New Positron Beam Applications: Positrons Reveal Lattice Defects in Functional Materials and Electronic Structure in Correlated Systems

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NEPOMUC

E21
Arbeitsgebiet
Physik mit Positronen

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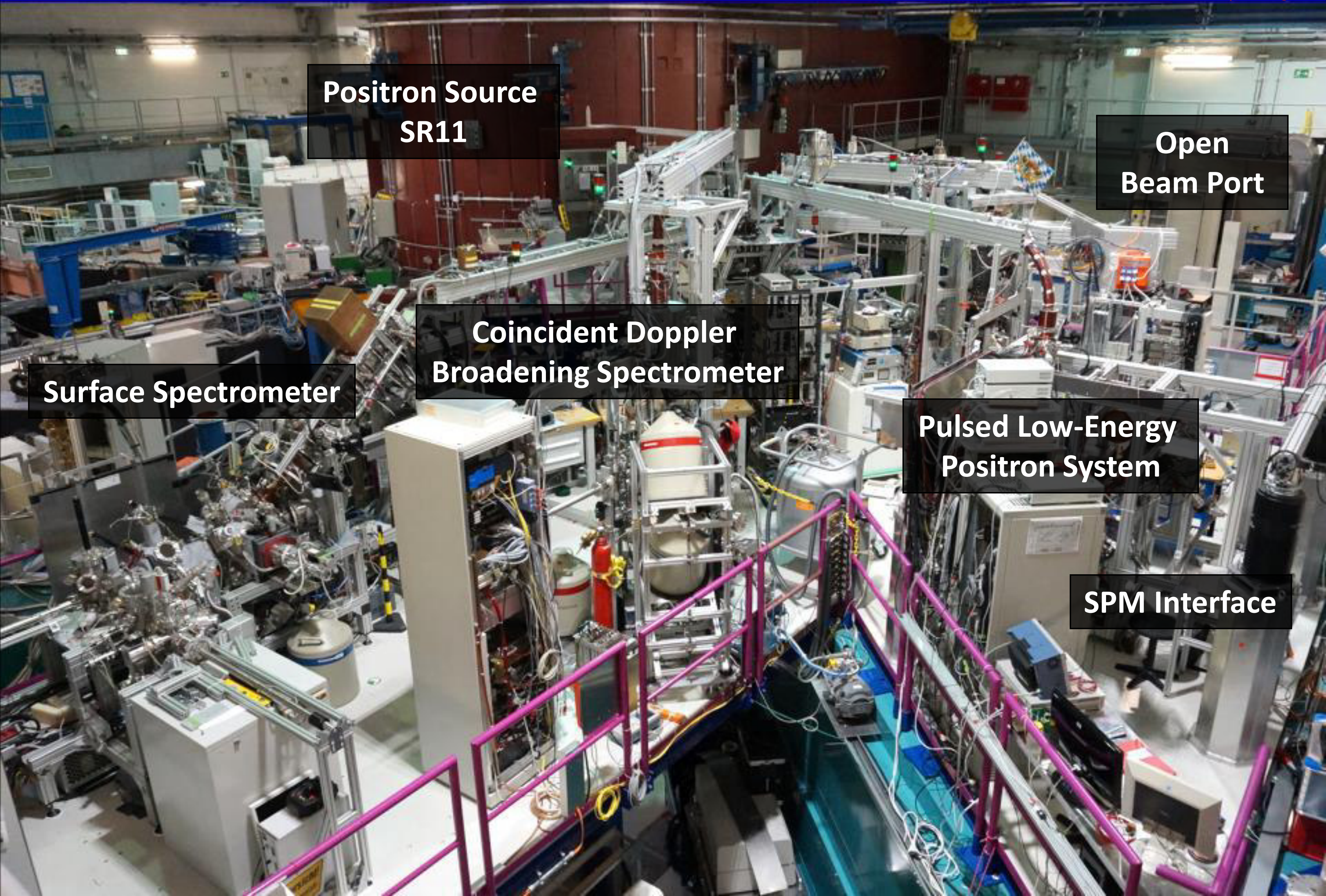
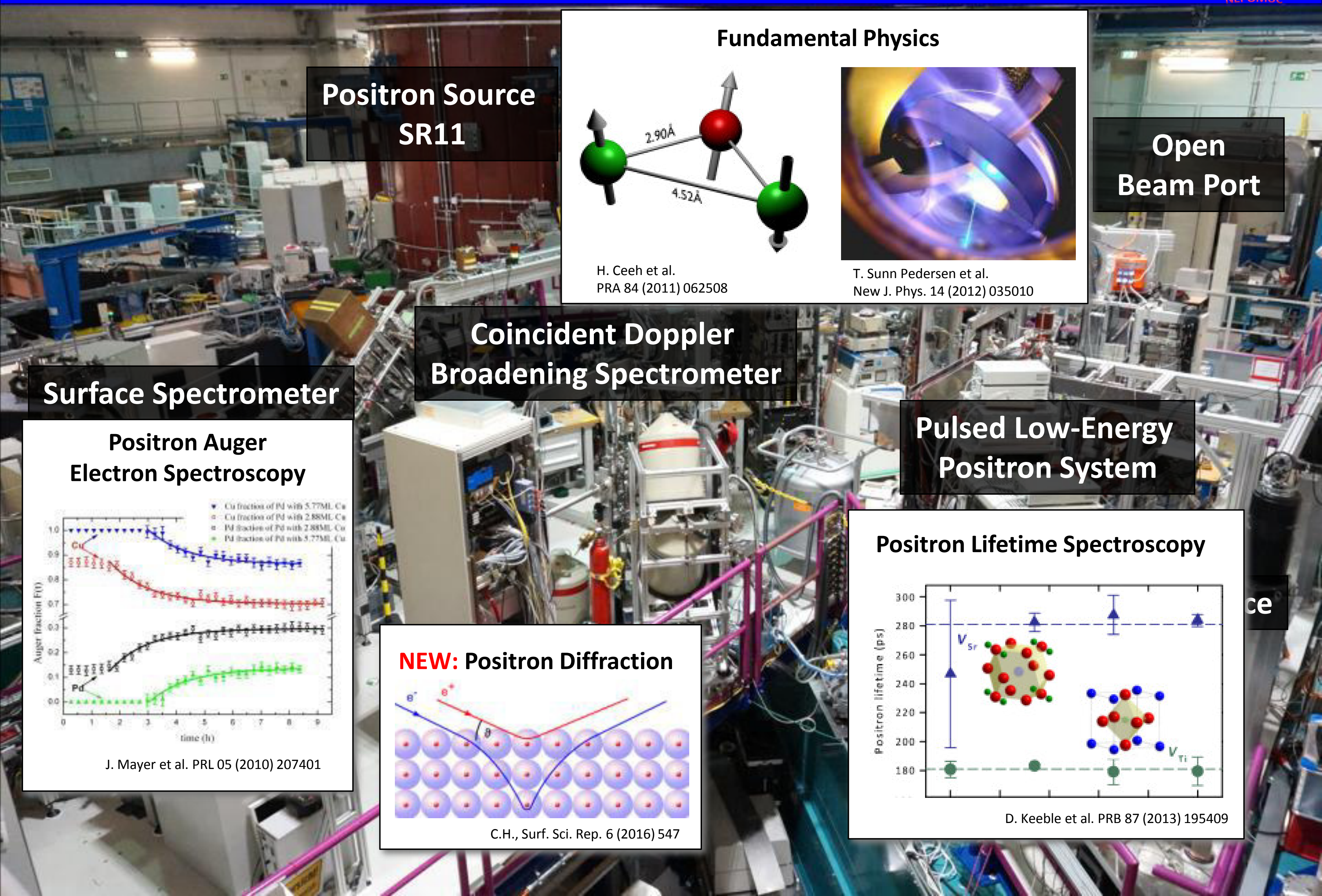
Positron Beam Facility at NEPOMUC



Positron Beam Facility at NEPOMUC



Outline

Positrons in Matter

Part I

(C)DBS at NEPOMUC

YBCO

AlCu6Mn

Part II

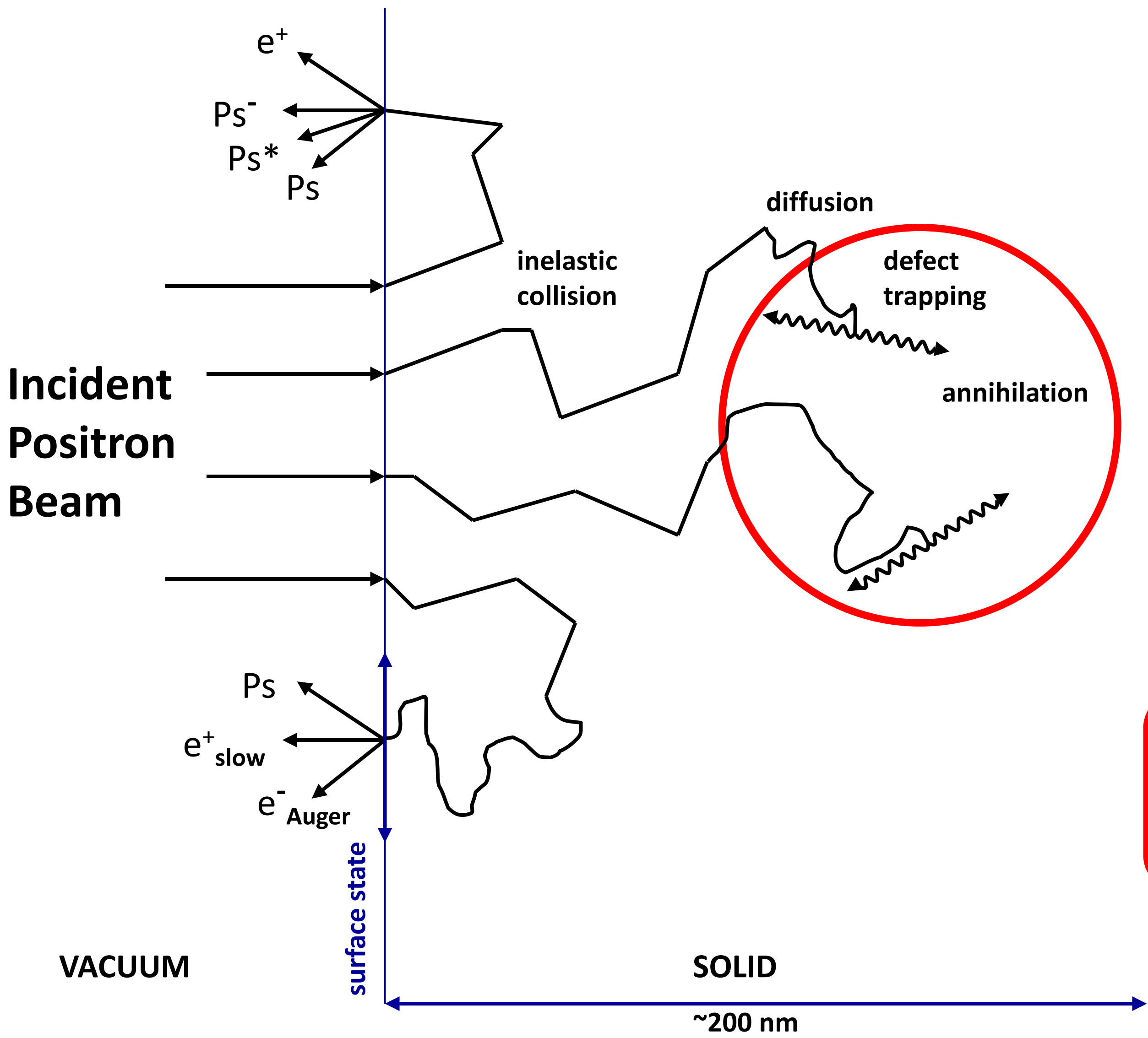
2D-ACAR with ^{22}Na

Ni

Cu_2MnAl

Conclusion & Future

Positrons in Matter



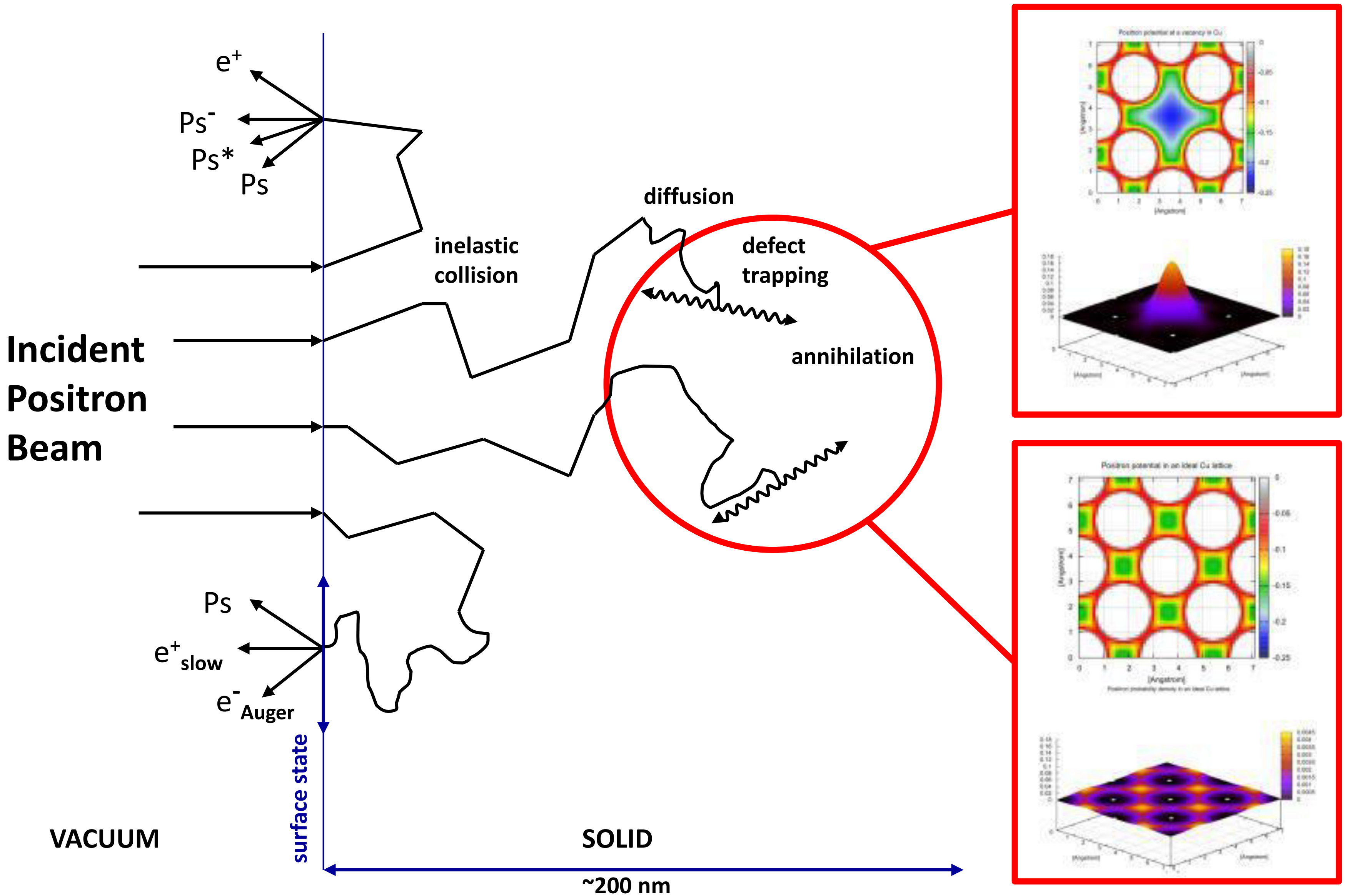
Positrons fate:

