Positrons Probing Matter:

Novel Applications Using Low-Energy Positron Beams

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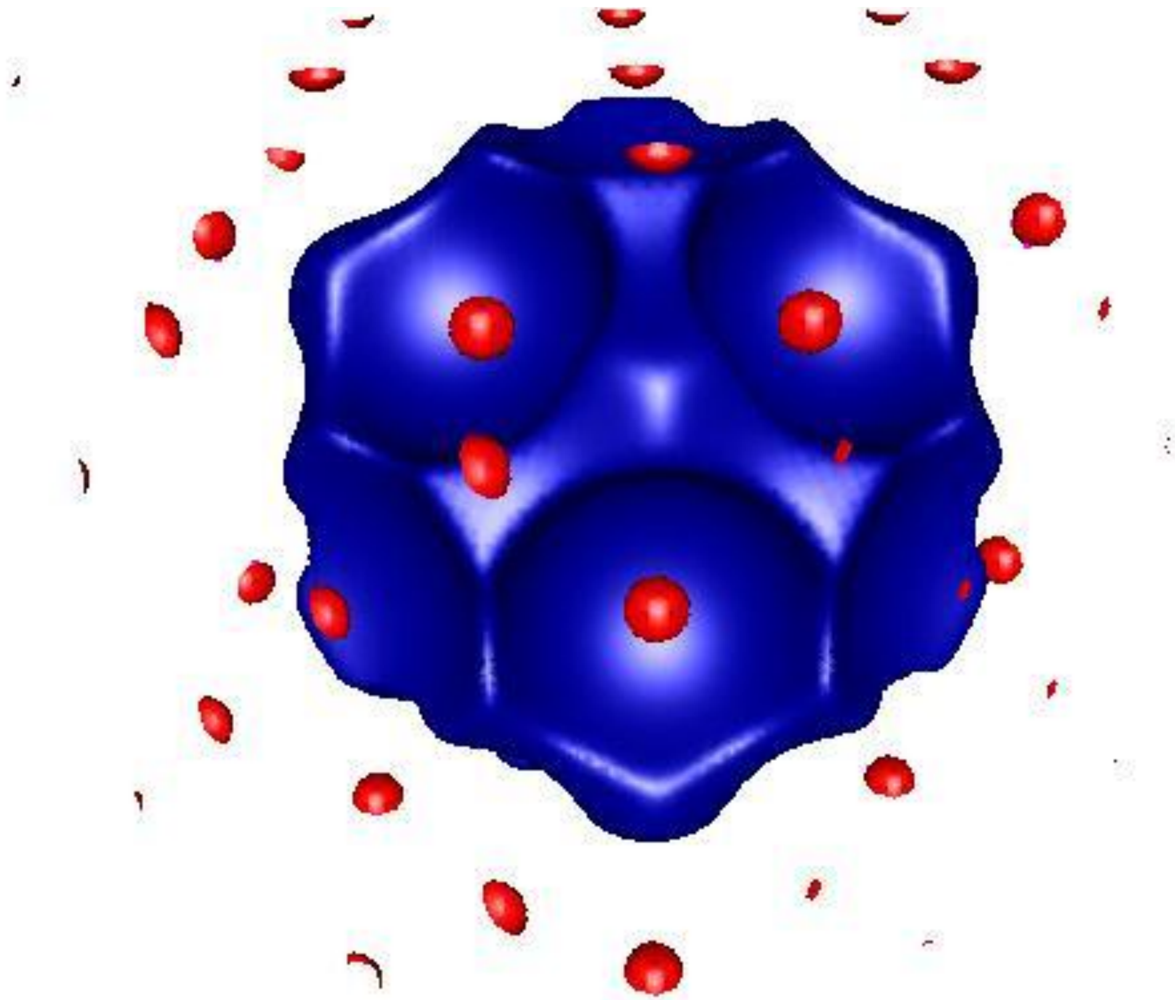
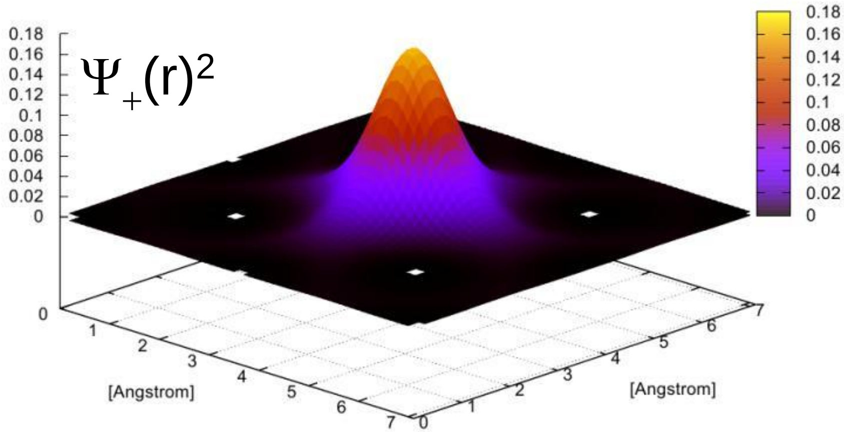
Physik mit Positronen

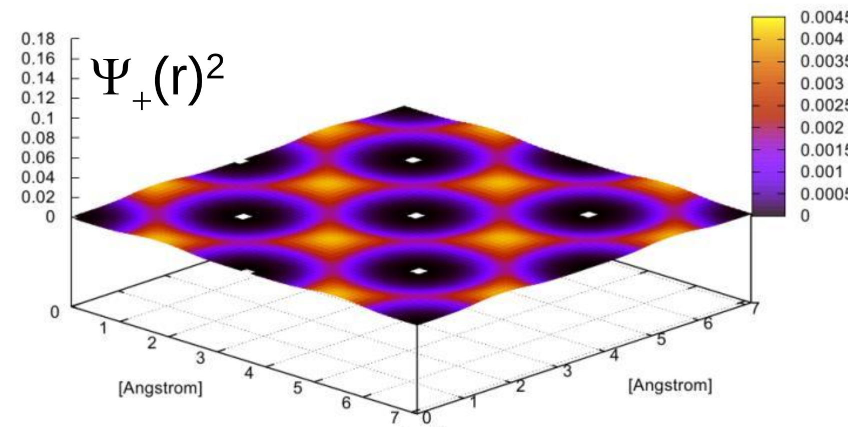


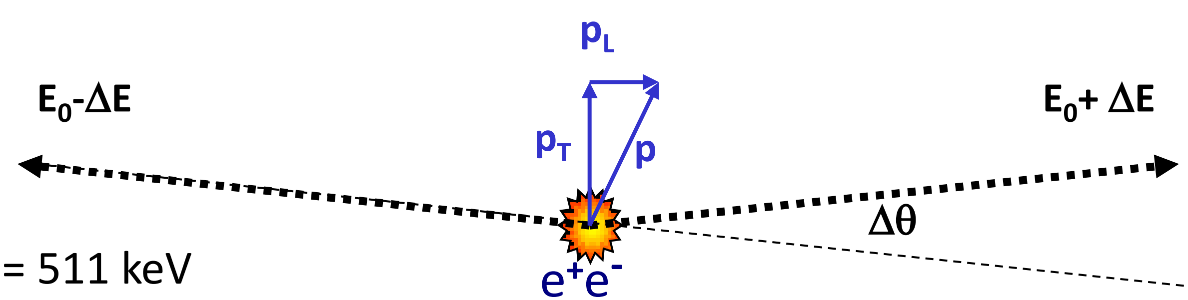
Positron Trapping in Vacancies

Mono-vacancy in Cu



Ideal Cu lattice





$E_0 = m_0c^2 = 511$ keV

Doppler-shift: $\Delta E = 1/2\ p_L c$

Angular deviation: $\Delta\Theta \approx p_T / m_0 c$

Positron Group at NEPOMUC

MLZ Heinz Maier-Leibnitz Zentrum

NEPOMUC

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DAAD

RISE

Japan Society for the Promotion of Science 日本学術振興会

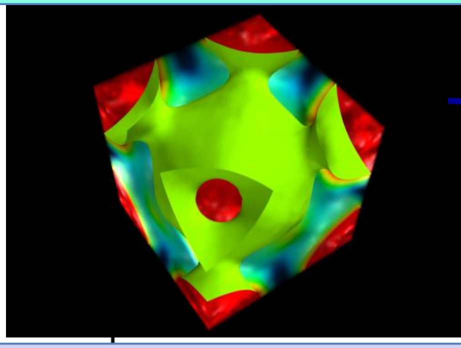
Universität Bayern e.V.

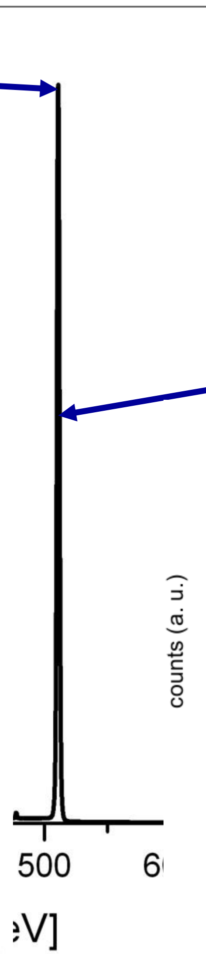
BaCaTeC Bavaria California Technology Center

GEFÖRDERT VOM Bundesministerium für Bildung und Forschung

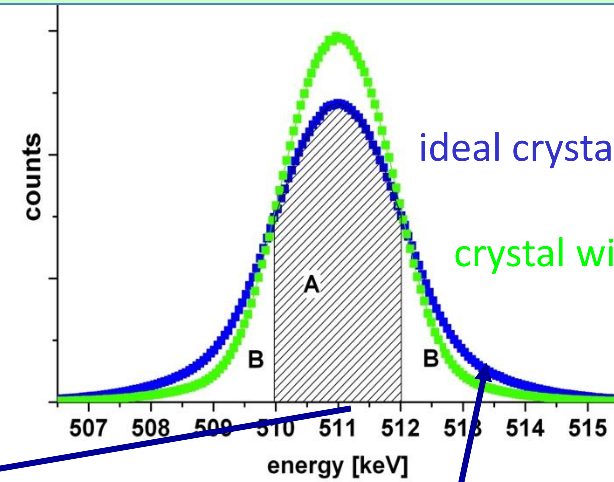
What we Measure:

