MAGNETIC MOLECULES DERIVED FROM HYDROGENATION OF POLYCYCLIC AROMATIC HYDROCARBONS

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The quest for spin polarized organic molecules is since long attracting a great deal of attention. Actual routes to produce magnetic organic-based materials commonly adopt a similar strategy: the addition of magnetic species (atoms, polyradicals, etc.). Here we explore an alternative approach that consists of hydrogenation of Polycyclic Aromatic Hydrocarbons (PAH). The proposal is illustrated by carrying out extensive mono- (Hartree Fock and DFT) and multi-determinantal (CISD and MCSCF) calculations on hexa-hydrogenated coronene $C_{24}H_{18}$ and hexahydrogenated corannulene $C_{20}H_{16}$ (see Fig. 1). The results indicate that high spin species (S=3 or S=2) are stable, its formation being favorable provided that the reaction involves atomic hydrogen. An attempt is made to rationalize these results in terms of Lieb's theorem for bipartite lattices [1], which states that the spin multiplicity of their ground states is |NA − NB | + 1, NA and NB being the number of atoms in each sublattice. C atoms in most PAHs, corannulene being an exception, actually form clusters of a bipartite lattice, i.e., graphene (such PAH are also known as alternant hydrocarbons). The spin of the ground state of hydrogenated coronene is in accordance with Lieb's theorem (S=3 in one of the molecules) in spite of the significantly different topologies of ab initio and Lieb's Hamiltonians. This is somehow compatible with results showing that spin states of PAH (corannulene included) (benzene, anthracene and twelve-fold hydogenated coronene) can be nicely fitted by means of Lieb (or Hubbard) Hamiltonian with a Coulomb repulsion of U = 3.3 eV and a hopping of t = 2.71 eV (charge states, however, cannot be fit with this Hamiltonian). Such a low value of U/t gives, notwithstanding, significant antiferromagnetic correlations in the molecules. Based on the strong antiferromagnetic correlation obtained in

PAHs, and in order to handle frustrated topologies, we suggest an extension of Lieb's theorem. ***According to the proposed alternating rule, the number of AB pairs should be maximized in order to optimize energy.*** On the other hand, such simple interacting models cannot describe the energies of the charged molecules; this suggests the need of a critical

References:

[1] E.H. Lieb, Phys. Rev. Lett. **62**, 1201 (1989).
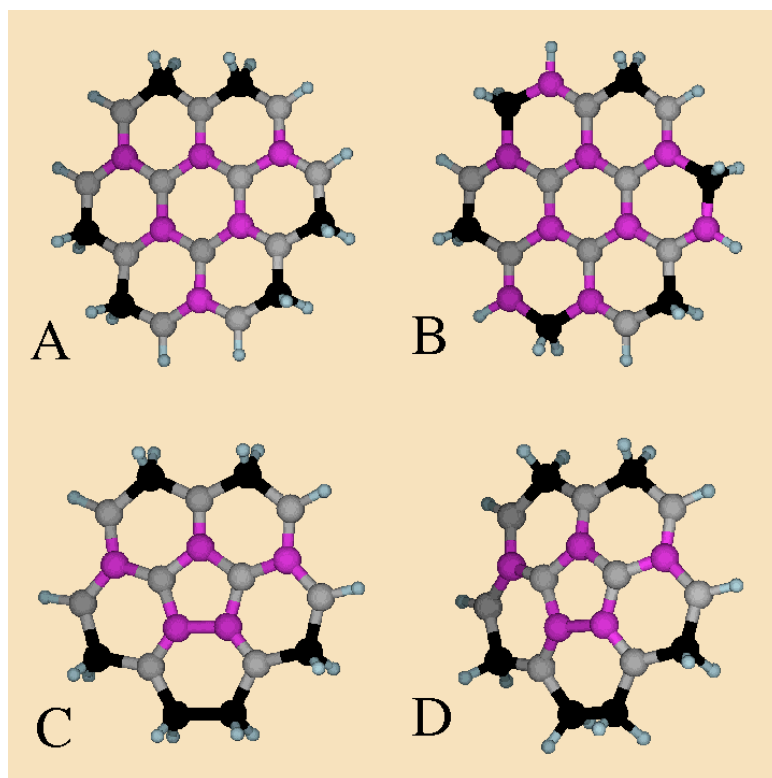
Fig. 1: 1,4,5,8,9,12-hexahydrocoronene (A, hereafter referred to as D_{3h} according to its symmetry group), 1,3,5,7,9,11-hexahydrocoronene (B, hereafter referred to as C_{3h}) and 1,4,5,6,7,10-hexahydrocorannulene (C planar and D curved, hereafter referred to as C_{2v}). Saturated carbons are represented by black symbols while dark grey (magenta) and light grey symbols correspond to carbons in one of the two sublattices. Note that corannulene is a non-alternant hydrocarbon, or, alternatively, a frustrated cluster of carbon atoms (note the fully magenta bond between two magenta atoms)