Angle Resolved Photoemission Spectroscopy

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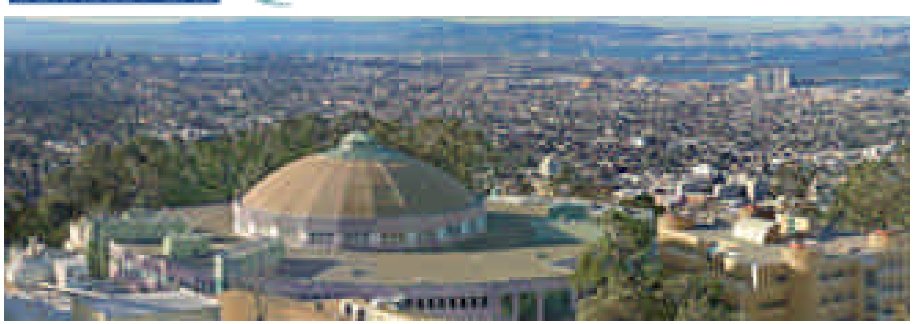
BERKELEY LAB



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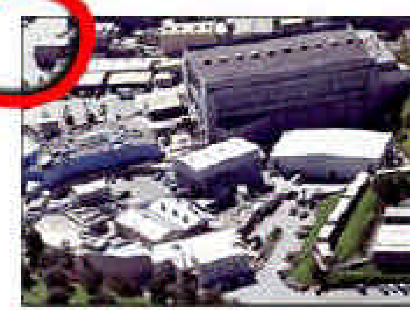
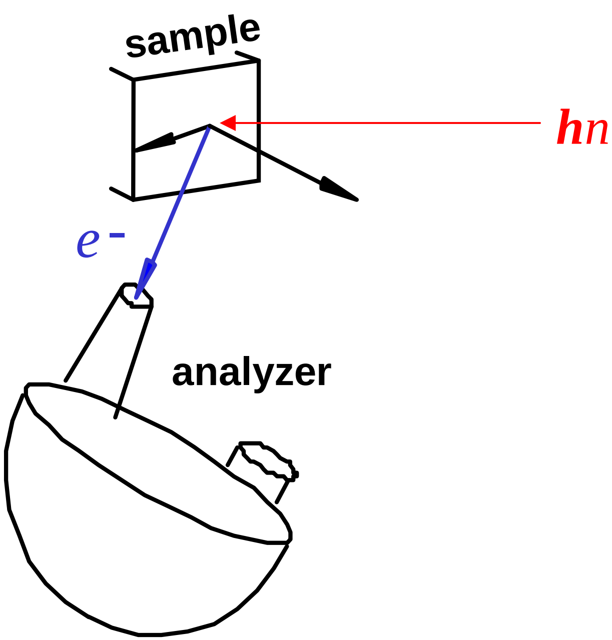
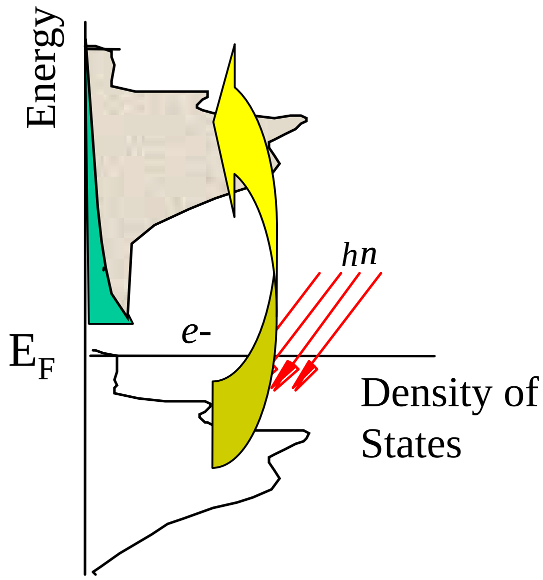
SSRL



Photoemission Spectroscopy





High K.E. Low B.E.

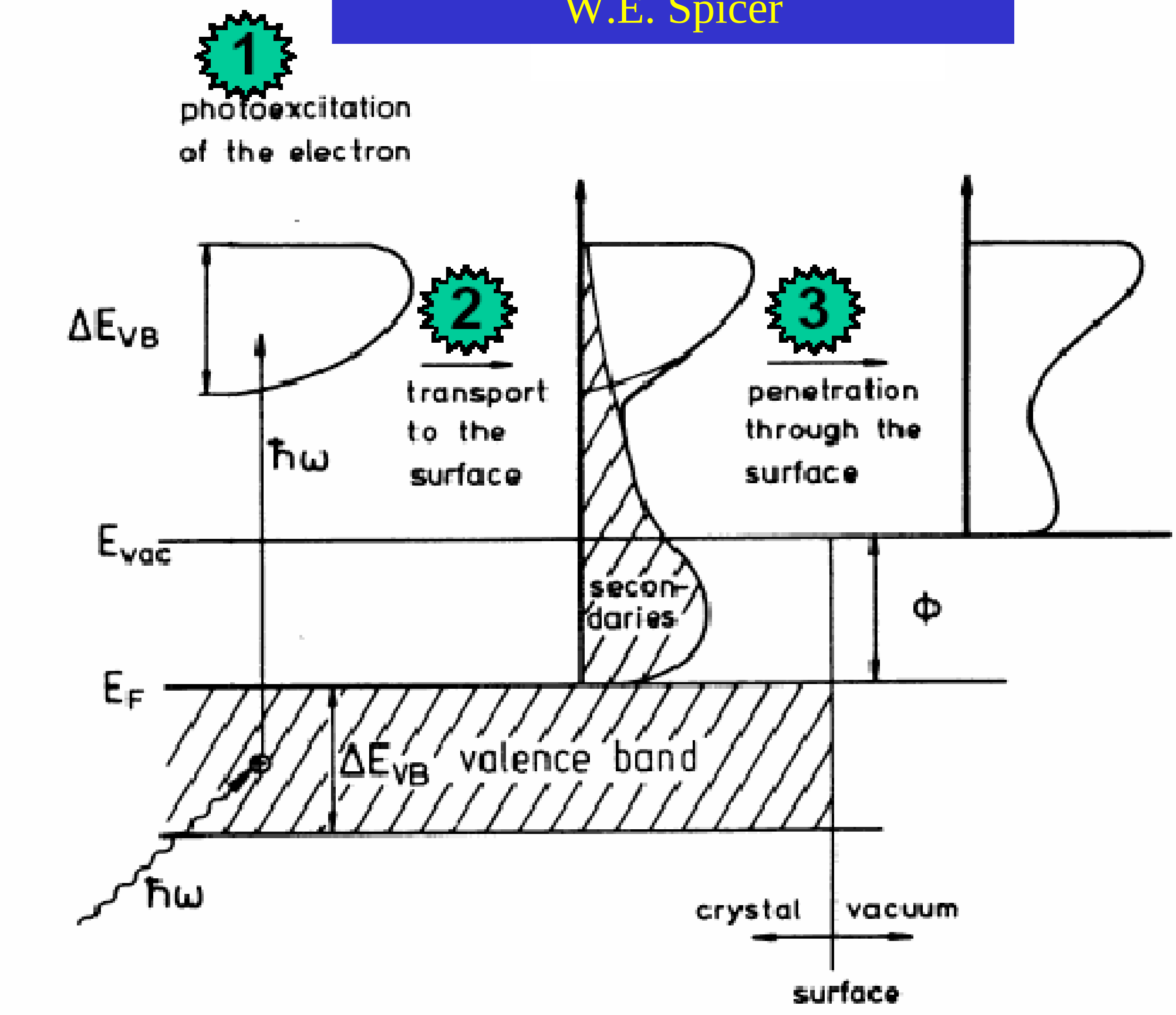
Low K.E. High B.E.

Primary electrons – no scattering events.
Contain information of density of states

Secondary electrons (inelastic background) – increases with decreasing kinetic energy.

$$E_{kin} = \hbar\omega - \Phi \; - \left| E_B \right|$$

Three Step Model
W.E. Spicer



Angle resolved photoemission spectroscopy

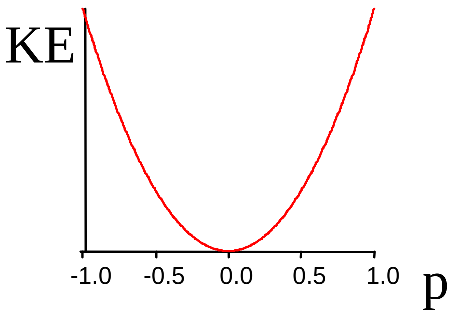
- Angle–dependent core level spectroscopy (not this talk)
  - To vary the surface sensitivity of an experiment
    - normal emission – more bulk sensitive
    - glancing emission – more surface sensitive
  - To perform X-ray photoelectron Diffraction (XPD)
    - Obtain local structural information (similar to EXAFS)

- Angle-dependent valence band spectroscopy (this talk)
  - To measure the k (momentum) dependence of valence band states
  - To measure electronic band dispersions and Fermi Surfaces
  - To measure symmetries of states
  - To obtain many-body phenomena (e.g. correlated electron systems).

Quantum numbers E,k

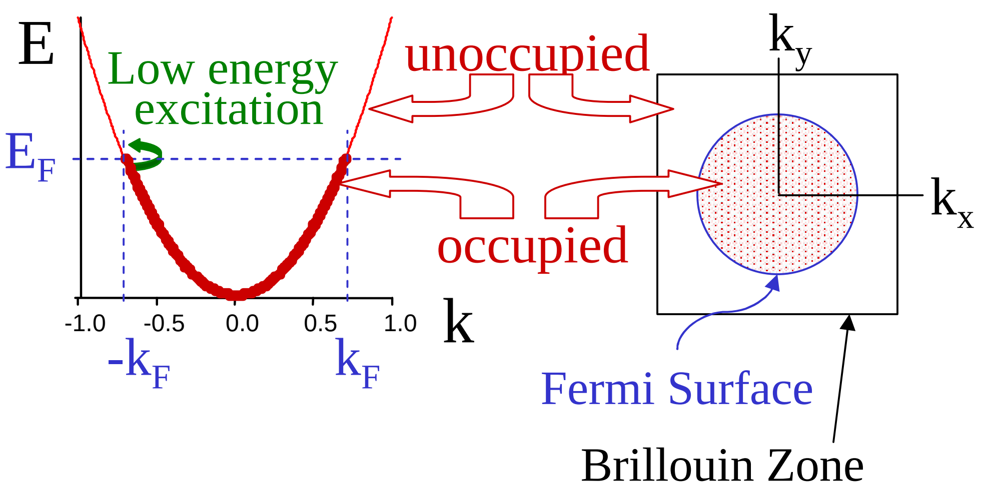
Newton : KE=0.5 $mv^2 = p^2/2m$

$p=mv$, $p=\hbar k$

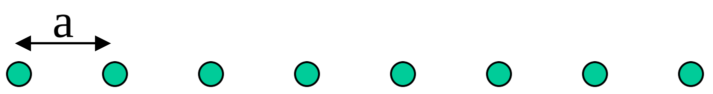


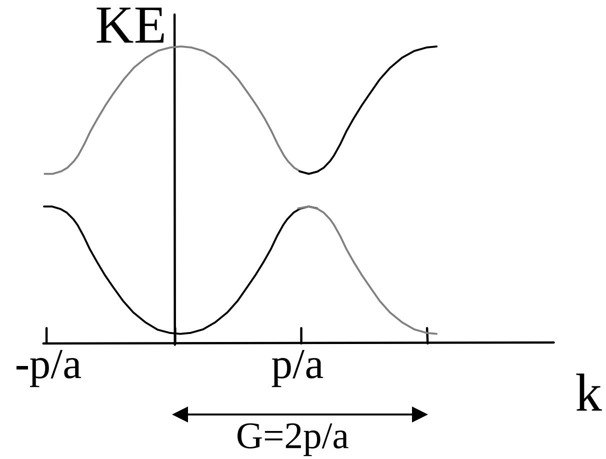
Energy (E) and momentum (p or k) are the most important quantum numbers in a solid. Specifying these specifies behavior of electrons.

Pauli Principle - each electron goes into it's own individual quantum state (new E,k). Fill lowest energy states first. Highest energy electrons are at Fermi Energy E_F, with a momentum $\hbar k_F$. These are the most important electrons. All low energy excitations come from near E_F.

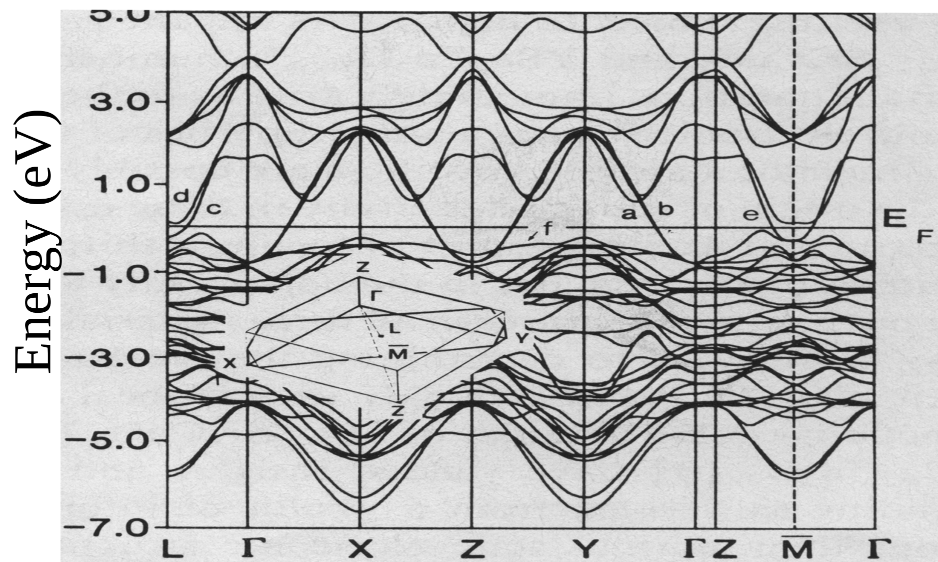


Electrons in a periodic potential





Band Structure calculation of $Bi_2Sr_2CaCu_2O_8$ (Bi2212).



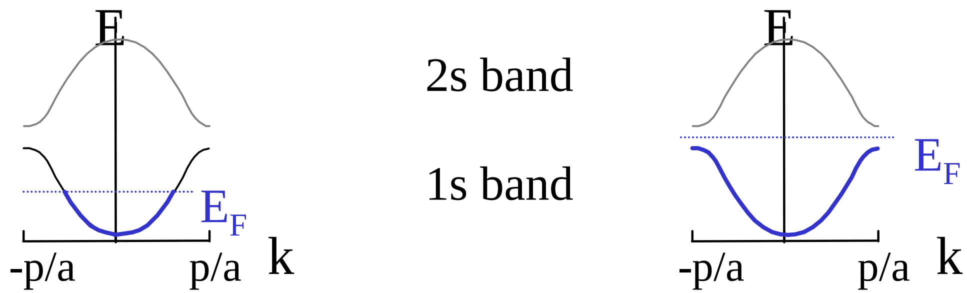
S. Massidda et al, Physica C **152**, 251 (1988)

Everything still mean-field or static - no correlation effects yet.

