Generalised Diffusion Equations for Anomalous Diffusion in Polymer Networks

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by
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This thesis is dedicated to my lovely wife
Jo, and beautiful children
Ayla, Jakob and Claudia.

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THE SUN IS SHINING...
Murray Ball.

Abstract

Fractional and generalised derivative equations have proven to be a powerful tool in the modelling of anomalous diffusion within *complex fluids* [1–3]. The suitability of these equations arises from to their capacity to include memory effects prevalent in the fluid [4], as well as allowing for the inclusion of external forces or relevant boundary values. Fractional derivative equations may be derived from underlying continuous-time random-walk models and these fractional derivative equations (and their generalisations) are simpler to deal with [5]. This thesis will investigate the ability of fractional derivative equations (and their generalisations) to model long timescale anomalous diffusion phenomena observed in some visco-elastic polymer networks. Recent work [6, 7] has suggested that within certain physical polymer networks there is a tendency for internal stresses to continuously build and dissipate. This phenomenon manifests itself within recordings of probe-particle mean squared displacements at long time scales. The dynamic behaviour of these networks parallels behaviour observed in earthquakes, earning the phenomena the name *gel* or *cyto* quakes [6, 8]. This link suggests that statistical features involved in other stress driven events may provide insight into the modeling of gel quakes. Both *temporal* and *spatial* considerations relevant to these quaking systems will be outlined and modelled within this work.

List of Abbreviations

MSD: Mean Squared Displacement

DWS: Diffusing Wave Spectroscopy

FPE: Fokker-Planck Equation

FFPE: Fractional Fokker-Planck Equation

GFPE: Generalised Fokker-Planck Equation

OU: Ornstein-Uhlenbeck

R(L)HS: Right (Left) Hand Side

PDF: Probability Density Function

RL: Riemann-Liouville

CTRW: Continuous Time Random Walk

CLT: Central Limit Theorem

GLT: General Limit Theorem

PD: Probability Density

PDC: Probability Density Current

LWWR: Lévy Walks With Rests

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1

Outline

Physical gels are characterised by dynamic cross-links which may be broken and reformed based on environmental factors. In recent years it has been observed that physical polymer gel networks can exhibit restructuring events, suggested to arise as internal stresses redistribute within the network [6, 7, 9]. Systems of this kind possess these internal stresses as a result of the network formulation and differing degrees of node binding strengths [6, 10]. These events have been coined *gel quakes* [6] and are driven by the liberation of stresses in the system causing rupturing of the network which subsequently reforms. It is these non-equilibrium events that are the initial motivation for this thesis. This thesis will develop mathematical descriptions of these diffusive behaviours, and outline possible experimentally accessible parameters. Given the diffusion takes part within a visco-elastic system it is expected that Doob's theorem and the central limit theorem will not hold [11]. Both of these theorems have assumptions which are not valid upon the occurrence of temporal correlations, beyond the instantaneous approximations of Markov. A suitable candidate for framing dynamics of this kind is to use the approaches found within the field of generalised diffusion. This field expands upon the classical descriptions of diffusion in terms of the Fokker-Planck equation, by incorporating non-Markovian features. To summarise the intentions of this work we aim to,

- Explore and develop a possible generalised diffusion model for the quaking dynamic observed in some physical polymer gels.
- Incorporate both spatial and temporal features of these *quakes*.
- Provide estimation and predictions for possible experimentally accessible parameters and characteristic features of the diffusive behaviours.

The thesis is structured as follows: Chapter 2 introduces relevant concepts, Chapter 3 introduces the modelling approach in a broad sense, and the latter portion of this Chapter presents the model incorporating the temporal features. Chapter 4 presents the first of two frameworks considered for the incorporation of the spatial features. Finally, Chapter 5 describes a second approach considered for encoding spatial features. The thesis then concludes with an outline of the future possible directions such work may take, considering both experimental and theoretical investigations, some predictions are also made for experimentally accessible parameters.

2

Introduction

2.1 Diffusion

Diffusion is a transport phenomena observed in a large number of natural systems. The phenomena itself refers to the tendency of certain quantities to *flow* down concentration gradients. A range of exotic scenarios can be described within the framework of diffusion such as chemical reactions [12], communication patterns [13] and economics [14] as well as the orbital energy and angular momenta of stars in close proximity to super massive black holes [15]. The mathematical formulation of diffusion is often described in terms of either a random walk [16], Langevin equation [17] or deterministic diffusion equation [18]. These formulations will be briefly outlined in this Chapter and further details will be provided regarding the random walks and the deterministic diffusion equation due to the particular relevance they have to the present work.

2.1.1 Historical Account

As early as 1785 the Dutch physicist Jan Ingenhousz observed the small flickering of coal dust particles on the surface of alcohol. The Scottish botanist Robert Brown observed [19] a similar pattern of irregular motion in his study of small pollen grains suspended in water. It was at a similar time that the famed mathematician Joseph Fourier formulated the heat conduction equation [20], which provided the basis for A. Fick to establish the diffusion equation in 1855 [18]. The erratic motion of Brown and Ingenhousz was formulated mathematically through the work of Christian Wiener in 1863 [21]. The work of Wiener was validated by the experimental findings of Guoy, thus establishing the kinetic theory explanation Wiener provided. It was Albert Einstein who first unified the stochastic description of Wiener with the deterministic diffusion equation by his derivation of the following relationship [22],

$$D = \int_0^{\infty} \langle \dot{x}(t_0)\dot{x}(t_0 - t)\rangle dt. \tag{2.1}$$

Eq. (2.1) relates the diffusion coefficient (associated with the diffusion equation) to the velocity autocorrelation function (ensemble averaged, denoted in this document by the used of the angled brackets $\langle \ldots \rangle$) for the diffusing particle, which in turn relates back to the stochastic force utilised in the approach of Langevin. Jean Baptiste Perrin managed to validate the work of Einstein through his own independent research [23]. Einstein's work also set the scene for the Fokker-Planck, Smoluchowski and Klein-Kramers theories and work later still by Ornstein and Uhlenbeck, Chandrasekhar, Montroll as well as others [24–27]. Still the mathematical treatment of Brownian motion is for the most part due to the work of Norbert Wiener who went on to demonstrate the continuous everywhere, differentiable nowhere (motivating approaches such as the Ito calculus [28]) nature of the Wiener motion (the model of natural Brownian motion). This work led to the understanding of the motion as a fractal or self-similar process in the spatial sense.

2.1.2 Langevin Approach

Langevin opted to describe things in a different way to the probabilistic approach of the diffusion equation, developing instead an approach based on the Newtonian dynamics of particles. His work in 1908 described the equation of motion for the particle, under the influence of a stochastic force. His formulation of the theory marked the beginnings of a field of mathematics known as *stochastic* differential equations [29]. Langevin's equation of motion included a systematic force $-\zeta \frac{d}{dt}x(t)$ attributed to the dissipation due to friction generated by the environment, as well as a rapidly fluctuating random force $F_R(t)$ again arising from the environment (Brownian fluctuation). Thus his equation appears as,

$$m\frac{d^2}{dt^2}x(t) = -\zeta \frac{d}{dt}x(t) + F_R(t), \tag{2.2}$$

where the fluctuating random force, $F_R(x)$ has the following properties

$$\langle F_R(t_1)F_R(t_2)\rangle = \Gamma\delta(t_1 - t_2)2,$$

where $\delta(t)$ is the Dirac delta function,

$$\langle F_R(t)\rangle = 0,$$

and $\Gamma = 2m\zeta k_B T$, k_B being Boltzmann's constant, m being the mass of the diffusing object, ζ being the friction coefficient of Langevin and T the temperature. Notably this relationship links the friction coefficient ζ to the strength of the environmental fluctuations and hence represents a manifestation of the famed *fluctuation dissipation theorem* [22]. One of the highlighted short comings of the standard Langevin approach is that it utilises what has been referred to as a local or *Markovian* friction coefficient ζ. The implication of this is that the environment by which friction is generated responds instantaneously to the motion of the particle. A modification to the Langevin equation which accounts for the non-instantaneous response of the system (non-Markovian) may be presented in the following generalised Langevin equation

$$m\frac{d^2}{dt^2}x(t) = -\int_0^t \zeta(t-t')\frac{d}{dt'}x(t')dt' + F_R(t). \tag{2.3}$$

This equation was also derived in the work of Zwangzig and Nordholm [30], as well as in the model by Caldeira and Legget [31]. The difference in the generalised Langevin approach is that the dissipation term depends on the past. The generalised Langevin equation aligns closely with the standard Langevin equation in the instance where the timescale of the memory kernel $\zeta(t)$ is much smaller than any other timescales of the system. In this scenario the following local approximation is satisfactory,

$$\int_0^t \zeta(t-t')\frac{d}{dt'}x(t')dt' \approx \zeta\int_0^t \delta(t-t')\frac{d}{dt'}x(t')dt' = \zeta\frac{d}{dt}x(t) \tag{2.4}$$

2.1.3 Fokker-Planck Approach

As mentioned an alternative approach to that of Langevin is to consider the time evolution of the position probability density function (PDF) underlying the diffusive process. This involves the use of deterministic equations such as the Fokker-Planck equation which reduces to the diffusion equation in the absence of an external force. The diffusion equation has been derived a variety of ways, and verified experimentally [32–34]. The equation itself appears as,

$$\frac{\partial}{\partial t}P(x,t) = D\frac{\partial^2}{\partial x^2}P(x,t). \tag{2.5}$$

Where in Eq. (2.5), $P(x,t)$ is the PDF describing the PD at each position x at time t and D is the diffusion coefficient. The solution to this equation (in the one dimensional Cartesian space, for an initial Dirac delta profile in x and $P(\pm\infty,t) = 0$) is of the following Gaussian form. The process it describes is often referred to as *Gaussian (or normal) diffusion*

$$P(x,t) = \frac{1}{\sqrt{2\pi Dt}}\exp\left(\frac{-x^2}{2Dt}\right). \tag{2.6}$$

The second moment of this probability density function may be found via the following relations

$$\int_{-\infty}^{\infty} x^2 P(x,t)dx = -\frac{\partial^2}{\partial k^2}\widehat{P}(k,t)\bigg|_{k=0}. \tag{2.7}$$

In the case of the Gaussian solution we have,

$$\int_{-\infty}^{\infty} x^2 P(x,t)dx = \langle x^2\rangle(t) = 2Dt. \tag{2.8}$$

When the diffusion is unbiased or without external force, the second moment and the mean squared displacement (MSD) are the same. Thus, a characteristic trait of Gaussian (or Fickian) diffusion is the linear time evolution of the second moment or MSD . In contrast, non-Fickian (anomalous) diffusion demonstrates non-linear time dependence for the MSD [35], (however, it is conceivable for the Gaussian form to still remain) [36]. Fickian diffusion brings with it further properties such as being *Stationary* and *Markovian*. We now explain these properties, whilst also deriving the previously mentioned *Fokker-Planck equation*. This equation describes diffusion in the presence of drift due to outside forces, and will set the stage for later comparison to the generalised forms more suited to the description of anomalous diffusion.

Continuous Stochastic Processes

The mathematical modeling of diffusion phenomena may be constructed using *stochastic processes*.

> **Definition 2.1.1.** A function $X(t)$ is defined to be a continuous time stochastic process if X is a stochastic variable and,
>
> $$\{X(t); t \in T\}$$
>
> where t denotes time, and the set T is not *finite* nor *countable*.

Let $f(t)$ to define a continuous time stochastic process, as in Definition 2.1.1 [37]. The *state space* of the process $f(t)$ is the entirety of values accessible to the process $f(t)$. The statistical properties of the process may be specified by an infinite set of multiple time-joint probability densities, which describe the *chain* of events occurring the in process. Take the following three event chain as an example,

$$P\big(f_3, t_3; f_2, t_2; f_1, t_1\big) df_3 df_2 df_1. \tag{2.9}$$

which provides the probability of the three events occurring *at* the three particular times provided. The multiple time-joint probability density function may be expressed as a product of a conditional PDF and an $n-1$ joint-time PDF as follows

$$P\big(f_3, t_3; f_2, t_2; f_1, t_1\big) = P\big(f_3, t_3 \big| f_2, t_2; f_1, t_1\big) P\big(f_2, t_2; f_1, t_1\big) \tag{2.10}$$

Markov Processes

A *Markov process* is a stochastic process in which the *memory* is restricted, thus the conditional probability density function is influenced only by the event immediately preceding the current event. If we take the previous three event example we have

$$P\big(f_3, t_3 \big| f_2, t_2; f_1, t_1\big) = P\big(f_3, t_3 \big| f_2, t_2\big). \tag{2.11}$$

Stationarity

Moreover, a stochastic process may be *stationary* such that the statistical properties of the process are time independent. As an example we have,

$$P(f_2, t_2 | f_1, t_1) = P(f_2, t_2 - t_1 | f_1). \tag{2.12}$$

Thus, the conditional PDF depends on the time difference between events rather than the times the events occur.

Chapmann-Kolmogorov Equation

The joint-time PDF describing the chain of events in the instance of a stationary Markovian process may be written as follows

$$P(f_3, t_3; f_2, t_2; f_1, t_1) = P(f_3, t_3 | f_2, t_2) P(f_2, t_2 | f_1, t_1) P(f_1, t_1).$$

Integrating both sides over the intermediate event f_2 removes the dependence of the left hand side on that event occurring. Rearranging and employing the stationarity leaves us with,

$$P(f_3, t_3 - t_1 | f_1) = \int P(f_3, t_3 - t_2 | f_2) P(f_2, t_2 - t_1 | f_1) df_2 \tag{2.13}$$

Eq (2.13) is known as the Chapmann-Kolmogorov equation or *Chain equation*.

Master Equation

In its current state the chain equation is non-linear in the conditional PDFs, however, in this derivation we follow along the path outlined by [38] in order to produce, ultimately, a more manageable equation. The approach begins by tidying up the notation, setting $f = f_3$, $t = t_3 - t_1$, $f' = f_2$, $f_0 = f_1$ and $t' = t_2$, the chain equation is then,

$$P(f, t | f_0) = \int P(f, t - t' | f') P(f', t' | f_0) df'. \tag{2.14}$$

If we consider the time difference to be small such that, $t - t' = \delta t << 1$ then we can approximate $P(f, \delta t | f')$ with a Taylor expansion about $\delta t = 0$,

$$P(f, \delta t | f') \approx P(f, 0 | f') + \delta t \left(\frac{\partial P(f, \delta t | f')}{\partial \delta t} \right) \Bigg|_{\delta t = 0} \quad \text{for} \quad \delta t << 1. \tag{2.15}$$

Inserting a Dirac delta function as the initial condition,

$$P(f, 0 | f') = \delta(f - f') \tag{2.16}$$

and defining

$$W(f, f') = \left(\frac{\partial P(f, \delta t | f')}{\partial \delta t} \right) \Bigg|_{\delta t = 0}. \tag{2.17}$$

Normalising Eq. (2.15) over f yields,

$$\int P(f, \delta t | f') df = 1 + \delta t \int W(f, f') df + \delta t \int a_0(f) \delta(f - f') df = 1. \tag{2.18}$$

The term involving $a_0(f)$ is inserted to satisfy the normalisation condition. Through rearranging Eq. (2.18) $a_0(f)$ can be identified as,

$$a_0(f) = \int W(x', f) dx'. \tag{2.19}$$

x' playing the role of the integration variable representative of the other states, shortly we will set $x' = f'$ when appropriate. Thus we now have,

$$P(f, \delta t | f') = (1 - \delta t a_0(f)) \delta(f - f') + \delta t W(f, f') \tag{2.20}$$

Plugging this back into the chain equation, we have

$$\begin{aligned} P(f, t | f_0) &= \int \left((1 - \delta t a_0(f)) \delta(f - f') + \delta t W(f, f') \right) P(f', t' | f_0) df' \\ &= (1 - \delta t a_0(f)) P(f, t' | f_0) + \int \delta t W(f, f') P(f', t' | f_0) df' \\ \frac{P(f, t' + \delta t | f_0) - P(f, t' | f_0)}{\delta t} &= - \int W(f', f) df' P(f, t' | f_0) + \int W(f, f') P(f', t' | f_0) df' \end{aligned} \tag{2.21}$$

Taking the limit $\delta t \to 0$ and using the definition,

$$\frac{\partial}{\partial t} P(f, t'|f_0) = \lim_{\delta t \to 0} \frac{P(f, t' + \delta t|f_0) - P(f, t'|f_0)}{\delta t} \tag{2.22}$$

after setting $t' = t$, we have

$$\frac{\partial P(f, t|f_0)}{\partial t} = \int W(f, f')P(f', t|f_0) - W(f', f)P(f, t|f_0)df', \tag{2.23}$$

where Eq. (2.23) is known as a *Master equation*, with $W(f, f')$ defining the flow of probability associated with the process transitioning from event f' to f. The equation represents a probability balance equation. The name arises from its first occurrence [39], where it was employed as a general equation from which more specific results were derived. We are now closing in on the final Fokker-Planck equation structure and have already employed the properties of both *Markovian* and *Stationary* processes. The final few steps begin with an expansion known historically as the Kramers-Moyal expansion [40]. In order to prepare the master equation for expansion via the Kramers-Moyal expansion, first the transition probability $W(f, f')$ is expressed as a function of the starting *state* and the size of the *jump* between states such that $W(f, f') \to W(f', \Delta f)$ where $\Delta f = f - f'$. With these alterations, and the substitution $f' = f - \Delta f$, the master equation (Eq. (2.23)) becomes,

$$\frac{\partial P(f, t|f_0)}{\partial t} = \int W(f - \Delta f, \Delta f)P(f - \Delta f, t|f_0) - W(f, \Delta f)P(f, t|f_0)d\Delta f. \tag{2.24}$$

The Kramers-Moyal expansion represents a Taylor series expansion of $P(f - \Delta f, t|f_0)$ about the state f (for details see Appendix A), which results in

$$\frac{\partial P(f, t|f_0)}{\partial t} = \sum_{n=1}^{\infty} \frac{(-1)^n}{n!} \frac{d^{(n)}}{df^{(n)}} \Big[\mu_{W,n}(f) \ P(f, t|f_0) \Big]. \tag{2.25}$$

where, the moments are found in the usual way via,

$$\mu_{W,n}(f) = \int_{-\infty}^{\infty} (\Delta f)^n W(f, \Delta f) d\Delta f. \tag{2.26}$$

2.1.4 Fokker-Planck Equation

The first two terms of the expansion in Eq. (2.25) represent *drift* and *diffusion* terms and typically for most situations it is a valid approximation to neglect higher order terms in Δf. In a more rigorous sense this approximation is valid due to Pawula's theorem which asserts that if any of the even moments $\mu_{W,2l}$, where $l \geq 1$, vanish then *all* the moments $\mu_{W,n}$ for $n \geq 3$ vanish and the expansion terminates after the second term [41]. A consequence of Pawula's theorem is that there are only three possibilities of the Kramers-Moyal expansion,

- $\mu_{W,2} = 0$, the expansion then terminates after the first term. This corresponds to a situation of drift only.
- One of the higher order even moments vanishes such that the expansion terminates after the second term
- The expansion does not terminate at any finite order and it remains an infinite series.

The second of these scenarios is the most significant to physical studies. A whole class of processes known as *Diffusion Processes* are described by Kramers-Moyal expansions that terminate after the second term. In such scenarios the expression takes the following form,

$$\frac{\partial P\left(f,t\middle|f_0\right)}{\partial t} = -\frac{\partial}{\partial f}\left[\mu_{W,1}(f)\ P\left(f,t\middle|f_0\right)\right] + \frac{1}{2}\frac{\partial^2}{\partial f^2}\left[\mu_{W,2}(f)\ P\left(f,t\middle|f_0\right)\right]. \tag{2.27}$$

This equation is known as the *Fokker-Planck equation*, perhaps one of the most widely used forms of the master equation in the context of continuous Markov processes.

Classifications of the Fokker-Planck Equation

There are several ways to classify the various forms of the FPE that may be generated [42]. The equations may be *linear* or *non-linear*, depending on the nature of the moments, $\mu_{W,n}$. Specifically if the moments are functions of the PDF the equation is non-linear. There also exists a classification based on the so-called *dimension of non-linearity*, d which is employed when the moments mentioned depend not on the PDF but on a number, n of it's moments, implying a non-linear FPE with dimension $d = n$ (the previous situation of dependence on the PDF implies a non-linear dimension $d = \infty$). It is also clearly useful to distinguish between univariate and multivariate

FPE's, as well as to distinguish between FPE's subjected to different boundary conditions. Non-linear FPE's may be derived in variety of ways and are often regarded as N-dimensional linear FPE's describing many body systems involving N sub-systems in the limit as $N \to \infty$ [43].

2.1.5 Applications of the Fokker-Planck Equation

Nonlinear FPE's have been applied across a great number of scenarios typically where collective phenomena are of importance. These equations have seen application in surface physics, studying growth and roughening phenomena [44, 45]. Wetting processes on surfaces have also been described utilising non-linear FPE's [46]. Within the field of plasma physics non-linear FPE's have also arisen as useful tools to describe the evolution of the velocity distribution for plasma particles [47–50] (transient and stationary solutions have been studied) and to describe the distribution of bunch particles within particle beams [51–54] among other phenomena [55, 56]. In terms of non-linear hydrodynamics non-linear FPE's have been used to describe flows in porous media [57, 58] and polymer fluids [59].

2.2 Anomalous Diffusion

The previous section has discussed aspects of what may be considered standard diffusion processes, describing the most prevalent stochastic and deterministic approaches to these systems. Specifically in the instance of a stationary Markovian process a derivation of the Fokker-Planck equation was presented. The standard diffusion equation describes Gaussian diffusion where the PDF solution possesses a second moment which grows linearly with time. When these features are not present, the diffusive behaviour is deemed *anomalous*. Anomalous diffusion is a transport process observed in a large variety of systems, across many length and time scales. As specific examples, it has been observed in charge carrier transport in amorphous semiconductors [60, 61], in flow in porous systems [62], in quantum optics [63, 64], as well as many other systems [65, 66]. Mathematically there are two main avenues for the treatment of anomalous diffusion, which amount to generalisations of the two main approaches to standard diffusion: either to consider the stochastic description afforded by the Langevin equation or to study the deterministic Fokker-Planck equation, where the latter encodes information regarding the evolution in time of the position PDF. Diffusive behaviour is most commonly characterised by the temporal behaviour of the second moment of the PDF [67],

$$\langle x^2\rangle(t) \propto t^{\alpha}. \tag{2.28}$$

The exponent, α is used to distinguish between the characteristic regimes. The behaviour is referred to as sub-diffusive ($\alpha < 1$), Fickian or normal ($\alpha = 1$) and super-diffusive ($\alpha > 1$). In other cases the exponent may vary in time known as *transient* diffusion [67]. All of these diffusive behaviours may be found in systems throughout the physical world [68–70]. One important consequence of the change in temporal behaviour of the second moment (caused by changes in the underlying dynamics) is the change in the form of the PDF from Gaussian to non-Gaussian. Gaussian forms are the expected consequence of the sums of independent identically distributed random variables, by virtue of the central limit theorem. Therefore in the context of anomalous diffusion there must be some fundamental features present which invalidate the use of the central limit theorem (CLT). However, before the discussion of these concepts, we first highlight the foundations of the CLT.

2.2.1 Central Limit Theorem

Consider the summation,

$$\zeta_n = \frac{\epsilon_1 + \epsilon_2 + \ldots \epsilon_n}{B_n} - A_n, \tag{2.29}$$

where ϵ_n represents a sampled random variable and the ϵ occurring in the summation are *independent and identically distributed*. There exists a well known limit theorem, namely the central limit theorem (CLT) which states that A_n and B_n will tend to $\sqrt{n}\mu/\sigma$ and $\sigma\sqrt{n}$, respectively for large n [71, 72]. Where μ and σ are the mean and variance of an appropriately scaled Gaussian distribution function. Thus, the Gaussian solution of the standard diffusion equation (Brownian diffusion) represents a consequence of the central limit theorem, in which the displacements represent the stochastic variables. Equivalently given a summation of the form above, one may inquire as to under what conditions the distribution functions underlying the random variables ϵ will combine to a stable distribution of ζ_n (see Definition 2.2.1). The conditions of convergence to a stable distribution are referred to as the *Domain of Attraction* for said stable distribution. The domain of attraction of the Gaussian distribution is outlined in Theorem 2.2.1.

Theorem 2.2.1. *Domain of Attraction for the Normal Distribution:*

If we consider the distribution function $F(x)$ to be the limit distribution of the normalised sum

$$\zeta_n = \frac{\epsilon_1 + \epsilon_2 + \ldots \epsilon_n}{B_n} - A_n, \tag{2.30}$$

where the ϵ represent independent identically distributed random variables. For suitably chosen constants $A_n, B_n > 0$ [73]. The distribution function $F(x)$ belongs to the domain of attraction of the normal distribution if the following relationship holds as $X \to \infty$

$$\frac{X^2 \int_{|x|>X} dF(x)}{\int_{|x|<X} x^2 dF(x)} \to 0. \tag{2.31}$$

This feature of the underlying distribution functions is the essence of the central limit theorem [73]. Within this domain of attraction A_n and B_n tend to $\sqrt{n}\mu/\sigma$ and $\sigma\sqrt{n}$, respectively.

Failure to Satisfy the Conditions of the Central Limit Theorem

In the instance of anomalous diffusion the conditions leading to the central limit theorem are not satisfied, which can occur as a consequence of *three* fundamental effects. These effects are known as the *Moses*, *Joseph* and *Noah* effects. The Joseph and Noah effects were outlined by Mandelbrot in his efforts to decompose the nature of anomalous diffusion into its root causes [74]. His work demonstrated the dominant causes to be either long time increment correlation (the Joseph effect) or due to increment distributions having heavy tails (the Noah effect). The Moses effect has been identified more recently in the work of Chen *et al.* [75], this effect is defined as the increment distributions being non-stationary. A somewhat recent piece of research carried out by Meyer *et al.* [76] discussed the relationship between the so-called Joseph, Noah and Moses exponents (defined within) and the well known Hurst exponent [77]. In carrying out this work they probed the diffusive behaviour of the modified Pomeau-Manneville map [78] both with analytic results and simulation. The results illustrated that the dominant exponent varied, which implied that the cause of the anomalous diffusion of this artificial system varied over various exponent values, a phenomena that may occur amongst natural systems as well.

2.2.2 Generalised Central Limit Theorem

Due to the breakdown of the CLT anomalous diffusion relies on another limit theorem namely, the Lévy-Gnedenko generalised limit theorem [73], (for the domain of attraction to a general stable law see Theorem 2.2.2). The theorem removes the necessity of zero mean and finite variance on the sequence of random variables, the result of which is the convergence to an α-stable distribution (see Eq. (2.35) and the convergence is not confined to the Gaussian distribution [79].

Stable Distributions

Definition 2.2.1. A stochastic variable X is considered to be *stable* (or Lévy α-stable [80]) in the broad sense if the following relationship holds for some positive constants, $a, b, c, g \in \mathbb{R}$

$$aX_1 + bX_2 \stackrel{d}{=} cX + g, \tag{2.32}$$

where X_1 and X_2 are independent copies of the stochastic variable X. If the above relationship is satisfied with $g = 0$ the variable is deemed *strictly* stable.

While stable distributions may also be defined in other equivalent ways, the above definition relies on the equivalence in distribution, $\stackrel{d}{=}$. Perhaps the most concrete way to characterise stable distributions is through the associated characteristic function of the distribution. If we have a probability density function, $P(x)$ the characteristic function is the Fourier transform of $P(x)$, denoted $\widehat{P}(k)$ (see Appendix C). This characteristic function is defined as,

$$\widehat{P}(k) = \langle e^{-ikx} \rangle = \sum_{n=0}^{\infty} \int_{-\infty}^{\infty} \frac{(-ikx)^n}{n!} P(x) dx = \sum_{n=0}^{\infty} \frac{(-ik)^n}{n!} M_{x,n} \tag{2.33}$$

$$M_{x,n} = \int_{-\infty}^{\infty} x^n P(x) dx \tag{2.34}$$

As can be seen the characteristic function represents a generating function for the moments of the distribution. The definition of a stable distribution in terms of a general characteristic function typically appears in the following form, known as the *canonical* form [73].

$$\ln \widehat{P}(k) = -|k|^{\alpha} \{1 + i\beta \frac{k}{|k|} \omega(k, \alpha)\}, \quad -1 \leq \beta \leq 1 \quad \text{and} \quad 0 < \alpha \leq 2 \tag{2.35}$$

where ω is given by

$$\omega(k,\alpha) = \begin{cases} \tan\left(\frac{\pi}{2}\alpha\right) & if \quad \alpha \neq 1 \\ \frac{2}{\pi}\log|k| & if \quad \alpha = 1 \end{cases} \tag{2.36}$$

Stable distributions are of note because they allow for features such as skewness and heavy tails to be incorporated into the probability density function. Aside from the generalised central limit theorem, one of the main reasons to use stable distributions for the statistical description of natural phenomena is the simple fact that there is a wide range of empirical evidence supporting it [81, 82].

2.2.3 Mathematical Models for Anomalous Diffusion

As alluded to in the initial outline regarding anomalous diffusion, there are two main branches of mathematical investigation into anomalous diffusion, the stochastic realisation (Langevin) and the deterministic ensemble (Fokker-Planck). Within these branches there are a variety of approaches to the modeling of anomalous diffusion (in the presence or absence of an external force field). Historically, the most prevalent have been, fractional Brownian motion [83], generalised diffusion equations (in the sense of O'Shaughnessy) [84], continuous time random walks (CTRW) [85–89], Langevin equations [90–92], generalised Langevin equations (GLE) [93, 94], or generalised master equations [95, 96]. For anomalous diffusion only the CTRW and GLE capture the combination of system memory and consideration of the form of the relevant PDF(s) underlying the dynamics. However, in the CTRW framework it is not straightforward to incorporate external force fields, boundary problems or to consider phase space dynamics. An alternative approach to those mentioned has garnered significant attention over the past few decades [97], the approach is to make use of fractional derivative equations. Fractional derivative (or generalised derivative) equations allow for the simple incorporation of boundary value problems as well as external fields, moreover even in the original works it was noted that these fractional derivative (or generalised) operators *naturally* account for memory effects which are associated with many complex systems.

Theorem 2.2.2. *Domain of Attraction to a Stable Distribution:*

The wider class of distribution functions of which are limit distributions for the infinite sum of Eq. (2.29) are known as stable distributions. In order that the distribution function $F(x)$ belongs to the domain of attraction for a stable law with a characteristic exponent $0 < \alpha < 2$ (see Eq. (2.33)) it is necessary and sufficient that [73][a]:

$$\frac{F(-x)}{1-F(x)} \to \frac{c_1}{c_2} \quad \text{as} \quad x \to \infty, \tag{2.37}$$

and for every constant, $k > 0$

$$\lim_{x\to\infty} \frac{1-F(x)+F(-x)}{1-F(kx)+F(-kx)} = k^{\alpha} \quad \text{as} \quad x \to \infty \tag{2.38}$$

The form of B_n in Eq. (2.29) must satisfy the following conditions:

$$nF(B_n x) \to \frac{c_1}{|x|^{\alpha}} \quad (x < 0), \tag{2.39}$$

$$n(1-F(B_n x)) \to \frac{c_2}{x^{\alpha}} \quad (x > 0), \tag{2.40}$$

$$\lim_{\epsilon\to 0}\lim_{n\to\infty} n\left\{\int_{|x|<\epsilon} x^2 dF(B_n x) - \left(\int_{|x|<\epsilon} x dF(B_n x)\right)^2\right\} = 0. \tag{2.41}$$

The form of A_n in Eq. (2.29) takes on one of the following forms, dependent on the value of α (see Eq. (2.33)):

$$A_n = \frac{n}{B_n}\int_{-\infty}^{\infty} x dF(x) \quad \text{for} \quad 1 < \alpha \leq 2 \tag{2.42}$$

$$A_n = n \, \mathrm{Im}\left\{\ln f\left(\frac{1}{B_n}\right)\right\} \quad \text{for} \quad \alpha = 1 \tag{2.43}$$

where f is the characteristic function of F.

$$A_n = 0 \quad \text{for} \quad \alpha < 1 \tag{2.44}$$

The convergence of these underlying distributions to a stable distribution, forms the essence of the generalised limit theorem and provides a basis for anomalous diffusion.

[a] Specifically, see page 175, with details of A_n highlighted in the footnote.

2.3 Fractional Calculus

While fractional calculus has existed as long as integer order calculus, the applications of fractional calculus were not immediately apparent. A common difficulty of fractional (or more accurately *arbitrary* [98]) order calculus is the lack of a simple geometric interpretation, which is present with the integer order counterpart. Indeed, many have attempted to capture the geometric essence of these operators [99] though a consensus on the matter is, for the most part, unavailable. Perhaps the most widely utilised forms of the fractional operators in the temporal sense, are the Riemann-Liouville (RL) and Caputo operators. The RL fractional derivative follows from generalising Cauchy's repeated integral formula whilst the Caputo form follows from generalising the Laplace transform (see Appendix C) expression for the derivative. The RL and Caputo derivatives may be defined as follows [99].

Definition 2.3.1. The fractional derivative of Riemann and Liouville, for arbitrary order α, applied to the function $f(t)$ is defined as

$$ {}_{0}^{RL}D_t^{\alpha} f(t) = \frac{1}{\Gamma(n+1-\alpha)} \left(\frac{d}{dt}\right)^{n+1} \int_0^t (t-t')^{n-\alpha} f(t') dt', \tag{2.45} $$

for $n \leq \alpha < n+1$ where $n \in \mathbb{Z}^{0+}$.

Definition 2.3.2. The fractional derivative of Caputo, for arbitrary order α, applied to the function $f(t)$ is defined as

$$ {}_{0}^{C}D_t^{\alpha} f(t) = \frac{1}{\Gamma(n-\alpha)} \int_0^t \frac{f^{(n)}(t')}{(t-t')^{1+\alpha-n}} dt', \tag{2.46} $$

for $n-1 \leq \alpha < n$ where $n \in \mathbb{Z}^{0+}$.

