AIP Journal of Mathematical Physics

Contributions to the Theory of Multiplicative Stochastic Processes

Ronald Forrest Fox

Citation: J. Math. Phys. **13**, 1196 (1972); doi: 10.1063/1.1666123
View online: http://dx.doi.org/10.1063/1.1666123
View Table of Contents: http://jmp.aip.org/resource/1/JMAPAQ/v13/i8
Published by the American Institute of Physics.

Contributions to the Theory of Multiplicative Stochastic Processes

Ronald Forrest Fox
School of Physics, Georgia Institute of Technology, Atlanta, Georgia 30332
(Received 7 February 1972; Revised Manuscript Received 9 March 1972)

The theory of multiplicative stochastic processes is contrasted with the theory of additive stochastic processes. The case of multiplicative factors which are purely random, Gaussian, stochastic processes is treated in detail. In a spirit originally introduced by theoretical work in nuclear magnetic resonance and greatly extended by Kubo, dissipative behavior is demonstrated, on the average, for dynamical equations which do not show dissipative behavior without averaging. It is suggested that multiplicative stochastic processes lead to a conceptual foundation for nonequilibrium thermodynamics and nonequilibrium statistical mechanics, of marked generality.

1. INTRODUCTION

The purpose of this paper is to present results in the theory of "multiplicative stochastic processes." The physical applications of this theory will be presented in a sequel to this paper.

Effective use of stochastic processes in physics was first achieved in the theory of Brownian motion.[1] The basic ideas were generalized by Onsager and Machlup in their theory of fluctuations and irreversible processes.[2] Further generalizations, which resulted in a general stochastic theory for the linear dynamical behavior of classical thermodynamical systems, close to but not yet in full equilibrium, were presented by Fox and Uhlenbeck.[3,4] The theory of Fox and Uhlenbeck includes the Langevin theory of Brownian motion and the Onsager and Machlup theory for irreversible processes as special cases. In addition, it includes the linearized fluctuating hydrodynamical equations of Landau and Lifshitz[5] and the linearized fluctuating Boltzmann equation as special cases.

In each of these special cases, and in the general theory, the mathematical description used involves either linear partial integro-differential equations or linear matrix equations which are inhomogeneous. The inhomogeneity is the stochastic "driving force" of the process. Consequently, we shall refer to these processes as "additive stochastic processes." The processes to be presented in this paper will be seen to involve homogeneous equations in which the stochastic "driving force" enters in a multiplicative way. These processes will, consequently, be called "multiplicative stochastic processes."

Multiplicative stochastic processes arise in a natural way in the field of nuclear magnetic resonance. The nature and history of this development may be found in a paper by Redfield.[6] Major generalizations of these ideas for other areas of physics have been presented by Kubo.[7-9] Kubo has also pursued the mathematical foundations for a theory of multiplicative stochastic processes in his work. The special attention paid to purely random, Gaussian, stochastic processes in this paper will serve to further clarify and support the spirit of Kubo's earlier work.

2. MATHEMATICAL PRELIMINARIES

The fundamental stochastic process to be considered here is the purely random, stationary, Gaussian process.[10] Let $\tilde{\varphi}(t)$ denote such a process. Processes with an average value of zero will be considered throughout. This is denoted by

$$\langle\tilde{\varphi}(t)\rangle = 0. \tag{1}$$

The mean square correlation is given by

$$\langle\tilde{\varphi}(t)\,\tilde{\varphi}(s)\rangle = 2\lambda\delta(t-s), \tag{2}$$

where λ is a constant. The purely random quality of the process is reflected in the presence of $\delta(t-s)$. The dependence upon time differences only, in (2), reflects the condition of stationarity. The Gaussian property may be introduced in terms of the higher order averaged products. All odd order averaged products are zero:

$$\langle\tilde{\varphi}(t_1)\cdots\tilde{\varphi}(t_{2n-1})\rangle = 0, \quad n = 1, 2, \cdots. \tag{3}$$

All even order averaged products are given by

$$\langle\tilde{\varphi}(t_1)\cdots\tilde{\varphi}(t_{2n})\rangle = \frac{1}{2^n n!}\sum_{p\in S_{2n}}\prod_{j=1}^{n}\langle\tilde{\varphi}(t_{p(2j)})\tilde{\varphi}(t_{p(2j-1)})\rangle$$
$$= \frac{1}{2^n n!}\sum_{p\in S_{2n}} 2^n\lambda^n\prod_{j=1}^{n}\delta(t_{p(2j)}-t_{p(2j-1)}). \tag{4}$$

$\sum_{p\in S_{2n}}$ denotes the sum over all permutations p of the symmetric group of order $(2n)!$, S_{2n}. Because the two orders of the arguments of a delta function give the same value and because each arrangement of factors in a product of delta functions gives the same value, each distinct term in (4) is $(2^n n!)$-fold redundant. Since S_{2n} is of order $(2n)!$ the expression in (4) has $[(2n)!/2^n n!] = 1\cdot3\cdot5\cdots(2n-1)$ distinct terms.[11]

3. ADDITIVE STOCHASTIC PROCESSES

The prototype for the application of stochastic processes to physical phenomena is found in the theory of Brownian motion.[1,3,10,12,13] The velocity $u(t)$ of a heavy particle with mass M in a fluid which is in thermal equilibrium obeys the Langevin equation

$$M\frac{du(t)}{dt} = -\alpha u(t) + \tilde{F}(t), \tag{5}$$

where α is the dissipative, friction coefficient, and $\tilde{F}(t)$ is a purely random, stationary, Gaussian driving force. It is thought that $\tilde{F}(t)$ corresponds with the true microscopic force on the heavy particle which is produced by a great quantity of collisions in rapid succession, between the heavy particle and the molecules constituting the fluid. From a point of view which considers time on a much longer scale than the scale determined by the time between collisions, the true force may be replaced by $\tilde{F}(t)$. This means that $M/\alpha \gg \tau_c$, where τ_c measures the microscopic collision correlation time, and M/α measures the relaxation time from the macroscopic viewpoint.

By assuming that $\tilde{F}(t)$ is purely random we have that

$$\langle\tilde{F}(t)\tilde{F}(s)\rangle = 2D\delta(t-s), \tag{6}$$

which means that microscopic collision correlations last effectively "no time" in the macroscopic time scale. In this way, a purely random process is used to describe a situation involving two distinct time scales: a microscopic time scale and a macroscopic time scale. Because the fluid remains in thermal equilibrium throughout the relaxation process, $\tilde{F}(t)$ is also stationary. The Gaussian property for $\tilde{F}(t)$ may be thought to be a consequence of the central limit theorem of probability theory since $\tilde{F}(t)$ results from the collective effect of large numbers of thermally randomized collisions. Using the equipartition of energy theorem leads one to the Einstein relation

$$D = K_B T \alpha \tag{7}$$

in which T is the temperature, and K_B is Boltzmann's constant. Equation (7) is the prototype of so-called fluctuation-dissipation theorems.[3,4]

Equation (5) is manifestly inhomogeneous and exhibits the "additive" quality of this stochastic process. The process described by (5) is a one-component stationary, Gaussian, Markov process. The generalization to N-component stationary, Gaussian, Markov processes has the form[3]

$$\frac{d}{dt} a_i(t) = \sum_j A_{ij} a_j(t) + \sum_j S_{ij} a_j(t) + \tilde{F}_i(t), \tag{8}$$

where $i = 1, 2, \ldots, N$, A_{ij} is an $N \times N$ antisymmetric, real matrix, S_{ij} is an $N \times N$ symmetric, real matrix with nonpositive eigenvalues, and $\tilde{F}_i(t)$ is an N-component purely random, stationary, Gaussian "driving force". The analog to (6) is

$$\langle \tilde{F}_i(t) \tilde{F}_j(s) \rangle = 2 Q_{ij} \delta(t-s), \tag{9}$$

where Q_{ij} is a symmetric matrix with nonnegative eigenvalues. Corresponding with (7) is the general fluctuation-dissipation theorem

$$Q_{ij} = \tfrac{1}{2} \sum_k (G_{ik} E^{-1}_{kj} + E^{-1}_{ik} G_{jk}), \tag{10}$$

where $G_{ij} = A_{ij} + S_{ij}$, and E_{ij} is the entropy matrix which appears in the second-order formula for the entropy

$$S(t) = S_0 - \tfrac{1}{2} K_B \sum_i \sum_j a_i(t) E_{ij} a_j(t). \tag{11}$$

E_{ij} is symmetric and positive definite. Note that (8) is also manifestly an "additive" stochastic process, with N components. The general physical applicability of (8)–(11) suggests that the interactions generated by a macroscopic system which is fluctuating about its equilibrium state may be characterized as purely random, stationary, Gaussian "forces."

4. MULTIPLICATIVE STOCHASTIC PROCESSES

An alternative usage for stochastic processes in the description of nonequilibrium processes is possible. The prototype for this alternative method will be called "frequency fluctuation dissipation." In Kubo's work this is the example of a harmonic oscillator with a randomly modulated frequency.[7]

Consider a harmonic oscillator described by the complex variable $a(t)$. The equation of motion is

$$\frac{d}{dt} a(t) = i\omega_0 a(t), \tag{12}$$

where $i = \sqrt{-1}$, and ω_0 is the frequency of oscillation. The solution is (12), is trivial, and is

$$a(t) = e^{i\omega_0 t} a(0). \tag{13}$$

Suppose that the oscillator is at temperature T, so that those physical properties which determine ω_0 exhibit thermal fluctuations. For instance, the length of a pendulum or the spring constant of a Hooke's law spring are such properties. As a consequence, the frequency of the oscillator will fluctuate. We will assume that this frequency fluctuation may be characterized by a purely random, stationary, Gaussian process $\tilde{\varphi}(t)$ with mean value zero. The properties of $\tilde{\varphi}(t)$ are given by (1)–(4). Equation (12) becomes

$$\frac{d}{dt} a(t) = i[\omega_0 + \tilde{\varphi}(t)] a(t). \tag{14}$$

The homogeneity of (14) is manifest, and the "multiplicative" nature of the stochastic process is evident. It will be proved that the average value of (14) is

$$\frac{d}{dt} \langle a(t) \rangle = (i\omega_0 - \lambda) \langle a(t) \rangle. \tag{15}$$

The solution to (15) is clearly a damped oscillation, whereas the solution to (14), without averaging, is oscillatory. This example must be distinguished from an example of damped oscillations which arises from the Brownian motion of a harmonic oscillator.[3]

Proof of Eq. (15): The formal solution to (14) is

$$a(t) = e^{i\omega_0 t} \exp\left(i \int_0^t \tilde{\varphi}(s)\, ds\right) a(0). \tag{16}$$

Therefore,

$$\langle a(t) \rangle = e^{i\omega_0 t} \left\langle \exp\, i\left(\int_0^t \tilde{\varphi}(s)\, ds\right)\right\rangle a(0). \tag{17}$$

However,

$$\left\langle \exp\left(i \int_0^t \tilde{\varphi}(s)\, ds\right)\right\rangle = \sum_{n=0}^{\infty} \frac{(i)^n}{n!} \left\langle \left(\int_0^t \tilde{\varphi}(s)\, ds\right)^n \right\rangle. \tag{18}$$

Using (3) gives, for odd $n = 2m - 1$,

$$\left\langle \left(\int_0^t \tilde{\varphi}(s)\, ds\right)^n \right\rangle = \int_0^t \cdots \int_0^t \langle \tilde{\varphi}(s_1) \cdots \times \tilde{\varphi}(s_{2m-1}) \rangle\, ds_1 \cdots ds_{2m-1} = 0. \tag{19}$$

Using (4) gives, for even $n = 2m$,

$$\begin{aligned}
\left\langle \left(\int_0^t \tilde{\varphi}(s)\, ds\right)^n \right\rangle
&= \int_0^t \cdots \int_0^t \langle \tilde{\varphi}(s_1) \cdots \tilde{\varphi}(s_{2m}) \rangle\, ds_1 \cdots ds_{2m} \\
&= \int_0^t \cdots \int_0^t \frac{1}{2^m m!} \sum_{p \in S_{2m}} 2^m \lambda^m \times \prod_{j=1}^m \delta(s_{p(2j)} - s_{p(2j-1)})\, ds_1 \cdots ds_{2m} \\
&= \frac{\lambda^m}{m!} \sum_{p \in S_{2m}} \int_0^t \cdots \times \int_0^t \prod_{j=1}^m \delta(s_{p(2j)} - s_{p(2j-1)})\, ds_1 \cdots ds_{2m} \\
&= \frac{\lambda^m}{m!} \sum_{p \in S_{2m}} \left(\int_0^t \int_0^t \delta(s_1 - s_2)\, ds_1 ds_2\right)^m \\
&= \frac{\lambda^m}{m!} (2m)!\, t^m.
\end{aligned} \tag{20}$$

Putting (20) and (19) into (18) gives

$$\left\langle \exp\left(i \int_0^t \tilde{\varphi}(s)\,ds\right)\right\rangle = \sum_{m=0}^{\infty} \frac{(i)^{2m}}{(2m)!}\frac{\lambda^m}{m!}(2m)!\,t^m = \sum_{m=0}^{\infty} \frac{(-\lambda t)^m}{m!} = e^{-\lambda t}. \quad (21)$$

Therefore, putting (21) into (17) gives (15). This completes the proof.

