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High-Fidelity 4D-STEM Enabled by Live Processing at 15’000 Detector Frames Per Second

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4D-STEM acquisition using fast pixelated detectors continues to have a strong impact on the electron microscopy community [1]. It has recently been shown that acquiring and (non-rigidly) registering series of 4D maps can substantially improve the quality of the data, especially for methods like center-of-mass analysis (COM) that rely on assuming correct beam positions and are sensitive to deviations [2]. However, the collection of 4D-STEM series produces massive amounts of data and it is normally necessary to either interrupt the microscopy session to pre-process on the side to roughly assess the data quality before resuming the next measurements or to store and process the data sets afterwards and only then have a full understanding of its quality. Therefore, the handling of those data sets is cumbersome and microscopy time and/or storage space are typically wasted in the process of acquiring good data, especially if one needs a series of 4D-STEM data.

Here, we present live processing at over 15’000 frames per second of a Dectris ELA direct detector [3] mounted on an IRIS spectrometer of a Nion HERMES microscope. The Dectris ELA detector is suitable for EELS measurements, and also for 4D-STEM. It combines the readout noise-free characteristics known from hybrid-pixel detectors with a relatively large number of pixels (1030x514), very high frame rates (up to 18’000 for 1030x130 pixel readout) and a high saturation current (0.8pA per pixel, especially relevant for nano-beam diffraction). The data is acquired, (pre-) processed and displayed using the Nion Swift software [4]. This software now allows to generate live updating images of customizable virtual detectors, such as annular dark-field, bright-field, and COM measurements at 66 µs dwell time [5]; close to values that are typically used when scanning with a normal integrating STEM detector. A 512x512 scan data set is acquired in ~17s.

Thus, 4D-STEM data sets can be evaluated from their virtual detector images as they are being acquired in a mode that resembles the live scanning of integrating STEM detectors. A customizable amount of 4D data sets is cached in the background on hard drive(s) and only the sets chosen by the operator based on the live data processing are permanently stored. The others are dynamically deleted as more data than the defined cache size is acquired (the oldest data sets drop out of the cache). For the data shown here, only the processed virtual detector data (five different detectors) was kept while the raw detector images were discarded. This allowed to acquire and store a series of 28 maps of 512x512 scan pixels of 130x130 detector pixels each resulting in 462 GB raw data as just 280 MB and therefore allows to routinely acquire high quality 4D-STEM series.

Fig. 1 shows a comparison of a single 4D-STEM map, summing 28 consecutively acquired maps, rigid registration of those maps (accounting for translations) and non-rigid registration (also accounting for artefacts from non-directional drift and scan noise using SmartAlign [6]) of a $SrTiO_3$ sample of roughly 15nm thickness, acquired using a convergence angle of 36mrad and 75pA beam current at 60kV. The rigid registration already greatly improves the data quality, and the non-rigid registration leads to even better data. The oxygen columns that are invisible in the HAADF images are clearly resolved.

A comparison of COM analysis using two different angular ranges is depicted in Fig.2 (a); the masks covering two different areas (as indicated in the inset) were applied and the result analyzed (this data was part of the acquisition shown in Fig. 1). The divergence of the COM analysis going out to larger angles better recovers the difference in atomic number of the atomic columns for this relatively thick sample (at 60kV). This is best seen in the inset, which has been created from averaging template-matched regions.

As the detector is mounted on the spectrometer, it is also possible to zero-loss filter the diffraction patterns. To assess the influence of the filtering, two data sets were acquired with and without filtering from adjacent sample areas of a roughly 50nm thick region of $SrTiO_3$. The result from 9 non-rigidly registered maps each is depicted in Fig. 2 (b) in the form of the COM signal and its divergence.

There seems to be no obvious benefit from the filtering for the COM analysis. The similar results might stem from the fact that plasmon scattering (which is the main contribution of the inelastic signal) is delocalized and symmetric (at least in zone axis) and has therefore no obvious effect for the COM. For other techniques that use more than just the first moment of the scattering distribution, like e.g. ptychography, the filtering should be very beneficial.

