Supplementary Information

Localization and Segmentation of Atomic Columns in Supported Nanoparticles for Fast Scanning Transmission Electron Microscopy

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Mean square displacement error

The error of the mean square displacement was calculated by propagating the mean positional error of the localization network. Each displacement d_i between points p_new and p_current is calculated as

$$d_\mathrm{i} = p_\mathrm{new} - p_\mathrm{current}$$

with corresponding displacement error

$$\Delta d_\mathrm{i} = \sqrt{(\Delta p_\mathrm{new})^2 + (\Delta p_\mathrm{current})^2} = \sqrt{2}\epsilon$$

where ϵ is the mean positional error of points predicted by the localization network. The expression for the error of the squared displacement, d_i^2, is then obtained through error propagation as

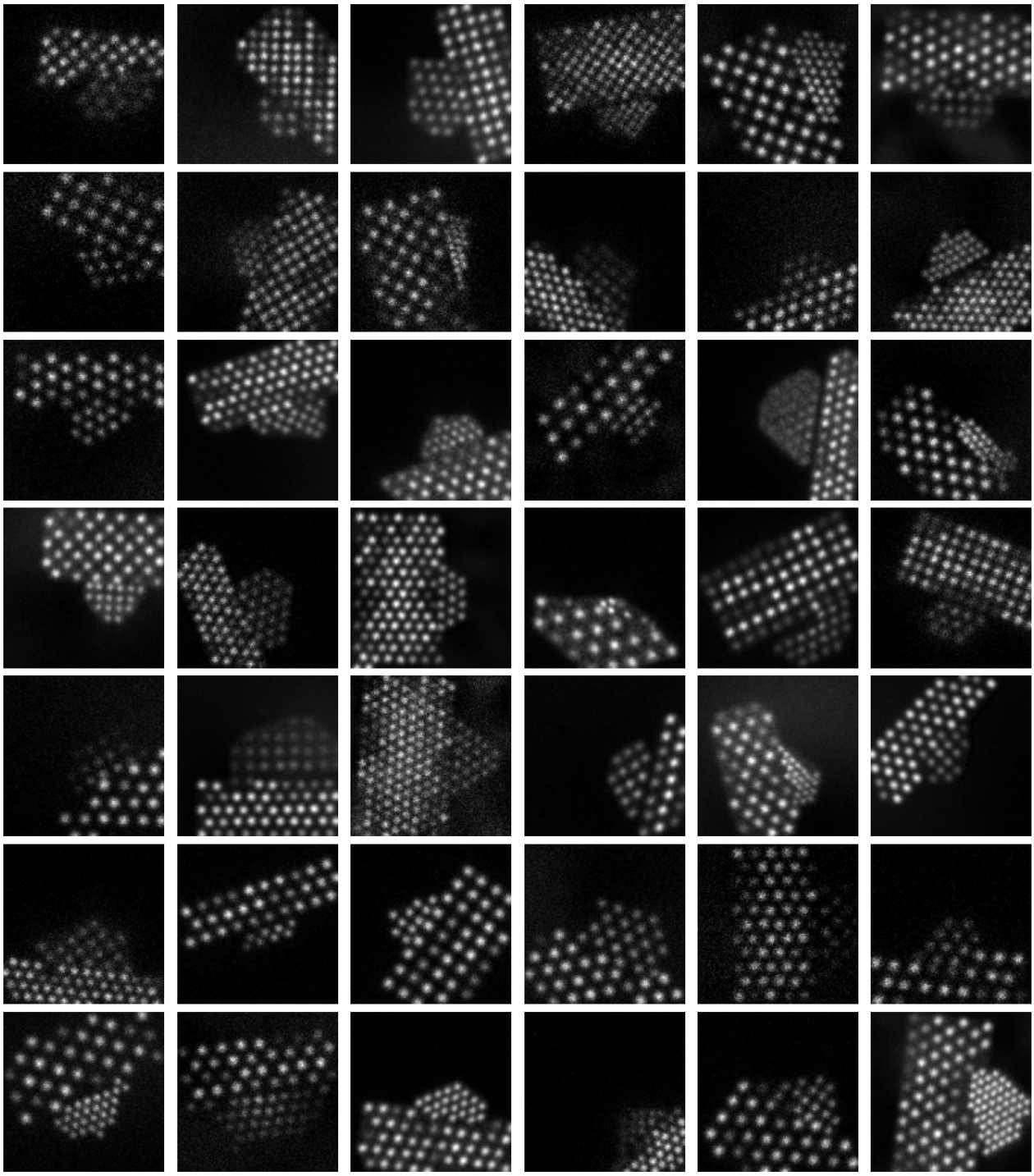
$$\Delta d_\mathrm{i}^2 = 2\sqrt{2}\epsilon|d_\mathrm{i}|.$$