Metals and Insulators

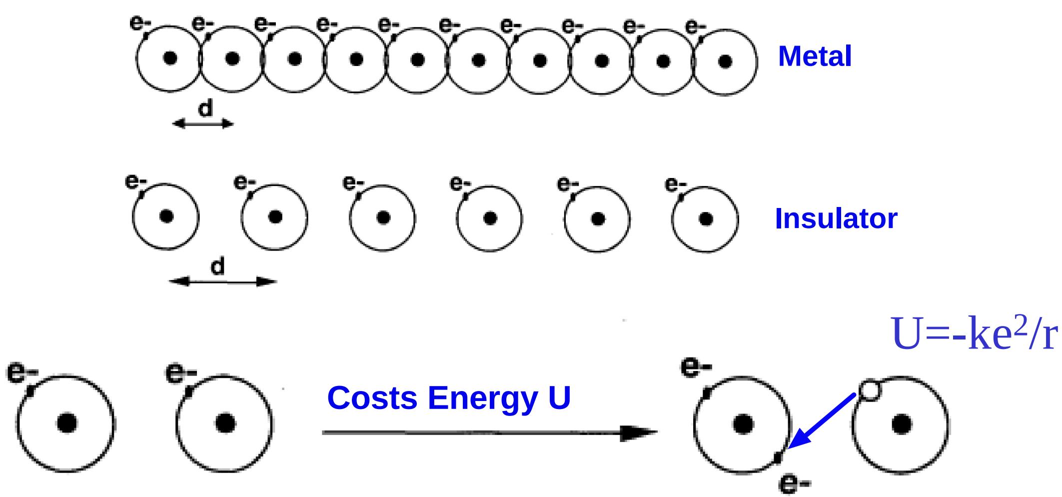
Band theory: E_F in a band --> Metal. E_F in a gap between bands --> insulator

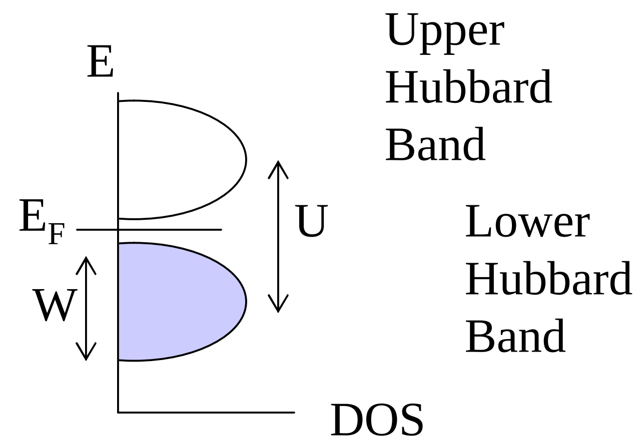
Solid H (metal)



Solid He (insulator)

Mott Insulators - Failure of this model

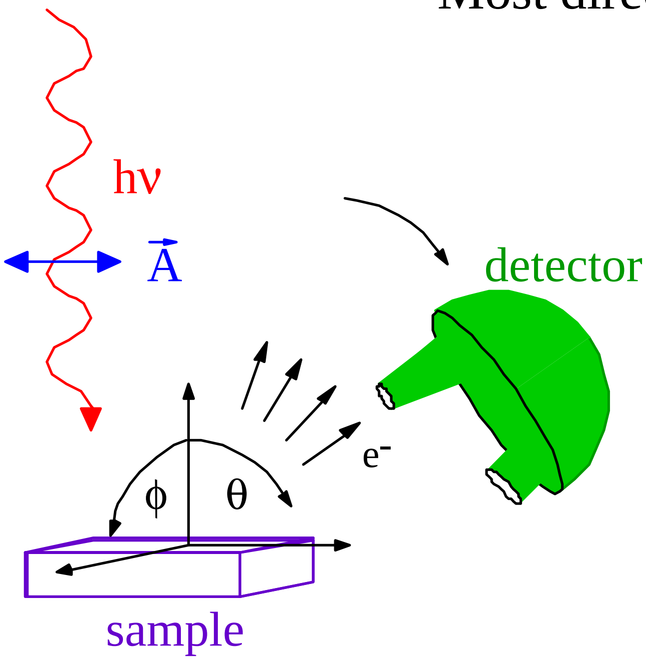




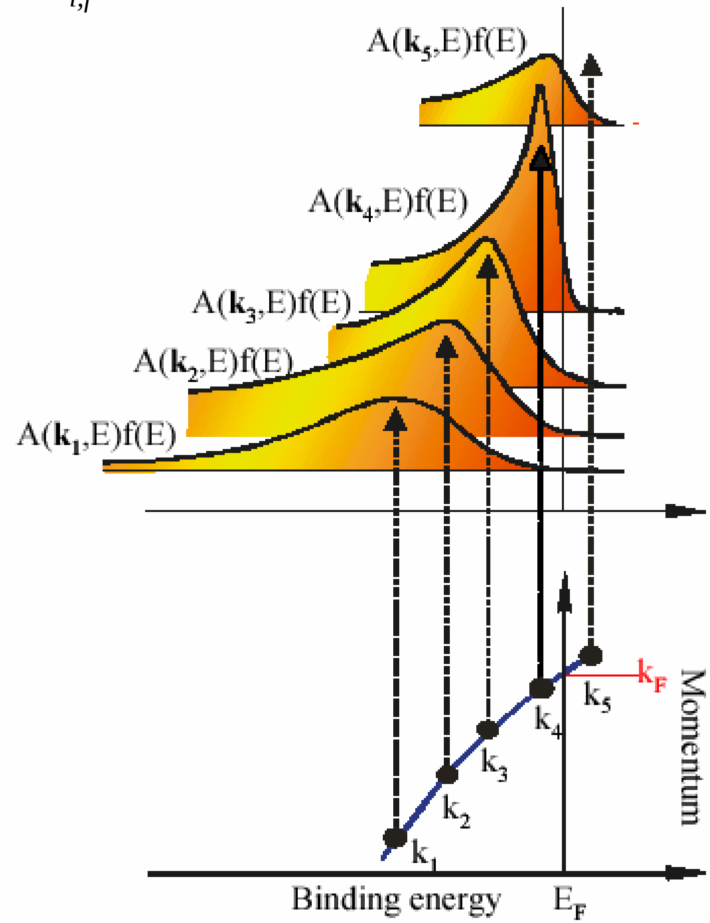
Effects most important in localized (d- and f-electron systems).
==> High Tc superconductors, Colossal Magnetoresistive oxides, etc.

Angle Resolved Photoemission (ARPES)
A momentum resolved spectroscopy

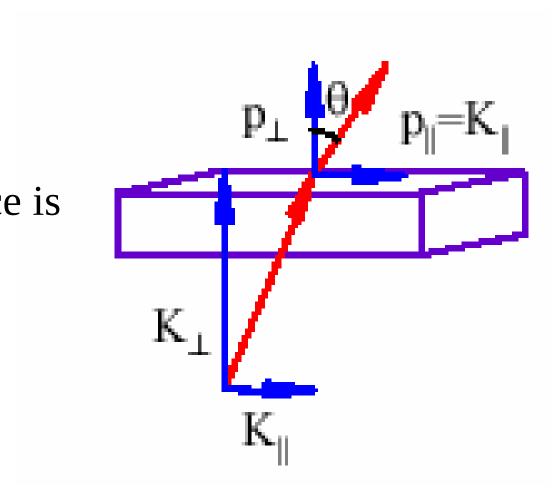
Most direct way to measure E vs. k of a solid.



$$\text{Intensity} \propto \sum_{i,f} \left| \langle f | \vec{\mathbf{p}} \bullet \vec{\mathbf{A}} | i \rangle \right|^2 A(\vec{\mathbf{k}}, E)\, f(E)$$



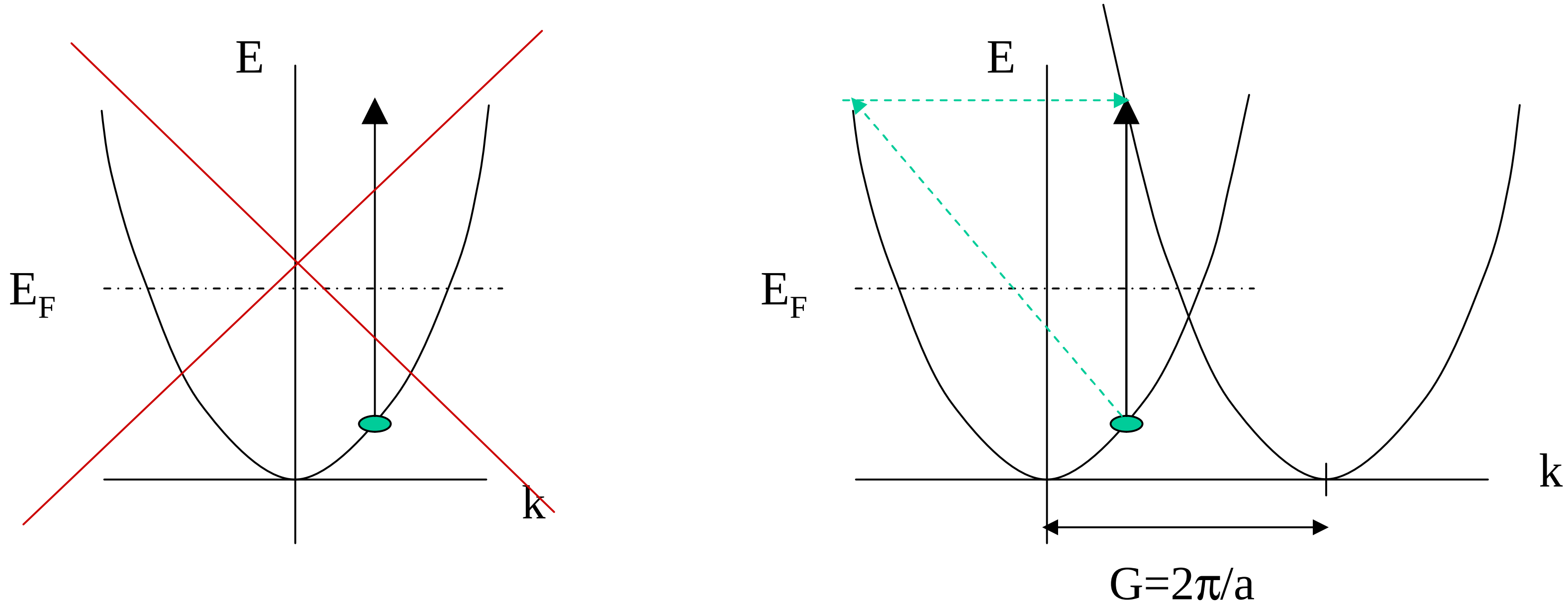
Electron momentum
Parallel to the surface is
conserved



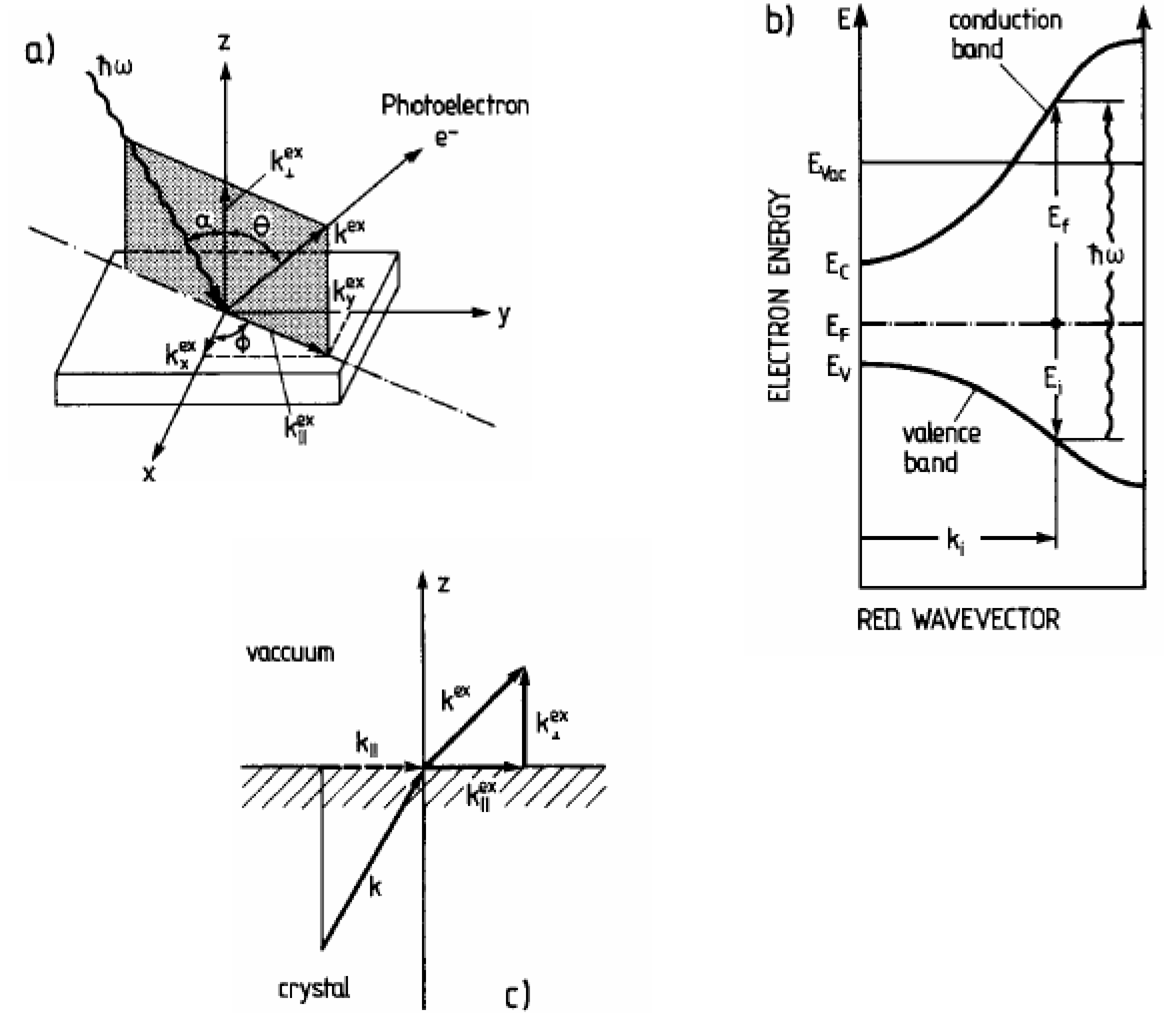
Momentum Conservation

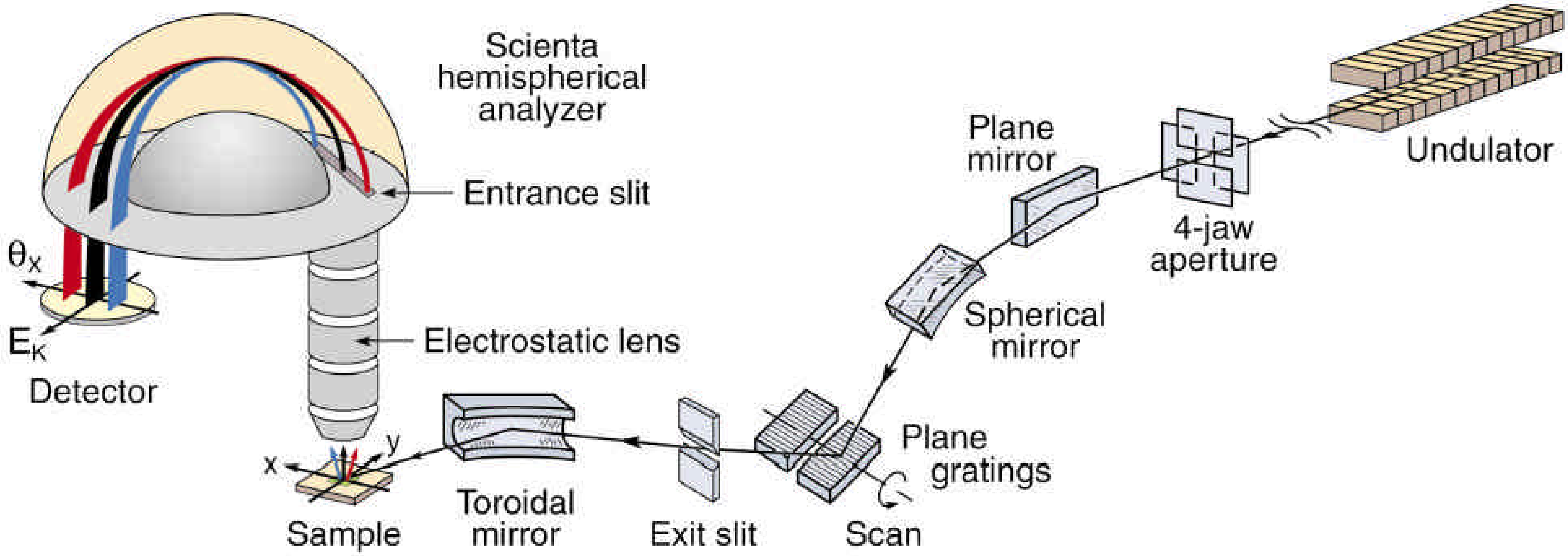
Photons of a few hundred eV or less carry negligible momentum compared to the typical electron momentum scales in a solid.

Therefore we consider "vertical" transition processes. For a free electron parabola there would be no final state and the process is forbidden.

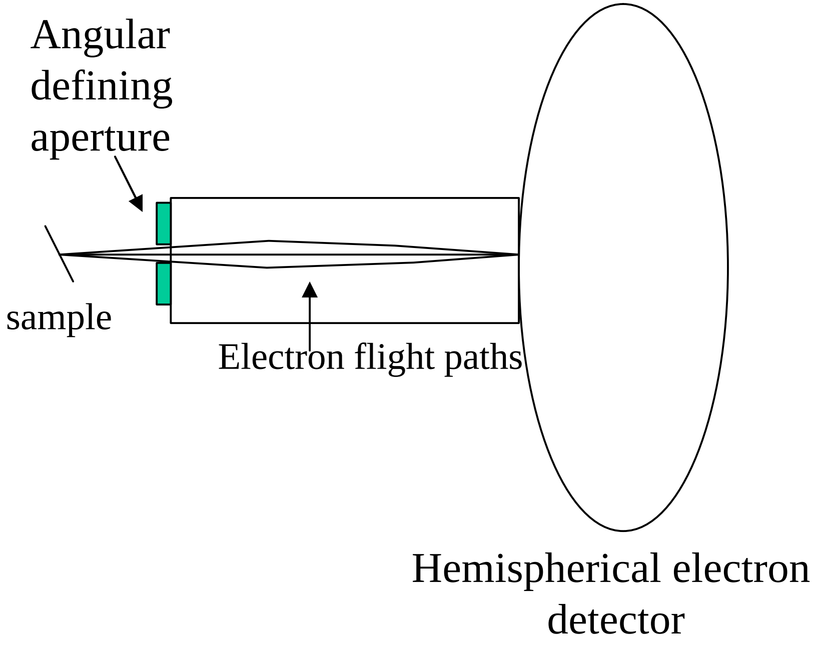


The vertical transition is allowed by considering the extended zone scheme and employing a reciprocal lattice vector $G=2\pi/a$ (the lattice degree of freedom takes care of the "missing" momentum).

