Aging Continuous Time Random Walks.

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We investigate aging continuous time random walks (ACTRW), introduced by Montroll and Bouc浇ad [J. Phys. A 29, 3847 (1996)]. Statistical behaviors of the displacement of the random walker \( \mathbf{r}(t) - \mathbf{r}(0) \) in the time interval \((0, t)\) are obtained, after aging the random walk in the time interval \((-t_a, 0)\). In ACTRW formalism, the Green function \( P(\mathbf{r}, t_a, t) \) depends on the age of the random walk \( t_a \) and the forward time \( t \). We derive a generalized Montroll–Weiss equation, which yields an exact expression for the Fourier double Laplace transform of the ACTRW Green function. Asymptotic long times \( t_a \) and \( t \) behaviors of the Green function are investigated in detail. In the limit of \( t \gg t_a \), we recover the standard non-equilibrium CTRW behaviors, while the important regimes \( t \ll t_a \) and \( t \sim t_a \) exhibit interesting aging effects. Convergence of the ACTRW results towards the CTRW behavior, becomes extremely slow when the diffusion exponent becomes small. In the context of biased ACTRW, we investigate an aging Einstein relation. We briefly discuss aging in Scher-Montroll type of transport in disordered materials.

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I. INTRODUCTION

Diffusion and relaxation in strongly disordered systems exhibits in many cases anomalous behaviors [1, 2, 3]. For example the diffusion of a test particle may become anomalous, namely the mean square displacement behaves like \( \langle r^2 \rangle \sim t^\alpha \) and \( \alpha \neq 1 \). A random walk framework, widely applied to describe anomalous diffusion is the continuous time random walk (CTRW) [4]. CTRWs are used to model many physical and chemical processes, for example: charge transport in disordered systems [5], protein folding dynamics [6, 7, 8], transport in low dimensional chaotic systems [9, 10, 11], and blinking behavior of single quantum dots [12, 13].

Anomalous diffusion processes may exhibit aging [10, 14, 15, 16], where vaguely speaking the age of the process controls the statistical properties of the random walk. Aging in diffusion processes yields an interesting perspective on dynamics in disordered medium, and more generally is used as a tool to probe complex systems such as spin glass, Anderson insulator, and colloidal suspensions [17] for a brief review. Monthus and Bouchaud [18] introduced a CTRW framework, which exhibits aging behaviors. We call this generalized CTRW, aging continuous time random walk (ACTRW). In this paper, we investigate properties of biased and non-biased ACTRWs in detail, in particular an exact expression for the Fourier double Laplace transform of the Green function is obtained, and asymptotic behaviors are investigated.

ACTRW might yield a phenomenological description of aging self diffusion dynamics in glasses [19], below the glass transition temperature \( T_g \) and then \( \alpha = T/T_g \) is temperature dependent (and see [20, 21, 22, 23] for related work). Recently [24] showed that ACTRW describes dynamics of an intermittent chaotic system. More generally, we expect that any random walk described by a CTRW process will exhibit aging, provided that aging initial condition discussed in the manuscript are satisfied. We note that other stochastic approaches to aging dynamics are based on a non-linear diffusion equation [25], and a generalized Langevin equation [26].

This paper is organized as follows. In Sec. II CTRW and ACTRW are introduced. We then derive an exact expression for the Fourier double Laplace transform of the ACTRW Green function \( P(r, t_a, t) \), thus generalizing the Montroll-Weiss equation (1) to include the effect of the age of the random walk (see Sec. III). Our result is based in part on recent work of Gordeche and Luck [27] who investigated renewal theory for processes with power law waiting time distributions. In Sec. IV we derive asymptotic behaviors of the Green function \( P(r, t_a, t) \) which are analyzed in detail. We then consider biased ACTRWs (Sec. V) and discuss an aging Einstein relation. Finally we briefly consider aging in Scher-Montroll type of transport as a possible application of this work. Note that a small part of our results was reported in [28], in the context of aging in chaotic transport.

II. CTRW AND ACTRW

One of the best well known random walk models is the CTRW introduced by Montroll and Weiss [30]. It de-
scribes a large class of random walks, both normal and anomalous and can be described as follows. Suppose a particle performs a random walk in such a way that the individual jump $r$ in space is governed by a probability density function (PDF) $f(r)$, and that all jump vectors are independent and identically distributed. The characteristic function of the position of the particle relative to the origin after $n$ jumps is $f^n(k)$, where $f(k)$ is the Fourier transform of $f(r)$. Unlike discrete time random walks, the CTRW describes a situation where the waiting time between jumps is not a constant. Rather, the waiting time is governed by the PDF $\psi(t)$ and all waiting times are mutually independent and identically distributed. Thus, number of jumps $n$ is a random variable.

Let $P_{MW}(r,t)$ be the Green function of the CTRW, the Montroll–Weiss equation yields this function in Fourier–Laplace ($k,u$) space:

$$P_{MW}(k,u) = \frac{1 - \psi(u)}{u} \frac{1}{1 - f(k) \psi(u)}.$$

All along this work we will use the convention that the arguments in the parenthesis define the space we are working in, thus $\psi(u)$ is the Laplace transform of $\psi(t)$. Properties of $P_{MW}(r,t)$ based on the Fourier–Laplace inversion of Eq. (1) are well investigated, see [3, 4] and Ref. therein. In particular, it is well known that asymptotic behavior of $P_{MW}(r,t)$ depends on the long time behavior of $\psi(t)$. Two classes of processes are usually considered. The first is the case when all moments of $\psi(t)$ are finite, the second class is the case where $\psi(t)$ is moment-less, corresponding to a situation where $\psi(t) \propto t^{-(1+\alpha)}$ and $0 < \alpha < 1$.

An important assumption made in the derivation of Eq. (1) is that the random walk begun at time $t = 0$. More precisely, it is assumed that the PDF of the first waiting time, i.e., the time elapsing between start of the process at $t = 0$ and the first jump event is $\psi(t)$. Thus the Montroll-Weiss CTRW approach describes a particular choice of initial conditions, called non-equilibrium initial conditions. The limitation of CTRW theory to a very particular choice of initial conditions, was an issue for debates in the early seventies [4].