The mean squared displacement is expressed as

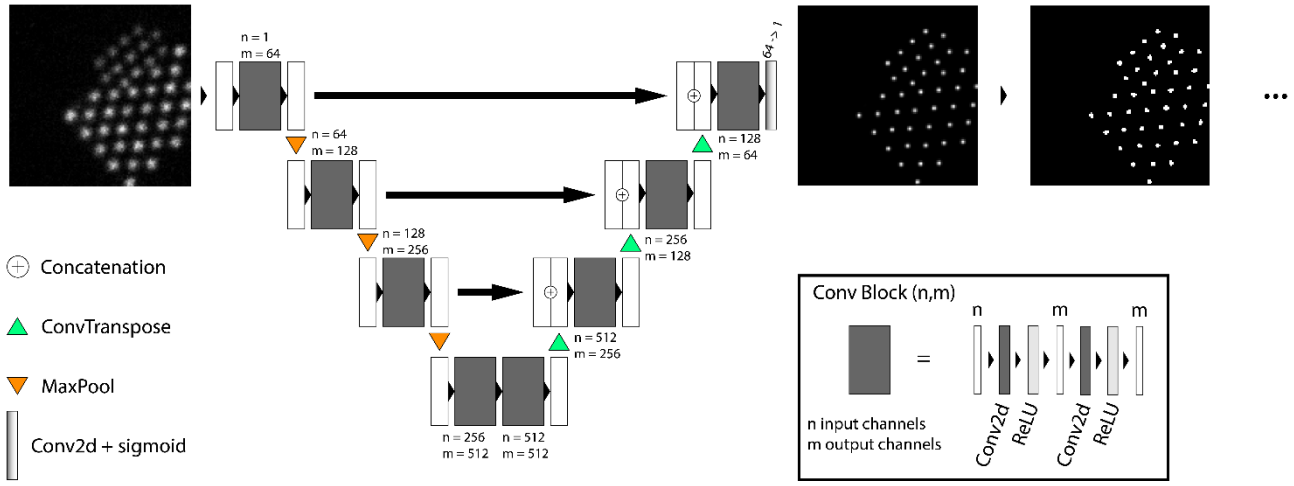
$$\mathrm{MSD} = \frac{1}{N}\sqrt{\sum_{i=1}^{N} d_i^2}$$

where N is the number of points in the sequence, with corresponding MSD error derived as

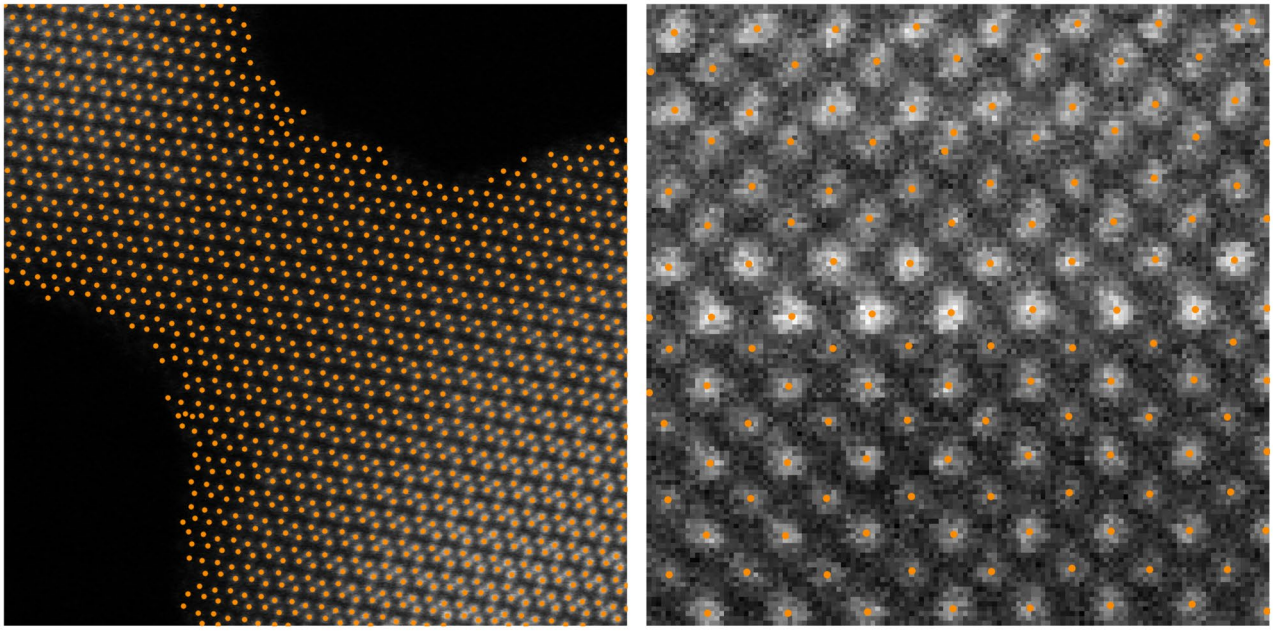
$$\Delta\mathrm{MSD} = \frac{1}{N}\sqrt{\sum_{i=1}^{N}(\Delta d_i^2)^2} = \frac{1}{N}\sqrt{\sum_{i=1}^{N}\left(2\sqrt{2}\epsilon|d_i|\right)^2}.$$



