Supplementary Information

Scan Parameters

O 1s Ptychography Scan:

- Zone-plate parameters: 50 nm outer zone width
- Ptychography scan paramaters: 1.25 μm diameter spot size, 8x8 pixel raster scan, 0.25 μm pixel size
- Camera: Andor CCD

Non-resonant (571 – 589 eV) scan

- Zone -plate parameters: 40 nm outer zone width
- Ptychography scan paramters: 2.5 μm diameter spot size, 10x10 pixel raster scan in 0.5 μm pixel size
- Camera: Tucsen Dhyana 95V2 BSI sCMOS Camera

Figure S1: Ptychography amplitude (a,b) and phase (c,d) images recorded at 571 eV of a single layer array of 220 nm SiO_2 nanospheres on a 100 nm Si_3N_4 membrane. The scale bar in each image represents 1 μm. A layer of photodeposition has been deposited in the upper left of these images due to previous spectroptychography scans of that area. The average amplitude and phase signal for the bare Si_3N_4 was extracted from the green region in images images (a) and (c), respectively and used to generate the corresponding spectra. The green region in images (b) and (d) was used to extract either the average amplitude or phase signal for the Si_3N_4 membrane with a layer of photodeposition. Amplitude (e) and phase (f) spectra for the bare Si_3N_4 membrane and the Si_3N_4 membrane with a layer of photodeposition.

Figure S2: Ptychography amplitude (a,b) and phase (c,d) images of a single layer array of 220 nm SiO_2 nanospheres on a 100 nm Si_3N_4 membrane recorded at 571 eV. The scale bar in each image represents 1 μm. A layer of photodeposition has been deposited in the upper left section of these images from previous spectroptychography scans. The average amplitude signal for the SiO_2 nanospheres on the Si_3N_4 membrane without (a) and with (b) photodeposition was extracted from the green regions to generate the corresponding amplitude spectra. The amplitude signal for the bare Si_3N_4 membrane was obtained from the red region in images (a) and (b) and used to normalize the amplitude signal from the green regions according to Beer's law to generate optical density spectra for the SiO_2 nanospheres on the Si_3N_4 membrane with and without photodeposition. The average phase signal was extracted from the green region in images (c) and (d) and used to produce phase spectra for the two regions. Amplitude (e), optical density (f), and phase (g) spectra for SiO_2 nanospheres on the Si_3N_4 membrane with and without photodeposition.

Figure S3: Ptychography amplitude (a,b) and phase (c,d) images of 220 nm SiO_2 nanospheres on a 100 nm Si_3N_4 membrane. Images (a) and (c) were recorded at 538.33 eV while images (b) and (d) were recorded at 536.52 eV. The scale bar in each image represents 0.6 μm. The amplitude and phase signal for the nanosphere cores was averaged from green region in images (a) and (c), respectively and the resulting data was used to generate the corresponding amplitude and phase spectra. The green region in images (b) and (d) were used to generate the average the amplitude (b) and phase (d) signal for the nanosphere shells to generate the corresponding nanosphere shell spectra. The amplitude signal for the bare Si_3N_4 membrane was extracted from the red regions in images (a) and (b). This was used to normalize the amplitude signal for the nanosphere cores and shells, respectively according to Beer's law to generate optical density spectra for the two sections of the SiO_2 nanospheres. Amplitude (e), optical density (f), and phase (g) spectra for the SiO_2 nanosphere cores (SiO_2 NS Core) and shells (SiO_2 NS Shells)

Calculation of Phase Spectra

Phase spectra were qualitatively calculated using the `ft.hilbert` routine in the `scipy.fftpack.hilbert` library,[1] using the following process:

1. Atomic transmission and optical density spectra were calculated for selected compounds using Henke atomic cross-sections,[2] within the program aXis2000.[3] The molecular compound, density and thickness are provided as inputs, and the aXis2000 program uses f_2 atomic scattering factors to provide atomic transmission and optical density spectra over a defined energy range. The aXis2000 program has access to Henke atomic cross-section data from 10 eV – 30 keV. In this case, atomic transmission and optical density spectra were generated from 10 eV – 3 keV as previous research[4] has shown that integration over the full 30 keV energy range isn't necessary to eliminate distortions in the phase spectra at soft X-ray energies.
2. A simple python code is used to obtain the qualitative phase spectra. This code consists of three steps:

- A simple routine to read in the atomic optical density spectra provided by step 1, above.
- The code calls the `ft.hilbert` routine from the `scipy.fftpack.hilbert` library, e.g.

```
PhaseSpectrum = - ft.hilbert(AbsorptionSpectrum)
```

- The qualitative phase spectra are written to a text file.

