ARPES: How to make it quantitative?

P. Aebi, Institut de Physique, Université de Neuchâtel, Switzerland

If it doesn't work for Cu, forget it!

Does it work for Cu? for HTc s? for CDW s?

(not so much new results / science, but "naive" questions)

(what are the different problems)

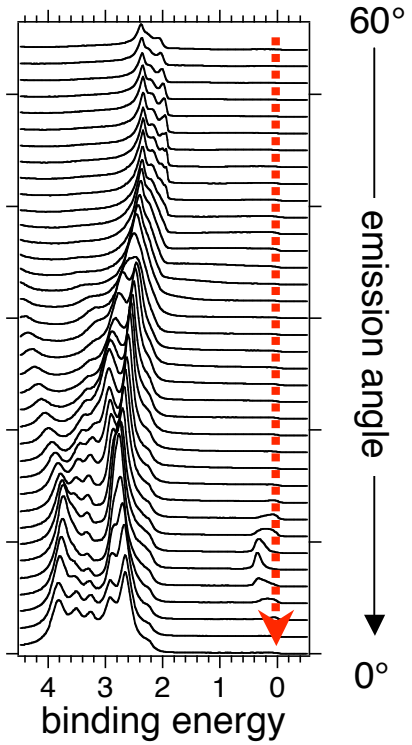
Modular approach:
separately understand

- physics of initial state ('everybody is interested')
- physics of final state ('nobody is interested')
- physics of coupling (matrix elements) ('nobody is interested')

F. Clerc, M. Bovet, C. Battaglia, L. Despont, H. Cercellier, M.G. Garnier

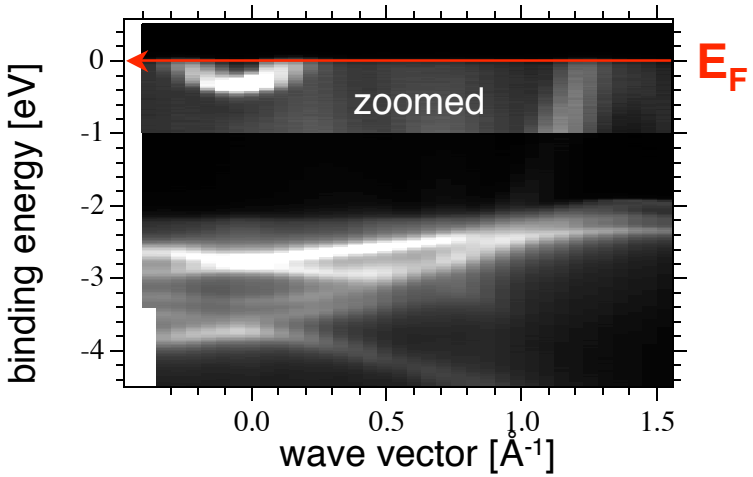
What we would like to do…

Measure spectra



E(k)

Bandmapping



Interpret line shape as

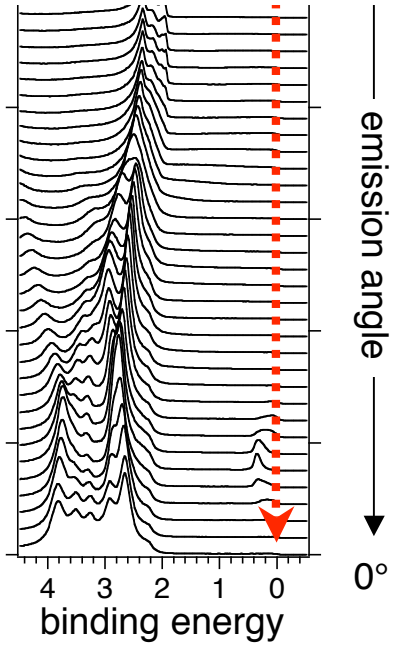
$$A(\vec{k},\omega) = \frac{1}{\pi}\,\frac{\Im m\Sigma(\vec{k},\omega)}{|\omega - \epsilon_{\vec{k}} - \Re e\Sigma(\vec{k},\omega)|^2 + |\Im m\Sigma(\vec{k},\omega)|^2}$$

Peak width: inverse life time

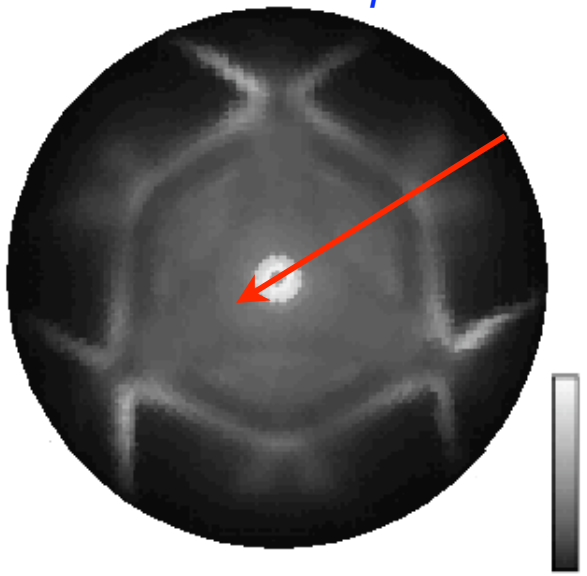
Band dispersion

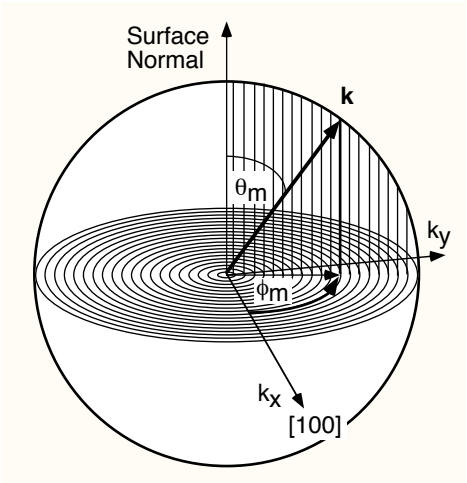
$\Sigma(\vec{k},\omega)$ **Self Energy**

What we would like to do…

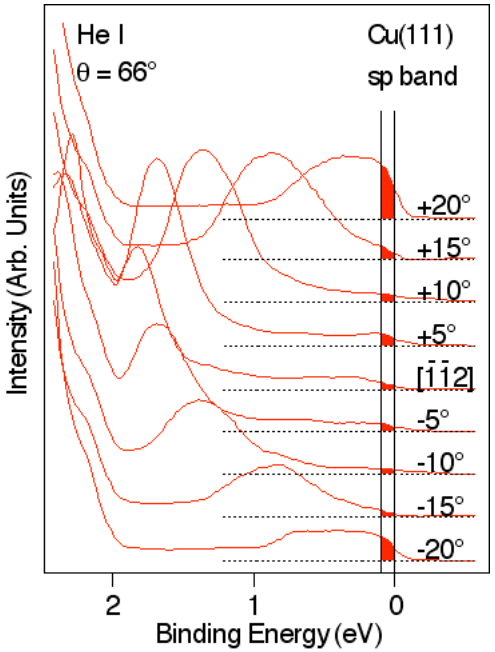


Measure maps…

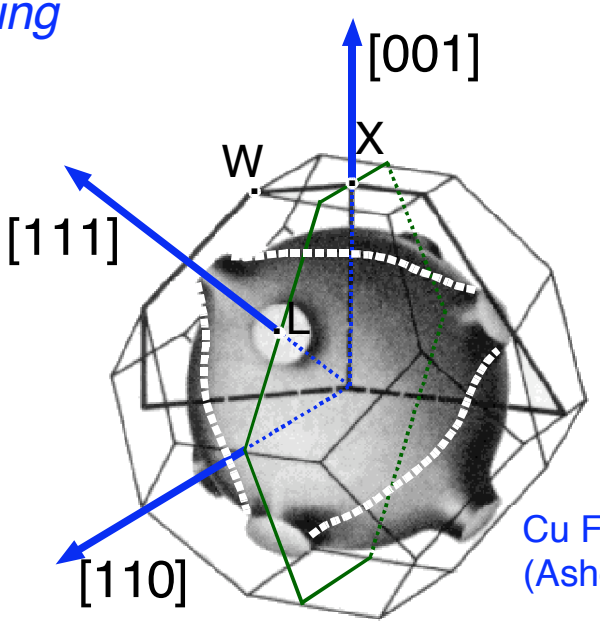




E_F mapping



and interpret as …
… Fermi surface



$k(E_F)$

Cu Fermi surface
(Ashcroft/Mermin)

We know: It is not as simple as that !

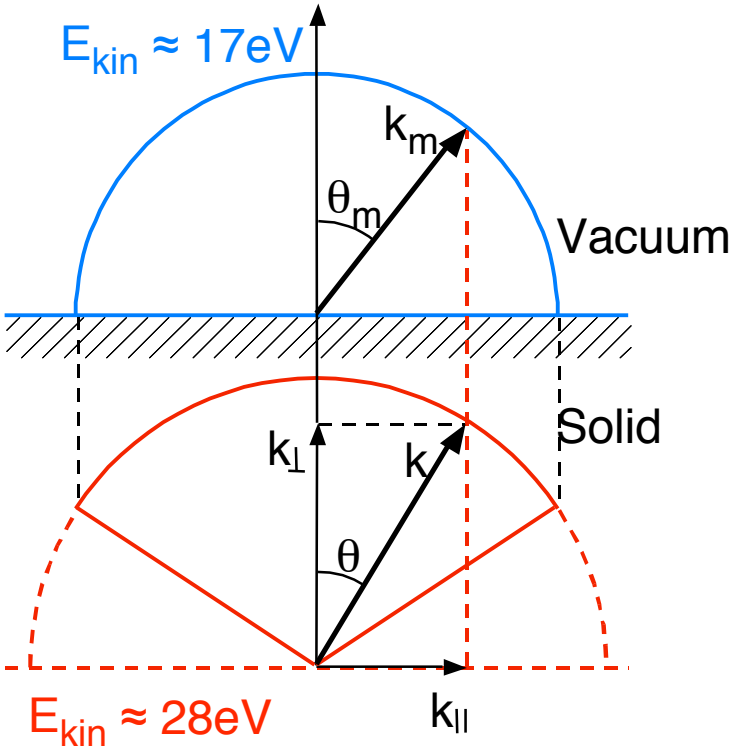
Presently, what can we do?