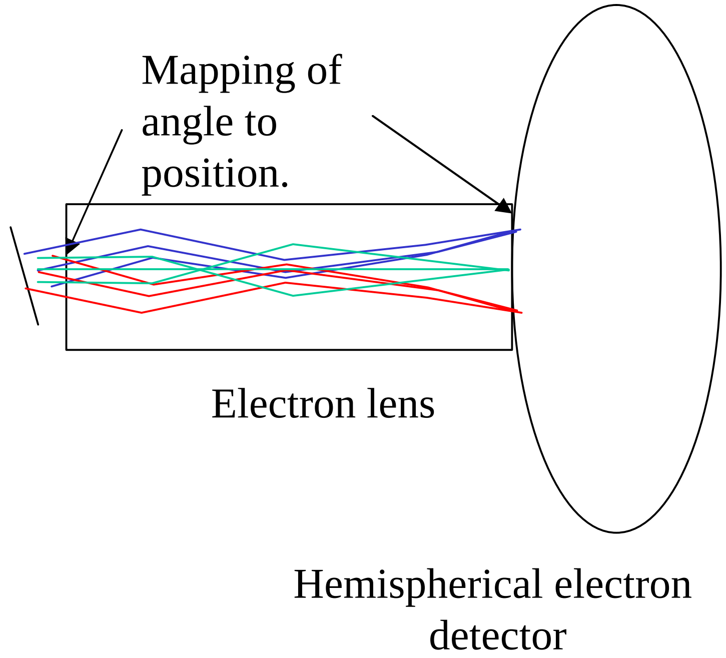

a)
$\hbar\omega$
z
Photoelectron
e^-
$k_\perp^{ex}$
α
Θ
k^{ex}
k_y^{ex}
y
k_x^{ex}
Φ
$k_\parallel^{ex}$
x
b)
E
conduction band
E_{Vac}
ELECTRON ENERGY
E_f
$\hbar\omega$
E_C
E_F
E_V
E_i
valence band
k_i
RED. WAVEVECTOR
z
vaccuum
k^{ex}
$k_\perp^{ex}$
$k_\parallel$
$k_\parallel^{ex}$
k
crystal
c)

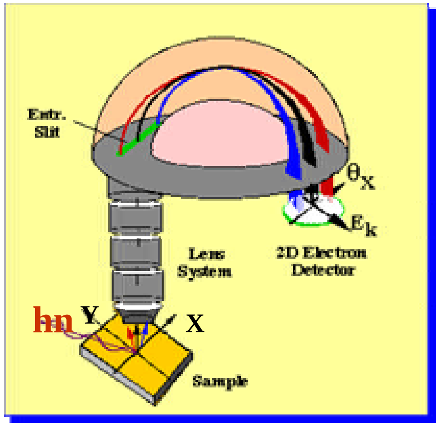

Scienta
hemispherical
analyzer
Entrance slit
θ_X
E_K
Detector
Electrostatic lens
x
y
Sample
Toroidal
mirror
Exit slit
Scan
Plane
gratings
Spherical
mirror
Plane
mirror
4-jaw
aperture
Undulator

Conventional mode of performing ARPES



Angle-mode in modern ARPES analyzers





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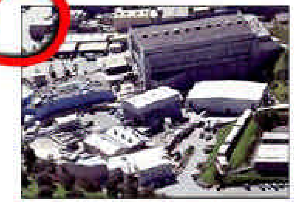

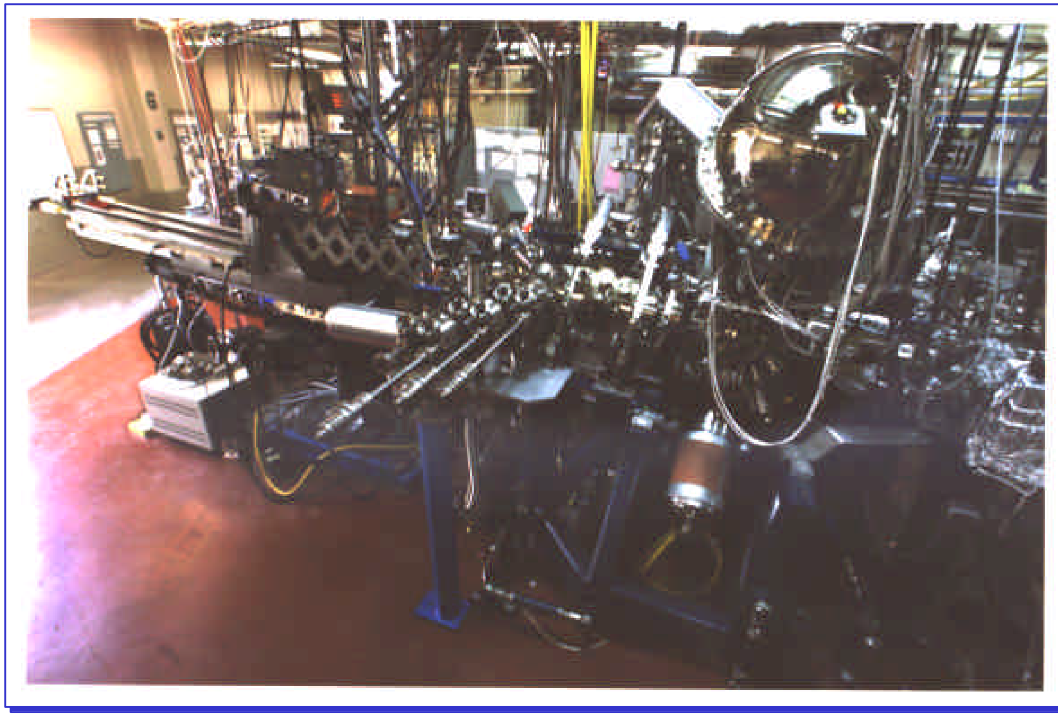

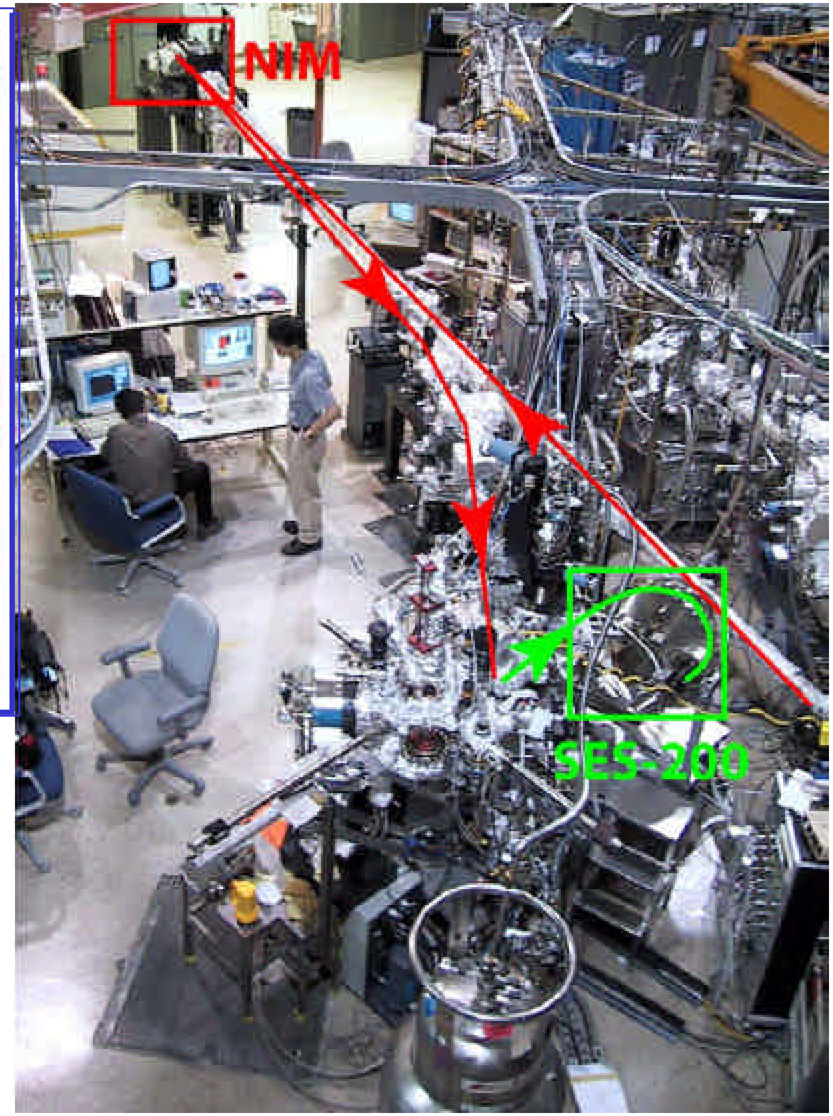

- UHV analysis chamber (10^{-11} Torr)
- 5 axis, He cooled sample manipulator
- Load-Lock transfer system
- Samples may be cleaved in UHV

PHYSICAL REVIEW B VOLUME 21, NUMBER 8 15 APRIL 1980

Angle-resolved photoemission, valence-band dispersions $E(\vec{k})$, and electron and hole lifetimes for GaAs

T.-C. Chiang, J. A. Knapp,* M. Aono,† and D. E. Eastman
IBM Thomas J. Watson Research Center, Yorktown Heights, New York 10598
(Received 3 December 1979)

$$E_f(\vec{k}) = \hbar^2 |\vec{k}|^2/2m + E_0 = \hbar^2(k_\parallel^2 + k_\perp^2)/2m + E_0$$

Final Bloch states. E_o = "bottom of Muffin tin" – starting point for parabolic band dispersions = -9.34 eV for GaAs.

$$E_f(\vec{k}) = E_i(\vec{k}) + h\nu$$

Direct or k-conserving transitions.

$$E_f = E_k + e\Phi$$

eϕ = work function of sample, E_k=kinetic energy

$$\hbar k_\parallel = (2mE_k)^{1/2}\sin\theta$$
$$= [2m(E_i + h\nu - e\Phi)]^{1/2}\sin\theta$$

$$\hbar k_\perp = [2m(E_k\cos^2\theta - V_0)]^{1/2}$$
$$= \{2m[(E_i + h\nu - e\Phi)\cos^2\theta - V_0]\}^{1/2}$$

V_o=E_o- eϕ ="Inner potential". Usually just a fitting parameter.

Normal emission: theta=0 $\hbar k_\parallel = 0$ $\hbar k_\perp = [2m(E_i + e\Phi - E_0)]^{1/2}$

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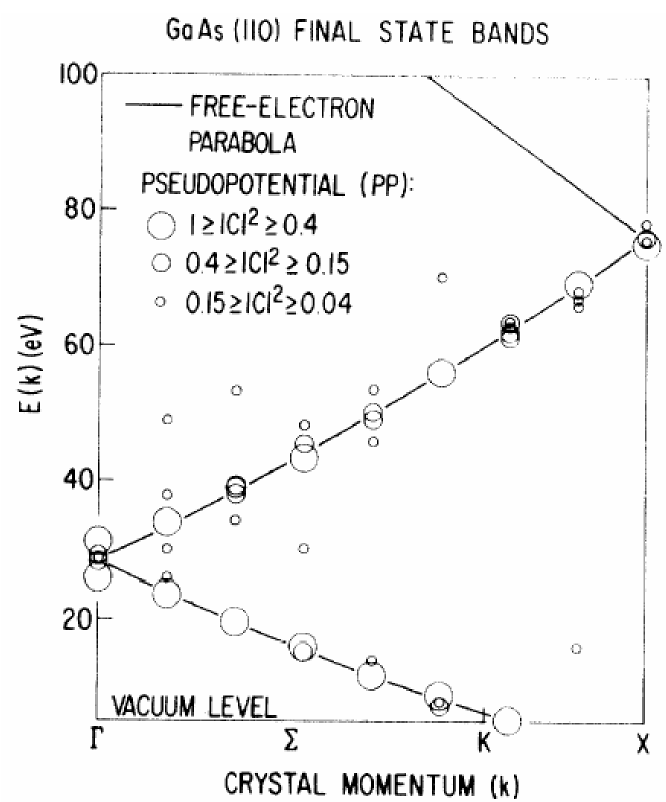

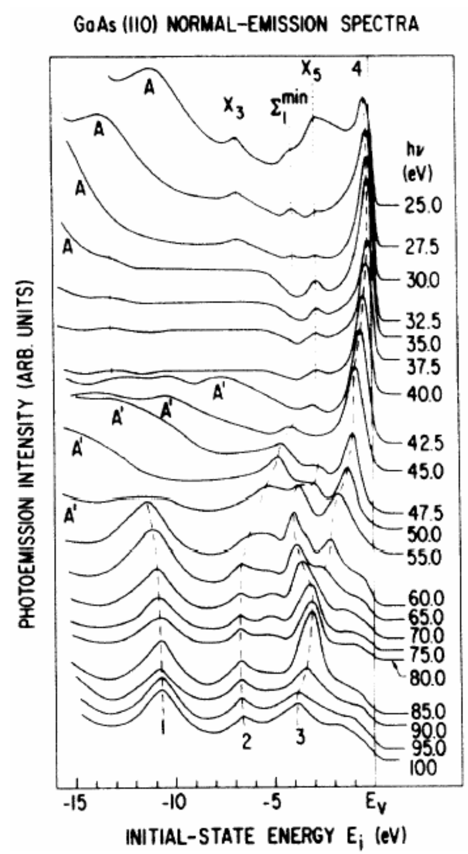

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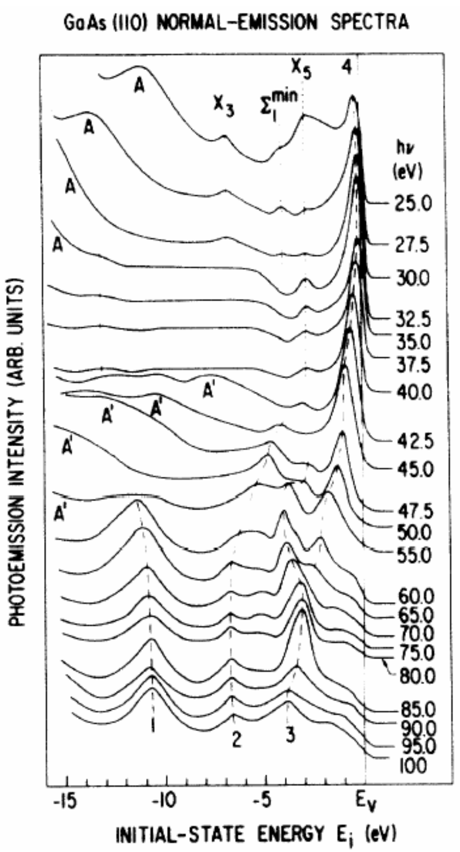

