Calculating Non-linear Properties of Closed- and Open-shell Species with EOM-CCSD: Theory and Examples

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Design of novel materials with enhanced non-linear response impacts many applications in chemical, biological, and materials sciences. Traditionally, design of non-linear materials has focused on closed-shell chemical systems. Open-shell systems are now gaining attention as candidates with enhanced non-linear response. However, the role of electronic structure of open-shell species in their non-linear response is unclear. We will present robust implementations for predicting various non-linear properties such as multiphoton cross sections and excited-state (hyper-)polarizabilities within the framework of the equation-of-motion coupled cluster singles and doubles (EOM-CCSD), which can efficiently describe the electronic structure of a variety of closed- and open-shell systems. Results for prototype closed- and open-shell species will be presented. The dependence of the non-linear properties on the open-shell character of the electronic structure will be discussed.