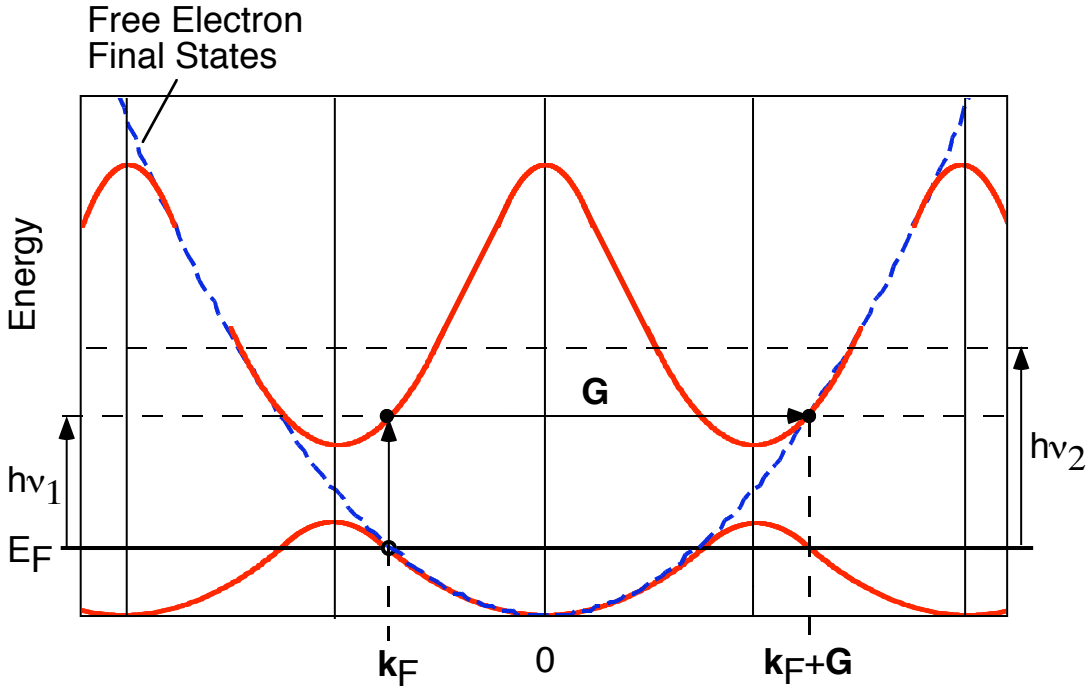
(as experimentalists)

Does it work for Cu?

… almost …

Direct transitions / Free electron final state





$$E^m_{kin} = h\nu - \Phi - E_B$$

$$|\vec{k}_{||}| = \frac{1}{\hbar}\sqrt{2mE^m_{kin}}sin\theta_m$$

$$|\vec{k}_{\perp}| = \frac{1}{\hbar}\sqrt{2m(E^m_{kin}+V_0)}\,cos\theta_m$$

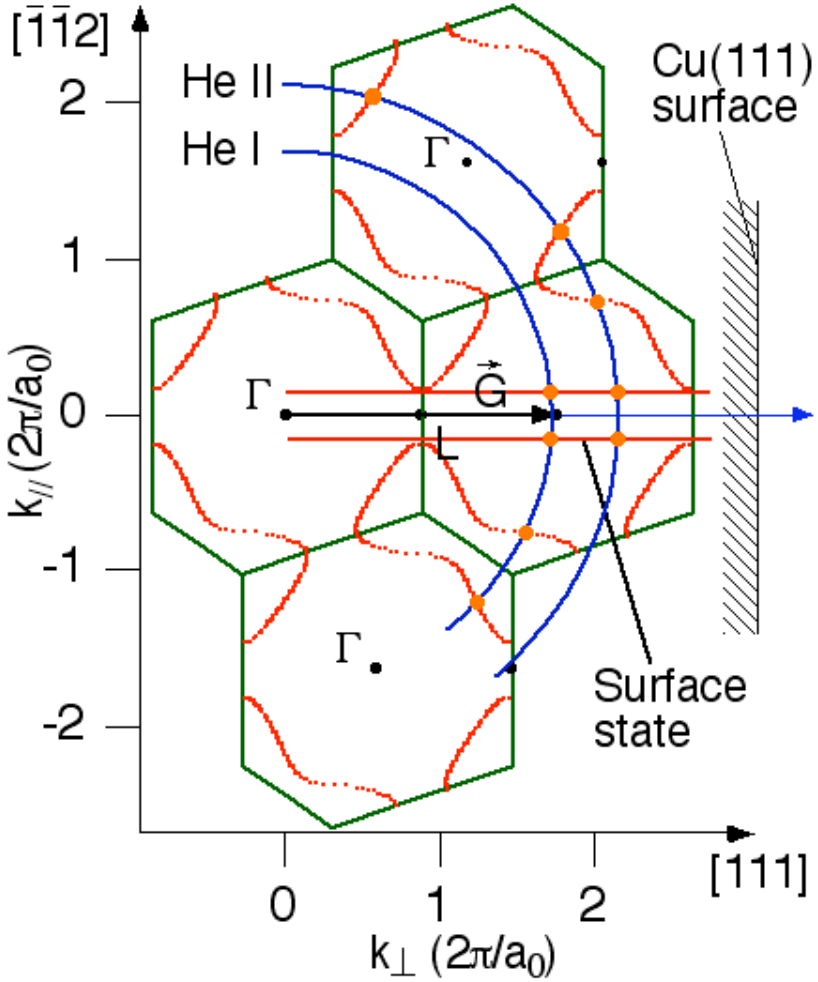
$$sin\theta = sin\theta_m\sqrt{\frac{E^m_{kin}}{(E^m_{kin}+V_0)}}$$

Refraction

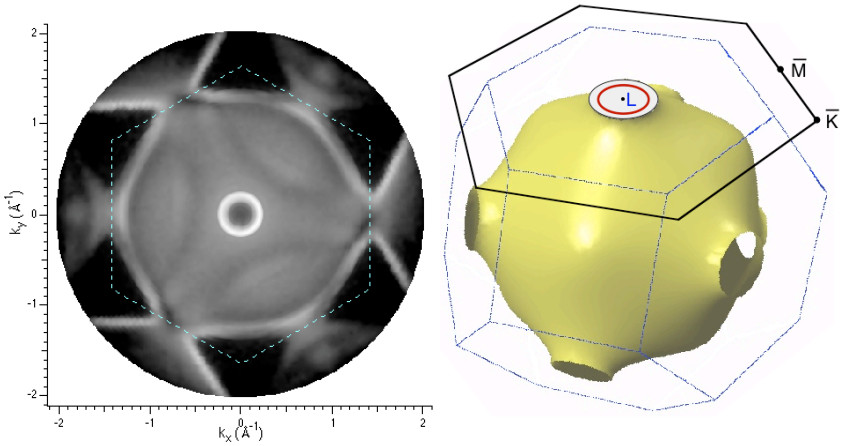
Given: hν, Φ, E_B, θ_m; adjust V_0 --> *k* is completely determined

Fermi surface mapping

Section along ($\bar{1}10$) plane in reciprocal space



Courtesy J. Osterwalder



In the photoemission process:

Energy conservation:

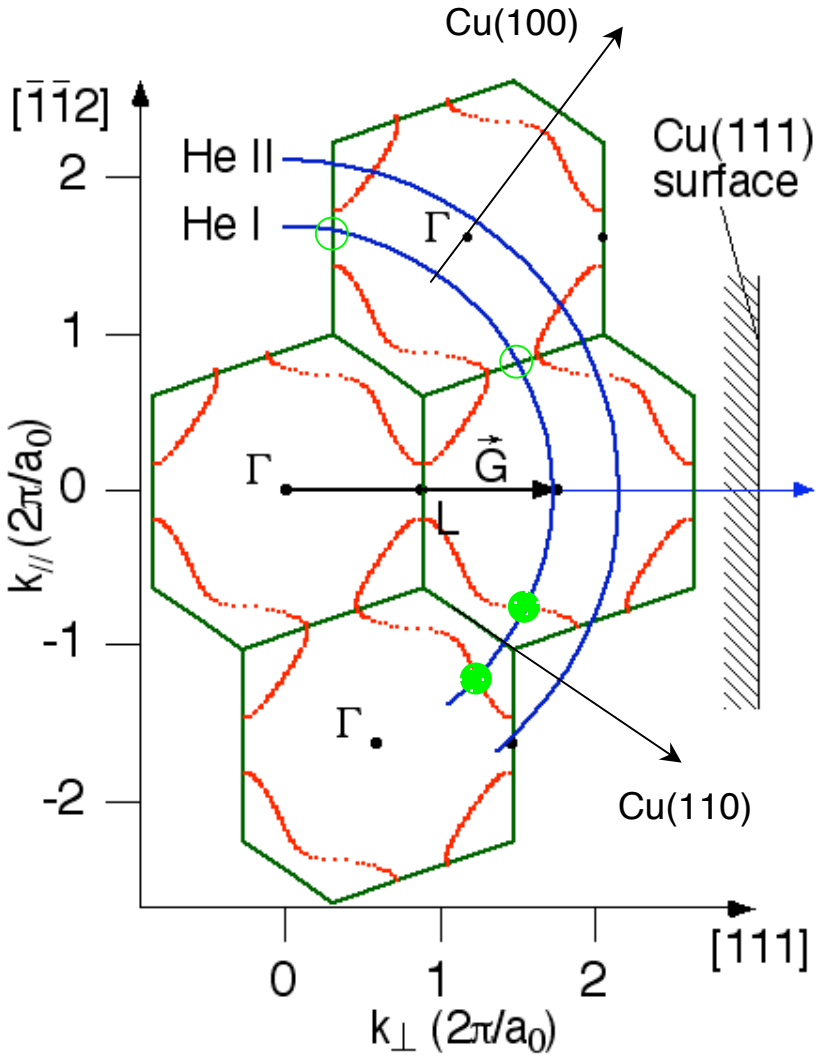
$$E_f = E_i + h\nu$$

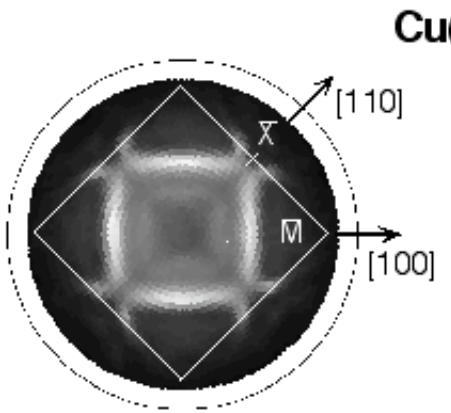
Momentum conservation:

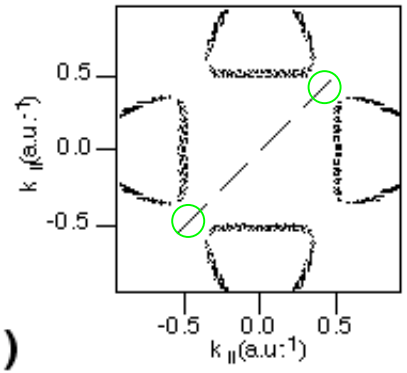
$$\vec{k}_f = \vec{k}_i + \vec{G}$$

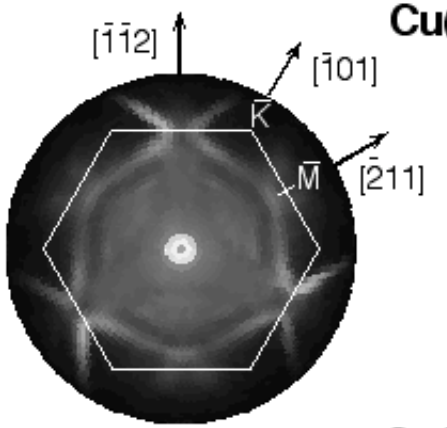
Surf. Sci. 307-309, 917 (1994)

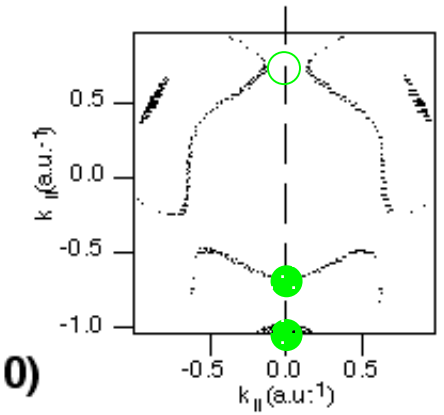
Fermi surface mapping: Cu surfaces He I

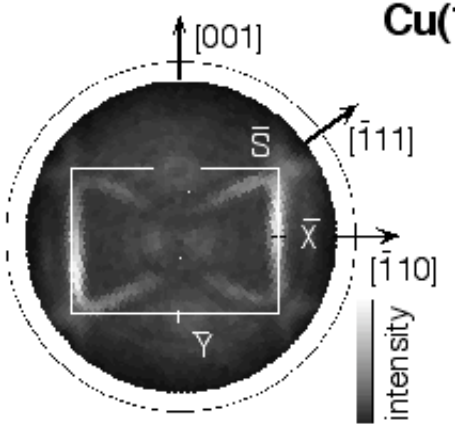


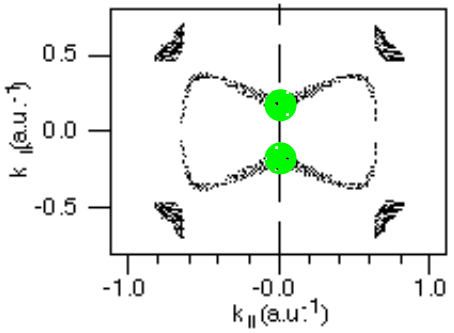






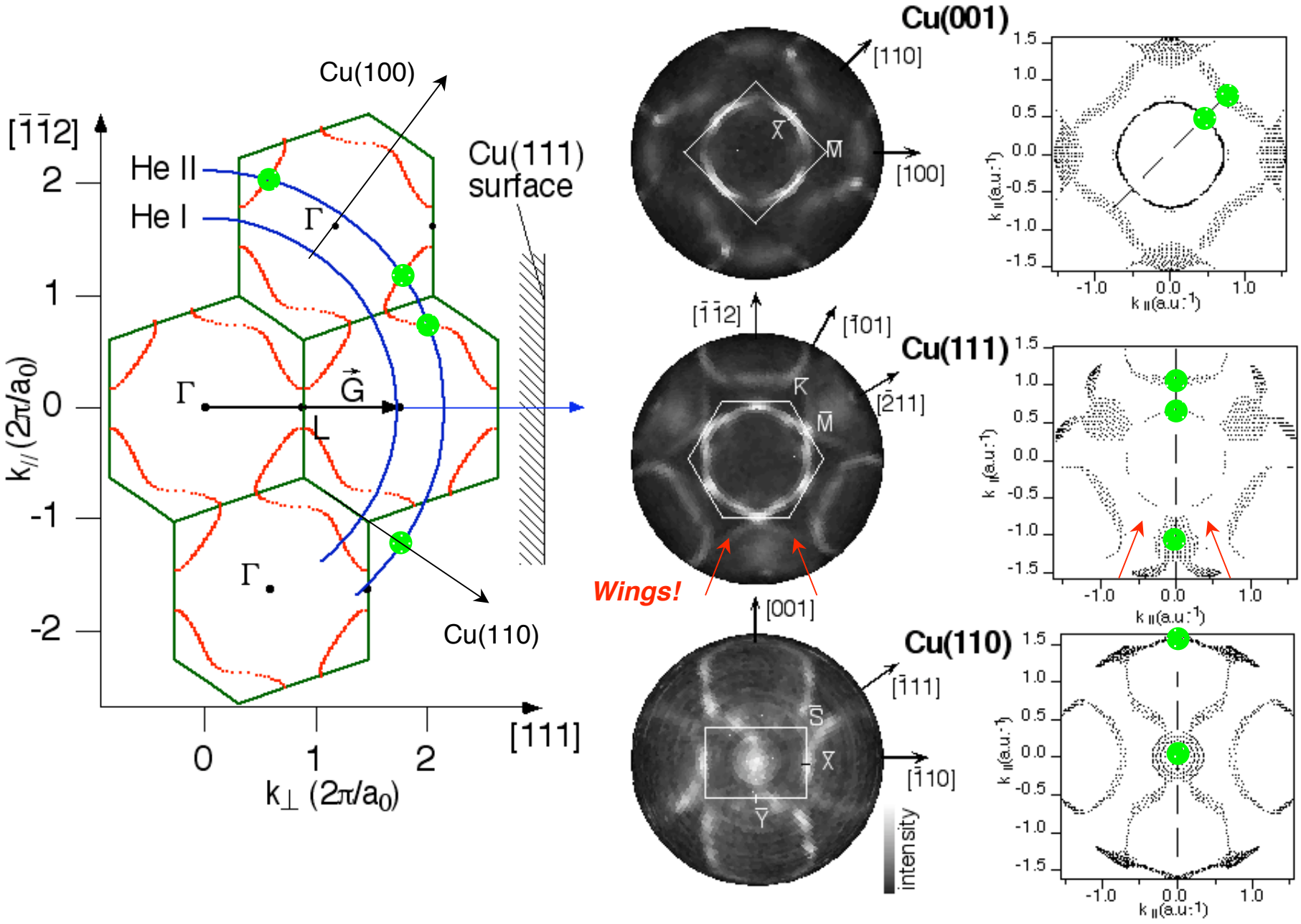




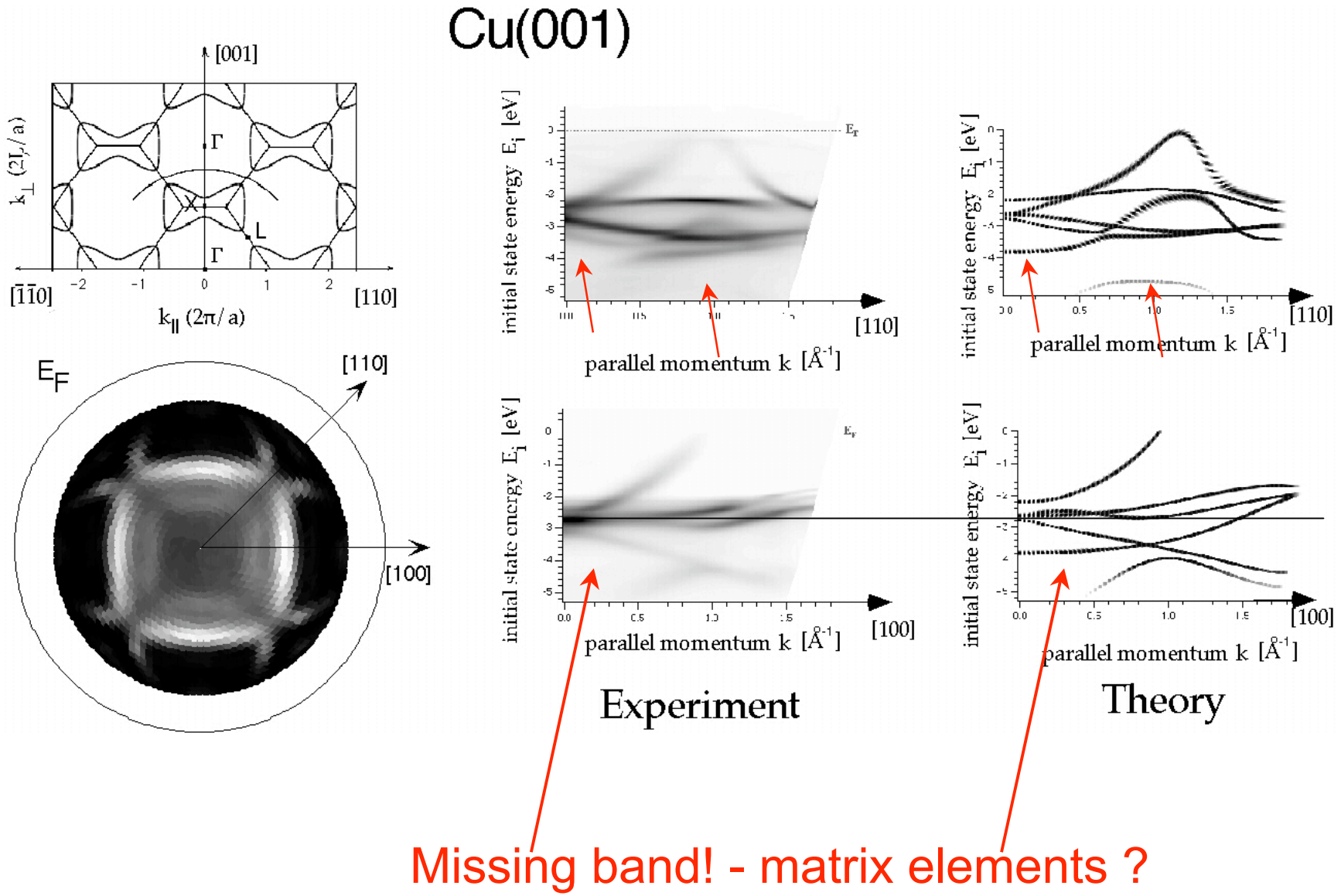


Surf. Sci. 307-309, 917 (1994)