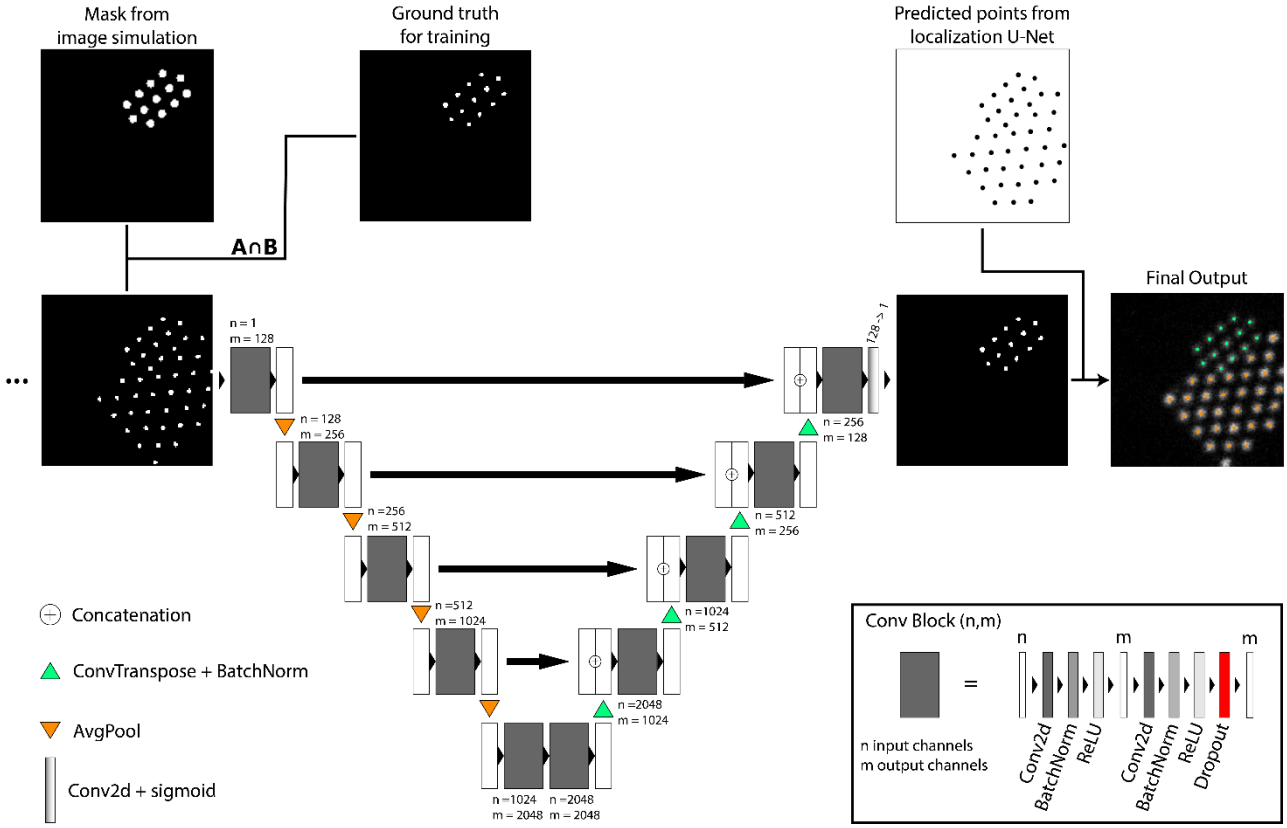
Supplementary Figure 1 An extract of 42 randomly selected images from training dataset.



