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Anomalous diffusion and phase relaxation

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The diffusion and relaxation of a phase are investigated on the basis of several stochastic models. A simple relation between the diffusional behavior of the extended phase and the relaxation of periodic phase observables is found in the case of Gaussian and Lèvy distributed increments. In these cases, an anomalous diffusion gives rise to a stretched exponential relaxation of phase observables. Continuous time random walks may lead, even in the case of normal diffusion, to a slow algebraic relaxation.

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I. INTRODUCTION

Diffusion is an important transport process of matter and energy in various physical, chemical, and biological systems [1,2]. In the seminal works of Smoluchowski, Einstein, and Langevin [3], the macroscopic spread of, say, the mass density of a specific substance diffusing in a background medium was related to the individual stochastic motion of the particles constituting the diffusing substance. Under quite general conditions, the irregular motion of the individual particles leads to a spread of the second moment of the mass distribution that is linear in time. However, striking deviations from linear behavior were observed under strong nonequilibrium conditions, or in disordered systems. For example, according to Richardson's law, the average square separation of a pair of particles passively moving in a turbulent flow grows with the third power of time [4]. Conversely, diffusion in disordered materials may proceed more slowly than linearly in time [5].

Strictly speaking, a diffusional spread of a quantity can only continue indefinitely if the space in which it takes place is infinitely extended. In a finite space, after some initial spread, the density of the diffusing quantity will relax toward a stationary distribution. A phase variable such as the one of a linear or nonlinear oscillator, of a classical wave or a quantum mechanical wave function, is by definition restricted to values ranging between 0 and 2π. In the absence of a phase locking mechanism there is no preferred value of the phase, and the phase may diffuse locally in the same way as an unrestricted variable. At sufficiently long times, however, the finiteness of the available phase space comes into play, and the mean values of phase observables relax to their stationary values. This mechanism determines the line shapes of atoms and molecules [6–8], and the quality of a laser [9], to name but a few examples.

Most theoretical investigations of phase relaxation were based on the assumption that the increments of the phase diffusion are Gaussian distributed. Then a particularly simple relation between the laws describing the spread in the hypothetically unrestricted case and the actual relaxation can be formulated [6]. Kubo also discussed the case of increments that are described by a discrete Markovian process [7]. In the present paper we discuss various classes of normal and anomalous diffusion processes, both Gaussian and non-Gaussian, and for the respective phase relaxations find very different laws ranging from fast Gaussian, exponential, and stretched exponential up to extremely slow algebraic decay.

The paper is organized as follows. In Sec. II we outline the relevant general relations between phase diffusion and phase relaxation in terms of the characteristic function. These relations are applied to processes with independent increments in Sec. III, to self-similar processes in Sec. IV, and to continuous time random walks in Sec. V. The paper closes with a discussion in Sec. VI.

II. DIFFUSION AND PHASE RELAXATION

An ever spreading process is called *normal diffusion* if the variance $\sigma^2(t)=\langle(x(t)-\langle x(t)\rangle)^2\rangle$ grows linearly in the time t, and *anomalous* diffusion if it grows with some power of t that is different from 1. Thus, diffusion is generally characterized by an algebraic spread of the variance in time,

$$\sigma_x^2(t)=D_\beta t^\beta, \tag{2.1}$$

where the exponent $\beta<1$ refers to subdiffusive behavior and $\beta>1$ to superdiffusive behavior and D_β is the (anomalous) diffusion constant [5]. Also, for very broadly distributed processes for which the second centered moments do not exist, diffusion can be defined in an analogous way using absolute centered moments of sufficiently low order p:

$$\langle|(x(t)-\langle x(t)\rangle|^p\rangle=D(p)t^{\beta(p)}, \tag{2.2}$$

where $\beta(p)$ may be a nonlinear function of p. In the latter definition the more special case of Eq. (2.1) is included.

In order to avoid confusion, we note that in mathematics, the notion of a diffusion process has a different meaning. It refers to a continuous Markov process which is driven by Gaussian white noise [10]. Here we do not restrict ourselves to Markovian or continuous processes, nor to processes driven by a Gaussian process. The relevant property we have in mind here refers to the unrestricted algebraic growth of the considered processes, which is characterized by Eqs. (2.1) or (2.2).

As for a prototypical random walk, the anomalous diffusion can be viewed as an accumulation of increments which, however, only in the case of normal diffusion can be independent. For anomalous diffusion, the increments are correlated according to an algebraic law. However, they do not depend on the actual state of the process if all possible states are equivalent, as we assume here.

