

Advances in STEM and EELS: New Operation Modes, Detectors and Software

A.L. Bleloch¹, N.J. Bacon¹, G.J. Corbin¹, N. Dellby¹, M.V. Hoffman¹, M.T. Hotz¹, P. Hrnčirik¹, N. Johnson¹, T.C. Lovejoy¹, C.E. Meyer¹, A. Mittelberger¹, B. Plotkin-Swing¹, G.S. Skone¹, Z.S. Szilagy, and O.L. Krivanek^{1,2*}

¹ Nion R&D, 11511 NE 118th St, Kirkland, WA, 98034, USA

² Department of Physics, Arizona State University, Tempe AZ 85287, USA

* Corresponding author: krivanek@nion.com

Advances in the capabilities of scanning transmission electron microscopes (STEMs) and electron energy loss spectrometers (EELS) over the last 2 decades have been remarkable. Modern-day monochromated, aberration corrected STEMs (MAC-STEMs) can give probe sizes of 1.1 Å at 30 kV operating voltage [1], EELS energy resolution of 4.2 meV also at 30 kV [2], and spatial resolution of ~0.3 Å through the use of 4D STEM and ptychographic reconstruction [3]. These achievements have been made possible by aberration correction, and by developments in monochromator, spectrometer and detector design. The electron-optical developments are now maturing, and further improvements in this area are likely to be more incremental. Here we summarize our work on endowing Nion electron microscopes with new capabilities via new operation modes, detectors and software.

One potentially transformative new capability is to map vibrational properties of materials in reciprocal space, i.e. to map the momentum-dependence of vibrational losses [4]. The needed data set is similar to the $(X, Y, \Delta E)$ energy loss data cube familiar from spectrum-imaging, but with the two spatial coordinates replaced by scattering angles θ_x and θ_y , typically rescaled into momentum space $q = 2\pi \theta / \lambda$. It promises to give similar information on phonon modes in solids as has been traditionally provided by neutron scattering [5] and more recently inelastic X-ray scattering, but from much smaller volumes. However, the signal is weak, for two reasons: a) the cross-sections are small, and b) when monochromating for 5-10 meV energy resolution, the electron beam current is typically only 3-15 pA.

Hage et al. [4] acquired ω - q line profiles (aka “ ω - q diagrams”) through the data cube serially, by defining the scattering angles accepted by the EEL spectrometer with a small entrance aperture and shifting the diffraction pattern over the aperture. This method is flexible but inefficient, resulting in acquisition times of tens of minutes or even hours per ω - q diagram. A more efficient collection method consists of placing a slot aperture in the spectrometer entrance plane, and adjusting a diffraction pattern projected onto the slot to encompass the momentum range of interest [6]. Fig. 1 shows an ω - q diagram obtained in this way along the Γ ->M line in h-BN. The diffraction pattern was rotated using the microscope’s post-sample lenses to project the desired Brillouin zone line onto the slot aperture, 125 μm wide and 2 mm long, and the data was summed over 30 acquisitions of 10 seconds each. The information is equivalent to Fig. 2 of [6], but the acquisition time was much shorter – about 5 minutes.

Acquiring ω - q data as above places high demands on the EELS detector. It needs to give a wide dynamic range (10^5 :1 and higher), narrow point spread function (PSF) so that the high intensity of strong Bragg discs does not “spill” into the much weaker signal at $\Delta E > 0$, $q > 0$ immediately next to the discs, and ideally single-electron sensitivity. The detector used to record the data for Fig. 1 was an ultra-low noise SCMOS camera lens-coupled to a P43 scintillator. Its main strength lies in combining high speed (400 Mpixels / second, i.e. 100 2kx2k frames per second, or 1000 EEL spectra of 2048x200 pixels

per second) with ultra-low noise (1.6 well e^- r.m.s.) read-out. However, it only gives detective quantum efficiency (DQE) >0.5 with about five 30 keV electrons per pixel, i.e. it is not able to detect single 30 keV electrons. Larger-pixel, fiber-optically coupled sCMOS cameras are able to detect single electrons, but their read-out is $\sim 5\times$ slower, and hence not as well optimized for fast elemental mapping.

