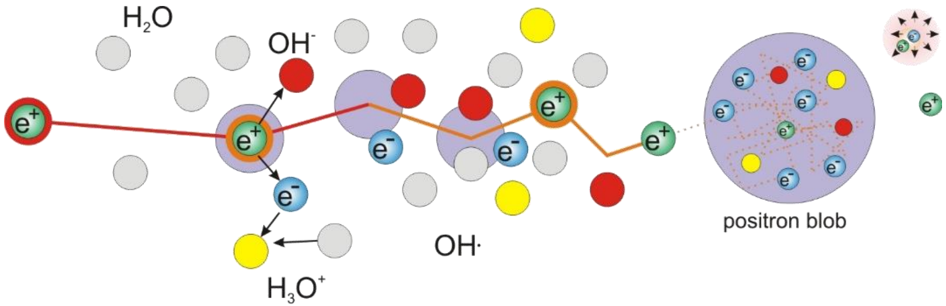


micro-solid oxide fuel cells

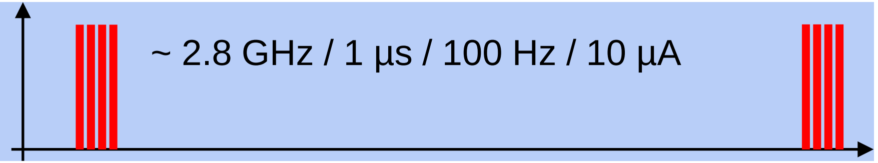
Positrons in matter – fundamentals to materials research

- porosimetry on the nm-scale
  - low-k SiO_2 for CMOS devices (DFG)
  - gas-separation membranes
- single$^+$-vacancy defect analysis
  - superconductors (H in Nb for SRF, YBCO)
  - FeCr, ODS, HEA for fission/fusion
  - semiconductors (ZnO, CIGS, Ge, Ga_2O_3,)
  - defect-induced magnetism (FeRh)
  - magneto-ionics (O_2 or N_2-mediated)
- chemical environment of defects
  - decoration, alloying, segregation
- positronium chemistry (medicine)