Angular Correlation of Annihilation Radiation – ACAR

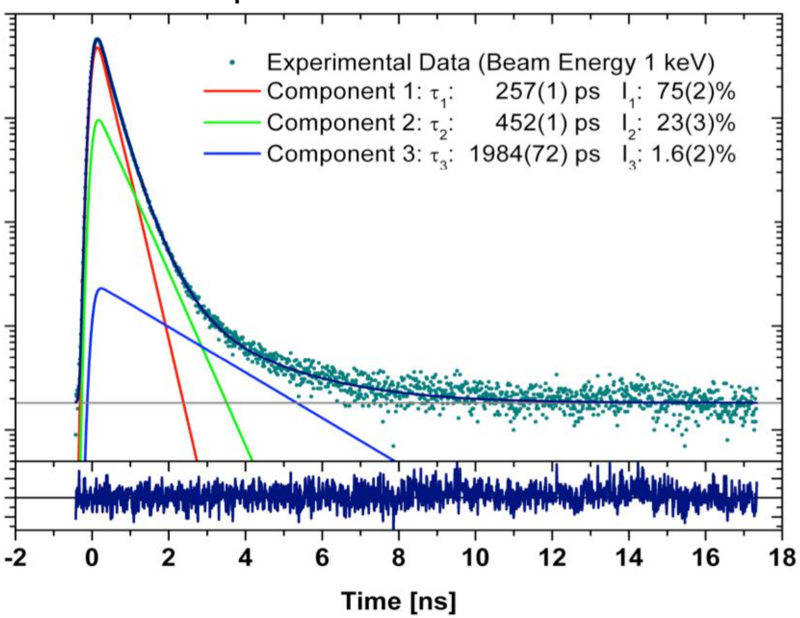


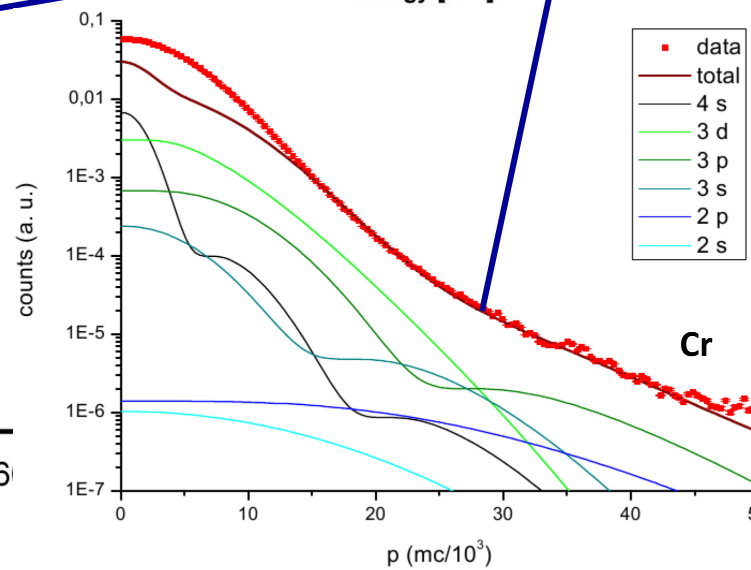


Doppler-Broadening Spectroscopy – DBS



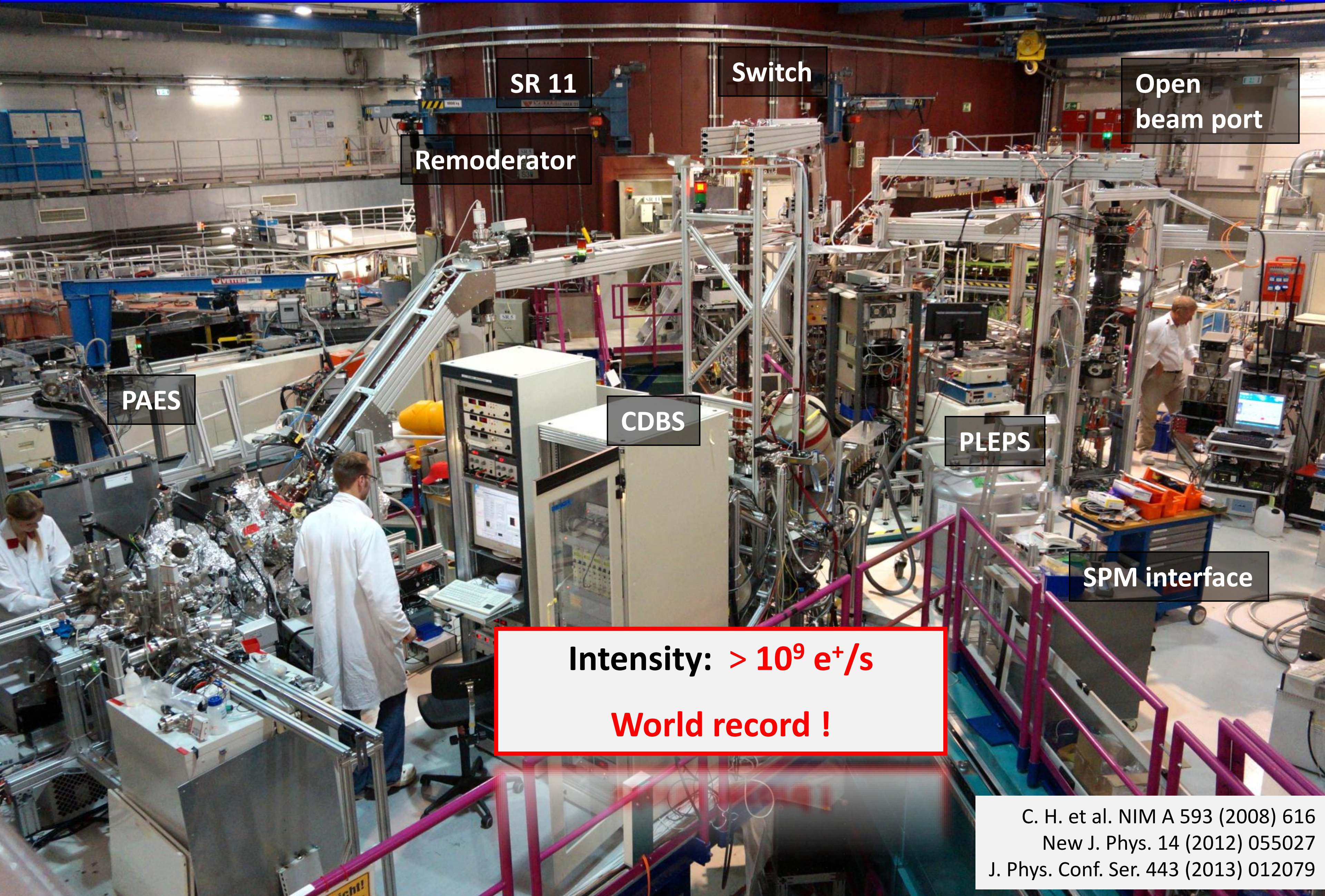
Positron Lifetime Spectroscopy – PLS





Coincident Doppler-Broadening Spectroscopy – CDBS

Positron Beam Facility at NEPOMUC

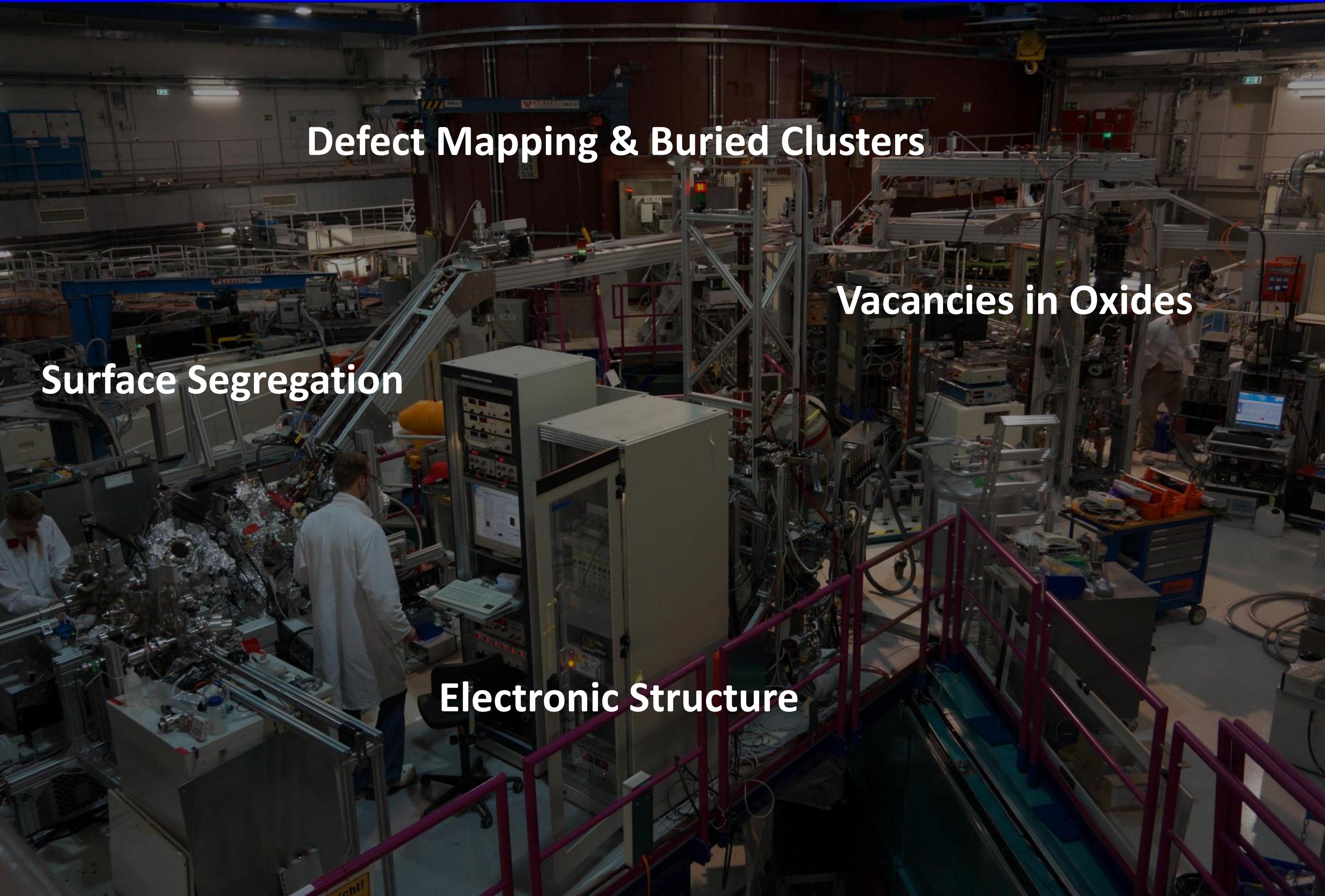


Intensity: > 10^9 e^+/s

World record !

C. H. et al. NIM A 593 (2008) 616
New J. Phys. 14 (2012) 055027
J. Phys. Conf. Ser. 443 (2013) 012079

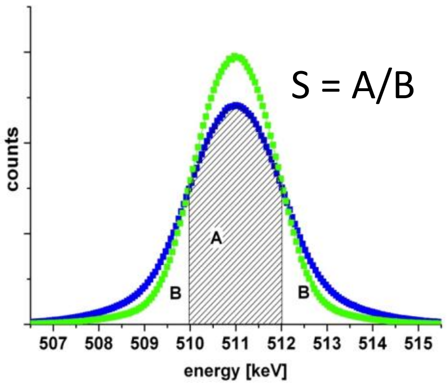
Positron Beam Experiments

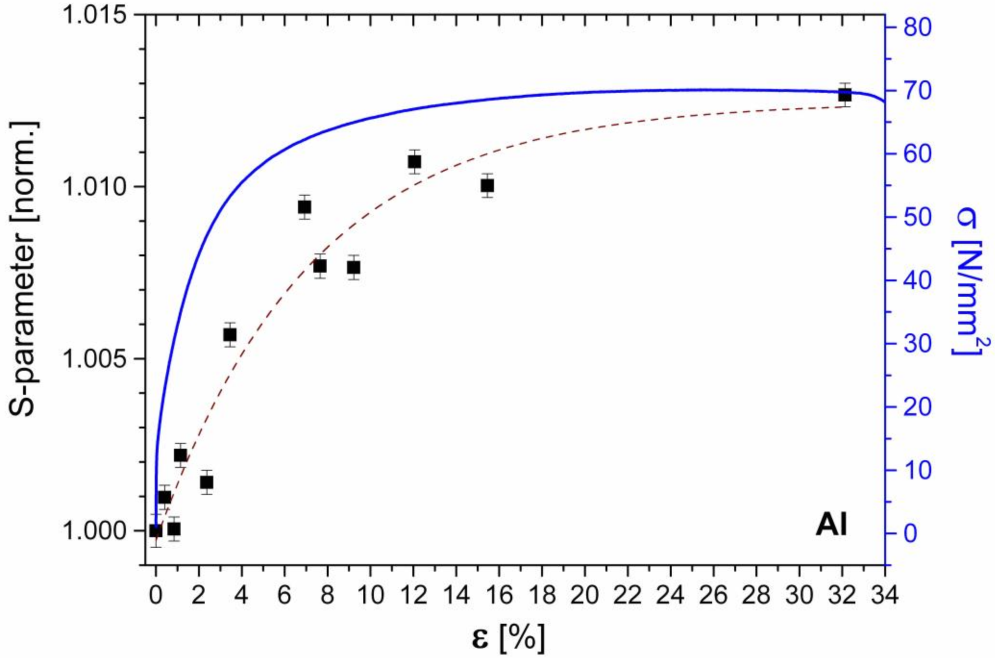


Plastic Deformation in Al and Al Alloys

Macroscopic ⟺ **Atomic scale**

- plastic deformation ⟺ dislocations, vacancy-like defects
- tensile tests → σ(ε) ⟺ DBS → **S-parameter**



