Soft-X-Ray ARPES Facility at SLS: Instrumentation and Applications to 3-Dimensional Systems

V.N. Strocov

Swiss Light Source, Paul Scherrer Insitute

Outline:

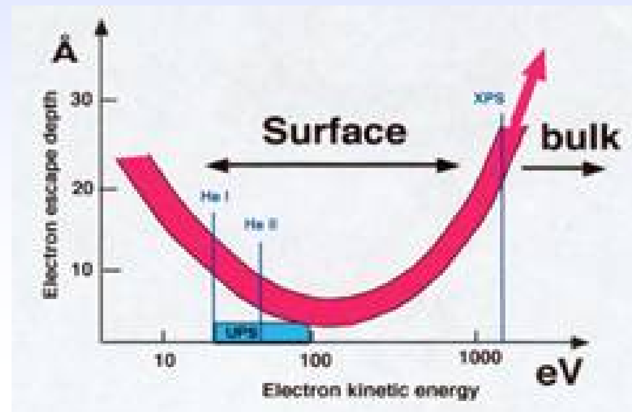
1. Why ARPES in the soft-X-ray range?
   - applications to 3D systems
2. Instrumentation:
   - ADRESS beamline
   - SX-ARPES endstation
3. First results

- test case: 3-dim band structure and Fermi surface of quasi-2D VSe_2
- overview: excitonic insulator $TiSe_2$; HTSC pnictides; heavy-fermion intermetallics; buried layers in LNO/STO heterostructures

Why going from UV to Soft-X-Ray ARPES ?

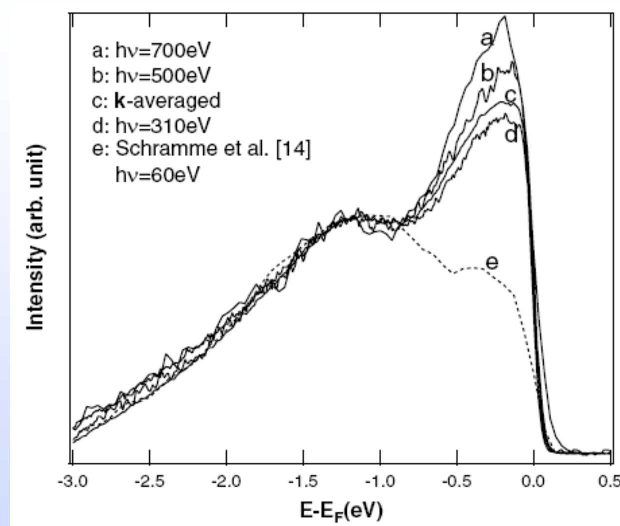
Virtue 1: Surface sensitivity

• 2-3 times increase in probing depth
⇒ through the distorted surface layer towards deeper atomic layers with bulk properties



Mott-Hubbard metal-insulator transition in V_2O_3
(Mo et al 2003)

• quasiparticle peak in the paramagnetic phase develops only in bulk

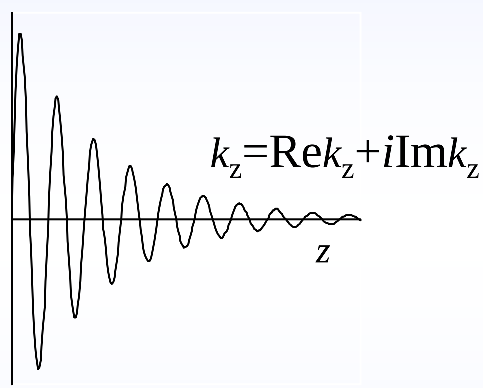


bulk sensitivity increases with *hν*

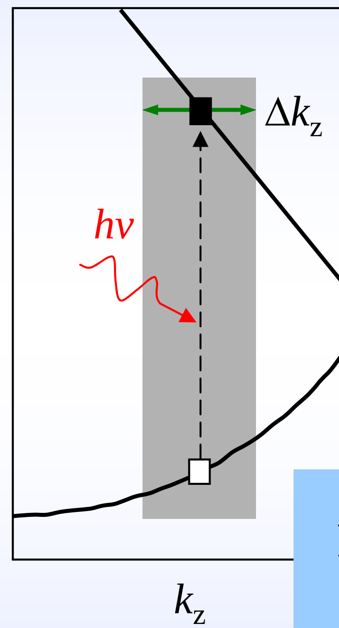
⇒ **soft-X-ray range** to increase bulk sensitivity

Virtue 2: Improvement of intrinsic k_z-resolution

- Concept of intrinsic k_z-resolution



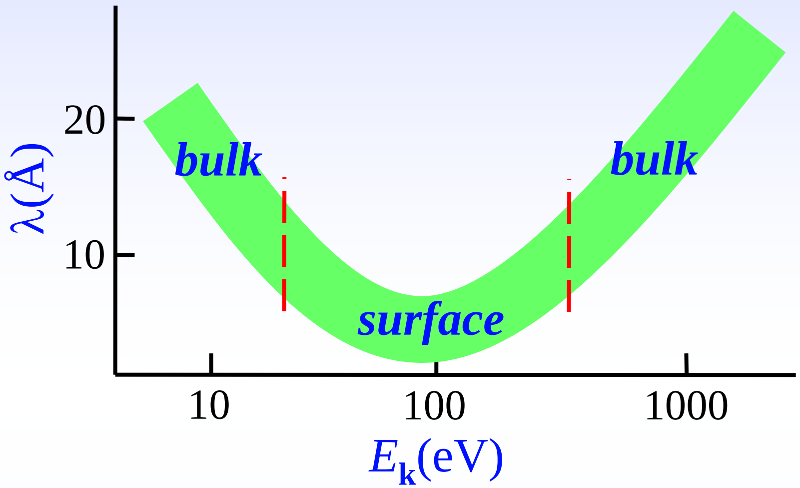
lifetime damping in surface-perpendicular z
$\Rightarrow$ broadening $\Delta k_z = \lambda^{-1} = 2\mathrm{Im}k_z$



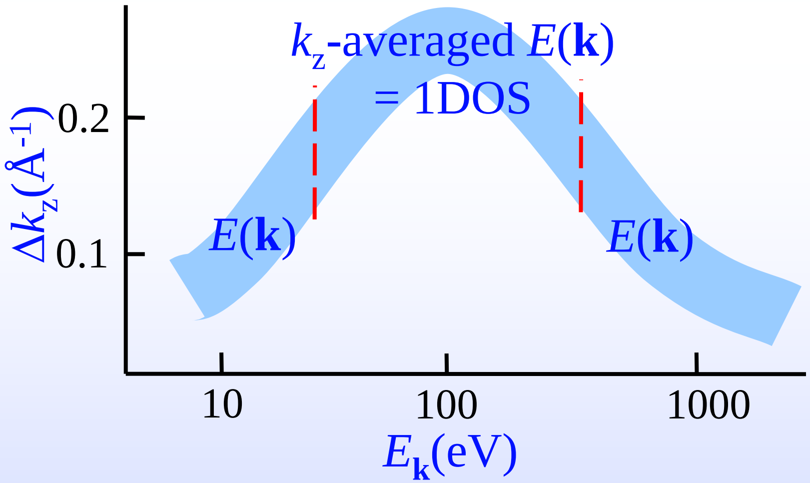
PE signal by *averaging* of $E(k_z)$ within Δk_z

- Δk_z broadening = *intrinsic k_z resolition*
- $\Delta k_z \ll k_z^{BZ}$ required to achieve accurate resolution in k_z

- Two-sides "universal" curve



$\Delta k_z = \lambda^{-1}$



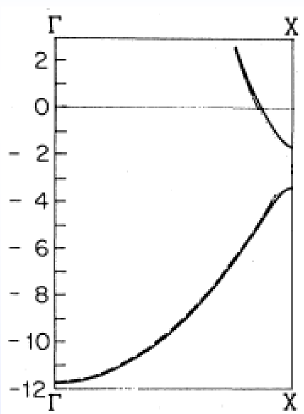
⇒ **soft-X-ray range** to increase the k_z-resolution and achieve $\Delta k_z << k_z^{BZ}$

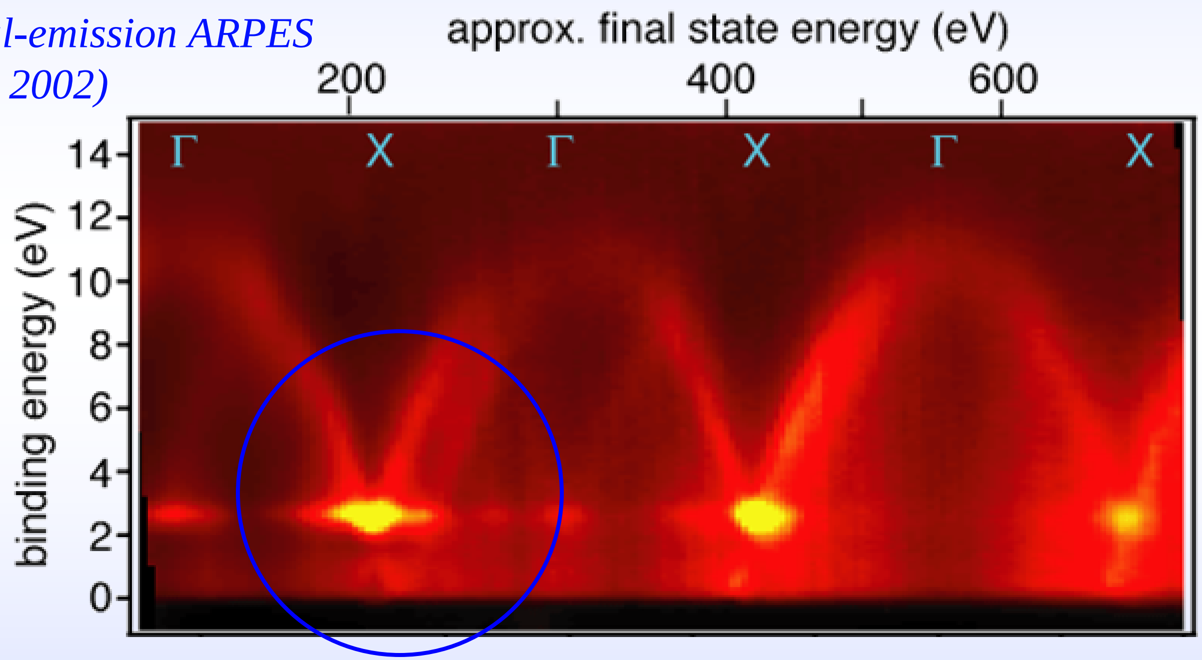
Virtue 3: Free-Electron Final States

- Final-state $E(k_z)$ is required to recover the valence band $E(k_z)$

- How far in energy do the non-free-electron effects carry on?

