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Mapping pm-scale Lattice Distortions and Measuring Interlayer Separations in Stacked 2D Materials by Interferometric 4D-STEM

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Two-dimensional (2D) materials possess a host of exotic properties, which can further be modified by stacking them into heterostructures. Since interactions between layers can alter their structural, electronic, and magnetic properties, these can be controlled for example through twist angles or stacking sequences [1]. A noteworthy example is bilayer graphene, which undergoes in-plane structural distortions and a corresponding restructuring of its electronic properties at low twist angles [2] that can result in correlated insulating states [3] and unconventional superconductivity [4]. The ability to map out structural distortions and their variations across materials with a range of twist angles and stacking sequences is therefore important for understanding and ultimately controlling both structural and electronic/magnetic properties of these materials. As a result, various transmission electron microscopy (TEM) techniques such as diffraction [5], dark-field and scanning TEM (STEM) imaging [6-8], and four-dimensional STEM (4D-STEM) [9-11] have recently been developed to provide access to this type of structural information.

Here, we describe an interferometric 4D-STEM approach for mapping picometer-scale structural reconstructions with nanometer resolution and measuring interlayer separations of few-layer materials [11]. By using a defocused STEM probe, diffracted beams focus in the same plane as the transmitted beam, perpendicular to the beam direction, and propagate to the far field. Diffracted beams that originate from separate layers in the material then interfere in reciprocal space where their Bragg disks overlap (Fig. 1). The positions of the resulting interference fringes are a function of the relative positions of the atoms in the layers under the probe, allowing information about structure to be extracted from these fringes. In the case of twisted bilayer graphene, this results in a linear shift of the fringe positions as a function of distance across the sample (Fig. 2a-c) plus small deviations away from this linear shift that occur due to structural distortions in the material. By mapping the phase of the interference fringes at every probe position in a 2D array and removing the linear component, the remaining pm-scale lattice distortions can be visualized with nm-scale spatial resolution (Fig. 2d-f). In addition, information about the separation of the layers can be extracted as well, since the rotation of the fringes carries information related to the positions of the interfering layers in the beam propagation direction.

Interferometric 4D-STEM therefore provides the unique ability to map three-dimensional information about the relative positions of atoms in separate layers of stacked 2D materials. This technique will therefore provide insights into the interplay between the structural and electronic/magnetic properties of stacked 2D materials, aiding in synthesis of heterostructures with novel properties [12].

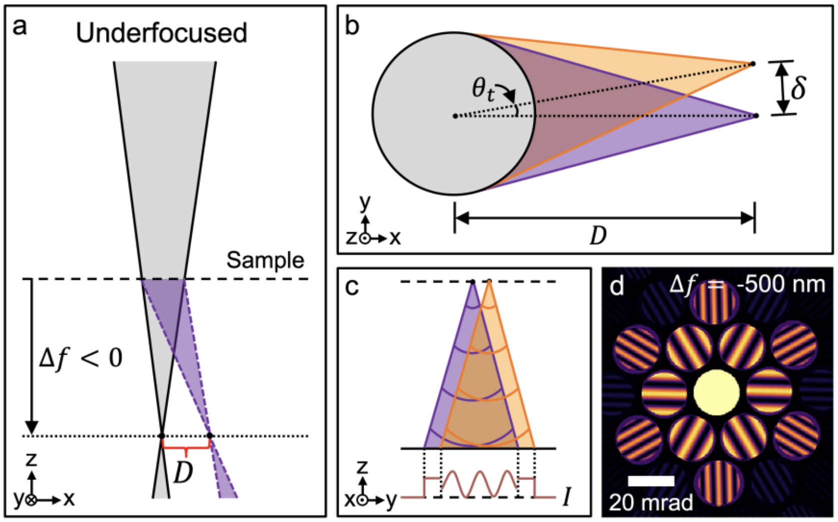

Figure 1. Schematic of reciprocal space fringe formation for interferometric 4D-STEM. (a) With a defocused probe, diffracted beams from each layer of a material focus in a plane perpendicular to the beam direction that contains the transmitted beam. (b) Diffracted beams from different layers of the material can be displaced from each other in this plane due to properties such as a relative twist between the layers. (c) These beams propagate to the far-field and interfere with each other where the corresponding Bragg disks overlap. (d) This interference is detected in diffraction patterns (simulated here) acquired at every probe position in a two-dimensional array across the sample. Figure reproduced, condensed, and recolored with permission from [11].

