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On the Environmental Modes
for the Generalized Langevin Equation

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Abstract

The generalized Langevin equation (GLE) is used widely in molecular science and time series analysis as it offers a convenient low-dimensional description for large systems. There the dynamical effect of the environment interacting with the low-dimensional system is expressed as friction and random force. The present paper aims to investigate explicit dynamical variables to describe the dynamical modes in the environment that are derived from the GLE and defined solely in terms of the time series of the observed variable. The formulation results in equations of motion without a memory term, and hence offers a more intuitive description than the GLE. The framework provided by the present study is expected to elucidate a multi-dimensional dynamics hidden behind the time series of the observed quantity.

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I. INTRODUCTION

In studying the dynamics of a system consisting of a huge number of atoms, it is often a good strategy to divide the total system into a *subsystem* and its *environment* rather than to treat all the atomic coordinates equally. The subsystem is a collection of a small number of selected variables that are central to the research interest, or are experimentally observable. The environment is defined as all the other degrees of freedom contained in the total system. While the subsystem is explicitly described with the dynamical variables, the effect of the environment is only implicitly included in the description, for example, as a frictional force exerted on the subsystem.

The generalized Langevin equation (GLE)[1–3] is one of such descriptions. There the time evolution of the subsystem is given by three terms: mean force, friction, and random force. The mean force represents the interaction within the subsystem and the average effect of the environment. It can be calculated as the force on the subsystem averaged over all the possible configurations of the environment weighted by a statistical distribution such as the Boltzmann distribution. The random force represents the kick from the environment that is determined by the environment and uncorrelated to the initial condition of the subsystem. The friction is the response of the environment to the subsystem. It depends on the history of the subsystem, thus making the GLE an integro-differential equation. By using the technique of projection operator, it has been proved[1–3] that any Hamiltonian system can be projected onto its subsystem obeying the GLE. Zwanzig[4] showed that the GLE with a linear friction term is exact when the environment is a collection of harmonic oscillators and the coupling to the subsystem is bilinear. Cortés *et al.*[5] went further to show that, when the couplings are linear in the subsystem coordinate but nonlinear in the other coordinates, the system still obeys the GLE with linear friction up to the first order in the system-environment coupling strength. The projection operator formalism has also been extended to non-Hamiltonian systems.[6]

In the field of molecular science, the Langevin-type formulation was the starting point of the traditional rate theories.[7,8] Kramers[7] derived analytical expressions for one-dimensional barrier-crossing rates by using the Fokker-Planck equation, which is equivalent to the Langevin equation, the Ohmic limit of the GLE. The study was followed by Mel'nikov and Meshkov[9] to obtain a formula that connects two limiting behaviors treated by Kramers.

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Langer[10] gave the multi-dimensional version of the rate formula. Extension to systems having retarded response of the environment, described by the GLE, was given by Grote and Hynes.[8] The useful formula derived by them was used in the analyses of reaction rates in molecular systems.[11–13] The dynamics of the barrier crossing was further studied in terms of the phase space structure in the neighborhood of a saddle point in the energy landscape,[14–24] which extends the nonlinear dynamical transition state theory developed for a long time.[25–32] By using the GLE, it was proved that, even in the existence of thermal noise, there exists a clear structure in the phase space that determines the occurrence of the chemical reaction.

In the application to specific molecular systems, it is needed to obtain the concrete functional forms of the potential and the friction term appearing in the GLE. Studies have been performed to obtain these functions from *ab initio* calculations or MD simulations[11–13,33–38] in molecular systems with many degrees of freedom. Rather than calculating them from the first principle by the projection operator formulation, the friction kernel is often calculated from the velocity autocorrelation function obtained by MD simulations.[39] Methods for direct calculation of the projected correlation, however, were also recently developed.[38] In addition to the molecular science, the GLE formulation has found its application also in the time series analysis of meteorological and financial data,[40] although its advantage over other models is under some criticism,[41] to which point we will come back later in the present paper. Another application of the GLE formulation was found in the problem of model reduction.[42] Starting with a rather complicated model for a cellular signaling process, the time series obtained by simulations with the full model was put into the GLE-based time series analysis. The reconstructed GLE provided a reduced one-dimensional model that has the same prediction ability with the original full model.

The success of the theoretical works using the GLE stems from the facts that the GLE is simple enough with only a small number of variables and that it is physically sound due to the exact derivation from the first principle using the projection operator. However, one may find difficulty in the intuitive interpretation of the friction term in the GLE that depends on the *history* of the subsystem. Physical picture of this "memory" term may be as follows: Suppose the environment is kicked by the subsystem at a certain time t'. This kick affects the configuration of atoms in the environment. The motion of the environmental atoms after time t' reflects to some extent the effect of the kick from the subsystem at time t'. This effect remains for some time after t'. Then, at time $t(> t')$, the subsystem feels a force from the

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environment that depends on the configuration of the environmental atoms at time t, the latter, in turn, depending on the kick by the subsystem in the past (at time t'). When the motion of the environment is not described with explicit variables (as is done by the GLE), this effect appears as if the subsystem were interacting with its past. Since the subsystem and the environment are continuously interacting with each other, the friction term is given by an integration of this effect over the range $0 < t' < t$ where 0 is the time at which we prepare the initial condition.

In fact, there have been several studies[15,18,24,43–50] to express this retarded effect of the environment *by explicit variables.* Dynamical variables are introduced to describe the motion that exists in the environment and gives rise to the memory term in the GLE. This concept dates back to the continued-fraction expansion of the friction kernel introduced by Mori.[51] Grigolini formulated a matrix-form equation of motion for the variables representing the dynamical modes in the environment.[43,45] It is later utilized to perform numerical simulations with GLE and to develop rate theories in the case of nonwhite noise.[45,52] Adelman[46,47] developed similar expansion in his molecular time scale GLE (MTGLE) and introduced harmonic chain representation that is equivalent to the GLE. Martens[49] developed another approach by decomposing the friction term. The phase space structure in the barrier crossing dynamics was discussed[15,18,24,50] following the latter approach. As pointed out by Bartsch[18], the dimensionality of the phase space in this approach remains to be two because the initial values of the environmental variables must be zero. The older approach[43,45–47,51] involves both the friction and the random force to define the environmental modes and the phase space dimensionality is the same with the number of variables, which appears to be intuitively more natural.

Contrary to describing all the atoms in the environment with explicit coordinates, the environmental modes in these works are introduced from the GLE and describe only those modes that are effectively coupled with the subsystem. The number of variables is kept much smaller than the number of all atoms in the total system. This description therefore keeps the simplicity offered by the GLE formulation while providing more intuitive physical picture because of the absence of memory terms. In addition to being a useful mathematical tool to cast the GLE into a more tractable form for numerical simulations and theoretical investigation of the phase space structure, it is also a physically interesting concept to represent in a simple way the collective motion of the environmental atoms, which otherwise

requires involved description with a huge number of coordinates.

Another important point noticed in Refs. 44,45 is that the description with such effective variables can cover more general situations than the original GLE. By using explicit variables for the environment, one can set values far from equilibrium for these variables enabling the simulation of "excited thermal bath," whereas the implicitly described thermal bath in the GLE is assumed to be in the thermodynamical equilibrium.

In the present paper, it is aimed to provide a formulation of such effectively expressed environmental modes (hereafter called EXEM) based on the multi-exponential form of the friction kernel. The assumption that the friction kernel can be sufficiently approximated by a multi-exponential form is proved to be equivalent to the assumption made in the previous works. In the recent approach of obtaining the friction kernel numerically from the MD simulations or observed time series,[11–13,33–38,40,41] the friction kernel can be obtained first numerically and then fitted to analytical forms. For example, the MD simulations[12,13] found the friction kernel fitted with sufficient accuracy to the sum of a small number of exponential and exponentially decaying trigonometric functions (note that the trigonometric function can also be expressed as an exponential function by using complex numbers). Thus the present paper takes the fitting of the friction kernel as the starting point.

By using the analytical multi-exponential form of the friction kernel as our starting point, it turns out that we can formulate the EXEM without explicitly evaluating the higher-order derivatives of the friction kernel, which were needed in the previous continued-fraction approaches.[43,45–47,51] In the cases where the friction kernel is given only numerically from the simulation or experimental data, this is of practical significance since the numerical evaluation of higher-order derivatives is often unstable. Moreover, by expressing our assumption in the form of multi-exponential friction kernel, the validity of the assumption can be evaluated readily as the residual error in the least-squares fitting of the friction kernel to the multi-exponential function. In the continued-fraction approaches, it was not directly clear at which point to truncate the expansion in order to obtain sufficient approximation. Sec. II of the present paper thus presents the formulation of EXEM by starting with the multi-exponential friction kernel, and proofs for the advantages mentioned above are concretely given. The definition of the EXEM in the present formulation is given completely in terms of the subsystem variable. It is therefore possible to calculate the values of those EXEM variables from the observed time series of the subsystem variable without requiring

any knowledge about the configuration of the environmental atoms. This is another great advantage in practical applications to time series analysis where we have only the time series of the observable quantity in hand. The theoretical formulation is followed by some specific examples in Sec. III in order to provide an idea of how the calculation with the present formulation proceeds. In particular, it is proved that, for some model cases including the Zwanzig Hamiltonian, the EXEM introduced in the present paper reproduces exactly the dynamical variables of the original total system. Concluding remarks and future outlook are given in Sec. IV.

II. THEORY

Let Q be the dynamical variable of the subsystem. To keep the formulation simple, the subsystem is considered to be one-dimensional. Extension to the case of multi-dimensional subsystems is straightforward. The GLE is given as

$$\ddot{Q} = f_{\mathrm{M}}(Q) - \int_0^t \gamma(t - t')\dot{Q}(t')\mathrm{d}t' + \xi(t), \tag{1}$$

where derivation with time t is expressed with the dot over the symbol. The first term $f_{\mathrm{M}}(Q)$ is the mean force acting on Q. The second term expresses the frictional force that depends on the velocity $\dot{Q}$ in the past. The third term $\xi(t)$ is the random force whose statistical distribution is uncorrelated to the initial condition of Q:

$$\langle Q(0)\xi(t)\rangle = 0, \qquad\qquad (\text{for } t > 0), \tag{2}$$

where the bracket denotes statistical average. The random force satisfies the fluctuation-dissipation theorem:

$$\langle \xi(t)\xi(t')\rangle = \langle \dot{Q}^2\rangle\gamma(t - t'). \tag{3}$$

As stated in Sec. I, the present theory assumes that the friction kernel γ is fitted with the multi-exponential form:

$$\gamma(\tau) \approx \sum_m c_m \exp\left((-\lambda_m + \mathrm{i}\omega_m)\tau\right)$$

$$= \sum_m c_m \exp\left(\mathrm{i}\alpha_m\tau\right), \tag{4}$$

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where c_m, λ_m, and ω_m are fitting parameters. The parameters λ_m and ω_m are real numbers. The former is responsible to the decay of $\gamma(\tau)$, while the latter allows oscillatory form of $\gamma(\tau)$. In the second line of Eq. (4), the two real parameters λ_m and ω_m are put together into one complex number α_m by

$$\alpha_m = \omega_m + \mathrm{i}\lambda_m. \tag{5}$$

In the case ω_m is not zero, the coefficients c_m should also be complex numbers and the summation over m in Eq. (4) should involve summation of complex conjugate pairs in order for $\gamma(\tau)$ to be real.

Note that the random force $\xi(t)$ is defined only for $t > 0$ since the GLE (1) describes the time evolution after the initial time $t = 0$ (note also Eq. (2)). The friction kernel $\gamma(\tau)$ is also defined only for $\tau > 0$ since the integration in Eq. (1) is performed only for the range $0 < t - t' < t$. However, one can extend the definition of γ by using the fluctuation-dissipation theorem Eq. (3). Namely, it must be an even function because the left hand side of Eq. (3) is symmetric with respect to the exchange of t and t'. Eq. (4) should accordingly be modified to

$$\gamma(\tau) = \gamma(-\tau) = \sum_m c_m \exp\left(\mathrm{i}\alpha_m|\tau|\right). \tag{6}$$

In what follows, the time non-local friction term, with the assumed multi-exponential kernel, is first decomposed into a set of additional variables in Sec. II A. This decomposition is basically equivalent to the previous approach in Refs. 15,18,24,49,50. As has been pointed out,[18] the dynamical variables thus introduced explore only a low-dimensional submanifold in the phase space. This is because the initial condition of these variables is restricted to be zero. This fact causes another problem concerning their physical relevance as dynamical variables. Since the initial time ($t = 0$) is the time at which we start the observation and does not carry great physical meaning in the equilibrium system, the peculiar restriction of the variables at the initial time implies that they may not be defined soundly on physical bases. In Secs. II B–II D, these two problems are solved by decomposing also the random force term and putting those variables together to define new additional variables. Each of the newly defined variables is a sum of a term coming from the friction term and one coming from the random force. Although these two terms depend on the time reference, the dependence cancel when their sum is taken, making the newly defined variables independent

of the time reference. The initial condition of these variables has therefore no restriction and the system explores the full dimensionality of the phase space. Sec. II B formulates the decomposition of the random force and the definition of the environmental variables. Sec. II C derives the equations of motion obeyed by the environmental variables, which turn out to be multi-dimensional and time-local in contrast to the GLE. Sec. II D then discusses and proves the above-mentioned advantages achieved by the decomposition of the random force.

A.   Decomposition of the friction term

Martens defined the environmental modes by decomposing the friction term. Formulation[50] presented here is slightly different from but equivalent to the original one[49] for non-degenerate cases. The equivalence is proved in Appendix A. Let

$$Z_m(t) = \int_0^t c_m \exp\left(i\alpha_m(t - t')\right) \dot{Q}(t') dt'. \tag{7}$$

Due to Eq. (4), the memory term of the GLE (1) can be replaced by the sum of Z_m:

$$\ddot{Q} = f_{\mathrm{M}}(Q) - \sum_m Z_m + \xi(t), \tag{8}$$

while the time evolution of Z_m derived from Eq. (7) is

$$\dot{Z}_m = i\alpha_m Z_m + c_m \dot{Q}. \tag{9}$$

Eqs. (8) and (9) give multi-dimensional equations of motion that are without a memory term but equivalent to the original GLE. The variables Z_m can thus be thought of as describing the motion existing in the environment. More precisely, these variables describe the part of the environment that are moving in response to the kick from the subsystem. While this treatment renders the intuitively difficult memory term into more explicit form of equations of motion, the random force $\xi(t)$ in Eq. (8) is still the same with that in the GLE, which is a colored noise satisfying Eq. (3). In the next subsection, we proceed to decompose the random force.

B. Decomposition of the random force

Let $\tilde{\gamma}(\omega)$ be the Fourier transform of $\gamma(\tau)$. From Eq. (6), elementary calculation gives

$$
\begin{aligned}
\tilde{\gamma}(\omega) &= \int_{-\infty}^{+\infty} \gamma(\tau) \exp\left(-i\omega\tau\right) d\tau \\
&= \sum_m \left\{ \frac{-ic_m}{\omega - \alpha_m} + \frac{ic_m^*}{\omega - \alpha_m^*} \right\},
\end{aligned}
\tag{10}
$$

where the star denotes complex conjugate.

