Eur. Phys. J. B (2017) 90: 205
DOI: 10.1140/epjb/e2017-80270-9

THE EUROPEAN
PHYSICAL JOURNAL B

Regular Article

Ageing effects in ultraslow continuous time random walks*

Aleksei V. Chechkin[1,2,a], Holger Kantz[3], and Ralf Metzler[1]

[1] Institute of Physics and Astronomy, University of Potsdam, 14476 Potsdam, Germany
[2] Akhiezer Institute for Theoretical Physics, Kharkov Institute of Physics and Technology, Kharkov 61108, Ukraine
[3] Max Planck Institute for the Physics of Complex Systems, 01187 Dresden, Germany

Received 9 May 2017 / Received in final form 28 June 2017
Published online 1 November 2017 –

Abstract. In ageing systems physical observables explicitly depend on the time span elapsing between the original initiation of the system and the actual start of the recording of the particle motion. We here study the signatures of ageing in the framework of ultraslow continuous time random walk processes with super-heavy tailed waiting time densities. We derive the density for the forward or recurrent waiting time of the motion as function of the ageing time, generalise the Montroll–Weiss equation for this process, and analyse the ageing behaviour of the ensemble and time averaged mean squared displacements.

1 Introduction

Deviations from the normal-diffusive law $\langle x^2(t)\rangle \simeq t$ of Brownian motion are routinely measured in a wide range of systems [1–8]. In particular, subdiffusion of the form $\langle x^2(t)\rangle \simeq t^\alpha$ with $0 < \alpha < 1$ is observed in the crowded and structured environment of biological cells [3–5,9–13] or their membranes [14–17]. Similarly, subdiffusion is observed in artificially crowded systems [18–20] and for in silico lipid membranes [21–25]. Also superdiffusion with $1 < \alpha < 2$ is often found in live cells due to active processes [26–28].

When the disorder of the system is increased beyond the above examples, instead of the power law form $\langle x^2(t)\rangle \simeq t^\alpha$ for the observed process we may also encounter logarithmically slow (ultraslow) anomalous diffusion with the mean squared displacement $\langle x^2(t)\rangle \simeq \ln^\gamma t$ with $\gamma > 0$. The prime example for ultraslow diffusion is Sinai diffusion, in which a random walk occurs in the potential landscape created by a seed random walk [29–34]. Logarithmic diffusion was found in non-linear maps [35] as well as for the random motion of particles in the homogeneous cooling state of granular gases with constant restitution coefficient [36,37]. In disordered channels, while a single particle would perform power-law anomalous diffusion of the above form $\langle x^2(t)\rangle \simeq t^\alpha$, due to excluded volume interactions in a single file of particles the mutual blocking gives rise to logarithmically slow diffusion [38]. Theoretical frameworks for ultraslow diffusion include ageing continuous time random walks [39], strongly localised diffusivity in heterogeneous diffusion [40,41], ultraslow scaled Brownian motion [42,43], and distributed order fractional diffusion equations [44,45].

Here we consider a model for ultraslow diffusion, based on continuous time random walks with super-heavy tailed waiting time densities [35,46,47]. These were introduced as a mean field, annealed description of Sinai diffusion, characterised by the quenched disordered environment. Random walks with super-heavy tailed waiting time densities were shown to share several important features with Sinai diffusion [47]. In particular, we here ask the experimentally relevant question how ageing changes the statistic of ultraslow motion. By ageing we understand the explicit dependence of physical observables on the delay t_a (the ageing time) between the original initiation of the system and the actual start of the observation. The occurrence of ageing signifies the non-stationary character of the measured system. This contrasts stationary dynamics such as regular Brownian motion or fractional Brownian motion, whose observables are independent of t_a [6,48]. Probing for ageing effects is thus an important diagnosis for the nature of observed diffusion processes [9,10,16,17]. A direct consequence of ageing in some systems is the population splitting of an ensemble of diffusing particles into those which are completely immobile during the measurement and others exhibiting more or less vivid motion [40,41,49,50].

We here develop the framework for the study of ageing effects in ultraslow continuous time random walks. In particular we obtain analytical expressions for the ensemble and time averaged mean squared displacements, the density of the recurrence or forward waiting times, as well as the Green's function (propagator) of the process. We proceed as follows. In Section 2 we first recall some basic definitions of the continuous time random walk framework. Section 3 then introduces the concept of super-heavy tailed waiting time densities and the associated

* Contribution to the Topical Issue "Continuous Time Random Walk Still Trendy: Fifty-year History, Current State and Outlook", edited by Ryszard Kutner and Jaume Masoliver.

[a] e-mail: achechkin@kipt.kharkov.ua

basic properties of the emerging ultraslow continuous time random walk. In Section 4 the forward or recurrent waiting time density is derived and evaluated in the limits of weak and strong ageing. The ageing analogue of the fundamental Montroll–Weiss equation is derived and analysed in Section 5. Sections 6 and 7 provide results for the ensemble and time averaged mean squared displacements, respectively. Finally, we draw our conclusions in Section 8.

2 Continuous time random walks

For a continuous time random walk process with given probability densities for the jump lengths and waiting times, $\lambda(x)$ and $\psi(t)$, respectively, the Greens' function $P_{MW}(x,t)$ for the initial condition $P_{MW}(x,0) = \delta(x)$ is given in terms of the Montroll–Weiss equation [1,2,51–53]

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

Here, we denote the Laplace transform

$$\psi(u) = \mathscr{L}\left\{\psi(t)\right\} = \int_0^\infty \psi(t) e^{-ut} dt, \tag{2}$$

and the Fourier transform with its inverse

$$\lambda(k) = \int_{-\infty}^{\infty} \lambda(x) e^{ikx} dx,$$
$$\lambda(x) = \frac{1}{2\pi}\int_{-\infty}^{\infty} \lambda(k) e^{-ikx} dk, \tag{3}$$

by explicit dependence on the Laplace and Fourier variables, u and k, respectively. We assume that the probability density for the jump lengths has a finite variance

$$\sigma^2 = \int_{-\infty}^{\infty} x^2 \lambda(x) dx, \tag{4}$$

such that its Fourier transform has the small wave number expansion [1,2,53]

$$\lambda(k) \sim 1 - \frac{\sigma^2}{2}k^2. \tag{5}$$

In the following we only consider this finite variance case for the density of jump lengths. The mean squared displacement of the process follows directly from equation (1) through inverse Laplace transform of

$$\langle x^2(u)\rangle = -\left.\frac{\partial^2}{\partial k^2} P_{MW}(k,u)\right|_{k=0}. \tag{6}$$

For a Poissonian waiting time density $\psi(t) = \tau^{-1}\exp(-t/\tau)$ the Laplace transform has the short Laplace frequency expansion

$$\psi(u) \sim 1 - u\tau, \tag{7}$$

so that in combination with relation (5) the Montroll–Weiss equation can directly be rewritten as

$$P_{MW}(k,u) \times [u + K_1 k^2] = 1. \tag{8}$$

With the Fourier–Laplace transform $1/u$ of the $\delta(x)$ initial condition, the inverse Fourier and Laplace transform of relation (8) is but the normal diffusion equation

$$\frac{\partial P_{MW}(x,t)}{\partial t} = K_1 \frac{\partial^2}{\partial x^2} P_{MW}(x,t), \tag{9}$$

with the diffusion coefficient $K_1 = \sigma^2/(2\tau)$ [1,2,53]. The associated mean squared displacement is $\langle x^2(t)\rangle = 2K_1 t$. A typical case considered in this framework is that of a one-sided Lévy stable form

$$\psi(u) = \exp\left(-[u\tau]^\alpha\right), \tag{10}$$

with $0 < \alpha < 1$ in Laplace space, corresponding to the power-law density

$$\psi(t) \simeq \frac{\tau^\alpha}{t^{1+\alpha}}. \tag{11}$$

In combination with the Montroll–Weiss equation (1) this gives rise to a fractional diffusion equation in (x,t)-space and the subdiffusive mean squared displacement [1,2,54–56]

$$\langle x^2(t)\rangle = 2K_\alpha t^\alpha, \tag{12}$$

where the generalised diffusion coefficient is given by $K_\alpha = \sigma^2/[2\tau^\alpha]$ [1,2,53–56]. In this formulation, the limit $\alpha = 1$ reduces exactly to Brownian motion, as then $\psi(u)$ transforms back to a Dirac delta form of $\psi(t)$, corresponding to the renormalisation argument of Hughes [57].

