Stochastic Formalism and Simulations of Quantum Dissipative Dynamics

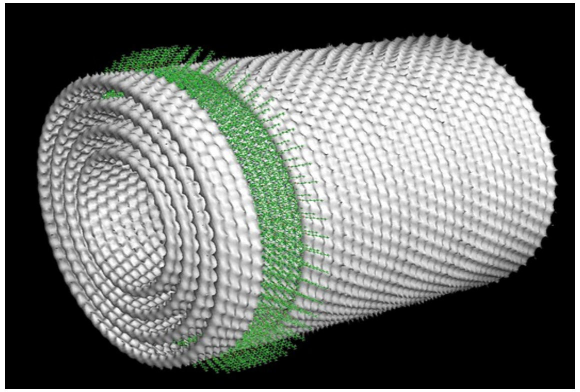
Jianshu Cao

Department of Chemsitry, MIT

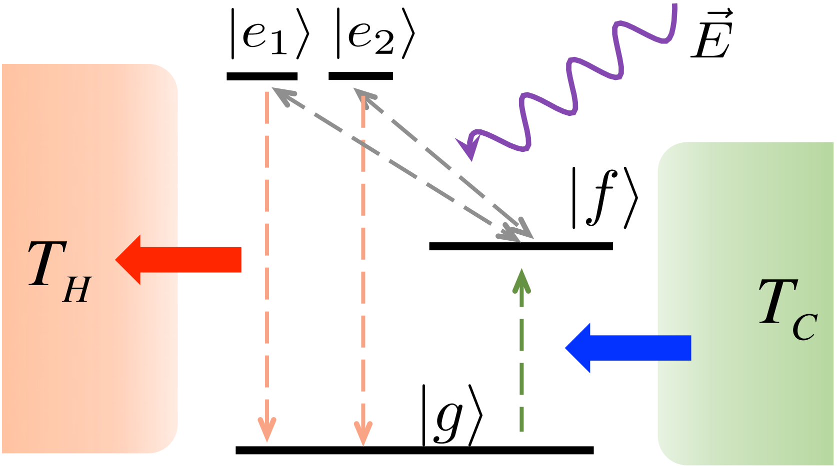
- Unified Stochastic Formalism (Chang-yu Hsieh)
- Stochastic Path Integral Simulations (Jeremy Moix)

Nov, 11, 2019, POSTECH, Korea
5^{th} KIAS Workshop
Quantum Information and Thermodynamics

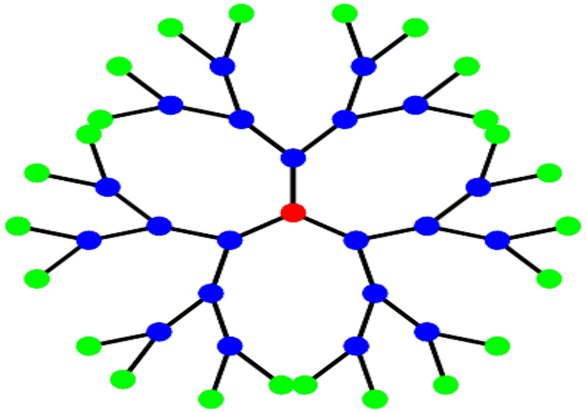
Applications to Open Quantum Processes



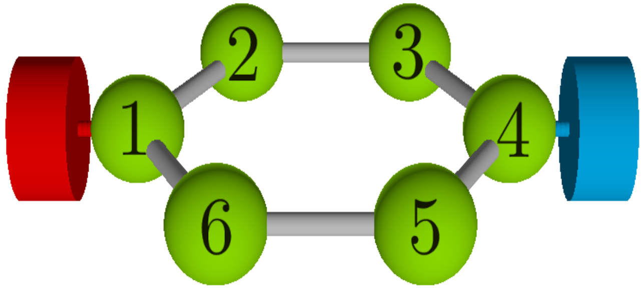
Quantum transport, PRL 116, 196803 (2016)



Quantum heat pumps NJP 18, p 023003 (2016)



Light-harvesting energy transfer PRL 110, 200402 (2013)



Heat transfer in Benzene Sci. Rep. 6, 28027 (2016)

Brownian Motion

Deterministic probability approach: Fokker-Planck Equation (1905)

$$\frac{\partial}{\partial t}P(x,t) = \frac{1}{2}\frac{\partial^2}{\partial x^2}\left[D(x,t)P(x,t)\right].$$

Ensemble averaging: $A(t) = \langle A(x) \rangle_{P(t)}$

Stochastic trajectory approach: Langevin Equation of Motion (1908)

$$m\frac{d^2\mathbf{x}}{dt^2} = -\eta\frac{d\mathbf{x}}{dt} + \boldsymbol{f}\,(t)\,.$$

Fluctuation-dissipation relation: $$\langle f_i\,(t)\,f_j\,(t')\rangle = 2\eta_{i,j}k_BT\delta\,(t-t')\,.$$

Stochastic averaging: $A(t) = \langle A[f(t)] \rangle_{f(t)}$

Later, Kubo et al showed the equivalence between HEOM and GLE with exponential noise

Dissipative Quantum Dynamics

Deterministic probability approach:

$$\frac{\partial \rho(t)}{\partial t} = -i\mathcal{L}\rho(t)$$

Ensemble averaging: $A(t) = \langle A(x) \rangle_{\rho(t)}$

HEOM, MCTDH, PIMC, QUAPI, etc.

Stochastic trajectory approach:

$$\frac{\partial \psi(t)}{\partial t} = -i[\hat{H} + f(t)\hat{V}]\psi(t)$$

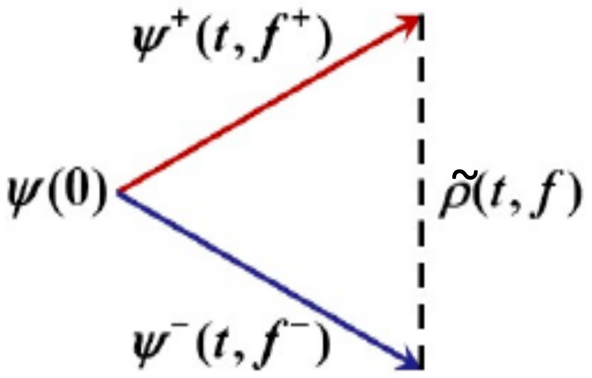
Fluctuation-dissipation relation: $\langle f(t) f(t') \rangle = C(t)$

Stochastic averaging: $A(t) = \langle A[f(t)] \rangle_{f(t)}$ [Each realization is unphysical, but the average is physical]

Quantum state diffusion, Stochastic Liouville equation, Stochastic path integral, etc

A Novel Method for Simulating Quantum Dissipative Systems

J. Cao, L. W. Ungar, and G. A. Voth, J. Chem. Phys. 104, 4189 (1996)



1 Monte Carlo sampling of the Gaussian functional

$$\exp(-S_{bath}[x_c(t)])$$

2. Propagation of the system wave-function

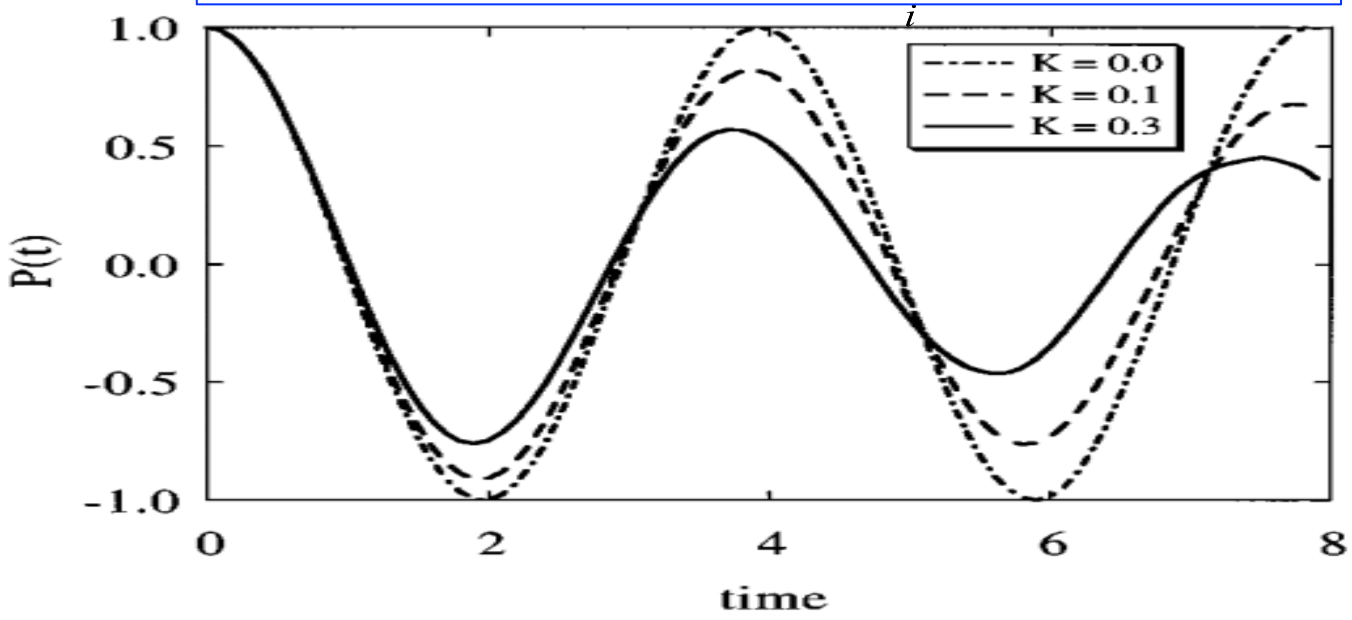
$$i\frac{\partial \psi}{\partial t} = [H_s + V(x_c(t), q)]\psi$$