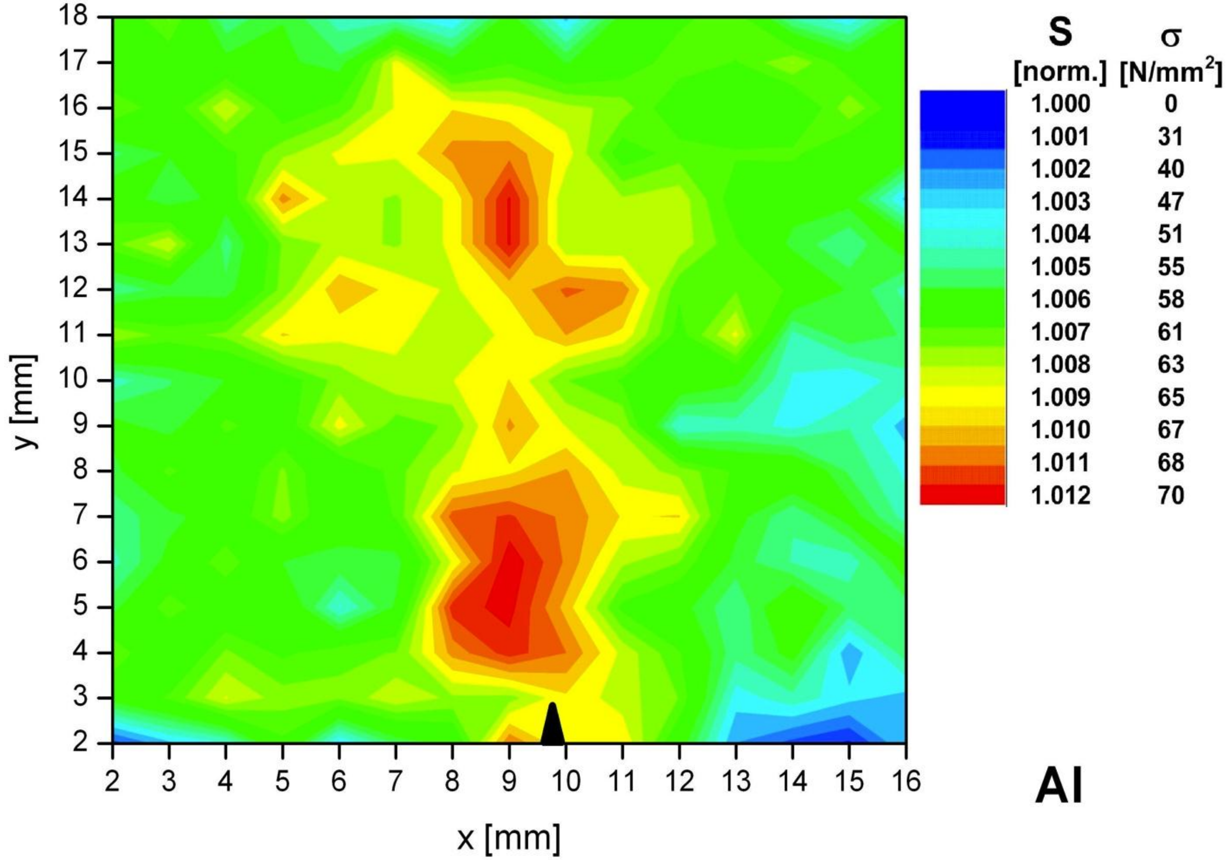


- **S(σ)** correlation almost material independent (Al, AlMgSi0.5, AlMg3)
- Correlation of S and ***locally acting σ*** by spatial resolved DBS

C. H. et al. PR B 81 (2009) 064102

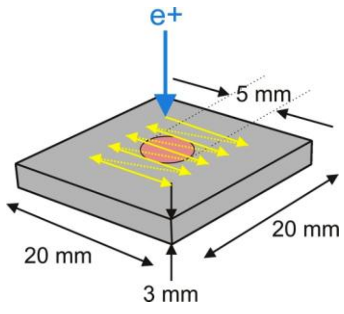
Plastic Deformation in Al and Al Alloys

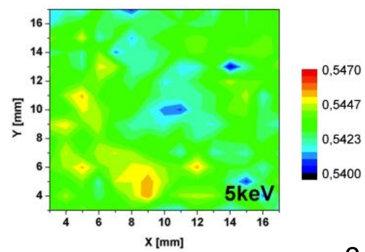
Aim: 2D defect mapping + visualization of local σ in asymmetrically shaped samples



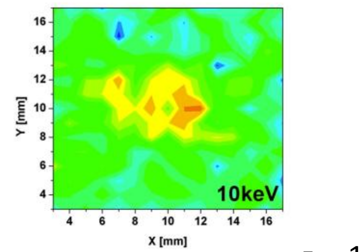
C. H. et al. PR B 81 (2009) 064102

Irradiation Induced Defects

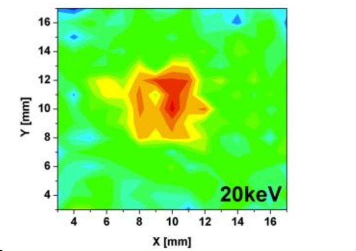




z_m=0.3µm



z_m=1.0µm

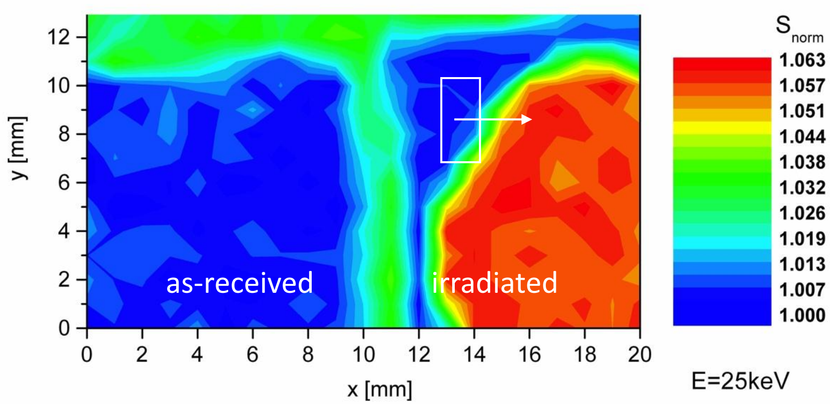


z_m=3.2µm

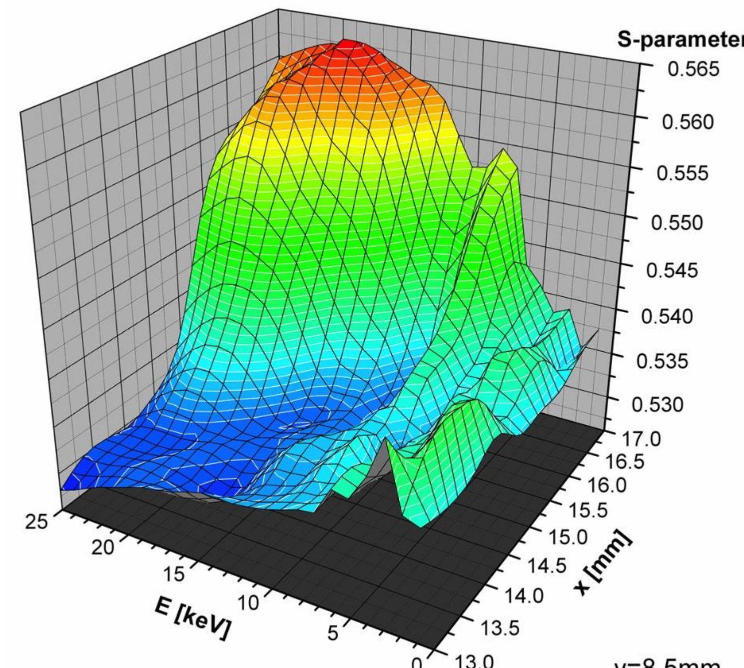
Simulation of fission fragments induced defects:

Zr^+ irradiated Zircaloy: 3 MeV, 2.5 10^{13} Zr^+/cm^2

Samples: R. Hengstler, AREVA NP GmbH



E=25keV



y=8.5mm

- Spatially resolved defect profile due to vacancy clusters

M. Stadlbauer, C. H. et al. PRB 76 (2007) 174104

R.M. Hengstler-Eger, C.H. et al. J. Nucl. Mat. 423 (2012) 170

Element Information

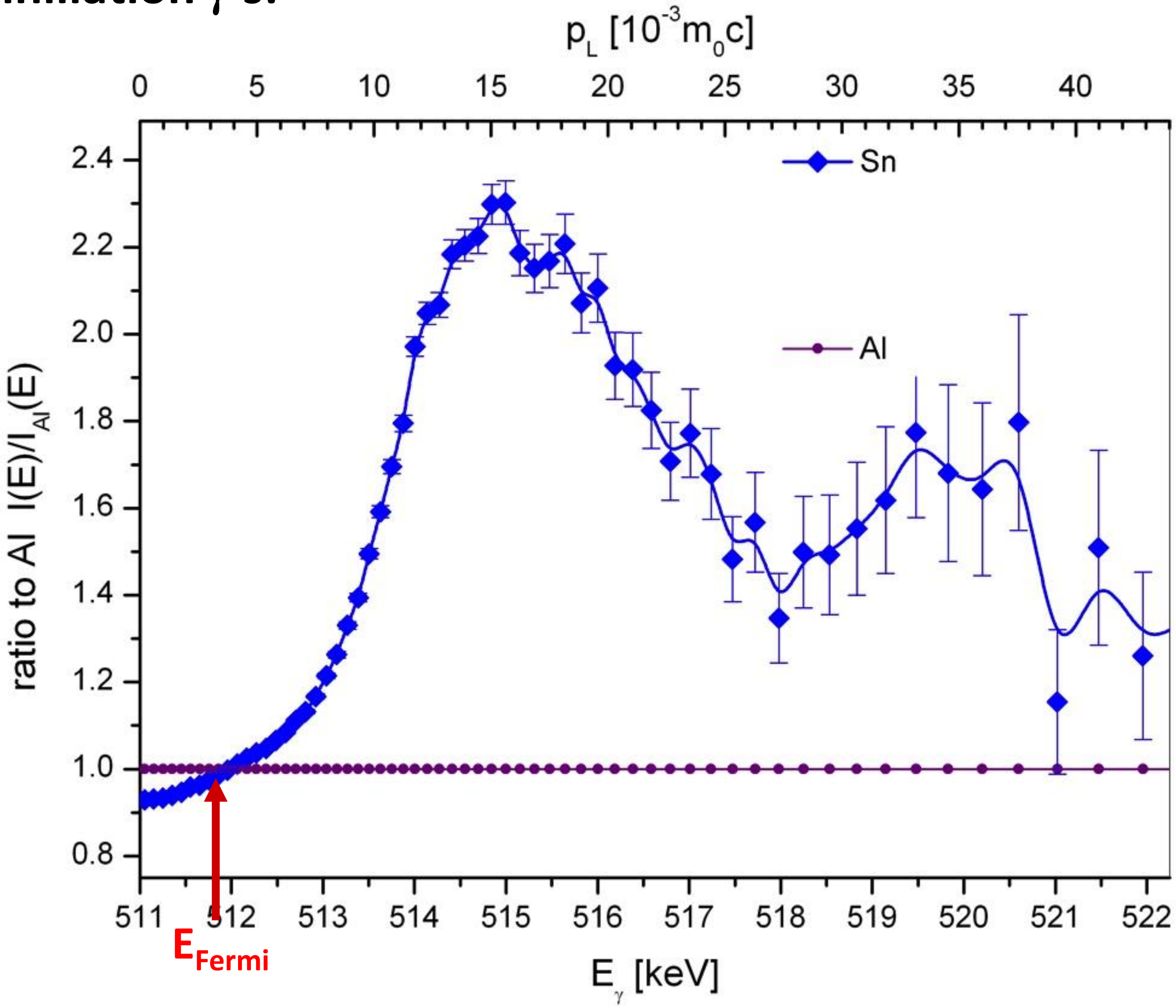
Coincident detection of annihiliation γ‘s:

(CDBS ratio spectra)

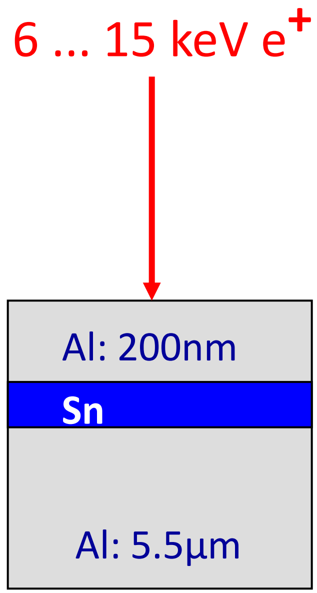
Questions:

- What is the elemental **sensitivity**?
- What about the elemental **selectivity**?

