k-resolved electronic structure by soft-X-ray ARPES: From 3D systems to buried heterostructures

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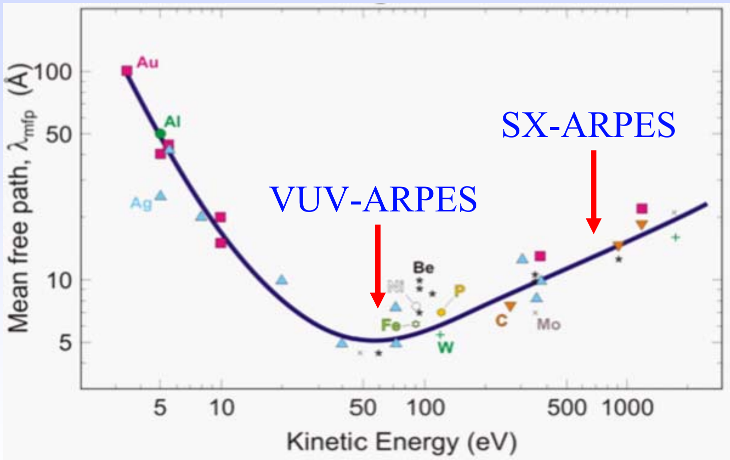
Outline

1.Why (and why not) ARPES in the soft-X-ray range?
- probing depth, 3D momentum resolution and resonant photoemission

2. Instrumentation

3. Spectroscopic abilities of SX-ARPES and results
- from 3D electronic structure to buried heterostructures and impurities

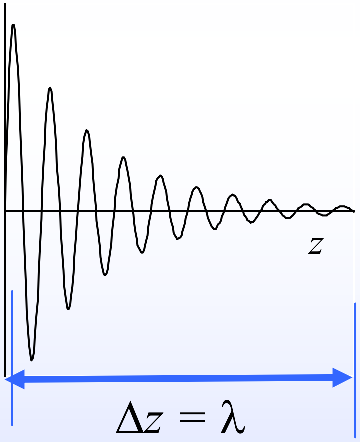
Why Soft-X-Ray ARPES ($h\nu$ ~ 500-1500 eV)?



Virtue 1: Increasing λ

- increasing bulk sensitivity
- buried impurities, interfaces and heterostructures

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



- reduction of Δk_z broadening
- sharply defined 3D **k**-vector
- combination with free-electron final states ⇒ 3D materials

Virtue 3: Elemental specificity through resonant photoemission

- combination with increasing λ ⇒ buried interfaces, heterostructures and impurities

Further virtues: Regular atomic-like matrix elements …

Challenges of SX-ARPES

• ΔE of a few tens meV (vs a few meV in VUV-ARPES)

• *e-ph* scattering destructive for **k**-resolution (photoelectron wavelength ~ thermal motion):

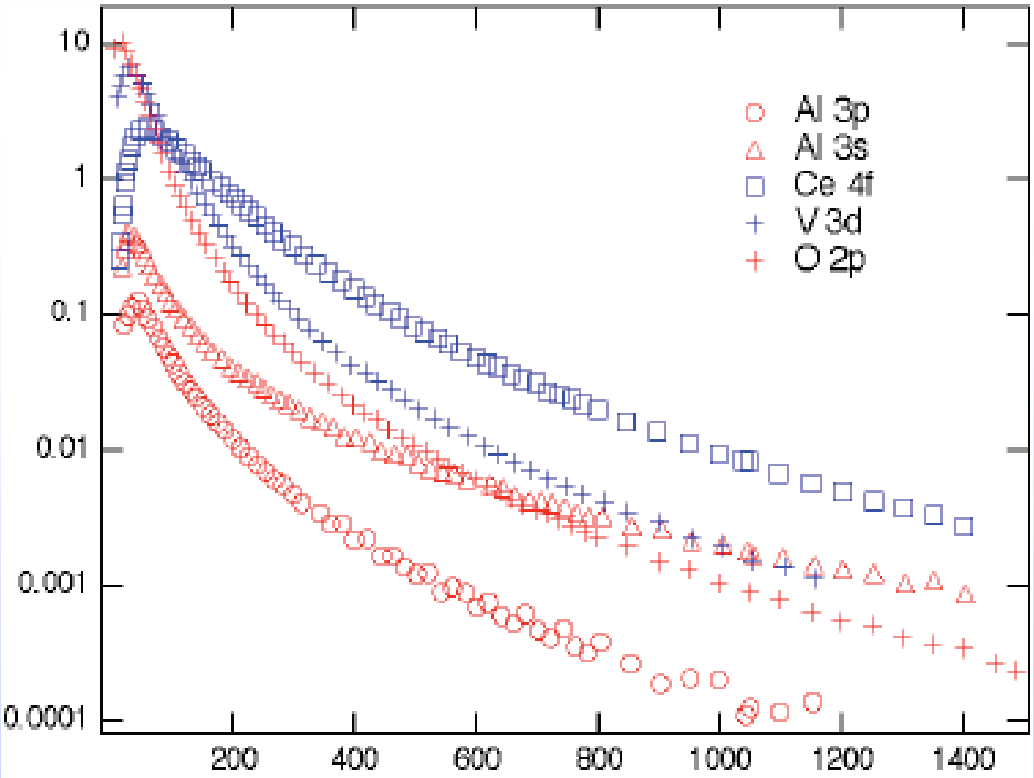
$$I^{coh} = W(T) I^{coh}_{T=0}$$

($W(T) = \exp\left(-\Delta G^2 U_0^2(T)\right)$, with $\Delta G^2 \propto E$ and $U_0^2 \propto T$)

