The Spin-Flip Bethe-Salpeter Equation approach, and applications to molecules and defects in solids

Description of the ground- and excited-state energies for open-shell systems is quite challenging for electronic structure methods, as these systems are poorly described by single-reference electronic-structure methods. The "spin flip" approach allows such methods to describe open-shell states as an excitation -- up or down in energy -- from a related single-reference high-spin state. Spin-flip time-dependent density-functional theory (TDDFT) has shown moderate successes in describing their energies. The GW/Bethe-Salpeter equations have a similar form to TDDFT, but provide an *ab initio* kernel that overcomes many problems of standard TDDFT approximations. We have implemented spin-flip Bethe-Salpeter, allowing more accurate calculations on molecules, and enabling spin-flip for extended systems such as defects in solids for quantum information. We consider specifically ethylene and the diamond NV^- center.