Monthus and Bouchaud [22] introduced a CTRW for an ongoing process, where the random walk process is assumed to start at some time $t = -t_a$ long before start of observation at time $t = 0$. In Fig. 1 a stochastic realization of number of jumps in such a process is shown. For such a random walk the Green function is denoted with $P(r, t_a, t)$ and $r$ is the displacement in the time interval $(0,t)$. Using scaling analysis, [22] have investigated basic properties of this random walk, mainly the behavior of the Fourier transform of the Green function. For $\alpha < 1$ the random walk exhibits interesting aging effects, hence as mentioned we call it ACTRW.

![Graph showing number of jumps in a renewal process](image)

**Fig. 1:** Number of jumps $i$ in a renewal process with $\psi(t) = \pi^{-1} t^{-1/2}(1+t)^{-1}$, i.e. $\alpha = 1/2$. The process starts at $t = -t_a$ with $t_a = 4000$. Observation of the process begins at time $t = 0$. $t_1$ is the time elapsing between $t = 0$ and first jump event in the forward time interval $(0,t)$, in the Fig. $t_1 = 2553$.

### III. ACTRW:- GENERALIZED MONTROLL–WEISS EQUATION

The ACTRW describes the following process, a particle is trapped on the origin for time $t_1$, then jumps to $r_1$, the particle is then trapped on $r_1$ for time $t_2$, and then it jumps to a new location; the process is then renewed. Thus, the ACTRW process is characterized by a set of waiting times $\{t_1, \ldots, t_n, \ldots\}$ and displacements $\{r_1, \ldots, r_n, \ldots\}$. The time elapsing between start of observation at $t = 0$, and the first jump event is denoted by $t_1$. Here we denote the PDF of the first waiting time $t_1$ with $h_{t_a}(t_1)$. In ACTRW the random walk started at $t = -t_a$, before the start of observation at $t = 0$, therefore $h_{t_a}(t_1)$ depends on age of the process $t_a$. The waiting times $\{t_n\}$, with $n > 1$ are independent and identically distributed with a common probability density $\psi(t)$. The jump length $\{r_1, \ldots, r_n, \ldots\}$, are independent identically distributed random variables, described by the probability density $f(r)$.

In contrast, in the Montroll–Weiss non-equilibrium CTRW, the age of the process is zero $t_a = 0$. And, for that case $h_{t_a}(t_1) = \psi(t_1)$.

Recently, Gordeche and Luck [21] investigated statistical properties of fractal renewal processes, among other things they obtain $h_{t_a}(t_1)$. Let $h_s(u)$ be the double Laplace transform of $h_{t_a}(t_1)$

$$h_s(u) = \int_0^\infty dt_1 \int_0^{t_1} dt_2 h_{t_a}(t_1) e^{-t_2 u},$$

\[ (2) \]
then according to \[31\]
\[h_s(u) = \frac{1}{1 - \psi(s)} \frac{\psi(s) - \psi(u)}{u - s}. \tag{3}\]

In Appendix A we re-derive Eq. \[3\] using a method which slightly differs than the one used in \[31\].

Two types of behaviors are found for \(h_{t_a}(t_1)\) \[32\]. The first case corresponds to a situation when average waiting time \(t = \int_0^{\infty} \psi(t)dt\) is finite, and then in the long aging time limit one obtains
\[\lim_{t_a \to \infty} h_{t_a}(t_1) = \int_{t_1}^{t_a} \psi(t)dt. \tag{4}\]

This type of initial condition is called equilibrium initial condition, it was investigated previously in the context of CTRW and related models \[34\] \[35\] \[36\]. Here we will mainly consider the second case corresponding to a power law waiting time PDF
\[\psi(t) \propto t^{-(1+\alpha)} \quad \text{with} \quad 0 < \alpha < 1, \tag{5}\]
when \(t\) is long. In Laplace \(t \to u\) space Eq. \[3\] reads
\[\psi(u) \sim 1 - Au^\alpha, \tag{6}\]
where \(u\) is small, and \(A\) is a positive parameter \[37\]. For example the one sided Lévy PDFs whose Laplace pair is \(\psi(u) = \exp(-Au^\alpha)\), or \(\psi(u) = 1/(1 + Au^\alpha)\) discussed below, belong to the class of functions described by Eqs. \[3\] \[38\]. In the limit of long aging times, \(t_a \gg A^{1/\alpha}\), these kind of probability densities yield
\[h_{t_a}(t_1) \sim \frac{\sin(\pi \alpha)}{\pi} \frac{t_1^\alpha}{t_1^\alpha (t_1 + t_a)}. \tag{7}\]

Note that this expression is independent of the exact form of \(\psi(t)\), except for the exponent \(\alpha\). When \(\alpha \to 1\) the mass of the PDF \(h_{t_a}(t_1)\) is concentrated in the vicinity of \(t_1 \to 0\), as expected from a ‘normal process’. Eq. \[3\] shows that as age of the process becomes older, we have to wait longer for first jumping event to occur. In a physical process, this may correspond to a particle in a disordered system which searches for a local energy minima in time interval \((-t_a, 0)\). In this case the longer the search takes place the deeper the minima found, hence in statistical sense the release time becomes longer as the process is older. In what follows, we will also use the double Laplace transform of Eq. \[3\]:
\[h_s(u) \sim \frac{u^\alpha - s^\alpha}{s^\alpha(u - s)}. \tag{8}\]

This equation can be derived by inserting the small \(u\) and \(s\) expansion of \(\psi(u)\) and \(\psi(s)\) given in Eq. \[3\], in Eq. \[3\].

Let \(P(r, t_a, t)\) be the Green function of the random walker, where as mentioned
\[r \equiv r(t) - r(0) \tag{9}\]
is the displacement in the time interval \((0, t)\). Hence, clearly initially \(r = 0\) at time \(t = 0\). Let \((i)\) \(p_n(t_a, t)\) be the probability of making \(n\) steps in the time interval \((0, t)\) and \((ii)\) \(P(k, s, u)\) be the double–Laplace –Fourier transform \((r \to k, t_a \to s, t \to u)\) of \(P(r, t_a, t)\). Then
\[P(k, s, u) = \sum_{n=0}^{\infty} p_n(s, u) f^n(k), \tag{10}\]
where \(p_n(s, u)\) is the double Laplace transform of \(p_n(t_a, t)\). As mentioned, \(f^n(k)\) in Eq. \[10\] is the characteristic function of a random walk with exactly \(n\) steps. Using the convolution theorem of Laplace transform we obtain
\[p_n(s, u) = \left\{ \begin{array}{ll} \frac{1 - s h_s(u)}{su} & n = 0 \\ h_s(u)\psi^{n-1}(u) \frac{1 - \psi(u)}{u} & n \geq 1. \end{array} \right. \tag{11}\]
Hence inserting Eq. \[11\] in Eq. \[10\], using Eq. \[3\], and summing over \(n\), we find the exact result
\[P(k, s, u) = \frac{1}{su} + \frac{[\psi(u) - \psi(s)] [1 - f(k)]}{u(s - u) [1 - \psi(s)] [1 - \psi(u) f(k)]}. \tag{12}\]

Eq. \[12\] is a generalization of the Montroll–Weiss equation \[1\] for ACTRW. Note that \(P(k = 0, s, u) = 1/(su)\) as expected from the normalization condition.

