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ICTP 40th Anniversary

SCHOOL ON SYNCHROTRON RADIATION AND APPLICATIONS
In memory of J.C. Fuggle & L. Fonda

19 April - 21 May 2004

Miramare - Trieste, Italy

1561/18

Photoemission as a Solid State Spectroscopy: Many-Body Effects, Applications to Complex Materials

Juerg Osterwalder

Photoemission as a Solid State Spectroscopy: Many-Body Effects, Applications to Complex Materials

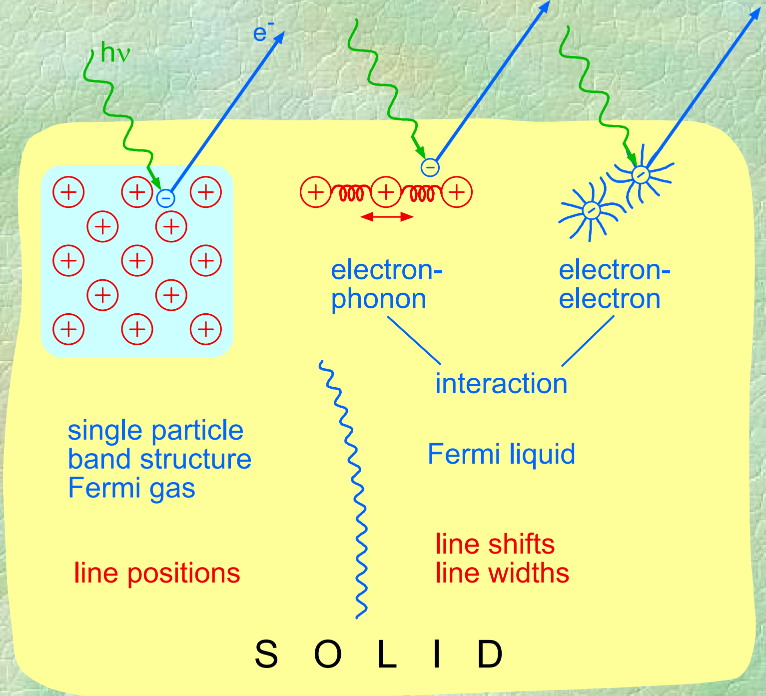
Jürg Osterwalder
Physik-Institut, Universität Zürich, Winterthurerstr. 190,
CH-8057 Zürich, Switzerland - osterwal@physik.unizh.ch
http://www.physik.unizh.ch/groups/grouposterwalder/

Lecture 4

- 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

Solid State Spectroscopy

Photoemission as a



⇒ 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

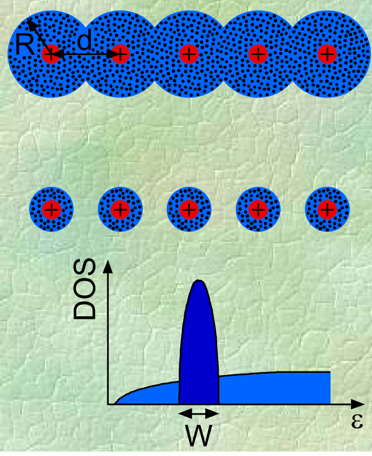
  → hot tropic in solid state physics
  (strongly correlated materials,
  Fermi liquids → non-Fermi liquids)

- 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 !

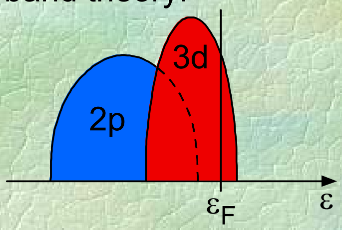
R << d : little overlap (d, f)
little dispersion
tight binding
→ correlation effects !

NiO: an Example for a Strongly Correlated System

NiO

$3d^8$ $2p^6$

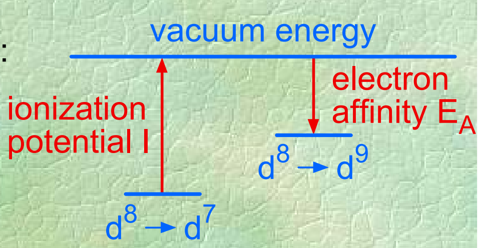
band theory:



→ predicts a metal

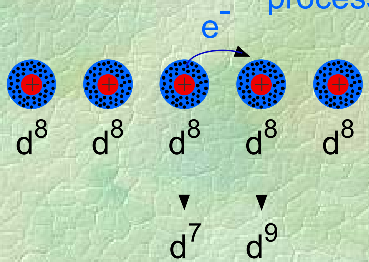
But: experiment shows insulating gap (4 eV) !

energetics:



Mott Insulator:

