Analyzing non-Markovian effects in large photosynthetic networks and open quantum systems

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In Short

- understanding the mechanisms of energy conversion in photosynthetic molecular systems
- study the time-resolved spectroscopy and relaxation dynamics of molecular complexes
- improve the distributed algorithms to solve the Hierarchical Equations Of Motion (HEOM)

Photosynthesis in plants fuels life on earth by converting solar radiation into chemical energy and by producing oxygen for respiration. Within the project we perform accurate simulation of energy transfer processes in photoactive pigment complexes. Understanding the directed energy transfer of an initial excitation of the antenna to the reaction center requires to model the dynamics on various timescales very accurately. This is achieved by regarding the molecular pigments as coupled entities, which convert part of the absorbed energy into heat (vibrations), while also retaining some properties of a coherent quantum state. We compare the theoretical results with experimental data by computing the time-resolved optical response caused by a series of time-delayed pump-probe pulses for models of Photosystem I and the photosynthetic apparatus of Green sulfur bacteria. The required simulations are computationally very demanding since we include electronic and vibrational degrees of freedom on an equal footing. Based on the Hierarchical Equation of Motion (HEOM) method for solving the quantum dynamics, we have developed a scalable, highly parallel computational package to simulate all optical spectra (DM-HEOM) [1,2]. With increasing number of pigments and decreasing temperature, the compute memory of the HEOM method increases exponentially. Our distributed memory DM-HEOM overcomes the memory barrier imposed by a single compute node by distributing the required memory across multiple nodes. This allows us to consider larger molecular complexes (100 pigments) and a wider range of temperatures (30K-300K) than previous implementations. DM-HEOM facilitates the computation of time- and frequency resolved spectra upon pulsed laser excitations with specific polarization sequences [1,3]. By comparing the HEOM results with more approximative methods, we uncover systematic limitations of perturbative approaches.

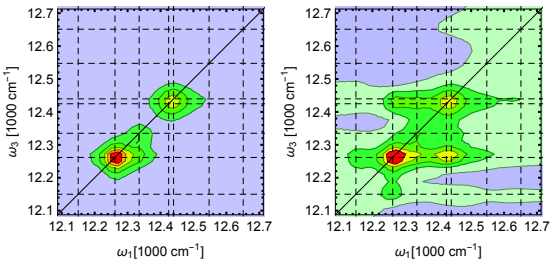

***Figure 1:** DM-HEOM calculation of the time-resolved spectroscopy of the photosynthetic Fenna Matthews Olson complex [1]. At a delay time of 40 femtoseconds, the left panel shows an all equal polarized case, while in the right panel two orthogonal polarization directions are used to enhance the visibility of off-diagonal signal.*

WWW

http://www.zib.de/members/kramer

More Information

[1] Kramer, T., Noack, M., Reinefeld, A., Rodríguez, M. & Zelinskyy, Y. *Efficient calculation of open quantum system dynamics and time-resolved spectroscopy with distributed memory HEOM (DM-HEOM).*Journal of Computational Chemistry (accepted, in press), https://arxiv.org/abs/1803.03498.

[2] Noack, M., Reinefeld, A., Kramer, T. & Steinke, T. *DM-HEOM: A Portable and Scalable Solver-Framework for the Hierarchical Equations of Motion.* In 19th IEEE International Workshop on Parallel and Distributed Scientific and Engineering Computing (PDSEC 2018) (2018).

[3] Kramer, T., Rodríguez, M. & Zelinskyy, Y. *Modeling of transient absorption spectra in exciton charge-transfer systems.* Journal of Physical Chemistry B 121, 463 (2016).

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