

## Distortion Correction in Scanning Transmission Electron Microscopy with Controllable Scanning Pathways

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Quantitative intensity and distance measurements in atomically resolved scanning transmission electron microscopy (STEM) imaging relies on the precision of the intensity and position of the images corresponding to the scanned sample area. Conventional STEM rastering modes are limited by drift distortion in the slow scan direction. It has been realized that the scan direction is a parameter that can be tuned to enhance STEM image quality. For example, the revolving STEM (RevSTEM) method utilizes sequential imaging at varying scan directions to effectively eliminate drift distortion from atomic resolution images [1]; however, the scan pattern within each frame is still limited by conventional STEM rastering modes and drift. By controlling the scanning trajectories in an aberration-corrected STEM, Jesse et al. have shown that it is possible to precisely position the probe along a predefined path [2]. An array of intensity data at frequencies up to 2 MHz can be simultaneously acquired and then reconstructed to form an interpretable 2D STEM image. This so-called general-scan STEM (G-STEM) approach opens up new possibilities for STEM imaging and quantitative analysis. For example, using a spiral-scanning path, fly-back can be avoided to save frame acquisition time for fast imaging of beam sensitive materials. This also enables the possibility for compressed sensing with an optimized scan path. However, the G-STEM images show complicated image distortions depending on the scan path that are not fully understood. Figure 1a shows a typical G-STEM spiral-scan path that was selected for equal sampling over the sequential turns of the spiral. Figures 1b and 1c show typical G-STEM annular dark field (ADF)-STEM images acquired from an outward and inward spiral scan across an interface between strontium titanate (STO) and lanthanum strontium manganite (LSMO) acquired at 100kV using a Nion UltraSTEM. The obvious distortion in the center is attributed to hysteresis and phase-lag of the scan system electronics and high velocity within the center during the transition from inward-to-outward spiraling.

Here, we propose a distortion correction algorithm that can be applied for any scanning trajectories along a predefined curve with an array of x-y coordinates  $(x_i, y_i)$ . Although the curve may fit some analytical functions, the actual movement of the STEM probe follows discrete points. The velocity  $\mathbf{v}(x_i, y_i)$  of the probe at point  $(x_i, y_i)$  is calculated as  $(x_i - x_{i-1}, y_i - y_{i-1}) / \Delta t$ , where  $\Delta t$  is inversion of read-out frequency, which is analogous to pixel dwell time in conventional STEM rastering mode. The acceleration  $\mathbf{a}(x_i, y_i)$  is defined as  $(\mathbf{v}(x_i, y_i) - \mathbf{v}(x_{i-1}, y_{i-1})) / \Delta t$ . The displacement of each probe position from the ideal position can then be compensated as a linear combination of velocity and acceleration using:

$$(dx_i, dy_i) = A\mathbf{v}(x_i, y_i) + B\mathbf{a}(x_i, y_i) \quad (1)$$

where A and B are coefficients that can be refined from experimental STEM images. Figures 1d and 1e show the corrected STEM image (with  $A = -17\Delta t$  and  $B = 0$ ) where the distortion is essentially removed. To aid in the calibration of scan electronics, a serpentine raster scan was generated such that the scan

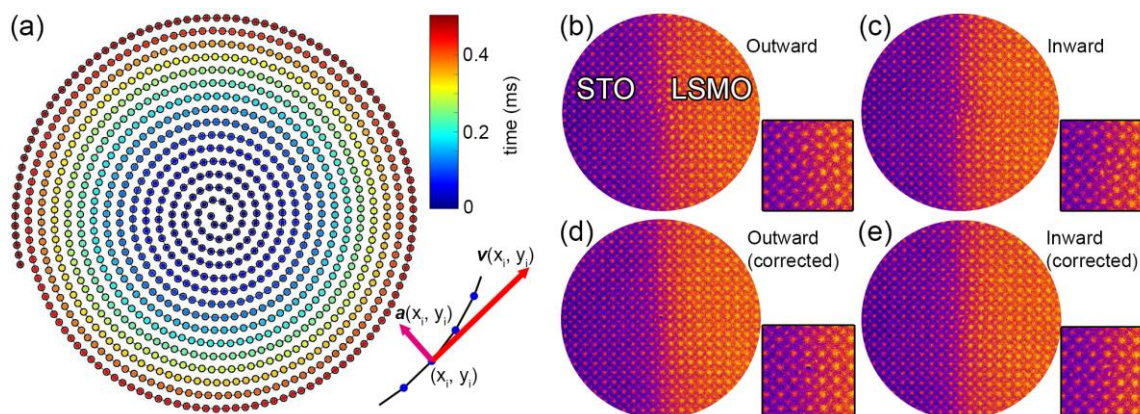
frequency is increased as the beam moves along the slow scan axis. Therefore, each line in the resulting image gives us information about scan distortion as a function of scan velocity. The frequency increases from 20 Hz to 10,000 Hz from top-to-bottom. The reconstructed forward-scanning image and the backward-scanning image are shown in Figures 2a and 2b respectively. Although atomic-columns are still distinguishable, the distortion clearly worsens as the frequency increases. Using the same A and B from the spiral-scan in Figure 1, distortion-corrected forward and backward images are shown in Figures 2c and 2d, respectively. Both images are correctly restored up to around 5000Hz, beyond which the sampling is too low for the scanned periodic structure. In addition to the scan electronics dynamic-based correction, we performed non-linear distortion corrections that only operate on the resulting images without considering the scan path. Future work involves using a more rigorous column indexing approach to quantify the residual higher order distortion [3].

#### References:

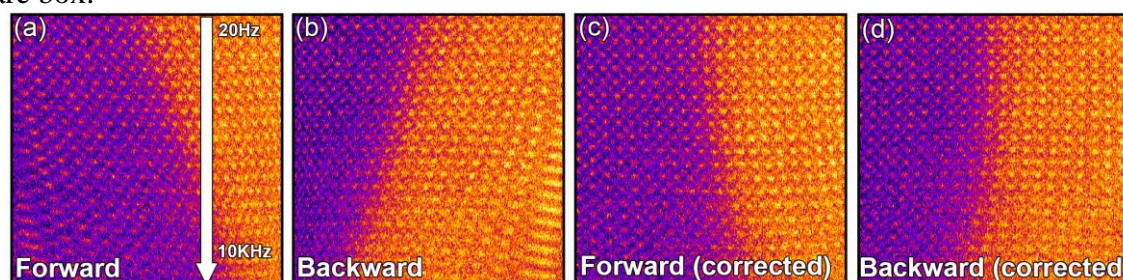
[1] X. Sang and J. M. LeBeau, *Ultramicroscopy* **138** (2014), p. 28.

[2] S. Jesse et al., *Small* **44** (2015), p. 5895.

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**Figure 1.** (a) A spiral scan that can scan both inward and outward. Reconstructed outward spiral image (b) and inward spiral image (c) acquired from a STO/LSMO interface along [001]. (d) and (e) are distortion corrected images of (a) and (b), respectively. The enlarged center of each image is shown in the square box.



**Figure 2.** Reconstructed forward (a) and backward (b) scans as a function of scan frequency for image calibration. The distortion corrected images are shown in (c) and (d).