

Fluctuations and nonlinear irreversible processes. II

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This paper forms the second part of a study which reexamines the relationship between fluctuations and nonlinear irreversible processes. The scope of the previous paper is generalized to include macroscopic variables which transform odd under time reversal. The fluxes of some of the variables may be purely reversible so that the diffusion matrix may be singular. The deterministic equations for nonlinear irreversible processes can again be derived from a minimum principle. The fluctuations of the macroscopic variables are treated on the basis of a Fokker-Planck equation which has the form derived from statistical mechanics by one of us. The conditional probability of the fluctuations is constructed as a path integral. The connection between the deterministic and the stochastic descriptions of the macroscopic dynamics is formulated in a covariant way, independent of the frame of coordinates. For that purpose, a metric tensor in the space of state variables is introduced. The form of the metric tensor is particularly simple in frames where the macroscopic variables are sums of molecular variables.

I. INTRODUCTION

Irreversible processes in macroscopic systems are described on the fundamental molecular level by statistical mechanics, and on a more phenomenological macroscopic level as a dynamical process of a complete set of macroscopic variables. In the macroscopic description the dynamical process is either considered as deterministic or stochastic, depending on the problem and on the degree of approximation adopted. In any case, the deterministic and stochastic descriptions are intimately connected because they emerge from the same underlying statistical mechanics.

It seems to be clear that the deterministic description comes out of the stochastic description in a deterministic limit in which fluctuations become negligible. It is less clear what exactly is involved in that limit and, furthermore, whether the stochastic description can be reconstructed in a unique way if only the deterministic description is known. In a preceeding paper¹ (henceforth referred to as I) two of us have investigated this question for systems where the stochastic macroscopic dynamics is furnished by the Fokker-Planck equation derived in an earlier paper by one of us (M.S.G.).² The analysis in I was restricted because the macroscopic variables treated were limited to those transforming even under time reversal. In the present paper we want to return to the same question, but allowing for all the complications due to the existence of

macroscopic variables with different time-reversal properties.

Before beginning a systematic exposition it may be worthwhile to elaborate on some aspects of the problem we wish to address. One might ask why we are dissatisfied with the widespread procedure³ for constructing macroscopic stochastic models from macroscopic deterministic equations by requiring detailed balance and the canonical form of the equilibrium distribution. The reason for our dissatisfaction with this procedure is the fact that both the deterministic description and the stochastic description are covariant, i.e., independent of the frame of coordinates in state space used, whereas the above procedure of relating these two descriptions is not covariant.⁴ In other words, physically different stochastic models are obtained from the same deterministic equations if the above procedure is applied in different frames of coordinates in state space.

A similar difficulty arises in formulating the deterministic limit of the stochastic description. If simply the diffusion term of the Fokker-Planck equation is neglected, the resulting "drift approximation" lacks covariance and leads to different results in different frames. We try to resolve these difficulties by bearing in mind that the physical quantities must transform correctly under arbitrary nonlinear transformations of the state variables.

In Onsager's work⁵ the connection between the deterministic theory of irreversible processes

and the stochastic theory of spontaneous fluctuations was made by postulating that the decay of a system from a given nonequilibrium state produced by a spontaneous fluctuation obeys, on the average, the deterministic laws. This work was limited to the linear regime. In I, two of us extended this connection between the deterministic and the stochastic level of description to nonlinear systems with even state variables. The deterministic limit was shown to be controlled by a parameter k , which was identified as Boltzmann's constant. The covariant formulation of the deterministic limit $k \rightarrow 0$ involved a metric in state space. In I, we made the simplest choice and used the transport coefficients to define a metric.⁶ Here, we question this choice and propose a different choice of the metric which is more satisfactory from a physical point of view.

The outline of the paper is as follows: In Sec. II we discuss the deterministic equations of motion for the irreversible process. The underlying molecular nature of the system is manifest in the form of the deterministic equations as a linear relationship between fluxes and forces. One of the difficulties due to the existence of macroscopic variables with different time-reversal symmetries is the possibility that some of the fluxes may be purely reversible. By way of example, we mention the mass flux in a fluid and the time rate of change of the position of a Brownian particle. Because of the intimate connection between irreversible fluxes and fluctuations, the lack of an irreversible flux points to a constraint on the fluctuations. This is apparent in the formulation of the deterministic equations according to a variational principle in which the purely reversible equations are treated as constraints.

In Sec. III we turn to stochastic motion. We first summarize the main results of a previous investigation² by one of us. The Fokker-Planck equation derived there is the basis for our further discussion. The stochastic description by the Fokker-Planck equation is related to the deterministic description by the transport equations. A covariant form of the Fokker-Planck equation is obtained. We show that the main quantity which is necessary to specify a precise and covariant relation between the deterministic and the stochastic description is the metric in state space. The appropriate choice of this metric is discussed in Sec. IV.

It is clear that the entropy must be defined differently in the deterministic and the stochastic descriptions, since the stochastic description contains more information about the state of the system than the deterministic one. In Sec. V we re-examine the properties of the entropies and the

form of the second law in both descriptions and discuss their relation. In Sec. VI we use our previous results to represent the conditional probability as a path integral. We obtain the general form of this representation of a stochastic process for a system with a diffusion matrix which may be singular. In Sec. VII we present our conclusions.

II. DETERMINISTIC MOTION

A. Transport equations

We consider a system described by a set $a = (a^1, \dots, a^I, \dots, a^N)$ of macroscopic variables. Choosing the variables so that they are even or odd in time, the time-reversal transformation in state space reads

$$\tilde{a}^I = \epsilon^I a^I, \quad \epsilon^I = \begin{cases} 1, & \text{for even variables} \\ -1, & \text{for odd variables.} \end{cases} \quad (2.1)$$

