The Impact of Artificial Intelligence on *In Situ* Electron Microscopy

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A primary goal of *in situ* electron microscopy is to follow the atomic-level structural evolution taking place in a material due to an applied stimulus. The *in situ* field started several years after the invention of the transmission electron microscope (TEM) in 1935 [1], and there have been constant developments in the areas of closed windowed cells and differentially pumped open cells. Each generation offers advantages over the previous technology, opening up new doors for scientific discovery. Wilbur Bigelow recognized the critical importance of instrumentation development with his work on hot stages and gas cells [2-4]. In recent times, new instrumentation based on direct electron detectors has almost eliminated detector noise, resulting in superior quality data with signal-to-noise ratios (SNR) associated only with electron counting.

To follow structural evolution with improved time resolution, short exposure times must be used, resulting in weak image signals, making it essential to develop approaches to mitigate the effect of noise. Artificial intelligence (AI) offers a potential path forward, with denoising techniques based on convolutional neural networks showing great promise for electron microscopy. We have had success in developing and applying supervised convolutional neural networks to denoise TEM data from catalytic nanoparticles [5, 6]. Recently, we have been looking at unsupervised denoising methods that are trained strictly using experimental data, eliminating the need to simulate large training datasets that might deviate from the real data. Such approaches become feasible when large quantities of data are available, as is the case with movies that are generated during *in situ* experiments. We have developed an unsupervised deep video denoiser (UDVD), which we are using to reveal the atomic-level structural dynamics in catalytic nanoparticles at time resolutions approaching one hundredth of a second [7].

To illustrate how the denoiser can reveal structural dynamics, a Pt catalyst, consisting of Pt nanoparticles supported on a CeO_2 support, was imaged during exposure to CO gas at varying temperatures and pressures [8]. Experiments were performed on a Thermo Fisher Titan environmental transmission electron microscope and images were recorded with a Gatan K3 direct electron detector with an electron dose rate of 600 $e^{-}Å^{-2}s^{-1}$ and a frame rate of 75 frames per second. The average number of electrons at each pixel in the vacuum region in each frame was 0.46± 0.7 to give an SNR of ~0.6. The UDVD denoising network uses a blind-spot architecture that estimates the intensity of a pixel by using its surrounding spatio-temporal neighborhood [7]. It can produce high-quality denoising even when trained exclusively on a single noisy video.

Figure 1A shows two images recorded 0.2 s apart from a small cluster of Pt particles in a CO atmosphere at room temperature. In the raw data, noise makes it difficult to determine the location and structure of the particle surfaces. The output from the denoiser clearly reveals the location and image contrast from the particle. To validate the UDVD denoiser, we compare a raw 40 frame averaged image with a denoised 40 frame average over the same time range. **Figure 1B** shows that there is good agreement between the two averages, suggesting that UDVD is making a reasonably accurate estimate of the pixel values.

Figure 2A shows a series of 9 consecutive denoised frames of exposure time 0.013s from a Pt nanoparticle. During the total time (~ 0.1s), the nanoparticle is seen to undergo significant structural transformations. The atoms making the initial particle edge (marked with a blue star in frame 1), composed of two intersecting (111) facets, appear to migrate onto the left (111) facet forming a short stacked double layer which is highly fluxional, as indicated by the continuous line contrast (frame 5). These atoms then re-arranged themselves into a new stable (111) layer and a very short, unstable (100) facet (frame 9). Throughout the process, the entire nanoparticle undergoes an anticlockwise rigid body rotation of ~10^{o}. **Figure 2B** shows the same 9 frames summed together to simulate the image that would be recorded with a 0.1s exposure time. While the SNR in the summed image is improved, there is no information about the complex series of elementary structural transformations that is revealed in the denoised higher temporal resolution image series. This demonstrates that AI combined with *in situ* TEM, will provide new insights into atomic-level structural dynamics that were previously inaccessible.

