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A Generalized Langevin Equation for Dealing with Nonadditive Fluctuations

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A suitable extension of the Mori memory-function formalism to the non-Hermitian case allows a "multiplicative" process to be described by a Langevin equation of non-Markoffian nature. This generalized Langevin equation is then shown to provide for the variable of interest the same autocorrelation function as the well-known theoretical approach developed by Kubo, the stochastic Liouville equation (SLE) theory. It is shown, furthermore, that the present approach does not disregard the influence of the variable of interest on the time evolution of its thermal bath. The stochastic process under study can also be described by a Fokker–Planck-like equation, which results in a Gaussian equilibrium distribution for the variable of interest. The main flaw of the SLE theory, that resulting in an uncorrect equilibrium distribution, is therefore completely eliminated.

KEY WORDS: Stochastic process of "multiplicative" type; stochastic Liouville equation; multidimensional Langevin equation; non-Markoffian Langevin equation; Fokker–Planck equation for non-Markoffian processes.

1. INTRODUCTION

The principal purpose of the present paper is to develop a new theoretical approach to the study of "multiplicative" stochastic processes such as

$$\frac{du}{dt} = i\omega(t)u \tag{1.1}$$

This is the well-known stochastic oscillator studied by Kubo.[1] The stochastic process described by Eq. (1.1) is termed "multiplicative" according to the nomenclature of Fox.[2,3] Within the context of the stochastic Liouville equation (SLE) theory,[1] the motion of the variable $\omega(t)$ is

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thought of as independent of the variable u. Furthermore, this stochastic process is assumed to be both Markoffian and Gaussian. As a consequence, its correlation function is given by

$$\langle \omega\omega(t) \rangle = \Delta^2 e^{-\gamma t} \tag{1.2}$$

These assumptions need not be regarded as being serious limitations. In fact, it has been shown[4–6] that the SLE theory can even be extended to the case where $\omega(t)$ is neither a Markoffian nor a Gaussian stochastic variable. As is well known, the main flaw of this theory depends on the fact that the motion of $\omega(t)$ is assumed to be independent of that of $u(t)$. As a consequence (see Appendix A) the correct equilibrium distribution of u is not obtained. Several attempts at correcting this flaw have been made.[7–9] Tokuyama and Mori[7] developed a new approach to the subject of generalized Brownian motion which complements the well-known theory of the generalized Langevin equation given by Mori in two previous papers.[10,11] Though, from a formal point of view, this theory should allow the back reaction of the variable of interest on its thermal bath to be taken into account, when actually dealing with the stochastic oscillator[7] they made the same main assumption as Kubo did,[1] i.e., that the stochastic process $\omega(t)$ is independent of $u(t)$.

Memoryless equations of the same kind as that found by Tokuyama and Mori have also been obtained by Hashitsume, Shibata, and Shingū[8] and Shibata, Takahashi, and Hashitsume.[9] Their approach, too, shares the main flaw of the SLE theory.

In the present paper, we shall focus our attention on a Gaussian stochastic variable $\omega(t)$ which in the absence of the u–ω coupling satisfies Eq. (1.2). Then we shall explicitly evaluate the motion of $\omega(t)$ when this interaction is turned on.

The theory developed in the present paper is based on an extension to the non-Hermitian case of the approach developed in previous papers.[4,5,12,13] In order to make this paper as much self-sufficient as possible, in the next section we shall summarize the main results of these papers.

In Section 3 we shall point out that a multiplicative form can straightforwardly be obtained by expressing the equation of motion of the variable of interest in the interaction picture. We also shall show that a complete knowledge of the stochastic properties of the resulting multiplicative variable is tantamount to replacing the unperturbed Liouvillian concerning the thermal bath with a diffusion operator playing the role of effective Liouvillian. Since the new total Liouvillian is no longer Hermitian, a suitable generalization of the Mori approach[10,11] is required.

In Section 4 we shall apply our approach to the special case of Kubo's stochastic oscillator. Concluding remarks can be found in Section 5.

2. THE CHAIN OF THE MORI VARIABLES AS A CONVENIENT BASIS SET FOR EXPANDING THE LIOUVILLE OPERATOR

In a well-known paper,[10] Mori showed that the equation of motion of a variable of interest, a, governed by the time-independent Liouville operator L,

$$\frac{da}{dt}(t) = iLa(t) \tag{2.1}$$

can be transformed into an equation of Langevin type,

$$\frac{da}{dt}(t) - i\omega a(t) + \int_0^t \varphi(t-s)a(s)\,ds = f(t) \tag{2.2}$$

where

$$i\omega \equiv (iLa, a^*)\cdot(a, a^*)^{-1} \tag{2.3}$$

$$\varphi(t) \equiv (f(t), f^*)\cdot(a, a^*)^{-1} \tag{2.4}$$

and

$$f(t) \equiv e^{tQiL}QiLa \tag{2.5}$$

Q is the projection operator onto the subspace orthogonal to a, defined as follows:

$$Q \equiv 1 - P \tag{2.6}$$

The operator P, in turn, is defined by

$$Pg \equiv (g, a^*)\cdot(a, a^*)^{-1}a \tag{2.7}$$

The previous results are based on the preliminary definition of a suitable scalar product (f, g^*) for two arbitrary variables f and g.

In a subsequent paper,[11] Mori showed that the Laplace transform of the "memory kernel" $\varphi(t)$ of Eq. (2.2) is given by the following infinite continued fraction:

$$\hat{\varphi}_0(z) \equiv \hat{\varphi}(z) = \cfrac{\Delta_1^2}{z - i\omega_1 + \cfrac{\Delta_2^2}{z - i\omega_2 + \ddots + \cfrac{\Delta_n^2}{z - i\omega_n + \hat{\varphi}_n(z)}}} \tag{2.8}$$

$$\hat{\varphi}_n(z) \equiv \int_0^\infty e^{-zt}(f_{n+1}(t), f_{n+1}^*)\cdot(f_n, f_n^*)^{-1} = \frac{\Delta_{n+1}^2}{z - i\omega_{n+1} + \hat{\varphi}_{n+1}(z)} \tag{2.9}$$

where

$$\Delta_j^2 \equiv (f_j, f_j^*) \cdot (f_{j-1}, f_{j-1}^*)^{-1} \tag{2.10}$$

$$i\omega_j \equiv (iL_j f_j, f_j^*) \cdot (f_j, f_j^*)^{-1} \tag{2.11}$$

It is useful to recall that

$$i\omega_0 \equiv i\omega \tag{2.11'}$$

The new dynamical variables f_j are generated by the recurrence equation

$$f_j \equiv iL_j f_{j-1} \qquad (j = 1, 2, \ldots, n) \tag{2.12}$$

$$f_0 \equiv a \tag{2.12'}$$

where ($iL_0 \equiv iL$)

$$iL_j \equiv \left(\prod_{r=0}^{j-r} Q_r \right) iL \tag{2.13}$$

$$Q_r \equiv 1 - P_r \tag{2.14}$$

$$P_r g \equiv (g, f_r^*) \cdot (f_r, f_r^*)^{-1} f_r \tag{2.14'}$$

Equation (2.5) provides a deterministic time evolution for the fluctuating force $f(t)$. This deterministic time evolution depends on the dynamics of the space complementary to that of the variable a. When this subsystem is characterized by an overwhelming number of freedom degrees, it is usually assumed[3] that $f(t)$ can be regarded as a stochastic force. As a consequence, Eq. (2.2) becomes a stochastic integrodifferential equation.

As is well known, any computational problem resulting from the presence of the stochastic force can be avoided by substituting the corresponding Fokker–Planck equation for the Langevin equation. In the case of the generalized Langevin equation of Eq. (2.2), this replacement has been attempted by several authors.[14–17] They found a time-convolutionless equation for the conditional probability, which in the Gaussian case assumes a form of Fokker–Planck type. Such an equation has been extended to dealing with the non-Gaussian[17,15] and nonlinear[17,18] case. As pointed out by Fox[16] and Hänggi et al.,[19] however, these equations are not bona fide Fokker–Planck equations, in that they do not provide a complete description of the stochastic process under study.

According to Refs. 4, 5, 13, 20, by using the results of the Mori theory expressed by Eq. (2.8), a completely different approach, resulting in a bona fide Fokker–Planck equation, can be followed. In fact, in Appendix B it is shown that if $\hat{\varphi}_n(z)$ is assumed to be independent of z, then Eq. (2.2) can be replaced by

$$\frac{d\mathbf{U}}{dt} = \mathbf{\Gamma U} + \mathbf{F}(t) \tag{2.2'}$$

where

$$\Gamma \equiv \begin{vmatrix} i\omega_0 & \Delta_1 & 0 & \cdots & 0 \\ -\Delta_1 & i\omega_1 & \Delta_2 & \cdots & 0 \\ 0 & -\Delta_2 & i\omega_2 & \cdots & 0 \\ \cdots & \cdots & \cdots & \cdots & \Delta_n \\ 0 & 0 & 0 & -\Delta_n & i\omega_n - \gamma_n \end{vmatrix} \tag{2.2''}$$

$$\gamma_n \equiv \hat{\varphi}_n(0) \tag{2.2'''}$$

and

$$\mathbf{F}(t) \equiv \begin{bmatrix} 0 \\ 0 \\ \vdots \\ F_n(t) \end{bmatrix} \tag{2.2^{IV}}$$

$\mathbf{U}(t)$ is a multidimensional stochastic variable with components U_0, $U_1, \ldots, U_n$. The first component $U_0(t)$ has to be identified with $a(t)$, Eq. (2.2). Only the last component directly undergoes a fluctuation–dissipation process expressed by

$$(F_n(t), F_n^*) = 2\gamma_n \delta(t) \tag{2.2^{V}}$$

As said before, we regard Eq. (2.2′) as a stochastic equation. The variable $\mathbf{U} \equiv (U_0, U_1, \ldots, U_n)$, when thought of as stochastic will be denoted by the new symbol $\mathbf{u} \equiv (u_0, u_1, \ldots, u_n)$. When $\mathbf{U}$ is replaced by $\mathbf{u}$, Eq. (2.2′) results in a multidimensional Langevin equation of the same kind as that studied by Fox and Uhlenbeck.[21] Since they were able to build up the corresponding Fokker–Planck equation, we can use their result to solve our problem. Thus we find (Appendix B) that the Fokker–Planck equation to be associated with Eq. (2.2) is

$$\frac{\partial}{\partial t_2} P_2(\mathbf{u}^{(1)}, t_1; \mathbf{u}^{(2)}, t_2) = \Gamma_{\mathbf{u}} P_2(\cdots) \tag{2.15}$$

where (we assume that $\omega_i = 0$)

$$\begin{aligned} \Gamma_{\mathbf{u}} = & -\Delta_1 \left(\frac{\partial}{\partial u_0^{(2)}} u_1^{(2)} - \frac{\partial}{\partial u_1^{(2)}} u_0^{(2)} \right) - \Delta_2 \left(\frac{\partial}{\partial u_1^{(2)}} u_2^{(2)} - \frac{\partial}{\partial u_2^{(2)}} u_1^{(2)} \right) \\ & + \cdots - \Delta_n \left(\frac{\partial}{\partial u_{n-1}^{(2)}} u_n^{(2)} - \frac{\partial}{\partial u_n^{(2)}} u_{n-1}^{(2)} \right) \\ & + \gamma_n \left(\frac{\partial}{\partial u_n^{(2)}} u_n^{(2)} + q_n^2 \frac{\partial^2}{\partial (u_n^{(2)})^2} \right) \end{aligned} \tag{2.15'}$$

Equation (2.15) is the bona fide Fokker–Planck equation to be associated with Eq. (2.2). Of course, this Fokker–Planck equation is only approximatively equivalent to Eq. (2.2). However, its accuracy increases as the parameter n increases. Equation (2.15) becomes completely equivalent to Eq. (2.2) for $n \to \infty$. From a practical point of view, the continued fraction of Eq. (2.8) is really useful only in the case of fast convergence.

