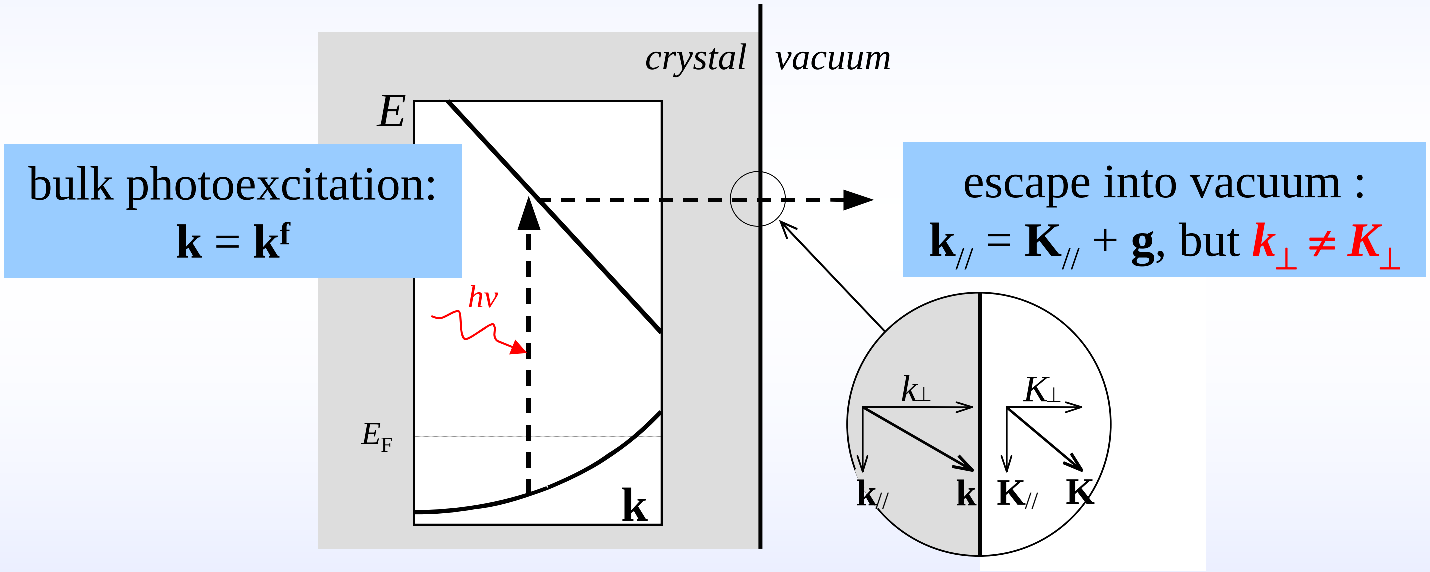
Photoemission final states from the low-energy to high-energy limit: Resolving electronic structure in 3-dim k-space

V.N. Strocov, Swiss Light Source

- Problem of 3-dim wavevector **k** in ARPES:
  - Final-state $k_\perp$ dispersions and lifetimes
- Low energies
  - Final states by VLEED
  - Properties of the final states (non-FE and self-energy effects)
  - 3-dim band mapping by VLEED+ARPES
- High energies
  - Where do the final states become FE-like?

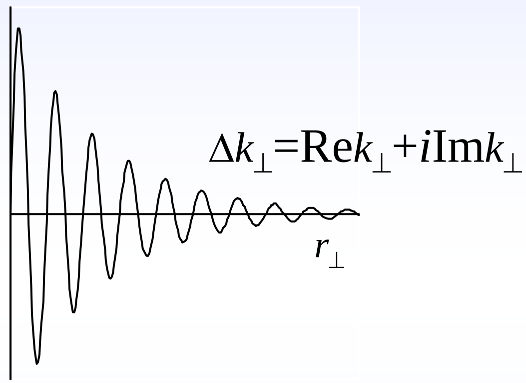
Problem of 3-dim k in ARPES

- Final-state dispersions

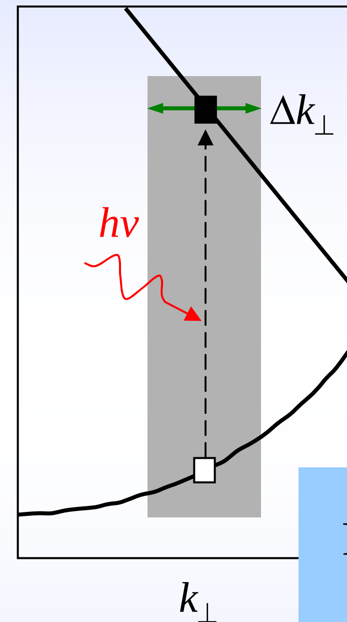


- resolving 3-dim **k** requires final-state $E(k_\perp)$
- low energies: non-free-electron and excited-state self-energy effects in the final states

- Final-state damping



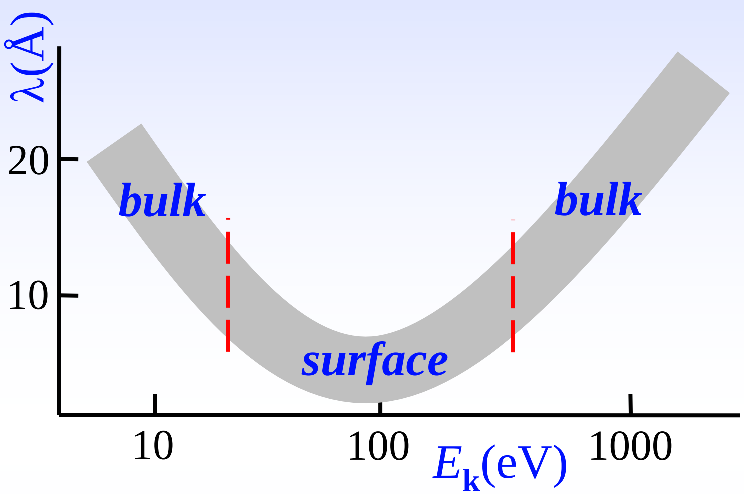
lifetime damping in $r_\perp$ ⇒
broadening $\Delta k_\perp = 2\mathrm{Im}k_\perp$



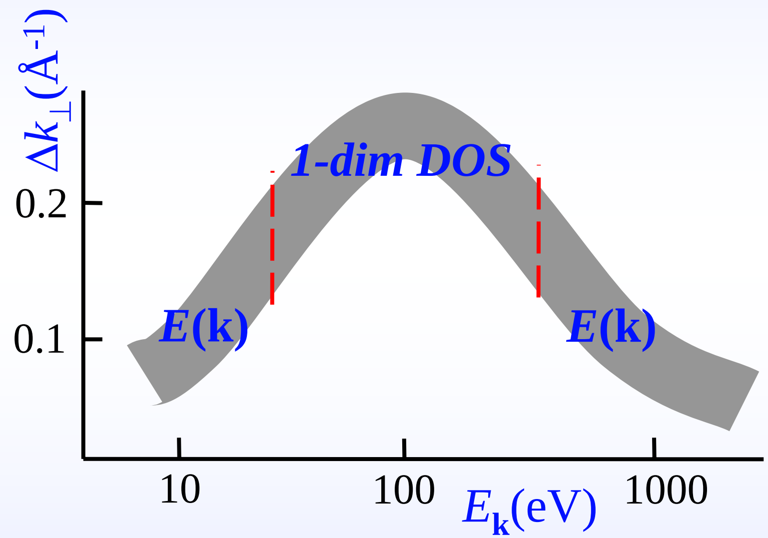
PE signal by *averaging* of $E(k_\perp)$ within $\Delta k_\perp$

- $\Delta k_\perp$ broadening = *intrinsic* $k_\perp$ *resolition*
- spectral peaks ≠ true quasiparticle $E(\mathbf{k})$: *intrinsic accuracy* of 3-dim band mapping

- Two sides "universal" curve



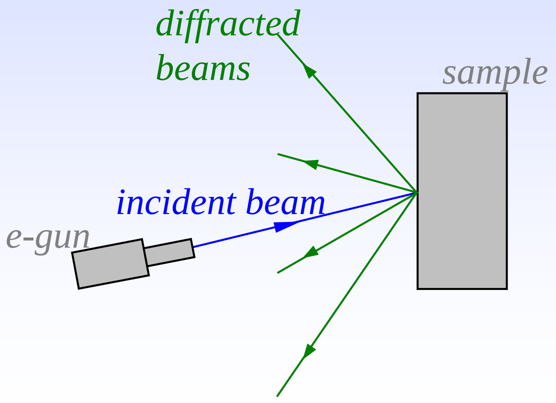
$$\Delta k_\perp = \lambda^{-1}$$



- ARPES regimes: $E(\mathbf{k})$ ($\Delta k_\perp << k_\perp^{BZ}$) → 1-dim DOS ($\Delta k_\perp >> k_\perp^{BZ}$) → $E(\mathbf{k})$ (*Feibelman & Eastman, 1974*)
- low energies: non-free-electron and self-energy effects in the final states by **Very-Low-Energy Electron Diffraction (VLEED)**
- high energies: free-electron final states

Low energies: Final states by VLEED

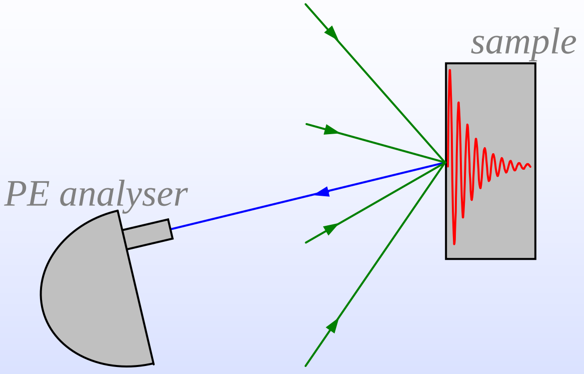
- What is VLEED?



