Photoemission from Valence Bands, Dispersion and Fermi Surface Mapping

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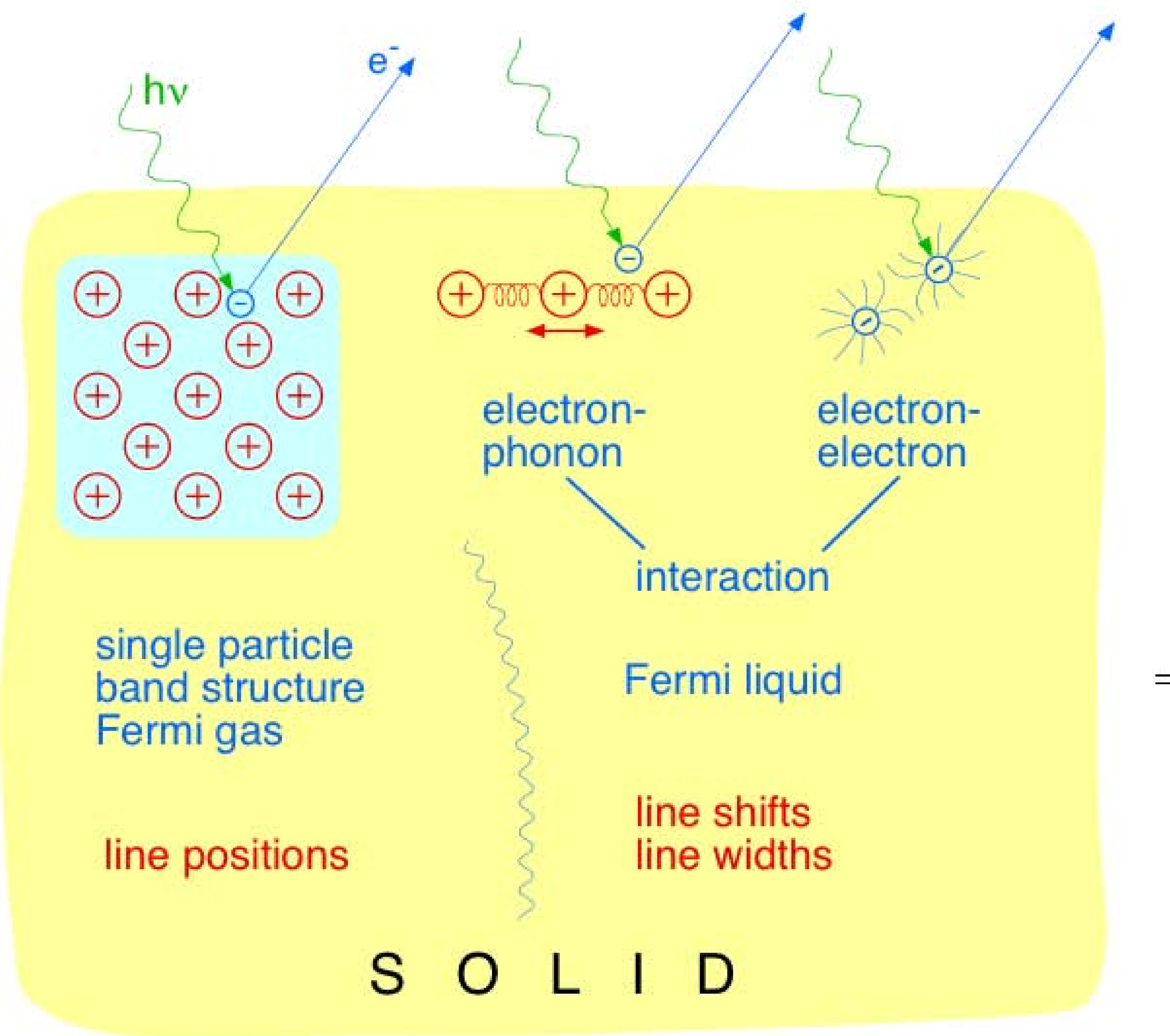
Lecture 2

- Many-Body Effects in Photoemission
- NiO: an Example for a Strongly Correlated System
- Theoretical Concepts: Sudden Approximation, Spectral Function
- Interpretation of Line Widths, Relation to Photoemission Experiments
- Energy Distribution Curves vs. Momentum Distribution Curves

Photoemission as a Solid State Spectroscopy



⇒ Understanding

electrical
magnetic
optical
superconducting
thermal

properties

Manybody Effects in Photoemission

So far: one-electron picture (Fermi gas !)

But: electrons interact with electrons
with phonons
with ...

Two types of phenomena:

- e⁻ - interaction in the initial state

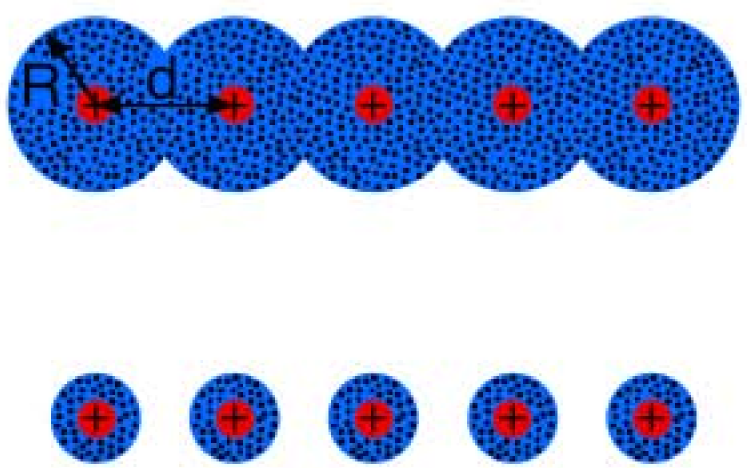
  → hot tropic in solid state physics (strongly correlated materials, Fermi liquids → non-Fermi liquids)

- e⁻ - interaction during the photoemission process

  → satellites, line shapes, line positions

  important for interpretation of spectra !

Two extreme types of states

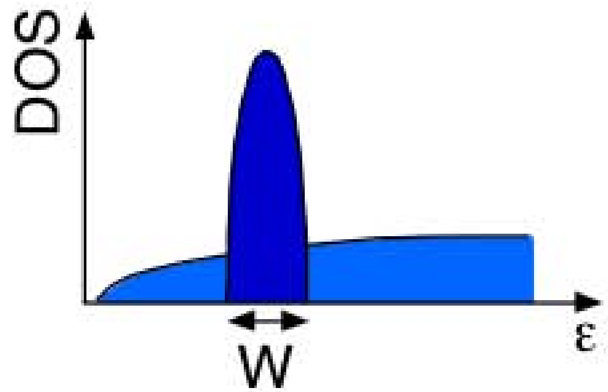


R ≈ d : large overlap (s, p)
large dispersion ε (k)
nearly free electrons

→ one-electron picture !

R << d : little overlap (d, f)
little dispersion
tight binding

→ correlation effects !

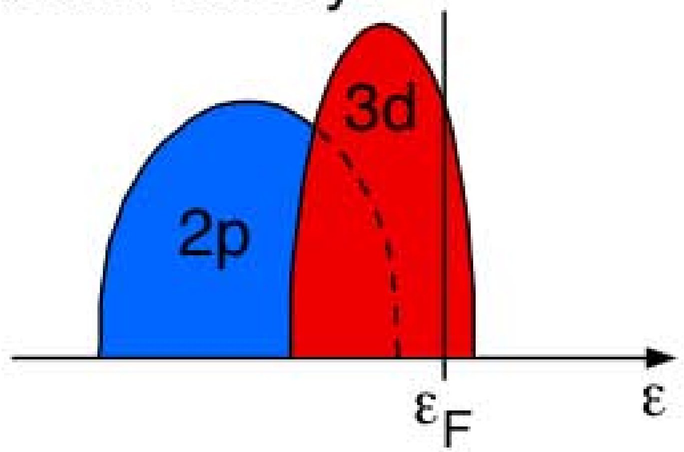


NiO: an Example for a Strongly Correlated System

NiO

3d^8 2p^6

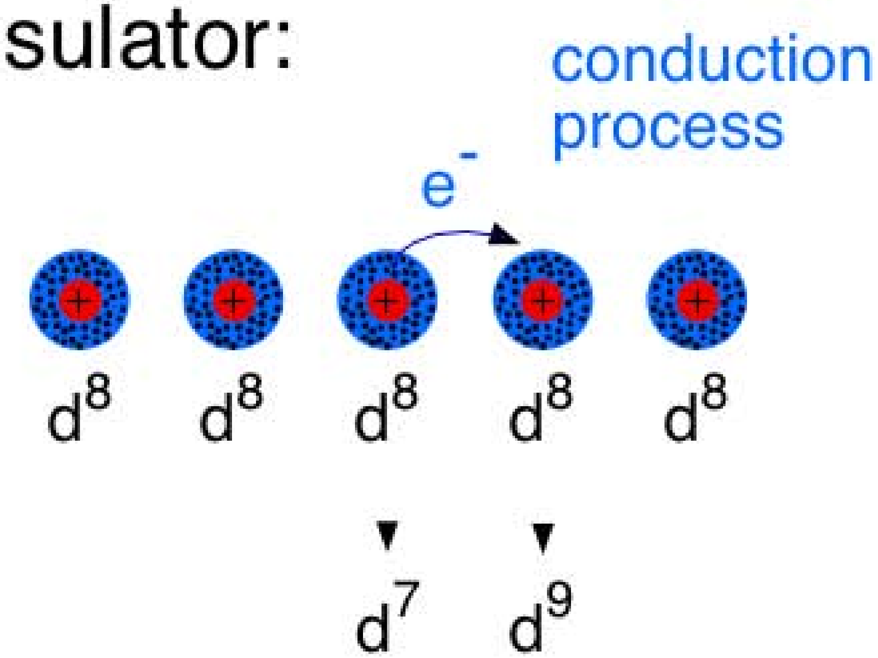
band theory:



→ predicts a metal

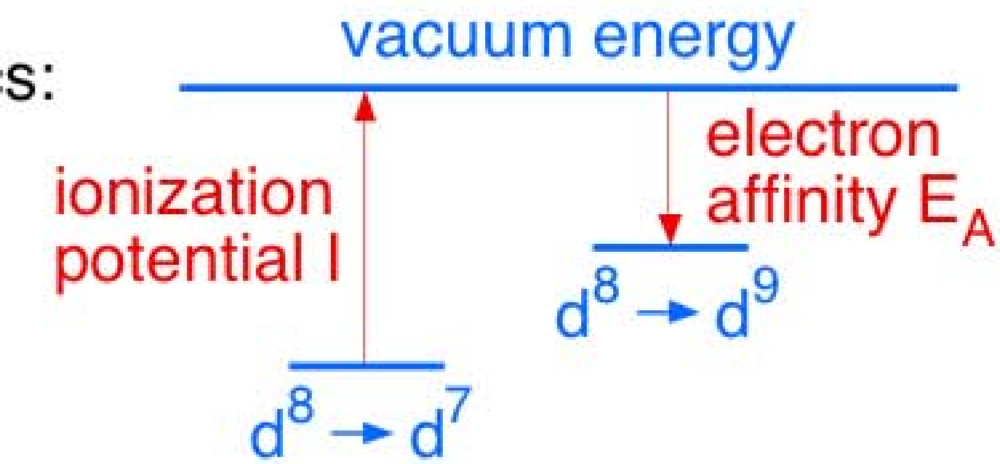
But: experiment shows insulating gap (4 eV) !

Mott Insulator:



"reaction": $2\,d^8 \rightarrow d^7 + d^9 - U$

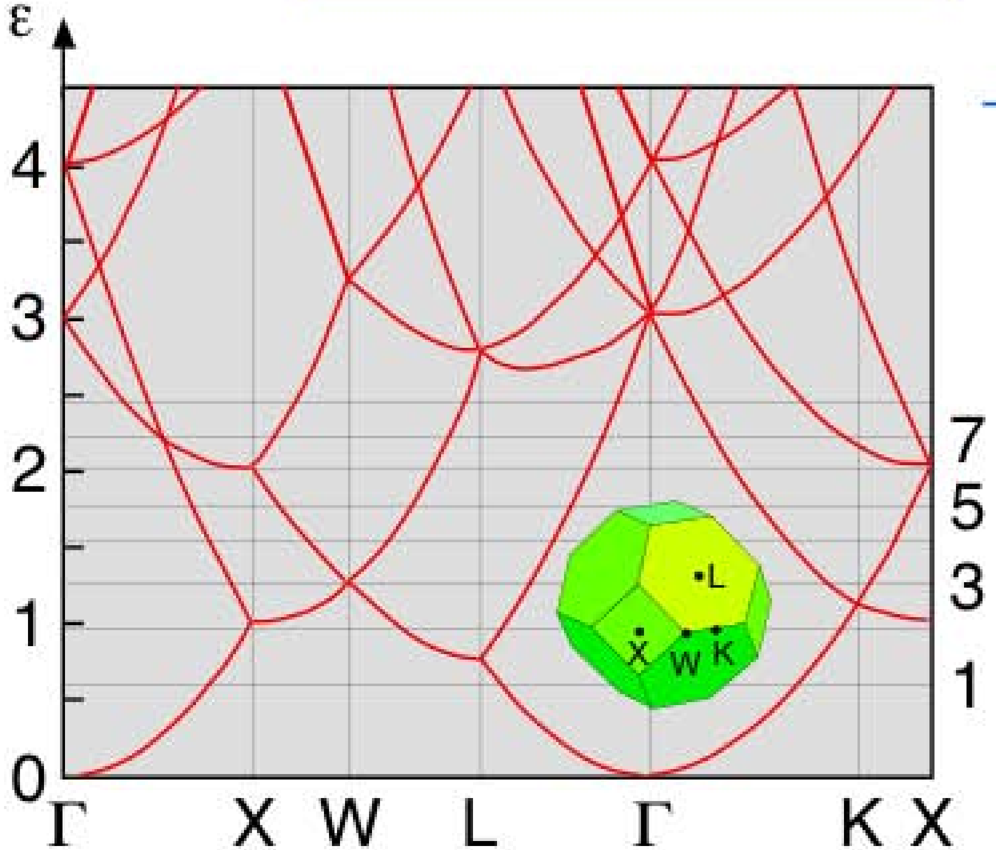
energetics:



$$U = I - E_A$$

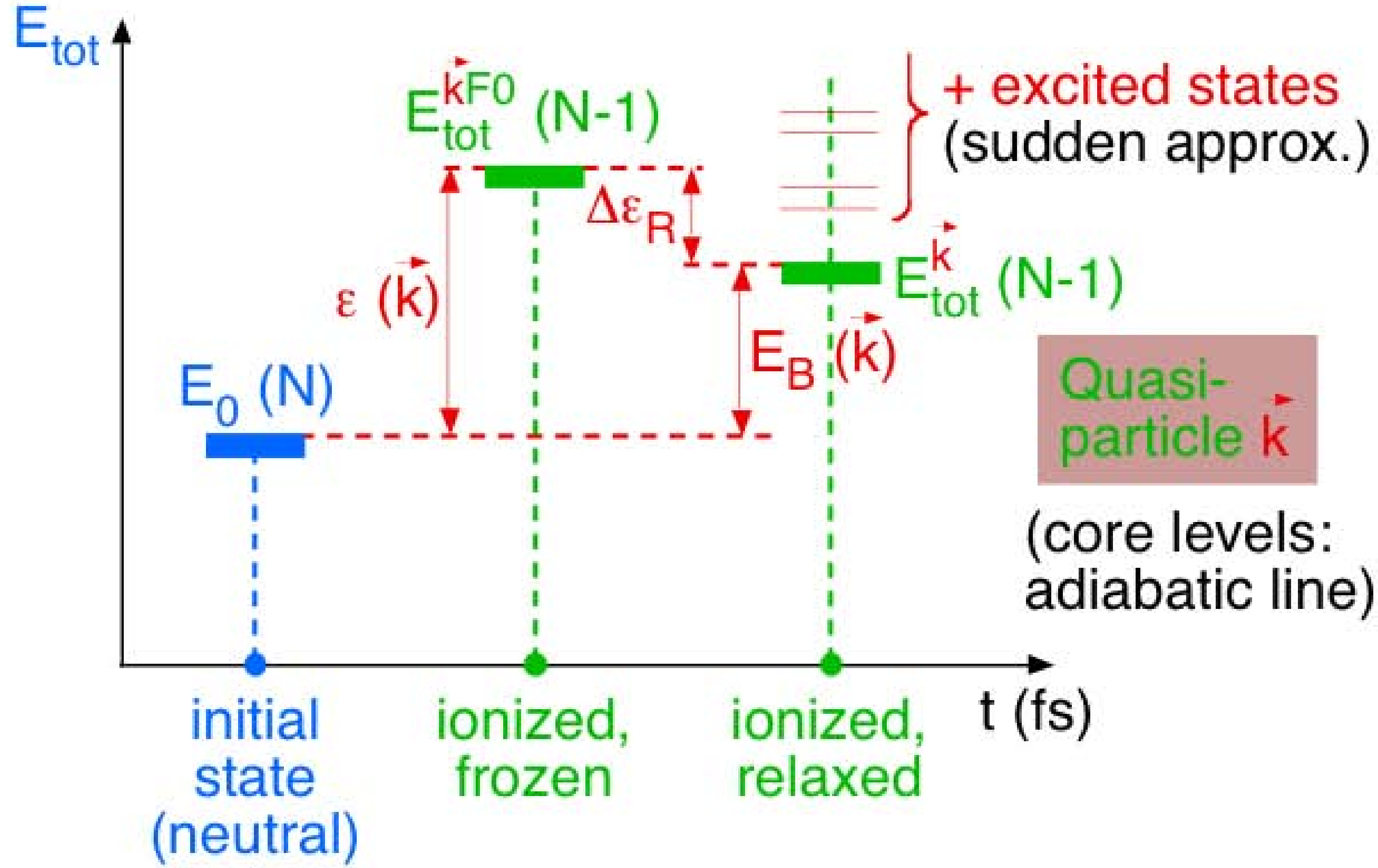
free ions : U = 18 eV
solid state: reduced (O charge transfer)