3 Super-heavy tailed waiting time densities

In the following we consider a waiting time density with extremely slow asymptotic behaviour of the form [47,58,59]

$$\psi(t) \sim \frac{\ell(t)}{t}, \tag{13}$$

valid in the long time limit. Here, $\ell(t)$ denotes a slowly varying function with the property $\ell(qt) \sim \ell(t)$ for $q > 0$ in the limit $t \to \infty$. We remind the reader of two important properties of the density $\psi(t)$ in equation (13): (i) Since for the slowly varying function $\ell(t)$ the condition $t^\rho \ell(t) \to \infty$ for all $\rho > 0$ at $t \to \infty$, all fractional moments of $\psi(t)$ are infinite. (ii) The slowly varying function $\ell(t)$ in equation (13) is not arbitrary in the sense that it must be compatible with the normalisation condition for $\psi(t)$, implying that $\ell(t)$ should decay faster then $1/\ln(t)$ at $t \to \infty$.

The special choice $\ell(t) = A/\ln^{1+\gamma}(t/\tau)$ for ℓ with $\gamma > 0$ leads to the Havlin–Weiss waiting time density [46]

$$\psi(t) \sim \frac{A}{t\,[\ln(t/\tau)]^{1+\gamma}}. \tag{14}$$

The (cumulative) stalling probability that no jump occurs up to some time t is given by [1,2,53]

$$\Psi(t) = 1 - \int_0^t \psi(t')dt' = \int_t^\infty \psi(t')dt', \tag{15}$$

corresponding to the relation

$$\Psi(u) = \frac{1-\psi(u)}{u}, \tag{16}$$

in Laplace space. For the special form (13) the stalling probability is also a slowly varying function, that is, $\Psi(\mu t) \sim \Psi(t)$ in the limit $t \to \infty$ [60]. With this property of slow variation and with relation (16) it follows that

$$\begin{aligned}\psi(u) &= 1 - u\Psi(u) = 1 - u\int_0^\infty \Psi(t)e^{-ut}dt\\ &= 1 - \int_0^\infty \Psi\left(\frac{q}{u}\right)e^{-q}dq,\end{aligned} \tag{17}$$

where we substituted $q = ut$. In the limit $u \to 0$ relevant for the long time asymptotic entering the Montroll–Weiss equation in the diffusion limit, we can replace $\Psi(q/u)$ in the above integral with $\Psi(1/u)$, which is independent of the integration variable. As the remaining integral equals unity, we thus get the asymptotic equality

$$\psi(u) \sim 1 - \Psi(t)\Big|_{t=1/u}. \tag{18}$$

Given that $\Psi(t)$ is slowly varying, we can express all our results without taking any specific form. To illustrate our results we use the generic form

$$\Psi(t) = \frac{\ln^\gamma \eta}{\ln^\gamma(\eta + t)}, \tag{19}$$

where the finite constant η guarantees convergence at short times. From this choice for the stalling probability we obtain the corresponding waiting time density by differentiation,

$$\psi(t) = \frac{\gamma \ln^\gamma \eta}{(\eta + t)\ln^{1+\gamma}(\eta + t)}. \tag{20}$$

Asymptotically, this is equivalent to the Havlin–Weiss form (14). Therefore, the asymptotic form entering equation (18) is

$$\Psi(t)\Big|_{t=1/u} \sim \frac{\ln^\gamma \eta}{\ln^\gamma(1/u)}, \tag{21}$$

in the limit $u \to 0$. Concurrently, the asymptotic behaviour of $\psi(u)$ in the small u limit for the specific choice of the regularised Havlin–Weiss form (20) is [46]

$$\psi(u) \sim 1 - \frac{\ln^\gamma \eta}{\ln^\gamma(1/u)}. \tag{22}$$

To obtain the mean squared displacement we use relation (6), yielding

$$\langle x^2(u)\rangle = \frac{\psi(u)}{u[1-\psi(u)]} \times \left(-\frac{\partial^2\lambda(k)}{\partial k^2}\right)_{k=0}. \tag{23}$$

Now, we use the asymptotic equality (18) to get

$$\langle x^2(u)\rangle \sim \frac{\sigma^2}{u\Psi(t)|_{t=1/u}} \times \Big(1 - \Psi(t)|_{t=1/u}\Big). \tag{24}$$

By Tauberian theorems [57,61], we then find the mean squared displacement

$$\langle x^2(t)\rangle \sim \sigma^2\left(\frac{1}{\Psi(t)} - 1\right), \tag{25}$$

valid in the long time limit. For the regularised Havlin–Weiss waiting time density (20) the concrete form becomes

$$\langle x^2(t)\rangle \sim \frac{\sigma^2}{\ln^\gamma \eta}\ln^\gamma(\eta + t). \tag{26}$$

To obtain the Green's function for the ultraslow continuous time random walk we rewrite the Montroll–Weiss equation (1) in the identical form

$$\begin{aligned}P_{MW}(k,u) &= \frac{1-\psi(u)}{u} + \frac{[1-\psi(u)]\psi(u)}{u}\\ &\quad\times\frac{\lambda(k)}{1-\lambda(k)\psi(u)}\\ &\sim \frac{\Psi(t)_{t=1/u}}{u} + \frac{\Psi(t)_{t=1/u}}{u}\\ &\quad\times\frac{1-\Psi(t)_{t=1/u}}{\sigma^2k^2/2 + \Psi(t)_{t=1/u}},\end{aligned} \tag{27}$$

where in the last step we took the limit of small k and u, taking into account relations (5) and (18). Inverse Fourier transform yields

$$\begin{aligned}P_{MW}(x,u) &\sim \frac{\Psi(t)_{t=1/u}}{u}\delta(x) + \frac{1-\Psi(t)_{t=1/u}}{u}\sqrt{\frac{\Psi(t)_{t=1/u}}{2\sigma^2}}\\ &\quad\times\exp\left(-|x|\sqrt{2\Psi(t)_{t=1/u}/\sigma^2}\right).\end{aligned} \tag{28}$$

This result is normalised, that is, $\int_{-\infty}^{\infty} P_{MW}(x,u)dx = 1/u$. We note that the spatial integral over the first part is of order $\Psi(t)_{t=1/u}/u$ while that of the second part is $1/u$. As in the limit $u \to 0$ we have that $\Psi(t)_{t=1/u} \to 0$ the first part can be neglected at $x = 0$.

4 Forward or recurrent waiting time

After defining the tools we need in the following and acquainting the reader with the otherwise quite unfamiliar ultraslow continuous time random walk process, we are now ready to derive the ageing properties of the process. This analysis is based on the probability density of the first or recurrent waiting time [49,50,62–65].

