



PAPER

On the derivation of a Nonlinear Generalized Langevin Equation

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Abstract

We recast the Zwanzig's derivation of a nonlinear generalized Langevin equation (GLE) for a heavy particle interacting with a heat bath in a more general framework. We show that it is necessary to readjust the Zwanzig's definitions of the kernel matrix and noise vector in the GLE in order to recover the correct definition of fluctuation-dissipation theorem and to be able performing consistently the continuum limit. As shown by Zwanzig, the nonlinear feature of the resulting GLE is due to the nonlinear dependence of the equilibrium map by the heavy particle variables. Such an equilibrium map represents the global equilibrium configuration of the heat bath particles for a fixed (instantaneous) configuration of the system. Following the same derivation of the GLE, we show that a deeper investigation of the equilibrium map, considered in the Zwanzig's Hamiltonian, is necessary. Moreover, we discuss how to get an equilibrium map given a general interaction potential. Finally, we provide a renormalization procedure which allows to divide the dependence of the equilibrium map by coupling coefficient from the dependence by the system variables yielding a more rigorous mathematical structure of the nonlinear GLE.

1. Introduction**1.1. State-of-art**

The generalized Langevin equation (GLE) is an important mathematical tool for investigating a wide class of both quantum and classical diffusive phenomena [1] and it is an extension of the Langevin equation [2]. The major advantage of GLE-based approaches is the reduction of the number of degrees of freedom which describe the system-environment interactions. For example, in case of a particle moving in a given environment, instead of solving the Newton's equations of motions for the system and for each particles of the environment, one can just focus on the velocity vector, U , of the diffusing particle, that is, the GLE reads:

$$\frac{dU}{dt}(t) = - \int_0^t \gamma(t - \tau) U(\tau) d\tau + F(t), \quad (1)$$

where $\gamma(t - \tau)$ is the friction function and, finally, $F(t)$ is a zero mean, stochastic, Gaussian colored noise and they are related through the fluctuation-dissipation relation (FDR) [3]:

$$\langle F(t)F(\tau) \rangle = k_B T \gamma(t - \tau), \quad (2)$$

where k_B is the Boltzmann constant and T the temperature. Despite the drastic reduction of the degrees of freedom, the GLE has shown to provide a reliable description of the physical diffusive processes. In fact, it has been employed in many fields of research, for instance, in Biophysics, for studying anomalous diffusion of proteins in lipid membrane [4–7], in Hydrodynamics, for studying how probe particles diffuse in viscoelastic materials [8–12], in Quantum Mechanics, for explaining the dissipation in quantum system on a more fundamental and general level [13–17] as well as in case of specific system such as the Caldeira-Leggett model [18, 19] and, in Chemistry, as a diffusion model for the chemical reactions [20, 21]. Nevertheless, the accuracy of the physical description provided by the GLE strictly depends on how one models the parameters and the time-dependence of the friction function. Its parametrization, in fact, is an open field of research which involves both

mathematical and physical studies [22–28] which provide numerically or analytically the correct description of the kernel function depending on physical system that one considers.

By definition, the diffusion is a dynamical process at non-equilibrium since the energy is not constant in time but it varies over time due to the interaction with the environment, thereby, it cannot be described through the Hamiltonian formalism. In fact, it is not possible to obtain a (generalized) Langevin equation from the Hamilton's equations to any Hamiltonian when the time dependence is not explicit. However, if one introduces a further Hamiltonian containing all the microscopic details of the environment explicating the interactions with the system, the energy of both the system and the environment can vary but their sum will be unchanged. The first attempts to derive a GLE adopting this approach have been done by Mori [29] and Ford *et al* [30] in the context of linear system-environment interactions and, then, by Zwanzig [31] extending the previous works to the case of non-linear interactions.

The nonlinear GLE obtained by Zwanzig in [31] is derived from a Hamiltonian function where the particles composing the environment oscillate harmonically around their global equilibrium configuration, denoted with the map a , which depends on the instantaneous system configuration. The equilibrium map, a , plays a fundamental role in the derivation since it contains the information about the coupling between the system and the environment and the (non) linearity feature of the GLE is determined by the (non) linear dependence of this map by the system variable.

As shown by Zwanzig [31] through a purely theoretical derivation, even in presence of a non-linear coupling, one can still provide a mathematical definition for the friction and noise terms and, thereby, the fluctuation-dissipation theorem [32] can be formulated. Unfortunately, as we will show in the upcoming section, by strictly applying the theoretical definitions derived in [31], one notices that the friction function is ill-defined since it does not contain the information about the coupling and this leads to obtain a tautology on the FDR and to an unbounded friction function in the continuum limit [31].

The first aim of this paper is to formalize and adapt the Zwanzig's derivation in order to provide a preciser definition of kernel and noise terms which is consistent with the continuum limit. The second aim is to underline the importance of the equilibrium map in the context of the GLE and to study its mathematical properties.

1.2. Introduction to the problem

In this section, we revisit and formalize the theoretical derivation of a GLE introduced by Zwanzig [31] putting in evidence what are the main inconsistencies and giving an insight about how to overcome them.

The physical system that Zwanzig considered is composed by a subsystem that we call *heavy particle* described by the variable $X := (Q, P)$ where Q is the generalized coordinate and P the conjugated momentum and a *heat bath* which is a set of N particles whose state is described by the collective variables $Y := (q, p)$ with the same meaning as before.

The whole Hamiltonian of this system is given by Zwanzig is [31]

$$H(X, Y) = H_s(X) + \frac{1}{2}[Y - a(X)]^T K [Y - a(X)], \quad (3)$$

where the second term represents the interaction Hamiltonian since it involves the variables of both heavy particle and heat bath through the $a(X)$ vector. In fact, in order to switch off the interactions, we have to impose somehow $a(X) = 0$ for each $X \neq 0$. Finally, the matrix K is a symmetric constant matrix.

The equation of motion for the heat bath and heavy particle variables are, respectively:

$$\begin{aligned} \frac{dY}{dt}(t) &= BK [Y(t) - a(X(t))], \\ \frac{dX}{dt}(t) &= V(X(t)) - A W(X(t)) K [Y(t) - a(X(t))] \end{aligned} \quad (4)$$

where A and B are two constant antisymmetric matrix, i.e.:

$$A = \begin{pmatrix} 0 & 1_{h,p} \\ -1_{h,p} & 0 \end{pmatrix}, \quad B = \begin{pmatrix} 0 & 1_{h,b} \\ -1_{h,b} & 0 \end{pmatrix},$$

such that the ranks of A and B are, in general, different and where $W(x) = \nabla_x a^T(X)$, $V(X) = A \nabla_x H_s(X)$.

