Open boundary condition-based modelling of disordered materials: the impact of the periodicity assumption in doped graphene

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Simulations accelerate the discovery and allow the deep understanding of materials. Periodic boundary conditions are used in basically all material simulations, in spite of the fact they are hard to realize experimentally. We apply the recursive open boundary and interfaces (ROBIN) method [1], i.e. the first method that extends the open boundary self-energies of the nonequilibrium Green's function (NEGF) method [2] to material and interface modelling. Since recursive NEGF methods significantly limit the computational costs, ROBIN allows to discretize millions of atoms in real space. In this way periodic, regular, or random distributions of atoms can be modelled explicitly. The periodicity assumption in modelling materials is assessed in detail with silicon dopants in graphene. Graphene is confirmed to produce a band gap with periodic substitution of 3% carbon with silicon in agreement with published periodic boundary condition calculations [3]. 3% randomly distributed silicon in graphene, however, only shifts the Dirac cone of graphene. The predicted shift agrees quantitatively with published experimental data [3]. Assuming periodicity elevates a small perturbation of a periodic cell into a strong impact on the material property prediction. Periodic boundary conditions can be applied on truly periodic systems only. More general systems should apply an open boundary method for reliable predictions.

References

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Figures

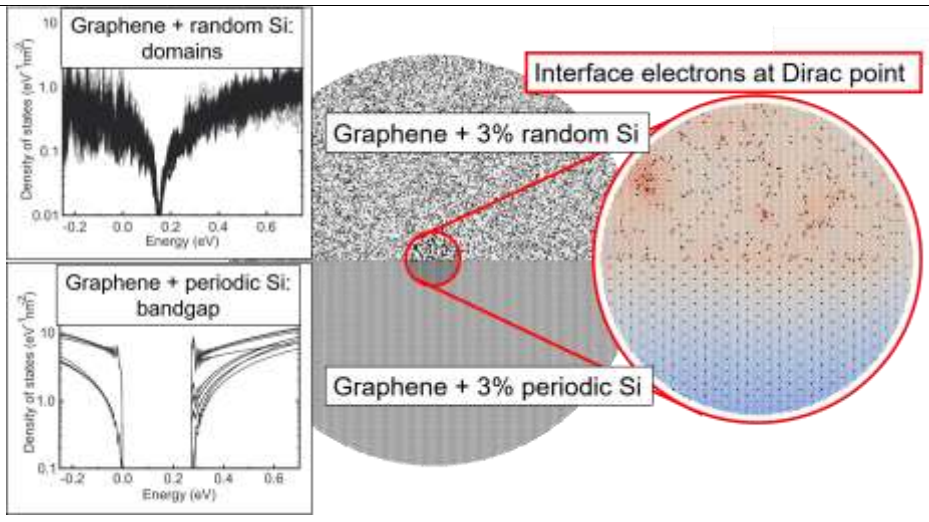

Figure 1: 200nm disc of graphene (carbon atoms are white) with 3% Si atoms (black) distributed randomly on the top, and periodically on the bottom of the disc. (right) Electronic density of states (DOS) of the centre 25 nm of the graphene disc solved with ROBIN at 10meV above the Dirac point of graphene. Carbon atoms are coloured according to the electronic DOS, silicon atoms are black. (upper left) The DOS solved in the ROBIN method of randomly distributed Si atoms in graphene does not show a bandgap. (lower left) Instead, the DOS of graphene with 3% periodically distributed silicon reproduces the 0.28eV band gap of Ref. [3].