## Advantages of Direct Detection and Electron Counting for High-energy Resolution and Monochromated Electron Energy Loss Spectroscopy Data Acquisition

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Direct detection cameras (DDCs) and particularly the Gatan K2 have revolutionized the cryo-TEM field as well as have strong advantages for in-situ TEM in both imaging and diffraction applications. It's already been shown that EELS applications can benefit from the improved PSF and the ability to count electrons. The improved PSF allows spectra to be acquired over larger energy ranges (low spectrometer dispersions) while maintaining sharp features and greatly reduced spectral tails. The ability to count electrons nearly eliminates the noise associated with detector readout and greatly reduces the proportional noise associated with detector gain variations. This effectively leaves the shot noise as the limiting noise source present. The implication for EELS acquisition is that fine structure analysis becomes more straightforward for typical conditions and even possible for the case of low signal levels. Here, we want to explore the effects of electron counting in the case of monochromated experiments where higher energy resolution is required.

As example of the advantages due to the reduced noise and PSF in the case of monochromated experiments, Figures 1c,d show the EELS spectra of Fe L<sub>2.3</sub>-edges at 708eV and Ti L<sub>2.3</sub>-edges at 456eV extracted from the Fe<sub>2</sub>O<sub>3</sub> and the SrTiO<sub>3</sub> areas within the green box region in the ADF STEM survey image in Figure 1b. Spectra were extracted in STEM mode and the probe was rastered across the SrTiO<sub>3</sub>/Fe<sub>2</sub>O<sub>3</sub> as shown in Figure 1b. The monochromator was excited to deliver an energy resolution of 0.1eV as shown in the low-loss spectrum in Figure 1a. The spectrum was acquired using the K2 (the DD detector for the experiment) in counting mode and the spectrometer setup with a dispersion of 0.1eV/channel that results in a spectrum with an energy range of ~400eV. In this way both the Ti L-edges at 456 and the Fe L-edges at 708eV are present in the same spectrum. Every detail in both the Ti L and the Fe L near edge fine structure can be observed leading to a more accurate and precise chemical analysis and comparison with the results from simulations. Figures 2 show the comparison between spectra acquired using the K2 and CCD (the IDC detector used for the experiment) with the spectrometer setup for both experiments with a dispersion of 0.1eV/channel. The monochromator was excited exactly in the same way resulting in an energy resolution of 0.1eV measured using the K2 as shown in the lowloss spectrum in Figure 1a. In the case of the CCD, a dispersion of 0.1eV/channel results in a spectrum with an energy range of 200eV, thus the contribution from the Ti  $L_{2,3}$ -edges at 456 is now missing. In the case of the K2, given the much reduced PSF, the energy resolution is such that the split of the Fe  $L_3$  peak is very pronounced whereas in the case of the CCD the split is just a shoulder, higher energy resolution would be needed in order to cope with the high PSF and give enough energy resolution for chemical analysis.



Figures 1. All data extracted using the K2 as detector. a) low-loss spectrum showing an energy resolution for the experiment of 0.1eV achieved with a dispersion of 0.1eV/channel; b) ADF STEM survey image, the green region is where EELS spectra were collected point by point; c,d) EELS spectra of Fe L- and Ti -edges at 708eV and 456eV extracted from the Fe2O3 and the SrTiO3 regions inside the green box in Figure 1b. Both the Fe and Ti L signals are extracted from the same spectrum



Figures 2: a) EELS spectra extracted using the K2 in green and the CCD in red. Both spectra are averaged across the entire interface in Figure 1b. for both the K2 and the CCD spectra, the spectrometer was setup with a dispersion of 0.1eV/channel resulting in a spectrum of 400eV for the K2 and 200eV for the CCD in energy range. The Ti L at 456eV is present in the spectrum extracted using the K2 but missing in the case of the CCD; b) comparison of the Fe L-edges spectra in the case of the K2 and the CCD. The energy resolution in the case of the K2 spectrum is much higher, more features and details in the fine structure can be observed.