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Mapping Local Structural and Electronic Properties of 2D Materials by Multi-dimensional STEM

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Two-dimensional (2D) materials often possess interesting physical and electronic phenomena that are not present in bulk materials. For example, graphene has energy bands with a linear dispersion, which leads to low-energy charge carriers that act as massless fermions [1]. In addition, many-body interactions in 2D transition metal dichalcogenides result in complex phase diagrams yielding superconducting and charge density wave states [2]. Recently, efforts towards furthering the unique properties of 2D materials have been pursued, for example by forming bilayer structures. By utilizing materials with different lattice constants or by introducing a twist angle between the layers, a real-space moiré pattern is generated in the material that can act as a periodic electron scattering potential and lead to reconstruction of the material's energy spectrum [3,4]. In certain circumstances this can dramatically alter the properties of the material, as was recently discovered for bilayer graphene, where a 'magic' twist angle resulted in flat bands and strongly correlated electrons that generated correlated insulating and unconventional superconducting states in the material [5,6].

While techniques commonly used to study 2D materials such as optical spectroscopies and electrical transport measurements allow various properties to be explored under a variety of conditions and with high sensitivity, they lack the spatial resolution to probe properties locally around structures such as moiré patterns. Scanned probe techniques can provide high-resolution information but are typically restricted to samples lying on a substrate, which can alter the physical and electronic structure of the overlying 2D material [3]. Scanning transmission electron microscopy (STEM) and electron energy-loss spectroscopy (EELS) on the other hand can provide a host of structural, compositional, and bonding state information about samples suspended over vacuum down to the atomic-scale [7,8], but the ability of these conventional techniques to directly probe properties such as the local charge distribution is limited. Recently, however, pixelated detectors have allowed an electron diffraction pattern to be recorded at each probe position, as shown schematically in Fig. 1a, which enables properties such as the local strain or electric field to be directly measured, in some cases at the sub-Å scale [9-11].

Here, we utilize multi-dimensional STEM techniques to study suspended 2D materials like graphene, as shown in Fig. 2b-g. By performing *in situ* heating of the materials we enable large contamination-free areas to be maintained, allowing us to reveal the effects of a twist angle on the structural and electronic properties of bilayer materials from atomic-scale to longer-range features of the moiré pattern. Our results demonstrate the utility of multi-dimensional STEM for characterizing the inherent local properties of 2D materials that are difficult to obtain by other means [12].

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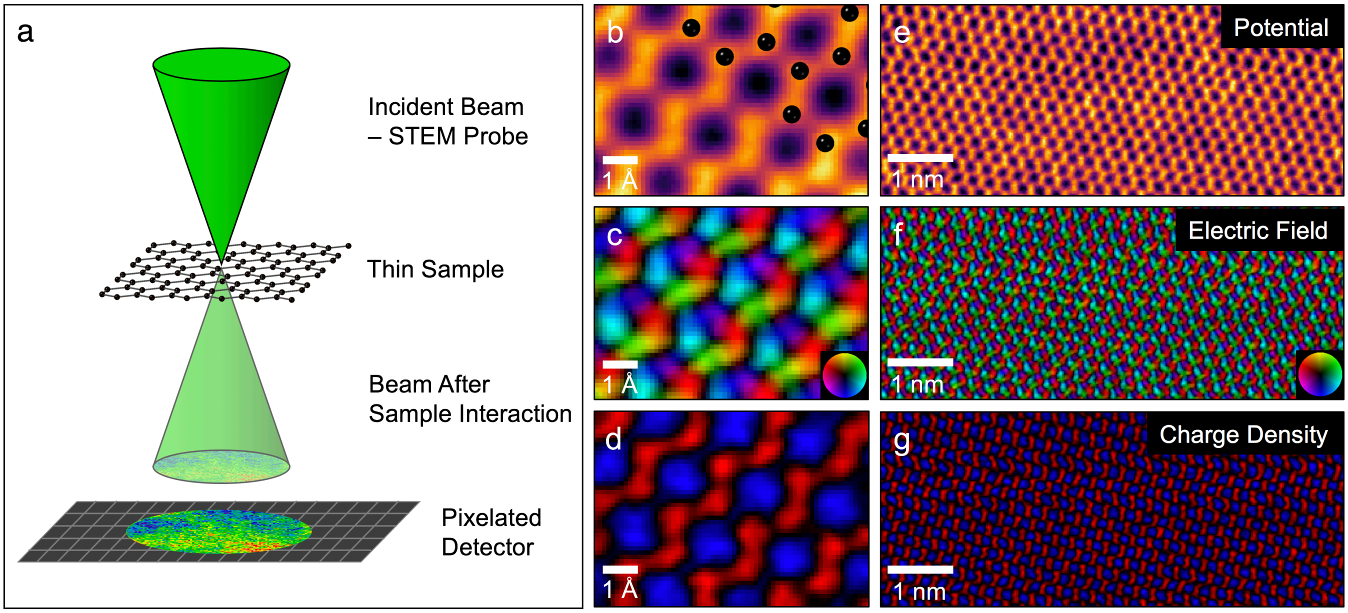

Figure 1. Schematic of center of mass STEM imaging and example of results for graphene. (a) As an incident electron probe passes through a sample, its phase is altered by the sample potential, producing variations in diffraction pattern intensity recorded on a pixelated detector. The resulting diffraction pattern center of mass shift is proportional to projected electric field at the measured probe position, which can be used to extract related properties such as projected sample potential and charge density. Acquiring diffraction patterns in a 2D array of probe positions allows projected potential, electric field, and charge density to be mapped. (b, e) Projected potential maps of graphene obtained from fields shown in (c, f). Graphene's crystal structure is also depicted in the upper-right corner of (b). (c, f) Projected electric field maps of graphene. Field strength and direction are given by inset color wheels. (d, g) Projected charge density maps of graphene obtained from electric fields shown in (c, f). Red and blue indicate positive and negative charge density, respectively. Note that electric field and charge density maps have been filtered by a gaussian with full width at half maximum of <0.5 Å to reduce high-frequency noise.