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## Severe slowing-down and universality of the dynamics in disordered interacting many-body systems: ageing and ultraslow diffusion

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### Abstract

Low-dimensional, many-body systems are often characterized by ultraslow dynamics. We study a labelled particle in a generic system of identical particles with hard-core interactions in a strongly disordered environment. The disorder is manifested through intermittent motion with scale-free sticking times at the single particle level. While for a non-interacting particle we find anomalous diffusion of the power-law form  $\langle x^2(t) \rangle \simeq t^\alpha$  of the mean squared displacement with  $0 < \alpha < 1$ , we demonstrate here that the combination of the disordered environment with the many-body interactions leads to an ultraslow, logarithmic dynamics  $\langle x^2(t) \rangle \simeq \log^{1/2} t$  with a universal 1/2 exponent. Even when a characteristic sticking time exists but the fluctuations of sticking times diverge we observe the mean squared displacement  $\langle x^2(t) \rangle \simeq t^\gamma$  with  $0 < \gamma < 1/2$ , that is slower than the famed Harris law  $\langle x^2(t) \rangle \simeq t^{1/2}$  without disorder. We rationalize the results in terms of a subordination to a counting process, in which each transition is dominated by the forward waiting time of an ageing continuous time process.



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Keywords: single-file diffusion, continuous time random walks, ageing

## 1. Introduction

Understanding effects due to many-body interactions represents one of the fundamental problems in physics and chemistry [1]. A generic model combining many-body interactions with the stochastic motion of the individual particles in the system is single file motion, in which diffusing particles with hard core, excluded volume interactions move in one dimension. The resulting collisions between the particles in the single file severely alter the Brownian law performed by any of the particles in the case when no excluded volume interactions were present. As shown by Harris in 1965, the motion of a labelled tracer particle in a single file of particles is characterized by the square-root scaling

$$\langle x^2(t) \rangle \simeq K_{1/2} t^{1/2} \quad (1)$$

of the mean squared displacement [2]. Theoretically and numerically, single file diffusion was studied extensively [3–13], in particular, regarding the effects of physical parameters such as the particle density, the system size, or external forcing. The so-called harmonization approach, which maps the long-time dynamics for such systems onto the motion of harmonically coupled beads with spring constants determined by the system's equation of state, shows that after integrating out all other particle co-ordinates, the motion of the labelled tracer particle is described by a fractional Langevin equation [14].

Wei *et al* in a seminal study provided direct experimental evidence for the single file law (1) of colloidal particles in circular grooves and corroborated the asymptotically Gaussian distribution of the labelled particle's position [15]. Channels for the study of single file motion can also be established by means of optical tweezers [16] to study the crossover from the initial free motion of the particles before collisions take place, to the single file regime with the  $t^{1/2}$  scaling (1) of the mean squared displacement [17]. Earlier, single file diffusion was also discovered in zeolites—so-called molecular sieves—by pulsed field gradient NMR methods [18]. Single file motion in micro- and nanochannels is studied in view of potential technological applications [19, 20]. One of these applications is promoted to be the slow release of drugs from nanochannel containers [20]. In biological contexts, single file motion arises in the transport of molecules through membrane channels [21].

Here we address the fundamental question of how disorder of the surrounding medium affects the dynamics of an interacting many-body system. In our prototype physical model the disorder leads to a single particle sticking time density of power law form with exponent  $\alpha$ . In this scenario we demonstrate that while a single, non-interacting particle in the presence of annealed disorder performs anomalous diffusion characterized by the power-law  $\langle x^2(t) \rangle \simeq t^\alpha$  of the mean squared displacement with  $0 < \alpha < 1$ , the additional many-body effects lead to the emergence of ultraslow, logarithmic motion of the form

$$\langle x^2(t) \rangle \simeq \log^{1/2} t. \quad (2)$$

Mathematically, we will show that we can understand this result from subordination of the many-body Harris result (1) in absence of the disorder to the dynamics of a counting process in an ageing system [22], leading to the universal 1/2 exponent of the ensuing logarithmic law (2). The remarkable slowing-down in our system, due to the conspiracy of disorder and strong

particle interactions, represents a generic effect for ultraslow dynamics in low-dimensional many-body systems. Such logarithmic time evolution is indeed experimentally observed in numerous experimental many-body studies, for instance, for DNA local structure relaxation [23], frictional strength [24], grain compactification [25], glassy systems [26], relaxation in Andersson insulators [27] and electron-glasses [28], paper crumpling under a heavy piston [29], as well as magnetization, conductance, and current relaxations in superconductors, spin glasses, and field-effect transistors [30].

## 2. Scale-free sticking times and single file dynamics

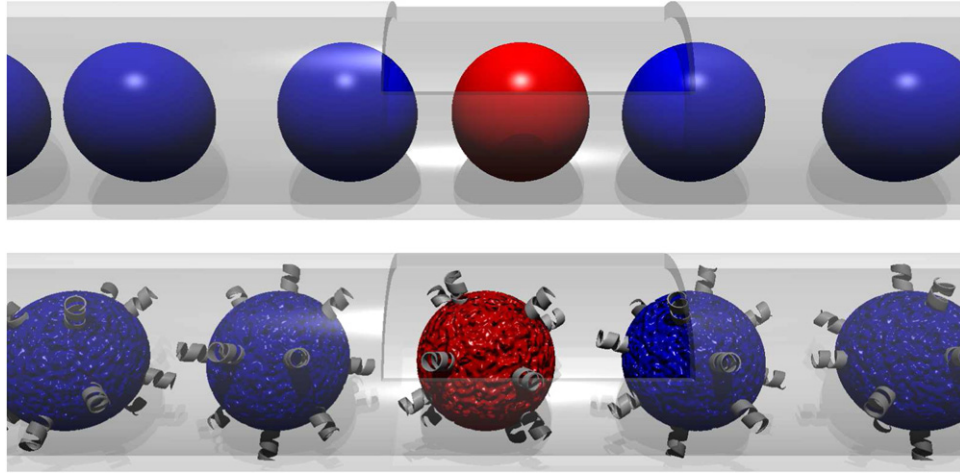
To introduce our system, let us start with a simple physical example. Imagine a *single* colloidal particle diffusing in a narrow fluidic channel. Its mean squared displacement  $\langle x^2(t) \rangle \simeq t$  will display the linear time dependence characteristic of Brownian motion. In contrast, if the same particle is made viscid by functionalization with ‘sticky’ ends and the channel surface complementarily coated, its motion will exhibit intermittent pausing events caused by transient binding to the channel surface with diffusive unbinding. The distribution of pausing durations  $\tau$  of the particle motion due to the sticking events is of power-law form  $\psi(\tau) \simeq \tau^{-1-\alpha}$  with  $0 < \alpha < 1$  in a certain temperature window, leading to the subdiffusive behaviour [31]

$$\langle x^2(t) \rangle \simeq t^\alpha. \quad (3)$$

This scenario of interrupted motion belongs to the family of the Scher–Montroll–Weiss continuous time random walk [32, 33], a renewal process with independent successive waiting times. The subdiffusion (3) emerges due to the divergence of the mean waiting time  $\langle \tau \rangle = \int_0^\infty \tau \psi(\tau) d\tau$  [34–36].

