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Direct transmission electron microscopic observation of the oriented edge-attachment processes between single-layer graphene flakes

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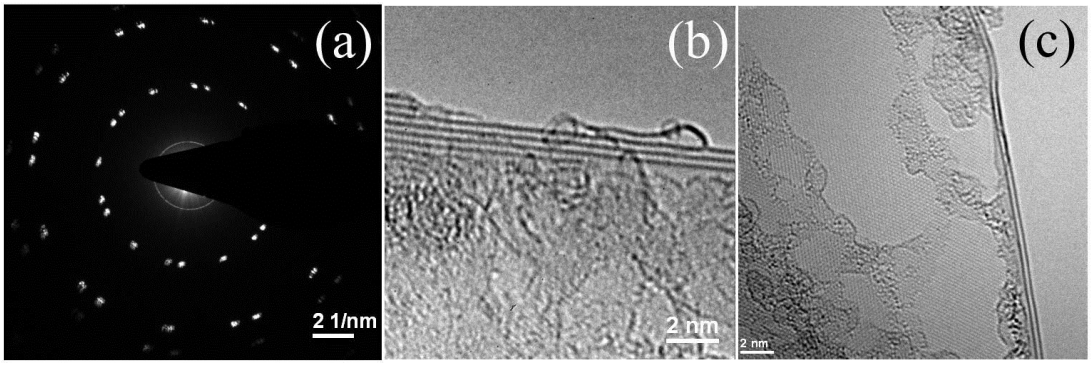


Figure s1. (a) SAED pattern of the graphene structure. The splitted diffraction spots indicate the multilayer nature of the sample. And typical TEM images obtained at the edge of the multilayer graphene structure: (a) normally observed thick region with layer > 3; (b) thin region with a double layer structure.

