Seniority-based Coupled Cluster Wavefunctions

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We explore exploitation of the seniority concept in the CC formalism in a more general way. For instance, pCCD is a Ω = 0 CC wavefunction that can be improved, after calculating the pair amplitudes, by incorporating S and/or T excitations (these are known as frozen pair CCD, fpCCD, schemes). But a different path, limiting from the outset the seniority sectors accessible via the excitation operators in the cluster operator, leads to a new family of CC methods that we call seniority-restricted CC (sr-CC). For instance, the sr-CCSD(0) wavefunction includes all the S excitations, but only the seniority zero doubles. Reassuringly, these new wavefunctions give great accuracy even in the strongly correlated regime yet retain the quality of traditional CC in the weakly correlated regime (while also preserving, in the worst case, the favorable scaling of the parent "standard" CC calculations).