It is useful to rewrite Eq. \[12\] in terms of the Montroll–Weiss Eq. \[1\], and \(p_0(s, u)\) in the first line of Eq. \[13\]:
\[P(k, s, u) = p_0(s, u) + h_s(u) f(k) P_{MW}(k, u), \tag{13}\]
The first term on the right hand side of this equation, describes random walks where the particle does not leave the origin (i.e. \(n = 0\)). The second term describes random walks where number of steps is greater than zero, it is given in terms of a convolution of \(h_s(u) f(k)\) with the Montroll-Weiss equation. This is expected since the only difference between ACTRW and the non-equilibrium CTRW, is the first waiting time distribution.

If the process is Poissonian, \(\psi(t) = \exp(-t)\), the Green function \(P(r, t, t_a)\) is independent of the age of the random walk \(t_a\). To show this we insert
\[\psi(u) = \frac{1}{1 + u}, \quad \psi(s) = \frac{1}{1 + s}. \tag{14}\]
in Eq. \[12\] and find
\[P(k, s, u) = \frac{1}{su + 1 - f(k)}. \tag{15}\]
Inverting to the double time domain
\[P(k, t_a, t) = e^{-[1 - f(k)]t}. \tag{16}\]
This result is independent of \(t_a\) as expected from a Markovian process. Assume that \(f(k) = 1 - m \mu |k|^\mu + \cdots\) for small values of \(k\) and \(\mu \leq 2\), implying that the
random walks is non biased. In the long time limit $P(\mathbf{k}, t_a, t) \sim \exp(-m_\mu k^{|t|})$, and either a Lévy behavior ($\mu < 2$) or a Gaussian behavior ($\mu = 2$) is found, as expected from the Gauss–Lévy central limit theorem \[3\]. In what follows, we investigate cases when this standard behavior does not hold.

IV. ASYMPTOTIC BEHAVIORS

Let us now consider basic properties of ACTRWs. While Eq. (12) is valid for a large class of random walks, including Lévy flights ($\mu < 2$), we will assume that variance of $f(x)$ is finite ($\mu = 2$). Special emphasis will be given to the case when $\psi(t)$ is moment-less $\alpha < 1$, since this regime exhibits interesting aging behaviors.

A. Mean Square Displacement

By differentiating Eq. (12) with respect to $k$ and setting $k = 0$, we obtain the moments of the random walk in a standard way. Assuming a non biased symmetrical random walk, we obtain

$$\langle r^2(s, u) \rangle = \frac{h_x(u)m_2}{u[1-\psi(u)]},$$

where $m_2 = \int r^2 f(|r|)dr$ is assumed to be finite. We consider power law waiting time PDFs as in Eqs (5,6), in the limit where both $u$ and $s$ are small, their ratio being arbitrary, we find

$$\langle r^2(s, u) \rangle \sim \frac{(u^\alpha - s^\alpha)m_2}{s^\alpha(u-s)Au^{1+\alpha}}.$$  \hspace{1cm} (18)

As shown below one can invert this equation exactly to the double time domain. However it is instructive to consider two limits first. If $u \ll s$, corresponding to $t \gg t_a$, we have

$$\langle r^2(s, u) \rangle \sim \frac{u^{-1-\alpha}m_2}{As}.$$ \hspace{1cm} (19)

While for $s \ll u$, corresponding to $t_a \gg t$, we have

$$\langle r^2(s, u) \rangle \sim \frac{m_2}{Au^2s^\alpha}.$$ \hspace{1cm} (20)

Inverting Eq. (18) and Eq. (21), we obtain

$$\langle r^2(t_a, t) \rangle \sim \begin{cases} \frac{m_2}{A(1+\alpha)} & t \gg t_a \\ \frac{m_2r^{-1}}{A(\alpha)} & t \ll t_a. \end{cases}$$ \hspace{1cm} (21)

This result is valid provided that both $t, t_a > A^{1/\alpha}$. In the limit $t \gg t_a$ we recover standard behavior found in non-equilibrium CTRW \[1]. In the aging regime, $t \ll t_a$ we find an interesting behavior. Independent of the exponent $\alpha$, the mean square displacement increases linearly with respect to the forward time $t$, as found in normal diffusion processes. In addition, the diffusion is slowed down as the age of the random walk $t_a$ is increased. This behavior is expected, due to statistically longer release times, from the initial position of the particle, as the age of the random walk is increased.

We now consider a specific choice of waiting time distribution:

$$\psi(u) = \frac{1}{1 + Au^\alpha},$$ \hspace{1cm} (22)

corresponding to \[4\]

$$\psi(t) = \frac{t^{-\alpha}}{A}E_{\alpha,\alpha} \left( -\frac{t^\alpha}{A} \right),$$ \hspace{1cm} (23)

where $E_{\alpha,\alpha}(x)$ is the generalized Mittag–Leffler function \[11\]. Inserting Eq. (22) in Eq. (17) we have

$$\langle r^2(s, u) \rangle = \frac{m_2}{A} \frac{u^\alpha - s^\alpha}{s^\alpha(u-s)u^{1+\alpha}}.$$ \hspace{1cm} (24)

Hence for this choice of waiting times, Eq. (18) is exact and not limited to the asymptotic regime. Inverting to the time domain using Eq. (7) we find

$$\langle r^2(t_a, t) \rangle = \frac{m_2}{A} \frac{\sin(\pi\alpha)}{\pi} \int_0^{t/t_a} \frac{(t/t_a - y)^\alpha}{y^\alpha(1+y)} dy.$$ \hspace{1cm} (25)

The solution of the integral is readily obtained, we find

$$\langle r^2(t_a, t) \rangle = \frac{m_2}{A} \frac{1}{\Gamma(1+\alpha)} [(t + t_a)^\alpha - t_a^\alpha].$$ \hspace{1cm} (27)

The right hand side of Eq. (27) describes the long time $t$, long time $t_a$, behavior of a large class of random walks with waiting time PDF satisfying $\psi(t) \propto t^{-(1+\alpha)}$ \[i.e., the right hand side of Eq. (23) is the double inverse Laplace transform of Eq. (18)]. Note that if $\alpha = 1$ in Eq. (27), the random walk does not exhibit aging.

