

Carbon fullerenes from first-principles many-body theories

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We investigate the accuracy of first-principles many-body theories at the nanoscale by comparing the low energy excitations of the carbon fullerenes C₂₀, C₂₄, C₅₀, C₆₀, C₇₀, and C₈₀ with experiment. Properties are calculated via the GW-Bethe-Salpeter Equation (GW-BSE) and diffusion Quantum Monte Carlo (QMC) methods. We critically compare these theories and assess their accuracy against available photoabsorption and photoelectron spectroscopy data. The first ionization potentials are consistently well reproduced and are similar for all the fullerenes and methods studied. The electron affinities and first triplet excitation energies show substantial method and geometry dependence. These results establish the validity of many-body theories as viable alternative to density-functional theory in describing electronic properties of confined carbon nanostructures. The comparison with experiments suggest that nodal errors in DMC might hinder the calculations of excitation energies in larger systems. Perspectives for the applications of Quantum Monte Carlo methods in nanostructures are discussed.