Fermi surface mapping: Cu surfaces He II



Band mapping: Cu(001) He I



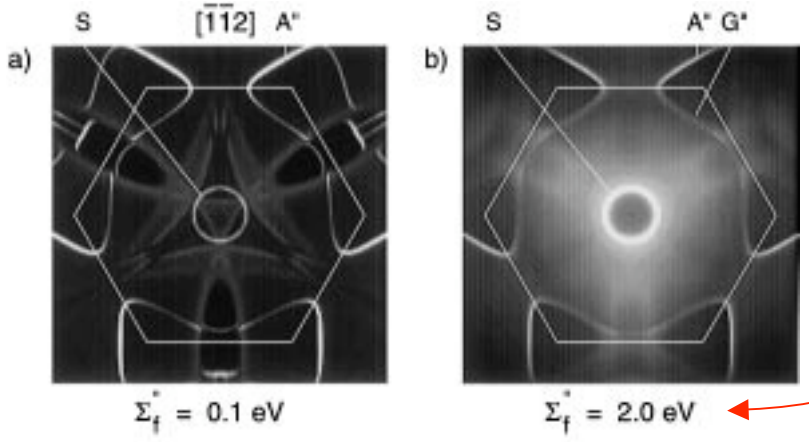
One-step model may account for this …

VOLUME 77, NUMBER 14 PHYSICAL REVIEW LETTERS 30 SEPTEMBER 1996

A Novel Direct Method of Fermi Surface Determination Using Constant Initial Energy Angle-Scanned Photoemission Spectroscopy

M. Lindroos[1,2] and A. Bansil[1]
[1]*Physics Department, Northeastern University, Boston, Massachusetts 02115*
[2]*Tampere University of Technology, P.O. Box 692, SF-33101, Tampere, Finland*
(Received 26 April 1996)

We show that a constant initial energy, angle-scanned (CIE-AS) photoemission spectrum for emission from the Fermi energy (E_F) contains Fermi surface (FS) signatures which originate from density of states type *indirect* transitions. Such previously unrecognized FS features in a CIE-AS spectrum would provide a robust and straightforward means of determining Fermi surfaces. Furthermore, the associated photointensity should yield a new window on $k_\perp$ dispersion related issues in materials. Extensive simulations of CIE-AS spectra from low index faces of Cu are presented within the framework of the one-step photoemission model in order to delineate the nature of these new spectral features. [S0031-9007(96)01334-8]



hν= 21.2 eV, unpolarized, would be nice to have it for hν= 64 eV

Final state broadening

For Cu the situation is not too bad …

BUT:
Free electron final state does NOT explain:
- DOS features, - indirect transitions,
- Missing band

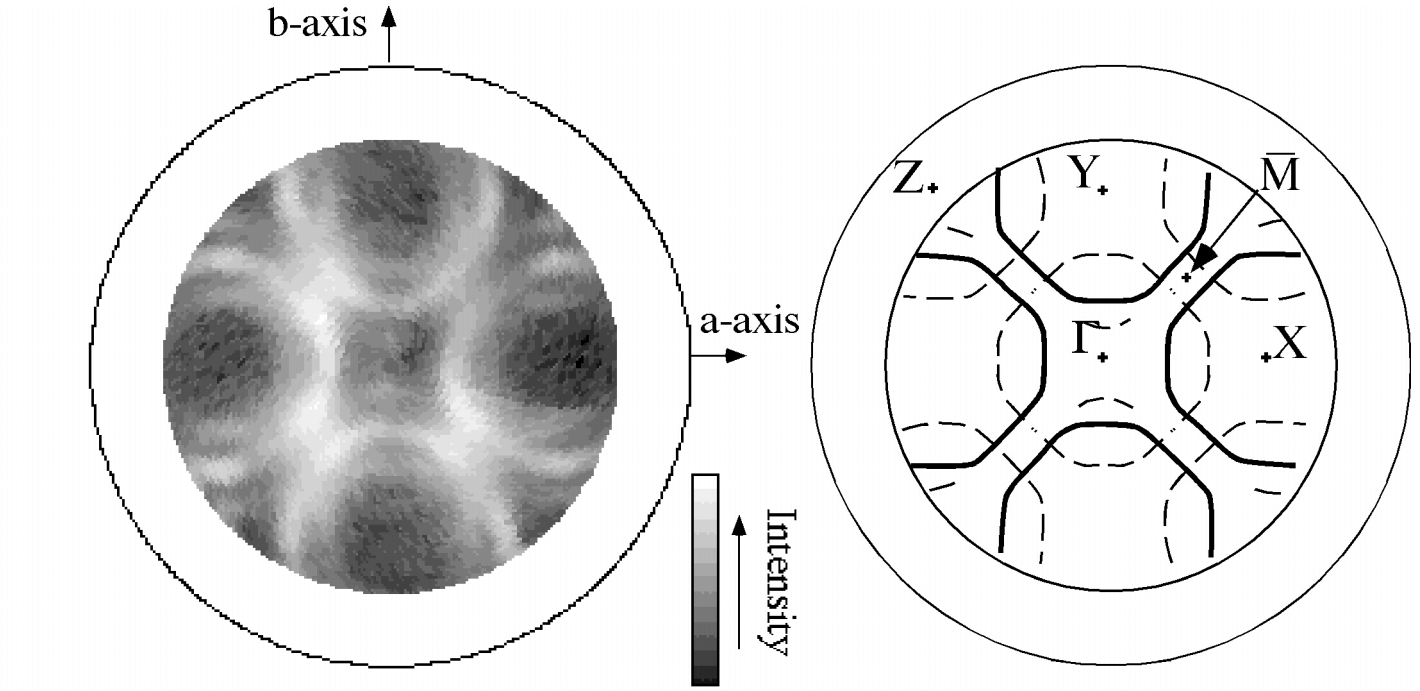
We need to consider:
-final state scattering
-lifetime broadening
-matrix elements

Is it because Cu is 3D?

What about 2D?

Quasi 2D system, a well-known example …

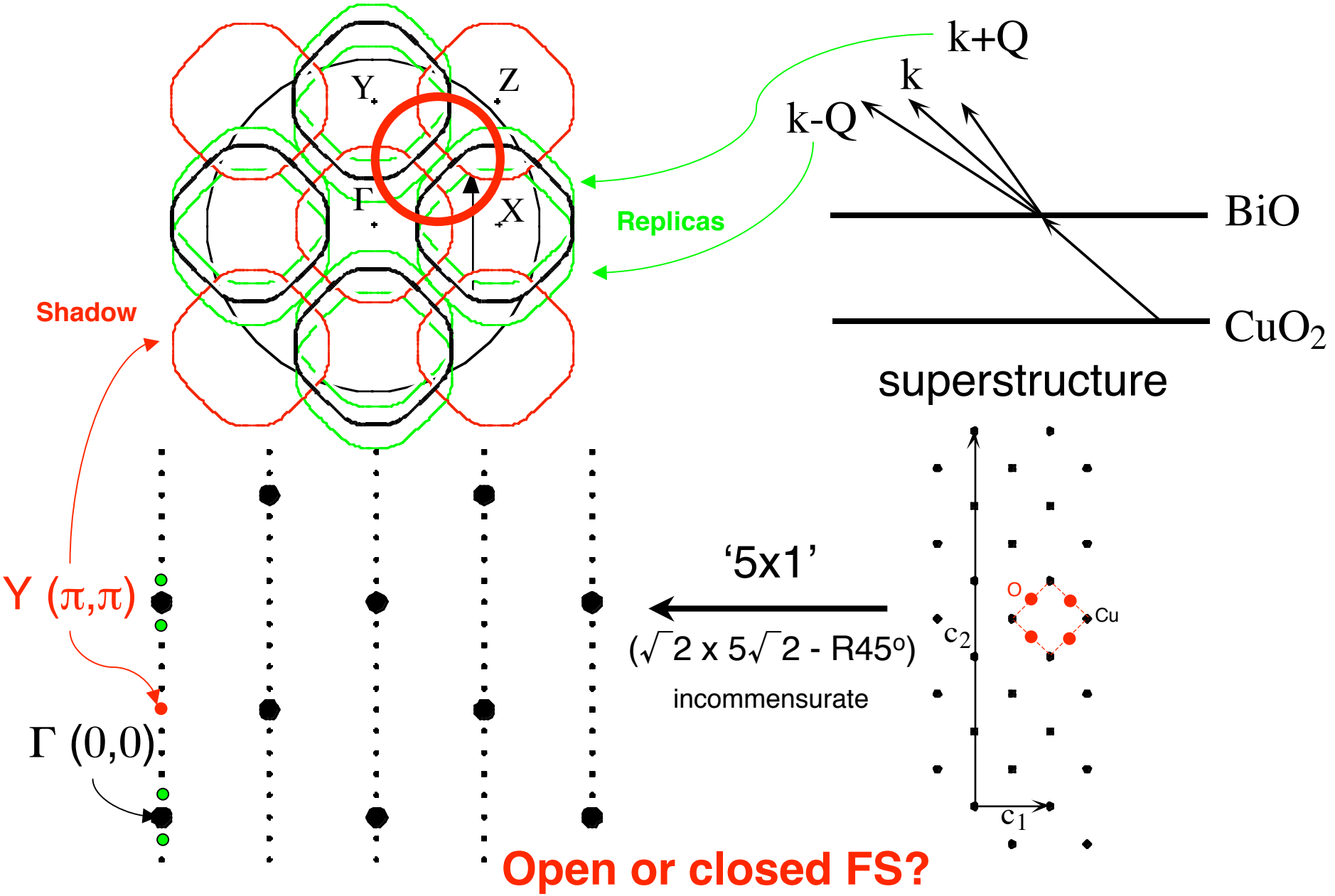
Bi2212: The joys of Fermi surface mapping …



… may turn into pitfalls ….

