*Journal of Statistical Physics, Vol. 107, Nos. 5/6, June 2002 (© 2002)*

# Thermodynamics of the General Diffusion Process: Time-Reversibility and Entropy Production

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*Received September 17, 2000; accepted December 31, 2001*

We introduce an axiomatic thermodynamic theory for the general diffusion process and prove a theorem concerning entropy and irreversibility: the equivalence among time-reversibility, zero entropy production, symmetricity of the stationary diffusion process, and a potential condition.

**KEY WORDS:** Elliptic equation; entropy production; invariant measure; macromolecular mechanics; thermodynamics.

## 1. INTRODUCTION

Diffusion processes are important models for many equilibrium and nonequilibrium phenomena. They are widely considered as a phenomenological approach to molecular systems with fluctuations. One of the well-known examples is the theory of polymer dynamics in an ambient fluid.[(2)] Recently, motivated by work on biological motor molecules which convert chemical energy into mechanical work,[(19, 5)] it becomes evident that a thermodynamic theory, both for equilibrium and more importantly for nonequilibrium steady-state (NESS), can be developed for the general diffusion process.[(25, 22)] For more details of the applications of diffusion models and the motivation for the present mathematical analysis, see ref. 26.

By thermodynamics, we mean the theory that connects key concepts such as entropy, heat, their respective production and dissipation, and

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0022-4715/02/0600-1129/0 

irreversibility with the stochastic dynamics. The essential difference between a synthetic polymer and a biological motor-protein is that the former has zero heat dissipation and entropy production while for the latter they are positive.[21] It should be noted and as we shall show that while the number of degrees of freedom in a stochastic model is not necessarily large, the random collisions with the solvent molecules, modeled by a Wiener process, provide a sufficient large interacting molecular system in which thermodynamics is valid.

Heat dissipation and entropy production also play important roles in the mathematical formulations for the NESS theory motivated by a computer simulation of driven fluids.[3] Numerical observations have led to a surge of mathematical analyses for NESS as either a dynamical system[34, 14] or a stochastic process.[11, 12, 1] A unifying mathematical feature of the entropy production has been established for the axiom-A system, the diffusion process, and interacting particle systems.[9, 15]

We consider the stochastic models in the form of the stochastic differential equation

$$\frac{\mathrm{d}x}{\mathrm{d}t} = b(x) + \Gamma\xi(t), \qquad x \in \mathbb{R}^n \tag{1}$$

where $\Gamma$ is a nonsingular matrix and $\xi(t)$ is the "derivative" of an $n$-dimensional Wiener process. Following the standard polymer theory,[2] $x$ can be thought of as the coordinates of $N$ "atoms" in a single macromolecule who stochastic dynamics in an ambient fluid is assumed to be overdamped.[26] Hence $n = 3N$. The corresponding Fokker–Planck equation is

$$\frac{\partial u}{\partial t} = \mathscr{L}^* u(t, x) \triangleq \nabla \cdot \left( \frac{1}{2} A(x)\, \nabla u - b(x)\, u \right), \qquad (A = \Gamma\Gamma^T) \tag{2}$$

$$u(0, x) = f(x) \tag{3}$$

where $\mathscr{L}^*$ denotes the adjoint of operator $\mathscr{L}$.

In mathematics, ref. 31 gave the first rigorous result on irreversibility and entropy production for the case of discrete-state Markov chains. A comprehensive treatment of this case is ref. 10. For the general diffusion process with bounded coefficients $A(x)$ and $b(x)$, related results were announced in refs. 32 and 33 using Girsanov formula. This approach, however, is not valid for the most applications with unbounded $A(x)$ and $b(x)$ on $\mathbb{R}^n$. For linear $b(x)$ in Eq. (1), the mathematical task is significantly simplified and the diffusion process is also Gaussian.[22] A related study of interacting particle systems can be found in ref. 16.

This paper focuses on the nonlinear stochastic differential equation (1). We assume:

(1) $A(x)=\{a_{ij}(x)\}$, $b(x)=\{b_j(x)\}$ are smooth;

(2) $\nabla\cdot b(x)\leqslant\mu_0$, where $\mu_0$ is a constant;

(3) uniformly elliptic condition $\sum_{i,j=1}^{n} a_{ij}(x)\,\xi_i\xi_j\geqslant r\sum_{i=1}^{n}\xi_i^2$, $\forall\xi\in\mathbb{R}^n$, where $r$ is a positive constant.

