Correlated Multivariant Ornstein-Uhlenbeck Process

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The Ornstein-Uhlenbeck process is a very useful method to account for many Markovian stochastic processes. It's multivariate representation is even more practical for physical processes. Here we discuss the multivariate Ornstein-Uhlenbeck process including correlated Wiener processes, for the purpose of tackling realistic physical problems such as chromorphores coupled to their respective phonon environments but interacting with a common bath.

Stochastic Processes Adapt Correlated Brownian Motions

We define the Ornstein-Uhlenbeck process $\mathbf{X}(t)$ in the vector form

$$d\mathbf{X}(t) = -A\mathbf{X}(t)dt + Bd\mathbf{W}(t), \tag{1}$$

in which A and B are coefficient matrices. $\mathbf{W}(t)$ is the Wiener process vector which is correlated through the correlation matrix

$$\rho(t, t') \equiv \delta_{tt'} d\mathbf{W}(t) d\mathbf{W}(t')^{\mathrm{T}}/dt. \tag{2}$$

The matrix elements are $\rho_{ij} = dW_i(t)dW_j(t)/dt$, called the Itô isometry in higher dimensions. Obviously $\rho_{ii} = 1$ according to the quadratic variation $(dW_t)^2 = dt$.

$\rho_{ij} \in [-1, 1]$. Anti-correlated∼non-correlated∼fully correlated.

According to the Itô's lemma, one can find the solution

$$\mathbf{X}(t) = e^{-At}\mathbf{X}(0) + \int_0^t e^{-A(t-t')} B d\mathbf{W}(t'), \tag{3}$$

where $\mathbf{X}(0)$ is the initial condition of the process $\mathbf{X}(t)$, the mean

$$\langle \mathbf{X}(t) \rangle = e^{-At} \langle \mathbf{X}(0) \rangle, \tag{4}$$

and the correlation function between time t and s is

$$\begin{aligned} \left\langle \mathbf{X}_t, \mathbf{X}_s^{\mathrm{T}} \right\rangle \equiv & \left\langle \left[\mathbf{X}_t - \langle \mathbf{X}_t \rangle\right] \left[\mathbf{X}_t - \langle \mathbf{X}_t \rangle\right]^{\mathrm{T}} \right\rangle \\ = & e^{-At} \left\langle \mathbf{X}(0), \mathbf{X}^{\mathrm{T}}(0) \right\rangle e^{-A^{\mathrm{T}} s} \\ & + \int_0^{\min(s,t)} e^{-A(t-t')} B \rho B^{\mathrm{T}} e^{-A^{\mathrm{T}}(s-t')} dt'. \end{aligned} \tag{5}$$

Eigenstate Relaxation

If $AA^{\mathrm{T}} = A^{\mathrm{T}}A$, for deterministic initial condition, one can find a unitary matrix S to diagonalize the coefficient matrix $SAS^\dagger = SA^{\mathrm{T}}S^\dagger = diag(\gamma_1, \gamma_2, \dots, \gamma_n)$, so does the correlation function $\langle \mathbf{X}_t, \mathbf{X}_s^{\mathrm{T}} \rangle = S^\dagger G(t,s) S$, where

$$[G(t,s)]_{ij} = \frac{\left(B\rho B^{\mathrm{T}}\right)_{ij}}{\gamma_i + \gamma_j} \left[e^{-\gamma_i |t-s|} - e^{-\gamma_i t - \gamma_j s} \right] \qquad (t \geq s), \quad (6)$$

$$[G(t,s)]_{ij} = \frac{\left(B\rho B^{\mathrm{T}}\right)_{ij}}{\gamma_i + \gamma_j} \left[e^{-\gamma_j |t-s|} - e^{-\gamma_i t - \gamma_j s} \right] \qquad (t \leq s). \quad (7)$$