Such stochastic dynamics with a power-law form for the distribution  $\psi(\tau)$  of sticking times  $\tau$  and  $0 < \alpha < 1$  are indeed observed for the single tracer particle motion in the cytoplasm [37] and in membranes [38] of living cells, as well as in reconstituted actin networks [39]. Moreover, they determine the blinking dynamics of single quantum dots [40] and the dynamics involved in laser cooling [41]. Physically, the form  $\psi(\tau)$  may arise from comb models [42] or random energy landscapes [43]. The divergence of the mean sticking time leads to ageing phenomena [45] and weak ergodicity breaking [46], with deep consequences for, inter alia, cellular processes [36, 47].

Biomolecules are intrinsically heterogeneous and the fabrication of nanosystems often includes surface roughness. It is therefore a natural step to consider the scenario of single file diffusion in a disordered environment. As our guiding example we consider the situation of a colloidal particle in a channel and ask what will happen if we surround this colloidal particle with identical particles (figure 1)? As the channel is narrow, individual particles cannot pass each other, thus forming a single file [2, 3, 48, 49]. When the colloidal particles and the channel walls are not coated this system serves as a prototype for strongly interacting classical particles in one dimension and displays the characteristic Harris scaling (1) of the mean squared displacement of the labelled particle [2, 3, 48, 49]. However, when we use functionalized, sticky particles, the motion of each individual particle becomes successively interrupted by sticking times  $\tau$  distributed according to the law  $\psi(\tau)$ . When the distribution  $\psi(\tau)$  is scale free in the case  $0 < \alpha < 1$ , our scaling arguments and extensive simulations show that the motion of the labelled sticky tracer particle becomes slowed down dramatically and assumes the square-



**Figure 1.** Schematic of a narrow channel containing a single file of  $N$  colloidal particles (blue). In our description we are interested in the stochastic motion of the labelled tracer particle (red), which is otherwise identical with its neighbours. Regular particles in a bare channel (top) perform single file motion characterized by Harris' law  $\langle x^2(t) \rangle \simeq t^{1/2}$ . For the case of functionalized, sticky particles (bottom) the motion becomes ultraslow,  $\langle x^2(t) \rangle \simeq \log^{1/2}(t)$ .

root logarithmic dynamics (2). We also find that even when a characteristic sticking time  $\langle \tau \rangle$  exists, as long as  $1 < \alpha < 2$  the motion of the labelled particle is still anomalous,  $\langle x^2(t) \rangle \simeq t^\gamma$  with a non-universal,  $\alpha$ -dependent dynamic exponent  $\gamma < 1/2$ . When  $\alpha > 2$ , we return to the regular universal  $1/2$  Harris scaling exponent.

### 3. Motion rules for the single file system

Let us now describe our simulation scheme. A *single walker* is updated following the simple continuous time random walk rules: on our one-dimensional lattice, jumps occur to left and right with equal probability, and the waiting times between successive jumps are drawn from the probability density

$$\psi(\tau) = \frac{\alpha}{\tau^\star (1 + \tau/\tau^\star)^{1+\alpha}}, \quad (4)$$

where  $\tau^\star$  is a scaling factor with unit of time. As noted above,  $\psi(\tau)$  given by equation (4) occur, for instance, for the motion in random energy landscapes [43]. In this particular scenario site-independence of the waiting time density corresponds to the motion in an annealed free energy landscape (the landscape is evolving on a time scale faster than  $\tau^\star$ ). However, we again emphasize that  $\psi(\tau)$  of the power-law form above occur in several other physical systems (see section 2). Practically, the waiting times become  $\tau = \tau^\star [r^{-1/\alpha} - 1]$ , where  $r$  is a uniform random number from the unit interval. After each jump the walker's clock is updated, algorithmically,  $T \rightarrow T + \tau$ , where initially  $T = 0$ . For the scale-free case,  $0 < \alpha < 1$ , i.e., infinite average waiting time  $\langle \tau \rangle$ , we obtain subdiffusive transport,  $\langle x^2(t) \rangle = 2K_\alpha t^\alpha / \Gamma(1 + \alpha)$  [35]. Here,  $K_\alpha = a^2 / [2(\tau^\star)^\alpha \Gamma(1 - \alpha)]$  is the anomalous diffusion coefficient with  $a$  the lattice spacing. For  $\alpha > 1$ ,  $\langle \tau \rangle$  is finite and we recover normal diffusion.

For the case of a single file of *many* excluded-volume walkers the motion of individual particles is updated in a similar fashion, with the convention that any jump leading to a double occupancy of lattice sites is cancelled. More specifically, the simulation steps are:

- (i) Assign initial positions to all the  $N$  particles (indexed by  $i = 1, \dots, N$ ). We position the labelled particle at the middle lattice point and randomly distribute equally many particles to the left and right. Each particle carries its own clock with timer  $T_i$ , and all clocks are initiated simultaneously,  $T_i = 0$ .
- (ii) Draw an independent random waiting  $\tau_i$  from equation (4) for each particle and add this to the timer,  $T_i \rightarrow T_i + \tau_i$ .
- (iii) Determine the particle  $j$  with the minimal value  $T_j = \min \{T_i\}$  and move particle  $j$  one step with probability 1/2 to the left or right, unless the chosen site is already occupied by another particle or is outside the finite interval of our simulations. In this case cancel the move.
- (iv) Add a new waiting time  $\tau_j$  chosen from  $\psi(\tau)$  to the timer of particle  $j$ , i.e.,  $T_j \rightarrow T_j + \tau_j$ , and return to (iii).

