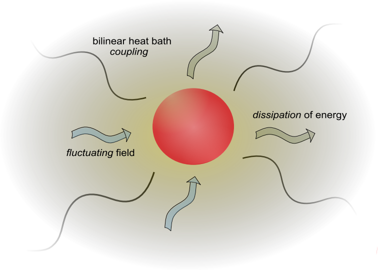
Time Correlated Blip Dynamics of Open Quantum Systems



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Mapping a reservoir to probability space

Bilinear system-reservoir complex

$$H(t) = H_S(t) + H_I + H_R \quad \text{where} \quad H_I = -q \cdot \mathcal{E} \quad \text{and} \quad H_R = \sum_k \hbar\omega_k b_k^\dagger b_k$$

Feynman-Vernon

$$\rho(q_f, q_f', t) = \int dq_i dq_i' \rho(q_i, q_i', t_0) \int \mathcal{D}[q_1]\mathcal{D}[q_2] e^{\frac{i}{\hbar}(S_S[q_1]-S_S[q_2])} F_{FV}[q_1, q_2]$$

Stochastic noise action $\implies$ $S_\zeta[q] = S_S[q] + \int_{\mathcal{C}} du \zeta(u) q(u)$

$$\underbrace{\langle \exp\left[\frac{i}{\hbar}\int_{\mathcal{C}} du \zeta(u) q(u)\right]\rangle_R}_{} = \exp\left[-\frac{1}{\hbar^2}\int_{\mathcal{C}} du \int_{u>u'} du' q(u) \langle \zeta(u)\zeta(u')\rangle_R q(u')\right]$$

$\zeta(u) \in \mathcal{C}$ **Gaussian**: characteristic functional

$$\langle \zeta(u)\zeta(u')\rangle_R = \hbar L(u-u') = \text{bath autocorrelation} \Rightarrow F_{FV}$$

Stochastic Liouville-von Neumann equation

Complex-valued stochastic sample dynamics - the **SLN equation**:

$$\frac{d}{dt}\rho_z(t) = -\frac{i}{\hbar}[H_S, \rho_z(t)] + \frac{i}{\hbar}\xi(t)[q, \rho_z(t)] + \frac{i}{2\hbar}\nu(t)\{q, \rho_z(t)\}$$

Sample average $\rho = \mathbb{M}[\rho_z]$ with respect to $z = (\xi(t), \nu(t))$:

$$\langle \xi(t)\xi(0)\rangle_R = \hbar L'(t), \quad \langle \xi(t)\nu(0)\rangle_R = 2i\hbar L''(t), \quad \langle \nu(t)\nu(0)\rangle_R = 0$$

Gaussian reservoir $$L(t) = \frac{1}{\pi}\int_0^\infty d\omega\, J(\omega)\frac{\cosh[\omega(\hbar\beta/2 - it)]}{\sinh(\hbar\beta\omega/2)}$$

Key benefits:

- general approach for open system quantum dynamics
- non-perturbative, time-local and non-Markovian

J. S., arXiv: 1608.03438v2 (accepted EPL, 2016)

J. S., Grabert, Phys. Rev. Lett. **88** (2002)

SLED - stochastic real noise and dissipation

Ohmic dissipation and scaling limit $\omega_c \to \infty$:

$$L''(t-t') \sim \gamma \frac{d}{dt}\delta(t-t')$$

EOM (**SLED**) for quantum system driven by real noise $\tilde{\zeta}(t)$:

$$\begin{aligned} \frac{d}{dt}\rho_{\tilde{\zeta}} &= \frac{1}{i\hbar}\left([H_S,\rho_{\tilde{\zeta}}] - \tilde{\zeta}(t)[q,\rho_{\tilde{\zeta}}]\right) \\ &+ \frac{\gamma}{2i\hbar}[q,\{p,\rho_{\tilde{\zeta}}\}] - \frac{m\gamma}{\hbar^2\beta}[q,[q,\rho_{\tilde{\zeta}}]] \end{aligned}$$

- equilibrium relaxation of populations ρ_{diag} governed by γ
- noise $\tilde{\zeta}$ induced dephasing time τ_D

Long range ($\sim \hbar\beta$), thermal correlator $L'(\tau)$:

$$\left\langle \tilde{\zeta}(\tau)\tilde{\zeta}(0)\right\rangle_R = L'(\tau) - \frac{2m\gamma}{\hbar\beta}\delta(\tau)$$

Proof of concept: SLN and SLED

Applications

- **optimal control**
  R. Schmidt et al., Phys. Rev. Lett. **107**, 130404 (2011)
  R. Schmidt et al., Phys. Rev. A **88**, 052321 (2013)
- **semiclassical dynamics**
  W. Koch et al., Phys. Rev. Lett. **100**, 230402 (2008)
- **spin dynamics**
  J. S., J. Chem. Phys. **296**, 159169 (2004)
- **bio-molecules** and **structured spectral densities**
  H. Imai et al., J. Chem. Phys. **446**, 134141 (2015)
- **work and heat**
  R. Schmidt et al., Phys. Rev. B **91**, 224303 (2015)
  J. S. and T. Motz, arXiv: 1606.04326v1 (2016)

Challenges ?