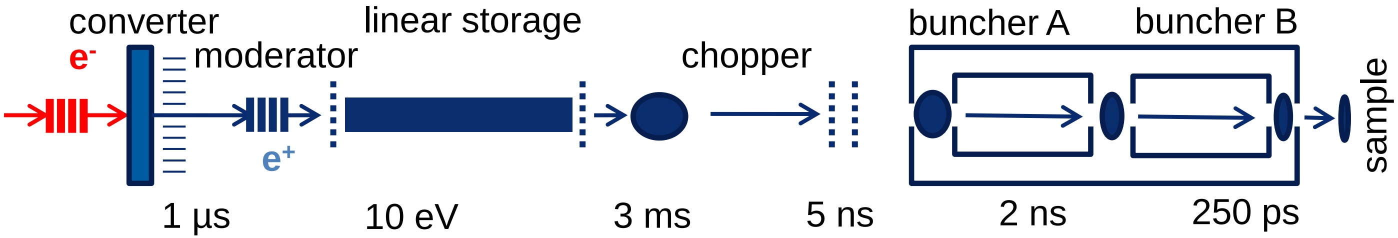


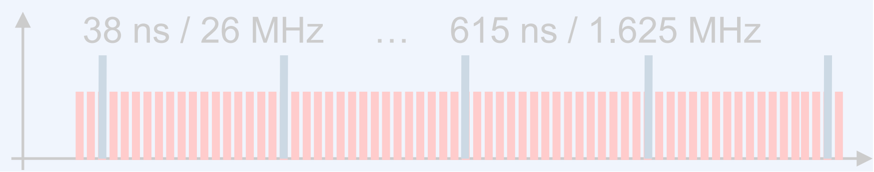


Accelerator-driven positron sources



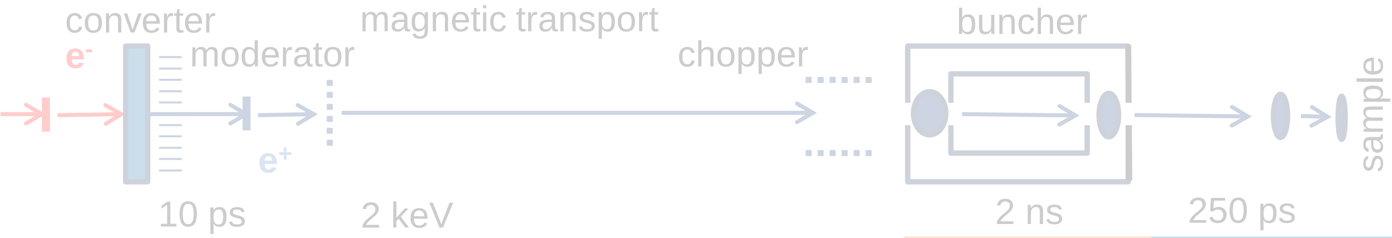
NC-LINAC in macro-bunched mode



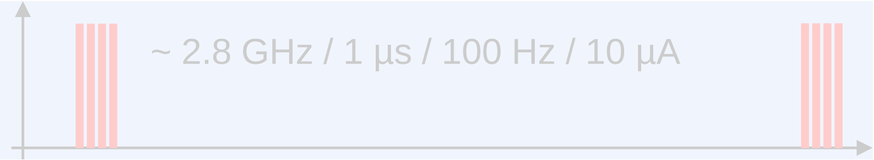


SC-LINAC in CW mode

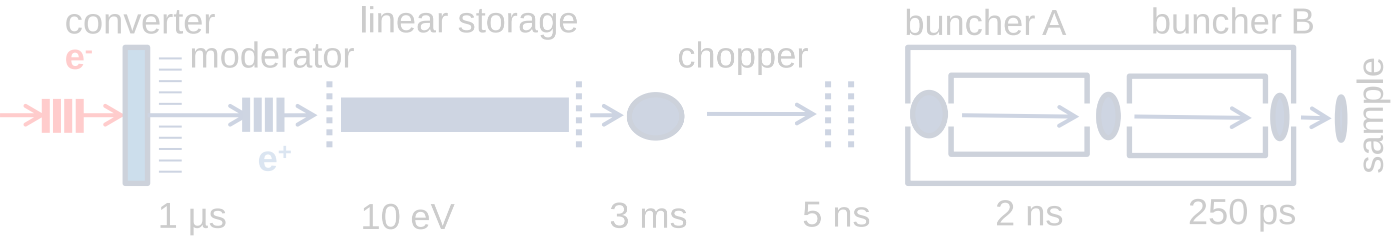
EPOS / MePS facility

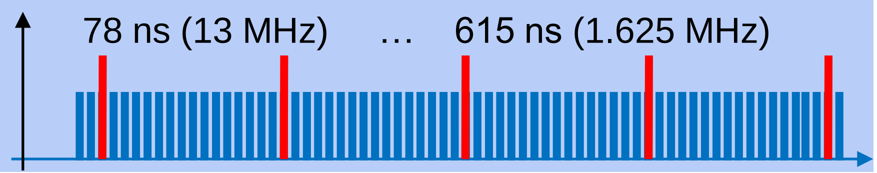


Accelerator-driven positron sources



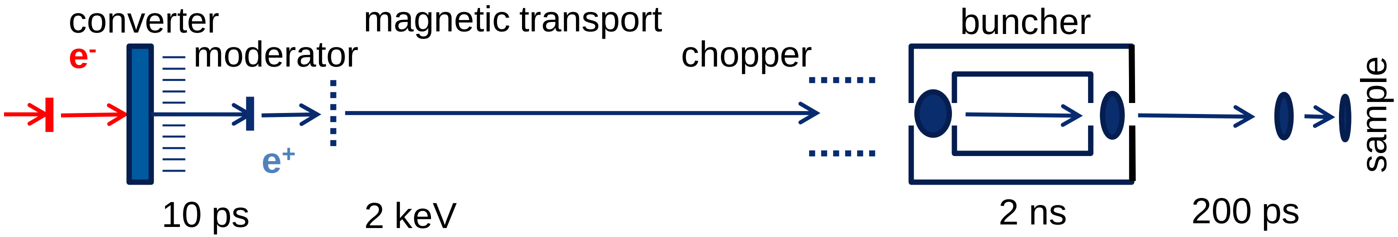
NC-LINAC in macro-bunched mode



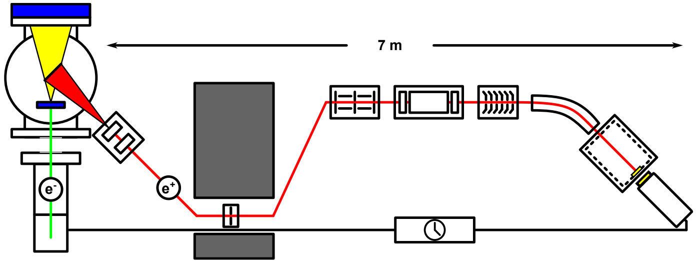


SC-LINAC in CW mode

EPOS / MePS facility



Positron User Facilities



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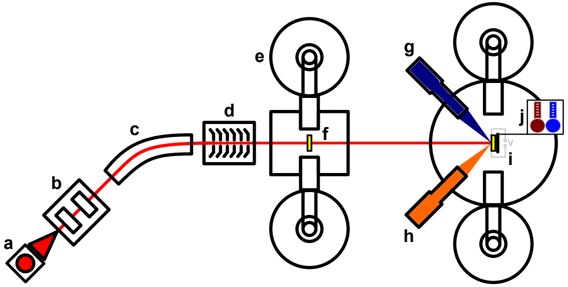
- ^{22}Na-based (2 GBq) e$^+$ beam