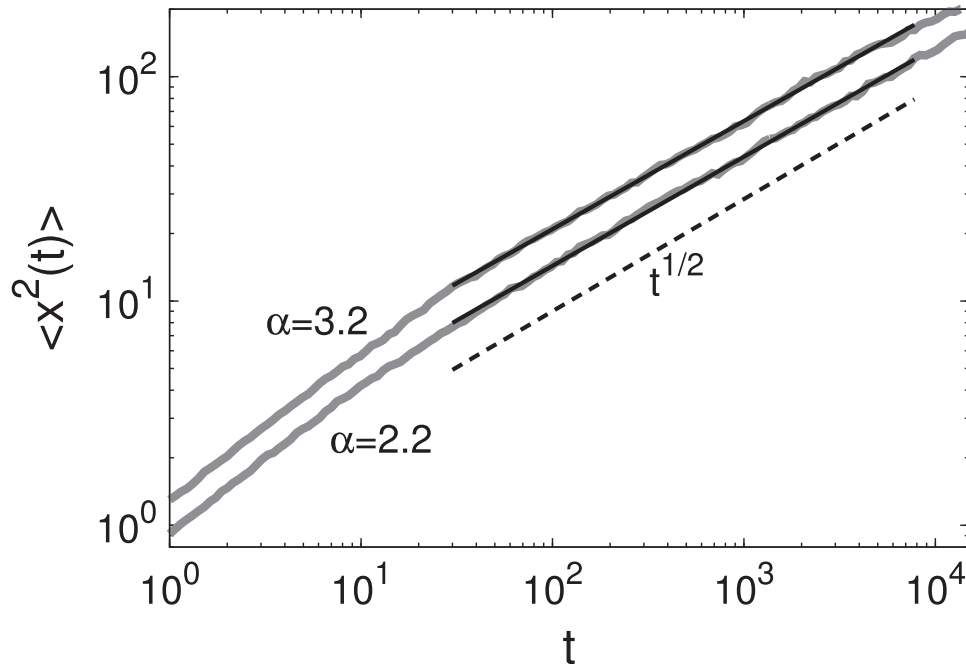
This algorithm is repeated until a designated stop time. The simulations presented herein, based on these simple update rules, were at the computational limit of a contemporary 3 GHz workstation.

The above motion scenario used in our stochastic simulations directly reflects the local nature of the physical problem (figure 1). Namely, when we follow individual, sticky particles in the channel, each binding and subsequent unbinding event will provide a different, random, waiting time. The waiting time densities for different particles are assumed to be independent. As rationalized by our picture of the sticky particles in the channel, physically this corresponds to the assumption that a particle's waiting time is set by properties of the local environment and that the local environment in between collisions cannot transfer 'knowledge' of the motion of one walker to another one. It is only when colliding that two particles affect one another. Our choice of cancelling moves when a particle collides with another particle comes from the underlying assumption, that overall the noise coming from a neighboring single file particle does not significantly distinguish itself from the noise of the heat bath, say water molecules, that would otherwise take up the space of the particle. Note that since we are in the overdamped limit the particles do not carry momentum that could be transferred in collisions. In a continuous model for the single file motion the motion rules would be less restrictive.

#### 4. Results and subordination arguments

For the case of the power-law waiting time distribution (4) with exponent  $\alpha > 2$ , we show results from extensive simulations based on above motion rules in figure 2. Our results reproduce the classical Harris scaling (1) of Brownian particles with scaling exponent 1/2. The fitted exponents from the simulations in figure 2 are  $0.49 \pm 0.01$  for  $\alpha = 2.2$  and  $0.48 \pm 0.01$  for  $\alpha = 3.2$ , thus corroborating our simulations approach. We note that the lattice sizes were throughout this study chosen sufficiently large as to avoid finite size effects, i.e., the levelling off of the mean squared displacement.



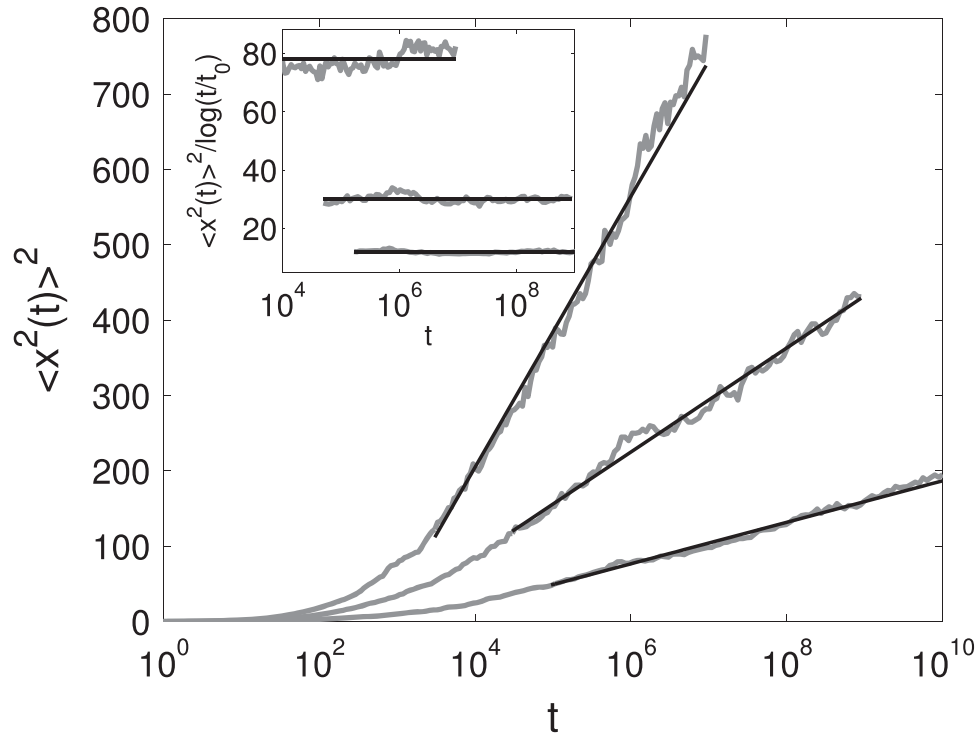


**Figure 2.** Mean squared displacement  $\langle x^2(t) \rangle$  for a single file system governed by the waiting time distribution (4) with the scaling exponents  $\alpha = 2.2$  and  $3.2$ . In both cases the mean squared displacement follows the Harris  $1/2$  scaling (dashed line) for long times. Parameters: scaling factor  $\tau^* = 1$ , lattice size  $L = 600$ , number of particles  $N = 201$ , so that the particle density is  $\rho = N/L \approx 0.3$ . The mean squared displacement was averaged from  $3.5 \times 10^3$  simulations.

