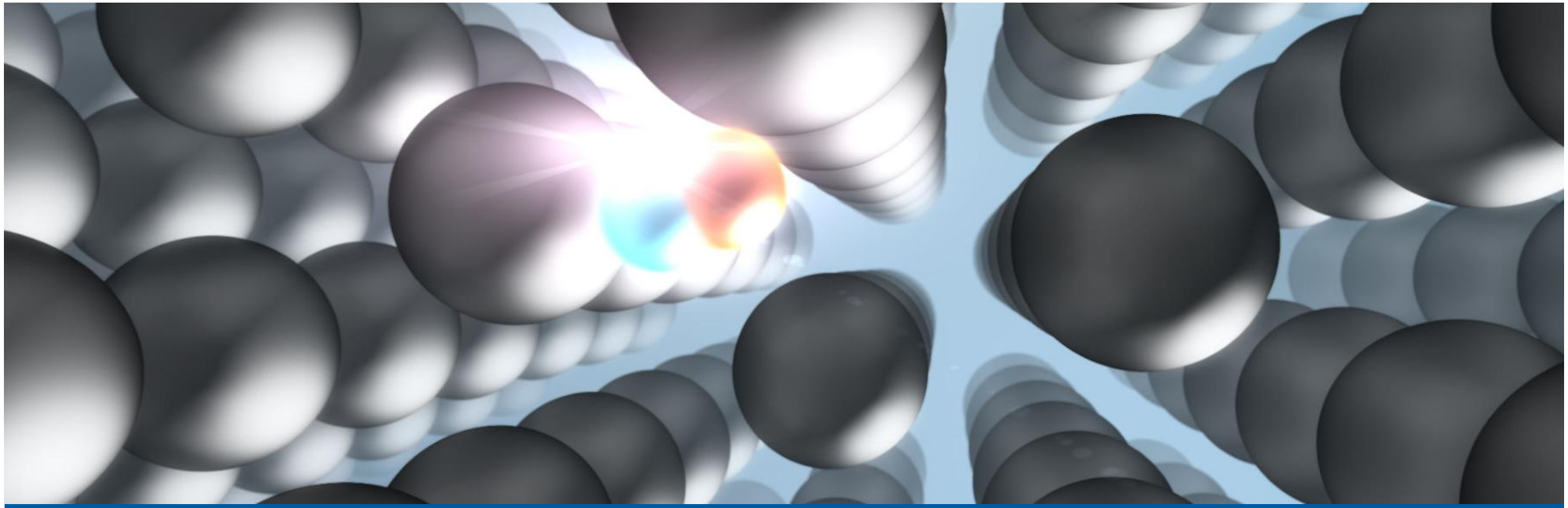


The high-intense pulsed positron source MePS

Recent developments and future plans

Institute of Radiation Physics · Nuclear Physics · Maik Butterling · m.butterling@hzdr.de · www.hzdr.de

The high-intense pulsed positron source MePS

Recent developments and future plans

M.O. Liedke, E. Hirschmann, A.G.A. Elsherif, K. Potzger, D. Stach, M. Görler, A. Hartmann, A. Wagner

Helmholtz-Zentrum Dresden-Rossendorf, HZDR, Germany

R. Krause-Rehberg, M. Elsayed, M. John, M. Jungmann, A. Müller

Martin-Luther-Universtiy Halle-Wittenberg, Germany

D. Enke, G. Dornberg
University Leipzig, Germany
➢ porous glasses for catalysts

S. Schulz, R. Ecke, N. Köhler
Fraunhofer ENAS, TU Chemnitz, Germany
➢ low-k dielectrics

H. Weise, M. Wenskat
DESY Hamburg, Germany
➢ Ni-hydrides in SC-Niobium

E. Menendez, J. de Roja
University Barcelona, Spain
➢ Magneto-ionics

M. Fujinami, L. Chiari
University Chiba, Japan
➢ H-embrittlement in Ni

M. Pietrow, R. Zaleski
University Lublin, Poland
➢ Ps light

Content

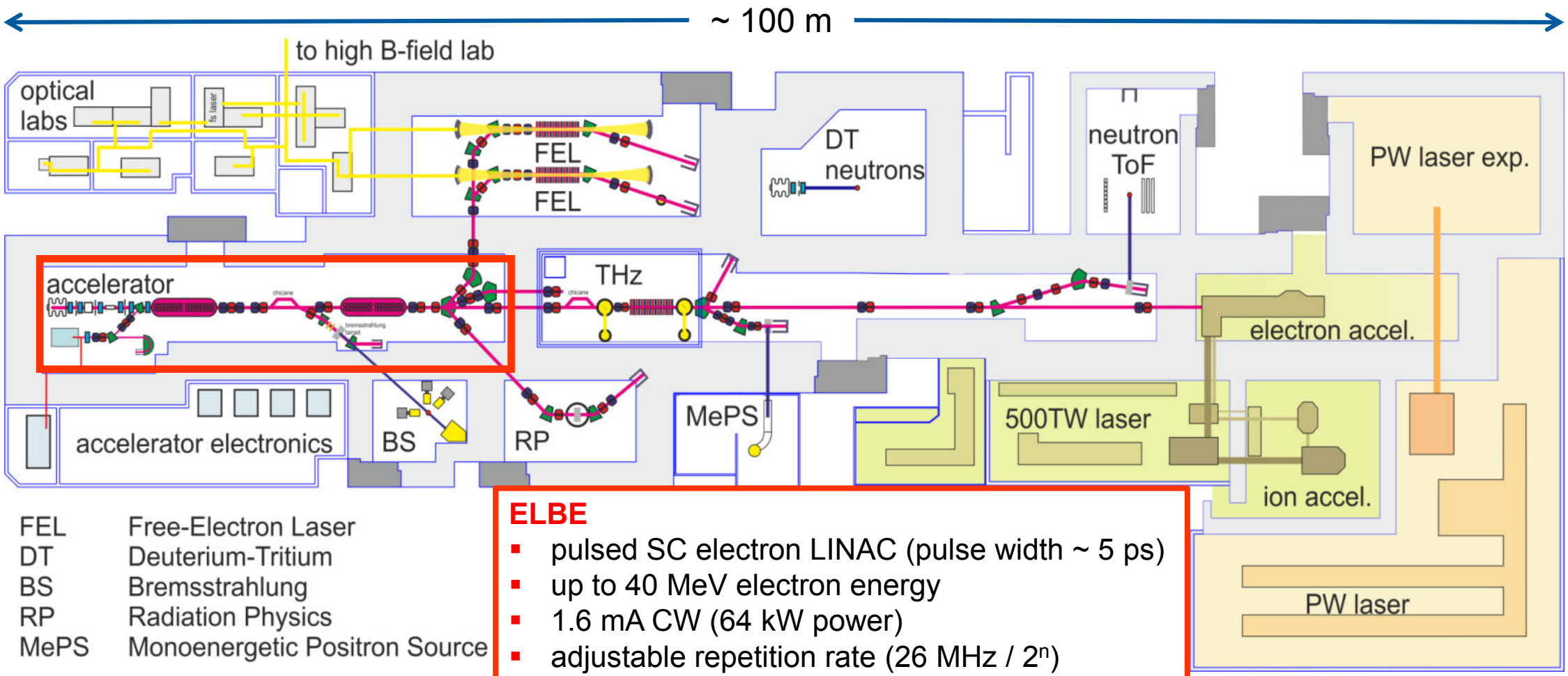
- Positron Annihilation Spectroscopy at the HZDR
  - Mono-energetic Positron Source - MePS
  - Optimized ultra-fast digital DAQ
- Controlled extracted sodium borosilicate (NBS) glass

} PhD work of E. Hirschmann

MARTIN-LUTHER-UNIVERSITÄT HALLE-WITTENBERG DRESDEN concept HZDR

Positron Annihilation Spectroscopy at the HZDR

The superconducting electron LINAC ELBE



FEL	Free-Electron Laser
DT	Deuterium-Tritium
BS	Bremsstrahlung
RP	Radiation Physics
MePS	Monoenergetic Positron Source

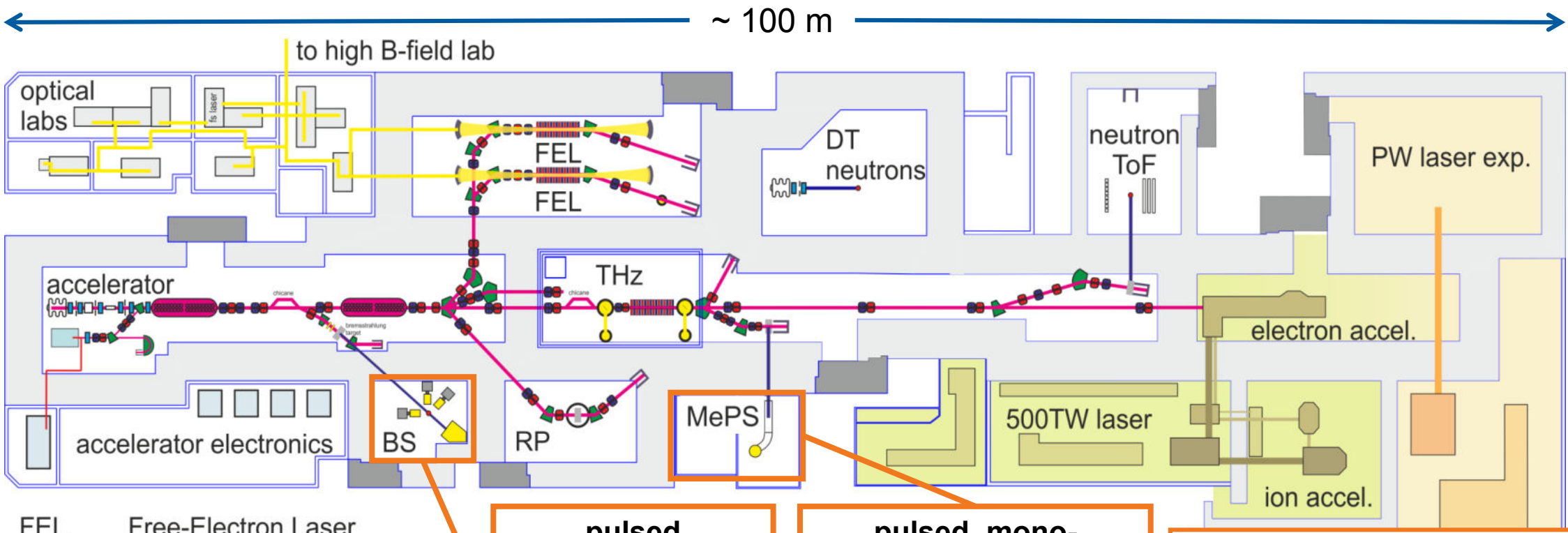
ELBE

- pulsed SC electron LINAC (pulse width ~ 5 ps)
- up to 40 MeV electron energy
- 1.6 mA CW (64 kW power)
- adjustable repetition rate (26 MHz / 2^n)

MARTIN-LUTHER-UNIVERSITÄT HALLE-WITTENBERG DRESDEN concept HZDR

Positron Annihilation Spectroscopy at the HZDR

The superconducting electron LINAC ELBE



FEL	Free-Electron Laser
DT	Deuterium-Tritium
BS	Bremsstrahlung
RP	Radiation Physics
MePS	Monoenergetic Positron Source

pulsed bremsstrahlung
16 MeV
Gamma-induced Positrons for bulk samples / liquids

pulsed, mono-energetic positrons
0.2 - 20 keV

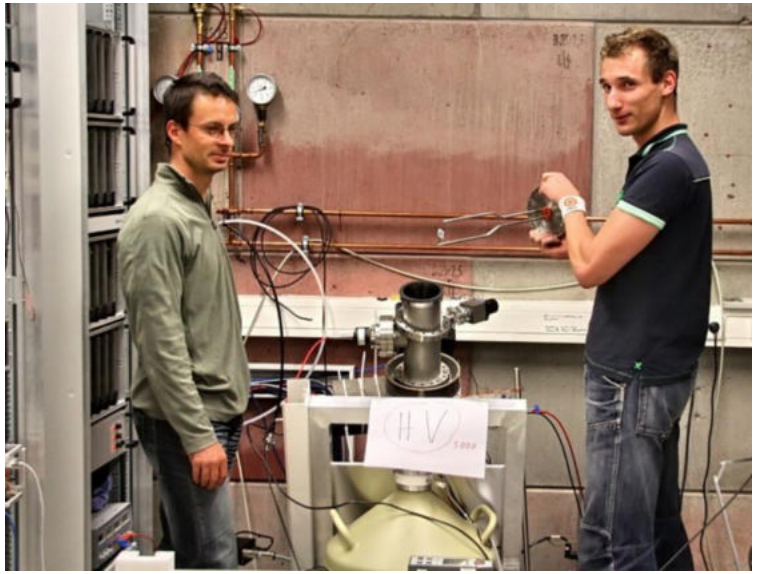
AIDA I & SPONSOR:
RI based mono-energetic positrons
+ 2 digital PALS systems

