Journal of Statistical Physics, Vol. 66, Nos. 1/2, 1992

Stretched-Exponential Decay Laws of General Defect Diffusion Models

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Received September 12, 1990; final June 26, 1991

We calculate a correlation function of a dipole which flips upon contact with one of the defects making generally non-Gaussian diffusions. Other than the memory effect in the fractal random walk model, the non-Gaussian property can be an origin of the stretched-exponential law of the correlation function.

KEY WORDS: Streched-exponential; relaxation; defect diffusion.

1. INTRODUCTION

Relaxation phenomena in diverse condensed-matter systems have a common feature.[1,2] Correlation functions obey a stretched-exponential law

$$\phi(t) = \exp[-(t/t_0)^\beta], \qquad 0 < \beta < 1 \tag{1.1}$$

for sufficiently large t.

One of the basic pictures (for others, see refs. 1–3) is that relaxation of a certain substance is triggered by contacts with other migrating substances. In the simplest system composed of a fixed dipole and many noninteracting defects making simple random walks, first introduced by Glarum[4] and later elaborated by Bordewijk,[5] the dipole makes an instantaneous flip upon contact with one of the defects. This model, hereafter referred as the defect diffusion (DD) model, provides a prototype of not only relaxation phenomena, but also the kinetics of diffusion-limited

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0022-4715/92/0100-0563$06.50/0

chemical reactions.[6] Furthermore, the model has many possibilities of extension (see refs. 7 and 8 for some of these[4]).

In n-dimensional space the DD model has[9]

$$S(t) \sim \begin{cases} \sqrt{t} & n=1 \\ t/\log t & n=2 \\ t & n=3 \end{cases} \tag{1.2}$$

as exponential parts in (1.1).

A class, called fractal random walk models,[10,11] was shown by Shlesinger and Montroll[12],[5] to cover the stretched-exponential law (1.1). Assuming that the distribution of the waiting time σ of the random walkers ($=$defects) obeys a power law

$$P(\sigma > t) \sim t^{-\alpha}, \qquad 0 < \alpha < 1 \tag{1.3}$$

instead of an exponential law of the simple random walk, they obtained

$$S(t) \sim \begin{cases} t^{\alpha/2} & n=1 \\ t^{\alpha} & n=3 \end{cases} \tag{1.4}$$

Bendler and Shlesinger[15] further discussed the origin of (1.3) as well as the temperature dependence of the constant t_0 in (1.1).

From a mathematical point of view, the fractal random walk models take into account non-Markovian effects. In view of (1.2), on the other hand, the DD model, not directly included in the fractal random walk models, partly covers the stretched-exponential law. In this paper we will present a class of DD models with stretched-exponential behavior, which is still Markovian but not necessarily Gaussian.

The present work is motivated by recent studies on the origin of the long-time tail in stochastic processes. The well-known Alder–Wainright effect of random motion of a hard sphere in viscous fluid, where the velocity correlation function of the sphere decays as $t^{-3/2}$, is a direct consequence of the non-Markovian effect in the so-called Stokes–Boussinesq–Langevin equation (see ref. 16 for review). Okabe[17] clarified this structure generally in his study of the KMO-Langevin equation (see ref. 18 for further development). On the other hand, a non-Gaussian property likewise gives rise to the long-time-tail behavior. In statistical physics many examples have been found exhibiting power-law decays of moment

[4] In particular, see ref. 7 for results when a multiplicity of relaxing substances is taken into account; introduction of the dynamics during contacts was done in ref. 8.

[5] See also similar studies on electron scavenging in refs. 13 and 14.

functions,[19] which has inspired a general treatment in the framework of diffusion processes by Minami *et al.*,[20] Ogura and Tomisaki,[21] and Tomisaki.[22]

This paper is constituted as follows. We begin Section 2 with a continuous version of a general DD model, and show that the problem is reduced to an evaluation of certain integral involving one-dimensional diffusion processes. We then sketch an asymptotics of the integral which leads to the stretched exponential decay laws. Section 3 is devoted to a proof of the asymptotics. Examples are given in Section 4. Bearing in mind an application to diffusion phenomena in the presence of dislocations and disclinations,[6] we discuss a simplest example of a DD model in Riemannian space. Section 5 is a summary, with a mention of a relation to other power laws in one-dimensional diffusion processes, decay forms of moment functions,[20–22] and size distributions of fractured area.[26]

2. DEFECT DIFFUSION MODELS

We study a continuous version of the Defect Diffusion (DD) model, workinig on n-dimensional Euclidean space instead of an n-dimensional lattice. Consider a system of a frozen-in dipole at the origin and N defects which, following a diffusion law, move independently outside of a ball $U_0 = \{\mathbf{x} \in R^n\colon\ |\mathbf{x}| \leqslant r_0\}$. Suppose that the dipole relaxes from the initial value $M(0)$ to 0 when one of the defects hits U_0 for the first time. Then the correlation function $\phi(t) = \langle M(t)\, M(0)\rangle / M(0)^2$ is given by the probability that none of the N defects hits U_0 up to time t,

$$\phi(t) = P(\tau_i > t,\ i = 1, 2,..., N) \tag{2.1}$$

Here τ_i is the time that the ith defect hits U_0 for the first time (put ∞ if it never hits). By the assumption of mutual independence of the N defects

$$\phi(t) = \prod_{i=1}^{N} P(\tau_i > t)$$

Let V be a domain containing U_0. Assuming that the initial distribution of the defects is uniform over the region $V \backslash U_0$ $(= \{x \in V\colon\ x \notin U_0\})$, we can transform $\phi(t)$ as

$$\begin{aligned}\phi(t) &= \prod_{i=1}^{N} \int_{V\backslash U_0} P(\tau_i > t \mid \mathbf{X}_i(0) = \mathbf{x})\, P(\mathbf{X}_i(0) \in d\mathbf{x})\\ &= \prod_{i=1}^{N} \frac{1}{|V\backslash U_0|} \int_{V\backslash U_0} d\mathbf{x}\, P(\tau_i > t \mid \mathbf{X}_i(0) = \mathbf{x})\end{aligned} \tag{2.2}$$

