Soft-X-Ray ARPES View of Three-Dimensional Electronic Structure

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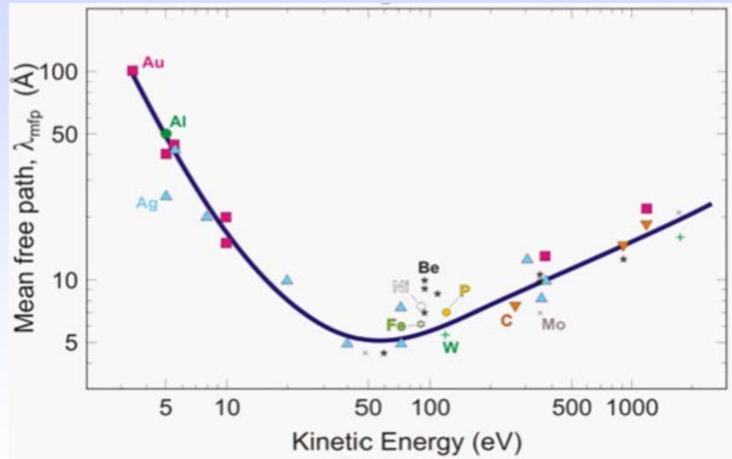
Outline:

1. Why ARPES in the soft-X-ray range?
   - applications to 3D systems
2. Instrumentation
3. Results

- 3-dim band structure and Fermi surface of quasi-2D VSe_2
- overview: HTSC pnictides; heavy-fermion intermetallics; fermiology of $NiS_{2-x}Se_x$, buried layers in LNO/STO heterostructures …

Why Soft-X-Ray ARPES?

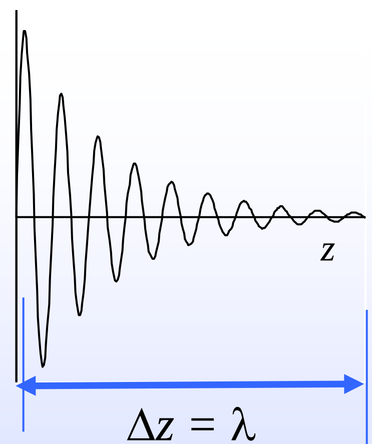
Virtue 1: Increasing λ

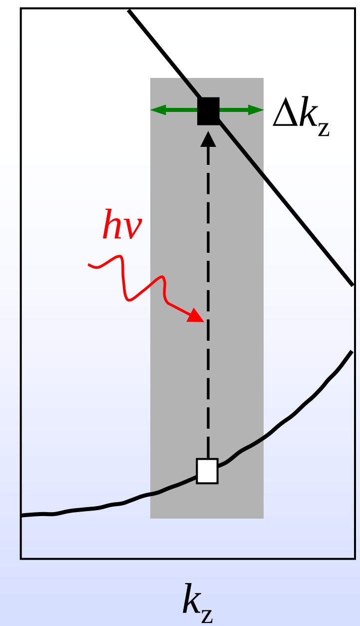


- increasing bulk sensitivity

⇒Virtue 2: Improving *intrinsic resolution* $\Delta k_z = \lambda^{-1}$

- ARPES signal by averaging $E(k_z)$ over Δk_z
- crucial for 3D systems



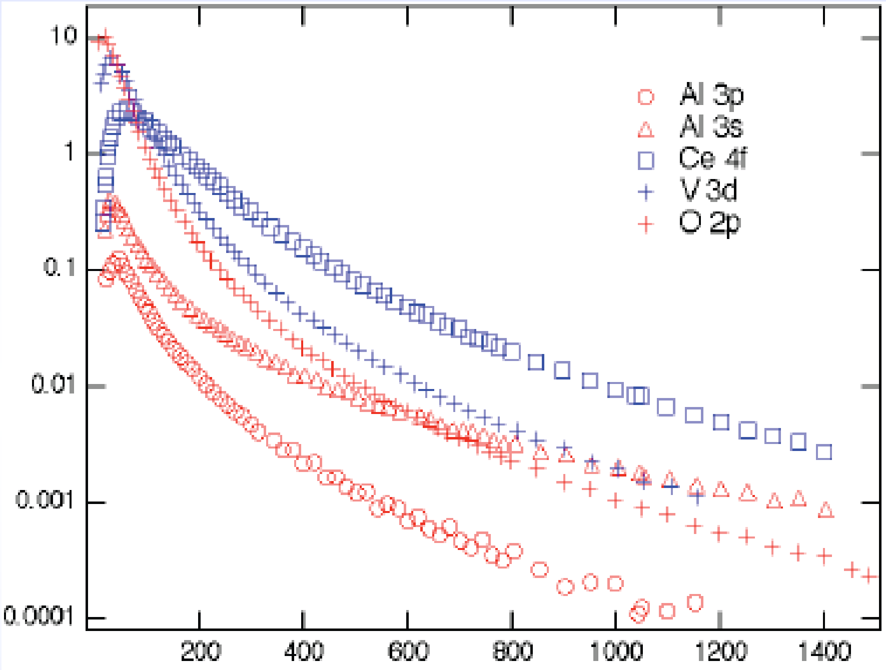


Virtue 3: Free-Electron Final States

- free-electron final-state $E(k_z)$ to find $k_z = \sqrt{\frac{2m}{\hbar^2}\left(E - V_0 - \frac{\hbar^2 k_{//}^2}{2m}\right)}$