It may happen, however, that the approximations suggested by the physical system under study lead us to a different kind of continued fraction, faster than the standard type of Eq. (2.8). An illuminating case is that where the thermal bath consists of two subsystems sequentially coupled with the variable of interest. Let us assume that the relaxation properties of the first subsystem, which depend on the interaction with the second one, are known. It is then possible to show that the standard Mori theory[11] is not suitable for exploiting the advantages of such an approximation. A first example is discussed in Appendix C. A second example of the same type is that of a magnetic spin interacting with a fluctuating magnetic field, the fluctuations of which are driven by the molecular motion. The theory of Brownian motion can then afford useful information on the mathematical properties of the stochastic variable [$\omega(t)$ in the case of Eq. (1.1)].

We would like to stress that the approximation expressed by Eq. (2.2‴) at first glance would seem to be equivalent to replacing the rigorous equation of motion given by Eq. (2.1) with the following one:

$$\frac{d}{dt} a(t) = iL_{\text{eff}} a(t) \tag{2.16}$$

L_{eff} is an approximate Liouville operator, which on the basis set of the first $n+1$ normalized variables of the standard Mori chain reads

$$iL_{\text{eff}} = \begin{bmatrix} i\omega_0 & \Delta_1 & 0 & \cdots & 0 & 0 \\ -\Delta_1 & i\omega_1 & \Delta_2 & \cdots & 0 & 0 \\ 0 & -\Delta_2 & i\omega_2 & \cdots & 0 & 0 \\ \cdots & \cdots & \cdots & \cdots & \cdots & \cdots \\ 0 & 0 & 0 & \cdots & i\omega_{n-1} & \Delta_n \\ 0 & 0 & 0 & \cdots & -\Delta_n & i\omega_n - \gamma_n \end{bmatrix} \tag{2.17}$$

However, even in the case when the truncation at nth order is exact, Eq. (2.1) and Eq. (2.16) are not completely equivalent. In fact, even if Eq. (2.17) results in the same autocorrelation function for the variable a as Eq. (2.16), its thermodynamical equilibrium is not correctly reproduced because L_{eff} does not take into account the fluctuating force acting on the last variable of the truncated Mori chain.

Let us now consider the following equation of motion:

$$\frac{du_0}{dt} = \Gamma_{\mathbf{u}} u_0(t) \tag{2.18}$$

This equation is based on the same central idea as that on which the approach of the next section is grounded. We mean the replacement of the rigorous Liouvillian L by the diffusion operator $\Gamma_{\mathbf{u}}$. It is reminiscent of the method followed by Mori and Fujisaka,[22] who used a generalized Fokker–Planck operator for driving the motion of the variable of interest. Equation (2.18) requires that an appropriate scalar product be defined. On intuitive ground one would be tempted to use the following definition: If $g_1(\mathbf{u})$ and $g_2(\mathbf{u})$ are two generic functions of the multidimensional stochastic variable $\mathbf{u}$, then

$$(g_1, g_2^*) \equiv \int d\mathbf{u}\, g_2^*(\mathbf{u}) g_1(\mathbf{u}) w_0(\mathbf{u}) \tag{2.19}$$

where $w_0(\mathbf{u})$ is the equilibrium distribution. In Appendix E, however, we shall use a more general definition including that of Eq. (2.19). In so doing, we shall show that the Laplace transform of $(u(t), u^*)$ results in the same continued fraction expansion as the Laplace transform of $(a(t), a^*)$. As far as the equilibrium distribution is concerned, however, Eq. (2.18) shares the flaw of Eq. (2.16). Equations such as Eq. (2.18) can be trusted only when used for evaluating the correlation function $(u_0(t), u_0^*)$. In order to obtain a correct equilibrium distribution the corresponding Langevin (or Fokker–Planck) equations have to be applied. This important remark has to be kept in mind throughout the remaining part of this paper.

3. EQUATION OF MOTION IN THE INTERACTION PICTURE AS STOCHASTIC PROCESS OF MULTIPLICATIVE KIND

Let us assume that the physical system under study be divided into two subsystems, **S** and **B**, interacting with each other. Let **S** be the subsystem of interest and a the corresponding dynamical variable. We mean that the variable $a \equiv a(0)$ only depends on the **S** phase space. The equation of motion of $a(t)$ can be written as follows:

$$\frac{da(t)}{dt} = i(L_{\mathbf{S}} + L_{\mathbf{I}} + L_{\mathbf{B}}) a(t) \tag{3.1}$$

where $L_{\mathbf{I}}$ is the Liouville operator concerning the interaction between **S** and **B**. Such a kind of partition of the Liouville operator L can also be founded on the standard Mori approach outlined in Section 2. We can call $L_{\mathbf{S}}$ the

part of L spanned by the "state" f_0 and L_B that spanned by all other "states" of the Mori chain. The interaction L_1 is then proportional to the parameter Δ_1. An approach based on assumptions concerning the modulation of this interaction from the remaining part of the Mori chain results in a new picture of the thermal bath. In this section we want to provide a theoretical approach to this new picture.

In the interaction representation Eq. (3.1) can be written as follows:

$$\frac{du(t)}{dt} = i\eta(t)u(t) \tag{3.2}$$

where

$$\eta(t) \equiv e^{-i(L_S+L_B)t}L_1 e^{i(L_S+L_B)t} \tag{3.3}$$

and

$$u(t) \equiv e^{-i(L_S+L_B)t}a(t) \tag{3.4}$$

To some extent, $\eta(t)$, Eq. (3.3), is reminiscent of $f(t)$, Eq. (2.2). Both $f(t)$ and $\eta(t)$ exhibit a deterministic dependence on the "irrelevant" part of the physical system under study. In Section 2 we remarked that when the "irrelevant" part involves a huge number of freedom degrees, it is convenient to replace the deterministic approach by a stochastic one. If this suggestion is followed, $\eta(t)$ has to be replaced by the stochastic variable $\omega(t)$ and Eq. (3.2) has to be identified with Eq. (1.1). The stochastic variable $\omega(t)$, in turn, in general obeys an equation of the same form as Eq. (2.2),

$$\frac{d\omega}{dt} = i\Xi\omega(t) - \int_0^t \varphi_\omega(t-\tau)\omega(\tau)\,d\tau + f_\omega(t) \tag{3.5}$$

We assume that $\hat{\varphi}_\omega(z)$ is given by a truncated continued fraction. Then, according to the theory of Section 2, we can replace Eq. (3.5) by

$$\frac{d\omega}{dt} = \Gamma_\Omega\omega \tag{3.6}$$

where Γ_Ω is the multidimensional Fokker–Planck operator to be associated with the generalized Langevin equation itself. We have to keep in mind that when the thermal bath driving the motion of $\omega(t)$ is assumed to be in its equilibrium state, Eq. (3.6) is equivalent to

$$\frac{d}{dt}\omega = \Gamma_\Omega^x\omega \equiv \Gamma_\Omega\omega - \omega\Gamma_\Omega \tag{3.6'}$$

As a consequence, if $\eta(t)$ is replaced by $\omega(t)$, Eq. (3.1) becomes

$$\frac{d}{dt}a = \mathcal{L}_0 a \equiv \left[i(L_S+\omega) + \Gamma_\Omega\right]a(t) \tag{3.7}$$

In fact, when written in the interaction picture, Eq. (3.7) reads

$$\frac{du}{dt} = i\omega(t)u(t) \tag{3.2'}$$

which is the stochastic counterpart of Eq. (3.2) [$\omega(t) \equiv e^{\Gamma_\Omega^\dagger t}\omega(0)$].

Though the total "Liouville operator" $i(L_S + \omega) + \Gamma_\Omega$ is not Hermitian, it is still possible to build up an expansion basis set of the same kind as that of the previous section (see Appendix D). The theory developed in Appendix D can be regarded as a generalization of that of Schneider[23] who, in turn, extended the Mori theory. In fact, Schneider[23] only dealt with pseudosymmetric systems, whereas the theory of Appendix D can be applied to operators of any mathematical type. Then, following the same approach as the corresponding one in Section 2, we can replace Eq. (3.7) with

$$\frac{d}{dt}\mathbf{u} = \boldsymbol{\Gamma}\mathbf{u}(t) + \mathbf{F}(t) \tag{3.8}$$

where

$$\boldsymbol{\Gamma} = \begin{pmatrix} \lambda_0 & \Delta_1 & 0 & \cdots \\ -\Delta_1 & \lambda_1 & \Delta_2 & \cdots \\ 0 & -\Delta_2 & \lambda_2 & \cdots \\ \cdots & \cdots & \cdots & \cdots \end{pmatrix} \tag{3.9}$$

The parameters λ_i and Δ_i can be obtained by the prescriptions of Appendix D. The dimension n of the variable $\mathbf{u}$ is determined by the required degree of accuracy in evaluating, for instance, the correlation function $(u_0(t), u_0^*)$. However, the matrix $\boldsymbol{\Gamma}$ can be truncated without any need of replacing the remaining part of the chain with an additional damping. In fact, a damping contribution is already present in the parameters λ_i, which are complex numbers also containing a real part.

