

Quantum-Chemistry-Enabled Molecular Design of Functional Nonbenzenoid Nanographenes: From π -Magnetism to Chiroptical Response

Author:

Junting Wang

Theoretical calculations and emerging machine-learning approaches have become essential for the molecular design of functional nanographenes, as even subtle shifts in topology, local aromaticity, curvature, and π -conjugation can profoundly alter their electronic, magnetic, and chiroptical properties. For nonbenzenoid systems in particular, quantum chemistry provides a crucial tool for interpreting complex experimental observations and establishing reliable structure–property relationships. This contribution outlines a series of high-performance computing (NHR/HPC) driven investigations into synthetic nonbenzenoid nanographenes and related carbon nanostructures. We employ a comprehensive computational workflow that integrates geometry optimization, open-shell spin-state analysis, spin-density mapping, aromaticity evaluation (NICS/ACID), and excited-state modeling via TD-DFT/ECD. Applying this toolkit to open-shell and π -magnetic systems—including concealed non-Kekulé molecules and structurally precise chiral nanographenes—enabled us to accurately rationalize their singlet–triplet energetics and spin-density distributions. We also extended this approach to highly defective curved nanographenes and asymmetric helical structures, uncovering the theoretical mechanisms behind their bandgap modulation, local antiaromaticity, and unique chiroptical responses. In summary, these investigations illustrate how chirality, conformation, and nonbenzenoid topology govern the interplay of π -magnetism and chiroptical behavior in carbon nanostructures. The resulting computational insights provide a clear pathway for the predictive structural design of novel nanographenes targeted at molecular magnetism, chiral optoelectronics, and next-generation spintronic devices.