Development and Applications of Monte Carlo Configuration Interaction

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While Full Configuration Interaction (FCI) is numerically exact it is computationally too demanding for all but the smallest of systems. Monte Carlo configuration interaction (MCCI) involves a stochastic search through the electronic Hilbert space to produce highly compact wavefunctions that are capable of representing full configuration interaction for both energies and properties, but at only a very small fraction of the state size.[1] MCCI treats static and strongly correlated systems in a *blackbox* manner, and is capable of generating diagnostics detailing the nature of a system's true correlated wavefunction.[2] The basics of the methodology will be introduced alongside details of its scope and accuracy. Examples from our recent work to be discussed will include studies of reactive potential energy surfaces,[3] static and frequency-dependent molecular electric properties,[4-5] electronic excitation energies and transition moments,[6] core-hole excited states,[7] and strongly correlated metal-metal bonding interactions.[8] Extensions of the basic methodology will also be discussed, including MCCI-PT2,[9] natural orbital approximations,[2,9,10] and application to mixed electronic/positronic systems.[11]

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