VLEED = measurement of $R(E)$ elastic reflectivity in the energy range below ~40 eV:

• weak $V_i \Rightarrow$ sensitivity to $E(\mathbf{k})$

- Why VLEED?



One-step ARPES theory: $I^{ph} = \left| \left\langle \Phi^{LEED*} \middle| \hat{A} \cdot \hat{p} \middle| \Phi^{i} \right\rangle \right|^2$

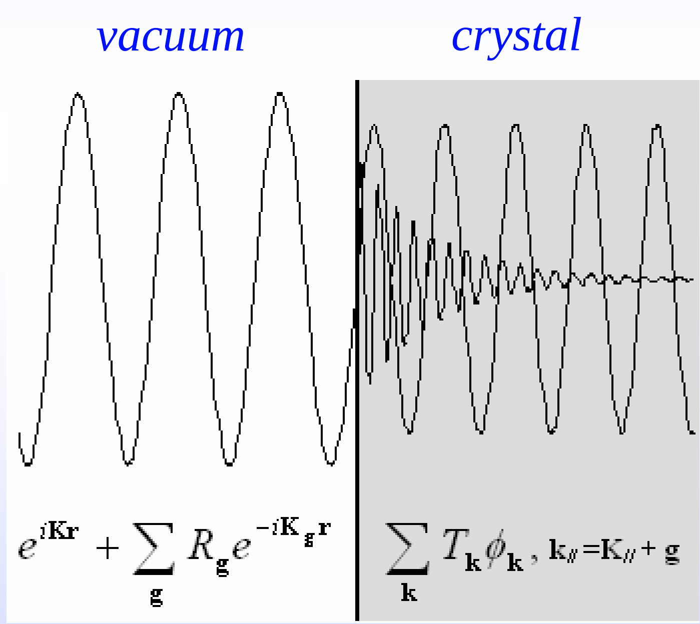
• direct connection to ARPES:
ARPES final state = time-inversed LEED state

• final-state energies in ARPES

Connection of VLEED to *E*(k)

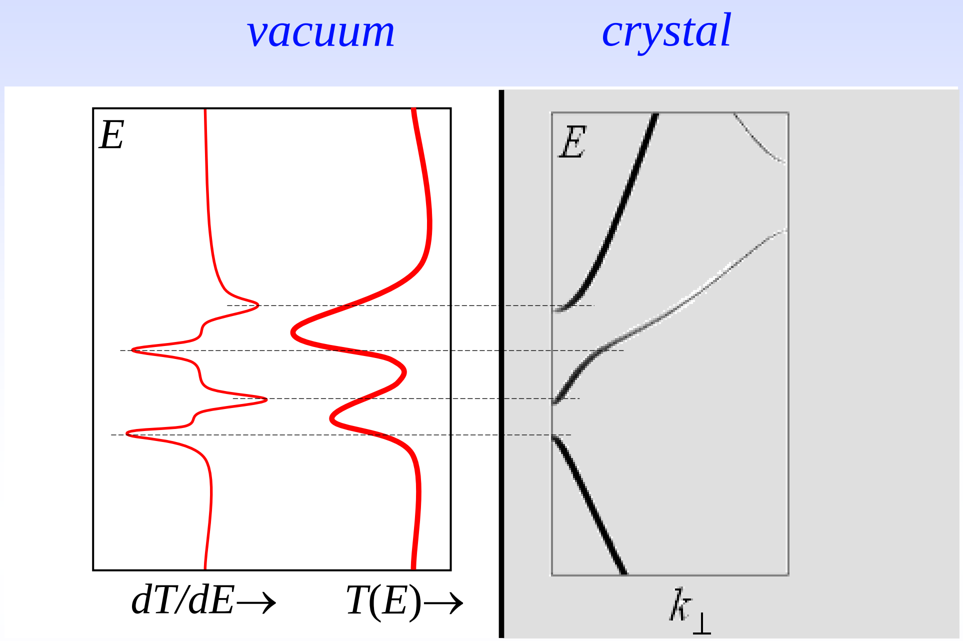
- elastic case (inelastic scattering = 0)

$R(E) = 1 - T(E)$ by matching vacuum and crystal wavefunctions:



VLEED spectrum $T(E) \rightarrow \quad \leftarrow \; E(k_{\perp})$ along $\mathbf{k}_{//} = \mathbf{K}_{//} + \mathbf{g}$

Final states by VLEED: Connection of VLEED to $E(\mathbf{k})$

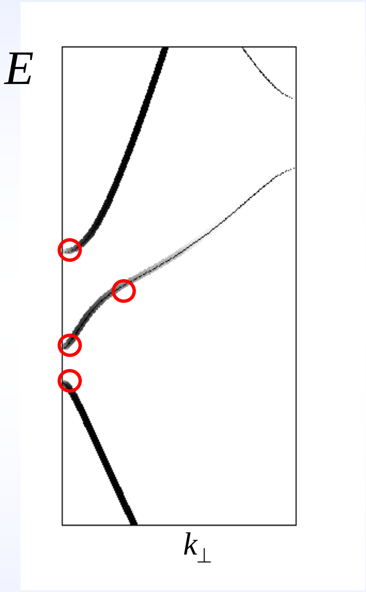


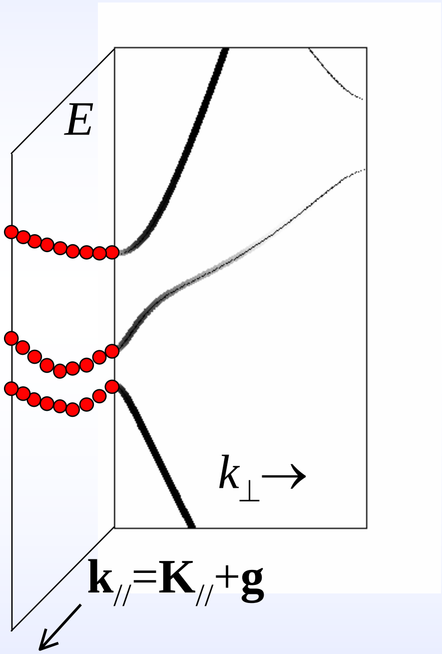
$T(E)$ minima/maxima → ← band gaps/dispersions ranges in $E(k_{\perp})$

dT/dE extremes → ← critical points in $E(k_{\perp})$

VLEED structures = critical points in $E(k_{\perp})$

Band mapping techniques



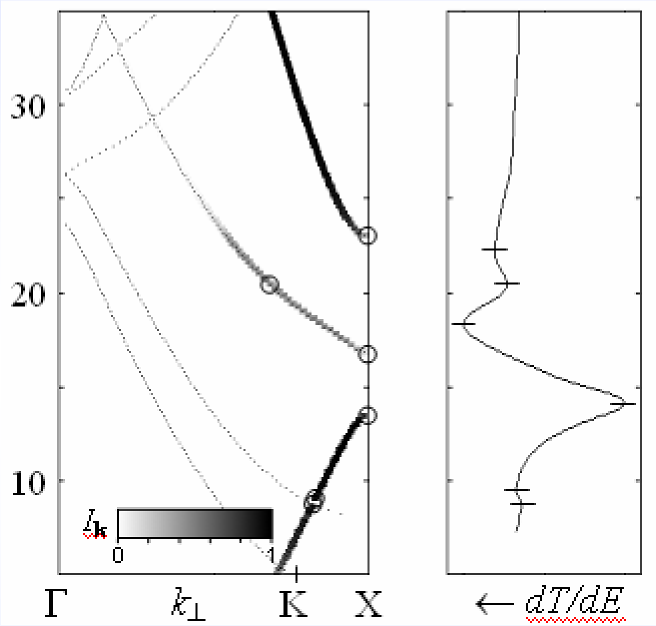


$E(k_\perp)$: fitting experimental critical points

$E(\mathbf{k}_{//})$: direct band mapping along symmetry lines

Effects of different bands

- normal-incidence VLEED on Cu(110)

