New Opportunities with Soft X-Rays for understanding Emergent Phenomena

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Division Deputy for Scientific Support

Advanced Light Source

Lawrence Berkeley National Laboratory

ic/ALSaerial/11-96

Grand Challenges !!

- **Energy problem** – search for 20 TWatts of energy,

  solar energy, hydrogen fuel, nuclear energy ??
- **Membrane Proteins** – from 3D structure of Macromolecules to understanding functions-dynamics
- **Understanding Emergent Phenomena** -

  Phenomena which are not the properties of the individual elementary components BUT of the assembly of such components;

  Strongly correlated electron systems - high Tc superconductor……
- **Spintronics, Quantum Computing**
- **Single atom/molecule imaging/spectroscopy....**

OUTLINE

- What are techniques of choice for understanding electronic properties of correlated systems?
- What facilities exist or are under development at the ALS?
  Explanation with a few *EXAMPLES*
- Future opportunities and critical parameters of next generation of light sources.

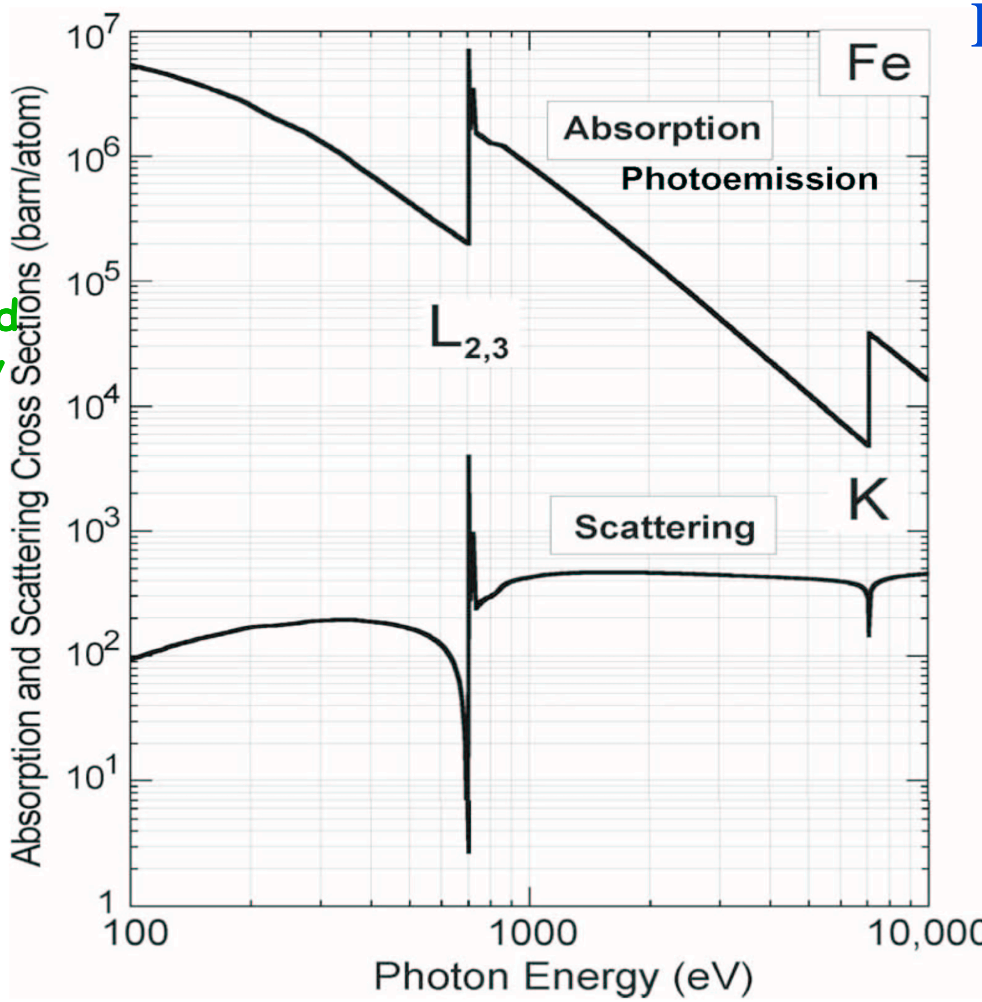
Understanding complex correlated phenomena require sharper and sharper tools

Why X-Rays (& not neutrons or electrons) ?

Tunable x-rays offer variable interaction cross section

Optical ↑ Electrons ↑

- Resonance effect
- Scattering combined with spectroscopy



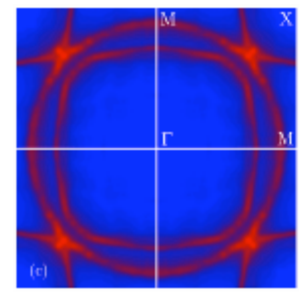
neutrons ←

Courtesy: Stohr

Co

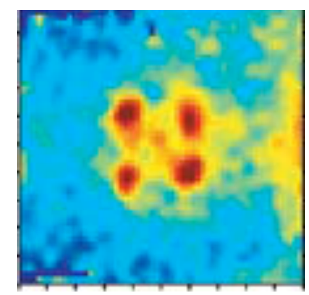
Fundamental Spectroscopies of Condensed Matter

Spectral functions (One-particle properties)
Correlation functions (two-particle properties)



1-particle response

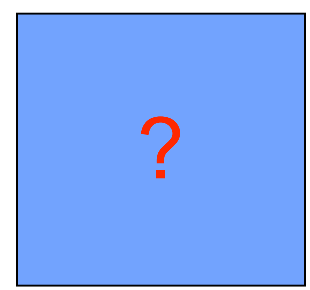
•Angle resolved photoemission (ARPES) :
Single-particle spectrum $A(k,\omega)$



2-particle responses

•<u>*Spin*</u> : Inelastic Neutron Scattering (INS) :
(neutrons carry magnetic moment)
Spin fluctuation spectrum $S(q,\omega)$

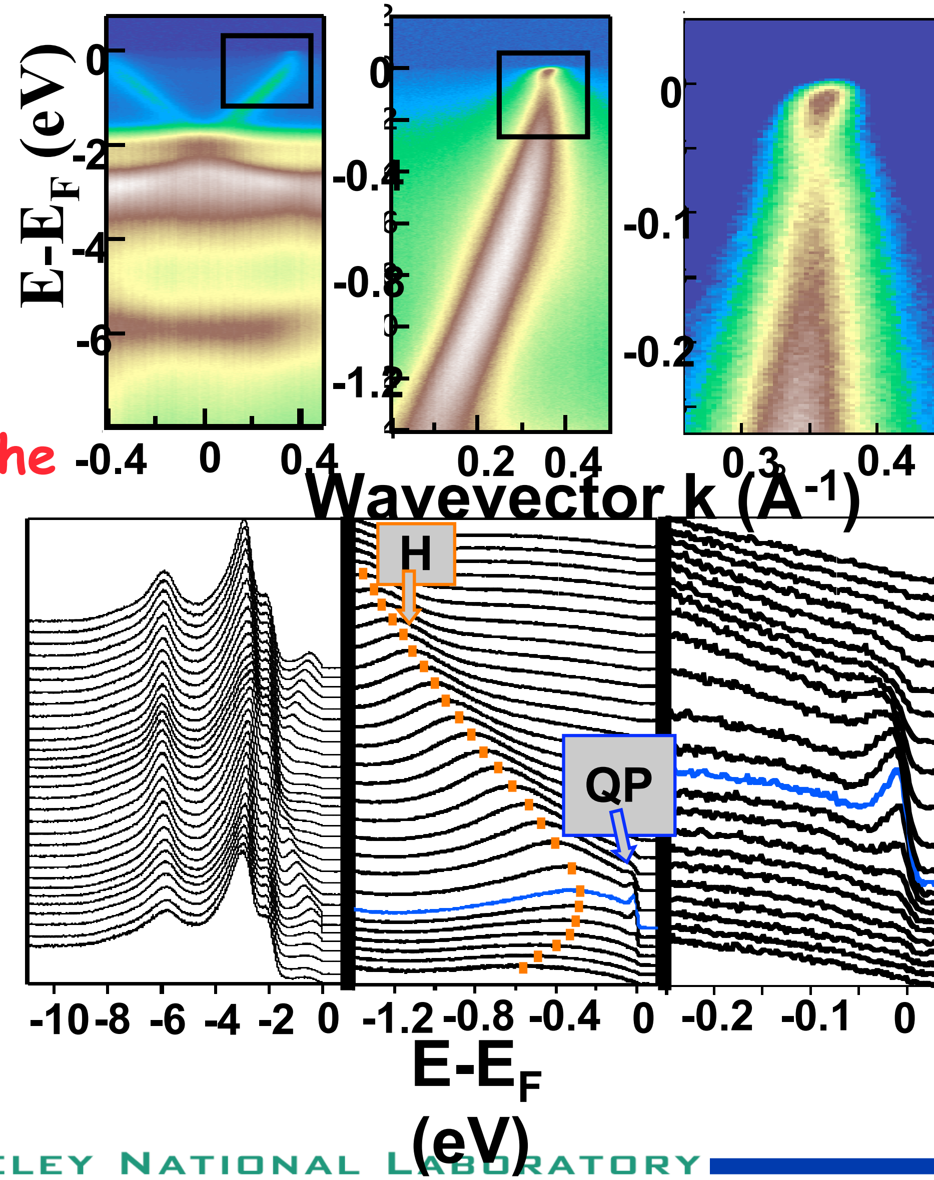
•<u>Charge</u> : Inelastic x-ray scattering (IXS) :
Coupled excitation in the
Charge Channel $N(q,\omega)$



(MERLIN/QERLIN (ALS); FEL)

Example 1: Quasiparticle in LSMO x=0.4

- Nodal quasiparticle:
- nodal-antinodal dichotomy in a *non* superconducting material
- Are the pseudogap state & the nodal-antinodal dichotomy hallmark of the superconductivity state?

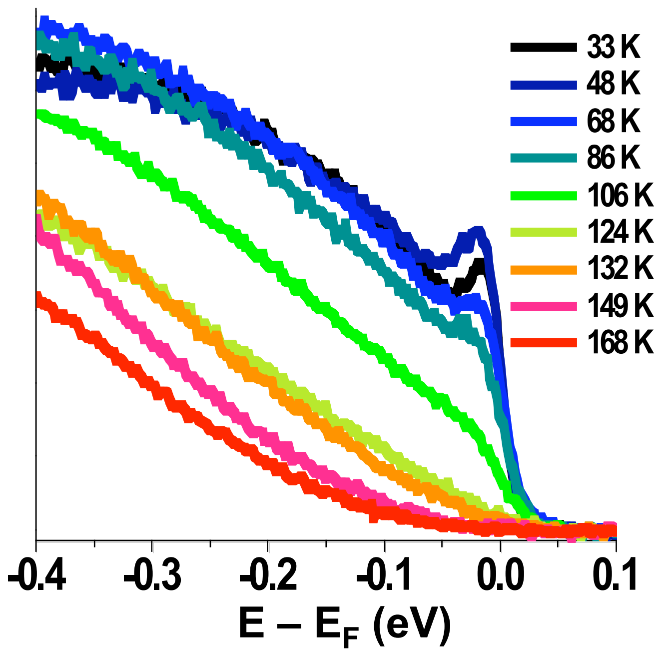


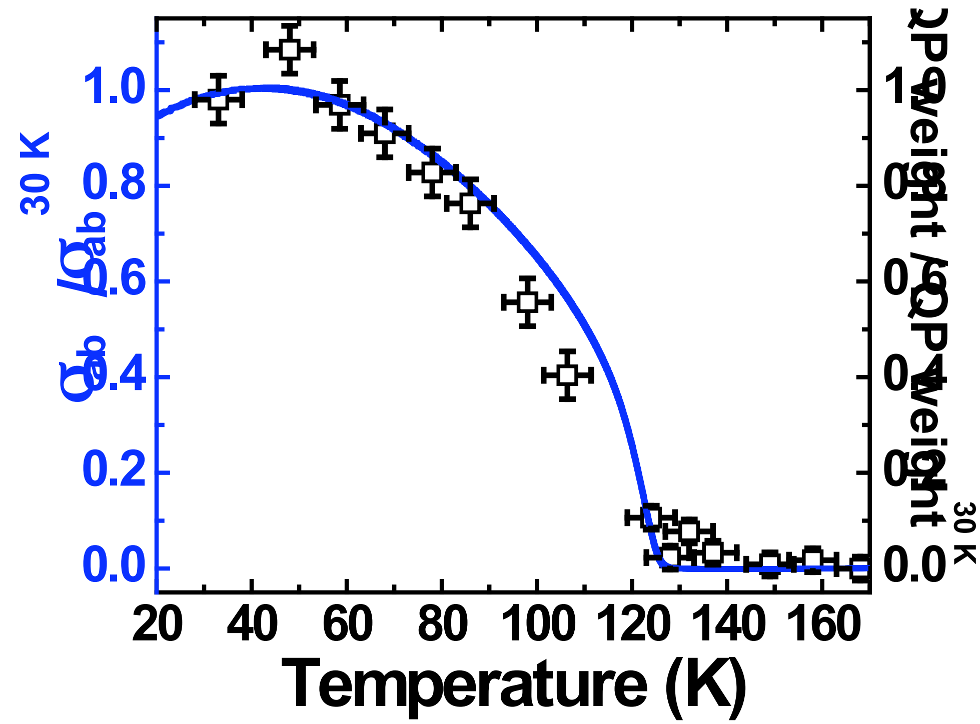
N. Mannella et al.
Nature 438, 474 (2005)