Since $\gamma(\tau)$ is a real and even function of τ, its Fourier transform must also be real and even for real ω. This requires that the sigular points α_m of $\tilde{\gamma}$ appear symmetrically with respect to both the real and the imaginary axes in the complex ω-plane (see Fig. 1). This can be easily seen from Eqs. (6) and (10): If $\alpha_m = \omega_m + i\lambda_m$, there must be $\alpha_{\overline{m}} = -\omega_m + i\lambda_m$ for some $\overline{m}$ in order for $\gamma(\tau)$ in Eq. (6) to be real. Then, the right hand side of Eq. (10) tells that $\alpha_m^* = \omega_m - i\lambda_m$ and $\alpha_{\overline{m}}^* = -\omega_m - i\lambda_m$ are also singular points of $\tilde{\gamma}(\omega)$. Thus any singular point of $\tilde{\gamma}(\omega)$ appears as a quadruple $\pm\omega_m \pm i\lambda_m$ for $\omega_m \neq 0$, or a pair $\pm i\lambda_m$ for $\omega_m = 0$.
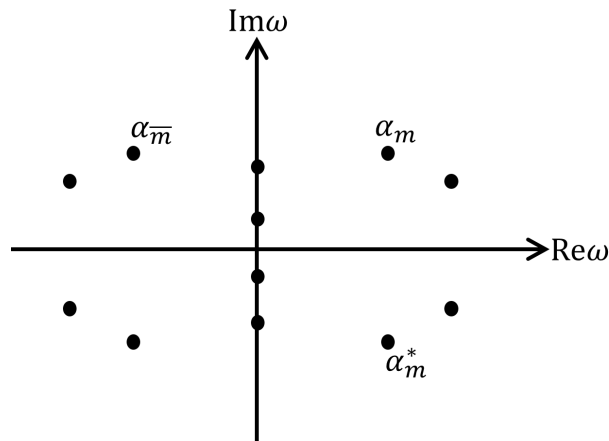
FIG. 1: Schematic picture for the distribution of the singular points of $\tilde{\gamma}(\omega)$ in the complex ω-plane.

Putting the terms in the right hand side of Eq. (10) into a common denominator and factorizing the numerator gives the following form:

$$
\tilde{\gamma}(\omega) = \frac{A \prod_n (\omega - \beta_n)(\omega - \beta_n^*)}{\prod_m (\omega - \alpha_m)(\omega - \alpha_m^*)}.
\tag{11}
$$

In order for $\tilde{\gamma}(\omega)$ to be real and even, the zeros of the numerator (β_n and β_n^*) must also appear symmetrically in a similar manner with Fig. 1. Let β_n and β_n^* in Eq. (11) be defined in such a way that $\text{Im}\beta_n > 0$ and $\text{Im}\beta_n^* < 0$.

The prefactor A in Eq. (11) must be a positive real number because $\tilde{\gamma}(\omega)$ is nonnegative for real ω.[47] The original proof reviewed in Ref. 47 citing Kubo's works[53,54] is based on the physical picture of dissipation. For the sake of completeness, a proof purely based on equations is given in Appendix B.

Now let us define a function $\tilde{g}$ with coefficients $\{b_m\}$ by

$$
\begin{aligned}
\tilde{g}(\omega) &= \sqrt{\frac{A}{2}} \frac{\prod_n \mathrm{i}(\omega - \beta_n^*)}{\prod_m \mathrm{i}(\omega - \alpha_m)} \\
&= \sum_m \frac{-\mathrm{i}b_m}{\omega - \alpha_m},
\end{aligned}
\tag{12}
$$

and its correspondent in the time domain

$$
\begin{aligned}
g(\tau) &= \frac{1}{2\pi} \int_{-\infty}^{+\infty} \tilde{g}(\omega) \exp(\mathrm{i}\omega\tau)\, \mathrm{d}\omega \\
&= \begin{cases} 0 & (\tau < 0) \\ \sum_m b_m \exp(\mathrm{i}\alpha_m \tau) & (\tau > 0) \end{cases}.
\end{aligned}
\tag{13}
$$

Note that, for real values of ω,

$$
\tilde{g}(-\omega) = \tilde{g}(\omega)^*
\tag{14}
$$

$$
\tilde{\gamma}(\omega) = 2\tilde{g}(\omega)\tilde{g}(-\omega).
\tag{15}
$$

The first equality is a consequence of the fact that $\{\alpha_m\}$ and $\{\beta_n^*\}$ are distributed symmetrically in the right and left half-planes (Fig. 1). The second equality can be easily seen from Eqs. (11) and (12).

We further define

$$
\tilde{h}(\omega) = \frac{1}{\tilde{g}(\omega)} = \sqrt{\frac{2}{A}} \frac{\prod_m \mathrm{i}(w - \alpha_m)}{\prod_n \mathrm{i}(w - \beta_n^*)},
\tag{16}
$$

$$
\tilde{k}_m(\omega) = \frac{-\mathrm{i}b_m}{\omega - \alpha_m} \tilde{h}(\omega),
\tag{17}
$$

and their correspondents in the time domain by the inverse Fourier transform:

$$
h(\tau) = \frac{1}{2\pi} \int_{-\infty}^{+\infty} \tilde{h}(\omega) \exp(\mathrm{i}\omega\tau)\, \mathrm{d}\omega.
\tag{18}
$$

$$
k_m(\tau) = \frac{1}{2\pi} \int_{-\infty}^{+\infty} \tilde{k}_m(\omega) \exp(\mathrm{i}\omega\tau)\, \mathrm{d}\omega.
\tag{19}
$$

Note that $\tilde{h}(-\omega) = \tilde{h}(\omega)^*$ due to Eqs. (14) and (16). Translated into the time domain, this means that $h(\tau)$ is a real-valued function. The functions k_m satisfy

$$\sum_m \tilde{k}_m(\omega) = 1, \tag{20}$$

because of Eqs. (12), (16), and (17). In the time domain, this corresponds to

$$\sum_m k_m(\tau) = \delta(\tau), \tag{21}$$

where δ is the Dirac delta function. We also note that the relation $(i\omega - i\alpha_m)\tilde{k}_m(\omega) = b_m\tilde{h}(\omega)$ obtained from Eq. (17) translates into the time domain as

$$\left(\frac{\mathrm{d}}{\mathrm{d}\tau} - i\alpha_m\right) k_m(\tau) = b_m h(\tau). \tag{22}$$

We are now ready to decompose the random force $\xi(t)$ by the following convolutions:

$$\eta(t) = \int_0^{+\infty} h(t - t')\xi(t')\mathrm{d}t', \tag{23}$$

$$\xi_m(t) = \int_0^{+\infty} k_m(t - t')\xi(t')\mathrm{d}t', \tag{24}$$

where we note the integration range is $t' > 0$ because the random force $\xi(t)$ is only defined for $t > 0$. Because of Eq. (21) we have

$$\sum_m \xi_m(t) = \xi(t), \tag{25}$$

and because of Eq. (22)

$$\frac{\mathrm{d}}{\mathrm{d}t}\xi_m(t) = i\alpha_m\xi_m(t) + b_m\eta(t). \tag{26}$$

C.   Effective equation of motion with environmental modes

Substituting Eq. (25) into Eq. (8) gives

$$\ddot{Q} = f_\mathrm{M}(Q) + \sum_m (-Z_m + \xi_m), \tag{27}$$

We can put Eqs. (9) and (26) together by defining

$$X_m = -Z_m + \xi_m. \tag{28}$$

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Putting together Eqs. (9), (26), and (27), we obtain the following equations of motion

$$\ddot{Q} = f_{\mathrm{M}}(Q) + \sum_m X_m,$$

$$\dot{X}_m = \mathrm{i}\alpha_m X_m - c_m \dot{Q} + b_m \eta(t), \qquad (29)$$

with the statistical property of the new random force η being that of white noise:

$$\langle \eta(t_1)\eta(t_2) \rangle = 2\langle \dot{Q}^2 \rangle \delta(t_1 - t_2), \qquad (30)$$

which is proved in Appendix C. The equation of motion (29) and Eq. (30) are the central result of this section. As mentioned in Sec. I, the variables X_m thus defined are considered to effectively represent the collective motion of the environmental atoms, which otherwise requires much involved description with a huge number of coordinates. Hereafter the variables X_m are called EXEM for effectively expressed environmental modes.

In the supplemental material,[55] the relationship between the environmental coordinates in the previous studies[43,45–47,51] and the variables X_m in the present work is established. It turns out that they are related in the form of linear transformations. The EXEMs in the present work correspond more directly to the stochastic normal modes considered in Refs. 45,46. The contribution of the present work firstly lies in the fact that it starts with the assumption of the multi-exponential form of the friction kernel. By postulating the explicit functional form of the friction kernel at the starting point, the present formulation avoids the evaluation of the high order derivatives of the friction kernel, which was needed in the previous continued-fraction approach. This is of practical significance in the case of friction kernels obtained numerically from simulations or data analyses, because the evaluation of high order derivatives is numerically unstable. Note also that the truncation of the continued fraction made in the previous studies is equivalent with the assumption of the multi-exponential form of the friction kernel, because it is proved in the supplemental material[55] that, if the truncation of the continued fraction at a certain order gives accurate description of the system, the friction kernel must be a multi-exponential function. A question of practical importance in the continued-fraction approach is when to truncate the continued-fraction expansion and how to judge if the approximation is accurate enough or not. In the present formulation, the question has become that of least-squares fitting. The quality of the fitting to $\gamma(\tau)$ with the multi-exponential form can be assessed with standard

statistical properties such as residual errors, coefficient of determination, information criteria, and so forth.[56] Finally, another contribution in the present work is the explicit forms (Eqs. (7), (24), and (28)) to give the environmental modes X_m in terms of the time series of $Q(t)$. This is of significant importance in the context of time series analysis where we start with only observed time series $Q(t)$ in hand.

Given γ in practice, or equivalently given $\{c_m\}$ and $\{\alpha_m\}$ in Eq. (4), one can calculate $\{b_m\}$ by elementary calculation of rational functions through Eqs. (10), (11), and (12). Alternatively, one can obtain $\{b_m\}$ by numerically solving the following equation:

$$c_m = 2\mathrm{i}b_m \sum_n \frac{b_n}{\alpha_m + \alpha_n}, \tag{31}$$

which can be proved as follows: Substituting Eq. (12) into Eq. (15) gives

$$
\begin{aligned}
\tilde{\gamma}(\omega) &= 2 \sum_m \frac{-\mathrm{i}b_m}{\omega - \alpha_m} \sum_n \frac{-\mathrm{i}b_n}{-\omega - \alpha_n} \\
&= 2 \sum_{m,n} \frac{b_m b_n}{\alpha_n + \alpha_m} \left(\frac{1}{\omega - \alpha_m} - \frac{1}{\omega + \alpha_n} \right).
\end{aligned}
\tag{32}
$$

Comparing the coefficient of $1/(\omega - \alpha_m)$ with Eq. (10), we obtain Eq. (31).

D. Initial conditions for EXEMs and the phase space dimensionality

As noted above (see discussion after Eq. (5)), for each $\alpha_m = \omega_m + \mathrm{i}\lambda_m$ whose real component ω_m is not zero, there must exist some $\overline{m}$ such that $\alpha_{\overline{m}} = -\omega_m + \mathrm{i}\lambda_m$, that is,

$$\alpha_{\overline{m}} = -\alpha_m^*. \tag{33}$$

For such $\overline{m}$, we can prove, by Eqs. (4), (12), and (14),

$$c_{\overline{m}} = c_m^*, \tag{34}$$

$$b_{\overline{m}} = b_m^*, \tag{35}$$

and, for real values of ω,

$$\tilde{k}_{\overline{m}}(\omega) = \tilde{k}_m(-\omega)^*. \tag{36}$$

It follows from Eqs. (7), (24), (28), (33), (35), and (36) that

$$k_{\overline{m}}(\tau) = k_m(\tau)^*, \tag{37}$$

$$\xi_{\overline{m}}(t) = \xi_m(t)^*, \tag{38}$$

$$X_{\overline{m}}(t) = X_m(t)^*. \tag{39}$$

In the supplemental material,[55] it is proved that the initial values of the decomposed random force satisfy

$$\langle \xi_m(0)\xi_n(0)\rangle = 2\langle \dot{Q}^2\rangle \frac{ib_m b_n}{\alpha_m + \alpha_n}. \tag{40}$$

On the other hand, it follows immediately from Eq. (7) that

$$Z_m(0) = 0. \tag{41}$$

The initial condition for X_m is therefore given by the same expression as Eq. (40)

$$\langle X_m(0)X_n(0)\rangle = 2\langle \dot{Q}^2\rangle \frac{ib_m b_n}{\alpha_m + \alpha_n}. \tag{42}$$

When one is to perform a numerical simulation obeying the GLE (1), the initial conditions for $\{X_m\}$ can be generated by random numbers whose variance and covariance satisfy Eq. (42). Note also Eq. (39), that is, $X_{\overline{m}}(0)$ must be complex conjugate to $X_m(0)$, where $\overline{m}$ is such that $\alpha_{\overline{m}} = -\alpha_m^*$.

Some previous studies[15,18,24,49,50] used $\{Z_m\}$ (or its linear transform) as environmental dynamical variables. In this case, the constraint $Z_m(0) = 0$ limits the degrees of freedom for the initial condition of the dynamical variables to two ($Q(0)$ and $\dot{Q}(0)$), which is less than the total number of dynamical variables. As a consequence, the phase space region accessed by the trajectories is limited to a two-dimensional subspace. Bartsch[18] has pertinently noticed this point and formulated the explicit construction of the two-dimensional phase space in the framework using $\{Z_m\}$. On the other hand, the initial condition for $\{X_m\}$ in the present theory is carried by $\xi_m(0)$ which is not constrained to zero but is distributed according to Eq. (42) in the phase space whose dimensionality is equal to the number of the dynamical variables (Q, $\dot{Q}$, and X_m's).

Additionally, it is proved in the supplemental material[55] that the values of X_m's are independent of the choice of $t = 0$. Note the time $t = 0$ appearing in the GLE (1) is the time at which the initial condition is prepared, or, in the context of time series analysis, the

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time at which one starts the observation. The definition of Z_m (Eq. (7)) explicitly includes the time $t = 0$ and therefore $Z_m(t)$ depends on the choise of when we start the observation (in particular, $Z_m(0) = 0$). As is shown in the supplemental material,[55] this dependence cancels with that of ξ_m, making the total X_m (Eq. (28)) unchanged when we change the starting time. This is another support for the physical soundness of the present EXEM since the physics should not change with our choice of the zero of the time for equilibrium systems.