B. Green Function

In this subsection we investigate asymptotic properties of the Green function $P(x, t_a, t)$, by considering the continuum approximation of Eq. (12). This approximation is expected to work in the limit where both the forward time $t$ and the aging time $t_a$ are long. A proof of the validity of this approach, is given in Sec. \[14\] for the one dimensional ACTRW. We assume a symmetric random walk, hence for small $|k|$, the following expansion is valid:

$$f(k) \sim 1 - \frac{1}{2} |k|^2 m_2 \frac{d}{A},$$ \hspace{1cm} (28)
where \( d \) is the dimensionality of the problem. We also use the small Laplace variable \( u \) expansion \( \psi(u) \sim 1 - Au^\alpha \) in Eq. (30). Inserting these expansions in Eq. (29), we obtain

\[
P(k, u, s) \sim \frac{s^\alpha u - u^\alpha}{s^{\alpha+1}u(u-s)} + \frac{(u^\alpha - s^\alpha)}{s^\alpha(u-s)Au^\alpha + |k|^2 |s|^2}. \tag{29}
\]

For convenience, and without loss of generality, we choose now to work in units where \( A = 1 \) and \( m_2/(2d) = 1 \).

Inverting Eq. (29) to the double time \( t, t' \) domain we find

\[
P(r, t, t') \sim p_0(t, t') \delta (r) +
\]

\[
\frac{\sin (\pi \alpha)}{\pi} \frac{1}{t_a \left( \frac{r}{t_a} \right)^\alpha \left( 1 + \frac{r}{t_a} \right)} \otimes P_{AMW}(r, t), \tag{30}
\]

where \( \otimes \) in Eq. (30) is the Laplace convolution operator with respect to the forward time \( t \), and in this limit

\[
p_0(t, t') \sim \frac{\sin (\pi \alpha)}{\pi} \int_{t/t_a}^\infty dx x^\alpha (1 + x). \tag{31}
\]

In Eq. (30), \( P_{AMW}(r, t) \) is the long time solution of the Montroll-Weiss equation, i.e., the Green function of the fractional diffusion equation [4],

\[
P_{AMW}(r, t) \equiv \mathcal{L}^{-1} \mathcal{F}^{-1}\left\{ \frac{u^{\alpha-1}}{u^{\alpha} + |k|^2} \right\} \tag{32}
\]

where \( \mathcal{L}^{-1} \mathcal{F}^{-1} \) is the inverse Laplace transform operator.

The Green function Eq. (30), is a sum of two terms. The first term on the right hand side of Eq. (30) is a singular term (i.e., the \( \delta (r) \) term). This term corresponds to random walks where number of steps in time interval \((0, t)\) is zero. Unless \( t \gg t_a \), this term cannot be neglected, since without it the Green function in Eq. (30) is not normalized. Thus ACTRW exhibits a behavior different from ordinary CTRWs, where realizations of random walks where number of steps is zero do not contribute to the asymptotic behavior.

In one dimension, we have

\[
P_{AMW}(r, t) = \frac{t}{\alpha |x|^{1+2/\alpha} t_{1/2}} \left( \frac{t}{|x|^{2/\alpha}} \right), \tag{33}
\]

where \( t_{1/2}(t) \) is the one sided Lévy stable PDF, whose Laplace pair is \( \exp(-u^{\alpha/2}) \). Hence the Green function solution of the ACTRW is

\[
P(x, t, t_a) \sim p_0(t, t_a) \delta (x) +
\]

\[
\frac{\sin (\pi \alpha)}{\pi} \frac{1}{t_a \left( \frac{t}{t_a} \right)^\alpha \left( 1 + \frac{t}{t_a} \right)} \otimes \frac{|x|^{-(1+2/\alpha)}}{\alpha 2^{1/\alpha} t_{1/2}} \left( \frac{|x|^{-(2/\alpha)}}{2^{1/\alpha}} \right), \tag{34}
\]

Scaling Eq. (34) with \( \tau \equiv t/t_a \) and \( q \equiv |x|/t^{\alpha/2} \) we obtain a scaled form of the Green function:

\[
P(x, t, t_a) \sim p_0(t, t_a) \delta (x) +
\]

\[
\frac{\sin (\pi \alpha)}{\pi} \frac{1}{\tau^{\alpha/2} \Gamma (1 + \alpha/2)} \left( \frac{\tau}{q^2} \right)^{\alpha/2} \tag{35}
\]

Below we analyze this expression in some detail.

In \( d \) space dimensions we have [12]

\[
P_{AMW}(r, t) = \alpha^{-1} r^{-d/2} \tau^{-d} \times
\]

\[
H_{12}^{\alpha} \left( 2^{-2/\alpha} r^{2/\alpha} t^{-1} \left| \frac{q (1, 1)}{d/2, 1/\alpha, (1, 1/\alpha)} \right) \right), \tag{36}
\]

Besides the \( d = 1 \) case, the Fox function solution \( H_{12}^{\alpha} \) is not generally tabulated, hence this solution is rather formal, though asymptotic behaviors of Eq. (36) are well investigated [43, 44]. A practical method of obtaining the solution of \( P_{AMW}(r, t) \) Eq. (30), using the inverse Lévy transform, is given in [43]. Using this method, we find the integral representation of the aging Green function in \( d \) dimension

\[
P(r, t, t_a) \sim p_0(t, t_a) \delta (r) +
\]

\[
\frac{1}{\alpha} \frac{\sin (\pi \alpha)}{\pi} \tau^{\alpha/2} \Gamma (1 + \alpha/2) \frac{t_{1/2}}{x^{1+2/\alpha}} \left( \frac{t}{|x|^{2/\alpha}} \right) \left( \frac{|x|^{-(2/\alpha)}}{2^{1/\alpha}} \right), \tag{37}
\]

where \( q = r/t^{\alpha/2} \) and \( \tau = t/t_a \). Similar to the one dimensional case, this solution shows the precise relation between ACTRWs and Lévy stable laws.