The entropy $S(a)$ is an even variable

$$S(\tilde{a}) = S(a). \quad (2.2)$$

As in I, we write the deterministic equations in Onsager's form

$$\dot{a}^I = f^I = L^{IJ} \chi_J. \quad (2.3)$$

We understand that a summation is to be carried out for repeated indices in all formulas except those with ϵ^I . The

$$\chi_I = \frac{\partial S}{\partial a^I} \equiv S_{I1} \quad (2.4)$$

are the thermodynamic forces and the L^{IJ} are the transport coefficients which may be functions of the state. The transport coefficients obey the reciprocal relations

$$L^{IJ}(\tilde{a}) = \epsilon^I \epsilon^J L^{JI}(a). \quad (2.5)$$

The matrix L^{IJ} is not symmetric in general. We may split it into a symmetric part

$$D^{IJ} = \frac{1}{2}(L^{IJ} + L^{JI}) \quad (2.6)$$

and an antisymmetric part

$$A^{IJ} = \frac{1}{2}(L^{IJ} - L^{JI}). \quad (2.7)$$

The deterministic drift f^I splits into

$$f^I = r^I + d^I, \quad (2.8)$$

where

$$r^I = A^{IJ} \chi_J \quad (2.9)$$

is the reversible part with the symmetry

$$r^I(\tilde{a}) = -\epsilon^I r^I(a), \quad (2.10)$$

and where

$$\dot{d}^I = D^{IJ} \chi_J \quad (2.11)$$

is the irreversible part with the symmetry

$$\dot{d}^I(\bar{a}) = \epsilon^I d^I(a). \quad (2.12)$$

Only the irreversible part contributes to the time rate of change of the entropy

$$\dot{S} = \chi_J \dot{a}^J = \chi_I d^I = D^{IJ} \chi_I \chi_J. \quad (2.13)$$

B. Reversible fluxes

The irreversible motion of the set a of macroscopic variables is due to the interaction of these variables with the large set of microscopic degrees of freedom. Often, some of the macroscopic variables do not couple directly to the microscopic degrees of freedom, and their fluxes are purely reversible. In such a case we may choose the set of macroscopic variables so that the first n variables $\{a^1, \dots, a^\alpha, \dots, a^n\}$ are those with purely reversible fluxes

$$\dot{a}^\alpha = f^\alpha \equiv r^\alpha. \quad (2.14)$$

We use the following convention: greek indices α, β, \dots run from 1 to n ; labeling the variables with purely reversible fluxes, small roman indices i, j, \dots run from $n+1$ to N , and large roman indices I, J, \dots run through the complete set from 1 to N . The first n equations of the set (2.3) of transport equations take the form (2.14) if the symmetric parts of some of the transport coefficients vanish

$$D^{\alpha\beta} = D^{\alpha i} = D^{i\alpha} = 0. \quad (2.15)$$

The remaining coefficients D^{ij} form a matrix in the subspace of the macroscopic variables with partly irreversible fluxes. The second law requires that D^{ij} be positive definite. The inverse D_{ij} of the matrix D^{ij} is defined by

$$D_{ij} D^{jk} = D^{kj} D_{ji} = \delta_i^k. \quad (2.16)$$

Despite the fact that the reversible deterministic equations are just a special case of the general transport equation (2.3), we must distinguish the variables $\{a^\alpha\}$ from the variables $\{a^i\}$. The macroscopic variables undergo fluctuations which are neglected in the deterministic theory. It is well known that these fluctuations are intimately connected with the irreversible motion of the variables a . Since the fluxes $\{\dot{a}^\alpha\}$ are purely reversible, the variables $\{a^\alpha\}$ will fluctuate only because of their couplings to the variables $\{a^i\}$ while the variables $\{a^i\}$ are driven directly by microscopic processes.

C. Variational principle

The differences between the $\{a^\alpha\}$ and the $\{a^i\}$ variables appear already in the variational principle for the deterministic equations where the purely reversible equations are treated as constraints. Introducing the Lagrangian

$$O(a, \dot{a}) = \frac{1}{2} D_{ij} (\dot{a}^i - f^i) (\dot{a}^j - f^j) \quad (2.17)$$

as well as the action functional

$$A(a(t), t_1 \leq t \leq t_2) = \int_{t_1}^{t_2} dt O(a(t), \dot{a}(t)), \quad (2.18)$$

we may ask for the path that minimizes the action among all paths that start out from $a(t_1) = a$ and that satisfy the constraints (2.14). To deal with the constraints we use the method of Lagrange multipliers and vary the functional

$$\int_{t_1}^{t_2} dt \{ O(a(t), \dot{a}(t)) - \eta_\alpha(t) [\dot{a}^\alpha(t) - f^\alpha(t)] \}.$$

As in I, the determination of the minimum path naturally divides into two steps. First we determine the minimum path connecting fixed initial and final states, which leads to the Euler-Lagrange equations

$$\begin{aligned} \frac{d}{dt} \eta_\alpha &= \frac{\partial O}{\partial a^\alpha} - \eta_\beta \frac{\partial f^\beta}{\partial a^\alpha}, \\ \frac{d}{dt} \frac{\partial O}{\partial \dot{a}^i} &= \frac{\partial O}{\partial a^i} - \eta_\alpha \frac{\partial f^\alpha}{\partial a^i}. \end{aligned} \quad (2.19)$$

Then the final state is varied, which leads to the additional requirements

$$\eta_\alpha = 0, \quad \frac{\partial O}{\partial \dot{a}^i} = 0. \quad (2.20)$$

We introduce the quantities

$$\eta_i = \frac{\partial O}{\partial \dot{a}^i} = D_{ij} (\dot{a}^j - f^j), \quad (2.21)$$

which, together with the Lagrange multipliers $\{\eta_\alpha\}$, form a complete set of conjugate variables η . The Euler-Lagrange equations (2.19) and the constraints (2.14) can then be transformed to the canonical equations

$$\begin{aligned} \dot{a}^I &= f^I + D^{IJ} \eta_J, \\ \dot{\eta}_I &= -\frac{1}{2} D_{JK}^I \eta_J \eta_K - f^J \eta_I. \end{aligned} \quad (2.22)$$

These equations are of the form

$$\dot{a}^I = \frac{\partial H}{\partial \eta_I}, \quad \dot{\eta}_I = -\frac{\partial H}{\partial a^I}, \quad (2.23)$$

where the Hamiltonian is given by

$$H(a, \eta) = \frac{1}{2} D^{IJ} \eta_I \eta_J + f^I \eta_I. \quad (2.24)$$

The additional requirements (2.20) may be written

$$\eta_I(t_2) = 0. \quad (2.25)$$

This condition picks from all paths that satisfy (2.22) the one with $\dot{a}^I = f^I$, $\eta_I = 0$, which is the deterministic path.

