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AB/AC Stacking Boundaries in Bilayer Graphene

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Boundaries, including grain boundaries and domain boundaries, are well known to modify the physical properties of materials. For example, it has been shown that certain types of grain boundaries in graphene can weaken the mechanical properties and degrade the electrical performance [1, 2]. Besides the well-studied grain boundaries, unique stacking boundaries can be present in bilayer graphene (BLG) at the interfaces of domains with the same crystal orientation but different stacking, *i.e.* AB and AC stacking. Recent studies have shown the coexistence of AB and AC stacking domains and the presence of domain boundaries [3]. However, little is known about the detailed atomic structure of these AB/AC stacking boundaries formed in BLG. In particular, it is not known if the stacking boundaries are atomically sharp like grain boundaries.

A CVD-grown bilayer graphene sample is shown in Figure 1. Tilted dark-field transmission electron microscopy (DF-TEM) imaging at 80 kV reveals the AB and AC stacking domains by their mirror-symmetric intensity during tilting (Figure 1a & 1b). Using the second order diffraction spot, the boundaries between the AB and AC stacking domains can be observed as dark lines in the DF-TEM image shown in Figure 1c. The decrease in DF-TEM image intensity across the stacking boundaries suggests that the transition from AB to AC stacking takes place over a width of a few nanometers.

The atomic structure of stacking boundaries is further investigated using annular dark-field (ADF) imaging on an aberration-corrected scanning transmission electron microscope (STEM) operated at 60 kV. The ADF image can be approximately interpreted as the convolution of the projected atomic positions in both graphene layers and the electron probe. While AB or AC stacking generates regular patterns with half of the carbon atoms overlapped, we observed irregular Moiré patterns at the stacking boundary regions. The precise pattern changes gradually across the stacking boundaries as shown in Figure 2. The observation demonstrates that the stacking boundaries are indeed continuous transitions from AB to AC stacking over a distance of several nanometers, which is consistent with the DF-TEM results.

Via density functional theory (DFT) and classical molecular dynamics calculations we further show that, unlike grain boundaries, the low-energy stacking boundaries are not sharp. Instead, they are nm-wide strain ripples that provide smooth transitions between the AB and AC stacking, in accord with the experimental observations. By optimizing the height, width and strain of the ripples using molecular dynamics, all the experimental STEM images can be reproduced by the simulated structural model (Figure 2a), confirming that the stacking boundaries exist as ripples. [4]

References:

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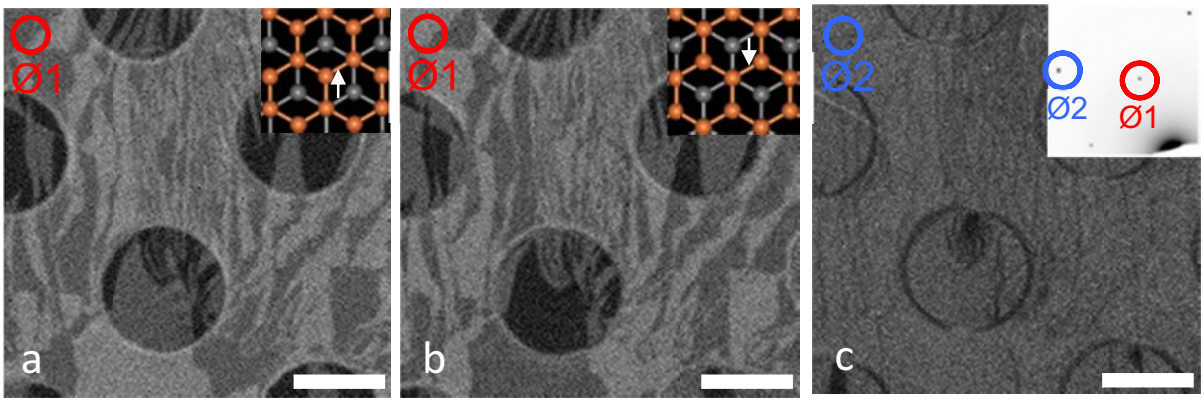

Figure 1. DF-TEM images using the first order diffraction peak Ø1 (red circle) tilted at a) 10° and b) -10°; and c) the second order diffraction peak Ø2 (blue circle) at zero tilt. Inset in a) & b): Schematic of AB and AC stacking in oBLG. Scale bars: 1 μm.

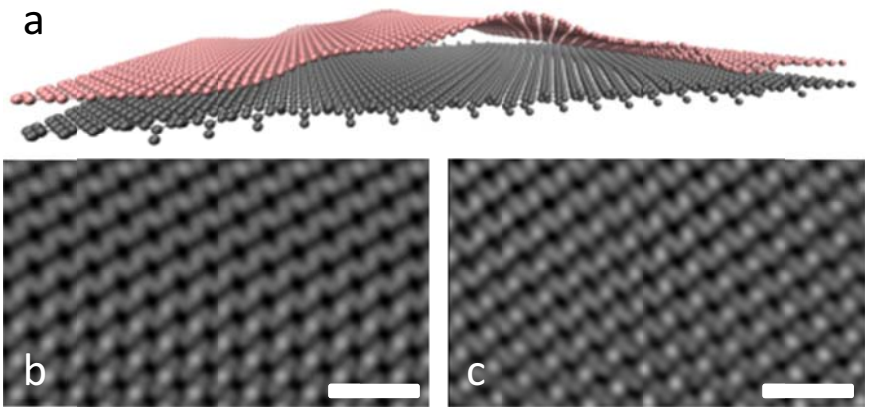

Figure 2. a) Atomic structural model of an AB/AC stacking boundary. b) Simulated STEM image using the above model compared with c) experimental ADF image, both varying gradually across the field of view. Scale bars: 0.5 nm.

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