Advances in Photoelectron Diffraction and Holography

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Relevance of photoelectron diffraction and holography

- geometric structure
- electronic structure, bonding
- magnetism
- low-dimensional systems
- holography (incl. theory as “artificial experiment”)
- XAFS, NEXAFS

Developments in PED theory

- photon polarization
  - linear, circular, unpolarized
- electron spin
  - spin-orbit, magnetic, spin-polarized holography
- multiple scattering
  - iteration, recursion, simultaneous & successive relaxation
- full potential (non-muffin-tin)
  - multiple scattering formalism
- electron hole
  - Coulomb waves
- Multi-Atom Resonant Photoemission (MARPE)

EDAC

F. Javier García de Abajo - EDAC - Netscape

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Electron Diffraction in Atomic Clusters

for Core Level Photoelectron Diffraction Simulations

Welcome to the EDAC home page.
This site allows performing on-line photoelectron diffraction calculations.
Multiple scattering (MS) of the photoelectron is carried out for a cluster representing a solid or molecule.
Scattering phase shifts and excitation radial matrix elements are calculated internally and there is no need for the user to provide them.

EDAC has been developed by F. J. García de Abajo in the framework of the collaboration between LBNL (Berkeley, California) and the Centro Mixto CSIC-UPV/EHU (San Sebastián, Spain). For theoretical details and application examples, see F. J. García de Abajo, M. A. Van Hove, and C. S. Fadley, submitted to Phys. Rev. B.

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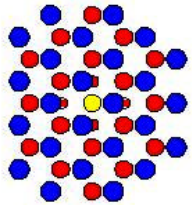
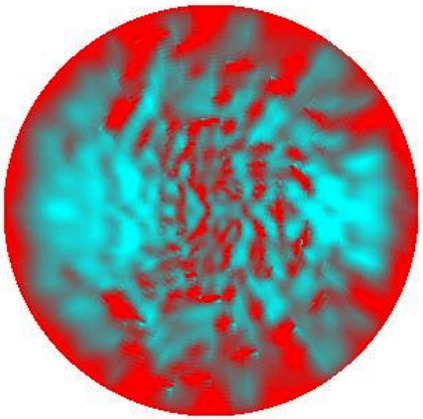
EDAC output for O/W(110)-(1x1)

Click on the figure to download data.



Left: representation of the cluster rocking around a line parallel to the *z* direction and passing by the **emitter (yellow atom)**. The dashed lines stand for the *xyz* axes. **Right:** top view of the cluster, where the *x*/*y* direction (not plotted) runs along the horizontal/vertical screen direction. Different atomic species have been assigned the colors O, W.



Polar scan of photoemission intensity (logarithmic scale). Blue/ Red regions correspond to high/ low intensity. The orientation is the same as in the top-view of the cluster. The distance to the center of the figure is proportional to the polar angle θ. The polar angle range is (0.0, 89.0) (in degrees).

Parameters used in the calculation:

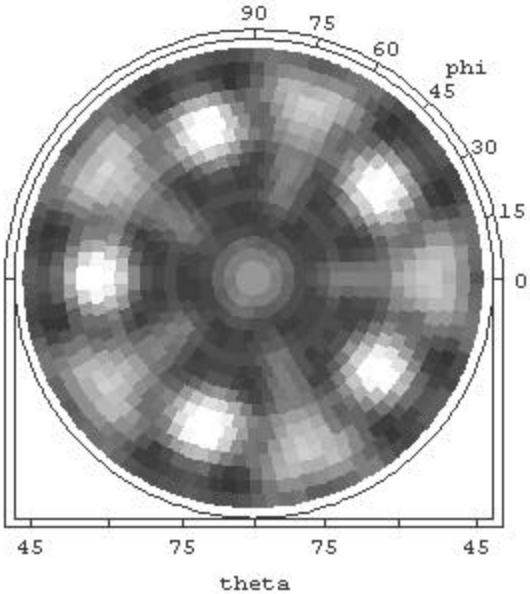
N=48 atoms
Scat. order=2
l_{max}=6
V_0=10.5 eV
K.E.=150 eV
p-pol. light
z_0=2.3 Å
i.m.f.p.=6 Å
Recursion method

Document: Done

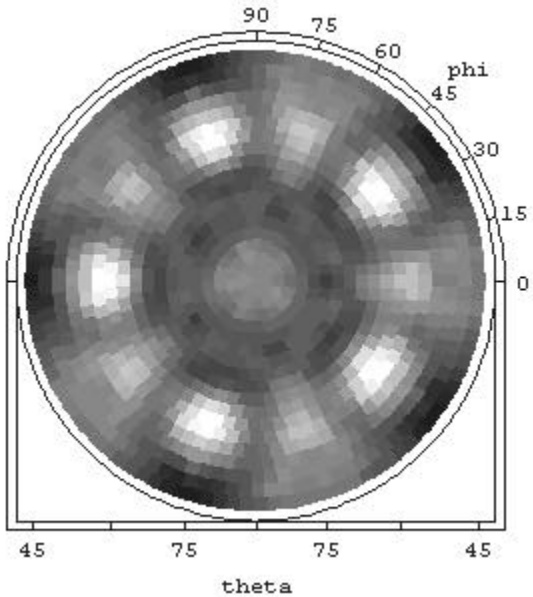
Multiple-scattering PED codes

- **MSCD** (R. Díez Muiño, F. Bondino)
  - photon polarization: linear, circular, unpolarized
  - domain averaging
  - full (non-spherical) potential
  - electron hole
- **EDAC** (F.J. García de Abajo)
  - new fast convergence (also for LEED)
  - photon polarization
  - spin polarization: relativistic (spin-orbit), magnetic (exchange)
  - small-cluster non-spin version <u>now interactive on-line</u>
- **will be available at http://electron.lbl.gov/**