Temperature evolution of the small QP peak linked to transport properties

and the metal-insulator transition in LSMO

Mannella et al



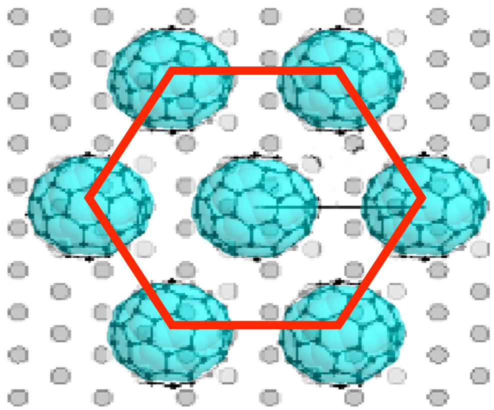


How could measurement of a microscopic electronic structure in certain part of the BZ could be related to the bulk macroscopic property?

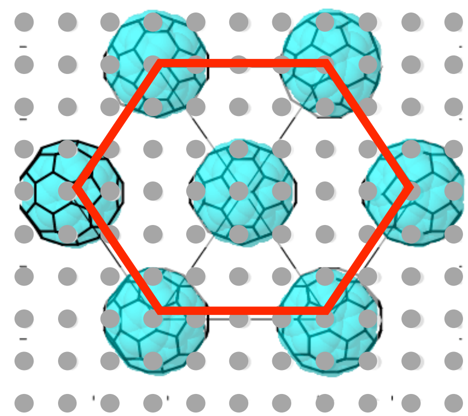
Example 2: Experimental Observation
(C_{60} on Ag (111) and Ag (100) Surfaces)

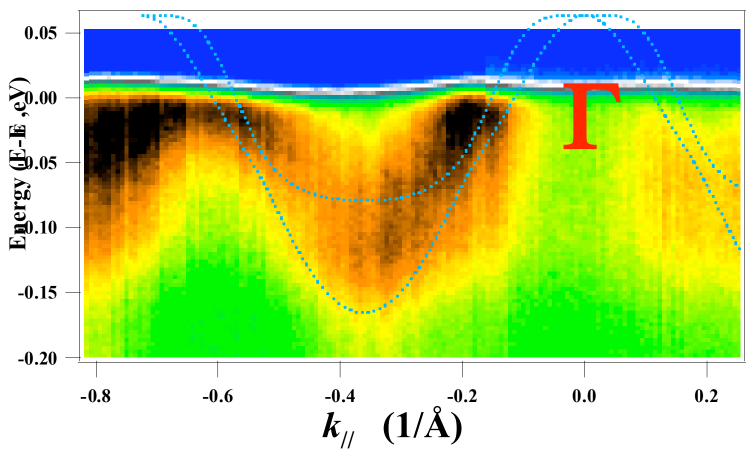
BERKELEY LAB

(111) surface

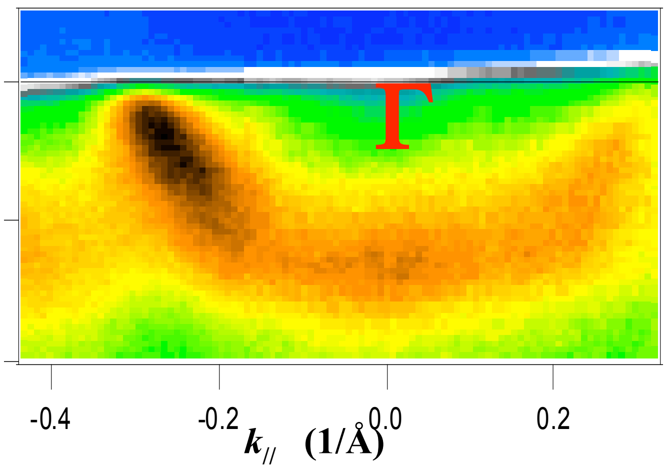


(100) surface





C_{60} ML / (111) surface

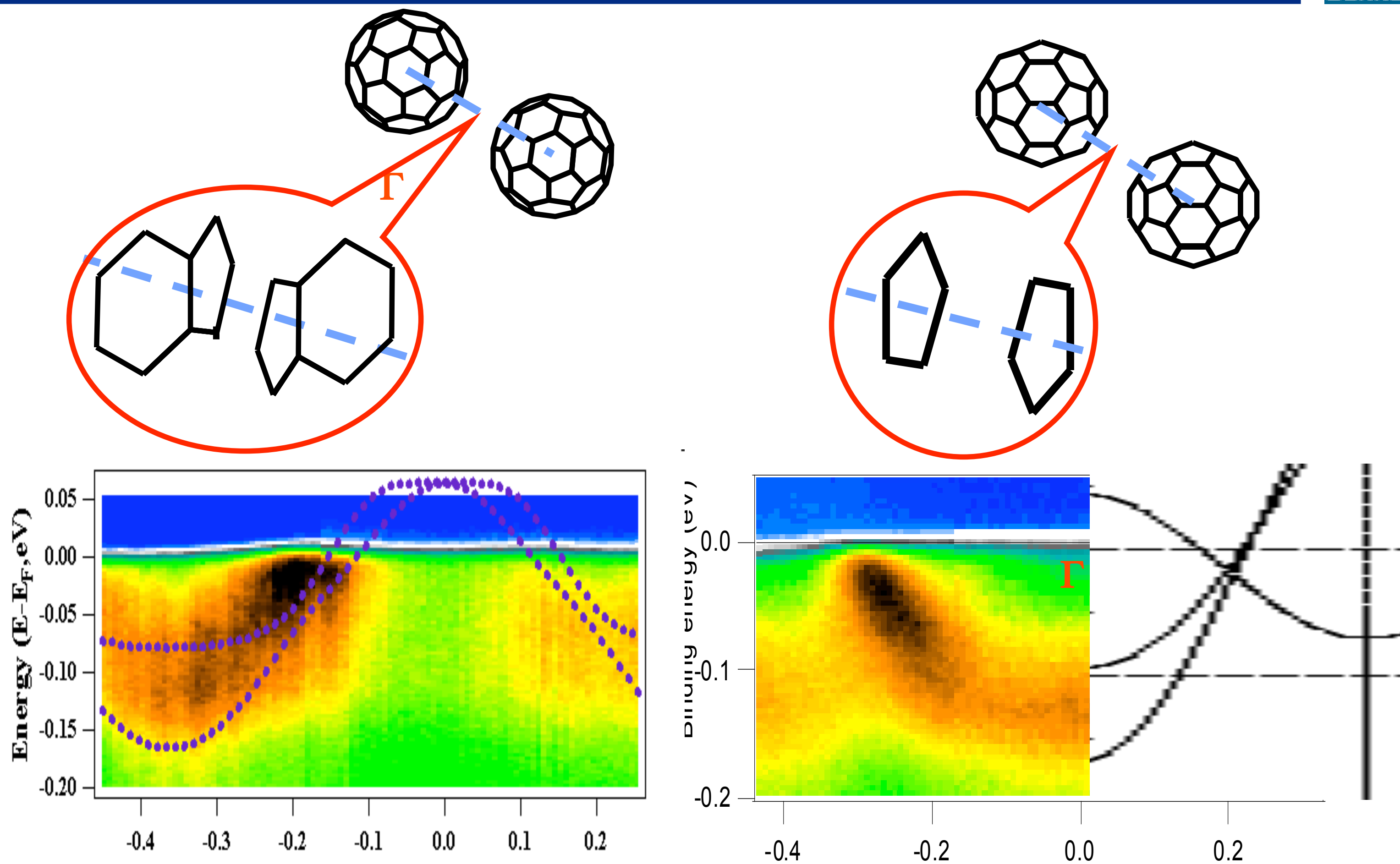


C_{60} ML / (100) surface

Same Hexagon Structure; Completely Different Dispersion!

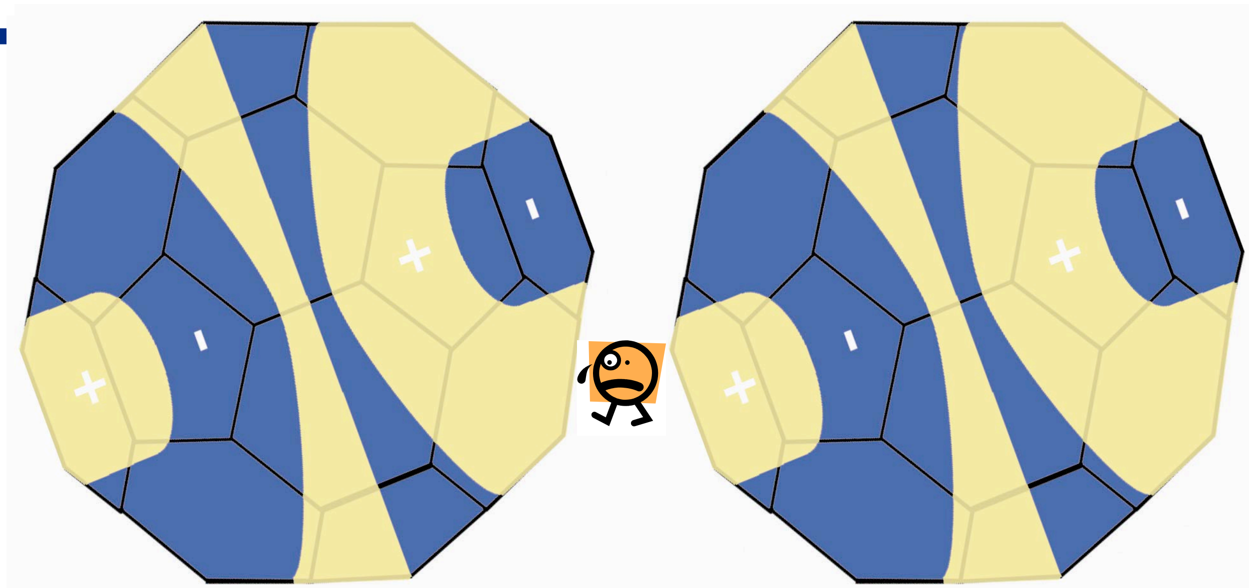
Brouet, Yang et al, Science, 300, 303 (2003); PRL (2004)