Physical density matrix obtained from $\rho = \mathbb{M}[\rho_z]$:

- non-unitary propagation of individual ρ_z trajectories

$$\underbrace{\frac{i}{2\hbar}\nu(t)\{q, \rho_z(t)\}}_{\Rightarrow \text{Tr}(\cdot)\neq 0,\ \text{non-unitary}} \quad \text{and} \quad \langle \nu(t)\nu(0)\rangle_R = 0$$

- asymptotic signal-to-noise deterioration

$$\propto \exp(-\alpha \cdot t)$$

Finite non-local system-reservoir interactions

Idea: projection on relevant and irrelevant part $\rho = \mathcal{P}\rho + \mathcal{Q}\rho$

$\Rightarrow$ *Nakajima-Zwanzig*

$$\dot{\rho}_{\mathcal{P}}(t) = \mathcal{P}\mathcal{L}(t)\rho_{\mathcal{P}} + \mathcal{P}\mathcal{L}(t)\int_0^t \mathrm{dt'}\, \exp_> \left[\int_{t'}^t \mathrm{ds}\ \mathcal{Q}\mathcal{L}(s)\right] \mathcal{Q}\mathcal{L}(t')\rho_{\mathcal{P}}(t')$$

Observation:

1. dephasing induced by $\mathcal{Q}\mathcal{L}(t)\mathcal{Q}$ superoperators
2. $\tau_D \ll t$ allows for raising lower integration bound

$$\int_0^t \longrightarrow \int_{t-\tau_m}^t$$

noisy propagation
over shorter intervals τ_m

Result: more efficient algorithm

Time Correlated Blip Dynamics - TCBD (1/2)

Reformulation of SLED:

$$\boxed{\mathcal{P}\rho\colon\ \rho \to \rho_{diag}}$$

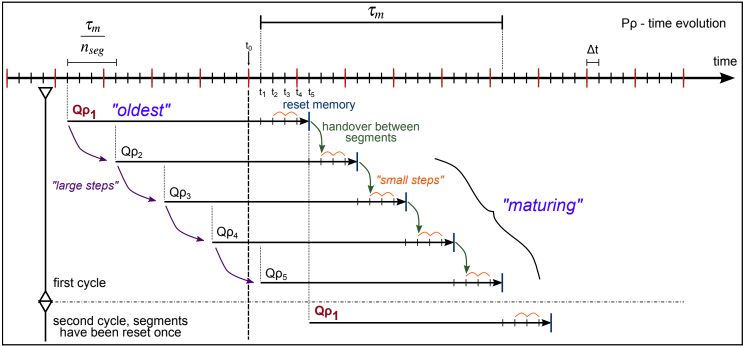
$$\frac{d}{dt}\begin{pmatrix}\rho_{\mathcal{P}}\\ \rho_{\mathcal{Q}}\end{pmatrix} = \begin{pmatrix}0 & \mathcal{P}\mathcal{L}_{det}\mathcal{Q}\\ \mathcal{Q}\mathcal{L}_{det}\mathcal{P} & \mathcal{Q}[\mathcal{L}_{det}+\mathcal{L}_{\zeta}]\mathcal{Q}\end{pmatrix}\begin{pmatrix}\rho_{\mathcal{P}}\\ \rho_{\mathcal{Q}}\end{pmatrix}$$

- deterministic sup. $\mathcal{L}_{det} \equiv \frac{1}{i\hbar}[H_S,\cdot] + \frac{\gamma}{2i\hbar}[q,\{p,\cdot\}]$
- stochastic superoperator $\mathcal{L}_{\zeta} \equiv \frac{i}{\hbar}[q,\cdot]\zeta(t)$

$\Rightarrow$ numerical solution via symmetric Trotter splitting

Time Correlated Blip Dynamics - TCBD (2/2)

Algorithm based on segmented time evolution of coherences:



TCBD approach:

- raise lower integration bound
- introduce multiple versions of $\mathcal{Q}\rho$: $\mathcal{Q}\rho_j$, $j \in \{1, 2, \ldots\}$

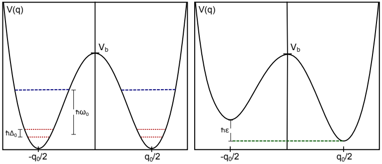
  $\Rightarrow$ stepwisely (τ_m/n_{seg}) increased initial conditions

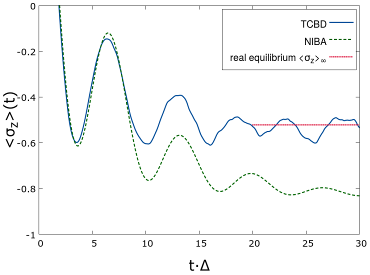
Dissipative two-state dynamics (1/2)

Spin-boson model:

$$H_S = \frac{\hbar\epsilon}{2}\sigma_z - \frac{\hbar\Delta}{2}\sigma_x$$

$$H_I = -\sigma_z \cdot \mathcal{E}$$





$\epsilon/\Delta = 0.5$, $\hbar\beta = 5\cdot\Delta^{-1}$, $K \approx 0.1$, $n_{samp} = 2000$

