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A phenomenological approach to anomalous transport in complex or disordered media

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Abstract

We aim to derive a phenomenological approach to link the theories of anomalous transport governed by fractional calculus and stochastic theory with the conductivity behavior governed by the semi-empirical conductivity formalism involving Debye, Cole-Cole, Cole-Davidson, and Havriliak-Negami type conductivity equations. We want to determine the anomalous transport processes in the amorphous semiconductors and insulators by developing a theoretical approach over some mathematical instruments and methods. In this paper, we obtain an analytical expression for the average behavior of conductivity in complex or disordered media via using the fractional-stochastic differential equation, the Fourier-Laplace transform, some natural boundary-initial conditions, and familiar physical relations. We start with the stochastic equation of motion called the Langevin equation, develop its equivalent master equation called Klein-Kramers or Fokker-Planck equation, and consider the time-fractional generalization of the master equation. Once we derive the fractional master equation, then determine the expressions for the mean value of the variables or observables through some calculations and conditions. Finally, we use these expressions in the current density relation to obtain the average conductivity behavior.

Keywords: Anomalous transport, fractional master equations, long-tailed distributions, Prabhakar derivative, Caputo regularization, Havriliak-Negami type behavior

1 Introduction

Transport dynamics in complex or disordered media that is also called anomalous transport, such as amorphous semiconductors and insulators, is presented and constructed through the stochastic and fractional calculus instruments. The anomalous transport process is governed by biased anomalous diffusion with asymmetric non-exponential or non-Gaussian distributions that involves the heavy- or long-tailed distributions or the probability distributions with power-law tails [1, 2].

Anomalous transport can be described through the time- and/or space-fractional drift-diffusion equation that involves anomalous diffusion and fractional drift processes. The manifestation of the anomalous diffusion is the occurrence of the second moment of displacement or the mean square displacement of the form $\langle z^2(t) \rangle \sim t^\alpha$, where $\alpha \neq 1$ and depending on the exponent α the diffusion process can be distinguished subdiffusion for the case $0 < \alpha < 1$ or superdiffusion for $\alpha > 1$ [3, 4].

Fractional drift can be interpreted through the time-fractional Fokker-Planck equation because that both drift and diffusion terms are affected by the time-fractional operator but in the space-fractional equation, the drift term is not affected by the space-fractional operator [5].

Stochastic processes that can be classified according to the index variable into two main group called discrete-time and continuous-time stochastic processes that both also have exponential probability distributions. Discrete-time stochastic processes involving such as binomial process, Bernoulli process, discrete-time random walk, etc define a basic random process and have normal or Gaussian distribution at some special limit cases [6–8]. Continuous-time stochastic processes involving such as the Wiener process, Ornstein-Uhlenbeck process, continuous time random walk, etc. define a diffusion or drift-diffusion process and have an exponential type probability distribution such as Gaussian(normal) or shifted Gaussian distribution [6–8].

Random walk in random environment or the continuous-time random walk(CTRW) are used analogical methods to perform anomalous transport process. The CTRW describes the random motion of a particle from one lattice site to another by waiting a stochastic time and stochastically jumping [9, 10]. A generalized master equation(GME) is obtained from a CTRW that can be derived from a step length and a waiting time probability distribution. GMEs defined through some special space- and time-dependent probability distributions even converge to the standard Fokker-Planck equation in the diffusion limit [11].

A CTRW model may be described by a GME and also the Langevin equation(LE) that is composed of deterministic and stochastic terms as a generalization of the second law of motion [12]. The LE can be transformed into the Fokker-Planck equation through deriving Kramers-Moyal expansion coefficients related to the Langevin equation or equation of motion for the probability density in the Langevin equation [13–15]. Both the GMEs and LEs have equivalent master equations(MEs) such Klein-Kramers(KKE), Fokker-Planck(FPE), Telegraph Equation(TE), Diffusion-Advection(DAE) equations that are each expressing different stochastic processes [16–18, 54].

Fractional dynamics have been considered to play a central role in modeling anomalous transport in complex media that are non-Markovian(memoryless) and non-local stochastic processes [20–22]. Though master equations can be obtained from the different GMEs and LEs that define normal transport processes, fractional generalization of the master equations is needed to understand the anomalous processes. Time- and space-fractional master equations arise from the long-tailed waiting time distribution and the long-ranged jump length distribution that are non-exponential and asymmetric in the presence of an external bias [23, 24].