In many cases, the states of a phase variable φ are equivalent, and the phase itself undergoes a diffusional process on a short time scale. However, by its very definition, a phase is only relevant up to multiples of 2π, and therefore an ever increasing spread of the variance of φ is impossible. Typically, one will instead expect a relaxational behavior of all functions of the phase. The only possible exceptions to this rule are periodic or quasiperiodic motions. Apart from these cases, in the asymptotic state reached for $t\rightarrow\infty$, an unlocked phase will be distributed according to the equipartition on the interval $[0,2\pi)$. However, an *extended* phase x, taking unrestricted real values, is conveniently defined as the sum of the phase increments up to a time t. It contains a winding number counting how often x can be wrapped on a circle with unit radius, additionally to the actual value of the phase, $\varphi = x \mod 2\pi$. The unrestricted phase therefore takes the form

$$x=\varphi+2\pi w, \tag{2.3}$$

where the winding number w is an integer number: $w\in\mathbb{Z}$. The way in which the probability distribution of the extended variable x spreads in time determines the law with which the phase relaxes.

All true phase observables are independent of the winding number, and, as periodic functions, linear combinations of the exponential functions $\exp\{inx(t)\}$, where n may be an arbitrary negative or positive integer: $n\in\mathbb{Z}$. Consequently, the mean values of all (periodic) functions of the phase φ can be expressed as linear combinations of the mean values of the exponential functions:

$$m_n(t)=\langle\exp\{inx(t)\}\rangle \quad \text{with} \quad n\in\mathbb{Z}. \tag{2.4}$$

In what follows, we will refer to $m_n(t)$ as the fundamental mean values of the phase. Obviously, these mean values coincide with the characteristic function

$$\Theta(u,t)=\langle\exp\{iux(t)\}\rangle \tag{2.5}$$

of the extended process $x(t)$ taken at the integer values $u=n\in\mathbb{Z}$:

$$m_n(t)=\Theta(n,t). \tag{2.6}$$

This simple relation is most important for the present paper. It has long been used in the stochastic theory of spectral line shapes [7] and motional narrowing in magnetic resonance and related fields [8]. In most of these cases the extended phase is assumed to be Gaussian.

For convenience, we collect some of the general properties of the characteristic function in Appendix A. Here we only mention the well known relation that gives the variance of $x(t)$ in terms of the first two derivatives of the characteristic function with respect to u at $u=0$:

$$\sigma_x^2(t)=-\frac{\partial^2\Theta(0,t)}{\partial u^2}+\left(\frac{\partial\Theta(0,t)}{\partial u}\right)^2. \tag{2.7}$$

Finally, we express the characteristic function $\Theta_w(u,t)$ of the winding number $w=(x-\varphi)/2\pi$ in terms of the statistics of the extended phase variable. The characteristic function is defined as

$$\Theta_w(u,t)=\sum_{w=-\infty}^{\infty}e^{2\pi iwu}p_w(t), \quad u\in[0,1), \tag{2.8}$$

where $p_w(t)$ denotes the probability that the winding number takes the value $w\in\mathbb{Z}$ at the time t. It can be expressed by the probability density $\rho(x,t)$ of finding the extended process at x at time t:

$$p_w(t)=\int_{2\pi w}^{2\pi(w+1)}dx\ \rho(x,t). \tag{2.9}$$

Using the Poisson sum formula $\Theta_w(u,t)$ can be expressed in terms of the characteristic function of the unrestricted phase x:

$$\Theta_w(u,t)=\sum_{w=-\infty}^{\infty}\frac{1-e^{-2\pi iu}}{2\pi i(u+w)}\Theta(u+w,t). \tag{2.10}$$

The variance of the winding number $\sigma_w^2(t)=-\partial^2\Theta_w(0,t)/\partial u^2+[\partial\Theta_w(0,t)/\partial u]^2$ and of the extended phase agree up to a factor in the limit of large times:

$$\sigma_x^2(t)\sim4\pi^2\sigma_w^2(t). \tag{2.11}$$

In the remainder, we will consider some models describing anomalous diffusion and determine the relaxation of the according phase variable.

III. PROCESSES WITH INDEPENDENT INCREMENTS

We start our discussion with the class of processes with independent increments, i.e., with processes $x(t)$ for which the increments $x(t_2)-x(t_1)$, $x(t_3)-x(t_2)$, $x(t_4)-x(t_3)$, etc. with $t_1<t_2<t_3<\cdots$ are mutually independent from each other [11]. If the increments moreover are stationary, i.e., if their distributions depend only on the time difference, say t_2-t_1, then, the characteristic function of processes with independent increments is an exponential function with an exponent that is linear in time [12]:

$$\Theta(u,t)=\exp\{t\Phi(u)\}, \tag{3.1}$$

where the function $\Phi(u)$ is the cumulant generating function per unit time. According to the definition of the characteristic function, $\Phi(u)$ vanishes at $u=0$ and $\Phi(0)=0$; also see Eq. (A2) below. Because the distribution of any process with stationary independent increments also is infinitely divisible [11], the cumulant generating function per unit time can be represented by the Lévy-Khinchin formula [12],

$$\Phi(u)=iua+\int_{-\infty}^{\infty}\left(e^{iux}-1-\frac{iux}{1+x^2}\right)\frac{1+x^2}{x^2}dF(x), \tag{3.2}$$

where a is a real constant and $F(x)$ is a bounded, monotonically nondecreasing function with $F(-\infty)=0$.