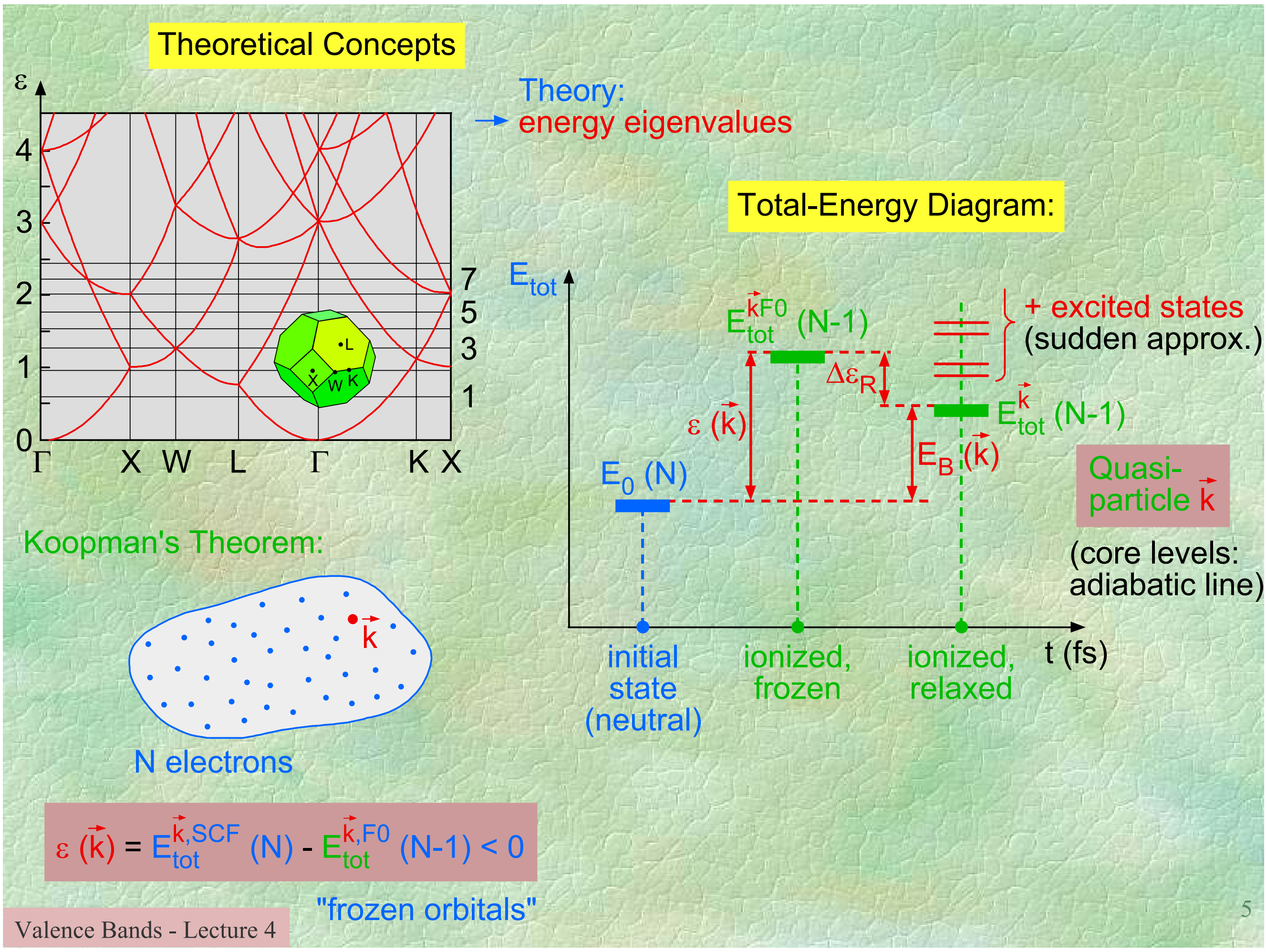
conduction process

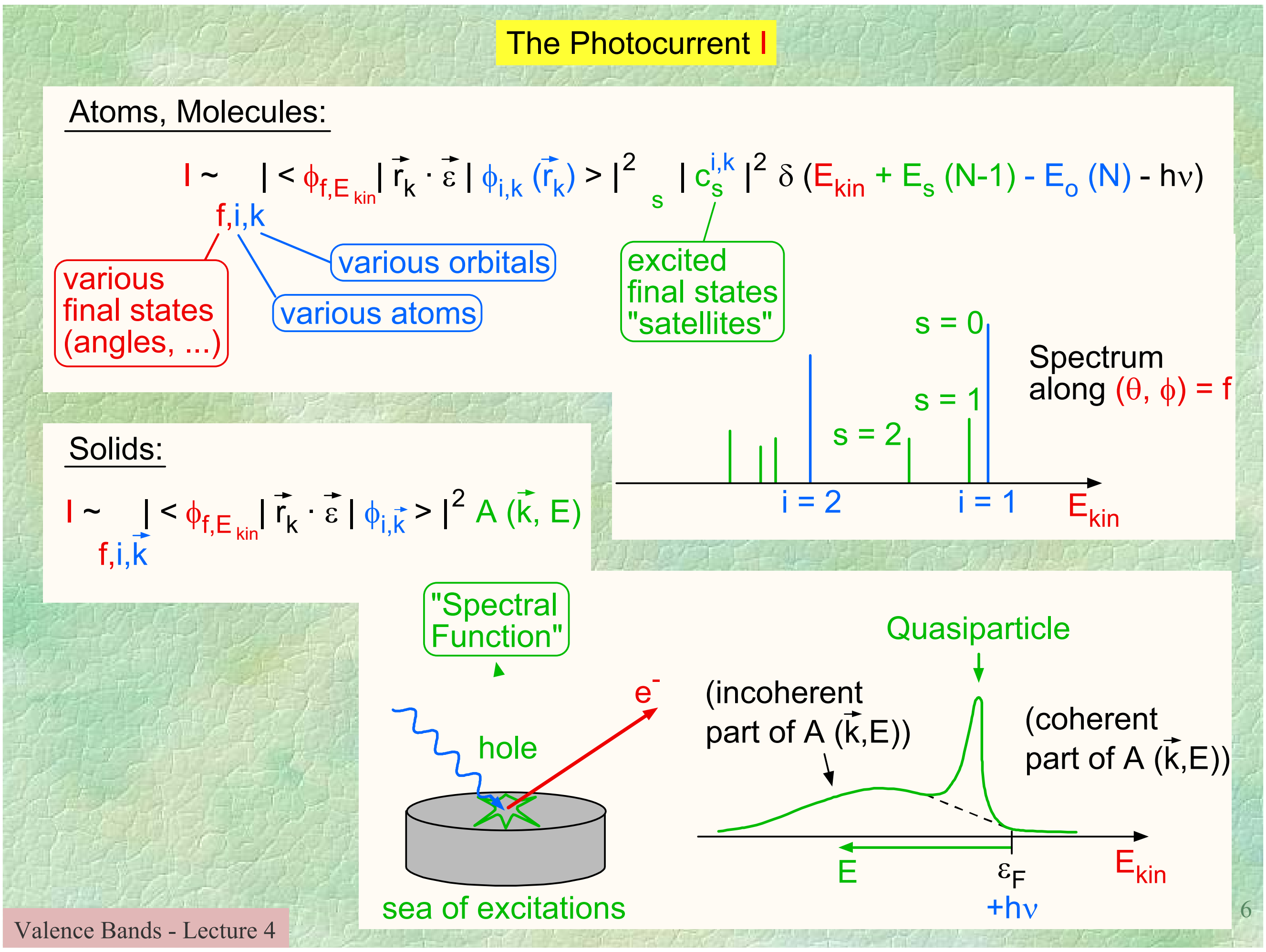


$U = I - E_A$

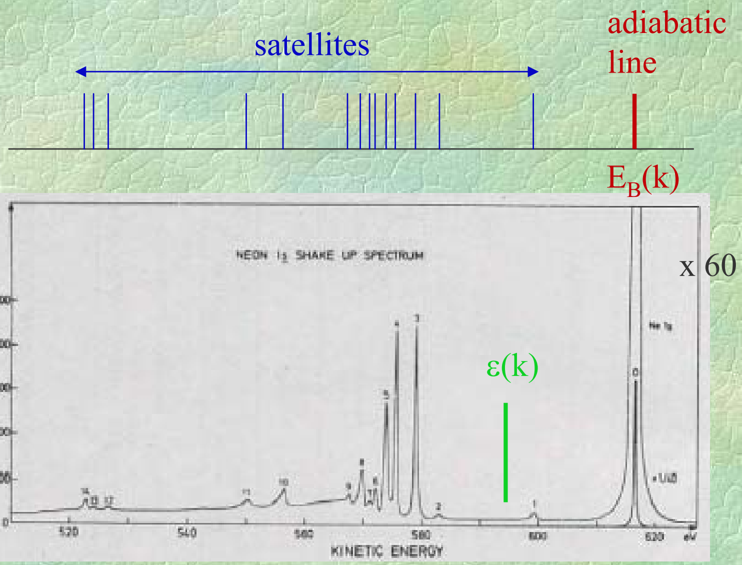
"reaction": $2\,d^8 \rightarrow d^7 + d^9 - U$

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


Theoretical Concepts
ε
0
1
2
3
4
Γ X W L Γ K X
7
5
3
1
L
X W K
Theory:
energy eigenvalues
Total-Energy Diagram:
E_tot
E_tot^kF0 (N-1)
Δε_R
ε (k)
E_0 (N)
+ excited states
(sudden approx.)
E_tot^k (N-1)
E_B (k)
Quasi-particle k
(core levels:
adiabatic line)
initial state (neutral)
ionized, frozen
ionized, relaxed
t (fs)
Koopman's Theorem:
k
N electrons
ε (k) = E_tot^k,SCF (N) - E_tot^k,F0 (N-1) < 0
"frozen orbitals"


The Photocurrent I
Atoms, Molecules:
I ~ Σ_f,i,k | < φ_f,E_kin | r_k · ε | φ_i,k (r_k) > |² Σ_s | c_s^i,k |² δ (E_kin + E_s (N-1) - E_o (N) - hν)
various final states (angles, ...)
various orbitals
various atoms
excited final states "satellites"
s = 0
s = 1
s = 2
Spectrum along (θ, φ) = f
i = 2
i = 1
E_kin
Solids:
I ~ Σ_f,i,k | < φ_f,E_kin | r_k · ε | φ_i,k > |² A (k, E)
"Spectral Function"
e⁻
hole
sea of excitations
Quasiparticle
(incoherent part of A (k,E))
(coherent part of A (k,E))
E
ε_F
+hν
E_kin

The Photocurrent from Neon 1s

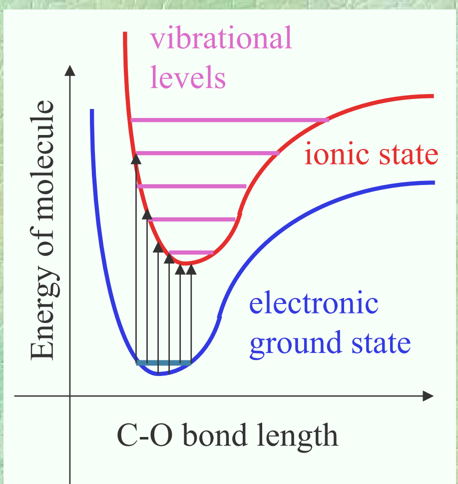


… and also Coupling to Lattice Degrees of Freedom - e.g. Adsorbate Vibrations

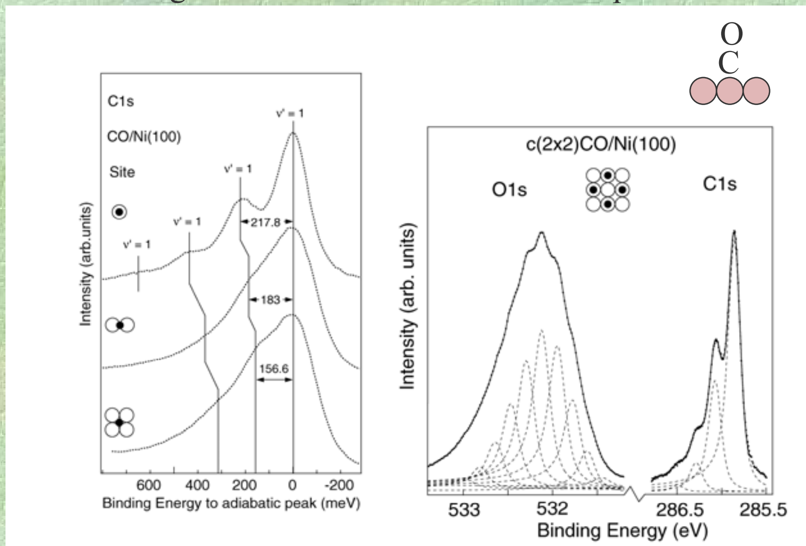
$$E_{kin} = h\nu - (E_{final} - E_{initial})$$

Photoelectron | Electronic or vibronic excitations | Ground state (chemical shift)

E_{final} depends on
… bonding site … where the photohole sits



=> Franck-Condon Principle



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

Spectral Functions

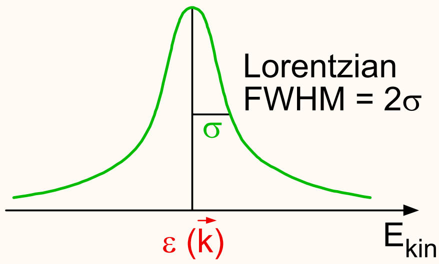
... can be calculated by Greens Function Formalism:

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

independent particles (no interactions):

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

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

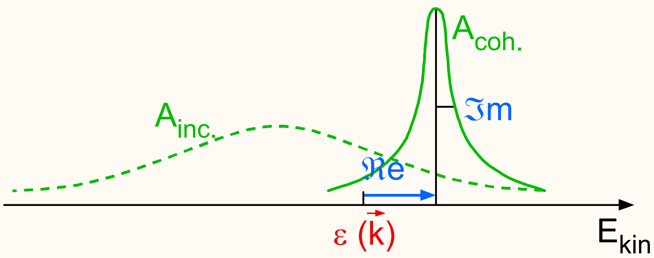


with interactions:

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

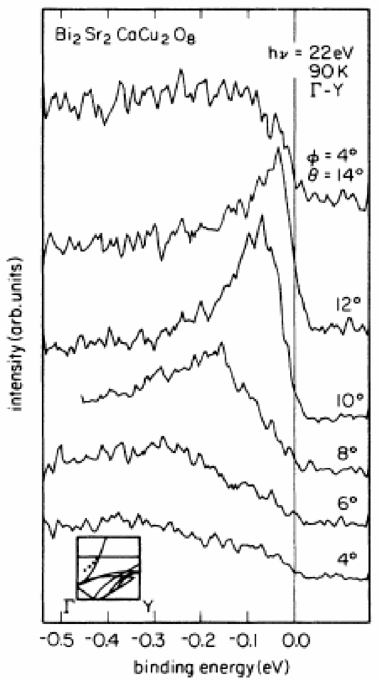
Self energy
=> need Hamiltonian of system !