III. STOCHASTIC MOTION

A. Statistical-mechanical Fokker-Planck equation

The stochastic theory deals with the fluctuations of the macroscopic variables, and is characterized by the conditional probability $p_t(a'/a)da'$ that the system will reach a state a' in the volume element da' within the time interval t if it starts out from the state a . In an earlier paper by one of us² it has been argued that $p_t(a'/a)$ is the Green's function solution of a Fokker-Planck equation of the form

$$\frac{\partial}{\partial t} p_t = \frac{\partial}{\partial a^I} \left(-v^I - w^{-1} \frac{\partial w \xi^{IJ}}{\partial a^J} + \frac{\partial}{\partial a^J} \xi^{IJ} \right) p_t. \quad (3.1)$$

The quantities w , v^I , and ξ^{IJ} are functions of the macroscopic variables, and they are determined as certain statistical-mechanical averages of molecular quantities; $w(a)$ is obtained by averaging the microscopic equilibrium distribution over a hypersurface $M(a)$ in phase space where the macroscopic variables have the fixed values a . It has the symmetry

$$w(\tilde{a}) = w(a), \quad (3.2)$$

and is the stationary solution of the Fokker-Planck equation (3.1). $v^I(a)$ is the average flux of the variable a^I on the hypersurface $M(a)$. It can be written

$$v^I = w^{-1} \frac{\partial}{\partial a^J} w \alpha^{IJ}, \quad (3.3)$$

where α^{IJ} is essentially given by the Poisson bracket⁷ of the variables a^I and a^J averaged over $M(a)$. The matrix α^{IJ} has the symmetries

$$\alpha^{IJ}(a) = -\alpha^{JI}(a), \quad (3.4)$$

$$\alpha^{IJ}(\tilde{a}) = \epsilon^I \epsilon^J \alpha^{JI}(a). \quad (3.5)$$

Consequently, v^I has the properties

$$\frac{\partial}{\partial a^I} v^I w = 0, \quad (3.6)$$

$$v^I(\tilde{a}) = -\epsilon^I v^I(a). \quad (3.7)$$

Finally, $\xi^{IJ}(a)$ is interpreted in statistical mechanics as a time integral over the average time correlation of the subtracted fluxes of the variables a^I and a^J on the hypersurface $M(a)$ where the subtracted fluxes are the microscopic fluxes subtracted by their averages v^I and v^J , respec-

tively. The matrix ξ^{IJ} is not necessarily symmetric, but it has the symmetry

$$\xi^{IJ}(\tilde{a}) = \epsilon^I \epsilon^J \xi^{JI}(a). \quad (3.8)$$

Since the $\{a^\alpha\}$ variables do not couple directly to the microscopic degrees of freedom, their fluxes are constant on every hypersurface $M(a)$ and their subtracted fluxes vanish. So do the integrands of those ξ^{IJ} coefficients which are correlations with a subtracted flux of an $\{a^\alpha\}$ variable, and we have

$$\xi^{\alpha\beta} = \xi^{\alpha i} = \xi^{i\alpha} = 0. \quad (3.9)$$

B. Connection between stochastic and deterministic motion

We wish to relate the coefficients of the Fokker-Planck equation (3.1) with the coefficients of the deterministic equations (2.3). By fixing this relation we define implicitly the limit in which the deterministic equations are obtained from the Fokker-Planck equation. The transport coefficients L^{IJ} are defined by

$$L^{IJ} = (1/k)(\alpha^{IJ} + \xi^{IJ}), \quad (3.10)$$

and hence

$$kA^{IJ} = \alpha^{IJ} + \frac{1}{2}(\xi^{IJ} - \xi^{JI}), \quad kD^{IJ} = \frac{1}{2}(\xi^{IJ} + \xi^{JI}), \quad (3.11)$$

where k is Boltzmann's constant. Equation (3.10) is a natural generalization of a corresponding definition in I. The coefficients so-defined have precisely the properties of Sec. II.

To define the entropy $S(a)$ we seek for a generalization of Boltzmann's principle that relates the entropy with the logarithm of the stationary distribution $w(a)$. In order that $w(a)da$ and $S(a)$ should both be independent of the representation of the state we put

$$w(a) \propto g(a)^{-1/2} e^{(1/k)S(a)}, \quad (3.12)$$

where g is the determinant of a metric tensor g^{IJ} in state space. The crucial question of an adequate definition of the metric tensor is discussed in Sec. IV. In Sec. V we show that, regardless of our choice of $g(a)$, the definition (3.12) makes $S(a)$ the deterministic limit of a stochastic entropy which is associated with the stochastic level of description. Using (3.3), (3.10), and (3.12), we can rewrite the Fokker-Planck equation (3.1) in the form

$$\frac{\partial}{\partial t} p_t = \frac{\partial}{\partial a^I} \left(-K^I + \frac{\partial}{\partial a^J} kD^{IJ} \right) p_t \quad (3.13)$$

with the Fokker-Planck drift

$$K^I = f^I + kg^{1/2} (L^{IJ} g^{-1/2})_{,J}. \quad (3.14)$$

Hence, for a given metric in state space, Eqs.

(3.10) and (3.12) establish a unique connection between the deterministic equations (2.3) and the Fokker-Planck equation (3.1).

It is easy to show that for a metric with the symmetry

$$g(\tilde{a}) = g(a) \quad (3.15)$$

the Fokker-Planck equation (3.13) satisfies the potential conditions⁸

$$\begin{aligned} w(\tilde{a}) &= w(a), \\ D^{IJ}(\tilde{a}) &= \epsilon^I \epsilon^J D^{IJ}(a), \\ \frac{\partial}{\partial a^I} K_{\text{rev}}^I(a) w(a) &= 0, \\ K_{\text{irr}}^I(a) w(a) &= k \frac{\partial}{\partial a^I} D^{IJ}(a) w(a), \end{aligned} \quad (3.16)$$

where

$$\begin{aligned} K_{\text{rev}}^I(a) &= \frac{1}{2} [K^I(a) - \epsilon^I K^I(\tilde{a})] \\ &= A^{IJ} \chi_J + k g^{1/2} (A^{IJ} g^{-1/2})_{|J}, \end{aligned} \quad (3.17)$$

$$\begin{aligned} K_{\text{irr}}^I(a) &= \frac{1}{2} [K^I(a) + \epsilon^I K^I(\tilde{a})] \\ &= D^{IJ} \chi_J + k g^{1/2} (D^{IJ} g^{-1/2})_{|J} \end{aligned} \quad (3.18)$$

are the reversible and irreversible parts of the Fokker-Planck drift, respectively. The conditions (3.16) guarantee the detailed balance of the Fokker-Planck process.