Al(100) normal-emission ARPES
(Hoffman et al 2002)



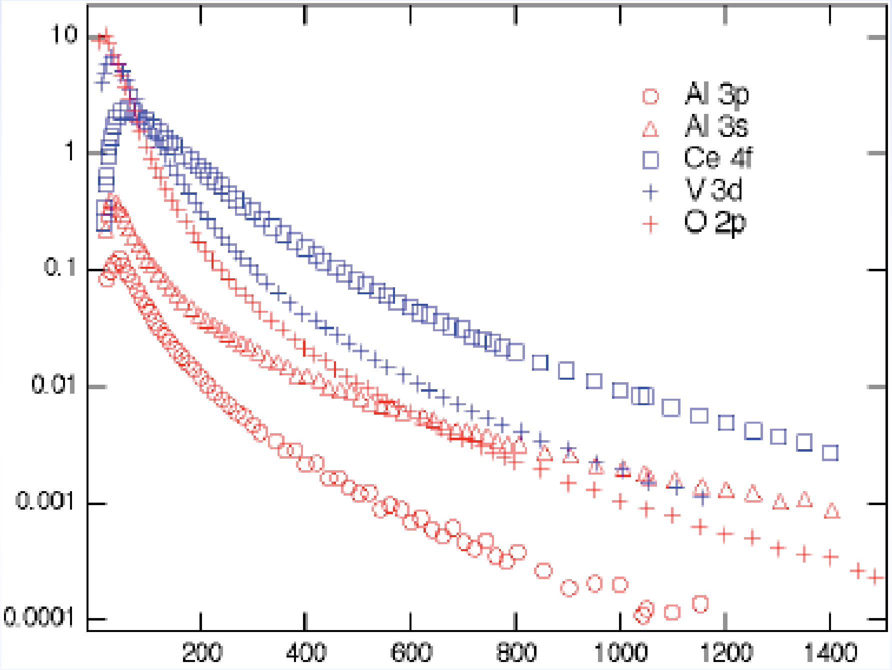


multi-k_z final states

• failure of FE-approximation below ~500 eV despite the free-electron nature of Al and rather high $h\nu \Rightarrow$ **SX-ARPES** to achieve FE final states

• Further virtues: Simplified matrix elements, …

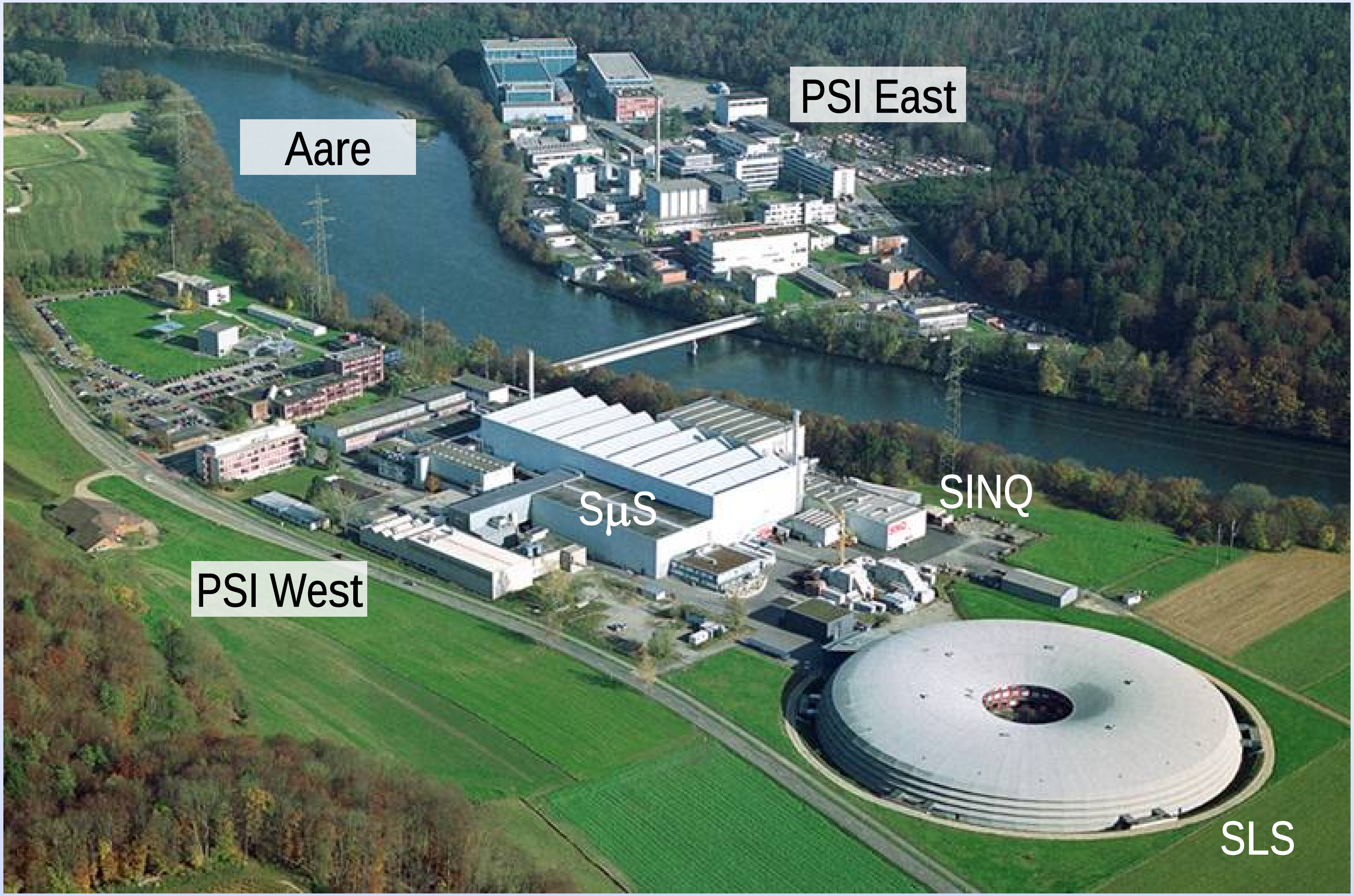
Problem: Small Crossection



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

SX-ARPES facility @ Swiss Light Source

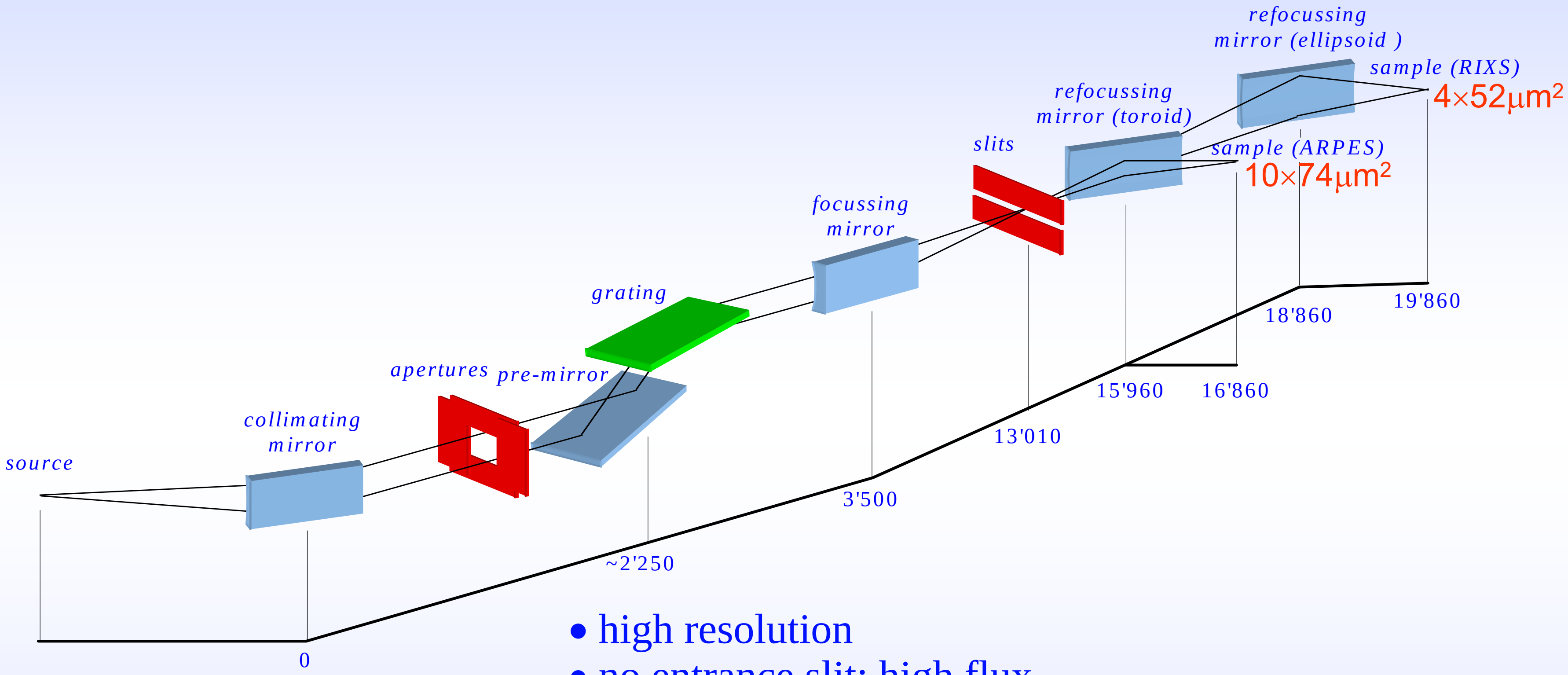
- Paul Scherrer Institute: Aerial view



ADRESS (*ADvanced RESonant Spectroscopies*) Beamline :

- soft-X-ray radiation with circular and 0-180^{o} variable linear polarizations
- energy range 300 – 1800 eV
- high resolution ΔE ~ 30 meV @ 1 keV
- collimated-light PGM optical scheme
- RIXS (~70 meV @ 1 keV) + ARPES (~30 meV @ 1 keV) endstations

Optical Scheme: Collimated-light PGM

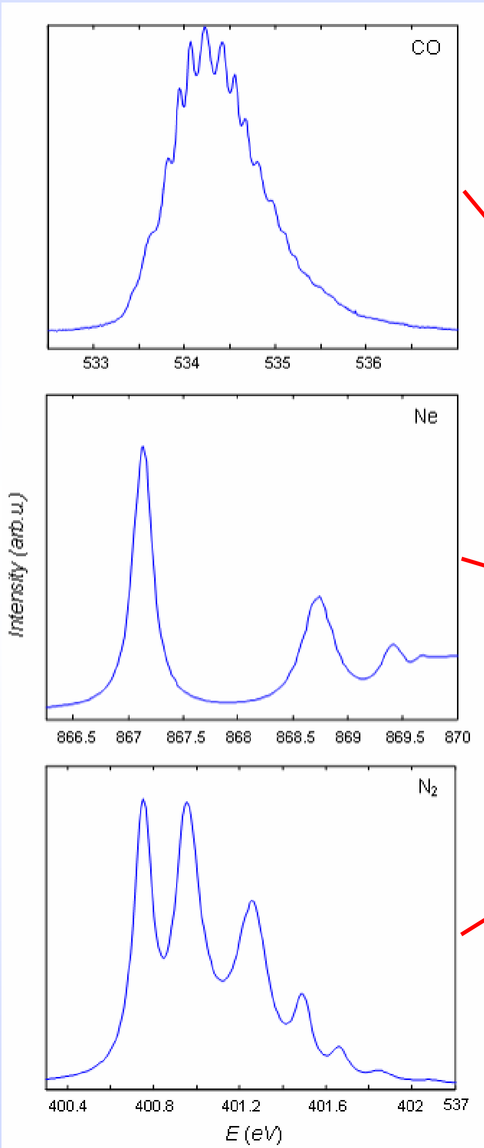


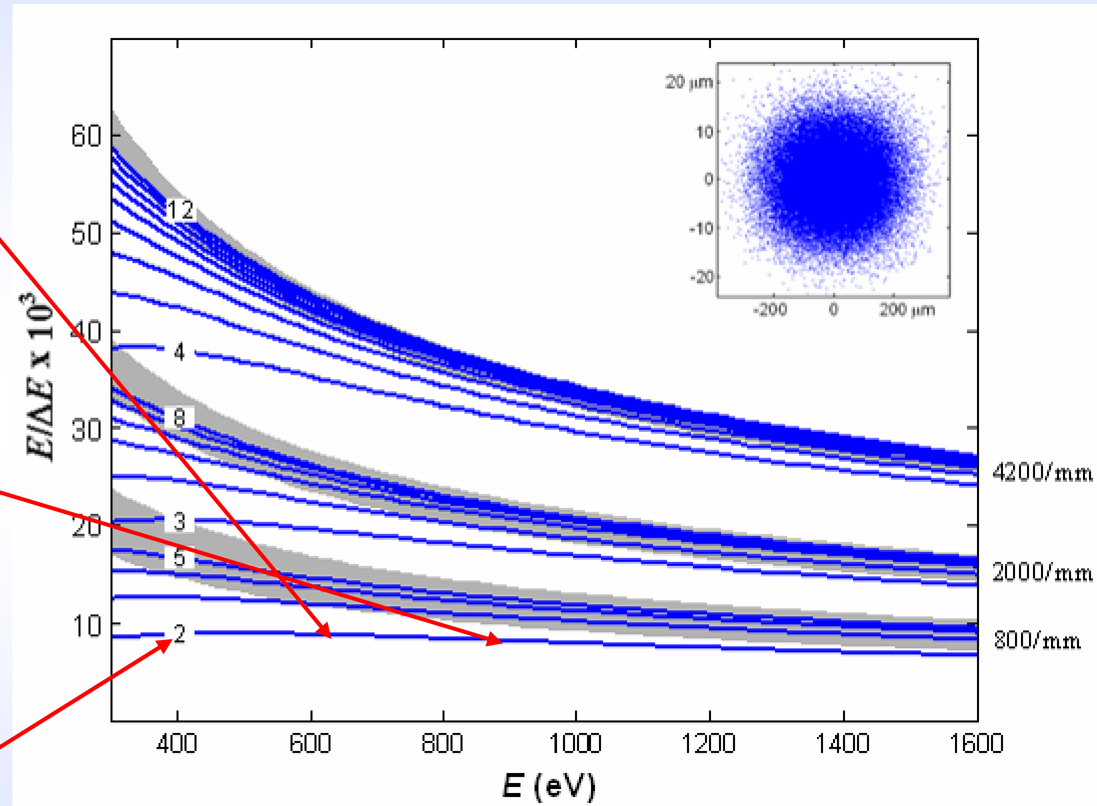
- high resolution
- no entrance slit: high flux
- wide energy range
- resolution, flux and HIOS optimization by C_{ff}
- proven design and flawless operation @ SLS