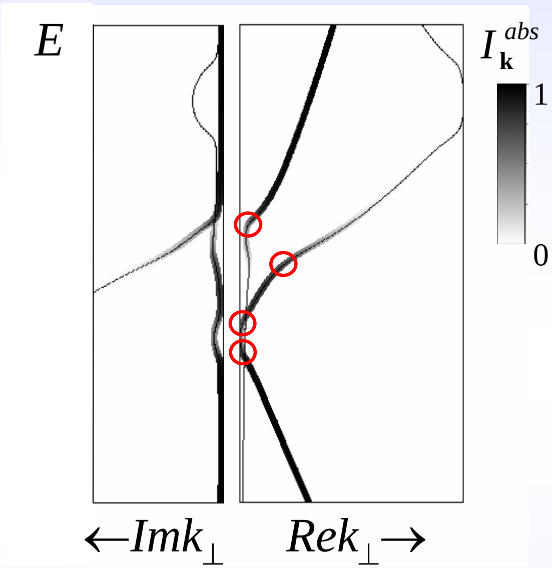


• VLEED spectrum ~ *conducting* bands: large *partial currents* $I_\mathbf{k} = |T_\mathbf{k}|^2 \upsilon_\perp$ (effective coupling to vacuum + transport into crystal)

• $I_\mathbf{k}^{\mathbf{VLEED}} \sim I_\mathbf{k}^{\mathbf{PE}}$: the same conducting bands effective in VLEED and ARPES

Effect of inelastic scattering ($V_i \neq 0$)

- damped Bloch waves with complex $k_\perp = \mathrm{Re}k_\perp + i\mathrm{Im}k_\perp$
- smooth $E(k_\perp)$ dispersions



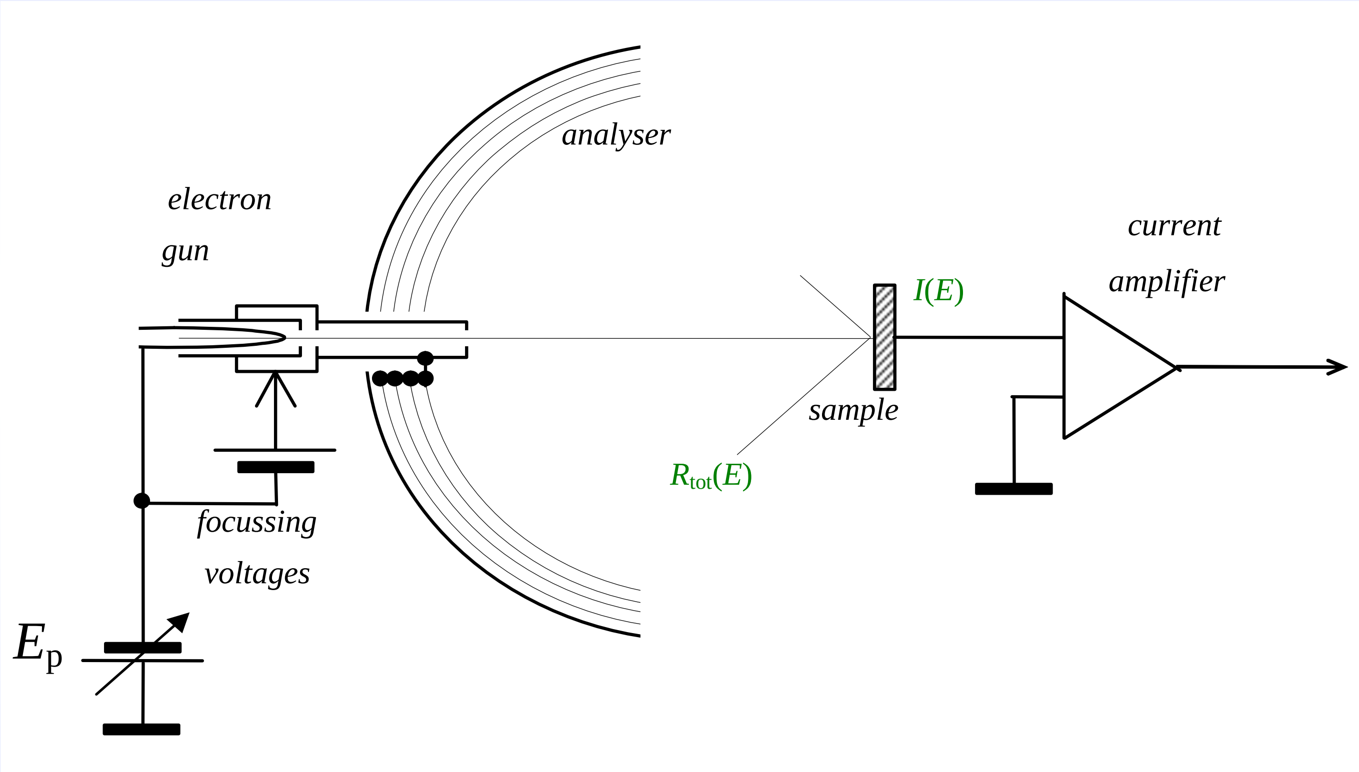
$$I_{\mathbf{k}}^{abs} \propto V_i \int_{crystal} \left| T_{\mathbf{k}} \phi_{\mathbf{k}} \right|^2 d\mathbf{r}$$

critical points = extremes in 1/curvature of $E(k_\perp)$

- VLEED structures { energies → critical points in conducting bands $E(k_\perp)$; broadening → $V_i = \hbar/\tau$ }

Experimental technique

- Experimental setup



- *retarding field* to maintain focusing down to E_p~0
- I_{gun}=*const* ⇒ measurements of $T(E)$ ∝ - integral reflectivity $R(E)$ in *target current*
- data acquisition time <1 min/spectrum

- Retarding-field: Angle dependences

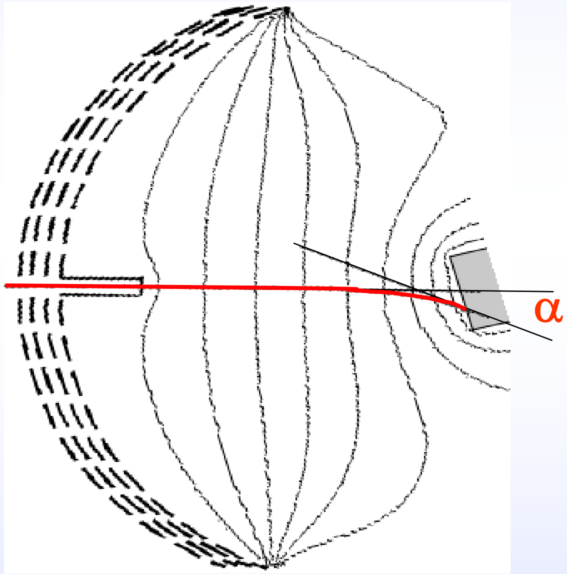
• α- and *E*-dependent deviation of off-normal trajectories and variation of incidence angle

• necessity to control $\mathbf{K}_{//}$:

- electrostatic ray-tracing

$$\sum_i \frac{\partial^2 U}{\partial x_i^2} = 0 \quad + \quad \frac{\partial^2 x_i}{\partial t^2} = -\frac{e}{m}\frac{\partial U}{\partial x_i}, \quad x_i = x, y, z$$

