

Approaches to Monitoring Structural Modification Using *In Situ* Electron Microscopy

Thomas W. Hansen^{1*}, William Bang Lomholdt¹, Matthew Helmi Leth Larsen², Cuauhtémoc Núñez Valencia², Jens Kling¹, Daniel Kelly¹, Pei Liu¹ and Jakob Schiøtz²

¹ DTU Nanolab, Technical University of Denmark, DK-2800, Kgs. Lyngby, Denmark

² Department of Physics, Technical University of Denmark, DK-2800, Kgs. Lyngby, Denmark

* Corresponding author: thwh@dtu.dk

Understanding the atomic-scale structure of materials and devices, as well as the potential structural changes that may occur during use in real-world operating conditions, is essential to developing next generation of functional materials. Such materials can provide new solutions within a diverse range of fields including chemistry, electronics, and medicine. In the energy sector, advanced nanomaterials are used in energy storage, power-to-X catalysts and many other devices. The core challenge of characterizing such materials is this: how do we obtain the most detailed atomic-resolution pictures of systems and devices under real-world conditions, without affecting them by our imaging process? State-of-the-art transmission electron microscopy techniques can obtain sub-angstrom spatial resolution and high spectroscopic and temporal resolution datasets of devices operating in gas or liquid environments. However, making these observations relevant to real world applications requires minimization of electron beam effects and reliable analysis of data acquired under low dose rate conditions.

Minimizing electron dose rate during data acquisition reduces the impact of imaging but at the cost of a reduced signal to noise ratio of the acquired data. The result is often that the data becomes so noisy that it is not analyzable using a manual approach. An automated approach which has gathered significant traction over the last decade is machine learning and convolutional neural networks. Well-trained networks, where noisy data is included in the training set have shown promising performance, even on *in situ* data. Figure 1 shows a gold nanoparticle imaged at in 4.5 Pa carbon monoxide at 300°C [1, 2]. The positions of atomic columns have successfully been located using a convolutional neural network. Analyzing sequences of such frames reveals the dynamic nature and structural variations due to changes in the environmental conditions. Data analysis by means of convolutional neural networks is also efficient for analyzing images of low-Z materials with poor contrast. Figure 2 shows a hole in a graphene sheet induced by an intense electron beam. Frames were subsequently acquired at low electron dose in order to minimize irreversible structural changes to the sample. The data was acquired at low acquisition times and summed to obtain images which could be analyzed with high accuracy [3, 4]. The figure shows both the output of the neural network from a set of summed frames as well as stress/strain maps derived from this data.

In this paper, we will discuss approaches to acquiring images of dynamic phenomena in order to provide statistically relevant, reliable and user unbiased materials science conclusions.

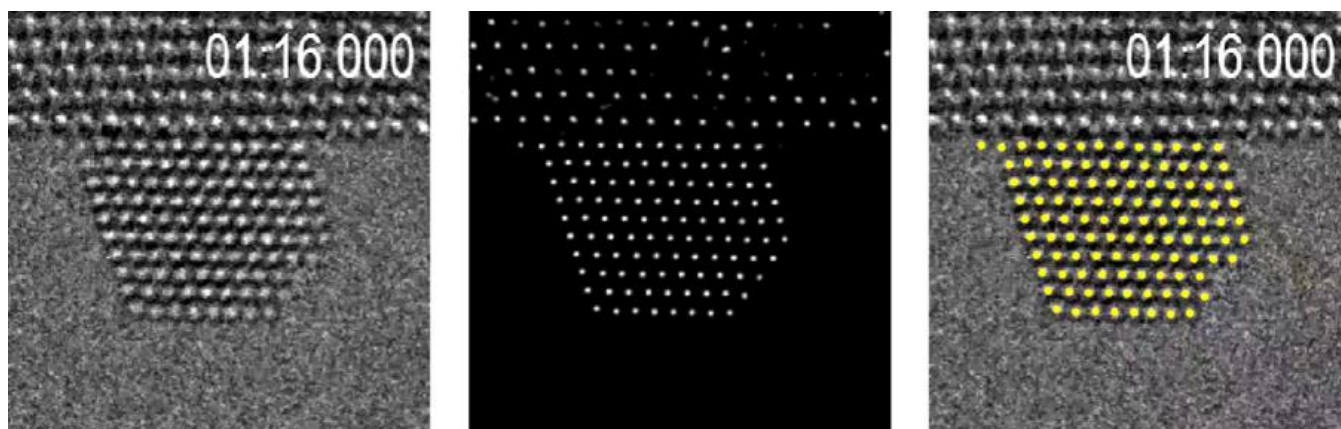


Figure 1: A gold nanoparticle on a cerium dioxide substrate acquired at 450°C in 4.5 Pa CO. Left: raw data; middle: output of a convolutional neural network; right: overlay of raw data and analysis output.

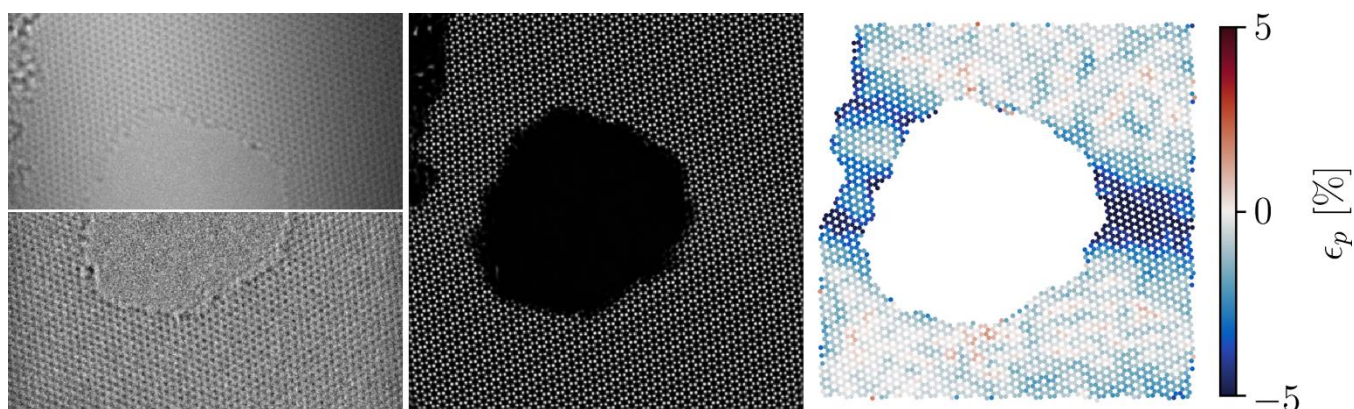


Figure 2: Electron beam induced hole in a graphene sheet. Left upper: raw data; left lower: renormalized summed frames; middle: output of neural network where atomic positions have been identified; right: strain mapped indicating the stress and strain in the vicinity of the hole.

References:

- [1] P. Liu, J. Madsen, J. Schiøtz *et al.*, *J. Phys.: Mater.* **3** (2020), 024009.
- [2] P. Liu, T. T. Wu, J. Madsen *et al.*, *Nanoscale* **11** (2019), 11885-11891.
- [3] J. Madsen, P. Liu, J. Kling *et al.*, *Advanced Theory and Simulations* **1** (2018), 12.
- [4] J. Madsen, P. Liu, J. B. Wagner *et al.*, *Adv. Struct. Chem. Imag.* **3** (2017), 14.