There is an intimate connection between these two formalisms which can be made apparent through the following manipulation. If $\alpha = 1-\beta$ and $0 < \beta < 1$, then $n = 0$ and

$$ {}_{0}^{RL}D_t^{1-\beta} f(t) = \frac{1}{\Gamma(\beta)} \frac{d}{dt} \int_0^t \frac{f(t')}{(t-t')^{1-\beta}} dt'. \tag{2.47} $$

Due to the convolution theorem, after Laplace transformation of Eq. (2.47), it appears as

$$\mathscr{L}\left[\frac{1}{\Gamma(\beta)}\frac{d}{dt}\int_0^t \frac{f(t')}{(t-t')^{1-\beta}}dt'\right] = \frac{1}{\Gamma(\beta)}u^{1+\beta}\widetilde{F(u)}. \tag{2.48}$$

Applying the Laplace inversion yields,

$$\mathscr{L}^{-1}\left[\frac{1}{\Gamma(\beta)}u^{\beta}\left(u\widetilde{F(u)}\right)\right] = \frac{1}{\Gamma(\beta)}\int_0^t \frac{f^{(1)}(t') + f(0)}{(t-t')^{1-\beta}}dt', \tag{2.49}$$

which is the Caputo fractional derivative plus a power law decaying contribution from the initial conditions [100]. Inspection of the form of the Caputo fractional derivative in Eq. (2.49) reveals that it generates an infinite sum of weighted infinitesimal contributions of the process over $[0, t]$. This integral represents the *memory* of the system and is of great importance to non-Markovian processes. Thus far we have encountered two formulations of the fractional derivative operators. In recent times many more generalisations have begun to emerge, a selection of these may be connected with a so-called memory Kernel embedded within the RL fractional derivative form [101]. Whilst others, that present entirely new forms, still are referred to as fractional derivatives [102].

2.3.1 Strict and Wide Sense Fractional Derivatives

Ortigueira and Machado presented an article in 2015 that was designed to directly address the *issues* with the number of generalisations emerging, the paper was titled "What is a fractional derivative" [103]. Their discussions outline the lack of any concrete framework from which fractional derivatives may be categorised, they present some historic discussions on the matter by Bertran Ross in 1975. His criteria (^{1}**P**) consisted of the following key points.

- ^{1}**P1** The derivative of an analytic function is analytic.
- ^{1}**P2** when the order is integer the fractional derivative reduces to the ordinary derivative. (Backwards compatibility)
- ^{1}**P3** The zero order derivative of a function returns the function itself.
- ^{1}**P4** The operator must be linear.

- ^{1}**P4** The index law holds, that is, $D^\alpha D^\beta f(t) = D^{\alpha+\beta} f(t)$ for $\alpha < 0$ and $\beta < 0$.

Ortigueira and Machado observe that these criteria were outlined at a time when many of the existing developments within the field of fractional calculus had yet to occur and with this in mind they extended the criteria above to the sets ^{2}**P** and ^{3}**P**. Their ^{2}**P** are outlined as follows and they refer to these criteria as the *wide sense criterion (WSC)*

- ^{2}**P1** Linearity: The operator should be *linear*.

- ^{2}**P2** Identity: The zero order derivative of a function returns the function itself.

- ^{2}**P3** Backward Compatibility: If the order of the fractional derivative is of integer value the result should align with that of the integer derivative counterpart.

- ^{2}**P4** Index Law: The index law should hold,that is, $D^\alpha D^\beta f(t) = D^{\alpha+\beta} f(t)$ for $\alpha < 0$ and $\beta < 0$.

- ^{2}**P5** Generalised Leibniz Rule:

$$D^\alpha\Big[f(t)g(t)\Big] = \sum_{i=0}^{\infty} \begin{pmatrix} \alpha \\ i \end{pmatrix} D^i f(t) D^{\alpha-i} g(t)$$

The ^{3}**P** criteria are referred to as the *strict sense criterion* (SSC). The SSC has all the same points as the ^{2}**P**,however, ^{3}**P4** becomes the following

- ^{1}**P4** Index Law: The index law should hold,that is, $D^\alpha D^\beta f(t) = D^{\alpha+\beta} f(t)$ for any α and β.

Their paper concludes by discerning whether common fractional derivatives such as the Grunwald-Letnikov, Riemann-Liouville and Caputo fit their criteria, the first two identified as satisfying the SSC and the third the WSC. They also test another so-called fractional derivative, the *conformable* fractional derivative, defined as

$$T_\alpha = \lim_{\epsilon \to 0} \frac{f(t + \epsilon t^{1-\alpha}) - f(t)}{\epsilon}. \tag{2.50}$$

This fractional derivative is identified as satisfying neither the SSC or the WSC and is hence identified as not being a fractional derivative. The conformable derivative was identified again as not being a fractional derivative by Tarasov where he demonstrated the requirement of non-locality for fractional derivatives in his paper "No Nonlocality. No Fractional Derivative" [104]. An additional critique of the conformable derivative was produced by Abdelhakim and Machado in their paper "A critical analysis of the conformable derivative" [105] where they demonstrated the intimate relationship between conformable and integer derivatives using three theorems proved within, thus "trivializing a large number of proofs and results obtained in the context of conformable derivatives". More recently an article titled "Desiderata of Fractional Derivatives" [106] was published providing a differing mathematical description of desired properties for fractional derivatives. This article touches on similar concepts to those listed above, but provides more careful analysis as to the domain of definition. They also make the point of not requiring a generalised Leibniz rule to hold but rather assert that the standard Leibniz rule hold at integer orders. The subject is still heavily discussed with many articles appearing to address the issue [107, 108]. It is, however, suggested that the best stance on these matters is to motivate the use of a generalised derivative in some form by allowing these physical system to provide clues for it's use. Thus, studies on the many different tools available within the field of generalised diffusion are meritable as it is conceivable physical systems exist within which these operators will most appropriately account for the non-Markovian features.

2.3.2 Generalised Diffusion

A branch of anomalous diffusion studies exists known as *Generalised Diffusion* and is concerned with the study of Generalised Fokker-Planck equations (GFPEs). In fact, fractional diffusion exists within the umbrella of this field. Most commonly the generalised derivatives utilised in this field are generalised time derivatives. These GFPEs appear as,

$$\frac{\partial}{\partial t}P(x,t) = \frac{\partial}{\partial t}\int_0^t \mathscr{K}(t-t')\frac{\partial^2}{\partial x^2}P(x,t')dt', \tag{2.51}$$

where $\mathscr{K}(t)$ has been called the memory kernel. The notion of memory is common in the pursuit of mathematical descriptions for anomalous diffusion, having appeared in both Langevin and FPE approaches. As mentioned above, the field of generalised diffusion is built upon diffusion equations with varying memory kernels. This field has it's origins around the early 2000s [109–111], though since that point it has continued to grow. In the years since there have been many differing memory kernels explored based on the Mittag-Leffler function and it's generalisations [112–116] (which have links to the generalised Cattaneo equations [117]). There have also been several forms connected with the notion of *distributed* polynomial orders in the kernel [118–120]. We now take a brief look at two forms,

Caputo-Fabrizio

The so called Caputo-Fabrizio [121] *generalised derivative* is defined with a memory kernel of the following form

$$\mathscr{K}(t) = \frac{\alpha}{1-\alpha} \exp\left(-\frac{\alpha t}{1-\alpha}\right), \tag{2.52}$$

for $\alpha \in [0,1]$. The influence this alteration has on the fractional diffusion process is to induce confined diffusion, aligning with results obtained via the over-damped Langevin equation [122]. The solution and time evolution of the second moment of this function were obtained by Tateishi *et al.* [101]. The documented relationship this operator has with stochastic resetting is also discussed therein.

Atangana-Baleanu

Another example of a non-local operator has following memory kernel [123],

$$\mathscr{K}(t) = bE_\alpha\left(-\frac{\alpha t^\alpha}{1-\alpha}\right), \tag{2.53}$$

for $\alpha \in [0,1]$, where $E_\alpha(t)$ is the Mittag-Leffler function (see Appendix B), which interpolates between the Lorentzian and exponential functions. The diffusive behaviour introduced via this operator is that of transient diffusion transitioning between normal diffusion and subdiffusion. This operator was also explored by Tateishi *et al.* [101].

2.3.3 Fractional Derivative Equations

Models incorporating fractional derivatives began receiving significant attention leading up to the 21st century when their application to anomalous transport processes was widely investigated. During this development a variety of research articles were published relating to fractional relaxation equations [124–127], fractional rheological models [128], fractional diffusion equations

[129, 130], fractional diffusion-advection equations [130, 131], and fractional Fokker-Planck equations [131, 132]. Fractional derivatives in time naturally incorporate memory effects of the same kind observed in nature, thus making them well suited in the treatment of non-Markovian diffusion processes. Moreover, these fractional diffusion equations are related to the hydrodynamic limit of continuous time random walks [133], thus in a sense they account for some of the limitations of CTRW mentioned earlier.

2.3.4 Continuous Time Random Walks

The continuous time random walk (CTRW) theory considers the motion of a *walker* as it progresses through time continuously. The framework was first described by Weiss and Montroll, and has been utilised to describe an enormous variety of stochastic processes [134]. In a CTRW the walker, or object of interest, progresses by taking steps of size x, after which it waits a time t prior to taking the next step. Both variables x and t are distributed according to a probability density function $\Psi(x,t)$. If the sizes of the steps and waiting times are uncorrelated the following expressions hold

$$\lambda(x) = \int_0^\infty \Psi(x,t)dt, \tag{2.54}$$

$$\omega(t) = \int_{-\infty}^\infty \Psi(x,t)dx, \tag{2.55}$$

where $\lambda(x)$ and $\omega(t)$ are the step-length and waiting time probability density functions, respectively. Equally, in the decoupled framework $\Psi(x,t)$ factors into the independent distributions $\lambda(x)$ and $\omega(t)$. From these quantities one may construct an arrival probability density, $\eta(x,t)$ describing the probability density of arriving at various positions x in time t, defined as

$$\eta(x,t) = \int_{-\infty}^\infty \int_0^t \eta(x',t')\lambda(x-x')\omega(t-t')dt'dx' + \delta(x)\delta(t). \tag{2.56}$$

This expression contains two terms, the first describes the probability associated with a walker at x' at time t' having made a jump of length $x-x'$ in the remaining time $t-t'$, summed over all x and causally relevant t. The second term represents the initial conditions here, that at time $t=0$ the walker is at a location defined by $\delta(x)$ (here at $t=0$ we assume the process begins at the origin, more generally we could use $\delta(x-x_0)$). The position PDF, $P(x,t)$ is then defined as the probability density of arriving and remaining at a position x at time t, defined as,

$$P(x,t) = \int_0^t \eta(x,t')\Phi(t-t')dt', \tag{2.57}$$

where $\Phi(t)$ is referred to as the survival PDF which provides the probability density for a waiting time longer than t, defined as

$$\Phi(t) = 1 - \int_0^t \omega(t')dt'. \tag{2.58}$$

Thus, Eq. (2.57) represents the probability density associated with a walker remaining at the position x for the at least the time $t - t'$. At this point it is easier to manipulate the series of equations by passing into the Fourier ($\mathscr{F} : \lambda(x) \to \widehat{\lambda}(k)$) Laplace ($\mathscr{L} : \omega(t) \to \widetilde{\omega}(u)$) space, by virtue of the convolution theorems for these transforms [135]. Transforming Eq. (2.56) and Eq. (2.58), then substituting into the Fourier-Laplace equivalent of Eq. (2.57) gives the following form for the probability density [136],

$$\widehat{\widetilde{P}}(k,u) = \frac{1-\widetilde{\omega}(u)}{u}\frac{1}{1-\widehat{\lambda}(k)\widetilde{\omega}(u)}. \tag{2.59}$$

Power Law Waiting Time Distribution

The functional forms of $\lambda(x)$ and $\omega(t)$ strongly influence the nature of the diffusion process. As an example of this suppose the function, $\omega(t)$ satisfies the power law distribution,

$$\omega(t) \sim \left(\frac{\tau}{t}\right)^{\alpha+1}, as \;\; t \to \infty. \tag{2.60}$$

The Laplace transform of this expression is provided as [137],

$$\widetilde{\omega}(u) \sim 1-(u\tau)^{\alpha}, as \;\; u \to 0. \tag{2.61}$$

Thus, the expression for the position PDF in the Fourier-Laplace space becomes

$$\widehat{\widetilde{P}}(k,u) = \frac{1}{u}\frac{\widehat{P}_0(k)}{(1+\frac{\sigma^2}{(u\tau)^{\alpha}}k^2)} \tag{2.62}$$

$\widehat{P}_0(k)$ being the Fourier transform of the initial condition $P(x,t)|_{t=0}$. Rearranging Eq. (2.62) and taking the inverse Fourier-Laplace transform yields

$$P(x,t) - P(x,0) = K_\alpha \, {}_0D_t^{-\alpha} \frac{\partial^2}{\partial x^2} P(x,t).$$

Taking the derivative of both sides with respect to t yields

$$\frac{\partial}{\partial t} P(x,t) = K_\alpha \, {}_0D_t^{1-\alpha} \frac{\partial^2}{\partial x^2} P(x,t). \tag{2.63}$$

K_α is the generalised diffusion coefficent, we have moved away from the D notation to avoid confusion with the generalised derivative operator. Eq. (2.63) is also known as the Riemann-Liouville fractional diffusion equation [138]. The Riemann-Liouville fractional derivative inserts a memory into the diffusive process connecting the present moment dynamics to dynamics that have occurred in the past. The order of the equation effects the amount of weighting the past receives. By manipulating Eq. (2.63) it may be cast in the following form

$${}_0D_t^\alpha \Big[P(x,t) - P_0(x) \Big] = {}_0D_t^\alpha P(x,t) - \frac{1}{t^\alpha \Gamma(1-\alpha)} P_0(x) = K_\alpha \frac{\partial^2}{\partial x^2} P(x,t). \tag{2.64}$$

This equation highlights the power law decay of the initial conditions, rather than only influencing the moment, $t = 0$. There are various approaches amongst the literature to solving fractional diffusion equations involving analytic or numerical approximations [139–143]. Historically solutions have been presented in terms of the Fox H-function (see appendix D), and this result is usually arrived at via inversion of both the Fourier and Laplace transforms. In certain examples amongst the literature a Mellin transform [144, 145] has been employed (see appendix C). As an example of the type of closed form solutions obtained in this fashion, consider the following Fox H-function,

$$P(x,t) = \frac{1}{\sqrt{4K_\alpha t^\alpha}} H_{1,1}^{1,0} \left[\frac{|x|}{\sqrt{K_\alpha t^\alpha}} \middle| \begin{matrix} (1-\alpha/2, \alpha/2) \\ (0,1) \end{matrix} \right]. \tag{2.65}$$

The second moment for this function evolves sub-diffusively as follows,

$$\langle x^2 \rangle(t) = \frac{2K_\alpha}{\Gamma(1+\alpha)} t^\alpha, \tag{2.66}$$

where if $\alpha \to 1$ the usual Gaussian statistics are obtained in terms of both $P(x,t)$ and $\langle x^2 \rangle$(t).

Lévy Jump Length Distribution

By modeling the jump length PDF using a Levy distribution it is possible to obtain the following form in the Fourier space,

$$\widehat{\lambda}(k) = \exp\left(-\frac{\sigma^{\mu}}{\tau}|k|^{\mu}\right), \tag{2.67}$$

with the following hydrodynamic limit approximation being valid,

$$\widehat{\lambda}(k) \sim 1 - \sigma^{\mu}|k|^{\mu} \quad as \quad k \to 0 \tag{2.68}$$

Inserting this expression into the Fourier-Laplace space equation for $P(k,u)$ gives us

$$\widehat{\widehat{P}}(k,u) = \frac{\widehat{P}_0(k)}{\left(u + \sigma^{\mu}|k|^{\mu}\right)}.$$

Laplace inversion of this provides the characteristic function discussed earlier for the Gnedenko generalised limit theorem. Alternatively, rearranging the expression before evaluating the double inversion leaves the following,

$$\frac{\partial}{\partial t}P(x,t) = K_{\mu}\ {}_{-\infty}D_x^{\mu}P(x,t),$$

where ${}_{-\infty}D_x^{\mu}$ is the Riesz-Feller [146] fractional derivative which aligns with the Caputo fractional derivative with the lower limit in the integral set to $-\infty$, and K_{μ} is the generalised diffusion constant. The use of the space fractional derivative introduces a connectedness in space that portrays the influence surrounding regions have on the dynamics of a given region. One of the short comings of this approach, or one of the challenges, is that the second moment under this regime is divergent. This has motivated some to consider alternative moments such as the fractional,

$$\int_{-\infty}^{\infty} x^{\mu}P(x,t)dx = \langle x^{\mu} \rangle(t). \tag{2.69}$$

Although the physical interpretation of a moment of this kind becomes a bit unclear. An alternative approach is to define the second moment through time dependent integral limits, such that it is considering the dynamics within a growing region,

$$\int_{L_1 t^{1/\mu}}^{L_2 t^{1/\mu}} x^2 P(x,t) dx = \langle x^2 \rangle_L(t) \tag{2.70}$$

It is possible to extend this model to incorporate space and time fractionality, however, this doesn't resolve the non-existence of the second moment.

Relationship Between the Memory Kernel and the Waiting Time Probability Density Function

Mentioned earlier was the structure of a general fractional derivative which can be defined as follows

$$\mathbf{G}_\alpha f(x,t) = \frac{\partial}{\partial t} \int_0^t f(x,t') \mathscr{K}_\alpha(t-t') dt', \tag{2.71}$$

where $\mathscr{K}_\alpha(t)$ is referred to as the memory kernel of this generalised fractional derivative. As was mentioned earlier it is known amongst the literature that GFPEs may be related to CTRW based approaches. Consider the generalised diffusion equation

$$\frac{\partial}{\partial t} P(x,t) = K_\alpha \mathbf{G}_\alpha P(x,t). \tag{2.72}$$

The Fourier-Laplace space solution becomes,

$$\widetilde{\widehat{P}}(k,u) = \frac{P_0(k)}{u(1 + K_\alpha \widetilde{\mathscr{K}}(u) k^2)}. \tag{2.73}$$

In the article produced by Tateshi *et al.* [101] they explored the role fractional derivative operators play in diffusion. One of the results they provided will be presented now, as it further identifies the link between CTRW processes and FDE. Firstly we must recall the Fourier-Laplace expression identified previously from the decoupled CTRW approach,

$$\widetilde{\widehat{P}}(k,u) = \frac{(1 - \widetilde{\omega}(u))}{u(1 + \widehat{\lambda}(k)\widetilde{\omega}(u))} = \frac{1}{(1 + K_\alpha \widetilde{\mathscr{K}}(u) k^2)} \tag{2.74}$$

$$\implies \frac{\widetilde{\omega}(u)}{\widetilde{\mathscr{K}}(u)(1 - \widetilde{\omega}(u))} = K_\alpha \frac{k^2}{(1 - \widehat{\lambda}(k))}.$$

After some final rearrangements this leaves us with

$$\widetilde{\omega}(u) = \frac{\frac{\widetilde{\mathscr{K}}(u)}{\tau_c}}{1 + \frac{\widetilde{\mathscr{K}}(u)}{\tau_c}} \tag{2.75}$$

$$\widehat{\lambda}(k) = 1 - K_\alpha \tau_c k^2, \tag{2.76}$$

where τ_c is the so-called characteristic time associated with the waiting time PDF in the underlying CTRW process. Thus, it becomes apparent that the various memory kernels alter the underlying waiting times of the CTRW process and therefore capture differing non-Markovian effects.

2.3.5 Fractional Fokker-Planck Equation from a CTRW

We will now describe a connection highlighted by Barkai *et al.* between the FFPE and the CTRW [5]. Their derivation begins with identifying some of the usual distributions that occur in CTRW theory. They define the survival probability, $\Phi(t)$ in the usual fashion which results in the probability of i jumps occurring in time t. $Q_i(t)$ appears in the Laplace space as

$$\widetilde{Q}_i(u) = \frac{1-\widetilde{\omega}(u)}{u}\widetilde{\omega}(u)^i. \tag{2.77}$$

They then go on to define the probability of finding the particle at site n at time t to be $P(n,t)$ and $P_i(n)$ to be the probability of being on site n after step i, then

$$\widetilde{P}(n,u) = \mathscr{L}\Big[\sum_{i=0}^{\infty} P_i(n) Q_i(t)\Big](u) = \frac{1-\widetilde{\omega}(u)}{u}\sum_{i=0}^{\infty} P_i(n)\widetilde{\omega}(u)^i. \tag{2.78}$$

The recurrence relation for $p_i(n)$ as described in [5] appears as

$$P_{i+1}(n) = R(n-1)P_i(n-1) + L(n+1)P_i(n+1), \tag{2.79}$$

where $R(n)$ and $L(n)$ are the probabilities of the walker moving right or left at the site n [5]. The authors then transition to the continuum limit of the equation for $P_i(n)$ such that $P_i(n) \to P_i(x)$, where it is now a probability density. The two terms of the above equation may be expanded about a point a as follows (using $R(n-1)P_i(n-1)$ as an example)

$$R(x-a)P_i(x-a) = R(x)P_i(x) + \frac{\partial}{\partial x}\big[R(x)P_i(x)\big](-a) + \frac{\partial^2}{\partial x^2}\big[R(x)P_i(x)\big]\frac{a^2}{2} + \dots \tag{2.80}$$

The authors omit terms of higher order and assume the system to be close to thermal equilibrium, permitting them to use the relationships

$$R(x) \approx L(x) \approx 1/2 \tag{2.81}$$

$$R(x) - L(x) \approx \frac{aF(x)}{2k_b T}. \tag{2.82}$$

$F(x)$ being some external force field. Taking the above into account, and inserting the truncated Taylor series forms into Eq. (2.79) results in the following expression,

$$P_{i+1}(x) = P_i(x) + \frac{a^2}{2}\left[\frac{\partial^2}{\partial x^2}P_i(x) - \frac{\partial}{\partial x}\left(\frac{F(x)}{k_b T}P_i(x)\right)\right]. \tag{2.83}$$

Rewriting Eq. (2.78) as (again having transitioned to the continuum limit),

$$\widetilde{P}(x,u) = \frac{1-\widetilde{\omega}(u)}{u}P_0(x) + \frac{1-\widetilde{\omega}(u)}{u}\sum_{i=1}^{\infty} P_i(x)\widetilde{\omega}(u)^i, \tag{2.84}$$

$$\widetilde{P}(x,u) = \frac{1-\widetilde{\omega}(u)}{u}p_0(x) + \frac{1-\widetilde{\omega}(u)}{u}\sum_{i=1}^{\infty}\left[p_{i-1}(x) + \frac{a^2}{2}\left[\frac{\partial^2}{\partial x^2}p_{i-1}(x) - \frac{\partial}{\partial x}\left(\frac{F(x)}{k_b T}p_{i-1}(x)\right)\right]\right]\widetilde{\omega}(u)^i. \tag{2.85}$$

Note that

$$\frac{1-\widetilde{\omega}(u)}{u}\sum_{i=1}^{\infty} p_{i-1}(x)\widetilde{\omega}(u)^{i-1}\widetilde{\omega}(u)^1 = \widetilde{P}(x,u)\widetilde{\omega}(u). \tag{2.86}$$

This gives

$$\widetilde{P}(x,u) = \frac{1-\widetilde{\omega}(u)}{u}p_0(x) + \left[\widetilde{P}(x,u) + \frac{a^2}{2}\left[\frac{\partial^2}{\partial x^2}\widetilde{P}(x,u) - \frac{\partial}{\partial x}\left(\frac{F(x)}{k_b T}\widetilde{P}(x,u)\right)\right]\right]\widetilde{\omega}(u). \tag{2.87}$$

The authors then define the waiting time distribution behaviour for large t to be,

$$\omega(t) \sim \frac{\alpha A_\alpha}{\Gamma(1-\alpha)t^{1+\alpha}} \tag{2.88}$$

as $t \to \infty$, where $\alpha \leq 1$. The Laplace transform of this expression is,

$$\widetilde{\omega}(u) \sim 1 - A_\alpha u^\alpha \quad as \quad u \to 0. \tag{2.89}$$

If $\alpha < 1$ the first moment of $\omega(t)$ will diverge. Inserting this expression into Eq. (2.87) gives us,

$$\widetilde{P}(x,u) = \frac{A_\alpha u^\alpha}{u} p_0(x)$$
$$+\left[\widetilde{P}(x,u) + \frac{a^2}{2}\left[\frac{\partial^2}{\partial x^2}\widetilde{P}(x,u) - \frac{\partial}{\partial x}\left(\frac{F(x)}{k_b T}\widetilde{P}(x,u)\right)\right]\right](1 - A_\alpha u^\alpha). \tag{2.90}$$

The authors then consider the limit as $a \to 0$ noting that in the standard diffusion approximation this limit is only meaningful when the average waiting time *and* the lattice spacing a approach 0. Considering the current discussion includes the possibility of diverging moments the authors assert the following ratio to be finitely bounded whilst a and $A_\alpha \to 0$, this defines K_α within their work,

$$\lim_{a^2 \to 0, A_\alpha \to 0} \frac{a^2}{2A_\alpha} = K_\alpha. \tag{2.91}$$

K_α being the generalised diffusion coefficient we have met previously. Multiplying Eq. (2.90) by $(A_\alpha u)^{-\alpha}$ yields

$$\widetilde{P}(x,u) - \frac{\delta(x)}{u} = K_\alpha u^{-\alpha}\left[\frac{\partial^2}{\partial x^2} - \frac{\partial}{\partial x}\frac{F(x)}{k_b T}\right]\widetilde{P}(x,u), \tag{2.92}$$

which upon Laplace inversion and subsequently taking the partial derivative with respect to t gives us,

$$\frac{\partial}{\partial t}P(x,t) = K_\alpha \, {}_0D_t^{1-\alpha}\left[\frac{\partial^2}{\partial x^2} - \frac{\partial}{\partial x}\frac{F(x)}{k_b T}\right]P(x,t), \tag{2.93}$$

which is the noteworthy *fractional Fokker-Planck equation*.

Generalised Non-Local Fokker-Planck Equation from a CTRW

It is perhaps worth pointing out that based on the hydrodynamic limit relationship between the CTRW waiting time distribution and the generalised memory kernel, as in Eq. (2.71), one may derive a generalised Fokker-Planck equation, this alternative derivation is *not* in the literature. This derivation follows the derivation outlined above, however, presents the waiting time distribution as

$$\widetilde{\omega}(u) = \frac{1}{1 + \frac{\tau_c}{\widetilde{\mathcal{K}}(u)}}. \tag{2.94}$$

Inserting this expression into Eq. (2.87) gives

$$\widetilde{P}(x,u) = \frac{1 - \frac{1}{1+\frac{\tau_c}{\widetilde{\mathcal{K}}(u)}}}{u} p_0(x) + \left[\widetilde{P}(x,u) + \frac{a^2}{2}\left[\frac{\partial^2}{\partial x^2}\widetilde{P}(x,u) - \frac{\partial}{\partial x}\left(\frac{F(x)}{k_b T}\widetilde{P}(x,u)\right)\right]\right]\frac{1}{1 + \frac{\tau_c}{\widetilde{\mathcal{K}}(u)}}. \tag{2.95}$$

Multiplying both sides by $1 + \frac{\tau_c}{\widetilde{\mathcal{K}}(u)}$ gives

$$(1 + \frac{\tau_c}{\widetilde{\mathcal{K}}(u)})\widetilde{P}(x,u) = \frac{\tau_c}{u\widetilde{\mathcal{K}}(u)} p_0(x) + \left[\widetilde{P}(x,u) + \frac{a^2}{2}\left[\frac{\partial^2}{\partial x^2}\widetilde{P}(x,u) - \frac{\partial}{\partial x}\left(\frac{F(x)}{k_b T}\widetilde{P}(x,u)\right)\right]\right]. \tag{2.96}$$

Simplifying and rearranging yields,

$$\widetilde{P}(x,u) - \frac{p_0(x)}{u} = \widetilde{\mathcal{K}}(u)\frac{a^2}{2\tau_c}\left[\frac{\partial^2}{\partial x^2}\widetilde{P}(x,u) - \frac{\partial}{\partial x}\left(\frac{F(x)}{k_b T}\widetilde{P}(x,u)\right)\right], \tag{2.97}$$

defining

$$\lim_{a^2 \to 0,\, \tau_c \to 0} \frac{a^2}{2\tau_c} = K_g. \tag{2.98}$$

Taking the Laplace inverse followed by the partial derivative with respect to t gives

$$\frac{\partial}{\partial t}P(x,t) = K_g \frac{\partial}{\partial t}\int_0^t \mathcal{K}(t-t')\left[\frac{\partial^2}{\partial x^2}P(x,t') - \frac{\partial}{\partial x}\left(\frac{F(x)}{k_b T}P(x,t')\right)\right]dt'. \tag{2.99}$$

Eq. (2.99) may be thought of as a *generalised Fokker-Planck equation*, where K_g is a generalised diffusion coefficient. In fact, non-Markovian FPE similar to this have a history dating back to the early works of Zwanzig. Since the work of Zwanzig, presenting a generalised framework for the non-Markovian FPE, the FFPE has occurred as a manifestation of these early insights. Interestingly, in recent times strong connections between GFPE and subordination approaches have been established and studied. The series of connections in this vein can be traced to the work of Sokolov [147] who with the aid of Chechin [148] established a collection of conditions upon which subordination schemes and generalised FPEs are equivalent.

Waiting Time Probability Density Functions Associated with Stress Redistributing Events

As has been mentioned already, the diffusive behaviour occurring at long timescales in the physical polymer systems we are interested in is suggested to be a *quake*-like phenomena. The study of both natural quaking events as well as simpler systems designed to mimic their features, has been a longstanding scientific field. A result of their widespread study is that many statistical properties of quakes have been documented, and in some cases universalities have been suggested. In terms of the temporal distribution of waiting times between events, it was proposed in 2004 [149] that natural quaking phenomena universally appear to adhere to a generalised Γ distribution. The generalised Γ distribution has the functional form,

$$w(t) = \frac{t^{\gamma-1}}{\tau^{\gamma}\Gamma\left(\gamma/\alpha\right)} \exp\left(-\frac{t^{\alpha}}{\tau^{\alpha}}\right). \tag{2.100}$$

This functional form encapsulates the shorter timescale *correlated* aftershocks as well as the longer timescale main shock behaviour, (the latter becomes Poissonian upon $\alpha \to 1$). It's appearance more broadly in both physical and model systems has been noted since Corral's original article [150–155]. It is therefore suggested that this is a suitable distribution to insert into the CTRW in our own diffusive modelling efforts. Of note also is the work of Hainzyl and Holler [156], who constructed a model network within which stress redistributing events may occur. They observed that the waiting time distribution function actually changed between a handful of functional forms, and that the gamma (or generalised gamma) distribution existed when the value of *stress transfer* was relatively large and the value of the *noise intensity* relatively small. Whilst their model is rather simplistic it does highlight potential trends that may be relevant to physical systems such as polymer gel networks.

2.3.6 Applications of Fractional Calculus

We now discuss some prior research where fractional derivative operators and their generalisations have been employed to describe aspects of polymer systems. Early work in applying fractional calculus to physical systems was motivated by the fact the stress-strain experiments on visco-elastic bodies [128, 157, 158] produced the following empirical law

$$\sigma(t) \propto t^{-\alpha}, \tag{2.101}$$

which represents an inverse power law of the stress function $\sigma(t)$. The use of fractional calculus was associated with the knowledge that a pure Hookean solid possessed the following stress-strain ($\epsilon(t)$, being the strain) relationship

$$\sigma(t) \propto \epsilon(t), \tag{2.102}$$

whilst in a Newtonian fluid the stress was proportional to the first derivative of the strain

$$\sigma(t) \propto \frac{d}{dt}\epsilon(t). \tag{2.103}$$

Thus the following relationship was proposed for visco-elastic materials

$$\sigma(t) \propto \frac{d^\alpha}{dt^\alpha}\epsilon(t), \quad \alpha \in [0,1]. \tag{2.104}$$

Experimental results under the condition of constant strain align with the relationship above if the Riemann-Liouville derivative is employed. Throughout the late 80s and early 90s interest grew in considering the generalisation of the diffusion equation to fractional order derivatives, initially time derivatives. These efforts were related to formulating and solving the Cauchy problem for the fractional diffusion equation. Again this work was motivated through experimental observations in which the mean squared displacement of a diffusing quantity evolves sub-linearly in time as,

$$\langle x^2\rangle(t) \propto t^{\frac{2}{d_w}}. \tag{2.105}$$

The exponent characterising the evolution depends on d_w which is defined to be the anomalous diffusion coefficient where $d_w > 2$ (just another means of characterising diffusion as mentioned in Eq. (2.28)). Early on a combination of theoretical, experimental and computer simulations provided the motivation for some researchers to propose that the PDF $P(x,t)$ satisfies the condition,

$$P(x,t)\Big|_{x=0} \propto t^{\frac{d_s}{2}}, \tag{2.106}$$

where, $d_s = \frac{2d_f}{d_w}$ is the *spectral* dimension. d_f is the Hausdorff dimension [159], providing a link to the fractal nature of the system. Also proposed by the same researchers was the following asymptotic behaviour for the PDF,

$$P(x,t) \approx t^{-d_s/2} e^{-\left(\frac{x}{\sqrt{\langle x^2(t)\rangle}}\right)^{\frac{d_w}{d_w-1}}}, \quad \text{for} \quad x/\sqrt{\langle x^2\rangle(t)} >> 1 \quad \text{as} \quad t \to \infty, \tag{2.107}$$

From these considerations the appropriate differential equation appears as

$$\frac{\partial^{2/d_w}}{\partial t^{2/d_w}} P(x,t) = x^{1-d_s} \frac{\partial}{\partial x} x^{d_s-1} \frac{\partial}{\partial x} P(x,t). \tag{2.108}$$

This equation reduces to standard diffusion in the instance $d_w = 2$. This early fractional diffusion equation was solved in the work of Metzler *et al.* [160] and the solution took the form

$$P(x,t) \propto t^{-d_f/d_w} H^{2,0}_{1,2}\left[\frac{x^d_w}{t} \middle|^{(1-d_f/d_w,1)}_{(1-d_f/d_w,d_w/2),(0,d_w/2)} \right]. \tag{2.109}$$

This result foreshadows the important role Fox H-functions play in solving fractional diffusion equations. Around the early 2000's several researchers [146] had established the link between the continuous time random walks framework of Montrol and Weiss to the fractional diffusion equation. Gorenflo and Mainardi [146] explored the space-time fractional diffusion equation surveying some of the general theory as well as demonstrating that time fractionality was due to an underlying non- Markovian CTRW. In 2007 Dubbeldam *et al.* [161] provided one of the earliest explicit descriptions of polymer dynamics via fractional diffusion in their paper describing polymer translocation through a nanopore. They outlined a PDF $P(s,t)$ describing the probability density associated with a segment of a polymer, s being located in the nanopore at time, t. The evolution of the PDF was provided through the Riemann-Liouville fractional diffusion equation, introduced due to their estimation that the second moment $\langle s^2\rangle(t)$ evolved sub-linearly. They obtained their solution through an eigenfunction summation approach with *reflecting-absorbing* boundary conditions with an initial condition of $P(s,0) = \delta(s - s_0)$. They went on to obtain the solution, then identified the first- passage time and compared it with Monte-Carlo simulation results, which demonstrated good agreement. They also suggested their scheme could be extended to include drag forces acting on the polymer strand.

