Supporting Information

Huang et al. 10.1073/pnas.0905193106

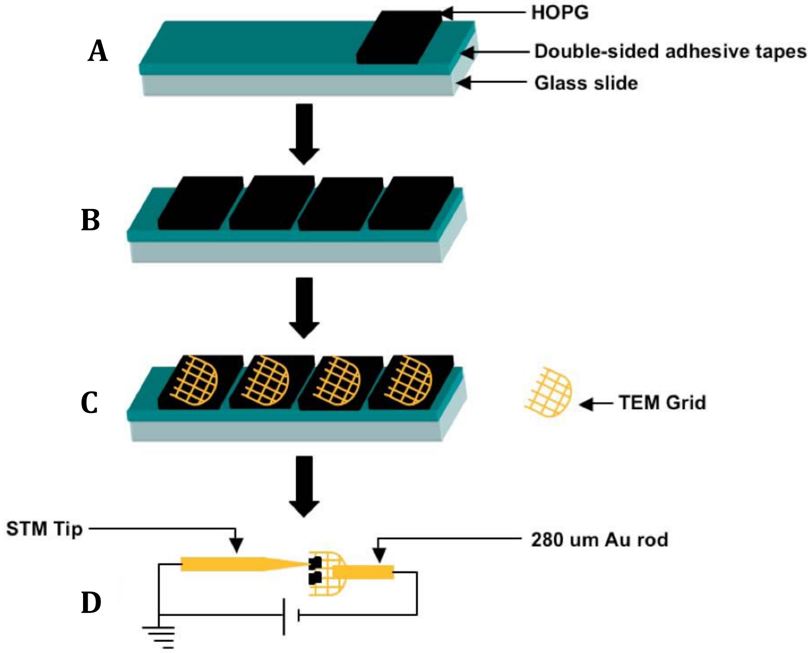


Fig. S1. TEM sample preparation procedures. Highly orientated pyrolytic graphite (HOPG) with a thickness of a few hundred μm was glued to a glass slide with a double-sided adhesive tape (Fig. S1 *A*). The HOPG was thinned down to transparent under an optical microscope by repeated peeling using a Scotch tape (Fig. S1*B*). A 200-mesh transmission electron microscopy (TEM) grid was cut into half, painted with conducting silver epoxy on the grid bars, and then glued on the transparent graphene sheet on the glass slide. Once the silver epoxy was cured, the half grid was lifted off from the glass slide (Fig. S1*C*). Graphene was attached to the grid (sketched in dark) after lifting off from the glass slide (Fig. S1*D*). The half TEM grid was glued to an Au rod of 280 μm and inserted into a Nanofactory TEM-scanning tunneling microscopy (TEM-STM) platform (Fig. S1*D*), in which a full functional STM is integrated into a TEM sample holder, allowing for in situ manipulation and measurements of individual graphene.