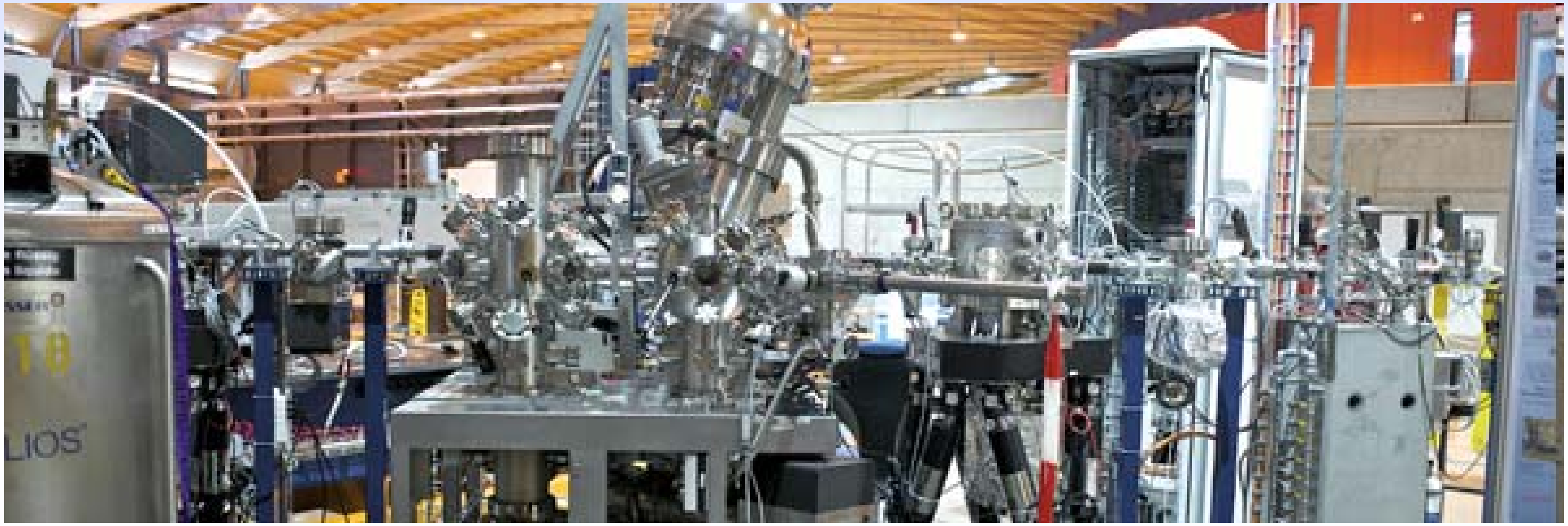
- liquid-He cooling

• loss of photoexcitation cross-section by 2-3 orders of magnitude

- efficient detectors and high photon flux instrumentation



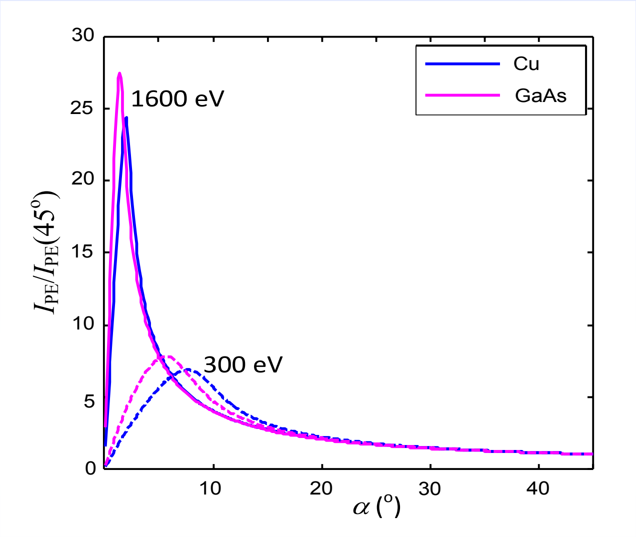
ADRESS (*ADvanced RESonant Spectroscopies*) Beamline at SLS



- RIXS (Resonant Inelastic X-ray scattering) and ARPES endstations
- soft-X-ray radiation with circular and 0-180^{o} variable linear polarizations
- energy range 300 – 1600 eV
- high resolution ΔE ~ 30 meV @ 1 keV
- collimated-light PGM optical scheme
- **flux up to 1.5x10^{13} ph/s/0.01%BW:** factor of 10 to 100 increase compared to best available beamlines ⇒ **breakthrough of the cross-section problem**

SX-ARPES Endstation @ ADRESS: Geometry

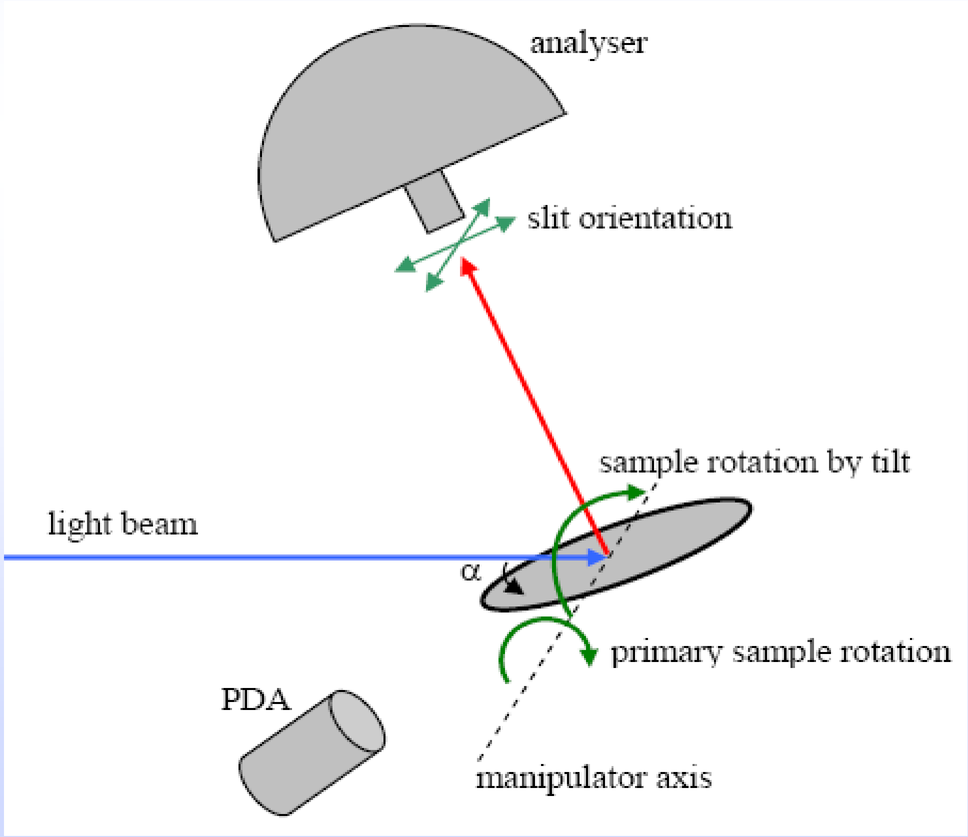
- **grazing incidence** at 20° to increase photoyield (factor of 2 compared to 45°)



V.S., J. Synchr. Rad **20** (2013)

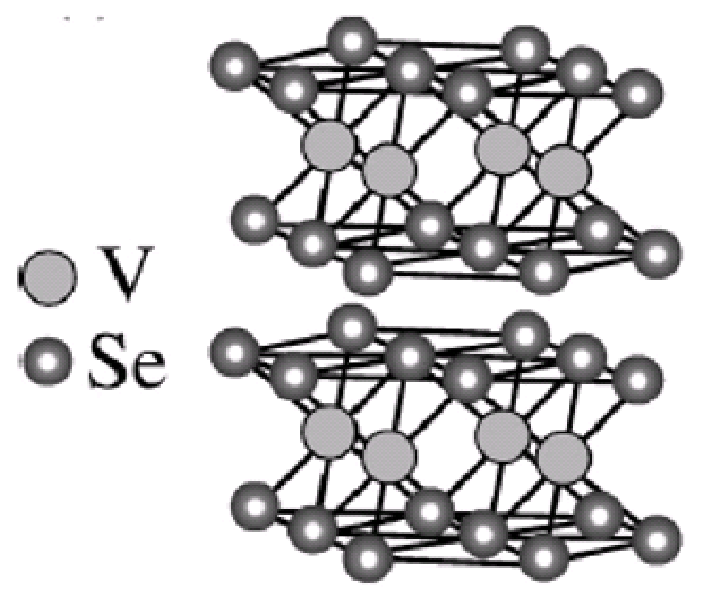
- **horizontal rotation axis** to balance the vertical (<20μm) and horizontal (74μm) light footprint

- rotatable analyzer: **parallel slit orientation** ⇒ symmetry analysis of the valence states



3D k-vector definition of SX-ARPES: Bandstucture and Fermi surface of VSe_2

- quasi-2D structure with weaker interlayer interaction
- significant 3D-lity due to Se $4p_z$ orbitals