Again in 2007 Li *et al.* [162] described the process of drug release from a polymer matrix through fractional diffusion. Their research followed on from their earlier work (2004) [162] which represented the first time this approach had been utilised, however, didn't consider it's application to polymer matrix systems. They described the process through a moving boundary scenario, whereby the boundary represented the diffusion front. They described the concentration $C(x,t)$ through a space-time fractional diffusion equation. They were also interested in finding the form of the underlying flux of diffusion from a generalised non-local Fick's law relationship. They solved the fractional diffusion equation in a dimensionless form and identified the form of the dimensionless flux in several different cases.

The FPE with the inclusion of a sink term has been used frequently to describe reaction processes. In 2008 Seki *et al.*[163] extended this approach to the instance of non-Markovian processes by the inclusion of the Riemann-Liouville fractional derivative. They looked predominantly at the behaviour of the survival PDF defining the probability density associated with a *particle* remaining in the described space. They considered their system contained within a harmonic potential surface and that the sink be localised. They considered both barrier-less and high activation energy arrangements. In the high activation energy case they demonstrated departure from the Kramers-Smoluchowski picture as exponential decay did not appear.

A slightly more technical paper was published in 2010 by Yu Luchko [164], this paper established a total of five theorems pertaining to the time fractional diffusion equation in the Caputo sense. Beginning with a random walk framework the fractional diffusion equation was first extracted under the usual considerations. With the equation in hand he then progressed to the consideration of some initial boundary value (IBV) problems, ultimately proving a suite of theorems (in some instances the proofs are included in his prior publications), most notably establishing the uniqueness of the solution of the time fractional diffusion equation in the Caputo sense for the given IBV problem. The nature of the IVB problem discussed is rather general and allows for the adaption to a wide variety of physical systems, thus establishing the capacity of this time fractional diffusion equation to provide meaningful results in such settings.

A paper by Basu *et al.* [165] published in 2012 addressed the diffusion of the density of ions in a gelatin-$LiClO_4$ network, over varying concentrations of the lithium salt. Their work was motivated by the recent focus away from polyethylene oxide towards more environmentally friendly polymer electrolytes with gelatin falling into this category. They go on to acknowledge other models which describe the impedance results for the system through construction of an equivalent circuit, they make the observation that models of this nature often have a "considerable number of free parameters" of which "it is difficult to interpret the physical significance of". This observation is of note because a similar approach exists amongst the study of visco-elastic systems and perhaps falls victim to the same shortcomings these authors have put forth. Fractional diffusion models are widely known to benefit from fewer parameters (that may also be experimentally fixed) than many of the mathematical counterparts in the anomalous diffusion realm, especially those of the nature outlined above. The researchers also benefited from the prior adaption of the fractional diffusion framework to the study of impedance spectroscopy of electrolytic cells. Basu *et al.* [166] were able to model impedance data across a wide range of lithium salt concentrations, extracting from these models a selection of model parameters including the Debye length. However, they observed the deterioration of their model in the regime of lithium concentrations > 12.5 (wt/wt), suggesting some additional features have been neglected in their modeling. Nevertheless, the article provides a step away from models with limited interpretability in terms of impedance data, towards a model with a smaller selection of experimentally accessible parameters. Fractional diffusion appeared again in 2016 in the context of polymer systems when it was utilised by Krasnov *et al.* [167] to describe the anomalous diffusion of hydrogen atoms in spider silk under varying external strains. The anomalous diffusion described in this article was detected across picosecond timescales and nanometer length scales, accessible with the employment of incoherent neutron spectroscopy. The modeling incorporated a fractional diffusion equation in the Caputo sense, describing the time evolution of the position PDF. They identified that the generalised diffusion coefficient in this scheme was independent of the scattering vector obtained, over the length scales investigated, they noted that it was monotone increasing (as was the anomalous exponent) with increasing applied tensile strain. Interestingly they note their fractional framework implicitly incorporates both diffusion due to absorbed water and due to the polymer chains themselves. However, these processes must occur at similar temporal regimes such that the diffusion does not become transient but rather remains sub-diffusive, unlike the quaking pectin gel system investigated in this thesis.

2017 saw the application [168] of the fractional diffusion equation to another gel system, the cross-linked polysaccharide gel beads known as *Sephadex*^{TM}. They looked specifically at the diffusion dynamics of water protons utilising magnetic resonance imaging. Their equation was fractional in both time and space, they record the values of both exponents and the generalised diffusion coefficient over $27-200ms$. The space fractionality in the system is shown to rather rapidly return to the standard value of two whilst the time exponent decreased throughout the probed times. They are able to assert that their anomalous exponents and generalised diffusion coefficient capture the porosity and tortuosity of the gel structure in the diffusion dynamics though the relationships between these quantities aren't explicitly provided.

2.4 Pectin Polymer Network Quakes

Pectin denotes a range of polysaccharide structures found abundantly within plant cell walls and plays a key role in the mechanical properties of the wall. Pectin polymer gels form under various conditions, the most abundant form consisting of polymer strands interacting via mediating calcium ions in a structure referred to as the egg box model. A less studied variant was explored in 2013 by Mansel et al. known as an acid induced gel [10]. The key binding between pectin chains in these acid induced gels is hydrogen bonding and they require a more particular pH environment to form. Structurally this polyelectrolytic polysaccharide alternates in solution between a two-fold extended helix and a more compact three-fold helical structure, based on the distribution of charges on the pectin chains. Dynamics (of the polymer network, and of probe microparticles) within this particular type of pectin gel was investigated by diffusive wavelength spectroscopy and multiple particle tracking, ultimately highlighting a particular diffusion phenomena apparent among many other out of equilibrium systems. The diffusion phenomena was studied by measuring the time of decorrelation of scattered light passing through the gel and allowing the behaviour of the mean squared displacements of probe particles to be obtained. The phenomena was discussed briefly within the same article where it was ruled out that the phenomena could be attributed to pore hopping, as had been observed in F-actin networks [169].

This motivated a follow up article to be produced, focusing again on long timescale dynamics, this time in terms of the ion induced gel system. Mansel *et al.* [6] were able to demonstrate that increasing the length of the binding junctions between polymer strands did not guarantee greater stability but rather could also enhance the driving of internal dynamics. Their work also highlighted the long spatial correlations apparent within the internal dynamics which remain correlated over length scales of up to millimeters. This process is not driven by thermal breakage of weak bonds but rather the competition of strong bond formation in multiple locations disrupting the network. Stress redistribution of this kind is believed to be closely tied to the same physical considerations that govern other non-equilibrium dynamics such as earthquakes. Experimental MSD data from quaking pectin gel systems may be seen in Fig. 2.1, where beyond values of t of the order 10^{-1} seconds the emergence of the new diffusive dynamic is revealed.

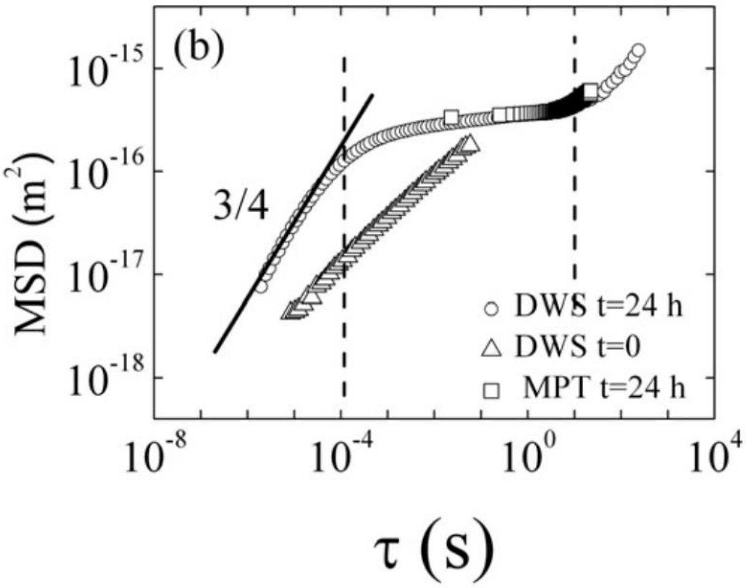

Figure 2.1: The graph displays experimentally obtained values for the MSD, as was published in [7]. Data is obtained by way of either diffusing wave spectroscopy (DWS) or multiple particle tracking (MPT). Also included in the figure is MSD data corresponding to data collected at the time of solution formation ($t = 0$) as well as at a time ($t = 24h$) when the polymer structure has formed. $\tau(s)$ represents the *lag time* measured in seconds, the lag time being the difference between the timestamps of the first and last position measurements.

3

Modelling Quaking Diffusion

In the system of primary interest in this research, the *overall* diffusive behaviour is proposed to arise from *two* separate underlying behaviours. The first is Brownian motion within a *semipermeable* polymer network, whilst the second contribution occurs as the network itself fluctuates as internal stresses redistribute (junctions rupture and reform). By considering the position PDF, $P(x,t)$ associated with diffusion within this system to be the result of these two independent behaviours, $P(x,t)$ may be constructed as,

$$P(x,t) = \int_{-\infty}^{\infty} P_{SR}(x-x',t)P_B(x',t)dx', \tag{3.1}$$

where $P_{SR}(x,t)$ and $P_B(x,t)$ are the stress redistributing and Brownian contributions, respectively.

3.1 Brownian Contribution

In this section the contribution due to Brownian motion within the network is discussed, and a model framework is proposed.

To motivate the model of choice we now outline the key influences acting upon a diffusive process within this system: The polymer strands provide a restoring force to the diffusing particle, and due to the viscoelastic nature of the system the diffusive process should be non-Markovian. The combination of these features can be encoded in a straightforward manner, within the Ornstein Uhlenbeck (OU) process of a FFPE [170]. Thus, we make use of this established framework to provide a description of the Brownian motion in the network, which dominates the short to intermediate timescale behaviour.

This framework has been studied in the past, it amounts to the scenario of diffusion that is impacted by the *past* gradient and restoring forces. In this scheme there exists a potential, $U(x) \propto x^2$. This equation has been studied in some detail dating back to 1984 in the works Weiss [171]. The solution as they demonstrated can be presented in the following way, obtained via separation of variables.

$$P_B(x,t) = \sqrt{\frac{m\omega^2}{2\pi k_B T}} \sum_{n=0}^{\infty} \frac{1}{2^n n!} E_\alpha\left(-\frac{nt^\alpha}{\tau^\alpha}\right) H_n\left(x\sqrt{\frac{m\omega^2}{4\pi k_B T}}\right) H_n\left(x\sqrt{\frac{m\omega^2}{4\pi k_B T}}\right) \exp\left(-\frac{x^2}{2}\frac{m\omega^2}{4\pi k_B T}\right), \tag{3.2}$$

With H_n being the Hermite polynomials and E_α the Mittag-leffler function. Such a PDF relaxes anomalously (with a non-Gaussian profile) from an initial position x_0 towards a Gaussian profile at the origin, this Gaussian profile is the stationary solution to the FFPE in the OU setting. The MSD corresponding to this form is displayed alongside experimental data below in Fig. 3.1, and the model can be seen to capture the first part of the observed data well.

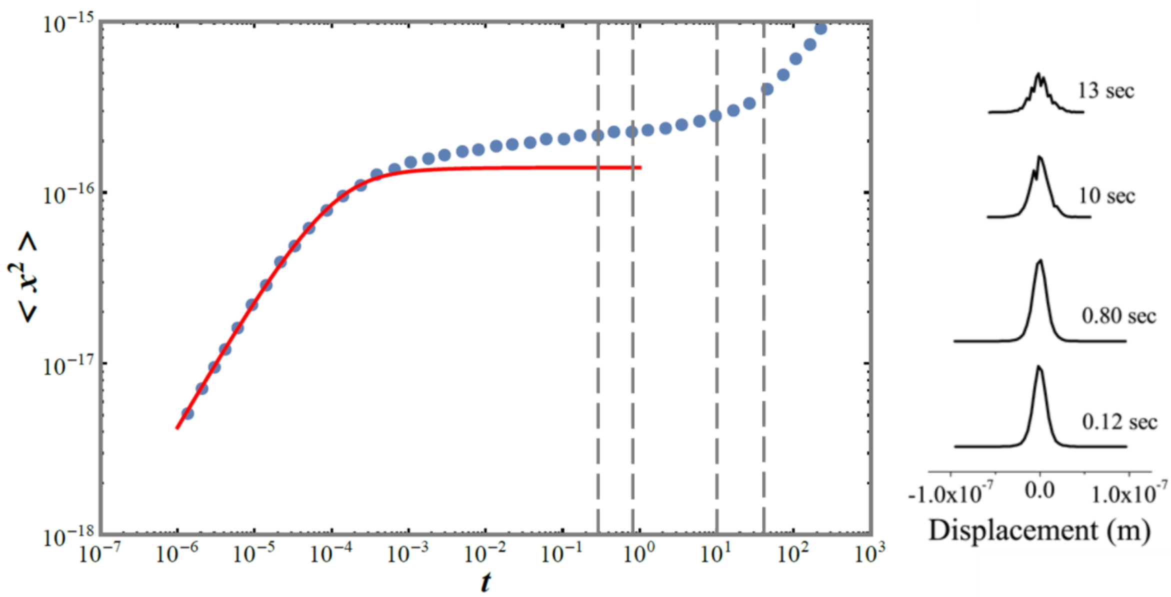

Figure 3.1: Time evolution of the MSD of the FFPE in the OU scenario (red) compared with the experimental MSD data of Mansel *et al.* [7]. The PDF plots are also adapted from the works of Mansel, and the vertical dashed lines indicate the times corresponding to the sampling of the Van Hove plots indicated on the RHS of the figure.

3.2 Stress Redistribution Contribution

This section will cover the diffusive implications of having the network itself restructure as internal stresses are liberated. In order to develop a mathematical description of this behaviour we begin with the CTRW process. The CTRW framework has been outlined in the introduction, and Eq. (2.59) represents a common starting point when constructing anomalous diffusion models. By altering the functional forms of the distributions, $\tilde{\omega}(u)$ and $\hat{\lambda}(k)$, non-local features, in both time and space can be incorporated in the corresponding generalised diffusion equation. A variety of models ranging in complexity have been described in the literature that attempt to capture statistical aspects of *stress redistribution* processes, akin to those of plate tectonics and more broadly in stick-slip systems [172, 173]. As mentioned in the introduction a common feature of both simulated and real world statistical data is that the waiting time distribution between successive events appears to be contained within the functional form of a *generalised* Γ *distribution* [174, 175]. The generalised Γ distribution is defined as,

$$\omega(t) = \frac{t^{\gamma-1}}{\tau^{\gamma}\Gamma(\gamma/\alpha)} \exp\left(\frac{-t^{\alpha}}{\tau^{\alpha}}\right). \tag{3.3}$$

In this Chapter the influence of waiting times of this form on diffusive processes is explored, with $\alpha = 1$ and $\gamma \in [0,1]$. Thus, the events exhibit random behaviour in the long time regime. The motivation for exploring this is the expectation that stress redistributing events within polymer networks will produce similar statistical signatures. The development of internal stresses and these restructuring events are expected to contribute to the dynamics of the system. The Laplace transform of Eq. (3.3) is,

$$\tilde{\omega}(u) = \frac{1}{\tau^{\gamma}(\frac{1}{\tau}+u)^{\gamma}}. \tag{3.4}$$

Substituting Eq. (3.4) into the Eq. (2.59), under the following conditions

$$P(x,t) = \delta(x) \quad \text{as} \quad t \to 0,$$

$$P(x,t) = 0 \quad \text{as} \quad x \to \pm\infty,$$

yields

$$\hat{\tilde{P}}(k,u) = \frac{1 - \frac{1}{\tau^{\gamma}(\frac{1}{\tau}+u)^{\gamma}}}{u} \frac{P_0(k)}{1 - \hat{\lambda}(k)\frac{1}{\tau^{\gamma}(\frac{1}{\tau}+u)^{\gamma}}}. \tag{3.5}$$

As a brief aside we note here (and from this point forward) that $P(x,t)$ corresponds to $P_{SR}(x,t)$ in Eq. (3.1).

Gaussian Jump Distribution

After inserting this new functional form of $\widetilde{\omega}(u)$, the effects of a Gaussian jump distribution $\lambda(x)$ are considered. This distribution may be represented, in the Fourier space, by the following approximation for small k (hydrodynamic limit),

$$\widehat{\lambda}(k) \sim 1 - \sigma^2 k^2, \quad \text{as} \quad k \to 0 \tag{3.6}$$

where σ^2 represents the variance. With some rearranging of Eq. (3.5),

$$\widehat{\widetilde{P}}(k,u) = \frac{1}{u}\frac{1}{1 + \frac{\sigma^2 k^2}{\tau^\gamma((\frac{1}{\tau}+u)^\gamma - \frac{1}{\tau^\gamma})}}. \tag{3.7}$$

By defining the ratio σ^2/τ^γ to be the generalised diffusion coefficient D_γ, with units m^2/s^γ (where $\gamma \to 1$ recovers the standard diffusion coefficient),

$$\widehat{\widetilde{P}}(k,u) = \frac{1}{u}\frac{1}{1 + \frac{D_\gamma k^2}{((\frac{1}{\tau}+u)^\gamma - \frac{1}{\tau^\gamma})}}. \tag{3.8}$$

3.2.1 Second Moment Behaviour

The transient nature of the process driving the temporal evolution of the system is apparent in the second moment of the PDF. The second moment, $\mu_2(t)$ is related to the mean squared displacement (MSD) in the following way,

$$\mu_2(t) = \langle x^2 \rangle(t) + \langle x \rangle^2(t). \tag{3.9}$$

Thus the MSD and second moment are equivalent in the present work, where $\langle x \rangle(t) = 0$, by virtue of beginning with a bias free CTRW. The second moment of the PDF in Eq. (3.8), may be obtained from the Laplace inversion of the relation

$$\widetilde{\langle x^2 \rangle}(u) = -\frac{\partial^2}{\partial k^2}\widehat{\widetilde{P}}(k,u)|_{k=0}. \tag{3.10}$$

After Laplace inversion of Eq. (3.10)

$$\langle x^2\rangle(t) = D_\gamma \exp\left(-\frac{t}{\tau}\right)\sum_{k=0}^{\infty}\left(\frac{t}{\tau}\right)^k t^\gamma E_{\gamma,1+\gamma+k}\left(\frac{t^\gamma}{\tau^\gamma}\right) \tag{3.11}$$

where $E_{a,b}(t)$ is the generalised Mittag-Leffler function [176]. Fig. 3.2 displays the behaviour of this expression. The processes evolves sub-diffusively $\langle x^2\rangle(t) \propto t^\gamma$, for $t \ll \tau$, prior to transitioning to Fickian behaviour, $\langle x^2\rangle(t) \propto t$, for $t \gg \tau$ as the random nature of long time scale events begins to manifest.

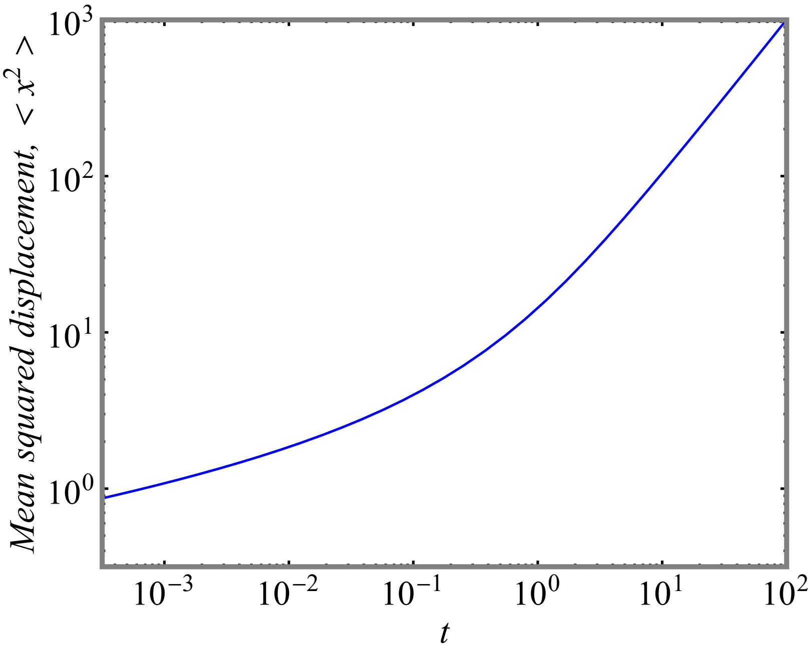

Figure 3.2: Second moment behaviour for $P(x,t)$, found in Eq. (3.12). With $\tau = \sigma = 1$ and $\gamma = 1/10$. The parameter τ plays the role of determining the approximate temporal scale on which the process relaxes to a Fickian process. The parameter γ corresponds to the inverse power law scaling on short time scales, and is therefore responsible for the sub-diffusive character apparent in the figure for small t.

3.2.2 Diffusion Equation

Applying the inverse Laplace and Fourier transforms to Eq. (3.8), the following generalised diffusion equation is obtained,

$$\frac{\partial}{\partial t}P(x,t) = D_\gamma \frac{\partial}{\partial t}\int_0^t \exp\left(-\frac{(t-t')}{\tau}\right)(t-t')^{\gamma-1}E_{\gamma,\gamma}\left(\frac{(t-t')^\gamma}{\tau^\gamma}\right)\frac{\partial^2}{\partial x^2}P(x,t')dt'. \tag{3.12}$$

Generalised diffusion equations have been investigated recently by Tateishi *et al.* [177] and have been shown to describe a range of different diffusive behaviours. Within this work Tateishi *et al.* also discuss the concept of a *memory kernel*, $\mathcal{K}(t)$, which represents the function being involved in the convolution with the standard diffusive component, $\frac{\partial^2}{\partial x^2}P(x,t)$ as mentioned in the introductory Chapter. The form of the memory kernel relevant to this work appears as

$$\mathcal{K}(t) = \exp(-t/\tau)t^{\gamma-1}E_{\gamma,\gamma}\left(t^{\gamma}/\tau^{\gamma}\right). \tag{3.13}$$

The memory kernel describes the influence past diffusive behaviour has on the present dynamics. The notion of a *memory kernel* has emerged historically in other frameworks, such as the generalised Langevin equation [22]. Fig. 3.3 highlights the comparison of this memory kernel with that found in the Riemann-Liouville fractional diffusion equation. The y-axis of Fig 3.3 refers to the value of the memory kernel, the phrase *memory weighting* is a reference to the following (equivalent) interpretation of the role of the memory kernel

$$\frac{\partial}{\partial t}\int_0^t \mathcal{K}(t-t')\frac{\partial^2}{\partial x^2}P(x,t')dt' = \int_0^t \mathcal{K}(t-t')\frac{\partial}{\partial t'}\frac{\partial^2}{\partial x^2}P(x,t')dt' + \mathcal{K}(t)\frac{\partial^2}{\partial x^2}P(x,0), \tag{3.14}$$

when expressed in this fashion it is possible to describe $\mathcal{K}(t-t')$ as a *memory weighting* which is involved in the infinite sum of the infinitesimal past diffusive components $\frac{\partial}{\partial t'}\frac{\partial^2}{\partial x^2}P(x,t')dt'$. For small values of t the functions have the same dependence on t, thus they account for the *recent memory* in the same way. However, as the value of t grows the two kernels deviate in behaviour, with the kernel of interest to this research decaying to a *constant* value. This reflects the transition back to a Markovian process on these timescales and the return to Fickian dynamics.

3.2.3 Probability Density Function

To obtain the PDF of interest in x-t space requires the Fourier-Laplace inversion of the following equation

$$\widehat{\widetilde{P}}(k,u) = \frac{1}{u}\frac{1}{1+\frac{\sigma^2k^2}{\tau^{\gamma}((\frac{1}{\tau}+u)^{\gamma}-\frac{1}{\tau^{\gamma}})}}, \tag{3.15}$$

which has been developed through a CTRW framework, with a Gaussian jump distribution and gamma waiting time distribution. This equation has been discussed once prior [178]. However,

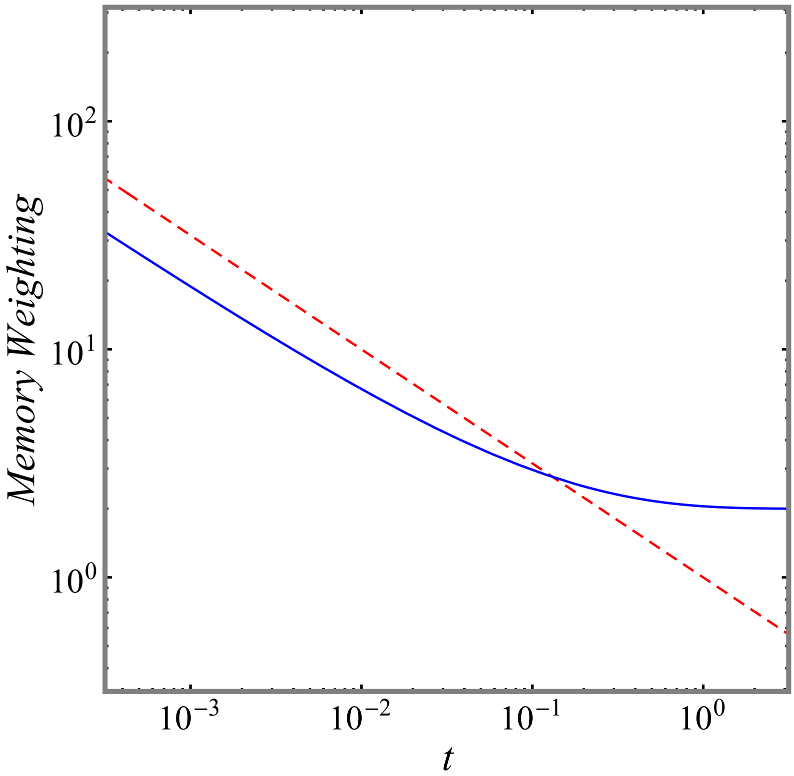

Figure 3.3: Comparison of the RL memory kernel (red) and memory kernel of Eq. (3.12) (blue). Here $\tau = \sigma = 1$ and $\gamma = \frac{1}{2}$. The parameter τ determines the relaxation time for the memory kernel from the RL behaviour back to a constant value. The relaxation to a constant value is realised in the plot above for $t > 1$.

in the application to physical systems, the Fox H-function solution, the short and long timescale asymptotic behaviour, and the means by which the Gaussian solution may be recovered upon setting $\gamma = 1$ are described for the first time. First the Fourier inverse transform of the expression is performed, readily identified by a common Fourier inversion result [179]

$$\widetilde{P}(x,u) = \mathscr{F}^{-1}\left[\widehat{\widetilde{P}}(k,u)\right] = \mathscr{F}^{-1}\left[\frac{1}{u}\frac{1}{1+\frac{\sigma^2 k^2}{\tau^{\gamma}((\frac{1}{\tau}+u)^{\gamma}-\frac{1}{\tau^{\gamma}})}}\right]. \tag{3.16}$$

Evaluating the Fourier transform gives us the following expression, in x-u space

$$\widetilde{P}(x,u) = \frac{\tau^{\frac{\gamma}{2}}}{2u\sigma}\sqrt{(\frac{1}{\tau}+u)^{\gamma}-\frac{1}{\tau^{\gamma}}}\exp\left(-\frac{|x|\tau^{\frac{\gamma}{2}}}{\sigma}\sqrt{(\frac{1}{\tau}+u)^{\gamma}-\frac{1}{\tau^{\gamma}}}\right). \tag{3.17}$$

Taking the inverse Laplace transform yields,

$$P(x,t) = \mathscr{L}^{-1}\Big[\frac{\tau^{\frac{\gamma}{2}}}{2u\sigma}\sum_{n=0}^{\infty}\Big(-\frac{|x|\tau^{\frac{\gamma}{2}}}{\sigma}\Big)^n\Big((\frac{1}{\tau}+u)^{\gamma}\Big)^{\frac{1}{2}+\frac{n}{2}}\Big(1-\frac{1}{\tau^{\gamma}(\frac{1}{\tau}+u)^{\gamma}}\Big)^{\frac{1}{2}+\frac{n}{2}}\frac{1}{\Gamma(n+1)}\Big](t). \tag{3.18}$$

Taking out the $1/u$ factor as the t space definite integral over $[0,t]$.

$$P(x,t) = \int_0^t \mathscr{L}^{-1}\Big[\frac{\tau^{\frac{\gamma}{2}}}{2\sigma}\sum_{n=0}^{\infty}\Big(-\frac{|x|\tau^{\frac{\gamma}{2}}}{\sigma}\Big)^n\Big((\frac{1}{\tau}+u)^{\gamma}\Big)^{\frac{1}{2}+\frac{n}{2}}\Big(1-\frac{1}{\tau^{\gamma}(\frac{1}{\tau}+u)^{\gamma}}\Big)^{\frac{1}{2}+\frac{n}{2}}\frac{1}{\Gamma(n+1)}\Big](t')dt'. \tag{3.19}$$

Invoking the *shift theorem* of the Laplace transform and utilising the binomial theorem leaves us with the expression

$$P(x,t) = \int_0^t \exp(-\frac{t'}{\tau})\mathscr{L}^{-1}\Big[\frac{\tau^{\frac{\gamma}{2}}}{2\sigma}\sum_{n=0}^{\infty}\Big(-\frac{|x|\tau^{\frac{\gamma}{2}}}{\sigma}\Big)^n (u^{\gamma})^{\frac{1}{2}+\frac{n}{2}}\sum_{m=0}^{\infty}\frac{\Gamma(\frac{3}{2}+\frac{n}{2})}{\Gamma(m+1)\Gamma(\frac{3}{2}+\frac{n}{2}-m)} \times\Big(-\frac{1}{\tau^{\gamma}u^{\gamma}}\Big)^m\frac{1}{\Gamma(n+1)}\Big](t')dt'. \tag{3.20}$$

This expression can be cast as a Fox H-function [180], by virtue of its series expansion [181]. Recognising the Fox H-function form allows for more straight forward manipulations, with regards to the inverse transforms and reduction to known results. For these reasons and others the H-function sees regular employment within the realm of fractional and generalised diffusion equations [182–186]

$$P(x,t) = \int_0^t \exp(-\frac{t'}{\tau})\mathscr{L}^{-1}\Big\{\frac{\tau^{\frac{\gamma}{2}}}{2\sigma}\sum_{m=0}^{\infty}\frac{1}{\Gamma(m+1)}\Big(-\frac{1}{\tau^{\gamma}u^{\gamma}}\Big)^m u^{\frac{\gamma}{2}}H^{1,1}_{1,2}\left[\frac{|x|\tau^{\frac{\gamma}{2}}u^{\frac{\gamma}{2}}}{\sigma}\middle|\begin{matrix}(-\frac{1}{2},\frac{1}{2})\\(0,1)(m-\frac{1}{2},\frac{1}{2})\end{matrix}\right]\Big\}(t')dt'. \tag{3.21}$$

The Laplace inversion of the Fox H-function, followed by the use of Eq. (D.15) in Appendix D, is now employed. Due to the flexibility of the coefficients, a_i and α_i, (by virtue of the value of p in the H-function) it can be cast in the following form,

$$P(x,t) = \frac{1}{2}\int_0^t \exp(-\frac{t'}{\tau})\sum_{m=0}^{\infty}\frac{1}{\Gamma(m+1)}\Big(\frac{t'^{\gamma}}{\tau^{\gamma}}\Big)^m\frac{1}{|x|}\frac{1}{t'}H^{2,0}_{2,2}\left[\frac{|x|\tau^{\frac{\gamma}{2}}}{\sigma t'^{\frac{\gamma}{2}}}\middle|\begin{matrix}(0,\frac{1}{2}),(\gamma m,\frac{\gamma}{2})\\(m,\frac{1}{2}),(1,1)\end{matrix}\right]dt'. \tag{3.22}$$

The behaviour of this solution across a range of timescales and γ values is portrayed in Fig. 3.4. This solution describes a system exhibiting short timescale sub-diffusive behaviour which transitions on longer time scales to normal diffusion and thus may capture many real world systems with this stress-redistribution phenomenology.

