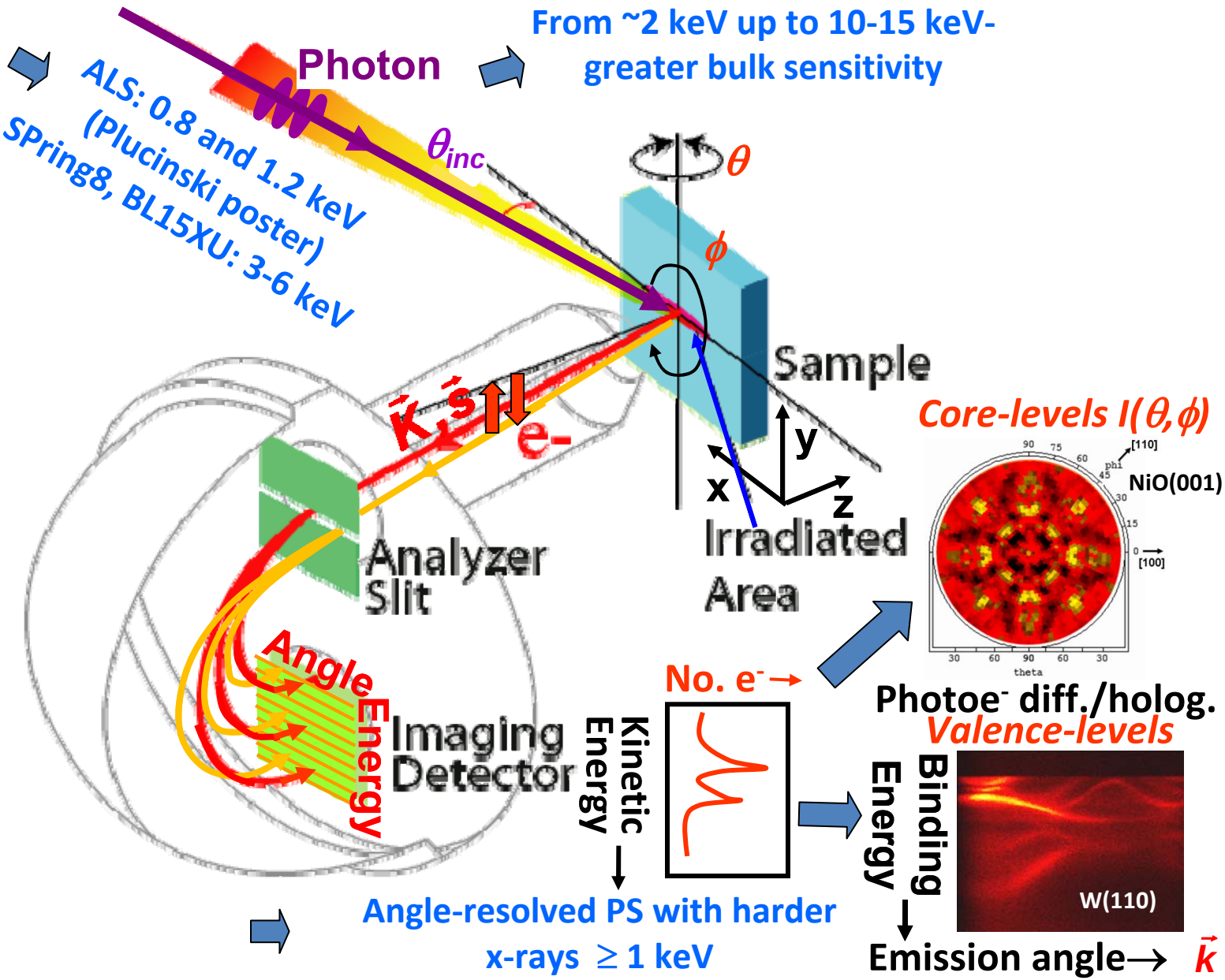
Angle-resolved photoemission in the multi-keV regime: first experiments and theory

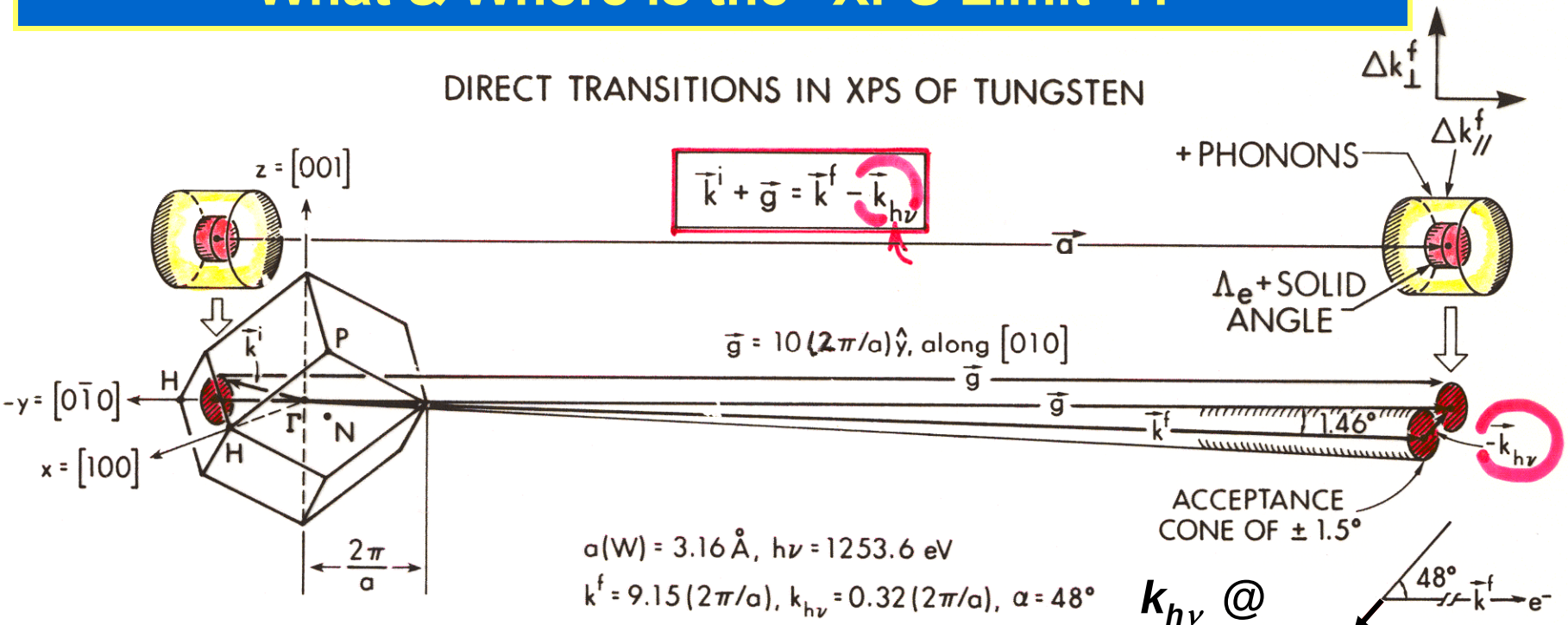
C. Papp, A. Gray, B. Balke, **S. Ueda,** K. Kobayashi, S. Sakai, H. Yoshikawa, Y. Yamashita, S. L. He, **L. Plucinski,** C. Schneider, **J. Minar,** J. Braun, H. Ebert , C. S. Fadley

Materials Sciences Division, LBNL; Physics, UC Davis; SPring8; National Institute for Materials Science, Tsukuba; Physical Chemistry, University of Munich, Germany; Solid State Physics, Jűlich Research Center, Germany

Looking at buried layers, interfaces, bulk properties with photoemission



Angle-Resolved Photoemission at High Energy--What & Where is the "XPS Limit"?:



$k_{h\nu}$ @ 1,253 eV

$k_{h\nu}$ @ 10,000 eV

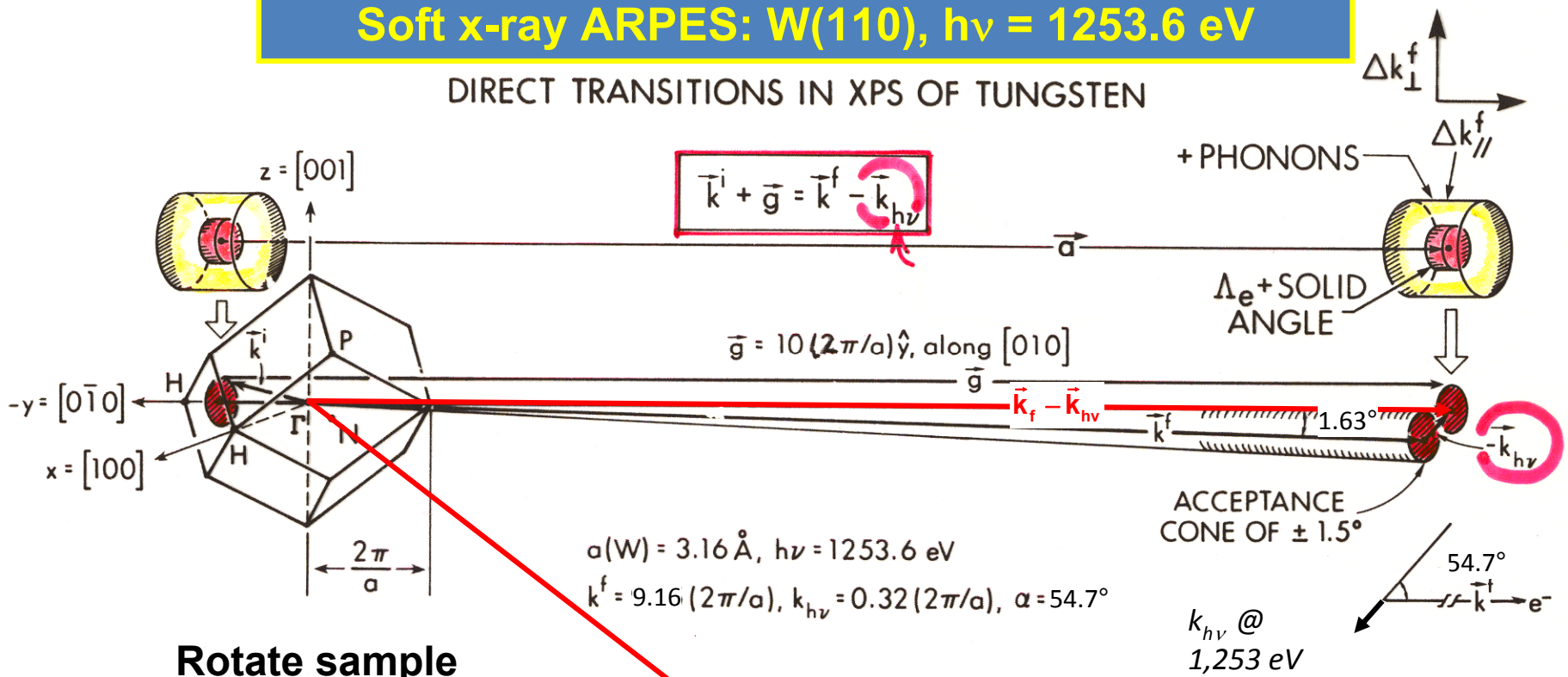
Additional effects at higher energies:

- **non-dipole--the photon momentum**
- **angular acceptance→B.Z. averaging**
- **lattice recoil, phonon creation→more B.Z. averaging, ~Debye-Waller factor =** $W(T) \approx \exp[-(k^f)^2 \langle u^2(T)\rangle] \approx \exp(-C_1 E_{kin} T)$

