Lecture: Introduction to ARPES

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Hongkong

Xingjiang Zhou

National Lab for Superconductivity Institute of Physics Chinese Academy of Sciences Beijing 100190, China

High-Tc Has Changed Landscape of Condensed Matter Physics



Experimental Tools for Superconductivity

DOE Report (2006)

BASIC RESEARCH NEEDS FOR SUPERCONDUCTIVITY

Report of the Basic Energy Sciences Workshop on Superconductivity, May 8-11, 2006



ARPES Techniques and Opportunities

- Angle-Resolved Photoemission Spectroscopy (ARPES). Wavelike quantum states of the electrons are defined in momentum space (k-space). ARPES allows direct determination of the complete momentum-space electronic structure, A(k, E), with remarkable energy and momentum resolution.
- Spectroscopic Imaging-Scanning Tunneling Microscopy (SI-STM). This is the complementary technique to ARPES that allows mapping of the energy-resolved quantum states in real space (*r*-space) with atomic resolution and yet over large sample areas.
- *Microwave/terahertz/infrared/optical spectroscopies*. These probe the electronic excitations and charge dynamics in both the frequency and time domains. This information is the key to understanding the dynamical interactions of the electrons.
- *Resonant elastic and inelastic x-ray spectroscopy*. Resonant elastic and inelastic x-ray scattering can now reveal spin and charge density waves and superlattices with tiny modulation amplitudes. This information is critically important for understanding spatially periodic electronic states of matter.
- *Neutron Scattering (NS).* High-intensity NS for example, from the Spallation Neutron Source will allow precision measurements of both magnetic ground states and the complete spectrum of magnetic excitations in high-temperature and exotic superconductors.
- $NMR/NQR/\mu SR$. NMR measures spin dynamics, NQR measures the charge heterogeneity and dynamics, and μSR measures nanoscale variation in local magnetic field strength. These are essentially local spin/charge probes, but without imaging capabilities.

ARPES on High Temperature Cuprate Superconductors

d-wave **Many-Body Effects Pseudogap** Superconducting gap ARPES on Superconducting gap **ARPES on Pseudogap ARPES** on Kink **Μ**(π, 0) d-wave-like symmetry -0.05 30 $(0,\pi)$ (π,π) Relative LEM (meV) () Э_-0.10 Ц Photoemission Intensity **X**= 20 • 0.03 0.05 (0,0) $(\pi, 0)$ ш **X 0.063** 0.075 -0.15 0.10 10 **0.12** ⊠ 0.15 **v** 0.18 ● UD10 Ding et al. 0.22 • UD46 Harris et al. 0.30 O UD78 Harris et al. Bi2212 UD78 Loeser et al. 0) Tc = 78K 0.06 0.04 0.02 k - k_F (A⁻¹) --- 20 K 15 30 45 0 -- 85 K Fermi surface angle (deg) -0.2 -0.1 -0.3 0 0.1 0.2 -0.4Energy Relative to the Fermi Level (eV) A. Lanzara et al., Nature 412(2001)510. Loeser et al., Science 273(1996)325. Z.-X. Shen et al., Phys. Rev.Lett.70(1993)1553. X. J. Zhou et al., Nature 423(2003)398. Ding et al., Nature 382(1996) 51.

ARPES on Topological Insulators



Y. Xia et al., Nature Phys. 5(2009)398.



Y. L. Chen et al., Science 325 (2009)178.

ARPES on Various Materials



Science 300(2003)303



Nature 438(2005)647





Nature 426(2003)540

Quantum Well



Nature 398(1999)132

Quasicrystal



Magnetic Materials



Nature 438(2005)474

CDW



Phys. Rev. Lett. 93(2004)126405

Content

>What is ARPES?

Brief history, ARPES process, ARPES setup (Light sources, electron analyzer), ARPES resolutions, Matrix element effect

>What can ARPES do?