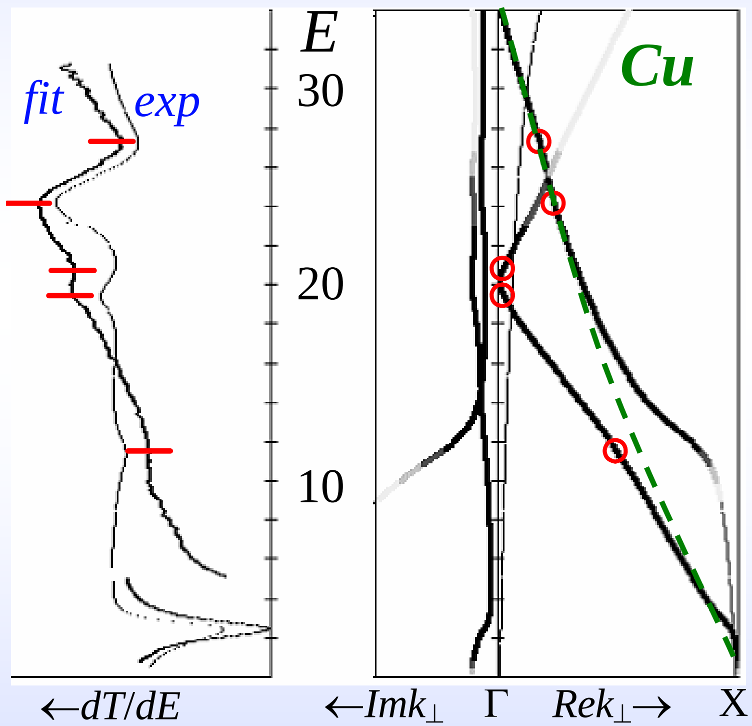
- $K_{//}(\alpha, E) = \sum_{m,n=0}^{2} A_{mn} E^m \alpha^n$ fitted to experimental points with well-defined $\mathbf{K}_{//}$



3. Experimental properties of the final states

Non-free-electron effects

- $E(k_\perp)$ from normal-incidence VLEED on Cu(100)

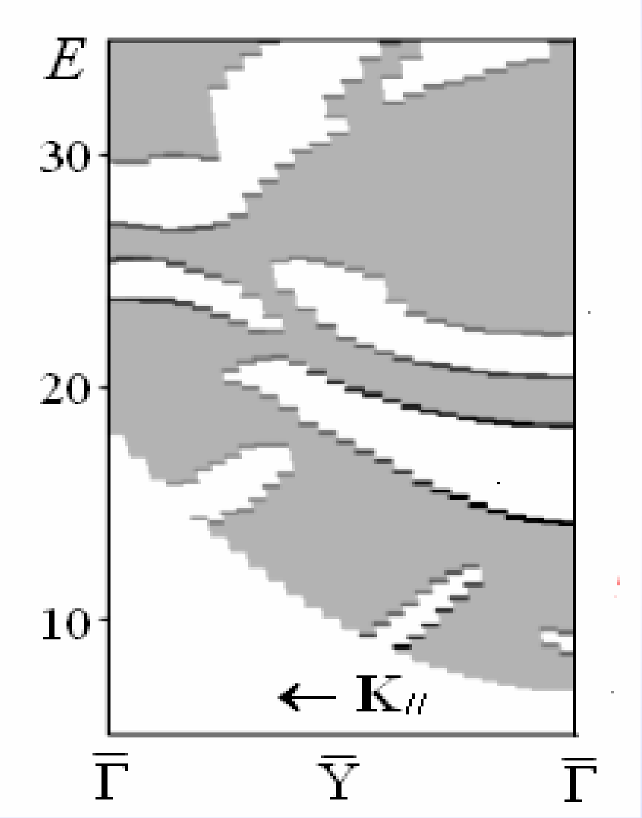


• deviations from FE-like model:

- non-parabolic dispersions (local m^*, V_{000})
- *multiband* composition (different $k_\perp$ available in the final state)

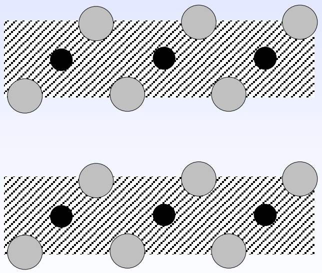
- $E(\mathbf{k}_{//})$ from angle-dependent VLEED on Cu(110)

VLEED $\mathbf{K}_{//}$ dispersions:
$T(E)$ maxima (gray) = $k_\perp$ dispersion ranges,
$T(E)$ minima (white) = band gaps
= *surface-projected* $E(\mathbf{k})$ of the final states



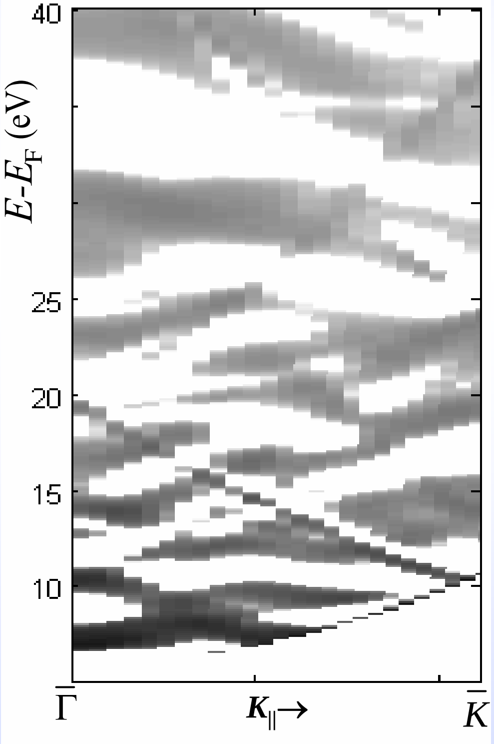
- deviations from FE-like dispersions closer to the BZ borders

- *E*(k) of layered material $TiTe_2$



• anisotropic quasi-2-dim properties

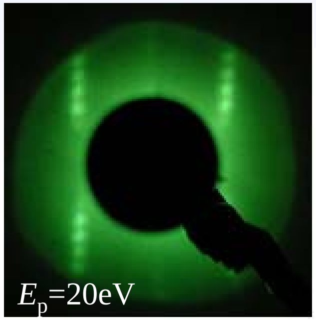
$E(k_{//})$ = VLEED $K_{//}$ dispersion

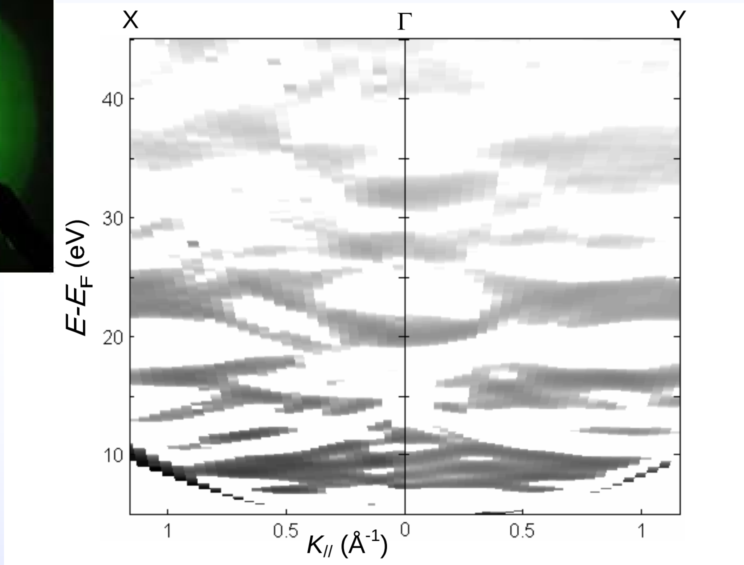


• prominent non-FE effects

• similar strength of non-FE effects in other layered materials (graphite, VSe_2, $TiTe_2$, $NbSe_2$…) due to sharp modulations of the crystal potential