This subtle characteristic of our approach requires some comments. Once the structure of the matrix $\boldsymbol{\Gamma}$ has been determined, the stochastic force $\mathbf{F}(t)$ has to be related to the dissipation part of the matrix $\boldsymbol{\Gamma}$ itself by a fluctuation–dissipation relation, which will afford a correct square average value for the variable of interest. Let us consider the case where the parameters λ_i are endowed with only the real part (see Appendix E). The fluctuation–dissipation relation of the Fox–Uhlenbeck theory[21] can then be written as follows (see also Appendix B):

$$(\mathbf{F}(t), \mathbf{F}^*) = 2\boldsymbol{\Lambda}\delta(t) \tag{3.10}$$

where $\boldsymbol{\Lambda}$ is a diagonal matrix, the nonvanishing elements of which are given

by

$$\gamma_i \equiv (\Gamma)_{ii} = -\lambda_i \tag{3.11}$$

In the non-Gaussian case, the higher-order moments of $\mathbf{F}(t)$ cannot be expressed in terms of Λ. However, the equilibrium square average value of the variable of interest is independent of their actual values.[24] On the contrary, the correlation functions $(u_0^{2n}(t), u_0^{2n}*)$ are significantly affected by any non-Gaussian contribution to the stochastic force. Our approach could provide a faithful description of such a non-Gaussian behavior as a result of further investigation on the physical properties of the system under study, whereas the "non-Gaussian" behavior of Kubo's stochastic oscillator (see Appendix A) is an artifact of the SLE theory.

Failing information on higher moments of $\mathbf{F}(t)$, we assume that this stochastic force is Gaussian. Then, by using the same approach as that which led us to Eq. (2.15), we obtain

$$\frac{\partial}{\partial t_2} P_2(\mathbf{u}^{(1)}, t_1; \mathbf{u}^{(2)}, t_2) = \Gamma_{\mathbf{u}} P_2(\mathbf{u}^{(1)}, t_1; \mathbf{u}^{(2)}, t_2) \tag{3.12}$$

where $\Gamma_{\mathbf{u}}$ is defined by

$$\Gamma_{\mathbf{u}} \equiv -\sum_{r=0}^{n-1} \Delta_{r+1} \left(\frac{\partial}{\partial u_r^{(2)}} u_{r+1}^{(2)} - \frac{\partial}{\partial u_{r+1}^{(2)}} u_r^{(2)} \right) + \sum_{r=0}^{n} \gamma_r \left[\frac{\partial}{\partial u_r^{(2)}} u_r^{(2)} + q_r^2 \frac{\partial^2}{\partial (u_r^{(2)})^2} \right] \tag{3.13}$$

In the case where the "memory kernel" of Eq. (3.5) is endowed with a finite lifetime, the theory developed in the present section leads to a generalization of the SLE theory along the same lines as those of Ref. 5. Let us recall, furthermore, that Eq. (3.12) has been built up by making the usual Gaussian assumption on the stochastic variable $\omega(t)$. We can then emphasize a further advantageous feature of our approach: The diffusion operator $\Gamma_{\mathbf{u}}$ when constructed along the lines of Refs. 24 and 6 would enable us to avoid this assumption.

It is important to stress that Eq. (3.8) leads to a generalized Langevin equation with a "memory kernel" the Laplace transform of which exhibits the generalized structure found in Appendix D. By following the lines of Ref. 5 (see Appendix F), we find that u_0 satisfies the following generalized Langevin equation:

$$\frac{d}{dt} u_0(t) = \lambda_0 u_0(t) - \int_0^t ds\, \varphi(t-s) u_0(s) + f(t) \tag{3.14}$$

where

$$\hat{\varphi}(z) = \cfrac{\Delta_1^2}{z - \lambda_1 + \cfrac{\Delta_2^2}{z - \lambda_2 + \ddots}} \tag{3.15}$$

However, Eq. (3.14) is not completely equivalent to the generalized Langevin equation derived by using the technique of Mori type of Appendix D, Eq. (D.38) with $k = 0$. In fact, as shown in Appendix F, the Laplace transform of $f(t)$ can be expressed in terms of the Laplace transforms of the components of $\mathbf{F}(t)$ as follows:

$$\hat{f}(z) = \hat{F}_0(z) + \cfrac{\Delta_1}{z - \lambda_1 + \cfrac{\Delta_2^2}{z - \lambda_2 + \cfrac{\Delta_3^2}{z - \lambda_3 + \cdots}}} \times \left\{ u_1(0) + \hat{F}_1(z) + \cfrac{\Delta_2}{z - \lambda_2 + \cfrac{\Delta_3^2}{z - \lambda_3 + \cfrac{\Delta_4^2}{z - \lambda_4 + \ddots}}} \times \left[u_2(0) + \hat{F}_2(z) + \cfrac{\Delta_3}{z - \lambda_3 + \cfrac{\Delta_4^2}{z - \lambda_4 + \cfrac{\Delta_5^2}{z - \lambda_5 + \ddots}}} \times (\cdots \quad \cdots) \right] \right\} \tag{3.16}$$

It means that the fluctuating force $f(t)$ also depends on the variables u_i ($i \geqslant 1$), which at $t = 0$ can be found far from their thermodynamical

equilibrium, whereas the generalized Langevin equation of the Mori theory, Eq. (2.2), and its generalized version of Appendix D as well, is derived in the presence of a thermal bath assumed to be in its equilibrium state. This is the reason why the present approach can be used to evaluate the spectroscopic effects of thermal bath excitations.[25] The problem of preparation has recently been studied within the context of different theoretical approaches.[26,27] A detailed comparison between the present method and those of Refs. 26, 27 could be the subject of a future stimulating investigation.

4. A COMPARISON BETWEEN THE SLE THEORY AND THE NEW APPROACH

The generalized Mori theory of Appendix D shares the main features of the standard Mori approach described in Section 2. This theory consists in building up a suitable basis set for expanding a non-Hermitian dynamical operator $\mathcal{L}_0$ such as that of Eq. (3.7).

In this reference framework, when the "quantum-mechanical" notation of Appendices D and E is used, $\mathcal{L}_0$ can be written as follows:

$$\mathcal{L}_0 = \sum_{r=0}^{\infty} |\tilde{m}_r\rangle\lambda_r\langle m_r| - \sum_{r=0}^{\infty} \Delta_{r+1}\{|\tilde{m}_r\rangle\langle m_{r+1}| - |\tilde{m}_{r+1}\rangle\langle m_r|\} \tag{4.1}$$

We can assume that $L_S = 0$. Furthermore the stochastic variable $\omega(t)$ is assumed to be both Gaussian and Markoffian. As a consequence Γ_Ω is replaced by

$$\Gamma_\omega \equiv \gamma\left\{\frac{\partial}{\partial\omega}\omega + \Delta^2\frac{\partial^2}{\partial\omega^2}\right\} \tag{4.2}$$

As shown in Appendix E, Eq. (4.1) then becomes

$$\mathcal{L}_0 = -\sum_{r=0}^{\infty} |\tilde{m}_r\rangle(r-1)\gamma\langle m_r| - \Delta\sum_{r=0}^{\infty}(r+1)^{1/2}\{|\tilde{m}_r\rangle\langle m_{r+1}| - |\tilde{m}_{r+1}\rangle\langle m_r|\} \tag{4.3}$$

We can use $\mathcal{L}_0$, Eq. (4.3), for evaluating the time evolution of $\omega(t)$. The main difference between the SLE theory and the present approach is that this time evolution is now determined also by the interaction between ω and the part of interest itself. In the case under study the stochastic variable ω has to be regarded as being an operator on the space spanned by the states $|m_r\rangle$ to be denoted by $\hat{\omega}$. Its nonvanishing matrix elements are then given by (see Appendix E)

$$-\langle\tilde{m}_r|\hat{\omega}|m_{r+1}\rangle = \langle\tilde{m}_{r+1}|\hat{\omega}|m_r\rangle = (r+1)^{1/2}\Delta \tag{4.4}$$

It is straightforward to check that if we replace $\mathfrak{L}_0$ by

$$\mathfrak{L}_0' \equiv -\sum_{r=0}^{\infty} |m_r\rangle(r-1)\gamma\langle\tilde{m}_r| \tag{4.3'}$$

the corresponding time evolution of ω as given by

$$\frac{d\hat{\omega}}{dt} = \mathfrak{L}_0'\hat{\omega} \tag{4.5}$$

completely satisfies the Gaussian requirements of Eqs. (A.2) provided that the moments of $\omega(t)$ are defined by

$$\langle\omega(t_1)\omega(t_2)\cdots\rangle \equiv \langle\tilde{m}_0|\hat{\omega}(t_1)\hat{\omega}(t_2)\cdots|m_0\rangle$$

However, when the total Liouville operator of Eq. (4.3) is used, the time evolution of ω is shown to exhibit significant deviations from the free decay behavior. Figure 1 shows the behavior of the correlation function $C_\omega(t)$

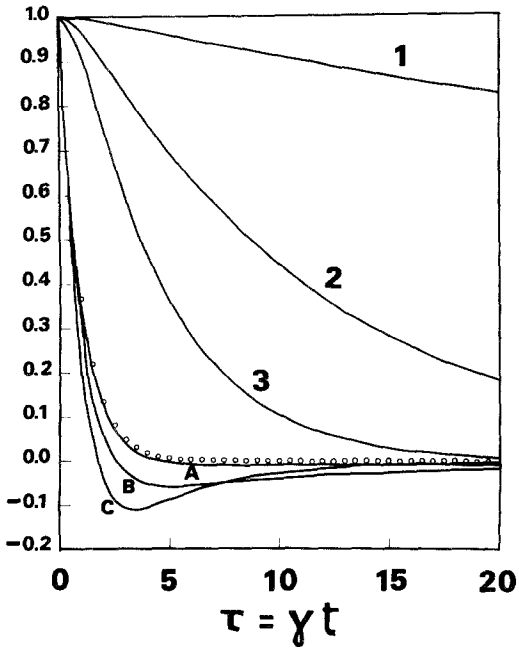

Fig. 1. Time evolution of the correlation functions $C_{u_0}(\tau) \equiv (u_0(\tau), u_0^*)$ (1, 2, 3) and $C_\omega(\tau) \equiv \langle\omega\omega(\tau)\rangle$ (A, B, C). The parameter τ is related to the time t by $\tau = \gamma t$. Values of the parameter $\Delta: 0.1\gamma$ (1, A), 0.3γ (2, B), 0.5γ (3, C). All correlation functions are evaluated by using the present approach. The curves 1, 2, and 3 coincide with the corresponding ones obtained by using the SLE theory, Eq. (A.6). The small open circles denote the free decay of the correlation function $C_\omega(\tau)$.