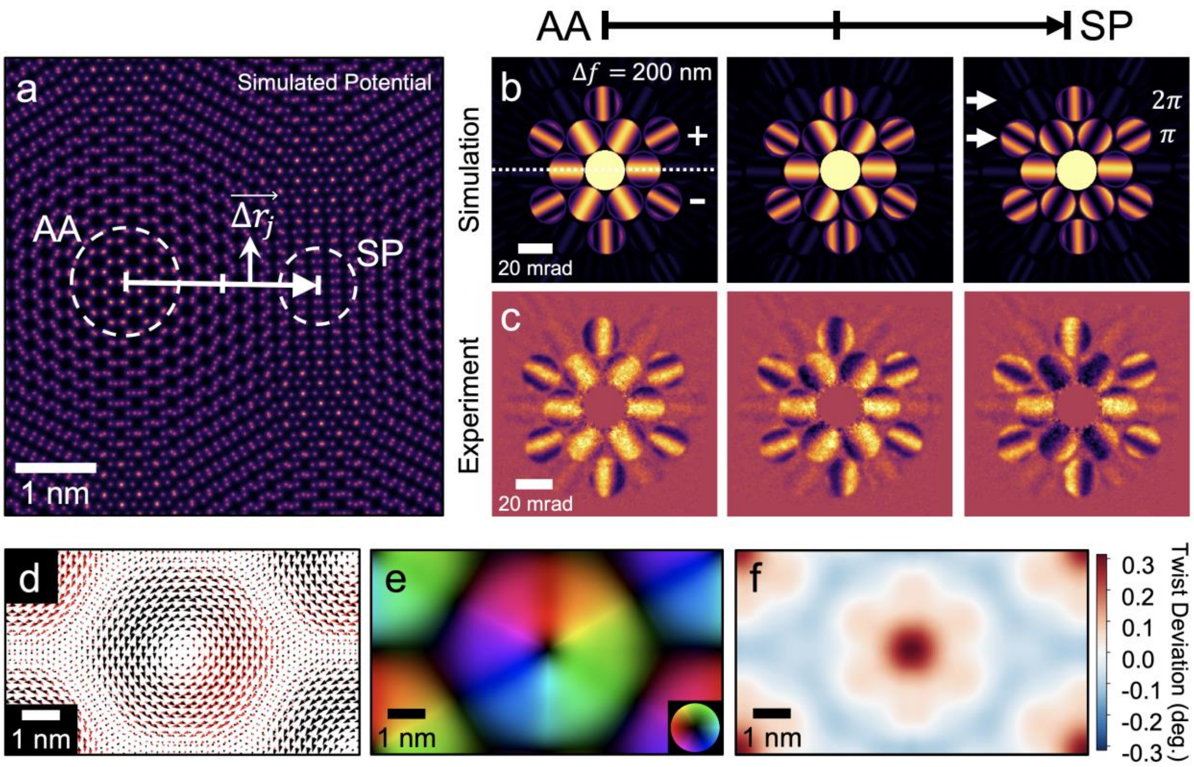

Figure 2. Mapping structural reconstructions in twisted bilayer graphene using interferometric 4D-STEM. (a) The positions, or phases, of interference fringes in the overlapped regions of Bragg disks originating from separate layers in a twisted 2D heterostructure are directly related to the relative positions of the atoms in the layers under the probe. Where the atoms from separate layers in bilayer graphene exactly overlap (AA stacking), these phases are zero. (b,c) The phases vary approximately linearly in the direction of the relative shift of the atom positions as a function of position away from the AA point. Simulated and experimental results are shown here for probe positions between an AA and SP region, where the atoms in one layer are shifted half of a unit cell in the direction of $\overrightarrow{\Delta r_j}$ in (a) with respect to the AA point. Mapping these phases and removing the linear portion of the result allows structural distortions to be visualized. (d) Density functional theory (DFT) shows that bilayer graphene with a twist angle of ~3 degrees has structural distortions in the top and bottom layers (black and red arrows, respectively) with maximum magnitudes of 4.6 pm. (e) Multislice electron scattering calculations of this structure demonstrate that interferometric 4D-STEM enables mapping of these pm-scale structural distortions. (f) From these distortions, properties such as local deviations in twist angle can also be mapped with nanometer resolution. Figure reproduced and condensed with permission from [11].

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