In view of Eq. (2.6), a process with independent increments leads to an exponential relaxation law for the fundamental mean values of the phase,

$$m_n(t)=e^{(-\kappa_n+i\omega_n)t}, \tag{3.3}$$

where the relaxation constants κ_n and frequencies ω_n can be expressed in terms of integrals of the function $F(x)$:

$$\begin{aligned}\kappa_n&=-\mathrm{Re}\ \Phi(n)\\ &=\int_{-\infty}^{\infty}[1-\cos(nx)]\frac{1+x^2}{x^2}dF(x),\\ \omega_n&=\mathrm{Im}\ \Phi(n)\\ &=na+\int_{-\infty}^{\infty}\left[\sin(nx)-\frac{nx}{1+x^2}\right]\frac{1+x^2}{x^2}dF(x).\end{aligned} \tag{3.4}$$

Here Re and Im denote the real and imaginary parts, respectively. The relaxation constants κ_n are positive for all admissible functions $F(x)$. Only if $F(x)$ is a constant apart from finite steps at nonzero integer multiples of 2π, $x=2\pi n$ and $0\neq n\in\mathbb{Z}$, do the relaxation constants vanish; $\kappa_n=0$. In this case, the extended process $x(t)$ moves in jumps of the length of an integer fraction of 2π. Wrapped onto the unit circle, this process periodically visits a discrete number of points, and the mean values $m_n(t)$ result as periodic functions of time.

If $F(x)$ has only a single jump at $x=0$ of the height σ^2 and is constant everywhere else, then the cumulant generating function per time becomes

$$\Phi(u)=iua-\frac{1}{2}\sigma^2u^2. \tag{3.5}$$

The extended variable $x(t)$ performs an ordinary Gaussian diffusion characterized by the diffusion constant $D_1=\sigma^2$, and the damping constants of the fundamental mean values are given by $\kappa_n=D_1n^2/2$. For general functions $F(x)$, no simple relation exists between the relaxation and diffusion constants. The latter may even diverge, whereas the relaxation constants are always finite.

IV. SELF-SIMILAR PROCESSES

By definition, the finite time distributions of a self-similar process are invariant under a joint rescaling of the time t by an arbitrary positive factor λ, and of the state variable x by the factor λ^{ζ} with a convenient scaling exponent ζ,

$$x(\lambda t)\overset{d}{=}\lambda^{\zeta}x(t), \tag{4.1}$$

where $\overset{d}{=}$ indicates equality in distribution. Consequently, a self-similar process lacks absolute scales of magnitude and time.

From scaling relation (4.1), one immediately recovers the diffusion law [Eq. (2.1)] with the scaling exponent $\beta=2\zeta$, provided that the variance of the process is finite, or, more generally, law (2.2) follows with the linear scaling exponent $\beta(p)=\zeta p$.

Below we will only make use of the single time distribution of the process $x(t)$. For the single time probability density $\rho(x,t)dx=\mathrm{Prob}(x\leqslant x(t)<x+dx)$ the scale invariance [Eq. (4.1)] implies

$$\rho(x,\lambda t)=\lambda^{-\zeta}\rho(\lambda^{-\zeta}x,t). \tag{4.2}$$

Consequently, the probability density at any time t is related to that at a reference time $t_0>0$ by

$$\rho(x,t)=f\left[\left(\frac{t_0}{t}\right)^{\zeta}x\right]\left(\frac{t_0}{t}\right)^{\zeta}, \tag{4.3}$$

where $f(x)=\rho(x,t_0)$ is the probability density at a reference time t_0. The characteristic function of $x(t)$ and, hence, all mean values $m_n(t)$ follow from that of $x(t_0)$,

$$m_n(t)=\Theta_f\left[n\left(\frac{t}{t_0}\right)^{\zeta}\right], \tag{4.4}$$

where

$$\Theta_f(u)=\int_{-\infty}^{\infty}dx\ e^{iux}f(x) \tag{4.5}$$

is the characteristic function of the reference distribution $f(x)$.

We note that the exponent is restricted to values $\zeta\leqslant 1$ if the increments of a self-similar process are stationary [13], i.e., the distribution of the increment $x(t)-x(s)$ depends only on the time difference $t-s$. Here we will restrict ourselves to stable distributions [12] as reference distributions, and start with the special case of a Gaussian distribution leading to so called fractional Brownian motion for the process $x(t)$.

A. Fractional Brownian motion

For a Gaussian reference distribution

$$f(x)=\frac{1}{\sqrt{2\pi\sigma_0^2}}\exp\left\{-\frac{x^2}{2\sigma_0^2}\right\}, \tag{4.6}$$

one obtains a self-similar process $x(t)$ known as fractional Brownian motion [14]. Here σ_0^2 is the variance of x at the reference time t_0. Using Eq. (4.4) we find that the fundamental mean values relax according to nonlinear exponential laws:

$$m_n(t)=\exp\left\{-\frac{\sigma_0^2}{2}n^2\left(\frac{t}{t_0}\right)^{2\zeta}\right\}. \tag{4.7}$$

The exponent 2ζ in this law is independent of n. For stationary increments of the phase, it may vary from zero to 2, covering the regimes of stretched exponential $0<2\zeta<1$, exponential $2\zeta=1$, and faster than exponential, including Gaussian, relaxation $1<2\zeta\leqslant 2$.