3. Average of the bath configurations

spin-boson model

Kondo constant:

$$J(\omega) = \frac{\pi \hbar K}{2}\omega e^{-\omega/\omega_c}$$



Stochastic Liouville Equation for Gaussian Bath

Replace bath with stochastic force if V = f(x) A(q)

$$\begin{cases} i\dfrac{d}{dt}|\psi^-(t)\rangle = [H_s + f^{-*}(t)A]|\psi^-(t)\rangle \\ i\dfrac{d}{dt}|\psi^+(t)\rangle = [H_s + f^{+}(t)A]|\psi^+(t)\rangle \end{cases}$$

stochastic RDM $\tilde{\rho}_s(t) = |\psi^+(t)\rangle\langle\psi^-(t)|$

$$\boxed{\frac{d\,\tilde{\rho}_s(t)}{dt} = -i[H_s, \tilde{\rho}_s] - iA\tilde{\rho}_s\,(t)f^+(t) + i\tilde{\rho}_s\,(t)Af^-(t)}$$

Fluctuation-dissipation relation: $\langle f\,(t)\,f\,(t')\rangle = C(t)$

SLE: Stockburger and Grabert, Phys. Rev. Lett., 88:170407, (2002): Cao, Ungar, Voth, JCP, 104, 6189 (1996)

Goal: Numerical Implementation and Generalize to any bath models

GHE

Generalized hierarchy equation

1. Bosonic bath
2. Fermionic bath
3. Spin bath (dual Fermion)
4. non-Gaussian bath

$$\tilde{\rho}(t) = |\psi^{+}(t)\rangle\langle\psi^{-}(t)|$$

SLE \ SW

SPI/SW

Stochastic path integrals

1. Imaginary time – thermal distribution
2. Absorption / Emission spectra
3. Multi-chromophor Forster rate

Hybrid

Deterministic + Stochastic

1. stochastic-HEOM (JCP139,13406, 2013)
2. Transfer tensor method (PRL 112, p11040, 2014)

System + Bath Quantum Dynamics

$$H = H_s + H_b + \mathrm{AB}$$

$$\rho(0) = \rho_s(0) \otimes \rho_b^{eq}$$

$$\frac{d\rho(t)}{dt} = -i\left[H, \rho(t)\right]$$

Three classes of bath models :

$$H_b = \sum_k \omega_k a_k^+ a_k$$

$$[a_k, a_j^+] = \delta_{k,j}$$

Bosons

$$H_b = \sum_k \omega_k c_k^+ c_k$$

$$\{c_k, c_j^+\} = \delta_{k,j}$$

Fermions

$$H_b = \sum_k \frac{1}{2} \sigma_k^z$$

$$[\sigma_k^x, \sigma_k^y] = i\sigma_k^z$$

Spins

Stochastically Decoupled Quantum Dynamics

$$\frac{d\rho(t)}{dt} = -i\left[H, \rho(t)\right]$$

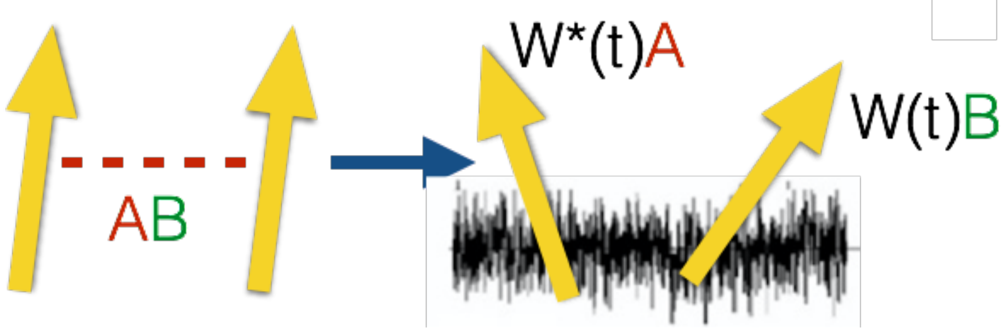
$$d\tilde{\rho}_s(t) = -i\,dt\left[H_s, \tilde{\rho}_s(t)\right] - \frac{i}{\sqrt{2}} A\tilde{\rho}_s(t)\,dW + \frac{i}{\sqrt{2}} \tilde{\rho}_s(t) A\,dV$$

$$d\tilde{\rho}_b(t) = -i\,dt\left[H_b, \tilde{\rho}_b(t)\right] + \frac{1}{\sqrt{2}} dW^* B\tilde{\rho}_b(t) + \frac{1}{\sqrt{2}} dV^* \tilde{\rho}_b(t) B$$

White Noise Statistics $\quad \overline{dW dW^*} = \overline{dV dV^*} = 2\,dt$

reduced density matrix

$$\rho_s(t) = \overline{\tilde{\rho}_s(t)\,\mathrm{Tr}_b\,\tilde{\rho}_b(t)}$$



Stochastic decoupling of bilinear coupling AB

Bath-induced Dissipations and Multi-time Correlation Functions

$$d\tilde{\rho}_b(t) = -i\,dt\left[H_b, \tilde{\rho}_b(t)\right] + \frac{1}{\sqrt{2}} dW^* B\tilde{\rho}_b(t) + \frac{1}{\sqrt{2}} dV^* \tilde{\rho}_b(t) B$$

Take trace and obtain formal solution

$$\boxed{\mathrm{Tr}_b \tilde{\rho}_b(t) = \exp\left(-\frac{1}{\sqrt{2}} \int_0^t (dW_s + dV_s) \mathcal{B}(s) \right)}$$

Analogous to the influence functional.

forward / backward path
In terms of noise realization

Bath-induced fluctuating field

The fluctuation and dissipation kernels as well as higher order responses encoded in bath's multi-time correlation functions.

Bath-induced fluctuating field

$$\mathcal{B}(t) = \mathrm{Tr}_b\left(B\,\tilde{\rho}_b(t) \right)$$

Bath-induced Dissipations and Multi-time Correlation Functions

$$\rho_s(t) = \overline{\tilde{\rho}_s(t)\,\boxed{\mathrm{Tr}_b\,\tilde{\rho}_b(t)}}$$

Bath-induced fluctuations

$$\boxed{\mathrm{Tr}_b\tilde{\rho}_b(t) = \exp\left(-\frac{1}{\sqrt{2}}\int_0^t (dW_s + dV_s)\mathcal{B}(s)\right)}$$

Analogous to the influence functional.

$$\mathcal{B}(t) = \frac{1}{\sqrt{2}}\int_0^t dW_s^*\Phi_{2,1}(t,s) + \frac{1}{\sqrt{2}}\int_0^t dV_s^*\Phi_{2,2}(t,s) +$$
$$+\left(\frac{1}{\sqrt{2}}\right)^3 \int_0^t\int_0^{s_1}\int_0^{s_2} dW_{s_1}^*\,dW_{s_2}^*\,dW_{s_3}^*\Phi_{4,1}(t,s_1,s_2,s_3) + \ldots +$$
$$+\left(\frac{1}{\sqrt{2}}\right)^3 \int_0^t\int_0^{s_1}\int_0^{s_2} dV_{s_1}^*\,dV_{s_2}^*\,dV_{s_3}^*\Phi_{4,8}(t,s_1,s_2,s_3) +$$

Only first two terms exist for Gaussian bath models.

Multi-time correlation functions convoluted with the noise histories.

Stochastic Liouville Equation (SLE)

A simple outline to obtain SLE,

$$\boxed{\frac{d\,\tilde{\rho}_s(t)}{dt} = -i[H_s, \tilde{\rho}_s] \mp iA\tilde{\rho}_s\,(t)f^+(t) + i\tilde{\rho}_s\,(t)Af^-(t)}$$

All distinguishing properties of various bath models are now hidden under the details of the noise: Complex-valued vs Grassmann-valued , Gaussian vs non-Gaussian etc.