[6] Closely related quantum mechanical problems were discussed in refs. 23–25.

where $|V\backslash U_0|$ is the volume of $V\backslash U_0$, and $\mathbf{X}_i(t)$ is the position of the ith defect at time t. Since all τ_i have the same distribution,

$$\phi(t)=\left[1-\frac{1}{|V\backslash U_0|}\int_{V\backslash U_0} d\mathbf{x}\, P(\tau_1\leqslant t\,|\,\mathbf{X}_1(0)=\mathbf{x}\right]^N$$

In the thermodynamic limit $V\nearrow R^n$, $N/|V|=\rho$ (fixed), the above integral converges to

$$\exp\left[-\rho\int_{R^n\backslash U_0} d\mathbf{x}\, P(\tau_1\leqslant t\,|\,\mathbf{X}_1(0)=\mathbf{x})\right]$$

which is further transformed to

$$\exp\left[-\frac{\rho\pi^{n/2}}{\Gamma(n/2+1)}\int_{r_0}^{\infty} d|\mathbf{x}|^n\, P(\tau_1\leqslant t\,|\,\mathbf{X}_1(0)=\mathbf{x})\right]$$

under the assumption of spherical symmetry that $P(\tau_1\leqslant t\,|\,\mathbf{X}_1(0)=\mathbf{x})$ depends solely on $|\mathbf{x}|$. Let ξ be a diffusion process on $[0, \infty)$ defined by

$$\xi(t)=|\mathbf{X}_1(t)|-r_0 \tag{2.3}$$

and let τ denote its first hitting time of the origin. Then the correlation function is finally written as

$$\phi(t)=\exp\left[-\frac{\rho\pi^{n/2}}{\Gamma(n/2+1)}\int_0^{\infty} d(x+r_0)^n\, P_x(\tau\leqslant t)\right], \tag{2.4}$$

so that our problem is to evaluate the integral

$$\int_0^{\infty} d(x+r_0)^n\;\; P_x(\tau\leqslant t) \tag{2.5}$$

Here $P_x(\tau\leqslant t)$ stands for $P(\tau\leqslant t\,|\,\xi(0)=x)$.

From a viewpoint of applications in physics, ξ may be assumed to be determined by Itô type stochastic differential equations

$$d\xi(t)=b(\xi(t))\,dt+a(\xi(t))\,dw(t) \tag{2.6}$$

with suitable functions $a(x)$ and $b(x)$ and a Brownian motion $w(t)$. Probability density $p(t, x)$ that $\xi(t)$ is found around x, i.e., $p(t, x)=P(\xi(t)\in dx)/dx$ follows a Fokker-Planck equation

$$\frac{\partial}{\partial t}\,p(t, x)=\frac{\partial}{\partial x}\,(-b(x)\,p(t, x))+\frac{1}{2}\frac{\partial^2}{\partial x^2}\,(a(x)^2\,p(t, x)) \tag{2.7}$$

(See, for example, ref. 34 for basics of stochastic calculus.) When $X_1(t)$ is an n-dimensional Brownian motion, $\xi(t)$ becomes the so-called Bessel process (shifted by $-r_0$ as defined in (2.3)), and $a(x)=1$ and $b(x)=\frac{1}{2}(n-1)/(x-r_0)$.

For the sake of mathematical convenience, we introduce two functions $s(x)$ and $m(x)$ which are defined by

$$s(x)=\int^x du \exp[-F(u)], \qquad m(x)=\int^x du 2a(u)^{-2} \exp[F(u)] \tag{2.8}$$

where

$$F(u)=\int^u 2b(v)\, a(v)^{-2}\, dv \tag{2.9}$$

With these functions Kolmogorov's backward operator $(a^2/2)d^2/dx^2+bd/dx$ is expressed by $d/dmd/ds$. In other words, the process ξ is completely characterized by the functions s and m. The function $s(x)$ and the measure $dm(x)$ are called the canonical scale and the speed measure, respectively.

For one-dimensional Brownian motion $[b(x)=0,\ a(x)=1]$, $s(x)=x$ and $m(x)=2x$. So we would like to know the asymptotics of (2.5) when s and m are generally nonlinear functions. Roughly speaking, if $s(x)$ and $m(x)$ grow in a power order

$$s(x)\sim x^{\lambda}, \qquad m(x)\sim x^{\mu} \qquad (\lambda>0,\ \mu>0) \tag{2.10}$$

as $x\to\infty$, then

$$\text{the integral (2.5)} \sim t^{n/(\lambda+\mu)} \tag{2.11}$$

as $t\to\infty$, i.e., we obtain the stretched exponential decay law with an exponent $\beta=n/(\lambda+\mu)$. We will give a proof in the next section.

Let us see intuitively what is meant by the condition (2.10). $m'(x)$ agrees with a formal stationary solution $p_{st}(x)$ of the Fokker–Planck equation (2.7). The condition $m(x)\sim x^{\mu}$ means that $p_{st}(x)$ is unnormalizable. So the defects, though initially uniformly distributed, have a tendency to diffuse to infinity. On the other hand, the condition $s(x)\sim x^{\lambda}$ means that the defects are recurrent (if they are reflected upon reaching the ball U_0), that is, the defects move about in a sufficiently dense manner.

The exponent in (2.11) can become greater than 1. So, let us next see how this is possible. It is known that the process $\xi(t)$ is given by modifying a Brownian motion $B(t)$ as

$$\xi(t)=s(B(\psi(t)))+\xi(0) \tag{2.12}$$

where $\psi(t)$ is a random function depending on $\{B(s)\}_{0 \leqslant s \leqslant t}$ through s and m. This function ψ effectively changes diffusion constants from 1 $[=\langle B(t)^2 \rangle / t]$ to $\langle \psi(t)^2 \rangle / t$, which, together with the scale $s(x)$, can accelerate the relaxation process. See Example 2 in Section 4 for further discussion on this point.