To conclude, Eq. (30) shows that the asymptotic solution of ACTRW is a sum of two terms. The first is a singular term, and the second is a convolution of the distribution of the first waiting time and the asymptotic Green function of the non-equilibrium CTRW.

C. Graphic Examples

In order to better understand the asymptotic behavior of the ACTRW, we perform numerical calculations to obtain the non-singular part of the Green function for different values of \( \alpha \) in one dimension. The one sided Lévy stable probability density in Eq. (33) was obtained using a numerical inverse Laplace transformation method [43, 44]. The calculated PDF was then used to evaluate
the convolution integral numerically to obtain the non-singular part of the Green function according to Eq. (35). For mathematical details on one sided Lévy stable laws, see Appendix in \[38\], and references therein.

In Figs. 3-5 we present the calculated non-singular part of the Green function for different \(\alpha\) in the aging behavior when \(\tau \ll 1\) to the non-equilibrium CTRW behavior when \(\tau \gg 1\) can be clearly seen in this figure, where the time ratio \(\tau = t/t_a\) is changed continuously. In addition, we observe a monotonic increase of the non-singular Green function as the scaled time \(\tau\) increases. Note that as the scaled \(\tau\) is increased the singular term is decreasing, hence we may think of this aging process, as if the singular part of the Green function, is feeding the non-singular part.

Figs. 3-5 show the scaled non-singular Green function versus \(q = |x|/t^\alpha/2\) for \(\alpha = 1/6, \alpha = 1/2, \) and \(\alpha = 5/6,\) respectively. In each figure, the scaled Green functions at several different \(\tau\)s are shown. A few general features can be seen in these figures. First, the Green function is clearly non-Gaussian for all cases, as we expected. A comparison between the shape of the Green function for \(\alpha = 1/6\) in Fig. 3 and for \(\alpha = 5/6\) in Fig. 5 clearly demonstrates that the deviations from Gaussian behavior are stronger for smaller \(\alpha\). In the limit of \(\alpha \to 1\) (not shown), we obtain a Gaussian Green function. Second, for \(\tau \gg t_a\) (i.e. \(\tau \gg 1\)), we recover the usual non-equilibrium CTRW behavior shown as the solid curves in Figs. 3-5. Finally, as \(\alpha\) becomes small, the convergence towards the non-aging behavior when \(t/t_a = \infty\) becomes extremely slow. For example, the case for \(\alpha = 1/6\) in Fig. 3 shows a significant deviation from the non-equilibrium CTRW behavior (solid line) when \(t/t_a = 10000\). As a result, the Green function exhibits aging behavior even when \(t/t_a\) is large, and an ACTRW treatment for dynamics in this regime will be essential.

D. Proof of Asymptotic Behavior for One Dimension

We now prove the validity of Eq. (30) using a method developed in \[38\]. The main idea is to show that moments of the ACTRW, are in the asymptotic limit described well by Eq. (30). For simplicity we assume a one dimensional symmetric random walk.

The moment generating function \(f(k)\) is expanded

\[
f(k) = 1 - m_2 k^2/2 + m_4 k^4/24 - m_6 k^6/720 + \cdots. \tag{38}\]

Where \(m_2, m_4\) etc are the moments of the jumps. Inserting this expansion in Eq. (14) we obtain the small \(k\) expansion of the ACTRW moment generating function:

\[
P(k, s, u) = \frac{1}{su} - \frac{h_s(u)}{u} \left\{ [1 + \Omega(u)] m_2 k^2/2 \right\}
\]

\[- [1 + \Omega(u)] (m_4 + 6\Omega(u)m_2^2) k^4/24
\]

\[
[1 + \Omega(u)] (m_6 - 30\Omega(u)m_2^2m_4 + 90\Omega^2(u)m_2^3) k^6/720 + \cdots \tag{39}\]

where

\[
\Omega(u) = \psi(u)/(1 - \psi(u)). \tag{40}\]

The moments \(\langle x^n(s, u) \rangle\) of the ACTRW are defined in the usual way

\[
P(k, s, u) = \sum_{n=0}^{\infty} \langle x^n(s, u) \rangle \frac{(ik)^n}{n!}. \tag{41}\]
Comparing Eq. (39) with Eq. (41) we have

\[
\langle x^2(s, u) \rangle = \frac{h_u(u)}{u} |1 + \Omega(u)|
\]

which means that normalization is conserved. For the second moment we obtain

\[
\langle x^2(s, u) \rangle = \frac{h_u(u)}{u} [1 + \Omega(u)]
\]

which is the same as Eq. (17). The fourth moment is more interesting:

\[
\langle x^4(s, u) \rangle = \frac{h_u(u)}{u} [1 + \Omega(u)] [m_4 + 6\Omega(u)m_2^2].
\]

Higher order moments are obtained in a similar way, for the sake of space they are not included here. Odd moments vanish due to the symmetry of the random walk.

One can easily see that the ACTRW n th moment \(\langle x^n(s, u) \rangle\) depends on the microscopic jump moments \(m_2, \ldots, m_n\). However in the limit \(u \to 0\)

\[
\langle x^n(s, u) \rangle \sim \frac{h_u(u)}{u \left(Au^\alpha\right)^{n/2} m_2^{n/2} n!}{2^{n/2}}.
\]

which depends on \(m_2\) but not on the higher order jump moments \(m_4, m_6\) etc. Thus the moments \(m_n\) with \(n > 2\) are the irrelevant parameters in this problem. Inserting Eq. (45) in Eq. (41) we have

\[
P(k, u, s) \sim \sum_{n=0}^{\infty} \frac{h_u(u)}{u} (ik)^n \left(\frac{m_2}{2Au^\alpha}\right)^{n/2}.
\]

Since we are interested in the limit where \(t\) and \(t_a\) are large, the ratio \(t/t_a\) being arbitrary, the corresponding Laplace variables \(u\) and \(s\) must approach zero their ratio being arbitrary. Therefore \(h_u(u)\) in Eq. (16) is given by its asymptotic form in Eq. (1). Inserting this expression for \(h_u(u)\) into Eq. (46), setting \(m_2 = 1\), and then summing over \(n\), we find an expression for \(P(k, u, s)\) that is the same as Eq. (29). Eq. (29) when transformed yields Eq. (30). To conclude we showed that Eq. (25) describes the small \(s, u\) behavior of the ACTRW moments, hence it follows that Eq. (30) describes the long time \(t\) and \(t_a\) behavior of the ACTRW Green function, the set of moments \(m_4, m_6\) etc are unimportant in this limit.

