Quasiparticle dynamics driving the electronic structure of molecules and solids

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Capturing the dynamics of electronic excitations in realistic systems containing more than a few electrons is one of the outstanding theoretical challenges. Dynamical quantum correlations mediate interactions and couplings between multiple excited states and represent an important driver of materials' optoelectronic characteristics. A predictive ab-initio theory is thus critical for understanding, predicting, and designing novel compounds with tailored (quantum) properties. I will discuss our approach to tackling the first-principles description of excitation dynamics using post-DFT methods in molecular and condensed systems (containing up to several thousands of electrons) and studying individual excited states, including their non-trivial interactions. Besides theoretical analyses and comparisons, I will exemplify these approaches in practical applications to quantum materials, e.g., exploring the correlated phenomena for localized moire states in twisted bilayer graphene and defect centers in diamond. Our theoretical framework uses real-time methods combined with ab-initio dynamical downfolding. Together with efficient low-scaling numerical techniques, it is generally applicable to (quantum) material science and chemistry problems and constitutes an ideal platform for simulating complex nanoscale systems, such as molecular assemblies or materials interfaces.