MARTIN-LUTHER-UNIVERSITÄT HALLE-WITTENBERG DRESDEN concept HZDR

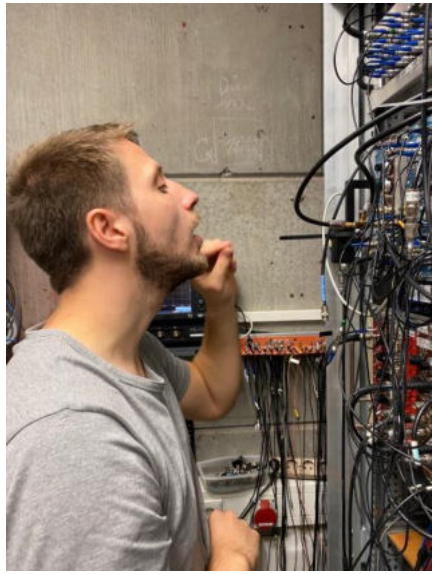
Mono-energetic Positron Source - MePS

A long journey

Reinhard Krause-Rehberg moving lead bricks (2008)



Marco Jungmann and Maik Butterling installing the very first sample at MePS (2008)

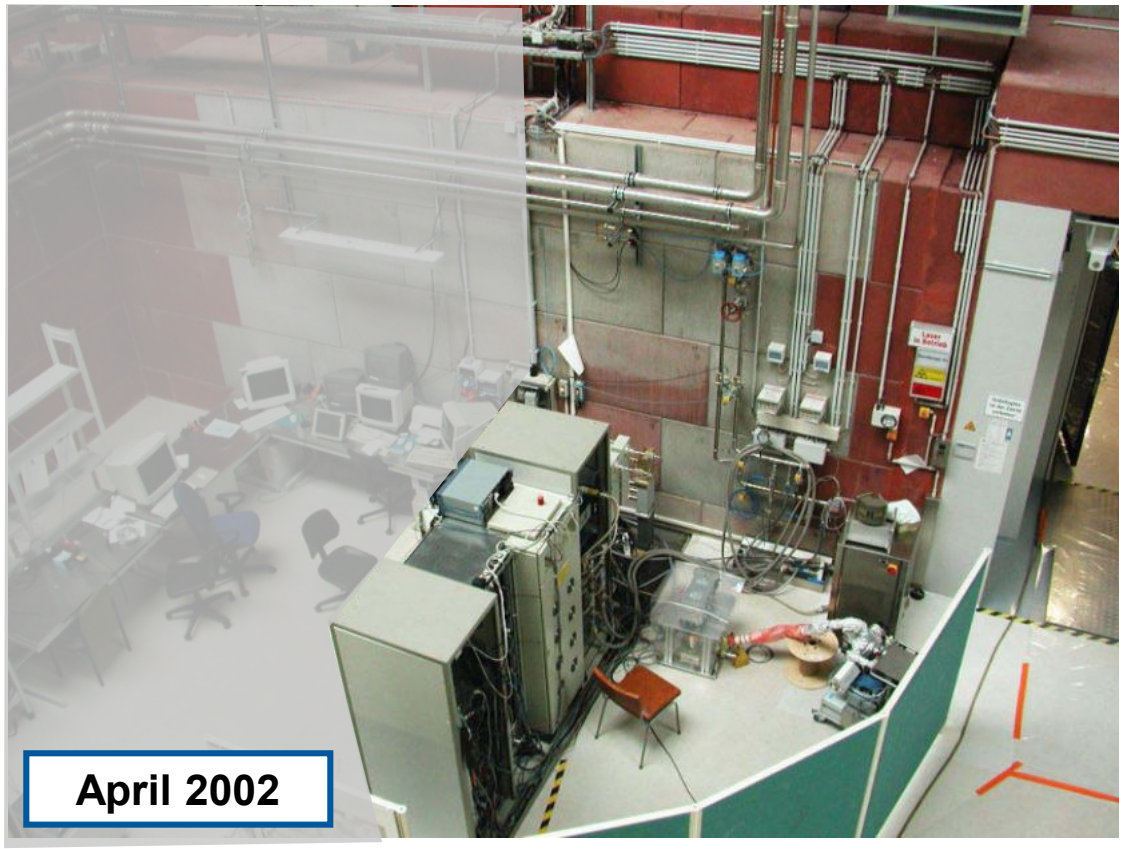


Eric Hirschmann looking for signals to process (2020)

Andreas Wagner praying for more positrons (2020)

Mono-energetic Positron Source - MePS

A long journey



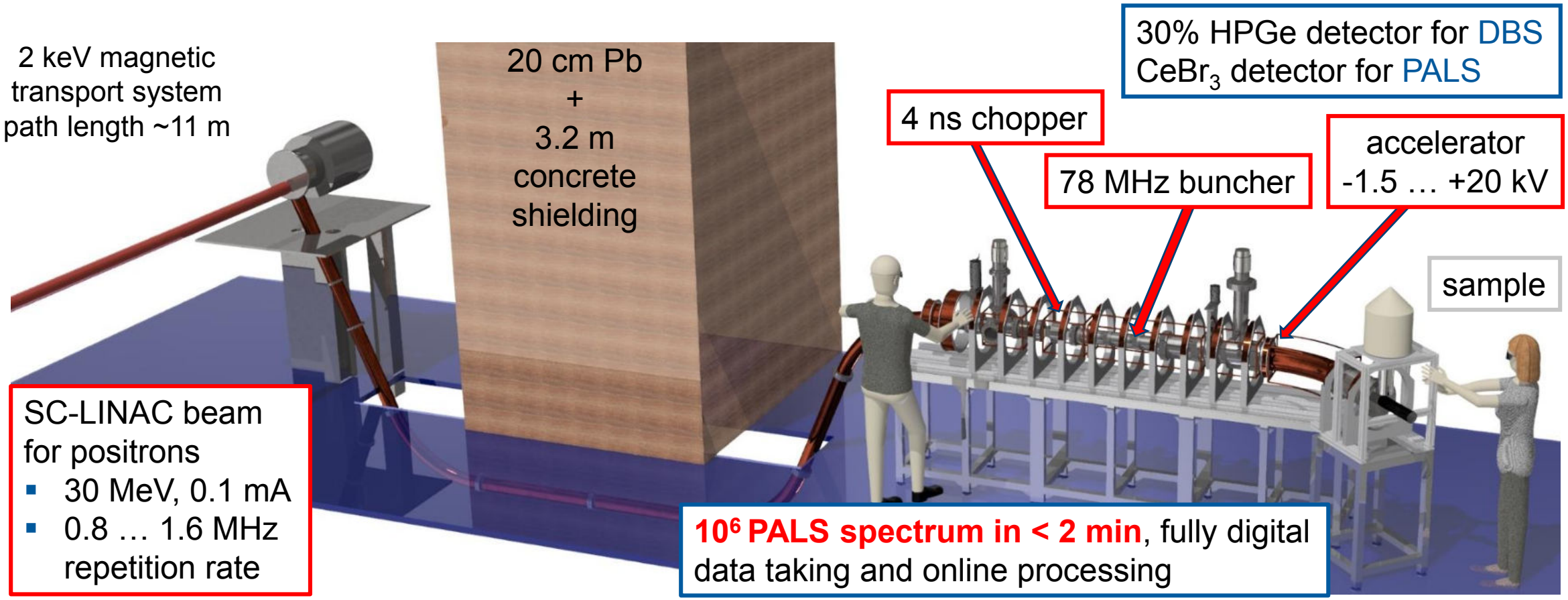
not even walls or a door around the lab

two beam lines on top of each other

MARTIN-LUTHER-UNIVERSITÄT HALLE-WITTENBERG DRESDEN concept HZDR

Mono-energetic Positron Source - MePS

Current setup and status

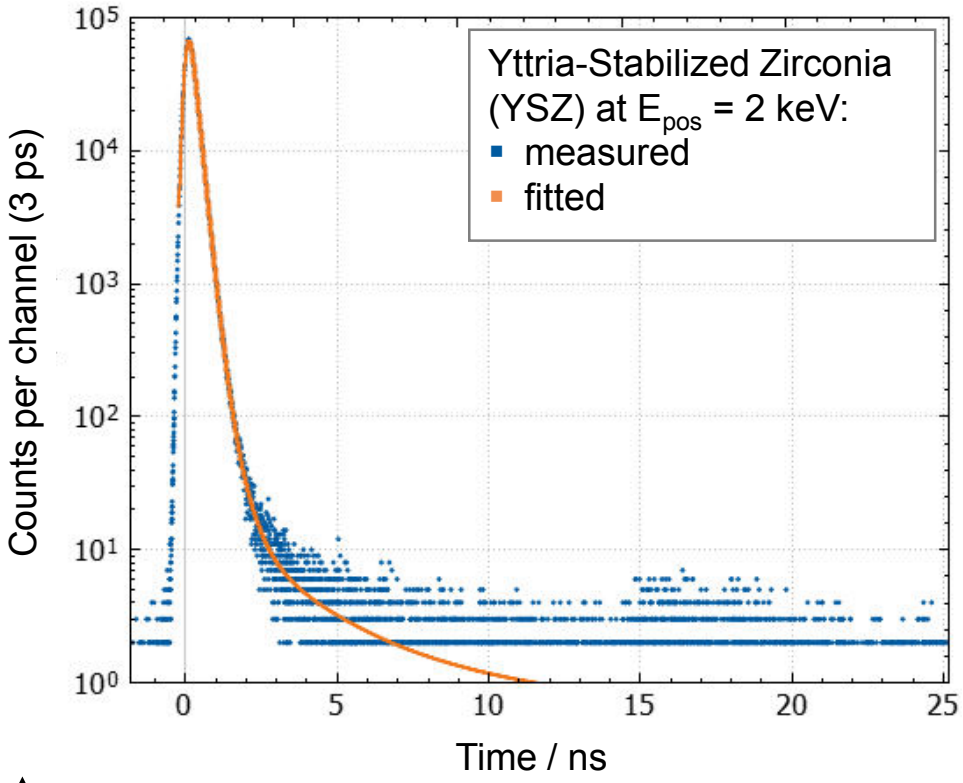


start → Positron Annihilation Lifetime spectroscopy ← stop

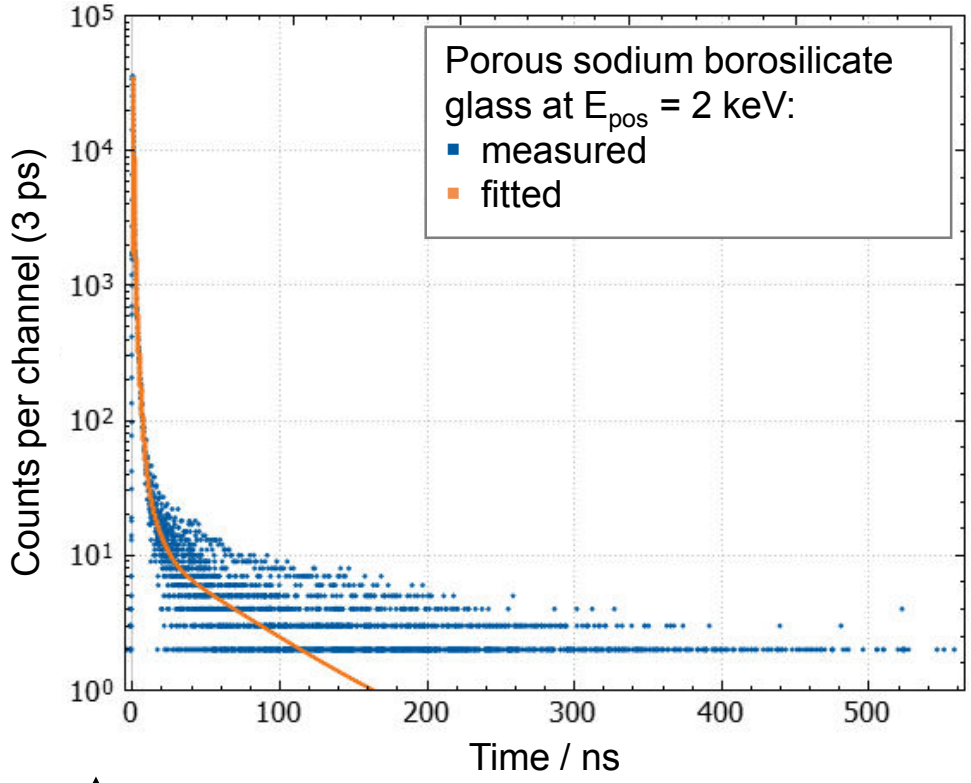
MARTIN-LUTHER-UNIVERSITÄT HALLE-WITTENBERG DRESDEN concept HZDR

