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doi:10.1017/S1431927622009965
Microsc. Microanal. 28 (Suppl 1), 2022
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Measuring 3D Chemistry with Fused Multi-Modal Electron Tomography

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In principle, electron tomography used in combination with energy dispersive X-ray (EDX) or electron energy loss (EELS) spectroscopy can characterize complex three-dimensional (3D) material chemistry at the nanoscale [1,2]. However in practice, chemical tomography using core excitation spectroscopy demands high electron doses that almost always exceed the specimen limits (e.g. > 10^7 e/Å^2). At best, researchers must choose between measuring 3D structure with high-angle annular dark-field (HAADF) tomography or characterizing chemistry along a single viewing direction. Recent developments in multi-modal data fusion paved new opportunities by substantially reducing the dose requirements for high-resolution chemical imaging [3]. In alignment with the principles of fused multi-modal electron microscopy, here we report extending its algorithmic framework into the third dimension.

Here we introduce fused multi-modal electron tomography, a technique that offers high SNR and high-resolution recovery of material chemistry in 3D by leveraging correlated information encoded within both HAADF and EDX / EELS. We demonstrate fused multi-modal tomography on a simulated composite material system (Fig. 1a) consisting of three elements (Ni, Co and O). The method virtually eliminates Poisson noise in the raw chemical tomograms (Fig. 1c) is efficiently recovers interior nanoparticles in the fused multi-modal reconstruction are efficiently recovered (Fig. 1d). Conventionally spectral maps are collected at every tilt; however we propose a new sampling strategy that efficiently minimizes dose by maintaining an asymmetric ratio (e.g. 1:5) between the number of HAADF and chemical images (Fig. 1d). Although sparse chemical measurements are severely underdetermined in Fourier space (Fig. 1c), fusing the two modalities ultimately fills in missing information (Fig. 1d), improving resolution and reconstruction quality.

We experimentally demonstrate fused multi-modal electron tomography on ZnS/CuS nanocrystals [4]. The radiation sensitivity of the 20 nm nanoparticles makes it challenging to map the distribution ZnS and CuS rich phases in 3D. Despite the noisy EELS maps, the recovered chemical tomograms quality is substantially improved (Fig. 2b,c) over traditional tomography as ~5-10 nm internal cavities and oxidize shells are clearly visible (Fig. 2a). These results demonstrate fused multi-modal tomography can

https://doi.org/10.1017/S1431927622009965 Published online by Cambridge University Press

substantially improve the quality and accuracy of chemical tomograms, even under minimal doses (~10^4 e/$Å^2$).

Fused multi-modal electron tomography opens a pathway to investigate 3D chemistry of nanomaterials by reducing total electron dose while maintaining high fidelity.

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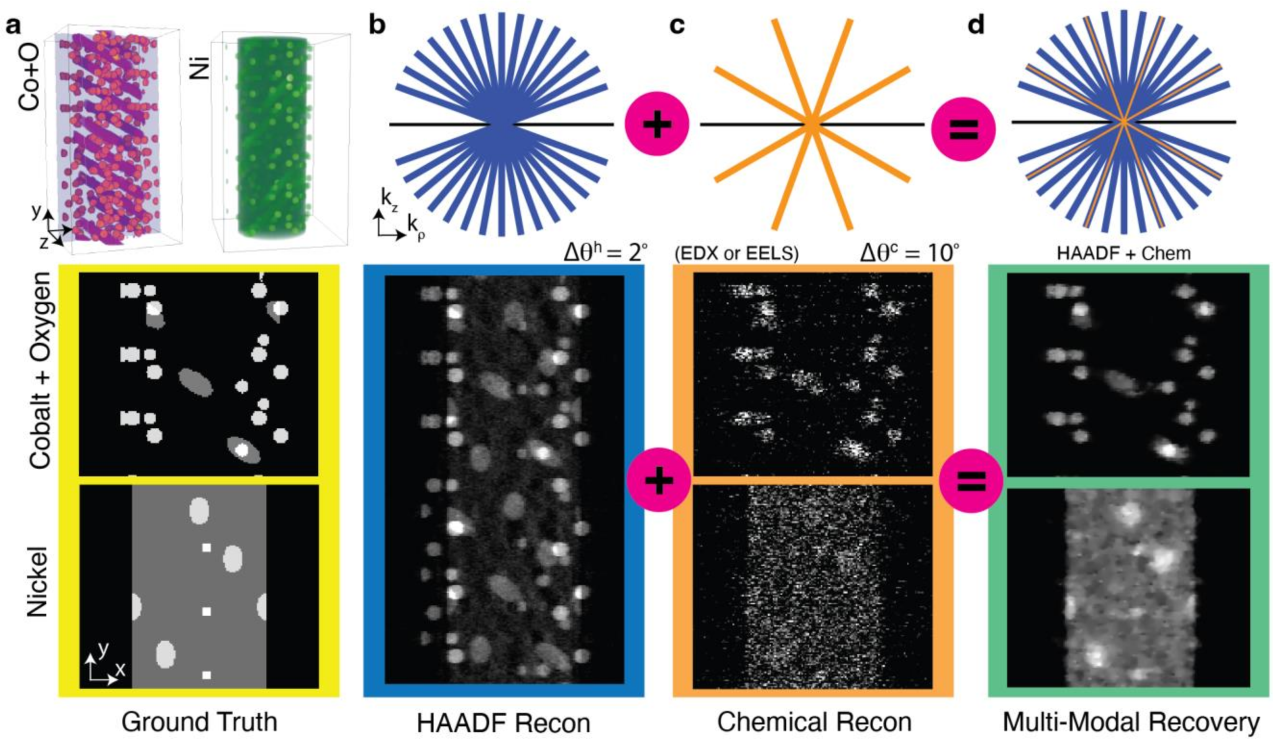