Summation Form

In Eq. (3.17) we invoke the shift theorem, then prior to the evaluation of the inverse Laplace transform we make use of the binomial theorem, before finally reforming the H-function structure and inverting. Carrying this out gives,

$$P(x,t) = \exp(-t/\tau)\frac{\tau^{\gamma/2}}{2\sigma}\sum_{j=0}^{\infty}\sum_{m=0}^{\infty}\frac{1}{\Gamma(m+1)}\left(\frac{-t^{\gamma}}{\tau^{\gamma}}\right)^{m}\frac{1}{t^{\frac{\gamma}{2}}}\left(\frac{t}{\tau}\right)^{j}H_{2,2}^{1,1}\left[\frac{\tau^{\gamma/2}|x|}{\sigma t^{\gamma/2}}\middle|_{(0,1)(m-1/2,1/2)}^{(-1/2,1/2)(1+j+\gamma m-\gamma/2,\gamma/2)}\right]. \tag{3.23}$$

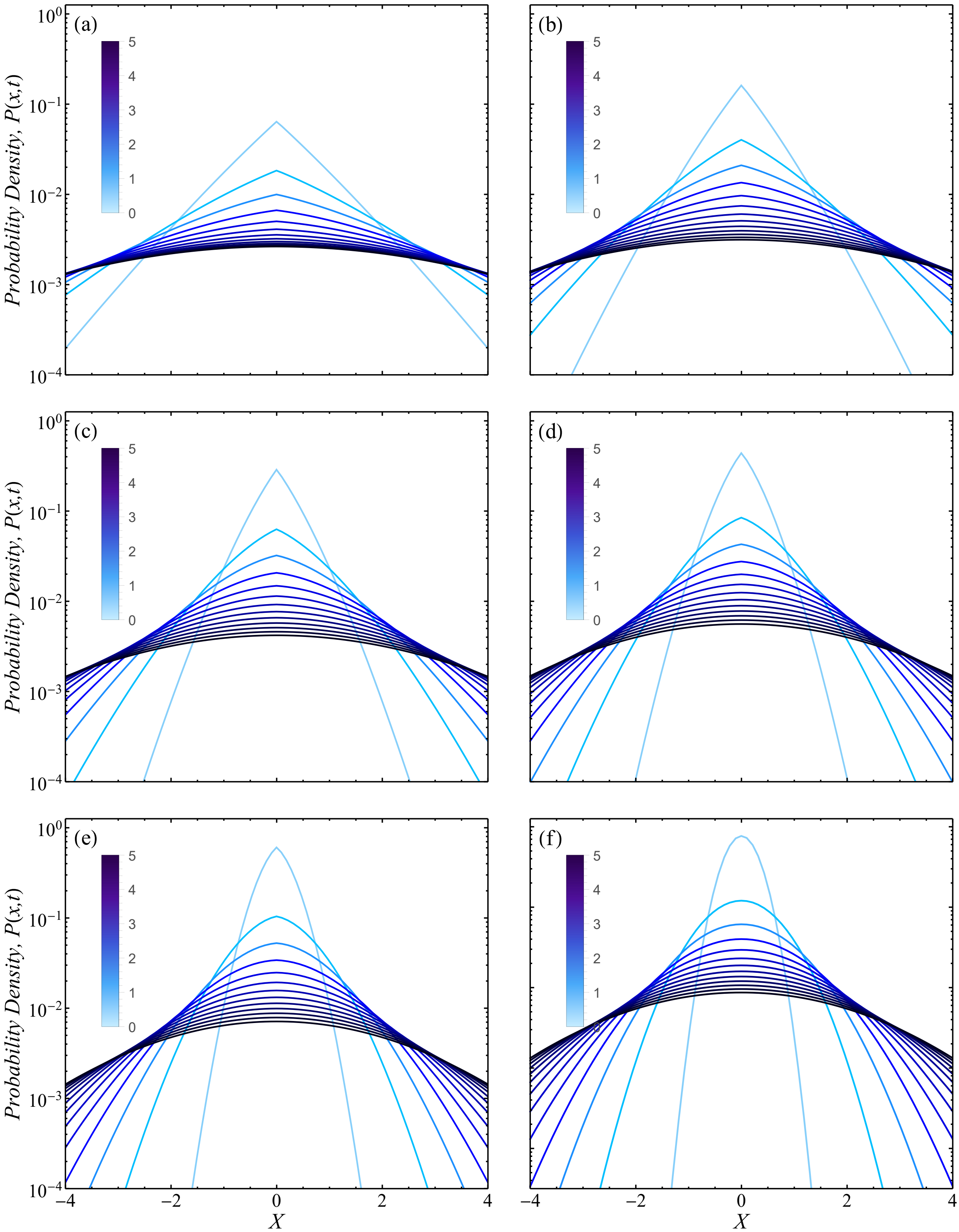

Figure 3.4: PDFs corresponding to increasing values of γ in Eq. (3.22), where X is dimensionless, $X = x/\sigma$. The value of γ ranges from $\gamma = 1/6$ (a), to $\gamma = 1$ (f), in increments of $1/6$. The colours correspond to units of time, t/τ, within the open range $(0, 5)$ (light blue to black). These figures have used the values $\tau = \sigma = 1$. τ plays the role of determining the time scale upon which relaxation to a Fickian process occurs. The anomalous exponent γ is associated with the short time scale fractional diffusion and it's impact is apparent through the PDF possessing a cusp like nature at the origin. For smaller values of γ the ensemble spreads slower than for larger values.

3.2.4 Recovery of the Gaussian Probability Density Function

We can test the behaviour of this result in several different ways, firstly by setting $\gamma = 1$ in Eq. (3.22), this should cause the solution to return to the solution of the normal diffusion equation. This change immediately allows for the reduction of the H-function by way of Eq. (D.12) of Appendix D. After the reduction, there is no longer any occurrence of the index m within the H-function structure, thus all that remains is the series form of the exponential function. Carrying out these steps leaves the form,

$$P(x,t) = \int_0^t \exp\left(-\frac{t'}{\tau}\right)\exp\left(\frac{t'}{\tau}\right)\frac{1}{2|x|}\frac{1}{t'}H_{1,1}^{1,0}\left[\frac{|x|\tau^{\frac{1}{2}}}{\sigma t'^{\frac{1}{2}}}\middle|_{(1,1)}^{(0,\frac{1}{2})}\right]dt', \tag{3.24}$$

where the exponential functions cancel one another, leaving,

$$P(x,t) = \int_0^t \frac{1}{2|x|}\frac{1}{t'}H_{1,1}^{1,0}\left[\frac{|x|\tau^{\frac{1}{2}}}{\sigma t'^{\frac{1}{2}}}\middle|_{(1,1)}^{(0,\frac{1}{2})}\right]dt'. \tag{3.25}$$

We now utilise Eq. (D.14) in Appendix D, and use it in conjunction with Legendre's duplication formula for the gamma function. These steps produce the following result,

$$P(x,t) = \frac{1}{\sqrt{4\pi}}\int_0^t \frac{1}{|x|}\frac{1}{t'}H_{1,2}^{2,0}\left[\frac{x^2\tau}{4\sigma^2 t'}\middle|_{(\frac{1}{2},1),(1,1)}^{(0,1)}\right]dt'. \tag{3.26}$$

Next Eq. (D.16) of Appendix D, is used in combination with taking the partial derivative with respect to t of both sides of the equation, thus producing,

$$\frac{\partial}{\partial t}P(x,t) = \frac{1}{\sqrt{4\pi}}\frac{-1}{|x|}\frac{1}{t}H_{1,2}^{1,1}\left[\frac{x^2\tau}{4\sigma^2 t}\middle|_{(1,1),(\frac{1}{2},1)}^{(0,1)}\right]. \tag{3.27}$$

By virtue of the known differential results for the H-function covered in Appendix D (specifically, Eq. (D.18)), the following result can be derived,

$$\frac{\partial}{\partial t}P(x,t) = \frac{1}{\sqrt{4\pi}}\sqrt{\frac{\tau}{\sigma^2}}\frac{\partial}{\partial t}\frac{1}{\sqrt{t}}H_{0,1}^{1,0}\left[\frac{x^2\tau}{4\sigma^2 t}\middle|_{(0,1)}\right]. \tag{3.28}$$

Taking the antiderivative of both sides, and utilising the initial conditions discussed earlier, namely the initial Dirac delta form over x, provides,

$$P(x,t) = \frac{1}{\sqrt{4\pi}}\sqrt{\frac{\tau}{\sigma^2}}\frac{1}{\sqrt{t}}H^{1,0}_{0,1}\left[\frac{x^2\tau}{\sigma^2 t}\bigg|_{(0,1)}\right], \tag{3.29}$$

which is the H-function form of the Gaussian PDF as expected.

3.2.5 Long Timescale Asymptotics

The H-function, present in Eq. (3.22), in its series form as defined in the article by Metzler, Tomovski and Sandev [181, 187] is,

$$\begin{aligned} H^{2,0}_{2,2}\left[\frac{|x|\tau^{\frac{\gamma}{2}}}{\sigma t^{\frac{\gamma}{2}}}\bigg|^{(0,\frac{1}{2}),(\gamma m,\frac{\gamma}{2})}_{(m,\frac{1}{2}),(1,1)}\right] &= \sum_{n=0}^{\infty}\frac{\Gamma(1-2m-2n)}{\Gamma(\gamma m-\gamma(m+n))}\frac{(-1)^n}{\Gamma(-m-n)\Gamma(n+1)(\Gamma(\frac{3}{2}))}\left(\frac{x^2\tau^{\gamma}}{\sigma^2 t^{\gamma}}\right)^{m+n} \\ &+\sum_{n=0}^{\infty}\frac{\Gamma(m-\frac{1}{2}(1+n))}{\Gamma(\gamma m-\gamma(1+n))\Gamma(-\frac{1}{2}-\frac{n}{2})}\frac{(-1)^n}{\Gamma(n+1)}\left(\frac{|x|\tau^{\frac{\gamma}{2}}}{\sigma t^{\frac{\gamma}{2}}}\right)^{1+n}. \end{aligned} \tag{3.30}$$

Because $\frac{1}{\Gamma(-m-n)} = 0$ for $m,n \in \mathbb{Z}^{0+}$, only the second term contributes. Inserting this result back into Eq. (3.22) yields,

$$\begin{aligned} P(x,t) = &\frac{1}{2}\int_0^t \exp(-\frac{t'}{\tau})\sum_{m=0}^{\infty}\frac{1}{\Gamma(m+1)}\left(\frac{t'^{\gamma}}{\tau^{\gamma}}\right)^m \\ &\times\frac{1}{|x|}\frac{1}{t'}\sum_{n=0}^{\infty}\frac{\Gamma(m-\frac{1}{2}(1+n))}{\Gamma(\gamma m-\gamma(1+n))\Gamma(-\frac{1}{2}-\frac{n}{2})\Gamma(n+1)}(-1)^n\left(\frac{|x|\tau^{\frac{\gamma}{2}}}{\sigma t'^{\frac{\gamma}{2}}}\right)^{(1+n)} dt'. \end{aligned} \tag{3.31}$$

Expressing the series over m as a Fox H-function,

$$\begin{aligned} P(x,t) &= \frac{1}{2}\int_0^t \exp(-\frac{t'}{\tau})\sum_{n=0}^{\infty}\frac{(-1)^n}{\Gamma(-\frac{1}{2}-\frac{n}{2})\Gamma(n+1)}\left(\frac{|x|\tau^{\frac{\gamma}{2}}}{\sigma t'^{\frac{\gamma}{2}}}\right)^{1+n}\frac{1}{|x|}\frac{1}{t'}H^{1,1}_{1,2}\left[-\frac{t^{\gamma}}{\tau^{\gamma}}\bigg|^{(\frac{3}{2}+\frac{n}{2},1)}_{(0,1),(1+\frac{\gamma}{2}+\frac{\gamma n}{2},\gamma)}\right]dt' \\ &= \frac{1}{2}\int_0^t \exp(-\frac{t'}{\tau})\sum_{n=0}^{\infty}\frac{(-1)^n}{\Gamma(-\frac{1}{2}-\frac{n}{2})\Gamma(n+1)}\left(\frac{|x|\tau^{\frac{\gamma}{2}}}{\sigma t'^{\frac{\gamma}{2}}}\right)^{1+n}\frac{1}{|x|}\frac{1}{t'}\,{}_1\Psi_1\left[\frac{t^{\gamma}}{\tau^{\gamma}}\bigg|^{(-\frac{1}{2}-\frac{n}{2},1)}_{(-\frac{\gamma}{2}-\gamma\frac{n}{2},\gamma)}\right]dt', \end{aligned} \tag{3.32}$$

where ${}_p\Psi_q$ is the Wright function [188]. The asymptotic behaviour of this function is described in the article by Wright published in 1940 [189]. For the Wright function of interest, these parameters take on the following values, which are defined in the work of Wright [189],

$$\begin{aligned}
\kappa &= \gamma \\
h &= \gamma^{-\gamma} \\
\theta &= -\frac{1}{2} - \frac{n}{2} + \frac{\gamma}{2} + \frac{\gamma n}{2} \\
Z &= \gamma(\gamma^{-\gamma}\frac{t^{\gamma}}{\tau^{\gamma}})^{\frac{1}{\gamma}} = \frac{t}{\tau} \\
A_0 &= \gamma^{\frac{n}{2}-\frac{\gamma}{2}-\frac{\gamma n}{2}}\gamma^{\frac{1}{2}+\frac{\gamma}{2}+\frac{\gamma n}{2}} = \gamma^{\frac{1}{2}+\frac{n}{2}}
\end{aligned}$$

$$I\left(\frac{t}{\tau}\right) = \left(\frac{t}{\tau}\right)^{-\frac{1}{2}-\frac{n}{2}+\frac{\gamma}{2}+\frac{\gamma n}{2}} \exp\left(\frac{t}{\tau}\right)\left(\sum_{m=0}^{M-1} A_m \left(\frac{t}{\tau}^{-m}\right) + O\left(\frac{t}{\tau}^{-M}\right)\right). \tag{3.33}$$

In the article by Wright, M is free to be chosen so long as $M \in \mathbb{Z}$. Therefore $M = 1$ is chosen, but the terms $O(\frac{t}{\tau}^{-1})$ are neglected, given the fact $t \to \infty$. Thus,

$$I\left(\frac{t}{\tau}\right) = \left(\frac{t}{\tau}\right)^{-\frac{1}{2}-\frac{n}{2}+\frac{\gamma}{2}+\frac{\gamma n}{2}} \exp\left(\frac{t}{\tau}\right)\left(\gamma^{\frac{1}{2}+\frac{n}{2}}\right). \tag{3.34}$$

$P(x,t)$ appears in this regime as,

$$\begin{aligned}
P(x,t) &= \frac{1}{2}\int_0^t \exp(-\frac{t'}{\tau})\frac{1}{|x|}\frac{1}{t'}\sum_{n=0}^{\infty}\frac{(-1)^n}{\Gamma(-\frac{1}{2}-\frac{n}{2})}\frac{1}{\Gamma(n+1)}\left(\frac{|x|\tau^{\frac{\gamma}{2}}}{\sigma t'^{\frac{\gamma}{2}}}\right)^{1+n} I\left(\frac{t'}{\tau}\right)dt' \\
&= \frac{1}{2}\int_0^t \exp(-\frac{t'}{\tau})\frac{1}{|x|}\frac{1}{t'}\sum_{n=0}^{\infty}\frac{(-1)^n}{\Gamma(-\frac{1}{2}-\frac{n}{2})\Gamma(n+1)}\left(\frac{|x|\tau^{\frac{\gamma}{2}}}{\sigma t'^{\frac{\gamma}{2}}}\right)^{1+n} \frac{t'}{\tau}^{-\frac{1}{2}-\frac{n}{2}+\frac{\gamma}{2}+\frac{\gamma n}{2}} \\
&\times \exp\left(\frac{t'}{\tau}\right)\left(\gamma^{\frac{1}{2}+\frac{n}{2}}\right)dt' \\
&= \frac{1}{2}\int_0^t \frac{1}{|x|}\frac{1}{t'}\sum_{n=0}^{\infty}\frac{(-1)^n}{\Gamma(-\frac{1}{2}-\frac{n}{2})}\frac{1}{\Gamma(n+1)}\left(\frac{\gamma^{\frac{1}{2}}|x|\tau^{\frac{1}{2}}}{\sigma t'^{\frac{1}{2}}}\right)^{1+n} dt',
\end{aligned} \tag{3.35}$$

where, expressing the series in its Fox H-function form yields,

$$P(x,t) = \frac{1}{2}\int_0^t \frac{1}{|x|}\frac{1}{t'}H_{1,1}^{1,0}\left[\frac{\gamma^{\frac{1}{2}}|x|\tau^{\frac{1}{2}}}{\sigma t'^{\frac{1}{2}}}\middle|\begin{matrix}(0,\frac{1}{2})\\(1,1)\end{matrix}\right]dt', \quad \text{as} \quad t \to \infty \tag{3.36}$$

This arrives at a Gaussian PDF in the same manner as demonstrated from Eq. (3.25). Interestingly the remnants of the prior non-Gaussian behaviour are visible in the occurrence of γ in the standard deviation of the resulting Gaussian.

3.2.6 Short Timescale Behaviour

It can be shown that in the regime where $\exp\left(-\frac{t}{\tau}\right) \approx 1$ (or $t \ll \tau$), taking the $m = 0$ term of the summation ($m = 0$ being the dominant term in the series, for small t) yields the solution to the RL fractional diffusion equation. Under these conditions Eq. (3.22) is approximated by,

$$P(x,t) \approx \frac{1}{2}\int_0^t \frac{1}{|x|}\frac{1}{t'}H_{1,1}^{1,0}\left[\frac{|x|\tau^{\frac{\gamma}{2}}}{\sigma t'^{\frac{\gamma}{2}}}\middle|_{(1,1)}^{(0,\frac{\gamma}{2})}\right]dt'. \tag{3.37}$$

Making use of the properties of the H-function (specifically Eq. (D.12) and Eq. (D.16)) [190], the following form of Eq. (3.37) can be found

$$\frac{\partial}{\partial t}P(x,t) = \frac{1}{2t|x|}(-1)H_{2,2}^{1,1}\left[\frac{|x|\tau^{\frac{\gamma}{2}}}{\sigma t^{\frac{\gamma}{2}}}\middle|_{(1,1),(1,\frac{\gamma}{2})}^{(0,\frac{\gamma}{2}),(1,\frac{\gamma}{2})}\right]. \tag{3.38}$$

The right hand side can be identified as the partial derivative of the RL solution (see Eq. (D.18) in Appendix D), with respect to time. Given the Dirac delta initial condition for $P(x,t)$, the RL solution appears after integration of both sides

$$P(x,t) = \frac{1}{2}\frac{1}{|x|}H_{1,1}^{1,0}\left[\frac{|x|\tau^{\frac{\gamma}{2}}}{\sigma t^{\frac{\gamma}{2}}}\middle|_{(1,1)}^{(1,\frac{\gamma}{2})}\right]. \tag{3.39}$$

3.3 Discussion

A functional form of the waiting time distribution, widely associated with the timing of stress driven displacements, was inserted into the CTRW. The corresponding generalised diffusion equation was then obtained, by making use of the small k approximation. The *memory kernel* of this equation was plotted in Fig. 3.3, where it was compared against the power law decay of the Riemann-Liouville kernel. The memory kernel represents the functional form of the *weighting* of past diffusive contributions. The memory kernel described within this Chapter exhibits power law decay which tails off to a constant value: this represents the transition from sub-diffusive behaviour towards standard diffusion. The MSD was obtained as well, plotted in Fig. 3.2, the behaviour is transient in nature, moving between sub-diffusive and diffusive regimes. The early sub-diffusive behaviour can be attributed to the power law decay observed on short timescales for the underlying waiting time distribution. This power law decay is indicative of a correlation of events on shorter timescales. The recovery of Fickian behaviour is a direct consequence of the exponential decay on longer timescales for the waiting time distribution. The solution to the generalised diffusion equation was obtained as an integral over an infinite series of Fox H-functions. The solution was arrived at through a combination of Fourier and Laplace transforms used in conjunction with the known properties of the H-function. It was further shown that the solution reduced to known and expected results across the relevant regimes of short and long timescale, as well as in the instance that the anomalous parameter γ took on the Fickian value of 1. This Chapter explores the implications of *stress-redistribution* type timing, on diffusion behaviour. Restructuring events, which may be described in this manner, have been observed amongst a range of soft materials such as dense colloids, foams, gels and granular fluids [9, 191–194]. It is therefore suggested that the diffusive contribution outlined in this article may be found across these systems. As such it is expected this model will provide a good description of diffusive behaviour in the system outlined in the outset of this work [7].

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STATEMENT OF CONTRIBUTION
DOCTORATE WITH PUBLICATIONS/MANUSCRIPTS

We, the candidate and the candidate's Primary Supervisor, certify that all co-authors have consented to their work being included in the thesis and they have accepted the candidate's contribution as indicated below in the *Statement of Originality*.

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4

Lévy processes

4.1 Overview

In the previous chapter a generalised diffusion equation was derived from an underlying CTRW, which possessed a timing distribution associated with stress redistributing systems. The intention of that work was to explore the ability of fractional or non-Markovian models to describe dynamics within polymer systems with stress redistributing features. It could be said that the *temporal* implications of gel quakes have been considered. We now turn to the spatial implications which may follow from *stress redistributing* processes within polymer systems. In order to capture these spatial features we employ non-Gaussian distributions of the displacements in the underlying CTRW framework. Specifically we look now to stable Lévy distributions, for the PDF describing jump lengths.

4.2 Lévy Flight

If the CTRW contains a Lévy stable distribution for the displacements then such behaviour corresponds to the following Fourier space, small k approximation

$$\widehat{\lambda}(k) \sim 1 - \sigma^{\mu}|k|^{\mu} \tag{4.1}$$

with $\mu \in [1,2]$. Inserting this into Eq. (2.59) yields the following expression for the PDF in the Fourier-Laplace space

$$\widehat{\widetilde{P}}(k,u) = \frac{1}{u} \cdot \frac{1}{1 + \frac{D_{\gamma,\mu}|k|^{\mu}}{((\frac{1}{\tau}+u)^{\gamma} - \frac{1}{\tau^{\gamma}})}}, \tag{4.2}$$

where $D_{\alpha,\mu} = \frac{\sigma^{\mu}}{\tau^{\alpha}}$ is the generalised space-time diffusion coefficient.

4.2.1 Generalised Diffusion Equation

From Eq. (4.2) we can derive a generalised diffusion equation. The generalised diffusion equation is of the form

$$\frac{\partial}{\partial t}P(x,t) = D_{\alpha,\mu}\frac{\partial}{\partial t}\int_0^t \exp(-\frac{(t-t')}{\tau})(t-t')^{\gamma-1}E_{\gamma,\gamma}(\frac{(t-t')^{\gamma}}{\tau^{\gamma}})\frac{\partial^{\mu}}{\partial |x|^{\mu}}P(x,t')dt', \tag{4.3}$$

where $\frac{\partial^{\mu}}{\partial |x|^{\mu}}$ is the Riesz space-fractional derivative defined

$$\frac{\partial^{\mu}}{\partial |x|^{\mu}}f(x) = \mathscr{F}^{-1}\big[|k|^{\mu}f(k)\big](x), \tag{4.4}$$

which in the x space is (for $1 < \mu \leq 2$),

$$\frac{\partial^{\mu}}{\partial |x|^{\mu}}f(x) = \frac{-1}{2\cos(\pi\mu/2)}\frac{1}{\Gamma(2-\mu)}\frac{\partial^2}{\partial x^2}\left(\int_{-\infty}^{x}\frac{f(x')}{(x-x')^{\mu-1}}dx' + \int_{x}^{\infty}\frac{f(x')}{(x'-x)^{\mu-1}}dx'\right). \tag{4.5}$$

Eq. (4.3) therefore represents the extension of the non-locality to include both temporal and spatial domains.

Probability Density Current

There exists a well known connection between the diffusion equation and the continuity equation. The continuity equation states that the total change in concentration at a location (probability density in this case) and the divergence of the concentration current at the same location (probability density current in this case) must be zero. Another way of expressing it is to state that in the instance of a conserved quantity the change in the amount in a region must be equal and opposite to the amount leaving the region,

$$P_t(x,t) = -\frac{\partial}{\partial x} J(x,t), \tag{4.6}$$

where J is the probability density current (PDC). Eq. (4.6) provides a useful starting point for investigations into how a generalised diffusion equation varies from the standard or *normal* case,

$$J(x,t) = -{}_0G_t^{1-\gamma} \frac{\partial^{\mu-1}}{\partial |x|^{\mu-1}} P(x,t), \tag{4.7}$$

where the operator ${}_0G_t^{1-\gamma}$ is defined as,

$${}_0G_t^{1-\gamma} f(t) = \frac{\partial}{\partial t} \int_0^t \exp(-\frac{(t-t')}{\tau})(t-t')^{\gamma-1} E_{\gamma,\gamma}(\frac{(t-t')^\gamma}{\tau^\gamma}) f(t') dt'. \tag{4.8}$$

In the form of Eq. (4.7) it is no longer accurate to describe the PDC as moving down the gradient of the PDF. In order to establish precisely what this PDC is sensitive to, we outline the mathematical relationship between it and the PDF

$$J(x,t) = -{}_0G_t^{1-\gamma} \left[\frac{-1}{\cos\left(\frac{\pi(\mu-1)}{2}\right)} \left({}_{-\infty}D_x^{\mu-1} P(x,t) + {}_xD_\infty^{\mu-1} P(x,t) \right) \right]. \tag{4.9}$$

Where the operators ${}_{-\infty}D_x^{\mu-1}$ and ${}_xD_\infty^{\mu-1}$ are the left and right Riemann-Liouville fractional derivatives (with $0 < \mu - 1 \leq 1$) [195], respectively,

$$J(x,t) =_0 G_t^{1-\gamma} \left[\frac{1}{\cos\left(\frac{\pi(\mu-1)}{2}\right)} \frac{1}{\Gamma(2-\mu)} \frac{\partial}{\partial x} \left(\int_{-\infty}^x \frac{P(x',t)}{(x-x')^{\mu-1}} dx' - \int_x^\infty \frac{P(x',t)}{(x'-x)^{\mu-1}} dx' \right) \right]. \tag{4.10}$$

Thus, there is still a gradient that the PDC will be directed down, however, the gradient is of the factor $\left(\int_{-\infty}^x \frac{P(x',t)}{(x-x')^{\mu-1}} dx' - \int_x^\infty \frac{P(x',t)}{(x'-x)^{\mu-1}} dx' \right)$. This object is positive or negative depending on the position x being considered, which follows from the symmetry of $P(x,t)$. Equally, Eq. (4.10) outlines a measure of the non-local allocation of probability density, it constructs a difference from the weighted sum of probability above and below the point of interest x. It is the gradient of this non-local description that guides the path of the flow of probability density. The presence of the generalised time derivative captures the non-local behaviour in time which persists over a regime connected with τ. The occurrence of the non-local behaviour in time has been motivated in the previous Chapter, as for the appearance of spatial non-local behaviour, that is motivated in a simple manner by the observation that the dynamics originate from a *network*.

4.2.2 Probability Density Function

Beginning with Eq. (4.2),the first modification made is to express it as a Fox H-function,

$$\widehat{\widetilde{P}}(k,u) = \frac{1}{u} H^{1,1}_{1,1}\left[\frac{D_{\gamma,\mu}|k|^{\mu}}{((\frac{1}{\tau}+u)^{\gamma} - \frac{1}{\tau^{\gamma}})} \middle|^{(0,1)}_{(0,1)} \right]. \tag{4.11}$$

Taking the inverse Fourier (cosine) transform, and following up with the inverse Laplace transform yields,

$$P(x,t) = \frac{1}{2\sqrt{\pi}} \int_0^t \exp\left(-\frac{t}{\tau}\right) \mathscr{L}^{-1}\left\{ \frac{1}{|x|} H^{2,1}_{1,3}\left[\frac{(u^{\gamma} - \frac{1}{\tau^{\gamma}})|x|^{\mu}}{2^{\mu} D_{\gamma,\mu}} \middle|^{(1,1)}_{(\frac{1}{2},\frac{\mu}{2})(1,1)(1,\frac{\mu}{2})} \right] \right\}(t')dt' \tag{4.12}$$

As a brief aside here it is pointed out that if only the Fourier inversion is evaluated, it is possible to construct an integral decomposition precisely as described by Chechin *et al.* [148], however, rather than a decomposition involving the Gaussian propagator it now involves the Lévy propagator (using the same Laplace-type transform structure). To our knowledge this has *not* been described in the literature. Prior to the full inversion of the Laplace transform above, we first rearrange the H-function. We achieve this by first expressing the H-function in its series form as described in the text by Mathai and Saxena [187]. This gives us the following result;

$$\begin{aligned} &\mathscr{L}^{-1}\left\{ \frac{1}{|x|} H^{2,1}_{1,3}\left[\frac{(u^{\gamma} - \frac{1}{\tau^{\gamma}})|x|^{\mu}}{2^{\mu} D_{\gamma,\mu}} \middle|^{(1,1)}_{(\frac{1}{2},\frac{\mu}{2})(1,1)(1,\frac{\mu}{2})} \right] \right\}(t) = \\ &\frac{1}{|x|} \mathscr{L}^{-1}\left\{ \sum_{n=0}^{\infty} \left(\frac{\Gamma\left(1 - \frac{2}{\mu}(\frac{1}{2}+n)\right)\Gamma\left(\frac{2}{\mu}(\frac{1}{2}+n)\right)}{\Gamma\left(\frac{1}{2}+n\right)\Gamma(n+1)} \frac{(-1)^n}{\frac{\mu}{2}} \left(\frac{\tau^{\gamma}|x|^{\mu}(u^{\gamma} - \frac{1}{\tau}^{\gamma})2}{2^{\mu}\sigma^{\mu}\mu} \right)^{\frac{2}{\mu}(\frac{1}{2}+n)} \right) \right. \\ &\qquad \left. + \sum_{n=0}^{\infty} \left(\frac{\Gamma\left(\frac{1}{2} - \frac{\mu}{2}(1+n)\right)\Gamma(1+n)}{\Gamma\left(\frac{\mu}{2}(1+n)\right)\Gamma(1+n)} (-1)^n \left(\frac{\tau^{\gamma}|x|^{\mu}(u^{\gamma} - \frac{1}{\tau}^{\gamma})}{2^{\mu}\sigma^{\mu}} \right)^{1+n} \right) \right\}(t). \end{aligned} \tag{4.13}$$

Now we use the binomial theorem to expand the $(u^\gamma - \frac{1}{\tau}^\gamma)$ terms,

$$(u^\gamma - \frac{1}{\tau}^\gamma)^A = u^{\gamma A}(1 - \frac{1}{u^\gamma \tau^\gamma})^A = u^{\gamma A} \sum_{m=0}^{\infty} \begin{pmatrix} A \\ m \end{pmatrix} \left(-\frac{1}{u^\gamma \tau^\gamma} \right)^m, \tag{4.14}$$

where $\begin{pmatrix} A \\ m \end{pmatrix}$ are the generalised binomial coefficients. Thus we have,

$$\begin{aligned}\mathscr{L}^{-1}\left\{\frac{1}{u|x|} H^{2,1}_{1,3}\left[\frac{(u^\gamma - \frac{1}{\tau^\gamma})|x|^\mu}{2^\mu D_{\gamma,\mu}}\Bigg|^{(1,1)}_{(\frac{1}{2},\frac{\mu}{2})(1,1)(1,\frac{\mu}{2})}\right]\right\}(t) = \mathscr{L}^{-1}\Bigg\{\sum_{n=0}^{\infty}\sum_{m=0}^{\infty}\Bigg(\frac{\Gamma\left(1-\frac{2}{\mu}(\frac{1}{2}+n)\right)\Gamma\left(\frac{2}{\mu}(\frac{1}{2}+n)\right)}{\Gamma\left(\frac{1}{2}+n\right)\Gamma(n+1)\Gamma(m+1)} \\ \frac{\Gamma\left(\frac{2}{\mu}(\frac{1}{2}+n)+1\right)}{\Gamma\left(\frac{\mu}{2}(\frac{1}{2}+n)+1-m\right)}\frac{(-1)^n}{\frac{\mu}{2}}\left(\frac{\tau^\gamma|x|^\mu u^\gamma 2}{2^\mu\sigma^\mu\mu}\right)^{\frac{2}{\mu}(\frac{1}{2}+n)}\Bigg)\left(-\frac{1}{u^\gamma\tau^\gamma}\right)^m \\ +\sum_{n=0}^{\infty}\sum_{m=0}^{\infty}\Bigg(\frac{\Gamma\left(\frac{1}{2}-\frac{\mu}{2}(1+n)\right)\Gamma(1+n)\Gamma(2+n)}{\Gamma\left(\frac{\mu}{2}(1+n)\right)\Gamma(1+n)\Gamma(m+1)\Gamma(2+n-m)}(-1)^n\left(\frac{\tau^\gamma|x|^\mu u^\gamma}{2^\mu\sigma^\mu}\right)^{1+n}\Bigg)\left(-\frac{1}{u^\gamma\tau^\gamma}\right)^m\Bigg\}(t).\end{aligned} \tag{4.15}$$

Which when put back into the Fox H-function form, and evaluating the Laplace inversion provides

$$P(x,t) = \frac{1}{2\sqrt{\pi}}\int_0^t \exp\left(-\frac{t'}{\tau}\right)\sum_{m=0}^{\infty}\frac{\frac{\mu}{2}}{\Gamma(m+1)}\left(-\frac{t'^\gamma}{\tau^\gamma}\right)^m\frac{1}{t'|x|}H^{2,2}_{3,4}\left[\frac{\tau^\gamma|x|^\mu}{2^\mu\sigma^\mu t'^\gamma}\Bigg|^{(1,1)(0,1)(\gamma m,\gamma)}_{(\frac{1}{2},\frac{\mu}{2})(1,1)(1,\frac{\mu}{2})(m,1)}\right]dt'. \tag{4.16}$$

The behaviour of this solution for a range of μ and γ values can be seen in Fig. 4.1.