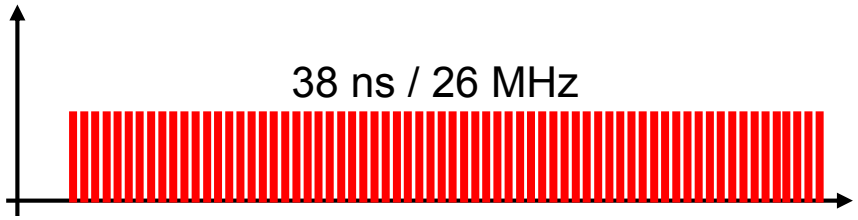
Mono-energetic Positron Source - MePS

Adjustable repetition frequency and sharp pulses of ELBE

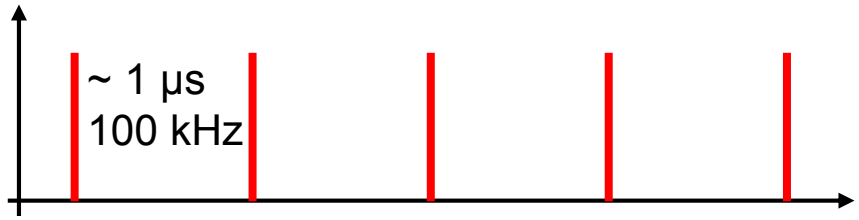


26 MHz / 2^n
(n = 0…8)





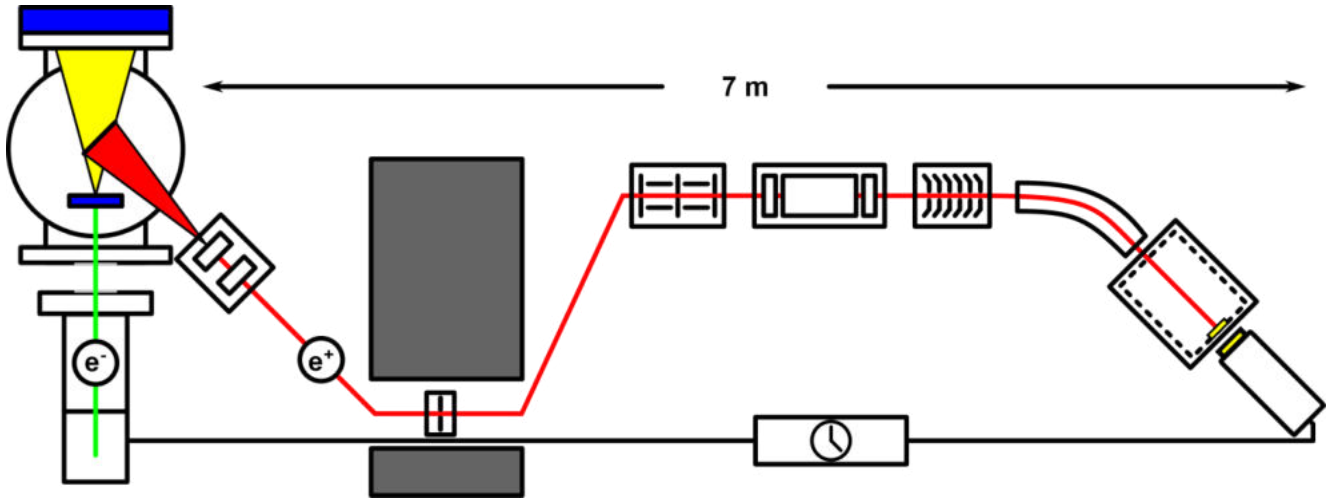
no limits in time window
no pulse pile-up
no background

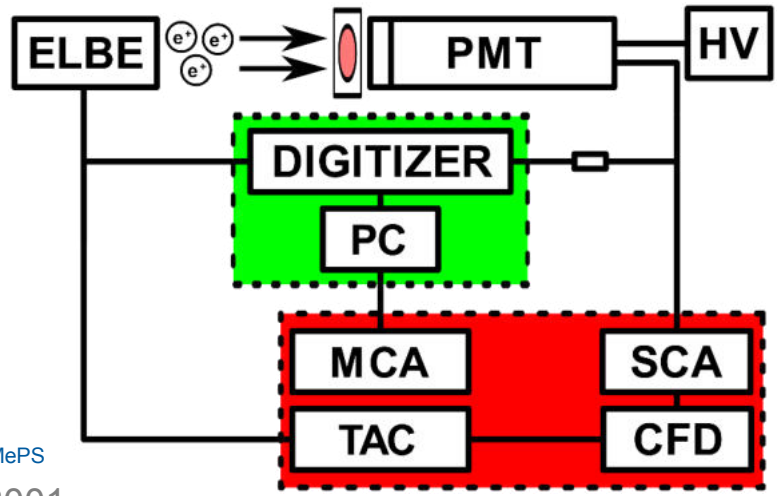


MARTIN-LUTHER-UNIVERSITÄT HALLE-WITTENBERG
DRESDEN concept
HZDR

Optimized ultra-fast digital DAQ

SPDevices digitizer





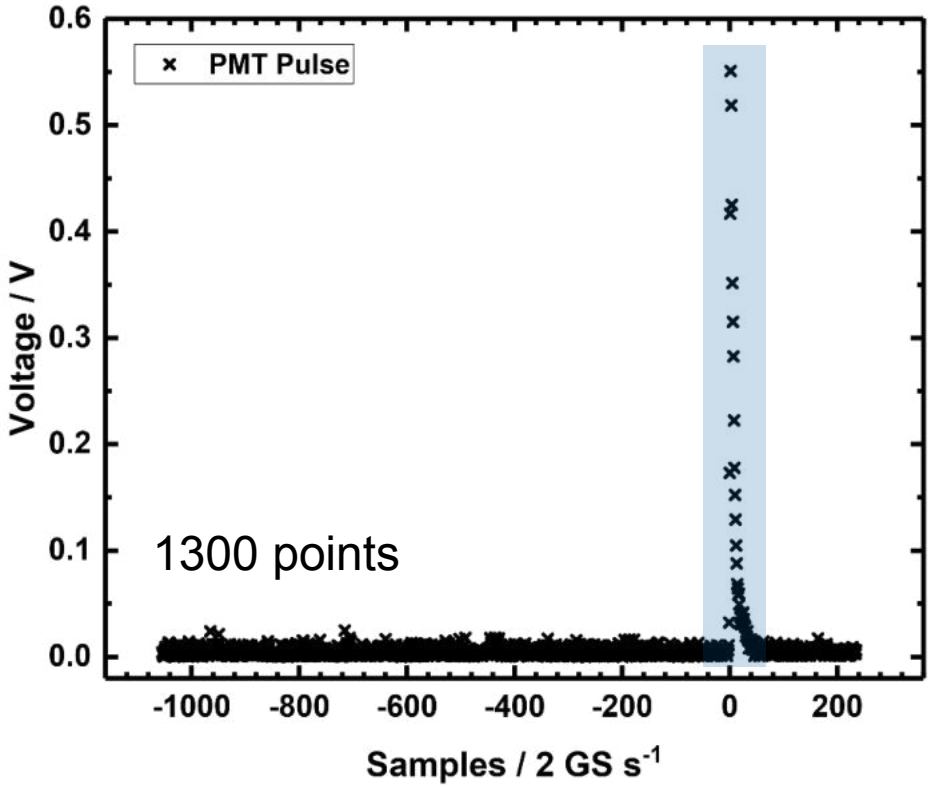
- SPDevices ADQ14-FWPD
  - 2 GS/s
  - 14-bit
  - 2 channels
- pulse detection firmware
- individual channel trigger
- no external trigger logic needed

