Ultra-fast Spectroscopy for High-Throughput and Interactive Quantum Chemistry

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Abstract

We present ultra-fast quantum chemical methods for the calculation of infrared and ultraviolet-visible spectra designed to provide fingerprint information during autonomous and interactive explorations of molecular structures. Characteristic spectral signals can serve as diagnostic probes for the identification and characterization of molecular structures. These features often do not require ultimate accuracy with respect to peak position and intensity, which alleviates the accuracy–time dilemma in ultra-fast electronic structure methods. If approximate ultra-fast algorithms are supplemented with an uncertainty quantification scheme for the detection of potentially large prediction errors in signal position and intensity, an offline refinement will always be possible to confirm or discard the predictions of the ultra-fast approach. Here, we present ultra-fast electronic structure methods for such a protocol in order to obtain ground- and excited-state electronic energies, dipole moments, and their derivatives for real-time applications in vibrational spectroscopy and photophysics. As part of this endeavor, we devise an information-inheritance partial Hessian approach for vibrational spectroscopy, a tailored subspace diagonalization approach and a determinant-selection scheme for excited-state calculations.

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