

## Mapping Magnetic Ordering With Aberrated Electron Probes in STEM

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Although magnetism originates at the atomic scale, existing spectroscopic techniques sensitive to magnetic signals only produce spectra with spatial resolutions on a larger scale. However, recently it has been theoretically argued that atomic-size electron probes with customized phase distributions can detect electron magnetic circular dichroism (EMCD) [1]. Based on this prediction we have recently shown that deliberately aberrated electron probes in scanning transmission electron microscopy (STEM) can be utilized to obtain chiral dichroic signals in materials via electron energy-loss spectroscopy (EELS) with high spatial resolution [2].

The experiments were performed in an aberration-corrected Nion UltraSTEM™ 100, equipped with a cold field emission electron source and a corrector of third and fifth order aberrations, operating at an accelerating voltage of 100kV [3]. EEL spectra were collected using a Gatan Enfina spectrometer, with 0.3 eV/channel dispersion, giving an energy resolution of 0.9 eV. The convergence semi-angle for the incident probe and the EELS collection semi-angle were 30 mrad and 48 mrad, respectively.

Figure 1 shows two examples of drift (affine)-corrected Z-contrast images and denoised EEL spectra that were acquired simultaneously from the room temperature C-type antiferromagnet LaMnAsO. The data were acquired with the beam along the *c*-axis using a corrected electron probe (Fig. 1a to 1c) and the  $C_{34} = 15 \mu\text{m}$  aberrated probe (Figs. 1d to 1f) [2].

A clear EMCD signature in the EEL spectra, defined as  $(\text{Mn}\uparrow - \text{Mn}\downarrow)$  presenting a change of sign in its integrated intensity between the  $L_3$  and  $L_2$  peaks, is only visible for the Mn L-edge acquired using the  $C_{34}$  aberrated probe. In Figs. 1e and 1f, this signal, shown in green, is positive at the  $L_3$  peak and negative at the  $L_2$  peak. For comparison, the Mn signal in 1b and 1c, shown in grey, does not display this distinctive signature, with both positive and negative components on both peaks.

We will discuss the experimental conditions necessary to reveal the magnetic ordering of individual atomic columns and atomic-size defects in materials [4].

### References:

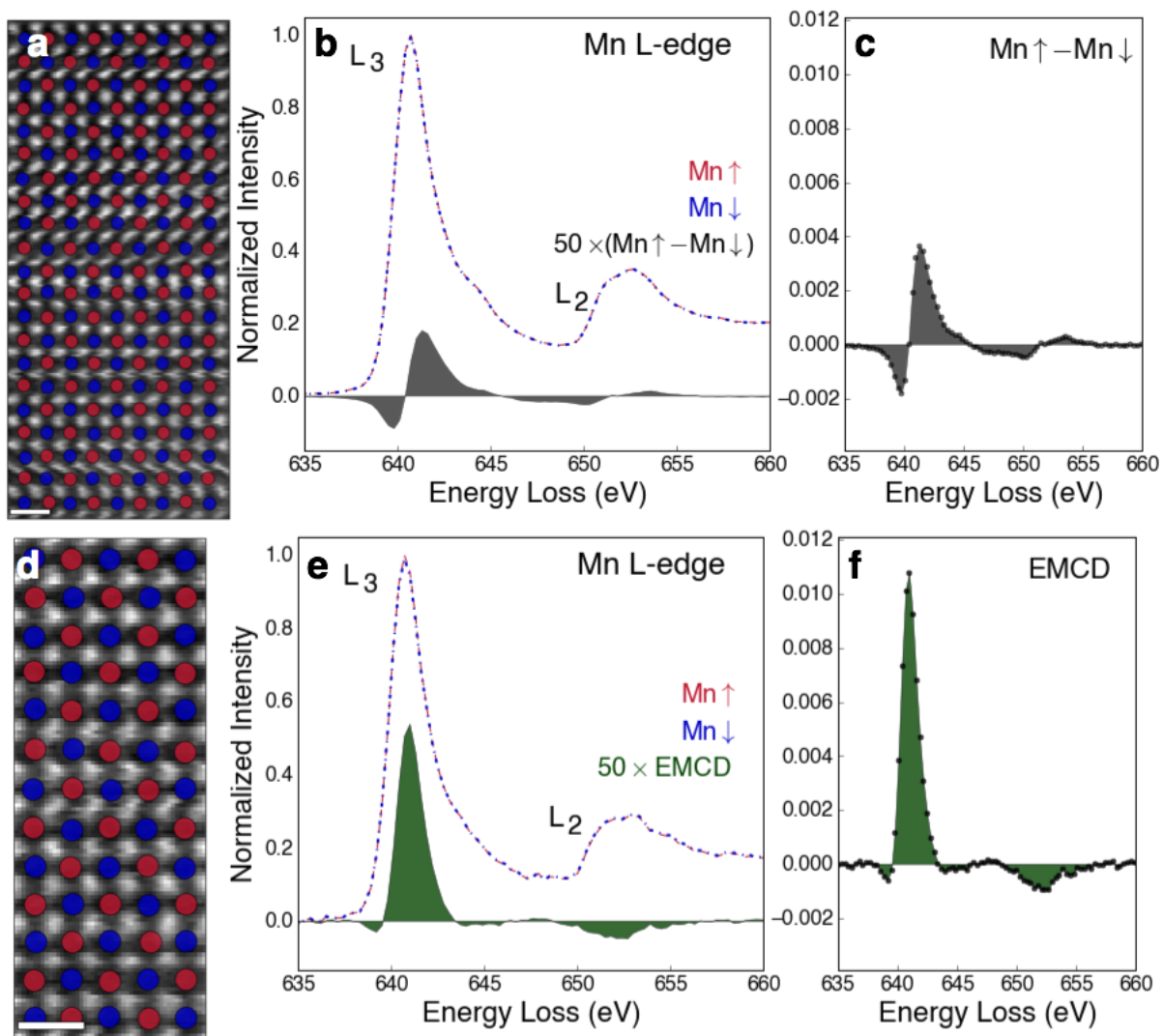
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**Figure 1.** Z-contrast STEM images and Mn L-edge spectra acquired simultaneously for LaMnAsO. (a) and (d) Z-contrast STEM images of LaMnAsO acquired using a corrected and a ( $C_{34} = 15 \mu\text{m}$ ) aberrated electron probe, respectively. The Mn L-edge electron energy-loss (EEL) spectra are shown in (b,c,e,f). The red and blue circles in (a) and (d) schematically highlight the assumed positions of the Mn $\uparrow$ /O and Mn $\downarrow$ /O atomic columns, respectively, that were selected to extract the Mn L-edge EEL spectra. (c) and (f) show the magnified Mn $\uparrow - \text{Mn}\downarrow$  spectra. An EMCD signal (e and f) is only present for the spectra acquired with the  $C_{34}$  aberrated probe. The scale bars are 0.5 nm and the radii for the schematic circles in (a) and (d) are 0.08 nm. Figure adapted from Ref. 2.