Activation of graphenic carbon through doping : A First principles study

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Boron and nitrogen doped graphene and carbon nanotubes are being increasingly explored as electro-catalysts particularly for the oxygen reduction reactions (ORR) which are central to fuel-cells. Independently they have also been reported to chemisorb each other, which is relevant to device fabrication as well as reinforcement of their mechanical endurance. Covering these two aspects, we reveal from first-principles, a new paradigm of activation of graphenic carbon, as a result of interplay between strain, inter-sub-lattice magnetic ordering and charge-transfer processes, initiated upon doping by boron or nitrogen in experimentally observed configurations. Our results indicate that substitutional doping can non-trivially alter the balance between pi-conjugation and the on-site Coulomb repulsion which are the two fundamental competing interactions inherent to graphene that determines its ground state physical and chemical properties. Similar mechanisms are proposed to be responsible as well for activation of carbon atoms doped in hexagonal boron-nitride in energetically favourable configurations.