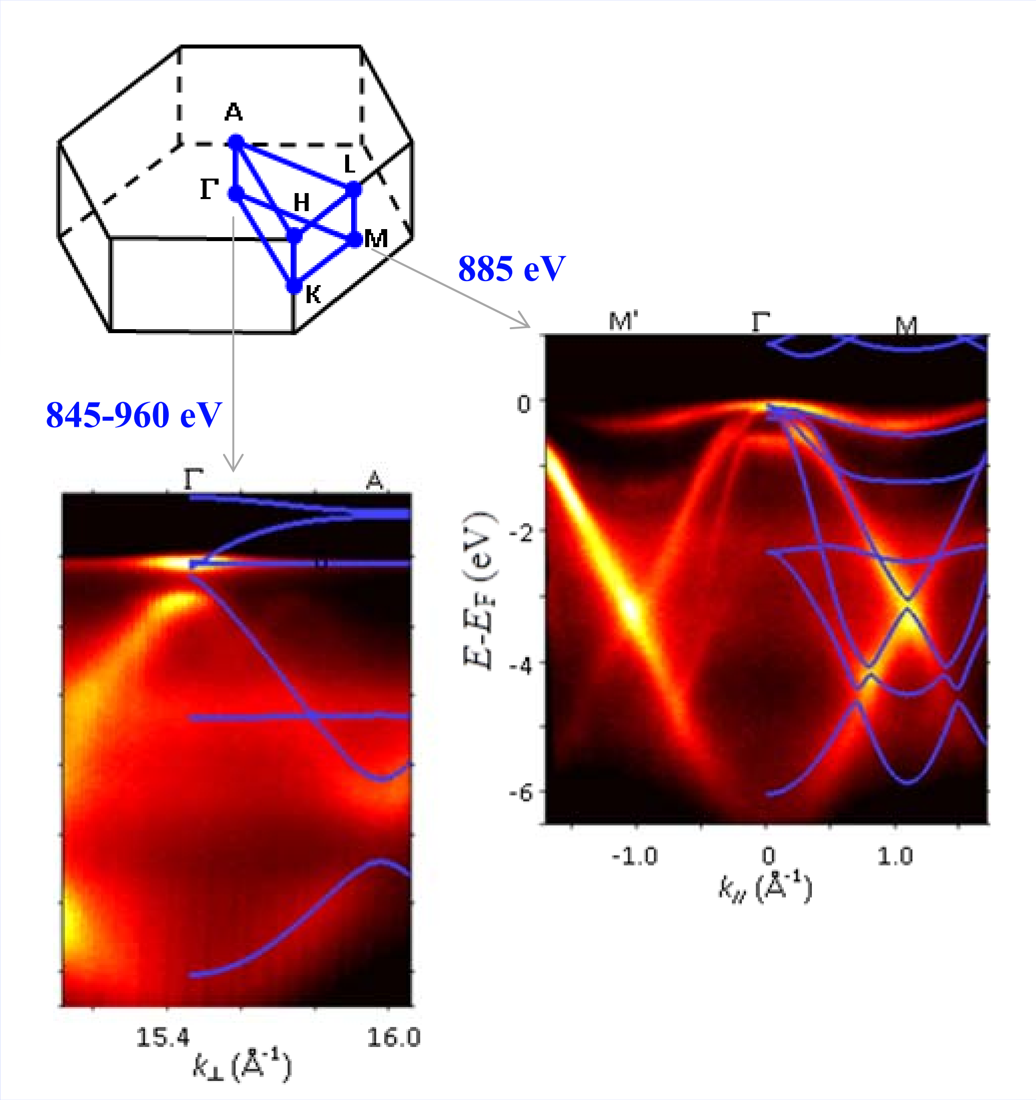


- Experimental *E*(k)

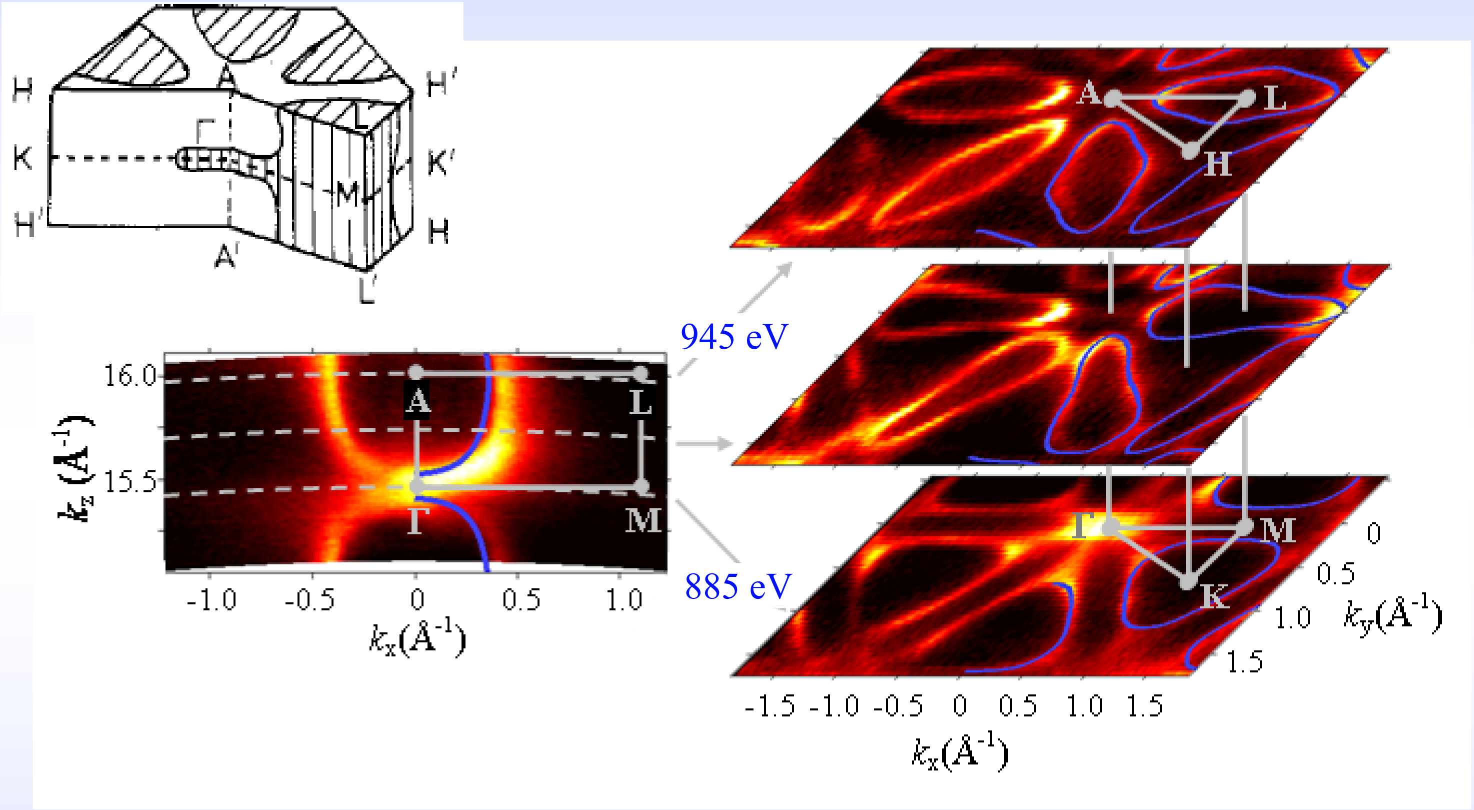
- k_z by varying *hv* around 900 eV
- ΔE~110 meV

⇓

- excellent statistics in a few min despite the cross-section loss (~1800 for V3*d* and 35 for Se 4*p* vs *hv*=50eV)
- intense and sharp structures => *e-ph* scattering effects (spectral weight transfer to 3D-DOS and **k**-broadening) are insignificant at our T=10.7K despite low T_D=220K
- significant 3D-lity of Se $4p_z$ orbitals
- agreement with GGA-DFT (*P. Blaha, TU Wien*)



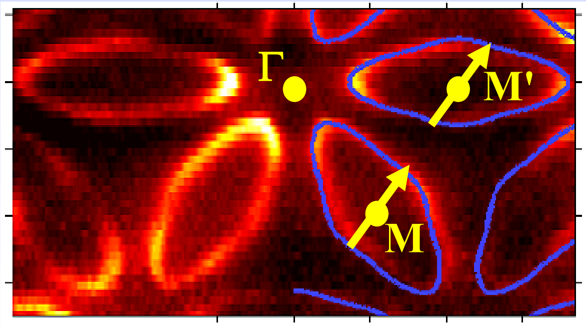
- Experimental 3D Fermi surface of VSe_2



• extraordinary clarity of the experimental data (no image enhancement):
Sharp definition of 3D wavevector and regular matrix elements in soft-X-ray energies

• agreement with GGA-DFT (*P. Blaha, TU Wien*)