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



Experimental Situation on Line Widths

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

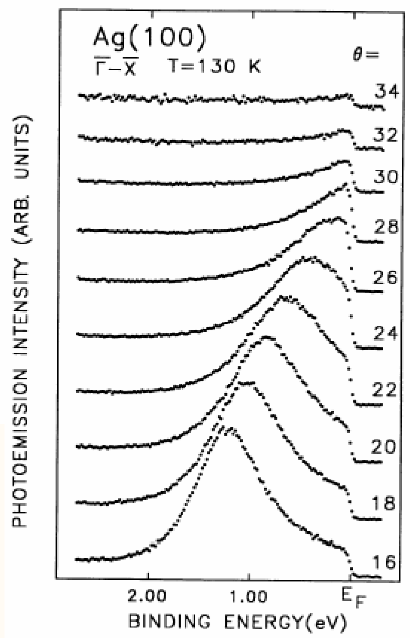


Experiment on HTC material:

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

=> non-Fermi-liquid ?

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



… but:

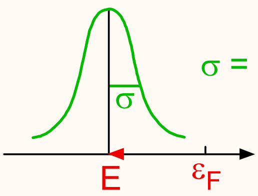
Silver (a much simpler material) even shows a growing line width towards EF !

?

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

⇒One needs to know the technique well !

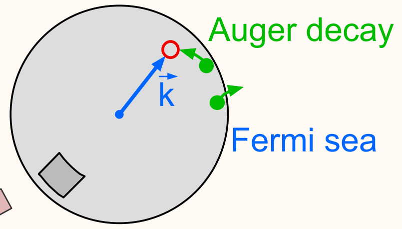
Interpretation of Line Widths



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

Q.P. Life time

dominant hole decay processes:

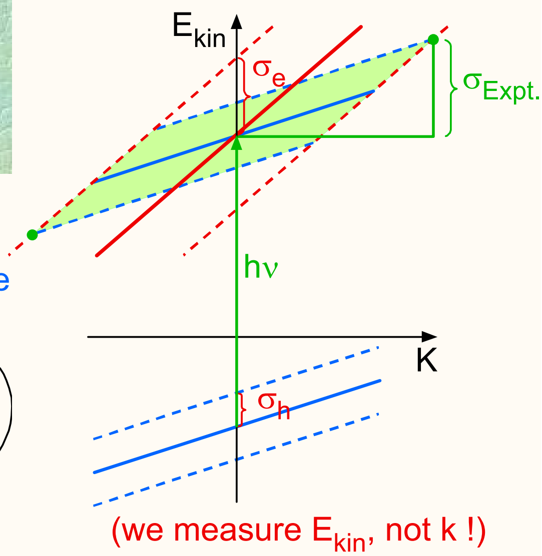


Fermi liquid theory

$\rightarrow \Delta t \sim \frac{1}{E^2}$

$\rightarrow \boxed{\sigma \sim E^2}$

Caution !



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

normal emission:

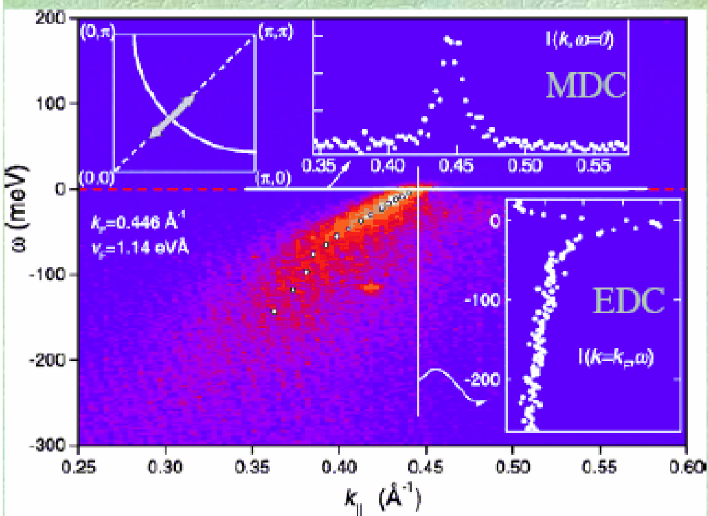
(P.Thiry, 1981)

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

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

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

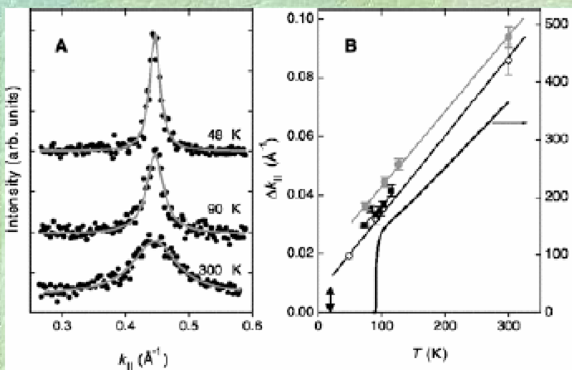
Example: Dispersion Plot from BSCCO High Temperature Superconductor



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:

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

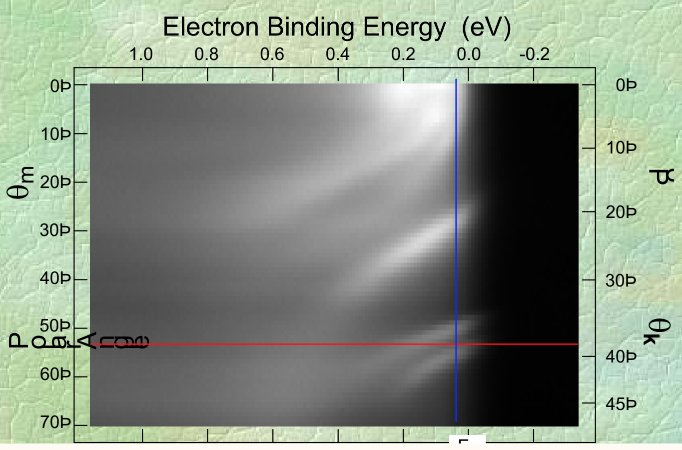
(ν_k is the electron group velocity)

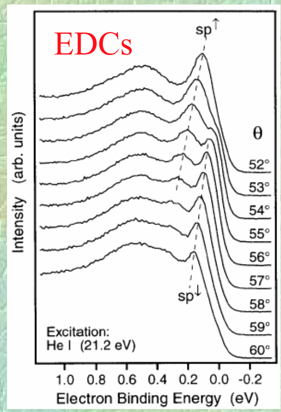


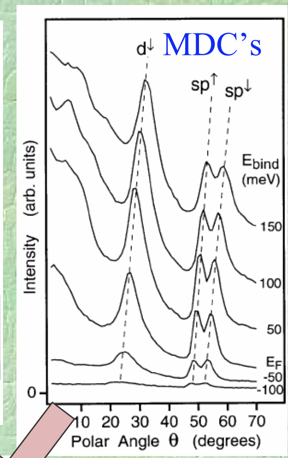
⇒ Connection between photoemission data and sample resistivity

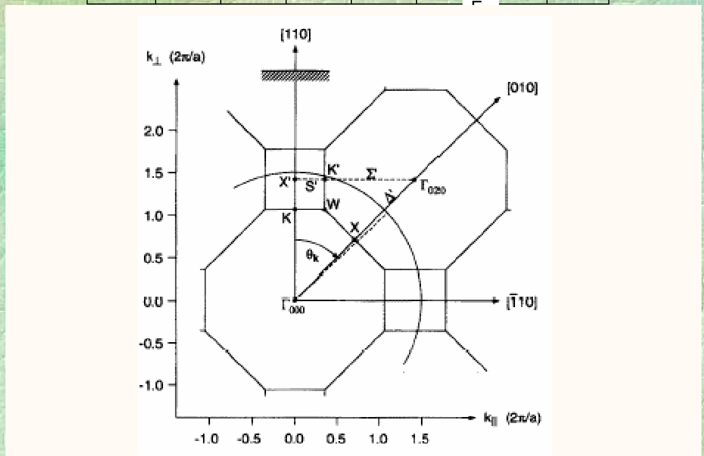
T. Valla et al. Science 285, 2110 (1999)

