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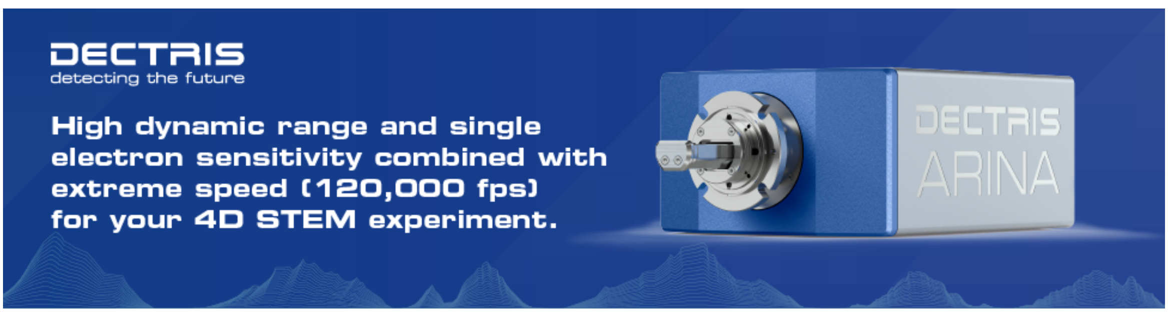

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Measuring 3D Chemistry at 1 nm Resolution with Fused Multi-Modal Electron Tomography

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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). To alleviate the high dose requirement for chemical tomography, researchers commonly sacrifice resolution by increasing the tilt increment (ranging between 5° to 15°) and constraining total dose within a tolerance that specimens can withstand (e.g., < 10^6 e/Å^2). Consequently, 3D resolution is degraded due to under sampling the total number projections for a fixed object size—as predicted by the Crowther criterion [3]. Fortunately, recent developments in multi-modal data fusion for electron microscopy is paving new opportunities by substantially reducing the dose requirements for high resolution chemical imaging [4].

Here we present 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. Data fusion solves an inverse problem that agrees with the chemical and HAADF measurements. Traditional chemical tomography is sparsely sampled in Fourier space (Fig. 1a) resulting in a tomographic reconstruction with artifacts and low SNR (Fig. 1b). Despite the limited sampling, chemical tomography hints at Co core and Mn shell distributions in Co_3O_4-Mn_3O_4 nanoparticles [5]. In contrast, HAADF tomography (Fig. 1c) produces high-resolution and high-SNR reconstructions of specimen structure from many measured projections but lacks chemical specificity (Fig. 1d). Here, fusing the chemical and HAADF modality fills in missing chemical information (Fig. 1e) producing reconstructions where the core-shell chemistry is clearly visible (Fig. 1g) at two-orders of magnitude less electron dose (~10^5 e/A^2) than an equivalent conventional approaches (~10^7 e/A^2).

Sub-nanometer 3D resolution of the chemical distributions is achievable with fused multi-modal electron tomography. We verify nanometer resolution in real space with Au - Fe_3O_4 nanoparticles (Fig. 2a) by measuring the edge sharpness of a single 3 nm Au nanoparticle whose half pitch resolution is visibly less than 1 nm (Fig. 2d). Exploiting shared information in both modalities achieves chemical 3D distributions with resolutions comparable to high-SNR HAADF reconstructions (1.006 nm x 1.032 nm x 1.019 nm).

Fused multi-modal electron tomography opens novel opportunities to investigate the 3D chemistry for a wide range of previously inaccessible nanomaterials.