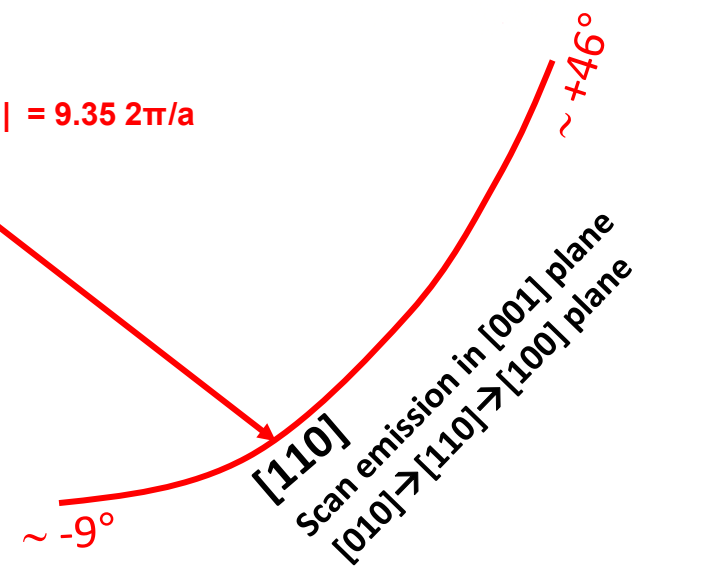
….the "XPS limit" of full B.Z. averaging and density-of-states (DOS) sensitivity

Hussain et al., Phys. Rev. B 22, 3750 ('80)

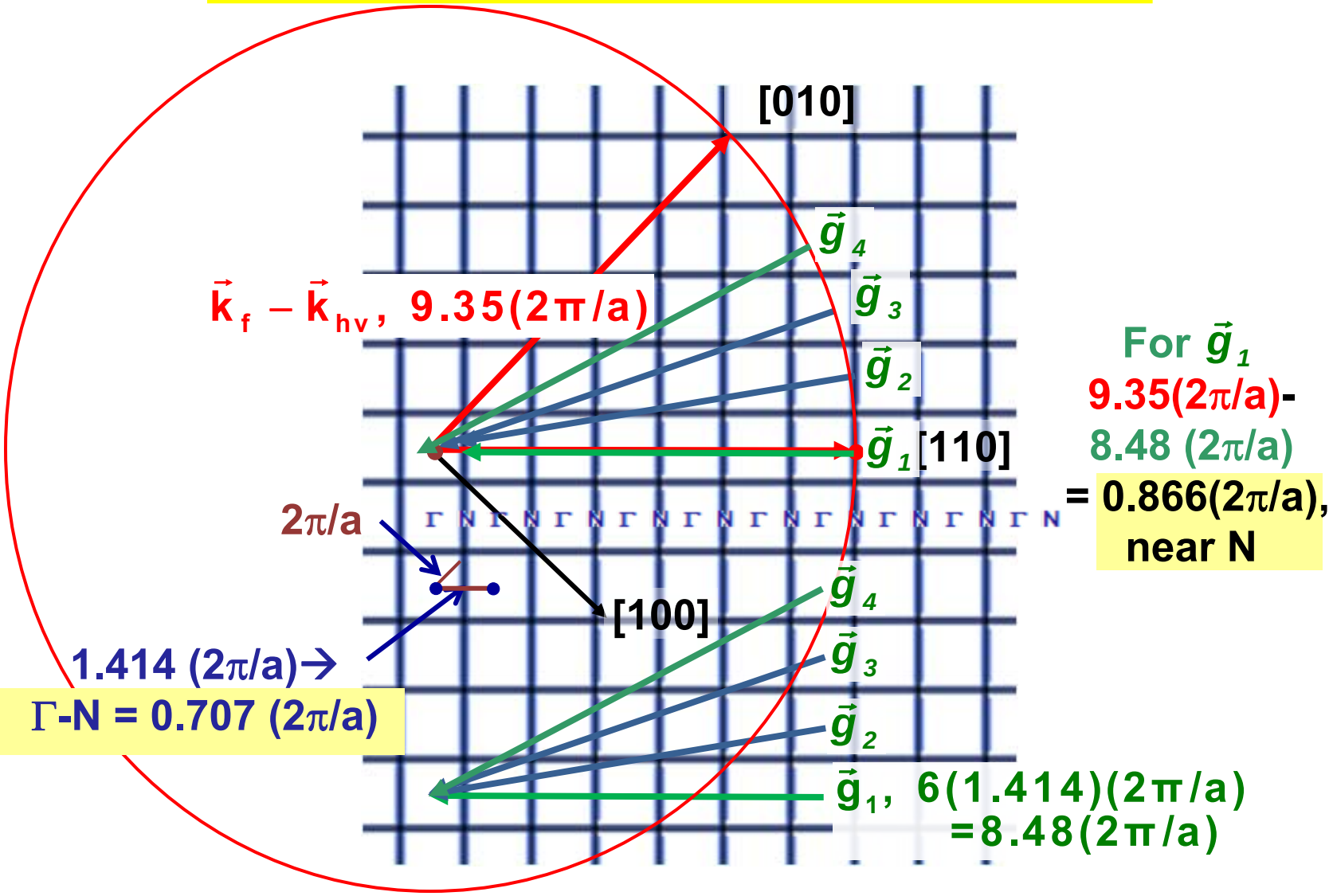
Soft x-ray ARPES: W(110), hν = 1253.6 eV



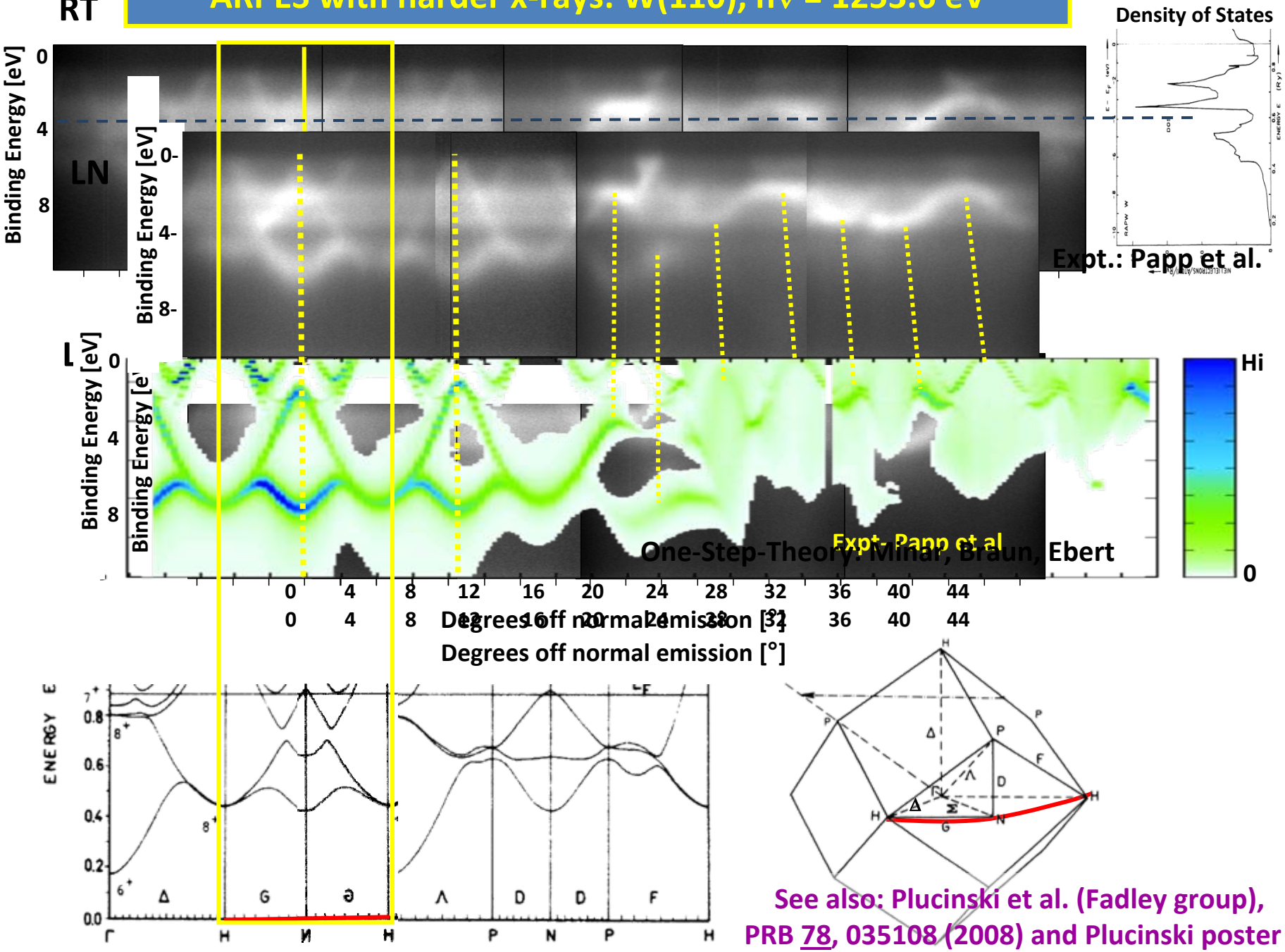
Rotate sample
Angle-resolving detector fixed w.r.t. light



Soft x-ray ARPES in an extended zone scheme: W(110), [001] plane, hν = 1253.6 eV



ARPES with harder x-rays: W(110), hν = 1253.6 eV



See also: Plucinski et al. (Fadley group), PRB 78, 035108 (2008) and Plucinski poster

Hard X-Ray ARPES: W(110), hν = 5945 eV
SPring8, BL15XU, Ueda et al.

The free-electron picture:

$(2\pi / a) = 1.988\ A^{-1}$

$h\nu = 5945\ eV$

$|\vec{k}_f| = 0.512E_{kin}^{0.5}(eV) = 39.48\ A^{-1} = 19.85\ (2\pi / a)$

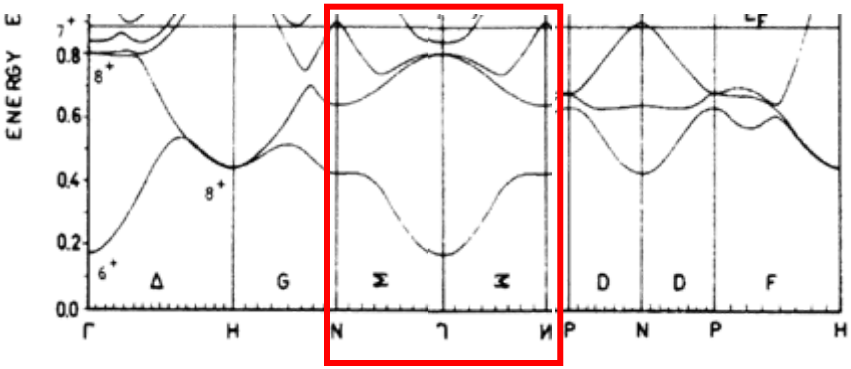
$|\vec{k}_{h\nu}| = 0.000507(h\nu(eV)) = 3.01\ A^{-1} = 1.51\ (2\pi / a)$

$|\vec{k}_f - \vec{k}_{h\nu}| = \left[39.48^2 + 3.01^2\right] = 39.59\ A^{-1} = 19.91\ (2\pi / a) --> 14.08\ [1.414((2\pi / a)]$

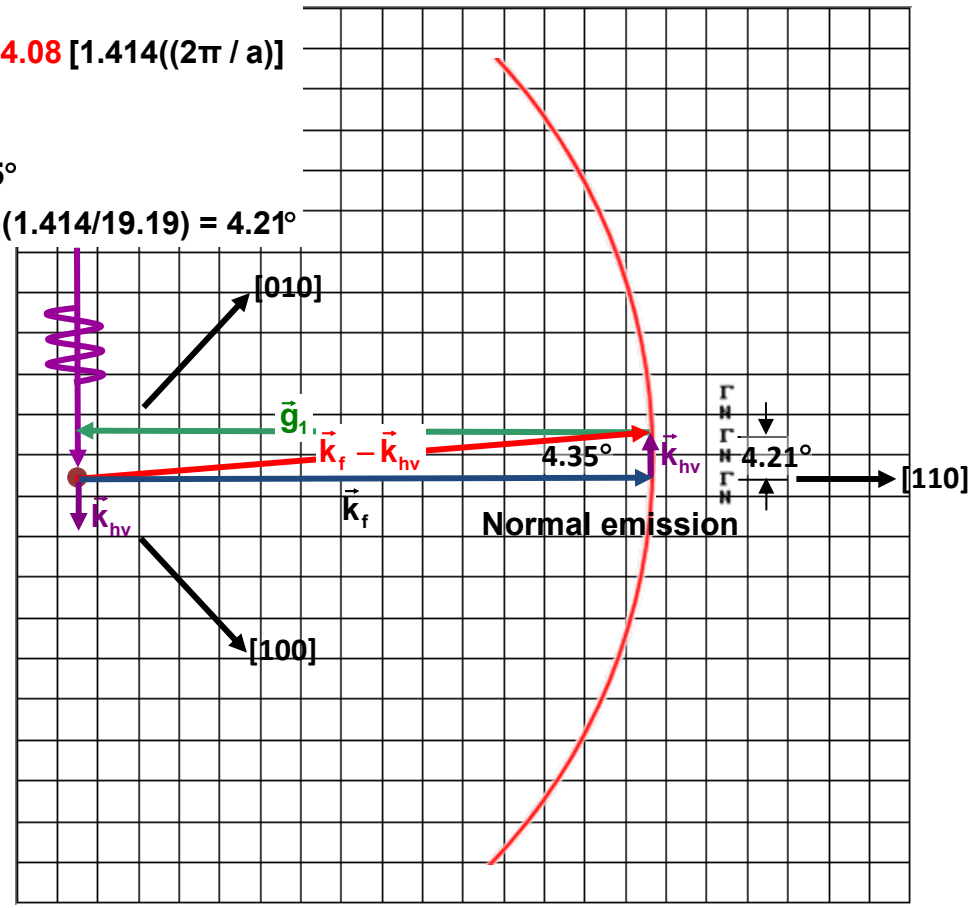
$|\vec{g}_1| = 14.00\ [1.414((2\pi / a)]$