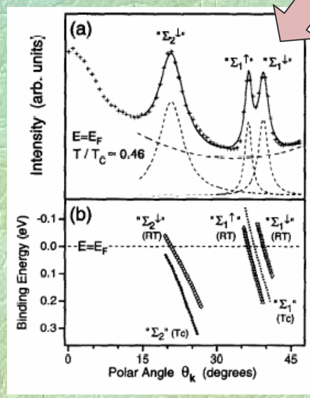
Extracting Energy Dispersion











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

School on Synchrotron Radiation and Applications, ICTP Trieste, May 3-6, 2004

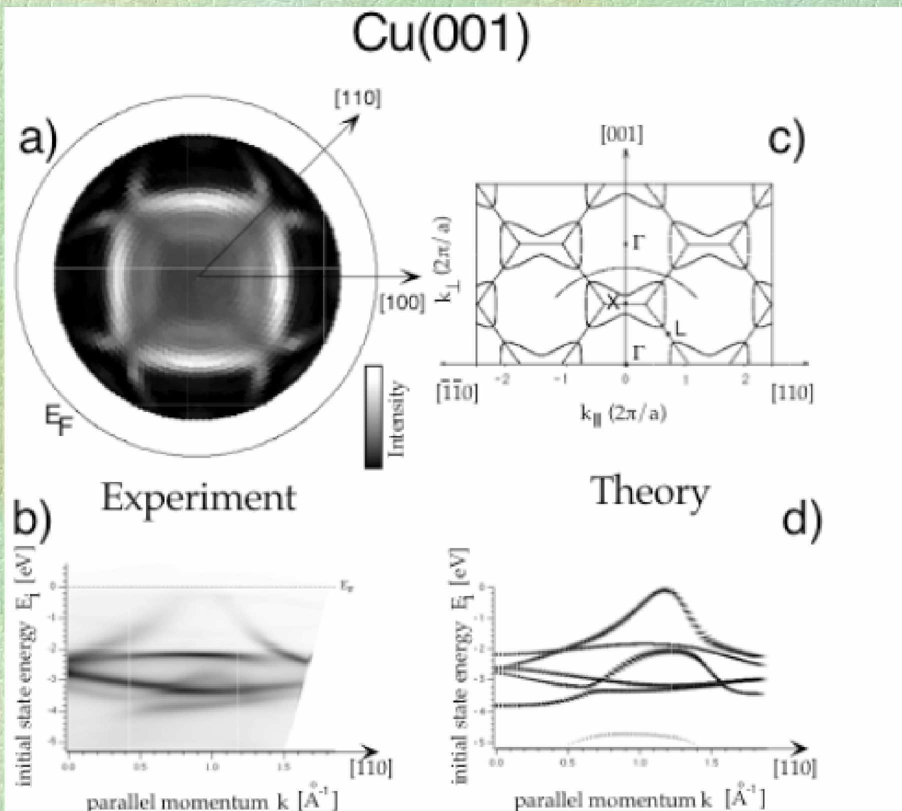
Photoemission as a Solid State Spectroscopy: Many-Body Effects, Applications to Complex Materials

Jürg Osterwalder
Physik-Institut, Unversität Zürich, Winterthurerstr. 190, CH-8057 Zürich, Switzerland - osterwal@physik.unizh.ch
http://www.physik.unizh.ch/groups/grouposterwalder/

Lecture 5

- Electron-Electron Interaction: Weak Effects in Photoemission from Cu
- Electron-Electron Interaction: Strong Effects in Photoemission from Ni and Co
- Electron-Phonon Interaction: Renormalization of Dispersion by Phonons
- Applications: Observation of a Giant Kohn Anomaly on H/Mo(110)
- Applications: Photoemission from High and Low T_c Superconductors

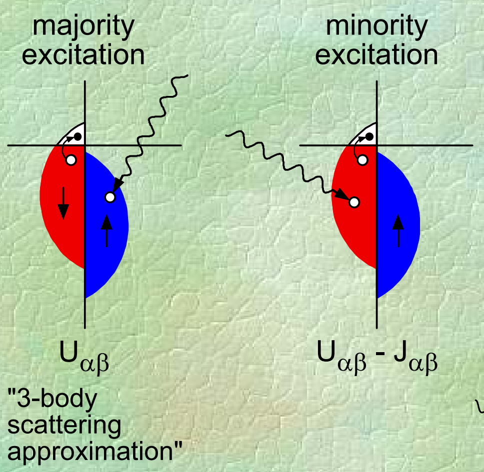
Weak Electron-Electron Interaction Effects in Photoemission from Cu



Courtesy P. Aebi, University of Neuchatel

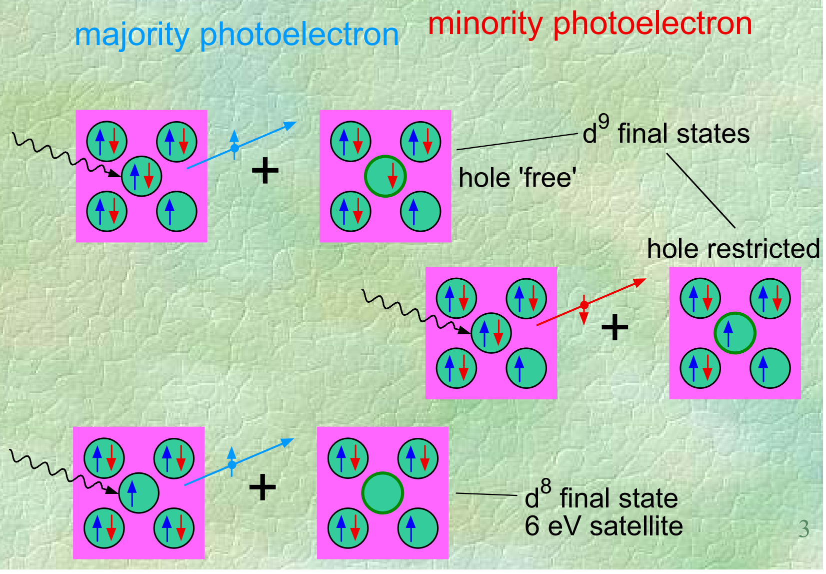
Valence Photoemission from Ni

$3d^{9.4}$ → correlation effects in the 3d channel



F. Manghi et al.,
PRL 73, 3129 (1994).

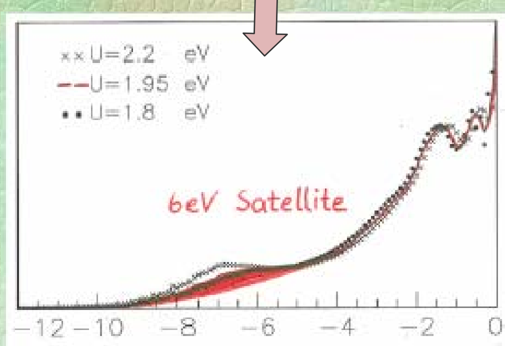
Pictorial:

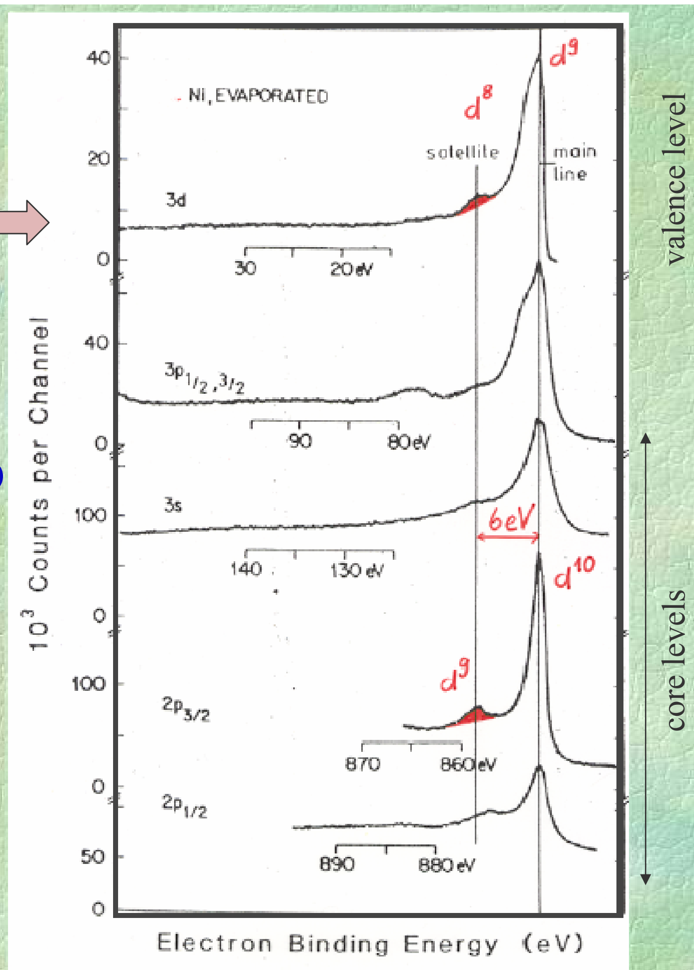


The 6 eV Satellite in Ni Metal

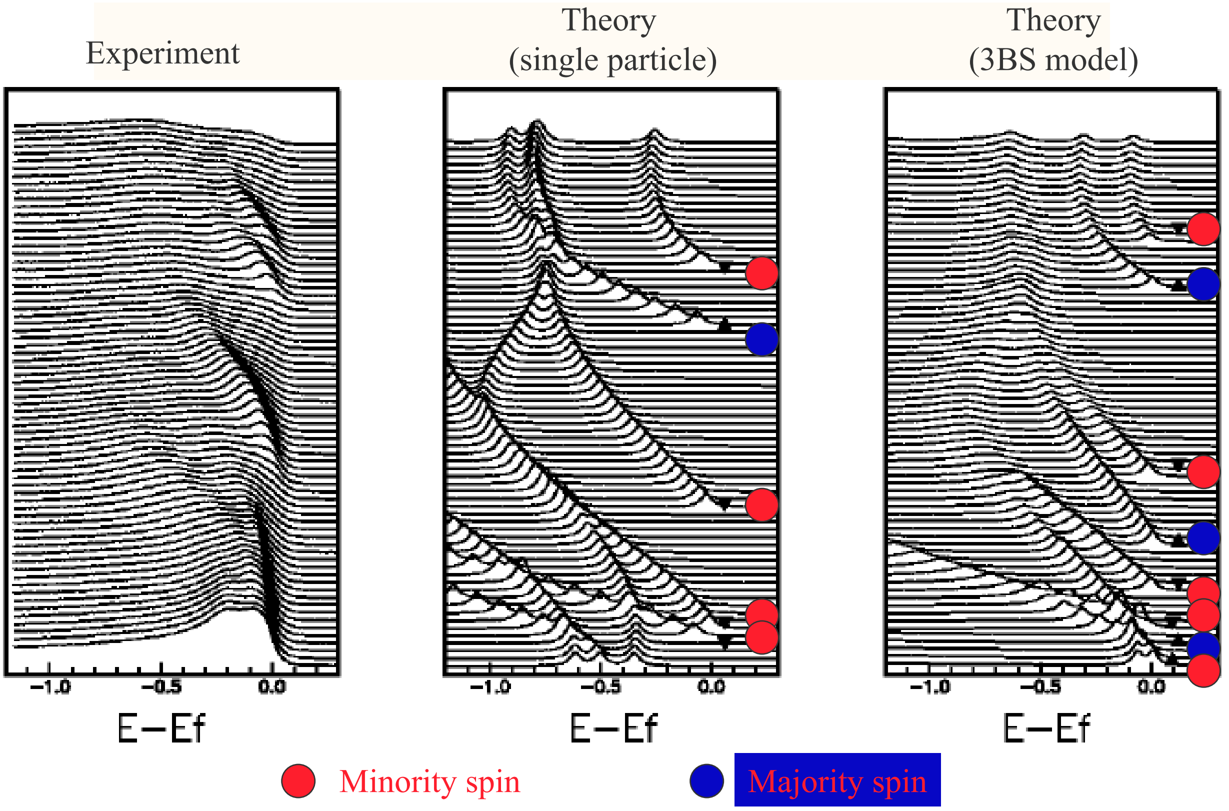
Experiments: Each photohole is accompanied by a 6 eV satellite
(S. Hüfner, Photoelectron Spectroscopy (Springer, Berlin 1995))