#### 4.1. Infinite mean sticking time with $0 < \alpha < 1$

Let us now turn to the case of interest with a diverging mean sticking time  $\langle \tau \rangle$  corresponding to the range  $0 < \alpha < 1$  of the scaling exponent. Simulations results are depicted in figure 3. Note that we plot the *square* of the mean squared displacement versus time, such that according to the logarithmic time evolution (2) of the mean squared displacement derived from a scaling argument below, we would expect a linear dependence using linear-log scales. Indeed, the numerical results support the universal square root-logarithmic time evolution at long times for all exponents  $\alpha$  (see also inset). The only asymptotic dependence on  $\alpha$  is that of the prefactor in figure 3.

To understand and quantify the system's dynamics we now obtain the mean squared displacement for the labelled particle from a scaling argument. First, however, recall that for the standard case of a Poissonian (exponential) sticking time distribution between jumps of the labelled particle in the single-file system, the long-time dynamics belongs to the class of fractional Langevin motion with Hurst exponent  $H = 1/4$  [5, 14]. For later argumentation we use a scaling argument to obtain the Harris law by using fractional Langevin results. We first notice that the time in between collisions is estimated as  $\tau_c = 1/(q^2 D)$ , i.e. it is the time it takes to diffuse a distance equal to the average interparticle distance,  $1/q$ . We now coarse-grain the dynamics in terms of  $\tau_c$  using the fact that  $t = n\tau_c$ , where  $n$  is the number of collisions. The position,  $x(n)$ , of the tracer particle after taking  $n$  such steps is  $x(n) = \sum_{i=1}^n \Delta x_i$ , where  $\Delta x_i$  is a random number taking the values  $\pm 1/q$  with equal probability. The  $\Delta x_i$ 's are strongly anti-



**Figure 3.** Squared mean squared displacement (grey) for a labelled particle in a single file governed by the waiting time distribution (4) with  $0 < \alpha < 1$ . Note the logarithmic abscissa. Black solid lines are fitted to  $\langle x^2(t) \rangle = c\sqrt{\log(t/t_0)}$  with fitting parameters  $c$  and  $t_0$ , see equation (2). Inset: square of the MSD divided by  $\log(t/t_0)$  as a function of time  $t$ . Our scaling argument predicts a horizontal line (black line) for long times, in good agreement with simulations (grey). Parameters: ensemble size  $5 \times 10^3$ , lattice size 100, number of particles  $N = 31$ .

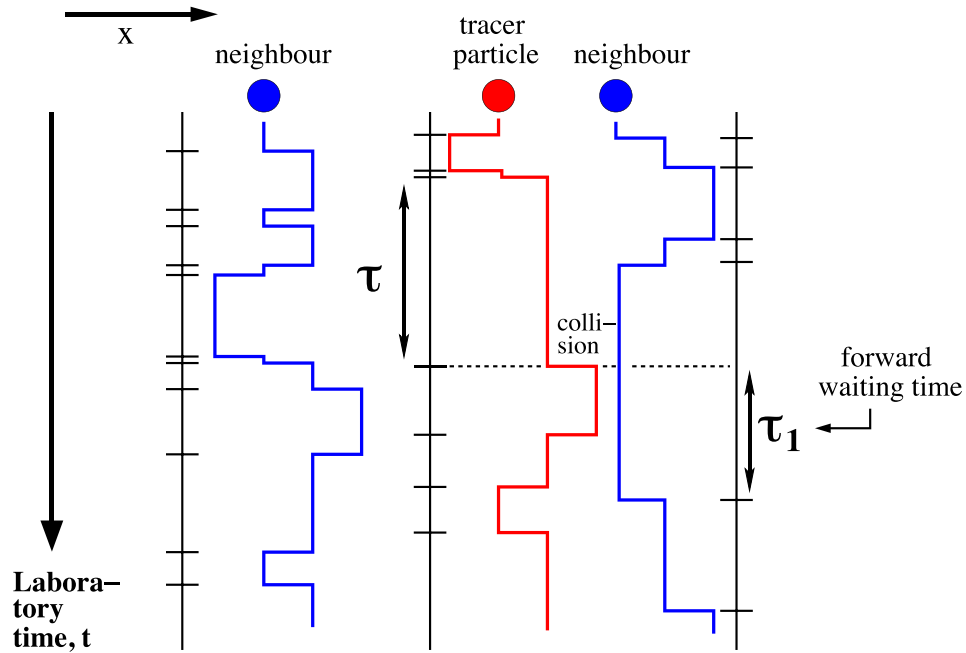
correlated,  $\langle \Delta x_i \Delta x_j \rangle \sim -|i - j|^{-3/2}$ , for fractional Brownian motion: if a particle finds itself to the left of its equilibrium position after a number of steps it is more likely to go right, due to collisions with its neighbours, in subsequent steps. Using this result we find that:

$$\langle x^2(n) \rangle = \sum_{i,j=1}^n \langle \Delta x_i \Delta x_j \rangle \sim C \frac{1}{q^2} n^{1/2}, \quad (5)$$

with a prefactor  $C = 2/\sqrt{\pi}$  [10]. The fact that we have  $\langle x^2(n) \rangle \propto \sqrt{n}$  rather than  $\propto n$  is due to the strong anti-correlation of the increments. If we plug  $n = t/\tau_c$  into equation (5) we recover the Harris result, equation (1) with correct prefactors [10].

Consider now the case of the functionalized, sticky particles and channel walls causing a non-Poissonian single file dynamics. The dynamics of the system is then characterized by motility periods, separated by blockage events when immobile neighbours are encountered, see figure 4. For scale-free waiting time distributions of the form (4) these blockage events are long compared with the duration of the motility periods, and thus the blockage events dominate the long time dynamics. In effect, at a given large time  $t$ , the number of jumps,  $n$ , which is larger than the step length,  $1/q$ , and thus contributes to the sum in equation (5), is smaller for power-law distributed waiting times compared to the case of exponential waiting time densities. In our scaling approach, we take this into account by converting the number of such steps  $n$  to the time





**Figure 4.** Illustration of our scaling argument and the forward (recurrence) waiting time, showing a particular realization of the trajectories of the tracer particle (red) and its immediate neighbours (blue). The horizontal direction corresponds to the motion of the particles along the  $x$  axis, the vertical line corresponds to the time axis. Impulses denote jump events for the respective particles. Individual particle waiting times  $\tau$  are drawn from the waiting time distribution (4), see the marked black vertical lines. The forward waiting time,  $\tau_1$ , is the time a particle needs to wait from colliding with a neighbour to the neighbour resuming its motion. The neighbour, in turn, needs to wait for the next particle in the single file, before it can progress further, etc. Our scaling argument (see section 4.1) is valid for long times, so the local motion of particles between blocking neighbours is not rate limiting.