Two-Time Statistics

$$\overline{f^+(t)f^+(t')} = C_{++}(t'-t)$$

$$\overline{f^+(t)f^-(t')} = C_{+-}(t'-t)$$

$$\overline{f^-(t)f^-(t')} = C_{--}(t'-t)$$

Bath's two-time correlation function (boson case)

$$\boxed{C_{+-}(t) = \alpha_B(t) = \int d\omega J(\omega)\left(\coth\frac{\beta\omega}{2}\cos(\omega t) - i\sin(\omega t)\right)}$$

Stockburger and Grabert, Phys. Rev. Lett., 88:170407, 2002

Stochastic Liouville Equation (SLE)

A simple outline to obtain SLE,

$$\boxed{\frac{d\,\tilde{\rho}_s(t)}{dt} = -i[H_s, \tilde{\rho}_s] \mp iA\tilde{\rho}_s\,(t) f^{+}(t) + i\tilde{\rho}_s\,(t) A f^{-}(t)}$$

All distinguishing properties of various bath models are now hidden under the details of the noise: Complex-valued vs Grassmann-valued , Gaussian vs non-Gaussian etc.

Two-Time Statistics

$$\overline{f^{+}(t)f^{+}(t')} = C_{++}(t'-t)$$

$$\overline{f^{+}(t)f^{-}(t')} = C_{+-}(t'-t)$$

$$\overline{f^{-}(t)f^{-}(t')} = C_{--}(t'-t)$$

Boson (complex-valued) vs Fermion (Grassmann-valued)

$$C_{++}(t'-t) = \begin{cases} \alpha_B(|t'-t|) & \text{Boson} \\ \theta(t-t')\alpha_F(t-t') - \theta(t'-t)\alpha_F(t'-t\quad) & \text{Fermion} \end{cases}$$

correlation functions with Bose-Einstein or Fermi-Dirac statistics.

Hsieh and Cao, JCP 148, p014103 (2018)

Stochastic Liouville Equation (SLE)

A simple outline to obtain SLE,

$$\boxed{\frac{d\,\tilde{\rho}_s(t)}{dt} = -i[H_s, \tilde{\rho}_s] \mp iA\tilde{\rho}_s\,(t) f^+(t) + i\tilde{\rho}_s\,(t) A f^-(t)}$$

All distinguishing properties of various bath models are now hidden under the details of the noise: Complex-valued vs Grassmann-valued , Gaussian vs non-Gaussian etc.

Two-Time Statistics

$$\overline{f^+(t) f^+(t')} = C_{++}(t'-t)$$

$$\overline{f^+(t) f^-(t')} = C_{+-}(t'-t)$$

$$\overline{f^-(t) f^-(t')} = C_{--}(t'-t)$$

Four-Time Statistics etc.

$$\overline{f^+(t_1) f^+(t_2) f^+(t_3) f^+(t_4)} = C_{++++}(t_2-t_1, t_3-t_2, t_4-t_1)$$

$$\cdots$$

$$\overline{f^-(t_1) f^-(t_2) f^-(t_3) f^-(t_4)} = C_{----}(t_2-t_1, t_3-t_2, t_4-t_1)$$

Deterministic Solutions for SLE

1. Formal Averaging over Stochastic Variables

$$\frac{d\,\overline{\tilde{\rho}_s(t)}}{dt} = -i\left[H_s, \overline{\tilde{\rho}_s(t)}\right] \mp iA\overline{\tilde{\rho}_s\,(t)f^+(t)} \pm i\overline{\tilde{\rho}_s\,(t)f^-(t)}\,A$$

$$= -i\left[H_s, \overline{\tilde{\rho}_s(t)}\right] \mp iA\overline{\tilde{\rho}_s\,(t)\,\underline{\mathcal{B}(t)}} \pm i\overline{\tilde{\rho}_s\,(t)\underline{\mathcal{B}(t)}}\,A$$

2. Define Auxiliary Density Matrices (ADM), m = 0 is RDM

$$\rho^{[m]} = \overline{\tilde{\rho}_s\,\mathcal{B}^m(t)}$$

3. Deterministic Equations of Motions

$$d_t[\overline{\tilde{\rho}_s\mathcal{B}^m}] = \overline{d_t[\tilde{\rho}_s]\mathcal{B}^m} + \overline{\tilde{\rho}_s d_t\mathcal{B}^m} + \overline{d_t\tilde{\rho}_s d_t\mathcal{B}^m}.$$

Deterministic Solutions for SLE

$$d_t[\overline{\tilde{\rho}_s \mathcal{B}^m}] = \overline{d_t[\tilde{\rho}_s]\mathcal{B}^m} + \overline{\tilde{\rho}_s d_t \mathcal{B}^m} + \overline{d_t \tilde{\rho}_s d_t \mathcal{B}^m}.$$

To turn the above into a closed hierarchy,

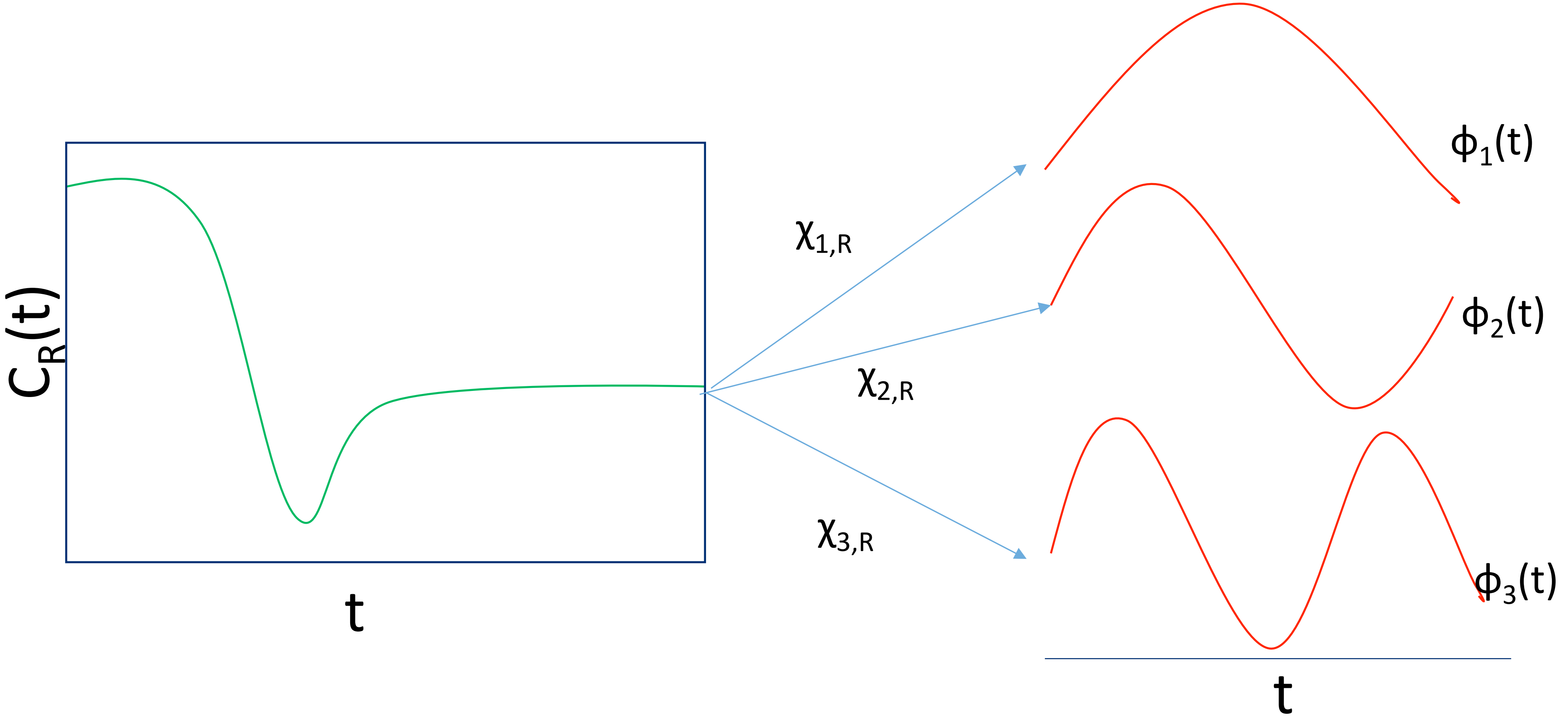
1. A suitable decomposition of stochastic field B(t) : exponentials, orthonormal basis etc.