Theory: 3BS Model
(F. Manghi et al., PRB 59, R10409 (1999)



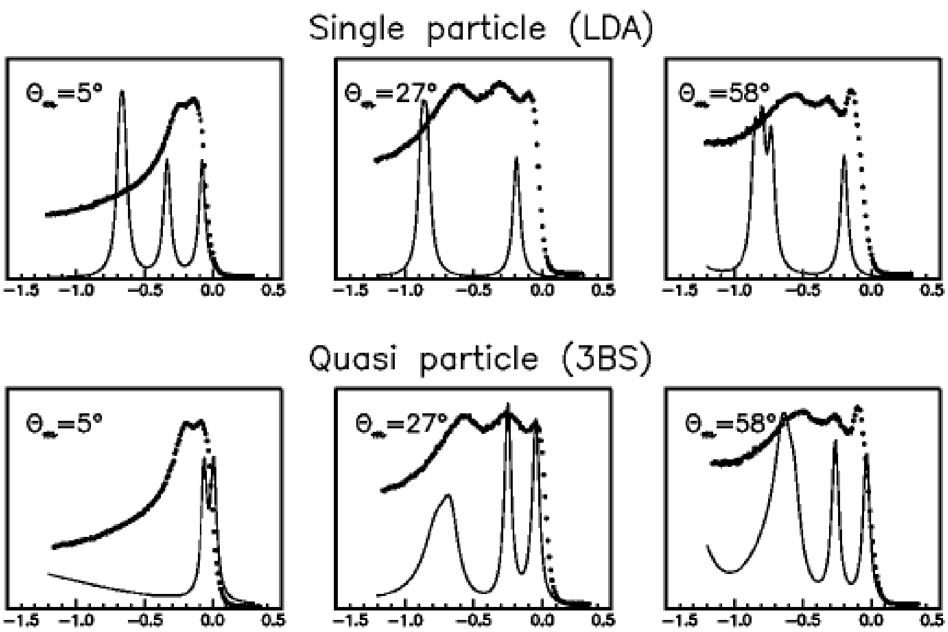


Renormalization of Band Dispersion by e-e Interaction: Ni



F. Manghi et al., PRB 59, R10409 (1999)

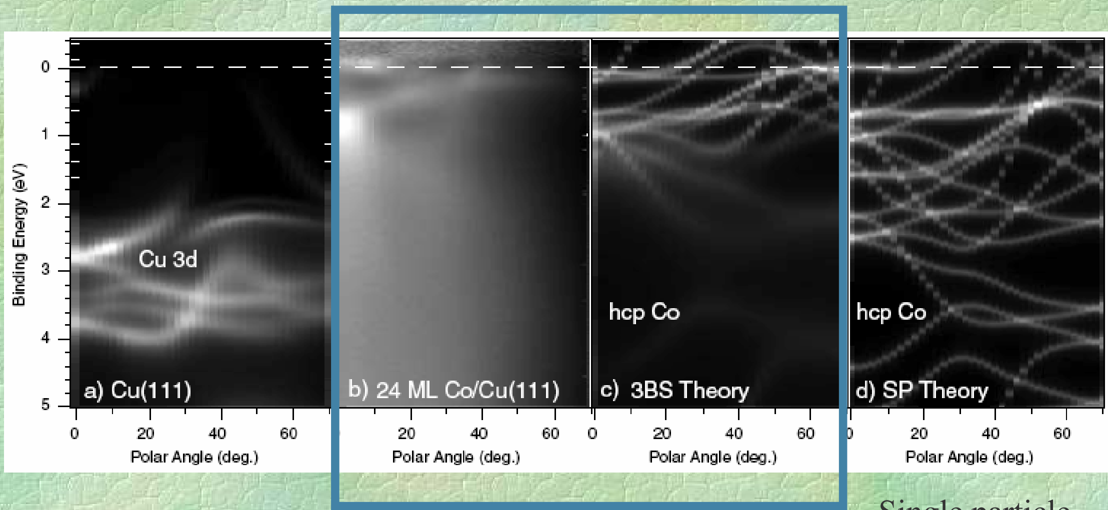
Quantitative Comparison of e-e Interaction Effects in Ni



F. Manghi et al., PRB 59, R10409 (1999)

Even Stronger Renormalization in Cobalt

Sample: Co(0001) / Cu(111)

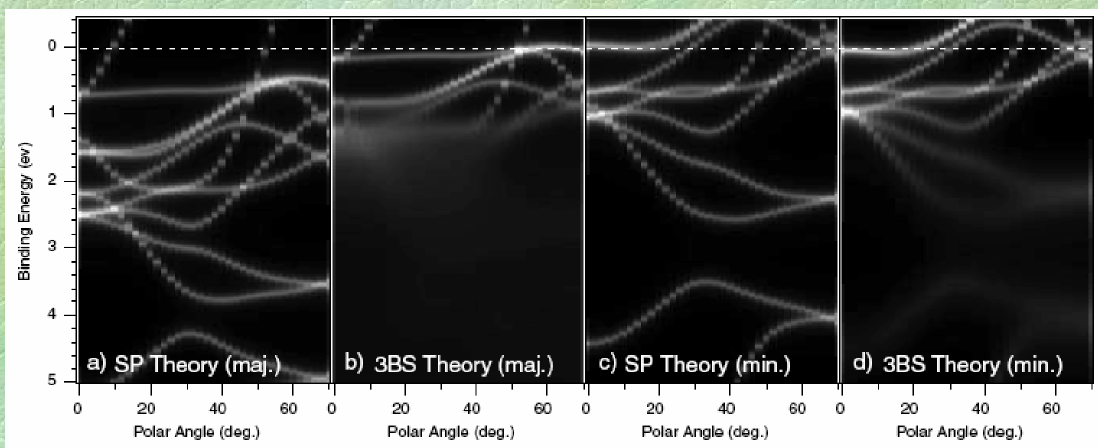


Single particle

Strong many-body effects !

S. Monastra et al., PRL 88, 236402 (2002)

Many-Body Effects are Strongly Spin Dependent



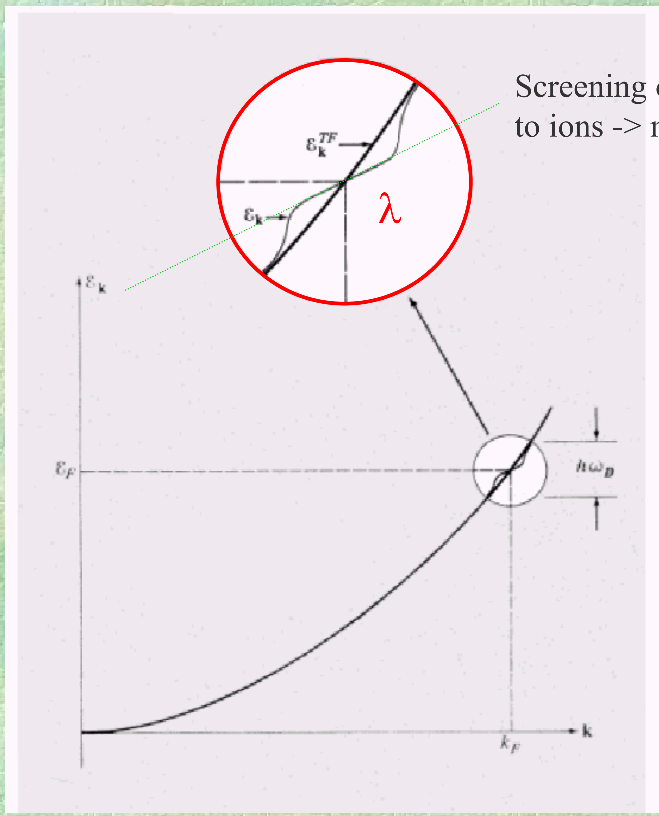
Majority spin

Minority spin

S. Monastra et al., PRL 88, 236402 (2002)

Electron-Phonon Interaction: Renormalization of Dispersion by Phonons

λ
electron-mode coupling constant



Screening of electrons due to ions -> m* increases !