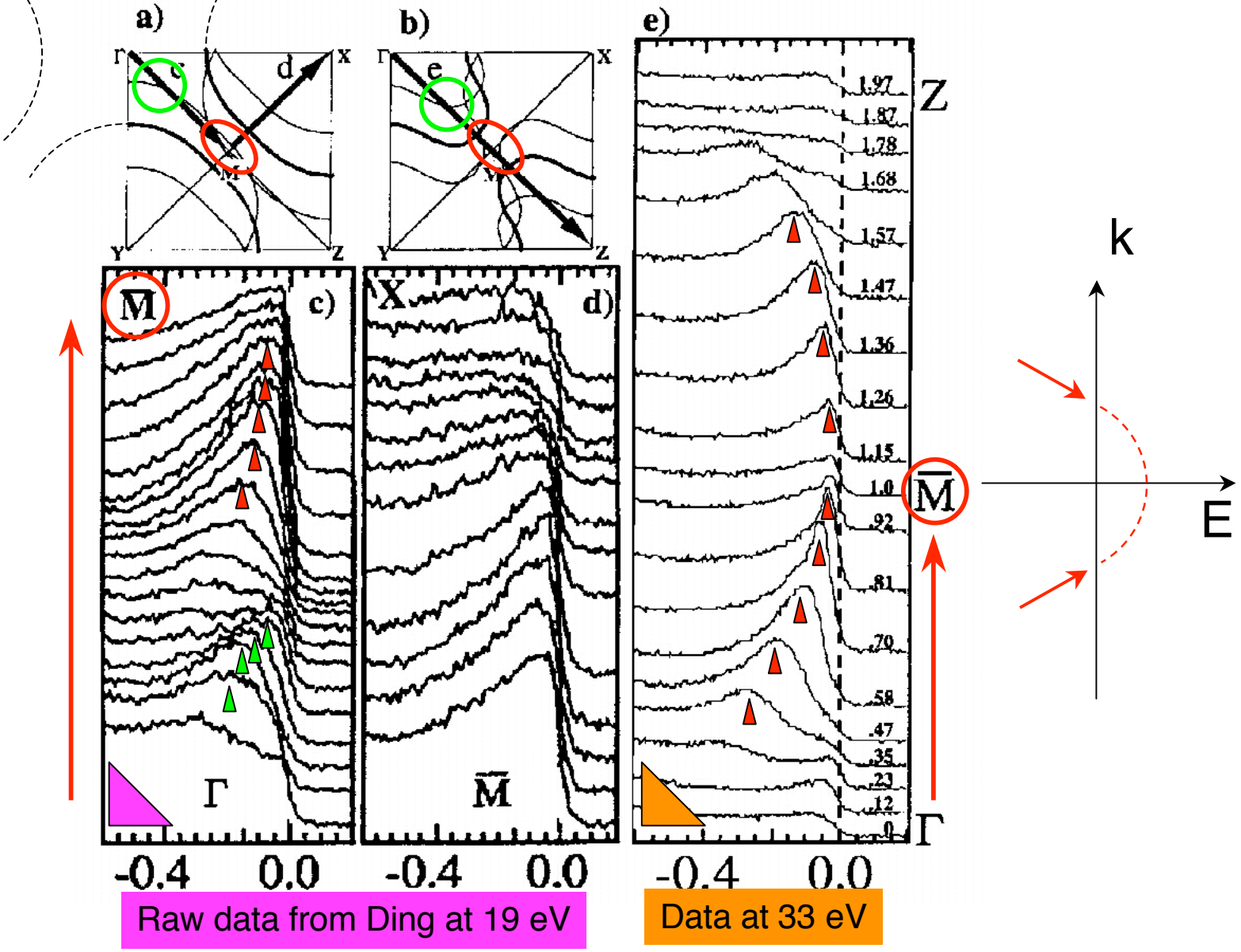
Phys. Rev. Lett. 72, 2757-2760 (1994)

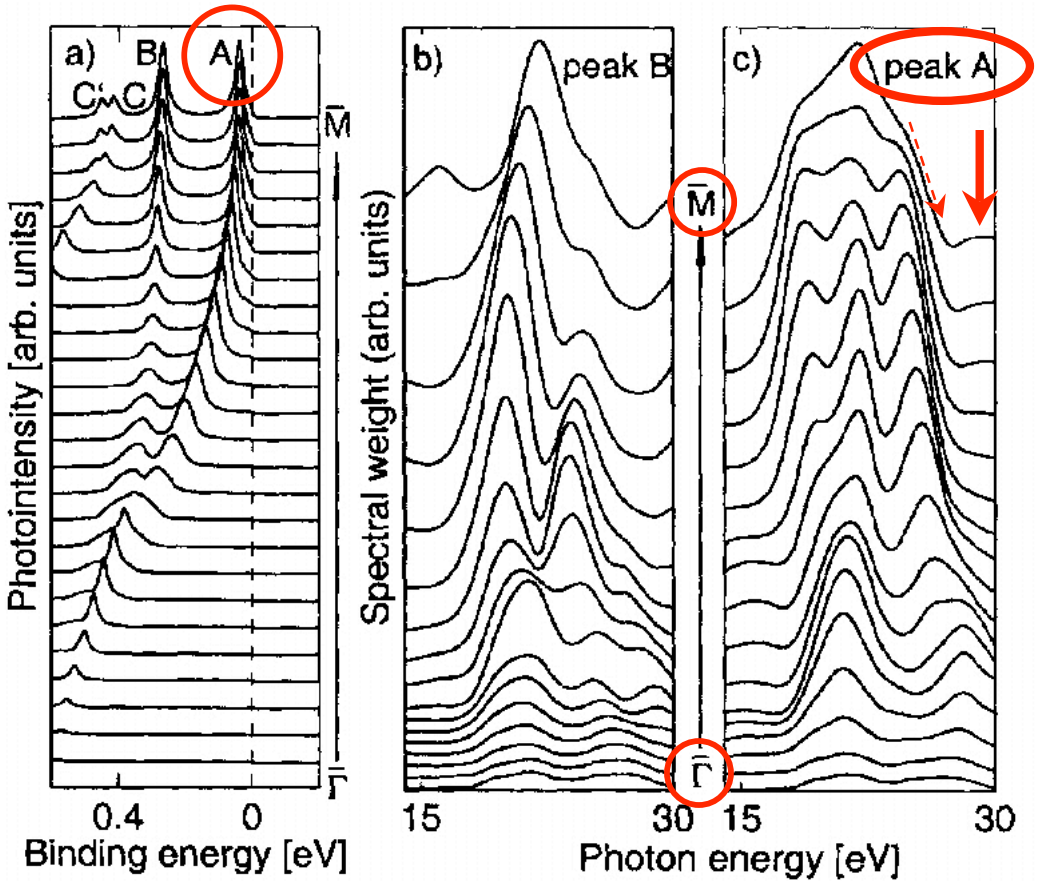
… complicated situation due to FS contour manifolds



Open or closed Fermi surface?

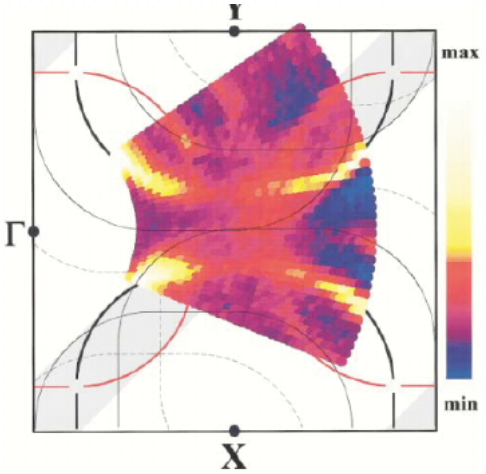
Y.-D. Chuang et al. PRL 83 (1999) 3717





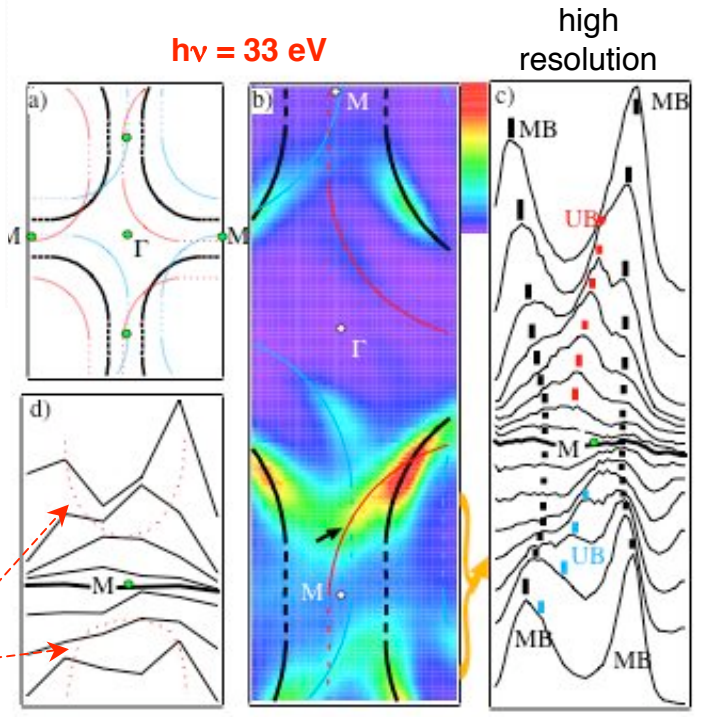
Bansil, Lindroos, PRL 83 (1999) 5154

Matrix elements play a crucial role



Borisenko et al., PRL 84 (2000) 4453

High resolution experiments



Fretwell et al., PRL 84 (2000) 4449

Bi2212: the situation is complicated despite its 2D character

We need to consider
-final state scattering ('5x1')-reconstruction
-matrix elements ($h\nu$ - dependence)

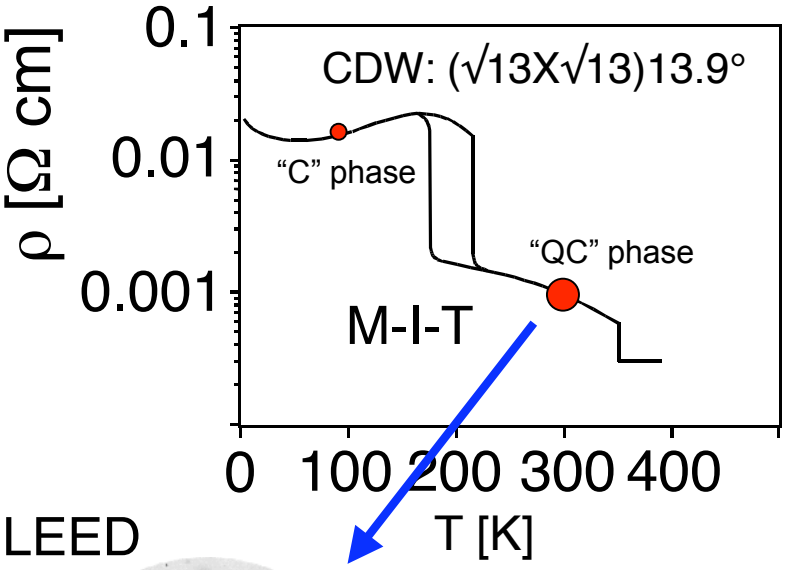
We should have a modular approach:
Module for computing:
Inverse LEED state
+ coupling to initial state
from arbitrary model