The phase spectra obtained in this manner are qualitative, in that the `ft.hilbert` transform lacks the constants and the 'Z' term of the full Kramers-Kronig relation.

References:

1. [1] scipy.fftpack.hilbert, docs.scipy.org, 2023.
2. [2] B.L. Henke, E.M. Gullikson, J.C. Davis, X-Ray Interactions: Photoabsorption, Scattering, Transmission, and Reflection at E = 50-30,000 eV, Z = 1-92, Atomic Data and Nuclear Data Tables 54 (1993) 181-342.
3. [3] A.P. Hitchcock, aXis2000, Hamilton, ON, 2023.
4. [4] C. Jacobsen, S. Wang, W. Yun, S. Frigo, Calculation of x-ray refraction from near-edge absorption data only, Optical Science and Technology, the SPIE 49th Annual Meeting, SPIE2004.

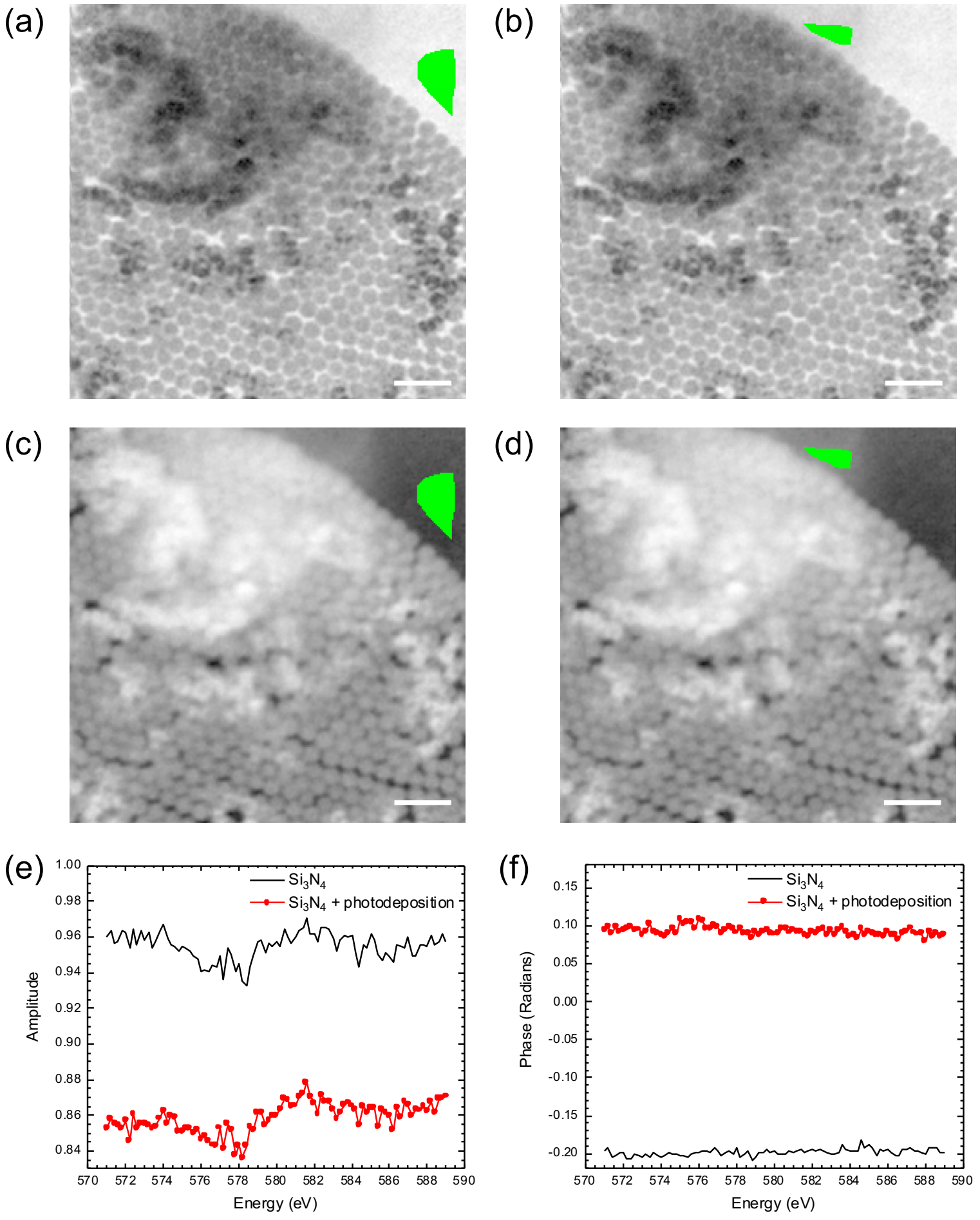
(a)
(b)
(c)
(d)
(e)
Amplitude
Energy (eV)
Si_3N_4
Si_3N_4 + photodeposition
(f)
Phase (Radians)
Energy (eV)
Si_3N_4
Si_3N_4 + photodeposition