Analytical *comparative* theory:

$\Rightarrow$ **NIBA**

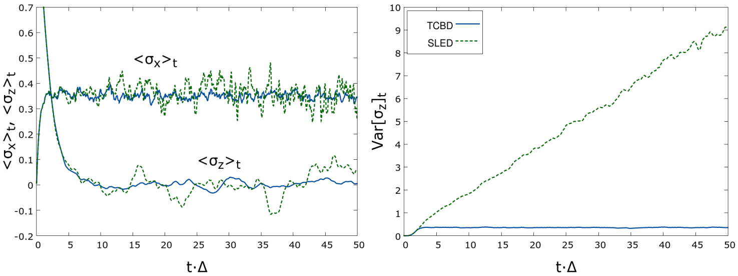
- approximative
- estimator for memory

$$t \gg \hbar\beta : \quad \tau_m \sim \frac{4\hbar\beta}{2K\pi}$$

U. Weiss, *Quantum Dissipative Systems*, World Scientific, Singapore, 4th edition (2012)

Dissipative two-state dynamics (2/2)

First and second order statistics for TCBD spin-boson dynamics:



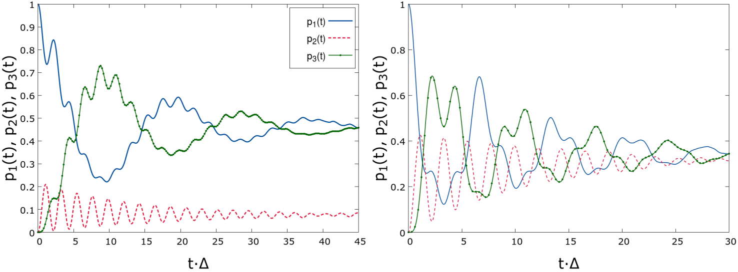
$\epsilon/\Delta = 0$, $\hbar\beta = 0.7 \cdot \Delta^{-1}$, $K = 0.25$, $n_{samp} = 2500$

$\Rightarrow$ significant improvement in **signal-to-noise** ratio

Dissipative three state system

Hamiltonian in spin-1 basis $\{\mathbb{1}, S_x, S_y, S_z\}$

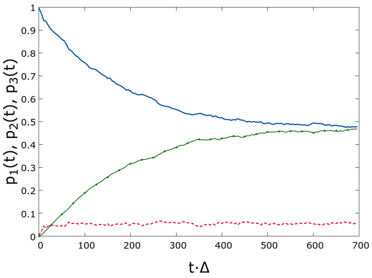
$$H_S = \frac{\hbar}{\sqrt{2}} \begin{pmatrix} 0 & \Delta & 0 \\ \Delta & \epsilon & \Delta \\ 0 & \Delta & 0 \end{pmatrix} \quad \text{and} \quad H_I = -S_z \cdot \mathcal{E}$$



$\epsilon/\Delta = 3$, $\hbar\beta = 5 \cdot \Delta^{-1}$, $K = 0.25$ $\quad$ $\epsilon/\Delta = 1$, $\hbar\beta = 7 \cdot \Delta^{-1}$, $K = 0.08$

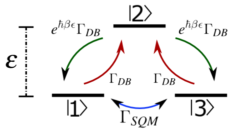
coherent regime

Dissipative 3LS - Transfer Rates (1/2)



$\epsilon/\Delta = 17$, $\hbar\beta = 5 \cdot \Delta^{-1}$, $K = 0.25$

incoherent regime



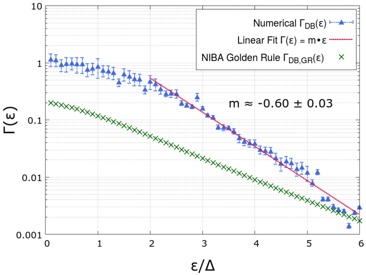
Three state population **transfer model**:

- thermally activated hopping Γ_{DB}
- superexchange tunneling Γ_{SQM}

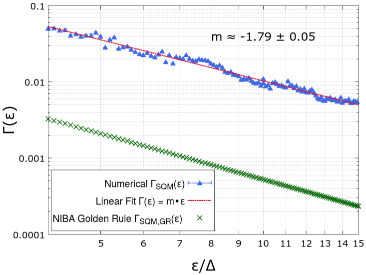
Dissipative 3LS - Transfer Rates (2/2)

Comparative model: **NIBA**, Fermi's **golden rule** rates

$\hbar\beta = 0.7 \cdot \Delta^{-1}, K = 0.25$



Arrhenius



Superexchange

Conclusion

Finite memory **TCBD** scheme:

- *improvement* of SLN/SLED stochastic sampling
- *memory length* τ_m tailored for application
- investigation of *coherent* and *incoherent* dynamics
  $\Rightarrow$ *transfer rates*
- extension to
  - *higher dimensional* systems
  - *anharmonic* oscillators

Thank You!