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## A NEW WIDE BAND GAP FORM OF HYDROGENATED GRAPHENE

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Single-layer graphene is a very promising material for future silicon-free nanoelectronics due to the peculiar properties of the  $p$  electrons, that give rise to the Dirac cones. A number of controlled techniques for energy band engineering have been proposed other than the actively pursued goal to obtain nanoribbons of controlled size and edge geometry. Among these adsorption of hydrogen atoms or other species, as well as substitution of carbon atoms with foreign species.

It has been recently shown that upon exposure to cold hydrogen plasma, H atoms stick to graphene to form pairs[1] and clusters[2], up to eventually reach complete saturation[3]. When complete hydrogenation on both graphene sides is achieved, one gets the so-called *graphane* [4], a structure with no  $\pi$  electrons left, hence a wide band gap insulator.

Instead of reaching full hydrogenation it has been proposed that few symmetrically arranged adsorbates, or more generally  $\pi$  defects, modify the bands responsible for the Dirac cones in graphene and open a wide gap in the band structure [5]. Based on this, we now propose a new form of partially hydrogenated graphene in which H atoms lay in *para* position to each other (see Figure 1), and form a honeycomb-shaped superlattice. This arrangement is shown to be favoured by progressive preferential sticking events in *para* positions[6], induced by the synergistic effect of graphene's aromatic nature and lattice distortions. With the help of DFT calculations we find this structure to be thermodynamically stable in ordinary laboratory conditions where hydrogen atoms are employed as H sources. Compared to graphane synthesis, this structure does not need plasma exposure on both graphene's faces, hence it might be obtained from supported graphene substrates.

The structure of Figure 1 shows disconnected patches of (aromatic) graphene, i.e., benzene-like rings in a saturated hydrocarbon matrix. Its electronic structure, as computed at the DFT level of theory and supported by correlated GW calculations, is shown in Figure 2.

Despite the hydrogen coverage is only 25%, this new material shows a band gap 5.2 eV wide - surprisingly close to that found in graphane (5.4 eV [7]) - as a consequence of the joint effect of quantum confinement and symmetry. Valence and conduction band edges at G (see Figure 3) resemble (weakly coupled) Highest Occupied Lowest Unoccupied molecular orbitals in benzene supporting long-lived localized excitations. This suggests that the proposed structure may also form an interesting substrate for experimenting quantum computing applications.

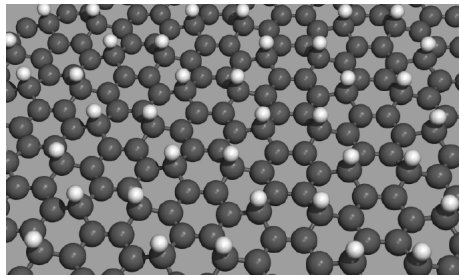


Figure 1: Lattice structure and unit cell of the partially hydrogenated graphene structure proposed.

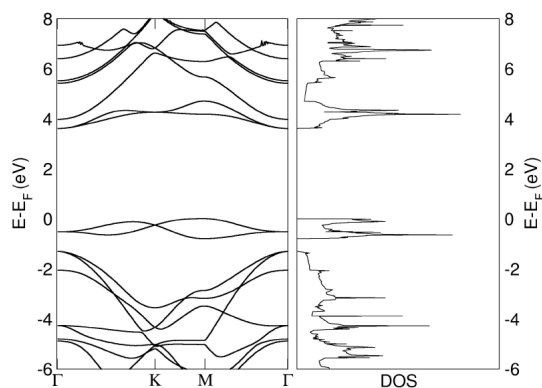


Figure 2: Band structure (left) and density of states (DOS, right) for the partially hydrogenated graphene structure as computed at the DFT-GGA level.

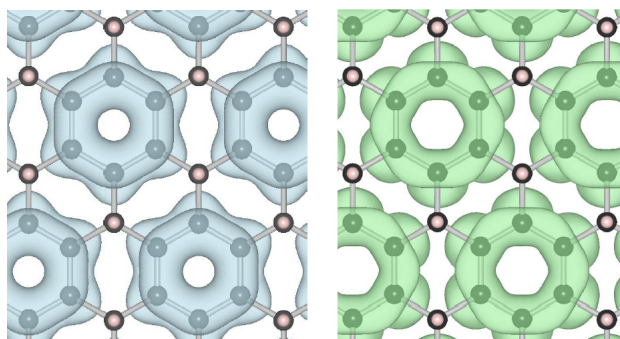


Figure 3: Isosurface of VBM (left) and CBM (right) taken at  $\Gamma$ .

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