E. Hirschmann et al 2021 JINST 16 P08001

MARTIN-LUTHER-UNIVERSITÄT HALLE-WITTENBERG DRESDEN concept HZDR

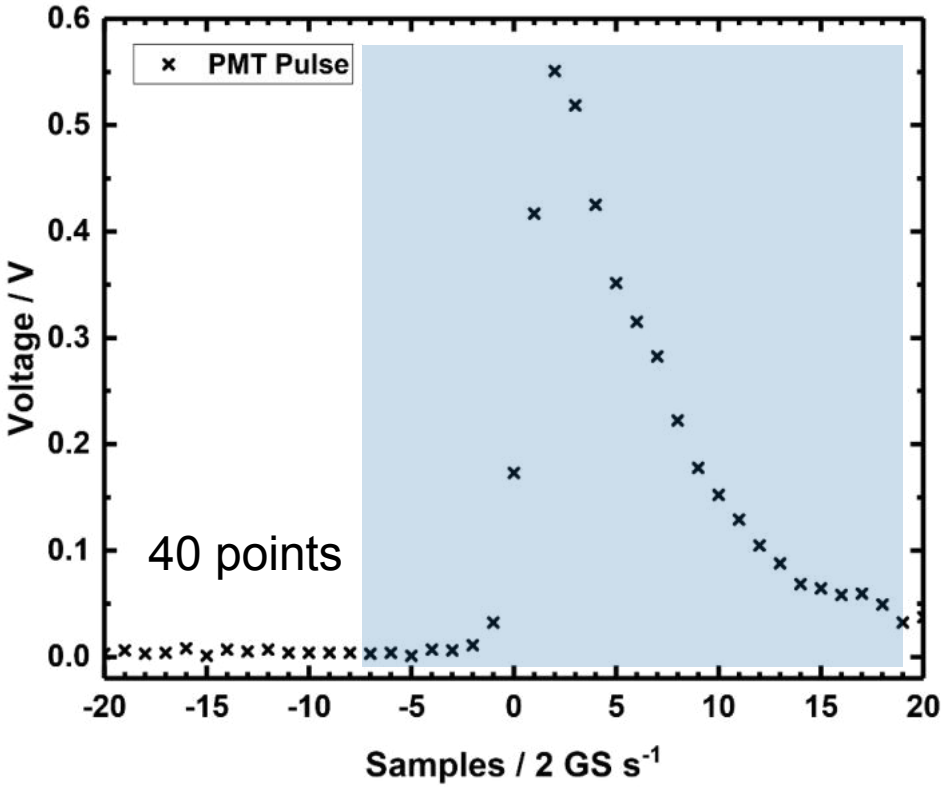
Optimized ultra-fast digital DAQ

Pulse detection firmware



Reduced data traffic

40 points instead of 1300 (650 ns 2 GS/s) per channel

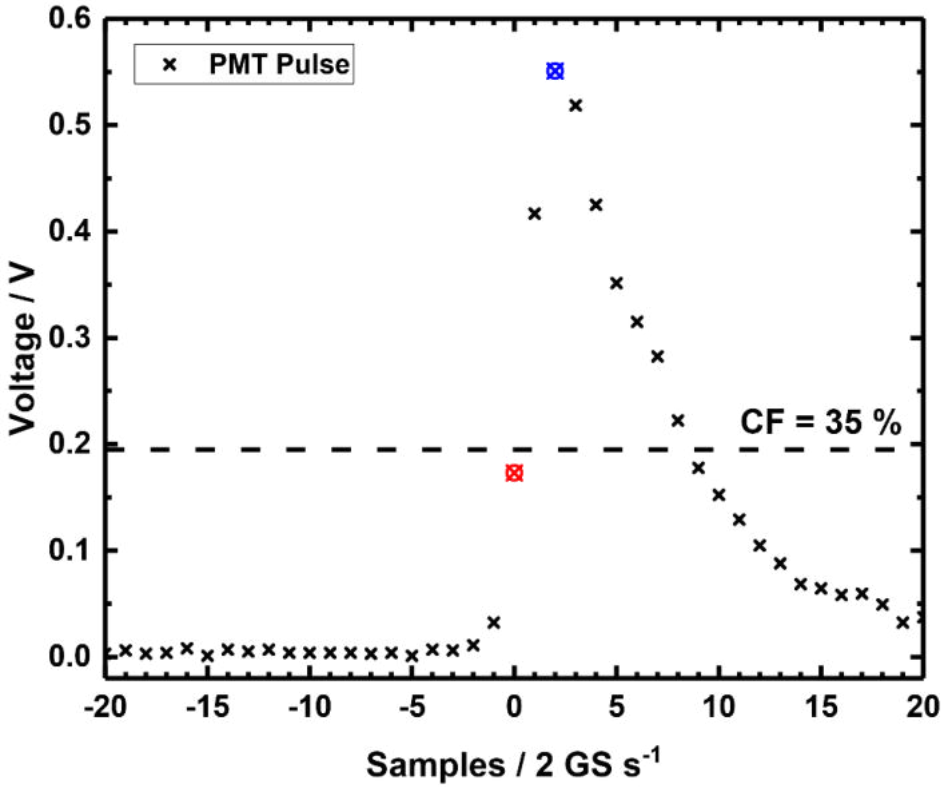


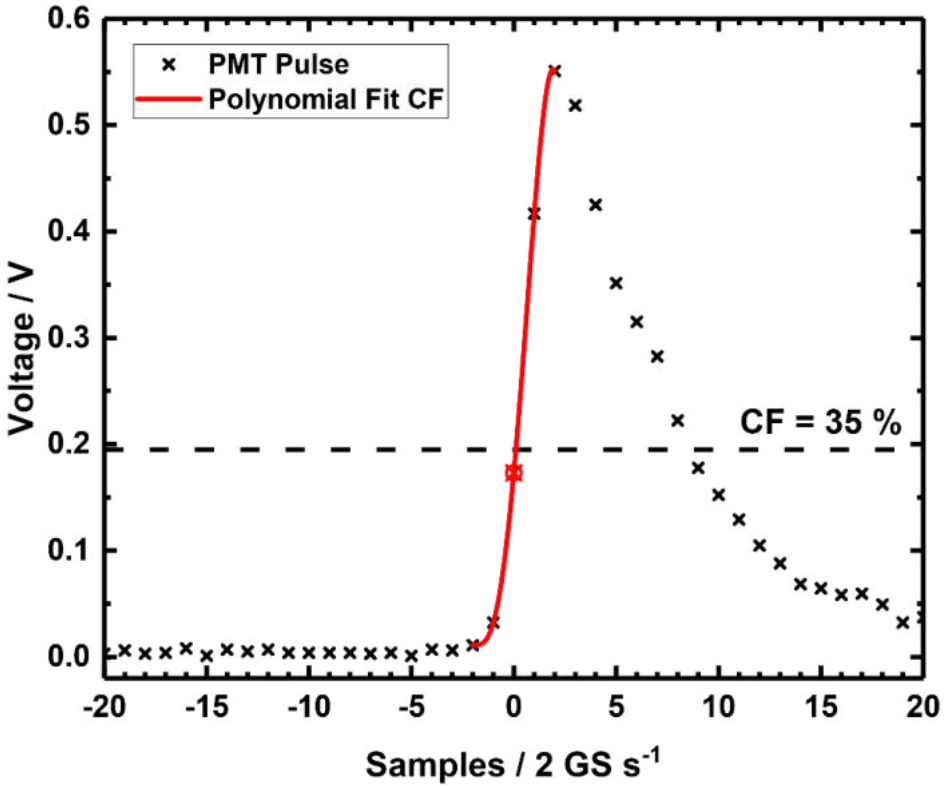
- FPGA-based trigger logic
- individual time stamp will be calculated from transferred global time stamp

E. Hirschmann et al 2021 JINST 16 P08001

Optimized ultra-fast digital DAQ

Online pulse interpolation and lifetime calculation





- necessary: pulse maximum and closest point to CF level of the inverted PMT signal
- usual idea: polynomial fit (3th or 4th degree) using 5 points around CF → time-consuming for high signal rates

E. Hirschmann et al 2021 JINST 16 P08001

Optimized ultra-fast digital DAQ

Householder method

- Polynom of 4th order

$$y(x) = a_0 + a_1 x + a_2 x^2 + a_3 x^3 + a_4 x^4$$

- Householder method

$$x_1 = \frac{a_0^*(2\, a_0^*\, a_1\, a_2 - {a_0^*}^2\, a_3 - {a_1}^3)}{{a_1}^4 - 3\, a_0^*\, {a_1}^2\, a_2 + 2{a_0^*}^2\, a_1\, a_3 + {a_0^*}^2({a_2}^2 - a_0^*\, a_4)}$$

shift the polynom to CF level: $a_0^* = a_0 - y_{CFD}$

- final time stamp of the pulse
  $t = t_{FPGA} + x_1$

Householder method

- numerical algorithms for solving the nonlinear equation f(x) = 0 (first order is equal to Newton's method)
- interpolation instead of polynomial fit
- high accuracy for already the first iterations

Performance

- avoid complex functions like roots, higher exponentials or case selections

➢ Interpolation of a polynomial of 4th order much faster than fitting a polynomial of 3rd order

E. Hirschmann et al 2021 JINST 16 P08001

Optimized ultra-fast digital DAQ

Performance optimisation

Qualitative improvements

- improved time resolution (now only limited by detector)
- PALS with adjustable binning: actually 3 ps/channel
- background reduction
- pulse pile-up reduction
- better energy window selection on 511 keV
- list-mode
- filters and corrections

Quantitative improvements

- reduced data traffic (pulse detection)
- multi-threading
- 120 kcps so far
  - ➢ dynamic measurements

cubic spline interpolation
5 kHz

cubic spline interpolation
+ pulse detection
95 kHz

cubic spline interpolation
+ pulse detection
+ multi-threading
375 kHz

Householder method
+ pulse detection
+ multi-threading
5600 kHz

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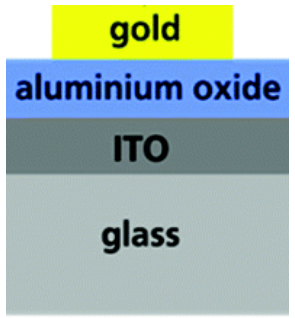
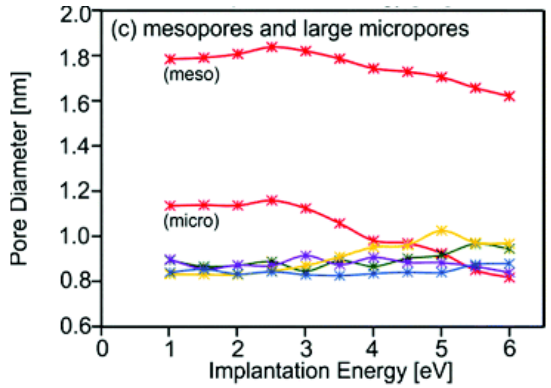
MARTIN-LUTHER-UNIVERSITÄT HALLE-WITTENBERG DRESDEN concept HZDR

Optimized ultra-fast digital DAQ

Recent measurement examples

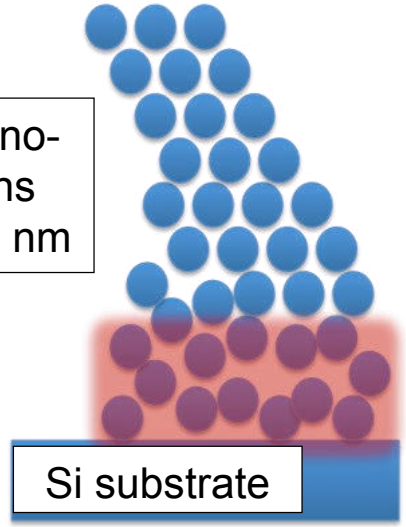


micro-pores (1…2 nm) in metal oxide double layer capacitors
Al_2O_3, ZnO



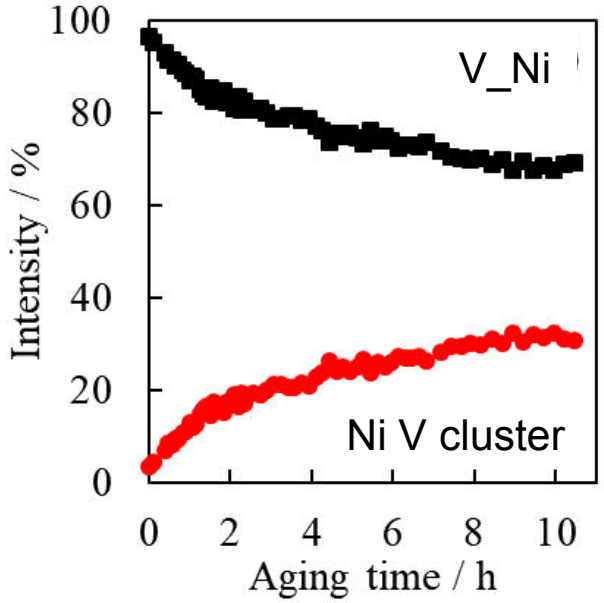
R.C. Hoffmann *et al.*, J. Mater. Chem. C 6 (2018), 9501
R.C. Hoffmann *et al.*, Chem. Eur. J. 27 (2021), 5422-5431
R.C. Hoffmann *et al.*, Dalton Trans. 50 (2021), 8811-8819

SiO_2 nano-columns
80°, 600 nm



A. García-Valenzuela *et al.*, Microporous and Mesoporous Materials 295 (2020),109968

Characterisation of micro-pores in plasma deposited SiO_x films
C. Hoppe *et al.* J. Phys. D 53 (2020), 475205



Vacancy cluster growth in Ni at RT after H charging

L. Chiari *et al.*, in press, Acta Materialia (2021), 117264

MARTIN-LUTHER-UNIVERSITÄT HALLE-WITTENBERG DRESDEN concept HZDR

Controlled extracted sodium borosilicate (NBS) glass

Controlled porous glass - part of PhD work of Eric Hirschmann

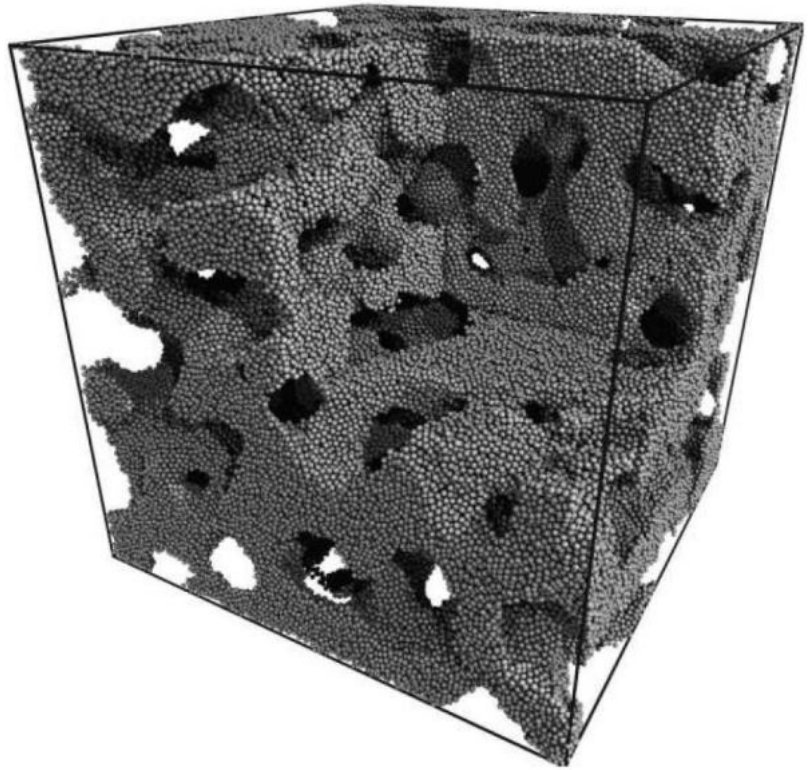
- high-silica glass that contains pores with a specific size distribution between 1 … 1000 nm

