Modelling metallic solids with coupled cluster theory beyond CCSD

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In silico modelling of materials promises the potential of a fast, safe, and reliable route to materials design. In recent years, highly accurate wavefunction methods, such as coupled cluster theory, which were traditionally developed for molecules, have been developed and tested for solids. Coupled cluster theory with perturbative triples (CCSD(T)) is known to give "chemical accuracy" and has a lower scaling ($O(N^7)$) than with full triples (CCSDT). However, due to an infrared divergence CCSD(T) cannot give meaningful results for metallic solids. We resum the divergent terms with ring diagrams, giving a new method, "ring-CCSDT", keeping the $O(N^7)$ run-time scaling of CCSD(T). ring-CCSDT therefore avoids the divergence and significantly improves upon CCSD when evaluating uniform electron gas ground state energies and bulk lithium structural properties. Besides CCSDT, we also investigated semi-empirical coupled cluster methods on metallic solids, such as distinguishable coupled cluster (DCSD and DCSDT) as well as spin-component scaled coupled cluster (SCS-CCSD, SCS-DCSD), improving upon their original coupled cluster theories.

V. A. Neufeld, T. C. Berkelbach, Phys. Rev. Lett. 131 (18), 1503 (2023).