In the limit $k \rightarrow 0$ the diffusion matrix kD^{IJ} of the Fokker-Planck equation vanishes, and there is no broadening of a distribution concentrated on one state. The center of such a distribution moves in the limit $k \rightarrow 0$ according to the deterministic equations (2.3).

C. Covariant form of Fokker-Planck equation

The Fokker-Planck drift K^I does not transform like a vector. However, following essentially a recent work⁶ of one of us, we can write the Fokker-Planck equation in a manifest covariant form if we introduce a scalar probability

$$\hat{p}_t = g^{1/2} p_t \quad (3.19)$$

which obeys the equation

$$\frac{\partial}{\partial t} \hat{p}_t = (-h^I \hat{p}_t + k D^{IJ} \hat{p}_{t;J})_{;I}. \quad (3.20)$$

The covariant derivatives are those of a Riemannian manifold with metric g^{IJ} . In particular, the covariant gradient of a scalar is

$$\psi_{;I} = \frac{\partial \psi}{\partial a^I} = \psi_{|I}, \quad (3.21)$$

and the covariant divergence of a vector ψ^I reads

$$\psi^I_{;I} = g^{1/2} \frac{\partial}{\partial a^I} (\psi^I g^{-1/2}). \quad (3.22)$$

The covariant drift vector h^I is given by

$$h^I = f^I + k g^{1/2} (A^{IJ} g^{-1/2})_{|J}. \quad (3.23)$$

The second term is k times the covariant divergence of the antisymmetric matrix A^{IJ} . This term is itself a vector.

At the end of this section we list some relations among the various drift vectors that hold in a basis where the $\{a^\alpha\}$ and $\{a^i\}$ variables are separated:

$$K^\alpha = h^\alpha = v^\alpha = A^{\alpha I} \chi_I + k g^{1/2} (A^{\alpha I} g^{-1/2})_{|I}, \quad (3.24)$$

$$f^\alpha = r^\alpha = A^{\alpha I} \chi_I, \quad (3.25)$$

$$K^i = h^i + k g^{1/2} (D^{ij} g^{-1/2})_{|j}, \quad (3.26)$$

$$h^i = L^{iJ} \chi_J + k g^{1/2} (A^{iJ} g^{-1/2})_{|J}, \quad (3.27)$$

$$f^i = L^{iJ} \chi_J. \quad (3.28)$$

The α components of K , h , and f are reversible, while the i components contain an irreversible part. The reversible parts K_{rev}^i , h_{rev}^i , and $f_{\text{rev}}^i = r^i$ are related by

$$K_{\text{rev}}^i = h_{\text{rev}}^i = A^{iJ} \chi_J + k g^{1/2} (A^{iJ} g^{-1/2})_{|J}, \quad (3.29)$$

$$f_{\text{rev}}^i = r^i = A^{iJ} \chi_J. \quad (3.30)$$

IV. METRIC IN STATE SPACE

So far we have not specified the metric g^{IJ} in state space, and its specification may merely seem like a matter of convenience. In fact, in the previous work we made the simplest choice and used the transport coefficients to define the metric. With that definition the theory was particularly simple, since metrical quantities derived from the transport coefficients like the "curvature" R then appear very naturally in the path integral. On the other hand, there is also a physical point involved in choosing the metric. It is clear from Sec. III that the definition of the metric is intimately connected with the question of the meaning of the deterministic theory. The very definition of the deterministic limit depends on the choice of the metric, e.g., the definition of the entropy S in Eq. (3.12). From this point of view, it now appears unnatural to identify the metric with the matrix of transport coefficients. After all, why should the entropy depend on the transport coefficients characterizing dynamical properties of the system? That is why we give here another, more physical, definition of the metric.

In statistical mechanics there are "natural" representations of the state a so that the macroscopic variables a are algebraic sums of molecular variables that depend on few microscopic degrees of freedom. We now define the metric

in such a way that the metric tensor is independent of the state a for those representations. Natural representations are related by linear transformation laws that change the determinant g by a constant factor only. Since such a factor does not matter in Sec. III, there is no need to give a more precise definition of the metric. Physically, this choice of the metric implies that the deterministic state associated with a given probability distribution by the deterministic limit $k \rightarrow 0$ is identical with the most probable state in a natural representation. This conforms to our intuition. However, the concept of the most probable state is not covariant. Hence we cannot identify the deterministic state with the most probable state in a general frame of coordinates.

In an arbitrary representation of the state the metric must be determined by means of the tensor transformation law. The knowledge of the metric is equivalent to knowing how a given set of macroscopic variables is related with a natural choice of the variables. With this definition of the metric the geometry of the state space is Euclidean. Equation (3.15) is automatically satisfied. The metric is the only quantity appearing in the covariant theory of the previous sections that distinguishes the natural representations. That distinction, however, is very desirable from a physical point of view. For instance, a linear system will behave linearly just in those representations, and nonlinearities are adequately read off from the form of the equations of motion in a natural representation.

In a natural representation the formulas which relate the deterministic and the stochastic description simplify considerably, since the g factors can be omitted everywhere. In these representations the usual procedure, mentioned in the Introduction, for constructing stochastic models from deterministic transport equations is justified, since it is consistent with the deterministic limit $k \rightarrow 0$. Further, the deterministic state associated with a distribution is just the most probable state and Boltzmann's formula connects the entropy S with the stationary distribution w in the usual way: $S \propto \log w$.

V. ENTROPY AND THE SECOND LAW

A. Entropy

In the stochastic theory the "state" of the system is characterized by a distribution $p(a)$ in state space, whereas the "state" of the deterministic theory is just a point a in the state space. For this reason, it is natural that the entropy as a function of the "state" be defined differently for the stochastic theory that includes fluctuations

and the deterministic theory that neglects them.

In a previous work² by one of us, the entropy of the stochastic theory has been defined on the basis of statistical-mechanical arguments. Following that work we put

$$\tilde{S}(p) = -k \int da p(a) \ln \frac{p(a)}{w(a)} + S_{\text{eq}}, \quad (5.1)$$

where S_{eq} is the entropy of the equilibrium distribution $w(a)$.