The analytical solution of the fractional master equation chosen for the corresponding transport problem can be obtained by the use of the Fourier-Laplace(FL) transform that allows changing the space-time with the FL domain [25–27]. When the fractional master equation and its FL transform have been derived, an analytical solution can be obtained for the probability density function(PDF), and then the natural boundary conditions for the PDF can be considered to avoid vanishing terms. The inverse FL transform of the PDF is usually given in terms of the Fox H-function. Through the parallel analytical derivation, the mean or expected value of the variables or observables can be obtained by using the same mathematical approaches.

In literature, three expressions called Cole-Cole, Cole-Davidson, Havriliak-Negami equations have been described according

to the experimental data on disordered systems for the dielectric relaxation. The corresponding differential equations of response-excitation or probability density functions, containing fractional operators in the time-domain have been obtained, and solved by Fourier-Laplace transform to yield the relaxation functions of the current semi-empirical formalism [28–32]. Though the relaxation models have been studied extensively, neither obtained a conductivity formalism via using the relaxation models based on the relaxation-conductivity analogy [33], also nor studied for deriving theoretical approaches via using stochastic-fractional dynamical instruments to obtain a conductivity formalism and interpret anomalous transport processes.

The anomalous transport in complex or disordered media involving amorphous semiconductors and insulators is governed by some nonlinear and nonlocal processes corresponding to analytically derived Debye and semi-empirical Cole-Cole(CC), Cole-Davidson(CD) and Havriliak-Negami(HN) type conductivity equations [34–38]. HN type conductivity equation may be derived analytically through using the mathematical instruments and approaches of the stochastic and fractional dynamics and this sheds light on the types of anomalous behaviors that occurred in the conductivity process. Starting with LE, getting the equivalent master equation and obtaining the mean values of the observables over this equation gives a chance to interpret the conductivity in complex media in terms of the anomalous drift and diffusion processes.

This paper is searching for a direct theoretical derivation of conductivity formalism in amorphous semiconductors and insulators by using stochastic-fractional dynamical instruments, and also for the interpretation of the formalism via the anomalous transport processes. In this work, the average conductivity behavior is obtained by using the mean velocity expression, that is derived from a fractional-stochastic differential equation under the natural boundary conditions, in the current density relation. In literature, it seems that the expression for the mean value of an observable is obtained in some papers but ever not used to derive an analytical average behavior of another observable via using a physical relation. The novelty of the work comes from that the expression for the mean velocity is obtained from the stochastic differential equation including fractional operators and used to derive an analytical average behavior of conductivity relating to anomalous transport processes in complex or disordered media. Our motivation is to construct the relationship between the semi-empirical conductivity equations especially HN type and the anomalous transport processes is governed by the approaches of fractional and stochastic dynamics. We want to determine the types of anomalous processes in complex media correlated with semi-empirical conductivity formalism and link the semi-empirical formulation with the analytical derivation of conductivity equations.

2 Method

The conductivity behavior in the amorphous semiconductor and insulators are governed by the semi-empirical conductivity equations that are also a manifestation of anomalous transport. The fractional-stochastic description of anomalous transport gives a chance to understand the nonlinear and nonlocal random processes that occur in disordered or complex systems or media. We can mimic the average abnormal behaviors in non-equilibrium systems by using the stochastic differential equations and their fractional generalizations.

The stochastic differential equations called LE, KKE, FPE, and their fractional generalizations are used to determine the

average behavior in the processes of anomalous transport and behavioral abnormalities in terms of long-ranged memory and jump length. Thus we have developed a phenomenological approach to realize the type of abnormal behaviors that occurs in the conductivity processes in complex or disordered media. Developing an analytical approach linking up a direct relationship between the fractional-stochastic theory and the semi-empirical conductivity formalism can be a milestone for further analytical studies and the understanding of the processes of anomalous transport.

The stochastic motion of a particle in position-velocity space can be described by the LEs [39]:

$$\frac{dv}{dt} = -\eta v + \frac{F(z)}{m} + \xi(t), \quad \frac{dz}{dt} = v \quad (1)$$

Here, m , v , z are the mass, velocity, and position of particle respectively, η is a friction coefficient, $F(z)$ is an external force, and $\xi(t)$ is Gaussian white noise.