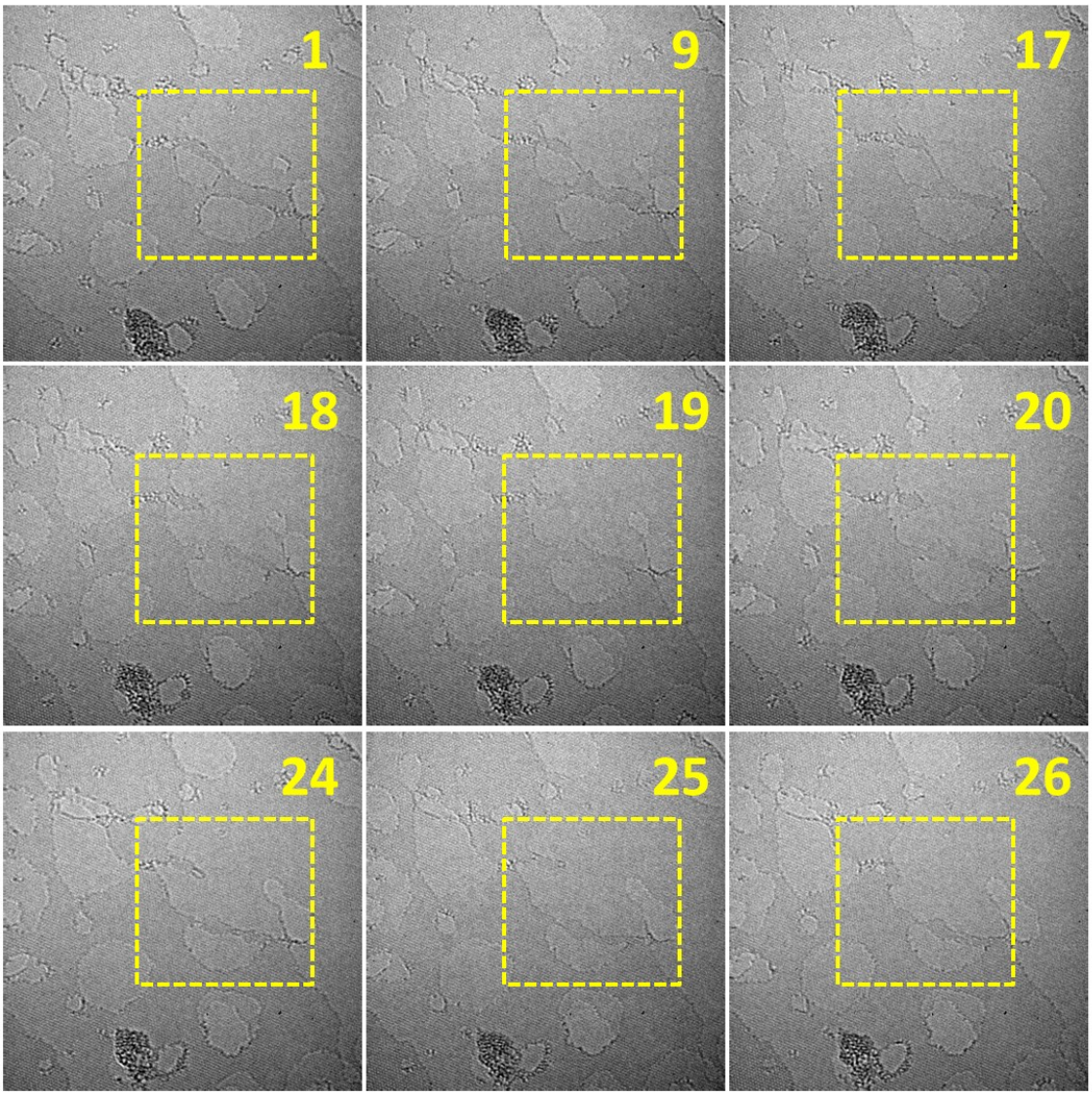
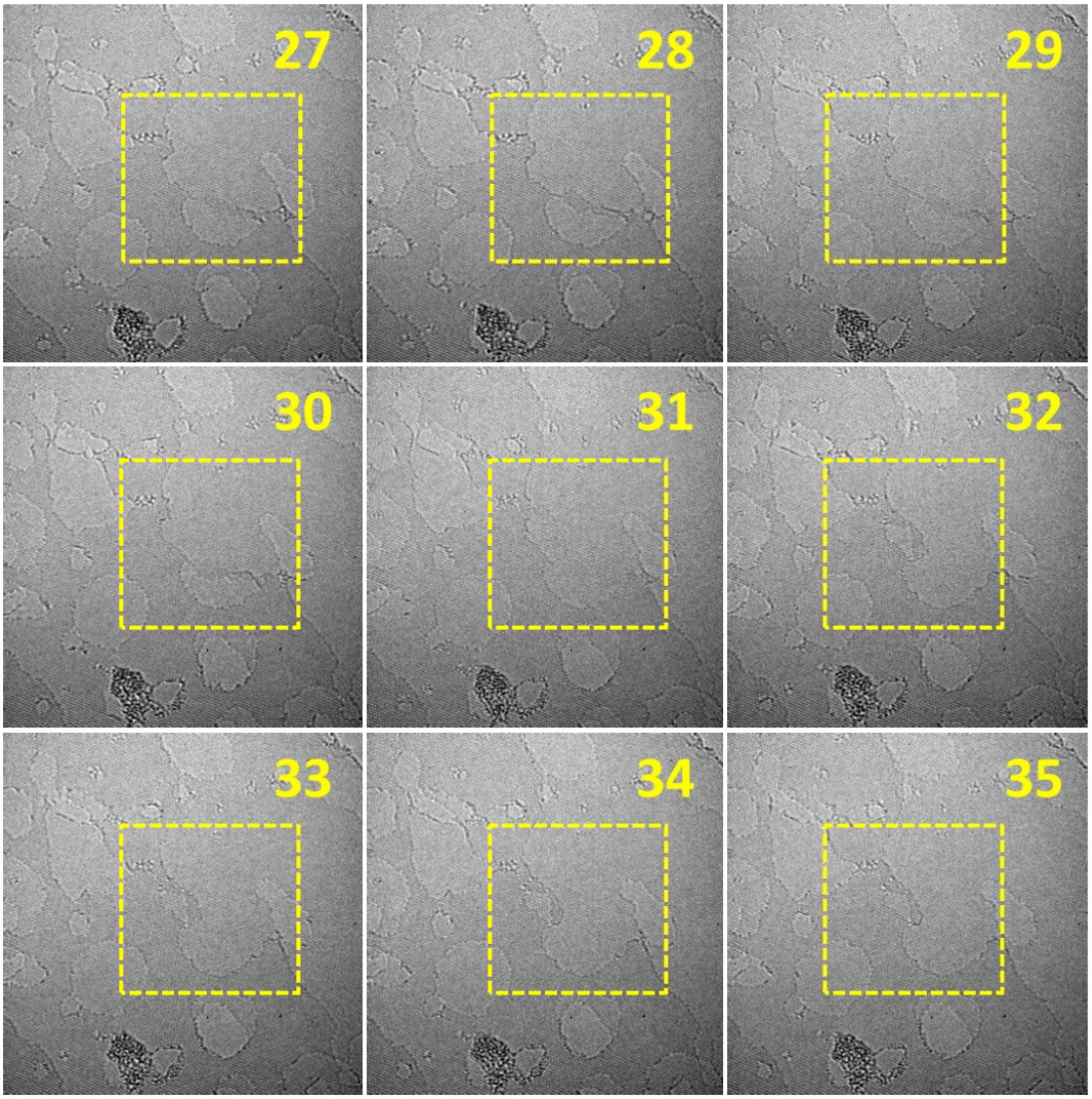