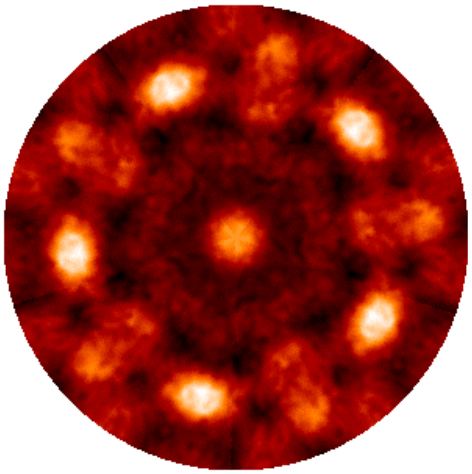
XPD AlPdMn - theory vs experiment



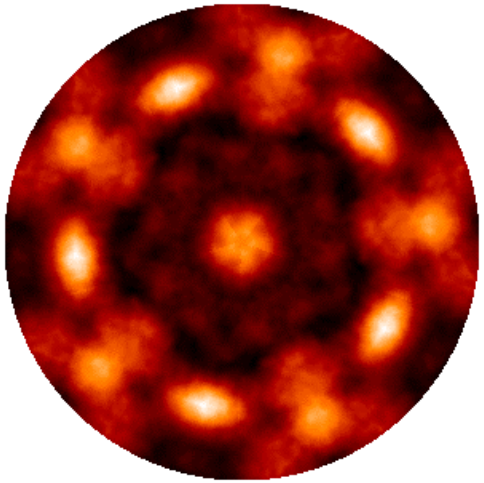
Al 2p th.



Pd 3d th.

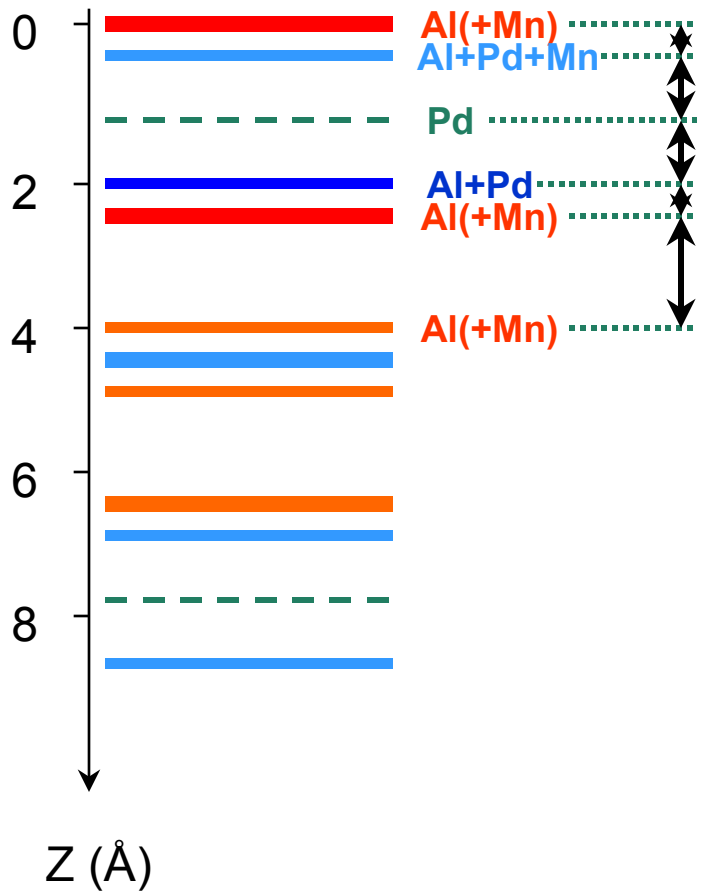


Al 2p exp.



Pd 3d exp.

XPD AlPdMn - PED vs LEED vs bulk interlayer spacings (Å)

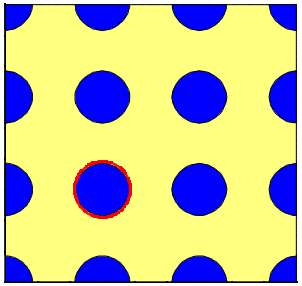


PED	LEED	(bulk)
0.29	0.38	(0.47)
0.60	0.75	(0.76)
1.14	0.85	(0.76)
0.54	0.47	(0.47)
1.56	1.56	(1.56)

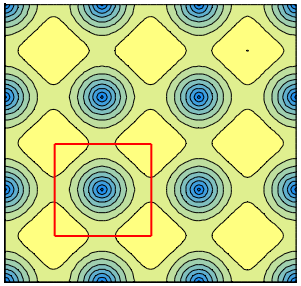
Valence PED

- **low final-state energy**
  - full (non-spherical) potential
  - electron hole
  - sensitive to bonding electrons
  - test on free oriented CO and N_2 molecules
- **valence level as initial state**
  - next step!
  - also sensitive to bonding electrons
  - coherent emission across surface
  - developing efficient methods with Schattke et al

