

## Application of Deep Unsupervised Convolutional Neural Networks to Denoise Large Temporally Resolved *In Situ* TEM Datasets

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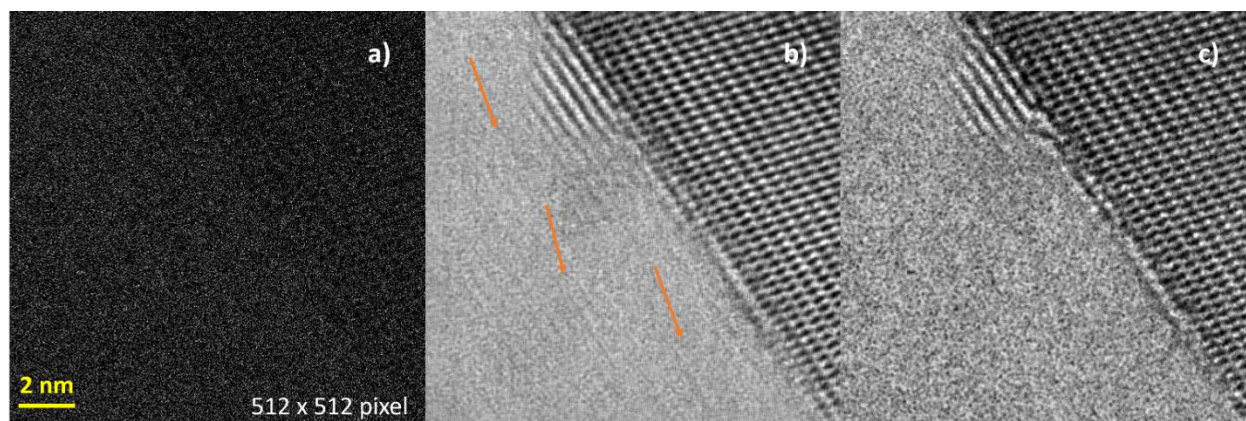
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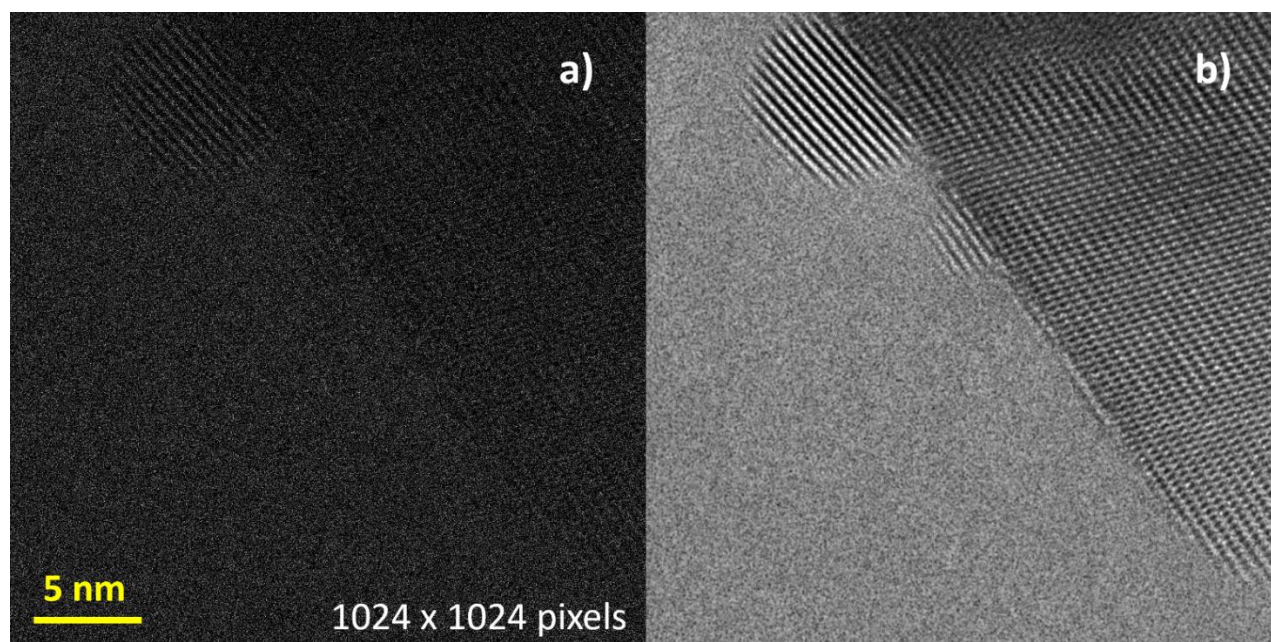
There is increasing recognition of the importance of structural dynamics in nanoparticles linked to catalytic processes [1, 2]. Our ability to gain insights into the functioning of these catalysts during reaction, depends on our ability to image the structural dynamics taking place at the surface/interface. Commercially available direct-electron detectors allow temporal resolutions up to the millisecond regime. However, the short exposure time images are degraded by the presence of Poisson noise associated with the low number of electrons contributing to each frame, hindering the visualization of structural dynamics. To overcome this drawback, convolutional neural networks (CNN) are being employed for denoising such noisy TEM images.