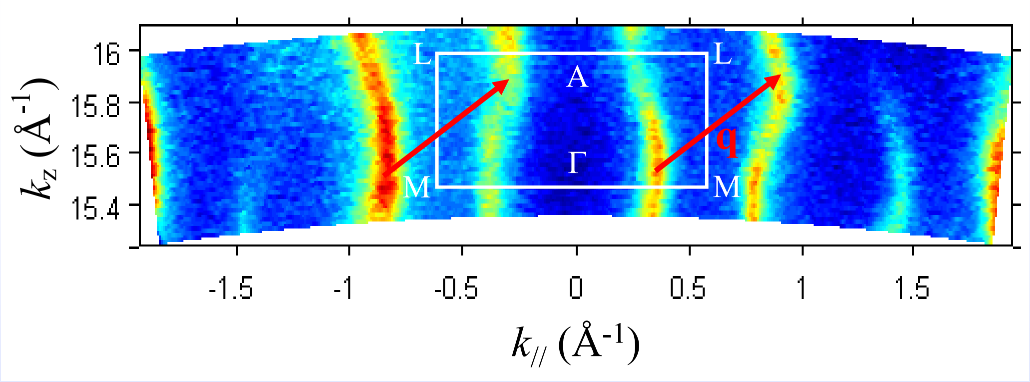
- Origin of 3-dimensional CDWs

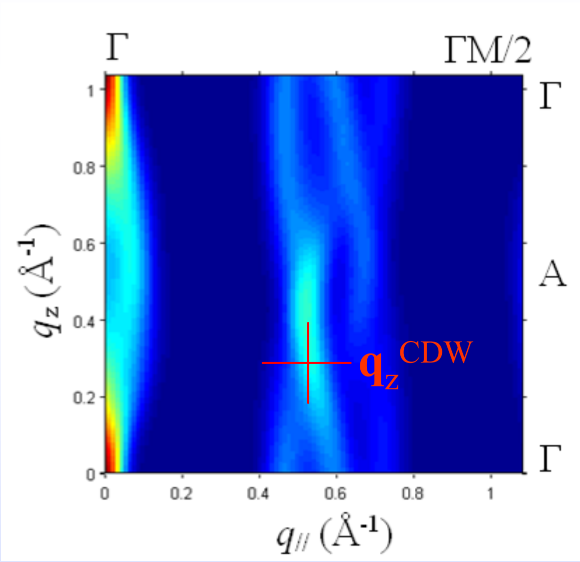


- Unusual 3-dimensionality of CDWs:

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

- Perpendicular FS cut in MLL'M' plane



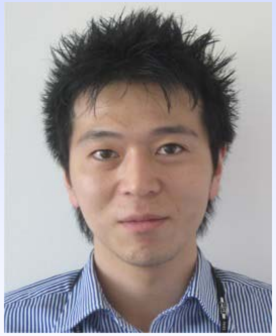


- 3D warping to support nesting close q_z^{CDW}

- Autocorrelation peak at $\mathbf{q}_{//}^{CDW}$ and shifted to q_z^{CDW} by commensurization

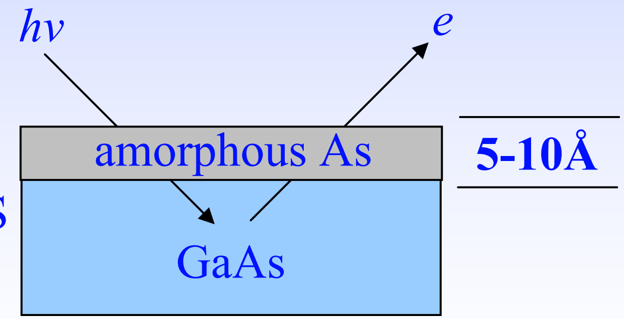
- V.S. *et al.*, PRL **109** (2012) 086401

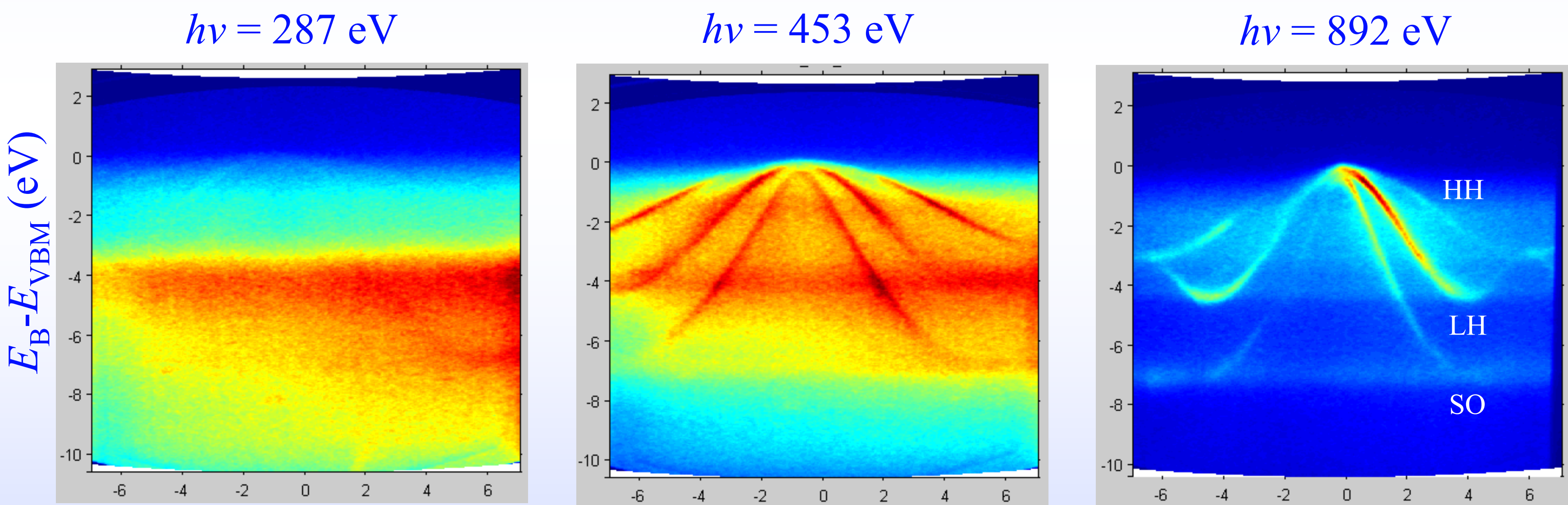
Penetrating ability of SX-ARPES: Band structure of GaAs through amorphous As layer



M. Kobayashi et al (SLS); samples: Uni Tokyo

- large λ required: Soft-X-ray ARPES

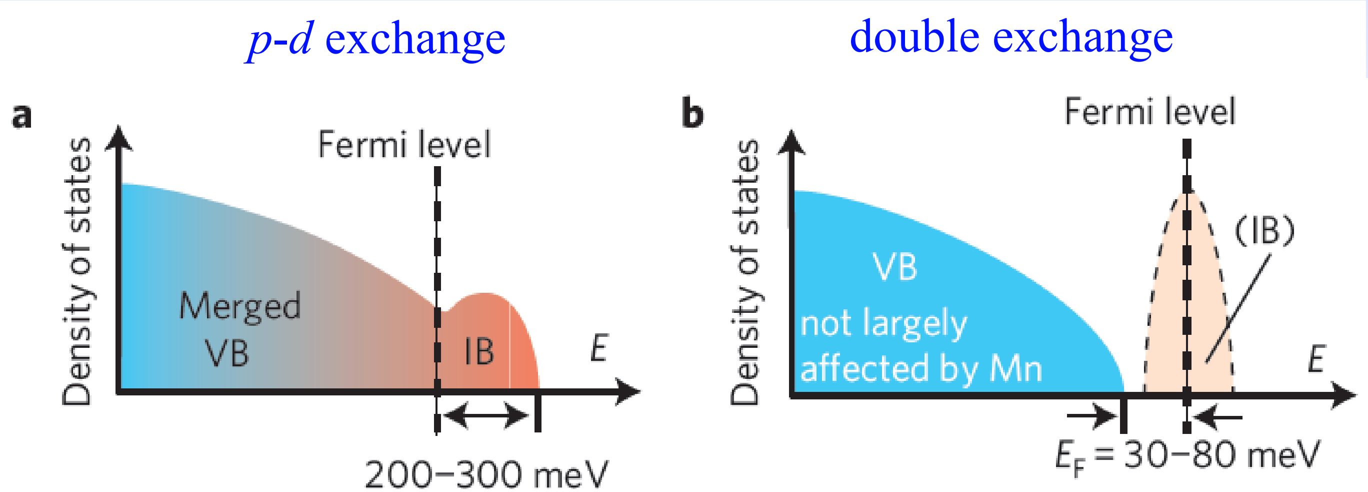




- acquisition time 3 min
- GaAs signal piles up with *hν*
- New diagnostics tool for MBE grown films: Applications in microelectronics

- M. Kobayashi *et al.*, Appl. Phys. Lett. **101** (2012) 242103

Ferromagnetism of diluted magnetic semiconductor GaMnAs

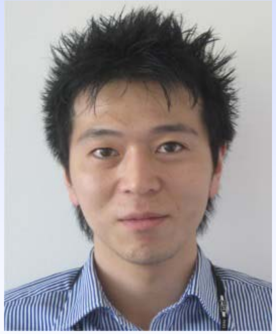


S. Ohya *et al*., Nat. Phys. (2011).

• Role of impurity vs band states in the ferromagnetism of GaMnAs:
– does Mn form an impurity band?
– energy alignment of the Mn impurity band?
– hybridization with the host GaAs bands?

