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The Time-Local View of Nonequilibrium Statistical Mechanics. II. Generalized Langevin Equations

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On a semiphenomenological level, generalized Langevin equations are usually obtained by adding a random force (RF) term to macroscopic deterministic equations assumed to be known. Here this procedure is made rigorous by conveniently redefining the RF, which is shown to be colored noise weakly correlated with the observables at earlier times due to the finite lifetime of microscopic events. Corresponding fluctuation-dissipation theorems are derived. Explicit expressions for the spectral density of the fluctuations are obtained in a particularly simple form, with the deviation of the line shape from the Lorentzian being related most explicitly to the spectral density of the RF. Well-known low-frequency expressions and the Einstein relation of (generalized) Brownian motion theory are modified so as to include lifetime effects. New sum rules are obtained relating dissipative quantities to contour integrals (in the complex frequency domain) over spectral densities or corresponding response functions. The Heisenberg dynamics of a complete set of macroobservables is shown to be equivalent to a generalized Orstein–Uhlenbeck stochastic process which is a non-Markovian process due to the lifetime effects.

KEY WORDS: Statistical mechanics; Langevin equations; Heisenberg dynamics; spectral density; line shape; colored noise; Ornstein–Uhlenbeck process.

1. INTRODUCTION

Generalized Langevin equations (GLE) have proven in recent years to provide a valuable conceptual basis for the mesoscopic description of a broad variety of irreversible processes, particularly in far-from-equilibrium

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situations.[1] Originally, such an equation was given by Langevin for describing Brownian motion as

$$\dot{v}(t) = -\zeta v(t) + X(t) \tag{1.1}$$

where v denotes the velocity of the Brownian particle of unit mass, ζ is the friction constant, and x is the random force (RF), assumed to be white noise. Equation (1.1) was obtained by adding the RF term to the systematic (or macroscopic) part $\dot{v} = -\zeta v$ in order to simulate the fluctuations of v around its systematic or frictional motion.

This is a phenomenological procedure. Exact GLEs for an arbitrary set of observables $A(t)$, $\{A(t)\colon A_1(t),\ldots, A_n(t)\}$ have been derived by Mori[2] directly from statistical mechanics by means of his projection operator technique. These exact equations differ from Eq. (1.1) in that the noise is colored and the friction term is a retarded one. Mori's equations are of wide use and form one of the cornerstones of nonequilibrium statistical mechanics in the convolution picture (cf. Section 2).

The convolutionless version of the GLE has been derived subsequently by Tokuyama and Mori and others.[3,4] These equations have the general structure of Eq. (1.1), i.e., the friction term is time-local, with ζ depending on time, however. They have found much less application than their convolution counterpart due to their more complicated structure (see Section 3 for details).

The present paper proposes a third type of GLE [cf. Eq. (4.2)], which is of the same structure as Eq. (1.1), i.e., with ζ a constant (autonomous friction term). Thus, all of the effects due to the finite duration of the microscopic events that are responsible for the fluctuations of $A(t)$ are relegated to the colored noise character of the RF (cf. Section 4.). This new ansatz has a number of important consequences. In particular, it leads to very simple expressions for the spectral density of $A(t)$ and gives explicit account of the modifications of the Lorentzian line shape due to the coloredness of the noise (Section 6) and of the modification of the Einstein relation of Brownian motion theory due to lifetime effects (Section 7). From the conceptual point of view, it seems interesting that at all levels of observation we may identify Heisenberg dynamics with a generalized Ornstein–Uhlenbeck process (Section 8.).

The paper rests in several respects on results obtained in Ref. 4 (referred to as I in the following). In order to make the present paper as self-contained as possible, we note the following results. The object of central interest in I was the correlation matrix $C(t)$, which is given in Liouville space notation as

$$C(t) = \langle A(t) | A^{+} \rangle \tag{1.2}$$

where A is a row matrix of n elements and A^+ its Hermitian counterpart. It is assumed that A forms a complete set, so that it comprises all of the slow variables of the system. Then, the analytical continuation into the lower half-plane of the Laplace transform $\tilde{C}(z)$ of $C(t)$ reveals two groups of singularities, macroscopic and microscopic ones. These are situated a distance of order t_R^{-1} or t_c^{-1}, respectively, below the real axis, where t_R (t_c) is a characteristic time of the slow (rapid) variables. This leads, by means of the theory of residua, to the decomposition (valid for $t>0$)

$$C(t)=C^{(+)}(t)+C^{(m)}(t) \tag{1.3}$$

of C into a macroscopic and a microscopic part. We note that $C^{(m)}(t)$ decays on a time scale given by t_c,[2]

$$C^{(m)}(t)=0, \qquad t \gg t_c \tag{1.4}$$

whereas $C^{(+)}$ decays with scale t_R.

The convolutionless equation of motion for $C(t)$ is

$$\dot{C}(t)= -I(t)\, C(t) \tag{1.5a}$$

where

$$I(t)= -\dot{C}(t)\, C^{-1}(t) \tag{1.5b}$$

On the other hand, $C^{(+)}$ obeys the autonomous equations

$$\partial_t C^{(+)}(t)= -I^{(+)}C^{(+)}(t) \tag{1.6}$$

where

$$I^{(\pm)}= \lim_{t\to \pm\infty} I(t) \tag{1.7}$$

with $I^{(+)}$ the transport kernel of the autonomous macrodynamics obeyed by the aged system. As a consequence of the above we obtain[3]

$$C^{(+)}(t)=e^{-It}\,\Delta \tag{1.8}$$

where

$$C^{(+)}(0)=:\Delta= \lim_{t\to \infty} e^{It}C(t) \tag{1.9}$$

[2] Equation (1.4) is a symbolic notation including, e.g., exponential but not algebraic decay, which has no time scale.

[3] We drop here and in the following the $(+)$ sign wherever possible, i.e., $I=I^{(+)}$, $\Delta=\Delta^{(+)}$,....

and we will frequently use

$$\Delta = F + \Gamma \tag{1.10}$$

where

$$F = \langle A \mid A^{+} \rangle \tag{1.11}$$

is the variance matrix of the equilibrium fluctuations of A and

$$\Gamma = O(\xi), \qquad \xi = t_c/t_R \tag{1.12}$$

We will also need

$$\mathscr{L} = I\Delta \tag{1.13}$$

which plays the role of the Onsager kinetic coefficient in the irreversible thermodynamics to be developed in a subsequent paper. Note that $\mathscr{L}$ deviates from its usual definition by terms of order ξ due to Eqs. (1.10) and (1.12).

In concluding this section, we want to emphasize that the above results were not derived from first principles, from considering the spectrum of the Liouvillean, say. Instead, all we have shown in I is that the above results are fully consistent with the convolution picture. The results to be derived in the present paper rest on the same footing, so that, e.g., the decay properties of the noise term [cf. Eq. (4.5)] are valid if and only if the property (2.6) holds.

A second remak concerns the completeness of the set A. The decay of noise correlations over molecular times can hold only if A also contains all bilinear and higher combinations of some primitive slow variables as they are introduced in mode–mode coupling theory.

Thus, we actually are dealing with a nonlinear GLE. Reducing A to the set of the linear variables only leads to the appearance of a long-time tail (algebraic decay) in RF correlation functions much in the same way as obtained in mode–mode coupling theory for the memory kernel $M(t)$ of the convolution picture. This will be treated in more detail in a separate paper.

2. GENERALIZED LANGEVIN EQUATION IN THE CONVOLUTION PICTURE

One of the main achievements of the customary convolution picture (CP) approach to nonequilibrium statistical mechanics consists in the derivation of a generalized Langevin equation (GLE), i.e., in rewriting the Heisenberg equations of motion

$$\dot{A}_k = iLA_k, \qquad k = 1,\ldots, n \tag{2.1}$$

for our set $A = \{A_1, ..., A_n\}$ of observables into the exact GLE

$$\dot{A}(t) = -i\Omega A(t) - \int_0^t dt' \, M^{ir}(t') \, A(t-t') + f(t) \tag{2.2}$$

where $f(t)$ denotes the so-called random force (RF) given by

$$f(t) = i[\exp(+i\hat{Q}L\hat{Q}t)] \, \hat{Q}L\hat{P}A(0) \tag{2.3}$$

where the projection operators $\hat{P}$ and $\hat{Q}$, with $\hat{P} + \hat{Q} = 1$, were introduced in I. In Eq. (2.2)

$$\Omega = \langle A | \, L \, | A^+ \rangle \, F^{-1} \tag{2.4}$$

and

$$M^{ir}(t) = \langle f(t+s) \, | \, f^+(s) \rangle \, F^{-1}, \qquad \forall s \tag{2.5}$$

which is called the fluctuation-dissipation theorem of the second kind relating the random force to the generalized transport kernel $M(t)$.

Equation (2.2) is exactly valid for any system and choice of observables, its physical meaning deriving from the decay properties of $M(t)$, which in the symbolic notation introduced in Section 1 are given by

$$M(t) = 0, \qquad |t| \gg t_c \tag{2.6}$$

provided the set A is a complete one, i.e., that it comprises all slow (with characteristic time t_R) parameters of the system.

