New theoretical tools for modeling nonlinear optical properties in closed- and open-shell systems.

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Design of novel organic materials with enhanced/tunable nonlinear response impacts many applications in chemical, biological, and materials sciences. Traditionally, design of nonlinear materials has focused on closed-shell motifs. Open-shell systems are gaining attention as candidates with enhanced nonlinear response. However, the role of electronic structure of open-shell species in their nonlinear response is unclear. We will present robust methods[1,2] for calculating two-photon absorption spectra and nonlinear properties such as excited-state dipole polarizabilities within the equation-of-motion coupled-cluster singles and doubles (EOM-CCSD) framework, which can accurately describe the electronic structure of a variety of closed- and open-shell systems. Results for prototypical closed- and open-shell organic molecules will be presented. We will also present theoretical tools that help in interpreting these results and establishing structure-property relationships for these nonlinear properties.

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References:

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