Angular deflection due to $\vec{k}_{h\nu}$ is : $\arctan(3.01 / 39.48) = 4.35°$

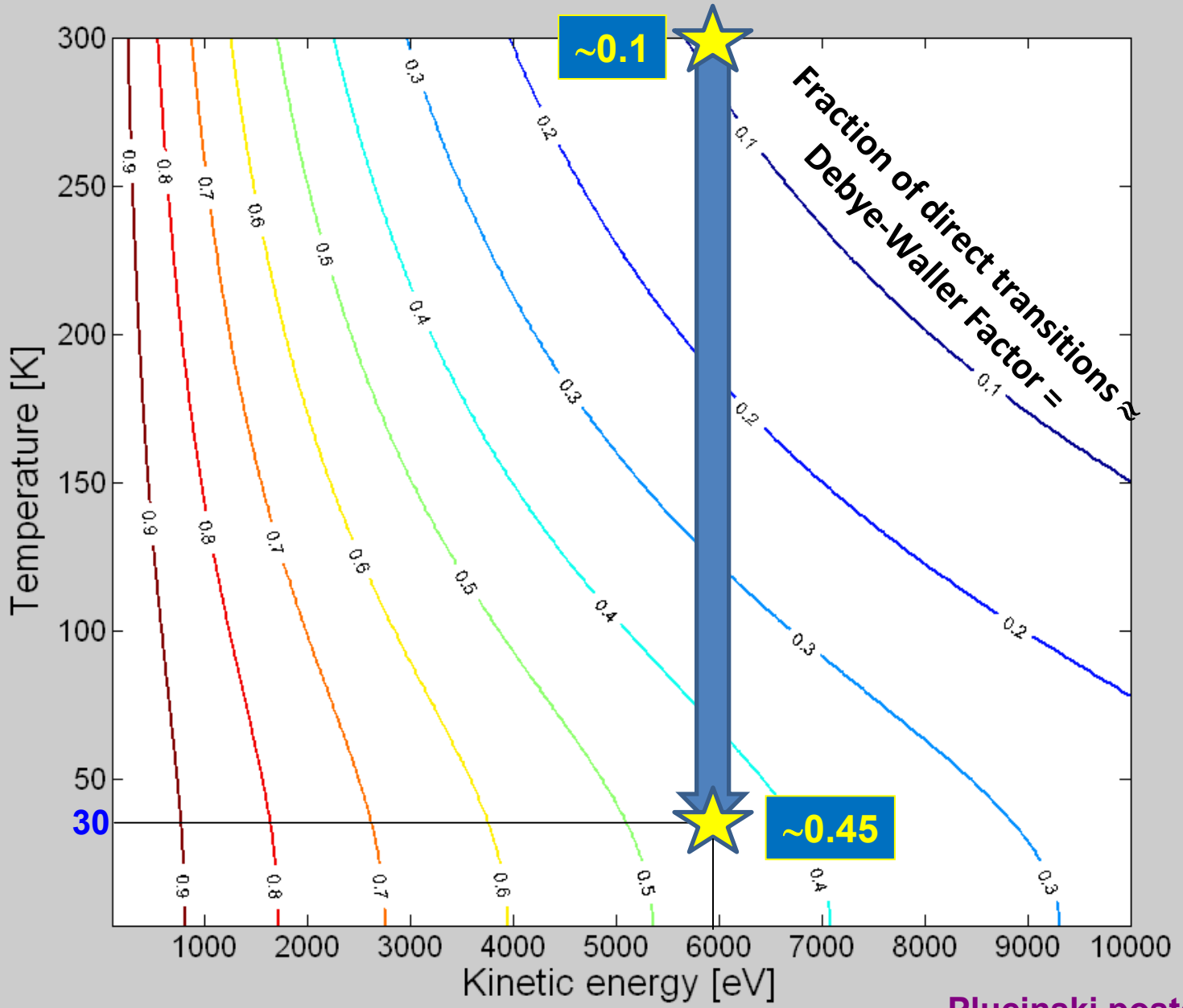
$\Gamma - N - \Gamma$ is $1.414(2\pi / a)$, so angular range of this is $\arctan(1.414/19.19) = 4.21°$



4.21°



Phonon effects—Qualitative systematics:Tungsten, 5954 eV

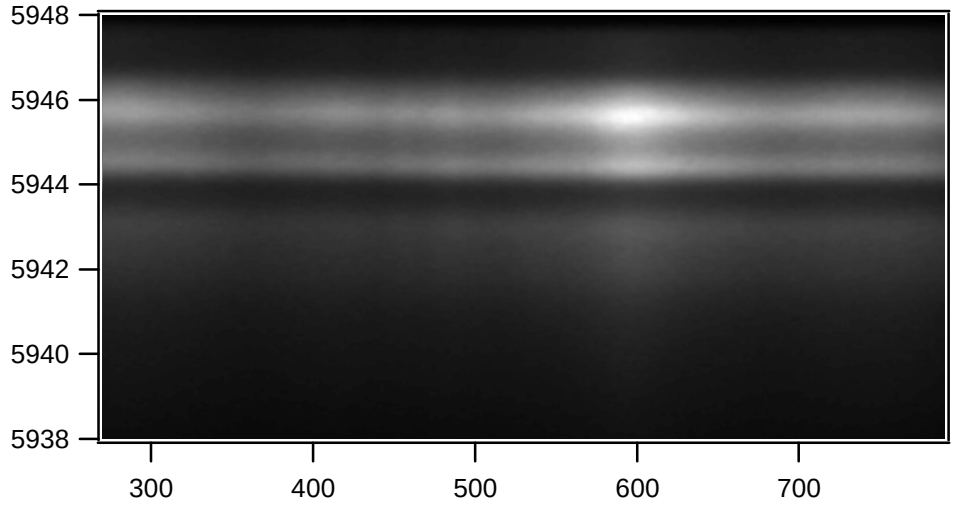


Plucinski poster

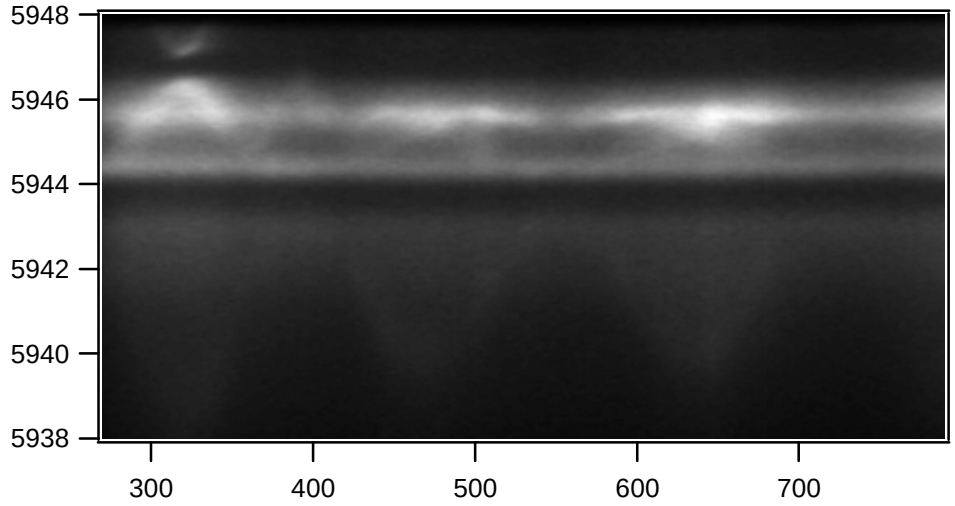
W(110)-Raw data, Temperature Dependence 5954 eV

39 channels = 1 deg

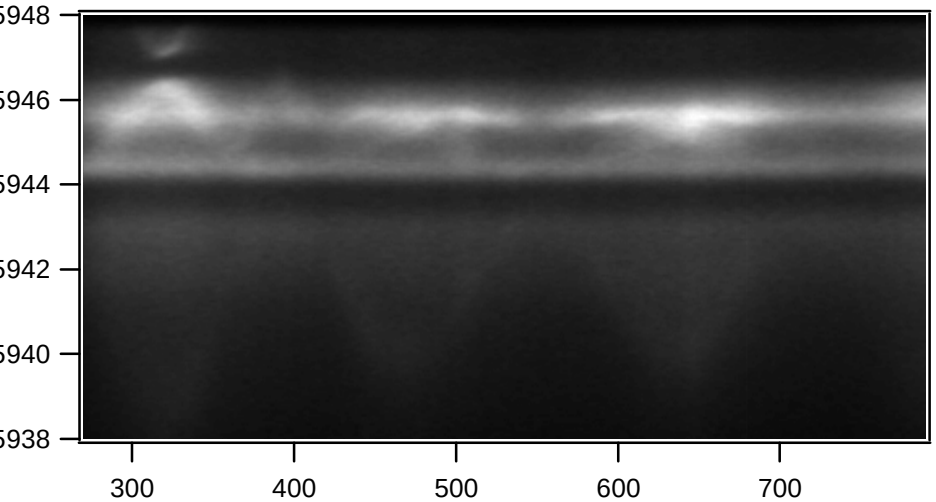
Room temperature: Density of states ("XPS limit") + x-ray photoelectron diffraction (Winkelmann poster)



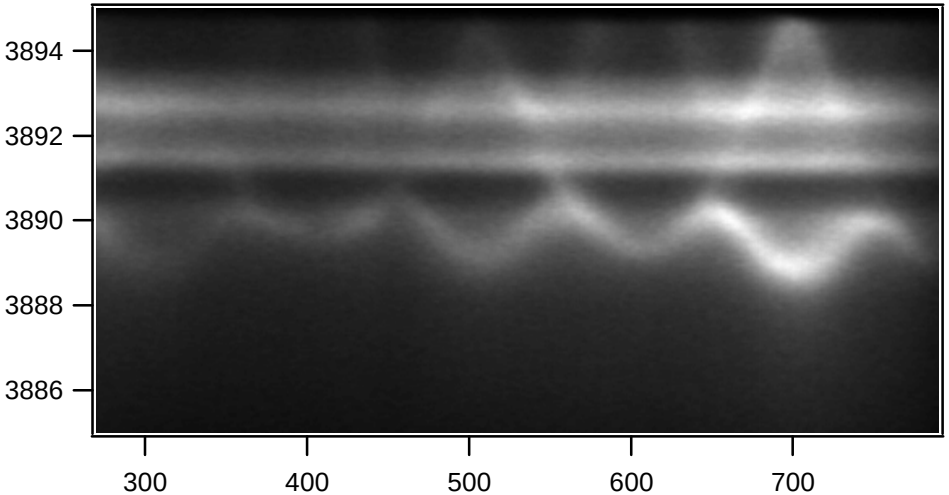
30 K: Mix of bands HX-ARPES and DOS+XPD



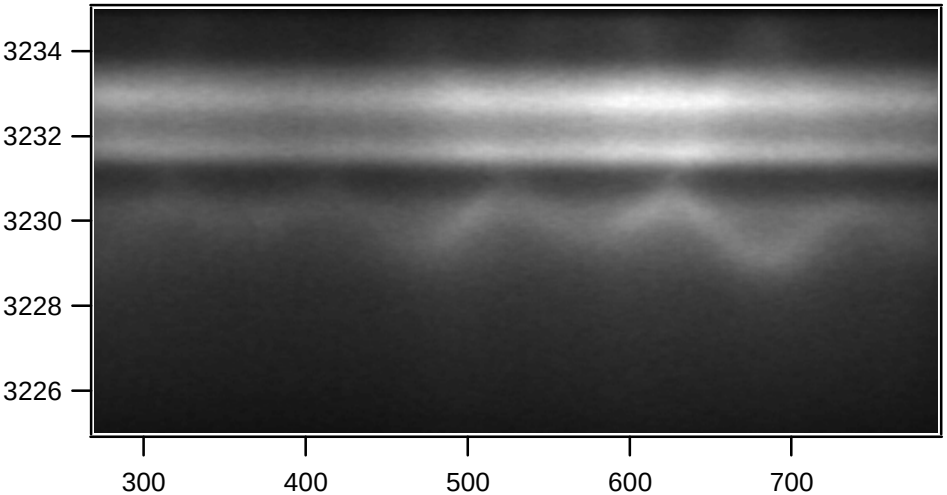
W(110)- Raw data, Dependence on photon energy: 6 keV, 3.9 keV, 3.2 keV