The paper is organized as follows. Section 2 provides heuristically the essential elements of the thermodynamic theory of diffusion[25, 22] including the definitions for entropy production rate and time-reversibility. It als states our main theorem on the thermodynamics of diffusion process. In order to provide a mathematically rigorous proof for the theorem, specifically to obtain the self-adjoint property in Eq. (13), Section 3 gives a summary for the relevant mathematical results and notations on the general diffusion process without proof.[36, 28, 29] With unbounded $a_{ij}(x)$ and $b(x)$ on $\mathbb{R}^n$, the standard method of integration by parts is not applicable. Finally in Section 4, the equivalence among time-reversibility, zero entropy production rate, symmetricity, and potential condition is established for the general minimal diffusion process. The paper concludes with Section 5. A mathematically more complete version of the present paper can be found in ref. 27.

## 2. THE THERMODYNAMIC FORMALISM OF THE DIFFUSION PROCESS

The most important concepts in thermodynamics are mechanical work, heat, and entropy. The thermodynamic theory of the diffusion process provides mathematical definitions for these three quantities. The entropy has the well-known definition $e[P]=\int_{\mathbb{R}^n} P(t,x)\log P(t,x)\,dx$ which is a functional of the probability density $P(t,x)$, the solution to Eq. (2). For more discussion on the Gibbs entropy and its physical interpretation, see ref. 23. The concept of mechanical work, energy and its dissipation are stochastic according to our model. They are related by energy conservation. Hence there is a functional of the diffusion trajectory $x(t)$: $W(t)=\int_0^t F(x)\circ dx(s)$ where $F(x)=2A^{-1}(x)\,b(x)$ and $\circ$ denotes the Stratonovich integral.[12] The mean heat dissipation rate (hdr), thus, is the expectation $\lim_{t\to\infty} E[W(t)/t]=\int_{\mathbb{R}^n} F(x)\,\mathscr{J}\,dx$, which is the product of force $F(x)$ and probability flux

$$\mathscr{J}=-\tfrac{1}{2}A(x)\,\nabla P(t,x)+b(x)\,P(t,x)$$

The force $F(x)$ in turn is the product of frictional coefficient $(2A^{-1})$ and velocity $b(x)$.

The rate of the increase of entropy is then

$$\dot{e}[P] = epr - h\,dr \tag{4}$$

where

$$epr = \int_{\mathbb{R}^n} 2(\mathcal{J} A^{-1}(x)\,\mathcal{J})\,P^{-1}(t, x)\,\mathrm{d}x$$

It is meaningful from thermodynamics point of view to identify the first term in Eq. (4) with the entropy production rate. In a time independent NESS, $\dot{e} = 0$, and the entropy production is balanced by the heat dissipation. Eq. (4) is the well-known equation for entropy balance. It is the central hypothesis of nonequilibrium thermodynamics.[18] The diffusion theory, therefore, provides the nonequilibrium thermodynamics with a mesoscopic equation of motion (Eq. (1)).

If the force $F(x) = -\nabla U(x)$ is conservative, $W(t)$ is bounded almost surely. In this case one can further introduce Helmholtz free energy $h[P] = u[P] - e[P]$ in which $u[P] = \int_{\mathbb{R}^n} U(x)\,P(x)\,dx$ is the internal energy and $\dot{u} = -h\,dr$, as expected due to energy conservation. Then $\dot{h} = -epr \leqslant 0$ with the equality hold true for an equilibrium process. This is the second law of thermodynamics applied to isothermal processes with canonical ensembles. For nonconservative force $F(x)$ without a potential, $W(t)$ increases without bound, and the free energy can not be defined. In this case, one writes the force in terms of Helmholtz–Hodge decomposition: $F(x) = -\nabla\phi + \gamma(x)$ where the $\gamma$ is related to the circulation of the irreversible process.[31, 19, 30] There is a geometric representation for the energy conservation as well. It can be shown that solving the stationary solution to Eq. (2) is equivalent to requiring[7]

$$\nabla \cdot \gamma - \nabla\phi \cdot \gamma = 0 \tag{5}$$

Then $F(x) \circ dx = -d\phi + \gamma \circ dx$ where the term on the left is work, and the terms on the right are mechanical energy and dissipated heat, respectively. Finally, Eq. (5) is also a generalization of the Tellegen theorem, i.e., if $\nabla \cdot \gamma = 0$, then $\nabla\phi \cdot \gamma = 0$.[10, 35]