B. Lévy processes

The Gaussian distribution is a special case of the Lévy, or stable distributions which result as solutions of the renormalization equation for the distribution of sums of independent identically distributed random numbers [15]. Here we will only be concerned with symmetric Lévy distributions, which are most conveniently characterized by their characteristic functions [12]

$$\Theta(u)=\exp\{-\sigma_0^\alpha|u|^\alpha\}, \tag{4.8}$$

where $\sigma_0>0$ is a reference scale and α is an exponent which is restricted to $0<\alpha\leqslant 2$. For $\alpha=2$ one obtains a Gaussian distribution. For $\alpha<2$ the probability density of a Lévy distributed random variable falls off as $\rho(x)\sim|x|^{-(1+\alpha)}$ [12]. Consequently, the moments $\langle|x|^p\rangle$ of the reference distribution then only exist for $0<p<\alpha$.

Using Eqs. (4.2) and (4.8), one recovers a power law with exponent $p\zeta$ for the pth moment of $x(t)$:

$$\langle|x(t)|^p\rangle=C_{p,\alpha}\sigma_0^p\left(\frac{t}{t_0}\right)^{p\zeta}, \tag{4.9}$$

where

$$C_{p,\alpha}=\frac{1}{2\pi}\int_{-\infty}^{\infty}dx|x|^p\int_{-\infty}^{\infty}du e^{iux-|u|^\alpha}. \tag{4.10}$$

This time-independent prefactor has a finite value for $0\leqslant p<\alpha$, and diverges for $p\geqslant\alpha$ if $\alpha<2$. For a process with stationary increments, the similarity exponent ζ is restricted to $\zeta<1/\alpha$ for $0<\alpha<1$ and $\zeta\leqslant 1$ for $1\leqslant\alpha\leqslant 2$ [13].

If x is an extended phase, then the fundamental mean values $m_n(t)$ relax according to a stretched exponential law following from Eq. (4.4):

$$m_n(t)=\exp\left\{-(n\sigma_0)^\alpha\left(\frac{t}{t_0}\right)^{\alpha\zeta}\right\}. \tag{4.11}$$

The stretching exponent $\alpha\zeta$ is less than or equal to 1 for $0<\alpha<1$, and less than or equal to α for $1\leqslant\alpha\leqslant 2$, provided the increments of the extended variable $x(t)$, and hence also those of the phase are stationary. So the same range of stretching exponents is covered as in the case of fractional Brownian motion although the respective diffusional behavior is very different.

V. CONTINUOUS TIME RANDOM WALKS

Another class of processes that may lead to anomalous diffusion and that have found various physical applications are continuous time random walks [16]. We will collect the relevant relations for these processes that will be needed here, and then discuss the ensuing phase relaxation for a few particular cases.

A. Expression for the probability density in terms of the combined jump and waiting time distribution

A continuous time random walk is characterized by the joint jump probability $\psi(x,t)$ giving the likelihood that the process pauses for the time t in a state until it makes a jump over the distance x. Note that this probability is independent of the absolute time and the actual state of the process, and, hence, is homogeneous both in time and space. The waiting time between jumps of arbitrary lengths is distributed according to the density

$$w(t)=\int_{-\infty}^{\infty}dx\ \psi(x,t). \tag{5.1}$$

The distribution of the jump width is given by

$$\lambda(x)=\int_0^{\infty}dt\ \psi(x,t). \tag{5.2}$$

For later use we introduce the conditional density of a jump of length x if the jump takes place at the time t after the previous jump:

$$\psi(x|t)=\frac{\psi(x,t)}{w(t)}. \tag{5.3}$$

The Fourier-Laplace transform of the probability density $W(x,t)$, giving the likelihood that the process has reached a distance x at time t from where it started, is known in terms of the respective transforms of the marginal waiting time distribution and the joint jump distribution [16]

$$\hat{\tilde{W}}(u,z)=\frac{1-\hat{w}(z)}{z}\frac{1}{1-\hat{\tilde{\psi}}(u,z)}, \tag{5.4}$$

where we denote Fourier and Laplace transformed functions by the same symbols as the original ones distinguished by a tilde and a hat, respectively:

$$\hat{\tilde{\psi}}(u,z)=\int_{-\infty}^{\infty}dx\int_0^{\infty}d\ te^{iux}e^{-zt}\psi(x,t). \tag{5.5}$$

The long-time behavior of the fundamental phase mean values and the variance of the extended process follow from Eq. (5.4) together with the general relations (2.6) and (2.7) in the limit of small z. The relation between the long-time and small-z behavior of a function and its Laplace transform, respectively, can often be obtained by means of the Tauber theorem [17]. This theorem relates an algebraic long-time behavior of a function $h(t)$, say $h(t)\sim t^{-\alpha}$, to an algebraic behavior of the corresponding Lapace transform $\hat{h}(z)$ for small z given by $\hat{h}(z)\sim z^{\alpha-1}$, and vice versa, provided $h(t)$ is non-negative and monotone at infinity; see Ref. [17] for a precise statement of the theorem.