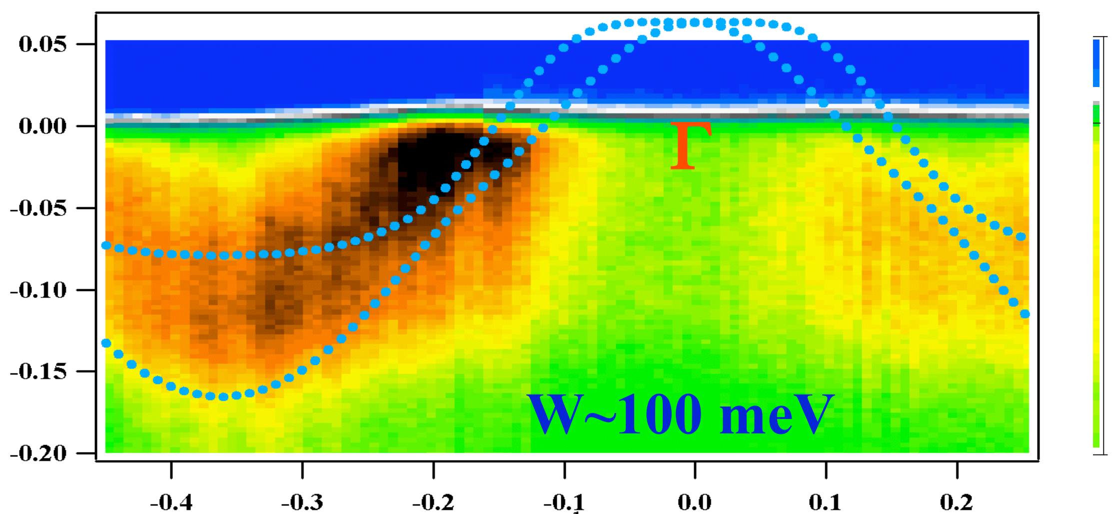
Combination of Experiment and Theory (strong orientation dependence)



Dotted Lines: Theory (Louie, Cohen et al) LDA);
2D Images: Experiments

C60 (Conclusion)





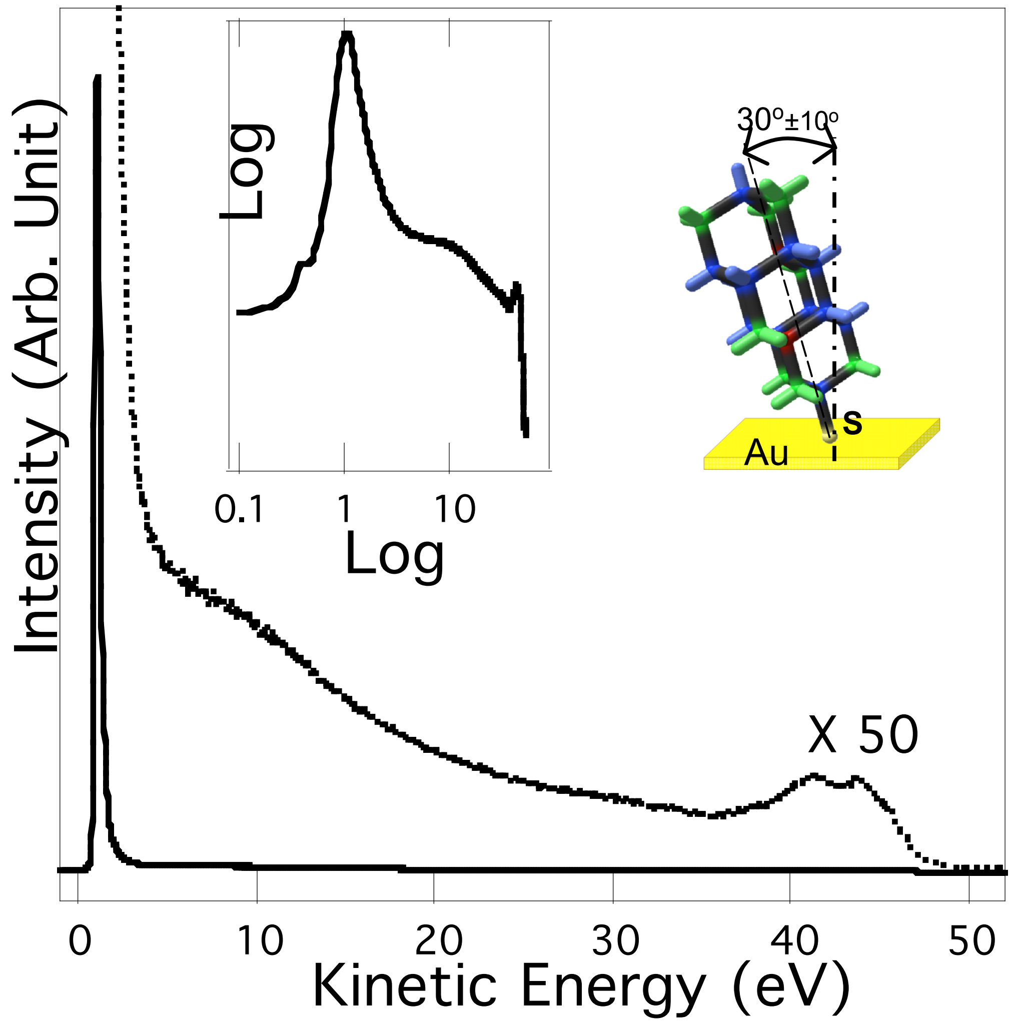
Yang, Brouet, Louie, Hussain, Shen et al

Science 300, 303(2003)

PRL93, (2004)

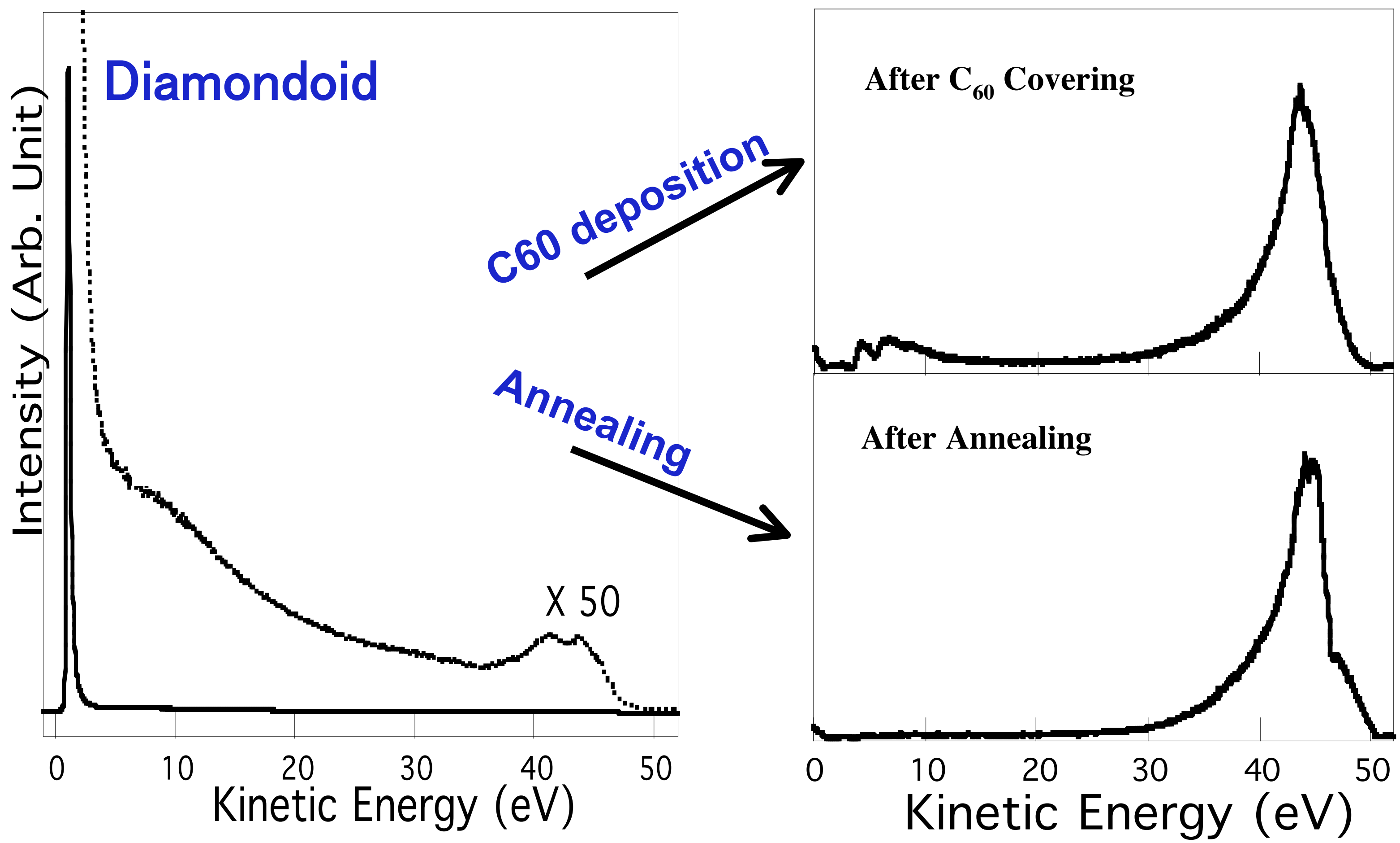
Example 3: Unique Electron Emission of Diamondoid

Possible Photocathode source for FEL, display panel…

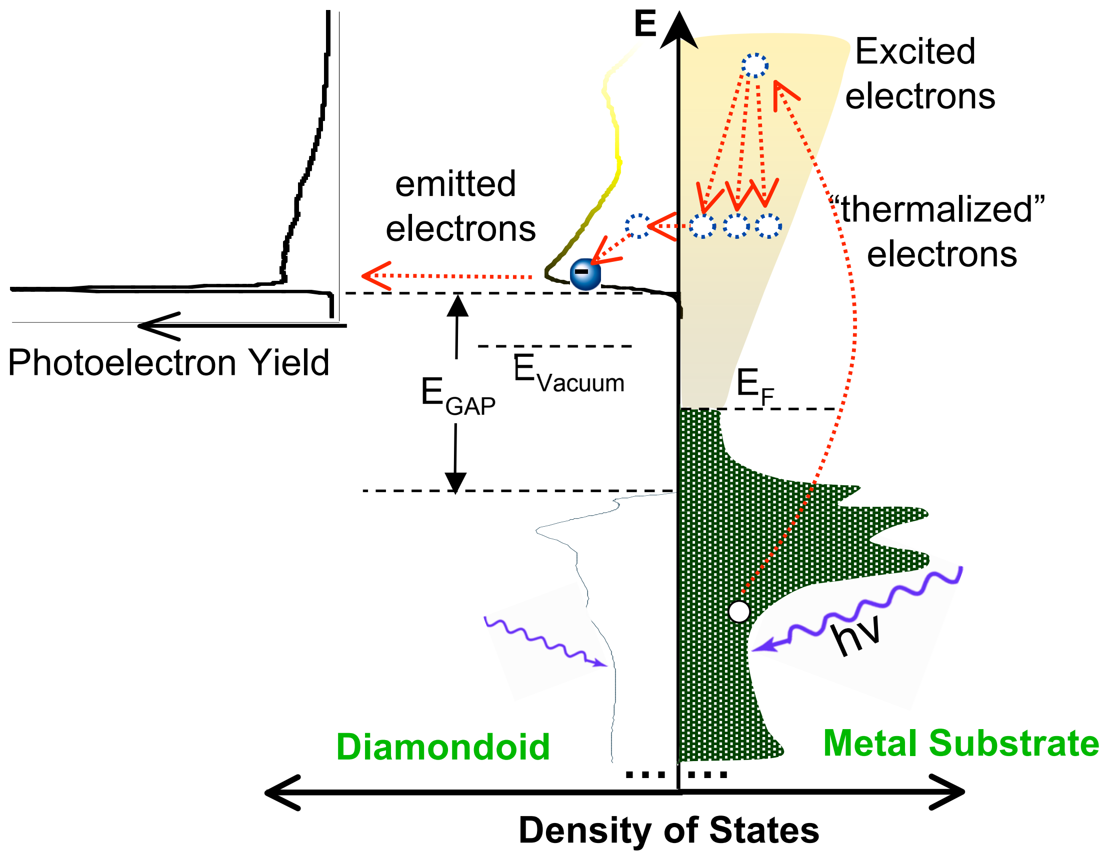


Chevron/Stanford/LBNL
(Yang, Shen, Hussain…
Patent filed
Science … (2007)