Application

- catalysts, filters, chemical and optical sensors

Control parameters

- temperature and time of heat treatment
  → pore size / structure
- extraction time → porous layer thickness



Model of a CPG with 47 % porosity, 3.23 nm mean pore diameter D. Enke, habilitation, Martin-Luther-Universität Halle-Wittenberg, 2005. L. D. Gelb *et al.* Langmuir 15/2 (1999) 305-308

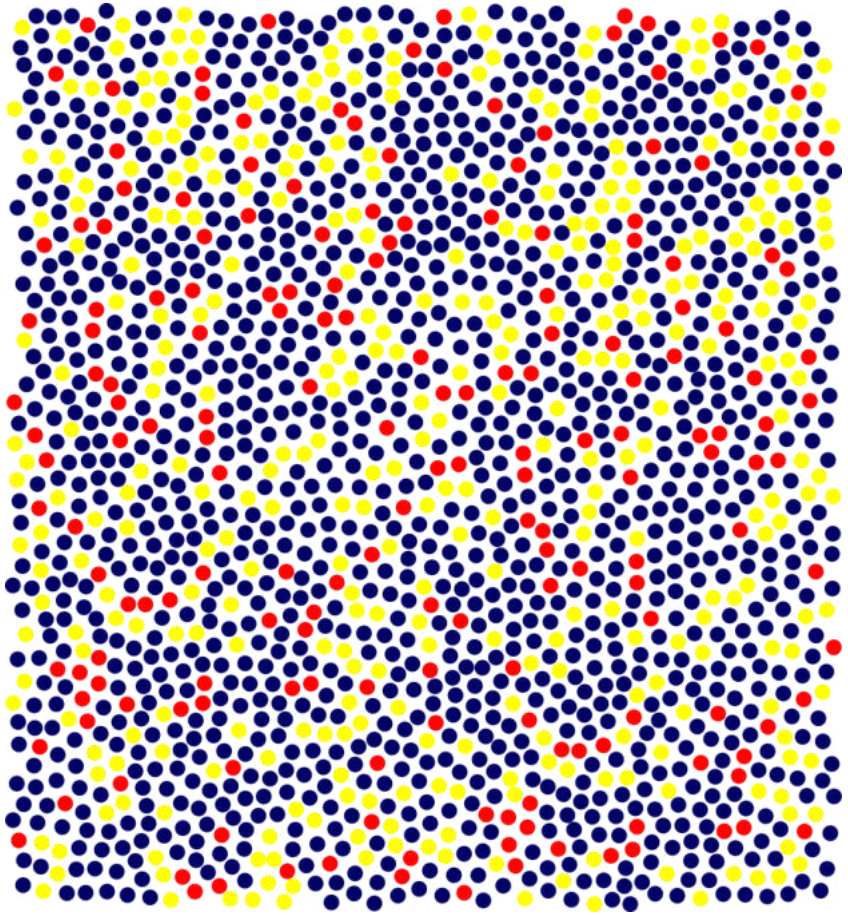
Controlled extracted sodium borosilicate (NBS) glass

Generation of NBS glass (D. Enke, University of Leipzig, Germany)

Material	Functionality	Weight % in CPG glass	Weight % in NBS glass
SiO_2	network former	50 - 70	70
B_2O_3	network former	15 - 40	23
Na_2O	network converter	3 - 10	7

VYCOR process:

- heat treatment of NBS glass



H. P. Hood et al., US2106744A (1938) and US2221709A (1940)

Controlled extracted sodium borosilicate (NBS) glass

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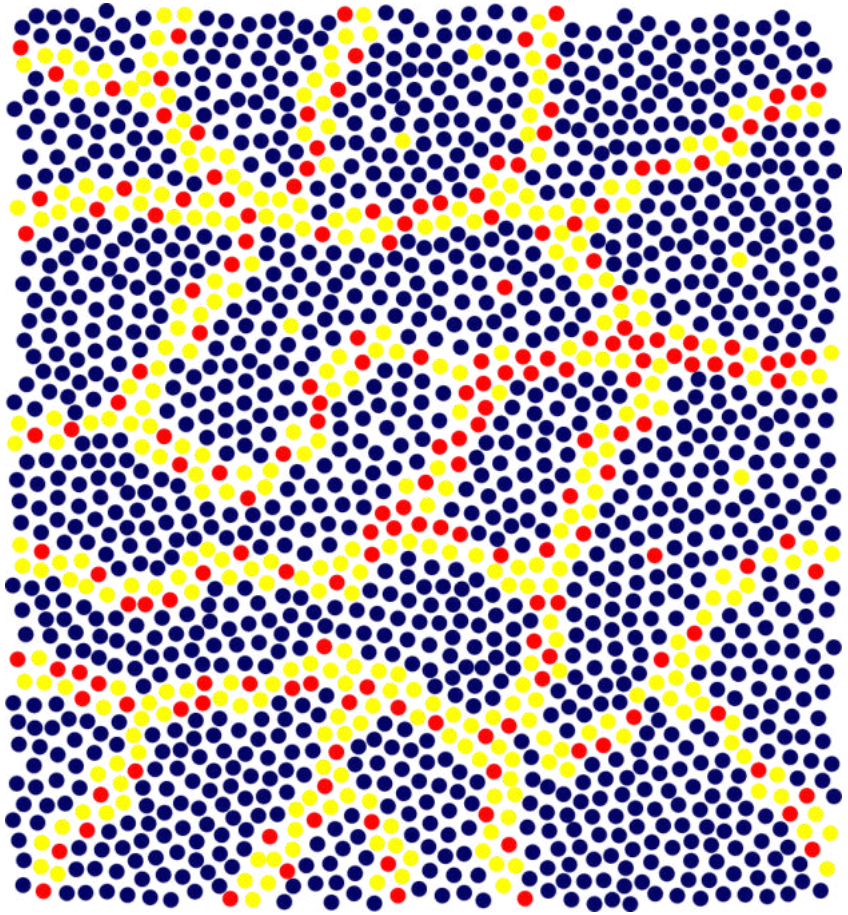

VYCOR process:

- heat treatment of NBS glass

→ spinodal phase separation

→ generation of sodium-rich borate phase and silicate phase

H. P. Hood et al., US2106744A (1938) and US2221709A (1940)

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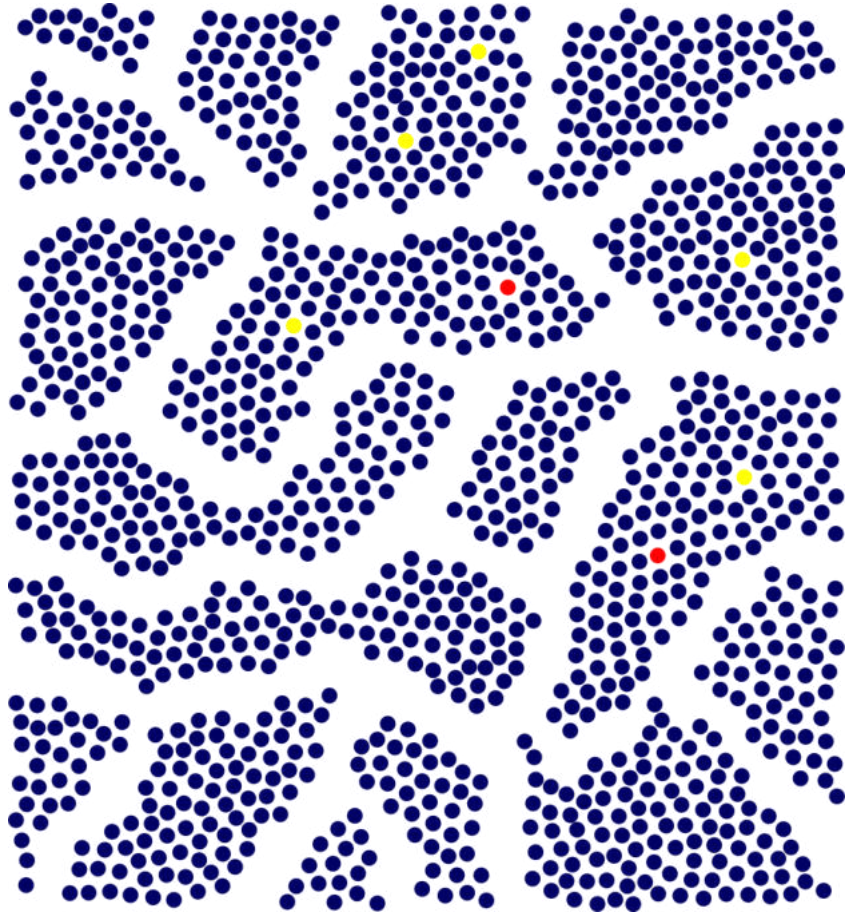


VYCOR process:

- heat treatment of NBS glass

→ spinodal phase separation

→ generation of sodium-rich borate phase and silicate phase

- extract sodium-rich borate phase by organic/anorganic acids
- silicate phase remains as stable, porous and glassy framework

H. P. Hood et al., US2106744A (1938) and US2221709A (1940)

Controlled extracted sodium borosilicate (NBS) glass

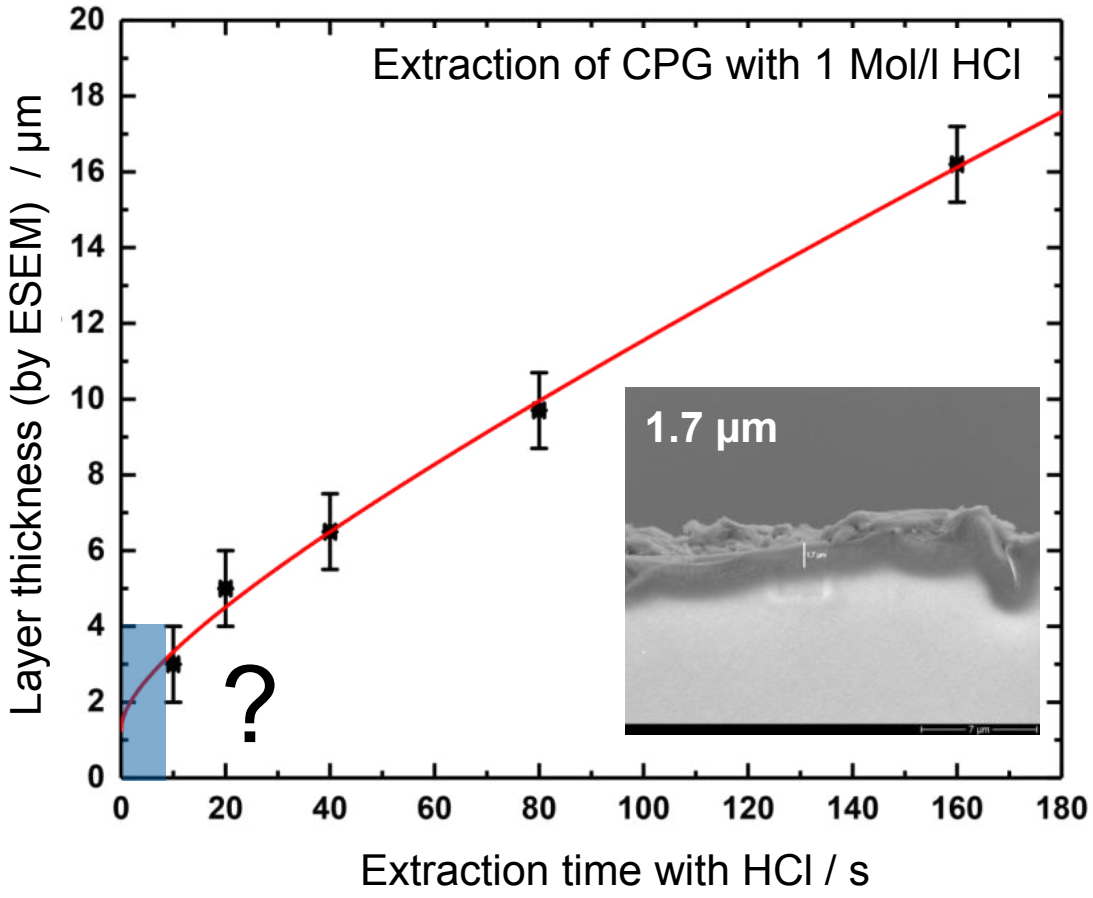
Porous layers on NBS glass

Generation of thin layers:

