

# Two-Component Spin-Orbit Coupled-Cluster Methods using Mean-Field Spin-Orbit Integrals

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A novel implementation of the two-component spin-orbit (SO) coupled-cluster singles and doubles (CCSD) method and the CCSD augmented with the perturbative inclusion of triple excitations (CCSD(T)) method using mean-field SO integrals is presented. The new formulation of SO-CCSD(T) features an atomic-orbital based algorithm for the particle-particle ladder term in the CCSD equation, which not only removes the computational bottleneck associated with the large molecular-orbital integral file, but also accelerates the evaluation of the particle-particle ladder term by around a factor of 4 by taking advantage of the spin-free nature of the instantaneous electron-electron Coulomb interaction. Benchmark calculations of the SO splittings for the thallium atom and a set of diatomic radicals as well as of the bond lengths and harmonic frequencies for a set of closed-shell diatomic molecules are presented. The basis-set and core-correlation effects in the calculations of these properties has been carefully analyzed.