Reconstruction of Angstrom resolution exit-waves by the application of drift-corrected phase-shifting off-axis electron holography

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Background

Off-axis electron holography is a phase retrieval technique which enables access to the full complex-valued exit-wave of thin samples. The potential distribution at interfaces obtained from the measured phase information is highly relevant for in-situ experiments. Combining off-axis holography with the capabilities of an environmental TEM offers the ability for exit-wave reconstruction under external bias and in catalysis relevant gases and material systems. However, the conventional holography Fourier-reconstruction suffers from a trade-off between spatial and phase resolution caused by the fringe spacing and visibility. To tackle the open fundamentally questions in catalysis research, e.g. the atomic structure of the electrolyte-solid interface, the identification of active reaction sites and the influence of surface faceting, atomic resolution is highly desired. Therefore, state-of-the-art phase retrieval techniques must be adapted to the particular requirements of in-situ studies. Phase-shifting electron holography bypasses the spatial resolution limit by real-space evaluation of hologram series. However, to reach atomic resolution in reconstructed hologram-series, special care is needed to correct sample and biprism drift.

Methods

Phase-shifting holography acquires a series of holograms formed by tilted incident waves. This results in a shift of the hologram fringes, that are modulated by the potential of the specimen. If specimen and biprism drift are carefully corrected, the cosine intensity dependency of the hologram series can be used for linear fits of the local amplitude and phase of the exit wave. This obviates the use of the low-pass aperture which is necessary for the conventional reconstruction of off-axis holograms in the Fourier domain. The upper bound of the spatial resolution is thus only limited by the performance characteristics of the instrument, while the low-frequency information is also retained.

Results

Previous implementations of phase-shifting holography have been limited by the independent drift of biprism and sample and allowing for medium spatial resolution. We improve the reconstruction process by introducing a drift correction scheme and demonstrate exit wave reconstruction on platinum. The reconstructed exit-waves show reliable phase information at the 1 Å information limit of the used Titan 80-300 kV environmental transmission electron microscope. Simultaneously, the omission of the trade-off between fringe spacing and visibility leads to phase resolutions up to 2π/452 rad at moderate birpism voltages of 250 V (fringe spacing 1 Å). The obtained phase and amplitude information is validated at a thin Pt sample due to the excellent matching to frozen-lattice multi-slice image simulations.

Conclusions

In conclusion, we demonstrate the successful method improvement of the phase-shifting holography reconstruction process by introducing novel drift correction of the mixed signals of biprism and sample drifts. The reconstructed exit-waves of a thin platinum sample show spatial resolution up to the 1 Å information limit of the microscope simultaneously with a phase resolution up to 2π/452 rad. The exit-waves are in excellent agreement with multi-slice frozen lattice image simulations and preserve the high- and low-frequency information. The published method is applicable in any TEM equipped with a single electron biprism and thus allows to achieve high resolution off-axis holography in various instruments including those for in-situ applications. A software implementation for the acquisition, calibration and reconstruction is provided. The combination of environmental TEM and high-resolution phase-shifting electron holography grants access to the platinum-water interface at the atomic scale in ongoing studies.

Keywords:

atomic-scale, off-axis-holography, exit-wave reconstruction, ETEM

Reference:

Q. Ru, G. Lai, K. Aoyama, J. Endo, and A. Tonomura, “Principle and application of phase-shifting electron holography,” Ultramicroscopy 55 (2), 209–220 (1994).

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