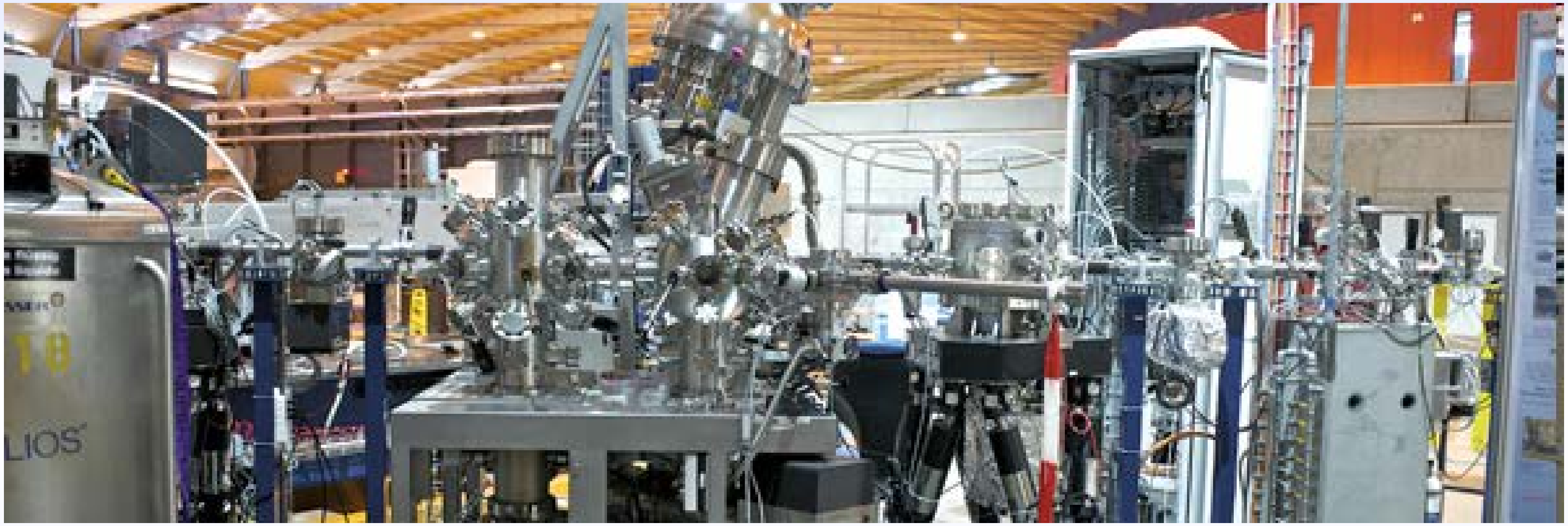
Further virtues: Simplified matrix elements, …

Crossection Problem of SX-ARPES



- dramatic drop of valence band crossection with *hv* (especially for *s*- and *p*-states): **photon flux required!**

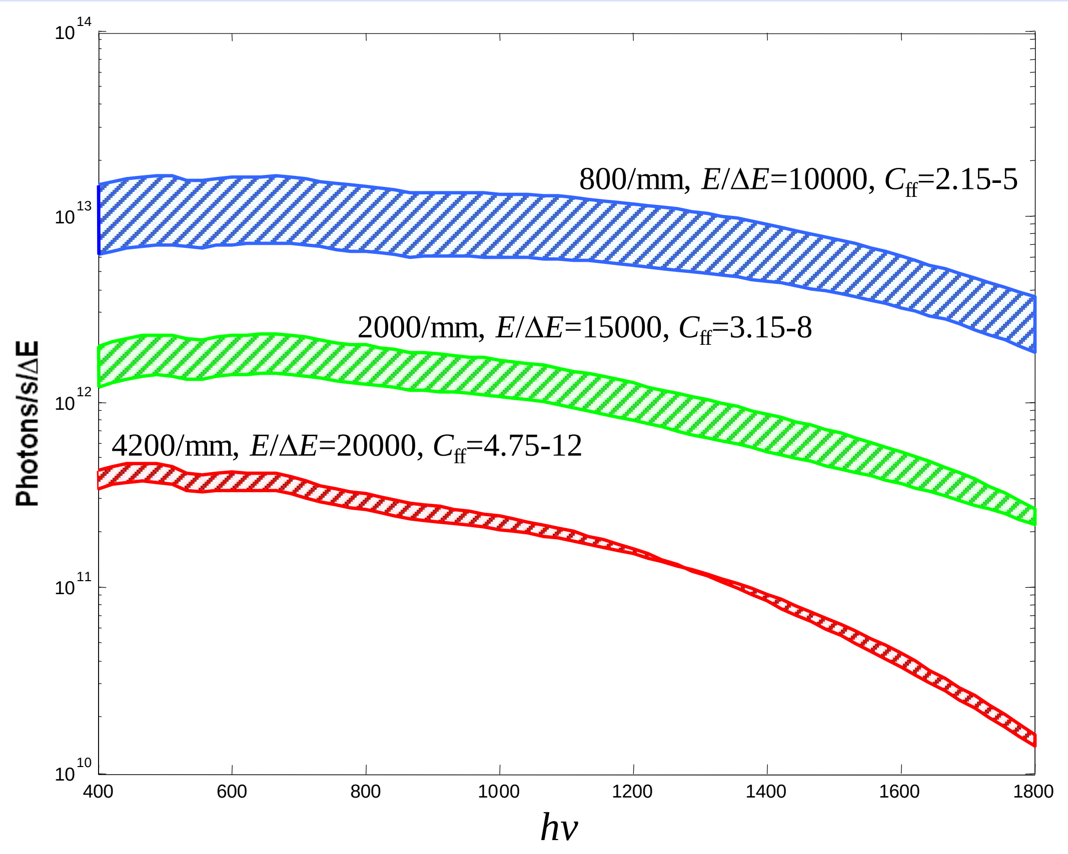
ADRESS (*ADvanced RESonant Spectroscopies*) Beamline at SLS