Fermi surface mapping, Gap measurement Many-body effects

>Latest and future development of ARPES

(Laser ARPES, spin-resolved, time-resolved, spatially-resolved)

Brief History of ARPES

Principle of Photoemission—Photoelectric Effect

Discovery (1887) German Physicist:

Hertz



Heinrich Rudolf Hertz (1857-1894) Photoelectric Effect



Explanation (1905) :



The Nobel Prize in Physics 1921



Albert Einstein (1879-1955)

"For his services to Theoretical Physics, and especially for his discovery of the law of <u>The Photoelectric Effect</u>"

Nobel Prizes in Photoemission Spectroscopy



The Nobel Prize in Physics 1924



Karl Manne

(1886 - 1978)

Georg

Siegbahn

"for his
discoveries
and research
in the field of
X-ray
spectroscopy "

"The X-ray spectra and the structure of the atoms"





1981

Kai M. Siegbahn (1918-2007) "for his contribution to the development of high-resolution electron spectroscopy "

The Nobel Prize in Physics

"Electron Spectroscopy for Atoms, Molecules and Condensed Matter"

Classifications of Photoemission Techniques

Photoemission	Angle- Integraed	Angle- Resolved
Spin-integrated	Ε	E , k
Spin-resolved	E , s	E, s, k

Angle-Resolved Photoemission Spectroscopy (ARPES)



First Angle-Resolved Photoemission Experiment of Band Mapping



Neville V. Smith (1942-2006)



N. V. Smith, M.M. Traum and F.J. DiSalvo Solid State Communications 15, 211 (1974)

Angle-Resolved Photoemission Spectroscopy (ARPES)

$$E_{K} = \hbar^{2}/2m_{e}(K_{out}^{2}) = \hbar^{2}/2m_{e}(K_{x,out}^{2} + K_{y,out}^{2} + K_{z,out}^{2})$$

$$K_{x,out} = \sqrt{2m_{e}E_{K}}sin\theta$$

$$K_{y,out} = \sqrt{2m_{e}E_{K}}cos\theta sin\beta$$

$$K_{z,out} = \sqrt{2m_{e}E_{K}}cos\theta cos\beta$$

$$E_{B} = hv - \Phi - |E_{K}|$$

$$k_{x,i} = 0.512\sqrt{E_{K}}sin\theta$$

$$k_{y,i} = 0.512\sqrt{E_{K}}sin\beta$$

Photoemission Process











Power of ARPES:

Direct Measurement of Fundamental Parameters





JOURNAL OF ELECTRON SPECTROSCOPY and Related Phenomena

Journal of Electron Spectroscopy and Related Phenomena 124 (2002) 289-315

www.elsevier.com/locate/elspec

Sudden approximation in photoemission and beyond L. Hedin^{a,b}, J.D. Lee^{c,*}

^aDepartment of Physics, University of Lund, Sölvegatan 14A, 22362 Lund, Sweden ^bMPI-FKF, Heisenbergstrasse 1, D-70569 Stuttgart, Germany

^cDepartment of Physics and Department of Complexity Science and Engineering, University of Tokyo, Bunkyo ku, Tokyo 113, Japan

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$$G(\mathbf{k},\omega) = \frac{1}{\omega - \epsilon_{\mathbf{k}} - \Sigma(\mathbf{k},\omega)}$$
$$A(\mathbf{k},\omega) = -(1/\pi) \operatorname{Im} G(\mathbf{k},\omega)$$

Challenge: At which point the sudden approximation breaks down and how?

Angle-Resolved Photoemission Spectroscopy (ARPES) - Direct Comparison between Theory and Experiment



ARPES: A Bridge between Theory and Experiment

- Conventinal Fermi Liquid: $Im\Sigma = -\beta\omega^2$ $Re\Sigma = \alpha\omega$
- Marginal Fermi Liquid:

ImΣ = $-\pi/2Cx$ ReΣ =CωIn(x/ωc) where x=max(|ω|,T)

Electron-Phonon Coupling System

$$\operatorname{Im}\Sigma = -\pi \int_{0}^{\infty} d\omega \alpha_{k}^{2}(\omega) F_{k}(\omega) [2n_{B}(\omega) + n_{F}(\omega + u) + n_{F}(\omega - u)]$$
$$\operatorname{Re}\Sigma = -\int_{0}^{\infty} d\omega \alpha_{k}^{2}(\omega) F_{k}(\omega) \ln \left| \frac{\omega + u}{\omega - u} \right|$$

ARPES Experimental Setup

ARPES Endstation on BL10.0.1, ALS, LBNL



Some Remarks about ARPES

- ARPES is a surface-sensitive technique,
 Jultra-high vacuum is necessary;
- ARPES uses electron emission angle to determine the momentum
 magnetic field is not welcome;
- Electrons have to travel to analyzer directly
 ARPES is not compatible with high-pressure, ultra-low temperature yet.



1. Synchrotron Light Sources;

2. Gas Discharge Lamp;

3. UV and VUV lasers.

What is Synchrotron?