3. ASYMPTOTICS OF THE INTEGRAL (2.5)

Let ξ be a conservative diffusion process on $[0, \infty)$ having a scale s and a speed measure dm with $\operatorname{supp}[dm] = (0, \infty)$. We assume that the origin is a regular boundary, so that we can put $s(0) = m(0) = 0$ without loss of generality, and that ∞ is a natural boundary. See ref. 26 for an expression of the boundary condition in terms of s and m. We further assume that s and m have asymptotic forms

$$\begin{aligned} s(x) &\sim x^{\lambda} K(x) \\ m(x) &\sim x^{\mu} L(x) \end{aligned} \tag{3.1}$$

as $x \to \infty$. Here λ, μ are positive constants, and functions K and L are slowly varying at ∞, i.e.,

$$\lim_{x \to \infty} K(cx)/K(x) = \lim_{x \to \infty} L(cx)/L(x) = 1 \tag{3.2}$$

for any $c > 0$.

Under the above assumptions for ξ we have the following theorem.

Theorem.

$$\text{Integral (2.5)} \sim C_{\lambda, \mu, n} [s^{-1}(k(t))]^n \qquad \text{as} \quad t \to \infty \tag{3.3}$$

Here k is the inverse of $x \mapsto x m(s^{-1}(x))$, i.e., $k(t) m(s^{-1}(k(t))) = t$, and

$$C_{\lambda, \mu, n} = \left(\frac{(\lambda + \mu)^2}{\lambda \mu} \right)^{n/(\lambda + \mu)} \frac{\Gamma((\lambda + n)/(\lambda + \mu))}{\Gamma(\lambda/(\lambda + \mu))} \tag{3.4}$$

Remark 1. Combining the theorem with (2.4), we see that the logarithm of the correlation function ϕ asymptotically satisfies

$$\log \phi(t) \sim -\frac{\rho \pi^{n/2} C_{\lambda, \mu, n}}{\Gamma(n/2 + 1)} [s^{-1}(k(t))]^n$$

Remark 2. In (3.3),

$$[s^{-1}(k(t))]^n \sim t^{n/(\lambda + \mu)} L_1(t)$$

with a certain function L_1 slowly varying at ∞. When K and L are constants, so is L_1, which means the stretched-exponential law for ϕ.

Proof of the Theorem. Consider a differential equation

$$\frac{d}{dm}\frac{d^+}{ds}g(x;\alpha)=\alpha g(x;\alpha),\qquad \alpha>0 \tag{3.5}$$

on $[0,\infty)$. Here d^+/ds is one-sided scale derivative defined by

$$\frac{d^+}{ds}f(x)=\lim_{h\downarrow 0}\frac{f(x+h)-f(x)}{s(x+h)-s(x)}$$

Equation (3.5) has a unique solution g_2 which is positive and decreasing as well as satisfying the initial condition $g_2(0;\alpha)=1$.[27] On the one hand, g_2 is represented as [ref. 28, Section 4.6, p. 129, Eq. (3b)]

$$g_2(x;\alpha)=E_x[\exp(-\alpha\tau)] \tag{3.6}$$

by using the probabilistic quantity τ, the time that ξ hits the origin for the first time. On the other hand, it is represented by a Laplace transform of a nonnegative quantity q_0 as [ref. 20, (3.20)]

$$g_2(x;\alpha)=\int_0^\infty e^{-\alpha t}q_0(t,s(x))\,dt \tag{3.7}$$

Here q_0 is given as a limit

$$q_0(t,y)=\lim_{x\downarrow 0}\partial p(t,x,y)/\partial x \tag{3.8}$$

where $p(t,x,y)$ is a fundamental solution of the operator $\partial/\partial t-d/d\hat{m}\,d^+/dx$ with respect to the measure $d\hat{m}$ defined by

$$\hat{m}(x)=m(s^{-1}(x)) \tag{3.9}$$

From (3.6) and (3.7) we have $dP_x(\tau\leqslant t)=q_0(t,s(x))\,dt$, so that

$$\text{integral (2.5)}=\int_0^\infty d(x+r_0)^n\int_0^t q_0(u,s(x))\,du \tag{3.10}$$

Next we take a constant $c>0$ and make a transformation of the measure $\hat{m}\to\hat{m}^{(c)}$

$$\hat{m}^{(c)}(x)=\hat{m}(cx)/\hat{m}(c) \tag{3.11}$$

By $q_0^{(c)}(t, y)$ we denote $q_0(t, y)$ when using $\hat{m}^{(c)}$ instead of $\hat{m}$. Then we have

$$q_0(t, x) = q_0^{(c)}\left(\frac{t}{c\hat{m}(c)}, \frac{x}{c}\right) \Big/ c\hat{m}(c) \tag{3.12}$$

Using these relations, we can rewrite the right-hand side of (3.10) ($\equiv I$) as

$$I = \frac{1}{c\hat{m}(c)} \int_0^\infty d(x+r_0)^n \int_0^t q_0^{(c)}\left(\frac{u}{c\hat{m}(c)}, \frac{s(x)}{c}\right) du$$
$$= \int_0^\infty d(s^{-1}(cx)+r_0)^n \int_0^{t/c\hat{m}(c)} q_0^{(c)}(u, x)\, du$$

We choose the constant c so that $t = \hat{m}(c)c$, i.e., $c = k(t)$, and get

$$I = [s^{-1}(c)]^n \int_0^\infty d\left(\frac{s^{-1}(cx)+r_0}{s^{-1}(c)}\right)^n \int_0^1 q_0^{(c)}(u, x)\, du$$

Since m is monotonically increasing, $t \to \infty$ implies $c \to \infty$, so that we only have to study the asymptotics of I as $c \to \infty$. As will be proved later,

$$\lim_{c\to\infty} I/[s^{-1}(c)]^n = \int_0^\infty dx^{n/\lambda} \int_0^1 q_0^*(u, x)\, du \tag{3.13}$$