In the deterministic limit the distribution $p(a)$ shrinks to a single point. Let us express $p(a)$ in terms of a scalar function $\psi(a)$ by

$$p(a) = N^{-1} g^{-1/2}(a) e^{(1/k)\psi(a)}, \quad (5.2)$$

where N is the normalization factor

$$N = \int da g^{-1/2}(a) e^{(1/k)\psi(a)}. \quad (5.3)$$

In the limit $k \rightarrow 0$ the distribution (5.2) sharply concentrates around the position a_{max} , where $\psi(a)$ takes its absolute maximum⁹; a_{max} is the deterministic state that we associate with the distribution $p(a)$. According to (3.12), we write the equilibrium distribution in the form

$$w(a) = N_{\text{eq}}^{-1} g^{-1/2}(a) e^{(1/k)S(a)}, \quad (5.4)$$

with

$$N_{\text{eq}} = \int da g^{-1/2}(a) e^{(1/k)S(a)}. \quad (5.5)$$

The absolute maximum of $S(a)$ is located at the deterministic equilibrium state a_{eq} .

We now insert (5.2)–(5.5) into (5.1). In the limit $k \rightarrow 0$ the integral can be done using the method of steepest descent, and we find

$$\lim_{k \rightarrow 0} \tilde{S}(p) = S(a_{\text{max}}), \quad (5.6)$$

where we have used $S(a_{\text{eq}}) = S_{\text{eq}}$. Since a_{max} is the deterministic state associated with $p(a)$, the definition (3.12) of the entropy S is in fact the $k \rightarrow 0$ limit of the entropy $\tilde{S}(p)$ defined in Eq. (5.1).

B. Second law

The Second Law demands that the entropy is increasing with time. Indeed, if we evaluate the time rate of change of $\tilde{S}(p_t)$ by means of (3.13), we find with (3.12) and (3.14) after simple transformations

$$\begin{aligned} \frac{d}{dt} \tilde{S}(p_t) &= k^2 \int da p_t(a) D^{ij}(a) \left(\ln \frac{p_t(a)}{w(a)} \right)_{,i} \left(\ln \frac{p_t(a)}{w(a)} \right)_{,j} \geq 0. \end{aligned} \quad (5.7)$$

The reversible part of the Fokker-Planck drift

$$K_{\text{rev}}^I = A^{IJ} \chi_J + k g^{1/2} (A^{IJ} g^{-1/2})_{,J} = k w^{-1} \frac{\partial}{\partial a^J} w A^{IJ} \quad (5.8)$$

does not contribute to the time rate of change of the entropy \bar{S} .

The inequality (5.7) derived within the stochastic theory cannot be lost in the $k \rightarrow 0$ limit. By inserting (5.2)–(5.5) into (5.7) we obtain for $k \rightarrow 0$

$$\frac{d}{dt} S(a(t)) = D^{ij}(a(t)) \chi_i(t) \chi_j(t) \geq 0, \quad (5.9)$$

where $a(t)$ is the deterministic state associated with $p_t(a)$. Equation (5.9) coincides with our previous result (2.13).

C. Note on reversible drift

It is worthwhile noting that there is generally a difference between the reversible drift K_{rev}^I that appears in the Fokker-Planck equation and the reversible part r^I of the deterministic drift. This is as it should be. The drift K_{rev}^I generates a reversible flow $-(\partial/\partial a^I) K_{\text{rev}}^I(a) p(a)$ in the space of distributions $p(a)$ which vanishes for the equilibrium distribution

$$-\frac{\partial}{\partial a^I} K_{\text{rev}}^I(a) w(a) = 0, \quad (5.10)$$

and which is orthogonal to $\delta \bar{S}(p)/\delta p(a)$ in the sense of

$$-\int da \frac{\delta \bar{S}(p)}{\delta p(a)} \frac{\partial}{\partial a^I} K_{\text{rev}}^I(a) p(a) = 0. \quad (5.11)$$

On the other hand, the drift $r^I(a)$ is a reversible flow in the state space which vanishes at the equilibrium state

$$r^I(a_{\text{eq}}) = 0 \quad (5.12)$$

and which is orthogonal to $\partial \bar{S}/\partial a^I = \chi_I$ in the sense of

$$\frac{\partial S(a)}{\partial a^I} r^I(a) = 0. \quad (5.13)$$

We are aware of the fact that there are models like the Van der Pol oscillator¹⁰ for which Eq. (5.12) is not satisfied. Such models require that elements of the matrix $A^{IJ}(a)$ approach infinity as a approaches a_{eq} . This is not compatible with the commutator representation of A^{IJ} which follows from statistical mechanics. We conclude that those models cannot describe a system whose stationary state is an equilibrium state. Indeed, the Van der Pol oscillator is often used as a simple model for lasers, which has been based on far-from-equilibrium statistical mechanics. Con-

ditions (5.10)–(5.13) express fundamental properties of the stochastic and deterministic theories of closed systems.

VI. PATH-INTEGRAL REPRESENTATION FOR CONDITIONAL PROBABILITY

A. Conditional probability for small τ

The conditional probability $p_t(a'/a)$ is the solution of the Fokker-Planck equation (3.13) with the initial condition $p_0(a'/a) = \delta(a' - a)$. In a basis where the $\{a^\alpha\}$ and $\{a^i\}$ variables are separated the evolution equation for $p_t(a'/a)$ reads

$$\begin{aligned} \frac{\partial}{\partial t} p_t(a'/a) = & \left(-\frac{\partial}{\partial a^\alpha} v^\alpha(a') - \frac{\partial}{\partial a^i} K^i(a') \right. \\ & \left. + k \frac{\partial^2}{\partial a^i \partial a^j} D^{ij}(a') \right) p_t(a'/a). \end{aligned} \quad (6.1)$$

From the theory of diffusion processes it is known that for a small time interval τ the conditional probability $p_\tau(a'/a)$ derived from (6.1) has the properties

$$\begin{aligned} \int da' p_\tau(a'/a) &= 1, \\ \int da' \Delta^\alpha p_\tau(a'/a) &= v^\alpha(a) \tau + O(\tau^{3/2}), \\ \int da' \Delta^i p_\tau(a'/a) &= K^i(a) \tau + O(\tau^{3/2}), \\ \int da' \Delta^\alpha \Delta^\beta p_\tau(a'/a) &= O(\tau^{3/2}), \\ \int da' \Delta^\alpha \Delta^i p_\tau(a'/a) &= O(\tau^{3/2}), \\ \int da' \Delta^i \Delta^j p_\tau(a'/a) &= 2k D^{ij}(a) \tau + O(\tau^{3/2}), \end{aligned} \quad (6.2)$$

where $\Delta^\alpha = a^\alpha - a^\alpha$ and $\Delta^i = a^i - a^i$. The third and higher moments are at least of order $\tau^{3/2}$.