References

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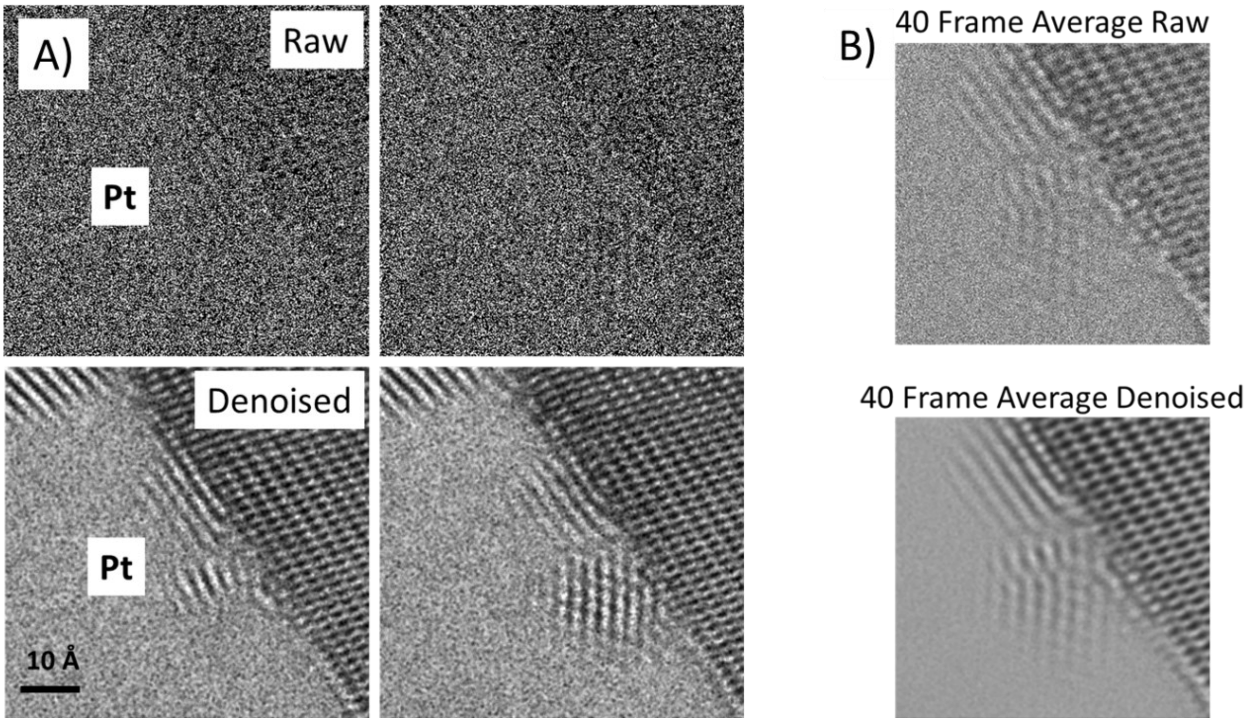

Figure 1. A) Images showing structural changes in irregularly shaped Pt nanoparticles at different points in time (left 0s, right 0.2s). Top row is raw data (0.013s exposure time), bottom row same frame after UDVD denoising. B) 40 frame average of raw data (upper) and denoised output (lower).

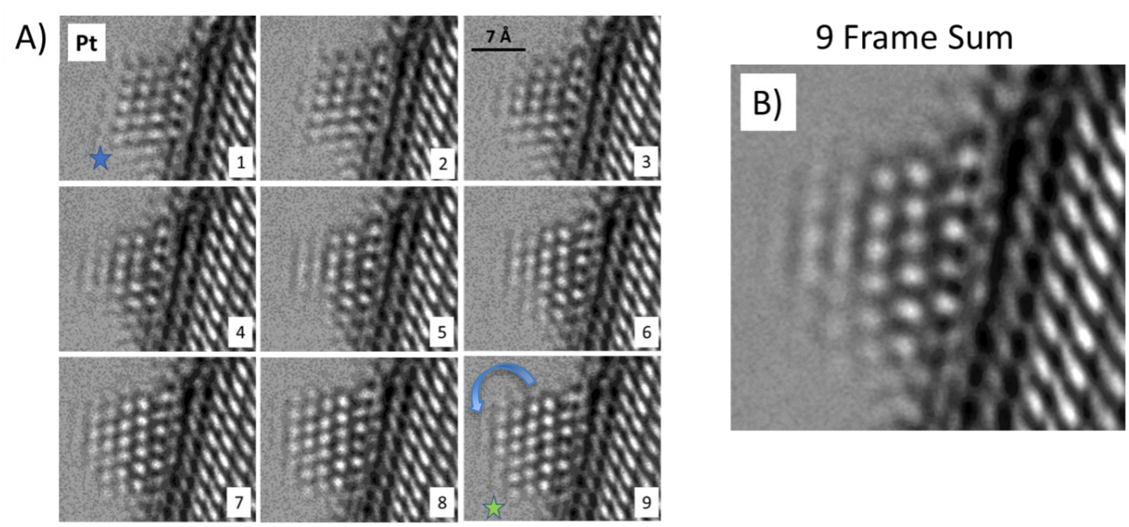

Figure 2. A) Consecutive series (1 – 9) of denoised images from Pt nanoparticle recorded every 0.013 s showing structural dynamics. Initial particle edge is marked with a blue star in frame 1 and final (100) very short surface marked with green star in frame 9. In frame 9, particle is rotated 10^{o} anticlockwise with respect to frame 1. B) Same 9 frames summed to give the equivalent image recorded in 0.1s.