Simulating Strongly Correlated Core-Excited States With Multireference Algebraic Diagrammatic Construction Theory

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Recently, X-ray spectroscopies have become widely used tools to investigate the electronic structure and dynamics of molecules and materials. Along with the experimental advances in X-ray spectroscopies, reliable interpretation of the X-ray spectra requires simulations of core-excited states using accurate electronic structure methods. Although many quantum chemical approaches have been extended to the simulations of core-level excitations, most of these methods assume that the chemical system probed by the X-ray light is weakly-correlated, i.e. its ground-state electronic structure is well described using a single (reference) electronic configuration.

In this talk I will present our group's recent work on the development of new methods for simulating core-excited states of strongly-correlated systems within the framework of multireference algebraic diagrammatic construction theory (MR-ADC). The MR-ADC approach is naturally suited for the simulations of core-level excitations combining several attractive features: (i) low computational cost (similar to that of multireference perturbation theories), (ii) Hermitian equations, and (iii) ability to calculate excitations from all molecular orbitals, including inner-shell and core. To access the high-energy core-excited states, we employ the core-valence separation technique that has been successfully used for the simulations of X-ray spectra with other theoretical methods. I will first give a brief overview of the MR-ADC theory, outline its implementation for simulating core-level excitations, and present applications of MR-ADC to core-ionized and core-excited states of molecules with significant multireference character in the ground electronic state.