Because of (15), it is clear why (14) is called frequency fluctuation dissipation. This is an example of a one-complex-component situation. There is also an N-complex-component generalization for multiplicative stochastic processes. However, it will later be shown that a multicomponent-complex situation is a special case of a multicomponent real variable generalization. Therefore, the multicomponent generalization will be given for the real variable case. The multicomponent case is proved using the purely random character of the stochastic "force" and the Gaussian property of its higher order averages.

Let $a_\alpha(t)$ for $\alpha = 1, 2, \ldots, N$ be an N-component real process which satisfies the equation

$$\frac{d}{dt} a_\alpha(t) = \sum_{\alpha'} [A_{\alpha\alpha'} + \tilde{A}_{\alpha\alpha'}(t)]\,a_{\alpha'}(t), \quad (22)$$

where $A_{\alpha\alpha'} = -A_{\alpha'\alpha}$ and $\tilde{A}_{\alpha\alpha'}(t) = -\tilde{A}_{\alpha'\alpha}(t)$. The matrix components of $\tilde{A}_{\alpha\alpha'}(t)$ will be assumed to be purely random, stationary, Gaussian processes with average values of zero, and therefore, we have

$$\langle \tilde{A}_{\alpha\alpha'}(t)\rangle = 0 \quad \text{for all } \alpha \text{ and } \alpha', \quad (23)$$

$$\langle \tilde{A}_{\alpha\beta}(t)\tilde{A}_{\mu\nu}(s)\rangle = 2Q_{\alpha\beta\mu\nu}\,\delta(t-s), \quad (24)$$

$$\langle \tilde{A}_{\mu_{2n-1}\nu_{2n-1}}(s_{2n-1}) \cdots A_{\mu_1\nu_1}(s_1)\rangle = 0, \quad (25)$$

$$\begin{aligned}\langle \tilde{A}_{\mu_{2n}\nu_{2n}}(s_{2n}) \cdots \tilde{A}_{\mu_1\nu_1}(s_1)\rangle &= \frac{1}{2^n n!} \sum_{p\in S_{2n}} \prod_{j=1}^{n} \langle \tilde{A}_{\mu_{p(2j)}\nu_{p(2j)}}(s_{p(2j)}) \times \tilde{A}_{\mu_{p(2j-1)}\nu_{p(2j-1)}}(s_{p(2j-1)})\rangle \\ &= \frac{1}{2^n n!} \sum_{p\in S_{2n}} 2^n \prod_{j=1}^{n} Q_{\mu_{p(2j)}\nu_{p(2j)}\mu_{p(2j-1)}\nu_{p(2j-1)}} \times \delta(s_{p(2j)} - s_{p(2j-1)}). \end{aligned} \quad (26)$$

The average value of (22) is

$$\frac{d}{dt}\langle a_\alpha(t)\rangle = \sum_{\alpha'} A_{\alpha\alpha'}\langle a_{\alpha'}(t)\rangle + \sum_{\alpha'}\sum_{\theta} Q_{\alpha\theta\theta\alpha'}\langle a_{\alpha'}(t)\rangle. \quad (27)$$

This is the generalization of (15).

The proof to (27) is found in the Appendix. Here, we will give a plausibility argument for (27) which is made rigorous by the more lengthy, rigorous, proof in the Appendix. The irreversibility implicit in (27) will be demonstrated following the plausibility argument.

From (22), by averaging, we get

$$\frac{d}{dt}\langle a_\alpha(t)\rangle = \sum_{\alpha'} A_{\alpha\alpha'}\langle a_{\alpha'}(t)\rangle + \sum_{\alpha'} \langle \tilde{A}_{\alpha\alpha'}(t)\,a_{\alpha'}(t)\rangle. \quad (28)$$

It is the second term on the right-hand side of (28) which needs simplification. Integrating (22) with respect to time between $t-\tau$ and t gives

$$a_\alpha(t) - a_\alpha(t-\tau) = \sum_{\alpha'} A_{\alpha\alpha'} \int_{t-\tau}^{t} a_{\alpha'}(s)\,ds + \sum_{\alpha'} \int_{t-\tau}^{t} \tilde{A}_{\alpha\alpha'}(s)\,a_{\alpha'}(s)\,ds. \quad (29)$$

Multiplying (29) by $\tilde{A}_{\beta\alpha}(t)$, and summing over α gives, upon averaging the sum,

$$\begin{aligned}\sum_\alpha \langle \tilde{A}_{\beta\alpha}(t)\,a_\alpha(t)\rangle - \sum_\alpha \langle \tilde{A}_{\beta\alpha}(t)\,a_\alpha(t-\tau)\rangle &= \sum_\alpha\sum_{\alpha'} A_{\alpha\alpha'} \int_{t-\tau}^{t} \langle \tilde{A}_{\beta\alpha}(t)\,a_{\alpha'}(s)\rangle\,ds \\ &+ \sum_\alpha\sum_{\alpha'} \int_{t-\tau}^{t} \langle \tilde{A}_{\beta\alpha}(t)\tilde{A}_{\alpha\alpha'}(s)\,a_{\alpha'}(s)\rangle\,ds. \end{aligned} \quad (30)$$

Now, it seems plausible that because of (24) and (26) that

$$\langle \tilde{A}_{\beta\alpha}(t)\,a_\alpha(t-\tau)\rangle = 0 \quad (31)$$

and

$$\int_{t-\tau}^{t} \langle \tilde{A}_{\beta\alpha}(t)\,a_{\alpha'}(s)\rangle\,ds = 0. \quad (32)$$

Using (31) and (32) in (30) will give

$$\sum_\alpha \langle \tilde{A}_{\beta\alpha}(t)\,a_\alpha(t)\rangle = \sum_\alpha\sum_{\alpha'} \int_{t-\tau}^{t} \langle \tilde{A}_{\beta\alpha}(t)\tilde{A}_{\alpha\alpha'}(s)\,a_{\alpha'}(s)\rangle\,ds. \quad (33)$$

Using the Gaussian property of $\tilde{A}_{\mu\nu}(t)$ and (23) makes it plausible that

$$\sum_\alpha\sum_{\alpha'} \int_{t-\tau}^{t} \langle \tilde{A}_{\beta\alpha}(t)\tilde{A}_{\alpha\alpha'}(s)\,a_{\alpha'}(s)\rangle\,ds = \sum_\alpha\sum_{\alpha'} \int_{t-\tau}^{t} \langle \tilde{A}_{\beta\alpha}(t)\tilde{A}_{\alpha\alpha'}(s)\rangle\langle a_{\alpha'}(s)\rangle\,ds. \quad (34)$$

For the right-hand side of (34) we use (24) and get

$$\begin{aligned}\sum_\alpha\sum_{\alpha'} \int_{t-\tau}^{t} \langle \tilde{A}_{\beta\alpha}(t)\tilde{A}_{\alpha\alpha'}(s)\rangle\langle a_{\alpha'}(s)\rangle\,ds &= \sum_\alpha\sum_{\alpha'} \int_{t-\tau}^{t} 2Q_{\beta\alpha\alpha\alpha'}\,\delta(t-s)\langle a_{\alpha'}(s)\rangle\,ds \\ &= \sum_\alpha\sum_{\alpha'} Q_{\beta\alpha\alpha\alpha'}\langle a_{\alpha'}(t)\rangle. \end{aligned} \quad (35)$$

Putting (35) with (34) into (33) gives

$$\sum_\alpha \langle \tilde{A}_{\beta\alpha}(t)\,a_\alpha(t)\rangle = \sum_{\alpha'}\sum_{\alpha} Q_{\beta\alpha\alpha\alpha'}\langle a_{\alpha'}(t)\rangle. \quad (36)$$

Returning to (28) with (36) gives (27), if we simply rename indices. This plausibility argument depends upon the truth of (31), (32), and (34). In the Appendix it is shown that the result obtained in (27) is rigorously achieved.

5. IRREVERSIBILITY

The irreversibility in (15) is obvious. That of (27) is less easily seen. To see that irreversibility arises from averaging, we will consider both $\sum_\alpha a_\alpha(t)\,a_\alpha(t)$ and $\sum_\alpha \langle a_\alpha(t)\rangle\langle a_\alpha(t)\rangle$ using both (22) and (27).

Using (22) and the antisymmetry of both $A_{\alpha\alpha'}$ and $\tilde{A}_{\alpha\alpha'}(t)$ gives

$$\frac{d}{dt}\sum_\alpha a_\alpha(t)\,a_\alpha(t) = 2\sum_\alpha\sum_{\alpha'} a_\alpha(t)A_{\alpha\alpha'}a_{\alpha'}(t) + 2\sum_\alpha\sum_{\alpha'} a_\alpha(t)\tilde{A}_{\alpha\alpha'}(t)\,a_{\alpha'}(t) = 0. \quad (37)$$

Therefore $\sum_\alpha a_\alpha(t) a_\alpha(t)$ is a conserved quantity during the unaveraged time evolution. Using (27) and the antisymmetry of $A_{\alpha\alpha'}$ gives

$$\frac{d}{dt}\sum_\alpha \langle a_\alpha(t)\rangle\langle a_\alpha(t)\rangle = 2\sum_\alpha\sum_{\alpha'}\langle a_\alpha(t)\rangle A_{\alpha\alpha'}\langle a_{\alpha'}(t)\rangle + 2\sum_\alpha\sum_{\alpha'}\sum_\theta \langle a_\alpha(t)\rangle Q_{\alpha\theta\theta\alpha'}\langle a_{\alpha'}(t)\rangle = 2\sum_\alpha\sum_{\alpha'}\sum_\theta \langle a_\alpha(t)\rangle Q_{\alpha\theta\theta\alpha'}\langle a_{\alpha'}(t)\rangle. \quad (38)$$

From (24) it follows that

$$\sum_\theta \langle \tilde{A}_{\alpha\theta}(t) A_{\theta\alpha'}(s)\rangle = 2\sum_\theta Q_{\alpha\theta\theta\alpha'}\delta(t-s). \quad (39)$$

Let y_α be an arbitrary N-component vector. Using (39) gives

$$\begin{aligned}\sum_\alpha\sum_{\alpha'}\sum_\theta y_\alpha Q_{\alpha\theta\theta\alpha'}y_{\alpha'} &= \sum_\alpha\sum_{\alpha'}\sum_\theta 2y_\alpha Q_{\alpha\theta\theta\alpha'}y_{\alpha'}\int_0^t \delta(t-s)ds \\ &= \sum_\alpha\sum_{\alpha'}\sum_\theta \int_0^t y_\alpha\langle\tilde{A}_{\alpha\theta}(t)\tilde{A}_{\theta\alpha'}(s)\rangle y_{\alpha'}ds \\ &= -\sum_\theta\int_0^t \left\langle\left(\sum_\alpha \tilde{A}_{\theta\alpha}(t)y_\alpha\right)\left(\sum_{\alpha'}\tilde{A}_{\theta\alpha'}(s)y_{\alpha'}\right)\right\rangle ds \\ &\leq 0. \end{aligned} \quad (40)$$

The last equality in (40) follows from the antisymmetry of the matrix $\tilde{A}_{\mu\nu}(t)$, and the inequality follows from the form of the integral. Putting the results expressed by (40) into (38) gives

$$\frac{d}{dt}\sum_\alpha \langle a_\alpha(t)\rangle\langle a_\alpha(t)\rangle = +2\sum_\alpha\sum_{\alpha'}\sum_\theta\langle a_\alpha(t)\rangle \times Q_{\alpha\theta\theta\alpha'}\langle a_{\alpha'}(t)\rangle \leq 0. \quad (41)$$

Therefore the quantity $\sum_\alpha \langle a_\alpha(t)\rangle\langle a_\alpha(t)\rangle$ shows a monotonic decrease to its equilibrium value. The inequality in (40) shows that the matrix $\sum_\theta Q_{\alpha\theta\theta\alpha'}$ is a symmetric matrix with nonpositive eigenvalues. If all the eigenvalues of $\sum_\theta Q_{\alpha\theta\theta\alpha'}$ are also nonzero, then the equilibrium state corresponds with $\langle a_\alpha\rangle = 0$ for all $\alpha = 1, 2, \ldots, N$. The possibility of zero value eigenvalues of $\sum_\theta Q_{\alpha\theta\theta\alpha'}$ corresponds with the possibility of certain linear combinations of the $\langle a_\alpha(t)\rangle$'s being conserved quantities during the overall approach to equilibrium. In this case equilibrium is not characterized by $\langle a_\alpha\rangle = 0$ for all $\alpha = 1, 2, \ldots, N$; for some α, $\langle a_\alpha\rangle \neq 0$.

