Exploring Spatiotemporal Limits for Atomic Resolution *In Situ* Electron Microscopy

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The remarkable capabilities of modern transmission electron microscopy have dramatically extended our vision, enabling us to see fundamental atomic level processes in ways that were previously not possible. Lena F. Kourkoutis recognized how compelling both qualitative and quantitative visual images are for exploring materials and talked about "seeing with electrons", her Cornell group's informal logo. Lena was aware of the importance of characterizing not only static materials' structures/compositions but also structural dynamics. Indeed, one of her first papers was an *in situ* electron microscopy study on the kinetics of Er diffusion on Si [1]. She used photoelectron emission spectroscopy to perform elegant experiments to investigate strain and diffusion on nucleation and growth processes. The images provided a compelling view of the surface processes taking place and, with suitable image processing, she was able to come up with kinetic parameters to quantify the observations. As her career developed, she became an expert in many techniques available in the scanning transmission electron microscope (STEM) and successfully applied these to a wide range of problems in energy and quantum materials. Like her mentor, David Muller, she was never happy with the microscopy instrumentation status quo and enjoyed pushing the techniques to higher levels. Her pioneering work on atomic resolution STEM imaging and spectroscopy of complex oxides at cryogenic temperatures demonstrated her willingness to push the tools to the limits. She was passionate about developing cryo-STEM techniques for the exploration of quantum materials and I am sure she would have been an international leader in this emerging field. She provided great service to the microscopy community including several leadership roles in the Microscopy Society of America and was also active in providing feedback and guidance at the federal level to funding agencies. It was my great privilege to work with Lena on organizing an NSF workshop to explore future directions for electron microscopy [2].

Characterizing solid-liquid and solid-gas interfaces is an ongoing challenge in the field of electron microscopy with potential to impact many technologies. Lena was particularly interested in the former while I concentrate more on the latter. In the area of *in situ* electron microscopy, there is a constant drive to improve temporal and spatial resolution. The spatio-temporal dream machine would be one that allows us to visualize atomic arrangement nears surfaces, interfaces and defects with spatial resolution $< 10^{-10}$ m, while simultaneously observing non-cyclic atomic dynamics on molecular time scales with temporal resolution of 10^{-12} s. Unfortunately, this dream may never become a reality for several reasons. At present we can either have high temporal (ultrafast TEM techniques ~ 10^{-12}s) or high spatial resolution (aberration corrected TEM/STEM) but we cannot have them both at the same time. There are many reasons for this apparent "*uncertainty principle*". On the ultrafast side, the Boersch effect limits spatial resolution and this appears to be fundamental. For TEM/STEMs lacking ultrafast technologies, the temporal resolution is in part limited by detector hardware readout rates. For the current generation of direct electron detectors, readout rates in excess of 10^4 Hz are now available with improving efficiencies and counting rates. It is not clear what will ultimately limit high quality readout rates, but megahertz seems likely in the near future and gigahertz may be achieved for at least some architectures.

Does this open up the future of being able to perform atomic resolution imaging, spectroscopy and diffraction with 1 Å spatial resolution and time resolution at the microsecond or nanosecond level? It will depend on the application, but signal-to-noise and radiation damage will usually be limiting factors. Fig. 1 shows the relationship between spatial precision, temporal precision, and electron dose for a typical atomic resolution phase contrast imaging experiments (similar graphs can be generated for STEM situations) [3]. If a spatial precision of 1 Å is desired, the sample exposure should be around 2 x 10^3 e/ Å^2. For temporal precisions of 1 ms, 1 μs or 1 ps, fluence rates of 2 x 10^6 e/ Å^2 /s, 2 x 10^9 and 2 x 10^{15} e Å^2 /s are required, respectively. These rates are clearly going to destroy many if not all materials. One may be able to use complex STEM raster patterns or pulsed electron beams to try and deliver the dose in different ways, but it is unlikely to substantially alter the conclusion. This analysis shows that as detector technology improves, radiation damage will be the primary limitation on temporal resolution so any strategies to mitigate beam damage should be aggressively pursued. Limiting the electron dose to minimize damage will result in datasets with high temporal resolution that are severely degraded by shot noise.

Many groups are managing the effect of noise with machine learning using either segmentation techniques (to determine atom positions, intensities etc.) or direct denoising of images, spectra, and diffraction patterns. We are particularly interested in unsupervised convolutional neural network approaches that make no prior assumptions about the form of the data stream. This is particularly important for our *in situ* investigations of structural dynamics in nanoparticles where the atomic resolution images are formed from crystals that constantly change their orientation and structure yielding images with continuously varying contrast. Noise reduction techniques are helpful but must be approached with caution since they can introduce artifacts. We will show practical examples of denoising on images, diffraction patterns and energy-loss spectra. Practical approaches to increase the degree of confidence in the output of such analyses will be discussed.

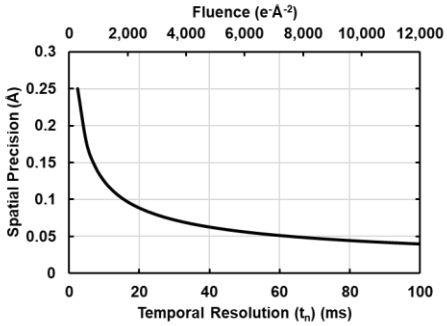

Fig. 1. Relationship between spatial precision, temporal resolution, and electron fluence (from [3]).

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2. NSF, *Developing a Future Vision for Electron Microscopy*. 2020: https://sites.coecis.cornell.edu/temfrontiers/.
3. Lawrence, E.L., et al.Microscopy and Microanalysis, 2020. **26**(1): p. 86-94.
4. We gratefully acknowledge the support of the following NSF grants (OAC 1940263, 2104105, CBET 1604971 and DMR 1840841). We also acknowledge the support from DOE grant BES DE-SC0004954. The authors acknowledge HPC resources available through ASU as well as the John M. Cowley Center for High Resolution Electron Microscopy at Arizona State University.