In view of Eq. (2.5), the RF is seen to be stationary. This is obvious already, since $f(t)$ obeys the equation of motion

$$\dot{f}(t) = +i\hat{Q}L\hat{Q}f(t) \tag{2.7}$$

for all times t. Thus, neither Eq. (2.1) nor Eq. (2.7) singles out any instant of time. The RF obeys

$$\langle f(t) \rangle_{\mathrm{eq}} = 0 \tag{2.8a}$$

and the orthogonality property

$$\langle f(t) \, | \, A^+(0) \rangle = 0, \qquad \forall t \tag{2.8b}$$

However, one easily convinces oneself that the orthogonality of Eq. (2.8b) is a nonstationary property in the sense that

$$\langle f(t_1) \, | \, A^+(t_2) \rangle = \varphi(t_1, t_2) \neq 0 \tag{2.8c}$$

in general. Thus, one may say the RF $f(t)$ is exactly orthogonal to (uncorrelated with) the observables A only at a single instant of time chosen arbitrarily at $t_2 = 0$.

We note without proof that for $t_1, t_2 > 0$ (<0) φ has, apart from terms of order ξ, the same properties as $Q^{(+)}(t_1 - t_2)$ ($Q^{(-)}$) as introduced in Section 4. In particular, f is strongly correlated with A for, say, $t_2 > t_1 \gg t_0$, where $\varphi \approx 2I \exp[-(t_2 - t_1) I]_F$ in the white noise limit.

The orthogonality property (2.8b) usually is considered essential for the reinterpretation of $f(t)$ as a random force (RF). The point of view taken in the present paper is different. We will argue below that certain correlations, such as expressed by Eq. (2.8c), between the observables and the RF are quite natural in realistic systems obeying the Heisenberg dynamics (2.1). Thus, there is no compelling physical reason for excluding such correlations at some particular instant of time, as in (2.8b). We will therefore drop this orthogonality requirement and thus obtain a theory that is time-local, completely stationary, and displays explicitly the existence of an autonomous macrodynamics that is hidden in the GLE (2.2). Before introducing this GLE, we will consider in the following section the GLE in the time-local picture as proposed earlier by several authors.

3. GENERALIZED LANGEVIN EQUATIONS IN THE TIME-LOCAL PICTURE (TLP)

The TLP analog of the GLE (2.2) has been obtained earlier by several authors in the framework of the so-called convolutionless projection operator method, the result being given usually as[3]

$$\dot{A}(t) = -I(t)\, A(t) + g(t) \tag{3.1}$$

where $I(t)$ was introduced in Eq. (1.5), but was also given as an explicit expression [cf. Eqs. (2.3)–(2.7) of Ref. 3] as

$$I(t) = i\Omega + \int_0^t ds\, \psi(s) \tag{3.2a}$$

and

$$\psi(t) = \langle g(t) \mid g^+(0) \rangle\, C^{-1}(t) \tag{3.2b}$$

The random force (RF)

$$g(t) = i[L - iI(t)]\, A(t) \tag{3.3}$$

may be written explicitly as

$$g(t) = [\exp(it\hat{Q}L\hat{Q})][1 - \hat{Q}d(t)]^{-1}\,\hat{Q}\dot{A}(0) \tag{3.4}$$

and

$$d(t) = 1 - \exp(-itL)\exp(it\hat{Q}L\hat{Q})$$

so that the explicit expressions of this TLP equation are much more difficult than the corresponding CP ones, which is probably why these equations have found only few applications so far.

The RF $g(t)$ obeys the orthogonality condition

$$\langle g(t) \mid A^{+}(0)\rangle = 0, \qquad \forall t \geqslant 0 \tag{3.5}$$

which again is a nonstationary property in the sense explained with regard to Eq. (2.8c). Further properties of $g(t)$ may be shown to be

$$\langle g(t_1) \mid g(t_2)^{+}\rangle = \gamma(t_1, t_2) \tag{3.6a}$$

where

$$\gamma(t_1, t_2) \to \gamma_{as}(t_1 - t_2), \qquad t_1, t_2 \gg t_c \tag{3.6b}$$

provided an autonomous macrodynamics exists, i.e., provided the limes in Eq. (1.7)

$$\lim_{t \to +\infty} I(t) = I \tag{3.7}$$

exists and is approached rapidly for $t \gg t_c$.

Thus, the RF $g(t)$ is seen to behave in a nonstationary way at short times, which is connected with the initial slip of pertinent correlation functions and hence with the memory of the initial preparation of the system.

Because of Eq. (1.7), the existence of an autonomous macrodynamics enters the GLE (3.1) explicitly. However, the theory is more complicated now than the corresponding CP approach given in Section 2. This is reflected, for instance, by the fluctuation-dissipation theorem (3.2b), which gets even much more complicated for the corresponding nonlinear theory. These difficulties can be traced back to the orthogonality requirement, which we will drop in the following.

4. THE NEW TIME-LOCAL GENERALIZED LANGEVIN EQUATION

The GLEs (2.2) and (3.1) introduced so far obviously not only single out a particular instant of time, but also possess some properties of nonstationarity connected with the existence of lifetime effects in the system. As discussed in more detail in I, these lifetime effects lead in the systematic parts of LEs to the convolution in Eq. (2.2) and to the time dependence of $I(t)$ in Eq. (3.1). In the random force terms they lead to the finite correlation time t_c of f or g, causing the noise to be colored in all realistic systems.[4] This is obvious physically, since the relaxation originates from microscopic events, binary collisions, e.g., which usually have a finite lifetime.

4.1. Formulation of the Langevin Equation

In the TLP approach presented in I it was found natural to split both the correlation matrix $C(t)$ and the expectation values $a(t)$ into a macroscopic and a microscopic part [cf. Eq. (1.3)] and correspondingly

$$a(t) = a^{(+)}(t) + a^{(m)}(t), \qquad t > 0 \tag{4.1a}$$

where $a^{(+)}$ denotes the macroscopic or irreversible branch of the time evolution of $a(t)$, whereas $a^{(m)}(t)$ is of purely microscopic origin. We note Eq. (1.4) (and hence $a^{(m)}(t) = 0$, $t \gg t_c$) and the fact that $C^{(+)}$ and $a^{(+)}$ obey the autonomous macrodynamics [cf. Eq. (1.6)]

$$\dot{a}^{(+)}(t) = -Ia^{(+)}(t), \qquad \dot{C}^{(+)}(t) = -IC^{(+)}(t) \tag{4.1b}$$

for all times t.

The idea of projection does not fit this picture very explicitly, as discussed extensively in I. This is also obvious from the GLEs (2.2) and (3.1), the systematic (or friction) parts of which are, so to say, not completely decoupled from the microscopic world, since they describe the time evolution of $a(t)$ and not of $a^{(+)}(t)$.

Let us therefore introduce a new kind of GLE by simply making the ansatz

$$\dot{A}(t) = -IA(t) + Z(t), \qquad -\infty < t < \infty \tag{4.2}$$

where

$$Z(t) = i(L - iI)\, A(t) \tag{4.3}$$

where I was introduced in Eqs. (1.7) and (3.7).

Equation (4.2) is a trivial reformulation of Eq. (2.1) and in fact is valid for any matrix I. However, if we want to consider Eq. (4.2) as a GLE, a

[4] Notable exceptions are classical hard-sphere systems.

necessary condition is that $Z(t)$ may be taken for a random force (RF), i.e., that for

$$K(t) = \langle Z(t+s) \mid Z(s)^+ \rangle, \forall s \tag{4.4}$$

we have

$$K(t) = 0, \qquad |t| \gg t_c \tag{4.5}$$

This condition fixes I in an unambiguous way to be just the kernel of the autonomous macrodynamics occurring in Eq. (4.1b). In fact, using Eqs. (1.3) and (1.4) together with (A.1a), one readily verifies that (4.5) is valid if and only if I corresponds to Eq. (4.1b). We note again that the ultimate condition for Eq. (4.5) to be valid consists in the assumption (see Section 1) that an autonomous macrodynamics exists, which is tantamount to the proposition that A represents a complete set or that Eq. (2.6) in the CP approach and hence Eq. (1.4) are fulfilled.

In view of Eqs. (2.6) and (4.5), Z behaves as "randomly" as f, so that at this level there is no qualitative difference between the two forces. However, for the behavior of the correlation matrix Q of the RF Z with A we find

$$Q(t) = \langle Z(t+s) \mid A(s)^+ \rangle, \qquad \forall s \tag{4.6a}$$

which is stationary, in contrast to Eqs. (2.8c) and (3.6a). From Appendix B we obtain

$$Q(t) = (\partial_t + I)\, C^{(m)}(t), \qquad t > 0 \tag{4.6b}$$

and hence

$$Q(t) = 0, \qquad t \gg t_c > 0 \tag{4.6c}$$

so that obviously *Z is correlated over a microscopic period of time with the observables A*. Consequently, we find, upon averaging our GLE (4.2) with some nonequilibrium ensemble $\rho(t_0)$, that the contribution of the RF does not vanish identically as is the case with GLEs introduced earlier. Instead, we find, in using, for example $\rho(t_0)$ of Eq. (2.3) of I and linearization from Eq. (4.2),

$$\dot{a}(t) = -Ia(t) + Q(t - t_0)\, a(t_0) \tag{4.7a}$$

so that we also have

$$Q(t) = [I - I(t)]\, C(t) = (\partial_t + I)\, C(t) \tag{4.8}$$

the interrelation of Q with the "memory" kernel (2.5) having been given earlier [cf. Eq. (3.13) of Ref. 5]. Note that corresponding to Eq. (4.7a) we also find

$$\dot{C}(t) = -IC(t) + Q(t) \tag{4.7b}$$

so that both the initial slip and the memory of the initial preparation are traced in the present theory to the colored noise character of the RF via the nonorthogonality of A with Z.

