

High-energy photoelectron diffraction: model calculations and comparison to first experimental data

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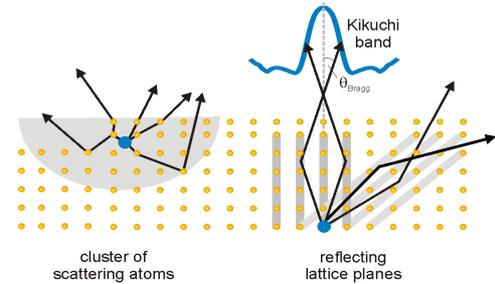
F Javier Garcia de Abajo

MOTIVATION

Photoelectron spectroscopy using hard X-rays (HXPES, HAXPES) can provide access to bulk stoichiometry and electronic structure of materials, due to the large inelastic mean free path of electrons at high kinetic energies (5-15nm at 10keV). Diffraction of such high energy photoelectrons (HXP) should provide additional element-selective crystallographic information. We show that such high-energy photoelectron diffraction can be quantitatively modelled using the dynamical Bloch wave theory of high-energy electron diffraction [1]. The HXP patterns are dominated by Kikuchi bands formed by Bragg reflection of photoelectrons at bulk lattice planes. Our approach makes HXP simulations computationally feasible at high energies and agrees with multiple-scattering cluster calculations [2] when compared to experimental XPD patterns at ~1keV. We then use these calculations to predict structural sensitivity in future HXP experiments, as well as to describe some first experimental HXP data from W(110).

PHOTOELECTRON DIFFRACTION

The scattering of photoelectrons from localized sources can be described in real space (multiple scattering cluster) and reciprocal space (dynamical theory of electron diffraction)

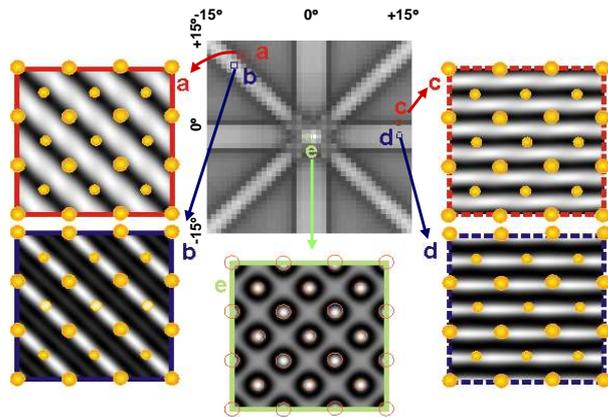


BLOCH WAVES AND KIKUCHI BANDS

Diffacted wave function is the sum of Bloch waves in the bulk 3D crystal:

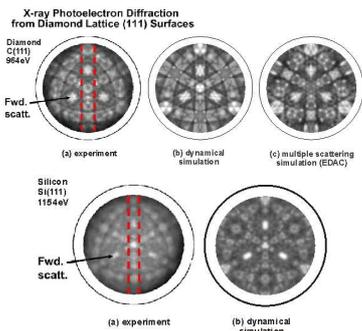
$$\Psi(\vec{r}) = \sum_j c_j \exp(i\vec{k}^{(j)} \cdot \vec{r}) \sum_{\vec{g}} C_{\vec{g}}^{(j)} \exp(i\vec{g} \cdot \vec{r})$$

Top middle panel: simplified nine-beam HXP Kikuchi pattern from an Fe bcc lattice at 15 keV kinetic energy and for an angular range of $\pm 15^\circ$ from the [001] surface normal. The areas marked by colored squares and letters a-e in the Kikuchi pattern each correspond to a specific detection direction of either high or low intensity. The other panels then show the (x, y) probability density distribution in the surface plane corresponding to (a)-(e) for 3x3 unit cells averaged along the z-axis [001] (white = high probability of ending up in the plane wave to the detection direction and black = low probability). The photoelectrons are created via localized excitations at the atomic positions (colored circles) and are diffracted into the detection directions with an intensity proportional to the overlap with the probability density of the diffraction process. The central lower pattern (e), green border) corresponds to the forward-scattering direction of the surface normal and shows how the symmetry of the lattice can confine the electrons to a channel along the zone-axis direction. The results in the other panels (a)-(d) explain the high or low level of intensity along the associated directions.

