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Dose-Efficient Defect Contrast with 4D-STEM

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Defects in materials play a crucial role in determining their properties [1-4]. Because these features often break local order, understanding symmetry is critical for determining structure-property relationships. Scanning transmission electron microscopy (S/TEM) is an especially apt tool to investigate these structural features, as its atomic to nanoscale resolution is on the same length scale as many important defects.

With S/TEM, the symmetry of a material can be studied in both real and reciprocal space. The versatility of 4D-STEM (Fig. 1A), a STEM experiment where a 2D diffraction pattern is recorded at each position in real space, makes it a particularly valuable method to study subtle changes in local order. Small perturbations in diffraction space can be probed with a variety of reconstruction methods that lead to high resolution real space maps of material properties [5, 6].

Using PRISM multislice simulations and a library of graphene structures [7, 8], we demonstrate how the application of virtual detectors can be used to efficiently enhance signals from defects. Graphene is a quintessential example of a weak phase object, meaning high doses are needed to get sufficient signal to noise in STEM for atomic resolution imaging. Substitutions and vacancies in graphene can strongly impact its properties, so we use this material as a case study to look for more efficient tools to map point defects [9].

Considering a monolayer of graphene (Fig. 1B) and using prior knowledge of its space group and diffraction space symmetry (Fig. 1C), we designed a set of virtual detectors with six-fold symmetry aligned to the bright field disk (Fig. 1D-E). When the purple and green regions are subtracted, the resulting image reflects the local order of the material. Moreover, as the detector is rotated, the image contrast changes from illuminating the coherent crystal lattice to highlighting the defects (Fig. 1F-I).

The dose can be further reduced through the incorporation of a phase plate in the probe forming aperture of a STEM (Fig. 2A). Phase plates have been shown to efficiently transfer information about light elements across a wide range of spatial frequencies [10]. A traditional ringed plate (Fig. 2B) can be used to enhance the signal from graphene (Fig. 2E&H). Alternatively, when rotated a 3-fold plate (Fig. 2C-D) can enhance either atomic (Fig. 2F&I) or defect (Fig. 2G&J) contrast in a more dose efficient manner.

In this presentation, we will show how a combination of phase plates and virtual detectors can be used to enhance contrast from important symmetry breaking defects. Moreover, we will explore how these effects depend on acquisition parameters and can be extended to more complex material systems [11].

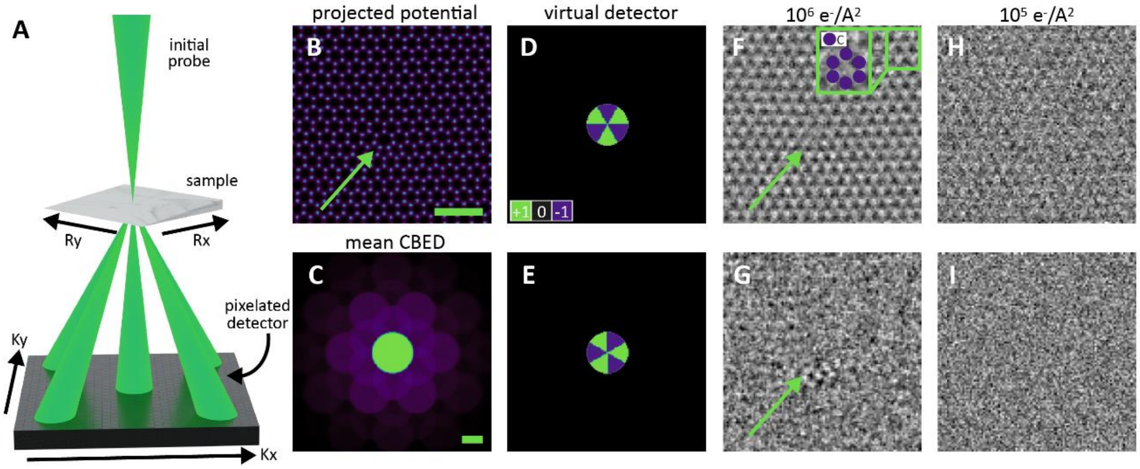

Figure 1. (A) 4D-STEM approach (B) Projected potential of graphene with a defect as indicated by arrow. (C) Mean converged beam electron diffraction pattern from simulated dataset. (D-E) virtual symmetry detectors for F-H and G-I respectively with inset legend. Detectors create (F-I) images that highlights either the coherent lattice or defect. Scale bar 1 nm or 15 mrad.

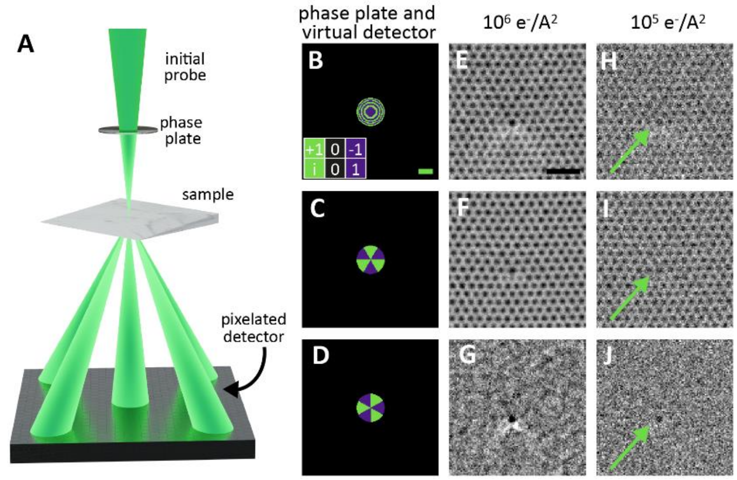

Figure 2. (A) When a phase plate is inserted in the probe forming aperture of the STEM, the defect contrast is enhanced. (B-D) Phase plate and matching virtual detectors with inset legend. (E-J) Depending on plate shape and orientation either the atomic or defect contrast can be enhanced. Scale bar 1 nm or 15 mrad.

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