6. COMPLEX COMPONENT CASE

A problem closely related to the real case just described involves N complex components $C_\alpha(t)$ for $\alpha = 1, 2, \ldots, N$ satisfying the equation

$$i\frac{d}{dt}C_\alpha(t) = \sum_{\alpha'} M_{\alpha\alpha'}C_{\alpha'}(t) + \sum_{\alpha'}\tilde{M}_{\alpha\alpha'}(t)C_{\alpha'}(t). \quad (42)$$

Both $M_{\alpha\alpha'}$ and $\tilde{M}_{\alpha\alpha'}(t)$ are complex Hermitian matrices. Therefore

$$M^*_{\alpha\alpha'} = M_{\alpha'\alpha} \quad \text{and} \quad \tilde{M}^*_{\alpha\alpha'}(t) = \tilde{M}_{\alpha'\alpha}(t). \quad (43)$$

$\tilde{M}_{\alpha\alpha'}(t)$ is also a purely random, stationary, Gaussian process with average zero. This implies, in analogy with (23)–(26) that

$$\langle \tilde{M}_{\alpha\alpha'}(t)\rangle = 0, \quad (44)$$

$$\langle \tilde{M}_{\alpha\beta}(t)\tilde{M}_{\mu\nu}(s)\rangle = 2Q'_{\alpha\beta\mu\nu}\delta(t-s), \quad (45)$$

$$\langle \tilde{M}_{\mu_{2n-1}\nu_{2n-1}}(s_{2n-1})\cdots\tilde{M}_{\mu_1\nu_1}(s_1)\rangle = 0, \quad (46)$$

$$\begin{aligned}\langle \tilde{M}_{\mu_{2n}\nu_{2n}}(s_{2n})\cdots\tilde{M}_{\mu_1\nu_1}(s_1)\rangle &= \frac{1}{2^n n!}\sum_{p\in S_{2n}}\prod_{j=1}^n \langle \tilde{M}_{\mu_{p(2j)}\nu_{p(2j)}}(s_{p(2j)}) \times \tilde{M}_{\mu_{p(2j-1)}\nu_{p(2j-1)}}(s_{p(2j-1)})\rangle \\ &= \frac{1}{2^n n!}\sum_{p\in S_{2n}} 2^n\prod_{j=1}^n Q'_{\mu_{p(2j)}\nu_{p(2j)}\mu_{p(2j-1)}\nu_{p(2j-1)}} \times \delta(s_{p(2j)} - s_{p(2j-1)}).\end{aligned} \quad (47)$$

It will now be shown that the analog to (27) is

$$\frac{d}{dt}\langle C_\alpha(t)\rangle = -i\sum_{\alpha'} M_{\alpha\alpha'}\langle C_{\alpha'}(t)\rangle - \sum_{\alpha'}\sum_\theta Q'_{\alpha\theta\theta\alpha'}\langle C_{\alpha'}(t)\rangle \quad (48)$$

and that $\sum_\theta Q'_{\alpha\theta\theta\alpha'}$ is Hermitian with nonnegative eigenvalues.

Each complex component $C_\alpha(t)$ may be written as

$$C_\alpha(t) = a_\alpha(t) + ib_\alpha(t), \quad (49)$$

wherein $a_\alpha(t)$ and $b_\alpha(t)$ are both real. $M_{\alpha\alpha'}$ and $\tilde{M}_{\alpha\alpha'}(t)$ may be written as

$$M_{\alpha\alpha'} = S_{\alpha\alpha'} + iA_{\alpha\alpha'}, \quad (50)$$

$$\tilde{M}_{\alpha\alpha'}(t) = \tilde{S}_{\alpha\alpha'}(t) + i\tilde{A}_{\alpha\alpha'}(t), \quad (51)$$

wherein $S_{\alpha\alpha'}, A_{\alpha\alpha'}, \tilde{S}_{\alpha\alpha'}(t)$, and $\tilde{A}_{\alpha\alpha'}(t)$ are defined by

$$S_{\alpha\alpha'} = \tfrac{1}{2}(M_{\alpha\alpha'} + M^*_{\alpha\alpha'}), \quad (52)$$

$$A_{\alpha\alpha'} = -\tfrac{1}{2}(M_{\alpha\alpha'} - M^*_{\alpha\alpha'}), \quad (53)$$

$$\tilde{S}_{\alpha\alpha'}(t) = \tfrac{1}{2}[\tilde{M}_{\alpha\alpha'}(t) + \tilde{M}^*_{\alpha\alpha'}(t)], \quad (54)$$

$$\tilde{A}_{\alpha\alpha'}(t) = -\tfrac{1}{2}[\tilde{M}_{\alpha\alpha'}(t) - \tilde{M}^*_{\alpha\alpha'}(t)]. \quad (55)$$

With (43) it is seen that $S_{\alpha\alpha'}, A_{\alpha\alpha'}, \tilde{S}_{\alpha\alpha'}(t)$, and $\tilde{A}_{\alpha\alpha'}(t)$ are real matrices and that $S_{\alpha\alpha'}$, and $\tilde{S}_{\alpha\alpha'}(t)$ are symmetric, while $A_{\alpha\alpha'}$ and $\tilde{A}_{\alpha\alpha'}(t)$ are antisymmetric. Using (49)–(51), (42) can be rewritten as

$$\frac{d}{dt}\begin{pmatrix}a_\alpha(t)\\ b_\alpha(t)\end{pmatrix} = \sum_{\alpha'=1}\begin{pmatrix}A_{\alpha\alpha'} & S_{\alpha\alpha'}\\ -S_{\alpha\alpha'} & A_{\alpha\alpha'}\end{pmatrix}\begin{pmatrix}a_{\alpha'}(t)\\ b_{\alpha'}(t)\end{pmatrix} + \sum_{\alpha'=1}^N\begin{pmatrix}\tilde{A}_{\alpha\alpha'}(t) & \tilde{S}_{\alpha\alpha'}(t)\\ -\tilde{S}_{\alpha\alpha'}(t) & \tilde{A}_{\alpha\alpha'}(t)\end{pmatrix}\begin{pmatrix}a_{\alpha'}(t)\\ b_{\alpha'}(t)\end{pmatrix}. \quad (56)$$

Note that $\binom{a_\alpha(t)}{b_\alpha(t)}$ is a column vector with $2N$ real-valued components. Denote it by $a'_\beta(t)$, where

$$a'_\beta(t) \equiv a_\beta(t) \text{ for } \beta = 1, 2, \ldots, N \quad (57)$$

and

$$a'_\beta(t) \equiv b_{\beta-N}(t) \quad \text{for } \beta = N+1, N+2, \ldots, 2N.$$

In the same spirit, $A'_{\beta\beta'}$ will denote the antisymmetric matrix

$$\begin{pmatrix} A_{\alpha\alpha'} & S_{\alpha\alpha'} \\ -S_{\alpha\alpha'} & A_{\alpha\alpha'} \end{pmatrix},$$

where

$$A'_{\beta\beta'} \equiv A_{\beta\beta'} \quad \text{for } \beta = 1, 2, \ldots, N \text{ and } \beta' = 1, 2, \ldots, N,$$
$$A'_{\beta\beta'} \equiv S_{\beta\beta'-N} \quad \text{for } \beta = 1, 2, \ldots, N \text{ and } \beta' = N+1, N+2, \ldots, 2N,$$
$$A'_{\beta\beta'} \equiv S_{\beta-N\beta'} \quad \text{for } \beta = N+1, N+2, \ldots, 2N \text{ and } \beta' = 1, 2, \ldots, N,$$

and

$$A'_{\beta\beta'} \equiv A_{\beta-N\beta'-N} \quad \text{for } \beta = N+1, N+2, \ldots, 2N \text{ and } \beta' = N+1, N+2, \ldots, 2N. \tag{58}$$

In a similar manner, define the $2N \times 2N$ real antisymmetric matrix $\tilde{A}'_{\beta\beta'}(t)$ in terms of $\tilde{S}_{\alpha\alpha'}(t)$ and $\tilde{A}_{\alpha\alpha'}(t)$. With these definitions (56) becomes

$$\frac{d}{dt} a'_\beta(t) = \sum_{\beta'=1}^{2N} A'_{\beta\beta'} a'_{\beta'}(t) + \sum_{\beta'=1}^{2N} \tilde{A}'_{\beta\beta'}(t) a'_{\beta'}(t), \tag{59}$$

which is a special case of (22).

In order to get the analogue of (27) for (59) it is necessary to determine the matrix $\sum_{\theta=1}^{2N} Q''_{\beta\theta\theta\beta'}$ defined by

$$\sum_{\theta=1}^{2N} \langle \tilde{A}'_{\beta\theta}(t) \tilde{A}'_{\theta\beta'}(s) \rangle \equiv 2 \sum_{\theta=1}^{2N} Q''_{\beta\theta\theta\beta'} \delta(t-s). \tag{60}$$

The left-hand side of (60) is computed by using the definition of $\tilde{A}'_{\beta\beta'}(t)$, (54), (55), and (45), in that order. The computation is straightforward and somewhat long. The results are

$$\sum_{\theta=1}^{2N} Q''_{\beta\theta\theta\beta'} = -\frac{1}{2}\left(\sum_{\theta=1}^{N} Q'_{\beta\theta\theta\beta'} + \sum_{\theta=1}^{N} Q'^{*}_{\beta\theta\theta\beta'}\right) \quad \text{for } \beta = 1, 2, \ldots, N \text{ and } \beta' = 1, 2, \ldots, N;$$
$$\sum_{\theta=1}^{2N} Q''_{\beta\theta\theta\beta'} = -\frac{i}{2}\left(\sum_{\theta=1}^{N} Q'_{\beta\theta\theta\beta'-N} - \sum_{\theta=1}^{N} Q'^{*}_{\beta\theta\theta\beta'-N}\right) \quad \text{for } \beta = 1, 2, \ldots, N \text{ and } \beta' = N+1, N+2, \ldots, 2N;$$
$$\sum_{\theta=1}^{2N} Q''_{\beta\theta\theta\beta'} = -\frac{i}{2}\left(\sum_{\theta=1}^{N} Q'_{\beta-N\theta\theta\beta'} - \sum_{\theta=1}^{N} Q'^{*}_{\beta-N\theta\theta\beta'}\right) \quad \text{for } \beta = N+1, N+2, \ldots, 2N \text{ and } \beta' = 1, 2, \ldots, N; \tag{61}$$
$$\sum_{\theta=1}^{2N} Q''_{\beta\theta\theta\beta'} = -\frac{1}{2}\left(\sum_{\theta=1}^{N} Q'_{\beta-N\theta\theta\beta'-N} - \sum_{\theta=1}^{N} Q'^{*}_{\beta-N\theta\theta\beta'-N}\right) \quad \text{for } \beta = N+1, N+2, \ldots, 2N \text{ and } \beta' = N+1, N+2, \ldots, 2N.$$

At this point, the use of (49)–(51) and (61) leads to (48) if one notices that

$$\sum_{\theta=1}^{N} Q'_{\alpha\theta\theta\alpha'} = \frac{1}{2}\left(\sum_{\theta=1}^{N} Q'_{\alpha\theta\theta\alpha'} + \sum_{\theta=1}^{N} Q'^{*}_{\alpha\theta\theta\alpha'}\right) - i\,\frac{i}{2}\left(\sum_{\theta=1}^{N} Q'_{\alpha\theta\theta\alpha'} - \sum_{\theta=1}^{N} Q'^{*}_{\alpha\theta\theta\alpha'}\right), \tag{62}$$

where

$$\frac{1}{2}\left(\sum_{\theta=1}^{N} Q'_{\alpha\theta\theta\alpha'} + \sum_{\theta=1}^{N} Q'^{*}_{\alpha\theta\theta\alpha'}\right)$$

and

$$\frac{i}{2}\left(\sum_{\theta=1}^{N} Q'_{\alpha\theta\theta\alpha'} - \sum_{\theta=1}^{N} Q'^{*}_{\alpha\theta\theta\alpha'}\right)$$

are both real matrices. Therefore, it has been justified that (42) and (48) are a special case of (22) and (27).

