Angle Resolved Photoemission Spectroscopy

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Photoemission Spectroscopy



Primary electrons – no scattering events. Contain information of the electron spectral function

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

$$E_{kin} = \hbar \omega - \Phi - E_B$$

Angle Resolved Photoemission (ARPES)

Most direct way to measure quantum mechanical "dance" of electrons in a solid.



Interested in critical details of the lowest energy interactions near E_F . \rightarrow Requirement for the highest spectral resolution and sensitivity.



Three Step Model W.E. Spicer



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).

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)

 $E_{f}(\mathbf{\bar{k}}) = \hbar^{2} |\mathbf{\bar{k}}|^{2} / 2m + E_{0} = \hbar^{2} (k_{\parallel}^{2} + k_{\perp}^{2}) / 2m + E_{0}$ Final Bloch states. E_{0} = "bottom of Muffin tin" – starting point for parabolic band dispersions = -9.34 eV for GaAs.

- $E_f(\mathbf{k}) = E_i(\mathbf{k}) + h\nu$ Direct or k-conserving transitions.
- $E_f = E_k + e\Phi$

- $e\phi$ = work function of sample, E_k =kinetic energy
- $\hbar k_{\parallel} = (2mE_{\rm b})^{1/2} \sin\theta$ Projection to parallel component of momentum

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

$$\begin{split} &\hbar k_{\perp} = [2m(E_{k}\cos^{2}\theta - V_{0})]^{1/2} \\ &= \{2m[(E_{i} + h\nu - e\Phi)\cos^{2}\theta - V_{0}]\}^{1/2} \end{split}$$

 $V_o = E_o - e\phi =$ "Inner potential". Usually just a fitting parameter.

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

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.

•Usually much better cleaved surfaces

Fermi Surface, Surface States, and Surface Reconstruction in Sr2RuO4

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FIG. 9. Photoemission results from Sr₂RuO₄: 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).



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GaAs (110) NORMAL-EMISSION SPECTRA



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Photoemission linewidths and quasiparticle lifetimes

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Measured linewidths $\Gamma_{\rm m}$ have a contribution from the lifetimes of the initial state (lifetime $\Gamma_{\rm i}$) and final state (lifetime $\Gamma_{\rm f}$).

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

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

$$\Gamma_m = \Gamma_i + \left| \frac{v_{i1}}{v_{f1}} \right| \Gamma_f \; .$$

 \mathbf{k}_{perp} (and hv) value with half maximum intensity k_{perp} (and hv) value with maximum intensity (cross section)





→3-10 times more bulk sensitive than standard ARPES Very helpful for studies of "bulk" physics.

M. P. Seah and W. A. Dench, Surf. Interface Anal. 1, 2 (1979).

Condensed-Matter Physics

Researchers Turn Up the Heat in Superconductivity Hunt

ARPES dispersion along the nodal line of $Bi_2Sr_2CaCu_2O_8$ T ~ 20K



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J.D. Koralek, D.S.D. et.al, Phys. Rev. Lett. 96, 017005 (2006)

Low photon energy ARPES

- Improved k and E resolution
- Improved bulk sensitivity
- •Reduced background
- •Decreased space-charge effect
- Increased final state lifetimes (less k_perp broadening)

Disadvantages of low-energy ARPES

- Potential issues with breakdown of the sudden-approximation
- Technically more challenging (Electron analyzers don't like low kinetic energy)
- Often a lack of matrix element/photon energy control
- Not many synchrotron beamlines.



CW to few hundred femtosecond, 80-100 MHz rep rate

Resolution and k-space effect



For the same angular resolution, the k resolution at low E is superior.k resolution translates to E widths if the peak is dispersive.

For nodal states & \pm .15 degree angular resolution,

5 meV broadening for hv=6eV, 38 meV for hv=52 eV.

• However – relatively small range of k-space accessible.