Motivation: Non-spherical effects in the scattering of photoemitted electrons



standard muffin-tin approximation: spherical cells



non-spherical cells

$j_l(r) \longrightarrow$ $j_l(r) + t_l\, h_l^+(r)$

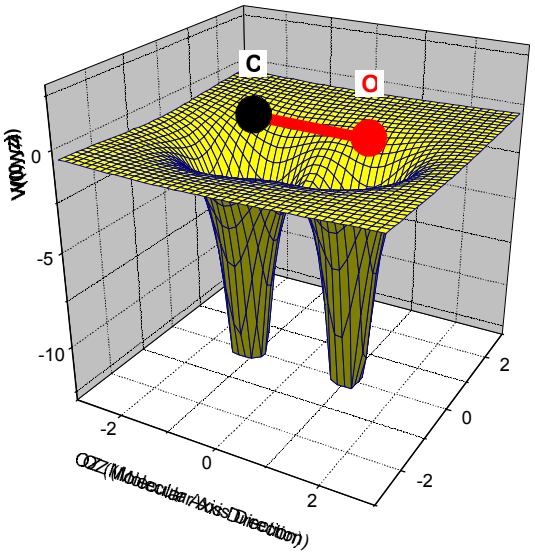
$j_l(r) \longrightarrow$ $j_l(r) + \Sigma[t_{lml'm'} h_{l'}^+(r)\, Y_{l'm'}(\Omega_k)]$

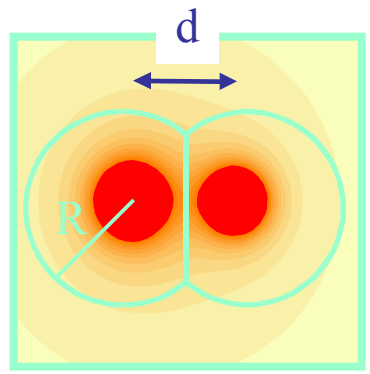
every partial wave scatters off the potential independently: phase shifts δ_l

$t_l = [1-\exp(2i\delta_l)]$

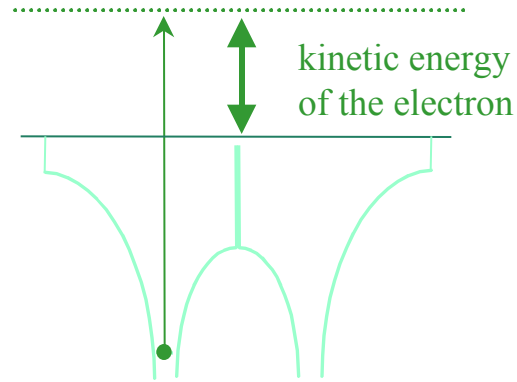
coupling between different partial waves: non-diagonal scattering T-matrices

Partitioning of potential

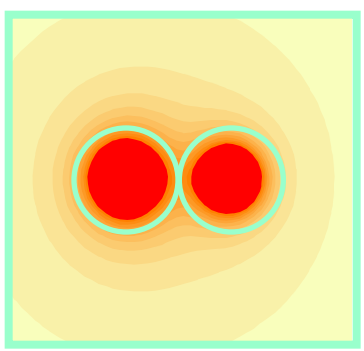




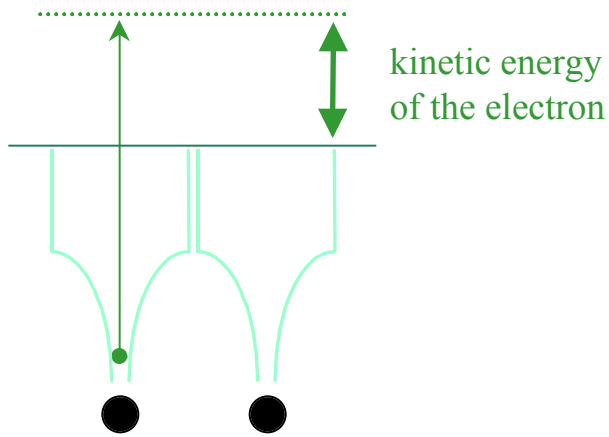
non-spherical potentials
R<d



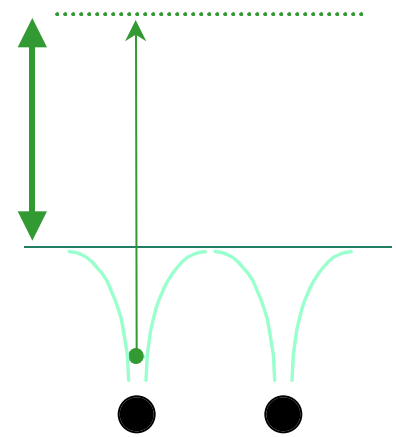
non-spherical multiple scattering
(NSMS)



spherical potentials



muffin-tin
multiple scattering
(MTMS)

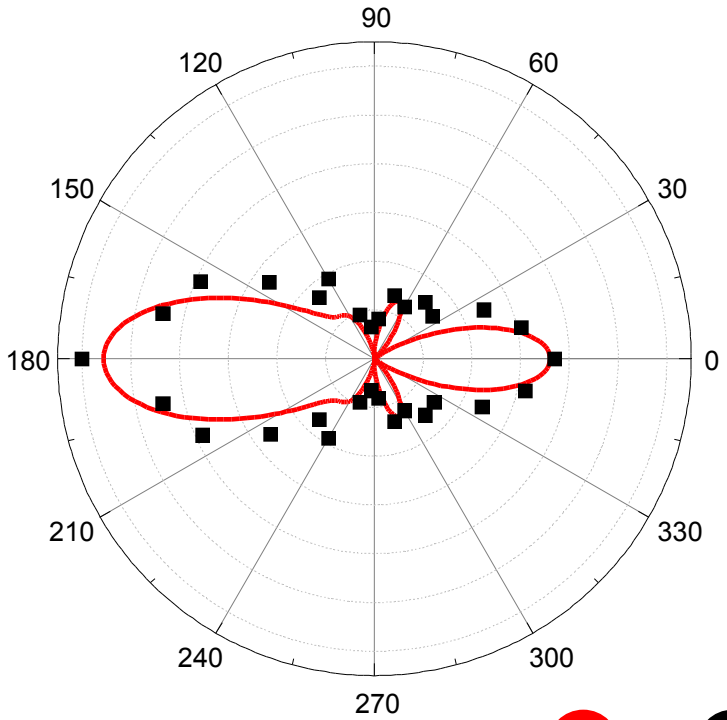


spherical atomic
multiple scattering
(SAMS)