III. SPECIFIC EXAMPLES

A. Double-exponential friction kernel

The first part of this section deals with a simple friction kernel with the aim of obtaining an idea of how the calculation of the present theory proceeds. Let us suppose a double-exponential kernel

$$g(\tau) = \exp(-\tau) + \exp(-4\tau). \tag{43}$$

Its Fourier transform is

$$
\begin{aligned}
\tilde{\gamma}(\omega) &= \int_{-\infty}^{+\infty} g(|\tau|) \exp(-\mathrm{i}\omega\tau)\mathrm{d}\tau \\
&= -\frac{\mathrm{i}}{\omega - \mathrm{i}} - \frac{\mathrm{i}}{\omega - 4\mathrm{i}} + \frac{\mathrm{i}}{\omega + \mathrm{i}} + \frac{\mathrm{i}}{\omega + 4\mathrm{i}} \\
&= \frac{10\omega^2 + 40}{(\omega - \mathrm{i})(\omega - 4\mathrm{i})(\omega + \mathrm{i})(\omega + 4\mathrm{i})} \\
&= 10\frac{(\omega + 2\mathrm{i})(\omega - 2\mathrm{i})}{(\omega - \mathrm{i})(\omega - 4\mathrm{i})(\omega + \mathrm{i})(\omega + 4\mathrm{i})}.
\end{aligned}
\tag{44}
$$

Comparison with Eqs. (11), (12), (16), and (17) implies $A = 10$ and

$$\tilde{g}(\omega) = \sqrt{5}\frac{(\omega + 2\mathrm{i})}{\mathrm{i}(\omega - \mathrm{i})(\omega - 4\mathrm{i})} = \frac{\sqrt{5}\mathrm{i}}{\omega - \mathrm{i}} - \frac{2\sqrt{5}\mathrm{i}}{\omega - 4\mathrm{i}}, \tag{45}$$

$$\tilde{h}(\omega) = \frac{\mathrm{i}(\omega - \mathrm{i})(\omega - 4\mathrm{i})}{\sqrt{5}(\omega + 2\mathrm{i})}, \tag{46}$$

$$\tilde{k}_1(\omega) = \frac{\sqrt{5}\mathrm{i}}{\omega - \mathrm{i}}\tilde{h}(\omega) = -\frac{\omega - 4\mathrm{i}}{\omega + 2\mathrm{i}} = -1 + \frac{6\mathrm{i}}{\omega + 2\mathrm{i}}, \tag{47}$$

$$\tilde{k}_2(\omega) = \frac{-2\sqrt{5}\mathrm{i}}{\omega - 4\mathrm{i}}\tilde{h}(\omega) = \frac{2(\omega - \mathrm{i})}{\omega + 2\mathrm{i}} = 2 - \frac{6\mathrm{i}}{\omega + 2\mathrm{i}}. \tag{48}$$

Inverse Fourier transformation gives

$$k_1(\tau) = -\,\delta(\tau) + 6\Theta(-\tau)\exp(2\tau), \tag{49}$$

$$k_2(\tau) = 2\delta(\tau) - 6\Theta(-\tau)\exp(2\tau), \tag{50}$$

where δ is the Dirac delta function and Θ denotes the Heaviside step function:

$$\Theta(\tau) = \begin{cases} 1 & (\tau > 0) \\ 0 & (\tau < 0) \end{cases}. \tag{51}$$

Then the definition of ξ_m (Eq. (24)) reads

$$\xi_1(t) = -\,\xi(t) + \int_t^{+\infty} 6\exp\left(2(t-t')\right)\xi(t')\mathrm{d}t',$$

$$\xi_2(t) = 2\xi(t) - \int_t^{+\infty} 6\exp\left(2(t-t')\right)\xi(t')\mathrm{d}t', \tag{52}$$

which gives the expression of ξ_m (and therefore X_m) in terms of the observed time series $Q(t)$, since $\xi(t)$ can be obtained from Eq. (1) once we obtain a time series $Q(t)$ of the observable. From Eq. (52) one can easily verify $\xi_1(t) + \xi_2(t) = \xi(t)$ corresponding to Eq. (25).

The integration in the definition of ξ_m (Eq. (24)) has ambiguity at $t = 0$ due to the delta function in k_m. To avoid the ambiguity, the definition of ξ_m must be understood as the following limit:

$$\begin{aligned} \xi_m(t) &= \int_{-0}^{+\infty} k_m(t-t')\xi(t')\mathrm{d}t' \\ &= \lim_{\epsilon \to +0} \int_{-\epsilon}^{+\infty} k_m(t-t')\xi(t')\mathrm{d}t', \end{aligned} \tag{53}$$

which makes Eq. (52) valid including $t = 0$. This argument applies throughout the present paper and supplemental material[55] whenever the integration over $0 \le t < +\infty$ appears.

B. Harmonic Oscillator

Let us next consider a one-dimensional harmonic oscillator given by the following Hamiltonian:

$$H(q, p) = \frac{1}{2}p^2 + \frac{1}{2}q^2. \tag{54}$$

In the context of time series analysis, this example was once used to criticize the use of the GLE.[41]

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The time evolution of the coordinates is given by

$$q(t) = q(0) \cos t + p(0) \sin t,$$
$$p(t) = p(0) \cos t - q(0) \sin t. \tag{55}$$

When one projects the system onto q, the following GLE is obtained:

$$\dot{q} = -\int_0^t q(t') \mathrm{d}t' + \xi(t). \tag{56}$$

The friction kernel in this case is a constant function $\gamma(\tau) \equiv 1$ and does not exactly fall into the multi-exponential form postulated in this paper. However, introducing a small damping factor in the Fourier transform may help:

$$\tilde{\gamma}(\omega) = \lim_{\epsilon \to +0} \int_{-\infty}^{+\infty} \exp(-\epsilon|\tau|) \gamma(\tau) \exp(-i\omega\tau) \mathrm{d}\tau$$
$$= \lim_{\epsilon \to +0} \frac{2\epsilon}{(\omega + i\epsilon)(\omega - i\epsilon)}, \tag{57}$$

which, before taking the limit, falls into the form of Eq. (10). Following the calculations in Sec. II (the example calculation in Sec. III A may also help), we obtain

$$\tilde{g}(\omega) = \frac{-i\sqrt{\epsilon}}{\omega - i\epsilon},$$
$$\tilde{h}(\omega) = \frac{i(\omega - i\epsilon)}{\sqrt{\epsilon}},$$
$$\tilde{k}_1(\omega) = 1. \tag{58}$$

(There is only one EXEM.) The equation of motion becomes, from Eq. (29),

$$\dot{q} = X_1,$$
$$\dot{X}_1 = -\epsilon X_1 - q + \sqrt{\epsilon}\eta(t).$$
$$\tag{59}$$

Finally, taking the limit $\epsilon \to +0$, we obtain

$$\dot{q} = X_1,$$
$$\dot{X}_1 = -q, \tag{60}$$

which reproduces the original equation of motion obtained from Eq. (54) with $X_1 = p$. Thus, although the constant kernel does not have a direct correspondence to the frequency

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of the original system, the GLE combined with the present EXEM analysis reproduce the information of the original system exactly only by observing the time series of $q(t)$ (and not $p(t)$) and obtaining its GLE (56).

Next consider taking the observable $Q = q^3$. The GLE becomes[41]

$$\dot{Q} = -\int_0^t \gamma(t-t')Q(t')\mathrm{d}t' + \xi(t),$$

$$\gamma(\tau) = \frac{45}{41} + \frac{144}{205}\cos\left(\sqrt{\frac{41}{5}}\tau\right). \tag{61}$$

The frequency and the amplitudes in the friction kernel have no direct relation to the frequency of the original system. This is a significant problem when we are to obtain insights about the original system through the analysis of the observed time series using the GLE, as pointed out by Ref. 41. Let us now examine what the GLE analysis combined with the present EXEM provides. Since one cosine function is a sum of two exponentials $(\cos(\omega\tau) = (\exp(\mathrm{i}\omega\tau) + \exp(-\mathrm{i}\omega\tau))/2)$, three EXEMs appear from the friction kernel of Eq. (61), two of which are a complex conjugate pair (see Eq. (39)). The calculation proceeds similarly as above by the help of a small damping factor ϵ. As the previous case, the coefficients b_m of the white noise $\eta(t)$ disappear when the limit $\epsilon \to +0$ is taken. The other coefficients appearing in the equation of motion can be read simply from the friction kernel (see Eq. (29)). Put in the matrix from, the result is

$$\frac{\mathrm{d}}{\mathrm{d}t}\begin{pmatrix} Q \\ X_1 \\ X_2 \\ X_3 \end{pmatrix} = \begin{pmatrix} 0 & 1 & 1 & 1 \\ -45/41 & 0 & 0 & 0 \\ -72/205 & 0 & \mathrm{i}\sqrt{41/5} & 0 \\ -72/205 & 0 & 0 & -\mathrm{i}\sqrt{41/5} \end{pmatrix}\begin{pmatrix} Q \\ X_1 \\ X_2 \\ X_3 \end{pmatrix} \tag{62}$$

The last two modes, X_2 and X_3, are complex conjugate to each other. Diagonalizing the matrix in Eq. (62) gives eigenvalues $\pm\mathrm{i}$ and $\pm 3\mathrm{i}$, which reflects the frequencies of the following time evolution

$$q(t)^3 = q(0)^3 \left(3\cos(t) + \cos(3t)\right)/4$$
$$+ 3q(0)^2 p(0) \left(\sin(t) + \sin(3t)\right)/4$$
$$+ 3q(0)p(0)^2 \left(\cos(t) - \cos(3t)\right)/4$$
$$+ p(0)^2 \left(3\sin(t) - \sin(3t)\right)/4. \tag{63}$$

In conclusion, it is suggested here that the time series analysis based on the GLE formulation[40] proceed further to obtain equations of motion expressed in EXEMs presented in this work. The latter is expected to be more effective to get insights into the original system than the coefficients and frequencies appearing in the friction kernel.

C. Zwanzig Hamiltonian

Zwanzig[4] showed that the GLE with a linear friction term is exact when the environment is a collection of harmonic oscillators and the coupling to the subsystem is bilinear. The Hamiltonian for such a system is given by

$$H = \frac{1}{2}P^2 + \frac{1}{2}\sum_j p_j{}^2 + V(Q) + \frac{1}{2}\sum_j \omega_j{}^2 \left(q_j - s_j Q\right)^2, \tag{64}$$

where P is the conjugate momentum to Q, and (q_j, p_j) is the canonical pair for the bath modes. The potential energy is given by the one-dimensional potential $V(Q)$ plus the harmonic potential with frequencies ω_j for the bath modes q_j coupled to Q with the coupling strength given by the constants s_j.

The Hamiltonian equations of motion derived from Eq. (64) are

$$\dot{Q} = P$$
$$\dot{P} = -\frac{\mathrm{d}V}{\mathrm{d}Q} + \sum_j \omega_j{}^2 s_j \left(q_j - s_j Q\right)$$
$$\dot{q}_j = p_j$$
$$\dot{p}_j = -\omega_j{}^2 \left(q_j - s_j Q\right) \tag{65}$$

The following variables are useful in the following calculations:

$$a_j = \frac{1}{2}\left\{\omega_j \left(q_j - s_j Q\right) + \mathrm{i}p_j\right\},$$
$$a_j^* = \frac{1}{2}\left\{\omega_j \left(q_j - s_j Q\right) - \mathrm{i}p_j\right\}, \tag{66}$$

with which the last two equations in Eq. (65) can be expressed as

$$\dot{a}_j = -\mathrm{i}\omega_j a_j - \frac{1}{2}s_j\omega_j \dot{Q}. \tag{67}$$

This can be formally solved as

$$a_j(t) = a_j(0)\exp(-\mathrm{i}\omega_j t) - \frac{s_j\omega_j}{2}\int_0^t \exp\left(-\mathrm{i}\omega_j(t - t')\right)\dot{Q}(t')\mathrm{d}t'. \tag{68}$$

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By inserting this into the first two lines of Eq. (65), the GLE for Q is obtained:

$$\ddot{Q} = -\frac{\mathrm{d}V}{\mathrm{d}Q} - \int_0^t \gamma(t - t')\dot{Q}(t')\mathrm{d}t' + \xi(t), \tag{69}$$

$$\gamma(\tau) = \sum_j \frac{(s_j\omega_j)^2}{2} \left\{ \exp\left(-\mathrm{i}\omega_j\tau\right) + \exp\left(\mathrm{i}\omega_j\tau\right) \right\} \tag{70}$$

$$\xi(t) = \sum_j s_j\omega_j \left\{ a(0)\exp(-\mathrm{i}\omega_j t) + a^*(0)\exp(\mathrm{i}\omega_j t) \right\} \tag{71}$$

The friction kernel of Eq. (70) can be put into the form of Eq. (4) by setting

$$c_{2j} = \frac{(s_j\omega_j)^2}{2}, \qquad\qquad \alpha_{2j} = \omega_j,$$

$$c_{2j+1} = \frac{(s_j\omega_j)^2}{2}, \qquad\qquad \alpha_{2j+1} = -\omega_j. \tag{72}$$

To proceed with the calculation, the small damping factor is again needed since the friction kernel contains pure oscillations. In Eq. (S2) in supplemental material,[55] it is shown that the convolution with k_m extracts only the term $\exp(\mathrm{i}\alpha_m t)$. Therefore, from Eq. (71), the decomposition of the random force is given by

$$\xi_m(t) = \int_0^{+\infty} k_m(t - t')\xi(t')\mathrm{d}t' = \begin{cases} s_j\omega_j a^*(0)\exp(\mathrm{i}\omega_j t) & (\text{for } m = 2j) \\ s_j\omega_j a(0)\exp(-\mathrm{i}\omega_j t) & (\text{for } m = 2j + 1) \end{cases}. \tag{73}$$

Together with

$$Z_m = \begin{cases} \dfrac{(s_j\omega_j)^2}{2} \displaystyle\int_0^t \exp\left(\mathrm{i}\omega_j(t - t')\right) \dot{Q}(t')\mathrm{d}t' & (\text{for } m = 2j) \\[2mm] \dfrac{(s_j\omega_j)^2}{2} \displaystyle\int_0^t \exp\left(-\mathrm{i}\omega_j(t - t')\right) \dot{Q}(t')\mathrm{d}t' & (\text{for } m = 2j + 1) \end{cases}, \tag{74}$$

the EXEMs are obtained as

$$X_m = -Z_m + \xi_m = \begin{cases} s_j\omega_j a_j^* & (\text{for } m = 2j) \\ s_j\omega_j a_j & (\text{for } m = 2j + 1) \end{cases}, \tag{75}$$

due to Eq. (68). This shows that, if the underlying total system is given exactly by the sum of the subsystem and harmonic oscillators bilinearly coupled to it (Eq. (64)), the present EXEM analysis reproduces correctly the dynamical variables (a_j, a_j^*) (excepting the inessential constant multipliers), which are related to the original dynamical variables (q_j, p_j) through a simple linear transformation.

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D. Molecular time-scale GLE

Adelman[46,47] suggested a different representation of the dynamics of a chemical system that reflects more directly the molecular vibrational frequency in the short time scale. The equation of motion, called the molecular time scale GLE (MTGLE), reads

$$\ddot{Q} = -\omega_{e0}{}^2 Q + \omega_{c1}{}^4 \int_0^t \theta_1(t - t')Q(t')\mathrm{d}t' + f_1(t), \tag{76}$$

with the fluctuation-dissipation relation

$$\left\langle \dot{f}_1(t)\dot{f}_1(t') \right\rangle = \langle \dot{Q}^2 \rangle \omega_{c1}{}^4 \dot{\theta}_1(t - t'). \tag{77}$$

The difference from the conventional GLE is that (i) the Einstein frequency ω_{e0}, which describes the molecular vibration in the short time scale, is used instead of the mean force, (ii) the position coordinate Q, instead of the velocity $\dot{Q}$, appears in the friction term, and (iii) the time derivative of the random force f_1 and the time derivative of the kernel θ_1 are involved in the fluctuation-dissipation relation.

