

Supporting Information for

The Liquid-Assisted Tip Manipulation: Fabrication of Twisted Bilayer Graphene Superlattices on HOPG

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In order to display the high-resolution topography of sample surface after the organic liquid treated, the HOPG samples were also measured in ultra-high vacuum chamber with liquid nitrogen temperature. It shows that most regions of the sample show clean graphite surface (Figure S1 (a)), while some small areas are covered by organic molecules (Figure S1(c),(d) and (f)). It demonstrated that no large area of organic film formed on the sample surface, and the arrangement of thiol molecules are locally ordered but with multi-domains, as shown in Figure S1 (d) and (f). The multi-domain ordered structure has been reported previously and can be attribute to the fact that the carbon chain length is too short to form the large-area ordered structures on HOPG surface. As a result, the organic molecules are very easy to mobile on surface during the STM scanning and even preferentially adsorb on the tip apex, which lead to increase the hydrophilic property of HOPG surface and induce the capillary force between the tip and surface (such as Figure S1(c)).

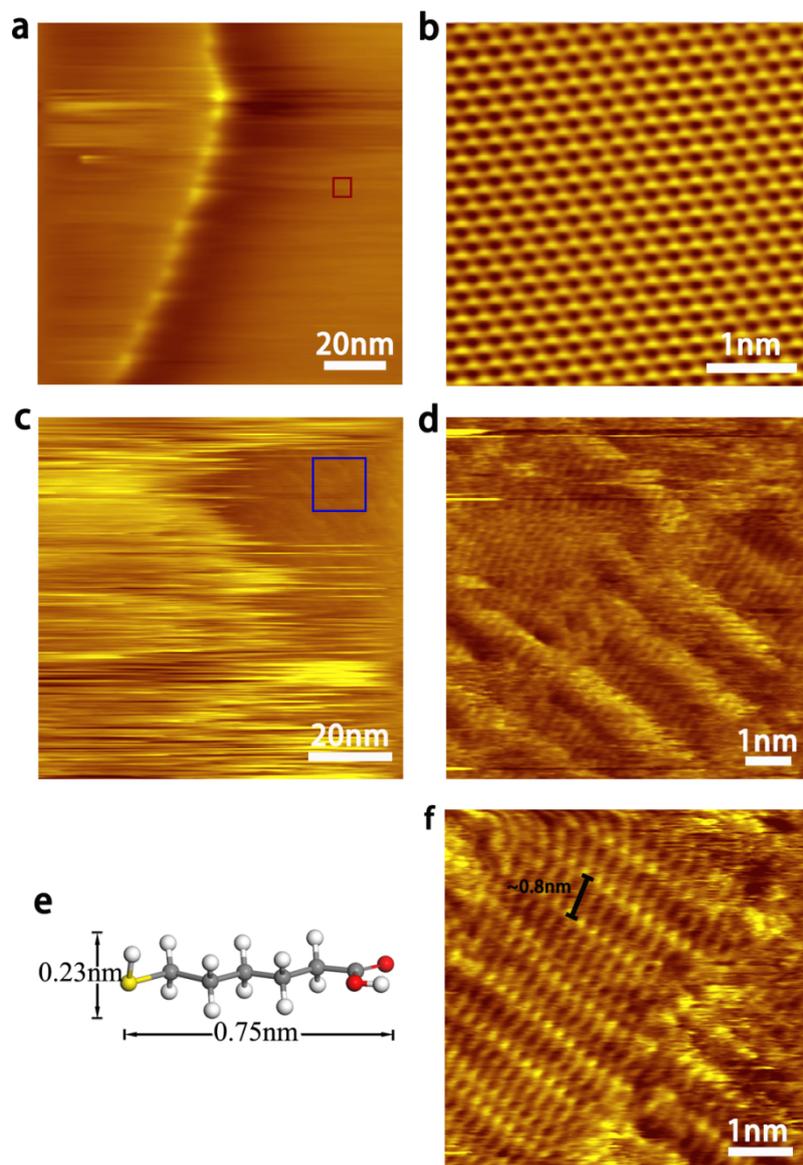


Figure S1 (a) A large area STM image of the HOPG surface treated by the organic molecules. (b) Atomic resolution zoom-in image from the red frame shown in panel (a) demonstrate hexagonal lattice structure of graphene. It shows no organic molecules cover the surface. (c) A typical STM image of the same sample as (a) but with the rough surface topography. (d) A zoom-in image from the blue frame shown in panel (c). (e) Molecular structure of $C_6H_{12}O_2S$. (f) A STM image obtained from a region near to that of (d). This image shows the molecular arrangement of the thiol on HOPG. All of above STM images were measured in ultra-high vacuum chamber with liquid nitrogen temperature.

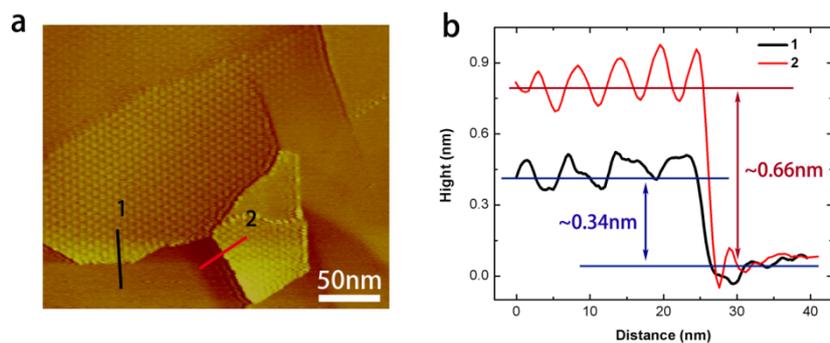


Figure S2 (a) STM image shown in Figure 2. (b) Profiles along the section lines 1 and 2 shown in panel a.

Figure S2 (a) shows a STM topological image of the folded graphene sheet. The height of the original graphene step is about 0.34 nm, displayed in Figure S1(b), which corresponds to one graphite interlayer distance. The height difference between the topmost layer and the third layer shown by the profile line 2 is around 0.66 nm, demonstrating that a graphene domain is folded onto the original sheet induced by the STM tip.