From (1), we can obtain the equivalent master equation called KKE that describes the general case of the Brownian motion [40].

$$\frac{\partial \rho(z, v, t)}{\partial t} = \left[-v \frac{\partial}{\partial z} + \frac{\partial}{\partial v} \left(\eta v - \frac{F(z)}{m} \right) + \frac{\eta k_B T}{m} \frac{\partial^2}{\partial v^2} \right] \rho(z, v, t) \quad (2)$$

Here, $\rho(z, v, t)$ is the probability distribution function in position and velocity space, k_B and T are the Boltzmann constant and the absolute temperature, respectively.

To describe PDF in only velocity-time space, the equation can be averaged out by integrating the equation over position in the absence of the external force and the KKE is reduced to the ordinary FPE.

$$\frac{\partial}{\partial t} f(v, t) = \eta \frac{\partial}{\partial v} (v f(v, t)) + D \frac{\partial^2}{\partial v^2} f(v, t) \quad (3)$$

The time-fractional generalisation of the FPE (3) by using Riemann-Liouville fractional derivative operator can be defined in the form [41–43]:

$${}^{RL}D_t^\alpha f(v, t) = \gamma_\alpha \frac{\partial}{\partial v} (v f(v, t)) + \kappa_\alpha \frac{\partial^2}{\partial v^2} f(v, t) \quad (4)$$

Here, γ_α is the fractional damping coefficient that has dimension $[\gamma_\alpha] = \text{sec}^{-\alpha}$ and κ_α is the fractional diffusion coefficient that has dimension $[\kappa_\alpha] = \text{cm}^2 / \text{sec}^{\alpha+2}$.

Since we can distinguish the fractional operators of non-integer orders and their corresponding physical effects, type of

phenomena and processes [44], instead we begin to consider for only time-fractional FPE of Prabhakar type that generalizes the Riemann-Liouville derivative and in this case, (3) can be written as follows [45,46]:

$$D_{\alpha,\rho,\lambda}^{\beta} f(v,t) = \gamma_{1-\rho} \frac{\partial}{\partial v} (vf(v,t)) + \kappa_{1-\rho} \frac{\partial^2}{\partial v^2} f(v,t) \quad (5)$$

After multiplying (5) by v , the second derivative of the right-hand side can be re-written as:

$$D_{\alpha,\rho,\lambda}^{\beta} f(v,t) v = \gamma_{1-\rho} \left[f(v,t) v + \frac{\partial f(v,t)}{\partial v} v^2 \right] + \kappa_{1-\rho} \left[\frac{\partial}{\partial v} \left(\frac{\partial f(v,t)}{\partial v} v \right) - \frac{\partial f(v,t)}{\partial v} \right] \quad (6)$$

Some of the terms can be eliminated and the others remain under the natural boundary conditions as follows [47].

$$\lim_{v \rightarrow \pm\infty} f(v,t) = \lim_{v \rightarrow \pm\infty} \frac{\partial f(v,t)}{\partial v} = 0; \quad \lim_{v \rightarrow \pm\infty} \kappa_{1-\rho} \frac{\partial}{\partial v} \left(\frac{\partial f(v,t)}{\partial v} v \right) \quad (7)$$

We can obtain the mean velocity through integrating (6) over v ,

$$D_{\alpha,\rho,\lambda}^{\beta} \int_{-\infty}^{+\infty} f(v,t) v dv = \lim_{v \rightarrow \pm\infty} \kappa_{1-\rho} \int_{-\infty}^{+\infty} \frac{\partial}{\partial v} \left(\frac{\partial f(v,t)}{\partial v} v \right) dv \quad (8)$$

after regulating the integrals as below,

$$D_{\alpha,\rho,\lambda}^{\beta} \langle v(t) \rangle = \lim_{v \rightarrow \pm\infty} \kappa_{1-\rho} \frac{\partial f(v,t)}{\partial v} v \quad (9)$$

and defining the PDF as an ensemble average,

$$f(v,t) = \langle \delta(v - \xi t) \rangle, \quad \frac{\partial f(v,t)}{\partial v} = \langle \delta'(v - \xi t) \rangle \quad (10)$$

here, ξ denotes the constant acceleration that has dimension $[\xi] = \text{cm}/\text{sec}^2$ and we can obtain the (9) as follows.

$$D_{\alpha,\rho,\lambda}^{\beta} \langle v(t) \rangle = \lim_{v \rightarrow \pm\infty} \kappa_{1-\rho} \langle \delta'(v - \xi t) \rangle v \quad (11)$$

and also we are familiar with some characteristics of the Dirac distribution.

