Holographic Atomic Images from Surface and Bulk W(110) Photoelectron Diffraction Data

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Although Gabor originally proposed using electrons in holography to form atomic-resolution images [1], it is only very recently that photoelectron diffraction data have been discussed as a practical method for achieving this [2,3]. Such data enable recording both the amplitudes and phases of the scattered waves (relative to a direct reference wave), thus in principle permitting the holographic reconstruction of atomic positions [2,3]. Photoelectron holography thus holds much promise of at least providing approximate starting structures that can be refined by more conventional comparisons of multiple-scattering calculations with experiment via *R*-factors. Criteria for optimizing the taking of such data and their holographic inversion have recently been published [4]. In the present study, such holographic photoelectron diffraction patterns have been measured for surface and bulk core-level-shifted W $4f_{7/2}$ photoemission from W(110) on ALS beamline 7.0, yielding a data set of unprecedented size and quality. These data have been compared to multiple scattering theory [5], and used as a test case, since the W(110) surface is known not to exhibit significant relaxation in its interlayer spacings relative to the bulk [6]. We have analyzed these experimental and theoretical results holographically in order to demonstrate the capabilities and limitations of photoelectron holography as a structural probe.

The experimental geometry at ALS beamline 7.0 is shown in Fig. 1(a). The W $4f_{7/2}$ photoelectron peak can be resolved into surface and bulk core-level-shifted components, as shown in Fig. 1(b). For each energy E (or wavenumber k) and direction studied, the W $4f_{7/2}$ peak was divided into surface and bulk emission intensities by integrating the areas under the higher-energy and lower-energy flanks of the two W $4f_{7/2}$ peaks, respectively, as shown by the shaded areas in Fig. 1(b). The brightness of the ALS undulator radiation permitted taking one of these spectra in ~10 sec, and doing the simpler area integration over bulk, surface, and high-energy background in ~1 sec.

Figures 2 and 3 respectively show the raw surface and bulk $I(\mathbf{k})$ data sets in **k**-space, for kinetic energies in vacuum of E = 39eV to 309eV (wavenumbers $k = 3.2\text{Å}^{-1}$ to 9.0Å^{-1}), for a polar takeoff angle range of $10° \leq \theta \leq 90°$(≡ surface normal). A total of ~20,000 unique intensities in the symmetry-reduced 1/4th of the solid angle above the sample was measured.

Figures 4(a) and 5(a) show holographic images reconstructed from the normalized surface and bulk $\chi(\mathbf{k})$ data sets of Figs. 2 and 3 in the vertical $(\bar{1}1\bar{2})$ plane, using the original multi-energy algorithm proposed by Barton [3a], and Tong and co-workers [3b]. The emitter position is indicated by a dashed square, and the ideal positions of the neighboring atoms are indicated by circles.