Because of (6.2), $p_\tau(a'/a)$ can be written in the form

$$p_\tau(a'/a) = \delta(\Delta^\alpha - \tau v^\alpha(a)) \Pi_\tau(\Delta^i, a) [1 + O(\tau^{3/2})], \quad (6.3)$$

where $\delta(\Delta^\alpha)$ is the n -dimensional δ function and where $\Pi_\tau(\Delta^i, a)$ is a distribution in the $\{a^i\}$ subspace with the properties

$$\begin{aligned} \int \prod_k da^k \Pi_\tau(\Delta^i, a) &= 1 + O(\tau^{3/2}), \\ \int \prod_k da^k \Delta^i \Pi_\tau(\Delta^i, a) &= K^i(a) \tau + O(\tau^{3/2}), \\ \int \prod_k da^k \Delta^i \Delta^j \Pi_\tau(\Delta^i, a) &= 2k D^{ij}(a) \tau + O(\tau^{3/2}). \end{aligned} \quad (6.4)$$

Higher moments are at least of the order $\tau^{3/2}$.

B. Modified Lagrangian

It is shown in Appendix A that $\Pi_\tau(\Delta^i, a)$ may be written

$$\Pi_\tau(\Delta^i, a) = \frac{1}{(4\pi k \tau)^{(N-n)/2} D(a')^{1/2}} \times \exp\left(-\frac{1}{2k} \bar{A}_\tau(\Delta^i, a)\right) [1 + O(\tau^{3/2})]; \tag{6.5}$$

$D(a)$ is the determinant of the matrix $D^{ij}(a)$, and $\bar{A}_\tau(\Delta^i, a)$ is given by

$$A_\tau(\Delta^i, a) = \int_0^\tau dt \bar{O}(a(t), \dot{a}(t))_{\min} \tag{6.6}$$

with the modified Lagrangian

$$\bar{O}(a, \dot{a}) = \frac{1}{2} D_{ij}(\dot{a}^i - l^i)(\dot{a}^j - l^j) + k D^{1/2}(l^i D^{-1/2})_{,i} - k(\ln D^{1/2})_{,i} a^\alpha + \frac{2}{3} k^2 R. \tag{6.7}$$

The integral in (6.6) is over the minimum path connecting the states a and $a' = (a^\alpha + v^\alpha \tau, a^i + \Delta^i)$. The drift l^i is defined by

$$l^i = K^i - k D^{1/2}(D^{ij} D^{-1/2})_{,j}, \tag{6.8}$$

and R is defined in terms of D^{ij} in the same way as the corresponding quantity in paper I is defined in terms of L^{ij} . Combining (6.3) and (6.5), we obtain for the conditional probability $p_\tau(a'/a)$ the expression

$$p_\tau(a'/a) = \frac{\delta(\Delta^\alpha - \tau v^\alpha(a))}{(4\pi k \tau)^{(N-n)/2} D(a')^{1/2}} \times \exp\left(-\frac{1}{2k} \int_0^\tau dt \bar{O}(a(t), \dot{a}(t))_{\min}\right) \times [1 + O(\tau^{3/2})]. \tag{6.9}$$

C. Path integral

The conditional probability for a finite time difference s can be obtained from (6.9) by repeated use of the Chapman-Kolmogorov equation. Essentially the same argumentation as in I, Sec. V B leads to

$$p_s(a'/a) = \lim_{M \rightarrow \infty} \int \frac{\delta(a_M^\alpha - a_{M-1}^\alpha - \tau v^\alpha(a_{M-1}))}{(4\pi k \tau)^{(M/2)(N-n)} D(a_M)^{1/2}} \prod_{m=1}^{M-1} \frac{da_m \delta(a_m^\alpha - a_{m-1}^\alpha - \tau v^\alpha(a_{m-1}))}{D(a_m)^{1/2}} \exp\left(-\frac{1}{2k} \int_0^s d\tau \bar{O}(a(t), \dot{a}(t))_{\min}\right), \tag{6.10}$$

where $a_0 = a$, $a_M = a'$, and $\tau = s/M$. Because of the δ functions, the succession of states a_m at times $t_m = m\tau$ is constrained by $a_m^\alpha - a_{m-1}^\alpha = v^\alpha(a_{m-1})\tau$, which ensures that only paths with differentiable

$a^\alpha(t)$ satisfying $\dot{a}^\alpha(t) = v^\alpha(t)$ contribute in the limit $M \rightarrow \infty$.

Since $\delta(\Delta^\alpha - \tau v^\alpha) = (1/\tau^n) \delta(\Delta^\alpha/\tau - v^\alpha)$ it is natural to introduce the integration measure

$$D[a(t)] \delta(\dot{a}^\alpha(t) - v^\alpha(t)) = \lim_{M \rightarrow \infty} \frac{g(a_M)^{1/2} \delta((a_M^\alpha - a_{M-1}^\alpha)/\tau - v^\alpha(a_{M-1}))}{\tau^{Mn} (4\pi k \tau)^{M(N-n)/2} D(a_M)^{1/2}} \prod_{m=1}^{M-1} \frac{da_m \delta((a_m^\alpha - a_{m-1}^\alpha)/\tau - v(a_{m-1}))}{D(a_m)^{1/2}}. \tag{6.11}$$

This measure is invariant under all transformations of the variables a that do not mix the $\{a^\alpha\}$ with the $\{a^i\}$ subset. With (6.11) we may abbreviate the limit (6.10) as the path integral¹¹

$$p_s(a'/a) = g(a')^{-1/2} \int D[a(t)] \delta(\dot{a}^\alpha(t) - v^\alpha(t)) \exp\left(-\frac{1}{2k} \int_0^s dt \bar{O}(a(t), \dot{a}(t))\right). \tag{6.12}$$

Equation (6.12) is a generalization of the corresponding result in I in two ways. First, it holds for a more general choice of the metric tensor; second, it is also valid for systems with a singular diffusion matrix. It is interesting to note from our result that it is not possible to obtain the modified Lagrangian (6.7) from the modified Lagrangian in I by simply taking appropriate elements of the matrix of transport coefficients equal to zero, because for such a limit there exists some arbitrariness for the definition of the δ function in the measure in Eq. (6.12).

