Extracting High Spatio-Temporal Information using Machine Learning from Pt Nanoparticles in CO Gas Environment

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In the field of *in situ* transmission electron microscopy (TEM), the availability of new direct electron detectors is opening up completely new areas of atomic-level materials characterization. The faster acquisition time offers the possibility of exploring structural dynamics with improved temporal resolution. However, for moderate electron doses, faster acquisition time result in poor signal-to-noise (SNR) in single image frames hindering in the extraction of structural information. Machine learning offers a potential path forward, with denoising techniques based on convolutional neural networks showing great promises for electron microscopy. 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 is revealing previously unseen atomic-level structural dynamics in catalytic nanoparticles at time resolutions approaching one hundredth of a second [1]. In this work, with the help of unsupervised video denoising techniques, we are investigating the structural dynamics/transformations taking place on a platinum (Pt) nanoparticle supported on ceria when exposed to gas environment inside the microscope [1]. Our particular interest is the dynamic strain field and structural transformations that are induced in the particle upon exposure to carbon monoxide (CO) gas. Pt supported on ceria is one of the most commonly used material to study CO oxidation reaction used in automotive emission control [2]. With the help from the UDVD denoiser, it is possible to map the evolution of the atomic-level strain fields with time resolutions approaching ~0.01s.

Pt supported on ceria was imaged at a frame rate of 75 frames per second using a Gatan K3 direct electron detector on a Thermo Fisher Titan environmental transmission electron microscope operated at 300 kV. The nanoparticle was imaged with an electron dose rate of 600 $e^{-}Å^{-2}s^{-1}$ at a CO gas pressure of 7 x 10^{-4} Torr inside the microscope. The UDVDcode was used to denoise the *in situ* dataset. The typical movie sequences were 1000 frames or more to ensure successful denoising with UDVD. Atomic column positions were located using a custom-written Gaussian peak fitting algorithm [3]. Strainmaps were generated from each frame by measuring the spacing between atomic columns and normalizing it to the bulk spacing along the equivalent direction.

Fig 1 shows the effect of the denoising on a single 0.013 sec frame from a Pt nanoparticle. In the denoised data, high SNR greatly facilitates the detection of the atomic column positions with much higher precision. With the help of denoised images, it can be seen (in **fig 2)** that, over the time of

13 frames (~0.15 sec), the Pt nanoparticle shows shearing of atomic columns at the (111) plane. The shear results in the introduction of a stacking fault where the usual "ABCABC" stacking of the (111) planes in the FCC Pt crystal structure (as marked in **fig 2a**) changes to an "ABAB" type stacking (as marked in **fig 2c**). During shearing, the shear plane appears as a streaked line (as marked in **fig 2b**) for about two frames suggesting some sort of dynamic transition state that lasts a few hundredths of a second. Also, during the shearing, the nanoparticle goes through a clockwise rigid body rotation of ~7.5° apparently triggered by the downward translation of the shearing plane (C6 in **fig 2b**).

Strain maps with a time resolution of 0.01s have been generated from the denoised data. Strain fields (mostly tensile in nature) are quite random and present over the entire nanoparticle with more severity near the surfaces which might be associated with the stretching of the Pt-Pt bond resulting from the CO adsorption. 3 instances are shown in **fig 2d, 2e, and 2f** where the strain fields next to the shearing plane show significant changes arising probably due to the shearing-induced distortions in the next layer of atoms (C6 layer shows a wiggle). For example, during the transition state of shearing, the layer marked as B5 shows a high degree of tensile strain compared to the other two cases. More such occurrences are being studied with the high temporal resolution using the strainmaps in the same gas environment to understand different structural transformations and associated active regions in the nanoparticle gas interactions.