$\equiv \langle \omega\omega(t) \rangle$ for increasing values of Δ. The agreement with the SLE theory is fairly good for small values of the "memory strength" parameter $g \equiv 2\Delta/\gamma$. On the contrary, when $g \sim 1$, the correlation function $C_\omega(t)$ can also have negative values and slowly attains a steady vanishing value for $t \to \infty$. Of course, the correlation function $C_{u_0}(t) \equiv (u_0(t), u_0^*)$ coincides with that given by the SLE theory.

As far as the equilibrium distribution of u_0 is concerned we can have recourse to Eq. (3.13). By using again the diffusion operator of Eq. (4.2) and the explicit expressions for the parameters Δ_i and λ_i given in Appendix E, we obtain

$$\frac{\partial}{\partial t_2} P_2(\mathbf{u}^{(1)}, t_1; \mathbf{u}^{(2)}, t_2) = -\sum_{r=0}^{\infty} (r+1)^{1/2} \Delta \left(\frac{\partial}{\partial u_r^{(2)}} u_{r+1}^{(2)} - \frac{\partial}{\partial u_{r+1}^{(2)}} u_r^{(2)} \right) P_2(\cdots)$$
$$+ \sum_{r=1}^{\infty} r\gamma \left\{ \frac{\partial}{\partial u_r^{(2)}} u_r^{(2)} + q^2 \frac{\partial^2}{\partial \left(u_r^{(2)}\right)^2} \right\} P_2(\cdots) \tag{4.6}$$

It is straightforward to check that the corresponding equilibrium distribution, $w_0(\mathbf{u})$, is

$$w_0(\mathbf{u}) = \prod_{r=0}^{\infty} \left[\frac{q e^{-(u_r^2/q^2)}}{(2\pi)^{1/2}} \right] \tag{4.7}$$

which results in a Gaussian distribution for the variable of interest u_0 when the other variables are wiped out. As a consequence, the higher-order correlation functions

$$C_{u_0}^{(n)}(t) \equiv \left(u_0^{2n}(t), u_0^{2n*}\right) \tag{4.8}$$

are shown to attain the correct value $C_{u_0}(\infty) = (2n-1)!!$, as required by the Gaussian assumption on the stochastic force $\mathbf{F}(t)$ done in the previous section. To obtain the simple equilibrium distribution of Eq. (4.7) we had to assume all the parameters q_r of Eq. (3.13) to have the common value q.

The basic aspects of our approach can be summarized as follows. The first step consists in replacing the true many-body thermal bath with a "reduced" equivalent one. This aspect is also shared by Adelman's approach, which has recently been proven successful for studying a wide class of liquid state as well as solid state phenomena.[28] In the present case the "reduced" thermal bath has to be proven to result in the same properties of the stochastic variable $\omega(t)$ as the true one. Appendix D teaches us how to do that without introducing any additional arbitrary parameter. The coupling Δ_i and the dampings γ_i are indeed expressed in terms of the parame-

ters Δ and γ, respectively, defining the correlation function of Eq. (1.2). The actual size of the chain of fictitious variables also depends on the statistical nature of the stochastic variable $\omega(t)$. For example, if $\omega(t)$ were a two-state Poisson stochastic variable, the equivalent "reduced" thermal bath would involve a fictitious variable alone. Any fictitious variable is affected by a damping, γ_i. Each damping γ_i, in turn, can be traced back to the presence of an additive stochastic force $f_i(t)$ satisfying the fluctuation–dissipation relation $(f_i(t), f_i^*) = 2q^2\gamma_i\delta(t)$ which results in a correct equilibrium distribution of the corresponding fictitious variable. This important requirement can be satisfied without affecting the unperturbed properties of the fluctuating frequency $\omega(t)$. The Fokker–Planck equation of Eq. (4.6) then shows that also the variable of interest attains a correct thermodynamical equilibrium. After replacing the true thermal bath with the "reduced" one, complete information on the dynamics of our system can straightforwardly be obtained, including the back-reaction of the variable of interest on the thermal bath itself. The actual time behavior of the stochastic variable $\omega(t)$ can be strongly affected by the couplings Δ_i.

5. CONCLUDING REMARKS

The basic idea of our approach consists in replacing a "multiplicative" relaxation process by an equivalent one of additive nature. In a recent paper,[27] Tokuyama transformed a general multiplicative stochastic process into an additive one. The corresponding Fokker–Planck equation was shown to contain a nonlinear drift term. Since, in general, a nonlinear drift term can be traced back to a "memory kernel,"[22] the present approach could be connected with that by Tokuyama. It should be stressed, however, that Tokuyama did not explicitly discuss the problem which has been the major topic under discussion in the present paper.

An interesting attempt of correcting the main flaw of the SLE theory has also been made by Stillman and Freed.[29] They applied rigorous constraint of detailed balance to the reversible drift terms in the Fokker–Planck equation plus simple modeling assumptions. They did not discuss the case of Kubo's stochastic oscillator. However, we think that their approach when applied to this problem will result in the same type of structure as that found in the previous section.

We believe that the results obtained in this paper can also be useful in the field of computer simulation of generalized Brownian motion. In such a stimulating field of research increasing interest is being devoted to the computer simulation of stochastic equations of motion.[30–32] In the non-Markoffian case the theory illustrated in Section 2 is a quite useful tool,[31]

provided that the continued fraction giving the "memory kernel" is not so slowly convergent as to result in a multidimensional Langevin equation of an enormous size. In some cases, unfortunately, as shown by Balucani et al.,[33] a truncation of Tokuyama–Mori kind has to be used for obtaining a good agreement with the "experimental" results. By using the theory developed in the present paper it is an easy matter to find the generalized continued fraction expansion which corresponds to the mathematical model used in Ref. 33. If we assume that the nth-order "state" of the standard Mori chain "feels" the remaining part of this chain acting as a "multiplicative" stochastic process, it is straightforward to show that the Laplace transform of $\varphi(t)$, Eq. (2.2), can be written as follows:

$$\hat{\varphi}(z) = \cfrac{\Delta_1^2}{z - i\omega_1 + \cfrac{\Delta_2^2}{z - i\omega_2 + \ddots + \cfrac{\Delta_n^2}{z - i\omega_n + \hat{\varphi}_n(z)}}} \tag{5.1}$$

where

$$\hat{\varphi}_n(z) = \cfrac{\Delta^2}{z + \gamma + \cfrac{2\Delta^2}{z + 2\gamma + \cfrac{3\Delta^2}{z + 3\gamma + \ddots}}} \tag{5.2}$$

The SLE theory would afford the suggestion of applying the new approach of Refs. 30–32 to the following set of differential equations:

$$\begin{aligned}
\frac{du_0}{dt} &= \Delta_1 u_0(t) \\
\frac{du_1}{dt} &= -\Delta_1 u_0(t) + \Delta_2 u_2(t) \\
&\vdots \\
\frac{du_n}{dt} &= -\Delta_n u_{n-1}(t) + \omega(t) u_n(t) \\
\frac{d\omega}{dt} &= -\gamma\omega(t) + f_\omega(t)
\end{aligned} \tag{5.3}$$

where $f_\omega(t)$ can be assumed to be a Gaussian white noise. However, as shown in Appendix A, the higher-order correlation functions $C_{u_0}^{(n)}(t)$, Eq. (4.8), would not be correctly reproduced.

The interesting suggestion of the present paper is that of using the following set of differential equations:

$$\begin{aligned}
\frac{du_0}{dt} &= \Delta_1 u_0(t) \\
&\vdots \quad\quad \vdots \\
\frac{du_n}{dt} &= -\Delta_n u_{n-1}(t) + \Delta u_n(t) \\
\frac{du_{n+1}}{dt} &= -\Delta u_n(t) + \sqrt{2}\,\Delta u_{n+2}(t) \\
\frac{du_{n+2}}{dt} &= -\gamma u_{n+2} - \sqrt{2}\,\Delta u_{n+1}(t) + \sqrt{3}\,\Delta u_{n+3} + f_1(t) \\
\frac{du_{n+3}}{dt} &= -2\gamma u_{n+3} - \sqrt{3}\,\Delta u_{n+2}(t) + \sqrt{4}\,\Delta u_{n+4} + f_2(t) \\
&\cdots
\end{aligned} \tag{5.4}$$

where the $f_r(t)$ are Gaussian stochastic functions satisfying

$$(f_r(t), f_r^*) = 2r\gamma\delta(t) \tag{5.5}$$

We hope to assess in a future investigation whether the basic idea of this paper can be successfully applied to the computer simulation of nonlinear nonequilibrium statistical processes.[32]

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APPENDIX A

Let us consider the stochastic process defined by

$$\frac{du}{dt} = i\omega(t)u(t) \tag{A.1}$$

where $\omega(t)$ is a stochastic variable of Gaussian kind defined by its moments as follows:

$$\langle \omega(t_1) \rangle = 0 \tag{A.2}$$

$$\langle \omega(t_1)\omega(t_2) \rangle = \Delta^2 e^{-\gamma(t_1 - t_2)} \qquad (t_1 \geqslant t_2) \tag{A.2'}$$

$$\langle \omega(t_1)\omega(t_2)\omega(t_3) \rangle = 0 \qquad (t_1 \geqslant t_2 \geqslant t_3) \tag{A.2''}$$

$$\begin{aligned} \langle \omega(t_1)\omega(t_2)\omega(t_3)\omega(t_4) \rangle = \Delta^4 \{ & e^{-\gamma(t_1 - t_2)} e^{-\gamma(t_3 - t_4)} \\ & + e^{-\gamma(t_1 - t_3)} e^{-\gamma(t_2 - t_4)} \\ & + e^{-\gamma(t_1 - t_4)} e^{-\gamma(t_2 - t_3)} \} \qquad (t_1 \geqslant t_2 \geqslant t_3 \geqslant t_4) \end{aligned} \tag{A.2'''}$$

and so on.