The EXEM formulated in Sec. II can similarly be defined by using the MTGLE as our starting point. Let us define γ by

$$\gamma(\tau) = \langle \dot{Q}^2 \rangle^{-1} \left\langle f_1(0)f_1(\tau) \right\rangle, \tag{78}$$

and postulate the multi-exponential form in the same manner as Eq. (4):

$$\gamma(\tau) = \sum_m c_m \exp\left(\mathrm{i}\alpha_m|\tau|\right). \tag{79}$$

The following relation holds due to Eq. (77):

$$\omega_{c1}{}^4 \dot{\theta}_1(\tau) = -\ddot{\gamma}(\tau). \tag{80}$$

Integrating this once and noting the boundary conditions $\lim_{\tau\to\infty} \theta_1(\tau) = 0$ and $\lim_{\tau\to\infty} \gamma_1(\tau) = 0$ because γ is the autocorrelation function of f_1, one obtains

$$\begin{aligned} \omega_{c1}{}^4 \theta_1(\tau) &= -\dot{\gamma}(\tau) \\ &= -\sum_m \mathrm{i}c_m\alpha_m \exp\left(\mathrm{i}\alpha_m\tau\right) \qquad \text{(for } \tau > 0\text{)}. \end{aligned}$$

$$\tag{81}$$

By defining the functions g, h, k_m in the same procedure with Sec. II staring from γ, the random force can be decomposed as

$$\xi_m(t) = \int_0^{+\infty} k_m(t - t') f_1(t') \mathrm{d}t'. \tag{82}$$

On the other hand, Z_m has to be defined in a slightly different equation reflecting the relation of Eq. (81):

$$Z_m = \int_0^t \mathrm{i}c_m \alpha_m \exp\left(\mathrm{i}\alpha_m(t - t')\right) Q(t') \mathrm{d}t'. \tag{83}$$

With these definitions, the EXEM is defined as

$$X_m = -Z_m + \xi_m, \tag{84}$$

and satisfies the following equations of motion

$$\ddot{Q} = -\omega_{\mathrm{e}0}{}^2 Q + \sum_m X_m$$

$$\dot{X}_m = \mathrm{i}\alpha_m X_m - \mathrm{i}c_m \alpha_m Q + b_m \eta(t)$$

$$\langle \eta(t)\eta(t') \rangle = \langle \dot{Q}^2 \rangle \delta(t - t'). \tag{85}$$

Adelman goes further to derive a model heat bath representation from Eq. (76). In the supplemental material,[55] it is shown that those bath coordinates and the EXEM introduced in the present paper are related through a linear transformation. As has already been discussed in Sec. II C, contribution of the present work should be found in (i) the assumption of the multi-exponential form of the friction kernel that is equivalent to the truncation of the continued-fraction expansion and avoids the need for the numerical evaluation of high order derivatives and (ii) the definition (Eqs. (82)–(84)) for the environmental modes given explicitly in terms of the observed time series $Q(t)$.

IV. SUMMARY AND OUTLOOK

Separation of the total system into the subsystem and its environment is one of the central concepts in the study of condensed phase dynamics. In the equation of motion for the subsystem, a memory term appears as a retarded response of the environment to the subsystem. It is possible to introduce effectively expressed environmental modes (EXEM)

that describe this retarded effect explicitly as dynamical motions existing in the environment. In the present paper, EXEMs were formulated with the assumption of multi-exponential form of the friction kernel appearing in the GLE. The derived equation of motion for EXEMs is equivalent to (or even more general than[44,45]) the original GLE, and describes the time evolution of the system only in terms of the current values of the variables (*viz.* without a memory term) and white noise. Since the memory term has vanished at the cost of increasing the number of variables, the newly introduced variables can be interpreted as effectively expressing the dynamical modes that exist in the environment and interacts with the subsystem. The framework is expected to provide a simple description with probably a far smaller number of variables than the all-atom description in molecular systems.

As a future outlook, it is suggested to apply the present formulation to any observed time series to obtain equations of motion that reflect the multi-dimensional dynamics hidden behind the observed time series, and, where possible, to examine the relation between the EXEM and other physical quantities existing in the same system. Since the EXEMs are explicitly given in terms of the observed time series $Q(t)$, we can obtain the time series of X_m's from the observed $Q(t)$. This time series can be compared with the time series of any other physical quantity available for the system (*e.g.* atomic coordinates of the solvents in molecular dynamics simulations). Examining their correlation would elucidate how the EXEMs obtained rather phenomenologically by the present formulation are related to the atomic coordinates in the environment, and hence it is expected to provide the molecular picture of the environmental modes.

Appendix A: Relation of the formulation in Sec. II A to the previous work

Martens[49] assumed a general friction kernel constrained by the requirement that it satisfies the following (arbitrary high order) differential equation

$$\gamma^{(n+1)} + \sum_{k=0}^{n} a_k \gamma^{(k)} = 0, \tag{A1}$$

where $\gamma^{(k)} = \mathrm{d}^k \gamma(\tau)/\mathrm{d}\tau^k$ and a_k's are constants. Factorizing the operator part, we can obtain

$$\left(\frac{\mathrm{d}}{\mathrm{d}\tau} - \mu_1 \right) \cdots \left(\frac{\mathrm{d}}{\mathrm{d}\tau} - \mu_{n+1} \right) \gamma(\tau) = 0, \tag{A2}$$

where μ_k's are the (generally complex) roots of the following algebraic equation

$$\mu^{n+1} + \sum_{k=0}^{n} a_k \mu^k = 0. \tag{A3}$$

To keep the discussion simple, let us assume that the roots $\{\mu_k\}$ are not degenerate. In this case, the solution to Eq. (A2) is given by

$$\gamma(\tau) = \sum_{m=1}^{n+1} c_m \exp(\mu_m \tau), \tag{A4}$$

with some constants c_m. Identifying the real and the imaginary parts of μ_m with $-\lambda_m$ and ω_m, respectively, Eq. (A4) is identical with Eq. (4). This proves that the assumption of the present formulation is the same with that of Ref. 49 when there is no degeneracy.

Martens proceeds to define the environmental modes by

$$z_0 = \int_0^t \gamma(t - t') \dot{Q}(t') \mathrm{d}t',$$

$$z_1 = \int_0^t \gamma^{(1)}(t - t') \dot{Q}(t') \mathrm{d}t',$$

$$\vdots$$

$$z_n = \int_0^t \gamma^{(n)}(t - t') \dot{Q}(t') \mathrm{d}t'. \tag{A5}$$

Substituting Eq. (A4) and recalling Eq. (7), one obtains the following relation

$$z_0 = \sum_m Z_m,$$

$$z_1 = \sum_m \mu_m Z_m,$$

$$\vdots$$

$$z_n = \sum_m \mu_m^n Z_m. \tag{A6}$$

Therefore the relation between the environmental modes $\{z_m\}$ of Ref. 49 and $\{Z_m\}$ defined in Sec. II A is simply the linear coordinate transformation with coefficients μ_m^n.

When there is degeneracy, the solution to Eq. (A2) becomes exponential functions multiplied by polynomials of τ. Eq. (11) would have to be modified to include multiple powers of $(\omega - \alpha_m)$.

Appendix B: Non-negativeness of the power spectrum

Let us evaluate the following integral in two ways.

$$\int_0^{+\infty} dt_1 \int_0^{+\infty} dt_2 \exp\left(-\epsilon t_1 - \epsilon t_2 + i\omega t_1 - i\omega t_2\right) \langle \xi(t_1)\xi(t_2) \rangle, \tag{B1}$$

where $\epsilon > 0$ is a damping factor to ensure the convergence of the integral. On one hand, Eq. (B1) is equal to

$$\left\langle \int_0^{+\infty} dt_1 \exp\left(-\epsilon t_1 + i\omega t_1\right) \xi(t_1) \int_0^{+\infty} dt_2 \exp\left(-\epsilon t_2 - i\omega t_2\right) \xi(t_2) \right\rangle,$$

$$= \left\langle \left(\int_0^{+\infty} dt_1 \exp\left(-\epsilon t_1 + i\omega t_1\right) \xi(t_1) \right) \left(\int_0^{+\infty} dt_2 \exp\left(-\epsilon t_2 + i\omega t_2\right) \xi(t_2) \right)^* \right\rangle,$$

$$= \left\langle \left| \int_0^{+\infty} dt \exp\left(-\epsilon t + i\omega t\right) \xi(t) \right|^2 \right\rangle, \tag{B2}$$

for real values of ω.

On the other hand, the fluctuation-dissipation relation Eq. (3) can be substituted into Eq. (B1). Let us separate the integration domain into two parts by

$$\int_0^{+\infty} dt_1 \int_0^{+\infty} dt_2 \cdots$$

$$= \int_0^{+\infty} dt_1 \int_{t_1}^{+\infty} dt_2 \cdots + \int_0^{+\infty} dt_2 \int_{t_2}^{+\infty} dt_1 \cdots . \tag{B3}$$

The first term becomes

$$\int_0^{+\infty} \mathrm{d}t_1 \int_{t_1}^{+\infty} \mathrm{d}t_2 \exp\left(-\epsilon t_1 - \epsilon t_2 + \mathrm{i}\omega t_1 - \mathrm{i}\omega t_2\right) \langle \dot{Q}^2 \rangle \gamma(t_2 - t_1)$$

$$= \int_0^{+\infty} \mathrm{d}t_1 \exp\left(-2\epsilon t_1\right) \int_0^{+\infty} \mathrm{d}\tau \exp\left(-\epsilon\tau - \mathrm{i}\omega\tau\right) \langle \dot{Q}^2 \rangle \gamma(\tau),$$

$$= \frac{1}{2\epsilon} \langle \dot{Q}^2 \rangle \int_0^{+\infty} \mathrm{d}\tau \exp\left(-\epsilon\tau - \mathrm{i}\omega\tau\right) \gamma(\tau), \tag{B4}$$

where the transformation $t_2 \mapsto \tau = t_2 - t_1$ of the integration variable has been performed on the second line. Similarly, the second term in Eq. (B3) becomes

$$\frac{1}{2\epsilon} \langle \dot{Q}^2 \rangle \int_0^{+\infty} \mathrm{d}\tau \exp\left(-\epsilon\tau + \mathrm{i}\omega\tau\right) \gamma(\tau), \tag{B5}$$

Putting them together, Eq. (B1) is equal to

$$\frac{1}{2\epsilon} \langle \dot{Q}^2 \rangle \int_{-\infty}^{+\infty} \mathrm{d}\tau \exp\left(-\epsilon|\tau| - \mathrm{i}\omega\tau\right) \gamma(\tau), \tag{B6}$$

Equating Eqs. (B2) and (B6), one obtains

$$\int_{-\infty}^{+\infty} \mathrm{d}\tau \exp\left(-\epsilon|\tau| - \mathrm{i}\omega\tau\right) \gamma(\tau) = \frac{2\epsilon}{\langle \dot{Q}^2 \rangle} \left\langle \left| \int_0^{+\infty} \mathrm{d}t \exp\left(-\epsilon t + \mathrm{i}\omega t\right) \xi(t) \right|^2 \right\rangle. \tag{B7}$$

If we take the limit of $\epsilon \to +0$, the left hand side becomes $\tilde{\gamma}(\omega)$, while the right hand side remains non-negative. This completes the proof for

$$\tilde{\gamma}(\omega) \geq 0 \qquad\qquad \text{for real } \omega. \tag{B8}$$

Appendix C: Statistical property of random force

The proof of Eq. (30) starts with noticing

$$h(\tau) = 0, \qquad\qquad \text{for } \tau > 0. \tag{C1}$$

This can be proved by noting that all the singularities β_n^* of $\tilde{h}(\omega)$ in Eq. (16) lies in the lower half of the complex ω-plane by the definition of β_n. Then, by closing the integration path by C^+ defined in Fig. 2,

$$h(\tau) = \frac{1}{2\pi} \int_{-\infty}^{+\infty} \tilde{h}(\omega) \exp\left(\mathrm{i}\omega\tau\right) \mathrm{d}\omega$$

$$= \frac{1}{2\pi} \int_{C^+} \tilde{h}(\omega) \exp\left(\mathrm{i}\omega\tau\right) \mathrm{d}\omega \qquad\qquad \text{(for } \tau > 0\text{)}$$

$$= 0. \tag{C2}$$
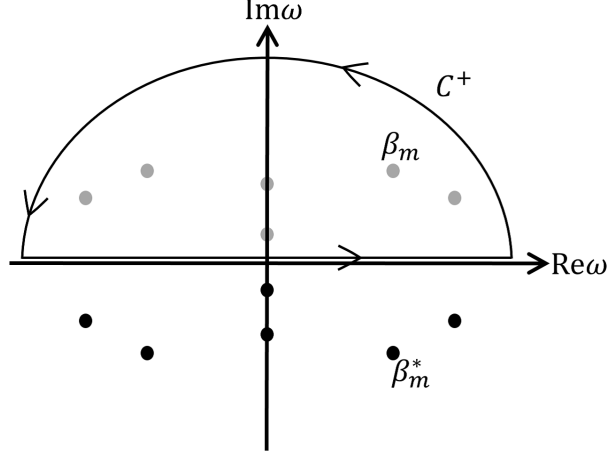
FIG. 2: Schematic picture for the distribution of the singular points of $\tilde{h}(\omega)$ in the complex ω-plane and the definition of the integration path C^+.

As a consequence, for positive t, we can extend the integration range in Eq. (23)

$$
\begin{aligned}
\eta(t) &= \int_0^{+\infty} h(t - t')\xi(t')\mathrm{d}t', \\
&= \int_{-\infty}^{+\infty} h(t - t')\xi(t')\mathrm{d}t', \qquad \text{(for } t > 0) \qquad \text{(C3)}
\end{aligned}
$$

since $h(t - t') = 0$ for $t' < 0$ if $t > 0$.

For positive t_1 and t_2, therefore,

$$
\begin{aligned}
&\langle \eta(t_1)\eta(t_2) \rangle \\
&= \int_{-\infty}^{+\infty} \mathrm{d}t_1' \int_{-\infty}^{+\infty} \mathrm{d}t_2' h(t_1 - t_1')h(t_2 - t_2')\,\langle \xi(t_1')\xi(t_2') \rangle, \\
&= \int_{-\infty}^{+\infty} \mathrm{d}t_1' \int_{-\infty}^{+\infty} \mathrm{d}t_2' h(t_1 - t_1')h(t_2 - t_2')\langle \dot{Q}^2 \rangle \frac{1}{2\pi} \int_{-\infty}^{+\infty} \mathrm{d}\omega \tilde{\gamma}(\omega) \exp\left(i\omega(t_1' - t_2')\right), \\
&= \langle \dot{Q}^2 \rangle \frac{1}{2\pi} \int_{-\infty}^{+\infty} \mathrm{d}\omega \tilde{h}(\omega)\tilde{h}(-\omega)\tilde{\gamma}(\omega) \exp\left(i\omega(t_1 - t_2)\right). \qquad \text{(C4)}
\end{aligned}
$$

By using Eqs. (16) and (15), this becomes

$$
\begin{aligned}
&= \langle \dot{Q}^2 \rangle \frac{1}{2\pi} \int_{-\infty}^{+\infty} \mathrm{d}\omega \frac{\tilde{\gamma}(\omega)}{\tilde{g}(\omega)\tilde{g}(-\omega)} \exp\left(i\omega(t_1 - t_2)\right), \\
&= 2\langle \dot{Q}^2 \rangle \frac{1}{2\pi} \int_{-\infty}^{+\infty} \mathrm{d}\omega \exp\left(i\omega(t_1 - t_2)\right), \\
&= 2\langle \dot{Q}^2 \rangle \delta(t_1 - t_2). \qquad \text{(C5)}
\end{aligned}
$$

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I. DUALITY BETWEEN THE KERNELS k_m AND EXPONENTIALS

The first part of this material is dedicated to the proof of the following relations, which will be useful in the subsequent sections.

$$k_m(\tau) = 0, \qquad\qquad \text{(for } \tau > 0) \qquad \text{(S1)}$$

$$\int_{-\infty}^{+\infty} k_m(t - t')\Theta(t')\exp\left(i\alpha_n t'\right) dt' = \delta_{mn}\Theta(t)\exp\left(i\alpha_n t\right), \qquad \text{(for } t > 0) \qquad \text{(S2)}$$

where α_m and $k_m(\tau)$ are defined in Sec. II B and Θ denotes the Heaviside step function:

$$\Theta(\tau) = \begin{cases} 1 & (\tau > 0) \\ 0 & (\tau < 0) \end{cases}. \qquad\qquad \text{(S3)}$$

In words, Eq. (S2) means that the convolution of the function $\Theta(t)\exp\left(i\alpha_n t\right)$ with the kernel $k_m(\tau)$ vanishes unless $m = n$. In the latter case the convolution keeps the original function $\exp\left(i\alpha_n t\right)$ unchanged for $t > 0$.

