H coverage defects in quasi-free-standing monolayer graphene on SiC

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Quasi-free-standing monolayer graphene (QFMLG) on SiC is obtained by intercalating H atoms at the interface between the SiC(0001) substrate and the carbon-rich buffer layer obtained by Si evaporation. The procedure produces a flat and completely sp^2 hybridized honeycomb lattice [1]. However, the mobility of QFMLG is lower than in suspended graphene, indicating the existence of carrier-scattering centers, identified as Si dangling bonds without H termination at the interface. In STM images, Si dangling bonds appear as bright or dark spots, depending on bias voltage. From their size and related STS spectra, two different types of spots have been detected, one smaller and brighter (in STM images), and one larger and darker. The amount and distribution of spots varies with H intercalation temperature, appearing in some cases located on a lattice with a ~6×6 SiC periodicity [2].

In order to clarify the nature of these spots, we evaluated the structural and electronic properties of QFMLG with defects in H coverage within the Density Functional Theory (DFT) framework, using a calculation setup previously tested in similar systems [3]. For calculations, we have used two kinds of supercells: the 6√3x6√3 (~1000 atoms) including up to two vacancies per cell, and a smaller one, √31x√31, which includes a slight rotation of graphene with respect to SiC (~0.7 deg), and a single vacancy per cell, but is considerably smaller (~280 atoms). This allowed extensively evaluating and comparing a large number of vacancies with size in the range 1-13 missing H atoms and with different translations relative to the graphene lattice [4]. Calculations show that inward bending is present in correspondence of the vacant sites, whose depth and width increase with the size of the vacancy.

The analysis of the electronic structure and DoS reveals localized electronic states between graphene and SiC, with energy and intensity depending on the vacancy size. This allows interpreting the different relative brightness of observed STM features. Besides, the extensive evaluation of the vacancy energies indicates that their stability increases up to the size of ~7 vacant Hs. Comparison of simulated and measured AFM, STM and STS data [5] indicates that the two kinds of spot can be attributed to three and four missing H atoms. The most favorable location conformation and location of vacancies with respect to the graphene lattice are also revealed, shedding some light on the H intercalation/evaporation process.

References:

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