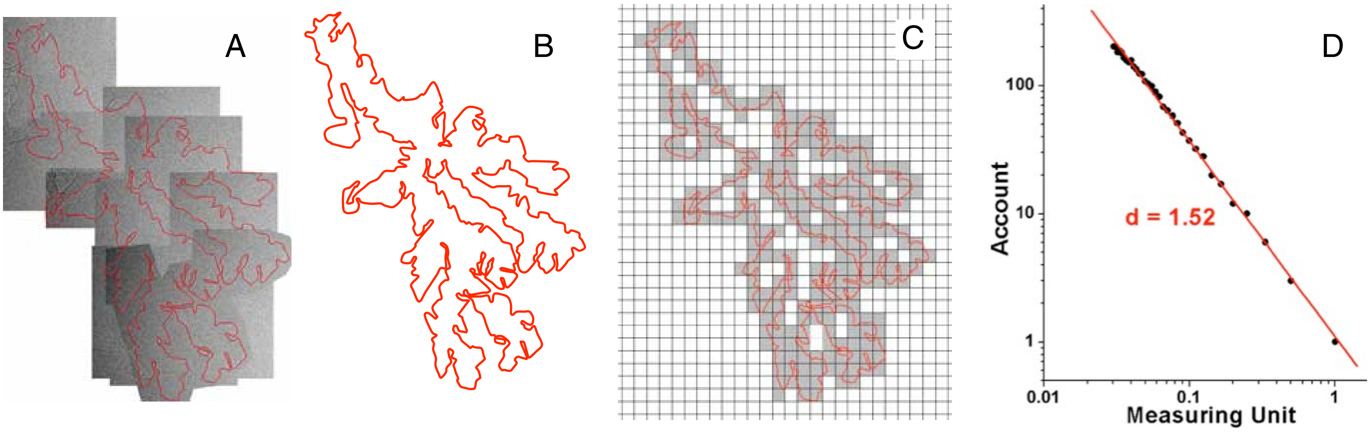


Fig. S2. Fractal dimension measurement procedures. The fractal dimension of the propagating front pattern was measured by the same method as that used to measure the coastline [Sapoval B, Baldassarri A, Gabrielli A (2004) *Phys Rev Lett* 93:098501]. First, a close loop was drawn by fitting the experimental propagating sublimation front (Fig. S2 *A* and *B*). Then the close loop was measured on a 2D square lattice. In the measurement, only those squares that intersect with the close loop were accounted (e.g., the gray squares in Fig. S2*C*). By varying the size of the squares, the number of accounted squares as a function of a square size (i.e., the measuring unit) was plotted (Fig. S2*D*). According to the definition of the fractal dimension, the fractal dimension of the loop or the propagating loop is calculated as:

$$d = -\frac{d\log(N)}{d\log(I)},$$

where N is the number of the accounted squares and I is the size of the squares or the measuring unit. For the propagating front shown in Fig. S2*A*, the fitted fractal dimension is 1.52.