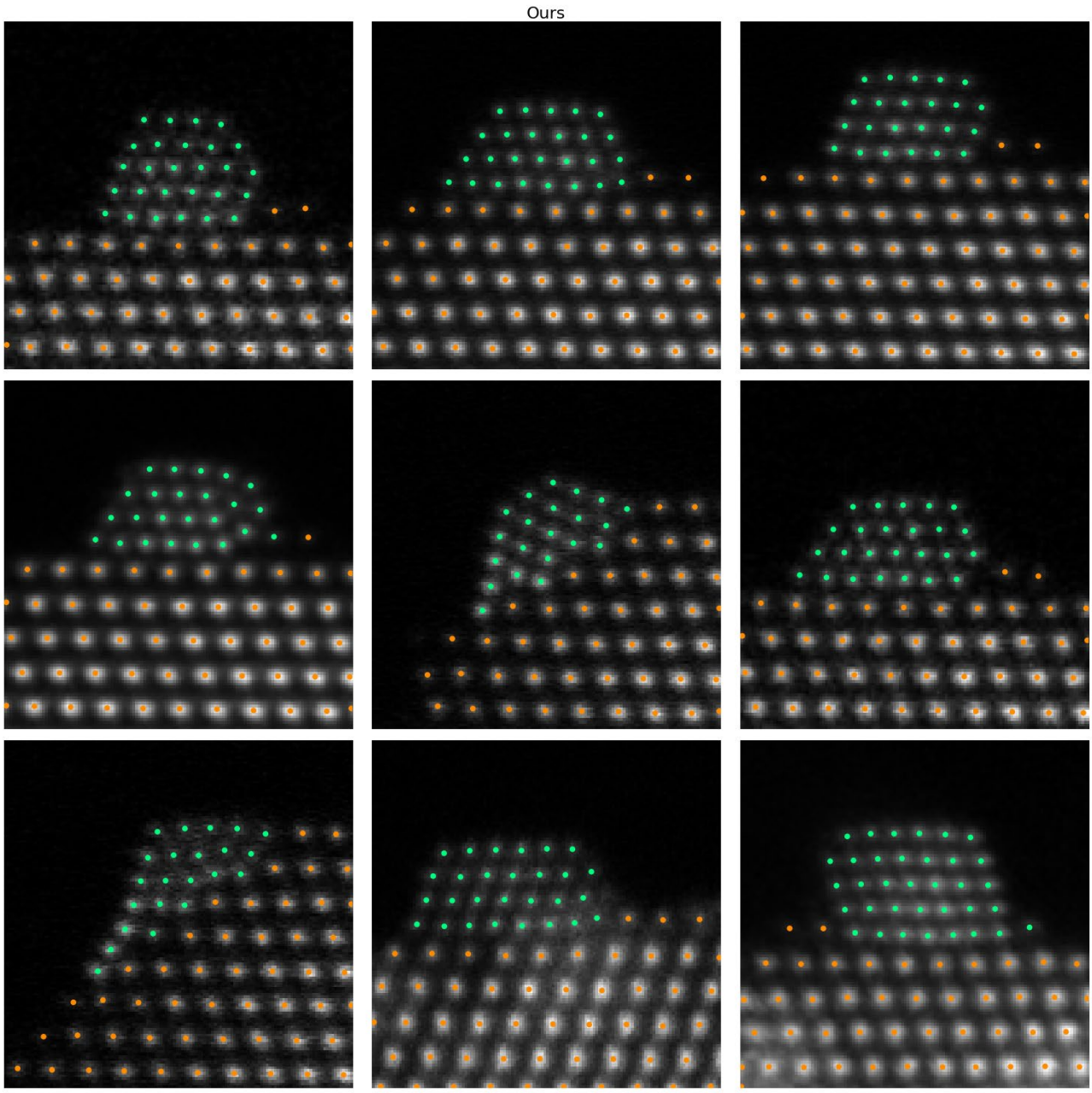
Supplementary Figure 2 Schematic of the localization network and related workflow.