Figure S1

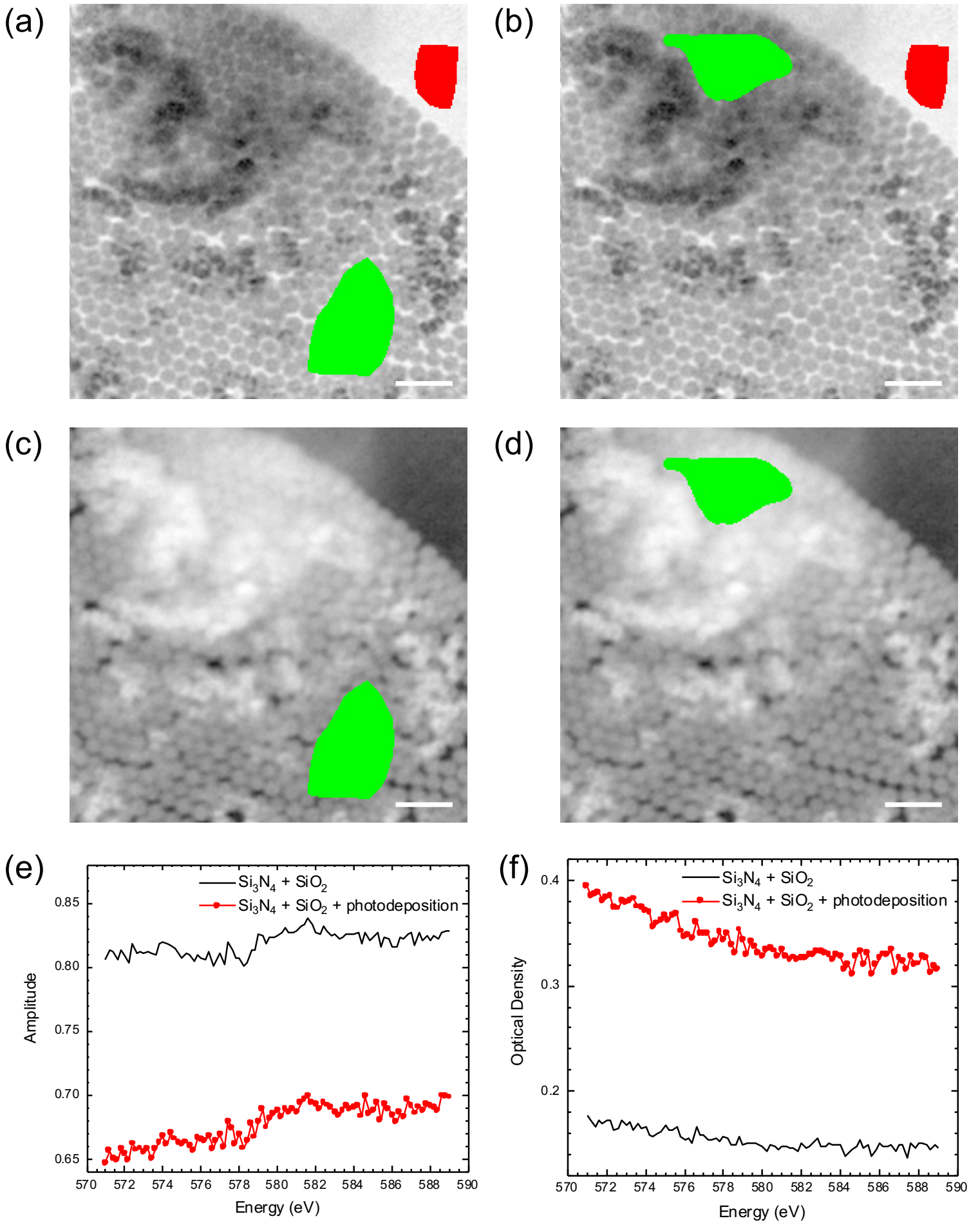
(a)
(b)
(c)
(d)
(e)
Si3N4 + SiO2
Si3N4 + SiO2 + photodeposition
Amplitude
0.85
0.80
0.75
0.70
0.65
570 572 574 576 578 580 582 584 586 588 590
Energy (eV)
(f)
Si3N4 + SiO2
Si3N4 + SiO2 + photodeposition
Optical Density
0.4
0.3
0.2
570 572 574 576 578 580 582 584 586 588 590
Energy (eV)