Renormalization of m* by

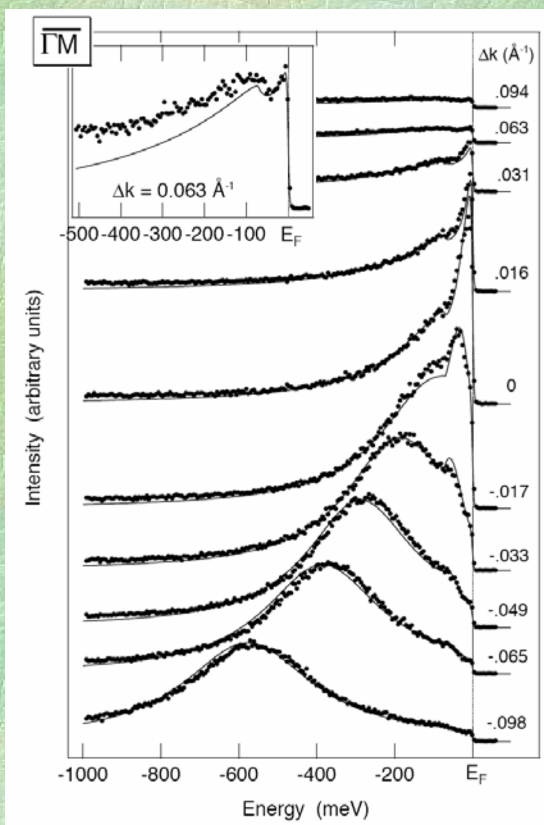
$Z^{-1} = 1+\lambda$

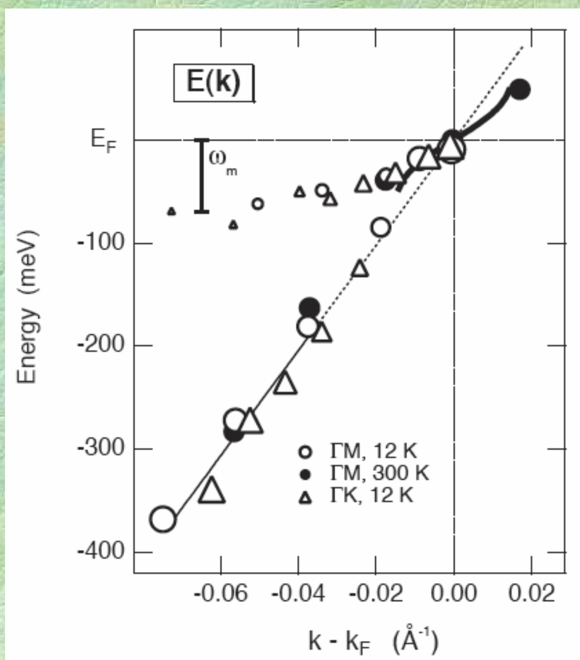
Some values for λ:

Cu 0.15
Be 0.24
Pb 1.50

Ashcroft, Mermin
"Solid State Physics"

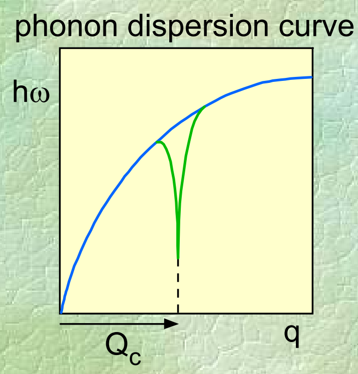
Observation of m* Renormalization in Beryllium

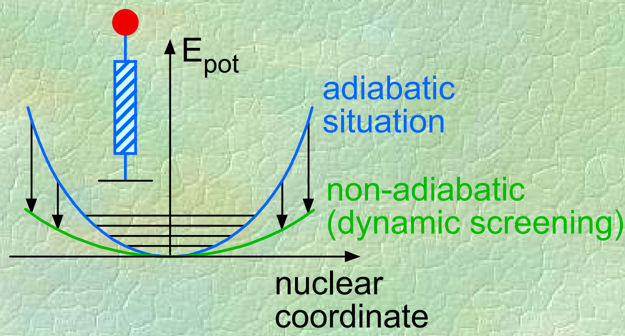




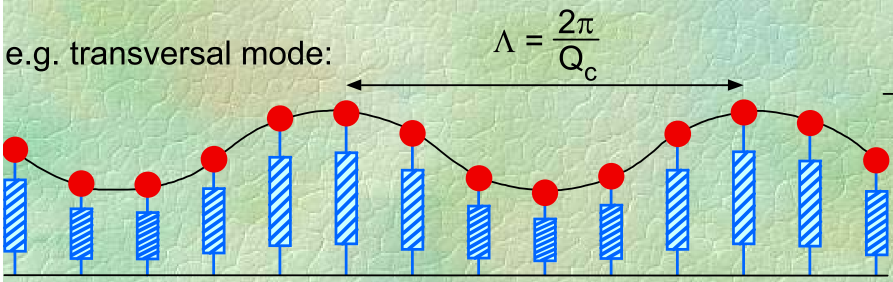
M. Hengsberger et al.,
PRL 83, 592 (1999).

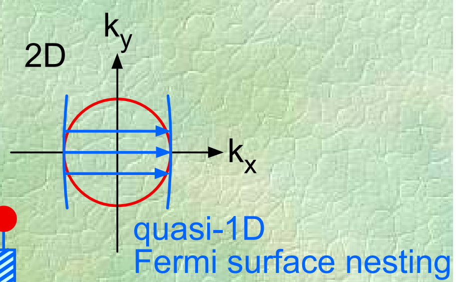
Giant Kohn Anomaly





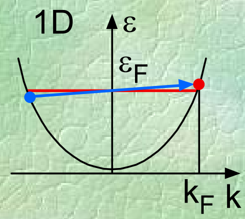
e.g. transversal mode:



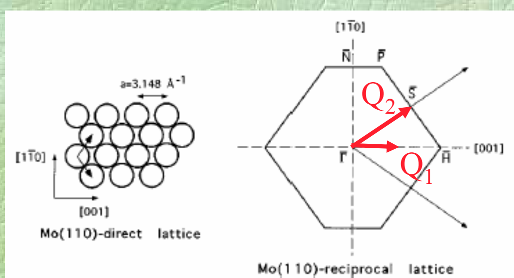


optimum screening:

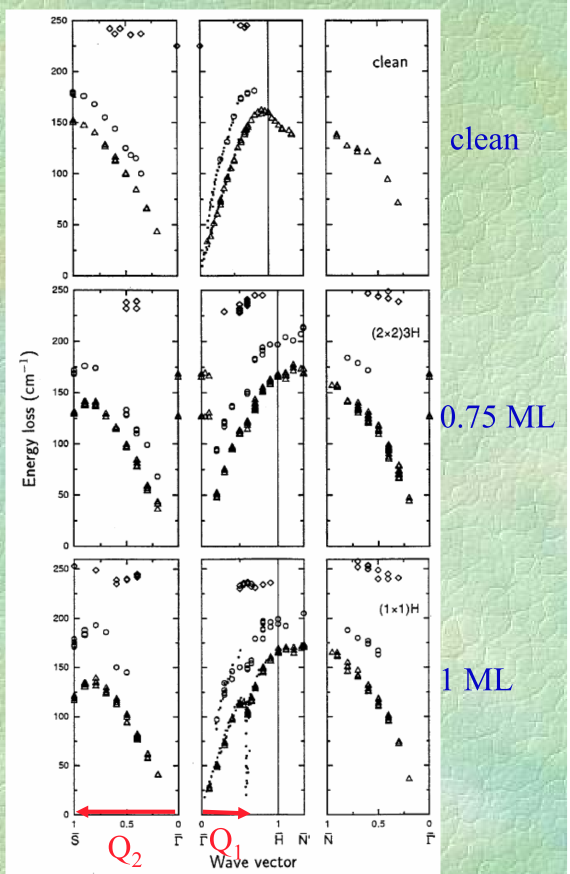
- standing wave with same wave number ($Q_c = 2\,k_F$!)
- large phase space for e^- - hole pair excitations (low ε / high q)



Surface Phonon Modes for H/Mo(110) - A Giant Kohn Anomaly ?



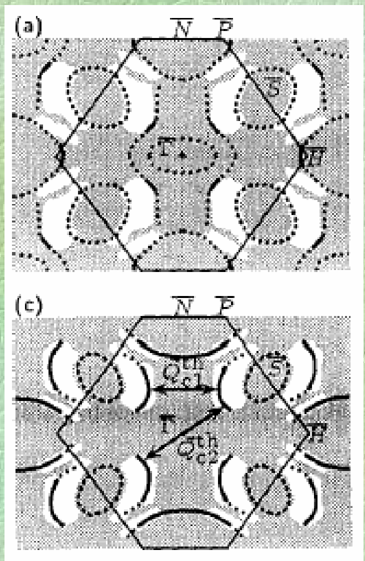
HREELS Data



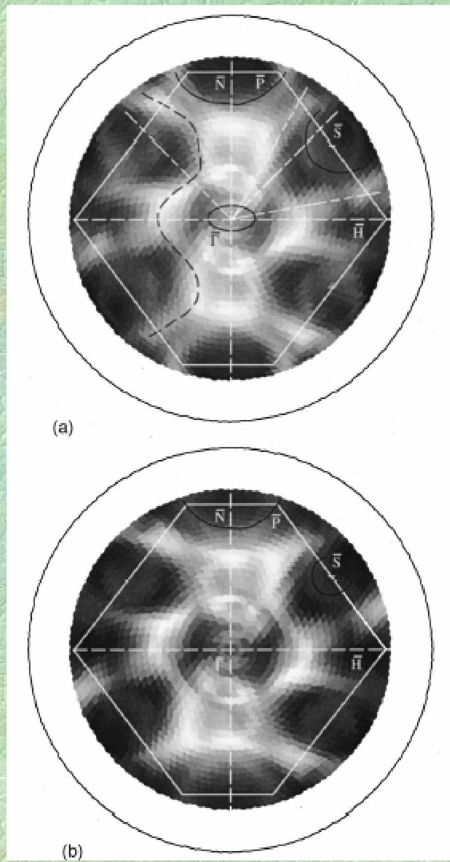
J. Kröger et al., PRB 55, 10895 (1997)

Fermi Surface Contours of Surface States on Mo(110)