6 keV
30 K
DW ≈ 0.45

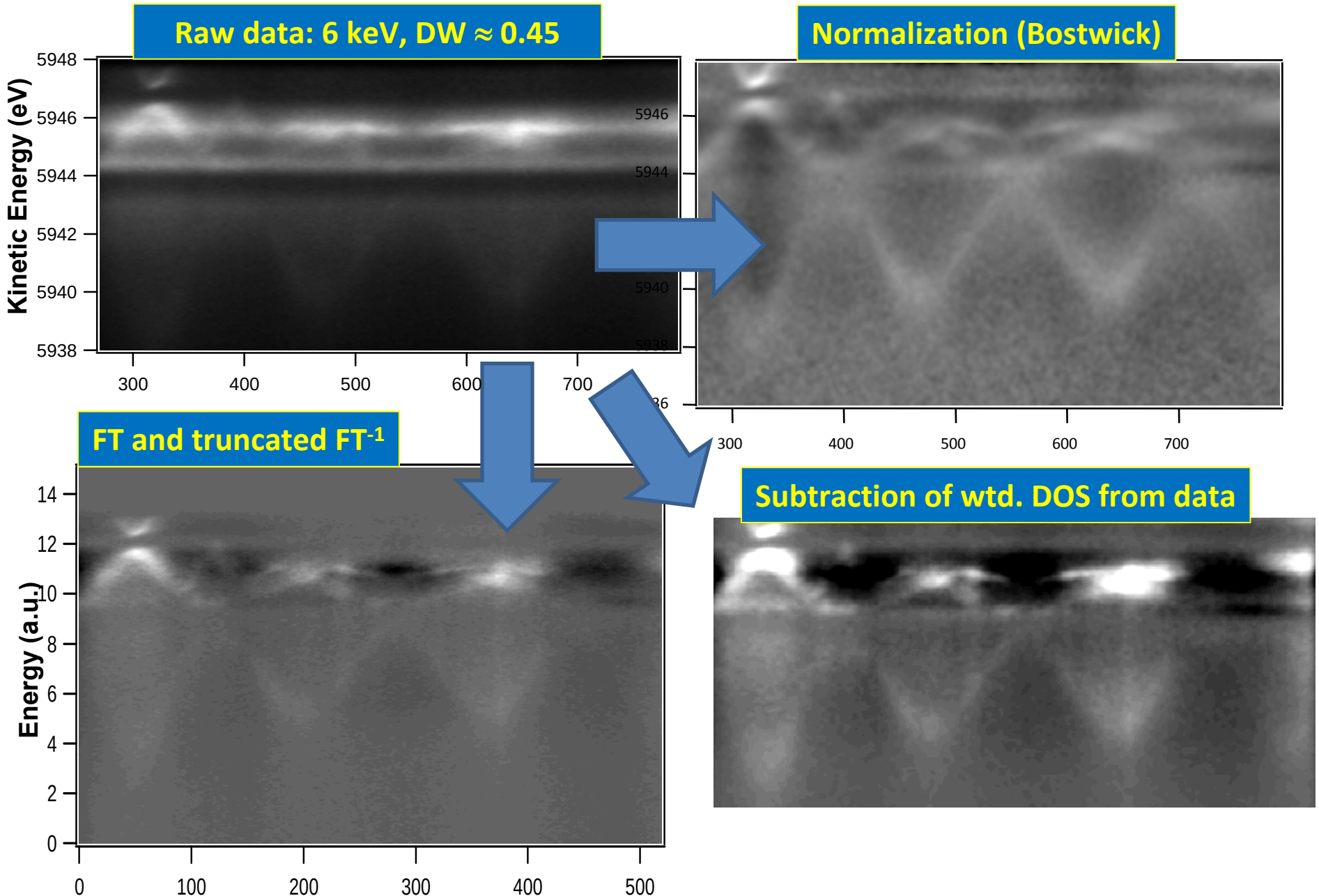


3.9 keV
30 K
DW ≈ 0.60

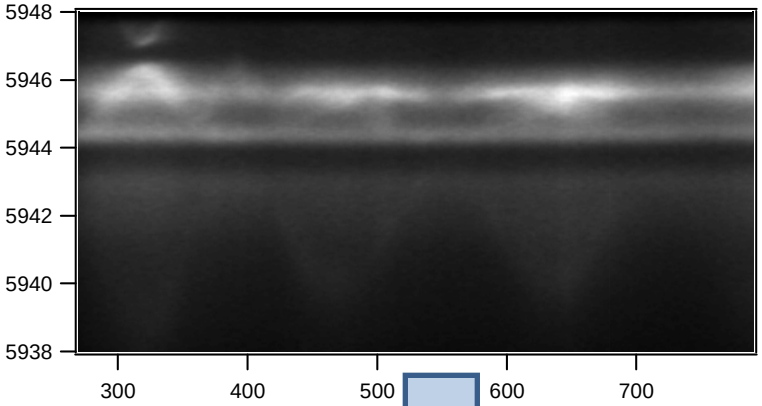


3.2 keV
300K
DW ≈ 0.28

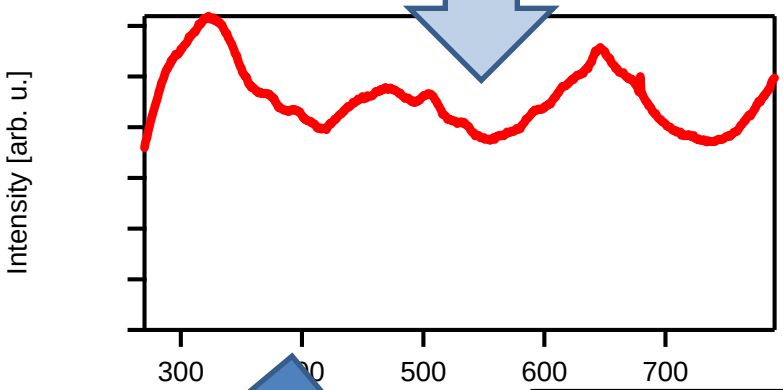
Correction for DOS and XPD: Several Methods



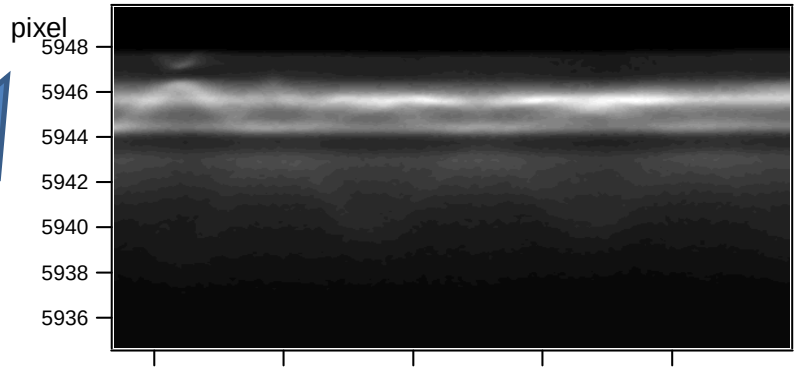
Normalization (Bostwick)



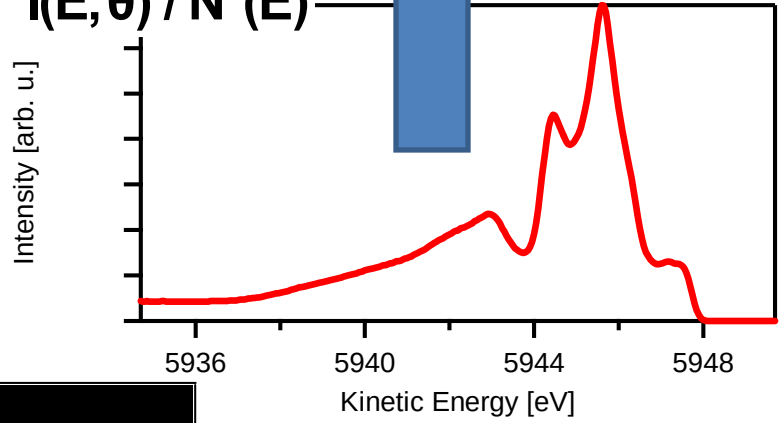
$$N(\theta) = \int I_{exp\,t}(E,\theta)dE$$



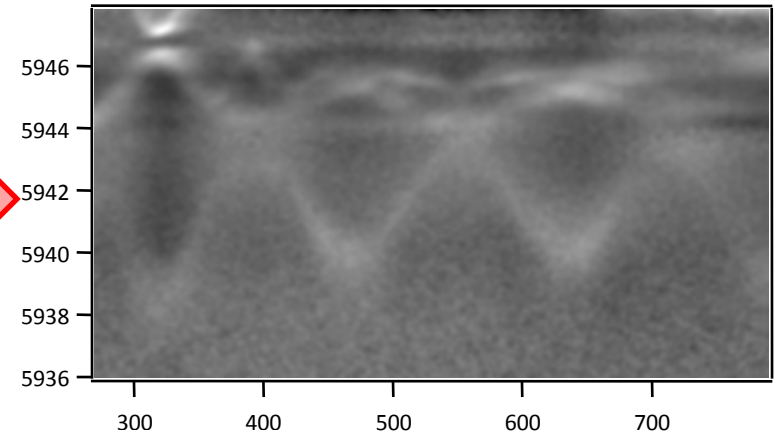
$$I(E,\theta) = I_{exp\,t}(E,\theta) / N(\theta)$$