Lebowitz and Spohn[12] also studied the generating function of $W(t)$ in term of the theory of large deviations: the limit

$$\lim_{t \to \infty} -\tfrac{1}{t} \log E[e^{-\lambda W(t)}]$$

is convex and possesses a certain symmetry with respect to $\lambda$. It is zero for equilibrium $W(t)$, and the symmetry generalizes positivity of hdr (=epr) in NESS.

The fundamental theorem of our thermodynamic theory is the mathematical equivalence between the time-reversibility and vanishing entropy production for the diffusion process (Theorem 1). We have the definitions for entropy production rate and time-reversibility:

**Definition 1.** The entropy production rate, epr, of a stationary diffusion process defined by Eq. (1) is

$$\frac{1}{2}\int (\nabla \log P(t,x) - 2A^{-1}b(t,x))^T A(\nabla \log P(t,x) - 2A^{-1}b(x))\, P(t,x)\, dx$$

In the stationary case, $P(t,x) = w(x)$.

**Definition 2.** A stationary stochastic process $\{x(t); t \in \mathbb{R}\}$ is time-reversible if $\forall m \in \mathbb{N}$ and every $t_1, t_2, \ldots, t_m \in \mathbb{R}$, the joint probability distribution

$$P(x(t_1), x(t_2), \ldots, x(t_m)) = P(x(-t_1), x(-t_2), \ldots, x(-t_m))$$

We now state the main theorem:

**Theorem 1.** For the stationary diffusion process defined by Eq. (1), the following four statements are equivalent:

(i) The process is time-reversible;

(ii) Its corresponding elliptic operator $\mathscr{L}^*$ is symmetric on $C_0^\infty(\mathbb{R}^n)$ with respect to a positive function $w^{-1}(x)$, $w(x) \in L^1(\mathbb{R}^n)$, i.e., $\int_{\mathbb{R}^n} w(x)\, dx < \infty$;

(iii) The process has zero entropy production rate (epr);

(iv) The force $F(x) = 2A^{-1}(x)\, b(x)$ has a potential function.

## 3. SOME RELEVANT MATHEMATICAL RESULTS

We denote

$$C(\mathbb{R}^n) = \{\text{bounded continuous function } f(x)\}$$

$$C_0(\mathbb{R}^n) = \{f \in C(\mathbb{R}^n) \mid \lim_{|x| \to \infty} f(x) = 0 \text{ uniformly}\}$$

$$\|f(x)\| = \sup_{x \in \mathbb{R}^n} |f(x)|$$

$\|\cdot\|$ is the norm on $C(\mathbb{R}^n)$ and $C_0(\mathbb{R}^n)$

The conjugate of the Fokker–Planck equation (2) is the Kolmogorov backward equation:

$$\frac{\partial u(t,x)}{\partial t}=\mathscr{L}u(t,x)=\frac{1}{2}\sum_{i,j=1}^{n}a_{ij}(x)\frac{\partial^2 u}{\partial x_i\,\partial x_j}+\sum_{i=1}^{n}b_i(x)\frac{\partial u}{\partial x_i}\qquad (t>0,\ x\in\mathbb{R}^n) \tag{6}$$

For the solutions to Eqs. (2) and (6), we have the following theorems:

**Theorem 2.** If the coefficients of Eq. (6) satisfy assumptions (1) and (3), then there exists a Banach space $\hat{C}(\mathbb{R}^n)$, $C_0(\mathbb{R}^n)\subset\hat{C}(\mathbb{R}^n)\subset C(\mathbb{R}^n)$, and the positive semigroup $T(t)$ generated by the solution to the Cauchy problem (2) and (3) with initial data $f(x)$ exists in $\hat{C}(\mathbb{R}^n)$.

The minimal solution to the Kolmogorov backward equation then is $T(t)\,f$. Solution uniqueness actually does not hold true for general $a_{ij}(x)$ and $b_i(x)$. The next theorem is about the Kolmogorov forward equation and the relation between the solutions to the two equations.