B. Factorizing joint jump distributions

If the jump length and the waiting time are independent of each other, the joint density $\psi(x,t)$ factorizes as

$$\psi(x,t)=\lambda(x)w(t), \tag{5.6}$$

and $\hat{\tilde{W}}(u,z)$ becomes:

$$\hat{\tilde{W}}(u,z)=\frac{1-\hat{w}(z)}{z}\frac{1}{1-\tilde{\lambda}(u)\hat{w}(u)}. \tag{5.7}$$

We will discuss this result in the cases of long and short rests.

1. Long rests, short jumps

Long rests are characterized by an algebraic decay of $w(t)$ at large times,

$$w(t)\sim t^{-(1+\alpha)}, \tag{5.8}$$

with an exponent $\alpha>0$. The corresponding Laplace transform at small arguments is then given by (see Appendix B)

$$\hat{w}(z)=\frac{1}{1+c|z|^{\alpha}}, \tag{5.9}$$

where c is a positive constant. The functional form correctly takes into account the normalization and the long-time behavior of the waiting time distribution. For the jump width we assume a Gaussian distribution so that short jumps prevail:

$$\tilde{\lambda}(u)=e^{-\sigma^2u^2/2}. \tag{5.10}$$

For the extended process, it is sufficient to consider the joint jump distribution for small values of u. For the Fourier-Laplace transformed density one then finds:

$$\hat{\tilde{W}}(u,z)=\frac{1}{z}\frac{1}{1+\frac{1}{2}\sigma^2u^2c^{-1}|z|^{-\alpha}}. \tag{5.11}$$

With Eq. (2.7) and the Tauber theorem this yields the expected diffusion behavior for the extended process:

$$\langle x^2(t)\rangle\sim\frac{\sigma^2}{2c\Gamma(\alpha)}t^{\alpha}. \tag{5.12}$$

Similarly, one obtains with [Eq. (2.6)] for the long-time behavior of the fundamental phase mean values:

$$m_n(t)\sim\frac{c}{\Gamma(\alpha)[1-\tilde{\lambda}(n)]}t^{-\alpha}. \tag{5.13}$$

Here one must in general not use the small-u expansion of the jump distribution. Note that the decay of the fundamental mean values of the phase is algebraic, and, hence, much slower than the stretched exponential relaxation that emerges from a fractional Brownian motion having the same diffusion exponent $2\zeta=\alpha$. The exponent describing the algebraic phase decay [Eq. (5.13)] is independent of the jump distribution which only determines the prefactor. In contrast to the diffusion law [Eq. (5.12)], the relaxation [Eq. (5.13)] is not restricted to short jumps but holds for arbitrary jump widths distributions.

2. Short rests, arbitrary jumps

An exponential waiting time distribution is characterized by the average waiting time. The probability for rests longer than this characteristic time rapidly decreases. In this sense, rests are typically short. The resulting continuous time random walk has independent increments and, consequently, the fundamental phase mean values relax exponentially; see Sec. III. However, an exponential relaxation is found for the wider class of waiting time distributions for which a mean waiting time exists:

$$\langle t\rangle=\int_0^{\infty}dt\,t\,w(t). \tag{5.14}$$

The Laplace transform for small u then behaves as (see Appendix B)

$$\hat{w}(z)=\frac{1}{1+\langle t\rangle z}. \tag{5.15}$$

For an arbitrary jump distribution $\lambda(x)$ this gives

$$\hat{\tilde{W}}(u,z)=\frac{\langle t\rangle}{1-\tilde{\lambda}(u)+\langle t\rangle z}. \tag{5.16}$$

The inverse Laplace transform yields the claimed exponential relaxation of the fundamental phase mean values:

$$m_n(t)=e^{-(1-\tilde{\lambda}(n))t/\langle t\rangle}. \tag{5.17}$$

The diffusion of the extended process is normal if the jump width distribution possesses a finite second moment.