- *E*(k) of high-T_c material $Bi_2Sr_2CaCu_2O_8$

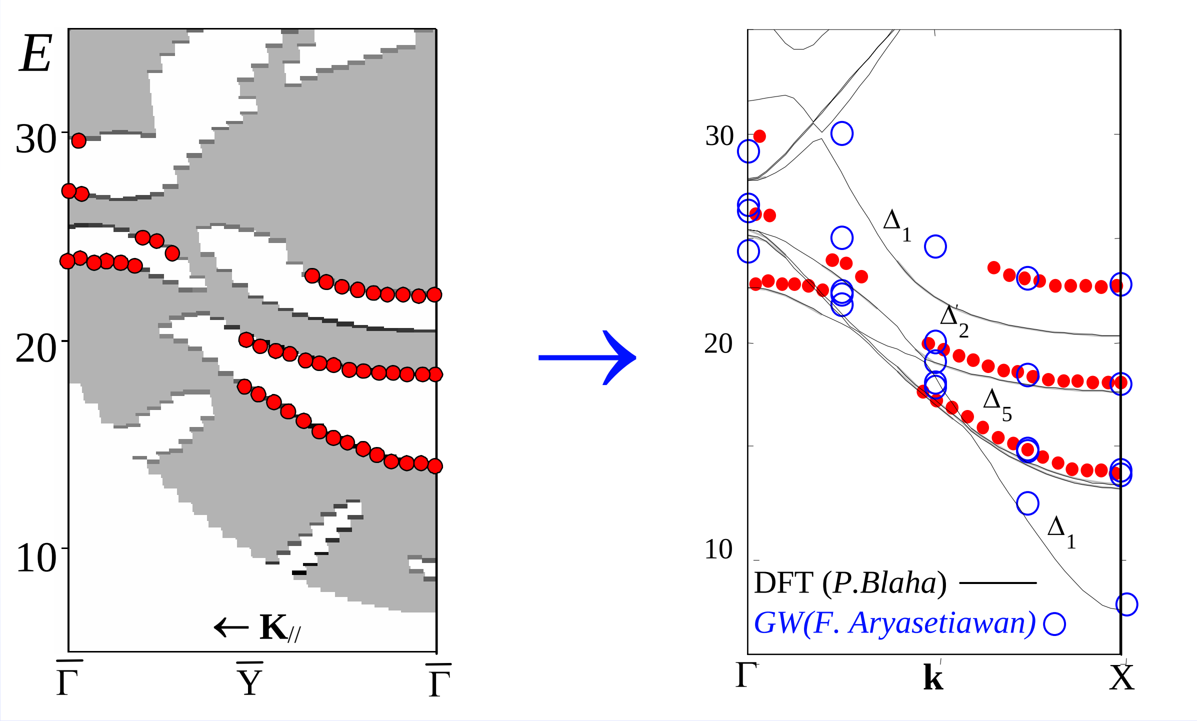




- 3D final-state dispersions
- highly structured final states => dramatic *hv* and $\mathbf{K}_{//}$ dependences in ARPES

Self-energy effects

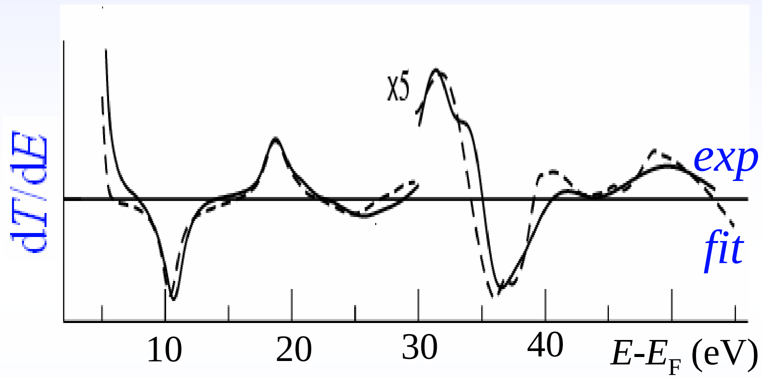
- ΔΣ(*E*,k) in Cu by angle-dependent VLEED on (110) surface



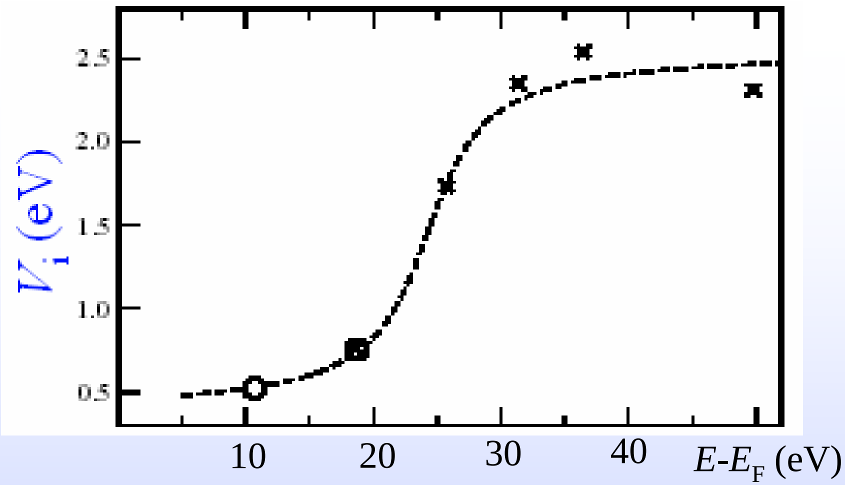
- Excited-state Σ(*E*,**k**,**k**') ≠ static V_{xc} ⇒ ΔΣ
- Band- and **k**-dependence of ΔΣ

Lifetimes

- $V_i(E)=h/\tau$ in graphite *(Barrett et al, 2005)*



• $V_i(E)$ by fitting the sharpness and relative amplitude of dT/dE structures



• Typically sharp increase of V_i at $\hbar\omega_p$

VLEED *vs* other spectroscopies for unoccupied states

- VLEED *vs* XAS and IPES

- resolution in 3-dim **k**
- single electron state (XAS: core hole excitonic effects)
- direct connection with ARPES
- experimental simplicity

- VLEED *vs* SEE

- Recent study on Cu(110) *(Bovet et al, 2004)*: **SEE and VLEED equivalent**
- Thermodynamic model of SEE (*Feder & Pendry*, 1978): $I_{\mathrm{SEE}}(E) \propto T_{\mathrm{VLEED}}(E) + bkg$

3-dim band mapping by VLEED+ARPES

- Connection between VLEED and ARPES

General connection: $I^{\mathrm{ph}} = \left| \left\langle \Phi^{\mathrm{LEED}*} \middle| \hat{\mathrm{A}} \cdot \hat{\mathrm{p}} \middle| \Phi^{\mathrm{i}} \right\rangle \right|^2$

Detailed connection between
the *partial absorbed currents*
and *partial photocurrents*:

$$I_{\mathbf{k}}^{\mathrm{ph}} \propto I_{\mathbf{k}}^{\mathrm{abs}} \cdot \left(\frac{1}{V_i} \frac{\partial k_\perp}{\partial E_i} \left| M_{\mathrm{fi}} \right|^2 \right)$$

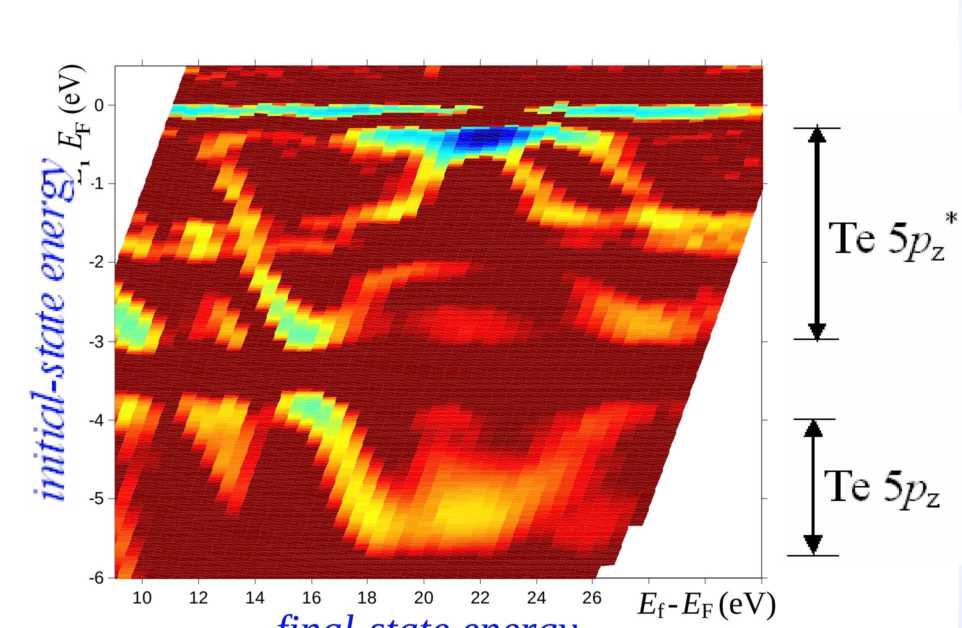
• VLEED coupling bands = PE dominant final bands

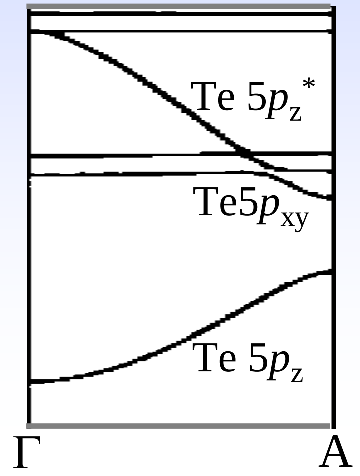
Final-state { $E(\mathbf{k})$, lifetimes } → Valence-band { $E(\mathbf{k})$ resolved in 3-dim $\mathbf{k}$, controlled intrinsic accuracy }

Mapping in $k_\perp$ (photon energy variation): Quasi-2-dim $TiTe_2$

- ARPES data

$-d^2I/dE^2$ intensity map (log scale)