  30 eV – 30 keV for CDBS coupled to

AIDA

- 35K -1300 K
- MBE, ion irradiation
- sheet resistance



W. Anwand, et al., Def & Diff. Forum 331 (2012) 25

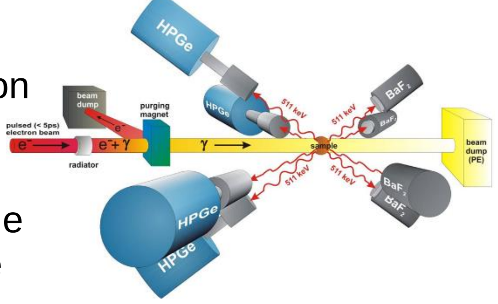
Mono-energetic Positron Source

- 35 MeV, 200 µA, 1.625 MHz electron beam drives
- 0.5 – 16 keV e$^+$ beam for PALS
  300 K – 550 K sample temperature
  typically **250 kcps event rate**
  (120 kcps at 511 keV)
- 10 Mevts PALS spectrum in 80 s, fully digital data taking and online processing

A.Wagner, et al., AIP Conf. Proc. 1970, 040003 (2018)

Gamma-ray induced Positron Source

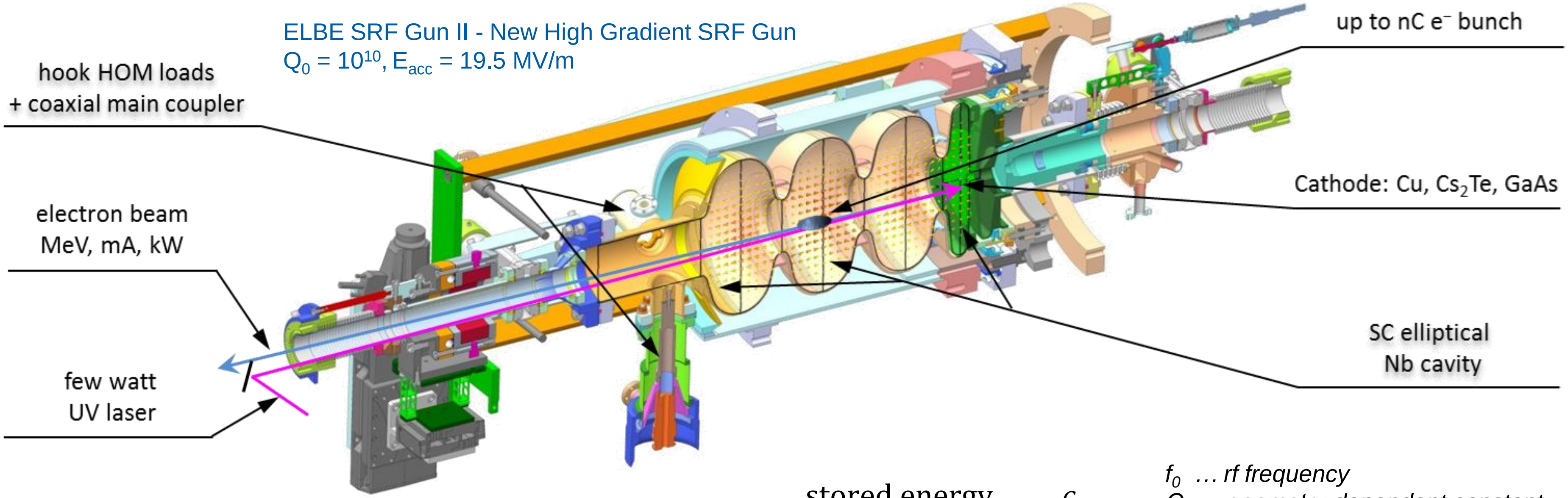
- 16 MeV, 700 µA electron beam generates bremsstrahlung
- e$^+$ formed inside sample
- Suitable for radioactive samples, gases, fluids