Theoretical Concepts

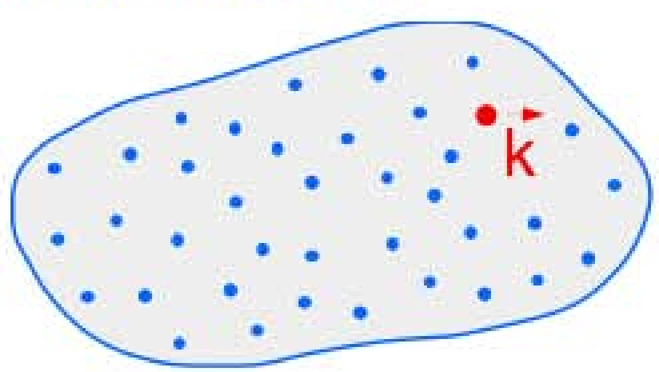


Theory: → energy eigenvalues

Total-Energy Diagram:



Koopman's Theorem:



N electrons

$$\varepsilon(\vec{k}) = E_{tot}^{\vec{k},SCF}(N) - E_{tot}^{\vec{k},F0}(N-1) < 0$$

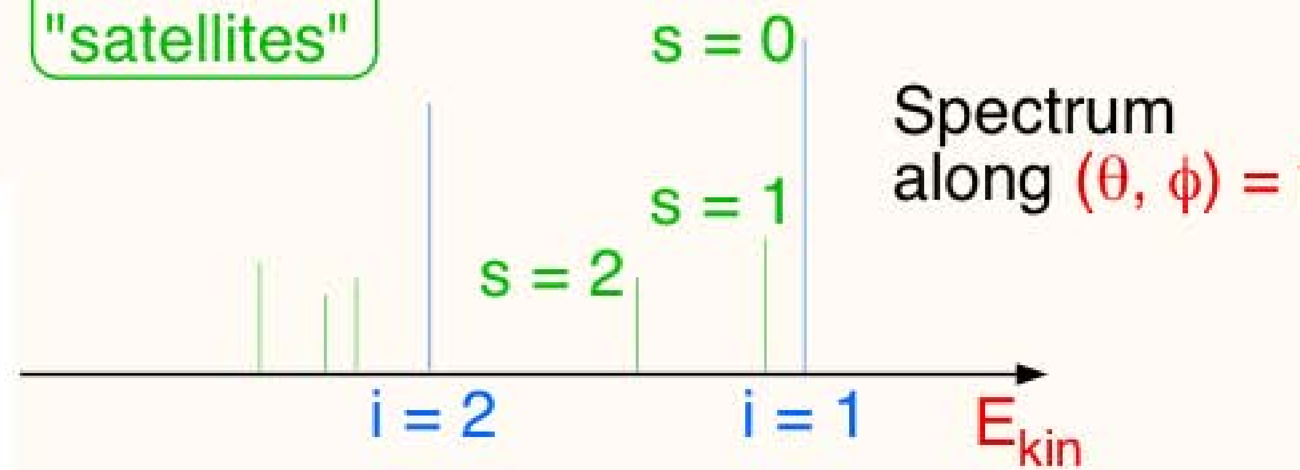
"frozen orbitals"

The Photocurrent I

Atoms, Molecules:

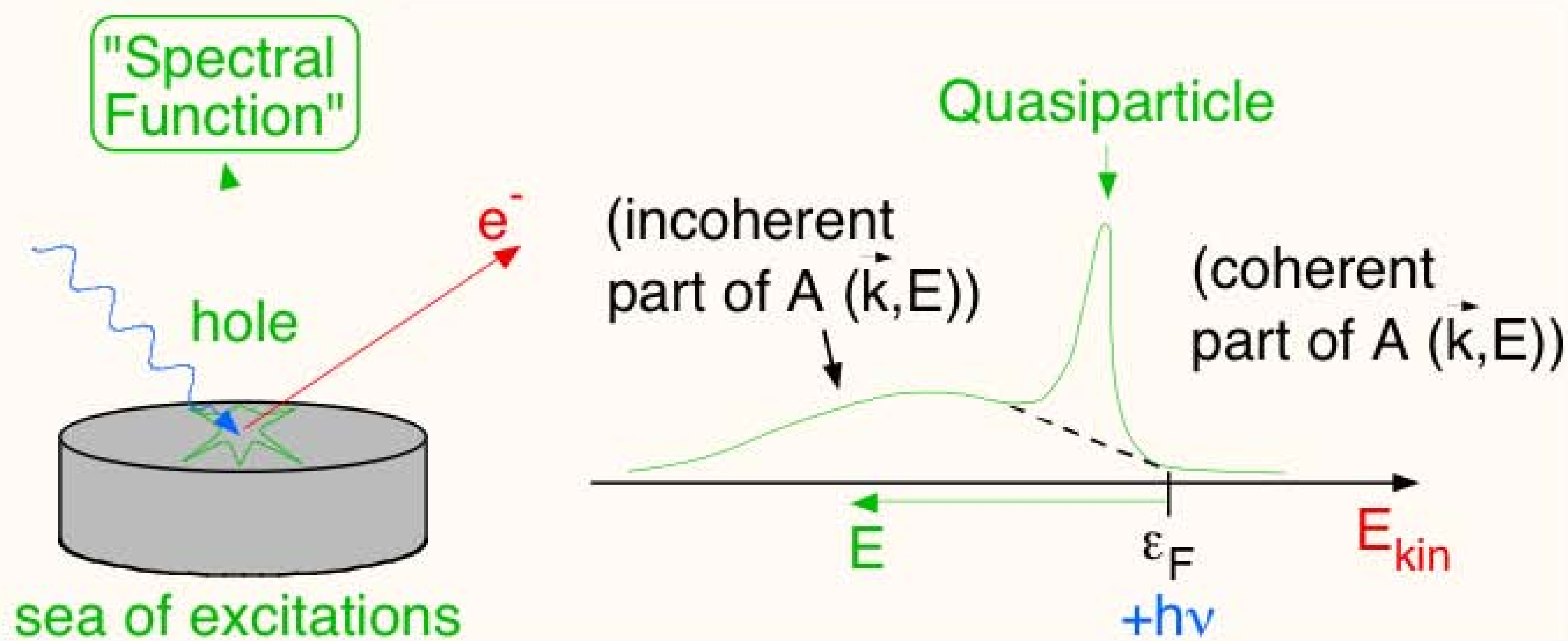
$$I \sim \sum_{f,i,k} | \langle \phi_{f,E_{kin}} | \vec{r}_k \cdot \vec{\varepsilon} | \phi_{i,k}(\vec{r}_k) \rangle |^2 \sum_s | c_s^{i,k} |^2 \, \delta\left(E_{kin} + E_s(N-1) - E_o(N) - h\nu\right)$$

- f: various final states (angles, ...)
- i: various orbitals
- k: various atoms
- $c_s^{i,k}$: excited final states "satellites"