Figure s2. (Continued) Detailed TEM image sequence shows the re-orientation, alignment and merging of the GFs. Yellow dashed rectangular highlight the interested region. Numbers is the sequence of the image frame.



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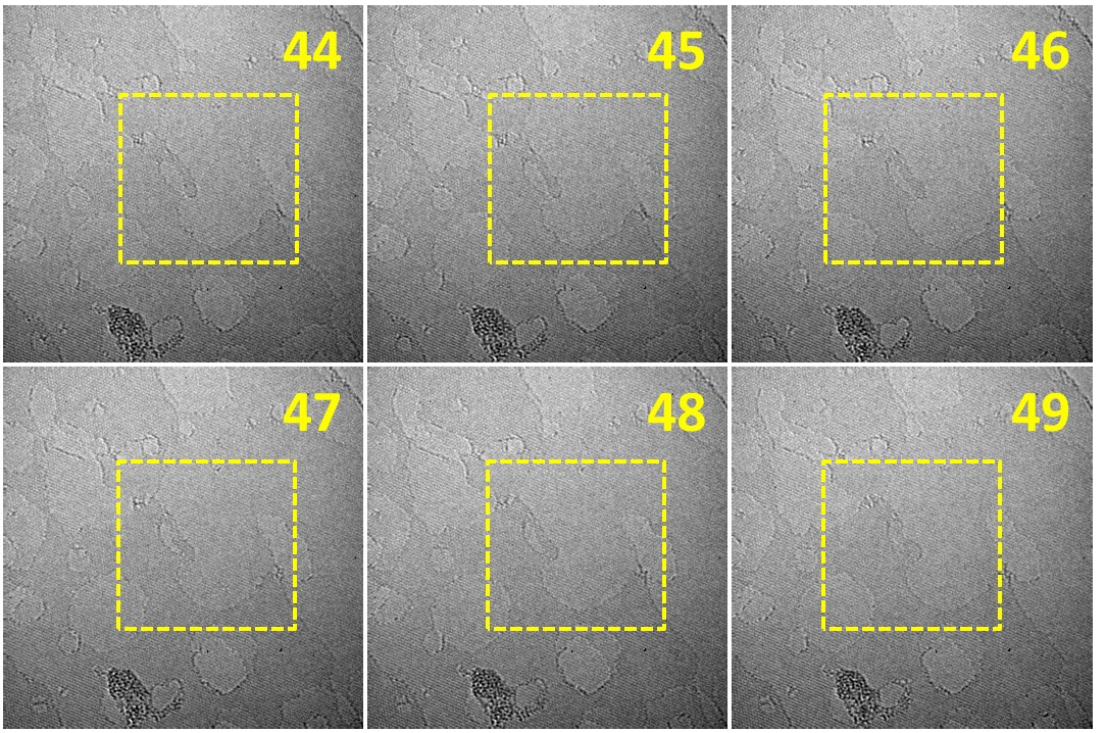