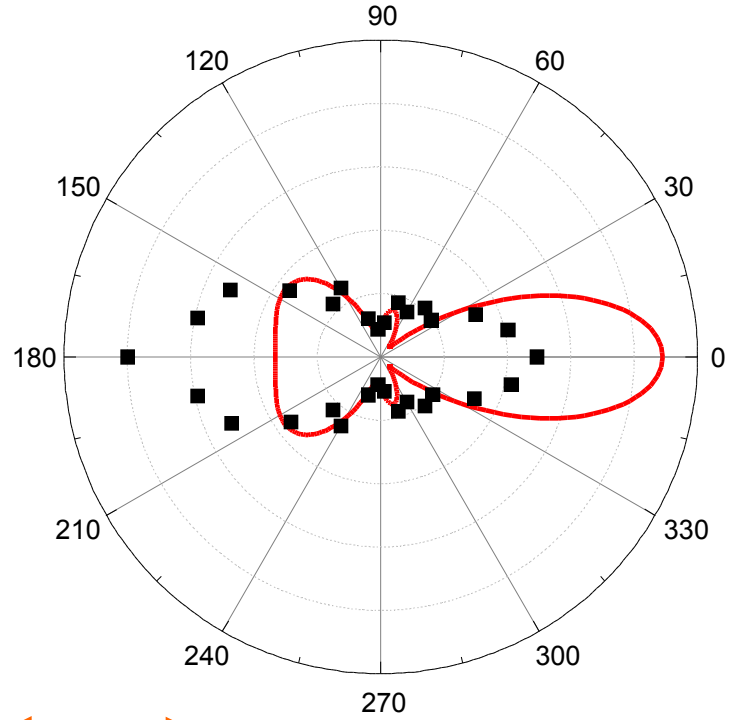
Effect of the Coulomb hole: Photoemission from C1s in CO

kinetic energy of the electrons E_k=21 eV

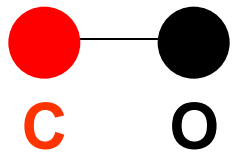
Coulomb potential included locally



Coulomb potential NOT included



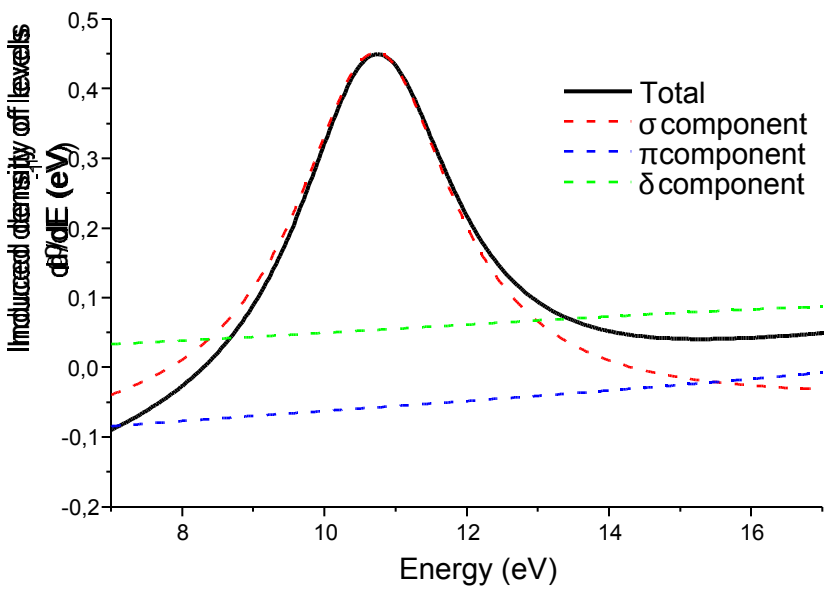
■ Experimental data: Landers *et al.*



light polarization

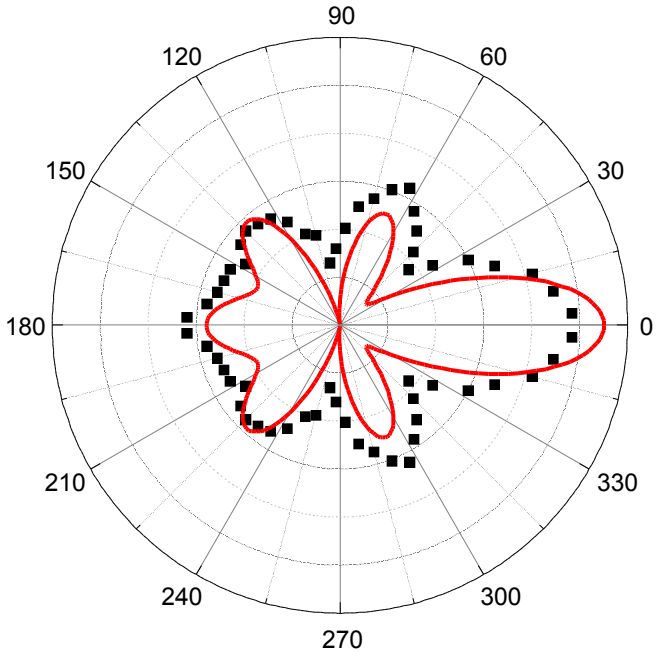
σ-shape resonance: Photoemission from C1s in CO

