Molecular properties, spectroscopy, and photochemistry. We use linear-response coupled-cluster methods, along with other ab initio approaches, to calculate molecular multipole moments and (hyper)polarizabilities and the effect of nuclear motion on these properties. We use first-principles theories to obtain rotivibrational, electronic, and rovibronic spectra of molecules and weakly bound species. We have demonstrated that the lowest excited state of methylcobalamin should be interpreted as metal-to-ligand charge-transfer excitation and that azulene possesses the doubly excited state below the ionization threshold, which can drive multi-photon ionization experiments related to Rydberg fingerprint spectroscopy. We have provided definitive information about structural, electronic, and spectroscopic properties of several organic biradicals and small metal nanoparticles, including, for example, beryllium, magnesium, silver, and gold clusters.

Reaction mechanisms and dynamics. We performed successful computational studies for several important organic chemistry reactions, including the Cope rearrangement of 1,5-hexadiene, cycladdition of cyclopentene to ethylene, thermal stremutations of cyclopropane, and isomerization of bicyclo[1.1.0]butane to buta-1,3-diene. We carried out unprecedented coupled-cluster calculations for CuO, and Cu2O, systems, relevant to oxygen activation by metalloenzymes, for photodissociation of NO and O2, and valence systems around 56Ni and its isotopes. We are looking for the alternative approaches to accurate calculations for many-fermion systems with pair-wise interactions, including the use of cluster expansions involving two-body correlation operators to represent nearly exact many-fermion states.