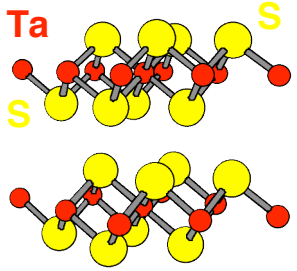
Many different models deal with initial state physics
(calculate: wavefunctions, greensfunction, $A(k,\omega)$)

Final state scattering (LEED, 'conventional')
(calculate inverse LEED state for known structure)

CDW system 1*T*-TaS_2

F. J. Di Salvo *et al*., Solid State Commun. **23**, 825 (1977)

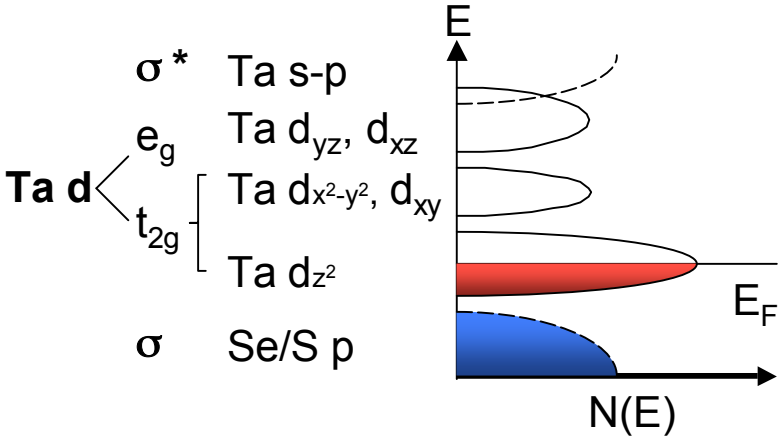




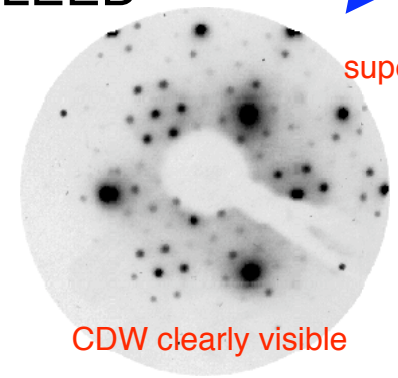
^{73}Ta [Xe]4f^{14}**5d^{3}6s^{2}**

^{16}S [Ne] **3s^{2}3p^{4}**

IONIC PICTURE

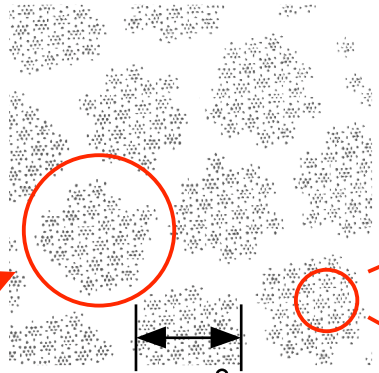


LEED



superspots

CDW clearly visible

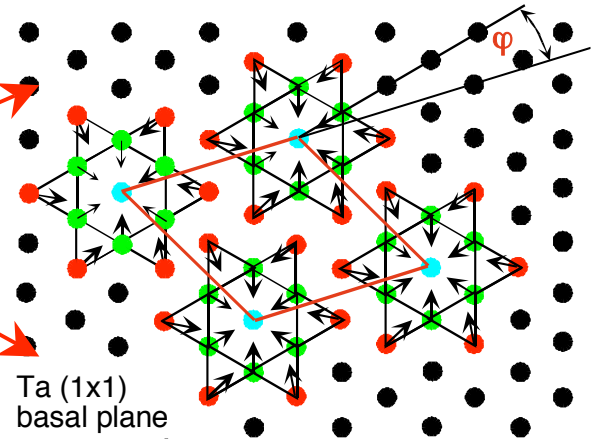


~73 Å

Domains of commensurate CDW

CDW: (√13X√13)13.9°

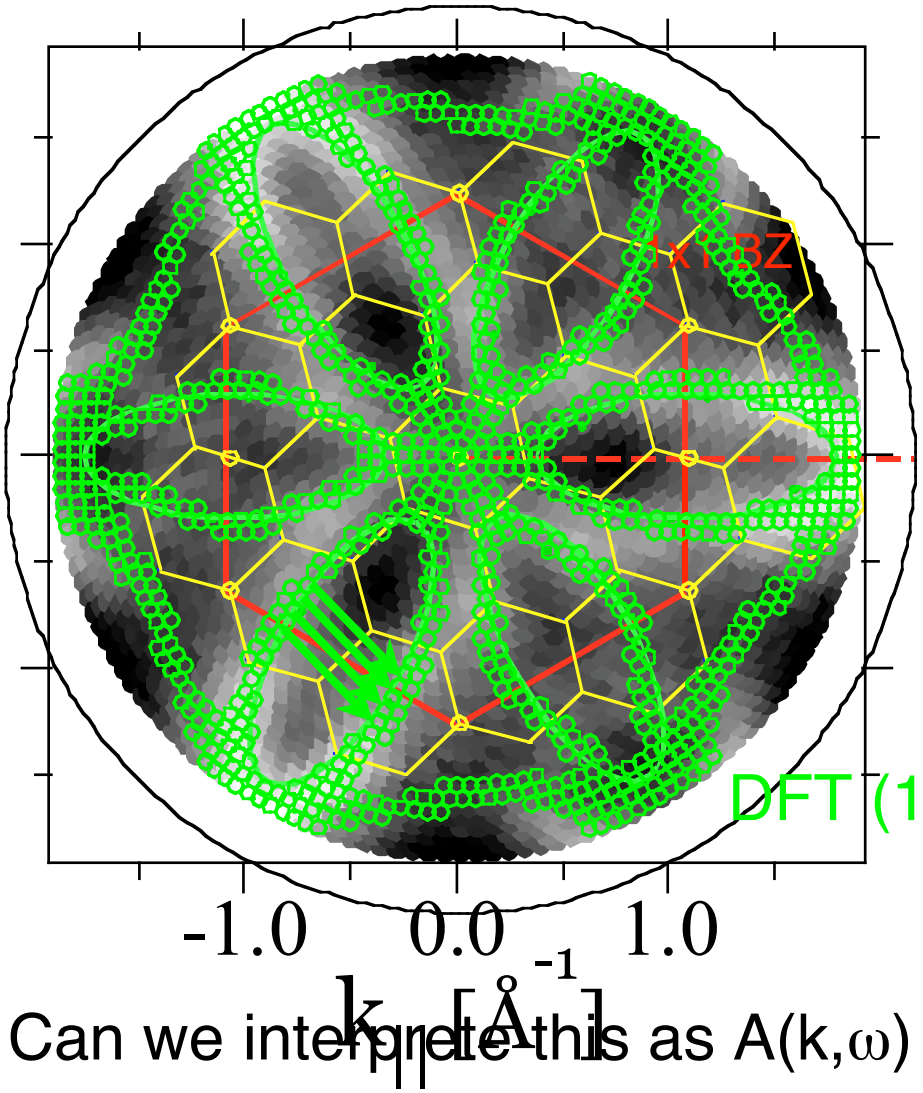
A. Spijkerman et al. Phys. Rev. B 56, 13757 (1997)



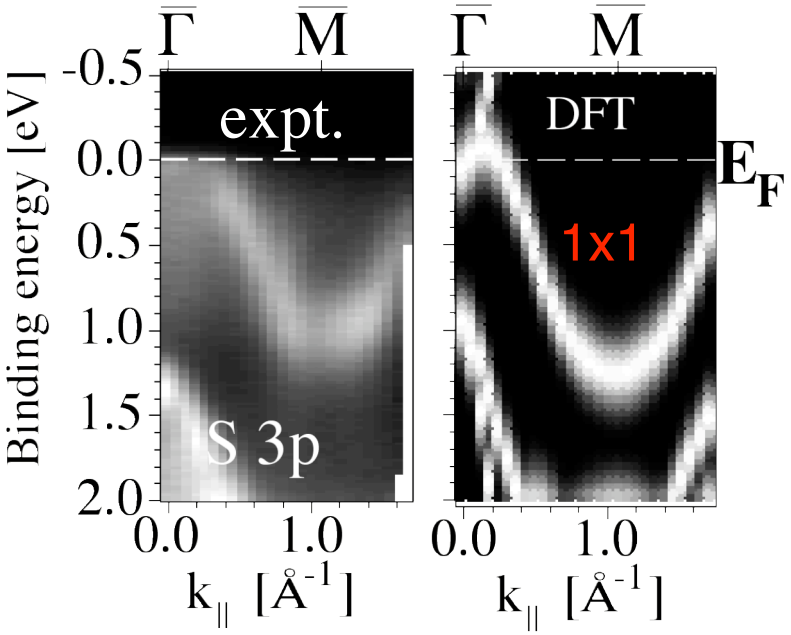
… what do we see in the experiment ...

Phys. Rev. B 69, 125117/1-125117/9 (2004)

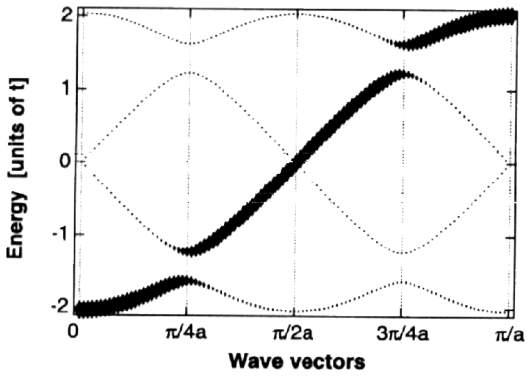
Experiment: FS mapping



Band mapping



No clear effect of CDW?



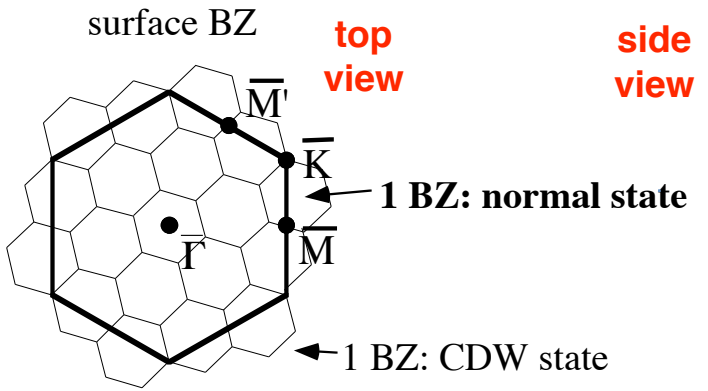
J.Voit et al. Science 290,501 (2001)

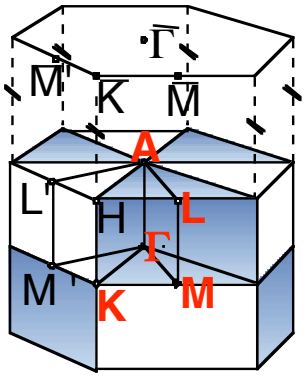
Can we interprete this as A(k,ω) ?