U١



Photon energy

Photon flux

Synchrotron Source for ARPES: BL10.0.1 at ALS, Berkeley

High Flux



- Wide Energy Range: 17-360 eV;
- > High Energy Resolution;
 - (<5meV between 20-50eV)
- > High Flux.

Electron Energy Analyzer



ARPES--Improved Resolution Brings New Discoveries

3rd Generation Synchrotron



Electron Energy Analyzer



	Energy Resolution	Angle Resolution
	(meV)	(degree)
Previous	20~40	~2
Now	<1	0.1

Ag(111) Surface State



G. Nicolay et al., Phys. Rev. B 63(2001)115415.

Evolution of Energy Analyzers: from 0D to 1D Angular Detection



Better Resolution Reveals Electron Kinkiness in Cuprates

Angular Resolution: 2 degrees





Dessau et al., PRL (1995).



Better Resolution Reveals Electron Kinkiness in Cuprates

Angular Resolution: 2 degrees



Dessau et al., PRL (1995).

Angular Resolution: 0.3 degree



Related Issues in Photoemission

Surface vs Bulk Property Measurements


Matrix Element Effect in Photoemission



Measured Signal

Single-particle spectral function —Intrinsic property of materials Fermi-Dirac Distribution

Matrix Element :

Depending on measurement conditions

Photon energy;
Photon polarization;
Momentum.

Bilayer Splitting in Bi₂Sr₂CaCu₂O_{8+δ}









A. I. Liechtenstein et al., PRB 54, 12505 (1996)

Calculations of Matrix Element Effect

Calculated



A. Bansil et al., Phys. Rev. Lett. 83(1999) 5154.



P. V. Bogdanov et al. Phys. Rev. B 64, 180505 (2001)D. L. Feng et al., PRL 86 (2001) 5550.Y. D. Chuang et al., PRL 87 (2001) 117002.

Challenge:

How to calculate Matrix Element Effect in correlated systems?

Light Polarization on Matrix Element Effect



To disentangle different orbitals

 $M_{ij}(E_f, K) \propto <\phi_{f,K}|A \cdot P|\phi_{i,K}>^2$

Related Issues in Photoemission

Photoemission Measured (Basically) Occupied States

Fermi-Dirac Distribution Function at Different Temperatures



ARPES Measurement of Electronic States above Fermi Level



300 K, Original

Divided by Fermi Function



Data Analysis in ARPES –

Energy Gap

ARPES on Superconducting Gap of Cuprates





ARPES Observation of Pseudogap



Bi2212 UD85K

> D. S. Marshall et al., Phys. Rev. Lett. 76 (1996) 4841;

A. G. Loeser et al., Science 273 (1996) 325;

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

Determination of Energy Gaps—EDC Symmetrization



D. F. Liu, X. J. Zhou et al., Nature Communications 3 (2012) 931.

How to Measure Energy Gap—EDC Symmetrization

$$I(k,\omega) = I_0(k,\nu,A) A(k,\omega) f(\omega) \qquad f(\omega) = \frac{1}{\exp(\frac{\omega}{k_z 7}) + 1}$$
$$I(k,-\omega) = I_0(k,\nu,A) A(k,-\omega) f(-\omega) \qquad f(-\omega) = \frac{1}{\exp(\frac{-\omega}{k_z 7}) + 1}$$

If
$$A(k,\omega) = A(k,-\omega)$$
,
Then $I=I(k,\omega)+I(k,-\omega) = I_0(k,v,A) A(k,\omega)$

Fermi distribution function is conveniently removed!

ARPES on Energy Gap--Single Layer FeSe



D. F. Liu, X. J. Zhou et al., Nature Communications 3 (2012) 931.

Superconducting Gap

1. Gap opens along the entire Fermi surface;

2. Gap opening has a particle-hole symmetry.



For superconducting gap, in fact, only one point at k_F satisfies $A(k,\omega)=A(k,-\omega)$

ARPES as a powerful tool

for many-body effects

Power of ARPES – A Probe for Many-Body Effects



$$A(k,\omega) = \frac{1}{\pi} \frac{\mathrm{Im}\,\Sigma}{\left[\mathrm{h}\omega - E_k^0 - \mathrm{Re}\,\Sigma\right]^2 + \left[\mathrm{Im}\,\Sigma\right]^2}$$



Many-Body Effects: Interaction of electrons with other entities such as other electrons, phonons, magnons and etc.