2. A correspondingly refined definition of auxiliary density matrices

Hsieh and Cao, JCP 148, p014103 (2018)

Orthonormal Function Expansions of Correlation Functions

Single variable for Gaussian $\quad \mathrm{C(t)} = \sum_{j=1}^{K} \chi_j \phi_j(t) \qquad \int_0^T ds\, w(s)\phi_i(s)\phi_j(s) = \delta_{ij}$



From Stochastic to Hierarchical Equations

1. A suitable basis set: orthonormal function basis (e.g. exponential functions)

$$\mathrm{C(t)} = \sum_{j=1}^{K} \chi_j \phi_j(t) \qquad \int_0^T ds\, w(s)\phi_i(s)\phi_j(s) = \delta_{ij}$$

2. Decomposition of stochastic field B(t)

$$\mathcal{B}(t) = \sum_{j=1}^{K} \chi_j a_j(t). \qquad a_j(t) \equiv \left(\frac{1}{\sqrt{2}}\right) \int_0^t dU_s^* \phi_j(t-s),$$

3. Correspondingly a refined definition of auxiliary density matrices (ADM)

$$\bar{\rho}_{\boldsymbol{n}}(t) = \overline{\tilde{\rho}_S(t) a_1^{n_1}(t) \cdots a_K^{n_K}(t)}$$

n_j = 0,1,2,3… for bosons
n_j = 0 / 1 for fermions

Hsieh and Cao, JCP 148, p014103 (2018)

Extended Hierarchy Equation for Boson Bath

$$\frac{d\bar{\rho}_{\mathbf{n}}}{dt} = -i\left[\hat{H}_{\mathbf{s}}, \bar{\rho}_{\mathbf{n}}\right] + i\sum_j \chi_j \left(A\bar{\rho}_{\mathbf{n}+\mathbf{1}_j} - \bar{\rho}_{\mathbf{n}+\mathbf{1}_j}A\right)$$
$$+i\sum_j \phi_j(0)\left(A\bar{\rho}_{\mathbf{n}-\mathbf{1}_j} + \bar{\rho}_{\mathbf{n}-\mathbf{1}_j}A\right)$$
$$+\sum_{j,j'} \eta_{jj'}\bar{\rho}_{\mathbf{n}_{j\to j'}},$$

exponential decay: HEOM (Tanimura)
related development: Stochastic derivation of HEOM (Shao)
extended-HEOM (Wu): quantum phase transition in subohmic spin-boson model, PRB 95, p214308 (2017)

Hsieh and Cao, JCP 148, p014103 (2018)

Hierarchy Equation for Fermions

$$\frac{d\bar{\rho}_{\mathbf{n}}}{dt} = -i\left[\hat{H}_s, \bar{\rho}_{\mathbf{n}}\right] + i\sum_j \chi_j \left(A\bar{\rho}_{\mathbf{n}+\mathbf{1}_j} + \bar{\rho}_{\mathbf{n}+\mathbf{1}_j}A\right) \underline{(-1)^{|\mathbf{n}|_j}(1-n_j)}$$
$$+i\sum_j \phi_j(0)\left(A\bar{\rho}_{\mathbf{n}-\mathbf{1}_j} + \bar{\rho}_{\mathbf{n}-\mathbf{1}_j}A\right)\underline{(-1)^{|\mathbf{n}|_j} n_j}$$
$$+\sum_{j,j'} \eta_{jj'}\bar{\rho}_{\mathbf{n}_{j\to j'}}\underline{(-1)^{|\mathbf{n}|_j+|\mathbf{n}|_{j'}} n_j(1-n_{j'})},$$

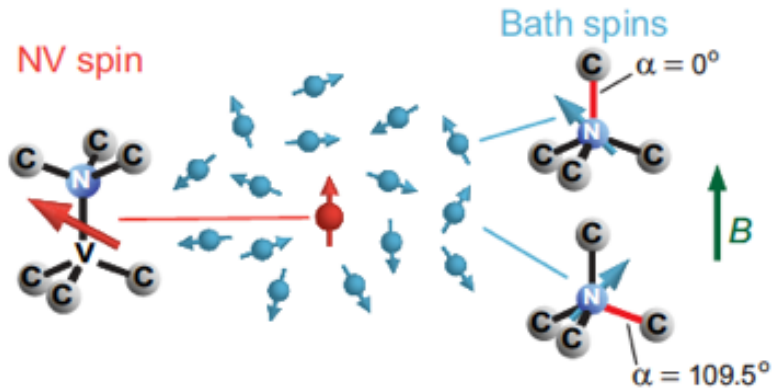
Pauli exclusion and negative sign due to anti-commutativity

finite-tiers for Fermions
correlation functions beyond exponential forms
application: dual fermion representation of spins

Hsieh and Cao, JCP 148, p014103 (2018); Early work on Fermion bath: Yan, Yu, Strunz, etc.

Spin-Based Quantum Devices

Nitrogen-Vacancy Spins in Diamond

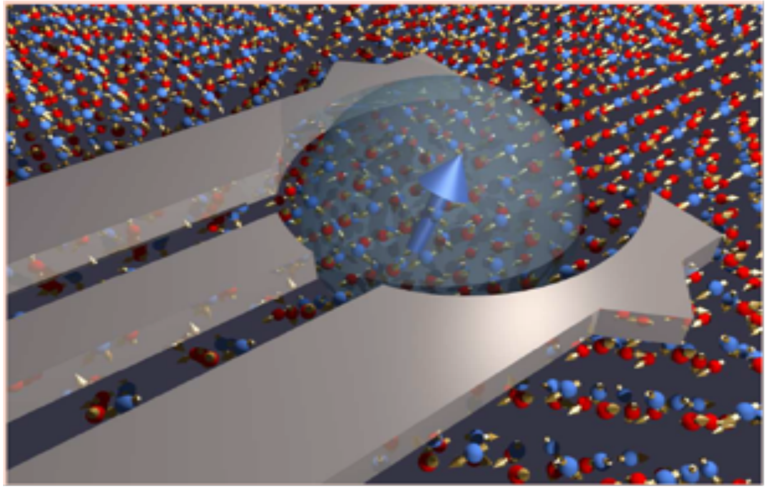


Applications:

quantum computing and
room-temperature ultra-
precision magnetic sensors.

Source of Spin Noises:
$1/r^3$ dipolar coupling to 10^1~10^2 impurity spins.

Spin-Based Qubit in Gated QM Dots



Coish et. al.

Source of Spin Noises:

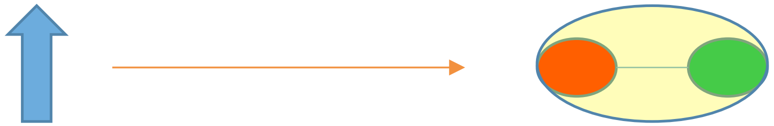
Hyperfine coupling and 10^4~10^6 nuclear spins

Casting Spin Bath as Fermionic Bath
Dual Fermion Representation

$$\sigma_k^x \Rightarrow \left(c_k^+ - c_k\right)\left(d_k^+ + d_k\right) \qquad \sigma_k^y \Rightarrow -i\left(c_k^+ + c_k\right)\left(d_k^+ + d_k\right) \qquad \sigma_k^z \Rightarrow -2\left(c_k^+ c_k - \frac{1}{2}\right)$$

C-fermion: Jordan-Wigner Transformation. Represent spin algebra with fermions

D-fermion: Correct the minus sign of fermion representations for multiple spins.

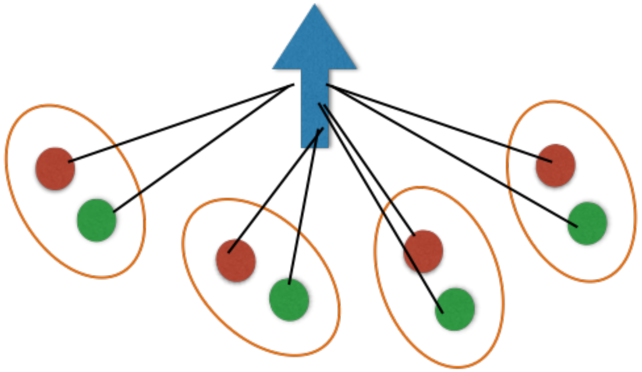


Phys. Rev. Lett. 91:207204, 2003.

The Effective Two-Bath Model

$$H = H_s + \sum_k \frac{\omega_k}{2}\sigma_k^z + \sigma_0^z \sum_k g_k \sigma_k^x$$

Dual-Fermion Mapping



$$H = H_s + \sum_k \omega_k \left(c_k^\dagger c_k - \frac{1}{2}\right) + i\sigma_0^z \sum_k g_k (c_k^\dagger + c_k)(d_k^\dagger + d_k)$$

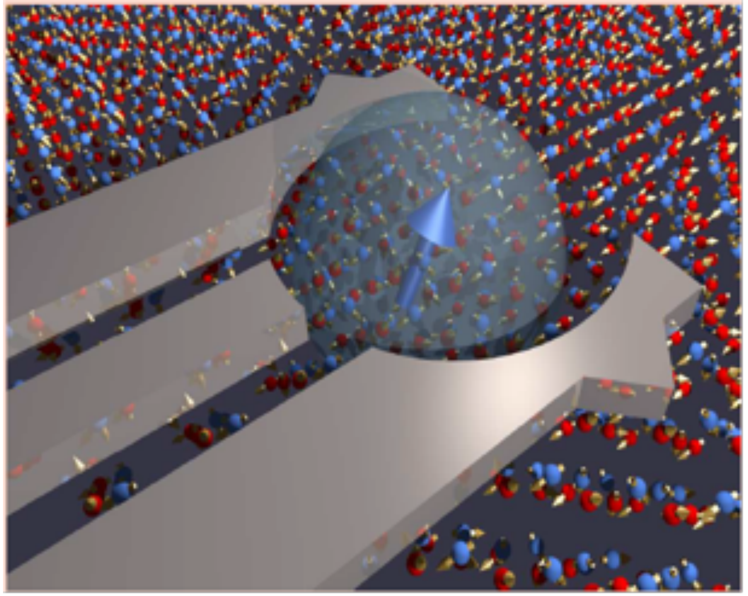
General Strategy:

1. Stochastically decouple C-Fermions and derive SLE for D-Fermions and Central spins.
2. Trace out the D-Fermions in SLE.
3. Formally average out the C-Fermions to obtain the spin equation.