- features of ADRESS to increase flux:

- glancing incidence angles on all mirrors
- optimization of the gratings groove densities and profiles

Resolution parameters





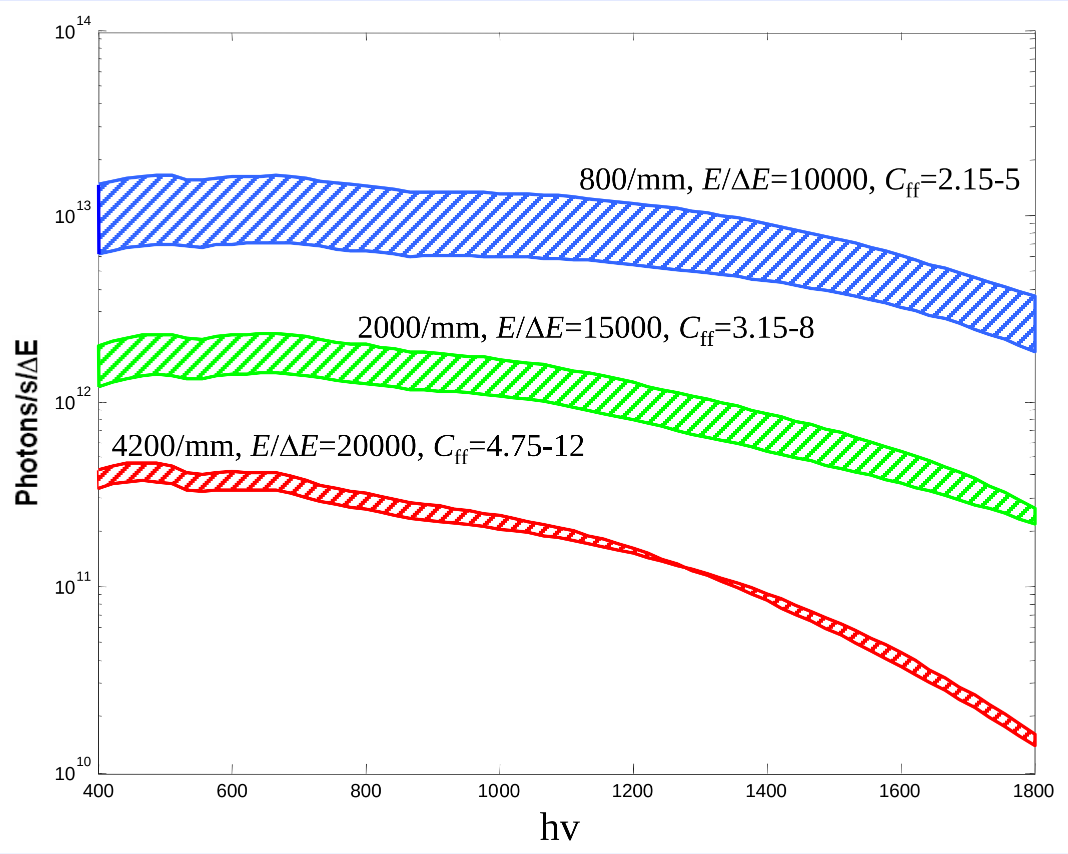
- FWHM spot size = 14.1×228 μm^2, almost no aberrations
- $E/\Delta E$ up to 33000 @ 4200/mm, C_{ff}=10
- optimal resolution coverage with 800, 2000 and 4200/mm

Flux parameters

• flat energy dependence with all gratings including 800/mm blazed

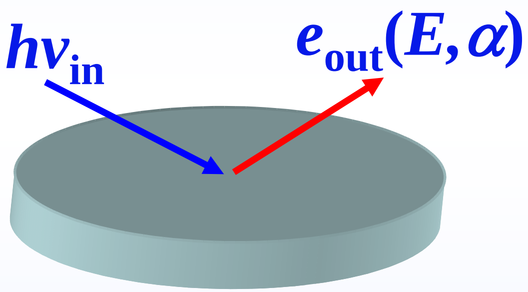
• flux-optimal C_{ff} increases with l/mm and energy

• 3×10^{11} to 1×10^{13} ph/s/0.01%BW @ 1 keV (experimentally confirmed): factor of 10 to 100 better compared to other soft-X-ray beamlines



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

ARPES Endstation @ ADRESS: Purpose



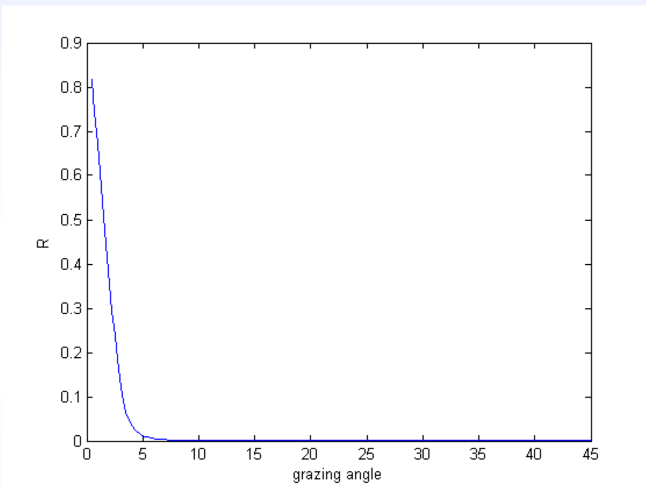
- high photoelectron energies (enhanced bulk sensitivity, free-electron final states …) with angular resolution $\Rightarrow$ soft-X-ray region

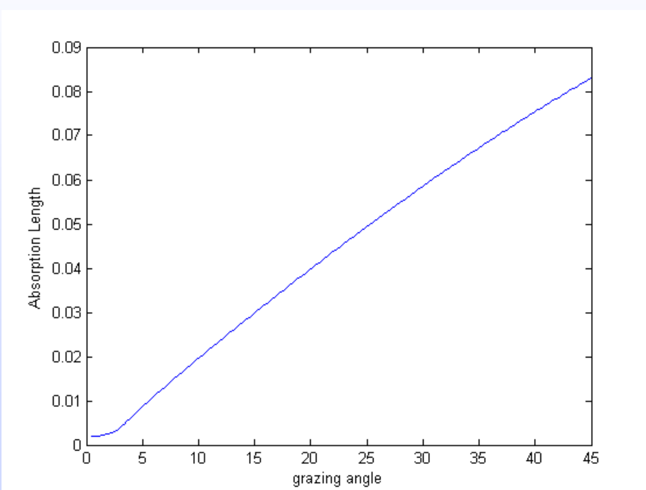
- electronic structure of complex materials (perovskites…) with resolution in 3-dim **k-**space

- compatibility with PLD thin film preparation

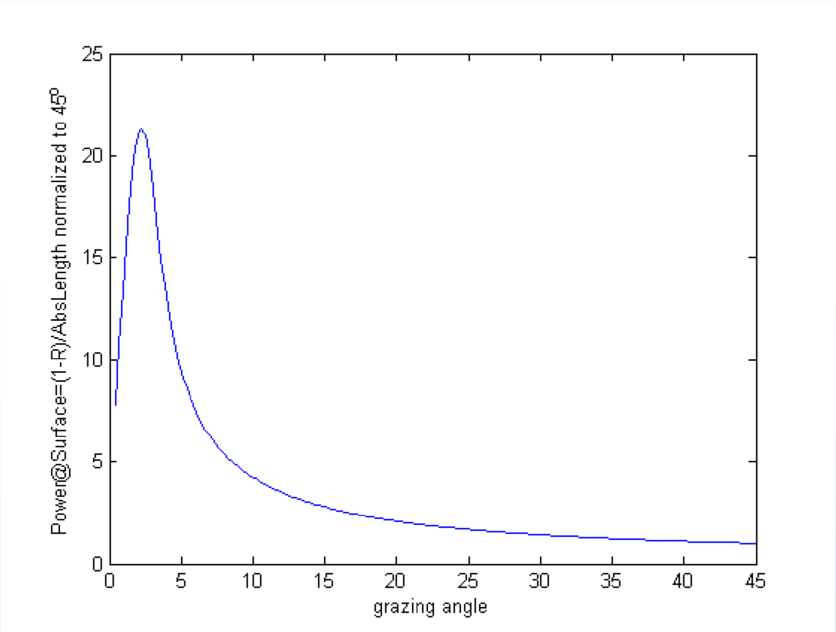
SX-ARPES Endstation @ ADRESS: Concepts

Experimental geometry concepts: Optimal light incidence angle





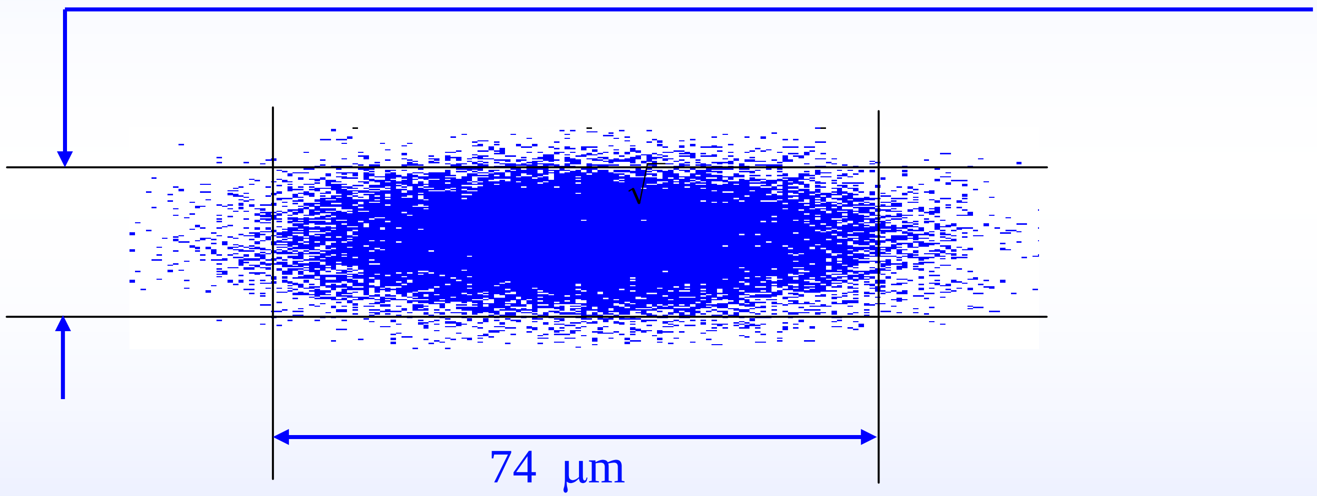
$\lambda_e << \lambda_{hv}$ => Photoelectron Yield $\propto (1-R)/\ \lambda_{hv}$



- photoelectron yield peak at glancing angles ~2.5º
- factor of 1.4 improvement @ 30º and 2.1 @ 20º

Experimental geometry concepts: Alignment of the light footprint

$[(\text{demagnified slit})^2 + (\text{refocusing aberration})^2]^{1/2} \sim 10\ \mu m$ @ $10\ \mu m$ slit