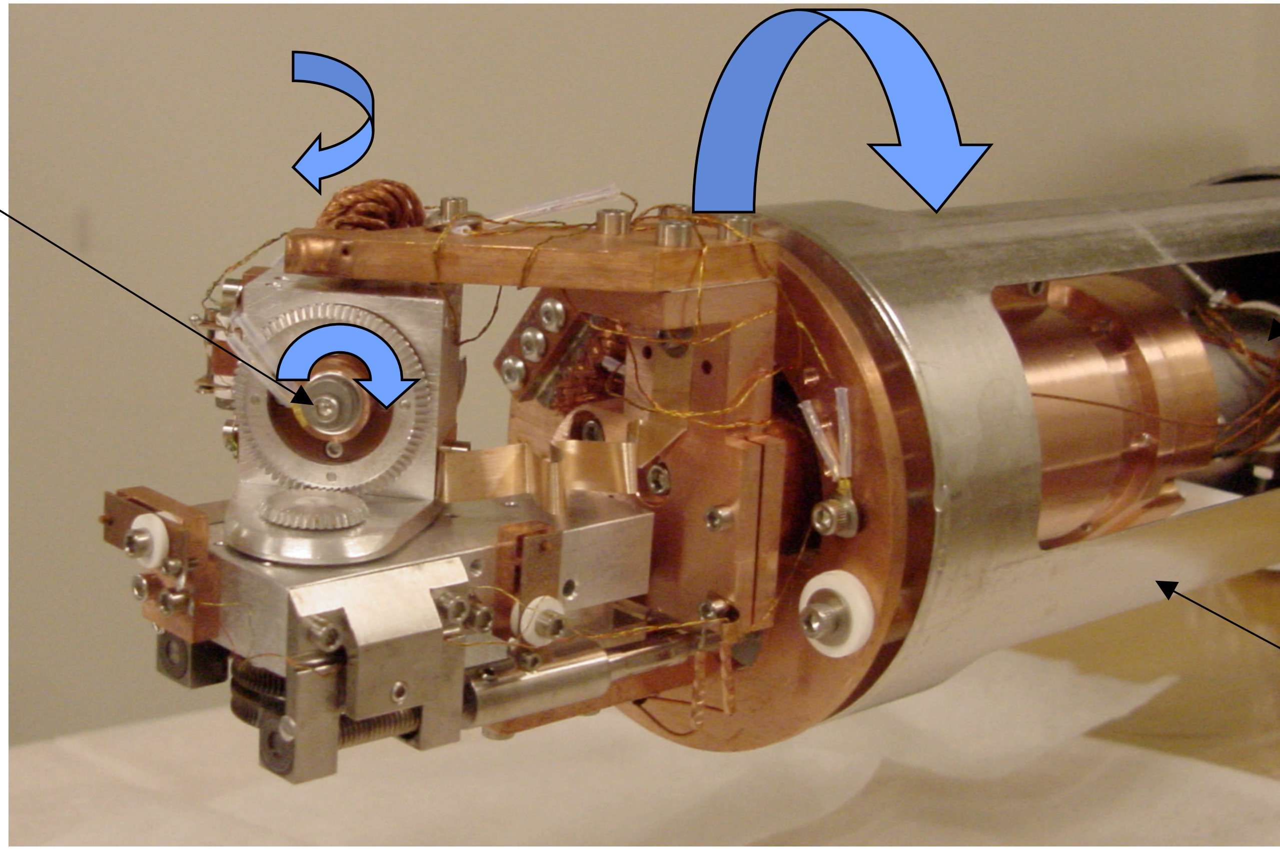
Surface Destruction Test



Electron Emission from Diamondoid



Low-Temperature Goniometer with Six Degrees of Freedom



Sample Position

Cryostat

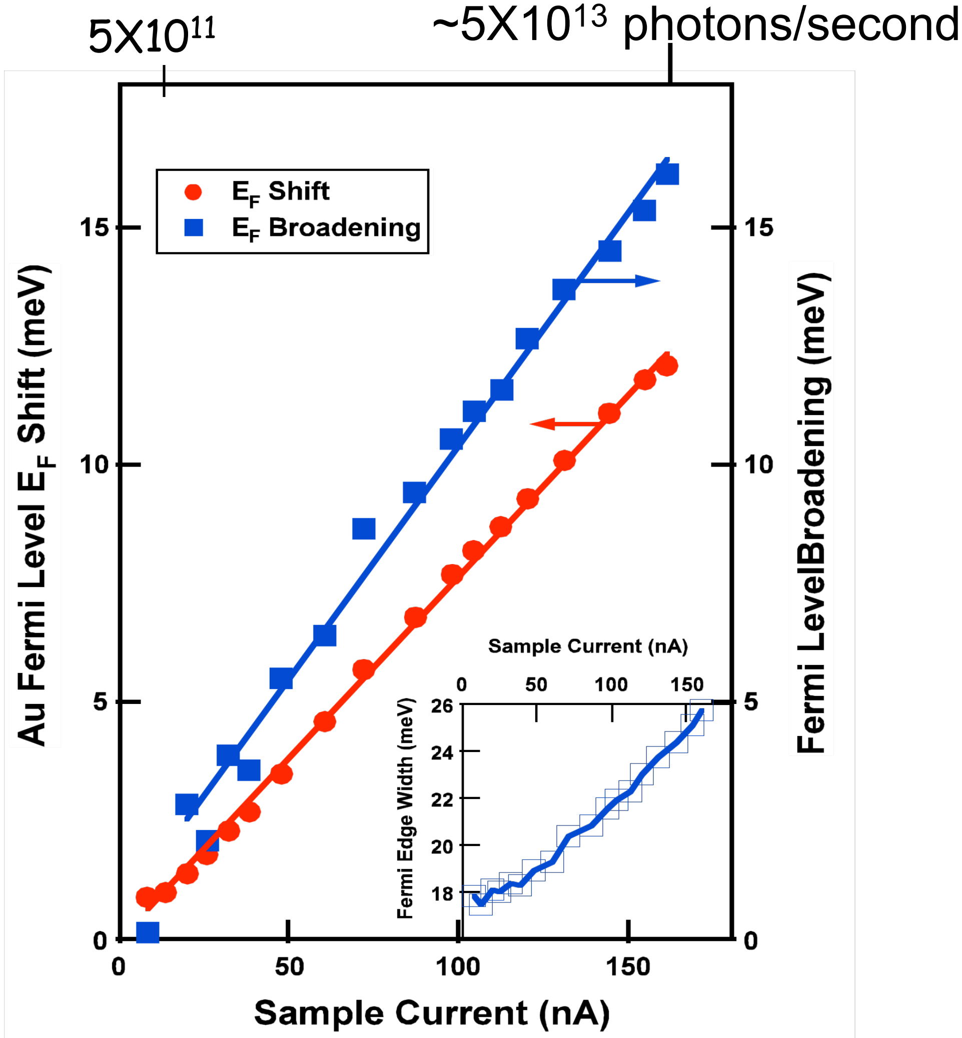
Support tube

(1). Six degrees of freedom: 3 rotational and 3 translational;
(2). Samp temperature ~10 K (no radiation shield);
(for MERLIN; ~3-5K)
(3). Stability of sample against temperature change

Designed and fabricated by John Pepper (ALS)

Space Charge Effect in Photoemission

Caution



X. J. Zhou, B. Wannberg, W. L. Yang, V. Brouet, Z. Sun, J. F. Douglas, D. Dessau, Z. Hussain, Z.-X. Shen, J. Electron Spectroscopy and Related Phenomena (2004).

Overview-New Opportunities @ ALS/LBNL !

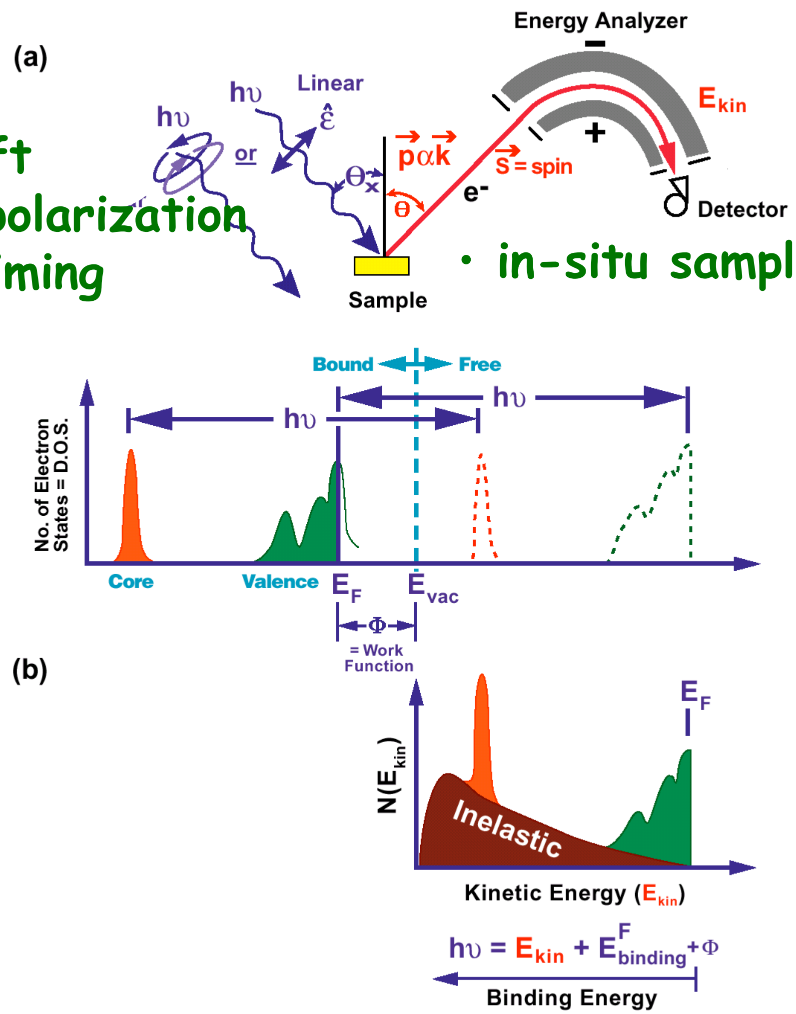
- New kind of Photoelectron Spectroscopy
  - Ambient Pressure Photoemission
    - In Situ (wet); slow dynamics – High speed (2 GHz 1D) Detector
    - High Energy/Momentum Resolution ARPES (non UHV compatible samples)
- New Opportunities with ARPES
  - High Throughput/high resolution Spin-Resolved Photoemission
  - Time-Resolved ARPES with TOF analyzer (high-resolution imaging (kx,ky)
  - Nano-ARPES
    - zone plate diffractive focusing/scanning (spatial res.=50-100nm),
    - 3D imaging TOF analyzer (x, y, spatially resolved & time → energy)
- High Resolution Inelastic Scattering
  - High resolution (<10meV) soft x-ray spectrograph
- meV Resolution Beamline (MERLIN)
  - Ultra-high resolution; quasi periodic undulator
  - Ultra-high resolution photoemission and inelastic scattering
- Next generation of FEL