The proof for Eq. (S1) can be done by noting the Fourier transform of k_m (Eqs. (16) and (17)):

$$k_m(\tau) = \frac{1}{2\pi}\int_{-\infty}^{+\infty} \tilde{k}_m(\omega)\exp\left(i\omega\tau\right) d\omega = \frac{1}{2\pi}\int_{-\infty}^{+\infty} \sqrt{\frac{2}{A}}\frac{-ib_m}{\omega - \alpha_m}\frac{\prod_{m'}(w - \alpha_{m'})}{\prod_n(w - \beta_n^*)}\exp\left(i\omega\tau\right) d\omega.$$

$$\text{(S4)}$$

The integrand of the last expression has no singularity at α_m because the factor $(\omega - \alpha_m)$ is contained in the numerator. In the main text, β_n's have been defined to satisfy $\text{Im}\beta_n > 0$. All the singular points β_n^* of the integrand lie therefore only in the lower half of the complex ω-plane. For $\tau > 0$, we can close the integration path with C^+ defined in Fig. 2:

$$k_m(\tau) = \frac{1}{2\pi}\int_{C^+} \sqrt{\frac{2}{A}}\frac{-ib_m}{\omega - \alpha_m}\frac{\prod_{m'}(w - \alpha_{m'})}{\prod_n(w - \beta_n^*)}\exp\left(i\omega\tau\right) d\omega = 0, \qquad \text{(for } \tau > 0) \qquad \text{(S5)}$$

which completes the proof of Eq. (S1).

The proof for Eq. (S2) starts with recalling that the Fourier transform of the convoluted function is given by the product of the Fourier transform of each function, and the Fourier transform of $\Theta(t)\exp\left(i\alpha_n t\right)$ is

$$\int_{-\infty}^{+\infty} \Theta(t)\exp\left(i\alpha_n t\right)\exp\left(-i\omega t\right) dt = \int_0^{+\infty} \exp\left(i\alpha_n t\right)\exp\left(-i\omega t\right) dt = \frac{-i}{\omega - \alpha_n}. \qquad \text{(S6)}$$

Therefore, the Fourier transform of the left hand side of Eq. (S2) is

$$\tilde{k}_m(\omega)\frac{-\mathrm{i}}{\omega - \alpha_n}. \tag{S7}$$

By inserting the definitions of $\tilde{k}_m$ (Eq. (17)) and $\tilde{h}$ (Eq. (16)), this becomes

$$\tilde{k}_m(\omega)\frac{-\mathrm{i}}{\omega - \alpha_n} = \left(\frac{-\mathrm{i}}{\omega - \alpha_n}\right)\left(\frac{-\mathrm{i}b_m}{\omega - \alpha_m}\right)\tilde{h}(\omega) = \frac{-b_m}{(\omega - \alpha_n)(\omega - \alpha_m)}\sqrt{\frac{2}{A}}\frac{\prod_{m'}(w - \alpha_{m'})}{\prod_n(w - \beta_n^*)}. \tag{S8}$$

By the inverse Fourier transformation, therefore, the left hand side of Eq. (S2) is equal to

$$\frac{1}{2\pi}\int_{-\infty}^{+\infty}\sqrt{\frac{2}{A}}\frac{-b_m}{(\omega - \alpha_n)(\omega - \alpha_m)}\frac{\prod_{m'}(w - \alpha_{m'})}{\prod_n(w - \beta_n^*)}\exp(\mathrm{i}\omega t)\mathrm{d}\omega. \tag{S9}$$

In the case of $m \neq n$, the numerator in Eq. (S9) contains $(\omega - \alpha_n)$ and $(\omega - \alpha_m)$ once for each. These factors therefore cancel. Since β_n^*'s are all in the lower half of the complex ω-plane, the integrand in Eq. (S9) has no singular point in the upper half plane. By closing the integration path by C^+ shown in Fig. 2, therefore, it is seen that Eq. (S9) is zero for $t > 0$.

In the case $m = n$, the integrand in Eq. (S9) has one singular point at $\omega = \alpha_m$ in the upper half plane. The result of integration is therefore given by the residue at $\omega = \alpha_m$ and becomes

$$\begin{aligned}
(2\pi\mathrm{i})\lim_{\omega \to \alpha_m}\frac{1}{2\pi}&\sqrt{\frac{2}{A}}\frac{-b_m}{(\omega - \alpha_n)(\omega - \alpha_m)}\frac{\prod_{m'}(w - \alpha_{m'})}{\prod_n(w - \beta_n^*)}\exp(\mathrm{i}\omega t) \\
&= -\mathrm{i}b_m\lim_{\omega \to \alpha_m}\sqrt{\frac{2}{A}}\frac{1}{(\omega - \alpha_m)}\frac{\prod_{m'}(w - \alpha_{m'})}{\prod_n(w - \beta_n^*)}\exp(\mathrm{i}\omega t) \\
&= -\mathrm{i}b_m\lim_{\omega \to \alpha_m}\frac{1}{(\omega - \alpha_m)\tilde{g}(\omega)}\exp(\mathrm{i}\omega t),
\end{aligned} \tag{S10}$$

where the definition of $\tilde{g}$ (Eq. (12)) has been inserted. Further, by using Eq. (12),

$$\lim_{\omega \to \alpha_m}(\omega - \alpha_m)\tilde{g}(\omega) = \lim_{\omega \to \alpha_m}\sum_{m'}\frac{-\mathrm{i}b_{m'}(\omega - \alpha_m)}{\omega - \alpha_{m'}} = -\mathrm{i}b_m. \tag{S11}$$

Inserting this into Eq. (S10) gives

$$\begin{aligned}
&= -\mathrm{i}b_m\lim_{\omega \to \alpha_m}\frac{1}{(-\mathrm{i}b_m)}\exp(\mathrm{i}\omega t) \\
&= \exp(\mathrm{i}\alpha_m t).
\end{aligned} \tag{S12}$$

This completes the proof of Eq. (S2).

2

II. STATISTICAL PROPERTY OF THE INITIAL CONDITION

From the definition of ξ_m (Eq. (24)) it follows that

$$\xi_m(0) = \int_0^{+\infty} k_m(-t')\xi(t')\mathrm{d}t'. \tag{S13}$$

By combining this with the fluctuation-dissipation theorem (Eq. (3)), we can calculate the variance and covariance of $\xi_m(0)$:

$$
\begin{aligned}
\langle \xi_m(0)\xi_n(0)\rangle &= \int_0^{+\infty}\mathrm{d}t_1'\int_0^{+\infty}\mathrm{d}t_2'\; k_m(-t_1')k_n(-t_2')\,\langle \xi(t_1')\xi(t_2')\rangle \\
&= \langle \dot{Q}^2\rangle \int_0^{+\infty}\mathrm{d}t_1'\int_0^{+\infty}\mathrm{d}t_2'\; k_m(-t_1')k_n(-t_2')\gamma(t_1'-t_2').
\end{aligned} \tag{S14}
$$

We further note that, from Eq. (15) and standard calculations of the Fourier transform,

$$
\begin{aligned}
\gamma(\tau) &= \frac{1}{2\pi}\int_{-\infty}^{+\infty}\tilde{\gamma}(\omega)\exp(\mathrm{i}\omega\tau)\mathrm{d}\omega = \frac{1}{2\pi}\int_{-\infty}^{+\infty}2\tilde{g}(\omega)\tilde{g}(-\omega)\exp(\mathrm{i}\omega\tau)\mathrm{d}\omega \\
&= 2\int_{-\infty}^{+\infty} g(\tau'+\tau)g(\tau')\mathrm{d}\tau',
\end{aligned} \tag{S15}
$$

and, from Eq. (12) and because of $\mathrm{Im}\,\alpha_m > 0$,

$$
\begin{aligned}
g(\tau) &= \frac{1}{2\pi}\int_{-\infty}^{+\infty}\tilde{g}(\omega)\exp(\mathrm{i}\omega\tau)\mathrm{d}\omega = \frac{1}{2\pi}\int_{-\infty}^{+\infty}\sum_m \frac{-\mathrm{i}b_m}{\omega-\alpha_m}\exp(\mathrm{i}\omega\tau)\mathrm{d}\omega \\
&= \sum_m b_m\Theta(\tau)\exp(\mathrm{i}\alpha_m\tau),
\end{aligned} \tag{S16}
$$

where $\Theta(\tau)$ has been defined in Eq. (51). Inserting these relations into Eq. (S14), we obtain

$$
\begin{aligned}
\langle \xi_m(0)\xi_n(0)\rangle &= 2\langle \dot{Q}^2\rangle\int_0^{+\infty}\mathrm{d}t_1'\int_0^{+\infty}\mathrm{d}t_2'\int_{-\infty}^{+\infty}\mathrm{d}\tau'\; k_m(-t_1')k_n(-t_2')g(t_1'-t_2'+\tau')g(\tau') \\
&= 2\langle \dot{Q}^2\rangle\int_0^{+\infty}\mathrm{d}t_1'\int_0^{+\infty}\mathrm{d}t_2'\int_{-\infty}^{+\infty}\mathrm{d}\tau''\; k_m(-t_1')k_n(-t_2')g(t_1'+\tau'')g(t_2'+\tau''), \\
&= 2\langle \dot{Q}^2\rangle\int_{-\infty}^{+\infty}\mathrm{d}\tau''\int_0^{+\infty} k_m(-t_1')g(t_1'+\tau'')\mathrm{d}t_1'\int_0^{+\infty} k_n(-t_2')g(t_2'+\tau'')\mathrm{d}t_2',
\end{aligned} \tag{S17}
$$

where a transformation of the integration variable $\tau' \mapsto \tau'' = \tau'-t_2'$ has been performed.

By Eq. (S1), the integration range of t_1'' and t_2'' can be extended to $(-\infty, +\infty)$, and

$$
\begin{aligned}
\int_0^{+\infty} k_m(-t_1')g(t_1'+\tau'')\mathrm{d}t_1' &= \int_{-\infty}^{+\infty} k_m(-t_1')g(t_1'+\tau'')\mathrm{d}t_1' \\
&= \frac{1}{2\pi}\int_{-\infty}^{+\infty}\tilde{k}_m(\omega)\tilde{g}(\omega)\exp(\mathrm{i}\omega\tau'')\mathrm{d}\omega \\
&= \frac{1}{2\pi}\int_{-\infty}^{+\infty}\frac{-\mathrm{i}b_m}{\omega-\alpha_m}\exp(\mathrm{i}\omega\tau'')\mathrm{d}\omega \\
&= \Theta(\tau'')b_m\exp(\mathrm{i}\alpha_m\tau''),
\end{aligned} \tag{S18}
$$

3

where Eqs. (17) and (16) have been used. Inserting this into Eq. (S17),

$$\langle \xi_m(0)\xi_n(0)\rangle = 2\langle \dot{Q}^2\rangle \int_0^{+\infty} \mathrm{d}\tau'' b_m \exp(\mathrm{i}\alpha_m\tau'')b_n \exp(\mathrm{i}\alpha_n\tau'')$$

$$= 2\langle \dot{Q}^2\rangle \frac{\mathrm{i}b_m b_n}{\alpha_m + \alpha_n}. \tag{S19}$$

This completes the proof for Eq. (40)

III.   INDEPENDENCE FROM THE CHOICE OF TIME ZERO

In the GLE (Eq. (1)), $t = 0$ strands for the time at which the initial condition is prepared for the dynamical simulation, or the time at which we start the observation. In an equilibrium system, the choice of the time zero is only arbitrary and is not related to the nature of the system. Therefore, it is desirable that any physical concept utilized for the analysis be independent of this choice.

Let us examine whether the concept of EXEM introduced in the present study depends on the choice of time zero or not. In the main text, the EXEM has been defined as

$$X_m(t) = -\int_0^t c_m \exp\left(\mathrm{i}\alpha_m(t-t')\right)\dot{Q}(t')\mathrm{d}t' + \int_0^{+\infty} k_m(t-t')\xi(t')\mathrm{d}t', \tag{S20}$$

for $t \geq 0$. Note, due to Eq. (S1), the lower limit of the second integral can be extended to $-\infty$ because, if $t' < 0$, the argument $t - t'$ is positive. Thus,

$$X_m(t) = -\int_0^t c_m \exp\left(\mathrm{i}\alpha_m(t-t')\right)\dot{Q}(t')\mathrm{d}t' + \int_{-\infty}^{+\infty} k_m(t-t')\xi(t')\mathrm{d}t'. \tag{S21}$$

Let the symbol ΔF be defined as

$$\Delta F(t) = \ddot{Q}(t) - f_{\mathrm{M}}\left(Q(t)\right)$$

$$= -\int_0^t \gamma(t-t')\dot{Q}(t')\mathrm{d}t' + \xi(t), \tag{S22}$$

whose physical meaning is the deviation of the instantaneous force $\ddot{Q}(t)$ (or acceleration, to be more precise) from the mean force $f_{\mathrm{M}}\left(Q(t)\right)$. The second equality is a consequence of the GLE (1). From Eqs. (S20) and (S22), the EXEM can also be expressed as

$$X_m(t) = -\int_0^t c_m \exp\left(\mathrm{i}\alpha_m(t-t')\right)\dot{Q}(t')\mathrm{d}t'$$

$$+ \int_{-\infty}^{+\infty} \mathrm{d}t' k_m(t-t')\int_0^{t'} \mathrm{d}t''\gamma(t'-t'')\dot{Q}(t'') + \int_{-\infty}^{+\infty} k_m(t-t')\Delta F(t')\mathrm{d}t'. \tag{S23}$$

4

Suppose now we shift the reference of the time from 0 to $t^{\mathrm{o}} > 0$ (the case of $t^{\mathrm{o}} < 0$ can be treated similarly). The definition of the EXEM changes to

$$X_m^{\mathrm{o}}(t) = -\int_{t^{\mathrm{o}}}^{t} c_m \exp\left(\mathrm{i}\alpha_m(t - t')\right) \dot{Q}(t')\mathrm{d}t'$$
$$+ \int_{-\infty}^{+\infty} \mathrm{d}t' k_m(t - t') \int_{t^{\mathrm{o}}}^{t'} \mathrm{d}t'' \gamma(t' - t'')\dot{Q}(t'') + \int_{-\infty}^{+\infty} k_m(t - t')\Delta F(t')\mathrm{d}t', \quad \text{(S24)}$$

for $t \geq t^{\mathrm{o}}$, where the superscript o in X_m^{o} has been introduced to distinguish the two definitions. Taking the difference of Eqs. (S23) and (S24) results in

$$X_m(t) - X_m^{\mathrm{o}}(t) = -\int_{0}^{t^{\mathrm{o}}} c_m \exp\left(\mathrm{i}\alpha_m(t - t')\right) \dot{Q}(t')\mathrm{d}t'$$
$$+ \int_{-\infty}^{+\infty} \mathrm{d}t' k_m(t - t') \int_{0}^{t^{\mathrm{o}}} \mathrm{d}t'' \gamma(t' - t'')\dot{Q}(t'') \quad \text{(S25)}$$

Since $X_m^{\mathrm{o}}(t)$ is defined only for $t \geq t^{\mathrm{o}}$, we only think of $t \geq t^{\mathrm{o}}$. Then, in the integral of the second term of Eq. (S25), we have $0 \leq t'' \leq t^{\mathrm{o}} \leq t \leq t'$, where the last inequality is due to Eq. (S1). Because of $t'' \leq t'$ we have

$$\int_{-\infty}^{+\infty} \mathrm{d}t' k_m(t - t')\gamma(t' - t'') = \int_{-\infty}^{+\infty} \mathrm{d}t' k_m(t - t') \sum_n c_n \exp\left(\mathrm{i}\alpha_n(t' - t'')\right)$$
$$= c_m \exp\left(\mathrm{i}\alpha_m(t - t'')\right), \quad \text{(S26)}$$

due to Eq. (S2). Substituting this into Eq. (S25) gives

$$X_m(t) - X_m^{\mathrm{o}}(t) = -\int_{0}^{t^{\mathrm{o}}} c_m \exp\left(\mathrm{i}\alpha_m(t - t')\right) \dot{Q}(t')\mathrm{d}t' + \int_{0}^{t^{\mathrm{o}}} c_m \exp\left(\mathrm{i}\alpha_m(t - t'')\right) \dot{Q}(t'')\mathrm{d}t''$$
$$= 0.$$

$$\text{(S27)}$$

This completes the proof that the definition of the EXEM is independent of the choice of time zero.