Here q_0^* denotes q_0 when we put $\hat{m}(x) = x^{\mu/\lambda}$ [$= \lim_{c\to\infty} \hat{m}^{(c)}(x)$], and is explicitly written as

$$q_0^*(u, x) = \left(\frac{\gamma}{(\gamma+1)^2}\right)^{1/(\gamma+1)} \frac{x}{\Gamma(1/(\gamma+1))u^{1+1/(\gamma+1)}} \exp\left(-\frac{\gamma x^{\gamma+1}}{(\gamma+1)^2 u}\right) \tag{3.14}$$

where $\gamma = \mu/\lambda$. Using (3.13) and (3.14), we carry out the integration to obtain $\lim_{c\to\infty} I/[s^{-1}(c)]^n = C_{\lambda,\mu,n}$, which establishes the relation (3.3)

The relation (3.13) will be proved by using a result by Ogura and Tomisaki (ref. 21, Lemma 4.1) concerning convergence of $q_0^{(c)}$ to q_0^*.

Lemma 1. For $u > 0$, $\eta > 0$, $r \leqslant 1$, we have:

(a) $\lim_{c\to\infty} q_0^{(c)}(t, y)/y = q_0^*(t, y)/y$ uniformly in $(t, y) \in [u, \infty) \times (0, \eta]$

(b) $\lim_{c\to\infty} \exp(-y^{\gamma+3})\, q_0^{(c)}(t, y) = \exp(-y^{\gamma+3})\, q_0^*(t, y)$ uniformly in $(t, y) \in [u, \infty) \times [\eta, \infty)$, where γ is the constant in (3.14)

(c) $\lim_{c \to \infty} \sup_{0 \leqslant y \leqslant \eta} \left| \frac{\int_0^u q_0^{(c)}(t, y)\, dt}{\int_0^u q_0^*(t, y)\, dt} - 1 \right| = 0$

(d) $\sup_{c \geqslant c_0, 0 < t \leqslant u, y \geqslant \eta} t^r y^{1-r} [q_0^{(c)}(t, y) \vee q_0^*(t, y)] < \infty$

for some $c_0 = c_0(u, \eta, r)$

We further need the following simple lemma.

Lemma 2. Let D be an interval in R^1 with endpoints a and b $(a < b)$. Let $F^{(c)}$ $(c > 0)$ be a family of functions which are continuous and increasing in D. We suppose $F^{(c)}$ converges as $c \to \infty$ to a function F in D.

(a) If D is bounded and

$$\sup_{c > 0} \{F^{(c)}(b) - F^{(c)}(a)\} < \infty \tag{3.15}$$

then

$$\lim_{c \to \infty} \int_D f(x)\, dF^{(c)}(x) = \int_D f(x)\, dF(x) \tag{3.16}$$

for any function f which is uniformly continunous and bounded on D.

(b) Suppose there exists an increasing sequence of bounded intervals D_i with endpoints a_i and b_i such that $\bigcup_i D_i = D$,

$$\sup_{c > 0} \{F^{(c)}(b_i) - F^{(c)}(a_i)\} < \infty \tag{3.17}$$

and

$$\lim_{i \to \infty} \sup_{c > 0} \int_{D \setminus D_i} f(x)\, dF^{(c)}(x) = 0 \tag{3.18}$$

for some uniformly continuous and bounded f on D_i. Then the relation (3.16) holds for the same f as in (3.18).

Proof of Lemma 2(a). For any $\varepsilon > 0$, we can find $\delta > 0$ and points $x_1, \ldots, x_m$ in D with $|x_i - x_{i+1}| < \delta$ such that $|f(x) - f(x_i)| < \varepsilon$ if $x_i < x \leqslant x_{i+1}$. Then we have

$$\left| \int_D f(x)\, dF^{(c)}(x) - \sum_i f(x_i) \{F^{(c)}(x_{i+1}) - F^{(c)}(x_i)\} \right| < \varepsilon \sup_c \{F^{(c)}(b) - F^{(c)}(a)\}$$

and a similar inequality for the difference between $\int_D f(x)\, dF(x)$ and its approximate Riemannian sum $\sum_i f(x_i)\{F(x_{i+1}) - F(x_i)\}$. On the other hand, the absolute value of the difference of the two approximate Riemannian sums can be made smaller than ε by taking c sufficiently large.

(b) In an inequality

$$\left|\int_D f(x)\,dF^{(c)}(x) - \int_D f(x)\,dF(x)\right|$$
$$\leqslant \left|\int_{D_i} f\,dF^{(c)} - \int_{D_i} f\,dF\right| + \left|\int_{D\setminus D_i} f\,dF^{(c)}\right| + \left|\int_{D\setminus D_i} f\,dF\right|$$

we can find D_i such that the second and third terms on the right-hand side are smaller than arbitrary ε. With this D_i, the first term can be made smaller than ε by (a) of this lemma if we take c sufficiently large.

Now let us continue the proof of the theorem. We first divide $I/[s^{-1}(c)]^n$ into two parts:

$$\begin{aligned} J_1 &= \int_{0+}^{1} dv^{(c)}(y) \int_0^1 dt\, q_0^{(c)}(t, y) \\ J_2 &= \int_1^{\infty} dv^{(c)}(y) \int_0^1 dt\, q_0^{(c)}(t, y) \end{aligned} \tag{3.19}$$

where

$$v^{(c)}(0, x] = \left(\frac{s^{-1}(cx) + r_0}{s^{-1}(c)}\right)^n - \left(\frac{r_0}{s^{-1}(c)}\right)^n \tag{3.20}$$

Let us begin with an estimation of the integral J_1. Take $\varepsilon > 0$ arbitrarily. By (c) of Lemma 1, there exists $c_1 = c_1(\varepsilon)$ such that if $c > c_1$, then

$$\left|\int_0^1 q_0^{(c)}(t, y)\,dt - \int_0^1 q_0^*(t, y)\,dt\right| < \varepsilon \int_0^1 q_0^*(t, y)\,dt \tag{3.21}$$

holds uniformly in $y \in (0, 1]$.