In an ageing continuous time random walk process we are interested in the physical situation that the experimental observation of the system starts only at an ageing time t_a after the original initiation of the system at $t = 0$. For any continuous time random walk process with a diverging characteristic waiting time scale

$$\langle t\rangle = \int_0^\infty t\psi(t')dt', \tag{29}$$

the waiting times typically become longer and longer. When the system is allowed to age for an appreciable time t_a, the likelihood that the start of the observation occurs during a long waiting period is significant. In such a scenario the probability to observe the first jump in the system is not given by $\psi(t)$ but by the modified statistic given by the probability density $h(t_1, t_a)$, referred to as forward or recurrent waiting time density [49,50,62–65]. Here t_1 is the time for the first jump to occur after start of the observation at $t_a > 0$. The dual Laplace transform of this forward or recurrent waiting time density

$$h(u, s) = \int_0^\infty \int_0^\infty h(t_1, t_a)e^{-ut_1-st_a}dt_1dt_a, \tag{30}$$

according to Cox [66], see also Godrèche and Luck [62], is given by the algebraic relation

$$h(u, s) = \frac{1}{1-\psi(s)} \times \frac{\psi(s)-\psi(u)}{u-s}. \tag{31}$$

It is straightforward to show that $h(t_1, t_a)$ is normalised with respect to t_1 and its Laplace inversion converges to the regular waiting time density $\psi(t_1)$ in the limit $s \to \infty$ corresponding to short ageing times. The properties of $h(t_1, t_a)$ for power-law waiting times are discussed in references [49,50,63–65].

The inverse Laplace transform of equation (31) is given by

$$h(t_1, s) = \frac{1}{1-\psi(s)} \times \left(e^{st_1}\psi(s) - \mathscr{L}^{-1}\left\{\frac{\psi(u-s+s)}{u-s}\right\}\right), \tag{32}$$

where in the last term in the curly brackets we expanded the argument of $\psi(u)$ by zero. This last term by help of standard theorems of the Laplace transformation becomes

$$\mathscr{L}^{-1}\left\{\frac{\psi(u-s+s)}{u-s}\right\} = e^{st_1}\int_0^{t_1} \mathscr{L}^{-1}\{\psi(u+s)\}dt_1' = e^{st_1}\int_0^{t_1} e^{-st_1'}\psi(t_1')dt_1'. \tag{33}$$

Combining result (33) with equation (32) we obtain the compact relation

$$h(t_1, s) = \frac{e^{st_1}}{1-\psi(s)}\left[\psi(s) - \int_0^{t_1} e^{-st_1'}\psi(t_1')dt_1'\right] = \frac{1}{1-\psi(s)}\int_{t_1}^\infty e^{st_1-st_1'}\psi(t_1')dt_1'. \tag{34}$$

From here we proceed for the limiting cases of long and short ageing times.

4.1 Long ageing time limit of the first or recurrent waiting time

We first consider the long ageing time limit $t_a \gg t_1$, corresponding to the inequality $st_1 \ll 1$ in the ageing time conjugated Laplace space. This implies that $\exp(st_1) \sim 1$. Moreover if we use the form $\psi(s) - \int_0^{t_1}\psi(t_1')\exp(-st_1')dt_1'$ for the integral in the nominator of equation (34) we also observe the inequality $st_1' < st_1 \ll 1$ and thus $\exp(-st_1') \sim 1$ in the integral. Therefore, with the use of relation (18) the forward waiting time density becomes

$$h(t_1, s) \sim \frac{\psi(s) - \int_0^{t_1}\psi(t_1')dt_1'}{1-\psi(s)} \sim \frac{1-\Psi(t)|_{t=1/s}}{\Psi(t)|_{t=1/s}} - \frac{1-\Psi(t_1)}{\Psi(t)|_{t=1/s}} = \frac{\Psi(t_1)}{\Psi(t)|_{t=1/s}} - 1. \tag{35}$$

Since $\Psi(t)|_{t=1/s} \to 0$ when $s \to 0$ we may neglect the term -1 on the right hand side, thus

$$h(t_1, s) \sim \frac{\Psi(t_1)}{\Psi(t)|_{t=1/s}}, \tag{36}$$

when $s \to 0$. Via the Tauberian theorem [57,61] the Laplace inversion of the remaining s dependence in the denominator becomes

$$\mathscr{L}^{-1}\left\{\frac{1}{\Psi(t)|_{t=1/s}}\right\} \sim \frac{d}{dt}\left(\frac{1}{\Psi(t)}\right) = \frac{\psi(t)}{\Psi(t)^2}. \tag{37}$$

Plugging this expression into the forward waiting time (36), we thus find

$$h(t_1, t_a) \sim \Psi(t_1)\frac{\psi(t_a)}{\Psi(t_a)^2}, \tag{38}$$

in our limit $t_a \gg t_1$. Specifically, for our regularised Havlin–Weiss waiting time density involving equations (20) and (21) we thus obtain

$$h(t_1,t_a) \sim \frac{\gamma}{\ln^{\gamma}(\eta+t_1)}\frac{\ln^{\gamma-1}(t_a)}{t_a}, \quad (39)$$

for $t_a \gg t_1$. Thus the leading order long waiting time behaviour of $1/t$ of the waiting time density $\psi(t)$ in expression (20) is replaced by the ageing time dependence $1/t_a$ here. The forward waiting time has an extremely slow logarithmic decay in t_1. Therefore, the first waiting time is significantly more spread than for the power-law case investigated in references [49,50,63–65].

4.2 Short ageing time limit of the first or recurrent waiting time

Let us now get to the limit of short ageing times corresponding to $t_a \ll t_1$ or $st_1 \gg 1$. Since in our numerical simulation η is the shortest time scale we have the additional inequality $t_1 \gg t_a \gg \eta$ or $1/t_1 \ll s \ll s_\eta = 1/\eta$. As typically we can choose $\eta \sim \mathcal{O}(1)$, this implies that $s \ll 1$. In the denominator of equation (34) we can therefore use the replacement $1-\psi(s) \sim \Psi(t)|_{t=1/s}$ and see that the integral in the numerator can be written as

$$\int_{t_1}^{\infty} e^{-st_1'}\psi(t_1')dt_1' = t_1\int_1^{\infty} e^{-st_1q}\psi(qt_1)dq. \quad (40)$$

Due the limit $st_1 \gg 1$ of interest, the latter integral will be dominated by its lower boundary, and thus

$$\int_{t_1}^{\infty} e^{-st_1'}\psi(t_1')dt_1' \approx \frac{t_1\exp(-st_1)\psi(t_1)}{st_1} = \frac{\exp(-st_1)\psi(t_1)}{s}. \quad (41)$$

Combining this result with expressions (34) and (18) we find that

$$h(t_1,s) \sim \frac{\psi(t_1)}{s\Psi(t)|_{t=1/s}}, \quad (42)$$

and thus

$$h(t_1,t_a) \sim \frac{\psi(t_1)}{\Psi(t_a)}. \quad (43)$$

For the regularised Havlin–Weiss waiting time density (20) this lead to

$$h(t_1,t_a) \sim \frac{\gamma\ln^{\gamma}(\eta+t_a)}{(\eta+t_1)\ln^{1+\gamma}(\eta+t_1)}, \quad (44)$$

valid in the limit $t_a \ll t_1$. In this short ageing time limit t_a only features in the logarithm in the nominator and thus only leads to minor corrections to the leading $[t_1\ln^{1+\gamma}(t_1)]^{-1}$ behaviour, as it should (compare with equation (20)).

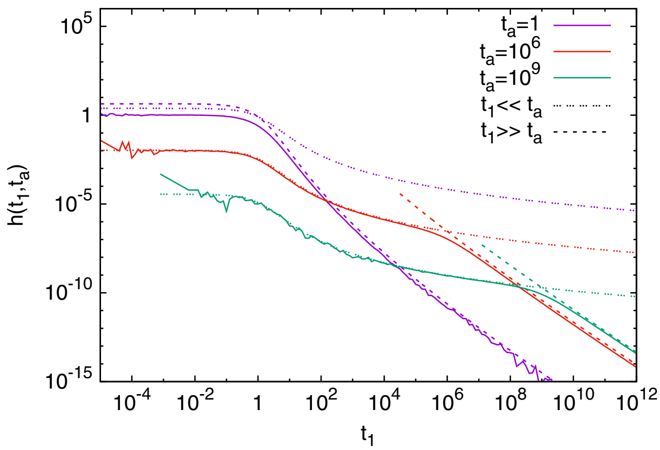

Fig. 1. Forward or recurrent waiting time density $h(t_1,t_a)$ as function of the forward waiting time t_1, shown for different ageing times t_a, for the case of the waiting time density (20) with $\gamma = 4$. In the strong ageing regime when $t_a \gg t_1$, relation (39) shows very good agreement with the data, while in the opposite limit $t_1 \gg t_a$ equation (44) agrees equally well with the data. The crossover times also nicely correspond to the crossover behaviour in the simulations results. We note that the relatively small discrepancy in the case $t_a = 1$ is due to the fact that in this case the limit $t_a \gg \eta$ is not fulfilled for the value $\eta = e$ chosen here.