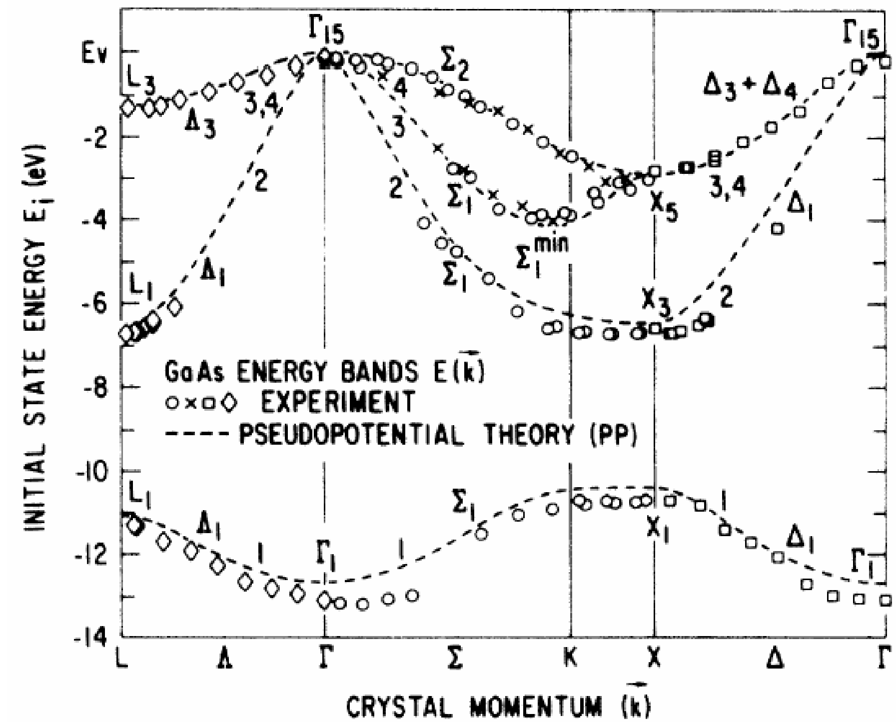

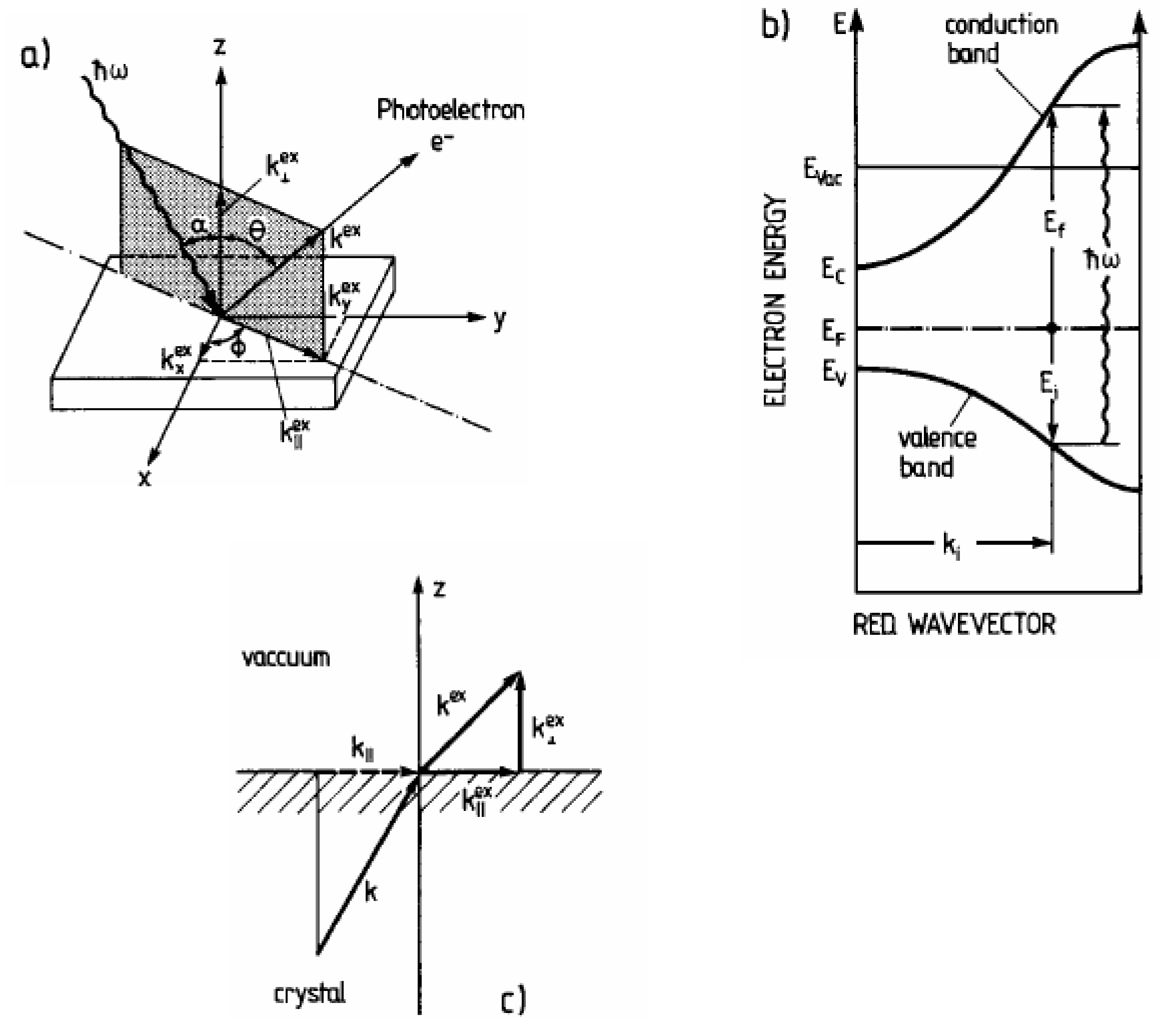

a)
ħω
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Photoelectron
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k∥ex
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b)
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band
EVac
Ef
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EV
Ei
valence
band
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RED. WAVEVECTOR
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vaccuum
kex
k⊥ex
k∥
k∥ex
k
crystal
c)

PHYSICAL REVIEW B VOLUME 47, NUMBER 23 15 JUNE 1993-I

Photoemission linewidths and quasiparticle lifetimes

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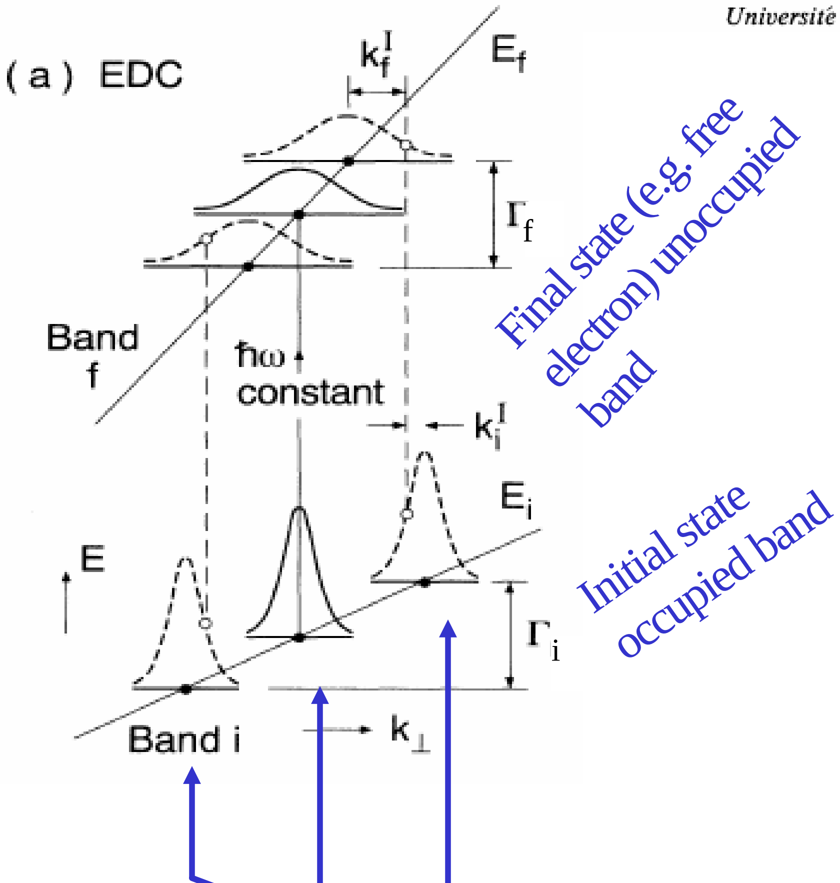


Measured linewidths Γ_m have a contribution from the lifetimes of the initial state (lifetime Γ_i) and final state (lifetime Γ_f).

$$\Gamma_m = \frac{\Gamma_i/|v_{i\perp}| + \Gamma_f/|v_{f\perp}|}{\left| \frac{1}{v_{i\perp}}\left[1 - \frac{m v_{i\|}\sin^2\theta}{\hbar k_\|}\right] - \frac{1}{v_{f\perp}}\left[1 - \frac{m v_{f\|}\sin^2\theta}{\hbar k_\|}\right] \right|}$$

Nearly 2D limit: $v_{i\,perp}$ small. Near isolation of Γ_i.

$$\Gamma_m = \Gamma_i + \left|\frac{v_{i\perp}}{v_{f\perp}}\right| \Gamma_f \ .$$

k_{perp} (and hν) value with half maximum intensity

k_{perp} (and hν) value with maximum intensity (cross section)

2D compounds

- Can ignore k_z dispersion.
- Need not vary photon energy to map out Fermi surface and high symmetry directions.
- Less final state broadening. Intrinsic initial-state linewidths can be studied.

Fermi Surface, Surface States, and Surface Reconstruction in Sr_2RuO_4

A. Damascelli, D. H. Lu, K. M. Shen, N. P. Armitage, F. Ronning, D. L. Feng, C. Kim, and Z.-X. Shen
Department of Physics, Applied Physics and Stanford Synchrotron Radiation Laboratory, Stanford University, Stanford, California 94305

T. Kimura and Y. Tokura
Department of Applied Physics, The University of Tokyo, Tokyo 113-8656, Japan ***and*** *JRCAT, Tsukuba, 305-0046, Japan*

Z. Q. Mao and Y. Maeno
Department of Physics, Kyoto University, Kyoto 606-8502, Japan ***and*** *CREST-JST, Kawagushi, Saitama 332-0012, Japan*

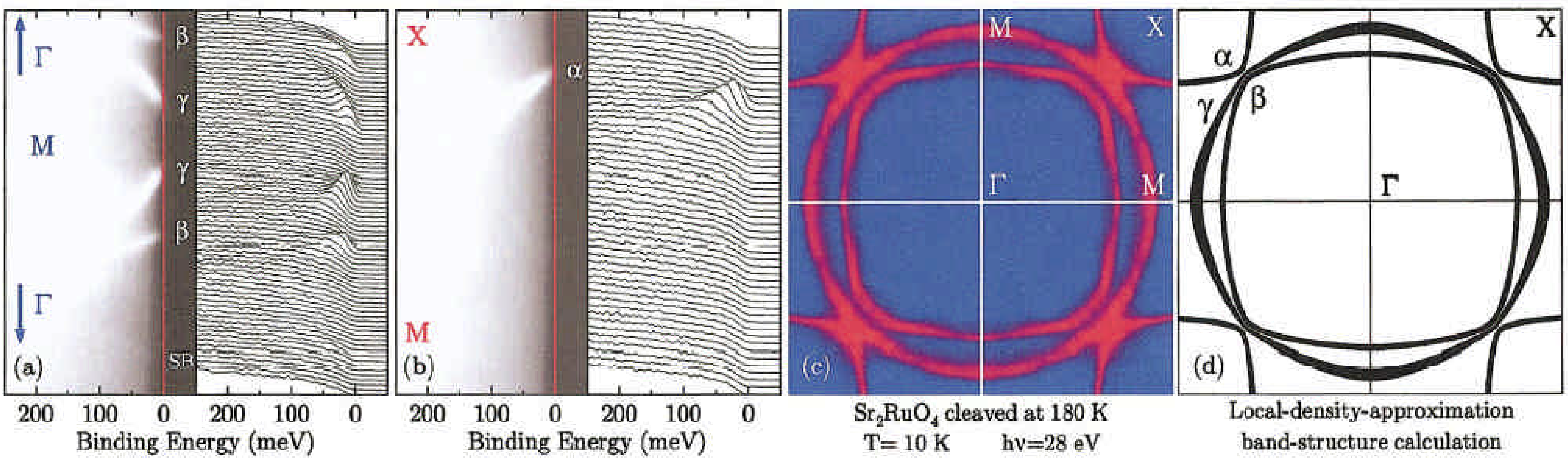
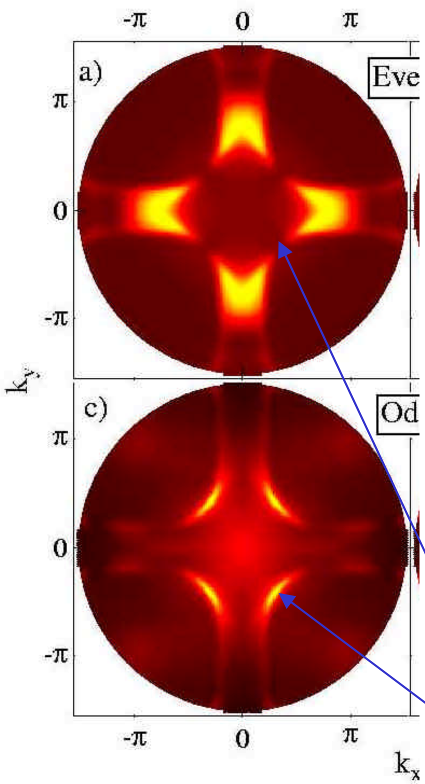


FIG. 9. Photoemission results from Sr_2RuO_4: ARPES spectra and corresponding intensity plot along (a) Γ-*M* and (b) *M*-*X*; (c) measured Fermi surface; (d) calculated Fermi surface (Mazin and Singh, 1997). From Damascelli *et al.*, 2000 (Color).

How different Fermi surface maps emerge in photoemission from Bi2212

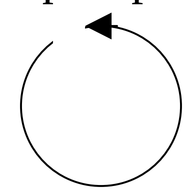
M.C. Asensio[1,2,*], J. Avila[1,2], L. Roca[1,2], A. Tejeda[1,2,*], G.D. Gu[3], M. Lindroos[4,5], R.S. Markiewicz[5], and A. Bansil[5]



Photon E field (Polarizaton direction)

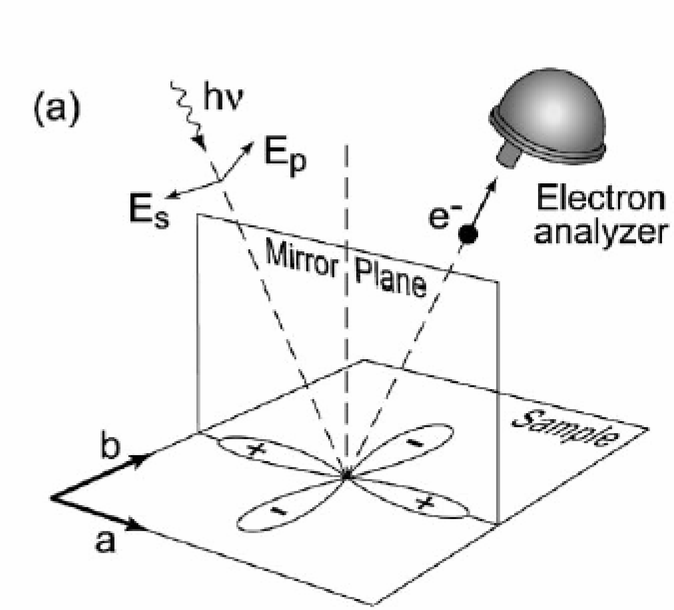
Electron emission direction

Sample spins



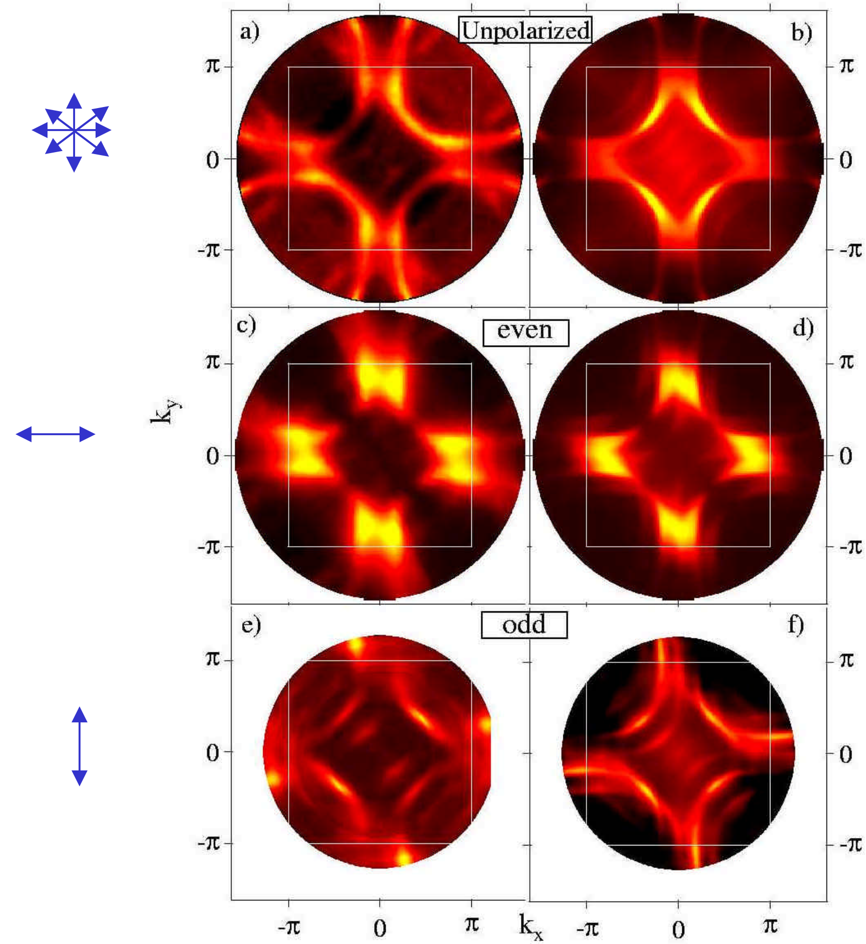
Zeros shifted by 45 degrees.

Symmetry Analysis



$$\langle \phi_f^{\mathbf{k}} | \mathbf{A}\cdot\mathbf{p} | \phi_i^{\mathbf{k}} \rangle \begin{cases} \phi_i^{\mathbf{k}} \;\; \text{even} \;\; \langle + | + | + \rangle \Rightarrow \mathbf{A} \;\; \text{even} \\ \phi_i^{\mathbf{k}} \;\; \text{odd} \;\; \langle + | - | - \rangle \Rightarrow \mathbf{A} \;\; \text{odd.} \end{cases}$$

E field



The matrix element is integrated over all space. The integration axis of interest here is perpendicular to a chosen mirror plane.

If net odd symmetry, then the matrix element integrates to exactly zero.