If the real parts of all A's eigenvalues are positive, one may find the stationary solution

$$\mathbf{X}_{\mathrm{s}}(t) = \int_{-\infty}^{t} e^{-A(t-t')} B d\mathbf{W}(t'), \tag{8}$$

and the correlation matrix

$$\left\langle \mathbf{X}_{\mathrm{s}}(t), \mathbf{X}_{\mathrm{s}}^{\mathrm{T}}(s) \right\rangle = \int_{-\infty}^{\min(s,t)} e^{-A(t-t')} B \rho B^{\mathrm{T}} e^{-A^{\mathrm{T}}(s-t')} d\mathbf{W}(t'). \tag{9}$$

Stationary Covariance and Correlation Function

We define the stationary covariance matrix

$$\sigma = \left\langle \mathbf{X}_{\rm s}(t), \mathbf{X}_{\rm s}^{\rm T}(t) \right\rangle, \tag{10}$$

then find a useful algebraic equation for stationary covariance matrix

$$A\sigma + \sigma A^{\rm T} = B\rho B^{\rm T}. \tag{11}$$

For $s < t$ the stationary correlation function can be written as

$$\begin{aligned} \left\langle \mathbf{X}_{\rm s}(t), \mathbf{X}_{\rm s}^{\rm T}(s) \right\rangle &= e^{-A(t-s)} \int_{-\infty}^{s} e^{-A(s-t')} B\rho B^{\rm T} e^{-A^{\rm T}(s-t')} dt' \\ &= e^{-A(t-s)}\sigma \qquad s < t, \end{aligned} \tag{12}$$

and

$$= \sigma e^{-A^{\rm T}(s-t)} \qquad s > t. \tag{13}$$

Regression Theorem

- The correlation function only depends on the time difference $|t-s|$ as expected for the stationary solution.
- Define stationary correlation matrix $G_{\mathrm{s}}(\tau)=\left\langle \mathbf{X}_{\mathrm{s}}(t),\mathbf{X}_{\mathrm{s}}^{\mathrm{T}}(t-\tau)\right\rangle$, obviously $G_{\mathrm{s}}(0)=\sigma$.
- The time evolution for $\tau>0$ is $G_{\mathrm{s}}(\tau)=e^{-A\tau}G_{\mathrm{s}}(0)$, same as that of the mean $\langle \mathbf{X}(t)\rangle = e^{-At}\langle \mathbf{X}(0)\rangle$.

$$\frac{d}{d\tau}\left[G_{\mathrm{s}}(\tau)\right]=\frac{d}{d\tau}\left\langle \mathbf{X}_{\mathrm{s}}(\tau),\mathbf{X}_{\mathrm{s}}^{\mathrm{T}}(0)\right\rangle=-AG_{\mathrm{s}}(\tau). \tag{14}$$

- A consequence of the Markovian linear nature of the process.
- Since $\sigma^{\mathrm{T}}=\sigma$, we have $G_{\mathrm{s}}(\tau)=\left[G_{\mathrm{s}}(-\tau)\right]^{\mathrm{T}}$.

Spectrum Matrix in Stationary State

One can find the spectrum matrix as the Fourier transform of the autocorrelation matrix $G_{\mathrm{s}}(\tau)$

$$
\begin{aligned}
S(\omega) &= \frac{1}{2\pi}\int_{-\infty}^{\infty} e^{-i\omega\tau}\, G_{\mathrm{s}}(\tau) d\tau \\
&= \frac{1}{2\pi}\left[(A+i\omega)^{-1}\sigma + \sigma\left(A^{\mathrm{T}} - i\omega\right)^{-1}\right] \\
&= \frac{1}{2\pi}\,(A+i\omega)^{-1}\, B\rho B^{\mathrm{T}}\,(A-i\omega)^{-1}\,. \qquad (15)
\end{aligned}
$$

2D Ornstein-Uhlenbeck Process - 1

- 2D Ornstein-Uhlenbeck SDEs

$$dX_1(t) = -\gamma_1 X_1(t)dt + \sigma_{11} dW_1(t) + \sigma_{12} dW_2(t),$$
$$dX_2(t) = -\gamma_2 X_2(t)dt + \sigma_{21} dW_1(t) + \sigma_{22} dW_2(t).$$