Via (45) it is seen that

$$\begin{aligned}
\sum_{\alpha}\sum_{\alpha'}\sum_{\theta} y^{*}_{\alpha} Q'_{\alpha\theta\theta\alpha'} y_{\alpha'}
&= \sum_{\alpha}\sum_{\alpha'}\sum_{\theta} 2 y^{*}_{\alpha} Q'_{\alpha\theta\theta\alpha'} y_{\alpha'} \int_0^t \delta(t-s)\,ds \\
&= \sum_{\alpha}\sum_{\alpha'}\sum_{\theta} \int_0^t y^{*}_{\alpha} \langle \tilde{M}_{\alpha\theta}(t) \tilde{M}_{\theta\alpha'}(s) \rangle y_{\alpha'}\,ds \\
&= \sum_{\theta} \int_0^t \left\langle \left(\sum_{\alpha} \tilde{M}^{*}_{\theta\alpha}(t) y^{*}_{\alpha}\right)\left(\sum_{\alpha'} \tilde{M}_{\theta\alpha'}(s) y_{\alpha'}\right)\right\rangle ds \\
&\geq 0.
\end{aligned} \tag{63}$$

Therefore, $\sum_\theta Q'_{\alpha\theta\theta\alpha'}$ has nonnegative eigenvalues, and with (48) it is seen that the quantity $\sum_\alpha \langle C^{*}_{\alpha}(t)\rangle \langle C_\alpha(t)\rangle$ shows a monotonic decrease to equilibrium; whereas from (42) it is seen that the quantity $\sum_\alpha C^{*}_{\alpha}(t) C_\alpha(t)$ is a time invariant. These results are analogs of (41) and (37), respectively.

7. COMPLEX BILINEAR FORMS

Starting with Eq. (42), it is possible to define the matrix $\rho_{\alpha\beta}(t)$ by

$$\rho_{\alpha\beta}(t) \equiv C^{*}_{\alpha}(t) C_\beta(t) \tag{64}$$

and to ask what the time dependence equations for $\rho_{\alpha\beta}(t)$ and $\langle \rho_{\alpha\beta}(t)\rangle$ are. One gets from (42)

$$i\frac{d}{dt}\rho_{\alpha\beta}(t) = \sum_{\alpha'}\sum_{\beta'} \left(L_{\alpha\beta\alpha'\beta'} + \tilde{L}_{\alpha\beta\alpha'\beta'}(t)\right)\rho_{\alpha'\beta'}(t), \tag{65}$$

wherein $L_{\alpha\beta\alpha'\beta'}$ and $\tilde{L}_{\alpha\beta\alpha'\beta'}(t)$ are defined by

$$L_{\alpha\beta\alpha'\beta'} \equiv \delta_{\alpha\alpha'} M_{\beta\beta'} - \delta_{\beta\beta'} M^{*}_{\alpha\alpha'}, \tag{66}$$

$$\tilde{L}_{\alpha\beta\alpha'\beta'}(t) \equiv \delta_{\alpha\alpha'} \tilde{M}_{\beta\beta'}(t) - \delta_{\beta\beta'} \tilde{M}^{*}_{\alpha\alpha'}(t). \tag{67}$$

Note that (43) implies that

$$L^{*}_{\alpha\beta\alpha'\beta'} = L_{\alpha'\beta'\alpha\beta} \quad \text{and} \quad \tilde{L}^{*}_{\alpha\beta\alpha'\beta'}(t) = \tilde{L}_{\alpha'\beta'\alpha\beta}(t). \tag{68}$$

Both indices α and β range over $1, 2, \ldots, N$. Therefore it is possible to think of $\rho_{\alpha\beta}(t)$ as an N^2 component "vector," and to think of $L_{\alpha\beta\alpha'\beta'}$ and $\tilde{L}_{\alpha\beta\alpha'\beta'}(t)$ as $N^2 \times N^2$ "matrices." Equation (68) suggests that these two "matrices" are Hermitian. Equation (67) shows that $\tilde{L}_{\alpha\beta\alpha'\beta'}(t)$ is a linear combination of two purely random, stationary, Gaussian processes and is, therefore, itself a purely random, stationary, Gaussian process. Consequently, in this way of viewing (65) it is seen that (65) is a special case of (42), as well as being derived from (42). Therefore, $\langle \rho_{\alpha\beta}(t)\rangle$ will obey an equation which is the analog to (48).

In order to get the equation for $\langle \rho_{\alpha\beta}(t)\rangle$ it is necessary to obtain the analog of $\sum_\theta Q'_{\alpha\theta\theta\alpha'}$ which appears in (48). Comparing (65) with (42) it is seen that one needs the analog of (45) which is

$$\langle \tilde{L}_{\alpha\beta\alpha'\beta'}(t)\tilde{L}_{\mu\nu\mu'\nu'}(s)\rangle = 2Q'_{\alpha\beta\alpha'\beta'\mu\nu\mu'\nu'}\delta(t-s). \quad (69)$$

In order to explicitly determine $Q'_{\alpha\beta\alpha'\beta'\mu\nu\mu'\nu'}$, one uses (67) and also (45). The exercise of a little algebra yields

$$Q'_{\alpha\beta\alpha'\beta'\mu\nu\mu'\nu'} = \delta_{\alpha\alpha'}\delta_{\mu\mu'}Q'_{\beta\beta'\nu\nu'} + \delta_{\beta\beta'}\delta_{\nu\nu'}Q'^{*}_{\alpha\alpha'\mu\mu'} - \delta_{\alpha\alpha'}\delta_{\nu\nu'}Q'_{\beta\beta'\mu'\mu} - \delta_{\beta\beta'}\delta_{\mu\mu'}Q'_{\alpha'\alpha\nu\nu'}. \quad (70)$$

Therefore, renaming indices leads to the analog of $\sum_\theta Q'_{\alpha\theta\theta\alpha'}$ which is

$$\sum_\theta \sum_{\theta'} Q'_{\alpha\beta\theta\theta'\theta\theta'\alpha'\beta'} = \delta_{\alpha\alpha'}\sum_{\theta'} Q'_{\beta\theta'\theta'\beta'} + \delta_{\beta\beta'}\sum_\theta Q'^{*}_{\alpha\theta\theta\alpha'} - Q'_{\beta\beta'\alpha'\alpha} - Q'_{\alpha'\alpha\beta\beta'}. \quad (71)$$

Use of (45) shows that

$$Q'^{*}_{\alpha\theta\theta\alpha'} = Q'_{\theta\alpha\alpha'\theta}. \quad (72)$$

If the left-hand side of (71) is defined to be $R_{\alpha\beta\alpha'\beta'}$, then (71) and (72) give

$$R_{\alpha\beta\alpha'\beta'} = \delta_{\alpha\alpha'}\sum_\theta Q'_{\beta\theta\theta\beta'} + \delta_{\beta\beta'}\sum_\theta Q'_{\theta\alpha\alpha'\theta} - Q'_{\beta\beta'\alpha'\alpha} - Q'_{\alpha'\alpha\beta\beta'}. \quad (73)$$

Consequently, the analog to (48) is

$$\frac{d}{dt}\langle \rho_{\alpha\beta}(t)\rangle = -i\sum_{\alpha'}\sum_{\beta'} L_{\alpha\beta\alpha'\beta'}\langle \rho_{\alpha'\beta'}(t)\rangle - \sum_{\alpha'}\sum_{\beta'} R_{\alpha\beta\alpha'\beta'}\langle \rho_{\alpha'\beta'}(t)\rangle \quad (74)$$

The analog to the proof to (63) may be applied to $R_{\alpha\beta\alpha'\beta'}$ by using (69). Therefore, for an arbitrary matrix $X_{\alpha\beta}$, which is thought of as an N^2 component "vector," it follows that

$$\sum_\alpha\sum_\beta\sum_{\alpha'}\sum_{\beta'} X^*_{\alpha\beta}R_{\alpha\beta\alpha'\beta'}X_{\alpha'\beta'} \geq 0. \quad (75)$$

Thus the eigen-"vectors" of $R_{\alpha\beta\alpha'\beta'}$ are really the eigenmatrices of a tetratic, and the eigenvalues are nonnegative. The case of a zero eigenmatrix, or eigen-"vector," is realized by using (69) and (67) which show that the identity matrix $\delta_{\alpha'\beta'}$ is an eigen-"vector" eigenmatrix with eigenvalue zero:

$$\sum_{\alpha'}\sum_{\beta'}\tilde{L}_{\alpha\beta\alpha'\beta'}(t)\delta_{\alpha'\beta'} = \sum_{\alpha'}\sum_{\beta'}[\delta_{\alpha\alpha'}\tilde{M}_{\beta\beta'}(t) - \delta_{\beta\beta'}\tilde{M}^*_{\alpha\alpha'}(t)]\delta_{\alpha'\beta'} = \tilde{M}_{\beta\alpha}(t) - \tilde{M}^*_{\alpha\beta}(t) = 0. \quad (76)$$

Therefore, it also follows that

$$\sum_{\alpha'}\sum_{\beta'} R_{\alpha\beta\alpha'\beta'}\delta_{\alpha'\beta'} = \sum_\theta R_{\alpha\beta\theta\theta} = 0. \quad (77)$$

The symmetry of $R_{\alpha\beta\alpha'\beta'}$, which follows from (69), implies

$$\sum_\theta R_{\theta\theta\alpha'\beta'} = 0. \quad (78)$$

Therefore,

$$\frac{d}{dt}\sum_\alpha\langle \rho_{\alpha\alpha}(t)\rangle = -i\sum_\alpha\sum_{\alpha'}\sum_{\beta'} L_{\alpha\alpha\alpha'\beta'}\langle \rho_{\alpha'\beta'}(t)\rangle - \sum_\alpha\sum_{\alpha'}\sum_{\beta'} R_{\alpha\alpha\alpha'\beta'}\langle \rho_{\alpha'\beta'}(t)\rangle = 0 \quad (79)$$

because of (78) and a result like (76) which follows from (66):

$$\sum_\alpha L_{\alpha\alpha\alpha'\beta'} = \sum_\alpha (\delta_{\alpha\alpha'}M_{\alpha\beta'} - \delta_{\alpha\beta'}M^*_{\alpha\alpha'}) = M_{\alpha'\beta'} - M^*_{\beta'\alpha'} = 0. \quad (80)$$

Therefore, $\sum_\alpha \langle \rho_{\alpha\alpha}(t)\rangle$ is a conserved quantity. Nevertheless, (75) guarantees that (74) shows irreversible behavior.

8. PURELY DIAGONAL BILINEAR BEHAVIOR

Again starting with (42), it is always possible to perform a unitary similarity transformation which diagonalizes $M_{\alpha\alpha'}$ since $M_{\alpha\alpha'}$ is Hermitian. $\tilde{M}_{\alpha\alpha'}(t)$ in the new representation will not necessarily be diagonal, but it will still be Hermitian and a purely random, stationary, Gaussian process. Therefore, without loss of generality, (42) can always be transformed into the form

$$i\frac{d}{dt}c_\alpha(t) = d_\alpha c_\alpha(t) + \sum_{\alpha'}\tilde{M}_{\alpha\alpha'}(t)c_{\alpha'}(t), \quad (81)$$

wherein the d_α are real numbers. This is equivalent with saying that $M_{\alpha\alpha'}$ is diagonal and is given by

$$M_{\alpha\alpha'} = d_\alpha\delta_{\alpha\alpha'}. \quad (82)$$

The program of Sec. 7 can again be carried through with the simple modification that

$$L_{\alpha\beta\alpha'\beta'} \equiv \delta_{\alpha\alpha'}M_{\beta\beta'} - \delta_{\beta\beta'}M^*_{\alpha\alpha'} = \delta_{\alpha\alpha'}\delta_{\beta\beta'}d_\beta - \delta_{\beta\beta'}\delta_{\alpha\alpha'}d_\alpha = (d_\beta - d_\alpha)\delta_{\alpha\alpha'}\delta_{\beta\beta'}. \quad (83)$$

Therefore, (74) becomes

$$\frac{d}{dt}\langle \rho_{\alpha\beta}(t)\rangle = -i(d_\beta - d_\alpha)\langle \rho_{\alpha\beta}(t)\rangle - \sum_{\alpha'}\sum_{\beta'} R_{\alpha\beta\alpha'\beta'}\langle \rho_{\alpha'\beta'}(t)\rangle. \quad (84)$$

Using this diagonal $-M_{\alpha\alpha'}$ representation,

$$\langle \tilde{M}_{\alpha\beta}(t)\tilde{M}_{\mu\nu}(s)\rangle = 2Q'_{\alpha\beta\mu\nu}[\delta_{\alpha\nu}\delta_{\beta\mu} + \delta_{\alpha\mu}\delta_{\beta\nu} + (1-\delta_{\alpha\nu})(1-\delta_{\beta\mu})(1-\delta_{\alpha\mu})(1-\delta_{\beta\nu})]\delta(t-s) \quad (85)$$

is a sufficient condition for the reduction of (84) into an equation involving only the diagonal elements of $\langle \rho_{\alpha\beta}(t)\rangle$. The Kronecker delta factors in (85) require that either α, β, μ, and ν are all different, or that either $\alpha = \beta$ *or* $\mu = \nu$, *or* $\alpha = \nu$ *and* $\beta = \mu$, *or* $\alpha = \mu$ *and* $\beta = \nu$, in order that the over-all quantity be nonzero.