Typical synchrotron beamline for ARPES





- •5 or 6 axis, He cooled sample manipulators
- •Load-Lock transfer system
- •Samples may be cleaved in UHV

Matrix Element for Photoemission

Perturbation Theory gives Fermi's Golden Rule for transition probability

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

For dipole allowed transitions,

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

PHYSICAL REVIEW B 69, 094515 (2004)

Bilayer splitting and coherence effects in optimal and underdoped Bi₂Sr₂CaCu₂O_{8+δ}

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 $\langle \phi_{f}^{\mathbf{k}} | \mathbf{A} \cdot \mathbf{p} | \phi_{i}^{\mathbf{k}} \rangle \begin{cases} \phi_{i}^{\mathbf{k}} \text{ even } \langle + | + | + \rangle \Rightarrow \mathbf{A} \text{ even} \\ \phi_{i}^{\mathbf{k}} \text{ odd } \langle + | - | - \rangle \Rightarrow \mathbf{A} \text{ odd.} \end{cases}$

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.



Sudden Approximation – does not appear to be a big issue.

•Old "rule of thumb" - need hv>15-20 eV. Based upon plasmon loss peaks in core level spectra.

•Our Expt: All low energy or "quasiparticle" physics found to be similar in the 6 eV spectra as for the 20-50 eV spectra (velocities, kinks, SC gaps). Deeper (phonon scale) loss peaks also clearly observed, and with similar intensity as in high hv expts.



•Can not fully be in the sudden approximation – not enough energy to excite certain loss features (high energy plasmons, Mott excitations, etc.)

Two dimensional electron detection



A.D. Gromko, University of Colorado Thesis

Resolution effects, gaps, and EDC and MDC dispersions (just following the peaks)



Base Simulation Fermi-Liquid like band dispersing through E_{F.}

Eres σ = .5 meV

 $\Delta = 0 \text{ meV}$

MDC fit is accurate EDC dispersion is not.





T=10K Energy Resolution $\sigma = 2.5 \text{ meV}$





EDC dispersion is accurate MDC is not

2D detection on the high Tc superconductor Bi₂Sr₂CaCu₂O₈

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. Koshizuka⁶



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

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2D detection on the high Tc superconductor Bi₂Sr₂CaCu₂O₈



Lorentzian MDC fits as as a function of temperature.

Broader peaks at higher T \rightarrow shorter photohole lifetimes.

Origin: Electron-electron scattering? Electron-phonon? Electron-impurity? The same mechanisms for scattering also affect other probes (optics, transport, etc.). Are the interactions responsible for the superconducting pairing?

Lorentzian EDC fitting (nodal OP Bi2212)

 $I_{ARPES} = [Lorentzian \times Fermi] + Background$



Nodal Quasiparticles in Bi2212



J.D. Koralek et al. Phys. Rev. Lett. 96, 017005 (2006)



Count rate (arb. units)

Anderson – effect of Gutzwiller projection on ARPES lineshape

$$G(k, \omega) = \iint dx \, dt \, e^{i(kx - \omega t)} t^{-p} / (x - v_F t)$$

$$p=.25(1-x)^2 \text{ x is doping level}$$

$$Intensity = \operatorname{Im}\{G\} = \operatorname{Im}\{\frac{f(\omega/T)}{[(v_F k - \omega) + i\Gamma]^{1-p}}\}$$

$$= f(\omega/T) \frac{\sin[(1-p)\cot^{-1}([\omega - v_F k]/\Gamma)]}{[(\omega - v_F k)^2 + (\Gamma)^2]^{(1-p)/2}}$$

$$f = 1/(1 + e^{\hbar\omega/k_B T})$$
Main empirical difference to FL: reduced background



P.A. Casey, J.D. Koralek, N.C. Plumb, D.S. Dessau, P.W. Anderson, Nature Physics 4, 210 (2008)

SUPERCONDUCTORS The electron shatters



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Coherent vs. Incoherent states

Different operational meanings for a coherent state.

- a) A true Landau quasiparticle
- b) A "sharp" spectral peak near E_F .
- c) A dispersive spectral peak, even if it is broad.