**Theorem 3.** If the coefficients of Eq. (2) satisfy the assumptions (1), (2) and (3), then there exists a Banach space $\tilde{C}(\mathbb{R}^n)$, $C_0(\mathbb{R}^n)\subset\tilde{C}(\mathbb{R}^n)\subset C(\mathbb{R}^n)$, and $\forall g\in\tilde{C}(\mathbb{R}^n)$, the solution of the Cauchy problem (2) and (3) with initial data $g(x)$ exists in $\tilde{C}(\mathbb{R}^n)$, which is denoted by $\tilde{T}(t)\,g$. Furthermore, $\forall f, g\in C_0(\mathbb{R}^n)$

$$\int (T(t)\,f)\,g\,\mathrm{d}x=\int f(\tilde{T}(t)\,g)\,\mathrm{d}x \tag{7}$$

The proof for Theorems 2 and 3 is based on and expands the classic work.[4, 17] The essential steps are

(i) $\forall n\in\mathbb{N}$ (the positive integers) on the bounded sphere $B_n\triangleq\{x\in\mathbb{R}^n\mid |x|\leqslant n\}$, one solves the elliptic equation.

(ii) By taking monotone limit of the above, $\forall\lambda>0$ and $\forall f\in C(\mathbb{R}^n)$, define $R_n(\lambda)\,f=u_n$ on $B_n$ with 0 elsewhere. $u_n$ is the solution to $(\lambda-\mathscr{L})\,u=fg_n$ with boundary condition $u|_{\partial B_n}=0$. $g_n(x)$: $\mathbb{R}^n\to\mathbb{R}$, is a sequence of smooth functions $\in C_0^\infty(\mathbb{R}^n)$ with $g_n(x)=0$ for $x\notin B_n$. Then as the limit of $R_n(\lambda)$ with $n\to\infty$, the resolvent operator $R(\lambda)$: $C(\mathbb{R}^n)\to C(\mathbb{R}^n)$, satisfying $\forall f\in C(\mathbb{R}^n)$, $(\lambda-\mathscr{L})\,R(\lambda)\,f=f$ in $\mathbb{R}^n$ and $\|R(\lambda)\|\leqslant\frac{1}{\lambda}$;

(iii) Using $R(\lambda)$, define a Banach space $\hat{C}(\mathbb{R}^n)$, satisfying $C_0(\mathbb{R}^n)\subset\hat{C}(\mathbb{R}^n)\subset C(\mathbb{R}^n)$;

(iv) The resolvent operators of $\mathscr{L}$ in $\hat{C}(\mathbb{R}^n)$ satisfy the conditions of Hille-Yosida theorem.[37] Hence we obtain the semigroup generated by $\mathscr{L}$ which is the solution to the Cauchy problem (3) and (6).

(v) $\mathscr{L}^* u$ in Eq. (2) contains a term $-\nabla \cdot (b(x)\, u(x))$, so the assumption (2) in Section 1 is required to apply the above steps to prove Theorem 3, which leads to its respective $\tilde{C}(\mathbb{R}^n)$.

For the $T(t)$ and $\tilde{T}(t)$ obtained above we have the following theorems:

**Theorem 4.** $\forall t > 0$, $x \in \mathbb{R}^n$, there is a regular measure $p(t, x, \mathrm{d}y)$, called transition function, which satisfies:

(1) $T(t)\, f(x) = \int p(t, x, \mathrm{d}y)\, f(y)$, $\forall f \in C_0(\mathbb{R}^n)$;

(2) Setting $\Gamma \in \mathscr{B}$, a Borel field generated by $\mathbb{R}^n$, $p(t, x, \Gamma)$ is a Borel measurable function;

(3) The transition functions satisfy the Kolmogorov–Chapman equation

$$p(t+s, x, \Gamma) = \int p(t, x, \mathrm{d}z)\, p(s, z, \Gamma) \quad \text{a.e.}$$

(4) For $\tilde{T}(t)$, there also exists a family of measure $\tilde{p}(t, x, \mathrm{d}y)$, satisfying the same property as $p(t, x, \mathrm{d}y)$; and

$$\tilde{p}(t, x, \mathrm{d}y)\, \mathrm{d}x = p(t, y, \mathrm{d}x)\, \mathrm{d}y \tag{8}$$

The following two theorems state the existence of a positive invariant probability density.