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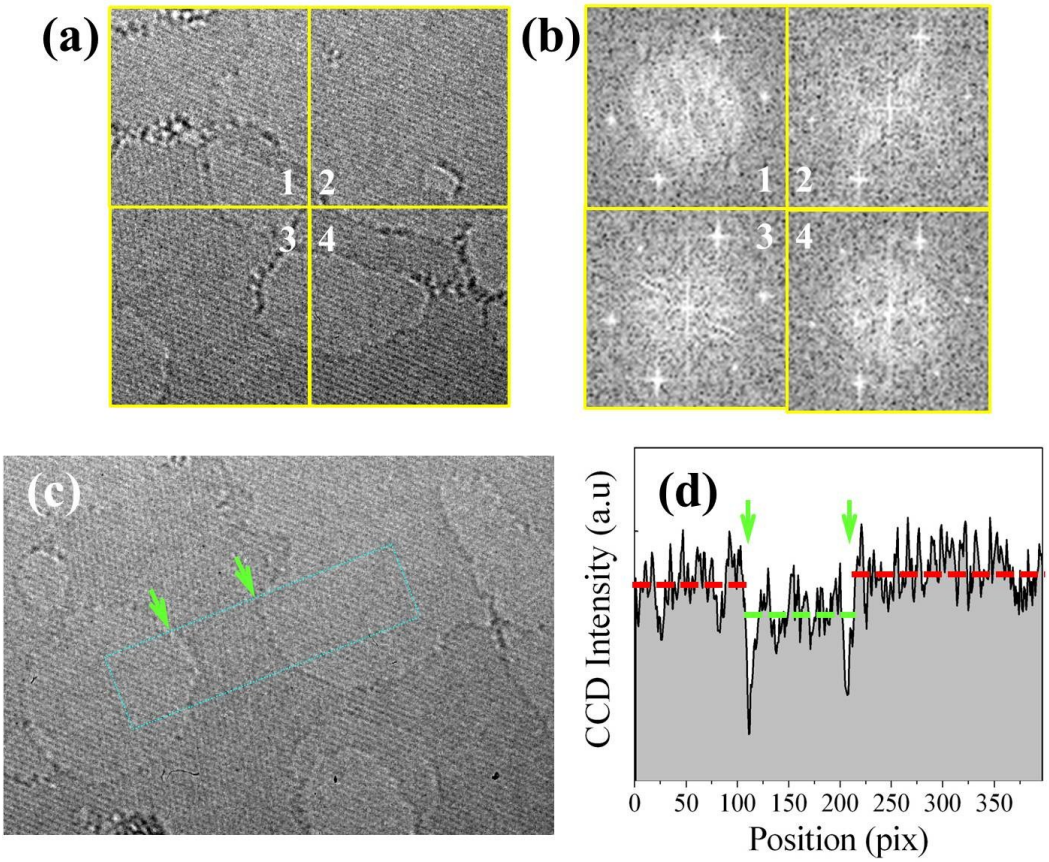

Figure s3. Survey of the TEM image by the fast Fourier transformation (FFT) pattern performed at different places indicated the single in-plane symmetry of the graphene layers. Single crystal structure can be observed as the FFT pattern normally performed at much smaller region. The relative larger select area aperture normally resulted in a polycrystal diffraction pattern as shown above. They are not contradicting with each other. (a) The TEM image. (b) FFT patterns performed at different position of the TEM image as indicated by “1” to “4”. (c) and (d) show the line profile contrast analysis of the multilayer graphene layer structure. Position of the line profile is shown in (c) with two green arrows indicating the layer edges. Dashed lines in (d) indicate the positions with different layers. Note higher CCD intensity indicates smaller thickness thus smaller graphene layers (see also the images simulation presented below). Sharp contrast dips indicated by green arrows come from the layer edge.

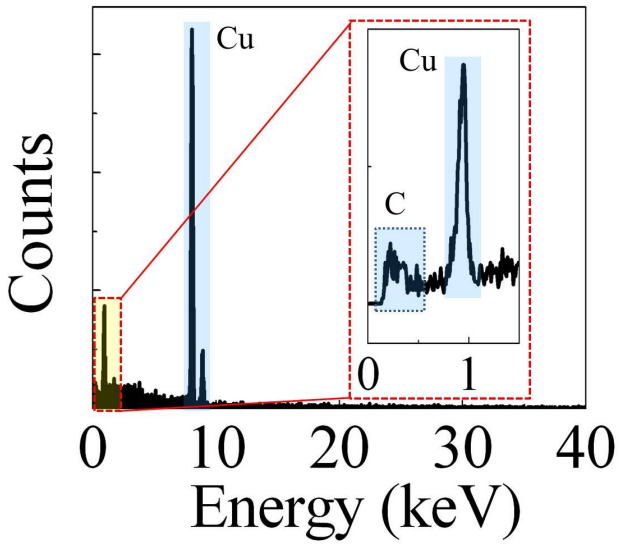

Figure s4. Typical X-ray energy dispersive spectrum (EDS) pattern obtained from the sample. Inserted figure is a magnified region (shaded with yellow) of the EDS spectrum as indicated.