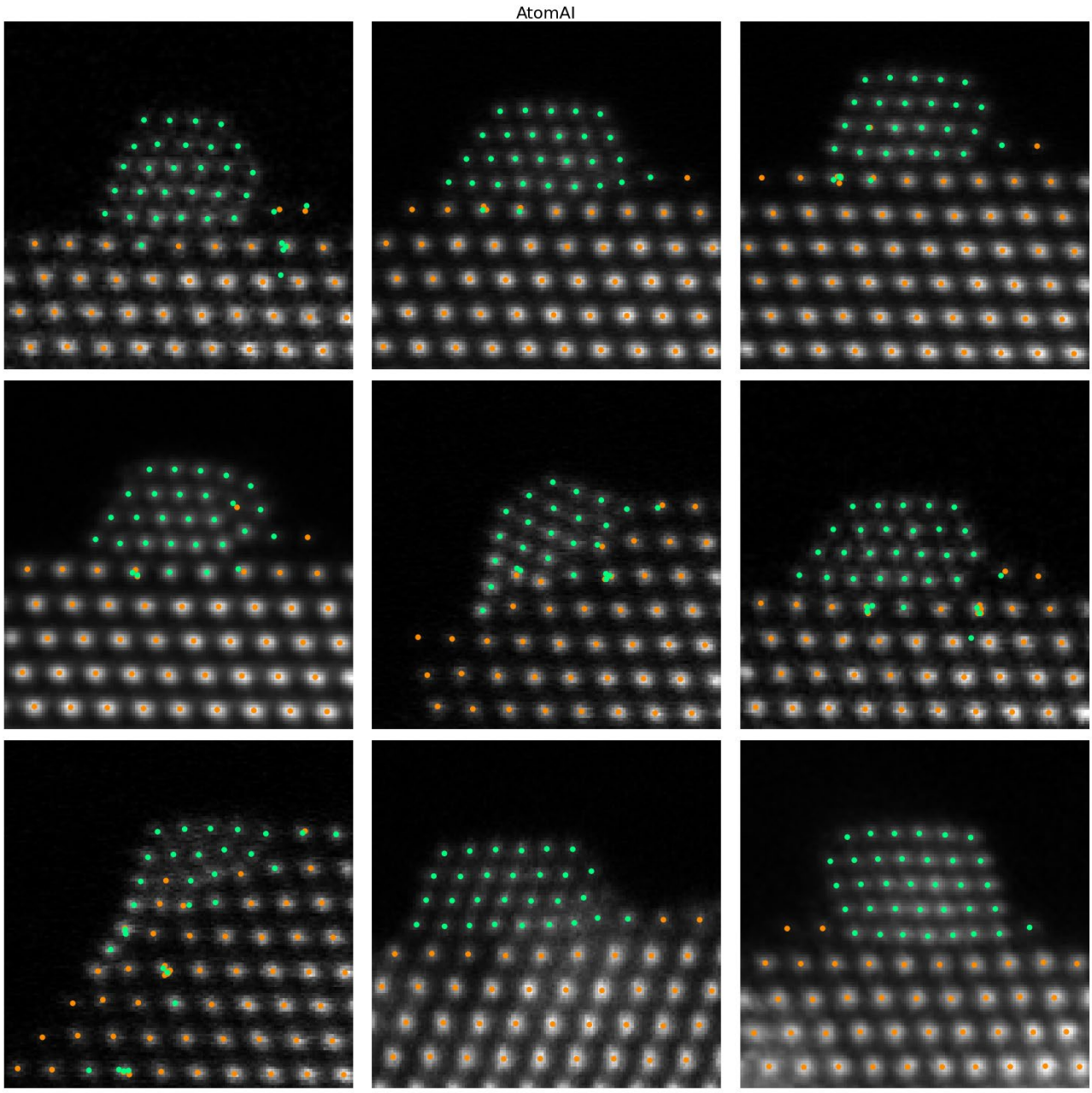
Supplementary Figure 3 The model applied to (left) a 1024x1024 image of monoclinic ZrO_2, a much larger image size than that of the training data, as well as a lattice geometry that the model was not trained on. (right). An HR-STEM image from the TEAM microscope of a perovskite interface.



