

Graphene under ultrahigh pressure: Local tuning of its electronic properties

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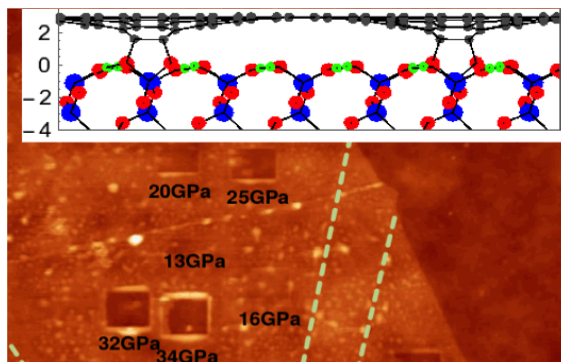
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Controlling graphene electronic properties is critical to promote its use in a variety of electronic devices. In this context, it is known, for example, that the degree of graphene coupling to the substrate is an important factor controlling graphene doping [1,2]. Very recently, it has been shown that graphene electronic properties can be locally tuned by applying ultrahigh pressures (> 10 GPa) using Atomic Force Microscopy (AFM). Graphene is modified in such a way that specific areas can be irreversibly flattened against an amorphous SiO₂ substrate, creating p-doped graphene regions. Furthermore, as proved by Raman Spectroscopy, Kelvin Probe Force Microscopy and Scanning X-ray Photoelectron Microscopy, the strength of the doping depends monotonically on the applied pressure [3]



AFM topographic image showing 600x600 nm² area modified under different pressures. Inset: chemisorbed configuration found by means of PBS-DFT simulations.

Aiming to understand these experimental findings, we have studied the binding mechanism between graphene and several SiO₂ surfaces by means of periodic boundary conditions (PBC) calculations based on density functional theory (DFT). Our results reveal that, for the four SiO₂ surfaces considered in this study, it exists a stable configuration in which the graphene is physisorbed, i.e., it is bound, by weak dispersion forces, at a distance ~3 Å. In addition, for some particular SiO₂ surfaces, we have found several configurations in which one or more C atoms are chemically bound to either the O or the Si atoms of the surface. Among these configurations, the ones retaining the honeycomb lattice are of special interest. Focusing on these latter configurations, we have run PBC-DFT calculations on coordinate path connecting the physisorbed and the chemisorbed configurations. Our simulations reveal that these two minima are connected by a pressure barrier of the order of 10-20 GPa, in agreement with experimental findings [3].

References:

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