An additional comment is on the concept of the random force in the GLE. Since the GLE contains the initial time, the definition of the random force depends on the choice of time zero. From Eq. (S22) one sees

$$\xi(t) = \Delta F(t) + \int_{0}^{t} \gamma(t - t')\dot{Q}(t')\mathrm{d}t'. \quad \text{(S28)}$$

When we change the reference of time to t^{o}, the definition of the random force changes to

$$\xi^{\mathrm{o}}(t) = \Delta F(t) + \int_{t^{\mathrm{o}}}^{t} \gamma(t - t')\dot{Q}(t')\mathrm{d}t'. \quad \text{(S29)}$$

5

where the definition of $\Delta F(t) = \ddot{Q}(t) - f_{\mathrm{M}}(Q(t))$ does not change since it is defined only by the physical quantities at time t. Thus, by changing the reference of time, the random force changes by

$$\xi(t) - \xi^{\mathrm{o}}(t) = \int_0^{t^{\mathrm{o}}} \gamma(t - t')\dot{Q}(t')\mathrm{d}t'. \tag{S30}$$

Although the variable X_m is defined by using the random force, the difference in the random force is canceled by the change of the term Z_m, making the total X_m eventually unchanged by the change of the reference of time.

IV. RELATION WITH THE BATH COORDINATES GENERATED BY THE CONTINUED-FRACTION APPROACH

In Ref. 43, the following equation of motion including bath coordinate $\boldsymbol{A}$ was derived through truncation of the continued-fraction expression of the Laplace transform of the friction kernel:

$$\ddot{Q} = f_{\mathrm{M}}(Q) + \Delta_1 A_1 \tag{S31}$$

$$\dot{A}_1 = -\Delta_1 \dot{Q} + \Delta_2 A_2 \tag{S32}$$

$$\dot{A}_2 = -\Delta_2 A_1 + \Delta_3 A_3 \tag{S33}$$

$$\vdots$$

$$\dot{A}_m = -\Delta_m A_{m-1} + \Delta_{m+1} A_{m+1} \tag{S34}$$

$$\vdots$$

$$\dot{A}_N = -\Delta_N A_{N-1} - \gamma_N A_N + f_N(t) \tag{S35}$$

$$\langle f_N(t) f_N(t') \rangle = 2\gamma_N \delta(t - t') \tag{S36}$$

Let us suppose this equation accurately describes the given system, that is, the truncation is made at sufficiently large N.

Eqs. (S32)–(S35) can be put into the following matrix form:

$$\dot{\boldsymbol{A}} = \boldsymbol{L}\boldsymbol{A} - \Delta_1 \dot{Q}\boldsymbol{e}_1 + f_N(t)\boldsymbol{e}_N, \tag{S37}$$

where $\boldsymbol{A}$, $\boldsymbol{e}_1$, and $\boldsymbol{e}_N$ are the following N-dimensional vectors

$$\boldsymbol{A} = \begin{pmatrix} A_1 \\ A_2 \\ \vdots \\ A_N \end{pmatrix}, \qquad \boldsymbol{e}_1 = \begin{pmatrix} 1 \\ 0 \\ \vdots \\ 0 \end{pmatrix}, \qquad \boldsymbol{e}_N = \begin{pmatrix} 0 \\ \vdots \\ 0 \\ 1 \end{pmatrix}, \qquad \text{(S38)}$$

and $\boldsymbol{L}$ is the following $N \times N$ matrix

$$\boldsymbol{L} = \begin{pmatrix} 0 & \Delta_2 & 0 & 0 & \cdots & 0 & 0 \\ -\Delta_2 & 0 & \Delta_3 & 0 & \cdots & 0 & 0 \\ 0 & -\Delta_3 & 0 & \Delta_4 & \cdots & 0 & 0 \\ \vdots & & \ddots & \ddots & \ddots & & \vdots \\ \vdots & & & \ddots & \ddots & \ddots & \vdots \\ 0 & & & & -\Delta_{N-1} & 0 & \Delta_N \\ 0 & 0 & 0 & \cdots & 0 & -\Delta_N & -\gamma_N \end{pmatrix}. \qquad \text{(S39)}$$

Eq. (S37) can be formally solved by introducing the eigenvectors of $\boldsymbol{L}$. Let the eigenvalues of $\boldsymbol{L}$ be $\mathrm{i}\alpha_1, \mathrm{i}\alpha_2, \ldots, \mathrm{i}\alpha_N$, and the corresponding right and left eigenvectors $\boldsymbol{u}_m$ and $\boldsymbol{v}_m^{\mathsf{T}}$, respectively:

$$\begin{aligned} \boldsymbol{L}\boldsymbol{u}_m &= \mathrm{i}\alpha_m \boldsymbol{u}_m \\ \boldsymbol{v}_m^{\mathsf{T}}\boldsymbol{L} &= \mathrm{i}\alpha_m \boldsymbol{v}_m^{\mathsf{T}}. \end{aligned} \qquad \text{(S40)}$$

Note that the right and left eigenvectors are not the same since the matrix $\boldsymbol{L}$ is not symmetric. The following calculation shows that the eigenvectors satisfy $\boldsymbol{v}_n^{\mathsf{T}}\boldsymbol{u}_m = 0$ for $m \neq n$:

$$\mathrm{i}\alpha_n \boldsymbol{v}_n^{\mathsf{T}}\boldsymbol{u}_m = \boldsymbol{v}_n^{\mathsf{T}}\boldsymbol{L}\boldsymbol{u}_m = \mathrm{i}\alpha_m \boldsymbol{v}_n^{\mathsf{T}}\boldsymbol{u}_m. \qquad \text{(S41)}$$

Further, let us normalize the vectors in such a way that the following is satisfied:

$$\boldsymbol{v}_n^{\mathsf{T}}\boldsymbol{u}_m = \delta_{nm}. \qquad \text{(S42)}$$

If we expand the bath coordinate $\boldsymbol{A}$ as

$$\boldsymbol{A} = \sum_m \tilde{A}_m \boldsymbol{u}_m \qquad \text{(S43)}$$

by using the eigenvectors, the equation of motion (Eq. (S37)) transforms to

$$\dot{\tilde{A}}_m = \mathrm{i}\alpha_m \tilde{A}_m - \Delta_1 \left(\boldsymbol{v}_m^\mathsf{T} \boldsymbol{e}_1\right) \dot{Q} + \left(\boldsymbol{v}_m^\mathsf{T} \boldsymbol{e}_N\right) f_N(t), \tag{S44}$$

which can be formally solved as

$$\tilde{A}_m(t) = \tilde{A}_m(0) \exp(\mathrm{i}\alpha_m t) + \left(\boldsymbol{v}_m^\mathsf{T} \boldsymbol{e}_N\right) \int_0^t \exp\left(\mathrm{i}\alpha_m(t - t')\right) f_N(t')\mathrm{d}t'$$
$$- \Delta_1 \left(\boldsymbol{v}_m^\mathsf{T} \boldsymbol{e}_1\right) \int_0^t \exp\left(\mathrm{i}\alpha_m(t - t')\right) \dot{Q}(t')\mathrm{d}t' \tag{S45}$$

Substituting A_1 obtained from $A_1 = \boldsymbol{e}_1^\mathsf{T} \boldsymbol{A}$ and Eqs. (S43) and (S45) into Eq. (S31) and comparing the result with the GLE, we obtain the following identification:

$$\gamma(\tau) = \sum_m \Delta_1{}^2 \left(\boldsymbol{e}_1^\mathsf{T} \boldsymbol{u}_m\right) \left(\boldsymbol{v}_m^\mathsf{T} \boldsymbol{e}_1\right) \exp(\mathrm{i}\alpha_m \tau),$$

$$\xi(t) = \sum_m \Delta_1 \left(\boldsymbol{e}_1^\mathsf{T} \boldsymbol{u}_m\right) \left\{ \tilde{A}_m(0) \exp(\mathrm{i}\alpha_m t) + \left(\boldsymbol{v}_m^\mathsf{T} \boldsymbol{e}_N\right) \int_0^t \exp\left(\mathrm{i}\alpha_m(t - t')\right) f_N(t')\mathrm{d}t' \right\}. \tag{S46}$$

Comparison with Eq. (4) yields

$$c_m = \Delta_1{}^2 \left(\boldsymbol{e}_1^\mathsf{T} \boldsymbol{u}_m\right) \left(\boldsymbol{v}_m^\mathsf{T} \boldsymbol{e}_1\right) \tag{S47}$$

Next we show that the solution to Eq. (31) is given by

$$b_m = \Delta_1 \sqrt{\gamma_N} \left(\boldsymbol{e}_1^\mathsf{T} \boldsymbol{u}_m\right) \left(\boldsymbol{v}_m^\mathsf{T} \boldsymbol{e}_N\right). \tag{S48}$$

To prove this, we first note the matrix $\boldsymbol{L}$ can be decomposed as

$$\boldsymbol{L} = \boldsymbol{D} - \gamma_N \boldsymbol{e}_N \boldsymbol{e}_N^\mathsf{T},$$

$$\boldsymbol{D} = \begin{pmatrix} 0 & \Delta_2 & 0 & 0 & \cdots & 0 \\ -\Delta_2 & 0 & \Delta_3 & 0 & \cdots & 0 \\ 0 & -\Delta_3 & 0 & \ddots & & 0 \\ \vdots & & \ddots & \ddots & \ddots & \vdots \\ 0 & & & -\Delta_{N-1} & 0 & \Delta_N \\ 0 & 0 & \cdots & 0 & -\Delta_N & 0 \end{pmatrix}, \tag{S49}$$

and the matrix $\boldsymbol{D}$ thus defined is antisymmetric: $\boldsymbol{D}^\mathsf{T} = -\boldsymbol{D}$. The definition of the left eigenvector $\boldsymbol{v}_m$, Eq. (S40), then becomes

$$\boldsymbol{v}_m^\mathsf{T} \boldsymbol{D} - \gamma_N \left(\boldsymbol{v}_m^\mathsf{T} \boldsymbol{e}_N\right) \boldsymbol{e}_N^\mathsf{T} = \mathrm{i}\alpha_m \boldsymbol{v}_m^\mathsf{T}. \tag{S50}$$

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Taking the transpose of Eq. (S50) and replacing the index m with n yields

$$-\boldsymbol{D}\boldsymbol{v}_n - \gamma_N \left(\boldsymbol{v}_n^\mathsf{T}\boldsymbol{e}_N\right)\boldsymbol{e}_N = \mathrm{i}\alpha_n \boldsymbol{v}_n. \tag{S51}$$

Multiplying $\boldsymbol{v}_m^\mathsf{T}$ from the left gives

$$-\boldsymbol{v}_m^\mathsf{T}\boldsymbol{D}\boldsymbol{v}_n - \gamma_N \left(\boldsymbol{v}_n^\mathsf{T}\boldsymbol{e}_N\right)\left(\boldsymbol{v}_m^\mathsf{T}\boldsymbol{e}_N\right) = \mathrm{i}\alpha_n \left(\boldsymbol{v}_m^\mathsf{T}\boldsymbol{v}_n\right). \tag{S52}$$

On the other hand, multiplying $\boldsymbol{v}_n$ on Eq. (S50) from the right gives

$$\boldsymbol{v}_m^\mathsf{T}\boldsymbol{D}\boldsymbol{v}_n - \gamma_N \left(\boldsymbol{v}_m^\mathsf{T}\boldsymbol{e}_N\right)\left(\boldsymbol{e}_N^\mathsf{T}\boldsymbol{v}_n\right) = \mathrm{i}\alpha_m \left(\boldsymbol{v}_m^\mathsf{T}\boldsymbol{v}_n\right). \tag{S53}$$

Summing Eqs. (S52) and (S53) gives (note $\left(\boldsymbol{e}_N^\mathsf{T}\boldsymbol{v}_n\right) = \left(\boldsymbol{v}_n^\mathsf{T}\boldsymbol{e}_N\right)$ since it is a scalar product)

$$-2\gamma_N \left(\boldsymbol{v}_m^\mathsf{T}\boldsymbol{e}_N\right)\left(\boldsymbol{v}_n^\mathsf{T}\boldsymbol{e}_N\right) = \left(\mathrm{i}\alpha_n + \mathrm{i}\alpha_m\right)\left(\boldsymbol{v}_m^\mathsf{T}\boldsymbol{v}_n\right). \tag{S54}$$

Now let us calculate the right hand side of Eq. (31) with b_m given by Eq. (S48), and see if it is equal to the left hand side of Eq. (31).

$$\begin{aligned}
2\mathrm{i}\sum_n \frac{b_m b_n}{\alpha_m + \alpha_n} &= 2\mathrm{i}\sum_n \frac{1}{\alpha_m + \alpha_n}{\Delta_1}^2 \gamma_N \left(\boldsymbol{e}_1^\mathsf{T}\boldsymbol{u}_m\right)\left(\boldsymbol{v}_m^\mathsf{T}\boldsymbol{e}_N\right)\left(\boldsymbol{e}_1^\mathsf{T}\boldsymbol{u}_n\right)\left(\boldsymbol{v}_n^\mathsf{T}\boldsymbol{e}_N\right) \\
&= {\Delta_1}^2 \sum_n \left(\boldsymbol{e}_1^\mathsf{T}\boldsymbol{u}_m\right)\left(\boldsymbol{v}_m^\mathsf{T}\boldsymbol{v}_n\right)\left(\boldsymbol{e}_1^\mathsf{T}\boldsymbol{u}_n\right) \\
&= {\Delta_1}^2 \sum_n \left(\boldsymbol{e}_1^\mathsf{T}\boldsymbol{u}_m\right)\left(\boldsymbol{e}_1^\mathsf{T}\boldsymbol{u}_n\right)\left(\boldsymbol{v}_n^\mathsf{T}\boldsymbol{v}_m\right) \\
&= {\Delta_1}^2 \left(\boldsymbol{e}_1^\mathsf{T}\boldsymbol{u}_m\right)\left(\boldsymbol{e}_1^\mathsf{T}\boldsymbol{v}_m\right),
\end{aligned} \tag{S55}$$

where the first line is substitution of Eq. (S48), in the second line Eq. (S54) was used, in the third line the orders of the inner products were exchanged, and in the fourth line the completeness $\left(\sum_n \boldsymbol{u}_n\boldsymbol{v}_n^\mathsf{T} = 1\right)$ of the eigensystem was used. From Eq. (S47) it is seen that this is indeed equal to c_m.