Using (3.24)–(3.28), and (6.8), we find

$$v^\alpha = f^\alpha + k g^{1/2} (A^{\alpha I} g^{-1/2})_{,I} \tag{6.13}$$

$$l^i = f^i + k g^{1/2} (A^{iJ} g^{-1/2})_{,J} + k D^{ij} [\ln(D^{1/2} g^{-1/2})]_{,j}.$$

With (2.17) and (6.7) we see that

$$v^\alpha \rightarrow f^\alpha, \quad \bar{O}(a, \dot{a}) \rightarrow O(a, \dot{a}), \quad \text{for } k \rightarrow 0.$$

Hence the path probability in the $k \rightarrow 0$ limit sharply concentrates about the deterministic path and fluctuations vanish. The explicit k dependence of

$\tilde{O}(\alpha, \dot{\alpha})$, which is given by Eqs. (6.7) and (6.13), can be used to set up a perturbation scheme where k is used as an expansion parameter in order to calculate fluctuation corrections to the deterministic theory.

VII. CONCLUSIONS

We have examined the deterministic as well as the stochastic theory of nonlinear irreversible systems. It has been shown that the deterministic equations of motion in Onsager's form (2.3) can be looked upon as a certain $k \rightarrow 0$ limit of a stochastic theory characterized by a Fokker-Planck equation [(3.13)]. The limit $k \rightarrow 0$ can only be defined in a covariant way, if we introduce a metric in state space. We have defined this metric in such a way that the metric tensor is constant whenever the macroscopic variables are sums of molecular variables on a kinetic model. Given the metric, we can reconstruct the Fokker-Planck equation (3.13) from the transport equations (2.3) by use of (3.12) and (3.14).

The connection of the deterministic with the stochastic theory is such that both have the well-known properties of theories describing the dynamics of closed physical systems. For instance, the reciprocal relations (2.5) are connected with the potential conditions (3.16) which are themselves a consequence of the microscopic reversibility. Further, the irreversible fluxes lead in both theories to an increase of the entropy.

Given the deterministic theory, fluctuations are often introduced by simply adding a random force to the deterministic equations of motion. The Langevin equations obtained by such a procedure are equivalent to a Fokker-Planck equation. However, the reversible part of the resulting Fokker-Planck drift coincides with the reversible deterministic drift, so that either the potential conditions or Eq. (5.13) will be violated in general. Hence the so-called Langevin assumption may not be correct for nonlinear systems with partly reversible fluxes.

The deterministic limit involves a metric in state space which has been defined here differently from our previous work I. This means that the deterministic equations of I are not precisely the same as here. The entropy S differs by a term proportional to the logarithm of the ratio of the invariant volume elements in state space. Hence the fluctuation hypothesis of I may not hold within the present theory. We believe that the new definition of the metric is more adequate and gives a more intuitive meaning to the deterministic limit, despite the fact that we now need, besides a tensor L^{ij} and a scalar S , an additional independent invariant volume element $g^{-1/2}(a)da$ to specify the theory on the phenomenological level.

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APPENDIX A: DERIVATION OF EQ. (6.5)

Let us define a Lagrangian

$$\hat{O}(a^\alpha, a^i(t), \dot{a}^i(t)) = \frac{1}{2} D_{ij} (\dot{a}^i - l^i) (\dot{a}^j - l^j) + k D^{1/2} (l^i D^{-1/2})_{,i} + \frac{2}{3} k^2 R, \quad (A1)$$

where D_{ij} and l^i are functions of $(a^\alpha, a^i(t))$; that means the $\{a^\alpha\}$ are for the moment being considered as fixed parameters. The Lagrangian defines an action

$$\hat{A}_\tau(\Delta^i, a^\alpha, a^i) = \int_0^\tau dt \hat{O}(a^\alpha, a^i(t), \dot{a}^i(t))_{\min}, \quad (A2)$$

where the integral is over the minimum path connecting $\{a^i\}$ and $\{a^i + \Delta^i\}$. Except for some differences in notation, Eqs. (A1) and (A2) coincide with Eqs. (5.8) and (5.9) in paper I, since the $\{a^\alpha\}$ are considered to be fixed.

If we now define l^i in analogy to Eq. (5.3) in I by (6.8), we can conclude from (5.5) and (5.2) in I that the distribution

$$\Pi_\tau(\Delta^i, a) = [1/(4\pi k \tau)^{(N-n)/2} D(a^\alpha, a^i + \Delta^i)^{1/2}] \times \exp\left(-\frac{1}{2k} \hat{A}_\tau(\Delta^i, a)\right) \quad (A3)$$

has the properties (6.4). Here we have taken into account that there are $N-n$ variables $\{a^i\}$.

Now, the $\{a^\alpha\}$ are not really fixed in the time interval $0 \leq t \leq \tau$; rather they depend on t according to

$$a^\alpha(t) = a^\alpha + v^\alpha t + O(\tau^{3/2}). \quad (A4)$$

Therefore we must consider the Lagrangian \hat{O} as a function of time-dependent $\{a^\alpha(t)\}$. Since for the present consideration terms of the order $\tau^{3/2}$ are negligible, we may look upon $a^\alpha(t)$ as a given function of t . By means of a Taylor series expansion of the Lagrangian in terms of $a^\alpha(t) - a^\alpha = v^\alpha t + O(\tau^{3/2})$ one shows easily that the time dependence of the $\{a^\alpha\}$ variables just adds the term

$$\delta_1 A_\tau = \frac{1}{4} D_{ij|1} v^\alpha \Delta^i \Delta^j \quad (A5)$$

to the action. This term, in order τ , is equivalent to the term

$$\delta_1 A_\tau = \frac{1}{4} D_{ij|1} v^\alpha 2k D^{ij} \tau = -k (\ln D^{1/2})_{,1} v^\alpha \tau. \quad (A6)$$