• HAXPES study in A.X. Gray *et al,* Nature Mat. 11 (2012) 957:
Mn weight below E_F; Ferromagnetism as a combination of the two models

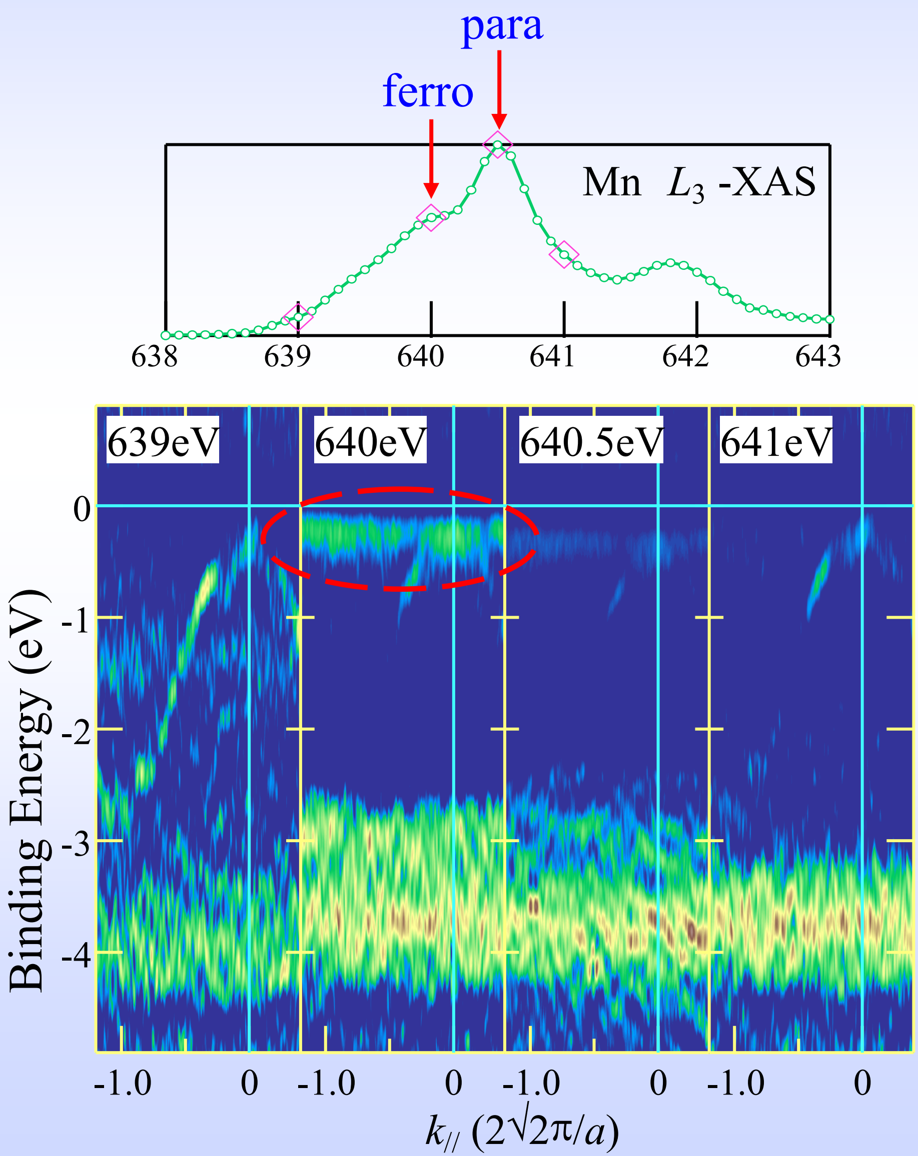
Elemental specificity of SX-ARPES: Resonant ARPES of diluted magnetic semiconductor GaMnAs



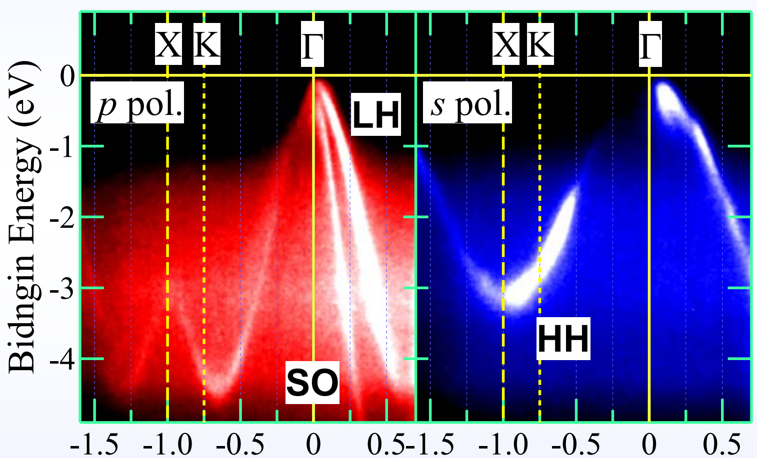
M. Kobayashi et al (SLS); samples: Uni Tokyo

- Mn concentration only 2.5% of Ga
⇒ hard to see unless resonantly enhance Mn 3*d* weight

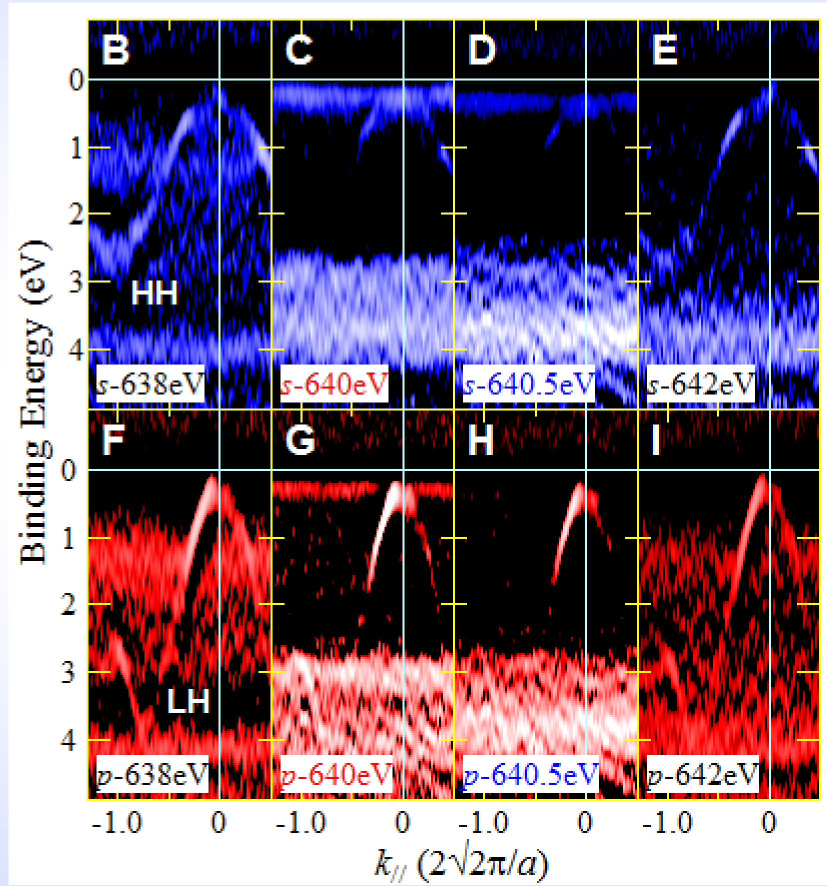
- Resonance on ferromagnetic XAS peak
⇒ ferromagnetic non-dispersive Mn 3*d* impurity band just below the VBM