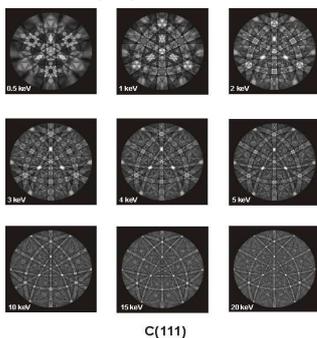


APPLICATIONS OF THE THEORY

Comparison to experiments at 1keV

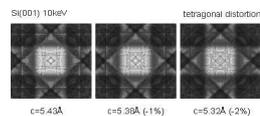


Simulation of high energy photoelectron diffraction from Diamond C(111)



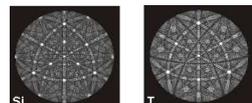
Predicted sensitivity of HXP to structure

Tetragonally distorted Silicon



Calculated photoelectron intensity $\pm 5^\circ$ away from the surface normal of Si(001) at 10 keV showing a fine structure which is very sensitive to a possible tetragonal distortion. The left panel corresponds to the cubic Si unit cell; for the other panels, the unit cell has been compressed parallel to the surface normal along the c-axis by 1% (middle) and 2% (right). Changes are clearly noticeable at 1% distortion.

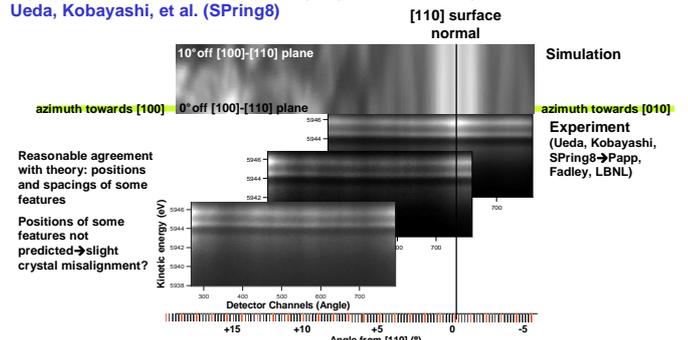
Impurity sites in Silicon



Calculated photoelectron diffraction pattern for a Si(111) surface at 6 keV photoelectron kinetic energy from impurities located in different sites in the Si lattice. Left: impurity located at the substitutional (Si) sites, right: impurity located at the tetrahedral interstitial (T) sites. Note the missing Kikuchi bands ("forbidden reflections") for the T sites.

FIRST HXP EXPERIMENTS

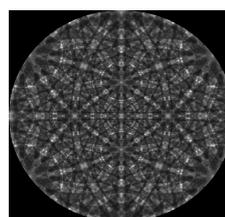
W(110) Valence-Band HXP from W(110) at 6kV, 300K by Ueda, Kobayashi, et al. (SPring8)



Reasonable agreement with theory: positions and spacings of some features

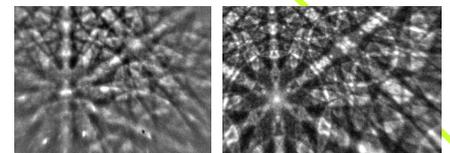
Positions of some features not predicted -> slight crystal misalignment?

Simulation: W(110) at 5946eV stereographic projection



Scanning Electron Microscopy: Electron Backscatter Diffraction (EBSD) from W at 6kV

Measured EBSD pattern „-6kV“ Dynamical simulation 6.3kV



Pattern courtesy of R. Saliwan Neumann, Bundesanstalt für Materialprüfung (BAM), Berlin, Germany

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