$t$  measured by the laboratory master clock. This translation from the number of steps  $n$  to  $t$  is referred to as subordination [50]. Formally, if we denote by  $H_n(t)$  the probability that the tracer particle has taken  $n$  steps up to time  $t$ , we invoke the transformation

$$\langle x^2(t) \rangle = \sum_n \langle x^2(n) \rangle H_n(t) \longrightarrow C \int_0^\infty n^{1/2} H_n(t) dn. \quad (6)$$

Here we assumed that the *subordinator*  $H_n(t)$  is slowly varying in  $n$  to replace the sum by an integral.

To proceed, we employ a scaling argument to relate  $n$  with laboratory time  $t$  in the limit of many jumps (long times). As argued above, the rate limiting step for the motion of the labelled particle is the time needed for a blocking neighbour to resume its motion. This time corresponds to the so-called forward waiting (recurrence) time,  $\tau_1$ , see figure 4 for an illustration. For processes with  $\psi(\tau) \simeq \tau^{-1-\alpha}$  for large  $\tau$ , see equation (4), one can obtain the probability density for  $\tau_1$ , which is found to be [44, 45]

$$\psi_1(\tau_1; t) = \frac{\sin(\pi\alpha)}{\pi} \frac{t^\alpha}{\tau_1^\alpha(\tau_1 + t)}, \quad (7)$$

where  $t$  is the current (laboratory) time. Note that the probability density for  $\tau_1$  has a longer tail  $\psi_1(\tau_1) \simeq \tau_1^{-\alpha}$  than the underlying waiting time density,  $\psi(\tau) \simeq \tau^{-1-\alpha}$ , in the strong ageing regime  $t \gg \tau_1$ .<sup>6</sup> However, for our system, the distribution of a single  $\tau_1$  is not sufficient to describe the time evolution of a tracer particle. The reason for this is, simply, that the tracer particle's neighbour, in turn, needs to wait for its neighbours to move, etc (see figure 4). In the long time limit we thus face a process, in which *every* step  $n$  is governed by the forward waiting time characteristic of the blockage events. Such a process was considered recently, and there it was shown that the average number of steps taken at time  $t$  scales as  $\log t$  [22]. Furthermore, the spread (standard deviation) of the corresponding probability distribution was shown to grow slower than  $\log t$ , implying that in the long time limit we may consider  $n \simeq \log t$  as a *deterministic* (scaling) relation between  $n$  and  $t$ .

The argument above leads us to the scaling ansatz for the subordinator  $H_n(t)$  in equation (6) for the scale-free case with  $0 < \alpha < 1$ . Namely, the subordinator should be expressed in terms of a scaling function  $f(n/\log t)$ . Imposing the normalization  $\int_0^\infty H_n(t) dn = 1$ , i.e., a jump necessarily occurs at some given time, we thus have the result

$$H_n(t) \simeq (\log t)^{-1} f(n/\log t) \quad (8)$$

valid in the limit of many jumps,  $n \gg 1$  (which implies  $t \gg \tau^*$ ). Combining this scaling form with equations (5) and (6) after the change of variables  $n \rightarrow n/\log t$  we indeed obtain the square-root logarithmic behaviour in equation (2). Interestingly, compared to the standard square root scaling of Brownian single file motion, the scale-free waiting time process introduces a logarithmic time. For the full range  $0 < \alpha < 1$  we find the universal  $1/2$  scaling of the logarithm. As we show in figure 3 this simple scaling argument combined with the results from [22] indeed accurately captures the dynamics of the many-body continuous time random walk system. We discuss this result further below.

#### 4.2. Finite mean sticking time with $\alpha > 1$

What happens when we turn to larger values of the anomalous exponent  $\alpha$ , such that the characteristic waiting time  $\langle \tau \rangle$  becomes finite? Similar to the observations in [22] it turns out that we need to distinguish two cases. Let us start with the case  $\alpha > 2$ . The results of [22] suggest a deterministic, linear scaling between  $n$  and  $t$ . Thus, we require the scaling form  $H_n(t) \simeq t^{-1} f(n/t)$ . In turn, this relates equation (5) to the time dependence

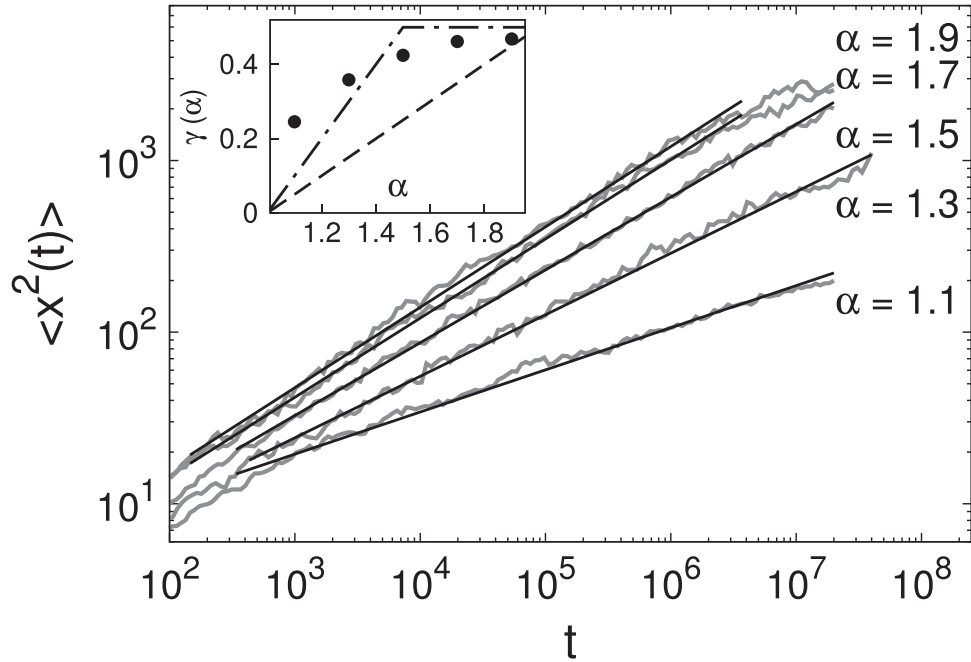
$$\langle x^2(t) \rangle \simeq t^{1/2}, \quad (9)$$

of the mean squared displacement and we recover the Brownian single file dynamics. This characteristic  $1/2$  scaling corresponding to Harris' law is confirmed in figure 2.