The heat-bath equation in (4) can be formally solved and we have:

$$Y(t) - a(X(t)) = \exp(BK t)[Y(0) - a(X(0))] + \int_0^t \exp(BK \tau) \frac{d}{d\tau} a(X(t - \tau)) d\tau. \quad (5)$$

The time-derivative of the $a(X)$ vector can be rewritten in terms of the W matrix, i.e.

$$\frac{d}{d\tau}a(X(t-\tau)) = -W^T(X(t-\tau))\dot{X}(t-\tau). \quad (6)$$

By replacing equation (5) into the first equation in (4) and by making use of equation (6), one gets

$$\begin{aligned} \frac{dX}{dt}(t) = & V(X(t)) + \int_0^t A W(X(t)) K \exp(BK \tau) W^T(X(t-\tau)) \dot{X}(t-\tau) d\tau \\ & - A W(X(t)) K \exp(BK t)[Y(0) - a(X(0))]. \end{aligned} \quad (7)$$

At this point, Zwanzig defined the noise vector

$$F(t) = -K \exp(BK t)[Y(0) - a(X(0))], \quad (8)$$

and the kernel matrix

$$L(t) = K \exp(BK t), \quad (9)$$

and, introducing the canonical partition function

$$\rho(p, q|Q) := \exp(-\beta H_b(p, q, Q)), \quad (10)$$

he formulates the fluctuation-dissipation relation (FDR)

$$\langle F(t) \cdot F(\tau) \rangle_\rho = k_B T \zeta(t-\tau), \quad (11)$$

where k_B and T , respectively, are the Boltzmann constant and the temperature whereas $\zeta := \text{Tr}[L]$ and Tr denotes the trace of a matrix.

1.2.1. Criticism

We now explain our main criticism to the Zwanzig's derivation showing that the definitions (8) and (9) provided by Zwanzig yield a FDR which is inconsistent with the standard definition [3]. The main reason of this inconsistency can be better explained by means of the following argument. In writing the Hamiltonian for two interacting systems, all the information about the nature of the interaction is contained in the potential function which gives a fully microscopic and detailed description of system. Conversely, in the (generalized) Langevin equation, one just takes into account a coarse-grained representation of these interactions encoded into two terms: the stochastic noise vector and the friction matrix or function. The role of the FDR is somehow to balance the energy dissipated by the friction with that supplied by the random noise. Consequently, the friction and noise term have to contain the information about the strength of the interactions. For example, in the easiest case of a standard Langevin equation (LE), i.e., $\dot{U}(t) = -\gamma U(t) + F(t)$, the second term on the right-hand side can be interpreted as a convolution between a kernel function $\zeta(t-\tau) \sim \gamma \delta(t-\tau)$ (δ is the Dirac delta distribution) and the velocity vector of the heavy particle, $U(t)$. In this case, it is also evident that the strength of the interaction is represented by the friction constant γ which is a coarse-grained description of the heavy particle-environment interactions. This feature must hold also in case of a GLE, that is, we expect that, in the Zwanzig's derivation, all the microscopic information contained in the potential function are condensed within the friction matrix L .

By inspection, we immediately realize that L and F , respectively, in equation (8) and (9) do not show this feature. In fact, (up to two constants k_B and T) L depends on B and K . Now, B is a constant antisymmetric matrix whereas K is the matrix associated to the oscillation frequencies of the heat bath particles when they do not interact with the heavy one¹.

Therefore, the FDR formulated by Zwanzig in (11) does not provide any balance.

Based on this observation, we now explicitly show that, introducing the same Hamiltonian considered by Zwanzig, that is: [31]

$$H = \frac{P^2}{2} + V(Q) + \sum_{j=1}^N \left[\frac{p_j^2}{2} + \frac{\omega_j}{2} \left(q_j - \frac{\gamma_j}{\omega_j^2} Q \right)^2 \right], \quad (12)$$

where we recall that capital letters represent the heavy particle variables.

By comparing (12) and (3), we note that the first two terms in the Hamiltonian above correspond to $H_s(X)$ whereas the terms in the square brackets correspond to $H_b(X, Y)$. Therefore, the Hamiltonian (12) can be deduced from that in equation (3) making explicit in the latter the position and momentum variables, i.e.:

¹ We can convince about it just observing the Hamiltonian $H_b(X, Y)$ in equation (3). By switching the heat bath-heavy particles interactions off means that $a(X) = 0$ for any $X \neq 0$ and this leads to $H_b(X, Y) = H_s(X) + Y^T K Y / 2$. This is the Hamiltonian of two non-interacting subsystems where the latter is nothing but a set of (non-interacting) harmonic oscillators.

$$\begin{aligned}
 H(P, p, Q, q) = & H_s(P, Q) + \frac{1}{2}[p - a_p(P, Q)]^T K_{pp}[p - a_p(P, Q)] \\
 & + \frac{1}{2}[q - a_q(P, Q)]^T K_{qq}[q - a_q(P, Q)] \\
 & + [p - a_p(P, Q)]^T K_{pq}[q - a_q(P, Q)].
 \end{aligned} \tag{13}$$

Now, by imposing $K_{pp} = 1$ and $a_p(P, Q) = 0, K_{pq} = K_{qp} = 0$ and setting $K_{qq} := \text{diag} \{ \omega_1^2, \omega_2^2, \dots, \omega_N^2 \}$ and $a_q^i(P, Q) \equiv a_q^i(Q) = \frac{\gamma_i}{\omega_i^2} Q$, equation (13) reduces to (12). We note that $\{ \omega_i^2 \}$ is the set of *free oscillation frequencies* of the heat bath particles whereas $\{ \gamma_i \}_{i=1}^N$ is the set of coupling coefficients which determine the strength of the heat bath-heavy particle interactions. Thus, a specific interaction can be switched off just imposing $\gamma_i \rightarrow 0$ which implies $a_q^i(Q) = 0$.

By specializing definitions (8) and (9) to this example, a cumbersome but straightforward calculation leads to the noise term

$$E_q(t) = -K_{qq} \cos(K_{qq}^{1/2} t) + K_{qq}^{1/2} \sin(K_{qq}^{1/2} t) p, \tag{14}$$

whereas for the kernel matrix, we have

$$L_{qq}(t) = K_{qq} \cos(K_{qq}^{1/2} t). \tag{15}$$

As already stressed, the friction matrix (15) does not depend on the coupling coefficients, γ_i and, thereby, the FDR is ill-defined as we will see in a moment. In fact, by plugging equations (14) and (15) into the definition (11) for the FDR, one gets:

$$\langle E_q(t) \cdot E_q(\tau) \rangle = k_B T \text{Tr} [K_{qq} \cos(K_{qq}^{1/2} (t - \tau))] \equiv k_B T \text{Tr} [L_{qq}(t - \tau)] = k_B T \sum_{i=1}^N \omega_i^2 \cos(\omega_i(t - \tau)). \tag{16}$$

It is evident that the friction function on the last right-hand side, that is:

$$\xi(t - \tau) := \sum_{i=1}^N \omega_i^2 \cos(\omega_i(t - \tau)), \tag{17}$$

just depends on the oscillation frequencies $\{ \omega_i \}_{i=1}^N$, therefore, the obtained FDR (16) does not recover the usual definition.