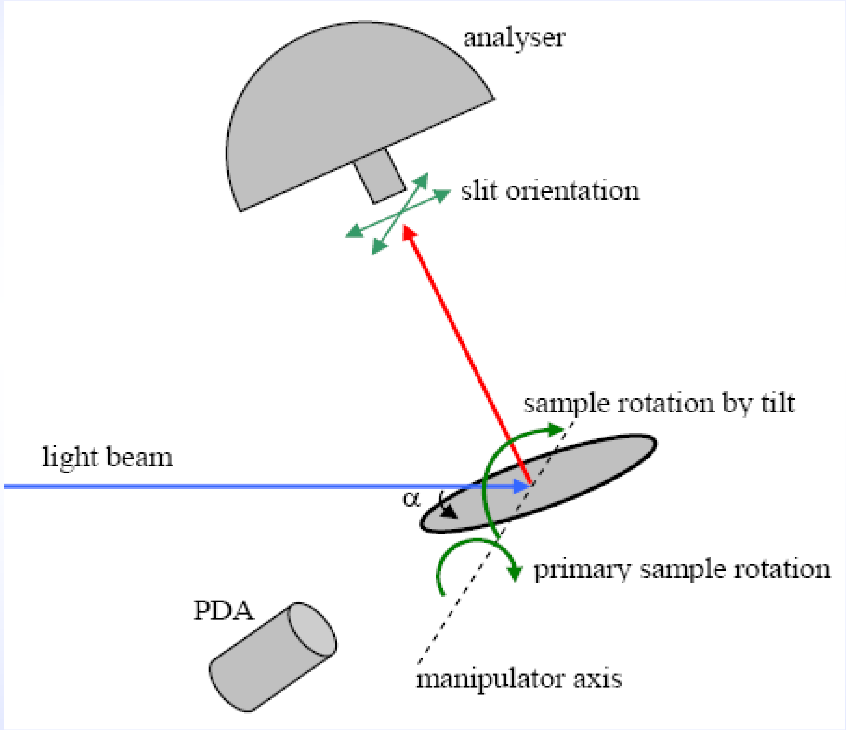


- *rotation around the horizontal axis* to align the horizontal and vertical spot size
- 100 μm slit => grazing incidence angle ~ 13.5°

Experimental geometry

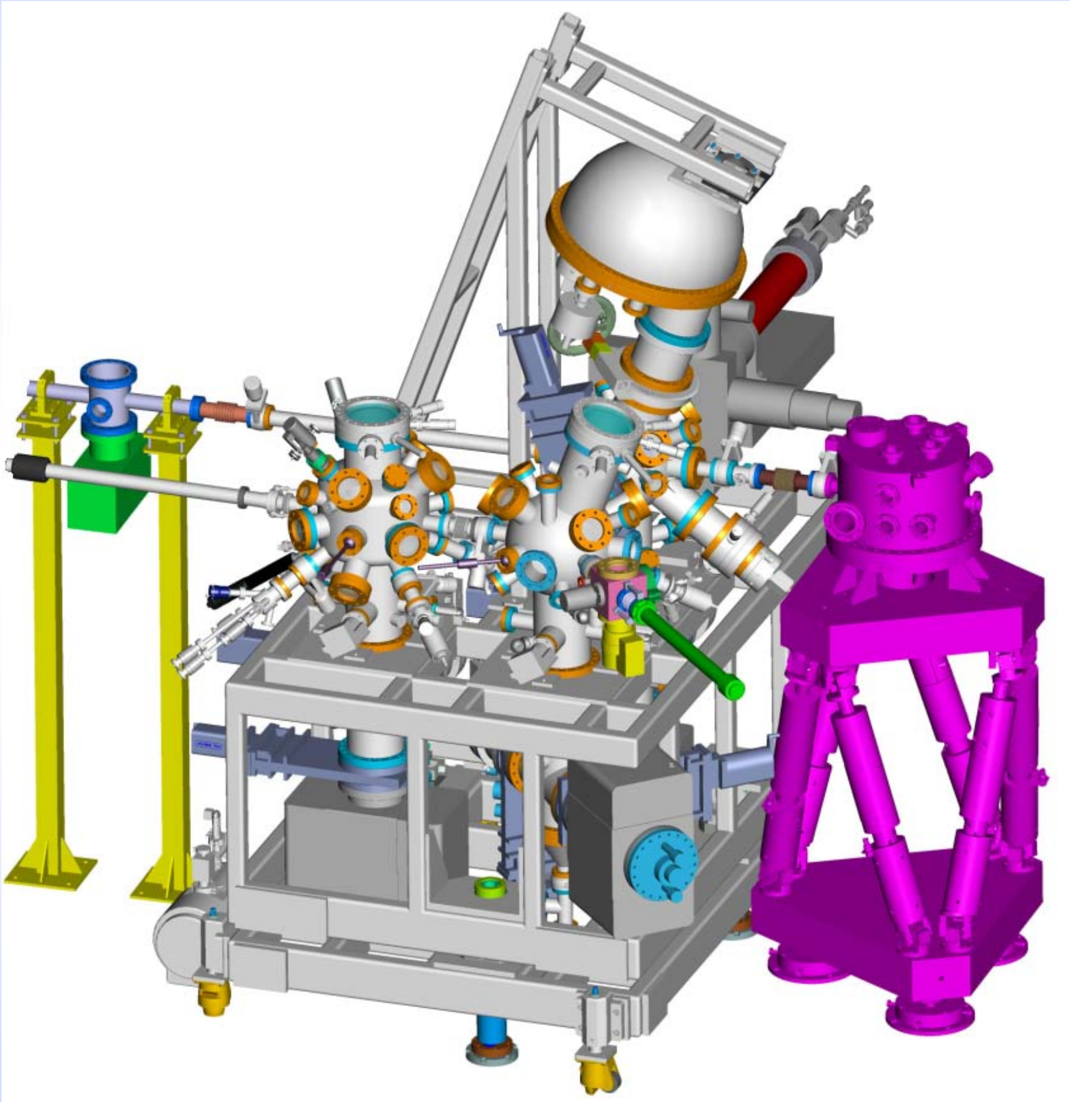
- Grazing incidence at 20^o // smaller vertical footprint with *horizontal* manipulator axis
- 2 operation modes:

- analyser slit // beam + tilt (selection rules)
- analyser slit ⊥ beam + primary rotation



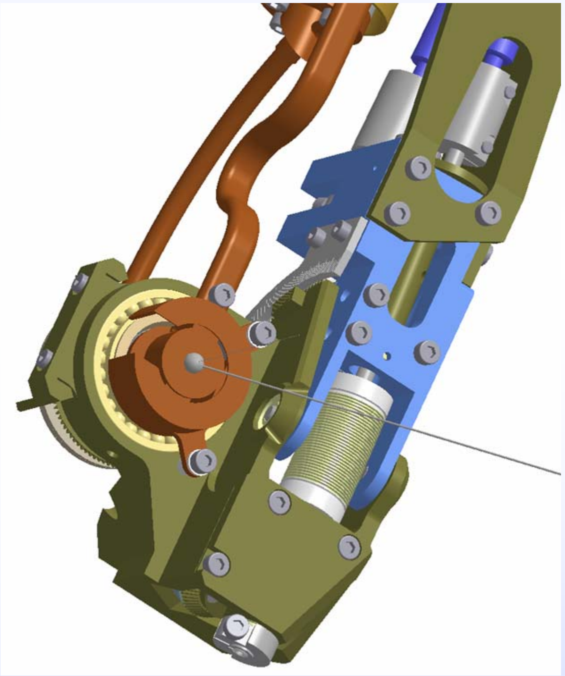
SX-ARPES Endstation @ ADRESS: Realization

- AC (analysis)
  TC (transfer & cleavage)
  PC (ion etching, thin films, chemistry)
- *one* sample transfer of cleaved samples to AC
- PLD port
- Easy switching between the RIXS and ARPES endstations by insertion of the RM



Manipulator

- $\Delta k_{//}=0.512\sqrt{E}\sin\alpha \Rightarrow$ high angular accuracy required
- Proprietary CARVING™ design by PSI + Uni Amsterdam
- 3 translation (resolution 5μm) + 3 angular (resolution 0.1°) DOFs
- L-He_2 cooling to 10.5K

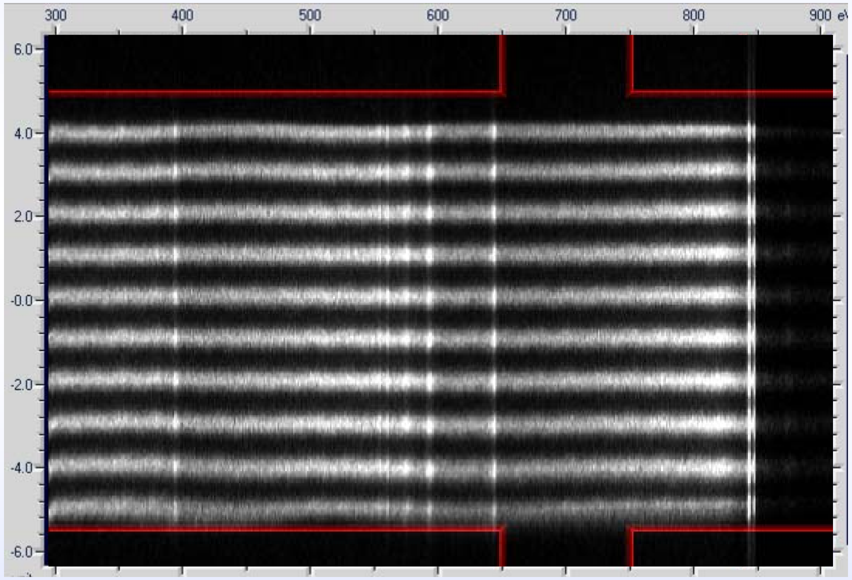


Analyser

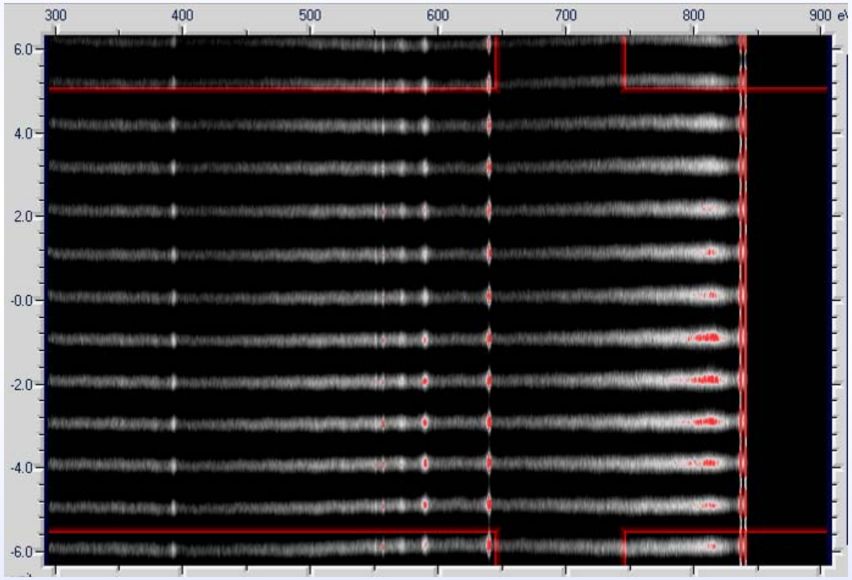
• PHOIBIOS-150 from SPECS: ΔE up to 3meV (beamline limited combined ΔE) and $\Delta\theta \sim 0.07^{o}$ @ 1 keV)

- Energy dependence of the angular dispersion measured with slit array (E_{pass} = 80 eV)

MAD mode (±4°)