C. Correlated waiting times and jump width

For the long-time behavior of both the extended diffusion and the phase relaxation, the asymptotic Fourier-Laplace transform of the jump distribution $\hat{\tilde{\psi}}(u,z)$ is important at small values of z. We can split off the Fourier transform of the jump width distribution $\lambda(x)$, and obtain

$$\hat{\tilde{\psi}}(u,z)=\tilde{\lambda}(u)-\chi(u,z), \tag{5.18}$$

where $\chi(u,z)$ is a function that vanishes for all u at $z=0$. We first consider the fundamental phase mean values which are determined by $\hat{\tilde{W}}(u,z)$ at integer values u. Typically, the absolute value of the Fourier transform of the jumps width distribution $|\tilde{\lambda}(u)|$ is less than 1 at finite values of u; see Eq. (A3). Hence one can neglect the small term $\chi(u,z)$ compared to $1-\tilde{\lambda}(u)$ in the denominator of Eq. (5.4), and obtain the same form for the Fourier-Laplace transformed density $\hat{\tilde{W}}(u,z)$ at small z as for independent jump widths and waiting times:

$$\hat{\tilde{W}}(u,z)=\frac{1-\hat{w}(z)}{z}\frac{1}{1-\tilde{\psi}(u)}. \tag{5.19}$$

Therefore, we find the same results for the relaxation of the fundamental mean values of the phase as for independent waiting times and jump width. If a finite mean waiting time exists the exponential relaxation [Eq. (5.17)] is recovered, and if the waiting time distribution decays only algebraically, i.e., $\chi(z)=cz^{\alpha}+o(z^{\alpha})$ with $0<\alpha<1$, then we again find the algebraic decay law [Eq. (5.13)].

The variance of the extended process is given by the general expression (2.7). Its Laplace transform with respect to time can be expressed in terms of the first derivatives of its Fourier-Laplace transformed density:

$$\begin{aligned}\hat{\sigma}_x^2(z)&=-\frac{\partial^2\hat{\tilde{W}}(0,z)}{\partial u^2}+\left(\frac{\partial\hat{\tilde{W}}(0,z)}{\partial u}\right)^2\\&=-\frac{1}{z(1-\hat{w}(z))}\frac{\partial^2\hat{\tilde{\psi}}(0,z)}{\partial u^2},\end{aligned} \tag{5.20}$$

where in the second line we used the particular form [Eq. (5.4)] of the probability density of a continuous time random walk. For the sake of simplicity we assumed that the joint jump distribution $\psi(x,t)$ is even in x and, hence, the first derivative of the Fourier-Laplace transform with respect to u vanishes at $u=0$. Accordingly, the long-time behavior of the variance is determined by the small-z dependence both of the second derivative of $\hat{\tilde{\psi}}(u,z)$ and of the waiting time distribution. The second derivative may scale in a different way than the waiting time distribution, and so the scaling exponent for the diffusion of the extended process may be different from the scaling exponent for the phase relaxation, which is completely determined by the waiting-time distribution. We will illustrate this with an example.

We consider the case of a Gaussian conditional jump width distribution given by

$$\psi(x|t)=\frac{1}{\sqrt{2\pi\sigma^2(t)}}e^{-x^2/2\sigma^2(t)}, \tag{5.21}$$

where the variance $\sigma(t)$ may itself diffusively grow with the waiting time t:

$$\sigma^2(t)=D_\beta t^\beta. \tag{5.22}$$

Hence, after a long waiting time a wide jump becomes more likely. The Fourier-Laplace transformed joint jump distribution then becomes

$$\hat{\tilde{\psi}}(u,z)=\int_0^\infty dt\,e^{-zt}e^{-(1/2)\sigma^2(t)u^2}w(t). \tag{5.23}$$

The waiting time distribution $w(t)$ is assumed to have an algebraic tail $\sim t^{-\alpha-1}$ and, consequently, for small z its Laplace transform assumes the form

$$\hat{w}(z)=\frac{1}{1+cz^{\alpha}}, \tag{5.24}$$

where c is a positive constant and $0<\alpha\leqslant 1$. Note that with $a=1$ exponential waiting times are also included in Eq. (5.24). For jump width exponents $\beta\geqslant 0$, the integral $t(u)=\int_0^\infty dt\,t\,e^{-(1/2)\sigma^2(t)u^2}w(t)$ exists for all $u\neq 0$ and for all waiting time distributions $w(t)$. Hence, for nonvanishing u and small values of z, one finds

$$\hat{\tilde{\psi}}(u,z)=\hat{\lambda}(u)-t(u)z+O(z^2). \tag{5.25}$$

Note that $t(u)$ diverges at $u=0$ if $\alpha<1$ and, then, instead of Eq. (5.25), Eq. (5.24) holds for $\hat{\tilde{\psi}}(0,z)=\hat{w}(z)$. Using these expansions for the small-z behavior of $\hat{\tilde{W}}(u,z)$ at $u\neq 0$ we find

$$\hat{\tilde{W}}(u,z)=cz^{\alpha-1}\frac{1}{1-\tilde{\lambda}(u)+t(u)z}. \tag{5.26}$$

For the fundamental phase mean values one obtains the same asymptotic long-time result as given in Eq. (5.13), i.e. an algebraic decay proportional to $t^{-\alpha}$ for $0<\alpha<1$. Note that the exponent is independent of the spreading of the jump width distribution, and only determined by the waiting time distribution $w(t)$. For $\alpha=1$, corresponding to an exponential waiting time distribution, we find an exponential relaxation of $m_n(t)$.