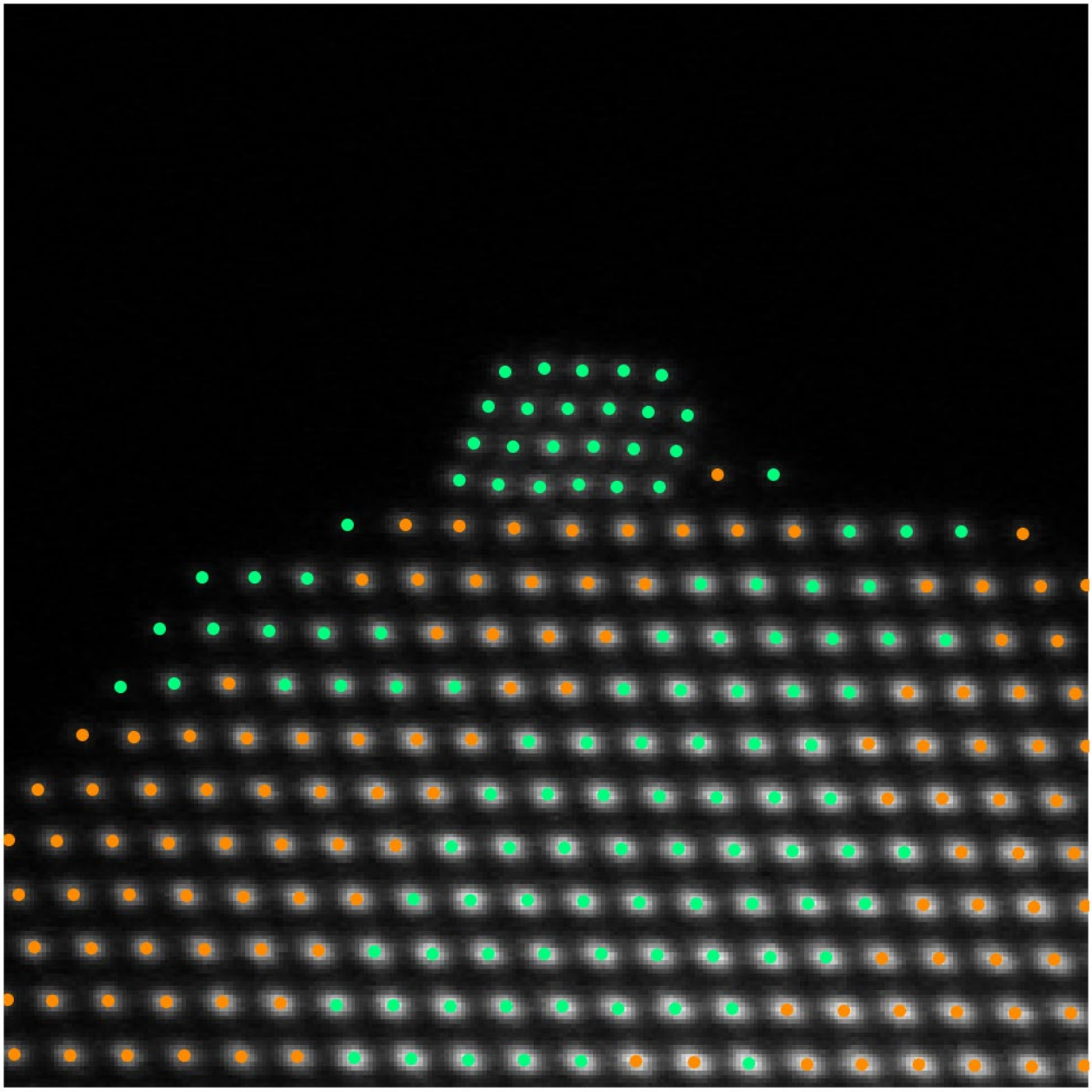
Supplementary Figure 4. Schematic of the segmentation network and related workflow.