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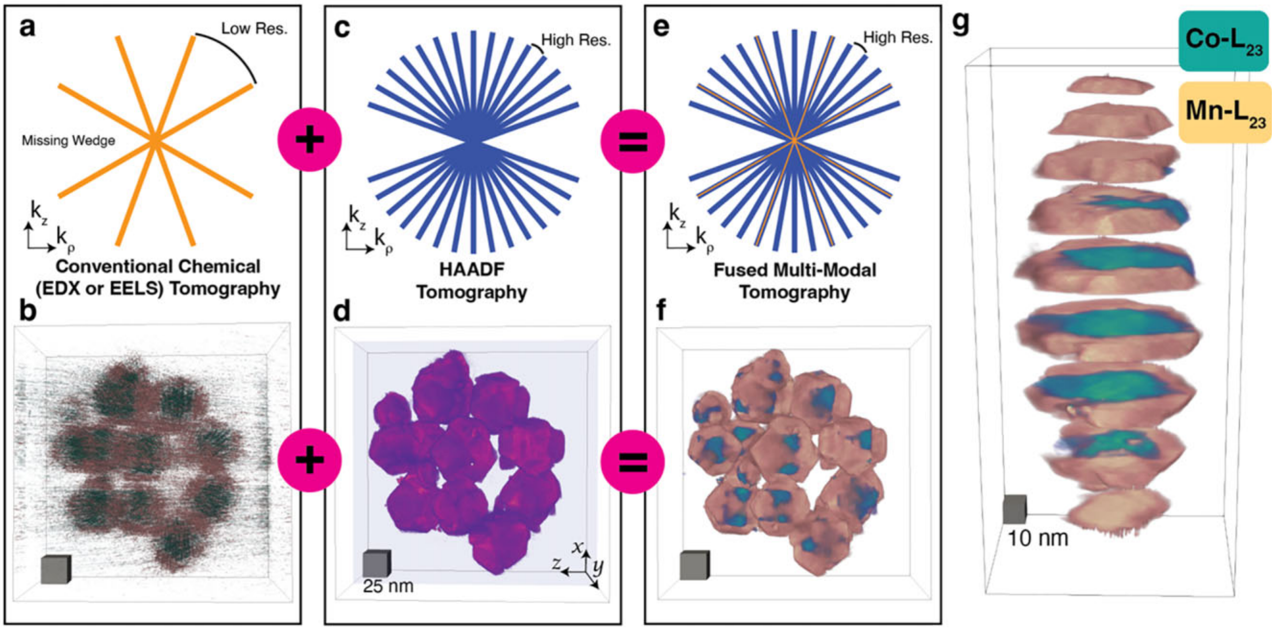

Fig. 1. Fused Multi-Modal Tomography of Core-Shell Co_3O_4-Mn_3O_4 Nanoparticles. **b**, Raw EELS reconstruction for the Co_3O_4 (green) and Mn_3O_4 (yellow) core-loss edges. **d**, The HAADF tomogram of Co_3O_4 - Mn_3O_4 nanoparticle tracks the structure of the specimen but fails to describe materials chemistry in 3D. **f**, The fused multi-modal reconstruction. **a,c,e,** Representation in Fourier space of the projections used to reconstruct the tomograms is shown above each 3D representation. **g**, Fused multi-modal tomogram of a single Co_3O_4-Mn_3O_4 nanoparticle.

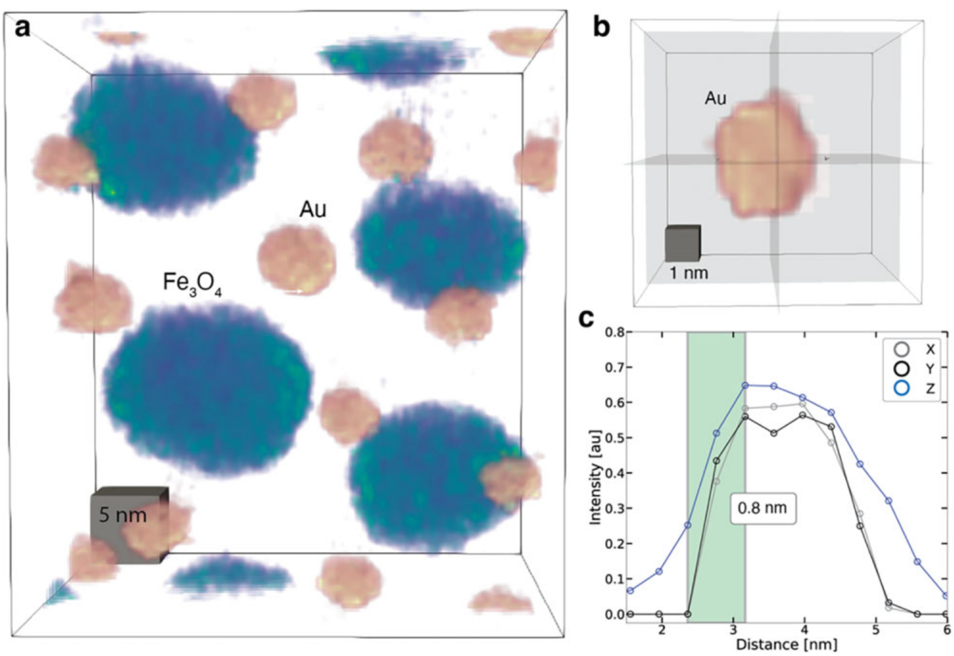

Fig. 2. Measuring 1nm Resolution for Au-Fe_3O_4 Nanoparticles. **a**, Fused EELS tomograms of Au-Fe_3O_4 nanoparticles. **b,c**, Line scan profiles of a 2.5 nm Au nanoparticle gives a resolution of 0.8, 0.8 and 1.2 nm along the x, y, and z directions.

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