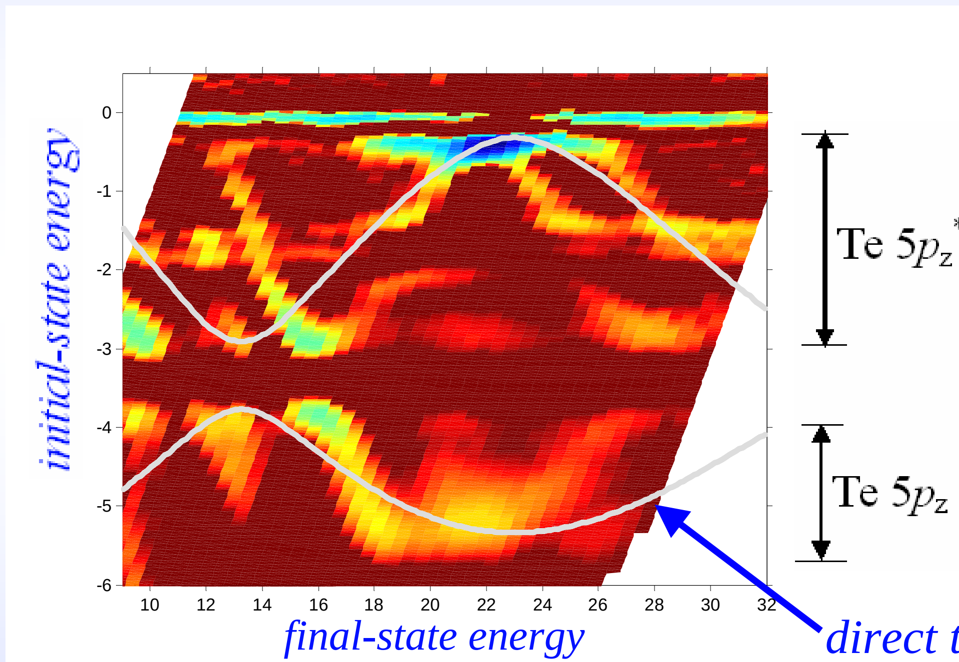


• non-parabolic and multiple Te $5p_z/p_z^*$ dispersions

Mapping in $k_\perp$ (photon energy variation): Quasi-2-dim $TiTe_2$

- ARPES data

$-d^2I/dE^2$ intensity map (log scale)



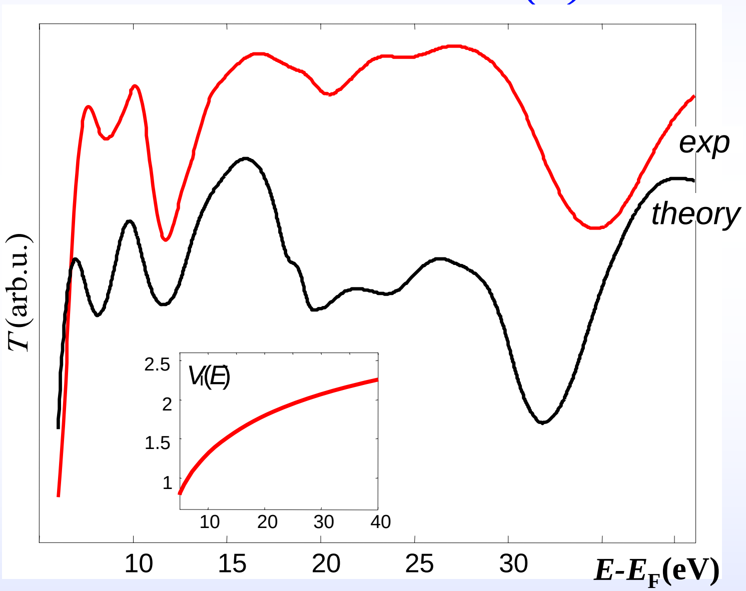
direct transitions to FE final bands with V_{000}=14.5 *eV*

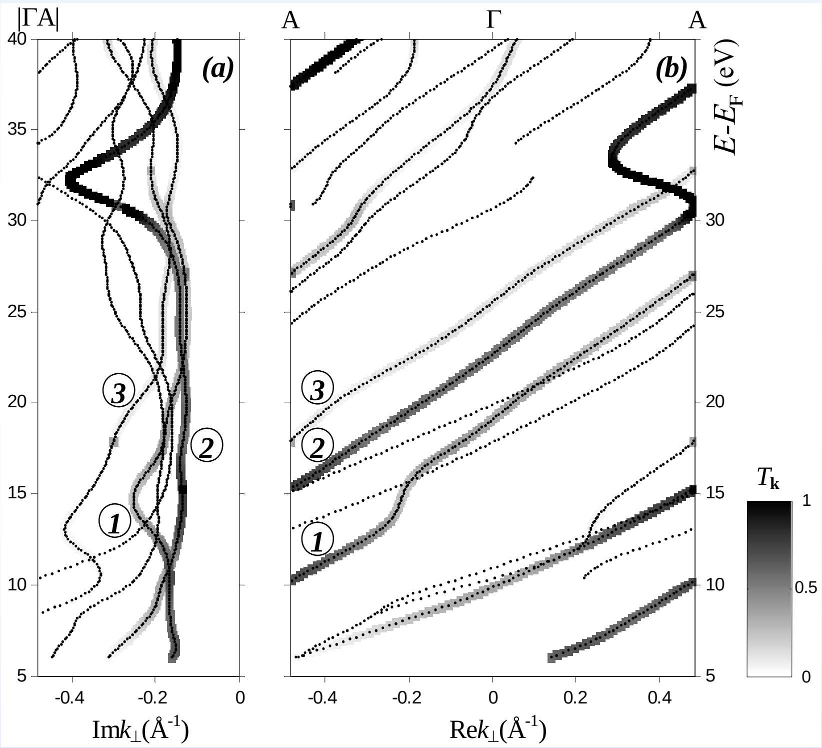
- failure of the FE approximation to describe the experimental Te $5p_z/p_z^*$ dispersions

- Final states

- final states by **k·p** + ELAPW calculations *(E.E. Krasovskii)* with $V_i(E)$ fitted to the experiment + $\Delta\Sigma$ corrections

normal-incidence T(E)

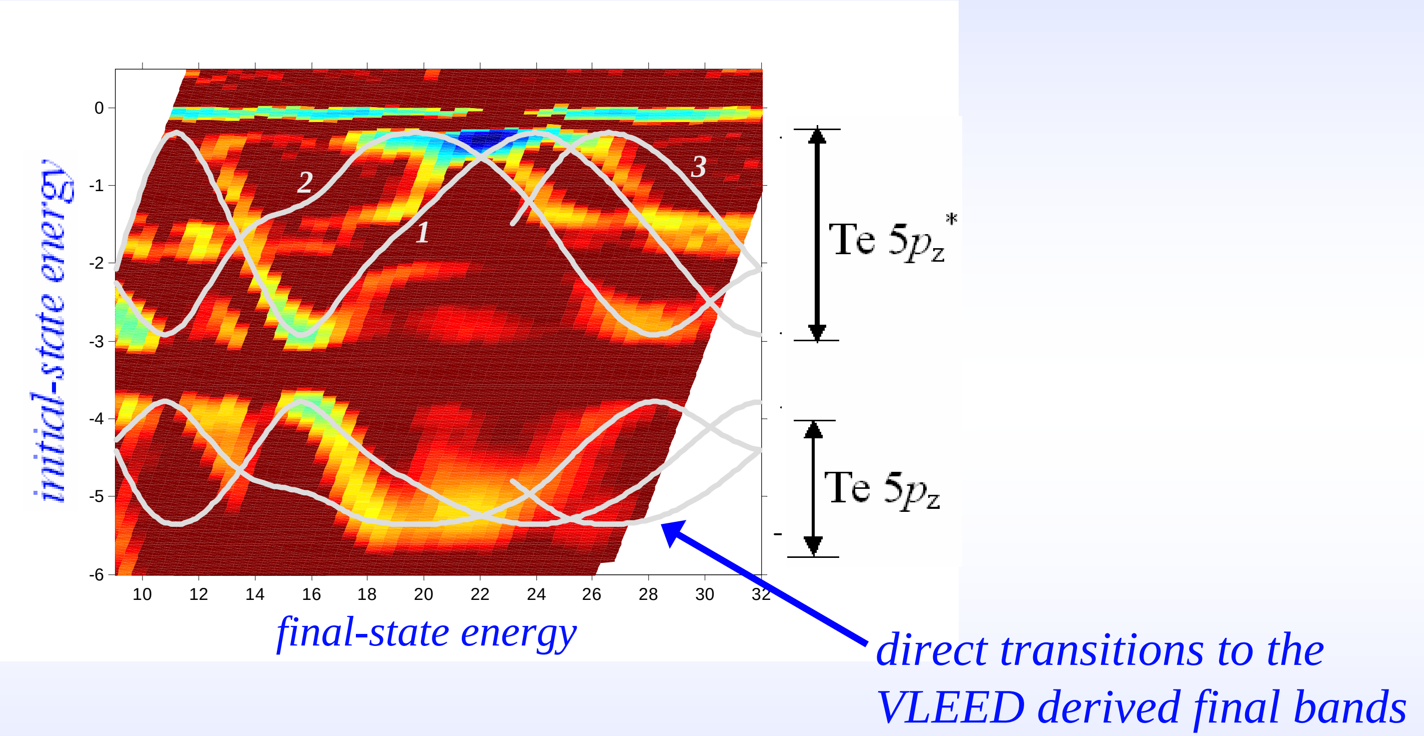




- dramatic non-FE effects: non-parabolic dispersions + multiband composition
- $\Delta k_\perp = \mathrm{Im}k_\perp \ll k_\perp^{BZ}$ – band structure regime