Matrix Element for Photoemission

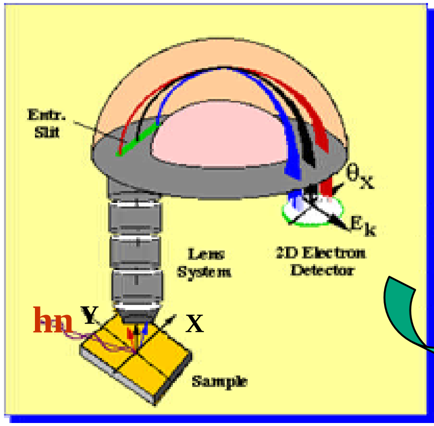
Perturbation Theory gives Fermi's Golden Rule for transition probability

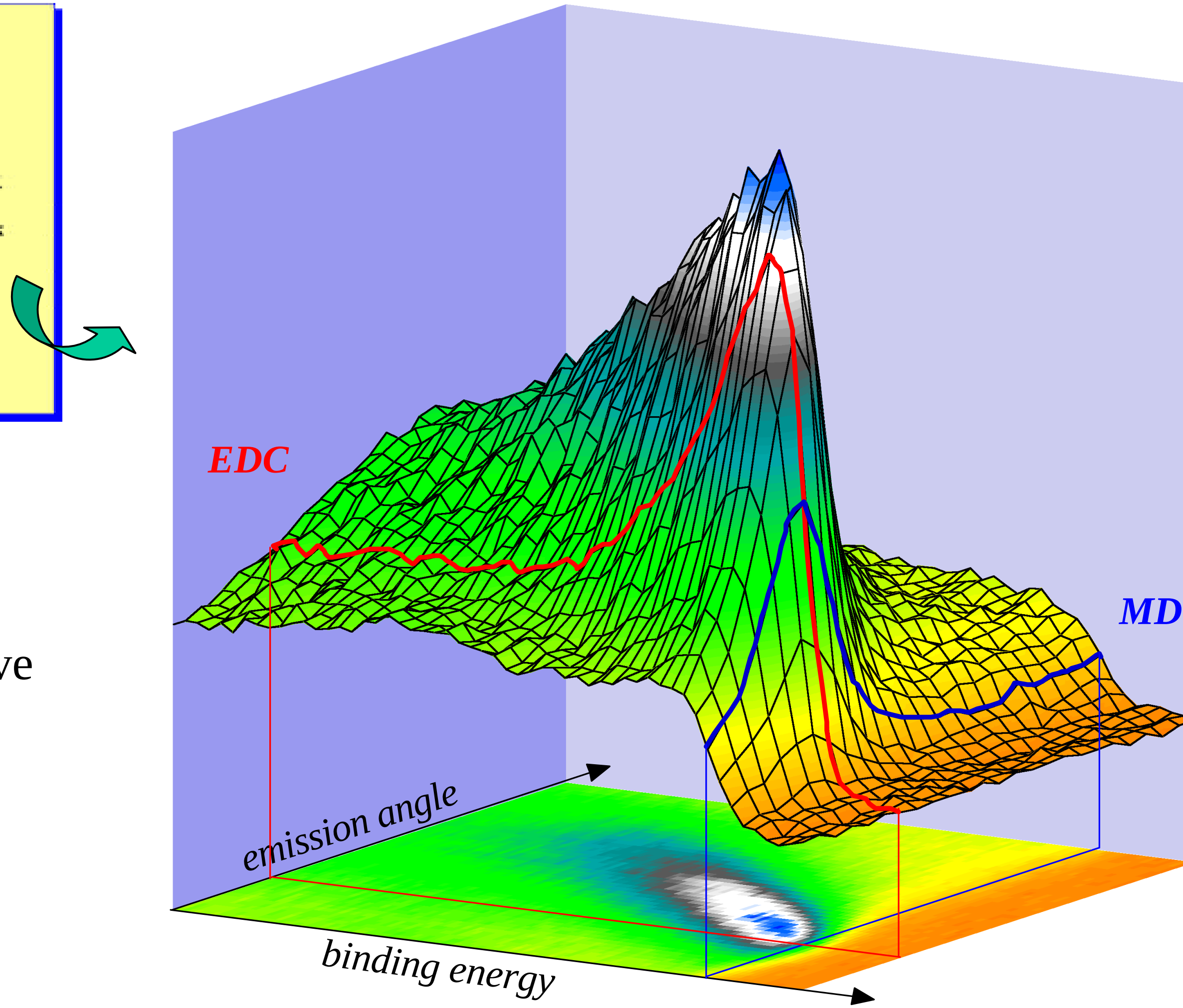
$$w = \frac{2\pi}{\hbar} \left| \langle \psi_f | H_{\text{int}} | \psi_i \rangle \right|^2 \delta(E_f - E_i - \hbar\omega)$$

For dipole allowed transitions,

$$H_{\text{int}} = \frac{e}{mc} \mathbf{A} \cdot \mathbf{p}$$

Two dimensional electron detection





Energy Distribution Curve (EDC)

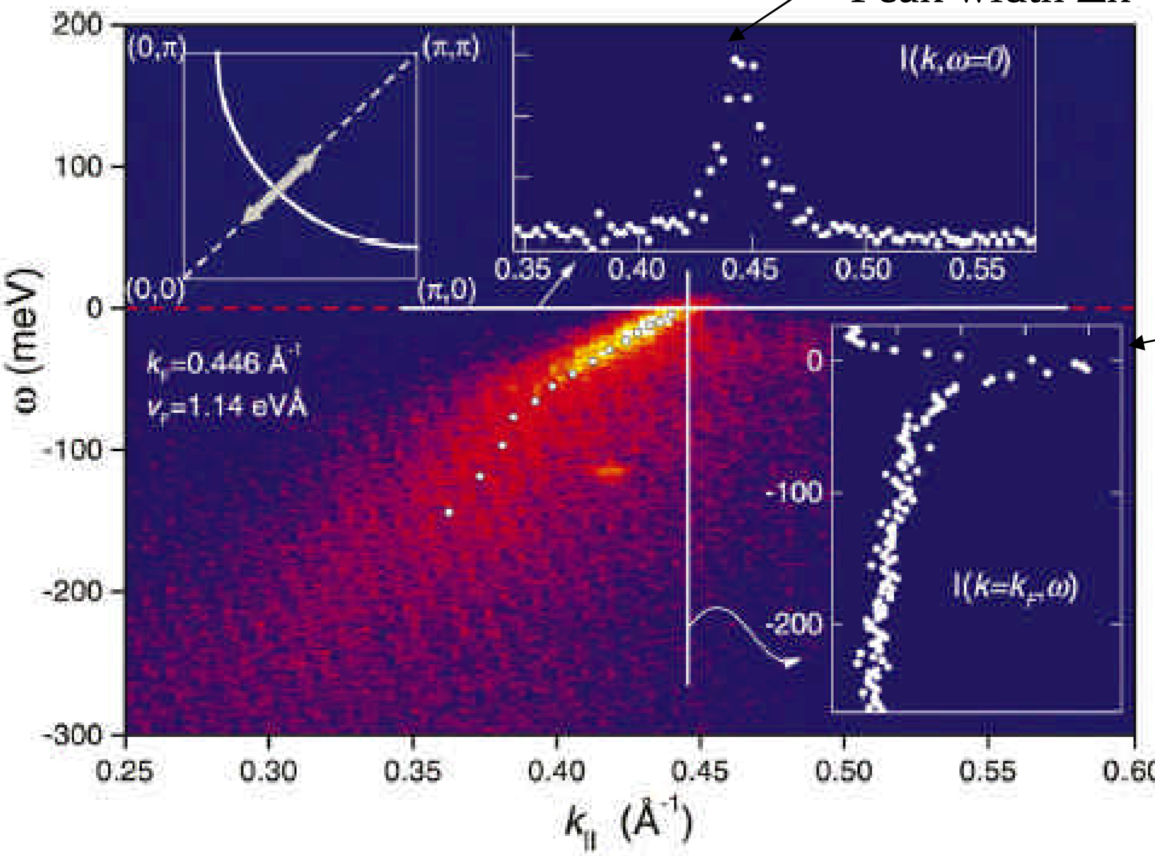
Momentum Distribution Curve (MDC)

A.D. Gromko, University of Colorado Thesis

2D detection on the high Tc superconductor $Bi_2Sr_2CaCu_2O_8$

T. Valla,[1] A. V. Fedorov,[1] P. D. Johnson,[1] B. O. Wells,[1,4]
S. L. Hulbert,[2] Q. Li,[3] G. D. Gu,[5] N. Koshizuka[6]

Momentum Distribution Curve (MDC)
Peak width $\Delta k = 1/l$: l=electron mean free path.



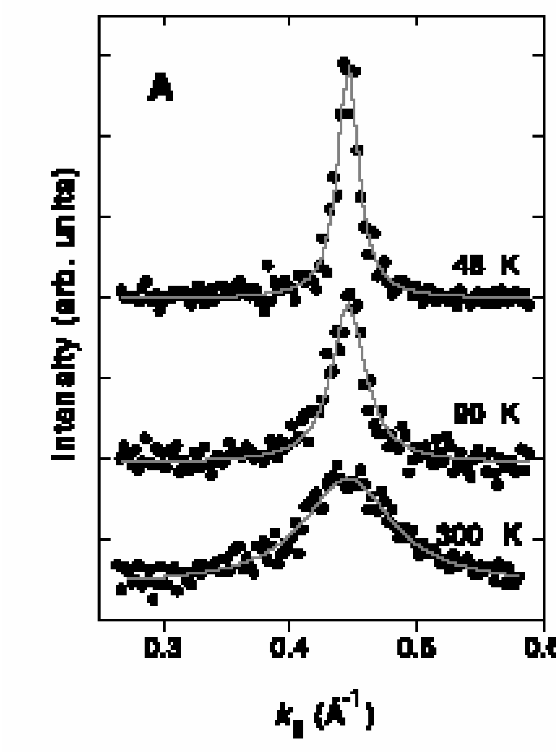
Energy Distribution Curve (EDC)
Peak width $\Delta E = \hbar/\tau$
$1/\tau$=scattering rate
τ=quasiparticle lifetime

$$\Delta E = \Delta k * dE/dk = \Delta k * v$$

MDCs are usually more symmetric than EDCs (simple Lorentzian). → easier to fit

2D detection on the high Tc superconductor $Bi_2Sr_2CaCu_2O_8$

Valla et al., Science (1999)



Lorentzian MDC fits as as a function of temperature.
Broader peaks at higher T →shorter photohole lifetimes.
Origin: Electron-electron scattering? Electron-phonon? Electron-impurity?
The same mechanisms for scattering also affect other probes (optics, transport, etc.).
Also the interactions responsible for the superconducting pairing?

VOLUME 83, NUMBER 10 PHYSICAL REVIEW LETTERS 6 SEPTEMBER 1999

Many-Body Effects in Angle-Resolved Photoemission: Quasiparticle Energy and Lifetime of a Mo(110) Surface State

T. Valla,[1] A. V. Fedorov,[1] P. D. Johnson,[1] and S. L. Hulbert[2]

$$A(\mathbf{k},\omega) \propto \frac{\mathrm{Im}\Sigma(\mathbf{k},\omega)}{[\omega - \varepsilon_{\mathbf{k}} - \mathrm{Re}\Sigma(\mathbf{k},\omega)]^2 + [\mathrm{Im}\Sigma(\mathbf{k},\omega)]^2}$$

“spectral function” = ARPES weight (k,ω)

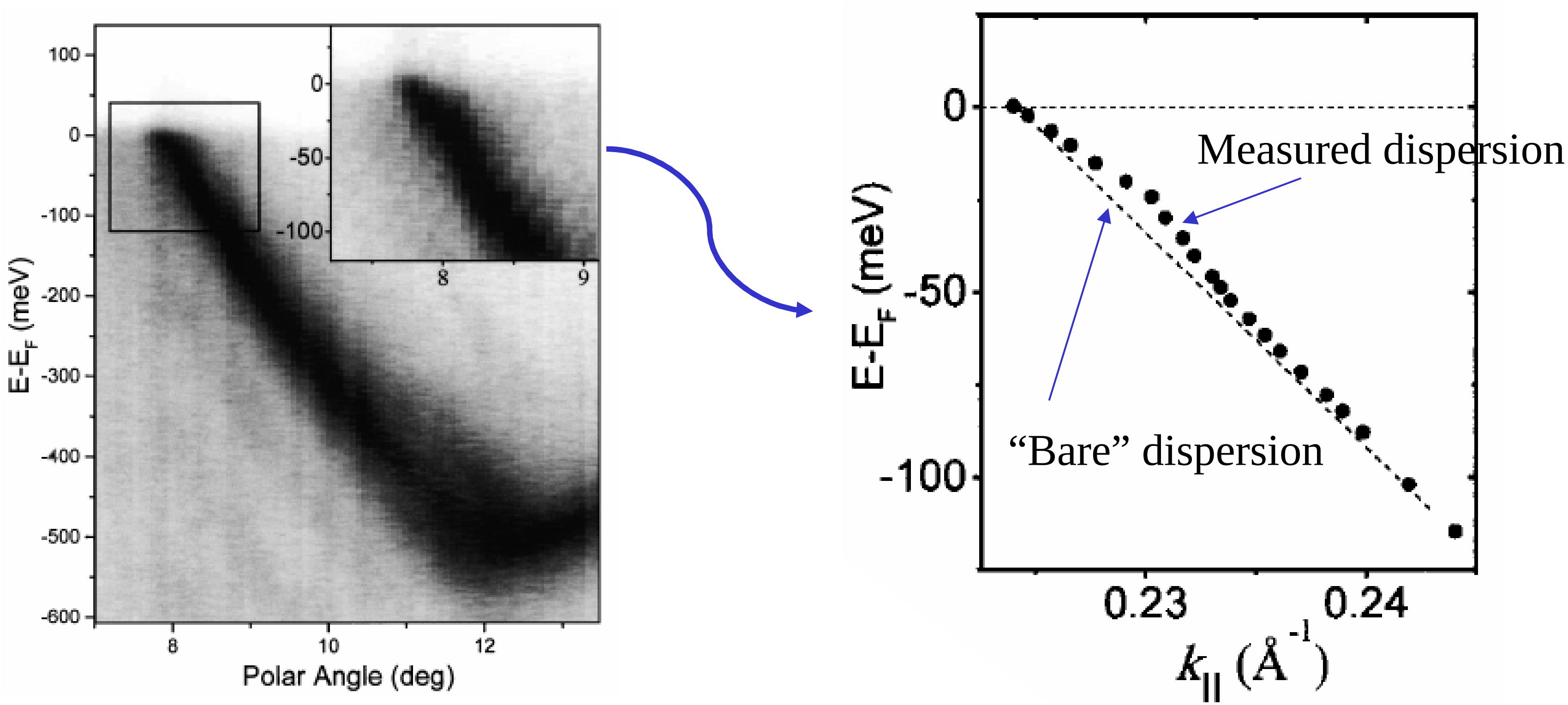


FIG. 1. ARPES intensity plot of the Mo(110) surface recorded along the $\overline{\Gamma}$-$\overline{N}$ line of the surface Brillouin zone at 70 K. Shown in the inset is the spectrum of the region around k_F taken with special attention to the surface cleanliness.

