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Energy transfer dynamics in structured environments

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Our aim is to present a novel approach to the numeric treatment of open quantum systems similar to [1]. On the basis of a stochastic Schrödinger equation we develop a hierarchy of equations of motion which can be used to simulate their time evolution. This method is then applied to energy transfer dynamics and absorption spectra in molecular aggregates.

Non-Markovian quantum state diffusion

- stochastic Schrödinger equation in the Hilbert space of the system [2]

$$\partial_t \psi_t = -iH\psi_t + LZ_t^*\psi_t - L^\dagger \int_0^t \alpha(t-s)\frac{\delta\psi_t}{\delta Z_s^*}\,ds$$

  - free Hamiltonian H
  - coupling operator L
  - bath correlation function α
  - complex noise Z_t with cov. α

- equation for state vectors $\Longrightarrow$ well suited for numerics
- density operator recovered as ensemble average over Z_t
- no approximation necessary so far

$\Rightarrow$ Functional derivation? Time non-locality? $\Longrightarrow$ Hierarchical Equations!

Hierarchical equations of motion

- Idea: derive equation of motion for problematic term

$$\psi_t^{(1)} := \int_0^t \alpha(t-s)\frac{\delta\psi_t}{\delta Z_s^*}\,ds$$

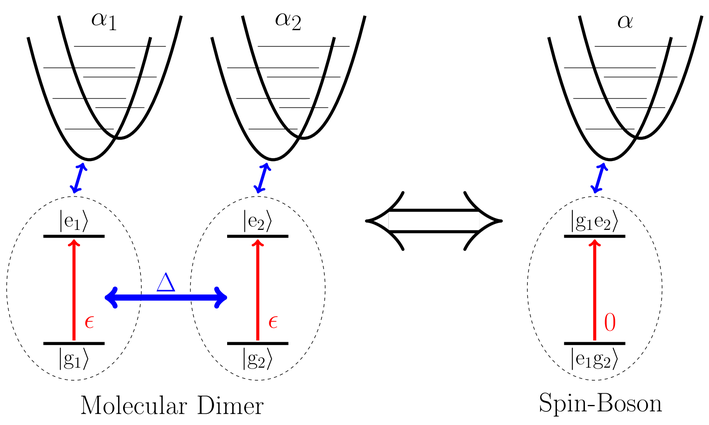
- simple form only for exponential correlation function $\alpha(t) \propto e^{-\gamma|t|-i\Omega t}$
- same structure as NMSSE, but with second order functional derivation

$\Rightarrow$ Infinite Hierarchy of state vectors $\psi_t^{(k)}$:

$$\partial_t \psi_t^{(k)} = (-iH - (\gamma + i\Omega)k + LZ_t^*)\,\psi_t^{(k)} + k\alpha(0)L\psi_t^{(k-1)} - L^\dagger\psi_t^{(k+1)}$$

- higher orders suppressed by $e^{-\gamma k}$ $\Longrightarrow$ truncation for numerical treatment

Energy transfer and absorption spectra in molecular dimers



Basic model of dimers

- two identical monomers with
  - electronic degrees of freedom → two level system
  - vibrational degrees of freedom → bath of oscillators
- coupling via dipol-dipol interaction
- simplification: only consider states with one excitation

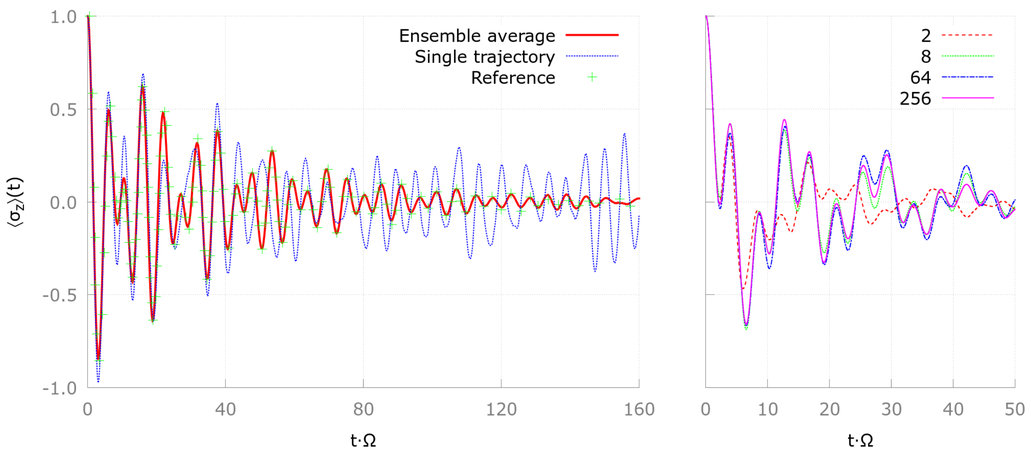
$\Rightarrow$ equivalent to symmetric Spin-Boson model with $H = \epsilon\mathbb{1} + \Delta\sigma_x$

- often few dominant modes in vibrational spectral density

$\Rightarrow$ fit α with sum of exponential functions

$\Rightarrow$ NMSSE contains sum of independent noise processes

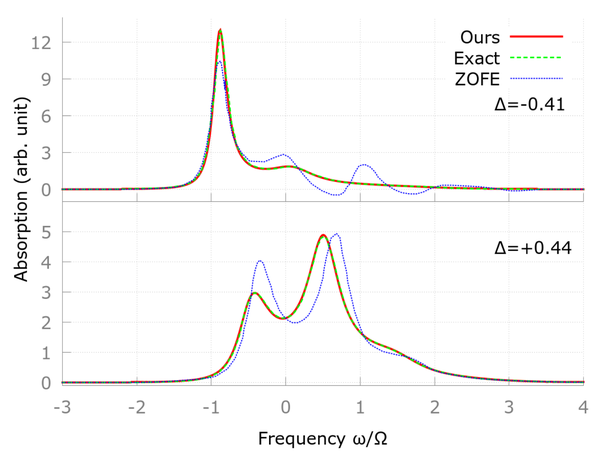
Dynamics of Spin-Boson model



Left picture Time evolution of weakly coupled Spin-Boson model in highly non-Markovian regime. Comparison between our approach and the analytical results of [3].

Right picture Same parameters but stronger coupling. Comparison between different numbers of hierarchies.

Absorption spectra



Absorption spectra of dimer for two coupling strenghts. ZOFE is an ansatz replacing the functional derivative with a time-dependent system operator [4]. Exact calculation is from [4] as well.

Conclusion and outlook

- efficient tool for small systems → apply to larger and more interesting systems

$\Rightarrow$ especially the inclusion of thermal noise and more complex spectral densities

- further comparison with established approaches (ZOFE [4], Tanimura [1], ...)
- better understanding of the underlying mathematics (e.g. of the functional derivative)

[1] Y. Tanimura, JPSJ 75, 8 (2006)

[2] L. Diósi, N. Gisin, W. T. Strunz, Phys. Rev. A 58, 3 (1998)

[3] P. Huang, H. Zheng, J. Phys: Condens. Matter 20, 395233 (2008)

[4] J. Roden, W. T. Strunz, A. Eisfeld, J. Chem. Phys. 134, 034902 (2011)

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