In conclusion, combining the novel live processing capabilities of Nion Swift with the Dectris ELA detector mounted to a Nion HERMES microscope allows to efficiently acquire 4D-STEM series that can be combined into high-fidelity maps. Although COM analysis seems to be robust in the presence of inelastic scattering from plasmons, other techniques should greatly benefit from being able to zero-loss filter 4D-STEM.

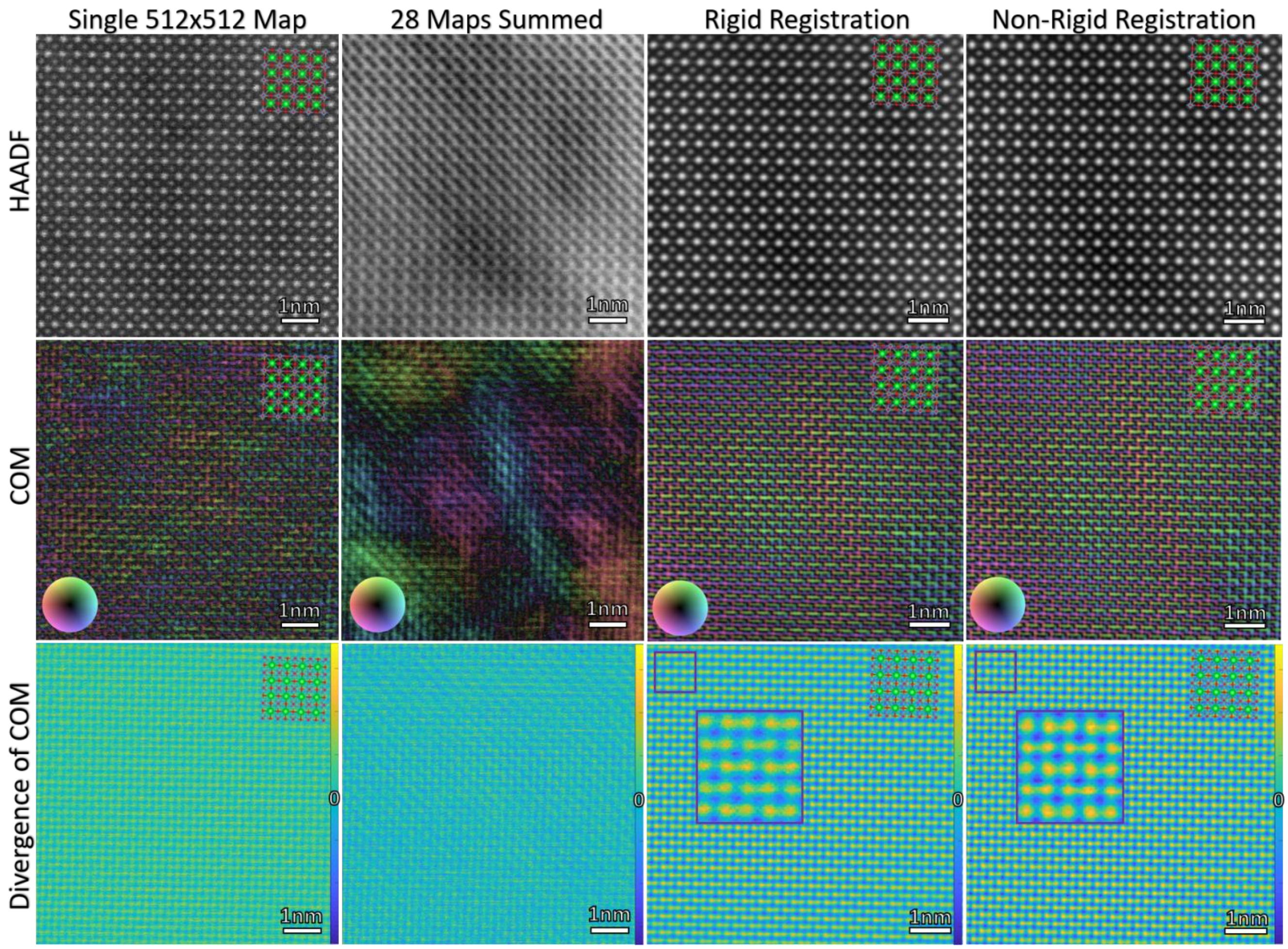

Figure 1. Comparison of a single 4D-STEM map and different registration of a series of 28 maps: HAADF image, center-of-mass analysis and its divergence for a single 512x512 pixel 4D-STEM map, the (not registered) sum of 28 maps, rigid registration (only translations) of those maps and the non-rigid registration result of a SrTiO3 sample of approximately 15nm thickness at 60kV acquired with 36mrad convergence angle and 75pA probe current. The data quality