The variance of the extended process is determined by the second derivative of the Fourier-Laplace transformed joint jump probability at $u=0$ [see Eq. (5.20)], which in this particular case is given by

$$\frac{\partial^2\hat{\tilde{\psi}}(0,z)}{\partial u^2}=-\int_0^\infty dt\;e^{-zt}\sigma^2(t)w(t). \tag{5.27}$$

If the growth of $\sigma^2(t)$ is so slow that the integral converges to a finite value for $z\to 0$, i.e., if $\alpha>\beta$, then a diffusive behavior of $x(t)$ with the exponent α of the waiting time results. If, however, $\beta>\alpha$, $\hat{\sigma}_x^2(z)$ diverges as $z^{-\beta+\alpha}$ and the diffusion of the extended process $x(t)$ is characterized by the exponent β, $\sigma_x^2(t)\sim t^{\beta}$, and, hence, is independent of α.

VI. CONCLUSIONS

We have compared the relaxational behavior of phase variables that results from different diffusion models of the respective extended phase. For processes with stationary independent increments, the relaxation is always exponential. According to the central limit theorem the diffusion of the extended phase is normal for long times if the second moments of the increments exist. If only absolute moments of the order $p<p_c$ exist, then Eq. (2.2) holds for sufficiently large times and for $p<p_c$ with $\beta(p)=p/p_c$.

For self-similar processes, the phase relaxation is given by the decay of the characteristic function of the unresticted phase, algebraically stretched by the similarity exponent. In the considered cases of Gaussian and Lévy processes, this leads to a stretched exponential relaxation of the phase with stretching exponents ranging from 0 to 2. This provides a simple generation mechanism of stretched exponential relaxation that also might be relevant to interpreting muon spin resonance relaxation [18].

For continuous time random walks, short rests which are characterized by a finite mean waiting time lead to exponential phase relaxation, and, if additionally the jumps are short, the accompanying extended diffusion is normal. Long rests, i.e., waiting time distributions without a finite mean value, give rise to algebraic decay. However, we note that, for correlated jump widths and waiting times, the exponents characterizing the algebraic phase decay and the diffusion may be different. In particular, the diffusion may be normal, but at the same time the phase relaxation may only be algebraic.

In all cases considered here one can also determine the scaling behavior of other than second centered moments. For the processes with independent increments and the self-similar processes, it is obvious that these moments scale according to Eq. (2.2) with a linear dependence of the exponent $\beta(p)$ on p, provided the considered moments exist. One can show that this is also true for all continuous time random walks. The general case of multifractal processes will be studied separately.

Although both the phase relaxation and the diffusion of the extended phase are determined by the same characteristic function, the two phenomena may appear quite unrelated. The mathematical reason for this discrepancy is that, while the diffusion is determined by the second derivative of the characteristic function at the wave number $u=0$, the behavior at integer wave numbers governs the phase relaxation.

Based on this observation, we suggest for the time series analysis of phases and of diffusional processes complementary strategies additionally to the existing methods. For a diffusion process, we propose not only to consider the growth behavior of the variance and higher centered moments, but also to introduce a fictitious period and to consider the relaxation of the resulting phase variable. Varying the period is tantamount to analyzing the full characteristic function. Also, if a phase is monitored, as is usually done in nuclear magnetic resonance and in muon resonance experiments, the diffusional aspect of the phase motion can be fully analyzed if the sampling rate of the data is high enough. Assuming a continuous motion of the considered phase, or at least a jumplike motion with jumps much smaller than the period, one can reconstruct the winding number by monitoring whether the period is left at 0 or 2π, leading to a change of the winding number by -1 or $+1$, respectively. The resulting extended (unwrapped) phase can then be analyzed by the various methods available for diffusion processes with respect to its statistical properties and its behavior in time. We hope to come back to this problem in a future investigation.

APPENDIX A: GENERAL PROPERTIES OF THE CHARACTERISTIC FUNCTION

The characteristic function of a random variable x is the Fourier transform of the probability density $\rho(x)$ (we suppress a possible dependence on time t which here is a mere parameter):

$$\Theta(u)=\int_{-\infty}^{\infty}dx\ e^{iux}\rho(x). \tag{A1}$$

This is always a continuous function of the real variable u, taking a value of 1 at $u=0$:

$$\Theta(0)=1; \tag{A2}$$

otherwise

$$|\Theta(u)|\leqslant 1. \tag{A3}$$

If the probability density $\rho(x)$ is a continuous function of x, the characteristic function $\Theta(u)$ vanishes for $|u|\to\infty$:

$$\lim_{|u|\to\infty}|\Theta(u)|=0. \tag{A4}$$

For further details, see Ref. [12].