Energy Distribution Curve(EDC) vs Momentum Distribution Curve (MDC)



EDC Dispersion– It Is Difficult to Fit EDCs

EDCs





MDC Dispersion: It Is **Easy** to Fit MDCs





Momentum k

Extract Electron Self-Energy from Dispersion and MDC Width

Spectral Function:
$$A(k,\omega) = \frac{1}{\pi} \frac{\mathrm{Im}\Sigma}{(\omega - v_0 k - \mathrm{Re}\Sigma)^2 + (\mathrm{Im}\Sigma)^2} \qquad (1)$$
$$= \frac{1}{\pi} \frac{\mathrm{Im}\Sigma/v_0^2}{(k - (\omega - \mathrm{Re}\Sigma)/v_0)^2 + (\mathrm{Im}\Sigma/v_0)^2}$$

Lorentzian Lineshape:
$$L(x) = \frac{H(\Gamma/2)^2}{(x-k_0)^2 + (\Gamma/2)^2}$$
 (2)

$$\frac{k}{k_0} = \frac{(E - \operatorname{Re}\Sigma)}{v}$$

Compare Equ. (1) and (2):

$$k_{0} = (E - \operatorname{Re}\Sigma) / v_{0}$$

$$\Gamma / 2 = \operatorname{Im}\Sigma / v_{0}$$

Therefore

$$\operatorname{Re} = E - k_{0}v_{0}$$
$$\operatorname{Im} = (\Gamma/2) * v_{0}$$

Dispersion and MDC Width -- Fitting MDCs



Manifestation of Many-Body Effects: Band Renormalization



Ashcroft-Mermin, Solid State Physics



Hengsberger et al., PRL 83(1999)592. S. Lashell et al., PRB 61(2000)2371. S. J. Tang et al., Phys. Stat. Solidi. 241(2004)2345.

Many-Body Effects in High Temperature Superconductors



X. J. Zhou, Z. X Shen et al., Nature 423, 398 (2003).

Latest Development –

Laser ARPES

Light Sources for Photoemission Spectroscopy



VUV Laser ARPES System at IOP



(Started development in early 2004, commissioned by the end of 2006) Guodong Liu, X. J. Zhou *et al.*, Rev. Sci. Instrum. 79 (2008) 023105.

Generation of VUV Laser (*hv*=6.994 eV)



KBe₂BO₃F₂ (KBBF): New Non-Linear Optical Crystal



C. T. Chen, Z. Y. Xu et al., Chin. Phys. Lett. <u>18</u>(8), 1081 (2001). J. Appl. Phys., Vol. 74, No. 11, 1 December 1993; J. Appl. Phys. 77 (6), 15 March 1995 Appl. Phys. Lett. 68 (21), 20 May 1996

KBe₂BO₃F₂ (KBBF): New Non-Linear Optical Crystal





C. T. Chen, Z. Y. Xu et al., Chin. Phys. Lett. <u>18(8)</u>, 1081 (2001).



China's crystal cache

A Chinese laboratory is the only source of a valuable crystal. David Cyranoski investigates why it won't share its supplies.

ne of Daniel Dessau's prized possessions is a small crystal of potassium peryllium fluoroborate (KBBF). Dessau, a solid-state physicist at the University of Colorado at Boulder, uses the crystal to convert the light of a US\$100.000 laser into a deep ultraviolet, a good wavelength for studying the surface of superconductors. But because the laser light gradually degrades the crystal, Dessau has to save it for special projects. "It is a beautiful crystal," he says. "It would really move the field forward — if people could get it." But Dessau can't get any more of it. Nor can Peter Johnson, a condensed-matter physicist at Brookhaven National Laboratory in Upton, New York, who was once promised it by Chuangtian Chen, the Chinese physicist who runs the only laboratory that knows how to make the crystals. And nor can any of a host of other solid-state physicists outside China. "There has been a limited release," says Johnson. "I don't know the politics behind it."