With $\{b_m\}$ thus obtained, the random force $\xi(t)$ of Eq. (S46) can be written as

$$\begin{aligned}
\xi(t) &= \sum_m \left\{\Delta_1 \left(\boldsymbol{e}_1^\mathsf{T}\boldsymbol{u}_m\right)\tilde{A}_m(0)\exp(\mathrm{i}\alpha_m t) + \int_0^t b_m \exp\left(\mathrm{i}\alpha_m(t-t')\right)\frac{f_N(t')}{\sqrt{\gamma_N}}\mathrm{d}t'\right\} \\
&= \sum_m \Delta_1 \left(\boldsymbol{e}_1^\mathsf{T}\boldsymbol{u}_m\right)\tilde{A}_m(0)\exp(\mathrm{i}\alpha_m t) + \int_0^t g(t-t')\frac{f_N(t')}{\sqrt{\gamma_N}}\mathrm{d}t',
\end{aligned} \tag{S56}$$

due to the definition of $g(\tau) = \Theta(\tau)\sum_m b_m \exp(\mathrm{i}\alpha_m \tau)$ (Eq. (12)). The definition of ξ_m (Eq. (24)) gives

$$\begin{aligned}
\xi_m(t) &= \int_0^{+\infty} k_m(t-t')\xi(t')\mathrm{d}t' \\
&= \Delta_1 \left(\boldsymbol{e}_1^\mathsf{T}\boldsymbol{u}_m\right)\tilde{A}_m(0)\exp(\mathrm{i}\alpha_m t) + b_m \int_0^t \exp\left(\mathrm{i}\alpha_m(t-t')\right)\frac{f_N(t')}{\sqrt{\gamma_N}}\mathrm{d}t',
\end{aligned} \tag{S57}$$

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where Eq. (S2) has been used for the first term and, for the second term,

$$
\int_0^{+\infty} \mathrm{d}t' k_m(t-t') \int_0^{t'} \mathrm{d}t'' g(t'-t'') \frac{f_N(t'')}{\sqrt{\gamma_N}}
$$

$$
= \int_0^{+\infty} \mathrm{d}t'' \int_{t''}^{+\infty} \mathrm{d}t' k_m(t-t') g(t'-t'') \frac{f_N(t'')}{\sqrt{\gamma_N}}
$$

$$
= \frac{1}{(2\pi)^2} \int_0^{+\infty} \mathrm{d}t'' \int_{-\infty}^{+\infty} \mathrm{d}t' \int_{-\infty}^{+\infty} \mathrm{d}\omega_1 \tilde{k}_m(\omega_1) \exp(i\omega_1(t-t')) \int_{-\infty}^{+\infty} \mathrm{d}\omega_2 \tilde{g}(\omega_2) \exp(i\omega_2(t'-t'')) \frac{f_N(t'')}{\sqrt{\gamma_N}}
$$

$$
= \frac{1}{2\pi} \int_0^{+\infty} \mathrm{d}t'' \int_{-\infty}^{+\infty} \mathrm{d}\omega_1 \int_{-\infty}^{+\infty} \mathrm{d}\omega_2 \tilde{k}_m(\omega_1) \tilde{g}(\omega_2) \exp\left(i\omega_1 t - i\omega_2 t''\right) \delta(-\omega_1+\omega_2) \frac{f_N(t'')}{\sqrt{\gamma_N}}
$$

$$
= \frac{1}{2\pi} \int_0^{+\infty} \mathrm{d}t'' \int_{-\infty}^{+\infty} \mathrm{d}\omega \tilde{k}_m(\omega) \tilde{g}(\omega) \exp(i\omega(t-t'')) \frac{f_N(t'')}{\sqrt{\gamma_N}}
$$

$$
= \frac{1}{2\pi} \int_0^{+\infty} \mathrm{d}t'' \int_{-\infty}^{+\infty} \mathrm{d}\omega \frac{-ib_m}{\omega - \alpha_m} \exp(i\omega(t-t'')) \frac{f_N(t'')}{\sqrt{\gamma_N}}
$$

$$
= \int_0^t \mathrm{d}t'' b_m \exp(i\alpha_m(t-t'')) \frac{f_N(t'')}{\sqrt{\gamma_N}}, \tag{S58}
$$

where in the third line the integration range of t' has been extended because $g(\tau) = \Theta(\tau) \sum_m b_m \exp(i\alpha_m \tau)$ is zero for $\tau < 0$.

Finally, the EXEM introduced in the present paper becomes, from Eq. (28),

$$
X_m = - \int_0^t c_m \exp\left(i\alpha_m(t-t')\right) \dot{Q}(t') \mathrm{d}t'
$$

$$
+ \Delta_1 \left(\boldsymbol{e}_1^\mathsf{T} \boldsymbol{u}_m\right) \tilde{A}_m(0) \exp(i\alpha_m t) + b_m \int_0^t \exp\left(i\alpha_m(t-t')\right) \frac{f_N(t')}{\sqrt{\gamma_N}} \mathrm{d}t',
$$

$$
= - \Delta_1{}^2 \left(\boldsymbol{e}_1^\mathsf{T} \boldsymbol{u}_m\right) \left(\boldsymbol{v}_m^\mathsf{T} \boldsymbol{e}_1\right) \int_0^t \exp\left(i\alpha_m(t-t')\right) \dot{Q}(t') \mathrm{d}t'
$$

$$
+ \Delta_1 \left(\boldsymbol{e}_1^\mathsf{T} \boldsymbol{u}_m\right) \tilde{A}_m(0) \exp(i\alpha_m t) + \Delta_1 \left(\boldsymbol{e}_1^\mathsf{T} \boldsymbol{u}_m\right) \left(\boldsymbol{v}_m^\mathsf{T} \boldsymbol{e}_N\right) \int_0^t \exp\left(i\alpha_m(t-t')\right) f_N(t') \mathrm{d}t',
$$

$$
= \Delta_1 \left(\boldsymbol{e}_1^\mathsf{T} \boldsymbol{u}_m\right) \tilde{A}_m(t), \tag{S59}
$$

where the Eq. (S48) for b_m, Eq. (S47) for c_m, and Eq. (S45) for $\tilde{A}_m(t)$ have been used. This shows that the EXEM is equal to the dynamical coordinate $\tilde{A}_m$ multiplied by a constant. Since $\tilde{A}_m$ is given by a linear transformation from the original bath coordinates A_m, it is concluded that the bath coordinate introduced by the continued-fraction approach and the EXEM in the present study are related through a linear transformation.

V. RELATION WITH THE MODEL HEAD BATH COORDINATES OF THE MT-GLE

In Refs. 46 and 47, Adelman derived a model heat bath representation from Eq. (76). This is achieved by (i) defining the sum of the second and third term as a new dynamical coordinate for the bath, (ii) successively deriving the MTGLE for the bath coordinate, and (iii) truncating when the random force can be approximated by a white noise. The resulting equations of motion read

$$\ddot{Q} = -\omega_{\mathrm{e}0}{}^2 Q + \omega_{\mathrm{c}1}{}^2 Q_1 \tag{S60}$$

$$\ddot{Q}_1 = \omega_{\mathrm{c}1}{}^2 Q - \omega_{\mathrm{e}1}{}^2 Q_1 + \omega_{\mathrm{c}2}{}^2 Q_2 \tag{S61}$$

$$\ddot{Q}_2 = \omega_{\mathrm{c}2}{}^2 Q_1 - \omega_{\mathrm{e}2}{}^2 Q_2 + \omega_{\mathrm{c}3}{}^2 Q_3 \tag{S62}$$

$$\vdots$$

$$\ddot{Q}_N = \omega_{\mathrm{c}N}{}^2 Q_{N-1} - \Omega_N{}^2 Q_N - \gamma_N \dot{Q}_N + \xi_{\mathrm{w}}, \tag{S63}$$

$$\langle \xi_{\mathrm{w}}(t)\xi_{\mathrm{w}}(t') \rangle = 2\langle \dot{Q}^2 \rangle \gamma_N \delta(t - t'), \tag{S64}$$

Let us suppose now that this set of equations describe the given system accurately, that is, the truncation is made at sufficiently large N. Eqs. (S61)-(S63) can be put into the following matrix form:

$$\frac{\mathrm{d}}{\mathrm{d}t} \begin{pmatrix} \boldsymbol{Q}_{\mathrm{b}} \\ \dot{\boldsymbol{Q}}_{\mathrm{b}} \end{pmatrix} = \begin{pmatrix} \boldsymbol{O} & \mathbf{1} \\ \boldsymbol{S} & -\gamma_N \boldsymbol{E}_N \end{pmatrix} \begin{pmatrix} \boldsymbol{Q}_{\mathrm{b}} \\ \dot{\boldsymbol{Q}}_{\mathrm{b}} \end{pmatrix} + \omega_{\mathrm{c}1}{}^2 Q \boldsymbol{e}_{N+1} + \xi_{\mathrm{w}} \boldsymbol{e}_{2N}, \tag{S65}$$

where $\boldsymbol{Q}_{\mathrm{b}} = (Q_1, Q_2, \ldots, Q_N)^{\mathsf{T}}$ is the column vector containing all the bath coordinates (but not Q), $\boldsymbol{O}$ and $\mathbf{1}$ denote the $N \times N$ zero and identity matrices, respectively, $\boldsymbol{S}$ and $\boldsymbol{E}_N$ are the following $N \times N$ matrices,

$$\boldsymbol{S} = \begin{pmatrix} -\omega_{\mathrm{e}1}{}^2 & \omega_{\mathrm{c}2}{}^2 & 0 & \cdots & \cdots & 0 \\ \omega_{\mathrm{c}2}{}^2 & -\omega_{\mathrm{e}2}{}^2 & \omega_{\mathrm{c}3}{}^2 & 0 & \cdots & 0 \\ 0 & \omega_{\mathrm{c}3}{}^2 & -\omega_{\mathrm{e}3}{}^2 & & & \vdots \\ \vdots & & & \ddots & & 0 \\ \vdots & & & & \ddots & \omega_{\mathrm{c}N}{}^2 \\ 0 & \cdots & & 0 & \omega_{\mathrm{c}N}{}^2 & -\Omega_N{}^2 \end{pmatrix}, \quad \boldsymbol{E}_N = \begin{pmatrix} 0 & \cdots & \cdots & \cdots & 0 & 0 \\ \vdots & \ddots & & & \vdots & \vdots \\ \vdots & & \ddots & & \vdots & \vdots \\ \vdots & & & \ddots & \vdots & \vdots \\ 0 & \cdots & \cdots & \cdots & 0 & 0 \\ 0 & \cdots & \cdots & \cdots & 0 & 1 \end{pmatrix}, \tag{S66}$$

and $\boldsymbol{e}_{2N}$ and $\boldsymbol{e}_N$ are the following $2N$-dimensional vectors:

$$\boldsymbol{e}_{2N} = \begin{pmatrix} 0 \\ 0 \\ \vdots \\ \vdots \\ \vdots \\ 0 \\ 1 \end{pmatrix}, \qquad \boldsymbol{e}_{N+1} = \begin{pmatrix} 0 \\ \vdots \\ 0 \\ 1 \\ 0 \\ \vdots \\ 0 \end{pmatrix} \prec N + 1 . \tag{S67}$$

Eq. (S65) can be formally solved by introducing the eigenvectors of the matrix

$$\boldsymbol{L} = \begin{pmatrix} \boldsymbol{O} & \boldsymbol{1} \\ \boldsymbol{S} & -\gamma_N \boldsymbol{E}_N \end{pmatrix} . \tag{S68}$$

Let the eigenvalues of $\boldsymbol{L}$ be $\mathrm{i}\alpha_1$, $\mathrm{i}\alpha_2$, ..., and $\boldsymbol{u}_m$ the corresponding right eigenvector:

$$\boldsymbol{L}\boldsymbol{u}_m = \mathrm{i}\alpha_m \boldsymbol{u}_m. \tag{S69}$$

Let $\check{\boldsymbol{u}}_m$ denote the upper N components of $\boldsymbol{u}_m$. Eq. (S69) leads to the following relations:

$$\boldsymbol{u}_m = \begin{pmatrix} \check{\boldsymbol{u}}_m \\ \mathrm{i}\alpha_m \check{\boldsymbol{u}}_m \end{pmatrix},$$

$$\left(\boldsymbol{S} - \mathrm{i}\alpha_m \gamma \boldsymbol{E}_N + \alpha_m{}^2 \right) \check{\boldsymbol{u}}_m = \boldsymbol{0}. \tag{S70}$$

The left eigenvectors of $\boldsymbol{L}$ are given by using $\check{\boldsymbol{e}}_m$ as follows:

$$\boldsymbol{v}_m^{\mathsf{T}} = \left(\check{\boldsymbol{u}}_m^{\mathsf{T}} \boldsymbol{S}, \quad \mathrm{i}\alpha_m \check{\boldsymbol{u}}_m^{\mathsf{T}} \right) \tag{S71}$$

$$\boldsymbol{v}_m^{\mathsf{T}} \boldsymbol{L} = \mathrm{i}\alpha_m \boldsymbol{v}_m^{\mathsf{T}}. \tag{S72}$$

That the vector $\boldsymbol{v}_m$ defined by Eq. (S71) satisfies the eigenvalue equation (S72) can be proved readily by taking the transpose of Eq. (S70).