Figure 1: Framework for fused multi-modal electron tomography. a, The simulated Co+O and Ni structures with 2D slices visualized below the tomograms. **b-c**, The HAADF and spectroscopic chemical (Co+O, Ni) reconstructions with their theoretical sampling in Fourier space shown above. **d**, The recovered chemical (Co+O, Ni) tomograms and a visualization of data fusion in Fourier space. The low-frequency information from the HAADF tomogram fills in intermediate missing wedges between adjacent tilts which dramatically improves reconstruction quality.

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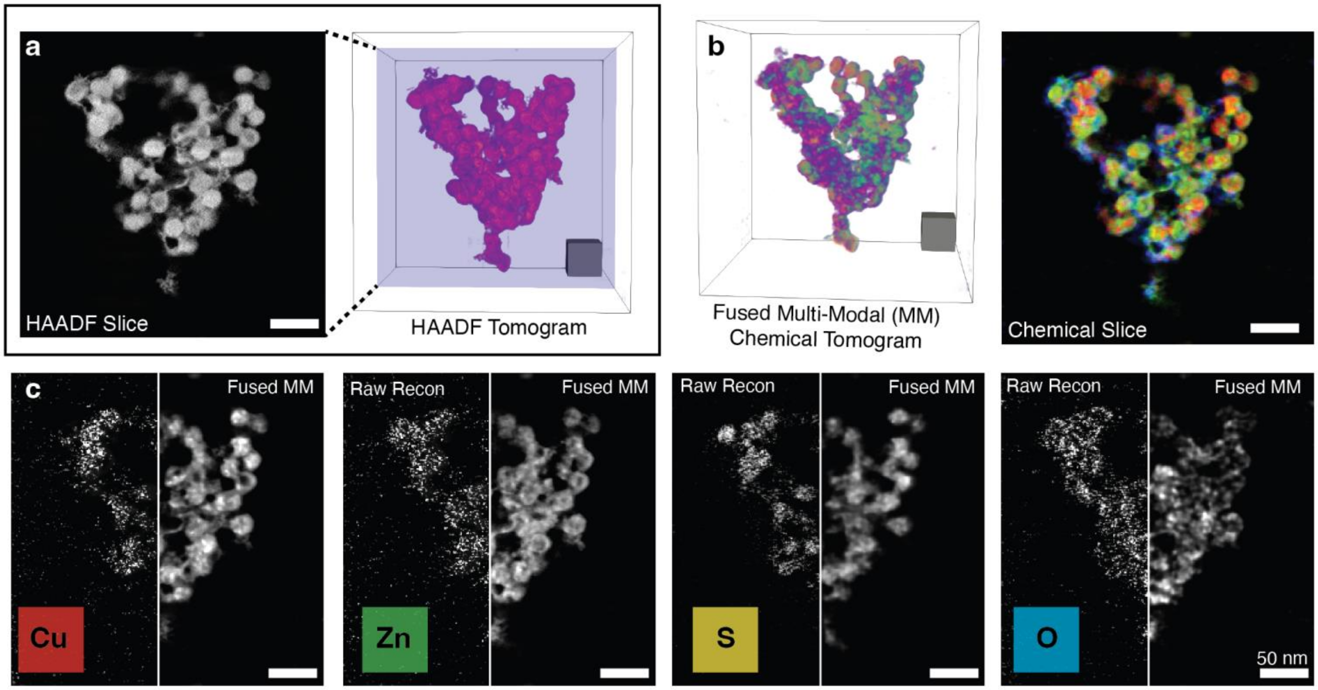

Figure 2: Demonstrating fused multi-modal electron tomography on ZnS/CuS nanoparticles. a, The HAADF reconstruction with a 2D slice shown on the left. **b**, The fused multi-modal reconstruction illustrating CuS or ZnS rich nanoparticles and oxidized shells. **c**, 2D slices of the chemical reconstructions with the noisy traditional reconstructions highlighted on the left of each image.

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