

Large-scale formation of single-crystal one-third-hydrogenated graphene with anisotropic electronic properties

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It is generally thought that hydrogen atoms can chemisorb on graphene in a perfect periodic manner forming new crystalline two-dimensional (2D) materials that exhibit unique electronic properties beyond pristine graphene, such as graphane (fully hydrogenated graphene), graphone (half hydrogenated graphene) and 2D C_xH_y (other partially hydrogenated graphene). So far, samples with small patches of periodically-hydrogenated graphene can only be produced. In previous works, we have fabricated highly ordered, millimeter-scale, continuous single-crystalline graphene monolayer on Ru(0001) [1, 2], which provides a template for selective adsorption of atoms [3,4] or molecules [5]. Here, we use graphene/Ru(0001) as a template for hydrogenation. We present atomically resolved scanning tunneling microscope (STM) images and low-energy electron diffraction (LEED) patterns that demonstrate the fabrication of periodically-hydrogenated graphene that is perfectly ordered over 4 millimeters. The as-fabricated hydrogenated graphene is highly ordered, with a $\sqrt{3} \times \sqrt{3}/R30^\circ$ period relative to the pristine graphene. As the ratio of hydrogen and carbon is 1:3, the periodically-hydrogenated graphene is named “one-third-hydrogenated graphene”(OTHG). Moreover, we show that, the OTHG is weakly coupled from the substrate and structurally anisotropic. Calculations show that the structural anisotropy of one-third hydrogenation results in an anisotropic electronic structure. This work provides an efficient method to produce large-scale crystalline functionalized graphene with specially desired properties.

References

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