LEARNING MOLECULAR HAMILTONIANS DIRECTLY FROM SPECTRA

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Assigning and interpreting the spectroscopy of large molecules and clusters often requires a manual trial-and-error approach. Here, we present our efforts to automate spectroscopic assignments through a data-driven approach. We will consider two cases: vibrational spectra of cold clusters and simulated electronic spectra of conjugated molecules. Our method first assumes a local mode form of the Hamiltonian and then performs an active search through the space of physically reasonable couplings that could be present in the system (e.g., anharmonicities for vibrations and electronic couplings for conjugated molecules). By finding the Hamiltonian(s) that can best fit the spectra, the assignment can be automatically performed. This approach employs a Bayesian-optimization-derived algorithm as its driver and does not require a large volume of initial training data. In this talk, we focus on applying the method to model problems, higher-level calculations of benchmark systems, and real experimental spectra found in the literature. Finally, we will present our efforts on modeling the framework's robustness to noisy input data.