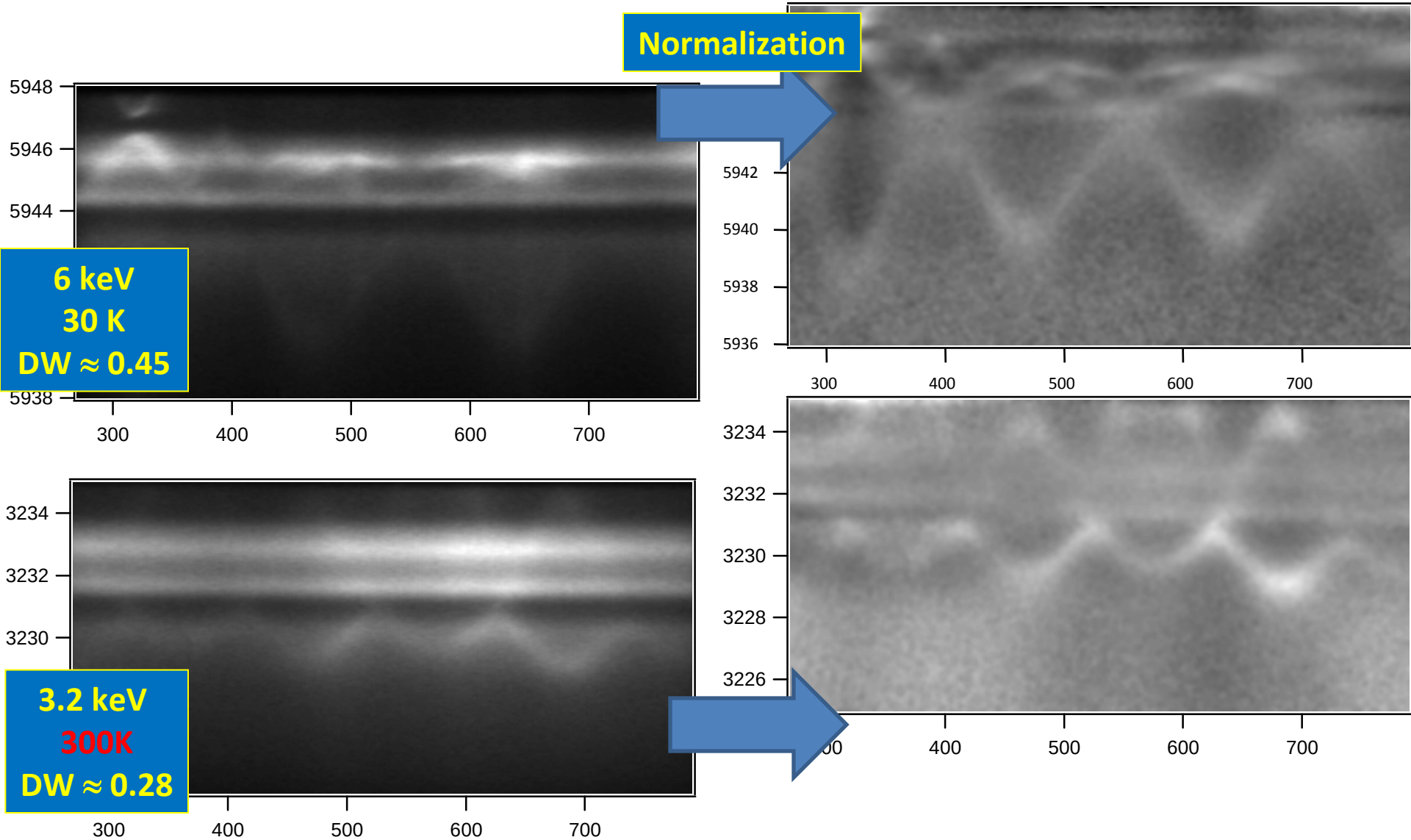
$$N'(E) = \int I(E,\theta)d\theta \approx DOS$$



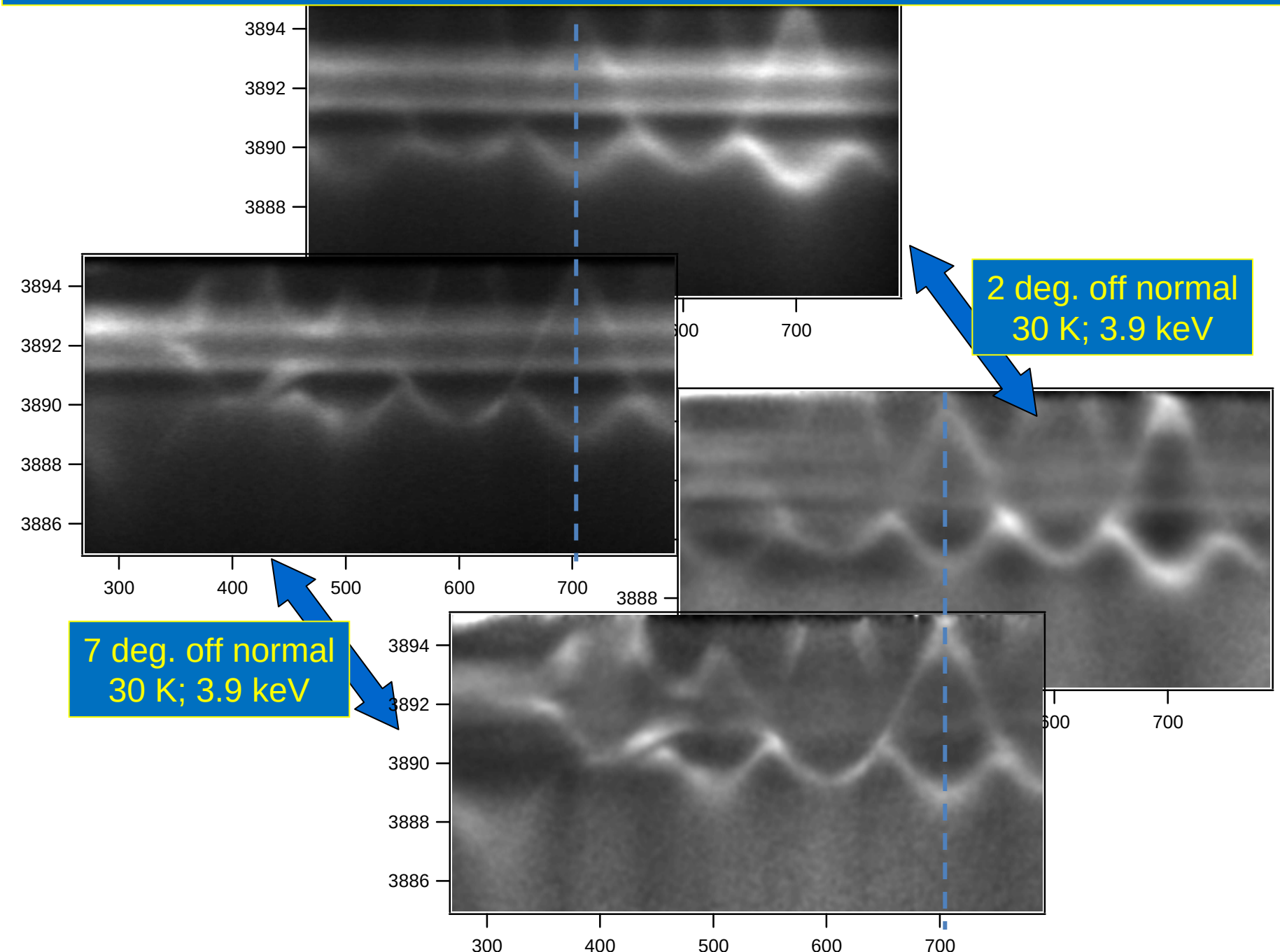
$$I'(E,\theta) = I(E,\theta) / N'(E)$$



Correction for DOS and XPD—6 keV and 3.2 keV

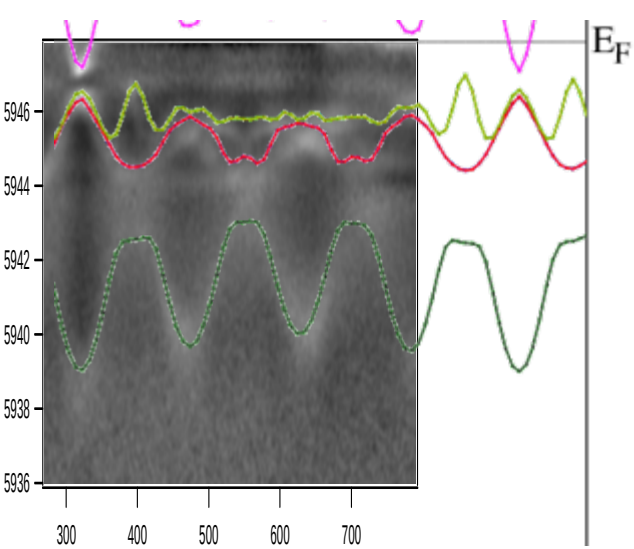


Correction preserves & enhances fine structure→tiling of detector images

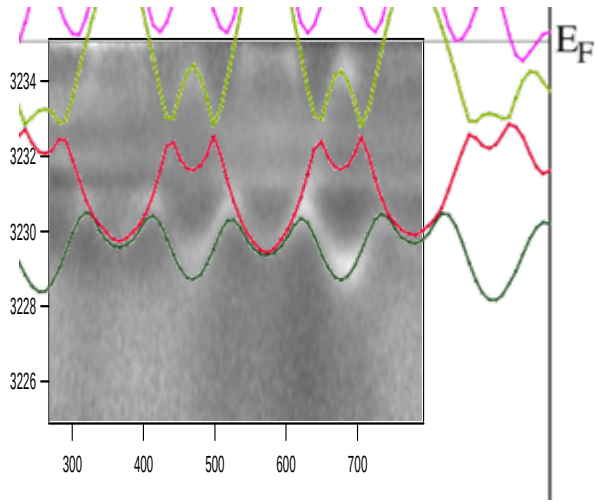


Comparison of normalized expt. to simple free-electron final-state theory (Plucinski poster)