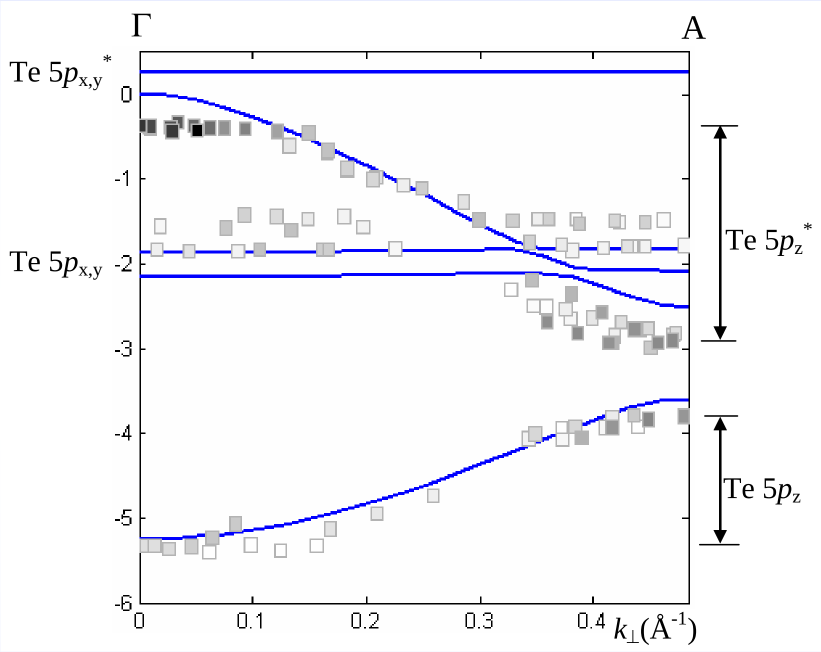
- ARPES data vs VLEED final states

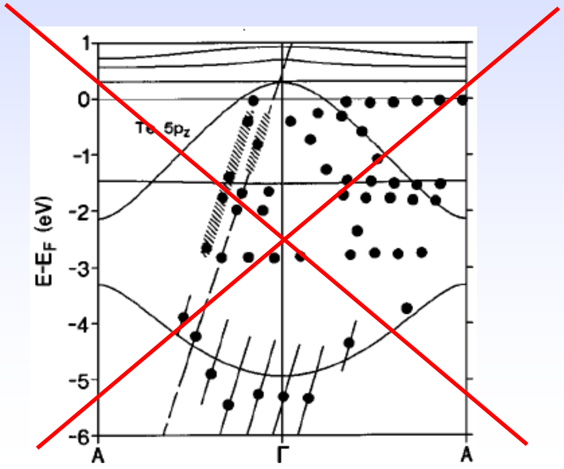
$-d^2I/dE^2$ intensity map (log scale)



- the VLEED derived final states (including the non-FE and ΔΣ effects) reproduce the experimental non-parabolic and multiple Te 5p_z/p_z* dispersions
- remnant disagreement due to intrinsic shifts from the direct transitions
- multiple final bands = common phenomenon of 'umklapp bands' or 'secondary cone' emission

- Band mapping



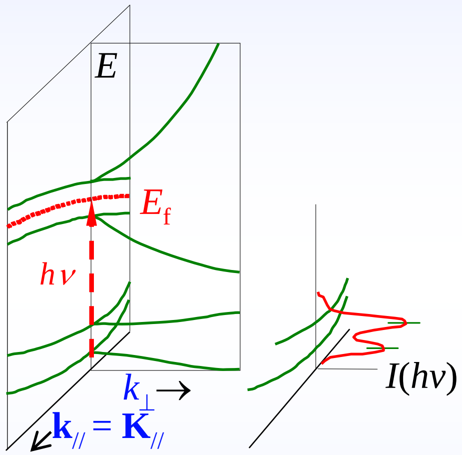


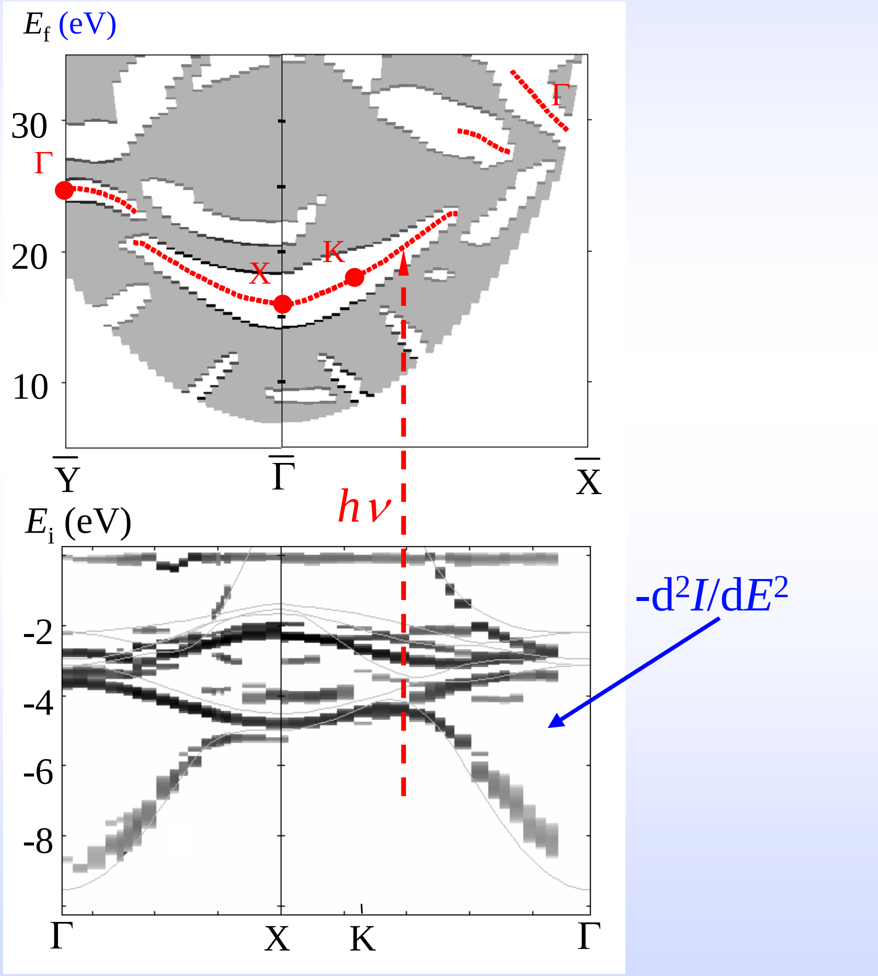
- consistent $E(k_\perp)$ in contrast to the FE final states
- Te 5p_z* does not cross $E_F \rightarrow$ no FS electron pocket in Γ
- true final states incorporating the non-FE and ΔΣ effects are essential (VSe_2, TiS_2 …)

Mapping in $K_{//}$ (emission angle variation)