- thermalization
  ~10^{-12} s
- diffusion
  ~10^{-10} s
  → ~100 nm
- defect trapping
- annihilation
  → 2 collinear γ-quanta

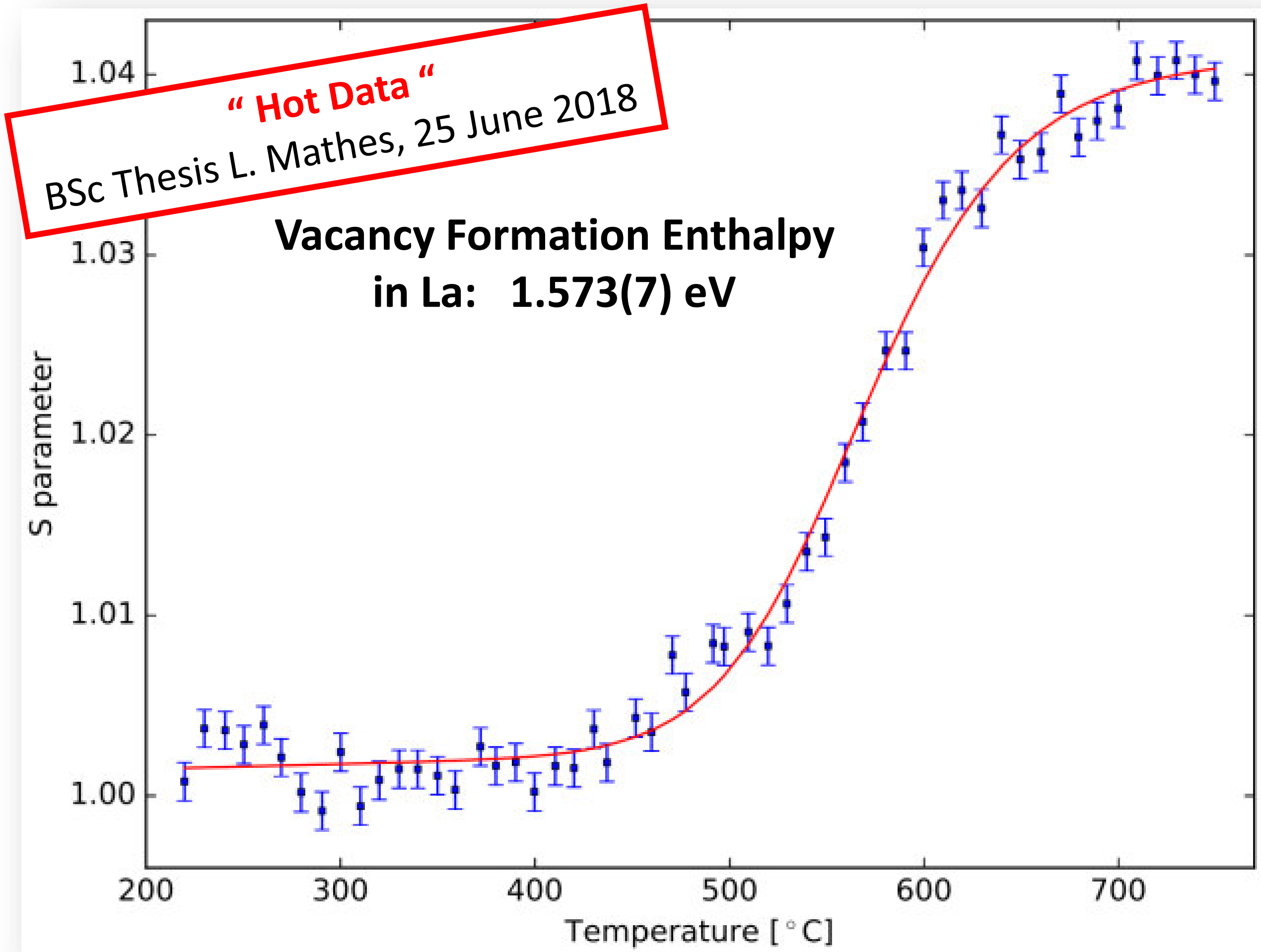
Experiment:

- positron lifetime τ
  → ρ (e-)
- Doppler-broadening ΔE
  Angular correlation $\Delta\Theta$
  → **p(e-)**
- Auger-electrons
  → E_b(e-)

Positrons in Matter

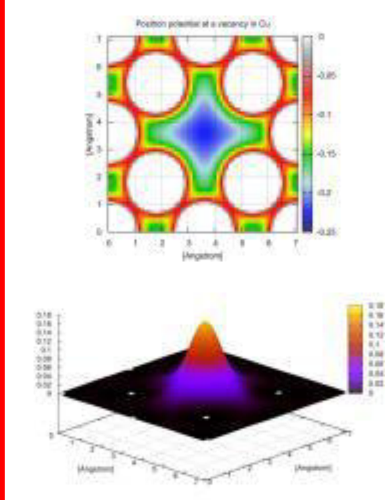


Positrons in Matter



“ Hot Data “
BSc Thesis L. Mathes, 25 June 2018

Vacancy Formation Enthalpy
in La: 1.573(7) eV



VACUUM

surface state

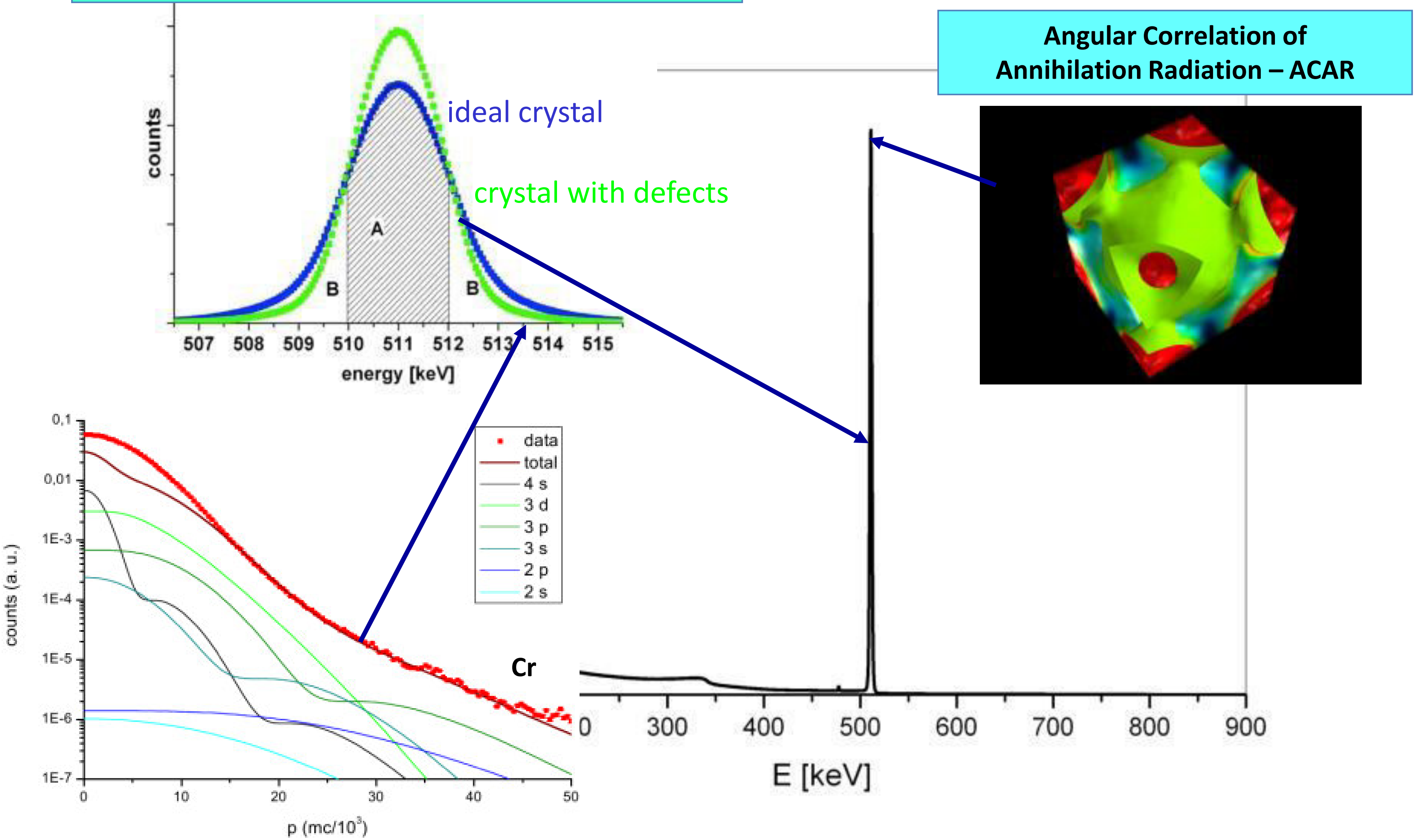
SOLID

~200 nm

What We Measure

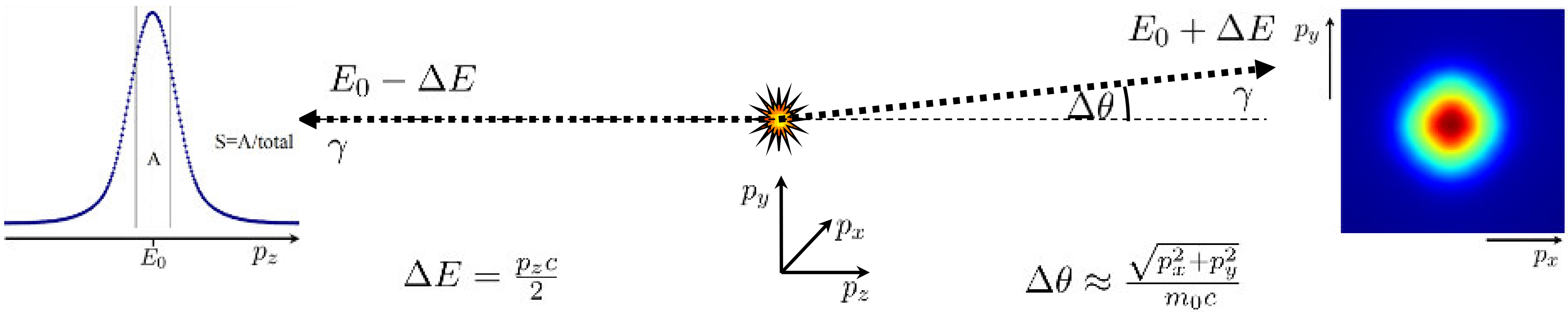
Doppler-Broadening Spectroscopy – DBS

Angular Correlation of Annihilation Radiation – ACAR



Coincident Doppler-Broadening Spectroscopy – CDBS

2γ - Annihilation & Electron Momentum



Observable	Doppler shifted γ ***energy***	***angle*** between γ's
Detector	high ***energy* resolution**	high ***spatial* resolution**
Method	**(C)DBS**	**2D-ACAR**
e^+ Source	NEPOMUC **e^+ beam**	**β^+ source** ^{22}Na
e^+ State	**localized**	**delocalized**
Physics	**defect spectroscopy & imaging**	**electronic structure, Fermi-surface**
Lab		MLL
Money	GEFÖRDERT VOM Bundesministerium für Bildung und Forschung	Deutsche Forschungsgemeinschaft DFG

