Supplementary information: Data-driven Coarse-grained Modeling of Non-equilibrium Systems

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1 Tikhonov regularization

As discussed in the paper, the construction of the force-momentum correlation function $D(t',t)$ or the memory kernel $K(t',t)$ in the discrete setting may yield an ill-conditioned linear system. Here not that when constructing $D(t',t) = \frac{\partial C(t',t)}{\partial t}$, we can convert it to $\int_{t'}^{t} D(t',t'')dt'' = C(t',t) - C(t',t')$, which can be also written as a linear system by applying some quadrature rules. In our paper, we choose the midpoint rule. The Tikhonov regularization[1,2] for a general ill-conditioned linear system can be expressed as:

$$(\boldsymbol{\mathcal{M}}_n^T \boldsymbol{\mathcal{M}}_n + \mu)\mathbf{f}_n = \boldsymbol{\mathcal{M}}_n^T \mathbf{g}_n \ , \tag{1}$$

where $\boldsymbol{\mathcal{M}}_n \in \mathbb{R}^{n\times n}$, $\mathbf{f}_n \in \mathbb{R}^{n\times 1}$, and $\mathbf{g}_n \in \mathbb{R}^{n\times 1}$. In the construction of $D(t',t)$, $\boldsymbol{\mathcal{M}}_n^{i,j} = \Delta t$ for $j \leq i$ (otherwise, $\boldsymbol{\mathcal{M}}_n^{i,j} = 0$), $\mathbf{f}_n^i = D(t'_n, t_{n+i-\frac{1}{2}})$, and $\mathbf{g}_n^i = C(t'_n, t_{n+i}) - C(t'_n, t'_n)$; while in the construction of $K(t',t)$, $\boldsymbol{\mathcal{M}}_n^{i,j} = \frac{1}{2}\Delta t(C(t'_i, t_{j-1}) + C(t'_i, t_j))$ for $i \leq j$ (otherwise, $\boldsymbol{\mathcal{M}}_n^{i,j} = 0$), $\mathbf{f}_n^i = K(t'_{i-\frac{1}{2}}, t_n)$, and $\mathbf{g}_n^i = -D(t'_{i-1}, t_n)$

In the following, we discuss how to determine the regularization parameter μ in Eq. (1). While a larger μ leads to a larger approximation error, a smaller μ results in weaker regularization and hence a less stable solution. Thus, choosing an appropriate value of μ is critical in the Tikhonov regularization, which has been widely studied in literature[3–5]. The principles followed to choose appropriate μ fall into either of the two categories according to whether they are noise-dependent or not. Considering the difficulty to accurately evaluate the noise in $D(t',t)$ after numerical differentiation, we can adopt a noise-independent principle to determine μ, e.g., based on the quasi-optimality criterion[5,6], which is described as follows.

Denoting the noisy data with the subscript δ, e.g., $\boldsymbol{\mathcal{M}}_{n,\delta}$ and $\mathbf{g}_{n,\delta}$, the regularized solution of Eq. (1) with parameter μ can be written as:

$$\mathbf{f}_{n,\delta}^{\mu} = (\boldsymbol{\mathcal{M}}_{n,\delta}^T \boldsymbol{\mathcal{M}}_{n,\delta} + \mu)^{-1} \boldsymbol{\mathcal{M}}_{n,\delta}^T \mathbf{g}_{n,\delta} \ . \tag{2}$$

We consider a geometric sequence of regularization parameters:

$$\mu_l := \mu_0 \eta^l, \ \ l \in \mathbb{N} \ , \tag{3}$$

for a fixed $\eta < 1$ and $\mu_0 > 0$. The regularization parameter by the quasi-optimality criterion: $\mu = \mu_{l^*}$, can then be obtained from:

$$l^* = \underset{l\geq 0}{\operatorname{argmin}} \, \|\mathbf{f}_{n,\delta}^{\mu_l} - \mathbf{f}_{n,\delta}^{\mu_{l+1}}\| \ , \tag{4}$$

where $\|\cdot\|$ denotes a norm, and L_2 norm is used herein. More details about the quasi-optimality criterion can be found in[7–9]. The convergence of the Tikhonov regularization combined with the quasi-optimality principle for regularizing ill-conditioned linear systems is discussed in[10].