M. Butterling, et al., Nucl. Instr. Meth. B 269, 2623 (2011)

Hydrogen and defects in Niobium – SC cavities

A. Arnold et al., Proc LINAC2014, Geneva, Switzerland



ELBE SRF Gun II - New High Gradient SRF Gun
$Q_0 = 10^{10}$, $E_{acc} = 19.5$ MV/m

$$Q_0 = 2\pi f_0 \frac{\text{stored energy}}{\text{dissipated power}} = \frac{G}{R_s}$$

f_0 … rf frequency
G … geometry-dependent constant
R_s … surface resistivity

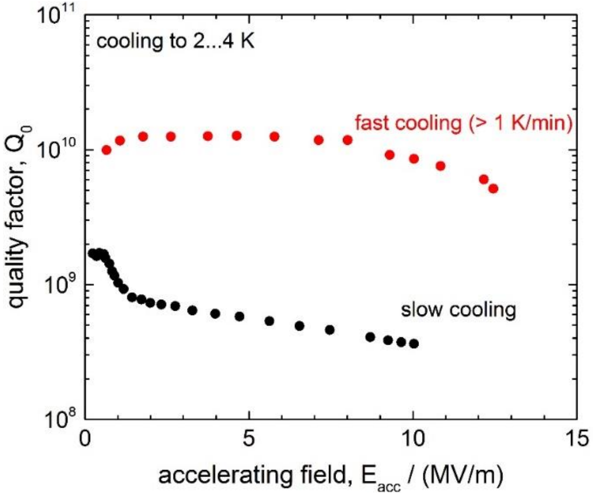
TESLA-type SRF cavities are used at CEBAF@JLAB, FLASH & XFEL (Hamburg), **ELBE@HZDR Dresden**, MESA (Mainz)

Cu: $R_s = 10$ mΩ, $Q_0 = 10^3 \dots 10^4$, max gradient 1 MV/m
Nb: R_s = few nΩ, $Q_0 = 10^{10} \dots 10^{11}$, max gradient 50 MV/m, operated at 2…4 K

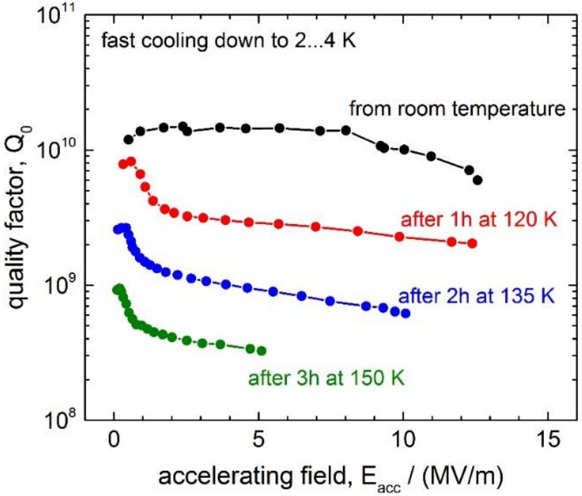
Thermal cavity treatment

- **Q disease**
- increase of the surface losses at cryogenic temperatures
- sets in even at low values of the applied accelerating field

significant losses for slow cooling



huge effect of temperature cycle



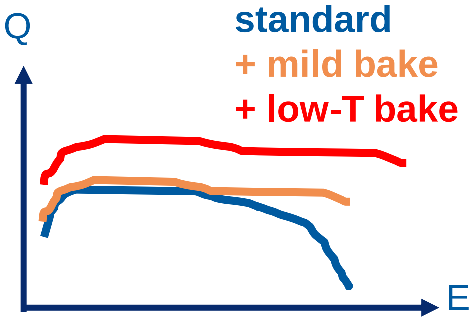
B. Aune *et al*., Proceedings of the 1990 Linac Conference, 253-255 (1990)
B. Bonin & R. Röth, Particle Accelerators 40, 59-83 (1992)
C. Antoine & S. Berry, AIP Conf. Proc.671, 176 (2003)