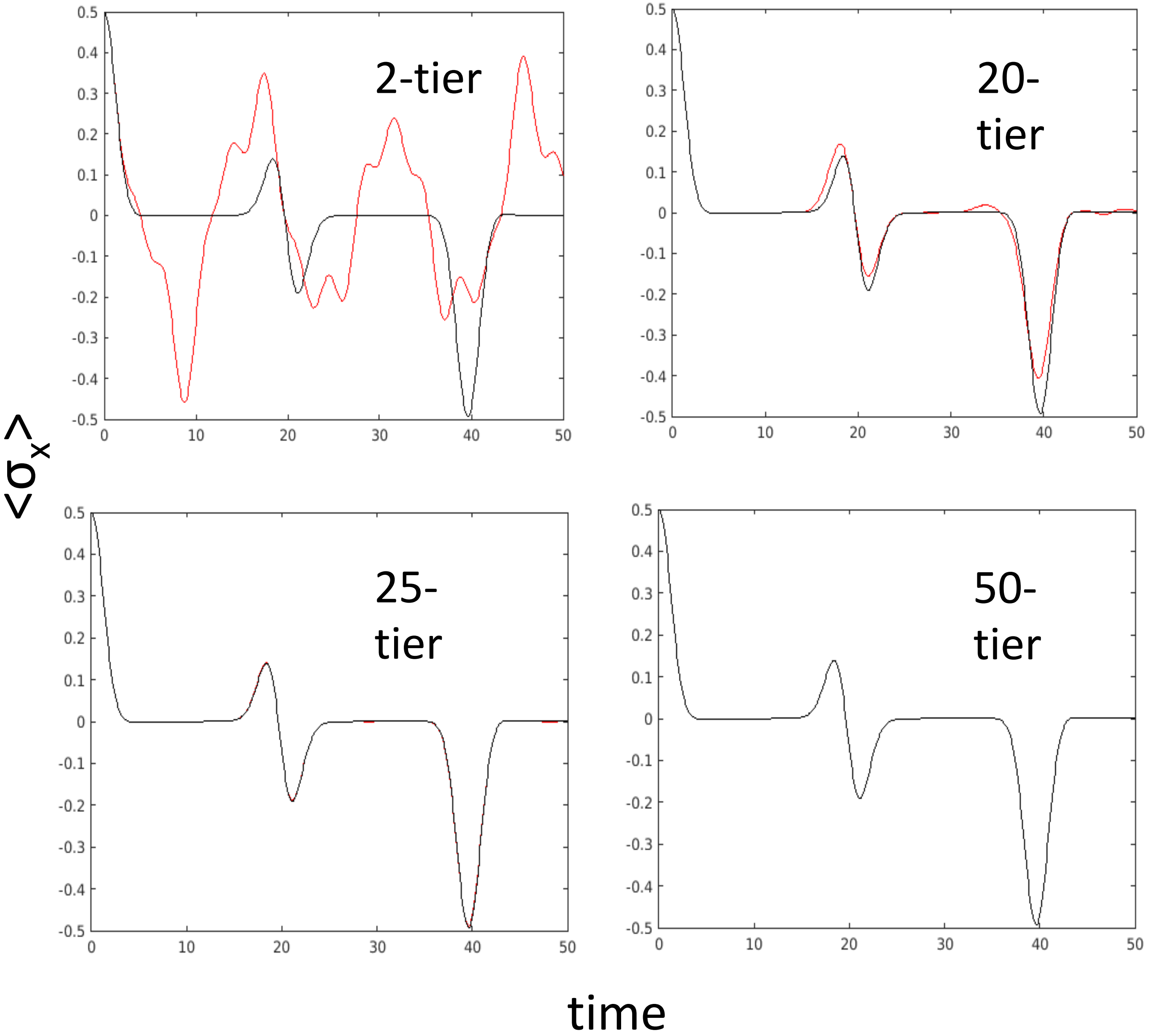
Hsieh and Cao, JCP 148, p014103 (2018)

Spin Bath: Fermionic Mapping

$$H = \frac{\omega_0}{2}\sigma_0^z + \sum_{k>0} \frac{\omega_k}{2}\sigma_k^z + \sigma_0^z \sum_{k>0} g_k \sigma_k^x$$



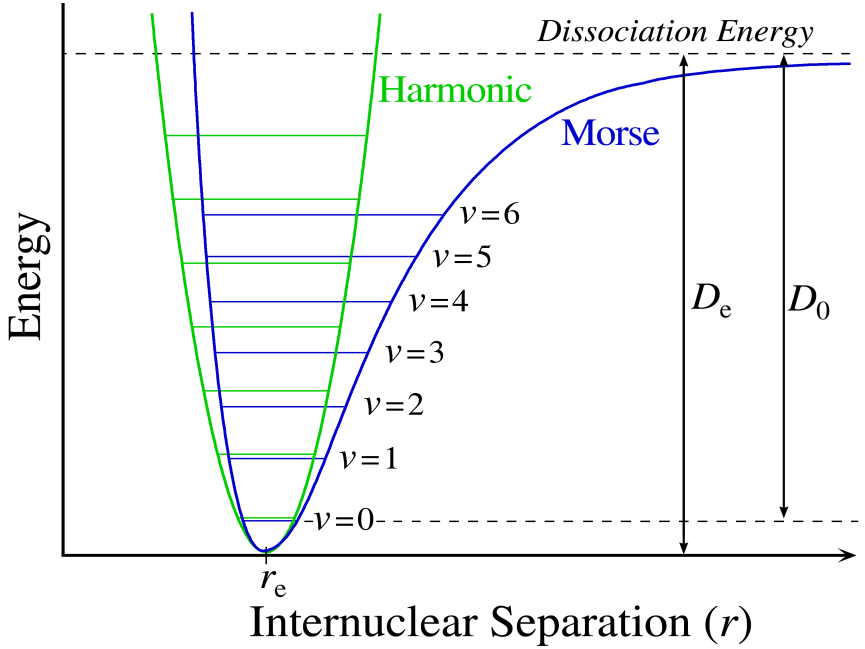
Coish et. al.



Condensed Phased Dynamics: Spin Bath as an Anharmonic Environment

anharmonic bath model

$$H = \frac{\epsilon}{2}\sigma_z + \Delta\sigma_x + \sum_{k>0}\left(\frac{1}{2}P_k^2 + D_k(1-e^{-\alpha_k X_k})^2\right) + \sigma_z \sum_{k>0} g_k X_k$$



Effective spin bath when only 2 bound states in each bath oscillator.

spin bath model

$$H = \frac{\epsilon}{2}\sigma_z + \Delta\sigma_x + \sum_{k>0}\frac{\Omega_k}{2}\sigma_k^z + \sigma_z\sum_{k>0}\tilde{g}_k\sigma_k^x$$

$$E_{v,k} = \omega_k\left(v+\frac{1}{2}\right) - \frac{\alpha_k^2}{2}\left(v+\frac{1}{2}\right)^2$$

Generalized Hierarchy Equation (GHE) : Anharmonic Bath

$$\partial_t \rho^{[\mathbf{A}_1][\mathbf{A}_2][\mathbf{A}_3]\cdots} = -i\left[H_s, \rho^{[\mathbf{A}_1][\mathbf{A}_2][\mathbf{A}_3]\cdots}\right] - i\sum_{n,m,\boldsymbol{j}} \chi_{\boldsymbol{j}}^{n+1,m}\left[A, \rho^{\cdots[\mathbf{A}_n+(m,\boldsymbol{j})]\cdots}\right]$$

$$-i\sum_{n,m,\mathbf{j}} \phi_{j_1}(0) A \rho^{\cdots[\mathbf{A}_{n-1}+(m',\boldsymbol{j}_1)][\mathbf{A}_n-(m,\boldsymbol{j})]\cdots} - i\sum_{n,m,\mathbf{j}} \phi_{j_1}(0) \rho^{\cdots[\mathbf{A}_{n-1}+(m',\boldsymbol{j}_1)][\mathbf{A}_n-(m,\boldsymbol{j})]\cdots} A$$

$$+\sum_{n,m,\mathbf{jj}'} \eta_{\mathbf{jj}'} \rho^{\cdots\left[a^n_{m\mathbf{j}} \to a^n_{m\mathbf{j}'}\right]\cdots}$$

(3-layers of hierarchy)

1. $[A_N]$ Block matrix accounts for the (N+1)-th cumulant expansion of the influence functional. In case of Gaussian bath, only $[A_1]$ contains non-zero elements.

