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Direct Mapping of Stacking Structure in Rotated Bilayer Graphene Using Aberration-corrected Transmission Electron Microscopy

Jong Min Yuk[1,3,4], Hu Young Jeong[1,2], Na Yeon Kim[1], Mi Jin Lee[1], Jeong Yong Lee[3,4], Zonghoon Lee[1]

[1.] School of Mechanical and Advanced Materials Engineering and [2.] UNIST Central Research Facilities, UNIST, Ulsan 689-798, Republic of Korea
[3.] Center for Nanomaterials and Chemical Reactions, Institute for Basic Science, Daejeon 305-701, Republic of Korea
[4.] Department of Materials Science and Engineering, KAIST, Daejeon 305-701, Republic of Korea

By layer-by-layer stacking various two-dimensional atomic crystals, it is possible to form multilayer heterostructures with designed electronic properties [1]. Bilayer graphene, especially, has attracted considerable interest because its electronic and optical properties provide useful characteristics not available to single layer graphene. A well-known example is opening tunable band gaps in Bernal-stacked bilayer graphene by applying transverse electric field [2], which makes them promising candidates for nanoelectronic devices. Even though these properties of bilayer graphene are radically varied depending upon their structures, including the stacking order [3], rotational angle and defects, their microstructural properties have not yet been studied in depth [4].

We present quick, accurate and large-area mapping of microstructures in rotated bilayer graphene, such as grains, defects and stacking rotation-angles. Because dark-field transmission electron microscopy (DF-TEM) images are sensitive to the alignment between the electron diffraction angle and the crystal orientation [5], we used DF-TEM to identify and visualize grains, defects and stacking structures, as shown in Fig. 1. By selecting each diffraction spot with an objective aperture, we are able to not only separate top and bottom layers, but also distinguish grains and grain boundaries (GBs) in each graphene layer. Moreover, we can acquire direct mapping of stacking structures in rotated bilayer graphene via Moiré patterns, which are performed by selecting two diffraction spots together of top and bottom layers with an objective aperture. We also used scanning electron diffraction with nanoparallel beam in scanning TEM mode, as shown in Fig. 2. These diffraction patterns show rotational angle change by sub-one degree across wrinkles in bilayer graphene. Figure 3 shows stacking structure of high-angle rotated bilayer graphene using high-resolution TEM (HR-TEM) in Cs corrected Titan Cube operated at a low kV. Our TEM study provides all crucial structural information of rotated bilayer graphene, which is sincerely demanded for their future development in optoelectronic and nanoelectronic devices.

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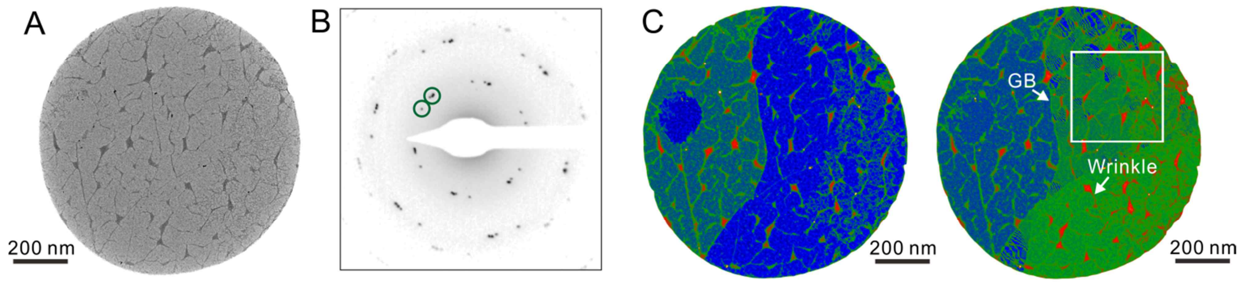

Figure 1. (A) Bright-field TEM image of artificially stacked bilayer graphene. (B) Electron diffraction pattern of (A). (C) Color-scaled DF-TEM images acquired by selecting diffraction spots in (B) with an aperture. Green indicates the corresponding grains with selected diffraction spots.

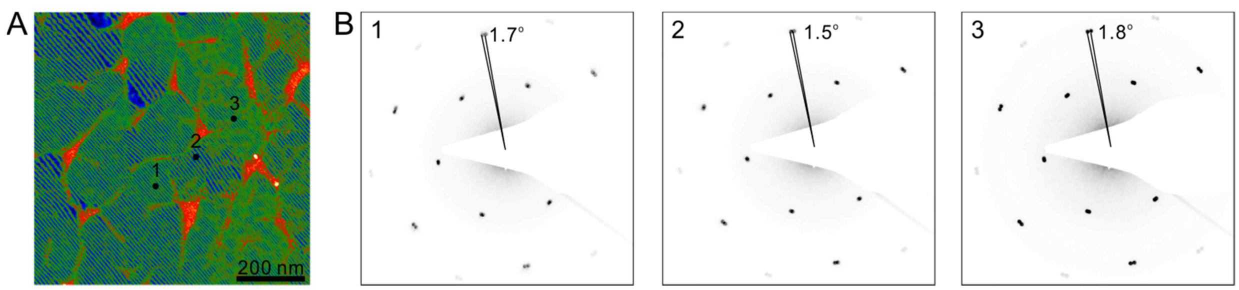

Figure 2. (A) Zoom-in DF-TEM image of rectangular region in Fig. 1(C). (B) Electron diffraction patterns acquired with nanoparallel beam at positions 1, 2 and 3 in (A).

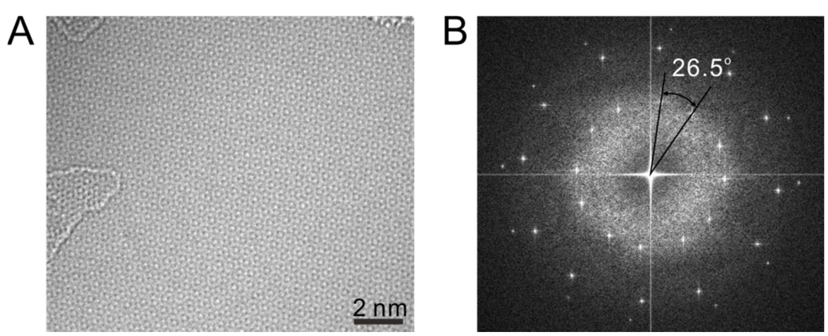

Figure 3. (A) HR-TEM image of high-angle rotated bilayer graphene. (B) Fast Fourier transformation of (A), displaying interlayer rotation angle of 26.5° between top and bottom layers.

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