- control layer thickness by time of acid extraction
- Douglas *et al.*: linear and square-root dependence of the layer thickness on extraction time

Open questions:

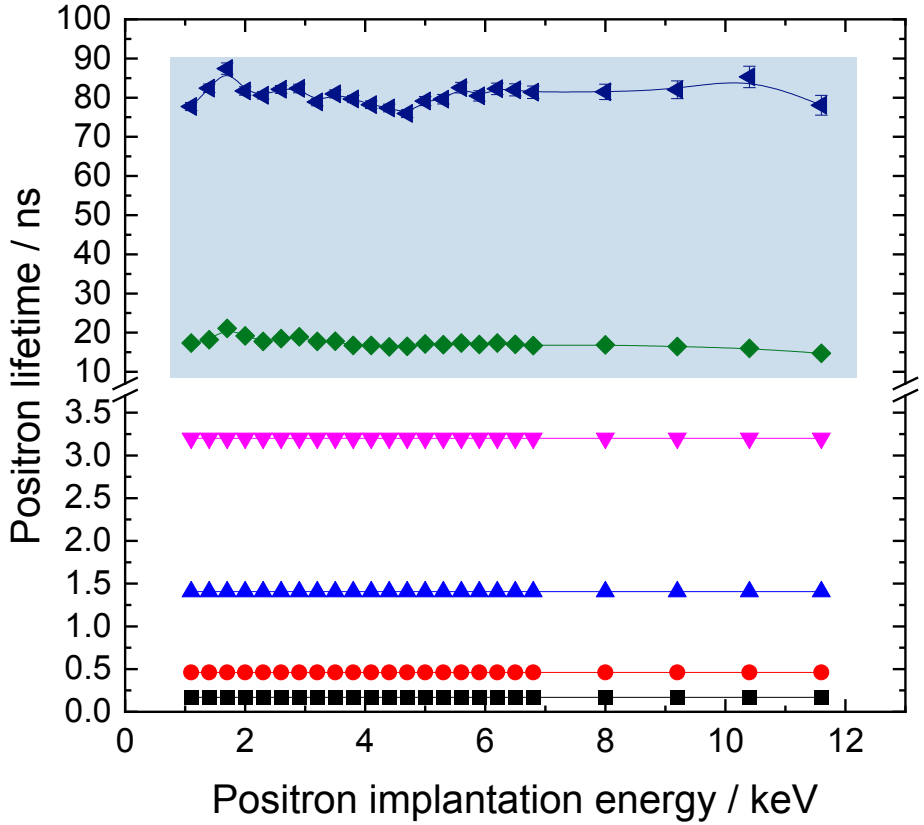
1. How to prepare layer of just a few 100 nm thickness?
2. Is it possible to determine thicknesses of rough and porous layers with mono-energetic PALS?
3. Is the model of Douglas still valid for very thin layers?



R. Douglas et al. / Journal of the American Ceramic Society 50/1 (1967)

Controlled extracted sodium borosilicate (NBS) glass

PALS measurements of NBS glass at MePS



meso-pores

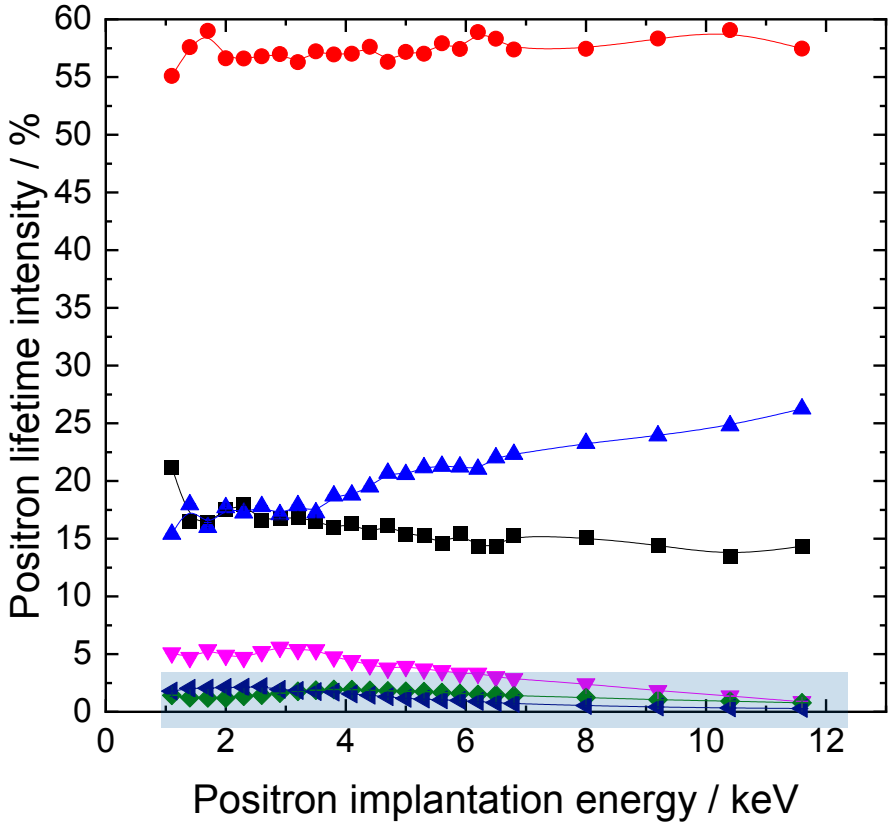
micro-pores

amorphous glass structure

free volume of glass

matrix/lattice material

bulk with p-Ps



- exemplary NBS glass sample (4 s extracted)

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Controlled extracted sodium borosilicate (NBS) glass

Thickness determination by Vehanen fit

- Makhovian implantation profile:

$$P(z,E) = \frac{m\, z^{m-1}}{z_0[E]^m} \mathrm{Exp}\left[-\left(\frac{z}{z_0[E]}\right)^m\right]$$

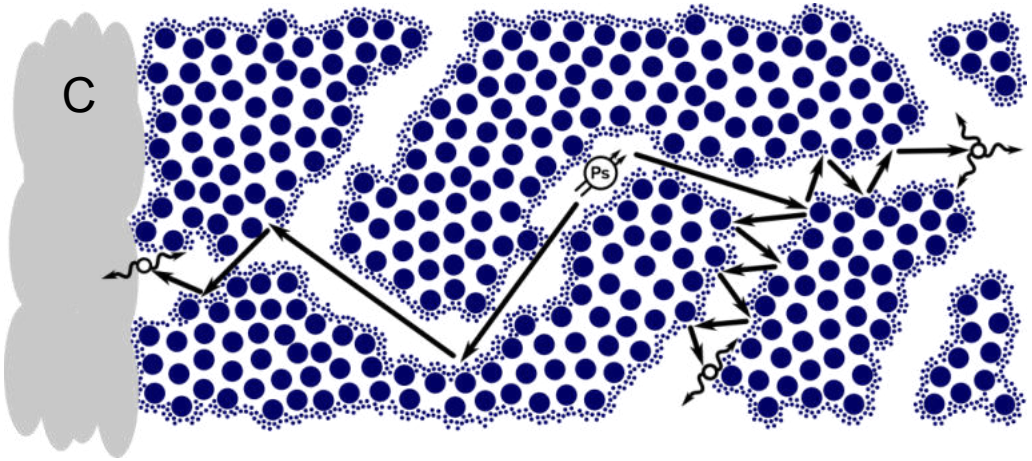
with $z_0(E) = \frac{A\, E^n}{\rho\, \Gamma\left[1+\frac{1}{m}\right]}$ and $\overline{z} = \frac{AE^n}{\rho}$

- Vehanen: **layered** structures:

$$I(z,E) = \sum_{i=1}^{n} I_i(z,E)\omega$$

$$I_C(z,E) = \int_0^d P_C(z,E)\, dz$$

- global fit of parameters A, m and n
- good agreement with *GEANT4* simulation by Dryzek *et al.*