Finally, we stress that, adopting the same example, Zwanzig writes a different friction function (which is also the correct one) and it reads (see equation (23) in [31]):

$$\zeta_Z(t - \tau) = \sum_{i=1}^N \left(\frac{\gamma_i}{\omega_i} \right)^2 \cos(\omega_i(t - \tau)). \tag{18}$$

This suggests that he maybe absorbed, ad hoc, the γ_i -coefficients within ζ .

Based on that, we conclude that the definition (18) should be adjusted in order to obtain the correct ζ function without any ad hoc-adjustment and this is the principal aim of the paper.

1.3. Notation

In this section, we formalize the derivation introduced in the previous section.

We consider a Hamiltonian function describing a classical physical system composed by a heat bath (\mathcal{P}) interacting with a heavy particle (\mathcal{B}). The heat bath consists of N particles with mass $m = 1$ smaller than the heavier particle mass M .

We assume that the system is embedded in the three dimensional physical space \mathbb{R}^3 so that the *phase space*, denoted by Γ , is a subset of $\mathbb{R}^{3N} \times \mathbb{R}^{3N} \times \mathbb{R}^3 \times \mathbb{R}^3$. Therefore, the heat bath conjugated coordinates are:

$$I^\alpha := \sum_{i=1}^N p_i^\alpha \mathbf{e}_i \in \mathbb{R}^N, \quad Q^\alpha := \sum_{i=1}^N q_i^\alpha \mathbf{e}_i \in \mathbb{R}^N, \quad \alpha = x, y, z, \tag{19}$$

where we have introduced the canonical base $\{ \mathbf{e}_i \}_{i=1}^N$ of \mathbb{R}^N . The heavy particle Hamiltonian conjugated coordinates are

$$\mathbf{X} := \sum_{\alpha=1}^3 X^\alpha \mathbf{e}_\alpha \in \mathbb{R}^3, \quad \mathbf{P} := \sum_{\alpha=1}^3 P^\alpha \mathbf{e}_\alpha \in \mathbb{R}^3.$$

The system Hamiltonian function, $\mathcal{H}: \Gamma \rightarrow \mathbb{R}$ can be split into three terms:

$$\mathcal{H}(\mathbf{P}, \mathbf{I}, \mathbf{X}, \mathbf{Q}) := H_{\mathcal{B}}(\mathbf{P}, \mathbf{X}) + H_{\mathcal{P}}(\mathbf{I}, \mathbf{Q}) + H_{INT}(\mathbf{Q}, \mathbf{X}),$$

where, as usual, we have:

$$\begin{aligned} H_{\mathcal{H}}(\mathbf{P}, \mathbf{X}) &= \frac{\|\mathbf{P}\|_{\mathbb{R}^3}^2}{2M} + V(\mathbf{X}), \\ H_{\mathcal{H}}(\mathbf{I}, \mathbf{Q}) &= \frac{\|\mathbf{I}\|_{\mathbb{R}^{3N}}^2}{2} = \sum_{i=1}^N \frac{\|\mathbf{p}_i\|_{\mathbb{R}^3}^2}{2}, \end{aligned} \quad (20)$$

where the potential function $V: \mathbb{R}^3 \rightarrow \mathbb{R}$ describes any *external* force which just acts on the heavy particle and it could have a generic dependence on \mathbf{X} .

The interaction term is given by:

$$H_{INT}(\mathbf{Q}, \mathbf{X}) := \frac{1}{2} \sum_{i,j=1}^N \langle \mathbf{q}_i - \mathbf{a}_i(\mathbf{X}), \mathbf{K}_{ij}(\mathbf{q}_j - \mathbf{a}_j(\mathbf{X})) \rangle_{\mathbb{R}^3}. \quad (21)$$

Thus, the interactions are described by the so-called *interaction matrix* which is a linear application $\mathbf{K}: \mathbb{R}^{3N} \rightarrow \mathbb{R}^{3N}$ acting on the heat bath degrees of freedom. Note that each \mathbf{K}_{ij} is a three dimensional matrix since also the spatial degrees of freedom are included.

The meaning of the interaction described by H_{INT} is to assume a harmonic approximation around the equilibrium positions $\mathbf{Q}^* := \mathbf{a}(\mathbf{X})$ of the heat bath system, where $\mathbf{a}: \mathbb{R}^3 \rightarrow \mathbb{R}^{3N}$ provides the equilibrium configuration of all the heat bath particles for a given $\mathbf{X} \in \mathbb{R}^3$.

The system of Hamilton's equations, then, reads:

$$\begin{aligned} \dot{\mathbf{X}}(t) &= \frac{\mathbf{P}(t)}{M}, \\ \dot{\mathbf{P}}(t) &= \nabla_{\mathbf{X}(t)} \mathbf{a}^T(\mathbf{X}(t)) \mathbf{K}(\mathbf{Q}(t) - \mathbf{a}(\mathbf{X}(t))) - \nabla_{\mathbf{X}(t)} V(\mathbf{X}(t)), \\ \dot{\mathbf{Q}}(t) &= \mathbf{I}(t), \\ \dot{\mathbf{I}}(t) &= -\mathbf{K}(\mathbf{Q}(t) - \mathbf{a}(\mathbf{X}(t))), \end{aligned} \quad (22)$$

where $\nabla_{\mathbf{X}(t)} \mathbf{a}: \mathbb{R}^3 \rightarrow \mathbb{R}^3 \times \mathbb{R}^{3N}$ and the superscript T denotes the transpose operation.

The system of equations above can be reduced to a GLE for the system position vector $\mathbf{X}(t)$. To do so, one first focuses on the heat bath equations (the last two equations) and, by just having a linear dependence on the \mathbf{Q} -coordinates, one interprets the factor $\mathbf{K}\mathbf{a}(\mathbf{X})$ as an external force. Hence, one exploits the Laplace transform method to obtain a formal solution $\mathbf{Q}^S(t)$ for the heat bath position vector. Actually, the Hamiltonian (21) presents a translational invariance since it only depends on $\mathbf{Q} - \mathbf{a}(\mathbf{X})$. This feature reflects on the formal heat bath solution and, as we will see later, we obtain an explicit mathematical expression of the heat bath solution directly in terms of $\mathbf{Q}^S(t) - \mathbf{a}(\mathbf{X}(t))$. Then, the solution can be plugged into the equation of motion of the heavy particle and the resulting equation explicitly takes the form of a GLE.