- Linear dichroism



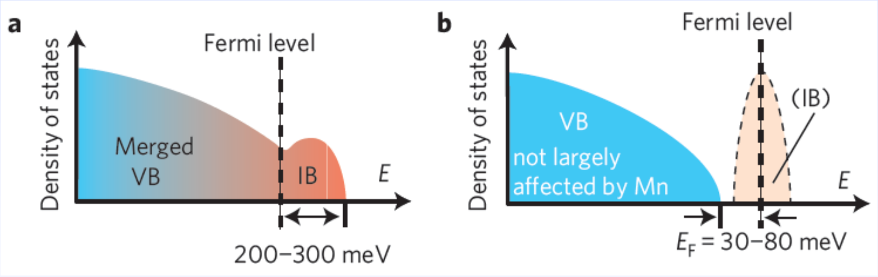
ferro
para

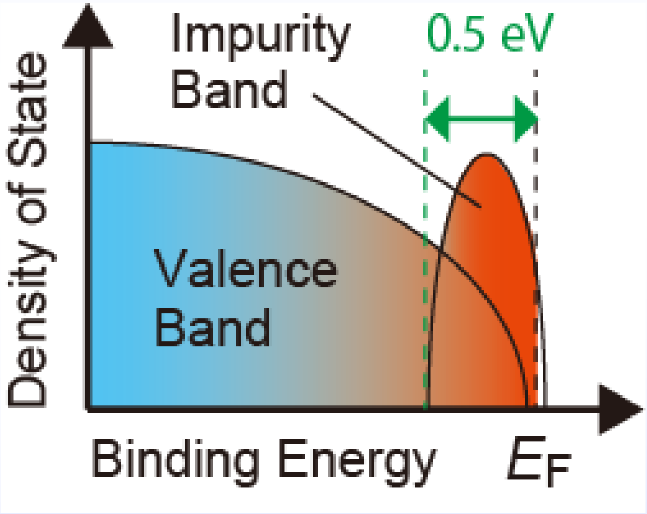


- Intensity at the ferromagnetic resonance

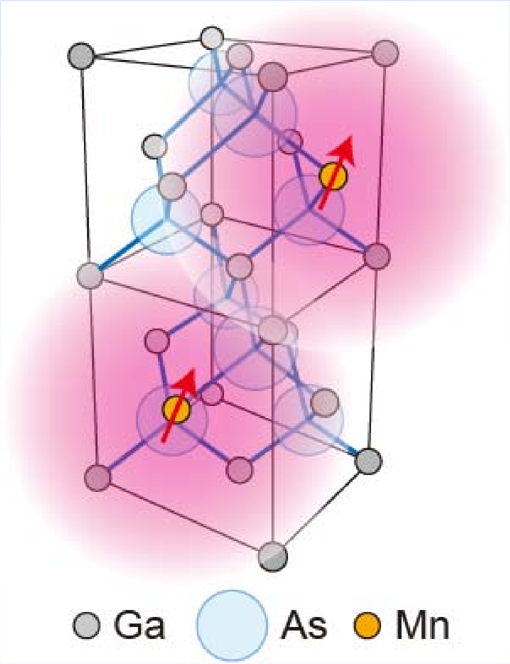
⇒ Mn 3*d* impurity band hybridizes with LH but only weakly with HH band

Ferromagnetism of diluted magnetic semiconductor GaMnAs





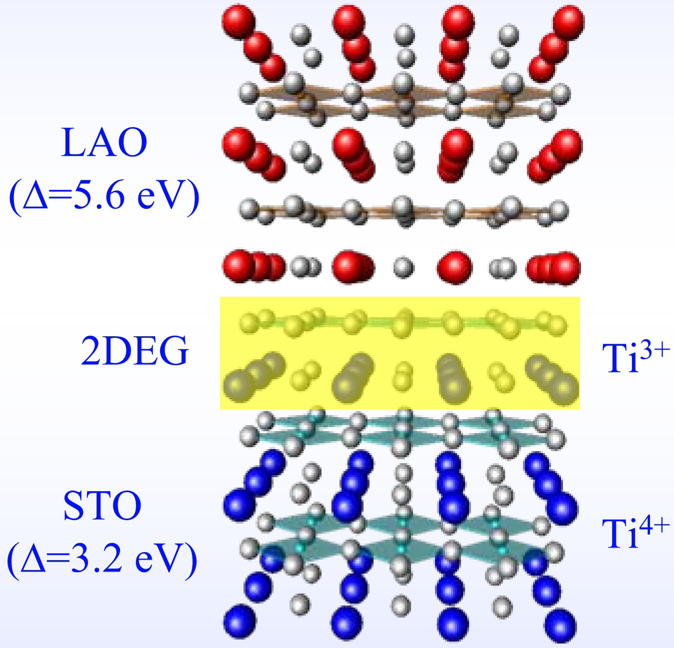
- Occupied Mn 3*d* impurity band hybridizing with GaAs host band



- Ferromagnetism induced by GaAs mediated exchange between Mn atoms
- Description starting from the Anderson impurity model

- M. Kobayashi *et al.*, http://arxiv.org/abs/1302.0063 (submitted to Phys. Rev. Lett.)

Buried interfaces: Resonant SX-ARPES of $LaAlO_3/SrTiO_3$



2DEG at the LAO/STO interface:

- electrons delivered by Ti^{3+} ions
- Critical LAO thickness > 4 u.c.

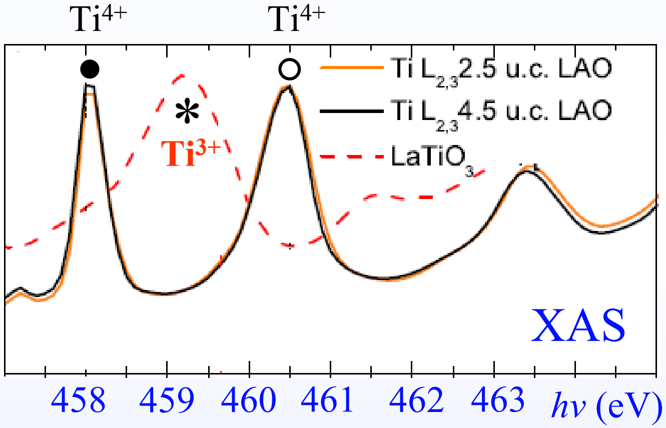
⇒ SX-ARPES required

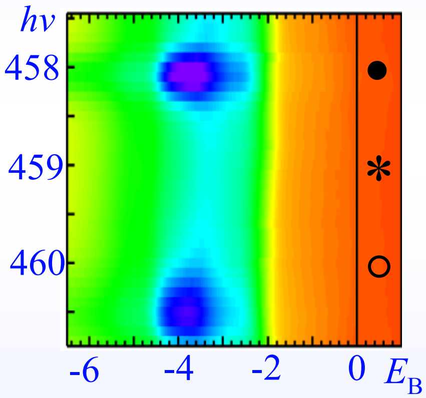
- Idea: Use Ti^{3+} **resonant** SX-ARPES to enhance the 2DEG signal

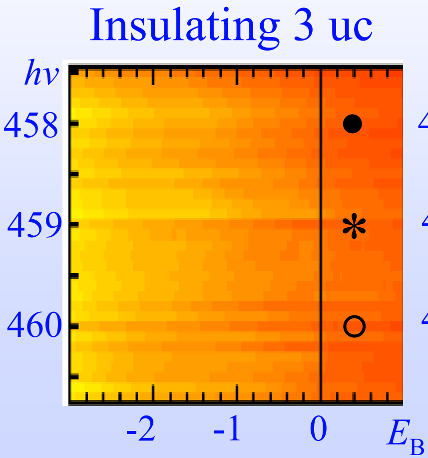
Depth Profiling of 2DEG at the $LaAlO_3/SrTiO_3$ interface