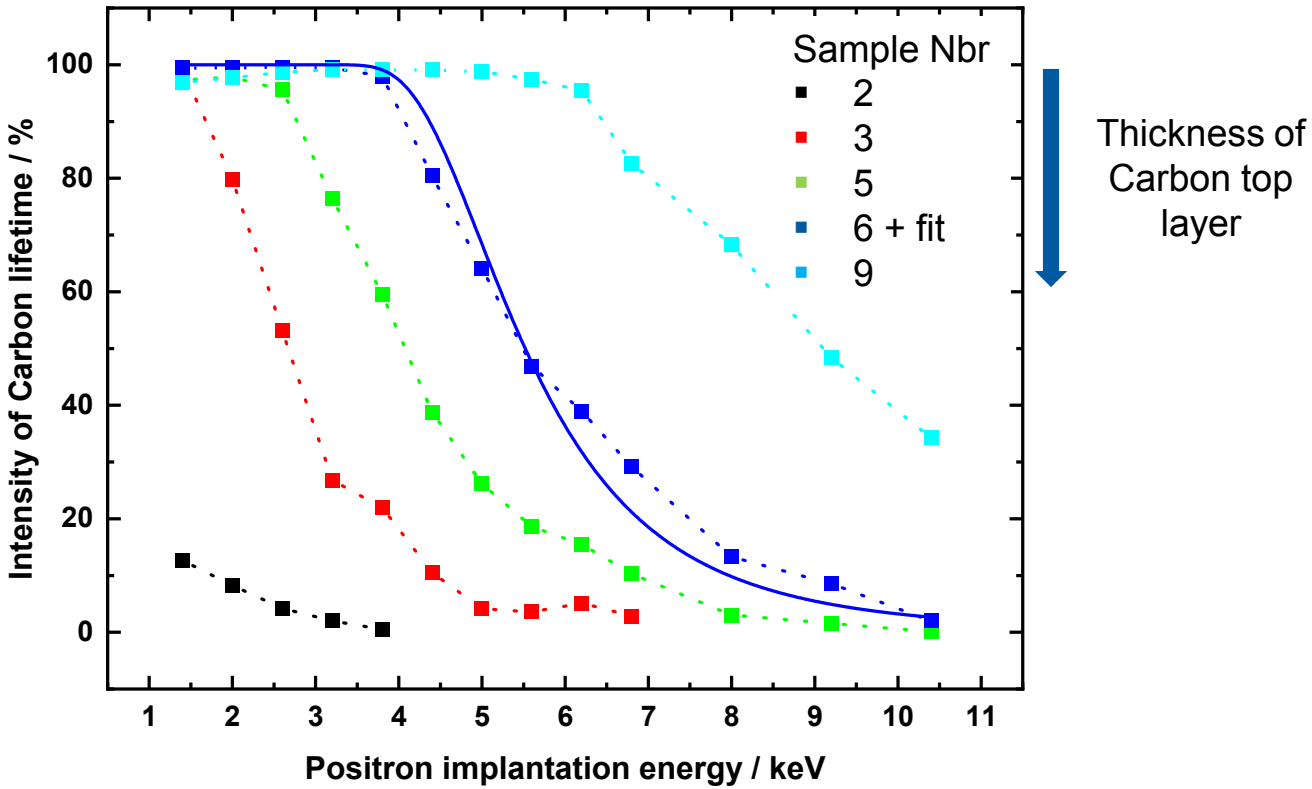


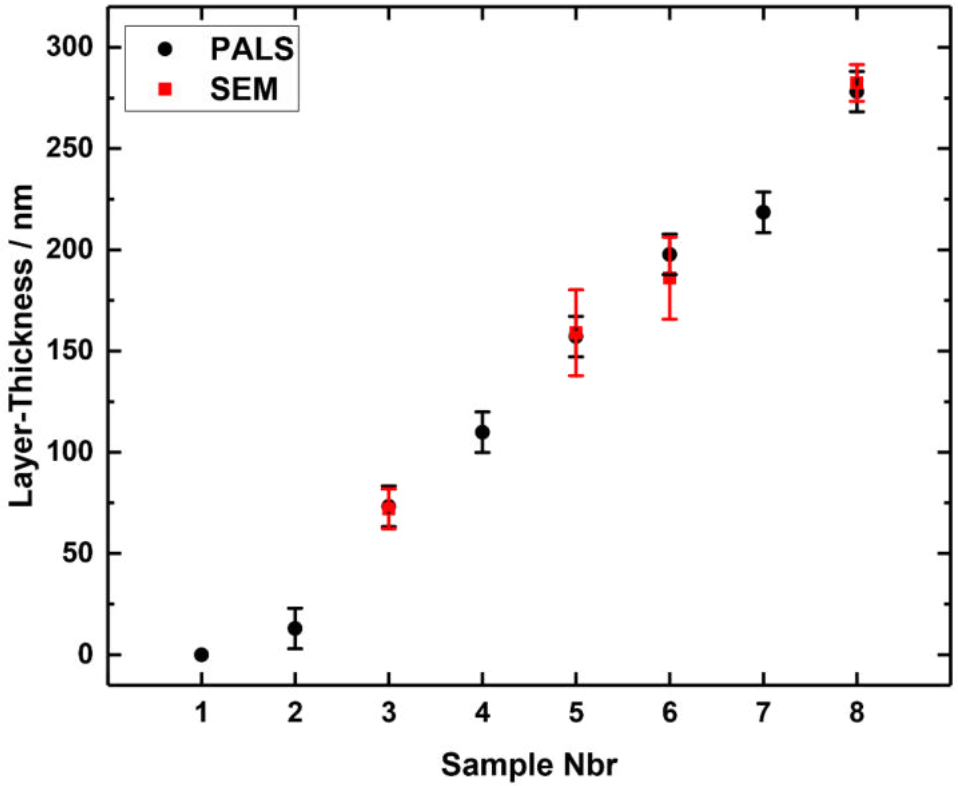
- CPG with different thick Carbon top layers
- evaluation of Vehanen model for rough and porous structures

A. Vehanen et al / Physical Review B 35 (1987) 4606
J. Dryzek *et al.*, NIM B 266,18 (2008) 4000-4009

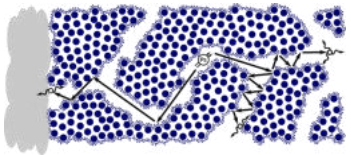
Controlled extracted sodium borosilicate (NBS) glass

Thickness determination by Vehanen fit - Verification on NBS glass with different Carbon top layer





- Vehanen fit works for porous structures
- good agreement between PALS and SEM



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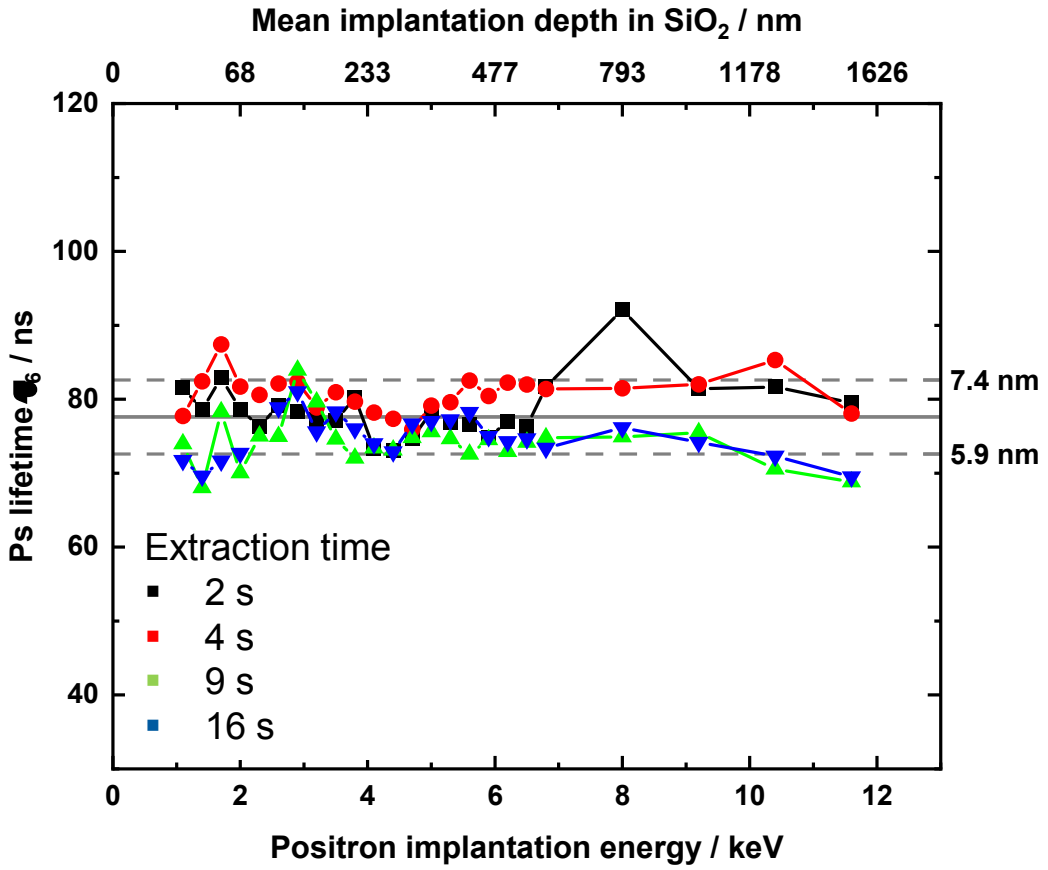
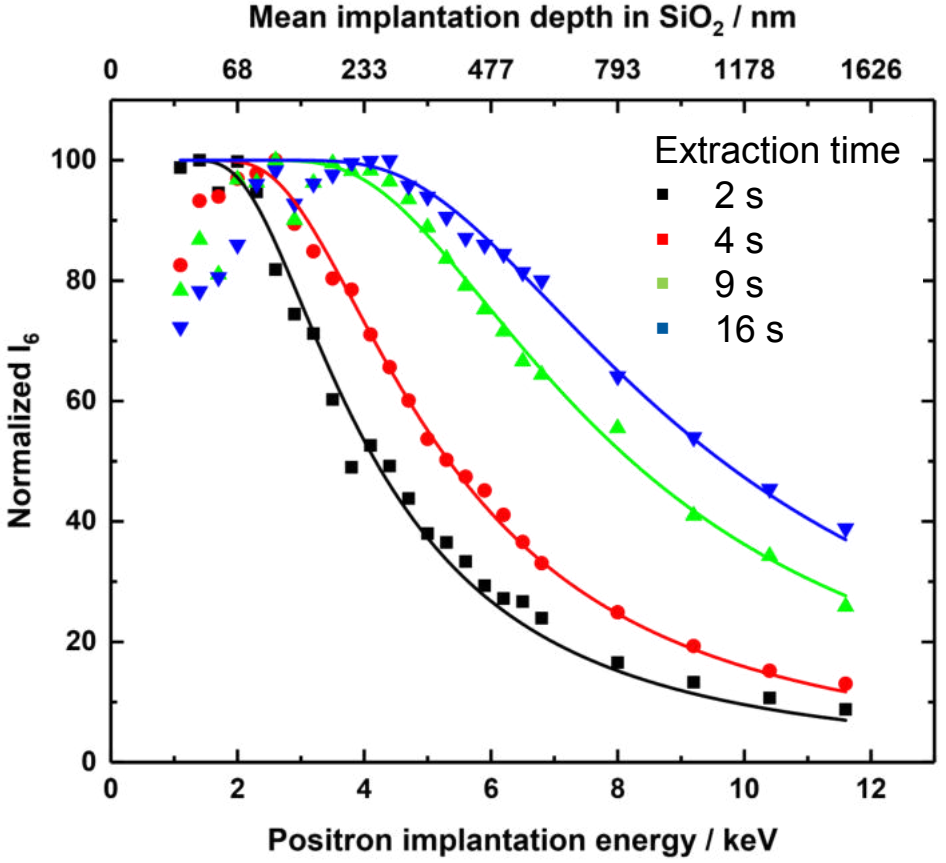
Controlled extracted sodium borosilicate (NBS) glass

Thickness determination by Vehanen fit - NBS glass with different extraction times





- pore size independent from extraction duration
- Vehanen fit of Ps component intensity

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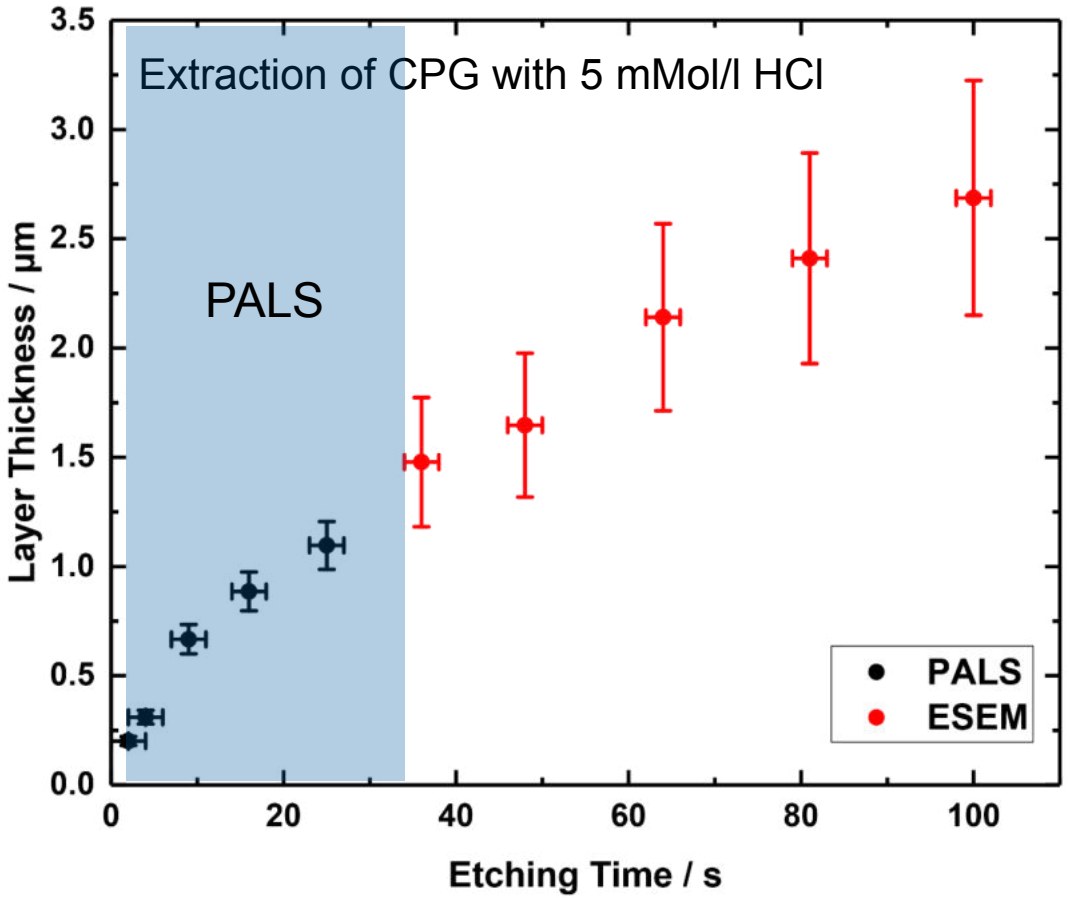
Controlled extracted sodium borosilicate (NBS) glass

Thin porous layers on NBS glass

1. How to prepare layer of just a few 100 nm thickness?
   - ➢ use very weak acid (5 mMol/l instead of 1 Mol/l HCl)
   - ➢ extract for only a few seconds

2. Is it possible to determine layer thicknesses of rough and porous structures with mono-energetic PALS?
   - ➢ PALS plus Vehanen fit is an easy tool for the determination of layer thicknesses even for rough, porous and insulating materials

3. Is the model of Douglas still valid for very thin layers?
   - ➢ square-root behaviour even for very short extraction times