- Two Wiener processes $dW_1(t)$ and $dW_2(t)$ are correlated through $\rho = dB_1(t)dB_2(t)/dt$.
- $\rho \in [-1, 1]$ covers from complete anti-correlation to full correlation. $\rho = 0$ is for the completely decoupled case.
- The solutions

$$X_1(t) = e^{-\gamma_1 t} X_1(0) + \sigma_{11} \int_0^t e^{-\gamma_1(t-s)} dW_1(s) + \sigma_{12} \int_0^t e^{-\gamma_1(t-s)} dW_2(s),$$
$$X_2(t) = e^{-\gamma_2 t} X_2(0) + \sigma_{21} \int_0^t e^{-\gamma_2(t-s)} dW_1(s) + \sigma_{22} \int_0^t e^{-\gamma_2(t-s)} dW_2(s).$$

2D Ornstein-Uhlenbeck Process - 2

- The means

$$\langle X_1(t)\rangle = \langle X_1(0)\rangle\, e^{-\gamma_1 t},$$
$$\langle X_2(t)\rangle = \langle X_2(0)\rangle\, e^{-\gamma_2 t}.$$

- The spectrum matrix

$$S(\omega) = \frac{1}{2\pi}\begin{bmatrix} \frac{\sigma_{11}^2+2\rho\sigma_{11}\sigma_{22}+\sigma_{12}^2}{\gamma_1^2+\omega^2} & \frac{\sigma_{12}\sigma_{22}+\sigma_{11}\sigma_{21}+\rho(\sigma_{12}\sigma_{21}+\sigma_{11}\sigma_{22})}{(\gamma_1+i\omega)(\gamma_2-i\omega)} \\ \frac{\sigma_{12}\sigma_{22}+\sigma_{11}\sigma_{21}+\rho(\sigma_{12}\sigma_{21}+\sigma_{11}\sigma_{22})}{(\gamma_1-i\omega)(\gamma_2+i\omega)} & \frac{\sigma_{21}^2+2\rho\sigma_{21}\sigma_{22}+\sigma_{22}^2}{\gamma_2^2+\omega^2} \end{bmatrix}.$$

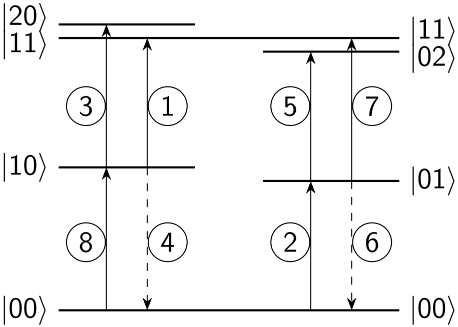
The correlation functions

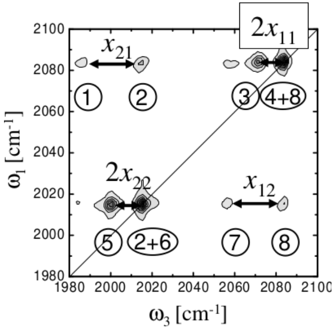
$$\mathrm{Cov}\left[X_1(t), X_1(s)\right] = \left\langle X_1(0)^2 \right\rangle e^{-\gamma_1(t+s)} + \frac{\sigma_{11}^2 + \sigma_{12}^2 + 2\rho\sigma_{11}\sigma_{12}}{2\gamma_1}\left[e^{-\gamma_1|t-s|} - e^{-\gamma_1(t+s)}\right],$$

$$\mathrm{Cov}\left[X_2(t), X_2(s)\right] = \left\langle X_2(0)^2 \right\rangle e^{-\gamma_2(t+s)} + \frac{\sigma_{21}^2 + \sigma_{22}^2 + 2\rho\sigma_{21}\sigma_{22}}{2\gamma_2}\left[e^{-\gamma_2|t-s|} - e^{-\gamma_2(t+s)}\right],$$