What is ordinary / exotic?

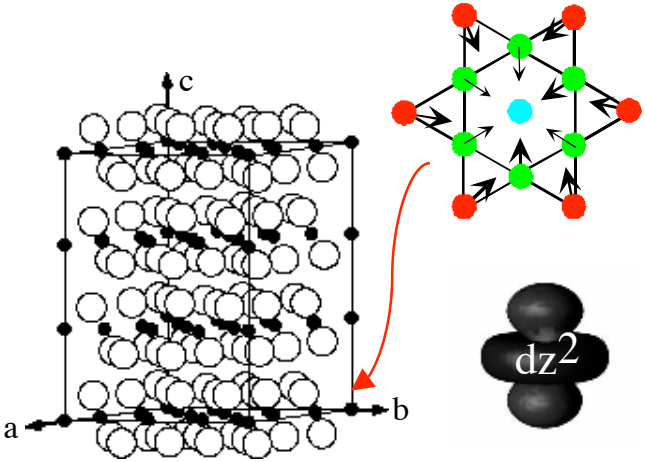
No clear gap opening?

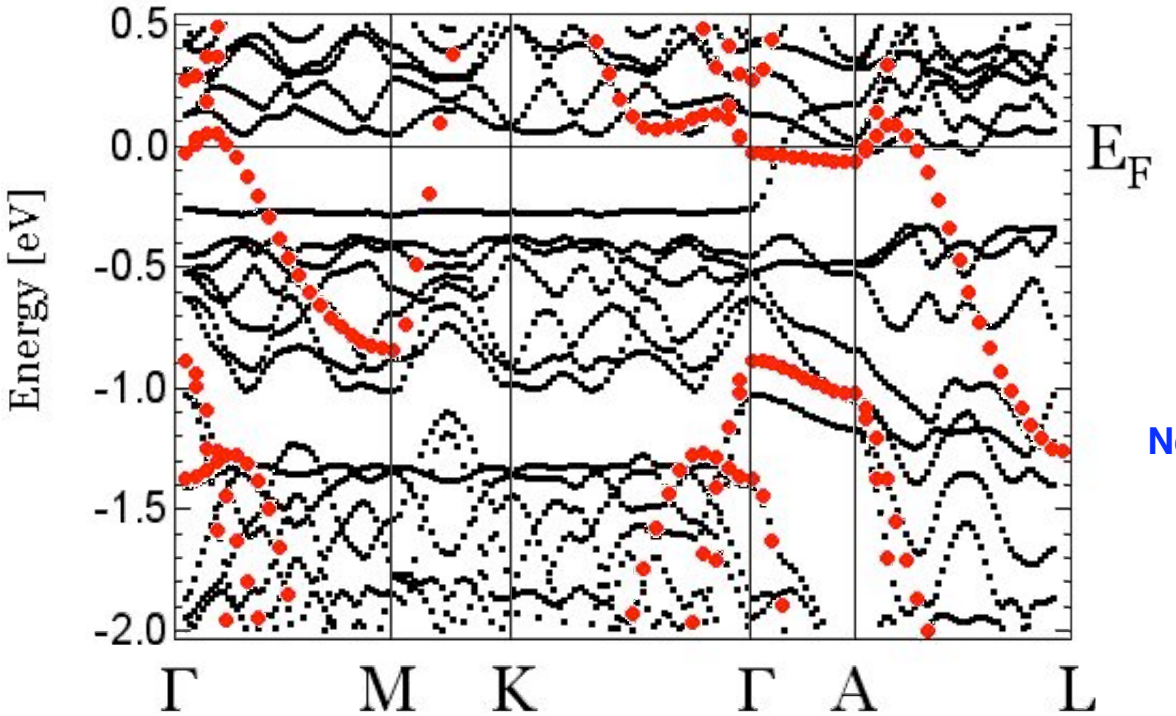
realistic* DFT calculation accounting for CDW





Bulk (1*T*)





Even the smallest CDW amplitude creates new BZs

Reconstructed band structure does not help
Need to quantify weight on new bands !

Influence on A(k,ω) ?

* calculation for expt. XRD structure

Phys. Rev. B 67, 125105 (2003)

CDWs: the situation is complicated despite its 2D character

?How to know?

-what is due to large unit cell (CDW) in initial state

-what is due to large unit cell (CDW) in final state

-what is due to A(k,ω) / quasi particles

-whether low intensity is due to absence of QP or ME

We should have a modular approach:

Many different models go into initial state physics
(calculate: wavefunctions, greensfunction, A(k,ω))

-Module for computing: A(k,ω) for large unit cell

Influence of potential with variable modulation strength

-Module for: Inverse LEED state for large unit cell
+ coupling (ME) to initial state

Experimental information on final states …

VLEED experiments !

VOLUME 81, NUMBER 22 PHYSICAL REVIEW LETTERS 30 NOVEMBER 1998

Absolute Band Mapping by Combined Angle-Dependent Very-Low-Energy Electron Diffraction and Photoemission: Application to Cu

V. N. Strocov,[1,2,*] R. Claessen,[1] G. Nicolay,[1] S. Hüfner,[1] A. Kimura,[3] A. Harasawa,[3] S. Shin,[3] A. Kakizaki,[4] P. O. Nilsson,[2] H. I. Starnberg,[2] and P. Blaha[5]

the method by application to Cu, and find significant deviation from free-electron-like behavior in the unoccupied states, and from density-functional theory in the occupied states. [S0031-9007(98)07792-8]

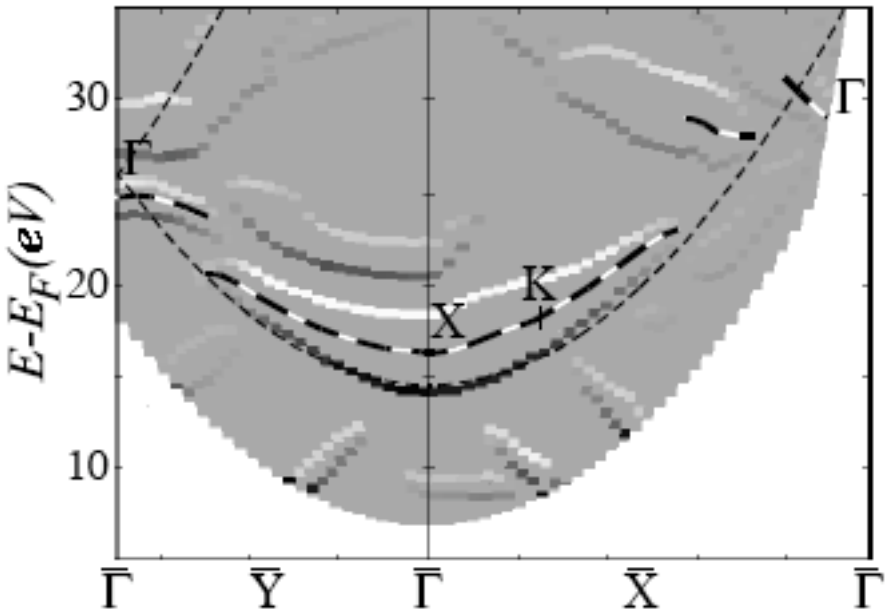


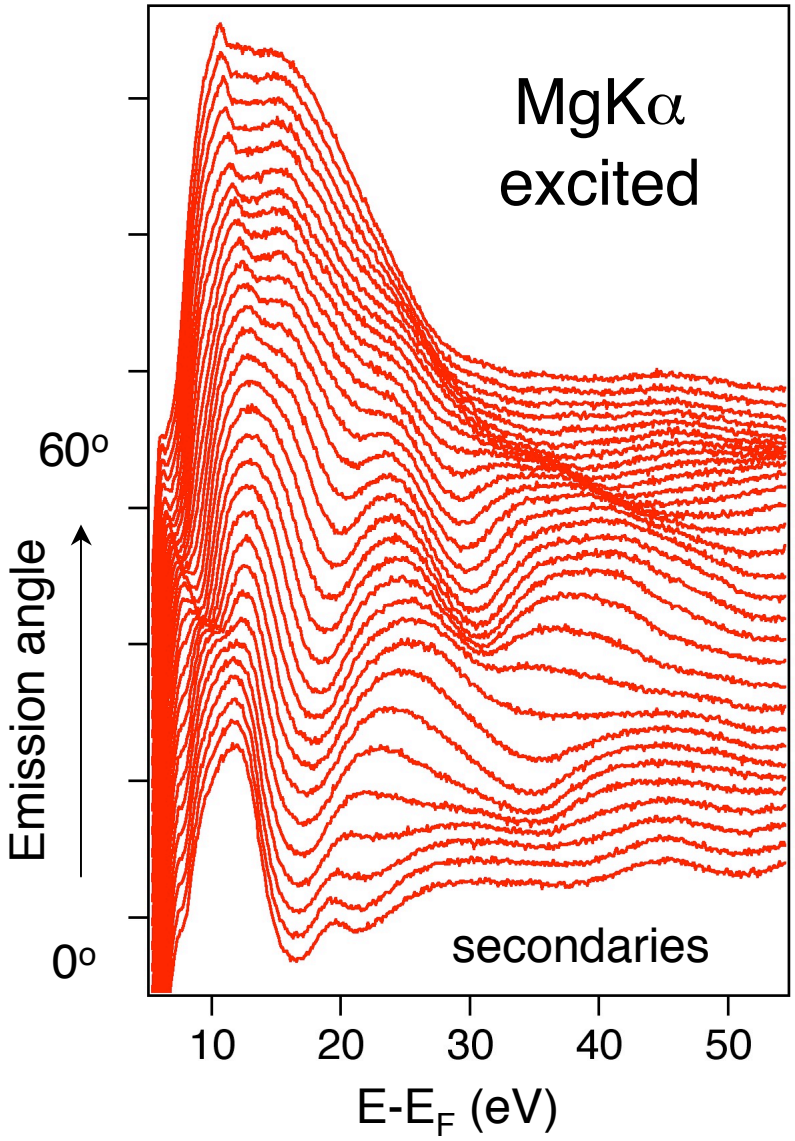
FIG. 3. Experimental upper bands derived from the VLEED dT/dE extrema. Their shading reflects a logarithmic gray scale proportional to $-\frac{d^2(dT/dE)}{dE^2}$, on top of the zero-level gray scale; dark (light) points correspond to dT/dE minima (maxima). The bold dashed curves show the bands of the final-state energies chosen for the CFS PE experiment, with the high-symmetry points in the $\Gamma KLUX$ plane indicated; the thin dashed curve is their free-electron approximation. The region below $E_{\text{vac}} + \frac{\hbar^2 K_\parallel^2}{2m} + 2$ eV is clipped.