The basic idea of Kubo's theory is as follows.[34,35] Equation (A.1) is integrated to

$$u(t) = \exp\left\{ i \int_0^t \omega(t_1)\, dt_1 \right\} u(0) \tag{A.3}$$

When the initial condition $u(0)$ is not a random one, the correlation function $\langle u(t)u(0) \rangle$ is proportional to the stochastic average of the exponential of Eq. (A.3). If the latter one, in turn, is developed in cumulants, we obtain

$$\langle u(t)u(0) \rangle / \langle u(0)u(0) \rangle = \exp\left\{ \sum_{m=1}^{\infty} \frac{(i)^m}{m!} \int_0^t \cdots \int_0^t \langle\langle \omega(t_1) \cdots \omega(t_n) \rangle\rangle \right\} \tag{A.4}$$

When Eqs. (A.2) are satisfied the only surviving cumulant is the following:

$$\langle\langle \omega(t_1)\omega(t_2) \rangle\rangle = \Delta^2 e^{-\gamma(t_1 - t_2)} \tag{A.5}$$

Then, from Eq. (A.4) we obtain

$$\langle u(t)u(0) \rangle / \langle u(0)u(0) \rangle = \exp\left[-(\Delta^2/\gamma)(e^{-\gamma t} - 1 + \gamma t) \right] \tag{A.6}$$

Since the equation of motion for u^n $(n > 0)$,

$$\frac{d}{dt} u^n = ni\omega(t)u^n(t) \tag{A.7}$$

can be derived from Eq. (A.1) by replacing $\omega(t)$ with $n\omega(t)$, the kind of approach which led us to Eq. (A.6) when applied to Eq. (A.7) results in

$$\langle u^n(t)u^n(0)\rangle/\langle u^n(0)u^n(0)\rangle = \exp\left[-(n^2\Delta^2/\gamma)(e^{-\gamma t} - 1 + \gamma t)\right] \quad \text{(A.8)}$$

The physical consequences of Eq. (A.7) are quite disappointing. When $t \to \infty$, $\langle u^n(t)u^n(0)\rangle$ vanish also for even values of n. In the case where $u(t)$ was the last variable of a standard Mori chain such as that resulting in the set of differential equations of Eq. (5.6), this undesired feature would spread along the chain and would affect even the variable of interest.

APPENDIX B

Let us define the variable of interest $\mathbf{U}$ as follows:

$$\mathbf{U} \equiv \begin{bmatrix} U_0 \\ U_1 \\ \vdots \\ U_n \end{bmatrix} \quad \text{(B.1)}$$

where

$$U_i \equiv f_i(f_i, f_i^*)^{-1/2} \quad \text{(B.2)}$$

and the f_i are defined by Eq. (2.12). By following the standard approach of Ref. 10, we obtain

$$\frac{d}{dt}\mathbf{U}(t) = i\boldsymbol{\Omega}(t)\cdot\mathbf{U}(t) - \int_0^t \boldsymbol{\Phi}(t-s)\cdot\mathbf{U}(s) + \mathbf{F}(t) \quad \text{(B.3)}$$

where

$$i\boldsymbol{\Omega} \equiv (iL\mathbf{U}, \mathbf{U}^*)\cdot(\mathbf{U}, \mathbf{U}^*)^{-1} \quad \text{(B.4)}$$

$$\boldsymbol{\Phi}(t) \equiv (\mathbf{F}(t), \mathbf{F}^*)\cdot(\mathbf{U}, \mathbf{U}^*)^{-1} \quad \text{(B.5)}$$

$$\mathbf{F}(t) \equiv e^{tQiL}QiL\mathbf{U}, \qquad \mathbf{F} \equiv \mathbf{F}(0) \quad \text{(B.6)}$$

We remark that, since the variables U_r are orthonormal,

$$Q = 1 - P = \prod_{r=0}^{\infty} Q_r \quad \text{(B.7)}$$

where $Q_r \equiv 1 - P_r$ and $P_r g \equiv (g, U_r^*)U_r$. As a consequence, the "random" force $\mathbf{F}(t)$ appears to be endowed with the following form:

$$\mathbf{F}(t) = \text{Col}\left[0, \ldots, e^{iQLt}iQLU_n\right] \quad \text{(B.8)}$$

We also point out that the orthonormal nature of the variables U_r is expressed by the following matrix equation:

$$(\mathbf{U}, \mathbf{U}^*) = \mathbf{I} \tag{B.9}$$

where $\mathbf{I}$ is the identity operator. Then, from Eqs. (B.8), (B.9), and (B.5), we obtain

$$\mathbf{\Phi}(t) = \begin{bmatrix} 0 & \cdots & 0 \\ \vdots & & \vdots \\ 0 & \cdots & \psi_n(t) \end{bmatrix} \tag{B.10}$$

where

$$\psi_n(t) = (U_{n+1}(t), U_{n+1}^*) = \left(e^{iQLt} iQLU_n, (QiLU_n)^*\right) \tag{B.10'}$$

By using the definitions of Eqs. (2.12)–(2.14′) and exploiting the Hermitian property of the projection operators P_r, we obtain

$$i(LU_j, U_{j+1}) = \Delta_{j+1} \tag{B.11}$$

$$i(LU_{j+1}, U_j) = -\Delta_{j+1} \tag{B.11'}$$

From Eqs. (B.2) and (B.10′) we also obtain that

$$\psi_n(t) = \varphi_n(t) \tag{B.12}$$

We recall that $\varphi_n(t)$ has been defined by Eq. (2.9). If we assume that

$$\varphi_n(t) = 2\gamma_n \delta(t) \tag{B.13}$$

we have

$$\hat{\varphi}_n(z) = \gamma_n \tag{B.14}$$

which coincides with Eq. (2.2‴). Then, from Eq. (B.3) by using Eqs. (B.12)–(B.14) we obtain Eq. (2.2′).

We can now remark that Eq. (2.2′) belongs to the family of the multidimensional Langevin equations studied by Fox and Uhlenbeck.[21] The result of this research has been reviewed by Fox.[3] The stochastic force $\mathbf{F}(t)$, Eq. (2.2′), has been assumed to be a Gaussian stochastic force with the stochastic properties

$$\langle F_i(t) \rangle = 0, \qquad \langle F_i(t) F_j(s) \rangle = 2Q_{ij}\delta(t-s) \tag{B.15}$$

We recall that the symbol $\langle \cdots \rangle$ denotes a stochastic averaging.[3] In Ref. 3 it is shown that a multidimensional Langevin equation such as Eq. (2.2′) has to satisfy a fluctuation–dissipation theorem expressed by the following equation [Eq. (I.2.16) of Ref. 3]

$$\mathbf{\Gamma E}^{-1} + \mathbf{E}^{-1}\mathbf{\Gamma}^{\dagger} = -2\mathbf{Q} \tag{B.16}$$

where the matrix **E** is defined as follows:

$$(\mathbf{E}^{-1})_{ij} \equiv \{u_i, u_j^*\} \tag{B.17}$$

and $\{\cdots\}$ denotes[3] an average on the equilibrium distribution of the stochastic variables u_i. If we identify $\{u_i, u_j^*\}$ with (U_i, U_j^*), by using Eq. (B.9) we have

$$\mathbf{E}^{-1} = (\mathbf{U}, \mathbf{U}^*) = \mathbf{I} \tag{B.18}$$

As a consequence, Eq. (2.2^V) completely agrees with Eq. (B.16).

Since our multidimensional Langevin equation satisfies the fluctuation–dissipation theorem of the theory developed by Fox and Uhlenbeck,[21] we can associate it with the Fokker–Planck equation found by them themselves [Eq. (I.2.40) of Ref. 3]. The normalized variables used in this appendix can be replaced by the usual ones simply by substituting u_i/q_i for u_i. Equation (2.15) thus is arrived at.

APPENDIX C

In this appendix we shall study a simple model of relaxation process, where the part of interest is sequentially coupled with two dissipation systems. A simple solution will be obtained by replacing a "reduced" model for the "complex" one. The "reduced" model will be shown to exhibit the same kind of structure as the Mori chain of variables of Section 2. However, the parameters corresponding to the real frequencies ω_i of Eqs. (2.8) and (2.17) will be shown to contain also an imaginary part.

First, consider the equation of motion

$$\frac{d}{dt}|e(t)\rangle = -i\mathcal{H}|e(t)\rangle \qquad (|e(0)\rangle = |e\rangle, \langle e|e\rangle = 1) \tag{C.1}$$

where $\mathcal{H}$ is the quantum-mechanical Hamiltonian defined by

$$\mathcal{H} \equiv |e\rangle\epsilon_e\langle e| + \sum_m |m\rangle\epsilon_m\langle m| + \sum_m \{|e\rangle v_m\langle m| + |m\rangle v_m\langle e|\} \tag{C.1'}$$

This kind of model Hamiltonian has widely been applied to the field of molecular radiationless decays. For a deeper understanding, the interested reader can consult several excellent reviews.[36] A reason for dealing here with this model has also to be seen in our desire to shed further light on the physical meaning of the Markoffian assumption of Section 2. Let us consider the case when the m manifold is so dense as to result in the same effects as a continuum set of states $\{|\epsilon\rangle\}$. This is just what is called Friedrichs model,[37] a simple scheme widely applied for studying nonequilibrium processes such as spontaneous emission.[38,39]

Equation (C.1) is of the same kind as Eq. (2.1). We can thus apply the standard Mori approach to the "variable" $|e(t)\rangle$. The scalar product between the two variables f and g used in Section 2 has to be replaced by the usual scalar product $\langle g|f\rangle$ between the two quantum-mechanical states $|f\rangle$ and $|g\rangle$. We have, for example,

$$|f_1\rangle = \int d\epsilon\, |\epsilon\rangle\langle\epsilon|V|e\rangle \tag{C.2}$$

$$\varphi(t) \equiv \langle f_1|f_1(t)\rangle = \int_{-\infty}^{+\infty} d\epsilon\, |\langle e|V|\epsilon\rangle|^2 e^{-i\epsilon t} \tag{C.3}$$

In the case where the coupling V is assumed to have the following Lorentzian form:

$$|\langle e|V|\epsilon\rangle|^2 \equiv A/\{(\epsilon - \epsilon_e)^2 + \gamma^2\} \tag{C.4}$$

we obtain

$$\hat{\varphi}(z) = \Delta_1^2/(z + i\epsilon_e + \gamma) \tag{C.5}$$

where

$$\Delta_1^2 = A\pi/\gamma \tag{C.6}$$

Therefore the infinite continued fraction expansion of Eq. (2.8) is shown to result in a truncated form, Eq. (C.5), which, in the case of the Lorentzian interaction of Eq. (C.4), is an exact expression for the Laplace transform of the "memory kernel" $\varphi(t)$.

