

MAGNETISM OF NANOCARBONS: FULLERENE C₆₀ AND GRAPHENE

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Odd electrons are a characteristic feature of nanocarbons. The term naturally covers terms “ π electrons”, “magnetic electrons”, and “dangling bonds” [1] and indicates that the number of valence electrons of each carbon atom of the species is larger by one than that of interatomic bonds formed by the atom. Due to enlarging length of the valence bonds of the species in comparison to the ones of classic π electron systems such as benzene or ethylene molecules a considerable weakening of the electron interaction occurs which causes a partial exclusion of odd electrons from the covalent bonding so that odd electrons, covalently bound in the classic molecules, become effectively unpaired in more complicated nanocarbons. These effectively unpaired electrons provide a partial radicalization of the species which results in a considerable enhancement of their chemical reactivity and magnetism.

Molecular magnetism of odd-electron systems is considered in terms of the Heisenberg Hamiltonian via the total spin S and the exchange integral J or the magnetic coupling constant. The constant determination is the central point of the magnetism study that crucially depends on the appropriate mapping between the Heisenberg spin eigenstates and the computationally determined electronic states.

Once being a perfect alternative to many-body configurational interaction, one-determinant unrestricted broken-symmetry Hartree-Fock approach applied to weakly interacting odd electrons provides a reliable determination of both pure-spin state energies and the relevant J value from the spin-contaminated eigenvalues of the UBS HF solutions [2]. In the case of even

number of “magnetic” (odd) electrons J is determined as $J = \frac{E_{S=0}^{UBS HF} - E_{S_{max}}^{PS}}{S_{max}^2}$. Here

$E_{S=0}^{UBS HF}$ and $E_{S_{max}}^{PS}$ are the energy of the UBS HF singlet state and the pure-spin state with maximal spin S_{max} that is the exact pure-spin single-determinant solution, respectively. Oppositely to the case, both magnetic coupling constant J and pure-spin states cannot be straightforwardly obtained within the one-determinant UBS DFT scope. Usually, additional particular procedures are needed to reach the goal.

The magnetization proceeds when J absolute value is small. This is of a particular importance for the systems with the singlet ground state due to the second-order character of the magnetic phenomenon in this case [3]. The performed UBS HF computations revealed that magnetic response of singlet nanocarbons was provided by a collective action of odd electrons and was size-dependent. The above criterion concerning the smallness of the J value is met when fullerene molecules are joined in carpet oligomers of a few nm in linear size [4] and when graphene is split into nm -size nanopanes [5]. The size-dependent origin of magnetism is well supported by experimental findings related to nanostructured polymerized fullerene crystals and activated carbon fibers consisting of nanographite domains. The explanation of disappearing magnetic response in single polymerized fullerene crystals and micro-size graphene is suggested.

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