4.2. Proporties of the Random Force

Before discussing this point further, we will give some additional properties of the RF Z. For this purpose we introduce a kind of interaction representation of the GLE (4.2), i.e., we introduce

$$A_w(t) = e^{It} A(t), \qquad Z_w(t) = e^{It} Z(t) \tag{4.9}$$

so that

$$\dot{A}_w(t) = Z_w(t), \qquad -\infty < t < \infty \tag{4.10}$$

which means that there is no friction term. This is no surprise, since the interaction representation may be understood as looking at the dynamics from a moving reference frame chosen such that the macrodynamics is just compensated and $A_w(t)$ is driven by fluctuations only.

In terms of Z_w we obtain (see Appendix B) for Γ, where

$$\Gamma = \Delta - F, \qquad \Delta = C^{(+)}(0) \tag{4.11}$$

the expression

$$\begin{aligned} \Gamma &= \int_0^\infty dt \, \langle Z_w(t) \mid A^+ \rangle, \qquad A = A(0) \\ &= \int_0^\infty dt \, e^{It} Q(t) \end{aligned} \tag{4.12}$$

As shown in I, we have

$$\Gamma = O(\xi), \qquad \xi = t_c / t_R \tag{4.13}$$

so that Γ vanishes for $\xi \to 0$, i.e., for a complete separation of microscopic (t_c) and macroscopic (t_R) time scales.

For the correlation matrix of Z_w with $\dot{A}$ we find from Eqs. (4.6)

$$R(t) = \langle Z(t+s) \mid \dot{A}(s)^{+} \rangle = -\dot{Q}(t) \tag{4.14a}$$

$$R_w(t) = \langle Z_w(t+s) \mid \dot{A}(s)^{+} \rangle = -\partial_t e^{It} \dot{C}(t) \tag{4.14b}$$

and from Eq. (B.5)

$$\mathscr{L}^{ir} = \mathscr{L} - i\Omega F = \int_0^{\infty} dt\, R_w(t) \tag{4.15}$$

where Ω was introduced in Eq. (2.4) and $\mathscr{L}$ in Eq. (1.13).

Moreover, from Eq. (4.14a), we obtain immediately, using $F = C(0)$ and $i\dot{C}(0) = \Omega F$, that

$$I^{ir} := I - i\Omega = \int_0^{\infty} dt\, R(t)\, F^{-1} \tag{4.16}$$

so that the irreversible part of the transport kernel and the matrix of generalized Onsager coefficients $\mathscr{L}$ are related to the correlation functions of Z and Z_w, respectively, with $\dot{A}$. In Brownian motion proper $\dot{A}$ is proportional to the total force acting on the particle. Hence, we may interpret $R(t)$ as describing the correlations between the friction and the total force, and Eqs. (4.15) and (4.16) may be viewed as establishing the relation between I^{ir}, $\mathscr{L}^{ir}$, and these correlations.

One also may derive a *fluctuation-dissipation* theorem (see Appendix C), i.e. (note $F = F^{+}$)

$$\begin{aligned} IF + FI^{+} = \int_{-\infty}^{0} dt\, [&\langle Z_w(t) \mid Z_w(0)^{+} \rangle \\ &+ \langle Z_w(0) \mid Z_w(t)^{+} \rangle] \end{aligned} \tag{4.17}$$

which may be viewed as relating the correlation matrix of the RF to the macroscopic decay properties of the system. A related expression is given in Eq. (7.3).

Finally, we note that our RF obeys the Heisenberg equation

$$\dot{Z}(t) = iLZ(t) \tag{4.18}$$

just as $A(t)$ itself does [cf. Eq. (2.1)], where the difference between A and Z consists in the initial condition

$$Z(t_1) = i(L - iI)\, A(t_1) = \dot{A}(t_1) + IA(t_1) \tag{4.19}$$

which may be viewed as considering $\dot{A}$ or the corresponding current with the macroscopic or systematic part $-IA$ subtracted away. The RF of the LE (2.2) obeys in turn [Eq. (2.7)]

$$\dot{f}(t) = i\hat{Q}L\hat{Q}f(t) \tag{4.20}$$

with initial condition

$$f(0) = i(L+\Omega)\, A(0) \tag{4.21}$$

so that $f(t)$ is governed by the projected dynamics given by $\hat{Q}L\hat{Q}$. This is usually considered as the reason for the decay property (2.6) of the correlation matrix of the RF (2.5). However, as seen from Eq. (4.5), the projection is not necessary to produce the ensuing randomness. Instead of projection, one may just as well use the subtraction procedure (4.19), which obviously is conserved under Eq. (4.18), so that in this way the systematic part is removed from Z at all times.

A different way of demonstrating the randomness of $Z(t)$ is found by noting that $Z(t) = (\partial_t + I)\, A(t)$ and that

$$(\partial_t + I)\, e^{-It} B = 0$$

where B is an arbitrary matrix. Consequently, the application of the operator $(\partial_t + I)$ to $A(t)$ destroys any contribution in $A(t)$ which obeys the autonomous macrodynamics. Thus, $Z(t)$ contains only the fluctuations around the latter which are of purely microscopic nature.

4.3. A Simple Example

Let us consider a simple example for the purpose of illustrating some of the special features of our GLE (4.2). For demonstrating the relationship of our GLE to Eq. (2.2), we do not start out from a specific microscopic model and calculate $Q(t)$ and $K(t)$ directly. Instead, we assume Eq. (2.2) is already known, $M(t)$ [cf. Eq. (2.5)] being given by the simple exponential ansatz

$$M(t) = be^{-ct} \tag{4.22}$$

where the case of a single observable is considered, so that $M(t)$ is a scalar function.

Using the results of I, Appendix A, we can easily obtain the correlation function $C(t)$ corresponding to Eq. (4.22) as

$$C(t) = \frac{1}{\lambda - I}\,(\lambda e^{-I|t|} - Ie^{-\lambda|t|})\, F = e^{-I|t|}\, \Delta - e^{-\lambda|t|}\, \Gamma \tag{4.23}$$

where $\Delta = [\lambda/(\lambda - I)]\, F$, $\Gamma = [I/(\lambda - I)]\, F$, and obviously $C(0) = F$ and $\dot{C}(0) = 0$, as it must be. The I and λ are connected with the parameters of $M(t)$ via $(x := 4b/c^2)$

$$\begin{aligned} I &= \frac{c}{2}\,[1-(1-x)^{1/2}] = \frac{b}{c}\,(1+x+\cdots) \\ \lambda &= \frac{c}{2}\,[1+(1-x)^{1/2}] = \frac{1}{c}\,(1-x-\cdots) \end{aligned} \tag{4.24}$$

since $I = iz_m$ and $\lambda = iz_m$. Note that $x = \xi + O(\xi)$.

For the given case of a single variable, we obtain from Eqs. (4.4) and (4.6a),

$$K(t) = -\ddot{C}(t) + I^2 C(t) = (I^2 - \partial_t^2)\, C(t) \tag{4.25}$$

so that, by means of Eq. (4.23),

$$K(t) = I(I+\lambda)\, e^{-\lambda|t|} F \tag{4.26}$$

Analogously, we find

$$\begin{aligned} Q(t) &= Ie^{-\lambda t}F, && t \geqslant 0, \\ Q(t) &= \frac{I}{\lambda - I}\,[2\lambda e^{It} - (\lambda + I)\, e^{\lambda t}]\, F, && t < 0 \end{aligned} \tag{4.27}$$

The behavior of these quantities is given in Fig. 1 for two different values of $\xi = t_c/t_R$. It is interesting to note that $K(t) = K(-t)$, whereas $Q(t)$ and $R(t)$ show a strong asymmetry with respect to time inversion. This is particularly marked in the limit $\xi \to 0$ (white noise), which we introduce for later use. For this purpose, we assume there is a slowness parameter q in the theory (see Section 4.3 of I), so that we may write $I = q^2 \bar{I}$. We introduce a scaled time $t^* = q^2 \bar{I} t$ and obtain for $q \to 0$, writing $x(t) = x[t^*]$,

$$K[t^*] = 2q^4 \bar{I}^2\, \delta(t^*)\, F \tag{4.28a}$$

as expected in that limit. Moreover,

$$Q[t^*] = 2q^2 \bar{I} \theta(-t^*)[\exp(-|t^*|)]\, F \tag{4.28b}$$

so that $Q[t^*] = 0$ for $t^* > 0$, and

$$R[t^*] = 2q^4 \bar{I}^2 [\delta(t^*) - \theta(-t^*) \exp(-|t^*|)]\, F \tag{4.28c}$$

Thus, we find that for $t^* > 0, Z$ is not correlated with A, whereas we observe strong correlations for $t^* < 0$; see Section 8 for a detailed discussion of this point.