1. Experimental details

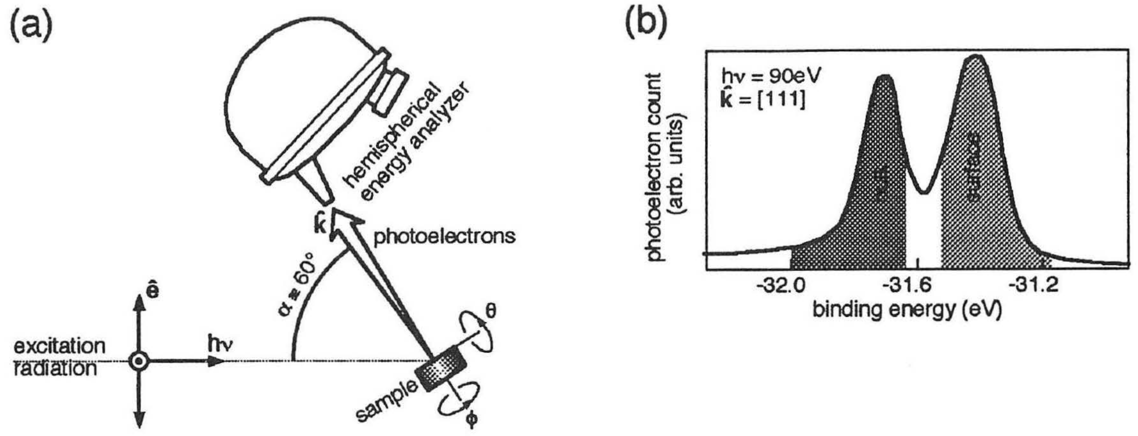
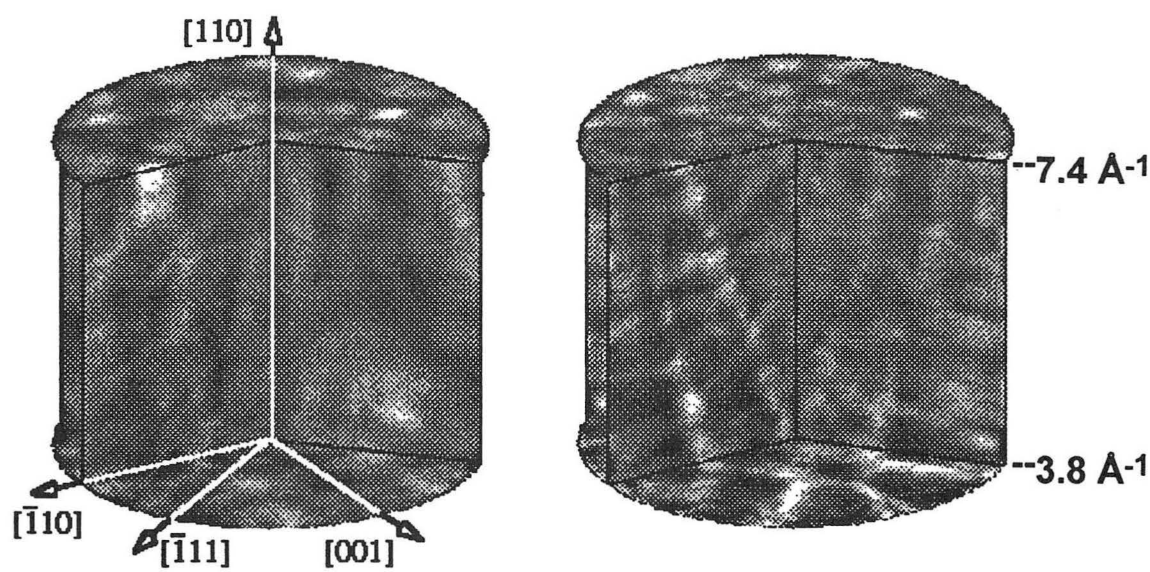


Figure 1. (a) Experimental geometry, including the orientation of the sample (where $\hat{\mathbf{n}}$ is the surface normal) with respect to the horizontal polarization vector ($\hat{\mathbf{e}}$) of the incident excitation radiation $h\nu$, and the exit photoelectron direction. (b) Typical W $4f_{7/2}$ x-ray photoelectron spectrum from W(110), indicating the surface and bulk core-level-shifted contributions used to generate the $I(\mathbf{k})$ intensity data points of Figs. 2-3.

2. Surface

3. Bulk



Figures 2-3. Volume representations of the normalized intensity data sets $I(\mathbf{k})$ for W $4f_{7/2}$ emission: 2 = surface and 3 = bulk. The vertical scale is wavenumber varying from 3.8Å^{-1} to 7.4Å^{-1} (corresponding to 56 eV to 211 eV in energy) and each plane is a stereographic projection of a nearly full-hemisphere data set taken over 0°≤θ≤80° and φ varying over 360°. The energies and polar angles have here all been adjusted to be inside the surface, using an inner potential of V_0 = 14eV.

Figures 4(b) and 5(b) show the equivalent images reconstructed from a fully-converged multiple scattering calculation [5]. There is in general good agreement between experiment and theory. Both surface images very well resolve the backscattering atom below the emitter, and reasonably well resolve the forward scattering atoms above the emitter. The two bulk images exhibit artifactual peaks near the emitter, but agree in the general elongated and slightly doubled shape of the forward scattering images, which are weaker in experiment as compared to theory.