Normalisation

The normalisation of Eq. (4.16) can be demonstrated using the Mellin transform, as shown before in the work of Sandev et al. [118]. After the evaluation of the Mellin transform, Eq. (4.16) becomes,

$$P(x,t) = \int_0^t \exp\left(-\frac{t'}{\tau}\right)\sum_{m=0}^{\infty}\frac{1}{\Gamma(m+1)}\left(\frac{t'^\gamma}{\tau^\gamma}\right)^m\frac{1}{t'}\frac{1}{\Gamma(\gamma m)\Gamma(1-m)}dt'. \tag{4.17}$$

Eq. (4.17) has an equivalent form in terms of the Fox H-function, after expressing it in this manner and taking the Laplace transform reveals

$$\frac{1}{u}H^{1,1}_{2,2}\left[-\frac{1}{(u+\tau)^\gamma\tau^\gamma}\Bigg|^{(1,\gamma)(1,1)}_{(0,1),(1,\gamma)}\right] = \frac{1}{u}\left(1+\frac{1}{(u+\tau)^\gamma\tau^\gamma}\right)^0 = \frac{1}{u}. \tag{4.18}$$

Reduction $\gamma = 1$

If $\gamma = 1$, Eq. (4.16) should reduce to the standard Lévy solution. The first step is to remove the factor $(-1)^m$ from the Fox H-function followed by taking the factor (-1) into the Fox H-function, both via the relations discussed in Skibinski et al. [190]

$$P(x,t) = \frac{1}{\sqrt{\pi}} \int_0^t \frac{(-1)}{t'|x|} H^{1,1}_{1,2}\left[\frac{\tau^\gamma |x|^\mu}{2^\mu \sigma^\mu t'} \Big|^{(0,1)}_{(\frac{1}{2},\frac{\mu}{2})(1,\frac{\mu}{2})} \right]. \tag{4.19}$$

Laplace transforming this to resolve the integral, provides the following expression for $\widetilde{P}(x,u)$

$$\widetilde{P}(x,u) = \frac{1}{\sqrt{\pi}} \frac{(-1)}{u|x|} H^{2,1}_{1,3}\left[\frac{\tau^\gamma |x|^\mu u}{2^\mu \sigma^\mu} \Big|^{(0,1)}_{(0,1)(\frac{1}{2},\frac{\mu}{2})(1,\frac{\mu}{2})} \right]. \tag{4.20}$$

The inversion of this followed by bringing the factor (-1) back into the H-function, enables the cancellation of the coefficient pair $(0,1)$

$$P(x,t) = \frac{1}{\sqrt{\pi}} \frac{1}{|x|} H^{1,1}_{1,2}\left[\frac{\tau^\gamma |x|^\mu}{2^\mu \sigma^\mu t} \Big|^{(1,1)}_{(\frac{1}{2},\frac{\mu}{2})(1,\frac{\mu}{2})} \right]. \tag{4.21}$$

From this expression you may simply remove μ from the Fox H-function (see known properties D), insert a symmetric coefficient pair $(1,\frac{1}{2})$ followed by the employment of the Legendre duplication formula on the coefficient pairs $(1,\frac{1}{2})$ and $(\frac{1}{2},\frac{1}{2})$ to complete the extraction of the standard Lévy form, as required [196].

$$P(x,t) = \frac{1}{|x|\mu} H^{1,1}_{2,2}\left[\frac{\tau|x|}{\sigma t^{\frac{1}{\mu}}} \Big|^{(1,\frac{1}{\mu})(1,\frac{1}{2})}_{(1,1)(1,\frac{1}{2})} \right]. \tag{4.22}$$

Reduction $\mu = 2$

If $\mu = 2$ the Gaussian propagator should be recovered and that is now demonstrated. Starting from Eq. (4.16) we set $\mu = 2$ which gives,

$$P(x,t) = \frac{1}{2\sqrt{\pi}} \int_0^t \exp\left(-\frac{t'}{\tau}\right) \sum_{m=0}^{\infty} \frac{1}{\Gamma(m+1)} \left(-\frac{t'^\gamma}{\tau^\gamma}\right)^m \frac{1}{t'|x|} H^{2,2}_{3,4}\left[\frac{\tau^\gamma x^2}{4\sigma^2 t'^\gamma} \Big|^{(1,1)(0,1)(\gamma m,\gamma)}_{(\frac{1}{2},1)(1,1)(1,1)(m,1)} \right] dt'. \tag{4.23}$$

We then combine coefficients by way of the Legendre duplication relation, and then reduce the H-function to give,

$$P(x,t) = \frac{1}{2}\int_0^t \exp\left(-\frac{t'}{\tau}\right)\sum_{m=0}^{\infty}\frac{1}{\Gamma(m+1)}\left(\frac{t'^{\gamma}}{\tau^{\gamma}}\right)^m \frac{1}{t'|x|}H_{2,2}^{1,1}\left[\frac{\tau^{\frac{\gamma}{2}}|x|}{\sigma t'^{\frac{\gamma}{2}}}\Bigg|_{(1,1)(m,\frac{1}{2})}^{(0,\frac{1}{2})(\gamma m,\frac{\gamma}{2})}\right]dt'. \tag{4.24}$$

This is the integral form identified for the Gaussian CTRW case. An alternate form may be found by first using the shift theorem for the Laplace inversion, rather than removing the $1/u$ factor as an integral from $0 \to t$. Keeping it in the u-space, it becomes $\frac{1}{u-\frac{1}{\tau}}$ and inverting the Laplace transform in it's entirety

$$P(x,t) = \exp\left(-\frac{t}{\tau}\right)\sum_{m=0}^{\infty}\sum_{j=0}^{\infty}\frac{\frac{\mu}{2}}{\Gamma(m+1)}\left(-\frac{t^{\gamma}}{\tau^{\gamma}}\right)^m\left(\frac{t}{B}\right)^j\frac{\sqrt{\pi}}{t|x|}H_{3,4}^{2,2}\left[\frac{\tau^{\gamma}|x|^{\mu}}{\sigma^{\mu}t^{\gamma}}\Bigg|_{(\frac{1}{2},\frac{\mu}{2})(1,1)(1,\frac{\mu}{2})(m,1)}^{(1,1)(0,1)(1+j+\gamma m,\gamma)}\right]. \tag{4.25}$$

Mean Squared Displacement

One short coming of this particular *Lévy* approach (in the context of physical *walkers*), is the difficulty in quantifying a traditional growth of the process, in terms of a physical space explored. As the second moment of the PDF is not defined and hence the *area* explored by the process is also not quantified in that manner. There exist extensions to this Lévy flight construction which provide an ability to maintain the second moment, however, still capture the Lévy flight aspects and hence super-diffusive behaviour.

4.3 Lévy Walk

The Lévy walk represents an extension of the Lévy flight framework, which allows for the second moment to be maintained. It has been described previously as the *proper* connection between Lévy type dynamics and the considerations of a physical system [67]. The point of difference between the Lévy walks and flights schemes is to impose the fact that it must take a finite amount of time to traverse a given displacement, and to remove the possibility of infinitely large displacements. Mathematically this is conditioned by way of a Dirac delta coupling between the displacement x and the waiting time t, such that given a time t the displacement is $|x| = vt$ where v is the

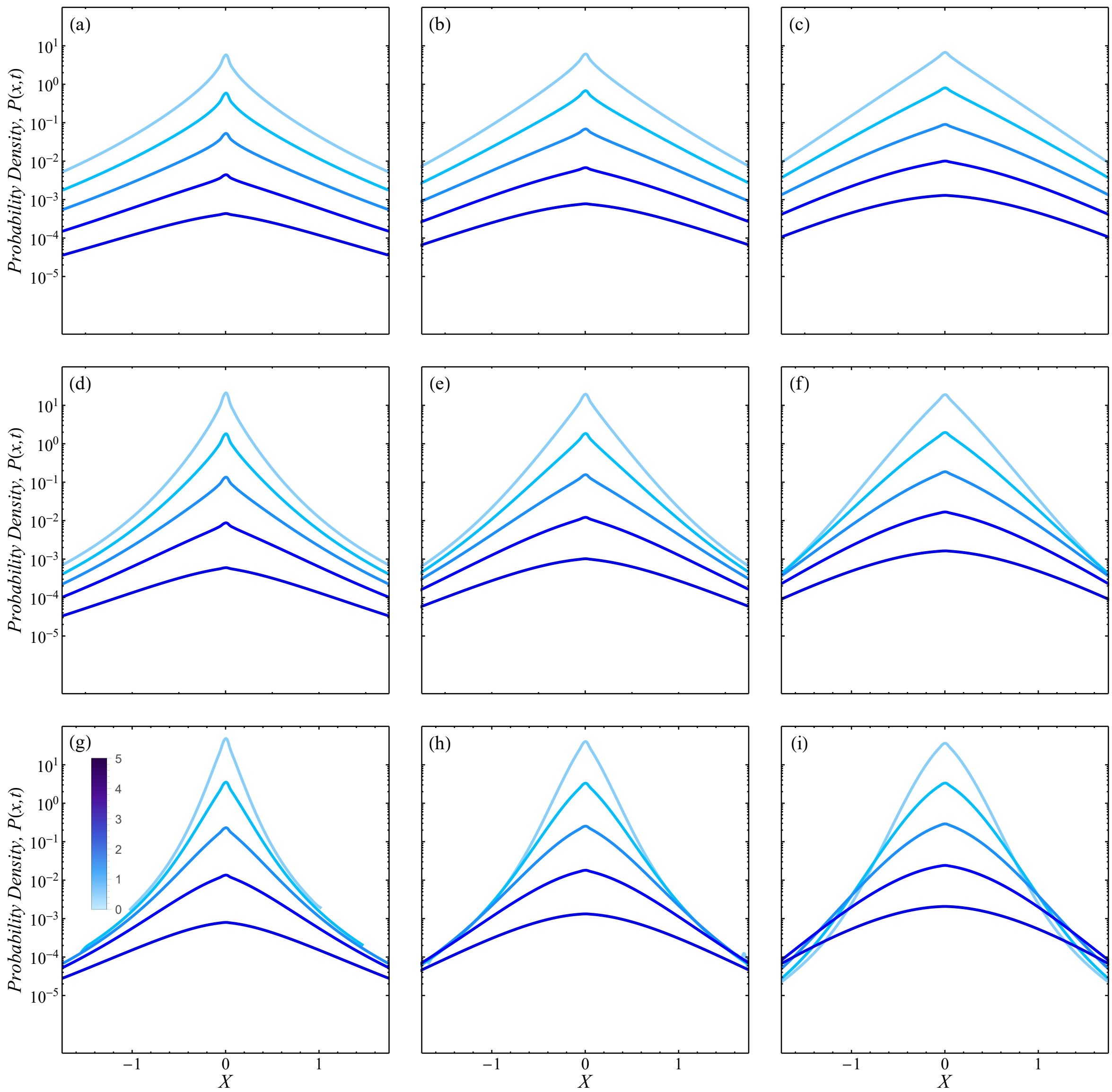

Figure 4.1: PDFs corresponding to increasing values of γ and μ in Eq. (4.25), where X is dimensionless, $X = x/\sigma$. The value of γ ranges from $\gamma = 1/4$ (first row), to $\gamma = 3/4$ (third row), in increments of $1/4$ and μ ranges from $3/2$ (first column) to $5/2$ (third column) in increments of $1/2$. The colours correspond to units of time, t/τ, within the open range $(0, 5)$ (light blue to dark blue). These figures have used the values of τ and σ to be $\tau = \sigma = 1$. These PDFs similar to those displayed for Eq. (3.22), are guided in their relaxation from RL type behaviour to Fickian (or Lévy) by the parameter τ. The additional parameter μ, associated with the Lévy stable distribution in the underlying CTRW, determines the degree of super-diffusive character. For larger values of μ the ensemble spreads more rapidly resulting in PDFs with larger tails.

magnitude of the characteristic velocity associated with the transit.

The Lévy walk framework outlines walker motion as consisting of trajectories during which the walker has a defined and constant velocity. At the end of this trajectory a new direction of motion is selected and the process repeats. Outlining this *Lévy walks* framework requires two features, the PDF for duration time of the motion, $\psi(t)$, and a PDF describing the *frequency* of the change in velocity v, $\rho(x,t)$. The construction of $\rho(x,t)$ is as follows,

$$\rho(x,t) = \int_{-\infty}^{\infty}\int_{0}^{t} \phi(y,t')\rho(x-y,t-t')dt'dy + \delta(t)P_0(x), \tag{4.26}$$

where $P_0(x) = P(x,t)|_{t=0}$. The transition probability density, $\phi(x,t)$ is *coupled* which constrains only walkers at a location $x-vt$ or $x+vt$ will reach x in the time t, and then change the direction of their velocities. The coupling is outlined as,

$$\phi(y,t') = \frac{1}{2}\delta\left(|y|-vt'\right)\psi(t'). \tag{4.27}$$

The second term in Eq. (4.26), outlines the initial conditions for the framework. For any given location the number of walkers in that location is given by

$$P(x,t) = \int_{-\infty}^{\infty}\int_{0}^{t} \Phi(y,t')\rho(x-y,t-t')dt'dy. \tag{4.28}$$

Here $\Phi(x',t')$ describes the probability density associated with the time required to traverse the spatial distance x' being larger than the value t'. The PDF $\Phi(y,t)$ is constructed from the product of the Dirac delta function (as in Eq. (4.27)) and the survival PDF $\Psi(t)$. By utilising the transformation properties of convolutions for both Fourier and Laplace transforms the following expression for the time dependent position PDF can be written,

$$\widehat{\widehat{P}}(k,u) = \frac{\left[\widehat{\widehat{\Psi}}(u+ikv)+\widehat{\widehat{\Psi}}(u-ikv)\right]\widehat{P}_0(k)}{2-\left[\widehat{\widehat{\psi}}(u+ikv)+\widehat{\widehat{\psi}}(u-ikv)\right]} \tag{4.29}$$

Through the use of differing functional forms of $\psi(t)$, a variety of equations capturing differing diffusive behaviours can be obtained, such as the well known Telegrapher's equation.

Lévy Walk With Rests

The Lévy walk framework as outlined above, describes a walker or particle in continuous motion. Another framework can be constructed which takes this flight motion and interrupts it with intermittent resting periods akin to the CTRW motion. This is aptly referred to as the Lévy walks with rests model (LWWR). The LWWR framework results in a natural splitting of the ensemble into two separate states which are resting, P_R and flying P_F. Thus, the PDF for the position of a walker in this framework, exists as the sum of these two states. In the LWWR model we first construct the PDF $\nu(x,t)$ describing the number of particles which have finished their rest period and are once again moving out of a given location x. This PDF is defined as

$$\nu(x,t) = \int_{-\infty}^{\infty}\int_0^t \psi_R(\tau)\int_0^{t-\tau} \phi(y,\tau_1)\nu(x-y,t-\tau-\tau_1)d\tau_1 d\tau dy + \psi_R(t)P_0(x), \quad (4.30)$$

where ϕ is once again the coupled transition PDF of the standard Lévy walks model and $\psi_R(t)$ is the waiting time PDF between flights. The flying and resting state position PDFs, are constructed as

$$P_F(x,t) = \int_{-\infty}^{\infty}\int_0^t \Phi(y,\tau)\nu(x-y,t-\tau)d\tau dy \quad (4.31)$$

and

$$P_R(x,t) = \int_{-\infty}^{\infty}\int_0^t \Psi(\tau)\int_0^{t-\tau} \phi(y,\tau_1)\nu(x-y,t-\tau-\tau_1)d\tau_1 d\tau dy + \Psi_R(t)P_0(x), \quad (4.32)$$

respectively, where Φ is the survival density of the Lévy walks and $\Psi(t)$ the probability that the walker is still in a resting state. As mentioned the total density is constructed from the sum of the two states and this sum can be expressed in the Fourier-Laplace space as,

$$\widehat{\widetilde{P}}_\Sigma(k,u) = \frac{\left[\widehat{\widetilde{\Phi}}(k,u)\widetilde{\psi}(u) + \widetilde{\Psi}(u)\right]\widehat{P}_0(k)}{1-\widetilde{\psi}(u)\widehat{\widetilde{\phi}}(k,u)}. \quad (4.33)$$

There is an interesting interplay that exists between the two states, highlighted by Zaburdaev and Chukbar [197] which is succinctly expressed by,

$$P_F(x,t) = \left(\frac{\langle\tau_f\rangle}{\langle\tau_R\rangle}\right)P_R(x,t). \quad (4.34)$$

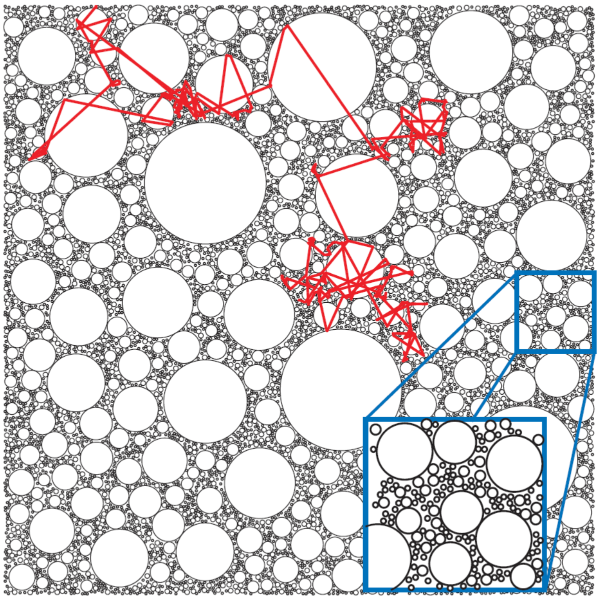

Figure 4.2: Simulated Lévy glass structure, adapted from [198]. The figure displays the path of light (red) within the Lévy simulated glass structure. The scattering of the light within the scale free structure results in Lévy like diffusion occurring.

This relationship is realised at longtime scales, and highlights how the average durations in each state ($\langle\tau_r\rangle$ or $\langle\tau_f\rangle$)ultimately dictate the balance between the states.

4.3.1 Physical System Examples

Prior to delving into an LWWR model of our own, we first provide a brief overview of the history of the subject.

Brief History

The Lévy walk (LW) is a coupled framework (either jump size to waiting time or vice versa), which has been in the literature since around the 80s. The early applications for the framework incorporated phenomena such as turbulent flows and chaos. Some early work in this regard was carried out by Schlesinger *et al* [199], with the coupling introduced via a Dirac delta function linking the PDF for jump lengths to the time, t. They were able to identify the form of the MSD for both turbulent and chaotic systems by considering the particular forms that the spatially dependent velocity fields may take. Their work recovered some known features of turbulent flow such as the t^3 scaling of Richardson's law. Their discussions on chaos revolved around Josephson junctions, where the *velocity field* is independent of the spatial variable, and in fact the spatial

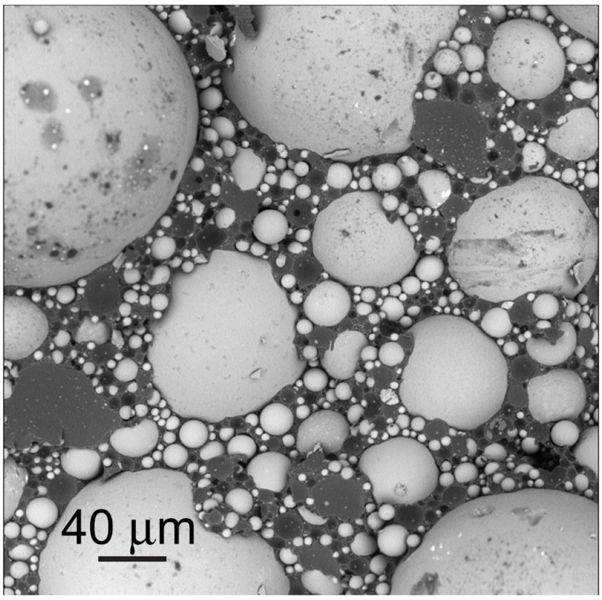

Figure 4.3: Lévy glass structure viewed via electron micrograph, adapted from [198]. Of note in the figure is the apparent scale free structuring of the glass, as was highlighted in the simulated glass structure.

variable is exchanged for a parameter connected to voltage fluctuations. Through the end of the 80s and into the early 90s research in the realms of both chaotic and turbulent systems continued to grow. Works in this vein were well contextualised in the review article by *Klafter et al.* titled *Strange Kinetics* [200]. This review article delved into the nature of chaotic dynamics and its propensity to generate what they refer to as *strange kinetics*. The chaotic systems in question are viewed in terms of their phase space, the strange kinetics being the tendency of fairly stable regions of the phase space to trap the particle between the border of the stable orbit and the edges of the chaotic sea. This behaviour is connected to Lévy walks due to the fact that along the long boundary structures motion is *flight-like*, whilst otherwise the motion is relatively more restricted. However, on top of these explorations into chaotic dynamics, new frontiers emerged for LWs in the form of biological systems [201]. Research into descriptions of DNA structures as well as new and peculiar surface diffusion dynamics were some examples of these new frontiers [202, 203]. There was a marked growth in the number of publications on LWs throughout the late 90s with the previously discussed themes carrying on, but the number of investigations in new topics such as finance and living organisms grew [204–208]. In the late 90s connections were beginning to emerge between fractional Fokker-Planck equations and underlying stochastic frameworks [209]. Similar connections were also being drawn between LW frameworks and slightly more exotic DEs such as the Telegrapher's equation. The dynamics of the LW model (both in its standard form and extended frameworks) are in many cases connected with more sophisticated equations than the decoupled CTRW. The decoupled CTRW at this point in time was garnering attention as it became apparent its connection with fractional diffusion. Equally the connection between LW style diffusion and disordered media was emerging as a key direction of future studies. A key paper in terms of Lévy diffusion studies emerged in 2008 authored by Barthelemy *et al.* [198] proposing a connection between Lévy flights and the diffusion of light through disordered media. Specifically they developed a material called *Lévy Glass* which they required to have self-similarity across many scales in a fractal manner. A simulated example of this structure is seen in Fig 4.2, with the Lévy flight path appearing in red. Their efforts were successful in generating Lévy glass materials and highlighting the descriptive power of the Lévy framework in the context of these materials. They later explored the structures further via electron microscopy as seen in Fig. 4.3. The study of Lévy diffusive systems only continued from this point. Shortly after the Lévy glass work of Barthelemy *et al.*, Vezanni *et al.* considered the similarities between Lévy walks and random walks on inhomogeneous superdiffusive media. In particular the random walk was occurring on a *Cantor graph*, one-dimensional graphs that were found to induce Lévy type behaviour through their fractal nature [210]. The diffusion of cold atoms through an optical lattice was covered by Barkai *et al* [211], in which the Lévy statistics were applied and developed. Barkai *et al.* paid close attention to the appropriate Langevin description for the equation of motion of a single atom, using this as a guide for their excursion into a Lévy probabilistic description. As alluded to in previous works relating to disordered media, there is a strong connection between Lévy statistics with their scale-free attributes. This attribute of Lévy walks has been drawn upon in multiple articles [212–214]. Another important theme that appears in disordered media is the concept of criticality [215]. Criticality is the notion that as a system approaches a location in the phase space where a phase transition will occur, there is a breakdown in the scale of the system. There are believed to be many useful features of maintaining a system at a state of criticality, such as optimal dynamic ranges, high sensitivity and improved computational power [217, 218].

In fact it is suggested that many of the most complex systems amongst the natural world may rely on *self-organized criticality*, to remove the shackles of a scale restricted environment and allow scale invariant events to take place [219]. As may be anticipated, the scale-invariant nature

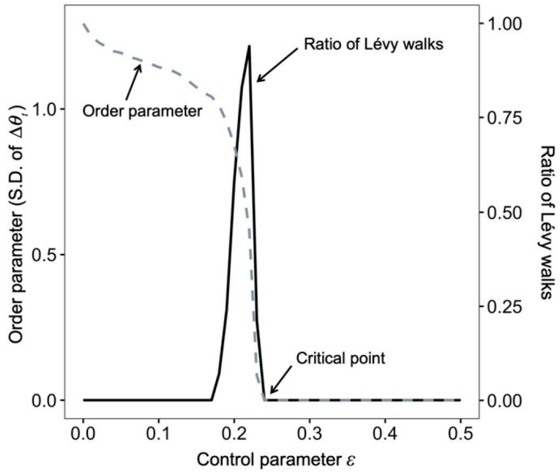

Figure 4.4: Occurrence of Lévy walks in relation to the order parameter value, as the critical point is approached, adapted from [216]. The figure demonstrates the intimate relationship between scale free structuring and Lévy walks. As the system begins to lose the scale separation of structures (encroaches upon the critical point), a sharp increase in the ratio of Lévy walks occurs. As the phase transition is completed the Lévy walks once again vanish.

of systems in criticality has been well mapped to Lévy behaviours such as the Lévy walk. It was identified in [216] that the prevalence of Lévy walks is greatly increased as criticality is approached, an adapted figure from their work may be seen in Fig. 4.4. Whilst the mathematical structure we touch on in this Chapter is explored in a general sense, we are still motivated through the initial foray into describing *quakes* in a polymer system. It is likely that the system exhibiting quaking phenomena may be verging on criticality and may enjoy scale invariant features. Coupled with other precursors, covered for Lévy walk behaviour, this provides sufficient motivation to warrant exploring this framework as an archetype for the *quaking dynamics* whether that is in gel systems or in other systems of a similar variety.

4.3.2 Stress Redistribution LWWR

We now examine a LWWR framework ultilising a Γ distribution for the waiting times between flights. This once again connects this framework with a possible description of stress redistribution driven diffusion. In this scenario the spatial behaviour is altered away from Gaussian distributed displacements to a delta function mediated coupling, as with the standard LW model.

4.3.3 Second Moment

The second moment for the process may be revealed by utilising the following relationship,

$$\langle x^2\rangle(t) = -\mathscr{L}^{-1}\left[\frac{\partial^2}{\partial k^2}\widehat{\widehat{P}}(k,u)\bigg|_{k=0}\right](t). \tag{4.35}$$

Evaluating the Laplace inversion in this expression provides the following expression for the second moment of the LWWR model above. We consider the difference between the inverse of the characteristic time of flight duration and rest periods to be defined as $\Delta = \frac{1}{\tau} - \frac{1}{\tau_R}$, and begin initially in the case $\Delta = 0$. Thus, for $\Delta = 0$, we find

$$\langle x^2\rangle(t) = 2v^2\left(\frac{t}{\tau}\right)^{1+\gamma+k} tE_{1+\gamma,3+\gamma+k}\left(\left(\frac{t}{\tau}\right)^{1+\gamma}\right). \tag{4.36}$$

Notably the behaviour begins in a superdiffusive regime with a characteristic exponent of $2+\gamma$ (green plot of Fig. 4.5), however, as time elapses the exponent relaxes back to a value of 1 suggesting a possible recovery of Fickian dynamics (red plot of Fig. 4.5).

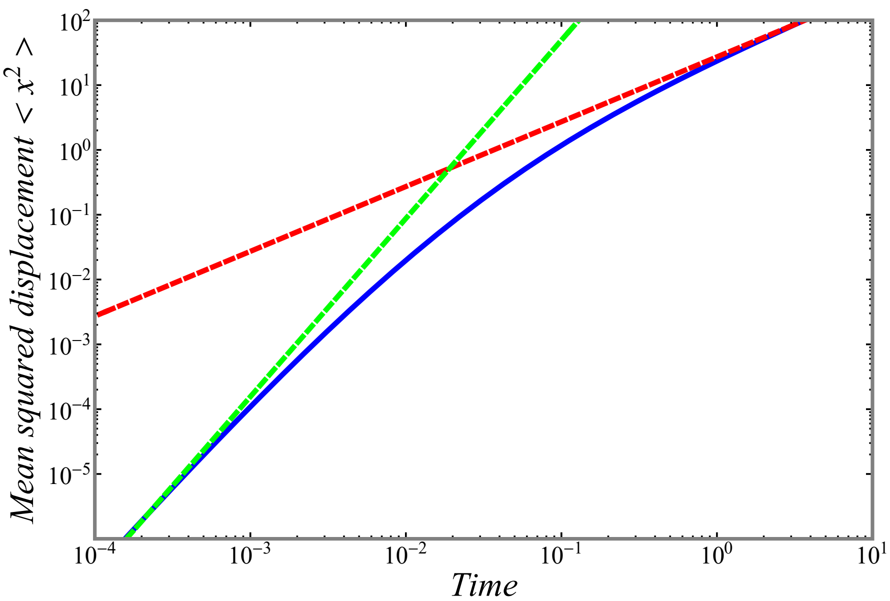

Figure 4.5: Second moment behaviour for $P_\Sigma(x,t)$, found in Eq. (4.36). With $\tau = 1/4, v = 4$ and $\gamma = 3/4$. Green dashed line is indicative of the super-ballistic short time diffusion, whilst the red dashed line is highlighting the eventual relaxation to a linear growth of the second moment, the timescale of relaxation is encoded by τ.

4.3.4 Probability Density Function

We now look to obtain the full PDF $P_\Sigma(x,t) = P_{Flight} + P_{Rest}$. For simplicity beginning with $\Delta = 0$,

$$\widehat{P_\Sigma}(k,t) = \exp(-t/\tau)\mathscr{L}^{-1}\left[\left(\frac{\frac{u^{1-\gamma}}{v^2\tau^{(1+\gamma)}}}{\left(\frac{u}{v}\right)^2+k^2} + \frac{1}{(u-\frac{1}{\tau})}\left(1-\frac{1}{\tau^\gamma u^\gamma}\right)\right)\frac{\left(\frac{u}{v}\right)^2+k^2}{\left(\frac{u}{v}\right)^2+k^2-\frac{u^{1-\gamma}}{v^2\tau^{(1+\gamma)}}}\right]. \tag{4.37}$$

Beginning with the Fourier inversion, this leaves,

$$P_\Sigma(x,t) = \exp(-t/\tau)\mathscr{L}^{-1}\Bigg[\Bigg(\frac{u^{1-\gamma}}{v^2\tau^{(1+\gamma)}}\frac{1}{\sqrt{\left(\frac{u}{v}\right)^2-\frac{u^{1-\gamma}}{v^2\tau^{(1+\gamma)}}}} + \frac{1}{(u-\frac{1}{\tau})}\left(1-\frac{1}{\tau^\gamma u^\gamma}\right)\Bigg(\frac{\frac{u^2}{v^2}}{\sqrt{\left(\frac{u}{v}\right)^2-\frac{u^{1-\gamma}}{v^2\tau^{(1+\gamma)}}}} - \sqrt{\left(\frac{u}{v}\right)^2-\frac{u^{1-\gamma}}{v^2\tau^{(1+\gamma)}}}\Bigg)\Bigg)\exp\left(-|x|\sqrt{\left(\frac{u}{v}\right)^2-\frac{u^{1-\gamma}}{v^2\tau^{(1+\gamma)}}}\right) + \frac{1}{(u-\frac{1}{\tau})\tau}\left(1-\frac{1}{\tau^\gamma u^\gamma}\right)\delta(x)\Bigg], \tag{4.38}$$

which after converting the exponential function to its H-function form, and invoking the Legendre duplication theorem in conjunction with an expansion theorem for the H-function leaves

$$P_\Sigma(x,t) = \frac{1}{\sqrt{\pi}}\exp(-t/\tau)\mathscr{L}^{-1}\Bigg[\frac{u^{-\gamma}}{v\tau^{(1+\gamma)}}\sum_{r=0}^{\infty}\frac{\left(\frac{1}{u^{1+\gamma}\tau^{1+\gamma}}\right)^r}{\Gamma(r+1)}\left(1+\frac{1}{(u-\frac{1}{\tau})}\left(1-\frac{1}{\tau^\gamma u^\gamma}\right)\right) H^{2,0}_{0,2}\left[x^2\left(\frac{u}{2v}\right)^2\Bigg|_{(0,1),(1/2+r,1)}\right] + \frac{1}{(u-\frac{1}{\tau})\tau}\left(1-\frac{1}{\tau^\gamma u^\gamma}\right)\delta(x)\Bigg]. \tag{4.39}$$

Evaluating the inverse Laplace transform, via known H-function properties gives us

$$P_\Sigma(x,t) = \frac{1}{\sqrt{\pi}}\exp(-t/\tau)\frac{t^\gamma}{\tau^{(1+\gamma)}}\sum_{r=0}^{\infty}\frac{\left(\frac{t^{1+\gamma}}{\tau^{1+\gamma}}\right)^r}{\Gamma(r+1)}\Bigg(\frac{1}{t}H^{2,0}_{1,2}\left[x^2\left(\frac{1}{2vt}\right)^2\Bigg|^{(\gamma+r+\gamma r,2)}_{(0,1),(1/2+r,1)}\right] + \sum_{k=0}^{\infty}\left(\frac{t}{\tau}\right)^k\Bigg(H^{2,0}_{1,2}\left[x^2\left(\frac{1}{2vt}\right)^2\Bigg|^{(1+\gamma+k+r+\gamma r)}_{(0,1),(1/2+r,1)}\right] - \frac{t^\gamma}{\tau^\gamma}H^{2,0}_{1,2}\left[x^2\left(\frac{1}{2vt}\right)^2\Bigg|^{(1+2\gamma+k+r+\gamma r,2)}_{(0,1),(1/2+r,1)}\right]\Bigg)\Bigg) + \delta(x) - \exp\left(-\frac{t}{\tau}\right)\frac{t^\gamma}{\tau^\gamma}E_{1,1+\gamma}\left(\frac{t}{\tau}\right)\delta(x). \tag{4.40}$$

The behaviour of this solution is displayed in Fig. 4.6 and it can be demonstrated that it recovers the solution obtained in Chapter 3. Notably with the $\Delta = 0$ case, whilst the Gaussian form is realised between the ballistic fronts, the short time cusp like behaviour is not present. This appears to be due to timescale overlap, and we will now explore the $\Delta \neq 0$ case, to determine whether this cusp-like behaviour may be revealed.

