# Atomic scale model of the platinum (111)-water interface revealed by angstrom resolution off-axis phase shifting holography

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Although the metal-water interface is the preeminent part of interest for a basic understanding of electrocatalysis and has become an extensively discussed topic in current catalysis research, the atomic structure of the electrolyte, the active sites of the reactions, and the role of single atoms and step edges on the catalyst surfaces remain elusive.

Environmental TEM research enables atomic-scale assessments of metal-water interfaces. We present an open-cell in-situ study of the interface between catalytically active platinum (111) and water vapor under experimentally feasible pressures. Under a water ambient condition, we measure the projected potential across the interface using the angstrom-resolution electron holography method [1]. The results obtained at various external bias conditions are compared to the atomic structures from ab initio molecular dynamics (AIMD) simulations [2]. We will discuss the metal-water interface structures and their natures from experiment and theory.

A two-electrode MEMS setup is exposed to 50 microbar water vapor, which forms an ultrathin condensed water layer at the Pt electrode surfaces. An in situ analysis is carried out by employing an image-aberration-corrected Titan environmental TEM. Off-axis phase-shifting electron holography is used in conjunction with an improved drift-correction scheme to reconstruct the exit-wave with a spatial resolution up to the information limit of the microscope (<1.0 Å).

## Results

The in-situ HRTEM image series and phase reconstructions identify dynamic platinum adatoms at the metal-water interface. The adatoms are residues of the metal-water interface formation and appear only after the electrode is exposed to the water. The existence of the adatoms may depend on the preparation procedures of the metal electrode. By comparing the quantitative frozen-lattice multi-slice simulations [3], we retrieve the specimen thickness at the edge and, subsequently, the coverage of the dynamic platinum adatoms. At the metal-water interface, the oscillations of the projected potential up to 5 Å above the platinum adatoms are monitored. We interpret the oscillations as preferential orientations of the water molecules in the first water layer with the help of predicted holography data from multi-slice simulations using the AIMD trajectories of the model interface configurations. Furthermore, we provide an outlook on the influence of external bias on the potential profiles and expectations based on AIMD-simulation for different surface conditions.

[1] Lindner, J., Ross, U., Meyer, T., Boureau, V., Seibt, M., & Jooss, C. (2024). Reconstruction of Angstrom resolution exit-waves by the application of drift-corrected phase-shifting off-axis electron holography. *Ultramicroscopy*, *256*, 113880.

[2] Huang, J., Zhang, Y., Li, M., Groß, A., & Sakong, S. (2023). Comparing Ab Initio Molecular Dynamics and a Semiclassical Grand Canonical Scheme for the Electric Double Layer of the Pt (111)/Water Interface. *The Journal of Physical Chemistry Letters*, *14*(9), 2354-2363.

[3] Barthel, J. (2018). Dr. Probe: A software for high-resolution STEM image simulation. *Ultramicroscopy*, *193*, 1-11.