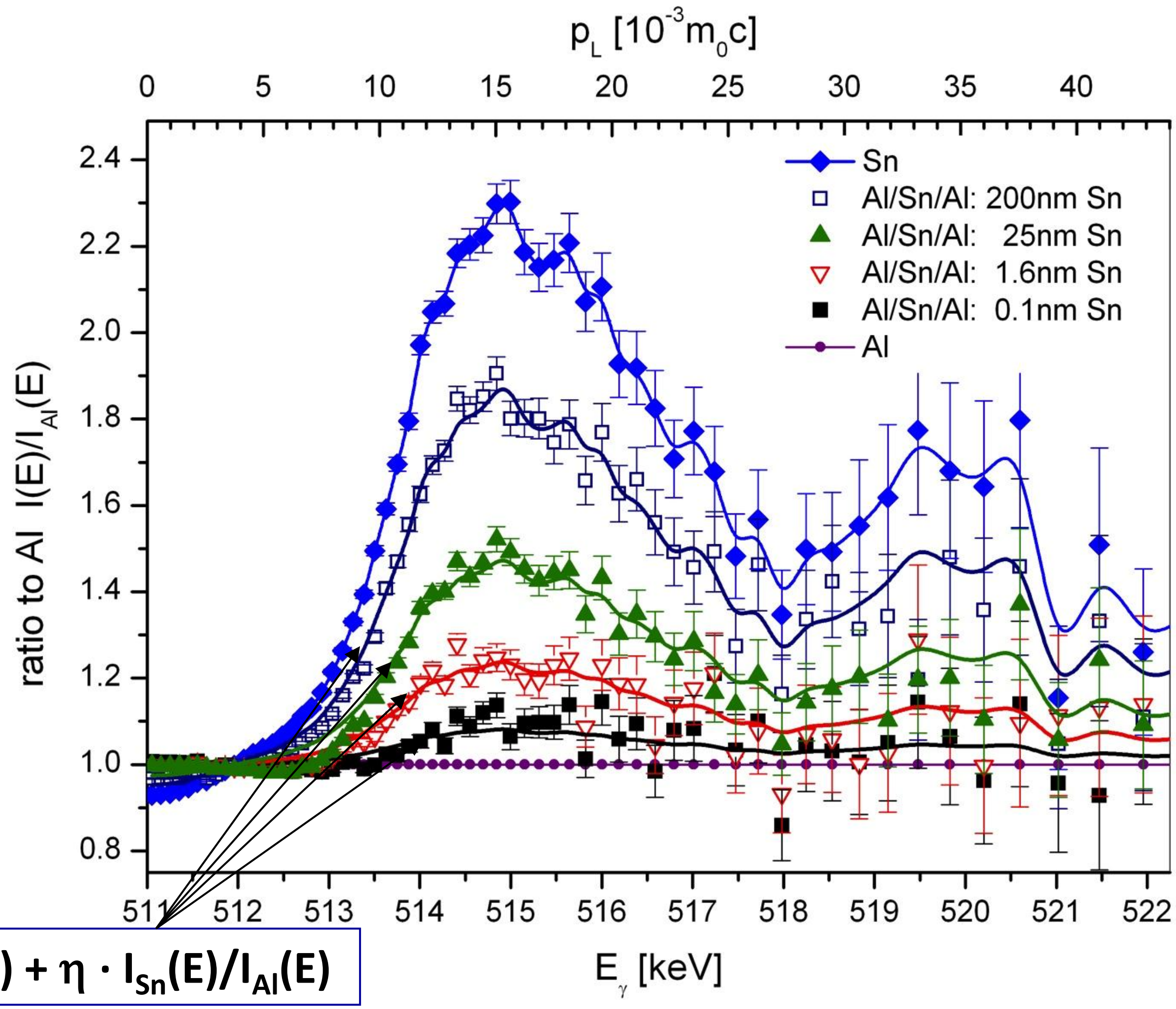
→ Can we see **buried Sn layers/clusters** with CDBS?



Buried Layers: Al/Sn/Al



Samples:
d_{Sn}=200, 25, 1.6, 0.1 nm

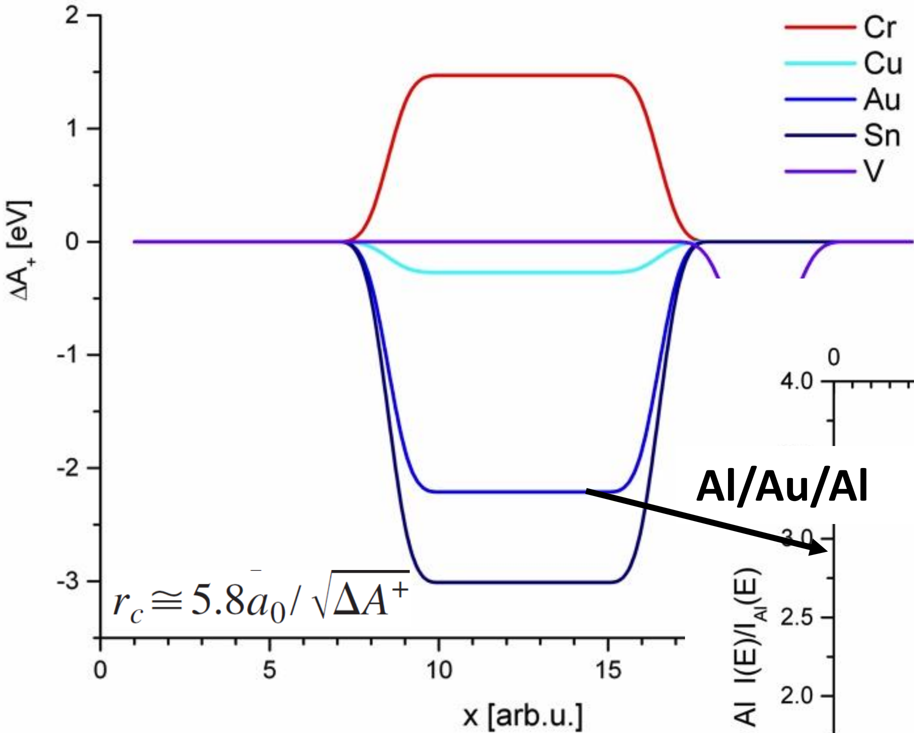


$$I_{ratio}(E) = (1 - \eta) + \eta \cdot I_{Sn}(E)/I_{Al}(E)$$

C. H. et al. PRB 77 (2008) 092105

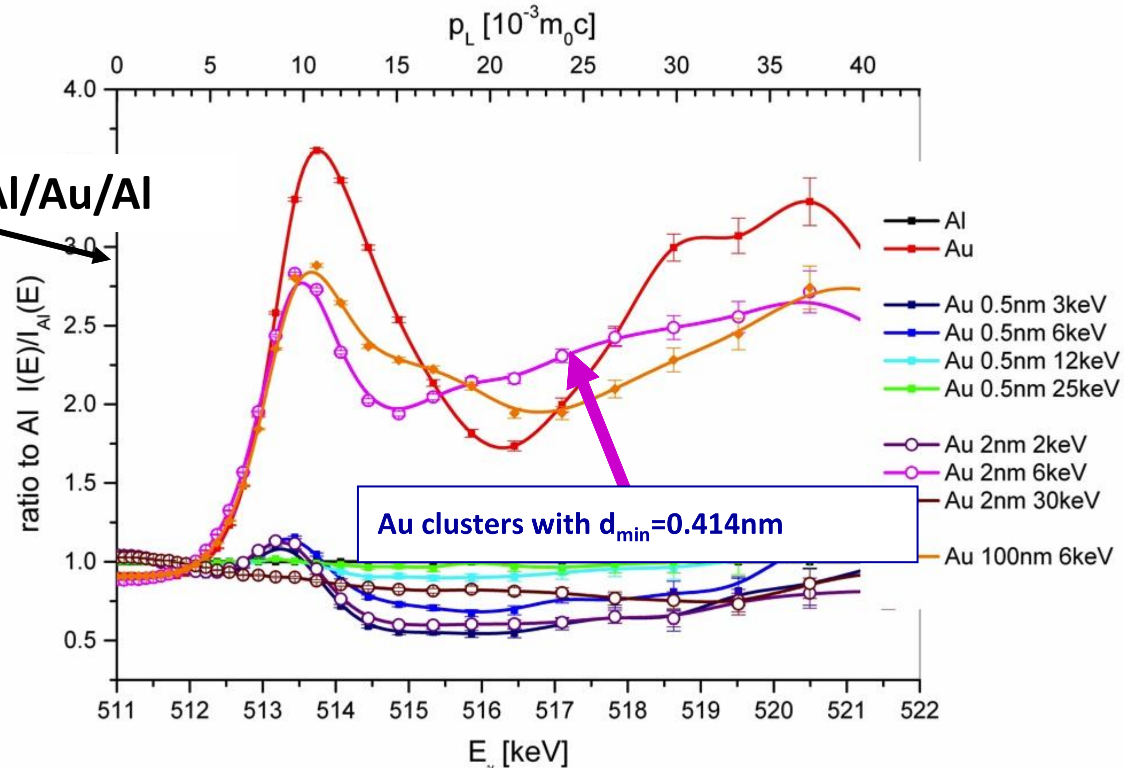
Variation of A^+ at Interface in Al/M/Al

Embedded layers in Al:



Al/Au/Al

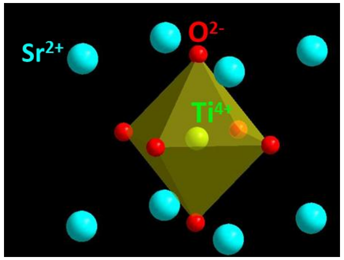
Similar result for Au layers:

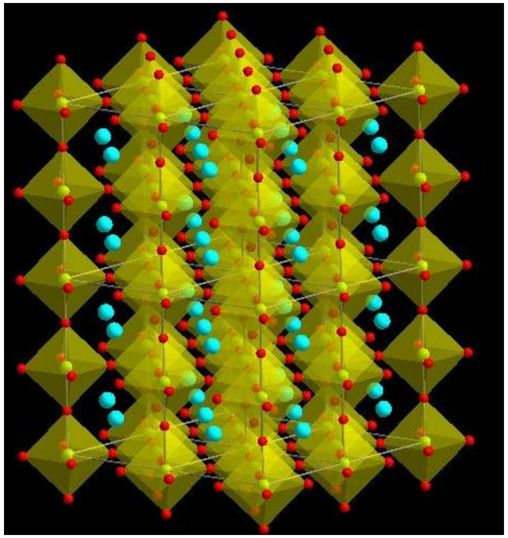


P. Pikart, C. Hugenschmidt et al. PR. B 84 (2011) 014106

Vacancies in Perovskite Oxide Thin Films

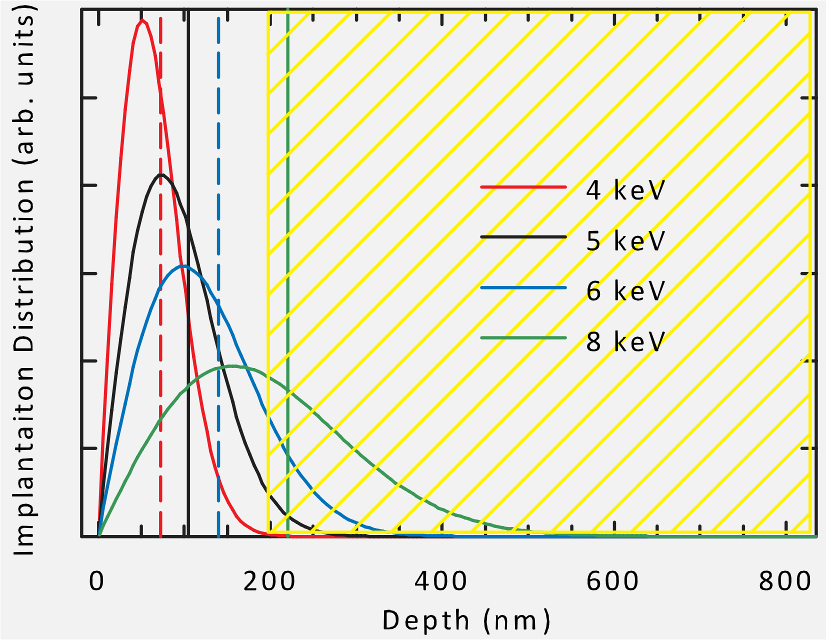
Pulsed laser deposited (PLD) homoepitaxial $SrTiO_3$





Aim:

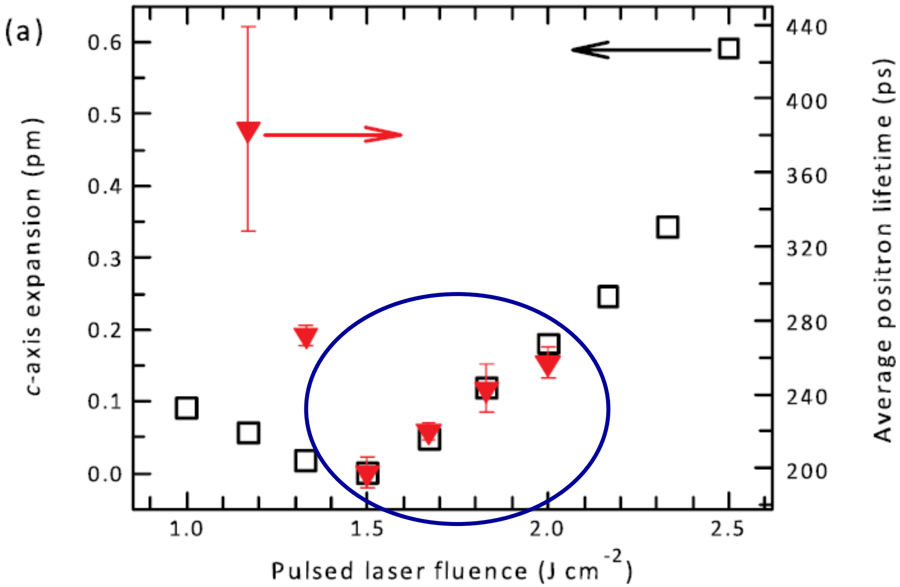
Determination of vacancy types for different process parameter