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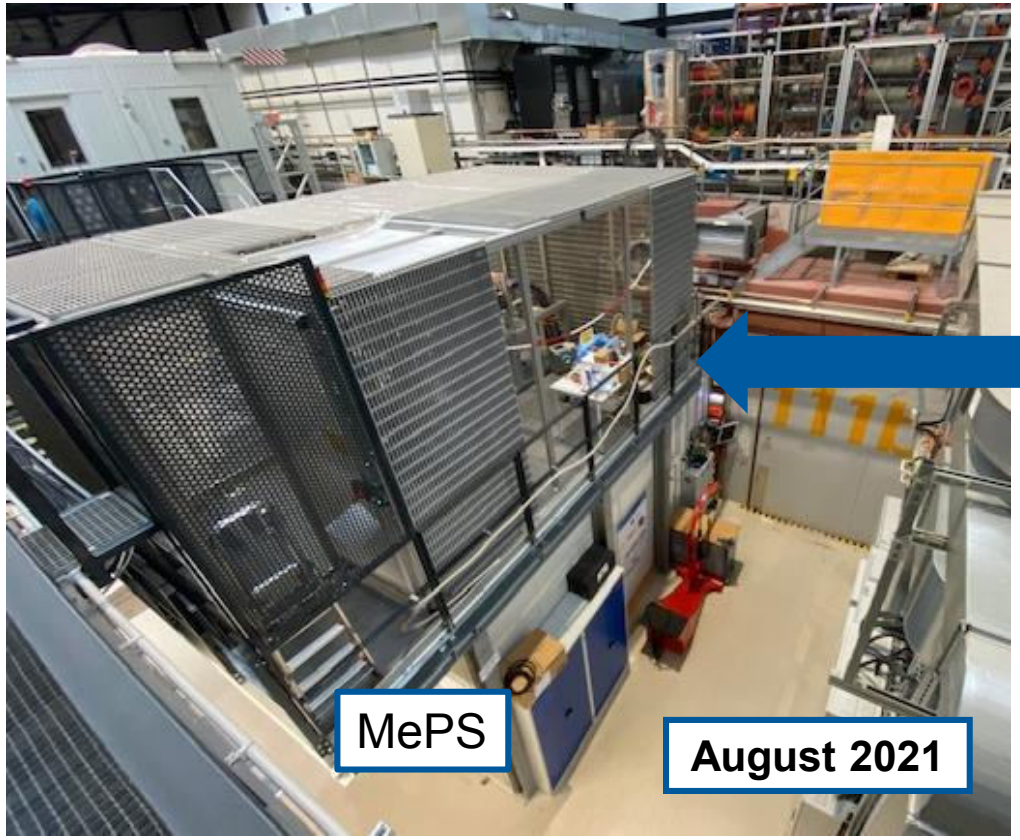
Outlook & Summary

Our second beam line



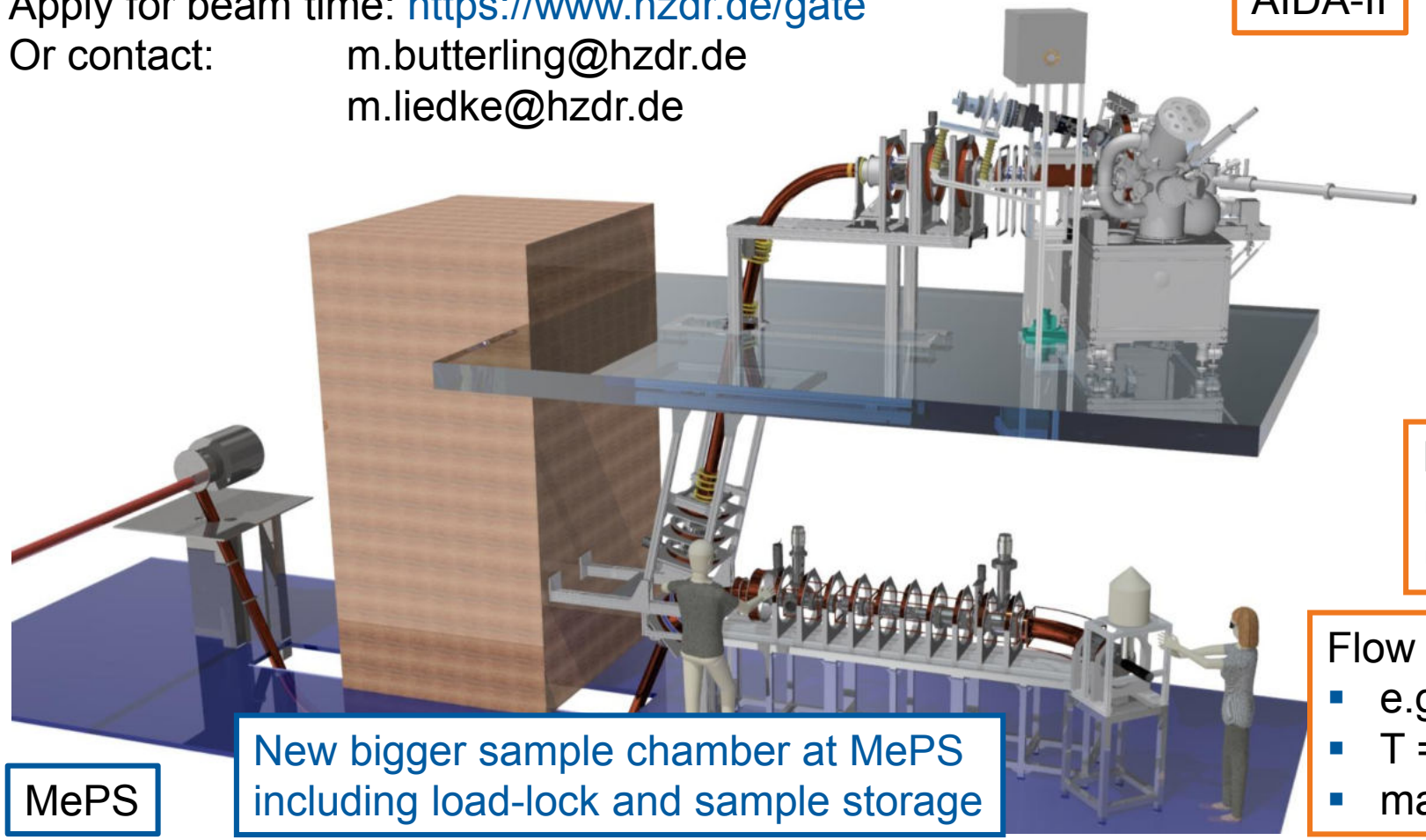
two beam lines on top of each other

What is hidden in that cage on top of the MePS beam line?

Outlook & Summary

Apparatus for in-situ Defect Analysis - PALS & More

Apply for beam time: https://www.hzdr.de/gate
Or contact: m.butterling@hzdr.de
m.liedke@hzdr.de



MePS

New bigger sample chamber at MePS including load-lock and sample storage

AIDA-II

Sample cooling/heating
- T = 50...1200 K

4-point probe resistance

Ion source
- noble gases, N, O, H
- I ≤ 20 mA
- E = 2 … 30 keV

Deposition:
- Fe, Co, Ni, Au, Ag, Cr, Pt, Mo
- rate: 0.1 - 0.5 Å/s

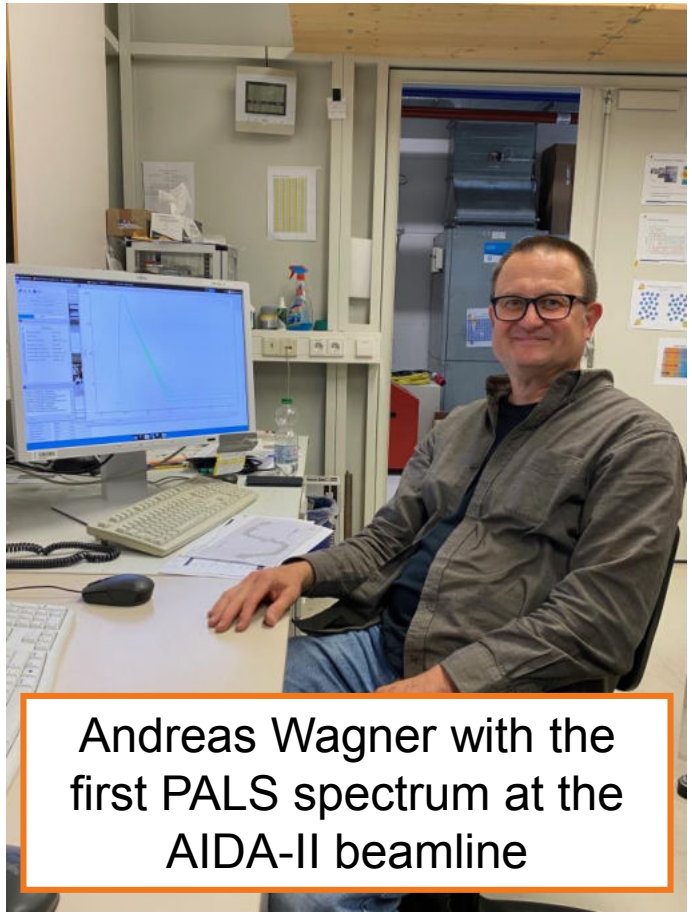
Flow Through High Pressure Reactor
- e.g. for H_2 gas loading
- T = 77 … 923 K
- maximum pressure: 20 bar

Outlook & Summary

Apparatus for in-situ Defect Analysis - PALS & More

28.08.2021: Comissioning of PALS beam line

AIDA-II at the EPOS beamline 29.08.2021



Andreas Wagner with the first PALS spectrum at the AIDA-II beamline

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Outlook & Summary

EPOS - a long journey from vision (2002) to reality - **Thank you Reinhard Krause-Rehberg**

- EPOS will be the combination of a positron lifetime spectrometer, Doppler coincidence, and AMOC
- high quality data for optimum defect characterization
- Main features:
  - high-intensity bunched positron beam
  - fast lifetime mode (single detector mode)
  - repetition time 77ns (13 MHz), but also longer for positronium studies (lifetime > 100ns)
- EPOS lab area is restricted to 25 m2 , thus only one beam line is planned

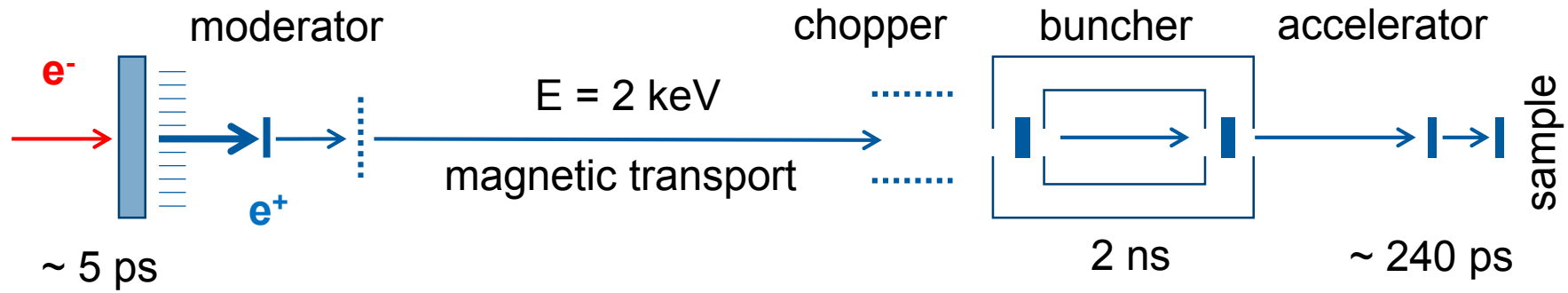
EPOS user meeting 2002 - Reinhards vision

- MePS + GiPS
- SC LINAC ELBE + improved beam alignment + digital PALS
- Timing resolution ~ 230 ps
- 120 kcps in PALS (10^6 counts in 80 s)
- PALS for thickness determination of thin, porous, rough NBS glass layers
- AIDA-II on rooftop sucessfully comissioned

Thank you for your attention

Positron Annihilation Spectroscopy at the HZDR

Accelerator-based positron facilities



MePS

Mono-**e**nergetic
Positron **S**ource

GiPS

Gamma-**i**nduced
Positron **S**ource

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DRESDEN concept
HZDR