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Atomic Resolution Convergent Beam Electron Diffraction Analysis Using Convolutional Neural Networks

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The combination of a fast direct electron camera and a modern scanning transmission electron microscopy (STEM) makes it possible to collect convergent electron beam diffraction (CBED) patterns at kHz frequency and sub-Å resolution in a technique called 4D STEM. One effective means to analyze such CBED data sets is by comparison to multislice simulations. That comparison is typically performed either by human eyes or using least square fitting (LSF) to match experimental patterns to a library of pre-computed simulations. Human vision is an excellent pattern discriminator but limited in availability and high cost. LSF requires full fitting of all experimental parameters, including probe and projector lens distortions, non-uniformity in illumination and aperture shapes, *etc.* Convolutional neural networks (CNNs), which have had huge success in computer vision, have substantial potential for automated, high speed analysis of CBED patterns, as recently shown by Xu and LeBeau for unit-cell average patterns [1]. Here we present a way to extract information from atomic resolution CBED patterns using one single CNN trained from simulated CBED pattern library labeled with the property of interest. $SrTiO_3$ was used as target material and atomic resolution thickness was predicted.

Experimental CBED patterns were collected on an aberration-corrected Titan STEM using a DE-16 direct electron camera. 512 x 512 pixel CBED patterns were acquired at 1100 frames per second (FPS) speed with camera acquisition was synchronized to STEM scan using a 24.5 mrad convergence half angle probe at 22 pA current. High angle annualr dark-field (HAADF) images were collected simultaneously with CBED patterns on a Fishione 3000 detector to provide an independent, column-by-column measurement of the local thickness from quantitative STEM [2].

CNNs of the VGG16 [3] architecture were trained using a simulated library generated by GPU accelerated multislice simulations [4], convolved with a 90 pm Gaussian kernel to account for spatial incoherence, then augmented by shift, flips, translations, rotations, shears, zooms, and image noise**.** The augmentation is important as it prevents overfitting of the CNN to the training dataset and help th CNN generalize its fitting capability. This also makes the trained CNN less sensitive to variations in the parameters which bedevil LSF. One CNN was trained for coarse thickness prediction in 2 nm increments up to 100 nm thickness, and one for fine prediction in 1 unit cell (0.4 nm) increments up to 35 nm. Training of coarse CNN ended with 80% prediction accuracy of synthetic data. The fine CNN ended with 60% accuracy.

Figure 1 shows examples thickness predictions. The top rows in Fig 1(a) and (c) are experimental CBED patterns averaged over a single Sr column from the experiment. The bottom rows show the simulated CEBDs matched by the CNN. The CNNs effectively match features in CBEDs to similar features in the simulation library. Figure 2 shows the thickness measured for many column by HAADF vs CBED / CNN as a function of the radius of integration for the CBED about the atomic columns. For the coarse CNN, about half of CNN predictions fall within ±2 nm of the HAADF thickness for all integration radii . For the fine CNN, half the predictions are within ±1 nm, except for the smallest radius (18 pm). Once trained, CNNs are fast to execute, providing near real-time thickness mapping for 4D STEM data sets [5].

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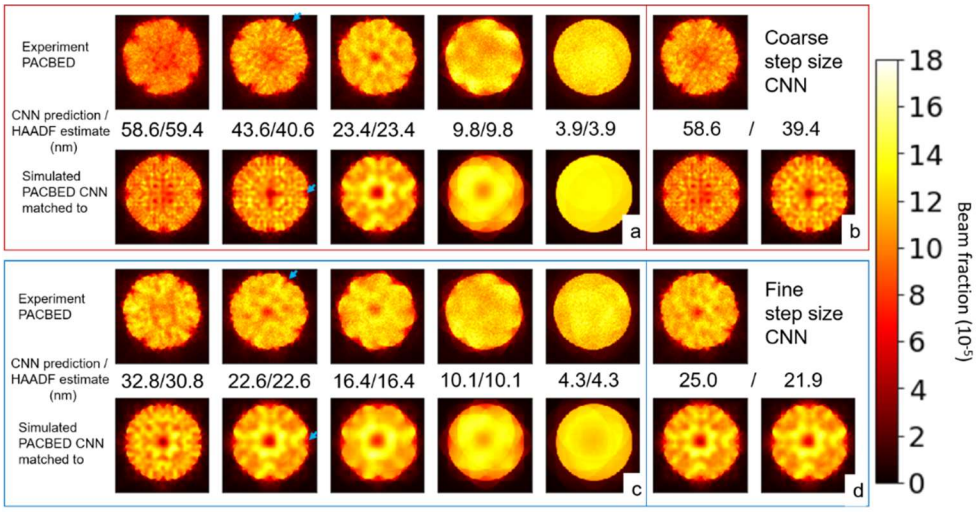

Figure 1. Examples of matches between experimental Sr-column CBED and simulated Sr-column CBED from CNN predicted thickness for a) coarse step size CNN and b) fine step size CNN. The sample is [100] oriented $SrTiO_3$.

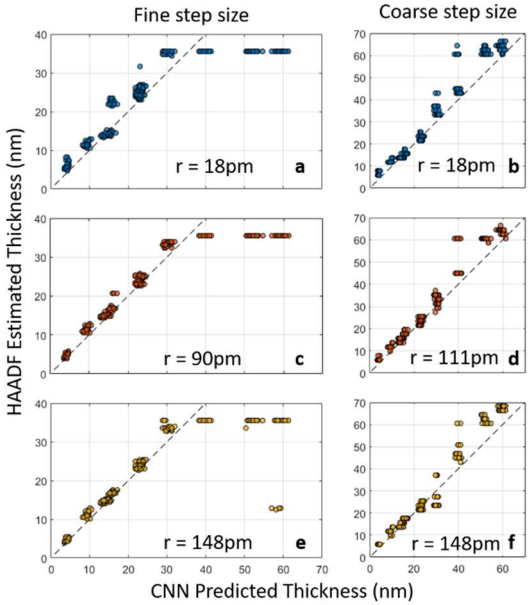

Figure 2. CNN with coarse and fine step size prediction compared to quantitative STEM measurement of thickness from the same atomic columns. *r* is the integration radius about the atomic column position in the CBED data set.

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