Note that since the noise level in the data of $C(t',t)$ can be easily estimated, a noise-dependent principle, such as the Morozov's discrepancy principle[3,11], can also be adopted to regularize the ill-conditioned numerical differentiation for obtaining $D(t',t)$ from $C(t',t)$.

S.W. (Shu Wang) and Z.M. (Zhan Ma) contributed equally to this work

2 Parameter matrices in the extended dynamics

The parameter matrices $\mathbf{A}_{ps} \in \mathbb{R}^{2N\times 1}$, $\mathbf{A}_{sp} \in \mathbb{R}^{1\times 2N}$, and $\mathbf{A}_{ss} \in \mathbb{R}^{2N\times 2N}$ are assembled as:

$$\begin{aligned}
\mathbf{A}_{ps} &= [\mathbf{A}_{ps,1}, \mathbf{A}_{ps,2}, \ldots, \mathbf{A}_{ps,N}], \\
\mathbf{A}_{sp} &= [\mathbf{A}_{sp,1}, \mathbf{A}_{sp,2}, \ldots, \mathbf{A}_{sp,N}]^T, \\
\mathbf{A}_{ss} &= \begin{bmatrix} \mathbf{A}_{ss,1} & 0 & \cdots & 0 \\ 0 & \mathbf{A}_{ss,2} & \cdots & 0 \\ \vdots & \vdots & \ddots & \vdots \\ 0 & 0 & \cdots & \mathbf{A}_{ss,N} \end{bmatrix} ,
\end{aligned} \tag{5}$$

where

$$\begin{aligned}
\mathbf{A}_{ps,i} &= \left[\sqrt{\frac{b_i}{2} - \frac{q_i c_i}{a_i}},\ \sqrt{\frac{b_i}{2} + \frac{q_i c_i}{a_i}}\right], \\
\mathbf{A}_{sp,i} &= \left[-\sqrt{\frac{b_i}{2} - \frac{q_i c_i}{a_i}},\ -\sqrt{\frac{b_i}{2} + \frac{q_i c_i}{a_i}}\right]^T, \\
\mathbf{A}_{ss,i} &= \begin{bmatrix} a_i & \frac{1}{2}\sqrt{4q_i^2 + a_i^2} \\ -\frac{1}{2}\sqrt{4q_i^2 + a_i^2} & 0 \end{bmatrix} .
\end{aligned} \tag{6}$$

Here, we can see $\mathbf{A}_{sp}^T = -\mathbf{A}_{ps}$. The matrix $\boldsymbol{\alpha}(t) \in \mathbb{R}^{2N\times 2N}$ is a diagonal matrix and assembled as:

$$\boldsymbol{\alpha}(t) = \begin{bmatrix} \alpha_1(t) & 0 & 0 & 0 & \cdots & 0 & 0 \\ 0 & \alpha_1(t) & 0 & 0 & \cdots & 0 & 0 \\ 0 & 0 & \alpha_2(t) & 0 & \cdots & 0 & 0 \\ 0 & 0 & 0 & \alpha_2(t) & \cdots & 0 & 0 \\ \vdots & \vdots & \vdots & \vdots & \ddots & \vdots & \vdots \\ 0 & 0 & 0 & 0 & \cdots & \alpha_N(t) & 0 \\ 0 & 0 & 0 & 0 & \cdots & 0 & \alpha_N(t) \end{bmatrix} . \tag{7}$$