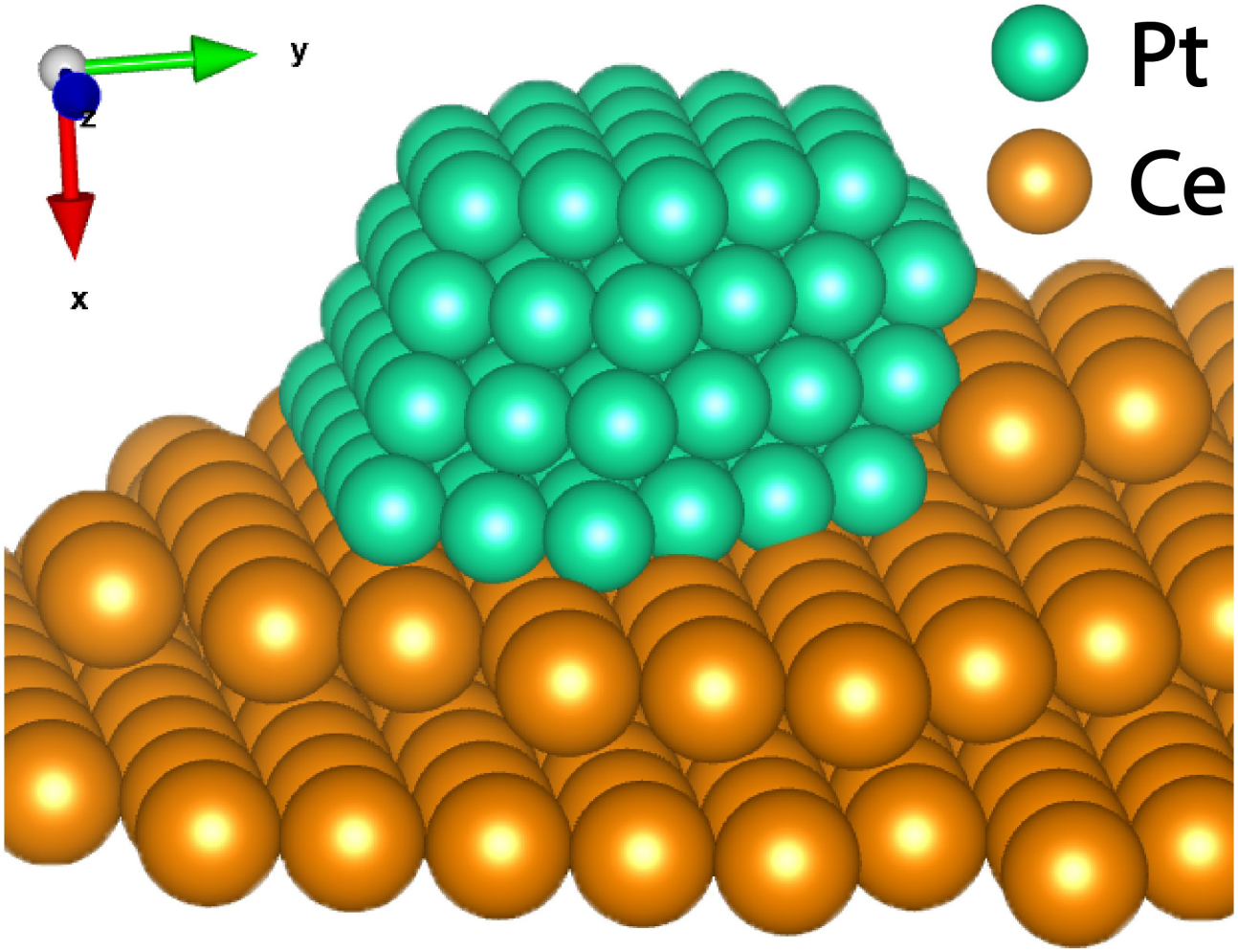
Supplementary Figure 5 The combined localization and segmentation model applied to a test set of experimental images.