Photoelectron Spectroscopy

- Spin detector
- High-Resolution TOF analyzer

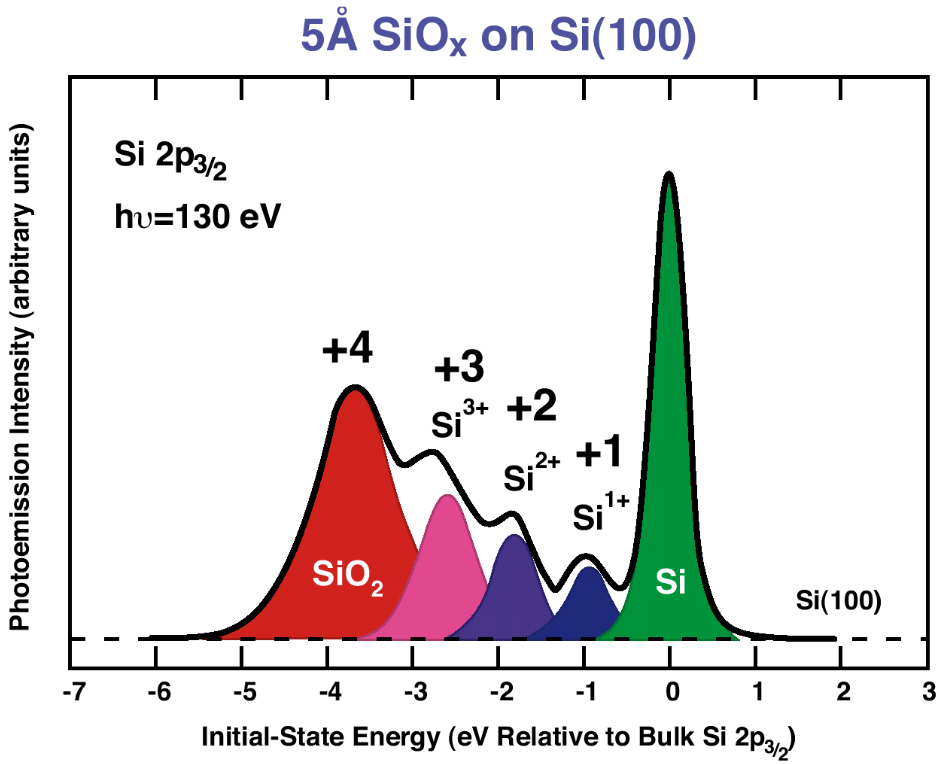
·Right/left Circular polarization
·Pulsed/timing

- in-situ sample environment

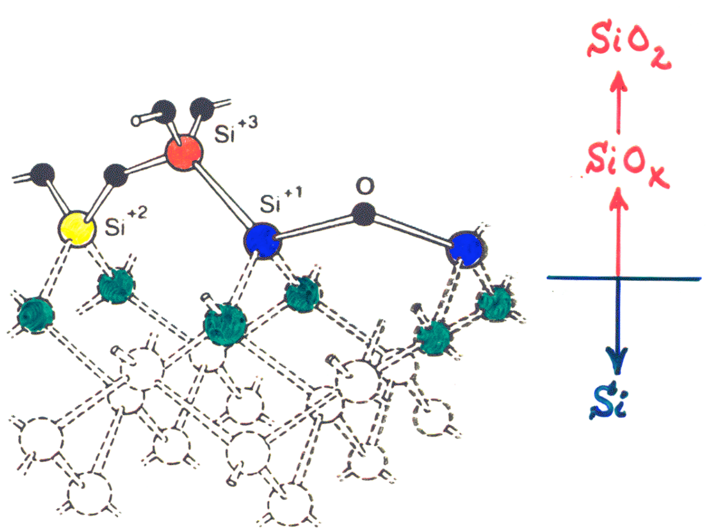


ag.zh/photospectros1/7-97

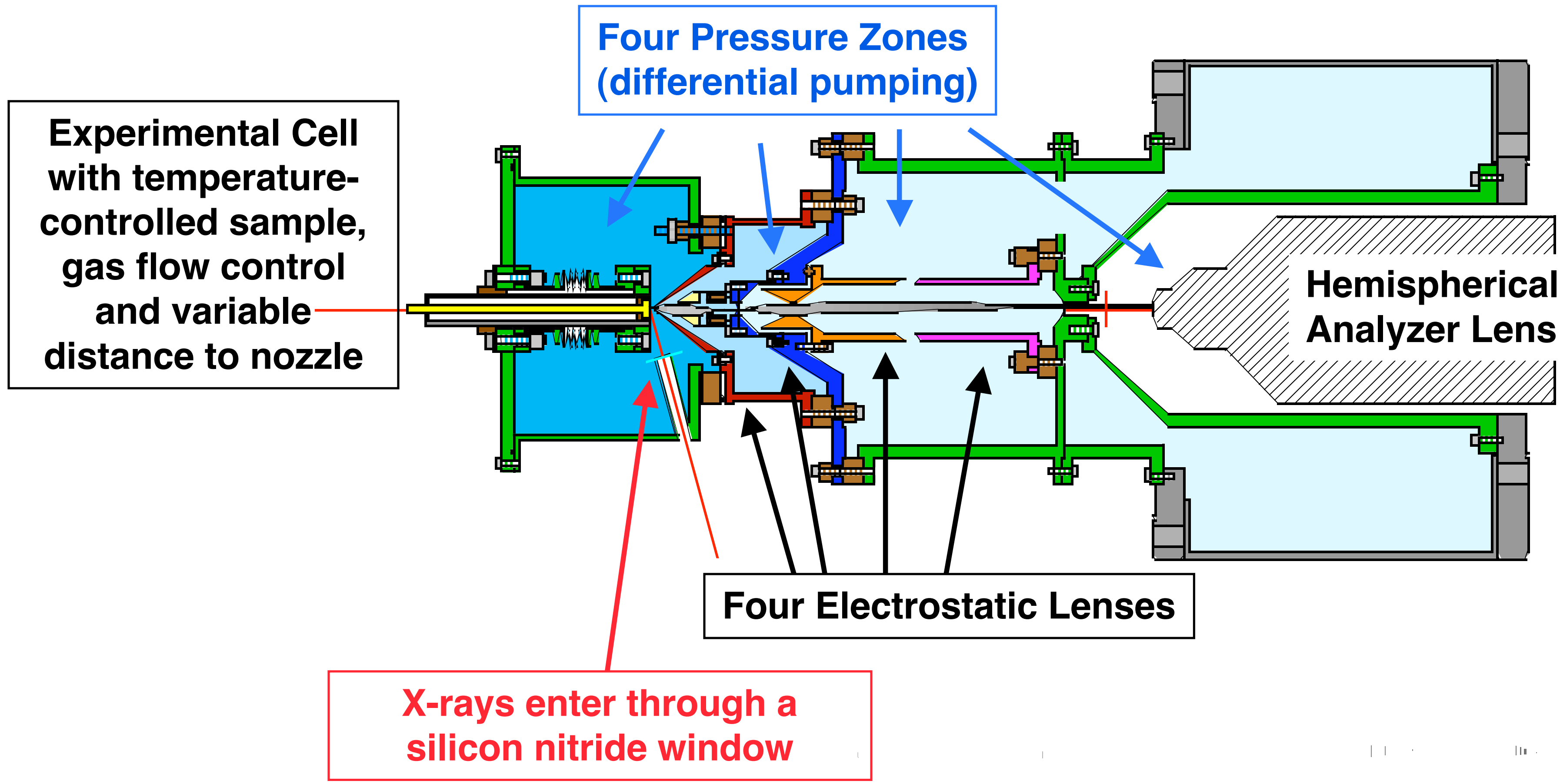
Oxidation of Silicon - Dynamics



(Himpsel et al, NSLS)



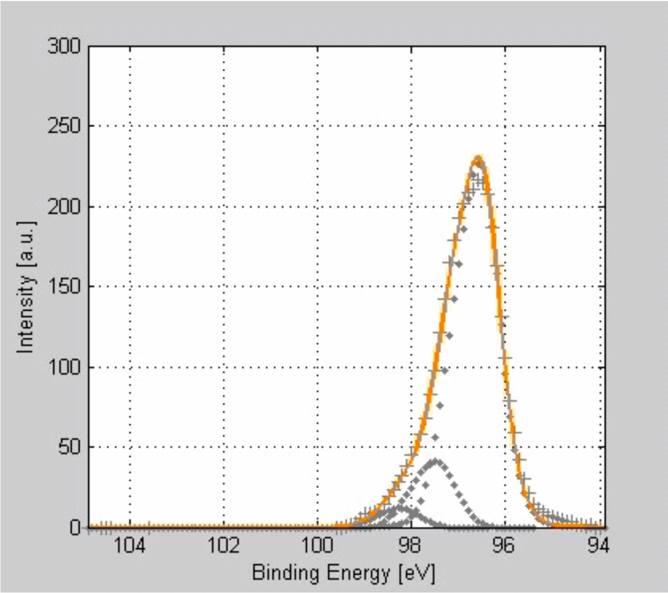
Ambient Pressure Photoemission: ~ 10 torr

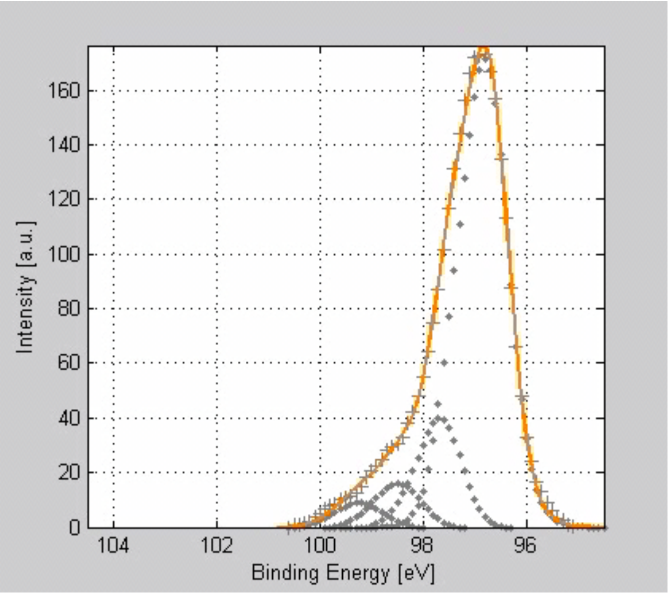


D.F. Ogletree, H. Bluhm, Ch. Fadley, Z. Hussain, M. Salmeron, Materials Sciences Division and Advanced Light Source, LBNL
Published in scientific instrument and methods(2004).

Oxidation dynamics

Strong temperature dependence





250°C | Si^{4+} | Si^{+0} Bulk

450°C | Si^{4+} | Si^{+0} Bulk

Si(100) oxidized by water vapour @ .1 torr

X-ray Spectroscopy of Condensed Matter

Quantum Number Selectivity:

✓ Absorption

$$\omega\ \varepsilon_2 \Rightarrow \Delta E = E_f - E_\iota$$

✓ Angle-integrated photoemission

$$N(E,\hbar\omega) \Rightarrow E_f, E_\iota$$

✓ Angle-resolved photoemission (also inelastic scattering)

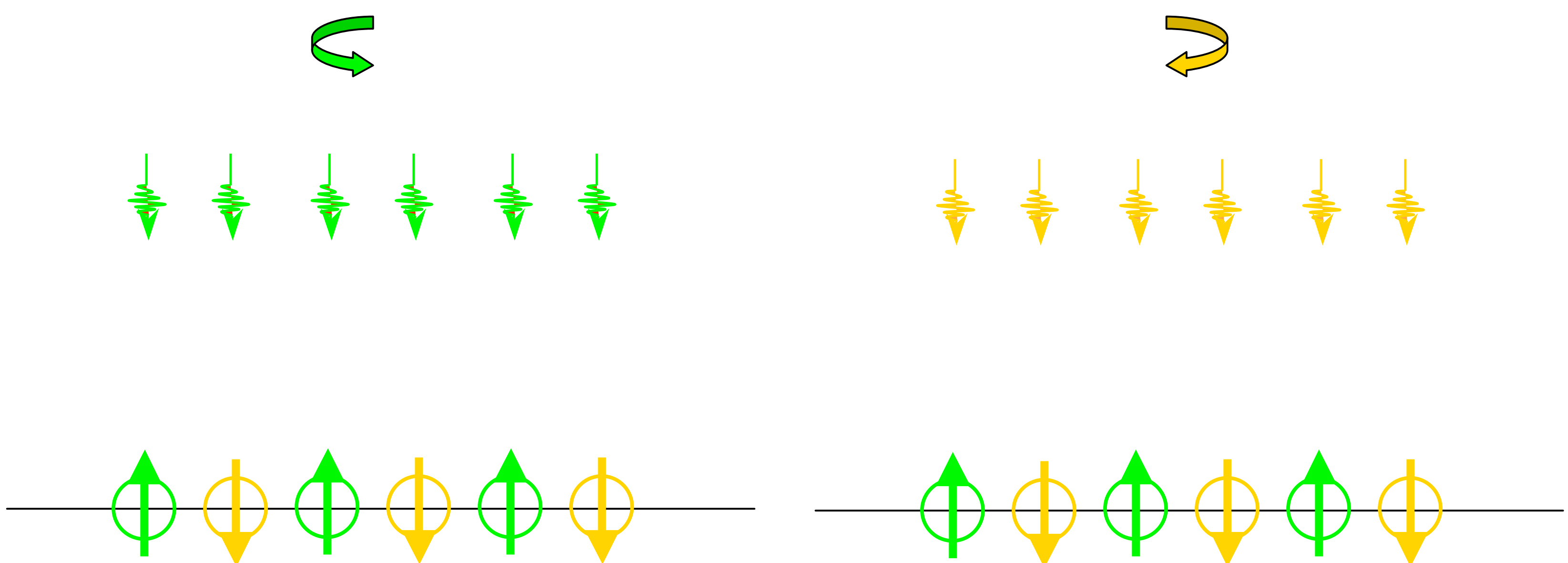
$$N(E,\hbar\omega,\theta,\varphi) \Rightarrow E_f, E_{\iota;}\ \vec{\kappa}$$