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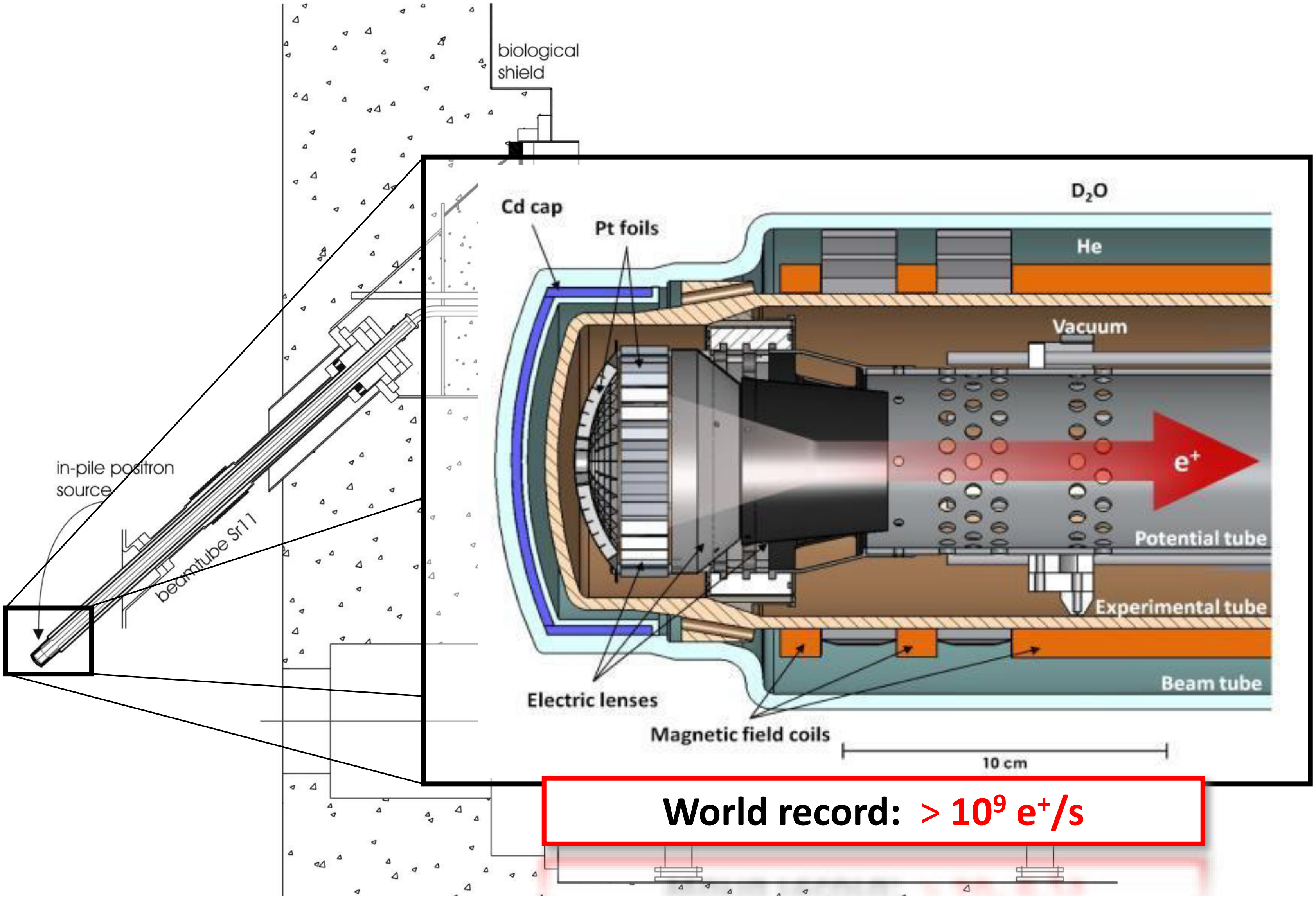
NEPOMUC

NEutron induced POsitron Source MUniCh



World record: > 10^9 e^+/s

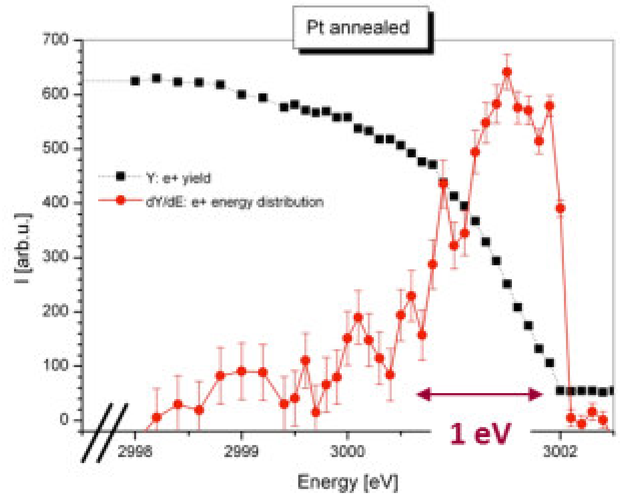
C. H. et al NIM A 593 (2008) 616; New J. Phys. 14 (2012) 055027; J. Phys. Conf. Ser. 443 (2013) 012079; Surf. Sci. Reports 71 (2016) 547

Positron Beam at NEPOMUC

- Pair production in Pt $\gamma \rightarrow e^+ e^-$
- Pt as converter **AND** moderator ϕ^+_{Pt} **= -1.95(5) eV**

E_{e+} ~ keV

$\Delta E < 2$ eV

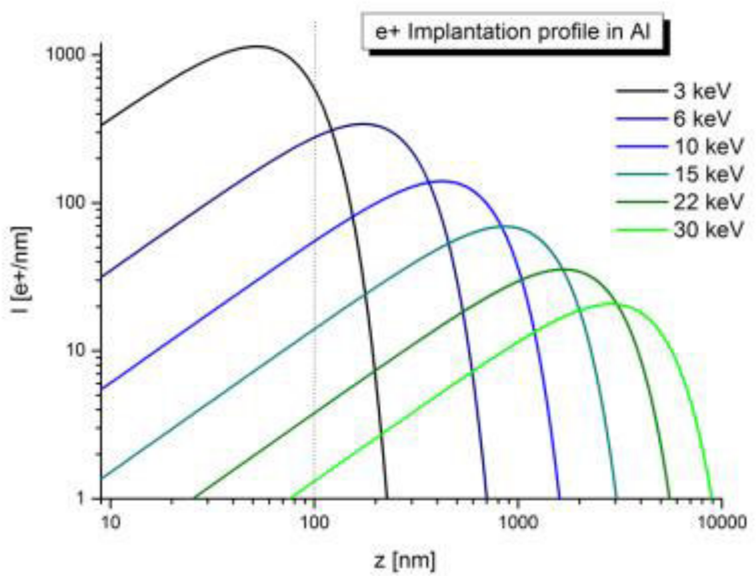


C. H. et al. NIM B 198 (2002) 220

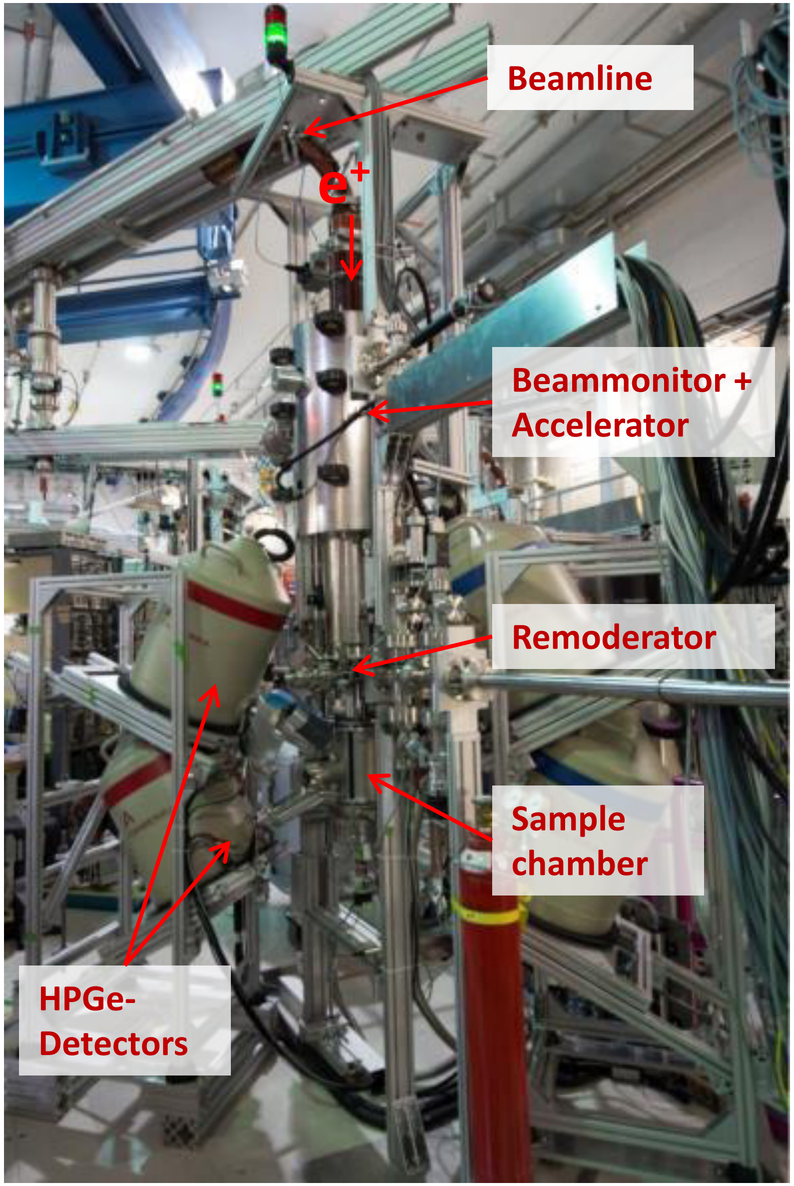
	Primary beam	Remoderated beam
E	**1 keV**	**20 eV**
d_{FWHM}	9 mm @5 mT	< 2 mm @4 mT
I_{beam}	**$1.1 \cdot 10^9$ e^+/s**	**$4\text{-}5 \cdot 10^7$ e^+/s**

- CDBS: Acceleration at sample

E = 0.1 – 30 keV

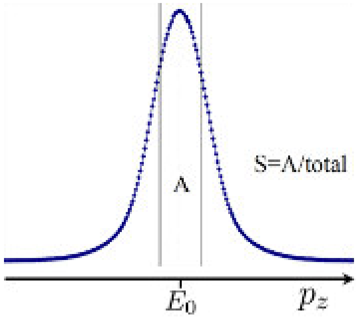


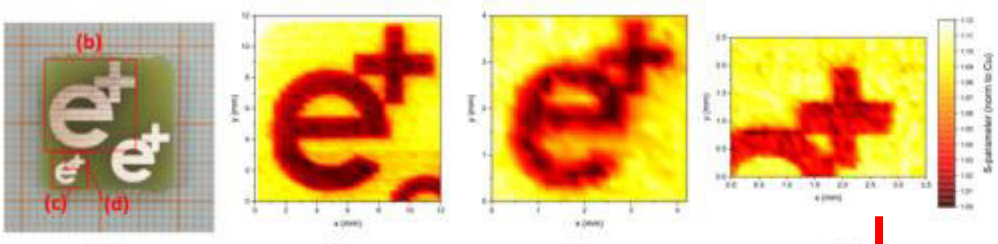
CDB Spectrometer at NEPOMUC



Doppler broadening spectroscopy:

- S-parameter:
  S(T), S(E), S(x, y)





(a) (b) (c) (d)

- NEPOMUC rem. beam: 253 µm
- **CDB microbeam: 49 µm**
  + aperture: 33 µm
- Scan area: 19x19 mm^2

T. Gigl, L. Beddrich, M. Dickmann, B. Rienäcker, M.Thalmayr, S. Vohburger, and C. Hugenschmidt, New J. Phys. 19 (2017) 123007

Superconductivity in YBCO

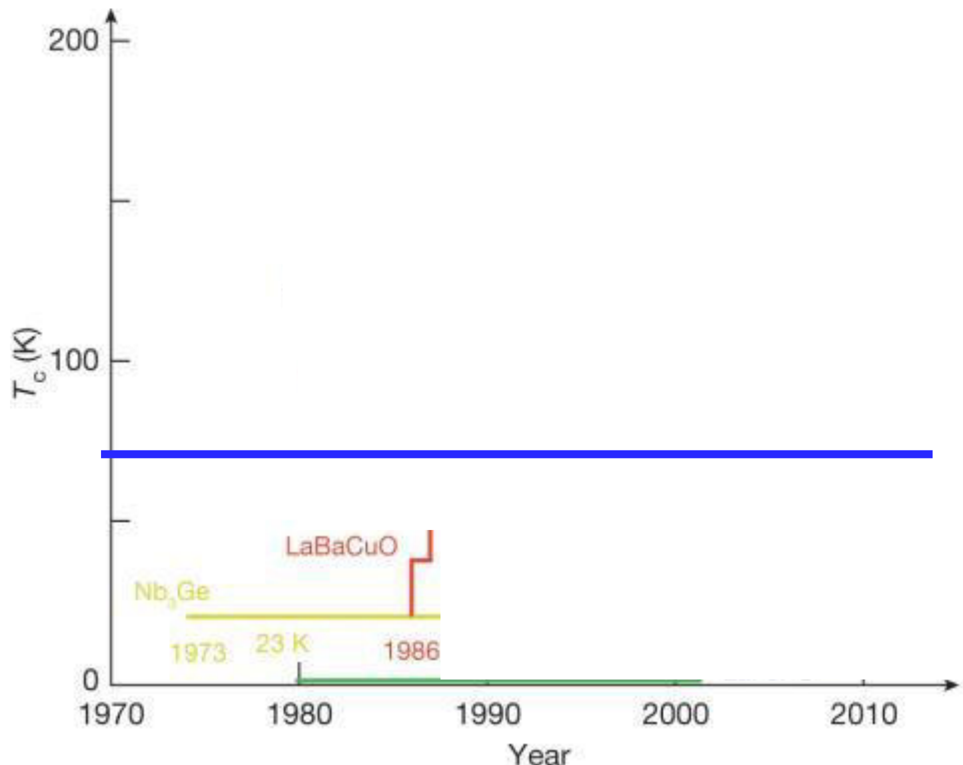
High-T_c Superconductivity in Cuprates

- **LaBaCuO $\quad T_c = 35$ K**

  1986 A. Müller & G. Bednorz
  → 1987 Nobel prize

- **$YBa_2Cu_3O_{6.93}$ $\quad T_c = 95$ K**

  1987 Maw-Kuen Wu & Chu Ching-Wu



B Keimer et al. Nature 518, 179 (2015)