Figure 1 shows the forward or recurrent waiting time density as function of t_1 and for different t_a for the case of he Havlin–Weiss waiting time density (20). Good agreement with the results (44) and (39) is observed.

5 Ageing Montroll–Weiss equation for ultraslow continuous time random walks

Which form does the analogue to the classical Montroll–Weiss equation (1) assume for ageing ultraslow continuous time random walks? Barkai showed how the ageing effects modify the Montroll–Weiss equation for power-law waiting time densities with diverging characteristic waiting time [63–65]. Similar to that approach we consider the ageing probability density function $P(x,t,t_a)$ depending on both the process time t and the ageing time t_a. For the convenience of the reader we summarise the related formalism in Appendix A. From these results we can rephrase the Fourier $(x \to k)$ and double Laplace transform $(t \to u$ and $t_a \to s)$ of $P(x,t,t_a)$ in the form

$$P(k,u,s) = p_0(u,s) + h(u,s)\lambda(k)P_{MW}(k,u), \quad (45)$$

where $p_0(t,t_a)$ denotes the probability of making no jumps up to time t given the ageing time t_a, see equations (A.2) and (A.3). The normalisation condition $P(k=0,u,s) = 1/(us)$ is fulfilled, as can be seen from equations (A.3) and (31). As observed in references [49,50] this way of writing the ageing Montroll–Weiss equation shows that the diffusing particles split up into a discrete, fully immobile fraction proportional to p_0 and those particles that perform a distribution of steps after the recurrence waiting

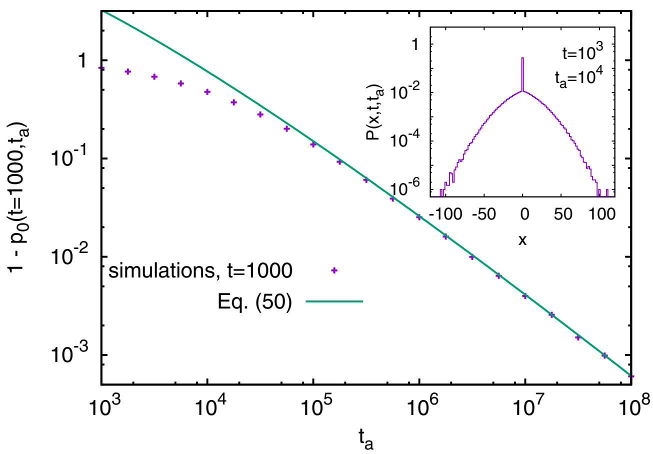

Fig. 2. Peak height $p_0(t,t_a)$ of the singular part of the probability density function $P(x,t,t_a)$, for the case of the waiting time density (20) with $\gamma=4$ and $\eta=e$. Good agreement of the simulations with the numerical evaluation of result (50) is observed in its regime of validity, $t_a \gg t$. Inset: Histogram representing the probability density function $P(x,t,t_a)$, from simulations of 10^6 trajectories. The peak according to equation (45) is distinct.

time. This population splitting is one of the characteristic features of ageing continuous time random walks with diverging characteristic waiting time [49,50].

Let us investigate the singular part p_0 in more detail for the regularised Havlin–Weiss waiting time density (20). In the strong ageing limit with $t_a \gg t \gg \eta$ we use our previous result (39) and insert this into equation (A.2), yielding

$$p_0(t,t_a) = 1 - \frac{\gamma \ln^{\gamma-1}(t_a)}{t_a}\int_0^t \frac{dt'}{\ln^\gamma(\eta+t')}. \tag{46}$$

For the concrete value $\gamma=4$ of Sinai-like diffusion case used in our numerical examples herein, the integral can be evaluated as follows. An integral of the form

$$\int \frac{dx}{\ln^4 x} = \int \frac{x dx}{x \ln^4 x}, \tag{47}$$

can, via integration by parts, be brought to the form

$$\int \frac{dx}{\ln^4 x} = -\frac{x}{3\ln^3 x} + \frac{1}{3}\int \frac{dx}{\ln^3 x}. \tag{48}$$

Repeated use of this trick leads to the form

$$\int \frac{dx}{\ln^4 x} = -\frac{x}{3\ln^3 x} - \frac{x}{6\ln^2 x} - \frac{x}{6\ln x} + \frac{1}{6}\int \frac{dx}{\ln x}. \tag{49}$$

Thus, we find the closed form expression

$$p_0(t,t_a) = 1 - \frac{4\ln^3 t_a}{t_a} \times I(t), \tag{50}$$

where

$$\begin{aligned} I(t) &= \int_0^t \frac{dt'}{\ln^4(\eta+t')} \\ &= -\frac{\eta+t}{6\ln^3(\eta+t)} \times \left(2+\ln(\eta+t)+\ln^2(\eta+t)\right) \\ &\quad + \frac{\eta}{6\ln^3\eta}\left(2+\ln\eta+\ln^2\eta\right) \\ &\quad + \frac{1}{6}\left(\mathrm{li}(\eta+t)-\mathrm{li}(\eta)\right), \end{aligned} \tag{51}$$

with the logarithmic integral

$$\mathrm{li}(t) = \int_0^t \frac{dt'}{\ln t'}. \tag{52}$$

In Figure 2 we show the peak height $p_0(t,t_a)$ of the singular part of the probability density function $P(x,t,t_a)$. We observe very good agreement with the numerical evaluation of result (50) for longer ageing times. In the inset of Figure 2 we also demonstrate the shape of the probability density function $P(x,t,t_a)$, illustrating the distinct peak versus the continuous remainder according to equation (45).

We note that it is worthwhile comparing the ageing properties of the annealed ultraslow continuous time random walk model studied here with those of the quenched Sinai model [29–34]. Thus, the probability corresponding to our $p_0(t,t_a)$ is given by equation (138) of reference [33] which in the strong ageing regime $t \ll t_a$ reads (in our notation, the upper index stands for the Sinai model)

$$\begin{aligned} p_0^{(S)}(t,t_a) &\sim 1 - \frac{4}{3}\left(\frac{\ln(t_a+t)}{\ln t_a} - 1\right) \\ &\sim 1 - \frac{4}{3}\frac{t}{t_a \ln t_a}. \end{aligned} \tag{53}$$

This behaviour is different from our result (50) for the annealed continuous time random walk formulation, clearly demonstrating the difference in the dynamics of the two models. As the probability is not a mean field quantity, this difference is not really surprising. We will see below, in contrast, that the ageing second moment of both models is indeed equivalent, similar to the non-ageing case studied in reference [47].