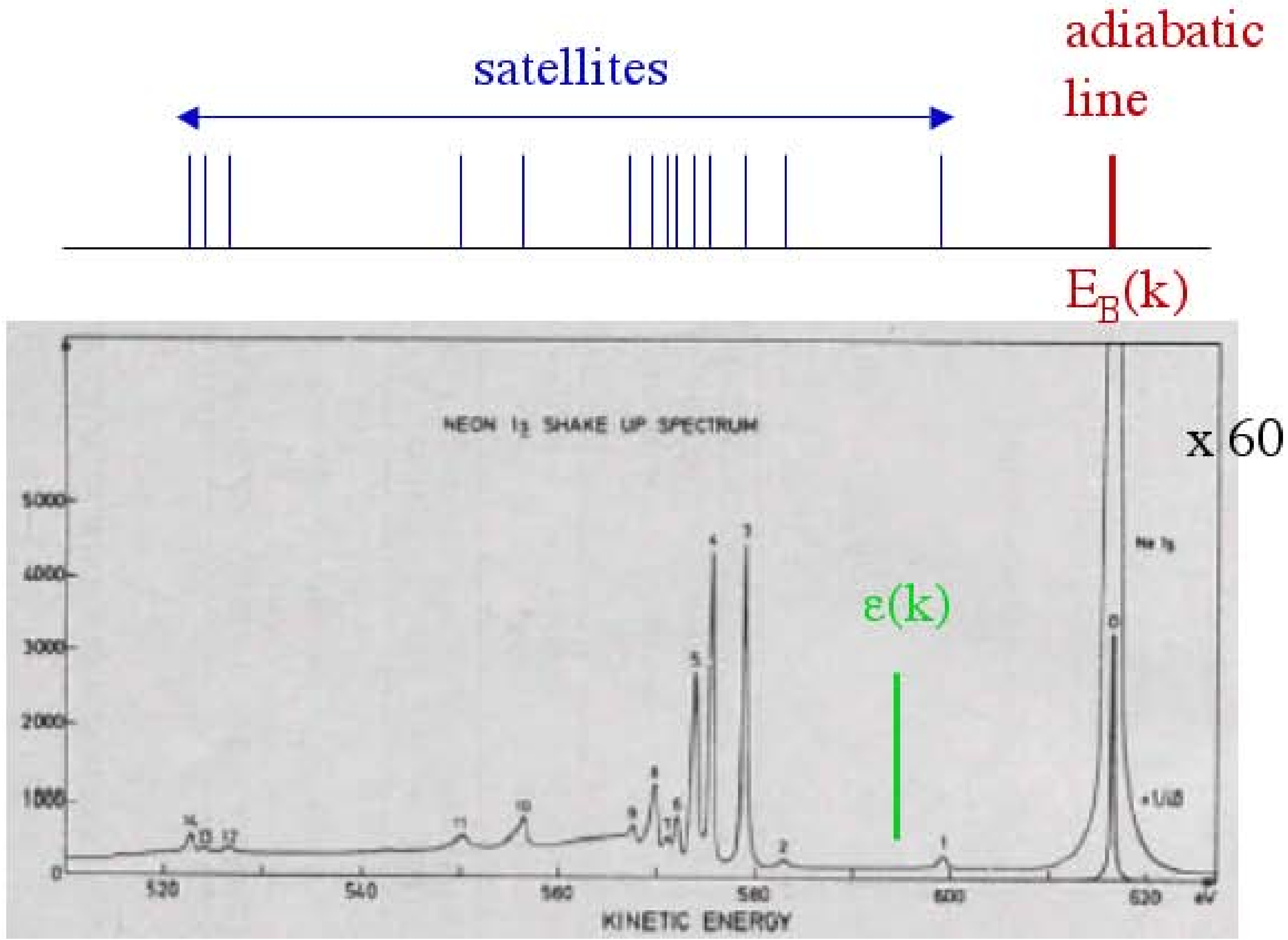


Solids:

$$I \sim \sum_{f,i,\vec{k}} | \langle \phi_{f,E_{kin}} | \vec{r}_k \cdot \vec{\varepsilon} | \phi_{i,\vec{k}} \rangle |^2 \, A(\vec{k}, E)$$



The Photocurrent from Neon 1s



… and also Coupling to Lattice Degrees of Freedom - e.g. Adsorbate Vibrations

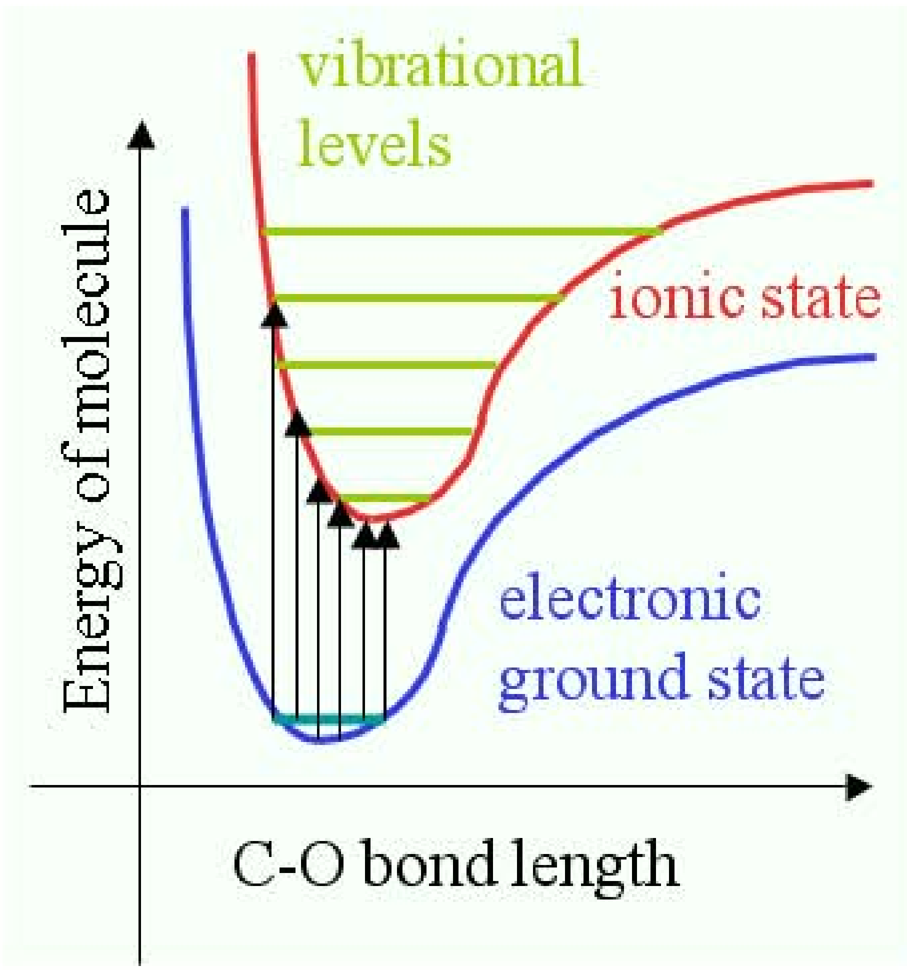
$$E_{kin} = h\nu - (E_{final} - E_{initial})$$

Photoelectron

Electronic or vibronic excitations

Ground state

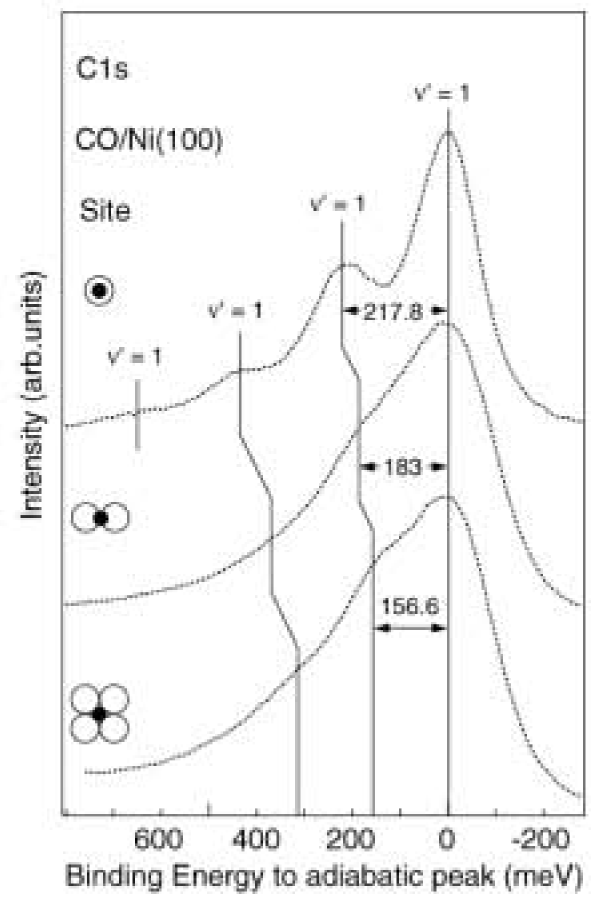
(chemical shift)