Curing Q disease

- rapid cool down inhibits the hydride formation
  - can cause damages due to thermal stress

- typical procedure[1,2] to increase Q and keep the H concentration below 10 ppm
  - chemical polishing (remove up to 100 µm), but incorporates H
  - baking at 1050 K for 3 h in vacuum (< 10^{-5} mbar) to remove H
    - several hundreds ppm of H remain in the lattice in the near-surface layer
  - chemical polishing (remove up to 100 µm), again incorporates H

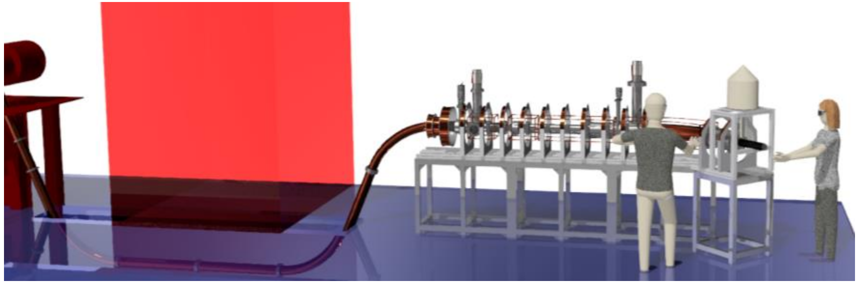
  - **mild bake:** at 390 K for 48 h (< 10^{-6} mbar)
    to avoid high-field Q slope (accidentally found!)
  - low-T bake[3]: only 350 K for first 2 h
    → Q losses reduced by factor 2
    → increased achievable accelerating field by >10%

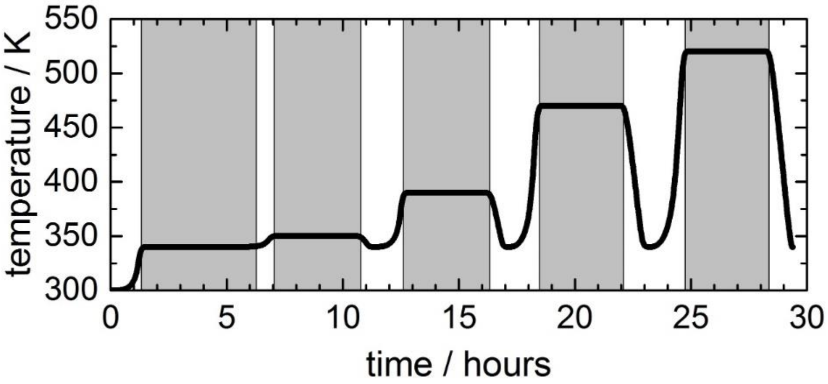


[1] P. Kneisel et al., AIP Con Proc 927, 84 (2007) JLAB
[2] G. Ciovati et al., Phys Rev Acc & Beams 13.2, 022002 (2010), JLAB
[3] A. Grassellino et al., arXiv 1806.09824 (2018)

Vacancy kinetics

- In-situ baking at MePS





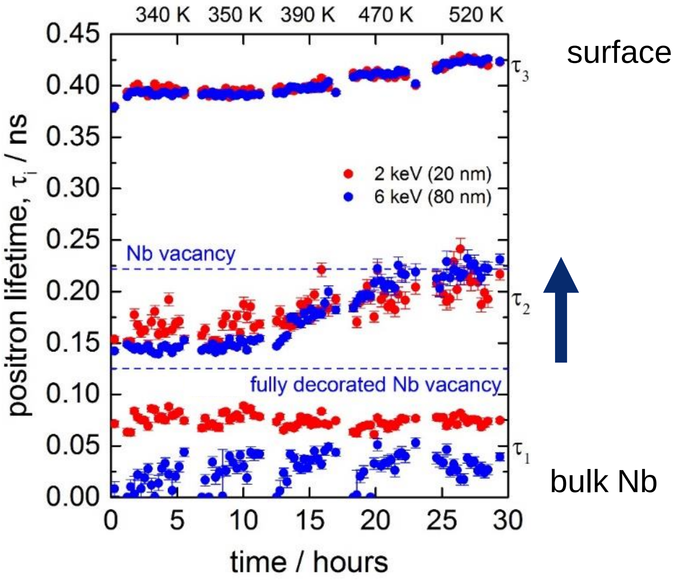
- **Mono-energetic Positron Source MePS at HZDR:**
- depth-resolved PALS up to 10 keV (in 2019)
- high intensity: 10^6 counts in 2 minutes
- well-suited for study of dynamic effects