Next, we change the factor in front of the exponential in Eq. (A4) to

$$1/(4\pi k \tau)^{(N-n)/2} D(a^\alpha + \Delta^\alpha, a^i + \Delta^i)^{1/2}.$$

Because

$$\begin{aligned} 1/D(a^\alpha + \Delta^\alpha, a^i + \Delta^i)^{1/2} &= [1/D(a^\alpha, a^i + \Delta^i)^{1/2}] \\ &\times \exp[-(\ln D^{1/2})_{,1} v^\alpha \tau] \\ &\times [1 + O(\tau^{3/2})], \end{aligned} \quad (\text{A7})$$

this change of the prefactor corresponds to a change of the action by

$$\delta_2 A_\tau = 2k(\ln D^{1/2})_{,1} v^\alpha \tau. \quad (\text{A8})$$

Finally, we add the term $-k(\ln D^{1/2})_{,1} v^\alpha$ to the Lagrangian \hat{O} . This yields the Lagrangian \tilde{O} defined in (6.7) and changes the action by

$$\delta_3 A_\tau = -k(\ln D^{1/2})_{,1} v^\alpha \tau. \quad (\text{A9})$$

Since the sum of $\delta_1 A_\tau$, $\delta_2 A_\tau$, and $\delta_3 A_\tau$ vanishes, we find that the distribution (A3) can also be written in the form (6.5) with an action \tilde{A}_τ defined by (6.6) and (6.7).

APPENDIX B

In I, the Lagrangian $\tilde{O}(a, \dot{a})$ which appears in the path-integral representation is defined in terms of the short-time propagator via the relation

$$\begin{aligned} p_\tau(a'/a) &= [1/(4\pi k \tau)^{n/2} L'^{1/2}] \\ &\times \exp[-(1/2k)\tilde{A}_\tau(a'/a)][1 + O(\tau^{3/2})], \end{aligned} \quad (\text{B1})$$

where the action is the time integral over the Lagrangian along the minimum path connecting $a(t)$ and $a'(t + \tau)$. The approximation of the short-time propagator is made in terms of the natural L^1 norm for distribution functions, i.e., $O(\tau^{3/2})$ means more rigorously

$$\int da' |p_\tau^{\text{ex}}(a'/a) - p_\tau^{\text{app}}(a'/a)| = O(\tau^{3/2}) \quad (\text{B2})$$

for $\tau \rightarrow 0$, where superscripts "ex" and "app" stand for "exact" and "approximate."

In Ref. 6 the short-time propagator is approximated in a different way. Considered as a function of the

initial and final states, the short-time propagator is singular for $\tau \rightarrow 0$. This singularity can be split off by putting

$$\begin{aligned} p_\tau(a'/a) &= [1/(4\pi k \tau)^{n/2} L'^{1/2}] \\ &\times \exp[-(1/2k)L_{,ij}\Delta^i\Delta^j][1 + Y(a, \Delta, \tau)], \end{aligned} \quad (\text{B3})$$

where $\Delta^i = a^{i'} - a^i$ and Y is a regular function which can be expanded in terms of Δ and τ in the usual way. In Ref. 6 the first few terms of the expansion of $Y(a, \Delta, \tau)$ in powers of Δ and τ are determined systematically. The Lagrangian \mathcal{L} is defined via this particular form by taking its limit $\Delta^i = \dot{a}^i \tau$, $\tau \rightarrow 0$. The particular form of the short-time propagator used in Ref. 6 can, in L^1 norm, not be distinguished from other forms of the same accuracy. While the Lagrangian \tilde{O} as defined in I does not depend on the form used, the Lagrangian \mathcal{L} as defined in Ref. 6 depends on that form. However, the particular form of the short-time propagator of Ref. 6 is well distinguished from all other forms, because it satisfies the Chapman-Kolmogorov condition¹² in order $\tau^{3/2}$, with respect to the strong topology used for the expansion in Ref. 6. The other forms satisfy this condition in order $\tau^{3/2}$ with respect to L^1 norm, only.

We now discuss how the definitions of the Lagrangians $\tilde{O}(a, \dot{a})$ in I and $\mathcal{L}(a, \dot{a})$ in Ref. 6 could be modified to become equivalent. The definition of I would, of course, be altered if we chose a different measure prefactor in Eq. (B1). Agreement with the result of Ref. 6 for the Lagrangian would be obtained if we replace in Eq. (B1)

$$L'^{-1/2} \rightarrow \left(\frac{L}{L'}\right)^{1/4} \left(\text{Det} \frac{\partial \tilde{A}_\tau(a'/a)}{\partial a^i \partial a^j}\right)^{1/2}. \quad (\text{B4})$$

The right-hand side arises as a natural measure prefactor in the WKB approximation to the short-time propagator.¹³ A corresponding change of the measure factor in the path integral of Ref. 6 would then make the two definitions of the path integral equivalent in the sense that one definition could be proven from the other as a theorem.

†Deceased.

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⁴The covariance is violated by the requirement of the canonical form of the equilibrium distribution which is not covariant.

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⁷The precise definition of the matrix α^{IJ} depends on the ensemble used. It is a different expression for a system with a microcanonically distributed total energy than for a system with a canonically distributed total energy. However, those differences are irrelevant if the variables a^I describe local properties of the system or properties of subsystems. Moreover, the present discussion applies to every situation if the matrix α^{IJ} is defined appropriately.

⁸R. Graham and H. Haken, Z. Phys. 243, 289 (1971).

⁹If the scalar function $\psi(a)$ has no absolute maximum there is no unique deterministic state associated with the distribution $p(a)$. As far as nonequilibrium states are concerned we can always restrict ourselves to

situations where the deterministic limit is uniquely defined. If the equilibrium distribution leads to an ambiguity for the definition of the deterministic equilibrium state, we can still define the deterministic limit by introducing a small symmetry-breaking term. However, in such a case fluctuations are not a small correction to the deterministic behavior.

¹⁰See, for example, Ref. 3(a).

¹¹In I and Ref. 6 slightly different definitions of path-integral representations have been used. The differences are discussed in Appendix B. For the sake of unity of presentation, we shall follow the line of reasoning of I.

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