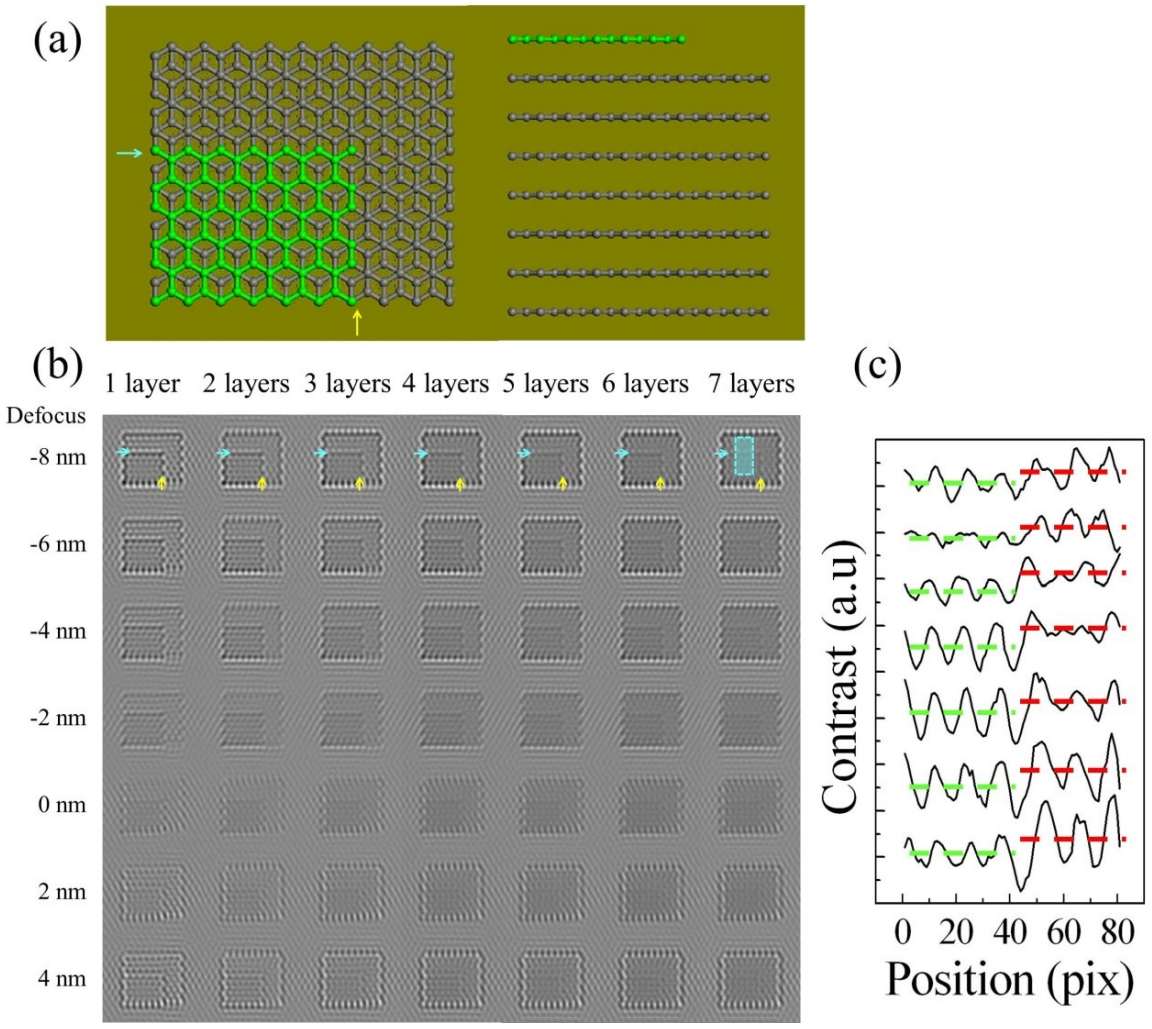

Figure s5. Image simulation for contrast interpretation. (a) Plan and cross-sectional view of the atomic model used for simulation. (b) Simulated images with different number of graphene layers and different defocus. Cyan and yellow arrow indicates the position of zigzag and armchair edge of the supported graphene layer, respectively. (c) Contrast line profile for the "7-layer" case with different defocus, from -8 to 4 nm (top to low). A cyan rectangular drawn in 7-layer with defocus = - 8 nm indicate the position used to compare the contrast.