- RIXS and ARPES endstations
- soft-X-ray radiation with circular and 0-180° variable linear polarizations
- energy range 300 – 1800 eV
- high resolution ΔE ~ 30 meV @ 1 keV
- collimated-light PGM optical scheme
- **flux up to 10^{13} ph/s/0.01%BW: factor of 10 to 100 increase compared to best available beamlines ⇒ breakthrough of the crossection problem**

ADRESS Beamline: Flux Parameters

- flat energy dependence with all gratings including 800/mm blazed
- 3×10^{11} to 1×10^{13} ph/s/0.01%BW (factor of 10 to 100 flux increase compared to BL25SU@SPring-8)



- excellent flux by virtue of (1) 2.4 GeV ring optimal for soft X-rays; (2) glancing angles on the mirrors; (3) blazed/lamellar and profile optimization of gratings; (4) optimal C_{ff}

⇒ ADRESS is ideal for the photon-hungry SX-ARPES

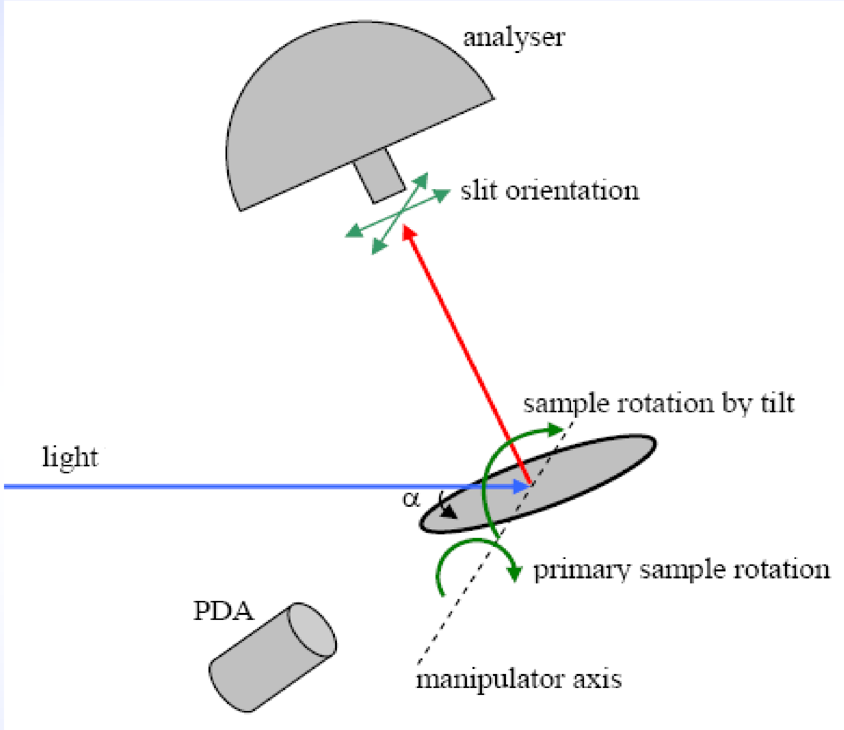
SX-ARPES Endstation @ ADRESS

- Geometry

• grazing incidence at 20° to increase photoyield (factor of 2 compared to 45°)

• horizontal rotation axis to balance the vertical (<20μm) and horizontal (74μm) light footprint

• 2 operation modes:
- analyser slit // beam + tilt (selection rules)
- analyser slit ⊥ beam + primary rotation



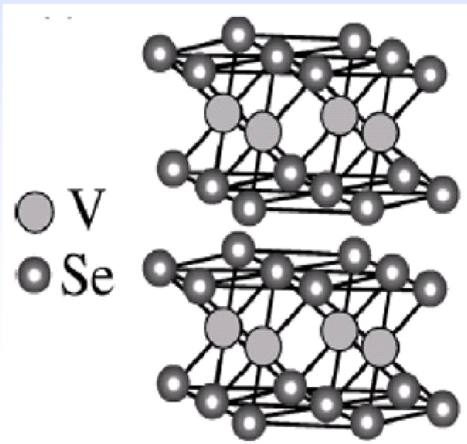
- Instrumentation

• CARVING™ manipulator:
- 3 angular DOFs (res 0.05°)
- L-He_2 cooling to 10.5K

• analyser PHOIBIOS-150: $\Delta k_{//}$=0.02Å^{-1}@1keV

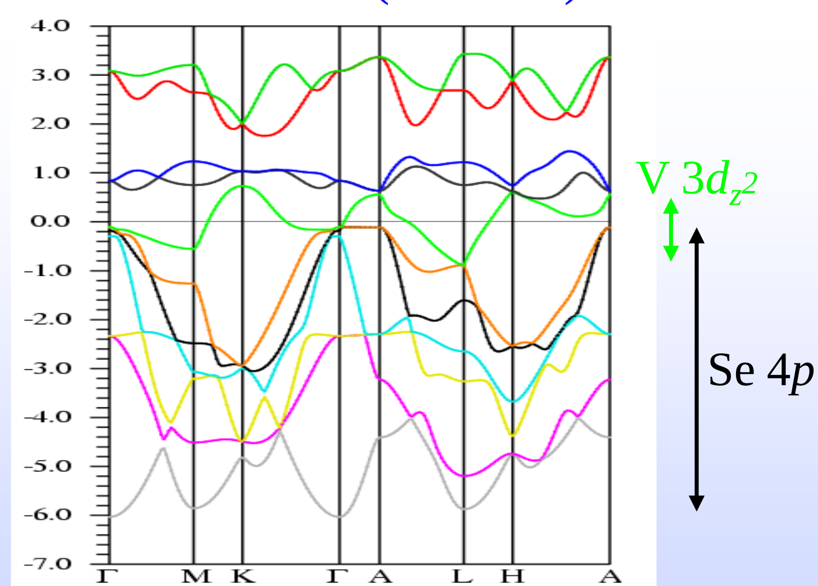
Test case: 3D bandstucture and Fermi surface of VSe_2