For the intermediate case,  $1 < \alpha < 2$ , the single file dynamics becomes rather subtle and different to the case  $\alpha > 2$ .<sup>7</sup> Direct use of the results in [22] would imply that  $\langle n \rangle \simeq t^{\alpha-1}$ , suggesting the scaling ansatz  $H_n(t) \simeq t^{\alpha-1} f(n/t^{\alpha-1})$ , where the prefactor is again due to

<sup>6</sup> This result is a consequence of the fact that for a scale-free distribution  $\psi(\tau)$  of waiting times ( $0 < \alpha < 1$ ), typically longer and longer waiting times  $\tau$  occur in the course of the process. More precisely, if a set of random numbers  $\tau_j$  are drawn from  $\psi(\tau)$  in equation (4), and if  $T = \sum_j \tau_j$  and  $\tau_{\max}$  is the largest of all  $\tau_j$ , then  $\tau_{\max}/T$  is a random variable with a probability density which is not concentrated on 0.

<sup>7</sup> The behaviour discussed here is in contrast to the unbiased single particle continuous time random walk with  $1 < \alpha < 2$  which has a simple Brownian mean squared displacement, just as for  $\alpha > 2$ .



**Figure 5.** Mean squared displacement  $\langle x^2(t) \rangle$  for a single file system with waiting time distribution (4) in the intermediate regime,  $1 < \alpha < 2$ . The fitted scaling exponent  $\gamma$  (inset) is compared to the predictions  $\gamma(\alpha) = (\alpha - 1)/2$  (straight dashed line) and  $\min\{\gamma(\alpha) = \alpha - 1, 0.5\}$  (kinked dash-dotted line) derived in the text. For the cases  $\alpha = \{1.1, 1.3, 1.5, 1.7, 1.9\}$  the MSD was averaged over  $\{750, 300, 750, 500, 450\}$  simulations runs. The system size was  $L = 4472$  and the number of particles  $N = 1341$ , except for  $\alpha = 1.3$  where we used  $L = 8944$  and  $N = 2683$ . Thus, the density,  $N/L$ , was  $\approx 0.3$  in all simulations.

normalization. This approach would yield the mean squared displacement

$$\langle x^2(t) \rangle \simeq t^{\gamma(\alpha)}, \quad (10)$$

with  $\gamma(\alpha) = (\alpha - 1)/2$ . As demonstrated in figure 5 (inset), this prediction for the scaling exponents does not agree well with the simulations. One possible issue is that for this intermediate case the spread of the number of jumps grows proportionally to the square root of the variance, and there is no deterministic relation between  $n$  and  $t$  as for the cases  $0 < \alpha < 1$  and  $\alpha > 2$  [22].

An improved argument turns out to be the following. Since the random walk is unbiased, a given particle can equally well escape in either direction from an interval confined by two blocking particles (see figure 4). Thus the labelled tracer particle only needs to wait for the blocked neighbour that moves first, corresponding to the minimum of two waiting times drawn from the forward waiting time density (7). The distribution of this minimum time will have the tail [51]

$$\tilde{\psi}_1(\tau_1) = 2\psi_1(\tau_1) \int_{\tau_1}^{\infty} \psi_1(\tau') d\tau' \simeq \tau_1^{-1 - (2\alpha - 2)}, \quad (11)$$

where we use the fact that in this case the distribution of the forward waiting time scales as  $\psi_1(\tau_1) \simeq 1/[\langle \tau \rangle \tau_1^\alpha]$ . The resulting mean squared displacement for the labelled particle thus

scales in the form of equation (10) where  $\gamma(\alpha) = \alpha - 1$  for  $1 < \alpha < 3/2$  and  $\gamma(\alpha) = 1/2$  for  $\alpha > 3/2$ . The saturation to the value  $\gamma(\alpha) = 1/2$  can be understood as follows. The distribution  $\tilde{\psi}_1(\tau_1)$  with saling index  $2(\alpha - 1)$  acts as waiting time distribution for the labelled particle. For a regular single walker CTRW process, when this index exceeds unity, the process becomes normally diffusive. Here, when the index  $2(\alpha - 1)$  exceeds unity, the scaling index  $\gamma$  comes back to the Brownian value  $1/2$ .

As seen from figure 5 (inset), this argument indeed leads to an improved agreement with the fitted exponents. Using the minimum of two waiting times instead of the direct subordination will not alter the scaling of the mean squared displacement for  $\alpha < 1$ , as the log  $t$  scaling was a result of the ageing of the waiting time distribution (i.e., its dependence on the time at which the waiting began), a property that will be carried over in the distribution of the minimum  $\tilde{\psi}_1$ . The mean squared displacement scaling for  $\alpha > 2$  is also unchanged by the modified argument above.

## 5. Discussion

We studied a physical model for the motion of interacting (excluded volume) particles in a strongly disordered environment. Building on the physical scenario of recent experiments in which sticky particles move along a complementary, functionalized surface, we assume that each particle performs a continuous time random walk with a power-law waiting time distribution  $\psi(\tau)$ . Each particle carries an individual clock whose timer triggers motion attempts according to this law  $\psi(\tau)$  and we attempt to move the particle whose timer expires first. Thus, while the update of the timers for each particle is a renewal process, the excluded volume interactions lead to strong correlations between the motion of the particles: when one particle attempts to move and finds the neighbouring lattice site occupied, typically the blocking particle is caught in a long waiting time period, and repeated attempts of motion by the mobile particle will be required. In the long time limit, we demonstrated from scaling arguments and extensive simulations that this many-body blockage scenario leads to an ultraslow logarithmic time evolution of the mean squared displacement of a labelled particle for  $\psi(\tau)$  with  $\alpha < 1$ . The  $1/2$  exponent of the logarithm is universal for any  $\alpha$  in that range. When the environment is less strongly disordered and the waiting time exponent  $\alpha > 1$ , the associated characteristic waiting time  $\langle \tau \rangle$  is finite. However, similar to biased continuous time random walk processes [52], there exists an intermediate regime for  $1 < \alpha < 2$ , which still exhibits anomalous scaling: the mean squared displacement has a power-law scaling with time, but the associated non-universal exponent is smaller than the value  $1/2$  for Brownian (Harris) single file motion. Our analysis shows that the subordination scheme based on a simple two-body correlation scheme needs to be replaced by higher order corrections. Only when the waiting time exponent  $\alpha$  exceeds the value 2, the process returns to Harris-type single file motion with  $\langle x^2(t) \rangle \simeq t^{1/2}$ .

