

## Atomically Resolved EELS Elemental and Fine Structure Mapping via Multi-Frame and Energy-Offset Correction Acquisition

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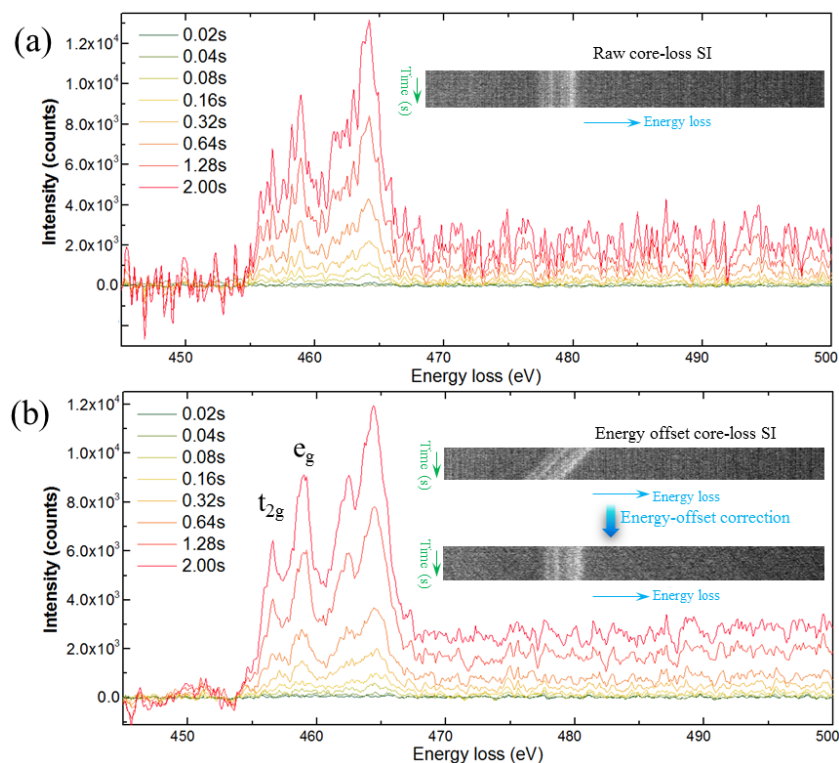
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Besides conventional imaging, an important capability of modern scanning transmission electron microscopy (STEM) is the integration of microanalysis techniques, such as electron energy-loss spectroscopy (EELS) and energy-dispersive X-ray spectroscopy (EDXS). It enables simultaneously probing of both local structural and chemical variations of materials at the atomic scale. In these measurements, a focused electron probe scans the sample and both the spatial and spectral information are acquired simultaneously. Recent advances in instrumentation hardware nowadays have made chemical analysis at atomic resolution readily possible. However, the acquisition and interpretation of atomically resolved spectra can still be problematic due to for instance image distortions or poor signal-to-noise ratio (SNR) spectra, especially for the investigating the energy-loss near-edge fine structures.