We will explicitly employ such a procedure in the next section after having studied more deeply the properties of the equilibrium map $\mathbf{a}(\mathbf{X})$ which plays a fundamental role in the definition of the kernel matrix and noise vector.

We conclude this section, mentioning that the procedure above described, called elimination of the heat bath degrees of freedom, has been already introduced by Kac *et al* [30] first and by Zwanzig [31] later. However, Kac *et al* just considered linear system-heat bath interactions that, in our notation, translates in defining the \mathbf{a} map to be $\mathbf{a}_i(\mathbf{X}) := \mathbf{X}$ and absorbing all the parameters representing the interaction strength within the \mathbf{K} matrix.

Zwanzig extended their system assuming a generic dependency of the \mathbf{a} map on the heavy particle position vector \mathbf{X} and since it could be nonlinear, the elimination procedure led to a nonlinear GLE.

2. Formal derivation of a nonlinear GLE

In this section, inspired by the work of Zwanzig, we will derive a nonlinear GLE and define the kernel matrix and noise term in such a way to be able (i) to recover the standard definition of FDR, (ii) to define a continuum limit without any ulterior adjustments.

In the section 2.1, we show that, by choosing a suitable potential function, one can always recast the starting Hamiltonian into an Hamiltonian of the same form given in equation (3). In section 2.2, we show that, an appropriate manipulation of the equilibrium map allows to naturally define the correct kernel matrix and noise vector.

2.1. General setting: study of the equilibrium map

Let us first clarify the role and the physical meaning of the map $\mathbf{a}_i: \mathbb{R}^3 \rightarrow \mathbb{R}^3$.

First of all, we note that each element of the set $\{\mathbf{a}_i(\mathbf{X})\}_{i=1}^N$ plays the role of an equilibrium configuration of the respective heat bath particle for any fixed $\mathbf{X} \in \mathbb{R}^3$. In fact, one can demonstrate [33] that such equilibrium map can be always derived once that a generic interaction potential is given.

In practice, given any Hamiltonian function of the form

$$\mathcal{H}(\mathbf{P}, \mathbf{I}, \mathbf{X}, \mathbf{Q}) := H_{\mathcal{B}}(\mathbf{P}, \mathbf{X}) + H_{\mathcal{D}}(\mathbf{I}, \mathbf{Q}) + \mathcal{V}(\mathbf{Q}, \mathbf{X}),$$

where $H_{\mathcal{B}}(\mathbf{P}, \mathbf{X})$ and $H_{\mathcal{D}}(\mathbf{I}, \mathbf{Q})$ are given in (20) and $\mathcal{V}(\mathbf{Q}, \mathbf{X})$ is a rather generic interaction term, one can define the global equilibrium map $\mathbf{a}(\mathbf{X}) := \{\mathbf{a}_1(\mathbf{X}), \dots, \mathbf{a}_N(\mathbf{X})\}$, imposing that any component of this map realizes the following requirement

$$\nabla_{\mathbf{q}_i} \mathcal{V}(\mathbf{Q}, \mathbf{X})|_{\mathbf{q}_i=\mathbf{a}_i(\mathbf{X})} = \mathbf{0}, \tag{23}$$

where we remind that $\mathbf{Q} = \{\mathbf{q}_1, \dots, \mathbf{q}_N\}$.

In other words, equation (23) provides an implicit relation between \mathbf{a} and \mathbf{X} . Therefore, the Taylor expansion (harmonic approximation) of the potential with respect to the equilibrium map \mathbf{a} reads:

$$\begin{aligned} \mathcal{V}(\mathbf{Q}, \mathbf{X}) &\approx \mathcal{V}(\mathbf{a}(\mathbf{X}), \mathbf{X}) \\ &+ \frac{1}{2} \sum_{i,j=1}^N \langle \mathbf{q}_i - \mathbf{a}_i(\mathbf{X}), \mathbf{K}_{ij}(\mathbf{q}_j - \mathbf{a}_j(\mathbf{X})) \rangle_{\mathbb{R}^3}, \end{aligned} \tag{24}$$

where $\mathbf{K}_{ij} := \nabla_{\mathbf{q}_i} \nabla_{\mathbf{q}_j} \mathcal{V}(\mathbf{Q}, \mathbf{X})|_{\mathbf{Q}=\mathbf{a}(\mathbf{X})}$ is, by construction, a constant matrix (in \mathbf{q}) which just contains the oscillation frequencies around the equilibrium positions associated to each heat bath particle. The first term in the expansion (24) could, in general, depends on \mathbf{X} and affects the equations of motion, however, if the dependence of the potential function, \mathcal{V} , on the \mathbf{X} - and \mathbf{q} -coordinates of the form $\|\mathbf{X} - \mathbf{q}_i\|$, such a factor is \mathbf{X} and \mathbf{Q} -independent and hence it could be dropped as we will notice in the next example. On the contrary, the second one can be identified with the interaction potential introduced in equation (21). Depending on the mathematical structure of the initial potential \mathcal{V} , the resulting equilibrium map $\mathbf{a}(\mathbf{X})$ can depend on \mathbf{X} in a general fashion.

2.1.1. Examples of a possible equilibrium map.

Let us consider a Lennard-Jones-like interaction potential of the form

$$\mathcal{V}_{LJ}(\mathbf{Q}, \mathbf{X}) := \sum_{i=1}^N 4\eta_i \left[\left(\frac{\sigma_i}{\|\mathbf{X} - \epsilon_i \mathbf{q}_i\|} \right)^{12} - \left(\frac{\sigma_i}{\|\mathbf{X} - \epsilon_i \mathbf{q}_i\|} \right)^6 \right], \tag{25}$$

where the parameters $\sigma_i, \eta_i \in \mathbb{R}$ have the dimension of an energy and a length, respectively; lastly, $\epsilon_i \in \mathbb{R}$ are dimensionless parameters. Note that we do not consider any mutual interaction between the heat bath particles. The requirement (23), namely:

$$\nabla_{\mathbf{q}_i} \mathcal{V}_{LJ}(\mathbf{Q}^*, \mathbf{X}) = \mathbf{0},$$

where $\mathbf{Q}^* = \{\mathbf{q}_1^*, \dots, \mathbf{q}_N^*\}$ is the equilibrium configuration of the heat bath, yields

$$\|\mathbf{X} - \epsilon_i \mathbf{q}_i^*\|^2 = 2^{1/3} \sigma_i^2, \quad \forall i \in [1, N].$$

This set of relations can be geometrically interpreted imagining that any particle, let us say, the i -th one, lies on a point of the sphere $\mathbb{S}_{R_i}^2$ with radius $R_i = 2^{1/6} \sigma_i$. The reason why any point on the sphere is equivalent to each other is due to the fact we do not have any mutual interaction between the heat bath particles.