Our study demonstrates how the addition of environmental disorder to a system with many-body interactions effects dramatic changes of the dynamics. While in a single body system the same disorder simply changes the scaling exponent in the power-law of the mean squared displacement, in the presence of the many-body interactions with mutual blocking ultraslow motion emerges. This is a fundamental physical effect. If a similar disorder could be engineered in microscopic systems, the ultraslow dynamics caused by the disorder might be of

high technological interest. For instance, it might lead to a significant improvement of the long-term retention of chemicals in engineered cylindrical nanochannels for usage in drug delivery assays, contemporary versions of which are based on regular, Harris' type single file motion [20]. Moreover, if the magnitude of the disorder and thus the scaling exponent  $\alpha$  of the waiting time distribution can be externally controlled, similar to the temperature effect in [31], even more versatile crossovers from single file type motion to regular motion may be achieved [19].

In [53, 54] another continuous time random walk-based generalization of single file motion was considered. However, their update rules for particles colliding with a neighbour are very different. One way to view their process is that of a castling, i.e., particles are allowed to move through each other (phantom particles), while the labels of the particles switch in this castling. Thus the labels will stay in the same order in the file and the tracer following a specific label. Alternatively, the rule can be stated as particles switching their clocks when they collide. [53, 54] found that with this rule the generalized single file dynamics acquires the mean squared displacement  $\langle x^2(t) \rangle \simeq t^{\alpha/2}$  for  $0 < \alpha < 1$ . This result is fundamentally different from our ultraslow result (2) based on an actual physical model, as we explicitly consider excluded volume effects [55]. In particular, different from the scenario studied in [55], we do not observe any particle clustering.

Finally, we put the ultraslow time evolution discovered here in perspective to other stochastic models with logarithmic growth of the mean squared displacement. The most famous process is that of Sinai diffusion of a single particle in a quenched, random force field in one dimension, leading to a  $\log^4(t)$  scaling of the mean squared displacement [56]. Despite the fundamental difference in the physical setting, ultraslow dynamics with very similar features—such as the form of the weak ergodicity breaking—is found for a renewal continuous time random walk with logarithmic waiting time distributions [58, 59]. Logarithmically slow time evolution also occurs in the motion of a Markovian particle with exponential position-dependence of the diffusion constant [57]. Our model is, to our knowledge, the first *interacting many-body* system for which tracer particle dynamics show the characteristic  $\log^{1/2}(t)$  dynamics. We expect our work to stimulate new research of the role of disorder in interacting many-body systems.

An interesting question is to what extent the ultraslow dynamics in the disordered single file system leads to the disparity between the mean squared displacement  $\langle x^2(t) \rangle$  considered here and its time averaged analogue used in the evaluation of many single particle tracking experiments. Such weakly non-ergodic behaviour breaking known from subdiffusive systems [36, 47] was indeed observed for other ultraslow systems [59] and, in a modified form, for the diffusion process with exponential position-dependence of the diffusion constant [57]. This effect in our disorder-generalized version of the single file system will be studied in detail in a forthcoming work.

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## References

- [1] Samaj L and Bajnok Z 2013 *Introduction to the Statistical Physics of Integrable Many-body Systems* (Cambridge, UK: Cambridge University Press)
- Stefanucci G and van Leeuwen R 2013 *Nonequilibrium Many-Body Theory of Quantum Systems: A Modern Introduction* (Cambridge, UK: Cambridge University Press)
- [2] Harris T E 1965 *J. Appl. Probab.* **2** 323
- [3] Levitt D G 1973 *Phys. Rev. A* **8** 3050
- [4] Fedders P A 1978 *Phys. Rev. B* **17** 40
- [5] Arratia R 1983 *Ann. Probab.* **11** 362
- [6] van Beijeren H, Kehr K W and Kutner R 1983 *Phys. Rev. B* **28** 5711
- [7] Kärger J 1992 *Phys. Rev. A* **45** 4173
- [8] Hahn K and Kärger J 1998 *J. Phys. Chem. B* **102** 5766
- [9] Nelson P H and Auerbach S M 1999 *J. Chem. Phys.* **110** 9235
- [10] Lizana L and Ambjörnsson T 2008 *Phys. Rev. Lett.* **100** 200601
- [11] Flomenbom O and Taloni A 2008 *Europhys. Lett.* **83** 20004
- [12] Barkai E and Silbey R 2009 *Phys. Rev. Lett.* **102** 050602
- [13] Ryabov A and Chvosta P 2011 *Phys. Rev. E* **83** 020106
- [14] Lizana L, Ambjörnsson T, Taloni A, Barkai E and Lomholt M A 2010 *Phys. Rev. E* **81** 051118
- [15] Wei Q-H, Bechinger C and Leiderer P 2000 *Science* **287** 625
- [16] Lutz C, Reichert M, Stark H and Bechinger C 2006 *Europhys. Lett.* **74** 719
- [17] Lutz C, Kollmann M and Bechinger C 2004 *Phys. Rev. Lett.* **93** 026001
- [18] Hahn K, Kärger J and Kukla V 1996 *Phys. Rev. Lett.* **76** 2762
- [19] Siems U, Kreuter C, Erbe A, Schwierz N, Sengupta S, Leiderer P and Nielaba P 2012 *Sci. Rep.* **2** 1015
- Pagliara S, Dettmer S L and Keyser U F 2014 *Phys. Rev. Lett.* **113** 048102
- [20] Yang S Y, Yang J A, Kim E S, Jeon G, Oh E J, Choi K Y, Hahn S K and Kim J K 2010 *ACS Nano* **4** 3817
- [21] Stern A S and Berg H C 2013 *Biophys. J* **105** 182
- [22] Lomholt M A, Lizana L, Metzler R and Ambjörnsson T 2013 *Phys. Rev. Lett.* **110** 208301
- [23] Brauns E B, Madaras M L, Coleman R S, Murphy C J and Berg M A 2002 *Phys. Rev. Lett.* **88** 158101
- [24] Ben-David O, Rubinstein S M and Fineberg J 2010 *Nature* **463** 76
- [25] Richard P, Nicodemi M, Delannay R, Ribière P and Bideau D 2005 *Nat. Mater.* **4** 121
- [26] Boettcher S and Sibani P 2011 *J. Phys. Condens. Matter* **23** 065103
- [27] Vaknin A, Ovadyahu Z and Pollak M 2000 *Phys. Rev. Lett.* **84** 3402
- [28] Orlyanchik V and Ovadyahu Z 2004 *Phys. Rev. Lett.* **92** 066801
- [29] Matan K, Williams R B, Witten T A and Nagel S R 2002 *Phys. Rev. Lett.* **88** 076101
- [30] Gurevich A and Küpfer H 1993 *Phys. Rev. B* **48** 6477
- Amir A, Oreg Y and Imry Y 2012 *Proc. Natl Acad. Sci. USA* **109** 1850
- Amir A, Borini S, Oreg Y and Imry Y 2011 *Phys. Rev. Lett.* **107** 186407
- Woltjer R, Hamada A and Takeda E 1993 *Electron. Devices IEEE Trans.* **40** 392
- Chowdhury D and Mookerjee A 1984 *J. Phys. F: Met. Phys.* **14** 245
- [31] Xu Q, Feng L, Sha R, Seeman N C and Chaikin P M 2011 *Phys. Rev. Lett.* **106** 228102
- [32] Montroll E W and Weiss G H 1969 *J. Math. Phys.* **10** 753
- [33] Scher H and Montroll E W 1975 *Phys. Rev. B* **12** 2455
- [34] Hughes B D 1995 *Random Walks and Random Environments Volume 1: Random Walks* (Oxford: Oxford University Press)
- [35] Metzler R and Klafter J 2000 *Phys. Rep.* **339** 1
- Metzler R and Klafter J 2004 *J. Phys. A: Math. Gen.* **37** R161
- [36] Barkai E, Garini Y and Metzler R 2012 *Phys. Today* **65** 29
- Metzler R, Jeon J-H, Cherstvy A G and Barkai E 2014 *Phys. Chem. Chem. Phys.* **16** 24128