Figure S2 (a-f)

(g)

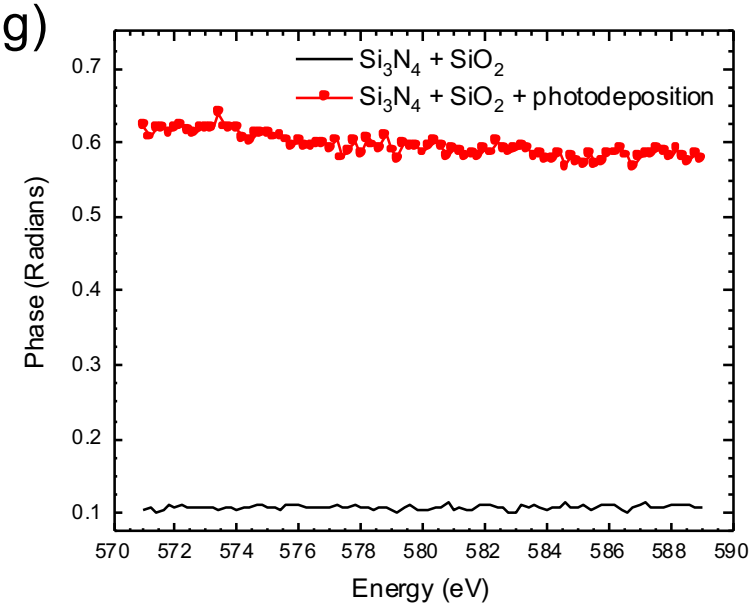
Si3N4 + SiO2
Si3N4 + SiO2 + photodeposition
Phase (Radians)
0.7
0.6
0.5
0.4
0.3
0.2
0.1
570
572
574
576
578
580
582
584
586
588
590
Energy (eV)

Figure S2 (g)

