Excited State Absorption: Benchmarking Quadratic Response TD-DFT against Higher Levels of Theory

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ABSTRACT

During excited-state absorption (ESA), an electronic transition between two excited states (ESs) occurs. ESA is a crucial phenomenon for probing and understanding light-matter interactions. ESA is an essential process in, e.g., organic optoelectronics, such as solar cells or lasers, and optical power limiting devices [1]. Accurate and computationally attainable ESA data are thus increasingly sought after. Quadratic-response TD-DFT (QR-TDDFT) is a promising method which can be used to obtain ESA properties at a low computational price [1, 2]. Here, we present a dataset of oscillator strengths (f), in three different gauges, and vertical excitation energies of electronic transitions between the ESs of 27 small and medium molecules from the QUEST database [3]. The reference values were obtained solely within the quadratic-response CC3 function formalism using a total of 8 Dunning correlation consistent basis sets. This allows to assess the performance of QR-TDDFT performed with both global and range-separated hybrids, as well as of lower-order wave function methods, e.g., CCSD, CC2, and ADC(2).

References

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