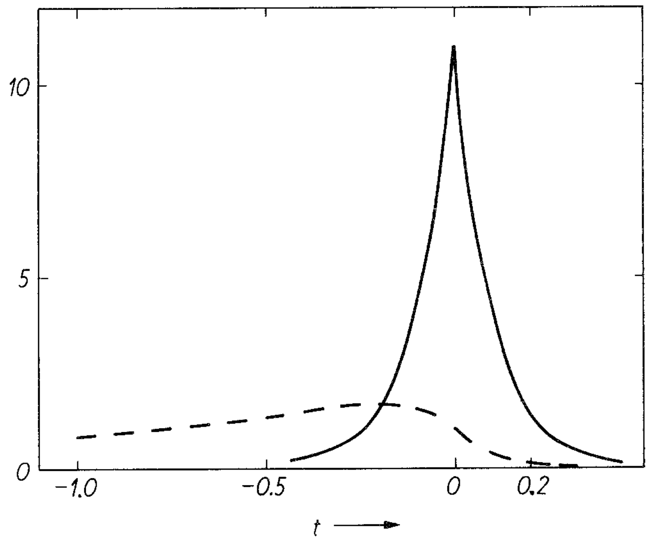

Fig. 1. The correlation functions (—) $K(t)$ (- -) and $Q(t) = Q^{(+)}(t)$ over time t for the model considered in Section 4.3. We put $F = 1$ and choose the parameters as $I = 1$ and $\lambda = 10$, so that lifetime effects are of the order of 10%.

5. EXPLICIT EXPRESSIONS FOR RANDOM FORCE CORRELATION MATRICES

The RF Z is qualified by its correlation matrices K, Q, and R as introduced in Section 4. If Z is of non-Gaussian nature, one also needs higher order correlators. The microscopic expressions given in Eqs. (4.4), (4.6), and (4.14) are still implicit, since they contain I, which usually is not known. Explicit expressions can be obtained in a straightforward manner by using the method of time-scale expansions (TSE) as introduced in Section 6 of I. For doing so we note that K and Q obey the decay properties (4.5) and (4.6c). Thus, we may find a time t^0 that is still of order t_c but is sufficiently large that K and Q are negligible for $t > t^0$.

Consequently, we need to consider times $t < t^0$ only. For simplicity, we study the case of A even only, so that Ω [cf. Eq. (2.4)] is equal to zero. We introduce [cf. Eq. (6.3) of I]

$$q(t) = \int_0^t dt'\, (t - t')\, \ddot{C}(t') \tag{5.1}$$

so that

$$C(t) = F + q(t) \tag{5.2}$$

Moreover, we observe

$$\dot{q}(t) = \int_0^t dt'\, \ddot{C}(t') = \dot{C}(t) \tag{5.3}$$

and write Eqs. (6.6) and (6.7) of I as

$$I = -\dot{q}(t^0) + \dot{q}(t^0)\, q(t^0) \tag{5.4}$$

keeping lifetime effects in leading order only, as will be done throughout the following.

Now, rewriting Eq. (4.7b) as

$$Q(t) = (\partial_t + I)\, C(t) \tag{5.5}$$

we find from introducing Eqs. (5.2) and (5.4) into (5.5) and after some manipulations

$$Q(t) = \langle Z(t) | A^+ \rangle = Q_0 + Q_1 + \cdots \tag{5.6a}$$

where

$$\begin{aligned} Q_0(t) &= -\int_t^{t^0} dt'\, \ddot{C}(t') \\ Q_1(t) &= -\int_0^{t^0} dt_1 \int_t^{t^0} dt_2 \int_0^{t_2} dt_3\, \ddot{C}(t_1)\, F^{-1} \ddot{C}(t_3) \end{aligned} \tag{5.6b}$$

which are the first members of the time scale expansion of $Q(t)$. By a similar argument we find

$$K(t) = K_0 + K_1 + \cdots \tag{5.7a}$$

where

$$\begin{aligned} K_0(t) &= -\ddot{C}(t) \\ K_1(t) &= \left[\int_t^{t^0} dt_1 \int_0^{t^0} dt_2 + \int_0^{t^0} dt_1 \int_0^{t} dt_2 \right] \ddot{C}(t_1)\, \ddot{C}(t_2)\, F^{-1} \end{aligned} \tag{5.7b}$$

and we note that in the case of a single observable

$$K_1(t) = \int_0^{t^0} dt_1 \int_0^{t^0} dt_2\, \ddot{C}(t_1)\, \ddot{C}(t_2)\, F^{-1} \tag{5.7c}$$

If the time scales given by t_c and t_R are sufficiently well separated so that lifetime effects are not too large, Eqs. (5.6) and (5.7) give a complete description of the behavior of $Q(t)$ and $K(t)$ for $0<t<t^0$. In practical applications, in many cases (i.e., if lifetime effects are negligible) it will even be sufficient to keep Q_0 and K_0 only, so that a very simple estimate of the behavior of the RF is obtained. The above procedure can also be carried over to the calculation of higher order correlators of the RF.

6. SPECTRAL DENSITY OF FLUCTUATIONS AND GENERALIZED LANGEVIN EQUATION

Additional physical insight into the GLE proposed in Eq. (4.2) can be obtained by studying the Fourier transform $\hat{C}(\omega)$ of the correlation matrix $C(t)$, where Fourier transforms are identified with a caret in the following, i.e.,

$$\hat{x}(\omega)=\int_{-\infty}^{\infty} dt\, e^{i\omega t}x(t) \tag{6.1}$$

for any function $x(t)$. The $\hat{C}(\omega)$ may be identified with the spectral density of the fluctuations of $A(t)$. It is directly accessible experimentally in many cases, e.g., by light or neutron scattering experiments. As is well known, $\hat{C}$ is connected with the Fourier–Laplace transform $\tilde{\tilde{C}}$,

$$\tilde{\tilde{C}}=\langle A|\frac{i}{z-L}|A^+\rangle \tag{6.2}$$

introduced earlier [cf. Eq. (3.2) of I by

$$\begin{aligned}\hat{C}(\omega)&=\tilde{\tilde{C}}(\omega+i\varepsilon)-\tilde{\tilde{C}}(\omega-i\varepsilon)\\&=2\pi\,\langle A|\,\delta(\omega-L)\,|A^+\rangle\end{aligned} \tag{6.3}$$

We note that $\hat{C}(\omega)$ is a Hermitian matrix,

$$\hat{C}(\omega)=[\hat{C}(\omega)]^+ \tag{6.4}$$

for real values of ω, due to Eq. (A.1). $\hat{C}$ is related to the Fourier transform $\hat{\chi}''$ of the response matrix or the matrix of response functions χ'' by the expression

$$\omega\hat{C}(\omega)=(2/\beta)\,\hat{\chi}''(\omega) \tag{6.5}$$

where

$$\chi''_{i,j}(t)=\tfrac{1}{2}\langle\{A_i(t),A_j\}\rangle_{\text{eq}} \tag{6.6a}$$

and

$$\chi''_{i,j}(t)=(1/2\hbar)\langle[A_i(t),A_j]\rangle_{\text{eq}} \tag{6.6b}$$

where $\{\cdots\}$ and $[\cdots]$ denote the Poisson bracket and the commutator in the classical and quantum cases, respectively.

The spectral density $\hat{C}$ and hence $\hat{\chi}''$ can be related to the Fourier transform of $M(t)$ [cf. Eq. (2.2)] by the dispersion relation, which in the particularly simple case of a single observable ($n=1$) reads [cf. Eq. (5.22) of Ref. 6]

$$\frac{1}{\beta\omega}\hat{\chi}''(\omega)=F\hat{M}(\omega)$$
$$\times\left\{\left[\omega-P\int\frac{d\omega'}{2\pi}\frac{\hat{M}(\omega')}{\omega-\omega'}\right]^2+\left[\frac{\hat{M}(\omega)}{2}\right]^2\right\}^{-1} \tag{6.7}$$

where P denotes the principal value. This expression corresponds to a modified Lorentzian line shape, to which it reduces for vanishing lifetime effects. It will be seen below that the ansatz (4.2) leads to expressions for the line shape that more explicitly show the deviations, due to lifetime effects, from the Lorentzian even in the case of several observables. These expressions also differ largely from those obtained[7-9] in the time-local approach based on Eq. (3.1).