4.3.5 Asymptotic Behaviour $t \to \infty$

Due to the similarity in structure of the three H-functions present in Eq. (4.40), we first explore the asymptotic nature of the first H-function and use this asymptotic form to gain an understanding of how the other two appear in the same regime. The first step in this process is to expand the H-function in its series form,

$$\frac{1}{\sqrt{\pi}}\exp(-t/\tau)\frac{t^{\gamma}}{\tau^{(1+\gamma)}}\sum_{r=0}^{\infty}\frac{\left(\frac{t^{1+\gamma}}{\tau^{1+\gamma}}\right)^{r}}{\Gamma(r+1)}\left(\frac{1}{t}H^{2,0}_{1,2}\left[x^{2}\left(\frac{1}{2\nu t}\right)^{2}\middle|^{(\gamma+r+\gamma r,2)}_{(0,1),(1/2+r,1)}\right]\right)$$
$$=\frac{1}{t}\frac{1}{\sqrt{\pi}}\exp(-t/\tau)\frac{t^{\gamma}}{\tau^{(1+\gamma)}}\sum_{r=0}^{\infty}\frac{\left(\frac{t^{1+\gamma}}{\tau^{1+\gamma}}\right)^{r}}{\Gamma(r+1)}\left(\sum_{n=0}^{\infty}\left(-\left(\frac{x}{2\nu t}\right)^{2}\right)^{n}\frac{\Gamma\left(1/2+r-n\right)}{\Gamma(n+1)\Gamma\left(\gamma+r+\gamma r-2n\right)}\right.$$
$$\left.+\sum_{n=0}^{\infty}\left(x^{2}\left(\frac{1}{2\nu t}\right)^{2}\right)^{1/2+r+n}(-1)^{n}\frac{\Gamma\left(-1/2-r-n\right)}{\Gamma(n+1)\Gamma\left(\gamma-r+\gamma r-2n\right)}\right) \tag{4.41}$$

The first series can be represented as an H-function via the first series form outlined in [187], whilst the second series is expressible as an H-function as per the second form outlined in [187], in both cases it is the summation over r that is used. Making these adjustments leaves,

$$\frac{1}{t}\frac{1}{\sqrt{\pi}}\exp(-t/\tau)\frac{t^{\gamma}}{\tau^{(1+\gamma)}}\sum_{r=0}^{\infty}\frac{\left(\frac{t^{1+\gamma}}{\tau^{1+\gamma}}\right)^{r}}{\Gamma\left(r+1\right])}\left(H^{2,0}_{1,2}\left[x^{2}\left(\frac{1}{2\nu t}\right)^{2}\middle|^{(\gamma+r+\gamma r,2)}_{(0,1),(1/2+r,1)}\right]\right)$$
$$=\frac{1}{t}\frac{1}{\sqrt{\pi}}\exp(-t/\tau)\frac{t^{\gamma}}{\tau^{(1+\gamma)}}\sum_{n=0}^{\infty}\left(-\frac{x^{2}}{(2\nu t)^{2}}\right)^{n}\frac{1}{\Gamma(n+1)}\left(H^{1,1}_{1,2}\left[-\frac{t^{1+\gamma}}{\tau^{1+\gamma}}\middle|^{(1/2+n,1)}_{(0,1),(1+2n-\gamma,1+\gamma)}\right]\right.$$
$$\left.+\frac{|x|}{\nu t}H^{1,1}_{2,1}\left[-x^{2}\left(\frac{1}{2\nu t}\right)^{2}\frac{t^{1+\gamma}}{\tau^{1+\gamma}}\middle|^{(\gamma+r+\gamma r,2)}_{(0,1),(1/2+r,1)}\right]\right) \tag{4.42}$$

For the first H-function above, we can make use of the Wright function asymptotics as outlined in [189]. We use the same approach as in Chapter 3, of setting $M = 1$. Using these results yields asymptotic behaviour of $(1+\gamma)^{n}\left(\frac{t}{\tau}\right)^{n-\gamma+1/2}\exp\left(\frac{t}{\tau}\right)$. For the second H-function we can use the asymptotic results contained within [179], which outlined the fact that this H-function form tends to 1 for large t. Hence the second H-function form is suppressed in the asymptotic regime and it's only the former which contributes giving

$$\begin{aligned}
&\frac{1}{t}\frac{1}{\sqrt{\pi}}\exp(-t/\tau)\frac{t^{\gamma}}{\tau^{(1+\gamma)}}\sum_{r=0}^{\infty}\frac{\left(\frac{t^{1+\gamma}}{\tau^{1+\gamma}}\right)^{r}}{\Gamma(r+1)}\left(H_{1,2}^{2,0}\left[x^{2}\left(\frac{1}{\nu t}\right)^{2}\Bigg|_{(0,1),(1/2+r,1)}^{(1+\gamma+r+\gamma r,2)}\right]\right)\\
&\approx\frac{1}{t}\frac{1}{\sqrt{\pi}}\exp(-t/\tau)\frac{t^{\gamma}}{\tau^{(1+\gamma)}}\sum_{n=0}^{\infty}\left(-\frac{x^{2}}{(\nu t)^{2}}\right)^{n}\frac{1}{\Gamma(n+1)}\left(\frac{t}{\tau}\right)^{n-\gamma+1/2}(1+\gamma)^{n}\exp\left(\frac{t}{\tau}\right)\\
&=\frac{1}{\sqrt{\pi}}\exp\left(-\frac{x^{2}(1+\gamma)}{\nu^{2}\tau t}\right)\left(\frac{\tau}{t}\right)^{-1/2}\quad as\ t\to\infty. \qquad (4.43)
\end{aligned}$$

This behaviour can be observed also in Fig. 4.6, where eventually the bounding behaviour of $|x|<\nu t$ outpaces the growth of the ensemble as the framework naturally relaxes back to a Fickian one.

4.3.6 $P(x,t)$, Without $\Delta=0$ Enforced

The prior derivation of $P_{\Sigma}(x,t)$ was under the assumption that the characteristic times τ and τ_r of the flight and rest times, were equivalent. We now explore the situation where they are different being cognizant of the fact the following derivation does require that the relationship $\tau_r\leq 2\tau$ holds. This relationship connects with the first moments of the respective flight duration and resting time PDFs as follows $\langle t_r\rangle=\gamma\tau_r\leq 2\langle t_f\rangle$. So if we have $\gamma\leq 1$, then the *average* resting time is no larger than twice the average flight time. We now present the implications of relaxing the requirement to $\tau_r\leq 2\tau$, letting $\Delta=\frac{1}{\tau}-\frac{1}{\tau_r}$ we have

$$\begin{aligned}
P_{\Sigma}(x,t)=\frac{1}{\sqrt{\pi}}\exp(-t/\tau_r)\mathscr{L}^{-1}&\left[\frac{u^{-\gamma}}{\nu\tau\tau_r^{\gamma}}\sum_{r=0}^{\infty}\frac{\left(\frac{1}{u^{1+\gamma}\tau\tau_r^{\gamma}}\right)^{r}\left(\frac{1}{1+\frac{\Delta}{u}}\right)^{r}}{\Gamma(r+1)}\left(1+\frac{1}{(u-\frac{1}{\tau_r})}\left(1-\frac{1}{\tau_r^{\gamma}u^{\gamma}}\right)\right)\right.\\
&\left.H_{0,2}^{2,0}\left[x^{2}u^{2}\left(\frac{1+\frac{\Delta}{u}}{\nu}\right)^{2}\Bigg|_{(0,1),(1/2+r,1)}\right]+\frac{1}{(u-\frac{1}{\tau})\tau}\left(1-\frac{1}{\tau^{\gamma}u^{\gamma}}\right)\delta(x)\right](t). \qquad (4.44)
\end{aligned}$$

We now look to use the generalised binomial theorem for the factors $\frac{1}{(1+\frac{\Delta}{u})^r}$ where $r \geq 1$. The term when $r=0$ is pulled out and evaluating this leaves

$$P_\Sigma(x,t) = \frac{1}{\sqrt{\pi}}\exp(-t/\tau_r)\mathscr{L}^{-1}\left[\frac{u^{-\gamma}}{\nu\tau\tau_r^\gamma}\left(1+\frac{1}{(u-\frac{1}{\tau_r})}\left(1-\frac{1}{\tau_r^\gamma u^\gamma}\right)\right)\right.$$
$$\sum_{m=0}^{\infty}\left(-\frac{\Delta}{u}\right)^m \frac{1}{\Gamma(m+1)} H^{3,0}_{1,3}\left[\frac{x^2u^2}{\nu^2}\Bigg|^{(0,2)}_{(m,2)(0,1),(1/2,1)}\right]$$
$$+\frac{u^{-\gamma}}{\nu\tau\tau_r^\gamma}\sum_{r=1}^{\infty}\frac{\left(\frac{1}{u^{1+\gamma}\tau\tau_r^\gamma}\right)^r\left(\frac{\Delta}{u}\right)^n}{\Gamma(r+1)}\frac{\Gamma(n+r)}{\Gamma(n+1)\Gamma(r)}\left(1+\frac{1}{(u-\frac{1}{\tau_r})}\left(1-\frac{1}{\tau_r^\gamma u^\gamma}\right)\right)$$
$$\left.\sum_{m=0}^{\infty}\left(-\frac{\Delta}{u}\right)^m \frac{1}{\Gamma(m+1)} H^{2,0}_{0,2}\left[\frac{x^2u^2}{\nu^2}\Bigg|^{(0,2)}_{(m,2)(0,1),(1/2+r,1)}\right] + \frac{1}{(u-\frac{1}{\tau})\tau}\left(1-\frac{1}{\tau^\gamma u^\gamma}\right)\delta(x)\right](t). \tag{4.45}$$

All that remains is to inverse the Laplace transform which then provides,

$$P_\Sigma(x,t) = \frac{1}{\sqrt{\pi}}\exp(-t/\tau_r)\sum_{m=0}\frac{(-\Delta t)^m}{\Gamma(m+1)}\frac{t^\gamma}{\nu\tau\tau_r^\gamma}\left(\frac{1}{t}H^{3,0}_{2,3}\left[\frac{x^2}{\nu^2t^2}\Bigg|^{(\gamma+m,2)(0,2)}_{(m,2)(0,1),(1/2,1)}\right]\right.$$
$$+\sum_{k=0}^{\infty}\left(\frac{t}{\tau_r}\right)^k\left(H^{3,0}_{2,3}\left[\frac{x^2}{\nu^2t^2}\Bigg|^{(1+\gamma+m+k,2)(0,2)}_{(m,2)(0,1),(1/2,1)}\right]-\frac{t^\gamma}{\tau_r^\gamma}H^{3,0}_{2,3}\left[\frac{x^2}{\nu^2t^2}\Bigg|^{(1+\gamma+m+k,2)(0,2)}_{(m,2)(0,1),(1/2,1)}\right]\right)$$
$$+\sum_{r=1}^{\infty}\frac{\left(\frac{t^{1+\gamma}}{\tau\tau_r^\gamma}\right)^r(\Delta t)^n}{\Gamma(r+1)}\frac{\Gamma(n+r)}{\Gamma(n+1)\Gamma(r)}\left(\frac{1}{t}H^{3,0}_{2,3}\left[\frac{x^2}{\nu^2t^2}\Bigg|^{(\gamma+\gamma r+r+n+m,2)(0,2)}_{(m,2)(0,1),(1/2+r,1)}\right]+\right.$$
$$\left.\left.\sum_{k=0}^{\infty}\left(\frac{t}{\tau_r}\right)^k\left(H^{3,0}_{2,3}\left[\frac{x^2}{\nu^2t^2}\Bigg|^{(1+\gamma+\gamma r+r+n+m+k,2)(0,2)}_{(m,2)(0,1),(1/2+r,1)}\right]-\frac{t^\gamma}{\tau_r^\gamma}H^{3,0}_{2,3}\left[\frac{x^2}{\nu^2t^2}\Bigg|^{(1+\gamma+\gamma r+r+n+m+k,2)(0,2)}_{(m,2)(0,1),(1/2+r,1)}\right]\right)\right)\right)$$
$$+\delta(x)-\exp\left(-\frac{t}{\tau_r}\right)\frac{t^\gamma}{\tau_r^\gamma}E_{1,1+\gamma}\left(\frac{t}{\tau_r}\right)\delta(x). \tag{4.46}$$

In Fig. 4.7, the time evolution of this solution is plotted from several values of γ. It can be seen now that this solution does allow for the occurrence of the cusp like central peak observed in the solution described in Chapter 3. Comparing the behaviours in Fig. 4.7, we see that the ballistic fronts are more heavily populated for smaller values of γ, this can be related back to the small γ values inducing a more pronounced *inverse power law* peak about small waiting times. Thus, when the Dirac delta coupled function samples this regime it results in a larger portion of the ensemble possessing Lévy type behaviour.

4.3.7 Corresponding Differential Equations

We now look to probe the connection between the LWWR structure explored above, and the corresponding differential equation, which is expected to be of the generalised variety

$$\widehat{\widetilde{P}}_{\Sigma}(k,u) = \left(\frac{\frac{\left(u+\frac{1}{\tau}\right)^{1-\gamma}}{v^2\tau^{(1+\gamma)}}}{\left(\frac{\left(u+\frac{1}{\tau}\right)}{v}\right)^2 + k^2} + \frac{1}{u}\left(1 - \frac{1}{\tau^{\gamma}\left(u+\frac{1}{\tau}\right)^{\gamma}}\right)\right) \frac{\left(\frac{\left(u+\frac{1}{\tau}\right)}{v}\right)^2 + k^2}{\left(\frac{\left(u+\frac{1}{\tau}\right)}{v}\right)^2 + k^2 - \frac{\left(u+\frac{1}{\tau}\right)^{1-\gamma}}{v^2\tau^{(1+\gamma)}}}. \tag{4.47}$$

This, as we know, exists as the sum of the PDFs for both *flying* and *resting* members of the ensemble, we therefore explore the form of the equations pertaining to these sub states. Firstly we will look at the connection between the states, expressing the dynamics of P_R in relation to P_F highlighting how movement between states is characterised,

$$\widehat{\widetilde{P}}_R(k,u) = \frac{\widetilde{\Psi}_r \widehat{\widetilde{\phi} P_F}}{\widehat{\widetilde{\Phi}}} + \widetilde{\Psi}_r \widehat{P}_0. \tag{4.48}$$

By virtue of our underlying exponential flight duration PDFs, we have $\phi(t) = \Phi(t)$

$$\widehat{\widetilde{P}}_R(k,u) = \widetilde{\Psi}_r \widehat{\widetilde{P}}_F + \widetilde{\Psi}_r \widehat{P}_0. \tag{4.49}$$

Substituting the appropriate forms in for Ψ_r, and rearranging

$$u\tau\widehat{\widetilde{P}}_R(k,u) = \left(1 - \frac{1}{\tau^{\gamma}(u+\frac{1}{\tau})^{\gamma}}\right)\widehat{\widetilde{P}}_F + \left(1 - \frac{1}{\tau^{\gamma}(u+\frac{1}{\tau})^{\gamma}}\right)\widehat{P}_0. \tag{4.50}$$

For small u this relationship reduces down after inverting the Fourier transform, to the following form in t-space,

$$\frac{\partial}{\partial t}P_R(x,t) = \gamma\frac{\partial}{\partial t}P_F(x,t). \tag{4.51}$$

This relationship amounts to the same description provided by Zbaeraev *et al,* and represents a steady fluctuation between states, where the exponent γ determines the ratio of the two. If the characteristic times are unequal ($\Delta \neq 0$) it is expected that they make an appearance in this expression. For short time scales the dynamics of the resting state are bolstered by a source term connected to the flight state, but reduced by an RL type derivative taken on the sum $P_F + P_0$. This is indicative of growth via the source but as time elapses this growth is stunted somewhat via this fractional derivative type term. This is especially the case in an initialized state of a Dirac delta function for the rest PDF and zero for the flight. Which is to say, the ensemble initiates in the rest state, therefore the source contribution far outweighs the *memory* term on small time scales. It's not until the flight state populates that the dynamics of the rest state are impeded. The full expression over all times can be expressed in the following way,

$$u\widehat{\widehat{P}}_R(k,u) = \frac{1}{\tau}\left(\widehat{\widehat{P}}_F + \widehat{P}_0\right) - \frac{1}{\tau}\left(\frac{1}{\tau^\gamma(u+\frac{1}{\tau})^\gamma}\right)\left(\widehat{\widehat{P}}_F + \widehat{P}_0\right) \tag{4.52}$$

$$\frac{\partial}{\partial t}P_R(x,t) = \frac{1}{\tau}\left(P_F + P_0\right) - \frac{1}{\tau}\frac{\partial}{\partial t}\left(\exp(-\frac{t}{\tau})\frac{t^{\gamma-1}}{\tau^\gamma}\right) * \left(P_F + P_0\right). \tag{4.53}$$

Again as outlined above in the long-time RL case, we see a balance between a P_F source term and a generalised derivative sink type term. For completeness we now outline the full form of the DE governing the dynamics of the LWWR model system. Rearranging Eq. (4.47), and carrying out both Laplace and Fourier inversions, the following differential equation is recovered,

$$\frac{1}{v^2}\left(\frac{\partial^2 P_\Sigma(x,t)}{\partial t^2} + \frac{2}{\tau}\frac{\partial P_\Sigma(x,t)}{\partial t} + \frac{P_\Sigma(x,t)}{\tau^2}\right) - \frac{1}{\tau^2\tau^{1+\gamma}}\left(\exp\left(-\frac{t}{\tau}\right)\frac{t^{\gamma-2}}{\Gamma\left(\gamma-1\right)}\right) * P_\Sigma(x,t)$$
$$= \frac{\partial^2 P(x,t)}{\partial x^2} + f_1(x,t). \tag{4.54}$$

The function $f_1(x,t)$ is defined as follows,

$$f_1(x,t) = \exp\left(-\frac{t}{\tau}\right)t^\gamma\left(\frac{P_0(x)}{t^2 v^2 \tau^{1+\gamma}} + P_{0,xx}(x)E_{1,1+\gamma}\left(\frac{t}{\tau}\right)\right) - P_{0,xx}(x) + P_0(x) \tag{4.55}$$

Eq. (4.54) bears the form of a generalised Telegraphers equation. The generalisation away from the more typical structure appears due to the convolution type source term, as well as the link to the initial condition via $f(x,t)$. Interestingly the original Telegraphers type equations can be rearranged to be the equation of Cattaneo, of which date back to 1948. The Cattaneo form is arrived at by modifying Ficks first law to include a time derivative of the probability current. The combination of this modified form with the continuity equation leads to the Telegraphers equation. The Cattaneo equation has been generalised to incorporate fractional derivatives, and several forms of this equations can be seen in the literature. The first generalisation was presented by Nonnenmacher [220], and this alongside two subsequent forms were outlined in the work of Compte and Metzler [221]. A fourth form was also put forward in a more recent survey published in 2020 by Metzler and Awad [222], within which they covered the prior three forms as well as some conditional solutions for these equations obtained as series over H-functions. They also noted the characteristic evolution of the second moment of the PDFs constructed from these equations which, depending on the type, tend to be some form of super diffusive behaviour which relaxes back to either Fickian or subdiffusive behaviour. The Cattaneo form of the generalised Telegraphers equation for our LWWR model can be expressed as,

$$\begin{aligned}&\frac{1}{v^2}\left(\frac{\partial}{\partial t}J(x,t)+\frac{2}{\tau}J(x,t)+\frac{1}{\tau^2}\int_0^t J(x,t'),dt'\right)-\frac{1}{v^2\tau^{1+\gamma}}\left(\exp\left(-\frac{t}{\tau}\right)t^{\gamma-1}E_{1,\gamma}(\frac{t}{\tau})\right)*J(x,t)\\&=-\frac{\partial^2 P_\Sigma(x,t)}{\partial x^2}+f_2(x,t),\end{aligned}\tag{4.56}$$

where $f_2(x,t)$ is defined by

$$\begin{aligned}f_2(x,t)=&\int P_0(x)dx\left(\exp\left(-\frac{t}{\tau}\right)\left(\frac{t^{2\gamma-4}}{v^4\tau^{1+2\gamma}}-\frac{t^{3\gamma-3}}{v^4\tau^{2+2\gamma}}+\frac{t^{\gamma-3}}{v^4\tau^{\gamma}}-\frac{t^{2\gamma}}{v^4\tau^{1+2\gamma}}+\frac{t^{2\gamma-3}}{v^4\tau^{2+2\gamma}}-\frac{t^{2\gamma-4}}{v^4\tau^{1+\gamma}}\right)\right)\\&-\frac{\partial P_0(x)}{\partial x}\exp\left(-\frac{t}{\tau}\right)t^{\gamma}E_{1,1+\gamma}\left(\frac{t}{\tau}\right).\end{aligned}\tag{4.57}$$

Eq. (4.56) deviates from the original form presented by Cattaneo, and also from the four generalisations. The deviations are the appearance of the integral over $J(x,t)$, as well as the occurrence of both the convolution with $J(x,t)$ and the intricate connection with the initial state of P_Σ which is encapsulated within $f_2(x,t)$. The convolution integral over $J(x,t)$ can actually be shown to be the sum of two Prahbakar type derivatives, which have been briefly studied within the context of generalised diffusion by Sandev *et al.* [223].

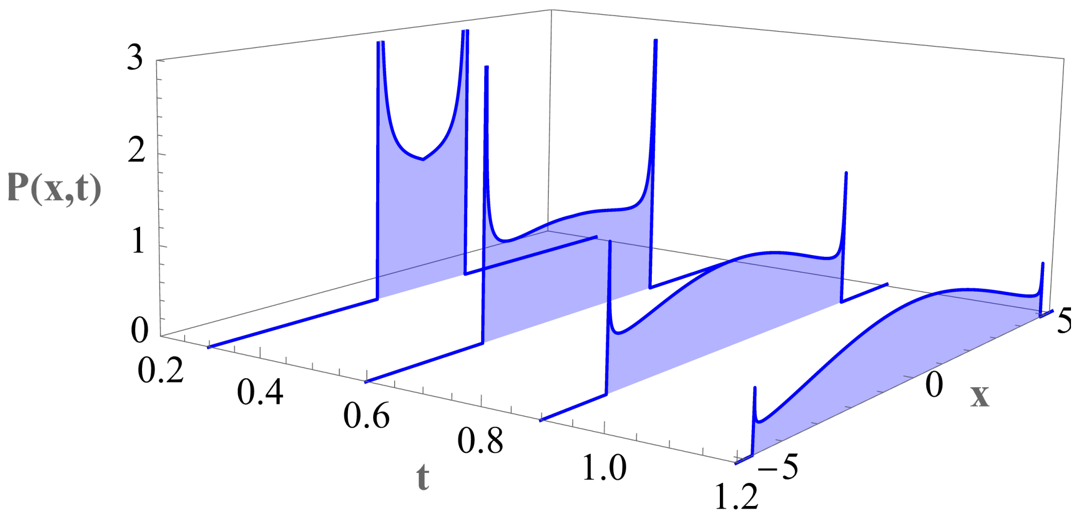

Figure 4.6: Time evolution of $P(x,t)$ from Eq. (4.40), $\gamma = 1/2, \tau = 1$. For early times the peaks at the boundary of the expansion are more pronounced, reflecting the fact that a larger portion of the ensemble is experiencing Lévy like motion for smaller times.

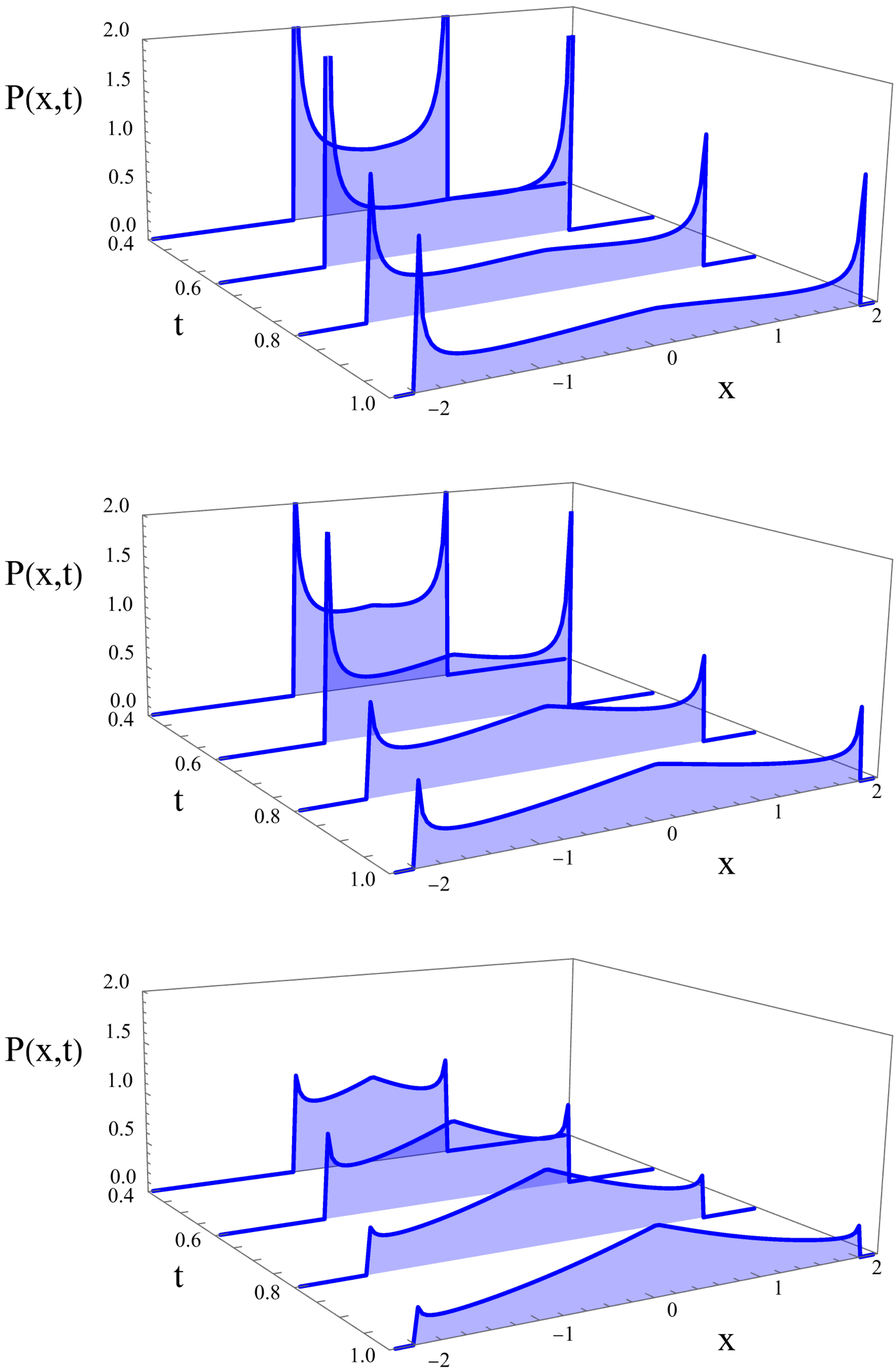

Figure 4.7: PDF solutions for $\Delta \neq 0$, in Eq. (4.46). Top to bottom corresponding to $\gamma = \frac{1}{4}, \frac{1}{2}, \frac{3}{4}$, respectively. $\tau_r = 2\tau$, $\tau = 1/2$, $\sigma = 1, \nu = \sigma/\tau$. The impact of τ_r and τ is to define the characteristic rest and flight periods, respectively. The interplay between τ_r and τ in this case where $\tau_r > \tau$ is to reveal the RL type features of the PDF solution within the bounds enforced by the Lévy coupling.

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Name of candidate:	Josiah Cleland
Name/title of Primary Supervisor:	Martin Williams
Name of Research Output and full reference:	
Generalised Fokker Planck equation with a stress redistribution type memory kernel	
In which Chapter is the Manuscript /Published work:	4
Please indicate:	
• The percentage of the manuscript/Published Work that was contributed by the candidate:	90
and	
• Describe the contribution that the candidate has made to the Manuscript/Published Work:	
Derivation of mathematical findings, writing up and producing figures.	
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Name/title of Primary Supervisor:	Martin Williams
Name of Research Output and full reference:	
Levy walk with gamma distributed rests	
In which Chapter is the Manuscript /Published work:	4
Please indicate:	
• The percentage of the manuscript/Published Work that was contributed by the candidate:	90
and	
• Describe the contribution that the candidate has made to the Manuscript/Published Work:	
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5

Coupled CTRW Model

This Chapter investigates the impact of a transient coupling in a CTRW between the PDF for displacements and the waiting time. The coupling is such that as the waiting time grows the coupling eventually dissipates. This coupling is proposed to be of physical relevance as it mimics the *storage* of energy prior to a disruptive event, the longer the period of internal stresses evolving the larger the potential displacement. However, the transient nature represents the fact the process has some limit as to how much storage is possible. The generalised diffusion equation (GDE) corresponding to this CTRW is then derived. The solution to this GDE is identified and the temporal behaviour of the second moment is presented. The asymptotic behaviour of the solution is explored, and the relaxation back to the uncoupled solution is demonstrated.

5.0.1 Coupled Continuous Time Random Walks

The CTRWs full description can be seen in the introductory Chapter. As alluded to, the coupling of the CTRW occurs through the distributions $\lambda(x)$ or $\omega(t)$, where they become the conditional distributions $\lambda(x|t)$ or $\omega(t|x)$. The precise manifestation of this conditional coupling has been explored in a few different ways historically [224–226]. One prominent approach is the so called *Lévy walks* framework which has been discussed in the previous Chapter. An alternative approach, first outlined in the discrete random walk of Wong *et al.* [227], has been extended in recent times by the work of Liu [228–230]. This particular coupling is introduced through the following conditional relationship for $\lambda(x|t)$,

$$\lambda(x|t) = \frac{1}{\sqrt{2\pi\sigma^2 g(t)}} \exp\left(-\frac{x^2}{2\sigma^2 g(t)}\right), \ g(t) > 0 \tag{5.1}$$

By taking the Fourier transform of this expression and considering the diffusion limit ($k << 1$), $\widehat{\lambda}(k|t)$ becomes,

$$\widehat{\lambda}(k|t) \sim 1 - \sigma^2 k^2 g(t), \quad \text{as} \quad k \to 0 \tag{5.2}$$

Combining this relationship with the Fourier-Laplace expression for the CTRW position PDF in Eq. (2.59)

$$\widehat{\widetilde{P}}(k,u) = \frac{1}{u} \frac{1}{1 + \sigma^2 k^2 \frac{\mathscr{L}[g(t)\omega(t)]}{1-\widetilde{\omega}(u)}}. \tag{5.3}$$

One interesting possibility of this mode of coupling, is that it allows for Fickian diffusive behaviour to be produced without the traditional decoupled signatures. Specifically, rather than requiring the waiting time to decay exponentially, it requires the following connection between $g(t)$ and $\omega(t)$

$$\omega(t) = \exp^{-\int_1^t \left(\frac{1-g'(t')}{g(t')}\right) dt'}, \tag{5.4}$$

where if $g(t)$ is a constant, then the waiting time distribution decays exponentially. However, in the coupled case, we can observe non-exponential waiting time decay whilst producing Fickian diffusion. A consequence of the interplay between the anomalous properties of the two underlying distributions is the coming together to produce linear growth of the second moment as well as Gaussian spreading.

5.0.2 Transient Coupling

The present Chapter explores a transient coupling of the jump distribution, $\lambda(x)$ to the waiting time, t. The coupling is such that the width of the distribution function for displacements grows with t, eventually reaching a constant width as the growth plateaus. The specific coupling function explored in this instance is,

$$g(t) = 1 - \exp(-t\alpha) \tag{5.5}$$

This function grows initially prior to tapering off, representative of the *decoupling* of the framework.

5.0.3 Generalised Diffusion Equation

Once again the waiting time distribution, $\omega(t)$ takes the form

$$\omega(t) = \frac{t^{\gamma-1}}{\tau^{\gamma}\Gamma(\gamma)} \exp\left(-\frac{t}{\tau}\right). \tag{5.6}$$

The influence of this particular waiting time distribution has been explored in Chapter 3. Rearranging and inverse transforming Eq. (5.3), with the respective functional forms of both $g(t)$ and $\omega(t)$ inserted, provides the following generalised diffusion equation:

$$\frac{\partial}{\partial t}P(x,t) = \frac{\partial}{\partial t}\int_0^t \exp\left(-\frac{t'}{\tau}\right)\sum_{n=0}^{\infty}\alpha\frac{(-\alpha)^n\Gamma(n+\gamma+1)}{\Gamma(n+2)\Gamma(\gamma)\tau^{\gamma}}t'^{n+\gamma}E_{\gamma,1+n+\gamma}\left(\frac{t'}{\tau}\right)\frac{\partial^2}{\partial x^2}P(x,t-t')dt'. \tag{5.7}$$

The memory kernel contained in Eq. (5.7) is plotted in Fig. 5.1 in black. For small t it grows indicating the past is weighted more heavily than the present, a consequence of the fact that within the time $t < \frac{1}{\alpha}$ a portion of the ensemble will be diffusively enhanced via the coupling built into the underlying CTRW. Thus, the contributions in dynamics from the coupled regime have a strong influence on present dynamics, but this connection fades and the decay to a constant value, as observed in the decoupled case.

5.0.4 Second Moment

The corresponding second moment for the PDF appears in u space as,

$$\widetilde{\langle x^2\rangle}(u) = \frac{1}{u}\frac{\mathscr{L}\{g(t)\omega(t)\}(u)}{1-\widetilde{\omega}(u)}. \tag{5.8}$$

In order to evaluate this second moment the Laplace transform of the right hand side must be evaluated

$$\mathscr{L}\{g(t)\omega(t)\}(u) = \mathscr{L}\left\{\left(1-\exp(-t\alpha)\right)\left(\frac{t^{\gamma-1}}{\tau^{\gamma}\Gamma(\gamma)}\exp\left(-\frac{t}{\tau}\right)\right)\right\}(u). \tag{5.9}$$

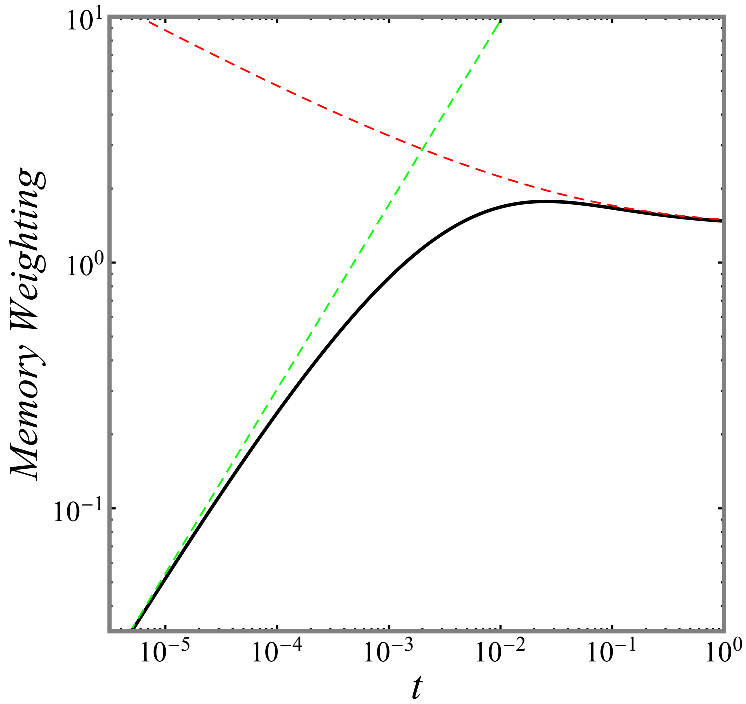

Figure 5.1: Comparison of the Riemann Liouville memory kernel (red) and memory kernel of Eq. (5.7) (black). Here $\tau = \sigma = 1, \alpha = 15$ and $\gamma = \frac{1}{2}$. The green dashed line represents the small t power law growth of the kernel, with the exponent gamma. Powerlaw *growth* in a memory kernel is indicative of super-diffusive behaviour as it implies the process is being enhanced in some manner proportional to its duration. The coupling and enhanced nature exists for a time period defined in association with the parameter α, after which the RL memory kernel form is maintained for some period prior to an eventual relaxation to a constant value. The duration of the intermittent RL memory kernel is confined by the relative values for α and τ.