“Kink effect”

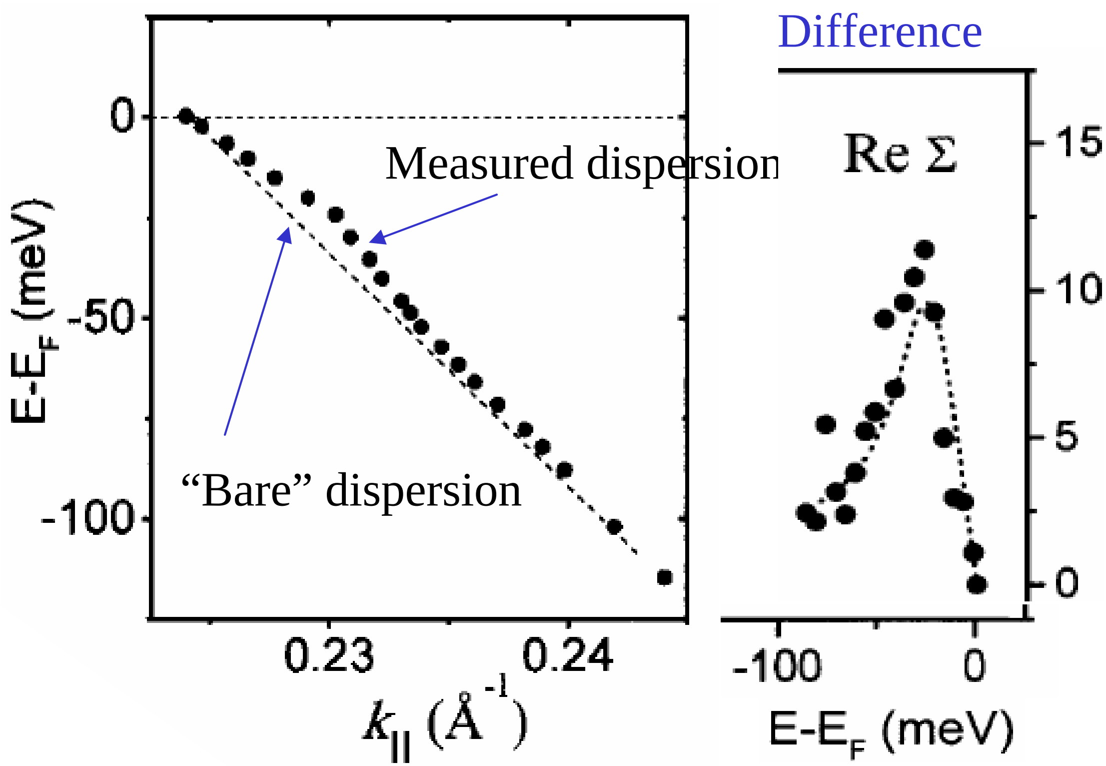
VOLUME 83, NUMBER 10 PHYSICAL REVIEW LETTERS 6 SEPTEMBER 1999

Many-Body Effects in Angle-Resolved Photoemission: Quasiparticle Energy and Lifetime of a Mo(110) Surface State

T. Valla,[1] A. V. Fedorov,[1] P. D. Johnson,[1] and S. L. Hulbert[2]

$$A(\mathbf{k},\omega) \propto \frac{\mathrm{Im}\Sigma(\mathbf{k},\omega)}{[\omega - \varepsilon_{\mathbf{k}} - \mathrm{Re}\Sigma(\mathbf{k},\omega)]^2 + [\mathrm{Im}\Sigma(\mathbf{k},\omega)]^2}$$

“spectral function” = ARPES weight (k,ω)

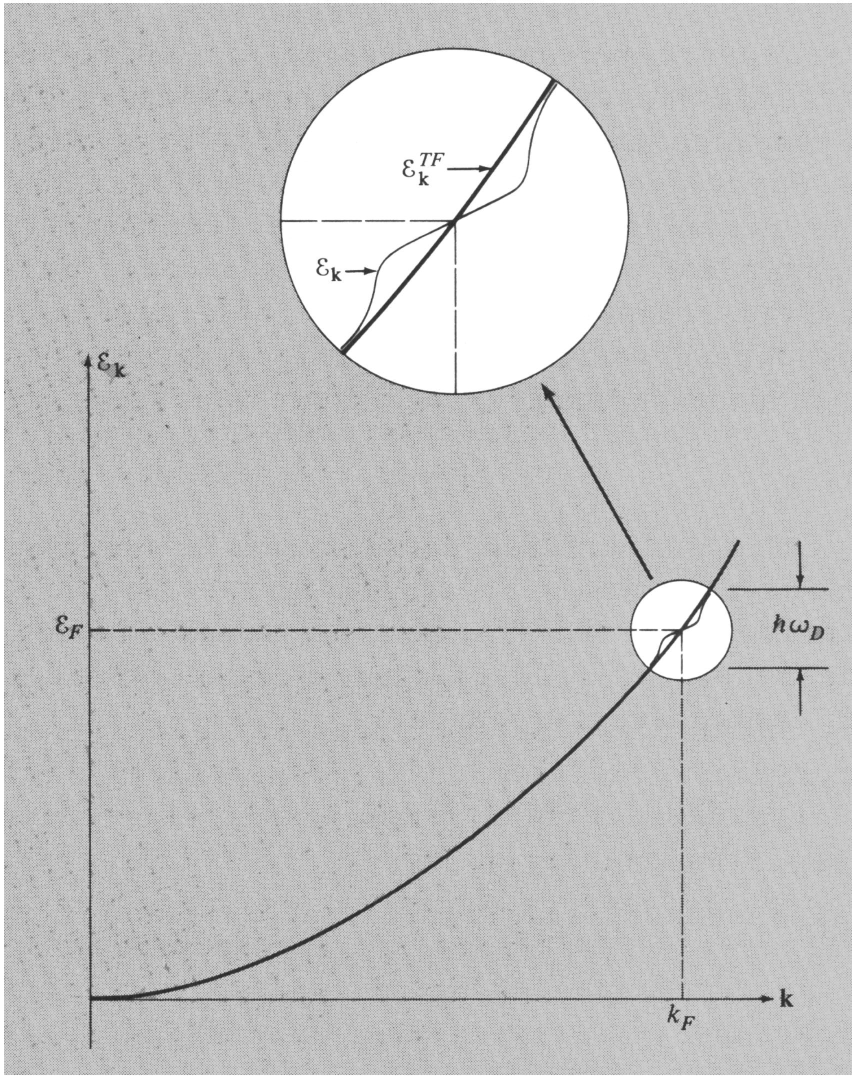


A(k,ω) peaks when $[\omega-\varepsilon_k-\mathrm{Re}\Sigma]=0$ or when
$\omega=\varepsilon_k+\mathrm{Re}\Sigma$
Bare band: $\mathrm{Re}\Sigma=0$
Measured: $\mathrm{Re}\Sigma$=finite.

Σ = electron “self energy”. Here the “kink” is due to electron-phonon scattering. (Phonon lives at kink scale or ~ 30 meV).

Changes in the carrier mass due to electron-phonon coupling only affects the near-E_F states

From Ashcroft and Mermin, Solid State Physics,1976

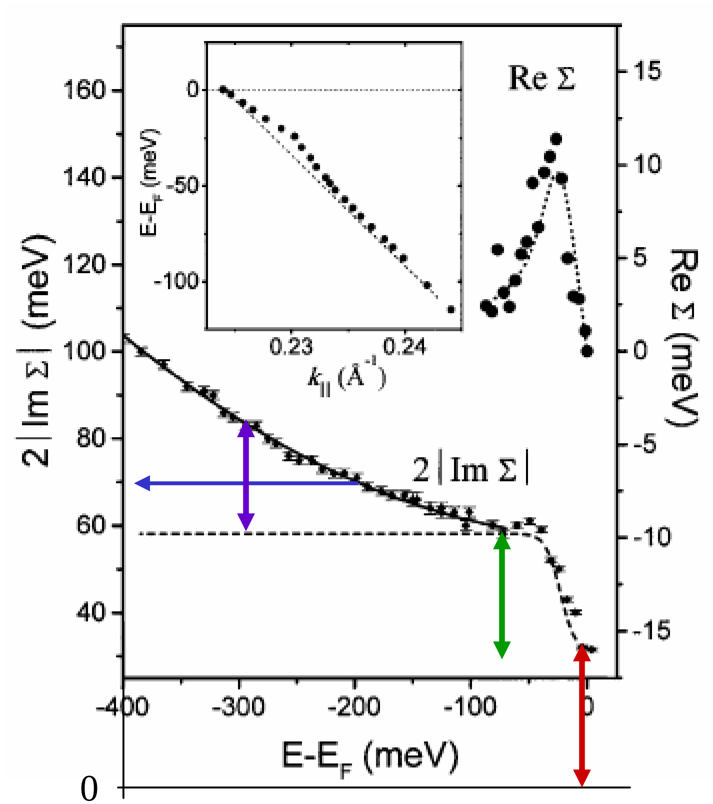


Many-Body Effects in Angle-Resolved Photoemission: Quasiparticle Energy and Lifetime of a Mo(110) Surface State

T. Valla,[1] A. V. Fedorov,[1] P. D. Johnson,[1] and S. L. Hulbert[2]

$$A(\mathbf{k},\omega) \propto \frac{\mathrm{Im}\Sigma(\mathbf{k},\omega)}{[\omega - \varepsilon_{\mathbf{k}} - \mathrm{Re}\Sigma(\mathbf{k},\omega)]^2 + [\mathrm{Im}\Sigma(\mathbf{k},\omega)]^2}$$

FWHM of quasiparticle peak



ImΣ = width of spectral peak
Measurable in the same spectra.

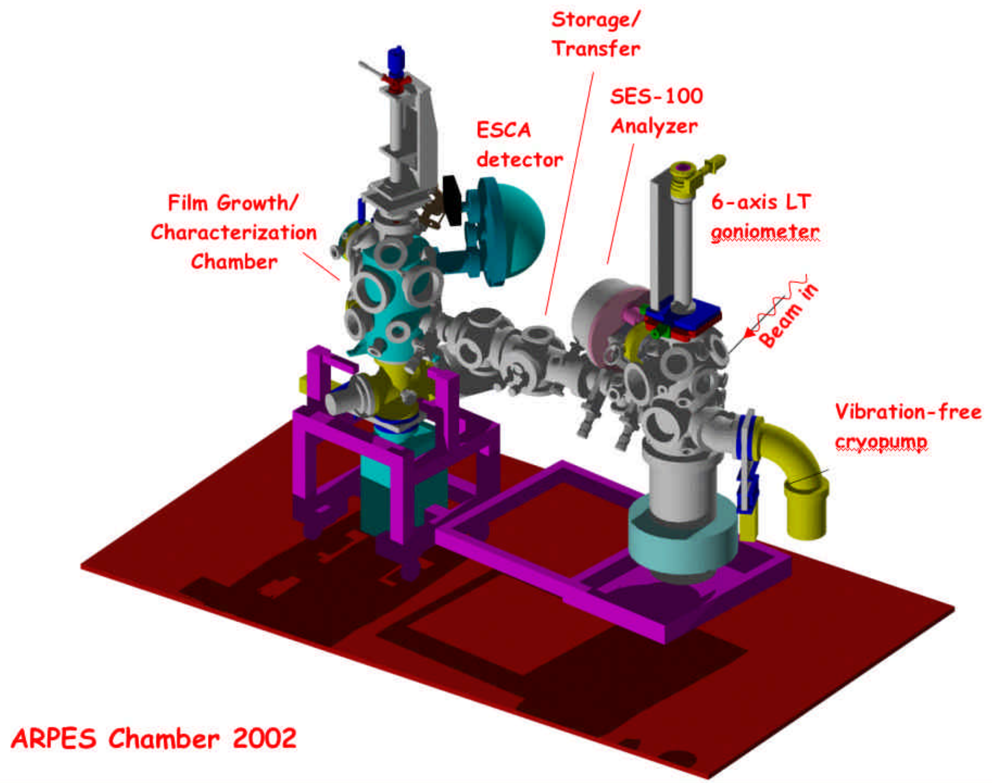
ImΣ and ReΣ related through Kramers-Kronig relations.

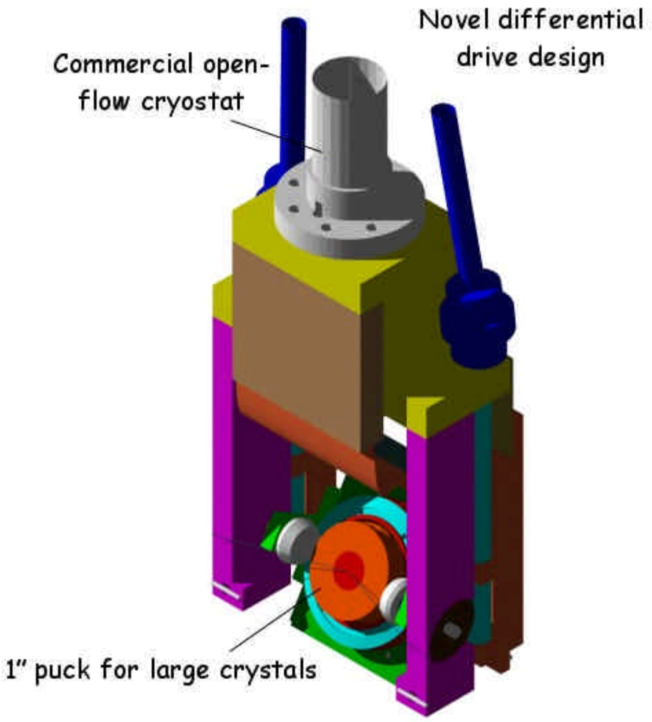
Electron-electron scattering

Coupling to phonons

Impurities, finite resolution, final state effects, etc.

Electronic Structure Factory at beamline 7, ALS





A data set.

Experimental issues:

a) Sample charging – for insulating or weakly insulating samples.
   - Vary the photon flux and look for energy shifts.
   - Raise the sample temperature
   - Electron flood gun to replenish lost electrons

b) Space-charge effect – for high beam intensities.
   - Shifts and broadens peaks.
   - Only an issue for highest beam intensities, highest resolution.
   - Test by adjusting beam intensity.
   - May defocus beam on sample.

c) Sample ageing during measurements
   - Gas chemisorption or physisorption. Warming may regenerate.
   - Gas leaving the sample (e.g. oxides). Low temp helps.
   - Photon beam damage. Lower energy photons may help.
   - Measure quickly!
   - Measure many samples, doing different aspects in a different order.

d) Surface/cleave quality
   - Especially relevant for high angular resolution experiments.
   - Defects/impurities/step edges.
   - Different work functions for different faces.

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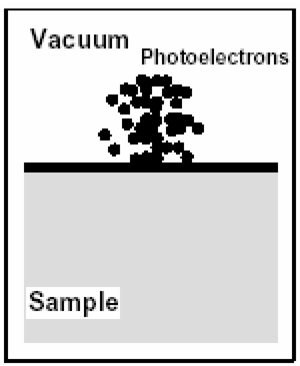
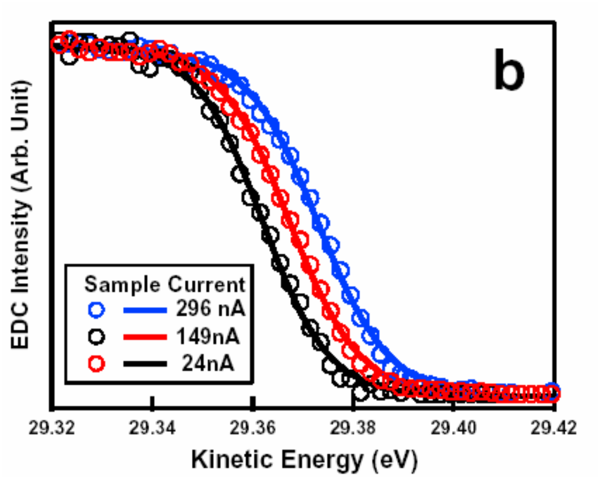
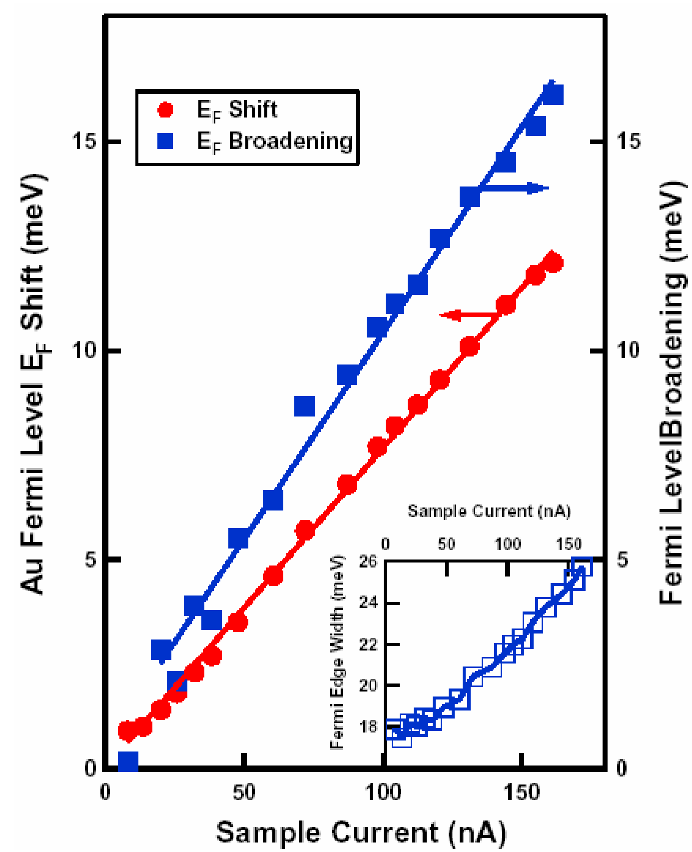
Space charge effect and mirror charge effect in photoemission spectroscopy

X.J. Zhou[a,b,*], B. Wannberg[c], W.L. Yang[a,b], V. Brouet[a,b], Z. Sun[d], J.F. Douglas[d], D. Dessau[d], Z. Hussain[b], Z.-X. Shen[a]