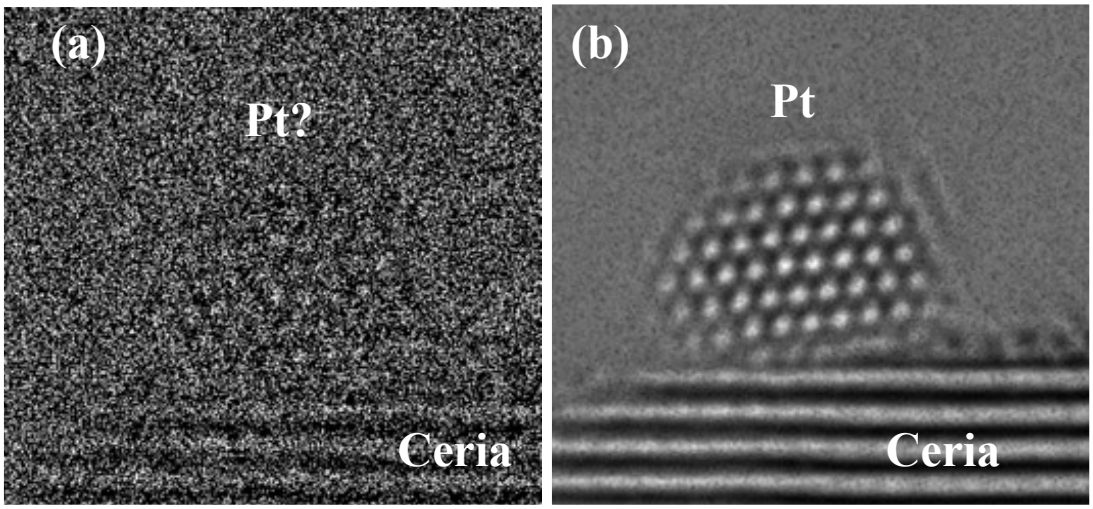

Figure 1: (a) and (b) show the raw data from a single frame and denoised single frame of the same nanoparticle of Pt supported on ceria respectively. The Pt nanoparticle is clearly visible in (b) while in (a) only faintly due to poor SNR.

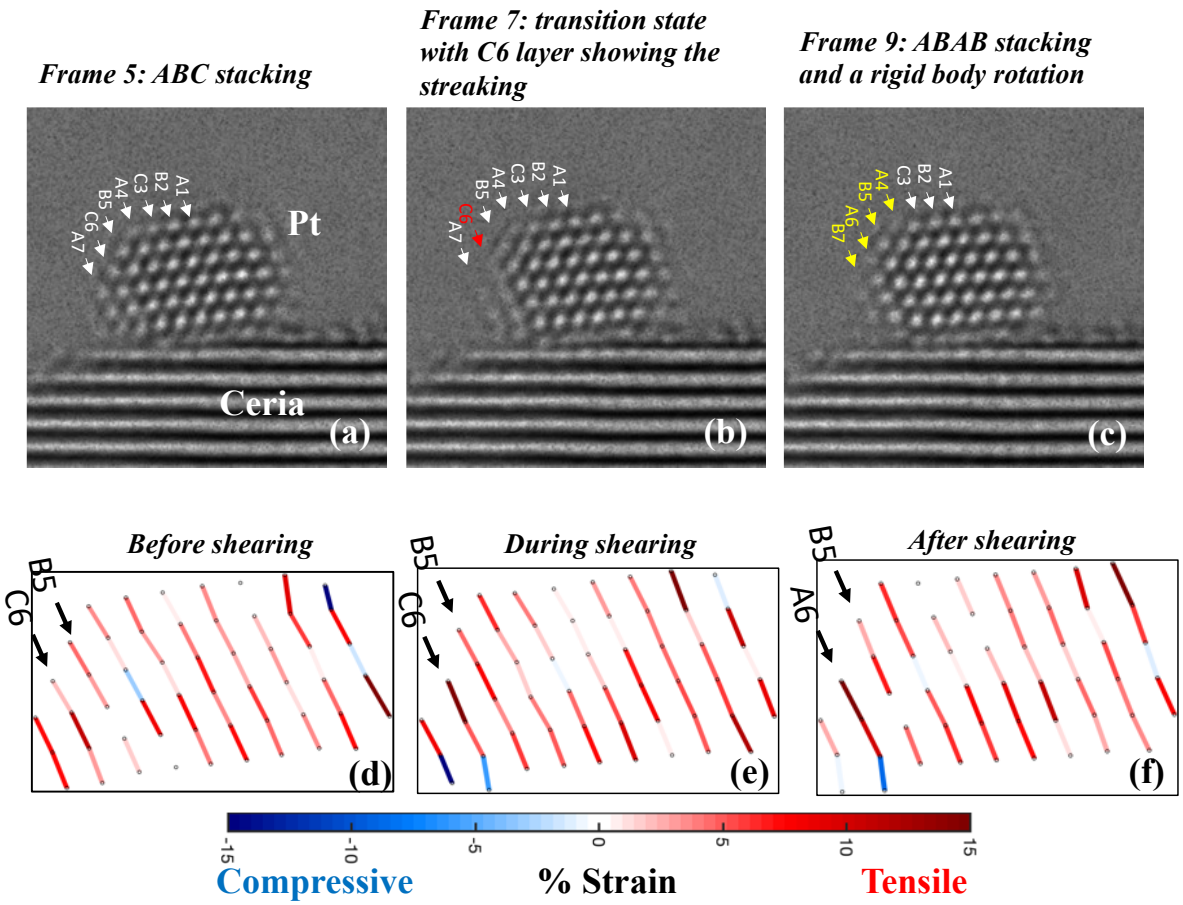

Figure 2: (a), (b) and (c) shows 3 frames over which the shearing and rigid body rotation takes place in the Pt nanoparticle supported on ceria. "ABCABC" stacking is marked in (a) which changes to a "ABAB" stacking (marked by yellow) in (c). In (b) transition is seen as a streaking of the atomic layer C6. In (d), (e), and (f) strainmaps are shown before, during, and after shearing respectively. C6 plane is undergoing the shearing while during the transition state in (e), high degree of tensile strain is visible in B5 layer. The color-bar represents tensile (red) and compressive (blue) strain fields.

References:

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