- Structure and electronic structure



- quasi-2D structure with weaker interlayer interaction

GGA-DFT (P. Blaha)



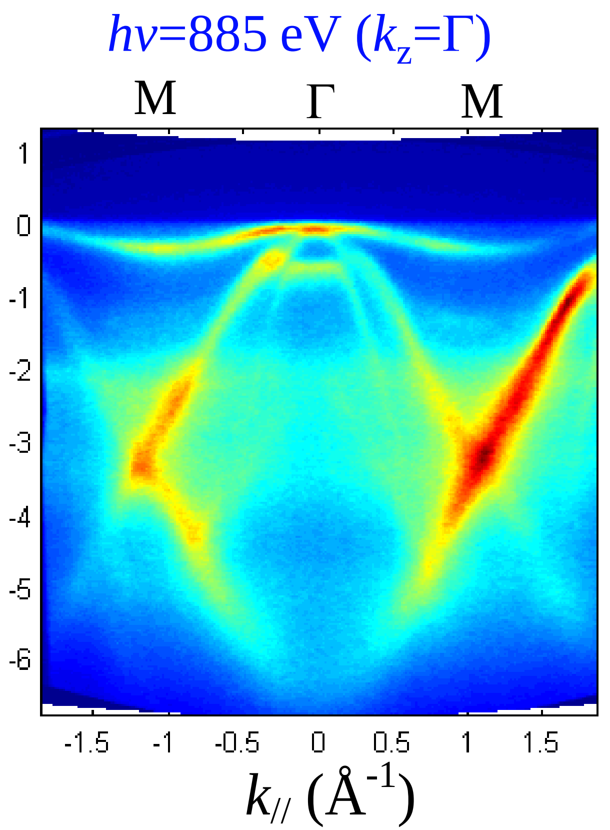
- significant 3D-lity due to V 3*d* and Se $4p_z$ dispersing along ΓA