3 Proof for the equivalence of the nsGLE and extended dynamics

Rewrite the extended dynamics as:

$$\begin{aligned}
\dot{P}_k(t) &= -\mathbf{A}_{ps}\boldsymbol{\alpha}(t)\mathbf{S}_k(t) \\
\dot{\mathbf{S}}_k(t) &= -\boldsymbol{\alpha}(t)\mathbf{A}_{sp}P_k(t) - \mathbf{A}_{ss}\mathbf{S}_k(t) + \mathbf{B}_s\boldsymbol{\xi}(t) ,
\end{aligned} \tag{8}$$

from which we solve for $\mathbf{S}_k(t)$ as:

$$\mathbf{S}_k(t) = \int_0^t e^{-(t-t')\mathbf{A}_{ss}}[-\boldsymbol{\alpha}(t')\mathbf{A}_{sp}P_k(t') + \mathbf{B}_s\boldsymbol{\xi}(t')]dt' + e^{-t\mathbf{A}_{ss}}\mathbf{S}_k(0) . \tag{9}$$

Substituting Eq. (9) into the first equation in Eq. (8) results in:

$$\begin{aligned}
\dot{P}_k(t) = &\int_0^t \mathbf{A}_{ps}\boldsymbol{\alpha}(t)e^{-(t-t')\mathbf{A}_{ss}}\boldsymbol{\alpha}(t')\mathbf{A}_{sp}P_k(t')dt' \\
&- \int_0^t \mathbf{A}_{ps}\boldsymbol{\alpha}(t)e^{-(t-t')\mathbf{A}_{ss}}\mathbf{B}_s\boldsymbol{\xi}(t')dt' - \mathbf{A}_{ps}\boldsymbol{\alpha}(t)e^{-t\mathbf{A}_{ss}}\mathbf{S}_k(0) .
\end{aligned} \tag{10}$$

Substituting into Eq. (10) the matrix form of the memory kernel Eq. (11):

$$K(t',t) = -\mathbf{A}_{ps}\boldsymbol{\alpha}(t)e^{-(t-t')\mathbf{A}_{ss}}\boldsymbol{\alpha}(t')\mathbf{A}_{sp} \ , \tag{11}$$

and the fluctuating force Eq. (12):

$$\tilde{F}_k(t) = -\int_0^t \mathbf{A}_{ps}\boldsymbol{\alpha}(t)e^{-(t-t')\mathbf{A}_{ss}}\mathbf{B}_s\boldsymbol{\xi}(t')dt' - \mathbf{A}_{ps}\boldsymbol{\alpha}(t)e^{-t\mathbf{A}_{ss}}\mathbf{S}_k(0) \ , \tag{12}$$

we obtain:

$$\dot{P}_k(t) = -\int_0^t K(t',t)P_k(t')dt' + \tilde{F}_k(t) \ , \tag{13}$$

which is just the nsGLE for each dimension of the CG variable $\mathbf{P}(t)$.

We next prove that such defined memory kernel and fluctuating force specified as Eq. (12) satisfy:

$$\langle \tilde{\mathbf{F}}(t') \cdot \tilde{\mathbf{F}}(t) \rangle = \langle |\mathbf{P}(t')|^2 \rangle K(t',t) \ , \tag{14}$$

the fluctuation-dissipation relation that holds for non-stationary processes. To proceed, the auto-correlation of the fluctuating force can be derived as:

$$\begin{aligned} \langle \tilde{F}_k(t')\tilde{F}_k(t) \rangle =& \langle \int_0^{t'} \mathbf{A}_{ps}\boldsymbol{\alpha}(t')e^{-(t'-t'')\mathbf{A}_{ss}}\mathbf{B}_s\boldsymbol{\xi}(t'')dt'' \int_0^t \boldsymbol{\xi}^T(t''')\mathbf{B}_s^T e^{-(t-t''')\mathbf{A}_{ss}^T}\boldsymbol{\alpha}^T(t)\mathbf{A}_{ps}^T dt''' \rangle \\ &+ \langle \mathbf{A}_{ps}\boldsymbol{\alpha}(t')e^{-t'\mathbf{A}_{ss}}\mathbf{S}_k(0)\mathbf{S}_k^T(0)e^{-t\mathbf{A}_{ss}^T}\boldsymbol{\alpha}^T(t)\mathbf{A}_{ps}^T \rangle \\ =& \int_{-\infty}^{t'} \int_{-\infty}^{t} \mathbf{A}_{ps}\boldsymbol{\alpha}(t')e^{-(t'-t'')\mathbf{A}_{ss}}\mathbf{B}_s \langle \boldsymbol{\xi}(t'')\boldsymbol{\xi}^T(t''') \rangle \mathbf{B}_s^T e^{-(t-t''')\mathbf{A}_{ss}^T}\boldsymbol{\alpha}^T(t)\mathbf{A}_{ps}^T dt'' dt''' \\ &+ \mathbf{A}_{ps}\boldsymbol{\alpha}(t')e^{-t'\mathbf{A}_{ss}} \langle \mathbf{S}_k(0)\mathbf{S}_k^T(0) \rangle e^{-t\mathbf{A}_{ss}^T}\boldsymbol{\alpha}(t)\mathbf{A}_{ps}^T \ . \end{aligned} \tag{15}$$

Since $\boldsymbol{\xi}(t) \in \mathbb{R}^{2N\times 1}$ satisfies $\langle \xi(t) \rangle = 0$ and

$$\langle \xi_i(t')\xi_j(t) \rangle = \begin{cases} 1 & i=j \text{ and } t'=t \\ 0 & \text{otherwise} \end{cases} \ , \tag{16}$$

where $\xi_i(t)$ and $\xi_j(t)$ denote the different elements of $\boldsymbol{\xi}(t)$, Eq. (15) can be written as:

$$\begin{aligned} \langle \tilde{F}_k(t')\tilde{F}_k(t) \rangle = & \int_0^{t'} \mathbf{A}_{ps}\boldsymbol{\alpha}(t')e^{-(t'-t'')\mathbf{A}_{ss}}\mathbf{B}_s\mathbf{B}_s^T e^{-(t-t'')\mathbf{A}_{ss}^T}\boldsymbol{\alpha}^T(t)\mathbf{A}_{ps}^T dt'' \\ &+ \mathbf{A}_{ps}\boldsymbol{\alpha}(t')e^{-t'\mathbf{A}_{ss}} \langle \mathbf{S}_k(0)\mathbf{S}_k^T(0) \rangle e^{-t\mathbf{A}_{ss}^T}\boldsymbol{\alpha}(t)\mathbf{A}_{ps}^T \ . \end{aligned} \tag{17}$$

By substituting $\langle \mathbf{S}_k(0)\mathbf{S}_k^T(0) \rangle = \frac{\langle |\mathbf{P}(t')|^2 \rangle}{d}\mathbf{I}$ and

$$\mathbf{B}_s\mathbf{B}_s^T = \frac{\langle |\mathbf{P}(t')|^2 \rangle}{d}(\mathbf{A}_{ss} + \mathbf{A}_{ss}^T)$$

into Eq. (17), we arrive:

$$
\begin{aligned}
\langle \tilde{F}_k(t')\tilde{F}_k(t)\rangle =& \int_0^{t'} \mathbf{A}_{ps}\boldsymbol{\alpha}(t')e^{-(t'-t'')\mathbf{A}_{ss}}\frac{\langle|\mathbf{P}(t')|^2\rangle}{d}(\mathbf{A}_{ss}+\mathbf{A}_{ss}^T)e^{-(t-t'')\mathbf{A}_{ss}^T}\boldsymbol{\alpha}^T(t)\mathbf{A}_{ps}^T dt'' \\
&+\frac{\langle|\mathbf{P}(t')|^2\rangle}{d}\mathbf{A}_{ps}\boldsymbol{\alpha}(t')e^{-t'\mathbf{A}_{ss}}e^{-t\mathbf{A}_{ss}^T}\boldsymbol{\alpha}(t)\mathbf{A}_{ps}^T \\
&=\frac{\langle|\mathbf{P}(t')|^2\rangle}{d}\mathbf{A}_{ps}\boldsymbol{\alpha}(t')e^{-t'\mathbf{A}_{ss}}\int_0^{t'} e^{t''\mathbf{A}_{ss}}(\mathbf{A}_{ss}+\mathbf{A}_{ss}^T)e^{t''\mathbf{A}_{ss}^T}dt''e^{-t\mathbf{A}_{ss}^T}\boldsymbol{\alpha}^T(t)\mathbf{A}_{ps}^T \\
&+\frac{\langle|\mathbf{P}(t')|^2\rangle}{d}\mathbf{A}_{ps}\boldsymbol{\alpha}(t')e^{-t'\mathbf{A}_{ss}}e^{-t\mathbf{A}_{ss}^T}\boldsymbol{\alpha}(t)\mathbf{A}_{ps}^T \\
&=\frac{\langle|\mathbf{P}(t')|^2\rangle}{d}\mathbf{A}_{ps}\boldsymbol{\alpha}(t')e^{-t'\mathbf{A}_{ss}}[e^{t''\mathbf{A}_{ss}}e^{t''\mathbf{A}_{ss}^T}]_{t''=0}^{t''=t'}e^{-t\mathbf{A}_{ss}^T}\boldsymbol{\alpha}^T(t)\mathbf{A}_{ps}^T \\
&+\frac{\langle|\mathbf{P}(t')|^2\rangle}{d}\mathbf{A}_{ps}\boldsymbol{\alpha}(t')e^{-t'\mathbf{A}_{ss}}e^{-t\mathbf{A}_{ss}^T}\boldsymbol{\alpha}(t)\mathbf{A}_{ps}^T \\
&=\frac{\langle|\mathbf{P}(t')|^2\rangle}{d}\mathbf{A}_{ps}\boldsymbol{\alpha}(t')e^{-(t-t')\mathbf{A}_{ss}^T}\boldsymbol{\alpha}^T(t)\mathbf{A}_{ps}^T \ .
\end{aligned}
\tag{18}
$$

Noting that $\langle \tilde{F}_i(t')\tilde{F}_i(t)\rangle$ is a scalar, $\boldsymbol{\alpha}(t)=\boldsymbol{\alpha}^T(t)$, and $\mathbf{A}_{ps}^T=-\mathbf{A}_{sp}$, we can further derive:

$$
\begin{aligned}
\langle \tilde{F}_k(t')\tilde{F}_k(t)\rangle =& \frac{\langle|\mathbf{P}(t')|^2\rangle}{d}(\mathbf{A}_{ps}\boldsymbol{\alpha}(t')e^{-(t-t')\mathbf{A}_{ss}^T}\boldsymbol{\alpha}^T(t)\mathbf{A}_{ps}^T)^T \\
=&\frac{\langle|\mathbf{P}(t')|^2\rangle}{d}(-\mathbf{A}_{ps}\boldsymbol{\alpha}(t)e^{-(t-t')\mathbf{A}_{ss}}\boldsymbol{\alpha}(t')\mathbf{A}_{sp}) \\
=&\frac{\langle|\mathbf{P}(t')|^2\rangle}{d}K(t',t) \ ,
\end{aligned}
\tag{19}
$$

from which we finally yield:

$$
\langle \tilde{\mathbf{F}}(t')\cdot\tilde{\mathbf{F}}(t)\rangle = \sum_{k=1}^{d}\langle \tilde{F}_k(t')\tilde{F}_k(t)\rangle = \langle|\mathbf{P}(t')|^2\rangle K(t',t) \ . \tag{20}
$$

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