Theory:



B. Kohler et al., PRL 74, 1387 (1995)

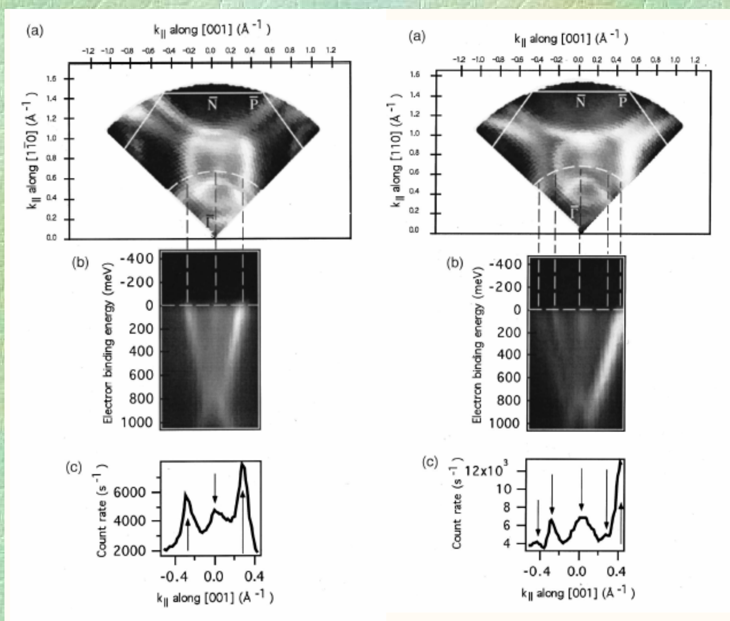


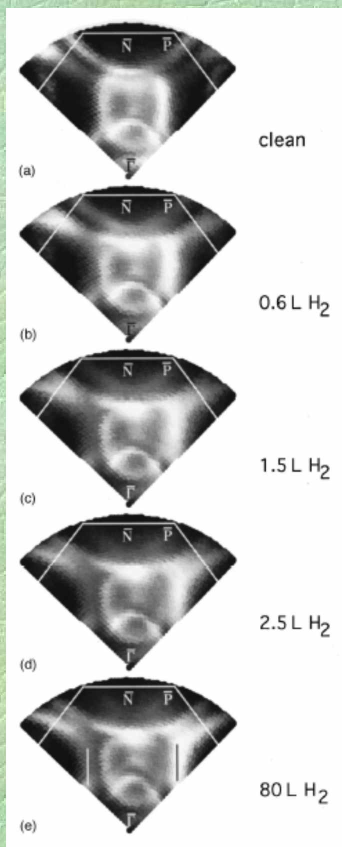
Clean Mo(110)

1 ML Hydrogen

J. Kröger et al., PRB 61, 14146 (2000).

Evolution of Fermi Surface Contours with Hydrogen Coverage

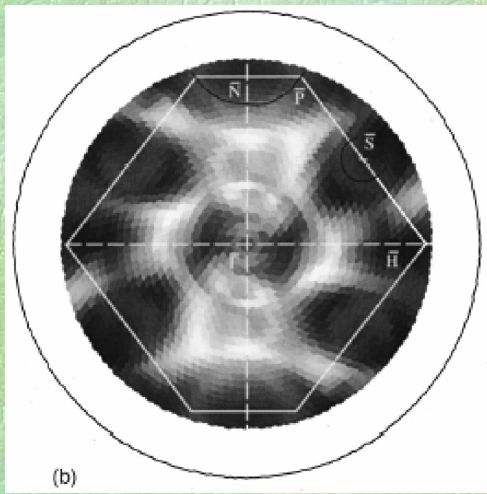


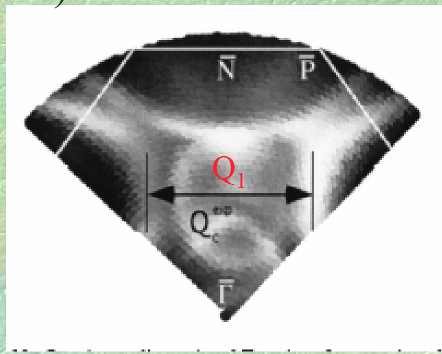


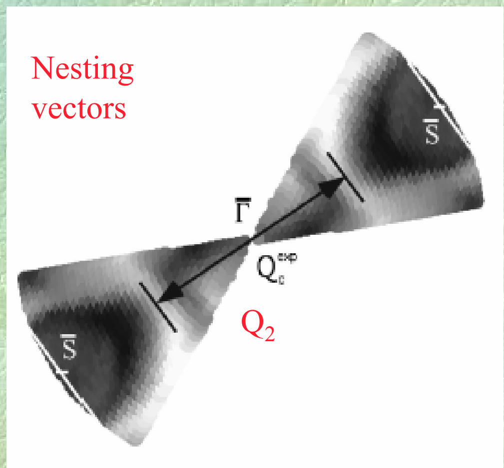
=> H-induced shift of Fermi surface contour

J. Kröger et al., PRB 61, 14146 (2000).

Fermi Surface Nesting on 1 ML H/Mo(110) Seen by Photoemission





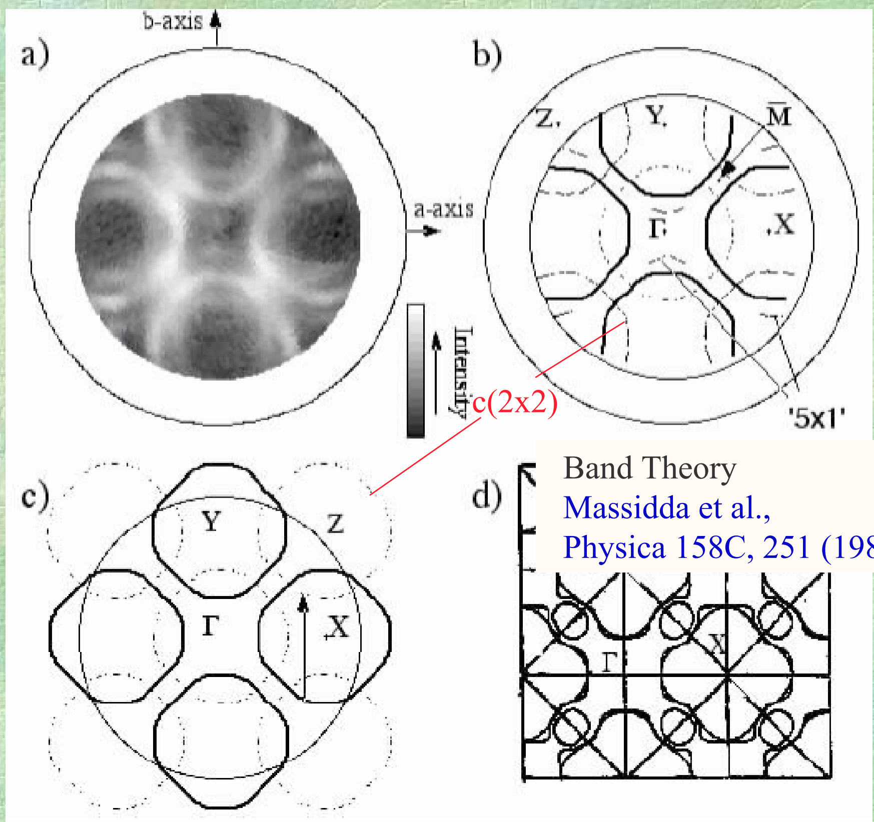


	Q_1(Å^{-1})	Q_2(Å^{-1})
EELS	0.90	1.22
PE	0.85	1.19
DFT	0.86	1.23

J. Kröger et al., PRB 61, 14146 (2000).

Fermi Surface Mapping on a High Temperature Superconductor: $Bi_2Sr_2CaCu_2O_{8+\delta}$ (001)

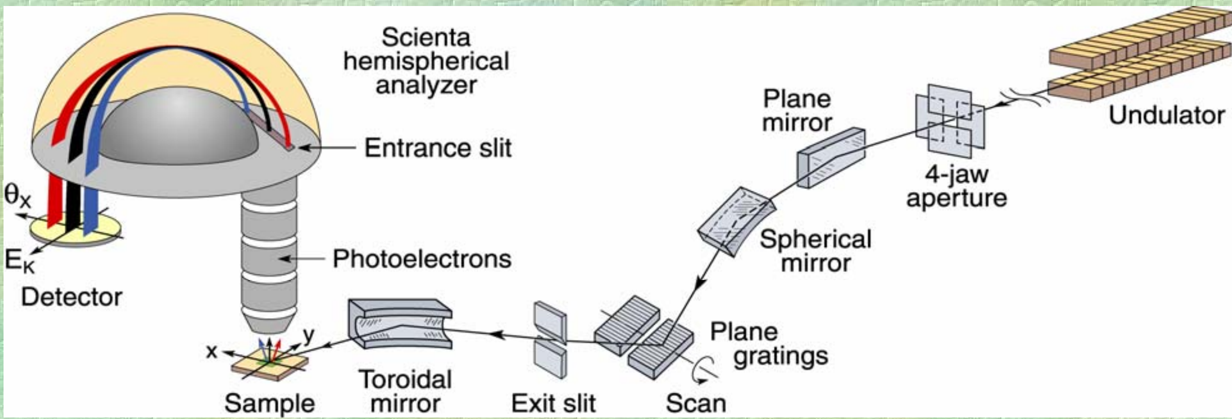
P. Aebi et al.,
PRL 72, 2757 (1994)



Band Theory
Massidda et al.,
Physica 158C, 251 (1988)

High-Resolution Photoemission

A state-of-the-art photoemission beamline at Stanford (SSRL)