### E. Behavior on the Origin

Using Eq. (23) we investigate the nonsingular part of the ACTRW on the origin. For \(d = 1\), we find

\[
P(x, t_a, t)|_{x=0} = t^{-\alpha/2} g \left( \frac{t}{t_a} \right),
\]

where

\[
g(z) = z^{\alpha/2} \frac{\sin (\pi \alpha)}{2\pi \Gamma(1-\alpha/2)} \int_0^z \frac{dy}{(1+y)^\alpha}.
\]

Hence

\[
P(x, t_a, t)|_{x=0} \sim \begin{cases} \frac{t_{-\alpha/2}}{2\Gamma(\alpha)(1-3\alpha/2)} \left(\frac{t}{t_a}\right)^{1-\alpha} & t \ll t_a \\ \frac{t_{-\alpha/2}}{2\Gamma(1-\alpha/2)} t \gg t_a. \end{cases}
\]

In the limit \(t >> t_a\) we recover standard CTRW behavior (4).

In Fig. 3 we present the behavior of the ACTRW on the origin. The ratio of the scaled nonsingular ACTRW Green function to the Montroll-Weiss non-equilibrium CTRW Green function on the origin, \(P(x, t, t_a)t^{\alpha/2}|_{x=0}/2\Gamma(1-\alpha/2)\), is plotted versus the scaled dimensionless time \(t/t_a\). This ratio approaches one in the limit \(t >> t_a\), showing that the ACTRW process will converge to the standard non-equilibrium CTRW behavior when \(t >> t_a\). It can be clearly seen in the Fig. 3 that for \(\alpha > 1/2\), the ACTRW process has roughly converged to the non-equilibrium CTRW limit when \(t/t_a \approx 1\), while for \(\alpha \to 0\), the crossover to CTRW limit becomes extremely slow. For example, when \(\alpha = 1/2\), large deviations from the CTRW limit are clearly observed even when \(t/t_a = 10^8\). Since the limit \(\alpha \to 0\) is important for several systems (7, 55, 9).
it becomes clear that when $\alpha$ is small, the convergence towards the standard CTRW results becomes extremely slow, and the aging effect is of importance even when $t > t_a$.

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{fig6}
\caption{The behavior of the non-singular Green function on the origin normalized by the long time solution of the Montroll–Weiss non-equilibrium CTRW. The convergence of ACTRW towards the CTRW result is extremely slow when $\alpha \to 0$.}
\end{figure}

From Eq. (12), we see that for $t \ll t_a$ the non-singular part of $F(x, t_a, t)|_{x=0}$ increases with time $t$ when $\alpha < 2/3$. This unusual behavior is not unphysical, because the singular delta function term is a decreasing function of time, and the total probability of finding the random walker in a small vicinity of the origin is decreasing monotonically with time as expected.

\section{V. BIASED ACTRW}

We now consider one dimensional biased ACTRW. We therefore use the small $k$ expansion of $f(k)$:

\begin{equation}
 f(k) = 1 + ikm_1 - \frac{k^2}{2} m_2 \cdots,
\end{equation}

where $m_1 > 0$ is the averaged jump length. Differentiating Eq. (12) once with respect to $k$ and taking $k = 0$ we find the mean displacement of the random walker in $s, u$ space:

\begin{equation}
 \langle x(s, u) \rangle = \frac{m_1 h_s(u)}{u [1 - \psi(u)]},
\end{equation}

where $h_s(u)$ is defined in Eq. (8). Differentiating Eq. (12) twice with respect to $k$, we find the second moment of the biased random walk

\begin{equation}
 \langle x^2(s, u) \rangle = \frac{h_s(u)}{u [1 - \psi(u)]} \left[ 2m_1^2 \psi(u) \frac{1}{1 - \psi(u)} + m_2 \right].
\end{equation}

1. Aging Einstein Relation

We now derive a relation between the mean square displacement in the absence of bias, and the mean displacement of the particle in the presence of bias, reflecting the fluctuation–dissipation relation valid within linear response theory (see [50, 51, 52, 53] for related work). The case of zero aging $t_a = 0$ were discussed in [54], where some conceptual problems of linear response theory for systems exhibiting anomalous type of diffusion was discussed (e.g., the non-stationarity of the process, the dependence of $\alpha$ on external field).

We assume that the random walk is on a one dimensional lattice with lattice spacing $a$, therefore

\begin{equation}
 f(x) = P_L \delta(x - a) + P_R \delta(x + a).
\end{equation}

Here $P_L + P_R = 1$, hence the jump moments in Eq. (50) are $m_1 = (P_R - P_L)a$ and $m_2 = a^2$. We assume that the process obeys local detailed balance, namely $P_L / P_R = \exp(-aF/k_bT)$ where $T$ is the temperature. Using these conditions, and the assumption of weak field $aF/k_bT \ll 1$, we have $m_1 \simeq a^2 F/2k_bT$. Using Eqs. (17) and (51), we find

\begin{equation}
 \langle x(s, u) \rangle = \frac{F}{2k_bT} \langle x^2(s, u) \rangle_0.
\end{equation}

The subscript $F$ in Eq. (14) indicates the presence of external field $F$. $\langle x^2(s, u) \rangle_0$ is the mean square displacement in the absence of a field, i.e. Eq. (52) with $m_1 = 0$ and $m_2 = a^2$. Since the equation holds for the $s,u$ domain, it holds also for the $t_a,t$ domain

\begin{equation}
 \langle x(t_a, t) \rangle = \frac{F}{2k_bT} \langle x^2(t_a, t) \rangle_0.
\end{equation}

Thus the mean square displacement of the particle in the absence of the field (the fluctuation) yields the mean displacement in the presence of a weak field. When the waiting times are exponentially distributed, we obtain the usual Einstein relation between mobility and diffusion constant, which is independent of the age of the process $t_a$. For experimental verification of Eq. (14) in the non-aging regime $t_a = 0$ and with $\alpha < 1$, see [51, 52, 53].