Let us take into account the case where a second dissipation manifold is present. We have thus the following Hamiltonian:

$$\mathcal{H} \equiv |e\rangle\epsilon_e\langle e| + \sum_m |m\rangle\epsilon_m\langle m| + \sum_m \{|e\rangle v_m\langle m| + |m\rangle v_m\langle e|\}$$
$$+ \sum_n |n\rangle\epsilon_n\langle n| + \sum_{nm} \{|n\rangle v_{nm}\langle m| + |m\rangle v_{nm}\langle n|\} \tag{C.7}$$

This model Hamiltonian has widely been applied for studying, for instance, the problem of vibrational relaxation[40] and that of multiphoton molecular dissociation.[41] We shall assume that interference effects among the states $|m\rangle$ can be neglected. The Markoffian assumption on the n–m interaction is avoided: We assume that any state $|m\rangle$ is coupled with the second dissipation manifold by an interaction of the same form as that of Eq. (C.4). The width of this Lorentzian n–m coupling is denoted by γ_2. By using the results obtained in the first part of this section, we can replace Eq. (C.7) with the following non-Hermitian Hamiltonian:

$$\mathcal{H} \equiv |e\rangle\epsilon_e\langle e| + \sum_m |m\rangle\epsilon_m\langle m| + \sum_m |d(m)\rangle(\epsilon_m - i\gamma_2)\langle d(m)|$$
$$+ \sum_m (|e\rangle\langle m| + |m\rangle\langle e|)v_m + \sum_m (|m\rangle\langle d(m)| + |d(m)\rangle\langle m|)v_2 \tag{C.8}$$

This is equivalent to writing the memory function $\varphi(t)$ as follows:

$$\varphi(t) = \sum_m v_m^2 \langle m|e^{-i\mathcal{H}''t}|m\rangle e^{-i\epsilon_m t}$$

$$= \sum_m v_m^2 \left[(\langle \pi_1 | m\rangle)^2 e^{-iE_{\pi_1}t} + (\langle \pi_2 | m\rangle)^2 e^{-iE_{\pi_2}t} \right] e^{-i\epsilon_m t} \qquad \text{(C.9)}$$

where $|\pi_1\rangle$, $|\pi_2\rangle$ and E_{π_1}, E_{π_2} are the eigenstates and the eigenvalues, respectively, of the Hamiltonian $\mathcal{H}''$ defined by

$$\mathcal{H}'' \equiv \begin{pmatrix} 0 & v_2 \\ v_2 & -i\gamma_2 \end{pmatrix} \qquad \text{(C.10)}$$

By assuming again that the e–m coupling is of Lorentzian kind with width γ_1, $\varphi(t)$ can be written as follows ($\epsilon_M = \epsilon_e$; $\langle \pi_r | M\rangle = \langle \pi_r | m\rangle$, $r = 1, 2$):

$$\varphi(t) = v_1^2 e^{-i\epsilon_M t} e^{-\gamma_1 t} \left[(\langle \pi_1 | M\rangle)^2 e^{-iE_{\pi_1}t} + (\langle \pi_2 | M\rangle)^2 e^{-iE_{\pi_2}t} \right] \qquad \text{(C.11)}$$

The same "memory kernel" as that of Eq. (C.11) is obtained by using the following effective Hamiltonian ($\epsilon_N = \epsilon_M$):

$$\mathcal{H}' \equiv |e\rangle\epsilon_e\langle e| + |M\rangle(\epsilon_M - i\gamma_1)\langle M| + |N\rangle(\epsilon_N - i[\gamma_1 + \gamma_2])\langle N|$$

$$+ v_1(|e\rangle\langle M| + |M\rangle\langle e|) + v_2(|M\rangle\langle N| + |N\rangle\langle M|) \qquad \text{(C.12)}$$

The standard Mori theory cannot account for this kind of chain. Infinite "virtual" states are thus required to simulate the presence of the damping γ_1 on the intermediate state $|M\rangle$.

APPENDIX D

We shall study a general equation of motion of the following kind:

$$\frac{d}{dt} A(t) = \mathfrak{L} A(t) \qquad \text{(D.1)}$$

where A can represent either a classical variable or a quantum-mechanical state. The latter case concerns the field of investigation alluded to in Appendix C, to which the result of the present study can thus be extended. The corresponding dynamical operator $\mathfrak{L}$ is defined as follows:

$$\mathfrak{L} \equiv -i\mathcal{H} \qquad \text{(D.1')}$$

where $\mathcal{H}$ can be an effective Hamiltonian of non-Hermitian nature such as those of Appendix C.

In the classical case

$$\mathfrak{L} = iL \qquad \text{(D.1'')}$$

where L can be the "effective Liouville" operator built up in Section 3. We shall use a "quantum-mechanical" formalism even for dealing with the classical case. Thus, the scalar product between two classical variables α and β, denoted in Section 2 by (α, β^*), will be given the new symbol $\langle \beta | \alpha \rangle$.

The basic idea of the following theoretical development, the aim of which is to extend the Mori approach[10,11] to the non-Hermitian case, consists in using a biorthogonal set of "states" rather than the standard basis set of Section 2. We shall build up a chain of "states," the first "state" of that chain being

$$|f_0\rangle = |A\rangle \tag{D.2}$$

Then we shall define the usual zero-order projection operator

$$P_0 \equiv |f_0\rangle\langle f_0|/\langle f_0|f_0\rangle \tag{D.3}$$

The "state" $|f_1\rangle$ is defined by

$$|f_1\rangle \equiv \mathfrak{L}_1|f_0\rangle \tag{D.4}$$

where

$$\mathfrak{L}_1 \equiv (1 - P_0)\mathfrak{L}_0 \tag{D.4'}$$

We define, together with the state $|f_1\rangle$, the corresponding left "state" $\langle \tilde{f}_1|$,

$$\langle \tilde{f}_1| \equiv \langle f_0|\mathfrak{L}_0(1 - P_0) \tag{D.5}$$

In the Hermitian case

$$\langle \tilde{f}_1| = -\langle f_1| \tag{D.6}$$

In general, $\langle \tilde{f}_1|$ is completely different from the usual dual state $\langle f_1|$ associated with $|f_1\rangle$. The projection operator P_1 will be defined as follows:

$$P_1 \equiv |f_1\rangle\langle \tilde{f}_1|f_1\rangle^{-1}\langle \tilde{f}_1| \tag{D.7}$$

The idempotent property is conserved, whereas the Hermitian one is lost. In general, in fact, $P_1 \neq P_1^\dagger$. The $(n + 1)$th-order "state" is defined by

$$|f_{k+1}\rangle \equiv \mathfrak{L}_{k+1}|f_k\rangle \tag{D.8}$$

where

$$\mathfrak{L}_{k+1} \equiv (1 - P_k)\mathfrak{L}_k \tag{D.9}$$

and

$$P_k \equiv |f_k\rangle\langle \tilde{f}_k|f_k\rangle^{-1}\langle \tilde{f}_k| \tag{D.10}$$

The corresponding left state is given by

$$\langle \widetilde{f_{k+1}}| \equiv \langle \tilde{f}_k|\mathfrak{L}_0(1 - P_0)\cdots(1 - P_k) \tag{D.11}$$

We have thus defined a biorthogonal basis set of vectors, i.e., a set of vectors satisfying the following orthogonality condition:

$$\langle \tilde{f}_{k'} | f_k \rangle = \delta_{kk'} \tag{D.12}$$

The time evolution of the state $|f_k\rangle$ as resulting from the interaction with the space spanned by $|f_{k+1}\rangle$, $|f_{k+2}\rangle$ and so on, is

$$|f_k(t)\rangle \equiv e^{\mathcal{L}_k t}\mathcal{L}_k |f_k\rangle \tag{D.13}$$

so that

$$\frac{d}{dt}|f_k(t)\rangle = \mathcal{L}_k |f_k(t)\rangle \tag{D.13'}$$

We can also write

$$|f_k(t)\rangle = P_k|f_k\rangle + (1 - P_k)|f_k(t)\rangle = \Phi_k(t)|f_k\rangle + |f_k'(t)\rangle \tag{D.14}$$

where

$$\Phi_k(t) \equiv \langle \tilde{f}_k | f_k(t)\rangle / \langle \tilde{f}_k | f_k \rangle \tag{D.15}$$

and

$$|f_k'(t)\rangle \equiv (1 - P_k)|f_k(t)\rangle \tag{D.16}$$

By using Eqs. (D.16), (D.13'), and (D.14), we obtain

$$\frac{d}{dt}|f_k'(t)\rangle = (1 - P_k)\mathcal{L}_k |f_k'(t)\rangle + \Phi_k(t)|f_{k+1}\rangle \tag{D.17}$$

which results in

$$|f_k'(t)\rangle = \int_0^t ds\, \Phi_k(s)|f_{k+1}(t - s)\rangle \tag{D.18}$$

Equation (D.14) can thus be written as follows:

$$|f_k(t)\rangle = \Phi_k(t)|f_k\rangle + \int_0^t ds\, \Phi_k(s)|f_{k+1}(t - s)\rangle \tag{D.14'}$$

We focus now our attention on

$$\langle \tilde{f}_k(t)| \equiv \langle \tilde{f}_k | \tilde{\mathcal{L}}_k e^{\tilde{\mathcal{L}}_k t} \tag{D.19}$$

satisfying

$$\frac{d}{dt}\langle \tilde{f}_k(t)| = \langle \tilde{f}_k(t)|\tilde{\mathcal{L}}_k \tag{D.20}$$

where

$$\tilde{\mathcal{L}}_k \equiv \tilde{\mathcal{L}}_{k-1}(1 - P_{k-1}) \qquad (\tilde{\mathcal{L}}_0 \equiv \mathcal{L}_0) \tag{D.21}$$

By following the same approach as that which led us to Eq. (D.14'), we

obtain

$$\langle \tilde{f}_k(t)| = \tilde{\Phi}_k(t)\langle \tilde{f}_k| + \int_0^t ds\, \tilde{\Phi}_k(s)\langle \tilde{f}_{k+1}(t-s)| \tag{D.22}$$

where

$$\tilde{\Phi}_k(t) \equiv \langle \tilde{f}_k(t)\,|f_k\rangle \cdot \langle \tilde{f}_k\,|f_k\rangle^{-1} \tag{D.23}$$