Adjustment of e+ energy to max sensitivity in 200nm layer

D. Keeble et al. PRL 105 (2010) 226102, PRB 87 (2013) 195409

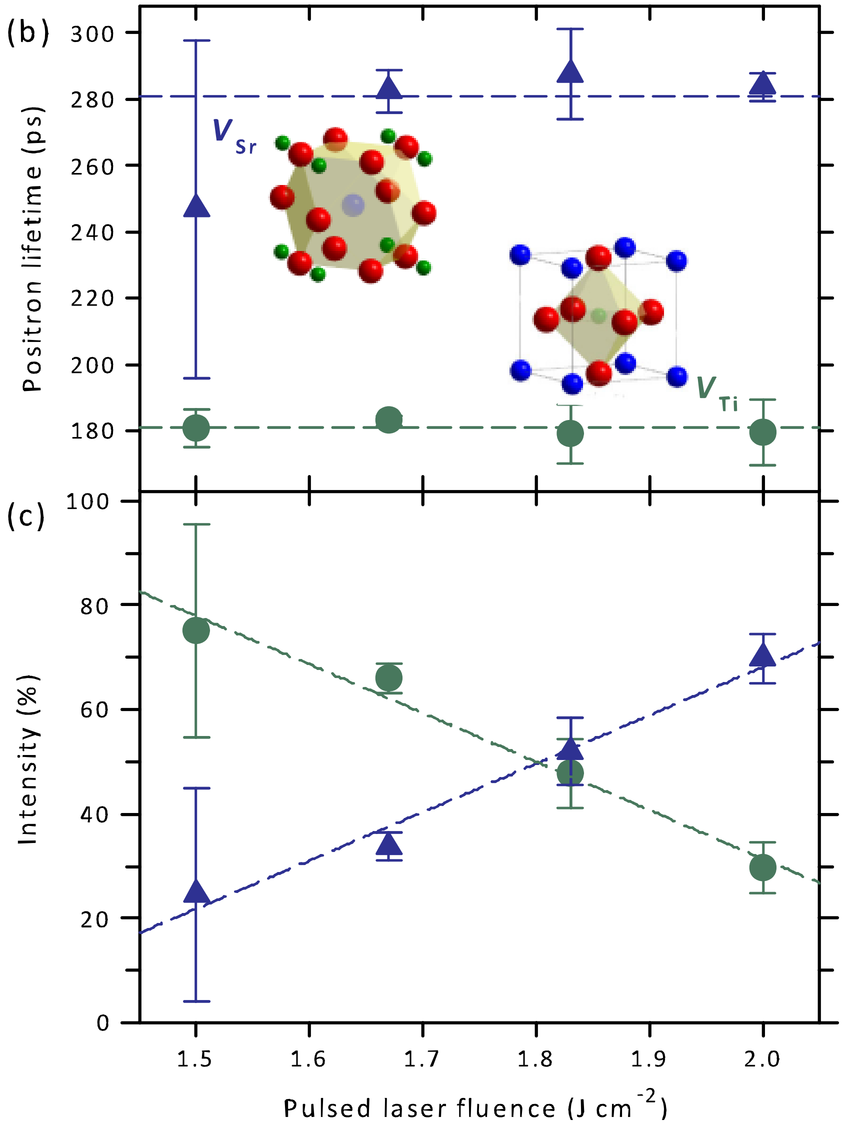
Results: $SrTiO_3$ Thin Films



→ c-axis parameter expansion (XRD) and **mean** positron lifetime

→ Vacancy **type** identified

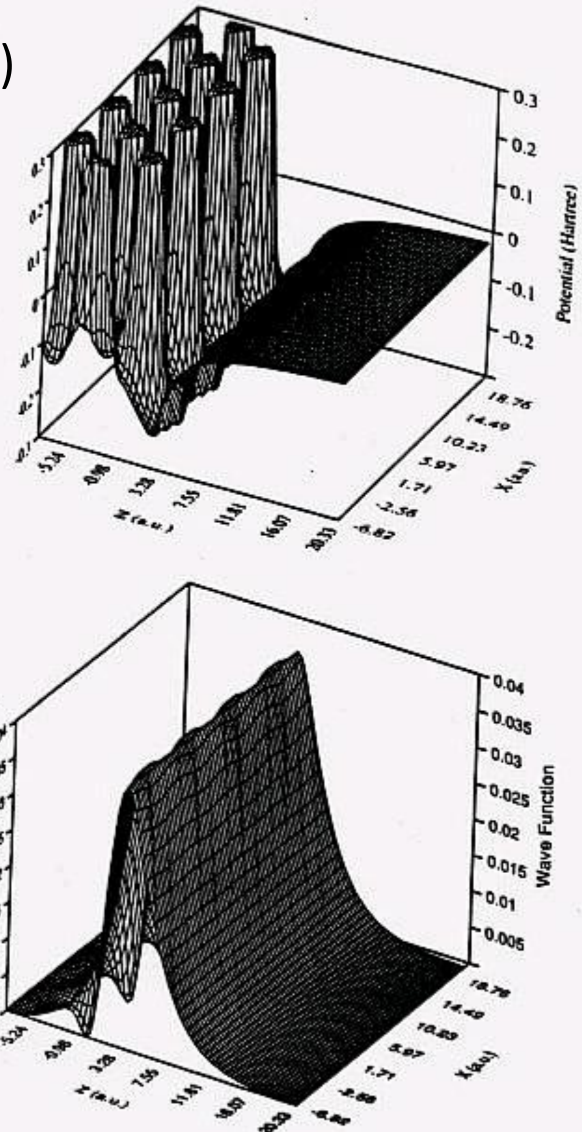
→ **Concentration** strongly depends on laser fluence



D. Keeble et al. PRL 105 (2010) 226102, PRB 87 (2013) 195409

The Positron at the Surface

Cu(100)



$\psi(r)^2_{e+}$

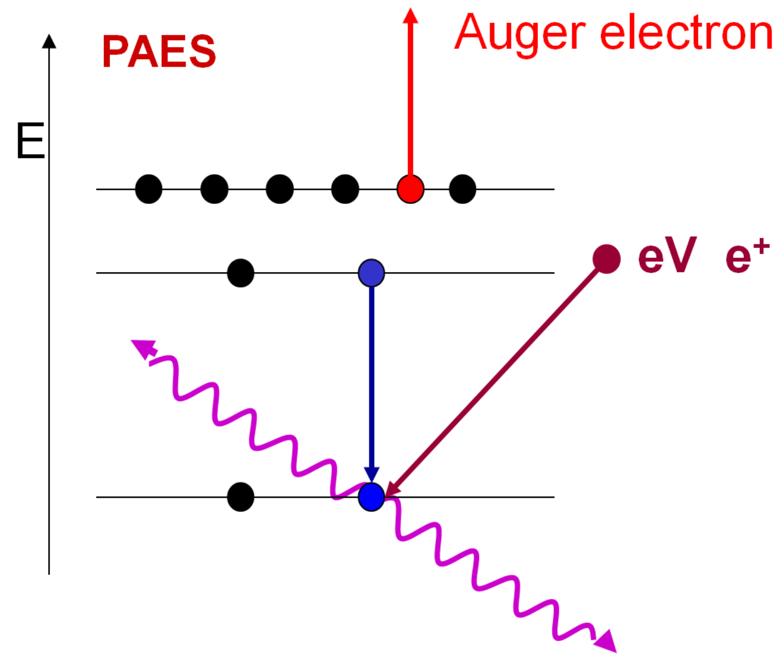
A. Weiss et al., 1992

Surface potential

→ Positron trapping

→ Annihilation at topmost layer

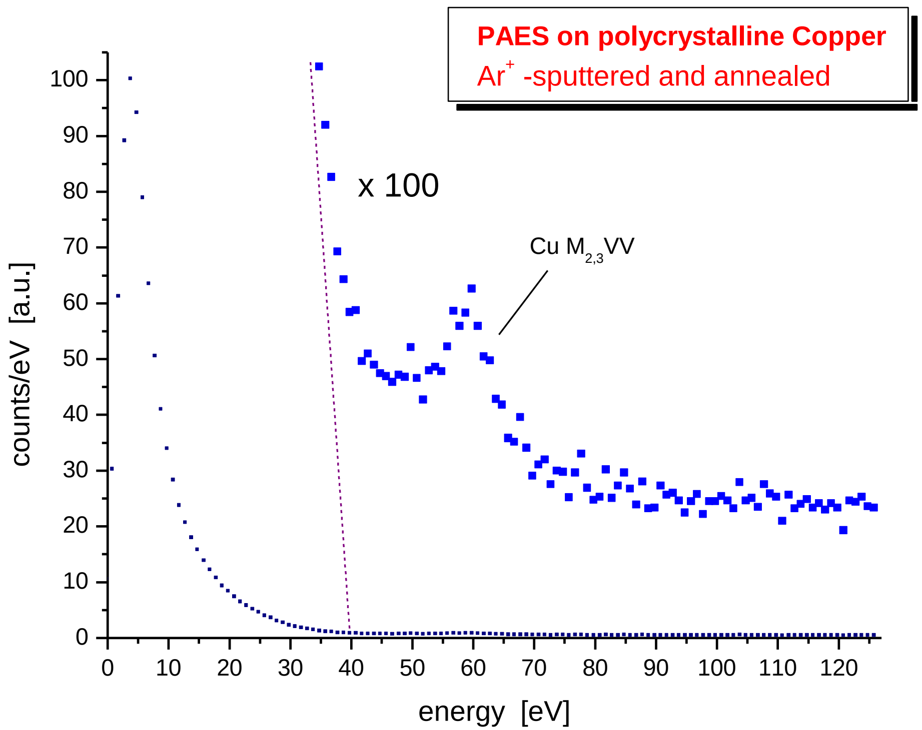
Positron Annihilation Induced AES

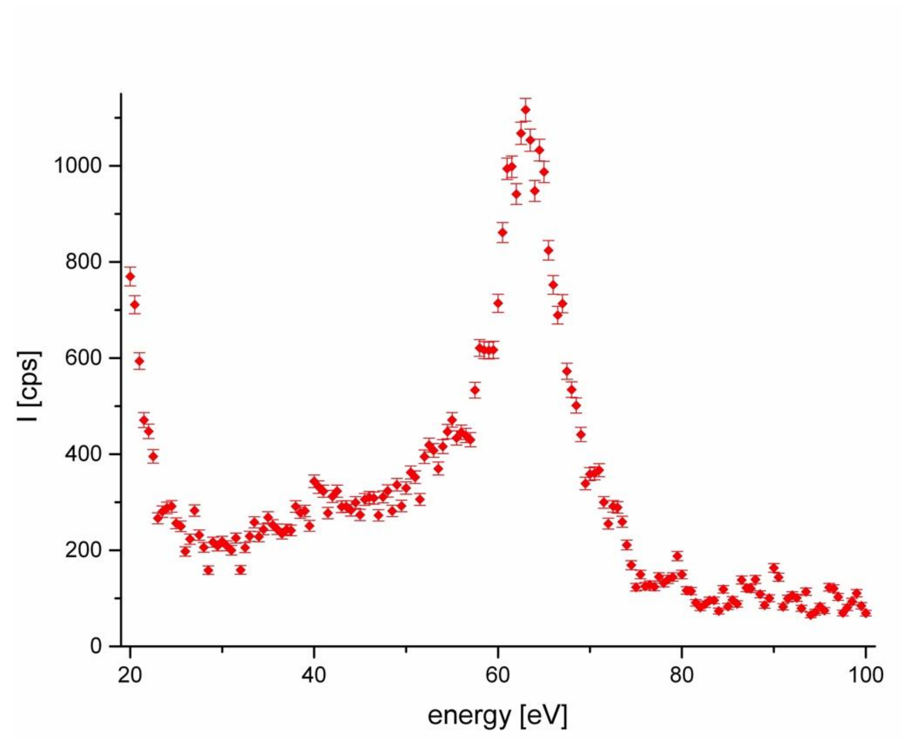


Positron Annihilation Induced AES

^{22}Na based lab beam: 8000 e⁺/s

NEPOMUC: ~4x10^7 e⁺/s
+ efficient e$^-$ detection



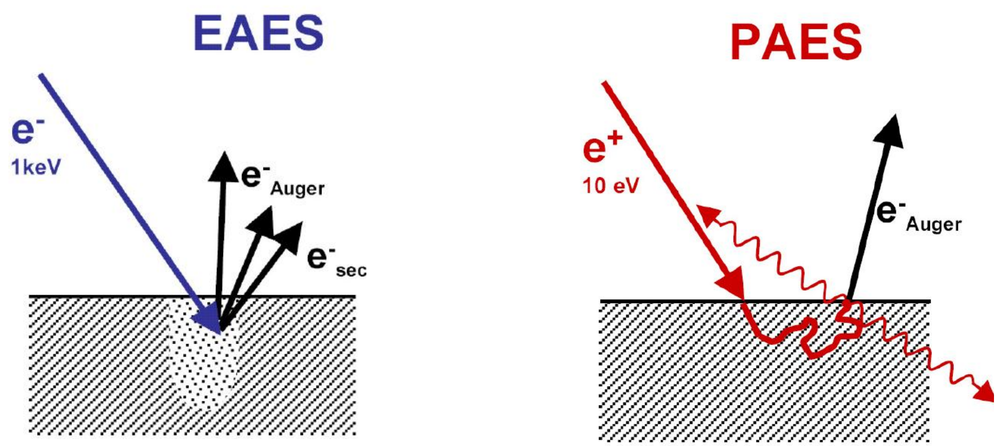


- Acquisition time: 20 days …
- Acquisition time: 7 min. !