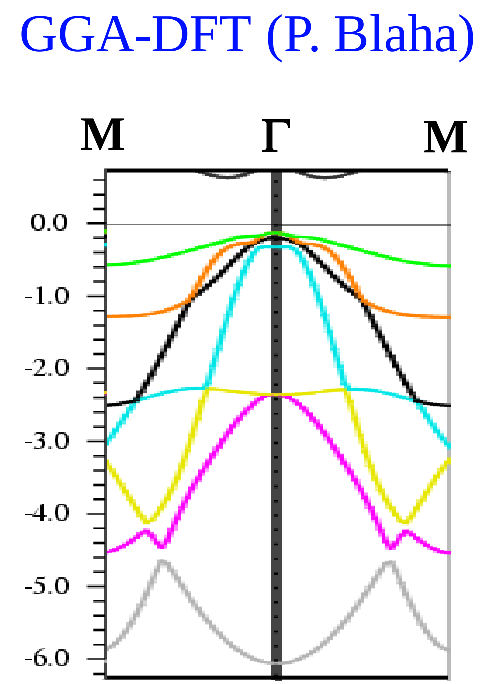
- 3D Band Structure

- T=**10.7 K**
- combined **Δ*E*~110meV**
- acquisition time **10 min**

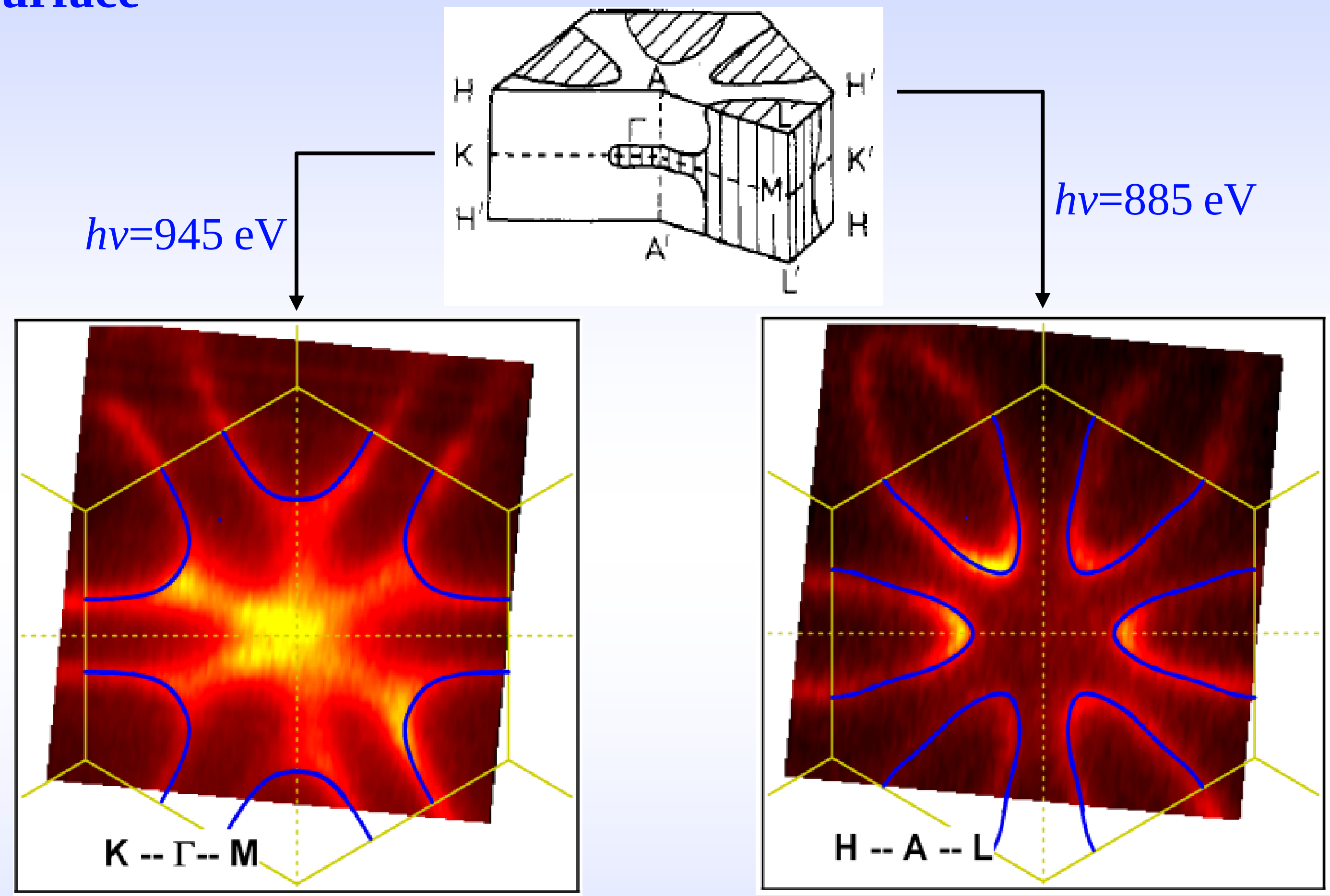
⇓

- excellent intensity not only for *d*-states, but also for *p*-states despite dramatic loss of crossection
- intense and sharp in $\mathbf{k}_{//}$ structures => Debye-Waller and phonon broadening are no prohibitive
- agreement with GGA-DFT
- evolution with *hv* => intrinsic Δk_z resolution ~ 0.05 $Å^{-1}$ (or λ ~ 20 Å)



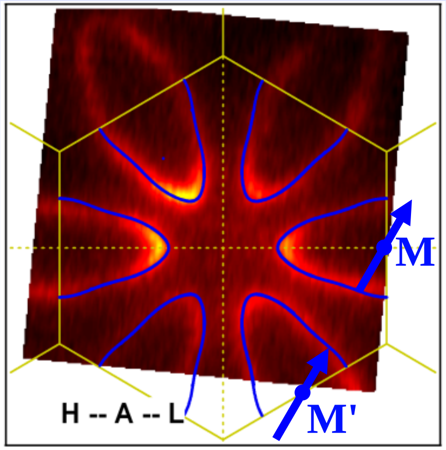


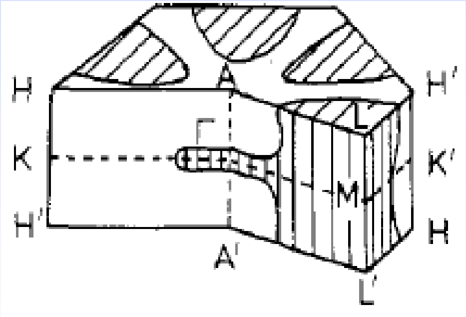
- Fermi surface



- combined ΔE~120meV; each image in 400 s; each map in < 5 hrs
- extraordinary clarity of the experimental data (no image enhancement)
- fantastic agreement with GGA-DFT, even the tiny warping in HAL

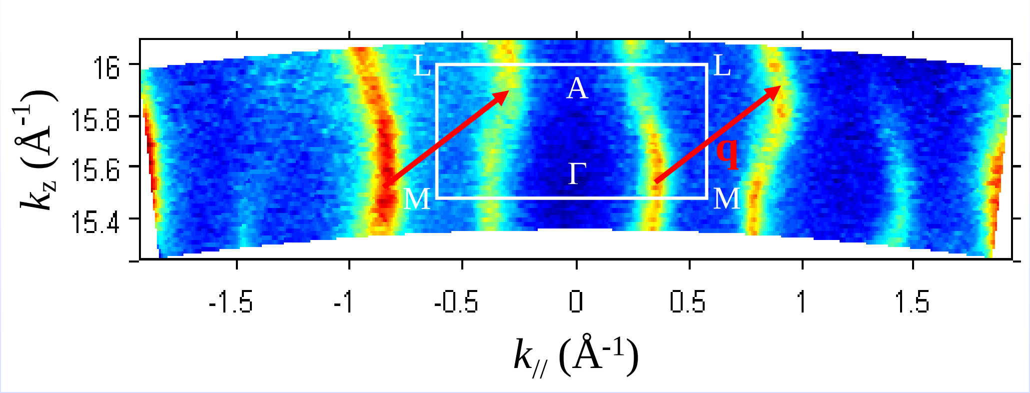
- Origin of 3-dimensional CDWs

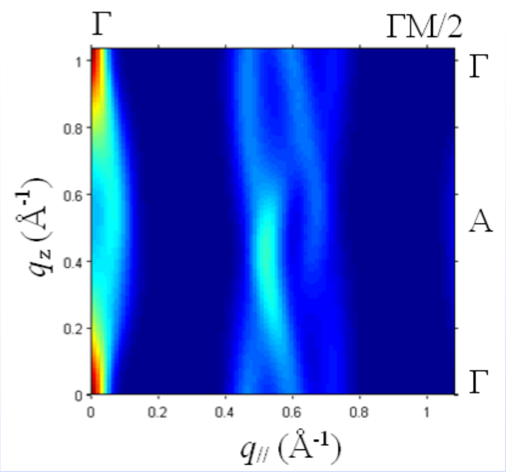




$\mathbf{q}_{CDW}=\mathbf{q}_{//}+\mathbf{q}_z$ ($q_z \sim k_z^{BZ}/3$)