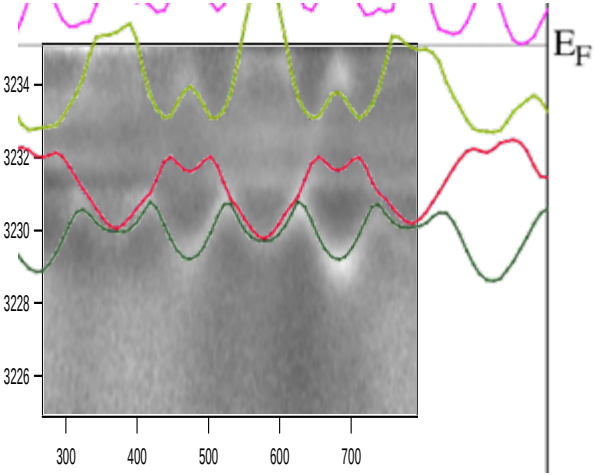
6 keV



3.9 keV

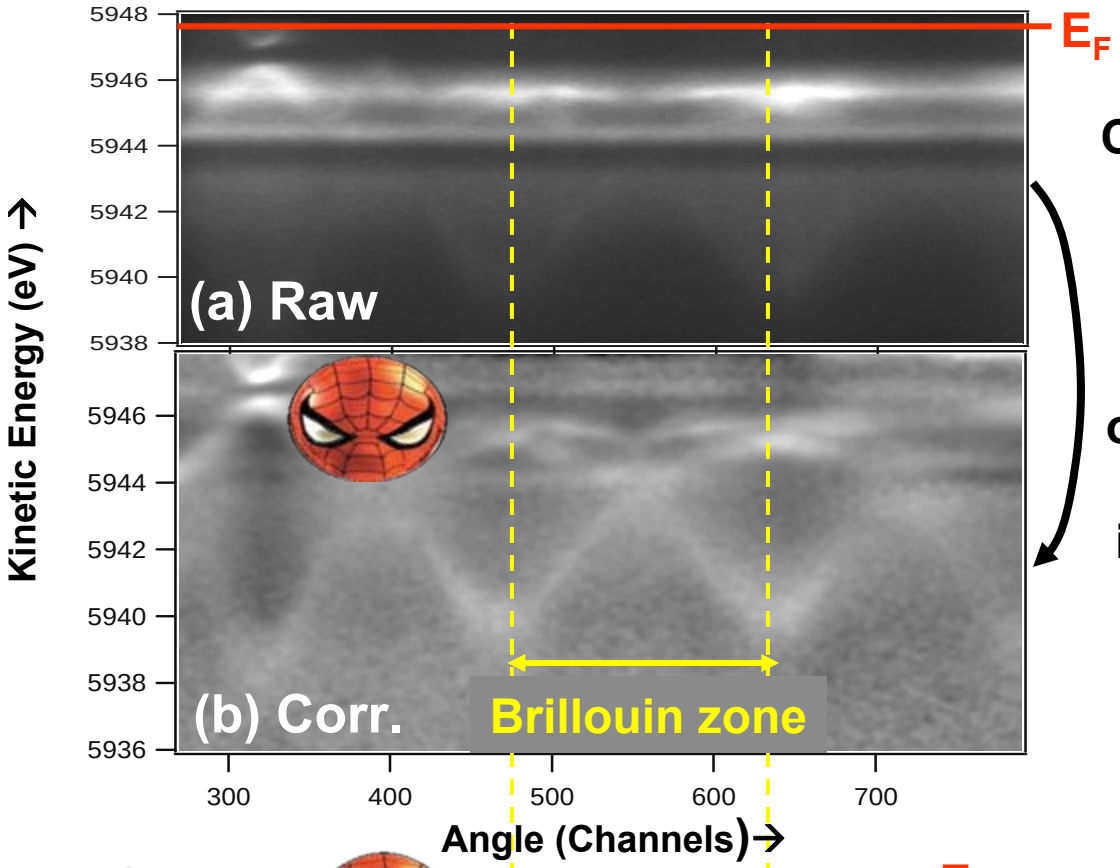


3.2 keV

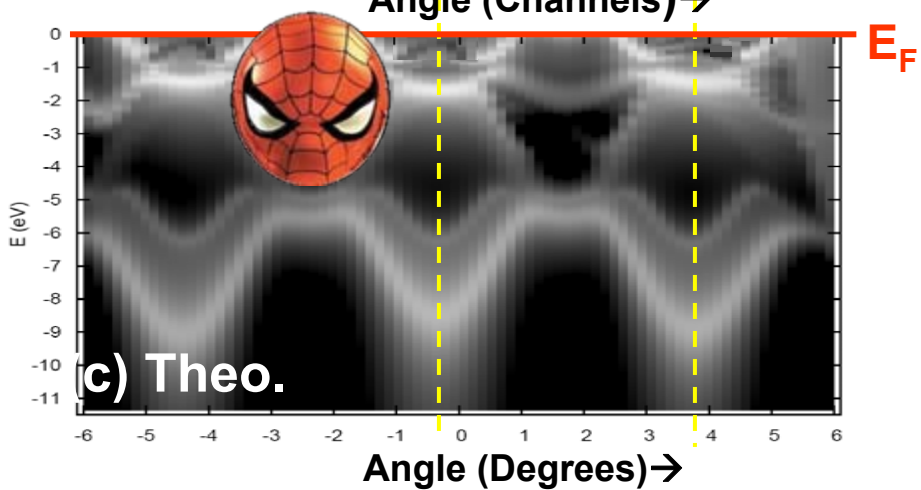


→Slight sample tilt of ca. 0.5 degrees

W(110), hν = 5,954 eV, T = 30K: Comparison to one-step theory, matrix elements

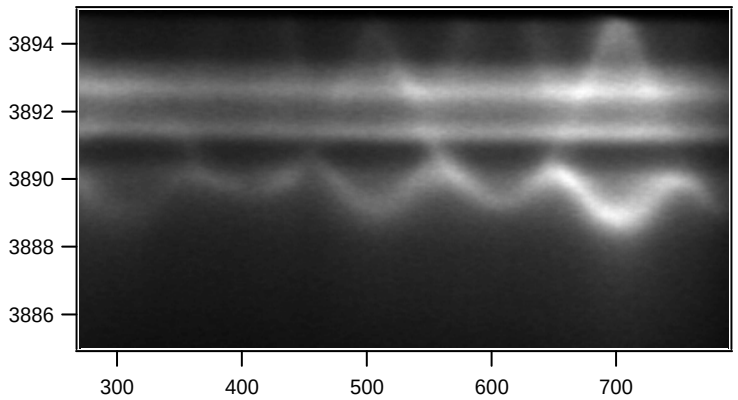


Corrected for phonon-induced density-of-states-like intensity

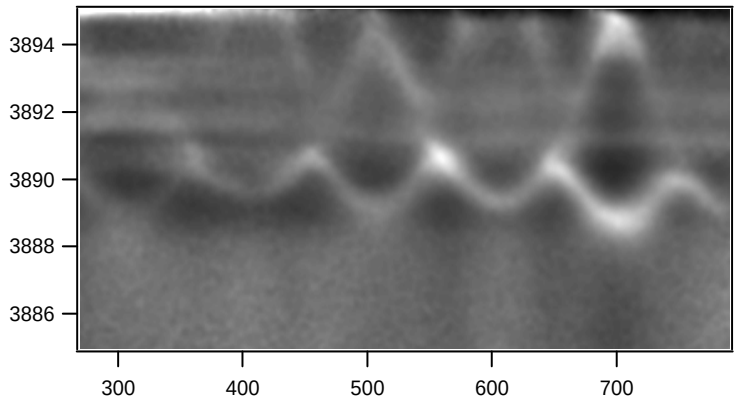


Expt.-Ueda, Kobayashi, SPring8
Data analysis-Papp, Gray, Plucinski, C.F.
Theory-Minar, Braun, Ebert

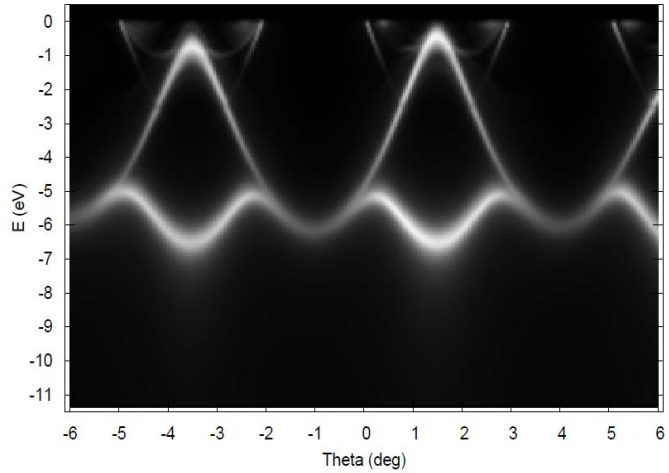
W(110), hν = 3,902 eV, T = 30K :Comparison to one-step theory, matrix elements



Raw data-3.9 keV, 30K (Ueda, Kobayashi, et al.)

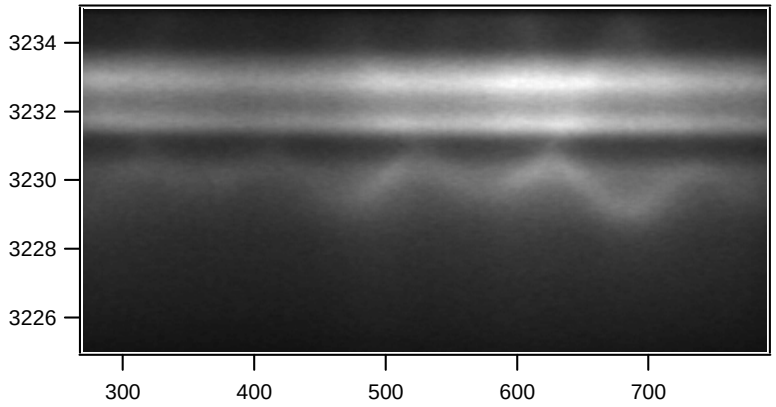


Corrected for phonon-induced density-of-states-like intensity (Papp, Gray, Fadley)

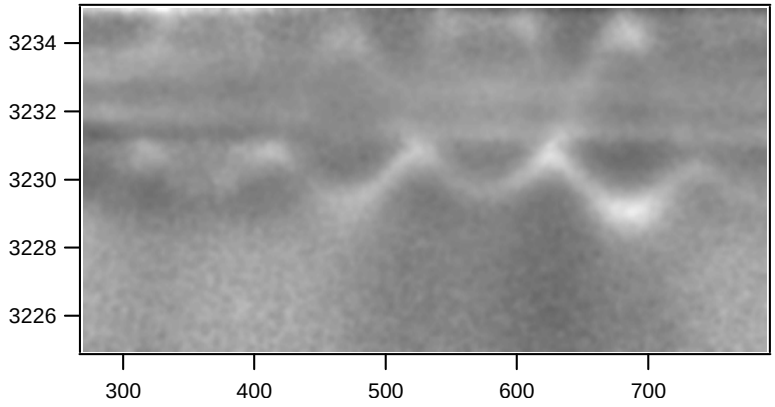


One-step theory (Minar, Braun, Ebert)

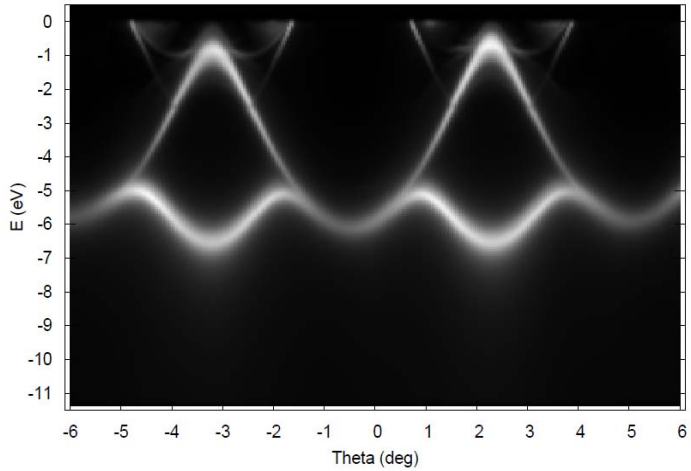
W(110), hν = 3,241 eV, T = 300K :Comparison to one-step theory, matrix elements



Raw data-3.2 keV, 300K (Ueda, Kobayashi, et al.)



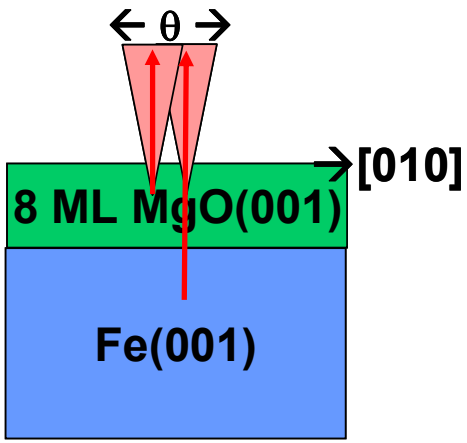
Corrected for phonon-induced density-of-states-like intensity (Papp, Gray, Fadley)



One-step theory (Minar, Braun, Ebert)

FUTURE APPLICATION TO MULTILAYER SPINTRONIC SYSTEMS

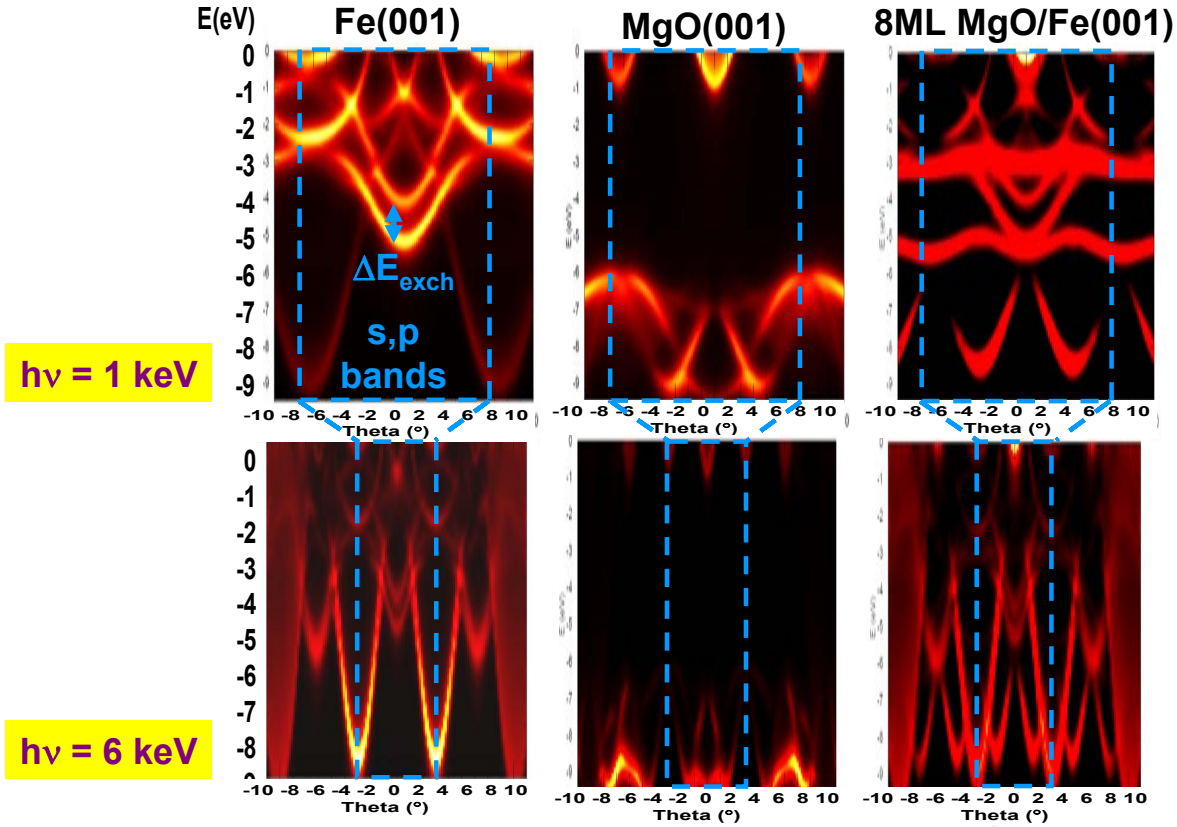
Angle-resolved photoemission through overlayer insulators: A new probe of buried layer/interface electronic structure? Fe/MgO