!!! Spin-polarized photoemission

$$(N_\uparrow - N_\downarrow)/(N_\uparrow + N_\downarrow) \Rightarrow E_f, E_{\iota;}\ \vec{\kappa}, \vec{\sigma}$$

Photoemission with circularly polarized light and spin detection

Selective excitations
(use of elliptically polarizing undulator)



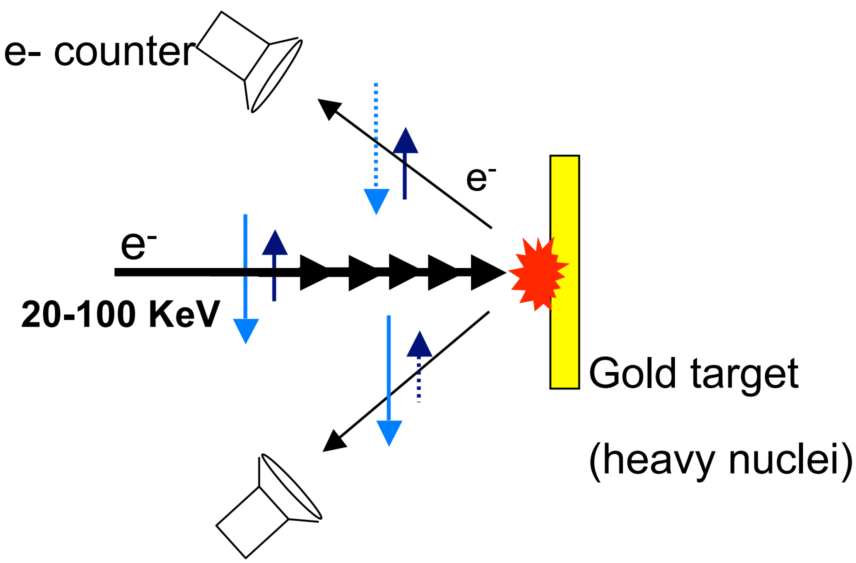
Courtesy: Yulin Chen

Spin detection (two schemes)

Mott Detector

Spin-orbit interaction

Hint = L · S



FOM ≤ 10^{-4}

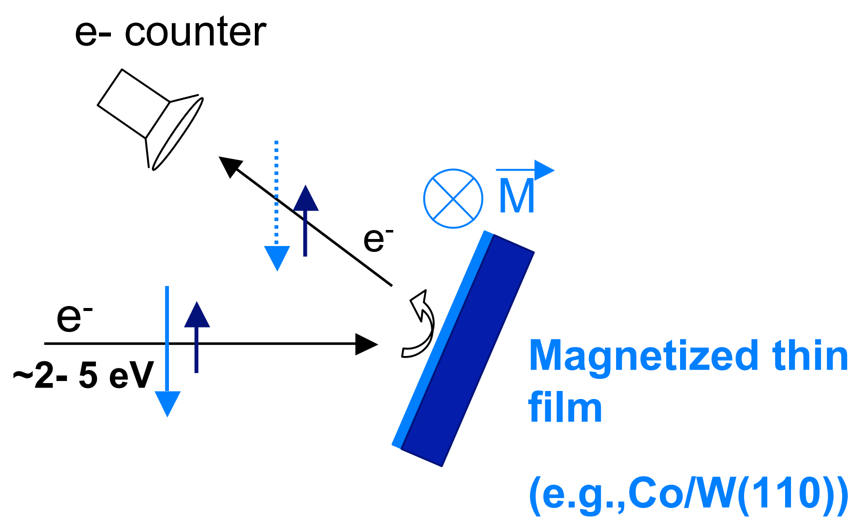
D.T. Pierce et al. 1988 +….

x 100

Exchange scattering interaction

Reflectivity contains a term:

α P · M



FOM ~ 10^{-2}

R. Bertacco et al. 2001
Hillebrecht et al. 2002
R. Zdyb and E. Bauer 2003

Spin-Resolved Photoemission (TOF Project)

"Time-of-Flight" energy analysis

Multichannel detection in time (energy):

~ 10-100 times more efficient than single channel dispersive analyzer

"Exchange Scattering" based spin analysis

~ 100 times more efficient than Mott Detector

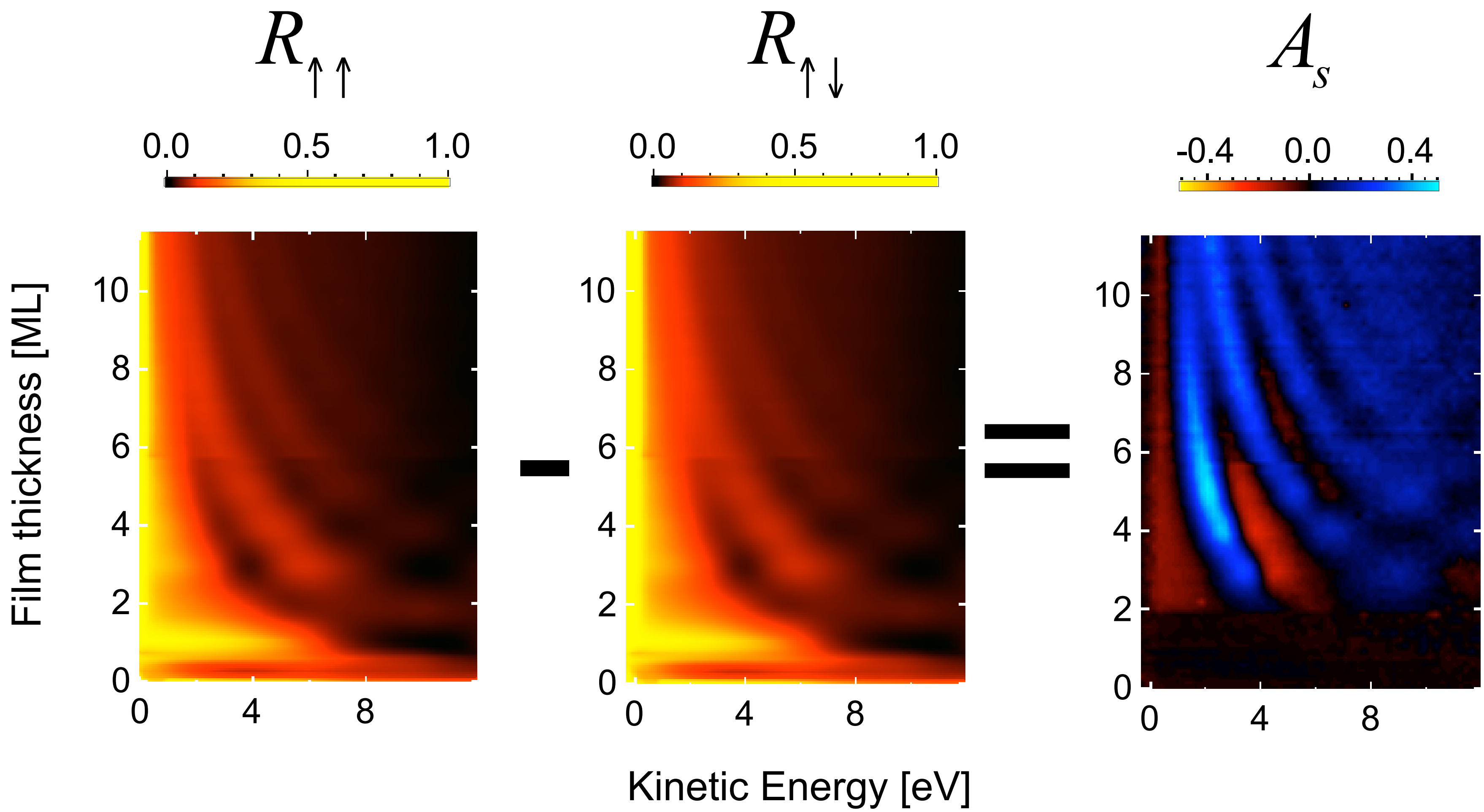
- **Spin-Resolved ARPES:** Improved efficiency & high resolution
  - Overall FOM: ~ 1000 times vs. existing (Mott det.+ dispersive analyzer)

energy resolution : ~ 10meV
Better statistics : > factor of 10

Note: TOF is inherently low noise detection as detector counts for short time; only the time window when electrons of interest arrive

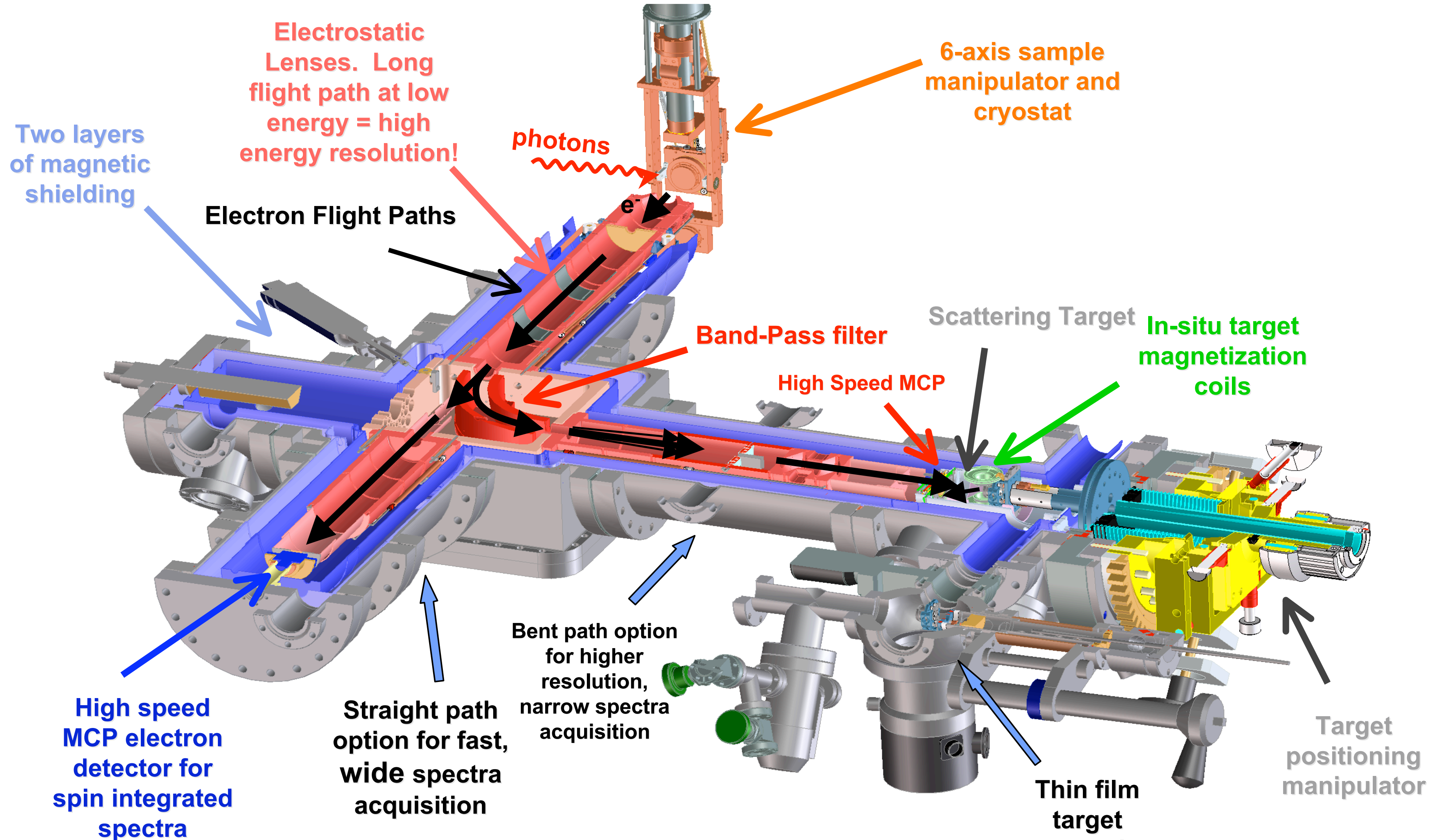
Graf, Schmid, Jozwiak, Hussain, Lanzara et al, PRB, 71, 144429 (2005)

