Theoretical Design of High-Spin Polycyclic Hydrocarbon Materials

Nicolas Suaud, Georges Trinquier, Jean-Paul Malrieu, Nathalie Guihéry

Laboratoire de Chimie et Physique Quantiques, Université Paul Sabatier, 118 route de Narbonne, Toulouse, France
suaud@irsamc.ups-tlse.fr

This work addresses the conception of purely-organic magnetic materials. A first part is dedicated to the conception of high-spin organic building blocks obtained from fused polycyclic hydrocarbons by converting selected HC(sp^2) sites into H_2C(sp^3) ones, guided by Ovchinnikov's rule. Some molecules involving three to twelve fused six-member carbon rings are presented. Unrestricted DFT calculations, including geometry optimizations, confirm the high-spin multiplicity of ground states. Spin-density distributions and low-energy spectra are further studied through geometry-dependent Heisenberg–Hamiltonian diagonalizations and explicit correlated *ab initio* treatments (DDCI), which all agree on the high-spin character of the suggested structures, and locate the low-lying states at significantly higher energies. In particular, the lowest-lying state of lower multiplicity is always found to be higher than kT at room temperature (at least ten times higher).

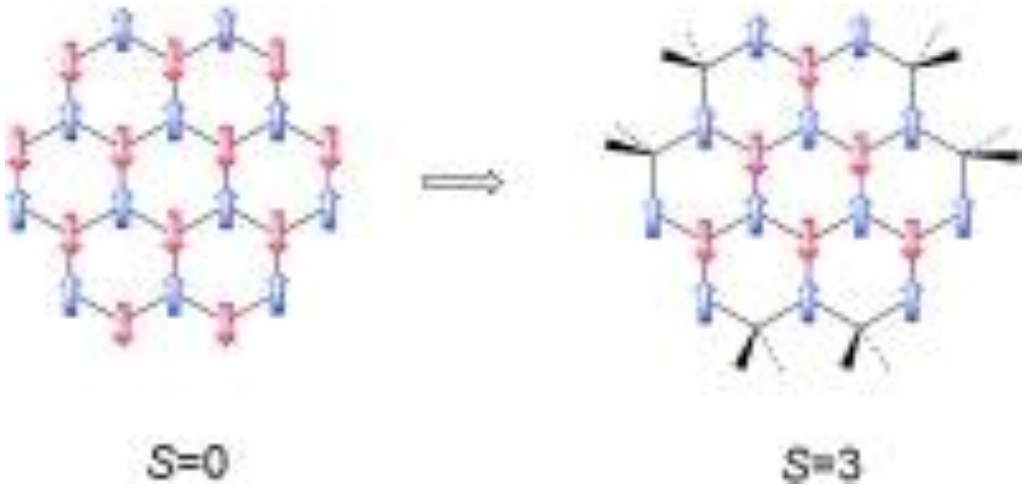
The second part is focused on the conception of ferro-, ferri- and antiferromagnetic periodic bi-dimensional lattices. For this purpose, the nature and magnitude of magnetic coupling between the high-spin units are analyzed as functions of the topology of the connection. Simple rules to predict the preferred spin ordering between the units are established, resulting from the interplay between two main physical factors, namely (i) the spin-polarization of the bridge, and (ii) the M_s-spin component of the connecting carbons on each magnetic unit. The relevance of these rules is confirmed by UDFT calculations performed on bi-unit fragments.

References

[1] Georges Trinquier, Nicolas Suaud, Jean-Paul Malrieu, Chem. Euro. J, **29** (2010) 8762-8772.
[2] Georges Trinquier, Nicolas Suaud, Nathalie Guihéry, Jean-Paul Malrieu, to be submitted.

Figures

Smart



saturation*: Saturating properly chosen vertices of polycyclic conjugated hydrocarbons may lead to high-spin organic units*