- Perpendicular FS cut in MLL'M' plane



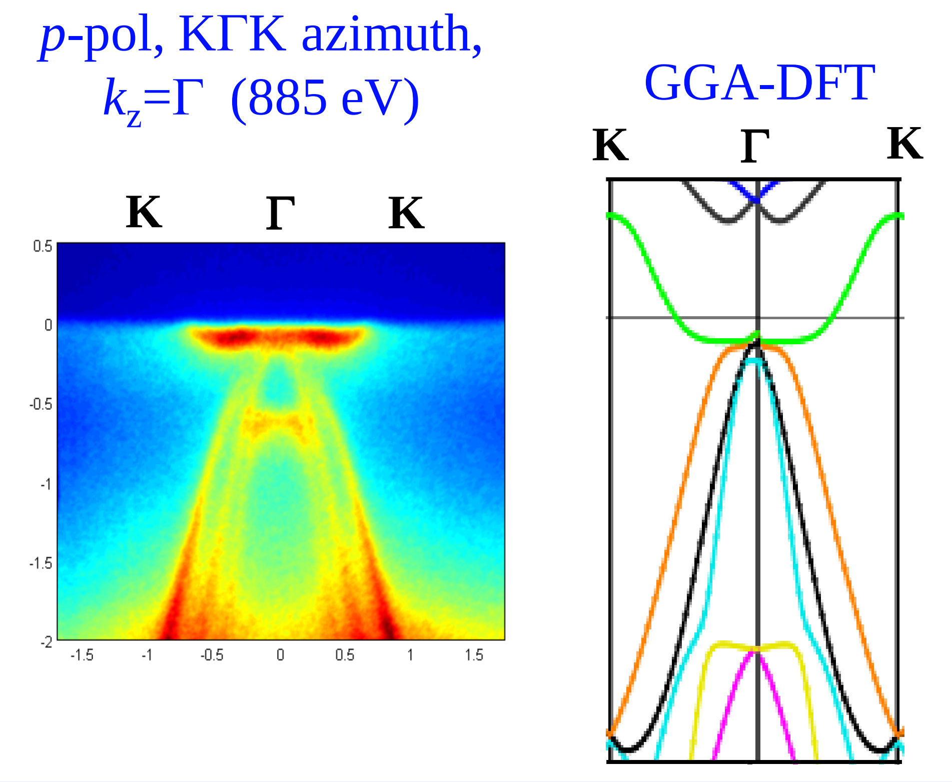


- 3D warping to support nesting with the experimental q_z

- Autocorrelation peak at the experimental q_z

Towards better resolution

- combined **Δ*E*~60meV**
- image in **40 min**
- well resolved bands

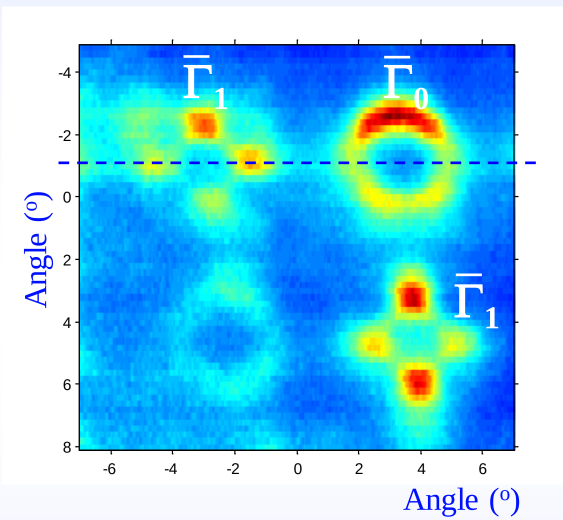


3D Fermi Surface of HTSC pnictide $Ba_{0.6}K_{0.4}Fe_2As_2$

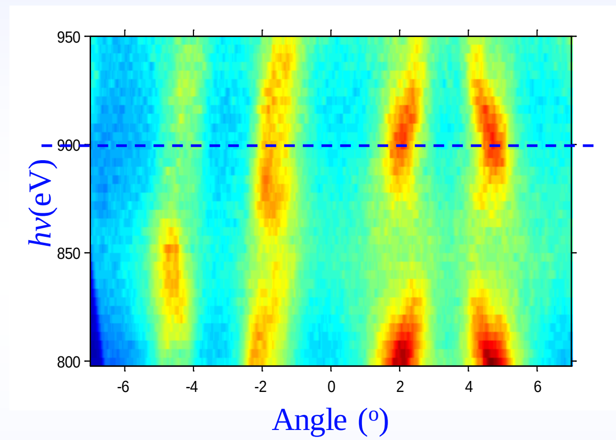
- pnictides are 3D materials

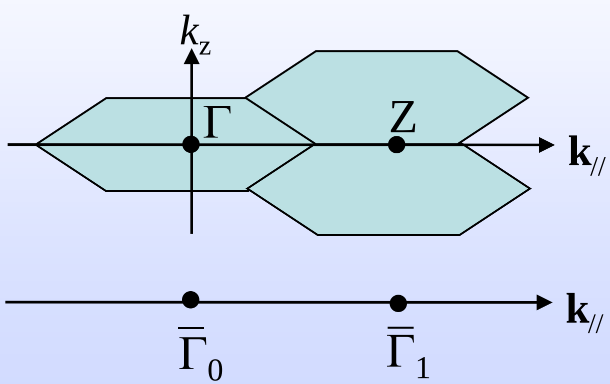
- measurements at *p*-pol, T=10.7K, ΔE~110meV