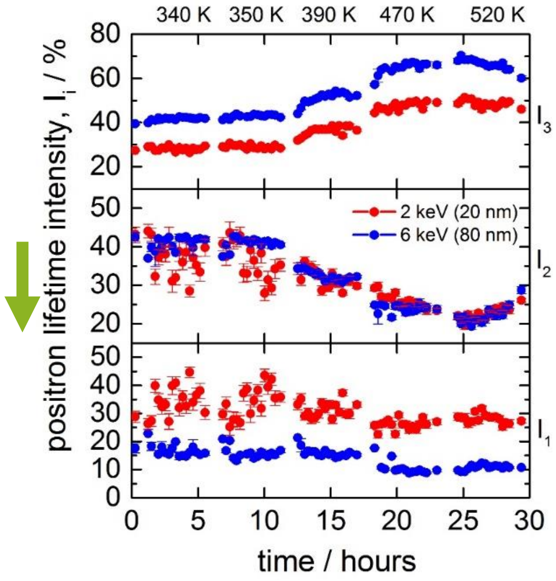
In-situ measurements

- in-situ baking at different temperatures for 4 hours and measuring at different energies all the time
- depth profile at 340 K after each annealing step

Vacancy dynamics

V+nH show complex dynamics near surface





Collaboration:
Hamburg U, DESY, HZ Berlin,
U Wuppertal, U Siegen,
U Kaiserslautern, U Bremen,
CU Prague, CTU Prague

RF layer w/ T_c =1.8 K

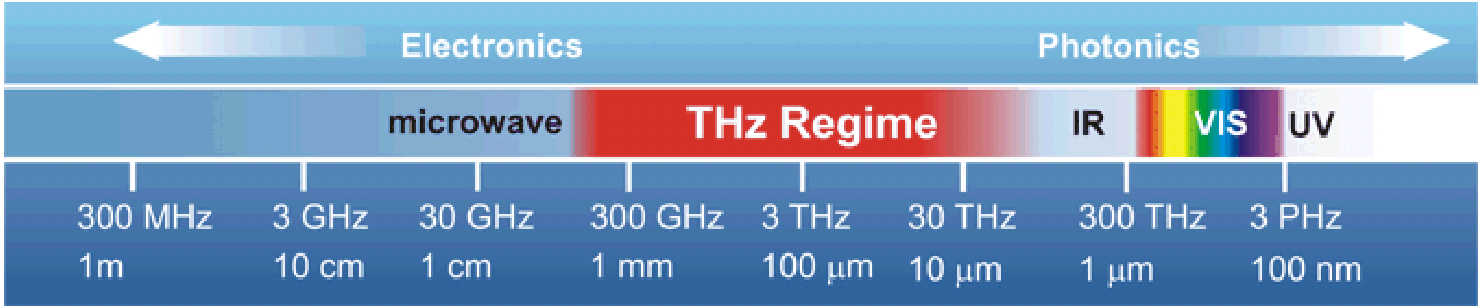
- T = 390 K: shows most significant changes (already after short times)
- T > 390 K: hydrogen released from complexes (τ_2 increases)
  v+nH concentration (I_2) decreases