Spin asymmetry (A_s): Co/W(110)
Experimental results using SPLEEM

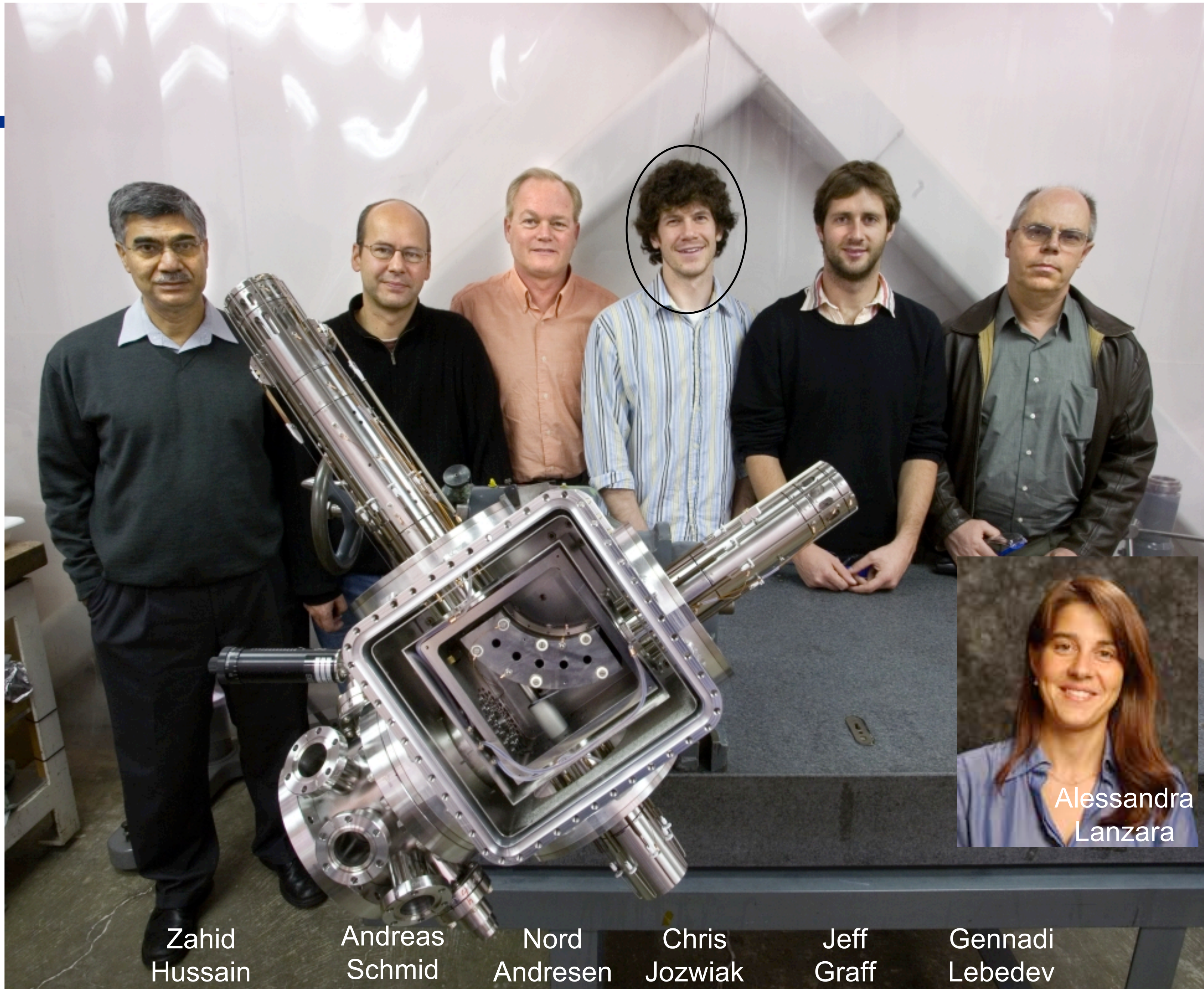


Graf, Schmid, Jozwiak, Lanzara, Hussain et al, PRB, 71, 144429 (2005)

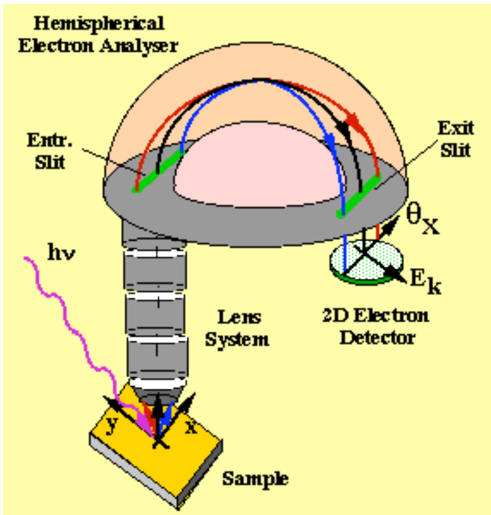
TOF Spin-Resolved Photoemission



(courtesy: Chris Jozwiac)

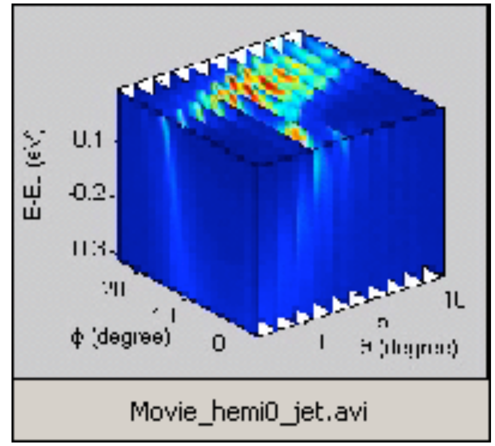

Zahid Hussain
Andreas Schmid
Nord Andresen
Chris Jozwiak
Jeff Graff
Gennadi Lebedev
Alessandra Lanzara

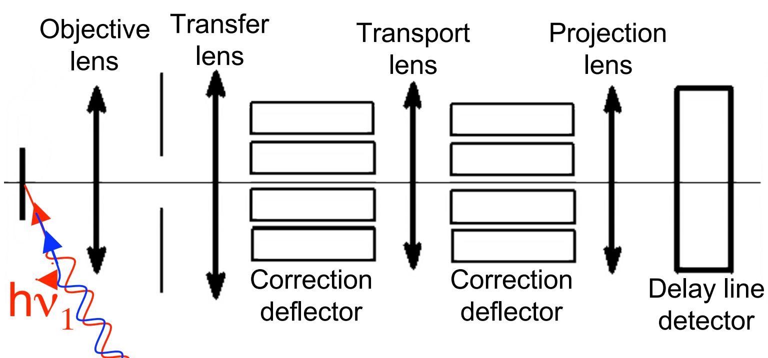
Time-Resolved Photoemission
Comparison of the Hemispherical Analyzer and the TOF Analyzer proposed



Currently used Hemispherical Analyzer (2D detection)

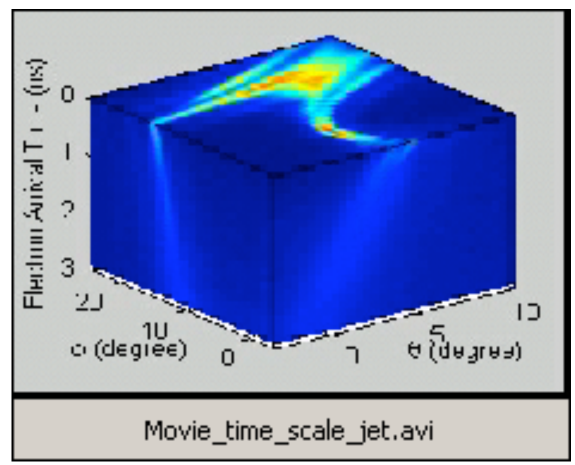
(Bi2212 Bi-layer splitting)





TOF Analyzer Proposed (3D detection)

(Bi2212 Bi-layer splitting)



Key Specifications:
Acceptance angle: 30 degree
Energy resolution: <=2meV (5eV Pass Energy)
Angular resolution: <=0.1 degree (~2mrad)
(comparable to Scienta analyzer but 100 times faster)

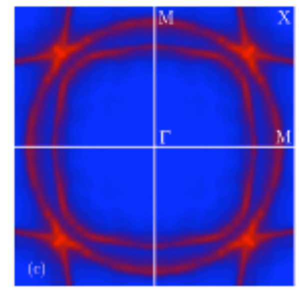
Design and a prototype system

Time-Scale of Various Phenomena

- Ultra-fast time regime: $\leqslant$ 200fs (ΔE >10meV)
    - Electron excitation/de-excitation (fs)
    - Bond breaking
    - Carrier-carrier scattering
    - Hole-optical phonon scattering
    - Charge density wave/charge transfer
    - Magnetic Dynamics
    - Relaxation of biological system after light absorption (Rhodopsin);
- Time regime: ~ 200fs - 2ps (ΔE ~1-10 meV)
    - Phase transition (diamond ⟷ graphite)
    - Carrier acoustic phonon scattering
- Time regime: > 1-100ps
    - Stripe fluctuation in High Temp Superconductor
    - Magnetic recording
    - Protein folding (ps-s)

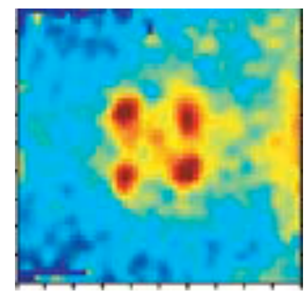
Fundamental Spectroscopies of Condensed Matter

Spectral functions (One-particle properties)
Correlation functions (two-particle properties)



1-particle response

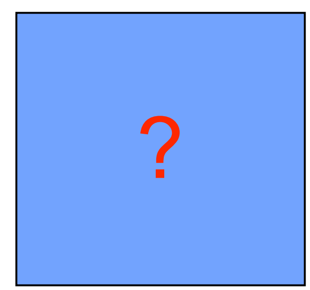
•Angle resolved photoemission (ARPES) :
Single-particle spectrum A(k,ω)