By using the biorthogonal properties of our basis set, after some algebra we obtain

$$\tilde{\Phi}_k(t) = \Phi_k(t) \tag{D.24}$$

and

$$\langle \tilde{f}_k(t)\,|f_{k+1}\rangle = \langle \tilde{f}_k|e^{\tilde{\mathcal{L}}_k t}|f_{k+1}\rangle = \langle \tilde{f}_k|e^{\mathcal{L}_k t}|f_{k+1}\rangle \tag{D.25}$$

From Eq. (D.13) we have

$$\frac{d}{dt}|f_k(t)\rangle = \mathcal{L}_k|f_k(t)\rangle = e^{\mathcal{L}_k t}\mathcal{L}_k|f_k\rangle \tag{D.26}$$

which by applying the property $1 = (1 - P_k) + P_k$ results in

$$\frac{d}{dt}|f_k(t)\rangle = \lambda_k e^{\mathcal{L}_k t}|f_k\rangle + e^{\mathcal{L}_k t}|f_{k+1}\rangle \tag{D.27}$$

where

$$\lambda_k \equiv \langle \tilde{f}_k|\mathcal{L}_k|f_k\rangle\langle \tilde{f}_k\,|f_k\rangle^{-1} \tag{D.28}$$

By applying again the resolution of unity to the second term on the right-hand side of Eq. (D.27), we have

$$\frac{d}{dt}|f_k(t)\rangle = \lambda_k|f_k(t)\rangle + |f_k\rangle\langle \tilde{f}_k|e^{\mathcal{L}_k t}|f_{k+1}\rangle\langle \widetilde{f_k}\,|f_k\rangle^{-1} + (1 - P_k)e^{\mathcal{L}_k t}|f_{k+1}\rangle \tag{D.29}$$

Equation (2.25) allows us to replace Eq. (2.29) by

$$\frac{d}{dt}|f_k(t)\rangle = \lambda_k|f_k(t)\rangle + |f_k\rangle\langle \tilde{f}_k(t)\,|f_{k+1}\rangle\langle \tilde{f}_k\,|f_k\rangle^{-1} + (1 - P_k)e^{\mathcal{L}_k t}|f_{k+1}\rangle \tag{D.30}$$

Inserting Eq. (D.22) for $\langle \tilde{f}_k(t)|$ into Eq. (D.30), we obtain

$$\frac{d}{dt}|f_k(t)\rangle = \lambda_k|f_k(t)\rangle - |f_k\rangle\int_0^t ds\, \Phi_{k+1}(t-s)\Phi_k(s)\Delta_{k+1}^2 + (1 - P_k)e^{\mathcal{L}_k t}|f_{k+1}\rangle \tag{D.31}$$

where

$$\Delta_{k+1}^2 \equiv -\langle \tilde{f}_{k+1}\,|f_{k+1}\rangle\langle \tilde{f}_k\,|f_k\rangle^{-1} \tag{D.32}$$

By multiplying Eq. (2.31) on the left by $\langle \tilde{f}_k|$, we obtain

$$\frac{d}{dt}\Phi_k(t) = \lambda_k \Phi_k(t) - \Delta_{k+1}^2 \int_0^t ds\, \Phi_k(s)\Phi_{k+1}(t-s) \tag{D.33}$$

By Laplace transforming Eq. (D.33), we have

$$\hat{\Phi}_k(z) = \Phi_k(0)/\left[z - \lambda_k + \Delta_{k+1}^2 \hat{\Phi}_{k+1}(z)\right] \tag{D.34}$$

As a consequence, we arrive at the following continued fraction:

$$\hat{\Phi}_o(z) = \cfrac{1}{z - \lambda_0 + \cfrac{\Delta_1^2}{z - \lambda_1 + \cfrac{\Delta_2^2}{z - \lambda_2 + \cfrac{\Delta_3^2}{\ddots \cfrac{\Delta_{n-1}^2}{z - \lambda_{n-1} + \Delta_n^2 \hat{\Phi}_n(z)}}}}} \tag{D.35}$$

which is more general than that by Mori, Eq. (2.8), since the expansion parameters λ_i and Δ_i are now complex numbers containing, in general, both a real and an imaginary part.

By Laplace-transforming Eq. (D.14′), we get

$$|\hat{f}_k(z)\rangle = |f_k\rangle \hat{\Phi}_k(z) + |\hat{f}_{k+1}(z)\rangle \hat{\Phi}_k(z) \tag{D.36}$$

Let us replace Eq. (2.34) into Eq. (2.36). We then obtain

$$|\hat{f}_k(z)\rangle = \left[|f_k\rangle + |\hat{f}_{k+1}(z)\rangle\right]\left(z - \lambda_k + \Delta_{k+1}^2 \hat{\Phi}_{k+1}(z)\right)^{-1} \tag{D.37}$$

which is the Laplace transform of the following equation:

$$\frac{d}{dt}|f_k(t)\rangle = \lambda_k |f_k(t)\rangle - \int_0^t ds\, |f_k(s)\rangle \varphi_k(t-s) + |f_{k+1}(t)\rangle \tag{D.38}$$

where

$$\varphi_k(t) \equiv \Delta_{k+1}^2 \Phi_{k+1}(t) \tag{D.39}$$

In the Hermitian case, for $k = 0$ the previous equation coincides with the generalized Langevin equation of Eq. (2.2).

As in Appendix B, it is useful to normalize the states $|f_m\rangle$. In the present case, we have to define a biorthonormal basis set of "states." They are defined as follows:

$$|m\rangle \equiv |f_m\rangle / \langle \tilde{f}_m | f_m \rangle^{1/2} \tag{D.40}$$

$$\langle \tilde{m}| \equiv \langle \tilde{f}_m| / \langle \tilde{f}_m | f_m \rangle^{1/2} \tag{D.40'}$$

As in Appendix B, it is then straightforward to show that

$$\langle \widetilde{m+1} | \mathcal{L}_0 | m \rangle = i\Delta_{m+1} \tag{D.41}$$

$$\langle \tilde{m} | \mathcal{L}_0 | m+1 \rangle = i\Delta_{m+1} \tag{D.41'}$$

$$\langle \tilde{m} | \mathcal{L}_0 | m \rangle = \lambda_m \tag{D.41''}$$

which are the only nonvanishing matrix elements of $\mathcal{L}_0$. $\mathcal{L}_0$ can also be given in the following equivalent form:

$$\langle \widetilde{m+1} | \mathcal{L}_0 | m \rangle = \Delta_{m+1} \tag{D.42}$$

$$\langle \tilde{m} | \mathcal{L}_0 | m+1 \rangle = -\Delta_{m+1} \tag{D.42'}$$

$$\langle \tilde{m} | \mathcal{L}_0 | m \rangle = \lambda_m \tag{D.42''}$$

which leads to Eq. (4.1).

APPENDIX E

In the present appendix we shall show that Eqs. (2.18) and (2.2′) lead to the same continued fraction for the variable of interest. In other words, we shall show that

$$(u_0(t), u_0^*) = (U_0(t), U_0^*) \tag{E.1}$$

In order to obtain this result we shall apply the generalized Mori theory of Appendix D.

First of all, we shall define a suitable zero-order basis set. By using the Hermite polynomials $\mathrm{He}_n(x)$,[42] we build the following "state":

$$p_m(x) \equiv e^{-x^2/2} \mathrm{He}_m(x) / R_m \tag{E.2}$$

where

$$R_m \equiv \left[m! (2\pi)^{1/2} \right]^{1/2} \tag{E.3}$$

For simplicity, we assumed q_n, Eq. (2.15′), to be equal to 1. The "states" $|p_m\rangle$ are an orthonormalized basis set provided that the scalar product $\langle p_n | p_m \rangle$ is defined as follows[43]:

$$\langle p_n | p_m \rangle = \int_{-\infty}^{+\infty} dx \, f_w(x) p_n(x) p_m(x) \tag{E.4}$$

where

$$f_w(x) \equiv e^{x^2/2} \tag{E.5}$$

We shall use the following important properties[43]:

$$\frac{d}{dx} |p_n\rangle = -(n+1)^{1/2} |p_{n+1}\rangle \tag{E.6}$$

$$x|p_m\rangle = (m)^{1/2} |p_{m-1}\rangle + (m+1)^{1/2} |p_{m+1}\rangle \tag{E.7}$$

Our basis set will then be defined by the following kind of direct products:

$$|n_0 n_1 n_2 \cdots\rangle \equiv |p_{n_0}(u_0)\rangle |p_{n_1}(u_1)\rangle |p_{n_2}(u_2)\rangle \cdots \tag{E.8}$$

Let us take into account the case where

$$\Gamma_u = -\Delta_1\left(\frac{\partial}{\partial u_0}u_1 - \frac{\partial}{\partial u_1}u_0\right) + \gamma_1\left(\frac{\partial}{\partial u_1}u_1 + \frac{\partial^2}{\partial u_1^2}\right) \tag{E.9}$$

The zero-order state of our generalized chain is defined as follows:

$$|f_0\rangle \equiv u_0|00\rangle \tag{E.10}$$

By applying Eq. (E.7) we can also write

$$|f_0\rangle = |10\rangle \tag{E.11}$$

By using Eqs. (E.6) and (E.7), $|f_1\rangle$, Eq. (D.4), can be written as follows:

$$|f_1\rangle = (1 - P_0)\Gamma_u|10\rangle = -\Delta_1|01\rangle \tag{E.12}$$

Equation (E.12) results in the same continued fraction (truncated at first order) as Eq. (2.2′). In fact

$$\lambda_1 = \langle \tilde{f}_1|\Gamma_u|f_1\rangle \cdot \langle \tilde{f}_1|f_1\rangle^{-1} = -\gamma_1 \tag{E.13}$$

The extension of this demonstration to the nth order is straightforward. For example, in the case where

$$\Gamma_u = -\Delta_1\left(\frac{\partial}{\partial u_0}u_1 - \frac{\partial}{\partial u_1}u_0\right) - \Delta_2\left(\frac{\partial}{\partial u_1}u_2 - \frac{\partial}{\partial u_2}u_1\right) + \gamma_2\left(\frac{\partial}{\partial u_2}u_2 + \frac{\partial^2}{\partial u_2^2}\right) \tag{E.14}$$

we have

$$|f_1\rangle = -\Delta_1|010\rangle \tag{E.15}$$

In fact, the contribution of the first term on the right-hand side of Eq. (E.14) is the same as the corresponding one of the previous case, whereas the contribution of the second term, when applied to $|000\rangle$, vanishes. It is also straightforward to show that

$$\left(\frac{\partial}{\partial u_0}u_1 - \frac{\partial}{\partial u_1}u_0\right)|010\rangle = -|100\rangle = -|f_0\rangle \tag{E.16}$$

As a consequence, this contribution vanishes when the projection operator $1 - P_0$ is applied. We have thus

$$|f_2\rangle = \Delta_1\Delta_2|001\rangle \tag{E.17}$$

In general

$$|f_j\rangle = (-1)^j\Delta_1\Delta_2 \cdots \Delta_j|0(0), 0(1), \ldots, 1(j), \ldots\rangle \tag{E.18}$$

It is then evident that $(u_0(t), u_0^*)$ results in the same continued fraction expansion as $(U_0(t), U_0^*)$. This result can also be regarded as a further check of the multidimensional Fokker–Planck equation of Section 2, Eq. (2.15′).