The right and left eigenvectors satisfy the relation $\boldsymbol{v}_n^{\mathsf{T}} \boldsymbol{u}_m = 0$ for $m \neq n$ as is shown by the following calculation:

$$\mathrm{i}\alpha_n \boldsymbol{v}_n^{\mathsf{T}} \boldsymbol{u}_m = \boldsymbol{v}_n^{\mathsf{T}} \boldsymbol{L} \boldsymbol{u}_m = \mathrm{i}\alpha_m \boldsymbol{v}_n^{\mathsf{T}} \boldsymbol{u}_m. \tag{S73}$$

Let us further assume that the vector $\check{\boldsymbol{u}}_m$ is normalized in such a way that

$$\boldsymbol{v}_n^\mathsf{T} \boldsymbol{u}_m = \delta_{mn}, \tag{S74}$$

where δ_{mn} denotes the Kronecker delta. Substitution of Eqs. (S70) and (S72) into Eq. (S74) leads to

$$\check{\boldsymbol{u}}_n^\mathsf{T} \boldsymbol{S} \check{\boldsymbol{u}}_m - \alpha_n \alpha_m \check{\boldsymbol{u}}_n^\mathsf{T} \check{\boldsymbol{u}}_m = \delta_{mn}. \tag{S75}$$

Now we perform the following eigenvector expansion

$$\begin{pmatrix} \boldsymbol{Q}_\mathrm{b} \\ \dot{\boldsymbol{Q}}_\mathrm{b} \end{pmatrix} = \sum_m y_m \boldsymbol{u}_m, \tag{S76}$$

to introduce a coordinate transformation from $(\boldsymbol{Q}_\mathrm{b}, \dot{\boldsymbol{Q}}_\mathrm{b})$ to (y_m). Substituting this into the equation of motion (Eq. (S65)) and multiplying $\boldsymbol{v}_m^\mathsf{T}$ from the left results in

$$\dot{y}_m = \mathrm{i}\alpha_m y_m + \omega_{\mathrm{c}1}{}^2 \left(\boldsymbol{v}_m^\mathsf{T} \boldsymbol{e}_{N+1}\right) Q + \left(\boldsymbol{v}_m^\mathsf{T} \boldsymbol{e}_{2N}\right) \xi_\mathrm{w}, \tag{S77}$$

which can be formally solved as

$$y_m(t) = y_m(0) \exp\left(\mathrm{i}\alpha_m t\right) + \int_0^t \omega_{\mathrm{c}1}{}^2 \left(\boldsymbol{v}_m^\mathsf{T} \boldsymbol{e}_{N+1}\right) \exp\left(\mathrm{i}\alpha_m(t-t')\right) Q(t') \mathrm{d}t'$$

$$+ \int_0^t \left(\boldsymbol{v}_m^\mathsf{T} \boldsymbol{e}_{2N}\right) \exp\left(\mathrm{i}\alpha_m(t-t')\right) \xi_\mathrm{w}(t') \mathrm{d}t'. \tag{S78}$$

Substituting this into Eq. (S76) gives $(\boldsymbol{Q}_\mathrm{b}, \dot{\boldsymbol{Q}}_\mathrm{b})$. Substituting this solution into Q_1 in Eq. (S60) one obtains

$$\ddot{Q} = -\omega_{\mathrm{e}0}{}^2 Q + \sum_m \omega_{\mathrm{c}1}{}^4 \left(\boldsymbol{e}_1^\mathsf{T} \boldsymbol{u}_m\right) \left(\boldsymbol{v}_m^\mathsf{T} \boldsymbol{e}_{N+1}\right) \int_0^t \exp\left(\mathrm{i}\alpha_m(t-t')\right) Q(t') \mathrm{d}t'$$

$$+ \sum_m \omega_{\mathrm{c}1}{}^2 \left(\boldsymbol{e}_1^\mathsf{T} \boldsymbol{u}_m\right) \left\{ y_m(0) \exp\left(\mathrm{i}\alpha_m t\right) + \int_0^t \left(\boldsymbol{v}_m^\mathsf{T} \boldsymbol{e}_{2N}\right) \exp\left(\mathrm{i}\alpha_m(t-t')\right) \xi_\mathrm{w}(t') \mathrm{d}t' \right\}, \tag{S79}$$

where $\boldsymbol{e}_1^\mathsf{T} = (1, 0, \ldots, 0)$. By comparing this with the MTGLE (76), the following identification is obtained

$$\theta_1(\tau) = \sum_m \left(\boldsymbol{e}_1^\mathsf{T} \boldsymbol{u}_m\right) \left(\boldsymbol{v}_m^\mathsf{T} \boldsymbol{e}_{N+1}\right) \exp\left(\mathrm{i}\alpha_m \tau\right)$$

$$f_1(t) = \sum_m \omega_{\mathrm{c}1}{}^2 \left(\boldsymbol{e}_1^\mathsf{T} \boldsymbol{u}_m\right) \left\{ y_m(0) \exp\left(\mathrm{i}\alpha_m t\right) + \int_0^t \left(\boldsymbol{v}_m^\mathsf{T} \boldsymbol{e}_{2N}\right) \exp\left(\mathrm{i}\alpha_m(t-t')\right) \xi_\mathrm{w}(t') \mathrm{d}t' \right\}, \tag{S80}$$

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Comparison with Eq. (81) yields

$$c_m = \mathrm{i}\alpha_m{}^{-1}\omega_{\mathrm{c}1}{}^4 \left(e_1^\mathsf{T} u_m\right) \left(v_m^\mathsf{T} e_{N+1}\right) \tag{S81}$$

Next we show that the solution to Eq. (31) is given by

$$b_m = \omega_{\mathrm{c}1}{}^2 \sqrt{\gamma_N} \left(e_1^\mathsf{T} u_m\right) \left(v_m^\mathsf{T} e_{2N}\right). \tag{S82}$$

The proof proceeds by calculating the right hand side of Eq. (31) with Eq. (S82). In the numerator, $b_m b_n$ is

$$b_m b_n = \omega_{\mathrm{c}1}{}^4 \gamma_N \left(e_1^\mathsf{T} u_m\right) \left(v_m^\mathsf{T} e_{2N}\right) \left(e_1^\mathsf{T} u_n\right) \left(v_n^\mathsf{T} e_{2N}\right). \tag{S83}$$

Here,

$$
\begin{aligned}
\left(v_m^\mathsf{T} e_{2N}\right) \left(v_n^\mathsf{T} e_{2N}\right) &= \left(v_m^\mathsf{T} e_{2N}\right) \left(e_{2N}^\mathsf{T} v_n\right) \\
&= -\alpha_m \alpha_n \check{u}_m^\mathsf{T} E_N \check{u}_n \\
&= -\frac{\alpha_m \alpha_n}{\mathrm{i}\alpha_n \gamma_N} \check{u}_m^\mathsf{T} \left(S + \alpha_n{}^2\right) \check{u}_n \\
&= \frac{\mathrm{i}\alpha_m}{\gamma_N} \left\{\delta_{mn} + \left(\alpha_m \alpha_n + \alpha_n{}^2\right) \check{u}_m^\mathsf{T} \check{u}_n\right\} \\
&= \frac{1}{\gamma_N} \left\{\mathrm{i}\alpha_m \delta_{mn} + \mathrm{i}\alpha_m \alpha_n \left(\alpha_m + \alpha_n\right) \check{u}_m^\mathsf{T} \check{u}_n\right\},
\end{aligned}
\tag{S84}
$$

where the third equality follows from Eq. (S70) and the fourth equality follows from Eq. (S75). Therefore,

$$
\begin{aligned}
2\mathrm{i} \sum_n \frac{b_m b_n}{\alpha_m + \alpha_n} &= 2\mathrm{i}\omega_{\mathrm{c}1}{}^4 \left(e_1^\mathsf{T} u_m\right) \sum_n \left\{\frac{\mathrm{i}\alpha_m}{\left(\alpha_m + \alpha_n\right)} \delta_{mn} + \mathrm{i}\alpha_m \alpha_n \check{u}_m^\mathsf{T} \check{u}_n\right\} \left(e_1^\mathsf{T} u_n\right) \\
&= -\omega_{\mathrm{c}1}{}^4 \left(e_1^\mathsf{T} u_m\right) \left\{\left(e_1^\mathsf{T} u_m\right) + \sum_n 2\alpha_m \alpha_n \left(\check{u}_m^\mathsf{T} \check{u}_n\right) \left(e_1^\mathsf{T} u_n\right)\right\}.
\end{aligned}
\tag{S85}
$$

Here the sum in the last part vanishes as follows

$$
\begin{aligned}
\sum_n 2\alpha_m \alpha_n \left(\check{u}_m^\mathsf{T} \check{u}_n\right) \left(e_1^\mathsf{T} u_n\right) &= 2 \sum_n \alpha_m \alpha_n \left(e_1^\mathsf{T} u_n\right) \left(\check{u}_n^\mathsf{T} \check{u}_m\right) \\
&= -2\mathrm{i} \sum_n \alpha_m e_1^\mathsf{T} u_n v_n^\mathsf{T} \begin{pmatrix} 0 \\ \check{u}_m \end{pmatrix} \\
&= -2\mathrm{i}\alpha_m e_1^\mathsf{T} \begin{pmatrix} 0 \\ \check{u}_m \end{pmatrix} \\
&= 0,
\end{aligned}
\tag{S86}
$$

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where Eq. (S72) has been used in the second line and the completeness relation ($\sum_n \boldsymbol{u}_n \boldsymbol{v}_n^\mathsf{T} = 1$) of the eigenvectors has been used in the third line. Therefore,

$$2\mathrm{i} \sum_n \frac{b_m b_n}{\alpha_m + \alpha_n} = -\omega_{\mathrm{c}1}{}^4 \left(\boldsymbol{e}_1^\mathsf{T} \boldsymbol{u}_m\right) \left(\boldsymbol{u}_m^\mathsf{T} \boldsymbol{e}_1\right) \tag{S87}$$

This is equal to Eq. (S81) because $\boldsymbol{v}_m^\mathsf{T} \boldsymbol{e}_{N+1} = \mathrm{i}\alpha_m \boldsymbol{u}_m^\mathsf{T} \boldsymbol{e}_1$ due to Eqs. (S70) and (S71).

With $\{b_m\}$ given by Eq. (S82), the random force $f_1(t)$ of Eq. (S80) can be written as

$$f_1(t) = \sum_m \left\{ \omega_{\mathrm{c}1}{}^2 \left(\boldsymbol{e}_1^\mathsf{T} \boldsymbol{u}_m\right) y_m(0) \exp\left(\mathrm{i}\alpha_m t\right) + \int_0^t b_m \exp\left(\mathrm{i}\alpha_m(t - t')\right) \frac{\xi_{\mathrm{w}}(t')}{\sqrt{\gamma_N}} \mathrm{d}t' \right\},$$

$$= \sum_m \omega_{\mathrm{c}1}{}^2 \left(\boldsymbol{e}_1^\mathsf{T} \boldsymbol{u}_m\right) y_m(0) \exp\left(\mathrm{i}\alpha_m t\right) + \int_0^t g(t - t') \frac{\xi_{\mathrm{w}}(t')}{\sqrt{\gamma_N}} \mathrm{d}t', \tag{S88}$$

due to the definition of $g(\tau) = \Theta(\tau) \sum_m b_m \exp(\mathrm{i}\alpha_m \tau)$ (Eq. (12)). The definition of ξ_m gives

$$\xi_m(t) = \int_0^{+\infty} k_m(t - t') f_1(t') \mathrm{d}t'$$

$$= \omega_{\mathrm{c}1}{}^2 \left(\boldsymbol{e}_1^\mathsf{T} \boldsymbol{u}_m\right) y_m(0) \exp\left(\mathrm{i}\alpha_m t\right) + b_m \int_0^t \exp\left(\mathrm{i}\alpha_m(t - t')\right) \frac{\xi_{\mathrm{w}}(t')}{\sqrt{\gamma_N}} \mathrm{d}t', \tag{S89}$$

where Eq. (S2) has been used for the first term and, for the second term,

$$\int_0^{+\infty} \mathrm{d}t' k_m(t - t') \int_0^{t'} \mathrm{d}t'' g(t' - t'') \frac{\xi_{\mathrm{w}}(t'')}{\sqrt{\gamma_N}}$$

$$= \int_0^{+\infty} \mathrm{d}t'' \int_{t''}^{+\infty} \mathrm{d}t' k_m(t - t') g(t' - t'') \frac{\xi_{\mathrm{w}}(t'')}{\sqrt{\gamma_N}}$$

$$= \frac{1}{(2\pi)^2} \int_0^{+\infty} \mathrm{d}t'' \int_{-\infty}^{+\infty} \mathrm{d}t' \int_{-\infty}^{+\infty} \mathrm{d}\omega_1 \tilde{k}_m(\omega_1) \exp(\mathrm{i}\omega_1(t - t')) \int_{-\infty}^{+\infty} \mathrm{d}\omega_2 \tilde{g}(\omega_2) \exp(\mathrm{i}\omega_2(t' - t'')) \frac{\xi_{\mathrm{w}}(t'')}{\sqrt{\gamma_N}}$$

$$= \frac{1}{2\pi} \int_0^{+\infty} \mathrm{d}t'' \int_{-\infty}^{+\infty} \mathrm{d}\omega_1 \int_{-\infty}^{+\infty} \mathrm{d}\omega_2 \tilde{k}_m(\omega_1) \tilde{g}(\omega_2) \delta(-\omega_1 + \omega_2) \exp(\mathrm{i}\omega_1 t - \mathrm{i}\omega_2 t'') \frac{\xi_{\mathrm{w}}(t'')}{\sqrt{\gamma_N}}$$

$$= \frac{1}{2\pi} \int_0^{+\infty} \mathrm{d}t'' \int_{-\infty}^{+\infty} \mathrm{d}\omega \frac{-\mathrm{i}b_m}{\omega - \alpha_m} \exp(\mathrm{i}\omega(t - t'')) \frac{\xi_{\mathrm{w}}(t'')}{\sqrt{\gamma_N}}$$

$$= \int_0^t \mathrm{d}t'' b_m \exp(\mathrm{i}\alpha_m(t - t'')) \frac{\xi_{\mathrm{w}}(t'')}{\sqrt{\gamma_N}}, \tag{S90}$$

where in the third line the integration range of t' has been extended because $g(\tau) = \Theta(\tau) \sum_m b_m \exp(\mathrm{i}\alpha_m \tau)$ is zero for $\tau < 0$.

Finally, the EXEM introduced in the present paper becomes, from Eq. (84),

$$
\begin{aligned}
X_m &= -\int_0^t \mathrm{i} c_m \alpha_m \exp\left(\mathrm{i}\alpha_m(t-t')\right) Q(t')\mathrm{d}t' + \omega_{\mathrm{c}1}{}^2 \left(\boldsymbol{e}_1^{\mathsf{T}}\boldsymbol{u}_m\right) y_m(0)\exp\left(\mathrm{i}\alpha_m t\right) \\
&\quad + b_m \int_0^t \exp\left(\mathrm{i}\alpha_m(t-t')\right) \frac{\xi_{\mathrm{w}}(t')}{\sqrt{\gamma_N}}\mathrm{d}t' \\
&= \int_0^t \omega_{\mathrm{c}1}{}^4 \left(\boldsymbol{e}_1^{\mathsf{T}}\boldsymbol{u}_m\right)\left(\boldsymbol{v}_m^{\mathsf{T}}\boldsymbol{e}_{N+1}\right)\exp\left(\mathrm{i}\alpha_m(t-t')\right) Q(t')\mathrm{d}t' + \omega_{\mathrm{c}1}{}^2 \left(\boldsymbol{e}_1^{\mathsf{T}}\boldsymbol{u}_m\right) y_m(0)\exp\left(\mathrm{i}\alpha_m t\right) \\
&\quad + \omega_{\mathrm{c}1}{}^2 \left(\boldsymbol{e}_1^{\mathsf{T}}\boldsymbol{u}_m\right)\int_0^t \left(\boldsymbol{v}_m^{\mathsf{T}}\boldsymbol{e}_{2N}\right)\exp\left(\mathrm{i}\alpha_m(t-t')\right)\xi_{\mathrm{w}}(t')\mathrm{d}t' \\
&= \omega_{\mathrm{c}1}{}^2 \left(\boldsymbol{e}_1^{\mathsf{T}}\boldsymbol{u}_m\right) y_m(t), \tag{S91}
\end{aligned}
$$

where Eq. (S82) for b_m, Eq. (S81) for c_m, and Eq. (S78) for y_m have been used. This shows that the EXEM is equal to the dynamical coordinate y_m multiplied by a constant. Since y_m is given by a linear transformation from the bath coordinates $(\boldsymbol{Q}_{\mathrm{b}}, \dot{\boldsymbol{Q}}_{\mathrm{b}})$, it is concluded that the bath coordinates introduced by Adelman for the MTGLE and the EXEM in the present study are related through a linear transformation.