On the other hand, $v^{(c)}$ is a measure on (0,1], and satisfies the properties $\sup_{c>0} v^{(c)}(0, 1] < \infty$ and $v^{(c)}(0, x] \to v^*(0, x]$ $(\equiv x^{n/\lambda})$ as $c \to \infty$ for $x \in (0, 1]$. Furthermore, $\int_0^1 dt\, q^*(t, y)$ $[= \Gamma(1/(\gamma+1))^{-1}\Gamma(1/(\gamma+1), \gamma y^{\gamma+1}/(\gamma+1)^2)$ if $y > 0]$ is uniformly continuous and bounded on (0, 1]. So by Lemma 2(a) we can find $c_2 = c_2(\varepsilon)$ such that for $c > c_2$

$$\left|\int_{0+}^1 dv^{(c)}(y) \int_0^1 dt\, q_0^*(t, y) - \int_{0+}^1 dv^*(y) \int_0^1 dt\, q_0^*(t, y)\right| < \varepsilon \tag{3.22}$$

From (3.21) and (3.22) we have, for $c > c_1 \vee c_2$,

$$\begin{aligned}
&\left| J_1 - \int_{0+}^{1} dv^*(y) \int_0^1 dt\, q_0^*(t, y) \right| \\
&\quad \leqslant \int_{0+}^{1} dv^{(c)}(y) \left| \int_0^1 q_0^{(c)}\, dt - \int_0^1 q_0^*\, dt \right| \\
&\qquad + \left| \int_{0+}^{1} dv^{(c)}(y) \int_0^1 q_0^*\, dt - \int_{0+}^{1} dv^*(y) \int_0^1 q_0^*\, dt \right| \\
&\quad \leqslant \varepsilon \left[\int_{0+}^{1} dv^{(c)}(y) \int_0^1 q_0^*\, dt + 1 \right] \leqslant M_1 \varepsilon
\end{aligned}$$

with a certain constant M_1 being independent of c, which guarantees

$$\lim_{c \to \infty} J_1 = \int_0^1 dv^*(x) \int_0^1 dt\, q_0^*(t, y) \tag{3.23}$$

Let us proceed to an estimation of J_2. Fix $r < 1 - n/\lambda$ and take $\varepsilon > 0$ arbitrarily. By (b) and (d) of Lemma 1, we can find $c_3 = c_3(\varepsilon, r, t)$ and $M_2 = M_2(r, t)$ such that if $c \geqslant c_3$,

$$q_0^{(c)}(t, y) \leqslant [q_0^*(t, y) + \varepsilon \exp(y^{\gamma+3})] \wedge M_2 t^{-r} y^{r-1}$$

holds for $y \geqslant 1$, so that the integral $J_3 = \int_1^\infty q_0^{(c)}(t, y)\, dv^{(c)}(y)$ satisfies

$$\varlimsup_{c \to \infty} J_3 \leqslant \varlimsup_{c \to \infty} \int_1^\infty [q_0^*(t, y) + \varepsilon \exp(y^{\gamma+3})] \wedge M_2 t^{-r} y^{r-1}\, dv^{(c)}(y) \tag{3.24}$$

On the right-hand side, it is not difficult to check the conditions of Lemma 2(b), where $D = [1, \infty)$, $D_i = [1, i)$, and $F^{(c)}(x) = v^{(c)}(1, x]$. The condition (3.17) is satisfied since

$$\lim_{c \to \infty} v^{(c)}(1, x] = x^{n/\lambda} - 1 \tag{3.25}$$

By integration by parts, we have

$$\int_i^\infty y^{r-1}\, dv^{(c)}(y) = -i^{r-1} v^{(c)}(1, i] - (r-1) \int_i^\infty y^{r-2} v^{(c)}(1, y]\, dy$$

Both terms in the right-hand side tend to 0 uniformly in c as $i \to \infty$ because of (3.20) and the inequality $r < 1 - n/\lambda$, which shows the condition (3.18) is satisfied.

Now we can apply Lemma 2(b) to (3.24), and see that the right-hand side of (3.24) becomes

$$\int_1^\infty [q_0^*(t, y) + \varepsilon \exp(y^{\gamma+3})] \wedge M_2 t^{-r} y^{r-1}\, dv^*(y)$$

where $v^*(0, x] = x^{n/\lambda}$ as before. Since $\int_1^\infty y^{r-1}\, dv^*(y) < \infty$, we have in the limit of $\varepsilon \downarrow 0$,

$$\begin{aligned}\overline{\lim_{c\to\infty}}\, J_3 &\leqslant \int_1^\infty q_0^*(t, y) \wedge M_2 t^{-r} y^{r-1}\, dv^*(y)\\ &\leqslant \int_1^\infty q_0^*(t, y)\, dv^*(y) \end{aligned} \tag{3.26}$$

by the dominated convergence theorem.

On the other hand, by (a) of Lemma 1, for arbitrarily fixed $\eta > 1$, there exists $c_4 = c_4(\varepsilon, \eta, t)$ such that if $c > c_4$,

$$q_0^{(c)}(t, y) \geqslant (1-\varepsilon)\, q_0^*(t, y)$$

holds for $y \in [1, \eta]$. So we have by Lemma 2(a)

$$\begin{aligned}\underline{\lim_{c\to\infty}}\, J_3 &\geqslant \underline{\lim_{c\to\infty}} \int_1^\eta (1-\varepsilon)\, q_0^*(t, y)\, dv^{(c)}(y)\\ &= (1-\varepsilon) \int_1^\eta q_0^*(t, y)\, dv^*(y)\end{aligned}$$