Sufficiency is demonstrated by using (85) in place of (45) in the calculation of $R_{\alpha\beta\alpha'\beta'}$ as determined by (73). This is equivalent with replacing the occurrence of $Q'_{\alpha\beta\mu\nu}$ in (73) with $Q'_{\alpha\beta\mu\nu}[\delta_{\alpha\nu}\delta_{\beta\mu} + \delta_{\alpha\mu}\delta_{\beta\mu} + (1-\delta_{\alpha\nu})(1-\delta_{\beta\mu}) \times (1-\delta_{\alpha\mu})(1-\delta_{\beta\nu})]$. The result, after a modicum of computation, is

$$R_{\alpha\beta\alpha'\beta'} = \delta_{\alpha\alpha'}\delta_{\beta\beta'}\Big(\sum_\theta Q'_{\beta\theta\theta\beta'} + \sum_\theta Q'_{\theta\alpha\alpha'\theta} + Q'_{\beta\beta\beta\beta} + Q'_{\alpha\alpha\alpha\alpha}\Big) - 2Q'_{\alpha'\alpha\beta\beta'}[\delta_{\alpha'\beta}\delta_{\alpha\beta'} + \delta_{\alpha\beta}\delta_{\alpha'\beta'} + (1-\delta_{\alpha'\beta})(1-\delta_{\alpha\beta'}) \times (1-\delta_{\alpha\beta})(1-\delta_{\alpha'\beta'})]. \quad (86)$$

Note that (86) implies that in order for $R_{\alpha\beta\alpha'\beta'} \neq 0$, then

$$\alpha = \beta \quad \text{if and only if} \quad \alpha' = \beta'. \quad (87)$$

Consequently, the expression for $R_{\alpha\beta\alpha'\beta'}$ given by (86) will reduce (84) to an equation for the diagonal elements of $\langle \rho_{\alpha\beta}(t)\rangle$ only. Define $P_\alpha(t)$ by

$$P_\alpha(t) \equiv \langle \rho_{\alpha\alpha}(t)\rangle. \quad (88)$$

By using (88), the diagonal equation resulting from (84) is

$$\frac{d}{dt} P_\alpha(t) = \sum_{\alpha'} [W_{\alpha\alpha'}P_{\alpha'}(t) - W_{\alpha'\alpha}P_\alpha(t)], \quad (89)$$

where $W_{\alpha\alpha'}$ is defined by

$$W_{\alpha\alpha'} \equiv 2Q'_{\alpha'\alpha\alpha\alpha'}. \quad (90)$$

Equation (90) holds because (86) leads to the result

$$R_{\alpha\alpha\alpha'\alpha'} = \delta_{\alpha\alpha'} \sum_\theta 2Q'_{\alpha\theta\theta\alpha} - 2Q'_{\alpha'\alpha\alpha\alpha'}. \quad (91)$$

Returning to (45) it is seen that

$$W_{\alpha\alpha'} \geq 0. \quad (92)$$

In addition, (79) may be rewritten using (88) to yield

$$\frac{d}{dt} \sum_\alpha P_\alpha(t) = 0. \quad (93)$$

9. FOKKER–PLANCK EQUATION

Because the stochastic "driving force" for the multiplicative stochastic processes presented here is always characterized as a purely random process, as well as a stationary, Gaussian process, the resulting over-all stochastic process is a Markov process. In this section, the Fokker–Planck equation which follows from the Markov property will be presented for the real N-component case. As was demonstrated in Secs. 6 and 7, the complex N-component case and the complex bilinear case are special cases of the real component case. Therefore, the Fokker–Planck equation presented below for the real component case is sufficiently general to cover all of these cases.

The Markov property alone does not necessarily lead to a Fokker–Planck equation. The following conditions are also necessary[14]:

$$\lim_{\Delta t\to 0} (1/\Delta t)\langle a_\alpha(\Delta t) - a_\alpha(0)\rangle \equiv A_\alpha[a_1(0)\cdots a_N(0)];$$
$$\lim_{\Delta t\to 0} (1/\Delta t)\langle [a_\alpha(\Delta t) - a(0)][a_\beta(\Delta t) - a_\beta(0)]\rangle \equiv B_{\alpha\beta}[a_1(0)\cdots a_N(0)]; \quad (94)$$
$$\lim_{\Delta t\to 0} \frac{1}{\Delta t} \prod_{j=1}^n [a_{\alpha_j}(\Delta t) - a_{\alpha_j}(0)] = 0 \quad \text{for } n \geq 3.$$

These conditions do indeed hold for the real N-component case as may be rigorously verified by application of the techniques developed in the Appendix for the solution to Eq. (22). Moreover, A_α and $B_{\alpha\beta}$ are explicitly found to be

$$A_\alpha(a_1\cdots a_N) = \sum_{\alpha'=1}^N \Big(A_{\alpha\alpha'} + \sum_{\theta=1}^N Q_{\alpha\theta\theta\alpha'}\Big)a_{\alpha'}, \quad (95)$$

$$B_{\alpha\beta}(a_1\cdots a_N) = \sum_{\alpha'=1}^N \sum_{\beta'=1}^N 2Q_{\alpha\alpha'\beta\beta'}a_{\alpha'}a_{\beta'}. \quad (96)$$

Using (95) and (96) when (94) is true for a Markov process leads to the Fokker–Planck equation[14]:

$$\frac{\partial}{\partial t} P(a_1(0)\cdots a_N(0)|a_1\cdots a_N t) = -\sum_{\alpha=1}^N \frac{\partial}{\partial a_\alpha}\Big[\sum_{\alpha'=1}^N \Big(A_{\alpha\alpha'} + \sum_{\theta=1}^N Q_{\alpha\theta\theta\alpha'}\Big) \times a_{\alpha'}P(a_1(0)\cdots a_N(0)|a_1\cdots a_N t)\Big] + \sum_{\alpha=1}^N\sum_{\beta=1}^N \frac{\partial^2}{\partial a_\alpha \partial a_\beta}\Big(\sum_{\alpha'=1}^N\sum_{\beta'=1}^N \times Q_{\alpha\alpha'\beta\beta'}a_{\alpha'}a_{\beta'}P(a_1(0)\cdots a_N(0)|a_1\cdots a_N t)\Big), \quad (97)$$

where $P(a_1(0)\cdots a_N(0)|a_1\cdots a_N t)$ is the probability that $a_1(t) = a_1$, $a_2(t) = a_2, \ldots,$ and $a_N(t) = a_N$ at time $t > 0$ if it was the case that $a_1(t) = a_1(0)$, $a_2(t) = a_2(0), \ldots,$ and $a_N(t) = a_N(0)$ at $t = 0$.

Define $R_{\alpha\alpha'}$ by

$$R_{\alpha\alpha'} \equiv \sum_\theta Q_{\alpha\theta\theta\alpha'}. \quad (98)$$

Equation (40) shows that $R_{\alpha\alpha'}$ has nonpositive eigenvalues.

By using (98), the summation over repeated indices convention, and leaving out the explicit a_α dependence of P leads to

$$\frac{\partial}{\partial t} P = -\frac{\partial}{\partial a_\alpha}[(A_{\alpha\alpha'} + R_{\alpha\alpha'})a_{\alpha'}P] + \frac{\partial^2}{\partial a_\alpha \partial a_\beta}[Q_{\alpha\alpha'\beta\beta'}a_{\alpha'}a_\beta P]. \quad (99)$$

Equation (37) implies that $\sum_\alpha \langle a_\alpha^2(t)\rangle$ is a time invariant. This property may also be seen directly from (99). Averages are given in terms of P by

$$\langle a_\alpha(t)\rangle \equiv \int a_\alpha P(a_1(0)\cdots a_N(0)|a_1\cdots a_N t)\,da_1\cdots da_N,$$
$$\langle a_\alpha(t)a_\beta(t)\rangle \equiv \int a_\alpha a_\beta P(a_1(0)\cdots a_N(0)|a_1\cdots a_N t)\,da_1\cdots da_N. \quad (100)$$

Therefore, using (99) leads to

$$\frac{d}{dt}\sum_\alpha \langle a_\alpha^2(t)\rangle = \int \sum_\alpha a_\alpha^2 \frac{\partial}{\partial t} P\,da_1\cdots da_N = -\int \sum_{\alpha''} a_{\alpha''}^2 \frac{\partial}{\partial a_\alpha}[(A_{\alpha\alpha'} + R_{\alpha\alpha'})a_{\alpha'}P]\,da_1\cdots da_N + \int \sum_{\alpha''} a_{\alpha''}^2 \frac{\partial^2}{\partial a_\alpha \partial a_\beta}[Q_{\alpha\alpha'\beta\beta'}a_{\alpha'}a_\beta P]\,da_1\cdots da_N$$

$$= 2 \int a_\alpha (A_{\alpha\alpha'} + R_{\alpha\alpha'}) a_{\alpha'} P\, da_1 \cdots da_N$$
$$+ 2 \int \delta_{\alpha\beta} Q_{\alpha\alpha'\beta\beta'} a_{\alpha'} a_{\beta'} P\, da_1 \cdots da_N$$
$$= 2R_{\alpha\alpha'} \langle a_\alpha(t) a_{\alpha'}(t)\rangle + 2Q_{\alpha\alpha'\alpha\beta'} \langle a_{\alpha'}(t) a_{\beta'}(t)\rangle$$
$$= 2R_{\alpha\alpha'} \langle a_\alpha(t) a_{\alpha'}(t)\rangle - 2R_{\alpha'\beta'} \langle a_{\alpha'}(t) a_{\beta'}(t)\rangle = 0. \tag{101}$$

The first equality follows from (100), the second equality follows from (99), the third equality follows from integration by parts, the fourth equality follows from (100), and the last two equalities follow from (98) and a renaming of indices. Therefore, it is seen that the time invariance of $\sum_\alpha \langle a_\alpha^2(t)\rangle$ is guaranteed by (97) or (99). Similarly, in the bilinear complex case, the time invariance of $\sum_\alpha \langle \rho_{\alpha\alpha}(t)\rangle$ will be guaranteed by the corresponding Fokker–Planck equation.

A discussion of the solutions to (99) for general N will be reserved for a sequel to this paper. Here it will suffice to present the complete solution to (99) for the Kubo oscillator which is a one complex component case, and as was proved in Sec. 6 corresponds with a two real components case.