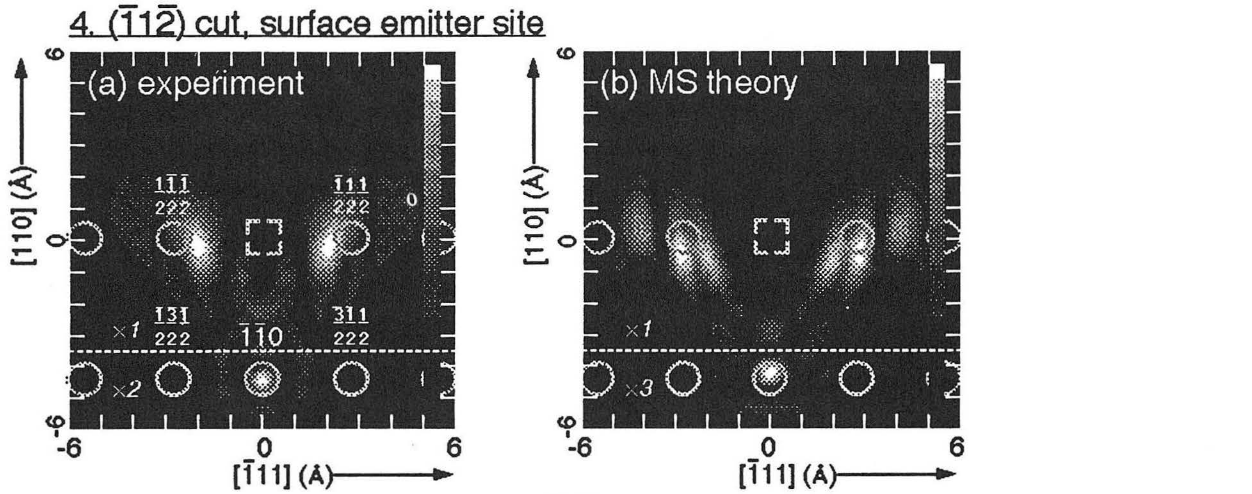


Figure 4. W(110) atomic images in the vertical ($\bar{1}1\bar{2}$) plane, as reconstructed from (a) the experimental W $4f_{7/2}$ surface-emission data set of Fig. 2; and (b) corresponding multiple scattering calculations. The surface emitter site at the origin is indicated by a dashed square, and the positions of the scatterers (assuming no surface relaxation) are indicated by circles. The nearest and next-nearest scattering positions have been labeled in panel (a). Axes are marked off in 1Å units. Image intensities for $z \leq -3.5$Å have been rescaled, with the scale factors indicated on the figures.

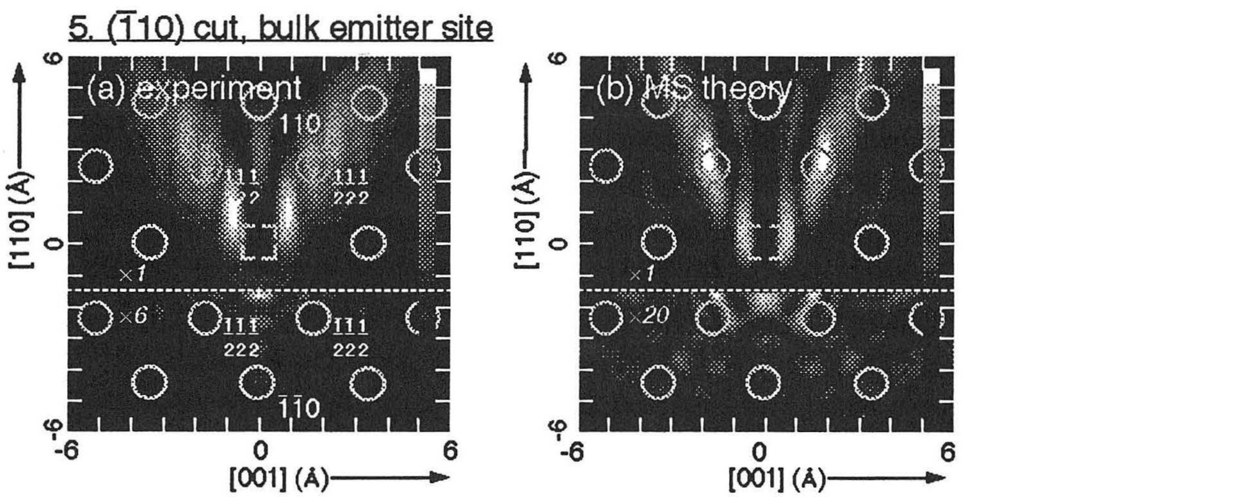


Figure 5. As Fig. 4, but for the bulk emission data set of Fig. 3, ($\bar{1}10$) plane, and its corresponding multiple scattering simulation. Image intensities for $z \leq -1.5$Å have been rescaled, with the scale factors indicated on the figures.

Although encouraging in several respects, particularly for the case of the surface emitters, both the experimental and theoretical images of Figs. 4-5 suffer from image aberrations and artifacts arising from anisotropies in the photoelectron reference wave and the resultant scattered waves, and possible residual multiple scattering effects. Several modified reconstruction algorithms have been proposed to account for the anisotropies mentioned, and these are found to exhibit varying degrees of success in improving such holographic images [4a,4b], including those for the present data [7]. Although further research into improved imaging algorithms and more tests against experimental data for other reference cases would certainly be worthwhile, the present data and analysis indicate that photoelectron holography can provide good-to-excellent *ab initio* estimates of the positions of the atoms beside and below surface emitters, and fair-to-good estimates of the positions of the atoms above bulk emitters. Such estimates could then be refined, using *R*-factor comparisons of experiment with model diffraction calculations for various structures.

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