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

Secondary beams: THz radiation – from electronics to photonics

$1\ \mathrm{THz} \mathrel{\hat{=}} 300\ \mu\mathrm{m} \mathrel{\hat{=}} 33\ \mathrm{cm}^{-1} \mathrel{\hat{=}} 4.1\ \mathrm{meV}$

oscillators **lasers**



Graphik: R. Huber

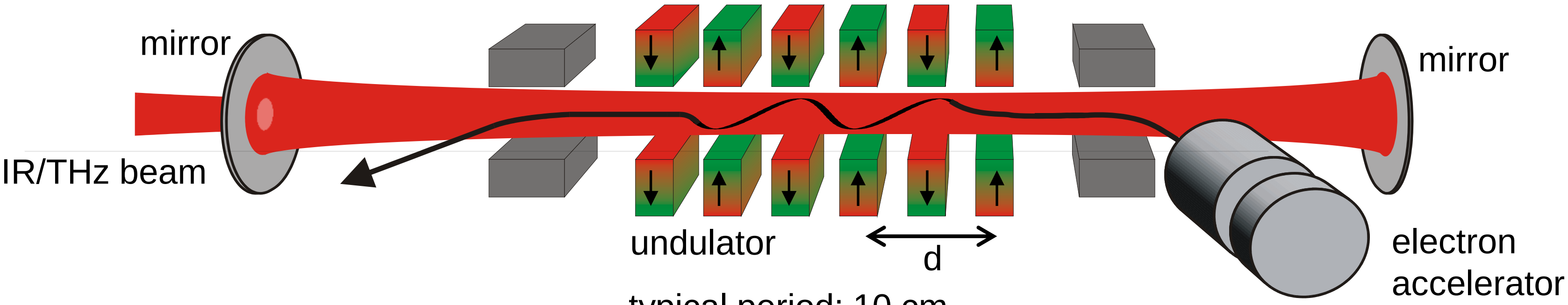
in various units:

Frequency: 0.3 – 30 THz
Wavenumber: 10 – 1000 cm^{-1}
Energy: ~1 – 100 meV
Wavelength: 1 mm – 10 µm

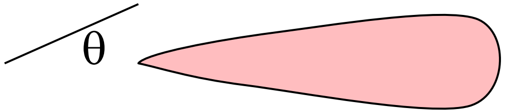
In some ranges no good laboratory (table-top) sources are available:
THz,
UV, X-ray

Secondary beams: Free-Electron Lasers

FEL – 1971 (Stanford), 2005 UV & 2017 X-ray (FLASH, DESY Hamburg)



typical period: 10 cm



Undulator parameter ***K*** = θ · γ = eBd/2πmc ~ 1

(θ is the maximum electron deflection angle,

1/γ is also the width of the emitted radiation cone)

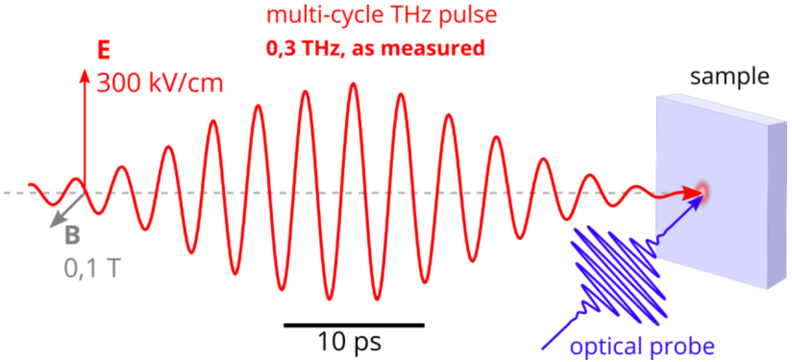
$$\boxed{\lambda_L = \frac{\lambda_u}{2\gamma^2}\left(1+\frac{K^2}{2}\right)}$$

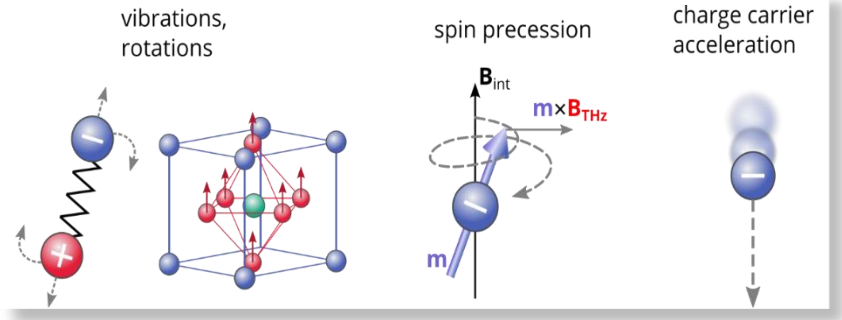
wavelength of undulator radiation

THz pulses as pump for nonlinear dynamics

DRIVE

THz pulses **drives** material into transient exotic phase
→ field (***E*** or ***B***) acts as **selective** (non)linear perturbation