2. N-th order cumulant contributions only emerge at (N-1)-th tier with closed system dynamics at zero-th tier.

3. When dealing with the Gaussian bath, the present approach reduces to the extended Hierarchical Equations of motions.

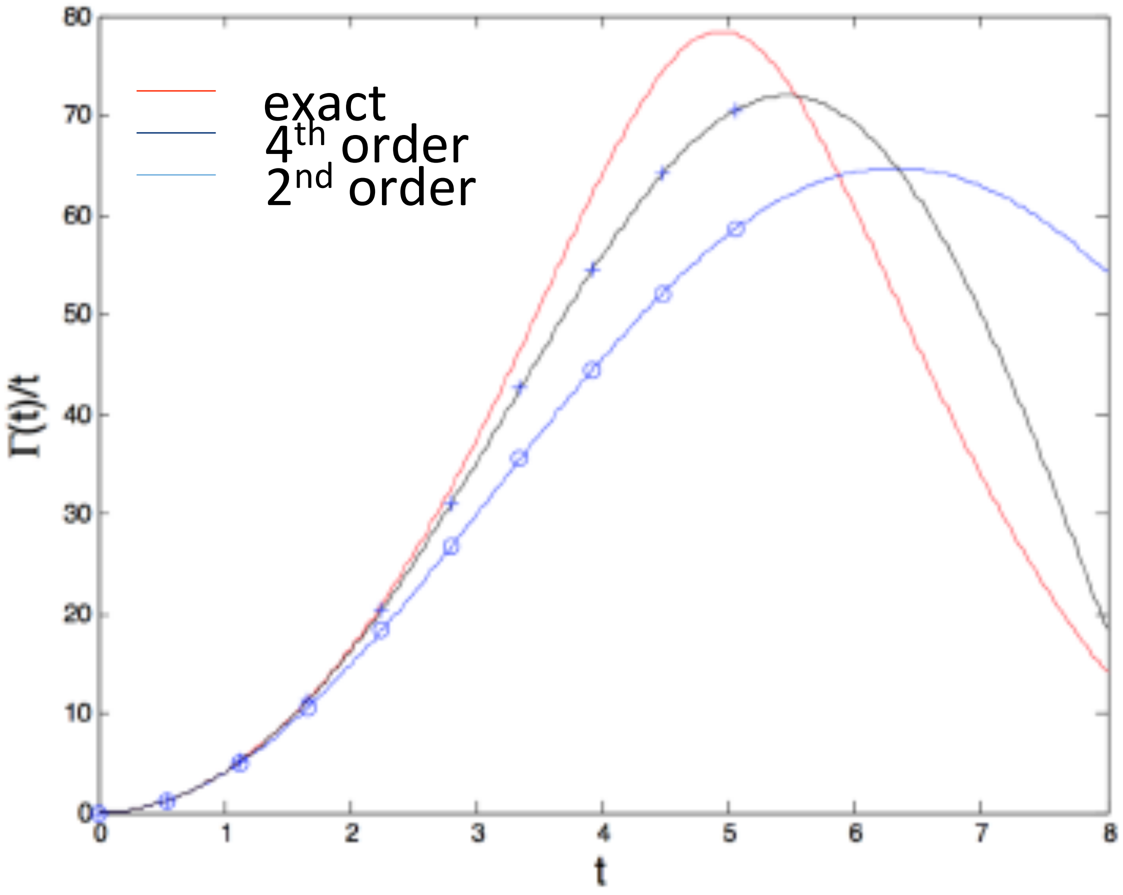
Spin Bath: 4-th Order Corrections

$$H = \frac{\omega_0}{2}\sigma_0^z + \sum_{k>0}\frac{\omega_k}{2}\sigma_k^z + \sigma_0^z\sum_{k>0} g_k\sigma_k^x$$

$$\langle\uparrow|\,\rho_s\,|\downarrow\rangle\,(t) = \langle\uparrow|\,\rho_s\,|\downarrow\rangle\,(0)e^{-i\epsilon t}e^{\Gamma(t)}$$

$$\Gamma(t) = \sum_k \ln\left[1 - \frac{4g_k^2}{\Omega_k^2}\left(1-\cos\Omega_k t\right)\right]$$

$$\Omega_k = \omega_k\sqrt{1+(4g_k^2/\omega_k^2)}.$$



Hsieh and Cao, JCP 148, p014104 (2018)

Summary

Paper I [JCP 148, p014103 (2018)]

The family of hierarchy equations provides a numerically exact description for generic quantum environments. Specifically, we derived hierarchy equations for Grassmann noise and non-Gaussian noise from the stochastic Liouville Equation.

Paper II [JCP 148, p014104 (2018)]

Spin bath is treated in two different approaches. Physical spins (such as nuclear spins) should be treated in the dual-fermion approach and go deep down the hierarchical tiers. Spin bath (as anharmonic condensed environment) is more conveniently handled by generalized hierarchy equation (GHE) approach, which goes beyond the linear response and the Gaussian assumption.

$$\tilde{\rho}(t) = |\psi^{+}(t)\rangle\langle\psi^{-}(t)|$$

SLE \ SW

GHE

Generalized hierarchy equation

1. Bosonic bath
2. Fermionic bath
3. Spin bath (dual Fermion)
4. non-Gaussian bath

SPI/SW

Stochastic path integrals

1. Imaginary time – thermal distribution
2. Absorption / Emission spectra
3. Multi-chromophor Forster rate

Hybrid

Deterministic + Stochastic

1. stochastic-HEOM
2. Transfer tensor method
   (PRL 112, p11040, 2014)

Hubbard-Stratonovich Transformation

1-Dimension (Gaussian integral):

$$\exp\left(a\frac{q^2}{4}\right) = \int d\xi \sqrt{\frac{1}{a\pi}} \exp\left(-\frac{1}{a}\xi^2 - q\xi\right)$$

N-Dimensions:

$$\exp\left(\frac{1}{4}\sum_i \sum_j q_i a_{ij} q_j\right) = \int \prod_i d\xi_i \left(\frac{1}{\det(a\pi)}\right)^{1/2} \exp\left(-\sum_i \sum_j \xi_i a_{ij}^{-1} \xi_j - \sum_i q_i \xi_i\right)$$

Infinite-Dimensions:

Influence Functional (non-local)

$$\exp\left(\frac{1}{4}\int_0^t dt' \int_0^t dt'' q(t') a(t'-t'') q(t'')\right)$$

$$= \int \mathcal{D}[\xi] \det\left(\frac{1}{a(t-t')\pi}\right)^{1/2} \exp\left(-\int_0^t dt' \int_0^t dt'' \xi(t') a(t-t')^{-1} \xi(t'') - \int_0^t dt' q(t') \xi(t')\right)$$

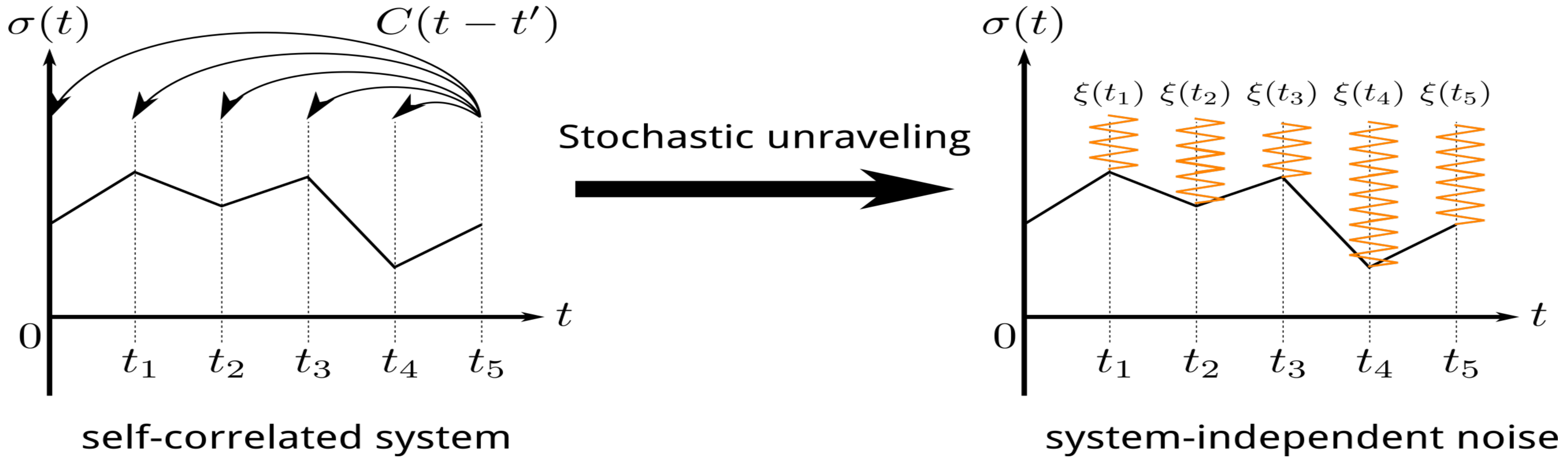
Auxiliary field

Time-local and linear

Stochastic Unraveling of Bath Entanglement

Hubbard-Stratonovich transformation

$$F[\sigma] = \prod_{m=1}^{N_A} \int \mathcal{D}[\xi_m] w_m \exp\left[-\frac{1}{2\hbar}\int_0^t dt' \int_0^t dt'' \xi_m(t')\xi_m(t'') C_m^{-1}(t'-t'') + \frac{i}{\hbar}\int_0^t dt' V_m^{A}(\sigma(t'))\xi_m(t')\right]$$



