MULTISCALE INVESTIGATION OF THE STRUCTURAL, ELECTRONIC AND TRANSPORT PROPERTIES OF PURE AND INTERCALATED GRAPHENE ON SiC SUBSTRATES

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Epitaxial graphene on silicon carbide presents a combination of characteristics that render it highly attractive for device integration: wafer-size scales and direct growth on semi-insulating substrates. Here, based on extended first-principles and semiempirical calculations we discuss the structural, electronic and transport properties of graphene grown on both the silicon and the carbon face of SiC(0001) substrates. We show the fundamental differences of the Si and the C-face reconstructions and discuss their importance for the conduction characteristics of the two systems. We moreover explore the alterations expected with hydrogen or lithium intercalation and derive statistical expressions for the quantum transport properties of the resulting quasi-freestanding graphene.

We start with a comparison of the surface reconstructions of the two faces of the SiC(0001) substrate. Si-face epitaxial graphene is characterized by the formation of a wide bandgap carbon-rich interface layer with a $6\sqrt{3} \times 6\sqrt{3}R30^o$ surface reconstruction, over which Bernel stacked graphene layers of excellent quality grow [1]. However the interface interaction pins the Fermi level of the system above the charge neutrality point of neutral graphene, giving rise to a significant n-type doping for the entire system. Moreover the $6\sqrt{3} \times 6\sqrt{3}R30^o$ reconstruction results in a significant substrate disorder below the buffer layer that can give rise to leakage currents through interface defects [2], compromising in a nontrivial way device-requested characteristics. On the other hand, growth on the C-face of SiC(0001) is less controllable with respect to the Si face but maintains a series of advantages regarding its electrical characteristics. The first carbon-rich layer here maintains a purely metallic character [3] and an important presence of π electrons, along with a non-negligible coupling with the substrate. The particularity of its geometrical configuration consists in a corrugated surface where small sp^2-bonded islands are terminated by carbon atoms that covalently bond with the substrate [Fig. 1]. Below these islands also the surface carbon atoms sp^2-hybridize, while their π bands are half-filled and present a ferromagnetic order due to the presence of strong electron exchange interactions [3]. In this case there exists no Fermi-level pinning effect, allowing for a plausible device operation starting from an off-state. Typical graphene-like characteristics (i.e. the Dirac cone) are recovered with the addition of a second graphene layer.

As a remedy for the negative influence of the buffer layer in Si-face epitaxial graphene, intercalation with lightweight group 1 elements has been proposed [4][5]. Here, starting from the $6\sqrt{3} \times 6\sqrt{3}R30^o$ reconstruction we study at an ab initio level structural and electronic properties considering hydrogen and lithium intercalation. In both cases the addition of the adatoms successfully passivates the dangling bonds of the interface and restores order in the substrate. Structurally, the buffer layer is transformed in a flat graphene sheet. However the electronic properties of these two systems significantly differ, since only in the case of hydrogen intercalation the substrate maintains a semi-insulating character. When it comes to lithium, the metallic bands originating from the Li adatoms penetrate throughout the entire SiC bandgap, giving rise to an overall metallic behavior.
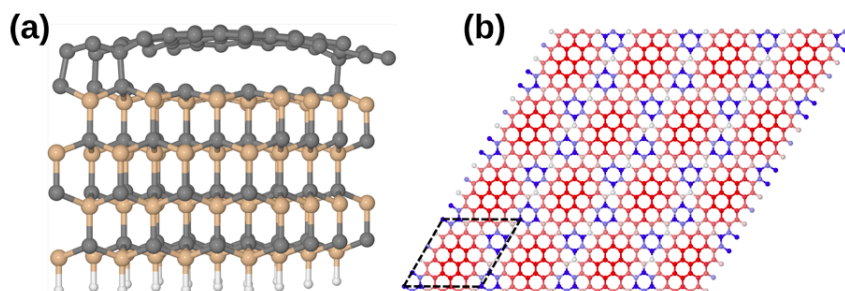

Figure 1: (a) Side view of a (5 × 5) graphene monolayer relaxed on a (4 × 4) 4H-SiC(000ø1) substrate. (b) Color map view of the first epitaxial graphene layer showing the bonding characteristics of the carbon atoms, where gradual red to blue coloring indicates sp^2 to sp^3 bonding. Dashed lines show the periodically reproduced unit cell.

As a result of a non-ideal intercalation process, sp^3-type defects can be formed in the graphene epilayer due to a covalent bonding between the carbon and the hydrogen/lithium adatoms. Within a statistical study [6] of finite defect concentrations we explore the quantum transport properties of the resulting disordered quasi-freestanding graphene in terms of transport gaps [7] and fluctuations in the conductance distribution. Computations here are based on accurately parameterized semiempirical Hamiltonians from first-principles calculations and non-equilibrium GreenÕs function techniques. Finally, we discuss how our results can be interpreted for a consistent analysis of data deriving from experimental measurements.

References

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