Applications of YBCO

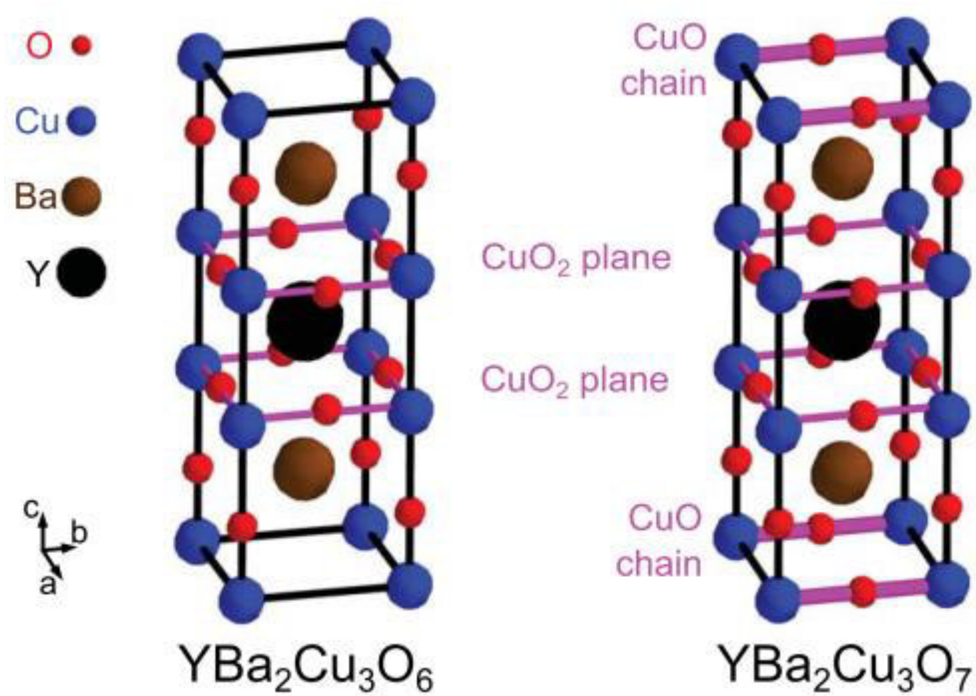
- **High-T_c model system**
- **Power transmission**
- **High field magnets**

http://eneff-industrie.info/projekte/2017/supraleiter-kabel-in-essen

Superconductivity in YBCO

Structure

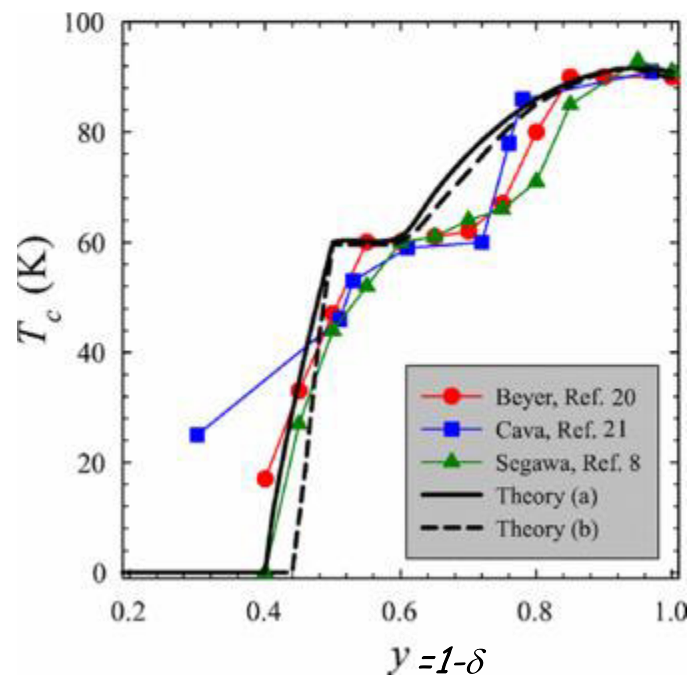
tetragonal | orthorombic



insulating | superconducting

Phase Diagram

$YBa_2Cu_3O_{6+y}$



Zaleski et al. PRB, 74 (2006) 014504

→ **Oxygen content crucial for T_c**

Single Crystalline $YBa_2Cu_3O_{7-\delta}$ Thin Films

Preparation

- Pulsed Laser Deposition (PLD) on STO substrate → **Thickness 210 nm** +/-10 nm
- Heat treatment (~400°C) → Reduction of oxygen content

Characterization

- Transport measurements
- XRD → c-axis parameter

  → δ and **T_c *mean*** values

Positrons

- DBS (x,y,E,T) & CDBS

  → **δ(x,y,z)** & **T_c(x,y,z)**

Sample	δ	T_c(K)	t_{temp}(min)	p_{temp}(mbar)
A1	0.191	90	n. a.	n. a.
A2	0.475	60	30	$2\cdot10^{-2}$ (O_2)
A3	0.641	60	30	10^{-7}
A4	0.791	25	50	10^{-7}

DBS on YBCO Thin Films

S(E) → depth dependent positron annihilation

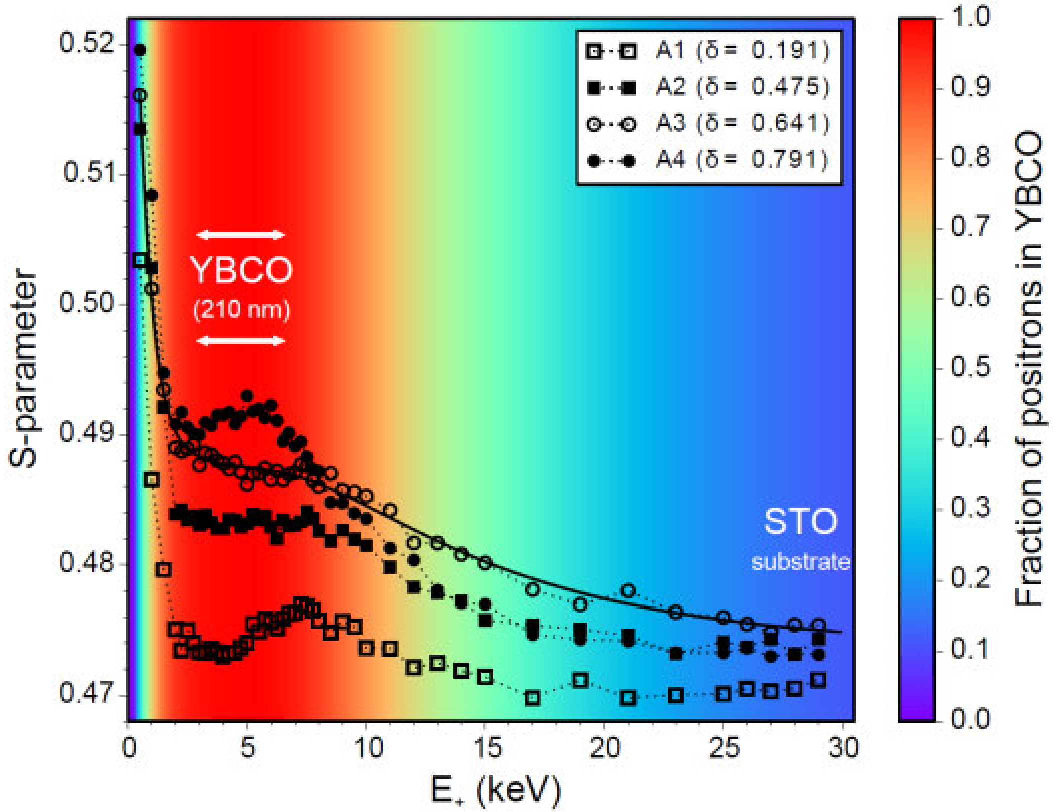
- Surface: E < 3 keV

  Steep increase of S

  → Low positron mobility

- YBCO film: E < 7.5 keV

- Interface YBCO/STO: 7.5 keV



Data fit:

- S(E) fit with a two-layer model: 210 nm YBCO on STO

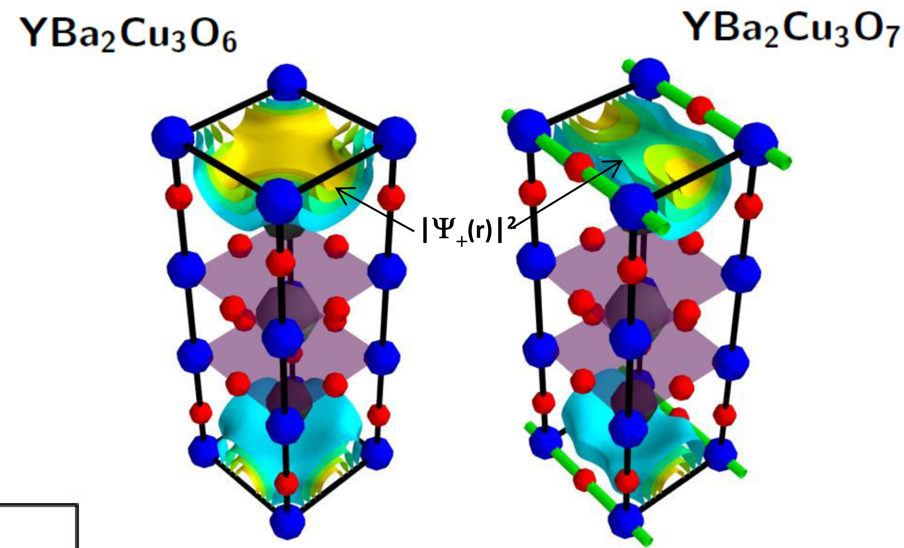
  → Fraction of positrons annihilating in YBCO layer

  → **Very short diffusion length: 1-2nm**

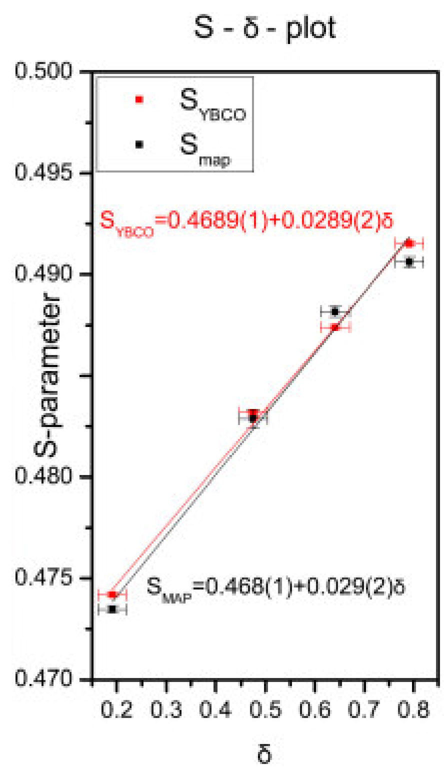
Oxygen Deficiency in YBCO

Positrons in $YBa_2Cu_3O_{7-\delta}$

- Positrons probe plane of oxygen deficiency
- Low positron mobility along c-axis
  → as observed in S(E)



- Linear correlation of
  δ (XRD) with **S** (DBS):

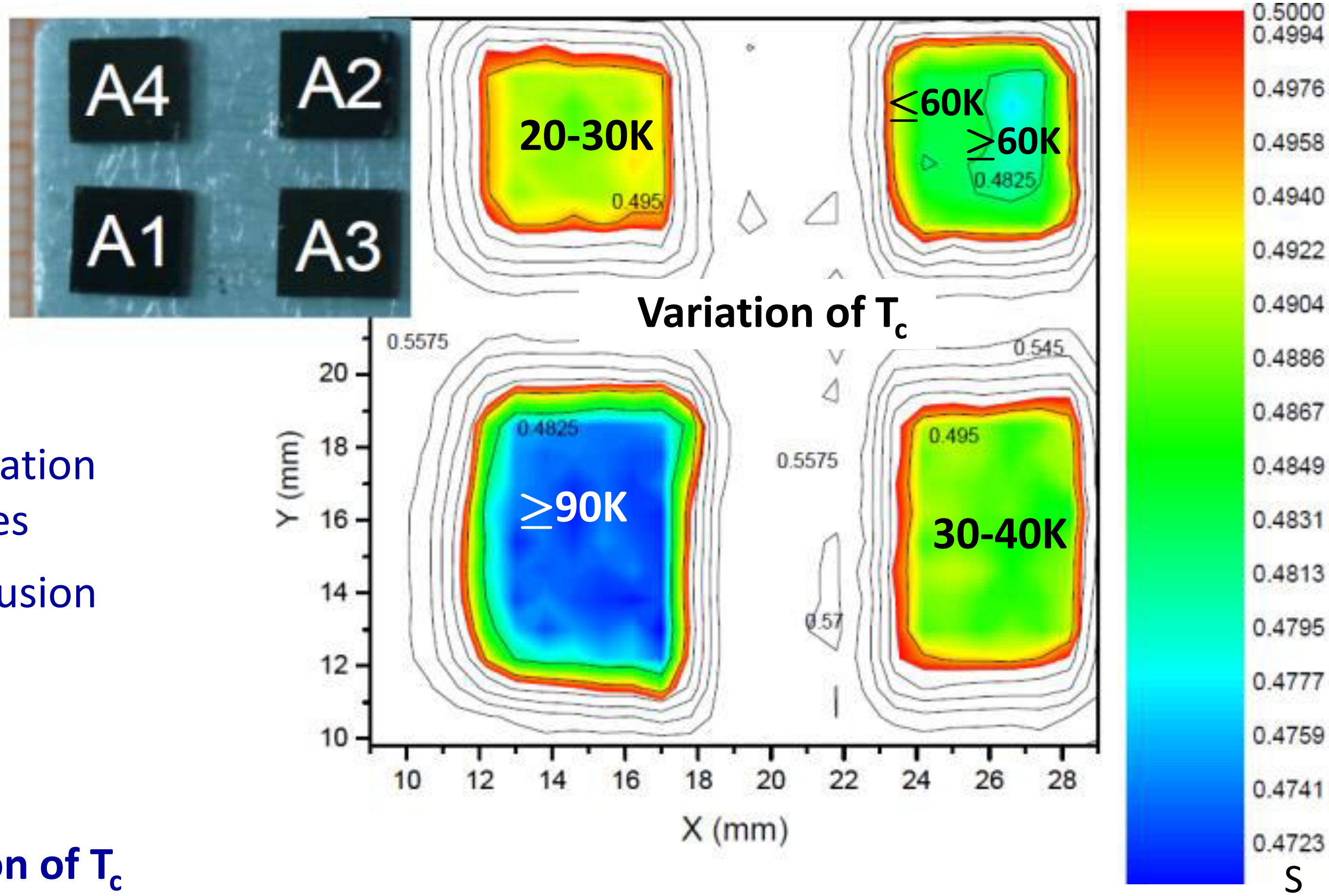


DBS with Positronbeam

- $YBa_2Cu_3O_{7-\delta}$: Measure **spatial distribution of δ** and determine **local variation of T_c**