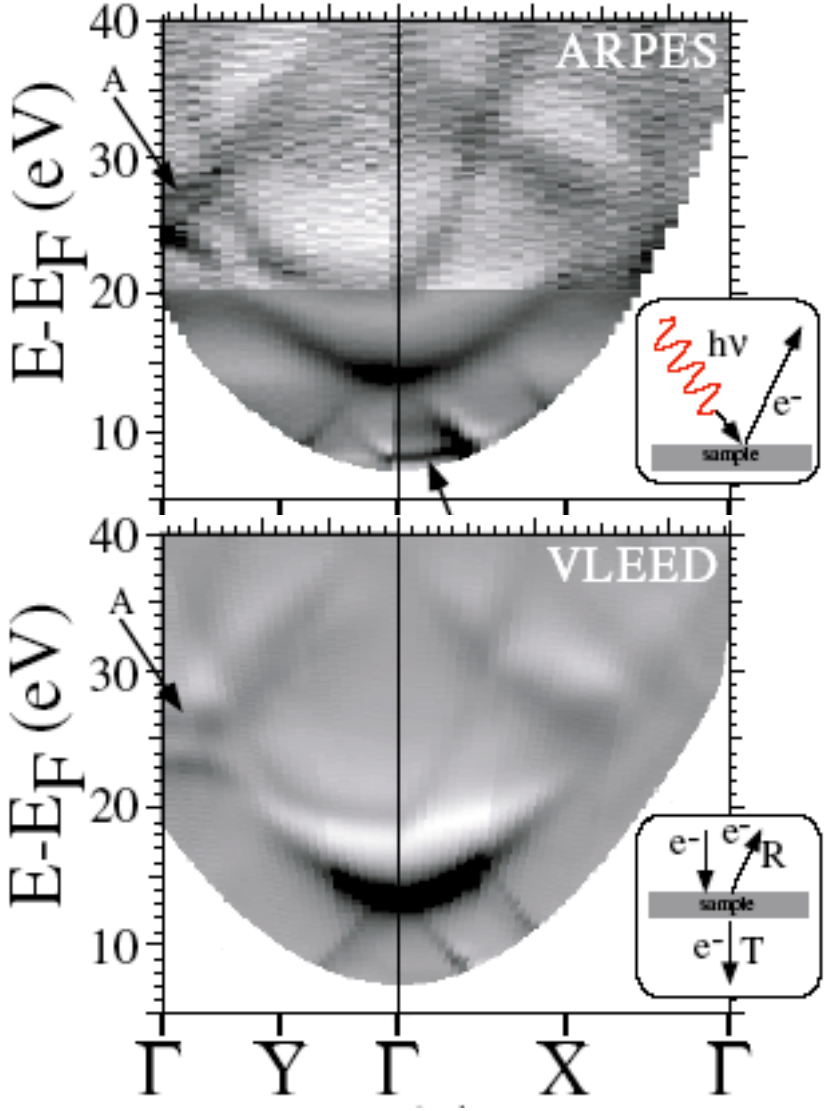
VLEED experiments on Cu(110)

Phys. Rev. Lett., 93, 107601/1 - 107601/4 (2004)

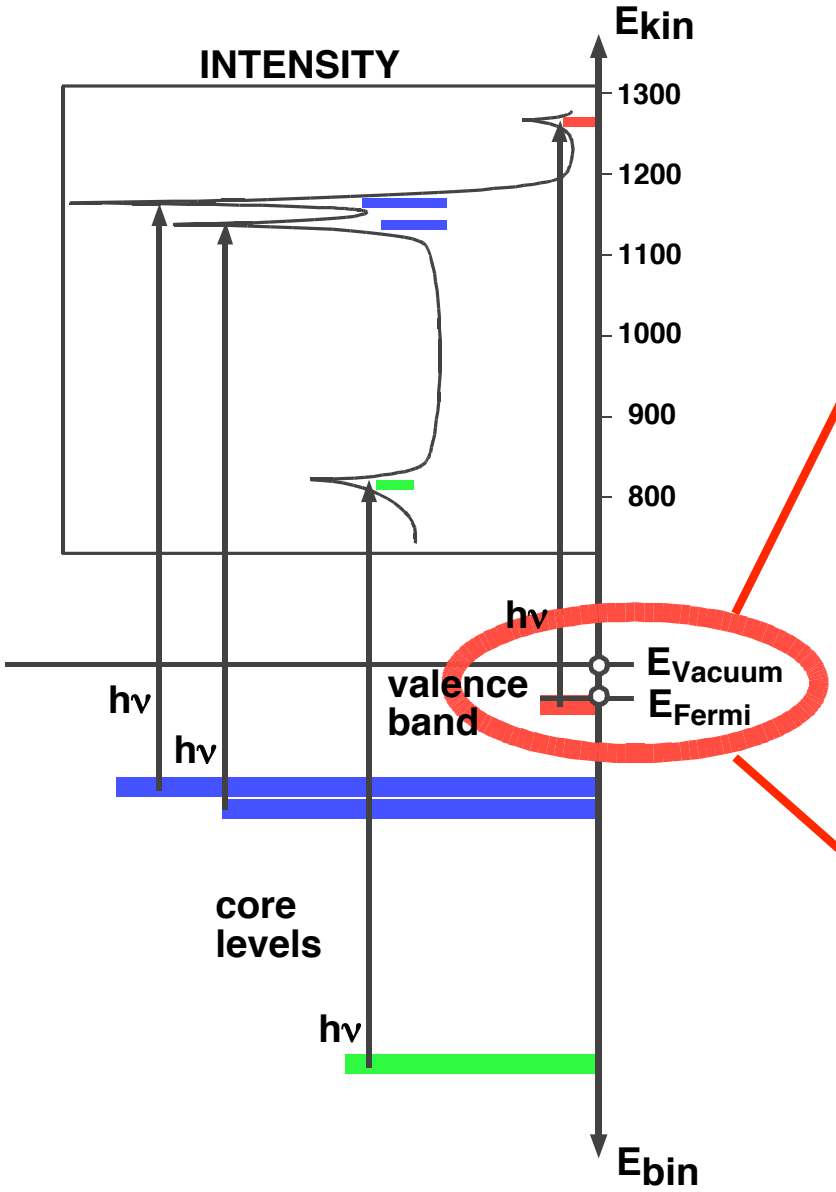
Experimental information on final states …

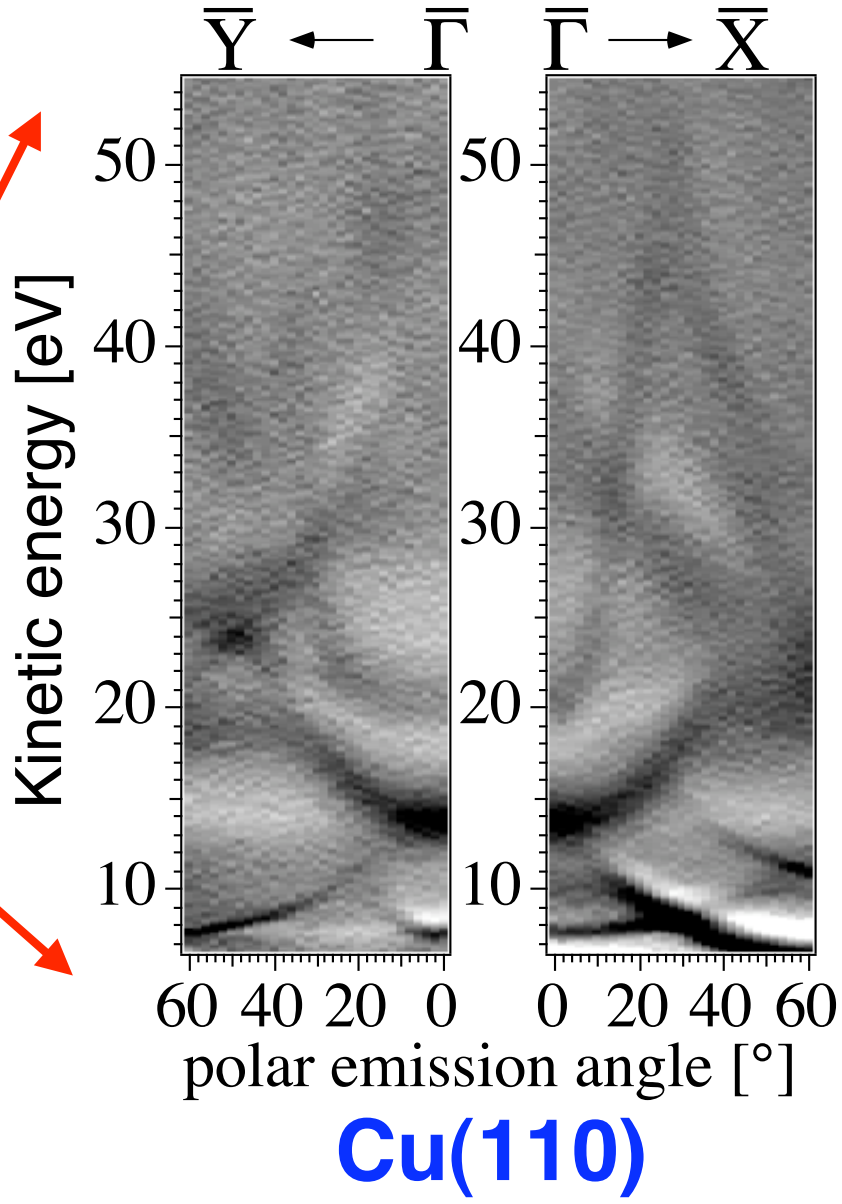
It works with secondary ARPES !





Photoemission … also for un-occupied states





Cu(110)

Conclusion / “Wish list”

Create standard (like in XRD)

democratisation

For final state calculation and Matrix elements,
independent of initial state physics

For ***arbitrary*** initial state physics (LDA, μ-model, etc.)
have a ***standard*** for final state calculation and Matrix elements

Example

Given:

- known (super)structure, different elements A, B,…
- known LDA bandstructure (generally accessible)
  or have a μ-model for physics

Would like to know:

- what hν, polarization, BZ (geometry) ?
  To see best states A, B,… details of μ-model

Photoemission (for quantitative)

Fermi surface- / spectral function- / empty band mapping