

Event-based hyperspectral EELS: towards nanosecond temporal resolution

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The acquisition of a hyperspectral image is nowadays a standard technique used in the Scanning Transmission Electron Microscope. It relates the spatial position of the electron probe to the spectral data associated with it [1]. In the case of Electron Energy Loss Spectroscopy (EELS), frame-based hyperspectral acquisition is much slower than the achievable rastering time of the scan unit (SU), which sometimes leads to undesirable effects in the sample, such as electron irradiation damage, that goes unperceived during frame acquisition [2]. In this work, we have developed an event-based hyperspectral EELS by using a Timepix3 application-specific integrated circuit detector with two supplementary time-to-digital (TDC) lines embedded. In such a system, electron events are characterized by their positional and temporal coordinates [3], but TDC events only by temporal ones. By sending reference signals from the SU to the TDC line [4], it is possible to reconstruct the entire spectral image with SU-limited scanning pixel dwell time and thus acquire, with no additional cost, a hyperspectral image at the same rate as that of a single channel detector, such as annular dark-field (Figure 1). To exemplify the possibilities behind event-based hyperspectral EELS, we have studied the decomposition of calcite (CaCO₃) into calcium oxide (CaO) and carbon dioxide (CO₂) under the electron beam irradiation (Figure 2). The present work has been recently published in ref. [5].

References:

[1] R. F. Egerton, *Electron Energy-Loss Spectroscopy in the Electron Microscope*, Springer, 2008.

[2] S. J. Pennycook, *The impact of STEM aberration correction on materials science*, Ultramicroscopy, 2017.

[3] R. Ballabriga et. al., *ASIC developments for radiation imaging applications: The Medipix and Timepix family*, Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment, 2017.

[4] A. Zobelli et. al., *Spatial and spectral dynamics in STEM hyperspectral imaging using random scan patterns*, Ultramicroscopy, 2020.

[5] Y. Auad et. al. *Event-based hyperspectral EELS: towards nanosecond temporal resolution*. arXiv preprint arXiv: 2110.01706. 2021.

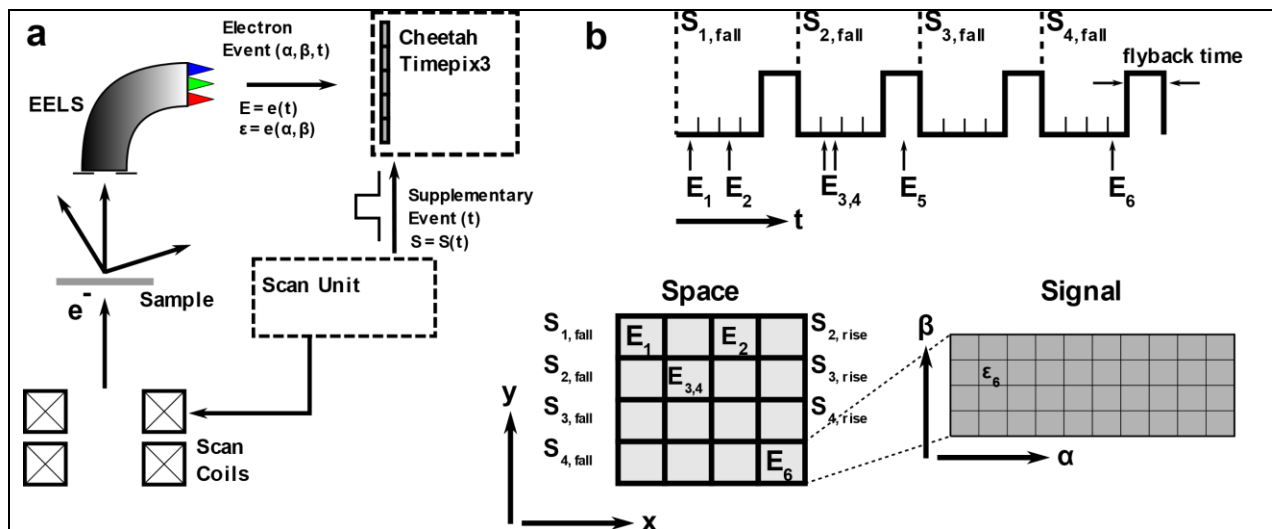


Figure 1: The hyperspectral data reconstruction process. (a) The scheme of the system used for data acquisition. The scan unit inputs temporal supplementary events, while individual electrons produce positional and temporal events. (b) We exemplify how the temporal information of both electrons and supplementary events can be used to arrange electrons in the reconstructed hyperspectral spatial data (x and y). Detector-pixel address information (α and β) is used to determine the spectral information of each spatial pixel.