Photoemission and inverse-photoemission spectra of $SrVO_3$ and $CaVO_3$ in the V 3d band region compared with a LDA band-structure calculation of Takegahara (1994). From Morikawa et al., 1995.



Angle-integrated photoemission spectra of $Ca_{1-r}Sr_rVO_3$ (Inoue *et al.*, 1995)

Bi-layer split band structure in x=0.36, 0.38 compounds



Different photon energies are utilized to pick up the A and B bands. Z. Sun et al., Phys. Rev. Lett. 97, 056401 (2006)

Temperature dependence of $(LaSr)Mn_2O_7 x=.4 T_c \sim 130K$



Bandwidth change : .06 eV/1.5 eV = 4%. Much less than the DE prediction of 30%. ==> DE relevant but not key effect.

T. Saitoh et al., PRB (2000)

k-Dependent Electronic Structure, a Large "Ghost" Fermi Surface, and a Pseudogap in a Layered Magnetoresistive Oxide

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Missing Quasiparticles and the Chemical Potential Puzzle in the Doping Evolution of the Cuprate Superconductors

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$Ca_{2-x}Na_xCuO_2Cl_2$



Q: If qp weight Z is strongly doping dependent, why is qp mass \sim constant with doping?

Changes in the carrier mass due to electron-phonon (or other electron-boson) 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. Hulbert2

 $A(\mathbf{k}, \omega) \propto \frac{\mathrm{Im}\Sigma(\mathbf{k}, \omega)}{[\omega - \varepsilon_{\mathbf{k}} - \mathrm{Re}\Sigma(\mathbf{k}, \omega)]^2 + [\mathrm{Im}\Sigma(\mathbf{k}, \omega)]^2} \quad \text{"spectral function"} = \mathrm{ARPES weight} (\mathbf{k}, \omega)$



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.



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

"spectral function" = ARPES weight (k,ω)



A(k, ω) peaks when [ω - ϵ_k -Re Σ]=0 or when $\omega = \epsilon_k + Re\Sigma$ Bare band: Re $\Sigma = 0$ Measured: Re $\Sigma =$ finite.

 Σ = electron "self energy". Here the "kink" is due to electronphonon scattering. (Phonon lives at kink scale or ~ 30 meV).

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



 $Im\Sigma = width of spectral peak$ Measurable in the same spectra.

Im Σ and Re Σ related through Kramers-Kronig relations.

Electron-electron scattering

Coupling to phonons

Impurities, finite resolution, final state effects, etc.

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



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



Isotope substitutions in Bi2212

Way to fingerprint a mode coupling as phonon originated or not



 ^{16}O Throughout this presentation

Raman on our samples:

- near full substitution of ¹⁸O for ¹⁶O
- ~ 3 meV softening with substitution

-Same samples as used by J.Lee and J.C. Davis for isotope studies using STM

Search for a low energy scale (few meV) shift of the nodal kink







Search for a low energy scale (few meV) shift of the kink, version 2

Kink energy analysis method for ARPES widths $(Im\Sigma)$

- a) Using ARPES widths (Im Σ), no assumed background is needed
- b) Take derivative to try to find a well-defined peak



Isotope Effect: Two methods, consistent results





Kink softening of 3.4 ± 0.5 meV



Nodal kink positively fingerprinted as originating from electron-phonon coupling.

H. Iwasawa, J.F. Douglas et al., (submitted)

Superconducting order parameter symmetry

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



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

Angle-resolved photoemission spectroscopy study of the superconducting gap anisotropy in Bi₂Sr₂CaCu₂O_{8+x}

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







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

J.D. Koralek, D.S.D. et al., Phys. Rev. Lett. **96**, 017005 (2006)

Distinct Fermi-Momentum–Dependent Energy Gaps in Deeply Underdoped Bi2212

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Possibility of two gaps:

- a) Near node opens at Tc, gap size tracks Tc
- b) At antinode (the pseudogap) stays open above Tc (to T*)

Competition or cooperation?