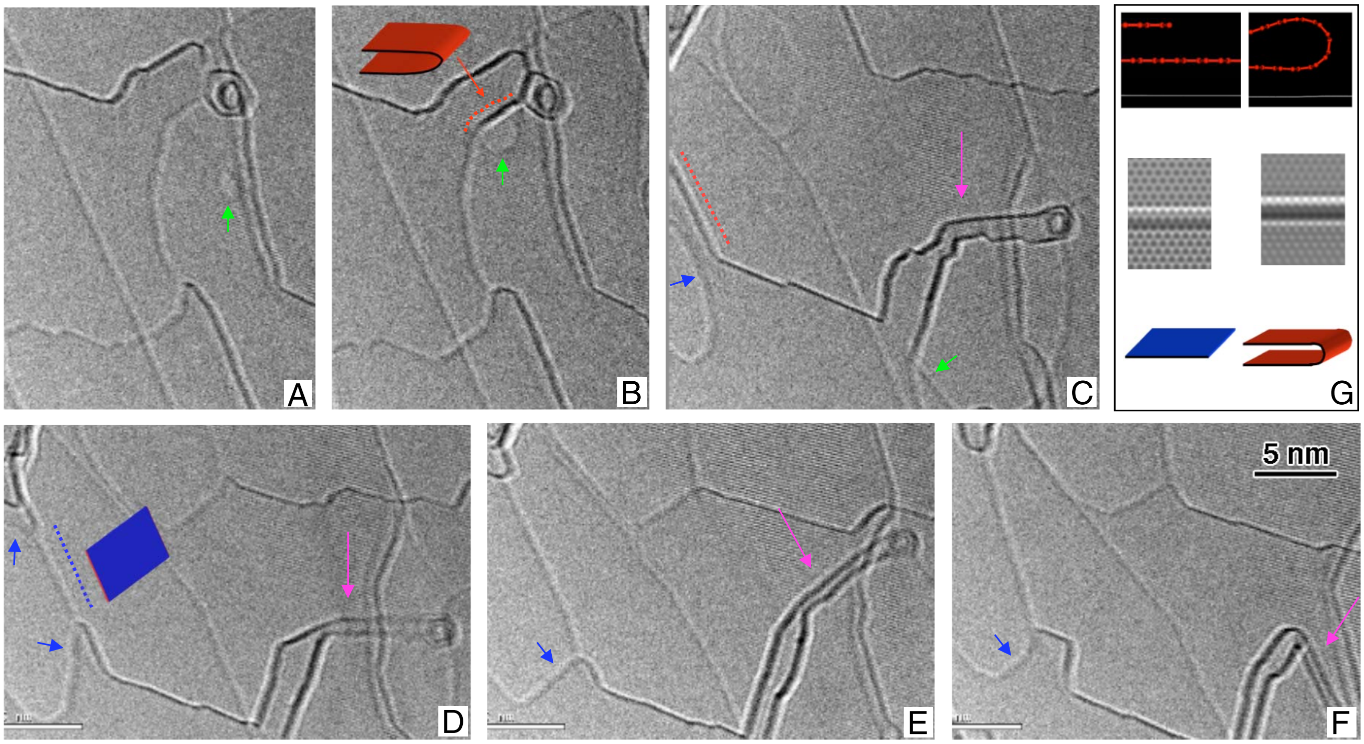


Fig. S3. Graphene edge evolution and their structural configuration identification. There are two different edge configurations: a MLE and a BLE, as shown in the first and the second columns in *G*, respectively. Edges marked by red-dotted lines (*B* and *C*) are BLEs, because when the vacancies marked by green arrows touched that edge, the vacancy etched away 1 graphene layer (*A*–*C*), leaving a bilayer graphene. From *C* and *D*, where another vacancy pointed out by blue arrows hit the edge highlighted in red in *C*, it etched away 1 layer of the bilayer graphene (*D*–*F*). So one can see the debris pointed out by pink arrows in *C*–*F* is indeed a carbon nanotube (because it has 2 layers at its edge), not a single graphene ribbon. The free-end of the tube was not bonded to the graphene substrate, and it could bend back and forth freely. There was actually a fullerene in the nanotube cap. Simulated images of the above 2 different edges (middle rows of *G*) qualitatively agree with the experimental images very well, say a BLE shows a stronger contrast than a MLE in both the experimental and the simulated images.

