Multi-Sun EELS: Ultra-High Energy Resolution combined with High Spatial Resolution and High Beam Current

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Since its introduction in 2014, vibrational spectroscopy in the electron microscope [1] has led to many remarkable results. Examples include distinguishing between ^{13}C vs ^{12}C in an amino acid (L-alanine) and mapping their distribution with ~30nm resolution [2], detecting the vibrational signal from a single defect in a solid [3], and recording the vibrational spectrum of a single atom [4]. Other examples can be found in a recent review of the progress [5]. The field continues to advance rapidly, facilitated by continuing advances in the monochromator and the spectrometer [6], and by the development of new operating modes. Figure 1(a) shows a monochromated zero loss peak (ZLP) acquired at 60 kV with a full width at half maximum (FWHM) of 3.2 meV, using a beam current of 0.3 pA.

Challenges do remain, however. A principal one is that monochromation reduces the available beam current very substantially. With an optimized cold-field-emission source, the ZLP is ~300 meV wide, and reducing the energy distribution to ~3 meV leaves <1% of the current. Increasing the unmonochromated beam current leads to larger crossover (probe) sizes at the sample and also at the energy selecting slit of the monochromator and at the EELS detector, and this results in worsened spatial and energy resolution. Selecting 300 pA as a reasonable compromise for the unmonochromated beam current gives < 3 pA monochromated current at the highest energy resolution, and this is not optimal for recording energy loss features which are 10^2-10^4 times less intense that the ZLP.

The root of the problem is that we need to minimize three parameters simultaneously: the energy, spatial and angular resolutions. This limits the "phase space" of the electron beam, and thus the current in the beam. Relaxing any one of the three constraints can lead to significantly larger beam currents. In many instances of practical analysis, such as when probing a material with an aloof beam [7], angular resolution is not a major concern, and illuminating the sample from many directions simultaneously can increase the beam current substantially. To do this, we set up an illuminating beam of narrow angular spread in one direction, a one-dimensional "nanodiffraction" condition (narrow in one direction, wide in the other). Then we use a modified optical regime in the monochromator to disperse different energy sub-beams in angle at the sample. The size of the probe on the sample in the energy dispersion direction is determined by the angular extent of each individual energy. We then adjust the spectrometer focus so that illumination sub-beams deviating from the average beam energy by ∂E are brought onto the same place on the EELS detector. Energy losses ΔE imparted at the sample arrive at different positions on the detector, i.e. the regular energy dispersion is maintained. The energy-selecting slit is not used in this mode—the energies that are allowed to pass are determined by the angular dispersion and the angular range selected by subsequent optics. In essence, we are using multiple sample-illuminating sub-beams that have different energies, but give identical spectra at the EELS detector that are summed. We call the technique "multi-sun EELS".

Figure 1(b) shows an aloof multi-sun EELS spectrum of guanine, a building block of DNA, acquired at 30 keV primary energy. The resolution FWHM is 5.5 meV, and the current in the spectrum 15 pA. The current admitted into the monochromator was 40 pA. Conventional filtering would give <1pA in this case. The spatial resolution, shown in the inset, is < 10 nm, sharper than the particle edge. The spectrum is rich with information: the features between 300 meV and 450 meV correspond to different types of hydrogen vibrations [7], the peak around 200 meV to the C=O bond, and the features down to ~25 meV correspond to the myriad vibrational modes of the guanine molecule. Note that for this spectrum, the data is plotted as Intensity*ΔE^2 vs. ΔE. This removes the ZLP by design (without subtraction) and improves the visibility of weak features.

It is also possible to create energy dispersion such that different energy sub-beams arrive at different positions on the sample, at the cost of spatial resolution in a particular direction [8, 9]. When the energy dispersion at the sample is made to align with the spectrometer dispersion direction, a variation of multi-sun EELS should be possible to "reassemble" the different illumination sub-beams while maintaining angular scattering information, such as for angle-resolved EELS [10], at much higher beam current.

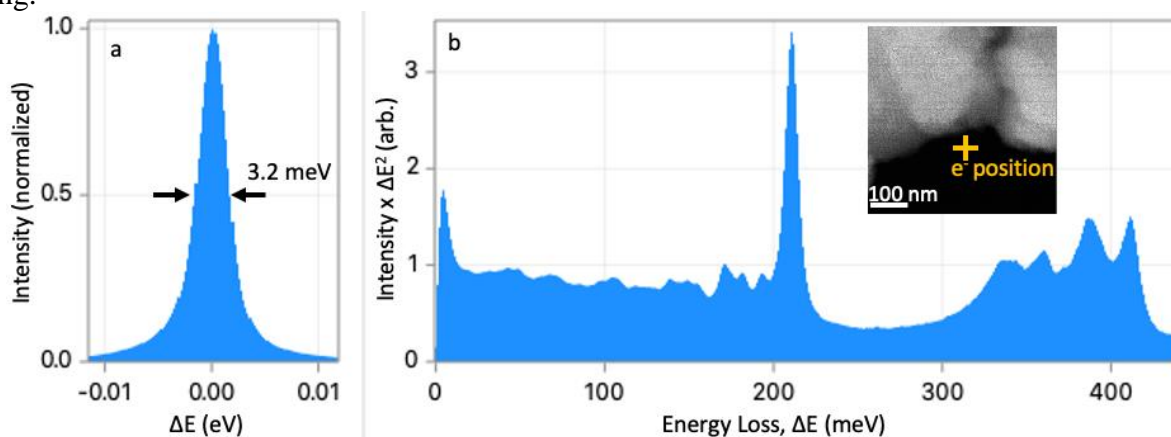Related developments and further applications of the multi-sun technique will be reported at the meeting.

Figure 1. (a) ZLP with 3.2 meV FWHM acquired at 60kV. Beam current is 0.33 pA, 60 s total (60,000 x 1ms, aligned) (b) aloof multi-sun EELS spectrum of guanine acquired at 30 kV with (inset) scanned image acquired in the same setting showing impact parameter ~55nm. Beam current is 15 pA, 40% of the current admitted to the monochromator (40pA). FWHM of ZLP is 5.5 meV (not shown). 90 seconds total (30,000 x 3 ms, aligned). Spectrum is plotted as Intensity*ΔE^2 vs ΔE, which removes the ZLP and increases visibility of weak features.

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