2-particle responses

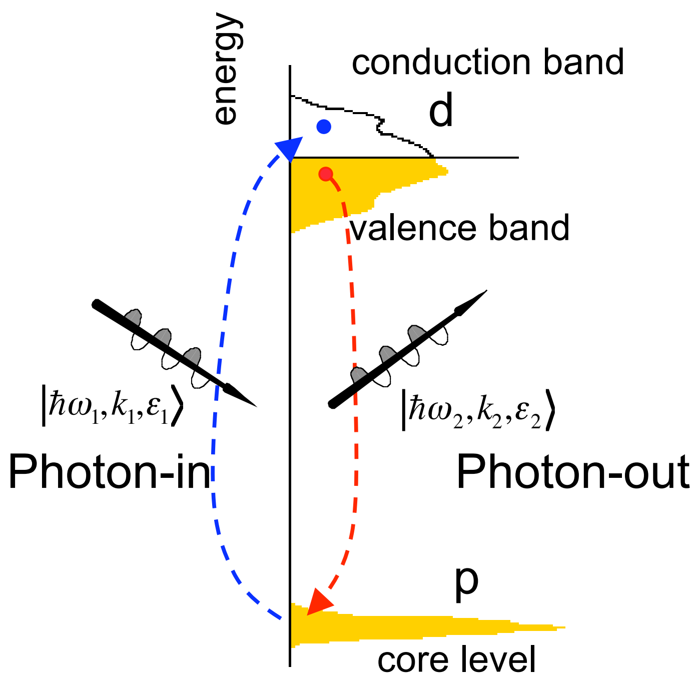
•Spin : Inelastic Neutron Scattering (INS) :
(neutrons carry magnetic moment)
Spin fluctuation spectrum S(q,ω)

•Charge : Inelastic x-ray scattering (IXS) :
Coupled excitation in the
Charge Channel N(q,ω)



(MERLIN/QERLIN (ALS); FEL)

Resonant Inelastic soft X-ray Scattering (*Raman* Spectroscopy with finite q)



Energy loss: $\omega=\omega_2-\omega_1$

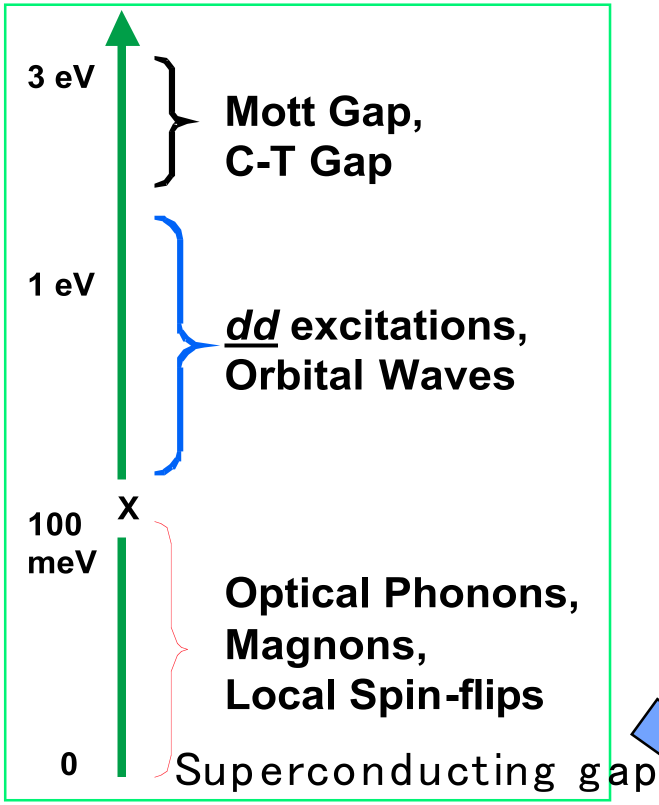
Momentum transfer: $q=k_2-k_1$

Resonance: $\omega_1 \sim \omega_{edge}$

Why???

- Can be applied in the presence of **magnetic/electric field**
- **Bulk sensitive** probe for studying unoccupied electronic states
- Optically forbidden **d-d** excitation
- Finite **q** transfer : spectroscopy of charge fluctuation
- Couples to **charge density** directly (Neutrons couples to spin).
- Energy Resolution **not** limited by the **core hole lifetime**: achieve k_BT resolution

Energy scales of various excitations

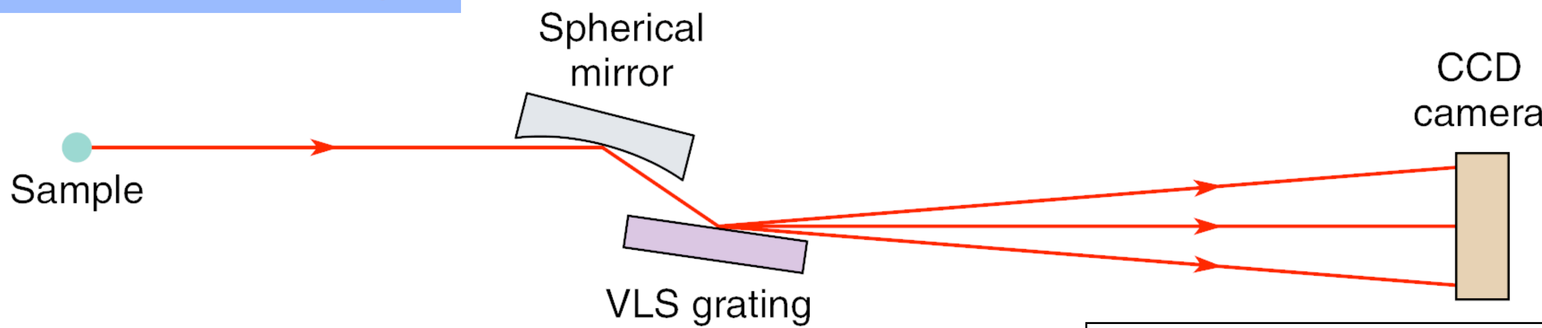


- Superconducting gap ~ 1 – 35meV
- Optical Phonons: ~ 40 - 70 meV
- Magnons: ~ 10 meV - 40 meV
- Orbital fluctuations (originated from optically forbidden d-d excitations): ~ 100 meV - 1.5 eV

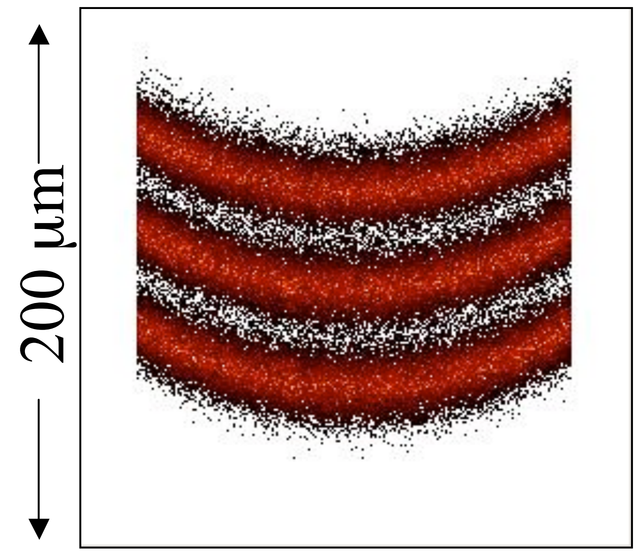
Requires study of many body excitations with energy resolution better than 10meV

meV Resolution VLS Spectrograph

Optical Design



Ray Traces

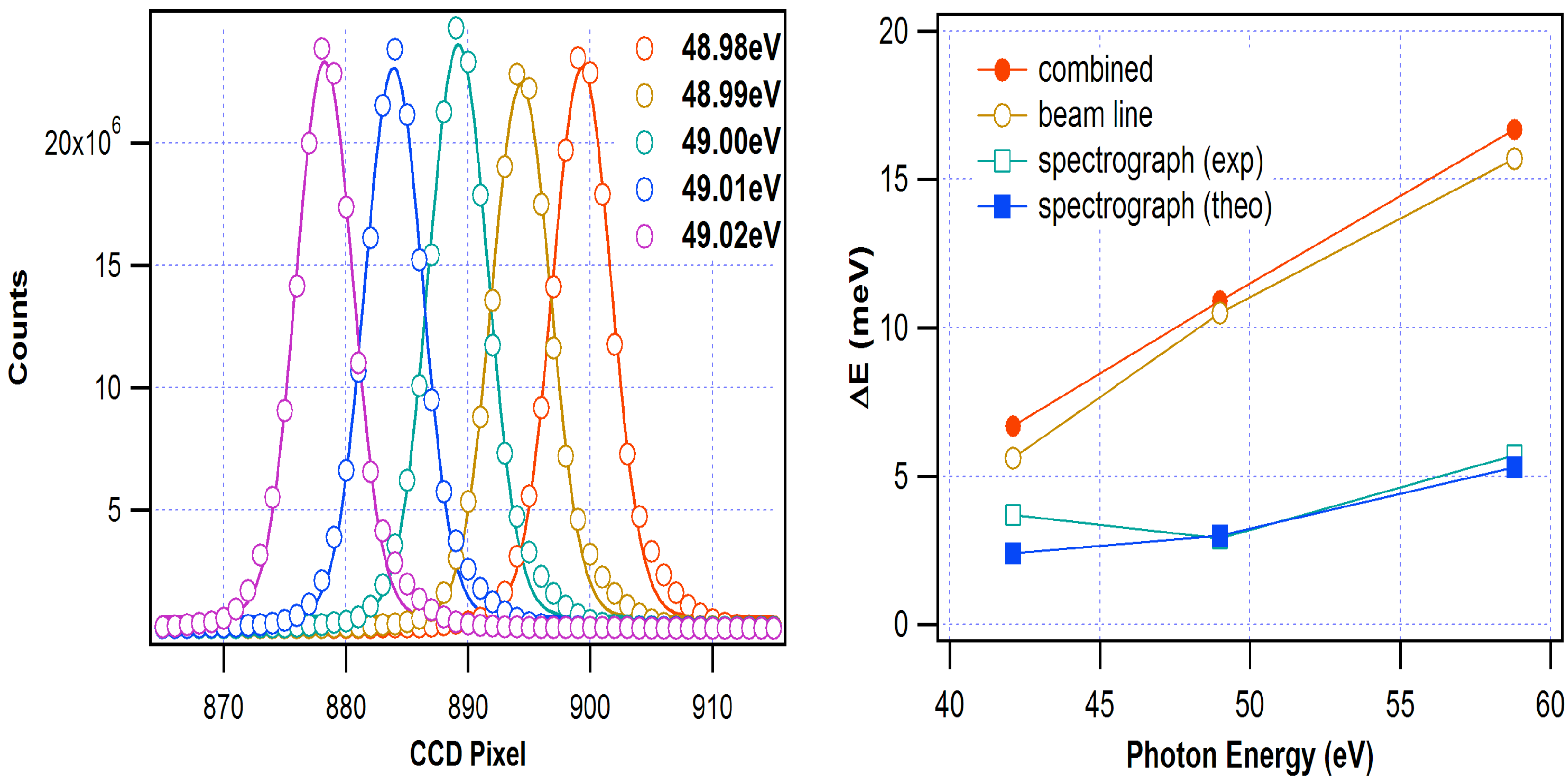


hν = 49 eV ± 5meV

- Calculated/measured Resolution
  3 meV (high efficiency)
- Overall length = 2 meters.
- Spectrograph for Merlin beamline
  (completion summer 2007)

Spectrograph: Energy Resolution Test

Straight beam with ~ 6μm source size BL 12.0



Acknowledgement

- **Ambient Photoemission:** Simon Mun, Charles Fadley…
- **ARPES:** ZX Shen(Stanford), Norman Mannella, Yulin Chen, Eli Rotenberg, Dan Dessau, Alexei Fedorov…
- **Spin Polarized Photoemission:** Chris Jozwiak, Gennadi Lebedev, Slim Chourou, Jeff Graff, Andreas Schmid, A. Lanzara (UC Berkeley/LBNL), Boris Sinkovic, Nord Andresen, Alexie Fedorov
- **MERLIN Beamline:** Yi De Chuang, Ruben Reininger, Malcolm Howells, John Bozek, Nicholas Kelez, Keith Franck, Rob Duarte, Tony Warwick, A.Lanzara, Zahid Hasan (Prineton)

Thank you for your attention