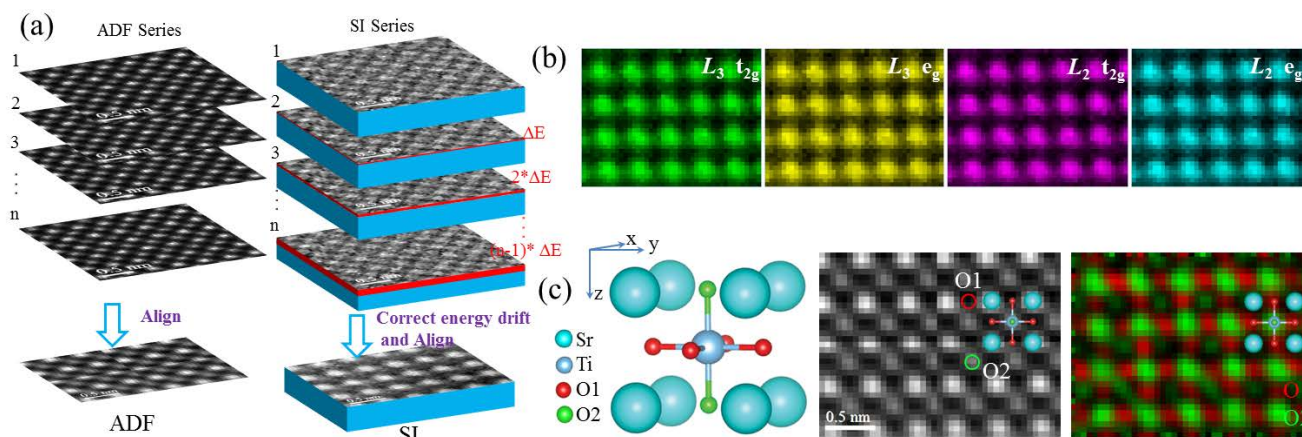
Here, by combining multi-frame spectrum imaging and automatic energy-offset correction, we report a simple, reliable, and step-by-step spectrum imaging (SI) technique for minimizing image distortions and improving the signal-to-noise ratio of the EELS spectra [1]. As widely used in STEM imaging, the fast-scanned multi-frame imaging technique can efficiently remove scan distortions in the ADF and ABF images [2]. The energy-offset correction technique significantly reduces correlated noise [3], as for successive spectra different camera pixels are exposed which precludes amplification of small gain normalization errors (Figure 1). We have implemented these techniques into STEM spectrum imaging for atomically resolved EELS elemental and fine structure mapping. Available methods for realizing the energy-offset on modern Gatan GIF quantum energy filters, and their reliability as well as influences on the final EELS energy resolution are tested and discussed. Using practical examples, we demonstrate that multi-frame SI and post-alignment can effectively suppress image distortions and improve the final elemental map quality. We demonstrate that the energy-offset correction method reduces the correlated noise and helps resolve weak features of the near-edge fine structures. The final SI with improved SNR enables the extraction of individual component maps of the Ti-L<sub>2,3</sub> near-edge fine structure and of a Ti-O-Ti bonding direction map at atomic resolution in SrTiO<sub>3</sub> (Figure 2), which has been theoretically predicted but extremely difficult to detect experimentally due to the poor SNR of the spectrum [4]. Combining multi-frame SI and auto energy-offset-correction we demonstrate that these techniques will open new opportunities for atomically-resolved EELS fine structure mapping. Moreover, this multi-frame SI technique also opens the possibility for managing the electron dose to reduce damaging of the sample [5], as well as the opportunity for atomically resolved imaging and SI at cryogenic temperatures.

### References:

- [1] Y Wang et al, *Ultramicroscopy* **184** (2018), p. 98.
- [2] Y Wang et al, *Ultramicroscopy* **168** (2016), p. 46.
- [3] M Bosman, V J Keast, *Ultramicroscopy* **108** (2008), p. 837.
- [4] M J Neish et al, *Phys. Rev. B* **88** (2013), p.115120.
- [5] L Jones et al, *Microscopy* (2018) <https://doi.org/10.1093/jmicro/dfx125>.



**Figure 1.** Comparison of the Ti- $L_{2,3}$  edge acquired by (a) the conventional method and the energy-offset correction method. The insets show the corresponding raw time series SI data.



**Figure 2.** Fine structure mapping of the Ti- $L_{2,3}$  and O-K edge in  $\text{SrTiO}_3$  via multi-frame and energy-offset correction acquisition. (a) Schematic diagram showing the processing of the multi-frame SI. (b) The individual components map of the Ti- $L_{2,3}$  edges:  $L_3 t_{2g}$ ,  $L_3 e_g$ ,  $L_2 t_{2g}$ , and  $L_2 e_g$ , respectively. (c) Structure of  $\text{SrTiO}_3$ . Viewed along the  $[001]$  zone axis, the O1 atoms have Ti-O bonds directed perpendicular to the  $z$ -axis and the O2 atoms have Ti-O bonds along the  $z$ -axis. Oxygen bonding direction maps extracted from multi-frame EELS SIs obtained by MLLS fitting to the O1 and O2 -K edge spectra.