is already greatly improved for the rigid registration, and the non-rigid registration still increases the signal-to-noise ratio (cf. inset).

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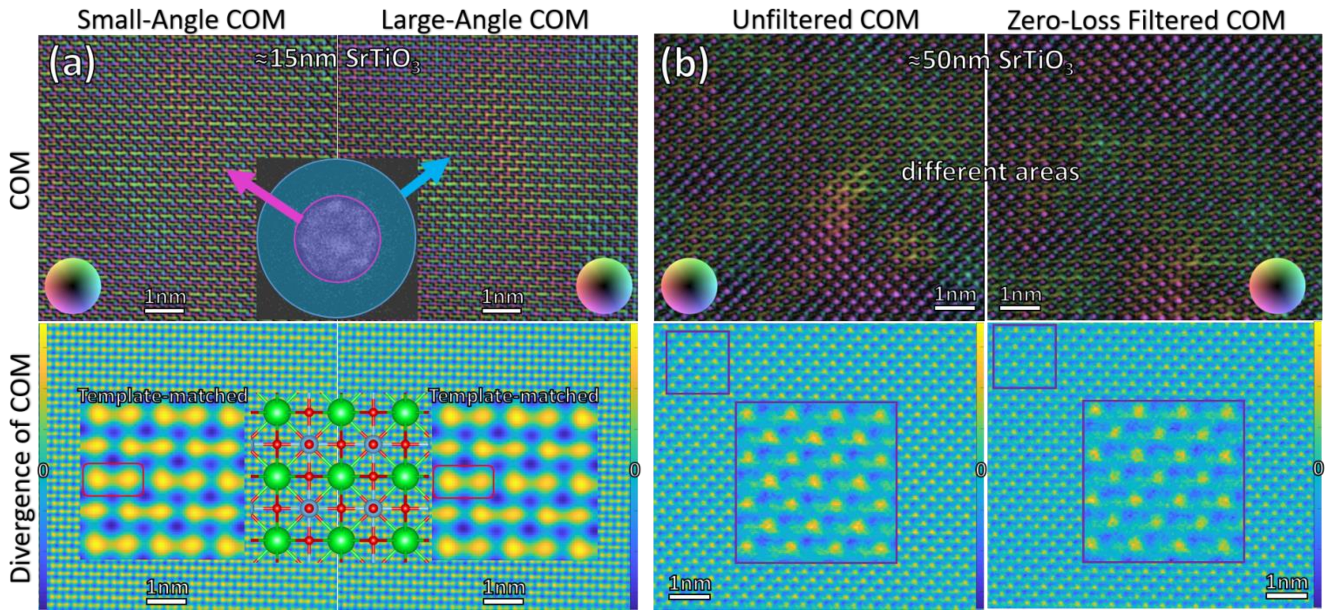

Figure 2. (a) Investigation of the influence of the range of scattering angles on the center-of-mass analysis; for small angles that are barely larger than the convergence angle (0-40mrad) and for larger angles that go out to twice that value (0-80mrad). The collection of higher angles allows to better distinguish different atoms in the divergence map (cf. red maker). (b) Comparison between normal and zero-loss filtered COM analysis from a thicker sample region. There seems to be no obvious benefit from filtering in this case. All data was acquired at 60kV using a convergence angle of 36mrad and 75pA probe current.

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