The Kubo oscillator is described by Eq. (12). Write $a(t)$ as $a(t) = a_x(t) + ia_y(t)$ where both $a_x(t)$ and $a_y(t)$ are real. In this way (12) becomes a special $N = 2$ case of (22), where

$$\mathbf{A} = \begin{pmatrix} 0 & -\omega \\ \omega & 0 \end{pmatrix} \quad \text{and} \quad \tilde{\mathbf{A}}(t) = \begin{pmatrix} 0 & -\tilde{\varphi}(t) \\ \tilde{\varphi}(t) & 0 \end{pmatrix}, \tag{102}$$

$$Q_{\alpha\alpha'\beta\beta'} = \lambda(\delta_{\alpha 1}\delta_{\alpha' 2}\delta_{\beta 1}\delta_{\beta' 2} + \delta_{\alpha 2}\delta_{\alpha' 1}\delta_{\beta 2}\delta_{\beta' 1} - \delta_{\alpha 1}\delta_{\alpha' 2}\delta_{\beta 2}\delta_{\beta' 1} - \delta_{\alpha 2}\delta_{\alpha' 1}\delta_{\beta 1}\delta_{\beta' 2}). \tag{103}$$

From (103) it is easy to compute $R_{\alpha\alpha'}$ as defined by (98) and this gives

$$\mathbf{R} = \begin{pmatrix} -\lambda & 0 \\ 0 & -\lambda \end{pmatrix}. \tag{104}$$

Using (102)–(104) in (99) for $N = 2$ gives

$$\frac{\partial}{\partial t} P = \omega \left(\frac{\partial}{\partial a_x} a_y - \frac{\partial}{\partial a_y} a_x \right) P + \lambda \left(\frac{\partial}{\partial a_x} a_x + \frac{\partial}{\partial a_y} a_y \right) P + \lambda \left(\frac{\partial^2}{\partial a_x^2} a_y^2 + \frac{\partial^2}{\partial a_y^2} a_x^2 - 2 \frac{\partial^2}{\partial a_x \partial a_y} a_x a_y \right) P. \tag{105}$$

At this point introduce polar coordinates: $a_x = r \cos\theta$ and $a_y = r \sin\theta$. This implies

$$\frac{\partial}{\partial a_x} = \cos\theta \frac{\partial}{\partial r} - \frac{\sin\theta}{r} \frac{\partial}{\partial \theta} \quad \text{and} \quad \frac{\partial}{\partial a_y} = \sin\theta \frac{\partial}{\partial r} + \frac{\cos\theta}{r} \frac{\partial}{\partial \theta}. \tag{106}$$

Using (106) in (105) leads, after a modicum of algebra, to

$$\frac{\partial}{\partial t} P = -\omega \frac{\partial}{\partial \theta} P + \lambda \frac{\partial^2}{\partial \theta^2} P, \tag{107}$$

where $P \equiv P(r(0)\theta(0)|r\theta t)$ and $P(r(0)\theta(0)|r\theta 0) = \delta(r - r(0))\delta(\theta - \theta(0))$. From (107) it is seen that P may be factored,

$$P(r\theta t) = R(rt)\, W(\theta t), \tag{108}$$

and (107) becomes two equations:

$$\frac{\partial}{\partial t} R(rt) = 0 \quad \text{and}$$
$$\frac{\partial}{\partial t} W(\theta, t) = -\omega \frac{\partial}{\partial \theta} W(\theta, t) + \lambda \frac{\partial^2}{\partial \theta^2} W(\theta, t). \tag{109}$$

With the initial condition for P given beneath (107) the solution to (109) for R is $R(rt) = \delta[r - r(0)]$. The solution for $W(\theta, t)$ with periodic boundary conditions is given by

$$W(\theta, t) = \frac{1}{\sqrt{4\pi\lambda t}} \sum_{K=-\infty}^{\infty} \exp\left(- \frac{(\theta - \theta(0) + 2K\pi - \omega t)^2}{4\lambda t} \right). \tag{110}$$

This describes a diffusion process on a circle coupled with a streaming term given by ωt. The complete solution to (105) is then given by

$$P(r(0)\theta(0)|r\theta t) = \delta[r - r(0)] \frac{1}{\sqrt{4\pi\lambda t}} \times \sum_{K=-\infty}^{\infty} \exp\left(- \frac{[\theta - \theta(0) + 2K\pi - \omega t]^2}{4\lambda t} \right). \tag{111}$$

It is possible to use (111) to reconfirm (15).

10. CONCLUDING REMARKS

The physical implications of the equations presented in this paper are relevant in the areas of nonequilibrium thermodynamics and nonequilibrium statistical mechanics. A fuller treatment of the appropriate physical interpretations for these equations will be presented in a sequel to this work. For the present it will suffice to indicate several immediately obvious points.

Additive stochastic processes have been used to explain Brownian motion by Langevin's equation, to explain nonequilibrium thermodynamics close to full equilibrium by the Onsager and Machlup equations, and to explain these first two cases, as well as the fluctuating hydrodynamic theory of Landau of Lifshitz, and the fluctuating Boltzmann equation, by the general theory of stationary, Gaussian, Markov processes presented by Fox and Uhlenbeck. All of these cases are limited to dynamical behavior near full equilibrium, and all of these cases are classical.

Multiplicative stochastic processes, as presented in this paper, suggest physical applications in the following cases. The most simple case is the case of frequency fluctuation for the harmonic oscillator, as was originally proposed by Kubo. The generalization to the real N-component case as given by (22) and (27) corresponds with the Liouville equation with a Hamiltonian that contains a fluctuating contribution to the overall energy.[9] Equation (22) is the matrix representation of the partial differential equation which provides the classical Liouville description. The complex N-component case corresponds with the Heisenberg matrix representation of the Schrödinger equation. Equation (42) is the relevant equation and contains a Hamiltonian which has a fluctuating contribution. Averaging (42) leads to (48) which depicts the decay of total probability as may be seen using (63). In order to avoid this physically unreasonable consequence, the density matrix formulation is presented by Eq. (65), and (74) corresponds with the averaged density matrix equation. Equation (79), in contrast with (48) and (63), implies conservation of total probability, even though (75) guarantees that (74) describes irreversible behavior for the whole averaged density matrix. In the literature (74) is referred to as the Redfield equation.[6] Here, the potential physical applicability of (74) is greater than the nuclear magnetic resonance context usually associated with Redfield's equation. In the special case in which (85) is realized, the Redfield equation (74) is seen to reduce to (89) and (92) which comprise the Pauli master equation for the diagonal elements of the average density matrix.[15]

All these cases show that multiplicative stochastic processes pertain to both classical and quantum mechanical considerations. The restriction of additive stochastic processes to physical applicability corresponding with dynamical behavior close to full equilibrium does not apply to multiplicative stochastic processes. This follows from the difference in the levels of description each case involves. In the additive stochastic process case the description is relatively macroscopic such as in fluctuating hydrodynamics, in the fluctuating Boltzmann equation, and in nonequilibrium thermodynamics. These levels of descriptions are usually nonlinear; but their linear approximations are required in order to obtain their stochastic description. The linearization step requires the restriction of applicability to near full equilibrium. In contrast, in the multiplicative stochastic process case the description is relatively microscopic such as in the fluctuating Liouville equation and in the fluctuating density matrix equation. The levels of description are intrinsically already linear, so that no linearization step is required, and, consequently, there is no corresponding attendant limitation to physical applicability.

The possible limitations to physical applicability of multiplicative stochastic processes arise with respect to the validity of introducing a part of the total Hamiltonian which is a purely random, stationary, Gaussian process. This consideration will be made in detail in a sequel to this work which stresses the physical context. For the present, simply note the existence of the rigorous theorem, the proof of which is found in the Appendix and the consequences of which are found in the text, for multiplicative stochastic processes "driven" by purely random, stationary, Gaussian "forces."

ACKNOWLEDGMENTS

The author is indebted to Professor Mark Kac and Professor George Uhlenbeck for their critical review of this work just prior to its final written rendering. The author is also indebted to the Miller Institute for Basic Research in Science for supporting this work during its inception while the author was a post doctoral fellow of the Miller Institute and associated with the Physics Department of the University of California at Berkeley.

APPENDIX: PROOF OF EQ. (27)

Define $R_\alpha(t)$ by

$$R_\alpha(t) \equiv \sum_{\alpha'} [e^{-\mathbf{A}t}]_{\alpha\alpha'} a_{\alpha'}(t), \tag{A1}$$

where $\mathbf{A}$ denotes the matrix with components $A_{\alpha\alpha'}$, and

$$[e^{-\mathbf{A}t}]_{\alpha\alpha'} \equiv \sum_{n=0}^{\infty} \frac{(-1)^n}{n!} (\mathbf{A}^n)_{\alpha\alpha'} t^n.$$

Also define $\tilde{L}_{\alpha\alpha'}(t)$ by

$$\tilde{L}_{\alpha\alpha'}(t) \equiv \sum_{\beta}\sum_{\beta'} [e^{-\mathbf{A}t}]_{\alpha\beta} \tilde{A}_{\beta\beta'}(t) [e^{\mathbf{A}t}]_{\beta'\alpha'}. \tag{A2}$$

Via (A1) and (A2), (22) may be written as

$$\frac{d}{dt} R_\alpha(t) = \sum_{\alpha'} \tilde{L}_{\alpha\alpha'}(t) R_{\alpha'}(t). \tag{A3}$$

Because $\tilde{\mathbf{L}}(t)$ and $\tilde{\mathbf{L}}(s)$ do not necessarily commute for $t \neq s$, (A3) must be solved using time-ordered integrals:

$$\begin{aligned} R_\alpha(t) = \sum_{\alpha'} \sum_{k=0}^{\infty} \int_0^t \int_0^{s_k} \int_0^{s_{k-1}} \cdots \int_0^{s_3} \int_0^{s_2} \\ \times \sum_{\mu_{k-1}} \sum_{\mu_{k-2}} \cdots \sum_{\mu_2} \sum_{\mu_1} \\ \times \tilde{L}_{\alpha\mu_{k-1}}(s_k) \tilde{L}_{\mu_{k-1}\mu_{k-2}}(s_{k-1}) \cdots \tilde{L}_{\mu_2\mu_1}(s_2) \\ \times \tilde{L}_{\mu_1\alpha'}(s_1) ds_1 \cdots ds_k R_{\alpha'}(0), \end{aligned} \tag{A4}$$

where $t \geq s_k \geq s_{k-1} \geq \cdots \geq s_2 \geq s_1 \geq 0$. Define $W^k_{\alpha\alpha'}(t)$ by

$$\begin{aligned} W^k_{\alpha\alpha'}(t) \equiv \int_0^t \int_0^{s_k} \cdots \int_0^{s_2} \sum_{\mu_{k-1}} \cdots \\ \times \sum_{\mu_1} \langle \tilde{L}_{\alpha\mu_{k-1}}(s_k) \cdots \tilde{L}_{\mu_1\alpha'}(s_1)\rangle ds_1 \cdots ds_k. \end{aligned} \tag{A5}$$

Equations (25) and (A2) imply that

$$W^k_{\alpha\alpha'}(t) = 0 \quad \text{for all odd } k. \tag{A6}$$

Consider all even k, such that $k = 2m$ for $m = 1, 2, \cdots$. Notice that Eqs. (26) and (A2) imply that

$$\begin{aligned} \langle \tilde{L}_{\mu_{2m}\nu_{2m}}(s_{2m}) \cdots \tilde{L}_{\mu_1\nu_1}(s_1)\rangle \\ = \frac{1}{2^m m!} \sum_{p\in S_{2m}} \prod_{j=1}^{m} \langle \tilde{L}_{\mu_{p(2j)}\nu_{p(2j)}}(s_{p(2j)}) \\ \times \tilde{L}_{\mu_{p(2j-1)}\nu_{p(2j-1)}}(s_{p(2j-1)})\rangle, \end{aligned} \tag{A7}$$

where

$$\begin{aligned} \langle \tilde{L}_{\mu\nu}(s) \tilde{L}_{\mu'\nu'}(s')\rangle \\ = \sum_{\alpha}\sum_{\alpha'}\sum_{\beta}\sum_{\beta'} \langle [e^{-\mathbf{A}s}]_{\mu\alpha} \tilde{A}_{\alpha\alpha'}(s) [e^{\mathbf{A}s}]_{\alpha'\nu} \\ \times [e^{-\mathbf{A}s'}]_{\mu'\beta} \tilde{A}_{\beta\beta'}(s') [e^{\mathbf{A}s'}]_{\beta'\nu'}\rangle \\ = 2 \sum_{\alpha}\sum_{\alpha'}\sum_{\beta}\sum_{\beta'} [e^{-\mathbf{A}s}]_{\mu\alpha} [e^{\mathbf{A}s}]_{\alpha'\nu} Q_{\alpha\alpha'\beta\beta'} \\ \times [e^{-\mathbf{A}s}]_{\mu'\beta} [e^{\mathbf{A}s}]_{\beta'\nu'} \delta(s - s'). \end{aligned} \tag{A8}$$

In particular, (A8) leads to

$$\begin{aligned} \sum_{\theta} \langle \tilde{L}_{\mu\theta}(s) \tilde{L}_{\theta\nu}(s')\rangle \\ = 2 \sum_{\alpha}\sum_{\beta'} [e^{-\mathbf{A}s}]_{\mu\alpha} \sum_{\theta} Q_{\alpha\theta\theta\beta'} [e^{\mathbf{A}s}]_{\beta'\nu} \delta(s - s'). \end{aligned} \tag{A9}$$

Using (A7) in (A5) for $k = 2m$ gives

$$W^k_{\alpha\alpha'}(t) = \int_0^t \int_0^{s_k} \cdots \int_0^{s_2} \sum_{\mu_k} \cdots \sum_{\mu_1} \sum_{\nu_k} \cdots \sum_{\nu_1} \times \delta_{\alpha\mu_k} \delta_{\alpha'\nu_1} \prod_{l=1}^{k-1} \delta_{\mu_l \nu_{l+1}} \frac{1}{2^m m!} \sum_{p \in S_{2m}} \prod_{j=1}^{m} \times \langle \tilde{L}_{\mu_{p(2j)}\nu_{p(2j)}}(s_{p(2j)}) \tilde{L}_{\mu_{p(2j-1)}\nu_{p(2j-1)}} \times (s_{p(2j-1)}) \rangle ds_1 \cdots ds_k. \quad \text{(A10)}$$