**Theorem 5.** If $\frac{1}{T} \int_0^T T(t)\, f(x)\, \mathrm{d}t$ does not converge to 0 for every $f \in C_0(\mathbb{R}^n)$, $x \in \mathbb{R}^n$, then there exists a positive linear functional $\Lambda$ on $\hat{C}(\mathbb{R}^n)$, which is invariant under $T(t)$: $\Lambda(T(t)\, f) = \Lambda(f)$. And corresponding to $\Lambda$, there is a regular measure $\theta(\mathrm{d}x)$, satisfying

$$\int T(t)\, f(x)\, \theta(\mathrm{d}x) \leqslant \int f(x)\, \theta(\mathrm{d}x) \qquad f \in C_0(\mathbb{R}^n) f \geqslant 0$$

Furthermore $\theta(\mathrm{d}x)$ has a density $\theta(x) > 0$.

**Theorem 6.** Under the conditions of Theorem 5, $\theta(x)$ is invariant under $T(t)$:

$$\int p(t, x, \mathrm{d}y)\, \theta(\mathrm{d}x) = \theta(\mathrm{d}y)$$

Since $T(t)$ has a family of transition functions $p(t, x, \mathrm{d}y)$ and an invariant measure $\theta(\mathrm{d}y)$, one can construct a stationary Markov process by

Kolmogorov theorem, whose transition probability functions are $\{\tilde{p}(t, x, \mathrm{d}y)\}$ and the initial distribution is $\theta(\mathrm{d}x)$. Furthermore, Theorems 5 and 6 together actually shows a weak form of the Foguel alternative given in ref. 13 where diffusions with bounded coefficients $A(x)$ and $b(x)$ are considered.

## 4. TIME-REVERSIBILTY AND ENTROPY PRODUCTION

We now establish the equivalence in Theorem 1.

*Proof.*

(i) $\Rightarrow$ (ii). This result is known to physicists. The proof for a discrete state Markov process is due to Kolmogorov. According to the definition of reversibility, with the transition function $\tilde{p}(t, x, dy)$ and the positive stationary measure $\theta(x)$, we have $\forall A, B \in \mathscr{B}$

$$\int_B \int_A \tilde{p}(t, x, \mathrm{d}y)\, \theta(y)\, \mathrm{d}x = \int_A \int_B \tilde{p}(t, y, \mathrm{d}x)\, \theta(x)\, \mathrm{d}y$$

By the standard method in probability, this leads to

$$\int_B \int_A \phi(x)\, \tilde{p}(t, x, \mathrm{d}y)\, \psi(y)\, \theta(y)\, \mathrm{d}x = \int_A \int_B \psi(y)\, \tilde{p}(t, y, \mathrm{d}x)\, \phi(x)\, \theta(x)\, \mathrm{d}y \tag{9}$$

where $\phi(x), \psi(x) \in C_0^\infty(\mathbb{R}^n)$. Differentiating both sides of (9) with respect to $t$ at $t = 0$, we have

$$\int_{\mathbb{R}^n} \phi(x)\, \mathscr{L}^*[\theta(x)\, \psi(x)]\, \mathrm{d}x = \int_{\mathbb{R}^n} \psi(y)\, \mathscr{L}^*[\theta(y)\, \phi(y)]\, \mathrm{d}y$$

Let $f(x) = \theta(x)\, \phi(x)$ and $g(x) = \theta(x)\, \psi(x)$, then $f$ and $g$ are two arbitrary functions in $C_0^\infty(\mathbb{R}^n)$. Since $\theta(x) > 0$,

$$\int_{\mathbb{R}^n} \theta^{-1}(x)\, f(x)\, \mathscr{L}^*[g(x)]\, \mathrm{d}x = \int_{\mathbb{R}^n} \theta^{-1}(y)\, g(y)\, \mathscr{L}^*[f(y)]\, \mathrm{d}y$$

Therefore, the operator $\mathscr{L}^*$ is symmetric with respect to the reciprocal of its stationary distribution $\theta(x)$: $w(x) = \theta(x)$.