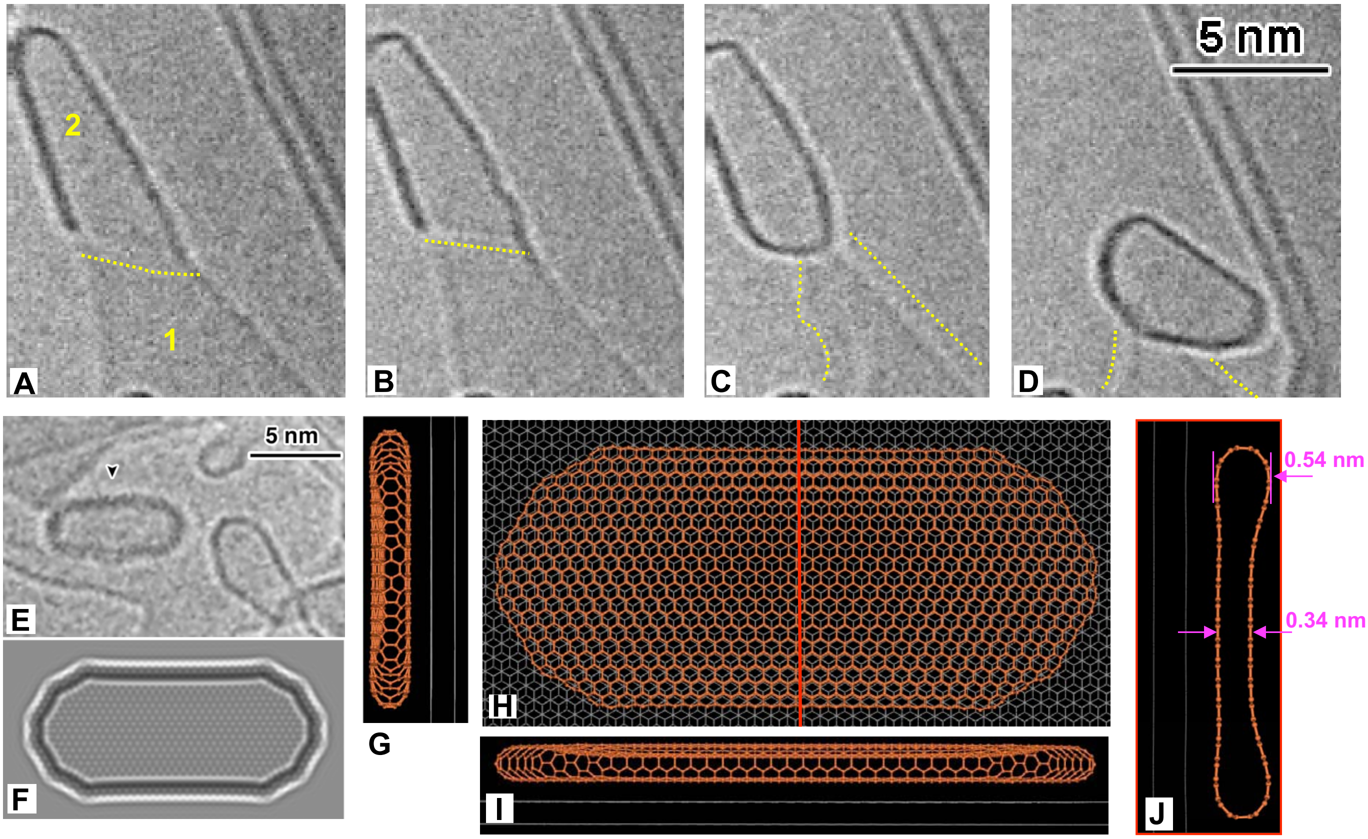
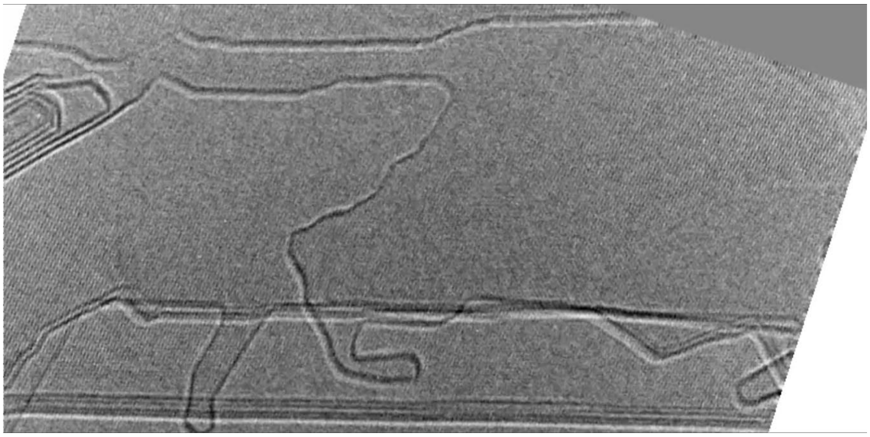
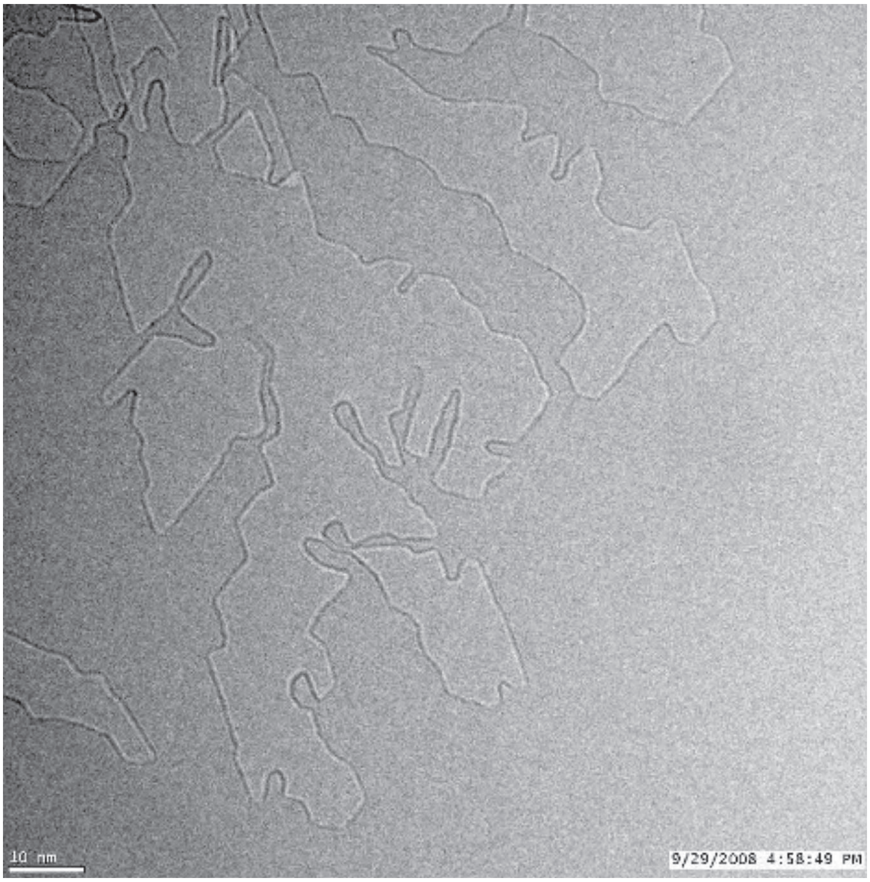


