Graphene on the reconstructed Pt(100) surface and its interaction with atomic hydrogen

L. Nilsson, M. Andersen, R. Balog, J. Bjerre, E. Lægsgaard, B. Hammer, F. Besenbacher, I. Stensgaard and L. Hornekær.

Interdisciplinary Nanoscience Center and Department of Physics and Astronomy, University of Aarhus, Denmark
Louis@inano.au.dk

Introduction

Despite an enormous development for graphene fabrication within the last eight years, the production of high quality, large scale graphene by cost efficient routes needs further improvements to meet industry standards [1]. A fundamental understanding of the growth of graphene is therefore of utmost importance. Here, we demonstrate, from an interplay between STM and DFT calculations, that a graphene sheet can be grown continuously on the hex-reconstructed Pt(100) surface even across step edges and domain boundaries in the platinum substrate [2].

To address the challenge of opening a band gap in graphene, we have recently presented results demonstrating a band gap opening in graphene on Ir(111) by patterned hydrogen adsorption [3]. The resulting patterned adsorption structure was facilitated by the interaction between the iridium substrate and the graphene sheet. To obtain a better understanding of the role of the substrate, we investigate the interaction between hydrogen atoms and graphene grown on the reconstructed Pt(100) surface by STM and temperature programmed desorption (TPD) experiments combined with DFT calculations.

Techniques

STM measurements were performed using the so-called Aarhus STM. The Pt (100) surface was cleaned by numerous cycles of 2 keV Ne sputtering and annealing up to 1000°C, combined with annealing in O_2 at 700°C, followed by flashes to 900°C. The cleanliness of the surface was checked with STM. Synthesis of graphene was typically carried out by exposing the Pt surface to 100-200 L of either ethylene or propylene (C_3H_6) at pressures in the low 10^{-7} torr range and a sample temperature of 700°C, with periodic flashes to 900°C. A commercial “Hydrogen Atomic Beam Source, HABS-40” was used to expose the surface to thermally cracked atomic hydrogen atoms.
The DFT calculations regarding rotated sheets of graphene on a Pt(111) surface were performed with the semi-local meta-GGA density functional M06-L implemented in the real-space projector augmented wave GPAW code.
The calculations regarding edge dislocation lines in graphene involved supercells with hundreds of atoms and were therefore (for reasons of computational efficiency) performed with the Siesta code. Exchange and correlation effects were described using the GGA functional of Perdew, Burke and Ernzerhof (PBE).

Results and Discussion

A continuous graphene sheet has been grown on the well-known Pt(100)-hex-R0.7 reconstruction[4]. Interestingly, the reconstruction of the platinum substrate is still present after the growth of graphene (figure 1). The graphene is found to grow across step edges and domain boundaries (figure 1) and thereby forming a continuous sheet. For domain boundaries of 7 degrees, the graphene is observed to rotate by incorporating defects similar to the Stone-Wales defects [5]. From STM it is revealed that the graphene sheet tends to have either an armchair or a zigzag direction aligned with the direction of the reconstruction of the platinum surface, however, other angles are also observed. The observed energy difference between different orientations of the graphene on the substrate is accordingly found to be small from DFT calculations.

We also present a combined STM and TPD investigation of the interaction of the graphene with atomic hydrogen. At low coverage, the hydrogen atoms are observed from STM to form dimer structures similar to those observed previously on HOPG, and the desorption energy found from TPD is also similar to that found for HOPG [6]. At higher coverage the reconstruction of the platinum substrate is lifted and extended structures are depicted by STM. From TPD a distinct shift in desorption energies is observed. The experimental data indicate that the platinum substrate plays an active role in stabilizing these adsorbate structures on graphene. The experimental findings are supported by DFT calculations.

References

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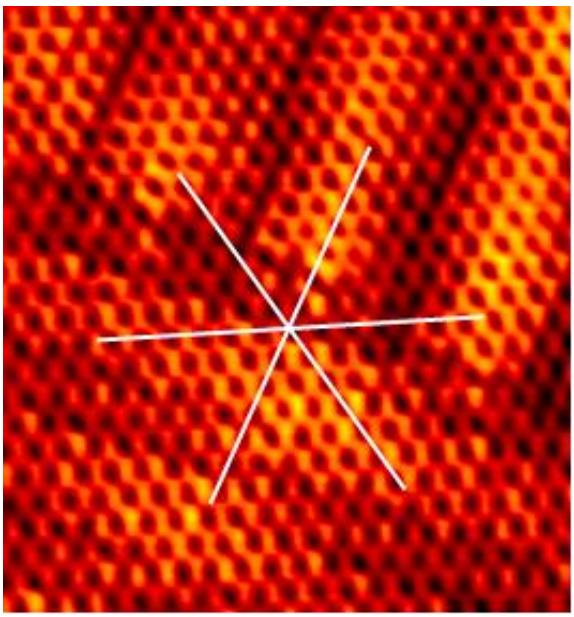
Figure 1



STM image of a graphene area with a domain boundary between two perpendicular domains of the underlying hex-reconstructed Pt(100) surface. The white lines indicate zigzag directions in the graphene.