Excited-state coupled-cluster frozen-density embedding for large systems

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Supermolecular calculations for systems containing an increased number of molecules suffer from two main drawbacks. These are, first, the non-linear and often steep scaling with system size and, second, a complex analysis due to delocalized orbitals or response vectors. These two drawbacks can be circumvented using frozen-density embedding (FDE) [1], which partitions the supersystem into the individual molecules and has been found to be a versatile tool to study molecules in complex environments.
Combining FDE with coupled-cluster methods can be achieved in a rigorous manner when using the Lagrangian approach, enabling an efficient treatment of excited-state response properties such as orbital-relaxed dipole moments and analytical gradients for coupled-cluster methods embedded in a density-functional theory (DFT) environment. Finally, the accuracy and efficiency is illustrated using biological model systems containing about 30 molecules consisting of about 360 atoms in total, for which orbital-relaxed excited-state properties can be computed on standard compute nodes using coupled cluster FDE [2,3].

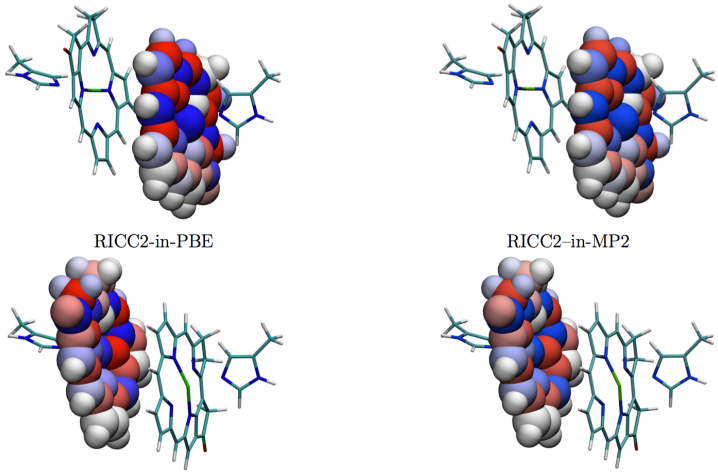

Figure 1: Orbital-relaxed excited-state atomic charges computed using FDE.

References

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3. J. Heuser, S. Höfener, *J. Chem. Theory Comput.* **14**, 4616 (2018).