Supplementary Information

A Contraction X-direction Y-direction Y-direction Y-direction Y-direction Y-direction

Scan-distortion Diagnosis

Figure S1. Scan-distortion analysis of an example ADF from the image-series. The x and y direction components of the scandistortion vector-field are shown, as well as histograms of their respective pixel values. The x-direction distortions are largely within \pm 10 pm. The y-direction is similar except for a vertical compression visible at the top of the raw image in Figure 7.

In Fig S1 the probe-offsets are represented as the shifts in x and y needed to correct the probe-positions. A similar vector-field exists for each of the 21 scan-frames in the dataset and these are stored with each image as meta-data. From these plots, we can determine that the approximate magnitude of the random probe-offsets is around ± 10 pm in x and ± 20 pm in y. With a pixel-size of 0.264 Å in the raw data, these random shifts are on the same order-of-magnitude.

Spatial-frequency Power Spectrum



Figure S2. Power-spectrum across the spatial dimensions of the 4D data-volumes; (left) from a single scan-frame, (right) from the cumulative 21 frames.



Figure S3. Amplitude plots of the first and third order disk-overlaps corresponding to Figure 7. After dosefusion, the chromatic damping envelope(Pennycook et al., 2015; Nellist & Rodenburg, 1994) is visible even for the third order overlaps.



Figure S4. Phase plots of the disk overlap regions for one example of the first-order Fourier components of the graphene lattice (top) and for a third-order component (bottom). For each one, both the single scan-frame (left) and the cumulative equivalent (right) are shown. For fair comparison, aberration correction and phase unwrapping has been applied to each. Amplitude plots for these overlaps are shown in the supplementary information.



Figure S5. Fourier transforms of the real-space phase-maps from (left) and single 4D-scan, and (right) from the fusion of 21 scans.