B. Straßer, C. Hugenschmidt, K. Schreckenbach
Radiat. Phys. Chem. 68 (2003) 627

J. Mayer, C. Hugenschmidt unpublished (2009)

Emission of Auger-Electrons



	EAES	PAES
currents:	**$I_{e-} > \mu A$**	$I_{e+} < pA$
setup:	**simple**	elaborate
beam energy:	~keV	**~10 eV**
e^-_{sec} background:	high	**"0"**
information depth:	several at. layers	**topmost at. layer**
Auger yield:	-	++

Pd & CuPd

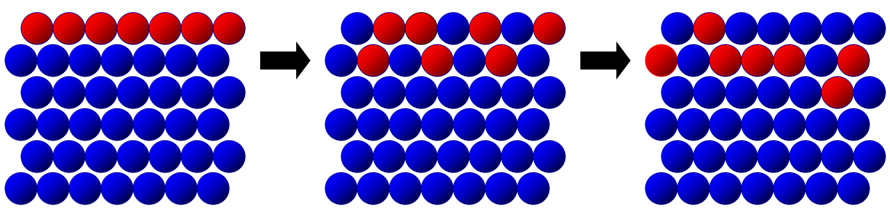
Application of Pd membranes:

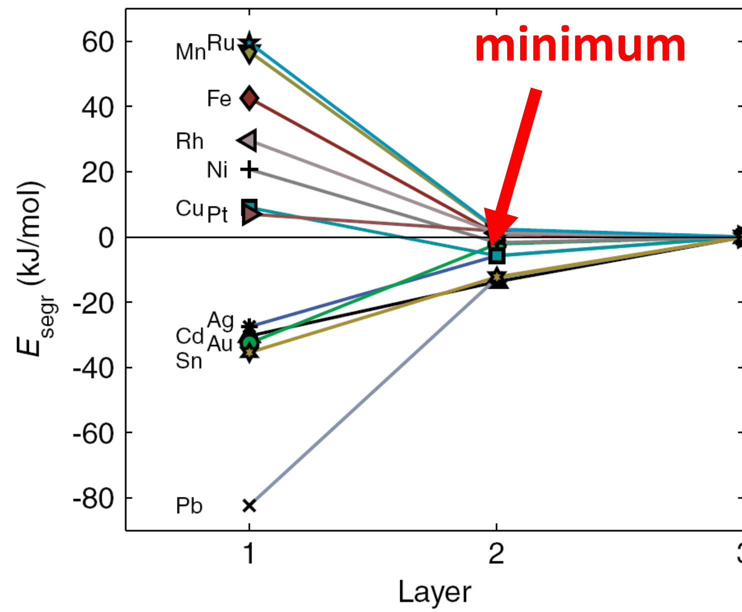
- heterogeneous catalysis
- H storage
- H purification
- CuPd → better H permeation

But:

- segregation at surface & grain boundaries predicted by theory
- calculated segregation energy:

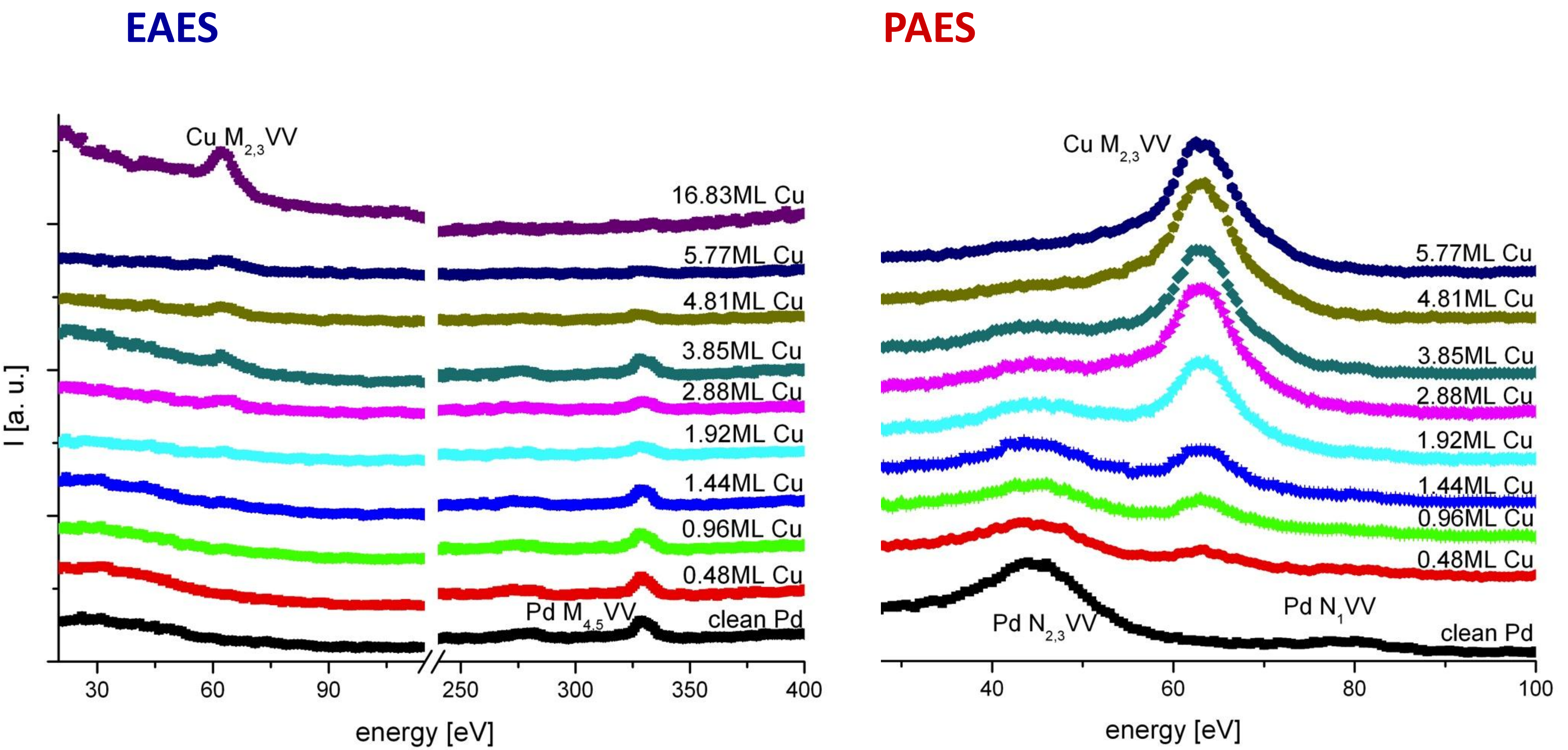
$E_{segr}(CuPd) \sim 60meV$





O.Lovvik, Surf.Sci. 583 (2005) 100

CuPd: Surface Selectivity

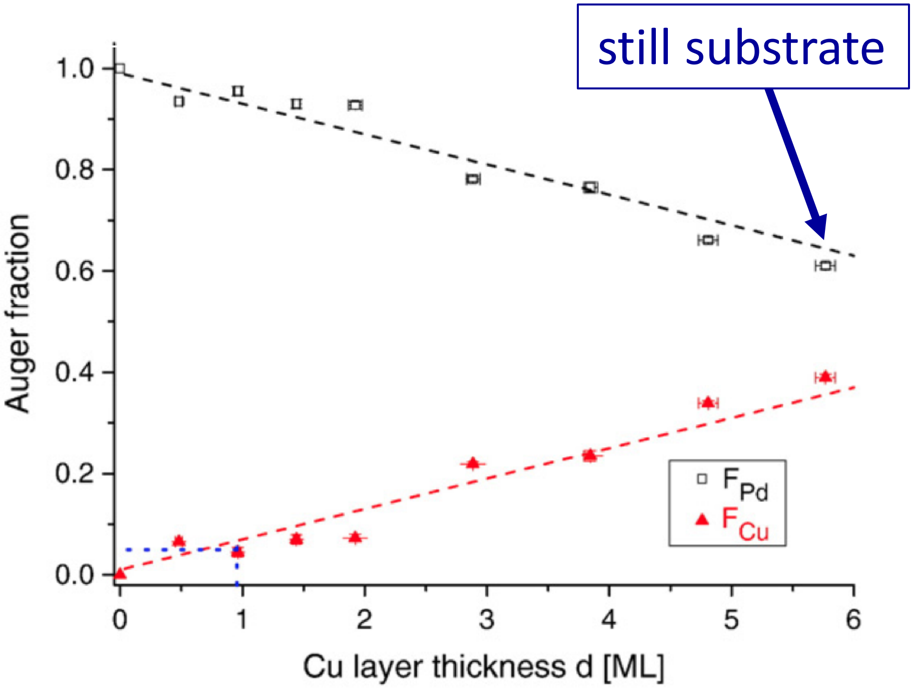


→ compare Auger fractions Cu & Pd

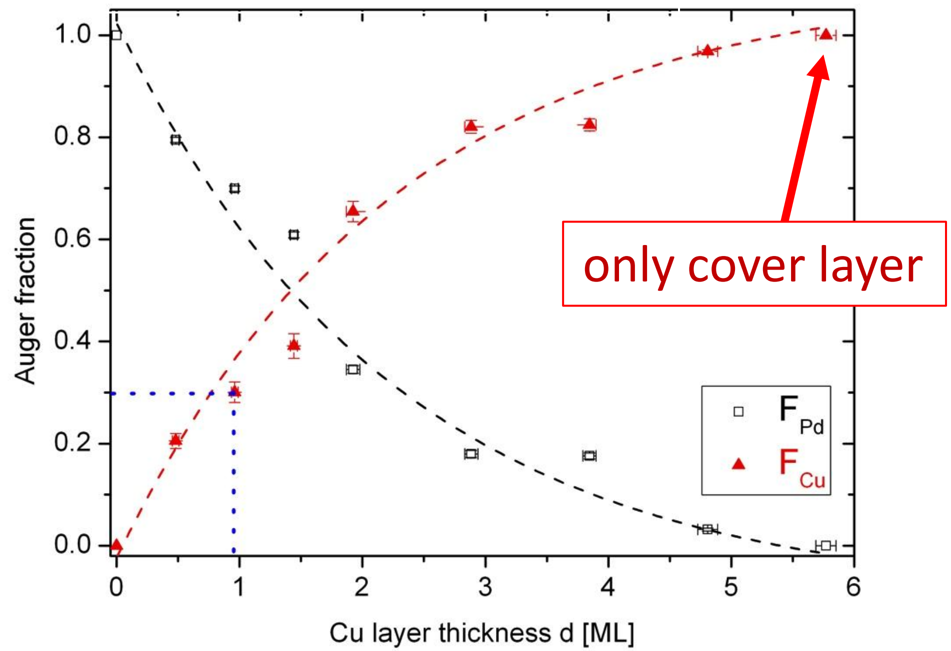
J. Mayer, C. H. et al. Surf. Sci. 604 (2010) 1772

(Sub-)ML Cu on Pd

EAES



PAES



Results

- > 1 ML Cu: Pd still visible → **Cu islands**
- 5.9 ML Cu: only Cu-Augers → **Pd surface completely covered with Cu**

(higher e^+ affinity to Pd)

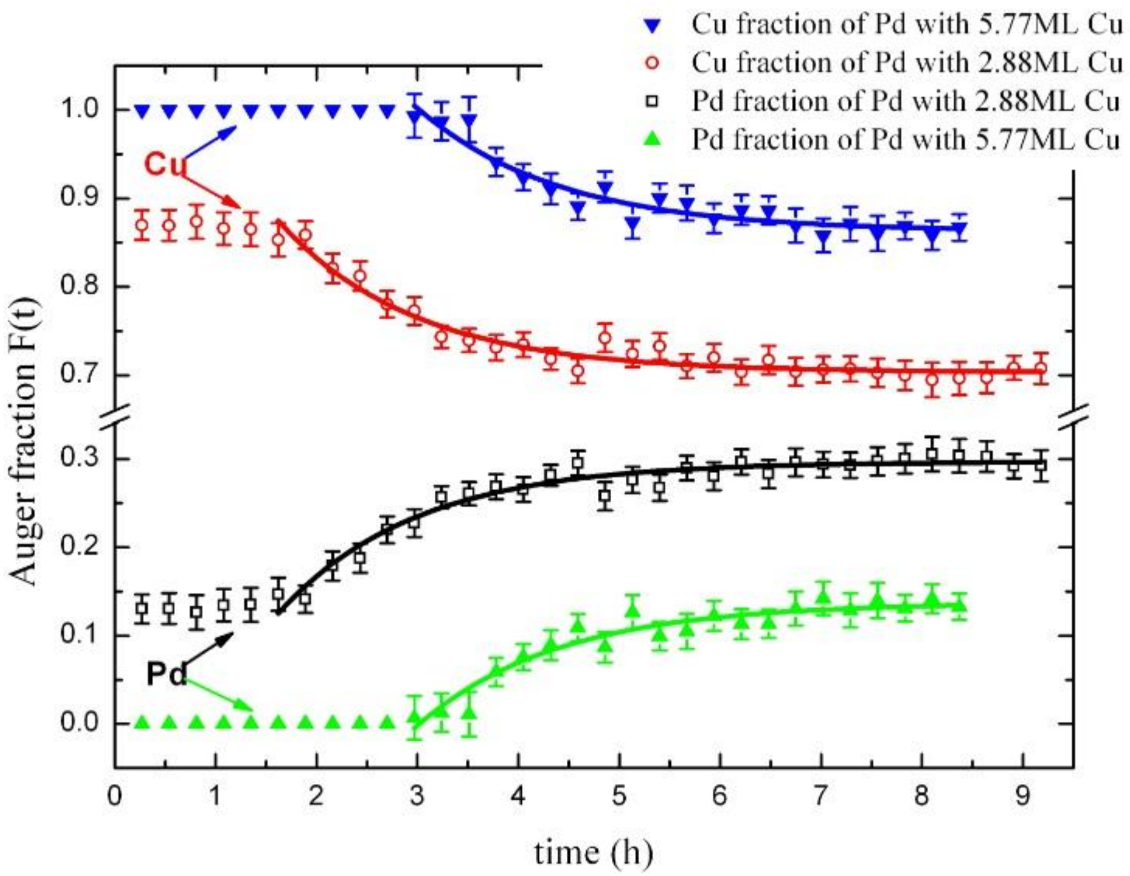
J. Mayer, C. H. et al. Surf. Sci. 604 (2010) 1772

Cu on Pd: Evolution of the Surface

Stability of Cu layer?