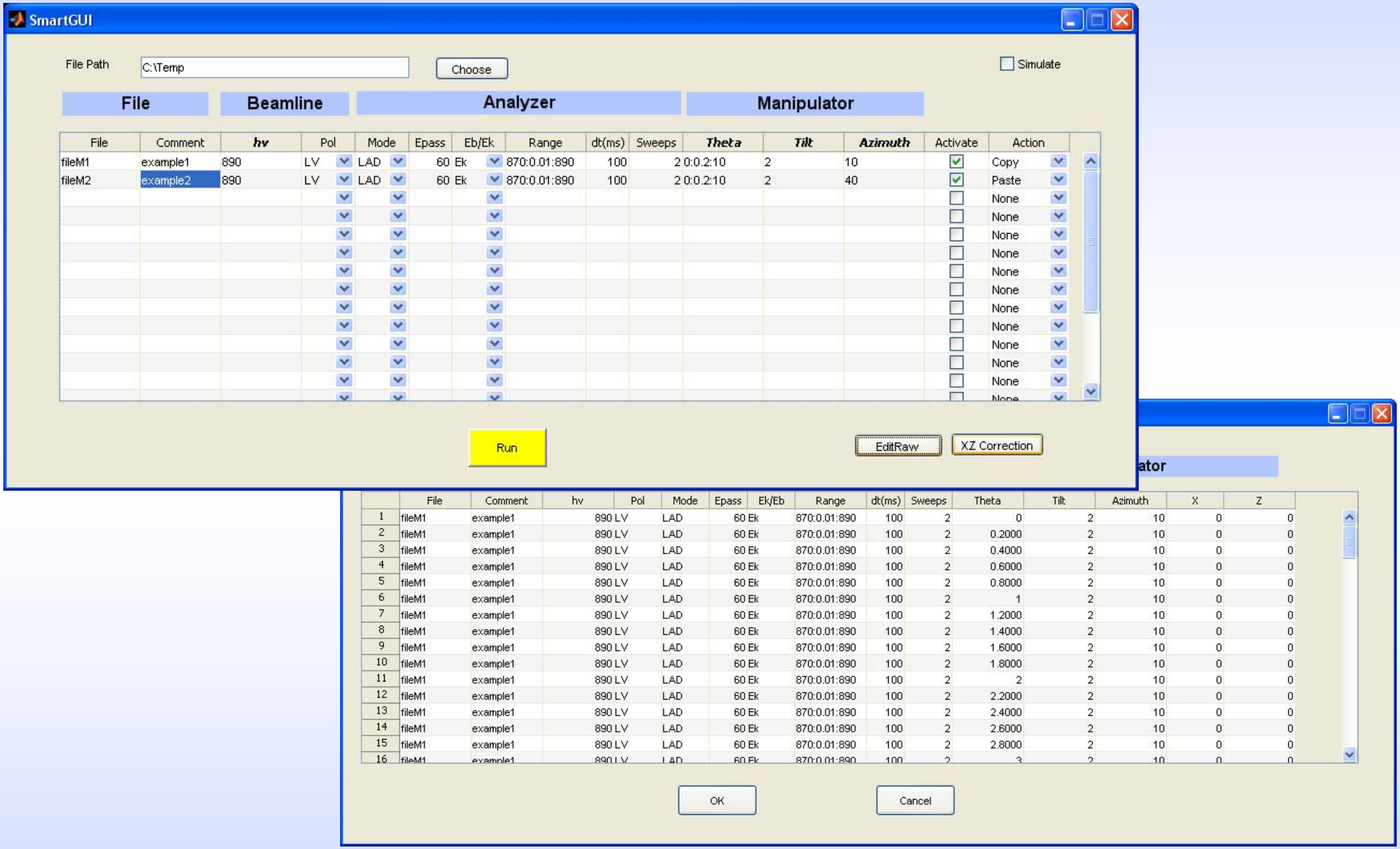


LAD mode (±6°)



• Linear and almost *E*-independent angular dispersion in the 300 - 900 eV range
• Minor problems (focal axis displaces with the operational mode up to 400 μm; illumination homogeneity). Is Scienta better?
• **Good overall performance allows reliable data acquisition**

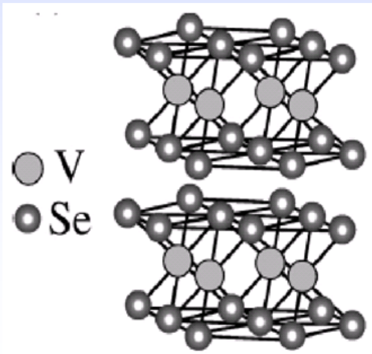
Software (X. Wang)

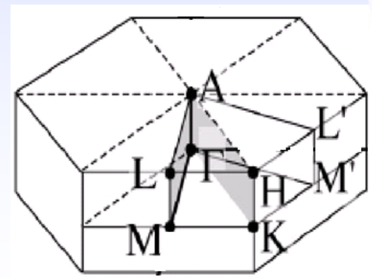


- "Smart Table" concept: vectorized *hv*, θ, tilt, azimuth
- Output data array in multidimensional hdf5 format

3D bandstucture and Fermi surface of VSe_2

- Structure and electronic structure

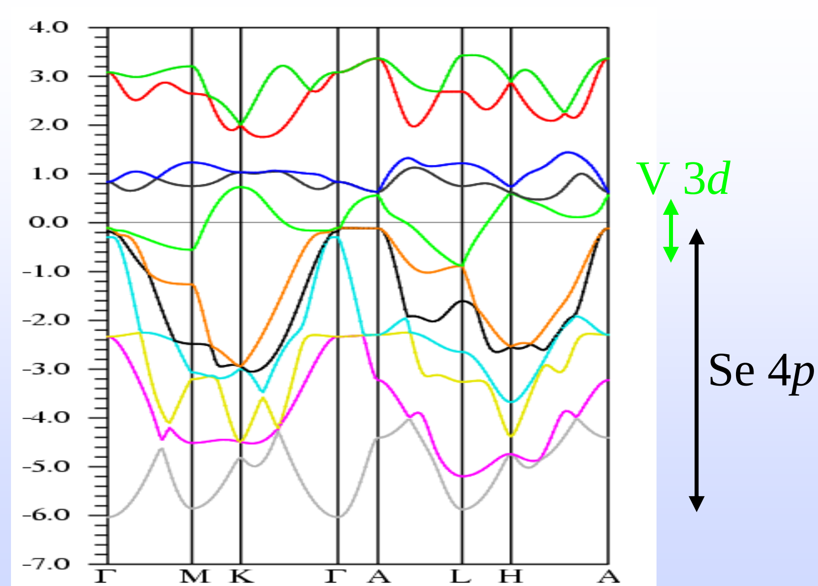




• 3-fold symmetry

• quasi-2D structure with weaker interlayer interaction

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



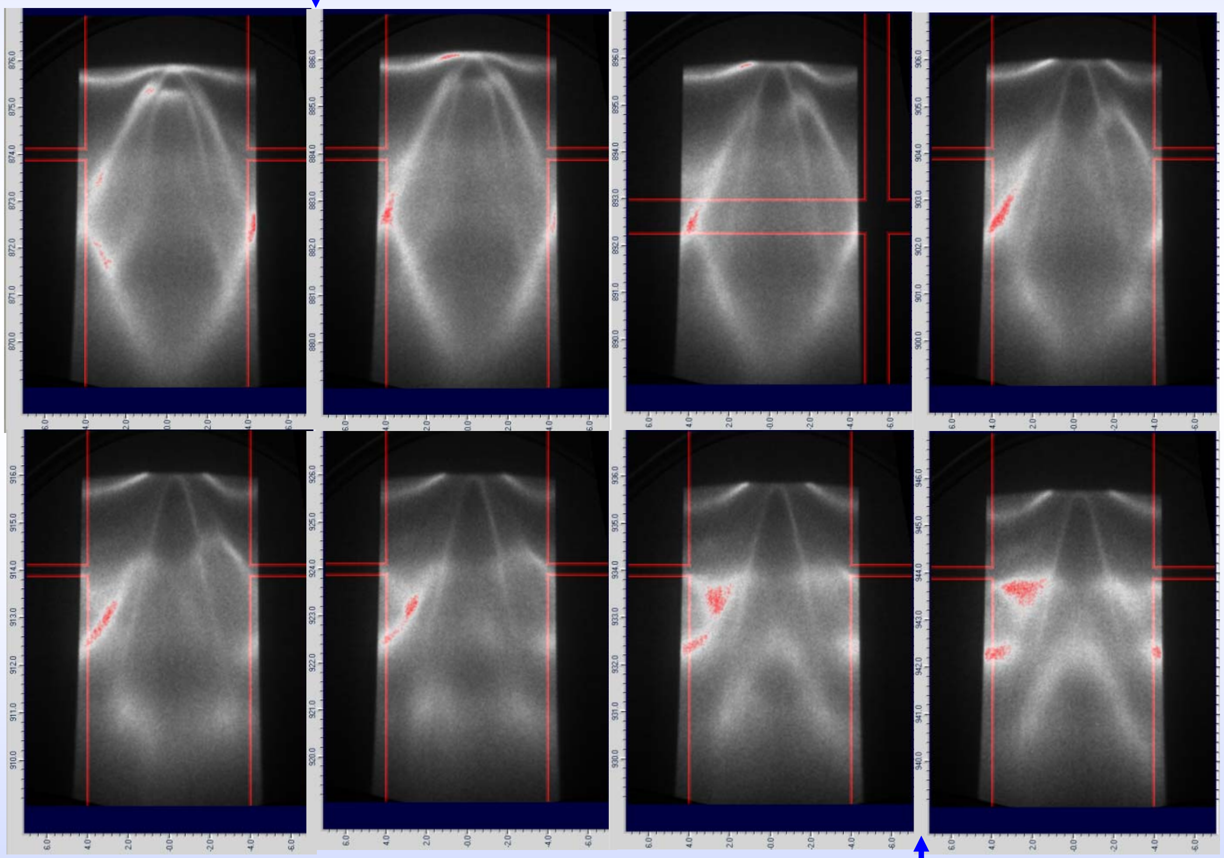
- Experimental results: k_z dispersion (MΓM azimuth)

- T=10.7K
- 800/mm, $slit_{BL}$=10μm, $slit_{AN}$=0.2mm, E_p=60V => combined **Δ*E*~120meV**
- **each image in 300 s**

⇓

- evolution of images => Δk_z~0.05 $Å^{-1}$ (or λ~20Å) - factor of 3-4 improvement compared to VUV-ARPES
- excellent intensity not only for *d*-, but also *p*-states despite dramatic loss of crossection
- intense and sharp in $\mathbf{k}_{//}$ structures => Debye-Waller and phonon broadening are no prohibitive

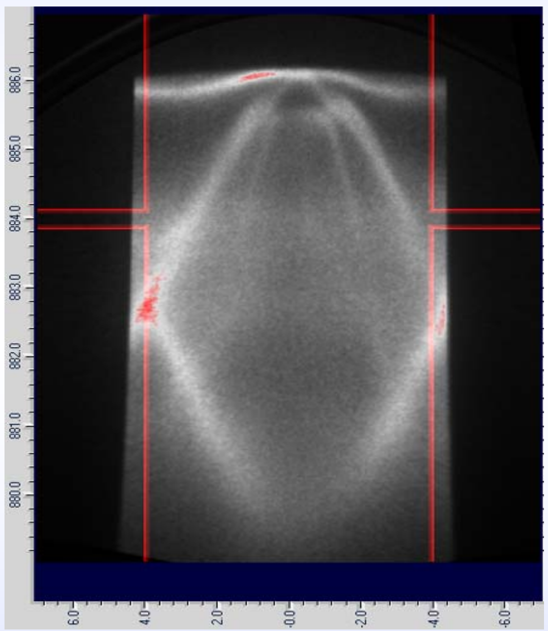
Γ~885 eV



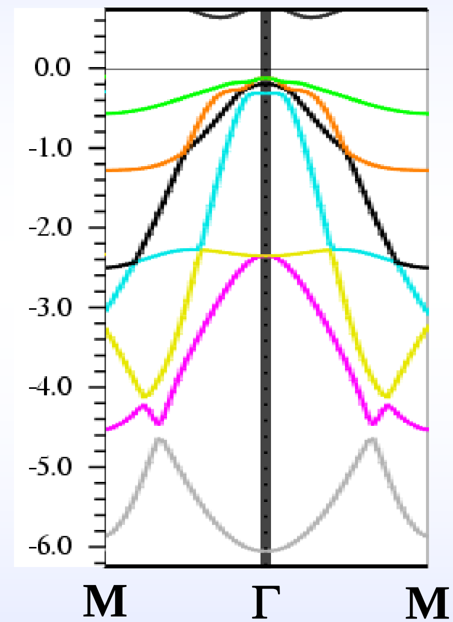
A~945 eV

- Comparison with DFT calculations

hν=885 eV (k_z=Γ)