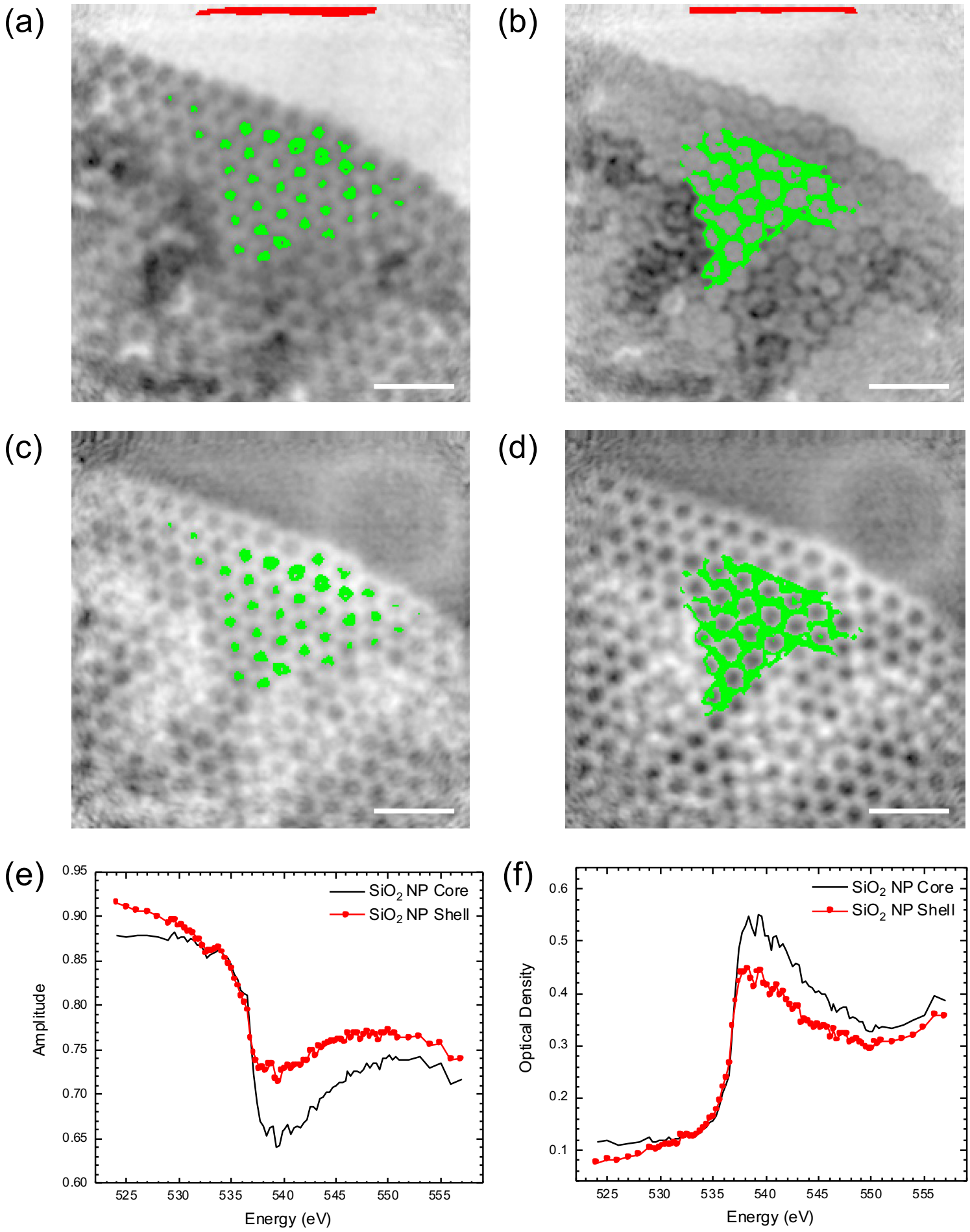
(a)
(b)
(c)
(d)
(e)
(f)
SiO2 NP Core
SiO2 NP Shell
Amplitude
Energy (eV)
Optical Density
Energy (eV)

Figure S3 (a-f)

(g)

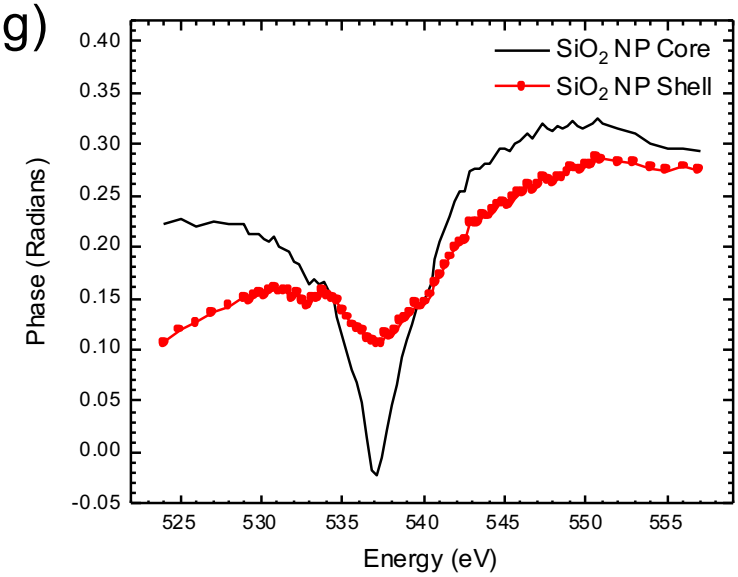
SiO2 NP Core
SiO2 NP Shell
Phase (Radians)
Energy (eV)
0.40
0.35
0.30
0.25
0.20
0.15
0.10
0.05
0.00
-0.05
525
530
535
540
545
550
555

Figure S3 (g)