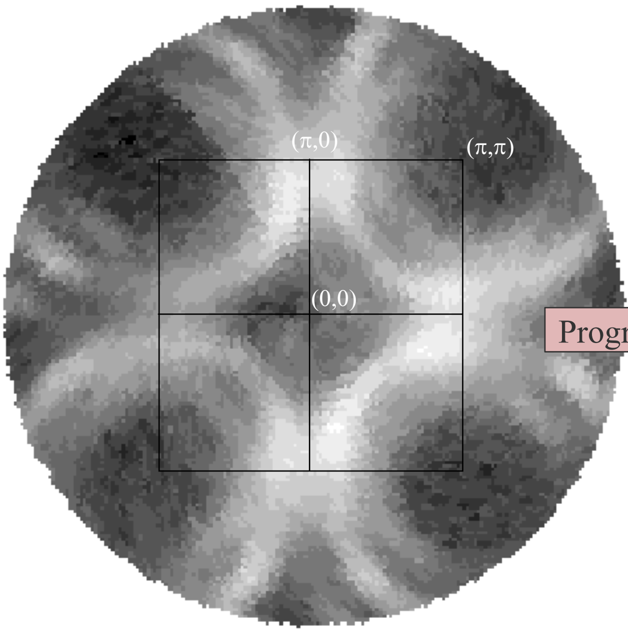


Highest resolution in energy and momentum

~ 2 meV in energy
~ 0.005 Å^{-1} in momentum

Courtesy Z.-X. Shen, Stanford University

Bi2212:
First Fermi surface mapping, superstructure contours

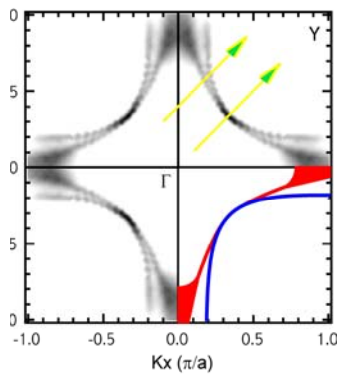


P. Aebi et al., Phys. Rev. Lett. 72, 2757 (1994).

Progress

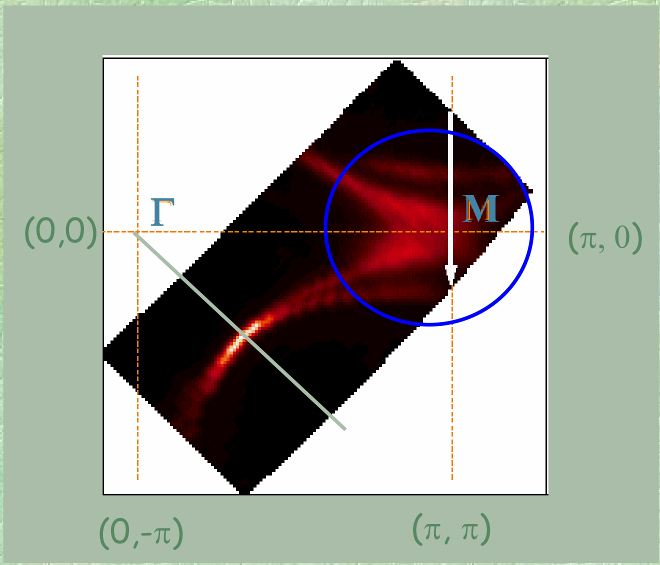
Pb-doped Bi2212:
-layer splitting

Two CuO layers per unit cell



gdanov et al.,
ys. Rev. B 64, 180505 (2001)

Details on the Fermi Surface of BSCCO

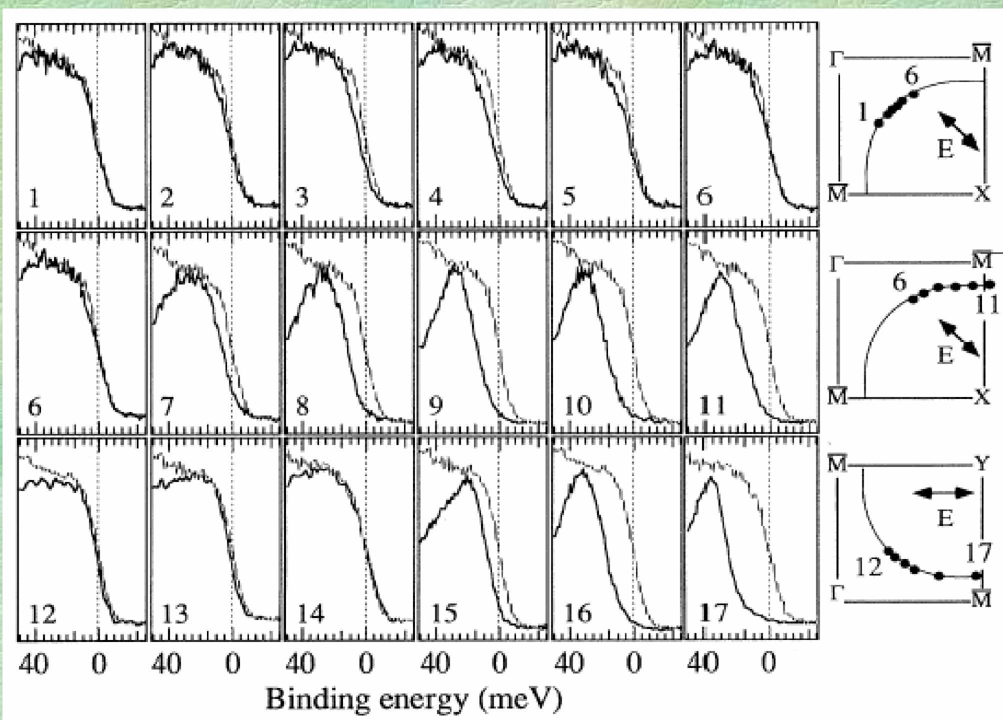


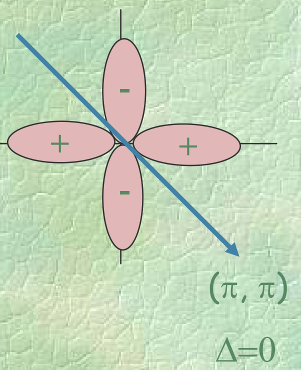
- Superstructure
- Superconducting Gap
- Bilayer Splitting

Courtesy Z.-X. Shen, Stanford University

Anisotropy of the Superconducting Gap in BSCCO

… need to measure spectra exactly on the Fermi surface !

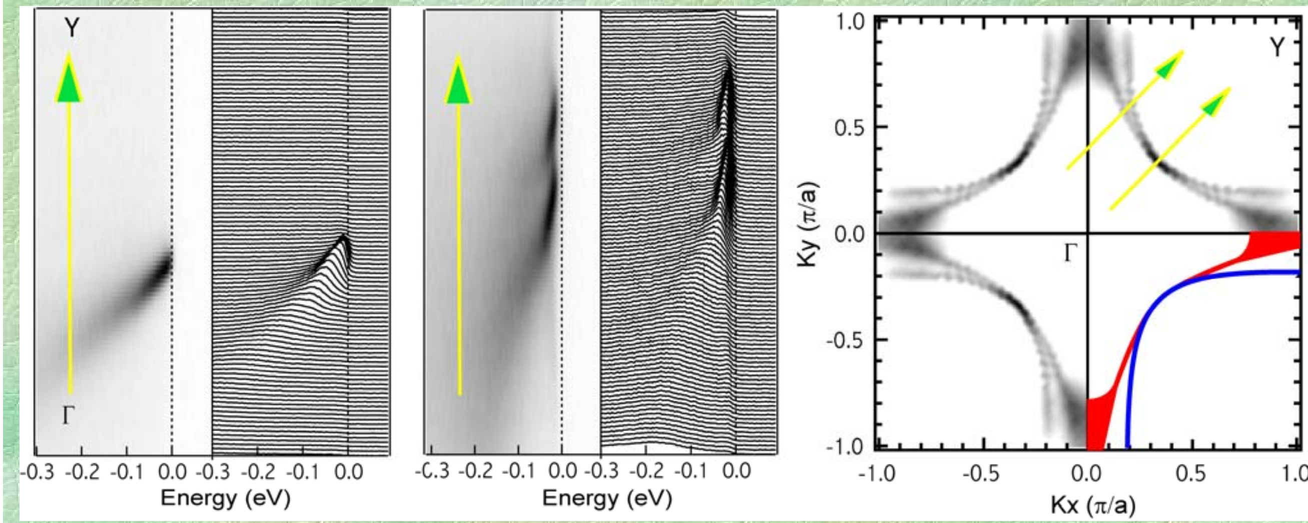




=> d-wave superconductor

H. Ding et al., PRL 74, 2784 (1995)

Pb-doped Bi2212: Bi-layer splitting

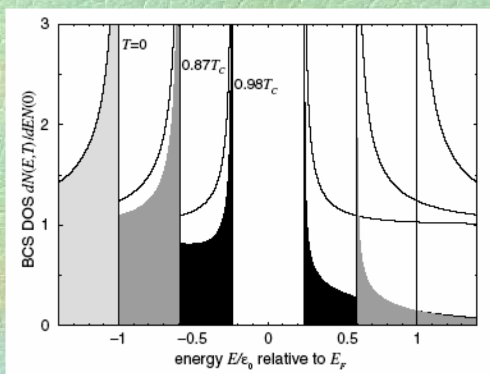


High energy and momentum resolution combined with improved sample enables the clear observation of bilayer splitting

Bogdanov et al. Phys. Rev. B 64, 180505 (2001)

In Conventional Superconductors it is Much Harder to See the Gap !

Density of states near E_F in the superconducting state: BCS model for different temperatures



Sample:

V_3Si

$T_c = 17$ K
$\Delta_{gap} = 5$ meV

Instr. Res. 2.9 meV !

F. Reinert et al.,
PRL 85, 3930 (2000)