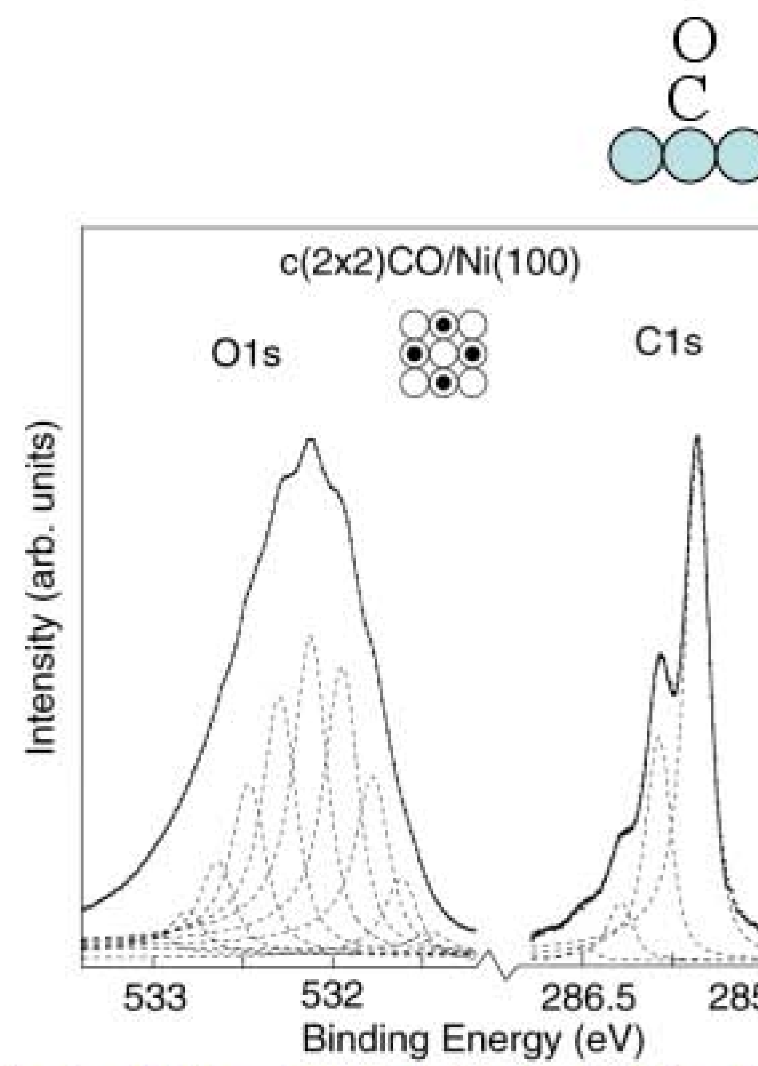
=> Franck-Condon Principle

E_{final} depends on

… bonding site



… where the photohole sits



Föhlisch et. al Phys. Rev. Lett. 81 (1998) 1730

Spectral Functions

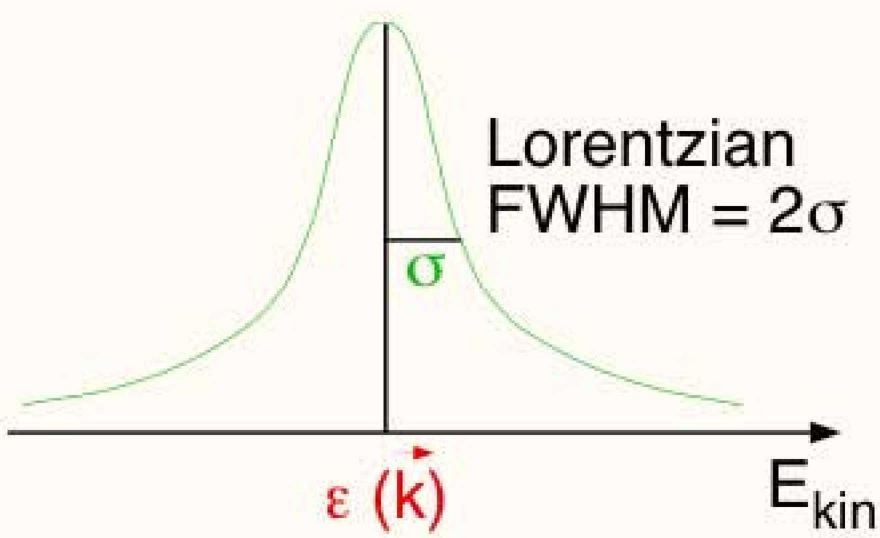
... can be calculated by Greens Function Formalism:

$$A(\vec{k},E) = \frac{1}{\pi} \Im m \{G_+(\vec{k},E)\}$$

independent particles (no interactions):

$$G_+^o(\vec{k},E) = \frac{1}{E - \varepsilon(\vec{k}) - i\sigma}$$

$$A(\vec{k},E) = \frac{1}{\pi} \frac{\sigma}{(E - \varepsilon(\vec{k}))^2 + \sigma^2}$$



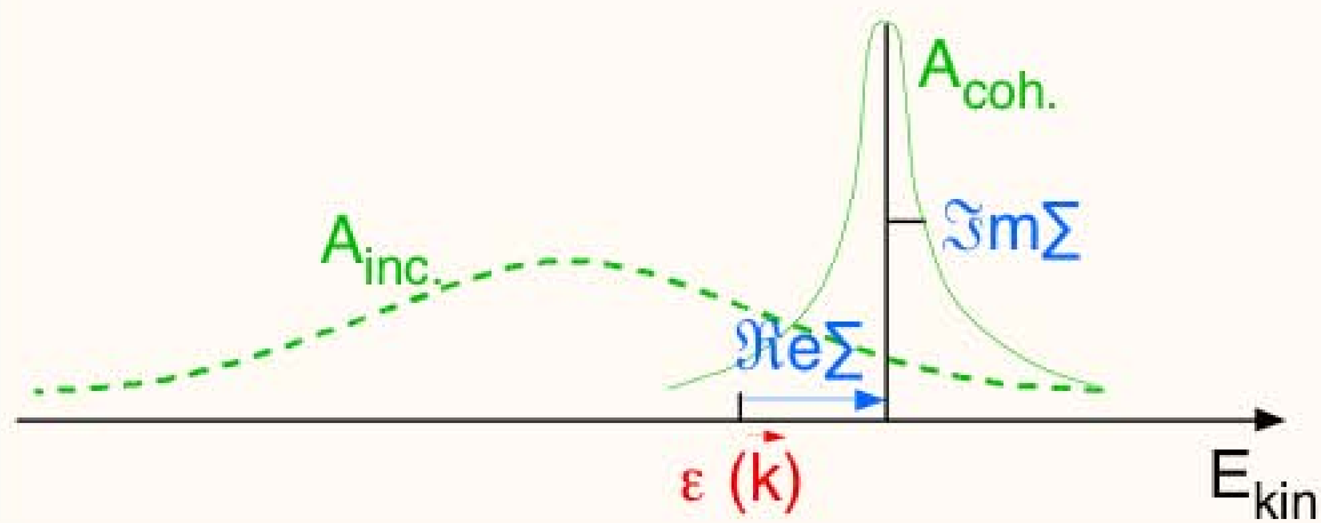
with interactions:

$$G_+(\vec{k},E) = \frac{1}{E - \varepsilon(\vec{k}) - \Sigma(\vec{k},E)}$$

Self energy

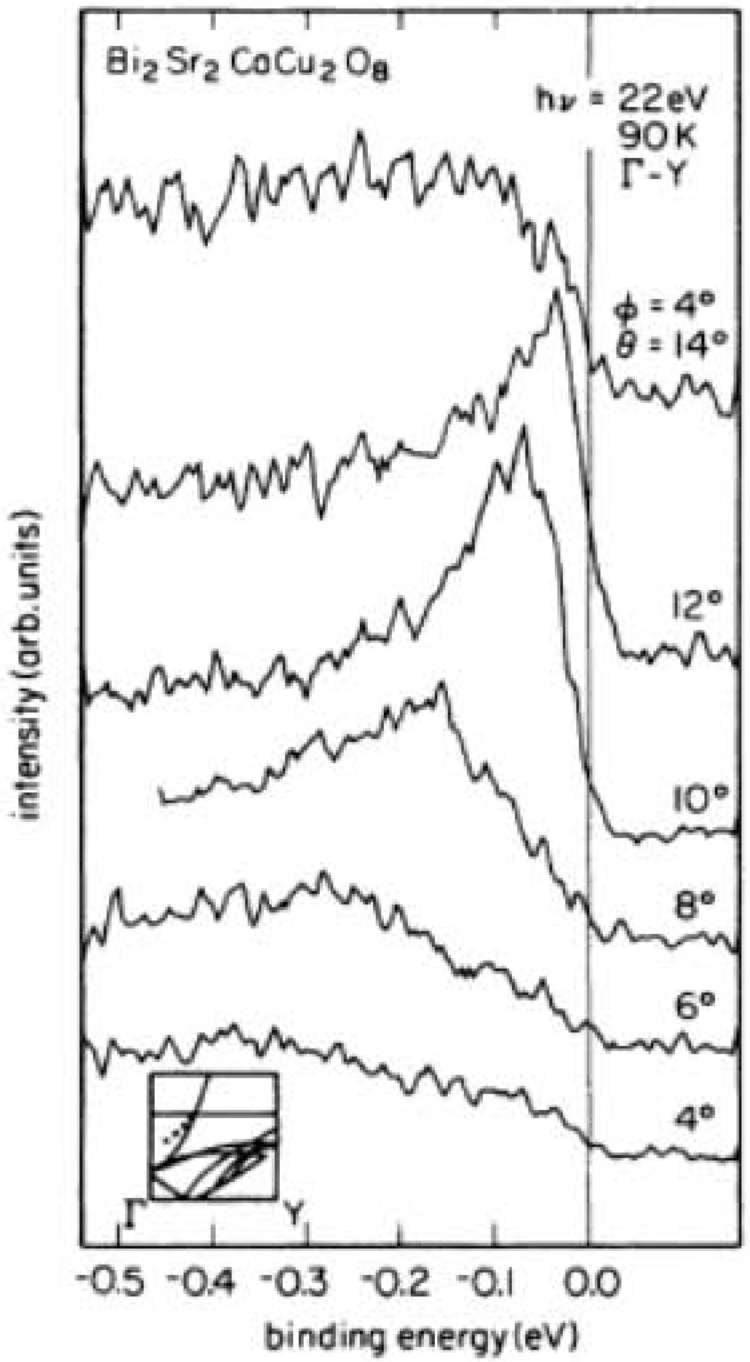
=> need Hamiltonian of system !

$$A(\vec{k},E) \sim \frac{\Im m\{\Sigma(\vec{k},E)\}}{[E - \varepsilon(\vec{k}) - \Re e\Sigma]^2 + [\Im m\Sigma]^2}$$



Experimental Situation on Line Widths

Fermi-liquid theory: Line width $\Delta E \sim (E-E_F)^2$



Experiment on HTC material:

$\Delta E \sim (E-E_F)$

=> non-Fermi-liquid ?