- [37] Jeon J-H, Tejedor V, Burov S, Barkai E, Selhuber-Unkel C, Berg-Sørensen K, Oddershede L and Metzler R 2011 *Phys. Rev. Lett.* **106** 048103
- Tabei S M A, Burov S, Kim H Y, Kuznetsov A, Huynh T, Jureller J, Philipson L H, Dinner A R and Scherer N F 2013 *Proc. Natl Acad. Sci. USA* **110** 4911
- [38] Weigel A V, Simon B, Tamkun M M and Krapf D 2011 *Proc. Natl Acad. Sci. USA* **108** 6438
- [39] Wong I Y, Gardel M L, Reichman D R, Weeks E R, Valentine M T, Bausch A R and Weitz D A 2004 *Phys. Rev. Lett.* **92** 178101
- [40] Stefani F D, Hoogenboom J P and Barkai E 2009 *Phys. Today* **62** 34
- [41] Bardou F, Bouchaud J-P, Emile O, Aspect A and Cohen-Tannoudji C 1994 *Phys. Rev. Lett.* **72** 203
- [42] Havlin S and Ben-Avraham D 1987 *Adv. Phys.* **36** 695
- [43] Monthus C and Bouchaud J-P 1996 *J. Phys. A: Math. Gen.* **29** 3847
- Ben Arous G, Bovier A and Gayraud V 2002 *Phys. Rev. Lett.* **88** 087201
- Burov S and Barkai E 2007 *Phys. Rev. Lett.* **98** 250601
- [44] Godrèche C and Luck J M 2001 *J. Stat. Phys.* **104** 489
- [45] Barkai E and Cheng Y C 2003 *J. Chem. Phys.* **118** 6167
- Schulz J H P, Barkai E and Metzler R 2013 *Phys. Rev. Lett.* **110** 020602
- Schulz J H P, Barkai E and Metzler R 2014 *Phys. Rev. X* **4** 011028
- Koren T, Lomholt M A, Chechkin A V, Klafter J and Metzler R 2007 *Phys. Rev. Lett.* **99** 160602
- [46] Bouchaud J-P 1992 *J. Phys. (Paris) I* **2** 1705
- Bel G and Barkai E 2005 *Phys. Rev. Lett.* **94** 240602
- Rebenshtok A and Barkai E 2007 *Phys. Rev. Lett.* **99** 210601
- Lomholt M A, Zaid I M and Metzler R 2007 *Phys. Rev. Lett.* **98** 200603
- Lomholt M A, Zaid I M and Metzler R 2009 *J. Biophys.* **97** 710
- [47] Höfling F and Franosch T 2013 *Rep. Prog. Phys.* **76** 046602
- [48] Rödenbeck C, Kärger J and Hahn K 1998 *Phys. Rev. E* **57** 4382
- [49] Lizana L and Ambjörnsson T 2008 *Phys. Rev. Lett.* **100** 200601
- [50] Feller W 1971 *An Introduction to Probability Theory and its Applications* vol 2 (New York, NY: Wiley)
- [51] Schmittmann B and Zia R K P 1999 *Am. J. Phys.* **67** 1269
- [52] Shlesinger M F 1974 *J. Stat. Phys.* **10** 421
- Margolin G and Berkowitz B 2000 *J. Phys. Chem. B* **104** 3942
- [53] Bandyopadhyay T 2008 *Europhys. Lett.* **81** 16003
- [54] Barkai E and Silbey R 2010 *Phys. Rev. E* **81** 041129
- [55] Flomenbom O 2011 *Europhys. Lett.* **94** 58001
- [56] Sinai Y G 1982 *Theory Probab. Appl.* **27** 256
- Golosov A O 1984 *Commun. Math. Phys.* **92** 491
- Comtet A and Dean D 1998 *J. Phys. A: Math. Gen.* **31** 8595
- [57] Cherstvy A and Metzler R 2013 *Phys. Chem. Chem. Phys.* **15** 20220
- [58] Havlin S and Weiss G H 1990 *J. Stat. Phys.* **58** 1267
- Dräger J and Klafter J 2000 *Phys. Rev. Lett.* **84** 5998
- Denisov S I, Yuste S B, Bystrik Yu S, Kantz H and Lindenberg K 2011 *Phys. Rev. E* **84** 061143
- Chechkin A V, Klafter J and Sokolov I M 2003 *Europhys. Lett.* **63** 326
- [59] Godec A, Chechkin A V, Barkai E, Kantz H and Metzler R 2014 *J. Phys. A: Math. Gen.* at press (arXiv:1406.6199)