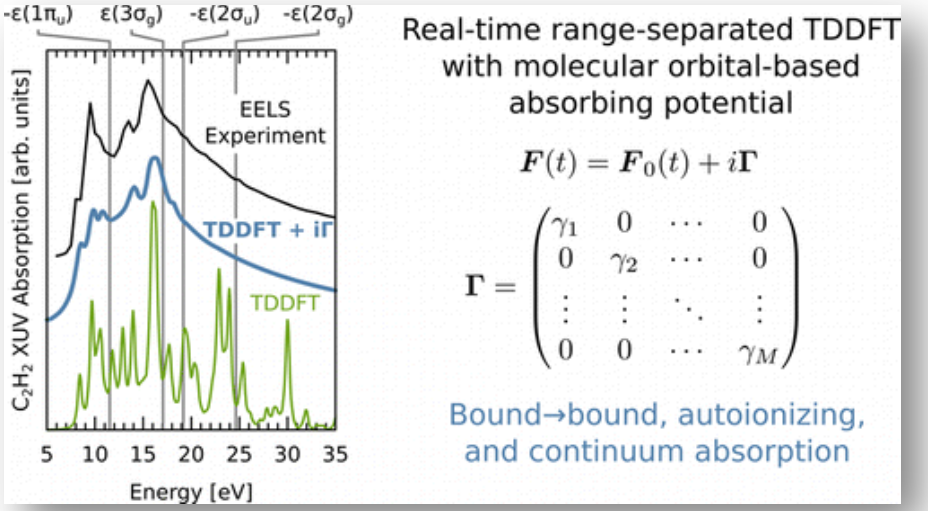
Near and Above Ionization Electronic Excitations with Non-Hermitian RT-TDDFT



RT-TDDFT computed extreme UV absorption spectrum of gas-phase acetylene with (blue) and without (green) an imaginary potential (iΓ), along with DFT Koopmans' IPs (gray). EELS data are shown in black.

Reference: K. Lopata, N. Govind, J. Chem. Theory. Comput, 9 (11), 4939-4946 (2013), Publication Date (Web): September 4, 2013

Work was performed at Pacific Northwest National Laboratory

Scientific achievement

Development of a RT-TDDFT formulation for capturing below and above-ionization excitations, mimic modeling to continuum

Significance and Impact

First-principles ionization dynamics, removes spurious high-energy finite basis artifacts, correct bound-to-bound transitions, metastable resonance states, consistent absorption shapes, prescription for accurate EAs and IPs

Research Details

- RT-TDDFT based on non-Hermitian density matrix propagation , atom-centered basis, tuned range-separated DFT, imaginary molecular orbital-based absorbing potential
- Computed extreme ultraviolet absorption for acetylene, water, Freon 12 agree well with EELS data over a broad range 0-50 eV