Hypermodal data fusion: Big data methods for nanoscale challenges

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The strong interaction of electrons with matter is a huge advantage wielded by the discipline of electron microscopy, with comparatively large scattering cross sections per unit dose emitted with sharp angular dependence in a highly confined geometry. A huge range of techniques related to imaging, diffraction, and spectroscopy has evolved to capture these diverse signals and convert them into scientifically meaningful information, permitting investigation of property domains ranging from local chemistry to crystal structure. These advantages are particularly evident when focused probes are scanned across the sample, as the volumes from which these diverse signals originate can be extremely laterally localized, potentially revealing the positions of individual atoms [1]. However, this diversity of detection techniques can also be a challenge, as many signals are acquired independently and demand specialized processing.

In this report, I describe a general experimental and analytical workflow called hypermodal data fusion that can directly couple disparate data types acquired from an arbitrary number of different detectors and/or collection modes. This yields a versatile statistical model in which variations between statistically significant features from any given dataset are directly correlated to features of any other coupled dataset. I begin by demonstrating how this approach can be used to link co-registered spectroscopy datasets (specifically EELS and EDX) to "extend" their respective spectral ranges [2,3]. I then show how this can be enhanced with blind-source separation to dissect highly complex functional materials into their various morphological, compositional, and electronic components, even estimating the pure underlying spectra at buried heterojunctions in core-shell structures [4]. Finally, I show how scanning diffraction can be included to correlate local crystal structure to spectrally significant features, even in the presence of strong spatial overlap. Figure 1 exemplifies this, where monochromated ELNES features corresponding to the crystal field splitting in compositionally similar TiO_2 polymorphs are directly correlated to individual Bragg reflections in each individual diffraction pattern. Subsequently, the reflections are automatically separated into the corresponding mean diffraction patterns from these individual phases, despite these nanoparticles (80% Anatase, 20% Rutile) being randomly distributed and overlapping.

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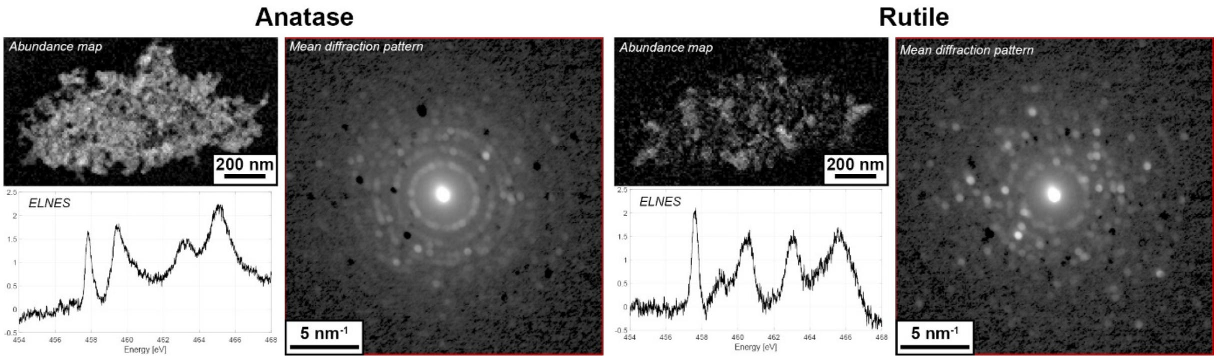

Figure 01 – Hypermodal data fusion between monochromated EELS and scanning diffraction. ELNES features corresponding to the crystal field splitting in TiO_2 polymorphs (Anatase and Rutile) were identified in the core-loss EELS dataset using non-negative minimum volume geometric unmixing. The resulting unmixing matrix can then be directly applied to the coupled scanning diffraction dataset. This allows diffraction features from each individual diffraction pattern (corresponding to Bragg disks) to be associated with a respective EELS spectrum in an unsupervised manner. The resulting mean diffraction patterns shown above closely correspond to simulated powder diffraction patterns from the respective polymorph, as determined by their characteristic ELNES features. This is despite the strong nanoparticle overlap evident in the shown abundance maps (which are subject to a sum-to-one constraint) Note that a third retrieved component corresponding to the Carbon support grid is omitted in this figure for clarity.