- Results on Cu

- Idea





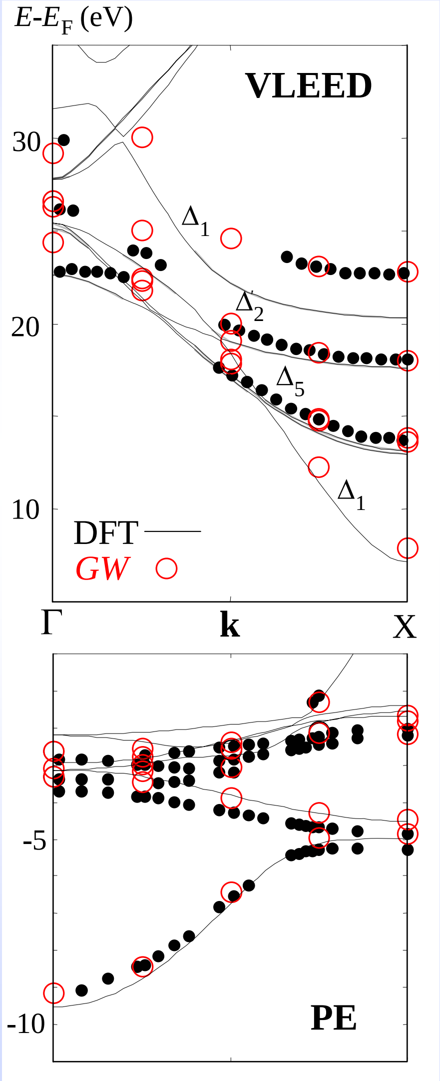
- Angle-dependent VLEED + Constant-Final-State ARPES:

- 1DOS maxima ⇒ intensity gain
- many directions at one surface
- direct image of valence band $E(\mathbf{k})$

- **k-** and band-dependent ΔΣ in the valence band

Self-energy effects in Cu

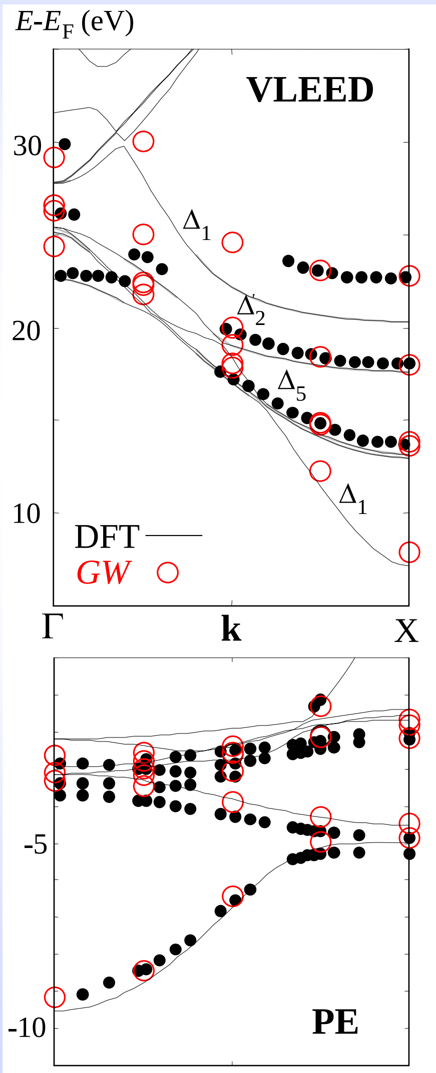
- VLEED experiment *vs* DFT and *GW*

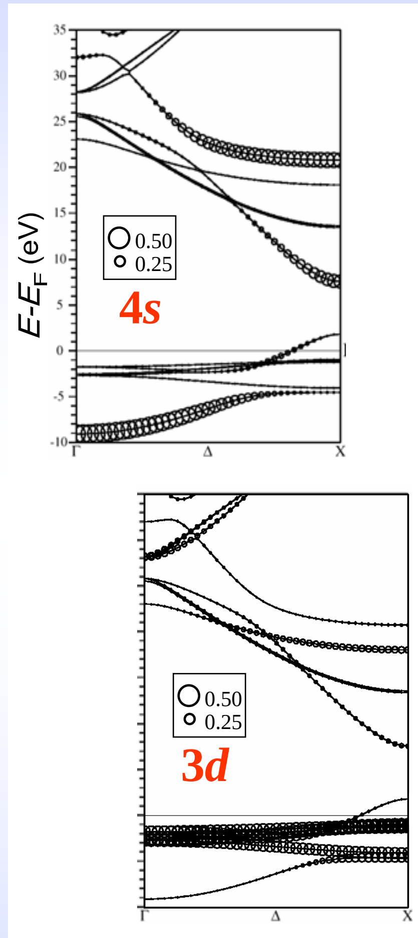


- Significant band- and **k**-dependent ΔΣ renormalization thru unoccupied and occupied *E*(**k**) despite weakly correlated nature of Cu
- Agreement with *GW* quasiparticle *E*(**k**)

(Strocov, Claessen, Aryasetiawan, Blaha & Nilsson, 2002)

- Mechanism: Non-local exchange



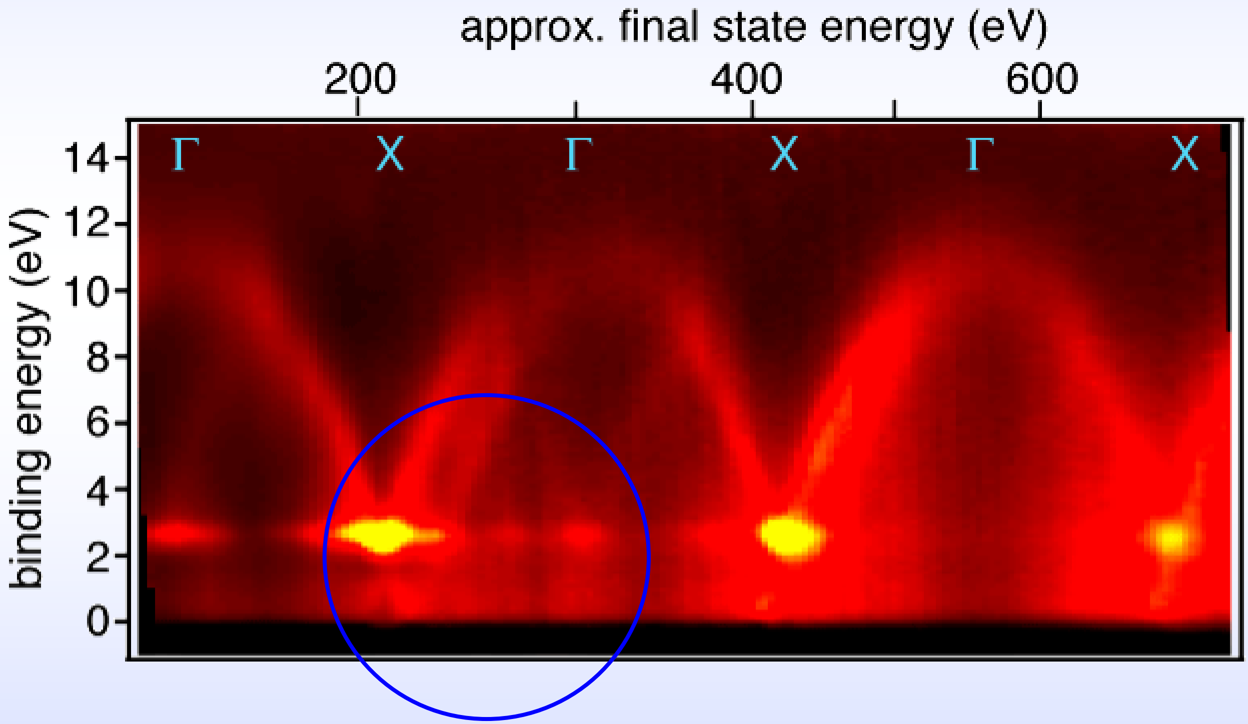


- 4*s* weight: anomalous ΔΣ>0
  3*d* weight: anomalous ΔΣ<0

- Mechanism: Exchange integral $-\sum_{\mathbf{q}} \int \phi^*_{\mathbf{k}}(\mathbf{r})\phi^*_{\mathbf{q}}(\mathbf{r})v(\mathbf{r}-\mathbf{r}')\phi_{\mathbf{q}}(\mathbf{r}')\phi_{\mathbf{k}}(\mathbf{r}')d\mathbf{r}d\mathbf{r}'$
couples to the valence *d*-electrons ⇒
more negative for *d*-states and less negative for *s*-states

High energies: Where do the final states become FE-like?

- Example: Normal-emission data on Al(100) *(Hoffman et al 2002)*



- non-FE multiband final states up to 400 eV

High energies: Soft-X-ray ARPES at Swiss Light Source

ADvanced RESonant Spectroscopies (ADRESS) Beamline :

- energy range 400-1800 eV
- resolution 35 meV @ 1 keV
- $3\text{x}10^{11}$ to $1\text{x}10^{13}$ photons/s/0.01%BW
- RIXS endstation:

- high-res spectrometer (70 meV @ 1 keV) by Politechnico di Milano
- rotating platform to study **q**-dependences

- ARPES endstation

- operation in spring 2008
- talk by L Patthey (Thursday, April 26)

Cooperations:

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R Claessen, Würzburg
P Blaha, Wien
E Krasovskii, Kiel
L Patthey, Swiss Light Source