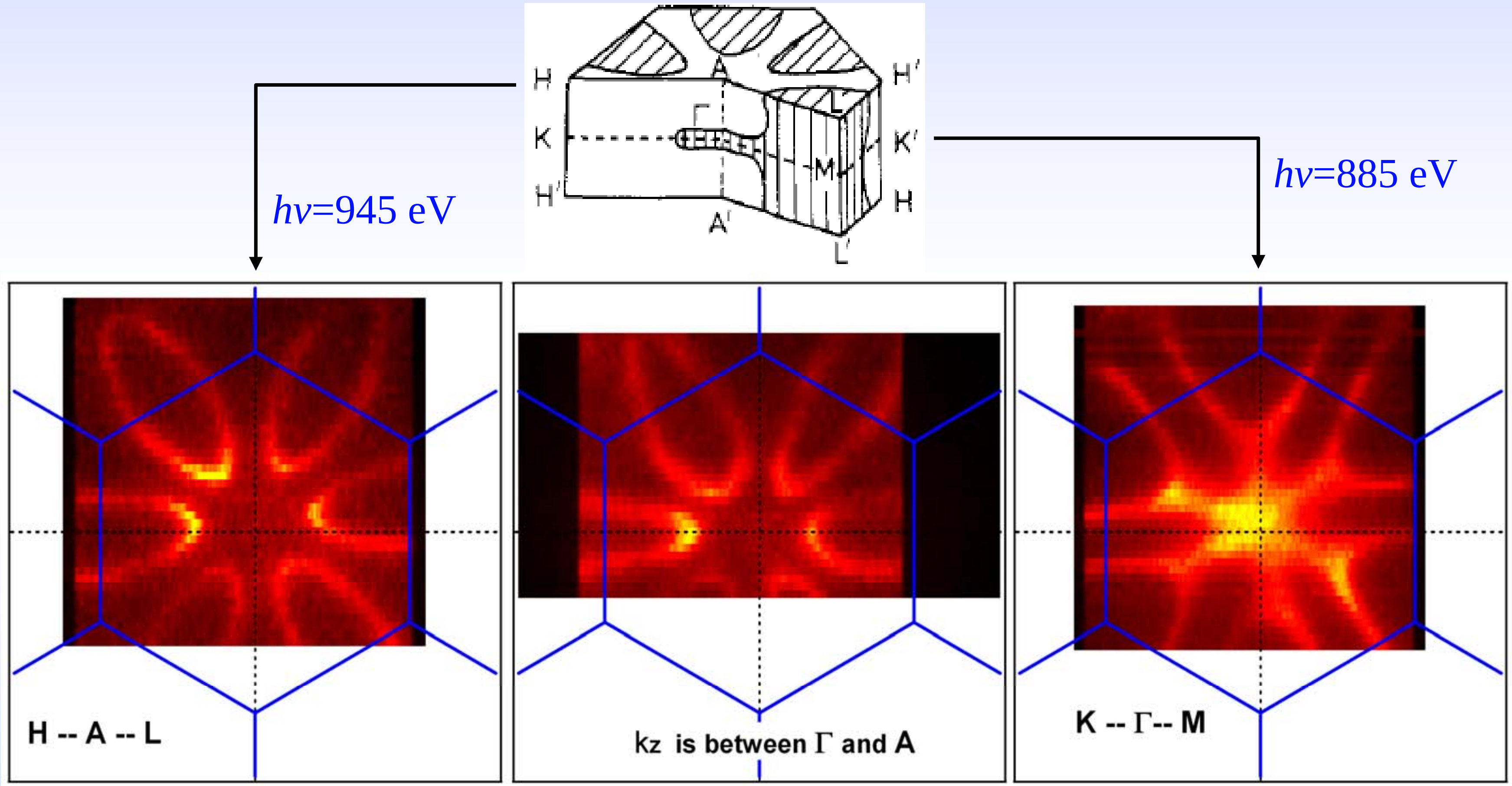


GGA-DFT (P. Blaha)



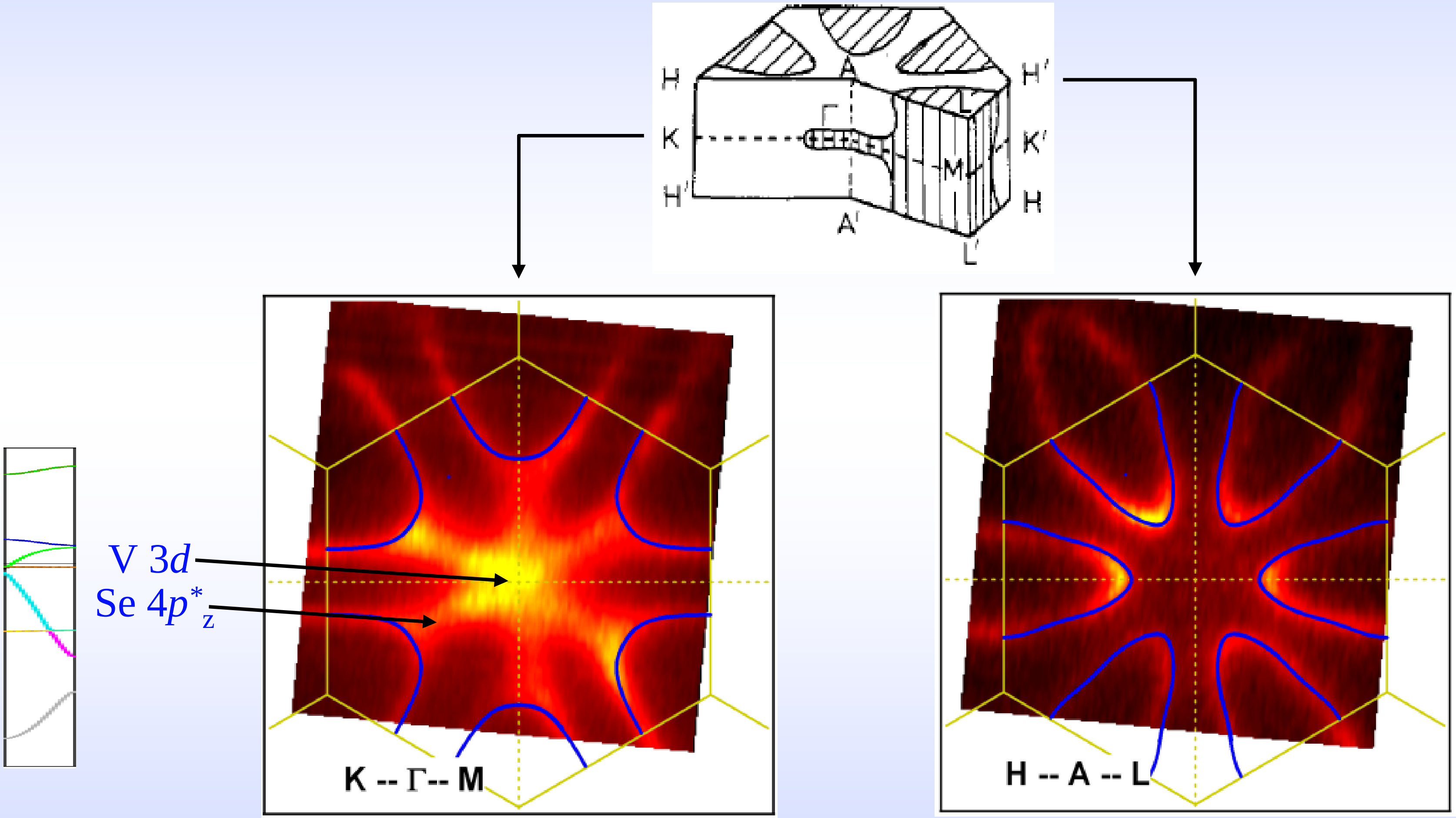
- excellent agreement

- Experimental results: Fermi surface



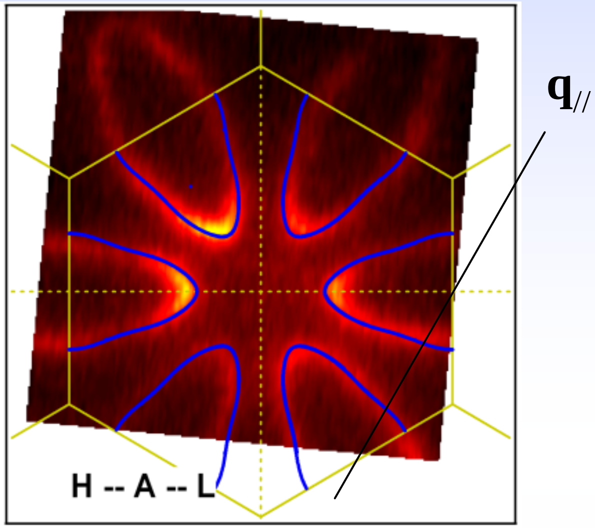
- $slit_{BL}$=10μm, $slit_{AN}$=0.2mm, E_p=60V => combined ΔE~120meV
- each image in 400 s; each map of ~40 images in less than 5 hrs
- extraordinary clarity of the experimental data without any image enhancement

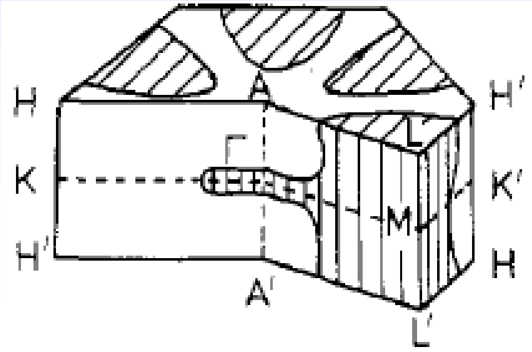
- Comparison with GGA-DFT calculations



- fantastic agreement, even the tiny warping in HAL
- V 3*d* seen in KΓM (Se 4p^*_z due to energy resolution)

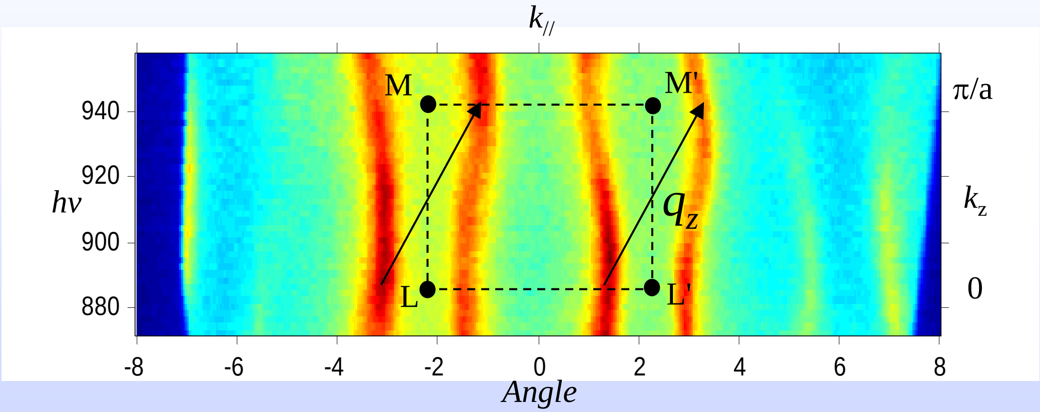
- Origin of CDWs





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

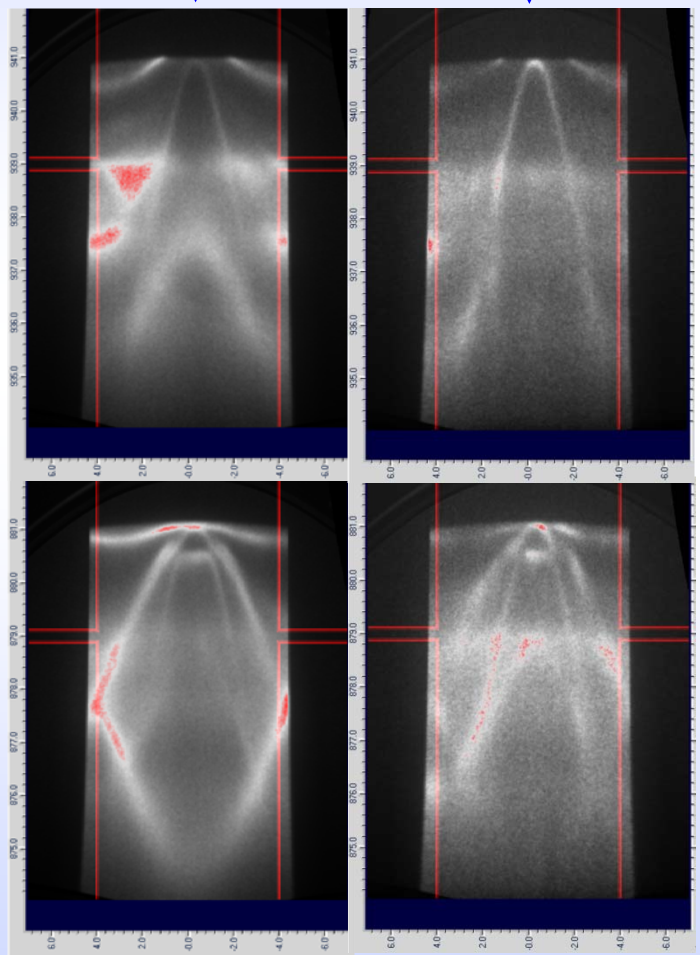
• Perpendicular cut in MLL'M' plane: 3D warping to support nesting with the experimental q_z