$$\begin{aligned}\mathrm{Cov}\left[X_1(t), X_2(s)\right] &= \langle X_1(0), X_2(0)\rangle\, e^{-\gamma_1 t - \gamma_2 s} \\ &\quad + \frac{\sigma_{11}\sigma_{21} + \sigma_{12}\sigma_{22} + \rho\sigma_{11}\sigma_{22} + \rho\sigma_{12}\sigma_{21}}{\gamma_1 + \gamma_2} e^{-\gamma_1 t - \gamma_2 s}\left[e^{(\gamma_1+\gamma_2)\min(s,t)} - 1\right], \\ \mathrm{Cov}\left[X_2(t), X_1(s)\right] &= \langle X_1(0), X_2(0)\rangle\, e^{-\gamma_2 t - \gamma_1 s} \\ &\quad + \frac{\sigma_{11}\sigma_{21} + \sigma_{12}\sigma_{22} + \rho\sigma_{11}\sigma_{22} + \rho\sigma_{12}\sigma_{21}}{\gamma_1 + \gamma_2} e^{-\gamma_2 t - \gamma_1 s}\left[e^{(\gamma_1+\gamma_2)\min(s,t)} - 1\right].\end{aligned}$$

2D Spectroscopy of Coupled Oscillators





- ◇ Oscillator anharmonicity ∼ diagonal peak splitting.
- ◇ Excited state coupling ∼ off-diagonal peak splitting.

[1] Hamm, P. and Zanni, M., *Concepts and Methods of 2D Infrared Spectroscopy*. Cambridge (2011)

[2] M. Khalil, N. Demirdöven, and A. Tokmakoff, *Phy. Rev. Lett.* **90**, 047401 (2003)

Model for Coupled Oscillators

$$\hat{H}_0(t) = \sum_i \hbar\omega_i(t)\hat{a}_i^\dagger \hat{a}_i + \sum_{i,j} g_{ij}\hat{a}_i^\dagger \hat{a}_j + \sum_i \frac{U_i}{2}\hat{a}_i^\dagger \hat{a}_i^\dagger \hat{a}_i \hat{a}_i$$

- ⋆ $\omega_i(t) = \omega_i + \delta\omega_i(t)$, $\langle \delta\omega_i(t) \rangle = 0$.
- ⋆ Linear correlation is frequency independent.
- ⋆ U_i is the exciton scattering/repulsion coefficient.
- ⋆ Fluctuation of the energy levels $\delta\omega_i(t)$ is governed by correlated multivariate Ornstein-Uhlenbeck process

$$\begin{aligned}
\delta\omega_1(t) &= -\gamma_1\delta\omega_1(t)dt + \sigma_{11}dW_1(t) + \sigma_{12}dW_2(t), \\
\delta\omega_2(t) &= -\gamma_2\delta\omega_2(t)dt + \sigma_{21}dW_1(t) + \sigma_{22}dW_2(t).
\end{aligned}$$

Interaction Picture

$$\hat{a}_0(t) = \exp\left[\frac{i}{\hbar}\int_0^t \hat{H}_0(\tau)d\tau\right]\hat{a}_0 \exp\left[-\frac{i}{\hbar}\int_0^t \hat{H}_0(\tau)d\tau\right].$$

Turn off the coupling between oscillators.

$$\hat{a}_i(t) = \exp\left[-iU_i t\hat{n}_i - i\int_0^t \omega_i(\tau)d\tau\right]\hat{a}_i(0).$$

Linear Response Function

$$
\begin{aligned}
S^{(1)} =& i \left\langle [\hat{\mu}(t), \hat{\mu}(0)]\, \rho(-\infty) \right\rangle \\
=& i \left\langle \sum_i \mu_i^2 \left[\left(e^{iU_i t} - 1 \right) \hat{a}_i^\dagger \hat{a}_i^\dagger + \left(e^{-iU_i t} - 1 \right) \hat{n}_i - 1 \right] \right. \\
& \left. \times \exp\left[i \int_0^t \omega_i(\tau) d\tau + iU_i t \hat{n}_i \right] \rho(-\infty) + c.c \right\rangle
\end{aligned}
$$

where $\omega_i(\tau) = \omega_i + \delta\omega_i(\tau)$, and $\hat{n}_i = \hat{a}_i^\dagger \hat{a}_i$.

Cumulant expansion ......