FS($\mathbf{k}_{//}$) @ $h\nu$ = 900 eV



FS(k_z) along Γ_1-Γ_2



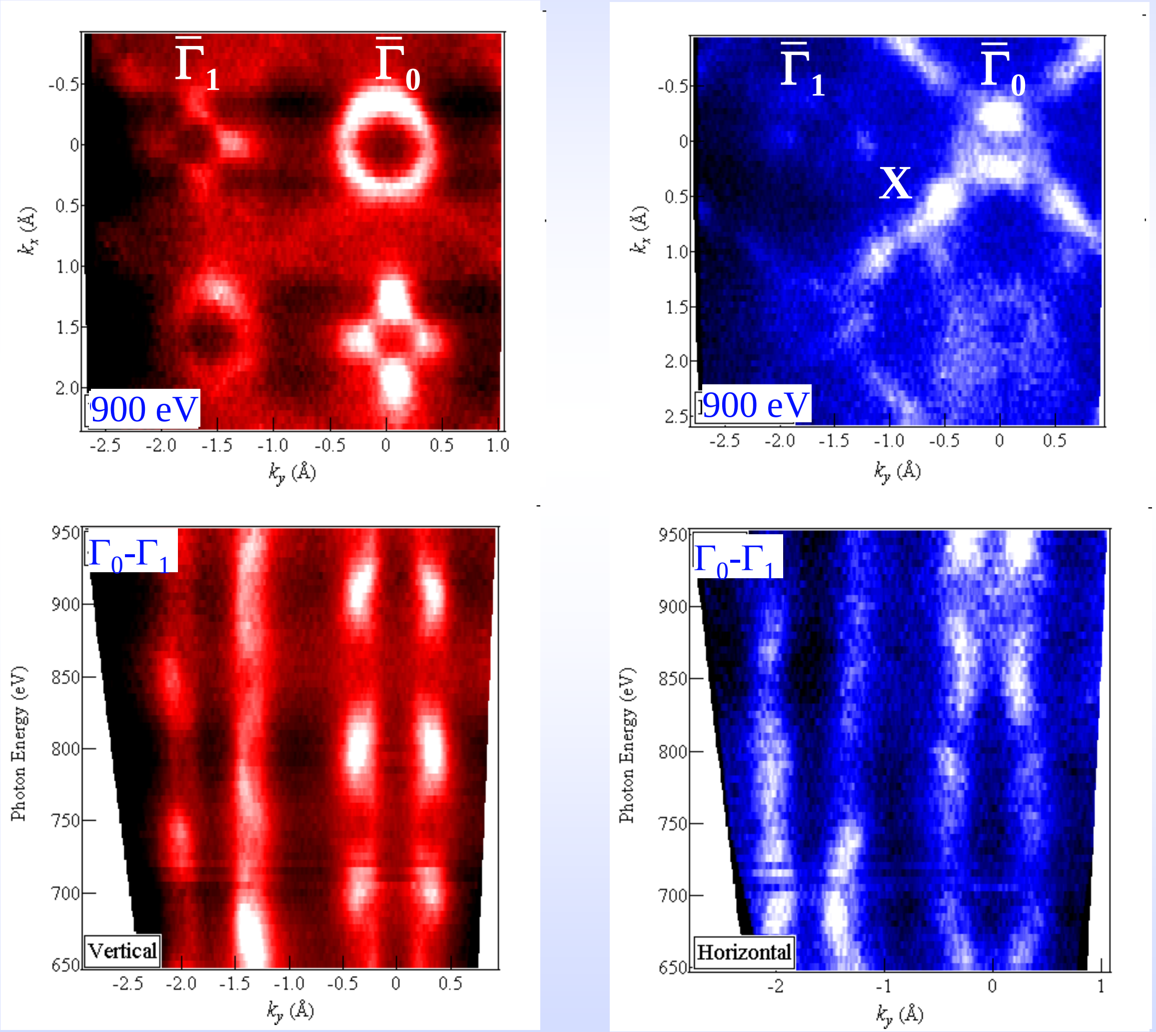


Beyond the 3-dimensionality:

- k_z dispersion along Γ_1 reduced compared to Γ_0

- linear polarization dependence

p-pol *s*-pol



- strong linear dichroism in the FS($\mathbf{K}_{//}$) and FS(k_z) maps

- Varying BZs and polarization ⇒ selective excitation of α, α' and β Fe 3*d* bands of the FS: matrix element effects due to intra-cell interference

3D bulk electronic structure of heavy-fermion $EuRh_2Si_2$

TU Dresden: M. Höppner, S. Danzenbächer, D. Vyalikh, S. Molodtsov

3D electronic structure of strongly correlated $NiS_{2-x}Se_x$

Fudan University : Y. Zhang, H. C. Xu, M. Xu and D. L. Feng

$PbBi_4Te_7$: Topological Surface State in 3D Sea

H. Dil, G. Landolt, B. Slomski, J. Osterwalder (PSI/Uni Zuerich)

k-resolved Fermi surface of $LaAlO_3/LaNiO_3$ heterostructures

Uni Wuerzburg: G. Berner, M. Sing and R. Claessen;
samples: MPI-FKF Stuttgart

Summary

• Virtues of soft-X-ray ARPES

- Enhanced λ, improved Δk_z resolution, free-electron final states, simplified matrix elements…