Therefore, every equilibrium position vector \mathbf{q}_i^* can be related to the heavy particle vector \mathbf{X} , introducing any unit vector $\mathbf{n}(\mathbf{X})$ ($\|\mathbf{n}(\mathbf{X})\|_{\mathbb{R}^3} = 1$) such that

$$\mathbf{q}_i^* := \frac{\mathbf{X}}{\epsilon_i} + \frac{2^{1/6} \sigma_i}{\epsilon_i} \mathbf{n}(\mathbf{X}), \quad \forall i \in [1, N].$$

In general, it could depend on \mathbf{X} , for example, such as $\mathbf{n}(\mathbf{X}) = \mathbf{X}/\|\mathbf{X}\|$. Thus, the equilibrium map may read:

$$\mathbf{a}_i(\mathbf{X}) := \frac{\mathbf{X}}{\epsilon_i} + \frac{2^{1/6} \sigma_i}{\epsilon_i} \mathbf{n}(\mathbf{X}), \quad \forall i \in [1, N]. \tag{26}$$

In so doing, the first and second term in harmonic expansion (24) read

$$\begin{aligned} \mathcal{V}_{LJ}(\mathbf{a}(\mathbf{X}), \mathbf{X}) &= -\sum_{i=1}^N \eta_i, \\ \mathbf{K}_{ii} &:= \text{Hess}[\mathcal{V}_{LJ}]_{ii}(\mathbf{a}(\mathbf{X}), \mathbf{X}) = \left(\frac{2^{2/3} 36 \eta_i \epsilon_i^2}{\sigma_i^2} \right) \mathbf{1}_{\mathbb{R}^3}, \end{aligned} \tag{27}$$

so that $\mathcal{V}_{jL}(\mathbf{a}(\mathbf{X}), \mathbf{X})$ can be dropped since constant and the potential function can be approximated as follows

$$\mathcal{V}_{jL}(\mathbf{Q}, \mathbf{X}) \approx \frac{1}{2} \sum_{i,j=1}^N \langle \mathbf{q}_i - \mathbf{a}_i(\mathbf{X}), \mathbf{K}_{ij}(\mathbf{q}_j - \mathbf{a}_j(\mathbf{X})) \rangle_{\mathbb{R}^3}. \tag{28}$$

Note that the factor in the \mathbf{K}_{ii} matrix of equation (27) has the physical dimension of a frequency, therefore, we define a new set of frequencies

$$\omega_i^2 := \frac{2^{2/3} 36 \eta_i \epsilon_i^2}{\sigma_i^2}, \quad \forall i \in [1, N], \tag{29}$$

which allows us to rewrite the initial set of parameters $(\eta_i, \epsilon_i, \sigma_i)$ in terms of a new one: $(\omega_i, \eta_i, \epsilon_i)$; in particular, the equilibrium map \mathbf{a}_i in equation (26) now reads:

$$\mathbf{a}_i(\mathbf{X}) := \frac{\mathbf{X}}{\epsilon_i} + 12 \frac{\sqrt{\eta_i}}{\omega_i} \mathbf{n}(\mathbf{X}), \quad \forall i \in [1, N]. \tag{30}$$

We note that if $X \in \mathbb{R}$ then we have $n(X) = X/|X|$ and this implies that $n(X) = \text{sgn}(X)$. Therefore, during the system dynamics, that is, along the equation of motion, the function $X = X(t)$ gives rise to a discontinuous equilibrium map since $n(X) = \text{sgn}(X(t))$ can change sign discontinuously in time passing from $+1$ to -1 and viceversa. Hence, the application to a one-dimensional case requires that the potential function is not of the form $V = V(|X - \epsilon_i q_i|)$.

By going back to the three-dimensional case, we observe that the interaction matrix \mathbf{K} does not depend on the parameters η_i and ϵ_i describing the interactions.

We will show that, in order to coherently define a continuum limit of the kernel function appearing in a GLE obtained through the elimination procedure, we have to *normalize* the equilibrium map as we show in the upcoming section.

2.2. Generalization of the equilibrium map

In order to make our derivation as general as possible, we assume to have n heavy particle position-dependent functions, $\{f_I(\mathbf{X})\}_{I=1}^n$, such that each \mathbf{a}_i map can be written as a sum of these functions weighted with different parameters $\eta_{i,I} \in \mathbb{R}$ which, in turn, represent the interaction strength.

Thus, we have:

$$\mathbf{a}_i(\mathbf{X}) := \sum_{I=1}^n \eta_{i,I} f_I(\mathbf{X}), \tag{31}$$

Essentially, the main trick is to separate, in the equilibrium map, the dependence by \mathbf{X} from the dependence by the parameters $\eta_{i,I}$, so that the f_I -functions do not depend on such parameters. Although each function f_I could have a generic \mathbf{X} dependence, without loss of generality, we assume that only the gradient of the first function f_1 equals the identity matrix in \mathbb{R}^3 . These assumptions can be formalized as follows

$$\nabla_{\mathbf{X}} f_1(\mathbf{X}) = \mathbf{1}_{\mathbb{R}^3}, \quad \nabla_{\mathbf{X}} f_I(\mathbf{X}) \neq \mathbf{1}_{\mathbb{R}^3}, \quad \forall I \in [2, N]. \tag{32}$$

The specific case provided in equation (26) can be obtained from the definition (31) posing $n = 2$ and $f_1(\mathbf{X}) := \mathbf{X}$ and $f_2(\mathbf{X}) := \mathbf{X}/\|\mathbf{X}\|$.