Imaging of δ and T_c Variation

DBS with Scanning Positron Beam → S(x,y)



- **S(δ) :**

→ Spatial δ variation within samples

→ Local out diffusion of oxygen

- **T_c(δ):**

→ **Local variation of T_c**

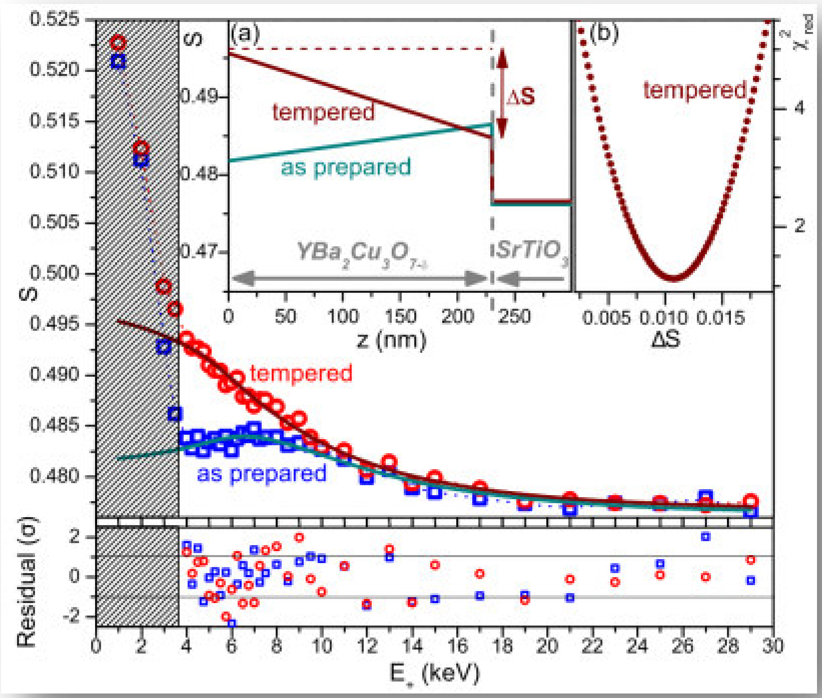
M. Reiner, T. Gigl, R. Jany, G. Hammerl, C. Hugenschmidt; Appl. Phys. Lett. **106**, 111910 (2015)

Depth Profile of T_c

DBS with Positron Beam → S(z)

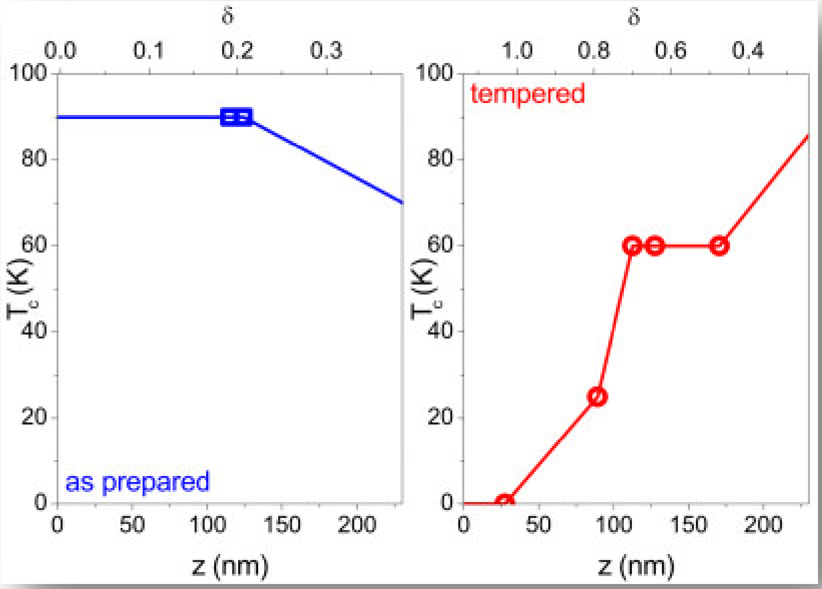
- 230nm YBCO film as prepared & after tempering at 400°C
- Use simple model for S(z) sufficient to explain $S(E_+)$

$S(E_+)$ → S(z)



δ(z) → T_c(z)

→



(dots: ref. samples)

M. Reiner, T. Gigl, R. Jany, G. Hammerl, C. Hugenschmidt; Physical Review B 97 (2018) 144503

Laser Beam Welded Al Alloy

Material: **AlCu6Mn** (EN AW 2219-T87)

- Age hardenable Al alloy
- High strength and low weight

Laser beam welding: (LBW)

- Beam spot & heat impact small
  → small heat affected zone (HAZ)
- Weld accuracy & reproducibility high

LBW of Al alloys:

- Weight reduction
  → replacement of steel and riveted joints

Sample preparation IWB at TUM **:**

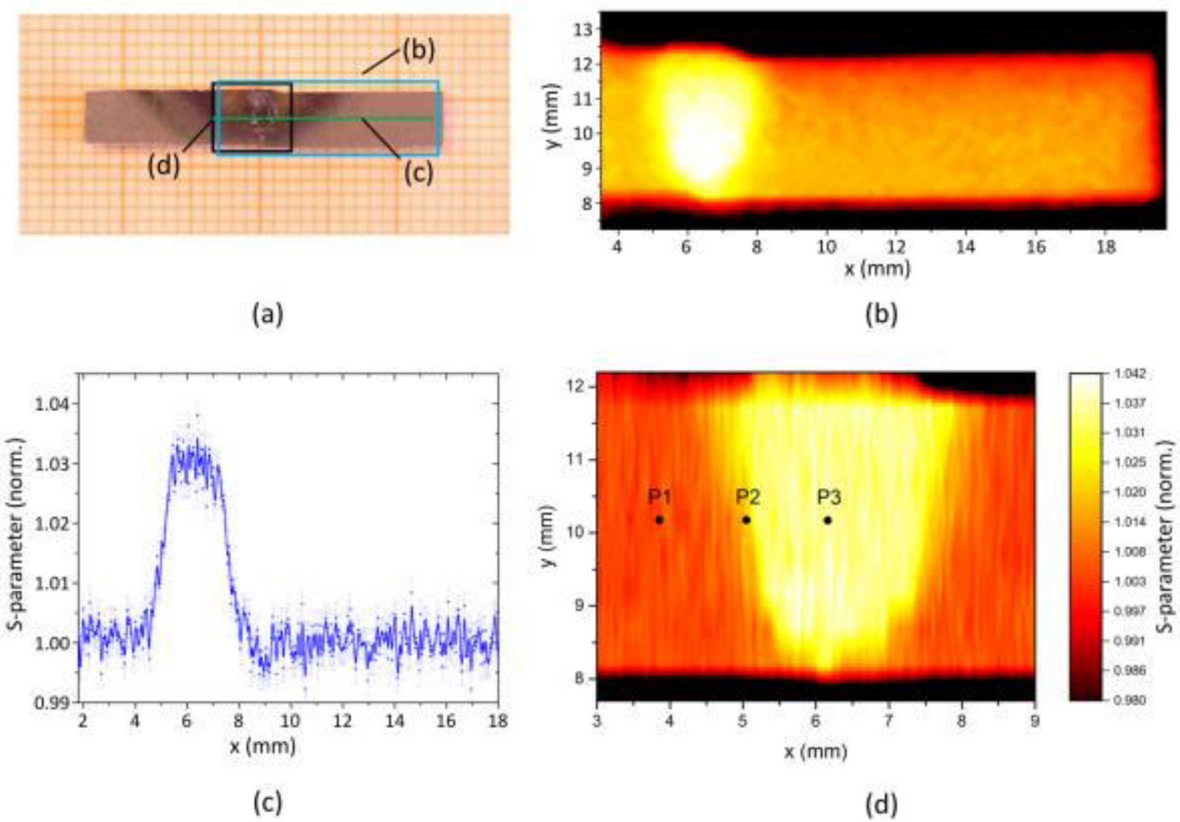
- Single-mode laser (IPG YLR-3000)
- Laser power: 2.6 kW
- Spot size: 50 µm
  Oscillating with 200 Hz, 0.2 mm amplitude
- Welding speed: 100 mm/s.

To be studied:

- Improvement of mechanical stability of joint
- What about (point) defects?
- Spatial distribution of precipitates?

Defect Mapping on a LBW

Sample: Laser beam weld (LBW) of **AlCu6Mn** (EN AW 2219-T87)



NEPOMUC rem. Beam
Overview **2D map**
Δx,y = 200 µm

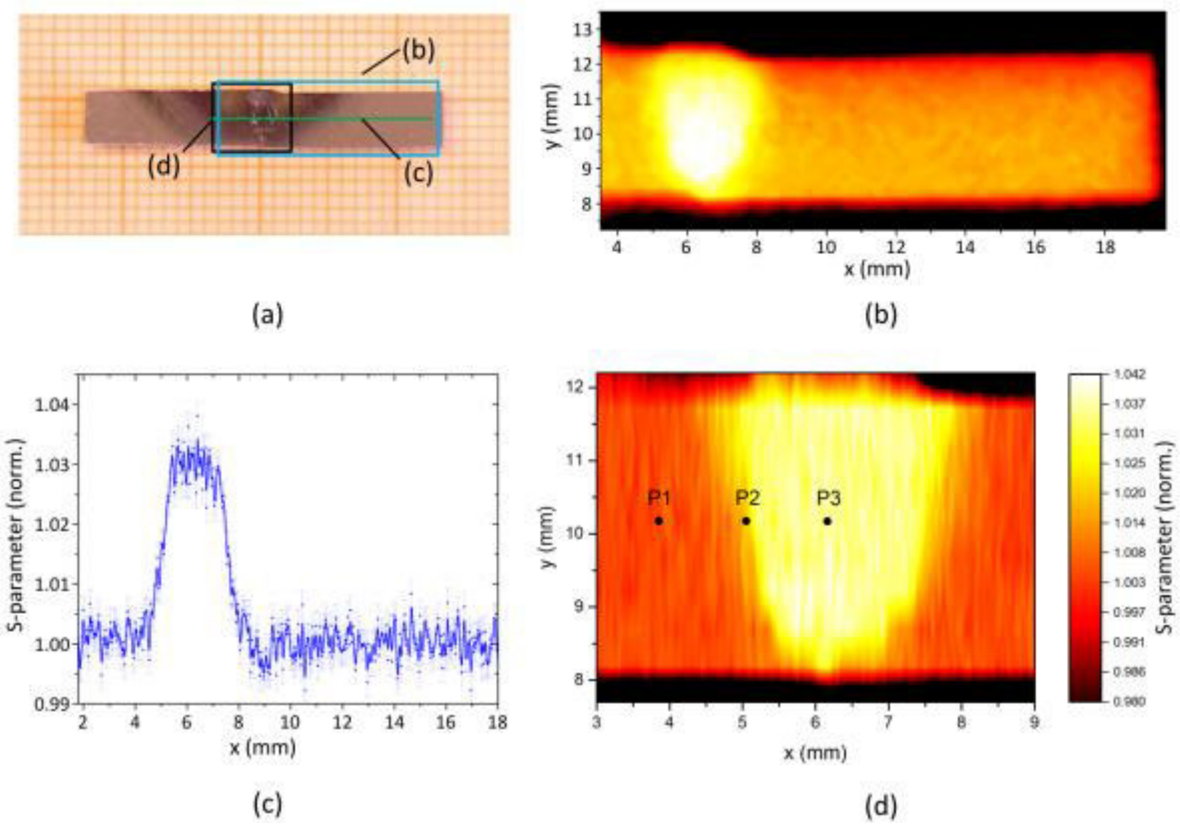
Positron microbeam
High resolution **line scan**
y = 10 mm, Δx = 50 µm

Positron microbeam
High resolution **2D map**
Δx = 50 µm, Δy =500 µm

T. Gigl, L. Beddrich, M. Dickmann, B. Rienäcker, M.Thalmayr,
S. Vohburger, and C. Hugenschmidt, New J. Phys. 19 (2017) 123007

Defect Mapping on a LBW

Results:



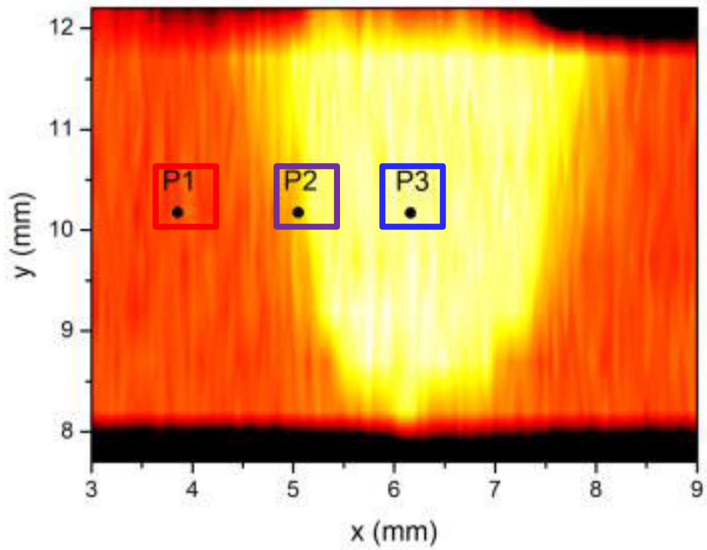
Gradient of S(y) outside LBW
→ defect gradient generated during **cold-rolling**

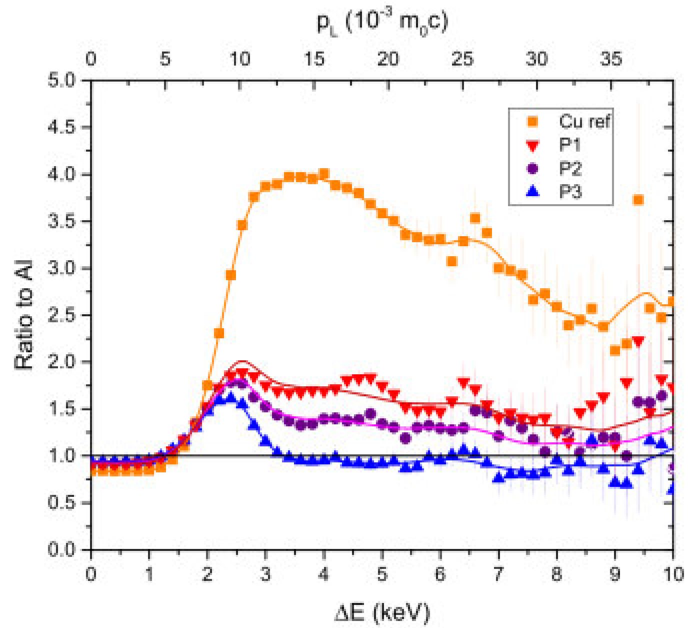
Sharp transition at LBW edges
→ well localized **small** heat affected zone (**HAZ**)

Drastic **increase of S in LBW**
→ creation and quenching of a large amount of **vacancy-like defects**

T. Gigl, L. Beddrich, M. Dickmann, B. Rienäcker, M.Thalmayr, S. Vohburger, and C. Hugenschmidt, New J. Phys. 19 (2017) 123007

CDBS at LBW of AlCu6Mn





P1, P2

- Significant contribution of Cu signature
  → positron trapping at **Cu rich precipitates**;
  in agreement with artificially age-hardened precipitates (Θ phase, Al_2Cu phase)

P3

- Disappearance of Cu signature
  → melting of Cu rich phases + rapid cooling
  → formation of **supersaturated solid solution**
- Appearance of confinement peak:
  → presence of **vacancies** (& V_{Al}-Cu?)

T. Gigl, L. Beddrich, M. Dickmann, B. Rienäcker, M.Thalmayr, S. Vohburger, and C. Hugenschmidt, New J. Phys. 19 (2017) 123007

Conclusion: Al Alloys & YBCO

LBW of Al Alloys:

- **2D defect imaging** within short time and <50µm resolution
- **Identification of Cu precipitates**

High T_c-Superconductors

- Positron ideal **probe for oxygen deficiency** δ in YBCO
- Non-destructive **spatial resolved determination of δ and T_c !**

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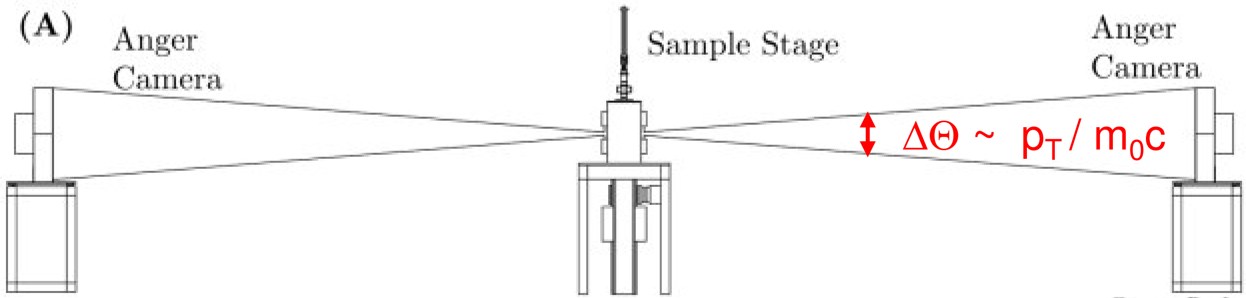
ACAR

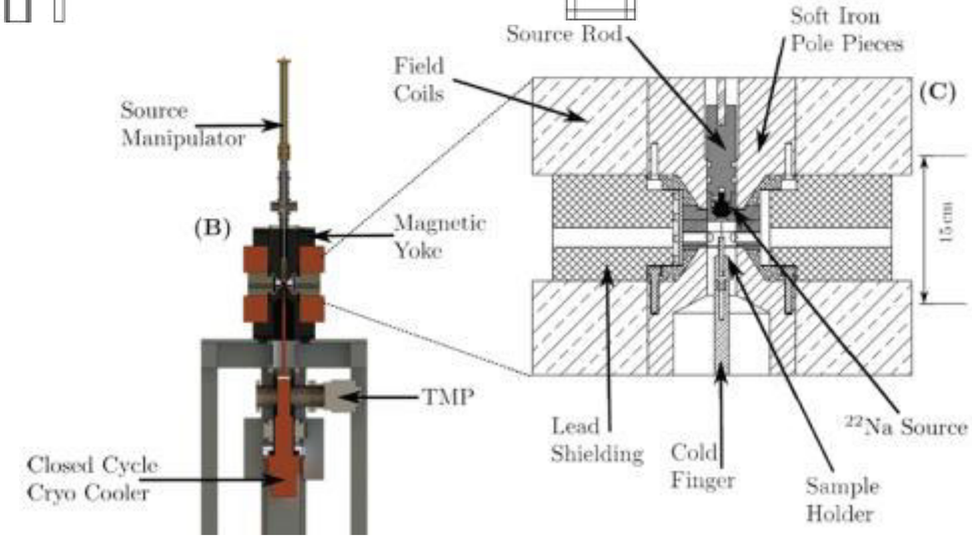
Angular Correlation of Annihilation Radiation

2D-ACAR spectrometer at TUM:



Applications & features:

- Determination of electronic structure
  → **Anisotropy of Fermi Surface**
- Special conditions **not** required
  → **T >> 0, no B-field**
- β^+ source
  → **spin-polarized ACAR**



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

Future:

- e^+ beam → surface, interface, thin layers, 2D electron systems…

2D-ACAR: Principle

Electron momentum density

$N(p_x, p_y)$: 2D projection of 3D electron momentum density

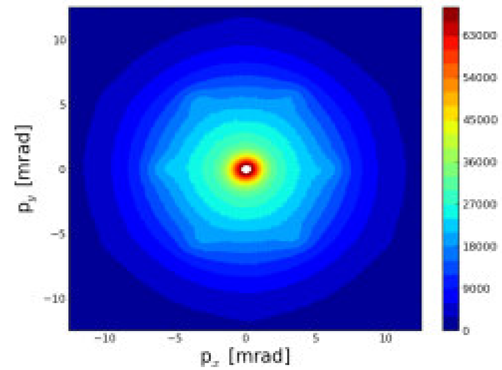
$$N(p_x, p_y) \propto \int \mathrm{d}p_z \sum_{i=\mathrm{occ.}} \sum_j \left| \int \mathrm{e}^{-i\mathbf{r}\,\mathbf{p}} \psi^+(\mathbf{r}) \psi^-_{i,j}(\mathbf{r})\, \mathrm{d}\mathbf{r} \right|^2$$

- product of positron and electron wavefunction
- in momentum space
- sum over all occupied states i in all bands j
- projecting allong the longitudinal component

2D-ACAR: Principle

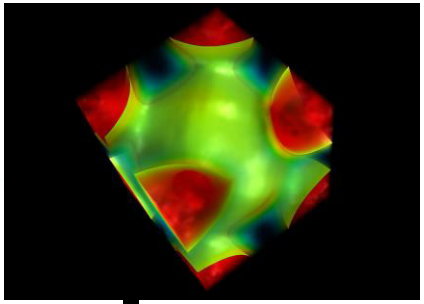
- 2D-ACAR spectrum:

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

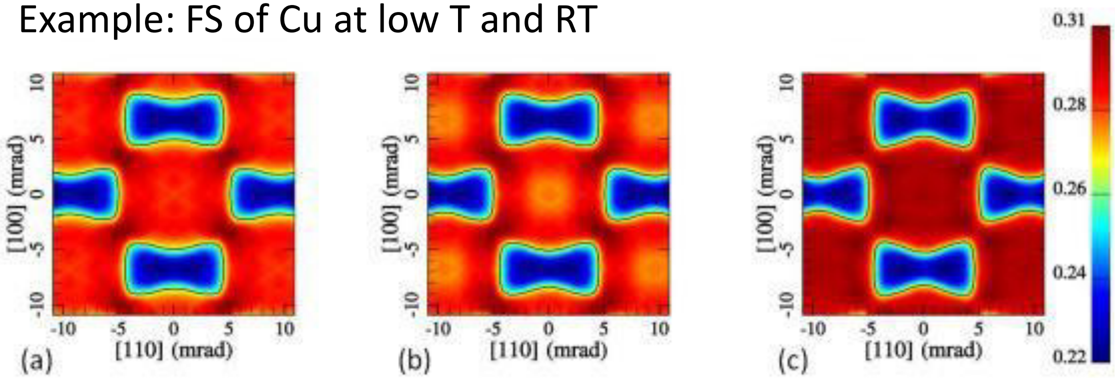


- Backfolding into first Brillouin zone

- **3D-reconstruction** of FS from 2D-projections
- **2D-Cuts** through FS



Example: FS of Cu at low T and RT



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

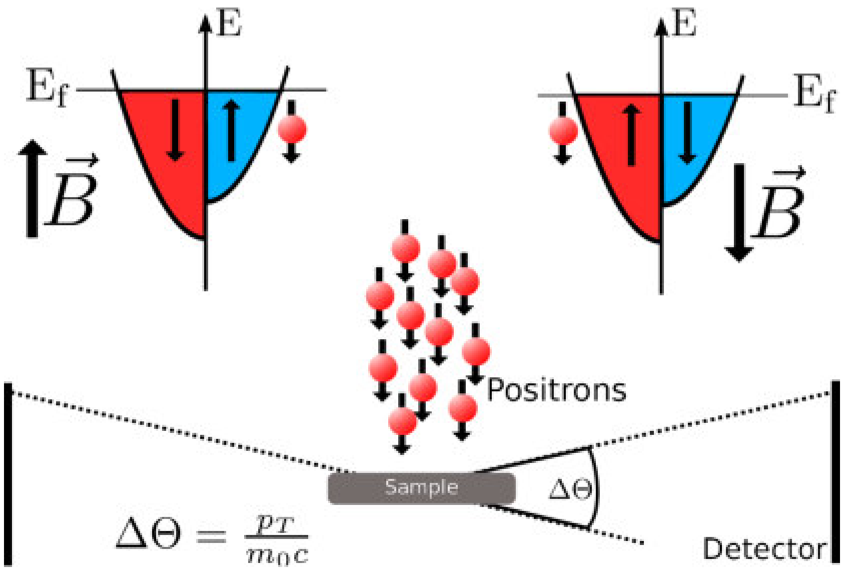
J. A. Weber et al.
J. Phys. Conf. Ser., 443(2013) 012092

Comparison with theory

Spin-Polarized ACAR

- Beta-decay → right-handed positrons
- Singlet 2γ-annihilation strongly favored
- Change magnetization of the sample

