Full k-Space Visualization of Photoelectron Diffraction

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INTRODUCTION

The development of photoelectron holography has promoted the need for larger photoelectron diffraction data sets in order to improve the quality of real-space reconstructed images (by suppressing transformational artifacts and distortions) [1-3]. The two main experimental and theoretical approaches to holography, the transform of angular distribution patterns for a coarse selection of energies or the transform of energy-scanned profiles for several directions, represent two limits to *k*-space sampling. The high brightness of third-generation soft x-ray synchrotron sources provides the opportunity to rapidly measure large high-density x-ray photoelectron diffraction (XPD) data sets with approximately uniform *k*-space sampling. In this abstract, we present such a photoelectron data set acquired for Cu 3*p* emission from Cu(001) [4].

Cu(001) is one of the most well-studied systems for understanding photoelectron diffraction structure [5] and for testing photoelectron holography methods [6-8]. Cu(001) was chosen for this study in part due to the relatively inert and unreconstructed clean surface, and it served to calibrate and fine-tune the operation of a new synchrotron beamline, electron spectrometer and sample goniometer [9]. In addition to Cu, similar 'volume' XPD data sets have been acquired for bulk and surface core-level emission from W(110), from reconstructed Si(100) and Si(111) surfaces [10], and from the adsorbate system of c(2x2) Mn/Ni(100) [11].

EXPERIMENT

Measurements were performed at the SpectroMicroscopy Facility (Beamline 7.0). The photoelectron diffraction system consists of a high precision two-axis goniometer for sample rotations and a PHI Omni 4 hemispherical electron spectrometer with multi-channel detection and variable-aperture input lens set for an angular acceptance less than ±1°. For rapid scanned-angle XPD measurements of bulk Cu 3*p*, a 0.4 eV wide energy-window was used to take 'snapshots' of the peak and higher kinetic energy background intensities at each sample direction. With dwell times less than 1 second per angle, a 482 point angular intensity pattern could be obtained in less than 15 minutes including ~50% motor motion overhead. Angular distribution patterns were acquired within a 45° azimuthal sector and then mirror- and four-fold symmetrized for Cu(001) to display the full angle hologram. Uniform increments in polar angle to 80° from normal emission and constant solid angle azimuth steps (inversely proportional to polar angle) were used to uniformly sample directions. Automated experimenter control of the undulator gap allows scanned-photon energy experiments to be routinely performed; however, the greater time overhead for undulator and monchromator exit slit motions make this data acquisition mode less efficient. For this reason the large Cu 3*p* XPD data set was acquired by sequential measurement of constant-energy angular distribution patterns.

RESULTS

The full Cu $3p$ XPD data set is represented as a cylindrical volume in Fig. 1. The cylinder is created by stacking 58 angular patterns acquired for an electron momentum range of 3.1 to 8.8 $Å^{-1}$ with 0.1 $Å^{-1}$ steps (37-295 eV kinetic energy range). The full volume represents greater than 185,000 intensities, resulting from the symmetrization of ~28,000 measurements acquired in less than three 8-hour synchrotron shifts. The angles and energies in Figures 1-3 correspond to coordinates outside the crystal and have not been corrected for refraction at the surface due to the crystal inner potential. Cutaways in the cylindrical volume in Fig. 1 show the interior diffraction structure. To highlight the richly detailed intensity variations, contrast has been enhanced by (a) correcting each pattern for inelastic attenuation near grazing emission, (b) normalizing each pattern to a constant integrated intensity, and (c) saturating the intense low kinetic energy peak at normal emission.

A second way of visualizing the data set is by playing a movie sequence of each diffraction pattern. Figure 2 shows a much coarser sampling of ten angular diffraction patterns extracted from the data volume. Energies with ~0.7 $Å^{-1}$ separation have been chosen to maximally illustrate the wide variety of angular patterns. A third way of visualizing the data set is by extracting energy dependent slices at fixed azimuths. Figure 3 shows energy-polar angle slices in two high symmetry azimuths separated by 45°. The slices are additionally mapped into a k_x-k_z coordinates in order to illustrate the polar emission angle.