6 Ageing ultraslow mean squared displacement

Differentiation of expression (A.7) twice with respect to the wave number k and setting $k=0$, we find that

$$\langle x^2(u,s)\rangle = \frac{\sigma^2 h(u,s)}{u[1-\psi(u)]}, \tag{54}$$

where, as above, σ^2 is the second moment of the jump length density $\lambda(k)$. With the property (18) for ultraslow

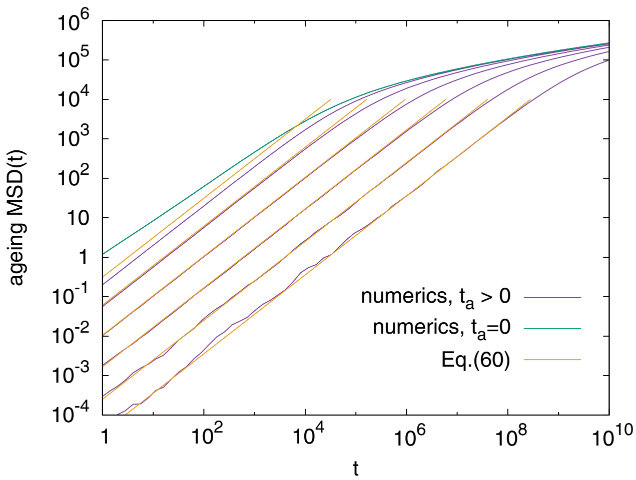

Fig. 3. Mean squared displacement $\langle x^2(t,t_a)\rangle$ of the ageing ultraslow continuous time random walk based on the waiting time density (20) with $\gamma = 4$ and $\eta = e$, as function of time t shown for the different ageing times $t_a = 0$ (top curve), $t_a = 10^4, 10^5, \ldots, 10^9$. Good agreement with the linear t-dependence predicted by the theoretical result (61), shown by the orange lines, is observed. At long times, according to result (58), all curves merge, independent of the ageing time t_a. Note that the crossover time between both regimes agrees nicely with the indicated ageing times. For each curve an average over 10^6 trajectories was performed.

motion we obtain

$$\langle x^2(u,s)\rangle \sim \frac{\sigma^2}{u(u-s)}\left(\frac{1}{\Psi(t)|_{t=1/s}} - \frac{1}{\Psi(t)|_{t=1/u}}\right). \quad (55)$$

For short ageing times with $t \gg t_a$ or $s \gg u$ we have that $1/\Psi(t)|_{t=1/s} \ll 1/\Psi(t)|_{t=1/u}$ and thus

$$\langle x^2(u,s)\rangle \sim \frac{\sigma^2}{us}\frac{1}{\Psi(t)|_{t=1/u}}, \quad (56)$$

such that

$$\langle x^2(t,t_a)\rangle \sim \frac{\sigma^2}{\Psi(t)}, \quad (57)$$

which matches exactly the mean squared displacement of the non-ageing ultraslow continuous time random walk [47]. For the regularised Havlin–Weiss waiting time density (20), this corresponds to [47]

$$\langle x^2(t,t_a)\rangle \sim \frac{\sigma^2 \ln^\gamma(\eta+t)}{\ln^\gamma \eta}, \quad t_a \ll t. \quad (58)$$

At long ageing times with $t_a \gg t$ or $u \gg s$ we have the opposite case, $1/\Psi(t)|_{t=1/u} \ll 1/\Psi(t)|_{t=1/s}$ and thus

$$\langle x^2(u,s)\rangle \sim \frac{\sigma^2}{u^2}\frac{1}{\Psi(t)|_{t=1/s}}, \quad (59)$$

such that

$$\langle x^2(t,t_a)\rangle \sim \sigma^2 t\frac{\psi(t_a)}{\Psi(t_a)}. \quad (60)$$

For the regularised Havlin–Weiss waiting time density we find

$$\langle x^2(t,t_a)\rangle \sim \frac{\sigma^2\gamma}{\ln^\gamma \eta}\frac{\ln^{\gamma-1}(\eta+t_a)}{\eta+t_a}t, \quad t_a \gg t, \quad (61)$$

according to which the leading order behaviour exhibits a normal diffusive scaling proportional to t, albeit with an ageing time corrected prefactor.

Figure 3 shows the mean squared displacement $\langle x^2(t)\rangle$ as function of time t for different ageing times, for the Havlin–Weiss waiting time density (20). Good agreement of the simulations results with the theoretical predictions is observed. Small fluctuations at very long ageing times are due to the fact that most of the trajectories do not exhibit any jumps at all up to some large time t. Thus the value of the MSD is determined by those few trajectories containing jumps, while the MSD curves get smoother for longer t.

Let us come back to our comparison with the ageing Sinai model [32–34]. The analogue to the ageing mean squared displacement for the Sinai case is given by equation (153) in reference [32–34] (note that their second moment of the relative displacement corresponds exactly to our $\langle x^2(t,t_a)\rangle$). Thus, in the strong ageing limit $t \ll t_a$, up to a numerical prefactor (see Eqs. (153) and (157) in that paper) and in our notation, we see that

$$\langle [x^2(t,t_a)]^{(S)}\rangle \sim \ln^4 t_a\left(\frac{\ln(t_a+t)}{\ln t_a} - 1\right) \sim \frac{\ln^3 t_a}{t_a}t, \quad (62)$$

which agrees with our result (61). Thus, the ageing mean squared displacement of the ultraslow continuous time random walk model coincides (up to a prefactor) with that of the quenched Sinai model. As mentioned above (and for the non-ageing case in reference [47]) in the mean field sense both the annealed ultraslow continuous time random walk and the quenched Sinai model are equivalent.

7 Ageing ultraslow time averaged mean squared displacement

In experiments tracking individual particles such as artificial or endogenous tracers in living biological cells [3–5,9–13] often few but long individual trajectories $x(t)$ of length T are measured. These are routinely evaluated in terms of the time averaged mean squared displacement

$$\overline{\delta_a^2(\Delta)} = \frac{1}{T-\Delta}\int_{t_a}^{T+t_a-\Delta}\Big(x(t+\Delta) - x(t)\Big)^2 dt. \quad (63)$$

When $t_a = 0$ this is the regular time averaged mean squared displacement. In the presence of an ageing period t_a before the measurement of length T, the ageing time explicitly features in the integration limits [49,50]. In order to extract analytical quantities for continuous time random walk processes, we introduce the additional average $\langle\cdot\rangle$ over an ensemble of N trajectories,

$$\left\langle\overline{\delta_a^2(\Delta)}\right\rangle = \frac{1}{N}\sum_{i=1}^{N}\overline{\delta_{a,i}^2(\Delta)} = \frac{1}{T-\Delta} \times \int_{t_a}^{T+t_a-\Delta}\left\langle\Big(x(t+\Delta)-x(t)\Big)^2\right\rangle dt. \quad (64)$$

For the evaluation of experiments, this additional averaging is often taken in order to produce a smoother behaviour of the time average.

To proceed we note that the mean squared displacement in the non-aged case is readily obtained from the equivalence with the mean number $\langle n(t)\rangle$ of steps up to time t, multiplied with the variance of the jump lengths [49,50,67,68],

$$\langle x^2(t)\rangle = \sigma^2\langle n(t)\rangle. \quad (65)$$

Due to relation (25) we see that the number of jumps can be reexpressed for ultraslow continuous time random walks in terms of the stalling probability,

$$\langle n(t)\rangle \sim \frac{1}{\Psi(t)}. \quad (66)$$

Thus,

$$\left\langle\Big(x(t_2)-x(t_1)\Big)^2\right\rangle = \sigma^2\Big(\langle n(t_2)\rangle - \langle n(t_1)\rangle\Big). \quad (67)$$

Plugging this into the definition (64) for the time averaged mean squared displacement, we find

$$\left\langle\overline{\delta_a^2(\Delta)}\right\rangle \sim \frac{\sigma^2}{T-\Delta} \times \int_{t_a}^{T+t_a-\Delta}\left(\frac{1}{\Psi(t+\Delta)}-\frac{1}{\Psi(t)}\right)dt. \quad (68)$$