APPENDIX B: GENERATING FUNCTION FOR WAITING TIMES

The Laplace transform $\hat{w}(z)=\int_0^\infty \exp\{-zt\}w(t)$ of a waiting time distribution is a generating function for the moments of the waiting time:

$$\langle t^n\rangle=(-1)^n\frac{d^n\hat{w}(0)}{dz^n}. \tag{B1}$$

If the nth derivative at $z=0$ does not exist, the respective moment of the waiting time diverges, and vice versa. Here we collect some analytical properties of the generating function for real, non-negative values of z.

(i) $\hat{w}(0)=1$. This follows immediately from the normalization of the waiting time distribution.

(ii) $\hat{w}(z)>0$ for all $0\leqslant z<\infty$, as is obvious from the definition of the moment generating function as a Laplace transform of a positive function.

(iii) $\hat{w}(z)$ is a nonincreasing function: $\hat{w}(z_1)-\hat{w}(z_2)=\int_0^\infty dt(\exp\{-z_1t\}-\exp\{-z_2t\})w(t)\geqslant 0$ for $z_1<z_2$. For a stronger growth property, see Ref. [17].

(iv) $\hat{w}(z)$ is infinitely often differentiable for all $z>0$.

(v) $\hat{w}(z)\to 0$ for $z\to\infty$ if there are no immediate jumps. This means that the probability for a jump within a short initial period of length τ vanishes as τ^γ with some positive exponent γ: $\int_0^\tau dt\, w(t)\leqslant c\tau^\gamma$ for $\tau\to 0$ with $\gamma>0$ and $c>0$.

Using these properties, one can represent the Laplace transformed waiting time probability in terms of another function $\chi(z)$,

$$\hat{w}(x)=\frac{1}{1+\chi(z)}, \tag{B2}$$

where $\chi(z)$ vanishes at $z=0$ and is an increasing smooth function for $z>0$ which goes to infinity for $z\to\infty$ if there are no immediate jumps in the sense of (v).

For a waiting time distribution possessing a finite first moment, $\chi(z)$ is differentiable at $z=0$, and starts to grow like

$$\chi(z)=\langle t\rangle z+o(z). \tag{B3}$$

For a waiting time distribution with an algebraic long-time tail, $w(t)\sim t^{-(1+\alpha)}$ with $0<\alpha<1$, $\chi(z)$ grows at $z=0$ like

$$\chi(z)=cz^{\alpha}+o(z^{\alpha}). \tag{B4}$$

[1] J.R. Manning, *Diffusion Kinetics for Atoms in Solids* (Van Nostrand, Princeton, NJ, 1968).

[2] J. Philibert, *Atom Movements, Diffusion and Mass Transport in Solids*, Les Edition de Physique (Cedex, Les Ulis, 1991).

[3] For a historical account, see S.G. Brush, *The Kind of Motion We Call Heat* (North Holland, Amsterdam, 1976).

[4] A.S. Monin and A.M. Yaglom, *Statistical Fluid Mechanics: Mechanics of Turbulence* (MIT Press, Cambridge, MA, 1975), Vol. II.

[5] J.-P. Bouchaud and A. Georges, Phys. Rep. **195**, 127 (1990); R. Metzler and J. Klafter, *ibid.* **339**, 1 (2000).

[6] P.W. Anderson and P.R. Weiss, Rev. Mod. Phys. **25**, 269 (1953).

[7] R. Kubo, in *Fluctuation, Relaxation and Resonance in Magnetic Systems*, edited by D. ter Haar (Olver and Boyd, Edinburgh, 1961), p. 23.

[8] A. Abragam, *The Principles of Nuclear Magnetism* (Oxford University Press, Oxford, 1961).

[9] H. Haken, *Laser Theory*, Encyclopedia of Physics, XXV/2c (Springer-Verlag, Berlin, 1970).

[10] N. Ikeda and S. Watanabe, *Stochastic Differential Equations and Diffusion Processes* (North-Holland, Amsterdam, 1981).

[11] W. Feller, *An Introduction to Probability Theory and Its Applications* (Wiley, New York, 1968), Vol. I.

[12] E. Lukacs, *Characteristic Functions* (Griffin, London, 1970).

[13] G. Samorodnitsky and M.S. Taqqu, *Stable Non-Gaussian Random Processes* (Chapman and Hall, New York, 1994).

[14] B.B. Mandelbrot and J.W. Van Ness, SIAM Rev. **10**, 422 (1968).

[15] M. Cassandro and G. Jona-Lasinio, Adv. Phys. **27**, 913 (1977).

[16] E.W. Montroll and M.F. Shlesinger, in *Nonequilibrium Phenomena II, from Stochastics to Hydrodynamics*, Studies in Statistical Mechanics, edited by J.L. Lebowitz (North-Holland, Amsterdam, 1984), p. 44.

[17] W. Feller, *An Introduction to Probability Theory and Its Applications* (Wiley, New York, 1968), Vol. II.

[18] Y.J. Uemura, in *Muon Science*, edited by S.L. Lee, S.H. Kilcoyne, and R. Cywinski (Institute of Physics, Bristol, 1999), p. 85.