From ALS undulator beamline 10.0.1



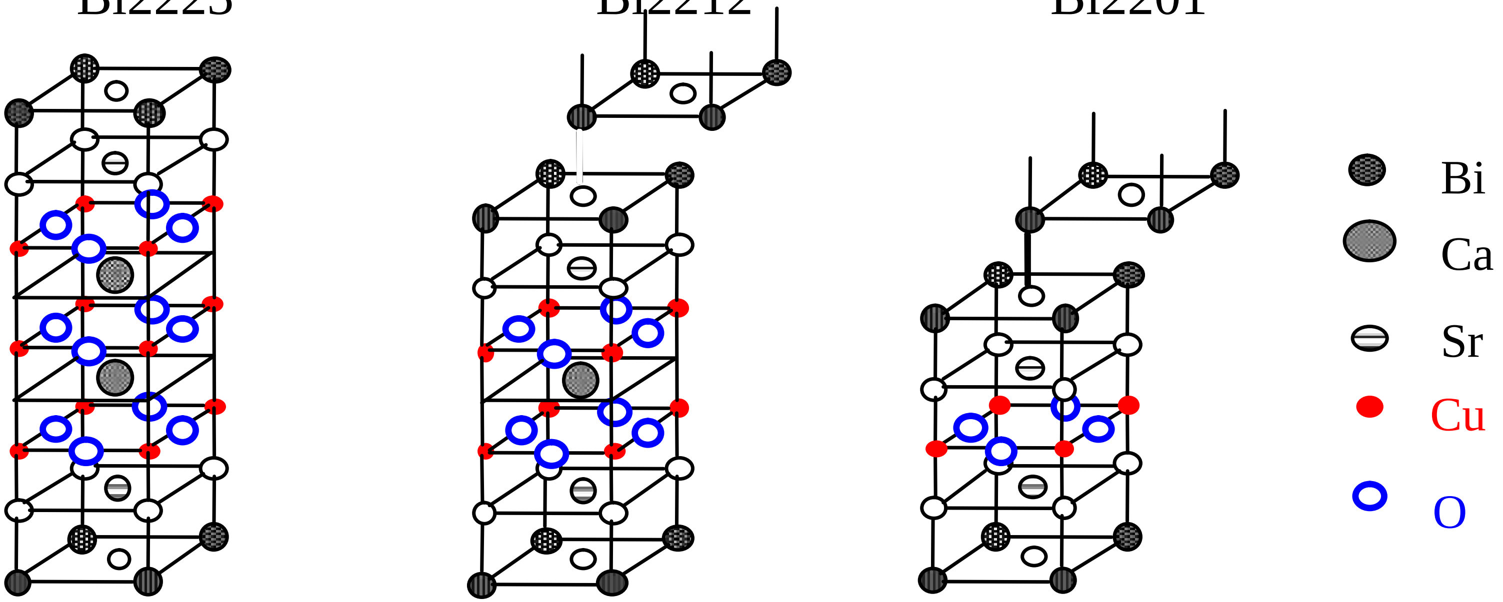




Bi-Sr-Ca-Cu-O family crystal structure

Superconductivity occurs in the CuO_2 planes

$Bi_2Sr_2Ca_2Cu_3O_{10}$ Bi2223 | $Bi_2Sr_2CaCu_2O_8$ Bi2212 | $Bi_2Sr_2CuO_6$ Bi2201



(3 CuO L) T_c= 105 K

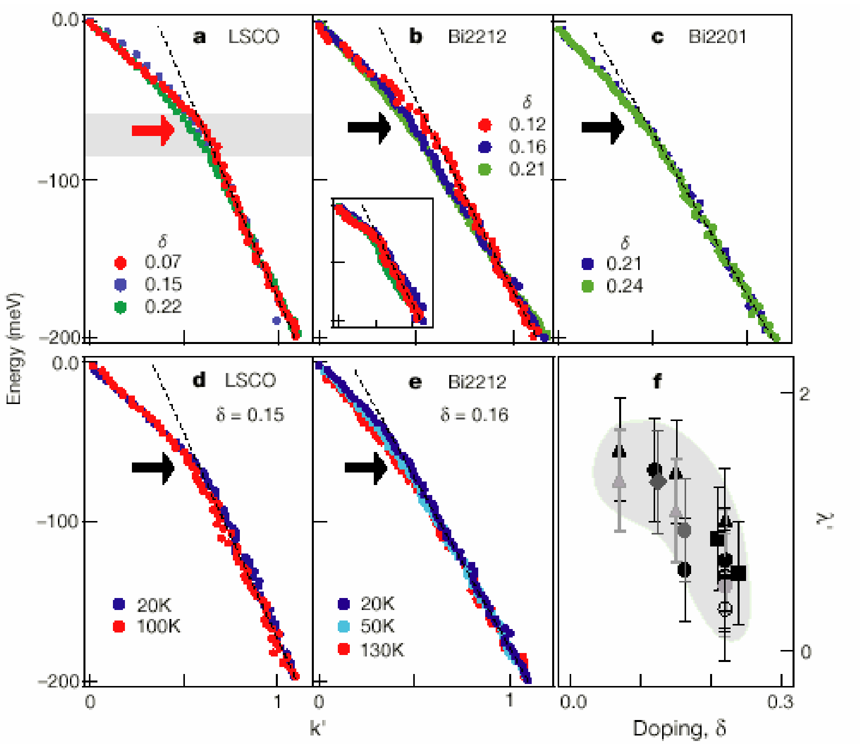
(2 CuO L) T_c= 92 K

(1 CuO L) T_c= 0 ~ 20 K

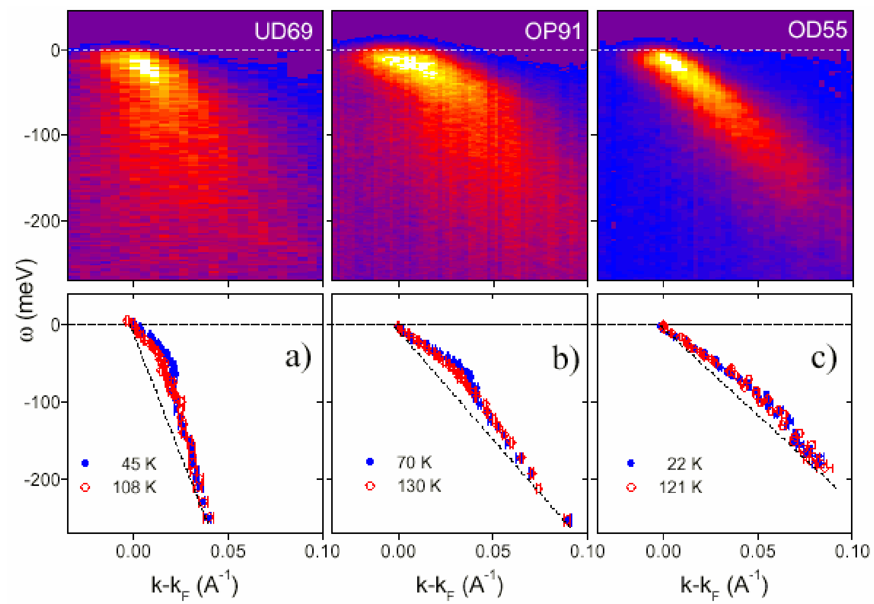
Main compound studied (Bi2212)

Recent ARPES results - kinks in HTSC's (p,p) direction (nodal direction of d-wave gap)

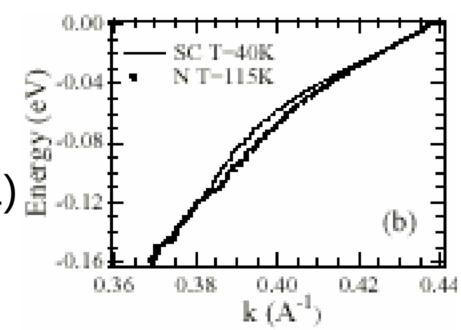
Stanford Group
Lanzara et al.
Nature **412**,510 (2001)



Brookhaven Group
Johnson et al.
cond-mat/0102260 (2001).



Argonne Group
Kaminski et al.
PRL **86**, 1070 (2001)

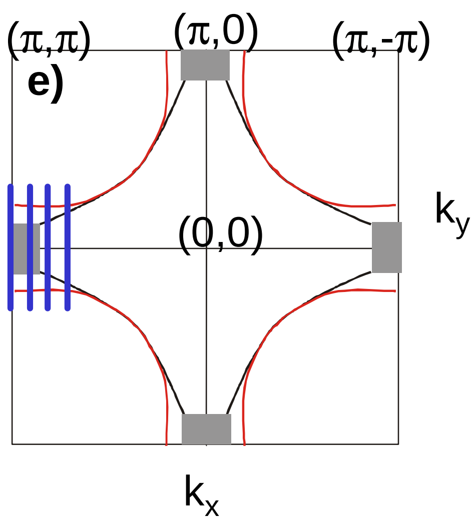


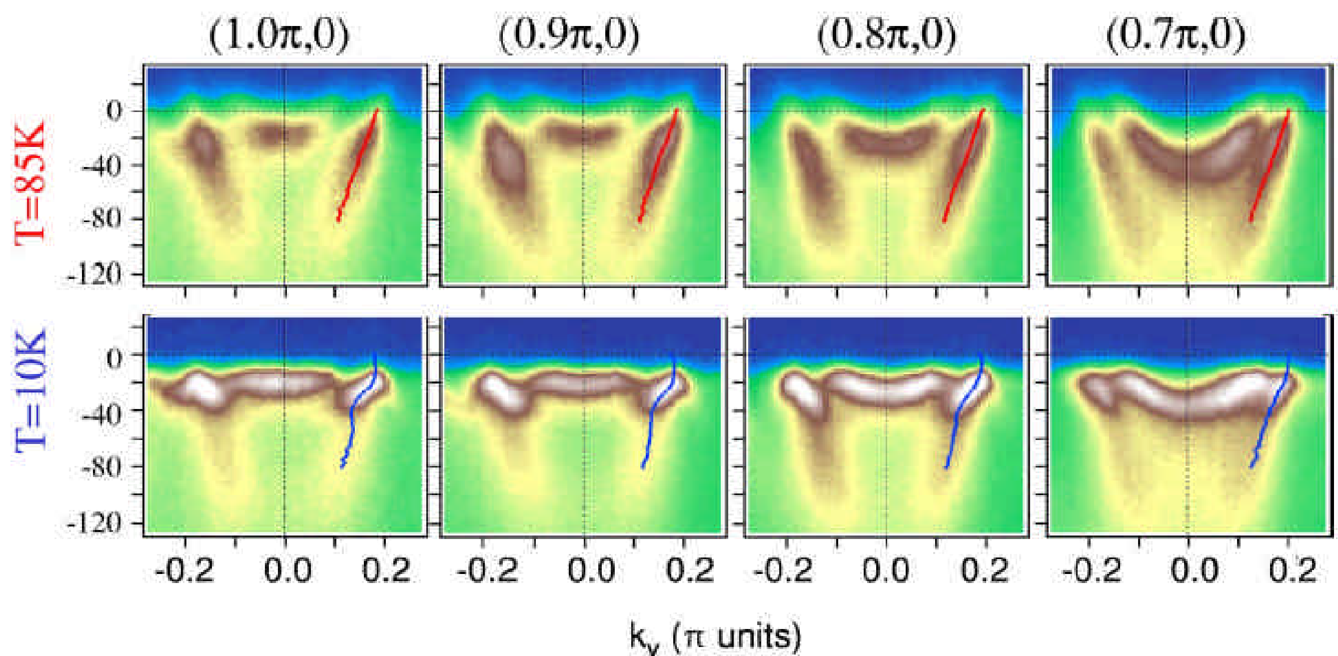
PHYSICAL REVIEW B **68**, 174520 (2003)

Mass-renormalized electronic excitations at (π,0) in the superconducting state of $Bi_2Sr_2CaCu_2O_{8+\delta}$

A. D. Gromko,[1] A. V. Fedorov,[1,2] Y.-D. Chuang,[1,2] J. D. Koralek,[1] Y. Aiura,[3] Y. Yamaguchi,[3] K. Oka,[3] Yoichi Ando,[4] and D. S. Dessau[1]

Kinks are strongly k-dependent.
Kinks are temperature-dependent (strong below T_c).
Difficult for phonons? Magnetic interactions instead?

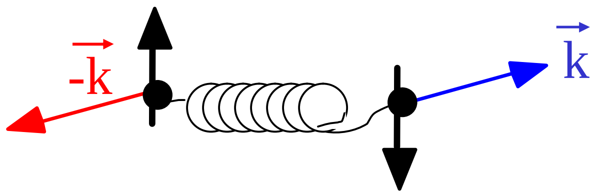




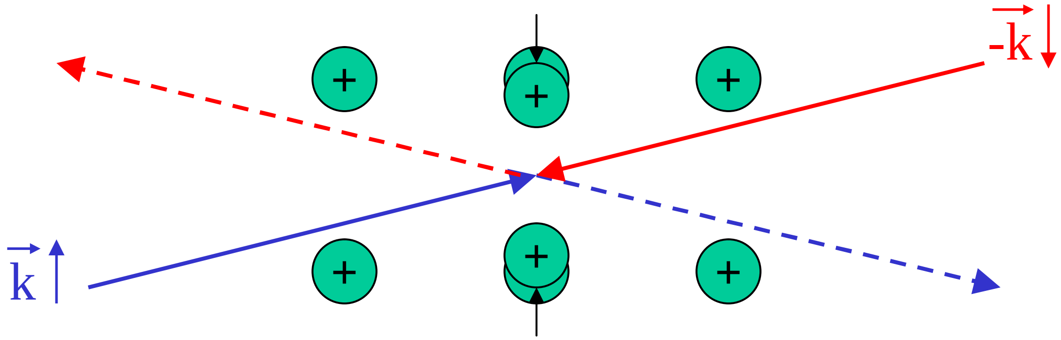
Reason for SC - formation of Cooper Pairs (two electrons form a Boson)

Pairs condense into macroscopic quantum SC state

Cooper Pair

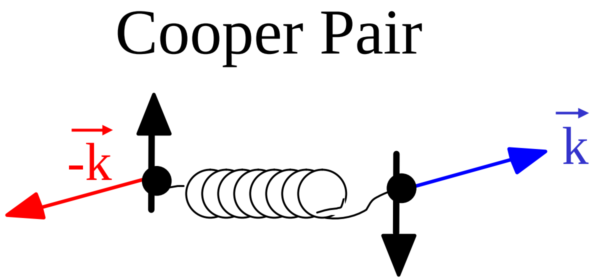


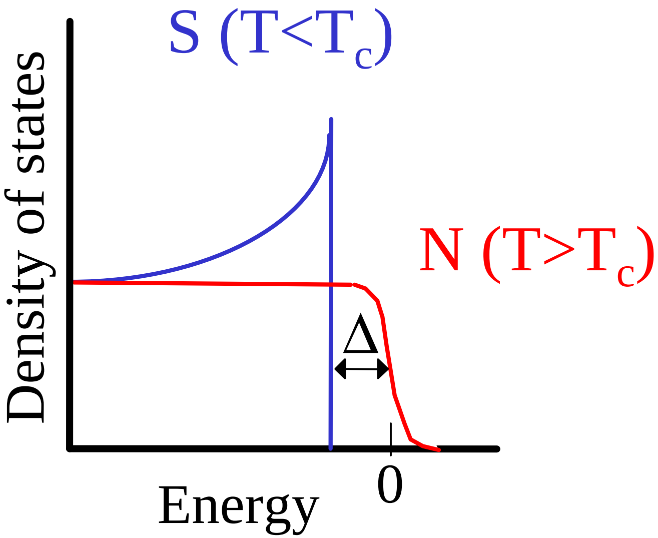
Conventional SC - pairing mediated by electron-phonon interaction



The superconducting gap ?

Energy to remove an electron from system - 1/2 of binding energy of the pair

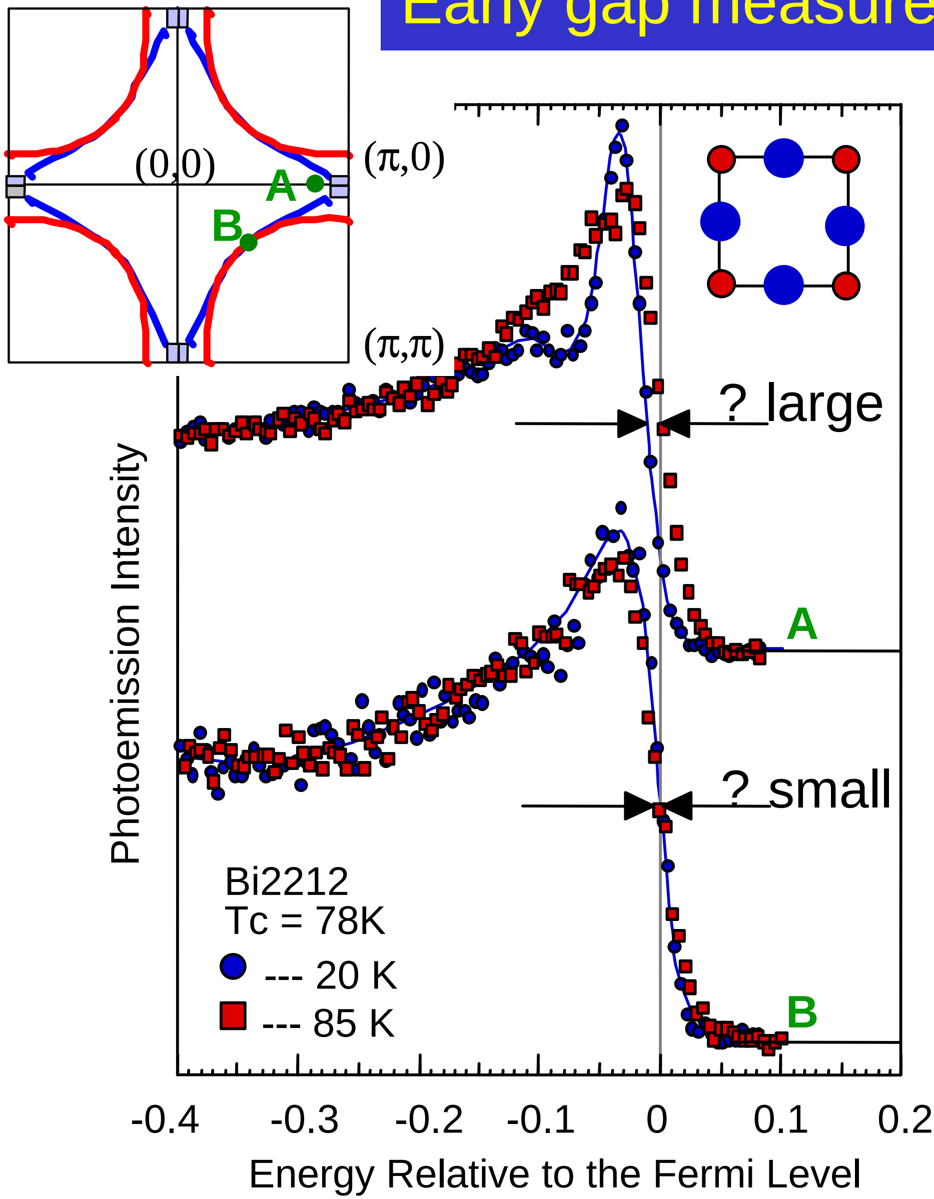




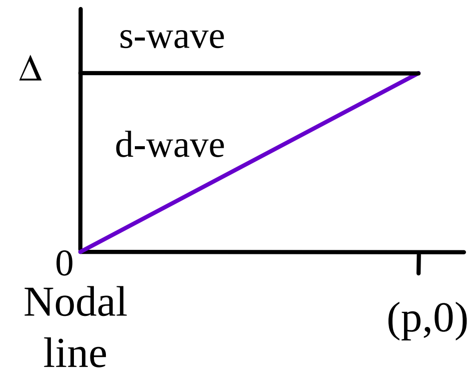
Conventional SCs: T_c~ 0- 30K, Δ ~ 1-2meV

HTSCs: T_c~ 100K, Δ ~ 20-40 meV

Early gap measurements on HTSCs



Gap magnitude maximal at (p,0), minimal or zero along (0,0)-(p,p) “nodal line.”
--> $d_{x^2-y^2}$ symmetry order parameter



Famous peak-dip-hump structure at (p,0).
--> interaction with some mode?

