Quantitative Chemically Specific Coherent Diffractive Imaging of Reactions at Buried Interfaces with Few-Nanometer Precision

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Keywords: Nanometrology, imaging, buried layers, chemical-specificity, high harmonic generation, extreme ultraviolet

Supporting Information

Sample Fabrication

The Cu structures were deposited by physical vapor deposition and patterned using I-line resist with a Canon GS22 followed by electroplating to reach a total height of 750 nm. Subsequently, 500 nm of SiO_2 was deposited using tetraethyl orthosilicate (TEOS).

Uncoated Sample Reflectivity Model Calculation

It is possible that a small amount of interdiffusion may occur between the Cu and its oxide layers, but we found that modeling diffusion by convolving the depth profile with a Gaussian having up to a 5 nm standard deviation did not significantly alter the theoretical reflectivity or phase.

Table S1. Reflectivity values of the uncoated sample

	Features	Substrate
Experiment	10.97 ± 1.26	6.77 ± 0.61
Model	11.10	6.45

Experimentally reconstructed mean reflectivity values (%) compared to modeled reflectivities of features and substrate on the uncoated sample. The error shown is the standard deviation from the mean of the reconstructed reflectivity in the masked regions

Coated Sample Reflectivity Model Calculation

Table S2. Coated Sample Reflectivity Values

	Features	Substrate
Experiment	2.04 ± 0.56	2.66 ± 0.35
Model	1.99	2.57

Experimentally reconstructed mean reflectivity (%) values compared to modeled reflectivities of features and substrate on the coated sample.

Height Maps

In the case of the uncoated sample, the model predicts the Fresnel phase shift due to the features is -96°, while the phase change from the substrate is -65°. In the case of the coated sample, the phase change from the features is -92°, while the phase change from the substrate is -43°.

For plotting purposes, the AFM images have been smoothed with a Gaussian filter with full width equal to the EUV microscope's pixel size in the case of the coated sample and twice the EUV pixel size in the case of the uncoated sample (to remove a one pixel artifact in the image around the letters). Without smoothing, the AFM images appear noisy because the feature heights are similar to the surface roughness of the sample.

Images were flattened using a 5th order polynomial surface fit to decrease substrate height variations and make the feature heights more easily comparable. Binary masks used to assign Fresnel phases in the CDI reconstructions were generated using the intelligent scissors algorithm[1].

Data Acquisition

For both samples, the ptychographic data set consisted of 270 diffraction patterns, collected with 3 μm step sizes between scan positions. A random offset of ±20% of the step size was added to each scan position to prevent periodic artifacts in the reconstructions.[36] The total EUV exposure time for the uncoated sample was 5.8 min, compared with 23.6 min for the Al-coated sample, with a total scanned area of 4270 μm^2 each. In future experiments, these exposure times can be reduced significantly (>100x) using optimized geometries and driving lasers.

Ptychography Reconstructions

Each of the reconstructions were run first using RAPTR-CDI for an initial 50 iterations, with position correction[2] implemented for another 3000 iterations afterwards. The multicolor reconstructions were both run using the wavelengths of the 25^{th}, 27^{th}, and 29^{th} harmonics and for approximately 2000 iterations.

The transverse spatial resolution of the EUV images is limited by the effective numerical aperture (NA) of the imaging system. Because there are no optics between the sample and detector, the NA is determined by the distance from the sample to the detector ($z = 38.5$ mm) and the size of the detector (D). The diffraction patterns were cropped to 512 x 512 to reduce computation time, so the effective detector size, D_{eff} is 512 x p, with pixel size p = 13.5 μm square. Therefore, the NA is $D_{eff}/2z = 0.09$ and the diffraction limited resolution is $\lambda/(2NA) =$ 162 nm.

The difference between the positions found using the position correction algorithm and the initial recorded positions were used to correctly determine the pixel size of the reconstructed images. This is necessary in general for ptychography CDI because any error in the measurement of the sample to detector distance or angle results in changes in the reconstructed pixel size. To solve for this change in the x and y directions, the difference between the corrected positions and the initial positions was fit to a plane. In particular, the scale factor necessary to correct the pixel size in the x-direction, a_x is $1-s_x$, where s_x is the slope of the plane fit to the position differences. Then, $d_x \cdot a_x$ is the corrected pixel size with d_x being the predicted pixel size in the absence of tilted plane correction and any error in the measured detector-sample distance. It was found that after solving for the correct pixel size the scaling of the EUV reconstructions agreed well with the SEM measurements, whereas without this scaling the reconstructions were stretched by up to 18% in a given direction.

To correct for error in the XY-calibration of the AFM used for comparison to the HHG CDI reconstructions, the AFM images in Figures 2 and 4 were scaled in the horizontal and vertical directions so that the transverse dimensions of the number “725” are in agreement with the SEM images shown in Figure 2.