Polarization dependence

- each image in 300 s
- selective excitation from different V 3*d* and Se 4*p* bands

LV (*s*-pol) LH (*p*-pol)



A (945 eV)

Γ (885 eV)

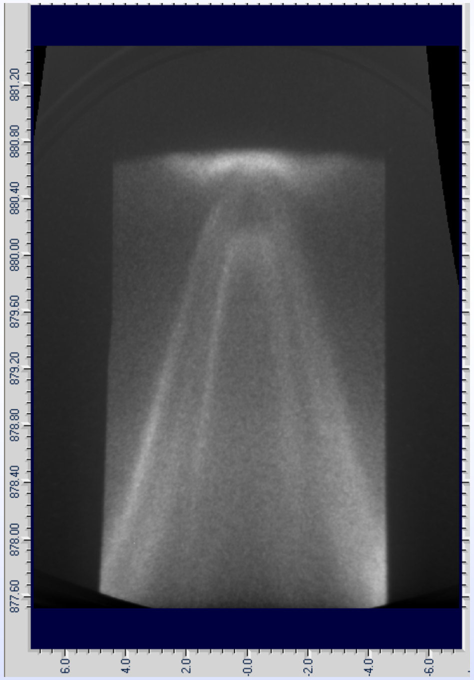
Towards better resolution

- 800/mm in 2nd order, slit$_{BL}$=10µm, slit$_{AN}$=0.2mm, E_p=30V => combined **Δ*E*~60meV**

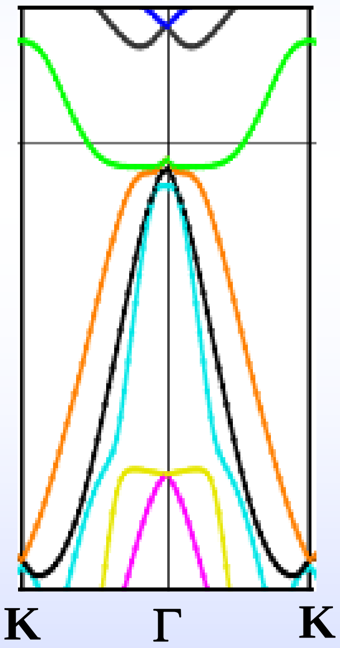
- **image in 2000 s**

- well resolved bands

LV-pol, KΓK azimuth,
k_z=Γ (885 eV)



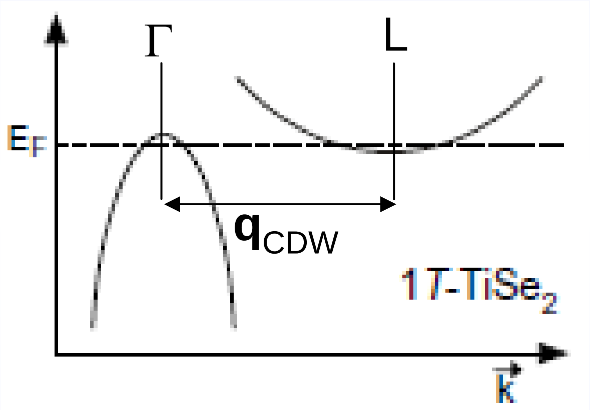
GGA-DFT



CDW band gap in excitonic insulator $TiSe_2$

Uni Fribourg: group of P. Aebi

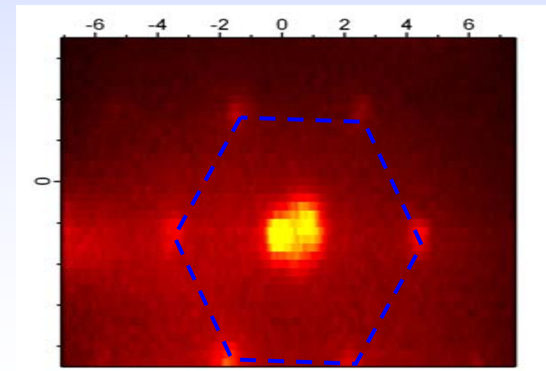
- CDWs due to excitonic coupling between the Γ and L points



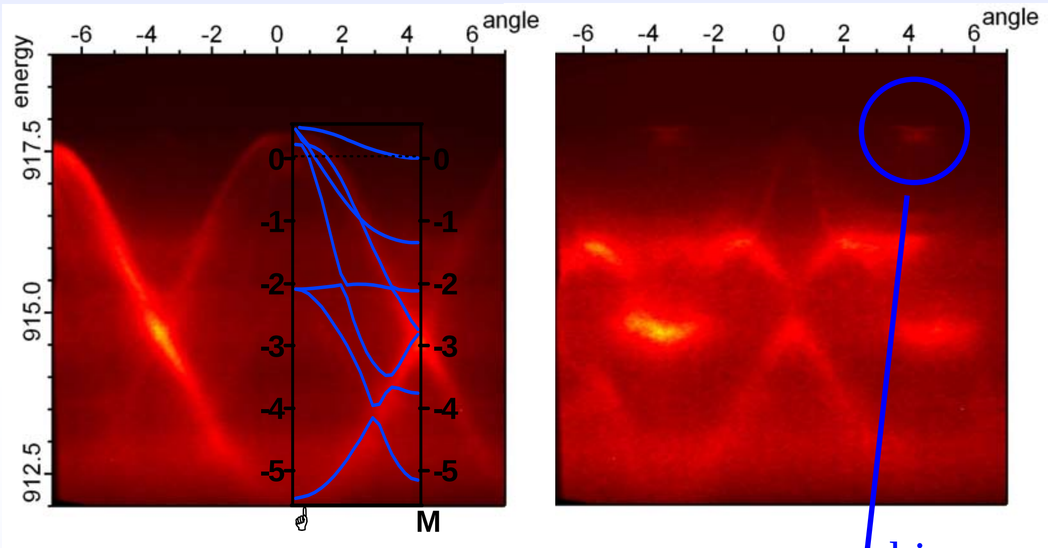
C. Monney *et al* 2010

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

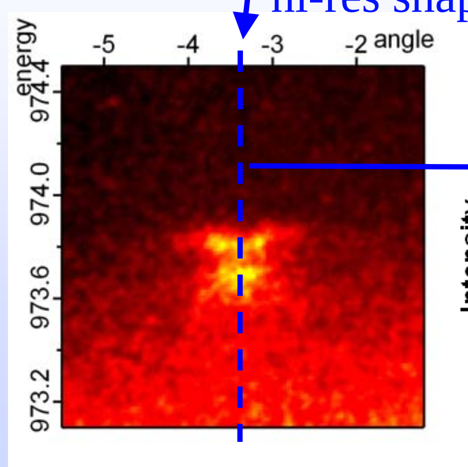
FS map @ k_z=A-point

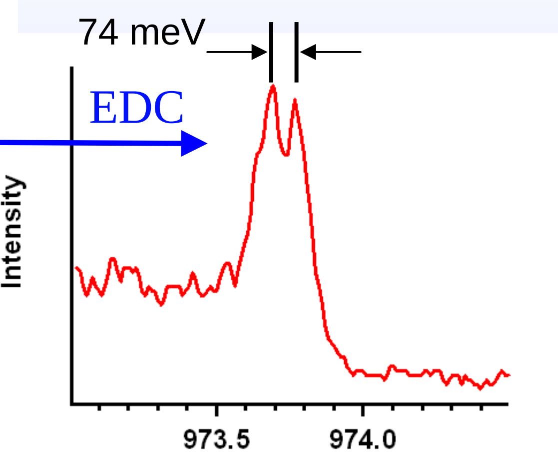


k_z=Γ-point ($h\nu$=922eV) A-point ($h\nu$=978eV)



hi-res snapshot ΔE~70meV



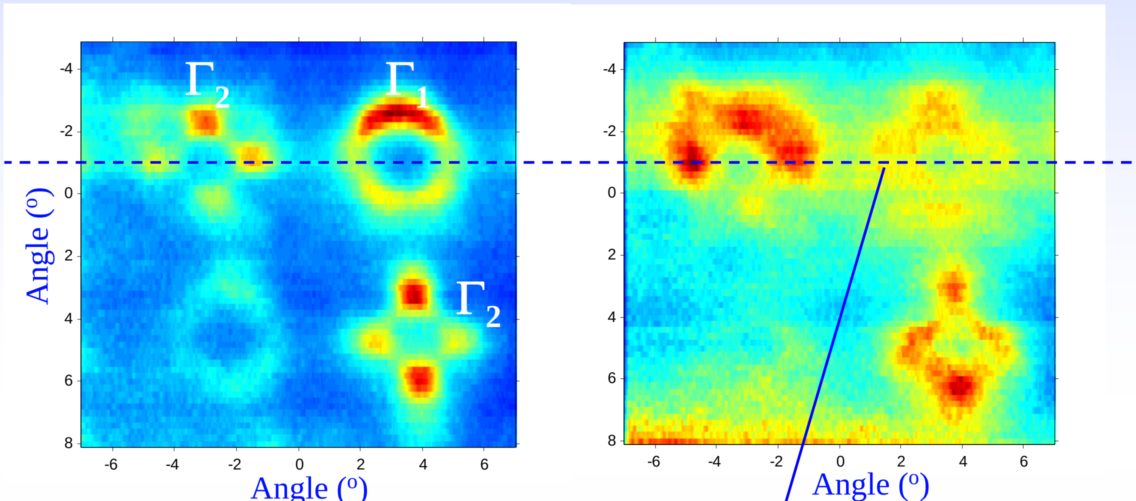


- CDW band gap between CBM in L and VBM replica from Γ

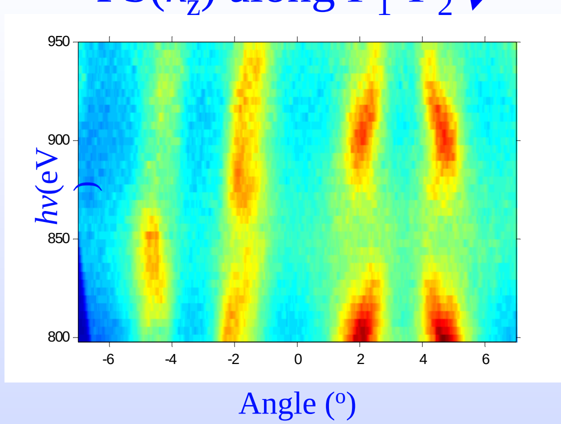
3D Fermi Surface of HTSC pnictide $Ba_{0.6}K_{0.4}Fe_2As_2$

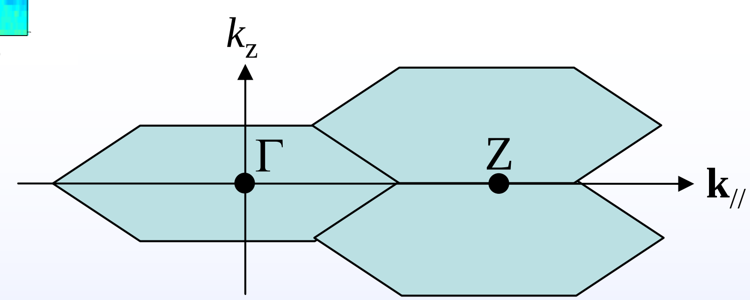
- measurements at *p*-pol, T=10.7K, ΔE~110meV

FS($\mathbf{k}_{//}$) @ $h\nu$ = 900 eV $h\nu$ = 850 eV



FS(k_z) along Γ_1-Γ_2





Beyond the 3-dimensionality:

- k_z dispersion along Γ_2 reduced to Γ_1
- the lower Γ_2 stays wheel-like

• matrix elements? effective halving the unit cell?