Using (A8) in (A10) gives

$$W^k_{\alpha\alpha'}(t) = \int_0^t \int_0^{s_k} \cdots \int_0^{s_2} \sum_{\mu_k} \cdots \sum_{\mu_1} \sum_{\nu_k} \cdots \sum_{\nu_1} \times \delta_{\alpha\mu_k} \delta_{\alpha'\nu_1} \prod_{l=1}^{k-1} \delta_{\nu_{l+1}\mu_l} \frac{1}{2^m m!} \sum_{p \in S_{2m}} 2^m \times \prod_{j=1}^{m} \sum_{\alpha_j} \sum_{\alpha'_j} \sum_{\beta_j} \sum_{\beta'_j} [e^{-\mathbf{A}s_{p(2j)}}]_{\mu_{p(2j)}\alpha_j} \times [e^{\mathbf{A}s_{p(2j)}}]_{\alpha'_j \nu_{p(2j)}} Q_{\alpha_j \alpha'_j \beta_j \beta'_j} [e^{-\mathbf{A}s_{p(2j)}}]_{\mu_{p(2j-1)}\beta_j} \times [e^{\mathbf{A}s_{p(2j)}}]_{\beta'_j \nu_{p(2j-1)}} \delta(s_{p(2j)} - s_{p(2j-1)}) ds_1 \cdots ds_k. \quad \text{(A11)}$$

This complex expression for $W^k_{\alpha\alpha'}(t)$ reveals its inner structure and leads to major simplifications. Define $f^{kp}_{\alpha\alpha'}(s_k \cdots s_1)$ by

$$f^{kp}_{\alpha\alpha'}(s_k \cdots s_1) \equiv \sum_{\mu_k} \cdots \sum_{\mu_1} \sum_{\nu_k} \cdots \times \sum_{\nu_1} \delta_{\alpha\mu_k} \delta_{\alpha'\nu_1} \prod_{l=1}^{k-1} \delta_{\nu_{l+1}\mu_l} \prod_{j=1}^{m} \sum_{\alpha_j} \sum_{\alpha'_j} \sum_{\beta_j} \times \sum_{\beta'_j} [e^{-\mathbf{A}s_{p(2j)}}]_{\mu_{p(2j)}\alpha_j} [e^{\mathbf{A}s_{p(2j)}}]_{\alpha'\nu_{p(2j)}} \times Q_{\alpha_j \alpha'_j \beta_j \beta'_j} [e^{-\mathbf{A}s_{p(2j)}}]_{\mu_{p(2j-1)}\beta_j} [e^{\mathbf{A}s_{p(2j)}}]_{\beta'_j \nu_{p(2j-1)}} \quad \text{(A12)}$$

for each $p \in S_{2m}$. Therefore, putting (A12) into (A11) gives

$$W^k_{\alpha\alpha'}(t) = \frac{1}{2^m m!} \sum_{p \in S_{2m}} 2^m \int_0^t \int_0^{s_k} \cdots \int_0^{s_2} f^{kp}_{\alpha\alpha'}(s_k \cdots s_1) \prod_{j=1}^{m} \delta(s_{p(2j)} - s_{p(2j-1)}) \times ds_1 \cdots ds_k. \quad \text{(A13)}$$

Now, define $I^{kq}_{\alpha\alpha'}(t)$ by

$$I^{kq}_{\alpha\alpha'}(t) \equiv \int_0^t \int_0^{s_k} \cdots \int_0^{s_2} f^{kq}_{\alpha\alpha'}(s_k \cdots s_1) \times \prod_{j=1}^{m} \delta(s_{q(2j-1)} - s_{q(2j-1)}) ds_1 \cdots ds_k \quad \text{(A14)}$$

for each $q \in S_{2m}$. Let the set N be given by

$$N \equiv \{q \in S_{2m} \mid \prod_{j=1}^{m} \delta(s_{q(2j)} - s_{q(2j-1)}) \text{ contains the factor } \delta(s_k - s_{k-1})\},$$

and let the set Z be defined by

$$Z \equiv \{q \in S_{2m} \mid \prod_{j=1}^{m} \delta(s_{q(2j)} - s_{q(2j-1)}) \text{ does not contain the factor } \delta(s_k - s_{k-1})\}.$$

Clearly, Z contains q's such that $\prod_{j=1}^{m} \delta(s_{q(2j)} - s_{q(2j-1)})$ contains the factor $\delta(s_k - s_l)$ for $l \neq k-1$. Since the order of the two time arguments in a delta function does not alter its value, and since the order of the delta functions in a product does not alter its value, then there are $2^m m!$ permutations in S_{2m} which yield identical $\prod_{j=1}^{m} \delta(s_{q(2j)} - s_{q(2j-1)})$. The number of permutations in N is $2m[(2m-2)!]$; because there are two ways of ordering s_k and s_{k-1} in $\delta(s_k - s_{k-1})$, there are m ways of ordering the product with respect to the factor $\delta(s_k - s_{k-1})$ and $m-1$ other factors, and there are $(2m-2)!$ ways of permuting the remaining $2m-2$ time variables. The number of permutations in Z is $2m(2m-2)[(2m-2)!]$; because there are two ways of ordering s_k and s_l in $\delta(s_k - s_l)$, there are m ways of ordering the product with respect to the factor $\delta(s_k - s_l)$ and $m-1$ other factors, there are $(2m-2)$ choices for $l \neq k-1$, and there are $(2m-2)!$ ways of permuting the remaining $2m-2$ time variables. In summary, it follows that $S_{2m} = N \cup Z$, $N \cap Z = \emptyset$, and $(2m)! = 2m[(2m-2)!] + 2m(2m-2)[(2m-2)!]$. If $g(s,s')$ is an arbitrary function of two time variables, then the preceding counting scheme leads to

$$\left\{\prod_{j=1}^{m} g(s_{q(2j)}, s_{q(2j-1)}) \mid q \in N\right\} = \left\{g(s_k, s_{k-1}) \prod_{j=1}^{m-1} g(s_{r(2j)} - s_{r(2j-1)}) \mid r \in S_{2m-2}\right\}. \quad \text{(A15)}$$

Each term on the left-hand side of (A15) is redundant $2m$ times if $g(s,s')$ is symmetric in s and s'. A special instance of (A15) is: $\{\prod_{j=1}^{m} \delta(s_{q(2j)} - s_{q(2j-1)}) \mid q \in N\} = \{\delta(s_k - s_{k-1}) \prod_{j=1}^{m-1} \delta(s_{r(2j)} - s_{r(2j-1)}) \mid r \in S_{2m-2}\}$. Equation (A15) will be useful later, and the redundancy factor $2m$ should be noted.

Using (A14) in (A13) gives

$$W^k_{\alpha\alpha'}(t) = \frac{1}{2^m m!} \sum_{p \in S_{2m}} 2^m I^{kp}_{\alpha\alpha'}(t) = \frac{1}{m!} \sum_{q \in N} I^{kq}_{\alpha\alpha'}(t) + \frac{1}{m!} \sum_{q \in Z} I^{kq}_{\alpha\alpha'}(t). \quad \text{(A16)}$$

It will now be shown that

$$I^{kq}_{\alpha\alpha'}(t) = 0 \quad \text{for each } q \in Z. \quad \text{(A17)}$$

Because $q \in Z$,

$$I^{kq}_{\alpha\alpha'}(t) = \int_0^t \int_0^{s_k} \cdots \int_0^{s_2} f^{kq}_{\alpha\alpha'}(s_k \cdots s_1) \times \delta(s_k - s_l)\delta(s_{k-1} - s_i) \times \prod_{j=1}^{m}{}' \delta(s_{q(2j)} - s_{q(2j-1)}) ds_1 \cdots ds_k, \quad \text{(A18)}$$

where $\prod_{j=1}^{m}{}' \delta(s_{q(2j)} - s_{q(2j-1)})$ is defined by

$$\prod_{j=1}^{m} \delta(s_{q(2j)} - s_{q(2j-1)}) \equiv \delta(s_k - s_l)\delta(s_{k-1} - s_i) \prod_{j=1}^{m}{}' \delta(s_{q(2j)} - s_{q(2j-1)}). \quad \text{(A19)}$$

Recall that $q \in Z$ implies that $l \neq k-1$. Therefore, there is some i such that $i \neq k$ and $i \neq l$ and $\delta(s_{k-1} - s_i)$ appears as a factor in the product

$\Pi_{j=1}^{m}\delta(s_{q(2j)}-s_{q(2j-1)})$. Note that $I_{\alpha\alpha'}^{kq}(0)=0$ and

$$\frac{d}{dt}I_{\alpha\alpha'}^{kq}(t)=\int_0^t\int_0^{s_{k-1}}\cdots\int_0^{s_2}f_{\alpha\alpha'}^{kq}(t,s_{k-1},\ldots,s_1)\times\delta(t-s_l)\delta(s_{k-1}-s_i)\prod_{j=1}^{m}{}'\delta(s_{q(2j)}-s_{q(2j-1)})\times ds_1\cdots ds_{k-1}. \tag{A20}$$

The time ordering of the integrals requires that

$$t\geq s_{k-1}\geq\cdots\geq s_l\geq\cdots\geq s_2\geq s_1\geq 0. \tag{A21}$$

The only singular contributions to the integrand of (A20) are in the product of delta functions since $f_{\alpha\alpha'}^{kq}(t,s_{k-1},\ldots,s_1)$ is a bounded integrable function as is seen from (A12). The integrations in (A20) are performed in the order $ds_1, ds_2,\ldots,ds_{k-1}$. After the ds_i integration, the $\delta(s_{k-1}-s_i)$ term will no longer be present, and the functional dependence of the remaining integrand will no longer be singular in s_{k-1} because no other delta function besides $\delta(s_{k-1}-s_i)$ contains s_{k-1} or s_i. For all $s_{k-1}<t$, the $\delta(t-s_l)$ term and (A21) imply that the integrand is zero. Therefore, only $s_{k-1}=t$ can contribute to the over-all integration with respect to s_{k-1}. Thus, when the ds_{k-1} integration is finally performed, the remaining integrand is zero for $s_{k-1}<t$ and is not singular in s_{k-1} anywhere in the interval $[0,t]$. Therefore, the Riemann integral over ds_{k-1} from 0 to t gives zero. This proves that for $q\in Z$, $(d/dt)I_{\alpha\alpha'}^{kq}(t)=0$. Coupling this result with $I_{\alpha\alpha'}^{kq}(0)=0$ implies that for each $q\in Z$ $I_{\alpha\alpha'}^{kq}(t)=0$ for all t. Consequently, (A17) is proved.

Returning to (A16), (A17) implies that

$$W_{\alpha\alpha'}^{k}(t)=\frac{1}{m!}\sum_{q\in N}I_{\alpha\alpha'}^{kq}(t). \tag{A22}$$

Using (A12) and (A14) yields

$$\begin{aligned}\sum_{q\in N}I_{\alpha\alpha'}^{kq}(t)&=\sum_{q\in N}\int_0^t\int_0^{s_k}\cdots\int_0^{s_2}f_{\alpha\alpha'}^{kq}(s_k\cdots s_1)\times\prod_{j=1}^{m}\delta(s_{q(2j)}-s_{q(2j-1)})ds_1\cdots ds_k\\&=\sum_{q\in N}\int_0^t\int_0^{s_k}\cdots\int_0^{s_2}\sum_{\mu_k}\sum_{\mu_{k-1}}\sum_{\nu_k}\sum_{\nu_{k-1}}\\&\quad\times\sum_{\mu_{k-2}}\cdots\sum_{\mu_1}\sum_{\nu_{k-2}}\cdots\sum_{\nu_1}\delta_{\alpha\mu_k}\delta_{\nu_k\mu_{k-1}}\delta_{\alpha'\nu_1}\\&\quad\times\prod_{l=1}^{k-2}\delta_{\nu_{l+1}\mu_l}\sum_{\alpha_m}\sum_{\alpha'_m}\sum_{\beta_m}\sum_{\beta'_m}[e^{-\mathbf{A}s_k}]_{\mu_k\alpha_m}\\&\quad\times[e^{\mathbf{A}s_k}]_{\alpha'_m\nu_k}Q_{\alpha_m\alpha'_m\beta_m\beta'_m}[e^{-\mathbf{A}s_k}]_{\mu_{k-1}\beta_m}\\&\quad\times[e^{\mathbf{A}s_k}]_{\beta'_m\nu_{k-1}}\prod_{j=1}^{m-1}\sum_{\alpha_j}\sum_{\alpha'_j}\sum_{\beta_j}\sum_{\beta'_j}\\&\quad\times[e^{-\mathbf{A}s_{q(2j)}}]_{\mu_{q(2j)}\alpha_j}[e^{\mathbf{A}s_{q(2j)}}]_{\alpha'_j\nu_{q(2j)}}Q_{\alpha_j\alpha'_j\beta_j\beta'_j}\\&\quad\times[e^{-\mathbf{A}s_{q(2j)}}]_{\mu_{q(2j-1)}\beta_j}[e^{\mathbf{A}s_{q(2j)}}]_{\beta'_j\nu_{q(2j-1)}}\\&\quad\times\delta(s_k-s_{k-1})\prod_{i=1}^{m}{}''\delta(s_{q(2i)}-s_{q(2i-1)})\\&\quad\times ds_1\cdots ds_k,\end{aligned} \tag{A23}$$

wherein $\Pi_{i=1}^{\prime\prime m}\delta(s_{q(2i)}-s_{q(2i-1)})$ is defined by

$$\prod_{i=1}^{m}\delta(s_{q(2i)}-s_{q(2i-1)})\equiv\delta(s_k-s_{k-1})\prod_{i=1}^{m}{}''\delta(s_{q(2i)}-s_{q(2i-1)}).$$