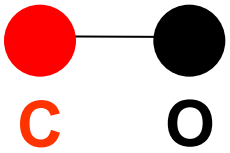
kinetic energy of the electrons E_k=10.4 eV



Density of levels induced in the continuum
by the CO^+ molecule:
the shape resonance is clearly seen,
and it shows a strong σ-character

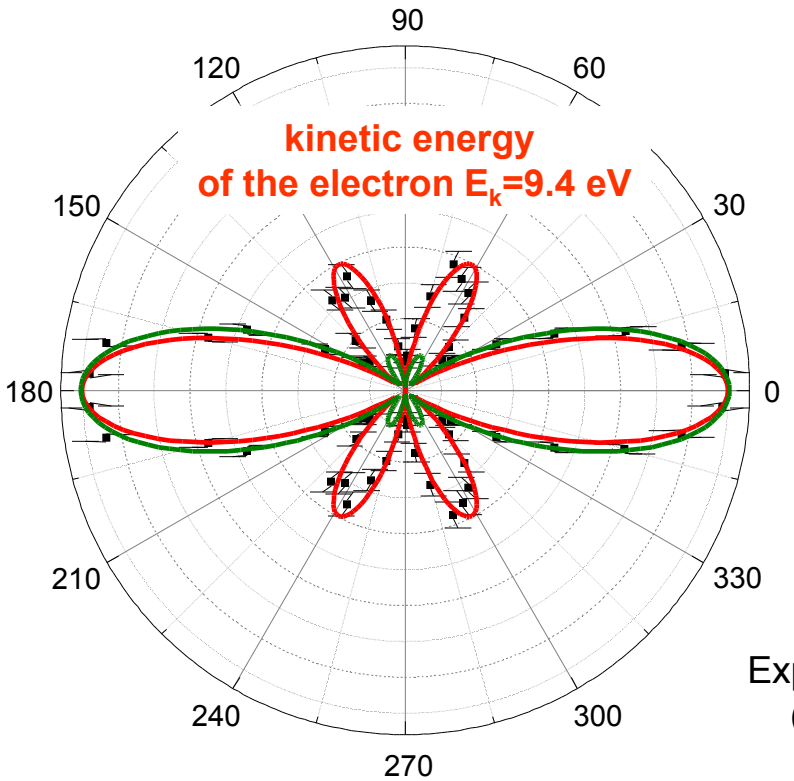
■ Experimental data:
Landers *et al.*

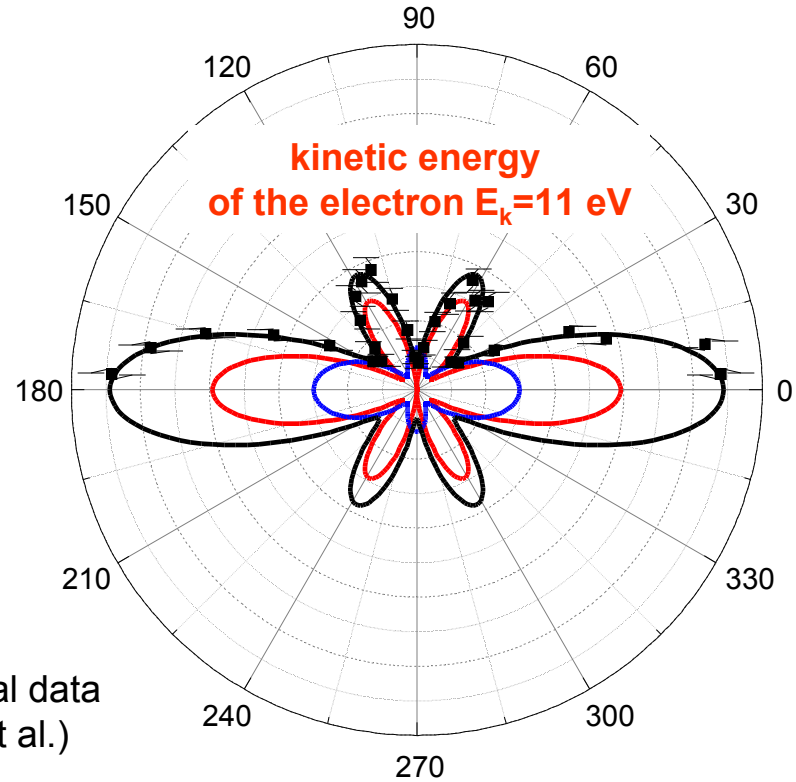




light
polarization

Shape resonance in homonuclear molecules: Photoemission from N1s in N_2





Experimental data (Weber et al.)

Non-Spherical Multiple Scattering (NSMS)

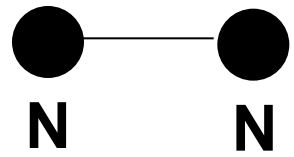
Spherical Atomic Multiple Scattering (SAMS)