Large intensity oscillations along the 45° polar emission angle in the {100} planes are observed for energies greater than 100 eV. These energy oscillations are recognized as arising from

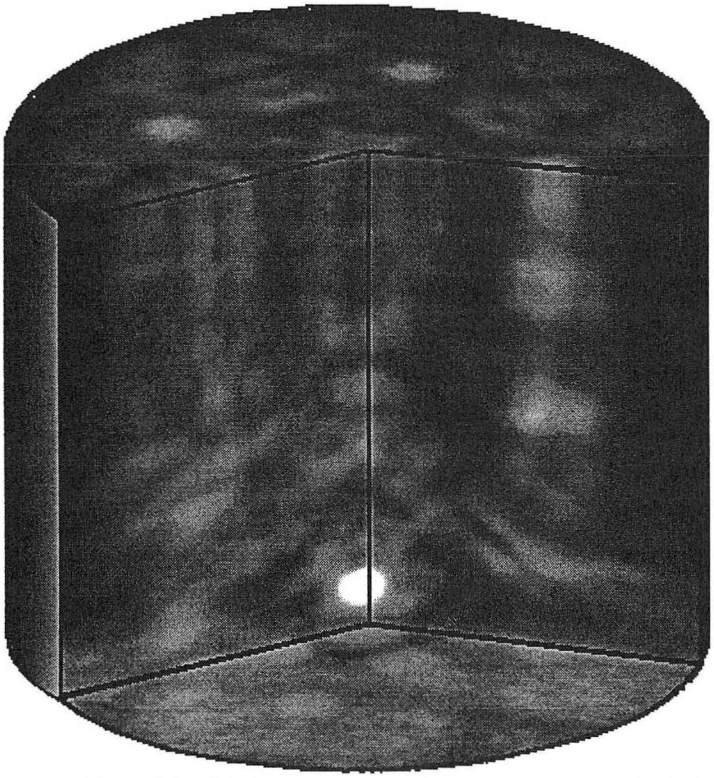

Figure 1. Cylindrical representation of the full Cu(001) Cu $3p$ volume XPD data set with cutaways to show interior diffraction structure. The vertical dimension is linear in electron momentum from k=3.1 to 8.8 Å-1. The radial dimension is linear in polar emission angle from 0° to 80°.

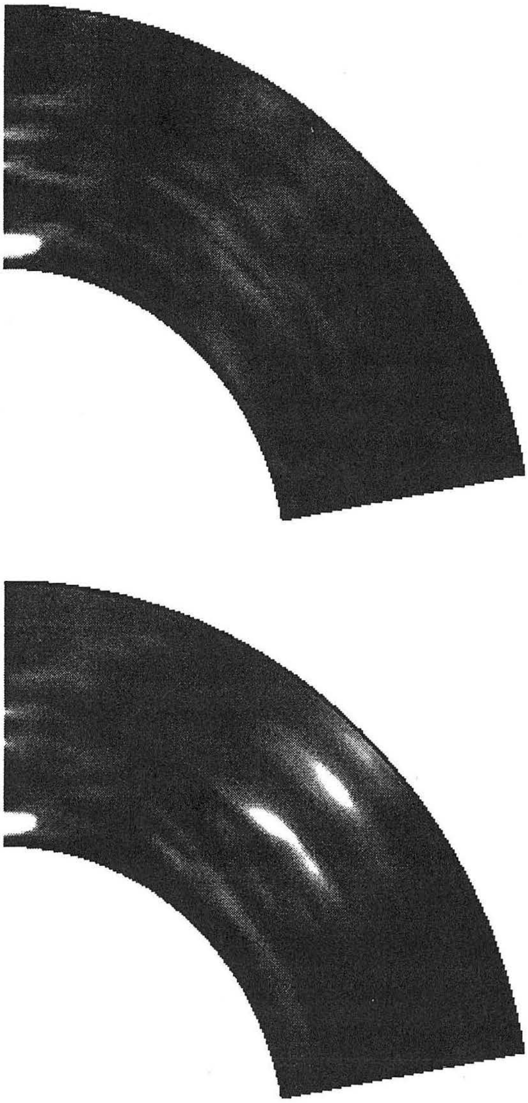

Figure 3. (above) Energy-dependent slices in the [110] (top) and [100] (bottom) emission planes mapped into *k*-space.