$$\delta(v) = \delta(-v), \quad \delta'(-v) = -\delta'(v); \quad \langle \delta'(v - \xi t) \rangle v = \langle \delta(v - \xi t) \rangle \quad (12)$$

We can evaluate the derivative of the Dirac δ -function in the light of the equations (12),

$$D_{\alpha, \rho, \lambda}^{\beta} \langle v(t) \rangle = \lim_{v \rightarrow \pm\infty} \kappa_{1-\rho} \langle \delta(v - \xi t) \rangle \quad (13)$$

the relationship analogue to existing between Riemann-Liouville and Caputo fractional derivatives can be given for also the regularized Prabhakar derivative in Caputo sense and (13) can be rewritten alternatively as [48, 49]:

$${}^C D_{\alpha, \rho, \lambda}^{\beta} \langle v(t) \rangle = \lim_{v \rightarrow \pm\infty} \kappa_{1-\rho} \langle \delta(v - \xi t) \rangle \quad (14)$$

the derivative in (14) can be redefined under the assumption $0 < \alpha\beta < 1$ and with the special choice of parameter $\rho = 1 - \alpha\beta$,

$${}^C D_{\alpha, 1-\alpha\beta, -\lambda}^{\beta} \langle v(t) \rangle = \lim_{v \rightarrow \pm\infty} \kappa_{\alpha\beta} \langle \delta(v - \xi t) \rangle \quad (15)$$

Thus we can introduce the fractional operator in the pseudo-differential form as follows [50, 51]:

$${}^C (\lambda + {}_0 D_t^{\alpha})^{\beta} \langle v(t) \rangle = \lim_{v \rightarrow \pm\infty} \kappa_{\alpha\beta} \langle \delta(v - \xi t) \rangle \quad (16)$$

we know $\lambda = \tau^{-\alpha}$ and then multiply each side by $\tau^{\alpha\beta}$,

$${}^C (1 + \tau^{\alpha} {}_0 D_t^{\alpha})^{\beta} \langle v(t) \rangle = \lim_{v \rightarrow \pm\infty} \tau^{\alpha\beta} \kappa_{\alpha\beta} \langle \delta(v - \xi t) \rangle \quad (17)$$

and we can change the domain of δ -function by using the generalized scaling property as follows:

$${}^C(1 + \tau^\alpha {}_0D_t^\alpha)^\beta \langle v(t) \rangle = \lim_{t \rightarrow \pm\infty} \frac{\tau^{\alpha\beta} \kappa_{\alpha\beta}}{\xi} \langle \delta(t) \rangle \quad (18)$$

Then the multiplier of δ -function has dimension $\left[\frac{\tau^{\alpha\beta} \kappa_{\alpha\beta}}{\xi} \right] = cm$ that can be denoted as x and derived as follows [52].

$$\frac{\tau^{\alpha\beta} \kappa_{\alpha\beta}}{\xi} \langle \delta(t) \rangle = x \langle \delta(t) \rangle = (x - x_0) \langle \delta(t - t_0) \rangle \Big|_{x_0=t_0=0} = \langle v(t) \rangle \quad (19)$$

Thus the term at the right-hand side of (18) is the velocity function and the equation can be redefined as:

$${}^C(1 + \tau^\alpha {}_0D_t^\alpha)^\beta \langle v(t) \rangle = \lim_{t \rightarrow \pm\infty} \langle v(t) \rangle \quad (20)$$

and the Fourier transform of (20) can be applied in consideration of the equations below [53,54].

$$\mathcal{F}[{}^C(1 + \tau^\alpha {}_0D_t^\alpha)^\beta \langle v(t) \rangle](w) = [1 + (i\omega\tau)^\alpha]^\beta \langle v(w) \rangle; \quad \mathcal{F}\left[\lim_{t \rightarrow \pm\infty} \langle v(t) \rangle\right](w) = \lim_{w \rightarrow 0} \langle v(w) \rangle = v_0 \quad (21)$$

We obtain the corresponding equation,

$$[1 + (i\omega\tau)^\alpha]^\beta \langle v(w) \rangle = v_0 \quad (22)$$

for the mean velocity equation, it can be also written as:

$$\langle v(w) \rangle = \frac{v_0}{[1 + (i\omega\tau)^\alpha]^\beta} \quad (23)$$