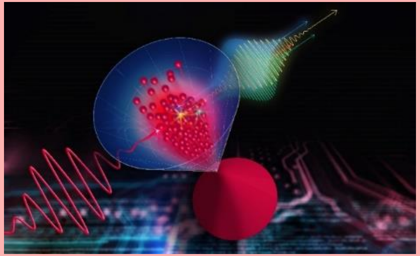
- Coherent **selective excitation** of relevant modes, avoiding parasitic electronic excitation
- How and on what timescales do degrees of freedom interact?

- Strong modifications of macroscopic properties possible → phase transitions, metastable states, tailored functionality, quasiparticle excitations

B. Green et al., *Sci. Rep.* **6**, 22256 (2016)
Deinert, J.-C., Kovalev, S. and Gensch, M., *Phys. Unserer Zeit* **54**, 12-17 (2023)

THz science cases

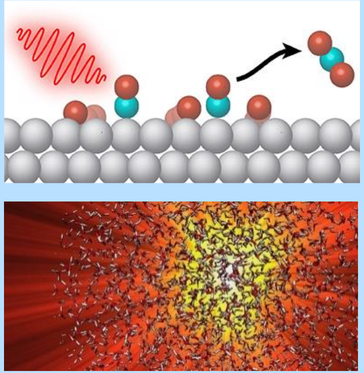
Control of **Materials** *with THz light*



- Electronic or structural phase transitions
- THz nonlinear optics
- Superconductivity
- Spin dynamics
- Exotic quantum properties

→ Materials and processes for information and energy technology

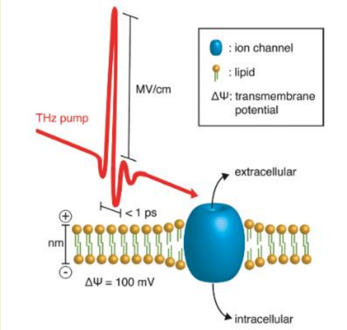
Selective **Chemistry**



- Trigger for reactions
- Steering reaction pathway
- Temperature jumps

→ Catalytic materials and processes, fundamentals of water chemistry

Biology *in highest electric fields*

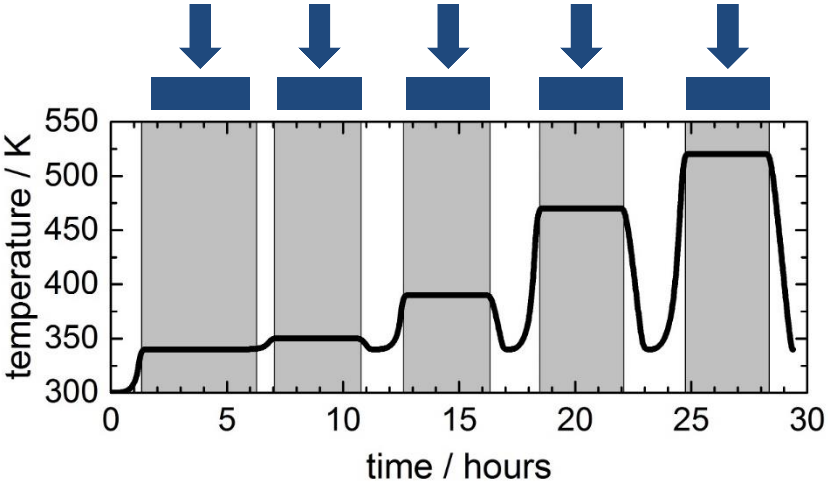


- Hydration shell dynamics
- Potential-driven processes in aqueous solutions

→ Elementary cellular mechanisms

Vacancy kinetics during in-situ low-T baking

- **In-situ Positron Annihilation Lifetime Spectroscopy at pELBE**



In-situ measurements

- in-situ baking at different temperatures for 4 hours and measuring at different energies all the time
- depth profile at 340 K after each annealing step

- focus on the vacancy dynamics: measurement during temperature treatment
- study two different material depths: 20 nm and 80 nm (RF layer ~ 100 nm)