High Precision STEM Imaging by Non-Rigid Alignment and Averaging of a Series of Short Exposures

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Precision in both high-resolution TEM and STEM imaging is fundamentally limited by signal to noise, but STEM encounters practical limits before the fundamental limit is reached. Because of the serial acquisition of the image, instabilities in the position of the probe or the sample introduce random and systematic errors in the positions of the atomic columns. As a result, 1 pm precision has been reported in TEM [1], but the best reported precision in STEM is 5 pm [2].

Instabilities that are fast compared to frame time and have zero mean can be removed by averaging over many frames. However, averaging with only a rigid translation between frames to account for sample drift often causes a loss of resolution and therefore does not reach the highest possible precision. We have developed a non-rigid registration scheme for series of STEM images. The registration of an image pair, R and T, is the task of transforming the two images into a common coordinate system. Taking R as reference image, the goal is to find a deformation ϕ of the image domain such that the composition of T and ϕ, $T \circ \phi$, agrees with R. Here we use a pixelwise, non-parametric mapping ϕ to model the deformation.

To find this deformation we employ a general variational framework. The objective functional is defined by a distance measure between $T \circ \phi$ and R, plus a penalty regularizer on the deformation ϕ which favors smooth ϕ. In order to cope with low signal-to-noise ratio images we use the negative of the normalized cross correlation (*i.e.* the normalized covariance) of $T \circ \phi$ and R as the distance measure. As the penalty term we use the integrated squared deviation of the Jacobian of ϕ from the identity matrix, which is called "Dirichlet energy" of the displacement.

To numerically solve the minimization problem we employ a multilevel approach from a coarse downsampling of the original images to the single-pixel level. At each level of resolution we use a regularized gradient descent based on the H^1 scalar product [4] with explicit time discretization and step size control [5].

Finally, the registration framework connecting two images is extended to handle the registration of hundreds of consecutive images back to a single reference image. Due to the extended acquisition time of such long series, the frames typically exhibit large scale drift plus local deformations, which requires special care. Our iterative process adjusts the transforms of all images $T_j \circ \phi_j$ to R in an attempt to minimize the combined penalty of all ϕ_j together.

Figure 1 shows the results of non-rigid averaging of a series of frames of Si [112]. The images were acquired on the a probe-corrected FEI Titan at 200 kV with a 24.5 mrad convergence angle, a 25 pA

probe, and collection angles of 54 to 270 mrad. The series consists of 512, 256×256 pixel images at a dwell time of 12.8 μs per pixel out of which 325 frames images were processed. Figure 1(a) is the first frame out of the series. Figure 1(b) is the average after non-rigid registration to the first frame. No other smoothing or processing has been applied to Figure 1(b). The effective dwell time per pixel in Figure 1(b) is 4.16 ms, but there is no loss of spatial resolution or spatial sampling. Close examination shows that there is a small variation in the distance between two horizontal rows of dumb bells in Figure 1(a) which has averaged out in Figure 1(b).

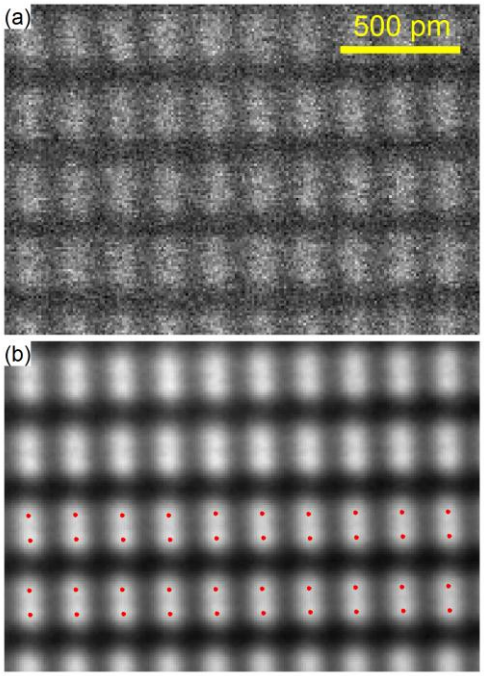

Figure 1: (a) The first of 512 frames of Si [112]; (b) Average of 325 frames registered to the first frame. No other smoothing or other processing has been done. The red dots are the positions of the columns identified by fitting.

Following Bals [3], we fit each of the Si [112] dumb bells to a sum of two Gaussian peaks to estimate the atom column positions, then calculate the interatomic distances. The precision of the positions is the standard deviation of the measured interatomic distances. The peak centers and distances are shown in Figure 1(b). By this measure, the precision in the 36 measured {220} distances is 0.8 pm, and the precision in the 20 measured {111} distances is 1.6 pm. The reason for the discrepancy is that the {220} distance is almost aligned with the scan direction, and the {111} distance is almost perpendicular.

Each pixel in the averaged image has an effective dwell time of 4.16 ms, and each dumb bell has an integrated scattered signal of 3.8×10^6 electrons. The large number of detected electrons creates the necessary signal to noise for high precision position determination and it means that the Poisson noise is only 0.05% of the signal, making very small changes in intensity from, *e.g.*, point defects detectable. Additional examples from compound materials and application to surface distortions on nanostructures will also be discussed [6].

References:

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