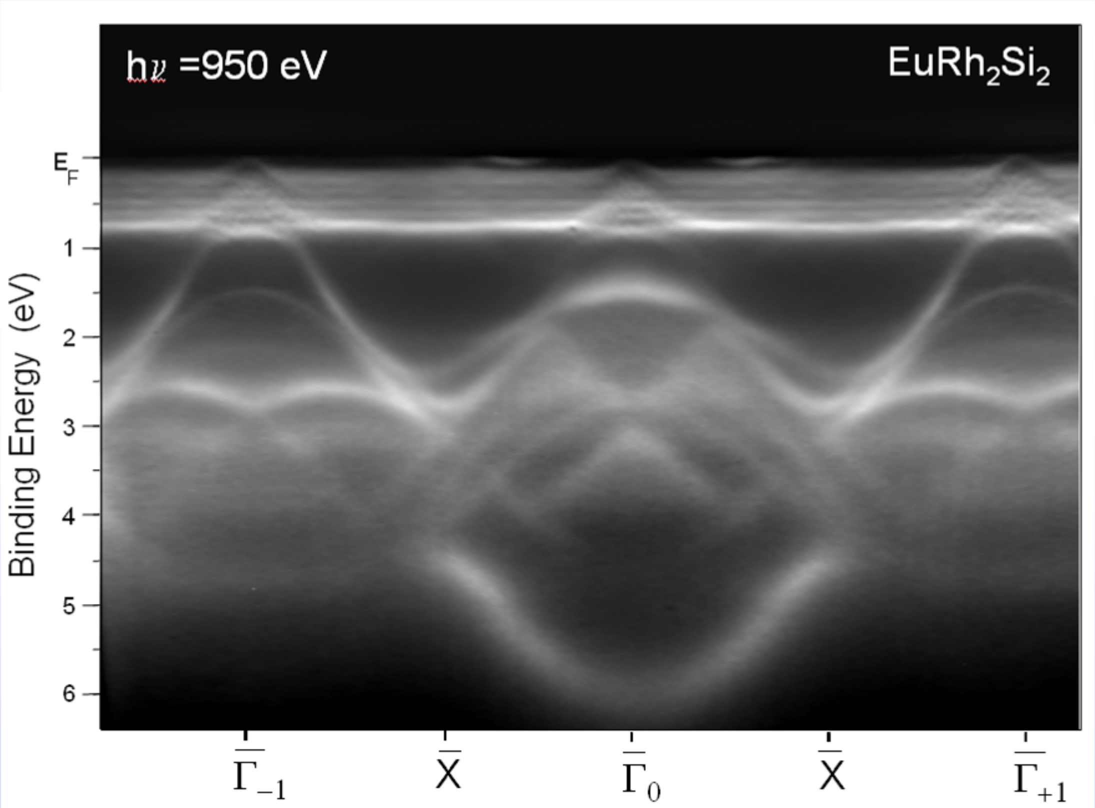
3D bulk electronic structure of heavy-fermion $EuRh_2Si_2$

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

- problems:

- electronic structure modification in subsurface region
- 3D effects

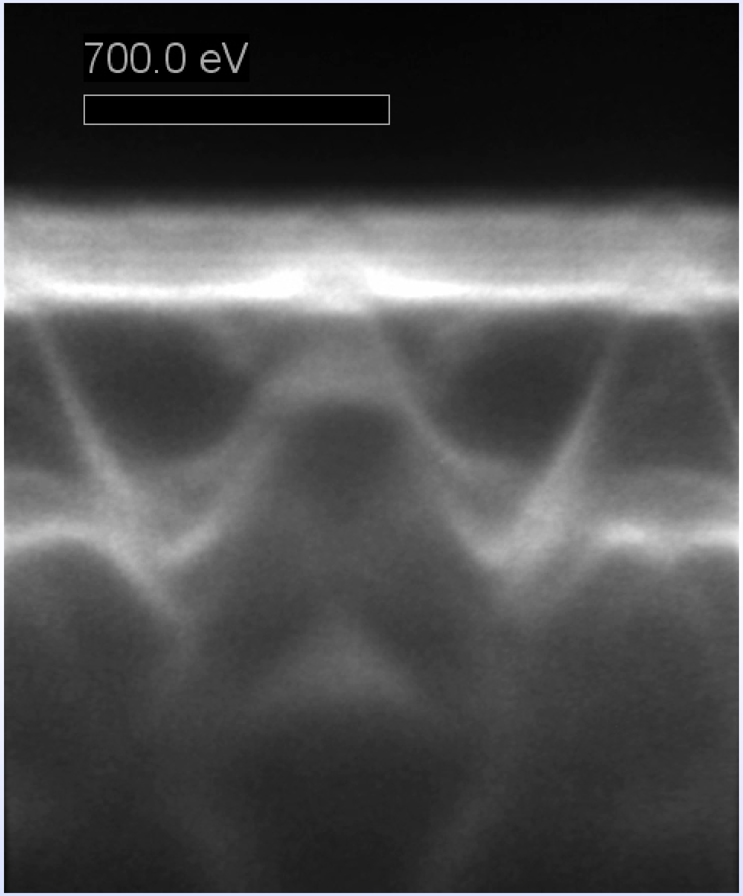
p-pol, T=10.7K, Δ*E*~110meV



- large photoexcitation crossection of the *f*-states
- excellent spectral contrast
- clearly resolved ^7F_J multiplet of the *f*-states and hybridization with extended states

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

- 3D dispersions by variation of *hν*

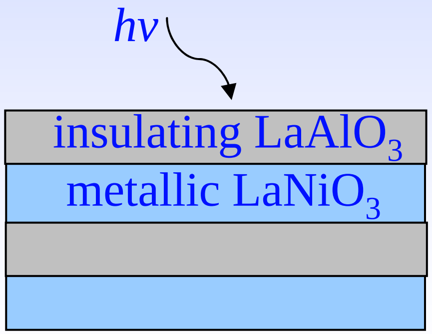


• prominent 3D character of the extended states

• k_z-dependent hybridization between the extended and *f*-states

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

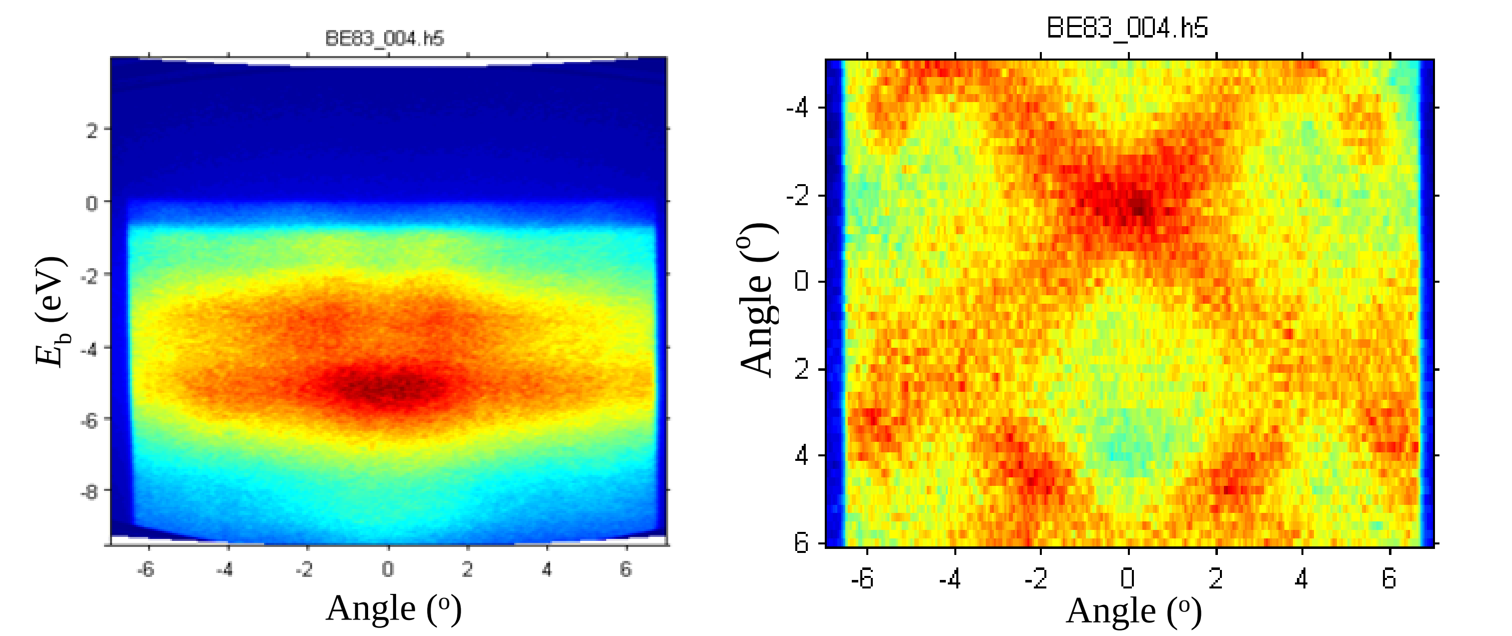
Uni Wuerzburg: M. Sing, G. Berner and R. Claessen



2 uc ~ 16Å

• large λ required

$h\nu$ = 1090 eV, ΔE~120meV, *p*-pol



• well-resolved Fermi surface

Conclusions

• Advantages of soft-X-ray ARPES:
- enhanced probing depth, improved resolution in k_z, free-electron final states, simplified matrix elements

• ADRESS beamline:
- energy range 300 - 1800 eV
- high res (ΔE~30meV@1 keV) and flux (up to 10^{13} ph/s/0.01%BW @ 1 keV)

• SX-ARPES endstation:
- He_2-cooled manipulator with 3 angular DOFs, rotatable analyser, user friendly
- image acquisition within ~5 min @ ΔE~110 meV and ~30 min @ ΔE~60meV; whole FS within a few hrs
- flux performance of ADRESS breaks through the valence band crossection problem

• Test case of VSe_2: Textbook clarity of k_z -resolved spectra and FS maps by virtue of free-electron final states and small Δk_z in the soft-X-ray region

• Further examples: excitonic insulator $TiSe_2$; HTSC pnictide $Ba_{0.6}K_{0.4}Fe_2As_2$; heavy-fermion intermetallic EuRh2Si2; buried layers in LNO/STO heterostructures …

$\Rightarrow$ **potential of SX-ARPES, in particular for 3D systems**

People

ADRESS Beamline

V.N. Strocov, M. Kobayashi, M. Shi, C. Hess, T. Schmitt, L. Patthey

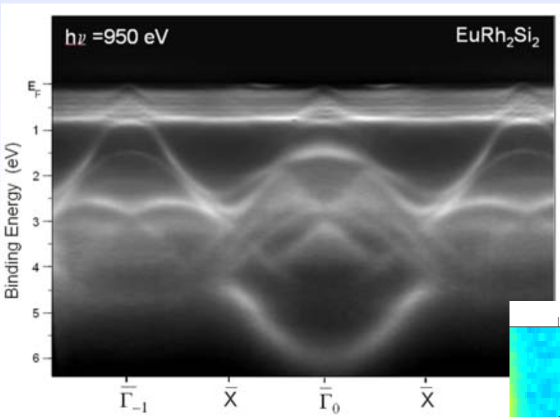
SLS stuff

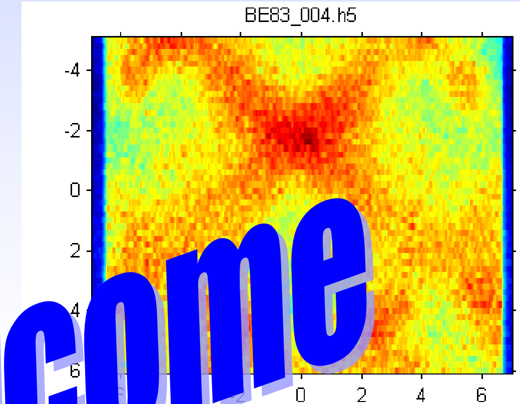
Optics group (U. Flechsig), ID group (T. Schmidt),
controls support (J. Krempasky, X. Wang) *et al*

ADRESS

PAUL SCHERRER INSTITUT PSI

SWISS LIGHT SOURCE SLS





Proposals Welcome