The best detector for ω - q diagrams and other EELS applications at 30-200 keV primary energy is likely to be a directly illuminated hybrid pixel detector, with single electron sensitivity and high read-out speed. Fig. 2. shows a core loss spectrum of h-BN recorded on a prototype of such a detector that uses a single Medipix3 chip. The spectrum comprises the zero loss peak (ZLP), BN plasmon and B K-edge, all recorded in the single electron detection mode, with DQE ~ 0.8 . The PSF is much better than for scintillator-based cameras: only about 1.2 pixels wide, and the drop-off of the intensity on the energy gain side is especially impressive: down to 2×10^{-5} within 3 pixels. In order to avoid saturation, the current in the ZLP was set to a low value of 0.7 pA and spread over 16 pixels in the non-dispersion direction. Had the ZLP been spread sideways to cover the full 256 pixel width of the sensor, a ZLP current of 11 pA could have been recorded, but this value is still too low for many practical applications. We are therefore developing a larger-format direct detection camera for EELS applications whose saturation threshold is about 10x higher, and expect to report on it at the meeting.

Other developments we intend to review at the meeting include testing the Nion microscope's ability to protect its cold field emission gun (CFEG) in the presence of large gas pressures at a sample in an aperture-type environmental gas cell, which showed that pressures up to ~ 10 torr at the sample are feasible with appropriate differential pumping; and new software developments, especially in the areas of probe, monochromator and spectrometer autotuning.

- [1] O.L. Krivanek et al., *Microsc. Microanal.* **24** (Suppl 1, 2018), 18.
 [2] O.L. Krivanek et al., *Ultramicroscopy* (2019) in press, doi.org/10.1016/j.ultramic.2018.12.006
 [3] Y. Jiang et al., *Nature* **559** (2018) 343-349, doi.org/10.1038/s41586-018-0298-5.
 [4] F.S. Hage et al., *Sci. Adv.* 2018;4:eaar7495 1-5.
 [5] P.C.H. Mitchell et al., *Vibrational Spectroscopy With Neutrons*, World Scientific (2005).
 [6] G. Mollenstedt, *Optik* **5**, 499 (1949).

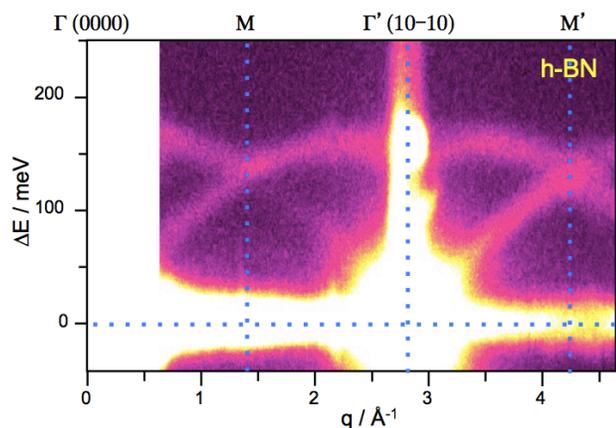


Figure 1. Experimental ω - q diagram of BN along the Γ -M line, recorded in parallel in 300 sec. Nion HERMES, 30 keV. Saturation was minimized by excluding the Γ point from the slot aperture.

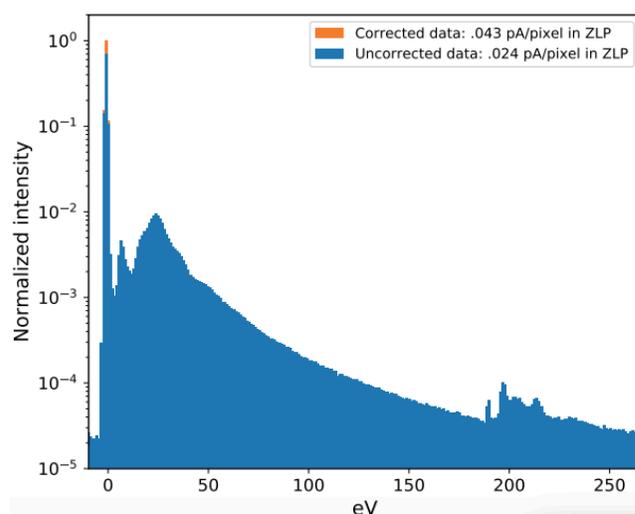


Figure 2. EELS of BN acquired with a Medipix 3 256x256 hybrid pixel detector. 30 keV, 20,000 x 0.5 ms exposures (total exposure 10 s).