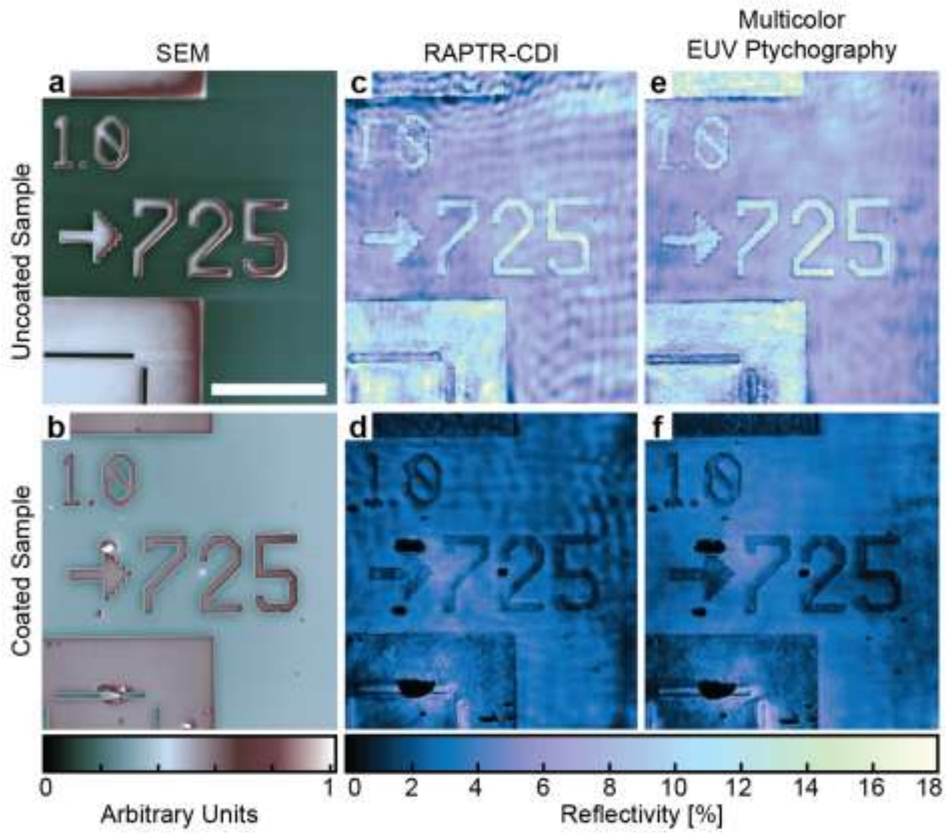

Figure S1. (a,b) SEM images for comparison. (c,d) RAPTR-CDI reconstructions, which return absolute reflectivity, but assume all diffraction is from exactly one harmonic. (e,f) Multicolor ptychography reconstructions demonstrating the enhanced fidelity provided by the multicolor algorithm. Because the multicolor algorithm only returns relative reflectivity, the reconstructions have been scaled such that their average reflectivity agrees with that of the RAPTR-CDI reconstructions.

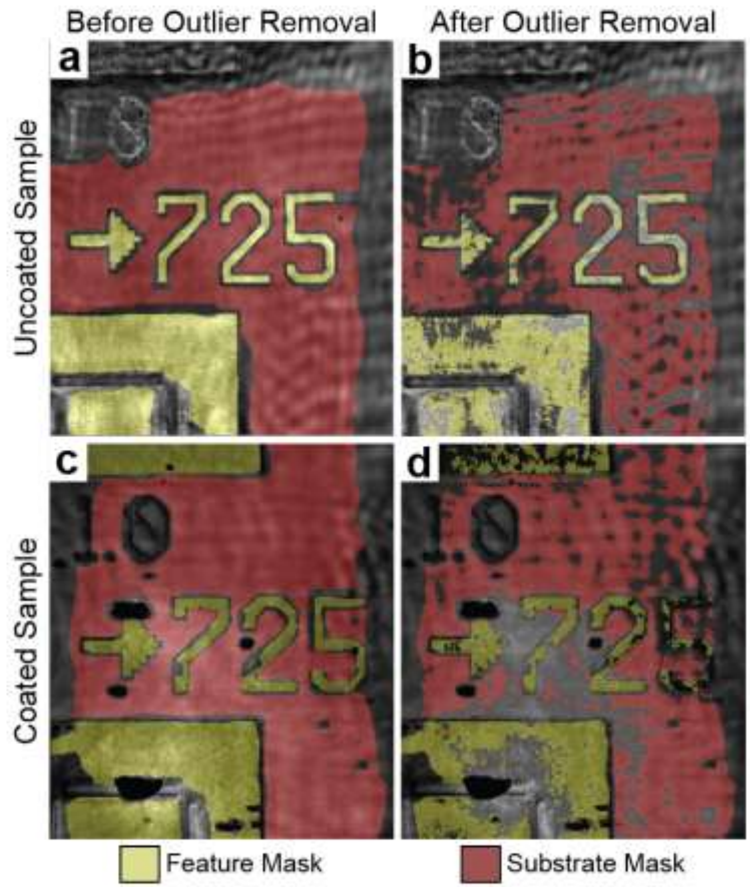

Figure S2. Masks used to calculate the average feature reflectivity (yellow) and substrate reflectivity (red) for the uncoated (b) and coated (d) sample are shown in the right column. Masks (b) and (d) were generated from masks (a) and (c) by excluding points further than one standard deviation from the mean reflectivity of the included areas. Masks (a) and (c) were made using the intelligent scissors algorithm[1]. Masks (b) and (d) were used to scale the multicolor reconstructions so that their average reflectivity matches the RAPTR-CDI reconstructions.

References

(1) Mortensen, E.; Barrett, W. *Comput. Graph. (ACM).* **1995**, 191–198.