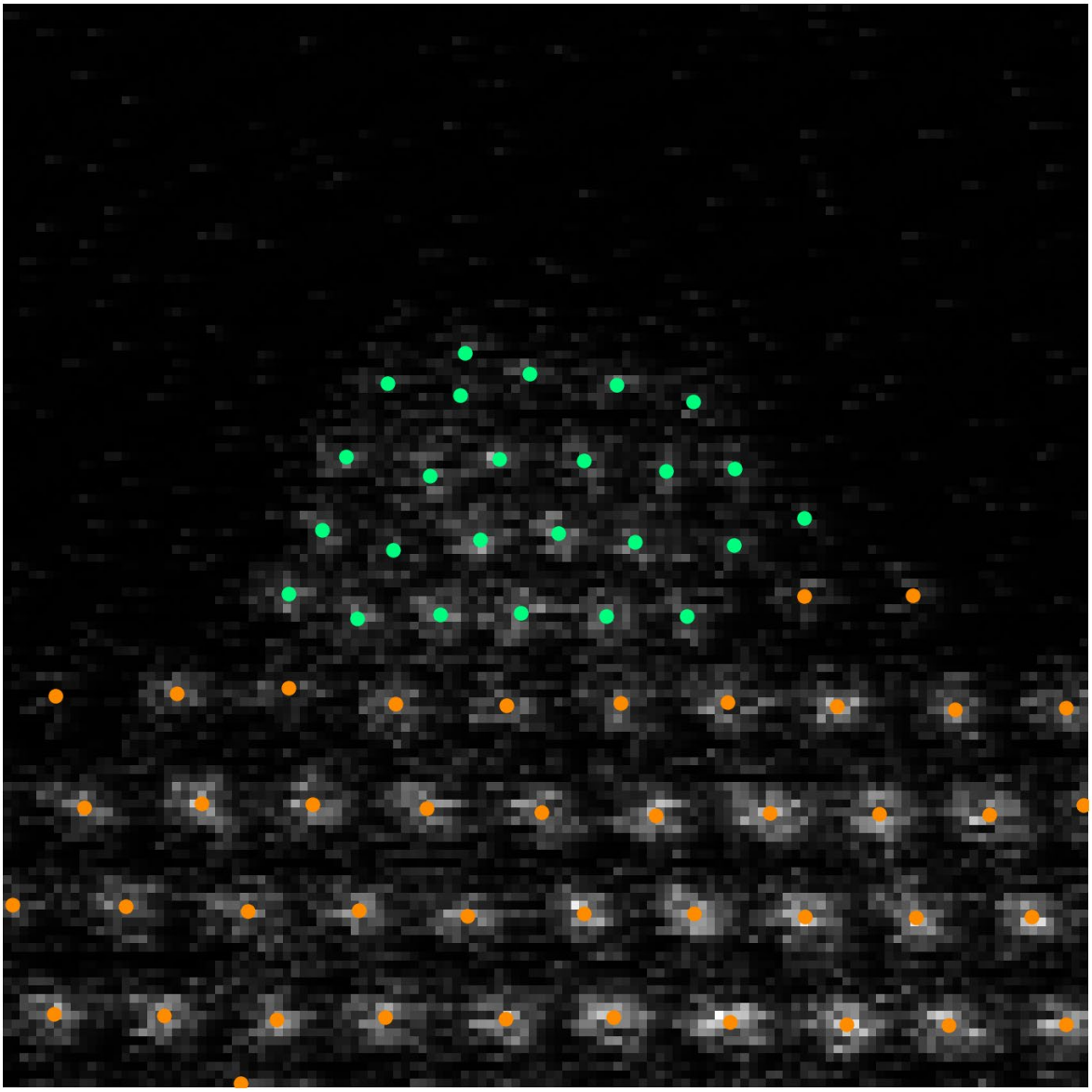
Supplementary Figure 6 AtomAI's *Segmentor* model applied to a test set of experimental images.



Supplementary Figure 7 Our model applied to a 256x256 particle-support interface image. Although the localization model finds all columns, the segmentation model does not generalize well to larger input sizes than 128x128.



Supplementary Figure 8 The Predicted particle structure of the particle in Fig. 7a**.** The estimated structure is inspired by a truncated octahedron and is built up of 118 Pt atoms.



Supplementary Figure 9 Column splitting in one of the sites exhibiting a high mean square displacement. The column to the right of the top left corner has split into two, this behavior can be observed more than once in the time-series. This site in Fig. 7 of the main text exhibits a high MSD which is likely correlated to the splitting.

Supplementary Video 1 The raw time-series of the structure in Figure. 7a of the main text. The time-series was recorded at 5 frames per second and is played back at 20 frames per second.

Supplementary Video 2 The raw time-series of the structure in Figure. 7b of the main text. The time-series was recorded at 5 frames per second and is played back at 20 frames per second.

Supplementary Video 3 The in-situ time-series of the structure in Figure. 7c of the main text. Each frame is the sum of itself and the next 19 frames. The time-series was recorded at 5 frames per second and is played back at 20 frames per second.