Z.-X. Shen, D.S. Dessau et al., Phys. Rev. Lett. **70**, 1553 (1993)

Superconducting order parameter symmetry

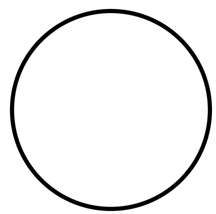
SC gap ? = magnitude of order parameter. Varies as a function of k in a d-wave SC

$$\Psi(r1,\sigma 1;r2,\sigma 2)=\psi(\text{orbital}) \bullet \chi(\text{spin})$$

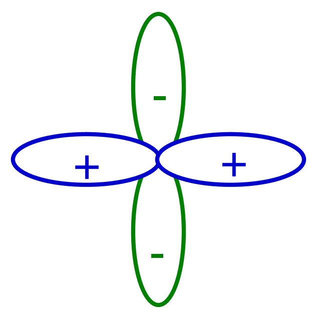
Antisymmetric under exchange

χ(spin) : known to be a singlet (S=0) ↓↑

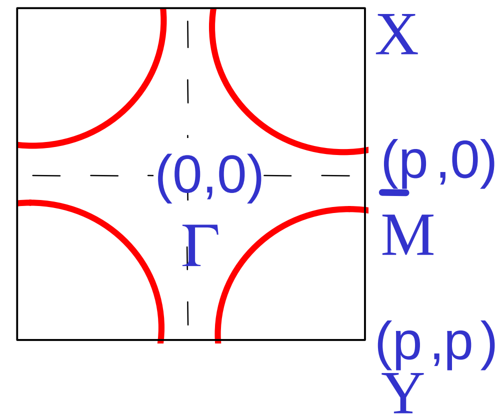
S = 0, l = 0
-- s-wave superconductor
(conventional SC)



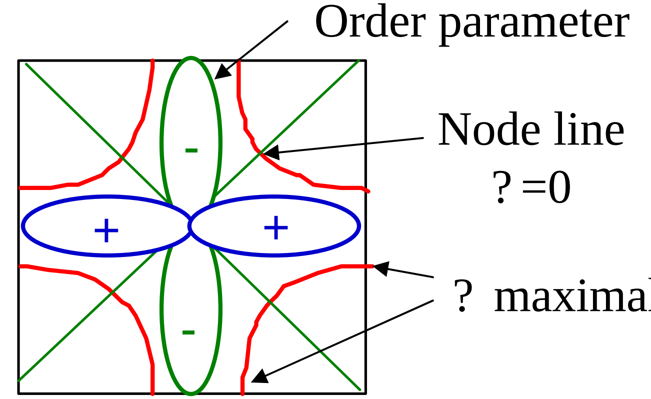
S = 0, l = 2
-- d-wave superconductor
(HTSCs - pretty sure)



Hole-like Fermi Surface



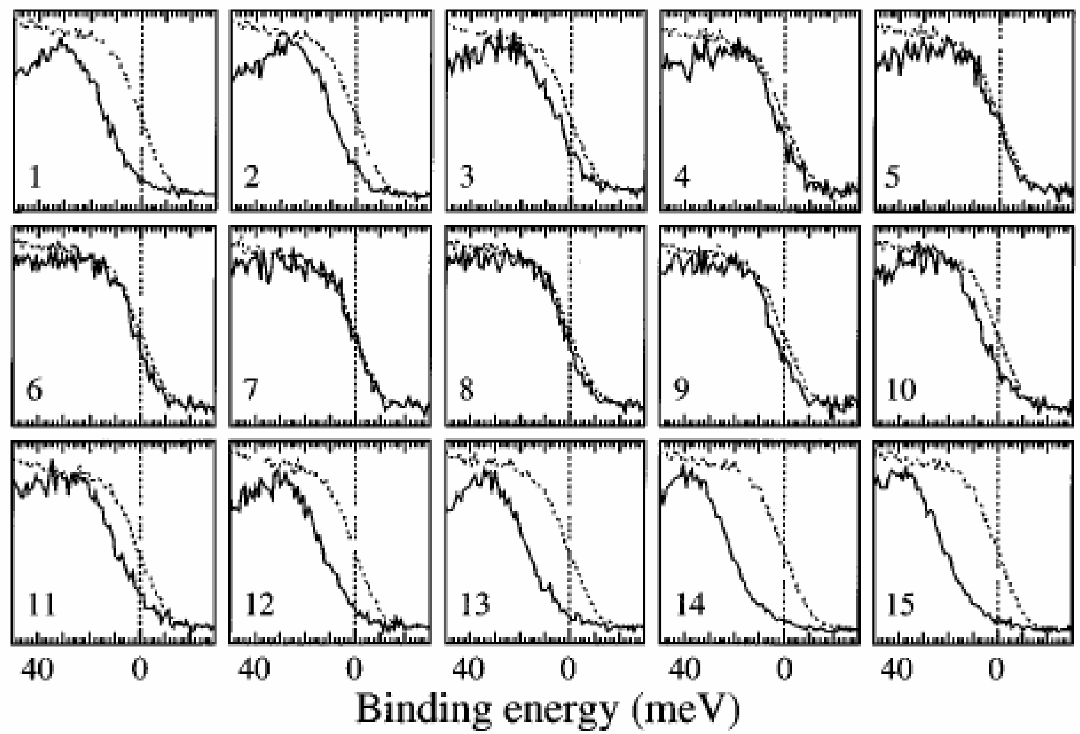
d-wave SC gap - maximal near (p,0)

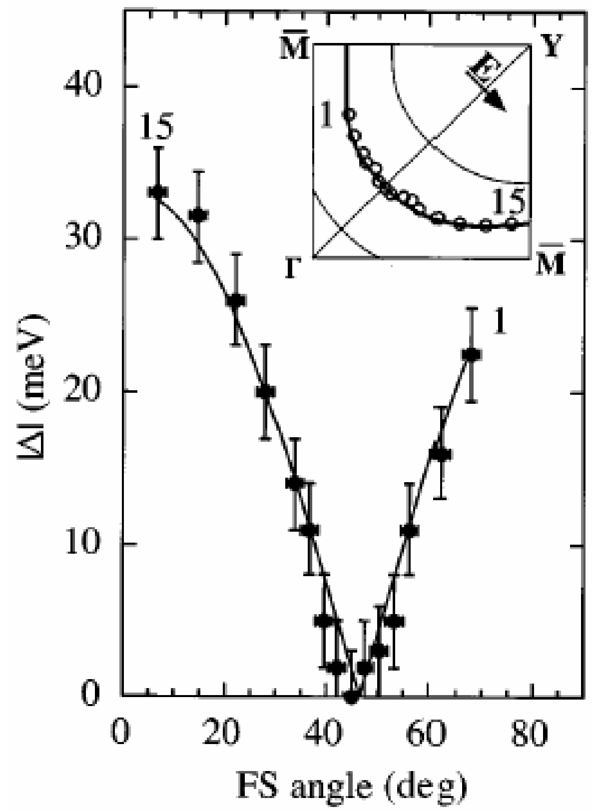


Z-X Shen, D.S. Dessau et al,
PRL **70**, 1553 (1993).

Angle-resolved photoemission spectroscopy study of the superconducting gap anisotropy in $Bi_2Sr_2CaCu_2O_{8+x}$

H. Ding, M.R. Norman, J.C. Campuzano, et al.



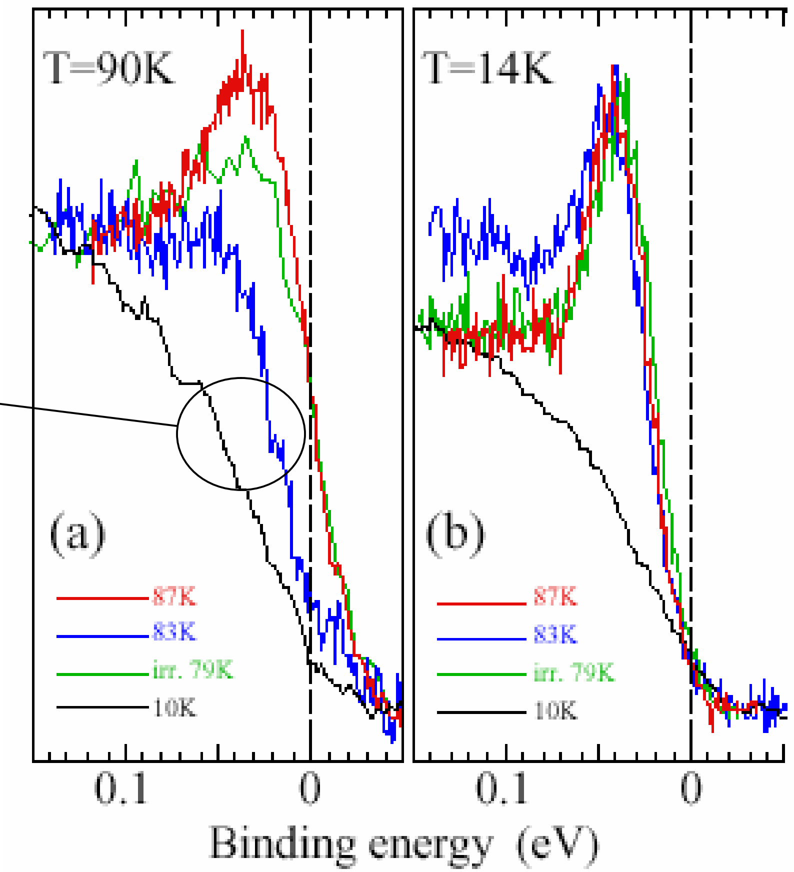


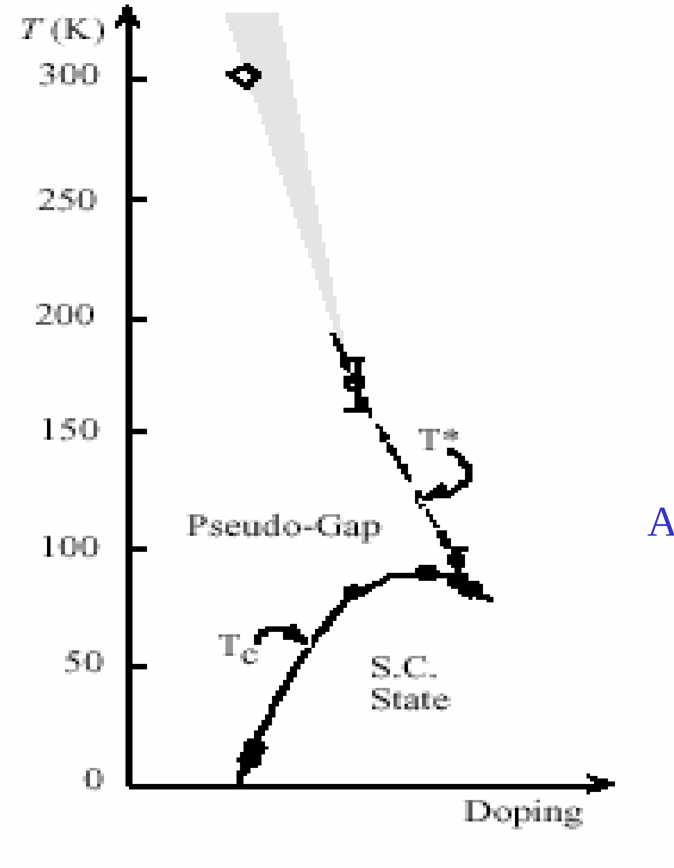
Spectroscopic evidence for a pseudogap in the normal state of underdoped high T_c superconductors

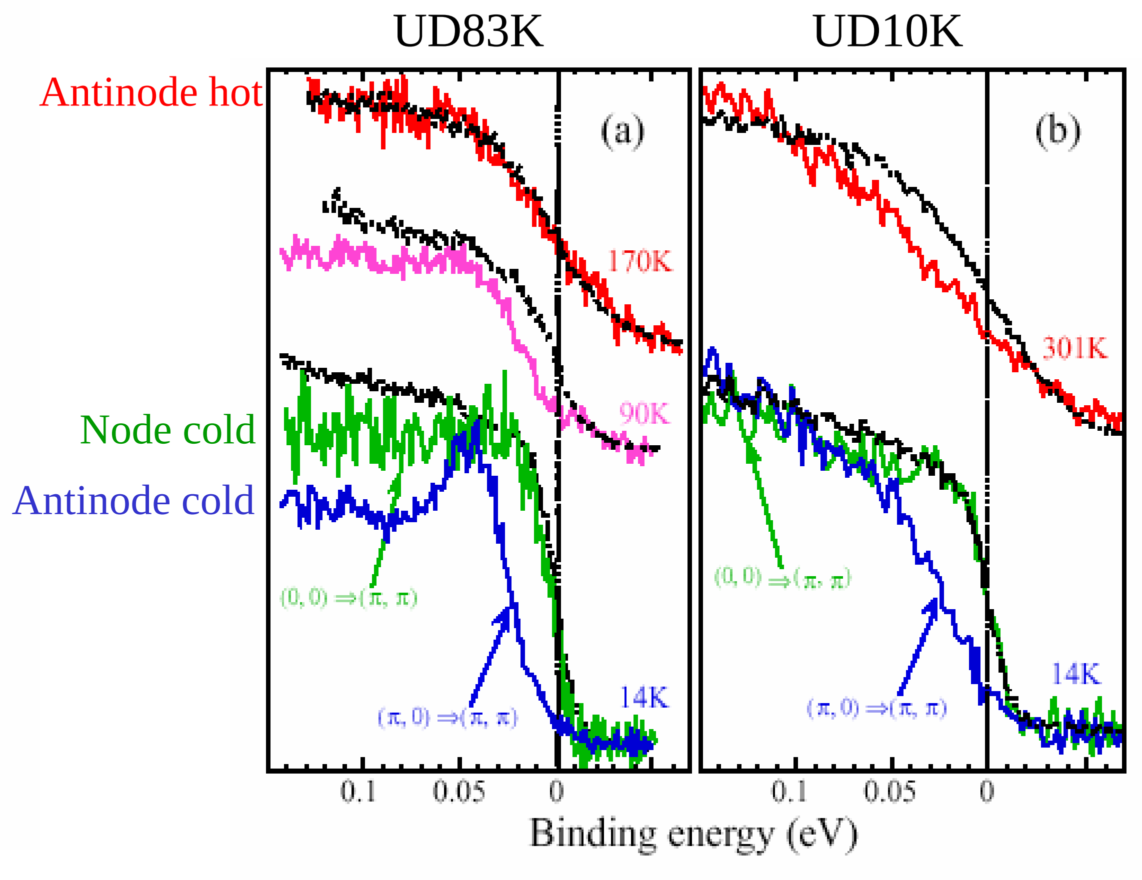
H. Ding et al. Nature ***382****, 51 (1996).*

antinode

UD samples "gapped" even above Tc.







Thought to exist between Tc and T* for UD samples.
Similar magnitude as SC gap.
Similar k-dependence as SC gap (d-wave).
Obtained from leading edge analysis.

Also
M. Norman et al, Nature ***392****, 157(1998).*
D.S. Marshall et al, PRL ***76****, 4841 (1996).*
A.G. Loeser et al. Science ***273****, 325 (1996).*