Fig. S4. In situ formation of flat fullurenes/nanopods on a graphene substrate. (*A-D*) In situ sequential HRTEM images showing the formation of a flat fullerene/nanopod on a graphene. Dotted-yellow lines mark MLEs. Numbers ''1'' and ''2'' mark the number of graphene layers. The MLE gradually sublimated along its edge, and eventually it reconstructed with a neighboring graphene layer, forming a nanopod/flat fullerene. (*E*) Another nanopod/flat fullerene (pointed out by an arrowhead). (*F*) A simulated image of a nanopod/flat fullerene as shown in (*G–J*). The simulated image (*F*) matches the experimental image (*E*) very well. (*G–I*) Three perspective views of the optimized nanopod/flat fullerene structure. (*J*) The cross-sectional view of the nanopod/flat fullerene (across the red line in *H*).



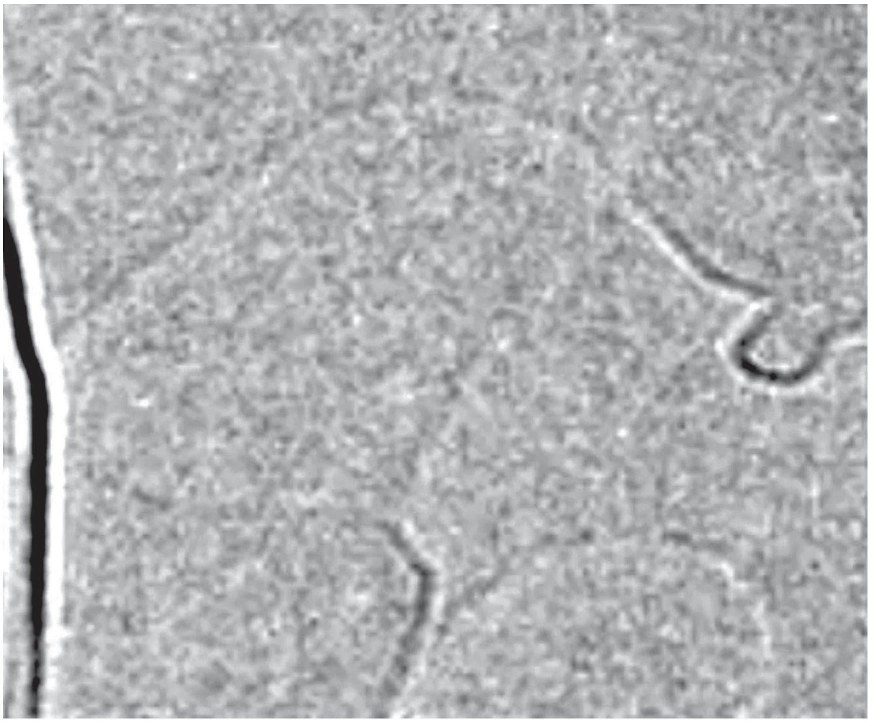
Movie S1. An in situ HRTEM movie showing a void propagation in graphene due to atom sublimation. The sublimation was caused by a high bias Joule heating and electron irradiation. The movie was recorded at 1 frame per second and played at 15× speed.

