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## Simulating the ultrafast dynamics of multi-mode multi-state molecular systems coupled to a dissipative environment

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Ultrafast processes in condensed phase photoexcited molecular systems involve the transition from a *coherent* dynamical regime –where a precise phase relation exists between different wave packet components –to *incoherent* “classical-like” dynamics. The decoherence process is driven the dissipation due to the surrounding molecular environment.

From the computational viewpoint, modelling the dynamics of nonadiabatic vibronic quantum systems interacting with fluctuating environments becomes especially challenging when the system’s high dimensionality precludes the calculation of its eigenstates. To overcome this limitation, a novel eigenstate-free formalism is introduced. This approach represents the open quantum system as a mixture of high-dimensional, time-dependent wave packets, governed by coupled Schrödinger equations [1], while the environment is modeled using a multi-component quantum master equation [2]. A computationally efficient implementation of this formalism employs a variational Gaussian/multiconfigurational time-dependent Hartree (G-MCTDH) ansatz for the wave packets and propagates the environment dynamics using hierarchical equations truncated at the first or second level.

The methodology is validated through simulations of multichromophoric aggregate dynamics, explicitly incorporating multiple vibrational modes, and through the study of vibrationally coherent symmetry-breaking charge transfer in a donor-acceptor-donor triad [3,4].

These results demonstrate the potential of the approach as a powerful quantum dynamical method for modeling complex system–bath interactions involving numerous degrees of freedom across multiple time scales.

[1] D. Picconi & I. Burghardt, *J. Chem. Phys.* **150**, 224106 (2019)

[2] D. Picconi, *J. Chem. Phys.* **161**, 164108 (2024)

[3] A. Aster, A.-B. Bornhof, N. Sakai, S. Matile, and E. Vauthey, *J. Phys. Chem. Lett.* **12**, 1052 (2021)

[4] D. Picconi, *J. Chem. Phys.* **156**, 184105 (2022)

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