Since ε and η are arbitrary, we have

$$\underline{\lim_{c\to\infty}}\, J_3 \geqslant \int_1^\infty q_0^*(t, y)\, dv^*(y) \tag{3.27}$$

which together with (3.26) shows that

$$\lim_{c\to\infty} J_3 = \int_1^\infty q_0^*(t, y)\, dv^*(y) \tag{3.28}$$

We fix $r < 1 - n/\lambda$ and apply (d) of Lemma 1. We can then find $c_5 = c_5(r)$ and $M_3 = M_3(r)$ such that

$$\int_1^\infty q_0^{(c)}(t, y)\, dv^{(c)}(y) \leqslant M_3 \int_1^\infty t^{-r} y^{r-1}\, dv^{(c)}(y)$$

for $t \in (0, 1]$. The last expression is dominated by $M_4(r)t^{-r}$, which is integrable in t on $(0, 1]$. Hence, using the dominated convergence theorem and (3.28), we have

$$\lim_{c \to \infty} J_2 = \lim_{c \to \infty} \int_0^1 dt\, J_3$$

$$= \int_0^1 dt \int_1^\infty q_0^*(t, y)\, dv^*(y) \tag{3.29}$$

The relations (3.23) and (3.29) complete the proof of the theorem.

4. EXAMPLES

Examples are named after the radial process of the particle 1, i.e., $R(t) = |\mathbf{X}_1(t)|$, and specified by the corresponding backward operators $\tilde{A} = d/d\tilde{m}\, d^+/d\tilde{s}$. When $\tilde{A}$, as is usual, is given formally as

$$\tilde{A} = \tilde{b}(x) \frac{d}{dx} + \frac{1}{2} \tilde{a}(x)^2 \frac{d^2}{dx^2} \tag{4.1}$$

$\tilde{s}$ and $\tilde{m}$ are expressed by

$$\tilde{s}(x) = \int^x du \exp[-\tilde{F}(u)], \qquad \tilde{m}(x) = \int^x du\, 2\tilde{a}(u)^{-2} \exp[\tilde{F}(u)] \tag{4.2}$$

where

$$\tilde{F}(u) = \int^u 2\tilde{b}(v)\, \tilde{a}(v)^{-2}\, dv \tag{4.3}$$

Lower bounds of the above integrals are suitably chosen. A change of them only gives rise to a linear transformation of $\tilde{s}$ and $\tilde{m}$ as $\tilde{s} \to C_1 \tilde{s} + C_2$, $\tilde{m} \to C_1^{-1} \tilde{m} + C_3$, which gives the same $\tilde{A}$. Since ξ is related to $R(t)$ $[= |\mathbf{X}_1(t)|]$ by (2.3), the scale s and the speed measure m of ξ [with $s(0) = m(0) = 0$] are given by $s(x) = \tilde{s}(x + a) - \tilde{s}(a)$, $m(x) = \tilde{m}(x + a) - \tilde{m}(a)$.

Example 1 (n-Dimensional Bessel process). This is the radius of an n-dimensional Brownian motion $(B_1(t), \dots, B_n(t))$; $R(t) = [\sum_{i=1}^n B_i(t)^2]^{1/2}$. As is well known, $\tilde{a}(x) = 1$, $\tilde{b}(x) = (n-1)/2x$. When $n = 1$

$$\tilde{s}(x) = x, \qquad \tilde{m}(x) = 2x \tag{4.4}$$

so that putting $n=1$, $\lambda=1$, $K(x)\equiv 1$, $L(x)\equiv 2$ in the theorem, we have

$$\phi(t)=\exp[-2(2/\pi)^{1/2}\rho t^{1/2}] \tag{4.5}$$

the stretched-exponential law (1.1) with $\beta=1/2$.

When $n\geqslant 2$,

$$\begin{aligned} &\tilde{s}(x)=\log x, && \tilde{m}(x)=x^2-1, && n=2 \\ &\tilde{s}(x)=(x^{2-n}-1)/(2-n), && \tilde{m}(x)=2(x^n-1)/n, && n\geqslant 3 \end{aligned} \tag{4.6}$$

These cases are not covered by the theorem and require more elaborate treatment. We will discuss it in a separate paper, where it is shown that

$$\text{Integral } (2.5)\sim\begin{cases} 2t/\log t & n=2 \\ [n(n-2)/2a^{2-n}]\,t & n\geqslant 3 \end{cases} \tag{4.7}$$

agreeing with expression (1.2) of the discrete DD model.

Example 2 (Self-similar diffusion process). The radial process $R(t)$ is said to be self-similar with parameter $H>0$ if $R(ct)$ is equivalent to $c^H R(t)$ for any $c>0$. It is characterized by[26]

$$\tilde{b}(x)=c_1 x^{1-1/H}, \qquad \tilde{a}(x)=c_2 x^{1-1/(2H)} \qquad (c_1>0,\ c_2>0) \tag{4.8}$$

or, by an application of (4.2) and (4.3),

$$\tilde{s}(x)=(x^\lambda-1)/\lambda, \qquad \tilde{m}(x)=2(x^\mu-1)/\mu c_2^2 \tag{4.9}$$

Here the parameters λ and μ are given by

$$\lambda=1-2c_1/c_2^2, \qquad \mu=-\lambda+1/H \tag{4.10}$$

We can apply the theorem as long as λ and μ are positive, i.e.,

$$1-1/H<2c_1/c_2^2<1 \tag{4.11}$$

and get the stretched-exponential law with an exponent nH.

The exponent nH can become greater than 1. This seemingly contradicts (4.7) if we recall that the Bessel processes are self-similar with $H=1/2$, $c_1=(n-1)/2$, and $c_2=1$. But when $n\geqslant 2$ the inequality (4.11) is violated. In other words, the restriction (4.11) implicitly affects the asymptotics of (2.5).