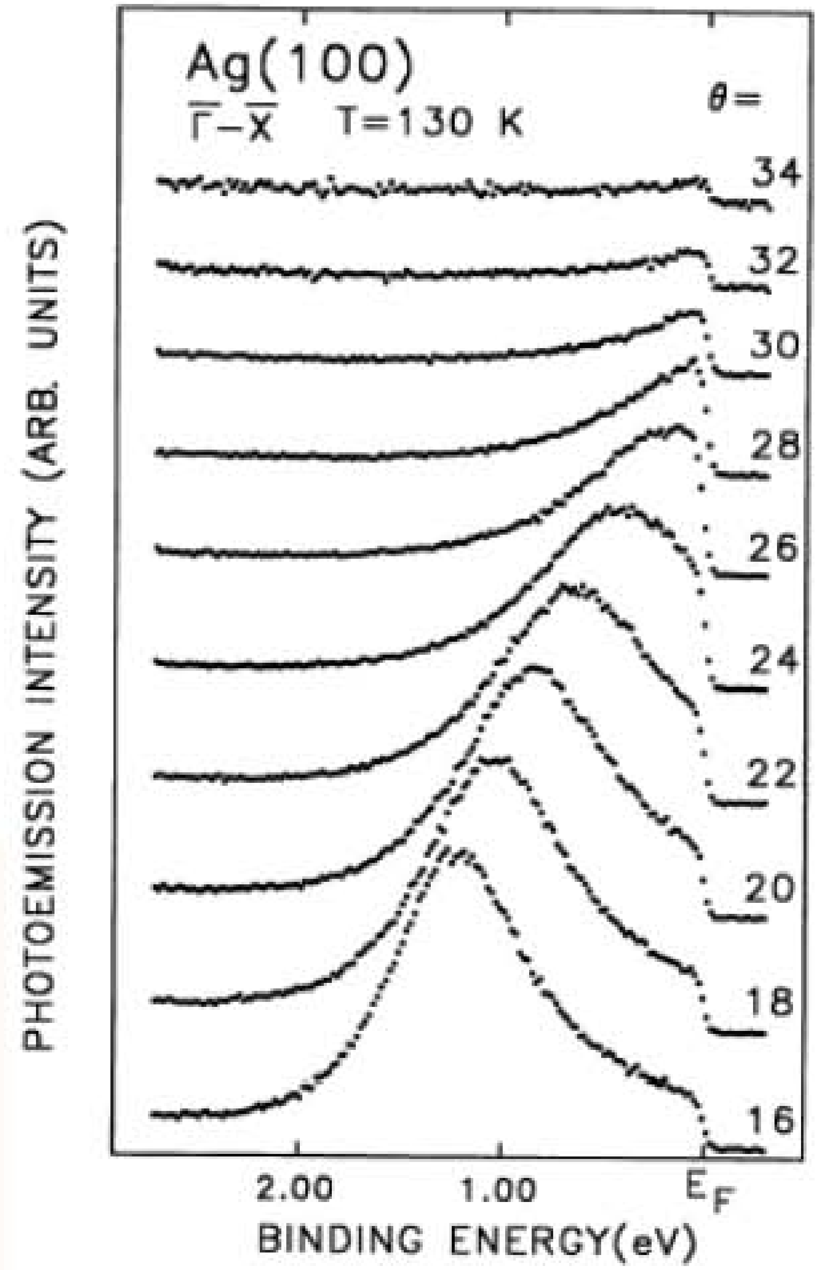
C. G. Olson et al., PRB 42, 381 (1990)

… but:

Silver (a much simpler material) even shows a growing line width towards EF !

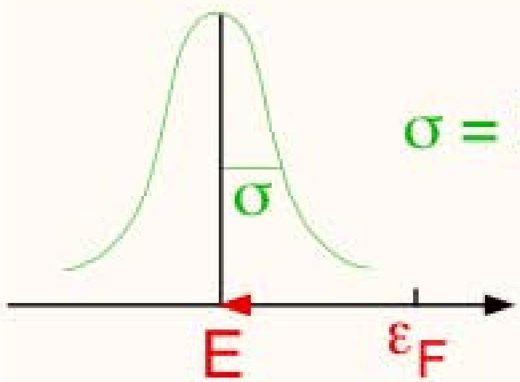
?

Y. Hwu et al., PRB 45, 5438 (1992)



⇒One needs to know the technique well !

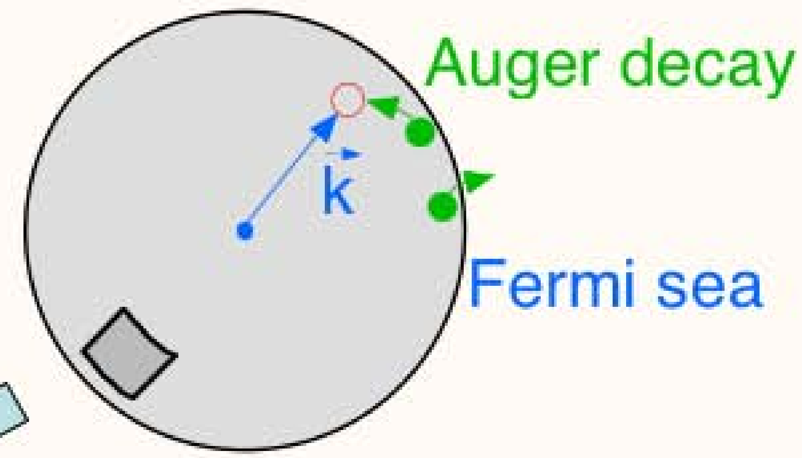
Interpretation of Line Widths



$$\sigma = \Im m \Sigma(\vec{k}, E) \sim \frac{\hbar}{\Delta t}$$

Q.P. Life time

dominant hole decay processes:

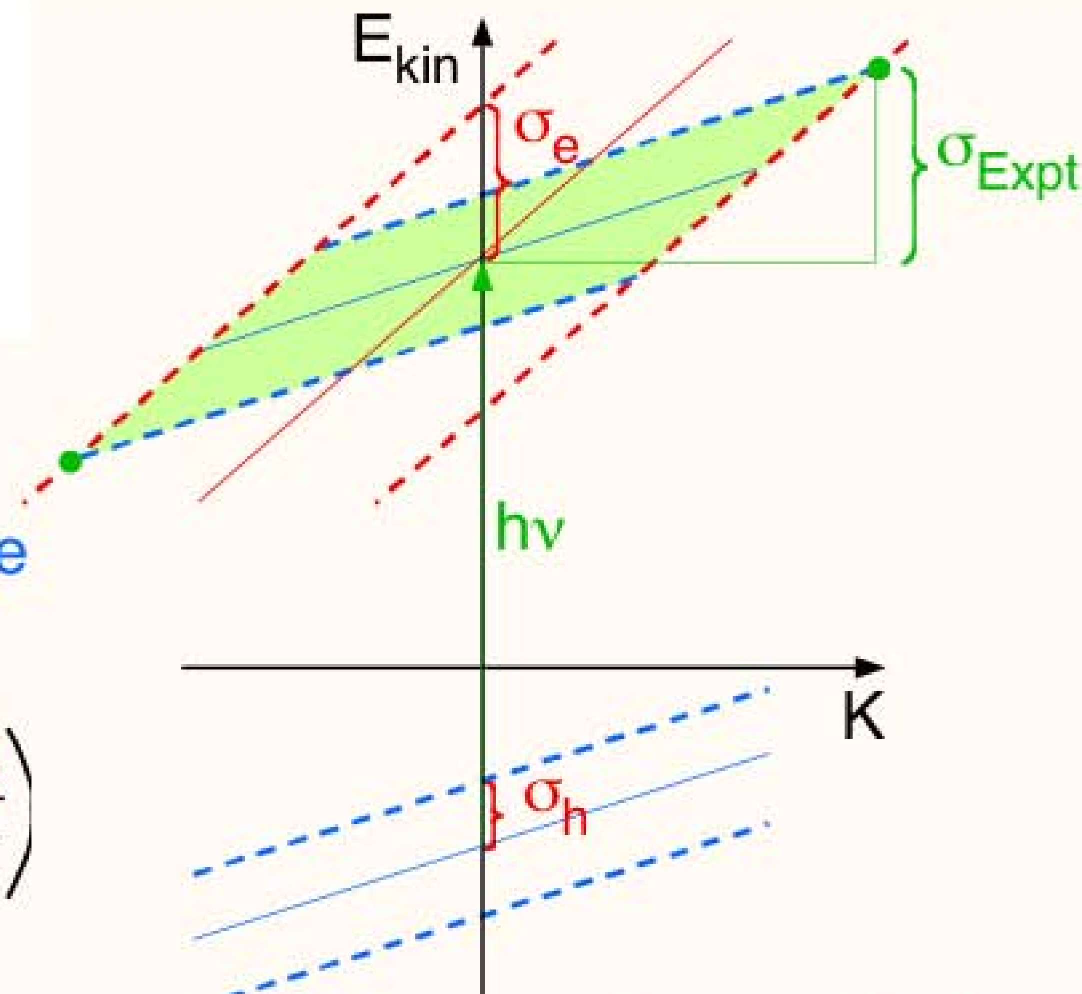


Fermi liquid theory

→ $\Delta t \sim \frac{1}{E^2}$

→ $\boxed{\sigma \sim E^2}$

Caution !



(we measure E_{kin}, not k !)