• SX-ARPES instrumentation at SLS

- ADRESS beamline: 300 - 1800 eV energy range, high res (ΔE~30meV@1 keV) and high flux (up to 10^{13} ph/s/0.01%BW @ 1 keV)
- Data acquisition rate of ~5 min @ ΔE~110 meV: breakthrough of the crossection problem

• Examples

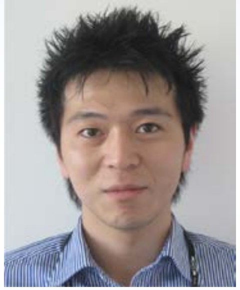
- 3D fermiology and CDWs in VSe_2: textbook clarity of 3D-resolved $E(\mathbf{k})$ and FS by virtue of free-electron final states and small Δk_z in the soft-X-ray region
- Further studies of 3D electronic structure: Band selection effects in HTSC pnictides, 3D hybridization effects in heavy-fermion intermetallics, 3D fermiology of strongly correlated $NiS_{2-x}Se_x$, buried layers in LNO/LAO heterostructures …

⇒ **high potential of SX-ARPES, in particular for 3D systems**

People

SX-ARPES team

V.N.S.
(BL Scientist)



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C. Hess
(BL Technician)

Collaborators

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(SIS beamline)

T. Schmitt
(RIXS)

L. Patthey
(Group Leader)

ADRESS

www.psi.ch/sls/adress/

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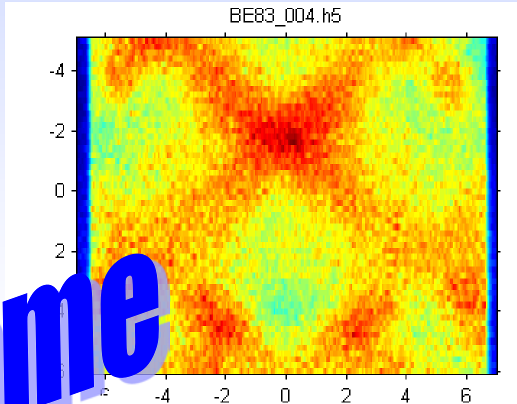
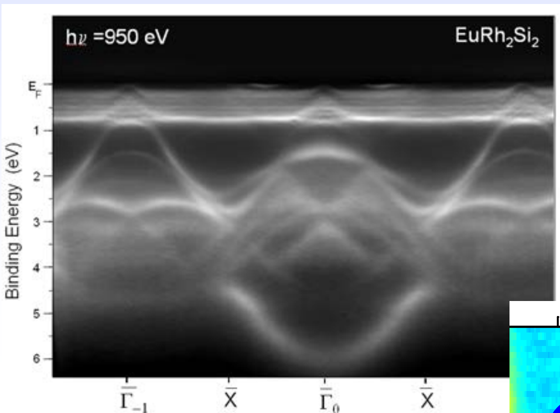
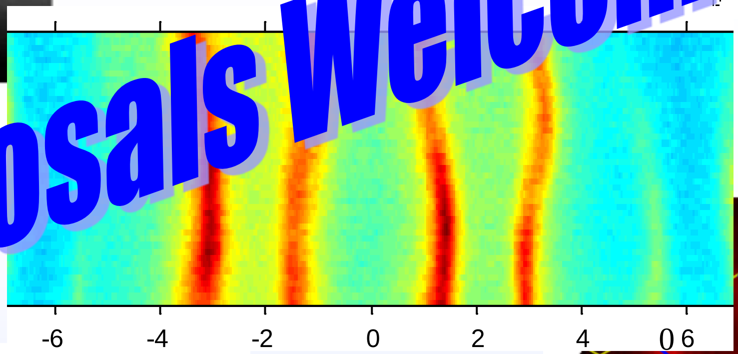
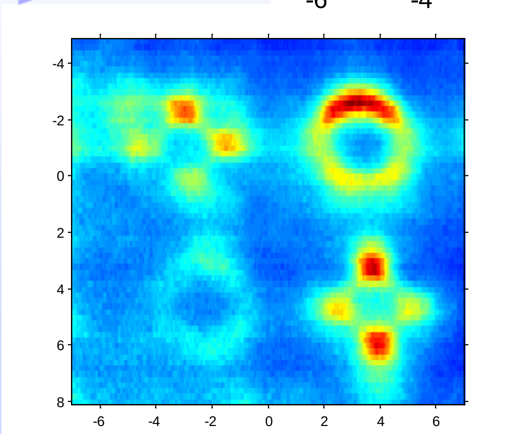
SWISS LIGHT SOURCE SLS

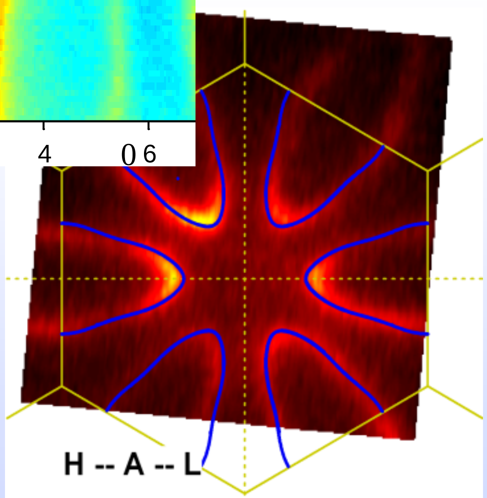






Proposals Welcome





Next call deadline: February 15