Supervised CNN have already demonstrated outstanding results [3], but they require TEM image simulation data and the generalization to other datasets is not always successful. However, unsupervised neural networks are directly trained on the experimental data and can adapt to new datasets to improve their performance, although there is still much work to be done in the implementation of these networks in the TEM field [4]. In this work, we have developed an unsupervised CNN for application to the experimental time-resolved *in situ* TEM datasets related to catalytic systems. Particularly, the unsupervised CNN is trained directly in a set of noisy videos trying to predict the value of a given noisy pixel using only the values of its spatial and temporal neighboring pixels, that is using the pixels around it in the same video frame and those in the same region of the previous and following video frames. Assuming that the expected value of a given noisy pixel is its clean value, the model tends to learn to denoise the images when trained in videos with a length of 30 frames or more. The time-series images used in this work are Pt/CeO<sub>2</sub> catalysts captured with a Gatan K3 camera at 75 frames per second. Different parameters like frame size and frame numbers are tuned to optimize the CNN performance. We are currently working on training new neural networks on a large noisy TEM dataset, which will learn and evolve in its denoising capabilities for future training datasets.

To evaluate the influence of dataset parameters on the performance of the developed CNN, the unsupervised denoising code has been applied to time-series with different frame sizes and frames numbers. Figure 1 (a) shows a noisy experimental TEM image and Figure 1 (b), (c) show the denoised output on two different 512 x 512 images datasets with lengths 400 and 1124 frames respectively. As indicated by the arrow marks in Figure 1(b), the 400-frame set shows a propagation of faint fringes into the vacuum. On the other hand, these artifacts disappear when the number of frames is increased (Figure 1(b)). In addition to the number of frames, the size of the image has also been assessed. Figure 2 (a) also shows the noisy experimental image with larger frame size and Figure 2 (b) represents a 1024 x 1024 image dataset with 1124 frames. As observed, the denoised output when increasing the number of pixels the performance gets better. These findings confirm that increasing the number of pixels gives better the denoising performance. Keeping this in mind, we will address the training of a more efficient model which will learn from our historical database and thus improve its performance on new unseen data [5].



**Figure 1.** (a) Shows the noisy experimental TEM image (b), (c) Represents the denoised output from unsupervised neural network on experiment TEM time series images of size 512 x 512 pixels with 400 and 1124 frames respectively.



**Figure 2.** (a) Shows the noisy experimental TEM image, and (b) Displays the denoised output from the unsupervised neural network on 1024 x 1024 pixel movie of 1124 frames.

### References :

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- [5] We gratefully acknowledge the support of the following NSF grants (OAC 1940263, OAC 1940097, CBET 1604971 and DMR 184084). We also acknowledge the support from DOE grant BES DE-SC0004954. The authors acknowledge HPC resources available through ASU, and NYU as well as the John M. Cowley Center for High Resolution Electron Microscopy at Arizona State.