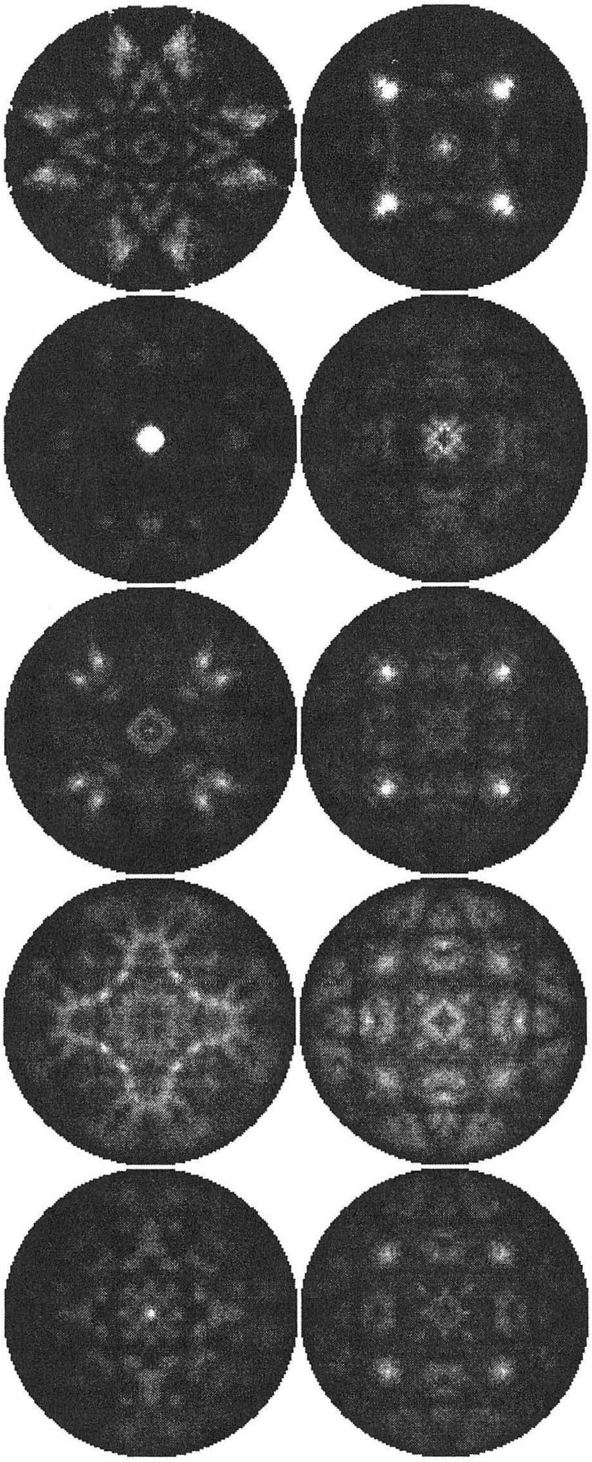

Figure 2. (left) Sampling of ten angular distribution patterns with kinetic energies: 39, 52, 67, 84, 111, 142, 176, 214, 250 and 295 eV.

back-scattering from nearest-neighbor atoms [12] along the [101] crystal axis. The path length difference for back-scattered photoelectrons (twice the near-neighbor bond distance) determines the dominant *k*-space period of the oscillations. The (100) energy slice in Fig. 3 also illustrates the angular width (~20°) of this cone of back-scattered intensity. For kinetic energies above 200 eV, the angular patterns in Fig. 2 begin to show the consistent appearance of arcs resulting from emission from atoms in deeper layers below the surface and Kikuchi-like scattering from atoms lying in low-index crystal planes [5].

Below 100 eV, rapidly varying diffraction structure is observed both in the dramatic changes in the angular patterns and in the outwardly sweeping intensity for decreasing energy in the energy slices. Additionally, an intense normal emission peak at 50 eV kinetic energy is observed. Contributing factors to this low energy diffraction behavior include: (i) the enhanced importance of multiple-scattering, (ii) rapid changes in the electron scattering factor, (iii) increased surface sensitivity, and (iv) the increasing effect of surface refraction. The detailed origins of these low energy features are being explored with single- and multiple-scattering calculations.

CONCLUSIONS

The continuous energy and angular variation of photoelectron diffraction intensities has for the first time been experimentally measured and visualized. The *k*-space range for the example presented, Cu 3*p* emission from Cu(001), illustrates the progression from low energies dominated by multiple-scattering to medium energies exhibiting strong back-scattering features. The detailed explanation of the rich diffraction structure will provide a test for scattering simulations. In addition, the data will be used as an experimental test bed for real-space reconstruction algorithms, where optimal *k*-space sampling, energy range selection, algorithm-specific artifacts and other issues are of concern [8].

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