- Cryocooling to suppress phonon smearing
- Standing-wave excitation to vary depth sensitivity

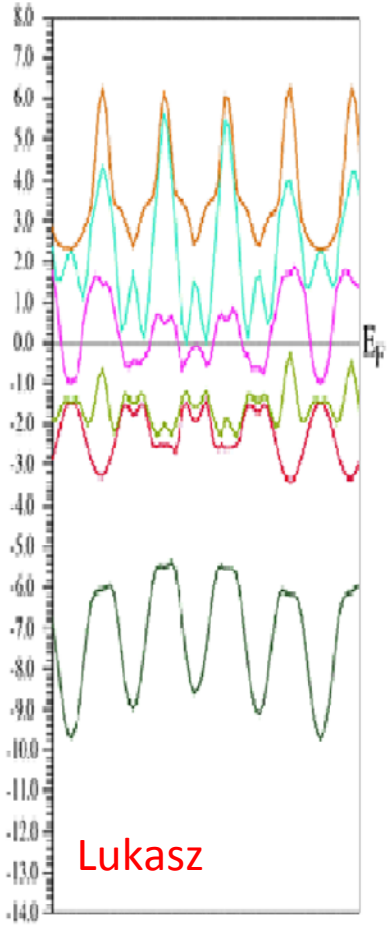
(See talks by Ebert & Papp, Poster by Plucinski)

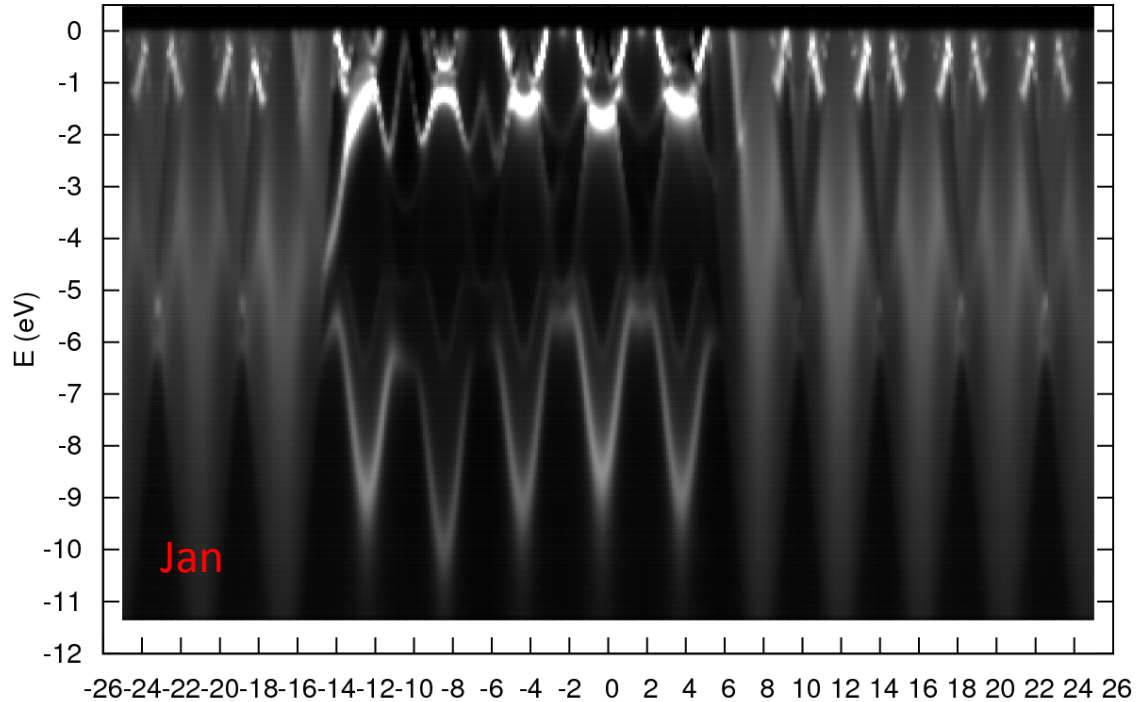
One-step photoemission theory: Ebert, Braun, Minar, C.F.

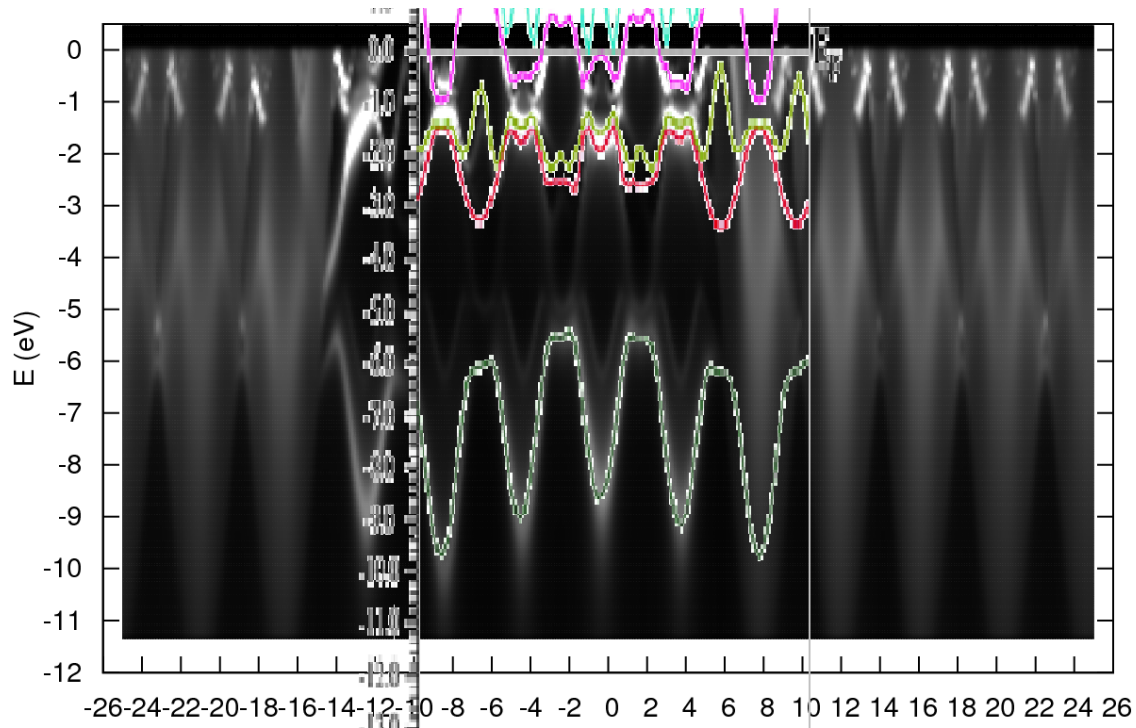


Conclusions

- Soft and Hard X-ray ARPES of W (110) demonstrated over the energy range 1.2-6.0 keV
  - Correction of data for phonon effects via several methods
  - Free-electron final-state calculations yield quick insight, e.g. on sample alignment
  - One-step photoemission theory with matrix elements agrees very well with expt.
- Better theoretical treatment of phonons needed (Ebert talk)
- Debye-Waller calculations for other systems suggest more bulk-sensitive electronic structure studies, esp. in 1-3 keV range (Plucinski poster)
- Future applications to depth-resolved electronic structure in multilayers?