It will be instructive to see how we get decay laws faster than exponential functions. It is known that one-dimensional diffusion processes are obtained by making nonlinear transformations and time changes to a Brownian motion B; $\xi(t)$ is expressed as

$$\xi(t) = s(B(\psi(t)) + \xi(0) \tag{4.12}$$

where $\psi(t)$ is a random function whose inverse is given by

$$\psi^{-1}(t) = \int_0^t ds\, \hat{m}'(B(s) + \xi(0)) \tag{4.13}$$

with $\hat{m}(x) = m(s^{-1}(x))$.[32] Let us consider a defect initially $\xi(0) = \sqrt{t}$ which will hit the dipole around time t if ξ is simply a Brownian motion. Using (4.9), we have

$$\psi^{-1}(t) = c_3 \int_0^t ds\, [\sqrt{t} + B(s) + 1/\lambda]^{\mu/\lambda - 1}$$

On the average we may set $B(s) \simeq -\sqrt{s}$, so that

$$\begin{aligned}\psi^{-1}(t) &\simeq c_3 \int_0^t ds\, (\sqrt{t} - \sqrt{s})^{\mu/\lambda - 1} \\ &= c_4 t^{1/(2\lambda H)}\end{aligned}$$

Here we have used $\lambda + \mu = 1/H$. Hence we have from (4.12)

$$\xi(t) \simeq s(B(c_5 t^{2\lambda H})) + \xi(0) \tag{4.14}$$

This approximate expression shows that the clock is modified from t to $t^{2\lambda H}$, and the space is scaled from x to x^{λ}; correspondingly, the time dependence of the relaxation function is changed as $\sqrt{t} \to (\sqrt{t})^{2\lambda H} = t^{\lambda H} \to t^{\lambda H/\lambda} = t^H$. This agrees with the index of the stretched-exponential decay law with $n = 1$.

The function $\psi(t)$ effectively changes diffusion constants from 1 $[= \langle B(t)^2 \rangle / t]$ to $\langle \psi(t)^2 \rangle / t$. Another mechanism has been considered in the continuous-time random walk model with a coupled spatial-temporal memory.[33] Suppose, for example, that the probability density of a jump occurring at time t behaves as $t^{-1-\alpha}$ $(0 < \alpha < 1)$; and that the conditional probability density of the jump going distance l behaves as $(2\pi t^{\beta})^{-1/2} \exp(-l^2/2t^{\beta})$ $(\beta > 1)$. Then the average of the square of the distance behaves like t^{β}. In the limit of continuous space these examples will converge to non-Markovian processes whose sample paths include jumps. The processes discussed in the present paper are Markovian and their sample paths are continuous. The change of the diffusion constants is caused by nonlinearity of the functions m and s.

The term $d(x+a)^n$ in (2.5) formally survives to give an $n/(\lambda+\mu)$ dependence on the dimension number n. The proof of the Theorem in Section 3 shows that the survival is guaranteed by the assumption $\lambda > 0$, $\mu > 0$. The first one means $\xi(t)$ is recurrent (when the defects are reflected upon hitting the dipole), that is, it reaches neighborhoods of any point within finite time. The process does not have a stationary probability distribution function, because a formal stationary density function $m'(x)$ of the Fokker–Planck equation is unnormalizable. Both conditions mean that the defects move densely around regions far away from the origin.

The Brownian motion in three dimension lacks the recurrence property. The two-dimensional case is critical; recurrent though $\lambda = 0$, and has no stationary distribution. The term $1/\log t$ in (4.7) may be regarded as a correction.

Example 3 (General case). Let us rewrite (4.1) as

$$\tilde{A} = \frac{\tilde{a}^2(x)}{2} e^{-\tilde{F}(x)} \frac{d}{dx} e^{\tilde{F}(x)} \frac{d}{dx} \tag{4.15}$$

where $\tilde{F}(x)$ is defined by (4.3). If $\tilde{a}(x) \sim x^{-\rho_1}$, $e^{-\tilde{F}(x)} \sim x^{\rho_2}$ as $x \to \infty$, then by (4.2)

$$s(x) \sim \frac{1}{\rho_2+1} x^{\rho_2+1}, \qquad m(x) \sim \frac{2}{2\rho_1 - \rho_2 + 1} x^{2\rho_1 - \rho_2 + 1} \qquad \text{as} \quad x \to \infty \tag{4.16}$$

as far as $\rho_2 + 1 > 0$ and $2\rho_1 - \rho_2 + 1 > 0$. Hence the exponent β in (1.1) $[= n/(\lambda+\mu)$ by Remark 2 of the theorem$]$ becomes $n/(2\rho_1 + 2)$, being independent of ρ_2.

Example 4 (Riemannian space). A modification of the discussion in Sections 2 and 3 gives us a DD model in Riemannian space. We sketch it using a simple example. Let ψ be a nonnegative C^∞ function on $[0, \infty)$ satisfying $\psi(0) = 0$, $\psi'(r) > 0$ $(r > 0)$, and

$$\psi(r) = r^\nu, \qquad r \geqslant 1 \tag{4.17}$$

In $(n+1)$ $(\geqslant 3)$-dimensional Euclidean space E^{n+1} we consider a surface M^n

$$x^{n+1} = \psi(r) \tag{4.18}$$

where

$$r = \left(\sum_{i=1}^{n} (x^i)^2 \right)^{1/2} \tag{4.19}$$

which is generated by rotating the curve $x^{n+1} = \psi(x^1)$, $x^2 = x^3 = \cdots = x^n = 0$ around the x^{n+1} axis. We suppose M^n is endowed with a metric induced by the Euclidean metric of E^{n+1}. Using the polar coordinate $\theta^1, \theta^2, ..., \theta^{n-1}, \theta^n = r$ of $x^1, x^2, ..., x^n$, the metric is written as

$$\sum_{i,j=1}^{n} g_{ij}\, d\theta^i\, d\theta^j = [1 + \psi'(r)^2]\, dr^2 + r^2\, d\Theta^2 \tag{4.20}$$