In the case where the effective operator under discussion is given by that of Eq. (3.7) and Γ_Ω is that of Eq. (4.2), we can define $|f_0\rangle$ as follows:

$$|f_0\rangle \equiv a|p_0(\omega)\rangle \tag{E.19}$$

In general the scalar product between two states $|F\rangle = b|q_1(\omega)\rangle$ and $|G\rangle = c|q_2(\omega)\rangle$, where $q_1(\omega)$ and $q_2(\omega)$ are functions of the space spanned by the Hermite polynomials, is given by

$$\langle G|F\rangle \equiv (b, c^*)\langle q_1|q_2\rangle \tag{E.20}$$

where (b, c^*) is defined according to the nature of the part of interest of our physical system and $\langle q_1|q_2\rangle$ has to be derived by the definition of Eq. (E.4).

Then, from the generalized Mori theory of Appendix D we have

$$\begin{aligned} |f_1\rangle &= (1 - P_0)(i\omega + \Gamma_\omega)a|p_0(\omega)\rangle \\ &= (1 - P_0)ia\omega|p_0(\omega)\rangle = ia|p_1(\omega)\rangle \end{aligned} \tag{E.21}$$

In order to obtain Eq. (E.21) we used Eq. (E.7). Furthermore, we took into account the fact that $|p_0(\omega)\rangle$ is the equilibrium eigenstate of Γ_ω [$\Gamma_\omega|p_0(\omega)\rangle = 0$]. In general, by using Eq. (E.7) and the property $(1 - P_{n-1})|p_{n-1}\rangle = 0$ we have

$$|f_n\rangle = i^n a(n!)^{1/2}|p_n(\omega)\rangle \tag{E.22}$$

As a consequence (recall the definitions of Δ_i^2's and λ_i's given in Appendix D)

$$\Delta_n^2 = n\Delta^2 \tag{E.23}$$

$$\lambda_n = -n\gamma \tag{E.24}$$

It is encouraging to remark that the continued fraction resulting from Eqs. (E.23) and (E.24) is the same as that provided by the SLE theory.[1]

In the present appendix we have used the "quantum-mechanical" formalism of the previous one. When translated into the "classical" notations of Section 2, the autocorrelation function of the variable of interest reads [we are now dealing again with the simple case of Eq. (2.18)]

$$\begin{aligned} (u_0(t), u_0^*) &\equiv \int d\mathbf{u}\, u_0^* u_0(t) w_0(\mathbf{u})\, d\mathbf{u} \equiv \int d\mathbf{u}\, u_0^* \exp(\Gamma_{\mathbf{u}} t) u_0 \exp(-\Gamma_{\mathbf{u}} t) w_0(\mathbf{u}) \\ &= \int d\mathbf{u}\, u_0^* \exp(\Gamma_{\mathbf{u}} t) u_0 w_0(\mathbf{u}) \end{aligned} \tag{E.25}$$

where $w_0(\mathbf{u})$ is the equilibrium distribution of the multidimensional variable $\mathbf{u}$. We can remark, in fact, that the general expression for $|f_0\rangle$ is

$$|f_0\rangle \equiv u_0|000\cdots\rangle \tag{E.10'}$$

Therefore,

$$\begin{aligned}\langle f_0|f_0(t)\rangle &= \langle 000\cdots|u_0^* \exp(\Gamma_{\mathbf{u}}t)u_0|000\cdots\rangle \\ &= \int e^{u_0^2/2}e^{u_1^2/2}\cdots e^{-u_0^2/2}e^{-u_1^2/2}\cdots u_0^* \exp(\Gamma_{\mathbf{u}}t)u_0 w_0(\mathbf{u})\,d\mathbf{u} \\ &= \int u_0^* u_0(t) w_0(\mathbf{u})\,d\mathbf{u}\end{aligned} \tag{E.26}$$

For $t \neq 0$ $u_0(t)$, Eq. (E.25), has to be regarded as being an operator acting even on $w_0(\mathbf{u})$. For $t = 0$ the definition of scalar product on which Eq. (E.25) is based is the same as that of Eq. (2.19).

This scalar product, of course, has been defined in such a way as to agree with the stochastic approach,[35] which provides $[t = t_2 - t_1 \geqslant 0]$

$$\langle u_0^* u_0(t)\rangle \equiv \int d\mathbf{u}^{(1)}\,d\mathbf{u}^{(2)}\,P_2(\mathbf{u}^{(1)}, t_1; \mathbf{u}^{(2)}, t_2)u_0^{(1)*}u_0^{(2)}w_0(\mathbf{u}) \tag{E.27}$$

By recalling that[35]

$$P_2(\mathbf{u}^{(1)}, t_1; \mathbf{u}^{(2)}, t_2) = \exp(\Gamma_{\mathbf{u}^{(2)}}t)\delta(\mathbf{u}^{(1)} - \mathbf{u}^{(2)}) \tag{E.28}$$

we obtain

$$\begin{aligned}\langle u_0 u_0(t)\rangle &= \int d\mathbf{u}^{(2)}\,u_0^{(2)}\left[\exp(D_{\mathbf{u}^{(2)}}t)u_0^{(2)*}\right]w_0(\mathbf{u}^{(2)}) \\ &= \int d\mathbf{u}\,u_0^* \exp(\Gamma_{\mathbf{u}}t)u_0 w_0(\mathbf{u})\end{aligned} \tag{E.29}$$

Equation (E.25) agrees with the previous one. $D_{\mathbf{u}}$ is the adjoint to $\Gamma_{\mathbf{u}}$ defined by the scalar product of Eq. (2.19) with $w_0(\mathbf{u}) = 1$.

APPENDIX F

By Laplace transforming Eq. (3.8) we obtain

$$z\hat{u}_0(z) - u_0(0) = \lambda_0\hat{u}_0(z) + \Delta_1\hat{u}_1(z) + \hat{F}_0(z) \tag{F.1}$$

$$z\hat{u}_1(z) - u_1(0) = \lambda_1\hat{u}_1(z) - \Delta_1\hat{u}_0(z) + \Delta_2\hat{u}_2(z) + \hat{F}_1(z) \tag{F.2}$$

$$z\hat{u}_2(z) - u_2(0) = \lambda_2\hat{u}_2(z) - \Delta_2\hat{u}_1(z) + \Delta_3\hat{u}_3(z) + \hat{F}_2(z) \tag{F.3}$$

$$\cdots$$

If this chain could be truncated at the second order ($\Delta_2 = 0$), from Eq. (F.2)

we would obtain

$$\hat{u}_1(z) = (z - \lambda_1)^{-1}\left[u_1(0) - \Delta_1 \hat{u}_0(z) + \hat{F}_1(z)\right] \tag{F.4}$$

which, when replaced in Eq. (F.1), results in

$$z\hat{u}_0(z) - u_0(0) = \lambda_0 u_0(z) - \frac{\Delta_1^2}{(z - \lambda_1)}\hat{u}_0(z) + \hat{F}_0(z) + \frac{\left[\hat{F}_1(z) + u_1(0)\right]}{(z - \lambda_1)}\Delta_1 \tag{F.5}$$

Equation (F.5) is the Laplace transform of the following generalized Langevin equation:

$$\frac{d}{dt}u_0(t) = \lambda_0 u_0(t) - \int_0^t \varphi(t - s)u_0(s)\,ds + f(t) \tag{F.6}$$

where

$$\hat{\varphi}(z) = \frac{\Delta_1^2}{z - \lambda_1} \tag{F.7}$$

$$\hat{f}(z) = \hat{F}_0(z) + \frac{\left[\hat{F}_1(z) + u_1(z)\right]}{(z - \lambda_1)}\Delta_1 \tag{F.8}$$

If the truncation is made at the third order ($\Delta_3 = 0$), from Eq. (F.3) we have

$$\hat{u}_2(z) = (z - \lambda_2)^{-1}\left[u_2(0) - \Delta_2 \hat{u}_1(z) + \hat{F}_2(z)\right] \tag{F.9}$$

When replaced into Eq. (F.2), it results in

$$z\hat{u}_1(z) - u_1(0) = \lambda_1 \hat{u}_1(z) - \Delta_1 \hat{u}_0(z) + (z - \lambda_2)^{-1}\Delta_2\left[\hat{u}_2(0) + \hat{F}_2(z) - \Delta_2 \hat{u}_1(z)\right] + \hat{F}_1(z) \tag{F.10}$$

from which

$$\hat{u}_1(z) = \left(z - \lambda_1 + \frac{\Delta_2^2}{z - \lambda_2}\right)^{-1} \times \left\{u_1(0) - \Delta_1 \hat{u}_0(z) + \hat{F}_1(z) + \frac{\Delta_2}{(z - \lambda_2)}\left[u_2(0) + \hat{F}_2(z)\right]\right\} \tag{F.11}$$

This equation, in turn, provides for the generalized Langevin equation of Eq. (F.6) the "memory kernel" given by the inverse Laplace transform of

$$\hat{\varphi}_0(z) = \frac{\Delta_1^2}{z - \lambda_1 + \Delta_2^2/(z - \lambda_2)} \tag{F.12}$$

and the following Laplace transform of the stochastic force:

$$\hat{f}(z) = \hat{F}_0(z) + \frac{\Delta_1}{z - \lambda_1 + \Delta_2^2/(z - \lambda_2)} \times \left\{ \hat{F}_1(z) + u_1(0) + \frac{\Delta_2}{(z - \lambda_2)} \left[u_2(0) + \hat{F}_2(z) \right] \right\} \quad \text{(F.13)}$$

By applying the same method to the higher-order cases, it is straightforward to show that the exact expressions for $\hat{\varphi}(z)$ and $\hat{f}(z)$ are given by Eqs. (3.15) and (3.16), respectively.

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