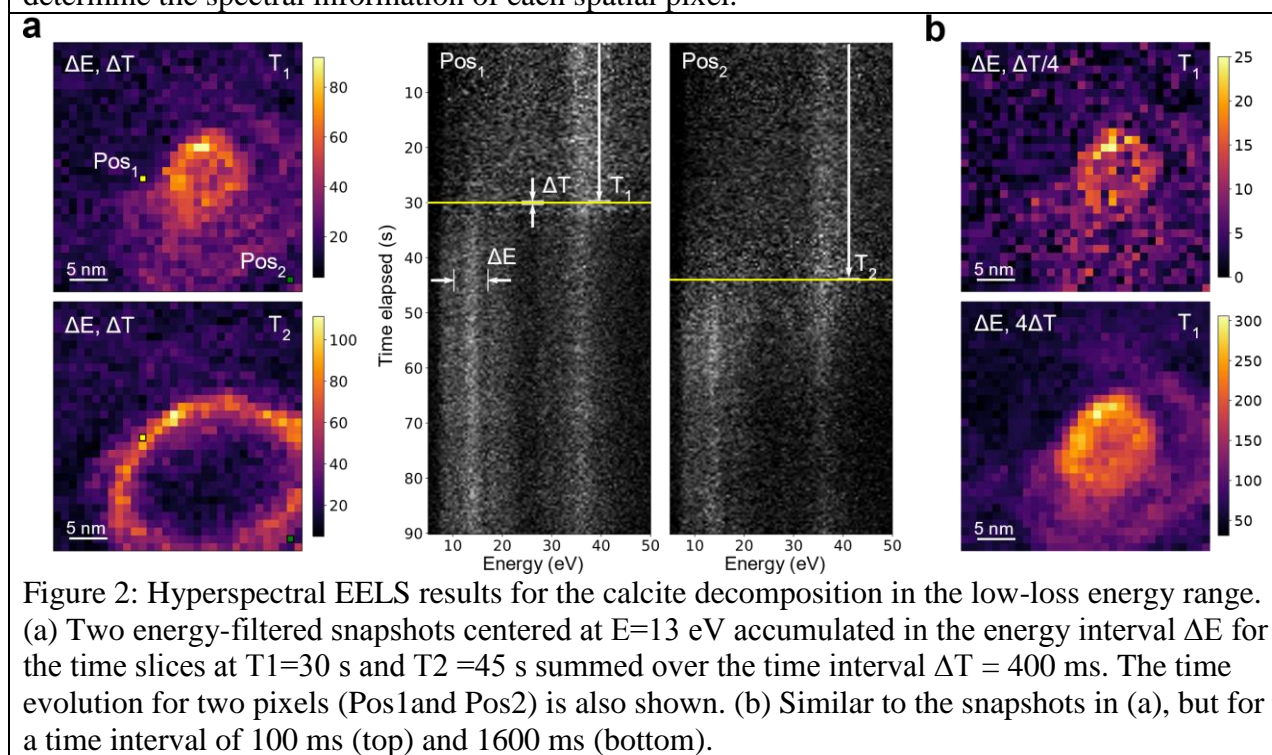


Figure 2: Hyperspectral EELS results for the calcite decomposition in the low-loss energy range. (a) Two energy-filtered snapshots centered at $E=13$ eV accumulated in the energy interval ΔE for the time slices at $T_1=30$ s and $T_2=45$ s summed over the time interval $\Delta T = 400$ ms. The time evolution for two pixels (Pos1 and Pos2) is also shown. (b) Similar to the snapshots in (a), but for a time interval of 100 ms (top) and 1600 ms (bottom).