Therefore, in this setting, the Hamiltonian function of the system is:

$$\begin{aligned} H_{\mathcal{B}}(\mathbf{P}, \mathbf{X}) &= \frac{\|\mathbf{P}\|_{\mathbb{R}^3}^2}{2M} + V(\mathbf{X}) + \frac{\|\mathbf{I}\|_{\mathbb{R}^{3N}}^2}{2} \\ &+ \frac{1}{2} \sum_{I,J=1}^n \sum_{i,j=1}^N \langle \mathbf{q}_i - \eta_{i,I} f_I(\mathbf{X}), \mathbf{K}_{ij}(\mathbf{q}_j - \eta_{j,I} f_I(\mathbf{X})) \rangle_{\mathbb{R}^3}. \end{aligned} \tag{33}$$

2.3. Alternative derivation

The system of Hamilton's equations of motion (22) based on the Hamiltonian (33) written as a system of second order differential equations reads:

$$\ddot{\mathbf{X}}(t) = \nabla_{\mathbf{X}(t)} \mathbf{a}^T(\mathbf{X}(t)) \mathbf{K}(\mathbf{Q}(t) - \mathbf{a}(\mathbf{X}(t))) - \nabla_{\mathbf{X}(t)} V(\mathbf{X}(t)), \tag{34}$$

$$\ddot{\mathbf{Q}}(t) = -\mathbf{K}(\mathbf{Q}(t) - \mathbf{a}(\mathbf{X}(t))). \tag{35}$$

We note that equation (35) is linear in the heat bath-variables \mathbf{Q} , this allows us to apply the Laplace transform method [24–26]. Hence, the Laplace transform of equation (35) is

$$\tilde{\mathbf{Q}}(s) = (\mathbf{K} + s^2 \mathbf{1})^{-1} [s \mathbf{Q}(0) + \dot{\mathbf{Q}}(0) + \mathbf{K} \mathbf{a}(\tilde{\mathbf{X}}(s))]. \tag{36}$$

Then, applying the inverse Laplace transform we get:

$$\mathbf{Q}(t) = \cos(\mathbf{K}^{1/2}t) \mathbf{Q}(0) + \mathbf{K}^{-1/2} \sin(\mathbf{K}^{1/2}t) \dot{\mathbf{Q}}(0) + \int_0^t \mathbf{K}^{1/2} \sin(\mathbf{K}^{1/2}(t - \tau)) \mathbf{a}(\mathbf{X}(\tau)) d\tau, \quad (37)$$

where the matrix-valued function is defined through the series representation:

$$\cos(\mathbf{K}^{1/2}t) = \sum_{n=0}^{\infty} \frac{(-1)^n}{(2n)!} \mathbf{K}^n t^{2n}, \quad \sin(\mathbf{K}^{1/2}t) = \sum_{n=0}^{\infty} \frac{(-1)^n}{(2n+1)!} \mathbf{K}^n t^{2n+1}.$$

In order to recover a GLE, in the convolution on the right-hand side the time derivative of the heavy particle variable has to appear. Therefore, we notice that the kernel matrix $\mathbf{K}^{1/2} \sin(\mathbf{K}^{1/2}(t - \tau))$ can be written as the derivative of $\cos(\mathbf{K}^{1/2}(t - \tau))$ and an integration by parts leads to:

$$\begin{aligned} \mathbf{Q}(t) &= \cos(\mathbf{K}^{1/2}t) \mathbf{Q}(0) + \mathbf{K}^{-1/2} \sin(\mathbf{K}^{1/2}t) \dot{\mathbf{Q}}(0) + \mathbf{a}(\mathbf{X}(t)) - \cos(\mathbf{K}^{1/2}t) \mathbf{a}(\mathbf{X}(0)) \\ &\quad - \int_0^t \cos(\mathbf{K}^{1/2}(t - \tau)) \nabla_{\mathbf{X}(\tau)} \mathbf{a}(\mathbf{X}(\tau)) \dot{\mathbf{X}}(\tau) d\tau, \end{aligned} \quad (38)$$

where $\dot{\mathbf{a}}(\mathbf{X}(\tau)) = \nabla_{\mathbf{X}(\tau)} \mathbf{a}(\mathbf{X}(\tau)) \dot{\mathbf{X}}(\tau)$.

Now, we note that the heat bath variable enter equation (34) under a specific combination, i.e., $\mathbf{K}[\mathbf{Q}(t) - \mathbf{a}(\mathbf{X}(t))]$, but this relation can be obtained in equation (38) that is:

$$\mathbf{K}[\mathbf{Q}(t) - \mathbf{a}(\mathbf{X}(t))] = G_{\mathbf{K}}(t) \dot{\mathbf{Q}}(0) + H_{\mathbf{K}}(t)(\mathbf{Q}(0) - \mathbf{a}(\mathbf{X}(0))) - \int_0^t [S_{\mathbf{K}}(t - \tau) \nabla_{\mathbf{X}(\tau)} \mathbf{a}(\mathbf{X}(\tau))] \dot{\mathbf{X}}(\tau) d\tau, \quad (39)$$

$$H_{\mathbf{K}}(t) := \mathbf{K} \cos(\mathbf{K}^{1/2}t), \quad G_{\mathbf{K}}(t) := \sin(\mathbf{K}^{1/2}t), \quad S_{\mathbf{K}}(t) := \mathbf{K} \cos(\mathbf{K}^{1/2}t), \quad (40)$$

Finally, we introduce a further canonical basis $\{\mathbf{e}_I\}_{I=1}^n$ of \mathbb{R}^n which allows us to define, respectively, the parameter matrix and the equilibrium map vector:

$$\mathbf{\Lambda} := \sum_{i=1}^N \sum_{j=1}^n \eta_{i,j} \mathbf{e}_i \otimes \mathbf{e}_j, \quad \mathbf{g}(\mathbf{X}) := \sum_{I=1}^n f_I(\mathbf{X}) \mathbf{e}_I, \quad (41)$$

such that

$$\nabla_{\mathbf{X}} \mathbf{a}(\mathbf{X}) = \mathbf{\Lambda} \nabla_{\mathbf{X}} \mathbf{g}(\mathbf{X}). \quad (42)$$

Hence, we plug equation (39) into equation (34) and we obtain:

$$\begin{aligned} M\ddot{\mathbf{X}}(t) &= \nabla_{\mathbf{X}(t)} \mathbf{g}^T(\mathbf{X}(t)) \mathbf{\Lambda}^T (G_{\mathbf{K}}(t) \mathbf{I}(0) + H_{\mathbf{K}}(t) [\mathbf{Q}(0) - \mathbf{a}(\mathbf{X}(0))]) \\ &\quad - \int_0^t \nabla_{\mathbf{X}(t)} \mathbf{g}^T(\mathbf{X}(t)) (\mathbf{\Lambda}^T S_{\mathbf{K}}(t - \tau) \mathbf{\Lambda}) \nabla_{\mathbf{X}(\tau)} \mathbf{g}(\mathbf{X}(\tau)) \dot{\mathbf{X}}(\tau) d\tau - \nabla_{\mathbf{X}(t)} V(\mathbf{X}(t)). \end{aligned} \quad (43)$$

In order to recover the common structure of a GLE, we note that the mathematical structure of the equation obtained above allows to define the kernel matrix and the noise term as follows:

$$\begin{aligned} \mathbf{L}(t - \tau) &:= \mathbf{\Lambda}^T S_{\mathbf{K}}(t - \tau) \mathbf{\Lambda}, \\ \mathbf{F}(t) &:= \mathbf{\Lambda}^T (G_{\mathbf{K}}(t) \mathbf{I}(0) + H_{\mathbf{K}}(t) [\mathbf{Q}(0) - \mathbf{a}(\mathbf{X}(0))]), \end{aligned} \quad (44)$$

We wish to stress a significant difference between the current definitions of noise vector and kernel matrix in equation (44) and the previous ones in equation (8) and (9). In the latter, the noise and kernel terms do not contain any information about the strength of the interactions represented by the coefficients $\gamma_{i,I}$ and all the parameters $\gamma_{i,I}$ are contained in the equilibrium map. In the former case, the parameters $\eta_{i,I}$ have been completely absorbed into the noise term and kernel matrix.