6.1. Spectral Density, Line Shape, and Random Force

Using Eq. (4.4), we may relate the correlation matrix of the random force (RF) to the spectral density,

$$\hat{K}(\omega)=(I-i\omega)\,\hat{C}(\omega)(I^{+}+i\omega) \tag{6.8a}$$

or for the case of a single observable ($n=1$)

$$\hat{K}(\omega)=(I^2+\omega^2)\,\hat{C}(\omega) \tag{6.8b}$$

Introducing

$$\hat{\mathscr{G}}(\omega)=(I-i\omega)^{-1} \tag{6.9}$$

we write correspondingly

$$\hat{C}=\hat{\mathscr{G}}\hat{K}\hat{\mathscr{G}}^{+} \tag{6.10a}$$

and for $n=1$

$$\hat{C}(\omega)=\hat{K}(\omega)/(\omega^2+I^2) \tag{6.10b}$$

In the same way we obtain expressions for the correlation matrices Q and R introduced in Eqs. (4.6a) and (4.14a), i.e.,

$$\hat{Q}(\omega) = \hat{K}(\omega)\,\hat{\mathscr{G}}(\omega)^+ = \hat{K}(\omega)\frac{1}{I^+ + i\omega} \tag{6.11a}$$

and

$$\hat{R}(\omega) = i\omega\hat{Q}(\omega) = \hat{K}(\omega) - \hat{K}(\omega)\,I^+\hat{\mathscr{G}}(\omega)^+ \tag{6.11b}$$

As is obvious from Eq. (6.10b), which is to be contrasted with Eq. (6.7), our approach based on the GLE (4.2) leads to rather explicit expressions in which the modification of the Lorentzian line shape due to lifetime effects is directly related to the properties of the RF contained in $\hat{K}(\omega)$. To discuss this point further, we taylor expand $\hat{K}$,

$$\hat{K}(\omega) = K_0 + \omega K_1 + \cdots \tag{6.12}$$

and note that $K_n = O(\xi^n)$, $n \geqslant 1$, as a consequence of the decay property (4.5). Thus, (6.12) corresponds to a time scale expansion of $\hat{K}$ that converges for all ω, where

$$|\omega| < \omega_0, \qquad \omega_0 = O(1/t_c) \tag{6.13}$$

Moreover, we find

$$K_l = 0, \qquad l = 1, 3,... \tag{6.14}$$

if $C(t) = C(-t)$, as is the case for A even [cf. Eq. (A.2b)].

It is interesting to consider the above expressions for small and large ω. In the latter case, i.e., for $\omega \gg t_R^{-1}$, we obtain from Eqs. (6.8a) and (6.5)

$$\hat{K}(\omega) = \omega^2\hat{C}(\omega) = \frac{2\omega}{\beta}\,\chi''(\omega), \qquad |\omega| \gg t_R^{-1} \tag{6.15a}$$

so that

$$\|\hat{K}(\omega)\| \sim |\omega|^{-\infty}, \qquad \omega \to \infty \tag{6.15b}$$

if the potential is such that all sum rule expressions do exist. Accordingly, we also obtain [cf. Eq. (6.11a)]

$$\hat{Q}(\omega) = -i\omega\hat{C}(\omega) = -(2i/\beta)\,\chi''(\omega), \qquad |\omega| \gg t_R^{-1} \tag{6.16a}$$

and

$$\|\hat{Q}(\omega)\| \sim |\omega|^{-\infty}, \qquad |\omega| \to \infty \tag{6.16b}$$

Thus, the high-frequency properties of $\hat{K}$ and $\hat{Q}$ are directly related to the properties of the spectral density or the response matrix. For low frequencies, i.e., $|\omega| \ll t_c^{-1}$, we find by introducing (6.12) into Eq. (6.10)

$$\hat{C}(\omega) = \hat{\mathscr{G}}(\omega)(K_0 + \omega K_1 + \cdots)\,\hat{\mathscr{G}}(\omega)^+ \tag{6.17a}$$

where explicit expressions for K_1 are given in Appendix D. For not too large ξ and ω, i.e., neglecting terms of order $(\omega t_c)^2$ or ξ^2 and higher, we obtain the following approximate relations:

$$\begin{aligned}\hat{C}(\omega) &= \hat{\mathscr{G}}(\omega)[(I - i\omega)\,\Delta^+ + \text{h.c.}]\,\hat{\mathscr{G}}(\omega)^+ \\ &= (I - i\omega)^{-1}\,\Delta + \text{h.c.}\end{aligned} \tag{6.17b}$$

Using Eq. (A.10), we obtain for the case of even $A(E=1)$ from Eq. (6.17b)

$$\hat{C}(\omega) = \hat{\mathscr{G}}(\omega)[\mathscr{L} + \mathscr{L}^+]\,\hat{\mathscr{G}}(\omega)^+ \tag{6.18}$$

where $\mathscr{L}$ was introduced in Eq. (1.13).

For $n = 1$ we obtain in particular

$$\hat{C}(\omega) = 2\mathscr{L}/(\Gamma^2 + \omega^2) \tag{6.19}$$

Equations (6.17)–(6.19) are low-frequency expressions of $\hat{C}$ which are correct if terms of order $(\omega t_c)^2$ and ξ^2 are negligible. Thus, they include lifetime effects in lowest order.

Low-frequency expressions for the spectral density have long been known. However, our expressions, say, Eq. (6.19), differ from the conventional ones [e.g., Eq. (121, 15) of Ref. 10] by the inclusion of lifetime effects into our theory. It is interesting to note that the regions of validity of the high- and low-frequency expressions given above overlap if ξ is not too large. In fact, Eq. (6.15a) is valid for $|\omega| \gg 1/t_R$, whereas Eqs. (6.17)–(6.19) are valid for $|\omega| \ll t_c^{-1}$. Thus, we find that at low frequencies, i.e., in the regions of the peaks at least, $\hat{C}$ is given by a (multivariate) Lorentzian, whereas the high-frequency (wing) region is given by the spectral density of the random force. For the case of even A, this is correct with lifetime effects of leading order included. In the even–odd case, we find that the Lorentzian already is biased by lifetime effects in lowest order, since $\Delta - \Delta^+ = O(\xi)$ in Eq. (6.17b),

6.2. Laurent Expansion of Spectral Density

In I the Laurent series expansion of the Fourier–Laplace transform $\tilde{C}(z)$ was shown to reveal interesting relations between macro and microphysics. A corresponding expansion also can be found for $\hat{C}(\omega)$. For this purpose we split $C(t)$ as

$$C(t) = C^{(M)}(t) + C^{(m)}(t), \qquad -\infty < t < \infty \tag{6.20}$$

where

$$C^{(M)}(t) = \theta(\pm t)[\exp(-\Gamma^{(\pm)}t)]\,\Delta^{(\pm)} \tag{6.21}$$

according to Eq. (2.23) of I, with θ denoting the Heaviside step function, and we note that $\Gamma^{(+)}$ and $\Gamma^{(-)}$ are of different sign [cf. Eq. (A.9a)], so that $C^{(M)}(t)$ is decaying for $t \to \pm\infty$.

From (6.21) we obtain, using Eqs. (A.7), (A.9), and (A.10),

$$\hat{C}^{(M)}(\omega) = \hat{\mathscr{G}}(\omega)(\Gamma - i\omega)\,\Delta^{+}\hat{\mathscr{G}}(\omega)^{+} + \text{h.c.} \tag{6.22a}$$

and for even A

$$\hat{C}^{(\mathrm{M})}(\omega) = \hat{\mathscr{G}}(\omega)[\mathscr{L} + \mathscr{L}^{+}]\,\hat{\mathscr{G}}(\omega)^{+} \tag{6.22b}$$

so that $\hat{C}^{(\mathrm{M})}$ is seen to agree with the low-frequency expression (6.17b), which is valid if ω and ξ are not too large.

The analytical continuation of $\hat{C}^{(\mathrm{M})}$ is readily obtained by simply considering Eq. (6.22a) for arbitrary complex values $\omega = \omega' + i\omega''$. Then, $\hat{\mathscr{G}}(\omega)$ ($\hat{\mathscr{G}}^{+}$) is found to have just n poles in the upper (lower) half-plane situated a distance of the order of t_R^{-1} away from the real axis. Consequently, for sufficiently large ω, i.e., $|\omega| > \bar{\omega}$, $\bar{\omega} = O(1/t_R)$, we find the Laurent expansion of $\hat{C}^{(\mathrm{M})}$,

$$\hat{C}^{(\mathrm{M})} = \sum_{k=-1}^{-\infty} \alpha_k \omega^{+k} \tag{6.23}$$

where the matrix coefficients α_k are found most easily by introducing the Laurent expansion of $\mathscr{G}$,

$$\hat{\mathscr{G}}(\omega) = \frac{i}{\omega} \sum_{n=0}^{\infty} \left(\frac{\Gamma}{i\omega}\right)^{n} \mathbf{1} \tag{6.24}$$

where $\mathbf{1}$ denotes the unit matrix, into Eq. (6.22), so that, for example,

$$\begin{aligned} \alpha_{-1} &= i(\Delta^{(+)} - \Delta^{(+)+}) = i(\Delta^{(+)} - \Delta^{(-)}) \\ \alpha_{-2} &= \mathscr{L} + \mathscr{L}^{+} = \mathscr{L}^{(+)} + \mathscr{L}^{(-)} \\ &\vdots \end{aligned} \tag{6.25a}$$

Moreover, one easily concludes that if $C(t)=C(-t)$, it follows that $\alpha_{-k}=0$ for $k=1, 3,\ldots$. Thus, for the case of all A even, we find

$$\alpha_{-1}=0 \tag{6.25b}$$

On the other hand, due to its rapid decay for $|t| \gg t_c$, we may Taylor expand $\hat{C}^{(\mathrm{m})}(\omega)$ as

$$\hat{C}^{(\mathrm{m})}(\omega)=\sum_{k=0}^{\infty} \alpha_k \omega^k \tag{6.26}$$

which is a time scale expansion valid for $|\omega| < \bar{\bar{\omega}}$, where $\bar{\bar{\omega}}=O(1/t_c)$. Now, inside its circle of convergence, Eq. (6.26) also may be used for complex values of ω, so that by adding Eqs. (6.23) and (6.26), we find

$$\hat{C}(\omega)=\sum_{k=-\infty}^{\infty} \alpha_k \omega^k \tag{6.27}$$

which is the desired Laurent expansion of $\hat{C}(\omega)$ valid in the annulus $\bar{\omega} < |\omega| < \bar{\bar{\omega}}$. We note that *the Laurent expansion provides us in a natural way the splitting of* $\hat{C}$ *into a microscopic and a macroscopic part* [cf. Eq. (6.20)], since $\hat{C}^{(\mathrm{m})}$ and $\hat{C}^{(\mathrm{M})}$ correspond just to the regular and principal parts, respectively, of the Laurent expansion of $\hat{C}$.