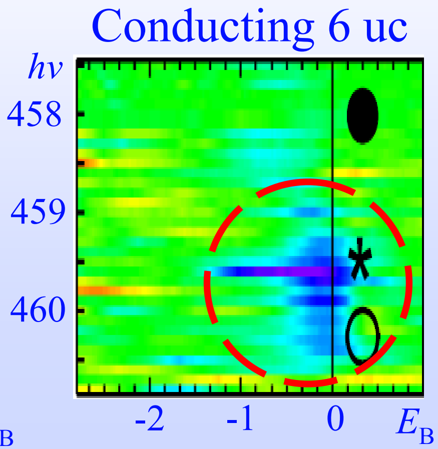
C. Cancellieri, M. Reinle-Schmitt et al; samples: Uni Geneve

- measurements @ RT ⇒ averaging in **k**-space
- comparison of insulating (3 uc LAO) and conducting (6 uc LAO) samples



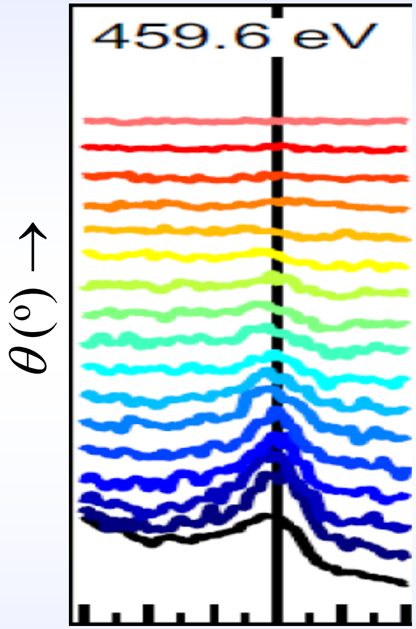






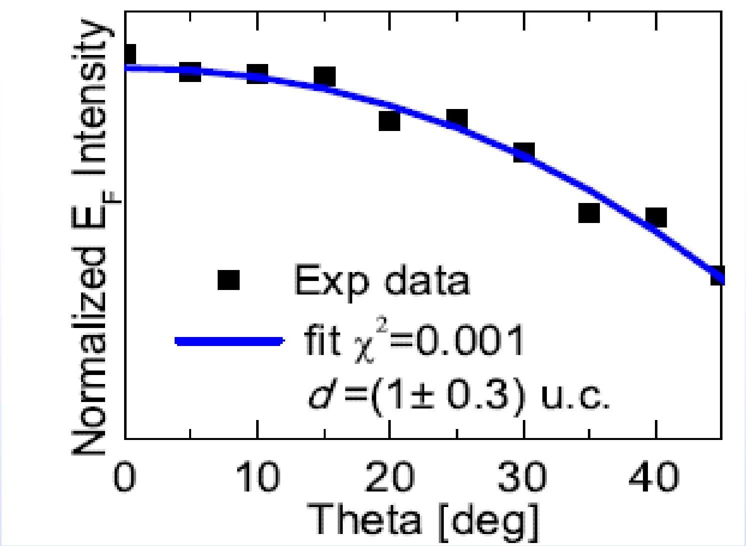
- Ti^{3+} "conductivity" peak near E_F ⇒ **2DEG**

- Angle-dependent XPS to resolve the 2DEG depth profile



$$I(\theta) = G(\theta, \phi) \int_0^\infty R(z) e^{-z/\lambda \cos\theta} \, \mathrm{d}z$$

- $I(\theta)$ = Laplace transform of $R(z)$



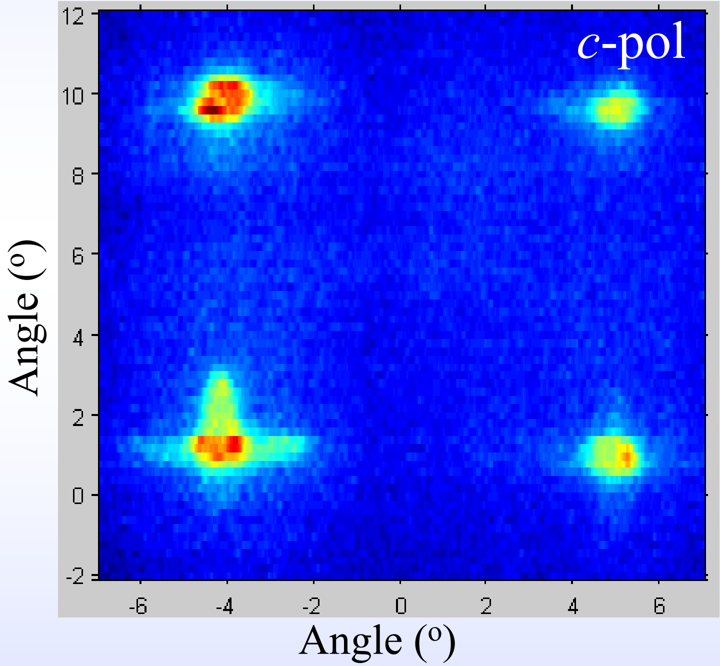
- **k**-integration due to RT

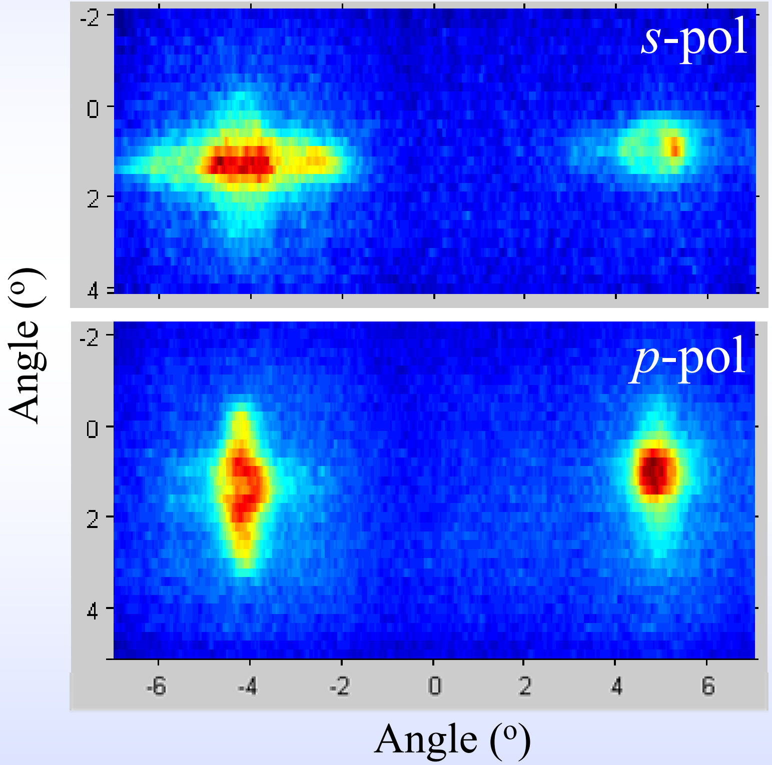
- 2DEG is located within **1.0±0.3 u.c.** on the STO side of the interface

- C. Cancellieri *et al.*, PRL **110** (2013) 137601

Fermi surface and bandstructure of the interface states

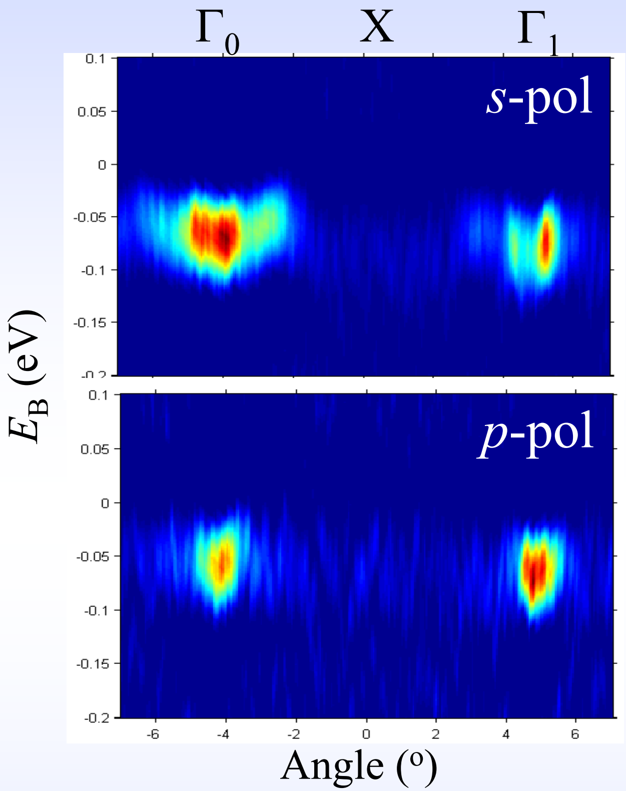
- T = 11 K to suppress *e-ph* scattering
- FS mapping at 2*p* resonance of Ti^{3+} at $h\nu$ = 460.2 eV
- Experimental ΔE ~ 80 meV