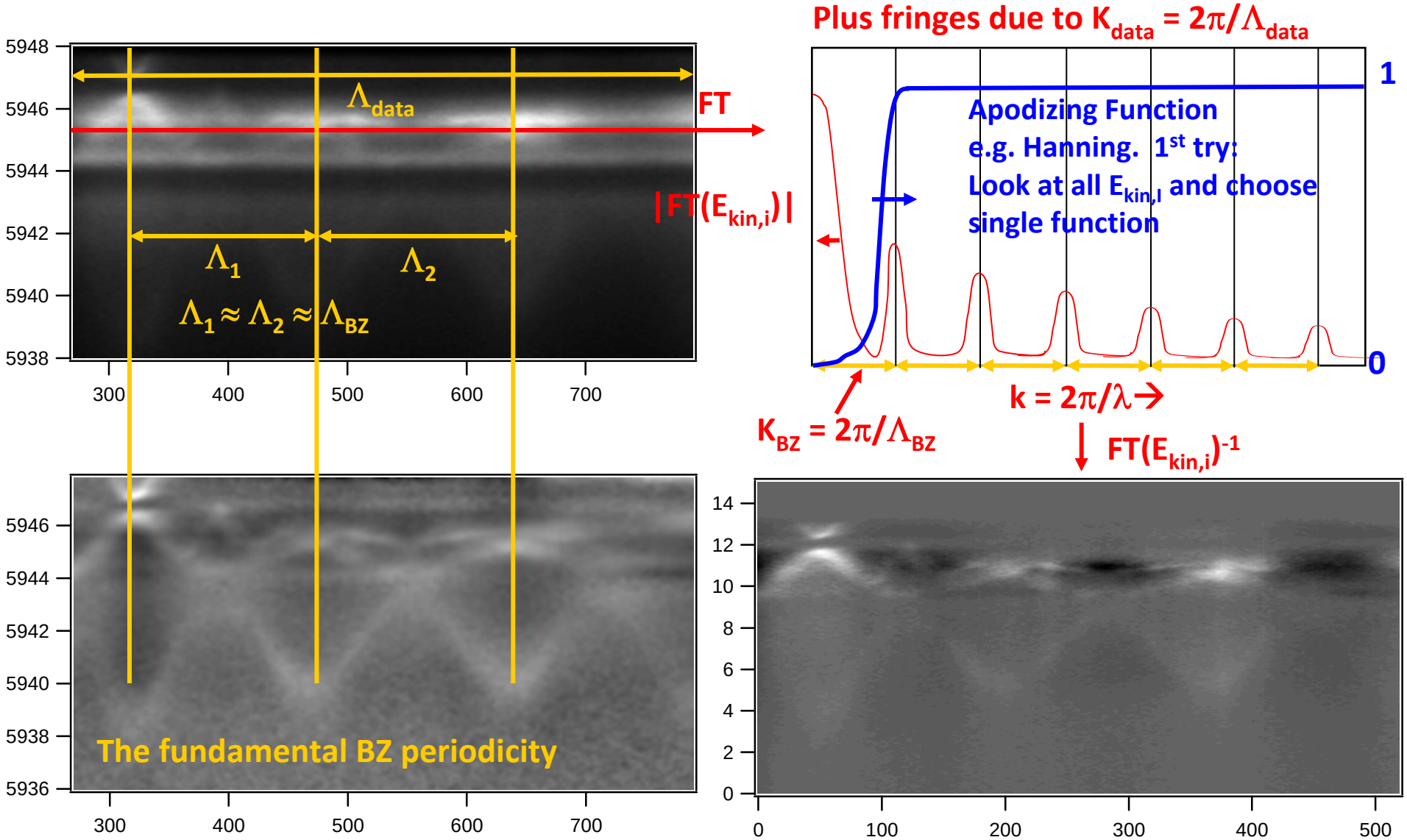




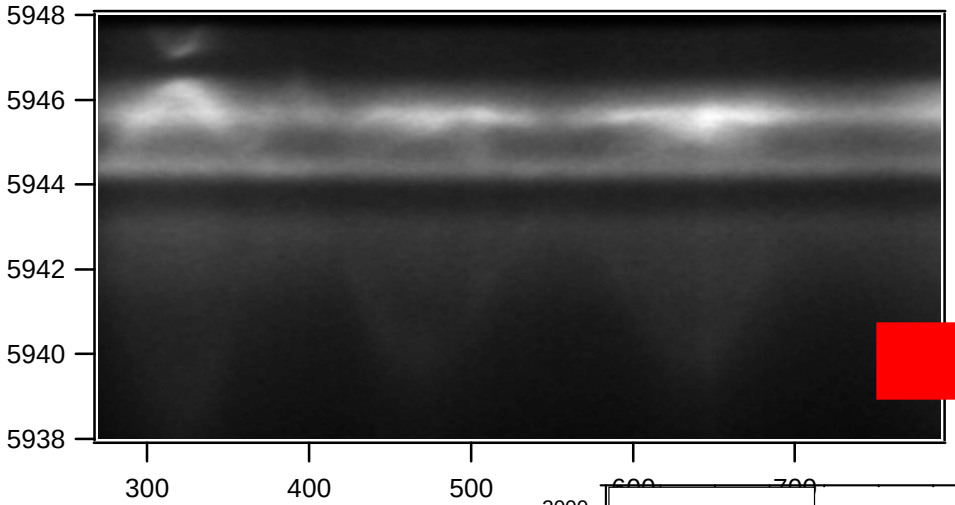


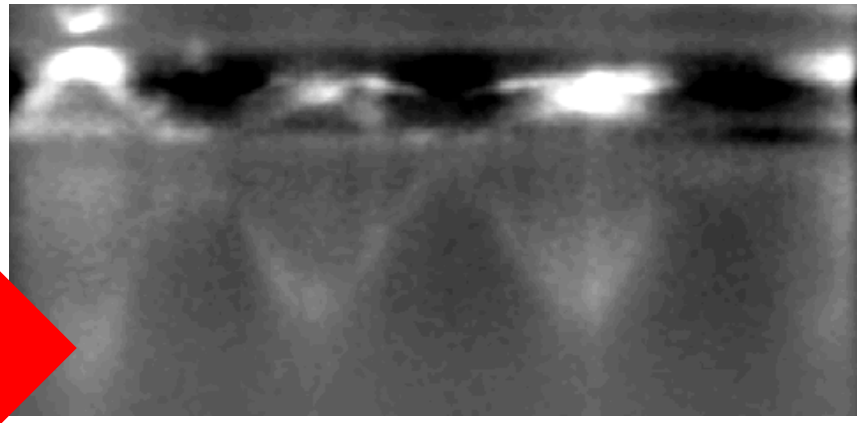
Comparison of Lukascz's to Jan's data at 6000eV.

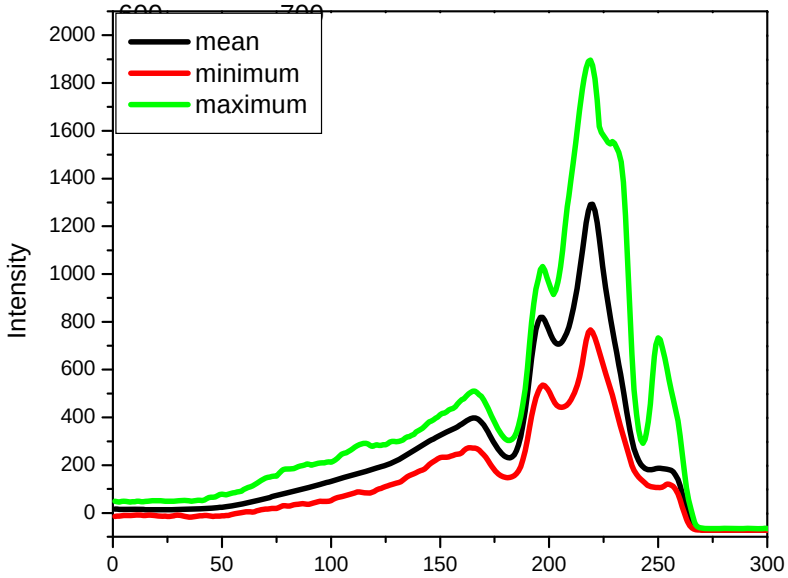
Schematic of FT correction of HXARPES data:



Subtraction







Mean turns out to be the best

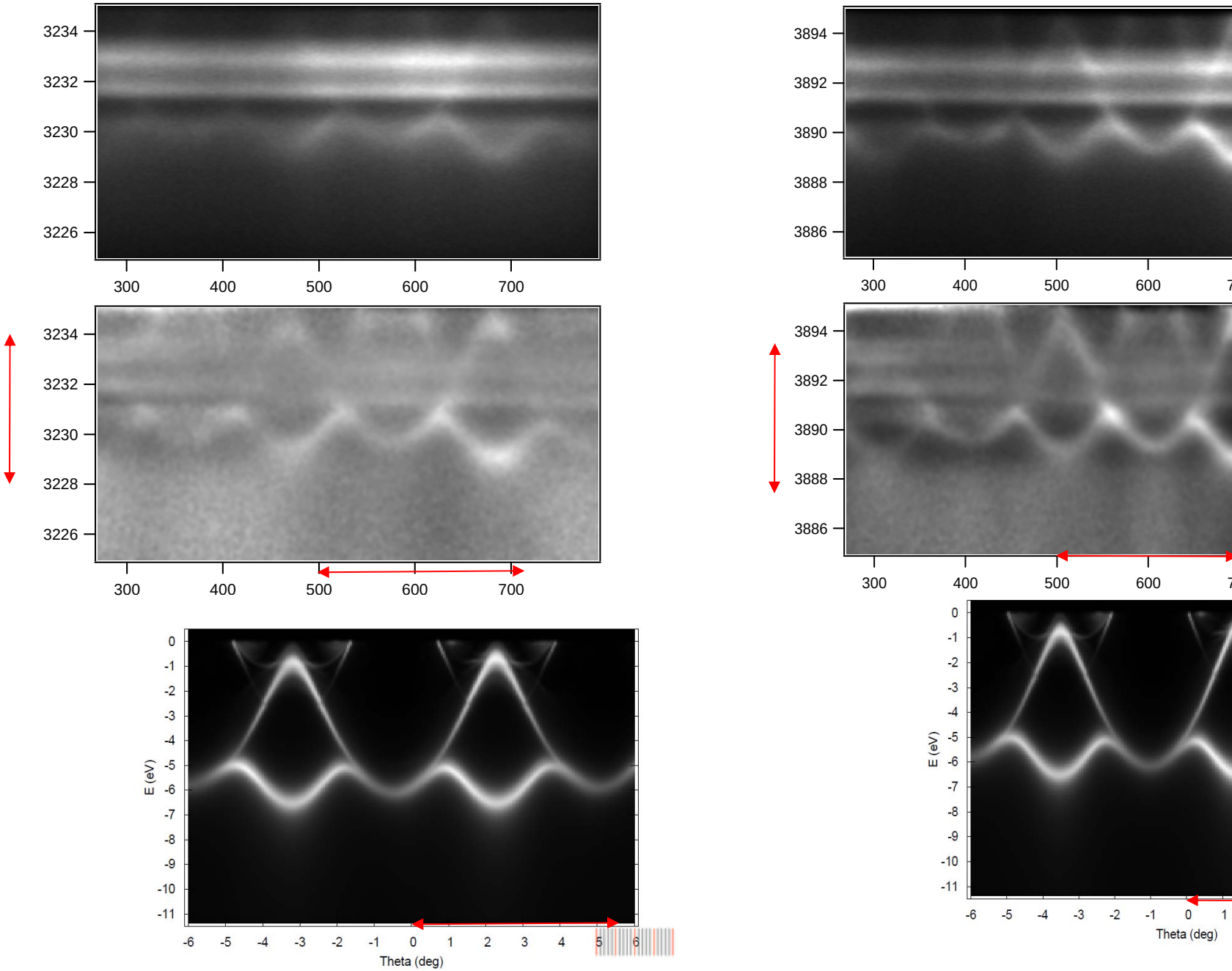
Alex Gray, CP, CSF

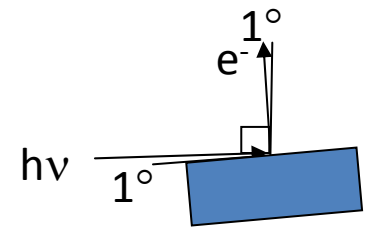
ata-3.2 keV,
emperature

n by
gle-
s

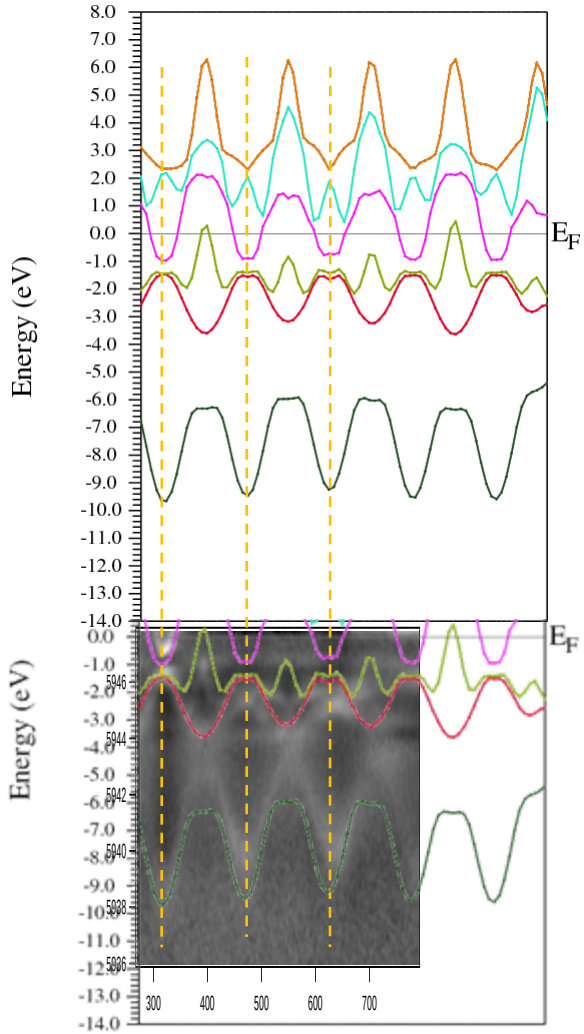
.

ory
.)

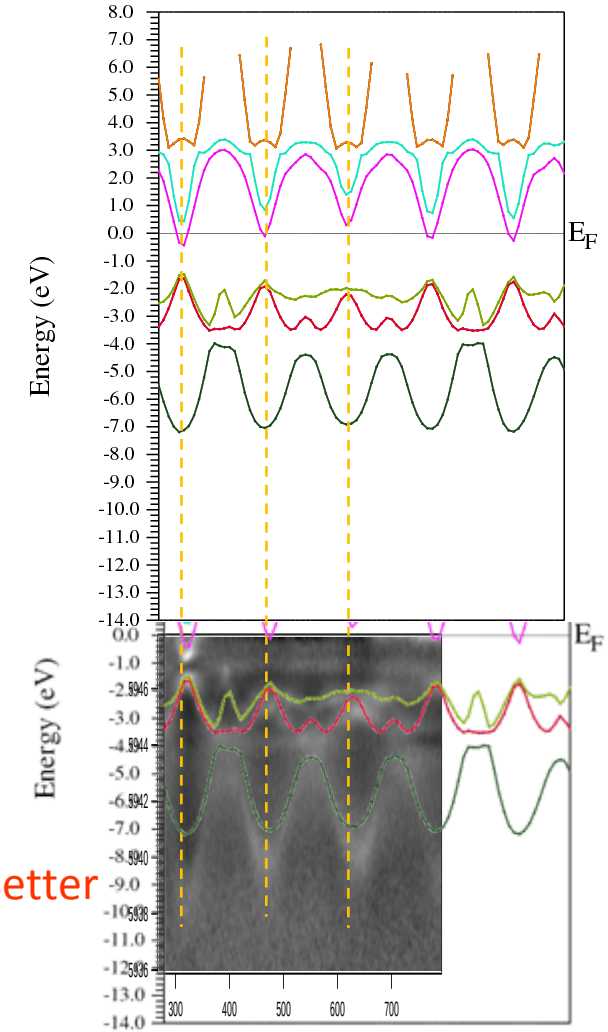




5953eV tilt 0 deg



5953eV tilt 1 deg



Better