6.3. Complex Sum Rules

By means of

$$\alpha_k=\frac{1}{2\pi i} \oint d\omega\, \omega^{-(k+1)}\hat{C}(\omega)$$

we find from (6.27), using Eqs. (6.25) and (6.5),

$$\frac{1}{2\pi} \oint d\omega\, \hat{C}(\omega)=\frac{1}{\beta\pi} \oint d\omega \frac{\hat{\chi}''(\omega)}{\omega}=\Delta^{(+)}-\Delta^{(-)} \tag{6.28}$$

and

$$-\frac{i}{\pi\beta} \oint d\omega\, \hat{\chi}''(\omega)=\mathscr{L}+\mathscr{L}^{+} \tag{6.29}$$

We note that the integration is to be carried out along a closed contour encircling the macroscopic but not the microscopic singularities of $\hat{C}(\omega)$, i.e., the contour must lie inside the annulus of convergence of the Laurent expansion.

The above expressions may be considered as a kind of sum rule in the complex frequency domain relating dissipative quantities to $\hat{C}$ or $\hat{\chi}''$, whereas the usual sum rules yield static or nondissipative quantities, such as

$$\frac{1}{\pi\beta}\int_{-\infty}^{\infty} d\omega\, \hat{\chi}''(\omega) = \Omega F \tag{6.30}$$

where $i\Omega F$ is just the nondissipative part of the matrix of Onsager coefficients $\mathscr{L}$, whereas the quantity $\mathscr{L} + \mathscr{L}^+$ will be seen in a subsequent paper to represent just the entropy production.

7. GENERALIZED DIFFUSION AND MODIFIED EINSTEIN RELATION

In Brownian motion theory, an important role is played by the diffusion constant (or tensor in anisotropic media) characterizing the asymptotic behavior of the mean squared displacement. In our general theory, we may introduce a corresponding quantity by considering the matrix

$$u(t) = \langle \delta x(t) | \delta x(t)^+ \rangle \tag{7.1a}$$

where

$$\delta x_k(t) = \int_0^t dt'\, A_k(t') = x_k(t) - x_k(0) \tag{7.1b}$$

so that $x_k(t)$ is a distance if $A_k(t)$ is interpreted as a velocity. Using the fact that the correlation matrix $C(t)$ is decaying for $t \to \pm\infty$, we find that for very large t

$$u(t) = t \int_{-\infty}^{\infty} C(t')\, dt' =: 2Dt \tag{7.2}$$

so that the Hermitian matrix D corresponds formally to the diffusion tensor.

Using Eqs. (4.4) and (4.3), we drive from Eq. (7.2)

$$2IDI^+ = \int_{-\infty}^{\infty} dt\, \langle Z(t) | Z^+ \rangle = K_0 \tag{7.3}$$

where K_0 was introduced in Eq. (6.12). *Equation (7.3) relates D to the correlation matrix of the* RF. To discuss this relation, we consider the case

$\xi \to 0$ (white noise limit) first. Then, K_0 agrees with the rhs of Eq. (4.17), i.e., we obtain

$$2D = I^{-1}F + \text{h.c.} \qquad \text{for} \quad \xi \to 0 \tag{7.4a}$$

which, if specified to the case of (one-dimensional) Brownian motion, is just the well-known Einstein relation

$$D = (kT/m)\, I^{-1} \tag{7.4b}$$

connecting the diffusion with the friction constant. Consequently, Eq. (7.4a) may be considered as the multivariate version of the Einstein relation of Brownian motion theory.

For the realistic (colored noise) case, one obtains instead of Eq. (7.4a) from (7.3) by means of (D.2)

$$2D = I^{-1}\Delta + \text{h.c.} + O(\xi^2) \tag{7.5}$$

which explicitly shows that the multivariate Einstein relation (7.4a) is modified if lifetime effects are not negligible, Eq. (7.5) including in first order the correction terms due to the finite duration of the binary collisions.

In the example considered in Section 4.3 we may calculate D explicitly, i.e., we obtain from Eqs. (7.3) and (4.26)

$$D = FI^{-1}(1 + I/\lambda) \tag{7.6}$$

which clearly shows the influence of the finite lifetime and obviously reduces to Eq. (7.4) for $\xi \approx I/\lambda \to 0$.

For a further discussion of the role of the general diffusion matrix D, we rewrite the low-frequency expression (6.17b) of $\hat{C}$ as

$$\hat{C}(\omega) = \frac{1}{W - i\omega\Delta^{-1}} + \text{h.c.} \tag{7.7}$$

where

$$W = \Delta^{-1}I \tag{7.8}$$

Let us consider the case of even A in the following. Then, we find from Eqs. (7.5), (7.7), and (7.8) and $W = W^{+}$ [prove by means of Eqs. (A.7), (A.9), and (A.10) for $E = 1$]

$$\hat{C}(\omega) = \frac{1}{D^{-1} - i\omega\Delta^{-1}}\,(2D^{-1})\,\frac{1}{D^{-1} + i\omega\Delta^{-1}} \tag{7.9a}$$

which for the case of a single observable reduces to

$$\hat{C}(\omega)=\frac{2D^{-1}}{D^{-2}+\omega^2 \Delta^{-2}} \tag{7.9b}$$

Again, these are correct low-frequency expressions if lifetime effects are not too large. Equation (7.9) expresses the Lorentzian directly in terms of the generalized diffusion matrix and the modified variance matrix of the fluctuations [cf. Eqs. (1.10)–(1.12)]. For vanishing lifetime effects, Eq. (7.9) reduces to well-known expressions [cf. Eq. (124, 6) of Ref. 10].

8. HEISENBERG DYNAMICS AS A GENERALIZED ORNSTEIN–UHLENBECK PROCESS

There are essentially two different points of view in looking at a GLE. On the one hand, by virtue of the deterministic equations of motion (4.20) or (4.18) for the random force (RF), there is a one-to-one correspondence between the solutions of the Heisenberg equation (2.1) and the corresponding GLE such as (2.2) or (4.2). On the other hand, due to the chaotic behavior of the RF, it is reasonable to reinterpret the latter as a stochastic process. Then, the GLEs become stochastic differential or integrodifferential equations which define the stochastic process $A(t)$. The properties of the RF and hence of $A(t)$ are now fully determined by specifying the RF correlation matrices such as Eqs. (2.5) and (4.4) together with all of the higher order correlators (if the process is non-Gaussian). If these correlators are calculated from the microscopic expressions for the RF, one finds that all correlators $C(t)$, e.g., of the stochastic process $A(t)$, agree with those obtained from the Heisenberg equations (2.1). In this sense one may say that Heisenberg dynamics and the stochastic process are completely equivalent to each other.

Now, the GLE (4.2) shows that $A(t)$ may be represented by a generalized Ornstein–Uhlenbeck (OU) process. In fact, let us consider Eq. (4.2) as a set of stochastic differential equations and consider the correlation matrix $C(\tau)$ in the stationary state, i.e.,

$$\lim_{t\to\infty} \langle\langle A(t+\tau)\, A^+(t)\rangle\rangle =: C(\tau) \tag{8.1}$$

where $\langle\langle\cdots\rangle\rangle$ denotes the stochastic average. Then, as is well known,[11] the Fourier transform of $C(\tau)$ is given just by Eq. (6.10a) in accordance with our proposition that $A(t)$ is repreented by a generalized OU process. By means of the Wiener–Khinchine theorem this may also be stated as

$$\langle\langle A_\omega A^+_{\omega'}\rangle\rangle = \pi\delta(\omega-\omega')\,\frac{1}{I-i\omega}\,\hat{K}(\omega)\,\frac{1}{I^+ +i\omega} \tag{8.2a}$$

where the stochastic quantity A_ω is formally given as the Fourier transform of $A(t)$. In this sense, we also have

$$\pi\hat{K}(\omega)\,\delta(\omega-\omega') = \langle\langle Z_\omega Z_{\omega'}^+\rangle\rangle \tag{8.2b}$$

and hence

$$\langle\langle A_\omega A_{\omega'}^+\rangle\rangle = \frac{1}{I-i\omega}\,\langle\langle Z_\omega Z_{\omega'}^+\rangle\rangle\,\frac{1}{I^+ + i\omega'} \tag{8.2c}$$

The OU process considered is generalized since the RF Z corresponds to colored noise. The ordinary OU process corresponding to white noise is a Markovian process with finite correlation time of order t_R. Colored noise then makes $A(t)$ a non-Markovian process. As a consequence, the correlation matrix $C(t)$ displays the initial slip. The important point is that in the wings the correlation matrix $C(t)$ still obeys the autonomous macrodynamics (cf. Fig. 1 of I) which governs the systematic part of the GLE (4.2). Thus, we may say that our GLE particularly clearly displays the basic fact that the non-Markovian process $A(t)$ is fully consistent with the existence of an autonomous macrodynamics. This is contained in the GLE (2.2) in a very implicit way only.