Incoherent sum

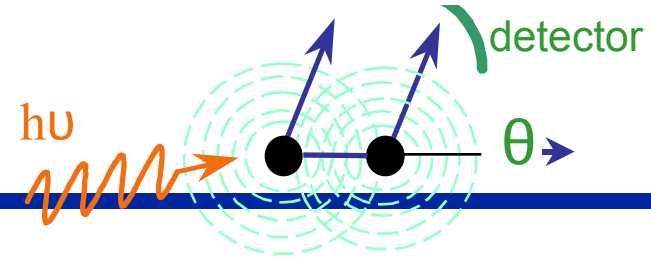
Contribution from symmetric orbital

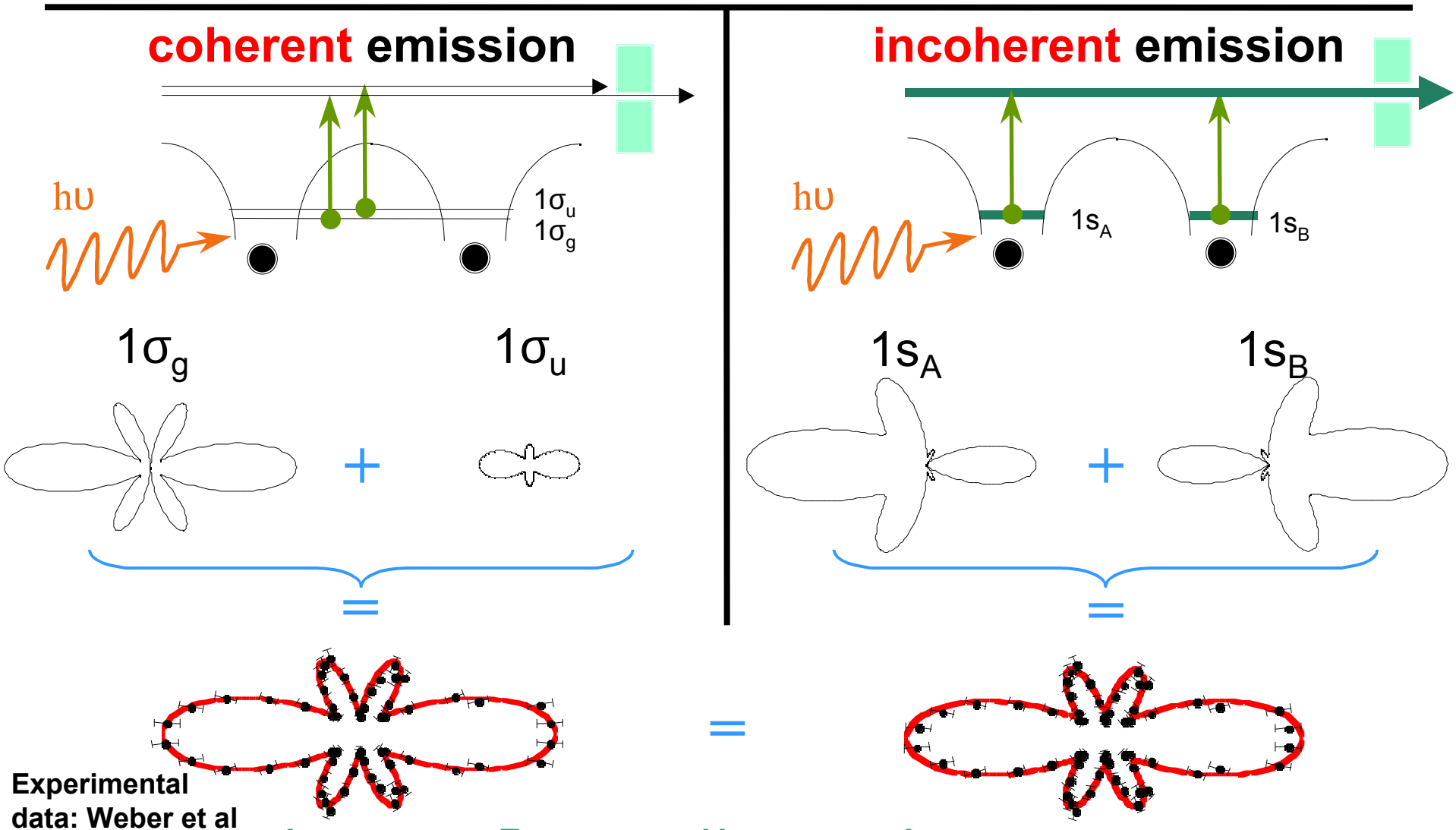
Contribution from antisymmetric orbital

light polarization



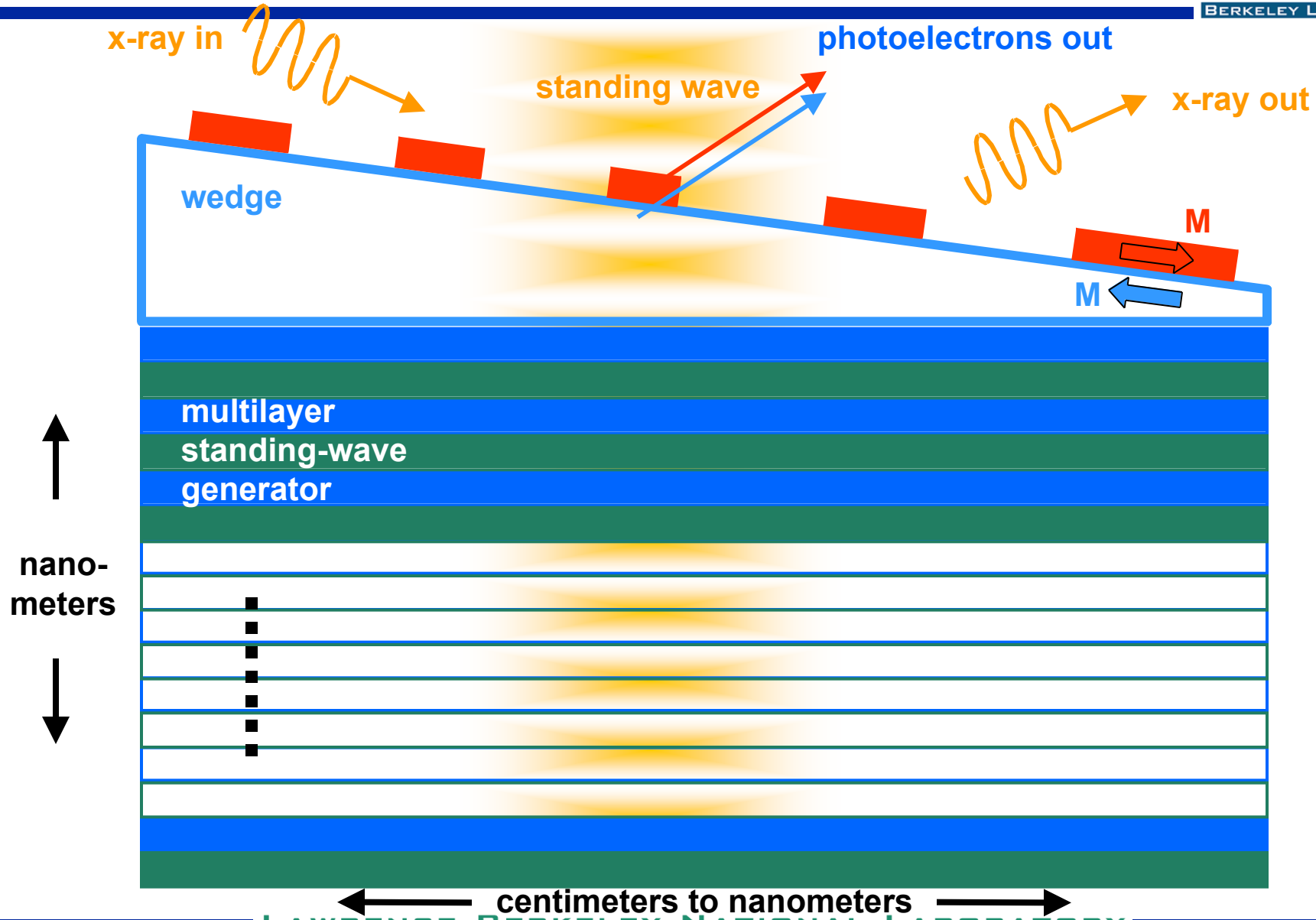
Coherence vs. incoherence in photoemission: N_2





Experimental data: Weber et al

probing nanoscale composition, structure and magnetism by scanning along wedge

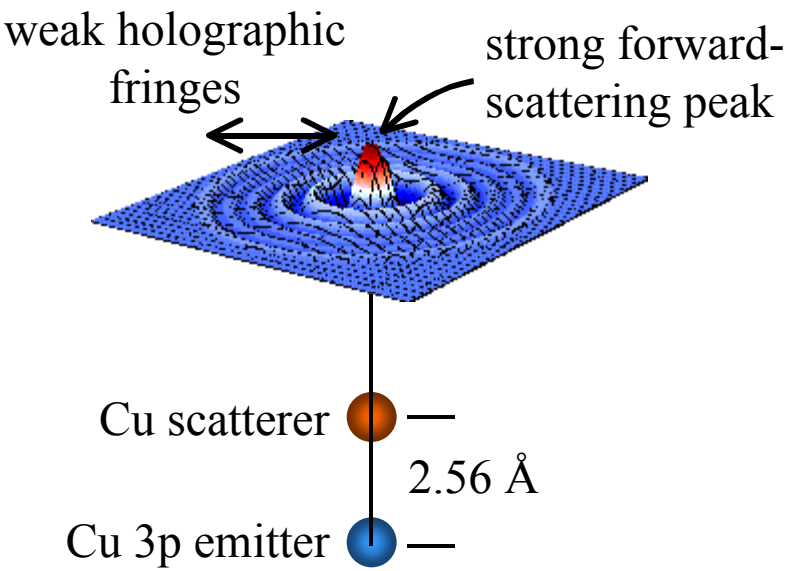


(after Yang et al)

Holography by contrast

- **Spin-Polarized Photoelectron Holography (SPPH)**
  - subtract spin-down from spin-up hologram
  - can give image of local magnetic order
- **Differential Photoelectron Holography (DPH)**
  - differentiate rel. to energy (k) to suppress forward-scattering peaks
  - gives cleaner holographic reconstruction
- **Resonant X-ray Fluorescence Holography (RXFH)**
  - subtract holograms taken across resonance
  - filters out non-resonant atoms
- **in general: look for contrast due to small changes**
  - image magnetism, relaxations, chemical substitution, …

Forward-Scattering Problem in PEH



Photoelectron hologram for the Cu-Cu nearest pair at E_k=323 eV (k=9.2 Å).

