Performance of the Two-Determinant Coupled-Cluster Method for Triplet and Open-Shell Singlet States of Silicon-Carbide Clusters

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We study the performance of the two-determinant (TD) coupled-cluster (CC) method, which, unlike conventional ground-state single-reference (SR) CC methods, can provide a naturally spin-adapted treatment of open-shell (low-spin) singlet and high-spin triplet electronic states. Comparisons are made with the results of equation-of-motion CC and, where appropriate, SR-CC calculations performed on topical silicon-carbide clusters. Alternative choices for reference orbitals are considered, as is a Brueckner variant. We also investigate the quality of TD-CC natural orbitals.