In fact, the politics is simple. The Chinese government is squeezing the crystal for every bit of academic and, eventually, commercial potential it can vield. In October 2008, the finance ministry sidestepped traditional scientific funding channels and started throwing 180 million renminbi (US\$26 million) at a three-year national project to find better ways to produce and use KBBF. China has selected a

handful of groups to work with Chen's crystal, including teams studying the newest type of superconductor, called pnictides. China's monopoly of this crystal is no fluke. At a time when materials scientists and solidstate physicists elsewhere are seeing a lack of

investment, their counterparts in China are surging ahead in a wide range of materials research for much the same reasons as they did with KBBF. The nation has accumulated a great depth of crystal-growing know-how over the past three decades; it has steadfast government support; and its scientists are willing to subsume

themselves in a large team effort and take on the often thankless, sometimes dangerous and always tedious trial-and-error task of synthesizing new materials. "Many great discoveries in this field come from putting things together and getting the temperature and timing just right," says Christos Panagopoulos, a materials researcher at Nanyang Technological Uni-

"doesn't require genius", he says. KBBF's ability to shorten the wavelength, and thereby boost the frequency, of laser light is an example of 'nonlinear' optics, a field that first blossomed in the 1960s as lasers became more

circumstances, light passing through water, glass or any other material will perturb the atoms only slightly, so that they vibrate in sync with the light wave. As a result, light can be reflected, refracted, scattered and absorbed ad infinitum without its frequency being affected. Nonlinear effects are evident only when the light is so intense that the vibrations it causes compete with the binding

You need a lot of quipment and you eed to move slowly Chuangtian Cher

versity in Singapore. The discovery process

widespread in laboratories. Under ordinary

forces on the atoms. When highly perturbed, as in the case of high-intensity lasers, the atoms can absorb the energy of the incoming light and re-emit the light with a frequency that is double, triple or even some

higher multiple of the original. A variety of materials have been discovered that can boost laser light to frequencies that the lasers alone cannot produce, and each has a set of signature frequencies that it can achieve.

China might easily have fallen behind in this field, as it did in so many others. Just as nonlinear optics started coming into its own, China was caught up in the Cultural Revolution, a particularly dark period starting in the mid-1960s when many academics were criticized as being elitist or impractical and sent to do farm work for 're-education'.

But Chen, now a spritely 71-year-old at the Technical Institute of Physics and Chemistry 953

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Nature Report, February, 2009

Advantages and Disadvantages of VUV Laser ARPES

Light Source	VUV Laser	Synchrotron
Energy Resolution (meV)	0.26	5~15
Momentum Resolution (Å ⁻¹)	0.0036	0.0091
	(6.994eV)	(21.1eV)
Photon Flux(Photons/s)	10 ¹⁴ ~ 10 ¹⁵	10¹²-10¹³
Electron Escape Depth (Å)	30~100	5~10
Photon Energy Tunability	Limited	Tunable
k-Space Coverage	Small	Large

Laser and Synchrotron are complementary.

VUV Laser Photoemission Lab at IOP



Angle-Resolved Photoemission Spectroscopy (ARPES)



Principle of Spin-Resolved ARPES



Mott Spin Detector



The efficiency of Mott Spin Detector is extremely low $\sim 10^{-4}$

Synchrotron Radiation-Based Spin ARPES



Energy Resolution ∆E is inversely proportional to Light intensity

In order to get decent signal intensity:

(1). Sacrifice of energy resolution to get high intensity;(2). Angle-integrated measurements.

Direct Evidence for a Half-Metallic Ferromagnet



National Synchrotron Light Source (NSLS) Brookhaven National Lab

1.Energy Resolution: 200 meV

2.Angle-Integrated.

J. H. Park et al., Nature 392 (1998) 794.
Spin-Resolved ARPES on Topological Insulators



Spin-Resolved



- Δk —Momentum Resolution
- **ΔE—Energy Resolution~80meV**

Done at Swiss Light Source

Advantages of Laser in Spin-Resolved Photoemission



Best energy resolution: ~50-100meV

(1).Narrow linewidth (0.26meV);
(2).Extremely high photon flux (10¹⁵ photons/s).

VUV Laser Spin-Resolved ARPES



Laser ARPES Based on Time-of-Flight Analyzer



Evolution of Energy Analyzers: from 0D to 1D Angular Detection



Time of Flight Analyzer: From 1D to 2D Angular Detection



Efficiency of Angle Detection Improved by 250 times

Other Developments and Future Developments

- Ultra-Low Temperature ARPES; He 3 pumping, Sample temperature<1 K;</p>
- Laser ARPES with Higher Photon Energy;
- > Time-Resolved ARPES;
- > Spatially-Resolved ARPES.

Summary

In the past two decades, ARPES has continuously experienced dramatic improvements;

This improvement still keeps going;

Every time there is a significant improvement on resolution, there are new findings.

Thanks