(ii) $\Rightarrow$ (iii). The differential operator $\mathscr{L}^*$ can also be rewritten as

$$\mathscr{L}^* f = \tfrac{1}{2} \nabla \cdot (A \nabla f) - (\nabla f) \cdot b(x) - f \nabla \cdot b(x)$$

The statement (ii) is

$$\int e^U g(x)\, \mathscr{L}^*[f(x)]\, \mathrm{d}x = \int e^U f(x)\, \mathscr{L}^*[g(x)]\, \mathrm{d}x$$

in which the positive $w(x) = e^{-U}$, $f$ and $g \in C_0^\infty(\mathbb{R}^n)$ are arbitrary functions. This leads to

$$\int e^U g(\tfrac{1}{2}\nabla \cdot (A\nabla f) - (\nabla f)\cdot b(x))\mathrm{d}x = \int e^U f(\tfrac{1}{2}\nabla \cdot (A\nabla g) - (\nabla g)\cdot b(x))\, \mathrm{d}x$$

Through integration by part, the first term on the left-hand-side (and similarly for the right-hand-side)

$$\int e^U g\nabla \cdot (A\nabla f)\, \mathrm{d}x = -\int e^U (\nabla g)\, A(\nabla f)\, \mathrm{d}x - \int e^U g(\nabla U)\, A(\nabla f)\, \mathrm{d}x$$

and we have

$$\int e^U g(\tfrac{1}{2}(\nabla U)\, A(\nabla f) + (\nabla f)\cdot b(x))\, \mathrm{d}x = \int e^U f(\tfrac{1}{2}(\nabla U)\, A(\nabla f) + (\nabla g)\cdot b(x))\, \mathrm{d}x$$

By a simple rearrangement, we have

$$\int e^U (g\nabla f - f\nabla g)\cdot(\tfrac{1}{2} A\nabla U + b(x))\, \mathrm{d}x = 0$$

Since $f$ and $g$ are arbitrary, we have $\frac{1}{2} A\nabla U + b(x) = 0$ in which $U = -\log w$. Therefore

$$\nabla \log w(s) - 2A^{-1} b(x) = 0$$

which means epr $= 0$.

(iii) $\Rightarrow$ (iv). The statement epr $= 0$ leads to

$$\tfrac{1}{2} A\nabla\theta(x) - b(x)\,\theta(x) = 0 \tag{10}$$

in which $\theta(x)$ is positive. Hence we have $2A^{-1}(x)\, b(x) = \nabla \ln \theta(x)$. That is $F(x)$ has a potential function.

(iv) $\Rightarrow$ (i). The equivalence between the potential condition, the symmetricity of the differential operator, and the time-reversibility are widely known to physicists. The usual proof involves integration by parts. However, for the unbounded $a_{ij}(x)$ and $b(x)$ on $\mathbb{R}^n$, boundary term vanishing becomes a delicate issue. Thus we use the results and notations given in Section 3 to circumvent the difficulty.

From the potential condition, i.e., Eq. (10) we know

$$\mathscr{L}^*\theta = \nabla\cdot(\tfrac{1}{2}A\nabla\theta - b(x)\,\theta) = 0 \tag{11}$$

With Eqs. (10) and (11), the operators $R_n(\lambda)$, $R(\lambda)$ in Theorem 2 and their corresponding $\tilde{R}_n(\lambda)$, $\tilde{R}(\lambda)$ have properties as follows.

First, $\theta R_n(\lambda)(\psi) = \tilde{R}_n(\lambda)(\theta\psi)$, where $\psi \in C_0^\infty(\mathbb{R}^n)$. This is because

$$\begin{aligned}
&\mathscr{L}^*(\theta R_n(\lambda)(\psi))\\
&\quad= \tfrac{1}{2}\theta\nabla\cdot A\nabla(R_n(\lambda)(\psi)) + \theta b(x)\cdot\nabla(R_n(\lambda)(\psi))\\
&\qquad+ \tfrac{1}{2}R_n(\lambda)(\psi)\,\nabla\cdot A\nabla\theta - R_n(\lambda)(\psi)\,b(x)\cdot\nabla\theta - R_n(\lambda)(\psi)\,\theta\nabla\cdot b(x)\\
&\qquad+ \nabla(R_n(\lambda)(\psi))\cdot A\nabla\theta - 2\theta\nabla(R_n(\lambda)(\psi))\cdot b(x)\\
&\quad= \theta\mathscr{L}(R_n(\lambda)(\psi)) + R_n(\lambda)(\psi)\,\mathscr{L}^*(\theta) + (\nabla\cdot R_n(\psi))(A\nabla\theta - 2\theta b(x))
\end{aligned}$$