→ Few ML of Cu on Pd

→ Time dependent PAES



Results

- **time constant 1.4h**

  >> surf. diffusion

- bulk (self) diffusion @RT

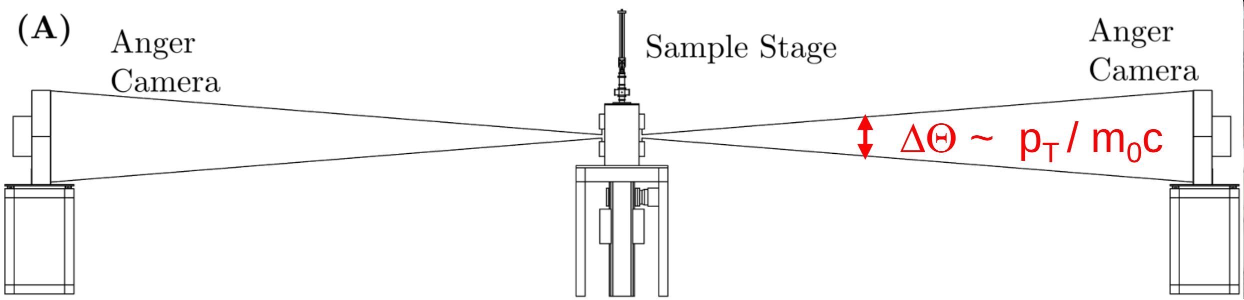
  > 1 nm/h

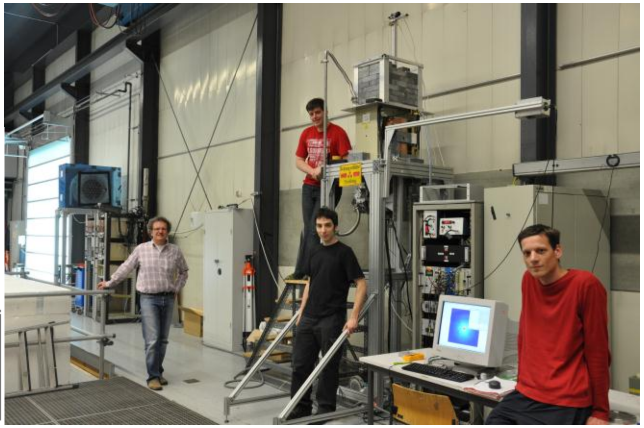
- **segregation of Cu/Pd !**

J. Mayer, C.H. et al. PRL 05 (2010) 207401

ACAR
Angular Correlation of Annihilation Radiation

2D-ACAR spectrometer at TUM:

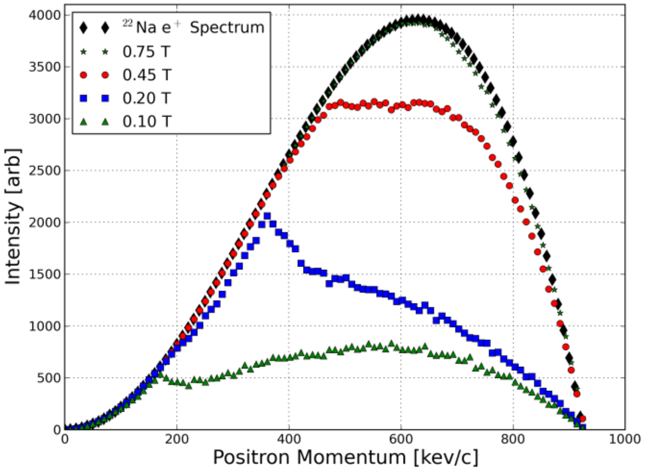


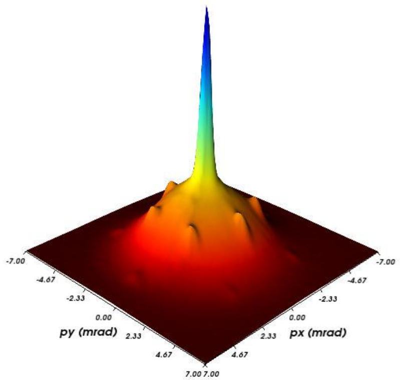


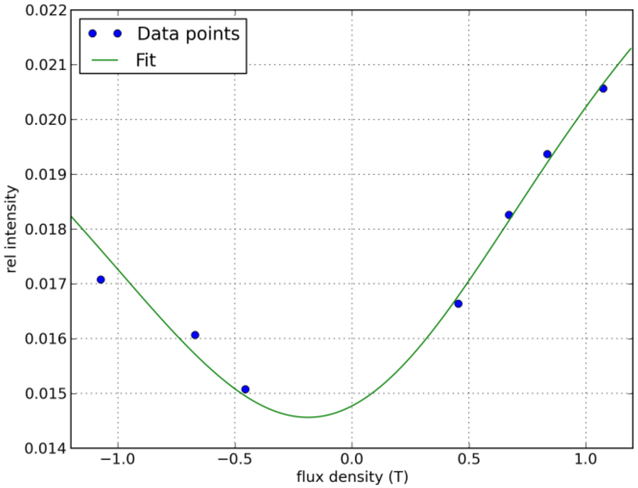
Ceeh et al. Rev. Sci. Instrum. 84, 043905 (2013)

^{22}Na:

- High transport efficiency
- **Polarized positrons:** $P = v_m/c = 0.368(5)$

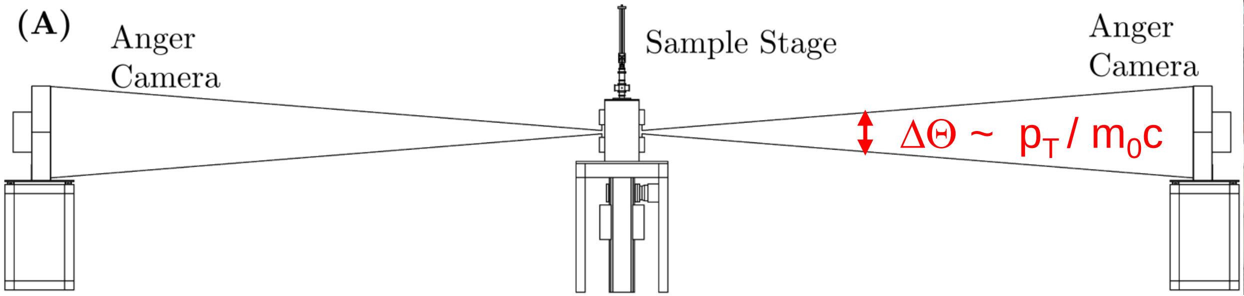


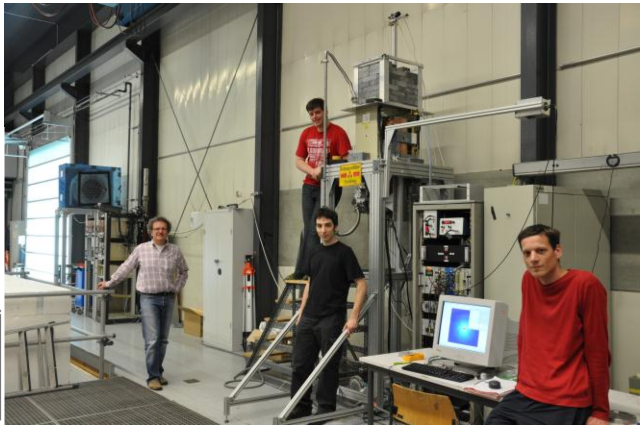




ACAR
Angular Correlation of Annihilation Radiation

2D-ACAR spectrometer at TUM:





Ceeh et al. Rev. Sci. Instrum. 84, 043905 (2013)

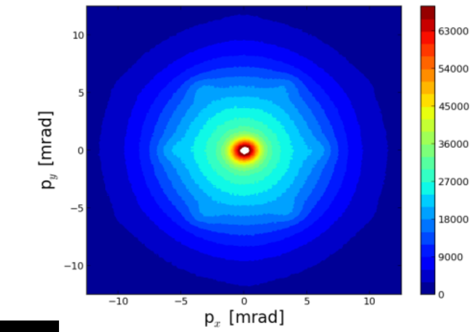
Application of ACAR:

- Determination of electronic structure
  → **Anisotropy of Fermi Surface**
- Special conditions not required
  → **T >> 0, no B-field, no UHV**
- β+ source → **spin-polarized ACAR**
- e+ beam → surface, thin layers, 2D el. systems (planned)

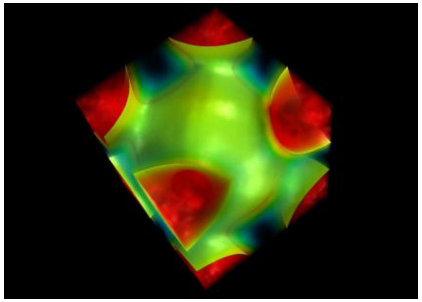
Principle of ACAR

1) Measure **2D-projection of TPMD** (Two-Photon Momentum Distribution) of an oriented single crystal
→ 2D-ACAR spectrum:

$$N(\theta,\phi) = N(p_x, p_y) = \left(\int \rho^{2\gamma}(\mathbf{p})dp_z\right) \otimes R(p_x, p_y)$$

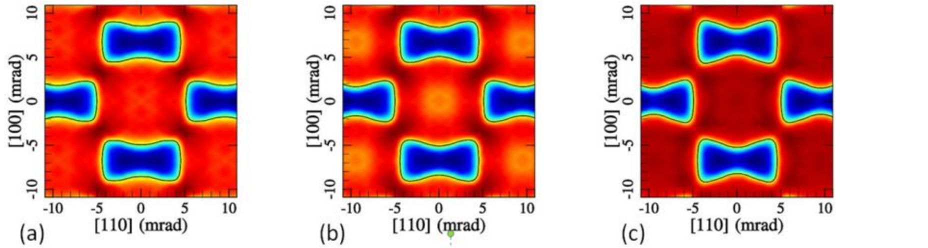


2) **3D-reconstruction** of Fermi surface (FS) from several 2D-projections



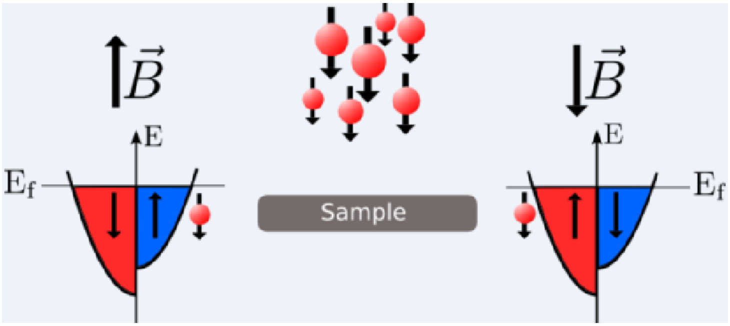
J. A. Weber et al. J. Phys. Conf. Ser. 443(2013) 012092

3) **2D-Cuts** through FS



10K **300K** **300K** (from 5 2D proj.)