  → positron predominantly probes either of the two spin bands

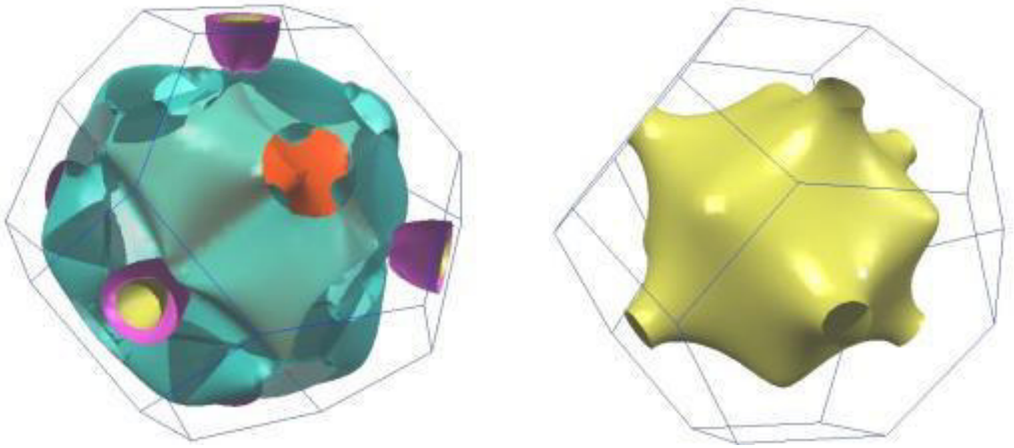


- Look at difference spectrum ⇧ - ⇩ : $N_{\pm}(p_x, p_y)$

Electron Correlations in Nickel

Nickel

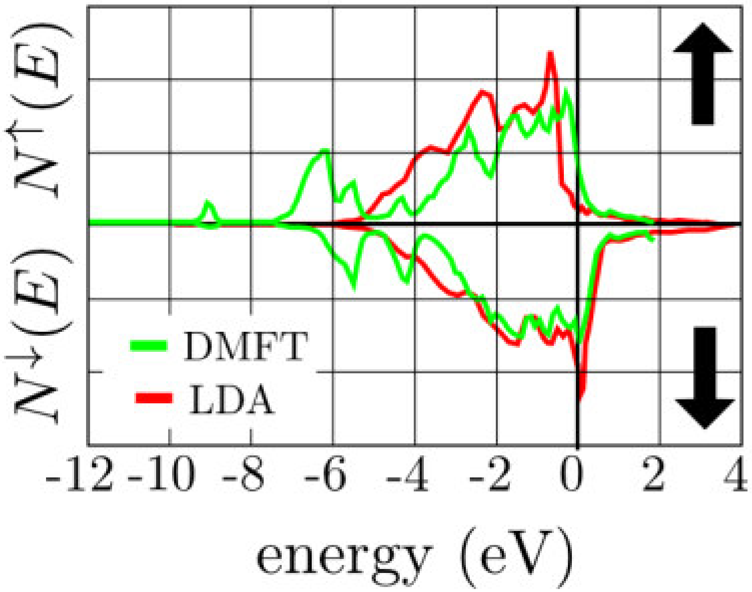
- Magnetic FCC metal
- One unpaired 3d-electron
- "simple" test case for theory and experiment



http://www.phys.u.edu

Theory

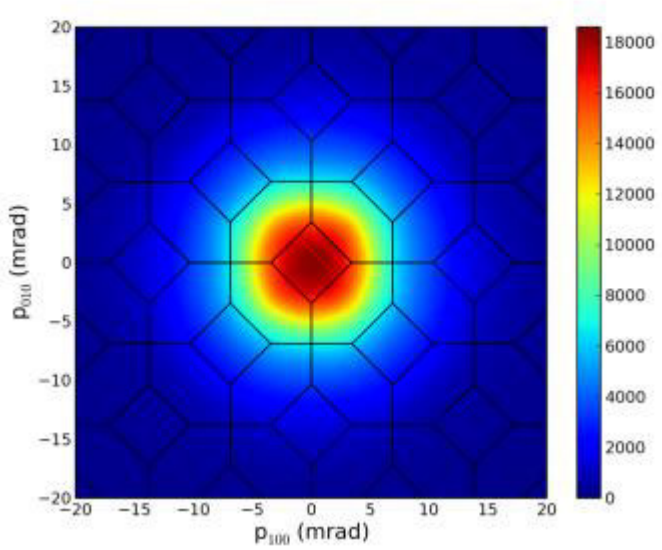
- 6eV satellite peak arises when correlations are included (DMFT)
- Electron states are relocated
- Effect of correlation change the appearance of the Fermi surface



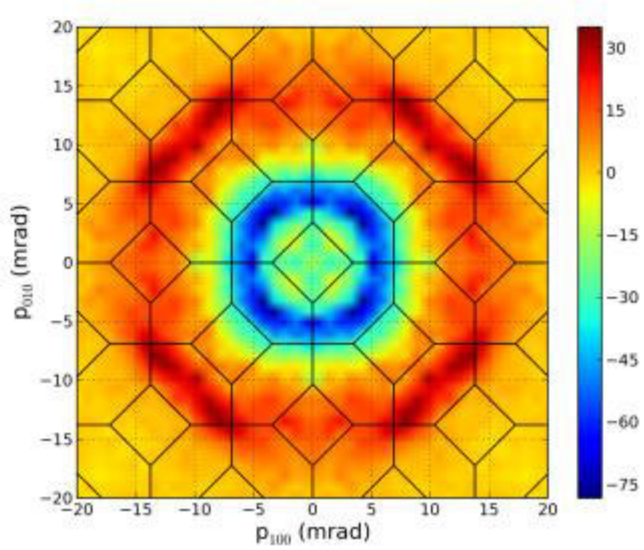
J. Kolorenc et al. Phys. Rev. B 85, 235136 (2012)

Nickel: 2D-ACAR Results

Raw spectrum

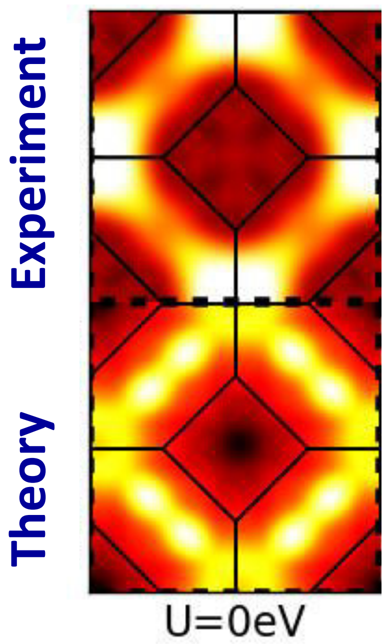


Difference



- Integration direction along <100>: 4-fold symmetry
- Magnetic difference spectrum exhibits the same symmetry

Nickel: Comparison with Theory

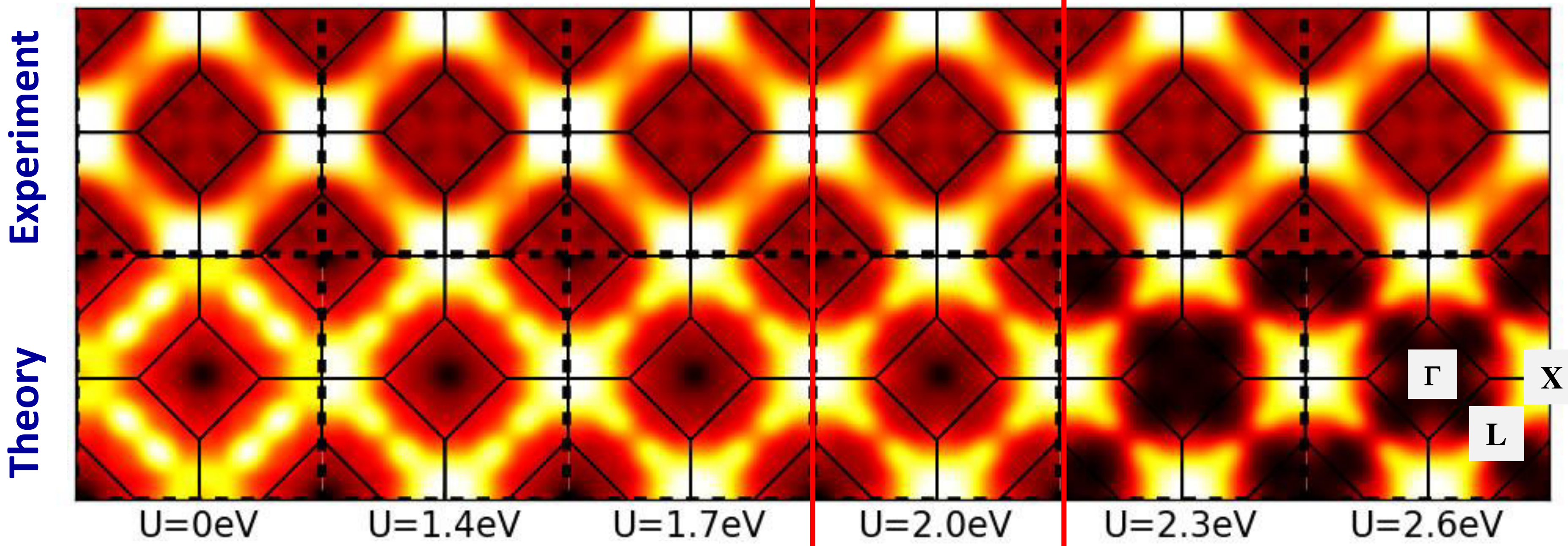


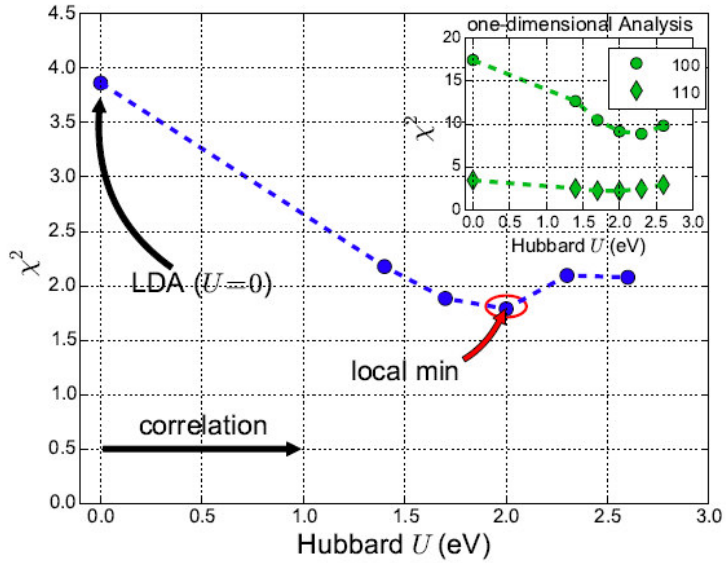
← LCW folded magnetic 2D projection along <100>

← Theory w/o e-e- correlation (convoluted with exp. resolution)

- Significant effect due to electronic correlations

Electron Correlation Strength in Nickel





Effect of correlations in Nickel

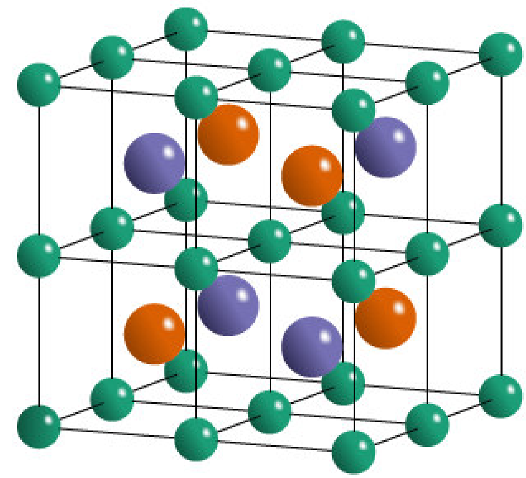
- Electron-electron correlation strength:
  U = 2.0(1) eV
- d-bands pushed below the Fermi level

H. Ceeh, J. A. Weber, P. Böni, M. Leitner, D. Benea, L. Chioncel, H. Ebert, J. Minar, D. Vollhardt, C. Hugenschmidt, Sci. Rep. 6, 20898 (2016)

Electronic Structure of Cu_2MnAl

Heusler compounds

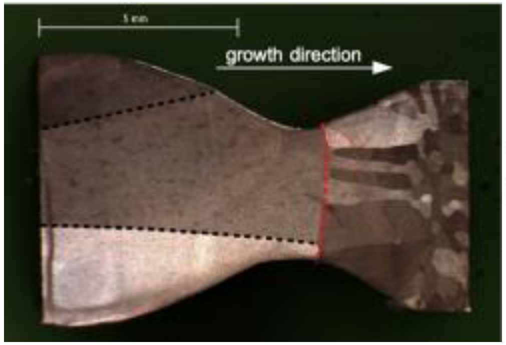
- F. Heusler 1903: ferromagnetic **Cu_2MnAl**
- Formula **X_2YZ** (Space group $L2_1$)
  X, Y transition metals
  Z non-magnetic/non-metallic element
- Large variety of electronic ground states:
  Insulating, (half-)metallic, semiconducting, ferromagnetic, superconducting



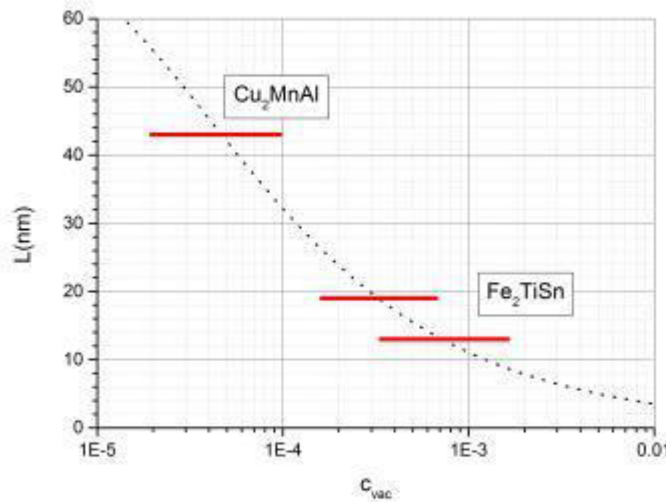
Single crystalline Cu_2MnAl

- Demanding to produce "large" & defect free samples
- Samples grown by optical floating zone