and Eq. (11) which leads to $\mathscr{L}^*(\theta R_n(\lambda)\,\psi) = \theta\mathscr{L}R_n(\lambda)(\psi)$. Thus $\theta R_n(\lambda)\,\psi$ satisfies

$$\begin{cases} (\lambda - \mathscr{L}^*)(\theta R_n(\lambda)\,\psi) = \theta(\lambda - \mathscr{L})\,R_n(\lambda)(\psi) = \theta\psi g_n & \text{in } B_n\\ \theta R_n(\lambda)(\psi)|_{\partial B_n} = 0 \end{cases}$$

According to the uniqueness of the solution in $B_n$,

$$\theta R_n(\lambda)(\psi) = \tilde{R}_n(\lambda)(\theta\psi) \tag{12}$$

Second, from (12), $\forall\varphi, \psi \in C_0^\infty(\mathbb{R}^n)$

$$\begin{aligned}
\int \psi g_n \tilde{R}_n(\lambda)(\theta\varphi)\,dx &= \int_{B_n} \psi g_n \theta R_n(\lambda)(\varphi)\,dx\\
&= \int_{B_n} (\lambda - \mathscr{L}^*)\,\tilde{R}_n(\lambda)(\psi\theta)\,R_n(\lambda)(\varphi)\,dx\\
&= \int_{B_n} \tilde{R}_n(\lambda)(\psi\theta)(\lambda - \mathscr{L})(R_n(\lambda)(\varphi))\,dx\\
&= \int_{B_n} \tilde{R}_n(\lambda)(\psi\theta)\,\varphi g_n\,dx\\
&= \int_{B_n} \tilde{R}_n(\lambda)(\psi\theta)\,\varphi g_n\,dx
\end{aligned}$$

Let $n \to \infty$, since $\psi$, $\varphi$ have compact support,

$$\int \psi \tilde{R}(\lambda)(\theta\varphi)\,\mathrm{d}x = \int \varphi \tilde{R}(\lambda)(\theta\psi)\,\mathrm{d}x$$

According to the theory of Laplace transformation, from the fact that $\psi\tilde{T}(t)(\theta\varphi)$, and $\varphi\tilde{T}(t)(\theta\psi)$ are continuous with $t$, we have

$$\int \psi \tilde{T}(t)(\theta\varphi)\,\mathrm{d}x = \int \varphi \tilde{T}(t)(\theta\psi)\,\mathrm{d}x \tag{13}$$

This leads to

$$\iint \psi(x)\,\tilde{p}(t, x, \mathrm{d}y)\,\theta(y)\,\varphi(y)\,\mathrm{d}x = \iint \varphi(y)\,\tilde{p}(t, y, \mathrm{d}x)\,\theta(x)\,\psi(x)\,\mathrm{d}y$$

The standard method of measure theory leads to

$$\int_A \int_B \tilde{p}(t, x, \mathrm{d}y)\,\theta(y)\,\mathrm{d}x = \int_B \int_A \tilde{p}(t, x, \mathrm{d}y)\,\theta(y)\,\mathrm{d}x$$

which means reversibility. ∎

## 5. CONCLUSIONS

We have provided the general diffusion process defined by nonlinear stochastic differential equations (1) with an axiomatic thermodynamic structure in a rigorous mathematical setup. We have introduced fundamental physical concepts of work, heat, entropy, entropy production, and time-reversibility. We demonstrate the fundamental principle of irreversibility by proving the equivalence between time-reversibility, vanishing entropy production, symmetricity of the stationary Markov process, and a potential condition. In a recent work on certain non-Markovian Gaussian processes,[20] it has been suggested that the equivalence between time-reversibility and equilibrium requires some additional conditions. A rigorous mathematical treatment of this problem remains to be developed.

## ACKNOWLEDGMENTS

We thank Professor Zhen-Qing Chen for many helpful discussions.

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