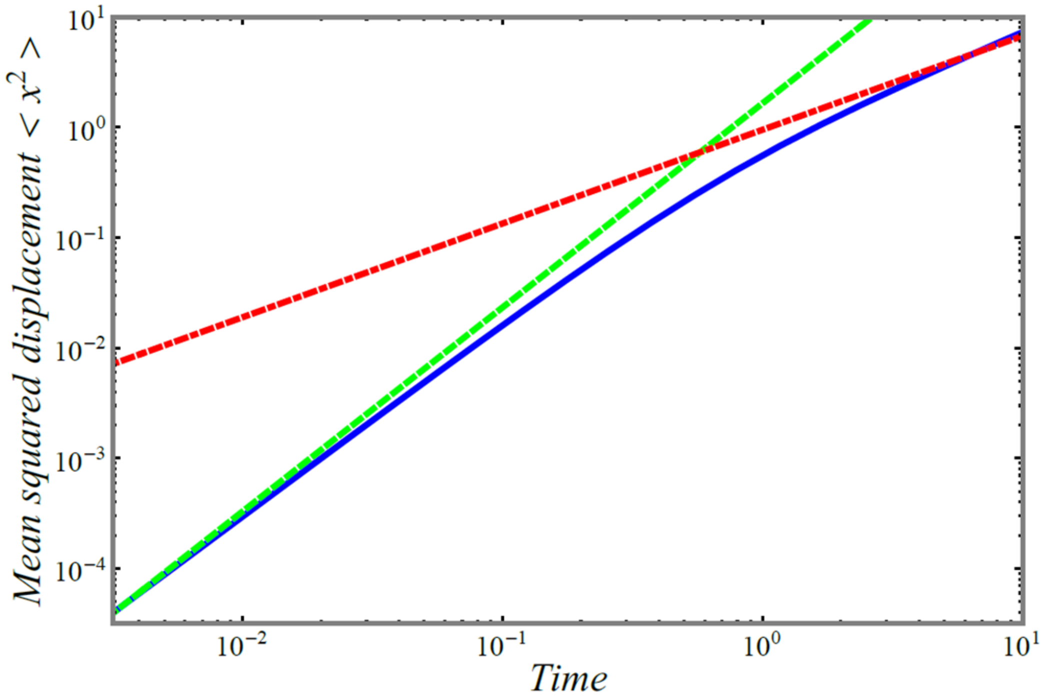

Figure 5.2: Second moment behaviour for $P(x,t)$, found in Eq. (5.11). With $\tau = \sigma = 1, \alpha = 2$ and $\gamma = 1/2$. The green dashed line reflects growth according to $1+\gamma$ whilst the red dashed line represents linear growth in time. The duration of the enhanced diffusion characteristics is associated with the parameter α, likewise the following sub-diffusive regime exists for a duration associated with τ prior to an eventual relaxation to Fickian behaviour for longer time scales.

Expressing the exponential function in it's series form, and adjusting the summation leaves

$$\mathscr{L}\{g(t)\omega(t)\}(u) = -\sum_{n=0}^{\infty} \mathscr{L}\left\{\frac{(-t\alpha)^{n+1}}{\Gamma(n+2)}\left(\frac{t^{\gamma-1}}{\tau^{\gamma}\Gamma(\gamma)}\exp\left(-\frac{t}{\tau}\right)\right)\right\}(u). \tag{5.10}$$

Substituting this back into the expression for $\langle x^2\rangle(t)$, and evaluating the Laplace transform gives

$$\langle x^2\rangle(t) = \exp\left(-\frac{t}{\tau}\right)\sum_{n,k=0}^{\infty} \alpha \frac{(-\alpha)^n\Gamma(n+\gamma+1)}{\Gamma(n+2)\Gamma(\gamma)\tau^{\gamma}} t^{1+n+\gamma} E_{\gamma,2+n+\gamma}\left(\frac{t^{\gamma}}{\tau^{\gamma}}\right) \tag{5.11}$$

In Fig.(5.2) the behaviour of the second moment is displayed. The second moment evolves from an initial characteristic exponent of $1+\gamma$ to eventually relaxing back to a slope of 1 as time elapses. Depending on the relationship between α and τ, there is some suggestion from the memory kernel that a fleeting sub-diffusive regime may exist.

5.0.5 Probability Density Function

The investigation into the t-x space form of the PDF begins with the Weiss equation, which can be expressed in the form of an H-function as,

$$\widehat{\widetilde{P}}(k,u) = \frac{1}{u} H^{1,1}_{1,1}\left[\frac{k^2\sigma^2 \mathscr{L}\{g(t)\omega(t)\}}{1-\widetilde{\omega}(u)} \Bigg|^{(0,1)}_{(0,1)} \right]. \tag{5.12}$$

Substituting the functional forms for $g(t), \omega(t)$ and $\widetilde{\omega}(u)$,

$$\widetilde{P}(k,u) = \mathscr{L}^{-1}\left[\frac{1}{u} H^{1,1}_{1,1}\left[\frac{k^2\sigma^2\left(1-\frac{(u+\frac{1}{\tau})^{\gamma}}{(u+\frac{1}{\tau}+\alpha)^{\gamma}}\right)}{(u+\frac{1}{\tau})^{\gamma}-\frac{1}{\tau^{\gamma}}} \Bigg|^{(0,1)}_{(0,1)} \right]\right]. \tag{5.13}$$

Evaluating the inverse cosine Fourier transform and inverting the argument of the H-function,

$$P(x,t) = \frac{1}{2\pi}\mathscr{L}^{-1}\left[\frac{\sqrt{\pi}}{u|x|} H^{2,0}_{0,2}\left[\frac{x^2((u+\frac{1}{\tau})^{\gamma}-\frac{1}{\tau^{\gamma}})}{4\sigma^2\left(1-\frac{(u+\frac{1}{\tau})^{\gamma}}{(u+\frac{1}{\tau}+\alpha)^{\gamma}}\right)} \Bigg|_{(1/2,1),(1,1)} \right]\right] \tag{5.14}$$

The Legendre duplication formula is now invoked, in order to combine to gamma functions present in the H-function structure. This modification is followed by the expansion (formula II in appendix D) of the H-function, with $\eta = (1-\frac{1}{u^{\gamma}\tau^{\gamma}})^{1/2}$,

$$\begin{aligned} P(x,t) =& \frac{1}{2}\exp\left(-\frac{t}{\tau}\right)\mathscr{L}^{-1}\Bigg[\frac{1}{\left(u-\frac{1}{\tau}\right)} \frac{u^{\gamma/2}}{\sigma\left(1-\frac{(u)^{\gamma}}{(u+\alpha)^{\gamma}}\right)^{1/2}} \\ & \sum_{m=0}^{\infty}\left(-\frac{1}{u^{\gamma}\tau^{\gamma}}\right)^{m} H^{1,1}_{1,2}\left[\frac{|x|u^{\gamma/2}}{\sigma\left(1-\frac{(u)^{\gamma}}{(u+\alpha)^{\gamma}}\right)^{1/2}} \Bigg|^{(-1/2,1/2)}_{(0,1),(m-1/2,1/2)} \right]\Bigg]. \end{aligned} \tag{5.15}$$

The next modification makes use of an expansion formula (formula III in appendix D) once more,with $\eta = \frac{1}{\left(1-\frac{(u)^{\gamma}}{(u+\alpha)^{\gamma}}\right)^{1/2}}$, its use constrains the values of u in the following fashion,

$$\Re\left\{ \frac{1}{\left(1-\frac{(u)^{\gamma}}{(u+\alpha)^{\gamma}}\right)} \right\} > 1/2, \tag{5.16}$$

which can be re-written as,

$$\Re\{u/\alpha\} > \frac{\cos\left(\frac{\pi}{\gamma}\right)\left(1-\cos\left(\frac{\pi}{\gamma}\right)\right)-\sin^2\left(\frac{\pi}{\gamma}\right)}{\left(1-\cos\left(\frac{\pi}{\gamma}\right)\right)^2+\sin^2\left(\frac{\pi}{\gamma}\right)}, \tag{5.17}$$

which holds for any $\alpha > 0$. In addition to the use of this expansion formula mentioned above, the removal of $\exp(-\alpha t)$ through the shift theorem and an additional expansion formula (formula II in appendix D) with $\eta = (1-\frac{\alpha}{u})^{\gamma/2}$ are applied. All this leaves the following form,

$$\begin{aligned} P(x,t) =& \frac{1}{4}\exp\left(-t\left(\alpha+\frac{1}{\tau}\right)\right)\mathscr{L}^{-1}\Bigg[\frac{1}{u}\sum_{k=0}^{\infty}\left(\frac{\left(\alpha+\frac{1}{\tau}\right)}{u}\right)\frac{(u-\alpha)^{\gamma/2}}{\sigma}\left(1-\frac{((u-\alpha))^{\gamma}}{(u)^{\gamma}}\right) \\ & \sum_{j,m,r=0}^{\infty}\left(-\frac{1}{u^{\gamma}\tau^{\gamma}}\right)^m\left(-\frac{\alpha}{u}\right)^j\frac{1}{\Gamma\left(j+1\right)}\frac{1}{\Gamma\left(m+1\right)}\left(\frac{(u-\alpha)^{\gamma}}{(u)^{\gamma}}\right)^r\frac{1}{\Gamma\left(r+1\right)} \\ & H^{1,2}_{2,3}\left[\frac{|x|u^{\gamma/2}}{\sigma}\middle|^{(\gamma m,\gamma/2),(-r-1/2,1/2)}_{(0,1),(m-1/2,1/2)(\gamma m+j,\gamma/2)}\right]\Bigg](t). \end{aligned} \tag{5.18}$$

The final step prior to the inversion of the Laplace transform is the employment of the binomial theorem for the factors containing $(1-\frac{\alpha}{u})$,

$$\begin{aligned} P(x,t) =& \frac{1}{4}\exp\left(-t\left(\alpha+\frac{1}{\tau}\right)\right)\mathscr{L}^{-1}\Bigg[\frac{1}{u}\sum_{k=0}^{\infty}\left(\frac{\left(\alpha+\frac{1}{\tau}\right)}{u}\right)^k\frac{1}{\sigma}u^{\gamma/2} \\ & \sum_{n=0}^{\infty}\left(-\frac{\alpha}{u}\right)^n\frac{1}{\Gamma\left(n+1\right)}\left(\frac{\Gamma\left(\gamma r+\gamma/2+1\right)}{\Gamma\left(\gamma r+\gamma/2+1-n\right)}-\frac{\Gamma\left(\gamma r+3\gamma/2+1\right)}{\Gamma\left(\gamma r+3\gamma/2+1-n\right)}\right) \\ & \sum_{j,m,r=0}^{\infty}\left(-\frac{1}{u^{\gamma}\tau^{\gamma}}\right)^m\left(-\frac{\alpha}{u}\right)^j\frac{1}{\Gamma\left(j+1\right)}\frac{1}{\Gamma\left(m+1\right)}\frac{1}{\Gamma\left(r+1\right)} \\ & H^{1,2}_{2,3}\left[\frac{|x|u^{\gamma/2}}{\sigma}\middle|^{(\gamma m,\gamma/2),(-r-1/2,1/2)}_{(0,1),(m-1/2,1/2)(\gamma m+j,\gamma/2)}\right]\Bigg](t). \end{aligned} \tag{5.19}$$

Evaluating the inverse Laplace transform and multiplying by a factor of 2, due to the evaluation of the integral over half the x space due to symmetry.

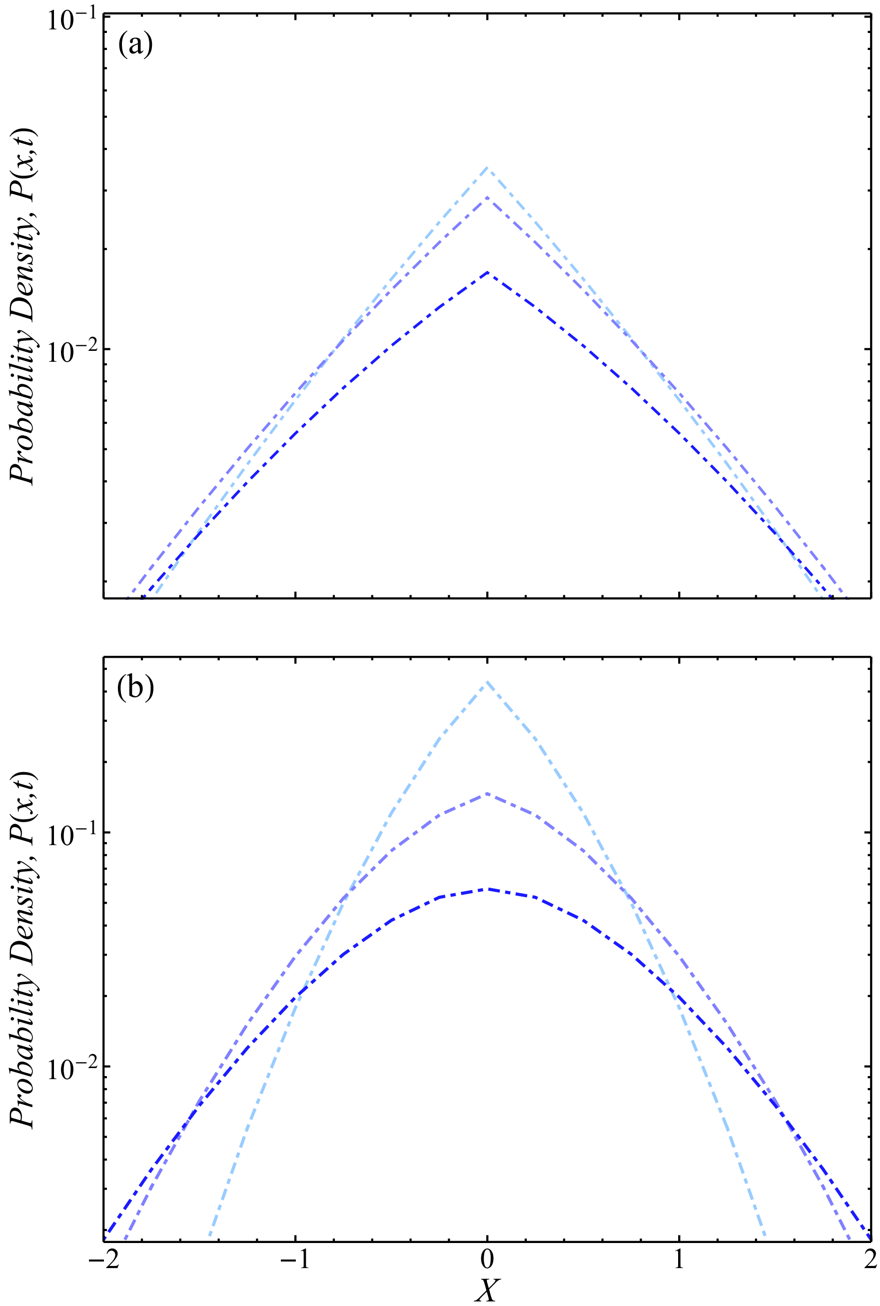

Figure 5.3: PDF plot corresponding to the solution to Eq. (5.7). Plot (a) corresponding to $\gamma = 1/4$, whilst (b) to $\gamma = 3/4$. In both cases $\tau = 1, \alpha = 5$. What is apparent from the comparison between the two plots is the more rapid spreading of the ensemble in the case of a larger value for gamma.

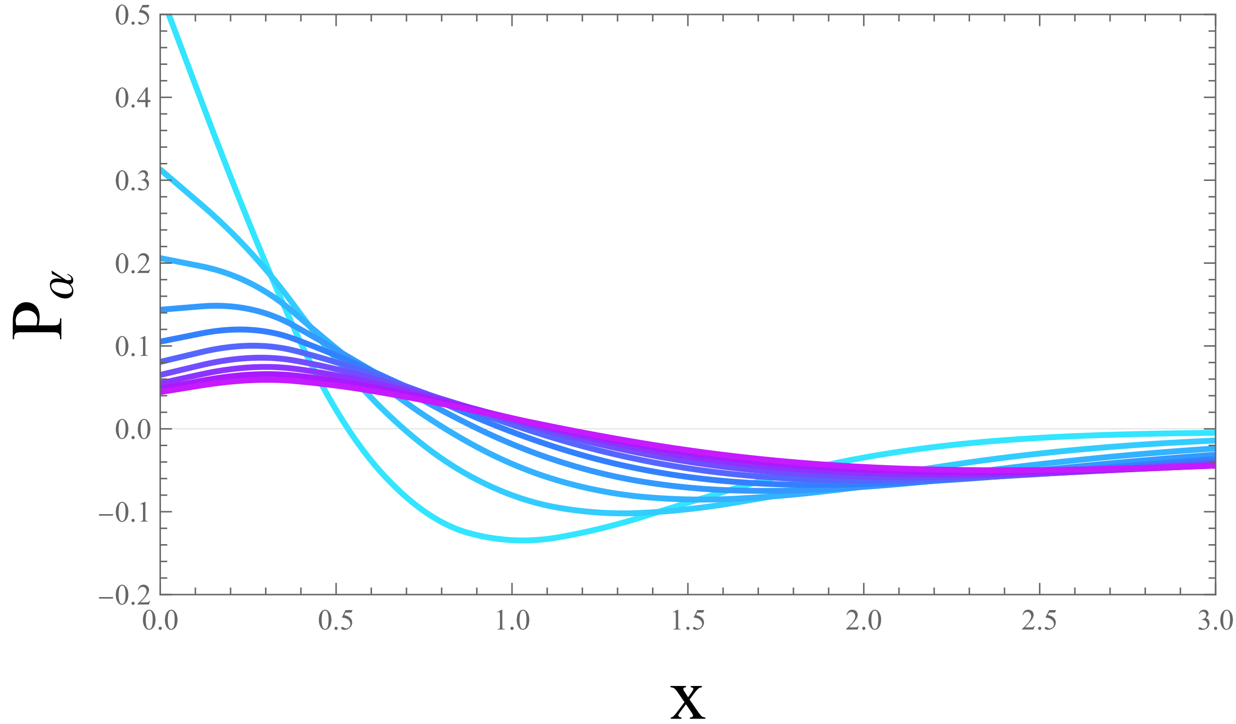

Figure 5.4: $P_\alpha(x,t)$ ranging between $t = .05$ (cyan) to $t = .5$ (purple), as outlined in Eq.(5.22). The trend overall in the plot is that the magnitude of P_α reduces with increasing t, reflecting the decoupling of the system. In general the impact of the coupling appears to relocate PD closer to the origin. However, since the PDF still expands super-diffusively this redistribution of PD results in a broader shape about the origin.

$$P(x,t) = \exp\left(-t\left(\alpha+\frac{1}{\tau}\right)\right)\frac{1}{2\sigma t^{\gamma/2}}\sum_{k,n,m,j,r=0}^{\infty}\left(\left(\alpha+\frac{1}{\tau}\right)t\right)^{k}$$
$$\frac{(-\alpha t)^n}{\Gamma(n+1)}\left(\frac{\Gamma\left(\gamma r+\gamma/2+1\right)}{\Gamma\left(\gamma r+\gamma/2+1-n\right)}-\frac{\Gamma\left(\gamma r+3\gamma/2+1\right)}{\Gamma\left(\gamma r+3\gamma/2+1-n\right)}\right)$$
$$\frac{(-\alpha t)^j}{\Gamma\left(j+1\right)}\frac{\left(-\frac{t^\gamma}{\tau^\gamma}\right)^m}{\Gamma(m+1)}\frac{1}{\Gamma(r+1)}H_{3,3}^{1,2}\left[\frac{|x|}{\sigma t^{\gamma/2}}\middle|_{(0,1),(m-1/2,1/2)(\gamma m+j,\gamma/2)}^{(\gamma m,\gamma/2),(-r-1/2,1/2)(1+k+n+\gamma m+j-\gamma/2,\gamma/2)}\right] \tag{5.20}$$

5.0.6 Coupling Impact

The solution to the diffusion equation associated with the small k approximation of a CTRW with a Γ distribution for the waiting time PDF has been identified in Chapter 3, in the absence of coupling. This result (Eq. (5.20)) can be extracted from the obtained form for the PDF solution discussed in Chapter 3,

$$P(x,t) = \exp\left(-\frac{t}{\tau}\right)\mathscr{L}^{-1}\left[\frac{1}{\left(u-\frac{1}{\tau}\right)}\frac{(u)^{\gamma/2}}{\sigma}\sum_{m,r=0}^{\infty}\left(-\frac{1}{(u)^{\gamma}\,\tau^{\gamma}}\right)^{m}\frac{1}{\Gamma(m+1)}\right.$$
$$\left. H_{1,2}^{1,1}\left[\frac{|x|\,(u)^{\gamma}}{\sigma}\middle|_{(0,1),(m-1/2,1/2)}^{(-1/2,1/2)}\right]\right](t) + P_{\alpha}(x,t) \tag{5.21}$$

$$P_{\alpha}(x,t) = \exp\left(-t\left(\alpha+\frac{1}{\tau}\right)\right)\frac{1}{2\sigma t^{\gamma/2}}\sum_{r,k,n,m,j=0}^{\infty}\left(\left(\alpha+\frac{1}{\tau}\right)t\right)^{k}\frac{(-\alpha t)^{n}}{\Gamma(n+1)}\frac{(-\alpha t)^{j}}{\Gamma(j+1)}$$
$$\frac{\left(-\frac{t^{\gamma}}{\tau^{\gamma}}\right)^{m}}{\Gamma(m+1)}\frac{\Gamma(\gamma r+3\gamma/2+1)}{\Gamma(\gamma r+3\gamma/2+1-n)}$$
$$\left(\frac{1}{\Gamma(r+2)}H_{3,3}^{1,2}\left[\frac{|x|}{\sigma t^{\gamma/2}}\middle|_{(0,1),(m-1/2,1/2)(\gamma m+j,\gamma/2)}^{(\gamma m,\gamma/2),(-r-3/2,1/2)(1+k+n+\gamma m+j-\gamma/2,\gamma/2)}\right]\right.$$
$$\left.-\frac{1}{\Gamma(r+1)}H_{3,3}^{1,2}\left[\frac{|x|}{\sigma t^{\gamma/2}}\middle|_{(0,1),(m-1/2,1/2)(\gamma m+j,\gamma/2)}^{(\gamma m,\gamma/2),(-r-1/2,1/2)(1+k+n+\gamma m+j-\gamma/2,\gamma/2)}\right]\right). \tag{5.22}$$

$P_{\alpha}(x,t)$ represents the redistribution of probability density due to the new dynamics introduced through the coupling and it's integral over x is zero as expected (this can be shown by using the Mellin transform properties of the H-function [179]). In Fig. 5.4 we can see how the introduction of coupling has adjusted the allocation of probability density across the position variable x. We see in earlier times (cyan) a bolstering of central peak, and an increase of PD about the shoulders of the peak. This is at the expense of the wings of the position PDF. This effect ultimately decays away rapidly as the framework self decouples.

5.0.7 Long Time Asymptotic behaviour

We expect $P_{\alpha} \to 0$ for large t. Expanding up the H functions into series form then constructing the H-function associated with the sum over "n" followed by taking the asymptotic behaviour for that function as $t >> \alpha$ reveals exponential decay sensitive to α. So as time elapses the only contribution that remains is the solution to the decoupled equation discussed previously. In the instance that the decoupled behaviour dies away first we still see the same asymptotic form for P_{α} though now the underlying decoupled solution is already in its Gaussian form as discussed in

Chapter 3 is

$$P_\alpha(x,t)=\exp\left(-t\left(\alpha+\frac{1}{\tau}\right)\right)\frac{1}{2\sigma t^{\gamma/2}}\sum_{r,k,n,m,j=0}^{\infty}\left(\left(\alpha+\frac{1}{\tau}\right)t\right)^{k}\frac{(-\alpha t)^{n}}{\Gamma(n+1)}\frac{(-\alpha t)^{j}}{\Gamma\left(j+1\right)}$$
$$\frac{\left(-\frac{t^{\gamma}}{\tau^{\gamma}}\right)^{m}}{\Gamma(m+1)}\frac{\Gamma\left(\gamma r+3\gamma/2+1\right)}{\Gamma\left(\gamma r+3\gamma/2+1-n\right)}$$
$$\sum_{p=0}^{\infty}\frac{\Gamma\left(1-\gamma m+\gamma p/2\right)}{\Gamma\left(p+1\right)\Gamma\left(3/2-m+p/2\right)\Gamma\left(1-j-\gamma m+\gamma p/2\right)}$$
$$\frac{1}{\Gamma\left(1+k+n+\gamma m+j-\gamma/2-\gamma p/2\right)}$$
$$\left(-\frac{|x|}{\sigma t^{\gamma/2}}\right)^{p}\left(\frac{\Gamma\left(5/2+r+p/2\right)}{\Gamma(r+2)}-\frac{\Gamma\left(3/2+r+p/2\right)}{\Gamma(r+1)}\right). \tag{5.23}$$

Compressing the series over n into an H-function structure leaves,

$$P_\alpha(x,t)=\exp\left(-t\left(\alpha+\frac{1}{\tau}\right)\right)\frac{1}{2\sigma t^{\gamma/2}}\sum_{r,k,m,j=0}^{\infty}\left(\left(\alpha+\frac{1}{\tau}\right)t\right)^{k}\frac{(-\alpha t)^{j}}{\Gamma\left(j+1\right)}\frac{\left(-\frac{t^{\gamma}}{\tau^{\gamma}}\right)^{m}}{\Gamma(m+1)}$$
$$\sum_{p=0}^{\infty}\frac{\Gamma\left(\gamma r+3\gamma/2+1\right)\Gamma\left(1-\gamma m+\gamma p/2\right)}{\Gamma\left(p+1\right)\Gamma\left(3/2-m+p/2\right)\Gamma\left(1-j-\gamma m+\gamma p/2\right)}$$
$$\left(-\frac{|x|}{\sigma t^{\gamma/2}}\right)^{p}\left(\frac{\Gamma\left(5/2+r+p/2\right)}{\Gamma(r+2)}-\frac{\Gamma\left(3/2+r+p/2\right)}{\Gamma(r+1)}\right)$$
$$H_{1,2}^{1,0}\left[\alpha t\left|\begin{matrix}(\gamma r+3\gamma/2+1,1)\\ \\(0,1)(\gamma/2-k-\gamma m-j+\gamma p/2,1)\end{matrix}\right.\right]. \tag{5.24}$$

Plugging in the asymptotic behaviour for the H-function for $t >> \alpha$, leaves the form

$$P_\alpha(x,t)=\exp\left(-t\left(2\alpha+\frac{1}{\tau}\right)\right)\frac{1}{2\sigma t^{3\gamma/2+1}}\sum_{r,k,m,j=0}^{\infty}\left(\frac{1}{\alpha t}\right)^{\gamma r}\left(\left(1+\frac{1}{\alpha\tau}\right)\right)^{k}$$
$$\frac{(-1)^{j}}{\Gamma\left(j+1\right)}\frac{\left(-\frac{\alpha^{\gamma}}{\tau^{\gamma}}\right)^{m}}{\Gamma(m+1)}\sum_{p=0}^{\infty}\frac{\Gamma\left(\gamma r+3\gamma/2+1\right)\Gamma\left(1-\gamma m+\gamma p/2\right)}{\Gamma\left(p+1\right)\Gamma\left(3/2-m+p/2\right)\Gamma\left(1-j-\gamma m+\gamma p/2\right)}$$
$$\left(\frac{-|x|\alpha^{\gamma/2}}{\sigma}\right)^{p}\left(\frac{\Gamma\left(5/2+r+p/2\right)}{\Gamma(r+2)}-\frac{\Gamma\left(3/2+r+p/2\right)}{\Gamma(r+1)}\right). \tag{5.25}$$

Because we have large t we can further simplify the expression by considering only the dominant term $r=0$

$$P_\alpha(x,t) = \exp\left(-t\left(2\alpha+\frac{1}{\tau}\right)\right)\frac{1}{2\sigma t^{3\gamma/2+1}}\sum_{k,m,j=0}^{\infty}\left(\left(1+\frac{1}{\alpha\tau}\right)\right)^k$$
$$\frac{(-1)^j}{\Gamma\left(j+1\right)}\frac{\left(-\frac{\alpha^\gamma}{\tau^\gamma}\right)^m}{\Gamma\left(m+1\right)}\sum_{p=0}^{\infty}\frac{\Gamma\left(3\gamma/2+1\right)\Gamma\left(1-\gamma m+\gamma p/2\right)}{\Gamma\left(p+1\right)\Gamma\left(3/2-m+p/2\right)\Gamma\left(1-j-\gamma m+\gamma p/2\right)}$$
$$\left(\frac{-|x|\alpha^{\gamma/2}}{\sigma}\right)^p\left(\frac{\Gamma\left(5/2+p/2\right)}{\Gamma\left(2\right)}-\Gamma\left(3/2+p/2\right)\right) \tag{5.26}$$

5.1 Discussion

This Chapter explored the implications of transient coupling between the jump probability density function and the waiting time within a CTRW framework. The implications were considered initially in terms of the generalised diffusion equation. The memory kernel was identified and plotted in Fig. 5.1, where it was compared against the memory kernel in the case of no coupling [196]. The memory kernel corresponding to Eq. (5.7) attributes relatively large weights to contributions within the coupled regime, prior to relaxing to a decay in line with the uncoupled case as obtained in Chapter 3. The kernel then continues its decay to a constant value, which is indicative of a relaxation to Fickian dynamics. The implications of this behaviour in the memory kernel is realised within the MSD as outlined in Fig. 5.2. The characteristic diffusion exponent transitions from a regime of super-diffusive behaviour towards return to a linear growth. The enhanced diffusion is generated as a result of the coupling in the underlying CTRW, and this fades to reveal a return to linear growth. The full solution to the generalised diffusion equation, Eq. (5.20) was obtained through the use of the Fox H-function. The solution required the employment of both transformation properties of this special function as well as a variety of general properties of the H-function. The resulting solution was expressed as several infinite series over Fox H-functions, and is displayed in Fig. 5.3. The solution includes the anomalous parameter gamma which regulates the nature of the anomalous waiting time in the CTRW, if $\gamma \to 1$ the waiting time distribution decays exponentially on all timescales. The characteristic coupling time α occurs as well, it provides a measure for how quickly the coupling decay occurs. Given the prior results of the present CTRW framework in the absence of coupling highlighted in Chapter 3, the impact of coupling was able to be explicitly isolated. The coupling impact was labeled P_α and its asymptotic behaviour was explored. It was identified that this contribution does indeed decay away to zero, in an exponential manner sensitive to α as demonstrated in the exponential decay factor in Eq. (5.26). The decay of this portion leaves the decoupled form as described in Chapter 3.

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STATEMENT OF CONTRIBUTION
DOCTORATE WITH PUBLICATIONS/MANUSCRIPTS

We, the candidate and the candidate's Primary Supervisor, certify that all co-authors have consented to their work being included in the thesis and they have accepted the candidate's contribution as indicated below in the *Statement of Originality*.

Name of candidate:	Josiah Cleland
Name/title of Primary Supervisor:	Martin Williams
Name of Research Output and full reference:	
Transiently coupled CTRW and corresponding diffusion equation	
In which Chapter is the Manuscript /Published work:	5
Please indicate:	
• The percentage of the manuscript/Published Work that was contributed by the candidate:	90
and	
• Describe the contribution that the candidate has made to the Manuscript/Published Work:	
Derivation of mathematical findings, writing up and producing figures.	
For manuscripts intended for publication please indicate target journal:	
Physical Review E	
Candidate's Signature:	Josiah Cleland Digitally signed by Josiah Cleland Date: 2022.04.12 16:49:43 +12'00'
Date:	
Primary Supervisor's Signature:	Williams, Martin Digitally signed by Williams, Martin Date: 2022.04.12 14:01:50 +12'00'
Date:	

(This form should appear at the end of each thesis chapter/section/appendix submitted as a manuscript/ publication or collected as an appendix at the end of the thesis)

6

Future work

6.0.1 Summary

In this thesis we constructed three frameworks, each of which can encode aspects of both temporal and spatial features of the dynamics in stress-redistributing (SR) systems, with polymer networks in mind. The universality of the timing of SR events was used to construct a new generalised diffusion equation and for the first time provide a detailed description of it's form and asymptotic behaviour using the Fox H-function and its properties. A new Lévy walks with rests (LWWR) framework was then constructed which incorporated the SR waiting time PDF. The LWWR model's position PDF and second moment were extracted using the Fox H-function properties. The asymptotic expansion of the LWWR model's solution was given, and the generalised diffusion equation (GDE) form was demonstrated. The connection between the GDE and generalised forms of the Cattaneo and Telegraphers equations was also discussed. The final model proposed a new mode of transient coupling between the displacements and waiting times in the CTRW. The position PDF and second moment were obtained using transformation methods and theorems relating to the Fox H-function. It is our hope that these solutions and frameworks will inform and stimulate future work in the interpretation of non-equilibrium quaking events and their manifold effects.