To proceed, we approximate the integrand by a first order Taylor expansion as

$$\frac{1}{\Psi(t+\Delta)}-\frac{1}{\Psi(t)} \sim \frac{1}{\Psi(t)+\Delta\frac{d\Psi(t)}{dt}}-\frac{1}{\Psi(t)} \sim \frac{1}{\Psi(t)}\left(\frac{1}{1+\Delta\frac{d\Psi(t)}{dt}\Psi(t)}-1\right) \sim -\Delta\frac{\frac{d\Psi(t)}{dt}}{\Psi^2(t)} = \Delta\frac{d}{dt}\left(\frac{1}{\Psi(t)}\right). \quad (69)$$

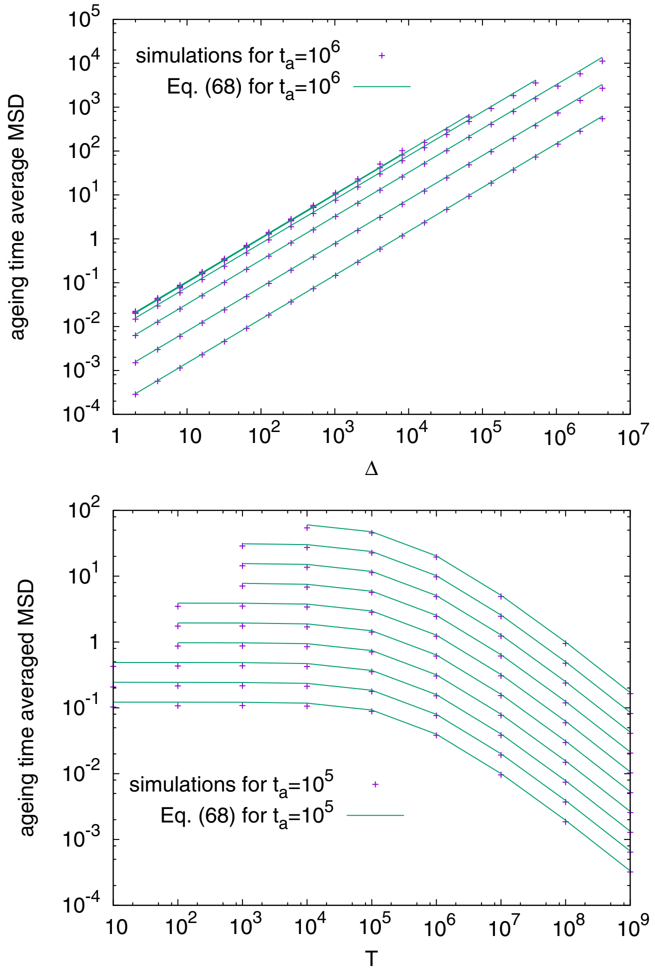

Fig. 4. Ageing time averaged mean squared displacement $\left\langle\overline{\delta_a^2(\Delta)}\right\rangle$ as function of the lag time Δ (Top) and measurement time T (Bottom) shown for the indicated ageing times, based on the waiting time density (20) with $\gamma = 4$ and $\eta = e$. In the top panel the different curves correspond to the different measurement times $T = 10, 10^2, \ldots, 10^9$ (top to bottom in this panel), where the curves for shorter T show significant overlap in the plot. In the bottom panel we varied the lag times as powers of 2, i.e., $\Delta = 2, 4, \ldots, 1024$ (bottom to top in this panel). Excellent agreement with the analytical result (70) is found in both panels.

For sufficiently small Δ we can therefore evaluate the integral in equation (68) and obtain (see Appendix B)

$$\left\langle\overline{\delta_a^2(\Delta)}\right\rangle \sim \frac{\sigma^2\Delta}{T}\left(\frac{1}{\Psi(T+t_a)}-\frac{1}{\Psi(t_a)}\right). \quad (70)$$

In the absence of ageing, $t_a \to 0$, the leading behaviour is

$$\left\langle\overline{\delta_a^2(\Delta)}\right\rangle \sim \left\langle\overline{\delta^2(\Delta)}\right\rangle \sim \sigma^2\Delta/[T\Psi(T)], \quad (71)$$

which matches the result found for the non-ageing ultraslow continuous time random walk in reference [47] (Fig. 4).

Let us illustrate our result (70) for the regularised Havlin–Weiss waiting time density (20). Plugging in the associated stalling probability (21), we find

$$\left\langle\overline{\delta_a^2(\Delta)}\right\rangle \sim \frac{\ln^\gamma(T+t_a+\eta)-\ln^\gamma(t_a+\eta)}{\ln^\gamma(T+\eta)} \times \left\langle\overline{\delta^2(\Delta)}\right\rangle, \tag{72}$$

in terms of the non-aged time averaged mean squared displacement. Thus, similar to other ageing anomalous diffusion processes [6,49,50,69–71], the physically relevant functional dependence on the lag time Δ is completely unaffected by t_a, the latter entering solely through a prefactor.

8 Conclusions

We here established the framework for ageing ultraslow continuous time random walk processes. In particular, we derived the expression for the forward or recurrent waiting time, the peak hight of the singular part of the ageing probability density function as well as the ensemble and time averaged mean squared displacements. Asymptotic results were presented for the limits of short and long ageing times. Good agreement is observed between our analytical results and stochastic simulations.

Ultraslow diffusion arises in numerous strongly disordered or interacting systems. In these ageing effects occur naturally in many experimental situations. The generalisation of the seminal continuous time random walk theory for the ultraslow case in the presence of ageing is therefore a relevant step towards the completion of the continuous time random walk framework.

RM acknowledges financial support from the Academy of Finland within the Finland Distinguished Professorship programme. RM and AVC acknowledge funding from the Deutsche Forschungsgemeinschaft, project ME 1535/6-1.

Author contribution statement

All authors were involved in the preparation of the manuscript. All authors have read and approved the final manuscript.

Appendix A: Ageing Montroll–Weiss formalism

The dual Laplace transform $t \to u$ and $t_a \to s$ and Fourier transform $x \to k$ of $P(x, t, t_a)$ can then be written in the form

$$P(k,u,s) = \sum_{n=0}^{\infty} p_n(u,s)\lambda^n(k), \tag{A.1}$$

where $p_n(t, t_a)$ is the probability to make n steps in an interval of length t after the ageing period t_a, and $\lambda(x)$ is our jump length density. The probability of making no jumps up to time t can be expressed by help of the recurrence waiting time.

$$p_0(t,t_a) = 1 - \int_0^t h(t_1', t_a)dt_1', \tag{A.2}$$

or

$$p_0(u,s) = \frac{1}{us} - \frac{h(u,s)}{u} = \frac{1-sh(u,s)}{us}. \tag{A.3}$$

A number $n \geq 1$ of jumps up to time t requires the first step and then $(n-1)$ regular steps, before a stalling time up to t,

$$p_n(u,s) = h(u,s)\psi^{n-1}(u)\frac{1-\psi(u)}{u}. \tag{A.4}$$

Note that in the absence of ageing ($t_a \to 0$) we readily find that $p_0(t, t_a \to 0) = 1 - \int_0^t \psi(t')dt' = \Psi(t)$, as it should. Now, from equation (A.1) we have (see Eq. (31) for $h(u,s)$)

$$\begin{aligned} P(k,u,s) &= p_0(u,s) + \sum_{n=1}^{\infty} p_n(u,s)\lambda^n(k) \\ &= \frac{1-sh(u,s)}{us} \\ &\quad + \sum_{n=1}^{\infty} h(u,s)\psi^{n-1}(u)\frac{1-\psi(u)}{u}\lambda^n(k) \\ &= \frac{1}{us} + \frac{\psi(u)-\psi(s)}{u(u-s)[1-\psi(s)]} \\ &\quad + \frac{[\psi(s)-\psi(u)][1-\psi(u)]}{u(u-s)[1-\psi(s)]} \\ &\quad \times \sum_{n=1}^{\infty}\psi^{n-1}(u)\lambda^n(k) \\ &= \frac{1}{us} + \frac{\psi(u)-\psi(s)}{u(u-s)[1-\psi(s)]} \\ &\quad \times \left(1 - [1-\psi(u)]\sum_{n=1}^{\infty}\psi^{n-1}(u)\lambda^n(k)\right) \\ &= \frac{1}{us} + \frac{\psi(u)-\psi(s)}{u(u-s)[1-\psi(s)]} \\ &\quad \times \left(1 - \sum_{n=1}^{\infty}\psi^{n-1}(u)\lambda^n(k) \right. \\ &\quad \left. + \sum_{n=1}^{\infty}\psi^{n}(u)\lambda^n(k)\right). \end{aligned} \tag{A.5}$$