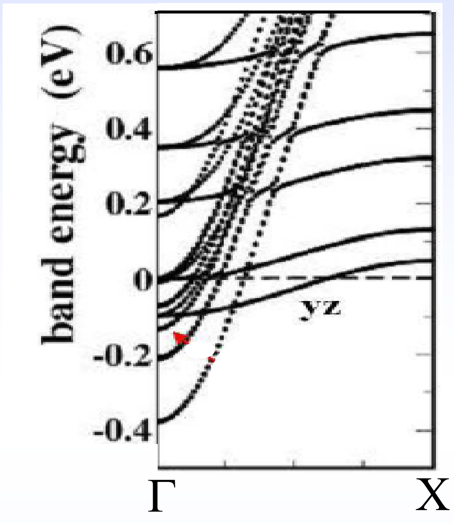




- FS shape of crossed $3d_{xy}$-like cigars (G. Berner *et al*, Phys. Rev. Lett. **110** (2013) 247601 - talk of M. Sing; N. Plumb *et al*, arXiv:1304.5948)
- different FS sheets depending on $\mathbf{K}_{//}$ and polarization

- Subband structure of the interface state





A. Filippetti, P. Ghosez & D. Fontaine

- composite interface state with subbands of different symmetries
- different sample preparations:

- Luttinger count of the FS area follows n_e from transport properties ⇒ coherent interface conductivity with insignificant contribution of ox-vacancies
- interface charge varies and differs from 0.5 *e*/u.c. (deviations from both structural deformation and polar catastrophe model)

- C. Cancellieri *et al.*, arXiv:1307.6943 (2013)

Comparison of VUV- and SX-ARPES

	VUV-ARPES	SX-ARPES
ΔE	meV-scale: quasiparticle interactions	tens-of-meV scale global valence band
probing depth λ	few Å: simple surfaces	~ 15 Å: bulk and buried heterostructures
k_z definition	$\Delta k_z \sim k_z^{BZ}$ and non-FE final states: mostly 2D systems	$\Delta k_z << k_z^{BZ}$ and FE final states: 3D systems

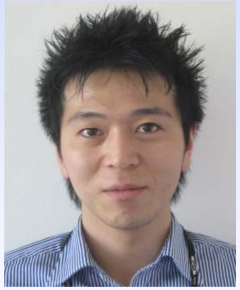
- SX-ARPES spectroscopic properties:

- 3D bulk band structure (FS and CDWs in VSe_2, FS in perovskite LSMO…)
- Enhanced probing depth: Band structure of GaAs behind thick cap layer …
- Elemental specificity through resonant photoemission: Ferromagnetic impurity band in GaMnAs, depth localization and Fermi surface of LAO/STO …
- Depth profiling with standing X-ray waves excited ARPES

- Flux performance of ADRESS ⇒ **From 3D materials to buried heterostructures**

SX-ARPES team

V.N.S.
(BL Scientist)



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J. Minar
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C. Fadley
(UC Davis)

…

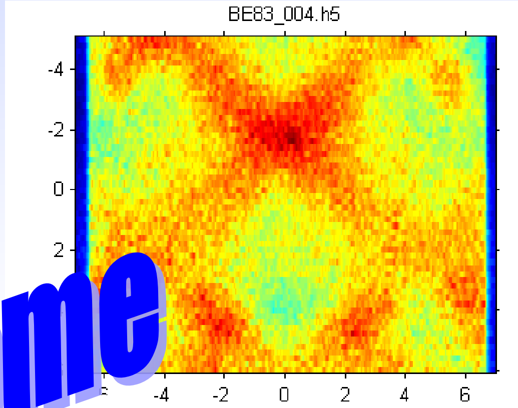
Support from PSI

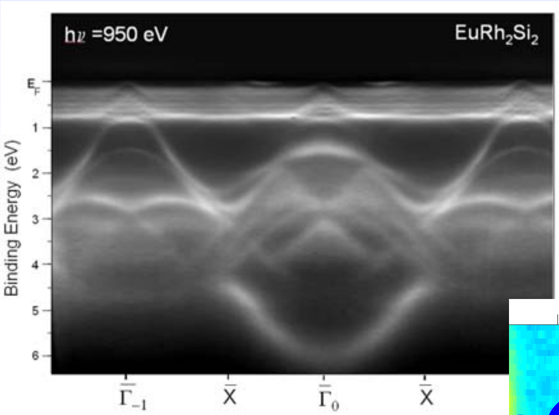
Optics (group of U. Flechsig), ID (group of T. Schmidt),
Controls (X. Wang, J. Krempasky) *et al*

www.psi.ch/sls/adress/

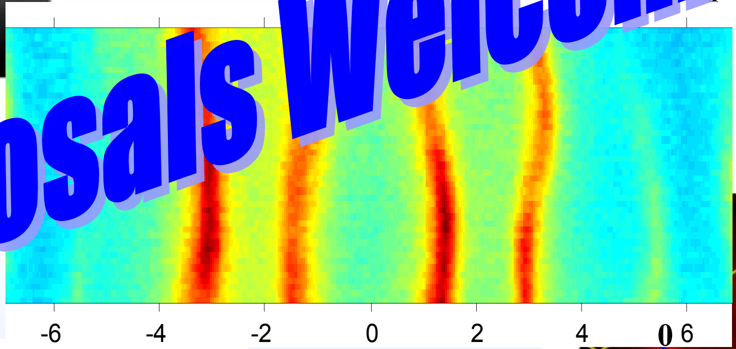
PAUL SCHERRER INSTITUT PSI

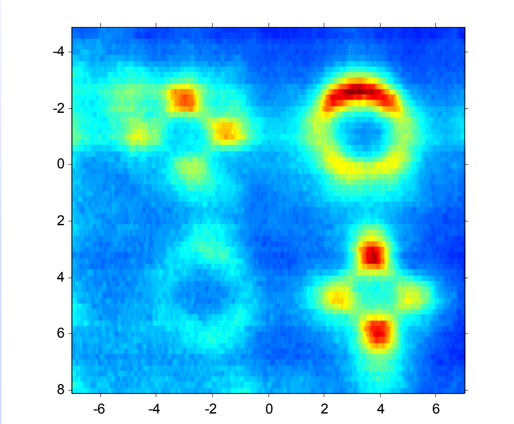
SWISS LIGHT SOURCE SLS

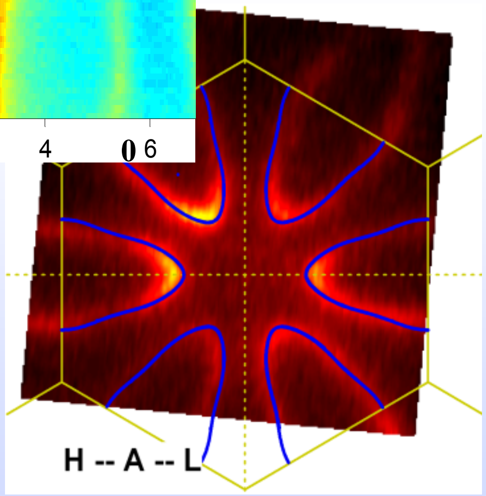




Proposals Welcome







Next call: September 15