Photoemission measures
- hole lifetime $\frac{1}{\sigma_h}$
- and photoelectron lifetime

normal emission:

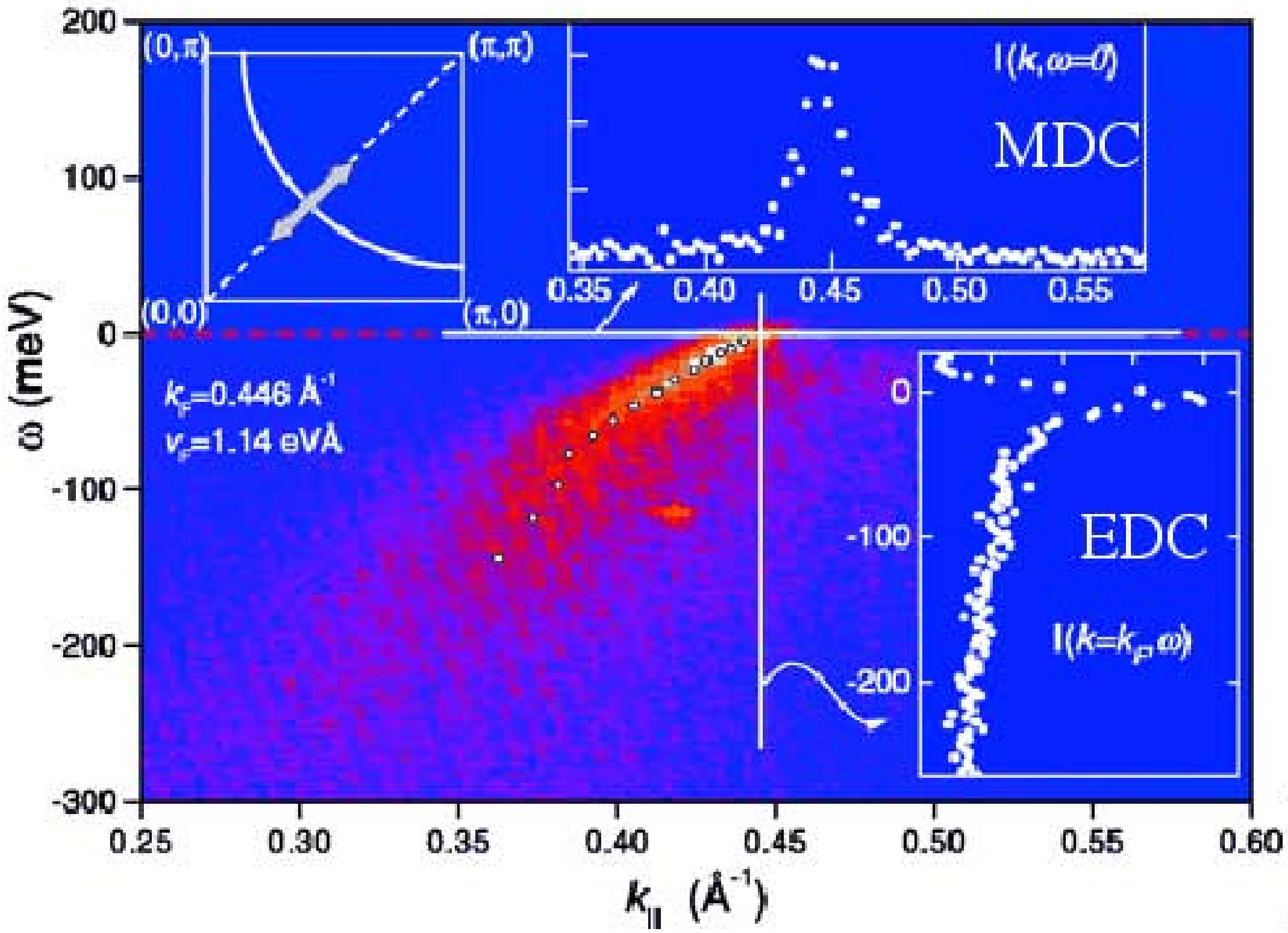
(P.Thiry, 1981)

$$\sigma_{Expt.} = \frac{v_e^\perp \cdot v_h^\perp}{v_e^\perp - v_h^\perp} \cdot \left(\frac{\sigma_h}{v_h^\perp} + \frac{\sigma_e}{v_e^\perp} \right)$$

group velocities $\frac{\partial \varepsilon}{\partial k_\perp}$

Energy Distribution Curves (EDCs) and Momentum Distribution Curves (MDCs)

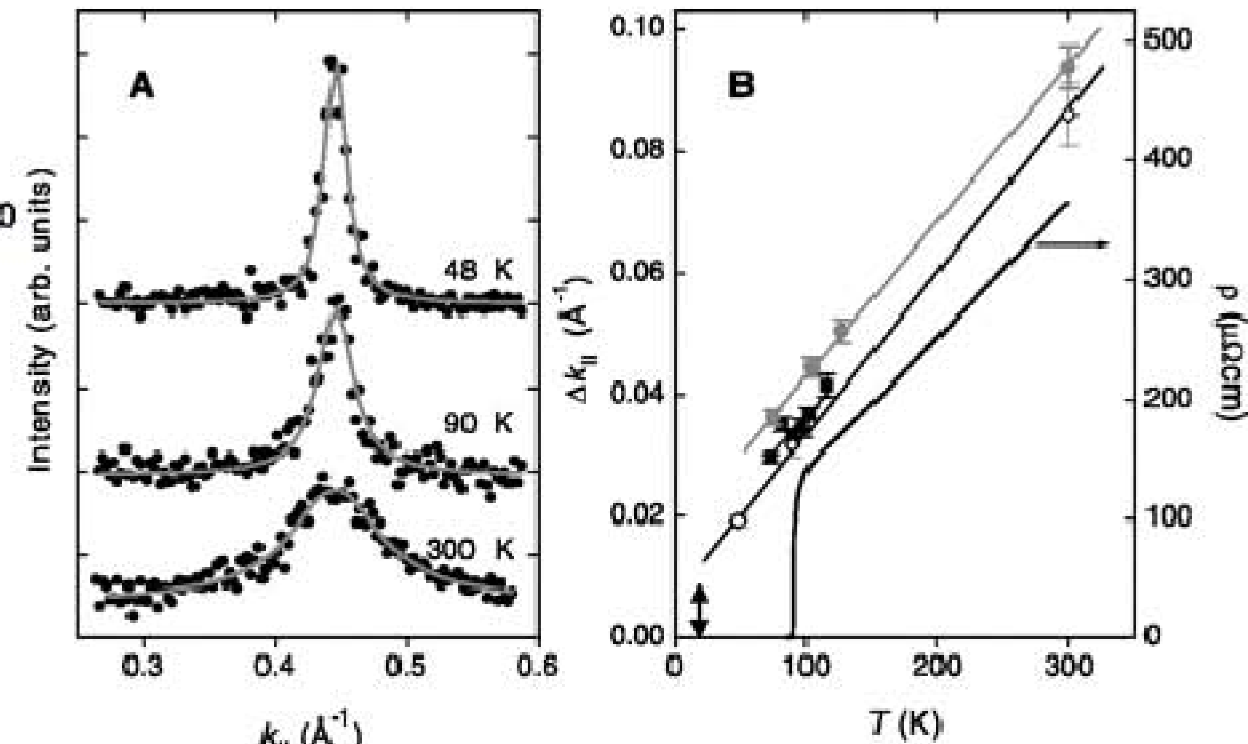
Example: Dispersion Plot from BSCCO High Temperature Superconductor



Line Width Δk in MDCs is related to the mean free path Λ of the electrons near E_F and thus to the scattering rate:

$$\hbar v_k \Delta k = \frac{\hbar v_k}{\Lambda} \approx | 2\Im\Sigma(\vec{k},\omega) |$$