2. Asymptotic Behavior of Biased ACTRW

From Eq. (11) we can derive the behavior of the mean displacement in exactly the same way as done in Sec. [54], and find

\begin{equation}
 \langle x(t_a, t) \rangle \sim \begin{cases} 
 \frac{m_1 t^\alpha}{A(1+\alpha)} & t \gg t_a \\
 \frac{m_1 t_a^{\alpha-1}}{A(\alpha)} & t \ll t_a.
\end{cases}
\end{equation}

For the second moment we use the small $s, u$ behavior of Eq. (52) and find

\begin{equation}
 \langle x^2(s, u) \rangle \sim \frac{1}{A u^{1+\alpha}} \frac{u^\alpha - s^\alpha}{s^\alpha (u-s)} \left[ 2m_1^2 / u^\alpha + m_2 \right].
\end{equation}
Inverting this equation using Eq. (56), we investigate now the dispersion:

$$
\sigma(t_a, t) \sim \sqrt{(x^2(t_a, t)) - \langle x(t_a, t) \rangle^2}.
$$

(58)

Considering first the \( u \ll s \) limit corresponding to \( t \gg t_a \) we recover Shlesinger’s result

$$
\sigma(t_a, t) \sim \frac{m_2 t^{2\alpha}}{A^2} \left[ \frac{2}{\Gamma(1+2\alpha)} - \frac{1}{\Gamma^2(1+\alpha)} \right] + \frac{m_2 t^\alpha}{\Delta \Gamma(1+\alpha)},
$$

(59)

hence if \( m_1 \neq 0 \) and \( \alpha < 1 \), one finds

$$
\sigma(t_a, t) \sim m_1 t^{\alpha}\sqrt{\frac{2}{\Gamma(1+2\alpha)} - \frac{1}{\Gamma^2(1+\alpha)}},
$$

(60)

For \( t \gg t_a \) the dispersion of the biased CTRW grows like the mean Eq. (56), a behavior very different than normal Gaussian diffusion.

Considering the \( s \ll u \) limit of Eq. (53) and using Eq. (54), we find for aging limit \( t \ll t_a \)

$$
\sigma(t_a, t) \sim \sqrt{\frac{m_2 t_a^{\alpha-1}}{A^2 \Gamma(\alpha)}} \frac{2 m_1}{A^2 \Gamma(\alpha) \Gamma(2 + \alpha)} - \frac{m_1 t_a^{2\alpha-2}}{A^2 T^2(\alpha)},
$$

(61)

Hence if \( m_1 \neq 0 \) and \( \alpha < 1 \), one finds

$$
\sigma(t_a, t) \sim \frac{m_1}{A} \sqrt{\frac{2}{\Gamma(\alpha) \Gamma(2 + \alpha)}} t_a^{1+\alpha},
$$

(62)

which is valid for \( t, t_a \gg A^{1/\alpha} \) and \( t \ll t_a \). As expected the dispersion decreases as age of the processes becomes older. Note that as \( \alpha \to 0 \), the first term in eq. (61) becomes important.

For \( \alpha = 1 \) one finds

$$
\sigma(t) \sim \sqrt{\frac{m_2 t}{A}},
$$

(63)

where in this case \( A \) has the meaning of the mean time between jumps. The dispersion in this case is independent of the age of the system \( t_a \), as expected from normal diffusion. We see that the dispersion in normal diffusion processes is controlled by the second moment of jump lengths \( m_2 \) (even when \( m_1 \neq 0 \)), while for CTRW and ACTRW \( m_1 \neq 0 \) is the relevant parameter.

VI. POSSIBLE APPLICATION: SCHER-MONTROLL TRANSPORT

In this section we briefly point out to one possible application of ACTRW. Scher and Montroll modeled transport in disordered medium based on CTRW theory. The fundamental reasons of why and when their approach is valid, while being the subject of much theoretical research \([53, 56]\), are not totally solved. What is clear is that on a phenomenological level, one can use the Scher–Montroll approach to fit behaviors of charge currents in a large number of experiments in very different systems. For example, transport of charge carriers in: organic photo-refractive glasses \([57]\), nano-crystalline \( T_2O_2 \) electrodes \([58]\), conjugated polymer system poly p-phenylene \([59, 60]\), and liquid crystalline zinc octakis \([61]\).

Scher and Montroll model such transport processes using \textit{non-equilibrium} biased CTRW theory with an effective waiting time distribution. In experiments, this corresponds to charge transport which is started at time \( t = 0 \), for example by a short photo flash applied on the system. After the initial triggering of the process, the charge carriers are transported using an external bias. For such initial conditions, we know that it is useful to assume that the physical transport process is described by the non-equilibrium biased CTRW.

In an aging experiment one would start the process, by an external impulse (e.g., a photo flash), then wait for an aging period \( t_a \), and only after that period add the external bias. In this case, biased aging CTRW might become a useful tool describing the aging transport. At this time it is still an open question if ACTRWs can be used to describe aging in real systems. Further it is not clear if aging in the above mentioned systems \([53, 58, 59, 60]\) is measurable, and if so do these very different systems exhibit any common aging effects in their transport?

In the non-aging experiments \([53, 58, 59, 60, 61]\) one measures the current of charge carriers, which according to the predictions of Scher and Montroll exhibit a universal behavior: \( I(t) \propto t^{1-\alpha} \) for short times and \( I(t) \propto t^{-\alpha-1} \) for long times. The transition time \( t_L \) between these two behaviors depends among other things on the length of the system. The short time behavior corresponds to \( I(t) \propto \frac{1}{t_a^\alpha}(x) \), with the non-equilibrium CTRW behavior \( \langle x \rangle \propto t^\alpha \), which yields immediately \( I(t) \propto t^{-1-\alpha} \). The long time behavior is more complicated, and is due to absorbing boundary condition.