Movie S1



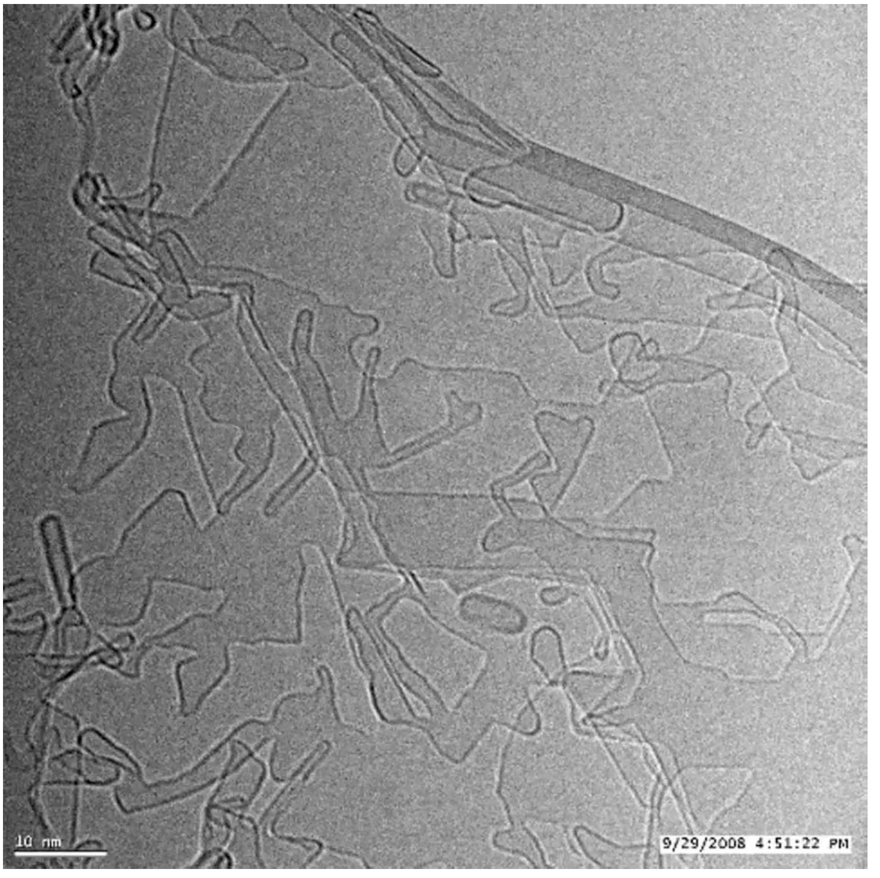
Movie S2. An in situ HRTEM movie showing the fractal sublimation of graphene. The movie was recorded at 1 frame per second and played at 25× speed.

Movie S2



Movie S3. An in situ HRTEM movie showing the reconstruction of MLEs to BLEs. The movie was recorded at 1 frame per second and played at 10× speed.

Movie S3



Movie S4. An HRTEM movie showing in situ formation of a large variety of nanostructures connected to graphene. The framed area shows a flat fullerene doing a random walk on the graphene. The movie was recorded at 1 frame per second and played at 20× speed.

Movie S4