

BAND GAP OPENING IN GRAPHENE INDUCED BY PATTERNED HYDROGEN ADSORPTION

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Introduction

Graphene has potential to become the ultimate material for future electronic devices [1]. However, graphene lacks a band gap around the Fermi level, which limits its application for circuit fabrication. Even though, it is well known that hydrogen treatment can modify graphene electronic behavior [2-5], the modification realized in experiments so far is of a rather uncontrolled nature. At present, confinement induced by hydrogen barriers appears to be a promising way to control the extent of graphene conductivity [4-6], raising a possibility for large-scale electronic circuits production by H adsorption on a pre-defined adsorption pattern. To utilize this idea, however, selective hydrogen adsorption on graphene needs to be achieved

Experimental

Epitaxial graphene was prepared by heating an Ir(111) crystal to 550 K for 100 s under 2×10^{-5} Pa partial pressure of ethylene, followed by a flash anneal to 1420 K [7]. This cycle was repeated seven times in order to obtain a high-quality graphene layer and to achieve complete surface coverage. The quality of the graphene layer was confirmed by STM, ARPES and Low-Energy Electron Diffraction. Atomic hydrogen was deposited onto the graphene layer at room temperature by means of a 2100 K hot H-atom beam with a flux of $\sim 10^{13}$ - 10^{14} atoms/cm² s. STM images were obtained by the Aarhus STM [8] with the sample held at room temperature.

ARPES data were recorded on the SGM-3 beamline at ASTRID synchrotron at the Institute for Storage Ring Facilities at Aarhus University [9]. The sample was kept at 100K while collecting the data sets. Determination of possible gap openings around the K point involve the potential risk of error due to small sample misalignments. To ensure that this is not the case, ARPES data sets were actually not taken as two-dimensional cuts in k-space, but as three dimensional maps with the photoemission intensity measured as a function of binding energy and a two-dimensional k-vector parallel to the surface.

Results and Discussion

Here we present investigations on atomic hydrogen interaction with graphene on Ir(111) by means of Scanning Tunneling Microscopy (STM) and Angle-Resolved Ultraviolet Photoelectron Spectroscopy (ARUPS) [10]. STM measurements reveal spatial selectivity for hydrogen adsorption onto the graphene/Ir(111) moiré pattern, Fig. 1. This preferential hydrogen adsorption leads to a structural and electronic modulation of the graphene sheet with the periodicity of the moiré superstructure.

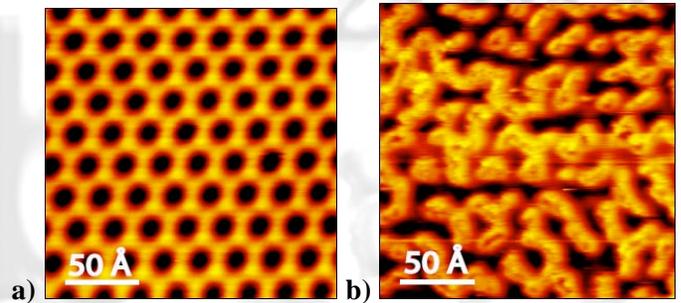


Fig. 1 a) STM image of clean graphene on Ir(111) showing the moiré superstructure b) after exposure to 30s of hot atomic hydrogen dose. The hydrogenation occurs along the bright parts of the moiré depicted in a).

From the DFT calculations we have found that the preferential hydrogen adsorption to certain moiré regions can be explained by the formation of graphane-like clusters. In these clusters carbon with attached hydrogen is pulled out of the surface, while the neighboring carbon is pulled down towards the substrate and forms a bond with Ir. This occurs alternatively for all carbons within the graphene hexagon thus forming a graphane- or diamond-like configuration. These graphane-like clusters represent a large barrier for the electron wave-function giving rise to a confinement effect and consequent bandgap opening equivalent to that known for antidot lattices [11]. The electronic band dispersion of such periodically modulated graphene is probed around the Dirac point using the ARUPS technique. The photoemission data,

indeed, confirm the appearance of a gap opening at the Fermi level. Moreover, we have found that the size of the gap is

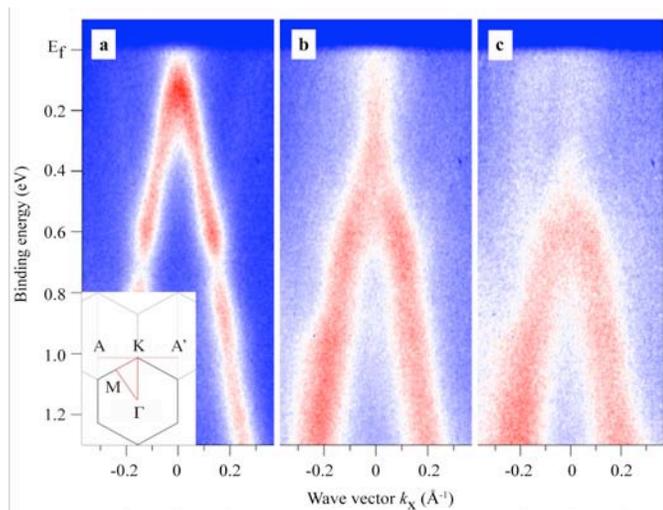


Fig 2 Observation of a gap opening in hydrogenated graphene: photoemission intensity along AKA' direction of the Brillouin zone (see inset) for a) clean graphene on Ir(111). b) graphene exposed to a 30 s dose of atomic hydrogen and c) graphene exposed to 50 s dose of atomic hydrogen.

coverage dependent and reaches a well-defined value after uniform hydrogen coverage is achieved, see Fig. 2c. Our experimental results thus validate the concept of band gap opening in graphene through patterned hydrogen adsorption.

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