In the framework of the (generalized) OU process it usually is felt natural to propose that the RF is not correlated with the initial value of $A(t_0)$, i.e.,

$$\langle\langle Z(t)\,A(t_0)\rangle\rangle = 0 \tag{8.3}$$

However, it must be noted that this property is not conserved during time evolution in general. Instead, though starting from Eq. (8.3), we find in the stationary state

$$\langle\langle Z_\omega A_{\omega'}^+\rangle\rangle = \langle\langle Z_\omega Z_{\omega'}^+\rangle\rangle\,\frac{1}{I^+ + i\omega'} = 2\pi\,\delta(\omega-\omega')\,\hat{Q}(\omega) \tag{8.4}$$

where

$$Q(t) = \lim_{s\to\infty}\,\langle\langle Z(t+s)\,A^+(s)\rangle\rangle \tag{8.5}$$

The qualitative properties of Q are seen best from Fig. 1 and the discussion given in Section 4.3. In the white noise limit we find that Z is still orthogonal to A for $t>0$, but not for $t<0$. For colored noise we find that Z is even for $t>0$ correlated over some time t_c with A, these correlations resulting from lifetime effects.

Hence, we may say that correlations of Z with A are a natural ingredient of the OU process described by the GLE (4.2) in the stationary state. The orthogonality requirement (8.3) is therefore seen to be rather alien to the OU process and it seems consistent to replace it with

$$\langle\langle Z(t+t_0)\, A(t_0)\rangle\rangle = Q(t), \qquad \forall t, t_0 \tag{8.6}$$

so that Eq. (4.6a) appears as a quite natural result.

The strong asymmetry of Q obviously is vital for reconciling the reversible equation (2.1) with the apparently irreversible GLE (4.2). However, it might be argued that this asymmetry has been introduced from outside. In fact, instead of the GLE (4.2), we also may consider the ansatz[5]

$$\dot{A}(t) = -I^{(-)}A(t) + Z^{(-)}(t) \tag{8.7}$$

Using Eq. (A.9a), which for the case of even A ($E = 1$) reads

$$I^{(+)} = -I^{(-)} \tag{8.8}$$

we find that the systematic part of Eq. (8.7) describes a decaying solution for $t \to -\infty$. Nevertheless, the RF correlation matrix $K^{(-)}$ obeys essentially the same properties as $K^{(+)}$ [cf. Eqs. (A.13a)–(A.13d)], so that Eq. (8.7) is as reasonable a description of $A(t)$ as Eq. (4.2) is.

The behavior of Q is crucial here. As seen from Eq. (A.14b), $Q^{(+)}$ is obtained from $Q^{(-)}$ essentially by time inversion. Hence, both in Eq. (4.2) and Eq. (8.7) the Z–A correlations are always weak (or vanishing in the white noise case) if considered in the direction of time where the frictional damping takes place [which is $t \to +\infty$ in Eq. (4.2) and $t \to -\infty$ in Eq. (8.7)], whereas we observe long-lived correlations if we take the opposite direction of time.

Moreover, Eqs. (4.2) and (8.7) are for $E = 1$ invariant against momentum inversion, or, more rigorously, against any transformation that converts iL into $-iL$. Hence, because of the iLt invariance of Eq. (2.1), we may say that the GLEs (4.2) and (8.7) no longer feel the direction of time associated with the microscopic motion. Thus, we are free to choose any one of the GLEs (4.2) and (8.7) and call forward the direction of time in which frictional damping occurs. This choice, then, is in agreement with our macroscopic experience, since it corresponds to a macroscopic description of the system in the sense of the phenomenological Langevin equation [cf. (1.1)]. In particular, under this choice, there are, apart from lifetime

[5] Note that we have used $I = I^{(+)}$, $Q = Q^{(+)}$, $Z = Z^{(+)}$, and $K = K^{(+)}$ so far for simplicity of notation.

effects, no correlations between Z and A, whereas there are strong correlations if we look backward in time, due to the asymmetry of $Q^{(\pm)}$.

Hopefully, the above discussion sufficiently clearly demonstrates our point of view that the irreversible look of the exact GLE (4.2) or (8.7) is not the result of an ad hoc introduced breaking of the time symmetry. Instead, our asymmetric formulation of the symmetric law (2.1) simply expresses the fact that the observables of our set A generally tend to relax once we observe at some time t_0 a macroscopic fluctuation of $A(t)$, and that this relaxation is observed if we look from t_0 into either the direction of $t \to +\infty$ or $t \to -\infty$.

In this formulation, the reversible nature of the underlying Heisenberg dynamics makes itself felt only in the time-asymmetric behavior of $Q^{(\pm)}(t)$, in particular, in the long-lived correlations observed when we look backward in time. In the description of any experiment, however, the GLE is used only forward in time, so that these correlations do not play any role. Hence, we may drop them. Then, our GLE describes a genuinely irreversible dynamics, which nevertheless is identical to the Heisenberg dynamics *if we use the GLE forward in time only.*

9. CONCLUDING REMARKS

The generalized Langevin equation (4.2) we have proposed in the present paper is particularly simple. Note that our GLE is actually a nonlinear equation, since the set A must also comprise all powers of some primitive variables contained in A. Although Eq. (4.2) is exact, it is identical in structure to the phenomenological GLE obtained from the ad hoc procedure of adding a noise term to an otherwise deterministic macroscopic equation. Equation (4.2) makes this procedure a rigorous one, with the result that the noise is colored and correlated with the initial value of A over some microscopic time t_c.

The latter fact is the most unusual feature of our approach. However, we hope to have clarified that this property is not as unphysical as it might seem, although it is rather unusual in the context of conventional stochastic calculus. Nonorthogonal noise has recently been discussed as a special quantum effect and necessary modifications of stochastic calculus have been given.[(12)] On the other hand, the functional-calculus approach developed recently[(13)] provides a very elegant way for dealing with the colored noise RF. A convenient modification of this approach in order to account for nonorthogonal noise would be highly desirable from the point of view of the present paper, since then the Fokker–Planck equation associated with the GLE (4.2) also could be obtained in terms of a time scale expansion (called τ-expansion in Ref. 13).

The correlations of the RF $Z(t)$ with the observables $A(t')$ at earlier times, i.e., for $t' < t$, arise from lifetime effects and vanish if the latter are negligible. The theory developed in the present paper clearly separates the autonomous macrodynamics from the lifetime effects. This is particularly clearly seen in the expressions for the spectral density, which essentially are products of multivariate Lorentzians with the spectral density $\hat{K}(\omega)$ of the RF [cf. Eqs. (6.10)], the Lorentzians corresponding to the autonomous macrodynamics, whereas the RF is represented by $\hat{K}(\omega)$, which depends on ω due to the lifetime effects.

In a subsequent paper it will be shown that this clear separation of macrophysics from microphysics obtained by our time-local formulation of the generalized Langevin equation will also lead to a convenient foundation of fluctuating irreversible thermodynamics.

APPENDIX A. SYMMETRY RELATIONS

Due to the reversibility in time of the microscopic motion, the matrix of correlation functions $C(t)$ is known to obey the relation

$$C(t) = \langle A(t) | A^+ \rangle = C^+(-t) \tag{A.1a}$$

or

$$C_{i,j}(t) = C^*_{j,i}(-t) \tag{A.1b}$$

where the asterisk denotes the complex conjugate. Moreover,

$$C_{i,j}(t) = \varepsilon_i \varepsilon_j C_{i,j}(-t) \tag{A.2a}$$

where $\varepsilon_i = +1$ or $\varepsilon_i = -1$ if the observable A_i is even or odd with respect to time reversal, respectively. Let us introduce a matrix E,

$$E_{i,j} = \varepsilon_i \, \delta_{i,j} \tag{A.3a}$$

with E obeying the properties

$$E^2 = 1, \qquad E^{-1} = E \tag{A.3b}$$

and obviously $E = 1$ if all of the A_i are even.

In terms of E, we may write Eq. (A.2a) as

$$C(t) = EC(-t) \, E \tag{A.2b}$$

and using (A.1)

$$C^+(t) = EC(t) \, E \tag{A.4}$$

For $C(0) = \langle A | A^+ \rangle =: F$ we find consequently

$$F = EFE = F^+ \tag{A.5a}$$

and

$$F_{i,j} = 0 \qquad \text{if} \quad \varepsilon_i \varepsilon_j = -1 \tag{A.5b}$$

as is well known.