The last expression in large round parentheses can be rewritten as

$$1+\sum_{n=1}^{\infty}\psi^n(u)\lambda^n(k)-\frac{1}{\psi(u)}\sum_{n=1}^{\infty}\psi^n(u)\lambda^n(k) =\frac{1}{1-\psi(u)\lambda(k)}-\frac{1}{\psi(u)}\left(\frac{1}{1-\psi(u)\lambda(k)}-1\right) =\frac{1-\lambda(k)}{1-\psi(u)\lambda(k)}. \quad (A.6)$$

We thus obtain the result for the ageing Montroll–Weiss equation,

$$P(k,u,s)=\frac{1}{us}+\frac{\psi(u)-\psi(s)}{u(u-s)[1-\psi(s)]}\times\frac{1-\lambda(k)}{1-\psi(u)\lambda(k)}, \quad (A.7)$$

which features the additional pre-factor proportional to the dual Laplace transform of the forward waiting time in comparison to the standard Montroll–Weiss equation. This result was obtained previously by Barkai [63–65].

It is instructive to rewrite the result (A.7) in terms of the regular Montroll–Weiss expression

$$P_{MW}(k,u)=\frac{1-\psi(u)}{u}\frac{1}{1-\psi(u)\lambda(k)}. \quad (A.8)$$

We expand equation (A.7) by $h(u,s)$ in the form

$$P(k,u,s)=\frac{1}{us}-\frac{h(u,s)}{u}+\frac{h(u,s)}{u}\times\left(1+\frac{u}{h(u,s)}\frac{\psi(u)-\psi(s)}{u(u-s)[1-\psi(s)]}\times\frac{1-\lambda(k)}{1-\psi(u)\lambda(k)}\right) =p_0(u,s)+\frac{h(u,s)}{u}\times\left(1-\frac{1-\lambda(k)}{1-\psi(u)\lambda(k)}\right), \quad (A.9)$$

from which equation (45) follows immediately.

Appendix B: Derivation of equation (70)

Here we confirm the result obtained in equation (70) by direct calculation of the integral (68) for the case of Sinai-like diffusion with $\gamma=4$. Plugging $\Psi(t)$ from equation (19) into equation (68) we use

$$\int\ln^4 x dx=x\ln^4 x-4xln^3x +12x\ln^2 x-24x\ln x+24x, \quad (B.1)$$

leading us to the somewhat lengthy expression

$$\left\langle\overline{\delta_a^2(\Delta)}\right\rangle=\frac{\sigma^2}{(T-\Delta)\ln^4\eta}\times\{(T+t_a+\eta)\ln^4(T+t_a+\eta) -(T+t_a+\eta-\Delta)\ln^4(T+t_a+\eta-\Delta) -(t_a+\Delta+\eta)\ln^4(t_a+\Delta+\eta) +(t_a+\eta)\ln^4(t_a+\eta) -4(T+t_a+\eta)\ln^3(T+t_a+\eta) +4(T+t_a+\eta-\Delta)\ln^3(T+t_a+\eta-\Delta) +4(t_a+\Delta+\eta)\ln^3(t_a+\Delta+\eta) -4(t_a+\eta)\ln^3(t_a+\eta) +12(T+t_a+\eta)\ln^2(T+t_a+\eta) -12(T+t_a+\eta-\Delta)\ln^2(T+t_a+\eta-\Delta) -12(t_a+\Delta+\eta)\ln^2(t_a+\Delta+\eta) +12(t_a+\eta)\ln^2(t_a+\eta) -24(T+t_a+\eta)\ln(T+t_a+\eta) +24(T+t_a+\eta-\Delta)\ln(T+t_a+\eta-\Delta) +24(t_a+\Delta+\eta)\ln(t_a+\Delta+\eta) -24(t_a+\eta)\ln(t+a+\eta)\}. \quad (B.2)$$

Now we assume that $\Delta\ll T,t_a$, expand all expressions and restrict ourselves to terms linear in Δ. For instance,

$$(T+t_a+\eta-\Delta)\times\ln^4(T+t_a+\eta-\Delta) =(T+t_a+\eta)\left(1-\frac{\Delta}{T+t_a+\eta}\right)\times\ln^4\left[(T+t_a+\eta)\left(1-\frac{\Delta}{T+t_a+\eta}\right)\right] \sim(T+t_a+\eta)\left(1-\frac{\Delta}{T+t_a+\eta}\right)\times\left[\ln(T+t_a+\eta)-\frac{\Delta}{T+t_a+\eta}\right]^4 \sim(T+t_a+\eta)\ln^4(T+t_a+\eta)\times\left[1-\frac{\Delta}{T+t_a+\eta}-\frac{4\Delta}{(t+t_a+\eta)\ln(T+t_a+\eta)}\right] \quad (B.3)$$

and

$$(t_a+\Delta+\eta)\times\ln^4(t_a+\Delta+\eta) \sim(t_a+\eta)\ln^4(t_a+\eta)\times\left[1+\frac{\Delta}{t_a+\eta}+\frac{4\Delta}{(t_a+\eta)\ln(t_a+\eta)}\right], \quad (B.4)$$

and so forth. After plugging in all these expression in to equation (B.2) successive terms turn out to cancel such that the linear approximation in Δ finally produces

$$\left\langle \overline{\delta_a^2(\Delta)} \right\rangle \sim \frac{\sigma^2 \Delta}{T \ln^4 \eta} \times \left[\ln^4(T + t_a + \eta) - \ln^4(t_a + \eta) \right], \quad \text{(B.5)}$$

which is equivalent to equation (70).