4) **Magnetic ACAR**: Measure TPMD with B-field parallel/antip. to P(e+):

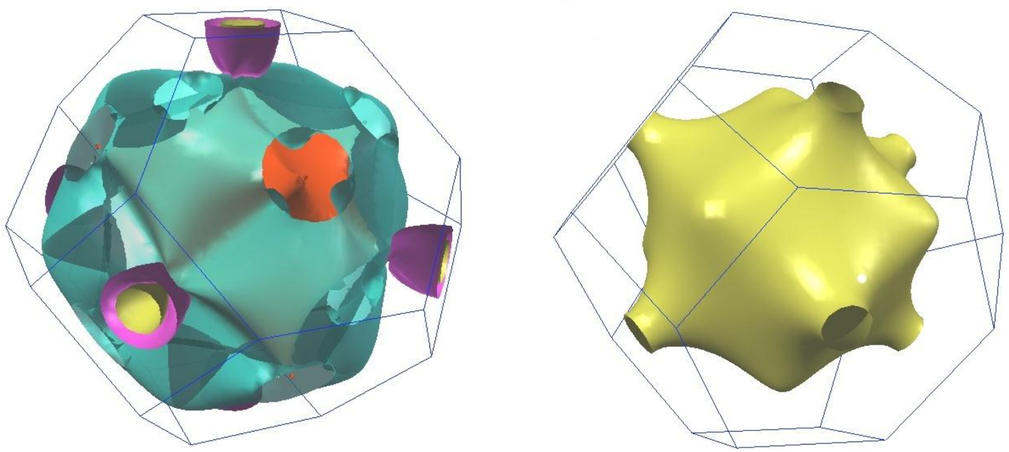


$$N_{\pm}(p_x, p_y) = \frac{\lambda_s}{4} \sum_i^{occ} \left[\frac{(1 \pm P)N_i^{\downarrow}}{\lambda^{\uparrow}} + \frac{(1 \mp P)N_i^{\uparrow}}{\lambda^{\downarrow}} \right]$$

Motivation

Nickel

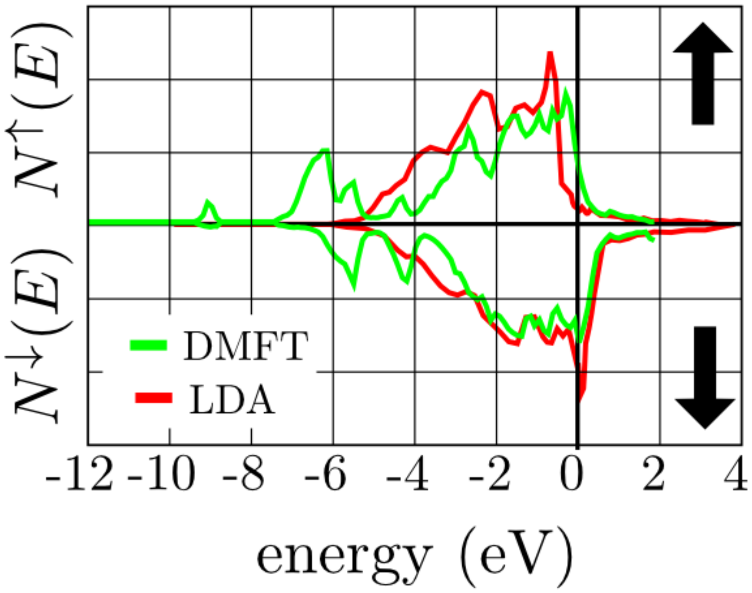
- Magnetic FCC metal
- One unpaired 3d-electron
- "simple" test case for theory and experiment



http://www.phys.ufl.edu

Theory

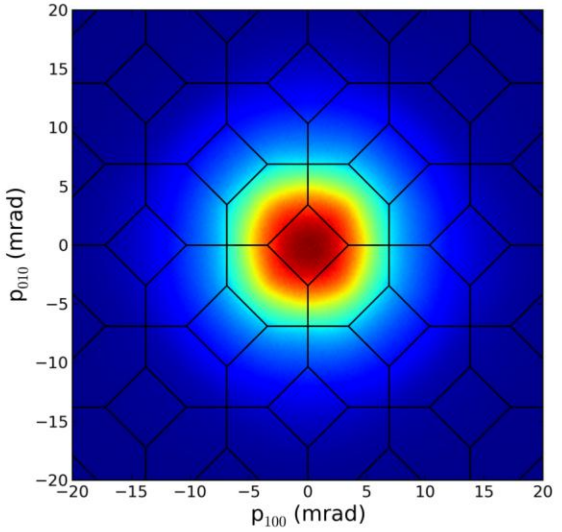
- 6eV satellite peak arises when correlations are included (DMFT)
- Effect of correlation change the shape of the Fermi surface



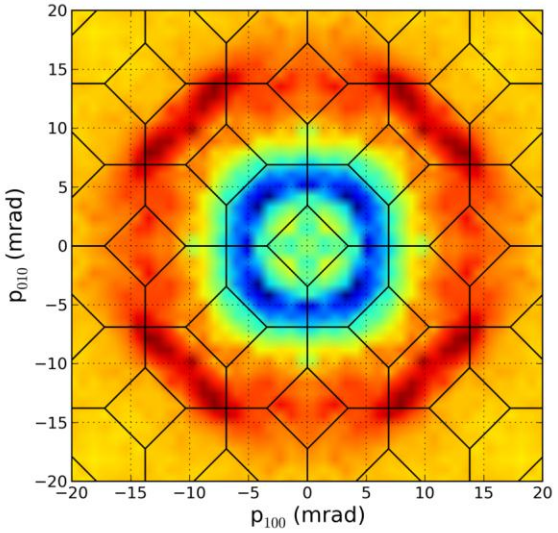
J Kolorenc, et al., arXiv:1202.6595v1

Results on Nickel

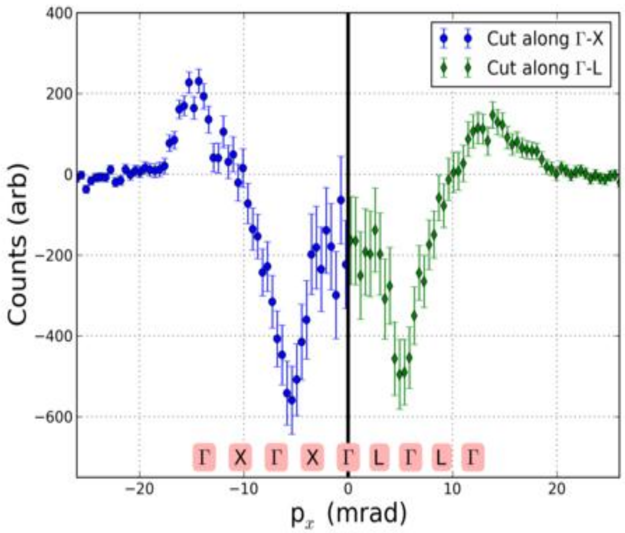
Raw spectrum



Difference



1D-Cuts



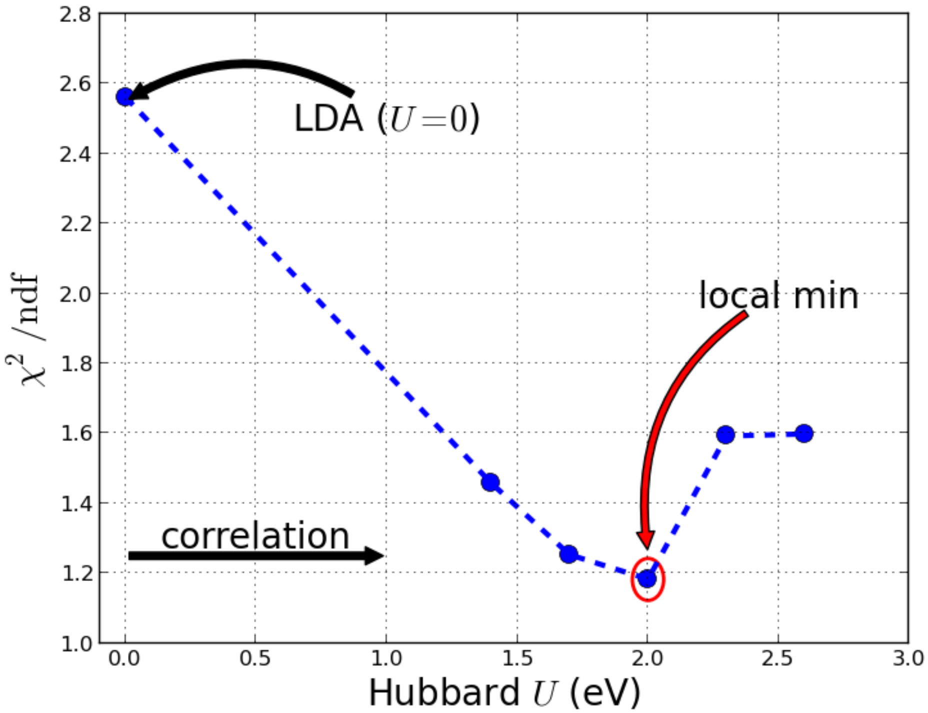
- Integration direction along <100>: 4-fold symmetry

Results (Preliminary! Publication in preparation; H. Ceeh et al.)

→ LCW-folding: transformation from p-space to k-space

- Magnetic difference spectrum exhibits the same symmetry

Electron Correlations



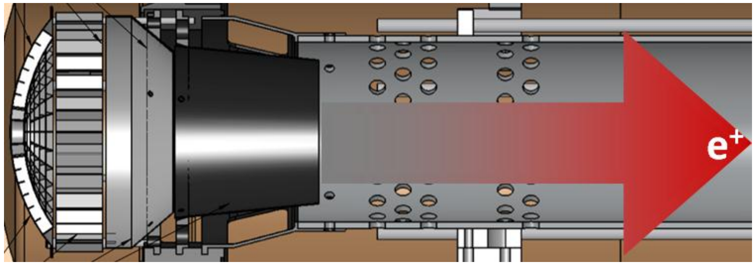
Results (Preliminary! Publication in preparation; H. Ceeh et al.)

- signifcant effect due to electronic correlations
- Hubbard U determines strength of correlation effects
- best agreement for U = 2.0 eV

Summary I

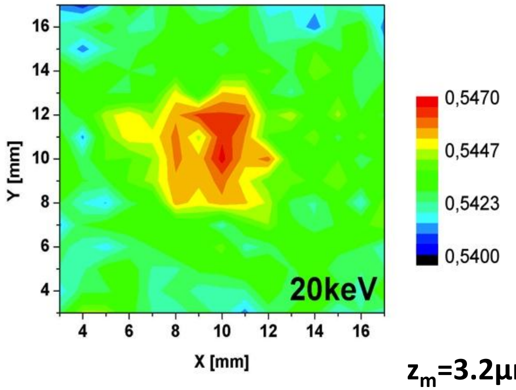
0) Positron as nano-probe:

- **extremely sensitive** to open volume defects & non-destructive
- high-intensity beam NEPOMUC: **>10^9 e+/s**
- **user facility**: NEPOMUC at MLZ Munich



1) Spatial resolved (C)DBS:

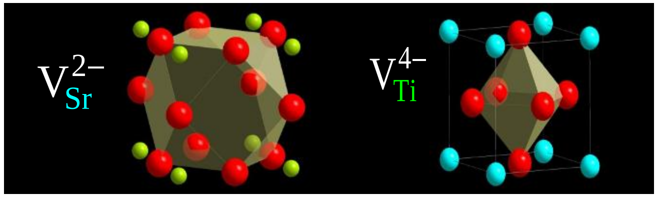
- **3D-defect imaging**
- clusters, layers, **chemical surrounding of defects**
- T-dependent defect annealing



z_m=3.2µm

2) Defect spectroscopy with e+ lifetime:

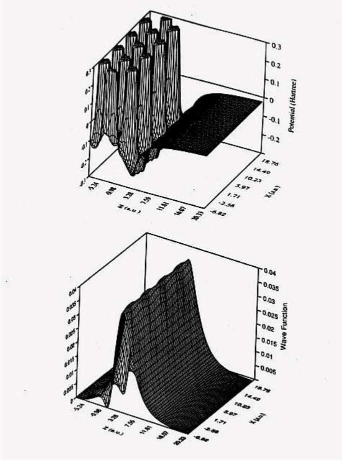
- type & concentration of **defects**
- **free volume**
- depth profile (beam)



Summary II

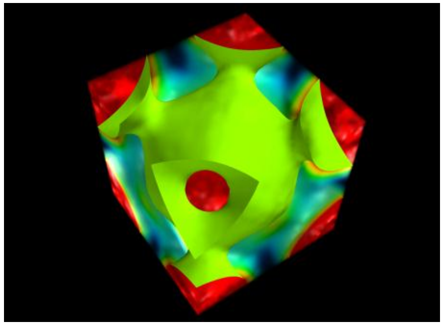
3) Surface physics with (t-dependent) PAES :

- **“no“ secondary electrons** & non-destructive
- top most atomic layer **sensitivity**
- surface **segregeation**



4) Electronic structure, Fermi surfaces:

- **T >> 0, no B-field**
- **spin-resolved ACAR**
- bulk, surface & **layers** (beam)



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Many sutdents

+

Many external users

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