We can obtain Havriliak-Negami type conductivity with regard to as done in the [38] by using the mean velocity in the mean current density relation as follows:

$$\langle J(w) \rangle = nq \langle v(w) \rangle = \langle \sigma(w) \rangle \langle E(w) \rangle \quad (24)$$

The mean current density is shown as:

$$\langle J(\omega) \rangle = nq \frac{v_0}{[1 + (i\omega\tau)^\alpha]^\beta} \quad (25)$$

We know the velocity for the initial condition

$$v_0 = \left. \frac{qE(\omega)\tau}{m} \right|_{\omega=0} \quad (26)$$

at this stage by using the relation above, we can also define the dc current density

$$\langle J(\omega) \rangle \Big|_{\omega=0} = \frac{nq^2\tau}{m} \frac{1}{[1 + (i\omega\tau)^\alpha]^\beta} E(\omega) \Big|_{\omega=0} = \langle \sigma(\omega) \rangle \langle E(\omega) \rangle \Big|_{\omega=0} \quad (27)$$

Let's we define the dc conductivity

$$\sigma_0 = \frac{nq^2\tau}{m} \quad (28)$$

We obtain the ac conductivity relation that is even known as Havriliak-Negami type conductivity.

$$\langle \sigma(\omega) \rangle = \frac{\sigma_0}{[1 + (i\omega\tau)^\alpha]^\beta} \quad (29)$$

We have developed a new method to understand anomalous transport phenomena through stochastic and fractional dynamical approaches and mathematical instruments. We see also that the semi-empirical Havriliak-Negami type conductivity relation can be derived by getting the observables over an induced form of the time-fractional Fokker-Planck equation with the special choice of drift-diffusion constants and under the natural boundary conditions. Thus, the method gives new ideas about the theoretical background of some semi-empirical equations that is derived to mimic the anomalous type behaviors. We have verified that the Prabhakar type derivative that is regulated in Caputo sense and served as the pseudo-differential equation is closely associated with the anomalous transport phenomena in amorphous semiconductors and insulators.

3 Discussions

We have constructed the relationship between the anomalous transport and the semi-empirical conductivity equation called Havriliak-Negami(HN) type conductivity equation through fractional and stochastic dynamical instruments. For the first step, we have defined the LE that is a stochastic equation of motion, derived its equivalent master equation called KKE, and obtained its simplified form called the ordinary Fokker-Planck equation. FPE is a master equation that is used to interpret normal drift-diffusion processes also called the Ornstein-Uhlenbeck process or Wiener process with drift. Then we have used the time-fractional FPE to determine the transport process according to the theories of anomalous transport.

We have proved that the HN type semi-empirical conductivity equation is correlated with the simplified form of the time-fractional FPE under some natural boundary conditions and in the same manner, we can interpret the HN type conductivity behavior as that arises from the long-tailed waiting time distribution and long-ranged memory according to this renewal process. Similarly, the other types of conductivity equations called Debye, CC, and CD have the same anomalous behaviors that introduce non-Markovian or memory features. Therefore, it can be said that the semi-empirical conductivity formulation is only related to memorial features of the anomalous transport not also with the long-ranged jump length corresponding to long-ranging correlations.

Although the differential equations of response-excitation function or the probability density function and their fractional generalizations corresponding to HN and the other type relaxation functions have been obtained with various methods in literature, any expression for the mean velocity have not been derived and not used to obtain the the average conductivity behavior using the mean current density relation. This paper does not only show that for the first time the analytical average conductivity behavior has been obtained equivalent to the semi-empirical conductivity formalism but also shows that the expression for the average conductivity behavior relevant to anomalous processes could be derived from the mean velocity expression that is obtained from a fractional-stochastic differential equation and used in a familiar physical relation. The novelty of this paper is that the average conductivity behavior has been derived using ever not tried method that uncovers the stochastic and non-linear nature behind the anomalous transport processes in complex or disordered media.

The time and space-dependent fractional FPE should be also worked to derive a complete conductivity theory and emerge all types of anomalous processes for amorphous semiconductors and insulators. This paper attempts to understand the relationship between the phenomena of anomalous transport governed by fractional calculus and stochastic theory and the conductivity behavior in complex or disordered media governed by the semi-empirical conductivity formulation. In this study, we have linked the semi-empirical HN type conductivity behavior and the FPE that is revisited under some natural boundary conditions. However, the master equations have to be reconsidered through some mathematical instruments of fractional and stochastic theory under some boundary and initial conditions to construct a further conductivity formalism.

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