Stockburger, Grabert, *Phys. Rev. Lett.* 88, 170407 (2002)

Full Real-Time Dynamics

Stochastic unraveling of the influence functional leads to:

1. Monte-Carlo wave-function for mixed states

   J. Stockburger and H. Grabert, PRL, 88, 170407, 2002.

2. Non-Markovian quantum state diffusion (NMQSD) for pure states

   L. Diosi, N. Gisin, and W. Strunz, PRA, 58, 1699, 1998.

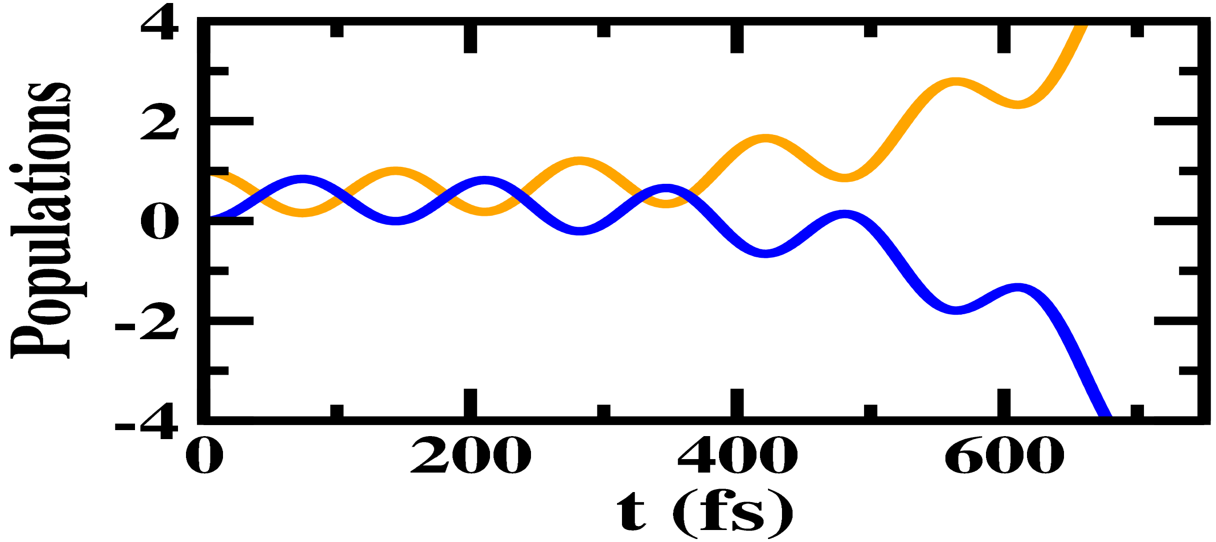
3. Stochastic sampling of harmonic baths

   J. Cao, L. W. Ungar, and G. A. Voth, J. Chem. Phys. **104**, 4189, 1996

Etc....

These methods are formally exact but do not converge well numerically

Divergence of real-time simulation and Solutions



The complex noise leads to divergence at long times

1) Hybrid Approach: combine stochastic dynamics and deterministic equation s-HEOM [Moix and Cao, JCP 139, 134106, (2013)]: use a fictitious temperature as a reference and sample the difference using stochastic path integrals.

2) Combination with transfer tensor method (PRL 112, p11040, 2014)

Hybrid Stochastic-Deterministic Dynamics

Proposal of simple partition:

1. Treat real fluctuations stochastically

   Generalized Haken-Strobl Model

2. Treat dissipation deterministically

   HEOM or QUAPI

Best of both methods:

- HEOM and QUAPI are difficult at low T
- Complex noise from dissipative part ruins pure stochastic methods

Simple Partition

Simple Stochastic Hierarchy Equations of Motion:

1. Hubbard-Stratonovich transformation of C_r
2. Hierarchy development of C_i

$$\dot{\rho}_0(t) = -i\hat{H}(t)^{\times}\rho_0(t) - i\hat{q}^{\times}\rho_1(t)$$

$$\dot{\rho}_n(t) = -i\hat{H}(t)^{\times}\rho_n(t) - n\omega_c\rho_n(t) - i\hat{q}^{\times}\rho_{n+1}(t) - n\lambda\omega_c\hat{q}^{\circ}\rho_{n-1}(t)$$

$$\hat{H}(t) = \hat{H}_s + \xi(t)\hat{q} \qquad \langle\xi(t)\xi(t')\rangle = C_r(t-t')/\hbar$$

Advantages:

1. T-independent hierarchy
2. Very efficient Monte Carlo convergence

Disadvantages:

1. Drude-Lorentz spectral density
2. Density matrix not wave-functions

Real-time Path Integral

Real-Time Propagator:

$$U(x',y';x,y;t) = \int \mathcal{D}[x_s]\mathcal{D}[y_s]e^{\frac{i}{\hbar}(S_s[x_s]-S_s[y_s])-\frac{1}{\hbar}\Phi[x_s,y_s]}$$

Influence Functional:

$$\Phi[x,y] = \underline{\int_0^t dt' \int_0^{t'} dt''\,[x(t')-y(t')]\,C_r(t'-t'')\,[x(t'')-y(t'')]}$$
$$+\,i\,\underline{\int_0^t dt' \int_0^{t'} dt''\,[x(t')-y(t')]\,C_i(t'-t'')\,[x(t'')+y(t'')]}$$

$$C_r(t) = \int_0^\infty \frac{d\omega}{\pi} J(\omega)\coth(\hbar\beta\omega/2)\cos(\omega t)$$

$$C_i(t) = \int_0^\infty \frac{d\omega}{\pi} J(\omega)\sin(\omega t)$$

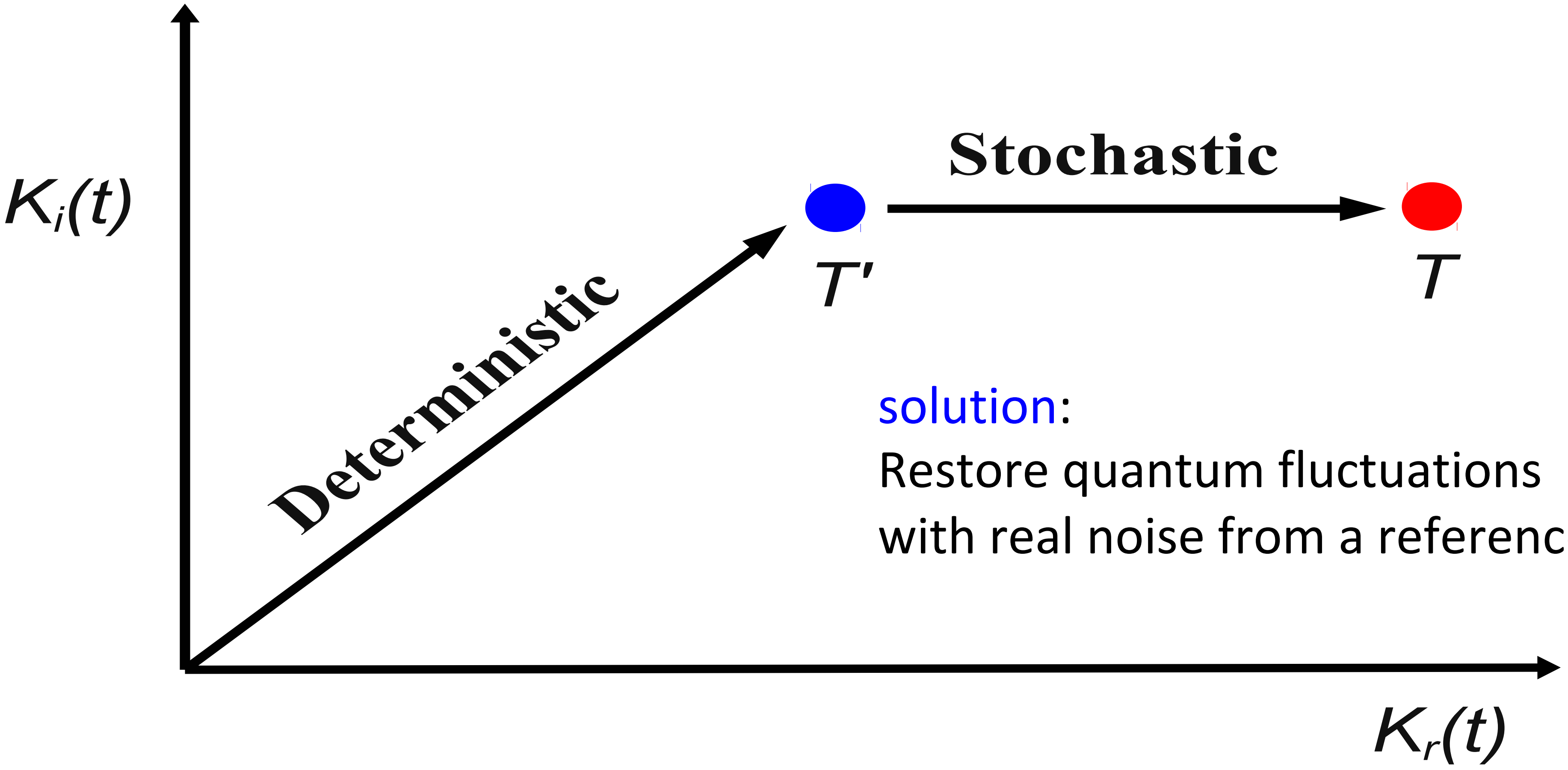
Fluctuations:

1. Purely Real
2. Increase with T/h

Dissipation:

1. Complex force
2. T-Independent

Stochastic HEOM



solution:
Restore quantum fluctuations
with real noise from a reference T'

J. Moix and J. Cao, JCP, 139, 134106, 2013: Related work by Strunz, Shao, etc.

Example I. TLS Energy Transfer Rates

$$P_1(t) = \frac{\chi_{eq} + e^{-(1+\chi_{eq})kt}}{1 + \chi_{eq}}$$

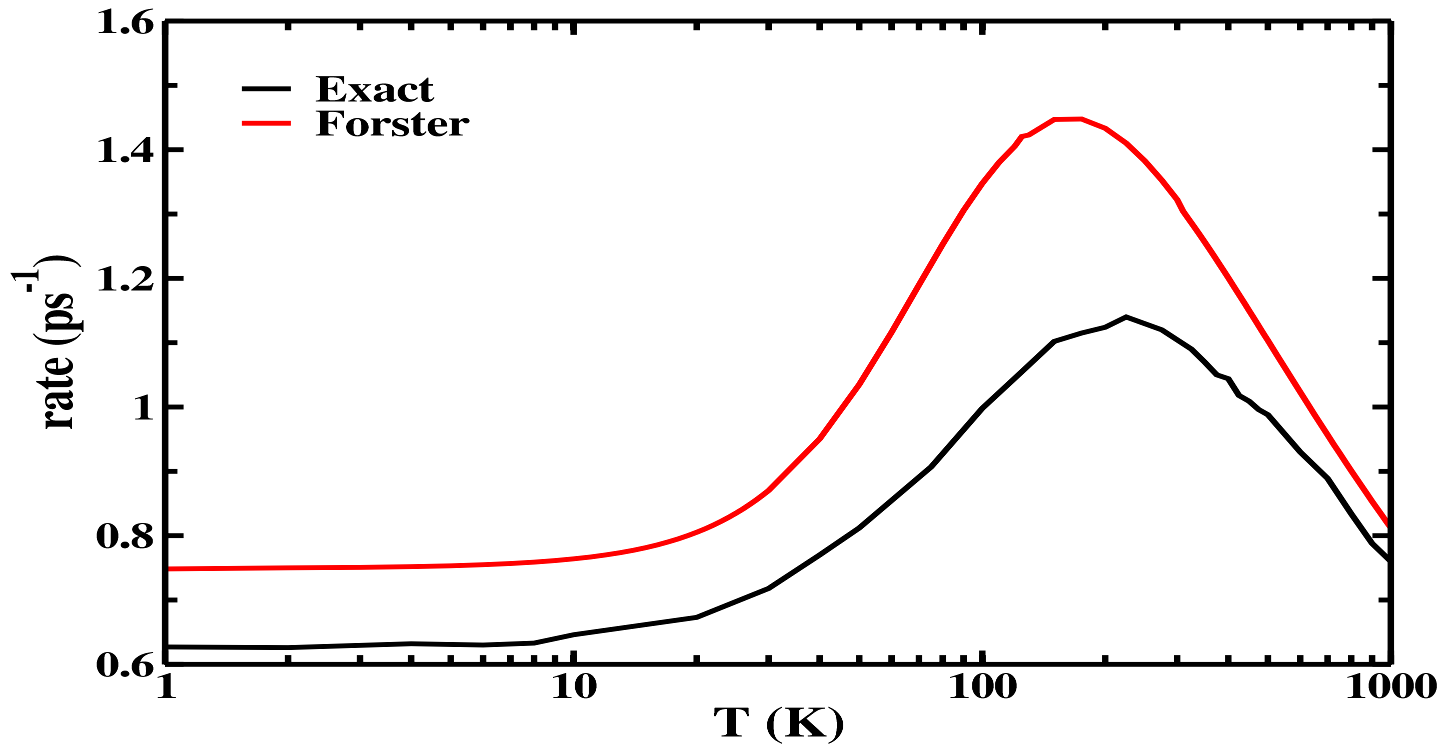
$$\chi_{eq} = P_1^{eq} / P_2^{eq}$$

$$\Delta = 50 \text{ cm}^{-1}$$

$$J = 20 \text{ cm}^{-1}$$

$$\lambda = 20 \text{ cm}^{-1}$$

$$\omega_c = 53 \text{ cm}^{-1}$$



Need higher order correction to FGR

J. Moix and J. Cao, JCP, 139, 134106, 2013

Example II: Concurrence Dynamics

Two Qubit Hamiltonian:

$$H_s = \omega_0\left(\sigma_1^z + \sigma_2^z\right) + J\sigma_1^x\sigma_2^x$$

$$H_{sb} = \sum_{k=1}^{2}\sigma_k^x\sum_j c_{kj}\left(b_{kj} + b_{kj}^\dagger\right)$$

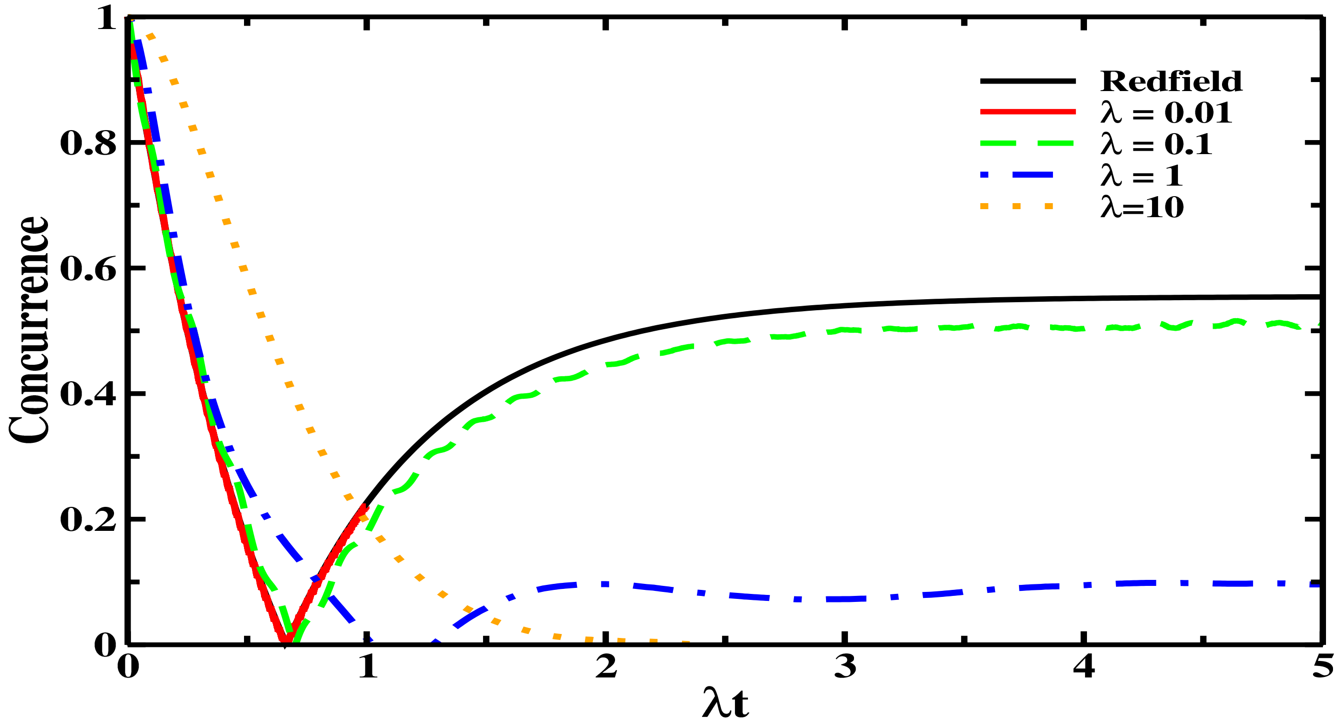
$$\omega_0 = 1.5J$$
$$\omega_c = 3J$$
$$\beta J = 50$$

Concurrence:

$$C = \max\left(0, \sqrt{\lambda_1} - \sqrt{\lambda_2} - \sqrt{\lambda_3} - \sqrt{\lambda_4}\right)$$

λ: sorted eigenvalues of spin-flipped state:

$$\tilde{\rho} = \left(\sigma_1^y \otimes \sigma_2^y\right)\rho^*\left(\sigma_1^y \otimes \sigma_2^y\right)$$



J. Moix and J. Cao, JCP, 139, 134106, 2013

Conclusion

A family of hierarchy equations are obtained from the SLE.

The hybrid stochastic-deterministic approach is promosing

Coworkers: Changyu Hsieh, Jeremy Moix

Funding: NSF and SMART

Thanks you for your attention!