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A Hybrid Approach to Calibrate the Affine Transformation Between Scan-Camera Coordinates for 4D-STEM Datasets

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Accurate geometrical calibration between scan-camera coordinates is critical in four-dimensional scanning transmission electron microscopy (4D-STEM) [1] for both quantitative imaging and ptychographic reconstructions. Such calibration is, however, not a trivial task since (*i*) 4D-STEM is more sensitive to the vortical nature of electron trajectories caused by the electromagnetic lenses compared to other STEM imaging modes that use detectors with circular symmetries; (*ii*) Most 4D-STEM experiments still need relatively long dwell time (*e.g.*, > 100μs per pixel) making them highly susceptible to instrumental instabilities and environmental disturbances.

We argue that existing calibration methods are not completely satisfactory. For instance, imaging registration methods [2-7] that require multiple 4D-STEM frames could still be practically challenging for most instruments. A popular method by J. Hatchel *et al.*, [8,9] which utilizes the electric-field information from the 4D-STEM dataset, did not address possible shearing and scaling of the scanning positions. Iterative methods like the serial cross-correlation method [10], annealing algorithm [11], nonlinear optimization [12], evolutionary refinement [13], or direct gradient of intensity patterns [14], may not work well if the initial scan-position error is large. The robustness of existing methods against factors such as residual aberrations is also not well understood.

In this presentation, we will introduce a *hybrid method* (**Figure 1**) based on two sub-routines, namely the *J-matrix method* and *the Fourier method*, and it can calibrate the uniform affine transformation between the scan-camera coordinates using raw data, without a priori knowledge about the crystal structure of the specimen. The *hybrid method* is found robust against scan distortions and residual probe aberrations. It is also effective even when defects are present in the specimen, or the specimen becomes relatively thick. We will demonstrate that a successful geometrical calibration with the *hybrid method* will lead to a more reliable recovery of both the specimen and the electron probe in a ptychographic reconstruction. We will also show that, although the elimination of local scan position errors still requires an iterative approach, the rate of convergence can be improved, and the residual errors can be further reduced if the *hybrid method* can be firstly applied for initial calibration. The code is made available as a simple-to-use tool to correct affine transformations of the scan-camera coordinates in 4D-STEM experiments.

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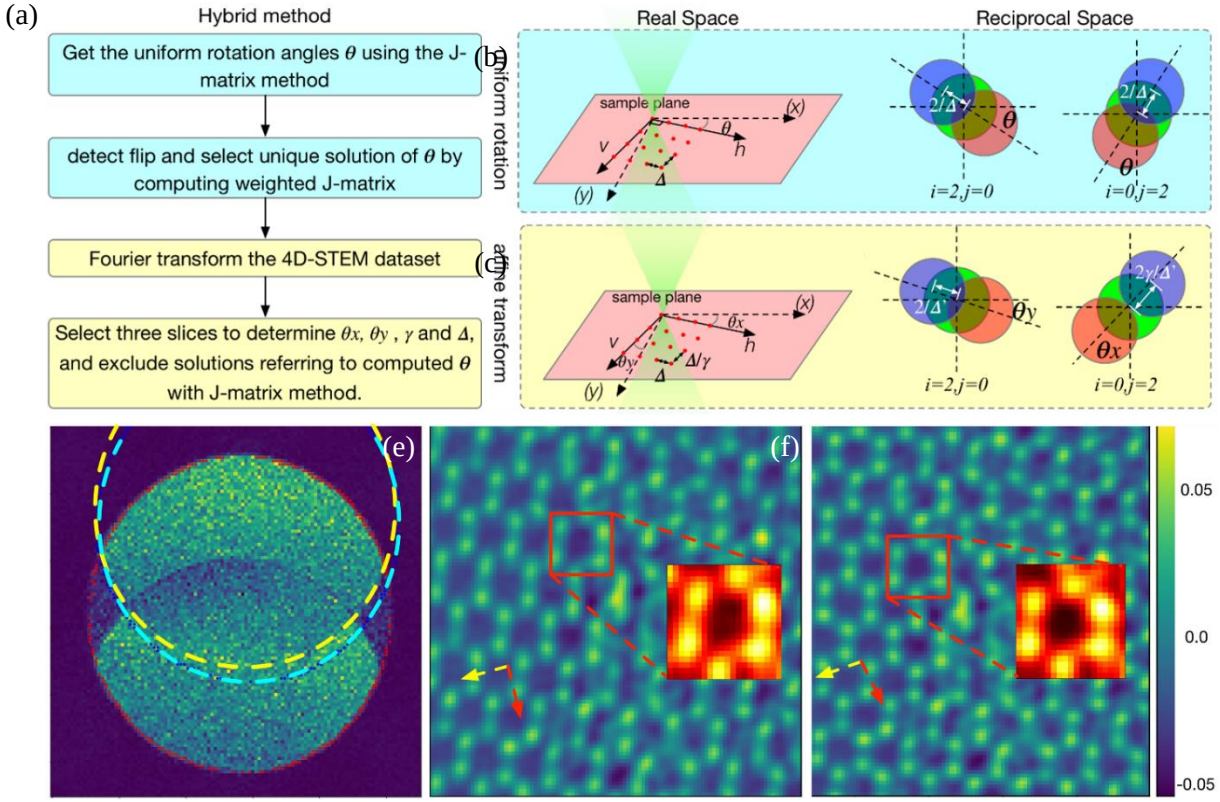

Figure 1. (a) The workflow of the *Hybrid method*. The *J-matrix method* will be used to first calibrate an uniform rotation angle θ between the scan-camera coordinates as shown in (b). The *Fourier method* will be applied next to calibrate the full affine transformation, separately considering the different rotation angles and scanning step sizes along the fast and slow scanning directions as shown in (c). (d) The difference between the first-order disk positions in a G-sets slice in an experimental 4D-STEM data on monolayer MoS_2, obtained from the geometrical parameters determined by the *J-matrix method* (cyan ring) and by the *hybrid method* (yellow ring). The corresponding reconstructed phase distribution using the single-side-band method with the geometrical calibration parameters determined by the *J-matrix method* and by the *hybrid method* are shown in (e) and (f), respectively. The MoS_2 lattice is clearly better reconstructed in the case of (f).

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