Here $d\Theta^2$ is the canonical metric on the unit sphere S^{n-1}. The expression (4.20) is readily derived by substituting (4.18) into the relation $\sum_{i,j=1}^{n} g_{ij}\, d\theta^i\, d\theta^j = \sum_{i=1}^{n+1} (dx^i)^2$ and using $\sum_{i=1}^{n} (dx^i)^2 = dr^2 + r^2 d\Theta^2$. When the metric is given by (4.20), the backward operator $\tilde{A}$ of the radial process of the Brownian motion on M^n is given by the radial part of $\frac{1}{2}\Delta$, i.e.,

$$\tilde{A} = \frac{1}{2[1 + \psi'(r)^2]^{1/2}\, r^{n-1}} \frac{d}{dr} \frac{r^{n-1}}{[1 + \psi'(r)^2]^{1/2}} \frac{d}{dr} \tag{4.21}$$

In view of (4.2) and (4.3), the canonical scale s and the speed measure dm are given by

$$s(r) = \int^r [1 + \psi'(u)^2]^{1/2}\, u^{1-n}\, du \tag{4.22}$$

$$m(r) = 2 \int^r [1 + \psi'(u)^2]^{1/2}\, u^{n-1}\, du \tag{4.23}$$

so that by (4.17)

$$s(r) \sim \frac{\nu}{\nu - n + 1} r^{\nu - n + 1}, \qquad m(r) \sim \frac{2\nu}{\nu + n - 1} r^{\nu + n - 1} \qquad \text{as} \quad r \to \infty \tag{4.24}$$

if $\nu > n - 1$.

A modification is required in the expression of the correlation ϕ; the volume element $d\mathbf{x}$ in (2.2) is changed by the invariant volume element $[\det(g_{ij})]^{1/2}\, dr\, d\theta^1 \cdots d\theta^{n-1}$; accordingly, $d(x + r_0)^n$ by $(n/2)\, dm(r + r_0)$ in (2.4) and (2.5), which now are denoted by (2.4′) and (2.5′).

An argument similar to the proof of the theorem shows that integral $(2.5') \sim m(s^{-1}(k(t)))$ as $t \to \infty$ up to a constant, so that from (4.24)

$$\text{integral } (2.5') \sim \text{const} \cdot t^{\beta}, \qquad \nu > n - 1$$

where the exponent β is given by

$$\beta = \frac{n + \nu - 1}{2\nu}$$

being dependent on ν.

Before closing the discussion of this example, we show how the parameter ν is related to the radial curvature $\mathscr{K}(r)$. By an introduction of the variable $\tilde{r} = F(r) \equiv \int_0^r [1 + \psi'(u)^2]^{1/2}\, du$, the metric (4.20) is written in a standard form $d\tilde{r}^2 + f(\tilde{r})^2\, d\Theta^2$ [ref. 29, (2.16)], where $f(r) = [F^{-1}(r)]^2$. By (2.18) of ref. 29,

$$\begin{aligned}\mathscr{K}(r) &= -f''(r)/f(r)\\ &= \psi'(r)\,\psi''(r)/r[1 + \psi'(r)^2]^2\\ &\sim (\nu - 1)/\nu^2 r^{2\nu}, \qquad \nu > 1\end{aligned}$$

In the case that the metric depends also on $\theta^1,\dots,\theta^{n-1}$, it will be possible to find lower and upper bounds in the asymptotics of integral (2.5′) by using the comparison theorems obtained in refs. 29 and 30.

5. DISCUSSION

As briefly mentioned in the introduction, several power laws have been found in different statistical mechanical situations involving general diffusion processes. We will review them in connection with the asymptotics of the canonical scale s and the speed measure dm.

The theorem in Section 3 shows that the asymptotic from of the relaxation function as $t \to \infty$ is determined by those of $s(x)$ and $m(x)$ as $x \to \infty$. This intuitively means the relaxation of the dipole after a long lapse of time is mainly triggered by defects at a long distance from the dipole. In this way is determined the asymptotic behavior of expectation values of moments as $t \to \infty$ for diffusion processes ξ discussed in refs. 19 and 20. With the assumption (3.1), we have for $f \in L_1(dm)$

$$E[f(\xi)] \sim t^{-\mu/(\lambda+\mu)} K_p(t), \qquad p \geqslant 1 \tag{5.1}$$

if ξ is recurrent.[20] Here $K_p(t)$ is a function slowly varying at ∞. See ref. 22 for transient cases.

On the other hand, in a problem of fracture, our concern is a size distribution of small fractured areas, which is determined by the asymptotics of $s(x)$ and $m(x)$ as $x \to 0$. In a simple model[26] it is assumed that the stress field is expressed by a one-dimensional diffusion process ξ and that a fractured area is identified with a maximal open interval on which $\xi(x) > 0$. If

$$s(x) \sim x^\lambda \qquad \text{and} \qquad m(x) \sim x^\mu \qquad \text{as} \quad x \to 0$$

then the cumulative number $N(l)$ of fractured areas whose length is greater than l behaves as

$$N(l) \sim \text{const} \cdot l^{-\lambda/(\lambda+\mu)} \qquad \text{as} \quad l \downarrow 0 \tag{5.2}$$

Here we can give a remark on self-similar cases (4.9). It is often assumed in the fractal theory that the index H has sufficient "physical" information. But the examples above show that this is not necessarily true. In fact, the asymptotics of neither the moment (5.1) nor the size distribution (5.2) can be expressed by H alone. We find another counterexample in the stretched-exponential decay law. Among processes with index $H = 1/2$, those with $\lambda > 0$ have a different exponent from those with $\lambda \leqslant 0$ (see Example 2 in Section 4).

Finally, the above analyses are closely related to those for the asymptotics at $t \to 0$ or ∞ of the transition probability density $p(t, x, y)$, a review of which is given by Tomisaki.[31]

ACKNOWLEDGMENTS

Part of this work was done while H.M.I. was at the Department of Mathematics, Rutgers University. He would like to thank Prof. J. L. Lebowitz for the hospitality extended to him. Y.O. was supported by the Air Force Office of Scientific Research Grant F4962085C0144.

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