References

1. R. Metzler, J. Klafter, Phys. Rep. **339**, 1 (2000)
2. R. Metzler, J. Klafter, J. Phys. A **37**, R161 (2004)
3. E. Barkai, Y. Garini, R. Metzler, Phys. Today **65**, 29 (2012)
4. F. Höfling, T. Franosch, Rep. Progr. Phys. **76**, 046602 (2013)
5. K. Nørregaard, R. Metzler, C.M. Ritter, K. Berg-Sørensen, L.B. Oddershede, Chem. Rev. **117**, 4342 (2017)
6. R. Metzler, J.-H. Jeon, A.G. Cherstvy, E. Barkai, Phys. Chem. Chem. Phys. **16**, 24128 (2014)
7. I.M. Sokolov, Soft Matter **8**, 9043 (2012)
8. Y. Meroz, I.M. Sokolov, Phys. Rep. **573**, 1 (2015)
9. J.-H. Jeon, V. Tejedor, S. Burov, E. Barkai, C. Selhuber-Unkel, K. Berg-Sørensen, L. Oddershede, R. Metzler, Phys. Rev. Lett. **106**, 048103 (2011)
10. S.M.A. Tabei, S. Burov, H.Y. Kim, A. Kuznetsov, T. Huynh, J. Jureller, L.H. Philipson, A.R. Dinner, N.F. Scherer, Proc. Natl. Acad. Sci. USA **110**, 4911 (2013)
11. I. Golding, E.C. Cox, Phys. Rev. Lett. **96**, 098102 (2006)
12. I. Bronstein, Y. Israel, E. Kepten, S. Mai, Y. Shav-Tal, E. Barkai, Y. Garini, Phys. Rev. Lett. **103**, 018102 (2009)
13. C. di Rienzo, V. Piazza, E. Gratton, F. Beltram, F. Cardarelly, Nat. Commun. **5**, 5891 (2014)
14. M. Weiss, H. Hashimoto, T. Nilsson, Biophys. J. **84**, 4043 (2003)
15. A. Honigmann, V. Mueller, S.W. Hell, C. Eggeling, Faraday Discuss. **161**, 77 (2013)
16. A.V. Weigel, B. Simon, M.M. Tamkun, D. Krapf, Proc. Natl. Acad. Sci. USA **108**, 6438 (2011)
17. C. Manzo et al., Phys. Rev X **5**, 011021 (2015)
18. J. Szymanski, M. Weiss, Phys. Rev. Lett. **103**, 038102 (2009)
19. W. Pan, L. Filobelo, N.D.Q. Pham, O. Galkin, V.V. Uzunova, P.G. Vekilov, Phys. Rev. Lett. **102**, 058101 (2009)
20. J.-H. Jeon, N. Leijnse, L.B. Oddershede, R. Metzler, New J. Phys. **15**, 045011 (2013)
21. G.R. Kneller, K. Baczynski, M. Pasenkiewicz-Gierula, J. Chem. Phys. **135**, 141105 (2011)
22. S. Stachura, G.R. Kneller, J. Chem. Phys. **40**, 245 (2014)
23. M. Javanainen, H. Hammaren, L. Monticelli, J.-H. Jeon, M.S. Miettinen, H. Martinez-Seara, R. Metzler, I. Vattulainen, Faraday Discuss. **161**, 397 (2013)
24. J.-H. Jeon, H.M. Monne, M. Javanainen, R. Metzler, Phys. Rev. Lett. **109**, 188103 (2012)
25. J.-H. Jeon, M. Javanainen, H. Martinez-Seara, R. Metzler, I. Vattulainen, Phys. Rev. X **6**, 021006 (2016)
26. A. Caspi, R. Granek, M. Elbaum, Phys. Rev. Lett. **85**, 5655 (2000)
27. D. Robert, T.H. Nguyen, F. Gallet, C. Wilhelm, PLoS ONE **4**, e10046 (2010)
28. J.F. Reverey, J.-H. Jeon, H. Bao, M. Leippe, R. Metzler, C. Selhuber-Unkel, Sci. Rep. **5**, 11690 (2015)
29. Ya.G. Sinai, Theory Prob. Appl. **27**, 256 (1982)
30. D.E. Temkin, Sov. Math. Dokl. **13**, 1172 (1972)
31. G. Oshanin, A. Rosso, G. Schehr, Phys. Rev. Lett. **110**, 100602 (2013)
32. L. Laloux, P. Le Doussal, Phys. Rev. E **57**, 6296 (1998)
33. P. Le Doussal, C. Monthus, D.S. Fischer, Phys. Rev. E **59**, 4795 (1999)
34. D.S. Fisher, P. Le Doussal, C. Monthus, Phys. Rev. E **64**, 066107 (2001)
35. J. Dräger, J. Klafter, Phys. Rev. Lett. **84**, 5998 (2000)
36. N.V. Brilliantov, T. Pöschel, *Kinetic theory of granular gases* (Oxford University Press, Oxford, UK, 2004)
37. A. Bodrova, A.V. Chechkin, A.G. Cherstvy, R. Metzler, Phys. Chem. Chem. Phys. **17**, 21791 (2015)
38. L.P. Sanders, M.A. Lomholt, L. Lizana, K. Fogelmark, R. Metzler, T. Ambjörnsson, New J. Phys. **16**, 113050 (2014)
39. M.A. Lomholt, L. Lizana, R. Metzler, T. Ambjörnsson, Phys. Rev. Lett. **110**, 208301 (2013)
40. A.G. Cherstvy, R. Metzler, Phys. Chem. Chem. Phys. **15**, 20220 (2013)
41. A.G. Cherstvy, A.V. Chechkin, R. Metzler, New J. Phys. **15**, 083039 (2013)
42. A. Bodrova, A.V. Chechkin, A.G. Cherstvy, R. Metzler, New J. Phys. **17**, 063038 (2015)
43. A. Bodrova, A.V. Chechkin, A.G. Cherstvy, H. Safdari, I.M. Sokolov, R. Metzler, Sci. Rep. **6**, 30520 (2016)
44. A.V. Chechkin, J. Klafter, I.M. Sokolov, EPL **63**, 326 (2003)
45. T. Sandev, A.V. Chechkin, N. Korabel, H. Kantz, I.M. Sokolov, R. Metzler, Phys. Rev. E **92**, 042117 (2015)
46. S. Havlin, G.H. Weiss, J. Stat. Phys. **58**, 1267 (1990)
47. A. Godec, A.V. Chechkin, E. Barkai, H. Kantz, R. Metzler, J. Phys. A **47**, 492002 (2014)
48. J. Kursawe, J. Schulz, R. Metzler, Phys. Rev. E **88**, 062124 (2013)
49. J.H.P. Schulz, E. Barkai, R. Metzler, Phys. Rev. Lett. **110**, 020602 (2013)
50. J.H.P. Schulz, E. Barkai, R. Metzler, Phys. Rev. X **4**, 011028 (2014)
51. E.W. Montroll, G.H. Weiss, J. Math. Phys. **10**, 753 (1969)
52. H. Scher, E.W. Montroll, Phys. Rev. B **12**, 2455 (1975)
53. J. Klafter, A. Blumen, M.F. Shlesinger, Phys. Rev. A **35**, 3081 (1987)
54. R. Metzler, E. Barkai, J. Klafter, Phys. Rev. Lett. **82**, 3563 (1999)
55. E. Barkai, R. Metzler, J. Klafter, Phys. Rev. E **61**, 132 (2000)
56. R. Metzler, E. Barkai, J. Klafter, Europhys. Lett. **46**, 431 (1999)
57. B.D. Hughes, *Random walks and random environments: random walks* (Oxford University Press, Oxford, UK, 1995), Vol. 1
58. S.I. Denisov, H. Kantz, Phys. Rev. E **83**, 041132 (2011)
59. S.I. Denisov, S.B. Yuste, Yu.S. Bystrik, H. Kantz, K. Lindenberg, Phys. Rev. E **84**, 061143 (2011)
60. S.I. Denisov, H. Kantz, Europhys. Lett. **92**, 30001 (2010)
61. W. Feller, *An introduction to probability theory and its application* (Wiley, New York, NY, 1970)
62. C. Godrèche, J.M. Luck, J. Stat. Phys. **104**, 489 (2001)
63. E. Barkai, Phys. Rev. Lett. **90**, 104101 (2003)

64. E. Barkai, Y.C. Cheng, J. Chem. Phys. **118**, 6167 (2003)
65. The forward waiting time can also be recovered as a temporal leapover: T. Koren, M.A. Lomholt, A.V. Chechkin, J. Klafter, R. Metzler, Phys. Rev. Lett. **99**, 160602 (2007)
66. D.R. Cox, *Renewal theory* (Methuen & Co. Ltd., London, 1962)
67. Y. He, S. Burov, R. Metzler, E. Barkai, Phys. Rev. Lett. **101**, 058101 (2008)
68. A. Lubelski, I.M. Sokolov, J. Klafter, Phys. Rev. Lett. **100**, 250602 (2008)
69. H. Safdari, A.V. Chechkin, G. Jafari, R. Metzler, Phys. Rev. E **91**, 042107 (2015)
70. H. Safdari, A.G. Cherstvy, A.V. Chechkin, A. Bodrova, R. Metzler, Phys. Rev. E **95**, 012120 (2017)
71. A.G. Cherstvy, A.V. Chechkin, R. Metzler, J. Phys. A **47**, 485002 (2014)