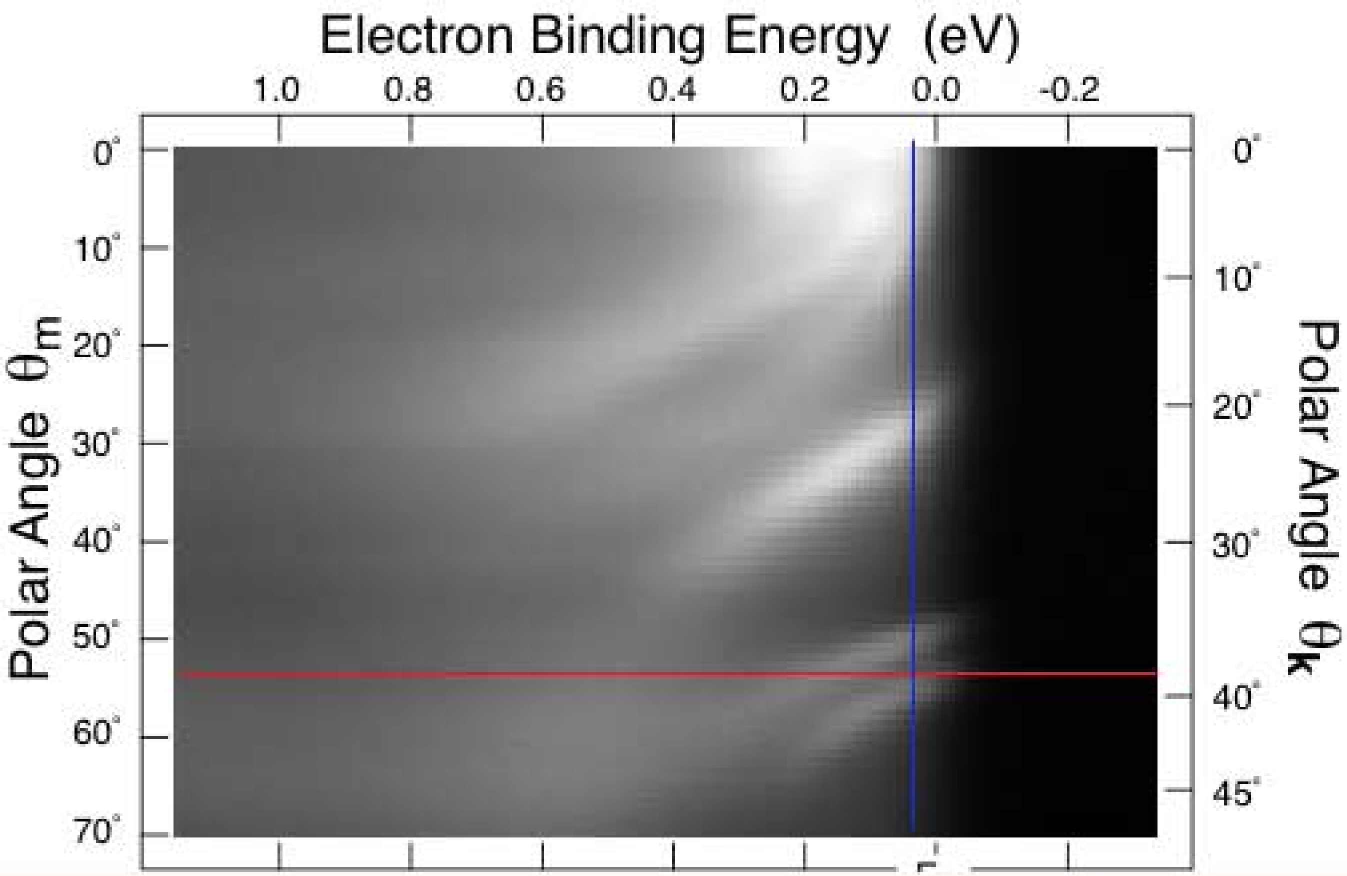
(v_k is the electron group velocity)

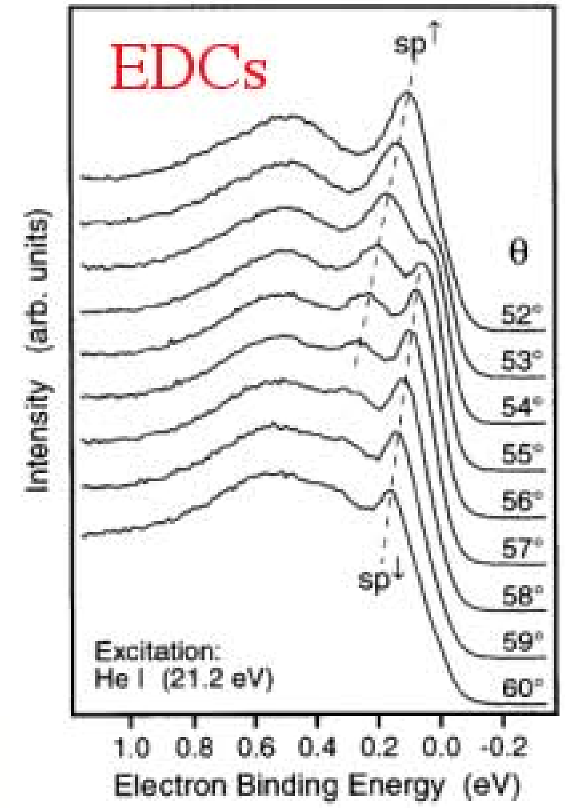


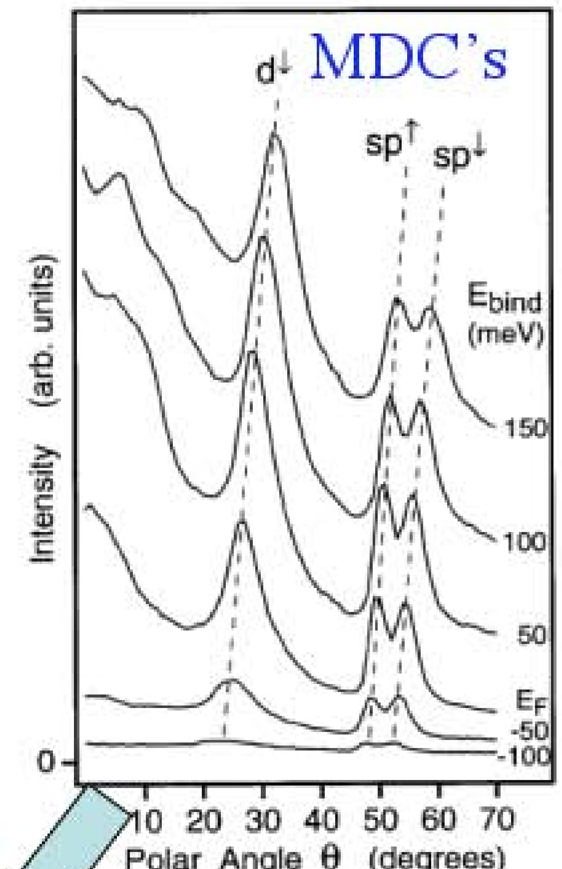
⇒ Connection between photoemission data and sample resistivity

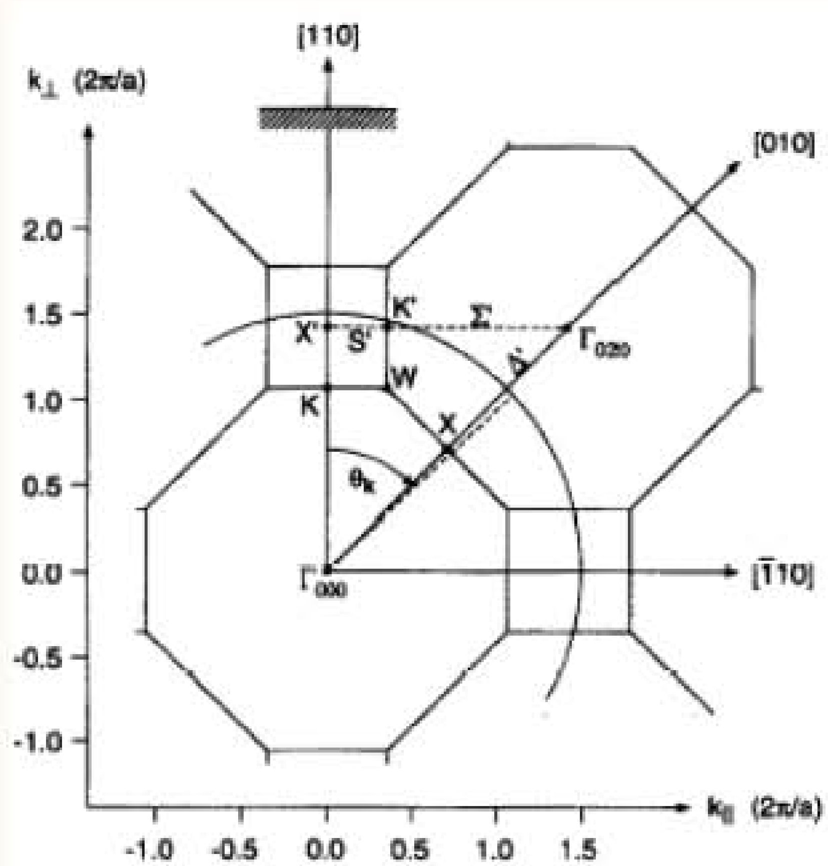
T. Valla et al. Science 285, 2110 (1999)

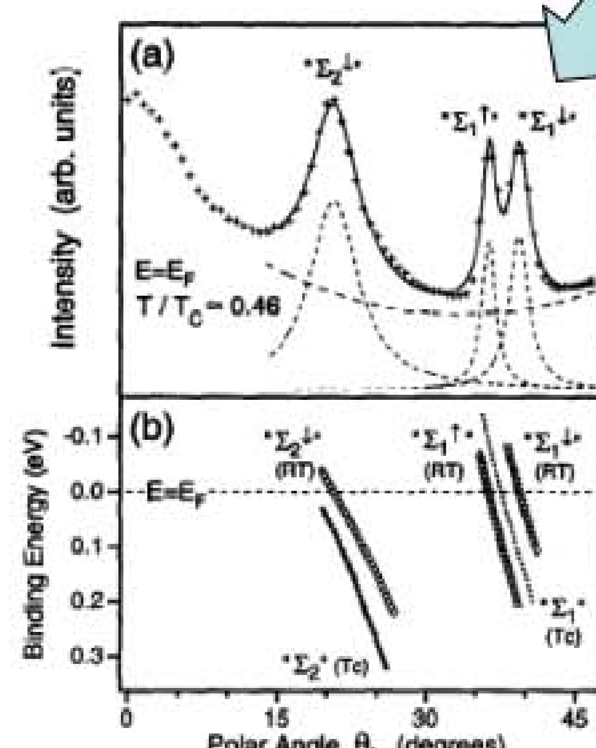
Extracting Energy Dispersion











… often more accurately from MDCs ! (simpler line shapes)