Therefore, we can recast equation (43) into a nonlinear GLE:

$$M\ddot{\mathbf{X}}(t) = \nabla_{\mathbf{X}(t)} \mathbf{g}^T(\mathbf{X}(t)) \mathbf{F}(t) - \int_0^t \nabla_{\mathbf{X}(t)} \mathbf{g}^T(\mathbf{X}(t)) \mathbf{L}(t - \tau) \nabla_{\mathbf{X}(\tau)} \mathbf{g}(\mathbf{X}(\tau)) \dot{\mathbf{X}}(\tau) d\tau - \nabla_{\mathbf{X}(t)} V(\mathbf{X}(t)). \quad (45)$$

An important step forward in the investigation of stochastic behaviors arising from interacting Hamiltonian systems would be made if we could solve equation (45) which is a nonlinear stochastic differential equation.

Two different approaches could be employed. From one side, one can attempt a semi-analytical approach exploiting the same method introduced in [34]. The authors showed that, even though we have a nonlinear differential equation, but we explicit know the time-dependence of the functions entering the equation², we can still apply the Laplace transform method to equation (45). However, this approach cannot always provide an exact solution but a series representation of the solution is possible. So far, this approach has been applied just for investigating nonlinear differential equations which are not stochastic but it could be extended to address stochastic differential equation. A more detailed study will be done in a future paper.

² In our case, this translates in choosing a specific potential function so that the equilibrium map is well-defined.

Another possible approach is the numerical one. In particular, under suitable condition on the kernel matrix, $L(t - \tau)$, a GLE can be converted into a stochastic Volterra equation where several numerical integration schemes have been developed such as the Euler and Milstein scheme [35].

We now show that adopting definitions (44), one naturally recover the FDR.

Hence, let us consider the heat bath Hamiltonian from equation (33), namely:

$$H_{\mathcal{D}}(\mathbf{I}, \mathbf{Q}) = \frac{\|\mathbf{I}\|_{\mathbb{R}^{3N}}^2}{2} + \frac{1}{2} \langle \mathbf{Q} - \mathbf{a}(\mathbf{X}), \mathbf{K}(\mathbf{Q} - \mathbf{a}(\mathbf{X})) \rangle_{\mathbb{R}^{3N}}. \tag{46}$$

The canonical probability density is given by:

$$\rho(\mathbf{X}(0) | \mathbf{Q}(0), \mathbf{I}(0)) := \frac{1}{Z} e^{-\frac{\beta}{2} \langle \mathbf{I}, \mathbf{I} \rangle_{\mathbb{R}^{3N}}} \cdot e^{-\frac{\beta}{2} \langle \mathbf{Q}(0) - \mathbf{a}(\mathbf{X}(0)), \mathbf{K}(\mathbf{Q}(0) - \mathbf{a}(\mathbf{X}(0))) \rangle_{\mathbb{R}^{3N}}}, \tag{47}$$

where Z is the canonical partition function

$$Z := \int e^{-\beta [\mathcal{H}_{\mathcal{D}}(\mathbf{I}(0), \mathbf{Q}(0), \mathbf{X}(0))]} d\mathbf{Q}(0) d\mathbf{I}(0), \tag{48}$$

and where $\beta^{-1} = k_B T$.

Let us note that, since \mathbf{X} is held fixed, we can introduce the following substitution:

$$\mathbf{x}(0) := \mathbf{Q}(0) - \mathbf{a}(\mathbf{X}(0)), \tag{49}$$

which does not affect the integration measure, i.e. $d\mathbf{x}(0) = d\mathbf{Q}(0)$, thereby, the canonical partition and density functions read:

$$\begin{aligned} \rho(\mathbf{X}(0) | \mathbf{x}(0), \mathbf{I}(0)) &:= \frac{1}{Z} e^{-\frac{\beta}{2} (\langle \mathbf{I}, \mathbf{I} \rangle_{\mathbb{R}^{3N}} - \langle \mathbf{x}(0), \mathbf{K}\mathbf{x}(0) \rangle_{\mathbb{R}^{3N}})}, \\ Z &:= \int e^{-\frac{\beta}{2} (\langle \mathbf{I}, \mathbf{I} \rangle_{\mathbb{R}^{3N}} - \langle \mathbf{x}(0), \mathbf{K}\mathbf{x}(0) \rangle_{\mathbb{R}^{3N}})} d\mathbf{x}(0) d\mathbf{I}(0), \end{aligned} \tag{50}$$

and the integration measure assumes manifestly a Gaussian form.

By exploiting equation (49), the noise vector in equation (44) becomes:

$$\mathbf{F}(t) := \mathbf{\Lambda}^T (G_{\mathbf{K}}(t) \mathbf{I}(0) + H_{\mathbf{K}}(t) \mathbf{x}(0)), \tag{51}$$

and the computation of the second order correlation functions can be done easily [30]:

$$\langle \mathbf{I} \rangle_{\rho} = \langle \mathbf{x} \rangle_{\rho} = \mathbf{0}, \quad \langle \mathbf{I} \otimes \mathbf{I} \rangle_{\rho} = \frac{\mathbf{1}_{\mathbb{R}^{3N}}}{\beta}, \quad \langle \mathbf{x} \otimes \mathbf{x} \rangle_{\rho} = \frac{\mathbf{K}^{-1}}{\beta}, \quad \langle \mathbf{x} \otimes \mathbf{I} \rangle_{\rho} = \mathbf{0}. \tag{52}$$