In this paper we have calculated the mean displacement of the biased ACTRW, without including the influence of the boundary. Thus we provide the aging corrections to the short time behavior of Scher–Montroll transport. According to Eq. (50), the Scher–Montroll behavior \( I(t) \propto t^{1-\alpha} \) is replaced with \( I(t) \propto t_a^{1-\alpha} \) when \( t \ll t_a \ll t_L \). This behavior is independent of \( t \), similar to behavior of normal currents. In this aging regime, the current decreases as \( t_a \) is increased, while in the non-aging case the current is decreasing when the forward time \( t \) is increased. The assumption made is that the dispersion of the probability packet during the aging period, is small compared with the length of the system. A detailed investigation of aging in Scher–Montroll transport systems will be given elsewhere.
VII. SUMMARY

We have derived an exact expression for the Green function of ACTRW in Fourier–double Laplace space. This generalized Montroll – Weiss equation describes dynamics of a large class of random walks. Since the CTRW describes a large class of physical and chemical systems, mainly disordered systems, we expect that ACTRW will be a valuable tool when aging effects are investigated in these systems. Interesting aging behaviors are found when the system turns non-ergodic, namely when the mean waiting time diverges, \( \alpha < 1 \). We note that also when the mean waiting time is finite, aging behaviors may be observed, however only within a certain time window.

We showed that asymptotic behavior of the Green function is related to a few parameters of the underlying walk \( \alpha, A, \) and \( m_2 \), while other informations contained in \( \psi(t) \) and \( f(r) \) are irrelevant. The Green function behavior is non Gaussian when \( \alpha < 1 \), it is related to Lévy’s generalized central limit theorem and to Gordeche–Luc’s fractal renewal theory. Unlike standard random walks or non-equilibrium CTRWs, the asymptotic Green function is a sum of two terms: a singular term corresponding to random walks where number of jumps is zero and a non-singular term corresponding to random walks where number of jumps is one or more. Finally, we note that the fractional Fokker-Planck equation framework, developed based on CTRW concepts, can be modified based on the results obtained in this manuscript to include aging effects. This topic is left for a future publication.

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VIII. APPENDIX A

In this Appendix, we derive Eq. (4) using a method which is slightly different then the one used in [37]. Consider a non-equilibrium renewal process which starts at time \( t = 0 \) (in the ACTRW this initial time is \( -t_a \)). Let \( t_i, i = 1,...N \) be dots on the time axis on which jumping events occur. Let \( t_{N+1} \) denote the time on which the first jump event occurred which is larger than \( t_a \), namely \( t_N < t_a < t_{N+1} \). Note that \( N \) itself is not fixed, it is a random variable. The time intervals between jumps events are denoted by \( \tau_i \equiv t_{i+1} - t_i \).

The random variable we are interested in is \( t_1 \), where \( t_1 \equiv t_{N+1} - t_a \). Since \( t_a \) is a parameter in the problem, knowledge of statistical properties of \( t_{N+1} \) yields the distribution of \( t_1 \). Hence let \( P_{t_a}(t_{N+1}) \) denote the PDF of the random variable \( t_{N+1} \). It is given by:

\[
P_{t_a}(t_{N+1}) = \sum_{N=0}^{\infty} \left( \frac{\delta(t_{N+1} - \sum_{i=1}^{N+1} \tau_i)}{I(t_{N} < t_a < t_{N+1})} \right)_N.
\]

Here \( I(t_{N} < t_a < t_{N+1}) = 1 \) if \( t_N < t_a < t_{N+1} \), otherwise it is zero. The average in Eq. (64) is

\[
\langle \cdots \rangle_N = \left( \Pi_{i=1}^{N+1} \int_0^\infty \psi(\tau_i)d\tau_i \cdots \right).
\]

We consider the double Laplace transform \( t_a \to s \) and \( t_{N+1} \to u \) of \( P_{t_a}(t_{N+1}) \) Eq. (44). \( P_s(u) \). Using \( t_N = \sum_{i=1}^{N} \tau_i \), we have:

\[
\int_0^\infty e^{-t_au}I(t_{N} < t_a < t_{N+1})dt_a = \frac{e^{-tu}s - e^{-t_{N+1}s}}{s} = \frac{e^{-s\sum_{i=1}^{N} \tau_i} - e^{-s\sum_{i=1}^{N+1} \tau_i}}{s},
\]

and

\[
\int_0^\infty e^{-u\tilde{t}_{N+1}}\delta(\tilde{t}_{N+1} - \sum_{i=1}^{N+1} \tau_i) \, d\tilde{t}_{N+1} = e^{-u\sum_{i=1}^{N+1} \tau_i}.
\]

Using Eqs. (66,67), we have from Eq. (44)

\[
P_s(u) = \sum_{N=0}^{\infty} P_s(u) = \sum_{N=0}^{\infty} \langle \exp \left( -u \sum_{i=1}^{N+1} \tau_i \right) \frac{\exp \left( -s \sum_{i=1}^{N} \tau_i \right) - \exp \left( -s \sum_{i=1}^{N+1} \tau_i \right)}{s} \rangle.
\]

Using the fact that the random variables \( \tau_i \) are independent and identically distributed, we have

\[
P_s(u) = \frac{1}{s} \sum_{N=0}^{\infty} \left[ \psi^N(u + s)\psi(u) - \psi^{N+1}(u + s) \right],
\]

where \( \psi(u + s) = \int_0^\infty \exp[-(u + s)\tau] \psi(\tau) \, d\tau \). Summing Eq. (68) we find

\[
P_s(u) = \frac{\psi(u) - \psi(u + s)}{s} \frac{1}{1 - \psi(u + s)}.
\]

Now the PDF \( h_{t_a}(t_1) \) is obtained from \( P_{t_a}(t_{N+1}) \) using \( t_1 = t_{N+1} - t_a \). According to definition of Laplace transform, we can write

\[
h_{t_a}(t_1) = \frac{\psi^{N+1}(t_1) - \psi^{N}(t_1)}{t_a}.
\]
\[ \mathcal{L}^{-1}_{u \rightarrow t_1, s \rightarrow t_a} \left\{ \int_0^\infty dt_a e^{-s t_a} \int_0^\infty dt_1 e^{-u t_1} h_{t_a}(t_1) \right\}, \quad (71) \]

where \( \mathcal{L}^{-1} \) is the double inverse Laplace transform. We use \( t_1 = t_{N+1} - t_a \) and find:

\[ h_{t_a}(t_3) = \mathcal{L}^{-1}_{u \rightarrow t_1, s \rightarrow t_a} \left\{ \int_0^\infty dt_a e^{-s t_a} \int_0^\infty dt_1 e^{-u(t_{N+1} - t_a)} P_{t_a}(t_{N+1}) \right\}. \quad (72) \]

Inserting Eq. (71) in Eq. (73), we find Eq. (3).

Hence it is easy to see that

\[ h_s(u) = P_{s-u}(u). \quad (73) \]
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