Atomic scale site and direction-specific patterning of freestanding monolayer graphene at elevated temperature

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Interest in the use of monolayer graphene in low-dimensional physics research and nanoscale devices has increased rapidly since its discovery in 2004 [1, 2]. Many applications require graphene to be patterned into ribbons or pores that have well-defined dimensions and edge configurations with respect to its crystallographic lattice[3, 4]. Since both theoretical and experimental studies indicate that the edges and geometries of graphene sheets can have a major influence on their electronic and magnetic properties[5], a highly reproducible technique for sculpting monolayer graphene in nano-scale is highly demanded. Compare to lithography method and STM etching, high energy electron beam induced radiation damage provides a fast way to sculpt free-standing graphene with ultimate precision, theoretically down to atomic resolution. We have previously shown that a transmission electron microscope (TEM) operated in high-resolution mode can be used to sculpt *multi-layer* graphene [6]. However, controllable sculpting of *mono-layer* graphene is difficult due to the extreme sensitivity of mono-layer graphene to electron beam.

By accurate control of electron beam irradiation damage and the use of *in-situ* heating in the TEM[7], we are now able to achieve site & direction-specific atomic scale sculpting of graphene (See Figure 1 A-D). In-situ heating is required for allowing the sculpting and imaging process of graphene operated at 500-700 °C, at which the self-repair of electron beam induced defects occurs rapidly, thus preventing the growth of point vacancies into big stable voids. The combination of destructive damage and constructive repairing allow one to accurately control the sculpting process. Our results show that for all the sculpted patterns, defect-free crystallinity is achieved up to the edge (See Figure 2); the atomically straight edges in <1-100> (zigzag) and <1-210> (armchair) directions can be obtained. In other directions, the edges of the ribbons are usually composed of zigzag and armchair fragments. The width of ribbon and the diameter of nano-pore can be precisely controlled down to less than 2nm. The whole pattering process is automatically executed by running a computer script, assuring the reproducibility of sculpting (See Figure 1 E-F).

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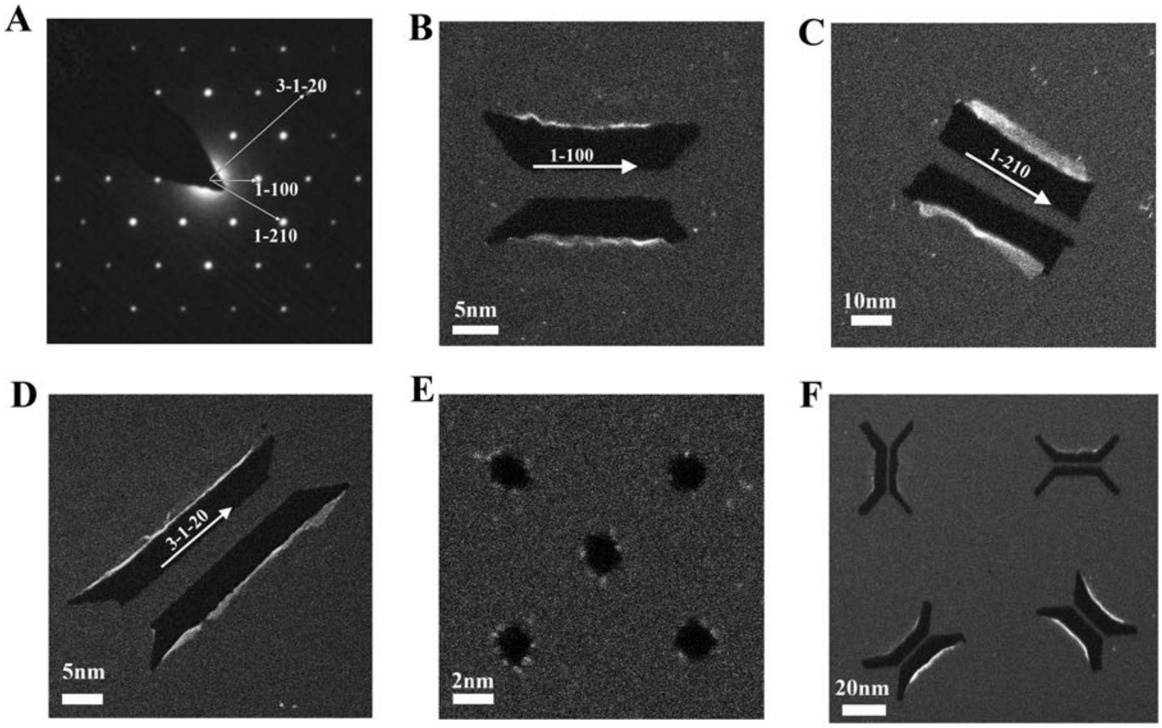

Figure 1 *(A) Diffraction of monolayer graphene (B-C) nano-ribbon along <1-100>,<1-210>,<3-1-20> orientation respectively. (E) Pattern of nanopores with 2nm diameter (F) Pattern of nanoribbon with 1.2nm width, created using computer scripts.*

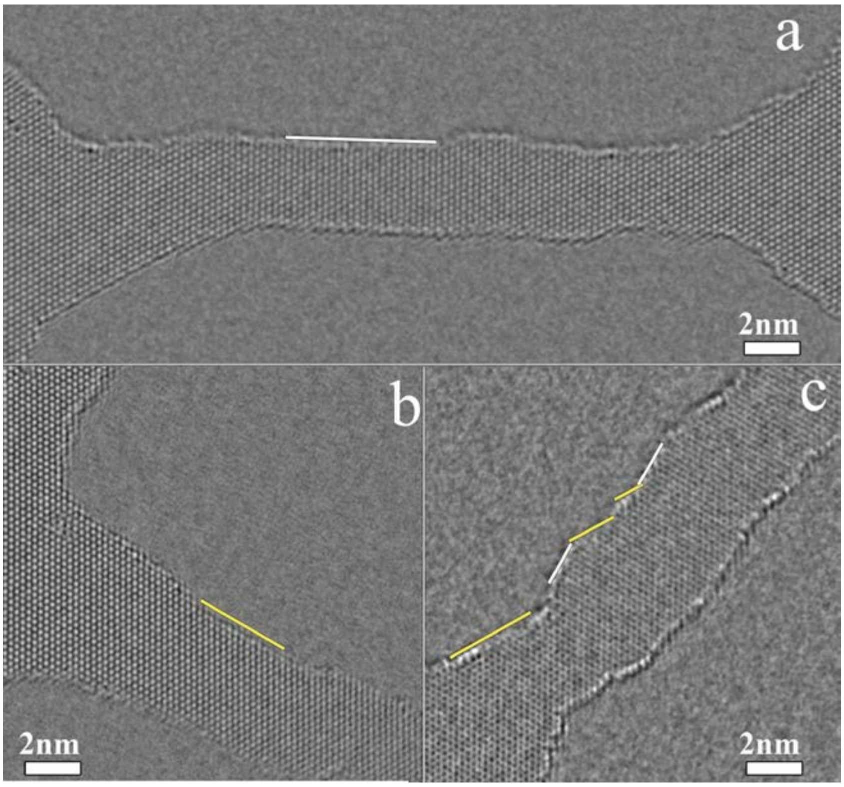

Figure 2 *(A-C) HREM images of nano-ribbons along <1-100>,<1-210>,<3-1-20> respectively, recorded at 80kV using FEI Titan 60-300 PICO TEM, equipped with Cs-Cc achro-aplanat image corrector.*