Transmission surface diffraction for µm-resolved in-situ studies of heterogeneous electrodes

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Processes at electrochemical interfaces often involve structural changes that are heterogeneous on the micrometre scale. We present a novel in-situ X-ray diffraction technique that uses high-energy photons and a transmission geometry for acquiring atomic-scale surface information with micrometre spatial resolution. The potential of this new technique is illustrated by in-situ studies of surface phase transitions and electrodeposition processes on model systems and real-world materials such as transition metal dichalcogenides (TMDs).

For large, high-quality single crystals, grazing incidence diffraction (GID) is a powerful tool for studying interfaces on the atomic scale. Using high photon energies and 2D detectors, a large portion of reciprocal space can be accessed in one image. [1] However, the large beam footprint prevents spatially resolved measurements.

We recently proposed an alternative geometry termed Transmission Surface Diffraction, in which the X-ray beam impinges along the surface normal and passes through the sample, allowing for a beam footprint of just µm dimensions. [2] Combined with high energies (70 keV), this geometry gives access to the full qz=0 plane of reciprocal space in a single acquisition, with diffraction peaks at intersections between the Ewald sphere and crystal truncation rods or adlayer rods (Fig. 1). This allows easy operando monitoring of unknown, incommensurate 2D phases, as no searches in reciprocal space are required. Proof-of-principle experiments on Au(111) (surface reconstruction, underpotential deposition of Bi) show that TSD is surface-sensitive down to partial monolayers.

Compared to GID, the main benefit offered by TSD is the small, µm-sized beam footprint on the sample, as a wide range of real-world interfaces exhibits heterogeneities on this scale. We deposited 30 monolayers (ML) of Au(111) on top of Si, modified the sample with mechanical scratches and then deposited an additional 20-300 ML of Pd. 2D-mapping of the electrode revealed significant changes in in-plane strain, domain rotation and disorder at the scratch edges, confirming that TSD can indeed resolve µm-size structure variations in nm-thin films.

To expand this technique to real-world materials, we used TSD to investigate Cu intercalation into 1T-TaS2, which was found to result in pronounced structural changes. These materials are typically prepared as µm-sized nanosheets, where TSD can offer unique in-situ insights into the surface structure of single 2D sheets.

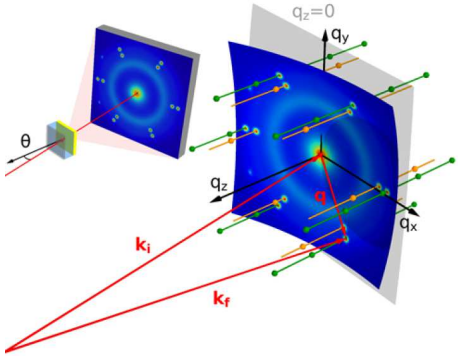

Figure 1. Real (left) and reciprocal space (right) geometry of TSD.

References:
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