"Non-Optical" Electron Scattering

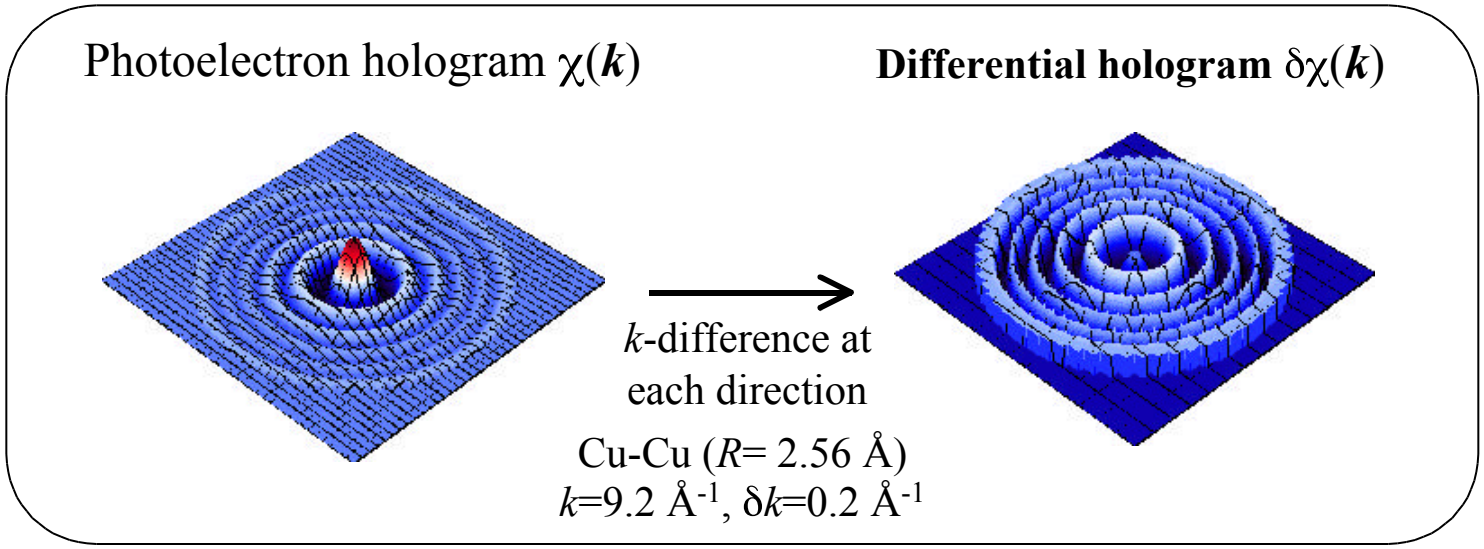
✓Large scattering cross section

✓Anisotropic scattering amplitude: forward-scattering for E_k> 100~200 eV

✓Scattering phase shift

✓Multiple-scattering

Idea of Differential Holography

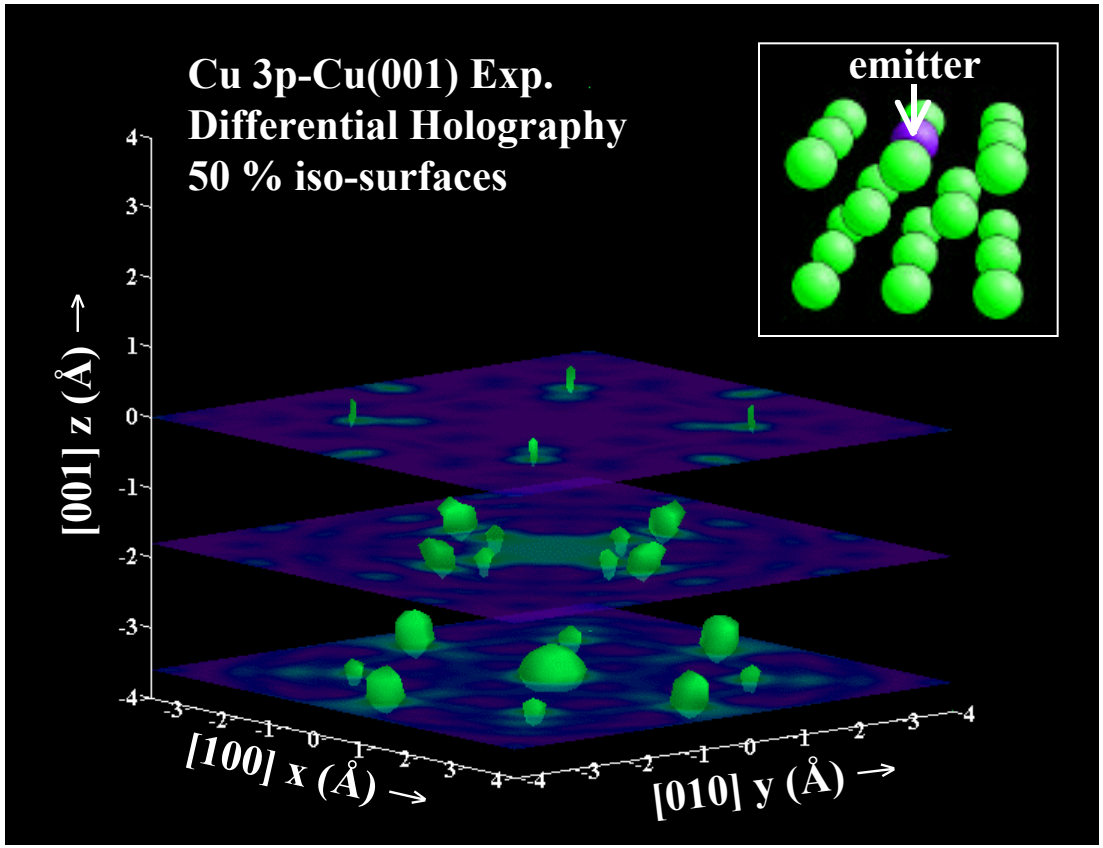


Owing to the lack of *k*-dependence of forward-scattering peaks, they can be removed by taking the difference of two holograms at slightly different *k*.

On the other hand, holographic oscillations in the form of cos[*kr*(1-cosθ)] will survive this subtraction because their phases are quite sensitive to the variation in *k*.

Therefore, by simply replacing χ by its *k*-derivative or *k*-difference δχ, the forward-scattering effects should be greatly suppressed.

3D Image of Cu(001) by Differential Holography



Three-dimensional Cu(001) atomic image reconstructed from the experimental hologram measured at ALS-BL7 by differential holography. Observable side-scattering and back-scattering atoms are suggested in the inset.