→ A. Neubauer, A. Bauer, C. Pfleiderer



A. Neubauer et al. NIM A 688 (2012) 66

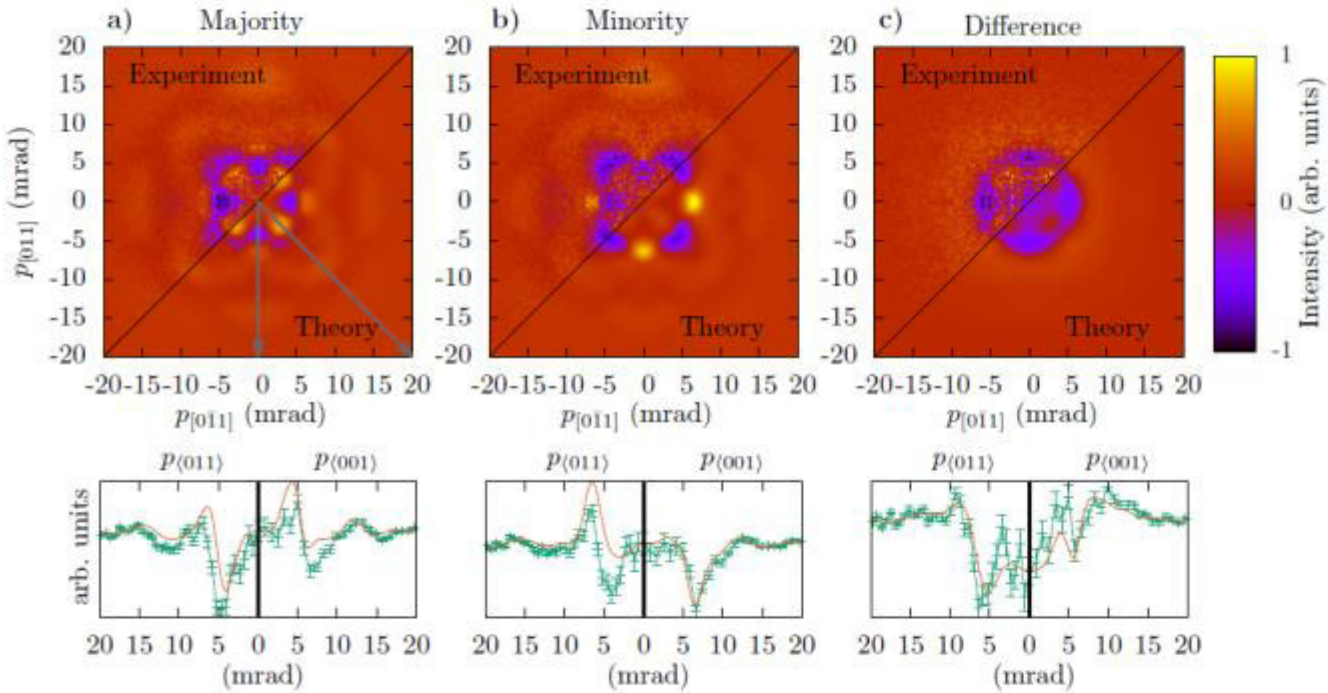


C.H. et al. Appl. Phys. A 119 (2015) 997

Spin-Polarized 2D-ACAR on Cu_2MnAl

Cu_2MnAl

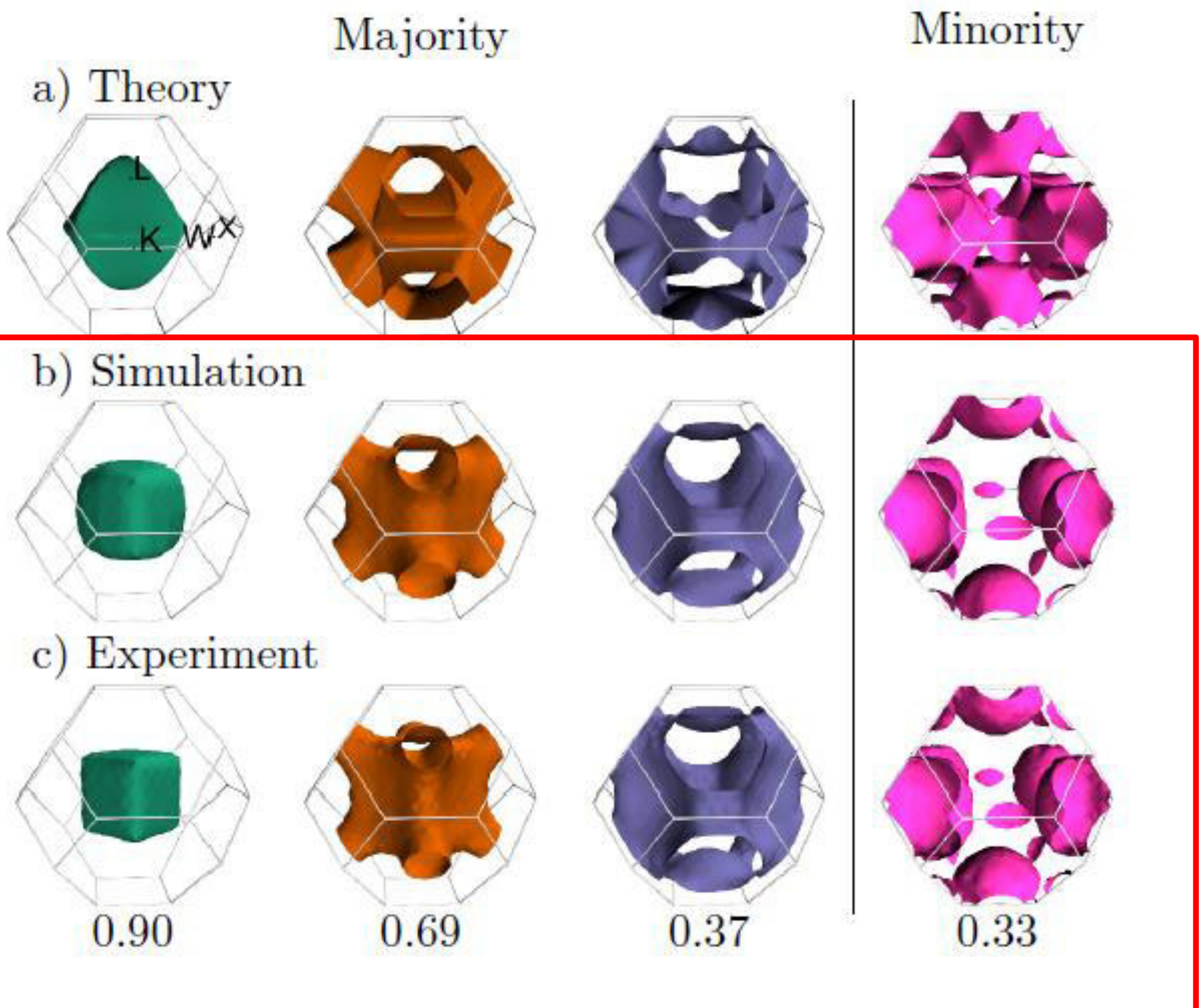
2D projections along [100] at RT → anisotropy of maj., min. density & difference



- Excellent agreement between theory and experiment
  → 3D reconstruction of Fermi sheets for individual spin channels

J. Weber, A. Bauer, P. Böni, H. Ceeh, S. B. Dugdale, D. Ernsting, W. Kreuzpaintner, M. Leitner, C. Pfleiderer, C. Hugenschmidt; PRL 115 (2015) 206404

Cu_2MnAl: 3D Reconstruction of Fermi Surface



Results

- Contribution of each individual Fermi sheet to magnetization !
- Total magnetization: 3.6(5) μB/f.u.

J. Weber, A. Bauer, P. Böni, H. Ceeh, S. B. Dugdale, D. Ernsting, W. Kreuzpaintner, M. Leitner, C. Pfleiderer, C. Hugenschmidt; PRL 115 (2015) 206404

Summary

- **World-class positron beam at NEPOMUC**
  - → ... & fundamental research

What about

even higher intensity?

- spin-polarized -

- **CDBS with scanning po...**
  - → spectroscopy and imaging of **defects**

- **Spin-resolved 2D-ACAR for electronic structure determination**

Evolution of the FS

from the surface to the bulk

- spin-resolved -

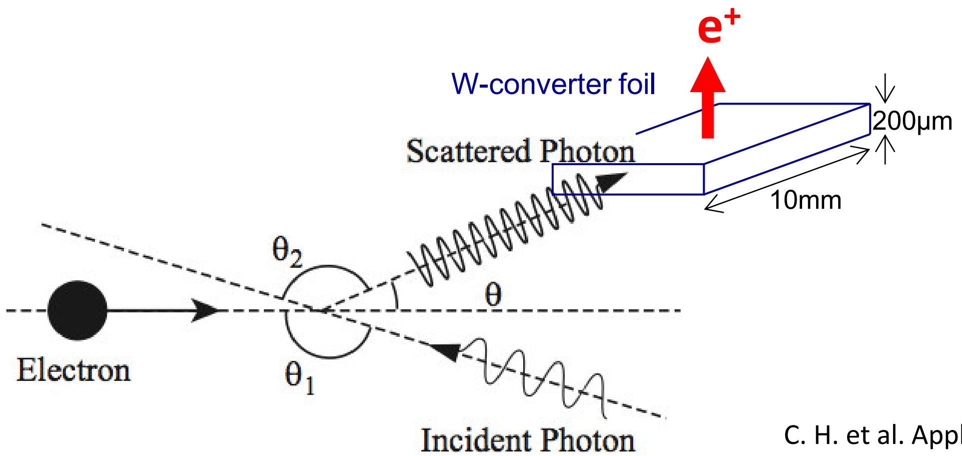
- **Experiment...**
  - → ... Al alloy
  - → Determination of **correlation strength** in Ni
  - → Revealing **spin-resolved Fermi sheets** in Cu_2MnAl

Plans for a Next-Generation Positron Beam

Inverse Compton Scattering & Positron Production

$\nu_{sc} \sim \gamma^2 \nu_0$

$\rightarrow \gamma \sim 1000$



C. H. et al. Appl. Phys. B 106 (2012) 241

Features:

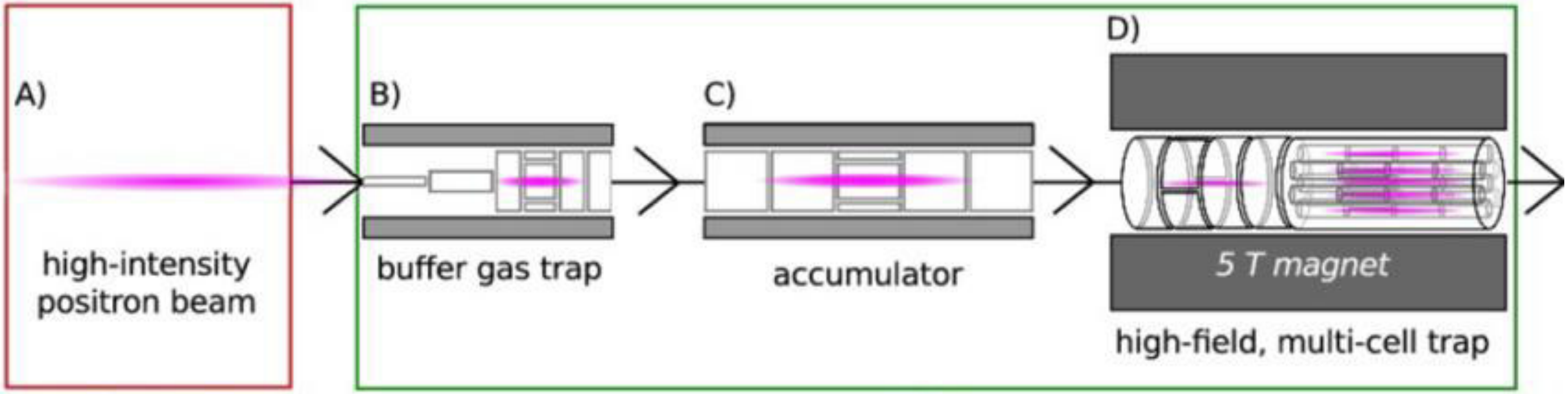
- Tiny diameter of γ beam → **high brightness**
- Narrow band width → no γ's with $E < 2mc^2$
- **Polarization** → spin-resolved positron beam experiments

ELI-NP positron source project:

- Flux of moderated positrons: $1\text{-}2\cdot10^6$ e+/s
- Degree of polarization: 31-45%

N. Djourelov et al. Rom. Rep. Phys. 68 (2016) S735 EU: ELI-NP/TDR/RA2/G2P/3/Jan2015

Ultra-Dense Positron Pulses



A) **NEPOMUC**: continuous positron beam (primary or remoderated)

B) **Cooling & trapping** in buffer gas trap

C) **Accumulation** in UHV

D) **Collection** and accumulation in multi-cell trap

- **Aim: Pulse of 10^9 e+ within few ns**

DFG TUM/IPP (HU 978/15-1)
DFG Uni Greifswald/IPP/TUM (HU 978/16-1)
ERC advanced grant, T. Pedersen, IPP: “Pair plasma”

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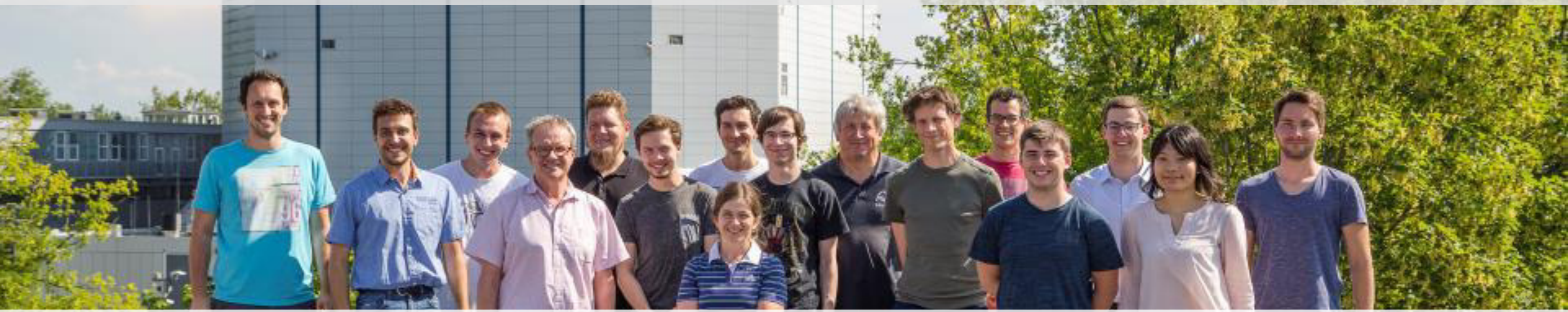

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