If

$$\prod_{l=1}^{k-2}\delta_{\nu_{l+1}\mu_l}=\delta_{\nu_{k-1}\mu_{k-2}}\prod_{l=1}^{k-3}\delta_{\nu_{l+1}\mu_l}$$

and (A15) are used, then the expression in (A23) becomes

$$\begin{aligned}\sum_{q\in N}I_{\alpha\alpha'}^{kq}(t)&=2m\sum_{r\in S_{2m-2}}\int_0^t\int_0^{s_k}\int_0^{s_{k-1}}\cdots\int_0^{s_2}\\&\quad\times\sum_{\nu_{k-1}}\sum_{\alpha_m}\sum_{\beta'_m}[e^{-\mathbf{A}s_k}]_{\alpha\alpha_m}\sum_{\theta_m}Q_{\alpha_m\theta_m\theta_m\beta'_m}\\&\quad\times[e^{\mathbf{A}s_k}]_{\beta'_m\nu_{k-1}}\delta(s_k-s_{k-1})\\&\quad\times\sum_{\mu_{k-2}}\cdots\sum_{\mu_1}\sum_{\nu_{k-2}}\cdots\\&\quad\times\sum_{\nu_1}\delta_{\nu_{k-1}\mu_{k-2}}\delta_{\alpha'\nu_1}\prod_{l=1}^{k-3}\delta_{\nu_{l+1}\mu_l}\prod_{j=1}^{m-1}\\&\quad\times\sum_{\alpha_j}\sum_{\alpha'_j}\sum_{\beta_j}\sum_{\beta'_j}[e^{-\mathbf{A}s_{r(2j)}}]_{\mu_{r(2j)}\alpha_j}\\&\quad\times[e^{\mathbf{A}s_{r(2j)}}]_{\alpha'_j\nu_{r(2j)}}Q_{\alpha_j\alpha'_j\beta_j\beta'_j}\\&\quad\times[e^{-\mathbf{A}s_{r(2j)}}]_{\mu_{r(2j-1)}\beta_j}[e^{\mathbf{A}s_{r(2j)}}]_{\beta'_j\nu_{r(2j-1)}}\\&\quad\times\prod_{i=1}^{m-1}\delta(s_{r(2i)}-s_{r(2i-1)})ds_1\cdots\\&\quad\times ds_{k-2}ds_{k-1}ds_k.\end{aligned} \tag{A24}$$

The factor $2m$ comes from the redundancy requirement discussed following relation (A15). Because $r\in S_{2m-2}$, it also follows that

$$\prod_{i=1}^{m}{}''(s_{q(2i)}-s_{q(2i-1)})=\prod_{i=1}^{m-1}\delta(s_{r(2j)}-s_{r(2i-1)})\ \text{for}\ q\in N,$$

and this has been used to get (A24) from (A23). Using (A12) for $k-2$ shows that (A24) is equivalent to

$$\begin{aligned}\sum_{q\in N}I_{\alpha\alpha'}^{kq}(t)&=2m\sum_{r\in S_{2m-2}}\int_0^t\int_0^{s_k}\sum_{\nu_{k-1}}\sum_{\alpha_m}\sum_{\beta'_m}\\&\quad\times[e^{-\mathbf{A}s_k}]_{\alpha\alpha_m}\sum_{\theta_m}Q_{\alpha_m\theta_m\theta_m\beta'_m}[e^{\mathbf{A}s_k}]_{\beta'_m\nu_{k-1}}\\&\quad\times\delta(s_k-s_{k-1})\int_0^{s_{k-1}}\cdots\int_0^{s_2}f_{\nu_{k-1}\alpha'}^{k-2\,r}\\&\quad\times(s_{k-2}\cdots s_1)\prod_{i=1}^{m-1}\delta(s_{r(2i)}-s_{r(2i-1)})\\&\quad\times ds_1\cdots ds_{k-2}ds_{k-1}ds_k\\&=2m\sum_{r\in S_{2m-2}}\int_0^t\int_0^{s_k}\sum_{\nu_{k-1}}\sum_{\alpha_m}\sum_{\beta'_m}[e^{-\mathbf{A}s_k}]_{\alpha\alpha_m}\\&\quad\times\sum_{\theta_m}Q_{\alpha_m\theta_m\theta_m\beta'_m}[e^{\mathbf{A}s_k}]_{\beta'_m\nu_{k-1}}\\&\quad\times\delta(s_k-s_{k-1})I_{\nu_{k-1}\alpha'}^{k-2\,r}(s_{k-1})ds_{k-1}ds_k.\end{aligned} \tag{A25}$$

The second equality in (A25) follows from (A14) for $k-2$.

By using (A13) and (A14) it also follows that

$$W^{k-2}_{\nu_{k-1}\alpha'}(s_{k-1}) = \frac{1}{(m-1)!}\sum_{r\in S_{2m-2}} I^{k-2\,r}_{\nu_{k-1}\alpha'}(s_{k-1}) \tag{A26}$$

because $k-2 = 2(m-1)$. Now, if one last quantity $N_{\alpha\,\nu_{k-1}}(s_k)$ is defined by

$$N_{\alpha\nu_{k-1}}(s_k) \equiv \sum_{\alpha_m}\sum_{\beta'_m}[e^{-\mathbf{A}s_k}]_{\alpha\alpha_m}\sum_{\theta_m} Q_{\alpha_m\theta_m\theta_m\beta'_m}[e^{\mathbf{A}s_k}]_{\beta'_m\nu_{k-1}}, \tag{A27}$$

then together with (A13), (A14), (A25), and (A26) this leads to

$$\begin{aligned} W^k_{\alpha\alpha'}(t) &= 2\sum_{\nu_{k-1}}\int_0^t\int_0^{s_k} N_{\alpha\nu_{k-1}}(s_k)\delta(s_k - s_{k-1}) \\ &\quad\times W^{k-2}_{\nu_{k-1}\alpha'}(s_{k-1})\,ds_{k-1}ds_k \\ &= \sum_{\nu_{k-1}}\int_0^t N_{\alpha\nu_{k-1}}(s_k)\,W^{k-2}_{\nu_{k-1}\alpha'}(s_k)\,ds_k, \end{aligned} \tag{A28}$$

wherein the ds_{k-1} integration with $\delta(s_k - s_{k-1})$ in the integrand and s_k as an integration limit introduced a factor of $\frac{1}{2}$ which cancelled the 2. By differentiation, (A28) gives

$$\frac{d}{dt}\,W^k_{\alpha\alpha}(t) = \sum_{\nu_{k-1}} N_{\alpha\nu_{k-1}}(t)\,W^{k-2}_{\nu_{k-1}\alpha'}(t). \tag{A29}$$

By returning to (A4) and (A5), (A29) permits the writing of

$$\begin{aligned} \frac{d}{dt}\langle R_\alpha(t)\rangle &= \sum_{\alpha'}\sum_{k=0}^{\infty}\frac{d}{dt}\,W^k_{\alpha\alpha'}(t)R_{\alpha'}(0) \\ &= \sum_{\alpha'}\sum_{m=0}^{\infty}\frac{d}{dt}\,W^{2m}_{\alpha\alpha'}(t)R_{\alpha'}(0) \\ &= \sum_{\alpha'}\sum_{m=1}^{\infty}\sum_{\nu} N_{\alpha\nu}(t)\,W^{2m-2}_{\nu\alpha'}(t)R_{\alpha'}(0) \\ &= \sum_{\nu} N_{\alpha\nu}(t)\sum_{\alpha'}\sum_{m=0}^{\infty} W^{2m}_{\alpha\alpha'}(t)R_{\alpha'}(0) \\ &= \sum_{\nu} N_{\alpha\nu}(t)\langle R_\nu(t)\rangle. \end{aligned} \tag{A30}$$

Using (A1) and (A27) finishes the proof of (27) with

$$\frac{d}{dt}\langle a_\alpha(t)\rangle = \sum_{\alpha'} A_{\alpha\alpha'}\langle a_{\alpha'}(t)\rangle + \sum_{\alpha'}\sum_{\theta} Q_{\alpha\theta\theta\alpha'}\langle a_{\alpha'}(t)\rangle.$$

It should be noted that this result is equivalent with the statement that the time-ordered integrals which arise in the formal solution to (22) yield nonzero quantities upon averaging, only if the product of delta functions which occurs is "properly ordered." By "properly ordered" is meant that

$$\prod_{j=1}^{m}\delta(s_{p(2j)} - s_{p(2j-1)}) = \prod_{j=1}^{m}\delta(s_{(2j)} - s_{(2j-1)}), \tag{A31}$$

since $\prod_{j=1}^{m}\delta(s_{(2j)} - s_{(2j-1)})$ in the integrand of the time-ordered integral leads to a nonzero quantity, whereas any other pairing of time variables leads to zero. Therefore, only permutations which satisfy (A31) give "properly ordered" delta function products. In the proof presented here this property has been arrived at by "peeling off" two time variables at a time, and noting that to get a nonzero result that the two time variables "peeled off" were "properly ordered" relative to all possible time variables.

Reference to these properties of "properly ordered" products are made in Sec. 9 with respect to rigorously justifying the Fokker–Planck equations given in that section.

1 P. Langevin, Compt. Rend. **146**, 530 (1908).
2 L. Onsager and S. Machlup, Phys. Rev. **91**, 1512 (1953).
3 R. F. Fox and G. E. Uhlenbeck, Phys. Fluids **13**, 1893 (1970).
4 R. F. Fox and G. E. Uhlenbeck, Phys. Fluids **13**, 2881 (1970).
5 L. D. Landau and E. M. Lifshitz, *Fluid Mechanics* (Pergamon, London, 1959), Chap. 17.
6 A. Redfield, *Advances in Magnetic Resonance* (Academic, New York, 1965), Vol. 1, pp. 1–32.
7 R. Kubo, *Fluctuations, Relaxation and Resonance in Magnetic Systems* (Oliver and Boyd, Edinburgh, 1962), pp. 23–68.
8 R. Kubo, "*Stochastic Processes and Statistical Mechanics of Irreversible Processes*," Unpublished lecture notes (1963).
9 R. Kubo, J. Math. Phys. **4**, 174 (1963).
10 M. C. Wang and G. E. Uhlenbeck, Rev. Mod. Phys. **17**, 323 (1945).
11 See Ref. 10, note III of the Appendix.
12 A. Einstein, *Investigations on the Theory of the Brownian Movement* (Dover, New York, 1956).
13 G. E. Uhlenbeck and L. S. Ornstein, Phys. Rev. **36**, 823 (1930).
14 See Ref. 10, Sec. 8.
15 K. Huang, *Statistical Mechanics* (Wiley, New York, 1963), p. 203.

Long-Wavelength Normal Modes of Crystals with Coulomb Interactions

John A. Davies

Department of Physics, Clark University, Worcester, Massachusetts 01610

(Received 3 March 1972)

An investigation is made of the behavior of the normal modes of vibration in the long-wavelength limit for infinite crystal lattices in which Coulomb interactions are present. The work is applicable to point ion models with any crystal structure. Rules are derived which are helpful in determining the long-wavelength behavior of the normal modes from symmetry considerations. A study is made of the conditions under which the branches of the phonon dispersion relations will approach definite frequencies in the long-wavelength limit. Finally, a number of examples are presented which illustrate the preceding analysis.

I. INTRODUCTION

The presence of Coulomb interactions in an infinite lattice has a marked effect on its lattice dynamics at long wavelengths. Neither the dynamical matrix nor the phonon dispersion relations have well-defined values at infinite wavelength for such lattices. In fact, in some cases, a branch of the dispersion relations will not even approach a definite frequency (independent of the direction of the propagation vector) as the propagation vector approaches zero.[1] The standard methods of group theory used to analyze the behavior of the dispersion relations at long wavelengths must