

Structural Analysis for Highly Aligned Graphene Fold on Cu(111) Substrate

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The intrinsic properties of graphene such as high in plane elastic modulus and low out of plane bending stiffness make the graphene to create a variety of new structures through fold, twist, and/or fracture [1]. According to the preceding research results, if the graphene becomes folded with specific stacking angle, it is expected to show novel characteristics such as interferometric effects when the magnetic fields are engaged into the graphene fold region [2]. It is also known that the charge confinement effect can be seen through the conductance test across the graphene fold [3]. Thus, graphene fold may be used as a promising material for valleytronic applications, particularly in protocols for quantum computation [4].

In general, when the graphene is synthesized through CVD method on the metal substrates, lots of ribbon-like graphene folds can be made during the cooling process after graphene growth at high temperature due to the difference in thermal contraction characteristics between grown graphene layers and used metal substrate. Typically, these graphene folds are randomly formed under the influence of crystal orientation of the substrate. However, by limiting the crystal orientation of the used substrate to a single orientation, it becomes confirmed that a highly aligned graphene fold can be formed aligned into one direction and parallel to each other [5-6].

Herein, we demonstrate the structural information of highly aligned graphene fold grown on a single crystal Cu(111) foil. Through the atomic resolution transmission electron microscopy, the exact stacking order and crystal orientation of fold region becomes clarified in both monolayer and multilayer graphene cases [7]. Figure 1 shows the graphene fold in monolayer graphene. It is not continuous but cracked into back-and-forth patterns. Always AB-stacked bilayer joint regions are formed at the joint that is connecting two alternating fold regions, and the cracks always propagate into two directions along zigzag or armchair directions. Figure 2 shows the graphene fold in multilayer graphene. Grown fold on multilayer graphene remains continuous with no cracks. This research clearly reveals the structural information of graphene fold and contributes to find the exact formation mechanism of graphene fold. Thus, this study will be able to provide basic knowledge for future research including the field of valleytronics [8].

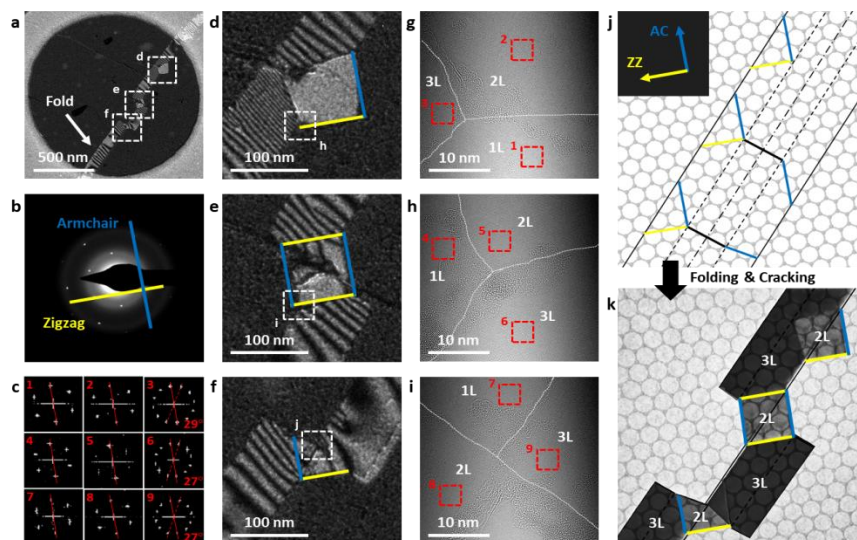


Figure 1. (a) DF TEM image showing graphene fold in monolayer graphene. (b) SAED pattern obtained from the whole region in (a). (d-f) Magnified Dark-field TEM images of the regions marked in (a). (g-i) AR TEM images showing graphene fold regions marked in (d-f). (c) FFT patterns obtained from the regions marked in (g-i). (j-k) A paper kirigami illustrating the structure of graphene fold in (a).

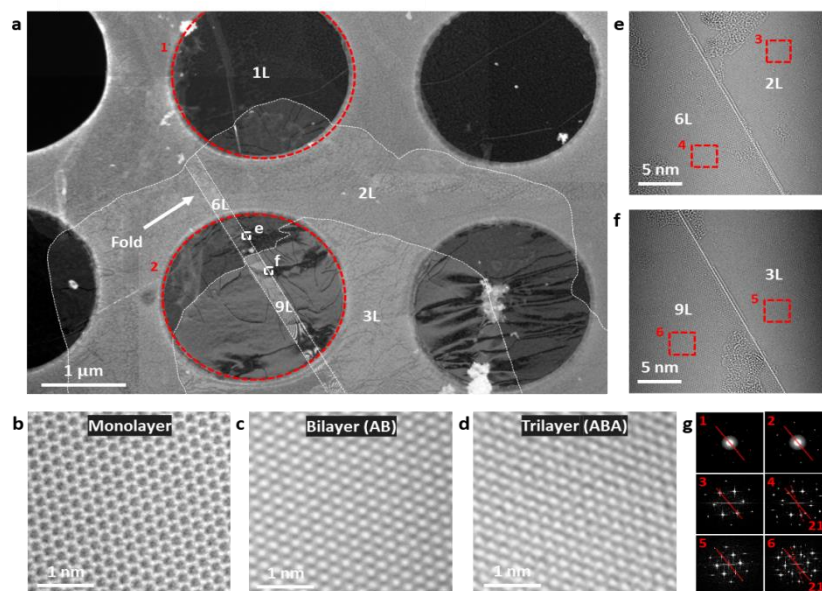


Figure 2. (a) Stitched DF TEM images showing graphene fold in multilayer graphene. AR TEM images captured from (b) monolayer, (c) bilayer, and (d) trilayer regions in multilayer graphene. Images show bilayer region is AB-stacked and trilayer region is ABA-stacked. Atomic resolution TEM images showing graphene fold in (e) AB-stacked bilayer, and (f) ABA-stacked trilayer graphene correlated to the marks in (a). (g) (1-2) SAED patterns and (3-6) FFT patterns obtained from the regions marked in (a-c). Both the graphene folds in bilayer and trilayer graphene show two sets of patterns.

References:

- [1] MK Blees et al., *Nature* **524** (2015), p. 204-207.
- [2] D Rainis et al., *Physical Review B* **83** (2011), p. 165403.
- [3] Y Wu et al., *Nano Letters* **18** (2018), p. 64.
- [4] D Faria et al., *Physical Review B* **101** (2020), p. 081410.
- [5] M Huang et al., *ACS Nano* **12** (2018), p. 6117.
- [6] D Luo et al., *Advanced Materials* **31** (2019), p. 1903615.
- [7] D Luo et al., *Advanced Materials [Online]*, (2022), p. 2110509.

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