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Super-Resolution for Real-Time Electronic Structure Theory in Complex Systems

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Calculating excited state spectra of large, complex systems is often prohibitively expensive with standard frequency-domain methods such as the Casida equations, the Bethe-Salpeter Equation, or Equation-of-Motion Coupled Cluster. Real-time methods provide an alternative, as all modes are excited simultaneously. However, long simulation times are required to resolve narrow spectral features with traditional Fourier signal analysis, significantly limiting system size, and any information about the molecular orbitals involved in each transition is lost. Super-resolution methods such as Compressed Sensing promise high-resolution spectra from much shorter signals but assume the spectrum to be sparse, an assumption which breaks down in larger systems where sharp features are embedded in a quasi-continuum of smaller nearby peaks. To overcome this, we combine recent, highly noise-tolerant super-resolution techniques with a physically motivated separation of transition dipoles into bright and quasi-continuum contributions in an integrated, self-validating workflow. Our approach extends prior reconstruction schemes [1, 2, 3] with substantially improved robustness, while also recovering transition-level molecular orbital insight, promising to become an alternative to frequency-domain methods, particularly for large systems or hybrid functionals. We demonstrate it on systems containing several hundred heavy atoms, achieving up to 20-fold speedups while maintaining spectral accuracy even for signals dominated by large continua.

[1] M. Kick et al., Nat. Commun. 15, 8001 (2024)
[2] A. Bruner et al., JCTC 12, 3741 (2016)
[3] X. Andrade et al., PNAS 109, 13928 (2012)