6.0.2 Future Work

There are several different paths for possible future extensions to this work: either continuing on the theoretical side, probing the model behaviours with simulations or testing the connections of the models with experimental results. On the theoretical front we believe that it would be of interest to extend the results to consider the *ergodic* properties of these frameworks. Many works on the topic have provided insight into the particular nature of ergodicity breaking that occurs within frameworks of a similar kind [231]. There are also interesting avenues to explore in extensions to the LWWR frameworks, such as those which explore the implications of *distributed velocities* [232]. Distributed velocities may be a more physically relevant mode of modelling, in contrast to the constant magnitude velocity included in Chapter 4. Simulations have proved to be a useful tool in other works and serve as a somewhat pseudo experimental perspective. There may be benefits to employing a simulation approach to bridge the gap between the analytic and the experimental. Experimentally it would be of great benefit to gather data on the waiting time PDF for the quaking phenomena and to do the same also for the displacements. In addition, more precise Van Hove plots (position PDFs) would allow limits to be placed on relevant parameter values. Many of the parameters are tied in with the displacement and waiting time distributions and accessing that information would experimentally fix these parameter values and ensure accurate model evaluation. These experimental investigations will aid greatly in reducing the current flexibility of the model parameters. Preliminary predictions for the model parameters can be generated from some existing data and these value will be discussed in the following subsection.

6.0.3 Parameter Predictions

Parameter	Value
α	0.75
k	$20\ \frac{pN}{\mu m}$
τ_α	0.0005 s

Table 6.1: Parameters from the Brownian contribution described in Chapter 3. These parameters originate from the FFPE formalism, α being the characteristic exponent present in the fractional derivative, k the spring constant associated with the gel network and τ_α the characteristic time associated with relaxation in this framework.

Taking the model proposed in Chapter 3, we can attempt to fit the experimental MSD data obtained in [9]. The Brownian component of the diffusion within the gel network was proposed to be well modelled by the FFPE in the instance of a linear restoring force and the extracted parameters associated with the modelling of this component are included in Table 6.1. The parameter α governs the characteristic exponent for small t, as such it is constrained by experimental values to be 0.75, a value known to be associated with small deformations of polymer chains. The parameter k represents the spring-constant associated with the gel network and although the value of $20\frac{pN}{\mu m}$ seems small when comparing to *stretching* experiments, carried out on individual polymers (that might be an order of magnitude or two higher) the deformations occurring are unlikely to be directly equivalent to a straightforward longitudinal stretching experiment and some difference here isn't too surprising. The parameter τ_α represents the characteristic time associated with a relaxation from some initial MSD value to the *thermal* equilibrium value [170].

The SR component of the system manifest at longer times can be modelled using the framework derived in Chapter 3 and the parameters required for the successful capturing of the experimental data are found in Table 6.2. The parameter γ reflects the degree of short timescale correlations between quaking events, smaller values reflecting a greater deviation away from random behaviour. The value of γ identified here is 0.275 which is less than half the figure identified for tectonic event timings (0.60) [149], but is larger than the value of a traditional block-slider model which has $\gamma = 0$ [233]. The characteristic time τ_γ identified here is 170s, the mean time between events, $\langle t \rangle$ is given by (for the Γ distribution)

$$\langle t \rangle = \gamma \tau_\gamma = 46.75s. \tag{6.1}$$

Therefore, the expected time between redistribution events in the gel network is 46.75s. Finally we have the generalised diffusion coefficient D_γ which takes the value $3.9 \times 10^{-17} \frac{m^2}{t^\gamma}$, which is in the range of normal diffusion coefficients for glassy polymers. These parameters, determined through the fitting of acquired MSD data-allow for the prediction of the functional forms of the both the position PDF for particles confined within the gel, as well as for the waiting time PDF of the SR events themselves. Future studies on experimental PDF forms and the waiting time PDFs for the quaking events will allow the predictions of these models to be tested.

Parameter	Value
γ	0.275
τ_γ	170 s
D_γ	$3.9\times10^{-17}\frac{m^2}{t^\gamma}$

Table 6.2: Parameters from the SR contribution described in Chapter 3, γ is the characteristic exponent of the Γ distribution associated with correlations between successive quake events, τ_γ is the characteristic time upon which quakes transition from correlated to uncorrelated and D_γ is the generalised diffusion coefficient associated with transport driven by the SR dynamic.

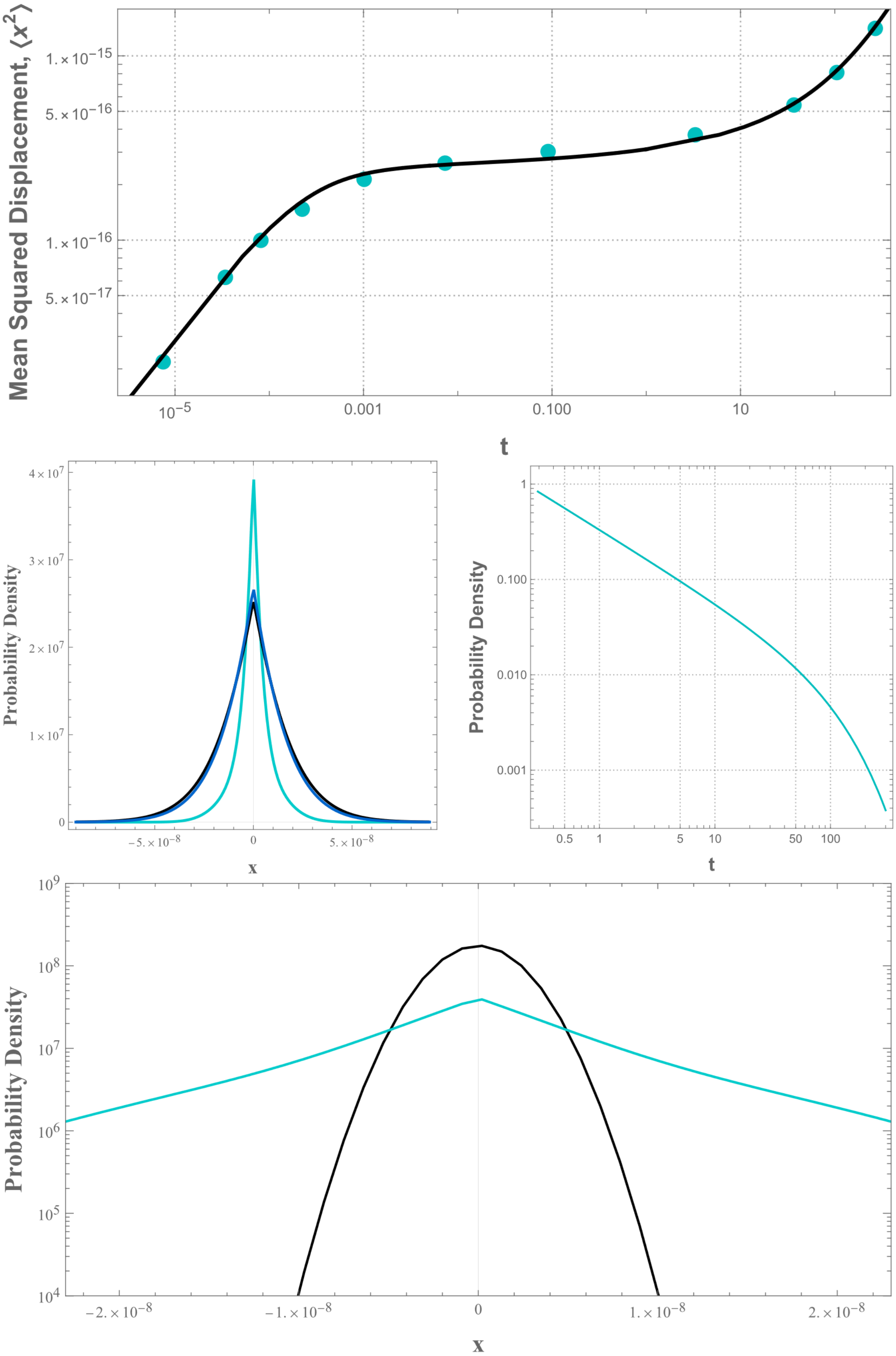

Figure 6.1: Top: Theoretical model (solid line) fitted to experimental values (blue points). Middle (left): PDF plots generated from the model described in Chapter 3 for $t = 0.8, 10$ and 13. Middle (right): The waiting time PDF corresponding to the fitted values. Bottom: The comparison of the PDF at $t = 0.8$ for $\gamma = 0.275$ and $\gamma = 1$, $\gamma = 1$ corresponds to a Gaussian profile which allows examination of where the greatest differences between a Gaussian PDF and the PDF of Chapter 3 would lie and thus helps guide further experimental enquiry.

Appendix A

Kramers-Moyal Expansion of the Master Equation

Before progressing with the derivation, we should briefly note that in casting the Master equation in the following form [38],

$$\frac{\partial P\big(f,t\big|f_0\big)}{\partial t} = \int W(f-\Delta f,\Delta f)P\big(f-\Delta f,t\big|f_0\big) - W(f,\Delta f)P\big(f,t\big|f_0\big)d\Delta f \tag{A.1}$$

we have employed the integral relationship below

$$\int_{-\infty}^{\infty} P(f')df' = -\int_{f+\infty}^{f-\infty} P(f-\Delta f)d\Delta f$$

$$= -\int_{\infty}^{-\infty} P(f-\Delta f)d\Delta f = \int_{-\infty}^{\infty} P(f-\Delta f)d\Delta f.$$

As supporting assumptions [38] for the Kramers-Moyal expansion we consider that the changes in state occur in small increments such that $W(f,\Delta f)$ is sharply peaked in jump size but varies slowly in initial state dependence. The final assumption is that the PDF $P(f,t\big|f_0)$ also varies slowly with the state f, and that the moments of the transition probability $W(f,\Delta f)$, $\mu_{W,n}(f)$ exist. These moments are found in the usual way via,

$$\mu_{W,n}(f) = \int_{-\infty}^{\infty} (\Delta f)^n W(f,\Delta f)d\Delta f. \tag{A.2}$$

The Kramers-Moyal expansion is based on the following Taylor series expansion

$$g(x+h) = \sum_{n=0}^{\infty} \frac{h^n}{n!}\frac{d^{(n)}}{dx^{(n)}}g(x). \tag{A.3}$$

We expand the first term of the Master equation as follows

$$\int W(f-\Delta f,\Delta f)P\big(f-\Delta f,t\big|f_0\big)d\Delta f=\sum_{n=0}^{\infty}\frac{(-\Delta f)^n}{n!}\frac{\partial^{(n)}}{\partial f^{(n)}}\Big[\int W(f,\Delta f)P\big(f,t\big|f_0\big)d\Delta f\Big]$$

$$=\sum_{n=0}^{\infty}\frac{(-1)^n}{n!}\frac{\partial^{(n)}}{\partial f^{(n)}}\Big[\int W(f,\Delta f)(\Delta f)^n d\Delta f\;\; P\big(f,t\big|f_0\big)\Big]=\sum_{n=0}^{\infty}\frac{(-1)^n}{n!}\frac{\partial^{(n)}}{\partial f^{(n)}}\Big[\mu_{W,n}(f)\;\;P\big(f,t\big|f_0\big)\Big]$$

$$=\Big[\int W(f,\Delta f)d\Delta f\;\;P\big(f,t\big|f_0\big)\Big]+\sum_{n=1}^{\infty}\frac{(-1)^n}{n!}\frac{\partial^{(n)}}{\partial f^{(n)}}\Big[\mu_{W,n}(f)\;\;P\big(f,t\big|f_0\big)\Big].$$

Substitution of this result back into the Master equation yields the Kramers-Moyal expansion of the master equation

$$\frac{\partial P\big(f,t\big|f_0\big)}{\partial t}=\int W(f,\Delta f)\;\;P\big(f,t\big|f_0\big)d\Delta f+\sum_{n=1}^{\infty}\frac{(-1)^n}{n!}\frac{\partial^{(n)}}{\partial f^{(n)}}\Big[\mu_{W,n}(f)\;\;P\big(f,t\big|f_0\big)\Big]$$

$$-\int W(f,\Delta f)P\big(f,t\big|f_0\big)d\Delta f$$

$$\therefore\;\frac{\partial P\big(f,t\big|f_0\big)}{\partial t}=\sum_{n=1}^{\infty}\frac{(-1)^n}{n!}\frac{\partial^{(n)}}{\partial f^{(n)}}\Big[\mu_{W,n}(f)\;\;P\big(f,t\big|f_0\big)\Big]\qquad\text{(A.4)}$$

Appendix B

Mittag-Leffler Function

B.0.1 Definitions

The Mittag-Leffler function is defined as [234]

$$E_\alpha(z) = \sum_{k=0}^{\infty} \frac{z^k}{\Gamma(1+\alpha k)}, \alpha \in \mathbb{C}, \Re(\alpha) > 0, z \in \mathbb{C} \tag{B.1}$$

with the generalised Mittag-Leffler function defined as

$$E_{\alpha,\beta}(z) = \sum_{k=0}^{\infty} \frac{z^k}{\Gamma(\beta+\alpha k)}, \alpha, \beta \in \mathbb{C}, \Re(\alpha) > 0, \Re(\beta) > 0, z \in \mathbb{C}. \tag{B.2}$$

The three parameter Mittag-Leffler function appears as

$$E^{\gamma}_{\alpha,\beta}(z) = \sum_{k=0}^{\infty} \frac{\Gamma(\gamma+k)}{\Gamma(\gamma)} \frac{z^k}{\Gamma(\beta+\alpha k)}, \alpha, \beta \in \mathbb{C}, \Re(\alpha) > 0, \Re(\beta) > 0, \Re(\gamma) > 0, \;\; z \in \mathbb{C}. \tag{B.3}$$

The Mittag-Leffler, E_α function interpolates between a stretched exponential at small values and and inverse power law behavior at large values. This function is general in nature and reduces to other, simpler functions at certain values of its parameters.

B.0.2 Some Special Cases:

$$E^{1}_{\alpha,1}(z) = E_{\alpha,1}(z) = E_\alpha(z) \tag{B.4}$$

$$E_0(z^2) = \frac{1}{1+z^2}, |z| < 1 \tag{B.5}$$

$$E_1(z^2) = e^{(-z^2)} \tag{B.6}$$

$$E^{\gamma}_{\alpha,\beta}(z^\alpha) = \frac{1}{\Gamma(\gamma)} H^{1,1}_{1,2}\left[z^\alpha \middle|^{(1-\gamma,1)}_{(0,1),(1-\beta,\alpha)} \right] \tag{B.7}$$

B.0.3 Laplace Transform:

$$\mathscr{L}\left[z^{\beta-1}E^{\gamma}_{\alpha,\beta}(\pm\eta z^{\alpha})\right](u) = \frac{u^{\alpha\gamma-\beta}}{\left(z^{\alpha} \mp \eta\right)^{\gamma}} \tag{B.8}$$

with $u, \alpha, \beta, \gamma \in C, \Re(\alpha) > 0, \Re(\beta) > 0, \Re(\gamma) > 0, \Re(u) > 0.$

Appendix C

Integral Transforms

C.1 Fourier Transform

In the one-dimensional case, the *Fourier transform* of a function $f(x)$ of a real variable $x \in \mathbb{R} = (-\infty, \infty)$ is defined as [235]

$$\mathscr{F}\left[f(x)\right](k) = \widehat{f}(k) = \int_{\infty}^{\infty} \exp(-ikx) f(x) dx, \quad k \in \mathbb{R}, \tag{C.1}$$

whereas the *inverse Fourier transform* is given by

$$\mathscr{F}^{-1}\left[\widehat{f}(k)\right](x) = f(x) = \frac{1}{2\pi} \int_{\infty}^{\infty} \exp(ikx) \widehat{f}(k) dk, \quad k \in \mathbb{R}. \tag{C.2}$$

The existence of the transform $\widehat{f}(k)$ is guaranteed if $f(x)$ is an integrable function and the integral converges. A sufficient (but not necessary) condition is to require that $f(x)$ be *absolutely integrable*, i.e., the integral

$$\int_{-\infty}^{\infty} |f(x)| dx \tag{C.3}$$

exists. In this case, $\widehat{f}(k)$ is absolutely convergent and, thus it is convergent.

C.2 Laplace Transform

The Laplace transform of a function $f(t)$ of a real variable $t \in \mathbb{R}^{+} = (0, \infty)$ is defined as [235]

$$\mathscr{L}\left[f(t)\right](u) = \widetilde{f}(u) = \int_{0}^{\infty} \exp(-ut) f(t) dt, \quad s \in \mathbb{C}. \tag{C.4}$$

For its existence, the function $f(t)$ must be such that

$$\exp(-\alpha t) |f(t)| \leq M, \quad \text{for} \quad \text{all} \quad t \in [0, \infty), \tag{C.5}$$

where M is a positive constant $\Re(\alpha) > 0$, indicating that the function $f(t)$ must not grow faster than a certain exponential function when $t \to \infty$. The *inverse Laplace transform* is given for $t \in \mathbb{R}^+$ by the formula

$$\mathscr{L}^{-1}\left[\widetilde{f}(u)\right](t) = f(t) = \frac{1}{2\pi i}\int_{\gamma-i\infty}^{\gamma+i\infty} \exp(ut)\widetilde{f}(u)du, \quad \gamma = \Re(u) > \sigma, \tag{C.6}$$

where σ is the infimum of u values for which the Laplace integral Eq. (C.4) converges, known as the abscissa of convergence.

C.3 Mellin Transform

The transform and inverse formula of Mellin are [**?**]

$$\widehat{\Phi}(p) = \int_0^\infty t^{p-1}\Phi(t)dt \tag{C.7}$$

$$\Phi(t) = \frac{1}{2\pi i}\int_{C-i\infty}^{C+i\infty} \widehat{\Phi}(p)t^{-p}dp \tag{C.8}$$

C.3.1 Mellin Transform of the Fox H function

$$\int_0^\infty x^{s-1}H_{p,q}^{m,n}\left[ax\middle|_{b_q,B_q}^{(a_p,A_p}\right]dx = a^{-s}\frac{\prod_{j=1}^{m}\Gamma\left(b_j+B_js\right)\prod_{j=1}^{n}\Gamma\left(1-a_j-A_js\right)}{\prod_{j=m+1}^{q}\Gamma\left(1-b_j-B_js\right)\prod_{j=n+1}^{p}\Gamma\left(a_j+A_js\right)} \tag{C.9}$$

where $a,s \in \mathbb{C}; -\min_{1\leq j\leq m}\Re(\frac{b_j}{B_j}) < \Re(s) < \max_{1\leq i\leq n}(\frac{1-\Re(a_j}{A_j}), |\arg a| < \frac{1}{2}\pi\alpha, \alpha > 0$.

Appendix D

Fox H-Function

The definition of the Fox H-function appears in terms of the Mellin-Barnes type integral as follows [187],

$$H_{p,q}^{m,n}(z) = H_{p,q}^{m,n}\left[z\left|\begin{matrix}(a_1,A_1),\ldots,(a_p,A_p)\\(b_1,B_1),\ldots,(b_p,B_p)\end{matrix}\right.\right]$$
$$= \frac{1}{2\pi i}\int_{\Omega}\theta(s)z^s\,ds, \tag{D.1}$$

where $\theta(s)$ is the ratio of products of gamma functions, hence the mention of Barnes in the integral name. Specifically we have

$$\theta(s) = \frac{\prod_{j=1}^{m}\Gamma(b_j - B_j s)\prod_{j=1}^{n}\Gamma(1-a_j+A_j s)}{\prod_{j=m+1}^{q}\Gamma(1-b_j+B_j s)\prod_{j=n+1}^{p}\Gamma(a_j - A_j s)}. \tag{D.2}$$

With the parameters defined such that, $0 \le n \le p, 1 \le m \le q, a_i, b_j \in \mathbb{C}, A_i, B_j \in \mathbb{R}^+, i = 1,\ldots,p, j = 1,\ldots,q$. The integration contour, Ω is chosen to run from $c - i\infty \to c + i\infty$ such that it avoids the poles of $\theta(s)$. There is a very useful expansion for the Fox H-function given in [236], it appears as

$$H_{p,q}^{m,n}(z) = H_{p,q}^{m,n}\left[z\left|\begin{matrix}(a_1,A_1),\ldots,(a_p,A_p)\\(b_1,B_1),\ldots,(b_p,B_p)\end{matrix}\right.\right]$$
$$= \sum_{h=1}^{m}\sum_{k=0}^{\infty}\frac{\prod_{j=1,j\neq h}^{m}\Gamma(b_j - B_j\frac{b_h+k}{B_h})\prod_{j=1}^{n}\Gamma(1-a_j+A_j\frac{b_h+k}{B_h})}{\prod_{j=m+1}^{q}\Gamma(1-b_j+B_j\frac{b_h+k}{B_h})\prod_{j=n+1}^{p}\Gamma(a_j - A_j\frac{b_h+k}{B_h})}\frac{(-1)^k z^{\frac{b_h+k}{B_h}}}{k!B_h}. \tag{D.3}$$

These functions are of great importance to anomalous diffusion as they provide a closed form in which to represent the non-Gaussian distributions that occur [237].

D.0.1 Expansion Formulae

Let m, n, p, and q be non-negative integers such that $1 \le m \le q, 0 \le n \le p$. Further, let $A_j, j = 1,\ldots,p$ and $B_j, j = 1,\ldots,q$ be positive numbers and $a_j, j = 1,\ldots,p$ and $b_j, j = 1,\ldots,q$ be complex numbers and $\mu > 0$ where

$$\mu = \sum_{j=1}^{p} B_j - \sum_{j=1}^{p} A_j. \tag{D.4}$$

Then if ω and η are complex numbers such that $\omega \neq 0$ and $\eta \neq 0$, then the following results hold:

Formula I

$$H_{p,q}^{m,n}\left[\eta\omega\middle|_{(b_1,B_1),\ldots,(b_q,B_q)}^{(a_1,A_1),\ldots,(a_p,A_p)}\right] = \eta^{\frac{b_1}{B_1}} \sum_{r=0}^{\infty} \frac{\left(1-\eta^{\frac{1}{B_q}}\right)^r}{r!} H_{p,q}^{m,n}\left[\omega\middle|_{(b_1+r,B_1),\ldots,(b_q,B_q)}^{(a_1,A_1),\ldots,(a_p,A_p)}\right] \tag{D.5}$$

where η is arbitrary for $m = 1$, and for $m > 1$ $|\eta^{\frac{1}{B_1}} - 1| < 1$, $\arg(\eta\omega) = B_1, \arg(\eta^{\frac{1}{B_1}}) + \arg(\omega)$, and $|\arg(\eta^{\frac{1}{B_1}})| < \frac{\pi}{2}$

Formula II

$$H_{p,q}^{m,n}\left[\eta\omega\middle|_{(b_1,B_1),\ldots,(b_q,B_q)}^{(a_1,A_1),\ldots,(a_p,A_p)}\right] = \eta^{\frac{b_q}{B_q}} \sum_{r=0}^{\infty} \frac{\left(\eta^{\frac{1}{B_q}}-1\right)^r}{r!} H_{p,q}^{m,n}\left[\omega\middle|_{(b_1,B_1),\ldots,(b_q+r,B_q)}^{(a_1,A_1),\ldots,(a_p,A_p)}\right] \tag{D.6}$$

where $q > m, |\eta^{\frac{1}{B_q}} - 1| < 1$ $\arg(\eta\omega) = B_q \arg(\eta^{\frac{1}{B_q}}) + \arg(\omega)$, and $|\arg(\eta^{\frac{1}{B_q}})| < \frac{\pi}{2}$

Formula III

$$H_{p,q}^{m,n}\left[\eta\omega\middle|_{(b_1,B_1),\ldots,(b_q,B_q)}^{(a_1,A_1),\ldots,(a_p,A_p)}\right] = \eta^{\frac{a_1-1}{A_1}} \sum_{r=0}^{\infty} \frac{\left(1-\eta^{-\frac{1}{A_1}}\right)^r}{r!} H_{p,q}^{m,n}\left[\omega\middle|_{(b_1,B_1),\ldots,(b_q,B_q)}^{(a_1-r,A_1),\ldots,(a_p,A_p)}\right] \tag{D.7}$$

where $n > 0, \Re\left(\eta^{\frac{1}{A_1}}\right) > \frac{1}{2}$, $\arg(\eta\omega) = A_1 \arg(\eta^{\frac{1}{A_1}}) + \arg(\omega)$, and $|\arg(\eta^{\frac{1}{A_1}})| < \frac{\pi}{2}$

Formula IV

$$H_{p,q}^{m,n}\left[\eta\omega\middle|_{(b_1,B_1),\ldots,(b_q,B_q)}^{(a_1,A_1),\ldots,(a_p,A_p)}\right] = \eta^{\frac{a_q-1}{A_q}} \sum_{r=0}^{\infty} \frac{\left(\eta^{-\frac{1}{A_p}}-1\right)^r}{r!} H_{p,q}^{m,n}\left[\omega\middle|_{(b_1,B_1),\ldots,(b_q,B_q)}^{(a_1,A_1),\ldots,(a_p-r,A_p)}\right] \tag{D.8}$$

where $p > n, \Re\left(\eta^{\frac{1}{A_p}}\right) > \frac{1}{2}$, $\arg(\eta\omega) = A_p \arg(\eta^{\frac{1}{A_p}}) + \arg(\omega)$, and $|\arg(\eta^{\frac{1}{A_q}})| < \frac{\pi}{2}$

D.0.2 Some Identities

Let m, n, p, and q be non-negative integers such that $1 \leq m \leq q, 0 \leq n \leq p$. Further, let $A_j, j = 1, \ldots, p$ and $B_j, j = 1, \ldots, q$ be positive numbers and $a_j, j = 1, \ldots, p$ and $b_j, j = 1, \ldots, q$ be complex numbers.Further, let r be a non-negative integer, δ and γ positive numbers, and ρ and τ complex numbers where ρ satisfies,

$$(\rho - 1 - \nu)/\delta \neq (a_j - 1 - \lambda)/A_j \quad (j = 1, \ldots, n; \nu, \lambda = 0, 1, \ldots). \tag{D.9}$$

It is also assumed that either, $\mu > 0$ and $z \neq 0$ in the Fox H function, where μ is defined in Eq. (D.4), or

$$0 \leq |z| < \prod_{j=1}^{p} A_j^{-A_j} \prod_{j=1}^{q} B_j^{B_j} = 1/\beta \quad \text{for} \quad \mu = 0. \tag{D.10}$$

Then, as detailed in [190],

$$H_{p+1,q+1}^{m,n+1}\left[z \middle| \begin{matrix} (\rho,\delta),(a_1,A_1),\ldots,(a_p,A_p) \\ (b_1,B_1),\ldots,(b_p,B_p),(\rho,\delta) \end{matrix}\right] = H_{p,q}^{m,n}\left[z \middle| \begin{matrix} (a_1,A_1),\ldots,(a_p,A_p) \\ (b_1,B_1),\ldots,(b_p,B_p) \end{matrix}\right], \tag{D.11}$$

$$H_{p+1,q+1}^{m+1,n}\left[z \middle| \begin{matrix} (a_1,A_1),\ldots,(a_p,A_p),(\rho,\delta) \\ (\rho,\delta),(b_1,B_1),\ldots,(b_p,B_p) \end{matrix}\right] = H_{p,q}^{m,n}\left| \begin{matrix} (a_1,A_1),\ldots,(a_p,A_p) \\ (b_1,B_1),\ldots,(b_p,B_p) \end{matrix}\right], \tag{D.12}$$

$$z^r H_{p,q}^{m,n}\left[z \middle| \begin{matrix} (a_1,A_1),\ldots,(a_p,A_p) \\ (b_1,B_1),\ldots,(b_p,B_p) \end{matrix}\right] = H_{p,q}^{m,n}\left[z \middle| \begin{matrix} (a_1+A_1\tau,A_1),\ldots,(a_p+A_p\tau,A_p) \\ (b_1+B_1\tau,B_1),\ldots,(b_p+B_p\tau,B_p) \end{matrix}\right], \tag{D.13}$$

$$(1/\gamma) H_{p,q}^{m,n}\left[z \middle| \begin{matrix} (a_1,A_1),\ldots,(a_p,A_p) \\ (b_1,B_1),\ldots,(b_p,B_p) \end{matrix}\right] = H_{p,q}^{m,n}\left[z^\gamma \middle| \begin{matrix} (a_1,A_1\gamma),\ldots,(a_p,A_p\gamma) \\ (b_1,B_1\gamma),\ldots,(b_p,B_p\gamma) \end{matrix}\right], \tag{D.14}$$

$$H_{p+1,q+1}^{m,n+1}\left[z \middle| \begin{matrix} (0,\delta),(a_1,A_1),\ldots,(a_p,A_p) \\ (b_1,B_1),\ldots,(b_p,B_p),(r,\delta) \end{matrix}\right] = (-1)^r H_{p+1,q+1}^{m+1,n}\left[z \middle| \begin{matrix} (a_1,A_1),\ldots,(a_p,A_p),(0,\delta) \\ (r,\delta),(b_1,B_1),\ldots,(b_p,B_p) \end{matrix}\right] \quad (p \leq q), \tag{D.15}$$

$$H_{p+1,q+1}^{m+1,n}\left[z \middle| \begin{matrix} (a_1,A_1),\ldots,(a_p,A_p),(1-r,\delta) \\ (1,\delta),(b_1,B_1),\ldots,(b_p,B_p) \end{matrix}\right] = (-1)^r H_{p+1,q+1}^{m,n+1}\left[z \middle| \begin{matrix} (1-r,\delta),(a_1,A_1),\ldots,(a_p,A_p) \\ (b_1,B_1),\ldots,(b_p,B_p),(1,\delta) \end{matrix}\right] \quad (p \leq q), \tag{D.16}$$

D.0.3 Successive Derivatives

Let m, n, p, and q be non-negative integers such that $1 \leq m \leq q, 0 \leq n \leq p$. Further, let $A_j, j = 1, \ldots, p$ and $B_j, j = 1, \ldots, q$ be positive numbers and $a_j, j = 1, \ldots, p$ and $b_j, j = 1, \ldots, q$ be complex numbers.Further, let r be a non-negative integer, δ be a positive number,

$$w^r \frac{d^r}{dw^r} H_{p,q}^{m,n}\left[w^\delta \middle| \begin{matrix} (a_1,A_1),\ldots,(a_p,A_p) \\ (b_1,B_1),\ldots,(b_p,B_p) \end{matrix}\right] = H_{p+1,q+1}^{m,n+1}\left[w^\delta \middle| \begin{matrix} (0,\delta),(a_1,A_1),\ldots,(a_p,A_p) \\ (b_1,B_1),\ldots,(b_p,B_p),(r,\delta) \end{matrix}\right]. \tag{D.17}$$

Now suppose that $w \neq 0, \infty$ For $\mu > 0$ and $0 < q/|w^\delta| < 1/\beta$ for $\mu = 0$. Then

$$w^r \frac{d^r}{dw^r} H_{p,q}^{m,n}\left[1/w^\delta \middle| \begin{matrix} (a_1,A_1),\ldots,(a_p,A_p) \\ (b_1,B_1),\ldots,(b_p,B_p) \end{matrix}\right] = (-1)^r H_{p+1,q+1}^{m,n+1}\left[w^\delta \middle| \begin{matrix} (1-r,\delta),(a_1,A_1),\ldots,(a_p,A_p) \\ (b_1,B_1),\ldots,(b_p,B_p),(1,\delta) \end{matrix}\right]. \tag{D.18}$$

D.0.4 Transformation properties

Laplace Transform

Let either $\alpha > 0, |\arg a| < \frac{1}{2}\pi\alpha$ or $\alpha = 0$ and $\Re(\delta) < -1$. Further assume that $\alpha > 0; \rho, \alpha, u \in C, \sigma > 0$, satisfy the condition: $\Re(\rho) + \sigma \min_{1\le j\le m}\left[\frac{\Re(b_j)}{B_j}\right] > 0$ for $\alpha > 0$ or $\alpha = 0, \mu \ge 0$; and $\Re(\rho) + \sigma \min_{1\le j\le m}\left[\frac{b_j}{B_j} + \frac{\Re(\delta)+\frac{1}{2}}{\mu}\right] > 0$ for $\alpha = 0$ and $\mu < 0$. Then for $\Re(u) > 0$, there holds the formula,

$$\mathscr{L}\left[t^{\rho-1}H^{m,n}_{p,q+1}\left[at^{\sigma}\Big|^{(a_p,A_p)}_{(b_q,B_q)}\right]\right](u) = u^{-\rho}H^{m,n}_{p,q}\left[au^{-\sigma}\Big|^{(a_p,A_p)}_{(b_q,B_q),(1-\rho,\sigma)}\right] \tag{D.19}$$

for $\Re(u) > 0, u \in C$.
With the inverse given by

$$\mathscr{L}^{-1}\left[u^{-\rho}H^{m,n}_{p,q}\left[au^{-\sigma}\Big|^{(a_p,A_p)}_{(b_q,B_q)}\right]\right](t) = t^{\rho-1}H^{m,n}_{p+1,q}\left[at^{\sigma}\Big|^{(a_p,A_p)}_{(b_q,B_q),(1-\rho,\sigma)}\right] \tag{D.20}$$

where $\rho, a, u \in C, \Re(u) > 0, \sigma > 0, \Re(\rho) + \sigma \max_{1\le i\le n}\left[\frac{1}{A_i} - \frac{\Re(a_i)}{A_i}\right] > 0, |\arg(a)| < \frac{1}{2}\pi\theta, \theta = -\sigma$.

Fourier Cosine Transform

$$\int_0^{\infty} x^{\rho-1}\cos(ax)H^{m,n}_{p,q+1}\left[bx^{\sigma}\Big|^{(a_p,A_p)}_{(b_q,B_q),(1-\rho,\sigma)}\right]dx = \frac{2^{\rho-1}\sqrt{\pi}}{a^{\rho}}H^{m,n+1}_{p+2,q}\left[ba^{-\sigma}2^{\sigma}\Big|^{(\frac{(2-\rho)}{2},\frac{\sigma}{2})(a_p,A_p)(\frac{(1-\rho)}{2},\frac{\sigma}{2})}_{(b_q,B_q),(1-\rho,\sigma)}\right] \tag{D.21}$$

where $a, \alpha, \sigma > 0, \rho, b \in C; |\arg b| < \frac{1}{2}\pi\alpha$;

$$\Re(\rho) + \sigma \min_{1\le j\le m}\Re\left(\frac{b_j}{B_j}\right) > 0; \Re(\rho) + \sigma \max_{1\le j\le n}\left[\frac{(a_j-1)}{A_j}\right] < 1$$

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