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Event-Streamed Spectral Imaging in an Aberration-Corrected AEM: A Robust Approach to High Spatial Resolution XEDS Elemental Mapping

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The development of aberration-correcting, multi-pole electron optics has greatly increased the resolving power of the analytical electron microscope (AEM). This technology has rendered the acquisition of atomic-resolution images nearly routine, and atomic-resolution chemical and bond-state mapping via electron energy-loss spectroscopy (EELS) have been achieved and are becoming widespread [1,2]. Nevertheless, a host of applications that could benefit immensely from atomic-resolution elemental-mapping remain inaccessible via EELS analysis due to the low signal-to-background of the characteristic inner-shell ionization edges, and thus poor EELS sensitivity, of many elements. In contrast, X-ray energy dispersive spectroscopy (XEDS) exhibits high and roughly equal sensitivity for all elements with $Z > 4$. However, this advantage is counteracted by the poor signal-collection efficiency of XEDS in the AEM due to the limited solid angle subtended by standard detectors within the limited space afforded by the high-resolution objective lens pole piece. Recent advances in detector design have shown promise for improving collection efficiency by overcoming these geometric constraints [3], but such detectors are not yet commercially available and their implementation in the aberration-corrected AEM does not appear to be imminent. The poor collection efficiency of current generation XEDS detectors can be somewhat mitigated by longer acquisition times; however, this solution inevitably exacerbates the problems of beam damage and spatial drift, which are particularly acute for characterization at ultra-high spatial resolution.

In this paper, we report the application of XEDS event-streamed spectral imaging (ESSI) [4] in an aberration-corrected AEM as a reliable method for the acquisition of ultra-high spatial resolution elemental maps. In contrast to the traditional method of acquiring a spectral image (SI), where the probe is scanned in a single pass over an area of interest with typical per-pixel dwell-times of a few tenths of a second, ESSI enables the collection of the XEDS SI over hundreds or even thousands of passes at rates and pixel densities typically reserved for imaging experiments. The ESSI approach has multiple advantages for aberration-corrected AEM, the first of which is that it provides an alternative, "on-the-fly" method of spatial drift-correction, where the relatively strong image signal is used as a reference to spatially register the more sparse XEDS data in each frame. Secondly, the electron dose imparted to the specimen is spread out over time by virtue of the short dwell-time per pixel (typically a few microseconds), thus reducing the effects dose-rate sensitive beam damage.

The advantages of ESSI over traditional single-pass SI for aberration-corrected AEM are demonstrated by application to a catalyst specimen comprised of palladium nanoparticles on a carbon support, as shown in Fig. 1. The top row of images was acquired using the single-scan approach, using a pixel size of ~ 1 nm. Fig. 1a is a traditional HAADF image, collected from the area of interest prior to the SI acquisition, and Figs. 1b and 1c are the HAADF signal intensity and Pd XEDS map extracted from an SI data cube acquired using a dwell-time of 60 ms per pixel and an overall frame time of 1200 s for the 128 x 128 pixel image. Spatial drift was corrected via periodic cross-correlation analysis of a reference image. The bottom row of images was acquired using ESSI

and real-time drift correction from a larger specimen area, but with a similar pixel size (~ 1 nm) to Figs. 1a-c. In this case, 6000 frames were acquired using a dwell time of 10 µs per pixel, resulting in a total frame time for each 256 x 256 image of 0.6 s. The total data set took approximately 1 h to acquire, and resulted in an overall per pixel signal comparable to that of the single-scan data set. It is obvious that, in the single-scan acquisition, the HAADF image in Fig. 1a, which provides a relatively accurate representation of the catalyst structure, differs markedly from that in Fig. 1b. The latter is plagued by scan faults, imprecise registration of the HAADF signal, and beam-induced specimen alteration, all of which are also present in the Pd-L_α map (Fig. 1c). In contrast, as shown in Fig. 1d, the spatial registration of the image intensity integrated over 6000 frames in the ESSI is excellent, which in turn results in the superior quality of the corresponding elemental maps (Figs. 1e and f). Further applications of this technique, and its potential for enabling robust XEDS characterization at ultra-high spatial resolution in the aberration-corrected AEM, will be discussed.

References

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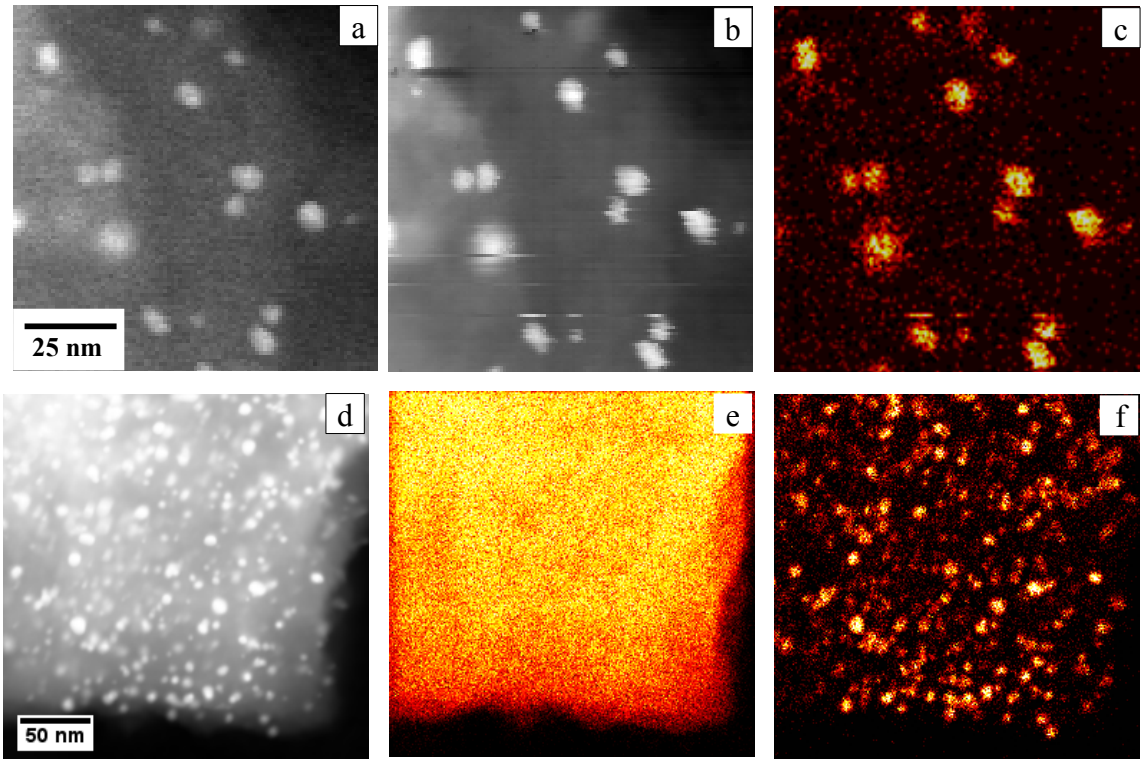

Fig. 1 – Characterization of a Pd/C supported metal catalyst using (a-c) single-scan and (d-f) event-streamed spectral imaging: (a,b) HAADF images acquired (a) prior to and (b) during the single-scan spectral image; (c) corresponding Pd-L_α XEDS elemental map; (d) HAADF image, (e) C-K_α and (f) Pd-L_α XEDS elemental maps acquired using event-streamed spectral imaging over 6000 frames from a larger area but with a pixel size and an overall dwell-time similar to those used in the single-scan approach.

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