Further relations are obtained by introducing the matrix $\mathring{I}(t)$, i.e., consider Eq. (2.24) of I and write

$$\partial_t C(t) = -I(t)\, C(t) = -C(t)\, \mathring{I}(t) \tag{A.6}$$

where obviously

$$\mathring{I}(t) = C^{-1}(t)\, I(t)\, C(t)$$

so that by means of Eqs. (2.33) and (2.34) of I we find

$$\mathring{I}^{(\pm)} := \lim_{t \to \pm\infty} \mathring{I}(t) = \Delta^{(\pm)-1} I^{(\pm)} \Delta^{(\pm)} \tag{A.7}$$

where

$$\Delta^{(\pm)} = \lim_{t \to \pm\infty} \exp(+I^{(\pm)} t)]\, C(t) \tag{A.8}$$

Now, using Eq. (A.2b) together with Eq. (3.7), we derive first

$$-I^{(-)} = EI^{(+)}E \tag{A.9a}$$

and by means of Eq. (A.1a)

$$\mathring{I}^{(+)} = -I^{(-)+} \tag{A.9b}$$

and correspondingly

$$\mathring{I}^{(-)} = -I^{(+)+} \tag{A.9c}$$

Using (A.8), (A.9b), and (A.9c), we also find

$$\Delta^{(+)+} = \Delta^{(-)} = E\Delta^{(+)}E \tag{A.10}$$

and consequently for $\mathscr{L}^{(\pm)} = I^{(\pm)} \Delta^{(\pm)}$

$$\mathscr{L}^{(+)} = \mathscr{L}^{(-)+} = E\mathscr{L}^{(-)}E \tag{A.11}$$

For the Fourier transforms introduced in Section 6, we obtain the following, valid for all real ω:

$$\hat{C}(-\omega) = E\hat{C}(\omega)\,E \tag{A.12}$$

corresponding to Eq. (A.2b). Using (A.12) together with Eqs. (6.8a) and (6.11a), we also derive

$$\hat{K}^{(-)}(-\omega) = E\hat{K}^{(+)}(\omega)\,E \tag{A.13a}$$

and

$$\hat{Q}^{(-)}(-\omega) = -E\hat{Q}^{(+)}(\omega)\,E \tag{A.14a}$$

which in time language read

$$K^{(-)}(-t) = EK^{(+)}(t)\,E \tag{A.13b}$$

and

$$Q^{(-)}(-t) = -EQ^{(+)}(t)\,E \tag{A.14b}$$

Moreover, we find from Eqs. (6.4) and (6.10a) that the $\hat{K}$ is Hermitian

$$\hat{K}(\omega) = [\hat{K}(\omega)]^{+} \tag{A.15a}$$

for all ω real. Consequently, we obtain

$$K(t) = [K(-t)]^{+} \tag{A.15b}$$

and using Eq. (A.13b),

$$K^{(-)}(t) = EK^{(+)}(t)^{+}\,E \tag{A.13c}$$

so that, for instance,

$$K_{1,1}^{(-)}(t) = K_{1,1}^{(+)}(t)^{*} = K_{1,1}^{(+)}(-t) \tag{A.13d}$$

APPENDIX B. DERIVATION OF EQS. (4.12) AND (4.15)

Let us consider the Fourier–Laplace transform of Eq. (4.2),

$$\tilde{\dot{A}}(z) = -I^{(+)}\tilde{A}(z) + \tilde{Z}^{(+)}(z) \tag{B.1}$$

from which we obtain

$$\tilde{A}(z) = \frac{i}{z + iI^{(+)}}\,[A + \tilde{Z}^{(+)}(z)],\ A = A(0) \tag{B.2}$$

Using Eq. (4.2) of I, we write

$$\Delta^{(+)} = \oint_{Ma} \frac{dz}{2\pi} \tilde{C}(z) = \oint_{Ma} \frac{dz}{2\pi} \langle \tilde{A}(z) | A^+ \rangle$$

$$= F + \oint_{Ma} \frac{dz}{2\pi} \frac{i}{z + i\Gamma^{(+)}} \langle \tilde{Z}^{(+)}(z) | A^+ \rangle \tag{B.3}$$

where [cf. Eq. (1.11)] $F = \langle A | A^+ \rangle$. Using $\Delta^{(+)} = F + \Gamma^{(+)}$, we obtain from Eq. (B.3)

$$\Gamma^{(+)} = \int_0^\infty dt\, [\exp(\Gamma^{(+)} t)] \langle Z^{(+)}(t) | A^+ \rangle \tag{B.4}$$

since $\langle \tilde{Z}^{(+)}(z) | A^+ \rangle = \tilde{Q}^{(+)}(z)$ has no singularities inside Ma because of Eq. (4.6c).

The corresponding equation (4.15) is obtained by starting from Eq. (4.3) of I, i.e.,

$$\mathscr{L} = -\oint_{Ma} \frac{dz}{2\pi} \tilde{\dot{C}}(z) = \oint_{Ma} \frac{dz}{2\pi} \langle \tilde{A}(z) | \dot{A}^+ \rangle$$

$$= i\Omega F + \int_0^\infty dt\, [\exp(\Gamma^{(+)} t)] \langle Z^{(+)}(t) | \dot{A}^+ \rangle \tag{B.5}$$

where $\dot{A} = \dot{A}(0)$.

APPENDIX C. DERIVATION OF THE FLUCTUATION-DISSIPATION THEOREM

We employ the common argument that

$$\langle A(t) | A(t)^+ \rangle = \langle A | A^+ \rangle = F, \qquad \forall t \tag{C.1}$$

As usual, we write the formal solution of Eq. (4.2) as

$$A(t) = U(t)\, A(0) + \int_0^t dt'\, U(t-t')\, Z^{(+)}(t') \tag{C.2}$$

where $U(t) = \exp(-\Gamma^{(+)} t)$. Introducing Eq. (C.2) into (C.1) and dropping all terms that vanish for $t \to \infty$, we find

$$F = \int_0^t dt_1 \int_0^t dt_2\, U(t-t_1)\, K(t_1 - t_2)\, U^+(t-t_2) \tag{C.3}$$

what is to be considered for large t, $t \gg t_R$, and we introduced [cf. Eq. (4.4)]

$$K(t_1 - t_2) = \langle Z^{(+)}(t_1) | Z^{(+)+}(t_2) \rangle \tag{C.4}$$

Consider $F = J_1 + J_2$, where

$$\begin{aligned} J_1 &= \int_0^t dt_1 \int_0^{t_1} dt_2\, U(t-t_1)\, K(t_1 - t_2)\, U^+(t-t_2) \\ &= \int_0^t dt_2 \int_{t_2}^t dt_1\, U(t-t_1)\, K(t_1-t_2)\, U^+(t-t_2) \\ &= \int_0^t \mathrm{ds} \int_0^s d\sigma\, U(s-\sigma)\, K(\sigma)\, U^+(\sigma) \\ &= \int_0^t ds\, [\exp(-I^{(+)}s)]\, V(s) \exp(-I^{(+)^+}s) \end{aligned} \tag{C.5}$$

and

$$V(s) = \int_0^s du\, [\exp(I^{(+)}u)]\, K(u)$$

By similar manipulations we easily find $J_2 = J_1^+$, as it must be since F is Hermitian.

Now, we obtain

$$I^{(+)}J_1 + J_1 I^{(+)+} = \int_0^\infty ds\, K(s) \exp(-I^{(+)+}s) \tag{C.6}$$

where we used partial integration, $V(0) = 0$, $V(s) \to$ const for $s \to \infty$, and finally put $t = \infty$. Collecting the above results, we finally obtain from (C.6), (C.4), and $F = J_1 + J_1^+$

$$\begin{aligned} &I^{(+)}F + FI^{(+)+} \\ &\quad = \int_0^\infty ds\, \langle Z^{(+)}(s) | Z^{(+)+}(0) \rangle \exp(-sI^{(+)+}) + \text{h.c.} \\ &\quad = \int_{-\infty}^0 ds\, \langle Z^{(+)}(0) | Z^{(+)+}(s) \rangle \exp(sI^{(+)+}) + \text{h.c.} \end{aligned} \tag{C.7}$$

and hence Eq. (4.17).

APPENDIX D. EXPRESSIONS FOR THE TAYLOR COEFFICIENTS OF $\hat{K}(\omega)$

The given Laurent expansion of $\hat{C}$ allows us to relate in a straightforward way the coefficients K_n, $n = 1,...,$ of the Taylor expansion (6.12), where K_n corresponds to the nth moment of $K(t)$, to the macroscopic quantities. Using the properties of the Laurent expansion and the fact that the Laurent expansion of $\hat{C}$ and the Taylor expansion of $\hat{K}$ are both convergent inside the annulus, we find by comparing Eqs. (6.17a) and (6.27) by means of Eq. (6.24)

$$\alpha_0 = \frac{1}{2} K_2 + O(\xi^3)$$

$$\alpha_{-1} = K_1 + \frac{1}{2} i(K_2 I^{(+)+} - I^{(+)} K_2) + O(\xi^3)$$

$$\alpha_{-2} = K_0 + i(K_1 I^{(+)+} - I^{(+)} K_1) + O(\xi^2)$$

Using Eq. (6.25a), we resolve this as

$$K_1 = i(\Delta - \Delta^+) + O(\xi^2) \tag{D.1}$$

and

$$K_0 = \Delta I^+ + I \Delta^+ + O(\xi^2) \tag{D.2}$$

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