Mountains and valleys of graphene

Understanding the electronic structure and the imaging contrast of the graphene nanomesh on metals

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Graphene superlattices, and among them graphene-based moiré structures, have attracted increased attention in recent condensed-matter studies because they can be used as a foundation for tailoring the transport properties of heterosystems. The formation of ordered commensurate moiré structures of graphene on different substrates leads to the cloning of Dirac cones in reciprocal space, which in turn modifies the electronic spectrum of carriers via the opening of minigaps around the Dirac point, deviates the energy band dispersion from the linear one, changes the effective mass and velocity of the carriers, etc. All these effects are a manifestation of the additional large-scale modulation potential originating from the moiré superlattice, which usually has a size of several nanometers.

One of the moiré lattice classes is a graphene layer on the close-packed surfaces of 4d and 5d transition metals, such as Ir(111), Rh(111), or Ru(0001) [1,2] (Fig. 1). Such graphene-metal systems are the subject of long-term surface-science studies, and they were proposed as substrates for ordered arrays of metallic clusters, which can then be used in storage technology or in catalysis as the behaviour of a cluster’s array could be modelled on the basis of a single element. The electronic or magnetic properties of such cluster arrays depend strongly on the underlying graphene-metal substrate. The crystallographic structure of these graphene-metal systems, and hence their electronic properties, are defined by the lattice mismatch of graphene and a metal surface as well as by the strength of the local interaction at the interface, leading to the observation of various moiré structures with different corrugations even for the same graphene-metal combinations [1,2]. Thus precise knowledge of the crystallographic structure of the graphene-metal system is crucial for the modelling of its electronic properties. Macroscopic diffraction experiments, such as low-energy electron diffraction (LEED), or local probe methods, such as scanning tunnelling microscopy (STM) and atomic force microscopy (AFM), can provide information about the symmetry of the system and the lattice alignment of graphene and metal [3,4]. The graphene moiré lattice corrugation on metal is extracted from a comparison of experimental and theoretical data, e.g., from the modelled STM images at different bias voltage [3] or from simulated $I(V)$ curves in LEED experiments [4]. In principle, AFM can do this job, but as was shown, the correct topography in this case is influenced by the residual electrostatic forces between the tip and the sample. In such AFM experiments, compensation of the local contact potential difference variation over the moiré lattice is not a trivial task. Also, because graphene is a very elastic material, the imaged topography in AFM experiments can be influenced by the indentation effect from the tip, which is placed in close vicinity to the surface. Therefore, the necessity of the tool, which gives full information about graphene moiré, is obvious, and our approach is based on complementary STM and AFM spectroscopy measurements in this case.

We performed an analysis of the crystallographic structure of the strongly corrugated graphene/Ru(0001) system employing a combination of state-of-the-art density-functional theory (DFT)
Figure 2: Interaction of W-tip with graphene/Ru(0001) by means of state of the art density functional theory and scanning probe microscopy and spectroscopy. (a) Frequency shift ($\Delta f$) as a function of the tip-sample distance measured at different places of graphene/Ru(0001) and the respective tip-sample interaction force ($F_z$) and energy ($E$). (b) Interaction energy ($E$) calculated at two different places of graphene/Ru(0001) and the respective tip-sample interaction force ($F_z$) as functions of the tip-sample distance ($z$). The inset shows the top view of the graphene/Ru(0001) structure (white star, hexagon, and rhombus mark ATOP, HCP, and FCC places, respectively). (c) Series of snapshots of the atomic configurations corresponding to the interaction of W-tip with graphene/Ru(0001) at the ATOP place taken for six different distances indicated with numbers 1-6 at (b). The cut is made perpendicular to the sample surface through the yellow line as shown in the inset of (b). Each structure is overlaid with the calculated difference electron density, $\rho = \rho_{\text{tip+sample}}(r) - [\rho_{\text{tip}}(r) + \rho_{\text{sample}}(r)]$. Red (blue) indicates accumulation (depletion) of the electron density.

and scanning probe microscopy and spectroscopy (STM/AFM) (Fig. 2) [5]. Our force spectroscopy data demonstrate the unexpected two-minima behaviour for the $\Delta f(z)$ and $F_z(z)$ channels for the ATOP places of the graphene/Ru(0001) structure that was successfully reproduced in the DFT calculations. We found that graphene at the ATOP places behaves like identical nano-drums (or nanoresonators) having a resonance frequency of about 1 THz and Young’s modulus, which is much smaller compared to the one for the free-standing-like graphene membranes. Our analysis also allows us to directly extract the value of the graphene corrugation from the direct comparison of the $\Delta f(z)$ curves for different places of the graphene structure, which can be used in further applications of SPM for graphene-based systems.

Recently the STM/AFM experiments were carried out for graphene on Ir(111), where similar nano-drum behaviour was observed. We intend to perform the respective DFT simulations in order to understand better properties of graphene on Ir(111) and to generalise our conclusions made previously for graphene/Ru(0001). Besides, the electronic properties of the graphene-bilayer/Ir(111) system will be studied within this project. In the latter case the topmost graphene layer is electronically decoupled from the metal substrate.

More Information


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