Electronic structure of the highly entangled polyradical nanographene with coexisting strong correlation and topological frustration

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In the recent work, [1] we have studied a highly entangled polyradical nanographene, which features multiple strongly correlated spins arising from the interplay of strong electron - electron correlation and topological frustration. The system involves fusing four [3]triangulene motifs onto the edges of a [3]rhombene to create a butterfly-shaped nanographene with a carefully crafted geometry that renders both sublattices (A and B) topologically frustrated. Additionally, this design creates a sufficiently large size (the largest size of fully fused open-shell nanographene by far) to trigger spin-symmetry breaking of occupied frontier orbitals through strong e-e interaction that dominates over the hybridization energy, yielding two more radicals. Combining these two magnetic origins results in a tetraradical nanographene with both ferromagnetic and antiferromagnetic coupling of correlated spins that is rarely found in fully aromatic PAHs possessing only a single magnetic origin. The analysis of the electronic structure was performed at the CASSCF/DLPNO-NEVPT2 level.

[1] Highly-Entangled Polyradical Nanographene with Coexisting Strong Correlation and Topological Frustration, S. Song, A. Pinar Solé, A. Matěj, G. Li, O. Stetsovych, D. Soler, H. Yang, M. Telychko, J. Li, M. Kumar, Q. Chen, S. Edalatmanesh, J. Brabec, L. Veis, J. Wu, P. Jelinek, J. Lu, 2023, accepted in Nature Chemistry