Furthermore, we notice that the noise, \mathbf{F} , is a mean-zero vector, namely: $\langle \mathbf{F}(t) \rangle_{\rho} = \mathbf{0}$. In order to explicitly write the fluctuation-dissipation relation (noise autocorrelation function), we first expand the tensor product, i.e.:

$$\begin{aligned} \mathbf{F}(t) \otimes \mathbf{F}(\tau) &= \mathbf{\Lambda}^T (G_{\mathbf{K}}(t) \mathbf{I}(0) + H_{\mathbf{K}}(t) \mathbf{x}(0)) \otimes (\mathbf{I}(0) G_{\mathbf{K}}^T(\tau) + \mathbf{x}(0) H_{\mathbf{K}}^T(\tau)) \mathbf{\Lambda} \\ &= \mathbf{\Lambda}^T (G_{\mathbf{K}}(t) \mathbf{I}(0) \otimes \mathbf{I}(0) G_{\mathbf{K}}^T(\tau) + H_{\mathbf{K}}(t) \mathbf{x}(0) \otimes \mathbf{x}(0) H_{\mathbf{K}}^T(\tau) \\ &\quad + H_{\mathbf{K}}(t) \mathbf{x}(0) \otimes \mathbf{I}(0) G_{\mathbf{K}}^T(\tau) + G_{\mathbf{K}}(t) \mathbf{I}(0) \otimes \mathbf{x}(0) H_{\mathbf{K}}^T(\tau)) \mathbf{\Lambda}. \end{aligned} \tag{53}$$

Thus, by exploiting equation (52), the average of the tensor product above becomes:

$$\langle \mathbf{F}(t) \otimes \mathbf{F}(\tau) \rangle_{\rho} = \beta^{-1} \mathbf{\Lambda}^T \mathbf{K} \cos(\mathbf{K}^{1/2}(t - \tau)) \mathbf{\Lambda}, \tag{54}$$

where we used equation (40) and knowing that $\cos(\mathbf{K}^{1/2}t)$ and $\sin(\mathbf{K}^{1/2}t)$ commute with any polynomial of \mathbf{K} .

We obtained in equation (54), the fluctuation-dissipation relation; in fact, by comparing the operator on the right-hand side above with equation (44), we explicitly have:

$$\langle \mathbf{F}(t) \otimes \mathbf{F}(\tau) \rangle_{\rho} = \beta^{-1} \mathbf{L}(t - \tau). \tag{55}$$

As proof of consistence and show-case example, let us finally apply such a derivation to the Hamiltonian system in equation (12) proposed by Zwanzig. Thus, the equilibrium map is given by

$$\mathbf{a}_i^H(\mathbf{X}) := \frac{\eta_i}{\omega_i^2} \mathbf{X}, \quad \forall i \in [1, N],$$

whereas the interaction matrix \mathbf{K} is

$$\mathbf{K} := \sum_{i=1}^N \omega_i^2 \mathbf{e}_i \otimes \mathbf{e}_i, \tag{56}$$

In this setting, it consists in imposing $f_i(\mathbf{X}) := \mathbf{X}$ with $\eta_i \equiv \eta_{i,1} = \eta_i / \omega_i^2$ and $\eta_{i,I} = 0$ for any $I \in [2, n]$. This implies that the matrix $\mathbf{\Lambda}$ in equation (41) reduces to

$$\mathbf{\Lambda} := \sum_{i=1}^N \frac{\eta_i}{\omega_i^2} \mathbf{e}_i \otimes \mathbf{e}_i,$$

whereas

$$S_K(t - \tau) = \sum_{i=1}^N \omega_i^2 \sin(\omega_i(t - \tau)) \mathbf{e}_i \otimes \mathbf{e}_i. \quad (57)$$

Therefore, the kernel matrix reduces

$$\mathbf{L}(t - \tau) := \sum_{i=1}^N \frac{\eta_i^2}{\omega_i^2} \cos(\omega_i(t - \tau)) \mathbf{e}_i \otimes \mathbf{e}_i, \quad (58)$$

whose trace coincides with the ζ_Z function (15) in the Zwanzig's result [31]. However, now, the continuum limit can be computed. We conclude this section discussing the importance of including all the parameters representing the interactions (in this case η_i) directly in the definitions of the kernel matrix and noise terms. In particular, this can be appreciated as soon as one computes the continuum limit of the friction matrix, which consists in assuming that the set of heat bath frequencies $\mathcal{S}_d := \{\omega_i\}_{i=1}^N$ becomes a continuum set $\mathcal{S} \subset \mathbb{R}$ together with a distribution function $g(\omega)$ of frequencies which depends on the specific interactions we considered.

In this limit, one replaces the sum over the frequencies with an integral [31]:

$$\sum_{i=1}^N \eta_i \longrightarrow \int_{\mathcal{S}} \eta(\omega) g(\omega) d\omega,$$

where $\{\eta_i\}_{i=1}^N$ is any set of coefficients and $\eta(\omega)$ is the associated frequency-dependent function in the continuum limit.

Now, if the kernel function is defined as in equation (17), i.e., without including the parameters γ_i , the continuum limit, which reads:

$$\xi(t - \tau) \longrightarrow \int_{\mathcal{S}} \omega^2 \cos(\omega(t - \tau)) g(\omega) d\omega,$$

cannot provide a bounded continuous kernel function.

Conversely, including the coefficients γ_i in the kernel as in (18), one assumes that the set $\{\gamma_i\}_{i=1}^N$ becomes a continuous function $\gamma(\omega)$ of the frequency variable, so that the continuum limit can be applied

$$\zeta(t - \tau) \longrightarrow \int_{\mathcal{S}} \frac{\cos(\omega(t - \tau))}{\omega^2} \gamma^2(\omega) g(\omega) d\omega, \quad (59)$$

choosing γ such that it remains bounded as $\omega \rightarrow \infty$.

The degree of freedom represented by the γ function can be fixed for requiring the boundedness of the kernel function. In practice, the remaining effort consists in providing the correct modeling, depending on the interaction nature, of the function $J(\omega) := \gamma^2(\omega)g(\omega)$ even called *density function*. with the existing techniques already introduced in the literature [19].

3. Conclusion

We have showed that in order to obtain a kernel matrix which contains the parameters of the interactions and, thereby, that admits a *thermodynamics limit* as proposed by Zwanzig, we have to pay attention to the definition of equilibrium map \mathbf{a} which plays a fundamental role.

In fact, its mathematical structure needs to be specified somehow so to distinguish the contributions given by the interaction parameters and the functional dependence on \mathbf{X} . In so doing, one obtains a nonlinear generalized Langevin equation (see equation (45)) where the kernel matrix and noise vector explicitly depend on the parameters $\eta_{i,l}$ which is impossible to obtain if we followed Zwanzig's procedure as we have shown in the introductory section. Finally, with these definitions, we can proceed in computing the continuum limit without introducing further assumptions for making the kernel function bounded.

Such a derivation could seem a matter of little consequence if we just have a heavy particle linearly interacting with a single heat bath system. In this case, it is always possible to understand how to readjust the kernel function definition (9) in order to include the parameters. However, if we had different kinds of heat baths which interact in a nonlinear way with the heavy particle, the adjustment previously mentioned cannot be applied anymore but a different derivation as that proposed in this paper should be considered.

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Data availability statement

All data that support the findings of this study are included within the article (and any supplementary files).

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